

# A THEORY FOR THE TRANSVERSE COLLAPSE OF WOOD PULP FIBRES

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## ABSTRACT

Theoretical considerations have led us to formulate a general equation for the collapse of wood pulp fibres in terms of transverse dimensions, transverse elastic modulus of the fibre wall, and the collapse pressure. This equation is in agreement with experimental results on the effects of fibre transverse dimensions on collapse. We have also developed an equation for the transverse elastic modulus of the fibre wall in terms of fibril angle and the orthotropic elastic constants. We can therefore confirm the dependence of collapse on the transverse elastic modulus through the effect of fibril angle on the collapse of chemical pulp fibres.

## INTRODUCTION

Fibre collapsibility is well recognized to be important for many physical properties of paper products. Numerous researchers have investigated the effects of transverse dimensions, and different chemical and mechanical treatments on the collapse of papermaking fibres [1–4]. It has been shown that fibre collapse is largely controlled by the collapse force that results from the papermaking processes, and two fundamental sets of fibre properties – the transverse dimensions and the mechanical properties of the fibre wall [4]. Previous experimental results have shown the effect of fibre transverse dimensions, such as wall thickness and perimeter, on the degree of fibre

collapse [4]. However, an equivalent understanding has not yet been fully extended to the effect of the mechanical properties of the fibre wall on collapse.

The mechanical properties of fibre depend largely on the chemical and physical structure of the fibre wall. Wood pulp fibres consist mainly of cellulose fibrils imbedded in a lignin and hemicellulose matrix. Although fibre wall properties can be modified through different pulping processes, they also depend strongly on the intrinsic structure of the fibre. The fibril angle, the angle between the fibrils and the fibre axis in the dominant  $S_2$  layer, is known to be one of the important factors that control the mechanical properties of wood pulp fibres. Previous experimental and theoretical work has shown that the longitudinal elastic modulus of wood pulp fibres decreases with increasing fibril angle [5].

Based on confocal laser scanning and polarization microscopies, techniques for measuring fibre transverse dimensions, collapse index, and fibril angle of individual wood pulp fibres have been developed [4,6,7]. We have also recently shown that fibril angle strongly affects the degree of fibre collapse [8]. At any given fibre transverse dimensions, fibres with low fibril angle tend to be more collapsible than fibres with high fibril angle. Similar to longitudinal elastic modulus, this behaviour signifies the strong dependence of the fibre transverse mechanical properties on its fibril angle. One of the major aims of this paper is to describe how fibre collapse, under surface tension during drying, depends on the fibre wall's transverse elastic modulus, itself a function of fibril angle of the  $S_2$  layer.

This paper starts with an empirical collapse equation proposed earlier [4]. With some assumptions and theoretical considerations, we propose a general equation for the collapse behaviour of a tubular pulp fibre in terms of transverse dimensions, transverse elastic modulus of the fibre wall, and the collapse pressure. This collapse equation will be verified in the context of our previous experimental work on the effect of transverse dimensions on collapse. To understand the dependence of collapse on transverse elastic modulus in the collapse equation, we first develop a theoretical equation for transverse elastic modulus of fibre wall in terms of fibril angle and the orthotropic elastic constants by modifying the longitudinal elastic modulus equation of Page *et al.* [5]. The collapse equation and the theoretical transverse elastic modulus will be shown to provide a good description of the effect of fibril angle on fibre collapse for low-yield chemical pulp fibres. The collapse behaviour of mechanical pulp fibres will also be discussed.

## COLLAPSE EQUATION

The collapse index, CI, for a fibre, based on its cross-sectional image, has been previously defined as the fractional loss of lumen area that results from fibre processing [4].

$$CI = 1 - \frac{LA}{LA_0}, \quad (1)$$

where LA is the lumen area measured from a fibre cross-sectional image, and LA<sub>0</sub> is the original lumen area of the fibre before collapse. An explicit equation relating CI to the transverse dimensions and the mechanical properties of fibre wall was shown to be [4],

$$CI = 1 - \exp[-a(\varepsilon, F)G^\beta], \quad (2)$$

where  $G$  is a fibre geometry factor which depends on fibre transverse dimensions, and  $\beta$  determines the sensitivity of collapse on this factor. With surface tension during drying as the dominant collapse force acting in the lumen,  $G$  can be set to  $LP/2\pi T$  for tubular fibres with  $LP \gg T$ ,  $LP$  being the fibre lumen perimeter and  $T$  the fibre wall thickness. The value for  $\beta$  was found experimentally to be nearly 2, based mainly on the collapse behaviour of earlywood pulp fibres [4]. The other term,  $a$ , in the exponent is considered to be a function of the transverse mechanical properties of the fibre wall,  $\varepsilon$ , and of the applied collapse pressure  $F$ . Fibres with higher  $a$  values are more collapsible. For fibres subjected to a similar  $F$ ,  $a$  is related to fibre wall compliance, and depends only on the transverse mechanical properties of the fibre wall.

