

ANALYSIS OF THE RELATIONSHIP BETWEEN MICROSTRUCTURE AND ELASTIC PROPERTIES OF THE CELL WALL¹

R. C. Tang

Department of Forestry, University of Kentucky, Lexington, Kentucky 40506

and

N. N. Hsu

Department of Engineering Mechanics, University of Kentucky, Lexington, Kentucky 40506

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ABSTRACT

A three-dimensional analysis of the relationship between the microstructure and the anisotropic elastic properties of the cell wall was made, using the theory of composite materials. In particular, the influence of the orientation of microfibrils in each layer, crossed helical structure, thickness of layers, and the spacing between the rectangular reinforced microfibrils to such properties were explored; spacing between microfibrils in each wall layer was found to be critical, and presence of crossed helices in the S_3 layer and the S_2 microfibril angles was found significant in relation to elastic properties. Numerical data of all elastic constants of the cell wall were evaluated for five hypothetical models that included the fibers of earlywood, latewood, and compression wood. Theoretical data of the axial Young's modulus of the wood fibers were compared with those values obtained from static tension tests and sonic tests by other investigators. The inadequacy of the technique used in the static tension tests of wood fibers was discussed, and a proper approach for such analysis was suggested.

Additional keywords: Cell-wall model, composite material theory, elastic constants.

INTRODUCTION

In recent years the problems of anisotropic shrinkage and uneven swelling in both bulk wood and papermaking fibers and the twisting of a wood fiber subjected to a tensile force have been intensively investigated by many researchers in studying wood mechanics (Cockrell 1946; Hosoi et al. 1958; Nakato 1958; Boutelje 1962; Ellwood and Wilcox 1962; Kelsey 1963; Barber and Meylan 1964; Harris and Meylan 1965; Sadoh and Christensen 1967; Sadoh and Kingston 1967; Ylinen and Jumppanen 1967; Barber 1968; Meylan 1968; Stamm and Smith 1969; Mark and Gillis 1970; Barrett et al. 1972). It was recognized that an adequate analysis cannot be made in

such problems unless a thorough knowledge of the mechanical properties of fibers is available.

Many investigators have performed tensile tests with delignified tracheids and fibers under air-dry conditions to evaluate their mechanical properties (Kollmann 1951; Wardrop 1951; Jayne 1959, 1960; Leopold and McIntosh 1961; Hartler et al. 1963; Britt and Yiannos 1964; Jentzen 1964; Kellogg and Wangaard 1964; Samuelsson 1964; Dinwoodie 1965; McIntosh 1965; Leopold 1966; Mark and Gillis 1970). Such tests supply information only on tensile strength and axial Young's modulus of delignified tracheids or fibers except the recent work, reported by Mark and Gillis (1970), which gives the degrees of rotation of a single fiber under tensile load. Their results show that vast differences exist not only between fibers of different species but also between fibers of earlywood and latewood of a single species. Of course, factors

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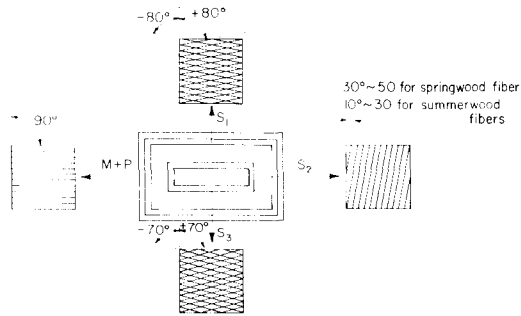


FIG. 1. Schematic diagram of the structure of a hypothetical fiber, showing microfibrillar directions.

such as isolation techniques, means of gripping fiber ends, span lengths, shapes of cross-section area and their measurement method, and environmental conditions may influence their results to a certain degree. However, it has long been recognized that the thickness of each layer in the cell wall of a wood fiber or tracheid and its microfibril angle have an important bearing on the mechanical properties of wood. For example, the microfibril angle is different not only between the earlywood and latewood tracheids but also between the tangential and radial walls (Mark 1967; Tang 1972); the secondary wall layers may be composed of a number of lamellae with varying microfibril angles (Wardrop 1964; Harada 1965; Dunning 1968). This indicates that a wide variation of mechanical properties in fibers or tracheids is expected.

From a theoretical approach, the mechanical properties of the cell wall of a hypothetical fiber have been evaluated two-dimensionally by Mark (1967); Cave (1968, 1969); Schniewind and Barrett (1969); Gillis (1970); Mark and Gillis (1970); and Schniewind (1970). It was shown that a three-dimensional analysis of hypothetical fibers supplies more information on the stress distribution in each layer of the cell wall (Tang 1972). In particular, such analysis gives the relative twisting angle of a single wood fiber that cannot be predicted by any two-dimensional analysis. In that study (Tang 1972), only two models of earlywood fibers with different

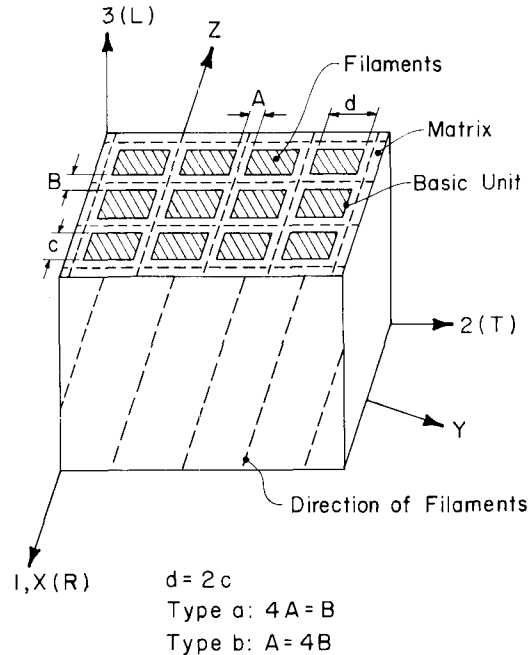


FIG. 2. An element from a layer of a fiber, showing the rectangular reinforcing filaments (microfibrils) with respect to its elastic coordinates (X, Y, Z) and geometric coordinates ($1, 2, 3$).

