Wood-fibre composites: Stress transfer and hygroexpansion

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KTH accounts for one-third of Sweden’s technical research and engineering education capacity at university level. Education and research cover a broad spectrum – from natural sciences to all the branches of engineering.

Part of this work was performed at Innventia AB. Innventia AB is a world leader in research and development relating to pulp, paper, graphic media, packaging and biorefining.
SUMMARY

Wood fibres is a type of natural fibres suitable for composite applications. The abundance of wood in Swedish forests makes wood-fibre composites a new and interesting application for the Swedish pulp and paper industry. For large scale production of composites reinforced by wood fibres to be realized, the mechanical properties of the materials have to be optimized. Furthermore, the negative effects of moisture, such as softening, creep and degradation, have to be limited. A better understanding of how design parameters such as choice of fibres and matrix material, fibre modifications and fibre orientation distribution affect the properties of the resulting composite material would help the development of wood-fibre composites.

In this thesis, focus has been on the fibre-matrix interface, wood-fibre hygroexpansion and resulting mechanical properties of the composite. The importance of an efficient fibre-matrix interface for composite properties is well known, but the determination of interface properties in wood-fibre composites is difficult due to the miniscule dimensions of the fibres. This is a problem also when hygroexpansion of wood fibres is investigated. Instead of tedious single-fibre tests, more straightforward, macroscopic approaches are suggested. Halpin-Tsai’s micromechanical models and laminate analogy were used to attain efficient interface characteristics of a wood-fibre composite. When Halpin-Tsai’s model was replaced by Hashin’s concentric cylinder assembly model, a value of an interface parameter could be derived from dynamic mechanical analysis. A micromechanical model developed by Hashin was used also to identify the coefficient of hygroexpansion of wood fibres. Measurements of thickness swelling of wood-fibre composites were performed. Back-calculation through laminate analogy and the micromechanical model made it possible to estimate the wood-fibre coefficient of hygroexpansion. Through these back-calculation procedures, information of fibre and interface properties can be gained for ranking of e.g. fibre types and modifications.

Dynamic FT-IR (Fourier Transform Infrared) spectroscopy was investigated as a tool for interface characterization at the molecular level. The effects of relative humidity in the test chamber on the IR spectra were studied. The elastic response of the matrix material increased relative to the motion of the reinforcing cellulose backbone. This could be understood as a stress transfer from fibres to matrix when moisture was introduced to the system, e.g. as a consequence of reduced interface efficiency in the moist environment. The method is still qualitative and further development is potentially very useful to measure stress redistribution on the molecular level.
PREFACE

This work has been carried out at KTH Solid Mechanics (2005-2007), KTH Fibre and Polymer Technology (2007-2010) and at Innventia AB. It has formed an integrated part of, and been financially supported by, the cluster program “New Fibres for New Materials III” sponsored by Södra, Mondi, Billerud, Korsnäs, Stora, M-real, BASF and Hartmann. The industrial parties are all gratefully recognized for their support.

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Slutligen, tack till min familj, till Martin och Hilda, för att ni delar mitt liv och finns vid min sida, för all glädje, kärlek och trygghet ni ger mig.
LIST OF APPENDED PAPERS

Paper A
Dynamic-mechanical properties of wood fibre reinforced polylactide: Experimental characterisation and micromechanical modelling
*Journal of Thermoplastic Composite Materials* 19(6): 613-637

Paper B
Effects of moisture on dynamic mechanical properties of wood-fibre composites studied by dynamic FT-IR spectroscopy
*Journal of Reinforced Plastics and Composites* 27(16-17): 1709-1721

Paper C
Characterization of interfacial stress transfer ability by dynamic mechanical analysis of cellulose fiber based composite materials
Manuscript submitted for publication

Paper D
Role of fibre-fibre and fibre-matrix adhesion in stress transfer in composites made from resin-impregnated paper sheets
*International Journal of Adhesion and Adhesives* 29(5): 551-557

Paper E
Moisture uptake and hygroexpansion of wood fiber composite materials with polylactide and polypropylene matrix materials
*Polymer Composites* 30(12): 1809-1816

Paper F
Contribution of wood fiber hygroexpansion to moisture induced thickness swelling of composite plates
*Polymer Composites* In Press
CONTRIBUTION REPORT

Contribution of the author to the appended papers:

**Paper A**
Experimental work
Modelling
Writing of paper

**Paper B**
Experimental work
Writing of paper

**Paper C**
Experimental work
Modelling
Writing of paper

**Paper D**
Joint efforts in experimental work
Interpretation of data
Writing of paper

**Paper E**
Joint efforts in experimental work
Joint efforts in interpretation of data
Writing of paper

**Paper F**
Experimental work
Joint efforts in modelling
Writing of paper
In addition to this thesis, the work has resulted in the following publications

**Dynamic-mechanical properties of wood fibre/polylactide**

**Dynamic-mechanical properties of wood-fibre reinforced polylactide: Experimental characterization and micromechanical modelling**

**Micromechanical approaches to development of improved wood-fibre biocomposites**

**Effects of relative humidity on load redistribution in cyclic loading of wood-fibre composites analysed by dynamic Fourier transform infrared spectroscopy**

**Stress transfer and failure in pulp-fiber reinforced composites: Effects of microstructure characterized by X-ray microtomography**

**Measuring fibre-fibre bonds in 3D images of fibrous materials**
Hygroexpansion of wood-fibre composite materials: Effects of cell-wall cross-linking and composition of thermoplastic matrix

Measuring fibre-fibre contact in 3D images of fibrous materials

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1 INTRODUCTION
During the last decade, environmental awareness has led to a considerably increased interest in developing sustainable materials to replace materials made from fossil-based resources. Polymers and reinforcing fibres from renewable resources, e.g. annual plants or wood, is one way to produce renewable and biodegradable composite materials for packaging and structural applications.

Wood and wood fibres are commonly used as structural material, e.g. as particle and flake board. In the pulp and paper industry, wood fibres are used to produce a wide variety of products with different properties: paper for printers, newsprint, paper for magazines, packaging materials such as board and corrugated board, paper for tissues and fluff products for diapers, to mention a few.

Due to their mechanical properties, wood fibres are also suitable as reinforcement in composite materials. The low density of natural fibres makes their specific properties comparable to those of commonly used glass fibres [1]. One application of natural fibre composites is interior panels in cars, where flax and hemp fibres are used as reinforcement in synthetic resins. Other promising applications for wood-fibre composites are packaging materials, furniture and non-structural building components.

Wood and other natural fibres offer many advantages compared to synthetic fibres, e.g. glass and carbon fibres. They are relatively inexpensive, and the cost of wood fibres lies in the lower region. They are derived from renewable resources and are biodegradable, and they are also less abrasive than the traditionally used synthetic fibres to equipment used in the manufacturing processes. These advantages have led to an increased interest in natural fibre composites. Many sources of natural fibres are utilized, e.g. wood [2-7], jute [8], flax [9], hemp [1], sisal [10], cotton [11], oil palm [12] and bamboo [13]. Compared to other natural fibres, wood has the advantage of around-the-year harvest, and a well-developed infrastructure for cutting, pulping, fibre treatment and preforming manufacture could essentially be provided by the well established pulp and paper industry. The benefits of wood fibres have led to intense research on wood-fibre composites; different types of fibre and pulping processes have been investigated [14-16] as well as suitable matrix materials [2] and methods for modification of both fibres and reinforced polymers to improve the interface properties [17, 18]. There is a general agreement that wood-fibre composites offer an important contribution to the composite field, and better understanding of the effects of the mentioned design parameters (types of fibres and fibre pre-processing, polymer and polymer modification as well as process conditions...
such as process temperature, pressure and shear flow) on composite properties would lead to improvement of the composite material properties.

Adequate adhesion between fibres and matrix is crucial to achieve optimal mechanical properties of wood-fibre composites and to make them appropriate as structural materials. The fibre surface is suitable for chemical modifications aiming to improving the interface properties. Fibre modifications can also be used to improve dimensional stability, i.e. reduce fibre hygroexpansion and absorption of moisture. The dimensional stability of wood fibres subjected to moisture is an Achilles’ heel of wood-fibre composites, since contact with moisture leads to softening and swelling of the fibres, and thereby to softening and deformation of the composite material. The wood-fibre hygroexpansion and interface properties hence have in common that they can readily be improved by chemical modifications of the wood fibres, but also that they are quite difficult to measure or determine experimentally. Single-fibre tests have been performed [e.g. 19-22], but these tests are time consuming and the variability is large. Measurements of composite samples are straightforward compared to single-fibre tests and can be considered to reflect the effective average behaviour of the fibres, since all fibres in the composite contribute to the composite properties. The scope of this thesis is to investigate ways of determining hygroexpansion properties of wood fibres and fibre-matrix interface properties from such wood-fibre composite measurements.

Micromechanical modelling is a powerful tool to predict wood-fibre composite properties or, if used backwards, to quantify properties of wood fibres and fibre-matrix interface. Micromechanical models are commonly and successfully used to predict thermal and elastic properties of both synthetic and natural fibre composite materials. The composite theory developed by Hashin and Rosen [23] as well as Halpin-Tsai’s [24], Tsai-Hahn’s [25] micromechanical models and Halpin and Pagano’s laminate approximation for short fibre composites [26] are commonly used to link the elastic properties of the composite constituents and composite microstructure to the elastic properties of the composites.

