

# A MODEL FOR THE PREDICTION OF FIBER ELASTICITY

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## ABSTRACT

A model is presented that enables the elastic properties of wood fibers to be estimated from the properties of its polymeric constituents, cellulose, hemicellulose, and lignin. The influence of the value of the axial stiffness of the cellulose crystal is demonstrated, its proper value being discussed in comparison with experimental data on fibers. The effects on fiber stiffness of the S2 fibril angle, the fibril angles of other layers, the crystallinity, and layer thicknesses are analyzed. The manner in which the effect of a variation in yield can be simulated by a change in shape factor of the reinforcing cellulose crystals is demonstrated, the cell wall thus being considered to be a discontinuous reinforced composite.

*Keywords:* Cell walls, cellulose, cellulose fibers, elastic strength, hemicelluloses, lignins, models.

## INTRODUCTION

In the search for an understanding of the elasticity of wood fibers, it is essential to derive theoretical tools to link the structure and mechanical properties of the components into a comprehensive composite model.

The elasticity of wood fibers may be predicted by recognizing the structural features of the cell wall in applying micromechanical theories coupled with classical lamination theory (Cave 1968; Mark 1972; Schniewind 1972). Hitherto the complexity of the structural model has been limited to *continuous* reinforcing elements. However, irregular zones are known to exist along the length of the microfibril (Krässig 1979), and the consequence of these for the mechanical properties of paper has also been discussed (Fellers et al. 1980; Page 1983). Especially under wet conditions when these zones can be considered soft, they have a special importance for the fiber properties.

This paper demonstrates how it is possible to introduce the concept of *discontinuous* reinforcing elements into the mathematical framework and thus take into account the effects of irregular zones in the microfibrils. Further it shows how different values suggested for the elastic constants of cellulose influence the theoretical prediction of the fiber moduli and how these compare with experimental values. Finally, the manner in which the length-to-diameter ratio of the reinforcing elements of cellulose crystallites may influence the fiber modulus is discussed and how changes in the slenderness of these reinforcing elements may be achieved by moisture uptake or cellulose hydrolysis. The influence of moisture content on the fiber moduli will be dealt with in a subsequent publication.

## CELL-WALL MODEL

The properties of wood fibers have been described in terms of various models of the structural organization of their components (Cave 1968; Cowdrey and Preston 1965; Hearle 1963; Mark 1967, 1972; Muench 1938; Schniewind 1966, 1972).

