INTERLAYER HYBRIDIZATION OF UNIDIRECTIONAL GLASS FIBRE COMPOSITES WITH SELF-REINFORCED POLYPROPYLENE

Y. Swolfs[1], X. Sun[1], L. Gorbatikh[1], I.Verpoest[1*], P. J. Hine[2], M. J. Bonner[2], I. M. Ward[2]

[1]*Department of Metallurgy and Materials Engineering, KU Leuven, Kasteelpark Arenberg 44, B-3001 LEUVEN (Belgium)*
[2]*School of Physics and Astronomy, University of Leeds, LS2 9JT, Leeds (UK)*
**Email address: ignaas.verpoest@mtm.kuleuven.be*

Keywords:
Self-reinforced composites, interlayer, hybrid, unidirectional glass fibre composites

Abstract
Unidirectional glass fibre layers have been hybridized with unidirectional self-reinforced polypropylene layers. Tensile tests and microCT have been performed to investigate and understand the properties of these hybrids. The tensile behavior is a combination of the stiff and strong glass fibre layers and the ductile self-reinforced layers. A hybrid effect of +10% was observed for the failure strain of the glass fibre layers. Tensile strength and absorbed energy showed mainly negative hybrid effects. This is attributed to the glass fibre misalignment, which is induced by the shrinkage forces of the self-reinforced layers. MicroCT measurements verified this hypothesis.

1 Introduction
Traditional fibre reinforced composites show early damage initiation and a low failure strain in tensile loading. Moreover, they easily develop delaminations in impact loading. The first reason for this brittleness is the brittle nature of the fibres. All commonly used reinforcing fibres have a low failure strain (carbon fibres < 2%, glass fibres < 3%), and even natural fibres like flax or hemp do not exceed these values. A second reason is the low transverse and shear strength of unidirectional impregnated fibre bundles (in textile composites) or layers (in laminates). A commonly applied solution is to use a more ductile matrix, but this has its limitations.

Another solution is to use ductile fibres. Self-reinforced composites (SRC) are an example of this approach. Self-reinforced polypropylene (SRPP) is the most popular SRC, mainly due to its low price, low density and very high toughness. The toughest SRPPs are made by hot compaction, a process developed by the University of Leeds [1]. Heavily drawn polypropylene (PP) tapes are used, with a stiffness of 10-15 GPa, a strength of 500-600 MPa and a failure strain of 10-15% (see [1-3]). The tapes are then woven into preforms. Finally, the preform is heated and the outer layers of the tapes melt and recrystallize to form the matrix phase. This eliminates the impregnation problems, which are often associated with thermoplastic matrix composites, because the matrix is created in situ. SRCs have a stiffness and strength of respectively 5 GPa and 180 MPa for a woven composite [1]. This is low compared to traditional glass and carbon fibre reinforced composites for the use in structural applications.

SRCs are tough but compliant, whereas traditional composites are brittle, but stiff and strong. To get a better toughness-stiffness balance, both composites can be combined into a hybrid composite. This has been shown to give rise to significant hybrid effects [4, 5], which are defined as deviations from the rule of mixtures. This means the properties can be better than the linear average of the properties of its constituents. A previous study by Taketa et al. [6] showed the potential of hybridization of carbon fibre composites with self-reinforced composites. The increase in the failure strain of the carbon fibre was proven to lie between +7 and +18%.

This paper will show results for self-reinforced composites hybridized with glass fibre. Because the glass fibre/polypropylene tape stiffness ratio is smaller than for carbon fibre/polypropylene tape, we believe the failure of the brittle glass fibre may introduce less damage into the self-reinforced composite. This may be beneficial for the toughness of the hybrid composite. This research focuses on unidirectional hybrid composites only.

2 Materials & testing methods

The hybrid composites described in this paper are made from unidirectional (UD) glass fibre polypropylene (GFPP) prepregs and unidirectional polypropylene tapes (SRPP). The GFPP prepregs were provided by Baycomp Performance Materials. The fibre content was measured according to ASTM D2584 and determined to be 34,1 vol%. The nominal thickness of the GFPP prepreg is 270µm. The PP tapes have a stiffness and strength in the range of 5-10 GPa and 400-600 MPa respectively.

