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THE STRESS STRAIN CURVE OF PAPER

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Abstract

The explanation of the in-plane tensile stress-strain curve of paper has long been a matter for debate. In an earlier study it was shown that the elastic modulus of paper is given by an equation $E_p = a\varphi E_f$, where a is a function of the orientation distribution of the fibres in the sheet, φ describes the efficiency of stress transfer between them, and E_f is the elastic modulus of the fibres. As a result of extensive work on the effect of various paper-making treatments on the stress-strain response of paper, we have now shown that the plastic regime can be described in a similar manner, that is to say, in terms of the visco-elastic properties of the fibres, the orientation factor, and the efficiency factor. It is concluded that the non-linear behaviour of the stress-strain curve of paper originates primarily from the properties of the component fibres and not from the sheet structure.

Introduction

Paper responds to strain in a manner shown in figure 1. For small strains it is elastic. At a certain stress it yields and the post-yield response is often approximately linear. If the stress is returned to zero after the yield point the sheet displays delayed elastic recovery and permanent set. Paper also exhibits other visco-elastic behaviour. For example it shows stress relaxation if held at constant strain, and creep at constant stress. The pulping, stock preparation and paper-making variables, as well as test variables such as temperature, relative humidity, and strain rate affect the shape of the stress-strain curve and the magnitude of its parameters. What are the mechanisms responsible for the various features of the stress-strain curve? Our contribution begins with a discussion of the factors that affect the elastic modulus of paper, and is followed by an extension of these ideas to the plastic regime.

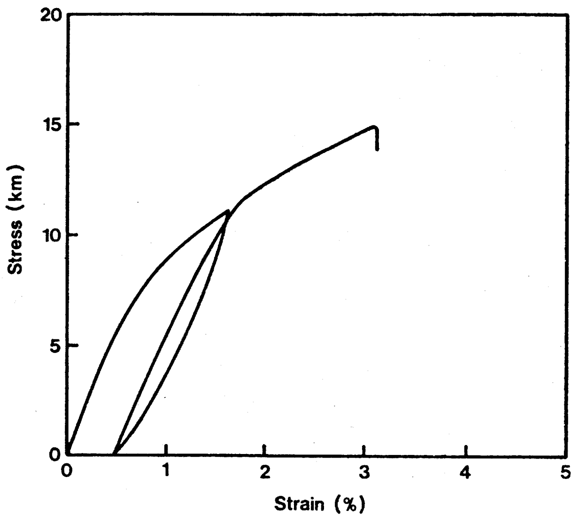

Fig 1—Stress strain curve of paper.

The Elastic Regime

Numerous authors have dealt with the problem of the elastic modulus of paper[1-17]. Mathematical equations have been derived from theoretical consideration of the deformation of a bonded random fibrous network. Generally these equations have not been tested adequately by experimental work and it is not clear that the assumptions on which the theories are based are valid.

In a recent extensive experimental study of the elastic modulus of paper(18-20) we have been able to reveal the paper-making factors that control modulus and elucidate the mechanisms by which they operate. Using the relevant mathematical analysis of Cox(1) and incorporating modern knowledge of sheet structure we have derived an equation for the elatic modulus of paper that fits all our data with remarkable precision. The equation is:

$$E_p = \frac{1}{3} E_f \left[1 - \frac{w}{L\ RBA}\sqrt{\frac{E_f}{2G_f}}\ \tanh\left(\frac{L\ RBA}{w}\sqrt{\frac{2G_f}{E_f}}\right)\right] \quad (1)$$

where E_p is the elastic modulus of paper
E_f is the axial elastic modulus of the component fibres
w is the mean fibre width
L is the arithmetic mean fibre length.
RBA is the relative bonded area of the sheet
G_f is the shear modulus of the component fibres for shear in the (L,w) plane.

The relative bonded area is calculated from the equation

$$RBA = (S_o - S)/S_o \quad (2)$$

where S is the optical scattering coefficient of the sheet
S_o is the optical scattering coefficient of the fibres in the unbonded state.

The derivation of the equation is given elsewhere(19): we will comment here on its meaning.

It is clear that the elastic modulus of paper depends on two quite separate factors. Firstly, it depends on the elastic properties of the fibrous material: secondly, it depends on the sheet structure. The structural factors arise as follows:

(a) <u>The factor of "one-third"</u>

This comes from a consideration of the stresses and strains in a homogeneous two-dimensional sheet of infinitely long fibres. The factor depends on the fibre orientation distribution, and may be calculated from it, if it is known. For a random orientation it is one-third.

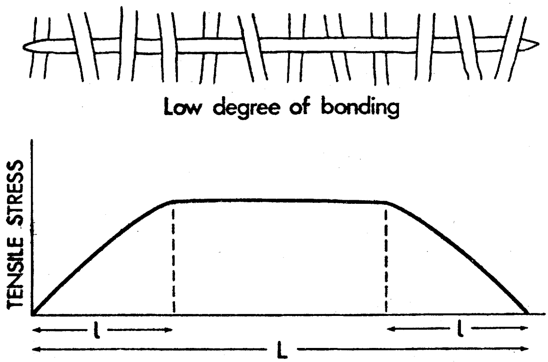

Fig 2—Tensile stress distribution along the length L of a fibre embedded in a matrix of other fibres. The stress transfer occurs over the length ℓ which depends of the degree of bonding; the sheet modulus falls short of the maximum by an amount that depends on the ratio ℓ/L.

(b) <u>The factor in brackets</u>

This describes the efficiency of the stress distribution between the fibres. Fibres in a paper sheet under strain have a distribution of stress along their length as shown in figure 2.

The stress at each fibre end must be zero and increase along the fibre length, being built up by the shear stresses in the adjacent fibres. The fibre is utilised more efficiently in a well-bonded sheet as shown in figure 3 since the stress is built up more rapidly. Similarly, each fibre is utilised more efficiently if it is long, rather than short. (It should be noted that an abrupt kink in a fibre behaves as a fibre end, since no load can be transmitted across it; the quantity L in equation (1) must therefore be considered as an effective fibre length for such fibres.)

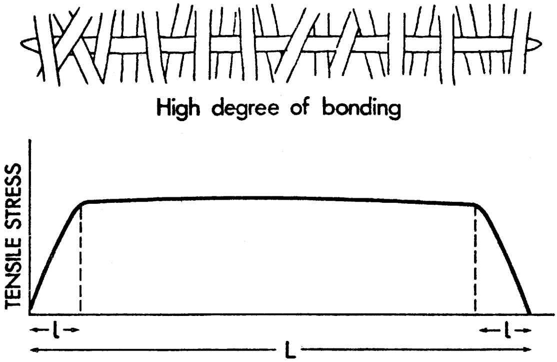

Fig 3—Stress distribution along the length of a fibre at high degree of bonding.

Equation (1) makes certain predictions that have been verified:

1. If the bonded area in a sheet is increased by a process that does not affect the elastic properties of the dry fibrous material, then the modulus of the sheet should rise with increasing bonding and reach a plateau. It is considered that increasing the wet-pressing pressure increases bonding without changing fibre properties and this has been used to test the prediction. Extensive data have been obtained, some of which are shown in figure 4. Curves which are least-square fits of Equation (1) demonstrate the validity of the equation.