In an attempt to formulate the effect of the transverse mechanical properties of the fibre wall on collapse, we assume that fibre collapse depends mainly on the transverse bending of the fibre wall. Fibres with more flexible walls that can be bent easily in the transverse direction are more collapsible. We further assume that the wall transverse deformations are within the elastic regime, that fibre walls are only subjected to plane stress, and that the thickness of fibre wall does not change during collapse. Therefore, the value of  $a$  in Equation (2) can be simply assumed to be a monotonically increasing function of fibre wall transverse compliance,  $S_T$ , which is the reciprocal of fibre wall transverse elastic modulus,  $E_T$ . As a first order approximation, we assume that

$$a(E_T, F) \propto \kappa(F) (S_T)^\lambda \propto F^\nu \left(\frac{1}{E_T}\right)^\lambda, \quad (3)$$

where  $\lambda$  and  $\nu$  are the powers that respectively determine the sensitivity of collapse on  $S_T$  and  $F$ . The term,  $\kappa(F)$ , which is also a monotonically increasing function of  $F$ , because higher fibre collapse occurs with increasing  $F$ , is assumed proportional to  $F^\nu$ . Since, by definition of elastic modulus, the pressure required to induce a similar deformation in the wall is proportional to the elastic modulus of the wall material, this allows us to conclude that  $\nu$  equals  $\lambda$ . Equation (3) can now be written as

$$a(E_T, F) \propto \left(\frac{F}{E_T}\right)^\lambda. \quad (4)$$

Basically,  $a(E_T, F)$  is a function of  $F/E_T$ , which according to Hooke's law, is proportional to the strain in the fibre wall in transverse direction. By substituting Equation (4) into Equation (2), a general equation for CI in terms of fibre geometry factor, transverse elastic modulus, and collapse pressure can now be written as,

$$CI = 1 - \exp \left[ - \left( \mu \frac{F}{E_T} \right)^\lambda G^\beta \right], \quad (5)$$

where  $\mu$  is a constant. It is worth noting that the dependence of CI on  $F$  in this formulation is consistent with the S-shaped load-deformation behaviour of a wet fibre in the region of fibre collapse [2].

From an analysis in Appendix A, the exponential term in Equation 5 is shown to relate to the square of the relative decrease in fibre thickness in the transverse direction for slightly collapsed fibres. This analysis also allows us to conclude that  $\lambda$  equals 2. Therefore, the general collapse equation for tubular fibres can now be written as

$$CI = 1 - \exp \left[ - \left( \mu \frac{F}{E_T} \right)^2 G^\beta \right]. \quad (6)$$

The relationship between  $\lambda$  and  $\beta$  in Equation (5) can be further established by understanding the relationship between  $F$  and  $G$ . Luce [9] performed a series of lateral compression tests on simulated wood fibres that had layered structure. It was found that the collapse force  $F$  required to close the lumen of these fibre models was linearly proportional to a shape factor, SF, defined as

$$\text{Collapse force} \propto SF = \frac{D^2 - D_i^2}{D^2 + D_i^2}, \quad (7)$$

where  $D$  and  $D_i$  are the outer and inner diameters of the simulated fibre. For fibres with thin walls,  $T \ll D$ ;  $T$  is the fibre wall thickness. Therefore,  $SF$  approximates to  $2T/D$ . It follows that  $SF \cong 2\pi T/LP = 1/G$  for thin-walled fibres. Therefore, the collapse force required to close the lumen of a fibre is inversely proportional to the fibre geometry factor, that is, Collapse force  $F \propto 1/G$ . It follows that the simulated fibres with the same  $FG$  values will have the same  $CI$ . If this conclusion is applied to the collapse Equation (5), it suggests that the powers  $\lambda$  and  $\beta$  that determine the sensitivity of collapse on the collapse force and the fibre's geometry factor respectively should be the same, and will both equal 2. Therefore, Equation (6) can now be written as

$$CI = 1 - \exp \left[ - \left( \mu \frac{F}{E_T} G \right)^2 \right], \quad (8)$$

for tubular fibres with layered structure. This equation also shows that the sensitivity of fibre collapse to  $1/E_T$  and  $G$  is similar. For example, if  $G = LP/2\pi T$ , in a fibre with a given  $LP$ ,  $CI$  should increase in a similar way if either the wall thickness or the transverse elastic modulus is reduced.

The dependence of collapse on the fibre geometry factor in Equation (8) is consistent with our experimental results where the value for  $\beta$  was found to be approximately 2 for low-yield kraft pulp fibres [4]. However, the dependence of collapse on the other term  $a$ , which was reasoned to be  $(F/E_T)^\lambda$  in Equation (4) with  $\lambda = 2$ , is yet to be confirmed. We will now provide experimental evidence to support the dependence of  $a$  on  $E_T$ , which itself is a function of the fibril angle.

## **EFFECT OF THE ELASTIC MODULUS OF THE FIBRE WALL ON COLLAPSE**

Recent work has demonstrated the effect of fibril angle on the collapse behaviour of chemical and mechanical pulp fibres [8]. The value of  $a$  was found to decrease as the fibril angle increased as shown in Figure 1 for two laboratory-made unbleached kraft pulps. These data were obtained mainly on the collapse of earlywood fibres subjected to surface tension during drying. We will now use these data to establish the dependence of  $a$  on the fibre's transverse elastic modulus as shown in Equation (4). To do that, we first

develop a theoretical equation for the transverse elastic modulus of the fibre wall in terms of fibril angle and the orthotropic elastic constants by modifying an existing equation for the longitudinal elastic modulus of the fibre.

