THE RELATIONSHIP BETWEEN THE TENSILE PROPERTIES OF NATURAL FIBRES AND THEIR UD COMPOSITES

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Abstract
In this paper a method is described to significantly reduce the amount of samples for single fibre testing. Futhermore, the relationship between single fibre properties and the composite behavior was investigated using this novel testing method. It was found that the tensile behavior of the technical fibre is not necessarily representative for their behavior in the composite. It is hypothesized that the intrinsic composite nature of the fibres is the main cause for the discrepancies.

1. Introduction

The accurate determination of tensile fibre properties is vital in assessing their potential for composite applications. Tensile tests on single fibres have been standardized by the ASTM C1337 standard. The complexity of the test method and the cumbersome sample preparation have led to the determination of fibre properties by back-calculation from unidirectional composite tests using a linear rule of mixtures.

However, in a number of circumstances the latter approach cannot be followed and single fibre testing is the only method that remains to determine the mechanical properties of the fibre. This may be the case when:

- the amount of fibres is limited and producing sufficiently large composite samples is difficult.
- the fibres are short and difficult to manipulate to produce UD samples.
- the fibres are not easily extractable, as is the case when exploring new types of natural fibres

This aim of this paper is doublefold. First, a method is presented to significantly speed up the single fibre testing process. Secondly, the validity of using single fibre testing to predict composite properties of natural fibres is investigated.

Technical plant fibres, used to reinforce polymers, are different from synthetic fibres in many aspects. One such difference is that all plant fibres are composite structures at multiple levels. The fibre lends its mechanical properties from crystalline or semi-crystalline regions that have cellulose chains, oriented in a preferential direction. These cellulosic regions are embedded in amorphous substances such as hemicellulose, pectin and/or lignin. Eventually a particular assembly of crystalline and amorphous regions forms the elementary or ultimate fibre. From a biological point of view this is considered as a single plant cell. This cell is in itself a composite at (sub-)micro scale, where cellulose chains act as a reinforcement in the amorphous matrix surrounding them.

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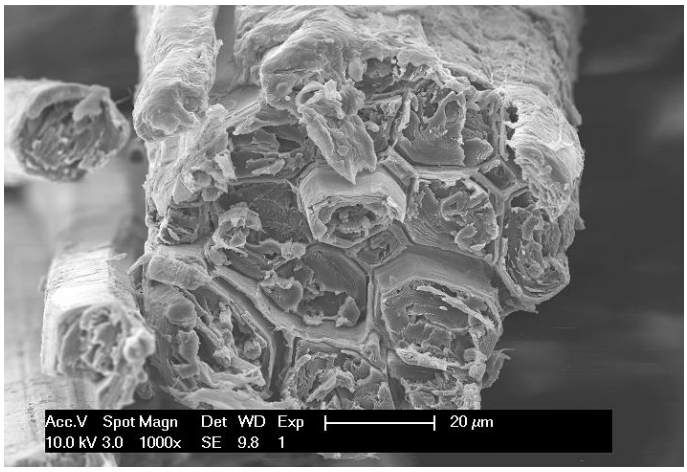

Figure 1. Microscopic image of a technical flax fibre, showing its composite structure of elementary fibres and interconnecting amorphous polymers.

A collection of elementary fibres is considered as a technical plant fibre. In this structure, the elementary fibres are held together by an amorphous matrix. When the elementary fibres are considered a homogenized entity, the structure can again be considered to be a composite, where the elementary fibres reinforce the surrounding interconnecting matrix. An example of such a structure for flax is shown in figure 1.

A composite material can be considered as a technical fibre reinforcing a thermoset or thermoplastic polymer. If the technical fibre is considered homogenous, then it is the technical fibre strength that determines the composite strength. Yet one can also consider the natural fibre composite as an array of elementary fibres, embedded in 2 types of polymers, first the amorphous material holding the elementary fibres together, then the polymer. This model implicates that the composite strength is determined by the elementary fibre strength. It is therefore hypothesized that the mechanical behavior of the single fibres is not necessarily representative for their behavior in a composite. The next sections will show the results of stiffness and strength measurements on different single technical fibres and compare them with the backcalculated properties from the resulting composites using a linear rule of mixtures.

