

# THE BEATING PROCESS

## PRIMARY EFFECTS AND THEIR INFLUENCE ON PULP AND PAPER PROPERTIES

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### *Synopsis*

*Developments in research on the beating process since the Cambridge symposium are reviewed with respect to changes in the individual fibres and the manner in which these primary effects influence pulp and paper properties. The following are regarded as primary effects—breaking of intrafibre bonds, external fibrillation and foliation, formation of fines and fibre shortening.*

*The breaking of intrafibre bonds can take place at various dimensional levels, but can be referred to changes in the hydrogen bond structure. Properties discussed include specific volume, specific surface, flexibility, flow resistance, drainage resistance, wet web strength, drying tensions, bonded area, density, tenacity, extensibility, Young's modulus, rupture energy, creep, tear factor and folding endurance.*

*Experiments on the influence of beating on the flow resistance and drainage resistance are described in some detail.*

### ***L'engraissement: les effets primaires et leur influence sur les propriétés de la pâte et du papier***

*Les auteurs font le point des progrès des recherches depuis le symposium de Cambridge sur les effets de l'engraissement sur les fibres unitaires et sur la façon dont ces 'effets primaires' influencent les propriétés de la pâte et du papier. Ils considèrent comme effets primaires—la rupture des liaisons intrafibrillaires, la fibrillation externe, le détachement de lamelles, la formation des fines et le*

*raccourcissement de la fibre. La rupture de liaisons intrafibrillaires peut se produire à de différents niveaux dimensionnels, mais peut se rattacher aux changements de la disposition des ponts hydrogène. Les caractéristiques examinées sont—le volume et la surface spécifiques, la flexibilité, les résistances à l'écoulement et à l'égouttage, la résistance de la feuille humide, les tensions de séchage, la surface de liaison, la tenacité, l'extensibilité, le module d'Young, l'énergie de rupture, la déformation plastique de sous tension, le coefficient de déchirement et la résistance au pliage.*

*Les auteurs décrivent en détail l'influence de l'engraissement sur les résistances à l'écoulement et à l'égouttage.*

### **Der Einfluss der Primärwirkungen bei der Mahlung auf Stoff und Papiereigenschaften**

*Die Stoff- und Papiereigenschaften werden von folgenden Primärwirkungen des Mahlprozesses beeinflusst—dem Aufbrechen der Brücken innerhalb der Fasern, der äusseren Fibrillierung und Folienbildung und der Bildung von Feinstoff und der Faserkürzung. Das Aufbrechen der Bindungen innerhalb der Fasern kann auf verschiedenen Dimensionsebenen erfolgen, beruht aber auf der Änderung der Wasserstoffbrückenstruktur. Im einzelnen handelt es sich bei den Eigenschaften um spezifisches Volumen, spezifische Oberfläche, Biegsamkeit, Strömungswiderstand, Entwässerungsvermögen, initiale Nassfestigkeit, Trocknungsspannung, gebundene Fläche, Dichte, Dehnung, Young-Modul, Bruchlast, Kriechen, Durchreissfestigkeit und Falzzahl. Es wurden Versuche über den Einfluss der Mahlung auf den Strömungswiderstand und das Entwässerungsvermögen beschrieben.*

### **Preamble**

**T**HE Cambridge symposium<sup>(1)</sup> in 1957 was concerned largely with the beating process and comprehensive and critical accounts were given of beating in relation to the mechanical properties,<sup>(2)</sup> hygrostability<sup>(3)</sup> and porous structure<sup>(4)</sup> of paper, the theory of the process,<sup>(5)</sup> energy considerations,<sup>(6)</sup> factors influencing the rate of response of the pulp<sup>(7)</sup> and the effects on individual fibres.<sup>(8)</sup> These provide an admirable foundation for future discussions. A monograph by Emerton,<sup>(9)</sup> which also appeared in 1957, an earlier review by Clark<sup>(10)</sup> and Rance's discussion of beating in relation to mechanical properties<sup>(11)</sup> are also valuable as points of departure. The present

contribution is an attempt to summarise and review some of the developments in the field of beating research during the last four years and to weave into the context of the account some of our own results and interpretations.

The complexity of the beating process has been often emphasised. On the one hand, a number of more or less separable effects are produced in the individual fibres; on the other, a number of pulp and paper properties of practical importance are affected, including flow behaviour, flocculation, sheet formation, drainage, wet web strength, response to pressing and drying, dimensional stability, porosity, interfibre bonding and mechanical properties of the final sheet and so on. If the ultimate object of research on beating is the practical one of improving efficiency and reducing costs, the best possibility of achieving this would seem to be in the development of ways of attaining the desired effects specifically, without large dissipative losses or the production of unnecessary changes in fibre structure. Such an approach calls for a clear understanding of the changes that take place in the fibre and the way in which they affect the properties of the pulp and paper.

Arising out of the requirements of pulp evaluation, much of the literature on beating deals with correlations between energy input and composite properties such as freeness or bursting strength. Data of this sort are however of only limited use in the development of a basic understanding of the process and, for this purpose, it is convenient to distinguish between the *primary* and *secondary* effects of beating.

The primary effects may be regarded as those that distinguish an individual beaten fibre from an unbeaten one and that cannot be resolved unequivocally into separate components. It is recognised, however, that in no beating process are all the fibres treated in the same way,<sup>(2)</sup> so that some fibres may show a disposition towards one or other of the primary effects. Thus, measurement of the change in a particular fibre property, made on a fibre population, will still be a property of the pulp—in the quantitative sense—rather than of an individual fibre.

The secondary effects may be taken to comprise changes in all the other properties of the pulp, wet web and final sheet that are brought about by beating, but that can be related only in a complex way to the changes in the individual fibres. Some of these depend on interaction between the fibre and the dispersion medium (for example, changes in flow resistance), others on interaction between the fibres themselves (for example, strength development), others on interaction both between fibres and between the fibre and the medium (for example, changes in drainage characteristics).

### Primary effects

THE main primary effects of beating may be listed as —

1. Breaking of some intrafibre hydrogen bonds and replacement by fibre-water hydrogen bonds.
2. External fibrillation and foliation.
3. Removal of layers or fibrils from the outer part of the cell wall, with formation of fines or crill.<sup>(12)</sup>
4. Shortening of the fibre.

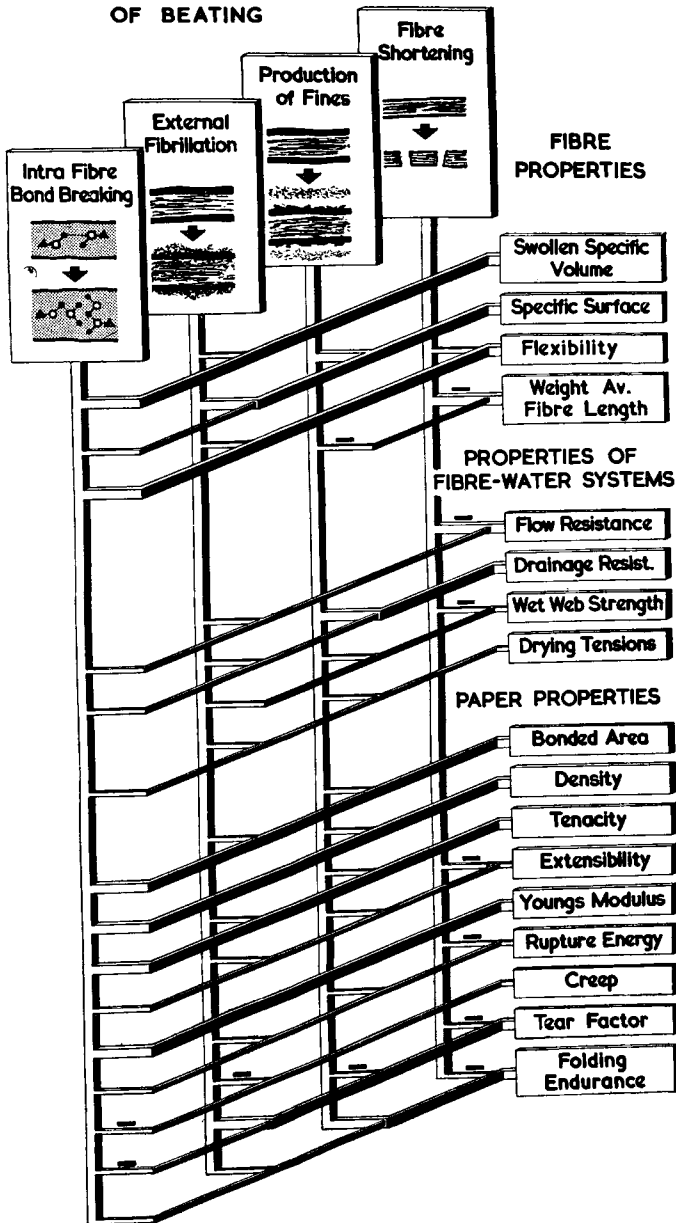
This classification of the effects lying at the root of the changes promoted by beating is a compromise between the molecular and morphological approaches, but, it is hoped, a realistic one. The breaking of intrafibre bonds is taken to include internal fibrillation and processes sometimes referred to as a *loosening* or an *opening up* of the fibre structure: these may involve separation of microfibrils on a scale much greater than molecular dimensions, but it is hard to differentiate between the various levels of splitting. It might be argued, again, that external fibrillation is a special case of intrafibre bond breaking or that removal of fibrils from the main body of the fibre is merely an advanced stage of external fibrillation. However, a fairly clear distinction can be made here between these processes, particularly when the ultimate consequences are considered.