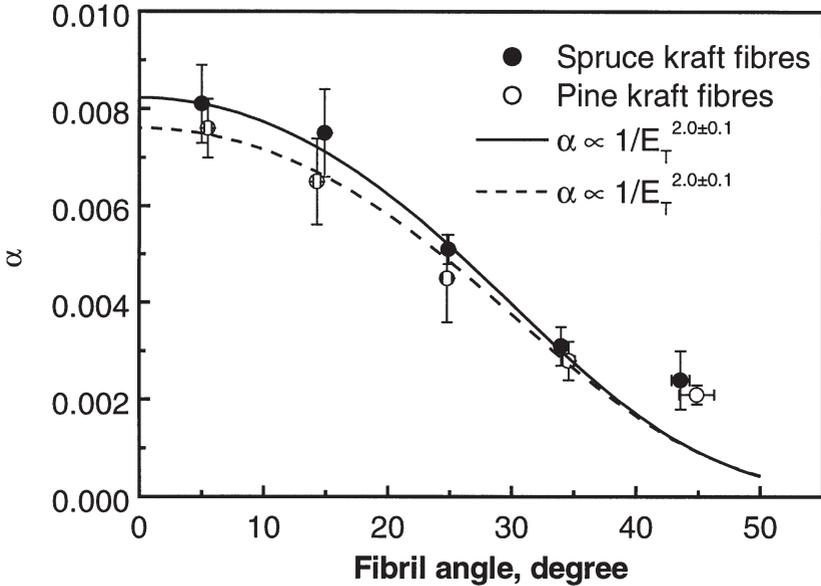
Page *et al.* [5] used orthotropic elasticity theory to derive for chemical pulp fibres an equation for the longitudinal elastic modulus,  $E_L$ , of a fibre wall in terms of fibril angle,  $\theta$ , and a set of four orthotropic compliance constants. This analysis considered only the effect from the  $S_2$  layer, and disregarded the effects from all other layers. Since the shear deformation of fibre wall was restricted in their experiments, the boundary condition for shear restraint was used. This equation has successfully described the dependence of  $E_L$  on  $\theta$  for undamaged chemical pulp fibres.

To derive a theoretical  $E_T$ , we will similarly assume that the  $E_T$  of the fibre wall depends only on the  $S_2$  layer, and the effects from other layers are absent. The fibres tested in our collapse work were dried on glass slides. The contact between the fibre and glass restricted shear deformation along the fibre. This corresponds to the same boundary condition as used by Page *et al.* [5]. Based on these simplifications and boundary conditions, there should be no difference in the formulation between  $E_L$  and  $E_T$  except that the direction of microfibrils would be referred to a different axis. Therefore, the  $E_L$  equation of Page *et al.* [5] can be modified to describe the transverse elastic modulus  $E_T$  of the fibre wall in our experiments as follows:

$$E_T(\theta) = E_L(90^\circ - \theta). \quad (9)$$

The equation for  $E_T$  in terms of fibril angle and four orthotropic compliance constants is presented in Appendix B. Equation (9) shows that, for  $\theta = 45^\circ$ ,  $E_T$  will be equal to  $E_L$ . For  $\theta = 0^\circ$ , the elastic modulus parallel to the fibril direction,  $E_2$ , determines the  $E_L$  of a fibre; for  $\theta = 90^\circ$ ,  $E_2$  determines the  $E_T$  of the fibre (see Figure B1). This  $E_T$  in Equation (B1) is used to describe the transverse tension and compression deformations in a fibre wall during fibre collapse.

By putting theoretical  $E_T$  of Equation (B1) into Equation (4), we are able to describe  $a$  in terms of  $\theta$ . If this equation for  $a(\theta)$  is correct, it should be able to describe the experimental data shown in Figure 1. However, the  $a(\theta)$  equation requires six other parameters:  $F$  and  $\lambda$  from Equation (4), and four orthotropic compliance constants of the fibre wall material,  $S_{11}$ ,  $S_{22}$ ,  $S_{66}$ , and  $S_{12}$  from Equation (B1). If all four orthotropic compliance constants were known, and these four constants and  $F$  were assumed to be similar for all fibres, the shape of  $a$  versus  $\theta$  curve as shown in Figure 1 will uniquely determine the  $\lambda$  value. For instance, when the orthotropic compliance constants for dry chemical pulp fibre walls in Table BI were used, the  $\lambda$  values



**Figure 1**  $a$  versus  $\theta$  for a never-dried spruce kraft pulp cooked to 47.1% yield with kappa number 29.0, and a once-dried pine kraft pulp cooked to 45.0% yield with kappa number 31.0 [8]. These  $\theta$  values represent the group mean fibril angles where fibres were divided into groups based on their fibril angles  $0^\circ$  to  $10^\circ$ ,  $10^\circ$  to  $20^\circ$ ,  $20^\circ$  to  $30^\circ$ , and so on. The  $a$  value was obtained for each group with  $\beta$  set to 2 in Equation (2)[4]. The curves are best fits of Equation (4) to the data with the theoretical  $E_T(\theta)$  described in Equation (C1) and the orthotropic compliance constants  $S_{11}/S_{66}$  set to 0.74. The best-fit values for  $\lambda$  were found to be  $2.0 \pm 0.1$  for both pulps.

were found to be  $2.2 \pm 0.1$  from the fits as shown in Figure B2. The present data sets can be fitted well using many different values of  $S_{11}$ ,  $S_{22}$ ,  $S_{66}$ , and  $S_{12}$ . However, only the best fit with the correct values of the material properties for the fibre wall will generate a correct  $\lambda$  value.

Most values quoted for compliance constants in the literature apply only to the dry fibre wall. These values cannot be used for the collapse of wet fibres during drying. Fibres soften when wet. The relative decrease in  $E_1$  (the elastic modulus perpendicular to the fibril direction) on wetting is much greater than that in  $E_2$  (the elastic modulus parallel to the fibril direction). The zero-span tensile strength tests of pulps with low fibril angle have shown that the strength of wet kraft pulp fibres can be up to 30% lower than that of dry

fibres [10], indicating that the difference between the longitudinal elastic modulus of dry and wet fibres can be generally small. However, the transverse elastic modulus of wet fibres has been shown to be three orders of magnitude lower than that of dry fibres [11]. Even for dry fibres,  $E_2$  is known to be an order of magnitude higher than  $E_1$  [5]. Therefore,  $S_{22}$  (or  $1/E_2$ ) should be a few orders of magnitude smaller than  $S_{11}$  (or  $1/E_1$ ) for wet fibres. The magnitude of  $S_{12}$  is also known to be very small compared to  $S_{11}$  even for dry fibres. When the fibres are wet, the magnitude of  $S_{12(\text{wet})}$  is less than  $(S_{11(\text{wet})}S_{22(\text{wet})})^{1/2}$  [12], and therefore  $S_{12}$  is also very small compared to  $S_{11}$ . These facts mean that both  $S_{22}$  and  $S_{12}$  can be set to zero without significant loss of accuracy. Because  $\lambda$  depends on the shape of the curve, only the relative magnitudes of the compliance constants are required for the fitting in order to obtain the best value for  $\lambda$ . Thus, with both  $S_{22}$  and  $S_{12}$  set to zero, only the ratio  $S_{11}/S_{66}$  is required to obtain a proper  $\lambda$ . The value  $\lambda$  obtained from such best fit depends on the input value of  $S_{11}/S_{66}$  as discussed in detail in Appendix C.

