Deformation and failure mechanism of secondary cell wall in Spruce late wood

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Abstract The deformation and failure of the secondary cell wall of Spruce wood was studied by in-situ SEM compression of micropillars machined by the focused ion beam technique. The cell wall exhibited yield strength values of approximately 160 MPa and large scale plasticity. High resolution SEM imaging post compression revealed bulging of the pillars followed by shear failure. With additional aid of cross-sectional analysis of the micropillars post compression, a model for deformation and failure mechanism of the cell wall has been proposed. The cell wall consists of oriented cellulose microfibrils with high aspect ratio embedded in a hemicellulose-lignin matrix. The deformation of the secondary wall occurs by asymmetric out of plane bulging because of buckling of the microfibrils. Failure of the cell wall following the deformation occurs by the formation of a shear or kink band.

1 Introduction

Softwood consists 95% of axially oriented 2 to 7 mm long cells known as tracheids and 5% of short parenchyma cells, mostly radially oriented in rays. Tracheids are interconnected through compound middle lamella. Each tracheid is composed of cell wall and lumen. The cell wall consists of a primary wall as outer layer and secondary walls S1, S2, and S3, successively positioned toward the lumen. It is found that S2 builds up 80% of the entire cell wall and is (especially in latewood) responsible for the extraordinary mechanical properties of wood [1]. The S2 wall layers consist of cellulose microfibrils in a matrix of lignin and hemicellulose, and hence can be thought of as a polymer-polymer composite [2]. The inclination of the microfibrils to the fiber axis is known as microfibril angle (MFA) and varies between 0 and 30° in S2 layers of normal wood [3].

Mechanical testing of individual fiber cells has been an area of active interest [4–7]. But, the mechanical behavior of individual cell wall components, especially the S2 wall (thickness of ~5 µm in latewood) is still an emerging area of research in wood mechanics. The hardness and elastic modulus of the S2 cell wall in latewood has been determined by nanoindentation [8]. Besides nanoindentation, another direct measurement on cell wall was reported recently in the form of cell wall bending, which was performed using an AFM tip mounted on a micromanipulator [9]. The cell walls were fabricated from the wood slice by using Focused Ion Beam (FIB) machining. While these nanoscale experiments provide useful insights into the S2 cell wall mechanics, it would be easier to interpret the mechanical response in a simple load case. Hence, in this study, the mechanics and deformation response of the S2 cell wall in latewood is studied by in-situ SEM compression of FIB machined micropillars.

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Fig. 1 (a) Overview showing locations used for FIB machining. (b) Pillar was milled exclusively from the S2 cell wall layer. (c) Micrograph showing complete pillar with a smooth surface obtained by tilting specimen by 50°. (d) High magnification image showing the thin Au-Pd coating on the top surface of the pillar

2 Materials and methods

2.1 Preparation of cell wall pillars

Longitudinal specimens of round spruce wood blocks (diameter ~ 3 mm; length ~ 8 mm) with a moisture content of about 6% were microtomed across the cell diameter using a Reichert–Jung Polycut E. Successive sections of 100, 10, and 1 µm thickness were cut from top surface using diamond knife with the speed of 500, 1000, 2000 rpm, respectively. Smooth surfaces of cell wall cross-sections were achieved after ~ 5 minutes sectioning of 1 µm slices. After microtoming, rectangular blocks are milled out of the round blocks using a razor blade. Specimens were coated with 10 nm thick gold-palladium to minimize charging and e-beam damage [10]. Cylindrical pillars with diameter of 3 µm and aspect ratio of ~ 2.5 were FIB machined from the longitudinal-radial side of the latewood cell walls using a Tescan Lyra FIB (Fig. 1). The FIB machining was carried out in two steps using a Ga^+ beam with energy of 30 keV. Initially, rough milling was performed with a high Ga^+ beam current

(3500 pA) to obtain a pillar of ~ 8 µm diameter in a crater of ~ 40 µm. Next, fine polishing was performed with low Ga^+ beam current (75 pA) to minimize beam damage and obtain the final pillar.

