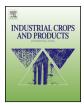


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# Evaluation of the potential of three non-woven flax fiber reinforcements: Spunlaced, needlepunched and paper process mats

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## ABSTRACT

This paper presents results from an experimental study of three types of non-woven preforms (needlepunched, spunlaced and mat manufactured using a paper-making process) intended as composite reinforcement. These are potentially very attractive for transport applications. First, the influence of processing on elementary fiber tensile properties is shown to be limited. Then the preforms are evaluated in polypropylene matrix composites and mechanical properties are determined. The structure of non-woven reinforcements is strongly dependent on the manufacturing route. By varying the fiber content it is shown that the most efficient reinforcement for flax fibers is the mat produced by paper processing. The new spunlaced reinforced composites are shown to have slightly lower tensile properties (15% lower strength, and 25% lower stiffness) compared to mat composites at equivalent volume fraction, but further optimization is possible for these materials. Based on the measured constituent properties micromechanics models have been used to estimate composite stiffness. A good correlation is obtained between test results and model predictions.

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# 1. Introduction

Flax fibers have been shown to possess good mechanical performances as a reinforcement for composite materials (Baley and Bourmaud, 2014) and these properties are constant from one year to the next (Lefeuvre et al., 2014). Oilseed flax fibers may also be used (Pillin et al., 2011), and the replacement of glass fibers by flax can result in a reduction in environmental impacts (Le Duigou et al., 2011). Polypropylene (PP) is a widely used polymer combining mechanical performance with low cost, so combining PP matrix with flax fibers provides recyclable composite materials with interesting properties (Bourmaud et al., 2013; Bourmaud and Baley, 2007). A combination of biodegradable polymers e.g., PLA and flax reinforcement offer the possibility to biodegrade the parts at the end-of-life (Alimuzzaman et al., 2014, 2013).

In plants the flax fibers support the stem and are assembled in bundles (Bourmaud et al., 2015; Morvan et al., 2003). Their properties are very anisotropic (Baley et al., 2006) as a result of their

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http://dx.doi.org/10.1016/j.indcrop.2015.10.008 0926-6690/© 2016 Published by Elsevier B.V. development during plant growth (Gorshkova et al., 2003). A retting step (usually performed by laying the stems on the ground in the field) is necessary in order to facilitate fiber extraction. This step must be closely controlled as it influences the mechanical properties of the fibers (Martin et al., 2013). The stems are then scutched in order to extract the fibers. After scutching, the fibers are in the form of scutched fibers or tows. Although the mechanical behavior of these two types of fiber are similar (Martin et al., 2014) their forms are quite different. The scutched fibers are assemblies of fiber bundles with lengths similar to those of the plant (aligned bundles) while the tows are made up of fibers without a particular orientation and containing a fraction of shives. The scutched fibers have a higher commercial value as after a single additional operation (combing to produce a ribbon) threads can be obtained. The tows require further mechanical operations.

Composite reinforcements can be continuous e.g., unidirectional, bidirectional, cross-ply, multidirectional, or discontinuous e.g., random chopped fiber mat. Each reinforcement architecture is designed for a particular domain of application. Fibers with a high aspect ratio and randomly distributed in-plane can be used to manufacture structural parts with a quasi-isotropic behavior. This type of 2 D reinforcement is useful for panel manufacturing, whereas unidirectional fibers are sought for beams.

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Several techniques are available to manufacture flax reinforcements. The spunlacing, needle punching and paper- making techniques are three non-wovens industry processes available to produce discontinuous flax fiber preforms. Spunlacing and needle punching are techniques used to give a web of fibers sufficient cohesion by mechanical bonding, while paper-making technique (wetlay) allows the production of a web where the fibers are bonded by hydrogen bonds.

The spunlacing technique also called hydroentanglement allows to bond fibers in a web by means of high-velocity water jets. The interaction of the water jets with fibers in the web increases the fibers entanglement and induces displacement and rearrangement of fibers segments in the web. The web of fibers is generally produced by carding.

During the needle punching the fibers of a web are mechanically entangled by reciprocating barbed needles through the web. The web is also generally produced by carding and then cross-lapped to increase the batt density.

The wetlay process is a technique derived from the papermaking technique. The general principle comes as follow. The chopped fibers are first poured in water and then transferred to a stirred water tank. The fibers are deposited on a drainage wire to produce a sheet. This sheet is then dried by suction.

The book of Russell (2006) gives additional information and references about non-wovens manufacturing.

Needlepunching can be used to produce felts with an areal density of  $100-4000 \,\mathrm{g}\,\mathrm{m}^{-2}$ . Spunlacing provides thin non-wovens of areal density  $15-400 \,\mathrm{g}\,\mathrm{m}^{-2}$  but experimentally, hydroentangled flax and hemp fabrics up to  $1500 \,\mathrm{g}\,\mathrm{m}^{-2}$  have been produced (Russell, 2006). Both techniques allow different fibers to be mixed e.g., flax and PP to manufacture semi-finished products. The wetlay process or paper process can be used to manufacture mats with an areal density between 50 and  $2500 \,\mathrm{g}\,\mathrm{m}^{-2}$ .