The value of  $S_{11}/S_{66} = 1.13 / 1.52 = 0.74$  for dry fibres reported by Page *et al.* [5] was used in this work. This choice was based on three reasons: first, the chemical fibres used in both sets of work were similar in yields, second, the changes in both  $S_{11}$  and  $S_{66}$  between dry and wet fibres were due mainly to the same softening of the hemicelluloses [11], and third, this value fits well to the data (Appendix C). The ratio of  $S_{11}/S_{66} = 0.74$  may seem high because when  $S_{11}$  is set to  $1.13 \times 10^{-10} \text{ Pa}^{-1}$ , the value  $G_{\text{sm}} (1/S_{66}) = 6.6 \text{ GPa}$  is high relative to other values reported in the literature. This relatively high value for  $G_{\text{sm}}$  may be a result of the shear-deformation restraint imposed on the fibres by the adhesion of fibres to glass. This condition is similar to the restraint on fibres from twisting by the test clamps used by Page *et al.* [5,13]. With  $S_{11}/S_{66}$  set to 0.74 and  $S_{22}$  and  $S_{12}$  set to zero,  $\lambda$  was found to be  $2.0 \pm 0.1$  from the best fits for both spruce and pine kraft pulp fibres as shown in Figure 1.

The good fits for both kraft pulp samples support the hypothesis that  $a$  is a function of  $E_T$  as proposed in Equation (4). The experimental value of  $\lambda$  has been shown to be around 2 when  $\beta$  was set to be 2. This is in excellent agreement with our conclusion based on theory that  $\beta = \lambda = 2$  as shown in Equation (8). Thus, both theoretical considerations and experimental results validate Equation (8) for describing the collapse behaviour of low-yield earlywood kraft fibres.

The transverse compliance of a wet fibre is shown to be a function of two orthotropic compliance constants  $S_{11}$  and  $S_{66}$ , with the effect from  $S_{22}$  being negligible (see Equation (C1)). This implies that transverse collapse of a fibre depends mainly on the elastic modulus transverse to the fibril direction and shear modulus in the (1,2) plane, but not on the elastic modulus in the fibril

direction. Therefore, fibre collapse is mainly determined by the physical properties of the inter-fibrillar materials, i.e., the level of cross-linking between the microfibrils. The extent of cross-linking, and therefore the resistance against fibre collapse can be decreased through various chemical and mechanical treatments.

It is known that for the collapse of rigid, thin-walled cylinders, the external collapse pressure is proportional to  $(T/D)^3$  [14]. On the other hand, the force that was required to collapse the simulated fibres with layered structure was found to be proportional to  $T/D$  [9]. The difference is mainly due to the very weak shear constraints between layers in the simulated fibres. Our present results for kraft fibres are more consistent with Luce's findings for simulated fibres, where  $F \propto E/G$ , than with the findings for the rigid-walled cylinder, where  $F \propto E/G^3$ . This implies that during collapse, wet kraft fibres have only very weak shear constraints between layers of fibrils in the fibre wall. However, for fibres with a higher concentration of cross-linking between the microfibrils (such as in mechanical pulp fibres with high lignin content), shear constraints between layers may become significant. The relationship between  $F$  and  $G$  for collapsing such fibres would lie between the layered and the rigid-wall structures. Therefore, the  $\beta$  values for mechanical pulp fibres may be larger than 2. In general, the ratio  $\lambda/\beta$  should be less than or equal to one but greater than or equal to  $1/3$ , i.e.,  $1 \geq \lambda/\beta \geq 1/3$ .

It would be important to check whether Equation (4) with  $\lambda = 2$  is consistent with the existing  $a$  and  $E_T$  data. In our earlier work,  $a$  values for low-yield kraft pulp and thermomechanical pulp (TMP) fibres were found to be 0.04 and 0.005 respectively [4]. The ratio  $a_{\text{kraft}}/a_{\text{TMP}}$  ( $= 0.04/0.005$ ) therefore was 8. Scallan *et al.* have used swelling experiments to show that the bulk elastic moduli of wet fibre walls were around 2.5 MPa and 9.5 MPa for kraft and TMP pulps respectively [11]. These values should approximately correspond to the transverse elastic moduli of low fibril angle fibres, because swelling occurs mainly in a direction transverse to the fibrils. The  $[E_{T(\text{TMP})}/E_{T(\text{kraft})}]^2$  reported by Scallan *et al.* is 14, which is of the same order of magnitude as the value 8 for  $a_{\text{kraft}}/a_{\text{TMP}}$  found from our work.