The PP tapes are wound on a frame using an automatic winding machine. One winding cycle produces a unidirectional bottom and top layer of about 90µm each. The GFPP prepregs are, depending on the stacking sequence, inserted at appropriate locations to create a layer-by-layer UD hybrid. This hybrid is put in between an aluminum mould and pressed for 5 minutes at 180°C and 50 bar. When naming the layups, a letter code is used. The “S” stands for a self-reinforced layer, while the “G” stands for a GFPP layer. The produced layups are shown in table 1, including the theoretical and measured fibre volume fractions. The conversion from glass fibre weight fraction to GFPP volume fraction was done using a GF and PP density of respectively 2540 kg/m³ and 900 kg/m³.

Lay-up	Theoretical vol% of GFPP	Measured vol% of GFPP
$(S_5)_s$	0	0
$(S_6GS_3)_s$	25	28.6 ± 0.3
$(S_4GS_2)_s$	33	35.7 ± 0.6
$(S_3GS_{3/2})_s$	40	38.8 ± 0.9
$(S_2GS)_s$	50	55.1 ± 1.2
$(S_2GS_2G_{1/2})_s$	53	52.8 ± 5.3
$(S_2GSG_{1/2})_s$	60	59.6 ± 6.6
$(SGSG_{1/2})_s$	70	72.0 ± 2.6
$(G_2)_s$	100	98.6 ± 0.59

Table 1. Layups and their volume fractions of GFPP

2.1 MicroCT characterization

The samples were characterized using X-Ray computed tomography system Phoenix Nanotom S. The sample size was 2mm x 2mm x 10mm. The images with voxel size of 1 µm were acquired applying a 60 kV voltage and a 240 µA current. Molybdenum was used as a target material. The exposure time was set to 500 ms to obtain enough contrast.

2.2 Tensile tests

Tensile tests were performed according to ASTM standard D3039 on a Instron 4505. The load cell was 100kN and the strain rate applied was 5%/min. The strains were measured with an optical extensometer. An optical strain is calculated every 250 ms. At least 4 samples were tested for every layup.

A schematic tensile graph is shown in figure 1. When the GFPP layers break, the specimen surface is severely damaged. This makes it impossible to use any kind of extensometer to measure the sample strain. In the shown tensile data, we have used optical strain measurement system up and till the ultimate tensile strength (UTS). The UTS is defined as the highest stress, which occurs when the glass fibres break (see figure 1). After the UTS has been reached, the tensile stress drops vertically, but not to zero as in traditional glass fibre reinforced composites. The SRPP layers are able to withstand the GFPP damage and continue to carry stress after the UTS. We used the displacement of the crosshead to calculate the strain in this second part. This displacement was corrected for the machine compliance using the part before the UTS, where both optical strain and crosshead displacement are available. Four important parameters were calculated from the corrected stress-strain data. Due to initial settling of the specimens, the tensile modulus was calculated between 0,5 and 1% strain. The other three parameters are schematically shown in figure 1. The UTS is the peak stress. The strain at which this UTS occurs, is labeled “strain at UTS”. A third parameter is the absorbed energy, which is calculated as the area underneath the stress-strain curve. Because the stress gradually decreases down to zero, a cut-off criterion is needed. The area was calculated up to the strain where the stress drops below 10 MPa.

