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Short communication

Moisture effects on the transverse compressive behaviour of single flax fibres

Anouk Chevallier [a,*], Wajih Akleh [a], Jason Govilas [a], Florian Boutenel [a], Violaine Guicheret-Retel [a], Johnny Beaugrand [b], Cédric Clévy [a], Vincent Placet [a]

[a] Université de Franche-Comté, SUPMICROTECH-ENSMM, CNRS, Institut FEMTO-ST, 25000, Besançon, France
[b] INRAE, UR 1268 BIA Biopolymères Interactions Assemblages, 44316, Nantes, France

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ABSTRACT

Studying the effects of moisture on the mechanical behaviour of single flax fibres, particularly in the transverse direction, is of key importance for the reliable use of biobased composites exposed to varying humidity levels. In this study, the apparent transverse Young's modulus evolution of single flax fibres is recorded through repeated compressive load/unload cycles conducted at three Relative Humidity (RH) levels— 40 %, 60 %, and 80 %. No significant changes in the apparent Young's modulus, determined from the unloading, were observed during transverse compression cycling or under increasing humidity conditions. The absence of apparent softening with the rise in RH is attributed to the expression of two antagonistic mechanisms: wall softening due to plasticization and structural stiffening linked to fibre compaction. Intriguingly, a noteworthy transverse stiffening is recorded at 40 % RH following the humidification and drying of the fibre. This outcome is ascribed to a hornification phenomenon.

1. Introduction

Biobased composites, reinforced with plant fibres such as flax, hemp and nettle, are the subject of an increasing number of studies, in line with the rising demand for these materials. They are highly attractive due to their favourable combination of high mechanical properties and environmental benefits [1]. Notably, flax fibre composites dominate the European bio-sourced market, primarily due to the advantageous conditions for flax cultivation in the north of France, Belgium and the Netherlands and the established facilities for fibre extraction and processing. Additionally, their tensile properties make them competitive with glass fibres [2].

However, unlike the latter ones which are not sensitive to humidity, plant fibres are hydrophilic therefore requiring specific considerations during the design and manufacturing of composite materials. When the Relative Humidity (RH) level rises in the composite's environment, its Moisture Content (MC) increases, causing the flax fibres to swell. Water molecules are adsorbed on the outer and the inner surface of the fibre and bonded to hydroxyl groups [3]. Nonetheless, this swelling is mitigated in the composite by the presence of the matrix. This can cause its plastic deformation and damage at the fibre/matrix interface, and sometimes even cracking and loss of mechanical performance [4]. This hinders the use of plant fibre composites in ever more high-performance applications subject to wide variations in MC.

The influence of moisture on the longitudinal tensile behaviour of single plant fibres has already been studied [5]. Moisture absorption leads to an apparent softening of the fibre under monotonic tensile loading. Placet et al. [6] explain this behaviour by the greater mobility of the cellulose microfibrils and the greater distance between them due to adsorption of water molecules by amorphous constituents. As a result, the reorientation of the microfibrils with loading direction is greater, as well as the slippage. In contrast, under cyclic loading, an intensification of the stiffening phenomenon is observed [7].

Knowledge of transverse properties of fibres is also essential for predicting composite mechanical performance. From the scutching and hackling phases, as flax fibres are extracted from the stems, to the production of the reinforcement, these fibres are exposed to substantial transverse stresses. These stresses are also present during composite manufacturing processes. In use, composites may be subjected to pure compression or bending. For this reason, Govilas et al. [8] measured at 50% RH level the apparent transverse Young's modulus (E_T) of single flax fibres which is 1.71 ± 0.39 GPa. The apparent transverse modulus was determined from Single Fibre Transverse Compression Test (SFTCT) by inverse method using the model derived from the one proposed by Jawad and Ward [9]. The influence of the geometric features of the plant fibres on the value identified for the transverse Young's modulus was also studied by finite element analysis by Govilas et al. [10]. The presence of a lumen leads to a decrease in apparent

* Corresponding author.
E-mail address: anouk.chevallier@femto-st.fr (A. Chevallier).

