

Z-DIRECTION PROPERTIES: THE EFFECTS OF YIELD AND REFINING

B. F. Berger and G. A. Baum
The Institute of Paper Chemistry, Appleton, Wisconsin, USA

ABSTRACT

The z-direction (ZD) elastic properties of paper have received little attention in the past because of measurement difficulties. This paper describes the effects of wet pressing, refining, and yield on three ZD elastic properties, C₃₃, C₄₄, and C₅₅. The elastic parameters were measured using ultrasonic methods on an unbleached kraft oak pulp. The ZD elastic parameters were very sensitive to wet pressing pressure. Increasing the level of refining or decreasing pulp yield produced increases in C₃₃, C₄₄, or C₅₅, which were greater than would be expected by wet pressing alone to the same density. A plausible explanation for this behavior is that the refining and yield changes also significantly change the ZD stiffness and shear stiffness of the fiber cell wall.

INTRODUCTION

The z-direction or thickness direction mechanical properties of paper have received much less attention than the in-plane properties because of the difficulty in measuring out-of-plane properties. Traditional measurements of elastic or strength properties of paper require that clamps or rigid platens be attached to the test specimen. In the thickness direction this usually means that adhesives must be used (except in compression) with the attendant adhesive penetration problems.

Wave propagation techniques can be used to measure the elastic properties of materials without requiring the use of adhesives. Such techniques have been adapted to paper (1-6). Because these measurements are non-destructive, it is possible to determine seven of the nine elastic parameters of paper (6) on a single specimen. This has led to a much better understanding of how machine and process variables separately and collectively affect the three-dimensional elastic response of paper. Some of the out-of-plane properties are far more

sensitive to certain process variables than are the corresponding in-plane properties. In addition, the research to date suggests that in many instances end use performance may be closely related to the (some times undetected) changes occurring in the z-direction during paper manufacture. Accordingly, the measurement of z-direction elastic properties should lead to an improved understanding of the manufacturing process and how it, in turn, relates to end use performance.

This paper briefly reviews some fundamentals and earlier work concerned with the effects of fiber orientation, wet pressing, and wet straining or drying restraints on paper z-direction elastic and strength properties, and then goes on to describe new results obtained for the effects of yield and refining.

BACKGROUND

Paper can be considered an orthotropic elastic material (2,3,6,7,8). An orthotropic material is one which has three mutually perpendicular planes of symmetry. For such a material the stresses, τ_{ij} , can be expressed in terms of the strains, ϵ_{ij} by

$$\begin{aligned} \tau_{11} &= C_{11}\epsilon_{11} + C_{12}\epsilon_{22} + C_{13}\epsilon_{33} \\ \tau_{22} &= C_{12}\epsilon_{11} + C_{22}\epsilon_{22} + C_{23}\epsilon_{33} \\ \tau_{33} &= C_{13}\epsilon_{11} + C_{23}\epsilon_{22} + C_{33}\epsilon_{33} \\ \tau_{23} &= 2C_{44}\epsilon_{23} \\ \tau_{13} &= 2C_{55}\epsilon_{13} \\ \tau_{12} &= 2C_{66}\epsilon_{12} \end{aligned}$$

The nine C_{ij} are called the elastic stiffnesses and have units of stress (Pa). Alternatively the nine stiffnesses can be written in terms of elastic compliances, S_{ij} , where $[S_{ij}][C_{ij}]=I$, or as engineering elastic constants. The latter include three Young's moduli, three shear moduli, and three Poisson ratios. While the elastic behavior of paper can be expressed in any of these three forms, the elastic stiffnesses are preferred since these may be measured directly using sound wave propagation techniques (6-9). Such techniques are valid as long as the wavelength of the sound wave is long compared to the characteristic dimensions of the fibers. In such cases the paper may be considered a homogeneous continuum.

The elastic stiffnesses C_{11} , C_{22} , C_{12} , and C_{66} are referred to as in-plane parameters since they are all defined in the MD-CD (or x-y or 1-2) plane. The elastic stiffnesses C_{33} , C_{44} , C_{55} , C_{13} , C_{23} are referred to as out-of-plane elastic parameters because they all involve the z-direction. It is this last group of "constants" (especially the first three) that is of interest to us in this paper. As will be discussed below, however, these quantities are not "constant" at all but are very sensitive to process conditions.

The appendix gives a brief description of how the engineering elastic constants are defined.

Previous work (for example, references 9-11) has shown that the in-plane and out-of-plane elastic properties of paper are very sensitive to paper machine process variables. It is well known, for example, that changes in rush-drag ratios (fiber orientation), wet pressing, or wet straining affect the in-plane Young's moduli. Less well understood is the affect of these and other variables on the out-of-plane elastic properties. Fleischman et al. (11) reported that C_{33} (related to E_{ZD}) was not sensitive to changes in fiber orientation (in the plane of the paper), but was extremely sensitive to wet pressing pressure and wet straining (wet draw) or drying restraints. Increased wet pressing pressure causes significant increases (up to tenfold) in ZD stiffness, C_{33} , and the out-of-plane shear stiffnesses. Fleischman's work also showed that the in-plane and out-of-plane elastic properties (and by implication other mechanical properties as well) are highly interrelated. That is, a change in a machine operating variable causes simultaneous changes in both in-plane and out-of-plane properties in quite predictable ways.

This is evident in the relationship that exists in paper between the shear modulus, G_{xy} , and the geometric mean of the in-plane Young's moduli (12), viz. $G_{xy} = a(E_x E_y)^{1/2}$. This expression was first deduced from considerations based on an isotropic material, for which $G = E/[2(1 + \nu)]$. This relationship also seems to hold for a number of orthotropic materials if the anisotropy is not too large and if E and ν are replaced by the geometric means (or some other suitable average) of the measured orthotropic parameters. Thus for the MD-CD plane in paper, perhaps it is not surprising that the

simple relationship holds for many (if not all) grades of paper manufactured on a Fourdrinier machine, since for these papers the anisotropy is usually less than three or so.

In reference 12 the value of a in the above expression was determined from measurements of the in-plane Poisson ratios to be 0.387 ± 0.007 (since $a^{-1} = 2(1 + (v_{xy}v_{yx})^{1/2})$). This value has since been confirmed for a large number of experimental and commercial papers in a number of laboratories. It appears to be quite insensitive to the method of paper manufacture or paper machine variables. Surprisingly, similar relationships have been found for the other two symmetry planes in the paper, even though the anisotropy in these planes is large. Specifically, for Fleischman's data (13), $C_{44} = 0.31(C_{22}C_{33})^{1/2}$ and $C_{55} = 0.25(C_{11}C_{33})^{1/2}$. The two coefficients in this case are determined from simple regressions, not from Poisson ratios, since the out-of-plane Poisson ratios are difficult to measure. The implication, however, is that shear in a given plane is related to the principal moduli in that plane.