In the following discussion, an attempt is made to relate fibre, pulp and paper properties to the main primary effects of beating. The relationships that will emerge from these considerations are foreshadowed in Fig. 1.

#### Breaking of intrafibre bonds

It is apparent from the infra-red spectra of cellulose and other hydroxy compounds that practically all the hydroxyl groups are involved in hydrogen bonding.<sup>(13)</sup> This conclusion is based on the frequency of the hydroxyl group stretching vibration (about  $3\ 300\text{ cm}^{-1}$ ), which is considerably lower than that observed for unbonded hydroxyl groups (about  $3\ 600\text{ cm}^{-1}$ ), for example, in water or alcohols in the vapour phase. The evidence for interfibre hydrogen bonding in paper is less direct, since the number of such bonds is only a very small fraction of the total number of hydrogen bonds in the paper<sup>(14)</sup> and could easily escape detection by spectroscopic methods. Various workers<sup>(14-19)</sup> have shown, however, that blocking the hydroxyl groups of the fibre surface by hydrophobic substituents drastically reduces the interfibre bonding, others have studied the strength of paper in media of different hydrogen-bonding capacity and of freeze-dried paper.<sup>(20)</sup> These investigations

**PRIMARY EFFECTS  
OF BEATING**



The primary effects are shown in a highly idealised way. A thick connecting member indicates that the primary effect is considered to exert a major influence, a thin member indicates a lesser influence. The absence of any connecting member does not necessarily mean that there is no relationship whatsoever, but it indicates that any such influence is considered to be quite small compared to the others that determine the property in question. A minus sign indicates an inverse relationship.

**Fig. 1**—Relationship between the primary and secondary effects of beating

lead to the conclusion that interfibre hydrogen bonds form during the drying of paper and provide the basic mechanism of paper strength.\*

Internal fibrillation needs to involve only a very small proportion of the hydrogen bonds initially present. According to Frey-Wyssling,<sup>(1)</sup> perhaps 100 cellulose chains pass through the cross-section of an elementary fibril in cotton, 2 000 through a microfibril and 500 000 through a macrofibril. The question of the dimensions of native cellulose fibrils has been reviewed by Rånby,<sup>(1)</sup> who quotes fibril widths of 80–100 Å and thicknesses of 30–40 Å for wood celluloses, corresponding to about 100 chains per cross-section. Partial fibrillation would thus give rise to only a small change in sorptive capacity, yet the tenacity with which water is held by the fibres would be increased and the rate at which it can be removed would be decreased. This is in fact what happens on beating.

The hydrogen bonds that are broken during beating in water appear to be confined mainly to the amorphous regions of the cellulose, since no large change in crystallinity is usually apparent on beating as assessed by X-ray diffraction,<sup>(10)</sup> sorption<sup>(10)</sup> or infra-red spectroscopy. Examination of the spectra of a purified eucalypt pulp and a pine kraft pulp, before and after beating for 40 000 rev in the Lampén mill, revealed negligible differences. The studies of Wijnman<sup>(21)</sup> certainly showed a moderate reduction in the average size of the crystalline regions; in this case, purified cotton was heavily beaten in the Jokro mill and the changes were accompanied by only a small reduction in the total crystalline fraction as estimated from hygroscopicity and X-ray measurements. Borruso<sup>(22)</sup> obtained similar results and found also that neutral sulphite semichemical pulps from spruce and poplar gave only a very small change in crystallinity after prolonged beating; mechanical pulps showed no effect. In Wijnman's experiments, a reduction in D.P. was also observed, but it is believed that, to the extent that covalent bonds are broken

\* Surprise and an element of scepticism are sometimes expressed at the rather rapid appreciation of the role of the hydrogen bond that has developed in the paper industry in recent years. We believe that there is at least one very sound and logical basis for this: the availability in many laboratories of the infra-red spectrometer. Prior to this development, the hydrogen bond appeared to many as a rather nebulous secondary valency that could be invoked to explain discrepancies in the behaviour of some simple compounds. Its ubiquity in polysaccharides, proteins and other polymeric materials had not been fully accepted: however, this fact is now forced continually upon the spectroscopist dealing with these materials. The effect of hydrogen bonding on the vibrations of the hydroxyl groups can be readily discerned and, even though the ultimate nature of the bond is still a matter of enquiry by chemical physicists, its existence and behaviour take on a reality comparable with that of the covalent bond. It is natural that efforts will be made to express the behaviour of paper in terms of the properties of the hydrogen bond and, however tentative and approximate these may be in the initial stages, they warrant every encouragement.

during normal beating operations, any deleterious effects would be overwhelmed by other effects leading to improved interfibre bonding.

On the basis of the hydrogen bond theory of the mechanical properties of cellulose sheets or paper, Nissan<sup>(23)</sup> has advanced an interpretation of the beating process, in which, on a comparison of papers from unbeaten and beaten pulps, the main effect is regarded as the rearrangement of intrafibre hydrogen bonds to form new interfibre hydrogen bonds. The number of bonds per unit volume, which on the hydrogen bond theory is proportional to the cube of the Young's modulus, is increased by the compaction of the amorphous region on drying the assemblage of beaten fibres. Support for these views was found in a correlation reported by Nordman,<sup>(24)</sup> for papers made from pulps beaten to various degrees, between the number of bonds per unit volume as calculated from Young's modulus and the same quantity calculated from the bond strength. This was determined from the slope of the line relating the increase in scattering coefficient to the irrecoverable energy loss in a straining/destraining cycle. Bonded area, used in the calculation of bond density, was determined by taking the scattering coefficient as a measure of unbonded surface area. The validity of the correlation was however questioned in discussion.<sup>(25)</sup>

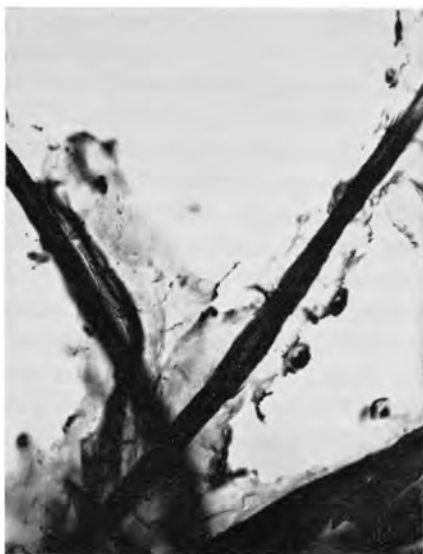
It may readily be agreed that the breaking of intrafibre bonds is an important effect of beating, but it assists analysis to view the replacement of these hydrogen bonds by fibre-water bonds as the primary effect and the development of a higher interfibre bond density on drying the beaten sheet as secondary.

