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Full Set of Elastic Constants of Spruce Wood Cell Walls Determined by Nanoindentation

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In the last years Nanoindentation (NI) has become a frequently used tool in material science. Since Wimmer et al. (1997) introduced NI in wood science for the characterisation of micro-mechanical properties, the validity of the obtained data and their significance is a matter of debate. As NI theory was initially developed exclusively for homogenous and isotropic materials, the interpretation of results from heavily anisotropic wood cell walls is not straightforward. Therefore the indentation modulus M (also called reduced modulus) typically determined by NI-tests is not comparable to the longitudinal elastic modulus obtained with other experimental techniques. As a consequence, NI on wooden cell walls has been mainly used for comparative purposes. In order to overcome this limitation, we present a new approach capable of identifying the stiffness tensor components of the secondary cell wall S2 for quantitative purposes.

The indentation modulus obtained by NI is a product of the stiffness tensor components. Under the assumption of transverse isotropy for the wood cell wall, it depends on five elastic parameters and also on the angle between the direction of indentation and the longitudinal cellulose microfibril axis. A model built by Jäger et al. (2010) was applied in order to draw a link between the indentation modulus M and the stiffness tensor. In this manner, the unknown elastic parameters can be back-calculated via minimization of the error between the modulus predicted by the model and the modulus measured in NI experiments. In order to find an explicit solution, NI tests have to be performed in at least five different angles between load direction and micro fibril orientation. The best solution of the algorithm delivered appropriate values for all elastic constants of the wooden cell wall.

Keywords:
Nanoindentation, mechanical properties, wood, cell wall, analytical modelling, stiffness tensor

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Introduction

In their COST action report Salmen and Burgert (2009) concluded that there is an increasing demand for a more detailed knowledge regarding structure/property relations at micro- or nanostructural level to improve utilisation and manufacture of wood materials. Mechanical properties in particular are not easy to determine at the microscopic level, particularly if all stiffness tensor components are of interest. For wood a variety of approaches was made to determine experimentally at least some of the different mechanical properties at the wood cell wall level.

Tensile tests on small specimens were one of the first approaches (Cowdrey and Preston, 1966) to estimate the *longitudinal* modulus of elasticity of the cell wall material by considering the actual density of the tissue and the assumed density of the wood substrate (Stamm, 1964) of 1,46g/cm³. Bergander and Salmen (2000) presented one of the few studies determining the elastic modulus of the fibre wall also in *transverse* direction. As tissue slices have their characteristic contact between the adjacent fibres Salmen and Burgert (2009) concluded that measurements on *individual fibres* are preferable. To this day a couple of research groups (Burgert et al., 2005; Eichhorn et al., 2001; Jayne, 1959; Mark and Gillis, 1973; Mott et al., 1995; Page et al., 1972) put a lot of effort in improving the technique and described its strengths and weaknesses. Above all there is still the fact that the fibres have to be isolated either chemically or mechanically (Burgert et al., 2005). Due to this necessary process step the structural integrity of fibres is often affected to different degrees. In a recent study Eder et al. (2009) concluded that single fibre testing is not a direct measurement of cell wall properties, since also cell geometry and size influence the material response determined by such experiments.

A new approach for determining the elastic modulus of a spruce cell wall was shown by Orso et al. (2006). They performed a *bending experiment* on a cantilever manufactured out of an individual cell wall with the help of a focussed ion beam (FIB). Uni-axial *micro compression tests* (Zhang et al., 2010) were recently described in literature to access cell wall properties. In the study mentioned, only yield stress and compressive strength were evaluated. Most of the approaches presented above consider properties in one anatomical direction only.

Nanoindentation is another promising technique applied on wood for the first time by Wimmer et al. (1997) to access the material behavior of the secondary cell wall. Since 1997 a couple of studies (Gindl et al., 2004; Gindl and Schoberl, 2004; Jäger et al., 2010; Jakes et al., 2009; Konnerth et al., 2009; Lee et al., 2006; Tze et al., 2007; Wang et al., 2006; Zickler et al., 2006) was done to develop the technique and to refine the understanding of acquired results. Up to now the evaluation of results on the anisotropic material wood is based in most cases on the procedure described by Oliver and Pharr (1992), which was developed for homogenous and isotropic materials. Consequently the obtained indentation modulus is not comparable with the longitudinal modulus of the wood cell wall. Using this procedure only qualitative results may be acquired, which can be used for comparative purposes only. In the following we show a procedure to identify the unknown parameters of the stiffness tensor (E_l, E_t, G_{tl}) based on nanoindentation measurements, assuming transverse isotropy for the wood cell wall.

