Hygroelastic behaviour of wood-fibre based materials on the composite, fibre and ultrastructural level

Răzvan Cristian Neagu

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KTH Solid Mechanics
School of Engineering Sciences
Royal Institute of Technology
SE-100 44 Stockholm
Sweden

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Part of this work was performed at STFI-Packforsk AB, Stockholm, Sweden. STFI-Packforsk is one of the world’s leading R&D companies in the fields of pulp, paper, graphic media, packaging and logistics. The activities range from basic research to direct assignments, where the competence is utilised to find solutions applicable at the customers.

This work was also associated to the framework of the Wood Ultrastructure Research Centre (WURC), a centre of excellence in the field of wood ultrastructure at the Swedish University of Agricultural Sciences (SLU). The centre's main task is to carry out basic research of industrial relevance.

KTH Solid Mechanics belongs to KTH (the Royal Institute of Technology). KTH provides one third of Sweden’s capacity for engineering studies and technical research at post-secondary level. KTH conducts top-notch education and research of a broad spectrum – from natural science to all branches of technology. KTH Solid Mechanics has about 50 employees working with undergraduate and graduate education, research and contract work. The research is combined to an equal extent by experiments, computation and theory. The main activities concern material mechanics, fracture mechanics, contact mechanics and biomechanics.

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SUMMARY

Wood fibres can be used as reinforcement in plastics for load carrying purposes. Some advantages compared with conventional man-made fibres are that wood fibres come from a renewable resource, have high specific stiffness and strength, are generally less hazardous to health, biodegradable, and can be manufactured at low cost and high volumes. A clear disadvantage with cellulose-based materials for structural use is their dimensional instability in humid environments. The hygroelastic properties are of high importance in materials development of improved wood-fibre composites. This work deals with the stiffness and hygroexpansion of wood fibres for composite materials. The long-term aim is to design engineered wood fibre composites based on better basic knowledge of wood fibres.

Mechanistic models have been used to link the fibrous microstructure with macroscopic composite engineering properties. The properties have been characterized experimentally for various wood-fibre composites and their fibre-mat preforms, by means of curvature measurements at various levels of relative humidity, as well as tensile and compressive tests. From these test results and microstructural characterization, the longitudinal Young’s modulus and transverse coefficient of hygroexpansion of wood fibres were identified by inverse modelling. Some effects of various pulp processes and fibre modifications on the elastic properties of the fibre were observed, illustrating how the mixed experimental-modelling approaches can be used in more efficient materials screening and selection.

An improved micromechanical analysis for wood-fibre composites has been presented. The model is more appropriate to combine with laminate analogy, to link fibre properties on the microscale to the macroscopic composite properties and vice versa. It also offers the possibility to include the effects of ultrastructure since it can account for an arbitrary number of phases. An approach to model ultrastructure-fibre property relations has been demonstrated. It includes analytical modelling of multilayered cylindrical fibres as well as finite element modelling of fibres with irregular geometry characterized with microscopy. Both approaches are useful and could be combined with experiments to reveal insights that can pave way for a firmer link between the wood fibre ultrastructure and wood fibre properties.

Keywords: wood fibres, ultrastructure, structure-property relations, microfibril angle, composites, characterization methods, hygroelastic properties, micromechanics, modelling, reinforcement potential
PREFACE

The work presented in this thesis has been carried out at STFI-Packforsk AB and KTH Solid Mechanics from March 2002 to September 2006. It has been financially supported by and an integral part of the research programme ‘New Fibres for New Materials’ at STFI-Packforsk and during the last two years also of the Wood Ultrastructure Research Centre (WURC).

This thesis is a result of collaboration between many people. I would like to thank all of you who have supported and encouraged me in its completion. First and foremost, I wish to express my sincere thanks to my supervisor Assistant Professor Kristofer Gamstedt who trusted and believed in me from day one forward. None of this would have been possible without your brilliant guidance, tireless dedication, endless enthusiasm and exceptional skills. I really appreciate you always taking time to listen and talk to me, both on and off the record.

My deep thanks and admiration to Dr. Mikael Lindström at STFI-Packforsk who made it possible for me to work at STFI-Packforsk and for helpful suggestions throughout the course of this work. Thank you for always being so genial and optimistic, always helping and simply making things smoother. If it would not have been for the initiative of Professor Peter Gudmundson I would never had come in contact with Kristofer and Mikael. Many thanks also for the help I received in preparation of my first paper. I gratefully acknowledge the help I received from all my co-authors. Dr. Fredrik Berthold at STFI-Packforsk is particularly acknowledged for providing numerous wood fibre composites which generated invaluable experimental data. I have been fortunate to cooperate with, and I particular want to thank, Professor Janis Varna and Lic. Eng. Erik Marklund at Luleå University of Technology for many stimulating interchanges of ideas. Dr. Stig Bardage at SLU is acknowledged for among others the microscopy data on wood fibres needed as input to modelling. Finally, I would also like to express sincere appreciation to all my colleagues at KTH Solid Mechanics and STFI-Packforsk who made this time an enjoyable and fun time after all.

Reserved until last is praise for my mom, dad and brother to whom this work is dedicated. It is ultimately their merit, and because of the difficult sacrifices they have made, that I have been able to accomplish this thesis. “Vă ofer această teză de doctorat ca omagiu și sper să vă aducă bucurie și înlimire sufletească. Vă mulțumesc din tot sufletul pentru toate eforturile și emoțiile care le-ați avut alături de mine.”