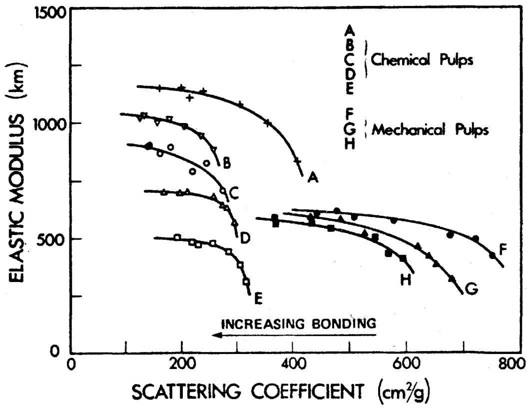

Fig 4—Data on elastic modulus and scattering coefficient for handsheets made from different pulps. The lines are least-square fits to Equation (1) allowing E_f, S_o and $(w/L)(E_f/2G_f)^{1/2}$ to float. The various pulps are: A. Laboratory-made, never-dried, unbleached, 49%-yield kraft pulp of black spruce, unbeaten. B. Laboratory-made, never-dried, unbleached, 65%-yield sulphite pulp of black spruce, Valley beaten to 600 ml CSF. C. Same pulp as B, unbeaten. D. Commercial, dried, bleached kraft pulp of Douglas fir, Valley beaten for 10 minutes. E. Commercial, dried, bleached kraft pulp of southern U.S. pine, unbeaten. F. Commercial stone groundwood of black spruce, 50 ml CSF. G. Laboratory-made refiner mechanical pulp of black spruce, 100 ml CSF. H. Commercial thermomechanical pulp of black spruce, 100 ml CSF.

2. If the fibre length is changed by a process that does not affect the elastic properties of the dry fibrous material or the relative bonded area, then the modulus should rise with increasing fibre length according to Equation (1). Figure 5 shows the data for sheets of differing fibre length and bonded area. Fibres of different lengths were prepared from the same original pulp sample by guillotining wet sheets. Handsheets of different bonded areas were obtained by changing the wet-pressing pressure. The various solid lines are simultaneous theoretical fits to Equation (1), again supporting the theory.

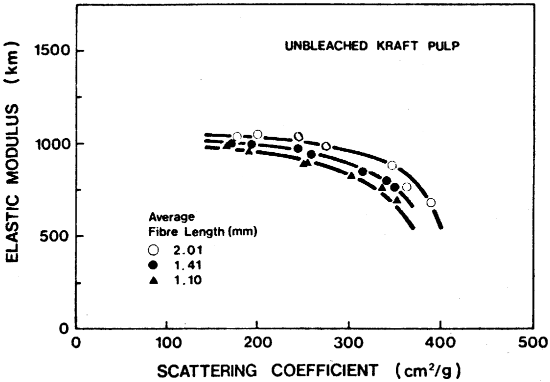

Fig 5—Data on elastic modulus and scattering coefficient for handsheets made from pulps of three different fibre-length distributions but having the same other fibre properties. The pulps were prepared from the same original pulp by guilloting wet handsheets. The pulp was a never-dried, laboratory-made, unbleached, 46%-yield kraft pulp of black spruce. The lines are fits to Equation (1) with E_f, $W(E_f/2G_f)^{1/2}$ and S_o kept the same.

3. If the fibres are long and the relative bonded area high, the elastic modulus of the paper should approach one-third that of the fibres. A plot of the sheet modulus against one-third of the measured fibre modulus for highly bonded sheets of long fibres is shown in figure 6. Again the agreement is impressive.

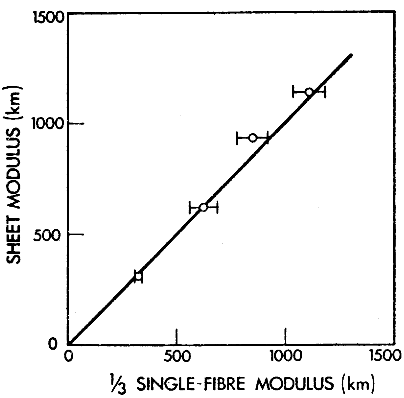

Fig 6—Plot of one-third of fibre modulus against maximum sheet modulus for four pulps in which the fibres were straight and free from gross kinks and crimps. They are: i. An unbeaten, unbleached, never-dried, laboratory-made, 47%-yield kraft pulp of black spruce. ii. An unbeaten, unbleached, never-dried, laboratory-made, 65%-yield sulphite pulp of black spruce. iii. A commercial thermo-mechanical pulp of black spruce. iv. A commercial sample of polyester fibres.

In summary, the elastic modulus of random sheets may be described by the following equation:

$$E_p = 1/3\, \varphi\, E_f \tag{3}$$

where φ is a factor, defined in Equation (1), that describes the efficiency of stress transfer in the fibres in a sheet. φ has a maximum value of 1.0 which it approaches for sheets of long, well-bonded fibres.

The Plastic Regime

The literature describing behaviour in the plastic regime contains two diametrically opposite viewpoints.

The first, the structuralist viewpoint, is most clearly stated in a 1956 paper by Rance(21). He stated that when paper is strained "...from the earliest phase of elastic bahaviour, through the apparent phases of visco-elastic flow and plastic yield, through strain hardening to ultimate fracture.... the whole sequence is visualised as elastic straining accompanied by progressive disruption of fibre to fibre bonds". In this view, the fibres are perfectly elastic and the breakage of fibre-fibre bonds causes the visco-elastic behaviour.

The second, the fibrous viewpoint, is well illustrated by the following quotation from Ebeling(22). "....the plastic region in the load/elongation curve <u>is not caused</u> by the breakage of fibre-fibre bonds, but is connected with the significant irreversible intra-fibre deformation at the molecular and supramolecular level of the cell wall structure".

Each view has its adherents. Rance(22), Nordman(23), Kallmes and Perez(10), Perez(12) and Craven(24,25) have tended to support the paper structuralist view while Page(26,27), Brezinski(28), Kubát(29), Giertz(11) and Ebeling(22) have supported the fibrous view. The matter has been discussed at length by Ebeling(30). There is however no hard evidence in the literature that unequivocally substantiates one viewpoint and discredits the other. Nor, if both mechanisms contribute, is there any evidence to indicate the proportion of each mechanism that operates under specified experimental conditions.

However, two important pieces of evidence have been obtained since the formulation of the paper structuralist viewpoint that reduce its persuasiveness.

The work of Page, Tydeman and Hunt(34) showed that the change in the optical scattering coefficient upon straining, observed by Nordman(32), is caused largely by the partial breakage of fibre-fibre contact areas. Some contact areas break completely during straining in the plastic regime, but they represent only a few percent of the total number.

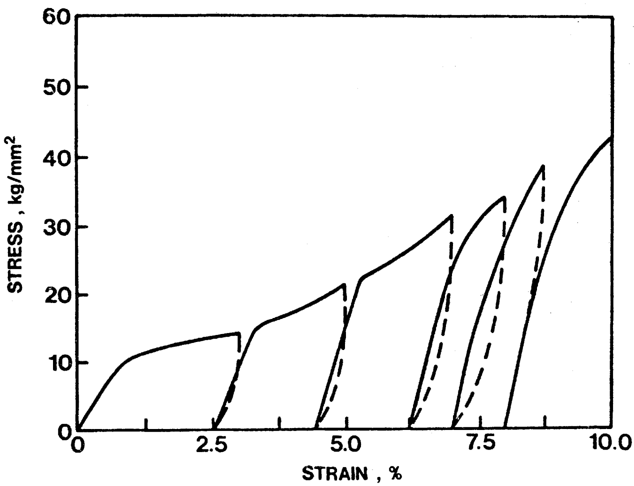

Fig 7—Typical stress-strain curve, after Dumbleton (38), for holocellulose summerwood fibres of long leaf pine. The fibres had been dried under a constant compressive strain of 10%.

Early work on the stress-strain curves of single wood-pulp fibres showed them to be linear or nearly so[33,34], and very different from the characteristic shape of the stress-strain curve of paper. This left considerable room for an explanation in terms of changes in paper structure. More recent work on the properties of single fibres has shown however that the stress-strain curves found in the early work are by no means typical, because the fibres were prepared by unusually careful methods: often holocellulose was used, gently disintegrated. As a result few dislocations or micro-compressions were introduced into the fibres.

In contrast, the fibres in paper sheets usually contain many micro-compressions. They are introduced progressively during defibering[35], bleaching[36], beating or refining[37] and finally at every fibre-fibre crossing in the sheet by the mutual interaction of fibres during drying[26].

When fibres containing many microcompressions are tested, the stress-strain curves resemble those of paper, showing an elastic regime, a yield point, and permanent set.