Some models are extended to thermoelastic properties, e.g. [27, 28], but the literature on hygroelastic properties is not that extensive, since the effect of moisture on dimensional stability of glass- and carbon fibre composites is small. The mechanics of linear thermo- and hygroelasticity are, however, essentially the same, and if the variation in moisture content in the different phases in the composite are accounted for, the models developed for thermoelasticity are valid also for hygroelasticity. Models of this type are originally developed with continuous synthetic fibres in mind, e.g. carbon and glass fibres, but are also applicable on natural fibre composites [29].
Micromechanical models of this type are used in Papers A, C and F, to evaluate the not so easily measured properties of fibre-matrix interface (Papers A and C) and wood fibre hygroexpansion (Paper F). Wood tracheids have high aspect ratios despite their short length [30, 31] and the composites studied in this thesis are based on intact, slender and well-separated wood fibres, which have not been broken down during a forceful manufacturing process such as injection moulding. For this reason, micromechanical models for continuous fibres are chosen over short-fibre approaches. Papers B, D and E focus on experimental techniques (studies of stress transfer in Papers B and D and of hygroexpansion in Paper E). The content of the different papers and how they are correlated is visualized in Figure 1.

This introduction is followed by a brief inventory of how stress is transferred in paper and composites and of how stress transfer and interface properties are commonly measured. Modelling approaches used by other authors to quantify interface properties are discussed. A description of the wood fibre ultrastructure is given, since the rather complex configuration and composition of wood fibres affect their physical and mechanical properties, making them anisotropic. The anisotropy of the mechanical properties and hygroexpansion of wood fibres, and simplifications thereof, are then discussed. Fibres and matrix materials used are presented in the section “Materials and Methods” where material preparations as well as experimental methods used are described. This is followed by the modelling strategies adopted for characterization of interface efficiency and wood fibre hygroexpansion. Results are presented and discussed and suggestions for future investigations are given.
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Figure 1: Schematic illustration of the appended papers.
1.1 FIBRE-MATRIX INTERFACE

In composite materials, stress is transferred between matrix and fibres. Several bonding mechanisms between fibres and matrix are possible, e.g. interdiffusion, chemical bonding and mechanical locking, as illustrated in Figure 2.

Interdiffusion is a bond formed by diffusion of polymer chains on one surface into the polymer network of the other phase. If groups on the surfaces of fibre and matrix form bonds, different kinds of chemical or physical bonding can occur. The bonds could be covalent, dipole or hydrogen bonds, or van der Waal forces. Coupling agents are commonly used to form chemical bonds between fibres and matrix. The strength of the bonds depends on type and amount of the chemical bonds or on the degree of entanglement and the amount of entangled chains.

Mechanical locking or keying occurs when the fluid matrix solidifies on a rough fibre surface. For synthetic fibres, which generally are rather smooth, this type of frictional bond is considered to give only a small contribution to the fibre-matrix interface strength. More than one bonding mechanism may occur, e.g. chemical bonding and mechanical locking, and types of bonding in the composite is naturally depending on the types of fibres and matrix. Also depending on the characteristics of the material, the bonded zone is described either as an interface, i.e. as a surface between fibres and matrix, or as an interphase, i.e. as a third material phase with properties between those of fibres and matrix.

In wood-fibre composites, however, the fibre-matrix interface is not the only possible stress transfer mechanism. Depending on types and amount of matrix and fibre-fibre contacts, stress transfer is possible also between fibres. This is how stress is transferred in paper and board, as illustrated in Figure 3.
While wet pressure is used to improve stress transfer between fibres, various methods, e.g. fibre modifications and polymer grafting, are used to improve fibre-matrix interface. Several methods and their effects on natural-fibre composite properties are described in the reviews by Bledzki and Gassan and Nabi Saheb and Jog [32, 33]. Methods applied to wood-fibre composites are described by e.g. Bledzki [17].

Figure 3: Illustration of stress transfer regions: fibre-matrix interface in composite material and fibre-fibre bond in paper application.

1.1.1 Stress transfer in paper and board

In paper and board, where stress is transferred between fibres, both the amount of fibre-fibre bonds and bond strength are of importance. In the literature, the amount of fibre bonds is commonly related to the relative bonded area (RBA), defined as the fibre to fibre bonded area divided by total fibre area. Several authors have correlated RBA to paper strength [34-36] and stiffness [37, 38]. A light scattering technique is commonly used to measure RBA [35, 39]. Free fibre surfaces reflect more light than bonded fibre segments and hence paper sheets with few fibre-fibre bonds, i.e. small RBA, reflect more light than a well consolidated sheet with many fibre-fibre bonds and large RBA. The reflected light of the total fibre area is found by measurements of unbonded reference sheets, and the RBA is then determined by comparing the scattered light from investigated samples and unbonded reference sheets. The study of hydrogen gas absorption has been suggested to give more accurate results since the nitrogen molecules are smaller than the wavelength of light [40, 41]. Surfaces inside lumens and micro cracks in the fibre wall will however also be identified as free surfaces, making these indirect methods imprecise [42].

A direct method to determine RBA has been presented by Yang et al [43] who used image analysis to study thin cross sections of paper sheets. Their study suggests that one fibre cross-section could be bonded to as many as five different fibres if lumen was not collapsed, compared to a maximum of two as suggested by other authors [44]. The method is limited by resolution and the fact that examination of the cross sections is done manually, which could introduce an error to the method since definition of fibre contacts may differ between operators. An improved version of this direct but approximate method is developed in Paper D, where X-ray tomography is
used for enhanced resolution and computerized image analysis tools are used to identify fibre-fibre contacts.

Bond strength has been studied in single-fibre tests [19-22, 45]. Different set-ups, e.g. fibre crossings and fibres pressed against thin shrives or cellophane, have been used to study shear strength of fibre bonds. However, these tests are difficult to perform; sometimes only about 30-50 % of the prepared specimens are deemed good enough to test and the variability of the test results is large, in some cases over 100 % [20]. Experiments of this type provide information of the strength of fibre joints, but to determine the strength of the actual fibre-to-fibre bonds, the bonded area in the joint must be determined. The surface of wood fibres is sometimes rough, and overlapping fibres are not necessarily bonded, or even in contact, over the entire overlapping area [46]. In the studies mentioned above, the bonded area is determined by light scattering techniques or simply by studying samples in a microscope.

Indirect test methods to characterise fibre-fibre bonding have been applied to avoid the difficulties of single fibre tests. $Z$-strength is such an indirect measure of the fibre-to-fibre bonding in paper sheets [42, 47]. If the fibres in a paper sheet have an in-plane orientation, the strength in the out-of-plane direction, i.e. $z$-direction, is mainly dependent on the strength and amount of fibre-fibre bonds. However, if some of the fibres have an out-of-plane orientation, which is commonly the case especially for commercial sheets, the $z$-strength will be dependent also on the axial fibre strength. A difference between the $z$-strength test and single-fibre methods is that the $z$-strength determines the bond strength normal to the fibre surface, while single-fibre tests are used to determine shear strength of the fibre to fibre bond. Shear strength and failure are believed to be the most important mechanisms in paper fracture. Despite this, and due to the simplicity of the $z$-strength tests, these methods for determination of fibre-fibre bonding strength are far more common than single-fibre tests.

1.1.2 Stress transfer in composite materials

The properties of the fibre matrix interface contribute to stiffness and strength of the composite and control the fracture behaviour of the material to a large extent. A weak interface gives poor stress transfer between matrix and fibres and the reinforcing abilities of the fibres are hence not fully utilized. At failure, fibres will be pulled out of the matrix, giving a ductile fracture. Strong interface gives stiffer and more brittle materials. At failure, the fibres will rupture rather than being pulled out of the matrix. It is thus important to control interface properties when designing a new material to obtain a combination of good mechanical properties.
Direct evaluation of stress transfer abilities is difficult due to the complex nature and the small scale of the fibre-matrix interface. For composite applications, single-fibre tests are used, e.g. fibre fragmentation and fibre pull-out, both illustrated in Figure 4.

![Illustration of single fibre tests for testing of interface properties: fibre fragmentation test and fibre pull-out test.](image)

Figure 4: Illustration of single fibre tests for testing of interface properties: fibre fragmentation test and fibre pull-out test.

The evaluation of the information assessed from these tests has been discussed by several authors. Désamont and Favre [48] and Favre and Merienne [49] pointed at the operational difficulties of the single-fibre pull-out test, but concluded that even though the single-fibre pull-out test offers a coarse simplification of the complex structure of the composite, important information about fibre-matrix interaction can be derived from the tests. Preparation of test specimens is considered cumbersome, even for synthetic fibres, which show less variability and are much longer, and thus easier to handle, than wood fibres. In their review on single-fibre fragmentation test, Tripathi and Jones [50] highlight the problem of correct interpretation of single-fibre fragmentation tests. Their review shows that it is difficult to predict macroscopic composite properties if the knowledge of the interphase is based solely on results from single-fibre fragmentation tests. This is explained by the somewhat unrealistic test conditions and the difficulty of separating interphasial strength from other mechanisms activated during the test, e.g. matrix yielding and cracking. However, for the ranking of different surface treatments with respect to interfacial shear strength, the single-fibre fragmentation test is very useful [51]. A different approach of single fibre tests is offered by Raman spectroscopy [52]. Through the study of changes in peak wavenumber shifts in the Raman spectra of stretched fibres embedded in polymeric matrix, information of interface efficiency is attained. The difficulties of handling and mounting single wood fibres are, however, the same as for other single fibre tests.