If the three equations above are multiplied (14), one obtains $C_{11}C_{22}C_{33} = K_1C_{44}C_{55}C_{66} + K_2$. The value of K_1 would be the inverse product of the three coefficients in the three separate relationships and K_2 would be expected to be zero. This relationship, again using Fleischman's data representing samples that had different levels of fiber orientation, wet straining, and wet pressing but essentially constant basis weights, is plotted in Fig. 1. The slope of the regression line in Fig. 1 is about 41 with an intercept, K_2 , not significantly different from zero. The densities resulting from the different processing conditions varied from about 0.4 to 1 gm/cm³. A change in any one or more of the three variables defines a point along the straight line. It may be that some other process variables, which affect the properties of the fiber cell wall, might also change the slope of the line. Two such variables, yield and level of refining, have been studied and the results are presented and discussed in the following sections.

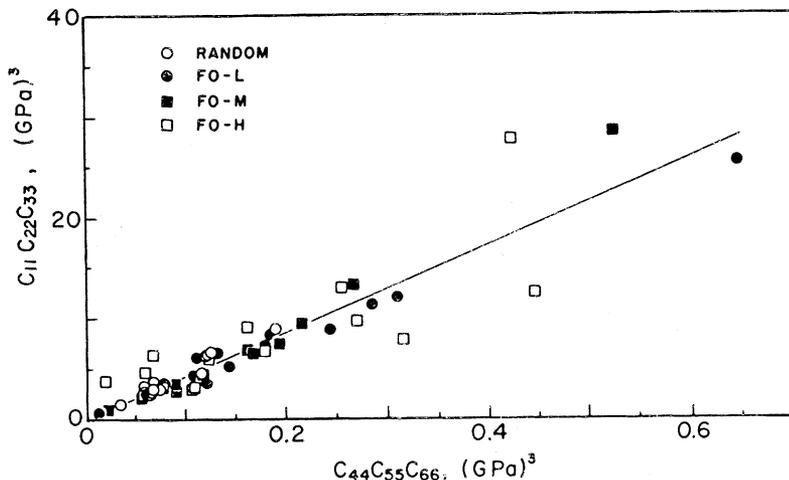


Fig 1—The product of the elastic axial stiffnesses vs. the product of the shear stiffnesses (Ref. 14).

THE EFFECTS OF YIELD AND REFINING ON ZD ELASTIC PROPERTIES

A red oak (*Quercus rubra* L.) was pulped to three yields using the kraft process by varying the cooking time and temperature (H-factor). Anisotropic sheets were prepared, using a Formette Dynamique, from each yield fraction using four refining levels in a Valley beater and four wet pressing pressures. The samples were dried under restraint in both the MD and CD. In addition, three levels of fiber orientation were used, but this work is not discussed here since the results are similar to those reported by Fleischman (11). Table 1 lists the various conditions studied.

The elastic stiffnesses for each sample were measured using ultrasonic wave propagation techniques (9). The calipers were determined using a soft rubber platen caliper gauge (15). Table 1 also presents six of the seven measured stiffnesses. (C_{12} is omitted from the table since it will not be discussed). Figure 2 shows C_{11} plotted against IPC (rubber platen) density. As expected, as the refining level is increased, the density at a given wet pressing level also increases, as does C_{11} . The

Table 1. Pulping and refining variables*.
(Last 3 digits of sample name are wet press pressure in psi.)

Name	IPC		Yield (%)	Kappa (%)	Beat time (min)	CSF (ml)	F0 (Arb.)	C11 (GPa)	C22 (GPa)	C33 (GPa)	C44 (GPa)	C55 (GPa)	C66 (GPa)
	Density (g/cm ³)	Basis Weight (g/m ²)											
1025	0.344	271	53.8	16.4	0	645	medium	4.36	2.13	0.0094	0.0193	0.0298	0.884
1050	0.431	270	53.8	16.4	0	645	"	6.1	2.94	0.0132	0.0286	0.0408	1.3
1100	0.576	270	53.8	16.4	0	645	"	8.68	4.31	0.0322	0.0524	0.0733	2.06
1250	0.582	271	53.8	16.4	0	645	"	8.5	4.03	0.0378	0.057	0.0791	1.93
2025	0.417	264	53.8	16.4	10	600	"	5.51	3.25	0.0218	0.0366	0.0517	1.3
2050	0.522	263	53.8	16.4	10	600	"	7.45	4.53	0.0344	0.0564	0.0773	1.84
2100	0.659	265	53.8	16.4	10	600	"	10	5.93	0.0729	0.0891	0.119	2.62
2250	0.658	266	53.8	16.4	10	600	"	9.93	5.87	0.075	0.0984	0.128	2.49
3025	0.658	274	53.8	16.4	35	330	"	10.4	6.1	0.137	0.14	0.172	2.84
3050	0.756	273	53.8	16.4	35	330	"	12.7	7.41	0.183	0.178	0.208	3.55
3100	0.865	275	53.8	16.4	35	330	"	14.1	9.32	0.298	0.212	0.242	4.1
3250	0.86	275	53.8	16.4	35	330	"	14.4	8.31	0.283	0.212	0.249	4.11
4025	0.733	263	53.8	16.4	50	170	"	12.9	7.36	0.223	0.186	0.215	3.53
4050	0.834	262	53.8	16.4	50	170	"	14.5	8.74	0.309	0.221	0.25	4.14
4100	0.955	273	53.8	16.4	50	170	"	16.1	9.73	0.447	0.248	0.285	4.73
4250	0.931	265	53.8	16.4	50	170	"	16.3	9.74	0.401	0.266	0.291	4.82
5025	0.593	265	56.5	31.7	35	450	"	8.82	5.06	0.0868	0.111	0.138	2.31
5050	0.696	263	56.5	31.7	35	450	"	10.7	6.97	0.129	0.143	0.172	2.93
5100	0.809	265	56.5	31.7	35	450	"	12.8	8.6	0.211	0.193	0.224	3.77
5250	0.812	268	56.5	31.7	35	450	"	12.7	8.47	0.207	0.173	0.207	3.66
6025	0.546	266	58.3	46.5	35	515	"	7.52	3.94	0.061	0.0775	0.105	1.92
6050	0.662	266	58.3	46.5	35	515	"	9.8	5.31	0.0977	0.107	0.141	2.54
6100	0.78	266	58.3	46.5	35	515	"	11.7	7.49	0.169	0.159	0.2	3.27
6250	0.778	269	58.3	46.5	35	515	"	11.5	6.77	0.172	0.151	0.195	3.17
7025	0.639	256	53.8	16.4	35	350	low	8.34	8.08	0.128	0.115	0.15	2.79
7050	0.736	260	53.8	16.4	35	350	"	9.79	9.24	0.178	0.18	0.18	3.37
7100	0.841	266	53.8	16.4	35	350	"	11.4	11.1	0.281	0.23	0.23	3.9
7250	0.827	267	53.8	16.4	35	350	"	12.1	10.4	0.266	0.207	0.207	3.89
8025	0.655	283	53.8	16.4	35	350	high	10.3	6.29	0.14	0.115	0.186	2.84
8050	0.755	286	53.8	16.4	35	350	"	12.8	7.35	0.198	0.175	0.217	3.47
8100	0.862	286	53.8	16.4	35	350	"	14.6	8.89	0.318	0.222	0.267	4.12
8250	0.845	286	53.8	16.4	35	350	"	14.4	8.88	0.289	0.217	0.259	4.12

