

Wood Physics/Mechanical Properties

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Moisture-dependent elastic and plastic properties of pine wood and bamboo cell wall layers measured by nanoindentation

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Abstract: A more systematic, multiscale characterization of the mechanical properties of lignocellulosic materials, such as wood and bamboo, is needed so that manufacturers can better tailor their properties for specific end-uses. Nanoindentation was used to assess the mesoscale elastic modulus (E_S^{NI}) and Meyer's hardness (H) of secondary cell wall layers (SCWL) in Moso bamboo, and S_2 SCWL and compound corner middle lamella (CCML) in loblolly pine. The anisotropic SCWL was tested in both longitudinal and transverse directions. The effects of moisture content (MC) were studied by testing specimens conditioned between 1 % and 92 % relative humidity (RH). Both pine and bamboo SCWL exhibited many of the same features in their MC- and orientation-dependent E_S^{NI} and H results, which suggested that the features were general characteristics of SCWL from different plants. A new transition in plasticity was observed around 2–4 % MC in which the SCWL transverse H had a sudden 20 % decrease. For comparison to literature plasticity S_2 SCWL uniaxial compression flow stress (σ) measurements made using micropillar compression testing, a H -derived uniaxial compression flow stress (σ_H) was calculated

using established methods. The σ_H compared well to the literature σ , which supported that both H and σ can be used to similarly study plastic deformation mechanisms.

Keywords: elastic modulus; hardness; nanoindentation; moisture; cell wall layers

1 Introduction

Sustainable lignocellulosic resources, including wood and bamboo, are poised to play a major role in meeting future needs for materials, chemicals, and fuels (Jakes et al. 2016; Ragauskas et al. 2006). However, lignocellulosic materials have some drawbacks that prevent them from reaching their full potential, including recalcitrance in biorefineries, susceptibility to decay, and moisture-induced dimensional instabilities, that limit their use as building materials in outdoor applications. To accelerate the progress in overcoming these limitations, an improved fundamental understanding of the multi-scale processing-structure-property-performance relationships controlling the mechanical properties is required. For example, bulk wood modifications that impart some dimensional stabilization and resistance to degradation, such as thermal modification (Esteves and Pereira 2008), also tend to decrease wood strength, resulting in wood with limited structural utility. Additionally, mechanical properties are a key factor in determining energy consumption during size reduction processes for overcoming recalcitrance in biorefineries. It has been shown that size reduction energy consumes a major portion, if not all, of the thermal energy in ethanol produced from woody biomass (Zhu and Pan 2010). An improved understanding would help identify ways of tailoring the composition, structure, and processing routes to optimize properties for specific end uses.

Trees and bamboo are different types of plants that produce lignocellulosic resources. Bamboos are fast-growing grasses whose woody tissue consists mostly of vascular bundles, including xylem, phloem, and thick-walled sclerenchyma fibers embedded in parenchyma cells (Grosser and Liese 1971). Trees have shoots that exhibit secondary growth, which

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results in concentric growth rings. The woody tissue structure of trees depends on whether they are softwood or hardwood. Softwood mostly consists of axial tracheids, whereas hardwoods have axial fibers and vessel cells (Wiedenhoef 2013).

Although wood and bamboo have distinct cellular structures (Wang et al. 2014), their mechanical properties are largely derived from similarly structured secondary cell wall layers (SCWL). SCWL consists of highly ordered stiff cellulose fibrils that are helically embedded in a compliant matrix of amorphous cellulose, hemicelluloses, and lignin (Parameswaran and Liese 1976; Salmén and Burgert 2009; Terashima et al. 2009). The angle between cellulose fibrils and the longitudinal axis of the cell is called the microfibril angle (MFA) (Figure 1). In softwood tracheids and hardwood fibers, there are typically three SCWL labeled S_1 , S_2 , and S_3 , as shown in Figure 1a. In contrast, bamboo adds additional cell wall layers as it matures, resulting in a polylamellate structure with varying numbers of alternating thicker and thinner SCWL (Figure 1b) (Parameswaran and Liese 1976). In both bamboo and wood, thicker SCWL have smaller MFA's. Cells in trees and bamboo are held together by a lignin-rich compound middle lamella (CML) cell wall layer. The larger volume of CML at the corner between cells is termed the corner CML (CCML).

Water is another important component of woody lignocellulosic resources such as wood and bamboo. As hygroscopic materials, cell walls readily adsorb and desorb water depending on the conditions of the local environment. Moisture content (MC) is defined as the mass of water in wood divided by the oven-dry mass of wood, and is typically expressed as a percentage (Glass and Zelinka 2010).

As hierarchical materials, the mechanical properties of woody tissues are derived from the properties and organization of their small-scale components (Hofstetter and Gamstedt 2009; Salmén and Burgert 2009; Tan et al. 2011). With regard to the mechanical properties, water is generally observed to have a plasticizing effect, in which the properties decrease with increasing MC (Tiemann 1906). Because thicker SCWL typically control the bulk mechanical properties, understanding their properties is an important step towards developing processing steps to optimize mechanical properties for specific end-uses. Some aspects have long been understood, such as the relationship between longitudinal elastic moduli and cellulose MFA (Cowdrey and Preston 1966). However, many other aspects remain to be elucidated, including plasticity, fracture, and the role of matrix polymers in the mechanical properties.

A major hindrance to progress is the lack of systematic SCWL mechanical property measurements over a wide range of environmental conditions and orientations (Salmén 2018). Past experiments to directly measure SCWL mechanical properties include bending of cantilevers ion milled from SCWL (Orso et al. 2006), compression of micropillar compression ion milled in an SCWL (Adusumalli et al. 2010), atomic force microscopy imaging methods (Li and Kasal 2025), and nanoindentation (Wimmer et al. 1997). Tensile experiments on individual fibers have also been conducted to estimate the mechanical properties of SCWL (Eder et al. 2013). In a nanoindentation experiment, a carefully shaped probe is pressed into a material following a prescribed load function. The applied load and indentation depth are