sets of helical angles were involved in the analysis. In both models, only the S_1 layer was assumed to be a crossed helical structure. Three different groups of elastic constants of crystalline cellulose were used in the defined calculation of layer elastic constants by an approach similar to Gillis' work (1970) on elastic moduli of a unidirectional composite with anisotropic rectangular reinforcement. Gillis' method can be applied only to orthotropic layered materials and will give only the approximate values of two-dimensional elastic constants with respect to their principal axes of elasticity. The elastic constants in the third direction were only approximated by using the law of mixture.

It follows from such data that all of the layer elastic constants with respect to the geometric axes of the fiber were calculated by using tensor transformations. Such calculations reveal that most layers will behave anisotropically with respect to the fiber geometric axes except for the layers

TABLE I. Characteristics of modified hypothetical fibers with crossed helices in both the S_1 and S_3 layers.

Fiber*	S_1		S_2		S_3			
	Area Fraction	Helical Angle	Area Fraction	Helical Angle	Area Fraction	Helical Angle		
SP-1	0.16	90°	0.3	$\pm 80^\circ$	0.2	$30^\circ - 50^\circ$	0.4	$\pm 70^\circ$
SP-2	0.15	90°	0.3	$\pm 80^\circ$	0.4	$30^\circ - 50^\circ$	0.15	$\pm 70^\circ$
OMP	0.07	90°	0.26	$\pm 80^\circ$	0.05	$30^\circ - 50^\circ$	0.0	--
SL-1	0.09	90°	0.13	$\pm 80^\circ$	0.7	$10^\circ - 30^\circ$	0.7	$\pm 70^\circ$
SL-2	0.07	90°	0.08	$\pm 80^\circ$	0.8	$10^\circ - 30^\circ$	0.05	$\pm 70^\circ$

*SP: Lignified fibers; OMP: Compression wood fibers; SL: Lateral fibers

with crossed helices or with a helical angle of 0° and 90° . This leads to complexity in the continuous determination of cell-wall elastic constants that are important and necessary to understand for the study of fiber mechanics. However, they cannot be determined by using Gillis' method (1970), and no literature on this subject has been found by the present authors. Therefore, in the previous analysis of a wood fiber under tensile forces, a layered anisotropic cylindrical model was considered because the cell-wall elastic constants are not available (Tang 1972). Also, in the previous investigation, we did not consider the size of the microfibril and the spacings between them, which are believed to be important in the analysis of the elastic behavior of the cell wall (Gillis 1970). So far as we are aware, such a suggestion has not been discussed. In addition, many investigators have shown the existence of crossed helices not only in the S_1 layer but also in the S_3 layer (Hodge and Wardrop 1950; Meiser 1955; Frei et al. 1957; Wardrop 1957; Harada 1965; Tang 1973). It is believed that some significant difference in elastic behavior of wood fibers will be revealed in the analysis of a cell-wall model with crossed helices in both the S_1 and S_3 layers. Seemingly, no such model has been investigated elsewhere.

More recently, Chou et al. (1972) have developed a more sophisticated method in deriving the overall elastic constants of three-dimensional layered anisotropic materials. Their method was adopted in this paper for computing the new elastic constants of each layer, as well as those of the

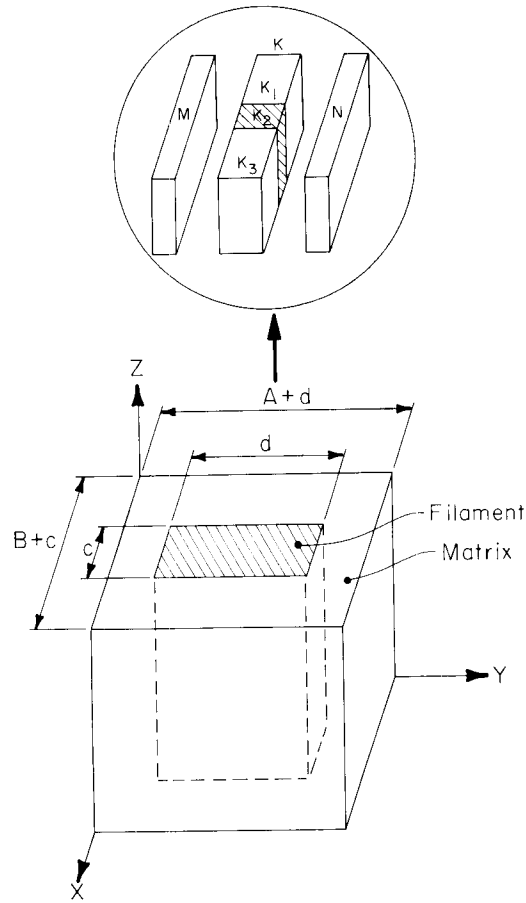


FIG. 3. A basic unit containing a single rectangular anisotropic filament with respect to its elastic coordinates. The X-direction is radial, the Z-direction corresponds to the filament length and the Y-direction completes an orthogonal set.

whole cell wall. Parameters being considered are the helical angle, the cross-sectional area of microfibrils, the spacing between these microfibrils, the ratio of filaments to matrix, the existence of crossed helices, and thickness of layers. Also, a comparison was made between the values of axial Young's modulus of the hypothetical fibers and of delignified fibers from mechanical tests. Furthermore, a suggestion on the improvement of accuracy in the determination of axial Young's modulus of a single fiber by static tension tests was discussed. Hopefully, the results can supply the fundamental information needed by re-

TABLE 2. Elastic constants* of filament and matrix in the cell wall with respect to their elastic coordinates (E and G in units 10^{11} dynes/cm²).

