UPCYCLING WASTE PLASTICS IN CONTINUOUS FIBRE REINFORCED COMPOSITES

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ABSTRACT

In general, recycled plastics have reduced mechanical properties as compared to virgin, thus often a lower market value. By using plastic waste streams as a matrix in fibre reinforced products, these products can be manufactured even cheaper, potentially with near-virgin properties. This research therefore looks at producing composites from continuous fibre reinforced tapes made from recyclates (rTapes).
Polypropylene (PP), polyethylene (PE) and polyester (PET) waste streams are characterised in terms of rheology, effect of contamination and overall mechanical behaviour of the rTape UD-composites and over moulded rTapes.
Bending and ILSS tests on the compression moulded rTape UD-composites, show no significant difference between using virgin material or recyclates. Bending test on overmoulded rTapes compared to unreinforced bars, showed the difference a reinforcement makes. A study on the effect of contamination shows a huge improvement of mechanical properties compared to its unreinforced samples, although mechanical properties were noticeable lower than reinforced virgin polymers.
This research thus shows that it is possible to produce rTape from plastic waste, resulting in cheaper composites with competitive mechanical properties, in comparison to their virgin counterparts. As such it shows the potential of plastic recycling with added value besides potential lower CO2-footprint and cost reduction in thermoplastic composites manufacturing.

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1. INTRODUCTION

1.1 Paper introduction

In this paper the research into different waste streams and the possibility to reinforce them with glass fibers is reported. Recycled plastics have in general poorer mechanical properties. By using the material as a matrix for fibre reinforced composites, the application possibilities are greater. This paper looks at different waste streams (PP, PE and PET), rTape production, and testing of matrix material and Tapes. In further research the possibilities of inserts and different processing techniques will be researched. The main goal is to show the differences between virgin material and similar recycled material, and the possibilities with waste materials.

1.2 Background

Thermoplastics are relatively inexpensive, lightweight and durable materials, which can be readily moulded into various products. The largest amount of polypropylene (PP) and polyethylene (PE) is found in non-structural applications such as packaging and electrics. [1]

In 2018 packaging waste generated was estimated at 174 kg per inhabitant in Europe. 'Plastic' waste is after 'paper and cardboard' the biggest group, 19% with 14.795.620 tons total [2].

The industrial interest in continuous fibre reinforced thermoplastic composites, particularly in the aerospace and automotive sector, is constantly increasing. In a composite material the fibres typically provide stiffness and strength, whereas the polymer ensures shape, load transfer, chemical resistance and appearance. By locally stiffening thermoplastic products, the material thickness can be reduced, which results in less material needed. Previous research showed that composite tapes can be produced from PP recyclates, and that weakening in recycled polymer parts due to contaminations can be compensated by using tape inserts [3].

1.3 Fibrerec project

One of the obstacles in the previous research, was the granulometry of the recyclate to assure a stable flow during tape production. Flexural properties of the recycled material compared with virgin material were of the same order of magnitude. This study proved the feasibility of manufacturing rTapes with PP. The current research has been extended to include PP, PE and Polyester (PET) recyclates. The goal is to define what kind of waste streams, can be used to produce rTapes, and compare them with a virgin benchmark. Apart from the production of rTapes, insert design and production are also researched, but are not part of this paper. The end goal of the entire research, is a demonstrator part produced with a rTape insert and overmoulded with a recycled thermoplastic matrix material.

2. EXPERIMENTATION

2.1 Materials

Three types of matrix material are researched: PP, PE and PET, Table 1. Advantex OCV E-CR glass T-30 2400 tex is used as a fibre for all materials, with a single filament tensile strength between 3,1 and 3,8 MPa [4].