### *External fibrillation*

It is generally accepted that during beating the outside of the fibre undergoes mechanical damage, so that projecting sheets or fibrils may be discerned by suitable microscope techniques. Considerable controversy persists, however, about the importance of external fibrillation in the development of strength properties on beating. Clark<sup>(26)</sup> maintains that the appearance of microscopic and sub-microscopic fibrils plays the most important role in strength development, although he admits that swelling and flexibility participate. Clark's views are in general conformity with those of Strachan,<sup>(27)</sup> a skilful microscopist, who emphasised the role of fibrillation of the outer layers of the fibre in facilitating the access of water, hence the rate and amount of swelling. This view is still acceptable, but it is difficult in the light of modern concepts of the hydrogen bond to subscribe to Strachan's explanation of interfibre bonding as arising primarily from mutual entanglement of the fibrils produced by beating.



(a) Unbeaten *Pinus radiata* holocellulose, lightly silvered ( $\times 140$ )



(b) *Pinus radiata* holocellulose, beaten 5 min in M.S.E. homogeniser ( $\times 140$ )



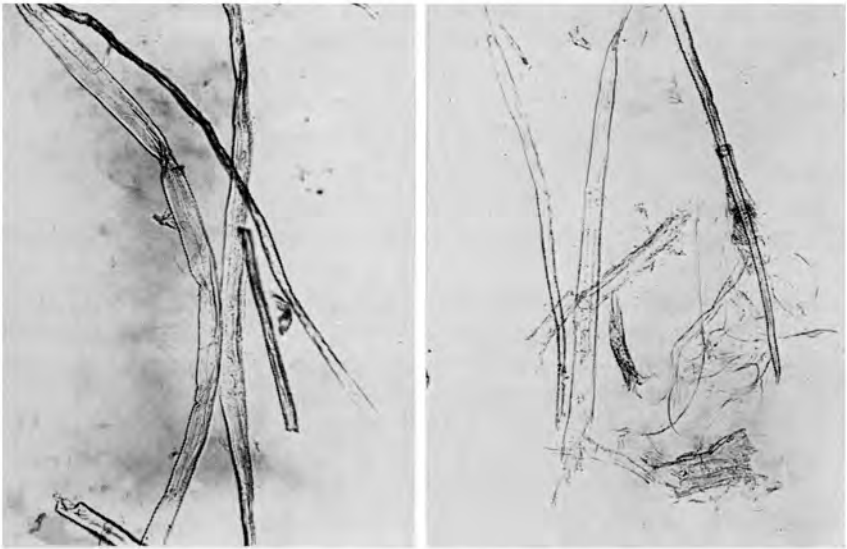
(c) Unbeaten spruce sulphite pulp, lightly silvered ( $\times 140$ )



(d) Spruce sulphite pulp beaten 72 min in Lampén mill, lightly silvered ( $\times 140$ )

**Fig. 2**





(e) Unbeaten spruce sulphite pulp, unsilvered ( $\times 90$ )

(f) Spruce sulphite pulp beaten 72 min in Lampén mill, unsilvered ( $\times 90$ )

**Fig. 2**



(g) Unbeaten *Eucalyptus regnans* holocellulose, unsilvered ( $\times 90$ )



(h) *Eucalyptus regnans* holocellulose, beaten 10 min in M.S.E. homogeniser, unsilvered ( $\times 90$ )



(i) Unbeaten *Eucalyptus regnans* holocellulose, heavily silvered ( $\times 90$ )



(j) *Eucalyptus regnans* holocellulose, beaten 10 min in M.S.E. homogeniser, heavily silvered ( $\times 90$ )

(Samples prepared by Miss V. Goldsmith, photographs by Dr. H. E. Dadswell)

**Fig. 2**—Silvering<sup>(10)</sup> as an aid to the detection of external fibrillation

The detection of fibrillar 'fuzz' on the outside of beaten fibres is facilitated by a light silvering treatment as advocated by Clark.<sup>(10)</sup> The contrast between *Pinus radiata* holocellulose before and after beating in the M.S.E. homogeniser is shown in Fig. 2*a* and 2*b*. A spruce sulphite pulp beaten in the Lampén mill also showed considerable development of external fibrillation accompanied by some fibre splitting (Fig. 2*c* and 2*d*). Somewhat similar effects could be observed in this material even without silvering (Fig. 2*e* and 2*f*). The change in *Eucalyptus regnans* holocellulose upon beating in the M.S.E. homogeniser could be detected readily only in silvered fibres (Fig. 2*g*–2*j*). Heavy silvering gave rise to the appearance of blobs on the fibre surface, which contrasted with the relatively smooth outlines of the unbeaten fibres (Fig. 2*i* and 2*j*).

An aspect of external fibrillation that was emphasised by Emerton<sup>(28)</sup> at Cambridge is that the material detached or partly detached from the surface is largely in the form of sheets or membranes rather than longitudinal strings. A study<sup>(29)</sup> of the material that normally passes into the whitewater revealed, however, the presence of fibrillar particles, the nature of which could more easily be explained as discrete fibrils than as rolled-up flakes. James and Wardrop<sup>(30)</sup> found by the shadow casting technique that a considerable amount of primary wall debris, largely derived from vessel elements, was formed on beating eucalypt pulps in the Lampén mill. In the case of *Pinus radiata* sulphate pulps, part of the outer layer of the secondary wall (S1) is also removed.

Asunmaa and Steenberg<sup>(31)</sup> applied phase contrast microscopy to an examination of very thin handsheets made from bleached spruce sulphite and bleached pine kraft pulps beaten in the P.F.I. beater. Their results showed that the S1 layer was often removed in beating and suggested that 'fibrillation' involved a splitting up of the S2 layer. Mutual interfibre contacts were observed between S1, S2 and the lumen.

### Formation of fines

In his 1957 review of the effects of beating on individual fibres, Giertz<sup>(8)</sup> suggested that swelling and the removal of the primary wall, by making the fibre more flexible, might be mainly responsible for the improvement in interfibre bonding and paper strength. It now appears<sup>(30,31)</sup> that both the P and S1 layers are removed on beating and contribute to the fibre debris or fines. In addition, Steenberg, Sandgren and Wahren<sup>(12)</sup> have found that an important part of the fine fraction of beaten pulps consists of loose slender, fibrillar particles—which they term *crill* and define as that part of the cellulosic

material that does not become entangled in the fibre plug during plug flow. Beating 'slime' is made up mainly of these particles.

Fines have a decisive influence on the drainage resistance of beaten pulps: Ingmanson and Andrews<sup>(32)</sup> found that the increase in drainage resistance on beating depends primarily on the amount of fines produced. Of the various methods of measuring specific surface, however, only drainage resistance accounts for the fines satisfactorily, according to Thode and Ingmanson.<sup>(33)</sup> Thus, the dye adsorption technique<sup>(34)</sup> gives a reasonable comparative estimate of external specific surface for whole fibres, but is insensitive to fines. The apparent influence of fines on mechanical properties differs according to the type of beater, the type of pulp, the size of the particles in the fines fraction and whether the fines are added to or subtracted from the pulp. The experiments of Stephansen<sup>(35)</sup> with the P.F.I. beater (cited by Steenberg<sup>(2)</sup>) supported Strachan's view that the fines contribute little towards the ultimate strength of the pulp. Murphy<sup>(36)</sup> found that the addition of fines (—150 mesh/in) had only a small effect on the strength of an unbleached eucalypt kraft pulp, though screening the same type of pulp after beating to various degrees of freeness in both the Lampén mill and the Valley beater reduced breaking length, stretch and burst factor. Goldsmith and Higgins<sup>(37)</sup> found that the removal of the colloidal fraction from a eucalypt kraft pulp that had been highly beaten in the Lampén mill had little effect on strength, but large decreases in breaking length, stretch and burst were observed on removing material through an 80 mesh/in screen. These effects were more pronounced for a eucalypt kraft than for a pine kraft pulp and were particularly marked in respect to folding endurance. Tear factor is exceptional in that it is increased on removing the fines fraction from a highly beaten pulp. The removal of crill<sup>(38)</sup> produces effects similar to the above on tensile strength and tear, as shown recently by Sandgren and Wahren. In earlier experiments on unbleached sulphate pulps beaten in the Lampén mill and Aylesford beater, Gartshore<sup>(39)</sup> showed that fines also could contribute considerably to paper strength.