FILAMENT	Elastic Constants								
	E_1	E_2	E_3	E_4	E_5	E_6	E_7	E_8	E_9
Case 1	8.13	1.69	0.75	0.70	0.018	0.009	-0.001	0.341	0.010
Case 2	0.29	0.21	0.10	0.12	0.033	0.017	-0.0002	0.021	0.0016
Case 3	0.22	0.22	0.10	0.10	0.030	0.030	-0.0001	0.130	0.0008

MATRIX	Elastic Constants								
	E_1	E_2	E_3	E_4	E_5	E_6	E_7	E_8	E_9
	0.10	0.20	0.20	0.077	0.077	0.077	0.000	0.000	0.000

* E_1 is the axial Young's modulus, E_2 is the radial modulus, and E_3 is the tangential modulus.

search foresters and paper technologists in studying fiber mechanics to predict, with some degree of certainty, the performance of fabrics subjected to service conditions as well as providing a better insight into the relationship between microstructure and fiber deformation.

HYPOTHETICAL WOOD FIBERS

In this study, on the basis of the concept originally developed by Wardrop (1964); we modified the models that have been examined two-dimensionally by Schniewind (1970) and Mark and Gillis (1970), so as to analyze them three-dimensionally. Two modifications were made on the selected models: (1) In order to take into account the variation of the helical angle in S_2 , and the difference between tangential and radial walls, we chose three cases: 30°, 40°, and 50° for earlywood fibers; 30°, 40°, and 50° for compression wood fibers; and 10°, 20°, and 30° for latewood fibers. (2) In order to consider the existence of crossed helices in the S_3 layer of a hypothetical fiber, the helical angle in S_3 was changed from 70° to $\pm 70^\circ$. These modified hypothetical fibers are tabulated in Table 1, and we feel that they adequately represent the various fibers of all wood species. The schematic diagram of the structure of a typical hypothetical fiber showing microfibrillar angles is pictured in Fig. 1.

ANALYSIS AND RESULTS

It is assumed that the element shown in Fig. 3, which contains a single anisotropic rectangular filament surrounded by iso-

TABLE 3. Volumetric proportions of filaments and matrix.

LAYER	MATRIX	FILAMENTS
$S_1 + P$	89.9	10.1
$S_1, S_2,$ and S_3	46.9	53.1

tropic matrix material, is the basic unit that repeatedly produces the layer shown in Fig. 2. The data on the elastic constants of the filament and the matrix, as well as the proportions of these two materials in each layer given in our previous report (Tang 1972), were used in the calculation. For the convenience of the readers, those data are listed in Tables 2 and 3.

Two important modifications in the analysis have been made for this investigation. First, we assume that the rectangular filaments are uniformly distributed in the matrix and that their long edges are parallel to the cell wall, while their short edges are perpendicular to the cell wall. The ratio of these two edges is assumed to be two to one (Fig. 2). This assumption was based on the concept of the cross sections of cellulose microfibrils developed by Frey-

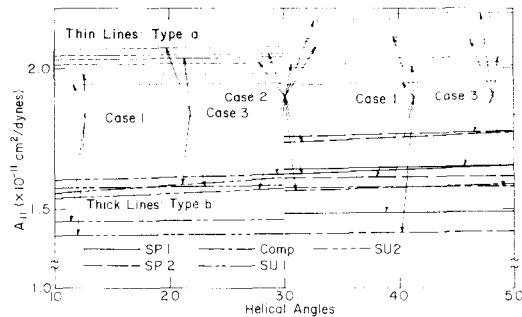


FIG. 4-16. Elastic compliances of cell wall versus helical angles in the S_2 layer for all the given cases of hypothetical fibers.

- Note: 1. The values given in Fig. 4 represent fibers either with or without crossed helices in the S_3 layer.
- 2. If a figure includes two parts, part a shows the values for fibers with crossed helices in the S_3 layer, while part b is for fibers lacking such structure.
- 3. The remaining figures are all for fibers with crossed helices in the S_3 layer.

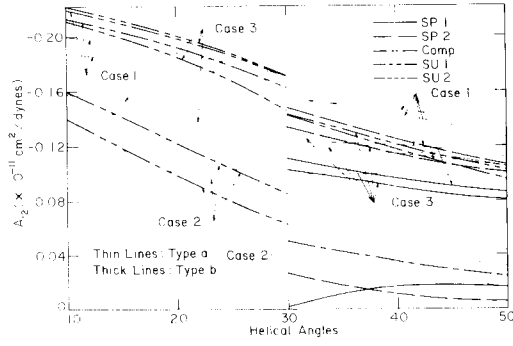


FIG. 5.

