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ORIGINAL ARTICLE

Kentaro Abe · Hiroyuki Yamamoto

The influences of boiling and drying treatments on the behaviors of tension wood with gelatinous layers in *Zelkova serrata*

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Abstract This study examined how boiling and drying treatments influenced various physical properties of the tension wood with gelatinous fibers (G-fibers) of a 29-year-old *Zelkova* branch. By boiling treatment, tension wood with numerous G-fibers contracted considerably in the longitudinal direction and the longitudinal Young's modulus decreased in spite of the water-saturated condition. The drying treatment caused green tension wood and boiled tension wood with numerous G-fibers to shrink longitudinally and increased their longitudinal Young's moduli. These specific behaviors in tension wood were highly correlated with the proportion of G-fibers in a specimen and were probably caused by the microscopic behavior of cellulose microfibril (CMF) in the gelatinous layers (G-layers). The longitudinal shrinkage of tension wood due to drying suggests the existence of a hygro-sensible, noncrystalline region in the CMF, which is abundant in the G-layer. Furthermore, the noncrystalline region in the CMF softens during boiling treatment, resulting in the reduction of the longitudinal Young's modulus in tension wood. The longitudinal contraction of tension wood with G-fibers by boiling might be caused by the tensile growth stress remaining in green G-layers. However, no changes were detected in the 004 *d*-spacing of cellulose crystal in tension wood from the boiling and drying treatments, regardless of the proportion of G-fibers.

Key words Tension wood · Gelatinous fiber · Gelatinous layer · Cellulose microfibril · *Zelkova serrata* M.

K. Abe · H. Yamamoto
Graduate School of Bioagricultural Sciences, Nagoya University, Nagoya 464-8601, Japan

K. Abe (✉)
Research Institute for Sustainable Humanosphere, Kyoto University, Uji, Kyoto 611-0011, Japan
Tel. +81-774-38-3657; Fax +81-774-38-3658
e-mail: abekentaro@rish.kyoto-u.ac.jp

Introduction

Hardwood species form tension wood on the upper side of branches and leaning stems to maintain mechanical strength against gravity. Tension wood forms gelatinous layers (G-layers) inside the secondary wall, and the structure and specific functions of the G-layer have been investigated for many years. Tension wood with G-layers have distinctive properties compared with normal wood, e.g., high tensile growth stress,[1–6] large longitudinal shrinkage due to drying,[4,6,7] and high longitudinal Young's modulus.[4] The combined effects of these properties often causes various defects during wood processing, and some studies have attributed the distinctive tension wood characteristics to the G-layer.[1–3,6] Therefore, determining the specific properties of tension wood and the G-layer would improve not only the processing efficiency but also the novel use for a wide range of hardwood species grown under various environmental conditions.

Reports have shown that the G-layer mainly consists of cellulose[8] and is free from lignin deposition.[9,10] Although some researchers have reported the presence of lignin in the G-layer,[3,11–13] the principal component of the G-layer is cellulose microfibril (CMF). Therefore, it is quite natural to assume that the specific behaviors of tension wood are caused by the characteristic properties of the CMF in the G-layer. However, compared with research on normal wood, studies on the physical properties of the CMF in the G-layer in tension wood remain scarce.

The purpose of this study was to examine the fine structures and properties peculiar to the CMF constituting the G-layer. We examined how boiling and drying treatments affect the behaviors of the G-layer, specifically, the changes in the longitudinal Young's modulus and longitudinal dimension. Ideally, the best way is to measure these behaviors directly by isolating the G-layer from the tension wood specimen. However, it is almost impossible to strip the G-layer from the secondary wall without imparting chemical or physical damage. Even if G-layers were to be successfully isolated without any damage, it would be difficult to mea-

