HOW THE MORPHOLOGY OF A FLAX FIBRE INFLUENCES ITS MECHANICAL BEHAVIOUR

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ABSTRACT

Owing to their low density, natural fibres like flax exhibit specific mechanical properties similar to those of glass fibres. Hence, their use as a reinforcement of polymeric matrices can be considered as a possible application in addition to the usual textile market. In order to get a better knowledge of their mechanical behaviour, tensile tests have been performed on several elementary flax fibres. The aspect of a typical stress-strain curve is described according to the different components present within the fibre. The results tend to prove that a flax fibre which is submitted to a tensile loading undergoes an internal reorganization of its constitutive elements. Besides, flax fibres / polyester composites were processed and tensile tested. Their tensile behaviour exhibits the same initial non-linearity as the one of a flax fibre.

1. INTRODUCTION

In the framework of the sustainable development carried out for many years by the European governments, green materials are getting more and more importance in many domains, especially in the automotive industry. Among these green materials, the composites made with a polymeric matrix and natural fibres often exhibit several advantages: a low cost of the raw materials, a competitiveness of the mechanical properties compared with the glass fibres reinforced polymers, a relative lightness, a worldwide availability of the fibres… Nevertheless, the difficulty to process such composites, the generally poor fibre-matrix adhesion and the lack of knowledge on the fibre behaviour are considered as the main hindrances to the development of these natural fibre based composites. In this paper, the flax fibre is studied from both mechanical and morphological points of view in order to enhance the understanding of its tensile behaviour and to model its multilayer structure. After a brief presentation of a flax fibre, a typical stress-strain curve of a fibre submitted to a tensile loading is described and a hypothesis of reorganization of the different polymers present in the cell walls is formulated. Then, flax fibres reinforced composites were processed by compression moulding and tensile tested up to rupture. The stress-strain curves and their mechanical properties are discussed and related to the flax fibre characteristics.

2. PRESENTATION OF A FLAX FIBRE

A flax fibre is a natural and biodegradable composite which exhibits good mechanical properties and low density (1.54). Its mean specific mechanical characteristics (i.e. reported to the material density) are 42 GPa and 675 MPa for the Young's modulus and the failure stress respectively [1], to be compared with those of the E-glass fibres which reach 30 GPa and 1000 MPa, respectively [2]. A flax fibre can be described as a 20 µm wide cylindrical composite with concentric layers of cell walls which differ in terms of chemical composition and morphology (see Figure 1). The thickest cell wall is itself made of microfibrils of cellulose which are embedded in a polysaccharidic matrix and lay at about 10° from the fibre axis [3]. The high volume fraction of cellulose (about 70 % [4-7]) confers to the flax fibre its good tensile properties. The size of the internal lumen can vary from 1 to 10 µm without any obvious correlation with the fibre diameter [8].

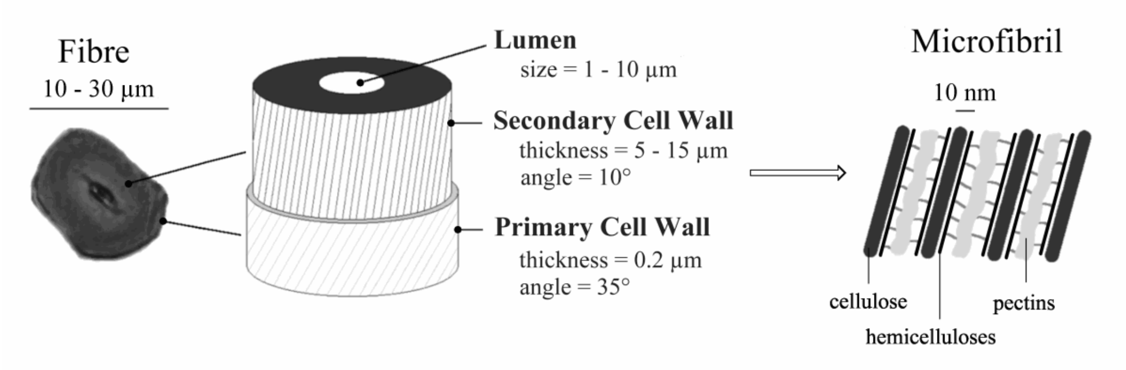


Figure 1: Microstructure of a flax fibre.