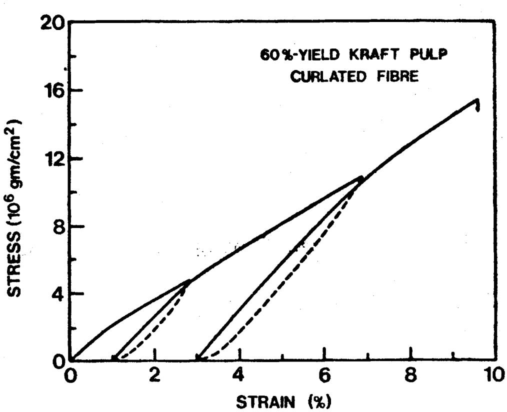

Fig 8—Typical stress-strain curve for 60%-yield kraft pulp fibres of black spruce in which microcompressions were introduced by high-consistency curlating action.

Figure 7 shows the results of Dumbleton[38]. In his work the micro-compressions were introduced artificially by applying an axial compressive strain to each fibre in the wet state.

Figure 8 is drawn from the work of Page et al.[39]. In this case the microcompressions were introduced by a high-consistency curling action.

In view of the foregoing, and in the light of the further evidence of this paper, we wish to claim that the non-linearity and visco-elasticity of the stress-strain curve of paper arise largely from the fibres. Moreover we claim that the stress-strain curve can be explained in exactly the same way that we have explained the elastic regime. In the elastic regime, the stress at any strain depends on the elastic properties of the fibres, and depends on structure only through the orientation factor (1/3) and the efficiency factor φ. Similarly, in the plastic regime the stress at any strain depends on the visco-elastic properties and on the structure through the orientation factor (1/3) and the efficiency factor φ. The orientation factor is constant during straining since for such small strains the change in fibre orientation is negligible. The efficiency factor may fall, however, during straining because of bond breakage.

This may be expressed formally in the following way:

$$E_p^* = 1/3 \ \varphi^* E_f^* \tag{4}$$

where E_p^* and E_f^* now represent the stress divided by strain for both the elastic and inelastic, time-dependent, regions of the stress-strain curve. φ^* now represents the strain-dependent efficiency factor.

The remainder of this paper provides evidence to support these claims, by examining the ways in which the stress-strain curve responds to certain treatments.

Investigation of influences on the stress-strain curve

CHANGE IN THE STRESS-STRAIN CURVE WITH:

(a) Fibre properties constant,
(b) Efficiency factor unchanged by paper-making treatments,
(c) Efficiency factor unchanged by straining.

We wish to consider, first, experiments where the stress-strain curve is changed while the dry fibre properties are held constant and also the efficiency factor is kept constant, close to unity. This occurs when the relative bonded area is high; the elastic modulus is then constant at one-third of the fibre modulus. Any change in bonded area during straining will have no effect on the efficiency factor since the bonded area is adequately high. In this case when bond strength or bonded area are changed, the shape of the entire stress-strain curve remains constant; only the end point changes. This is shown schematically in fig. 9.

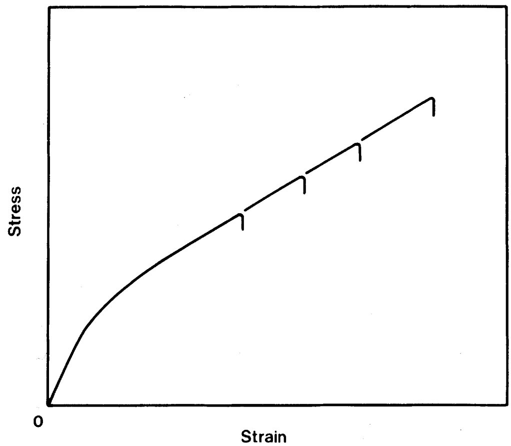

Fig 9—Schematic representation of the change in the stress-strain curve of paper with treatments that do not change the dry fibre properties and the efficiency factor.

We have tested this claim by the following three experiments.

1. Effect of bond strength additives

We have changed bond strength with two additives, locust bean gum and Hyamine 2389. Locust bean gum is known to increase fibre-fibre bond strength and Hyamine reduces it. Some of the physical properties of the sheets are shown in Table 1.

	Control	With Bonding Agent	With Debonding Agent
Elastic modulus (km)	830	836	830
Tensile strength (km)	8.08	9.13	6.54
Strain at failure (%)	3.77	4.08	2.56
Scattering Coefficient (cm^2/g)	242	211	267
Sheet Apparent Density	0.680	0.682	0.671

Table 1

The effect of changing bond strength on the properties of sheets made from a commercial, dried, bleached kraft pulp of softwood beaten for 3000 revolutions in a PFI Mill.

The gross structure of the sheet has clearly been maintained constant; the density is unchanged. Although the scattering coefficient has changed, the bonding is still high enough not to affect the elastic modulus. The effect on tensile strength is as expected. The locust bean gum increases tensile strength compared with the control and the Hyamine reduces it. The stress-strain curves are identical (figure 10). They differ only in the termination point, and when superimposed, can hardly be distinguished.

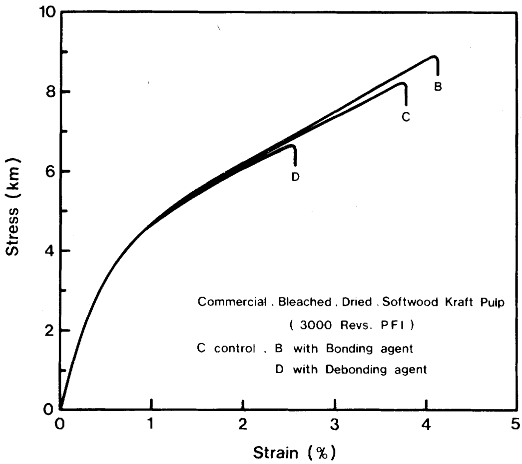

Fig 10—Stress-strain curves for handsheets made from a commercial, dried, bleached kraft pulp of softwood beaten for 3000 revolutions in a PFI mill. Bonding and debonding agents were added to the beaten pulp before sheetmaking.

2. Effect of change in bonded area by wet pressing

The bonded area of sheets made from a never-dried, unbleached kraft pulp of black spruce was changed by wet pressing. The fibres were straight and flexible so that the modulus had reached its plateau at the lower pressing pressures. Increasing the bonded area increased both tensile strength and strain at failure, but as can be seen from the curves of figure 11, it had no effect on the shape of the stress-strain curve.

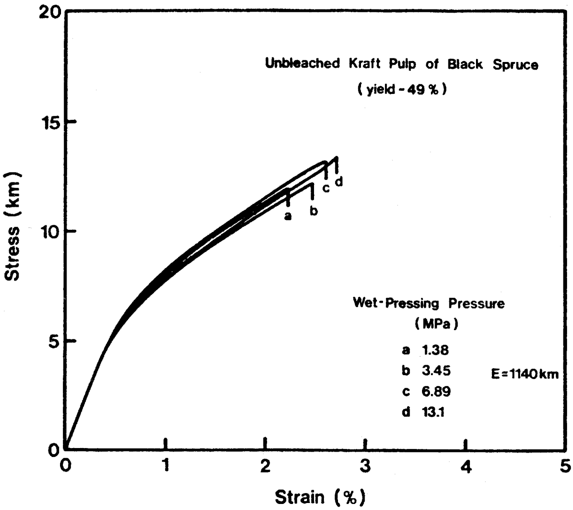

Fig 11—Stress-strain curves for handsheets prepared at different wet-pressing pressures from a laboratory-made, never-dried, unbleached, 49%-yield kraft pulp of black spruce.

Similar results are shown in Figure 12 for another never-dried, unbleached kraft pulp of black spruce.

