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NANOMECHANICAL BEHAVIOR OF WOOD CELL WALLS OBSERVED BY DIFFERENT INDENTATION LOADING PREREQUISITES

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ABSTRACT

The variations of nanomechanical behavior of wood cell walls under different peak loads, loading times, and holding times were studied. Samples were separately loaded to preset peak loads of 100, 150, 200, 250 and 300 μN. Changes in the micromechanical properties were tracked in the longitudinal direction to determine change values of the elastic modulus and hardness. Moreover, the creep behavior was also analyzed under different holding times. It was found that the longer the holding time, the larger the creep ratio of all of the samples, and the creep rate decreased slowly with longer loading times. Finally, when the peak load was larger, the displacement rate and strain rate increased, but the strain rate in each test exhibited a tendency to become constant after 10 s.

KEYWORDS: Cell wall, creep behavior, elastic modulus, hardness, mechanical properties, nanoindentation.

INTRODUCTION

Nanoindentation technology was originally proposed by Oliver and Pharr (1992) and its effect was tested by nanoscale indentation load, displacement relation, and the method chosen to analyze the mechanical properties of the material. The significant features of this technology are the high resolutions of force and displacement. During testing, a computer records the load and head displacement at the same time, and automatically calculates the elastic modulus, relaxation modulus, hardness, and creep values (Rho and Pharr 1999). Wimmer et al. (1997) first proposed the use of nanoindentation technology to measure the longitudinal modulus of the secondary wall of wood tracheids. The longitudinal elastic modulus of a tracheid chord and its distribution in the direction of cell-wall thickness were studied. The key parameters were measurement values of the hardness and elastic modulus of the tracheid walls in the longitudinal direction of spruce wood, specifically peak load, the depth at peak load, and the initial unloading contact stiffness.

When applied to testing the hardness and elastic modulus of wood secondary walls, peak load must be determined and then a formula is selected based on the test curve model followed by determination of the loading and unloading times. Therefore, the values of the peak load and indentation depth are crucial to the accuracy of the experiment. Gindl and Schoberl (2004) discovered the correlation rule of the longitudinal modulus and hardness of the S2 layer of the secondary wall in the tracheid of spruce wood. In the range 0–50 nm, the longitudinal hardness and elastic modulus increase sharply with increasing indentation depth. The better the quality of the sample surface, the smaller the range of the non-reliable work area. In terms of the surface quality of samples, when the indentation depth is greater than 50 nm, the influence of the surface quality of the sample on the experimental results can be ignored. The surface quality of the cell wall has an impact on the test data, and the indentation depth of testing is important. Jiang et al. (2004), Wimmer et al. (1997), and others have conducted research in this field but these studies did not deal with the creep properties of wood cell walls.

Creep testing according to general procedures requires many standard-sized samples and thus is somewhat time-consuming. In this regard, there have been many attempts to estimate creep behavior through simple indentation experiments. The indentation creep tests have many advantages, e.g., the testing procedure is simple and easy to set up and only a small sample is needed. However, one of the most powerful advantages may be the fact that one can estimate the small-scale creep properties and their local change through the test, which is applicable not only to micro- or nanoscale structures in the electronics industries, but also to relatively large-scale components, such as the weld heat-affected zones in which complex microstructural gradients exists. Moreover, the importance of small-scale creep research has been continuously increasing in these days of the "nano-age" because the creep becomes more active on the nanoscale (Wang et al. 2007, Guisbiers and Buchaillot 2008, Wang et al. 2009). Indentation creep research accelerated in the late 1980s with the development of instrumented indentation techniques, which made it possible to systematically investigate the time-dependent mechanical response by analyzing the indentation load-displacement (P-h) curves without the observation of hardness impressions (Pethica et al. 1983, Oliver and Pharr 1992, Oliver and Pharr 2004).

In the tests, the variations of the hardness and elastic modulus of wood cell walls under different peak loads and different loading times were studied first. Second, the creep behavior of wood cell walls under different peak loads and holding times were studied. Finally, the creep rate, indentation displacement rate, and creep strain rate were studied. The issues raised in precisely estimating small-scale hardness, elastic modulus, and creep properties are discussed in this paper, and an attempt is made to propose possible novel ways of obtaining more reliable data.

MATERIAL AND METHODS

Pinus massoniana Lamb. is a fast-growing softwood species that is mostly planted across southern and eastern China. In this research, a 40-year-old *Pinus massoniana* Lamb. was chosen from Nanping, Fujian Province, China. Specimens with dimensions of 10×8×8 mm in longitudinal, radius, and tangential directions, respectively, were prepared from one log. A total of 15 specimens were prepared for nanoindentation and obtained from the same latewood growth ring. The surface of the sample was polished using an ultrathin-section machine. The surface of the sample was then repaired with a glass knife and finally smoothed by diamond knives (Habelitz et al. 2001, Sunita et al. 2004).

