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Swelling of natural fibre bundles under hygro- and hydrothermal conditions: Determination of hydric expansion coefficients by automated laser scanning

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A B S T R A C T

The effect of humidity conditions on the moisture content and dimensional variations of natural fibre bundles from several botanical origins with contrasting biochemical and structural characteristics is investigated. Results highlight wide variations in water uptake and swelling behaviour of fibre bundles according to plant species. Two main swelling mechanisms are identified: (i) a microscopic swelling due to the sorption of *bound* water in the cell walls and the middle lamella, and (ii) a macroscopic swelling related to the formation of *free* water in pores and lumens, which induces anisotropic deformation of bast fibre bundles cross section. Cross-sectional hygro- and hydroexpansion coefficients are determined and studied in relation with the structural features of plant fibre bundles. These results constitute key data for the predictive modelling of “in-service” mechanical behaviour of biocomposites.

Keywords:

A. natural fibres

B. Fibre deformation

D. Moisture

1. Introduction

Composite materials are subjected to various environmental stresses such as humidity, temperature or UV radiation. This exposure to environmental conditions is one of the primary causes of their accelerated ageing. These ageing processes require special attention in order to evaluate the evolutionary behaviour of composite materials in service-life, and particularly those of biocomposites reinforced with natural fibres [1]. Indeed, the dimensional stability and the mechanical behaviour of natural fibres are strongly influenced by hygro- and hydrothermal conditions [2], related to relative humidity (RH) and immersion conditions, respectively.

Depending on moisture content, natural fibres may contain water in two states: (i) the water bound to the different biopolymers constituting the cell walls and the middle lamella, primarily via the formation of hydrogen bonds with hydroxyl groups –OH and (ii) the *free* water which is in the micro- and macropores of cell walls, filling the voids (lumens) and retained by capillary forces [3–6]. Fig. 1 schematically shows the swelling processes occurring in wood cells according to moisture content. As the relative humidity increases, water vapour penetrates and is absorbed into the cell walls and the middle lamella via

the formation of hydrogen bonds, thereby inducing significant cell swelling. At a certain moisture content in the cells, which depends on the plant species considered, the *bound* water saturates the cell walls and the middle lamella: the water saturation point of the plant cells is then reached, ~20–40% for wood cells [7]. Finally, cellular cavities (lumens and porosities) are filled by the *free* water until total saturation.

Several measurement techniques are used to quantify moisture content in natural fibres as a function of relative humidity. In particular, (i) DVS (Dynamic Vapour Sorption) which is a gravimetric technique that measures the amount of water absorbed by a sample as a function of relative humidity at equilibrium (or water activity $a_w = P/P_0$ with P the water vapour pressure at the surface of the considered substrate and P_0 the saturation vapour pressure of water). Several authors have used this technique to determine water sorption isotherms from a dry sample, or desorption isotherms from a water-saturated sample [8–10]; (ii) WRV (Water Retention Value) which is a centrifugation and weighing method developed for wood pulps to determine their water retention value after immersion. WRV is determined according to ISO 23714 standard [11].

The processes of water sorption and swelling of natural fibres are complex because of their biochemical, structural and morphological

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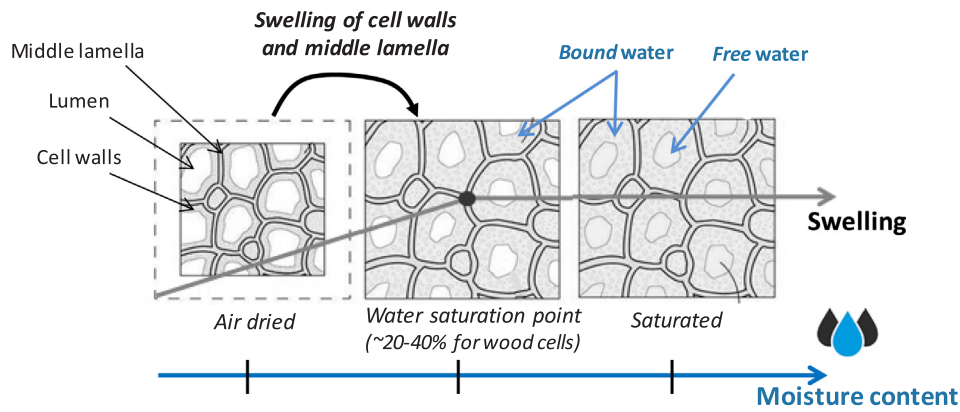