Table 1: Thermoplastic Materials

Thermoplastic Materials	Origin
PP – Polypropylene	
PP (virgin)	Benchmark
rPP	Post-consumer rigid plastic. Blend of different sorts of PP. Contains < 10% PE[1]
PEPP 50/50 (virgin)	Mix of 50% PE and 50% PP, mixed by hand
rPEPP 20/80	Post-consumer rigid plastic. Blend of different sorts of PE and PP. Contains < 20% PE, on average 14%[1]
PE – Polyethylene	
LDPE (virgin)	Benchmark
HDPE (virgin)	Benchmark
rPE	Post-consumer rigid plastic. Blend of different sorts of PE. Contains < 10% PP[1]
rPE food packaging	Collected, separated and cleaned food packaging. In pellet form. Contains 12 % aluminum[2]
PET – Polyester	
PET (virgin)	Benchmark
rPET	Post-consumer plastic[1]

rPET bottles	Post-consumer plastic bottle[1]
rPET textile	Post-consumer plastic from textile waste[3]

[1]Supplied by Van Werven BV, the Netherlands
[2]Supplied by Veolia BV, the Netherlands
[3]Supplied by Frankenhuis BV, the Netherlands

2.2 Physical testing

2.2.1 Melt Flow Index

In this research, the Melt Flow Index (MFI) is used as an indicator for the processability during the tape production: an MFI between 50-70 g/10 min is considered ideal. Higher or lower values result in a lower quality tape because of impregnation problems.

MFI testing is done according to EN ISO 1133:1999 [5], on a Kason XNR 400A MFI tester. Per material type, 3-4 batched are prepared of 5-6 grams of material. The test is done with the prescribed temperature, for PP 230 °C, for PE 190 °C and for PET 270 °C, and a load of 2,16 kg.

2.3 Mechanical testing

Mechanical testing is done to create a clear overview of the properties of the different materials, compare them to their benchmark, and compared them to each other or other waste stream.

2.3.1 Three-point bending laminates rTapes

To determine the bending modulus and bending strength three-point bending is done on consolidated specimens made of rTapes. Tests are done according to EN ISO 14125:1998 [6], on a Testometric M500-100CT with a three-point bending setup and a load cell of 100 kN. First, bars are compression moulded from tapes at 230°C for 2 minutes, followed by slowly cooling to room temperature (260°C for PET) using a 200x20 mm mould, by stacking the tapes on top of each other to achieve the required thickness. Out of this specimen the desired test specimens are cut. For each sample, a minimum of five specimens is used. A test speed of 2 mm/min and a preload of 20 N is used. The test will be stopped at 10% drop in force. The main goal of these tests is to determine the bending modulus and strength.

2.3.2 Three-point bending overmoulded rTapes

To determine the bending modulus and bending strength, three-point bending is done on specimens with and without inserts made from rTapes, overmoulded with a matching matrix material. Tests are done according to EN ISO 14125:1998 [6], on a Testometric M500-100CT with a three-point bending setup and a load cell of 5 kN. First samples are injection moulded without reinforcement, and to show the reinforcing effect of the same matrix material, a rTape insert is added to the injection molding process. Specimen sizes of 64x10,3x4 (lxwxh) are on average taken from the samples. For each sample, a minimum of five specimens is tested. A test speed of 2 mm/min and a preload of 20 N is used. The test will be stopped at 10% drop in force. The main goal of these tests is to determine the bending modulus and strength of the reinforced and unreinforced samples.

2.3.3 Interlaminar shear strength

To determine the interlaminar shear strength, tests are done according to EN ISO 14130:1997 [7], executed on a Testometric M500-100CT with a load cell of 100 kN. Specimen size of 20x10x2 (lxwxh) mm were taken from the moulded plates as described above. A span (L) of 10 mm, a test speed of 1 mm/min and a preload of 20 N is used. At least five specimens are tested for each sample.

3. RESULTS

3.1 Melt flow index results

The MFI results are shown in Figure 2 and Figure 1. For the **PP (**Figure 2), it was found that:

- Virgin PP has the highest MFI;
- rPP and rPEPP (20/80) have more or less the same value (around 20g/10 min);
- PEPP (50/50) MFI is around 40 g/10 min: the high content of HDPE affects the increasing of the viscosity of the blend system.

Different in-house studies revealed possibilities to increase the MFI, for example by blending virgin PP and rPP. Increasing the amount of virgin PP, resulted in a linearly decreasing viscosity. Another possibility is using the additive Zebra – flow T056 (5% peroxide concentrated), provided by Zebra-Chem Gmbh: by adding 1 wt% of the additive during extrusion, the MFI of the blend increases by 60%.

