MECHANICAL BEHAVIOUR OF GLASS FIBRE REINFORCED THERMOPLASTIC TAPES MADE OF RECYCLED POLYMERS

Franziska Regel[1], Nicole Claussnitzer[1,2] and Ferrie W.J. van Hattum[1]

[1]Group of Lightweight Structures, Saxion University of Applied Sciences
M.H. Tromplaan 28, 7513 AB Enschede, The Netherlands
Email: f.regel@saxion.nl, web page: http://www.saxion.nl/lightweight

[2]Lightweight Structures and Polymer Technology, Chemnitz University of Technology
Str. der Nationen 62, 09111 Chemnitz, Germany

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ABSTRACT

Plastics are relatively inexpensive, lightweight and durable materials, which can be readily moulded into various products. However, current levels of their usage and disposal generate severe environmental problems. Recycling is one method for reducing their environmental impact. Despite there are still many obstacles that restrict recycling, such as the difficulty to obtain polymers in nearly pure composition particularly from post-consumer waste. In a continuous fibre reinforced composite material, fibres are substantially responsible for the desired stiffness and strength: any effect on the mechanical properties due to non-virgin matrix properties are expected to be marginal.

The main objective of this study was to evaluate the potential of recycled polymers as matrix material for glass fibre reinforced thermoplastic tapes. The polymer materials studied were a virgin polypropylene and various post-consumer and post-industrial recyclates predominantly made of polypropylene. Additionally blends with a masterbatch featuring a combination of heat stabilisers and coupling agents were prepared to investigate its effect on processability and properties. Unidirectional preforms were manufactured by tape winding and compression moulded to composite plates. Four point bending, interlaminar shear strength and Charpy impact tests were conducted to analyse the mechanical behaviour. Overall, the current study proves the feasibility of manufacturing glass fibre reinforced thermoplastic tapes using recyclates. Mechanical properties of tapes made of recycled polymers are mostly at the same order of magnitude as of the virgin polymer. Values obtained for some polymers containing additive could be further increased. The results indicate the potential of using recycled polymers for cost-efficient and sustainable tape manufacturing without a loss of properties.

1 INTRODUCTION

Plastics have become one of the most universally used materials in global industries. Polypropylene (PP) and polyethylene (PE) account for about 50% with an annual demand of about 23 Mt of the total European plastic consumption of 49 Mt in 2015 [1]. The use and demand of PP and PE is steadily increasing due to their versatility, good processability, mechanical properties and low costs. However, current levels of their usage and disposal generate severe environmental problems. One of the reasons why thermoplastics are preferred over thermosets is their better recyclability. Recycling of plastics is one method for reducing environmental impact and resource depletion. It provides opportunities to reduce oil usage, CO_2 emissions and quantities of waste requiring disposal in order to comply with European directives and regulations [2, 3]. However, the recycling process of post-consumer waste proved to be complicated. Due to thermo-mechanical degradation during processing the mechanical properties of recyclates generally do not meet virgin material properties [4]. Maintaining a constant quality of recycled materials coming from consumers is difficult because of the many polymer types and grades used. Furthermore small levels of pollution are potentially affecting processability and properties. In industry therefore, a large effort is made to separate materials and obtain recycled polymers as clean and pure as possible, thus adding costs to the material price, which is generally still lower than its virgin counterpart.

High-quality unidirectional (UD) fibre reinforced polymeric tapes are widely applied in the aerospace industry using automated placement processes. However, the need for economical thermoplastic composite (TPC) materials in industrial applications outside high performance areas is rapidly expanding. The weight saving trend in the automotive sector is increasingly important due to more stringent CO_2 emission policies. Short and long fibre reinforced thermoplastics have made a major contribution to lightweight components. However, due to fundamental material limitations the automotive industry is steadily investigating overmoulding of continuous fibre reinforced composites to reduce the total material amount and costs, improve the thermo-mechanical performance, and even increase processing speeds [5-7]. Tape material can be used as local UD reinforcement or to produce tailored blank laminates with various patterns. Thus, TPC tapes will advance solutions for transportation and consumer goods markets. A potential application is the lining of pipes and pressure vessels due to the chemical and permeation resistance of thermoplastics.