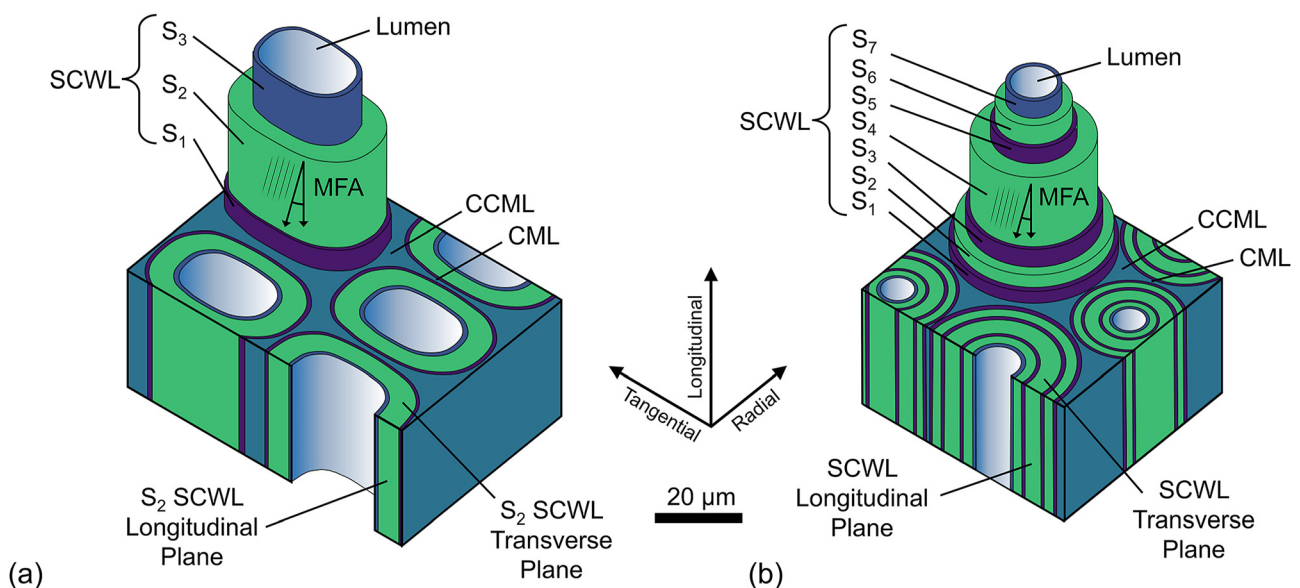


Figure 1: Schematics of the three-dimensional microstructure in (a) wood and (b) bamboo illustrating lumina, secondary cell wall layers (SCWL), compound middle lamella (CML), corner CML (CCML), and cellulose microfibril angles (MFA).

continuously measured during the experiment. Mechanical properties, such as elastic modulus and hardness can then be calculated from resulting load-depth trace. In principle, tensile, bending, and compression experiments are easier to interpret than nanoindentation because these uniaxial experiments have a much simpler stress state than the complex stress state beneath a nanoindentation probe. However, nanoindentation is the only experimental technique capable of simultaneously measuring elastic and plastic properties over a wide range of relative humidity (RH) (Yu et al. 2011), temperature (Konnerth and Gindl 2008), and over the full range of orientation in the SCWL (Jäger et al. 2011b). Nanoindentation can also be readily used to assess the time-dependent viscoelastic (Zhang et al. 2012) and viscoplastic (Jakes et al. 2008b) properties of wood cell walls.

Mechanical property measurements over wide ranges of time, temperature, or moisture content have the potential to not only provide useful information about how properties change under different conditions, but also provide insights into the microphysical processes causally linked to the mechanical properties (Lakes 2009). Insights about the deformation mechanisms are often gained from distinct “signatures” or features observed in the plots of the condition-dependent mechanical properties. For example, a sudden drop in the temperature-dependent elastic modulus plot of an amorphous polymer likely indicates the material passed through its glass transition (Lakes 2009). Hardness strain rate sensitivity in nanoindentation creep experiments can also be related to physical characteristics of plastic deformation mechanisms (Puthoff et al. 2009).

The aim of this study was to address the need to better understand the moisture-dependent mechanical properties in lignocellulosic cell walls by using Berkovich quasistatic nanoindentation to assess the moisture-dependent elastic modulus and hardness in pine and bamboo cell wall layers. Experiments were performed at closely spaced RH intervals, especially at lower RH, to produce detailed moisture-dependent plots of hardness and elastic modulus to help identify mechanical signatures in the data. Thick SCWL in bamboo and pine were tested in both the transverse and longitudinal planes. CCML in pine was also tested. The resulting data were analyzed both to examine how elastic and plastic properties change with moisture and to gain new insights into the microphysical processes controlling the properties. Results were compared to nanoindentation results in the literature. Nanoindentation hardness was also compared to literature plasticity measurements in wood S₂ SCWL that have been made using micropillar compression experiments.

2 Materials and methods

The lignocellulosic materials tested were latewood loblolly pine (*Pinus taeda*) and vascular fiber bundles from the dense outer region of Moso bamboo (*Phyllostachys edulis*). Two 5-mm cubes nominally oriented along the three primary material axes were prepared from each material. For each material, a transverse face and a tangential-longitudinal face were bonded to the end of stainless-steel cylinders (8 mm in diameter, 9.5 mm long) using a thin layer of 5-min epoxy. Nanoindentation surfaces were prepared without epoxy embedment following established protocols (Jakes and Stone 2021). First, a pyramid with an apex in the region of interest was carefully shaped in each specimen using a hand razor. The specimens were then fitted using a Leica EM UC7 ultramicrotome (Wetzlar, Germany) equipped with a diamond knife. Surfaces for nanoindentation were prepared by removing 200-nm-thick sections from the apex until a surface of 100 μm to 300 μm on a side was prepared.

A Bruker-Hysitron (Minneapolis, Minnesota, USA) TriboIndenter[®] equipped with a Berkovich probe was used. The machine compliance, probe area function, and tip roundness effects were determined from a series of 80 nanoindentations in a fused silica standard using the load function and procedures described in (Jakes 2018; Stone et al. 1991). Following the calibration reporting procedure prescribed in (Jakes 2018): Values for the square root of the Joslin-Oliver parameter of $1.217 \pm 0.002 \mu\text{m}/\text{N}^{1/2}$, elastic modulus of $72.0 \pm 0.2 \text{ GPa}$, and Meyer’s hardness of $9.13 \pm 0.03 \text{ GPa}$ (uncertainties are standard errors) were assessed for fused silica calibration nanoindentations with contact depths between 35 and 211 nm; no systematic variations of machine compliance or Joslin-Oliver parameter were observed in the systematic SYS plot analysis over this range of contact depths.

The relative humidity (RH) inside the nanoindentation enclosure was varied from 1 % and 92 % using an Instru-Quest (Coconut Creek, Florida, USA) HumiSys[™] HF RH generator. The specimens were conditioned inside the nanoindenter enclosure at each RH for at least 36 h before the experiments and the RH was maintained during the experiments. The temperature inside the enclosure varied with the laboratory temperature and was measured between 24 °C and 26 °C during the experiments. The calibration of the temperature and RH sensor inside the nanoindentation enclosure was verified using a Control Company (Webster, TX, USA) 4085 Traceable[®] Hygrometer Thermometer Dew-Point Meter.