Handling of the preforms during composite manufacturing requires a sufficient resistance in the dry state. The resistance of the needlepunched and spunlaced non-wovens is given by mechanical entanglement of the fibers, while the resistance of the wet laid mat is only provided by hydrogen binding between the fibers.

In Europe, the automotive sector is the main user of non-woven reinforced thermoplastic composites, usually for non-structural applications (Chen et al., 2008; Fages et al., 2012; Mieck et al., 1996; Shah, 2013). The main drivers for use of plant fiber composites in automotive applications are the demand for lightweight parts, which leads to a lower fuel consumption, and good recycling possibilities of the components made with thermoplastic polymers, reducing the waste disposal problem (Huda et al., 2008). Other factors that are promoting their applications in the automotive sector are: reduction of greenhouse emissions, competitive pricing, technical advantages e.g., longer lifetime of tools, growth opportunities for agriculture, and societal benefits i.e., health and safety improvements. These parts are mainly processed by stamping or compression molding. Weight reduction is sought for interior applications but acoustic and thermal insulation are also required. For the latter parts may be processed with up to 70% void content.

Thermoplastic matrix composites reinforced with flax nonwovens have been studied previously. Needlepunched flax composites were investigated by (Garkhail et al., 2000; Mieck et al., 1996; Oksman, 2000; Stamboulis et al., 2000). Wet laid flax mats composites were studied by (Bodros et al., 2007; Bos et al., 2006; Roussière et al., 2012). Spunlaced flax reinforced composites were studied by Chen et al. (2008).

Reinforcements with fibers randomly oriented in-plane, or mats, are interesting when quasi-isotropic properties are sought. Roussière et al. (2012) showed that such mats can be produced from flax fibers using paper-making techniques. When these mats are impregnated with a PLA matrix they show tensile properties which are similar to those of glass mat reinforced unsaturated polyester. Knowing the elastic properties of the elementary flax fibers (Roussière et al., 2012) showed that it is possible to estimate the composite properties. This requires knowledge of the in-plane fiber distribution, which is not completely random. The degree of division of fiber bundles and their geometry i.e. the aspect ratio, must also be known.

During composite manufacture the heating cycle will also affect the properties of the fibers (Bourmaud and Baley, 2010) so it is essential to control temperature and time. (Gourier et al., 2014) showed that exposure at 190 °C for 8 min, typical parameters for PP molding, did not significantly affect flax fiber properties. The reinforcing efficiency of flax fibers is strongly dependent on the interface areas between fibers and matrix so bundle division plays an important role (Coroller et al., 2013). This division depends on the technology used to prepare the reinforcement.

In this study three different technologies have been examined, which allow reinforcements to be produced with fibers randomly oriented in the laminate plane: these are non-wovens produced by spunlacing or needlepunching, and mats produced by paper processing techniques. Although these three do not address exactly the same markets they can all be used with natural fibers. The choice of one or another technology depends on the tools available, the cost/performance ratio and the drapability of the preform. Mat is the name generally given to random reinforcements in the composite material industry, particularly short glass fibers, and it can be applied to the paper industry products.

The aim here is to compare the reinforcement capacities of a modified PP when it is reinforced by layers of flax fibers produced by spunlacing, needlepunching and paper processing. To allow a comparison, it is necessary to know the mechanical properties of elementary fibers, and the microstructure of both preforms and composite materials. First the influence of processing on the reinforcement structure and the mechanical properties of the elementary fibers has been examined. In a second section the reinforcement in the composite and resulting composite properties are studied. The composite stiffness has then been analyzed using micromechanics models.

## 2. Materials and methods

#### 2.1. Flax non-wovens manufacturing methods

Three types of flax non-wovens were studied in this paper. Needlepunched and spunlaced non-wovens were manufactured from the same flax batch, random fiber mats were studied as a reference material, manufactured using a paper-making process. The flax plants, of the Suzanne variety, were grown in France in the region Nord-Pas-de-Calais in 2009. At maturity, flax stems were pulled from the ground and laid on the field to allow dew-retting. After harvesting, the flax stems were scutched by Van Robaeys Frères (Killem, France). Scutched fibers were cut to a length of 50 mm and refined in a Laroche opening machine. After refining, the flax bundles measured 42  $\mu$ m in diameter and 38 mm in length. The average aspect ratio was approximately 1000. Both spunlaced and needlepunched non-wovens were manufactured with refined fibers.

Spunlaced non-wovens were manufactured by Norafin Gmbh. Refined flax fibers were dry laid by carding to produce a web, before being consolidated by hydroentanglement. The water jet pressure was up to 100 bar. Following hydroentanglement, water was removed by suction and the non-woven was air dried ( $180 \,^{\circ}$ C) before final roll-up. The surface density of the non-woven was  $80 \, g \, m^{-2}$ .

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