Muench (1938) emphasized the helical winding of microfibrils in the cell wall,

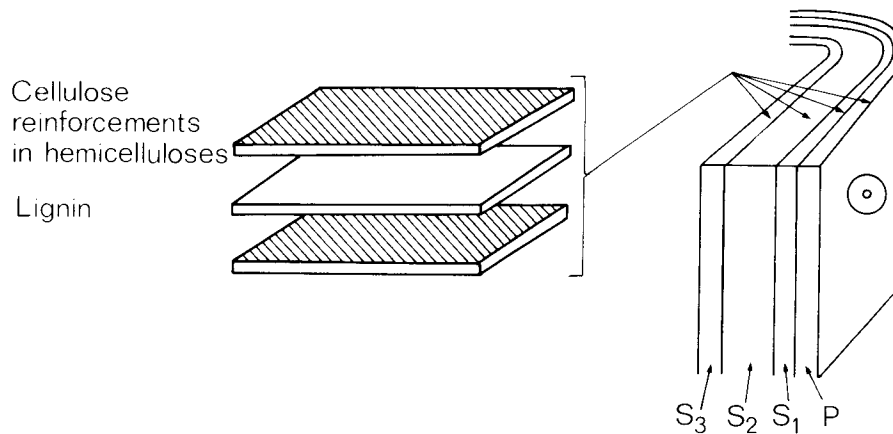


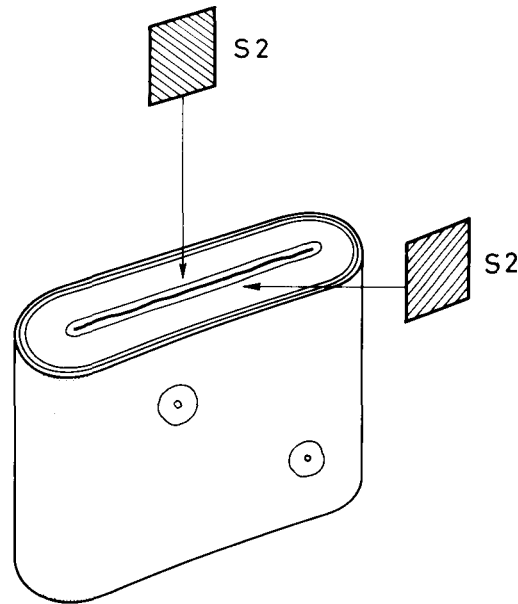
FIG. 1. Model representation of the cell-wall layers in a tracheid.

considered the cell wall to be composed of several sets of helical springs embedded in a matrix of noncrystalline material. Mark (1967) and Schniewind (1966) have taken into consideration the different layers in the tracheid cell wall and have developed a cell-wall model based on the concept of a laminated structure. The model used in this paper is based on this latter approach, assuming each cell-wall layer to consist of cellulose microfibrils embedded in a matrix of hemicelluloses. The lignin present is considered to exist in separate isotropic lamellae located in the middle of each cell-wall layer, as is schematically shown in Fig. 1.

This model involves a certain degree of simplification. Kerr and Goring (1975) have, for instance, concluded on the basis of electron microscope studies that the cell wall has an interrupted lamella structure where the dimension of a given lignin or carbohydrate entity is greater in the tangential direction of the fiber wall than in the radial direction. They also concluded that the hemicelluloses not only exist as a matrix around the microfibrils but must also be present in the lignin-containing entities. The presence of covalent bonds between lignin and hemicelluloses (Eriksson et al. 1980) indicates a close association between these components. On the other hand, at least a portion of the hemicellulose appears to be associated with the cellulose (Keegstra et al. 1973; Kerr and Goring 1975). There are also indications that the hemicelluloses show some degree of preferred orientation associated with the cellulose alignment (Fushitani 1973; Liang et al. 1960) so that the hemicellulose may not behave as an isotropic material. Yet there is still no evidence that the wood polymers are miscible in the sense that they mechanically react as a homogeneous component, i.e., exhibiting a single glass transition temperature. It is therefore here considered that the cellulose, hemicellulose, and lignin are components of a heterogeneous composite.

The laminate model structure of wood fibers is shown in Fig. 2. Well-beaten fibers or fibers of low yield can be considered to be collapsed, i.e., the square fiber has been flattened out so that its inner surfaces contact each other. In this case the angle of the fibrils in a layer in the front fiber wall is opposite to that in the back fiber wall. Thus the fibers can be viewed as *anti-symmetric* laminates.

Unless otherwise stated, the S2 layer of the fiber is here considered to have a



**fiber wall element**

FIG. 2. Fiber wall element of tracheid. The element consists of the layers P, S1, S2, S3, S3, S2, S1, P.

fibril angle of 20°, while all the other layers have a fibril angle of 70°. The thicknesses of the respective layers are taken as 16% for P plus S1, 76% for S2 and 8% for S3.

The material constants of the components of the cell wall needed to calculate the fiber properties are still not established in detail. Both experimental results and theoretical considerations have, however, given values that are sufficiently reliable for the scope of this model. The data used are summarized in Table 1. For hemicellulose, transverse isotropy is assumed. The ratios between the moduli are in accordance with the suggestions of Cave (1978).

#### RELATIONS FOR COMPOSITE MODELS

The fundamental steps in the cell-wall model calculation are first an analysis of the micromechanics of the composite material plies and then a combination of these through lamination theory to estimate the properties of the total structure of the fiber.