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stiffness, whereas increased ellipticity and flattening tend to increase it. Nevertheless, to the best of the authors' knowledge, the influence of the MC on the transverse behaviour has not been studied.

Thus, the aim of this work lies in studying the moisture effects on the transverse compressive behaviour of flax fibre using SFTCT in controlled environment.

2. Materials and methods

2.1. Fibre selection, separation and apparent cross-section measurement

Single fibres are extracted from a unidirectional flax tape (FlaxTape™, EcoTechnilin SAS, Valliquerville, France). They are selected randomly and isolated manually from the raw material. For the apparent cross-section measurement, each end of the fibre is glued using a photocurring adhesive (Dymax, 3099, GmbH, Wiesbaden, Germany) onto a support, called tab, to enable them to be positioned in a Fibre Dimensional Analysis System (FDAS 770, Dia-Stron Ltd, Hampshire, UK). The fibre length between the two tabs is 12 mm.

The apparent cross-section of each flax fibre is measured using ombroscopy technique (FDAS 770) in a dedicated enclosure at three different RH levels: 40%, 60% and 80%. The measurement of the apparent cross-section at different RH level with the FDAS 770 as well as the sources of uncertainty on this measurement are described in Supplementary Data 1. The average minimum apparent transverse dimension - $\overline{D_{min}}$ - and maximum apparent transverse dimension - $\overline{D_{max}}$ - are the respective averages of the minimum and the maximum apparent cross-section dimensions.

The transverse cross-section of the fibre is observed, during the SFTCT, through a microscope lens paired with a camera. To this end, the flax fibres are, beforehand, cut in half of their length using a tailored guillotine, flat and perpendicular to the fibre axis. The guillotine operation is described in Supplementary Data 2. One of the two parts of the fibre is installed in the transverse compression device.

Sixteen flax fibres are prepared for SFTCT.

2.2. Single fibre transverse compression experimental setup

The single fibre transverse compression experimental setup used was developed at FEMTO-ST Institute and described in [11]. Its use is described in Supplementary Data 3. In this in-house setup, a single fibre is positioned between two platens and compressed diametrically. The originality of this device lies in the upper platen, which is itself the force and displacement sensor. This sensor is a compliant structure made up of two parts: (i) the upper part moved by the linear actuator, (ii) the lower part in direct contact with the fibre.

It is through the movement of the High-Precision Fiducial Markers that the force and displacement are calculated as described in [12]. The force range is up to 350 mN with a precision of 10 μN. The displacement range is up to 500 μm with a precision of 30 nm.

The experimental device is housed in a separate enclosure from the FDAS's and is connected to a humidity controller (HumiSys HF InstruQuestInc., Coconut Creek, USA). RH level can vary in the enclosure from 20% to 80%.

2.3. Loading protocol

Each flax fibre is subjected to mechanical loading at different hygrometric conditions following the pathway described in Supplementary Data 4. Each mechanical loading step consists of a repetition of six loading/unloading cycles with the same amplitude, 100 μm of linear actuator displacement, corresponding to a transverse force of approximately 65 mN on the fibre. The linear actuator speed is $10\,\mu m\,s^{-1}$. At the fibre level the strain rate is approximately $1\,s^{-1}$ to reduce the expression of viscoelastic behaviour.

When the humidity in the enclosure has stabilized at the set point, a preload is applied to the fibre. This preload, ranging from 2.5 mN to 30 mN, is applied to translate the fibre until its free end comes into contact with the lower platen. In most cases, the largest transverse dimension of the fibre cross-section is positioned parallel to the compression platens, as described in [13]. The positioning of the fibre is checked with the camera, which observes the cross-section of the fibre. Pure compression can then take place. When plotting the force/displacement curves, the preload is subtracted from the force and the displacement associated to this preload is subtracted from the displacement. The preload and the repeated loading/unloading cycles are first applied to the fibre when the humidity has stabilized at 40% RH, then repeated at 60% RH and 80% RH. Finally, the mechanical preload and loading are applied again at the first RH level (40%). During the change in humidity, the fibre is not subjected to any mechanical preload or loading.

