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Mechanical properties of polylactic acid (PLA) composites reinforced with unidirectional flax and flax-paper layers

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ABSTRACT

Natural fibers are gaining much interest in composite materials because of their potential to replace conventional glass fibers. In particular, unidirectional flax fiber composites have shown excellent mechanical properties with different thermoset matrices. However, few data are available when they are made with a biodegradable thermoplastic matrix. This paper compares the mechanical properties of two types of unidirectional flax composites using polylactic acid as the matrix: one made with layers of aligned flax rovings alone and the other containing an additional paper layer fabricated using paper making techniques. The results show that specific tensile properties of the flax/PLA (252 $MPa \cdot cm^3 \cdot g^{-1}$) and flax-paper/PLA (217 $MPa \cdot cm^3 \cdot g^{-1}$) composites are similar to those made using woven glass fabrics impregnated with epoxy (227–278 $MPa \cdot cm^3 \cdot g^{-1}$). Very high impact strength (600 J/m) was also obtained for UD flax-paper composites compared to unreinforced resin (15 J/m). Together these mechanical properties are promising for industrial applications of this new reinforcement.

1. Introduction

NFCs will play a key role in the future of composite materials. Driven by their eco-friendly aspect, their good specific mechanical properties (comparable to those of glass fiber composites) and their cost effectiveness [1], flax fiber composites in particular have shown potential for the automotive and construction sectors [2].

Current flax reinforcements come in different shapes, ranging from isotropic mats [3], knitted quasi-isotropic or orthotropic reinforcement [4–6], up to isotropic air-laid or unidirectional non-woven fabrics [7,8]. Natural fiber mats are usually cheap. They can be laid down using fast spraying techniques. They also present high permeability to liquid resin, enabling parts fabrication by various liquid composite molding (LCM) techniques, such as the resin infusion (RI) and resin transfer molding (RTM) processes. On the other hand, mat reinforcements are not optimal for load bearing applications where weight is a key design factor. Knitted reinforcement (fabrics) can generally offer better mechanical properties depending on the knitted structure and the yarn design [4]. However, the production of natural fiber fabrics requires more time. The yarns must also be twisted a minimum number of turns per meter in order to induce the strength required for production on textile machines. Not only this torsion reduces the yarn permeability, but also it affects negatively the composite tensile properties due to fiber misalignment [9].

Unidirectional (UD) reinforcements are the most suitable structures to maximize the stiffness and strength of laminated composite parts designed for load bearing applications. Unidirectional prepregs are used in thermostamping and compression molding processes and dry fabrics are used in liquid composite molding processes such as RTM and vacuum assisted RTM (VARTM). While enabling light weight design by optimising the composite's layers orientations, UD fabrics can be somewhat difficult to manipulate. The stitching locally reduces the fibers alignment and the risk of inducing misalignment of the parallel individual fibers during manipulation remains important. These characteristics may negatively affect the expected mechanical properties. Stitching the UD reinforcement also creates damages to the fibers when the needles are pushed through the reinforcement. This fabrication constraint can also affect mechanical properties.

To avoid these drawbacks, the development of a new flax-paper reinforcement was initiated with the objective to maintain a good cohesion between the aligned flax fibers (for molding) and avoid disturbing and damaging the fibers during reinforcement fabrication [8]. This is achieved by using a thin porous paper layer backed to the aligned flax fibers, which acts as the fiber binder (Fig. 1). Flax and paper layers are assembled using processes inspired from papermaking [10]. This ensures a good adhesion between both types of fibers, reduces the impact of paper on the mechanical

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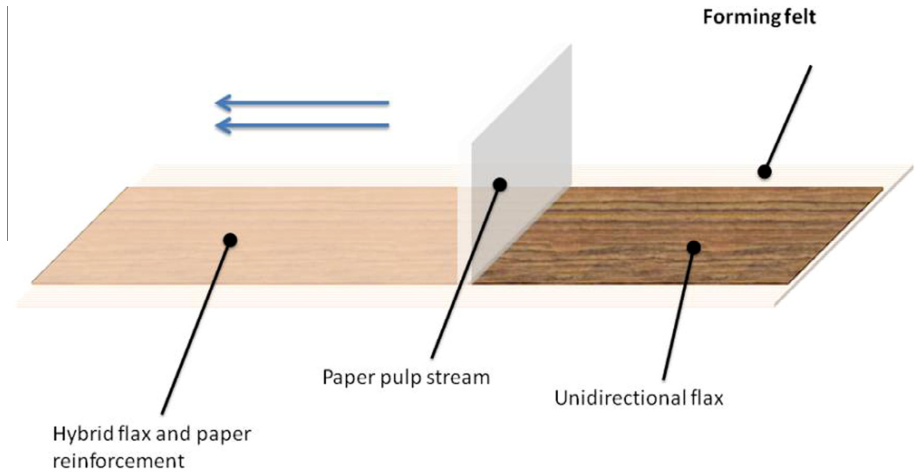