Nanoindentation experiments were designed to minimize the effects of the natural variability inherent in

lignocellulosic materials by testing the same cell wall or daughter tracheids at different RH levels whenever possible. On the pine transverse surface, a single row of daughter tracheids was chosen, and at each RH, eight nanoindentations were placed on the tangential sides of the S_2 SCWL within a double cell wall. At each RH, six to eight nanoindentations were also placed in the CCML of the pine transverse plane. On the pine tangential-longitudinal surface, 4–5 nanoindentations were placed on the longitudinal plane of an S_2 SCWL at each RH. On the bamboo transverse surface, all transverse plane nanoindentations were placed on fibers from a single bundle of vascular fibers and 11–18 nanoindentations were placed in thick SCWL across 5–8 fibers at each RH. On the bamboo longitudinal surface, 6–7 nanoindentations were performed at each RH along the thick SCWL. On the longitudinal surfaces in both pine and bamboo, cells cut approximately in half along the longitudinal direction with exposed lumina on the surface were chosen. This ensured that the tested SCWL did not have other cell wall layers immediately below the tested surface. The specimens were first conditioned at 1 % RH and experiments were performed in absorption. All pine experiments were performed at 1 %, 3 %, 5 %, 8 %, 12 %, 17 %, 23 %, 30 %, 39 %, 56 %, 77 %, and 88 % RH. Bamboo SCWL experiments on the transverse plane were performed at 1 %, 4 %, 7 %, 10 %, 14 %, 18 %, 40 %, 67 %, and 78 % RH, and on the longitudinal plane at 1 %, 3 %, 5 %, 8 %, 11 %, 12 %, 17 %, 23 %, 30 %, 39 %, 56 %, 77 %, and 92 % RH. The moisture content (MC) of the different cell wall layers was assumed to be that of the bulk material. The MC of pine S_2 SCWL and CCML were both estimated using the absorption isotherm of loblolly pine (Zelinka and Glass 2010). The MC of bamboo SCWL was estimated using the absorption isotherm of Moso bamboo taken from the dense outer region of a culm (Wei et al. 2021).

The nanoindentation protocols and analyses followed the guidance provided by Jakes and Stone (2021) to minimize the effects of surface detection errors, structural compliances arising from nearby free edges and cellular flexing, dirty probes, displacement drift, nanoindenter calibration issues, and nanoindenter performance issues. Briefly, a Berkovich probe was used to obtain z-height scanning probe microscopy (SPM) images to help position the nanoindentations in the cell wall layer of interest. SPM images were obtained using a 1 Hz scanning rate and a 1–2 μ N load setpoint. The multi-load function described in (Jakes et al. 2015) was used in this study. The maximum load was varied between 0.22 mN and 0.45 mN with decreasing loads at higher RH to maintain similarly sized nanoindentations across all RH conditions. The resulting experiments had total depths that varied from 170 nm to 220 nm. From the SPM images of residual nanoindentations, any nanoindentation

that was not completely contained within the SCWL or CCML was excluded from the analysis. For nanoindentation analysis, the structural compliance method (Jakes et al. 2008a) was employed to remove potential artifacts caused by edge effects and specimen-scale flexing at each nanoindentation location. Unloading segments with contact depths less than 35 nm, which were found to be affected by the tip roundness effects in the fused silica calibrations, were excluded from the structural compliance analysis. After correcting the data for structural compliance, the Meyer hardness (H) was calculated using

$$H = \frac{P_0}{A_0} \quad (1)$$

where P_0 and A_0 are the load and contact area, respectively, immediately prior to each unloading segment. A_0 was calculated using the probe area function and the Oliver–Pharr contact depth (Oliver and Pharr 1992). The effective modulus (E_{eff}) of contact was calculated using

$$E_{\text{eff}} = \frac{S}{A_0^{1/2}} \quad (2)$$

where S is the contact stiffness calculated by fitting the Oliver–Pharr power law function (Oliver and Pharr 1992) from to 40–95 % of the maximum load of each unloading segment. The diamond probe contributions to E_{eff} are accounted for in the calculation of the nanoindentation elastic modulus (E_s^{NI}) using

$$\frac{1}{E_{\text{eff}}} = \frac{\pi^2}{2\beta} \left(\frac{1 - \nu_s^2}{E_s^{NI}} + \frac{1 - \nu_d^2}{E_d} \right) \quad (3)$$

where E_d is the Young's modulus of diamond (1,137 GPa), ν_d is the Poisson's ratio of diamond (0.07), and ν_s is the Poisson's ratio assumed for the tested SCWL or CCML (0.45) (Wimmer et al. 1997). The numerical factor β was assumed to be 1. The material isotropy assumption implicit in Equation (3) is violated in SCWL nanoindentations because cellulose microfibrils cause orientation effects (Gindl and Schoberl 2004; Jäger et al. 2011a). Protocols have been developed to use Berkovich nanoindentation experiments to calculate transverse isotropic elastic constants of SCWL in which the plane of isotropy is perpendicular to the longitudinal axes of the cellulose fibrils (Jäger et al. 2011a,b). Arzola-Villegas et al. (2025) recently used these protocols to calculate the longitudinal elastic modulus and transverse elastic modulus in the S_2 SCWL of loblolly pine conditioned at 0 %, 33 %, 75 %, and 94 % RH. A comparison was made between nanoindentation elastic moduli calculated with the isotropic material assumption like E_s^{NI} in Equation (3) to the longitudinal and transverse elastic moduli calculated using the transverse isotropic material

assumption. Across all moisture levels tested, for experiments in which the indentation direction was parallel to the longitudinal axes of the cellulose fibrils the longitudinal elastic moduli were 6–8 GPa higher than E_S^{NI} . The transverse elastic moduli were about 1 GPa lower than E_S^{NI} in experiments with the indentation direction perpendicular to the longitudinal axes of the cellulose fibrils. The protocols to calculate transverse and longitudinal moduli of SCWL include performing nanoindentation experiments over the full range of possible angles between the indentation direction and cellulose fibril orientation. Unfortunately, it was not practical to perform such experiments at all the closely spaced moisture levels tested in this study. Therefore, only measurements in the transverse and longitudinal planes of the SCWL were made. The large number of moisture levels tested were prioritized to help identify mechanical signatures in the MC-dependent data that could provide insights into deformation mechanisms. Therefore, the “NI” superscript was included to indicate that the assessed elastic modulus is not the Young’s modulus typically calculated using Equation (3). H and E_S^{NI} were calculated for each unloading segment in the multi-load nanoindentation. Results from unloading segments affected by tip roundness effects (contact depths less than 35 nm) were excluded. After excluding data affected by tip roundness, no data exhibited any systematic size dependence. Therefore, for each specimen and RH level, all the results from the remaining unloading slopes were averaged and used to calculate the standard deviations and standard errors.

Micropillar compression experiments are another type of experiment used to measure the plastic properties in wood S_2 SCWL (Adusumalli et al. 2010). In micropillar compression experiments, micropillars of S_2 SCWL are machined using a focused ion beam. The micropillars are then compressed using a flat punch probe in a nanoindenter. Mechanical properties, such as uniaxial compression flow stress (σ), can be calculated from the resulting stress-strain data. Although H and σ are both quantitative measurements of plasticity, they are assessed under different stress states. Therefore, an empirical conversion calculation was used to compare literature micropillar σ results to nanoindentation H results obtained in this study. The H -derived uniaxial flow stress (σ_H) can be calculated from H using the Tabor-Marsh-Johnson correlation (Johnson 1970; Marsh 1964; Tabor 1948, 1970)

$$\sigma_H = \frac{H}{k} \quad (4)$$

where k is the constraint factor that depends on the probe geometry and E^*/σ . E^* is the Hertzian contact modulus defined as

$$E^* = \frac{\pi^{1/2}}{2\beta} E_{\text{eff}} \quad (5)$$

where the numerical factor β was again assumed to be 1.

