Mechanosorptive creep in single hemp fibres

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MECHANOSORPTIVE CREEP IN SINGLE HEMP FIBRES

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ABSTRACT

The literature on the time-dependent behaviour of single bast fibres such as flax and hemp is extremely poor. This is however particularly important in view of the development of models able to predict the long-term behaviour of plant fibres and plant fibre composites (PFCs). So, the aim of this study is to characterise the creep behaviour of elementary hemp fibres, and particularly the influence of relative humidity on it.

Single hemp fibres are shown to exhibit both instantaneous deformation and delayed, time-dependent deformation when tensile loaded. The creep behaviour appears to be a logarithmic function of time with a high deformation rate during the primary creep and a lower and constant one during the secondary creep. The creep rate is also highly influenced by the humidity and humidity variations. Much greater creep in cyclic humidity conditions than in a constant environment at the high-humidity is observed only for high rates of relative humidity variation. This mechanosorptive effect is consistent with sorption-induced stress-gradient explanations.

INTRODUCTION

Natural fibres derived from annual plants are attractive candidates to reinforce organic matrix in high performance composite applications. This use requires an accurate understanding of their mechanical properties and the development of an efficient tool for the design of reliable structure. In the last years, many efforts were concentrated on the characterisation of the tensile properties of bast fibres under quasi-static loading [1-6]. In contrast, the time-dependent behaviour has almost not been examined [7]. To the best of the author's knowledge, no information of the creep behaviour of cellulosic fibres derived from annual plants, such as hemp and flax, are available in literature. However, the integration of the viscoelastic behaviour in the predictive mechanical models of plant fibres and PFCs is necessary to ensure the reliability of the designed structures. So, the aim of this study is to provide the basis for the development of a constitutive law integrating the time-dependent behaviour of the single bast fibres derived from hemp plants.

As sometimes hypothesized in literature, and considering some similarities in their organization and composition [8], the creep behaviour of bast fibres could be similar to some of the wood fibres. However, this hypothesis needs to be confirmed. So, the aim of this study is to investigate the time-humidity dependent behaviour of elementary hemp fibres.

Anyway, the knowledge developed in the "wood" community can still be of great benefit to the "plant fibre reinforced composites" community and a detailed state-of-the-art is proposed before the presentation of the experiments and results collected on hemp fibres.

MECHANOSORPTIVE CREEP OF SINGLE WOOD FIBRES – A SHORT REVIEW OF LITERATURE

If the time-dependent behaviour of wood, wood tissue and paper has been widely studied from several decades [9-12], only a limited number of papers is related to the creep behaviour of single wood fibres [13-18]. Their creep behaviour has been studied in the context of their use in the paper and packaging applications. Elementary wood fibre has been shown to exhibit both instantaneous elastic deformation and delayed, time-dependent deformation when subjected to an externally applied load, and a permanent set when the load is removed [13-15]. Wood fibres also exhibit increases in creep compliance with increasing moisture content. Several authors also observed much greater creep in cyclic humidity conditions than in a constant environment at the highest humidity [16-18]. This accelerated creep phenomenon, induced by the sorption and desorption of water in the fibre wall, is known as mechanosorptive (MCS) effect.

