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# Influence of Processing Parameters and Composition on the Effective Compatibilization of Polypropylene–Poly(ethylene terephthalate) Blends

*The effects of the addition of different functionalized compatibilizers on toughness, morphology and rheological properties of a polypropylene (PP) – poly(ethylene terephthalate) (PET) (85–15 wt%) blend were studied. The three compatibilizers compared were: (Styrene Ethylene Butylene Styrene)-grafted-(glycidyl methacrylate); (Styrene Ethylene Butylene Styrene) – grafted – (maleic anhydride); (polyolefin) – grafted – (glycidyl methacrylate), abbreviated to: SEBS-g-GMA, SEBS-g-MA and POE-g-GMA respectively. The effective grafting content was the same for all three compatibilizers. Before the comparison of the different compatibilizers was done, first the effects of three different processing temperatures and three different compatibilizer contents were investigated, based on the addition of SEBS-g-GMA. The compatibilization effect was significantly improved with an increase in processing temperature from 250 to 300 °C. The toughness was increased with almost a factor two and a decrease in the average domain size of the dispersed phase was observed. An increase in compatibilizer content from 0.25 to 2.5 wt% resulted in a finer dispersity as well as in a steep increase in toughness, which was noted to approach the brittle-to-ductile transition. The comparison of the three compatibilizers was subsequently done at the most promising processing temperature and content: 300 °C and 2.5 wt%. The results showed that the addition of SEBS-g-MA and POE-g-GMA had a less significant positive effect on the compatibilization compared to SEBS-g-GMA. The difference is attributed to a higher reactivity for GMA compared to MA and a higher possibility for migration towards the PP-PET interface for the SEBS chain compared to the POE chain.*

## 1 Introduction

Plastics have become one of the most used materials in our lives. In Europe (EU-27+N/CH) alone, 57 metric ton of plastics are produced every year, of which near 46 metric ton are converted within the EU itself. This produces over 25 metric ton of yearly post-consumer plastic waste, of which only 6,6 metric ton are currently recycled. The rest goes to either energy recovery (8,9 metric ton) or landfill (38,1 metric ton) (PlasticsEurope, EuPC, et al. 2011). Plastic waste management is high on the European Commission's environmental agenda (European Commission, 2013) and professional organizations like Plastics Europe often refer to the potential impact of 'zero plastics to landfill', the imaginary timing of which has recently been pushed to 2025 instead of 2020 (Plastics Europe, 2015).

One of the product categories in which the demand for recycling has increased is the carpet industry. The most prominent type of household carpet is the cut-pile carpet, which is typically made of nylon or polyester yarns with a polypropylene backing (Jain, Pandey et al., 2012). In the recycling process, the upright yarns can be shaven off and recycled as a high-quality secondary material (EPA, 2009). This, however, leaves the backing with a short 'stubble' of remaining fiber.

This study considers the case of blends of polypropylene (PP) carpet backings with a minor fraction of remaining poly(ethylene terephthalate) (PET) shorn fiber. Such blends can also originate within products such as pressed carpet fibers and needle felt. A simple sorting process is not possible, since the two materials are physically attached to each other. This makes blending the only other option to mechanically recycle the carpet (production) waste. A major obstacle here is the thermodynamic immiscibility of PP and PET, due to their difference in chemical nature (van Krevelen and Te Nijenhuis, 2008). The untreated blend will end up as a two-phase morphology with poor interfacial adhesion, which leads to poor mechanical properties. One such mechanical property is the impact strength, which decreases significantly after blending the two incompatible polymers (Lei et al., 2009).

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The morphology is partly determined by the viscosity ratio and interfacial tension, and the average domain size of the dispersed phase is commonly described by the equation of Taylor (Eq. 1) (Wu, 1987):

$$\alpha_n = \frac{4\gamma_{12}p^x}{G \eta_m} \quad (1)$$

where  $\alpha_n$  = number of average particle size,  $\gamma_{12}$  = interfacial tension,  $v$  = viscosity ratio  $\times$  dispersed phase/matrix,  $G$  = effective shear rate,  $x = 0.84$  for  $p > 1$  and  $-0.84$  for  $p < 1$ ,  $\eta_m$  = viscosity of the matrix.

The interfacial tension also determines the interfacial adhesion and is therefore a crucial property in the determination of the compatibility. Compatibilizers can minimize the interfacial tension by preferentially locating themselves at the interface and interacting with both polymers (Pracella et al., 2002). The principle of this is illustrated in Fig. 1 (adapted from Ryan, 2002). If a compatibilizer is unable to become located at the interface, the maximum mechanical improvement cannot be obtained. The compatibilization effect can be promoted by the additional use of a functional group grafted onto the compatibilizer's chain. The functional group improves the physical and/or chemical interaction and results in a better-compatibilized blend (Heino et al., 1997; Pracella and Chionna, 2001). This study focuses on the effect of different compatibilizer-functional group combinations for use in a PP-PET blend, which is intended to be mixed via twin-screw compounding and subsequent processing by injection molding.

PET has functional groups like the carboxylic and/or hydroxyl end groups, contrary to PP which does not have any obvious reactive groups. The functionalized compatibilizers we discuss here are therefore mainly selected based on the interaction of the functional group with the PET phase and interaction of the rest of the compatibilizer with the PP phase is based on expected miscibility.

The most promising functional groups that are reactive towards PET are: Glycidyl Methacrylate (GMA) and Maleic Anhydride (MA) (Sun et al., 1996; Yildirim and Yurtsever, 2012). The final selection of the functionalized compatibilizers will contain these groups. The compatibilization effect of the chain is combined with impact modification by the use of a compatibilizer with an elastomeric character (Greco, 1998). The final choice for the functionalized compatibilizers is based on their commercial availability: styrene ethylene butylene styrene (SEBS)-g-GMA, polyolefin (POE)-g-GMA and SEBS-g-MA. SEBS is a block-copolymer, whose outer blocks have a low interfacial tension with PET and its inner EB blocks have a low interfacial tension with PP. The POE has a low in-

terfacial tension with PP only as illustrated in Fig. 1. Previous studies (Ihm and White, 1996; Heino et al., 1997; Papadopoulos and Kalfoglou, 2000; Pracella et al., 2002) to the effectiveness of compatibilizers can be found in literature, but quite often they compare (commercially available) compatibilizing agents with different grafting contents. As the grafting content is known to influence the degree of reaction (Sun et al., 2011) and thereby the compatibilization effect, it remains quite difficult to draw straightforward conclusions as to the actual effectiveness of the individual compatibilizer chains and their grafted functional groups. Within the current study, the grafting content is therefore explicitly the same for all compatibilizers. Additionally, a low grafting content is taken to avoid agglomeration of the functional groups (Sun et al., 2011) and to maintain economic viability of the proposed solutions.