R. Cristian Neagu
August 2006, Stockholm
LIST OF APPENDED PAPERS

This thesis contains an introduction and the following appended papers:

**Paper A**

**Paper B**

**Paper C**

**Paper D**

**Paper E**

**Paper F**
The following article and conference contributions are also a result of the PhD research project:


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MOTIVATION

Background

Structural polymer composite materials are commonly based on fossil carbohydrates as a raw material. It is desirable to replace some of these materials with new materials based on renewable resources such as cellulose, which is used in nature as a structural material in plants and trees. Wood and other natural-fibre reinforced polymers have large potential as structural materials and there has been increased interest in the use of lignocellulosic fibres as a load bearing constituent in composite materials [1-5]. This is due to environmental reasons and because they offer high performance at low cost [6]. Wood fibres are renewable, recyclable, biodegradable and possess a combination of properties, such as relatively high aspect ratio, excellent specific stiffness and strength, low density, and low cost. In terms of stiffness, typical values are summarized in Table 1, and one can see that wood and other natural fibres compare quite well with glass fibres especially given their lower density.

Table 1. Comparison of density and Young’s modulus of wood fibres with those of other natural and man-made fibres. (Paper C)

<table>
<thead>
<tr>
<th>Fibre</th>
<th>Density (g/cm³)</th>
<th>Young’s modulus (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spruce kraft</td>
<td>1.2</td>
<td>40</td>
</tr>
<tr>
<td>Pine kraft</td>
<td>1.2</td>
<td>32</td>
</tr>
<tr>
<td>Birch kraft</td>
<td>1.2</td>
<td>38</td>
</tr>
<tr>
<td>Eucalyptus kraft</td>
<td>1.2</td>
<td>35</td>
</tr>
<tr>
<td>Single cellulose</td>
<td>1.5</td>
<td>100</td>
</tr>
<tr>
<td>Cotton</td>
<td>1.5-1.6</td>
<td>9.5</td>
</tr>
<tr>
<td>Jute</td>
<td>1.3-1.45</td>
<td>34</td>
</tr>
<tr>
<td>Flax</td>
<td>1.43-1.52</td>
<td>37</td>
</tr>
<tr>
<td>Hemp</td>
<td>1.47-1.50</td>
<td>50</td>
</tr>
<tr>
<td>Kefaf</td>
<td>1.5</td>
<td>53</td>
</tr>
<tr>
<td>Ramie</td>
<td>1.5</td>
<td>84</td>
</tr>
<tr>
<td>Sisal</td>
<td>1.5</td>
<td>16</td>
</tr>
<tr>
<td>E-glass</td>
<td>2.6</td>
<td>76</td>
</tr>
<tr>
<td>S-glass</td>
<td>2.5</td>
<td>86</td>
</tr>
<tr>
<td>Aramid</td>
<td>1.44</td>
<td>62-128</td>
</tr>
<tr>
<td>Carbon</td>
<td>1.8</td>
<td>200-345</td>
</tr>
</tbody>
</table>

The properties of natural fibres show large variability and the values listed in Table 1 are average values of the longitudinal Young’s modulus. The scatter is explained by differences in fibre structure due to the overall environmental conditions during growth [7]. This scatter is
one disadvantage of the natural fibres compared with man-made fibres. Wood fibres show lower variability than many other cellulose-based fibres, such as fibres from annual crops, although the latter may have better mechanical properties [2]. Another drawback, compared with conventional inorganic fibres for composites, is the inherent susceptibility to moisture expansion of cellulosic fibres. Cellulosic-fibre reinforced plastics tend to swell considerably at water uptake and as a consequence mechanical properties, such as stiffness and strength, are deteriorated [8-10].

**Applications**

The largest market share for composite applications is held by glass fibre reinforced plastics followed by carbon fibre composites. The wood-fibre composites should first be compared with conventional contending materials like glass mat thermoplastics (GMT) and various sheet moulding compounds (SMC). The automotive and building industries are two examples where research and development efforts are being made to use lignocellulosic fibres as reinforcement in plastics. Automotive applications, e.g. Figure 1, include door panels, car roofs, package trays, load floors, spare tire covers, etc. In the building sector, typical applications are decking, fencing, railing, windows and doors. Other potential areas of application of wood-fibre composites are in e.g. the packaging and furniture industries.

![Figure 1. Concepts for exterior automotive parts made of natural fibre composites [5]. Potential applications for wood fibre composites?](image)

The bulk of wood composites used today is based on wood chips or flour rather than slender separated wood fibres [11]. In other words, the wood material is often used as filler instead of reinforcement. This causes severe stress concentration from the wood particles. The use of oriented separated fibres with relatively large aspect ratio should assure considerably
improved reinforcement. Wood fibres are found in abundance both as virgin pulp and waste products from saw, pulp and paper mills, as well as recycled paper.

Hygroelastic properties

There is a long and relatively mature experience to produce wood pulp fibres in large volumes for paper and board applications. In principle, there are two ways of fibre extraction, mechanical and chemical pulping, and combinations thereof. In mechanical pulping the fibres are softened and separated from each other using temperature, humidity and mechanical forces. In chemical pulping, substances of the middle lamella are chemically dissolved to an extent that makes liberation of fibres possible without further mechanical treatment. Depending on the pulping process, the chemical composition, fibre microstructure, fibre strength, flexibility and ability to adhere to other fibres or matrix materials vary widely between different types of wood fibres [12]. Wood pulp fibres are processed in a way to optimize the properties of the paper, which relies heavily on the fibre-fibre bond. Since flexibility is necessary to assure large contact area over fibre-fibre bonds through which stress is transferred [13], the pulp fibres are not optimized to be as stiff as possible.

In composites the stress is transferred through the matrix and an efficient fibre-matrix interface is therefore desirable. Structural composites are generally preferred to be stiff. The constituent fibres should therefore be as stiff as possible, and oriented in the primary direction of loading. Nevertheless, some of the conventional methods used in pulp and paper production may very well be employed in the manufacture of wood-fibre mats for composite applications. This would mean rapid and inexpensive production of suitable reinforcement with a relatively low variability of properties, if the process could be tuned differently to achieve optimal composite properties.