Although the MCS effect has been studied since the late 1950s for wood and paper materials [10, 17-20], the topic is highly contentious and there is no widely accepted explanation for single wood fibres. Its existence is widely debated in the scientific community since negative results have been published [13-16]. Effectively, the review of literature shows that the conclusions regarding the MCS effect for single fibres are mixed; some fibre exhibit accelerated creep [17-18], whereas others seem to resist [13-15]. According to Haberger *et al*. [16] the widespread lack of observance of single-fibre accelerated creep is due to an experimental detail rather than a fundamental difference in material behaviour. Authors attributed the absence of accelerated creep to an inadequate moisture cycling rate. Habeger *et al*. [16] demonstrated experimentally for several hydrophilic fibres, such as Kevlar, lyocell, ramie and nylon 6-6, that the MCS effect happened only because of the choice of humidity cycling parameters. They argue that accelerated creep is a phenomenon consistent with sorption-induced stress-gradient explanations [16,21]. The recent results of Lindström *et al*. [22] on nanocellulose materials seems to contradict this hypothesis since the MCS creep is not significantly affected by the through-thickness moisture gradient. Authors suggest that the MCS effect in this type of materials could be attributed to the interfibril bonds or possibly to the fibrils themselves. Others mechanisms and levels of explanation can also be proposed. Dong *et al*. [18] pointed out for example the influence of the fibre morphology and more exactly on the microfibril angle (MFA) on the MCS creep. At high microfibril angles, the MCS effect could be non-existing.

MATERIALS AND METHODS

Plant material

Hemp fibres (Cannabis sativa L.) tested in this study, were procured from the LCDA Company in France. They were delivered in a jumbled state. Bundles of fibres were washed in water for 72 h at 30°C in order to facilitate the extraction of elementary fibres.

Microscopic examination

The isolated single fibres were firstly examined using polarised light microscopy (Nikon Eclipse LV 150), to determine their outer diameters. The average diameter of each fibre was computed by taking ten measurements along its length.

Tensile creep test on elementary fibre

A Dynamic Mechanical Analyser (DMA Bose Electroforce 3230) was used to perform the tensile creep tests. Creep tests were preferred to relaxation tests considering the problem of fibre buckling encountered during relaxation tests. No rotation of the fibre is allowed during tensile loading. The single fibres were glued on thin paper. After the glue drying; the paper frame supporting each elementary fibre was clamped onto the testing machine. Thereafter the fibre was conditioned to the required relative humidity up to the fibre reached moisture equilibrium. The paper frame was cut before the beginning of each test. Several experiments were performed on carbon fibres to ensure that the glue and the extremity of the paper frame do not contribute to any creep.

The clamping length was 10 mm. The applied force was measured with a load sensor of 2 N with a resolution of about 1 mN, and the displacement was measured using a LVDT with a resolution of about 0.5 μm. To achieve the control of the environment around the natural fibres the machine was implemented with a relative humidity (RH) generator. It is designed to inject humidity inside the sample chamber using a heated transfer line with a flow rate between 500 ml/min and 5 l/min. The RH is controlled inside the sample chamber using a temperature and a humidity sensor placed inside the chamber, a few centimetres from the sample. The typical operating range is 10% to 90%. The volume of the chamber (approximately 250 mm^3) was designed to ensure a RH variation rate from 1%/min to 25%/min.

Fibres were subjected to static loads. The sample elongation was measured and the strain calculated based on the elongation divided by the initial length of the fibre. The stress was calculated using the applied force and the evaluated initial cross-section of the fibre. The applied load was calculated using the fibre cross-section to ensure a same stress for all the fibres.

RESULTS

Creep / recovery at constant humidity

Fig. 1 shows the evolution of the fibre deformation when submitted to a typical creep-recovery test. During the recovery step, a small tensile load is maintained to avoid buckling of the fibre. When loaded, the fibre exhibit both instantaneous deformation ($\varepsilon_{instantaneous}$) and delayed, time-dependent deformation ($\varepsilon_{delayed}$). With stress, sufficient energy is supplied to overcome secondary bonds which defined the initial macromolecular structure of the amorphous components. The packing molecules tend to align themselves in the direction of the stress by moving into new positions. The extension is found to increase rapidly at first (during the 15 first minutes) and more slowly later. This two creep components are classically called primary creep and secondary creep. For reason of time, the tertiary creep is not considered in this study. As in many cases, the creep of elementary hemp fibres appears to be a logarithmic function of time. When the load is removed, the fibre contracts, more rapidly at first again. However the retraction rate after the first time of recovery is higher than for the secondary.