There has been some discussion on the role of fines during the beating process<sup>(33,40)</sup> and controversy, particularly in the U.S.S.R., on the advantages of fractionating the pulp during beating. In one of the later articles, Reizinsh and Kalninsh<sup>(41)</sup> contend that fractional beating represents a significant advance in paper technology in that there is a large decrease in power consumption and, on the basis of laboratory tests, an increase in strength properties. It is apparent, however, that these views are not universally accepted.<sup>(33)</sup>

*Fibre shortening*

The shortening of cellulose fibres during beating was studied theoretically by Corte,<sup>(42)</sup> who characterised the process by the probability of shortening a fibre of a certain length in a given time. The length distribution at any beating time could be calculated and equations, which were experimentally verifiable, were derived for the change in mean fibre length for purely squeezing and purely cutting actions. The studies of Nordman and Niemi<sup>(43)</sup> confirmed the usually accepted view that a ball mill does not cut the fibres as much as beaters equipped with knives. Cottrall,<sup>(44)</sup> discussing the mechanical effects of beating, made a distinction between transverse subdivision of the fibre by sharp and by blunt cutting. The relative extent to which direct cutting of the fibres by the bars and transverse breaking of the fibres in a strong shear field would contribute to shortening was discussed also by Giertz,<sup>(8)</sup> who cited the studies of Rance and co-workers<sup>(45)</sup> in support of the view that fibres may be broken in a high velocity or acceleration gradient. When these possibilities are considered, it is understandable that the extent of fibre shortening during beating will depend largely on the type of pulp.

As a modification of the rather widely accepted view that fibre length is directly related to strength, the idea is now gaining ground that it is of secondary importance, except in so far as tearing strength and folding endurance are concerned. This concept is really a corollary of the hydrogen bond theory of mechanical behaviour, in which not only interfibre bonding, but the bond system within the amorphous regions of the fibre largely determine prerupture tensile properties and, to a lesser extent, rupture itself. Higuchi<sup>(46)</sup> regards interfibre bonding as the more important factor in strength on the basis of beating studies on hardwood pulp and similar conclusions have been reached in various laboratories. Kane<sup>(47)</sup> has concluded from an analysis of the forces on individual fibres in the deformed paper web that the fibre length changes encountered in beating do not usually have a pronounced effect on tensile strength.

In order to obtain reliable information on the effect of cutting the fibre, one must isolate this variable from the other primary effects of beating. One way in which this can be done is by making transverse cuts across layers of holocellulose prepared from a very long-fibred wood, such as *Araucaria klinkii*, then separating the fibres by alkali treatment. This method was developed by Higgins and Goldsmith<sup>(48)</sup> in work on the flow of fibre suspensions and was applied by Watson and Dadswell<sup>(49)</sup> to the study of the effect of fibre length on paper properties. They found that tear factor and folding endurance were very strongly dependent on fibre length, in accordance with all the previous evidence (different biological origin, fractionation

experiments, etc.), but that breaking length and burst were influenced to a much smaller extent—for example, breaking length was found to double over the range of mean fibre length 2–8 mm.

### *Specific volume, specific surface and flexibility*

CHANGES in swollen specific volume, specific surface and flexibility are of such significance to a wide range of pulp and paper properties that they tend to be regarded as fundamental beating effects. As they can be referred back to morphological and molecular changes in the fibre structure, we prefer to regard them as composite effects that are of basic significance only in that, with the reduction in fibre length, they are measurable changes in the properties of the fibre itself. In principle at least, specific surface, specific volume and flexibility can be measured in physical units in a more precisely defined way than quantities such as the degree of external or internal fibrillation, although in practice various difficulties and uncertainties arise.

The changes in specific surface, specific volume and flexibility have a composite origin in terms of the primary beating effects and it may be an oversimplification to look upon specific surface as a measure of tearing or peeling of fibrils from the fibres and specific volume as a measure of the separation of filaments within the fibre. The increase in specific volume (swelling) also will be facilitated by the removal of part of the restraining primary wall as shown by experiments in which balloon swelling is induced in damaged fibres by cuprammonium<sup>(6)</sup> or phosphoric acid.<sup>(36,50)</sup> Thus, in addition to the breaking of interfibre bonds, external fibrillation and foliation and the formation of fines will play a part in the degree of swelling attained. These three primary effects will clearly contribute also to the increase in true specific surface, but with a quite different emphasis. Flexibility, too, will be influenced greatly by the breaking of intrafibre bonds, by the removal of outer restraining walls and by fibre shortening.

Ingmanson and Andrews<sup>(32)</sup> have studied the effect of beating on specific surface and specific volume, a topic that was introduced by Robertson<sup>(51)</sup> at the Cambridge symposium. The method used was essentially that of Robertson and Mason,<sup>(52)</sup> who applied the Kozeny-Carman equation to the passage of water through porous fibre pads. From this point of view, the filtration resistance can be resolved into specific surface and fibre specific volume, providing the apparent density of the wet mat is known. No change in density was observed with beating, the external surface areas of both fibres and fines were linear functions of beating time and fibre specific volume increased rapidly, then levelled off. The bulk compressibility was found to

increase in proportion to the specific volume. Thode and Ingmanson<sup>(33)</sup> then found that the swollen specific volume was directly related to tensile strength, *per medium* of plasticisation. Robertson<sup>(51)</sup> found similar correlations, but showed that they depended on specific surface and could be vitiated by fibre damage, say, in the case of a sulphite pulp or under drastic refining conditions. An interesting sidelight of the work of Thode and Ingmanson was that very good agreement was obtained, for fines-free pulp, between the centrifugal method of Jayme and Rothamel<sup>(53)</sup> and the filtration resistance method of measuring swollen specific volume.

Useful as these developments are, it should be borne in mind that the Kozeny-Carman equation involves a number of uncertainties<sup>(54)</sup> that have not yet been resolved for porous media of even the simplest structure. It might well be argued, for instance, that the fibre/water system lends itself more naturally to an analysis of permeability based on the resistance to liquid flow past filamentous elements in the system than to one based on a capillary model. Thus, the drag equation of Emersleben<sup>(55)</sup> might provide a starting point for such an analysis. The mathematics involved would be more complex and filtration resistance would be resolvable into different components from those emerging from the Kozeny-Carman analysis. The case for an alternative approach is strengthened by the observation of Ingmanson, Andrews and Johnson<sup>(56)</sup> that the Kozeny 'constant' is a function of porosity.

The influence of swelling on the mechanical properties of paper can be shown in various ways—for example, by introducing hydrophobic groups into the cellulose molecule or by treating the fibre with hydrogen-bond-breaking reagents. If the swollen specific volume can be increased by other means in addition to the mechanical treatment, it would be expected that a stronger paper would be produced and Kress and Bialkowsky<sup>(57)</sup> found that beating rate did increase with the capacity of the pulp to swell in the beating liquid. Work in this laboratory<sup>(58)</sup> has shown that small increases in beating rate could be obtained when kraft pulps were beaten in aqueous solutions of ethylenediamine of low concentration, but that the improvement could easily be overwhelmed by fibre damage. Similarly,<sup>(59)</sup> the beating rate of a bleached kraft pulp was increased slightly when the pulp was pretreated with alkali at low concentration; at higher concentration, the rate decreased. Borruso<sup>(22)</sup> observed an increase in the amorphous fraction when pulps were treated after beating with sodium hydroxide solutions of less than mercerising strength. Cohen<sup>(60)</sup> found that, when a eucalypt kraft pulp from dense wood (that is, consisting of thick-walled fibres) was soaked in 25 per cent monoethanolamine solution for 24 h before beating, considerable improvements in burst, breaking length, stretch and folding endurance were recorded.