In fibre reinforced materials the fibres are substantially responsible for the desired material's stiffness and strength, while the polymeric matrix binds the fibres, serves to transfer loads and provides in particular its shear strength. Thus, the effect of potentially lower properties of recycled polymers might be less critical for the final material's performance. However, final properties are mainly determined by the strength of the fibre-polymer interphase. When the adhesion to the matrix material is weak fibres do not act as effective reinforcement. High adhesive strength can only be expected when strong polar interactions or chemical bonds exist. Therefore, various studies have focused on methods to increase the compatibility of fibre and matrix surfaces by fibre surface treatments using coupling agents or changes in the polymer matrix such as alkali treatments [8, 9].

It is known that the melt viscosity of a polymer generally decreases due to recycling, which is attributed to a decrease of its molecular weight [10]. A loss in chain length results in weaker intermolecular bonds, thus the material flows easier and the melt flow index (MFI) is higher. Hence, for the production of a fibre reinforced thermoplastic tape is the decreased viscosity of post-consumer polymers actually beneficial. On the contrary, the lower the MFI, the higher the viscosity, and consequently the more difficult the impregnation of a fibre roving. However with regard to mechanical properties, a lower MFI results in higher elongation and toughness as long and more branched chains entangle more due to stronger intermolecular forces [11].

The main objective of this study was to evaluate the potential of recycled post-industrial and post-consumer polymers as matrix material for glass fibre (GF) reinforced thermoplastic tapes. The mechanical performance of tapes made of recycled PPs was compared with a tape made of virgin PP. Moreover, the findings from mechanical testing were used to investigate the effect of an additive on the tape properties.

2 EXPERIMENTAL METHODS

2.1 Materials and manufacturing

The polymer materials in this study were a virgin polymer, and various post-consumer and post-industrial recyclates, as listed in Table 1. Special emphasis in the selection was given on the relationship between the MFI, purity and processability. Although the base polymer of all materials was PP, the recyclates would contain a certain amount (typically below 10%) of PE. Moplen is the virgin material of choice in regular tape manufacturing and hence used as a benchmark for the recycled polymers.

Additionally, blends of each polymer with the masterbatch ADD-VANCE THC 831 (produced and supplied by Addcomp) were prepared to analyse its effect on processability and properties. As recommended by the supplier the blends contained 4 wt% of additive. ADD-VANCE THC 831 is a masterbatch developed for producing long fibre reinforced PP components in the automotive industry. It features a combination of heat stabilisers and coupling agents, aimed at increasing bonding, thermal and UV stability. The coupling agents provide a strong and stable bond between the polymer matrix and the sizing of the glass fibres. Blends of polymers and additive are referred to in the following as Polymer4831.

Name	Designation	Colour	MFI
Moplen RP348U	Moplen	Black	75 g/10min*
Veolia PPC BL5005XB	Veolia	Black	46-54 g/10min*
QCP ExPP 158A	ExPP	Grey	70 g/10min*
Monogrind	Monogrind	All	n.a.
Resinex	Resinex	Black	n.a.

Table 1. Characteristics of polymers used (*ISO 1133, 230°C, 2.16 kg)

Apart from Monogrind all polymers were supplied as granules. The Monogrind recyclate was supplied in flake-form, offering a higher potential for cost reduction as it is uncompounded. It can be seen in Fig. 1(c) that the material is more heterogeneous as compared to polymers supplied in granular shape.

(a) (b) (c)

Figure 1. Samples of polymeric materials: Veolia (a), ExPP (b) and Monogrind (c)

Continuous fibre reinforced thermoplastic tapes were produced and supplied by CompTape BV. In comparison to high-quality tapes used in aerospace applications, the produced tapes are characterised by a lower smoothness, narrow tolerances on width and thickness, and a fibre mass fraction of about 60 wt%. The PPG TufRov 4599 glass fibre was used as reinforcing fibre, which is specially tailored for the use in PP matrices. The manufactured tape had a width of about 7.0 mm and a thickness of about 0.4 mm.

