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## EFFECT OF STRUCTURE ON THE MECHANICAL PROPERTIES OF PAPER

#### W. H. ALGAR, Department of Cellulose Technology, Technical University of Norway, Trondheim, Norway

*Synopsis*—Recent literature relating to the structure of paper and its load/ elongation behaviour is reviewed.

Following an outline of the more important load/elongation features and the effects on them of testing conditions and sheet variables, paper structure is considered in terms of the fibre (its strength, conformability and response to drying tension), the interfibre bond (its structure, area, frequency, strength and energy) and the sheet geometry. The structural changes that occur in the sheet as a whole and in its various elements during elongation and rupture are also described.

Theories relating mechanical properties to sheet structure are summarised in three categories—general theories, quantitative theories for the prediction of sheet elasticity and the application of the Griffith crack theory to paper. Some concluding remarks are offered about the significance, particularly in sheet elasticity theories, of the ability of a paper to distribute load evenly over all its structural elements.

#### INTRODUCTION

AT THE 1961 Oxford symposium, the structure of paper was discussed in detail and several new theories presented to explain its mechanical behaviour under various specified conditions. At the same time, important new evidence was produced about the basic structural elements—the fibres and the inter-fibre bonds—and their configurations in the sheet. Since that time and no doubt inspired by that symposium, there have been further advances made in both the theoretical and experimental aspects. The purpose of this paper is to survey these developments, providing background information that may be of value in the deliberations of this more specialised 1965 symposium and at the same time establishing some basic continuity between the two symposia.

The paper will comprise four main sections. The first will present an outline of what are considered to be the important mechanical properties of paper in this present context. The second section will summarise relevant knowledge of the structure of paper, including the structural changes occurring in elongation and rupture. Next, the various theories that have been proposed to relate structure to mechanical properties will be described, with some emphasis on those relating to the quantitative prediction of paper elasticity. In the final section, the theories will be discussed briefly and some general comments offered.

#### BASIC MECHANICAL PROPERTIES

FROM the wide range of mechanical properties now measured for paper, only those derived from or closely associated with the load/elongation curve will be considered in this review. Such properties can be regarded as basic in the sense that, on the one hand, they are the most likely to relate closely to structure, whereas they can be used on the other hand, either directly or in combination, to describe many of the mechanical use requirements of paper. Moreover, they give important information on the whole course of prerupture behaviour. Empirical, complex and solely destructive tests such as burst and tear will not be discussed.

#### Outline of load/elongation behaviour

As demonstrated in the pioneering work of Gibbon, Farebrother, Steenberg and Rance and their colleagues, paper exhibits the properties of a viscoelastic material. Its load/elongation curve is usually interpreted as an initial elastic regime followed by a yield point at which plastic behaviour starts to become significant. When paper is elongated past its yield point to a load not previously applied to it, then allowed to contract to zero load, it exhibits an immediate elastic recovery. On standing, it will contract further by an amount known as its *delayed elastic recovery*, the remaining elongation being its permanent set. If, after loading and contraction, the paper is then elongated again at the same rate to the load at which contraction was commenced, a hysteresis loop will be formed, indicating irreversible energy absorption. The elongation branch of this loop may have a greater initial slope than the original curve, indicating strain-hardening or mechanical conditioning. As loading is now continued, the curve then continues in approximately the same path as it followed before contraction, until rupture occurs. If further contraction/ elongation cycles are made from the same load, however, the irreversible energy absorption becomes less and the initial slopes of their elongation branches steeper, until, after several cycles, virtually constant values are reached.

With prolonged sinusoidal cycling between two fixed elongations, however, the irreversible energy absorption may pass through a minimum and then rise again. This behaviour, occurring after the elastic modulus of the paper has increased to a maximum value, has been regarded as evidence for *fatigue* in the sheet.<sup>(1)</sup> It is apparently possible for *fatigue recovery* to occur, as, on allowing the paper from such an experiment to stand, it was found that irreversible energy absorption and elastic modulus returned almost to their original values.

Prestraining of paper under impact conditions causes a decrease in the initial slope of the load/elongation curve, the phenomenon of *dilatancy*. Paper can be said also to follow the *Boltzmann superposition principle*, in so far as its elongation is determined not only by the load acting at the time of observation, but also by the entire loading history of the sample.

Stress relaxation takes place when paper is loaded and maintained at constant elongation. If, after loading, it is contracted to a low load, then maintained at constant elongation, relaxation recovery (upward stress 'relaxation') can occur. The rate of stress relaxation appears to be a linear function of initial stress and, upon extrapolation to zero rate, a value can be obtained for the level of internal stress in the sheet.<sup>(1-3)</sup> Rupture of the sheet can occur during stress relaxation.<sup>(4)</sup>

The elongation of paper maintained under constant load is known as *creep*: that part which is recoverable when the load is removed being termed *primary creep* (which is equivalent to elastic recovery) and that which is not recoverable, *secondary creep* (which is equivalent to permanent set). Creep occurs at all loads; however, for the apparent elastic regime, its rate is negligibly slow in normal load/elongation testing. In successive creep/ recovery cycles, the amount of secondary creep is reduced, but is never zero.<sup>(5)</sup>

This has described visco-elastic behaviour in the plane of the sheet. Limited measurements made in the direction normal to this plane have indicated somewhat similar behaviour.<sup>(6,7)</sup>

It is pertinent to recall at this stage the rheological axioms of Reiner quoted by Rance<sup>(4)</sup> some years ago in a similar context—

- (i) Every real material possesses all rheological properties.
- (ii) Every simple behaviour is a degeneracy of a more complex one.

Some load/elongation properties of paper are illustrated in Fig. 1.

#### Some factors affecting load/elongation behaviour

AMONG testing conditions, a faster rate of elongation increases rupture load, but is without appreciable effect on either elongation or elastic modulus.<sup>(8)</sup> Increase in relative humidity increases elongation, but decreases rupture load and elastic modulus; a rise in temperature decreases all properties.<sup>(9)</sup> If slippage at the clamps is prevented, decrease in strip length does not change the shape of the load/elongation curve, as earlier claimed, but it does increase rupture properties in accordance with the lower probability of





A. Load/elongation, illustrating a strain-hardening cycle

XA = immediate elastic recovery, AB = delayed elastic recovery, OB = permanent set, R = rupture point Shaded area = irreversible energy adsorption in hysteresis

The inset shows the effects that can be obtained on continued sinusoidal cycling between two fixed strains, according to Kubát, Nyborg & Steenberg<sup>(1)</sup>

**B.** Stress relaxation

The inset shows the method used by Kubát, Nyborg & Steenberg<sup>(1)</sup> to calculate the internal stress (OS) in paper

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weak spots in the shorter strip.<sup>(10)</sup> On the other hand, optical length change measurements on dead-loaded strips have shown elastic modulus to increase with decreasing strip length.<sup>(11)</sup>

Perhaps the most dramatic effect on the load/elongation curve is that due to the degree of restraint applied to the sheet during drying. A sheet dried free to shrink has a lower elastic modulus and generally a lower rupture load, but a higher elongation than one dried under restraint. Moreover, its load/ elongation curve often exhibits an upward curvature just before the rupture point. The extent to which these changes occur appears to depend on the change of length during drying and the shrinkage tendency of the pulp.<sup>(2-14)</sup> Fibre orientation results in a higher elastic modulus and rupture load in the direction of orientation, but its effect on rupture elongation is not so clearly established. In general, elongation in the orientation direction tends to decrease with increasing fibre orientation, but the opposite behaviour has been observed, particularly for very high orientations.<sup>(4, 15)</sup> Drying tension effects interact strongly with those of fibre orientation.<sup>(16, 17)</sup>

Increase in the degree of bonding in the sheet by wet pressing or beating increases both slopes of the load/elongation curve and rupture load and elongation. At a given density, the beaten sheet generally has a higher elastic modulus than one that has been pressed only.<sup>(18)</sup> At higher beating degrees, elastic modulus can reach a limiting value, fairly constant for a wide range of pulps<sup>(19)</sup> and post-yield slope and rupture elongation often pass through maxima.<sup>(20)</sup>

There is little systematic data available on the effects of pulp type and quality on the shape of the load/elongation curve. One difficulty here is the choice of a suitable basis for comparison—as in pulp evaluation generally. There is evidence, however, that at the same bonding degree, elastic modulus increases with decrease in lignin content.<sup>(21)</sup> Moreover, the way in which elastic modulus varies with bonding degree appears to be different for sulphite and sulphate pulps from the same wood.<sup>(22)</sup> For a given sheet density change produced by beating, the elastic moduli for pine and spruce pulps increase more than do those for birch and beech pulps.<sup>(23)</sup> For the same pulp, increasing fibre length has been found to increase both rupture load and post-yield slope.<sup>(24)</sup>

The effect of pulp quality on rupture properties alone has been widely studied, but this work is of only limited interest in the present context, without corresponding prerupture data, particularly elastic modulus.

#### STRUCTURAL CHARACTERISTICS

#### The fibre

THE significance of individual fibre properties varies considerably among

the different theories relating the structure and mechanical properties of paper. Some, which view paper essentially as a molecular assemblage, place little weight upon them, whereas others, dealing with the mechanics of the network, require extensive data of this type. Two requirements that appear essential here are, firstly, that of measuring distribution functions as well as mean values because of the heterogeneity of woodpulp fibres and, secondly, that of ensuring that the properties measured relate to the behaviour of the fibres in the condition in which they exist in the sheet, not to some idealised condition. Some recent work on individual fibre properties will now be described.

**Fibre strength properties**—The first major work in this area was that of Jayne<sup>(25)</sup> who measured load/elongation curves for holocellulose fibres from Douglas fir, cypress and white spruce. He found that these had the same general shape as the curves obtained for paper and gave the following values—

Rupture stress	$3.4 - 9.9 \times 10^9 \text{ dyn/cm}^2$
Elastic modulus	$18 - 43 \times 10^{10} \text{ dyn/cm}^2$
Rupture elongation	1.9-3.2 per cent
Elongation at vield point	0.8–2.0 per cent

Summerwood fibres were found to be stronger than springwood fibres and to have a higher elastic modulus.

Kallmes & Bernier<sup>(22)</sup> found similar load/elongation behaviour to Jayne, with elastic moduli in the range  $27-34 \times 10^{10}$  dyn/cm<sup>2</sup>. More recently, Kallmes, Stockel & Bernier<sup>(26)</sup> have found that the elastic modulus for bleached kraft softwood pulp fibres increases on beating.