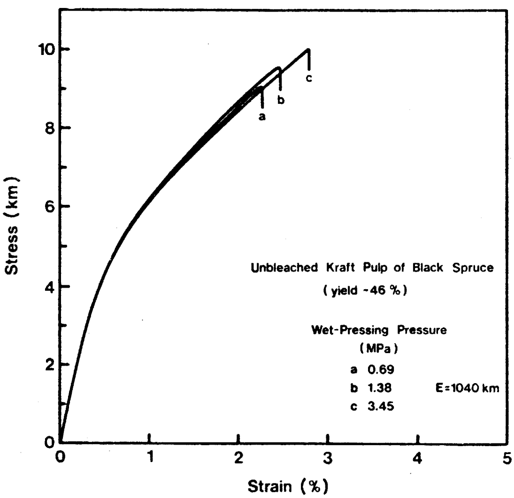

Fig 12—Stress-strain curves for handsheets prepared at different wet-pressing pressures from a laboratory-made, never-dried, unbleached, 46%-yield kraft pulp of black spruce.

3. Effect of change in bonded area by beating

The bonded area has been changed by beating a never-dried, unbleached kraft pulp of eastern Canadian white pine. The fibres in the unbeaten pulp were straight, thin-walled and flexible so that the modulus of the sheets was already on the plateau. The tensile strength and strain at failure were both increased by beating. The shape of the stress-strain curve remained constant, as shown in figure 13, and only the end points of the curves were changed by beating.

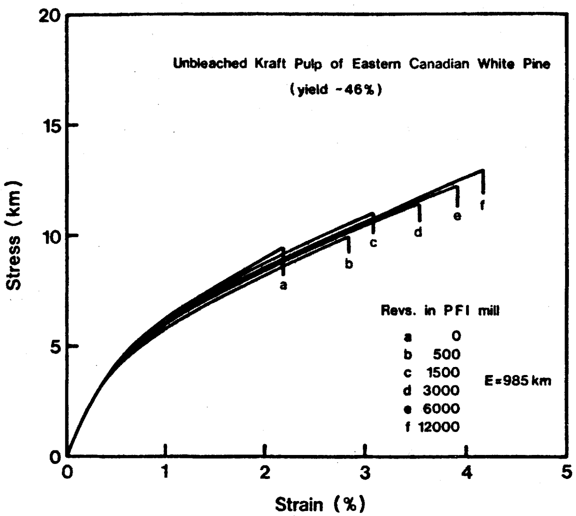

Fig 13—Stress-strain curves for handsheets prepared at different beating levels from a laboratory-made, never-dried, unbleached, 46%-yield kraft pulp of eastern Canadian white pine.

Similar results were obtained (figure 14) for a never-dried, unbleached kraft pulp of black spruce.

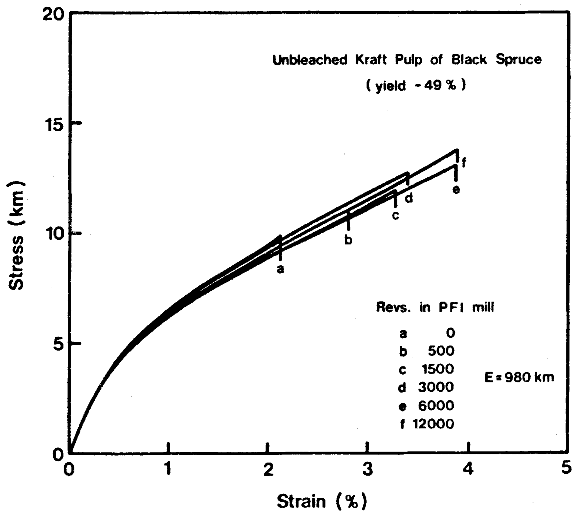

Fig 14—Stress-strain curves for handsheets prepared at different beating levels from a laboratory-made, never-dried, unbleached, 49%-yield kraft pulp of black spruce.

<u>CHANGE IN THE STRESS-STRAIN CURVE WITH:</u>

(a) Fibre properties constant,
(b) Efficiency factor changed by paper-making treatments,
(c) Efficiency factor unchanged by straining.

In certain experiments, the stress-strain curve may be changed by keeping the fibre properties constant while changing the efficiency factor. For example, if the bonded area is changed by varying the wet-pressing pressure for sheets in which the bonded area is below that necessary to bring the modulus to the plateau, the efficiency factor will change. If bond strength is sufficiently high, the change in bonded area during straining will be small and the efficiency factor will remain sensibly constant during straining. The result will be to produce stress-strain curves as shown in figure 15a.

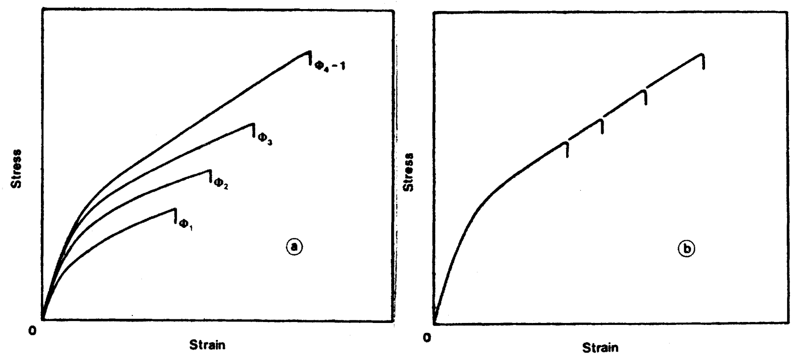

Fig 15—(a) Schematic representation of the stress-strain curves for handsheets with constant fibre properties but varying efficiency factors. By dividing these curves by appropriate efficiency factors, they can be transposed to a single curve (b).

It follows that all these stress-strain curves may be brought to a single one by dividing each by the efficiency factors determined from the ratio of the modulus of each curve to the modulus reached at high bonded area. The series of curves transposed in this way will appear as a single curve as shown in Figure 15b.

The following experiments illustrate this:

1. Effect of change in bonded area by beating

A series of stress-strain curves obtained by beating a never-dried, unbleached kraft pulp of southern U.S. pine is shown in Figure 16a. Although the pulp is never dried, the thick-walled fibres create a poorly bonded sheet in the unbeaten state so that the modulus has not reached its plateau. When the stress-strain curves are transposed, by dividing by the efficiency factor, they superimpose as shown in Figure 16b.

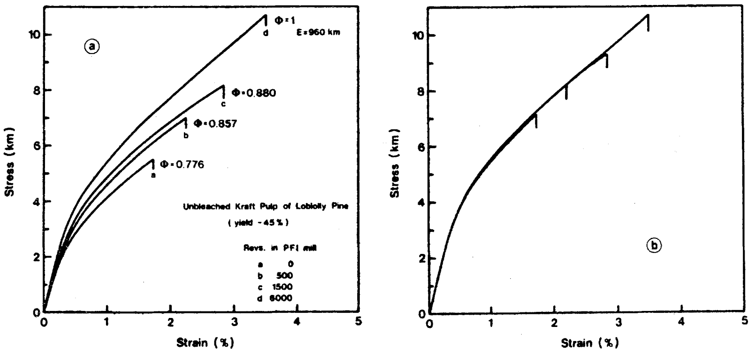

Fig 16—(a) Stress-strain curves for handsheets prepared at different beating levels from a laboratory-made, never-dried, unbleached, 45%-yield kraft pulp of Loblolly pine. The transposed curves are shown in Figure (b).

Similar results were obtained (figures 17a and 17b) for a never-dried, unbleached kraft pulp of Douglas fir.