For the micromechanical considerations, the Halpin-Tsai equations (Halpin and Kardos 1976), which account for the shape of the reinforcing material, have been chosen. This equation is given as:

$$E/E_m = \frac{1 + \zeta\eta V_f}{1 - \eta V_f} \quad (1)$$

TABLE 1. Data for mechanical properties of cell-wall components.

cellulose	
$E_x$ (N/m <sup>2</sup> )	$13.4 \cdot 10^{10}$ (Sakurada et al. 1962)
$E_y$ (N/m <sup>2</sup> )	$2.72 \cdot 10^{10}$ (Mark 1967)
$G$ (N/m <sup>2</sup> )	$0.44 \cdot 10^{10}$ (Mark 1967)
$\nu_x$	0.1 (Mark 1967)
$\ell/d$	50,000 (estimated)
hemicellulose	
$E_x$ (N/m <sup>2</sup> )	$8 \cdot 10^9$ (Cousins 1978)
$E_y$ (N/m <sup>2</sup> )	$4 \cdot 10^9$ (estimated)
$G$ (N/m <sup>2</sup> )	$2 \cdot 10^9$ (estimated)
$\nu_x$	0.2 (estimated)
lignin (isotropic)	
$E$ (N/m <sup>2</sup> )	$4 \cdot 10^9$ (Cousins 1976)
$G$ (N/m <sup>2</sup> )	$1.5 \cdot 10^9$ (estimated)
$\nu$	0.33

$E$  = Young's modulus.  $G$  = shear modulus.  $\nu$  = Poisson's ratio.  $\ell/d$  = reinforcement shape factor; crystallite length divided by diameter.

where

$$\eta = (E_f/E_m - 1)/(E_f/E_m + \zeta) \quad (2)$$

and  $E$  is the modulus of the composite,  $E_f$  that of the reinforcement and  $E_m$  that of the matrix,  $V_f$  is the volume fraction of the reinforcement and  $\zeta$  is a shape factor for the reinforcing elements. For the modulus along the axis of the reinforcements,  $\zeta = 2(\ell/d)$ , where  $\ell/d$  is the length-to-diameter ratio, i.e., the aspect ratio of the reinforcing elements. For a continuous reinforcement in the length direction, this factor  $\ell/d$  thus approaches infinity.

With this mathematical tool, it is possible to characterize various effects of the disordered regions in the cellulose microfibrils. Thus, if the cellulose crystallites alone are considered as reinforcements, the effect of cellulose hydrolysis or a softening by water of the disordered cellulose may be simulated. When the influence of other fiber characteristics is emphasized, it may still be adequate to consider the entire microfibril as the reinforcing element. Normally, under dry or humid conditions, a value of 50,000 for  $\ell/d$  has been chosen to simulate the "infinitely" long reinforcements while still recognizing their true limits.

The calculation of the properties of the fiber follows classical lamination theory as outlined in the Appendix.

The classical lamination theory assumes that the laminate is thin and that the normal to the middle plane remains straight and of constant length during deformation. Wood fibers have, of course, a finite width and a rather substantial thickness and are thus far from ideal. However the tenfold ratio of width to thickness makes the assumptions not too crude.

When fibers and wood samples are tested in the longitudinal direction, the strain  $\epsilon_1$  is given by:

$$\epsilon_1 = A_{11}^* N_1 + A_{12}^* N_2 + B_{16}^* K_6 \quad (3)$$

where  $A_{11}^*$ ,  $A_{12}^*$ ,  $B_{16}^*$ ,  $N_1$ ,  $N_2$  and  $K_6$  are, respectively, stiffness components, normal forces, and the twisting.