In scope for this study are the compatibilization effects on mechanical, morphological and rheological properties of a PP-PET blend. The effect of processing temperature on the compatibilization effect is included in this study, since it can have an influence the viscosity ratios, on the degradation of both polymers, the reactivity of the functional group towards the PET phase, and thus on the final mechanical properties of the blend. In addition, the compatibilizer content was varied to study the combined impact modification effect, based on a possible change in ligament thickness that determines the impact modifying effect of the elastomeric particle (Bacci et al., 2013). The critical ligament thickness is determined by the matrix and lies between 0.1 and 0.8,  $\mu\text{m}$  for PP (Premphet and Paecharoenchai, 2002; Zhang et al., 2004; Lin et al., 2011).

## 2 Materials and Methods

### 2.1 Materials

The PP (Domolen 1400N) used is a nucleated homopolymer and has a density of 0,91  $\text{g}/\text{cm}^3$ . The PET (Eastar Copolyester 6763) used is glycol-modified which decreases the crystallization of the material resulting in a highly amorphous structure. It has a density of 1,27  $\text{g}/\text{cm}^3$  and a recommended processing temperature range of 250 to 270  $^{\circ}\text{C}$  (Eastman, 2014).

All PP-PET blends were manufactured with a composition of 85 : 15 wt%, PP:PET.

The three compatibilizers are SEBS-g-GMA, POE-g-GMA and SEBS-g-MA. SEBS-g-GMA (product name: RG901) and POE-g-GMA (product name: RG702) are supplied by Shanghai Jianqiao Plastic LTD., Shanghai, PRC, SEBS-g-MA (product name: Kraton FG1901 G) is supplied by Kraton, Amster-

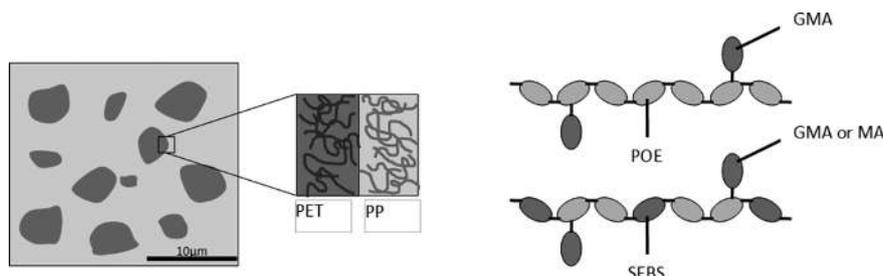


Fig. 1. Schematic representation of the effect of a compatibilizer on the blend. Chemical units in the used compatibilizers are color coded to indicate the polymer they will have an affinity for (PP or PET). Adapted from Al-Abdulrazzak and Jabarin, 2002

dam, The Netherlands. All compatibilizers have a functional group grafting content of 1.5 wt%.

## 2.2 Processing

Before each processing step the PET, the blends and all compatibilizers were dried during at least 12 hours in an oven at 60 °C.

All blends were compounded on a co-rotating twin screw extruder ZSK 18 MEGAlab (Coperion, Niel, Belgium), with a screw diameter of 18 mm and L/D ratio of 40. Processing parameters were set to screw speed of 150 min<sup>-1</sup> and feed rate of 3 kg/h. Previous modeling of the machine has shown that the effective shear rate in the screw lies between 50 and 100 s<sup>-1</sup>, with a residence time of 100 s (Delva et al., 2015). The die is a 2 mm round filament. This filament is led through a water bath of 15 °C and then immediately shredded into pellets. A large batch of PP:PET 85:15 blends was made as a precursor (at temperature profile 250 °C from Table 1) before the addition of compatibilizing agents. To this precursor blend, compatibilizers were consequently added at a controlled rate via the side feeder of the twin screw extruder, which is positioned at the fourth of ten screw zones, at the location of a set of right-handed kneading blocks and just after first degassing.

In the first step of this study the effect of temperature on the compatibilization effect was studied. This was done using the arbitrary addition of 2.5 wt% SEBS-g-GMA to the PP-PET blend. The compatibilized blend was mixed in the co-rotating twin screw extruder at three different temperature profiles: 250, 270 and 300 °C, as shown in Table 1. The temperature profile of 270 °C is the suggested temperature limit of processing for the used PET. The highest temperature profile was experimentally determined based on the appearance of the material coming out of the extruder. The other processing conditions of the extruder were the same as during blend preparation. The compatibilization effects were analyzed based on mechanical tests, analysis of the morphology and rheology measure-

ments. After the analysis of the properties, the most promising temperature was chosen for the subsequent steps. This was the profile of 300 °C, which was then used for all consecutive blend compounding.

The second step consists of the comparison of three different compatibilizer contents: 0.25, 1 and 2.5 wt%, again using SEBS-g-GMA. As a reference, virgin PP processed at the temperature profile of 250 °C was used. After the analysis of the properties, the most promising compatibilizer content was determined for the final part of this study. In this last part, the compatibilization effect of SEBS-g-GMA was compared to SEBS-g-MA and POE-g-GMA, for a compatibilizer content of 2.5 wt%.