*Red oak

effect of yield, in the narrow range studied, is not very great, although it can be seen that the higher yield samples tend to have a lower C_{11} stiffness at constant density. The out-of-plane properties, however, seem to be much more sensitive to yield and refining than the in-plane properties. Figure 3 plots C_{33} against density for the samples refined at different levels but at constant yield. The effects of refining and wet pressing on C_{33} are quite large; over the density range studied C_{33} increases by a factor of about 45. This is 4 or 5 times greater than the changes in C_{33} found by Fleischman (13) resulting from wet pressing over the same density range. At a constant density of 0.7 g/cm^3 , for example, C_{33} increases by a factor of two due to the refining. The effect of increasing yield on C_{33} is shown in Fig. 4. At constant density, increasing yield causes a decrease in ZD stiffness. At a density of 0.7 g/cm^3 the decrease is about 40%. These results could be interpreted in terms of decreased interfiber bonding at the higher yield levels. The scattering coefficients were measured for some of the samples at a wavelength of 700 nm^* . At densities near 0.7 g/m^3 the decrease in yield from 58.3 to 56.5% produced a decrease in scattering coefficient of about 5% and a decrease from 56.5 to 53.8% gave a 25% increase in scattering coefficient. Increased refining (0 to 50 minutes) decreased the scattering coefficient about 12% at the same density. From these results it appears that the observed 200% and 40% increases in Figs. 3 and 4, due to increased refining and decreased yield, respectively, cannot be explained by changes in interfiber bonding only. The results thus also suggest that decreased yield and increased refining lead to a stiffening of the cell wall in the dried sheet. Presumably this would represent increased intrafiber bonding in the cell wall.

Figures 5 and 6 show that C_{44} and C_{55} , respectively, vary with density and refining just like C_{33} . Increases in refining level on these two out-of-plane shear stiffnesses has a greater effect than just increasing density by wet pressing. The behavior with yield changes is also like that observed for the out-of-plane stiffness, as shown in Fig. 7 for C_{55} . The results for C_{44} are similar.

*The scattering coefficients of the heavy basis weight samples were measured at 700 nm in order to minimize scattering to get enough energy through the sheet to make the measurement. This should be permissible since the results are used in a comparative way.

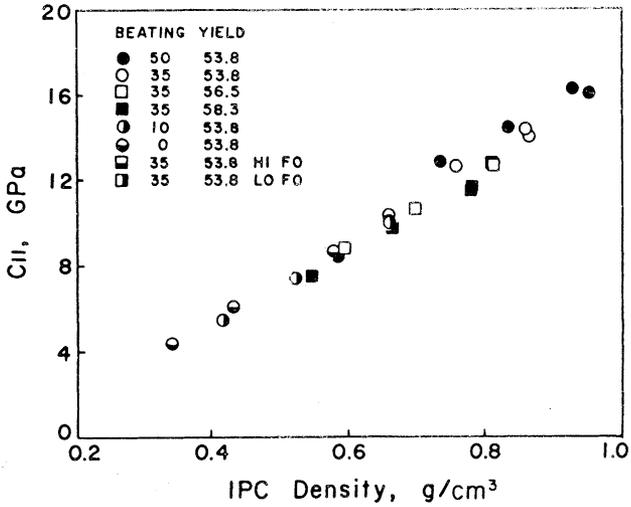


Fig 2—Elastic stiffness C_{11} vs. density at different refining and yield levels.

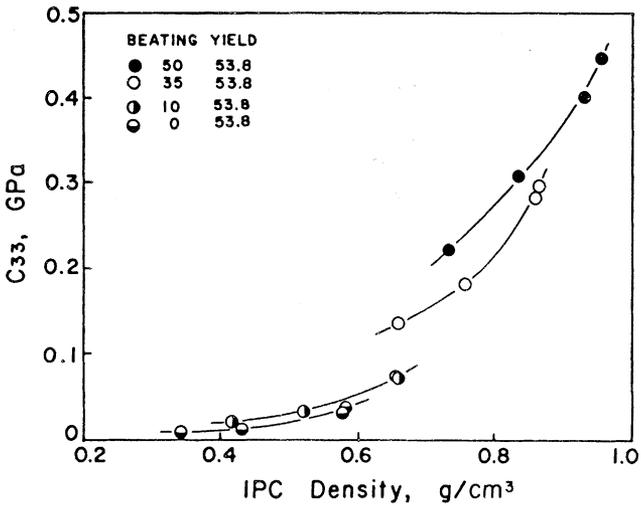


Fig 3—Elastic stiffness C_{33} vs. density at constant yield and four levels of refining.

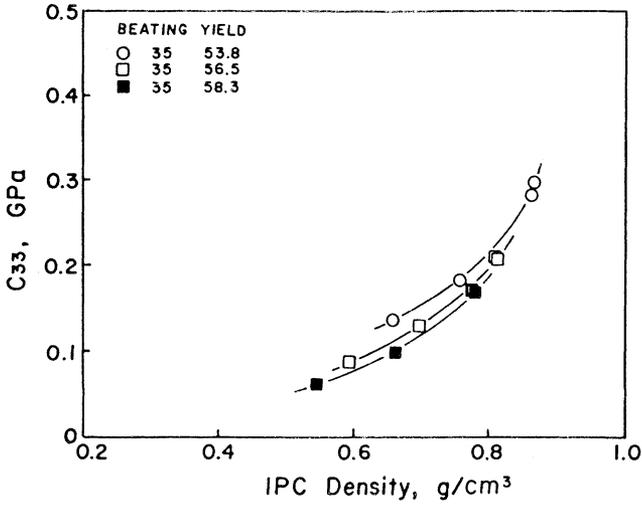


Fig 4—Elastic stiffness C_{33} vs. density at constant refining and three yield levels.

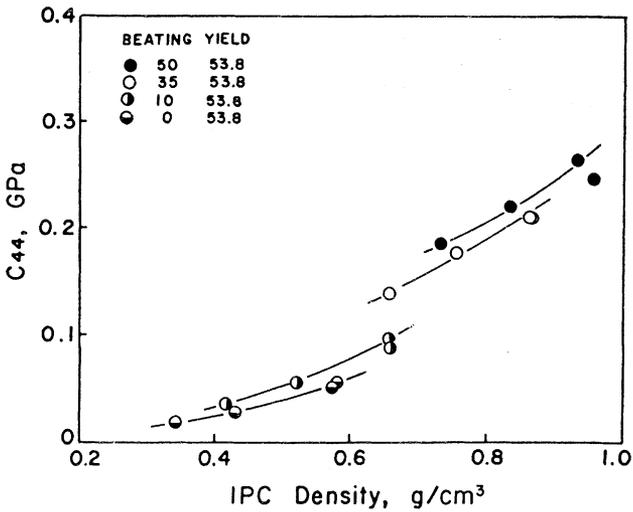


Fig 5—Elastic shear stiffness C_{44} vs. density at constant yield and four refining levels.

