Biocomposites based on plant material

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Abstract

Fibre reinforced composite materials offer superior specific mechanical properties in reference to their weight. In the past years, composite materials such as carbon or glass reinforced plastics (CFRP or GFRP) are used increasingly in all sectors of transportation and for industrial or leisure products. The composite consists of a load bearing fibre architecture, usually in the form of a continuous fabric architecture, and an embedding matrix, usually a thermoset such as epoxy. With regard to the energy efficiency and carbon footprint, due to their lightweight nature, these composite materials in general offer interesting properties, if applied in long-term operations. However, the raw materials used for the production of both typical fibre materials and thermoset resins are still based on crude oil, and the refining and processing up to the semi-finished good consume a significant amount of embodied energy. In this study, composites made of glass or flax fibres and resin systems based on condensed tannin and furfuryl alcohol, both extracted or derived from plant tissues, were manufactured using vacuum infusion (VI) and resin transfer moulding (RTM) processes. The results show that mechanical properties close to common fiber/resin combinations like glass fiber and epoxy or phenolic resins can be reached by these materials. Copyright © 2018 VBRI Press.

Keywords: Fiber reinforced composites, tannin, biocomposites, resin transfer molding.

Introduction

Fiber reinforced plastics (FRPs) or composites are increasingly used in all sectors demanding low weight and high stiffness and strength. They consist of a load bearing fiber and an embedding matrix. Due to their excellent lightweight design properties (specific stiffness and specific strength, relative to their density), they are ideally used in fast moving or accelerated systems [**1**], thus helping to save energy and reduce CO_2 footprint. However, as most of the raw materials used today (fibers and matrices) are oil-based products and required high amounts of energy for their production [**2**], the embodied energy of the finished component might be high. Thus, an effective energy saving is only achieved after long-term operation [**3**]. Renewable fibers or matrices are used for some applications, however a combination of natural fibers and a resin system based on renewable sources does not exist today. Most bio-based resins are only blended with renewable material but based on common systems like epoxy [**4**]. A sustainable, green composite system has to provide interesting material properties, comparable to systems available for the targeted applications. Furthermore, it has to be processible in terms of cycle time, part quality (pores and dry spot formation) and fiber-matrix bonding. The resin system used in this study, based on a tannin-furfuryl alcohol (TFA) formulation, is targeted for applications with the highest requirements concerning Fire Smoke Toxicity (FST). Due to the chemistry of the system, it is comparable to today's phenolics, which are widely used in aeronautics and railway applications [**5**].

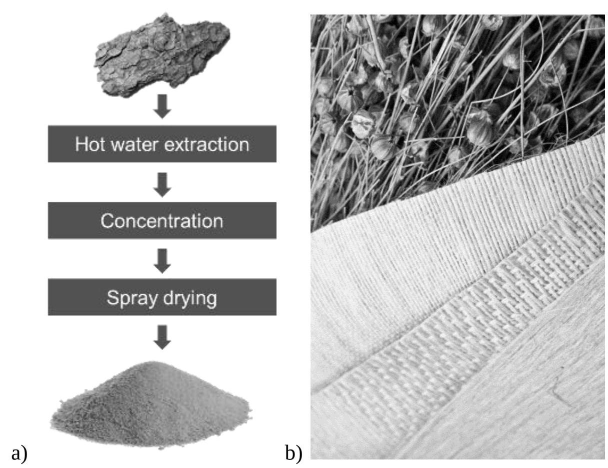

Fig. 1. Biocomposite based on plant material: a) Tannin sourcing from tree bark; b) flax fibers [BComp].

Tannins are natural polyphenolic compounds that can be recovered from wood tissues by hot-water extractions [**6**,**7**]. Furfuryl alcohol is produced by controlled reduction of agroforestry wastes [**8**]. No solvents are needed for processing. In combination with a natural fiber, in this case a flax fabric, a true biocomposite results (**Fig. 1**). The amount of material coming from non-renewable sources is minimized and limited to the hardener (p-Toluenesulfonic acid, pTSA) and auxiliary materials.

Experimental

The focus of this study is to prove the feasibility of the flax-TFA system in terms of processing in

a) hand lamination,
b) vacuum infusion,
c) resin transfer molding (RTM).