## 2.3 Mechanical Testing

The samples for mechanical testing are prepared in a horizontal injection molder of type BOY 22S, Dr. BOY GmbH, Neustadt-Fernthal, Germany. The three temperature profiles of the compounding step are combined with three corresponding temperature profiles of the injection molder and are given in Tables 1 and 2 respectively. Hydraulic injection pressure was set at a relative 40% (of maximum) and switchover to holding pressure was after 30 mm injection length, to a relative pressure of 15% (for 250 °C and 270 °C) or 25% (for 300 °C) for 8 seconds. The coolant of the mold was set to 15 °C. The samples had the dimensions of 100 × 10 × 4 mm and were notched with a depth of 2 mm at location of 40 mm from the top. The impact tests were performed following ISO 180 of notched Izod impact test. The measurements were carried out at room temperature by making use of the izod-02 equipment of Zwick-Roell, Venlo, The Netherlands, with a falling hammer with energy of 1 Joule. Impact testing was chosen over tensile testing, as it has been shown that Young's modulus is high insensitive to particle size in a binary system (Fu et al., 2008).

Profile	T1	T2	T3	T4	T5	T6	T7	T8	T9
250	200	230	230	235	235	240	245	250	250
270	200	230	230	240	250	260	265	270	270
300	200	230	240	250	260	270	280	290	300

Table 1. Temperature profile of twin screw extruder, given from zone 1 to 9 in °C

Profile	T1	T2	T3	T4	T5
250	180	220	230	250	250
270	185	240	255	270	270
300	185	260	285	300	300

Table 2. Temperature profile of injection molder, given from zone 1 to 5 in °C

## 2.4 Scanning Electron Microscope

The samples used for analysis of the morphology were obtained in the same way as for mechanical testing after which they were cryofractured. The fractured surfaces were coated with a layer of gold to avoid charging of the samples and analyzed at room temperature by XL30S FEG or Nova Nano-SEM 450 scanning electron microscopes (SEM, FEI, Eindhoven, The Netherlands). An acceleration voltage of 10 kV was used during these analyses. Indicative sizing of PET domains was performed with ImageJ software (ImageJ, NIH, Maryland, USA). Twenty random PET particles were measured per graph.

## 2.5 Rheological Measurements

The rheological measurements of the pure components were performed on samples prepared from granular that were pressed between two hot plates at a temperature of 200 °C (Fontijne, Holland). The measurements of the blends excluding and including compatibilizer were performed on sampled sections from the specimen prepared in the same way as for the mechanical tests. These sampled sections were pressed to a flat and circular shape with a diameter 25 mm and a thickness of about 3 mm.

The dynamic mechanical measurements were performed by using a TA instruments ARG2 rheometer (TA Instruments, Asse, Belgium) between two parallel plates. Temperature dependent measurements were done at a constant angular frequency of 100 rad/s corresponding to the typical shear rate in the compounder. For analysis of the interfacial interaction, tests were performed at a constant temperature of 220 °C. The latter analysis was done at lower frequencies, since at lower frequencies the (elastic) contribution of the interface is dominant compared to that of the components.

## 3 Results and Discussion

### 3.1 Processing Temperature

The main objective of this step is to compare the effect of different processing temperatures on the compatibilization effect of a functionalized compatibilizer. Arbitrarily, SEBS-g-GMA is chosen as compatibilizer in this step.

The effects of processing temperature on the impact strength of pure PP and of the PP:PET (85:15 wt%) blend excluding and including 2.5 wt% SEBS-g-GMA are shown in Table 3. The impact strength for virgin PP as well as for the blend excluding compatibilizer were unaffected by a change in processing temperature from 250 to 300 °C. However, the strength of the blend including SEBS-g-GMA was significantly improved upon an increase in processing temperature.

The SEM images of the binary and ternary blends are shown in Fig. 2. The PET phases in the binary blends are poorly dispersed and show large domain sizes that increase with increase in processing temperature. For uncompatibilized blends, this is a known phenomenon as the enthalpy of mixing (which adversely affects the miscibility) will increase with increasing temperature (Higgins et al., 2010). The coarse morphology is therefore a result of the high resulting interfacial tension (Wu, 1987). The increase in domain size with temperature is likely to be promoted by an increased deviation of the viscosity ratio from a value of one as explained by the Taylor equation (Eq. 1). Looking at the temperature dependence of dynamic viscosity of the virgin materials in Fig. 3, it is observed that the viscosity ratio of 1 for PP and PET would be reached around 295 °C. Afterwards, the two curves diverge once more and the ratio increases above 1. In practice, this crossover point will be at lower temperatures. Zero-shear viscosity for PET was experimentally determined as 2500 Pa s for virgin PET, but this falls to 1500 Pa s after a single processing step. As this

Experiment	PP wt%	PET wt%	CA type	CA wt%	T °C	Impact J/m <sup>2</sup>
T <sub>b</sub>	100	–	–	–	250	2,44 ± 0,06
T <sub>b</sub>	100	–	–	–	270	2,40 ± 0,07
T <sub>b</sub>	100	–	–	–	300	2,35 ± 0,08
T <sub>b</sub> , CA%, FG	85	15	–	–	250	1,20 ± 0,01
T <sub>b</sub>	85	15	–	–	270	1,23 ± 0,03
T <sub>b</sub>	85	15	–	–	300	1,21 ± 0,02
T <sub>b</sub>	85	15	SEBS-g-GMA	2,5	250	1,45 ± 0,11
T <sub>b</sub>	85	15	SEBS-g-GMA	2,5	270	1,91 ± 0,11
T <sub>b</sub> , CA%, FG	85	15	SEBS-g-GMA	2,5	300	2,21 ± 0,10
CA%	85	15	SEBS-g-GMA	0,25	300	1,23 ± 0,06
CA%	85	15	SEBS-g-GMA	1	300	1,24 ± 0,03
FG	85	15	SEBS-g-GMA	2,5	300	1,39 ± 0,14
FG	85	15	POE-g-GMA	2,5	300	1,40 ± 0,16

Table 3. Overview of all the impact results, in function of composition, compatibilizing agent (CA), amount of CA (proportional to the 100% of the PP-PET blend) and processing temperature (T). Izod impact is given as mean ± standard deviation. For clarity, it is marked which result lines are used for which comparative experiments: T<sub>b</sub> = the effect of blending temperature, CA% = the effect of wt% compatibilizing agent, FG = effect of the grafted functional group