3 Results

SPM images of the triangular-shaped residual Berkovich nanoindentation impressions in different cell wall layers and orientations are shown in Figure 2. The cell wall layers tested in these unembedded specimens had smooth surfaces that were suitable for nanoindentation. Free edges at the lumen surfaces and interfaces between different cell wall layers were also visualized and used to ensure that the nanoindentations were completely contained within the cell wall layer of interest. These free edges and heterophase interfaces violate the typical assumption in nanoindentation that the tested material is a homogeneous half-space. The open cellular structure in unembedded wood and bamboo also violates the rigid support assumption because the specimen may flex under loading. Therefore, multiloading nanoindentations and the structural compliance method were utilized to correct the nanoindentation results from potential artifacts arising from specimen-scale flexing or edge effects (Jakes and Stone 2021; Jakes et al. 2008a).

The MC-dependent bamboo and pine E_S^{NI} are plotted in Figure 3. In SCWL, the orientation of highly ordered stiff cellulose fibrils is the dominant factor that causes

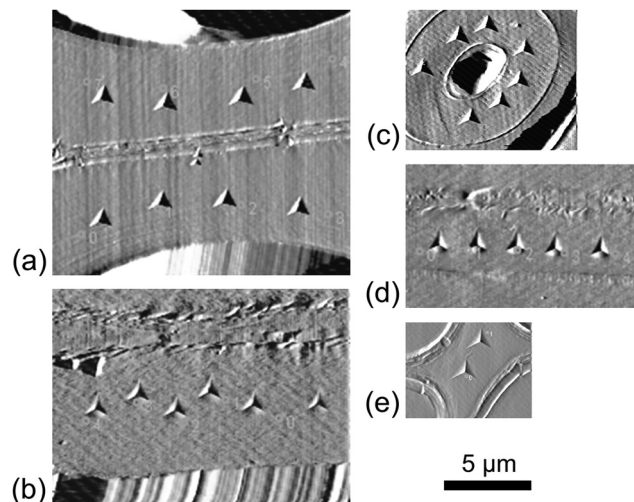


Figure 2: Scanning probe microscopy (SPM) images of nanoindentations in (a) transverse plane S_2 secondary cell wall layer (SCWL) of pine, (b) longitudinal plane S_2 SCWL of pine, (c) transverse plane SCWL of bamboo, (d) longitudinal plane SCWL of bamboo, and (e) transverse plane compound corner middle lamellae (CCML) of pine. SPM images were slope-shaded to better visualize the surface features.

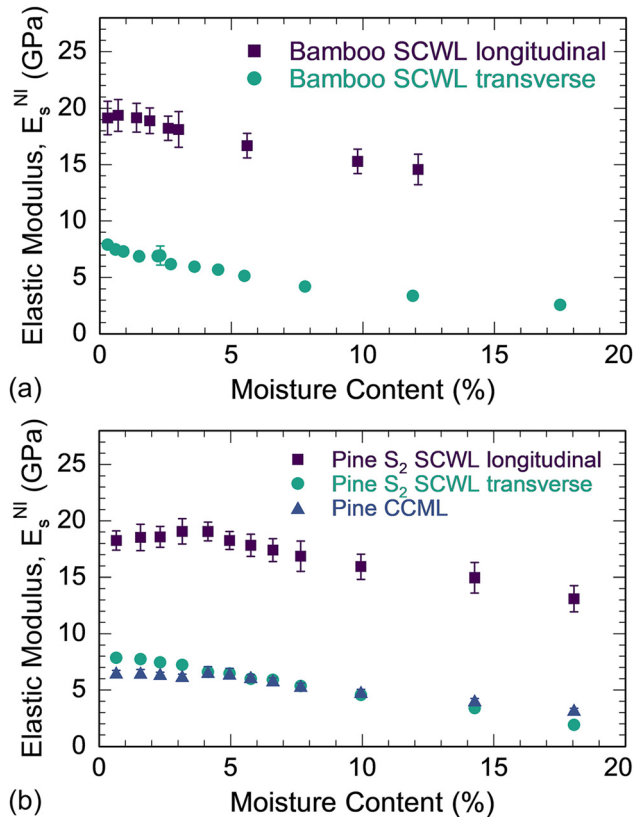


Figure 3: Nanoindentation elastic modulus (E_s^{NI}) as a function of calculated moisture content for (a) bamboo secondary cell wall layers (SCWL) and (b) pine S₂ SCWL and corner compound middle lamella (CCML). The longitudinal and transverse directions indicate the wood anatomical direction parallel to the direction of the applied force during nanoindentation. The bamboo E_s^{NI} results were previously reported in (Youssefian et al. 2017). Plotted error bars represent standard deviations. The error bars for the standard errors are smaller than the symbols.

anisotropic elastic properties (Jäger et al. 2011a; Salmén and Burgert 2009). As expected, the longitudinal E_s^{NI} , which was assessed from nanoindentations placed on the transverse plane and measured the elastic moduli in the direction close to parallel to the longitudinal axes of the cellulose fibrils, was much higher than the transverse E_s^{NI} , which measured the elastic modulus in the direction nearly perpendicular to the longitudinal axes of the fibrils. The SCWL transverse E_s^{NI} values from both materials were similar to those of the pine CCML E_s^{NI} . CCML lacks cellulose fibrils and is comprised of approximately 80 % lignin and 20 % hemicelluloses (Rowell et al. 2013). The similarity between the SCWL transverse E_s^{NI} and CCML E_s^{NI} supports the idea that the transverse elastic modulus is dominated by the properties of the more compliant matrix polymers, which include lignin and hemicelluloses (Salmén 2004).

The E_s^{NI} plotted in Figure 3 was calculated using Equation (3), which assumes that the material tested is elastically isotropic. This isotropy assumption is violated for SCWL (Gindl and Schoberl 2004; Jäger et al. 2011a). Based on previous experiments to determine anisotropic longitudinal elastic moduli and transverse elastic moduli in latewood loblolly pine tested from 0 % to 94 % RH (Arzola-Villegas et al. 2025), the SCWL longitudinal E_s^{NI} were likely underestimated by about 6–8 GPa compared to anisotropic longitudinal elastic moduli, and the SCWL transverse E_s^{NI} were likely overestimated by about 1 GPa compared to anisotropic transverse elastic moduli. Fortunately, the magnitudes of the offsets between E_s^{NI} and the anisotropic elastic moduli in the previous experiments did not depend on moisture. Therefore, the trends in the MC-dependent E_s^{NI} in Figure 3 are expected to be similar to those that would have been observed if the anisotropic elastic moduli were measured at the same moisture levels.