2. Materials and methods

2.1. Flax, bamboo and coir fibres

Hackled flax fibres (*Linum Usitatissimum L.*) were acquired from Lineo NV as FlaxTape® 200, a unidirectional flax tape with an areal density of 200 g/m². The prior processing conditions of the fibres are not known. The density of the fibres is 1.44 g/cm³.

Bamboo fibres (*Guadua Angustifolia K.*) were mechanically extracted from the culm by a technique developed at KU Leuven and parenchyma tissues on the fibre surface were removed by combing the fibres multiple times. The density of the fibres is 1.44 g/cm³.

Coir fibres (*Cocos Nucifera*) were extracted at Can Tho University, Vietnam, using a mechanical method not requiring any chemicals or retting procedure, maintaining the fibre length as much as possible. Fibre density is 1.3 g/cm³.

All fibres were conditioned at 20°C and 50% relative humidity (RH) for minimum 24 hours prior to testing.

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2.2. Single fibre tests

The average cross-sectional area of each individual fibrewas determined gravimetrically with an analytical balance, accurate to 0.01 mg. C-shaped frames were cut from paper containing silicon carbide particles with an average grain size of 18.3 µm so that the gauge length of the fibres was 50 mm, as shown in figure 2. Technical flax fibres were glued to the frames, at both ends, with a thin, low strength adhesive to ensure the sand paper grains were responsible for fibre gripping. Optical flags approximately 3 mm in diameter, were attached to the fibres in the gauge length region. These markers were sprayed with black paint to produce a speckle pattern, suitable for optical strain registration. Tensile tests were carried out on an Instron 5985, equipped with a 100 N load cell at a crosshead displacement rate of 1.5 mm/min. Strain was registered optically with a 2D Digital Image Correlation set-up of Limess GmbH and results were processed with Vic2D software. Single fibre tests were executed at 50% RH and a constant temperature of 20°C.

2.3. Unidirectional composite production

A thermosetting bisphenol A diglycidyl ether resin, Epikote 828 LVEL, was used to impregnate the fibres. To initiate the crosslinking reaction, 15.2 g of 1,4-cyclohexanediamine (Sigma Aldrich) was added per 100 g of resin. The pure matrix material has a strength of 75 MPa, a stiffness of 2.9 GPa and a strain to failure of 4% [1]. All composite samples were produced by vacuum assisted resin infusion (VARI). Fibres were dried for 24 h at 60°C. Unidirectional tensile test specimen dimensions were 1 cm x 20 cm x 2 mm and were produced to size in a custom build mould. The desired fibre volume fraction was 40% and was obtained by weighing the fibres prior to impregnation by the resin. After composite production, the fibre volume fraction was corrected by measuring the effective composite thickness.

2.4. Composite tensile tests

Composite samples were conditioned for at least 7 days at 50% RH and 20°C prior to testing. An Instron 4467, equipped with a 30 kN load cell, was used to test the composite samples in tension. Strain was registered with a 50 mm gauge length extensometer. Sand paper was used to prevent slippage of the sample in the clamps. The crosshead displacement rate was set to 1 mm/min.

3. Results and discussion

3.1. Optical strain measurement in single fibre testing

When a single fibre is loaded in tension, a contact extensometer cannot be used for strain measurement for evident reasons. As described in ASTM C1337, the absence of an extensometer requires a correction to be applied to the measured stiffness. In the standard this is done by introducing the system compliance, C_s. This parameter accounts for the part of the deformation that is not attributed to deformation of the samples, as described by equation 1 where ΔL is the crosshead displacement, Δl_f is the effective elongation of the fibre and F is the force.

$$\Delta L = \Delta l_f + C_s F \tag{1}$$

Applying Hooke's law and dividing by F, equation 1 can be rewritten as in equation 2 where l_0 is the intial gauge length of the fibre and A the cross-sectional area.

$$\frac{\Delta L}{F} = \frac{l_0}{A}\frac{1}{E_f} + C_s \tag{2}$$

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It can now be seen that a plot of $\Delta L/F$ values versus l_0/A values for different gauge lengths, l_0, produces a straight line with a slope of $1/E_f$. The standard suggests taking at least three different gauge lengths, resulting in a large number of single fibre tests required to obtain a reliable fibre modulus.