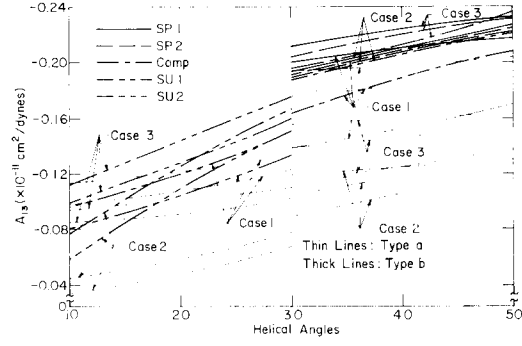


FIG. 6.

Wyssling in 1954 and Preston in 1965. Second, we assume that the spacings between adjacent filaments in the directions parallel to the cell wall, defined as A, and those in the orthogonal directions, defined as B, are not necessarily equal. Two types of arrangement have been assumed in the calculation: namely, Type a— $4A = B$; Type b— $A = 4B$ (see Fig. 2). No experimental data on the spacings between adjacent microfibrils are available at this time, but we believe that the range of these assumptions may cover most existing cases in the cell wall of a wood fiber. We are not aware of the above-mentioned assumption having been considered elsewhere in the study of the elastic behavior of wood fibers. By applying the method developed by Chou et al. (1972), the values of all the elastic compliances A_{ij} for the cell wall of hypothetical fibers in Table I were computed and their results were plotted in Figs 4-16, where

$$A_{ij} = \begin{bmatrix} A_{11} & A_{12} & A_{13} & A_{14} & 0 & 0 \\ A_{21} & A_{22} & A_{23} & A_{24} & 0 & 0 \\ A_{31} & A_{32} & A_{33} & A_{34} & 0 & 0 \\ A_{41} & A_{42} & A_{43} & A_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & A_{55} & A_{56} \\ 0 & 0 & 0 & 0 & A_{65} & A_{66} \end{bmatrix}$$

and $A_{ij} = A_{ji}$.

These elastic compliances may be grouped in terms of the equivalent "technical constants" as follows:

- I. $A_{11} = 1/E_R$, $A_{22} = 1/E_T$, $A_{33} = 1/E_L$;
- II. $A_{12} = -\mu_{TR}/E_R = -\mu_{RT}/E_T$, $A_{13} = -\mu_{LR}/E_R = -\mu_{RL}/E_L$, $A_{23} = -\mu_{LT}/E_T = -\mu_{TL}/E_L$;
- III. $A_{44} = 1/G_{TL} = 1/G_{LT}$, $A_{55} = 1/G_{RL} = 1/G_{LR}$, $A_{66} = 1/G_{RT} = 1/G_{TR}$;
- IV. $A_{56} = \nu_{RT, LR}/G_{RL} = \nu_{LR, RT}/G_{RT}$;
- V. $A_{14} = \eta_{TL, R}/E_R = \eta_{R, TL}/G_{TL}$, $A_{24} = \eta_{TL, T}/E_T = \eta_{T, TL}/G_{TL}$, $A_{34} = \eta_{TL, L}/E_L = \eta_{L, TL}/G_{TL}$;

where E is the directional Young's modulus, G is the shear modulus, μ is the Poisson's ratio, ν is the constant of Chentsov, and η is the constant of mutual influence, and the subscripts L, R, and T are the longitudinal, radial, and tangential directions in the cell wall, respectively. The data for the same

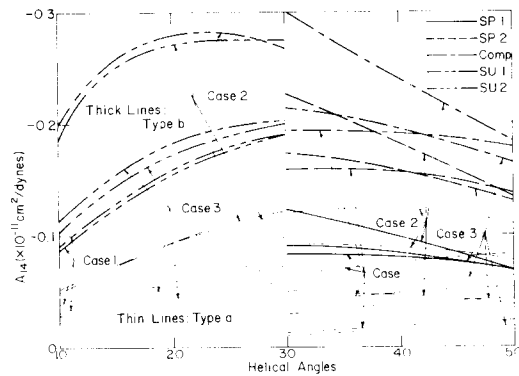


FIG. 7.

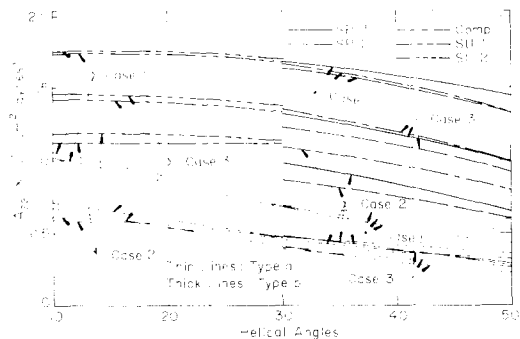


FIG. 8a.

model but with the assumption that no crossed helices occur in the S_3 layer were also calculated. For simplicity only the principal elastic compliances were plotted in the related figures for comparison. All of these elastic compliances were calculated by the following process: (1) We consider each individual layer, with reference to its elastic axes, to be composed of numerous repeated basic units and consisting of three layered elements, K, M, and N, where the K element also consists of three sublayered elements, K_1 , K_2 and K_3 (see Fig. 3). The fractional volume of these elements in each layer was determined by using the data for volumetric proportions of filaments and matrix as given in Table 3; the spacings between these filaments were those mentioned in the previous section. By using the elastic constants of filament and matrix given in Table 2, the elastic compliances of the K elements were first calculated. The elastic compliances of the basic units for

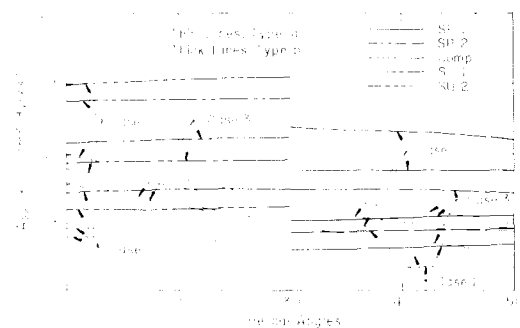


FIG. 8b.