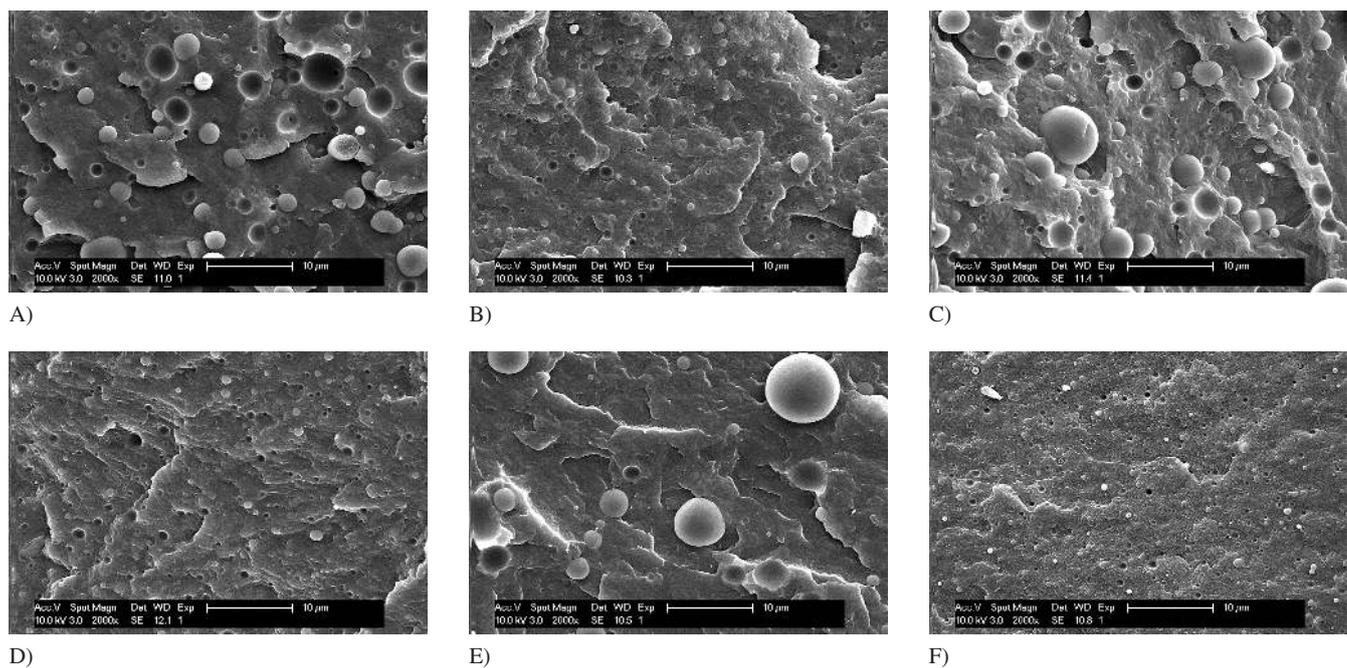


Fig. 2. SEM images of (A, C, E) PP:PET (85:15 wt%) blend excluding compatibilizer and (B, D, F) including 2.5 wt% SEBS-g-GMA as compatibilizer processed at the temperature profile of: A, B) 250°C, C, D) 270°C, E, F) 300°C

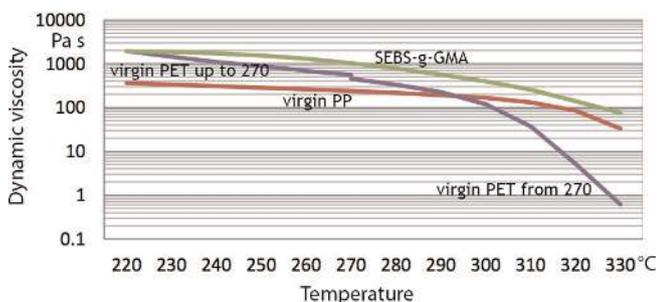


Fig. 3. Dependence of dynamic viscosity on the processing temperature profile. Measured at an angular frequency of 100 rad/s

degradation of PET is ongoing during the actual processing step, it is hard to predict by how far the effective viscosity curve of PET will be lowered, thus decreasing the temperature at which both curves intersect.

An indicative sampling of PET particle diameters is summarized in Table 4. It can be observed that, for the not compatibilized blends, the PET domains are not only significantly larger on average, but the spread on the particle sizes is very large, indicating insufficient homogeneous dispersion. For the tern-

ary blend including 2.5 wt% SEBS-g-GMA, a clear improvement in dispersion and reduction in domain size was observed at all processing temperatures compared to the blend without compatibilizer. In addition, a slight decrease in average particle size and an even more even distribution of the PET phase was obtained with an increase in processing temperature, contrary to the trend that was found without the compatibilizer. The improved morphology indicates an increased compatibilization with an increase in processing temperature and this corresponds well to the results of the mechanical impact test.

The contrary trend to the one that was observed for the binary blends suggests that another compatibilizing mechanism must be at work than just the approximation of viscosity values between PET and PP. In Fig. 3, the difference in viscosity of PP and SEBS-g-GMA indeed becomes smaller and approaches a ratio of one with increasing temperature. Moreover, interfacial tension is known to decrease with rising temperatures for larger molecular mass polymers (Wu, 1982; Biresaw et al., 2003). This results in an increased possibility of migration of SEBS-g-GMA into the PP matrix and towards the PP-PET interface (Babaei and Arefazar, 2014). Seeing how the impact strength increases significantly for higher compatibilizer content, while the respective decrease in domain size of the dispersed phase is only slight, it can therefore be deduced that the interfacial ad-

Figure	2A	2B	2C	2D	2E	2F
mean ( $\mu\text{m}$ )	2,54	1,00	2,50	0,96	2,77	0,76
stdev ( $\mu\text{m}$ )	1,07	0,49	1,50	0,27	2,10	0,16

Table 4. Indicative particle sizes (measured diameter) of PET domains in the PP matrix, per image from Fig. 2

hesion must have increased as well. This increased reactivity of the functional group towards the PET phase may result from two causes: 1) thermally enhanced activation of the reaction and a significant shift of the equilibrium towards the product side (Sun et al., 1996) or 2) increase in the amount of reactive groups of PET due to an increased thermal degradation and transesterification at higher processing temperatures (Dhavalikar et al., 2003). Moisture was excluded by the drying treatment of the materials, but no stabilizing agents (additional to those present in the standard PP) were added to the blend, so oxidative enhancement of the thermo-mechanically induced degradation during processing is quite possible (Al-AbdulRazzak and Jabarin, 2002; Assadi et al. 2004).