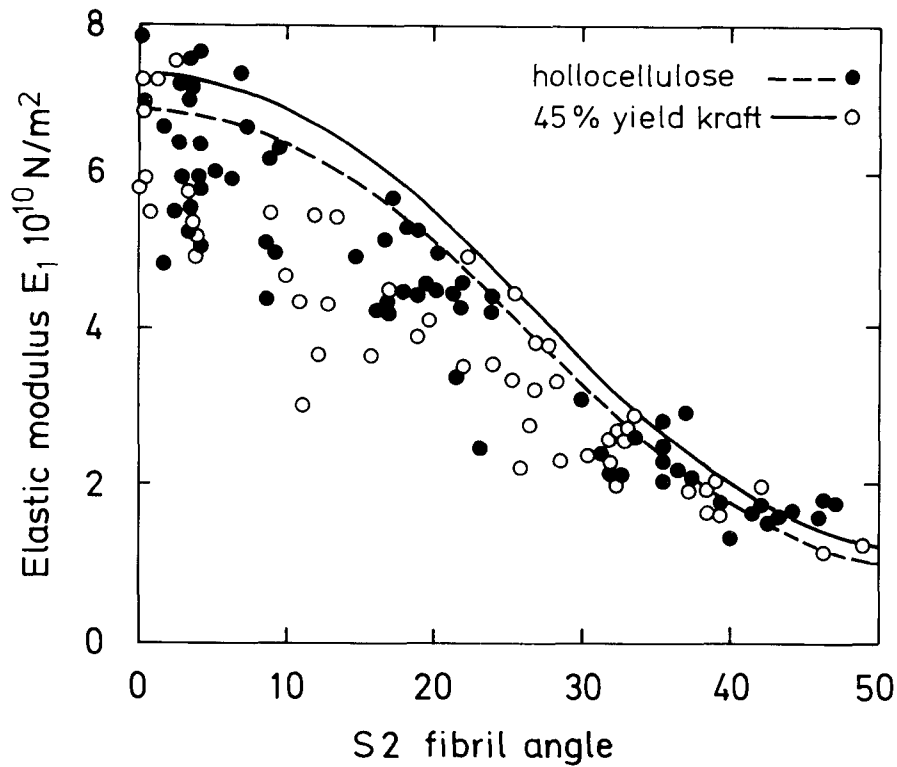


FIG. 3. The elastic modulus of pulp fibers as a function of the S2 mean fibril angle. The points refer to single fiber measurements by Page et al. (1977). The curves are calculated assuming the holocellulose fibers to be lignin-free and to contain 35% hemicellulose and the kraft fibers to contain 8% lignin and 23% hemicellulose.

If the mounting is considered to be firm, the twisting  $K_6$  is equal to zero. The transverse force  $N_2$  is also zero giving the engineering modulus:

$$E_1 = N_1 / \epsilon_1 h = 1 / A_1^* h \quad (4)$$

where  $h$  is the thickness of the laminate.

#### COMPARISON WITH EXPERIMENTS

##### *Fibril angle dependency*

Since the S2-layer of the fiber wall represents such a large proportion of the total fiber wall, the fibril angle of this layer will have a pronounced influence on the properties of the fiber. This fact was clearly demonstrated by Page et al. (1977) in their extensive measurements of the elastic modulus of single fibers of a black spruce. Their data for fibers prepared from a holocellulose pulp and from a kraft pulp of 45% yield are shown in Fig. 3, together with the results of calculations based on the present cell-wall composite model, assuming that the holocellulose fibers were lignin-free with a hemicellulose content of 35% and that the kraft fibers contained 8% lignin and 23% hemicellulose. It should be noted, as Page has pointed out, that many of the fibers measured were damaged during the process of isolation. These fibers contain crimps, microcompressions, dislocations etc.

TABLE 2. Various values reported for the elastic constants of crystalline cellulose.

	$E_x(10^{10}\text{N/m}^2)$	$E_y(10^{10}\text{N/m}^2)$	$G_{xy}(10^{10}\text{N/m}^2)$
Treloar (1960)	56.5		
Mark (1967)	111.3	27.2	4.4
Jaswon et al. (1968)	56.6	75.9	0.8
Jaswon et al. (1968)	56.5	33.6	0.4
Gillis (1969)	246	16.4	0.17
Gillis (1969)	319	37.3	0.39
Experimental value of Sakurada et al. (1962)	134		

and show a lower modulus within the span at a given fibrillar angle. Thus it may be expected that the highest value obtained for the elastic modulus of the fibers should represent the undamaged fibers with which the calculated values are to be compared. The fact that the fibers from the kraft cook generally seem to have a lower stiffness than the holocellulose fibers, contrary to the calculations, may be due to the fact that the former fibers have been more severely treated during the isolation process.