To take advantage of wood fibres in composites, they should be gently defibrated, have high aspect ratios and retain a relatively high stiffness and strength. It should also be emphasized that wood fibres are not inert but offer the possibility of being further tailored for improved properties by means of chemical, enzymatic or mechanical modifications e.g. Ref. [14]. To this end, the mechanical properties and dimensional stability of wood-fibre composites must be better understood. Concurrently, an increased understanding of the mechanical behaviour
of wood fibres is needed before they can be used with confidence as load bearing constituents in structural components.

The main engineering properties of composites to consider at the material selection stage are stiffness, hygroscopic dimensional stability, strength and fracture toughness. For structural applications with cellulose based composite materials, the most relevant properties are probably stiffness and dimensional stability. Knowledge of the hygroelastic behaviour of the constituent wood fibres is essential. Consequently, it is important to understand how the hygroelastic behaviour is governed by the nano- or ultrastructure of the fibres.

**Scope and aim**

Methods for materials development are needed to build a foundation for the necessary link between choice of raw materials, processing techniques, microstructure and the macroscopic mechanical properties of the material. Therefore suitable methods to characterize wood fibre hygroelastic properties need to be developed. An approach must be based on the recognition that an optimal balance of properties can be achieved by understanding the integration of fundamental properties at multiple scales from the (i) nanoscopic ultrastructural (e.g. cell-wall layer properties) to microscopic (e.g. fibre properties), to (ii) mesoscopic (e.g. fibre orientation distribution), to (iii) macroscopic (composite material) to (iv) component (structural application) level. A schematic of the length scales that need to be considered is given in Figure 2.

Figure 2 also shows some example of material design decisions that can be made on each scale and that will be critical to the performance of the composite and the final product. These include choice of fibre type, i.e. species, and fibre extraction process, i.e. pulping process (e.g. chemical or mechanical) and further treatments such as delignification. Cyclone fractionation could be made to differentiate between thin-walled earlywood and thick-walled latewood fibres [15]. The ultrastructure is of course important to consider. For instance, the difference in mechanical properties between earlywood and latewood fibres can be explained by a difference in the inclination of the reinforcing cellulose fibrils within the cell wall, i.e. microfibril angle [16, 17]. Besides, any pulping process is likely to alter the ultrastructure, which could also be modified chemically for improved properties.
Hence, the choice of raw material and extraction process and further modification will determine the properties of the fibres on the ultrastructural and microscopic level. These will influence the microstructure of the composite, e.g. in terms of the aspect ratio of the fibres [18]. However the microstructure of the composite is controlled by the selection matrix.
material (e.g. thermoset or thermoplastic) and manufacturing method of the composite, e.g. impregnation of fibre-mat preforms or hot pressing of commingled wood-thermoplastic preforms. Interfacial binding approaches and fibre orientation distribution are also of importance for the microstructure.

The aim of the thesis is to demonstrate some ways to assess the micromechanical efficiency of the fibres in providing improvement in some important engineering properties of the composite, i.e. increased stiffness and suppressed hygroinstability. The approaches used consider hygroelastic properties of wood fibres at different scales as shown in Figure 2. The thesis focuses on:

- test methods to determine the extent to which the wood fibres contribute to the stiffness of the composite, as well as the contribution of the fibres to the hygroexpansion of the composite at variable moisture content (Paper A-C)
- improved micromechanical modelling of wood fibre composites (Paper D)
- modelling ultrastructure-property relations in wood fibres for an increased understanding of their hygroelastic behaviour (Paper E-F)

HIERARCHICAL MATERIAL STRUCTURE

Wood has a complex material structure designed to assure the biological function and structural integrity of the tree [19]. It consists predominantly of tracheids (or wood fibres), long hollow cells of square to circular cross-section with tapered ends, bonded together in radial files with their long axes parallel with the axis of the tree. Figure 3 shows the hierarchical structure of wood from the tree down to the molecular level of cellulose.

Knowledge and understanding of the wood fibre structure is essential in the development of an appropriate modelling approach. A comprehensive review on the wood and fibre structure described from the microstructural level down to the ultrastructural level is given by Neagu [20].
Figure 3. Hierarchical structure from the tree to the molecular level (Illustration: Airi Illiste).
**Microstructure**

Wood fibres exhibit variability in fibre cross-sectional dimensions [21], not to mention variability along the fibre length, curl of the fibres etc., see e.g. Ref. [22]. Typical wood pulp fibres are about 1-3 mm in length and 20-40 \( \mu \text{m} \) in width [18]. The cross-sectional shape varies from being thick-walled boxlike for most of the latewood fibres to a relatively slender rectangular form for the thin-walled earlywood fibres, as shown in Figure 4a.

![Figure 4](image)

(a) (b)

Figure 4. (a) Light microscopy images of Norway spruce showing a transverse section with transition between earlywood and latewood. The scale bar length is 100 \( \mu \text{m} \). (b) Scanning electron micrograph of bordered pits in a softwood pulp fibre.

Pits, i.e. openings that permit passage of fluid in wood, are a particularity of the wood fibres [23]. The scanning electron microscopy (SEM) image in Figure 4b shows bordered pits in a softwood pulp fibre. Pits are an important morphological characteristic of wood fibres since they represent discontinuities in the fibre structure. From a structural mechanics point of view these can be seen as natural defects that cause stress concentrations which might result in crack initiation and ultimate structural failure [24].

The microstructure of tracheids influences properties such as wood density and aspect ratio of pulp fibres. The mechanical properties on the other hand are controlled by the composition and the structure on the nanometre level of the tracheid cells, often termed ultrastructure.
Wood fibres are multiphase composite materials with a highly intricate ultrastructure. This structural domain which is critical to the mechanical behaviour of wood fibres has been reviewed in Paper E. The cell wall material of wood fibres is made of several layers surrounding the lumen e.g. Ref. [25]. Cell wall layers shown in Figure 5a are designated as: the primary wall (P), the outer layer of secondary wall (S1), the middle layer of secondary wall (S2) and the inner layer of secondary wall (S3). The layers are themselves composite materials made of polymeric cellulose microfibrils aggregated into larger oriented structures embedded in a matrix of hemicellulose and lignin e.g. Refs. [26, 27] (Figure 5b). The distribution of wood polymers across the cell wall is not uniform, and cellulose and hemicellulose content is greater throughout the secondary wall layers than in P [28]. The S1, S2, and S3 are lignified to approximately the same extent.