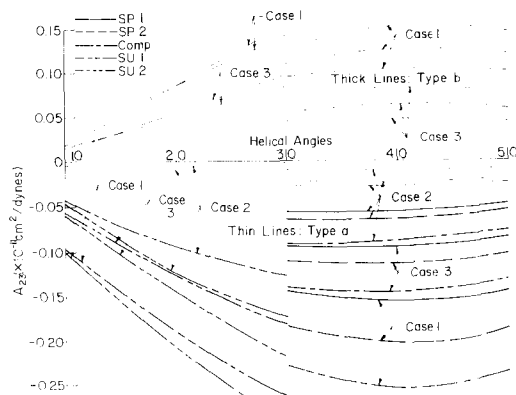


FIG. 9.

each layer were then determined. In other words, the layer elastic compliances with respect to their elastic coordinates were obtained. Then, by using tensor transformations with the data for the helical angle of each layer as listed in Table 1, the layer elastic compliances with respect to the cell-wall axes were determined. (2) We consider that the cell wall of a wood fiber consists of four layers, namely, primary wall and middle lamella (M + P); S_1 layer, S_2 layer, and S_3 layer. By using the data for the area fraction of each layer given in Table 1 and the results from (1) above, the cell-wall elastic compliances with respect to their geometric axes were determined.

To summarize the numerous variables involved in these determinations, we have one

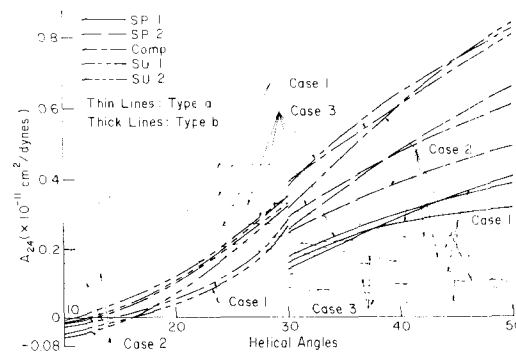


FIG. 10

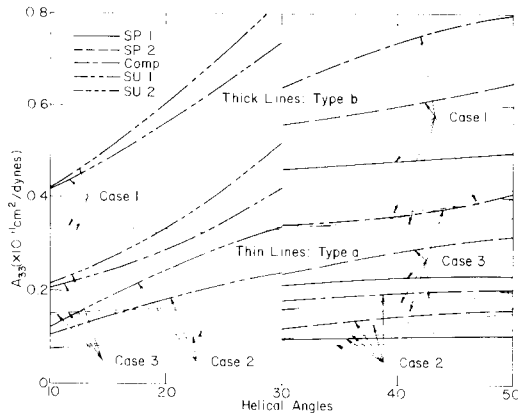


FIG. 11a.

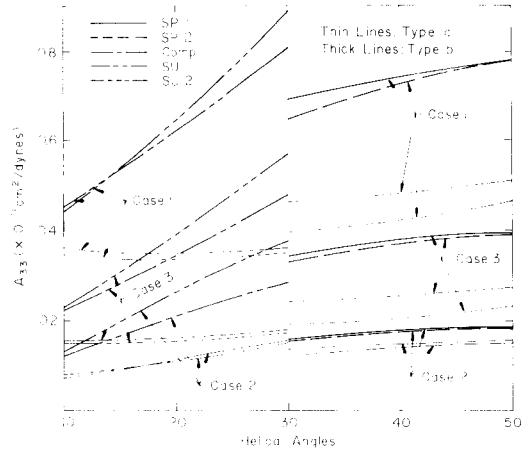


FIG. 11b.

set of elastic constants for matrix, three cases of elastic constants for the microfibrils, a fixed ratio of the volume of filaments (microfibrils) to matrix in the $M + P$, S_1 , S_2 , and S_3 layers, two types of spacing between microfibrils, and five models of hypothetical fibers with various orientations of helical angles in the S_2 layer and fractional volume of layers. It is too involved to list all the equations used in this calculation. However, the general equation was given by Chou et al. (1972). We would like to note here that one should use proper coordinates in the determination of the elastic constants in each process.

THEORETICAL VERSUS EXPERIMENTAL RESULTS

In the tension tests, the wood fiber was considered by all the investigators mentioned earlier as an isotropic hollow cylinder with both ends fixed and subjected to a tensile force (P) (Fig. 20). They assumed that the axial Young's modulus (E_L) of a wood fiber with an outer radius r_o and inner radius r_i when it is subjected to a tensile force P , can be determined by the formula $E_L = P/[\pi(r_o^2 - r_i^2)\epsilon_L]$, where ϵ_L is the strain over the linear portion of the load-deflection curve recorded from the tension tests.

It is well known that wood fibers behave anisotropically, and the relationships between stress and strain are very complicated. Therefore, the axial Young's modulus

of wood fibers determined from this formula is only an approximation. This is a good approximation provided that the wood fiber is not twisted before or after the tension test. However, a single tracheid or fiber will twist immediately after being picked up from the water. A scanning electron photomicrograph of such a twisted single Virginia pine tracheid is shown in Fig. 19. In other words, the wood fibers will be in the form of a coiled tubular spring with a very large helical pitch angle rather than in the form of a straight cylinder even before the tension test (Fig. 20).

