

REVEALING FIBRIL ANGLE IN WOOD SECTIONS BY ULTRASONIC TREATMENT

Chih-Lin Huang

Scientist

Weyerhaeuser Company

Weyerhaeuser Technology Center, 1E19

Tacoma, WA 98477

(Received January 1994)

ABSTRACT

An effective method for inducing cell-wall checks in a microtomed wood radial section by ultrasonic treatment is described. Congo Red enhances cell-wall fibrillation in ultrasonic-treated sections. The amount of fibrillation and checking increases as ultrasonic treatment continues. However, too much fibrillation generated either by prolonged ultrasonic treatment or by Congo Red enhancement may visually obscure the checks. Less than three hours of ultrasonic treatment at 47 kHz frequency was found to be satisfactory to induce checks in 15- μ m-thick southern pine sections. The ultrasound check-inducing technique is most effective on southern pine fibers with large diameters and high fibril angles.

Keywords: Cell-wall checking, Congo Red, fibril angle, southern pine, ultrasound.

INTRODUCTION

The orientation of the microfibrils in the secondary cell wall is an important anatomical characteristic of wood. Fibril angle (orientation of S2 microfibrils with respect to the longitudinal axis of the cell) is closely related to the mechanical properties of wood and pulp fibers. Fibril angles tend to be greater in the juvenile wood zone and decrease radially outward with age. Low fibril angles are associated with long fiber lengths. Low fibril angles also are related to high wood strength, low longitudinal shrinkage, and high tangential shrinkage (Megraw 1985). Fibril angle of a pulp fiber has been shown to relate to mechanical properties such as elastic modulus and strength of the fiber (Mark and Gillis 1973).

There are direct and indirect methods for measuring fibril angle in wood sections and pulp fibers. Among the indirect methods are polarized light and X-ray diffraction techniques. Polarized light major extinction position (M.E.P.) techniques are low-cost but tedious procedures. X-ray diffraction analysis of a wood section is less tedious but averages together the fibril angle values of a large number of cells. On the other hand, fibril angle can be

measured directly from images of the S2 microfibrils in the microscope. The microfibrillar orientation can be revealed by iodine crystal deposition, fluorescence microscopy, replication, or inducing weathering checks (Crosby and Mark 1974). Significant discrepancies were observed among indirect methods (Boyd 1973; Meylan 1967), whereas a close agreement was found among direct methods (Meylan 1967). Direct methods give more accurate and consistent results and should be used when a precise fibril angle measurement is required (Crosby and Mark 1974).

Weathering procedures have been used to improve the image of microfibrils in fluorescence microscopy and iodine crystal deposition methods (Marts 1955; Senft and Bendtsen 1985). However, the checks induced by swelling and shrinking of wood are often inconsistent and sporadic (Miniutti 1964). A procedure to generate uniformly distributed checks on both earlywood and latewood fibers would improve the efficiency of direct fibril angle measurement methods.

Fibrillation and delamination of the fiber cell wall are the main structural changes observed in refined pulp fibers (McIntosh 1967; Page and De Grace 1967; Mohlin and Lid-

brandt 1980). Direct dyes have been used in the refining process to enhance refining effects. A 50% increase in refining efficiency was reported using Congo Red (Laine et al. 1980). In an aqueous solution of a direct dye, the dye molecules are linked by hydrogen bonds and become agglomerates or colloids. The elongated structure of these dyes make them able to link with the parallel cellulose molecules by multiple hydrogen bonds (Mark et al. 1986). Such an intrafiber colloidal association of dye molecules causes a reduction of the cohesive forces between layered planes of cellulose and consequently the delamination of the cell wall during refining processes (Laine et al. 1980).

An ultrasonic wave has an alternating rarefaction and compression half-cycle. During the compression half of the cycle, certain sizes of cavities (bubbles) in the liquid medium are suddenly collapsed, creating powerful shock waves, and generating a large amount of mechanical and thermal energy in the liquid. Such a phenomenon (known as cavitation) is the most striking property of ultrasound (Cracknell 1980). Ultrasonic beating has been used to study the structural changes of pulp fibers during refining processes. Dislocations in the S2 layer, removal of the S1 layer, swelling of the S2 layer, and then the fibrillation in the S2 layer have been observed on ultrasonic-treated wood fibers (Iwasaki et al. 1962). The main objective of this study was to evaluate the use of ultrasonic energy and Congo Red in revealing fibril angle in the cell wall in wood sections.