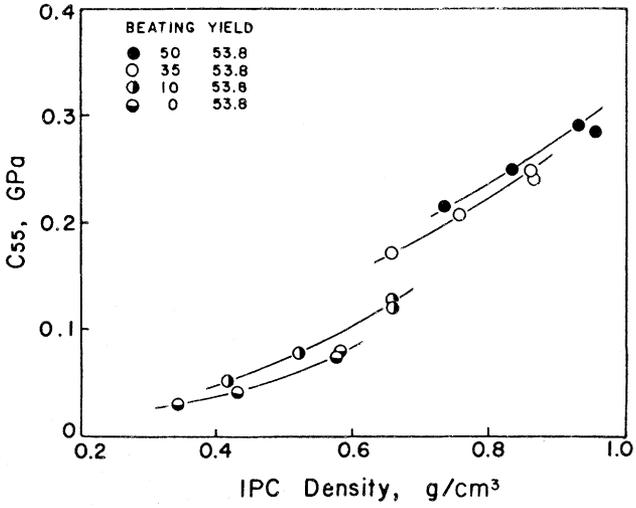


Fig 6—Elastic shear stiffness C_{33} vs. density at constant yield and four refining levels.

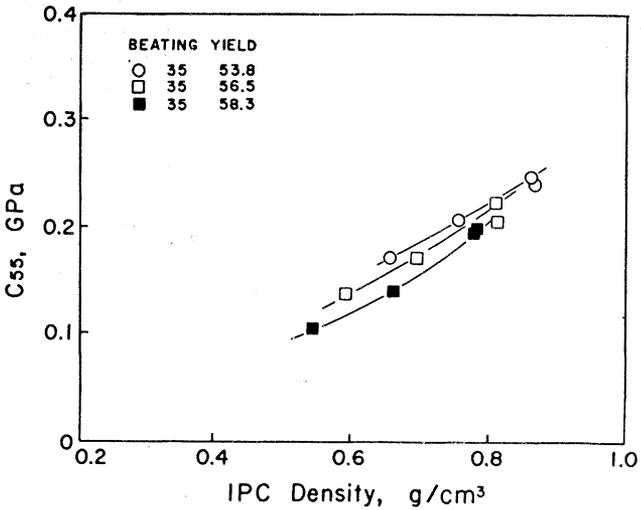


Fig 7—Elastic shear stiffness C_{55} at constant refining and three yield levels. The results for C_{44} are similar.

It appears that on a relative basis the ZD properties are much more sensitive to changes in yield or refining than the in-plane properties. If increased refining or decreased yield leads to a stiffer (less deformable) fiber cell wall in the transverse direction in the dried sheet, they would also increase the fiber cell wall shear stiffness. It would be difficult to separate the effects of increased interfiber bonding and increased intrafiber bonding, however, resulting from the changes in processing conditions.

The relationships between shear stiffness and the geometric mean of the two extensional stiffnesses in the same plane are shown in Fig. 8-10. Figure 8 is the situation for the MD-CD plane. The data form a nearly linear relationship between G_{66} and $(C_{11}C_{22})^{1/2}$ with a slope of 0.397 ∓ 0.014 . This slope should be compared with the previously mentioned value of 0.388. There does not, however, appear to be any major effects due to changes in yield or refining. Figures 9 and 10 show the relationships in the CD-ZD and MD-ZD planes, respectively. In both cases, a linear relationship exists at the lowest levels of the out-of-plane properties, but the data deviates from this behavior at the higher values. Note that in Figs. 8-10, the points which deviate from a simple linear relationship are those for the two highest refining levels and highest pressing pressures. In Fig. 8, for the MD-CD plane, the tendency seems to be toward slightly greater slope (points above the line) while in the CD-ZD plane (Fig. 9) and MD-ZD plane (Fig. 10) the data points fall beneath the line.

In some respects the results in Fig. 9 and 10 are surprising because the original argument which leads to a relationship between shear stiffness and the geometric mean of the in-plane Young's moduli assumes low anisotropy as discussed above. In the CD-ZD and MD-ZD planes in paper, however, the anisotropy ratios can easily be greater than 100 and are very sensitive to wet pressing and wet straining conditions (16). Hence, one could not anticipate relationships like those shown in Figs. 9 and 10.

In the earlier paper dealing with the relationship between shear and extensional stiffnesses in the MD-CD plane (12), it was observed that the relationship seems to hold until the anisotropy ratio R_{12} ($=C_{11}/C_{22}$) became greater than about 3.5. A

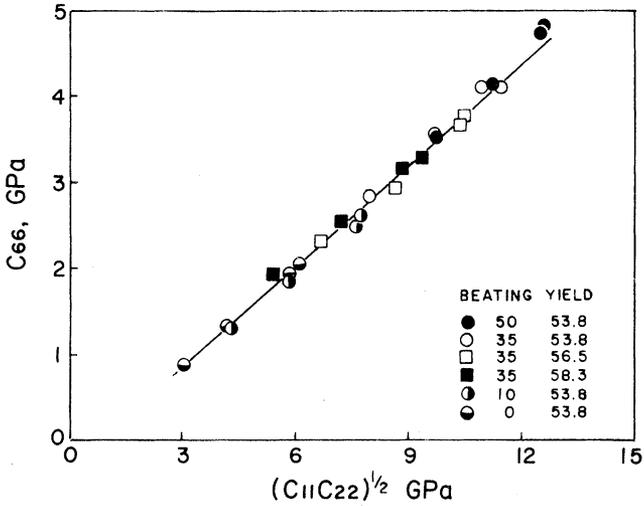


Fig 8—Elastic shear stiffness C_{66} plotted against the geometric mean of the in-plane elastic stiffnesses.

