'Nature Wins': development of 100 % bio-based thermoplastic composite materials

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With global oil reserves declining and high CO_2-levels causing environmental damage, the future calls for the development of an intrinsically sustainable economy. Among multiple solutions, materials from renewable resources will play a significant role in solving these problems. In the field of composite materials, this can be achieved by developing alternative fibres and matrices. The resulting composite material needs to:

1. be recyclable,
2. contain renewable raw materials,
3. have a high performance/weight ratio.

To address these issues, Centexbel, SLC-Lab and ITA (RWTH Aachen) collaborated on the research project 'NATURE WINS' (Cornet). In this research, novel composite materials based on hybrid textiles from thermoplastic biopolymer (PLA) and natural fibres (flax) were developed. A wide range of possible applications can be found for this type of composites in various fields, e.g. semi- and non-structural parts in transportation, parts in sports & leisure equipment, as well as panels in construction and design.

Introduction – the 'Nature Wins' processes

Natural fibre composites still have some hurdles to overcome in order to be widely applicable. Among others, natural fibre textiles are often not easy to impregnate, due to poor wetting of the fibres by polymeric matrices. This usually also leads to poor adhesion between fibre and matrix. In addition, the fibres need to have a well-defined orientation in the composite to obtain the maximum mechanical properties in the desired direction(s), which is also more difficult to achieve with natural fibres due to their discontinuity.

To overcome these obstacles, 'Nature Wins' follows specific processing routes (see diagram in fig. 1) that involve intimately blending flax fibre and PLA staple fibre to create a hybrid fabric (yarn, fabric, mat). These textile materials form the basis for the developed composites, which are manufactured by melting

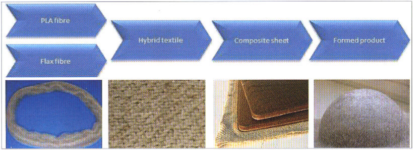

Fig. 1:
Diagram of the process routes followed in 'Nature Wins'. Both woven and nonwoven textiles were developed.

the PLA at high temperature and under pressure so that it flows between the flax fibres (impregnation or consolidation). Because the PLA is pre-mixed well with the flax, the flow distance is smaller during pressing, and an improved impregnation is expected.

Besides the development of processes for hybrid fabrics and the consolidation process, techniques for press forming thermoplastic natural fibre composites were also developed. Press forming into a shape can be done directly from the hybrid fabric, but usually a pre-consolidated composite sheet is used.

Following aspects were addressed in the research:

4. compatibility of flax and PLA,
5. fibre spinning from PLA,
6. hybrid yarns, nonwovens and woven fabrics from PLA and flax,
7. consolidation of composites,
8. press forming of nonwovens and woven fabrics,
9. environmental assessment of developed materials.

Results

1. Compatibility of flax and PLA

In the initial phase of 'Nature Wins' it was examined whether treatment of flax fibres is necessary to improve adhesion with the PLA matrix. As a measure for the adhesion, the interfacial shear strength (IFSS) was measured by means of the micro-drop technique. In this technique, a drop of PLA is deposited on a single fibre. Using the set-up described in fig. 2, the force needed to pull off the solidified drop is measured. The higher the force required to detach the droplet from the fibre, the higher is the IFSS. These tests showed that flax and PLA exhibit very good adhesion, which made further fibre treatment unnecessary. This is in contrast to the system flax-polypropylene, which has very poor interfacial adhesion.

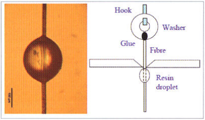

Fig. 2: left: PLA micro-droplet on a flax fibre
right: diagram of the pull-out test

2. PLA fibres

Several PLA grades were selected based on viscosity, crystallinity and melting temperature. A low viscosity matrix material is especially interesting, since this facilitates the impregnation of the fibres. Unfortunately, the mechanical properties of low viscosity (and thus low molecular weight) PLA are generally lower. Therefore, blends were made of very low viscosity PLA (MVI 50) and a high-viscosity standard (fibre) grade PLA (MVI 5).

Centexbel demonstrated that both the standard and the PLA blends can be smoothly processed in multifilament extrusion. However, this is only the case when the resin has been pre-dried to a water content of less than 300 ppm. Otherwise hydrolytic degradation during extrusion deteriorates the properties of the material.

The achieved tenacity of the yarns only has to be sufficient for textile processing, because the PLA will melt when the composite is produced. As can be seen in fig. 3, the tenacities achieved with different processing conditions and materials are between 0,9 and 0,23 N/tex. As demonstrated by the difference in tenacity between MVI 5 spun at 190° C and MVI 5 spun at 220° C, higher melt stress (caused by increased viscosity at 190° C) improves tenacity. Melt stress induces an oriented crystal structure and increases crystallinity, with the result that the fibre has good mechanical properties.

The PLA blends can only be processed with specifically tuned extrusion temperature. Other parameters (cold drawing temperatures and velocities) can be kept the same as for the standard grade. Fig. 3 shows that the PLA blend (with 30 % MVI 50) only exhibits a minor loss in strength. On the other hand, the blends have a much lower viscosity and are therefore better suited to flow between flax fibres.