In this paper, an alternative approach, based on optical extensometry, is proposed that requires less single fibre tests (the standard suggests testing at least three samples per gauge length) and results in a more reliable value for the fibre Young's modulus. Direct strain measurement on a single fibre is possible using optical extensometry when optical flags are attached to its surface. Therefore, a liquid mixture of naphta and titanium dioxide was carefully applied around the fibre and left to dry. The titanium dioxide particles adhere to the fibre and provide a base for optical extensometry. These dots were speckled with black elastomeric paint enabling the processing with image correlation software.

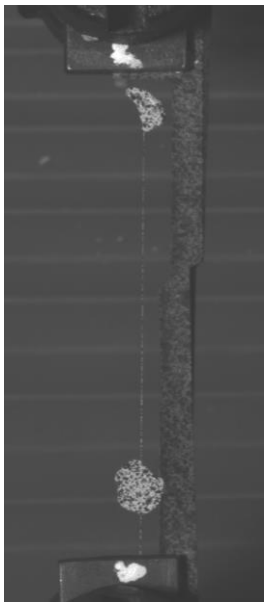

Figure 2. Mounted testing frame containing a single fibre with optical flags attached to it.

From the results obtained in a validation experiment on fibres with well known properties, carried out by the authors, it is clearly seen that the correction method suggested by ASTM C1337 significantly underestimates the stiffness, whereas the result obtained from the optical strain measurement corresponds well with the Young's modulus of the fibre. This result shows that the assumption of a constant system compliance, as put forward in the standard, does not hold in all cases. Indeed, if slippage was to occur between fibre and frame or frame and clamp, this would result in a non-constant system compliance rendering the method invalid. The test results of this validation method will be published in a journal paper, which is already submitted.

3.2. Technical natural fibre properties

Using the technique described above, the stiffness and strength of flax, bamboo and coir fibres were measured. The resulting stress-strain curves of the single technical fibre tests are shown in figure 3 and the results are summarized in table 1.

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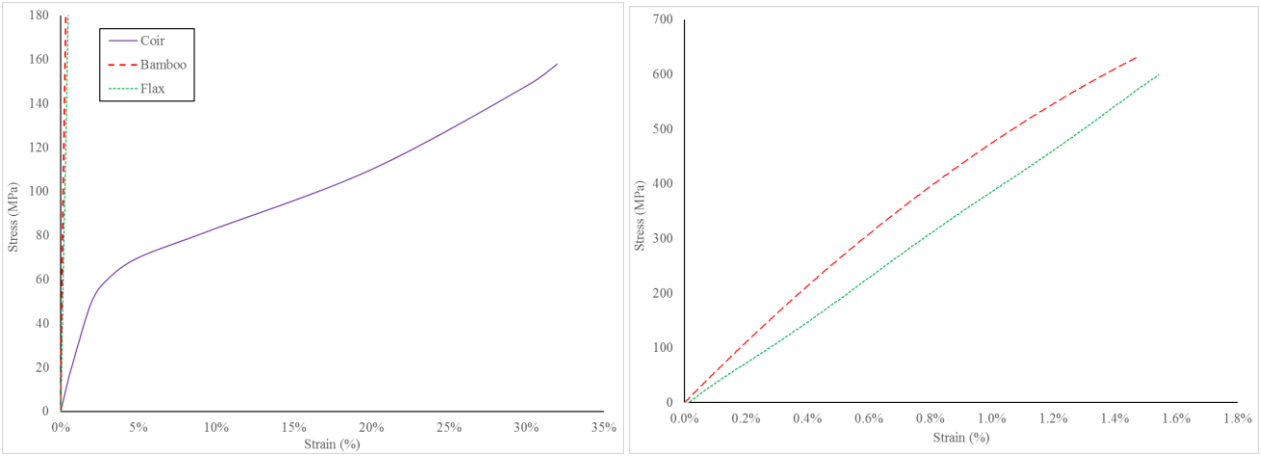

Figure 3. Stress-strain curves of flax, bamboo and coir single technical fibres. Strain was measured using optical extensometry.