Rectangular plates with dimensions of approx. 250×250×4mm were produced. The parts manufactured with these processes were then characterized using a universal testing machine Shimadzu AG-X 250kN. In this first stage, the Young's modulus, tensile strength and interlaminar shear strength (ILSS) were addressed and compared to other resin systems, mainly epoxy and phenolic. As an alternative to flax fibers, glass fibers were processed as well. The flax fiber reinforcement is an ampliTex 5031 (BComp, Fribourg, Switzerland) in quasi-isotropic layer sequence, the TFA system was used in two different mixtures, allowing to influence important process parameters such as resin reactivity, viscosity and thus cycle time. This is controlled by the addition of pTSA as a catalyzer. TFA systems for FRPs are not currently commercially available and were purposely formulated for this study.

For the production in the hand lamination process, the resin system has to provide relatively long processing times. Hand lamination in this context was used for a first trial with the TFA system. This process is however not widely used today because of the time and cost intensive characteristics. The flax-TFA system was processible. However, part quality in terms of pinholes and porosity inside the component was poor and vacuum compression as a second process step is recommended. As a fraction of the water content of the system was removed during curing, micropores are likely to generate.

Vacuum infusion is significantly faster than hand lamination, as the fiber reinforcement is first stacked to its final layer sequence, sealed with a vacuum bag and the resin is then pressed through the reinforcement by applying vacuum gate to vent side. Thanks to the low viscosity of the TFA resin, vacuum infusion was feasible as with other low-viscosity systems. The infusion time for a plate of 250 × 250 mm took from 16 to 20 minutes, afterward the component had to be cured for 12 hours. Based on the exothermal curing reaction, which leads to an increasing viscosity over time, the process was only feasible using the resin system with the slowest reaction time, as otherwise the flow front came to a stop and a not fully impregnated part resulted.

In RTM processing, illustrated in **Fig. 2.**, the same fiber stackup as before was used, but placed inside a cavity between the lower and the upper die (**Fig. 3**). A closed mold process resulted, allowing to apply pressure during injection and after the complete impregnation of the part (post pressure). The plates manufactured using the RTM process showed significantly better properties compared to hand lamination or vacuum infusion. Processing time was reduced to approx. 20 minutes (from start of injection to demolding of the finished part). The tool was heated to 60°C and the resin was injected at room temperature with a pressure difference of 4.9 bar (0.1 bar vacuum pressure, 4 bar injection pressure). The elevated mold temperature in combination with the exothermal curing reaction helped to get rid of the water contained in the system (visible as steam bubbles in the hose on the vent side of the tool). By applying post pressure, pores could be minimized, which provided significantly better part quality compared to the other processes. A post cure cycle for 3 hours at 80°C is recommended. Based on these findings, all further experimental work has been carried out using RTM as manufacturing process.

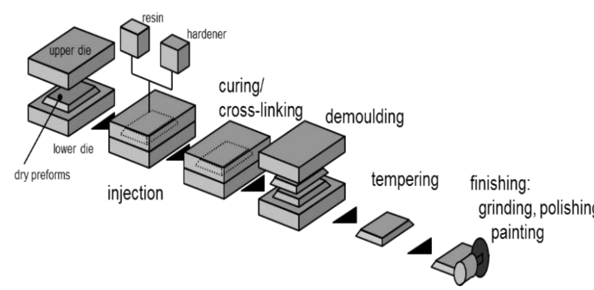

Fig. 2. Resin Transfer Molding process (injection, curing, demolding)

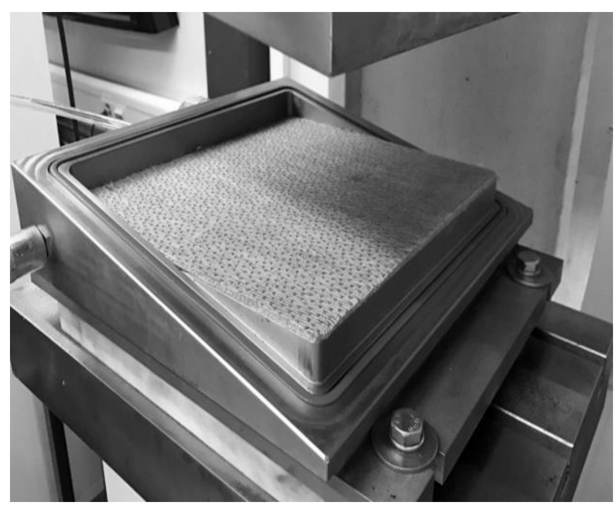

Fig. 3. RTM processing using plate tool; dry flax fiber preform placed on lower die (mounted on press)

Several plate specimens have been produced by the RTM process. The manufacture has proven to be robust, with no scrap parts and a constant quality concerning surface and visible pores.