2.4. Identification of the apparent transverse Young's modulus

The apparent transverse Young's modulus, E_T, is identified using a Jawad and Ward's model [9] modified for an elliptical cross-section fibre as described in [8] and in the Supplementary Data 5. The fibre is assumed to be elastic, homogeneous and transversely isotropic. This model relates the transverse displacement U of the fibre to the linear compressive force F measured during the test, the radius R and ν_{TT} the minor Poisson's ratio. It is considered that two parallel and rigid platens compress the fibre. The half-contact width between the fibre and one of the platens is noted b. The problem is carried out as plane strain with a Hertz contact between the fibre and the platens. A high anisotropy between the longitudinal and transverse directions of the fibre is also assumed. ν_{TT} is chosen equal to 0.07 given measurement on PA11 [13] and sensitivity analysis on the Jawad and Ward's model [11]. Given U, F and R, E_T is identified by inverse method using Eq. (1).

$$U = \frac{4F}{\pi} S_{11} \left(\operatorname{arcsinh}\left(\frac{R}{b}\right) + \ln 2 \right) - \frac{2F}{\pi} \left(S_{11} + S_{12}\right) + \frac{4F}{\pi} S_{12} \frac{R}{b} \left(\sqrt{1 + \left(\frac{R}{b}\right)^2} - \frac{R}{b} \right) \tag{1}$$

with

$$b = \sqrt{\frac{4FR}{\pi} S_{11}} \tag{2}$$

and $S_{11} = \frac{1}{E_T}$, $S_{12} = -\frac{\nu_{TT}}{E_T}$, terms of the compliance tensor $\overline{\overline{\overline{S}}}$.

2.5. Statistical analysis

A statistical analysis is carried out to determine whether the distributions at the different RH levels of apparent transverse Young's moduli for the five fibres at the six unloadings are significantly different. The Shapiro–Wilk test with a significance level set at 5% shows that the distribution for each of the RH levels of the six Young's moduli identified during the five unloadings is not normal. Thus, a Kruskal–Wallis test followed by a post-hoc test, Dunn's test, is then performed with a fixed significance level of 5%. At the end of these tests, the statistical distributions of the apparent transverse Young's moduli that are not significantly different are indicated by the same letter.

3. Results and discussion

Five among the sixteen flax fibres are successfully tested all over the testing protocol. The reasons for the failures are mainly of two types: issues with acquiring force and displacement data without measurement artifacts, and the rigid body movement of the fibre during the test due to insufficient preload. Testing and post-processing for one fibre, successfully or unsuccessfully, last approximately six hours.