Table 1. Single fibre stiffness (E_f), strength (σ_f) and strain to failure (ε_f) of flax, bamboo and coir single technical fibres.

Fibre	E_f (GPa)	σ_f (MPa)	ε_f (%)
Coir	3.3 ± 0.4	138 ± 33	30 ± 5
Bamboo	50.7 ± 6.9	568 ± 190	1.2 ± 0.4
Flax	46 ± 11	695 ± 191	1.6 ± 0.3

From figure 3 and table 1 it is clear that coir has the largest strain to failure and lowest modulus. This is due to its high microfibrillar angle [2]. Furthermore, it can be seen that the tensile behaviour of flax and bamboo fibres is quasi linear. For flax, the behavior is completely different from that of the elementary fibre. The latter typically display non-linear tensile behavior, possess a tensile strength that is up to two times higher and exhibit failure strains around 2-2.5% [3]–[5]. This discrepancy can be explained considering the composite nature of the fibre. It has been established that the interconnecting hemicelluloses and pectins that bind the elementary fibres are inherently weak and compliant [6], [7]. These may cause additional deformations during fibre loading by elasto-plastic shearing of the interphases. Damage in this interphase, present before or developed during the test, may lead to premature failure of the technical fibre.

3.3. Relating fibre properties to the unidirectional composite behavior

The stress-strain curves of all unidirectional composites are shown in figure 4 and summarized in table 2.

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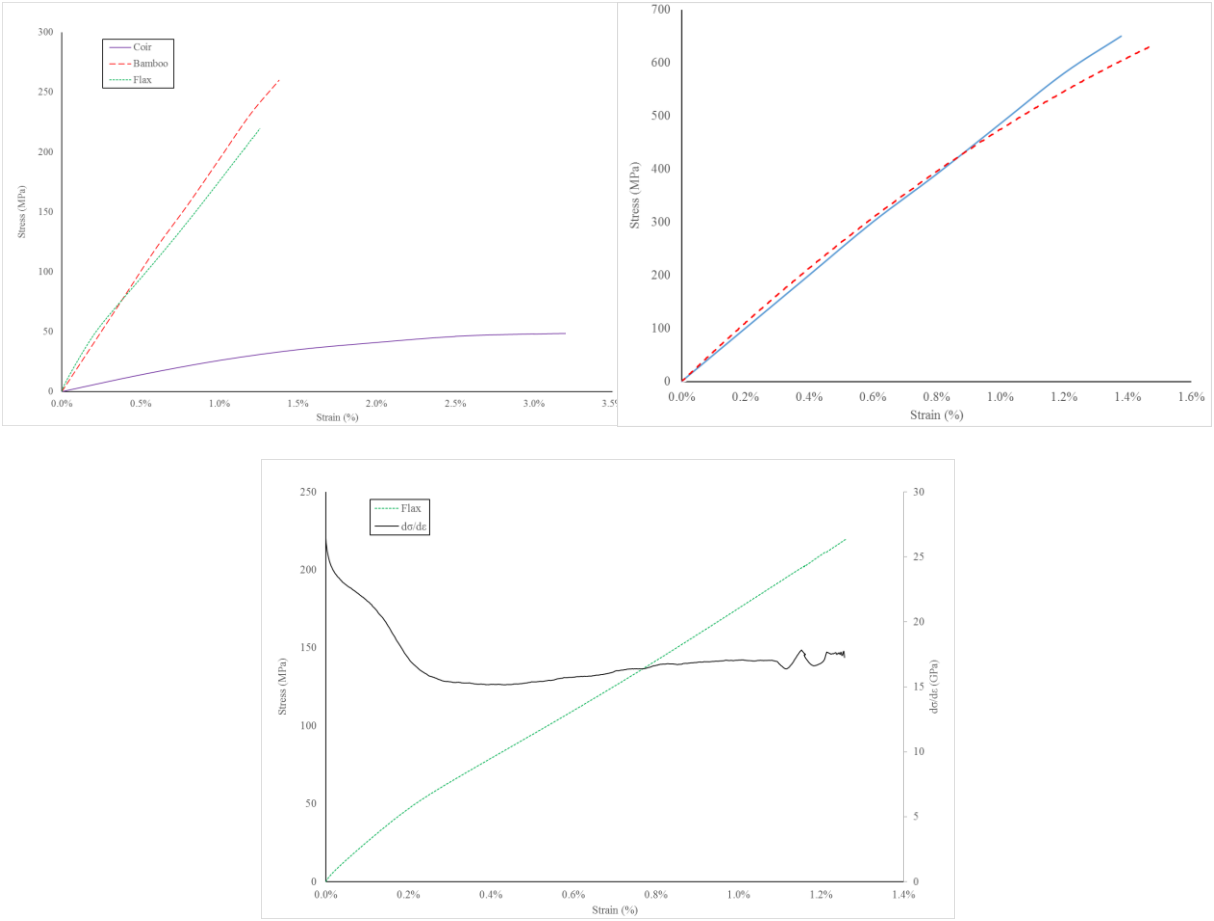