Fig. 1. Swelling and water state according to moisture content in wood cells (adapted from [12]). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

features. In this regard, these processes are influenced by several factors, such as the lumen size, the microfibrillar angle (MFA) [13], cellulose crystallinity, accessibility to hydroxyl groups, amorphous polysaccharides/biopolymers amounts and their relative hydrophilic/hydrophobic character. Moreover, swelling, water organization and its interactions with cell walls biopolymers are highly heterogeneous when considering the different scales of natural fibres structures. In the case of (ligno-)cellulosic substrates, a high hysteresis between the sorption and desorption isotherms is usually observed. The origins of this phenomenon are still debated in literature and although there is still no consensus, several authors attribute it to softening and relaxation processes related to water interactions with amorphous biopolymers present in the cell walls [14,15] and their glass transition temperature. Based on molecular modelling of water molecules interactions with crystalline cellulose - hemicelluloses - lignin models, Charlier and Mazeau [16] and Kulasinski et al. [17] showed that water mobility and its organization are heterogeneous at the microscale within cell wall structures, e.g. higher density of sorbed water molecules is found at cellulose/hemicellulose interfaces [17]. At the macroscale, Karmaker et al. [18] studied the swelling of jute fibre bundles cross-sections in water. Dry jute fibre bundles were compared to the same fibre bundles immersed for two weeks in water. Optical microscopy observations showed that cross-sectional swelling was heterogeneous along the fibre bundles and that maximum swelling occurred in the regions where the transverse dimensions of the fibre bundles were the smallest. According to the authors, in these thinner areas, the elementary fibres would be more packed within the fibre bundle so that during water sorption, the swelling of each elementary fibre causes a larger increase in diameter in these areas.

All these studies highlight the interest of accurately measuring the swelling of natural fibres in relation with their moisture content and structural features. These data are essential to better describe and understand the swelling processes of natural fibres that directly influence the processing and the in-service life behaviour of biocomposites. In particular, it should be pointed out that the swelling of natural fibres can induce modification of the stress state at the fibre/matrix interface in biocomposites, and contribute to an improvement in interfacial adhesion [19,20], but also generate stress levels that can locally damage the matrix, and thus cause an irreversible degradation of the composite [21].

The aim of this work is to characterize, under variable and controlled hygro- and hydrothermal conditions (i.e. 20%, 50%, 73% RH and immersion, respectively), the moisture content and the dimensional variations of fibre bundles from several botanical origins with contrasting biochemical, structural and morphological characteristics. An automated laser scanning technique [22], conducted either in a climate chamber or in an immersion cell, is used to evaluate the variations in the cross-sectional dimensions of the fibre bundles in relation with their

moisture content determined by DVS and WRV measurements. Swelling processes are discussed, and hygro- and hydroexpansion coefficients are determined and studied in relation with the specific structural features of the different plant fibre bundles.

2. Experimental

2.1. Selection of fibre bundles

Fibre bundles were extracted from various phylogenetic plant species with distinct functions in planta. These fibre bundles have contrasting morphometric characteristics as described in Garat et al. [22], microstructures and biochemical compositions [23]. Three of them belong to eudicots and are commonly called 'bast fibres'. These fibre bundles are weakly lignified (< 5%), have a low MFA (< 10°) and a support function of the stem in planta: flax *Linum usitatissimum* (Alizée, Grandvilliers, Picardie, 2012), hemp *Cannabis sativa* (Fedora 17, Champagne Ardenne, 2012) as annual fibre crop (after retting) and nettle *Urtica dioica* (2014, Lorraine) as a perennial herbaceous plant. The other two species are from perennial monocots. These fibres are highly lignified (> 9%) and have an higher MFA (> 20°): sisal *Agave sisalana* (FRD, Fibre Recherche Développement®, Troyes) coming from the leaf of agave and palm tree *Phoenix dactylifera* (Al-Ahsa, Saudi Arabia) present in the form of mats of foliar sheath surrounding palm tree stems. The biochemical compositions and average MFA of these different batches of fibre bundles are given in Table 2.

2.2. Characterization of fibre bundles under controlled hygro- and hydrothermal conditions

2.2.1. Determination of moisture content

The determination of the water sorption isotherms and moisture contents at given RH for the different plant fibre bundles were carried out by Dynamic Vapor Sorption (DVS) analysis. These tests were conducted over a relative humidity range of 0 to 90% RH, at RH intervals of 10% with a sample quantity of about 5 mg.

A centrifugation method adapted from ISO 23714 [11] was used to evaluate the Water Retention Value (WRV). Batches of 0.3 g of fibre bundles were immersed in deionized water for 5 min at 23 °C and then placed in tubes to be centrifuged at 3000 g for a period of 30 min, and finally weighed to determine the mass M_f (in g) of fibres. WRV values were determined according to Eq. (1) after drying for 10 min at 105 °C in a Sartorius-MA160 infrared moisture analyser (Grosseron SAS, Coueron, France) and weighing of the anhydrous mass of fibres M_s (in g).