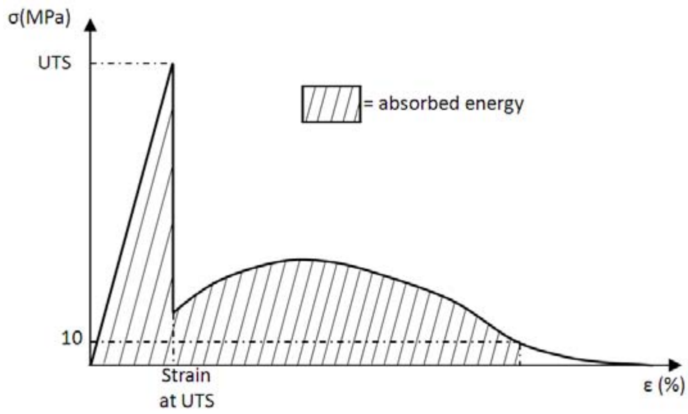

Figure 1. Schematic tensile graph indicating some of the calculated parameters

Before going into detail on the hybrid composites, we first analyse both its constituents. The layup $(G_2)_s$ contains 4 layers of the GFPP prepreg. This layup was determined to have a stiffness of 24 ± 4 GPa and a strength of 610 ± 67 MPa. The orientation of the glass fibres seems to be good, as seen in figure 2. Only a very limited amount of fibres are slightly misaligned.

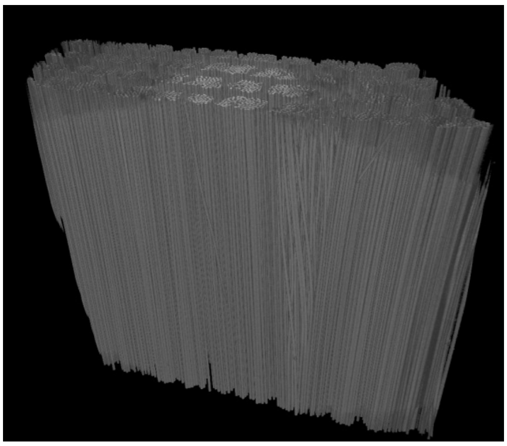

Figure 2. MicroCT image of glass fibre alignment in $(G_2)_s$

The layup $(S_5)_s$ contains 10 unidirectional self-reinforced layers, without any GFPP layers. These samples were produced at 180°C, which is actually too low to create enough matrix material. We chose this temperature anyhow, because in the hybrid composites the PP in the GFPP layers also contributes matrix material. By choosing this low temperature, the PP tapes have less opportunity for stress relaxation and better mechanical properties are retained. The stiffness and strength are 4.3 ± 0.03 GPa and 274 ± 28 MPa. The alignment of the SRPP layers was not verified. This is very difficult because PP matrix and tape have almost equal physical properties. Consequently, microCT does not show any contract between both phases.

2.3 Hybrid SRPP/GFPP

Tensile graphs of some hybrid SRPP/GFPP samples are shown in figure 3. When comparing the hybrid graphs with the SRPP and GFPP graphs, it is clear that a mixed behavior is obtained. When the GFPP layers fail, a vertical load drop is observed. Afterwards, the SRPP is the only load carrying element.

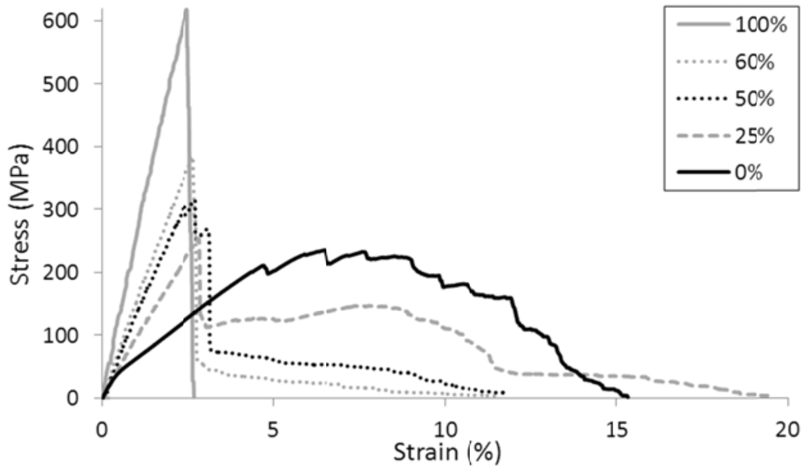

Figure 3. Tensile diagrams of hybrid SRPP/GFPP

These tensile graphs are analyzed in depth to explore the hybrid effect. All graphs are plotted as a function of the GFPP vol%, as previously shown in table 1. The results for the tensile modulus are shown in figure 4. The rule of mixtures is indicated by the dashed line. As expected, most of the data correspond rather well with the rule of mixtures.