Figure 4. Left: Stress-strain curves of flax, bamboo and coir fibre composites (volume fraction of 40%). Right: Back-calculated stress-strain curve of a bamboo fibre from the composite (blue line) and measured stress-strain curve on a technical bamboo fibre. Below: Flax composite stress-strain curve with the local gradient of this curve as a function of strain.

It can be seen from figure 4 (right) that the response of the bamboo fibre composite to tensile loads is similar to that of technical bamboo fibres. Stiffness, strength and failure strain are identical when the values are backcalculated from table 2 with the linear rule of mixtures.

For coir fibre composites, the strain to failure is significantly reduced compared to the technical fibres. However, stiffness and strength are maintained. The tenfold decrease in failure strain can be related to the constraining effect of the matrix on the reorientation of the fibrils in the elementary coir fibres. It was mentioned before that their microfibrillar angle is high, approximately 30-49°. During tensile loading of a fibre these fibrils will tend to align themselves with the loading direction. In a single fibre test this movement is largely unrestricted but in a composite, the matrix prevents rotation of the fibrils. This may lead to limited strain development in the fibre as reflected by the limited strain to failure.

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Table 2. Composite initial stiffness ($E_{c1,}$), final stiffness (E_{c2}), strength (σ_c) and strain to failure (ε_c) of flax, bamboo and coir unidirectional composites (volume fraction of 40%).

Composite	$(d\sigma/d\varepsilon)_{c1}$ (GPa)	$(d\sigma/d\varepsilon)_{c2}$ (GPa)	σ_c (MPa)	ε_c (%)
Coir/epoxy	3.0 ± 0.3	NA	54 ± 2	3.1 ± 0.1
Bamboo/epoxy	19.5 ± 1.0	NA	255 ± 18	1.3 ± 0.1
Flax/epoxy	27.4 ± 2.1	18.1 ± 1.7	255 ± 7	1.2 ± 0.1

Finally, in the case of flax fibre composites, the stress-strain curve exhibits non-linear behavior. As seen in figure 4 (below). Therefore an initial stiffness of the composite is calculated between 0.03 and 0.08% strain, $(d\sigma/d\varepsilon)_{c1}$, and a final stiffness between 0.8% and 1.0% strain, $(d\sigma/d\varepsilon)_2$. This shape of the stress-strain curve is very similar to that of elementary fibres but not technical fibres. Moreover the back-calculated initial stiffness is approximately 50% higher than the stiffness of technical flax fibres. This leads to the belief that elementary fibre behavior dominates the tensile response of the composite. However, the tensile strength of the composite is very close to that of the technical fibres which could indicate that the technical fibre may dominate the tensile failure of the composite by flaws that are intrinsically present inside these fibres. Strain to failure is also reduced slightly but the decrease is much less pronounced than in the case of coir fibres because the microfibrillar angle of elementary flax fibres is much lower.

4. Conclusion

Single fibre tests are a useful tool to predict the composite properties of fibres. Optical strain measurements can help reduce the amount of fibres needed to accurately determine the fibre stiffness. However, when testing natural fibres, the tensile behavior of the fibres is not necessarily representative for their behavior inside a composite. The composite modulus seems to be dominated by the modulus of the elementary fibres, whereas composite strength seems to be dominated by technical fibre strength. This is likely due to their multi level composite microstructure.

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