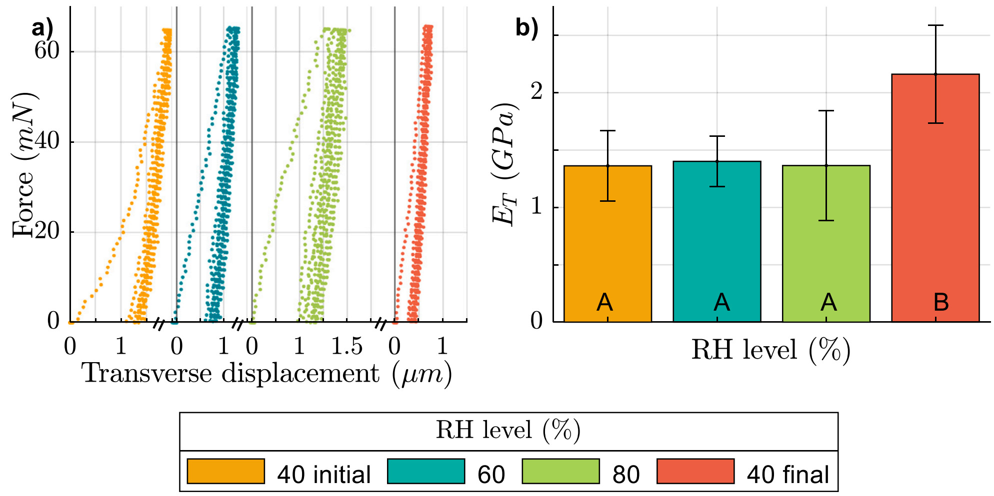

Fig. 1. (a) Example of compressive force/transverse displacement curves for a fibre at different RH levels (b) Average apparent transverse Young's modulus identified at the six unloadings for the five fibres at the different RH levels with two standard deviation error bar.

Supplementary Data 6 shows the average minimum and maximum apparent cross-section dimensions at 40%, 60% and 80% RH levels, used as an input of the Jawad and Ward modified model.

For each fibre, the compression force/transverse displacement curves obtained at different RH levels are plotted, as it is in Fig. 1(a) for one of them. The others are plotted in the Supplementary Data 7. The average apparent transverse Young's modulus identified at the six unloadings for the five fibres are presented in Fig. 1(b). The values of the average moduli for each fibre at each RH level are presented in the Supplementary Data 7.

At the initial 40% RH level, the average E_T of flax fibre is 1.36 ± 0.31 GPa. This value is of the same order of magnitude as indicated by Govilas et al. [8], who reported a range of 1.10 ± 0.23 GPa to 2.18 ± 0.91 GPa at 50% RH level under repeated progressive loading. For each fibre at the initial 40% RH level, the identified Young's moduli are in the same range along the six loading/unloading cycles as Supplementary Data 8 shows. Their maximum difference along the six unloadings is 0.5 GPa observed for the fibre 1. Mikczinski et al. [14] investigated the changes in transverse rigidity during transverse compression cycling on wood fibres. Unlike this work, their findings revealed an initial hardening of fibres that still contain lignin (spruce fibres) compared to fibres with a very low lignin content (pine fibres) which exhibited a weakening tendency. However, for these authors, the fibres have undergone more loading/unloading cycles compared to this work.

In Fig. 1(b), it can be also observed that the average apparent Young's moduli present no statistically significant difference at the chosen significance level at the initial 40%, the 60% and the 80% RH levels.

In this figure (Fig. 1b), it is important to note that the average apparent Young's modulus at the final 40% RH level shows a statistically significant difference at the chosen significant level from the average apparent Young's modulus at the initial 40% RH level, though for some of the five fibres no clear difference is observed (Supplementary Data 7). A decrease in the E_T could be expected with increasing RH level as it is observed for the apparent tensile Young's modulus. Indeed, it is well understood that water sorption in the fibre cell walls leads to plasticization of the amorphous hydrophilic constituents and a decrease in the fibre cell wall stiffness. In the case of transverse compression, the reduction in cell wall stiffness results, for a given loading, in greater deformation of the fibre (structural effect), which should lead to a lower apparent stiffness as shown by Govilas et al. [10]. Conversely, a flattening of the fibre and an increase in its ellipticity are possible independently or jointly due to repeated transverse compression. As shown by Govilas et al., these structural changes lead to a greater contact zone area of the fibre and the platens and therefore an increase in the apparent transverse stiffness of the fibre. Cellulose plasticization and in the meantime structural changes in the fibre have antagonistic effects on the E_T. Neither of these effects seems to systematically overcome the other at any of the first three RH levels tested.

Three hypotheses could explain the increase of the apparent Young's modulus at the final 40% RH level compared to the initial 40% RH level after undergoing loading/unloading cycles at different increasing RH levels.