McIntosh & Leopold<sup>(27)</sup> noted that the alkaline extraction of loblolly pine holocellulose reduced both fibre strength and cross-sectional area. Later, McIntosh<sup>(28)</sup> reported holocellulose rupture loads of  $8.6 \times 10^9$  dyn/cm<sup>2</sup> for summerwood and  $4.7 \times 10^9$  dyn/cm<sup>2</sup> for springwood. Normal yield kraft pulp from the same wood after delignification gave values of 5.9 and  $3.7 \times 10^9$  dyn/cm<sup>2</sup>, respectively. Intermediate values were obtained for high-yield kraft pulps. Hartler, Kull & Stockman<sup>(29)</sup> showed that, as for cotton fibres, the rupture load of spruce holocellulose fibres increased by about 25 per cent over the range 30–65 per cent rh. Britt & Yiannos<sup>(30)</sup> have found rupture loads of 7.1 and  $6.3 \times 10^9$  dyn/cm<sup>2</sup> for spruce unbleached kraft and bisulphite pulps, respectively.

Because of its importance in his structural theory of paper elasticity, Page<sup>(31)</sup> has measured the load/elongation behaviour of single fibres after axial compression—a condition which he believes exists in the bonded lengths of fibres in the sheet—and found elastic modulus to be reduced. Recent direct measurements of the density of the fibre wall by Jayme & Krause,<sup>(32)</sup> Worrall and Yiannos have been surveyed by Stone.<sup>(33)</sup> These indicate it to be considerably lower than previously believed. For example, a density of 0.88 g/cm<sup>3</sup> was found by Yiannos for both spruce unbleached kraft and sulphite pulps. These low values suggest the presence of a considerable pore volume in the fibre wall, a condition which affects the significance of stress calculations based on fibre wall cross-sectional areas.

Fibre conformability and collapse—The present conception of the consolidating and bonding mechanisms occurring in the sheet places considerable importance on the wet plasticity of the fibre, that is to say, its ability to conform to the shape of a crossing fibre under the influence of the compacting action of the surface tension forces present. A closely related fibre property is that of ease of collapse during the compaction and drying stages. Both of these properties are determined to a large extent by the wet stiffness of the fibres, which in turn depends very much on the size and shape of their crosssections. Collapse can be imagined as greatly increasing conformability in the direction in which it occurs, because of the change in cross-section and therefore of bending stiffness. Moreover, a larger potential surface becomes available for bonding with other fibres lying in essentially horizontal planes.

Wet stiffness of fibres is generally regarded as an index of their conformability, although of course stiffness over the whole drying range is probably important. Wet stiffness measurements have been made by several groups, notably Mason and co-workers<sup>(34)</sup> and more recently by Samuelsson<sup>(35)</sup> who showed that, at the same kappa number, sulphate woodpulp fibres are stiffer than sulphite fibres, that stiffness falls with decreasing kappa number and with increased beating and that springwood fibres are more flexible than summerwood ones.

The importance of fibre collapse has been discussed by Robertson & Mason<sup>(36)</sup> and later by Robertson,<sup>(37)</sup> who used as an indicator to degree of collapse the difference between the critical moistures for a pulp, determined by the hydrostatic tension and dye migration techniques. Kallmes & Eckert<sup>(38)</sup> have described a method for measuring the degree of fibre collapse in a sheet by direct microscopical observation in dark field illumination.

It should be noted that these properties of conformability and collapse are important not only in determining the degree of bonding in the sheet, but also for the final size and shape of the cross-sections of the load-bearing structural elements, a most important factor in establishing quantitative relationships between structure and strength.

*Effect of drying the fibre under stress*—Because fibres in the paper sheet are generally dried under some tension, it is possible that their strength properties in this condition are different from those measured after un-

#### Effect of structure on mechanical properties

restrained drying. To decide this important question, Jentzen<sup>(39)</sup> studied the effect of load during drying on the load/elongation properties of longleaf pine holocellulose. He found that a load applied to the wet fibre produced an elongation dependent upon the load, but, when drying commenced, a further sudden elongation took place. This appeared to be load-independent and was different for springwood and summerwood. As drying continued, a load-dependent contraction occurred. The total effects were found to be the development of a permanent set in the fibre, increases in its elastic modulus





stress calculated on solid cross-section of fibre

(up to 212 per cent), rupture stress (up to 85 per cent) and rupture energy (up to 39 per cent) and an increased orientation of the crystallites. Rupture elongation decreased (up to 40 per cent) and no change was noted in the degree of crystallinity. In general, the springwood fibres underwent much larger changes than did the summerwood. Elongation/contraction cycling was shown to increase the elastic modulus of the fibres dried under no load (about 45 per cent in three cycles). Cross-section areas used in the calculation of rupture stress, elastic modulus and rupture energy were calculated by dividing the mass per unit length of the tested fibres by the density of cellulose,  $1.55 \text{ g/cm}^3$ .

13—с.р.w. п

The explanation suggested for the length increase on drying was contraction of the fibril helix diameter caused by evaporation of water, with consequent lengthening in the axial direction. This should be independent of drying load, but related to initial fibril helix angle, two conditions which were fulfilled. The changes in mechanical properties because of the drying under load were attributed to the increase in crystallite orientation and a more even distribution of stress by relative movement of the fibrils (microfibrils) in the wet fibre rather than movement of molecules in the crystalline core. Assuming the S2 layer of the fibre wall to determine the elastic properties of the fibre, the maximum value found for the elastic modulus was  $70 \times 10^{10}$  dyn/cm<sup>2</sup> for springwood and  $72 \times 10^{10}$  dyn/cm<sup>2</sup> for summerwood. These values were believed to be close enough to the theoretical value for perfectly oriented crystalline cellulose— $90 \times 10^{10}$  dyn/cm<sup>2(40)</sup>—to conclude that the fibres have a crystalline core, in suport of the finged fibril theory of Hearle.<sup>(41)</sup>

Some of Jentzen's results, illustrating the effect of drying load on the load/elongation properties of fibres, are shown in Fig. 2, together with an example of the same effect for paper.

#### The interfibre bond

Bond structure revealed by microscopical methods-Microscopical evidence indicates that interfibre bonding occurs for the most part over a relatively large fraction of the contact area of two crossing fibres, drawn together by the consolidating processes operating during drying. Asunmaa & Steenberg<sup>(42)</sup> have shown in an electron microscope study of interfibre bonds that areas of contact up to 100  $\mu^2$  occur and that this contact is so intimate that it is suggestive of chemical bonding. Such areas were found to occur for the following fibre wall combinations-S1-S1, S1-S2 and S2-S2. Page, Tydeman & Hunt<sup>(43)</sup> and later Kallmes & Eckert<sup>(38)</sup> regard areas of optical contact between fibres as completely bonded, even though optical contact can be obtained at an interfibre spacing considerably greater than that required for molecular bonding. On the other hand, Javme & Hunger<sup>(44)</sup> maintain that the loosened microfibrils and thin lamellae that can be observed in the electron microscope at the surfaces formed by interfibre bond rupture are the real sites of bonding, amounting to only 10-20 per cent of the apparent contact area. Their investigation also showed that interfibre bonds formed during drving can break, if the shrinkage forces acting become too strong.

Buchanan & Washburn,<sup>(45)</sup> in scanning electron microscope studies, have shown that some interfibre bonding does occur via fibrillar and laminar material, even for unbeaten pulps, but that this does not make a significant contribution to bonding in the sheet as a whole, except at high beating degrees. Fibrils and fibre fragments appear to be more important in interfibre bonding in mechanical woodpulps than in chemical woodpulps. When the interfibre bond was sufficiently strong, as for example after beating, bond rupture no longer occurred at the original fibre surfaces, but deeper in the fibre wall, resulting in a transfer of material from one fibre to the other and a roughened surface appearance. Increased delignification also was shown to foster this behaviour. Similar observations have been made by Helle,<sup>(46)</sup> who believes that the cleaner rupture surfaces shown by high-yield pulps may be due to a higher fibre coherence.

An important contribution in this field is that of Page & Tydeman,<sup>(47)</sup> who demonstrated that bonding at a fibre crossing in a sheet dried free to shrink will produce a longitudinal contraction of one fibre because of the strong transverse shrinkage of the other. This contraction is believed to take the form of longitudinal microcompressions in the bonded lengths of the fibres and to be accompanied in the unbonded fibre segments by an equivalent shortening, either by microcompressions or by kinking, to ensure no change in the relative positions of the fibres at their crossings. (See also section *Theories based on paper as a fibrous network.*)

Area and frequency of bonds—Although many investigators have studied the total and relative bonded areas (RBA) in paper, there is in comparison only a small amount of data available on the area and frequency of the individual interfibre bonds, despite the obvious importance of these factors in developing a meaningful concept of sheet structure.

Data of considerable importance have been obtained by Page, Tydeman & Hunt,<sup>(43)</sup> who made direct microscopical measurements of the frequency of interfibre bonding in a sheet and the percentage of each fibre crossing that was optically bonded. For a bleached spruce sulphite pulp, they found that beating to 310 CSF increased mean bond area from 567  $\mu^2$  to 932  $\mu^2$ , at the same time increasing the frequency of bonding to a level at which virtually the whole of the fibre length was bonded on one side or the other. For one side of the fibre only, the projected interbond distance decreased from 34.0  $\mu$  to 13.2  $\mu$ . The mean width of the fibres was 44  $\mu$ . Drying tension was shown to have only a minor effect on bond size and no detectable effect on bond frequency.

Kallmes & Bernier<sup>(48)</sup> have established mathematical relationships between the frequency of interfibre bonding, RBA and fibre width. From experimental determination of the latter two factors, they calculate that, for unbeaten sheets with RBA of 30–50 per cent, the frequency of bonding is such that  $\frac{1}{3}-\frac{1}{5}$  of the fibre length is free. In well-beaten sheets, with RBA about 75 per cent, only a negligible free fibre length (about 5 per cent) remains.

Bond strength and bond energy-The failure of interfibre bonds during the

straining of paper is widely held to be one of the major factors determining its load/elongation behaviour, at least in the plastic range. The mode of failure of the bonds and the force and energy required have been the subject of numerous investigations.