Methods

Elastic modulus and hardness

The bottoms and tops of the blocks were parallel to each other and exactly perpendicular to the cell walls' longitudinal axes. All of the samples were placed on a flat stage and a straight-edge was used to verify that all of the sample angles were 90°. Then, all of the qualified samples were divided into different groups for testing different loads, loading times, and holding times. The specimens were fixed onto a metal sample holder and made into a pyramid shape. Finally, the apex was smoothed with a diamond knife in an ultramicrotome. Specimens were prepared following the procedure described by Meng et al. (2013). On the basis of the theory of nanoindentation, the hardness (H) and reduced elastic modulus (Er) were calculated from the load-displacement data by the following equation for hardness, described by Oliver and Pharr (1992):

$$H = \frac{P_{\max}}{A} \qquad \text{(GPa)} \qquad (1)$$

where: $P_{\max}$ - the peak load determined at a maximum depth in an indentation cycle, (μN)
A - the projected contact area between indenter and sample, (nm^2)

The sample's elastic modulus (*Er*) was calculated as follows:

$$Er = \frac{\sqrt{\pi}}{2\beta} \frac{S}{\sqrt{A}} \qquad \text{(GPa)} \qquad (2)$$

where: S (stiffness) - the slope of the line of the unloading curve in the load-displacement plot, (GPa)
β - correction factor correlated to indenter geometry (β=1.034 for a Berkovich indenter),
A - the projected contact area (nm^2).

Viscoelastic properties during nanoindentation

Viscoelastic material exhibits both viscous and elastic behavior. Samples were loaded to the preset peak load within certain period of time, followed by a certain holding time during indenting. The indentation creep ratio (C_i) was defined as the relative change in indentation depth while the applied load remained constant during the holding time, and it can be calculated as follows (Konnerth and Gindl 2006):

$$C_i = \frac{h_2 - h_1}{h_1} \times 100 \qquad (\%) \qquad (3)$$

where: h_2 - the final penetration depth at the end of the holding segment, (nm)
h_1 - the depth at the end of the loading segment, (nm)

The nanoindentation displacement rate ( ) is expressed by:

$$\dot{h} = \frac{dh(t)}{dt} \qquad (\mathrm{nm \cdot s^{-1}}) \tag{4}$$

where: h - the instantaneous indenter penetrating depth, (nm)
t - loading time, (s)

The indentation strain rate ε_1 was calculated according to Wang et al. (2009):

$$\varepsilon_1 = \frac{1}{h}\frac{dh}{dt} \qquad (\mathrm{s^{-1}}) \tag{5}$$

where: h is the indenter displacement monitored during creep, (nm).

RESULTS AND DISCUSSION

Cell-wall modulus and hardness

The cell-wall thickness of *Pinus massoniana* Lamb latewood was approximately 5-13 μm for nanoindentation tests. Several indentations are marked by green circles in Fig. 1. Each sample was loaded to preset peak loads of 100, 150, 200, 250, and 300 μN within 5 s, followed by a holding time of 5 s during indenting, then unloading within 5 s.

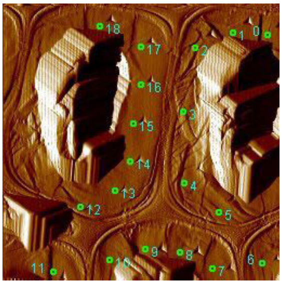

Fig. 1: Images of scanning probe microscopy of wood cell wall of Pinus massoniana Lamb.

Fig. 2 presents the typical load-displacement curves in nanoindentation tests under different peak loads.

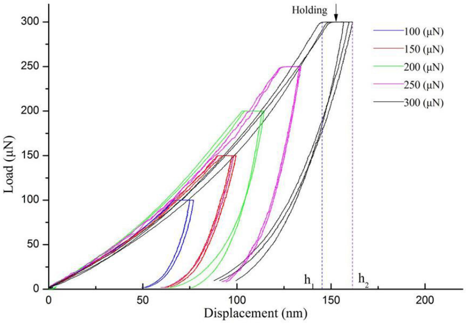

Fig. 2: Typical load-displacement curves of wood cell wall under different peak loads.

The summary of the measurements with the average modulus (Er) values of the secondary cell wall is shown in Fig. 3. The test value of the modulus varied with several factors, including the surface roughness of wood, different indentation loadings, surface hygroscopy, residual stress, and determination of the contact zero point of the test (Alcala et al. 2002, Cheng and Cheng 1998, ISO 14577:2002).

Fig. 3 shows the results of the indentation experiments. The elastic modulus was 17.27 0.52 GPa and the hardness 0.488 0.017 GPa under a peak load of 250 μN. These results are in accord with similar effects described and discussed in the literature (Yan et al. 2011).