3. TENSILE BEHAVIOUR OF A FLAX FIBRE

When a flax fibre is tensile loaded up to rupture, its stress-strain curve is often initially non-linear until a deformation of about 1.5 % and then becomes linear (see Figure 2a). The discrepancies between the different slopes can reach 20 GPa, which makes it difficult the determination of the Young's modulus. As the behaviour during unloading is a real indicator of the elasticity of a material and as the value of the slope during this stage is very close to the one of the final slope (see Figure 2b), this last one is used to estimate the Young's modulus of each fibre. The mean mechanical characteristics calculated from about 200 stress-strain curves of tensile tested flax fibres at a cross-head displacement rate of 1 mm/min are gathered in Table 1.

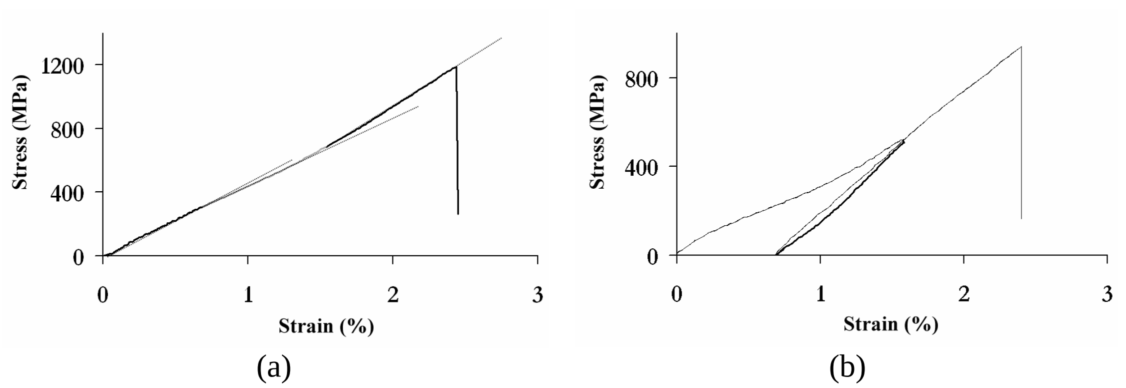


Figure 2: (a) Typical stress-strain curve of an elementary flax fibre showing different slopes; (b) example of stress-strain curve of a loaded-unloaded-reloaded flax fibre.

Diameter (µm)	Strength (MPa)	Ultimate strain (%)	Young's modulus (GPa)
17.8 ± 4.4	1025 ± 441	2.2 ± 0.9	53 ± 21

Table 1: Mean mechanical properties of the 200 tensile tested flax fibres.

On the basis of these 200 stress-strain curves, a hypothesis has been made about the deformation of a flax fibre under tension: the microfibrils would tend first to reorganize themselves along the fibre axis within the polysaccharidic matrix, and then would deform elastically until rupture. It would mean that the microfibrils (most of which being initially oriented at about 10° of the fiber axis [3]) would align themselves so that their final orientation would lay around 0°. The geometrical considerations associated to this assumption are illustrated in Figure 3.

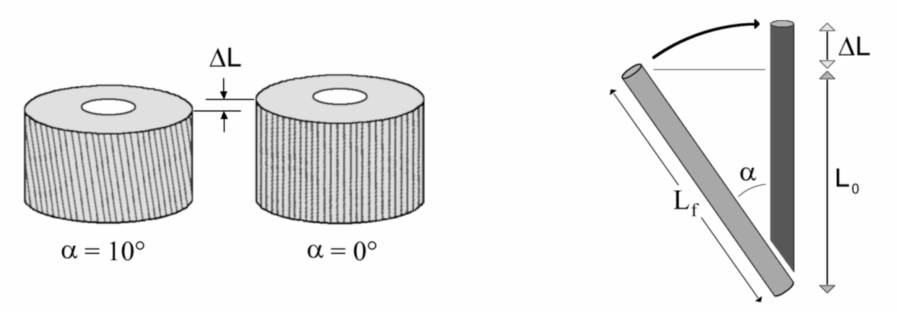


Figure 3: Illustration of the reorientation of the microfibrils within the thickest cell wall of a flax fibre.