Overall, these results indicate an increase in compatibilization effect of the functionalized compatibilizer SEBS-g-GMA for PP-PET with an increase in processing temperature.

### 3.2 Compatibilizer Content

The study of this part concerns the combined compatibilization and impact modification effect. Based on the significantly improved compatibilization with increasing temperature, the highest processing temperature profile of 300 °C was used for the comparison of different compatibilizer contents.

The impact strengths for the blends including SEBS-g-GMA functionalized compatibilizer contents of 0, 0.25, 1 and 2.5 wt% are compared in Table 3. The values of these blends are compared to the one of pure PP processed at the temperature profile of 250 °C, which was used as reference. No significant change was obtained after the addition of 0.25 and 1 wt% SEBS-g-GMA compared to the value of the blend excluding compatibilizer. However, the addition of 2.5 wt% compatibilizer results in a steep increase of the impact strength and indicates a good compatibilization. This is due to the interfacial adhesion that follows from compatibilization, which allows for improved stress transfer across the interface between the PP matrix and the PET dispersed particles (Paul and Barlow, 1980; Robeson, 2007).

The SEM images of the blends including different contents of SEBS-g-GMA are provided in Fig. 4. A significant decrease in average domain size and a finer dispersion of the minor PET phase was obtained at all compatibilizer contents compared to the morphology of the blend excluding compatibilizer.

The addition of 1 wt% SEBS-g-GMA at a processing temperature profile of 300 °C results in the same order of average particle size compared to the morphology obtained after the addition of 2.5 wt% SEBS-g-GMA at a temperature profile of 250 and 270 °C. While this confirms the earlier results of a better dispersive and distributive mixing for the higher blending temperature of 300 °C, these results also give an insight into the role of the amount of compatibilizer used. As the impact strength for 1 wt% at 300 °C is significantly lower than those of the 2.5 wt% blends at 250 and 270 °C, it is implied that the difference in potential encapsulation of the PET particles by the compatibilizer also has an important effect on impact modification and that the 1 wt% compatibilizer is simply not enough to obtain improved surface interactivity (and thus improved impact properties) between the PP and PET phases throughout the entire blend. Additionally, the critical ligament thickness below 1 µm (Premphet and Paecharoenchai, 2002; Zhang et al., 2004; Lin et al., 2011) seems to be approached by the addition of 2.5 wt%. The observed steep increase in impact strength supports the impact modifying effect. As ligament thickness decreases, micro-fractures and shear bands will be less likely to occur, this increasing the blend's toughness (Bucknall and Paul, 2013). The brittle-to-ductile transition has not occurred yet since only a relatively small increase in impact strength was noticed, if we compare our results to the increase obtained in an earlier study (Bacci et al., 2013).

### 3.3 Structural Interactivity

The main objective of this step is the comparison of the compatibilizing effect of the three different functionalized compatibilizers: SEBS-g-GMA, POE-g-GMA and SEBS-g-MA. For comparison the blends are processed at the temperature profile of 300 °C and compared at a content of 2.5 wt%, based on the previous results.

Table 3 shows the results of the impact tests. The impact strength increases by the addition of all functionalized compatibilizers to the PP-PET blend. However, the increase is most significant for the addition of SEBS-g-GMA. The addition of SEBS-g-MA and POE-g-GMA resulted in roughly the same lower impact strength.

The SEM images of the blend including the functionalized compatibilizers are shown in Fig. 5. The addition of POE-g-

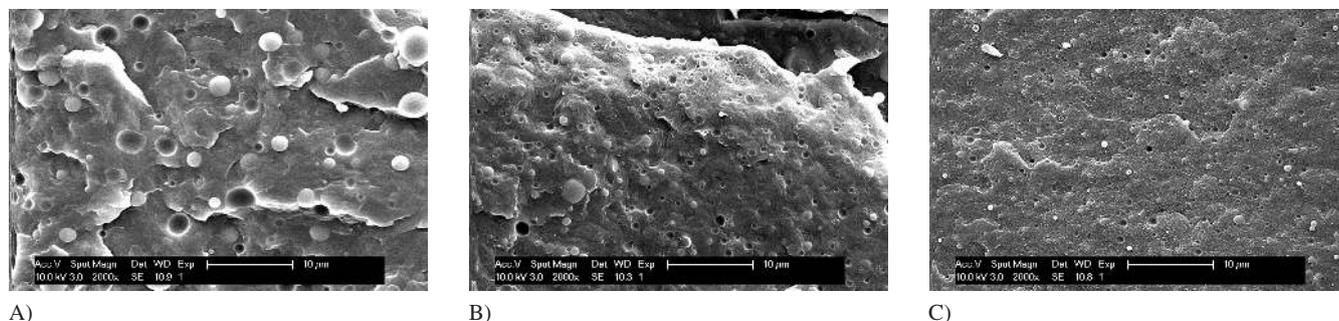


Fig. 4. SEM images of PP/PET (85:15 wt%) blend including (A) 0.25, (B) 1 and (C) 2.5 wt% SEBS-g-GMA processed at the temperature profile of 300 °C

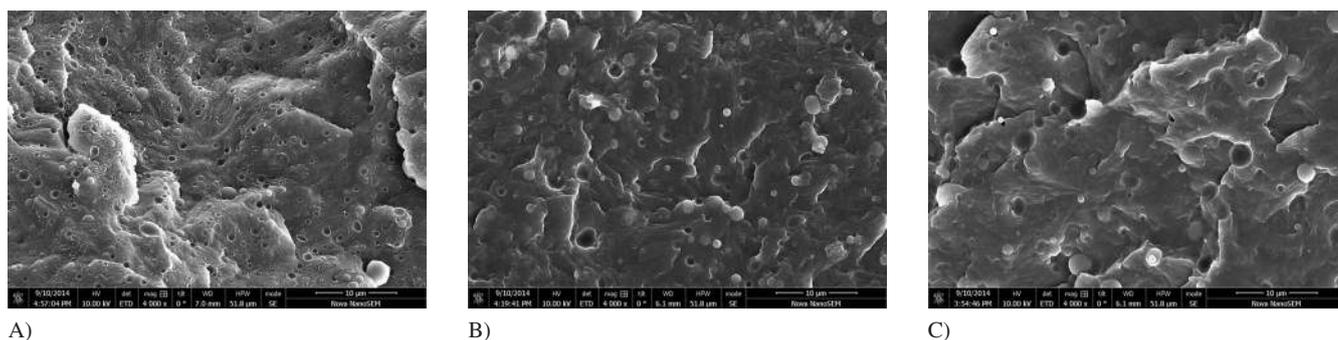


Fig. 5. SEM images of PP-PET blend including 2.5 wt% (A) SEBS-g-GMA, (B) SEBS-g-MA and (C) POE-g-GMA as compatibilizer processed at the temperature profile of 300 °C