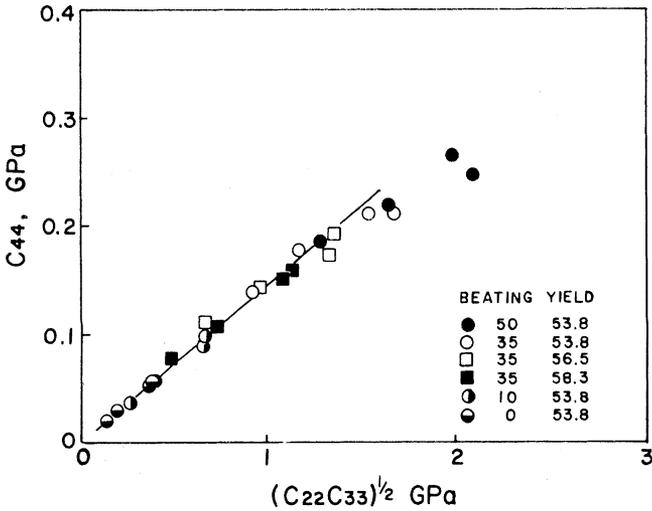


Fig 9—The elastic shear stiffness C_{44} plotted against the geometric mean of the elastic stiffnesses in the CD-ZD plane.

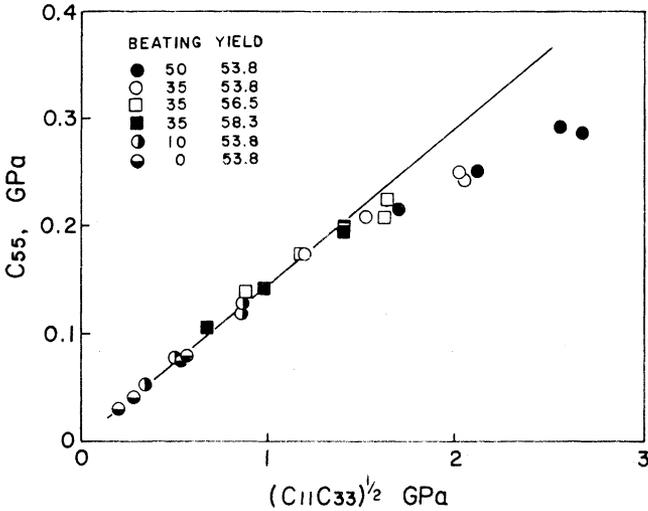


Fig 10—The elastic shear stiffness C_{55} plotted against the geometric mean of the elastic stiffnesses in the MD-ZD plane.

plot of the ratio of $C_{66}/(C_{11}C_{22})^{1/2}$ versus anisotropy for this study is shown in Fig. 11, but the range of R_{12} is so small that it is difficult to draw any conclusions concerning the coefficient. The results in Fig. 11 do imply, however, that one can expect greater anisotropy due to fiber orientation or/and drying restraint effects for unbeaten and/or high yield pulps. That is, the poorly bonded pulps in Fig. 11 seem to have greater values of R_{12} . The situation in the other two planes is depicted in Figs. 12 and 13. For either case the ratio is constant at the highest anisotropies, but decreases with decreasing anisotropy below about 60 in the CD-ZD plane and 110 in the MD-ZD plane. The significance of this is uncertain, but it would seem that the ratios should approach the in-plane value of 0.3 to 0.4 as the MD-ZD or CD-ZD anisotropies decreased toward lower numbers typical of the MD-CD plane. If so, a minimum must occur in the curves of Figs. 12 and 13 at an anisotropy between 1 and 25.

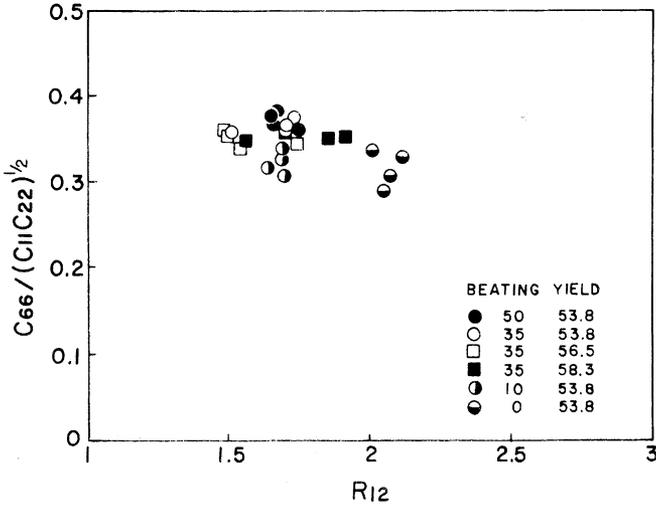


Fig 11—The ratio $C_{66}/(C_{11}C_{22})^{1/2}$ plotted against the anisotropy ratio $R_{12}(=C_{11}/C_{22})$

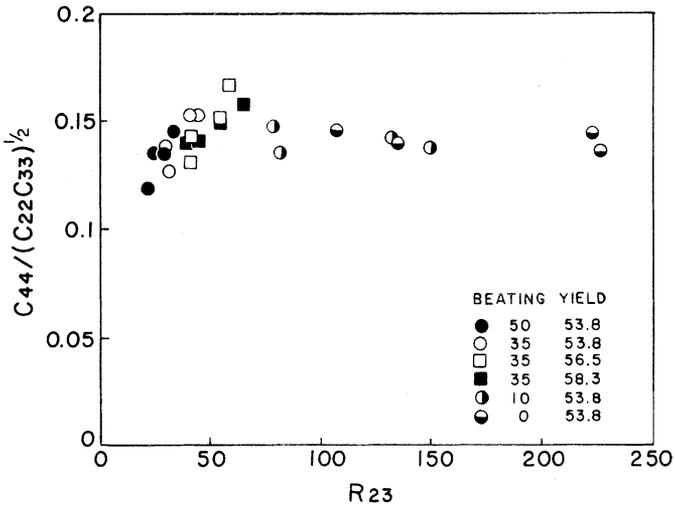


Fig 12—The ratio $C_{44}/(C_{22}C_{33})^{1/2}$ plotted against the anisotropy ratio in the CD-ZD plane $R_{23}(=C_{22}/C_{33})$

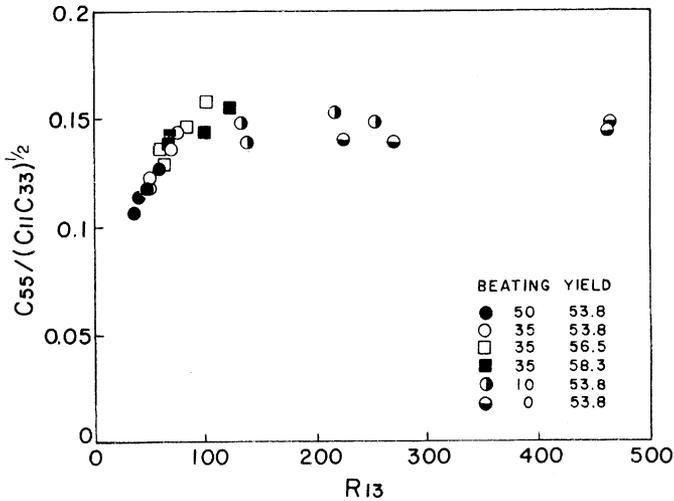


Fig 13—The ratio $C_{55}/(C_{11}C_{33})^{1/2}$ plotted against the anisotropy ratio in the MD-ZD plane $R_{13} (= C_{11}/C_{33})$

The product of the three extensional stiffnesses plotted against the product of the three shear stiffnesses is shown in Fig. 14. There is a definite upward curvature in the data. The quantities plotted in Fig. 14 are interchanged from those in Figs. 8-10, so that the curvature in Fig. 14 is actually consistent with the trends shown in Figs. 9 and 10. Note also that two additional data sets are included in Fig. 14, representing two different levels of fiber orientation (random and high) at the lowest yield level and 35 minute refining level (see Table 1). These additional sets show that data resulting from changes in fiber orientation still fall along the curve. The curve is best fit with a power law relationship, $C_{11}C_{22}C_{33} = 161.1 (C_{44}C_{55}C_{66})^{1.024}$, where $R = 0.996$. For Fleischman's data in Fig. 1 the power law relationship is $C_{11}C_{22}C_{33} = 47.3 (C_{44}C_{55}C_{66})^{1.061}$ with $R = 0.971$.