$$WRV (\%) = \frac{M_f - M_s}{M_s} \times 100 \quad (1)$$

2.2.2. Dimensional measurements

A fibre dimensional analysis system (FDAS, Diastron Ltd, Hampshire, UK) was used to measure the cross-sectional dimensions of fibre bundles with a nominal length of 30 mm and typical diameters ranging from 50 to 250 μm at 50% RH (see also in Ref. [22]), and thus estimate their cross-sectional area (CSA) that is generally variable along plant fibre bundles. This experimental method based on automated laser scanning of the object, as well as the sample preparation protocol, have been described in detail in a previous study [22]. During the measurement, the fibre bundle sample is put under slight tension to maintain it straight, and then translated and rotated, so as to collect up to 600 values of apparent diameter per revolution for each cross-section. Each analysed cross-section were separated by a longitudinal pitch of 560 μm . In the end, about 25 000 values of apparent diameters are collected for each fibre bundles allowing to take into account the irregularity of the fibre bundles along their length and the non-circularity of their cross-section. As detailed in Garat et al. [22], an elliptical model of the fibre bundle cross-sections using statistical data processing of apparent diameters was applied in order to estimate their median CSA.

In order to analyse the influence of relative humidity on the cross-sectional dimensions of the fibre bundles, the dimensional analysis apparatus (FDAS) was placed in a climatic chamber supplied by ETS (Electro-Tech Systems Inc, Pennsylvania, USA) to control the temperature at 23 °C and relative humidity at 20, 50 and 73%. An immersion cell (DSM, Dynamic Swelling Module, Diastron Ltd., Hampshire, United Kingdom) was implemented on the FDAS device to measure the cross-sectional dimensions of the fibre bundles immersed in deionized water at 23 °C. It should be noted that under immersion conditions, the device allows the dimensional analysis of the sample for only one cross-section at a time. For each plant species and each hygro/hydrothermal condition, up to 20 fibre bundles were characterized after being acclimatized 13 h at 23 °C and at the studied RH (i.e. 20, 50, 73%) or at 50% RH before immersion.

3. Results and discussion

3.1. Influence of hygro/hydrothermal conditions on moisture content

The equilibrium moisture content of the different plant fibre bundles as a function of the RH level was quantified from the sorption curves obtained by DVS analysis. Whatever the plant species considered, the shape of the curves is identical and of sigmoid type (type II); it results from the combination of different sorption mechanisms. Indeed, the sorption isotherms can be divided into three zones [10,24,25]. In the first zone related to Langmuir's mode (RH < 10%), the water is mainly 'chemically' absorbed and bound via hydrogen bonds to the hydroxyls of amorphous cellulose and hemicelluloses and the carboxyls of pectins, present in the cell walls and the middle lamella. In the second zone that can be described by Henry's law (10% < RH < 65%), the water is rather 'physically' sorbed within the porous microstructure of the fibres where the water can freely diffuse. Finally, beyond 65% RH, water concentration is high which involves the formation of *free* water loosely bonds in the macropores and lumens until water saturation of fibre bundles.

Fig. 2 shows the evolution of the moisture content of fibre bundles as a function of relative humidity (DVS measurements) and in immersion (WRV measurements). Between 0 and 50% RH, the moisture content of the various plant species is low and similar, i.e. $5.3 \pm 0.4\%$ on average. Between 50 and 73% RH, there is an increase in moisture content up to about $13.7 \pm 1.7\%$ regardless of the plant species considered. On the other hand, in the vicinity of the saturation point of the cell walls (moisture content of about 20–40% as determined for wood cells) and under immersion conditions (formation of *free* water in the pores and lumens), the moisture content increases considerably and differences are observed depending on the plant fibre species. The palm fibre bundles have the lowest moisture contents in immersion,

$48.8 \pm 2.0\%$, followed by sisal, flax and hemp fibre bundles with moisture contents of $60.4 \pm 1.0\%$, $60.9 \pm 0.7\%$ and $62.8 \pm 0.7\%$, respectively. It is noted that nettle fibre bundles are highly hygroscopic, with moisture content in immersion of $112.2 \pm 1.2\%$, almost twice as much as flax and hemp fibre bundles.

Variations in moisture contents of the different fibre bundles in immersion conditions should primarily come from morphological differences, especially the amount and size of open pores and lumens. The contrasting biochemical composition between the plant species considered may also induce substantial differences in water sorption of the cell walls under controlled hygrothermal conditions. Pejic et al. [26] studied the influence of non-cellulosic biopolymers such as lignin on the water sorption of hemp fibre bundles, and showed that the decrease in lignin content tends to increase the water retention capacity of hemp fibres. Fig. 2 shows that the palm fibre bundles having the highest lignin contents (Table 2) also have low moisture content in overall. In contrast, the very high moisture content of nettle fibre bundles above 80% RH compared to other bast fibres could also originate from a poor retting inducing high pectin content and stronger hygroscopic character (Table 2).

Moisture data obtained from DVS and WRV measurements will now be used to analyse the morphological and dimensional variations of the fibre bundles under the different hygro- and hydrothermal conditions according to their moisture content.