Fibre collapse results from deformation and bending of the fibre wall and the corners. In the absence of external pressure, the lumen collapses under surface tension as the water in it evaporates, while the fibre wall is wet and pliable. The fact that these deformations are expected not to be purely elastic makes a comprehensive theoretical treatment of fibre collapse very difficult. In order to apply known orthotropic elasticity theory to the relationship between  $\theta$  and  $a$ , the deformation and bending of the wet fibre wall during collapse were assumed to be elastic and dependent only on the transverse elastic modulus of the  $S_2$  layer of the wet fibre wall. It was also assumed that

the deformation of fibre corners would show the same dependence on  $\theta$  as in the  $S_2$  layer. The theoretical  $E_T$  equation presented in Appendix B applies only to the  $S_2$  layer of the fibre wall. The fact that this equation fits both the spruce and pine data so well suggests that the deformation for wet fibres can be modeled in the elastic regime, and the  $S_1$  and  $S_3$  layers do not significantly contribute to the transverse elastic modulus of the low-yield kraft fibres. The latter may be due to the fact that if lignin and hemicellulose are largely removed during kraft pulping, the cross-linking between  $S_1$  and  $S_2$  layers for wet fibres is weak. However, if the cross-linking is strong such as in the lignin-rich mechanical pulp fibres, the effect of high fibril angle of the  $S_1$  layer on  $E_T$ , and therefore on fibre collapse may be significant.

## COLLAPSE BEHAVIOUR OF MECHANICAL PULP FIBRES

The above analysis has explained satisfactorily the collapse behaviour of chemical pulp fibres. Can this analysis describe the collapse behaviour of mechanical pulp fibres? Our work has shown that the value of  $a$  for mechanical pulp fibres also decreased as the fibril angle  $\theta$  increased [8]. Although the general collapse Equation (6) will apply to all types of fibres, it should be recognized that mechanical pulp fibres are different from chemical pulp fibres in three ways.

1. The physical properties of mechanical pulp fibres are quite different from that of chemical pulp fibres because they have different constituents in their walls. This will lead to a different value for  $\beta$ , which will possibly be greater than 2, as discussed earlier. Moreover, the orthotropic compliance constants for mechanical pulp fibres will be different from those of the chemical pulp fibres.
2. For chemical pulps, we assumed that all fibres received similar delignification treatment, and the orthotropic compliance constants for the wall were the same for all fibres. However, this will not be the case for mechanical pulp fibres. Mechanical pulping is heterogeneous, and the nature and the amount of mechanical treatment each fibre receives depend on its morphology, and possibly the fibril angle. For example, it has been shown that latewood fibres have a larger reduction in wall thickness during mechanical pulping than the earlywood fibres, and that earlywood fibres tend to break more frequently [15]. Therefore, the orthotropic compliance constants for the fibre wall will be different for different fibres.
3. The  $S_1$  and  $S_3$  layers play little role in chemical pulp fibres, but can have a significant effect on the mechanical pulp fibres. Therefore, the theoretical

$E_T$  in Equation (B1), which only considered the effects from the  $S_2$  layer, will be an oversimplification for mechanical pulp fibres. Furthermore, the removal of  $S_1$  layer, which will be different for different fibres, will make calculation of  $E_T$  more complex.

In summary, some of the assumptions made for the chemical pulp fibres will not be justifiable for mechanical pulp fibres, and these issues will be discussed in a separate publication.

## CONCLUDING REMARKS

A general equation that describes the collapse of wood pulp fibres in terms of a fibre geometry factor  $LP/2\pi T$ , transverse elastic modulus  $E_T$ , and the external collapse pressure is proposed. This equation is consistent with the experimental results on the effect of fibre transverse dimensions on collapse. Combined with a theoretical  $E_T$  of the  $S_2$  layer of the fibre wall, this equation also provides a good description of the effect of fibril angle on fibre collapse for low-yield chemical pulp fibres. Both theory and experiments consistently show that the dependence of fibre collapse on  $1/E_T$  and on  $LP/2\pi T$  are similar.

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## APPENDIX A

*A physical interpretation of the term  $(\mu F/E_T)^\lambda G^\beta$  in Equation (5) and the theoretical value for  $\lambda$ .*

The exponential term  $(\mu F/E_T)^\lambda G^\beta$  in Equation (5) is a dimensionless parameter. For a slightly flattened fibre,  $(\mu F/E_T)^\lambda G^\beta$  is much less than one. Since  $e^{-x} \approx (1 - x)$  for  $x \ll 1$ , Equation (5) can be rewritten as

$$CI \approx \left( \frac{\mu F}{E_T} \right)^\lambda G^\beta. \quad (\text{A1})$$

If the original fibre lumen is assumed to be circular in cross section with radius  $r$  and area  $LA_0$ , then a slightly flattened fibre will be assumed to be elliptical in shape and will have the same lumen perimeter  $LP$  as the original fibre. When  $\delta r$  is small compared to  $r$ , as in a slightly flattened fibre, it can be shown that

$$CI = 1 - \frac{LA}{LA_0} \approx \left(\frac{\delta r}{r}\right)^2, \quad (A2)$$

where  $(\delta r/r)$  approximates the fractional decrease in fibre thickness for thin-walled fibres as shown in Figure A1a. Equation (A2) can also be obtained on fibres with an original square lumen, subsequently flattened to be a rectangle as shown in Figure A1b. Equation (A2) shows that for a slightly flattened fibre,  $CI$  is equivalent to the square of the fractional decrease in fibre thickness. The use of fractional decrease in fibre thickness to define the degree of collapse has been reported earlier by Görres *et al.* [16].