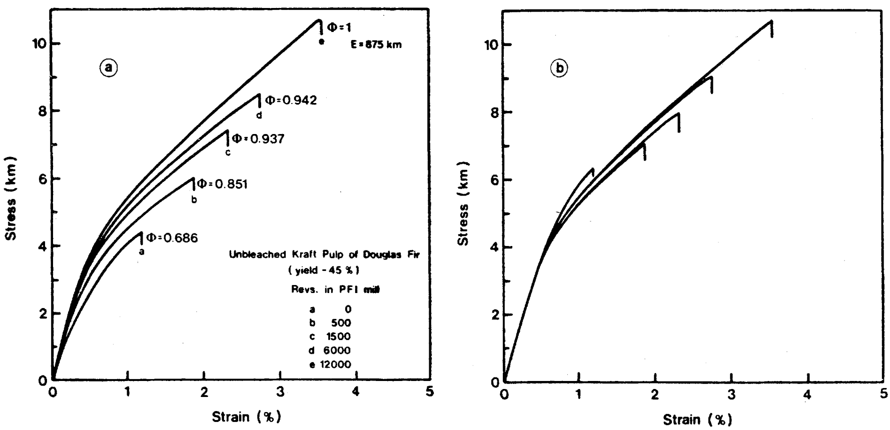

Fig 17—(a) Stress-strain curves for handsheets prepared at different beating levels from a laboratory-made, never-dried, unbleached, 45%-yield kraft pulp of Douglas fir. The transposed curves are shown in Figure (b).

2. Effect of change in bonded area by wet pressing

A series of stress-strain curves obtained by varying the wet-pressing pressure on sheets made from a never-dried, unbleached kraft pulp of black spruce is shown in figure 18a and is shown transposed in figure 18b.

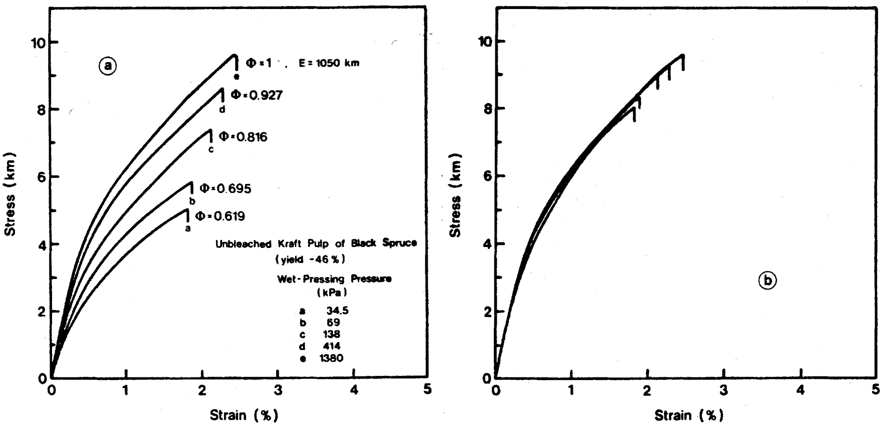

Fig 18—(a) Stress-strain curves for handsheets prepared at different wet-pressing pressure from a laboratory-made, never-dried, unbleached, 46%-yield kraft pulp of black spruce. The transposed curves are shown in Figure (b).

Similar results were obtained (figures 19a and 19b) for another never-dried, unbleached kraft pulp of black spruce.

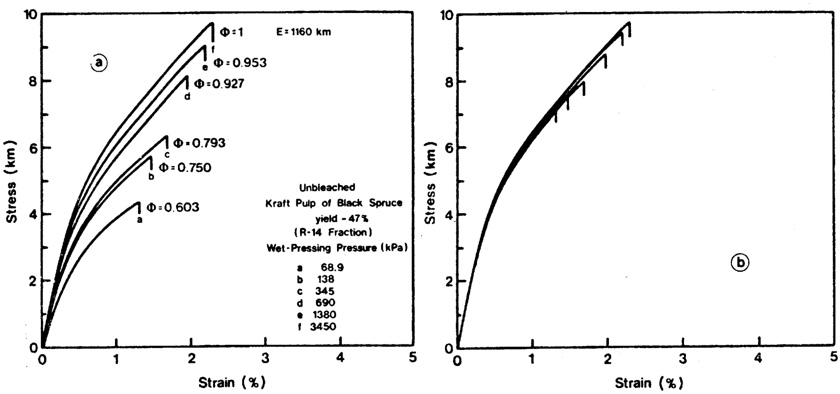

Fig 19—(a) Stress-strain curves for handsheets prepared at different wet-pressing pressures from R-14 Bauer-McNett fraction of a laboratory-made, never-dried, unbleached, 47%-yield kraft pulp of black spruce. The transposed curves are shown in Figure (b).

Thus, all the foregoing treatments give a single curve, when transposed.

CHANGE IN STRESS-STRAIN CURVE WITH:

(a) Fibre properties constant,
(b) Efficiency factor changed by paper-making treatments,
(c) Efficiency factor changed during straining.

These are cases where both the bonded area and the bond strength are low. Because of bond breakage during straining the modulus drops and the efficiency factor changes. The consequence of this is that the stress-strain curves do not superimpose when transposed. An example of this effect is shown in figure 20. The addition of Hyamine in appreciable quantities has the effect of reducing both the bond strength and the bonded area. The stress-strain curves when transposed are shown in figure 21.

In these cases of low bond strength, the elastic modulus falls appreciably during straining, since the efficiency factor falls, due to breakage of fibre-fibre bonds. This phenomenon has been observed by Taylor(40). In the above experiment, the modulus of the control sample did not change during straining, while the modulus of the sample treated with 20% Hyamine dropped 24% upon straining. If this drop in efficiency factor is taken into account, the stress-strain curves of the samples superimpose upon transposition.

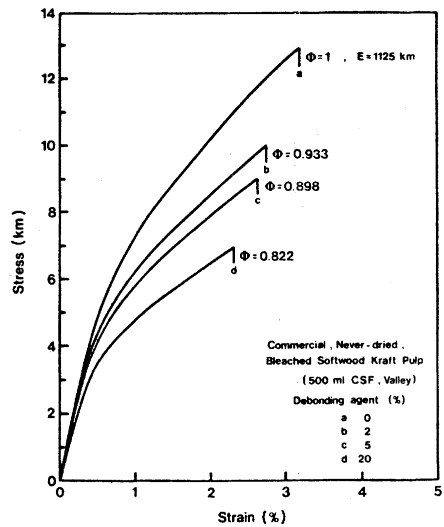

Fig 20—Stress-strain curves for handsheets prepared from a commercial, never-dried, bleached kraft pulp of softwood, beaten to 500 ml CSF in a Valley beater. Different amounts of Hyamine 2389, a debonding agent, were added to the beaten pulp before sheetmaking.

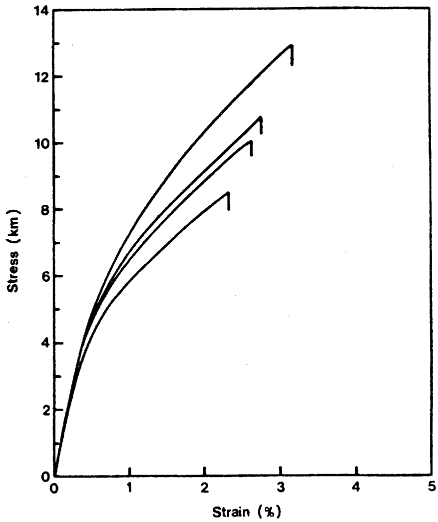

Fig 21—The transposed curves of the stress-strain curves shown in Figure 20.

CHANGE IN STRESS-STRAIN CURVE WITH:

Fibre Properties Not Constant

There are many treatments that affect the fibre properties controlling the shape of the stress-strain curve, and they fall into two main classes. Firstly, there are treatments that either introduce crimps and microcompressions into the fibre, or remove already existing crimps. Secondly, there are treatments that affect the molecular mobility of the interfibrillar matrix of the fibre wall. When such treatments are given it would be expected that the stress-strain curve would change its shape completely, and it would not be possible to superimpose them by the procedures of the earlier section.

1 Effect of introduction of kinks and microcompressions

Kinks and microcompressions may be introduced either by mechanical action applied to the pulp suspension or by the shrinkage stresses during drying.

An example of the effect of high-consistency curling is shown in Figure 22. The introduction of microcompressions has created an extensible sheet as discussed elsewhere(37). For our purposes it is important to note that the curves cannot be transposed to match the curve of the untreated fibres.