Only very small effects were observed in the unbeaten pulp or when the monoethanolamine treatment followed beating. These experiments lead to the conclusion that for normal fibres the amount of swelling that takes place on beating in water is not far below the optimum. It should be recalled that swollen specific volume increases rapidly in the early stages of beating<sup>(32)</sup> and tends towards an equilibrium and it is undesirable to exceed the optimum level (compare Robertson<sup>(51)</sup>). When the beating rate is low, however, as in the case of fibres from dense woods, the use of chemical aids to swelling may be technically advantageous; this does not necessarily imply economic feasibility when factors such as cost and corrosion are taken into account.

For flexibility, the method of measurement proposed by Forgacs, Robertson and Mason<sup>(61)</sup> has been applied to beating and further results have appeared.<sup>(62)</sup> Flexibility is assessed by classifying a fibre population according to the type of rotational orbit assumed by each individual fibre when the suspension is sheared under standard conditions. As beating proceeds, (i) the fraction of fibres in rigid or springy orbits decreases; (ii) the fraction of fibres in orbits of greater curvature increases initially as the internal structure is ruptured and swollen, but then decreases as some of them develop hinge joints and (iii) the proportion of hinge joints increases steadily. The jointed fibre probably represents an extreme form of localised damage of the P and S1 walls. The development of flexible orbits (including the damaged fibres) shows a trend with beating similar to that of tensile strength and density, which is taken<sup>(62)</sup> as evidence for a relationship between the ability of fibres to deform and the extent of bonding. It is reasonable to expect a relationship between the ease of deformation of the fibre in bending (flexibility) and in the lateral compression that is involved in sheetmaking, but this will probably be least marked in the earlier stages of beating, before structural anisotropy has been reduced by internal bond-breaking.

### ***Secondary effects: fibre/water systems***

#### *Flow resistance*

THE resistance to flow of a fibre suspension depends on more than one of the primary beating effects in such a way that opposite general effects may be produced under different beating conditions or with different types of pulp. In the early work of Brecht and Heller<sup>(63)</sup> on the friction losses involved in the flow of stock through pipes, an indication was given that the resistance of shorter-fibred stock was lower than that of long-fibred stock. This suggests that one effect of beating should be to reduce the effective viscosity of the system: however, other factors have to be considered. The hydrodynamic



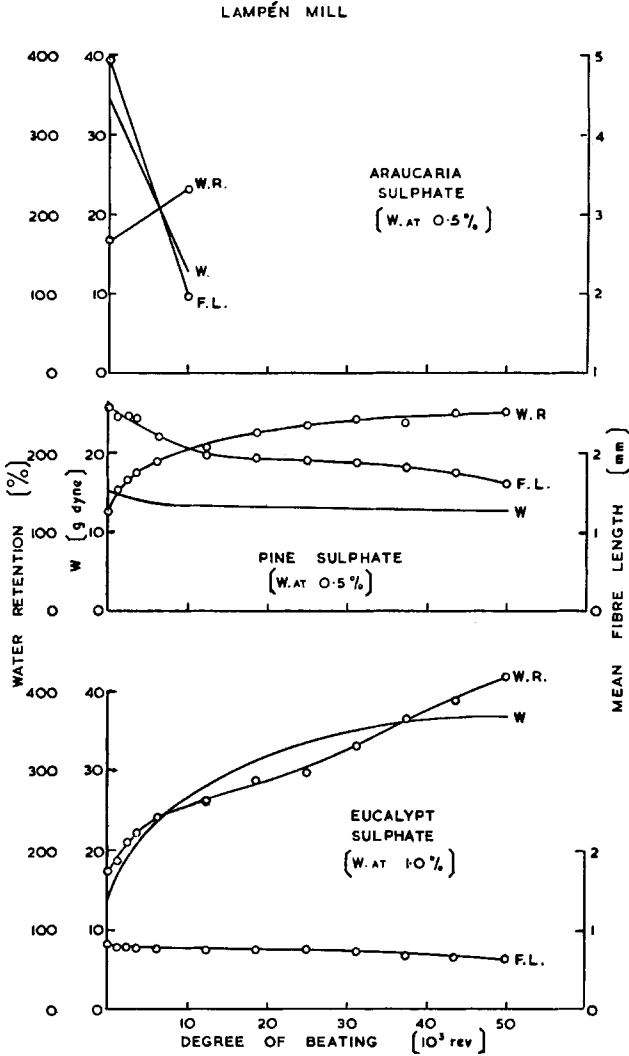
specific volume affects the volume fraction of the system occupied by the fibre<sup>(61,64)</sup>—that is, the hydrodynamically effective concentration. Thus, swelling induced by beating will tend to increase flow resistance. Interaction takes place between dispersed fibres even at very low concentrations and the network compressibility (the volume change under compression) also is believed to be of importance in plug flow.<sup>(61)</sup> By analogy with the comparatively well-known behaviour of polymer solutions, it might be thought that the increases in flexibility and external fibrillation on beating would tend to bring about respectively a decrease and an increase in effective viscosity.

Murley<sup>(65)</sup> observed that groundwood pulps beaten in different instruments gave in one case an increase in viscosity and in the other a decrease, the lower viscosity corresponding to a lower proportion of long fibres and a higher proportion of fines. Another indication of the effect of beating on viscosity is given by Steenberg and Johansson,<sup>(66)</sup> who found that the resistance to flow as measured at high rates of shear decreased when unbleached sulphite pulp was beaten from 16° to 50° S.R. Brecht and Heller<sup>(63)</sup> had found that with different pulps the friction loss could increase, decrease or remain the same on beating. Goldsmith, de Yong and Higgins<sup>(64)</sup> found that the resistance to flow of a eucalypt sulphate pulp rose sharply on beating in the Lampén mill or Valley beater, that of a pine sulphate pulp decreased slightly in the Lampén mill and rose slightly in the Valley beater, that of an *Araucaria* sulphate pulp dropped very rapidly in the Lampén mill (Fig. 3 and 4).

These apparently conflicting results can be understood more clearly by reference to the primary effects of beating. Changes in fibre length and water retention,<sup>(63)</sup> reflecting transverse cutting and swelling,<sup>(63)</sup> respectively, were also measured. The swelling value may be regarded as a measure not only of the breaking of intrafibre bonds, but to some extent of external fibrillation and foliation, combining those primary effects that exert a positive effect on flow resistance, whereas fibre cutting has a negative effect. The general effect of beating could therefore depend on whether fibre shortening or swelling predominates.

Thus, in Fig. 3, the eucalypt pulp on beating in the Lampén mill undergoes a very large increase in swollen specific volume (as shown by the water retention curve), which gives rise to very large viscosity increases, despite the fibre shortening that also occurs. With the very long-fibred *Araucaria* pulp (see below), even a short beating is sufficient to cause a very large reduction in fibre length, which, despite increased water retention, reduces the viscosity to a marked degree. For the pine pulp, the effects of fibre shortening and hydration just about balance and the viscosity is not greatly changed.