GMA and SEBS-g-MA resulted in a more or less equal and finer morphology compared to the morphology of the blend excluding compatibilizer (Fig. 2). An even finer dispersion was obtained after the addition of SEBS-g-GMA. The SEM image of the blend including SEBS-g-GMA also shows a rougher interface, which can be related to a better interfacial adhesion (Babaei and Arefazar, 2014). These results, in combination with the results of the impact strength indicate that the best compatibilization effect can be obtained by the addition of SEBS-g-GMA.

More insight is obtained by analyzing the results of the rheology measurements. The results were analyzed at the lower frequencies since this is more sensitive to interfacial contributions and avoids contributions from molecular interactions that dominate at high frequencies.

Figure 6 illustrates the dynamic viscosity of the pure components, where the reference PET was also passed through the compounder at the temperature profile of 300 °C to take any possible degradation into account. The dynamic viscosities of all compatibilizers are large compared to PP and PET and also show a higher sensitivity to shear rate. The higher shear sensitivity can be a result of a combination of; 1) a higher molecular weight; 2) a broader molecular weight distribution; and/or 3) a more complex network structure. The lower dynamic viscosity

of SEBS-g-MA compared to SEBS-g-GMA can be related to a lower molecular weight or to the difference in functional group. GMA is expected to be more reactive towards itself compared to MA, based on their chemical structure, and therefore has a higher possibility for network formation.

The order of magnitude of the dynamic viscosity decreases after processing and blending PP and PET compared to the values of the pure components. This is most probably caused by the lower inter-chain interactions due to the high interfacial tension. However, the addition of only 2.5 wt% of one of the compatibilizers already results in a significant increase, which increase is not related to the order of dynamic viscosities of the pure compatibilizers. This supports the findings of an earlier study, that the interfacial tension plays a more important role in the determination of the compatibilization effect compared to the viscosity (Babaei and Arefazar, 2014). The dynamic viscosity of the blend depends not only on the viscosity and elasticity of the blend components but also on the interfacial interactions and therefore is a good measure for the effectiveness of compatibilization. The increase in dynamic viscosity after the addition of the compatibilizers can be related to the increased interfacial interaction and/or increase in interfacial area.

The order of dynamic viscosity of the binary and ternary blends can be related to the morphology: the blend excluding compatibilizers showed the coarsest morphology and had the lowest dynamic viscosity; the blend including SEBS-g-GMA showed the finest dispersion and had the highest dynamic viscosity. The level of dispersion and the value of the dynamic viscosity for the blend including POE-g-GMA and SEBS-g-MA are in between. The better interfacial adhesion for the blend including SEBS-g-GMA, based on the SEM images, is also supported by the large increase in dynamic viscosity and the impact testing results, the combination of which is typically seen as a validation of component interactivity in the blend (Heino et al., 1997; Khonakdar et al., 2013).

The influence of the interface also plays an important role in the dynamic storage modulus. In addition, the storage modulus can also indicate possible network formation that results in steady-state values at the lower frequencies via the standard linear solid model. The storage modulus can also be influenced by a changed contribution of the dispersed PET phase due to a change in interfacial adhesion.

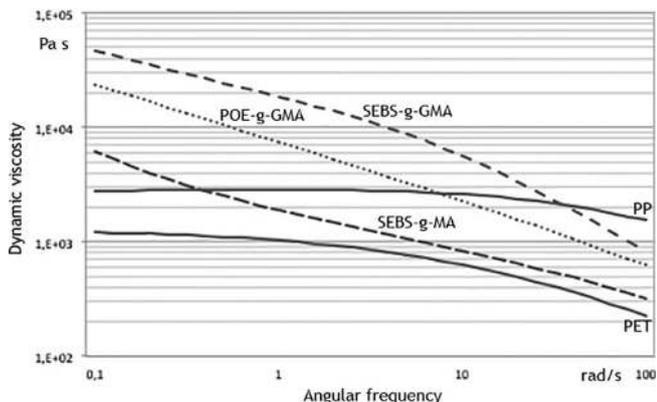


Fig. 6. Dynamic viscosity of pure components versus angular frequency, measured at a temperature of 220 °C

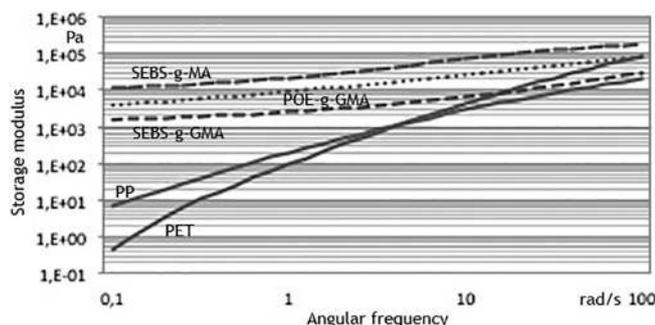


Fig. 7. Storage modulus of pure components versus angular frequency, measured at a temperature of 220°C

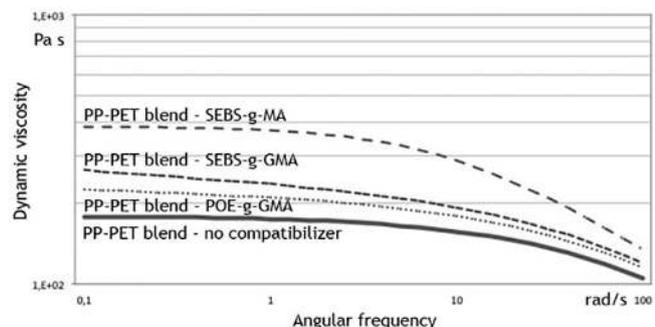


Fig. 8. Dynamic viscosity of PP-PET blend excluding and including different functionalized compatibilizers (SEBS-g-GMA, SEBS-g-MA, POE-g-GMA), processed at the temperature profile of 300°C, versus angular frequency, measured at a temperature of 220°C

