Moisture induced softening and swelling of natural cellulose fibres in composite applications

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Abstract. Composites based on natural cellulose fibres are susceptible to moisture. The fibres as well as the composites will inevitably soften and swell as moisture is absorbed. The intention of the present paper is to shed some light on the mechanisms behind softening and swelling. Also references to modelling work are made, to predict the moisture-induced dimensional stability. Characterisation techniques and models of such kind can be useful in choosing suitable fibres for improved moisture resistance, and identifying the main controlling parameters which affect the engineering consequences of moisture absorption. Understanding of the mechanisms and the main contributions to swelling can rationalise materials development. The examples shown in this review attempt to show the benefits by experimental mechanics and modelling in development of moisture resistant cellulose composites.

1. Introduction

1.1. Background
The use of natural cellulose fibres in composite materials has increased substantially during the last few decades. High-end applications with strict demands on mechanical performance typically advocate glass or carbon fibres. For high-volume applications, where low cost is more important than the mechanical performance, cellulose fibres can serve as an alternative to e.g. glass fibres. The main drawback of cellulose fibres is their affinity to moisture [1], which limits the outdoor applications, at least in climates which are not consistently dry [2]. In a short-time perspective, the moisture sensitivity of cellulose is caused by the abundance of accessible hydrogen bonds between hydroxyl end groups [3]. Of course, the moisture sensitivity can be reduced by chemical modification of the cellulose fibres, immobilizing and preventing the hydroxyl groups to adsorb moisture excessively [4, 5]. Any such chemical modification, is however going to lead to higher costs, and reducing the green features of this renewable and biodegradable raw material [6]. Many studies have focused on different strategies to chemically modify cellulose and characterising moisture absorption, swelling or reduction in tensile properties [7-9]. Such empirical studies are of course necessary in the quest to develop cost-effective and environmentally friendly alternatives to mitigate the disadvantages caused by the presence of moisture. In parallel to such empirical studies, it is also of interest to analyse the moisture absorption from a mechanics point of view. Suitable models can then be derived to link the moisture induced-deformation on the fibres to that of the composite [10]. The transfer of the stiffness drop due to moisture absorption from fibre to composite can also be studied [11, 12]. The purpose of the present article is to show how micromechanics can prove itself useful in linking effects of moisture on the
fibre level to the composite level. The main engineering properties in this regard is stiffness and dimensional stability. Swelling and elastic properties are amenable to micromechanical modelling, which is typically more complicated for e.g. strength properties, controlled by local inhomegeneities. The advantage of micromechanics and correlations with experiments is that simulations can be made how to constrain swelling and softening phenomena by e.g. changing the microstructure and composition of the composite. Such exercises can provide some advice in how cellulose composite with improved moisture resistance can be made. All models should, if possible, be based on observed physical mechanisms. Although strength is presently not the main focus, figure 1 below shows how experimental observations can be used to identify fibre pull-out and slippage at the fibre-matrix interface as key mechanism in the reduced strength of a wood-fibre composite.

Figure 1. Appearance of fracture in a dry or moist wood-fibre reinforced composite [13]: (a) tortuous crack growth around fibres in the dry composite, and (b) effortless pull-out of fibres from the matrix, indicative of a weakened fibre-matrix interface of moist composite.

1.2. Moisture transport

In regard of effects of moisture on cellulose composites, we will focus only softening and swelling within suitable working climates. For extended time, higher temperatures and consistently high ambient relative humidity, mould formation is a limiting factor for the use of cellulose-based materials. For these limiting cases, the reader is referred to other resources [14-15]. Furthermore, the transient regimes are not considered here. Only steady-state equilibrium conditions are taken into account. For relatively thin composite components and slow changes of the surrounding relatively humidity, the diffusions processes will be fast compared with the cycles of moisture content. In e.g packaging and furniture applications, the component dimensions and time scales are thought to be such that equilibrium conditions can be assumed for most part of the lifetime. For bulkier components or faster changes in surrounding climate, it is worthwhile to investigate the transport phenomena. If the moisture diffusion is slow, it is possible that there is not enough time for the material to absorb a sufficient amount of moisture to result in any noticeable degradation in stiffness or dimensional changes. Assuming Fickean diffusion, it is quite straightforward to characterise the diffusivity of composites with time-lapse weight measurements [16-18]. A particularity of composite diffusion is that diffusion is inherently anisotropic [19, 20], with higher in-plane diffusion rates where the fibres
(typically the most hydrophilic phase) are normally oriented [21]. In practice, diffusion is typically characterised only in the out-of-plane direction for relatively thin composites plates with sealed edges to promote essentially one-dimensional moisture transport. However, in composite components in service, only one-dimensional through-thickness diffusion cannot be assumed. It is likely that exposed fibrous edges can accelerate the rate of moisture uptake. In general, an impermeable coating and sealed edges, can slow down the moisture uptake and prevent undesired softening and swelling [22, 23].