Combining Equations (A1) and (A2) yields

$$\left(\frac{\mu F}{E_T}\right)^{\lambda/2} G^{\beta/2} = \left(\frac{\delta r}{r}\right), \quad (A3)$$

which shows that the term  $(\mu F/E_T)^{\lambda/2} G^{\beta/2}$  is equivalent to the relative change in fibre thickness. If  $\lambda$  is set to be 2, we have

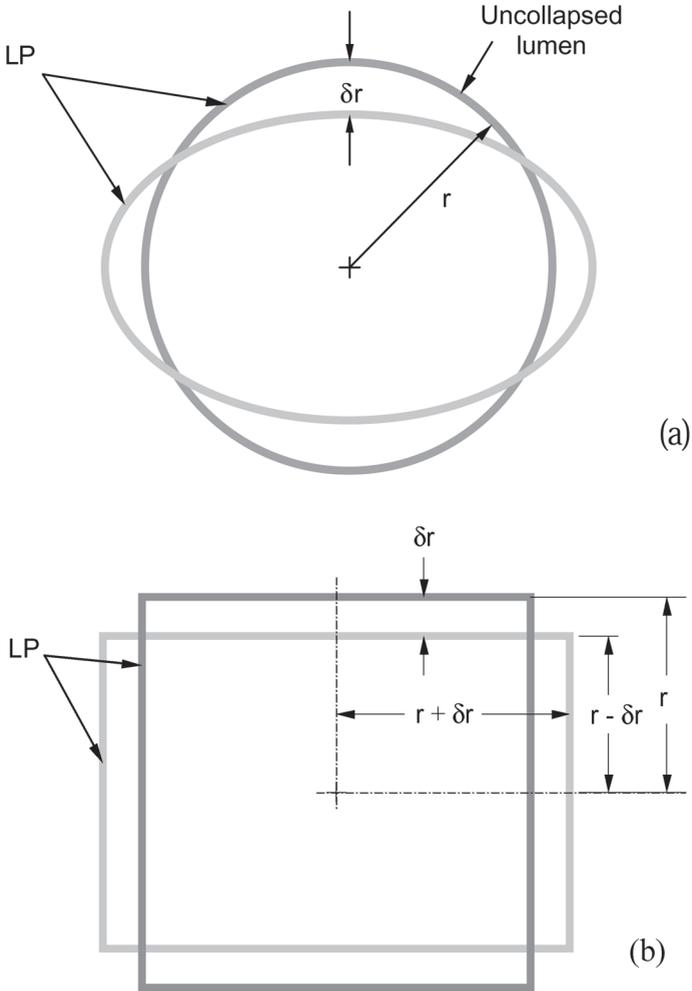
$$\frac{\mu F G^{\beta/2}}{E_T} = \left(\frac{\delta r}{r}\right). \quad (A4)$$

The change in fibre thickness  $\delta r$  during fibre collapse is considered proportional to the transverse deflection or bending of the fibre wall. Equation (A4) is consistent with the general formulation for a bending beam where the degree of deflection or bending is proportional to  $F/E$ , the strain of the fibre wall [14]. Hence, this analysis provides an argument for  $\lambda$  to equal 2.

## **APPENDIX B**

### *The transverse elastic modulus of a fibre wall*

The transverse elastic modulus of a fibre wall in terms of fibril angle and orthotropic compliance constants can be obtained by modifying the longi-

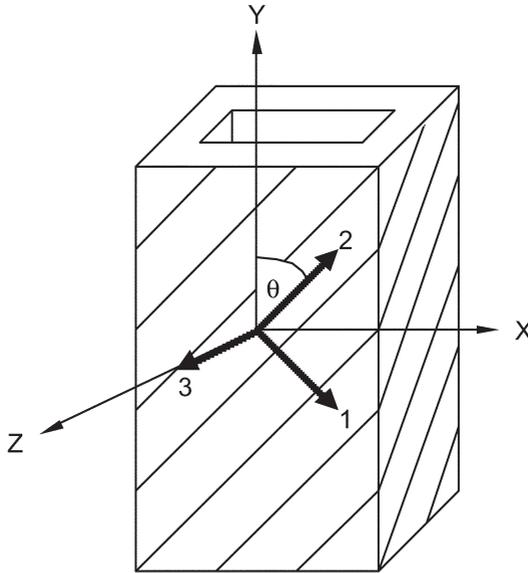


**Figure A1** The flattening of a fibre with (a) a circular and (b) a square lumen of area  $LA_0$  to form a fibre with an elliptical or a rectangular lumen of area  $LA$ . The decrease in fibre thickness is  $2\delta r$ . The lumen perimeter is assumed to be the same before and after flattening.

tudinal elastic modulus equation of Page *et al.* [5]. According to the angle modification discussed in Equation (9), the equation for  $E_T$  is

$$S_T(\theta) = \frac{I}{E_T(\theta)} = \frac{\cos^4\theta[(S_{11} + S_{22} + 2S_{12})S_{66} - 4(S_{11}S_{22} - S_{12}^2)] + 2\cos^2\theta[2(S_{11}S_{22} - S_{12}^2) - (S_{22} + S_{12})S_{66}] + S_{22}S_{66}}{S_{66} - 4\sin^2\theta\cos^2\theta(S_{66} + 2S_{12} - S_{11} - S_{22})}, \quad (\text{B1})$$

where  $\theta$  is the fibril angle and  $S_{11}$ ,  $S_{22}$ ,  $S_{12}$  and  $S_{66}$  are the principal compliance constants of the fibre wall. The principal directions (1, 2 and 3) of the fibre wall and the fibre axes (x, y and z) follow the conventions used by Page *et al.* [5].  $E_1$  and  $E_2$  respectively are the elastic moduli of the fibre wall in directions transverse and parallel to the fibril direction (Figure B1).