For the **PE** (Figure 2) it was found that the MFI of the virgin HDPE is seven times greater than the recycled PEs. Mixing virgin HDPE with rPE does not give a big change in the MFI. This can be due to presence of strong bonds within the chains or to the absence of compatibilizers.

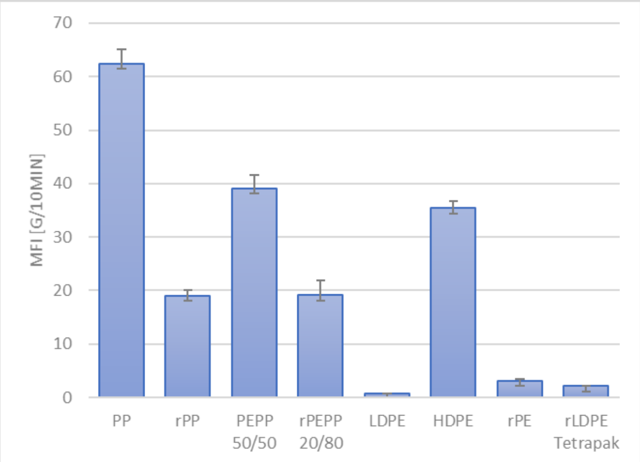

Figure 2: MFI PP and PET

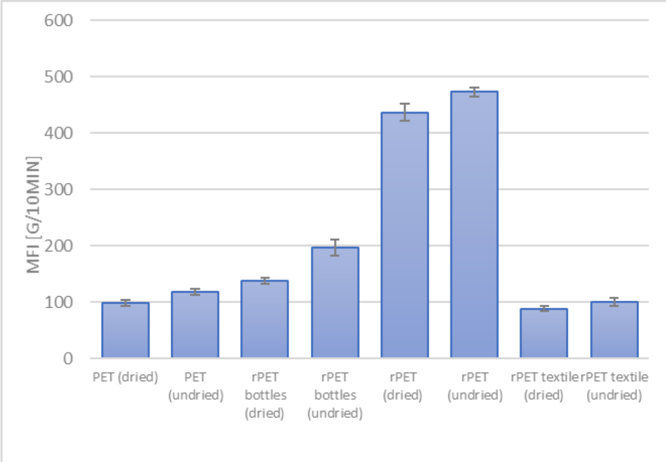

Figure 1: MFI PET

For the **PET** (Figure 1) it was found that:

- Virgin PET and rPET textile have the same MFI value (around 100 g/10 min),
- rPET bottle and rPET have very high MFI (> 400 g/10 min).

Since moisture content affects the MFI results, the difference between dried PET and undried PET is also shown: as expected, removing water is important, since this acts as a plasticizer.

In general, recycled PE and PP have lower MFI than the virgin ones, instead recycled PET has higher MFI than the virgin. This is due to the degradation of the thermoplastics (physical aging) and to the chain scission reactions, that produce smaller polymer molecules. During the aging of the polymer two phenomena can occur in the polymer chain: (1) a cross-linking or (2) chain scission reaction. If cross-linking occurs, more bonds among polymers are formed and this tends to decrease the MFI value (increase in viscosity). If chain scission occurs, the molecular weight of the chains decreases and thus results in an increase in MFI value.

3.2 Tape production results

From the selected materials four types had an MFI too low or high for use in the tape production process: LDPE, rPE, rPE food packaging, rPET. For the different types of PE, the recommendation would be to use an additive to increase the MFI. This rPET proved not to be suitable for this process, because of its significantly higher MFI.

Most materials were processable without any issues. rPP and rPEPP 20/80 rTapes were producible but looked visually less successful than the other tapes. In previous research an additive was used for the rPP tapes, this improved the quality drastically, and improved the mechanical testing results.

3.3 Three-point bending results rTape laminates

The introduction of glass fibres in the polymeric matrix increased the flexural moduli, compared to the pure polymer. In general, in terms of bending modulus (Figure 4). Recycled rTapes have a lower E modulus then virgin Tapes;

- GPP shows the highest bending modulus, so is the stiffer material;
- GrPET and GPEPP present the lowest bending moduli;
- GPEPP (50/50) and GrPEPP (20/80) have comparable bending modulus, even though one is virgin and one is recycled material. This is due to the different percentage PP.