To avoid cumbersome single fibre tests, the dynamic mechanical analyser (DMA) technique has been suggested for evaluation of interface properties during cyclic loading, i.e. not at interfacial failure as in the single-fibre tests described above. Kubát et. al [53] used dynamic mechanical thermal analysis (DMTA) to investigate the effects of a coupling agent in a glass sphere-polymer composite. They suggested that the coupling agent creates an
interface between fibres and matrix, and derived a parameter to compare efficiency of interfaces. Simple models were used to derive an interface parameter and the authors concluded that the method was convenient, albeit approximate. Afaghi-Khatibi and Mai [54] used DMTA to study the effect of cyclic fatigue on interface properties in a carbon fibre-epoxy resin composite. They concluded, in agreement with Kubát et al., that the DMA technique can be used to detect the presence of different interfaces in composite systems, but that results should be interpreted with care. In this thesis interface is studied through DMA-measurements in Papers A and C.

In Paper C, a more sophisticated model for interface studies developed by Hashin [55], was employed. The model is based on his previous composite models where perfect fibre-matrix interface is assumed, i.e. on composite cylinder assembly (CCA) and generalized self consistent scheme (GSCS), but extended with parameters for interface properties to investigate the effect of imperfect interface on composite properties. In the CCA model, concentric composite cylinders (fibres coated with matrix) of same fibre-matrix fractions but different, smaller and smaller, radius, are stacked together to form a composite. The microstructure is shown in Figure 5a. In the GSCS model, a fibre surrounded by matrix is embedded in a composite substrate with the effective properties of the composite, Figure 5b. In [55], where the effects of an imperfect interface on mechanical properties of composite materials are studied, both models are used in parallel and deliver the same results. The matrix is assumed to be isotropic while the fibres have anisotropic material properties. The interface is assumed to be elastic and is represented by three interface parameters as shown in Figure 5c. For derivation of the interface parameter $\mu_r$ only the GSCS model is used. In a following publication, the model was extended to cover linear viscoelastic properties of a fibre matrix interphase [56].

![Figure 5](image)

**Figure 5:** Fibre arrangement in (a) CCA-model: Fibres (black) coated with matrix (white), (b) GSCS-model: Fibre (black) coated with matrix (white) embedded in composite substrate (grey) and (c) elastic interface parameters $\mu_A$, $\mu_T$ and $\mu_R$ as described in [55].
Nairn [57] used shear-lag analysis of concentric cylinders to investigate effects of an elastic, imperfect interface. Data from fragmentation studies of a carbon fibre-epoxy system were used to exemplify how an imperfect interface parameter can be derived from experimental data with the shear-lag analysis derived. The shear-lag analysis presented in his work describes axial interface properties, compared to Hashin’s model which also includes interface parameters tangential and normal to the fibre surface. Wu et al [58] predicted the transverse shear modulus of a three phase system consisting of coated spheres or fibres embedded in a matrix. The inclusion-coating interface was assumed to be perfect, while the coating-matrix interface was imperfect and elastic, like a spring layer of vanishing thickness. Their results showed the same trends as presented by Hashin [55] for the investigated transverse shear modulus of the fibre composite.

1.2 WOOD FIBRES AND THEIR HYGROEXPANSION

The physical and mechanical properties of wood fibres, e.g. hygroexpansion, stiffness and strength, depend on various factors like species, growth conditions and pulping process. The influence of these parameters on wood fibre properties is briefly described in the following.

1.2.1 Structure of wood fibres

Both softwoods (e.g. pine and spruce) and hardwoods (e.g. birch) are used in the Swedish pulp and paper industry. Softwoods generally offer long (~2-3 mm) and flexible fibres, while hardwood fibres are shorter (~1 mm) and stiffer. Differences are also seen between fibres grown during springtime (earlywood) and during the summer (latewood), where earlywood fibres are larger and have thinner cell walls than the denser latewood fibres [31]. The properties of pulp fibres are highly dependent on the method of fibre extraction, i.e. the pulping process. In chemical pulp, wood is separated into fibres chemically, while mechanical treatment, i.e. grinding, is used to extract the fibres in mechanical pulp. Mechanical pulping renders fibres that are stiff and straight with high bending stiffness. The fibres are short and thick and less collapsed compared to fibres from chemical pulps and contain relatively high amounts of lignin and hemicellulose. Mechanical pulp is used for e.g. newsprint, while the slender and flexible fibres from chemical pulping processes are used in e.g. copy paper [31].

Wood fibres are hollow and have a layered structure with primary and secondary walls, as illustrated in Figure 6a. The primary wall is thin compared to the secondary walls and consists to a large extent of lignin. In the secondary walls, lignin and hemicellulose are reinforced by cellulose fibrils. In the thick S2 layer the fibril orientation is close to parallel to the fibre axis, while fibrils in the thinner S1 and S3 layers have a clear off-axis orientation [59]. The inner (S3) layer surrounds lumen, the hollow centre of
As a consequence of the fibril orientation in the layered structure, the wood fibres are anisotropic with different properties in longitudinal, radial and tangential directions, illustrated in Figure 6b. The off-axis orientation of the microfibrils gives a spiral structure, with a resulting coupling between twist and extension.

Several authors have investigated the longitudinal Young's modulus of longer natural fibres, i.e. hemp and flax, by single fibre tests [29]. Transverse Young’s modulus and shear properties are not available in the literature to the same extent due to the difficulties in experimental determination of these properties. The values found are often obtained through studies of the natural-fibre composite or of the cell wall structure of the fibre [29, 61]. Wood fibres are small also in the longitudinal direction, which makes single-fibre measurements to determine longitudinal Young’s modulus cumbersome and sometimes uncertain [62]. Transverse properties are found in similar manners as for longer natural fibres, with studies of the fibre cell wall. In the determination of transverse properties, wood fibres are often regarded as transversely isotropic and the anisotropy ratio, i.e. the ratio of longitudinal to transverse Young’s modulus is determined [62, 63]. The properties in radial direction are hence assumed to be equal to those in the tangential direction. This simplification, employed in Papers A, C and F, is considered reasonable, since differences between properties in radial and tangential directions are small compared to the properties in the longitudinal direction. For small microfibril angles and for constrained fibres in composites, the helical structure and twist-extension coupling is generally ignored.
1.2.2 Wood fibre hygroexpansion

Hygroexpansion and moisture induced softening of the hydrophilic wood fibres are perhaps the most severe drawbacks of wood-fibre composites. Hygroexpansion of the fibres leads to deformation of the composite component and the stiffness and strength of the composite are decreased by the moist induced softening of the fibres. If hydrogen bonds between fibres and in fibre-matrix interfaces are reduced by the absorbed moisture, stiffness and strength are further diminished and in the presence of moisture, cellulose is also more susceptible to microbial attacks.

In papermaking, where additives are used to increase the wet-strength of paper, focus has been on preserving the fibre-fibre bonds. In the field of natural fibre composites, efforts have been made to reduce the hygroexpansion of the reinforcing material, and different approaches have been suggested. Treatment with acetylation is one of the most studied methods presented in the literature. It is reported to increase dimensional stability and decrease hygroexpansion of wood and natural-fibre based materials. Unfortunately, stiffness and strength are also decreased by the acetylation treatment [64-68]. Various cross-linking reactions have been used to improve the dimension stability and wet-strength of paper. According to the review by Caulfield and Weatherwax [69], formaldehyde has been of primary interest for fibre cross-linking since it has been reported to increase wet-strength and decrease moisture sorption of paper. Wood is however not completely stabilized by the reaction with formaldehyde according to the later review by Bledzki et al. [17]. Instead of formaldehyde, the effects of cross linking with butanetetracarboxylic acid (BTCA) on hygroexpansion of wood-fibres for composite applications are investigated in Paper E.

The hygroexpansion of wood fibres shares a common trait with fibre-matrix interface properties in that it is difficult to measure the effects through single-fibre tests. The small dimensions, natural variability and the anisotropic swelling of the fibre make single-fibre tests of fibre swelling cumbersome. Attempts to assess information of wood fibre swelling from the swelling of paper sheets have been made [70, 71]. This is, as discussed by the authors, not without difficulty, since paper is a heterogeneous and porous material. Some of these difficulties can be avoided if back-calculation is performed from well consolidated wood-fibre composites instead of paper sheets, which is the approach employed in Paper F. Modelling of hygroexpansion of the fibre cell wall, where hygroexpansion of the constituents cellulose, lignin and hemi-cellulose are used as input parameters, is an alternative way to asses information about wood-fibre hygroexpansion [72].
Similar to the mechanical properties, the hygroexpansion and moisture-induced deformation of fibres is complex due to the complex structure of the wood fibre; the hygroexpansion in radial, tangential and longitudinal directions vary and the fibres tend to twist since the fibrils are not parallel to the fibre axis [73]. The anisotropic swelling also leads to other changes of the form of the fibre. Transverse swelling of the fibre cell wall straightens buckled fibres making them longer and opens the fibre cross section from the elliptic or rectangular cross section of dry fibres, to the more circular cross section. These form changes are not directly linked to the fibre coefficient of hygroexpansion. In the case of fibre elongation, generally only a minor part of the elongation is caused by actual elongation of the fibre correlated to the longitudinal coefficient of hygroexpansion. The main part of the elongation is explained by straightening of the buckled fibre, initiated by the radial swelling of the fibre cell wall [74]. When determining the coefficients of hygroexpansion, which are material parameters, it is therefore important to separate actual swelling of the fibre and other geometrical effects that might contribute to changing the form of the fibre.
2 MATERIALS AND METHODS
In this thesis, interface properties in wood-fibre composites are studied and new methods for interface characterisation are investigated. The aim has been to avoid the cumbersome single-fibre tests and coarse DMA methods for the benefit of simpler and more straightforward methods, including more suitable micromechanical models with a balance of simplicity and accuracy. Hygroexpansion of wood-fibres and wood-fibre composites have been studied with the same goals: to simplify measurements by avoiding single fibre tests. Single-fibre tests give, however, well-defined, local measurements, and could be used to validate macroscopic characterization techniques.