2.2 Mechanical testing of cell wall pillars

The micropillars were compressed in displacement control (strain rate of 10^{-3} /s) using an indigenously built in-situ SEM (Zeiss DSM962) nanoindenter. A detailed description of the in-situ SEM micro-compression is available elsewhere [11]. A conical flat punch diamond indenter with tip diameter of around 6 µm was used for compression of the micropillars. The micropillars were imaged a priori and post compression by a cold emission SEM (Hitachi 4800) for studying the cell wall deformation and failure mechanisms. Post compressed pillars were also imaged by field emission SEM after cutting in half (perpendicular to the diameter) using the FIB. All measurements were carried out in an environment of 10^{-6} mbar and 23°C implying that the micro-compression was carried out with 0% moisture content of the cell wall.

Fig. 2 Engineering stress-strain curve of the pillar milled from S2 cell wall. The stress drop in the stress-strain curve corresponds to the kink or shear band indicated in the micrograph. The yield point is calculated by considering the intersection of tangents drawn to the elastic and plastic regimes as shown in the micrograph

Fig. 3 Distinct biphasic behavior was observed in the stress-strain response of a pillar. Post compression HRSEM micrographs of opposite faces of the pillar revealing (a) uniform bulging of the pillar and axial cracks running across the surface wrinkles and (b) Axial crack in a plausible bordered pit. The corresponding in-situ SEM micropillar compression video is provided as supplementary information

3 Results and discussion

Engineering stress-strain curves obtained from in-situ SEM compression of micropillars FIB machined in the S2 cell wall are shown in Figs. 2–4. Yielding is observed as a distinct change in slope from the elastic regime, followed by an apparent “strain-hardening” due to an increase in cross-sectional area of the pillar. A distinct biphasic behaviour can be observed in the stress-strain curve shown in Fig. 3. The biphasic response is characterized by a distinct second change in slope following the primary or first yield point [12, 13]. In accordance to Keckes et al. [14], we assume that the biphasic behavior is observed due to misorientation of the microfibrils.

The yield strength (σ_y) of the cell wall was estimated to be 158 ± 21 MPa (average of five pillars) by considering the intersection of tangents drawn to the elastic and plastic regimes. These values are in good agreement with the values obtained by Gindl and Teischinger [15], who estimated the cell wall strength of Spruce wood in axial compression by considering the cell wall density. They also showed that the compression strength of the cell wall does not depend on MFA. The hardness (H) of Spruce wood-S2 cell wall has been estimated to be 0.47 ± 0.2 GPa [16]. Therefore, the H/σ_y ratio of the S2 cell wall is approximately three, which is comparable to metals. This analysis reveals that the composite nature of the dry cell wall allows accommodation of larger plastic deformation than previously imagined [17].

Fig. 4 Engineering stress-strain curve of a pillar showing severe bulging and axial cracks running across the surface wrinkles. Micro-compression was performed at 0% moisture content

Fig. 5 Simplified model of defect free S2 cell wall (pillar) deformation during micro-compression. Since the model is proposed considering the S2 cell wall as fiber reinforced composite in which fibers are aligned parallel to the loading axis (microfibril angle is 0°), deformation occurred by out-of-plane buckling and subsequent shear failure. Surface wrinkles formed on the surface of the pillar are due to the FIB induced damage

As mentioned in literature [15, 18], the cell wall can be thought of as a fiber-reinforced composite, where the microfibrils are reinforcing the matrix of hemicellulose and lignin. Post-compression HRSEM imaging of the micropillars reveals bulging and a wrinkle pattern on the surface (insets in Figs. 2–4). It is interesting to note that the deformation mechanism is not axisymmetric (Fig. 3b). In addition to a finite MFA effect, this could be due to structural inhomogeneities such as a narrow bordered pit, which tears and forms a crack visible on the surface. The corresponding in-situ SEM micropillar compression video is provided as supplementary information.