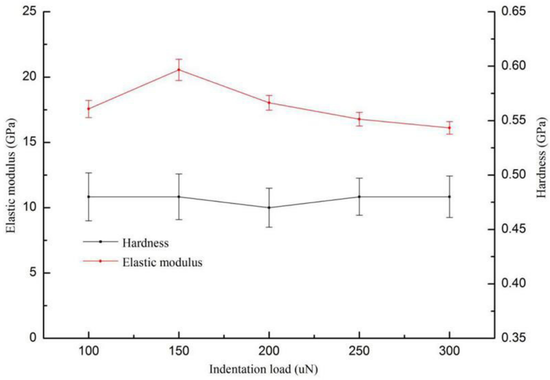

Fig. 3: Mechanical properties of wood cell walls under different peak loads.

An empirical formula to estimate the hardness and elastic modulus of wood was given in Yan's study on the longitudinal mechanical properties of the cell wall of *Pinus massoniana* Lamb. as related to moisture content and measured hardness and elastic modulus of the cell walls, as follows:

$H = 0.769 - 0.02\mathrm{x}$
$R^2 = 0.98$

where: H - the hardness of the cell walls of *Pinus massoniana* Lamb.,
x - their moisture content,
R^2 the coefficient of determination of the formula.

In addition,

$E_r = 23.16 - 0.45\mathrm{x}$
$R^2 = 0.759$

where: Er - the elastic modulus of the cell walls of *Pinus massoniana* Lamb.,
x - their moisture content,
R^2 - the coefficient of determination.

When the peak load increased from 100 to 300 μN, the indentation load has almost no influence on the hardness of all of the samples, as can be seen from Fig. 3.

Kollmann and Cote (1968) reported a strong relationship between maximum crushing strength and hardness. Janka evaluating the results of hardness tests of 280 wood species, found an empirical relationship between hardness (*H*) and crushing strength (σ): $H=(2\sigma-0.05)$ (GPa). The peak load has no effect on the hardness. Hardness is only related to the crushing strength of the cell walls of wood. Similar effects have been described and discussed in the literature (Kollmann and Cote 1968). The test values of the modulus were improved when the indentation peak load was changed from 100 to 150 μN, but the change was not obvious when the indentation peak load was changed from 150 to 300 μN. Tze et al. (2007) found an empirical relationship between modulus and displacement from the curve. The displacement also increased when the indentation peak load was changed from 100 to 150 μN, and then the modulus improved. Although the displacement increased, the modulus changed little when the indentation peak load was changed from 150 to 300 μN. Similar effects have been described and discussed in the literature (Tze et al. 2007).

In the test, each sample was loaded to preset loading times of 10, 50, 100, 200, and 300 s with a peak load of 250 μN, followed by a holding time of 5 s during indenting and then unloading within 5 s. Fig. 4 presents the typical load-displacement curves in nanoindentation tests under different loading times. The test values of the modulus were not obviously changed when the indentation loading time was changed from 10 to 300 s. The test value of hardness was 0.488 GPa when the indentation loading time was 10 s. The values of hardness were 0.601, 0.582, 0.586, and 0.588 GPa for indentation loading times of 50, 100, 200, and 300 s, respectively.

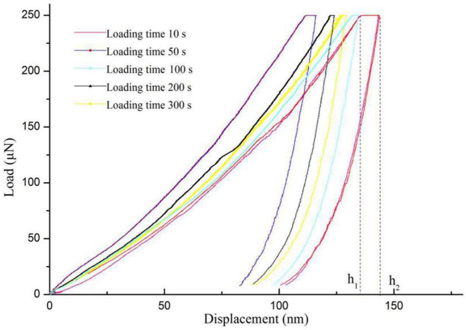

Fig. 4: Typical load-displacement curves of wood cell wall under different loading time.

The change was not obvious after the loading time increased to 50 s, as shown in Fig. 5.

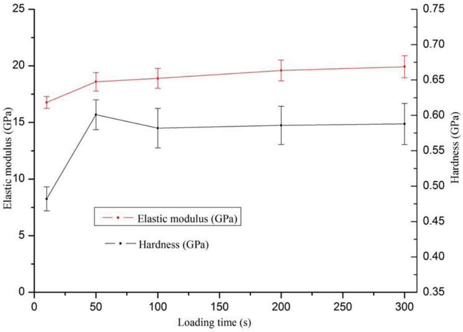

Fig. 5: Mechanical properties of wood cell walls under different loading time.