(a) (b)

Figure 2. Compression moulding of plates made of fibre reinforced thermoplastic tape: wound plate placed in mould (a), compressed plate (b)

Tape winding was used to manufacture UD reinforced preforms. To produce a composite plate with a thickness of about 2 mm six layers of tape were needed. The wound preforms were transferred to a compression mould for forming and consolidation of fibre reinforced thermoplastic plates (Fig. 2).

The GF reinforced PP was heated up to a temperature of 230°C for 8 min, and thereafter compressed for 2 min with a pressure of 0.25 MPa.

2.2 Calcination

Calcination tests were carried out according to EN ISO 1172 to determine the fibre volume fraction (V_f) of the manufactured plates. Specimens were cut from each plate with dimensions of about 50 x 25 x 2 mm^3, which corresponds to a mass of 3-4 g. The density values used for the GF were 2.55 g/cm^3 and for the PP 0.9 g/cm^3. The V_f was determined to relate the fibre content to material properties and further used to analytically calculate the elastic modulus.

2.3 Four point bending test

The flexural behaviour was analysed according to the EN ISO 14125 four point bending test. A universal testing machine with a 50 kN load cell was used. The tests were performed at a displacement rate of 2 mm/min. Six specimens were cut from each plate with standard dimensions compliant with class III for UD composite materials. The specimen length coincided with the longitudinal fibre direction. The measured data were used to calculate flexural moduli as well as the flexural stress-strain behaviour.

2.4 Interlaminar shear strength test

Interlaminar shear strength (ILSS) testing was carried out using the short beam shear test according to EN ISO 14130. It was aimed at studying the interlaminar shear behaviour of the different recycled polymer tapes and evaluate the composite plate quality. Six specimens were machined from the same composite panels used for the four point bending specimens. A universal testing machine was used to perform ILSS tests at a displacement rate of 1 mm/min. The measured data were used to calculate the apparent interlaminar shear strength.

2.5 Charpy impact test

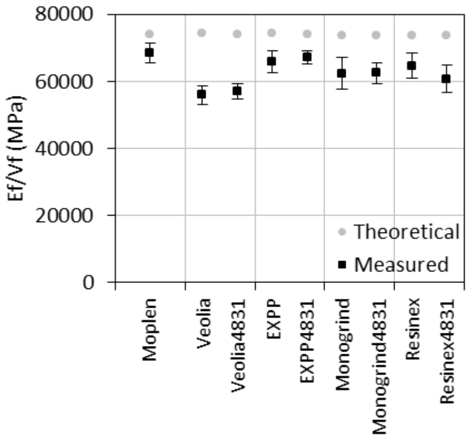
Charpy impact tests were conducted according to EN ISO 179-1 to evaluate the impact behaviour of the different thermoplastic tapes. Five unnotched specimens were subjected to flatwise impact using a test machine equipped with a 4 J hammer. The measured data were used to calculate the Charpy impact strength.

3 RESULTS

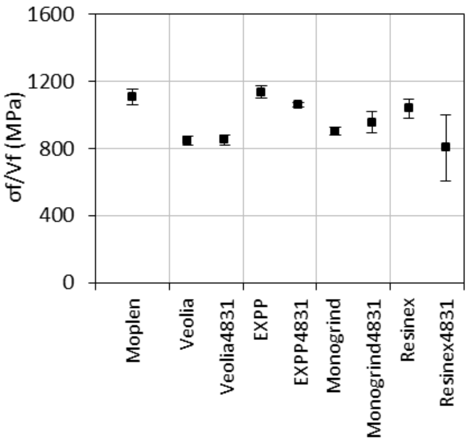
The study showed that GF reinforced thermoplastic tapes made of post-consumer and post-industrial recyclates can be readily produced without major obstacles. The processing parameters used for the virgin material could be applied for the recycled materials without major changes. The only obstacle observed was the granular flow in the extruder hopper for recyclates with irregular shapes and sizes, which was easily solved by additional size reduction. Composite plates were manufactured without major problems. Visually observed quality differences can be related to the impregnation process and the polymer viscosity. However, aesthetical requirements like smoothness and colour are often not relevant for the type of (semi-) finished products made of thermoplastic tapes.