If the initial twisting stresses in the fiber can be ignored, then the method for the determination of axial Young's modulus in a

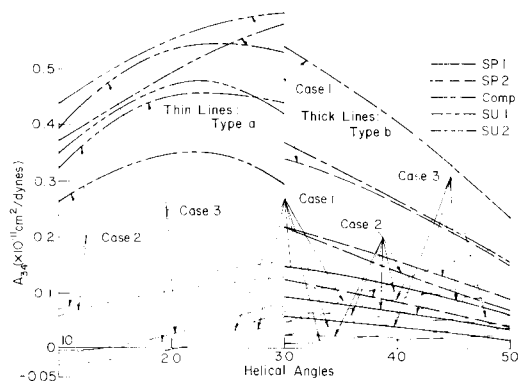


FIG. 12.

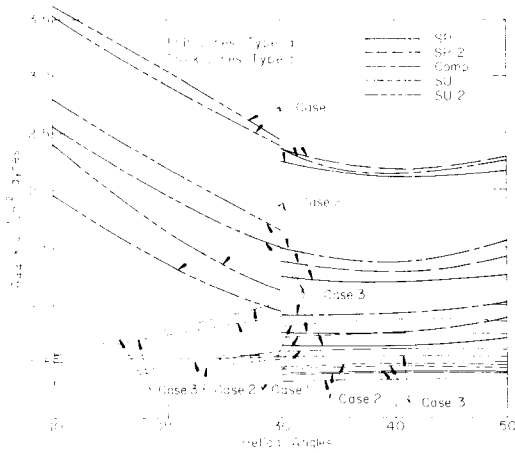


FIG. 13a.

coil spring of anisotropic materials can be modified and used in the analysis of tension tests of a fiber. Such a method was reported by Mark et al. in 1969. A detailed discussion and analysis on the mechanical properties of wood fibers using such an approach will be presented in a forthcoming report.

It is difficult to compute the theoretical values of axial Young's moduli of a wood fiber because the radial and tangential walls of a fiber behave not only anisotropically themselves but differently as well. However, for a first approximation, if the anisotropy of the cell wall can be neglected and the wood fibers are assumed to be of

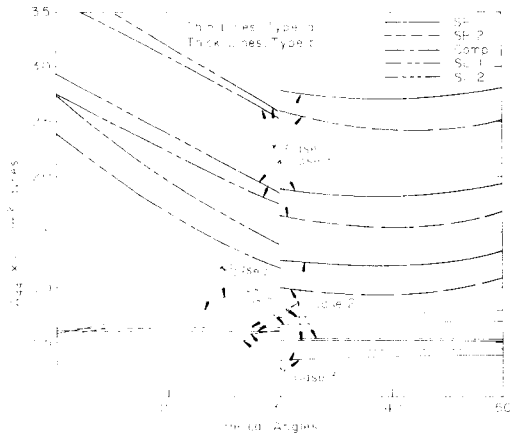


FIG. 13b.

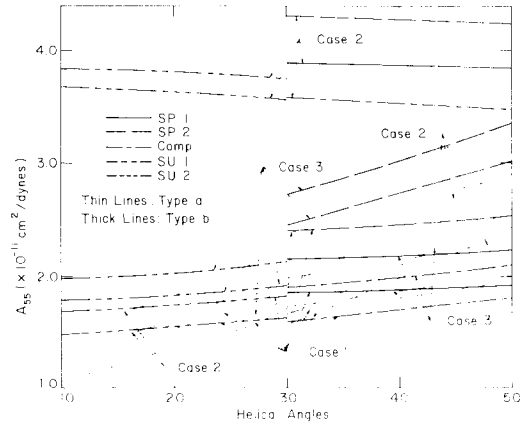


FIG. 14a.

square cross section with equal volume of radial and tangential walls, then the axial Young's modulus of a hypothetical fiber can be determined by applying the method developed by Paul in 1960 on the prediction of elastic constants of multiphase materials, provided there is only one value of axial Young's modulus for the entire cell wall. We assumed that such a value is equal to the average of the axial Young's moduli of the radial and the tangential walls, and the results of all the given cases of hypothetical fibers were calculated and plotted in Figs. 17 and 18. For comparison, the values obtained from the static tension tests of delignified fibers (Jayne 1960; Jentzen 1964; Samuelsson 1964; and Leopold 1966) as well as from the sonic tests

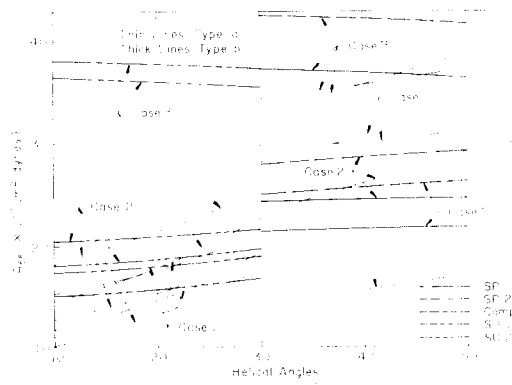


FIG. 14b.