2.1 MATERIALS AND MANUFACTURING
Three different polymers were used as matrix material. The composites investigated in the interface studies in Papers A, B and C were wood-fibre reinforced polylactide (PLA). Polylactide is a thermoplastic and biodegradable polymer derived from starch-rich plants like maize and wheat. Polylactide is rather brittle, but the adhesion to wood and natural fibres is good compared to more hydrophobic, non-polar polymers, and several studies have shown that it is suitable as a matrix material in natural-fibre composites [3, 75]. In Paper E, where hygroexpansion of wood fibres and wood-fibre composites was investigated, both polylactide and polypropylene (PP) were used as matrix material. Polypropylene is petroleum-based, inexpensive and commonly used in both natural- and synthetic-fibre composites. Composites with a polylactide matrix, polypropylene matrix and a mixed polylactide-polypropylene matrix (50 wt % of each) were used, but only well consolidated composites with a polylactide matrix were further investigated in Paper F. In the study of stress transfer in Paper D, the matrix material was an epoxy vinyl ester. Like polypropylene, epoxy vinyl ester is petroleum-based and commonly used in composite applications, but contrary to polylactide and polypropylene that are thermoplastic, epoxy vinyl ester is a thermoset polymer.

The reinforcing wood fibres were softwood pulp fibres (fully bleached in Papers A-C and unbleached in Paper D) or bleached birch pulp from industrial pulp (Papers E and F). The fibre treatment studied was butane-tetracarboxylic acid (BTCA) modification. BTCA modification prevents the fibres from swelling by cross-linking of the hydrogen groups in the fibre cell wall [76]. Both modified and untreated reference fibres were used for composite manufacturing and the results were compared.

The thermoplastic polymers, polylactide and polypropylene, were delivered as short fibres. Since both matrix material and reinforcement - wood fibres -
were delivered as fibres, a wet-forming technique much similar to the method used to make paper sheets for laboratory investigations was applied: Wood and polymer fibres are mixed in water and when the water is removed, a commingled wood- and polymer-fibre sheet is formed. Sheets with even, in-plane fibre distribution (Papers A, C, E and F) and oriented fibre sheets (for the FT-IR study in Paper B) were produced. After drying, the sheets were hot pressed, which caused the polymer fibres to melt and form a void-free matrix. The sheets were pressed individually in Papers A, B and C to obtain thin composites, while in Papers E and F sheets were stacked before pressing to render thicker composite plates.

In Paper D, where a thermoset resin was used, a resin transfer moulding (RTM) was used to manufacture the composites. Wood-fibre sheets were formed and vacuum suction was used to remove air and fill the sheets with the resin. The filled sheets were then cured in an oven to solidify the resin, and thereby stiff composites were prepared. Both RTM and hot-press moulding result in composites with slender fibres, enabling improved mechanical properties as compared to injection-moulded wood-fibre composites, where the fibre length is degraded in the severe shear flow during processing.

2.2 EXPERIMENTAL CHARACTERISATION

Experimental work was performed to study fibre-matrix interface properties and wood-fibre hygroexpansion. Some of the collected data was used as input to the modelling approaches for interface characterization and determination of wood-fibre coefficient of hygroexpansion, as described in section “2.3 Modelling tools” and presented in detail in Papers A, C and F. The subjects of interest for experimental characterisation in this thesis are given below, together with experimental methods used.

2.2.1 Material characterisation

Polylactide, the most employed matrix in this thesis, is a semi-crystalline polymer and its mechanical properties depend on the degree of crystallinity. When mechanical properties of polylactide film are used as input in the modelling sections, and the predicted results are compared to measured composite data, it is important to establish whether the degree of crystallinity of polylactide film and of composite samples are the same. Therefore, the crystallinity of pure polylactide film and composite samples (of the type studied in Papers A-C) was studied through DSC measurements.

To evaluate the effect of BTCA modification on water absorption of wood fibres the water retention value of modified and untreated reference fibres was determined. A standardized centrifuging method was used; after
centrifuging, the pulp is weighed, dried and reweighed. The water content corresponds to the weight loss and the water retention value is expressed as a percentage of water content to the dry weight of the sample [77].

A microscopy survey was performed to study the microstructure of selected composite materials and the presence of voids, cracks, fibre agglomeration and filled lumens was observed. Small samples were embedded in epoxy and the cross sections were gently polished before the electron microscope (ESEM) examination.

2.2.2 Mechanical properties

Linear viscoelastic mechanical properties, i.e. Young’s modulus, $E$, and loss factor, $\tan \delta$, of the composites, wood-fibre sheets and pure polylactide film used in Papers A and C were determined with dynamic mechanical analyzer (DMA). Cyclic testing was performed in dry and humid conditions to study the influence of moisture on the materials and generate data needed for the micromechanical models presented for interface characterization. A smaller DMA-equipment connected to the FT-IR was used to generate Young’s modulus and $\tan \delta$ of the composite, wood-fibre and neat polylactide samples tested in Paper B.

In Paper D the stiffness and strength of composite plates, wood-fibre sheets and pure resin samples were determined with quasistatic tensile tests. In Paper E, where the effects of fibre treatment, choice of matrix and fibre fraction were studied, three-point-bending tests were performed to compare stiffness and strength of the samples. Tensile tests are preferable due to a uniform and uniaxial stress field in the gauge section, although the flexural tests were chosen for practical reasons in cases where the manufactured composite plates were too small to machine standard dog-bone specimens.

2.2.3 Bonds and bond strength

In Paper B, thin wood-fibre polylactide composites were studied with Fourier Transform Infrared technique (FT-IR). Stretching and bending of molecular bonds in cellulose and polylactide could be observed as the samples were subjected to cyclic loading. Comparison of these motions when samples were tested under dry and humid conditions was performed. Observed differences could be interpreted in terms of fibre-matrix stress transfer-ability. FT-IR spectroscopy was used in Paper A and in Paper B, where a more detailed evaluation of the technique was performed.

In Paper D, the degree of consolidation in wood-fibre sheets was studied with $\gamma$-strength tests, i.e. strength in the out-of-plane direction, as described in section “1.1.1 Stress transfer in paper and board”. This was further studied with X-ray microtomography at beamline ID19 at the European
Synchrotron Radiation Facility (ESRF) in Grenoble, France. The tomograph gives three-dimensional (3D) images of the samples with high resolution (0.7 µm × 0.7 µm × 0.7 µm). Image analysis was performed over the tested volumes to obtain a measure of the fibre-fibre contact area.

The procedure of the image analysis method used is presented in Figures 7-8. In the tomography of a sample volume, Figure 7, wood fibres are clearly seen. Figure 8a shows a cross section of the sample volume. Fibre voxels are white and surrounding non-fibre voxels (matrix material) are black. The lumen voxels are then identified, grey colour in Figure 8b, and fibre contacts are identified by letting rays run through the image in the z-direction, i.e. thickness direction, of the sample. Any time a ray passes between two separate lumen areas without touching any non-fibre voxels, a contact is considered to be found.

![Figure 7: Wood-fibre composite sample studied by tomography.](image1)

![Figure 8: Illustration of image analysis procedure. (a) Cross section of wood-fibre composite sample. (b) A ray (dark grey) passes through identified lumens (grey) surrounded by fibre wall (white). Average fibre diameter ~30 µm.](image2)

For each x- and y-coordinate, a ray is computed and the total contact area is defined as the number of identified contacts for all rays. Similarly, the total fibre area is defined as the total number of times the material changes between fibre wall and non-fibre along the rays. The measure of the relative amount of fibre contact area is obtained by dividing the total contact area by
the total fibre area. A more detailed description of the image analysis tools used is given in [78]. The method presented will not detect collapsed fibres, which leads to an underestimation of the fibre contact area, in particular for chemical pulps. Furthermore, if two fibres are close enough (0.7 µm), a fibre contact will be identified whether the fibres are bonded or in contact or neither. This may lead to an overestimation of the fibre contact area. The aim of the study was not, however, to attain an exact value of fibre contact area, but to compare the degree of bonding of different composite samples and the corresponding effect on composite stiffness and strength. For that purpose, the developed method should suffice.

2.2.4 Hygroexpansion and vapour and water sorption

Dynamic vapour sorption system (DVS) was used to study vapour absorption of the materials used in Papers A and C. The temperature was kept constant and the moisture content of small samples of composite, wood-fibre mat and PLA was determined. Every fifth hour the relative humidity in the test chamber was increased in 10 % relative humidity steps from the initial dry condition to the final 90 % relative humidity. The samples used for the studies of hygroexpansion in Papers E and F were too big for the DVS to be used. Instead, samples were dried in an oven to obtain the dry weight and kept in a sealed humidity chamber for the moisture absorption test. Weight and thickness of the samples was continuously and manually collected during the time of the test.