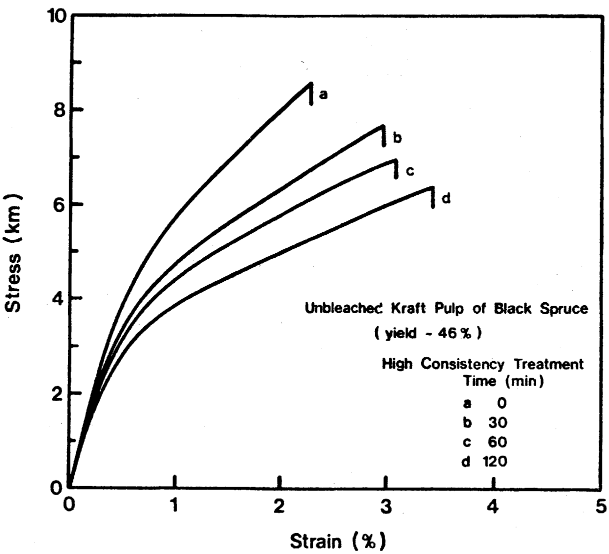

Fig 22—Stress-strain curves for handsheets prepared from a never-dried, laboratory-made, unbleached, 46%-yield kraft pulp of black spruce, which had been treated in a Hobart kitchen mixer at 20% consistency for different times. This treatment introduces kinks and micro-compressions into the fibres.

The same effect is present in sheets dried without restraint as shown in Figure 23. Again the curves do not superimpose when transposed.

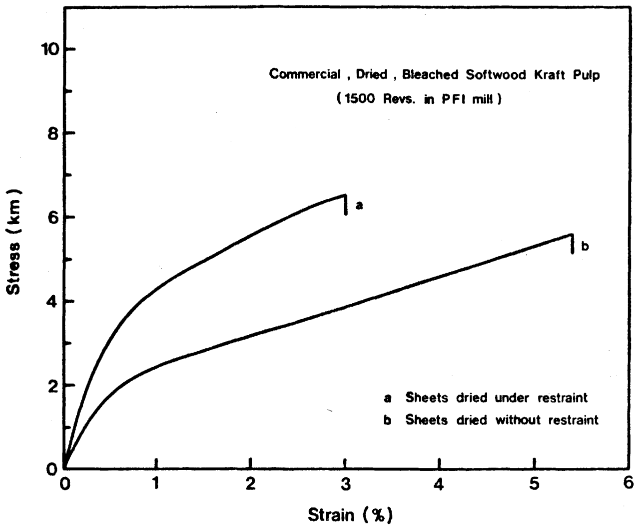

Fig 23—Stress-strain curves for handsheets made from a commercial, dried, bleached kraft pulp of softwood, beaten for 1500 revolutions in a PFI mill, and dried with and without restraint.

2 Effect of change in molecular mobility of the interfibrillar matrix

The interfibrillar matrix, consisting generally of hemicellulose and lignin, may be made more mobile by increasing the relative humidity, increasing the temperature or decreasing the strain rate. Many stress-strain curves of these effects have been given in the literature and some of them are reproduced here in figures 24-26. Again our interest in these curves is that they do not superimpose when transposed.

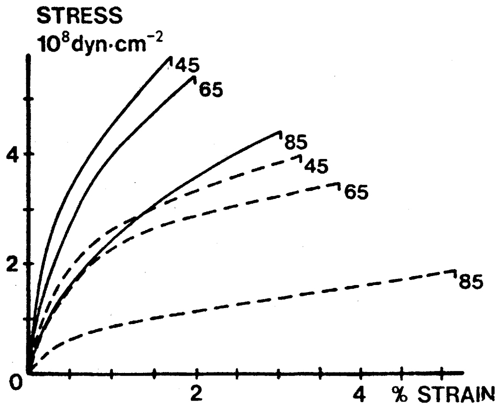

Fig 24—Stress-strain curves, after Kubát and Nyborg (41), of MG unbleached kraft paper, 58 g/m^2, at different relative humidities (numbers in the figure). Strain rate: 0.042% s^{-1}. MD ———— CD. – – – – – –

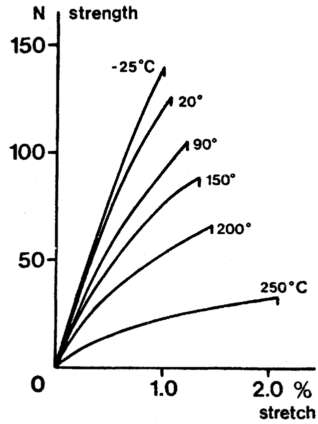

Fig 25—Stress-strain curves, after Salmen and Back (42), at different temperatures, of a dry fluting of 112 112 g/m^2 weight, in the machine direction. Strain rate: 0.17% s^{-1}.

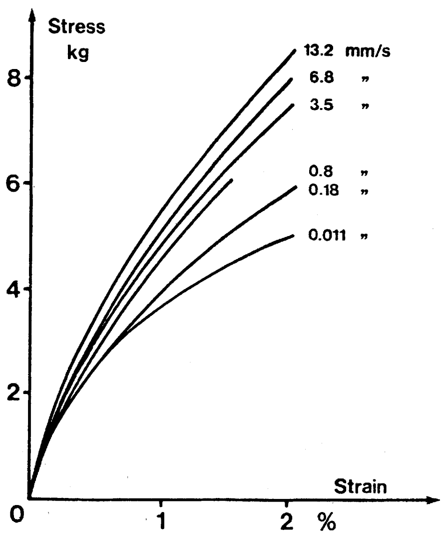

Fig 26—Stress-strain curves, after Andersson and Sjöberg (43), of MG kraft pulp at different rates of elongation, tested in the machine direction.

Discussion

A major problem that has faced paper physicists for twenty years is now resolved. The paper structural theory of the origin of the plastic regime, as originally proposed by Rance[21], is discredited: visco-elasticity comes from within the fibres. Bond breakage during straining can affect the stress-strain curve by reducing the efficiency factor but its effect is usually small.

Although the picture presented here relies on extensive evidence, perhaps the most decisive is that of Figure 10, showing the effect of bond strength on the stress-strain curve. During this experiment the change in scattering coefficient was recorded as shown in figure 27.

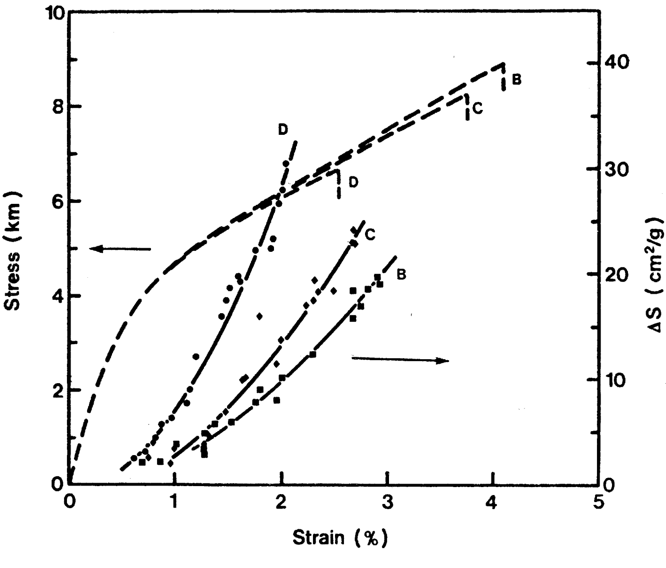

Fig 27—The change in scattering coefficient with strain for the handsheets of Figure 10. Strips 38 cm wide and 10 cm long were strained to different loads at an elongation rate of 0.5 cm/min. Scattering coefficient was measured at three different positions on each strip before and after straining to determine the change in scattering coefficient.

The increase in scattering coefficient is highest for the sheets treated with the debonding agent and lowest for the sheets treated with the bonding agent, confirming that the bond strength has been changed. Yet the sheets have identical stress-strain curves. Only the end point has changed.