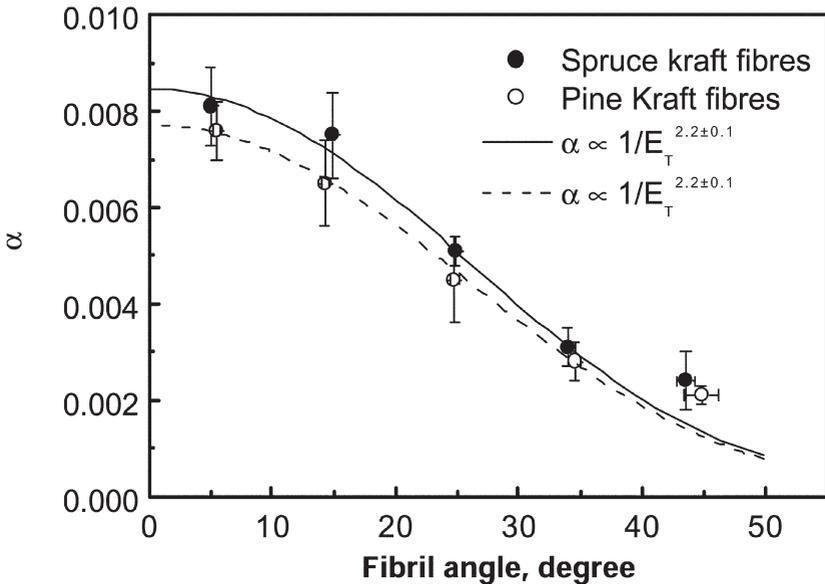


**Figure B1** Schematic diagram of cellulose microfibrils in the  $S_2$  layer of a wood pulp fibre wall. The fibril angle,  $\theta$ , is defined as indicated.

Table B1 shows the orthotropic compliance constants obtained by Page *et al.* [5] for dry chemical pulp fibres of yields similar to those used in our work. When these compliance constants were used to fit our  $\alpha$  versus  $\theta$  results, the  $\lambda$  values were found to be  $2.2 \pm 0.1$  for both spruce and pine kraft pulp fibres as shown in Figure B2.

**Table B1** Orthotropic compliance constants of a dry chemical pulp fibre wall [5].  $G_{sm}$  and  $\nu_{21}$  are the shear modulus and Poisson ratio in the (1, 2) plane.

	$10^{-10} \text{ Pa}^{-1}$		$10^{10} \text{ Pa}$
$S_{11}$	1.13	$E_1$	0.88
$S_{22}$	0.13	$E_2$	7.69
$S_{66}$	1.52	$G_{sm}$	0.66
$S_{12}$	-0.04	$\nu_{21}$	0.31



**Figure B2** Best fits of Equation (4) to the  $\alpha$  versus  $\theta$  data for spruce and pine kraft pulp fibres with the orthotropic compliance constants for dry chemical pulp fibres shown in Table B1. The data points in the  $40^\circ$  and  $50^\circ$  range were not included in the fits as they contained relatively insufficient measurements [8]. The best-fit values for  $\lambda$  were found to be  $2.2 \pm 0.1$  for both pulps.

**APPENDIX C**

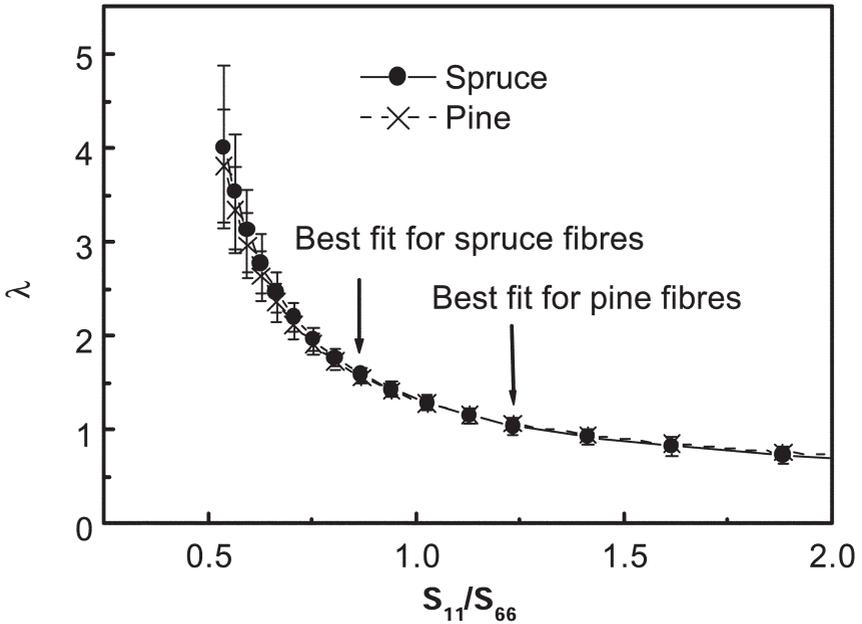
$\lambda$  as function of  $S_{11}/S_{66}$

If the principal compliance constants  $S_{12}$  and  $S_{22}$  are set to zero for a wet fibre wall, the transverse compliance in Equation (B1) becomes

$$S_T(\theta) = \frac{1}{E_T(\theta)} = S_{11} \frac{\cos^4 \theta}{1 - 4\sin^2 \theta \cos^2 \theta (1 - S_{11}/S_{66})} \quad (C1)$$

This equation shows that the relative dependence of  $S_T(\theta)$ , hence  $a$ , on  $\theta$  will depend only on the ratio  $S_{11}/S_{66}$ .

Figure C1 shows the best-fit values of  $\lambda$  as function of  $S_{11}/S_{66}$  for the  $a$  versus  $\theta$  data in Figure 1.  $\lambda$  increases in a similar way for both spruce and pine kraft fibres as  $S_{11}/S_{66}$  decreases. If both  $\lambda$  and  $S_{11}/S_{66}$  are allowed to vary freely,



**Figure C1** Values for  $\lambda$  as a function of  $S_{11}/S_{66}$ . The best-fit values for  $\lambda$  and  $S_{11}/S_{66}$  are also indicated.

the best fit values for  $S_{11}/S_{66}$  and  $\lambda$  are 0.85 and 1.63 for spruce and 1.23 and 1.1 for pine.