Failure in shear is believed by many to be predominant, the evidence for this having been summarised by Van den Akker.<sup>(49)</sup> Experimental determinations of the shear strengths of interfibre bonds have been made by Mayhood, Kallmes & Cauley,<sup>(50)</sup> McIntosh<sup>(28)</sup> and Schniewind, Nemeth & Brink.<sup>(51)</sup> For chemical woodpulp fibres, the shear strength values ranged from as low as  $2.0 \times 10^8$  dyn/cm<sup>2</sup> to  $6.9 \times 10^9$  dyn/cm<sup>2</sup>. Bonds between summerwood fibres gave higher values than those between springwood fibres, bond strength increased with pulp yield<sup>(28)</sup> and, for the limited data available, the effect of beating appeared negligible.<sup>(50)</sup>

No direct measurements of the energy required to rupture an interfibre bond yet appear to have been made. Of the indirect methods that by Nordman<sup>(52)</sup> has been most widely studied. In this method, bond strength values (erg/cm<sup>2</sup>) are calculated from the linear relationship found between the mechanical energy lost in elongation/contraction cycles (erg/g) and the corresponding increase in light scattering coefficient (cm<sup>2</sup>/g). For a range of chemical woodpulps, rag and esparto pulps, these were unaffected by beating degree, but appeared to be related to chemical composition. The range of bond strength values found was  $1.6-8.2 \times 10^5$  erg/cm<sup>2</sup>, but these cannot be related to any defined bonded area in the sheet, as the light scattering coefficient is merely a dimensionless ratio, divided by the substance of the sheet. In more recent work on birch semi-chemical pulp, Nordman & Göttsching<sup>(53)</sup> found that the bond strength increased with beating, a result ascribed to the different chemical nature of this pulp from those used in the earlier study.

Stone<sup>(54)</sup> has repeated Nordman's approach, but also determined surface area change by the nitrogen adsorption method. For this, bond energy values of about  $2 \times 10^4$  erg/cm<sup>2</sup> were obtained, whereas the light scattering method gave values 30–50 times this. Both values gave a curve passing through a maximum when plotted against xylan content of the pulp used.

Stone points out that in interpreting these results no distinction can be made between internal and external fibre surfaces, yet it appears that nitrogen adsorption may measure only the external surfaces of water-dried fibres, as Kallmes & Eckert<sup>(38)</sup> have recently shown that the ratio of the surface area determined in this way to that determined by direct microscopical measurement is close to  $\pi/2$ , which would be expected if the external surfaces were comprised of closely packed, parallel, semicircular fibrils.

Stone suggests that the high bond energy values found for paper compared

with those calculated by Stamm<sup>(55)</sup> for the surface free energy of gels, including cellulose, indicate that the bulk of the energy used must be consumed by mechanisms other than the simple creation of new surfaces.

#### Sheet geometry

ALTHOUGH much is already known in a semi-quantitative way of the effects of factors such as fibre orientation, flocculation and fines distribution on the structure and behaviour of the sheet, it is not yet possible to describe a real paper structure in precise mathematical terms. The case for developing a suitable geometry for this purpose was stated as early as 1950 by Van den Akker,<sup>(56)</sup> who saw it as a necessary step in obtaining an adequate understanding of the interrelationships between the structure and mechanical properties of paper.

The major activity in this field is that begun by Kallmes &  $Corte^{(57)}$  and since continued with various co-workers.<sup>(38, 58-62)</sup> Their first consideration has been the statistical geometry of a thin, randomly oriented sheet, the socalled 2-D sheet. This idealised structure is assumed to consist of flat fibres of rectangular cross-section, arranged in a sheet substantially no more than two fibres thick. Bonding is assumed to occur only between the horizontal fibre surfaces. The mid-point of each fibre axis is located randomly and each fibre axis oriented randomly. It is further assumed that a number of these 2-D sheets can be treated together to simulate normal weight sheets. When this is done, arbitrary levels of bonding must be assumed between the various layers. This model has already been used to assist in establishing relationships between sheet structural parameters and sheet properties, in particular sheet elastic modulus.

It has been demonstrated in this work also that the structure of a random, idealised sheet can be described completely by two parameters, fibre width and relative bonded area. It is suggested that for many purposes handsheet structure could be described in a similar manner, an approach that would be especially valuable in pulp evaluation comparisons, particularly if they were made after extrapolation to 100 per cent RBA.

In their most recent publication, Kallmes & Bernier<sup>(62)</sup> have described how the above model can be modified to simulate both flocculated and dispersed sheets, also fibre-oriented sheets. RBA has been used to describe the sheet structures so produced.

#### Structural aspects of sheet elongation and rupture

**Changes in the fibres**—When a sheet is elongated to rupture, no fibres break outside the rupture zone itself. This observation has been reported by Ranger & Hopkins<sup>(11)</sup> and by Helle.<sup>(63)</sup> Moreover, elongation appears not to alter

the relative positions of fibres in the sheet nor to straighten out kinks or bends in them. Observing this in a sheet dried without restraint and having an elongation at rupture of 10 per cent, Helle concluded that such a high elongation, several times greater than what a single fibre could tolerate without rupture, must be due to the straightening out of longitudinal microcompressions in the fibres as postulated by Page & Tydeman.

In the rupture zone itself, Van den Akker, Lathrop, Voelker & Dearth<sup>(64)</sup> found that fibres were either broken or pulled out from the sheet. The fraction broken increased with beating (from 40 to 71 per cent), with wet pressing and with the use of bonding additives. They also observed that many fibres in the rupture zone broke after rupture had commenced. Some other factors affecting the ratio of broken to pulled-out fibres have been studied by Helle.<sup>(63, 65)</sup> Sulphite woodpulp gave a higher ratio than sulphate pulp of the same breaking length and the ratio increased with pulp yield and with rate of loading in the test. This last result was interpreted by Helle as implying that paper rupture does not occur at a certain degree of bond breaking, but is rather an event initiated by the breaking of a fibre. In the case of sheets dried without restraint, the ratio of broken fibres to pulled-out fibres decreased, despite the higher degree of bonding in such sheets as evidenced by their lower opacity. Such behaviour was ascribed to the inability of sheets dried without restraint to distribute an applied load evenly over all their structural units. An increase in the relative humidity at which the test is made has been shown by Houen<sup>(66)</sup> also to reduce the fraction of broken fibres in the rupture zone.

Changes in the bonds-During elongation, bond rupture first becomes appreciable as the yield point in the load/elongation curve is reached. Evidence for this is to be found from various experiments. Nordman<sup>(53)</sup> showed that sheet opacity increased sharply in this region. Kubát<sup>(67)</sup> found that electrical noise, absent in the elastic region, became intense after the yield point. The observations by Corte, Kallmes & Jarrot<sup>(68)</sup> of amplified noise from the sheet undergoing elongation would seem also to support the above, although in their experiments some noise-and therefore presumably bond breaking-generally occurred right from the start of elongation. The nature of bond failure in a strip that has been elongated to rupture has been studied in detail by Page, Tydeman & Hunt.<sup>(69)</sup> They found that about half of the bonds were not visibly affected by the elongation and that only 5 per cent of the total bonds were completely broken. Most of the bonds affected showed partial breakage equivalent to less than 50 per cent of their optically bonded area. Beating appeared not to affect this distribution of bond breakage. The mean size of the bonds that broke completely was 510  $\mu^2$ , whereas that for all bonds was 930  $\mu^2$ .

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For sheets dried under uni-directional tension, elongation in the direction of drying tension resulted in less bond breaking than elongation in the freeto-shrink direction. Moreover, it was estimated that the bonds broken in the free-to-shrink direction would be more likely to relieve stress in adjacent fibre segments than those broken during elongation in the tension direction.

It is suggested by these investigators that the high proportion of partial bond breakage found is due to a combination of factors, including a time dependence for complete bond breaking and the relief of stress on the bond brought about by changes in the neighbouring structure and in the bond itself.

Changes in general sheet structure—When paper is elongated, it contracts in width and generally increases in thickness. The ratios of these changes to the elongation of the sheet are known as Poisson's ratios. Few comprehensive studies have been made of these basic changes in sheet dimensions. For a range of commercial papers and handsheets, Brecht & Wanka<sup>(70)</sup> found values in the range 0.1-0.45 for Poisson's ratio in the plane of the sheet. It was independent of substance and, at a given elongation, decreased with increased beating, but increased with relative humidity and with tension applied during drying (in the direction of elongation). It tended to increase with the elongation at which it was measured, at least for handsheets. Ranger & Hopkins<sup>(11)</sup> reported Poisson's ratios of 0.95 and 0.64 for plate-dried beaten sulphite and sulphate handsheets, respectively; 0.78 and 0.40 for the machine- and cross-directions of a commercial sack kraft pulp. On the other hand, Rothman<sup>(71)</sup> found commercial kraft papers to give values in the range 0.2-0.5, the actual values depending in a rather complicated manner on the load at which they were measured.

Thickness increase on elongation has been reported by Maynard,<sup>(72)</sup> who viewed it as evidence that the paper structure starts to disintegrate before rupture occurs. Ranger & Hopkins<sup>(11)</sup> have shown that, for unbeaten and lightly beaten sheets, thickness increases right from the beginning of elongation. As beating degree increases, the rate of thickness increase falls, particularly at small elongations and, for the most highly beaten sheet tested, a small decrease was in fact observed up to an elongation of 1 per cent. Above this value, however, thickness again began to increase. It was suggested that this behaviour was caused by bond rupture and strain line formation from a very early stage in the case of the unbeaten and lightly beaten sheets, in keeping with the 'almost negligible initial straightline section of the load/ strain curves in such cases'. This implies that thickness increase during elongation should occur only in the plastic regime of the load/elongation curve. The thickness increases measured ranged up to 6 per cent.

Other general structural changes that occur in the sheet on elongation in

the plastic regime are those indicative of a decrease in interfibre bonding such as the increased ease with which a layer of fibres can be removed from the sheet surface,<sup>(11)</sup> an increase in opacity, already partly described in the section dealing with bond strength and energy and an increase in porosity and change in pore size distribution as demonstrated by Sanborn.<sup>(73)</sup>

Paper is far from being a uniform material, however, and many of the changes occurring during elongation reflect this character. Nordman<sup>(53)</sup> has found that opacity changes on elongation vary irregularly both with time in the one position and for different positions in the sheet. Ranger & Hopkins<sup>(11)</sup> have made careful measurements of elongation in different parts of a test strip by optical methods and found it to vary markedly from millimetre to millimetre. For a mean rupture elongation of 1.3 per cent, a range of values (0.2-4.7 per cent) was found. Additional evidence for the importance of a statistical component in rupture behaviour can be found in the work of Wink et al.,<sup>(10)</sup> already briefly referred to, which showed that the effect of test strip length on rupture properties could be explained by the existence of randomly distributed weak spots in the sheet. A direct measurement of the effect of substance variability on the course of sheet rupture has been made by Tydeman & Hiron,<sup>(74)</sup> who prepared point-to-point substance maps of test strips, which were then elongated to rupture. The paths followed by the rupture lines were found to follow approximately paths of minimum substance.

Perhaps the most remarkable structural change occurring during paper elongation is that of strain line formation. These lines of increased opacity appear at characteristic angles to the direction of load when well-beaten translucent sheets or wetted sheets are elongated in their post-yield regions. They have been described by Rance,<sup>(20)</sup> who likened them to the Luder's lines that are formed in the initial stages of the plastic region of the load/ elongation behaviour of metals. Ranger & Hopkins,<sup>(11)</sup> however, have disagreed with Rance's views and have developed a new theory for their origin, which will be discussed later.