1.3. Vapour sorption
Diffusion processes show non-linear behaviour in the sense that the diffusivity depends on the local moisture content [24] and the two-phase transport of free and bound moisture, respectively [25]. Even under equilibrium conditions, non-linear behaviour presents itself in terms of a non-linear relationship between ambient relative humidity and the moisture content of the material. An example for a wood species is shown in figure 2. Dynamic-vapour sorption isotherm curves (equilibrium moisture content vs. surrounding relative humidity) show non-linearity in particular for high levels of relative humidity, close to the fibre saturation point where a noticeable amount of free moisture precipitates in the voids of the material [26]. Another feature in the relative humidity-moisture content relation is that absorption and desorption curves are not identical. There is a limited but undeniable hysteresis during climate cycling due to dissipation processes in moisture transport [27, 28]. Nevertheless, in the present context of moderate climate conditions significantly below the saturation level, the sorption isotherm can be regarded as linear and the difference between sorption and desorption can be neglected in practical micromechanical models.

![Figure 2](image_url)

Figure 2. Non-linear sorption isotherm curve of wood measured by dynamical vapour sorption test, with equilibrium moisture content against relative humidity of the ambient atmosphere [29]. The measurements show a saturation point at about 80% relative humidity, and an absorption-desorption hysteresis.
1.4. Temperature effects
The physical climate affecting moisture content also includes the temperature. The temperature is known to markedly affect moisture content even though the relative humidity is kept constant [30]. Most studies on moisture effects of cellulose composites are carried out at room temperature, around 20-25°C, which is acceptable for composites intended for indoor applications. Essentially, there is a lack of experimental data for the temperature dependency of the properties of moist cellulose composites. This is especially the case of high processing temperatures, where extrapolation of a few published measurements may be necessary [31, 32]. In the present work, however, we consider working conditions at constant ambient temperatures, although the hygroelastic models could be generalised to include also effects of the temperature.

1.5. Relative humidity vs. moisture content
Moisture is typically either characterised in terms of relative humidity in surrounding the specimen under investigation or specifically the moisture content (relative moisture mass) in the specimen itself. The two measures are intimately linked, and the relationship is characterised by dynamic vapour sorption (DVS) isotherms. The relative humidity is easily characterised by hygrometers in the ambient air, whereas the moisture content necessitates weighing of sample and comparison with its dry weight. The relative humidity thus has the advantage of straightforward measurement, but provides only and indirect moisture-level characterisation. The moisture content, in contrast, offers a direct intrinsic measure of hygroscopic state of the material, although it is not always practical to quantify experimentally. In the literature, both relative humidity and moisture content are used to describe the state of the material. Since they can be linked by DVS measurements, micromechanical hygroelastic models based on the two different measures are mutually transferable, which applies also to the methods presented in this paper.