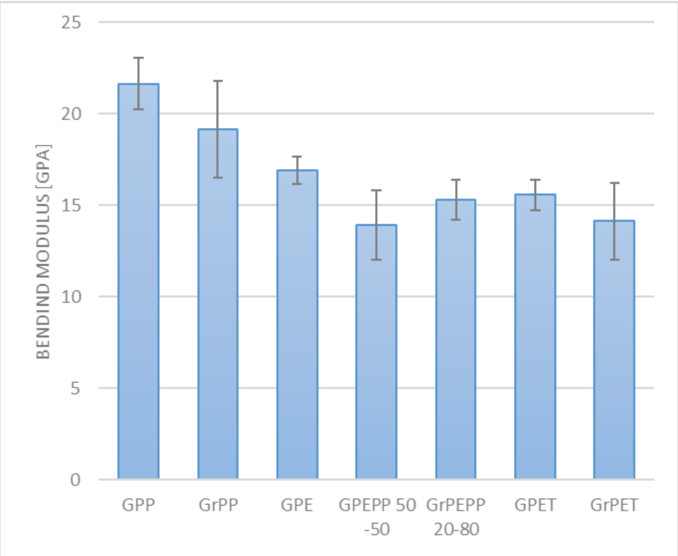

Figure 4: Bending modulus.

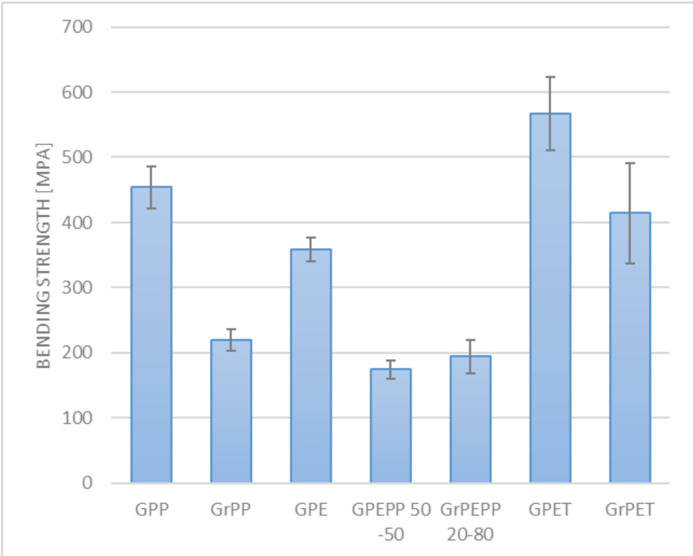

Figure 3: Bending strength.

In terms of bending strength (Figure 3):

- The bending strength of the rTape is lower than the bending strength of the virgin Tape;
- GPET and GPP have higher bending strength and GPEPP has lower bending strength.

The best performing material in bending modulus, bending strength and ILSS is the virgin GPP, as expected. The worst performing material in bending modulus, bending strength and ILSS is the virgin GPEPP (50/50). Some materials show lower bending strength, which is most likely attributed to poor adhesion between fibres and matrix. Currently, no coupling agents were used to improve the mechanical properties.

In general, recycled materials show lower bending modulus than the virgin ones. This is due to the physical aging, degradation and further thermal and mechanical treatment suffered by the material over time, that strongly decrease the mechanical properties.

3.4 Three-point bending results overmoulded rTapes

With these test results a comparison between unreinforced and reinforced material and virgin vs recycled can be observed.

In general, for PP in terms of bending modulus and strength (Figure 5 and Figure 6):

- **GPP vs. PP**: Bending modulus increases by a factor of 3,5 and the strength 1,2;
- **GrPP vs. rPP**: Bending modulus increases by a factor of 2,15 and the strength 0,85;
- **GPP vs. GrPP**: As expected, the recycled material performs less than the virgin in both bending modulus (factor 0,63) and bending strength (0,51);

In general, for PE in terms of bending modulus and strength (Figure 5 and Figure 6):

- **GPE vs. PE**: Bending modulus increases by a factor of 6,25 and the strength 2,67;

In general, for PEPP blends in terms of bending modulus and strength (Figure 5 and 6):

- **GPEPP vs. PEPP**: Bending modulus increases by a factor of 1,83 strength 0,96;
- **GrPEPP vs. rPEPP**: Bending modulus increases by a factor of 1,97 and the strength 0,63;
- **GPEPP vs. GrPEPP**: In this case the recycled blend performs better than the virgin blend, maybe due to the lower percentage of PE in the recycled blend.