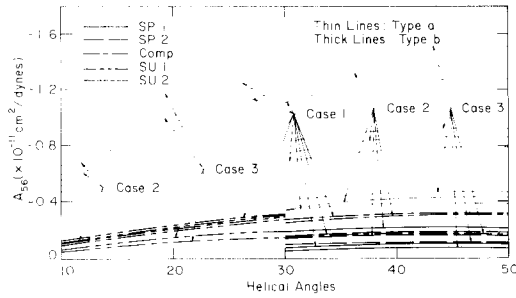


FIG. 15.

of thin wood sections (Yiamos and Taylor 1967) are also given in the related figures.

DISCUSSION AND CONCLUSIONS

During the past two decades, many foresters have researched the strengthening of paper-making wood fibers by altering control of the microfibrillar angle of the cell wall through tree breeding. Although this procedure may result in a wood fiber of greater tensile strength, its effect on the many other mechanical properties is unknown. From the results of this investigation, however, it is evident that the mechanical properties of wood fibers are correlated not only with the microfibrillar angle in the S_2 layer and the thickness of individual layers, as has been reported by other investigators before, but also with the crossed helices in the S_3 layer and the spacings between the microfibrils.

The results shown in all the figures indi-

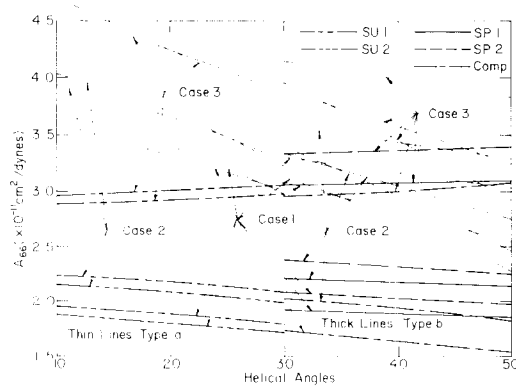


FIG. 16a.

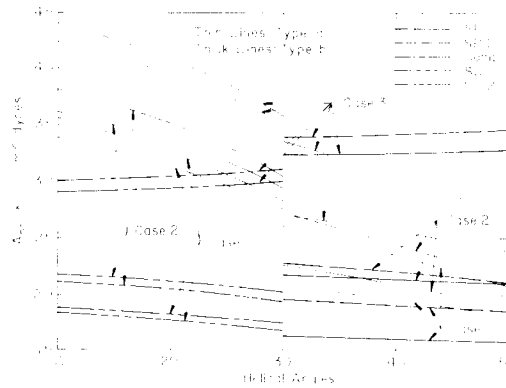


FIG. 16b.

cate that the influence of spacings between the microfibrils in each layer is much greater than the microfibrillar angle in the S_2 layer to the variation of cell-wall elastic compliances. It has been found that the values of directional Young's moduli E_{L_1} and E_T and shear moduli G_{TL} and G_{RL} of

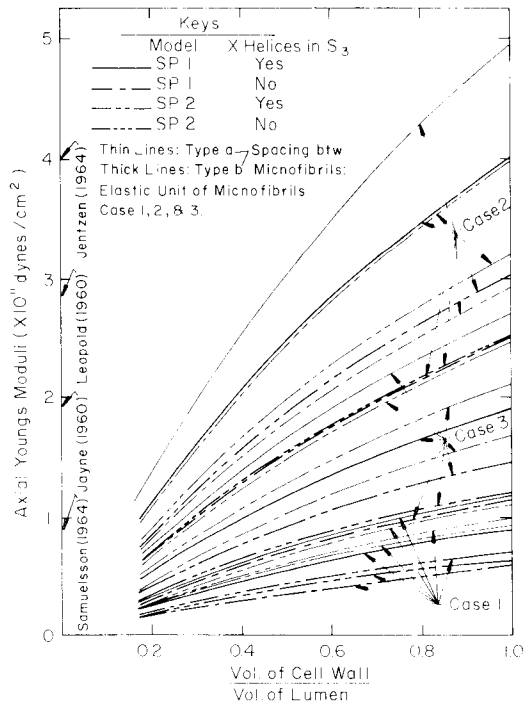


FIG. 17. Axial Young's moduli of earlywood fibers versus the ratio of volume of cell wall to cell lumen.

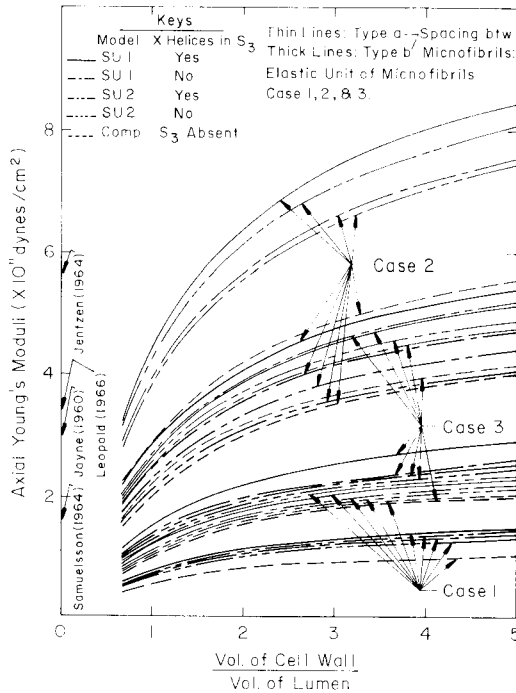


FIG. 18. Axial Young's moduli of latewood and compression wood fibers versus the ratio of volume of cell wall to cell lumen.

cell walls in the fibers with Type a spacings between the microfibrils are greater than those with Type b arrangement, regardless of the particular elastic constants for the microfibrils used in the calculation. However, the fibers having crossed helices in the S_3 layer, disregarding the arrangement of microfibrils, have a relatively higher value of cell-wall elastic constants E_L and G_{LT} than those without crossed helices, while there is no distinguishable difference in the values of E_R . The variation of the remaining elastic compliances is related to the existence of crossed helices in the S_3 layer, but no definite trend can be predicted.