1.6. Natural cellulose fibre composites
Natural cellulose fibres in composites come in many forms. On an industrial scale, the largest class in terms of material is WPC (wood polymer composites). Strictly speaking, the reinforcing phase is constituted by wood particles acting as fillers rather than reinforcing fibres. WPC components are manufactured by extrusion or injection moulding with a polyolefin matrix, such as polypropylene or polyethylene. The limited aspect ratio of the wood particles, i.e. a length to width ratio of about 2-5, is a limiting factor in the mechanical properties of WPC, in particular regarding tensile strength [34]. In fact, for particulate WPC possessing relatively poor fibre-matrix interfaces, the composite strength shows a strong correlation with the interfacial strength, and composite strength can be used to rank the efficiency of fibre surface treatment for improved interfacial stress transfer [35]. For applications with higher demands on the mechanical performance, it is better to use high-aspect ratio reinforcements based on long plant fibres or unbroken wood fibres. When processed in by high-volume techniques, such as injection moulding, these slender fibres are however likely to degrade in dimensions, and eventually reach small aspect ratio like WPC particles with limited reinforcing capabilities [35]. Compression moulding for thermoplastic matrix composites [36] and resin-transfer moulding for thermoset matrix composites [37] are better options for retained fibre slenderness and improved mechanical properties. The fibre-length degradation caused by high shear flow along the screws in injection moulding machine or extruder [38] are then avoided. At the cost of processability, choosing longer fibres usually mean clear improvements in mechanical properties for load-carrying applications, such as stiffness, strength and dimensional stability. For each specific application, a cost-performance assessment must be made in order to select the fibre type and a suitable manufacturing technique. The environmental impact is in the balance pan of cost, which is an important driving force for development of cellulose-fibre composites. In other words, a transition from petroleum-based materials to renewable bio-based materials is afflicted with additional costs, at least during a transition period. From a mechanics point of view, the use of longer fibres from wood (forestry) or plants (agriculture) only has an influence on the geometry of the fibres and their mechanical properties. The
woody cells are shorter and have inferior stiffness and strength, due to a lower cellulose content and a
higher off-axis orientation of the cellulose microfibrils in the cell wall, as compared with plant fibres
from annual crops. A comparison of wood and plant fibres for composite applications has been
compiled by Madsen and Gamstedt [39].

1.7. Materials modification and design
From a micromechanical perspective, natural cellulose fibre composites are composed of a continuous
phase, i.e. a polymer matrix, in which discrete reinforcements are embedded, i.e. the cellulose fibres.
Moisture effects of such composites are mainly attributable to the fibre constituent, since the cellulose
fibres are usually more hygroscopic than the polymer matrix. In some specific examples, such as
cellulose-reinforced starch, the softening and swelling could stem from the very hydrophilic matrix
[13]. Composites based on unmodified starch matrix are however not a candidate for load-carrying
applications. For cellulose fibres in general, it is safe to assume that the moisture effect  is
predominantly caused by the moisture uptake by the fibres. This is also the case for most biopolymer
matrix materials, such as polylactide, which are still less hydrophilic than the fibres, at least in the
transverse direction [10]. Modifications to control or suppress moisture effects are therefore mostly
focused on the cellulose fibres. In contrast to man-made synthetic fibres, such as glass and carbon
fibres, the natural fibres are not inert and offer many chemical routes to modify their properties. The
fibres are also porous on the nanometre scale [40], which facilitate access of low-molecular weight
chemicals.

Focussing on the main component, i.e. the natural fibre, it can also be regarded as a composite
material on the cell-wall level [41]. Natural cellulose fibres from plants and wood are composed of
crystalline cellulose microfibrils embedded in an amorphous biopolymer matrix composed of
hemicellulose and lignin. On this ultrastructural level, the crystalline cellulose microfibrils are
relatively inert, whereas the hemicellulose-lignin matrix is the moisture-sensitive component.
Hemicellulose has more accessible hydrogen bonds and is less fatty than lignin, and is hence the more
hydrophilic of these two matrix biopolymers [42]. Pulp fibres can be cooked to remove the hydrophilic
biopolymers while retaining most of the crystalline cellulose, leading to higher moisture resistance.
This is the case for dissolving pulp consisting almost entirely of cellulose, which can be processed to
form highly consolidated stiff and moisture-resistant composites [43]. Chemical cross-linking of
remaining hemicellulose improves the moisture resistance by creating a tighter molecular network.
Although the hydroxyl groups are active functionalities in such cross-linking, there is such an
abundance of accessible hydrogen bonds that far from all groups are used in for cross-links. The
hemicellulose still remains hydrophilic, but to a lesser extent than the unmodified material [44].