The weight fraction (W_f) of the supplied thermoplastic tapes was given as 60 wt%. A V_f of 33.5%, which corresponds to a W_f of about 59 wt%, was determined for the tape made of virgin PP. The V_f of the plates made of post-consumer recyclates were in the range of 29% to 39%, which corresponds to W_f of 54wt% to 64wt%. The lowest value was determined for the plate made of Veolia and the highest value for the plate made of Resinex with additive. Though variations exist, related to the processability of the material, still the average is relatively close to the benchmark value of the Moplen tapes.

Flexural properties are presented in Fig. 3. For a better comparison of the different TPC the moduli as well as the flexural strengths were normalised over the determined V_f for each material. The values used to determine the theoretical moduli with the Rule of Mixtures (RoM) were elastic moduli of 72 GPa and 1 GPa for the GF and for the matrix material, respectively. The measured flexural moduli are between 16.4 GPa and 24.3 GPa. The flexural strengths are in the range of 250 MPa and 380 MPa. Small variations can be observed between the different materials, but most of them actually are at the same level as the benchmark material. The value obtained for the Moplen material agrees well with the analytical prediction, whereas the measured moduli of the post-consumer recyclate composites are lower than the analytically predicted values. Assuming that processing parameters and fibre orientation are similar for all wound plates and differences in fibre content was accounted for, these differences can be likely attributed to differences in material quality or deviation of the actual recycled polymer property from the assumed value. The TPC made of Veolia presents the lowest flexural properties. Modulus and strength are 18% and 23% lower, respectively, compared to the virgin material. The data obtained for the Monogrind and Resinex show the greatest degree of variability. In particular, the flexural strength of the plates made of Resinex with additive show the largest deviation (Fig. 3(b)). While the additive seems to improve the flexural moduli mostly, an obvious improvement of the flexural strength due to its presence cannot be observed.



(a)



(b)

Figure 3. Normalised flexural properties determined for plates made of GF reinforced thermoplastic tapes: flexural modulus (a), flexural strength (b)

Fig. 4 presents representative flexural stress versus flexural strain curves for all wound plates. It can be seen that the maximum flexural strain of the Moplen material is the highest. As reported in literature [4] recycled PP exhibits a higher crystallinity compared to virgin PP. Additionally the average molecular weight decreases due to (re)processing, which results in drops of the elongation at break. However, it can be seen that the flexural strain can be partially improved because of the additive.

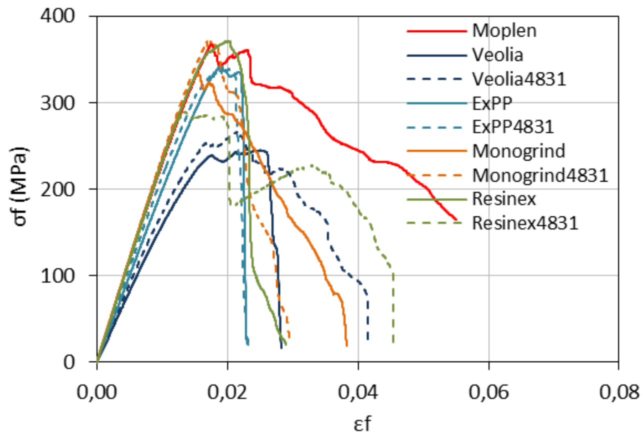


Figure 4. Representative flexural stress vs. flexural strain curves for plates made of GF reinforced thermoplastic tapes

Fig. 5 presents ILSS obtained for all manufactured plates. The ILSS values, which are in the range of 20 MPa and 25 MPa, were normalised over the V_f. The ILSS of plates made of post-consumer recyclates are similar to the ILSS of the benchmark plate made of Moplen tape. The selected additive only increased the values obtained for plates made of ExPP. As observed for the flexural strength, the deviation of the ILSS of the Resinex with additive is distinctly large.