The storage moduli of the pure components are shown in Fig. 7. The steady-state values of the compatibilizers at the lower frequency limit indicate no significant network formation. SEBS-g-MA and POE-GMA still show a small fraction of network formation after addition to the PP-PET blend as shown in Fig. 8. This is not the case after the addition of SEBS-g-GMA to the blend. This can be a result of a better encapsulation of the PET phase resulting in a smaller possibility for network formation. The lower storage modulus after the addition of SEBS-g-GMA can also be a result of a larger contribution of the PET phase, which as a pure material has a lower storage modulus compared to PP. The larger contribution can be caused by a better interfacial adhesion as was supported by the SEM images of Fig. 5.

#### 4 Conclusion

The first part of this study showed that the compatibilization effect of a PP:PET (85:15 wt%) blend by SEBS-g-GMA improves with an increase in processing temperature. This can be related to a combination of increased possibilities of migration towards the PP-PET interface and increase in reactivity of the compatibilizers towards the PET phase.

The second part showed that a minimal compatibilizer content is needed to have a significant compatibilization effect. This can partly be related to the impact modifying effect which

starts to play a role by the approach of the critical ligament thickness and can result in a brittle-to-ductile transition.

The last part showed that the compatibilization effect of a PP:PET (85:15 wt%) blend by a compatibilizer with a content of 2.5 wt% and at the processing temperature profile of 300°C was the highest for SEBS-g-GMA compared to SEBS-g-MA and POE-g-GMA. This can be the result of a larger reactivity of GMA compared to MA and larger possibility of migration of the compatibilizer towards the PP-PET interface of SEBS compared to POE as a result of the styrene blocks in SEBS that have a low interfacial adhesion with PET as well.

In conclusion can be said that the highest compatibilization effect can be obtained by an optimal combination of reactivity of the compatibilizer with the PET phase while also migration towards the PP-PET interface is needed. Extra studies are recommended to study the whole range of compatibilizer contents and its effect on reactivity, modification and migration rate.

#### References

- Al-Abdulrazzak, S., Jabarin, S. A., "Processing Characteristics of Poly(ethylene terephthalate): Hydrolytic and Thermal Degradation", *Polym. Int.*, **51**, 164–173 (2002), DOI:10.1002/pi.813
- Assadi, R., Colin, X. and Verdu, J., "Irreversible Structural Changes during PET Recycling by Extrusion", *Polymer*, **45**, 4403–4412 (2004), DOI:10.1016/j.polymer.2004.04.029
- Babaei, A., Arefazar, A., "Effect of Maleated Styrene/Ethylene-Co-Butylene/Styrene on the Dispersed Particles Size and Mechanical Properties of Polyamide 6/Poly(styrene-co-acrylonitrile)/Poly(styrene-B-(ethylene-co-butylene)-B-styrene) Ternary Blends", *J. Macromol. Sci. Part B Phys.*, **53**, 1406–1420 (2014), DOI:10.1080/00222348.2014.895634
- Bacci, D., Toaldo, A. and Scaini, M., "A Mathematical Formulation of the Brittle/Ductile Transition of Impact Modified Polymers", *J. Macromol. Sci. Part B Phys.*, **52**, 1438–1452 (2013), DOI:10.1080/00222348.2013.769838
- Biresaw, G., Carriere, C. G. and Sammler, R. L., "Effect of Temperature and Molecular Weight on the Interfacial Tension of PS/PDMS Blends", *Rheol. Acta*, **42**, 142–147 (2003), DOI:10.1007/s00397-002-0265-8
- Bucknall, C. B., Paul, D. R., "Notched Impact Behaviour of Polymer Blends: Part 2: Dependence of Critical Particle Size on Rubber Particle Volume Fraction", *Polymer*, **54**, 320–329 (2013), DOI:10.1016/j.polymer.2012.11.019
- Delva, L., Ragaert, K., Allaer, K., Gaspar-Cunha, A., Degrieck, J. and Cardon, L., "Influence of Twin-Screw Configuration on the Mechanical and Morphological Properties of Polypropylene – Clay Composites", *Int. J. Mater. Prod. Technol.*, ??Volume, Pages?? (2015)
- Dhavalikar, R., Yamaguchi M. and Xanthos, M., "Molecular and Structural Analysis of a Triepoxide-Modified Poly(ethylene terephthalate) from Rheological Data". *J. Polym. Sci., Part A: Polym. Chem.*, **41**, 958–969 (2003), DOI:10.1002/pola.10641
- Eastar Copolyesters Data Sheet, Eastman, ??Town, Country?? (2014)
- Environmental Protection Agency (EPA), "Opportunities to Reduce Greenhouse Gas Emissions through Materials and Land Management Practices", U.S. Environmental Protection Agency Office of Solid Waste and Emergency Response, ???Town, USA?? (2009)
- European Commission, "A European Strategy on Plastic Waste in the Environment", Green Paper, European Commission, Brussels (2013)
- Fu, S.-Y., Feng, X.-Q., Lauke, B. and Mai, Y.-W., "Effects of Particle Size, Particle/Matrix Interface Adhesion and Particle Loading on Mechanical Properties of Particulate-Polymer Composites", *Composites Part B-Engineering*, **39**, 933–961 (2008)