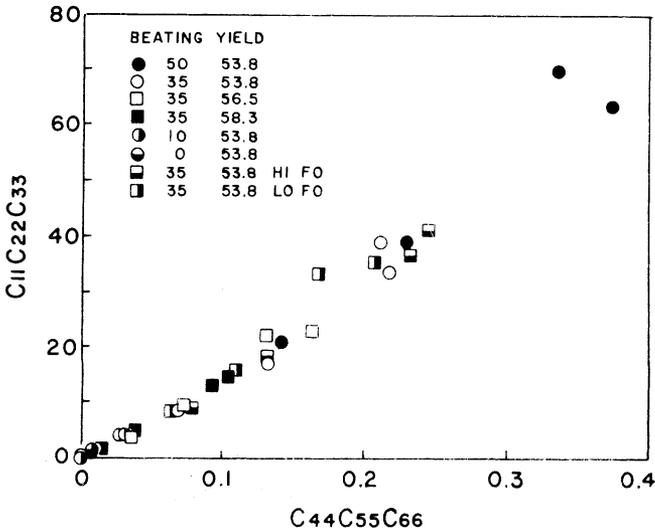


Fig 14—The product $C_{11}C_{22}C_{33}$ plotted against the product of the shear stiffnesses $C_{44}C_{55}C_{66}$. Compare to Fig 1.

DISCUSSION AND CONCLUSIONS

The results shown in Figs. 3-7 clearly show the large effects in ZD properties caused by changes in yield, refining, and wet pressing. The changes with yield and refining are greater than would be expected from densification caused by wet pressing alone. Thus it appears that changes in the cell wall brought about by increased refining or decreased yield (lignin and hemicellulose removal) may directly impact the measured ZD properties of the paper. This would be consistent with the work of Seth and Page (17) who discovered that the stress-strain behavior of paper is directly related to the stress-strain behavior of the fiber cell wall for well bonded sheets. There should be quite a difference, of course, between straining a sheet in the ZD compared to the MD-CD plane.

Figure 15 attempts to illustrate these differences. The upper figure (a) depicts an unstrained cross-section of paper. Figure 15b shows the situation for a uniaxial strain in the MD

or CD. Some fibers are primarily strained along their axis (deformed in their width-axis (W-A) and thickness-axis (T-A) planes), while others experience only shear strains in their width-thickness (W-T) plane or axis-thickness (A-T) plane. Since the collapsed fibers tend to lie in the MD-CD plane, the interfiber bonds, are primarily stressed in a shear mode in the MD-CD plane. Figure 15c shows the case for ZD straining. Here the fibers predominantly experience transverse strains, but some deformation in the fiber W-A plane must also occur. The fiber-fiber bonds in this case are primarily strained in the ZD, in contrast to the situation in Fig. 15b. Figure 15d depicts shear in the MD-ZD or CD-ZD plane. This case would seem to be a sort of a combination of Figs. 15b and 15c, since all of the types of stresses must exist in most of in the fibers and bonds.

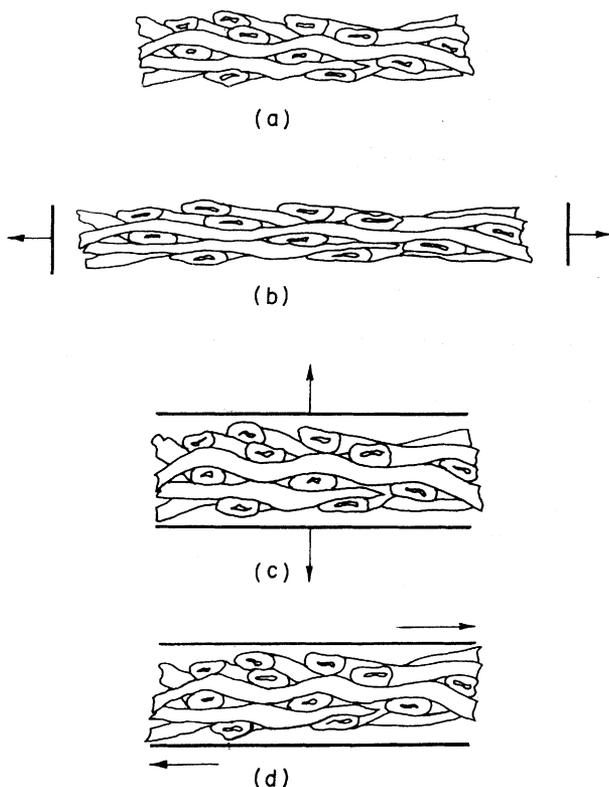


Fig 15—Modes of deformation in the MD-ZD or CD-ZD plane. (a) undeformed state; (b) MD or CD straining, (c) ZD straining, (d) shear strain in the MD-ZD or CD-ZD plane.

The representation above with respect to the bonds probably is too simple, since the microcompressions resulting from drying must impart a three-dimensional character to the fiber-fiber bonds. This 3D character would likely produce a fiber-fiber bond shear strength (in the MD-CD plane) greater than that expected for two smooth surfaces bonded together and placed in shear. The same argument may apply in the case of ZD straining. It is known, for example, that wet straining or drying restraints dramatically reduce C_{33} or ZD tensile strength (ZDT). Since these process variables would also reduce the occurrence of microcompressions, perhaps it is the latter which contribute to the greater ZD stiffness and strength for unstrained sheets or sheets dried with no restraint. That is, perhaps microcompressions lead to greater ZD stiffnesses and strengths because of the three dimensional character they impart to the bond.