#### THEORIES RELATING STRUCTURE AND MECHANICAL PROPERTIES

THE extensive experimental observations that have now been made on the behaviour of paper and of which only those relating to load/elongation properties have been summarised in the foregoing have given rise to a number of theories relating sheet structure to mechanical properties. Some of these theories, which stem from a variety of premises, will now be considered in three groups—

*I*. General theories that set out to explain the complete mechanical behaviour of paper or, at least, a considerable part of it.

2. Quantitative theories for the prediction of sheet elasticity from fibre properties. (These justify special treatment, because of the increased interest in them in recent years.)

3. Application of the Griffith crack theory to paper.

#### General theories

THE general theories can be classified according to the level of organisation considered to play the dominant role in determining mechanical behaviour; theories treating paper essentially as a molecular assemblage will thus be described separately from those whose starting point is a network of fibres and bonds.

In another class again is the rheological approach to the mechanical behaviour of paper. Ideally, this seeks only to interpret behaviour in terms of an analogue, without any necessary commitment about the nature of the physical units involved and is not therefore a theory relating the structure of paper to its mechanical properties. Nevertheless, the approach has structural implications justifying its inclusion.

**Theories based on paper as a molecular assemblage**—Nissan and coworkers<sup>(75,76)</sup> have developed a theory relating the mechanical behaviour of paper to the properties of its hydrogen bonds. It assumes that no distinction is necessary between intra- and interfibre hydrogen bonds and considers the sheet not as an arrangement of bonded fibres, but as a hydrogen-bonded continuum.

Using the Morse function to describe the potential energy of the hydrogen bond, a relationship is derived expressing stress as a function of strain, elastic modulus of the sheet and a second 'coefficient of elasticity'. The last two can be calculated from the constants in the Morse function for the hydrogen bond, an assumed hydrogen bond energy and the number of hydrogen bonds per unit volume taking part in the straining process. Different relationships are obtained, depending upon whether a one-dimensional or three-dimensional hydrogen bond model is used: these are believed to represent different limiting cases, with the behaviour of paper falling between them.

The theory predicts that, at vanishing strains, the elastic modulus of paper should be proportional to the cube root of the number of hydrogen bonds per unit volume taking part in straining and that rupture energy should be linearly related to the same parameter. A strong criticism of the theory has recently been made by Page.<sup>(77)</sup>

The mechanical behaviour of paper at high beating degrees has also been regarded as that of a typical hydrogen-bonded solid by McKenzie,<sup>(19)</sup> who found that the elastic moduli of papers made from a number of different pulps all reached the same limiting value on beating, namely, about  $1.0 \times$ 

 $10^{11}$  dyn/cm<sup>2</sup>, which is similar to the elastic modulus for ice crystals, but lower than that for single cellulose fibres. Post-yield slopes for the pulps also tended to a common limiting value at the same beating degrees, for which limiting values were obtained for the elastic moduli.

Theories based on paper as a fibrous network—Rance<sup>(20,4)</sup> has proposed a comprehensive theory of paper strength that centres around the strength of the interfibre bonds and the distribution of stress in the sheet. The theory states that, when a sheet of medium density is elongated, fibres that are bent or curved are straightened out and are then themselves elongated, though not beyond their elastic limit. Interfibre bonds are considered to be weaker than the fibres and, as elongation proceeds, bond rupture by shear begins to occur at an early stage at points where stress is concentrated. This process is seen to lead to a progressive decrease in the load-carrying section of the sheet, with a resultant increase in the stress concentration on those units that are presumably still firmly bonded in the network. The process is therefore one of 'progressive stress concentration in a disintegrating medium', with final fracture occurring at a specific state of disintegration. Similar mechanisms apply in creep and stress relaxation. In the latter case, bond rupture reduces the total load, but, as the load-bearing section is simultaneously reduced, the stress on it remains substantially constant. Structural dislocation and progressive stress concentration are also held to explain the phenomena of permanent set and strain hardening, but the concept of frictional drag between the fibres is introduced to account for the time-dependence of reversible deformation.

Rance developed this picture from the following experimental evidence-

*I*. The non-linearity of the relationship between total elastic deformation and relaxed load or dead load in stress relaxation experiments or creep experiments, respectively.

2. The absence of asymptotic or equilibrium values in creep and stress relaxation.

3. The constancy of rupture elongation over an extremely wide range of elongation rates in both creep and load/elongation measurements.

Rance suggested that an appropriate rheological model might be constructed on the basis of this theory and called for the development of a 'mathematical statement' to define the structure of paper so that its interrelationships with rheology could be fully explored.

A similar need was also seen by Van den Akker<sup>(56)</sup> when he advocated the development of some special sort of statistics to calculate the stress and strain in the fibres and shear stress in the interfibre bonds. These ideas culminated in his presentation at the 1961 Oxford symposium of a quantitative structural theory for paper elasticity and a descriptive structural interpreta-

tion of paper behaviour in the plastic regime.<sup>(78)</sup> His elasticity theory sets out from the assumption that the web is 'dried under restraint so that the various forces in a fibre element come into existence with the initial infinitesimal straining of the whole web and are linear functions of the strain to which the web is subjected'. Fibre characteristics are described by distribution functions; sheet deformation is assumed to occur by the elongation, bending and shear of fibre segments and by the angular displacement of bonds. A more detailed account of this theory is given later.

The plastic regime is considered to be signified by the onset of bond breaking, generally starting at a strain of about 0.005. Bond breaking, intrafibre creep—and, possibly, fibre failure at higher strains—continue until ultimately the sheet is so weakened that rupture occurs.

In the plastic range, the bonds are considered to be stressed by a combination of the anisotropic shrinkage forces, owing to the fibres themselves and either torque or tension forces arising from the strain in the sheet. These stress combinations are shown to result in high shearing forces at the bond peripheries. As strain increases, the fibres nearly parallel to the direction of strain contribute most to the load and are under substantial tension. It is shown that the end bonds on such fibres should break first, after which only a slight additional strain will suffice to cause the rest to rupture, the fibre then ceasing to contribute to the load. This mechanism results in a decreasing slope to the load/elongation curve, a decrease in the effective mass carrying the load and a change in the angular distribution function of the fibres. Torque failures also occur, but it is not until higher strain that they have an important effect on load.

Van den Akker emphasises that a reasonably accurate mathematical theory for the plastic regime would be very involved and should include appropriate treatment of the variability in bond and fibre strength. He draws attention to the unreal nature of his model in that it assumes that each fibre segment is of such configuration that it has its tension (or compression) and flexural and shear stress all linearly related to sheet strain.

In the theories of Rance and Van den Akker, bond rupture during elongation of the sheet is considered to occur largely through shear and tension. Bond rupture by a peeling action, however, is the central point in the theory of Ranger & Hopkins,<sup>(11)</sup> who set out to explain the tensile behaviour of paper by considering the significance of the width decrease and thickness increase that occur on elongation. They believe that the contraction in width of the test strip results in some fibres becoming compressed, then buckling. Unless the sheet is very dense, this is seen to lead to rupture of the bonds between these fibres and other fibres held in tension by a peeling action that requires only low stress. Such a mechanism is believed to be the main mode of bond rupture in the sheet and to account for the opacity increase observed in the plastic region of the load/elongation curve.

If, in a local area where such bond rupture occurs, there is also shear or slip between adjacent areas of the sheet, neighbouring fibres are likely to become more compressed and so a self-propagating mechanism is started. It can be shown mathematically that this is most likely to occur along diagonal lines, whose direction can be calculated. These lines of bond rupture, therefore increased opacity, are said to be identical with the strain lines that can be observed in many papers under load.

Propagation of a strain line started in this manner continues until it is stopped by fibres in tension; as stress continues to rise, the process then starts elsewhere and results in the observed pattern of intersecting lines. For strong papers, an extensive pattern can be formed, but, as fibre bonding is low in weak papers, there is less likelihood that fibres in tension are able to terminate a strain line. Thus, in such papers, it is possible for complete rupture of the strip to occur along one strain line, a phenomenon that can be readily observed.

Reduced strain line formation should be accompanied by an increase in rupture elongation, which would then tend to be distributed evenly over the whole strip. This effect might be expected to operate in the machine-direction of a machine-made paper, because it would contain more fibres oriented in the tension direction, therefore able to terminate strain lines and in a shorter interbond fibre length in the cross-direction, which would reduce buckling tendency. This is generally not the case, however, unless the interacting effect of drying tensions is eliminated, as in the experiments of Toroi,<sup>(15)</sup> in which this expected higher machine-direction elongation was observed.

Strain line formation is postulated to occur also in the sheet under the influence of the machine-direction drying tension operating on the papermachine. A pre-existing strain line pattern of this type is believed to reduce rupture load in any subsequent elongation in a direction at rightangles to the original tension direction.

The theory explains the permanent set of paper as a uni-directional frictional effect that arises when tension on the strip is relaxed and the fibre 'mats' on the two sides of the strain line move together again under the influence of the fibres in tension. It cannot explain, however, the behaviour of paper dried without tension. Strain lines are not found in such paper and, moreover, there is some evidence that thickness may in fact decrease when it is strained. It is suggested that this behaviour may be due to a slackness in the fibres, reducing the possibility of bond breaking by peeling, which is a prerequisite for strain line formation.

The shrinkage properties of the fibres are seen by Page & Tydeman<sup>(47)</sup> to

play a dominant role in the structure/strength relationship for paper. From microscopical evidence that showed that the longitudinal shortening of the fibres in a sheet dried without tension was the same as that of the sheet itself and an order of magnitude higher than that of the individual fibres, they concluded that a longitudinal contraction of the crossing fibres takes place at the bond. This is caused by the strong transverse shrinkage of one fibre acting on the other and assumes of course that bonding between the two has already commenced while the fibres are still able to yield to the contracting forces. This longitudinal contraction at the bonds (microcompression) is equal to the contraction of the sheet as a whole and is accompanied by an equal contraction in the length of the fibre segments. Kinks are formed in long segments and microcompressions in the shorter ones.

Such a structure may be expected to have a lower elastic modulus than that for a tension-dried sheet, in which such microcompressions are not apparent. This was demonstrated by a model load/elongation experiment, using strips of brass of constant cross-section, but kinked to various degrees. Kinking was found to decrease elastic modulus, but not to affect the linearity of the initial part of the curve. It also caused the yield point to occur at an earlier stage, presumably because of the introduced stress concentrations.

In the elongation of a sheet dried without tension, it was suggested that the microcompressions at the bonds tend to be pulled out, leading eventually to partial rupture at the perimeter of the interfibre bonded area. This rupture generally does not proceed to completion, but the microcompressions are released as it takes place, thereby increasing elongation and permanent set. It follows from this suggested mechanism that the load/elongation behaviour will be influenced to a large extent by bond strength, in particular in the post-yield range.