Our results also clearly show that, even after a prolonged time of retraction, a significant residual extension or permanent set remains. The instantaneous recovery (ε_{iR}), when the applied load is released, is extremely small in comparison to the initial instantaneous deformation. Only a portion of the delayed deformation is recovered after 3 hours. The existence of irreversible strain (ε_i) was already observed for these fibres under monotonic tensile loading, and even for small strain levels [23]. This was attributed to complex phenomena involving stick-slip mechanisms [24] and strain-induced crystallisation of cellulose. In the amorphous regions, and after the molecular reorganisation induced by stress, the bond reformation is assumed to be associated to a lock-in phenomenon. This could explain the large permanent set observed upon removal of the external stress.

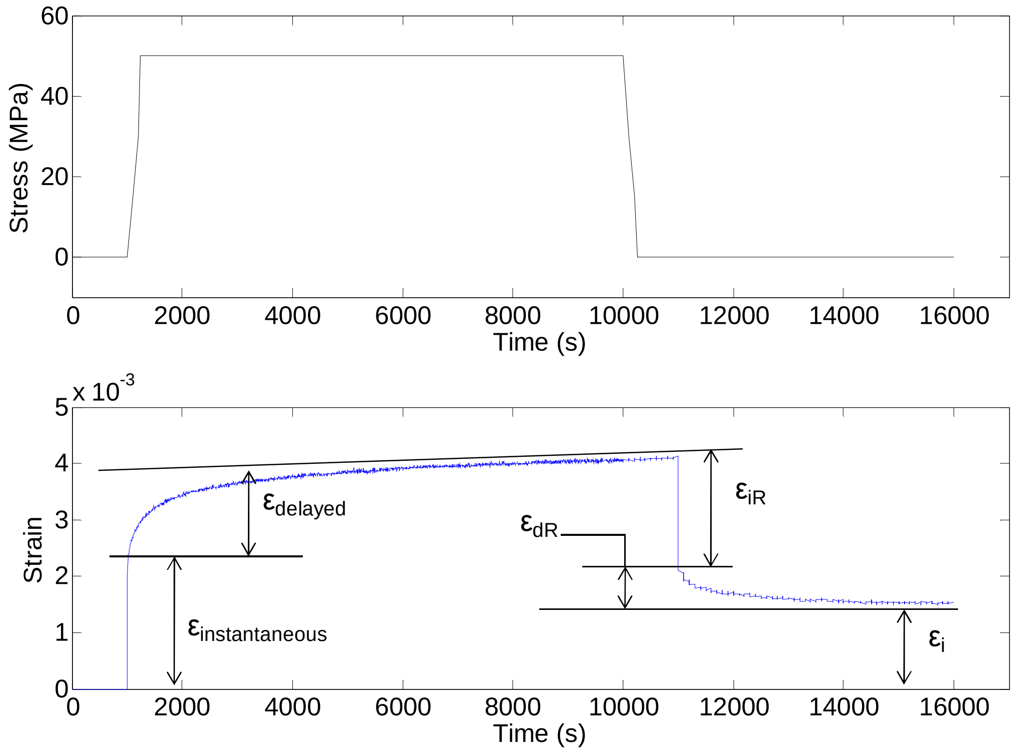

Figure 1: Creep-recovery test on a single hemp fibre. Evolution of tensile stress and strain as a function of time. T = 25°C±1.5°C, RH = 50%±1.5%.

For most of the viscoelastic materials, the primary creep is considered as the time-dependent recoverable portion of the delayed deformation, the secondary creep the portion of the total sample deformation which is nonrecoverable at the test conditions after removal of the load. For hemp fibre, as a consequence of the lock-in phenomenon, a portion of the primary creep is also non recoverable. Whatever its physical or microstructural origin, these time-dependent phenomena with large permanent set could affect the dimensional stability and other performance characteristics of natural fibres reinforced composites, and has to be taken into account.