According to this figure, the length L_f of a α-disoriented microfibril can be linked to its projection L_0 along the fibre axis by

$$L_0 = L_f \cos\alpha \qquad \text{Eq. 1}$$

The tensile loading of the fibre leads to a global extension ΔL defined by

$$\Delta L = L_f - L_0 \qquad \text{Eq. 2}$$

which brings about

$$\Delta L = L_0\left(\frac{1}{\cos\alpha} - 1\right) \qquad \text{Eq. 3}$$

that is, a global deformation of

$$\varepsilon = \ln\left(1 + \frac{\Delta L}{L_0}\right) = -\ln(\cos\alpha) \qquad \text{Eq. 4}$$

Using the value of 10° quoted in the literature, this last equation leads to a deformation of the fibre of about 1.5 %. When looking at the 200 stress-strain curves of flax fibres (some of them are displayed in Figure 4a), this deformation seems to correspond roughly to the end of the non-linear part of the curves. Other natural fibres, whose microfibril angle is known, have been tested in the same way. Some of the obtained stress-strain curves are shown in Figure 4bcd. In Table 2 are compared the literature data on the microfibril angle of these fibres with the angle calculated on the basis of their experimental stress-strain curves. The results tend to corroborate the hypothesis formulated above, that is the alignment of the cellulose microfibrils with the fiber axis during the first stages of a tensile loading.

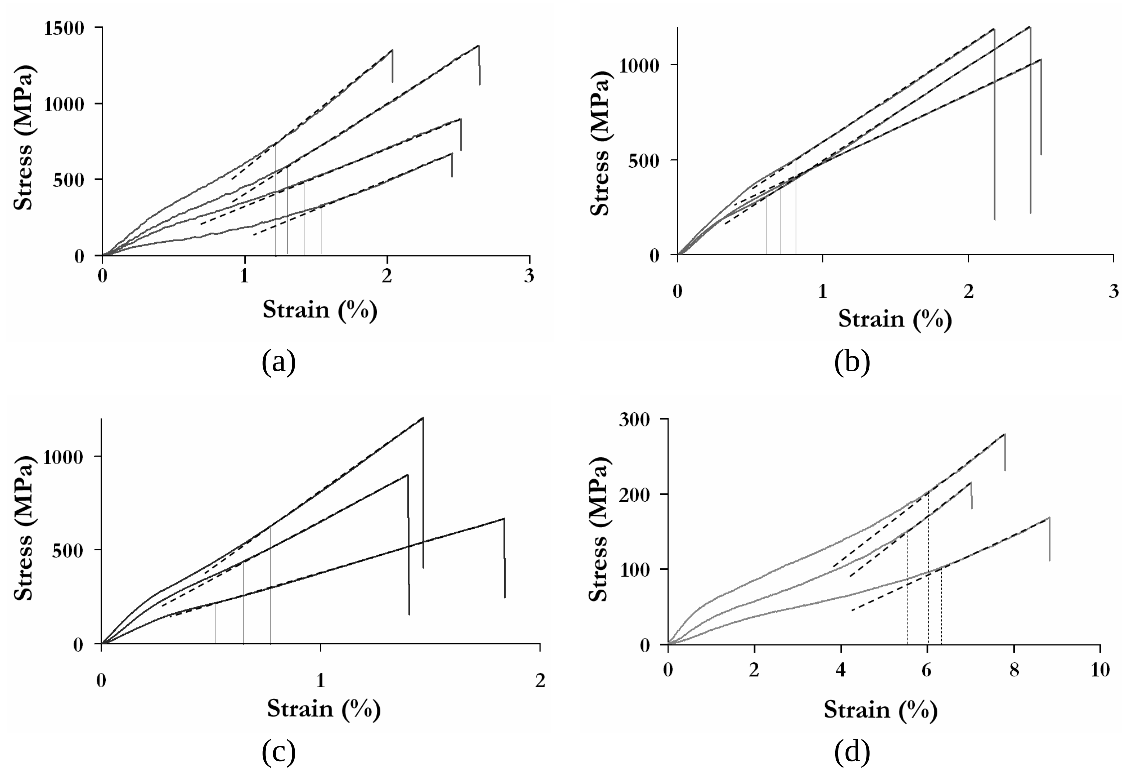


Figure 4: Stress-strain curves of natural fibres showing the deformation at the beginning of the final linear zone: (a) flax, (b) hemp, (c) ramie, (d) cotton.