MATERIALS AND METHODS

Two kiln-dried 2 × 4 southern pine (*Pinus taeda* L.) lumber specimens, one with low fibril angle (Sample A) and the other with high fibril angle (Sample B), were selected. One-centimeter-long, one-growth-ring-wide blocks were cut from these two specimens. The blocks were aspirated in water, then 15- μ m-thick radial sections were microtomed from these blocks and the sections were kept in water.

Nine sections from each block were put into

a 47-mm polystyrene petri dish with either water or 1% Congo Red solution. The level of liquid was marked on the wall of the petri dishes, and the petri dishes were placed in an ultrasonic cleaner (Branson 3200, frequency 47 kHz). At the end of each hour of ultrasonic treatment, water was added to each petri dish to the marked level to maintain the concentration of the solution. After 0, 1, and 3 hours of ultrasonic treatment, three sections were taken out of each petri dish, rinsed with water, and mounted on a microscope slide for observation. Observations and photomicrography were performed using polarized light and ordinary transmitted light microscopy on a Zeiss Universal research microscope.

RESULTS AND DISCUSSION

Fibrillation induced by ultrasound

Sections were observed in the microscope after ultrasonic treatment, and observations were made on the degree to which they showed bundles of microfibrils (shreds of cell wall still attached to the sections). Perhaps due to the thick cell walls and lower fibril angles, latewood fibers appeared to be fibrillated more than earlywood fibers. Fibrillation was observed earlier and to a greater degree on sections from low fibril angle Sample A than Sample B. The microfibrils attached to the sections from Sample A were much longer than those attached to sections from Sample B. Because the S2 microfibrils of Sample A (low fibril angle wood) are almost parallel to the microtomed edges of the cell walls, perhaps a longer segment of the microfibril could be separated by a given ultrasonic beating than those of Sample B, whose fibril angle is almost 45°.

More and finer microfibrils were observed on sections ultrasonically treated in Congo Red solution than on those in water. The mechanism for fibrillation enhancement by Congo Red on pulp fibers has been described as a weakening of the bonding between cellulose strands by intrafiber aggregates of the colloidal dye molecules (Laine et al. 1980). The same mechanism might also be applied to explain



FIG. 1. Radial sections of low fibril angle Sample A ultrasonically treated for two hours and photographed using polarized light microscopy. (a) Earlywood. (b) Latewood. (450 \times). The arrows are pointed to the checks in S2 layer. Note that the checks in the latewood fibers are almost parallel to the longitudinal axis of the cell.



FIG. 2. Radial sections of high fibril angle Sample B ultrasonically treated for two hours and photographed using polarized light microscopy. (a) Earlywood. (b) Latewood. (450 \times). The arrows are pointed to the checks in S2 layer. The microfibrils appeared curved near the edges of the cell walls of both earlywood and latewood fibers.