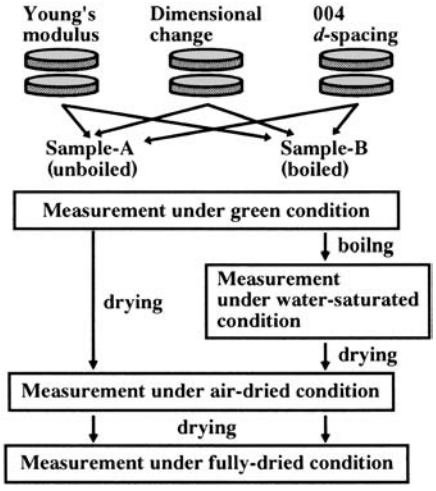

Fig. 1. Sample preparation and analysis

sure the Young's modulus from such an extremely small sample. Thus, in the present study, the influence of boiling and drying treatments on the physical properties of tension wood was investigated in relation to the proportion of G-fibers in a specimen. The study revealed the physical properties of the G-layer as well as the CMF in the G-layer. Furthermore, to investigate how boiling and drying treatments affect the CMF in the G-layer, posttreatment changes in the 004 *d*-spacing of cellulose crystal in tension wood were measured by X-ray diffractometry.

Experimental

Sample preparation

A branch of a 29-year-old mature *Zelkova serrata* M. (keyaki), 11.2 cm in diameter and cut to 105 cm in length, was examined. Six disks were cut from the branch, and two disks were assigned to each measurement of the longitudinal Young's modulus, longitudinal dimension, and 004 *d*-spacing (Fig. 1). Specimens were prepared from eight points around the circumference of each disk. The specimen sizes for the longitudinal Young's modulus and longitudinal dimension measurements were 60 (L) × 9 (T) × 4.5 (R) mm and 54 (L) × 10 (T) × 5 (R) mm, respectively. Crosscut sections (5 mm thick) were used for the 004 *d*-spacing measurement.

For the specimens prepared from one of the two disks, each measurement was performed when green, air-dried, and then fully dried conditions at room temperature (20°C). Air-dried and fully dried specimens were prepared for 1 month in closed chambers using a NaCl-saturated solution and P_2O_5 powder, respectively. The specimens measured according to this procedure are hereafter called Sample A. In order to investigate the influence of boiling on tension wood with G-fibers, the specimens prepared from other disks were boiled for 20 min after measuring in the green condition; specimens were held in a closed chamber with H_2O until reaching room temperature and then the measurements were performed again. The procedure thereafter was similar to that for Sample A, and the specimens in this procedure are hereafter called Sample B. X-ray measurement was performed in the water-saturated condition instead of the green condition for both samples.

The weight of each specimen was measured immediately before the measurements of Young's modulus, longitudinal dimension, and 004 *d*-spacing. After all measurements had been completed, each specimen was dried at 105°C for 24 h and then weighed. The average equilibrium moisture contents of the specimens in the air-dried and fully dried conditions were 14.3% and 0.65%, respectively.

Longitudinal dimensional change

The longitudinal dimensions of specimens under each moisture condition were measured using a dial-gauge comparator with a reading accuracy of 0.001 mm.[14] The drying shrinkage under each moisture condition for Sample A was calculated based on the length of the specimen in the green condition. For Sample B, the dimensional change due to boiling and the drying shrinkage were calculated based on the length of the specimen in the green condition and the length after boiling treatment, respectively.

Longitudinal Young's modulus

Tensile tests were performed under each moisture condition using a material testing machine. For each specimen, two strain gauges were bonded at the center of both flat-sawn grain surfaces. Both ends of the specimens were attached to the testing machine chucks, and then a tensile load was applied in the longitudinal direction. The longitudinal Young's modulus was calculated based on the average strain at both sides according to a strain meter.

Longitudinal *d*-spacing

The 004 *d*-spacing of each specimen was measured to examine the longitudinal dimensional change in the cellulose crystal in tension wood with G-layers by boiling and/or drying using an X-ray diffractometer (XD-D1w; Shimadzu, Kyoto, Japan) reflection technique. The incident X-ray used CuKα radiation ($\lambda = 0.154$ nm) at a power of 35 kV, and 35 mA passed through a Ni filter. The scattering angles (2ψ) for the 004 reflections ranged from 32° to 37°. To prevent changes in moisture content during the X-ray measurements, moisture-conditioned air was circulated through the sample cell.[15] The *d*-spacings were calculated using the Bragg equation based on the peak position of the (004) reflection in the obtained equatorial diffraction curve.