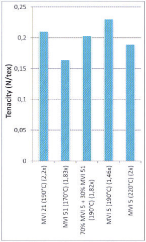

Fig. 3: Tenacity of PLA multifilaments from different PLA grades. Extrusion temperature and cold draw ratio are mentioned in brackets.

3. Hybrid textile

ITA used flax fibres (38 mm length) and PLA staple fibres (from filaments made at Centexbel) to develop intimately blended yarns and nonwovens. Woven fabrics were also made, based on the blended yarns. For all these materials, 40 vol% of flax was used (corresponding to 50 wt%).

Flax/PLA yarn

Flax sliver and PLA staple fibre were drawn together to provide a mixed sliver (see fig. 1), from which yarns were spun by means of rotor spinning. ITA used a Box-Behnken design of experiments to identify the influence and optimum values of the process parameters: (i) draft (delivery of the sliver), (ii) twist per meter and (iii) rotor speed. The best results achieved were at 1440 tpm twist, with a tenacity of 2.8 cN/tex. The applicability of these yarns for composites was assessed by SLC-Lab.

Nonwovens

For the nonwovens, ITA used flax fibre (60 mm) and PLA staple fibre (60 mm). Both fibres are mixed during the 'opening' of the fibre bales (blending). After one or more mixing steps, the fibres are air-laid down into a web. Multiple layers of which are connected by needle punching to the desired thickness (and density). Multiple mixing steps are expected to increase the homogeneity of the nonwoven. Needle punching increases the nonwoven's strength and density, also making it thinner, which makes it easier to handle in the composites processes.

Fig. 4: flax/PLA air laid and needle punched nonwoven

4. Composite processing

For textile-based thermoplastic composites consolidation is often done in a thermal press. Ideally, when sufficient production volume can be achieved in industry, this would be a continuous press or so-called *double belt press*. However, in this research static presses were used. The process is as follows:

1. fabric (yarn, fabric or nonwoven) is heated under (light) pressure to the processing temperature. The polymer should be completely melted at this stage;
2. pressure is increased and the material is held at constant temperature while the polymer impregnates the fibrous material;
3. once consolidation is achieved, the material is cooled under high pressure.

SLC-Lab determined the best processing temperature, pressure and pressing time for flax/PLA by evaluating porosity through microscopy. It is already known from literature that when processing flax, the temperature should be kept under 190-200° C to avoid degradation. The optimum processing parameters were established separately for UD composites and nonwoven composites.

Unidirectional composites

Unidirectional (UD) composites have a specific structure where all the fibres (or yarns) are oriented in exactly the same direction (the "0°-direction"). Fibre orientation has a very strong influence on mechanical properties, thus this type of composite allows the easy interpretation of the mechanical properties. Furthermore, it can be used as representative element when predicting properties of textile composites of multiple layers. SLC-Lab made UD composites by parallel winding of flax/PLA yarns from ITA. The optimum process parameters were established as: pressure 15 bar, temperature 190° C, dwell time 10 min. SLC-Lab used these for further characterisation of the UD composites.

Even though microscopy showed a minimal amount of voids, which indicates good impregnation, the mechanical properties of the resulting unidirectional composites were low. The measured tensile modulus of 11.0 GPa is less than 50 % of the theoretically predicted value for a UD flax/PLA composite (26 GPa). The explanation for this is that the flax fibres are strongly disoriented by the rotor spinning process. The twisting causes them to form a helix around the yarn axis (see fig. 5). Microscopy showed that the mean angle of the fibres is 45° to the 0° axis of the UD composite (instead of the desired minimal deviation from the 0° orientation). The theoretical tensile modulus for a UD flax/PLA composite with the fibres oriented at 45° was determined as 11.7 GPa, which corresponds well to the experimental value measured in this study.

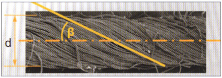

Fig. 5: flax yarn showing the mean angle of the fibres to the yarn axis β. This angle should be minimised in composite preforms[1].

Due to these results, new yarns are being developed with lower twist and higher fibre orientation. Composites with woven fabrics will be produced with these new yarns and will be evaluated.

Nonwoven composites

Nonwoven composites are made by simply stacking one or multiple layers of hybrid nonwoven in a mould and consolidating these following the procedure explained above. The development of nonwoven composites was performed in two phases.

Consolidation quality was verified by two parameters: mechanical properties of the nonwoven and void content through microscopy. To optimise consolidation, it was found that two parameters are important:

- blending steps: first consolidation tests showed one blending step was not sufficient. The resulting plates were not homogeneously consolidated; with dry spots. To limit these dry spots, consolidation temperature was kept at 185° C for all tests. The best result was found when three blending steps were used;
- flax fibre length: fibre length was varied between 38, 50 and 80 mm. The composite with 50 mm long fibres give the best composite properties.