The stiffness of the cell wall can primarily be attributed to the crystalline cellulose microfibrils which are aligned fairly parallel and trace a spiral around the cell wall. The hemicellulose molecules tend to be aligned with the cellulose chains [27] and may function as coupling agent between the cellulose and the lignin [30]. The omnipresence of hydroxyl groups in primarily hemicellulose is the main cause of hygroexpansion of the cell wall due to moisture absorption. Hemicellulose are polar hydrophilic polymers, and therefore the mechanical properties are strongly affected by changes in moisture content [31]. Lignin is an amorphous polymer and its structural function in the cell wall can be characterized as filling
up the cavities between the cellulose microfibrils, making the cell wall rigid and preventing buckling induced by longitudinal compression [32].

The fibre hygroelastic parameters depend primarily on the microfibril angle (MFA), which is the angular deviation of the microfibrils in the cell wall layers relative to the longitudinal cell wall axis [17, 30, 33-36]. The MFAs differ in each of the S1, S2 and S3 layers, but usually follow an S-Z-S arrangement [37]. A helix is defined as being either S or Z when the direction, as viewed from the outside of the fibre, is the same as the centre stroke of the letter. The most critical of layers is the S2 layer as it comprises a major part of the cell wall and has a dominant effect on the chemical and physical properties of the fibres. The thick S2 layer is considered to consist of microfibrils that are organized in a steep Z-helix. For Norway spruce, typical MFA is about 30˚ for earlywood fibres and about 2˚-10˚ for latewood fibres [38]. However, there is a large inherent variation of MFA within and between different wood fibres [39]. Moreover, the surface composition and surface morphology of the fibres is variable [40, 41] and of great importance for stress transfer between fibres over the fibre-matrix interface.

Ultrastructural changes occur during any fibre extraction processes. These include reduction of the MFA in the cell wall [42, 43], cellulose fibril aggregate enlargement [44, 45], formation of pores within the matrix of hemicelluloses and lignin, loosening of the P and/or S1 layer from the underlying layers [41], cell wall damage like fibrillation of the S1/S2 layers or delamination of the S2 layer into concentric lamellae [24], intensification of naturally occurring and creation of new e.g. kinks, microcompressions, etc [46]. These alterations at ultrastructural level reflect in turn the changes in fibre property at larger scales on, e.g. fibre dimensions and mechanical properties (reduced stiffness and strength [17, 33, 47]).

**MECHANICS APPROACH**

The hygroexpansion is intimately linked with the elastic properties of the constituents and their microstructural arrangement. For instance, constraints from stiff adjacent phases will subdue hygroexpansion and cause increase of internal stresses, which could result in interfacial failure. The hygroscopic and elastic properties are therefore commonly treated in terms of the resulting hygroelastic behaviour. To compare different fibre qualities, the coefficient of hygroexpansion and the Young’s modulus of the fibres are suitable measures.
The hygroelastic response of the composite can then be determined for a given microstructure. Similarly, the hygroelastic response of wood fibres can be determined from the behaviour on the ultrastructural level.

**Composite-fibre link**

Practical analytical/numerical-experimental methods for rapid characterization of the hygroelastic properties of wood fibres have been developed (Paper A-C). The methods are based on back-calculation, which is a common way to determine microscopic or local properties from a macroscopically determined property [15, 48-50]. The local property is implicitly found from the measured global property value through a mechanics model. These mixed analytical/numerical-experimental methods are directly applicable as engineering tools for material design. They are preferable to direct testing on single fibres [16, 17, 30, 33, 51-56] for fast characterization, albeit on the expense of being an indirect quantification. Testing of individual fibres is time consuming and requires a large amount of measurements since the fibre-to-fibre variability is large as mentioned earlier [7, 12].

The mixed experimental analytical work is summarized in Table 2. The methods involve a number of macroscopic experiments such as

- curvature or curl measurements on fibre mats at different levels relative humidity to determine the hygroexpansion coefficient of the fibres,
- tensile stiffness index measurement and elastic compression tests of loose fibre mats, and
- tensile tests of wood fibre composite specimens to determine the effective Young’s modulus of the fibres.

The macroscopic tests require micromechanical models to link to the fibre properties on the microscale to the macroscopic elastic behaviour. Therefore the experimental methods are combined with analytical models at different scales, which all together share the same approach. The fibre properties on the microscale are used to link the microscopic deformation mechanisms to the macroscopic elastic behaviour of fibre mats and composites. Experimental microstructural characterization was done to determine the fibre content, fibre orientation and dimensional variability of the fibres which are needed as input to modelling.
Table 2. Summary of the mixed experimental-analytical methods to determine hygroelastic properties of wood fibres. Filled dots ‘●’ indicate experiments necessary for microstructural characterization.