Further insights into the contributions of lignin and hemicelluloses to the cell wall elastic properties were gained by analyzing the MC dependence in Figure 3 for unique mechanical signatures. The pine CCML had a relatively constant E_s^{NI} of up to approximately 5 % MC, after which the E_s^{NI} decreased with increasing MC. In SCWL longitudinal E_s^{NI} , E_s^{NI} increased or remained relatively constant below 5 % MC for pine and below 2 % MC for bamboo, and then decreased with increasing MC. A similar mechanical signature with an initial increase at low moisture levels was observed in both bulk wood longitudinal elastic moduli (Gerhards 1982) and in elastic modulus measurements of lignin (Cousins 1976). In a previous study using atomistic simulations to interpret these bamboo E_s^{NI} results, the increase in longitudinal E_s^{NI} at low MC was attributed to the increases in stiffness of the lignin or lignin carbohydrate complexes (LCC) caused by the first water molecules entering molecular-scale ‘holes’ in their structures and creating new hydrogen bonds that enhance their overall stiffness (Youssefian et al. 2017). The same mechanism is likely responsible for the increases at low MC in pine longitudinal E_s^{NI} and lignin-rich CCML E_s^{NI} , which indicates that the behavior is a general characteristic of SCWL from different plants. In contrast to the longitudinal E_s^{NI} , both the pine and bamboo transverse E_s^{NI} decreased over the entire range of MC tested. This behavior is consistent with the plasticizing effect of water over the entire range of tested moisture conditions, as observed in the previous atomistic simulations (Youssefian et al. 2017). The transverse E_s^{NI} trends are also consistent with experimental measurements of bulk wood transverse elastic moduli (Gerhards 1982) and elastic moduli measurements of hemicelluloses (Cousins 1978).

Although it is well known that the orientation of stiff cellulose fibrils dominates the orientation-dependent elastic properties of SCWL (Jäger et al. 2011a; Salmén and Burgert 2009), there is increasing evidence that different matrix polymers have different effects depending on the orientation. These E_S^{NI} results are consistent with previous experiments and literature reviews of wood mechanical properties across multiple length scales, indicating that lignin has a larger effect than hemicelluloses when the elastic properties are measured in the longitudinal direction, whereas hemicelluloses have a larger effect than lignin in the transverse direction (Jakes et al. 2019; Youssefian et al. 2017). Therefore, when processing is expected to modify the molecular structure of matrix polymers, both longitudinal and transverse mechanical properties need to be assessed to fully study the potential effects of processing on SCWL elastic properties.

The dependence of H on MC in pine and bamboo is plotted in Figure 4. Compared to the E_S^{NI} , there was much less anisotropy in SCWL H . Previous studies have also observed that H is less dependent on MFA than E_S^{NI} (Arzola-Villegas et al. 2025; Eder et al. 2013; Gindl et al. 2004; Wanju et al. 2014). H did not increase at low MC like the E_S^{NI} . H decreased with increasing MC across the entire range of the tested MC. However, there was a transition in the behavior at approximately 2 % MC for bamboo and 4 % MC for pine. Below these values of MC, the SCWL H values were nearly identical in the longitudinal and transverse directions in the bamboo and pine. However, above these values of MC, SCWL transverse H in both bamboo and pine exhibited a sudden drop of approximately 20 %, or 100 MPa. The pine longitudinal SCWL and CCML exhibited a noticeable inflection in H at approximately 4 % MC, with MC having a much larger softening effect at MC values above 4 %. The transitions in MC-dependent H at approximately 2–4 % MC, especially the drastic drop in the SCWL transverse H , likely indicated a change in the deformation mechanism. The similar behavior of pine and bamboo suggests that this transition at 2–4 % MC is a common characteristic of SCWL from different plants. Although the increase in the elastic modulus occurred at a similar MC (Figure 3), it remains uncertain whether these transitions in H were also related to the same molecular-scale mechanism of lignin being stiffened by water-filled molecular-scale holes in the lignin molecular structure. The deformation mechanisms causing this transition are currently unknown. However, a discussion of SCWL plastic deformation mechanisms and future nanoindentation experiments to further study this transition in plastic properties will be included in the discussion section.

The MC-dependent E_S^{NI} and H values generally agree with the values reported for Berkovich nanoindentations in

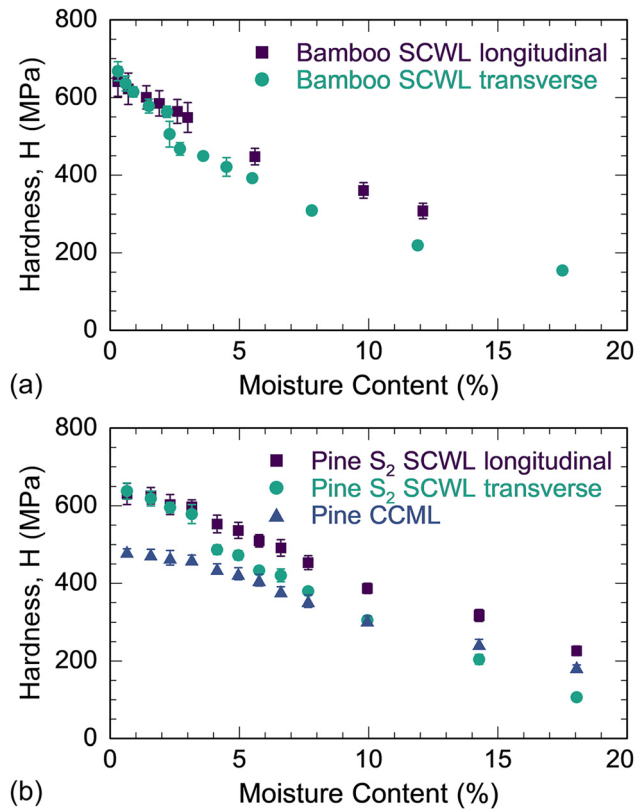


Figure 4: Nanoindentation Meyer's hardness (H) as a function of calculated moisture content for (a) bamboo secondary cell wall layers (SCWL) and (b) pine S₂ SCWL and corner compound middle lamella (CCML). Longitudinal and transverse indicate the wood anatomical direction parallel to the direction of applied force during nanoindentation. Plotted error bars are standard deviations. Error bars for standard errors are smaller than the symbols.

the literature. The values in Table 1 are for nanoindentation in the SCWL transverse planes, which assesses longitudinal properties. The studies tested only three to six levels of MC within the hygroscopic range. None of the studies in Table 1 reported a constant or increasing elastic modulus at low MC. However, for studies with at least two moisture levels at or below 6 % MC, the elastic modulus either increased or remained constant before decreasing at higher levels of moisture, which is consistent with the results for pine and bamboo in Figure 3. It is only with the closely spaced data in this study that low-MC trends become obvious. Similar to the current H results in Figure 4, the literature hardness decreased with MC over all ranges, except for the 27° MFA data of Masson pine that had anomalous behavior with its 11 % MC hardness slightly higher than its 8 % MC hardness (Wanju et al. 2014). Among the different studies, there was more scatter in the elastic modulus magnitudes than in the hardness, but this was likely caused by differences in MFA and how the elastic moduli were determined. Differences in