Using a Mutronic Diadrive 2000 CNC mill, specimens for mechanical testing and micrographs had been cut out. The specimens were arranged and numbered in a way to identify close-to-gate and far-from-gate positions, as part quality usually diminishes towards the vent size of RTM components [**9, 10**].

Results and discussion

Concerning the processing of the proposed material, RTM production delivered the best results. However, this is the case with nearly all resin systems, as two good surfaces (because of the closed tooling) exist and post pressure enhances part quality significantly.

The preliminary analysis for FST properties have been done using a cone calorimeter test. This allows the determination of the total heat release and total smoke release. Comparisons were between composites having TFA or synthetic phenolic resins for matrix and reinforced with glass fibers, as a flax/phenolic combination was not available. The results are shown in **Fig. 4**. It could be shown that the TFA-glass composite has comparable properties in terms of heat release, and better properties concerning smoke release.

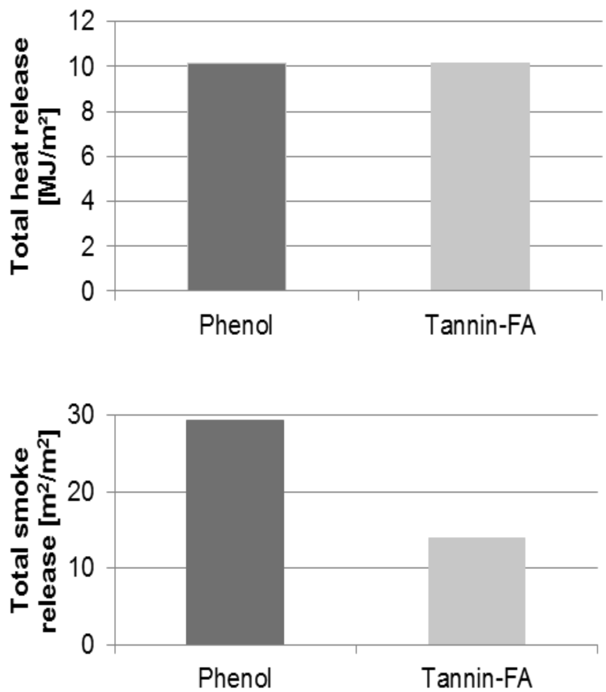

Fig. 4. Results from FST testing (cone calorimetry): total heat release and total smoke release.

Mechanical properties (tensile modulus, tensile strength, max. strain, ILSS) have been examined for the following resin/fiber combinations:

- TFA/Glass
- TFA/Flax
- Phenol/Glass
- Epoxy/Glass
- Epoxy/Flax

The combination Phenol/Flax was not processed. As phenol/glass is widely used in several applications today, it is a good benchmark for the new system. The results for the glass reinforced material are given in **Fig. 5**.