From a practical engineering top-down approach, one of the first issues to address is which fibre to
choose with regard to dimensional stability and softening of the composite. The micromechanics of
the cell wall, its ultrastructure and targeted chemical modification schemes are then not of primary
concern. Presently, we will only address natural cellulose fibres and composites containing such fibres
with regard to their hygroelastic behaviour. Models and experiments on this length scale could
potentially be used in materials development in ranking different fibre candidates [12], and provide
improvements in experimental design [45].

2. Moisture-induced softening

2.1. Driving forces
So-called green composites, i.e. composite materials at least partially made from renewable resources,
contain almost always lignocellulosic biopolymers, typically from annual plants or wood. These have
been processed and treated chemically, at least to some extent. Their ‘eco-friendliness’ may thus vary,
although they should be better than synthetic composites from petrochemical resources regarding
carbon dioxide emissions from a total life-cycle perspective. The lignocellulosic biopolymers are rich
in hydroxyl groups, which form hydrogen bonds as one of the most important secondary bonds along
the polymer chains. Apart from the interior of the cellulose crystallites, these hydrogen bonds are relatively accessible to water molecules which can diffuse into the biopolymer. Absorption of water molecules at the hydrogen sites will lead to weakening of the strong hydrogen bonds, and macroscopically a lower stiffness. Decrease of stiffness, typically characterised by a Young’s modulus measured in a tensile test, is known as moisture-induced softening, and is generally an undesired property of load carrying cellulose-fibre composites. Since the fibre is moisture sensitive constituent compared with the stress-transferring matrix, the softening of the fibres is of interest, and how the softened fibres contribute to the softened stiffness of the overall composite. In general, if the stiffness of fibres in a moist state of interest is known, then the least softened fibre type can be identified. It is also this fibre type that will lead to the least softened composite. Techniques to measure stiffness of fibres, in try and moist states, are therefore useful. The minuteness and variability of natural cellulose fibres make it cumbersome and time-consuming to characterise individual fibres [46], and many fibres must be tested to gain a statistically representative average.

2.2. Softening of fibres

Stiffness testing of fibres under moist conditions is essentially the same as under standard ambient condition. The only difference is that the climate is controlled in the room where the test is carried out, or that the test rig is mounted in a climate chamber with a controlled relative humidity. The fibres should be in equilibrium state, i.e. that all transients of moisture transport have vanished and the fibres are in a steady state with respect to their moisture content. The small dimensions of the natural fibres usually mean that equilibrium state is attained in a short period of time, comparable or even shorter than the preparation and mounting time before testing. However, to be confident that this is the case, it is recommended to weight the fibres, normally a larger fibre mass to reach a practical total weight, and check that the weight remains constant over time in the targeted climate.

Direct testing of single fibres would give the best actual value of the fibre stiffness along the axial direction. In addition to required dexterity, patience and sensitive load cells and deformation measurements, there is also a challenging large variation in stiffness which can be explained natural variability in fibre geometry and the orientation of the microfibrillar angle, i.e. the orientation of the aligned cellulose crystallites in the main cell wall layer with regard to the fibre axis [47]. The properties of the main cell-wall layer, the S2 layer, can be regarded as a material property, at least relatively speaking as compared with the properties of the fibre itself. However, for practical purposes the engineering stiffness on the fibre level is more suitable. In contrast to e.g. glass fibres, natural cellulose fibres are significantly anisotropic, with considerably higher compliance transverse to the axial direction of the composite. Recognising the fibre anisotropy is important in composites, where the fibres are typically multidirectional. In fact, the transverse properties have a higher impact on the stiffness of the in-plane stiffness of a randomly oriented composite, as predicted by the Tsai-Pagano relation [48]. Furthermore, it can be shown that the shear properties of the fibres have considerable influence on the macroscopic in-plane stiffness of a randomly-oriented fibre material [49]. The simplest description of the elastic anisotropy of natural fibres would be transversely isotropic elasticity, quantified by 5 elastic constants, of which 4 are independent. The major and minor Poisson ratios are assumed to have less influence on the overall composite stiffness than the Young’s moduli in the axial and transverse direction, and the shear modulus. It is only the axial Young’s modulus that is relatively straightforward to characterise experimentally. Estimation of the transverse and shear modulus usually requires an indirect test method since it is not directly feasible to apply pure shear loading or transverse loading to single fibres. In practice, the off-axis elastic parameters, i.e. all but the axial Young’s modulus, are estimated by assuming a fixed ratio to the axial stiffness [41]. These assumptions should, if possible, be based on physical reasoning, related experimental results or micromechanical modelling. This illustrates that there is a lack of reliable experimental data on the properties of the cellulose fibres.