The interfibre variation in creep behaviour is quite large. So, to quantify the influence of the RH level on creep behaviour, we preferred in this study working on a same fibre instead of using a statistical approach. Previous works performed in our team [3,4,23] show that a significant change in mechanical properties, in particular in rigidity, and also permanent strains are induced by successive tensile loadings or moisture variations under tensile stress. Hence, to evaluate the influence of RH on both instantaneous and delayed response of the fibre, recovery periods have to be intercalated between creep stages. Series of creep/recovery tests were made successively on a same fibre at different humidity levels. Fig. 2 shows, for instance, the evolution of the strain as a function of time and RH level. The delayed deformation reaches its minimum value at 60% RH, and increases when the RH is decreased or increased with respect to this value. This is consistent with the hypothesis of existence of a threshold RH value corresponding to the saturation of the hydroxyl groups in the fibre [4].

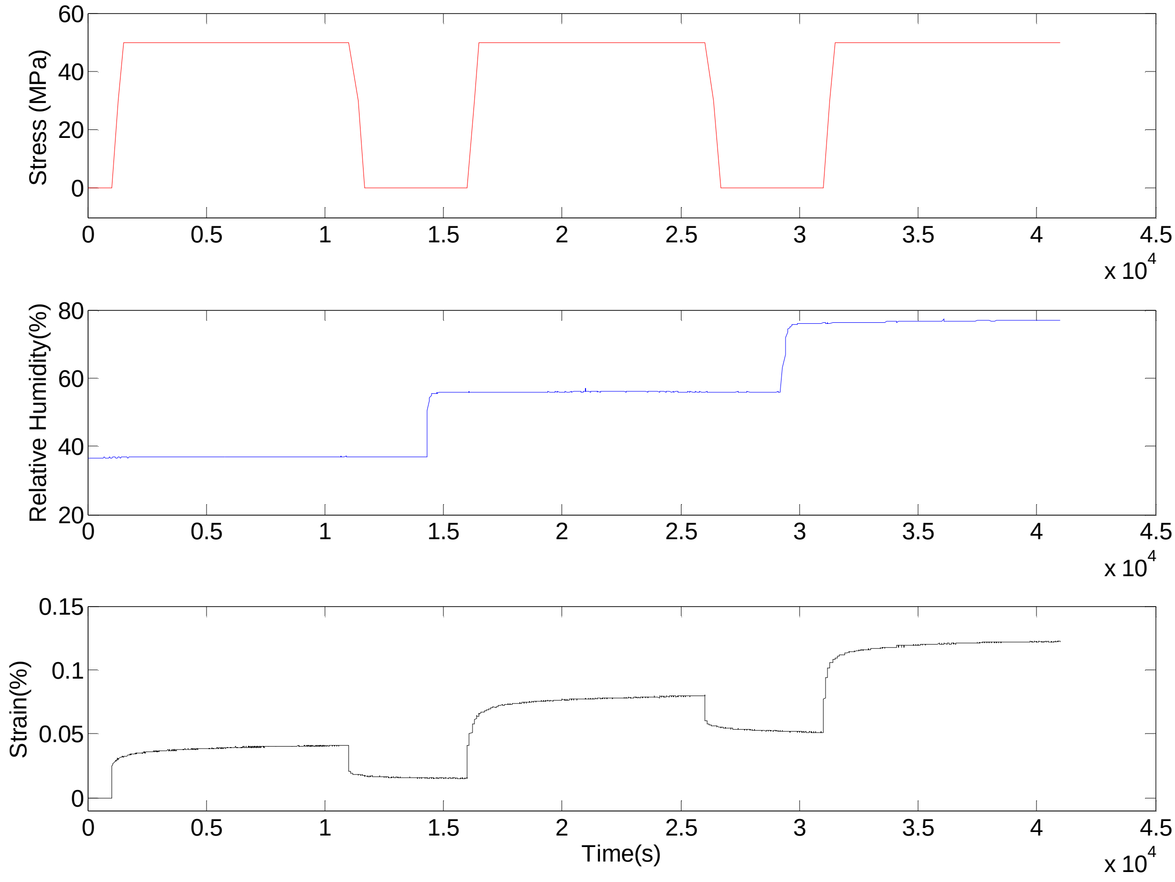

Figure 2: Successive creep-recovery tests on a single hemp fibre at different RH levels. Evolution of tensile stress, RH and strain as a function of time. T = 25°C±1.5°C.