- For the same RH level, the fibre may present different dimensions at hygroscopic equilibrium depending on whether this state has been reached following wetting or drying. Indeed, the sorption isotherms for flax fibres [15] show a hysteresis between water sorption and desorption. For the RH level considered (40%) and considering the sorption isotherms for flax fibres [15], its MC during sorption is 5% and 7% during desorption, so ΔMC is equal to 2%. According to the work of Lu et al. [15], the radial swelling coefficient, β_r, of flax fibres is 1.2. For a fibre with a nominal diameter, D, of 20 µm, the variation in diameter, $\Delta D = \frac{D\beta_r \Delta MC}{100}$, between an initial RH level of 40% and a final RH level of 40% is estimated at 0.48 µm. However, a variation of 0.48 µm for a 20 µm diameter fibre leads to an increase in the transverse Young's modulus identified by the Jawad and Ward model of only 0.02 GPa. The difference observed between the two average apparent transverse Young's moduli at the initial and final 40% RH level is 0.8 GPa which is much greater than 0.02 GPa. Therefore, the hysteresis phenomenon on the sorption isotherms leading to transverse dimensional variation of the fibre between sorption and desorption has only a minor impact on the observe change in its apparent transverse stiffness.
- Another explanation could lie in couplings between sorption and mechanical loading in particular in the mechano-sorptive effect. Mechano-sorption refers to a phenomenon observed when the material is subjected simultaneously to mechanical loading and sorption resulting from changing in hygrothermal conditions. It was demonstrated in literature that this phenomenon induces an acceleration in creep for wood under tension and also compression [16]. Mechano-sorptive creep has also been observed for wood fibres [17] and hemp fibres [7] under tensile loading. In the present study, care was taken (the load and preload are removed during the RH level changes) to decouple mechanical loading from sorption and desorption phases, eliminating the risk of expressing mechano-sorptive effect.

- Thus, the last hypothesis to explain this transverse apparent stiffening of the flax fibres when coming back to the 40% RH level after steps at higher humidity levels is hornification. Hornification is a term used in wood as well as pulp and paper literature, which refers to the stiffening of the polymer structure of the cellulosic materials constituting the cell wall upon drying or water removal [18]. Its origin has frequently been associated with the formation of irreversible or partly reversible hydrogen bonding upon drying [18]. For flax fibres, this phenomenon was hypothesized as the potential explanation for the stiffness increase of fibres in organic matrix composites submitted to moisture cycling. For wood as well as pulp and paper fibres, this phenomenon is supposed to be irreversible, even though this point is open to discussion, indicating that the phenomenon would only manifest after the first drying. The reversibility of this phenomenon for flax fibres should be further investigated to confirm that the observed transverse stiffening is related to drying induced modification of the cellulosic macromolecules constituting the fibre cell wall.

4. Conclusion

The findings of this study indicate that the surrounding RH has minimal impact on the apparent transverse stiffness of flax fibres, when transitioning from a RH level of 40% to 80%.

Interestingly, a more statistically significant increase is noted upon reverting to a RH level of 40%. By excluding assumptions of errors in fibre diameter induced by hysteresis in sorption/desorption and mechano-sorption phenomenon, the hypothesis of hornification emerges as the most plausible explanation.

Thus, the prediction of the transverse behaviour of composite materials including flax fibres at specific relative humidity levels can be considered by implementing these results in a finite element code. Furthermore, this study underscores the challenges associated with discerning the effects of surrounding environment on fibre transverse behaviour. This is especially challenging given the substantial measurement uncertainties inherent in conducting transverse compression tests at this scale and with such morphologically complex objects.

To validate the observed trends, replication of these tests on larger fibre quantities is desirable. Automation of the testing process is also essential to achieve this goal.

CRediT authorship contribution statement

Anouk Chevallier: Writing – original draft, Visualization, Validation, Software, Investigation. **Wajih Akleh:** Writing – review & editing, Validation, Software, Investigation. **Jason Govilas:** Writing – review & editing, Software. **Florian Boutenel:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Violaine Guicheret-Retel:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Johnny Beaugrand:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Cédric Clévy:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Vincent Placet:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work the authors used OpenAI ChatGPT in order to improve language and readability. After using this tool/service, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.compositesa.2024.108509.

Data availability

Data will be made available on request.

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