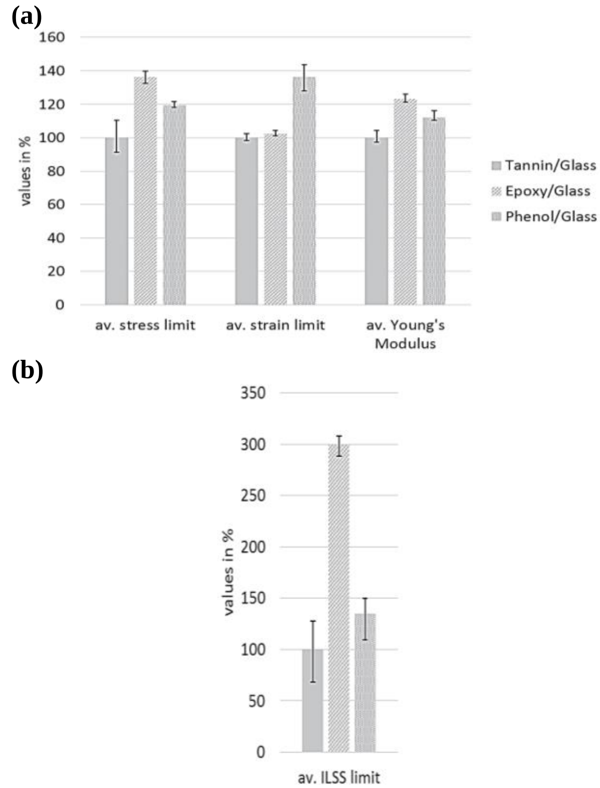

Fig. 5. Results from mechanical testing. a) Comparison of tensile strength, max. strain and Young's modulus for Tannin/Glass, Epoxy/Glass and Phenol/Glass composites (Tannin/Glass set to 100%); b) Comparison of interlaminar shear strength ILSS for Tannin/Glass, Epoxy/Glass and Phenol/Glass (Tannin/Glass set to 100%).

The results for the flax fiber reinforced composites are illustrated in **Fig. 6**.

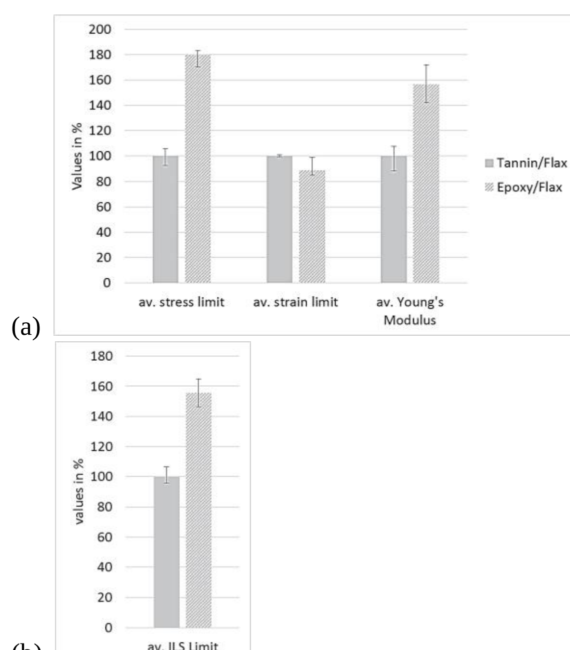

Fig. 6. Results from mechanical testing. a) Comparison of tensile strength, max. strain and Young's modulus for Tannin/Flax and Epoxy/Flax composites (Tannin/Flax set to 100%); b) Comparison of interlaminar shear strength ILSS for Tannin/Flax and Epoxy/Flax (Tannin/Flax set to 100%)

The results of the mechanical characterization illustrated the potential of the new, yet unoptimized TFA resin system. Combined with glass fibers, the mechanical properties from tensile testing were within a 40% range when compared with either epoxy or phenol resin materials. The combination of flax fibers and epoxy resin shows significantly better mechanical properties than the TFA/flax combination, because the base epoxy resin has better mechanical properties than the TFA system, and the fiber reinforcement was quasi-isotropic. The ILSS value, dominated by the properties of the matrices, was on the same level for the phenolic and the TFA system, and significantly better for the epoxy system. This is a result of the sizing of the fibers (glass and flax), which is optimized for the commonly used epoxy resins and which has to be modified when changing to the TFA (or phenol) resin system.

Conclusion

The objective of this study was to determine the feasibility of TFA natural fiber composites in terms of processing and determine the most important FST and mechanical properties. Using the RTM process and not yet optimized fiber and resin materials, it was possible to produce plate specimens which show similar mechanical properties to synthetic phenolic systems. FST properties assessed in a preliminary cone calorimeter test were promising and on the same level as synthetic phenolic systems.

Fiber matrix adhesion between the natural fiber and the TFA matrix will be a key element in further improving the mechanical properties of RTM laminates. It is assumed that the pre-treatment of the fiber available today is not completely compatible with the resin system. Additionally, due to the water generated during the injection/curing cycle, measures to further enhance porosity and prevent the creation of micro voids will be necessary. This might be addressed by either optimizing the formulation of the resin system or modified process control, or a combination of both.

Author’s contributions

Conceived the plan: GAB, DS, SB; Performed the expeirments: DS, OS; Data analysis: GAB, DS, OS, SB, IM; Wrote the paper: GAB, SB. Authors have no competing financial interests.

Supporting information

Supporting informations are available from VBRI Press.

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