Natural Fibre	Flax	Hemp	Ramie	Cotton
Microfibril angle (°) (literature data [9])	10	6.2	7.5	25
Intermediate deformation (%) (experimental data)	1.4 ± 0.7	0.73 ± 0.16	0.65 ± 0.23	5.7 ± 2.7
Microfibril angle (°) (calculated using Eq. 4)	9.4 ± 2.4	6.9 ± 0.8	6.4 ± 1.2	18.4 ± 4.9

Table 2: Literature data [9] and calculated values of the microfibril angles of some natural fibres.

In the last stage prior to rupture, the aligned microfibrils would then not only deform, but also slide one over another and the amorphous polymers would shear, which could explain that the slope in this part of the stress-strain curve does not reach the rigidity of cellulose, estimated at 137 GPa [10], but hardly half of this value (cf. Table 1).

4. TENSILE BEHAVIOUR OF FLAX FIBRES / POLYESTER COMPOSITES

Flax fibres were used to reinforce unidirectionally an unsaturated polyester matrix. Composites plates were made by compression moulding, which allows achieving a large interval of fibre volume fractions (here: from 10 % to 40 %). Their porosity was estimated at about 5 %. Forty samples for mechanical testing were cut in these plates and tensile tested up to rupture. Typical stress-strain curves of the matrix and of a composite are presented in Figure 5.

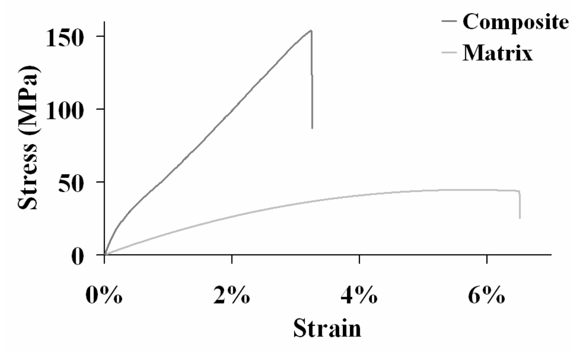
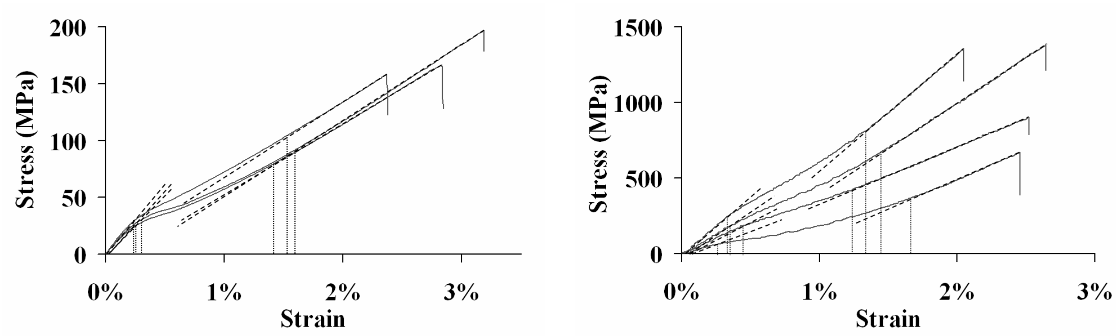


Figure 5: Typical stress-strain curves of the polyester matrix and of a flax fibre reinforced composite (t_v^f = 19 %).