The quite distinct response of eucalypt pulp to beating, either in the ball



The changes in fibre length (F.L.) and water retention (W.R.) are used to interpret the effects on  $W$ . The flow experiments were carried out with aqueous suspensions in a rotational viscometer<sup>(64)</sup> and  $W$  is the force required to produce an arbitrary rate of revolution of the rotor

**Fig. 3**—The effect of beating in the Lampén mill on the flow resistance  $W$  of eucalypt, pine and klinki pine (*Araucaria*) pulps

or bar type of beater, is of interest to paper manufacturers using this material or possibly other hardwood pulps. The treatments in both the Valley beater and the Lampén mill were carried out under the same conditions for the eucalypt and the pine pulp. This permitted a comparison of the response of

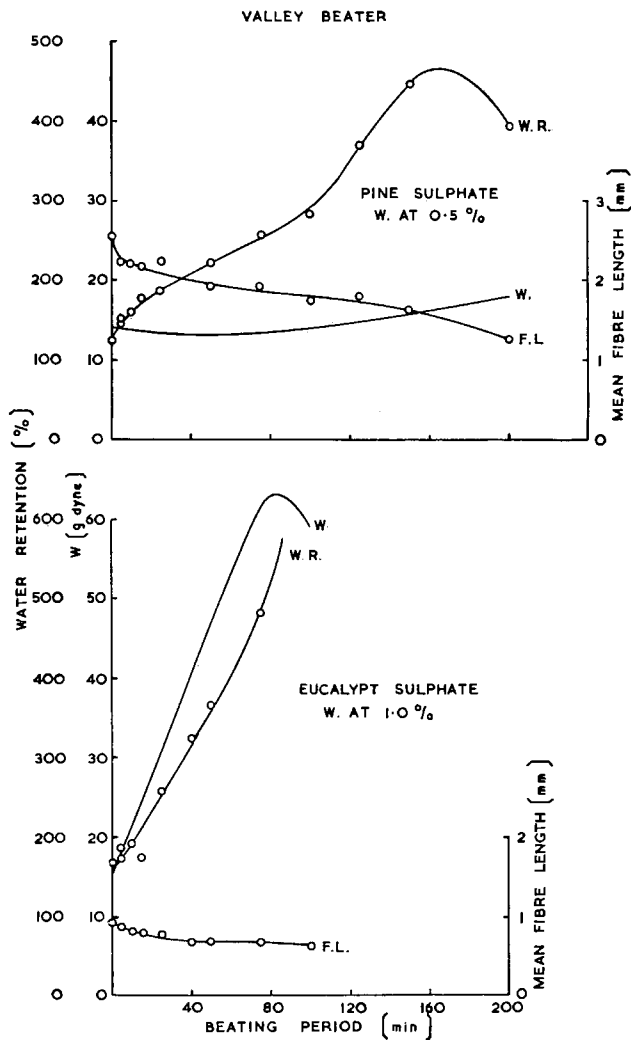
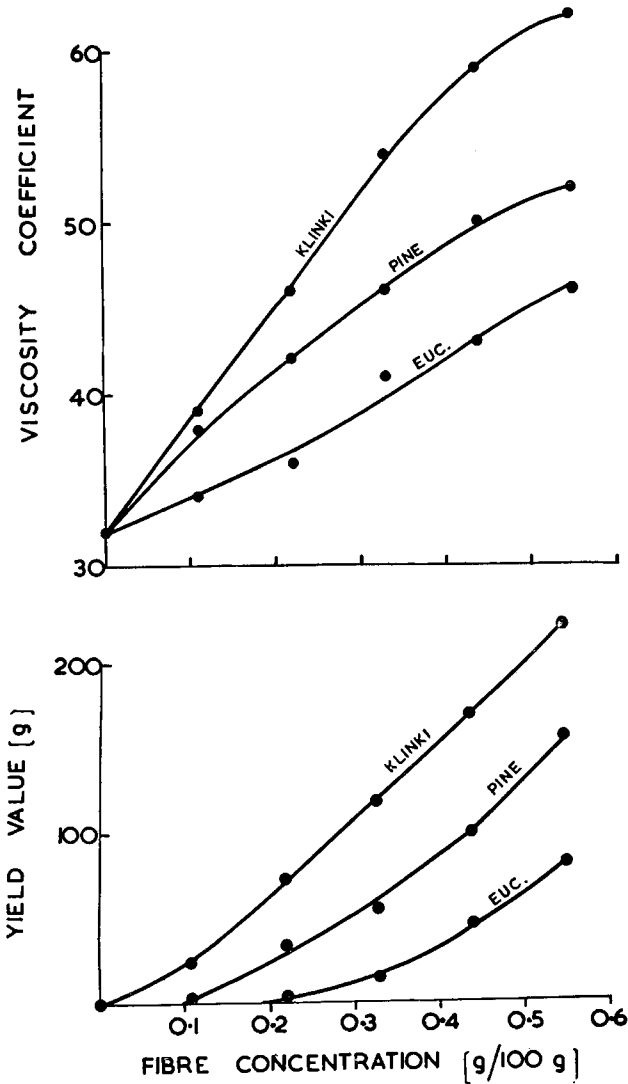


Fig. 4—The effect of beating in the Valley beater on the flow resistance of eucalypt and pine pulps (as in Fig. 3)

the different fibre types, although in one case—the eucalypt pulp in the Valley beater—the conditions deviated from those usually considered to be optimum. Thus, the eucalypt pulp at the beginning of beating offered little resistance to circulation in the beater and the fibre concentration appeared to be rather low for the fibres in the beating zone to offer the most satisfactory cushioning to the impact of the roll on the bedplate. As beating proceeded, the stock thickened and beat in the normal way, but circulation could still be achieved after prolonged beating. With the somewhat higher concentration conventionally used to obtain the most satisfactory initial beating, the circulation of the stock virtually ceases after beating periods similar to those used in this experiment.

It is evident that the change in flow resistance on beating must exert a very considerable influence on the beating process itself, the progress of which is, conversely, dependent on the effective viscosity of the pulp/water system at any particular instant. The observations recorded above may thus assist us to understand the markedly different response to beating of eucalypt and long-fibred pulps.

The flow resistance shown in Fig. 3 and 4 is arbitrarily defined as the force required to produce a given rate of revolution in the rotational viscometer. With water as the dispersion medium, sedimentation, flocculation and other complicating effects adversely affected the reproducibility of the results, although it was possible with care to obtain flow curves showing that the system at low concentrations deviates from Newtonian behaviour and takes on Bingham characteristics. In order that sedimentation should be negligible during the course of an experiment, a dispersion medium of high viscosity (golden syrup) was used in later work.<sup>(48)</sup> The results were far more reproducible and serve to characterise the pulp. The development of non-Newtonian behaviour with increasing concentration and its appearance at lower concentrations as the length and cross-section of the fibres increase (from eucalypt through pine to klinki) are shown in Fig. 5 and 6. This is relevant to beating in that it gives another indication of the effect of fibre length, though fibre shortening can be isolated completely from the other primary effects of beating by the technique mentioned earlier, in which sharp fractions of fibre segments of different lengths are prepared from klinki pine (*Araucaria klinkii*) holocellulose. This species grows in New Guinea and has much longer fibres than most woods; the weight average length is more than 5 mm, which makes it especially suitable for this purpose. The nominal segment lengths were restricted to the range 1–4 mm so as to reduce the number of segments at the end of the native fibre—that is, of final length less than the nominal length of the ‘twice-cut’ segments in the fraction.



The fibre dimensions are shown in Fig. 6

Lower—Yield value derived from Bingham flow curves

Upper—Viscosity coefficient derived from the slope of the flow curves; units equivalent to poises

Fig. 5—The influence of fibre concentration on flow properties of eucalypt, pine and klinki pine pulps, dispersed in golden syrup<sup>(48)</sup>

Flow curves of these fibre fractions as dispersions of various concentrations in golden syrup were determined as before. Variables that had previously influenced the results in an uncontrolled (and often unknown) way were now eliminated.