Area percentage of G-fiber

After all measurements were completed and the oven-dried weights of specimens were measured, the area percentage

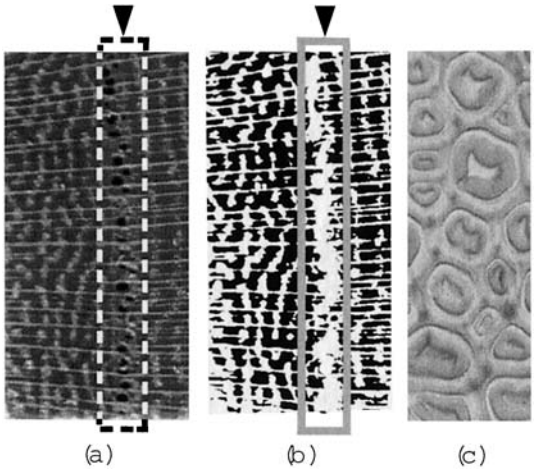

Fig. 2. **a** Captured image and **b** binarized image of a specimen. Vessel regions were corrected to white regions (*arrowhead*). **c** G-fibers observed under a light microscope

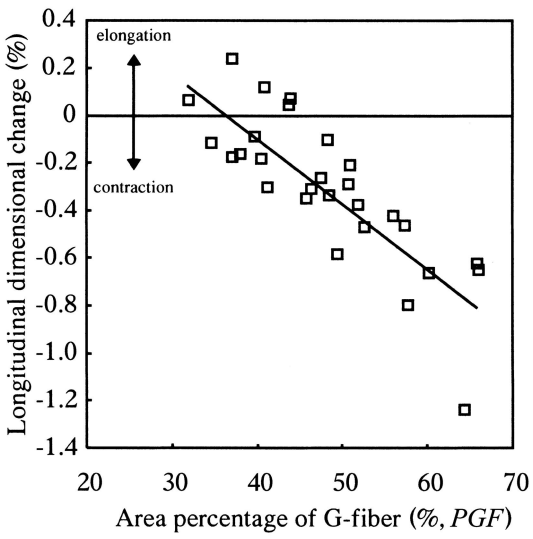

Fig. 3. Relationship between longitudinal dimensional change from green to water-saturated condition due to boiling and area percentage of G-fiber for Sample B

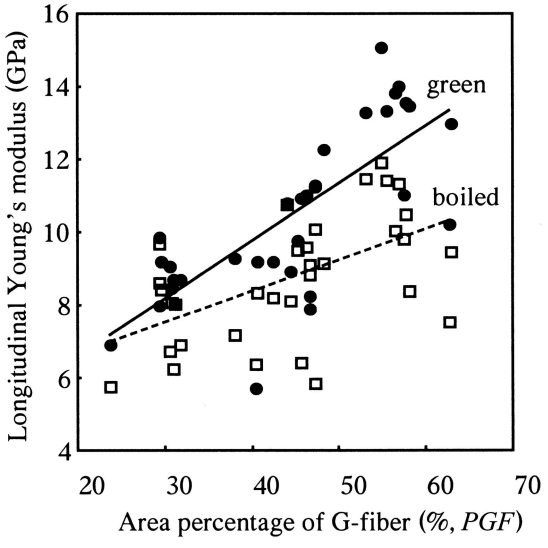

Fig. 4. Relationships between the longitudinal Young's modulus and the area percentage of G-fiber for green specimens and boiled specimens of Sample B

of G-fibers to the whole cross-sectional area (PGF) of each specimen was measured. In the cases of specimens measured for longitudinal dimensional change and the Young's modulus, the crosscut plane was prepared by cutting the center of a specimen, and then directly transformed into image data with an image scanner.