The calculations assume that the lumen is collapsed and that the opposing fiber walls are bonded together, whereas Page et al. (1977) assume that the cell-wall corners transmit the shear stresses between the walls and thus restrict shear deformation. Their boundary conditions are, however, essentially equivalent to a restriction across the lumen. The calculated curves in Fig. 3 are in good agreement with a curve calculated by Page et al. (1977), but in the present case, the curve is predicted from moduli of the individual components and does not merely represent a fit to the experimental data.

There is still a lot of uncertainty as to the proper values of the elastic moduli of the cellulose microfibrils. The axial stiffness of the cellulose in ramie fiber has been measured by Sakurada et al. (1962) as  $13.4 \times 10^{10} \text{ N/m}^2$ , but there are no measured data for the other stiffnesses. Several attempts have been made to compute cellulose stiffness from structural models (Gillis 1969; Jaswon et al. 1968; Mark 1967; Treloar 1960) but none of these have yet proved entirely satisfactory. These estimates for the stiffnesses of cellulose are given in Table 2. The calculations of Treloar (1960) and Jaswon et al. (1968) gave values far below experimental data, while Mark's estimates come close to the value for the axial stiffness measured. Gillis (1969) reached higher values of the cellulose stiffness by also considering deformations of interchain hydrogen bonds. However, as pointed out by Gillis (1969) himself, his values represent only an upper limit for the axial stiffness of the cellulose since several degrees of freedom are neglected in the calculations.

In Fig. 4, results obtained by incorporating different moduli for the cellulose in the present model are compared with actual fiber measurements, the value of  $E_x = 24.6 \times 10^{10} \text{ N/m}^2$  estimated by Gillis (1969) and the value of  $E_x = 13.4 \times 10^{10} \text{ N/m}^2$  measured by Sakurada et al. (1962) being compared with the measurements of Page et al. on holocellulose fibers of black spruce. The measured value of the modulus of cellulose crystallites clearly gives a far better estimate for

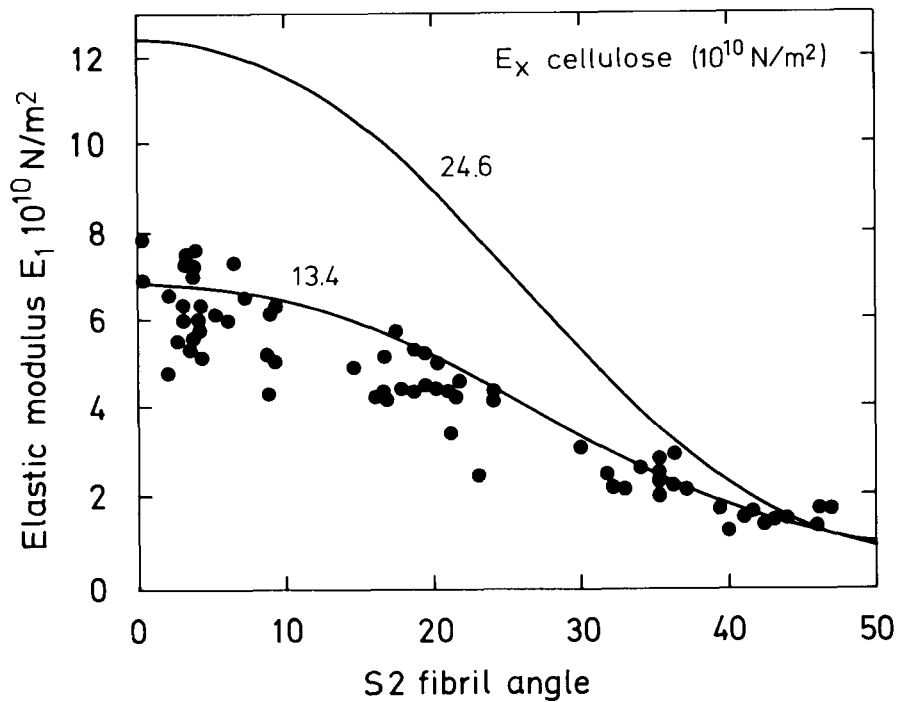


FIG. 4. The influence of the cellulose stiffness on the elastic modulus of pulp fibers as a function of the S2 mean fibril angle. The curves show calculated values assuming the value of the axial stiffness of cellulose estimated by Gillis (1969),  $24.6 \cdot 10^{10} \text{ N/m}^2$ , and that measured by Sakurada et al. (1962),  $13.4 \cdot 10^{10} \text{ N/m}^2$ . Points refer to single fiber measurements by Page et al. (1977).

the fiber modulus than does the theoretical value derived by Gillis, although Mark and Gillis (1973) have suggested that Gillis' theoretical values might be the more correct ones.