2.3 MODELLING TOOLS

In Papers A and C, the fibre-matrix interface was studied through DMA-measurements and linear viscoelastic material properties were used. In Paper F, where the wood fibre coefficient of hygroexpansion was determined, only the hygroelastic properties were considered. In all three papers (A, C and F), the modelling section is divided into micromechanical models and laminate analogy as illustrated in Figure 9.

![Figure 9: Illustration of the length scales used in the models.](image_url)
The micromechanical model links the mechanical properties of the constituents, fibres and matrix, and interface properties to the mechanical properties of a hypothetical, unidirectional composite lamella. Laminate analogy is then used to predict the mechanical properties of a composite material with any given in-plane fibre orientation. This is made through a summation of auxiliary, unidirectional lamellas that are added together to a composite of given fibre distribution using classic laminate theory.

The stress-strain correlation of purely elastic materials is given by Young’s modulus, \( E \). For viscoelastic materials, storage modulus, \( E' \), and loss modulus, \( E'' \), are used. Young’s modulus can then be expressed as a complex value, \( E' + iE'' \), where the storage modulus corresponds to the real component and the loss modulus to the imaginary component,

\[
E' = E' + iE''.
\]

The energy loss in a material can be expressed as the loss angle, \( \delta \). The loss angle is defined as the time delay between stress and strain, and is related to the loss modulus and the storage modulus through

\[
\tan \delta = \frac{E''}{E'}
\]

and commonly referred to as the loss factor, \( \eta \),

\[
\eta = \tan \delta.
\]

Micromechanical models, as Halpin-Tsai’s [24] and Hashin’s [55] models used in this study, are generally derived for purely elastic materials. The correspondence principle, however, can easily be used to extend the validation of the equations to the linear viscoelastic case [79-81]. The rule of mixtures, used to predict the longitudinal Young’s modulus of a unidirectional composite lamina, \( E_L \), is transformed from

\[
E_L = V_f E_f + V_m E_m
\]

where \( E \) is Young’s modulus, \( V \) is volume fraction, ‘f’ and ‘m’ denote fibre and matrix properties, respectively, and \( f1 \) means in axial fibre direction, coinciding with the longitudinal direction, \( L \), of the composite lamella, to

\[
E_L^* = V_f E_f^* + V_m E_m^*
\]

Micromechanical models, as Halpin-Tsai’s [24] and Hashin’s [55] models used in this study, are generally derived for purely elastic materials. The correspondence principle, however, can easily be used to extend the validation of the equations to the linear viscoelastic case [79-81]. The rule of mixtures, used to predict the longitudinal Young’s modulus of a unidirectional composite lamina, \( E_L \), is transformed from

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\[
E_L^* = V_f E_f^* + V_m E_m^*
\]
where $E_1^\ast$, $E_\|^\ast$ and $E_m^\ast$ contain both storage (real) and loss (imaginary) components according to Equation 1.

Many micromechanical models, as Halpin-Tsai’s model [24] used in Paper A, assume perfect interface between fibres and matrix. A comparison between predicted energy loss in a material and measured actual energy loss, when a material with imperfect interface is subjected to loading, could hence give indications on the efficiency of the interface. An imperfect interface results in stress redistribution from fibre to matrix, which affects the dissipation during cyclic loading.

In Paper A, where this model was used for the transverse and shear properties at the level of a unidirectional ply, viscoelastic material properties of wood-fibres and polylactide were used as input data. Using a laminate analogy, the damping of a wood-fibre-polylactide composite with perfect interface could hence be predicted by the model. The predicted damping could then be compared to experimentally determined data, and since tests were performed under both dry and humid conditions, the effect of humidity on energy losses in the material could be analyzed.

As in Paper A, where composite damping and Young’s modulus are predicted, micromechanical models are commonly used to predict composite properties from known properties of its constituents. Due to the difficulty of determining wood-fibre and interface properties, it can be useful to employ the micromechanical models the other way around, starting with experimentally determined composite properties and then predicting contributing properties of wood-fibres or interface. This approach was used in Paper C, where Hashin’s micromechanical model for imperfect elastic interface [55] was used in the search for a quantitative measure of the interface. The model with an elastic interface was chosen over the viscoelastic interphase, since the fibre-matrix interface in the investigated material is considered to be thin and hence the damping effects of the interface should be small. Hashin’s model includes three interface parameters $\mu_A$, $\mu_T$, and $\mu_R$, as discussed earlier and illustrated in Figure 5. The relations between the interface parameters are not known. To reduce the number of unknowns, the three interface parameters were assumed to be equal and were replaced with one interface parameter, $\mu$.

In Paper F, a method similar to the back-calculation approach used for interface studies in Paper C was utilized to determine the coefficient of hygroexpansion of untreated reference fibres and BTCA modified fibres. The fibres were used as reinforcement of a polylactide matrix and the thickness swelling of the composites was monitored as the samples were allowed to reach equilibrium in a humid environment. The out-of-plane
The micromechanical model was constituted by Hashin’s [27, 82] thermoelastic expressions, here used to describe hygroelastic behaviour. A difference between temperature and moisture content is that the former generally is known and constant in the constituent materials at equilibrium conditions, while moisture content in the constituents is most likely to differ depending on the hydrophilicity of the materials. Therefore, the relative humidity, which, similar to temperature, is constant, was used to describe hygroexpansion.

Hashin’s model was employed by McCartney and Kelly [28], who studied the out-of-plane thermal expansion of composite laminates. They found that constraints from neighbouring plies had a significant effect on the out-of-plane thermal expansion. The influence of in-plane stresses in the composite laminate should therefore be addressed when the out-of-plane hygroexpansion is considered. As in the previous studies, the fibres were assumed to be transversely isotropic in stiffness, but also proportionally in hygroexpansion, which was supported by the results of Schulgasser [83]. Only linear elastic and linear hygroexpansion properties were considered, and the fibre-matrix interface was assumed to be perfect.

The material parameters required as input to the micromechanical models, and hence to determine interface properties or fibre hygroexpansion through back-calculation, are the complex Young’s modulus of composite material, wood fibres and polylactide, the fibre volume fraction and distribution and the anisotropy ratio of the fibres. For the studies of hygroexpansion, the coefficients of hygroexpansion of composites and polylactide are required. For the composite material and the isotropic polylactide, these properties are simply determined through dynamic mechanical analysis and thickness measurements. Since direct measurements of wood fibres are difficult and uncertain, the properties of the wood fibres were determined through measurements on well-consolidated wood-fibre mats and back-calculation from laminate analogy and micromechanical models. The approach, derived by Neagu et al. [71], was similar to procedures used in Papers C and F.

In Papers C and F, as well as in the wood-fibre stiffness back-calculation, the summation over the lamellas in the laminate analogy was made with the
assumption of constant in-plane strain and zero stress in the out-of-plane direction. In Paper A, the summation was performed under constant stress over the lamellas. In paper technology, both methods are used; uniform stress tends to underestimate the stiffness of paper, while the assumption of uniform strain leads to an overestimation of the stiffness [84]. The latter is considered the most accurate for these dense composites, albeit the difference in predicted stiffness is not believed to be large.
3 RESULTS AND DISCUSSION

In this chapter, the results from experimental work and modelling tools will be presented and discussed. The quality of the manufactured composites was evaluated with scanning electron microscope (ESEM). Observations from the microscopy survey correlated well with trends seen in Young’s modulus of the samples, as discussed in “3.1.2 Mechanical properties”. The importance of an efficient interface is emphasized and results from the dynamic FT-IR spectroscopy are presented in “3.1.3 Bonds and bond strength”, followed by results from the experimental work on hygroexpansion and moisture absorption. Finally, the results obtained from the modelling tools, where interface properties and wood-fibre hygroexpansion were investigated, are presented and discussed.

3.1 MATERIALS AND EXPERIMENTAL CHARACTERISATION

3.1.1 Materials and manufacturing

In this work, four different polymer systems were used as matrix material: polylactide, polypropylene, a mixture of polylactide and polypropylene and epoxy vinyl ester. As reinforcement, bleached fibres (Papers A, B, C, E, F) and unbleached softwood fibres (Paper D), are used. In Papers E and F the effects of BTCA modification on wood-fibre hygroexpansion and absorption was studied. The BTCA modification was considered successful, with lower water retention value and less moisture absorption than the reference fibres. The weight gain due to moisture sorption was 27 % for the untreated fibres and 18 % for the BTCA modified fibres. A negative effect however, was an increased tendency to form aggregates in the composite manufacturing process. This was a problem particularly in combination with the polypropylene matrix, resulting in poor adhesion and void-filled composites.

Differential scanning calorimetry (DSC) was performed on polylactide composites (of the type studied in Papers A-C) and on pure polylactide samples to investigate whether the degree of crystallinity was the same in the polylactide films studied to generate input data to the micromechanical models (used in Papers A and C) as in the composite samples. No difference could be seen between the polylactide film and the composite sample, and the crystallinity was determined to approximately 43 %. Polylactide has previously been suggested for wood/natural-fibre composite applications [3, 85] and its affinity to wood fibres has been reported to be high. The adhesion of the hydrophobic polypropylene to wood-fibre is not as good as that of the hydrophilic polylactide, which was supported by the microscopy surveys performed in this thesis.
Wood-fibre composites: Stress transfer and hygroexpansion

Figure 10a: Micrograph of polylactide composite with 70% reference fibres. The darker regions are voids.

Figure 10b: Micrograph of polylactide-polypropylene composite with 40% reference fibres. Fibres are coated with polylactide (light grey); only few fibres are surrounded by polypropylene (dark grey).