MFA have a much larger effect on elastic modulus than on hardness (Arzola-Villegas et al. 2025; Eder et al. 2013; Gindl et al. 2004; Wanju et al. 2014). Some studies also assumed different variable values, such as for SCWL ν_s and β , when using Equation (3) to calculate E_s^{NI} . Other studies reported a different type of indentation modulus, such as the Hertzian contact modulus defined in Equation (5), instead of E_s^{NI} . The effects of assuming different variable values or using different types of indentation moduli is primarily to change the magnitudes of the elastic moduli, but not their trends with moisture. Therefore, the elastic modulus trends with moisture are more easily comparable than their magnitudes. Rindler et al. (2019) also tested Norway spruce SCWL in the transverse plane at 6 %, 9 %, and 12 % MC. Their data was not included in Table 1 for comparison because average values were not reported. However, they did report that there was no statistical difference in elastic modulus nor hardness caused by changes in MC in their study.

Wagner et al. (2015) reported Berkovich nanoindentation CCML elastic modulus and hardness for Scots pine, Norway spruce, Common yew, European oak, and European beech. CCML properties were measured at 10 %, 40 %, 60 %, and 80 % RH, which correspond to 4 %, 8 %, 11 %, and 15 % MC using the loblolly pine isotherm used in this study (Zelinka and Glass 2010). Both elastic modulus and hardness consistently decreased with increasing MC for all five wood species, which is consistent with CCML E_s^{NI} in Figure 3b and H in Figure 4b over the same MC range. At 4 % MC, the CCML elastic moduli ranged from 7.8 to 9.7 GPa

for the five wood species and decreased to the range of 4.5–5.5 GPa at 15 % MC. Similarly, the CCML hardness decreased from 400 to 470 MPa at 4 % MC to 150–300 MPa at 15 % MC. While the loblolly pine CCML H in Figure 4b falls within the ranges reported for the five wood species, the E_s^{NI} in Figure 3b were lower at about 6.5 GPa for 4 % MC and 3.5 GPa for 15 % MC. The difference in elastic moduli values was largely because of how the elastic moduli were calculated. Wagner et al. (2015) reported reduced moduli, which is equivalent to the Hertzian contact modulus defined in Equation (5). A reduced modulus would be about 25 % higher than the E_s^{NI} calculated using Equation (3) with the assumed CCML Poisson's ratio of 0.45. This 25 % difference accounted for almost all the difference in magnitude between the pine CCML E_s^{NI} in Figure 3b and the range of elastic moduli reported for the five wood species. Additionally, the effect of moisture softening from 4 % MC to 15 % MC was similar at about 3 GPa for both the pine E_s^{NI} in Figure 3b and the five wood species reported by Wagner et al. (2015).

4 Discussion

4.1 Comparison of SCWL H to micropillar compression experiments

Micropillar compression experiments have also been used to study plasticity in wood S_2 SCWL. Uniaxial experiments are

Table 1: Literature values for the moisture-dependent secondary cell wall layer elastic modulus and hardness measured by Berkovich nanoindentation.

Species	MFA ^a	MC ^b range tested (%)	Number of MC levels tested	Elastic modulus over MC range (GPa)	Hardness over MC range (MPa)	Reference
Masson pine	N/A	5 to 13	6	21 to 17	650 to 450	Yu et al. (2011)
Moso bamboo	N/A	4 to 11 ^c	5	23 to 20	600 to 470	Wang et al. (2013)
Masson pine	16°	5 to 11	3	18 to 16	420 to 350	Wanju et al. (2014)
Masson pine	27°	5 to 11	3	15 to 13	420 to 370	Wanju et al. (2014)
Masson pine	38°	5 to 11	3	14 to 13	450 to 340	Wanju et al. (2014)
Spruce wood	10°	3 to 14 ^c	4	21 to 13	600 to 200	Bertineti et al. (2015)
Loblolly pine	31°	0 to 18	3	14 to 10	575 to 300	Meng et al. (2015)
Scots pine	13°	4 to 15 ^c	4	20 to 15	490 to 255	Wagner et al. (2015)
Norway spruce	13°	4 to 15 ^c	4	22 to 15	510 to 240	Wagner et al. (2015)
Common yew	27°	4 to 15 ^c	4	15 to 12	475 to 320	Wagner et al. (2015)
European oak	3°	4 to 15 ^c	4	20 to 16	495 to 240	Wagner et al. (2015)
European beech	7°	4 to 15 ^c	4	19 to 15	495 to 270	Wagner et al. (2015)

^aMicrofibril angle (MFA). ^bMoisture content (MC). ^cOnly the RH was reported in these studies. The MC values were calculated using the same loblolly pine isotherm used in this study (Zelinka and Glass 2010).

conventionally used to study plasticity mechanisms in materials, such as to calculate their activation energies and volumes (Swallowe and Lee 2006), in part because their stress states are simpler compared to hardness tests. However, the small scale micropillar compression experiments in S_2 SCWL are very challenging experimentally. It would not be practical to use them to study plasticity over wide ranges of strain rates, temperature, moisture, and orientation, which would be needed to study plasticity mechanisms in wood cell walls. Nanoindentation hardness measurements over these conditions will be needed. It is therefore informative to compare uniaxial compression flow stress (σ) measurements from micropillar compression experiments to nanoindentation H because although σ are typically used in conventional analyses, future studies in wood cell walls will likely rely on H . For the comparison, Equation (4) was used to calculate the H -derived uniaxial flow stress (σ_H). The application of this procedure to calculate σ_H from Berkovich nanoindentation H in wood cell wall layers was motivated by previous experiments in synthetic amorphous and semicrystalline polymers (Jakes et al. 2012, 2024). In these experiments, σ_H calculated from Berkovich nanoindentation H agreed closely with literature uniaxial compression σ results for all five synthetic polymers tested. However, the application of Equation (4) for experiments in wood cell wall layers may be more qualitative currently because the effects of composite structures and anisotropy on the calculation have not been thoroughly studied. Nevertheless, it is important to attempt correlations between H and σ to compare both types of plasticity measurements and so that eventually they can be better understood.

Following Puthoff et al. (2009), it is convenient to define k for a Berkovich probe as a function of H/E^* , as shown in Figure 5. For each unloading segment analyzed, H/E^* was calculated and used with Figure 5 to calculate the k used in Equation (4) to determine σ_H from H . The experimental H/E^* ratio varied depending on the type of cell wall tested, orientation, and MC. The ratios are plotted in Figure 6, and generally decreased with increasing MC. The ratio was lower for the SCWL longitudinal experiments because SCWL E^* was at least a factor of two higher than the transverse E^* , whereas H did not vary much with orientation. The range of H/E^* values for experiments in bamboo and pine are also indicated in Figure 5 and show that the values of k needed for the conversions ranged from 2.6 to 3.