3.2. Morphological and dimensional variations as a function of moisture content

3.2.1. Morphological variation: Evolution of the cross-sectional shape factor

Fig. 3 shows the evolution of the median cross-sectional shape factor (α), i.e. the median value calculated from the ratios *maximum diameter/minimum diameter* of each cross-section of the fibre bundle considered, for the different plant fibre bundles and according to the humidity conditions. The cross-sectional shape factor is a morphometric indicator of the non-circularity of fibre bundles cross-section [22], and its evolution is studied here according to the moisture content in each type of fibre bundles. In the case of palm and sisal fibre bundles, moisture content has little influence on shape factors that remain close to 1, i.e. these fibre bundles retain substantially circular shaped cross-sections upon swelling. Indeed, their median shape factors remain similar, about 1.20 ± 0.06 and 1.29 ± 0.14 respectively for palm and sisal, over the entire range of moisture content and related humidity conditions (from 20% RH to immersion conditions). It can therefore be concluded that their cross-sectional swelling is isotropic.

On the other hand, bast fibre bundles have higher and more dispersed median cross-sectional shape factors, which are influenced by the moisture content, in particular in immersion conditions. The cross-sectional swelling of these fibre bundles is anisotropic because it is characterized by an increase in the minimum diameter much greater than that of the maximum diameter, leading to a significant decrease in their cross-sectional shape factor under immersion conditions (corresponding to moisture content > 60% and hence much greater than the saturation point of the cell walls, in the domain where *free* water is formed). Indeed, the increase in the minimum diameter for flax and hemp fibre bundles is about twice that of the maximum diameter, and up to three times higher for nettle fibre bundles. The hemp, flax and nettle fibre bundles thus have median cross-sectional shape factors in immersion of 1.82 ± 0.26 , 1.92 ± 0.42 and 1.98 ± 0.69 respectively, which implies an increase in their circularity under these humidity conditions.

This result is contrasting with macroscopic wood specimens for which *bound* water plays a predominant role in their dimensional changes. Within a stem, the elementary fibres that grow on the periphery of the central lacuna, between the cortical parenchyma and the phloem/xylem, are grouped into packed fibre bundles. Therefore, we assume that this mode of intrusive growth of elementary fibres [27], as

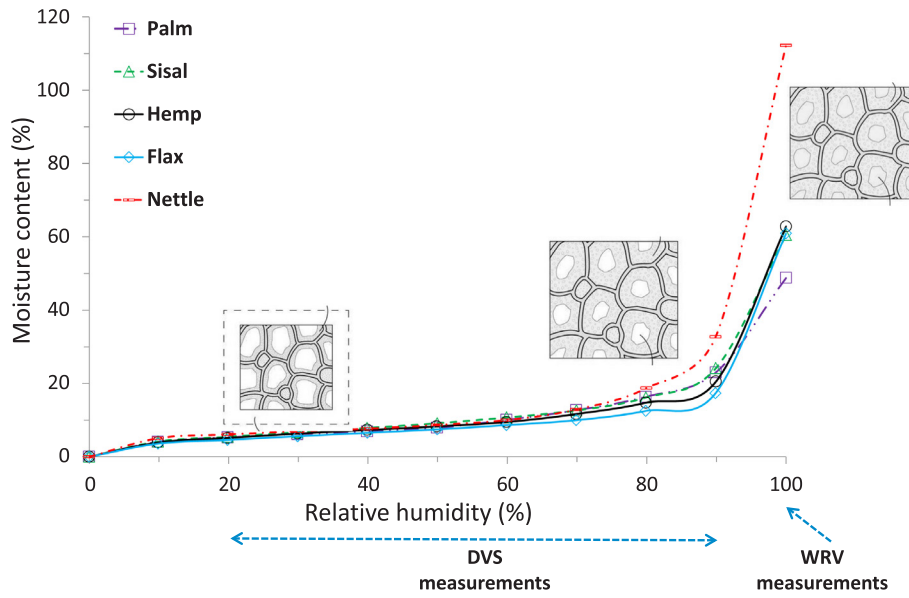


Fig. 2. Moisture content of fibre bundles in relative humidity (DVS) and in immersion (WRV) conditions at 23 °C for the different plant species. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

well as the progressive drying of the cells at the end of growth, after harvesting and cutting of the stems or leaves, cause or at least contribute to the anisotropic swelling phenomenon observed during rehydration of bast fibre bundles in immersion conditions. Thus, the lower the thickness of the cell walls, the greater the flattening effect during desiccation and the greater will be the decrease in the shape factor of bast fibre bundles during rehydration. Consequently, the geometry of their cross-section tends towards a more circular shape.

3.2.2. Dimensional variation: Evolution of the median cross-sectional area (CSA) and swelling mechanisms

The influence of the moisture content on the dimensional variations of the fibre bundles, and in particular on their cross-sectional swelling, was analysed. The cross-sectional swelling G_s of a fibre bundle is defined as follows (Eq. (2)):

$$G_s(\%) = \frac{S_i - S_0}{S_0} \times 100 \quad (2)$$

where S_i is the median CSA of the fibre bundle considered for a given humidity condition (in μm^2) and S_0 its median CSA at 50% RH (in μm^2).