In the case of ZD properties, however, it should be realized that the transverse fiber properties are the ones that are important, as Fig. 15c and 15d above try to illustrate. Since most fibers in paper lie in the (MD-CD) plane of the paper, it is the ZD properties of the fibers themselves that influence the ZD sheet properties. There are two things to look at with respect to fiber ZD properties. First there is the question of lumen collapse, directly related to the conformability of the fiber which in turn is affected by the pulp yield and level of refining (among other things). The more refining and the lower the lignin content the more conformable the fiber becomes in the wet state and the greater the likelihood it collapses under "normal" pressing pressures or surface tension forces. Once the lumen has collapsed it must stay collapsed if high ZD stiffness (or strength) is required in the paper product. This means that (hydrogen) bonds must form across the collapsed lumen, otherwise it would open up under ZD loading and act as a low modulus region (18). One would expect lignin removal and internal fibrillation, caused by extensive beating, would contribute greatly to this ZD stiffness in the dried fiber.

The other factor which would contribute to fiber ZD properties is the presence or lack of the lignin and hemicellulose "matrix" in the secondary cell wall. A wood pulp fiber is an excellent example of a "fiber reinforced composite" in which

the fibrils in the cell wall are the "fiber" in the composite. The lignin and hemicelluloses are the matrix material of the composite. When the lignin is removed by pulping and the fibrils can bond to each other, (perhaps in conjunction with the hemicelluloses), a much more homogeneous and stiffer structure results in the dried fiber. This "stiffer" fiber in the ZD then leads to a stiffer and stronger paper in the ZD. The removal of both lignin and hemicellulose may produce a situation where "fibril to fibril" bonds cannot readily occur, thereby weakening the fiber cell wall. It has been shown that lignin and hemicellulose removal does decrease fiber-fiber bond strength (19). It would seem likely that without sufficient matrix material to distribute the loads between fibrils, the cell wall is more apt to fail in a brittle fracture mode during ZD straining.

While the above arguments concerning the effects of yield and refining on the ZD elastic properties of paper and fibers seem reasonable and agree with the data, they are, of course, only conjecture. Only a few studies have been carried out examining fiber ZD properties and the nature of fiber-fiber bonds strained in a normal (to the bond surface area) direction (e.g. Refs. 18 and 20). Perhaps this is an area which deserves more attention.

The results presented in Figs. 8 through 14, while not completely understood, have some practical applications. For example, since the shear modulus in any plane is related to the geometric mean of the Young's moduli in that plane, process variables like fiber orientation or wet straining, which change anisotropy do not affect the shear modulus if the anisotropy ratio is less than three or so. One might use the shear stiffness values when studying furnish changes or wet pressing changes since the shear values would be insensitive to other changes which might be occurring simultaneously (e.g. fiber orientation or wet straining). This scheme has already been used in the case of on-machine measurements to help separate changes in the furnish from changes in machine operating variables for the eventual purpose of papermachine control (21). For those who model containers or other structures, the simple relationships between shear and extensional stiffnesses can often make the model simpler or minimize the amount of data gathering (or guessing) necessary. A value for the in-plane

shear modulus, for example, which would be needed in most box or tube models is not easily measured, but can be estimated from the Young's moduli which can be easily measured.

The results suggest that in many cases fewer than nine elastic parameters will be required to obtain a good description of the mechanical response of paper and how it is impacted by changes in process variables. It is important to note, however, that one or more z-direction parameters must be included in the description. We believe that a more thorough look at z-direction properties, and how they are influenced by machine variables, will be a fruitful path to follow.

APPENDIX: ENGINEERING ELASTIC CONSTANTS

The meaning of the engineering elastic parameters can be understood by referring to Figs. 16 to 18. Figure 16 defines the three principal directions. The machine direction is referred to as MD (or x or 1), the cross direction as CD (or y or 2), and the thickness direction as ZD (or z or 3). If we apply a uniaxial stress to the sample in any one of these three directions we could deform the small element in one of the modes shown in Fig. 17. The ratio of the applied stress to the resultant strain (at small strains) is defined as the elastic modulus or Young's modulus in the direction of straining. The three modes of deformation shown thus result in three Young's moduli: E_{md} , E_{cd} , and E_{zd} . In addition, for any of the three modes shown in Fig. 17, the Poisson ratio would be defined as the ratio of the lateral contraction to the axial extension in the direction of straining. For the upper left hand figure, for example, two Poisson ratios could be defined since the specimen contracts in both the CD and ZD. These would be referred to as ν_{cd-md} and ν_{zd-md} , or ν_{yx} and ν_{zx} , respectively. The three modes of deformation shown in Fig. 17 thus yield six Poisson ratios, but only three of these are independent. These are normally taken to be ν_{xy} , ν_{xz} , and ν_{yz} . The elastic stiffnesses, C_{ij} , are related to the Young's moduli and Poisson ratios. For example $C_{11} = E_{md}/(1-\nu_{xy}\nu_{yx})$, etc. Figure 18 shows three modes of shear deformation, where the applied stresses are parallel to one of the principal directions. In these cases, a push or pull on opposite sides (or faces) of the specimen results in a shear deformation. The three independent shear stiffnesses shown correspond to each of the three planes of symmetry. The elastic parameters E_{md} , E_{cd} , G_{xy} (G_{md-cd}),

and ν_{xy} (ν_{md-cd}) are referred to as in-plane elastic constants because they are all defined in the MD-CD plane, whereas the parameters E_{zd} , G_{xz} , G_{yz} , ν_{xz} , and ν_{yz} are called out-of-plane elastic constants because they all involve the z-direction.

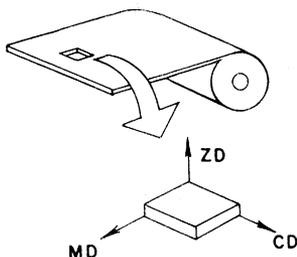


Fig 16—Principal directions assigned to paper.

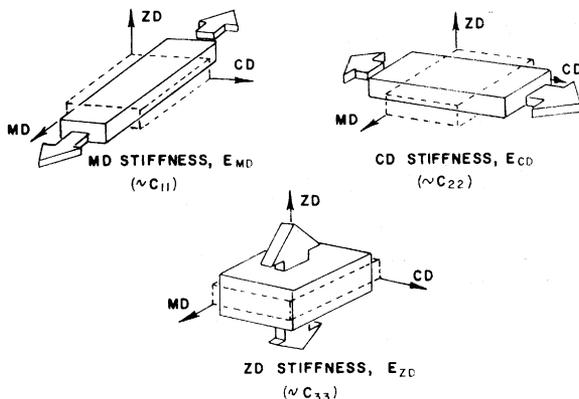


Fig 17—Three modes of deformation in uniaxial tension.

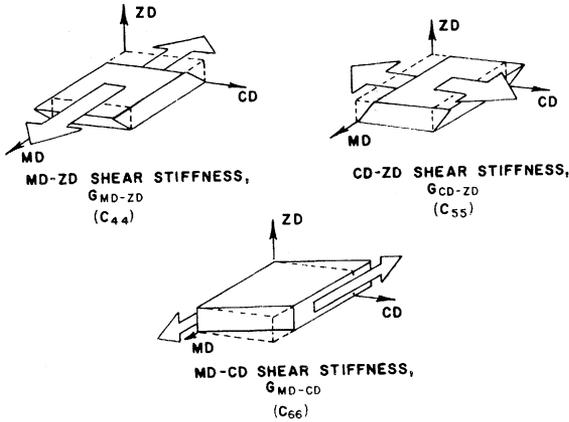


Fig 18—Three modes of deformation in shear.