The regions containing the G-fibers were distinguished by the binarization of the captured image of a specimen using NIH Image (v. 1.63; National Institutes of Health, Bethesda, MD, USA) image processor; PGF of the specimen was then determined. However, because this method cannot distinguish the regions with G-fibers and vessels, the effect from vessels was manually eliminated. Figure 2 shows the captured scanner image of a specimen and its binarized image. Furthermore, thin crosscut sections prepared using a sliding microtome from some specimens were stained with both safranine and fast green and then observed under a light microscope. Thereby, the G-fibers with thick G-layer were clearly observed, as shown in Fig. 2c, and it confirmed that the G-fiber domains in the binarized image observation were consistent with those observed by light microscope for the same specimen.

Results

The relationships between the various physical properties of tension wood and the area percentage of G-fiber (PGF) were clarified to investigate the influences of boiling and drying treatments.

Influence of boiling

Figure 3 shows the relationships between the longitudinal dimensional change from the green to water-saturated condition due to boiling and PGF for Sample B. For the tension wood with a large amount of G-fiber, the longitudinal contraction due to boiling was clearly observed in spite of water-saturated condition, increasing with PGF. On the other hand, the longitudinal elongation due to boiling sometimes occurred when PGF was below 45%.

Figure 4 shows the relationships between the longitudinal Young's modulus and PGF for Sample B. The longitudinal Young's moduli of the green specimen and the boiled specimen clearly increased with PGF. However, the slope of the regression line for the boiled specimen was clearly smaller than that for the green specimen, indicating that boiling treatment decreased the longitudinal Young's modulus of the G-fiber in the tension wood.

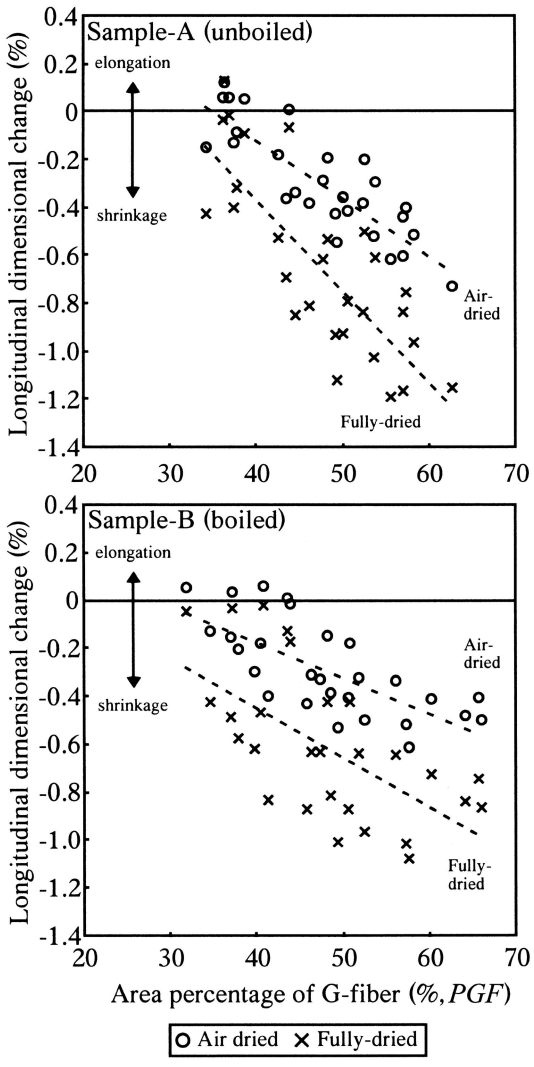

Fig. 5. Relationships between the longitudinal dimensional change due to drying shrinkage and the area percentage of G-fiber for Sample A (unboiled; *upper*) and Sample B (boiled; *lower*). The dimensional changes of Sample A and Sample B were calculated from green condition and the boiled condition, respectively