Fig. 4 shows the evolution of the median cross-sectional swelling of the different plant fibre bundles as a function of their moisture content. For moisture contents less than 5% (i.e. at 20% RH, see Fig. 2), a slight shrinkage is observed, ranging from $-1.2 \pm 0.1\%$ to $-3.3 \pm 1.0\%$ for the palm and sisal fibre bundles, respectively. The flax and hemp fibre bundles have shrinkage of $-2.8 \pm 0.6\%$ and $-3.0 \pm 1.8\%$, respectively. This phenomenon is more pronounced in the case of nettle fibre bundles, reaching $-6.9 \pm 2.9\%$. For moisture contents between 10.8% and 14.7% depending on the type of fibre bundles (i.e. at 73% RH, see Fig. 2), the swelling is relatively low for all plant species ($< 10\%$) with low dispersion. In this range of moisture contents

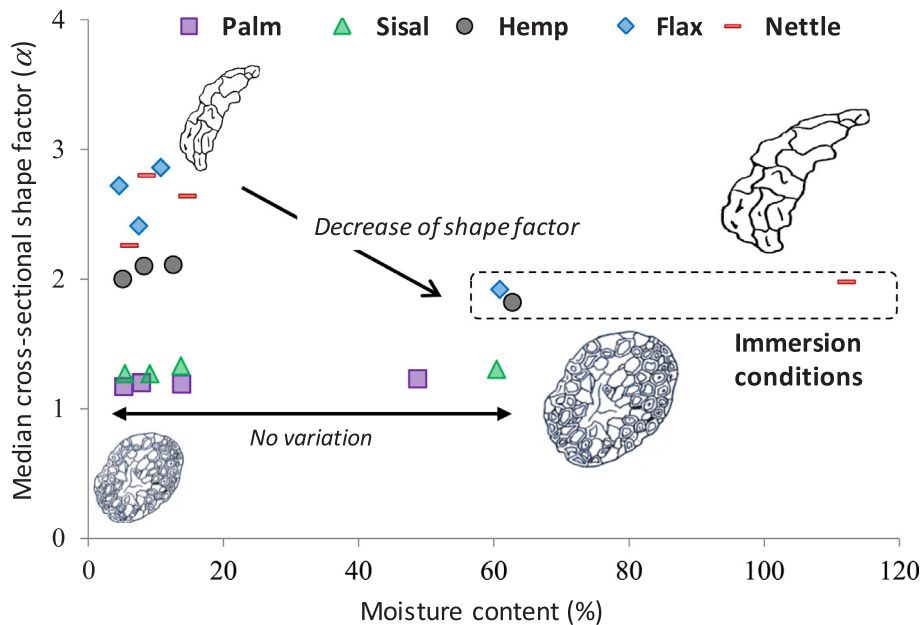


Fig. 3. Evolution of the median cross-sectional shape factor (α) of the fibre bundles as a function of moisture content at 23 °C for the different plant species. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

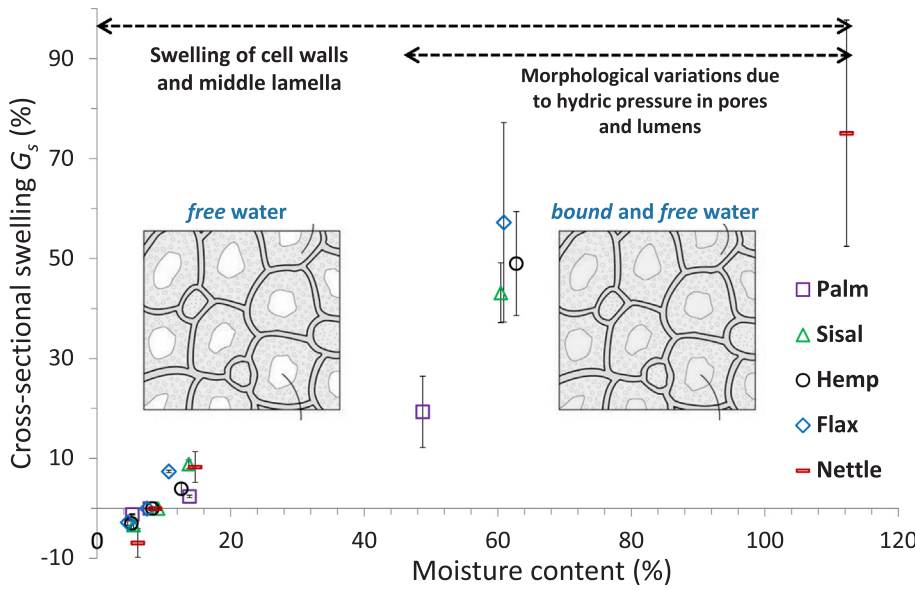


Fig. 4. Median cross-sectional swelling of the fibre bundles (normalized to the median CSA at 50% RH) as a function of moisture content at 23 °C for the different plant species. Proposal of two concomitant swelling mechanisms: (i) microscopic swelling of cell walls and middle lamella due to the sorption of *bound* water, (ii) macroscopic swelling related to anisotropic deformation of bast fibre bundles cross section, possibly induced by hydric pressure of *free* water in the pores and lumen. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

ranging from 5% to 15% (corresponding to relative humidity ranging from 20% RH to 73% RH), the swelling of fibre bundles is mainly associated with processes occurring at the level of cell walls and middle lamella, and related to the sorption of *bound* water by the biopolymers.