Materials and Methods

Three steps are required for the identification of elastic properties of the wood secondary cell wall material by means of nanoindentation tests. In the first step, nanoindentation tests on wood cell walls need to be performed for different indentation angles, i.e., the angle between indentation direction and the microfibril orientation. From this experimental procedure the indentation modulus is achieved as a function of the indentation angle. In order to determine elastic material constants, a model developed by Jäger at al. (2010) relating the indentation moduli to the elastic material constants of the wood cell wall material is applied, which constitutes the second step. Finally, the unknown elastic parameters are determined in the third step by means of a parameter identification procedure.

Nanoindentation test

A Spruce wood (picea abies) specimen with microfibril orientation characterized with a SilviScan-3^{TM} system (http://www.csiro.au/science/SilviScan.html),(Evans et al., 1999) was taken for micromechanical analyses. One single latewood band (Fig. 1, A) with a tangential extension of app. 2mm and a longitudinal extension of app. 7 mm was selected and subdivided in five different specimens. The specimens were prepared according to the procedure described in detail by Konnerth et al. (2008). After embedding the specimens were rotated around the tangential anatomical axis by the angle α (Fig. 2) in order to achieve five specimens with different fiber to indentation axis orientations. Considering the MFA of the latewood band these inclinations resulted in ten different microfibril to indentation axis orientations ranging from 0° to 90°. Additionally an unintended but existing inclination β (Fig.2) was considered. Both fiber inclinations, α and β, were determined by incident light microscopy for each specimen after final preparation.

Micromechanical experiments were performed on a Hysitron TriboIndenter system (Hysitron Inc., Minneapolis, USA) equipped with a three-sided pyramid diamond indenter tip (Berkovich type). In order to minimize the variation in (micro-) mechanical properties due to anatomical differences of the tested tissues, only cells within the last seven latewood cell rows (Fig. 1, B) of the single late wood band where tested, which were all located in direct proximity. One indent was performed in each radial and tangential latewood cell wall (Fig 1, C) resulting in 13 to 16 indents per fibril orientation. Indents were performed in load-controlled mode using a step-load function with partial unloading after 5 different peak loads (maximum load 500µN). A load function with partial unloading was selected in order to monitor possible influences of specimen compliance (Jakes et al., 2009). In any event, the evaluated indents where kept small enough to keep plastic deformation zones distant from the interface to the embedding material.

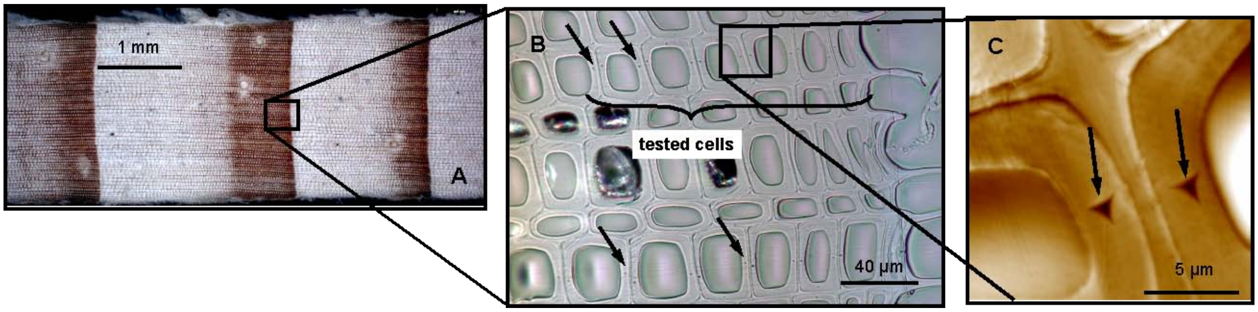

Fig. 1. Spruce wood sample A as source for SilviScan measurments. Incident light microscopy image B shows tested tracheids in the last cell rows of the late wood band with 45° inclination around the anatomical tangential axis. C scanning probe microscopy image generated with the indenter tip as used for positioning prior to testing and for control of accurate positioning after the indentation. Indent positions marked with black arrow heads.