It appears first that the addition of flax fibres greatly reduces the ductility of the polyester matrix, but notably increases its strength and rigidity. Secondly, as in the case of the elementary flax fibre, the stress-strain curve exhibits an initial non-linearity and a final linear part (see Figure 6). The values of deformation at the transitions between the 3 different behaviours are about the same for the composites (a) and for the elementary flax fibres (b), even if they exhibit a wider scattering in the last case. This similarity would imply not only that the synergy between the fibres and the matrix acts since the very beginning of the tensile loading of the composites, but also that, within the composites, the fibres are likely to deform as they do individually (cf. § 3). This would be in agreement with a good adhesion between the flax fibres and the polyester matrix.



(a) (b)

Figure 6: Stress-strain curves of some flax fibre / polyester composites (a) and of some elementary flax fibres (b), highlighting the similarities in their tensile behaviour.

The non-linearity of the composite stress-strain curves raises again the question about the determination of the Young’s modulus. That is why both the initial and the final slopes of the curves have been estimated, as well as the strength and the ultimate strain for each of the 40 composite samples, displayed in Figure 7 as a function of the fibre volume fraction. The mean values obtained for these parameters are gathered in Table 3.

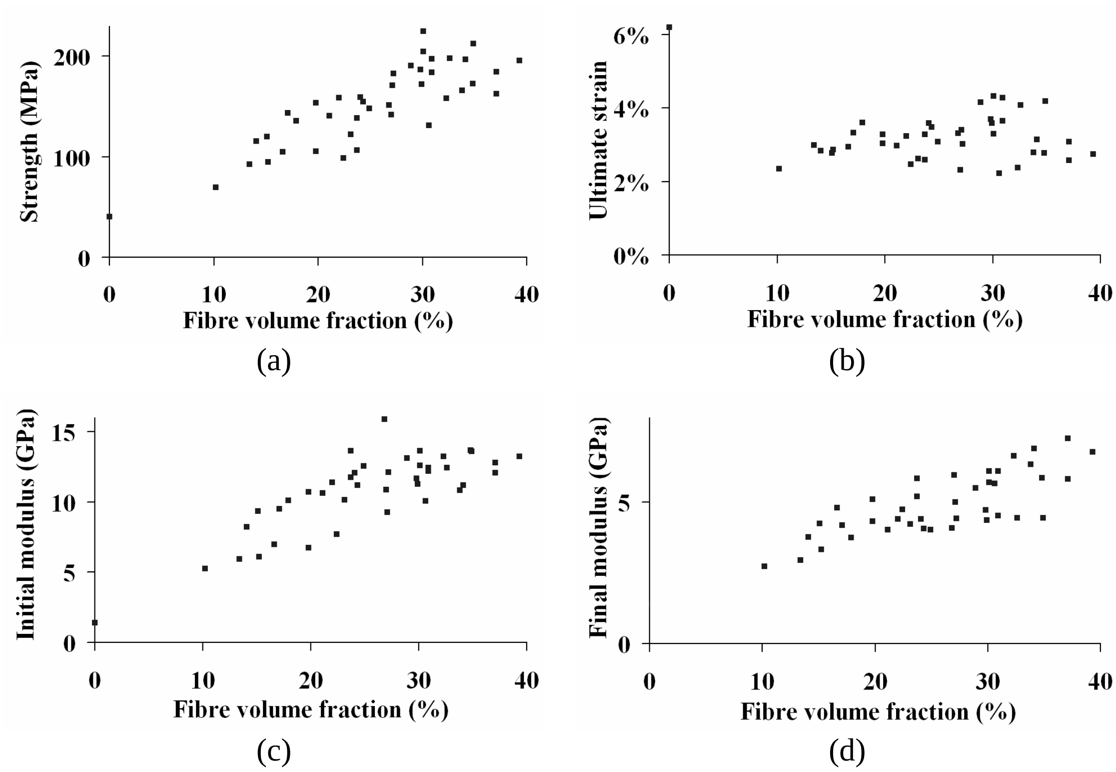


Figure 7: Evolution of the mechanical properties as a function of the fibre volume fraction.

	t^v_{fibres} (%)	$\sigma_{rupture}$ (MPa)	$\varepsilon_{rupture}$ (%)	$E_{initial}$ (GPa)	E_{final} (GPa)
Matrix	-	42 ± 8	6.2 ± 2.4	1.4 ± 0.2	-
Composites	25.8 ± 7.4	154 ± 37	3.2 ± 0.6	10.9 ± 2.4	4.9 ± 1.1

Table 3: Mean mechanical properties of the polyester matrix and of the flax fibre / polyester composites.