The work on properties of the single-fibre[39] has shown that it is the misalignment of the fibrils with the fibre axis that is responsible for the visco-elastic behaviour. This misalignment may be either natural as in the case of fibres of high fibril angle, or, more commonly, induced, as in the case of microcompressed fibres. The mechanism of the curve is made clear in Figure 28. As a microcompressed fibre is strained elastically, the interfibrillar structure undergoes shear. As with most non-crystalline polymers, at a certain shear stress, the matrix begins to flow, allowing the micro-compression to be drawn out. If the straining is arrested, the stress-strain curve will show all the features of permanent set, delayed elastic recovery, creep, and stress relaxation, arising from the stresses in the interfibrillar matrix. In the final stages of straining before failure the stress-strain curve may steepen as the fibre straightens out.

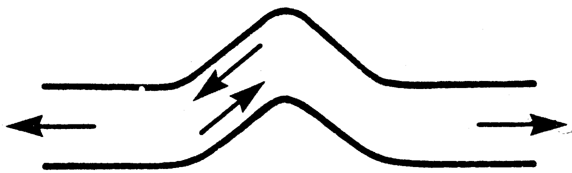

Fig 28—Shear in the interfibrillar structure of a microcompressed fibre under tensile strain.

This study now incorporates, interprets, and unifies many previous concepts of the stress-strain behaviour of paper. For example the springs and dashpots of Steenberg[44] are now seen as describing the behaviour of the interfibrillar matrix under stress, rather than the response of the paper structure. The work of Nissan[45] may be interpreted as an attempt to tackle the behaviour of this hydrogen bonded entity. The built-in stresses discussed by Kubát[29] are also matrix properties, as are the temperature, humidity, and strain-rate dependent properties.

The loss of modulus that occurs upon straining is indeed caused by bond breakage. However, it has only a minor effect on the shape of the stress-strain curve and the attempt by Kallmes and Perez[10] to attribute the entire shape of the curve to this cause is incorrect.

Only one major concept in the literature is not included in our model. It is the idea that because of fibre-fibre bond breakage, a major redistribution of stress occurs during straining and this controls the shape of the curve. The insensitivity of the stress-strain curve to bond breakage (shown by figure 27) together with the weight of the other evidence of this paper demands that this concept be rejected.

Conclusion

The stress-strain curve of paper is controlled by paper structure through a factor that takes into account the orientation of fibres in the sheet and a factor that describes the efficiency of the stress distribution in the fibres. In most cases these factors are relatively constant throughout the stress-strain curve and in combination they relate the curve of the fibres to the curve of the sheet. Non-linearity of the curve and the visco-elastic properties of paper originate from within the fibre wall. Bond breakage can play a modest role in the shape of the stress-strain curve by reducing the value of the efficiency factor during straining.

Experimental Techniques

Standard CPPA-TS procedures were generally followed. Handsheets of 60 g/m^2 basis weight were made and, unless stated otherwise, were presed in a laboratory press at the standard wet-pressing pressure of 414 kPa on the sheet. In the experiments where the wet-presssing pressure was varied, the pressures ranged from 34.5 kPa to 13.1 MPa.

The scattering coefficient was determined from the reflectance measurements made on sheets at 681 nm wavelength with a Zeiss Elrepho photometer. The stress-strain curves were obtained on 100mm long, 15mm wide specimens with a floor-model Instron universal testing instrument and are mean curves for ten specimens. The elongation rate was 10mm/min. The stress was calculated by dividing the load per unit sample width by the basis weight of the sheet, and is expressed as a length. It is the stress that is borne only by the solid fraction of the sheet.

All measurements were made at 22°C temperature and 50% relative humidity.

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Discussion

Discussion following paper presented by Dr. D.H. Page

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

Firstly, let me congratulate Dr. Seth and Dr. Page for an excellent paper. I look forward to the research efforts of future generations to prove or disprove their hypothesis. Their single fibre results and the application of these to apparent plastic behaviour were particularly impressive.

However, I was surprised at their neglecting the role of inhomogeneity of sheet structure. Because in any real paper the fibres are flocculated to a greater or lesser extent, there must always be more and less active areas in a sheet. Thus their efficiency factor ought to contain information from both the formation index, and the activity factor.

The activity factor is controlled by:

a. the degree of shrinkage actually permitted during drying.

b. the shrinkage potential (related to the amount of refining and the fibres′ response to it).

c. the amount of flocculation.

The figures below (reproduced from my Ph.D. thesis, referenced by Seth and Page) illustrate the role of fibre activation.

In the top one the load elongation behaviour of three laboratory handsheets made in different ways from the same bleached kraft pulp is shown. Curve A relates to a sheet made from a highly refined pulp, wet pressed, and allowed to dry freely. Curve B refers to a handsheet made from a well-refined pulp, wet pressed, and allowed to dry freely. Curve C shows the response of a handsheet made from well-refined pulp, wet pressed, and ring dried.

Both freely dried sheets, curves A and B, show excessive response to the initial load i.e. both show an apparently increasing modulus with increasing extension. I interpret this as meaning that more material is being subjected to the load as it increases. The associated heat transfer curves (A and B on the lower figure, taken simultaneously) demonstrate Kelvinian behaviour, implying that the loading response is governed by elastic deformation.

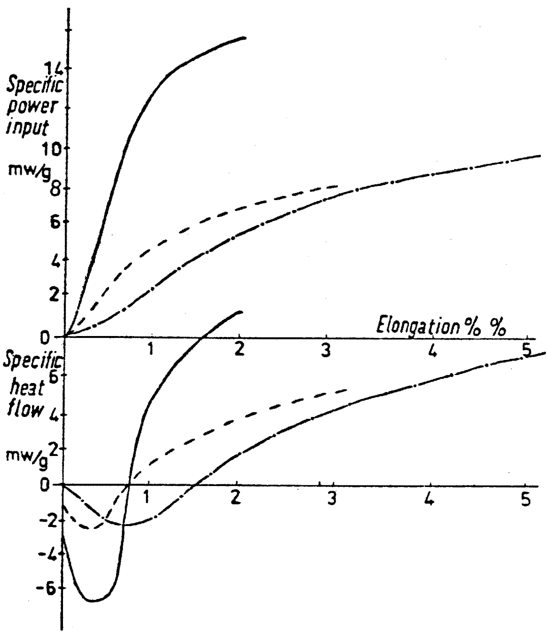

Fig 1

In the case of curve C (the ring dried handsheet) the refining induced shrinkage potential of the cell wall has made the fibrous material much more active. Thus the apparent modulus of this sheet is very high. Also the associated heat transfer is very high, emphasising that a large number of available fibres is undergoing elastic deformation.

After further loading the zone of so-called plastic deformation is reached, where irreversible heat generation commences in the paper due to the release of strain energy. The onset of the irreversible structural change associated with this behaviour occurs sooner in handsheet C than in the other two.

This is because a high proportion of the fibrous material was active from the commencement of straining. In handsheets A and B (especially) the onset of plastic deformation occurs only after considerably more straining because it is necessary first to load all the inactive regions of the sheet. Thus even the apparently elastic region of curve B (and to a lesser extent that of curve A) contains irreversible structural changes (inter- and intra-fibrous) as the less active areas become loaded. This has the effect of prolonging the apparently elastic deformation zone, and of smoothing the transition to the apparently plastic zone.

This is why I feel the authors' efficiency factor should make allowance for the heterogeneity of the structure, by way of the degree of flocculation and the relative fibre activity (governed by the refining and drying history).

Dr. D.H. Page, Paprican

We have investigated the effect of formation variability and found that it has a serious effect on tensile strength since points of weakness occur where the fibre population is low. The stress-strain and elastic modulus curves behave, not surprisingly, similarly. If allowance is made for the change in mass from one point to another then to a first approximation the effect can be removed. This may not, however, be true for machine made paper since we confined our investigations to handsheets. As paper weight increases there is an increase in the number of micro-compressions in the sheet, and thus there are more regions in the sheet where plastic flow can occur.