<table>
<thead>
<tr>
<th>Method</th>
<th>Experimental Modelling</th>
<th>Paper</th>
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<tr>
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<td>Macroscopic behaviour</td>
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<td>Fibre orientation</td>
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<td>Micromechanics</td>
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<td>Transverse hygroexpansion</td>
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<td>coefficient</td>
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<td>tensile stiffness index</td>
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<td>measurements</td>
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<td>Fibre mat compression</td>
<td>Applied pressure vs.</td>
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<td>Fibre mat tensile test</td>
<td>Tensile stiffness index</td>
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<td>Composite tensile test</td>
<td>Elastic moduli in</td>
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<td>principal material</td>
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<td>The hygroexpansion model is</td>
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<td>variation of fibre</td>
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|                           | orientation distribution through the thickness of the asymmetric fibre mat to determine the average coefficient of transverse hygroexpansion of the fibres from macroscopic curl measurements. The coefficient of hygroexpansion in the longitudinal direction of the fibre is generally negligible compared with the transverse coefficient of hygroexpansion, in particular for wood fibres with a small MFA [59] (Paper F). The micromechanical model for hygroexpansion from the fibre level to the fibre mat is to large extents inspired by the work of Schulgasser [58, 59]. The two methods of data reduction are considered for the wood fibre mats to determine the longitudinal Young’s modulus of the fibres. Modelling of the elastic compression in the thickness direction of a fibre assembly with an in-plane fibre orientation distribution and a distribution in fibre cross-sectional dimensions was used in the out-of-plane test. The compression model draws on work by Toll and Månsen [60] and Alkhagen [61]. The compression model is applicable on fibre mats with low density and long fibres. For fibre
mats of fairly high density with strong bonds and long fibres, laminate theory can be used to model their in-plane elastic behaviour. The fibre mat can be considered as a homogeneous laminate which can be modelled with laminate theory. The elastic behaviour of an individual fibre embedded in the fibre mat is based on the assumptions outlined by Schulgasser and Page [62].

For tensile loading of composites a laminate analogy can be applied to relate the in-plane short-fibre composite to a laminate of unidirectional plies, cf. e.g. Ref. [66]. This should be sufficient for wood fibre composites with planar fibre orientation distributions. The engineering constants for a unidirectional ply are obtained with the Composite Cylinder Assemblage (CCA) model [63-65]. The longitudinal Young’s modulus of the fibres can be identified as the parameter that minimizes the difference between the experimental data and the calculated values predicted by the model.

The method developed to estimate the transverse hygroexpansion coefficient of wood fibres could be used to screen fibre qualities suitable for dimensionally stable composites. Three methods have been used to determine the stiffness of wood fibres from simple macroscopic tests, i.e. tensile testing of wood-fibre composites, tensile testing of unimpregnated fibre mats and compression of fibre mats. They are all candidate methods for quality control and screening of suitable fibres for stiff composite applications. All methods are quantitative in the sense that the microstructure is accounted for in terms of fibre orientation, and fibre volume fraction as well as the fibre dimensional variability for the compression test method. The methodology could become useful for quantitative assessment of e.g. different types of candidate fibres for composite applications, processing parameters for tuning the pulp process and different fibre modifications and treatments for improved hygroelastic performance.

Micromechanics for wood fibre composites

The methods presented above have of course their limitations. The results depend on the assumptions of hygroelastic behaviour of the fibres and on the chosen micromechanical formulation. Predicted results are only as accurate as the applied micromechanical theory. The complicated microstructure of wood fibres was described earlier. To facilitate modelling and make use of existing micromechanical theories [63-65], simplifications and assumptions must be made with regard to the microstructure. It is usually simplified to a fibrous structure with a
high intrinsic degree of mechanical anisotropy that is stiff in the direction of the cellulose microfibrils and more compliant in the perpendicular directions. Thus, the fibres are assumed to be transversely isotropic (Paper A-C). The presence of lumen and the extension-twist interaction due to the helical structure of the fibres are also overlooked (Paper C). Therefore, a need for developing micromechanical models more appropriate for wood fibre composites was identified. An analytical model for composites with arbitrary number of phases (i.e. N-phase composite) with orthotropic properties of the constituents has been developed (Paper D).

The elastic properties of a wood fibre composite depend on the properties of the constituent fibres (phases), their relative volume fractions and on the microarchitecture, i.e. the fibre orientation distribution e.g. (Paper A), the fibre shape and the size of the lumen e.g. (Paper B), and whether it is filled with resin (Paper C) or not [67]. Other important aspects are the ultrastructural features that govern the fibre properties (Paper E-F). Laminate analogy can be applied to describe the elastic behaviour of wood fibre composites with planar fibre orientation distribution (Paper C). The wood fibre composite is thus treated as a stack of unidirectional layers of different orientation and with a fixed layer volume fraction obtained from the experimentally determined fibre orientation distributions, e.g. (Paper A, C). The key feature in this approach is determination of the elastic properties of a unidirectional layer.

To estimate or bound the overall elastic moduli of unidirectional composites, the auxiliary problem of a one or two-phase cylinder embedded in an unbounded matrix of another material, subjected to uniform conditions at infinity, has been used extensively [63-65, 68-70]. Many of these micromechanical models were developed for two-phase composites [63-65, 68] and are therefore not directly applicable to wood fibres if the lumen and the properties of the different cell wall layers need to be included. Extensions to multilayered fibres, i.e. N-phase composites, have been made although they are confined to transversely isotropic constituents [69-71], whereas wood fibres and their cell layers are orthotropic. Moreover, previous models are not applicable to hollow-cored fibres, e.g. Ref. [69]. The micromechanical model developed in Paper D is valid for orthotropic phase materials and for an arbitrary number of phases. The micromechanical model is a straightforward extension of the CCA model of Hashin [63-65] and the self-consistent model of Christensen and Lo [68].
The novelty lies in its applicability to multilayered hollow fibres with orthotropic material properties. The present micromechanical model was applied to

- identify the constituent parameters affecting a certain composite macroscopic property,
- evaluate the significance of the fibre anisotropy on the composite stiffness, and
- give guidelines for evaluation of possible errors when the fibre is considered as transversely isotropic.

The present micromechanical analysis is appropriate to use for wood fibre composites in combination with laminate analogy, to link to fibre properties on the microscale to the macroscopic composite properties and vice versa. It is also possible to include the effects of ultrastructure since it can account for an arbitrary number of phases. However, a better understanding of ultrastructure-fibre property relations is needed before reliable scale integration from the ultrastructural level of the constituents to the composite macroscopic properties can be made.