In general, for PET in terms of bending modulus and strength (Figure 5 and Figure 6):

- **GPET vs. PET**: Bending modulus increases by a factor of 1,44 and the strength 0,8;
- **GrPET (tex) vs. rPET (tex)**: Bending modulus increases by a factor of 1,8 and the strength 0,86;
- **GPET vs. GrPET (tex)**: In this case the recycled blend performs better than the virgin blend based on the bending modulus, and the bending strength performs less.

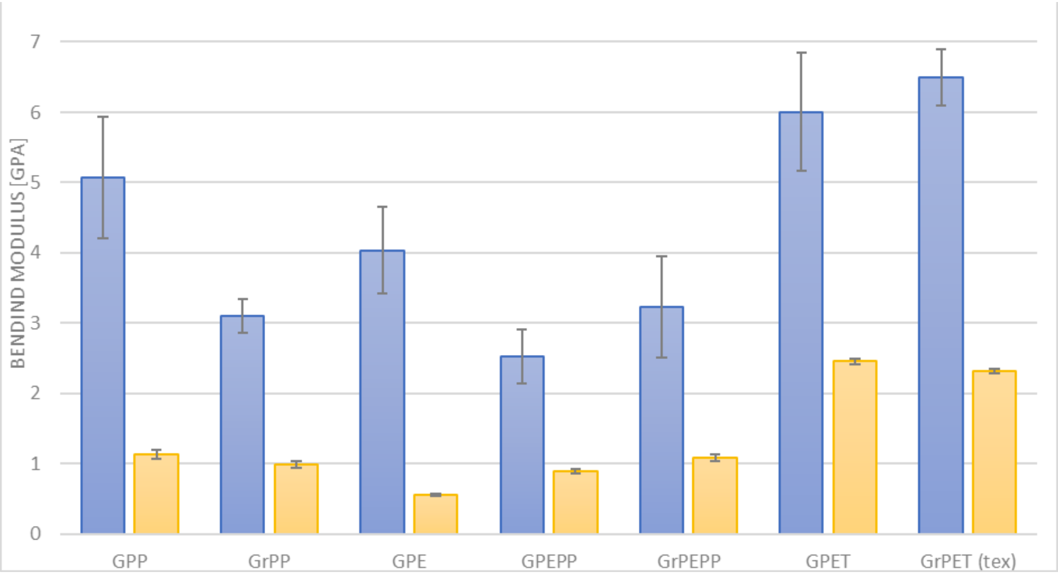

Figure 5: Bending modulus: Reinforced (blue) vs. unreinforced (yellow)