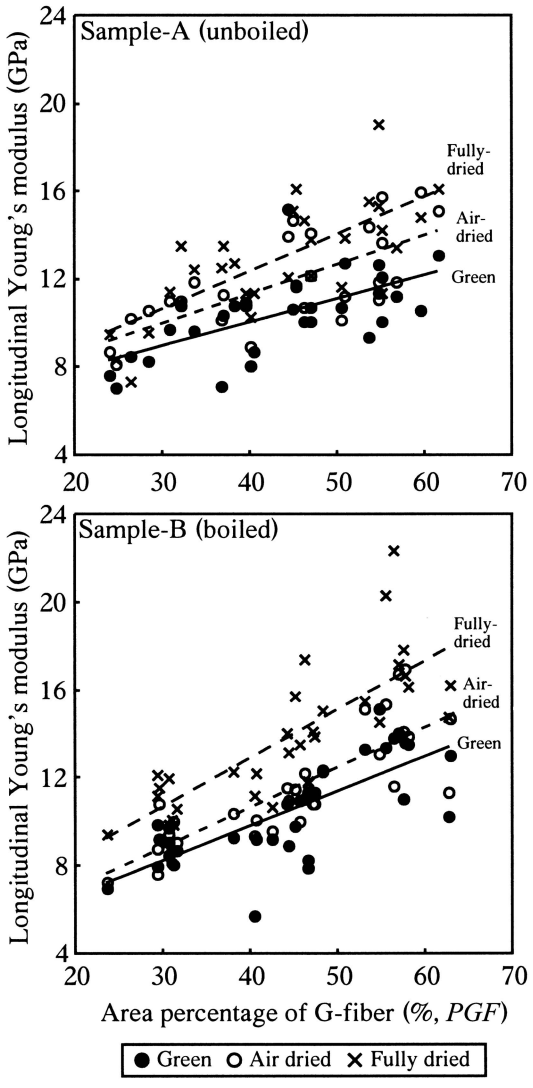

Fig. 6. Relationships between the longitudinal Young's modulus under each condition and the area percentage of G-fiber for Sample A (unboiled; *upper*) and Sample B (boiled; *lower*)

Influence of drying

Figure 5 shows the relationships between longitudinal dimensional change due to drying and PGF for Sample A and Sample B. The shrinkages for Sample A and Sample B under each moisture condition were measured based on the length of the specimen when green and the length after boiling treatment, respectively. The longitudinal shrinkages under both air-dried and fully dried conditions increased with increasing PGF for both samples, and the shrinkages for Sample A were similar to those for boiled Sample B. As well as the dimensional change due to boiling, longitudinal elongation was sometimes observed after drying when PGF was below 45%.

Figure 6 shows the longitudinal Young's modulus for each moisture condition in relation to the proportion of G-fibers for Sample A and Sample B. Under all moisture conditions, the longitudinal Young's modulus increased with PGF regardless of whether the specimen was boiled or not. The longitudinal Young's moduli of all specimens increased as the specimens were further dried. No clear difference in both longitudinal Young's moduli for each moisture condition was observed between boiled and unboiled specimens.

004 *d*-Spacing

Table 1 illustrates the linear correlation coefficients between 004 *d*-spacing and the area percentage of G-fibers (PGF) for Sample A (unboiled) and Sample B (boiled). Because tension wood with G-layers contains more cellulose than normal wood, it was thought that distinctive boiling and drying effects would be observed on the cellulose crystal in wood cell walls. However, this study found no significant relationships between the 004 *d*-spacings under

Table 1. Linear correlation coefficients between 004 *d*-spacing and the area percentage of gelatinous fibers for Sample A (unboiled) and Sample B (boiled)

Sample	Green	Boiled	Air-dried	Fully dried
Sample A ($n = 32$)	0.10	–	0.09	0.03
Sample B ($n = 32$)	0.36	0.12	0.32	0.20

Table 2. Average 004 *d*-spacings under each condition for Sample A (unboiled) and Sample B (boiled)

Sample	Green	Boiled	Air-dried	Fully dried
Sample A ($n = 32$)	0.26	–	0.26	0.26
Sample B ($n = 32$)	0.26	0.26	0.26	0.26

all moisture conditions and PGF for both Sample A and Sample B. Moreover, Table 2 demonstrates that the average 004 *d*-spacings after boiling and/or drying treatment were the same.