This being the said, the current results do not show sufficiently high mechanical properties, i.e. the best plates exhibit a tensile modulus of 6.5 GPa. This is much lower than 11 GPa, the expected value for a nonwoven flax/PLA composite. Microscopy has shown that voids were the cause of this, indicating that the processing parameters and the blending quality were not yet optimal. Therefore, SLC-Lab will perform final tests at somewhat higher temperature, pressure and with longer dwell time.

5. Press forming

The process for manufacturing a shaped component from thermoplastic composite is depicted in Fig. 6. The press forming process is characterised by a high potential for automation and low cycle times, which makes it especially suitable for mass production.

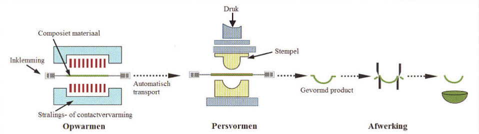

Fig. 6: subsequent process steps for press forming thermoplastic composite (f.l.t.r): heating (infrared), transport to the mould, press forming the component, edge trimming.

SLC-Lab developed matched metal moulds and processing parameters for press forming nonwovens on the one hand, and multiple layers of woven fabric on the other. In principle, the textile can be pressed straight into the desired shape, but much better results are achieved when a pre-consolidated sheet is used. When using a matched metal mould, it is very important to use the correct sheet thickness to match the cavity in the die.

The mould is always accompanied by a clamping device that applies a force on the sheet. This is used to control the movement of the sheet (or textile) into the cavity as the mould closes.

Fig. 7 shows an example where the clamping force was too high, causing the nonwoven to stretch and thin out in the centre. Especially when forming a woven textile composite, the clamping force is very important. Insufficient feeding of the sheet into the mould will cause it to tear, whereas over-feeding will cause wrinkles.

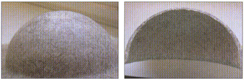
Fig. 7: formed and consolidated nonwoven. The figure on the right shows the importance of controlling the clamping force to achieve the desired thickness, with a thinner cross section in the centre of the dome.

Press formed demonstrator: car seat

SLC-Lab collaborated with the Groep T University College of Leuven to develop a seat for a "Formula Student" racing car out of flax/PLA composite. The material properties of the fabric and composite were used to design both the seat and its mould. Fig. 8 shows the mould as it was built and the form that the seat should take. SLC-Lab are currently still running forming trials to make the demonstrator.

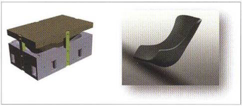
Fig. 8: mould and design impression of the "Formula Student" racing car seat

6. Environmental assessment

Centexbel performed a cradle-to-gate analysis comparing flax/PLA woven composites to standard glass/PP woven composites. The results show that by using bio-based resources, the impact on global warming can be greatly decreased. In the case of flax, the CO_2-emissions are even negative, due to its use of CO_2 to grow. However, the eutrophication and acidification that accompany the use of chemicals during agriculture still form an impact that need to be reduced.

Fig. 9 shows the use of non-renewable energy for the raw materials and production processes for a woven flax/PLA composite, compared to a woven glass/PP composite, and both with a fibre volume fraction of 40 vol%. It seems there is no real gain to be achieved by using renewable resources, but this is mainly due to the energy-intensive spinning operation needed for the current bio-based composites (see 'yarn production' in Fig. 9) [2,3,4]. The same comparison was made for composites based on nonwovens, where the textile processes are completely parallel for flax/PLA on the one hand and glass/PP on the other. In that case the bio-based alternative shows a significant reduction in embodied non-renewable energy.

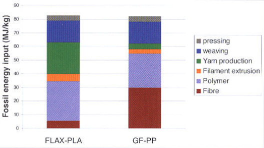

Fig. 9: comparison of embodied energy in flax/PLA and glass/PP woven composites

Conclusions

This research has shown that it is possible to produce 100% bio-based thermoplastic composites with minimal void content from intimately mixed textile preforms. Flax/PLA is a very interesting fibre-matrix combination, due to the inherent compatibility of these materials. Composite sheets from hybrid natural fibre textiles perform well in the press forming process, thus making it feasible to further develop efficient mass production processes for this type of material. However, the mechanical properties achieved are below expectations and too low for commercial application. In the yarn based composites, this is mainly caused by fibre twist introduced by the rotor spinning process. This shows the importance of keeping the twist as low as possible when spinning hybrid yarns for composites. The spinning operation also has a large energy use, which partially neutralises the positive effect of using bio-based raw materials, in comparison to standard composites. In the nonwoven composites, the reduction in properties was caused by voids. This indicates that the properties can be increased by improving the composite consolidation process. If so, these materials may form a very good alternative to nonwoven glass composites.

Based on the conclusions of this study, future research in this field should further address the issue of developing natural fibre preforms that are both cost- and performance-effective.

Acknowledgements

Centexbel, ITA Aachen, FKT and SLC-Lab would like to thank the CORNET program, IWT (grant No. 100385) and AIF for making this research possible.

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