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# Mechanical analysis of elementary flax fibre tensile properties after different thermal cycles

Clément Gourier, Antoine Le Duigou, Alain Bourmaud *, Christophe Baley

*University of South Brittany, LIMATB, C. Huygens Research Center, 56100 Lorient, France*

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ABSTRACT

Knowledge of the mechanical properties of biocomposite components, especially flax fibres, is of great interest for understanding reinforcement mechanisms. The aim of this work is to study the mechanical behaviour of flax fibres after different thermal treatments, representative of three thermoplastic composites process temperatures. Firstly, a specific analysis was conducted to identify the three different types of mechanical behaviours and study the fibre performances according to the thermal treatments. Secondly, the microfibrillar realignment process was carefully studied thanks to an analysis of the tangent Young's modulus – strain curves and of the stress at different phases of the loading. Then, to better understand the influence of the temperature, TGA experiments were carried out on flax fibres after the thermal cycles. We highlighted a significant decrease of the mechanical performances at 250 °C which could be explained by some evolutions of their mechanical behaviour induced by structural modifications of the cell walls.


## 1. Introduction

Natural fibres, such as hemp and flax, are considered potential alternatives to glass fibre for composite materials. Natural fibres show good longitudinal mechanical properties [1–3], low density (1.5 compared to 2.5 for glass) [4] and offer environmental benefits evidenced by life cycle analysis [5]. Among plant fibres, flax could be considered as being the most interesting due to its mechanical properties and its availability at European scale [6].

A schematic synthesis of the flax fibre is shown in Fig. 1. The elementary fibre could be assimilated to a composite itself, where the partly crystalline (around 70%) cellulose mesofibrils are arranged in spirals [7] within an amorphous polysaccharide matrix made up of structural hemicelluloses (EOH) and matrixpectins (EH) [8], and oriented at an angle of around 8–10° [9] with respect to the fibre axis. This complex and multi-layer structure exhibits several levels of interfaces and provides interesting mechanical properties.

Mechanical properties i.e. Young's modulus, stress and strain at break of flax could be measured on elementary fibres. The complex biochemical composition and structure of flax fibre previously described has consequences in its tensile behaviour. Duval et al. [10] have shown for single hemp fibres that overall tensile behaviour can be divided into three distinct categories: the first one is truly elastic (TI), the second, includes loss of linearity (TII) and the third one is complex with either elastic and plastic behaviour (TIII). The same distinctions were evidenced by Placet et al. [11] and Pickering et al. [12], always on hemp fibres. Such observations could be due to the differences of biochemical composition and structure of fibres inside the same batch. Therefore, the change of fibre structure could be followed by carefully studying tensile curves of flax fibres as described by Lefeuvre et al. [13] on several varieties of flax fibres. They proved that samples exhibiting a large proportion of the third type of behaviour were characterised by high tensile properties. In a previous work, Aslan et al. [14] found that green (non-retted) fibres exhibited a nearly linear TI behaviour while retted cottonized flax fibres showed TI behaviour as well as a complex non-linear TIII stress–strain behaviour; they explained the differences by the presence of defects induced by the mechanical treatments.

Due to the nature of the constituents of the cell walls, the temperature, and as a consequence, the water content, have a strong influence on the mechanical properties of plant fibres. Indeed, water could act as a polysaccharides plasticizer [15] by penetrating the amorphous zones [16] and induce a decrease in the stiffness or a cellulose fibrils division [17]. After heating for 14 h at 105 °C [18], it was shown that the tensile strength of the fibres is strongly affected by this drying cycle by becoming damaged and the loss of interactions between mesofibrils and pectin matrix. Moreover, Gassan and Bledzki [19] have shown that the mechanical properties of jute and flax fibres start to be affected by the temperature

* Corresponding author.
*E-mail address:* alain.bourmaud@univ-ubs.fr (A. Bourmaud).

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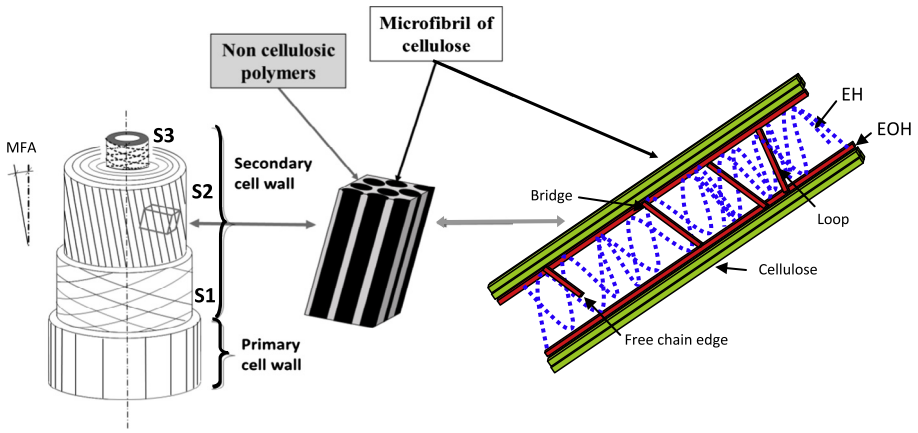