- Greco, R.: Impact Modifiers: (1) Mechanisms and Applications in Thermoplastics. Plastic Additives, Pritchard, G. (Ed.), Chapman and Hall, London, p. 375–385 (1998)
- Heino, M., Kirjava, J., Hietaoja, P. and Seppala, J., “Compatibilization of Polyethylene Terephthalate/Polypropylene Blends with Styrene-Ethylene/Butylene-Styrene (SEBS) Block Copolymers”, *J. Appl. Polym. Sci.*, **65**, 241–249 (1997), DOI:10.1002/(SICI)1097-4628(19970711)65:2<241::AID-APP4>3.0.CO;2-O
- Higgins, J. S., Lipson, J. E. G. and White, R. P., “A Simple Approach to Polymer Mixture Miscibility”, *Philos. Trans. R. Soc. London, Ser. A*, **368**, 1009–1025 (2010)
- Ihm, D., White, J., “Interfacial Tension of Polyethylene Polyethylene Terephthalate with Various Compatibilizing Agents”, *J. Appl. Polym. Sci.*, **60**, 1–7 (1996), DOI:10.1002/(SICI)1097-4628(19960404)60:1<1::AID-APP1>3.0.CO;2-T
- Jain, A., Pandey, G., Singh, A. K., Rajagopalan, V., Vaidyanathan, R. and Singh, R. P., “Fabrication of Structural Composites from Waste Carpet”, *Adv. Polym. Technol.*, **31**, 380–389 (2012), DOI:10.1002/adv.20261
- Khonakdar, H. A., Jafari, S. H., Mirzadeh, H., Kalaei, M. R., Zare, D. and Saehb, M. R., “Rheology-Morphology Correlation in PET/PP Blends: Influence of Type of Compatibilizer”, *J. Vinyl Add. Tech.*, **19**, 25–30 (2013), DOI:10.1002/vnl.20318
- Lei, Y., Wu, Q., Clemons, C. M. and Gu, W., “Phase Structure and Properties of Poly(ethylene terephthalate)/High-Density Polyethylene Based on Recycled Materials”, *J. Appl. Polym. Sci.*, **113**, 1710–1719 (2009), DOI:10.1002/app.30178
- Lin, Y., Chen, H., Chan, C. and Wu, J., “Effects of Coating Amount and Particle Concentration on the Impact Toughness of Polypropylene/CaCo<sub>3</sub> Nanocomposites”, *Eur. Polym. J.*, **47**, 294–304 (2011), DOI:10.1016/j.eurpolymj.2010.12.004
- Papadopoulou, C., Kalfoglou, N., “Comparison of Compatibilizer Effectiveness for PET/PP Blends: Their Mechanical, Thermal and Morphology Characterization”, *Polymer*, **41**, 2543–2555 (2000), DOI:10.1016/S0032-3861(99)00442-5
- Paul, D. R., Barlow, J. W., “Polymer Blends (or Alloys)”, *J. Macromol. Sci. Rev. Macromol. Chem. Phys.*, **C18**, 109–168 (1980), DOI:10.1080/00222358008080917
- Plastics Europe, “Plastics - the Facts 2014/2015. An Analysis of European Plastics Production, Demand and Waste Data”, Plastics Europe, Brussels (2015)
- Plastics Europe, EuPC, EuPR and EPRO, “Plastics - the Facts 2011. An Analysis of European Plastics Production, Demand and Recovery for 2010”, Plastics Europe, Brussels (2011)
- Pracella, M., Chionna, D., “Functionalization of Styrene-Olefin Block Copolymers by Melt Radical Grafting of Glycidyl Methacrylate and Reactive Blending with PET”, *Macromol. Symp.*, **218**, 173–182 (2001), DOI:10.1002/masy.200451418
- Pracella, M., Rolla, L., Chionna, D. and Galeski, A., “Compatibilization and Properties of Poly(ethylene terephthalate)/Polyethylene Blends Based on Recycled Materials”, *Macromol. Chem. Phys.*, **203**, 1473–1485 (2002), DOI:10.1002/1521-3935(200207)203:10/11<1473::AID-MACP1473>3.0.CO;2-4
- Premphet, K., Paecharoenchai, W., “Polypropylene/Metallocene Ethylene-Octene Copolymer Blends with a Bimodal Particle Size Distribution: Mechanical Properties and their Controlling Factors”, *J. Appl. Polym. Sci.*, **85**, 2412–2418 (2002), DOI:10.1002/app.10886
- Robeson, L., “Chapter 6 Properties of Polymer Blends”, in *Polymer Blends - A Comprehensive Review*, Hanser, Munich (2007), DOI:10.3139/9783446436503
- Ryan, A., “Polymer Science - Designer Polymer Blends”, *Nat. Mater.*, **1**, 8–10 (2002), DOI:10.1038/nmat720
- Sun, S., Zhang, M., Zhang, H. and Zhang, X., “Polylactide Toughening with Epoxy-Functionalized Grafted Acrylonitrile-Butadiene-Styrene Particles”, *J. Appl. Polym. Sci.*, **122**, 2992–2999 (2011), DOI:10.1002/app.34111
- Sun, Y., Hu, G., Lambla, M. and Kotlar, H., “In Situ Compatibilization of Polypropylene and Poly(butylene terephthalate) Polymer Blends by one-Step Reactive Extrusion”, *Polymer*, **37**, 4119–4127 (1996), DOI:10.1016/0032-3861(96)00229-7
- Van Krevelen, D., Te Nijenhuis, K.: *Interfacial Energy Properties. Properties of Polymers*, Elsevier, Amsterdam (2008)
- Wu, S.: *Polymer Interface and Adhesion*, Marcel Dekker, New York (1982)
- Wu, S. H., “Formation of Dispersed Phase in Incompatible Polymer Blends - Interfacial and Rheological Effects”, *Polym. Eng. Sci.*, **27**, 335–343 (1987), DOI:10.1002/pen.760270506
- Yildirim, E., Yurtsever, M., “A Comparative Study on the Efficiencies of Polyethylene Compatibilizers by Using Theoretical Methods”, *J. Polym. Res.*, **19** (2012)
- Zhang, L., Li, C. and Huang, R., “Toughness Mechanism in Polypropylene Composites: Polypropylene Toughened with Elastomer and Calcium Carbonate”, *J. Polym. Sci. Part B: Polym. Phys.*, **42**, 1656–1662 (2004), DOI:10.1002/polb.20035

*Date received: May 19, 2015*

*Date accepted: November 29, 2015*

Bibliography  
DOI 10.3139/217.3124  
Intern. Polymer Processing  
XXXI (2016) 2; page 179–187  
© Carl Hanser Verlag GmbH & Co. KG  
ISSN 0930-777X