The calculated σ_H is plotted in Figure 7. Some features in the MC-dependent σ_H are similar to those in H in Figure 4, such as the sudden drop in transverse σ_H at 2 % MC for bamboo and 4 % MC for pine. The σ_H in Figure 7 can be compared to literature wood S_2 SCWL plastic properties assessed using

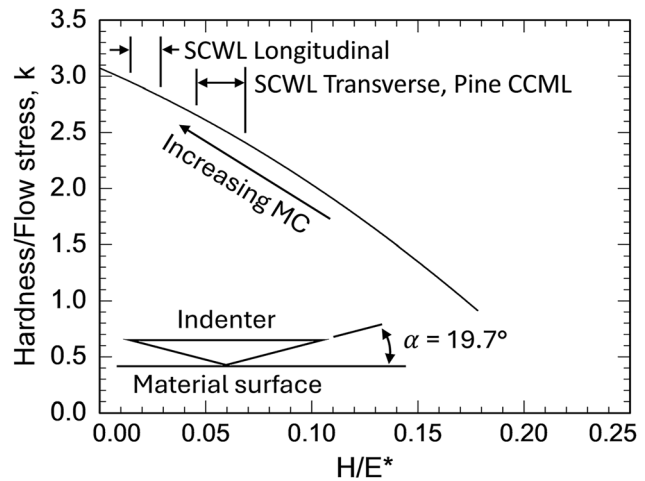


Figure 5: The constraint factor k (Equation (4)) as a function of the ratio of Meyer's hardness to the Hertzian contact modulus (H/E^*) for a Berkovich probe (cone equivalent $\alpha = 19.7^\circ$) based on the analysis of the Tabor-Marsh-Johnson correlation (Puthoff et al. 2009). The ranges for experiments in the secondary cell wall layers (SCWL) and compound corner middle lamella (CCML) are indicated. Longitudinal and transverse indicate the wood anatomical direction parallel to the direction of applied force during nanoindentation. The value of k generally increases with increasing moisture content (MC).

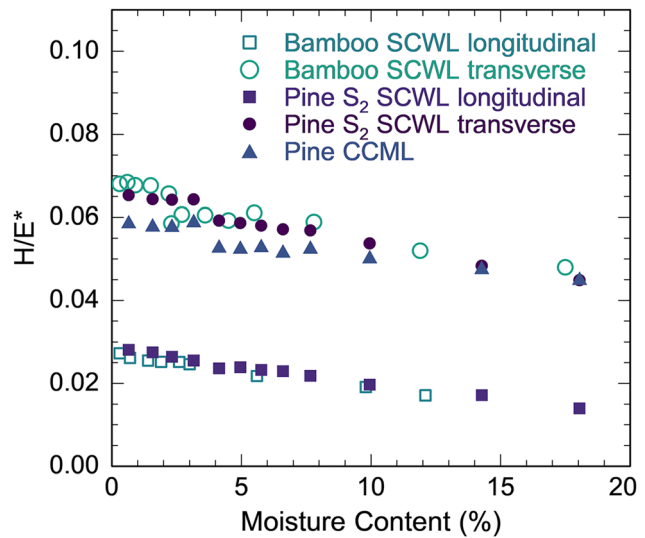


Figure 6: Ratio of Meyer's hardness to Hertzian contact modulus (H/E^*) as a function of calculated moisture content for bamboo secondary cell wall layers (SCWL) and pine S_2 SCWL and corner compound middle lamella (CCML). Longitudinal and transverse indicate the wood anatomical direction parallel to the direction of applied force during nanoindentation.

micropillar compression experiments (Table 2). Micropillar plastic properties are often reported as the yield stress, which

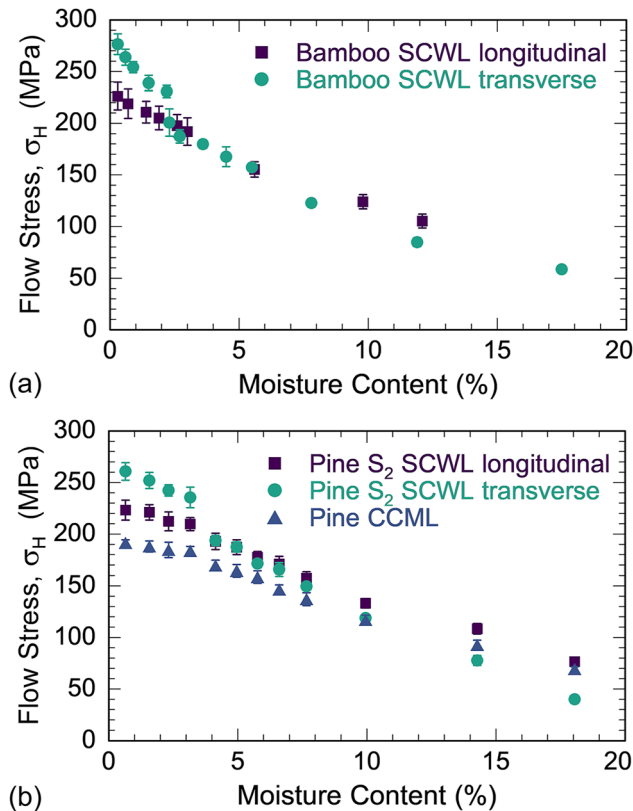


Figure 7: Nanoindentation Meyer's hardness-derived flow stress (σ_H) as a function of calculated moisture content for (a) bamboo secondary cell wall layers (SCWL) and (b) pine S₂ SCWL and corner compound middle lamella (CCML). Longitudinal and transverse indicate the wood anatomical direction parallel to the direction of applied force during nanoindentation. Plotted error bars are standard deviations. Error bars for standard errors are smaller than the symbols.

is the σ on a stress-strain curve at which plastic yielding is first observed. However, an obvious yielding point was not observed in the S₂ SCWL micropillar experiments, and different studies have used different criteria to define the yield stress, as indicated in Table 2. For normal wood, the reported yield stresses ranged from 147 to 183 MPa at 0 % MC, and from 111 to 137 MPa at 8 % MC. All the micropillar compression experiments were performed on the S₂ SCWL in the longitudinal direction. Compared to the SCWL longitudinal σ_H , these values are about 20–35 % lower at 0 % MC and about 10–30 % lower at 8 % MC. However, the σ_H was likely different because it is a measurement at a different representative strain. For the micropillar studies in which stress-strain curves were reported, the strains corresponding to the yielding stress were estimated to range from 3 % to 6 % (Table 2), which is lower than the estimated 7–10 % representative strain beneath a Berkovich probe (Johnson 1985; Kermouche et al. 2008). Therefore, for the studies with reported stress-strain curves a uniaxial σ at 8 % strain was estimated (Table 2). The uniaxial σ at 8 % strain ranges from 175 to 270 MPa for normal wood at 0 % MC, which includes the σ_H of 225 MPa for both pine and bamboo at 0 % MC in the longitudinal direction. Despite the uncertainties in the micropillar stress and strain measurements and the applicability of using Equation (4) to calculate σ_H for complex materials like SCWL, this agreement supports that nanoindentation and micropillar compression experiments are similarly assessing plasticity. Both types of measurements will likely play complementary roles in future studies of plasticity in wood cell wall layers.