An alternative method to single fibre tensile test, is to characterise the longitudinal fibre stiffness by compression of fibre mats, and back-calculate the fibre stiffness. The fibre network can be
interpreted an in-plane assembly of Bernoulli-Euler beams, which are deformed between contact points of adjacent fibres. The average distance between contact points necessarily decreases during elastic compaction. Toll and Månson [50] have developed a model which predicts the Young’s modulus of cylindrical glass fibres from compaction of in-plane randomly oriented fibre mats in the thickness direction. Their model can be modified to describe compaction of wood fibres, provided the contact points are sufficiently far away from one another (low-density fibre mats), and that the variation in second moment of area of are used in the expressions of the bending stiffness [51]. From load-displacement data of compression tests of fibre mats, the volume fraction of fibres $V_f$ in the mat can be determined with respect to the applied pressure $P$, if the grammage, initial thickness and cell-wall densities are given. For cylindrical fibres, it can be shown that the applied pressure can be expressed as [50]

$$P_c = \frac{512}{5\pi^2} E_{11} f^4 V_f^5$$

(1)

where $f$ is an integration of a weighted autocorrelation function of the orientation distribution. The dimensions of the fibre cross-section play an important role, since the fibre segments are bent between the contact points as the fibre assembly is compressed. Although the variation of cross-sectional geometry is relatively small, it can have a measurable impact on the compaction behaviour [51 52].

From compression test and the expression in equation (1) or statistical counterparts [51], the Young’s modulus of the fibre can be determined, and e.g. compared with results from the tensile tests briefly presented above. The stiffness of flax fibres has been estimated both by compaction of fibre mats and tensile testing of fibres [53], with comparable results. Fair agreement was obtained for the first compaction of a pristine fibre mat relatively low density and well-separated fibres.

Testing of moist fibres or fibre mats is more difficult than testing of dry fibres, given the softened state of the fibres. This can lead to difficulties in clamping the fibre ends for single fibre tensile tests. For the compaction test, the softened fibres may induce buckling of the hollow natural cellulose fibres [54]. Despite the difficulties to handle and test soft and moist fibres, the characterisation techniques are the same as for drier fibres.

2.3. Softening of composites

The stiffness of moist composites is measured in the same way as composites at normal ambient laboratory conditions. The stiffness of composites with moist softened composites can be predicted by micromechanical models and laminate analogy, similar to non-moistened composites [41]. Unless the matrix is an extremely hydrophilic biopolymer, the composite softening can be attributed directly to the softening of the cellulose fibres by a micromechanical model for a hypothetical unidirectional lamella [55] and laminate analogy to account for the in-plane fibre orientation distribution [41]. Empirical micromechanical models based on Cox’ network model can also be used to relate the microstructure, the fibre stiffness and that of the composite [56]. For the contribution of a unidirectional lamella, the simple rule of mixtures can be used for the longitudinal elastic properties. For transverse properties, the Halpin-Tsai model [57] or models from Hashin [58], can be used to estimate the stiffness parameters. From elastic properties of a unidirectional lamella, a laminate analogy can be used to scale up to the composite plate, as shown schematically in figure 3. Given the fibre orientation distribution for a short fibre composite or mat, the global stiffness matrix $[\mathbf{Q}]$ can be described as
\[
\mathcal{Q} = \frac{1}{\pi} \int_{-\pi/2}^{\pi/2} \mathcal{T}(\theta)^T \mathcal{Q} \mathcal{T}(\theta) p(\theta) \, d\theta
\]

where \(\mathcal{T}(\theta)\) is the transformation matrix for a rotation angle of \(\theta\) radians, and \([\mathcal{Q}]\) is the local stiffness matrix. It is assumed that \(p(\theta)\) is symmetric, which is typically the case for simple geometries like plates used to manufacture test coupons. This means that the material is globally orthotropic, and the stiffness matrix is described by five elastic constants, namely the longitudinal and transverse Young moduli, the major and minor Poisson ratios and the shear modulus. The components in the global stiffness matrix can be determined from standardised macroscopic testing, and the orientation distribution can readily be found by image analysis of scanned sections of the fibre mat [59] or image analysis of X-ray micro computed tomography scans [60]. From the stiffness matrix, the Young’s modulus of the composite can be found as a function of the elastic properties of the fibres. Such a model can be used both to investigate the effect of softened fibres on the stiffness of the composite, or conversely to identify the fibre stiffness based on macroscopic measurements of the composite stiffness, provided that the interrelation of the four independent transversely isotropic elastic properties of the fibre has been set [41, 61].