Figure 10c: Micrograph of epoxy composite with 18% (volume fraction) fibres. High affinity between epoxy and fibres, resin filled lumens and no voids are seen.
Micrographs of some of the different materials studied are presented in Figure 10a-c. The wetting of the polylactide matrix on the wood fibres was good as was fibre dispersion. Lumens of uncollapsed, thick-walled latewood fibres were filled. However, for high fibre fractions, as in Figure 10a where approximately 70% reference fibres were used, a substantial amount of voids are seen (since the density of polylactide, approximately 1.3 g/cm$^3$, (data provided from manufacturer), is close to that of the wood fibres 1.5 g/cm$^3$ [86], weight and volume fractions of fibres in polylactide composites are nearly the same). With only 30% matrix material, larger fibre regions remained unimpregnated, which resulted in cracks and air pockets.

When polypropylene was used as matrix material, the tendency for the fibres to agglomerate was increased and cracks between fibres and matrix were seen also at lower fibre contents. The result of the 50/50 mixture of polylactide and polypropylene is shown in Figure 10b, where the polylactide surrounds the fibres like a coating. This gave a material with fewer voids and cracks compared to the case where pure polypropylene was used. Similar results have been presented by Huda et al. [3] in their study of wood fibres in polylactide/polypropylene blends. The micrograph of one of the composites manufactured in Paper D shows that epoxy vinyl ester impregnation using RTM led to excellent wetting. Filled lumens were observed and no voids were seen, Figure 10c.

### 3.1.2 Mechanical properties

The mechanical properties of the composites studied showed clear improvement from polymer properties to composite properties when the polymers were reinforced, Figure 11.

![Figure 11: Young’s modulus of neat and reinforced polymers.](image)
This confirms that the manufacturing of the composites were successful and that fibres and matrix interact well. The trends from the microscopy survey can also be found in the stiffness of the composite samples. The wood-fibre reinforced epoxy resin (Figure 10c), where filled lumens but no voids or cracks could be observed, shows the highest improvement in Young’s modulus when fibres are added to the matrix. The Young’s modulus of polylactide is also considerably improved when reinforced with fibres (40 % fibres added to polylactide and polypropylene in Figure 11). When hydrophobic polypropylene was used, the composite showed less improvement of mechanical properties, especially for higher fibre contents and BTCA-modified fibres, which correlates well with the observations of agglomerated fibres and voids found e.g. in the micrograph in Figure 10b.

Mechanical testing was performed on the well consolidated, low fibre fraction polylactide composites. No effect of the BTCA modification could be seen. This is an advantage of BTCA modification as compared to e.g. acetylation, where mechanical properties have been reported to be impaired by the fibre treatment [65-68]. Although these results are promising, they should be treated with care, since few samples were used and the scatter was considerable. The highest modulus was found for composites with 40 % fibres, followed by those with 30 % fibres. The results for higher fibre fractions were lower due to insufficient wetting, in agreement with results presented by other authors [87].

### 3.1.3 Bonds and bond strength

The importance of good fibre-matrix adhesion was emphasized by the results in Paper D where effects on fibre-matrix interface and fibre-fibre bonds on composite properties were investigated. Wood-fibre mats with different levels of consolidation were manufactured through solvent exchange and subsequent pressing. Composites were then made by epoxy resin impregnation of the wood-fibre sheets, Table 1. The mechanical properties of composites and sheets were then determined. In paper mechanics, where the thickness of the sometimes fluffy low basis-weight samples is difficult to measure, the properties specific stiffness and strength are commonly used. These are commonly referred to as tensile stiffness index and tensile strength index in the paper research community. Such properties are also used in composite mechanics when thin composite plates are designed, and are simply defined as Young’s modulus and strength, respectively, divided by density. For the unimpregnated wood-fibre sheets, z-strength was determined, and both unimpregnated sheets and composites were studied by microtomography.
Table 1: Notation and characteristics of wood-fibre sheets and composites studied in Paper D.

<table>
<thead>
<tr>
<th>Description of sheet</th>
<th>Consolidation</th>
<th>Sheet</th>
<th>Appearance</th>
<th>Composite</th>
</tr>
</thead>
<tbody>
<tr>
<td>Iso-propanol, wet pressure: 0 bar</td>
<td>low</td>
<td>S1</td>
<td>Fluffy</td>
<td>C1</td>
</tr>
<tr>
<td>Iso-propanol, wet pressure: 0.5 bar</td>
<td>low</td>
<td>S2</td>
<td>Fluffy</td>
<td>C2</td>
</tr>
<tr>
<td>Iso-propanol, wet pressure: 2 x 0.5 bar</td>
<td>high</td>
<td>S3</td>
<td>Intermediate</td>
<td>C3</td>
</tr>
<tr>
<td>Reference</td>
<td>highest</td>
<td>S4</td>
<td>Thin</td>
<td>C4</td>
</tr>
</tbody>
</table>

The $\zeta$-strength tests confirmed that different levels of consolidation had been reached, Figure 12. After visual examination of the samples, this result was not surprising. Two of the sheets, here denoted S1 and S2, were fluffy with few fibre-to-fibre bonds, while the remaining two sheets, S3 and S4, were better consolidated.

Figure 12: $\zeta$-strength of sheets S1-S4.

The different amounts of fibre contacts in the sheets were visible in image analysis studies of the X-ray computer tomography images of the wood-fibre sheets. It was also suggested that these differences were maintained during the RTM process. This implies that composites with different amounts of fibre contacts between the reinforcing fibres had been manufactured. No effect of these fibre contacts could be seen on the composite properties. In Figure 13, C1 and C2 represent composites made from the fluffy sheets S1 and S2, respectively, and C3 and C4 are composites made from the better consolidated sheets S3 and S4, respectively.
The improved matrix properties suggest good stress transfer between the polymeric matrix and the reinforcing fibres, and since no influence of the amount of fibre contact could be seen, the fibre-fibre bonding is not considered to be an important stress transfer mechanism in this type of wood-fibre composites. Fibre bonds could play a more important role in wood-fibre composites, where the fibre fraction is high or where the fibre-matrix interface is weak. High fibre contents and poor fibre-matrix interface are however known to result in poor mechanical properties of wood-fibre composite materials, hence making them less interesting for structural applications.

Dynamic FT-IR spectra show small changes in spectral intensity induced by the straining of the tested sample. Instant and time delayed responses can be seen in the in-phase and out-of-phase spectra, respectively. The wave-numbers on the horizontal axis of the dynamic FT-IR spectra correspond to energy levels related with molecular vibrations characteristic for molecular bonds. Molecular bonds affected by the straining of the sample, i.e. bonds subjected to stretching or bending, can thereby be identified. The evaluation of dynamic FT-IR spectra of composite materials requires information of the absorbance of the constituent materials, i.e. the reinforcing cellulose in the wood fibres and polylactide, the matrix material. In-phase spectra of wood-fibre mat, polylactide film and composite material are presented in Figure 14.
Figure 14: In-phase spectra of wood-fibre mat, polylactide film and composite material at 0 % relative humidity.

The signals at 1447, 1389, 1369 and 1265 cm$^{-1}$ (corresponding to CH$_3$ asymmetric bend, CH$_3$ symmetric bend, CH and CH$_3$ symmetric bend and a combination band with contributions from CH bend and COC stretching, respectively) [88] were considered characteristic of the PLA matrix. The signals at the split peak 1423/1435, 1377 and 1323 cm$^{-1}$ (COH bend, CH bend and CH$_2$ bend, respectively) [89] were considered characteristic of the wood fibres. Previous studies by Hinterstoisser and Salmén have shown that it is mainly the cellulose in the wood fibres that contribute to the absorption at 1423/1435 cm$^{-1}$ and 1323 cm$^{-1}$ [90].

To visualize the relative response of the two constituents as moisture was induced, a normalization of the in-phase spectra was done with respect to the peak at 1435 cm$^{-1}$, reflecting the backbone deformation of cellulose [91]. The normalized in-phase spectra for the composite material at different levels of relative humidity are presented in Figure 15, where shifts from cellulose to polylactide is seen when the relative humidity increases. In the 1400 to 1450 cm$^{-1}$ region, where the dry and 60 % relative humidity spectra (light grey and grey lines) have their maxima at the cellulose peak 1435 cm$^{-1}$, the 80 and 90 % relative humidity spectra show responses from the polylactide peak at 1447 cm$^{-1}$. At dry conditions (light grey curve) the composite peak between 1350 and 1400 cm$^{-1}$ is dominated by the peak at 1377 cm$^{-1}$, which is characteristic for the fibre. However, when the relative humidity is increased (grey, dark grey and black lines), the peak at 1389 cm$^{-1}$, characteristic for polylactide, becomes visible as a shoulder. Furthermore, increased response is seen at wavenumber 1323 cm$^{-1}$, representing response...
from CH$_2$ bend in a side group in the cellulose molecule. This motion of the side-group is believed to be affected by the behaviour of the surrounding matrix rather than directly representative for the backbone movement of cellulose. Increased response is also seen in the region around 1265 cm$^{-1}$, where the main contribution origins from polylactide. The relative stress redistribution from the fibres to the matrix, as seen in the dynamic FT-IR spectra when humidity was increased, could be explained by reduced stress transfer ability as the interface absorbs moisture, and the matrix hence has to carry a larger part of the load.