**Fig. 1.** Schematic representation of flax fibre showing helix arrangement of cellulose fibrils and of the chemical structures in the S2 layer of flax cellulosic fibres. (EH: matrix non cellulosic polymers and EOH: structural non-cellulosic polymers) [10]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

at around 170 °C. These results were corroborated by Placet [20], on hemp fibre bundles and for different temperatures. In order to improve the thermal resistance of plant fibres, it is possible to apply thermal treatments inspired by wood technology such as Duralin® [21].

Thanks to their mechanical performances, plant fibres are commonly used for thermoplastics reinforcement; their incorporation ensures a well-known improvement of the matrix' mechanical properties [22]; moreover, they provide a positive effect on the shrinkage due to their interesting aspect ratio [23]. When they are associated with bio-based and/or biodegradable polymer, the obtained biocomposites can be compostable and recyclable composites [24]. Nevertheless, the majority of thermoplastic biopolymers currently used for biocomposite manufacturing require a process temperature of over 150 °C and a few could be used under 200 °C which leads to plant fibre degradation. Many works highlight thermal degradation of flax fibre [18,19,25–29] but very few deal with effect of temperature on mechanical behaviour.

This present study deals with the effects of a different thermal cycle, in temperature and time, typically used for compounding and processing by the injection of flax fibre with PBS, PLA and PA11. The fibres' tensile properties were evaluated and analysed using a Weibull statistical approach. Then, this mechanical behaviour was linked to property variations and changes to the components organization. Finally, TGA measurement and SEM observations were carried out to complete the mechanical analysis.

## 2. Material and methods

### 2.1. Flax fibres

Flax plants, variety Marylin (2003), were cultivated on the sandy loam plateau in Neubourg (Normandy). The stems were pulled and submitted to dew-retting for about one month until the harvest. Then, they were scutched and hackled by the company Coopérative de Teillage de Lin du Plateau du Neubourg (CTLN, Le Neubourg, Normandy, France).

### 2.2. Thermal cycles

Fibres were exposed to three different temperatures (140, 190 and 250 °C) in a laboratory oven for a similar period of 8 min each, representative of a compounding (3 min) followed by an injection molding cycle (5 min) for PBS, PLA and $PA_{11}$. These different times were measured by some of the authors [27]. These thermal cycles and polymers were chosen due to the wide range of temperature, properties, behaviour and therefore applicability.

A Volca muffle furnace was used and thermal cycles were done in air atmosphere similarly representative of a conventional compounding and injection process.

### 2.3. Tensile tests on single fibres

Tensile tests were performed on 70 elementary fibres for each batch. A gauge length of 10 mm was chosen. The samples were clamped on a MTS type tensile testing machine equipped with a 2 N load cell. These tests were performed in a temperature and relative humidity controlled laboratory (23 ± 0.5 °C, 48 ± 2% RH). Longitudinal tensile properties (Young modulus, strain and stress at break) were determined for each single fibre in accordance with the NFT 25-501-2 standard. The compliance of the loading frame was taken into account.

The mean diameter value of each fibre, required to calculate Young's modulus and strength, is an average value from five points measured all along the fibre with an optical microscope.

Water uptake of natural fibres after thermal treatments is quite high [18] and the numerous samples required a long manipulation time. As water influences the mechanical properties, it was essential to let the fibres recover their hygrometric equilibrium to avoid scattered and non-representative results [26].

When breaking properties present relatively high standard deviation and when brittle material is assumed, the weakest link theory can be applied with the Weibull theory. The Weibull cumulative distribution assumes that only one kind of flaw leads to failure [30] and is typically given by Eq. (1):

$$P_f = 1 - \exp\left[-\left(\frac{\sigma}{\sigma_0}\right)^m\right] \quad (1)$$

where $P_f$ is the probability of failure, $\sigma$ is the tensile strength, $\sigma_0$ is the Weibull scale parameter, m is the Weibull modulus or shape parameter. Weibull shape (m) and scale parameter are estimated by plotting the probability of failure $P_f$ as a function of tensile strength in Weibull coordinates. In order to decrease strength calculation uncertainty due to diameter estimation, Weibull statistics could be done by using strain at break based on the Virk et al. [31] recommendation. The probability index (Eq. (2)) is the lowest partial estimator for sample sizes more than 40 in accordance with Trustrum and Jayatilaka [32]:

$$P = \frac{i}{n+1} \quad (2)$$