Except for the ultimate strain, the association of flax fibres to the polyester matrix enhances its properties quasi linearly with the fibre volume fraction. The evolution of the composite strength with the amount of fibres can give information about the strength of the flax fibres used in the composites. It is worth underlining that these ones are not elementary but rather in the shape of small bundles of a few fibres linked by a pectic matrix; in this form, they are generally called “technical fibres” and exhibit smaller properties than the elementary fibres [11-12].

As the ultimate strain of the matrix is much higher than the one of the reinforcement, the fibres are likely to break first. The fibre volume fraction above which the fibres failure is expected to lead to the composite rupture is

$$t^*_{fibre} = \frac{\sigma^R_{matrix} - \sigma^*_{matrix}}{\sigma^R_{fibre} - \sigma^*_{matrix} + \sigma^R_{matrix}} \qquad \text{Eq. 5}$$

with σ^R_{matrix} the matrix strength (42 MPa), σ^R_{fibre} the fibre strength (500 - 1500 MPa) and σ^*_{matrix} the stress undergone by the matrix when the fibres break (at about 2 % deformation); this last stress is about 30 MPa according to Figure 5. This calculation

leads to a value for t^*_{fibres} of 1 - 2 %, which implies that the rupture of all the composites processed in this study is likely to be initiated by fibre rupture and not by the matrix failure.
When the fibres begin to break, the stress undergone by the matrix is so high that it fails suddenly, leading to a composite strength of

$$\sigma^R_{composite} = \sigma^R_{fibres} t_{fibres} + \sigma^*_{matrix} (1 - t_{fibres}) \qquad \text{Eq. 6}$$

According to Figure 7a and by imposing σ^*_{matrix} as the initial y-coordinate, the slope of the regression line leads to a fibre strength of 500 MPa. This value, which approaches the lower limit of elementary fibre strength, is in good agreement with the literature data on technical fibres [11-12].

Considering now the initial Young's modulus of the composites, the same analysis as above leads to a value for the technical fibres of 36 GPa, which is lower than the value obtained for elementary flax fibres. Due to the similarity between the stress-strain curves of the composites and of the flax fibres (cf. Figure 6), the interface fibre/matrix is supposed to be well bounded. After the reorganisation of the fibres as described before, *i.e.* at a level of deformation of 1-2 %, it is conceivable that only a fraction of fibres would be efficiently acting as a reinforcing material (for example, the best impregnated, the most resistant or even the best aligned fibres). In this case, a modified law of mixture can be applied:

$$E_{composite} = \lambda t^v_{fibres} E_{fibres} + \left(1 - \lambda t^v_{fibres}\right) E_{matrix} \qquad \text{Eq. 7}$$

with λ a factor of efficiency. The regression slope which fits the best the experimental composite modulus values (E_{fibres} = 36 GPa and E_{matrix} = 1.4 GPa) leads to λ = 39 %. This would mean that less than half of the fibres present in the composites would really reinforce the matrix in the last stage of the loading. The examination (for instance by X-radiography) of the composites during their tensile deformation could help confirming or infirming such a low reinforcement effect of the polyester by flax fibres.

5. CONCLUSION

In this paper, an attempt was made to correlate the mechanical properties of flax fibres with their morphological features. A simple model has been proposed to explain the non-linear tensile behaviour of a single fibre. This model, based on a progressive alignment of the cellulose microfibrils along the tensile axis, has been successfully applied to other vegetable fibres. Nevertheless the microstructural modifications involved in the reorientation process have not yet been clearly identified. The unidirectional composites obtained by the association of technical flax fibres with a polymeric matrix exhibit a non-linear mechanical behaviour similar to the one of the single fibres. This would be in agreement with a good natural adhesion between the flax fibres and the polyester matrix. Moreover, the composites show good mechanical properties although only a fraction of the fibres seems to participate to the mechanical reinforcement during the last stage of the loading.

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