Preston's data (1960) for sisal fibers of low fibril angle indicate, however, a stiffness far above that normally experienced for wood and wood fibers. However, assuming a fiber based on the range of variation in composition and the relative importance of various layers normally found for sisal fibers (Lock 1962), these data of Preston are comparable with those predicted by the use of Sakurada's value for the elastic modulus of cellulose as illustrated in Fig. 5. According to Wilson (1971), sisal fibers are composed of 8% lignin, 11% hemicellulose, and 81% cellulose. The calculations given are based on this composition taking into consideration the fact that for the fibers used by Preston a few percent of the lignin may be lost due to the retting prior to the measurements (Balashov et al. 1957). The differences between the measured and calculated values may in part be due to a variation in lignin content with change in fibrillar angle. It is known that the lignin content changes from about 12% for the high fibrillar angles to about 6% for the low fibrillar angles (Lock 1962).

In view of the complex structure of the natural cellulosic fibers, there are obviously a number of variables to be considered in the development of models for

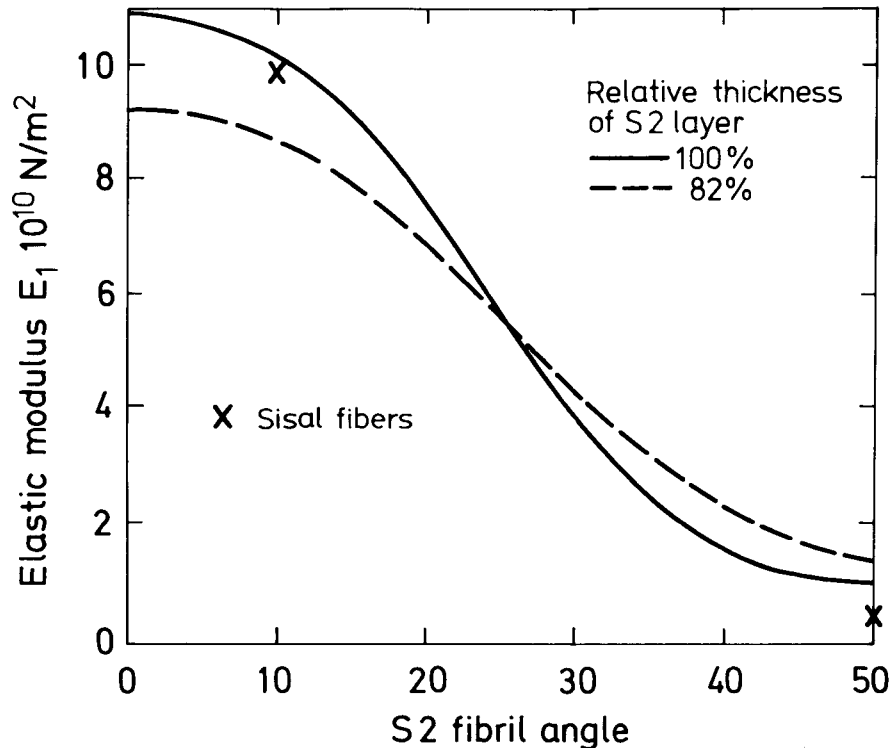


FIG. 5. The elastic modulus as a function of the S2 mean fibril angle estimated for sisal fibers. Points refer to measurements by Preston (1960).

the mechanical properties. Among the important variables are the amount of amorphous matrix component, the relative proportion of the S2 layer, and the fibril angles of the S1 and S3 layers.