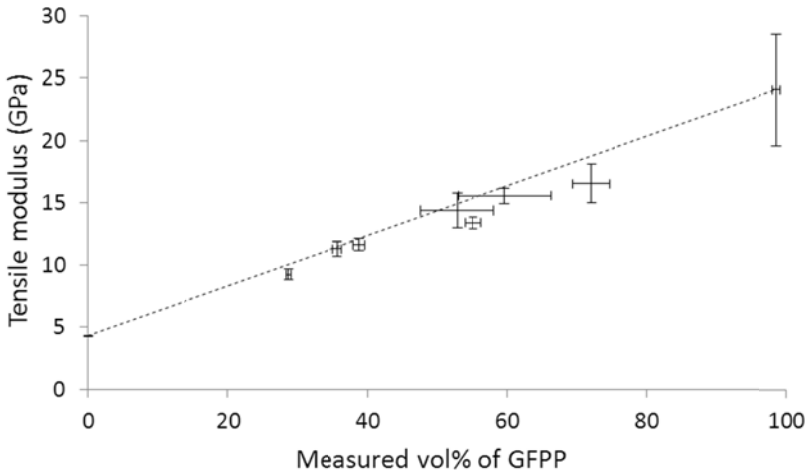

Figure 4. The modulus of hybrid SRPP/GFPP at various GFPP vol%.

The results for UTS are shown in figure 5. The rule-of-mixtures is in this case a bilinear line. The hybrids with less than 40% GFPP reasonably coincide with the rule of mixtures. The hybrids with a UTS of more than 40% GFPP clearly are below the rule of mixtures and thus show a negative hybrid effect.

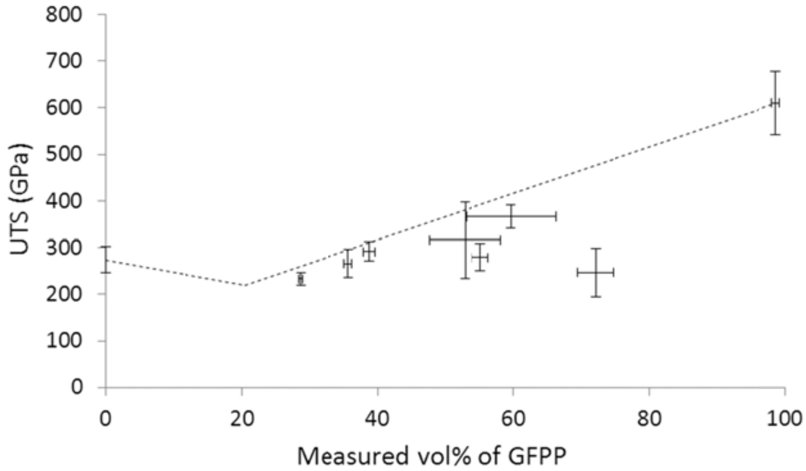

Figure 5. The ultimate tensile strength (UTS) of hybrid SRPP/GFPP at various GFPP vol%.

The results for the strain at the UTS are shown in figure 6. In this case, the rule of mixtures is a horizontal line. This is because this strain is related to the GFPP layers. Simple models predict that the failure strain of the GFPP does not change when a second material is added. A slightly positive hybrid effect can be seen for some samples. This hybrid effect is however smaller than +10% and not statistically significant at a 5% significance interval.