\[\text{Figure 3.} \quad \text{Schematic illustration of a laminate analogy for a short-fibre composite, i.e. with finite length natural fibre reinforcement. The relatively slender fibres have an in-plane random distribution given by the fibre orientation distribution } p(\theta) \text{, which defines the relative proportion of a small orientation interval in the laminate model.}\]

\[\text{It should be noted that it is not only the fibres that contribute to the softening of the composite, even if the matrix is relatively hydrophobic. The fibre-matrix interface can be susceptible to moisture uptake. If there the interfacial compatibility is limited, e.g. by a hydrophobic polymer matrix, like most polyolefins thermoplastics, the wetting of the fibres during manufacturing can be incomplete resulting in gaps between the reinforcement and the surrounding matrix. Capillary effects can then promote moisture uptake along the interfacial gaps. The high concentration of water can then promote breakage of hydrogen bonds at the interface, leading to a decrease in stiffness due to reduced fibre-matrix stress transfer [62]. The contribution of a weakened interface to a reduced stiffness is expected}\]
to be particularly important if fibre-matrix is susceptible to moisture adsorption. This is expected if the adhesion is poor, meaning capillary channels along the interfaces, and a hydrophilic composite in general, i.e. moisture is readily transported into the composite interfaces.

3. Moisture-induced swelling

3.1. Driving forces
Swelling is here defined as increase in dimensions as moisture is absorbed. The mechanisms are essentially the same as those for softening, i.e. increase of compliance as moisture is absorbed; water molecules diffuse into the material from the surroundings, and break tight hydrogen bonds, thus creating additional space between molecular chains of the lignocellulosic fibres. Therefore, swelling and softening go hand in hand, sometimes also quantitatively [61]. When the hygroexpansion coefficient is reduced by a material modification, the compliance of the material is typically reduced by approximately the same relative amount. Among the constituents of the cell wall of natural fibres, the cellulose crystallites do not swell, since their tight crystalline packing does not allow moisture uptake. Adsorption on the surface crystallites is possible, or in the amorphous or disordered cellulose regions interspacing the crystallites in the cellulose microfibrils. Lignin and in particular hemicellulose, on the other hand, have a large amount of accessible hydroxyl groups and breakable hydrogen bonds [42]. Any attempt to chemically modify the fibres to reduce the dimensional instability should be directed to these biopolymers. An example of cross-linking pulp fibres with butantetracarboxylic acid (BTCA) to increase the resistance to swelling is presented by Almgren et al. [10].