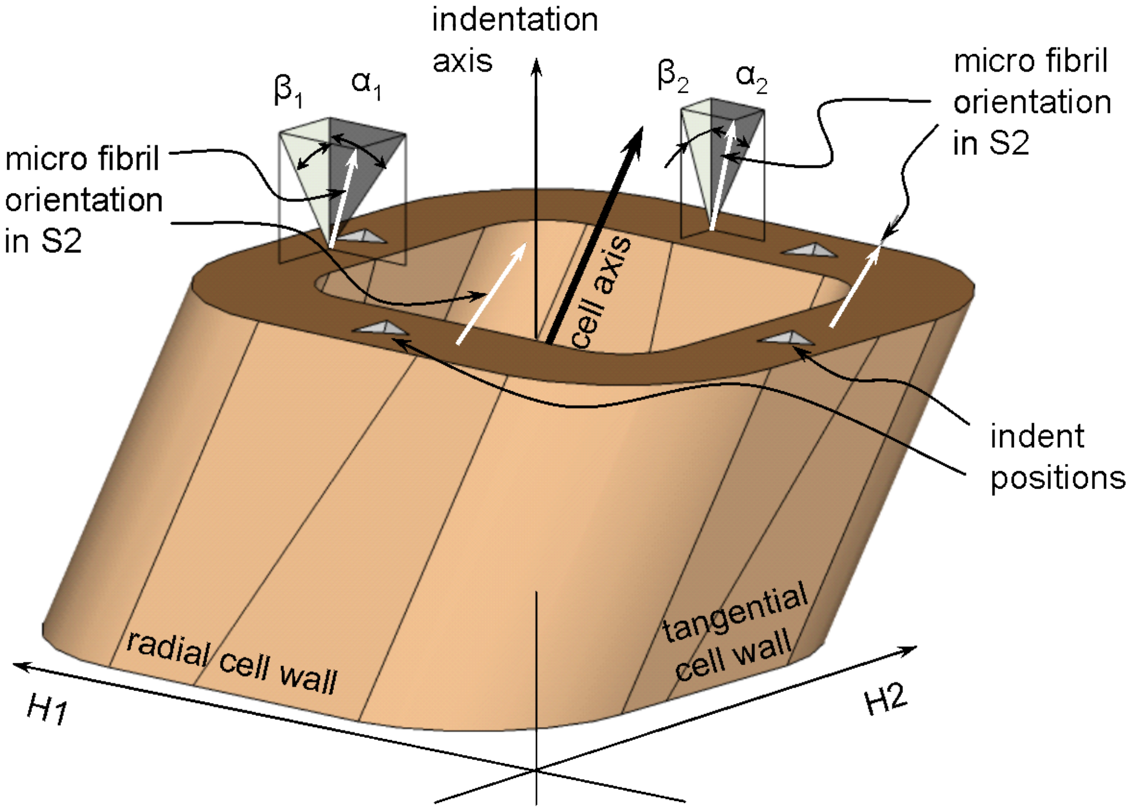

Fig. 2. Overview of indentation test design in wood cells inclined in two directions (α and β). H1 and H2 axes in horizontal plane, α inclination around H2 axis; β inclination around H1 axis.

Model for orthotropic nanoindentation

For anisotropic materials, the indentation modulus is a complicated function of the elastic material constants and depends on the indentation direction and the indenter shape. Closed form solutions are only available for the orientation of the major material axis (Delafargue and Ulm, 2004). For the present study a model which gives solutions for all possible indentation directions is required. Such solutions are given by Vlassak et al. (2003) and Swadener and Pharr (2001) which provide general implicit solutions for conical indentation into a anisotropic material. These solutions give the required relation between indentation modulus, elastic constants and indentation direction relative to the principal material directions. In the present paper the model presented by Vlassak et al. (2003) is employed. The same model has already been employed to describe the indentation response of wood cell walls (Jäger et al., 2010) which were considered to be transversely isotropic. For a more detailed description the reader is referred to the original papers mentioned above.