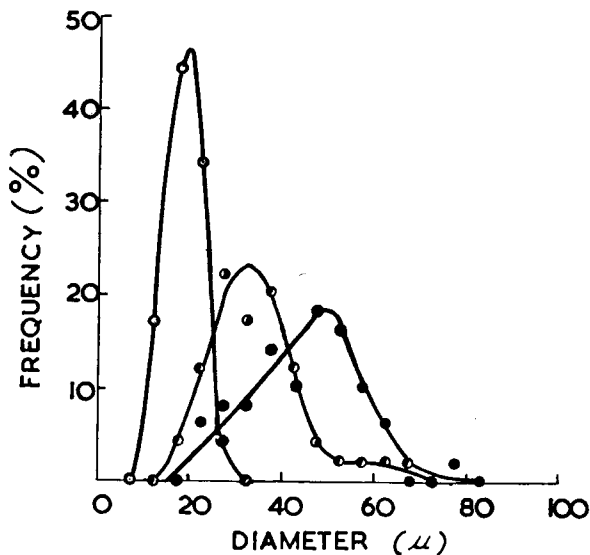
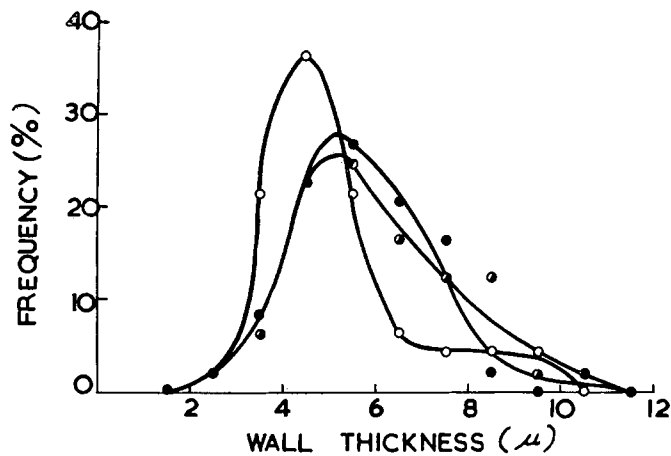
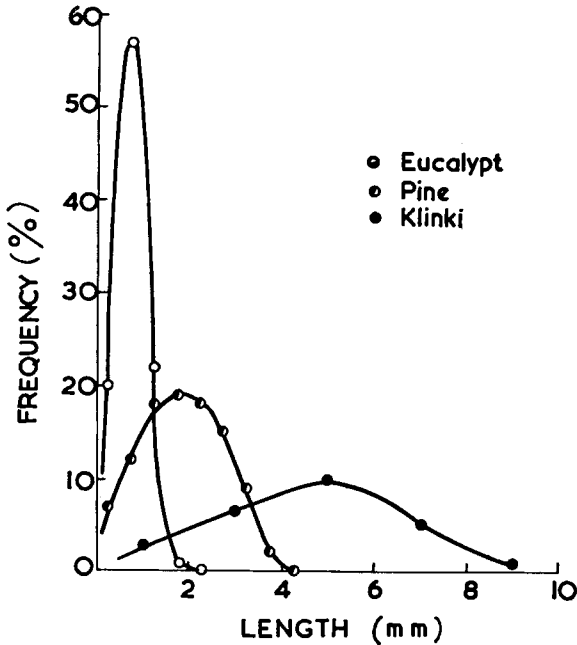


Fig. 6



Curves drawn through histograms with class interval  $5 \mu$  for fibre diameter,  $1 \mu$  for wall thickness,  $0.5 \text{ mm}$  for fibre length of eucalypt and pine and  $2 \text{ mm}$  for fibre length of klinki pine (for comparison frequency for klinki pine is plotted on basis of  $0.5 \text{ mm}$  intervals)

**Fig. 6**—Distribution of fibre length, fibre diameter and wall thickness within the eucalypt, pine and klinki pine pulps<sup>(47)</sup>

As a digression, one such set of variables significant in comparing different pulps rather than in assessing beating effects comprises the cross-sectional dimensions of the fibre, which, for some purposes, can be conveniently combined into a single quantity, the *specific fibre length* of the pulp. This can be defined as the total length of fibre per gram of pulp. If the fibres are considered as hollow cylinders of outer radius  $r_1$ , lumen radius  $r_2$  and wall density  $\rho$ , the specific fibre length is given by  $1/\pi \rho (r_1^2 - r_2^2)$ ; it is independent of mean fibre length (except in so far as this may happen to be related biologically to  $r_1$  and  $r_2$ ). For the eucalypt, pine and klinki pulps of Fig. 6, the specific fibre lengths are, respectively,  $4.96/\rho$ ,  $2.03/\rho$  and  $1.44/\rho \text{ km/g}$ , where  $\rho$  refers to the saturated cell wall. Cutting across the fibre does not change the specific fibre length.

Analogies are evident between the basic factors influencing the flow of

fibre dispersions and those determining the viscosity and rheological behaviour of colloidal solutions of linear unbranched macromolecules. It may be expected, for example, that the weight average fibre length will control the viscosity of the dispersion more closely than the number average. The relative significance of these quantities in respect to papermaking was indicated by Clark.<sup>(67)</sup> In the present work, they were calculated from 100 measurements of fibre or segment length on each fraction, with the results shown in Table 1.

An apparent intrinsic viscosity,  $[\eta]$ , corresponding to each value of the weight average fibre length may also be calculated in the usual way at  $\lim_{c \rightarrow 0} (\eta_{sp}/c)$  where  $c$  is the concentration, expressed in this case in g fibre/100 g dispersion. Linear regressions of  $\eta_{sp}/c$  on  $c$  were fitted to the data and the intercept on the  $\eta_{sp}/c$  axis was taken as  $[\eta]$ . The values found are shown in the last column of Table 1.

TABLE 1—NOMINAL, NUMBER AVERAGE AND WEIGHT AVERAGE FIBRE LENGTHS OF PULPS DERIVED FROM KLINKI PINE HOLOCCELLULOSE, WITH CORRESPONDING INTRINSIC VISCOSITIES  $[\eta]$

Nominal fibre length, mm	Number average fibre length	Weight average fibre length	Intrinsic viscosity, $[\eta]$
1	0.77	0.80	0.22
2	1.52	1.78	0.47
3	2.26	2.70	0.74
4	2.39	3.15	1.11

These experiments confirm beyond doubt that fibre cutting during beating tends to reduce the effective viscosity, although this may be overwhelmed by other primary beating effects.

Flow studies are of interest not only in connection with stock pumping and other phases of the fluid mechanics of fibre/water systems, but also in connection with wet web strength. Forgacs, Robertson and Mason<sup>(61)</sup> showed that the wet web strength in the vicinity of 15 per cent fibre concentration could be correlated with the tensile strength at 0.8 per cent concentration of the coherent fibre network. This in turn was found to be related inversely to the turbulent friction factor. Murley<sup>(65)</sup> showed that a clear correlation existed between pulp viscosity and both wet and dry strength for a series of groundwood pulps taken from the grinder pits under operating conditions.



*Drainage resistance*

An electrical device has been described earlier<sup>(68)</sup> whereby the rate of decline of the surface of the pulp suspension in the Technical Section standard sheetmachine could be recorded during the making of a handsheet. From such records, an approximate drainage resistance could be calculated by a direct application of Darcy's equation. This procedure involved the clearly defined assumption that the friction losses in the outlet system of the sheetmachine could be neglected. These losses have now been evaluated by carrying out tests with water only; in the absence of a pulp pad, the static head  $s$  is equal to the pipe friction loss  $av^n$ , where  $v$  is the rate of decline of liquid in the cylinder,  $a$  and  $n$  are constants. Plots of  $\log s$  against  $\log v$  were linear and gave values of 2.75 and 2.0 for  $a$  and  $n$ , respectively. Thus, the true pressure  $p$  in inches of water across the forming pad is given by—

$$p = s - 2.75 v^2$$

The validity of this expression for the pressure to be used in Darcy's equation was verified by separate experiments with a number of perforated celluloid sheets, which provided a range of drainage resistances, each constant during a particular test and related to each other in a known way. These observations showed<sup>(69)</sup> that the computed drainage resistance was in fact constant with time during the process of emptying the cylinder of the machine and that the resistances measured in the various experiments stood in the same relationship to each other as those predicted from the numbers of holes in the celluloid sheets.

The resistance obtained from Darcy's equation by dividing the pressure across the pad by the volume rate of flow is corrected at each point of a particular drainage run by a factor  $h_o/(h_o-h)$ , where  $h_o$  is the initial height of the surface of the pulp suspension above the wire and  $h$  is the height at the point of observation. This involves the assumption that the pad thickness at any time during formation is proportional to the decrease in head.

Independent support for the validity of this method of determining drainage resistance was provided by a series of experiments at different concentrations, in which the final sheets covered a range of basis weight 30–240 g/m<sup>2</sup>. It was found that basis weight and drainage resistance were proportional—as expected.