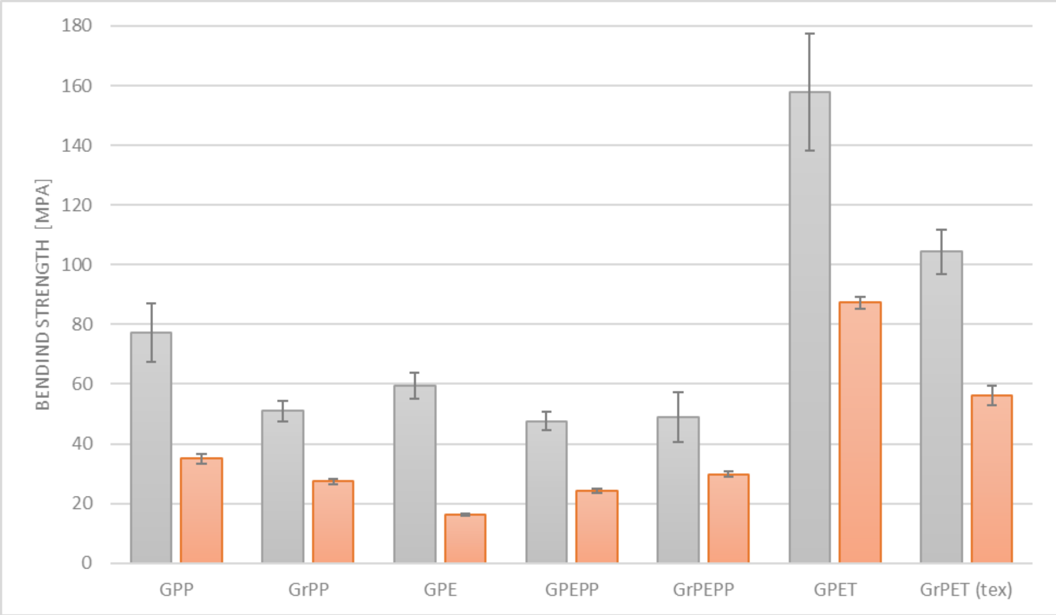

Figure 6: Bending strength: Reinforced (gray) vs. unreinforced (red)

3.5 Inter laminar shear strength results

The ILSS values are shown in Figure 7. According to the results:

- Recycled materials have lower ILSS compared to the virgin material: this is due to the decrease of the mechanical properties with the aging of material;
- Both GPEPPs have the lower ILSS value: this is due to the incompatibility between PP and PE. No coupling agents were used.

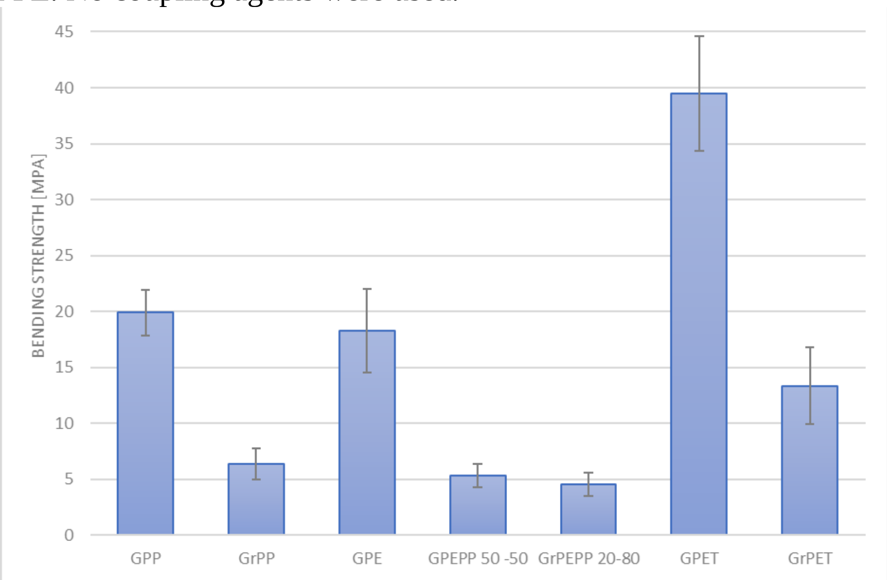

Figure 7: Bending strength.

Also, recycled PP shows a low ILSS, in comparison to the virgin value: previous research conducted on the same material revealed a higher ILSS value, close to the value of the virgin material. This gap exists because of different reasons such as the different way of processing and the use of additives, improving fibre-matrix adhesion and thus ILSS.

4. CONCLUSIONS

This paper shows the feasibility of producing thermoplastic composites with acceptable mechanical properties from glass fibres and recycled polymers. The following conclusions can be drawn from the results:

- The stiffness increases significantly by adding glass fibres to the polymeric matrices: both in laminated rTapes and overmoulded rTapes
- Recycled polymers have lower mechanical properties, due to the physical aging;
- The MFI of PET can be reduced by drying it for a certain time in the oven;
- Blending PE and PP does not give optimal results, caused by incompatibility between the two phases formed during melting. Coupling agent can be used as compatibilizers.

For further improvement, the use of stabilizers, in order to prevent material degradation and use of coupling agents to improve interfacial adhesion between matrix and fibre should be considered to obtain better mechanical properties.

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