**Ultrastructure-fibre link**

Probably the least established link to date is the one between the ultrastructural and fibre level. The spatial and directional distribution of the hemicellulose, lignin and cellulose microfibrils, in the layered cell-wall structure, has a strong influence on fibre properties such as stiffness and hygroexpansion. For instance, the helical structure of the cell-wall layers implies that axial deformation is coupled with torsion. This coupling is often not accounted for in micromechanical models *(Paper C-D)*. However, it is possible to study the relation between cell-wall structure on nano and micron scales and fibre properties using similar theoretical tools applied at higher length scales, i.e. *(Paper A-D)*. It would then be possible, at least semi-quantitatively, to assess the influence of ultrastructure (fibril angle, fibril content and distribution) and mesostructure (as the layered cell-wall structure) on the fibre properties that in turn control the engineering properties of wood fibre based materials. In *(Paper E-F)* approaches to model ultrastructure-hygroelastic property relations are presented.

*(Paper E)* re-examines information on the ultrastructure that can be useful for modelling the hygroelastic behaviour of wood fibres. Also attempts to model ultrastructure-property relation that have been carried out over the years are reviewed. The review was compiled to form a basis for modelling approaches that aim for better predictions of the hygroelastic behaviour of
wood fibres. It also led to recommendations of specific problems on ultrastructure-property relations that are potentially rewarding to investigate. These include among others modelling the behaviour of fibres with the irregular geometry characterized by microscopy. As a result, an example of FE modelling of geometrically well-characterized fibres was given.

Geometrical data of latewood Norway spruce fibres, obtained using microscopy and three-dimensional reconstruction by Bardage [22], was used to build a FE model. Ultrastructural homogenization to obtain the property of each cell wall layer was done by applying the CCA model [64] in a recursive manner. The subsequent analysis showed that the modelling approach is valuable to assess the influence of the commonly neglected irregular shape on elastic behaviour and stress state in wood fibres. Analysis of the stress distribution in a fibre segment with real geometry in combination with a two-dimensional failure criterion (i.e. Tsai-Hill criterion) allowed for prediction of probable locations of failure in accordance with experimental observations [72]. Comparison was also made with an analytical model which assumes cylindrical fibre shape and is presented in Paper F. Predictions of the elastic properties made with analytical modelling of cylindrical fibres and with FE modelling of irregularly-shaped fibres were in agreement. The stress state and failure predictions only showed qualitative similarity.

In Paper F, a wood fibre is modelled as an assembly of coaxial hollow cylinders made of orthotropic material with helical structure. The hygroelastic response of the assembly due to axisymmetric loading and moisture content changes is obtained by solving the corresponding boundary value problem. The solution scheme is based on the state space approach combined with the transfer matrix [73-75]. This method is more convenient for analysing multilayered systems than conventional methods based on the stress function approach [76-78]. A novel feature in Paper F is the extension to include the hygroexpansion behaviour. Simplifying assumptions are that the hygroexpansion is linear and that the moisture content is independent on residual stresses. Moreover, a simple analytical ultrastructural homogenization method [71] is used to determine properties of the cell wall layers from properties of the main wood polymers. This method is applicable to multiphase composites such as the cell wall material. It is more appropriate to use than micromechanical models for two-phase composites, e.g. Refs. [79, 80], which are frequently applied to determine cell wall layer properties, e.g. Refs. [76, 77]. The validity of the method was confirmed with FE calculations [81] and an
analytical model based on a stress function approach [82]. The applicability of the model was
demonstrated with simulations on a typical softwood fibre, with properties taken from
literature (Paper E), to investigate the effect MFA on the hygroelastic properties of wood
fibres. It could be concluded that the predicted hygroelastic response captured experimentally
measured behaviour.

Paper E-F show a possible approach to model hygroelastic behaviour of wood fibres that
aims to bridge the gap between the ultrastructure and microscopic fibre properties. They are
both useful to reveal insights that can pave way for a firmer link between the wood fibre
ultrastructure and wood fibre properties.
SUMMARY OF APPENDED PAPERS

Paper A. Influence of wood-fibre hygroexpansion on the dimensional instability of fibre mats and composites

One of the disadvantages of cellulose-based fibres is their susceptibility to absorb water and swell. A handy macroscopic test method to determine the hygroexpansion coefficient of the wood fibres from experiments on fibre mat performs has been developed. It has been showed that the fibre orientation distribution varies through the thickness in fibre mats manufactured by dynamic sheet formation. This gives rise to a change in curvature if relative humidity is changed (see Figure 6a-b).

Figure 6. Curvature measurements at (a) 0% RH and (b) 50% RH showing curl around the MD axis. (c) Transverse hygroexpansion of the of softwood kraft fibres as function fibre elastic anisotropy.

Measurements of the curvature change can be used constructively to estimate the hygroexpansion coefficient of the fibres (see Figure 6c). This was done using an approach that links the macroscopic hygroexpansion behaviour to the fibre properties on the micro scale, with classical laminate mechanics and a micromechanical model. The effect of the change in thickness of the fibre mat was accounted for. Necessary input data in the laminate micromechanics model is the fibre orientation distribution through the thickness, the fibre mat grammage, the elastic properties of the fibre, and thickness and curvature measurements. The method could be used to quantitatively assess the hygroexpansion behaviour of wood fibres and could serve to rank the suitability and potential of different wood fibres as composite reinforcement where dimensional stability is of importance.
Experiments on unbonded systems of fibres, i.e. fibre mats, together with micromechanical models provide alternative ways to characterize the fibre properties. Candidate fibre materials could then be screened at an earlier stage in the processing chain and the most suitable type of fibre for a certain composite application could be singled out. Compression experiments have been performed on fibre-mats (Figure 7a) to determine the pressure vs. fibre volume fraction. From this relation an effective value of the longitudinal Young’s modulus of the fibre has been determined using a statistical model and Bernoulli-Euler beam theory developed by Toll and Månson [60]. The modelled deformation unit in shown in Figure 7b.