Creep behavior under different holding times

Samples were loaded to the preset peak load of 250 μN within 5 s, followed by separate holding times of 5, 10, 50, 100, 200, and 300 s; the creep depth increased with increasing holding time. Creep behavior is evident from Fig. 2 that creep occurs even in the elastic regime and that the observed creep behavior is mainly time-dependent plastic deformation. In conventional loading-unloading indentation tests, wood samples exhibit creep behavior during the application of different peak forces. The investigation of creep behavior in this study was restricted to constant loading force and strain rates were determined by means of the indentation holding section data. As illustrated in Fig. 6, there are different creep ratios under different holding times; the longer the holding time, the larger the creep ratio of all samples.

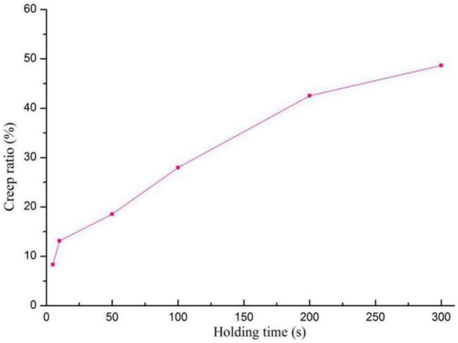

Fig. 6: Creep ratios of wood cell walls under different holding times.

At the same time, with longer loading times, the creep rate increased slowly. When the loading time exceeded 200 s, the creep rate changed only slightly. In Fig. 6, the low creep rates after 200 s prove that the late stage of the holding steps of these indentations have eliminated most of the creep component of the cell walls. Similar effects have been reported in the literature (Xing et al. 2016).

Creep behavior under different peak loads

Samples were separately loaded to preset peak loads of 100, 150, 200, 250, and 300 μN within 5 s, followed by a holding time of 5 s during indenting. The indentation displacement rate was defined as in Eq. 4. Fig. 7 presents the typical holding displacement rate curves in nanoindentation tests under different peak loads, and it shows that the real-time displacement changes intuitively. The indent displacement rates for each test presents different curves under different peak loads during the indentation holding step; the larger the load, the higher the displacement rate. The indent displacement rates for each test exhibited a tendency to become constant after 10 s, as shown in Fig. 7, proving that the holding steps of these indentations eliminated most of the creep component of the cell walls. Similar effects have been reported in the literature (Xing et al. 2016).

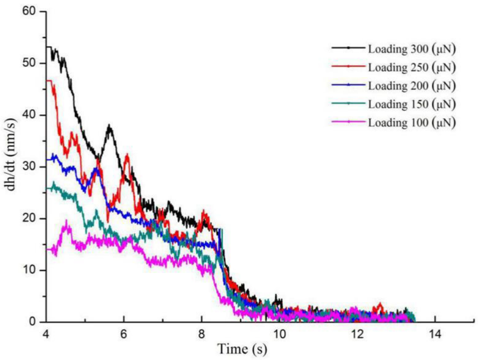

Fig. 7: Displacement-rate curves of wood cell wall under different peak loads during indentation holding step.

Strain rate is an important form of creep behavior. Fig. 8 shows that the indentation creep strain rate at the holding stage changed significantly as a function of time; in addition, the creep strain rate dropped quickly at the beginning and then became stable. Fig. 8 also indicates that different loads lead to different strain rates of wood cell walls.

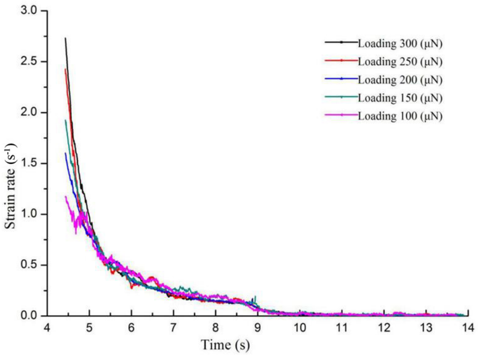

Fig. 8: Strain rate curves of wood cell wall under different peak loads.

At the same time, the figure also shows the low displacement rates after 10 s at different test peak loads, further proving that the loading and holding steps of these indentations have eliminated the highest creep component of the cell walls. At the loading stage, the strain rate decreases most rapidly, and the strain rate decreases slowly at the holding stage (5-10 s). Similar effects have been described and discussed in the literature (Wikberg and Maunu 2004, Jalaludin et al. 2010, Yin et al. 2011, Li et al. 2016).

CONCLUSIONS

The indentation peak load and indentation loading time had influence on the hardness, modulus, and creep of all of the samples tested. Regarding the results obtained, the modulus of the cell walls showed an obvious increase when the indentation peak load was increased from

100 to 150 μN, and then it remained constant from 150 to 300 μN. The indentation peak load had no influence on the hardness of all of the samples. Both modulus and hardness showed an obvious increase when the indentation loading time was changed from 10 to 50 s, and then remained constant from 50 to 300 s. Regarding creep behavior, the longer the holding time, the larger the creep ratio of all of the samples. The indentation displacement rate and the creep strain rate increased with increasing peak load and then remained constant after 10 s.

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