In the case of a sheet dried under uni-directional tension, microcompressions are not formed in the direction of the drying tension. When such a sheet is elongated in this direction, fewer bonds are found to break than if the elongation were done in the cross-direction. This behaviour is said to be explained by the above theory, presumably because the absence of microcompressions at such bonds would imply a more uniform stress concentration, therefore a higher rupture strength.

A special characteristic of this theory is that it stresses the importance of the behaviour of the bonded parts of the fibres when the sheet is being elongated. This follows logically, of course, from the earlier observation of these workers that in most papers there is very little fibre length that is not bonded on one side or the other. The theory therefore indicates that not only the longitudinal, but also the transverse Young's moduli of the fibres should be of importance in determining load/elongation behaviour of the sheet.

#### Effect of structure on mechanical properties

In an early publication. Van den Akker<sup>(56)</sup> drew attention to the heterogeneous nature of the fibre properties in a sheet and to the likely existence of a 'relative tightness and looseness' among the free fibre segments, which he believed to be determined to some extent by the degree of stress equalisation occurring during drving. Related to this theme is the theory of bond formation proposed by Craven<sup>(2, 12)</sup> from studies on the effects of drying tensions on the load/elongation curve and on stress relaxation. This theory states that, for any given tension in the drying sheet, there exists a critical moisture content, above which interfibre bonds are not formed. An increase in drving tension at this moisture content is believed to result in the pulling apart against surface tension forces of fibres that otherwise would have formed a bond. Thus, the higher the tension applied during drying, the lower the moisture level at which bonds begin to form and the narrower the range of moisture contents over which the formation takes place. High drving tension can therefore be thought to produce interfibre bonds that are under more uniform conditions of frozen-in stress than those occurring in a sheet dried free to shrink. In the latter case, the sheet would contain some bonds having zero stress, also some in compression.

When a tension-dried sheet is initially strained, therefore, the initial slope of its load/elongation curve is greater than that for the sheet free to shrink, as more of its bonds share the load. Beyond the elastic limit, bonds are imagined to break in reverse order to their order of formation. Thus, in the tensiondried sheet, nearly all the bonds in the rupture zone are imagined to fail together, resulting in the high rupture load and low elongation characteristic of such sheets.

Stress relaxation tests showed that for a wide range of papers relaxation rate was higher for sheets dried under light tension than for those dried under heavy tension: in comparison, degree of beating and pulp type had insignificant effects. As a sheet with a wide variation in bond stress would be expected to relax faster than a sheet in which stress was shared evenly by all bonds, this constitutes additional support for the proposed theory.

Complementary to this picture of the effect of drying tension on bond heterogeneity is the contribution of Majewski,<sup>(79)</sup> who partly dried sheets without tension to various predetermined lengths, then measured their developed stresses during the remainder of drying. Load/elongation curves on the dried strips showed that the behaviour of the paper was elastic only up to the maximum level of stress developed during drying, indicating a correspondence between the bonds initially taking the load during elongation and those formed in the final part of drying, during which strip length was held constant.

A somewhat similar view to that of Craven was offered by Schulz<sup>(13)</sup> after studies on the effects of strain applied during drying on the load/elongation

and creep properties of paper. With increasing drying strain, creep rate passed through a minimum, rupture load and elastic modulus through corresponding maxima. This behaviour was ascribed to a structural change in the sheet caused by the increasing drying strain and requiring more of the 'elements' in the sheet to share the load. The hypothesis was put forward that this could be the sliding of fibres over each other in the wet sheet to more effective relative positions, though this would occur at the expense of interfibre bonding. The observed maxima in rupture load and elastic modulus were thus the net effects of two opposing mechanisms, one increasing uniformity of load distribution, the other decreasing interfibre bonding.

An interpretation of many aspects of the load/elongation behaviour of paper in terms of active and passive fibre segments has been suggested by Giertz.<sup>(18,80)</sup> Active segments are defined as those that immediately share the load when paper is first elongated; thus, the larger the number of active segments, the higher the elastic modulus of the paper.

Segments are considered to become active if, during the drying process, they are straightened out under the influence of the transverse shrinkage of the crossing fibres at the interfibre bonds. In accordance with the ideas of Page & Tydeman, therefore, the number of active segments will be high in a sheet dried under restraint, low in a sheet dried free to shrink. A further reason that some fibre segments may not be active is the finding by Jentzen that fibres extend on drying under tension (see the section on drying the fibre under stress).

An unbeaten sheet has a low elastic modulus, because it has a low proportion of active segments, owing to its low degree of interfibre bonding. As the number of interfibre bonds is increased by beating or pressing, the segments will become shorter and transverse fibre shrinkage at the bonds will have a proportionally larger stretching effect on them. The number of active segments therefore increases and, with it, the elastic modulus. Moreover, with beating, the effect is enhanced because of the increased transverse fibre shrinkage at the bond. The increases in modulus occurring on elongation/contraction cycling or on rewetting and drying under restraint are also both ascribed to increases in the proportion of active segments in the sheet.

The concept can also be employed to explain the behaviour of the high stretch papers made by, for example, the Clupak process and the liquid ammonia process of Arlov & Snaprud.<sup>(81)</sup> In both cases, microscopical examination shows the presence of many kinked fibre segments, indicating a low proportion of active segments.

The rheological approach—The outstanding contribution in this field was made by Steenberg and his school, who introduced rheological method for the interpretation of the mechanical properties of paper.<sup>(82, 83)</sup> A consequence of this approach was the adoption of the Eyring visco-elastic model and a subsequent modification to serve as approximate analogies for the behaviour of paper. A physical correspondence between the models and the way in which various linked structural elements might behave in the sheet was at one time suggested,<sup>(84)</sup> but, apart from this, no direct structural meaning was attributed to them, though their acceptance did imply certain interpretative consequences—for example, that some type of flow occurred when paper was elongated and that this could be characterised by an apparent viscosity and the volume of a flowing unit calculated.<sup>(85)</sup>

The Steenberg school has not elaborated a comprehensive theory relating mechanical properties to sheet structure. Their interpretations of various phenomena generally relate back to the molecular and micellar levels rather than to the structural level at which fibres and bonds are the basic entities; they thereby imply that it is the properties of the material rather than those of the fibre network that determine mechanical behaviour. Ivarsson,<sup>(85)</sup> for example, has suggested that the mechanical conditioning of paper might be due to an organisation of 'crystalline elements'. Stress relaxation and fatigue in paper are also implicitly associated with intrafibre processes in recent papers by Kubát and co-workers.<sup>(1,3)</sup>

Moreover, Steenberg<sup>(86)</sup> has suggested the presence of longitudinal microcompressions in fibres to explain the permanent set introduced when a sheet is first elongated, thus partly anticipating the findings of Page & Tydeman.<sup>(47)</sup>

#### Quantitative theories for the prediction of sheet elasticity

OVER the past 14 years, several quantitative theories have been proposed for the prediction of sheet elasticity from the structural properties of the sheet and the elastic properties of its fibres. These assume various structural models for the sheet and consider its elastic deformation to be the resultant effect of the deformation of the individual fibres or fibre segments caused by various combinations of extension, shear, bending and interfibre bond rotation.

The first of these theories published was that of Hurley,<sup>(87)</sup> who developed simple equations relating sheet elasticity to sheet density and fibre elasticity. As the mathematical expression derived required correction factors to give even moderate agreement with experiment and as it was based on a model treating the sheet as a regular grid, it will not be considered further, except to note that it did take into account the separate response of the bonded parts of the fibres, as well as the unbonded segments, when calculating the general response of the sheet to an applied load. This aspect has been considered in none of the subsequent theories, except the two most recent ones.

#### Effect of structure on mechanical properties

The other theories have considered paper generally as a random network of randomly oriented fibres, although some include provision for the effect of preferential fibre orientation. In chronological order, the theories are those of Cox 1952,<sup>(88)</sup> Le Cacheux 1953,<sup>(89)</sup> Onogi & Sasaguri 1957,<sup>(90)</sup> Litt 1961,<sup>(91)</sup> Van den Akker 1961,<sup>(78)</sup> Kallmes & Bernier 1961,<sup>(22)</sup> Campbell 1963,<sup>(92)</sup> Kallmes, Stockel & Bernier 1963,<sup>(26)</sup> and Page 1963.<sup>(93)</sup> The earlier theory of Kallmes & Bernier was modified in the paper of Kallmes, Stockel & Bernier and will be denoted by Kallmes I. This paper also includes a modification of the theory of Van den Akker, denoted as Kallmes II. Page has given two alternative versions of his theory, denoted as Page I and II.

Assumptions made in all the theories, sometimes implicitly, are that-

*I*. The whole sheet responds immediately and uniformly on the initial infinitesimal application of an external load.

2. Fibres lie substantially in the plane of the sheet (three-dimensional treatments are also given by Cox and Onogi & Sasaguri).

3. No interfibre bonds are ruptured in the range in which the paper is exhibiting elastic behaviour.

Other major assumptions that vary from theory to theory and form the basis of a useful classification are that—

4. The strain of a fibre segment is the same as that of the sheet in its immediate neighbourhood. Theories making this assumption have been sometimes called *uniform strain* theories, but the designation *linear network* theory\* is more appropriate—Cox, Le Cacheux, Campbell, Van den Akker, Kallmes II, Page II.

5. The force on all fibre elements is uniform throughout the sheet—Onogi & Sasaguri, Litt, Kallmes I, Page I.

6. The strain energy in the bonded parts of the fibres can be distinguished from that in the unbonded segments—Page I and II, Kallmes I and II.

The various modes of fibre segment deformation considered in the different theories can be summarised as follows—

7. Extension only—Cox, Le Cacheux, Campbell, Page (for the special case of equal biaxial strain).

8. Extension and bending-Onogi & Sasaguri.

9. Extension and shear-Litt.

10. Extension, shear and bending-Kallmes I.

11. Extension, shear, bending and bond rotation-Van den Akker, Kallmes II.

The various theories will now each be briefly described. In Table 1, some of the derived expressions for Young's modulus have been set out in a

\* The author is indebted to Dr J. A. Van den Akker for suggesting this designation 14—c.p.w. II

common form to facilitate comparisons. A basic similarity among the theories in each class can be readily seen.