(Because of the complexity of Prof. Ebeling's question, the following written reply to his question has been included.)

The phenomenon that Prof. Ebeling describes seems to occur in systems different from the ones we have studied. Stress-strain curves that are concave to the stress axis (as also reported by Corte) are in our opinion caused by the lack of flatness of the sheets tested. Because of cockle, curl or wrinkles, that are common in sheets dried without tension, the plane strain condition needed for determination of the modulus is not met. We would not wish to incorporate such an effect in our efficiency factor. It is worth noting that formation has no effect on modulus for sheets that are plate dried. For sheets that are freely dried it is quite understandable that formation will have an effect since by influencing the local drying rate, it will influence the degree of cockle on the sheet.

Dr. D. Wahren, IPC

Since you use the term `modulus´ in a very specific manner I would like to ask whether your theory can predict any of the features of the micro-strain modulus?

Dr. D.H. Page

It doesn´t specifically, because the micro-strain modulus depends upon the modulus of the individual fibre, which might be kinked. At any point on the overall curve the micro-strain modulus will change according to how many micro-compressions have been removed, which will probably not be known. The theory does however predict that because of bond breakage the modulus will fall. This it does.

Dr. N.K. Bridge, PIRA, UK

What is the effect of changing the rate of strain, especially of increasing it? Are the factors affecting stress-strain behaviour still the same, or do they change in relative importance?

Dr. R.S. Seth, Paprican

I believe the answer to the first of those questions is given in figure 26 of our paper, which is taken from published literature. The answer to the second is `yes´.

Dr. D.H. Page

On a slightly different, but related topic, changing the temperature or humidity changes the stress-strain curve of a fibre since it changes the shear stress behaviour of the inter-fibrillar matrix. In other words, reported studies of the effects of temperature and humidity on the stress-strain behaviour of paper, have really been studies of these parameters' effects on the shear behaviour of the inter-fibrillar matrix.

Mr. P. Howarth, UMIST, UK

If this theory were expanded to include machine-made paper, then the numerical factor of 1/3 would presumably have to change to account for the differing machine and cross direction properties. Is this correct? If so, would this mean that machine and cross direction stress-strain curves could be superimposed simply by a change in a numerical factor?

Dr. D. H. Page

The answers are respectively 'yes' and 'no'. The efficiency factor would be larger in the machine and smaller in the cross direction. However, because the shrinkage stresses which produce micro-compression lie mainly in the cross direction, the average cross direction fibre is different from the average machine direction fibre. Thus the stress-strain curves will have different shapes and will not superimpose.

Mr. B. Radvan, Wiggins Teape, UK

Firstly, I would like wholeheartedly to join Prof. Ebeling in congratulating you on this work, which is excellent, and which has proved invaluable in my own researches both as a predictive tool, and as an aid to understanding how to improve a given stress-strain experiment.

I am going to confine what I have to say to discussing the 'modulus', since that is what I have been working on these last few years.

The heart of the argument is your figure 6, where you show with remarkable precision how the sheet modulus is 1/3 of the modulus of the constituent fibres. It is important to look closely at the theory which gives rise to this factor.

In his original paper, Cox relies on assumptions about infinitely long fibres, which, by some means unspecified, are loaded to the same strain as the whole sheet, then, by simple integration, the factor of 1/3 is derived. This has, of course, been much used in the theory of composites since then, and both the factors introduced by the present authors have been much used in the same context, though with one big difference. In the theory of composites there is always a matrix present, and the formula $1/3\ \varphi\ E_F$ is used for the volume fraction of the total. In the present case there is no volume fraction since the air plays no part. Thus there is a problem in describing how the stress is transferred from one fibre to another.

We could describe paper as being composed of a number of fibres which do not touch one another, the fibrous fraction whose contribution would be 1/3 the strength of a fibre, embedded in a matrix of identical fibres. The question then arises as to how we should evaluate the strength contribution from the matrix.

A second partition into non-connected and other fibres could then be made, and the argument repeated until all fibres had been considered. In this way we could account for the factor of 1/3, but the argument seems artificial and not very convincing.

I suggest that the Cox formula does not apply to paper because there is no matrix and that we must resort to contrived explanation to make it fit the facts. You do not have such an explanation in your paper.

Dr. D.H. Page

(Edited reply received after the symposium.)

Let me argue this way. With infinitely long fibres, randomly orientated, loaded only by the clamps, then you will agree that the elastic modulus is one third that of the fibres. You will agree too that bonding the fibres at crossing points doesn't change the position. Now introduce fibre ends by cutting. Every time one is cut, the stress in the fibre close to the end relaxes and is taken up by shear in the crossing fibres. This loss in strain energy implies a loss in modulus from the maximum value of one third. Our efficiency factor describes this loss.

Mr. B. Radvan

This is the familiar argument presented by Cox, but in composite theory there is a matrix volume fraction which is not the case in the non-composite paper. This is the point I was trying to make.

Dr. D. H. Page

If my previous argument is simply accepted then it is not necessary to consider composites.

Prof. B. Steenberg, Chairman

I am sorry but we must cut short this interesting discussion in order to allow others the chance to ask questions.

Prof. R.W. Perkins, Syracuse University, USA

I am interested in the stress-strain curves of individual fibres, not in paper, and their relationship to those fibres within a sheet. Surely, within a sheet the inter-fibre bonds restrain the deformation of the individual fibre, so that I would expect there to be a large difference between the in and out of sheet cases. As an example, you demonstrated a yield point in your cine film; now I don't know if this was a metallic-type yield, or an internal fracture of, say, the S_1 wall. In the latter case then the inter-fibre bonding within the sheet would change this behaviour. Would you please comment on this?

Dr. D.H. Page

We believe that the reason for the yield point is the shear within the fibrils of the S_2 layer. We do not believe that the presence of other fibres will significantly affect this. What it would affect, however, is the post-yield behaviour, the buckling, since the fibre is not free to contract as it was when alone. However, we have not considered the post-yield behaviour of the sheet: our presentation is restricted to consideration of the elastic deformation. We do not believe it would affect, to any major degree, the stress-strain curve of the fibre.

Mr. P.E. Wrist, Mead Corporation, USA

I understand you to have defined the elastic modulus at a point on the stress-strain curve as the ratio of total stress to total strain at the point. In the early Hookean phase this modulus is a constant whose value is the same as that defined by the ratio of a small increment of stress to a small increment of strain around the point (the micro-strain modulus). Beyond the yield point these two quantities begin to assume different values. It would seem to me that in attempts to understand the rheological behaviour of a fibre or a network, particularly in the plastic deformation regime, the micro-strain modulus would be more likely to be useful than the average value over the entire Hookean and plastic regimes. Have you considered the use of the micro-strain modulus in your analysis?

Dr. S.R. Seth

I believe we have discussed this on page three of our paper, where the modulus is observed to change on load cycling.

Mr. A de Ruvo, STFI, Sweden

Your formula predicts only the extensional modulus, and fibres are known to be highly anisotropic. Would your theory predict the same extensional modulus for two sheets made with fibrillar angles of 0^{o} and 45^{o}?

Dr. D.H. Page

No, the modulus drops for large fibrillar angles, reflecting the change in fibre modulus. This has been confirmed experimentally by us and in Australia.

Prof. H. Kropholler, UMIST, England

Would you be prepared to indicate where you envisage the usefulness, predictive ability, and application of the work you have just described to lie?

Dr. D.H. Page

People are interested in the stress-strain curve of paper for all sorts of reasons. The elastic modulus is of importance in connection with stiffness of the sheet, stretch to break, tensile energy absorption, and so on. We have used the theory to make predictions in specific areas. It has also enabled manufacturers to be more specific about their problems and describe product changes more accurately. For example, it is now possible to resolve a given problem into definitive terms, e.g. inadequate bonding or fibre length. The theory gives useful, practical results, which is really all one can say. Ben Radvan's opening compliments support this viewpoint.