Parameter identification procedure

The identification of elastic material constants of the tested wood cell wall material is based on an error minimization procedure. In this procedure, the experimentally measured indentation moduli M^{exp} are compared to the indentation moduli M^{pred} predicted by the model for orthotropic indentation presented by Vlassak et al. (2003). Since the cell wall material is assumed to be transversely isotropic, the elastic behavior is defined by 5 elastic constants. A parameter study performed by Jäger at al. (2010) showed, that the influence of the Poisson's ratios ν_{tt} and ν_{tl} on the indentation modulus is negligible compared to the influence of the elastic moduli E_l, E_t, and G_{tl}. Hence, only the elastic moduli are identified in the course of the error minimization procedure and both Poisson's ratios are set equal to 0.3. For a more detailed description of the error minimization procedure the reader is referred to the original paper of Jäger et al. (2010).

Results and Discussion

The measured indentation moduli M are displayed together with the corresponding indentation angles in Fig. 3. A clear decrease of M with increasing indentation angles from approx. 19 GPa for 0° to approx. 6 GPa for 90° is evident to a similar extent for both radial as well as tangential cell walls. This confirms the assumption of transversely isotropic material behavior. The absolute values of M agree well with data given in the literature ranging from 8 GPa (spruce compression wood with MFA=50°) to 18,0 GPa (Spruce with MFA=0°) (Gindl et al., 2004). The influence of the angle between indentation direction and microfibril orientation on the indentation modulus is already well known (Gindl et al., 2004; Konnerth et al., 2009; Tze et al., 2007). The novelty of the present data set is that it provides experimental results for indentation angles between 0° and 90° from one latewood zone only, all tested under the same conditions, which minimizes the effect of possible variations from moisture content or chemical composition on M. This unique data set serves as basis for the identification of the elastic material parameters of the wood cell wall material following the procedure outlined in Jäger et al. (2010).

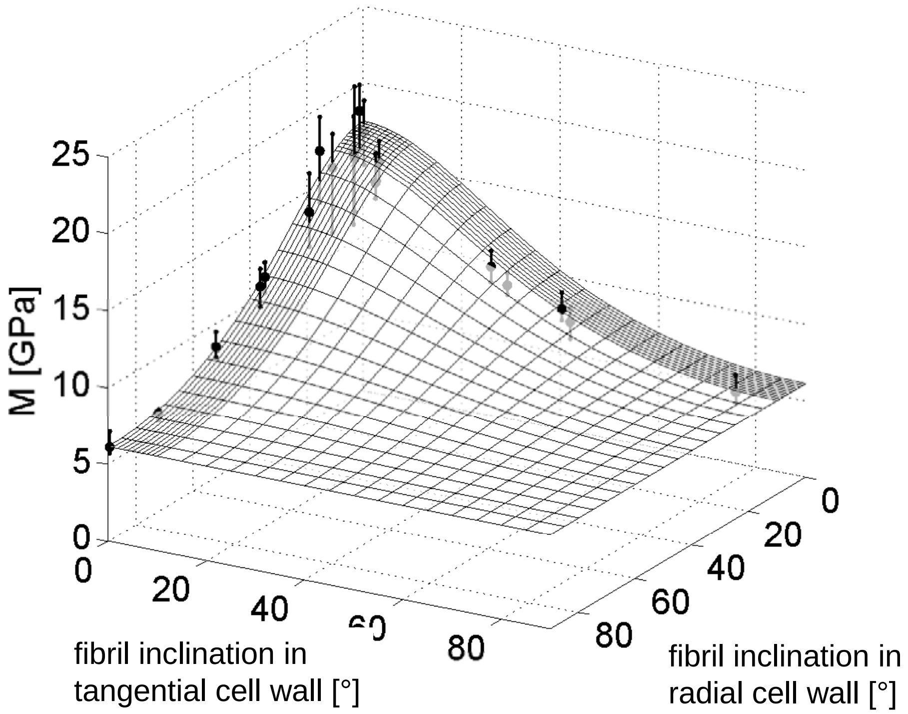

Fig. 3. Influence of the angle between fibril orientation and indentation direction on the indentation modulus. The experimental scatter is indicated by error bars showing minimum and maximum values.