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Transcription of Discussion

SESSION 2 FIBRES continued

Chairman D.H. Page

Z-DIRECTION Properties: The Effects of Yield and Refining

by B.F. Berger and G.A. Baum

Prof K.J. Ebeling Helsinki University of Technology,
Finland.

Is there perhaps a better explanation to the observed result of E_{ZD} increasing with refining than the increase in the shear stiffness of the cell wall on the closing of the lumen due to collapse of the cell wall. If you plot the scattering co-efficient against sheet density, the increased wet pressing gives a small decrease in light scattering, whereas refining gives a large decrease. As the bonded area is increased much more with refining and as the transverse shrinkage potential of the cell wall becomes more pronounced with refining, one would expect a more pronounced 'activity', i.e. readiness to carry load after refining in comparison to wet pressing. When one takes into consideration that the cell wall is more swollen after refining, it would seem to me that the transverse shrinkage induced activation of the cell wall material is also very important in addition to collapse in explaining the results you observed. In other words, a lateral phenomena of shrinkage forces contributes to Z-directional increase of "activity".

Prof G.A. Baum Thank you for your comment. It seems to me that swelling of the fibre due to refining promotes the local rearrangement of fibrils or layers of fibrils. If the rearrangement of fibrils (due to surface tension forces) results in more well bonded (connected) network of fibrils after drying, the result is an increase in the fibre cell wall transverse and shear stiffnesses. I believe that is your point. Aren't we saying the same thing?

Ebeling Yes, I suppose we are.

Chairman

I certainly like the idea of fibre collapse because the range of beating times and yields over which you were working, is exactly the range of yields over which collapse is changing rapidly for a Kraft pulp.

S.U. Hossain Kimberly Clark Corp., Neenah, U.S.A.

If you do your refining in the presence of a small surfactant molecule, that is if you can get it in between the lamellae, would you reduce the stiffening? Would this be a way of making a naturally stiff fibre less stiff?

Baum If what you suggest does happen, our measurements may be a method of quantifying it. But that hasn't really been explored. The measurements are sensitive to anything that you do to the paper in the way of changing process variables or fibre properties.

Dr. J. Parker Bowater Tech. Services, Gravesend,
England

Are your relationships obtained on uncalendered paper or on handsheets and secondly, do you know what effect calendering would have on the relationships that you describe?

Baum Yes to both questions. We have looked at handsheets, commercial papers, and laboratory sheets made on an anisotropic sheet former, so we know that the relationships described here do hold for a variety of papers, including commercial grades. We have just started work on calendering, so I will have to report on that at a later time. To date, all the results have been on uncalendered sheets, at least the laboratory sheets.

Dr. R.H. Marchessault Xerox Research, Mississauga, Canada

The measurement of crystal preferred orientation can be quantified using X-Ray diffraction. Beating or refining would have a pronounced influence on making the C-axis of the cellulose unit cell reside preferentially "in plane". Have you monitored this and tried to correlate your data with this preferred crystal orientation?

Baum We have not made any attempt to look at Z-direction properties in terms of fibre crystal orientation. I should qualify my earlier comment. Fibre orientation in the plane of the paper is not a factor in Z-direction properties. You can change jet/wire speed ratios, change fibre orientation in the plane and it does not affect the out-of-plane properties. Things affecting out-of-plane properties are primarily refining, wet pressing and wet draws or shrinkage during drying. In any case, the out-of-plane properties are important in operations such as bending, folding, edge compression and internal bond strength. You need to learn to control the out-of-plane properties on the paper machine. Z-direction fibre orientation is something which is difficult to study because we don't know how to measure fibre orientation in the Z-direction.

Parker You can't measure fibre orientation, but perhaps you could measure crystal orientation.

Baum Presumably, we could measure crystal orientation using X-Ray techniques. We have looked at dielectric properties of sheets. These are largely related to the orientation of the fibres or fibrils in the fibre cell wall. This was the work of Fleischman, reported at the Cambridge Symposium 1981. He showed that wet straining or stretching of the wet web did not cause any increase in fibre orientation in the direction of straining. The effects observed were apparently due to better distribution of stresses and not the alignment of the fibres in the plane. It is possible the out-of-plane fibre orientation was changed.

B. Radvan Wiggins Teape R & D Ltd., Beaconsfield, England

Would measuring the Poisson ratio in the thickness direction be difficult? Isn't it an easier property to measure?

Baum Your question relates to one of the three constants that I have not talked about, a Poisson ratio out-of-plane. I don't agree that it is easily measured. If you take dry paper and stretch it enough, it actually gets thicker. The same thing happens for wet paper in an open draw. That is not a Poisson effect at all but a large strain phenomenon which is outside of elasticity theory. It says that an extension of one unit, for example, produces a contraction of more than one unit. This sort of behaviour might be explained by a "picket fence" type model. We haven't actually done much work in this area, however. We have measured out-of-plane Poisson ratios using wave propagation techniques. These were also reported at the 1981 Symposium.

Prof J. Marton Westvaco, Laurel, U.S.A.

You said that the Z-direction properties are invariate and gave an example that the jet/wire ratio does not change modulus for example. I don't agree completely with this statement. Radvan reported that the fibres buckle if the stack meshes and the layers intermingle somewhat, which means that there might be a change in Z-direction strength. When you alter the fibre alignment on the wire, they may intermingle quite a lot, so it is not an ideal case at all.

Baum If you are talking about an increase or change in Z-direction fibre orientation, then you would expect the out-of-plane properties to be strongly affected. My point was only for changes in fibre orientation in the plane. Such changes in the plane do not affect the elastic properties in the Z-direction direction, at least in our experiments.

Prof H. Kropholler UMIST, Manchester, England

Referring to B. Radvan's question, would you care to comment about Poisson ratios and Z-direction stresses? Are there any formulae or published results?

Baum If you want figures for elastic properties, we have published a number of those in the past, so for a given stress, you can calculate the strains in the paper. In fact, there is a table in the 1981 Cambridge Symposium proceedings, which lists elastic parameters for two or three paper materials. You can define six Poisson ratios for paper, but only three are independent. A Poisson ratio is defined as the ratio of the lateral contraction to the axial extension. For example, you could apply the stress in the Z-direction and look at the contraction in the X-direction and define a Poisson ratio ν_{XZ} or you could apply the stress in the Z-direction and measure the contraction in X-direction to obtain ν_{ZX} . They are not the same number, but they are related through the anisotropy in that plane. One is extremely small and one is very large. In fact, one of them is greater than one which at first is hard to imagine.