Fig. 1. Flax and paper reinforcement fabrication concept.

properties because it is thin, and paves the way for high volume fabrication of the reinforcement using established papermaking processes. Moreover, paper binders allow the use of low twist natural fiber yarns for the fabrication of the UD reinforcements while avoiding the negative effect of needles on the integrity of the reinforcement. This means that better mechanical properties can be expected, because less damage is induced to the fibers, with an enhanced impregnation capacity of the final reinforcement considering the use of low twist yarns.

As far as the matrix is concerned, most works realized with high viscosity PLA have used the injection or compression molding processes. Short fiber compounds are typically used and relatively low mechanical properties are obtained. For example Oksman et al. [11] and Bax and Müssig [12] both used flax and PLA in compression and injection molding processes, respectively, and obtained tensile and stiffness properties in the range of 53 ± 3.1 MPa and 54 ± 4.57 respectively. Alimuzzaman et al. [7] used a UD flax reinforcement obtained by an air-laid technique mixed with a compression process, but again the properties obtained were relatively low (tensile strength of 80.3 MPa and Young's modulus of 9.9 GPa). Some authors have used paper fibers [13–15] with the injection molding process to obtain parts made of short fibers with low mechanical properties.

Until now biopolymers like PLA found market acceptance in the packaging and medicine sector, but they are still underestimated as engineering resins due to the poor thermo-mechanical properties [16]. It is thus mainly used in biomedical applications such as resorbable prostheses, biodegradable sutures, medical implants and scaffolds for tissue engineering [17]. For a use as engineering resin, PLA suffers from some shortcomings, such as low ductility and toughness, low heat distortion temperature, small rate of crystallization, high sensitivity to moisture and fast degradation by hydrolysis [18]. The degradation of PLA under aerobic conditions present in compost is highly temperature dependant, with essentially complete biodegradation in 3–4 months at 55–60 °C [19]. At temperatures well below the glass transition temperature, dramatically reduced rates of reaction can be observed. Data on hydrolysis of semicrystalline PLA fibers shows a drop in the hydrolysis rate of more than two orders of magnitude as the temperature is reduced from 55 to 23 °C [19]. Nonetheless, PLA has inferior moisture barrier properties compared to those of synthetic polymers and the management of moisture penetration of polylactide (PLA) is extremely important during the manufacturing, shipping, storage, and end-use of PLA products [20]. Moreover, the glass transition temperature (T_g) of PLA (around 55 °C) is such that it is brittle at room temperature while its elastic modulus drops rapidly when reaching Tg [11,21]. So PLA alone has nonsatisfying impact resistance and low heat distortion temperature, which usually affect the properties of composites using PLA as matrix. For example, Plackett et al. [22] found no statistical difference in mean impact resistance between the PLA processed at 190 °C and a PLA/jute composite processed at 200 °C. Bax and Müssig [12] observed that the impact strength of PLA/flax composites increases with increasing fiber proportion, but the highest value is still 31% lower than the value for pure PLA. As raised by Beldzki et al. [16], it seems that the reinforcing effect of several natural fibers reduce the ductility of composites made of PLA, thus resulting in lower impact strength [11,22,23]. So more developments are required to make PLA efficient at higher temperatures and avoid degradation when used over a long period of time. Therefore and with technological advances allowing to avoid these drawbacks, manufacturing of PLA light-weight composite parts with a high impact resistance would lead to new application fields.

To our knowledge, the potential of PLA in terms of mechanical properties when combined to continuous UD natural fibers has never been demonstrated, considering the high melt viscosity of PLA and inherent difficulty to impregnate the fibers without affecting their orientation. In this research a high viscosity biosourced PLA matrix is used to impregnate UD flax reinforcements with a compression molding process. The mechanical properties of composites made of UD flax alone and UD flax with a paper layer binder are compared. Two objectives are pursued: First, show it is possible to obtain good quality unidirectional composite laminates (with well oriented fibers and high mechanical properties) when using a viscous PLA matrix and second, demonstrate the potential in terms of composite mechanical properties of a new flax-paper reinforcement for which the paper binder is used to maintain the UD flax fibers in each layer of reinforcement. Results for tensile tests, flexural tests, and impact tests are presented and discussed, along with optical microscopy images of the two composites under study. To support the results, comparison with literature data for composites based on UD and fabric plies is also presented.

2. Experimental

2.1. Materials

As supplied flax and paper fibers were used in this work to obtain reference properties for future works involving comparisons with treated fibers. As a first approach, avoiding treatments also