3.2. Swelling of fibres
Moisture-induced swelling is considerably larger in the radial or transverse direction compared with that of the axial direction. This is due to the microfibril direction which is relatively close to the axis of the fibre. Since the swelling is large in the hemicellulose and lignin matrix surrounding the relatively inert cellulose microfibrils, the swelling of the fibres will be predominantly in the transverse direction where the constraint effect of the stiff microfibrils is very limited. The swelling is normally quantified by the hygroexpansion coefficient which can be defined as the change in strain per change in moisture content, or alternatively per change in relative humidity in the surrounding medium at equilibrium conditions. As for fibre stiffness, the best way to obtain a direct measure of the fibre welling is to measure it directly on single fibres. This is however even more cumbersome for fibre swelling than fibre tensile testing, since the fibres generally twist and bend during moisture uptake due to the off-axis orientation of the microfibrils and the non-prismatic and irregular shape of the cross-section along the fibre. Joffre et al. [63] investigated the dimensional changes of a single native spruce fibre subjected to climate changes in relative humidity with X-ray micro computed tomography at high resolution in a synchrotron. The experimental results were compared with finite element simulations accounting for large deformations, with the hygroexpansion coefficients in the longitudinal and transverse directions as the only fitting parameters. The elastic properties needed to be corrected to account for the influence of the changing environment, since moist fibres are more compliant and more prone to deformation when subjected to hygroscopic internal stresses. This approach is schematically outlined in figure 4. The irregular and varying individual shape of natural fibres require that the detailed geometry of specific fibres need to be taken into in such calculations. Only compression softwood fibres [61] or plant fibres with a near prismatic geometry [64] allow analytical modelling. The example shown in figure 4 with a mixed numerical-experimental approach where a finite-element updating scheme was used to simulate the swelling of an earlywood spruce fibre going from the experimental fibre geometry at 47% relative humidity to the predicted geometry of the fibre in the moist state at 80% relative humidity at equilibrium conditions. The hygroexpansion coefficients were identified by comparing the predicted and the experimental three-dimensional fiber geometry in the wet state. The obtained values are 0.17 strain per change in relative humidity transverse to the
microfibrils in the cell wall, and 0.014 along the microfibrils. It should be noted that this value is for untreated fibres in the green state. Any further treatment like pulping, hornification from drying (leading to microfibril aggregation) or chemical processing would most likely result in reduced hygroexpansion, since the free swelling is restrained by further cross-linking in the cell wall. The hygroexpansion of the helical cell wall, although at a moderate microfibril angle of 5-10° for wood fibres and even less for most plant fibres, is not the same as that of the fibre. Considering the lumen and the tubular helix structure, the fibre swelling can be calculated from the cell-wall swelling [61].

![Figure 4](image_url)

**Figure 4.** Comparison of 3D geometries of swollen single fibres from experimental measurements (synchrotron X-ray computed tomography images) and from numerical simulations (finite elements). The fitting parameter and the hygroelastic properties of the fibre.

### 3.3. Swelling of composites

The swelling of the composite is mainly attributed to the swelling of the hydrophilic fibres. Since the fibres in the composite are generally embedded in a relatively hydrophobic matrix, the surrounding matrix should restrain the swelling of the fibres. A micromechanical model has been established to predict the swelling of embedded fibres based on experimentally characterised microstructural parameters and hygroelastic properties of the constituents [60], and the predicted swelling was found to be in concert with direct measurement of various wood-pulp fibre composites by means of three-dimensional X-ray microtomographic images. The internal stresses developed during a change of moisture content can be sufficiently large to induce damage or cracking in the material. This is particularly the case of the local fibre dispersion results in tensile stresses normal to weak fibre-matrix interfaces. Such a situation is however more likely in drying than in wetting.

The model based on classical laminate theory used by Almgren et al. [10] has been used to link the fibre swelling to the out-of-plane swelling of in-plane oriented composite plates. The swelling in the thickness direction is larger than the in-plane swelling of composites (see figure 5), since the fibres tend to orient themselves in the plane of manufactured plates. This seems to be the case for both injection moulded [60] and compression moulded composites [12, 59]. For unsymmetric fibre orientation distributions with regard to the composite mid-plane, not only swelling in the thickness direction, but also a significant plate bending can take place [59], which can also be related to the fibre swelling. In order to avoid such obvious shape deformation, a symmetric lay-up is recommended.
Out-of-plane swelling is typically larger than the in-plane swelling due to the predominantly in-plane fibre orientation.

4. Final remarks
It can be summarised that the inevitable moisture induced softening and swelling of natural cellulose fibre composites is a nuisance in most engineering applications. In parallel to the development of new processing routes and modification schemes to suppress these undesired effects, it is useful to gain better understanding of the underlying mechanisms and develop models that can predict the reduction in stiffness and dimensional instability. The intention of this review paper is to provide some examples of such attempts, using a variety of experimental techniques and numerical and analytical composite mechanics models. The success of a quantitative mechanics-based approach in facilitating and rationalising the materials development process for improved moisture resistance lies in integrating the chemical/processing approach with methods derived from experimental mechanics. Multidisciplinarity and openness to collaborate over scientific and engineering fields are keywords in such a process. The mechanics based methods should preferably be straightforward to use and pedagogically explained.

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