Author	Type of theory	Expression for Young's modulus (Y)
Cox	Linear network	$\frac{1}{3} \cdot \frac{D}{d} \cdot E$
Le Cacheux	Linear network	$\frac{1}{3}\left(1-\frac{2g}{L}\right)\cdot\frac{D}{d}\cdot E$
Campbell	Linear network	$\frac{1}{3} \cdot \frac{D}{d} \cdot E$
Onogi & Sasaguri	Uniform force	$\left \frac{8}{\pi^2}\left[\frac{1}{g^2/3r^2+1}\right]\frac{D}{d}\cdot E\right $
Litt	Uniform force	$\frac{8}{\pi^2} \left[ \frac{1}{g^2/w^2 + 1} \right] \frac{D}{d} \cdot E$
Kallmes I	Uniform force	$\frac{8}{\pi^2} \left[ \frac{2IG}{aGg^{*2} + 2EI + 2GI} \right] \frac{D}{d\tau} \cdot E$
Van den Akker	Linear network	$\frac{1}{3} \left[ 1 + \left[ \frac{4IG}{aGg^2 + 12EI + 2GI} \right] \frac{D}{d} \cdot E \right]$
Kallmes II	Linear network	$\frac{1}{3} \left[ 1 + \frac{16IG}{3aGg^{*2} + 36EI + 8GI} \right] \frac{D}{d\tau} \cdot E$

 
 TABLE 1—EXPRESSIONS FOR YOUNG'S MODULUS OF PAPER DERIVED IN VARIOUS QUANTITATIVE THEORIES FOR PAPER ELASTICITY

a = cross-section area of fibre wall (cm<sup>2</sup>)

 $d = \text{density of fibre wall } (g/cm^3)$ 

- $D = \text{sheet density } (g/cm^3)$
- E =Young's modulus of the fibre (dyn/cm<sup>2</sup>)
- g = fibre segment length (cm)—distance between mid-points of adjacent bonds
- $g^* =$  unbonded fibre segment length (cm)
- G = modulus of rigidity of the fibre for shear stress along cross-section and parallel to the shearing force (dyn/cm<sup>2</sup>)
- $I = \text{moment of inertia of fibre cross-section about a neutral axis in the z-direc$ tion (cm<sup>4</sup>)
- L = fibre length (cm)
- r =fibre radius for circular fibres (cm)
- w = fibre width for rectangular fibres (cm)
- Y = Young's modulus of the sheet (dyn/cm<sup>2</sup>)
- $\tau = \text{fibre curl}$

Notes: 1. These equations are not necessarily as published, all having been reduced as far as possible to a comparable form

- 2. Average values are implied for fibre properties
- 3. All equations refer to planar mats

Table 1 excludes the expressions for three-dimensional fibre orientation derived by Cox and by Onogi & Sasaguri, as well as the distribution functions for fibre properties that are included in the general forms of the expressions derived by Onogi & Sasaguri, Van den Akker and Kallmes. Instead, average values are used for the sake of simplified presentation.

#### Uniform strain theories of Cox, Le Cacheux and Campbell

The purpose of Cox's study was primarily to determine how the elasticity of paper and other materials depends on the orientation of the fibres. The model used comprised a homogeneous plane or solid mat of long, thin, straight fibres, loaded at their ends, having negligible flexural stiffness, with no fibres in compression. When the load was applied, the fibres were assumed to extend in straight lines.

The elastic behaviour of these bodies was derived by considering the application of biaxial tensile strains at rightangles. Young's modulus, shear modulus and Poisson's ratio for both planar and solid isotropic mats were calculated. The effects of various fibre orientation distributions were determined for the planar mat only. Moderately good agreement with experimental results was found, when the theory was applied to a resin-bonded board.

The assumptions of Le Cacheux differed in only minor aspects from those of Cox. Fibre segments, rather than whole fibres, were considered and the strain in these was assumed to be the same as that of the sheet in their immediate neighbourhood. His expression for Young's modulus was derived by first calculating the force in one segment, then summing the number of segments in unit cross-section of an isotropic sheet.

The expression derived for Young's modulus is identical with that of Cox, except that it contains the term (1-2g/L) to account for the free ends of fibres that do not contribute to the developed tension. The magnitude of this effect is generally small enough to ignore. Le Cacheux found his theory predicted approximately the Young's modulus of paper made from ramie fibres of known Young's modulus.

Campbell's approach was essentially the same as those of Cox and Le Cacheux and led to the same expressions for Poisson's ratio  $(\frac{1}{3})$  and Young's modulus. In his paper, however, Campbell used extensional stiffness instead of Young's modulus, as it avoids the uncertainty of sheet thickness determination and is, he suggests, a more fundamental paper property. (Extensional stiffness is the product of Young's modulus of the sheet and its thickness.)

Using the experimental values of Kallmes & Bernier<sup>(22)</sup> for single fibre Young's modulus, Campbell's calculated values for sheet extensional stiffness were higher than those found experimentally, a discrepancy ascribed by him to the fact that the pulp used was not beaten. He stressed that his theory is valid only when a certain level of interfibre bonding has been reached. The extensional stiffness—430 kgf/cm ( $60 \text{ g/m}^2$  sheet)—predicted by his equation was therefore to be regarded as a maximum limiting value.

Campbell collected the reported values of limiting extensional stiffness from a number of investigations and found that, for a wide range of chemical woodpulps, they all lay closely about 400 kgf/cm (60 g/m<sup>2</sup> sheet). From this observation, he concluded that limiting extensional stiffness may, as an ultimate simplification, be considered as merely proportional to the substance of the sheet.

# Uniform force theories of Onogi & Sasaguri, Litt and Kallmes, Stockel & Bernier

The uniform force theory of Onogi & Sasaguri assumed that the number of interfibre bonds in the sheet is large enough for the uniform transmission of an external force and that the resultant deformation is uniform. They derived an expression for sheet elasticity by considering initially the deformation of a straight segment by extension and bending. After introducing distribution functions for segment length, fibre cross-sectional area (assumed circular), fibre moment of inertia and angular orientation in the plane of the sheet, the deformation of the sheet was found, thus the Young's modulus. The expression was simplified by introducing average values for fibre properties and a uniform planar distribution for segment orientation. When this was combined with the expression derived for sheet density, it resulted in that given in Table 1. The simplified form was used by the authors to derive further expressions for Young's modulus, assuming various z-direction segment orientation distributions. The theory predicts that the Young's modulus of the sheet should also be proportional to the third power of the sheet density. Experimental results presented gave power values ranging 2.0 - 2.8.

Litt's uniform force theory assumed that the fibre segments are deformed by axial extension and shear. The segments were considered to be of flat, rectangular cross-section, with 'no built-up stress in the network, as happens when paper is dried under tension'.

As can be seen from Table 1, the expression derived for Young's modulus, which is very similar to that of Onogi & Sasaguri, requires a value for the fibre segment length. By suitable choice of an empirical relationship between segment length and sheet density, good agreement was obtained between the derived expression and experimental curves relating Young's modulus of a sheet to its density, at least above a certain minimum sheet density. Litt also suggested that his theory may be applicable in a semi-quantitative way to the post-yield region and gave examples of its use in calculating the energy loss per unit area involved in bond rupture.

The Kallmes, Stockel & Bernier modification of the earlier uniform force theory of Kallmes & Bernier appears to have been made partly because of criticism by Tydeman & Hiron<sup>(94)</sup> of the assumption that all the strain energy in the sheet resides in the fibre segments and none in the bonded parts of the fibres. Strong justification for this criticism is found in the experimental finding that very little fibre length exists in most papers that is not bonded on one side or the other.<sup>(43)</sup>

In its modified form, the theory considers that the strain in a fibre, arising when a force is applied to the sheet, is separable into two parts—one applying uniformly throughout the whole fibre length and equal to what would be developed if the fibre were in a completely bonded sheet, the other being an additional strain, applying only in the unbonded parts. The first of these is believed to be a fibre property, the second a property both of the fibre and of the sheet geometry. A general expression is then derived for the elasticity of the sheet, incorporating the different strain behaviours of the bonded and unbonded regions.

Thereafter, the theory follows a parallel development to the earlier theory of Kallmes & Bernier. In both theories, a segment is considered as the length of fibre between the mid-points of two adjacent fibre crossings; whereas its bonded length was ignored in the earlier theory, it has been taken into account in this modification. The average total deformation of this segment is calculated as the sum of the contributions made by extension, shear and bending. An expression is then derived for the deformation of a chain of such segments, extending from one clamp to the other in a strip of very thin (2-D) sheet, imagined to be held in a straining device. The total strain in the sheet is thus obtained and, to determine the total force acting on the clamps, the number of fibres held in each is calculated. The foregoing assumes that each fibre at a clamp line is the end of a segment chain and that all such chains transmit force and deform in the same manner.

Young's modulus is calculated from the total strain and the total force on the clamps, thence the contributions of both the bonded and unbonded parts of the fibres to sheet strain. This theory will be discussed further in connection with the Kallmes, Stockel & Bernier modification of Van den Akker's linear network theory.

### Linear network theory of Van den Akker and its modification by Kallmes, Stockel & Bernier

This theory includes bond rotation as a source of segment deformation,

along with extension, shear and bending. Other assumptions not already mentioned are that the fibres are ribbon-like and flat, that the angle between a segment and a crossing fibre is constant up to the onset of failure, that fibre segments subjected to axial compressive stress do not buckle and that the segments are initially straight. Van den Akker first calculates the contribution to the load on the sheet made by one segment, then introduces the number of segments per unit area in the sheet to obtain the total load. Distribution functions are included for segment length, segment orientation, fibre cross-section area and its moment of inertia about a neutral axis in the z-direction. From the derived tensile force in each direction of the sheet Poisson's ratio is calculated, thence Young's modulus and the modulus of rigidity of the sheet for shear strain in the xy-plane.

The modification of this theory by Kallmes, Stockel & Bernier again takes into account the relative contributions of the bonded and unbonded fibre lengths to the total strain of the sheet. The expression derived for Young's modulus (given in simplified form in Table 1) can be seen to be quite similar to Van den Akker's original expression, except for the introduction of unbonded segment length.

This modified theory and the modified uniform force theory were compared with one another and with experimental results by considering the curves obtained when the so-called modulus ratio was plotted against relative bonded area. Modulus ratio is defined as  $Y\tau\gamma/WE$ , where  $\gamma$ =cellulose density, W=sheet substance and the other symbols are as in Table 1. The theoretical curves are calculated from relationships established earlier between segment length and RBA.

It was found that these two theories gave quite different results, the linear network theory giving the higher modulus ratio over the whole RBA range with the difference between the two becoming less as RBA increased. The experimental results lay between the two somewhat closer to the linear network theory curve and on a line that met the latter at 100 per cent RBA.