![Figure 7. (a) Structure of a wood fibre mat. (b) Modelled deformation unit. (c) Confocal microscopy image of softwood kraft fibre cross-sections.](image)

The model is adopted for wood-fibre cross-sections (i.e. box-like structure for latewood and flat lamellar structure for earlywood) and also takes in account the dimensional variability of the fibres (see Figure 7c). Confocal laser scanning microscopy and image analysis have been used to determine the variation in cross-section. A laminate model to determine the fibre stiffness from back-calculation of the fibre mat stiffness has also been used.

Results from the two methods compare well. The two methods are complementary, since the compression method is appropriate for low-density fibre mats and while the laminate model works better on high-density fibre mat. The two methods provide quantitative results since the fibre volume fraction, orientation distribution and fibre dimensional variability are taken into account. This is the main advantage as the potential of various types of fibres can be investigated, even if the microstructures are different.
Paper C. Stiffness contribution of various wood fibers to composite materials

A quantitative analytical experimental method has been developed to determine the contribution of the fibres to the elastic properties of the composite. A large variety of composites based on various wood fibres in an epoxy vinyl ester matrix were investigated. The microstructure of a composite is shown in Figure 8a. A micromechanical model (see Figure 8b) and classical laminate mechanics were used to relate the elastic properties of the fibres to the elastic properties of the composite.

![Figure 8](image)

Figure 8. (a) Microstructure of a wood fibre/epoxy vinyl ester composite. (b) Model microstructure, the composite cylinder assemblage (CCA) model.

The contributing Young's moduli of the fibres in the longitudinal direction were back-calculated from tensile tests of the composites. One finding is that there was an optimum in fibre stiffness as a function of lignin content and that unbleached fibres are more suitable than bleached fibres for use as stiffening reinforcement.

It was also found that industrially pulped hardwood fibres had higher stiffness than corresponding softwood fibres. The effects of hornification, prehydrolysis and sulphite processing were also investigated. The results indicate that mild defibration process should be used, that does not damage the cell-wall structure so that the inherent high stiffness of the native fibres can be retained. The proposed method works well to rank wood fibre candidates in terms of their contribution to the composite stiffness. The composite tensile test is preferable since it is closer to the end use.
Paper D. Stiffness of aligned wood fiber composites: Effects of microstructure and phase properties

A micromechanical model for $N$-phase composites with orthotropic properties of constituents has been developed. The model is a straightforward generalization of Hashin’s concentric cylinder assembly model [64] and Christensen and Lo’s [68] generalized self-consistent approach. The effect of wood fibre anisotropy and their geometrical features on wood fibre composite elastic parameters was analysed. It was found that most macroscopic properties are governed by only one corresponding property at the cell wall level. Several of the unknown anisotropic constants characterizing the wood fibre are not affecting the stiffness significantly and rough assumptions regarding their value would suffice. The errors introduced by application of Hashin’s model [64] and neglecting the orthotropic nature of the cell wall were evaluated and shown to reach almost 20% (overestimation). The results are very important in attempts to back-calculate the fibre properties.

![Figure 9. The effect of different fibre volume fractions on composite transverse shear modulus ($G_{23}$) with empty (○) and filled (●) lumen fraction of 0.04, and empty (○) and filled (●) lumen fraction of 0.36.](image)

The role of lumen (whether it is filled with the matrix material or not) has a very large effect on the composite shear properties (see Figure 9). The effect of geometrical deviations from circular cross-section was analysed using the finite element method. The effect of circular, elliptical or square fibre cross-section on composite properties is insignificant in the case of filled lumen. If the lumen is empty the only properties significantly affected are the transverse shear modulus and Poisson’s ratio. The present micromechanical analysis is suitable for wood fibre composites and provides the option to study effects of ultrastructure since it can account for an arbitrary number of phases.
Ultrastructure of wood fibres has a strong influence on properties such as stiffness and hygroexpansion. This structure-property relation can be modelled with e.g. composite mechanics in order to assess the influence of ultrastructure on the fibre properties that in turn control the engineering properties of wood fibre composites and other wood-based materials. This paper reviews some of the existing knowledge of ultrastructure of softwood fibres and of modelling of the hygroelastic properties of these fibres. Rather comprehensive information on the ultrastructure is presented that can be useful in modelling the hygroelastic behaviour of wood fibres. Many attempts to model ultrastructure-property relation that have been carried out over the years have been useful in revealing valuable insights that can help clarify experimentally determined behaviour of wood fibres. Still, many modelling approaches in literature are of limited applicability, not the least when it comes to geometry of the fibre structure. Therefore, an example of finite element modelling of geometrically well-characterized fibres is given.

Figure 10. Deformation behaviour of a fibre segment with a MFA of 10° in S2 under applied axial strain of 1% for boundary conditions of (a) free rotation and (b) no rotation.

The approach is shown to be useful to assess the influence of the commonly neglected irregular shape on elastic behaviour (e.g. Figure 10a-b) and stress state in wood fibres. Predictions of the elastic properties made with analytical modelling of cylindrical fibres and with finite element modelling of geometrically characterized fibres are in concert. The stress state and failure predictions only show qualitative similarity. Nevertheless, modelling fibres with the irregular and more realistic geometry in combination with experiments on single fibres is required for a better and more quantitative understanding of the hygroelastic behaviour and particularly failure of wood fibres.
Paper F. Modelling of effects of ultrastructural morphology on the hygroelastic properties of wood fibres

An analytical modelling approach has been used for prediction of hygroelastic response, and assessment of the stresses in thick-walled multilayered cylinder models of wood fibres. The hygroelastic response of the layered assembly due to axisymmetric loading and moisture content changes was obtained using an efficient solution scheme based on the state space approach and the transfer matrix method. This was combined with an analytical ultrastructural homogenization method, used to link hygroelastic properties of constituent wood polymers to properties of each layer. The validity of the method was confirmed with finite element calculations and an analytical model based on a stress function approach.