The amount of matrix material has a large impact, the absolute moduli for the fiber being higher the less matrix material is present.

The relative thickness of the S2 layer changes the general shape of the fibrillar dependency curve appreciably, i.e., the higher the relative thickness of the S2 layer the higher is the modulus at low fibrillar angles, and the lower is the modulus at higher fibrillar angles, as exemplified for holocellulose fibers in Fig. 6.

Although the S1 and S3 layers are much thinner than the S2 layer, the fibrillar angles of these layers will have a significant influence on the fiber moduli, as seen in Fig. 7. Especially at high fibrillar angles of the S2 layer, the angles of the S1 and S3 layers become important. Mark (1972) also pointed out the influence of the S1 fibril angle on the elastic modulus, but this calculation was for the single fiber wall. In his case, the effect was not very accentuated for S1 fibril angles above 60° when very high fibril angles of the S2 layer, i.e., above 40°, were considered. In fact the higher angle of the S1 and S3 layers, i.e., 80°, gives a stiffer fiber than a S1 and S3 angle of 60° will render, a fact not apparent from the data of Mark (1972). This behavior is due to the general angular dependency of the elastic



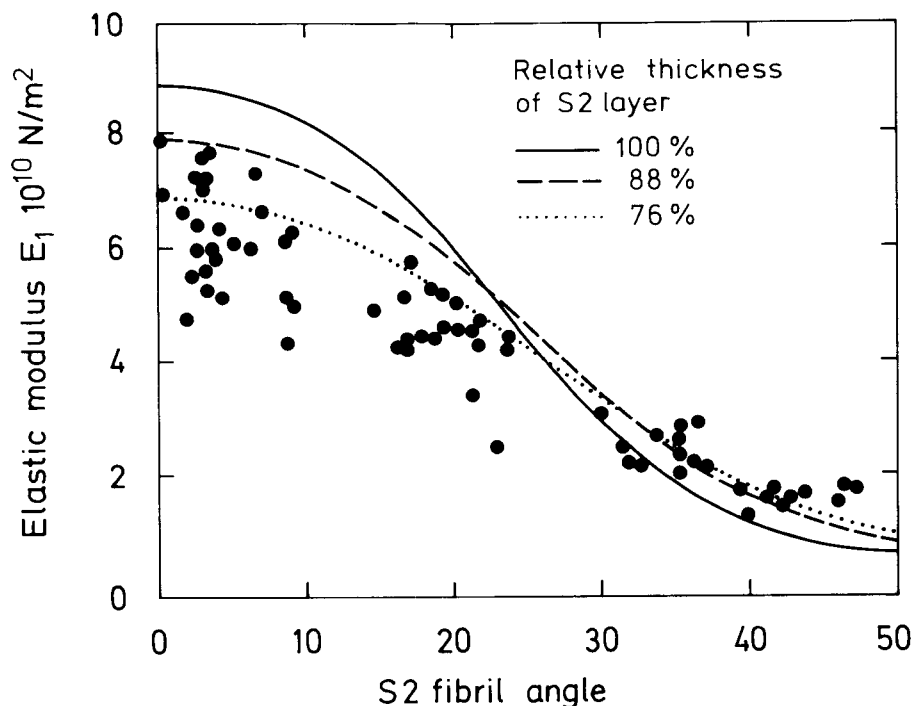


FIG. 6. The influence of the relative thickness of the S2 layer on the elastic modulus of wood fibers as a function of the S2 mean fibril angle. Points refer to single fiber measurements by Page et al. (1977).

modulus for an orthotropic material, which for the S1 and S3 layers has a minimum at about 60°.

This analysis suggests that the characterization of the single fiber has to be extremely accurate in order to rule out other reasons for variations in the data obtained. Today methods are available to determine yield level and crystallinity of a single fiber. Further refinement in predictive power demands knowledge of fibrillar distributions and the proportions of the various layers in the fiber wall.

It seems, however, that the approach used in this study gives a fairly good estimate of the single fiber modulus. Apparently the other factors demand experimental efforts that do not seem warranted in the light of their small contribution to the modulus.