![Dynamic FT-IR response](image)

**Figure 15:** In-phase spectra of composite material at different levels of relative humidity, italic for PLA peaks.

### 3.1.4 Hygroexpansion and vapour and water sorption

Effects of moisture on the interface properties of a wood-fibre/polylactide composite were analysed. To separate effects of moisture-induced softening and swelling from changes in interface properties, the influence of moisture on constituents and composite material were analyzed. Moisture absorption of wood-fibre mat, polylactide film and composite material is shown in Figure 16a, thickness swelling is presented for wood-fibre mat and composite material, Figure 16b. The thickness swelling of the polylactide film is not included since it was too small to measure using standard procedures.
Figure 16: Effects of moisture on (a) mass, (b) thickness, (c) Young’s modulus and loss factor.
Experimentally determined absorption of wood fibres and polylactide was used to predict composite mass change through the linear rule of mixture. The predicted data, 7.4%, was somewhat higher than the experimentally determined value, 6.7%. This small difference is more likely explained by material heterogeneity of the small composite samples than by constrained hygroexpansion of the wood fibres in the thin composite. The mass increase of polylactide was less than 1% when humidity was increased from dry conditions to 80% relative humidity. It was therefore not surprising that the thickness increase of polylactide film was small. The effects of moisture on Young’s modulus and loss factor of composite material are seen in Figure 16c.

The BTCA modification was considered successful at fibre level since both moisture absorption and water retention values were reduced for BTCA modified fibres. The modified fibres showed potential as reinforcement in polylactide composites, but were not compatible with polypropylene, resulting in poorly consolidated composites with high swelling. Figure 17 shows hygroexpansion of composite materials tested under humid and immersed conditions.

![Figure 17: Out-of-plane hygroexpansion for composites tested under humid conditions.](image)

The moisture induced swelling of polylactide and polylactide-polypropylene composites is reduced when BTCA modified fibres are used, most noticeable for higher fibre contents. For polypropylene composites, the effect of BTCA modified fibres is the opposite, with higher hygroexpansion than when the reference fibres are used. It is also seen that for reference fibres the choice of matrix is not crucial to moisture induced composite swelling, suggesting that the more effective wood-fibre/polylactide interface
does not prevent swelling of the composites. For composites where BTCA modified fibres are used, increased swelling is seen when polypropylene is used as matrix material. This could be explained by the increased agglomeration of BTCA modified fibres in polypropylene, leading to concentrations of the fibres and microcracks, and thereby also to high local strains.

The remaining thickness change after redrying of the samples follows the same trends, Figure 18. BTCA modification of fibres in polylactide composites leads to smaller residual out-of-plane strains, explained by less damage caused by the restrained swelling of the modified fibres. The change from polylactide to polypropylene when reference fibres were used led to an increased remaining thickness change, suggesting that a stronger interface could lead to reduced residual strains. This effect was also seen for BTCA modified fibres.

![Figure 18: Remaining thickness change after drying of moisture saturated samples.](image)

### 3.2 MODELLING TOOLS

In the first modelling approach in this thesis, Halpin-Tsai's micromechanical model for transverse and shear stiffness [24] was used, which assumes perfect interface between fibres and matrix. A laminate analogy was used to account for the in-plane fibre orientation distribution. Measurements were performed under dry and humid conditions on composites and their constituents, polylactide film and wood-fibre mat, and the predicted results of Young’s moduli and loss factors were compared to experimentally determined data for the composites. The elastic prediction, i.e. of Young’s
modulus, was close to the measured data (92% of the experimentally determined values for both dry and humid conditions), while the prediction of the specific damping was only \( \approx 65 \% \) of the measured data for both dry and humid conditions, Figure 19. The larger mismatch in damping prediction could be subscribed to losses induced by stress redistribution in the fibres and matrix. Moisture induced softening and swelling of both wood-fibre mat and composite material was observed and accounted for in the model, but no effects of moisture on the fibre-matrix interface could be proved.

![Graph showing Young’s modulus and tan(δ) for dry and humid conditions](image)

**Figure 19:** Measured and predicted Young’s modulus and tan(δ).

When Halpin-Tsai’s micromechanical model was replaced with Hashin’s micromechanical model for an imperfect interface [55], an interface parameter could be derived. In the materials investigated in this thesis, no fibre coatings or other surface modifications are used to improve the fibre-matrix interface. Interdiffusion from fibres to matrix is therefore not believed to contribute to the strength of fibre-matrix interface in these materials. Instead, chemical bonding through hydrogen bonds on fibre surfaces and in polylactide matrix and mechanical locking are considered more important to the interface strength and thereby in the stress transfer between matrix and fibres. The assumption of elastic interface is therefore deemed to be reasonable and the interface is hence described as a surface with vanishing thickness. As in the previous study, measured mechanical data of polylactide and wood-fibres were used as input to the model. The interface parameter included in the micromechanical model was adjusted to fit the predicted composite properties to experimentally determined data and thereby a value of the interface parameter was found. This was performed for samples tested under both dry and humid conditions.

Figure 20 shows predicted values of Young’s modulus (absolute value of the complex stiffness) for both dry (solid grey line) and humid (solid black line) conditions. As the humid prediction is lower than the dry prediction, the softening of the material is seen. For low and high values of the interface parameter, plateau values are seen, corresponding to interfacial debonding.
and strong interface, respectively. In the intermediate region, the predicted curves cross the experimentally determined values (dashed line, grey and black for dry and humid conditions, respectively). It is seen that the intersection occurs at the same value of the interface parameter for both dry and humid conditions. Hence, no effect of moisture on the interface could be seen, supporting the results from the previous study.

![Figure 20](image)

**Figure 20:** Predicted Young’s modulus (absolute value of the complex stiffness) as a function of the interface parameter and experimentally determined values under dry and humid conditions: Grey line for dry conditions and black line for humid conditions, predicted and measured values are represented by solid and dashed lines, respectively.

The results are hence complementary to those indicated by molecular effects characterized by dynamic FT-IR spectroscopy. The dynamic FT-IR data reflect changes on the molecular level, whereas the micromechanical model offers a link between micro- and macroscale. Molecular groups might locally have a different time-dependent behaviour than the polymer chain as a whole, which determines the macroscopic behaviour of the polymer.

A back-calculation strategy resembling the method used to back out the interface parameter was also used to determine the coefficient of hygroexpansion of single wood fibres. Again, Hashin’s micromechanical model [23, 82] was used. As in the previous studies, the properties of the known constituents were used as input to the model, and the sought parameter was identified by comparing the experimental macroscopic property with the corresponding predicted value. The wood-fibre hygroexpansion was thus determined by fitting the predicted value as closely as
possible to experimentally determined data of the out-of-plane hygro-expansion. This procedure was carried out for composites reinforced with BTCA modified and untreated reference fibres. It was found that the BTCA modification reduced the wood-fibre coefficient of hygroexpansion in the transverse direction from 0.28 to 0.12 % strain / % relative humidity.

The great advantage of modelling approaches, such as the methods developed in Papers A, C and F, is the simplicity of measurements and the fast and easy evaluation of tests. The models are however dependent on input data on mechanical properties of the composite constituents, fibres and matrix. Finding reliable input data and making realistic simplifications of the complex ultra- and microstructure of the fibre and composite, respectively, are hence considered to be the main issues. The lack of material data for different fibre types and feasible ways to experimentally determine fibre modulus and anisotropy make assumptions necessary. Reliable fibre data would increase the accuracy of the predicted results, and thereby of the sought interface parameter or coefficient of hygroexpansion.
4 SUMMARY OF PAPERS

PAPER A

Dynamic-mechanical properties of wood-fibre reinforced polylactide: Experimental characterization and micromechanical modelling

To avoid single-fibre tests for evaluation of interface properties in wood-fibre polymer composites, new methods for an indirect study of stress transfer and interface efficiency were investigated. Halpin-Tsai’s micromechanical model was used together with laminate analogy to predict mechanical properties, i.e. Young’s modulus and loss factor, of a wood-fibre composite material. The model, extended to viscoelastic material properties, assumes perfect interface between fibres and matrix. Energy loss in the cyclically loaded composite material caused by imperfections in the fibre-matrix interface will hence be neglected in the model. The predicted data were then compared to experimentally determined data. For the elastic property, Young’s modulus, the predicted and experimentally determined data correlated well, while the experimentally determined loss factor was significantly larger than the predicted one. Energy losses due to the imperfections of the interface are an important reason for this mismatch, which therefore can be used as a measure of interface efficiency. The study was performed under both dry and moist conditions. Softening of the materials was observed, as illustrated in Figure 21, where Young’s modulus in dry and humid conditions is presented. No effect of moisture on the fibre-matrix interface could be seen.

![Figure 21: Experimentally determined Young’s modulus in dry (0 % RH) and humid (80 % RH) conditions.](image)

Dynamic FT-IR (Fourier Transform Infrared) spectroscopy was tried to investigate changes of molecular losses pertaining to fibres and matrix. The results are promising, but the technique is not yet fully developed for stress transfer evaluation of composite materials.
PAPER B
Effects of moisture on dynamic mechanical properties of wood-fibre reinforced polylactide studied by dynamic FT-IR spectroscopy

Dynamic FT-IR (Fourier Transform Infrared) spectroscopy was evaluated for stress transfer investigations in a wood-fibre/polylactide composite material. With this technique, specific molecular bonds are detected and studied as the sample is subjected to cyclic load. The purpose of the study was to investigate whether this method could be used to evaluate stress transfer between fibres and matrix in a wood-fibre composite material. When a polymer sample is strained, the polymer structure is reoriented. This can be seen as an increase or decrease of absorbed IR radiation at a given energy. Stretching of covalent bonds is seen as frequency shifts and changes in width of the absorption maxima in the dynamic FT-IR spectra. Time-resolved spectra make it possible to obtain instant and time-delayed response of each individual molecular vibration. Dynamic FT-IR spectra can hence be divided into in-phase (instant) and out-of-phase (time-delayed) spectra. In-phase and out-of-phase spectra for the composite in the region 1500 to 1250 cm⁻¹ are shown in Figure 22. The dominance of the in-phase spectrum is a result of the elastic nature (with low loss factor) of the composite material under dry test conditions.