Discussion

When discussing the behaviors of the G-layer from the experimental results using a macroscopic tension wood specimen, we need to make clear whether the detachment of the G-layer from the lignified secondary wall occurs during boiling and/or drying treatments in advance. We first guessed that boiling treatment may have detached the G-layer from the secondary wall, thus causing the reduction of the Young's modulus. However, the light microscope observation in this study could not detect distinct detachment of the G-layer. Moreover, based on the experiment using tension wood samples of *Populus* sp., Clair et al.[16] concluded that the oven-drying treatment causes no delamination between the G-layer and the lignified wall. With reference to those facts, it is natural to consider that the observed relationships between the longitudinal physical properties of tension wood and the area percentage of G-fiber are caused by the behaviors of the G-layer due to the boiling and drying treatments. Moreover, because the G-layer mainly consists of cellulose and has a microfibril angle of nearly zero, the estimated longitudinal behavior of the G-layer is considerably influenced by the structural or physical changes in CMF.

Influence of boiling

The longitudinal contraction due to boiling (Fig. 3) probably resulted from the tensile growth stress that remained in tension wood with G-fibers. This suggestion is supported by other studies[3,4] that reported the positive relation between tensile growth strain and PGF. These findings suggest that the CMF itself generates the high tensile stress that is distinctive in tension wood with G-fibers.

The result shown in Fig. 4 suggests the reduction of the longitudinal Young's modulus of the G-layer is due to boiling. We first guessed that boiling may have detached the G-layer from the secondary wall, thus causing the reduction. However, it is natural to consider that no distinct detachment of the G-layer occurred by the boiling treatment because the longitudinal drying shrinkage and the longitudinal Young's modulus for tension wood after boiling increased with increasing PGF. Thus, it should be considered that the reduction in the longitudinal Young's modulus was caused by a boiling-induced softening of the matrix region that fills the gaps between the CMFs. However, it is insufficient that only this factor could have decreased the longitudinal Young's modulus of the G-layer, which mainly consists of cellulose. It thus indicates that boiling treatment affects the noncrystalline regions in the CMF and then decreases the longitudinal Young's modulus of the CMF itself.

Influence of drying

Figure 5 illustrates the longitudinal shrinkage in the G-layer caused by drying. This result shows that more longitudinal shrinkage occurred in the G-layer than in other layers and suggests the existence of noncrystalline, hygro-sensible zones in the CMFs.[17] Moreover, because the boiled specimen shrunk longitudinally no less than the unboiled specimen, it suggests that the shrinking mechanisms due to boiling and drying are different.

The longitudinal Young's modulus increased as the drying proceeded regardless of PGF (Fig. 6). In the case of tension wood with less G-fibers, the longitudinal Young's modulus of the matrix substance in the cell wall generally increases. However, a similar increase was observed for tension wood with high amounts of G-fibers, suggesting that the longitudinal Young's modulus of the CMF also increases due to water desorption mainly from noncrystalline regions in the CMFs, as mentioned above. Moreover, almost the same results for the longitudinal Young's modulus were obtained for each moisture condition regardless of whether specimens were boiled. Softening of the matrix substance and the noncrystalline regions in the CMFs caused by boiling therefore did not affect the longitudinal Young's modulus when drying.

004 *d*-spacing

We first expected that longitudinal contraction of cellulose crystals in the CMFs would result from boiling. However, such contraction could not be practically observed in this study (Table 2). Similarly, the longitudinal change in 004 *d*-spacing after drying did not occur. Other studies have found reduction in 004 *d*-spacing during drying in normal coniferous wood.[15,18] These studies concluded that the shrinking matrix substance, which was tightly bonded with the CMFs, compressed the CMFs longitudinally during drying. However, a small amount of matrix substance in the G-layer probably cannot compress the CMFs during drying. Because all specimens in this study had a PGF of 20% or more, reduction of the 004 *d*-spacing due to drying matrix could not be observed.

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