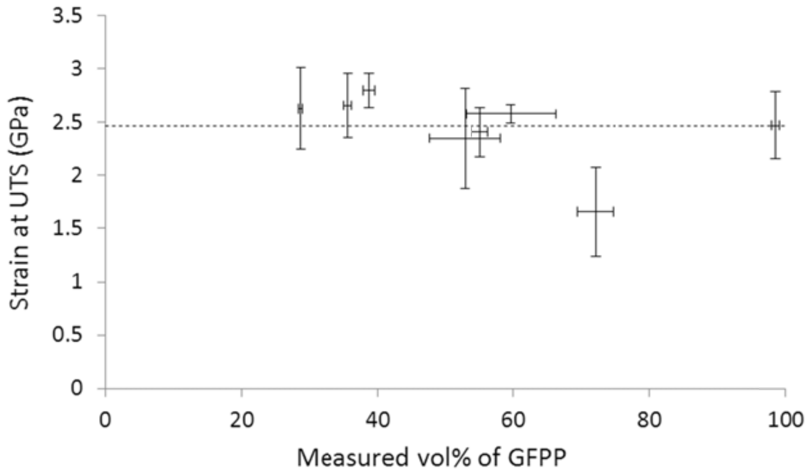

Figure 6. The strain at UTS of hybrid SRPP/GFPP at various GFPP vol%.

Finally, the absorbed energy is also calculated for each hybrid. The rule of mixtures is a straight line in this case, see figure 7. All hybrids have a significantly lower absorbed energy than expected from the rule of mixtures. A negative hybrid effect is present in all cases. This is due to the GFPP layers damaging the SRPP layers when they fracture at about 2,5% strain.

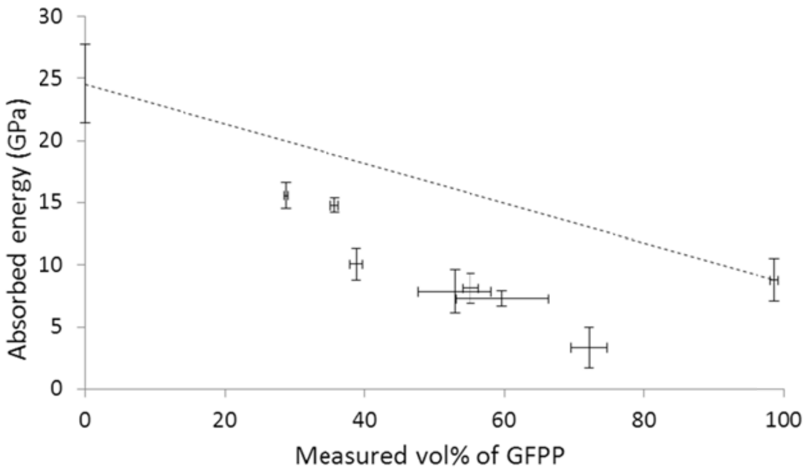

Figure 7. The absorbed energy of hybrid SRPP/GFPP at various GFPP vol%.

In conclusion, the hybrid effect for tensile modulus is slightly negative for some layups, while it is mostly negative for UTS and absorbed energy. This indicates some fibre misalignment may have occurred due to the shrinkage stresses of the PP tape. Because of the high degree of orientation of the PP tapes, they have a strong tendency to shrink during the hot compaction process [7]. This can distort the glass fibre alignment and explain the negative hybrid effect. This hypothesis was tested using microCT on a few hybrid layups. Figure 8 shows the results for $(S_2GSG_{1/2})_s$ and $(SGSG_{1/2})_s$, layups with theoretical fibre volume fractions of respectively 60% and 70%. This illustrates the glass fibre misalignment in $(S_2GSG_{1/2})_s$, while hardly any misaligned fibres are observed in $(SGSG_{1/2})_s$. This is surprising, because the most negative hybrid effects were observed for $(SGSG_{1/2})_s$. Because of the very small sample size, the microCT sample might not be representative for the entire plate. Samples from different locations within the same plate are needed to obtain more information.