Creep under cyclic humidity conditions: mechanosorptive effect

Fig.3 shows the evolution of the creep strain of a same elementary fibre as a function of time at constant humidity level (75%) and under cyclic humidity between 15% and 75% with different rate in humidity variation. The results confirm the observation of Haberger *et al*. [16], i.e. that the time parameters of the humidity cycling must be matched to the sorption time of the sample. For low rates, this match is not respected, the moisture gradient in the fibre wall is certainly weak and the creep is not accelerated. This result can clearly explains that accelerated creep was not observed in the past for cellulosic fibres.

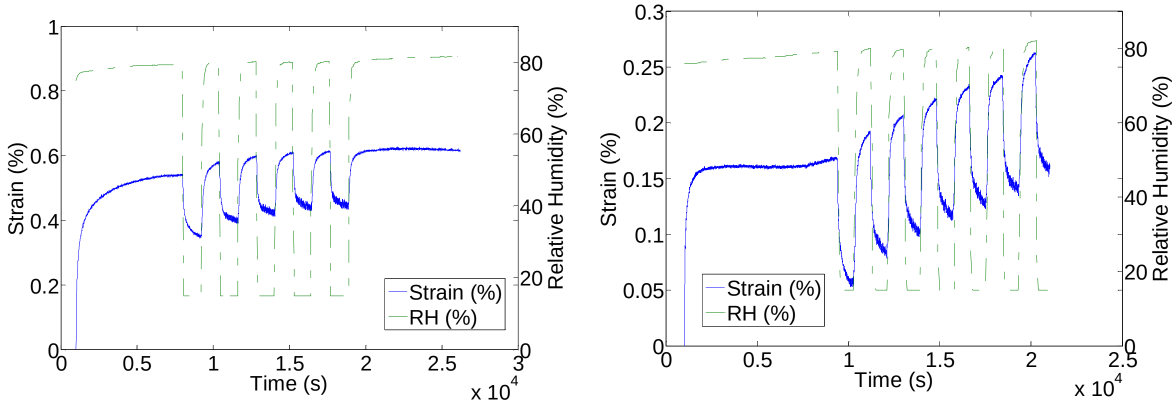

Figure 3: Evolution of creep strain of single hemp fibres as a function of logarithmic time for varying RH between 15% and 85%. Left: RH variation rate = 8%RH/min. Right: RH variation rate = 20%RH/min. Constant Tensile Stress = 50 MPa, T = 25°C±1.5°C.

CONCLUSION

Single hemp fibres exhibit significant delayed deformation when submitted to tensile load. When the fibres are unloaded, large permanent deformations are observed. This unrecoverable deformation origins from instantaneous and time-dependent mechanisms since only a portion of both instantaneous and delayed deformations is recovered.

Creep of single hemp fibres is also influenced by environmental relative humidity. The delayed deformation reaches it minimum value at 60% RH, and increases when the RH is decreased or increased with respect to this value. This is consistent with the hypothesis of existence of a threshold RH value corresponding to the saturation of the hydroxyl groups in the fibre, a decrease of the free volume and of the macromolecule mobility in the amorphous components.

Creep rate under cyclic humidity conditions exceeds any constant creep rate within the cycling range if the rate of RH variation overpasses 10 % RH/min for hemp fibres of about 30 µm in diameter and very small lumen. If the question of the origin of this mechanosorptive remains opened, our result seems to be consistent with sorption-induced stress-gradient explanations proposed for wood fibres.

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