![Dynamic FT-IR spectra of composite material, in-phase response, 90° polarization.](image)

As the environment in the test chamber was changed from dry to humid, shifts from cellulose to polylactide peaks were seen. These could be explained by a weakening of the interface, resulting in decreased stress transfer, and thereby increased load carrying by the matrix. Softening in the transverse direction of the fibres could also result in a larger proportion of the load being carried by the matrix and thereby contributing to the observed shift.
PAPER C
Characterization of interfacial stress transfer ability by
dynamic mechanical analysis of cellulose fiber based
composite materials
The modelling approach from Paper A was developed from comparison of
predicted and measured energy loss to derivation of an interface parameter.
As in Paper A, a two step model including micromechanical model and
laminate theory was used. Hashin’s micromechanical model for elastic
interface was utilized and expanded to cover viscoelastic material properties
of reinforcing fibres and matrix through the correspondence principle. The
assumption of elastic interface was chosen over the viscoelastic interface,
since the interface in the composite studied is deemed to be thin and hence
lack significant damping abilities, Figure 23.

Figure 23: Illustration of fibre, interface and matrix imbedded in a composite substrate,
as described by the general self consistent scheme.

The wood-fibre reinforced polylactide studied in Paper A was used also in
this study. The mechanical properties of the composite and its constituents
were hence known from dynamic mechanical analysis, and the interface
parameter was thus the only unknown parameter. This interface parameter
could consequently be determined through back calculation. To further
investigate the effect of moisture on interface efficiency, the interface
parameter was derived for composite samples saturated in dry and humid
environments.

When interface parameters were chosen to fit the experimental data as
closely as possible, no difference between dry and humid states was seen.
This suggests that moisture absorption leads to softening and mechanical
dissipation in the hydrophilic wood fibres and thermoplastic matrix, rather
than in loss of interfacial stress transfer ability, confirming the findings in
Paper A.
POPER D
Role of fibre-fibre and fibre-matrix adhesion in stress transfer in composites made from resin-impregnated paper sheets

While stiffness and strength of paper are highly dependent on the degree of fibre-fibre bonding, stress is transferred through the fibre-matrix interface in synthetic composites. In wood-fibre composites, it is not obvious through which of these mechanisms stress is mainly transferred and how they contribute to stiffness and strength of the composite. Therefore, an investigation of the importance of fibre-fibre bonds and fibre-matrix interface in wood-fibre composites was performed.

Stiffness and strength of paper sheets and wood fibre composites with varying degree of fibre-fibre bonding were experimentally determined. The results showed that in contrast to paper properties, composite properties are not dependent on the degree of fibre-fibre bonding, Figure 24.

![Figure 24: Young's modulus or specific stiffness of neat matrix material and composite materials with different degree of fibre-fibre bonding.](image)

Image analysis tools were used to study the paper sheets and composites used in the study. The results suggest that the fibre network in the paper sheets remained intact through the manufacturing of the composites (resin transfer moulding, RTM, was used). The degree of fibre-to-fibre bonding is therefore believed to be the same in the investigated paper and composite samples. It is hence suggested that the stress in thermoset wood-fibre composites is mainly transferred from the matrix to the fibres and not from fibre to fibre.
PAPER E
Moisture uptake and hygroexpansion of wood fiber composite materials with polylactide and polypropylene matrix materials

The effects of the material parameters matrix composition, fibre volume fraction and fibre modification on material properties connected to moisture and swelling, i.e. hygroexpansion, moisture and water uptake and time of diffusion were investigated. A total of 24 different types of composites, see Table 1, were manufactured. The matrix material was polylactide, polypropylene or a 50-50 mixture of the two. Fibre loading was 30, 40, 60 or 70 %, and both butanetetracarboxylic acid (BTCA) modified fibres and untreated reference fibres were used. Changes in thickness and weight were monitored as the composites were allowed to reach equilibrium in a humid environment or immersed in water.

Table 2: Composition of samples tested in Paper E.

<table>
<thead>
<tr>
<th>Constituents</th>
<th>Fibre volume fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polymer</td>
<td>Wood fibre</td>
</tr>
<tr>
<td>PLA</td>
<td>Untreated</td>
</tr>
<tr>
<td>PLA</td>
<td>BTCA modified</td>
</tr>
<tr>
<td>PP</td>
<td>Untreated</td>
</tr>
<tr>
<td>PP</td>
<td>BTCA modified</td>
</tr>
<tr>
<td>PLA-PP</td>
<td>Untreated</td>
</tr>
<tr>
<td>PLA-PP</td>
<td>BTCA modified</td>
</tr>
</tbody>
</table>

The BTCA modification reduced the moisture uptake and the water retention value of the fibres by 33 and 69 %, respectively. Also for the results for polylactide and polylactide-polypropylene composites, where polylactide coated the wood fibres, the BTCA modification showed positive effects with a clear decrease of out-of-plane hygroexpansion. However, the treatment led to increased fibre agglomeration and poor results for polypropylene composites where increased swelling and damage accumulation were observed.
Contribution of wood fibre hygroexpansion to moisture induced thickness swelling of composite plates

One of the main drawbacks of wood fibre based composite materials is their tendency to swell due to moisture uptake in wet and moist environments. The main contribution to the hygroexpansion usually comes from the hydrophilic wood fibres. In order to compare and rank different types of fibres, quantification of the coefficient of hygroexpansion of wood fibres would be of interest. Single-fibre tests are cumbersome and tedious due to the miniscule dimensions of the fibres. Therefore, a method based on macroscopic measurements on wood-fibre composite materials is suggested. An inverse method to estimate the transverse coefficient of hygroexpansion of wood fibres is investigated. The outlined method is based on back-calculation from measured moisture induced thickness swelling of composite plates, through laminate theory and composite micromechanics derived by Hashin.

The transverse coefficient of hygroexpansion was determined for BTCA modified, cross linked fibres and untreated reference fibres. The two types of fibres were used for the manufacturing of thin composite plates. Poly-lactide, used as matrix material, was reinforced with 30 and 40 % wood fibres. Thickness swelling of the composite materials was monitored as composite samples were allowed to saturate in a humid environment. Samples were then re-dried and the reversible swelling was used as input to the model, since the model is limited to reversible hygroexpansion.

Table 3: Transverse coefficient of hygroexpansion for reference and BTCA modified fibres. Details on estimated values found in the literature are given in Paper F.

<table>
<thead>
<tr>
<th>Type</th>
<th>$\beta (\epsilon/RH)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reference (40 wt %)</td>
<td>0.28</td>
</tr>
<tr>
<td>BTCA (30 and 40 wt %)</td>
<td>0.12</td>
</tr>
<tr>
<td>Estimated from paper properties</td>
<td>0.2</td>
</tr>
<tr>
<td>Estimated by FEM simulations</td>
<td>0.12–0.13</td>
</tr>
<tr>
<td>Estimated from wood samples</td>
<td>0.21</td>
</tr>
</tbody>
</table>

The estimated values corresponded well with values found in the literature, Table 3. The transverse coefficient of hygroexpansion of the BTCA modified fibres is lower than the earlier reported values, suggesting that the modification was successful.
5 FUTURE WORK

The aim of this thesis has been to find more straightforward and simple methods to measure the fibre-matrix interface properties in wood-fibre composites as well as wood-fibre hygroexpansion. Micromechanical models have been used to characterise fibre-matrix interface efficiency and wood-fibre hygroexpansion from macroscopic measurements of composite materials. In the studies of interface efficiency, comparisons between dry and humid conditions were made. Further testing, e.g. determination of the interface parameters and wood-fibre coefficient of hygroexpansion of composites with treated fibres vs. composites with untreated fibres, would validate and demonstrate the practical use of the models. The models could then be used to rank surface modifications and fibre treatments and thereby serve as a useful tool in the development of wood-fibre composite materials. The proposed methods should be compared with other microscopic methods to characterize fibre hygroexpansion and interfacial stress-transfer, to validate whether the macroscopic methods could be used not only for ranking different materials, but also in an absolute quantitative sense. The next step would be to put the methods into practical use in more rationalized materials development.

Dynamic FT-IR was used to study stress transfer at the molecular level as thin composite samples were subjected to cyclic loading. Interesting differences between dry and humid test conditions were seen. Shifts from reinforcing cellulose to the matrix were observed and could be interpreted as stress redistribution from fibres to matrix, possibly explained by a weakening of the interface and the constituents as moisture was introduced to the system. The scatter in e.g. molecular loss factor was however, considerable, and the method remained qualitative. A quantification of the dynamic FT-IR technique for interface characterisation would significantly simplify the evaluation of stress transfer ability in wood-fibre composite materials.
6 LITERATURE


60 Brändström, J., (2002), Morphology of Norway Spruce tracheids with emphasis on cell wall organisation, Doctoral thesis, Department of Wood Science, SLU, Uppsala


Wood-fibre composites: Stress transfer and hygroexpansion