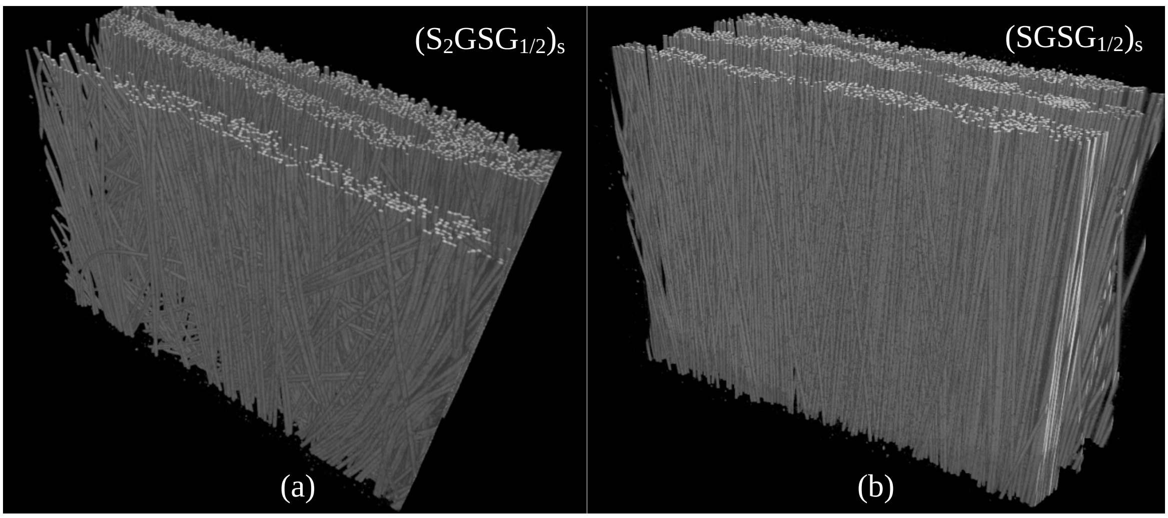

Figure 8. MicroCT image of hybrid layups (a) $(S_2GSG_{1/2})_s$ and (b) $(SGSG_{1/2})_s$

3 Conclusion

Interlayer hybrids of glass fibre and self-reinforced polypropylene have been made. A small hybrid effect of less than 10% was seen for the failure strain of the glass fibre. Ultimate tensile strength (UTS), strain at UTS and absorbed energy however, showed mainly negative hybrid effects. This is most likely due to the shrinkage of the PP tapes, which distorts the alignment of the glass fibres. The misalignment hypothesis was proven by microCT measurements.

4 Acknowledgements

The work leading to this publication has received funding from the European Union Seventh Framework Programme (FP7/2007-2013) under the topic NMP-2009-2.5-1, as part of the project HIVOCOMP (grant agreement n° 246389). The authors thank the Agency for Innovation by Science and Technology in Flanders (IWT) for the grant of Y. Swolfs. The help of technicians K. Van de Staey, B. Pelgrim and M. Adams is gratefully acknowledged.

5 References

[1] Ward I.M., Hine, P.J. The science and technology of hot compaction. Polymer, 45, pp. 1413-1427 (2004).
[2] Hine P.J., Ward, I.M., Teckoe, J. The hot compaction of woven polypropylene tapes. Journal of Materials Science, 33, pp. 2725-2733 (1998).
[3] Teckoe J., Olley, R.H., Bassett, D.C., Hine, P.J., Ward, I.M. The morphology of woven polypropylene tapes compacted at temperatures above and below optimum. Journal of Materials Science, 34, pp. 2065-2073 (1999).
[4] Hayashi T. On the improvement of mechanical properties of composites by hybrid composition. Proc. 8th Intl. Reinforced Plastics Conference, pp. 149-152 (1972).
[5] Aveston J., Sillwood, J.M. Synergistic fiber strengthening in hybrid composites. Journal of Materials Science, 11, pp. 1877-1883 (1976).
[6] Taketa I., Ustarroz, J., Gorbatikh, L., Lomov, S.V., Verpoest, I. Interply hybrid composites with carbon fiber reinforced polypropylene and self-reinforced polypropylene. Composites Part A: Applied Science and Manufacturing, 41, pp. 927-932 (2010).
[7] Le Bozec Y., Kaang, S., Hine, P., Ward, I. The thermal expansion behaviour of hot-compacted polypropylene and polyethylene composites. Composites Science and Technology, 60, pp. 333-344 (2000).