Table 2: Summary of literature values for plasticity measured by micropillar compression testing of S₂ secondary wood cell wall layers in wood.

Wood species	MFA ^a	Moisture condition at testing ^b	Yield stress (MPa)	Strain at yield stress ^c	Flow stress at 8 % strain ^e (MPa)	Reference
Spruce	N/A	Vacuum (0 % MC)	158 ^d	6 %	175	Adusumalli et al. (2010)
Loblolly pine	15°	38 % RH (8 % MC)	111 ^e	N/A	N/A	Zhang et al. (2010)
KerANJI	6°	38 % RH (8 % MC)	137 ^e	N/A	N/A	Zhang et al. (2010)
Norway spruce (normal wood)	5°	Vacuum (0 % MC)	147 ^e	3 %	220	Schwiedrzik et al. (2016)
Norway spruce (compression wood)	35°	Vacuum (0 % MC)	45 ^e	5 %	125	Schwiedrzik et al. (2016)
Beech	N/A	Vacuum (0 % MC)	183 ^f	3 %	270	Klímek et al. (2020)

^aMicrofibril angle (MFA). ^bExperiments performed in a vacuum were set at 0 % MC. MC values for experiments performed at a given RH were calculated using the same isotherm as that used in this study (Zelinka and Glass 2010). ^cValues estimated from stress-strain curves published in reference when available. ^dYield stress determined by intersection of lines drawn tangent to elastic and plastic deformations. ^eYield stress determined using the 0.2 % strain offset criteria. ^fYield stress determined from the point where the tangent distorted at an angle of 3° with respect to the initial linear portion of the stress-strain curve.

4.2 Plastic deformation mechanisms in wood cell walls

The nanoindentation H results in this study contributed new insights into the plastic deformation mechanisms in wood cell wall layers. Plastic deformation is irreversible and accommodated at the molecular scale by permanent rearrangement of the molecular structure. The mechanisms controlling the plasticity of wood cell walls have not been fully elucidated. Also, in wood a portion of the inelastic deformation is caused by viscoelastic, or time-dependent, recoverable strain, which is often neglected when discussing plasticity in wood cell walls. Nevertheless, in SCWL, initial yielding is proposed to be dominated by shear failures of the matrix polymers between cellulose fibrils (Fratzl et al. 2004; Hofstetter et al. 2008; Keckes et al. 2003). As deformation progresses, additional mechanisms, such as fibril buckling or kinking, have also been proposed (Adusumalli et al. 2010; Gindl and Teischinger 2002; Schwiedrzik et al. 2016). Hofstetter et al. (2008) suggested lignin controls yielding based on a favorable comparison of results from their multiscale mechanical model to experimental bulk wood strengths. Their model assumed that lignin controlled the bulk wood strength and had a shear strength of 20.2 MPa. They did not explore the possibility of other wood polymers, such as hemicelluloses, controlling bulk wood strength. Schwiedrzik et al. (2016) estimated a shear yield stress of 15 MPa for a wood polymer matrix based on micromechanical modeling and SCWL micropillar compression experiments. When it was assumed that lignin controlled the yielding, they calculated a 17 MPa stress in the lignin phase.

During the nanoindentation experiments in this study, the plastic deformations beneath the Berkovich probe were likely accommodated by multiple mechanisms. The 7–10 % effective strain beneath a Berkovich probe (Johnson 1985; Kermouche et al. 2008) was higher than the 3–6 % strain reported for the initial yielding in micropillar compression experiments (Table 2). Therefore, plastic deformation beneath the Berkovich probe likely progressed beyond the proposed initial yielding stage of shear failures in the matrix and likely also included deformation mechanisms, such as fibril buckling or kinking. By geometric necessity, at least some fibril buckling and kinking near the probe surface must have occurred to form clear residual nanoindentation impressions in the SPM images in Figure 2.

Despite the uncertainties in mechanisms controlling the plasticity in these Berkovich nanoindentation experiments, the observed trends in the MC-dependent H (Figure 4) and σ_H (Figure 7) data provided additional insights into the

complexities of plastic deformation in wood cell walls. The orientation effects support the differences in plasticity mechanisms in the SCWL when tested in the longitudinal or transverse directions. The sudden drops or inflections present in the data at approximately 2–4 % MC also indicate a likely moisture-dependent change in the plasticity mechanism. Although additional work is needed to better understand the mechanisms causing this transition in plasticity, its discovery in this work provides strong motivation for more future detailed study.

More detailed studies can be accomplished using advanced nanoindentation techniques like broadband nanoindentation creep (BNC). BNC assesses viscoplastic properties over 4–5 decades of strain rate in small volumes of polymeric materials like wood cell walls (Jakes et al. 2008b, 2012, 2024). BNC results obtained over a range of temperatures or moisture can be used to perform a thermal activation analysis to characterize the thermodynamic and physical nature of the plastic deformation mechanisms (Puthoff et al. 2009). Such experiments and analyses performed in different wood cell wall layers and orientations would provide valuable insights into the plastic deformation mechanisms in wood and how they may change with moisture, such as the 2–4 % MC transition observed in this study. Employing nanoindentation probes with different geometries, such as blunter and sharper pyramidal probes, can also facilitate the study of plasticity at different effective strains (Kermouche et al. 2008).

5 Conclusions

Nanoindentation was employed to measure the MC- and orientation-dependent elastic and plastic properties of SCWL in pine and bamboo. The MC-dependent pine CCML properties were also measured. As expected, the orientation of the cellulose fibrils with respect to the nanoindentation direction had the largest effect on the measured SCWL E_S^{NI} . However, the observed trends in the MC-dependent E_S^{NI} , including the slight increase in the SCWL longitudinal E_S^{NI} below 5 % MC and the continuous softening in the SCWL transverse E_S^{NI} , supported that lignin and hemicelluloses affect the SCWL elastic properties differently depending on the testing orientation. When tested in the longitudinal direction, lignin had a larger effect than hemicelluloses, whereas in the transverse direction, hemicelluloses had a larger effect. For plastic properties, trends in the MC-dependent data for both H and σ_H revealed a transition in behavior around 2–4 % MC. Although less sensitive than the

elastic properties, the plastic properties also had some orientation effects. More work is needed to better understand the mechanisms of plasticity in lignocellulosic cell walls.

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