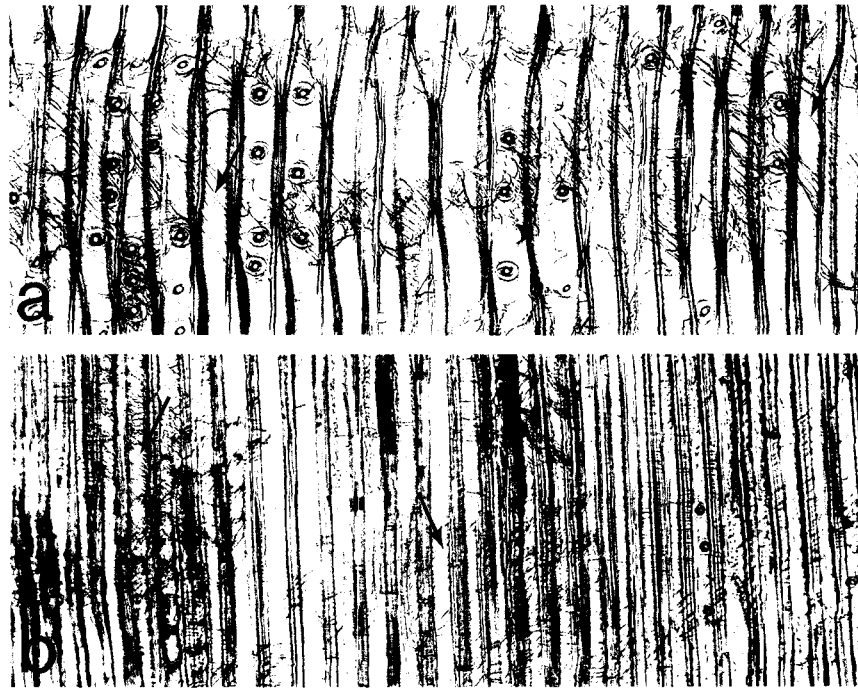


FIG. 3. Ultrasonically treated for two hours and air-dried sections from high fibril angle Sample B, checks are visible under the ordinary transmitted light illumination ($140\times$). (a) Earlywood, (b) Latewood. The arrows are pointed to the checks in S2 layer. The horizontal checks (possibly the orientation of S3 microfibrils) in some latewood fibers are also visible.

the fibrillation enhancement of Congo Red on ultrasonic-treated wood sections.

Checks induced by ultrasound

The fibril angle as determined by measuring the angle of the cell-wall checks was much lower in Sample A than in Sample B (Figs. 1 and 2). The fact that the rupture and removal of the S1 layer precede any other significant morphological changes has been found in previous studies on the effects of ultrasound on wood fibers (Iwasaki et al. 1962; Institute of Paper Chemistry 1972; Laine et al. 1976). Therefore, apparently only areas with exposed S2 layer on a wood section are subject to delamination by ultrasonic treatment. Most checks induced by ultrasonic treatment were found to be in the S2 layer or in areas of the S3 layer where the lumen of the fiber was exposed (Fig. 3). Patches of exposed S2 layer were found throughout the $15\text{-}\mu\text{m}$ -thick sections exam-

ined in this study. While microfibrils were observed to be torn apart on some exposed S2 areas, the fibril angle could be estimated using checks in an intact portion of the cell wall near those areas. When the sections were observed after mounting in water, the checks on the fiber walls were more distinct under polarized light illumination (Figs. 1 and 2). Once the sections were dried, the checks were also visible under ordinary transmitted light microscopy (Fig. 3).

The amount of checking observed increased as the ultrasonic treatment continued for up to three hours. At that point the large amount of fibrillation created by the ultrasonic treatment visually obscured the checking in low fibril angle Sample A. After three hours of ultrasonic treatment, the latewood zone in the section from Sample A was almost completely covered by microfibrils.

In latewood fibers with very low fibril angles, checks were easier to observe on ray cross fields



FIG. 4. Ultrasonically treated for two hours on section from low fibril angle Sample A showing checks and pits in ray cross field. The arrows are pointed to the vertical checks. Note that the angles of the checks in respect to the fiber axis are lower than those of the pit aperture ($450\times$).

where the fibrillation was insignificant. In a ray cross field, the angles of orientation of the checks were found to be slightly lower than the angles of the pit apertures (Fig. 4). The ultrasound check-inducing technique was observed to be most effective on southern pine fibers with larger diameters and high fibril angles. Checks in fibers with high fibril angles are helically curved, narrow, and closely spaced, while the checks in fibers with low fibril angles are straight, wider, and widely spaced.

The effects of Congo Red on enhancing checks in ultrasonically-treated sections were too small to be detected. Because Congo Red enhances fibrillation, which may visually obscure the checks, treatment with Congo Red may not improve the efficiency of revealing the S2 microfibril orientation in ultrasonically-treated wood sections.

In the author's experience, weathering checks are more difficult to induce on earlywood sections than on latewood sections. Also the iodine crystal deposition patterns on S2 microfibrils in some weathered specimens are

confounded with those of S1 microfibrils, which makes fibril angle measurement difficult. Using the ultrasonic treatment, checks were successfully induced on sections of these difficult specimens, and their fibril angles were measured with confidence. The only limitation of this technique is that too much fibrillation may be created on low fibril angle specimens that may visually obscure the checks.