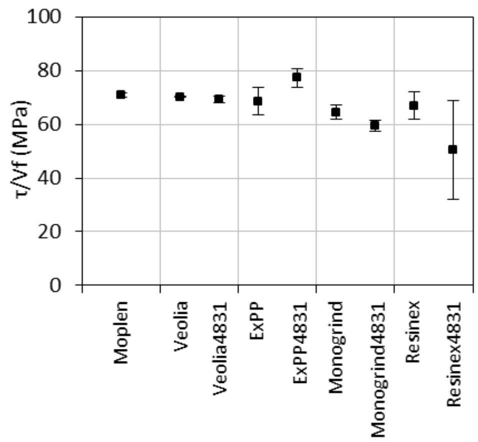


Figure 5. Normalised ILSS determined for plates made of GF reinforced thermoplastic tapes

Fig. 6 presents normalised Charpy impact strengths for all manufactured plates. The values obtained are in the range of 63 kJ/m^2 and 109 kJ/m^2. The impact strengths of all plates made of post-consumer recyclates are lower than the impact strength of the benchmark plate made of Moplen tape. An improvement of the impact strength due to the additive can only be observed for plates made of ExPP. The impact results show a distinct decrease in fracture toughness for recycled PPs. According to Meijer [12] a high molecular weight is beneficial for the strain-to-break and impact strength of polymers. As reported by Aurrekoetxa et al. [11] and Elloumi et al. [13], the observed changes can be thus related to a decrease in molecular weight and chain length. Long chains contain more C-C linkages to stretch. A reduction in chain length due to reprocessing causes failure to occur at smaller strains and facilitates further crystallisation.

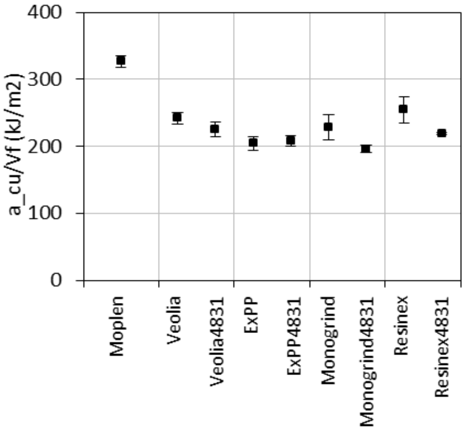


Figure 6. Normalised Charpy impact strength determined for plates made of GF reinforced thermoplastic tapes

4 CONCLUSIONS

The current study proves the feasibility of manufacturing GF reinforced thermoplastic tapes using post-consumer and post-industrial recyclates. Flexural properties and ILSS of thermoplastic tapes made of recycled polymers are at the same order of magnitude as of the virgin polymer. However, the Charpy impact strength of all tapes made of recyclates is lower than the strength obtained for the virgin material. The experimental value of the flexural modulus of the virgin material agrees well with the analytical prediction of the elastic modulus, whereas the measured moduli of the composites made of post-consumer recyclates are lower than their analytically predicted values. Assuming that processing parameters, fibre content and fibre orientation are similar for all TPC plates, differences can be attributed to lower moduli of the recycled polymers. The impact strengths of all plates made of post-consumer recyclates are lower than the impact strength of the virgin PP. The addition of the masterbatch ADD-VANCE THC 831 can improve the flexural modulus and ILSS of some TPC. However the fracture strain is for all recycled polymers lower than the strain observed for the virgin material. Overall, the results indicate the potential of using recycled polymers for cost-efficient and sustainable tape manufacturing without major property loss.

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