Calculation of the relative contributions of the bonded and unbonded fibre lengths to total strain for the linear network theory showed that the contribution of the unbonded length was only slightly larger than its occurrence. This was taken to mean that bending, postulated to be the only mode of deformation taking place solely in the unbonded lengths, appears to be of relatively minor importance, in contrast to the earlier uniform force theory of Kallmes & Bernier, from which bending was calculated to be the major contributor (about 75 per cent) to sheet strain, with the minor contributions being made by shear and extension. In this new study, however, bond rotation (not considered earlier) and shear 'appear to contribute significantly'.

#### Effect of structure on mechanical properties

#### The theory of Page

For simplicity, Page considered the case of a sheet under equal biaxial strain, a condition that he assumed results in all fibres extending to an amount equivalent to this strain, without shear or bending occurring. He derived an expression for the biaxial modulus of an isotropic sheet by equating its strain energy to that of the sum of the strain energies of the unbonded and bonded fibre regions, respectively.

Assuming that for two bonded isotropic fibres the longitudinal stress in one is shared equally with a crossing fibre in the bonded region, he then established the following relationships between the Young's modulus and Poisson's ratio for the fibres and those for the sheet for conditions both of constant strain in the sheet and of constant stress—

Constant strain 
$$\frac{Y'}{1-\nu_p} = \frac{E}{2} \left[ \frac{A(1+\nu_f)}{(1-\nu_f)} + 1 \right]$$
  
Constant stress  $\frac{Y'}{1-\nu_p} = \frac{E}{2-A(1+\nu_f)}$ 

where Y' = Young's modulus of the paper, in force per unit area fibre crosssection

 $\nu_p$  = Poisson's ratio for the sheet

 $v_f$  = Poisson's ratio for the fibres

E = Young's modulus of the fibre

A = the proportion of fibre volume that is in biaxial strain

(As these expressions cannot be compared directly with those derived from the other theories, they are excluded in Table 1.)

Page also indicated how the effect of fibre anisotropy could be taken into account and suggested that it might not be so very large.

The theory was tested by measuring the strain of sheets loaded equally in four directions at rightangles. From such measurements, the ratio  $(1 - \nu_p)/Y'$  can be calculated directly. A plot of this ratio against A, determined experimentally, approximated to a straight line as required by the theory. Fibre Young's modulus calculated from the intercept of this line was found to be  $30 \times 10^{10}$  dyn/cm<sup>2</sup>, in good agreement with published experimental values. Page believed that these results, although insufficient for verification of the theory, were 'certainly encouraging'.

#### Application of the Griffith crack theory to paper

INTERPRETATION of the rupture behaviour of paper in terms of the Griffith crack theory has been attempted by Nissan<sup>(95)</sup> and Balodis.<sup>(96)</sup> In this theory, final rupture is supposed to be initiated by the occurrence of small

structural defects, originally present in the material and that grow in size as deformation of the sample takes place. The theory, which predicts that the rupture stress of an essentially elastic material will be proportional to the square root of its elastic modulus, has been found valid for the brittle fracture of numerous materials and, in a modified form, for the ductile fracture of metals.

From a wide range of load/elongation data, Nissan found that rupture stress of paper was proportional to the elastic modulus to the power of 1.2 only and concluded that the theory did not apply.

Balodis measured the load/elongation properties of paper strips in which small cuts had been made and, from the theory, calculated fracture energy per unit area of new surface created in rupture. Contrary to the requirements of the theory, this was found to vary somewhat with the length and position of the cut, behaviour ascribed by Balodis to the partial plastic nature of the paper. Nevertheless, fracture energy per unit area of new surface was shown to be related in a general way to elastic modulus for a range of papers and Balodis concluded that, to a first order approximation, the Griffith crack theory does apply to paper, provided the initial cut length exceeds the length of the structural elements of the material. He suggested, moreover, that Nissan's findings need not be inconsistent with the theory, as the relationship tested by him implies constant fracture energy for all papers, a condition that Balodis demonstrated did not apply.

#### CONCLUDING REMARKS

THE MAIN purpose of this review has been to present in systematic fashion some of the essential features of the extensive experimental observations that have now been made on the structure of paper and its mechanical behaviour and to describe some of the theories that have been proposed to relate these to one another. A critical review of this whole large field, although desirable, is not attempted here; instead, these concluding remarks will be confined to certain aspects of the theories just outlined.

Each in their explicit form, none of the theories yet proposed appears adequate for the satisfactory interpretation of all the experimental data that has been collected in this field. Of those based on the fibrous network structure as the significant level of organisation, Rance's theory is the most comprehensive and, at least up to the 1961 symposium, was probably the most widely accepted, particularly because of the support given it by Nordman's ingenious experiments relating mechanical behaviour to apparent bond breaking.

Considering first the plastic regime, it is clear that our knowledge of the

interfibre bond and its rupture mechanisms has now been extended well past the relatively simple requirements of the Rance theory, which was more concerned with the consequence of bond rupture than its mechanism. The new theories relating the strength and structure of paper, which have arisen as a result of this new knowledge, are perhaps understandably rather diverse in nature; moreover, in some cases, their range of applicability is limited or uncertain. A first task then is to explore them further, to determine to what extent they may be complementary and to bring about whatever unification may be possible. Notwithstanding this eventuality, however, it is likely that the final picture will not be a simple one.

Among the quantitative theories for predicting the elastic modulus of paper, it is unfortunate that more work has not yet been done with the equal biaxial strain approach of Page, as the initial results (though few) are promising and the technique used eliminates some of the structural complications that arise in normal uniaxial strain. Moreover, it includes consideration of the elastic moduli in both directions of the fibre, which would appear to be clearly demanded by the present state of knowledge of the interfibre bond and the high proportion of bonded area in a normal sheet.

Of the other theories, although most predict Young's modulus to within an order of magnitude, none gives close agreement with experiment at low beating degrees. As the interfibre bonding in the sheet is increased, however, the linear network theories of Campbell and of Kallmes, Stockel & Bernier (which tend to become identical) both give predictions that approximate to the experimental results.

A prerequisite for the satisfactory development of quantitative relationships between structure and mechanical properties would appear to be the ability to describe the structure of a sheet in an exact manner. Kallmes and his group have already shown how this can be done in the case of an ideal sheet from measurements of relative bonded area and fibre width. In the case of real paper, however, an important further factor to enter is the way in which a sheet distributes an applied load between its constituent elements, be they fibre segments, interfibre bonds or even fibrils or molecules. This has long been recognised as important in determining mechanical behaviour and has been considered again more thoroughly in recent studies. Giertz views the distribution of load between fibre segments as the central point in a theory of paper elasticity and Craven and Schulz have provided evidence through stress relaxation and creep measurements that increased strain on the sheet during drving tends to a more uniform load distribution. Ideally, one should be able to define and measure the ability of a sheet to distribute load, as this property is just as important in determining many aspects of the strength of paper as is the degree of interfibre bonding.

These considerations are very relevant in the quantitative theories for paper elasticity. These theories should show the best agreement with observation for sheets that have been strained during drying to give maximum elastic modulus, a condition approaching uniform load distribution in the sheet. It seems doubtful if the usual practice of drying without any dimensional change will be adequate to attain this maximum, as Majewski and Schulz, for example, have both shown that elastic modulus continues to increase with actual extension during drying. Experimental values from sheets dried to maximum Young's modulus may particularly improve the agreement with predicted values at low beating degrees, since the low shrinkage forces occurring during the normal drying of such sheets may be insufficient to bring about the structural changes required for uniform load distribution.

Even so, the manner in which improved load distribution is brought about by tension drying is not clear. As Giertz suggests, the effect may be one of straightening out segments and rendering more of them active; on the other hand, following Jentzen, it may be essentially an intrafibre mechanism, resulting in a more effective utilisation of the substance of segments already active. Whatever the mechanism, the effect appears to justify further consideration in sheet elasticity theories, as too does Jentzen's finding that the elastic modulus of pulp fibres is markedly affected by the tension under which they are dried and his advice that, for meaningful comparison with corresponding sheet properties, it should be therefore measured only after the fibres have been subjected to the same drying tension that applies in the sheet.

#### Acknowledgement

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#### References

- 1. Kubát, J., Nyborg, L. and Steenberg, B., Svensk Papperstidn., 1963, 66 (19), 754-764
- 2. Craven, B. D., Appita, 1962, 16 (2), 57-67
- 3. Johansen, F. and Kubát, J., Svensk Papperstidn., 1964, 67 (20), 822-832
- 4. Rance, H. F., Tappi, 1956, 39 (2), 104-115
- 5. Brezinski, J. P., Tappi, 1956, 39 (2), 116-128
- 6. Gavelin, G., Svensk Papperstidn., 1949, 52 (17), 413-419
- 7. Ivarsson, B., Tappi, 1956, 39 (2), 97-104
- 8. Malmberg, B., Svensk Papperstidn., 1964, 67 (17), 690-692

- 9. Andersson, O. and Berkyto, E., Svensk Papperstidn., 1951, 54 (13), 437-440
- Wink, W. A., Hardacker, K. W., Van Epperen, R. H. and Van den Akker, J. A., *Tappi*, 1964, 47 (1), 47–54
- Ranger, A. E. and Hopkins, L. F., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 277-310
- 12. Craven, B. D., Appita, 1961, 15 (2), 59-67
- 13. Schulz, J. H., Tappi, 1961, 44 (10), 736-744
- 14. Gates, E. R. and Kenworthy, I. C., Paper Tech., 1963, 4 (5), T145-T152
- 15. Toroi, M., Paper & Timber (Finland), 1959, 41 (5), 271-273, 275-279
- 16. Danielsen, R. and Steenberg, B., Svensk Papperstidn., 1947, 50 (13), 301-305
- 17. Prusas, Z. C., Tappi, 1963, 46 (5), 325-330
- Giertz, H. W., Proceedings of EUCEPA/European TAPPI Conference, Venice, 1964 (in print)
- 19. McKenzie, A. W., Austral. J. appl. Sci., 1960, 11 (4), 451-461
- 20. Rance, H. F., *Mechanical Properties of Wood and Paper*, Ed. R. Meredith (North Holland Publishing Co., Amsterdam, 1953)
- 21. Luner, P., Kärnä, A. E. U. and Donofrio, C. P., Tappi, 1961, 44 (6), 409-414
- Kallmes, O. J. and Bernier, G. A., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 369–388
- 23. Malmberg, B., Svensk Papperstidn., 1964, 67 (3), 69-74; (16), 617-621
- 24. Arlov, A. P., Norsk Skogind., 1959, 13 (10), 342-351
- 25. Jayne, B. A., Tappi, 1959, 42 (6), 461-467
- Kallmes, O. J., Stockel, I. H. and Bernier, G. A., Pulp & Paper Mag. Can., 1963, 64 (10), T449–T456
- 27. Leopold, B. and McIntosh, D. C., Tappi, 1961, 44 (3), 235-240
- 28. McIntosh, D. C., Tappi, 1963, 46 (5), 273-277
- 29. Hartler, N., Kull, G. and Stockman, L., Svensk Papperstidn., 1963, 66 (8), 301-308
- 30. Britt, K. W. and Yiannos, P. N., Tappi, 1964, 47 (7), 427-431
- 31. Page, D. H., What we are doing (B.P. & B.I.R.A.), 1963, (32), 25-26
- 32. Jayme, G. and Krause, T., Holz Roh- u. Werkstoff, 1963, 21 (1), 14-19
- 33. Stone, J. E., Pulp & Paper Mag. Can., 1964, 65 (1), T3-T12
- Forgacs, O. L., Robertson, A. A. and Mason, S. V., Fundamentals of Papermaking Fibres, Ed. F. Bolam (Technical Section, B.P. & B.M.A., Kenley, 1958), 447–473
- Samuelson, L. G., Svensk Papperstidn., 1963, 66 (15), 541–546; 1964, 67 (22), 905–910
- Robertson, A. A. and Mason, S. G., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 639-647
- 37. Robertson, A. A., Svensk Papperstidn., 1963, 66 (12), 477-497
- 38. Kallmes, O. J. and Eckert, C., Tappi, 1964, 47 (9), 540-548
- 39. Jentzen, C. A., Tappi, 1964, 47 (7), 412-418
- 40. Meyer, K. H. and Lotmar, W., Helv. Chim. Acta, 1936, 19 (1), 68-86
- 41. Hearle, J. W. S., J. Text. Inst., 1962, 53 (8), P449-P464
- 42. Asunmaa, S. and Steenberg, B., Svensk Papperstidn., 1958, 61 (18b), 686-695