Under immersion conditions, the median cross-sectional swelling increases considerably and large differences are observed depending on the plant species. There is also greater dispersion on the swelling values. The palm fibre bundles had the lowest swelling, $19.3 \pm 7.1\%$, followed by the sisal fibre bundles with $43.2 \pm 5.9\%$. The hemp, flax and nettle fibre bundles showed greater swelling, $49.0 \pm 10.4\%$, $57.2 \pm 19.9\%$ and $75.1 \pm 22.6\%$, respectively. The anisotropic swelling observed in immersion for bast fibre bundles (flax, hemp and nettle) contributes to the increase of their cross-sectional area, and hence to the measured median cross-sectional swelling. A secondary process is implied here in the swelling of fibre bundles. Indeed, the formation of *free* water in large quantities in the lumens and porosities appears to contribute significantly to the cross-sectional swelling of fibre bundles, due to the morphological variations described above, and possibly induced by the hydric pressure exerted on the cell walls.

The swelling of the fibre bundles from the different plant species is therefore governed by two main processes (Fig. 4): (i) a microscopic swelling, at the scale of cell walls and middle lamella, due to the sorption of *bound* water, (ii) a macroscopic swelling related to the formation of *free* water in the pores and lumen (by capillary condensation at high RH or during immersion) which induces anisotropic deformation of bast fibre bundles and an additional increase in their cross-sectional swelling. Contrary to wood specimens, the water saturation point of the cell walls in natural fibre bundles is thus difficult to determine because, at high relative humidity between 80 and 99% RH, the swelling of the cell walls due to sorption of *bound* water and the morphological variations related to the formation of *free* water by capillary condensation processes are concomitant. Therefore, we rather assume a continuum of swelling processes occurring at the microscopic and macroscopic scales without a water saturation point of the cell walls being clearly defined. Experiments in an intermediate relative humidity range (i.e. between 80 and 99% RH) would nevertheless be interesting in order to complete these experimental data. It should be noted that the measuring devices used for this study cannot be used in this range of relative humidity because of the risks of water condensation which would damage the apparatus. A rise in temperature above 23 °C could allow limiting condensation and investigating higher relative humidity (potentially up to 90% RH). However, thermally induced phenomena, as polymer relaxation processes, would also have to

be considered.

3.3. Determination of hydric expansion coefficients and relation with structural features

The study of the morphological and dimensional variations of the fibre bundles as a function of their moisture content makes it possible to determine their hydric expansion coefficients, defined in this study as the cross-sectional swelling G_s (%) induced by a variation of 1% in moisture content Δm (%). Therefore, it is the surface expansion coefficients β_s of the fibre bundles cross-section that are determined.

Besides, we have previously shown that the swelling of fibre bundles results from two phenomena that strongly depend on hygro- and hydrothermal conditions. The swelling of the cell walls and the middle lamella due to sorption of *bound* water is the main process at stake under moderate hydrothermal conditions. In immersion conditions or when the relative humidity is sufficiently high to generate the formation of *free* water (by capillary condensation), significant morphological variations also contribute to the swelling of bast fibre bundles. It is therefore necessary to differentiate the cross-sectional hygroexpansion coefficient $\beta_{s,hygro}$ (Eq. (3)) and the cross-sectional hydroexpansion coefficient $\beta_{s,hydro}$ (Eq. (4)), defined as follows in this study:

$$\text{From 20 to 73\% RH } \beta_{s,hygro} = \frac{G_s(\%)}{\Delta m(\%)} \quad (3)$$

$$\text{In immersion conditions } \beta_{s,hydro} = \frac{G_s(\%)}{\Delta m(\%)} \quad (4)$$

Fig. 5 shows the cross-sectional hygroexpansion coefficients $\beta_{s,hygro}$ for the different plant fibre bundles, calculated between 20 and 73% RH at 23 °C. This coefficient varies from 0.42 to 1.70 per change in moisture content depending on the plant species. For comparison, Le Duigou et al. [19] determined a radial hygroexpansion coefficient for elementary flax fibres (from Flax Tape, Lineo®) of 1.14 per change in moisture content based on diameter measurements conducted in an Environmental Scanning Electron Microscope (ESEM, at 23 °C, steam pressure 5–10 mbar, from 20% to 98% RH). Assuming the isotropic radial expansion of an equivalent cylinder undergoing the same surface hygroexpansion coefficient β_s , a straightforward geometric calculation leads to Equation (5) which allows estimating an equivalent radial hygroexpansion coefficient β_r for flax fibre bundles in this study:

$$\beta_r = \sqrt{1 + \beta_s} - 1 \quad (5)$$

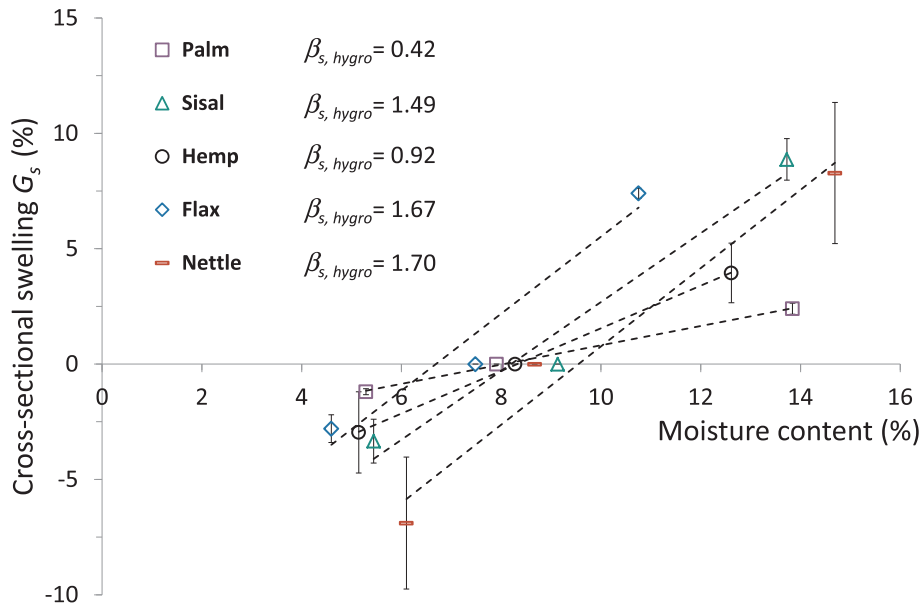


Fig. 5. Determination of cross-sectional hygroexpansion coefficients $\beta_{s,hygro}$ of fibre bundles measured between 20% and 73% RH at 23 °C for the different plant species. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The resulting β_r value is 0.63 for flax fibre bundles. This value is lower than that obtained by Le Duigou et al. [19] for an elementary flax fibre. The origin of the flax fibre may explain that difference, at least in part. Indeed one can hypothesize that the flax bundles and elementary flax fibres compared here are distinct in term of biochemistry and thermal history. In particular, the flax tape used in Le Duigou et al. [19] has been industrially modified, and could be more sensitive to moisture induced swelling due to structural degradation or variation in biochemical composition. Besides, the extent of swelling of a fibre bundle should be different from an elementary fibre. Indeed, the swelling of elementary fibres included within a bundle is more restricted due to intercellular cohesion with surrounding elementary fibres as compared to an isolated elementary fibre that can swell more freely. More investigations in that way would shed light on those differences.

Viguié et al. [28] assessed hygroexpansion coefficients of dense lignocellulosic fibrous networks (with folding board as case study). They found hygroexpansion coefficients of 0.13 and 0.098 in the cross direction (from 20% to 50% RH), at the macroscopic and mesoscopic scales (based on digital correlation of images obtained by X-ray microtomography) respectively. Joffre et al. [29] determined a local hygroexpansion coefficient (transverse to the microfibrils within the cell walls) for wood fibres (spruce, *Picea abies*) of 0.45 per change in moisture content (23 °C, from 47% to 80% RH) based on X-ray microtomography measurements (Synchrotron, Grenoble) and a numerical identification method.

Although all these hygroexpansion coefficients are of the same order of magnitude, these different results illustrate the importance of the measuring method and relative humidity range but also the dimensions and the nature of the object under consideration. Indeed, according to the scale (macro/meso/micro) and the type of plant fibres under study, the induced variations in microstructure and biochemical composition will have an influence on the value of the hygroexpansion coefficient. Note also the strong anisotropy of the microstructures encountered in plant fibres that generates very significant differences between the transverse and longitudinal hygroexpansion coefficients, the latter being very low especially for plant fibres with a low MFA (e.g. 0.037 for wood fibres in Joffre et al. [29]).

Similarly, the cross-sectional hydroexpansion coefficients $\beta_{s,hydro}$ were determined for the different plant fibre bundles and are reported in Table 1. In general, we found that these coefficients are lower than the cross-sectional hygroexpansion coefficients $\beta_{s,hygro}$, in particular for

Table 1

Cross-sectional hygroexpansion $\beta_{s,hygro}$ and hydroexpansion $\beta_{s,hydro}$ coefficients of fibre bundles at 23 °C for the different plant species.

| Plant species | $\beta_{s,hygro}$ | $\beta_{s,hydro}$ |
|---------------|-------------------|-------------------|
| Palm | 0.42 | 0.47 |
| Sisal | 1.49 | 0.84 |
| Hemp | 0.92 | 0.90 |
| Flax | 1.67 | 1.07 |
| Nettle | 1.70 | 0.72 |

fibre bundles which have high moisture content and/or wide morphological variations in immersion conditions, case of flax and nettle (Figs. 3 and 4). This indicates that sorption of bound water within the cell walls and the middle lamella plays a predominant role in the swelling of fibre bundles. It is also noticeable that, for phylogenetically distant species such as palm and hemp, the measured values of $\beta_{s,hygro}$ and $\beta_{s,hydro}$ expansion coefficients are similar.