The value of E_L of the cell wall decreases while E_T increases with increasing microfibrillar angles in the S_2 layer, but the value of E_R remains almost unchanged in the fibers when their microfibrils possess the same elastic constants (see Figs. 4, 8a, 8b, 11a, 11b). The value of G_{LR} decreases

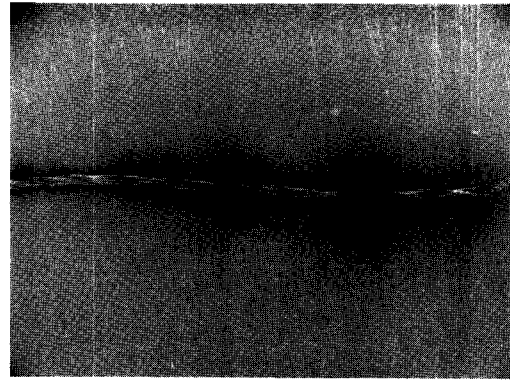


FIG. 19. A scanning electron photomicrograph of self-twisted Virginia pine tracheid.

with increasing microfibrillar angles in the S_2 layer except in those having Type b spacings between the microfibrils and with case 3 elastic constants (see Figs. 14a and 14b). Such a situation is reversed for the value of G_{RT} and no trend can be followed in the variation of G_{TL} . All of these variations indicate that there will be a difference between the elastic properties of radial and tangential walls if they have different microfibrillar angles in the S_2 layer. The influence of such facts on the remaining elastic constants is somewhat complicated. We do not discuss them here because they are less important in the evaluation of mechanical properties of materials.

From Figs. 17 and 18, it can be seen that the values of axial Young's modulus for delignified fibers obtained from static tension tests by other investigators fall in the range of theoretical values of hypothetical fibers evaluated from this analysis. It should be noted here that the data are only approximations. Since the delignified fibers were self-twisted before the tension tests (see Fig. 19), then the value of axial Young's modulus obtained from such tests will be the spring constant of a twisted fiber rather than the elastic constant of a straight fiber. Generally speaking, the spring constant of a coil spring will be relatively lower than the elastic constant of the material in its original axial direction. In addition, the effects of delignification on the fibers have not been taken into account

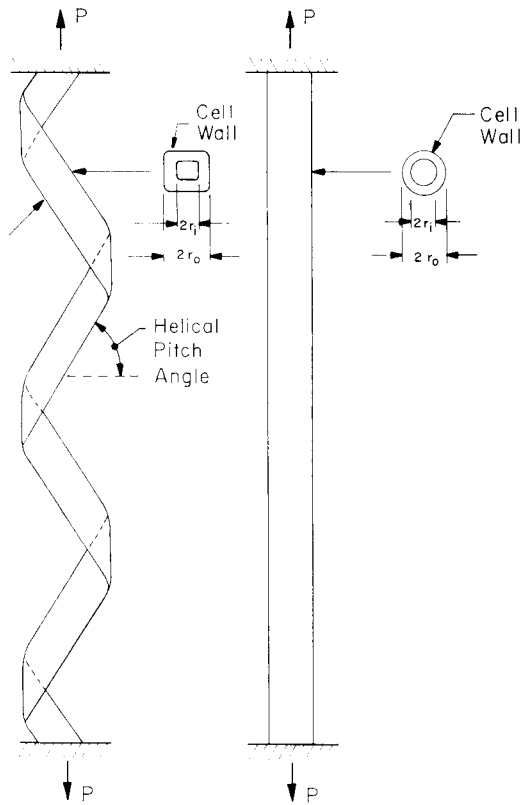


FIG. 20. A comparison of the tension test of a pretwisted fiber and a straight fiber.

in their determinations. For example, these experimental values for delignified fibers were relatively lower than the average value (6.922×10^{11} dynes/cm² of nine species) determined by Yiannos and Taylor (1967) from sonic tests of thin microtome wood sections, which obviously included both earlywood and latewood fibers. This is understandable because the axial Young's modulus determined by sonic velocity is essentially an intrinsic property of the material. In the determination of the theoretical axial Young's modulus of a hypothetical fiber, the anisotropy of the cell wall was neglected, the cross section of the fiber was assumed to be square, and the volumes of radial and tangential walls in the fiber were assumed to be equal. It is believed that one could obtain more reliable data of the elastic properties of wood

fibers either from static tension tests or from a theoretical analysis, if the above-mentioned facts are taken into account in their respective determinations.

By cross-examining the results of all the hypothetical fibers, it was found that the crossed helices in the S_3 layer are equally as important as the microfibrillar angles in the S_2 layer in affecting the strength properties of wood fibers. However, if the elastic properties and the dimensions of microfibrils in the cell wall of all wood species are virtually similar, and the volumetric properties of filaments (microfibrils) and matrix in each layer are of the ratios we assumed, then the mechanical properties of wood fibers are influenced chiefly by the spacings between the microfibrils in each layer, whereas the influences of the crossed helices in the S_3 layer and the microfibrillar angles in the S_2 layer are still significant but not critical. Hopefully, the results presented in this investigation can supply some fundamental information to foresters and paper technologists on the improvement of wood fiber quality through tree breeding.

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