- Page, D. H., Tydeman, P. A. and Hunt, M., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 171-193
- 44. Jayme, G. and Hunger, G., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 135-170
- Buchanan, J. G., and Washburn, O. V., Pulp & Paper Mag. Can., 1962, 63 (10), T485-T493
- 46. Helle, T., Norsk Skogind., 1963, 17 (10), 276, 378, 383, 385
- 47. Page, D. H. and Tydeman, P. A., Formation & Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 397-413
- 48. Kallmes, O. and Bernier, G. A., Tappi, 1963, 46 (2), 108-114
- 49. Van den Akker, J. A., Tappi, 1959, 42 (12), 940-947
- 50. Mayhood, C. H., Kallmes, O. J. and Cauley, M. M., Tappi, 1962, 45 (1), 69-73
- 51 Schniewind, A. P., Nemeth, L. J. and Brink, D. L., *Tappi*, 1964, **47** (4), 244–248
- Nordman, L. S., Fundamentals of Papermaking Fibres, Ed. F. Bolam (Technical Section, B.P. & B.M.A., Kenley, 1958), 333–347
- 53. Nordman, L. S. and Göttsching, L., Das Papier, 1963, 17 (6), 237-246
- 54. Stone, J. E., Pulp & Paper Mag. Can., 1963, 64 (12), T528-T533
- 55. Stamm, A. J., Tappi, 1957, 40 (9), 765-770
- 56. Van den Akker, J. A., Tappi, 1950, 33 (8), 398-402
- 57. Kallmes, O. and Corte, H., Tappi, 1960, 43 (9), 737-752
- 58. Kallmes, O. and Corte, H. and Bernier, G., Tappi, 1961, 44 (7), 519-528
- 59. Corte, H. and Kallmes, O. J., *Formation and Structure of Paper*, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 13-46
- 60. Kallmes, O. and Bernier, G., Tappi, 1963, 46 (2), 108-114
- 61. Kallmes, O., Corte, H. and Bernier, G., Tappi, 1963, 46 (8), 493-502
- 62. Kallmes, O. and Bernier, G., Tappi, 1964, 47 (11), 694-703
- 63. Helle, T., Norsk Skogind., 1964, 18 (3), 92-97
- Van den Akker, J. A., Lathrop, A. L., Voelker, M. H. and Dearth, L. R., *Tappi*, 1958, 41 (8), 416–425
- 65. Helle, T., Svensk Papperstidn., 1963, 66 (24), 1 015-1 030; Lic. tech. thesis, Technical University of Norway, 1962
- 66. Houen, P., Private communication to be published in Norsk Skogind.
- Kubát, J., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 393–394
- 68. Corte, H., Kallmes, O. and Jarrot, D., Paper Maker, 1961, 142 (7), 61, 62, 64–66, 68–70, 72
- Page, D. H., Tydeman, P. A. and Hunt, M., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 249-270
- 70. Brecht, W. and Wanka, R., Das Papier, 1963, 17 (4), 141-147
- Rothman, M., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 316
- 72. Maynard, C. R. G., Proc. Tech. Sect. P.M.A., 1948, 29 (2), 470

- 73. Sanborn, I. B., Tappi, 1962, 45 (6), 465-474
- 74. Tydeman, P. A. and Hiron, A. M., What we are doing (B.P. & B.I.R.A.), 1964, (35), 9-21
- Sternstein, S. S. and Nissan, A. H., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 319–347
- 76. Nissan, A. H. and Sternstein, S. S., Tappi, 1964, 47 (1), 1-6
- 77. Page, D. H., Tappi, 1963, 46 (12), 750-756
- 78. Van den Akker, J. A., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 205-241
- Majewski, Z. J., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 414–415
- 80. Giertz, H. W., Norsk Skogind., 1964, 18 (7), 239-244, 246-248
- 81. Arlov, A. P. and Snaprud, S. I., Norsk Skogind., 1964, 18 (5), 172-178, 180
- 82. Steenberg, B., Svensk Papperstidn., 1947, 50 (6), 127-140
- 83. Steenberg, B., Svensk Papperstidn., 1947, 50 (15), 346-350
- 84. Ivarsson, B. and Steenberg, B., Svensk Papperstidn., 1947, 50 (18), 419-432
- 85. Ivarsson, B., Svensk Papperstidn., 1948, 51 (17), 383-388
- 86. Steenberg, B., Pulp & Paper Mag. Can., 1949, 50 (3), 207-244
- 87. Hurley, R. E., Tappi, 1951, 34 (9), 390-395
- 88. Cox, H. L., Brit. J. appl. Phys., 1952, 3, 72-79
- 89. Le Cacheux, P., Papeterie, 1953, 75 (10), 259, 661, 663-665, 667, 669, 671, 673
- 90. Onogi, S. and Sasaguri, K., Tappi, 1961, 44 (12), 874–880; J. Jap. TAPPI, 1957, 4 (11), 233–238
- 91. Litt, M., J. Coll. Sci., 1961, 16 (3), 297-310
- 92. Campbell, J. C., Appita, 1963, 16 (5), 130-137
- 93. Page, D. H., RA/T/105 (B.P. and B.I.R.A., Kenley, 1963)
- 94. Tydeman, P. A. and Hiron, A. M., Paper Tech., 1962, 3 (4), 315, 316, 319
- Nissan, A. H., Formation and Structure of Paper, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 119–130
- 96. Balodis, V., Austral. J. appl. Sci., 1963, 14 (4), 284-304
- 97. Arlov, A. P. (Dr. tech. thesis, Technical University of Norway, 1958)

## Discussion

**Dr O. L. Forgacs**—On listening to the discussion of the stress/strain theories of paper, I wonder whether we are not losing sight of the purpose of this work.

There are surely two objectives, one short-term and one long-term. The short-term one is to arrive at a simplified and approximated theory for the stress/strain curve, which is immediately useful to applied problems. Therefore, any theoretical treatment, whether based on springs and dashpots or any other device, is quite acceptable, if—and only if—it fulfils a pragmatic function.

The long-term objective is to obtain a true insight into the behaviour of the paper network under applied stresses. It seems to me that, to accomplish this, the arrangement and behaviour of the individual fibre elements and the bonds under stress must be thoroughly understood before a realistic model for theoretical treatment can be constructed.

Is it not possible that the controversy today over the choice of models arises through our occasional neglect to define objectives?

**Dr H. F. Rance**—Before we leave this particular paper, I would like to raise one very small point that might otherwise be missed when we go to the more general discussion. I mention this, because it relates to the paper by Ihrman & Öhrn, which was billed to be in this session but was taken earlier. You will remember that Ihrman said that compacted greaseproof paper showed a decrease in thickness under tensile strain and he suggested that it differed in this respect from other compacted papers; it appeared to follow that the compaction of wet-beaten papers was different in principle from the compaction of free-beaten papers. In fact, Algar has rightly pointed out that ordinary wet-beaten papers like greaseproof even before compaction decrease in thickness under tensile strain. I want to thank him for reminding us of it, as it tends to clarify the apparent anomaly that was left over from our compaction session.

Dr J. A. Van den Akker—Algar's task of reviewing the experimental and theoretical researches on the structure and properties of paper was indeed a

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tremendous one and he has done an excellent job. It is natural that he has used the terminology implanted by workers in the field, but some of this I believe to be misleading. In directing some comments on nomenclature, therefore, it should be clear that the latter are not critical of his efforts.

Some authors have classified theories of sheet elasticity as falling under uniform force and uniform strain. I suggest that the latter is a poor designation of theories like the one I presented at the 1961 Oxford symposium, because of an implied parallelism with *uniform force*, a parallelism that, in fact, does not exist. In the uniform force classification, the basic assumption is that all the fibres in the sheet are extended by the same force—which is simply poor physics. The implied parallelism in *uniform strain* theories is that all the fibres in the sheet are extended by the same strain, which is not the case. (In my 1961 Oxford theory, for example, axial fibre strains and forces vary with orientation from a positive maximum to a negative maximum.) Persons working in the field have come to interpret *uniform strain* as referring to the assumption that the strain is uniform throughout the sheet: but this misses the main point of a so-called uniform strain theory. The essential assumption of such a theory, in so far as strain is concerned, is only that the extension and flexure of a fibre element is dictated by the strain of the sheet in the immediate neighbourhood of the fibre element. One of the next steps in development of the theory of paper structure is that of departing from the idealised assumption of uniform strain in the sheet: the basic ideas already developed in theories that are improperly classified under uniform strain should be retained, rather than discarded, when the step towards non-uniform sheet strain is taken. I suggest therefore that we adopt a new designation for elasticity (low strain) theories that properly incorporate the principles of physics for the assumed models. If there is to be a uniform force classification, why not a non-uniform force theory?or, to break away completely from this type of designation, a term like *linear* network theory would be acceptable for the *elastic regime* of the stress/strain relationship of paper.\*

<sup>\*</sup> Developing from this comment, later consultation between Drs Algar and Van den Akker led to Dr Algar's agreement for the nomenclature in his paper as published to be changed to agree with Dr Van den Akker's 1961 symposium paper and it appears in this revised form in these transactions—Ed.