Table 2 proposes a correlation (Pearson correlation coefficient, r) between the cross-sectional hygroexpansion coefficient $\beta_{s,hygro}$ and the available structural features of the cell walls of the plant fibre bundles, i.e. their biochemical composition and their MFA. This analysis shows a strong (negative) linear relationship of the $\beta_{s,hygro}$ coefficient with the lignin content and the MFA (i.e. Pearson coefficients r of -0.76 and -0.78 , respectively). Thus, high lignin contents and high MFA as those found in palm fibre bundles tend to limit the water retention capacity and reduce the cross-sectional swelling (Fig. 4), hence decreasing the cross-sectional hygroexpansion coefficient (0.42 for palm fibre bundles). Conversely, the low lignin content and low MFA found in flax fibre bundles are correlated with a higher cross-sectional hygroexpansion coefficient (1.67). Statistical approaches such as multivariate analyses on larger sets of data in terms of cell walls biochemical composition, MFA and integrating other descriptors such as the composition/structure of the middle lamellae, the size of the lumens... could help to better understand the contribution of these different microstructural parameters to the hygroexpansion of plant fibre bundles.

4. Conclusion

The effect of hygro- and hydrothermal conditions on moisture content and dimensional variations of different plant fibre bundles

Table 2

Correlation (Pearson coefficient, r) between cross-sectional hygroexpansion coefficients $\beta_{s,hygro}$ (20–73% HR; 23 °C) and composition in cellulosic and non-cellulosic polysaccharides (by HPLC analysis), lignin (Klason method) and MFA (average values taken from literature [23,30–32]) of the fibre bundles from the different plant species.

| Plant species | $\beta_{s, hygro}$ | Cellulose (%) | Hemicelluloses(%) | Others (pectins...) (%) | Lignin (%) | MFA (°) |
|-----------------------|--------------------|---------------|-------------------|-------------------------|------------|---------|
| Palm | 0.42 | 45.1 ± 3.4 | 27.7 ± 4.9 | 11 | 16.2 ± 0.3 | 44 |
| Sisal | 1.49 | 60.1 ± 1.2 | 23.3 ± 2.1 | 9 | 9.3 ± 0.1 | 20 |
| Hemp | 0.92 | 68.8 ± 1.3 | 9.8 ± 1.8 | 17 | 4.7 ± 0.3 | 9 |
| Flax | 1.67 | 64.9 ± 2.9 | 9.9 ± 3.9 | 21 | 3.7 ± 0.3 | 8 |
| Nettle | 1.70 | 51.7 ± 0.9 | 9.9 ± 1.2 | 34 | 4.6 ± 0.2 | 8 |
| Pearson Coef. (r) | | 0.36 | -0.58 | 0.56 | -0.76 | -0.78 |

(palm, sisal, hemp, flax, nettle), with contrasting biochemical and structural characteristics, was studied by DVS and WRV measurements and an automated laser scanning of cross-sectional dimensions conducted in a climate chamber (20%, 50%, 73% RH) and an immersion cell. The results evidenced different processes of water uptake and swelling according to the plant species and the hygro/hydrothermal conditions: (i) a microscopic swelling, at the scale of the cell walls and the middle lamella, due to the sorption of *bound* water and (ii) a macroscopic swelling related to the formation of *free* water in pores and lumens (by capillary condensation at high RH or during immersion) which induces anisotropic deformation of bast fibre bundles and contributes to their cross-sectional swelling. These results made it possible to determine the cross-sectional hygro- and hydroexpansion coefficients of the different plant fibre bundles studied, ranging from 0.42 to 1.70. A correlation between the cross-sectional hygroexpansion coefficient and some structural characteristics of the fibre bundles cell walls (biochemical composition and MFA) was found, thus demonstrating a dependence on lignin content and MFA. These results open interesting perspectives for the predictive modelling of “in-service” mechanical behaviour of biocomposites that would better take into account the swelling of natural fibres in relation with their structural features.

CRedit authorship contribution statement

William Garat: Methodology, Validation, Investigation, Writing - review & editing. **Nicolas Le Moigne:** Conceptualization, Methodology, Writing - original draft, Writing - review & editing, Visualization. **Stephane Corn:** Conceptualization, Methodology, Writing - review & editing. **Johnny Beaugrand ::** Conceptualization, Resources, Writing - review & editing. **Anne Bergeret:** Conceptualization, Supervision, Funding acquisition.

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