In conclusion, at a given temperature and moisture content, yield, crystallinity, and fibrillar orientation of the S2 layer are the primary parameters necessary to predict the single fiber modulus. The effect of moisture content will be described in a subsequent paper.

#### *Effect of disordered zones*

It is generally recognized that sulphite pulps are stiffer than sulphate pulps. This fact can, as Page (1983) has suggested, be traced to the difference in fiber properties in the variation of the cellulose crystallite length. In fact, Page (1983) has shown

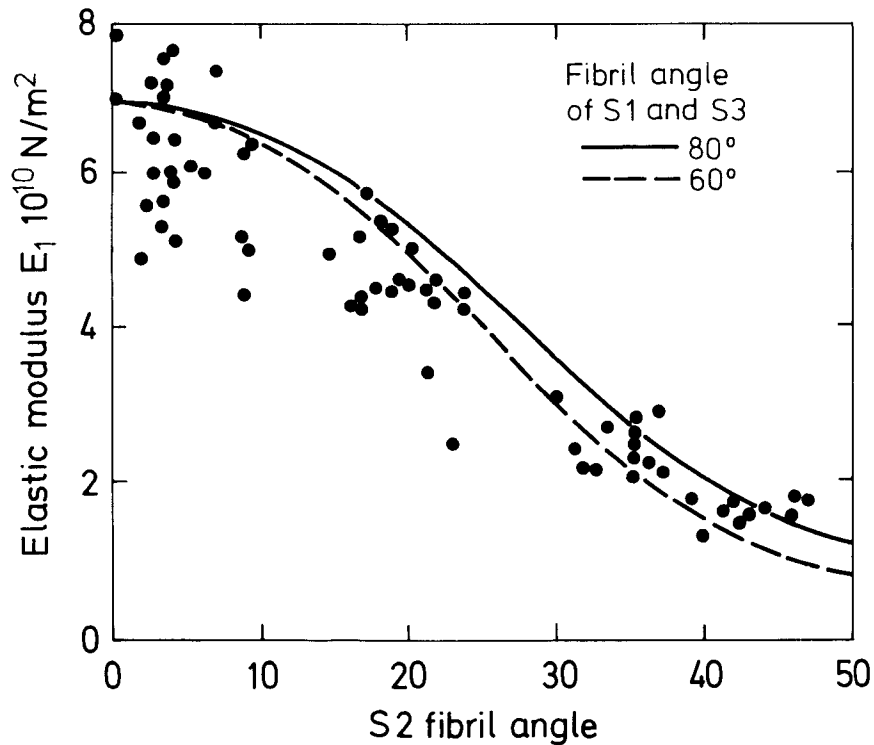


FIG. 7. The influence of fibrillar angle of the S1 and S3 layers on the elastic modulus of wood fibers as a function of the S2 mean fibril angle. Points refer to single fiber measurements by Page et al. (1977).

that an increased hydrolysis, i.e., reduction in the crystallite length, will successively lower the fiber stiffness.

This behavior may be mathematically treated by considering the cellulose crystallites merely as the reinforcing elements in a matrix of hemicellulose. This then implies that the disordered cellulose zones have the same properties as the amorphous hemicelluloses, which is probably not entirely correct. Figure 8 shows the effect on the tensile modulus of a hypothetical fiber of a change in the shape factor  $l/d$  for the reinforcing crystallites under these conditions. With the cellulose crystallite diameter considered to be about 25 Å and the crystallite length for a holocellulose fiber about 2,000 Å, the shape factor would be about 80, whereas a pulp fiber would have a shape factor of about 30 to 40. For fibers that have been mercerized, i.e., treated with higher concentrations of alkali, the shape factor may be as low as 5 to 10 (Krässig 1979). Thus for calculations on pulp fibers, the reduction in stiffness when taking the shape factor into account is not particularly pronounced compared with the original assumption that the entire microfibrils act as reinforcements. However when the cellulose is mercerized, there is certainly a noticeable effect in the calculations. This behavior has also been noticed in single fiber measurements by Page (1983).

A comparison may also be made with regenerated fibers. For normal rayon