Elastic material constants of wood cell walls

As already mentioned, the cell wall material is assumed to be a transversely isotropic material with five unknown elastic constants. The Poisson's ratios ν_{tt} and ν_{tl} are held constant at a value of 0.3 as parameter studies (Jäger et al., 2011) have shown that their influence on the indentation modulus is negligible. In order to study the influence of the experimental scatter, the parameter identification is performed for the mean values of the experimental data, as well as for the minimum and maximum values. The obtained elastic moduli of the cell wall material and the corresponding error between model response and experimental data are given in Table 1. Together with the two Poisson's ratios ν_{tt} and ν_{tl} all components of the elastic stiffness tensor of the cell wall material are now available. A comparison of the elastic parameters obtained for the different data sets shows that the shear modulus G_{tl} remains approximately constant, whereas the moduli in transverse and longitudinal direction are found to increase from the minimum to the maximum by 28 % and 79 %, respectively.

Fig. 4(a) shows a comparison of experimentally measured indentation moduli and the model response computed for the determined sets of elastic moduli. An excellent agreement is found between model response and experimental data over the whole range of indentation angles. This is also obvious from the correlation plot computed for the mean values and depicted in Fig. 4(b). The good agreement over the whole range of indentation angles confirms again the assumption of transversely isotropic material behavior.

Table 1: Elastic material parameters of cell wall material (E_l, E_t, and G_{tl}) and corresponding error determined via parameter identification for minimal, mean, and maximum values of experimental results.

Exp. data	E_t [Gpa]	E_l [GPa]	G_{tl} [GPa]	Error [%]
minimum	4.29	19.30	4.55	11.20
mean	4.54	26.32	4.84	7.91
maximum	5.50	34.56	4.26	8.41

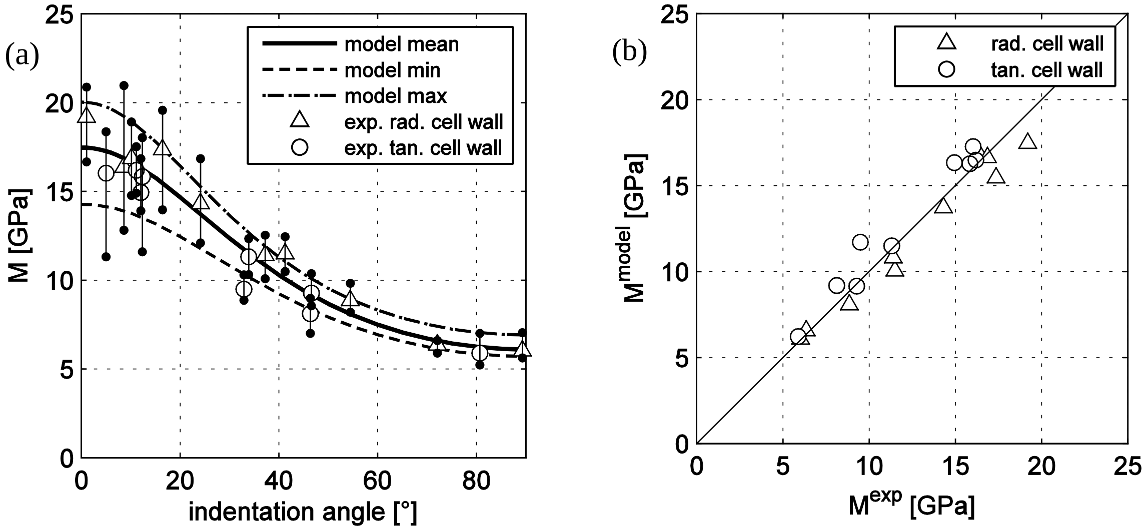

Fig. 4. Comparison of experimental results and model response obtained from parameter identification: (a) variation of indentation modulus M with indentation angle, and (b) comparison of predicted and measured indentation moduli (mean values); the solid line indicates locations of perfect agreement.

Conclusion

For the first time all elastic components of the stiffness tensor of the wood cell wall were successfully determined based on experimental nanoindentation measurements. Varying indentation directions with respect to the orientation of the cellulose microfibrils resulted in a function of the indentation modulus depending on indentation angles over the whole range from 0° to 90°. The experimentally obtained indentation moduli were fitted by a model for orthotropic indentation presented by Vlassak et al. (2003), giving the elastic material constants of the cell wall material. The applied parameter identification procedure (Jäger et al., 2011) gives access to the full stiffness tensor of wood cell wall material. In general, this procedure is applicable for all types of transversely isotropic but also anisotropic materials. The excellent agreement between experimental results and model response supports the assumption that the mechanical behavior of the cell wall material can be approximated by transverse isotropy.

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