The out-of-plane bulging of the pillar suggests that buckling of the microfibrils plays a critical role in the deformation of the cell wall. This can be understood as follows: the cellulose microfibrils (crystalline component) are stiffer than the hemicellulose-lignin matrix (amorphous components) [2]. Hence, while the microfibrils are susceptible to buckling because of their high aspect ratio, their high volume fraction and bonding with the softer amorphous matrix leads to uniform out of plane bulging of the pillar. Formation of a kink or shear band may be due to failure of the

matrix, which also leads to transverse fracture of the microfibrils. Figure 5 shows a simplified schematic illustration of the deformation and failure sequence of the S2 cell wall pillars (Figs. 2–4). Further evidence to support this hypothesis was obtained by HRSEM imaging of longitudinal cross-sections (FIB machined) of pillars exhibiting severe bulging (Figs. 6a and b). The cross-sections reveal heterogeneous pockets, which expose the buckled microfibrils.

Longitudinal cross-sections of the pillars were FIB machined to understand the unusual wrinkle patterns that form on the surface of the pillars post compression. Figure 6(c and d) shows that the wrinkles are formed only as film at the surface of the pillar. This external skin formation may be a result of FIB induced damage caused by interaction of Ga^+ ions with the hemicellulose and lignin. It has been shown that FIB machining of micropillars in GaAs leads to the formation of an amorphous layer approximately 10 nm thick [19]. Though the influence of a thin amorphous skin during compression is expected to be negligible, the impact of Ga^+ damage is a subject of future studies [20]. Additionally, microcracks observed on the wrinkled surface are restricted to the surface (Fig. 7a and b). These axial microcracks formed

Fig. 6 HRSEM micrographs of longitudinal FIB cross-sections of the deformed pillar (shown in Fig. 4) revealing (a) Two distinct pockets corresponding to the severe bulging and (b) Protrusion of micro-fibrils normal to the plane of the image (c) Skin-restricted nature of the wrinkle pattern and (d) Wavy nature of surface wrinkle pattern. White ovals in (a) and (c) are added to aid explanation in caption

on the wrinkle pattern occur due to the circumferential or Hoop stress developed on the surface as a result of pillar bulging.

It is important to note that a load drop in stress was observed occasionally within the plastic regime suggesting localized failure (Fig. 2). The formation of a kink or shear band at approximately 45° to the axis of the pillar is thought to be the failure mechanism of the cell wall, which gives rise to the stress drop in the plastic regime (inset of Fig. 2 and Fig. 7c). The formation of a kink or shear band is known as the failure mechanism of fiber-reinforced composites in compression [21], as well as in the longitudinal compression of macroscopic wood blocks [22]. Poulsen et al. [23] have shown that the incipient kinking in macroscopic Spruce wood specimens during compression occurs due to large local misalignments of the fibers. This analogy can be extended to the failure of the S2 cell wall, which is also a fiber-reinforced composite at a much smaller length scale. Thus, the shear band formation occurs as a result of failure of the hemicellulose-lignin matrix, which is aided by the buckling and misorientation of the microfibrils.

Fig. 7 HRSEM micrographs showing wavy nature of surface wrinkles (a, b), axial cracks in the wrinkle pattern restricted to the surface (b) and distinct shear band formation (c) shown by arrow in (a)

4 Conclusion

The deformation and failure response of S2 cell wall was studied by compression of FIB machined micropillars. The S2 cell wall revealed a biphasic engineering stress-strain response on compression and large scale plasticity. The deformation and failure of the S2 cell wall micropillar occurs by out of plane bulging and shear band formation, respectively. A simplified model for the deformation and failure sequence is discussed by considering the S2 cell wall as fiber reinforced composite in which fibers are aligned parallel to the loading axis.

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