Since shear modulus is normally smaller than elastic modulus,  $S_{66}$  should be greater than  $S_{11}$ , and  $S_{11}/S_{66}$  should be less than one. Figure C1 shows that the  $\lambda$  versus  $S_{11}/S_{66}$  fit deteriorates very quickly (as indicated by the error bars) as  $S_{11}/S_{66}$  is decreased below the best-fit value. The value for  $S_{11}/S_{66}$  must be greater than 0.65 for the standard error in  $\lambda$  to be within 10%. Therefore, a reasonable value for  $S_{11}/S_{66}$  should be in the range of 0.65 to 1, and the value of Page *et al.* of 0.74 is within this range.

## Transcription of Discussion

# A THEORY FOR THE TRANSVERSE COLLAPSE OF WOOD PULP FIBRES

*Ho Fan Jang*

Pulp and Paper Research Institute of Canada

*Kari Ebeling* UPM-Kymmene Corporation

How does the processing of the fibres for instance refining affect the collapsibility and its dependence on the fibril angle. Most of the papermakers use chemical fibres after refining. We all know that refining greatly increases collapsibility. Does it increase the collapsibility in different ways depending of the microfibrillar angle?

*Ho Fan Jang*

That is a very good question; the transverse elastic modulus for low-yield chemical pulp fibres is quite low. Most of these fibres have a high degree of collapse to start with. Further refining of low yield chemical pulp fibres will not increase their collapsibility significantly. However, mechanical pulp fibres, which have much higher transverse elastic modulus, do not collapse readily. I believe that refining will increase the collapsibility of these fibres differently, depending on their properties such as fibril angle.

*Tad Mahoney* JM Huber

I would think that the collapsibility would depend on the degree of fibre swelling, in addition to the fibril angle, so I am wondering if that enters into your model or into your thinking?

*Derek Page* Institute of Paper Science & Technology

Maybe I can help you out here. Are you thinking that the elastic modulus of the fibre in the more swollen state would be less, and that would lead you to more collapse the more swollen the fibre is?

*Discussion*

*Ho Fan Jang*

Yes, you are right. The swelling will decrease the transverse elastic modulus of the fibre wall; therefore for a given collapse pressure, more swelling of the fibre will lead to more collapse.

*Lars Wågberg* Mid-Sweden University

I wonder if you checked how you're measured values would scale if you change the surface tension of your solutions? You say that the collapse comes from the surface tension forces and I simply wonder if you have checked that you get the right scaling if you change the surface tension of the water?

*Ho Fan Jang*

The surface tension affects the collapse pressure. If we reduce the surface tension, the collapse pressure will decrease. However, we haven't done any experiments to demonstrate this.

*Ian Parker* APPI/Monash University

The results that you show from Derek Page's work are presumably for the elastic modulus of dry fibres, but you are dealing with wet fibres. Presumably the shear modulus is going to be lowered considerably by the water that you have in the fibres, one would also anticipate that as you break the bonding between fibrils then you will get more water going in between different fibrils within the fibre. Would you like to comment on that?

*Ho Fan Jang*

We don't have to use the absolute values of the elastic modulus from the dry fibre to fit our data. What we used is the ratio of  $S^{11}/S^{66}$ , which I would expect to be similar for both dry and wet fibres.

*Raj Seth* Paprican

I would like to respond to the question that Kari Ebeling asked. As far as bleached softwood chemical pulps are concerned, their fibres are mostly collapsed. Because these fibres have seen considerable delignification and incidental mechanical treatment, the transverse elastic modulus of the fibre walls is very low; the fibres collapse readily. Thus, collapse is not an issue for most

reinforcing pulps (unless they have unusually thick-walled fibres). We have confirmed this for several commercial pulps.

*Kari Ebeling*

Paper making is a process of trying to make money, so how you refine is of key importance. Depending how you fibrillate the cell wall you get tremendous increase in the collapse or you get a little increase in the collapse and that shows in the physical properties like the bulk. The key objective is to keep the strength increase in refining and retain the other properties that tend to decrease as refining progresses?

*Raj Seth*

As I said earlier, fibre collapse is not an issue for reinforcing pulps as their fibres are mostly collapsed. However, for high-yield pulps, particularly mechanical pulps, as their fibres are lignin rich and therefore stiff, refining would promote their further collapse.

*Derek Page*

I am very puzzled by the use of twice the surface tension divided by the lumen perimeter. As far as I am concerned, surface tension forces are given by twice the surface tension divided by the radius of curvature of the water meniscus. Why would you move away from the physical theories to something that is not at all the same? Twice the inner capillary like the lumen, the collapsing force is  $2t$  divided by the radius of curvature of the lumen, but you haven't used that.

*Ho Fan Jang*

Perhaps I should clarify that the symbol  $T$  in our collapse equation is referred to fibre wall thickness, but not surface tension. The collapse force  $F$  contains the surface tension.

*Derek Page*

I don't think that it is the same.

## *Discussion*

*Ho Fan Jang*

Are you saying that I should use the lumen radius, instead of the lumen perimeter?

*Derek Page*

Yes, you should use the minimum radius that the lumen has collapsed to, at the time when the water meniscus runs down the lumen. That gives the maximum pressure that the fibre has been exposed to. On another point have you taken into account S1 and S3 thickness in any of your work. In some species, particularly southern pines they have a very thick S1 and S3 layer and being transversely wound would have a high resistance to collapse. For the first time we may be understanding why southern pines behave in the peculiar way that they do because they tend to be unusually resistant to collapse, compared to their wall thickness.

*Ho Fan Jang*

I agree. We believe that compared to the S2 layer, the effects of the S1 and S3 layers are not significant for the chemical pulp fibres that we chose for our experiments. However, for some fibres, as you just mentioned, the effects can be significant, and need to be included in the transverse elastic modulus equation,  $E^T$ . I would also like to emphasize that although the S1 layer is much thinner than the S2 layer, it could affect fibre collapse because of its high fibril angle.