Simulations on a typical softwood fibre with properties taken from literature were made. Predicted hygroelastic response (e.g. Figure 11) captured experimentally measured behaviour. Results showed that the twist-extension coupling induced by the helical structure of the cell wall has significant effect. It was also shown that the ultrastructure, i.e. the microfibril angle, will control the hygroexpansion in the same way as it controls the compliance of the cell wall. No reduction or increase of the expansion in a given direction can be achieved without concomitantly reducing or increasing the compliance in the direction of interest in the same proportion. When fibres are subjected to changes in moisture content, twisting was shown to be the main deformation mechanism. Qualitative failure trends comparable with experimental observations could be established with a stress analysis and a simple plane-stress failure criterion. The analytical modelling approach represents a simple and powerful approach to study effects of ultrastructural morphology on hygroelastic properties of wood fibres.
SUGGESTIONS FOR FUTURE INVESTIGATIONS

Composites, made of wood fibres and polymers, are relatively novel materials yet to reach their full potential. A long-term ambition is to tailor the ultra- and microstructure of the fibres and composites in order to obtain optimal material performance for a certain application. The mechanistic approach in this thesis has been used to devise engineering tools to determine the fibre contribution to the macroscopic hygroelastic properties of the composite, which could contribute to the development of wood-based composites in structural applications. A better understanding of the processing, ultrastructure, microstructure, damage mechanisms and mechanical properties is still needed. Experimental micromechanics combined with modelling should provide a viable way for a continuation which should look closer at e.g. transient moisture sorption and barrier properties as well as damage accumulation and strength of the composites and wood fibres.

Apart from problems arising from dimensional instability in wood fibre composites, mechanical properties such as stiffness and strength are known to deteriorate if too much moisture is absorbed. If composites based on wood fibres are to be used, it would be valuable also to estimate how the dimensions of the composite structure changes due to moisture ingress and subsequent drying on an annual time scale. The first step would be to determine the diffusion coefficient of the composite. Similar to the hygroelastic schemes above, i.e. Paper A-C, it would then be possible to assess the contribution of the fibres and the fibrous microstructure to the moisture absorption rate of the composite, and thereby determine which kind of fibres are the most suitable to retard moisture sorption.

To devise methods for strength prediction of the composites, is more challenging since the strength generally depends on the most severe microstructural heterogeneity. The strength is intimately linked with the microarchitecture, i.e. aspect ratio, dispersion and orientation of the fibres, the fibre-matrix interface, the fibre strength distribution, etc. On the fibre level, the fragmentation test developed to characterize the strength distribution of conventional composite fibres made of glass or graphite, may very well be used also for wood fibres. The method is particularly suitable for natural fibres, since these generally show a large scatter in strength properties, which is quantified by the strength distribution. The influence of cell wall modification on the strength of fibres can then be investigated with significantly better accuracy than e.g. the zero-span tensile test (cf. Ref. [83]). Along these lines attention should
be given to development of experimental techniques to quantify interfacial adhesion and modelling of damage mechanisms. The work could for example result in: determination of other important mechanical properties like fracture toughness, techniques to compare different chemical modifications of the fibre surfaces or fibre bulk for improved stress transfer between fibres in the composite, etc.

A continuation should also look more closely on the link between the ultrastructure and the fibre properties. A similar mechanistic approach may be undertaken at the ultrastructural level. The efforts should be directed towards experimental characterization of the ultrastructural response, so that mechanistic models (e.g. Paper E-F) could be used to relate it to the fibre level. Hence, any theoretical approach should be accompanied by independent and extensive experimental measurements on each analysed property.

Ultrastructural characterization by silica casting [84] of fibres is a promising method to provide more accurate structural models of wood fibres. Preliminary results from silica casts of wood fibre mats show with quite good confidence that it is possible to determine the MFA on average for different pulp fibres with environmental SEM. It is also possible to obtain geometrical data for fibre-fibre bonds in paper and fibre mats that may be useful as input to a composite model in an attempt to predict the hygroexpansion behaviour of fibres from the dimensional changes of fibres due to the formation of fibre-fibre bonds.

Modelling the behaviour of fibres with the irregular geometry characterized should continue and be combined with experiments. Analysis of the stress distribution in a “real” fibre and the effect of structural inhomogeneities such as bordered and cross field pits would contribute to a better understanding of failure mechanisms. There is also need for a more comprehensive and realistic modelling of the effect of the helical structure of the fibres. The effect of a progressive change of MFA through the thickness of the cell wall and its implications on the stress distribution and stress transfer within the cell wall should be investigated.

It would be desirable to experimentally validate the twist-extension coupling effect on the relation between the anisotropic hygroexpansion and elastic behaviour, i.e. the expansion in a given direction is basically proportional to the inverse of the Young’s modulus in that direction [59] (Paper F). This might be useful in order to develop a practicable plan for experimental work to determine the hygroelastic properties of the cell wall and its
constituents. In any case, better knowledge of influence of time, temperature and moisture content on the elastic properties of the wood polymers is required. Furthermore, it should be a good idea to check if the experimentally proven linearity between rotational twist and moisture content in single fibres [85] can be used to determine the hygroexpansion coefficients of wood fibres.

An attempt to scale up ultrastructure-fibre effects to macroscopic engineering properties of the composite should be made. It would be desirable to investigate how simplifications (i.e. neglecting twist-extension coupling) limit the predictive capabilities of micromechanical models for wood fibre composites, e.g. Paper D. This could be the first step towards a systematic multiscale approach which includes scale bridging and consideration of interactions across multiple scales. This is of importance if the long-term ambition to tailor the ultra- and microstructure of the fibres and composites should be achieved.
REFERENCES


Hygroelastic behaviour of wood-fibre based materials