CONCLUSIONS

The ultrasound check-inducing technique in wood sections is most effective on southern pine fibers with large diameters and large fibril angles. The amounts of checking and fibrillation increase as ultrasonic treatment continues. Adding Congo Red enhances the fibrillation on fibers and gives a colorful presentation of layered cell-wall structure under polarized light illumination. However, the amount of fibrillation generated by prolonged ultrasound treatment or by Congo Red addition may visually obscure the cell-wall checks in the wood sections. For $15\text{-}\mu\text{m}$ -thick southern pine sec-

tions, less than three hours of ultrasonic treatment at 47 kHz frequency produces cell-wall checks that reveal fibril angle.

ACKNOWLEDGMENTS

The author would like to thank Ronald V. Zarges and Norman P. Kutscha for reading the manuscript, Robert A. Megraw and Pedro G. Armenta for the stimulating discussions, and Joanne Doolittle for the darkroom work.

REFERENCES

- BOYD, J. D. 1973. Appraising methods of measuring microfibril angles. *Wood Science* 6(1):95-96.
- CRACKNELL, A. P. 1980. Ultrasound as a form of energy in industry and medicine. *In Ultrasonic*. Crane, Russak & Co., New York, NY.
- CROSBY, C. M., AND R. E. MARK. 1974. Precise S2 angle determination in pulp fibers. *Svensk Papperstidn.* 17: 636-642.
- INSTITUTE OF PAPER CHEMISTRY. 1972. Effect of ultrasonics on fibers and pulp, an annotated bibliography. Institute of Paper Chemistry S56. Appleton, WI.
- IWASAKI, T., B. LINDBERG, AND H. MEIER. 1962. The effect of ultrasonic treatment on individual wood fibers. *Svensk Papperstidn.* 65:795-816.
- LAINE, J. E., J. M. MACLEOD, H. I. BOLKER, AND D. A. I. GORING. 1976. Application of ultrasound in pulp and paper technology. *Pulp Paper Res. Inst. Can. PPR No.* 148.
- , T. SLOAN, AND G. G. ALLAN. 1980. Modification of the refining process by additives, particularly Congo Red. Pages 237-251 *in International symposium on fundamental concepts of refining*. Appleton, WI.
- MACINTOSH, D. C. 1967. The effect of refining on the structure of the cell wall. *Tappi* 50(10):482-488.
- MARK, H. F., N. M. BIKALES, C. G. OVERBERGER, AND G. MENGES. 1986. Dyeing. Page 233 *in Encyclopedia of polymer science and engineering*, 2nd ed., vol. 5. John Wiley & Sons, New York, NY.
- MARK, R. E., AND P. P. GILLIS. 1973. The relationship between fiber modulus and S2 angle. *Tappi* 56(4):164-167.
- MARTS, R. O. 1955. Fluorescence microscopy for measuring fibril angles in pine tracheids. *Stain Technol.* 30(5): 243-248.
- MEGRAW, R. A. 1985. Fibril angle. Pages 49-62 *in Wood quality factors in loblolly pine*. Tappi Press.
- MEYLAN, B. A. 1967. Measurement of microfibril angle by X-ray diffraction. *Forest Prod. J.* 17(5):51-58.
- MINIUTTI, V. P. 1964. Microscale changes in cell structure at softwood surfaces during weathering. *Forest Prod. J.* 14(12):571-576.
- MOHLIN, U-B., AND O. LIDBRANDT. 1980. Changes in fiber structure due to refining as revealed by SEM. Pages 61-74 *in International symposium on fundamental concepts of refining*. Appleton, WI.
- PAGE, D. H., AND J. H. DE GRACE. 1967. Delamination of fiber walls by beating and refining. *Tappi* 50(10):489-495.
- SENF, J. F., AND B. A. BENDTSEN. 1985. Measuring microfibrillar angles using light microscopy. *Wood Fiber Sci.* 17(4):564-567.