In assessing the effect of beating on drainage resistance, it is necessary to make the comparisons at constant pressure across the pad. This eliminates the effect of differences in porosity that may arise from the immediate effects of stress on the deformation of the fibre network, but does not necessarily

correct for the smaller time-dependent rheological effects that may arise from differences in pressure history. For each beating point, a pressure/resistance curve was plotted, from which the resistance at a pressure of 20 in water was read. The results were converted to the *average specific*

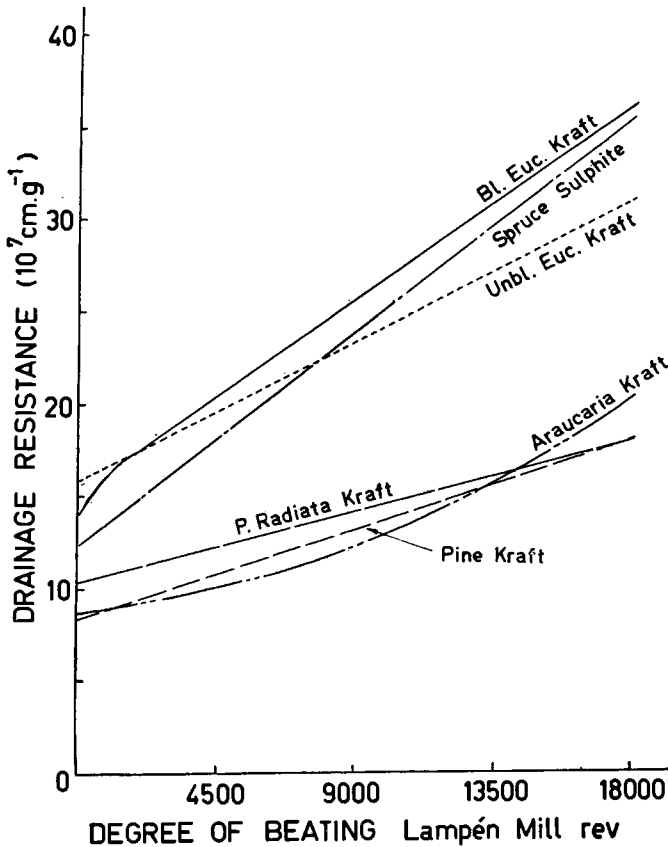


Fig. 7—Change in drainage resistance at constant pressure (20 in water) with extent of beating in the Lampén mill for various pulps

*filtration resistance* as defined by Ingmanson and Whitney<sup>(70)</sup> by working in c g s units and introducing the cross-sectional area of the pad, the fluid viscosity and the mass of particles in the pad.

The influence of Lampén mill beating on the drainage resistance at constant pressure of a number of pulps is shown in Fig 7. Over this beating

range, the deviation from linearity between energy input and drainage resistance is not pronounced. The short-fibred eucalypt kraft pulps show both a higher initial drainage resistance and a higher beating rate than the long-fibred kraft pulps. The highest beating rate is shown by a sulphite pulp, in accordance with the well-known differences in the rate of strength development between sulphite and sulphate pulps.

There is some evidence that the drainage resistance of unbeaten pulps is controlled by the external specific surface,  $S_E$ . From the Kozeny-Carman equation, the permeability should vary inversely with  $S_E^2$ —that is, the drainage resistance should vary directly as  $S_E^2$ , if the porosity is constant. If we consider the pad of unbeaten fibres as composed of a laterally random assemblage of long, hollow cylinders of uniform diameter, the effective porosity will be independent of the fibre diameter. The water-filled lumen would not be expected to affect the flow of liquid past the fibre to any significant extent, since the tubes are very long compared with their internal radius and they lie at rightangles to the mean flow path. Hence, in the absence of external fibrillation or fines, the effective porosity may be taken as constant and the external specific surface should control the drainage resistance.

Neglecting end surfaces, the external specific surface  $S_E$  will be given by the product of the circumference of the cross-section of the fibre,  $2\pi r_1$ , and the specific fibre length defined earlier, that is—

$$S_E = 2\pi r_1 [1/\pi \rho (r_1^2 - r_2^2)] = 2r_1/\rho (r_1^2 - r_2^2)$$

Values of  $S_E$  calculated from the data of Fig. 6 are given in Table 2, with the corresponding drainage resistances for three unbeaten kraft pulps. The radii  $r_1$  and  $r_2$  were measured on the wet fibres<sup>(47)</sup> and  $\rho$  thus refers to the density of the saturated cell wall. If this is taken as the same for unbeaten pulps prepared by the same pulping process, then the constancy of the quantity in the last column of Table 2 will be a measure of the proportionality between  $S_E^2$  and  $R$ .

TABLE 2—RELATIONSHIP BETWEEN EXTERNAL SPECIFIC SURFACE  $S_E$  COMPUTED GEOMETRICALLY AND DRAINAGE RESISTANCE  $R$  FOR THREE UNBEATEN KRAFT PULPS

<i>Pulp</i>	$r_1, \mu$	$r_2, \mu$	$\rho S_E, cm^{-1}$	$R, cm/g$	$\rho^2 S_E^2/R$
Eucalypt	9.0	4.1	2 800	$1.56 \times 10^8$	$5.0 \times 10^6$
Pine	17.2	11.2	2 020	$0.84 \times 10^8$	$4.9 \times 10^6$
Klinki	22.0	16.3	2 020	$0.84 \times 10^8$	$4.8 \times 10^6$

The values  $r_1$  and  $r_2$  are mean external radius and lumen radius, respectively

The change in drainage resistance was investigated over a wider range of beating than that shown in Fig. 7 for a eucalypt kraft pulp and a pine kraft pulp. Results for the Lampén mill and Valley beater are shown in Fig. 8 and 9, respectively. The linear increase in drainage resistance with beating is maintained for both pulps in the Lampén mill, but it is difficult to see a theoretical basis for this behaviour. The trends were observed at concentrations corresponding to basis weights of 60 and 120 g/m<sup>2</sup>. The more

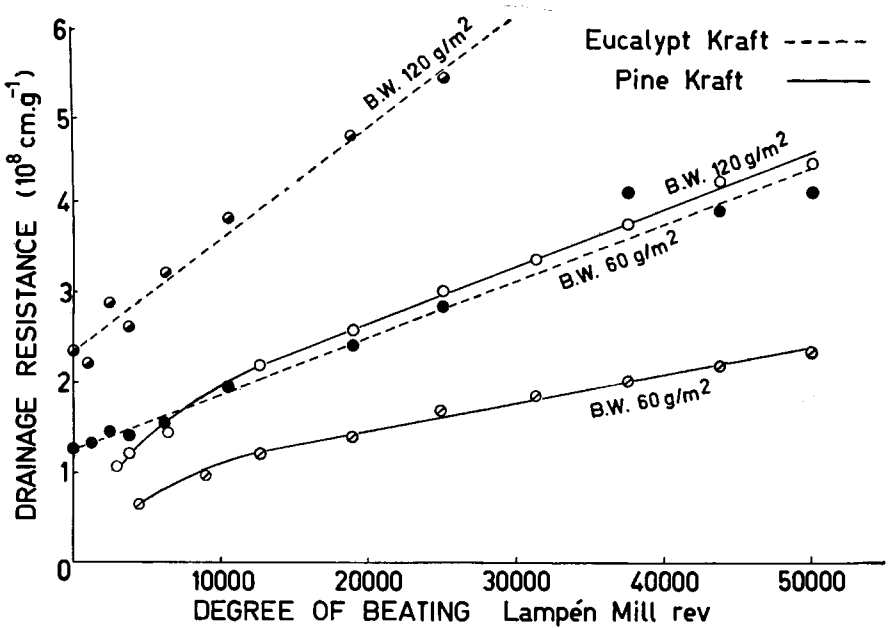


Fig. 8—Change in drainage resistance of eucalypt and pine kraft pulps on prolonged beating in the Lampén mill

usual exponential relationship<sup>(32)</sup> between drainage resistance and beating time was observed for both pulps in the Valley beater. As Ingmanson and Andrews<sup>(32)</sup> point out, the increase depends primarily on the amount of fines produced and this may explain the differences brought about by the two beaters.

The different response to beating of hardwood and softwood pulps, mentioned earlier, is illustrated in Fig. 10, in which the co-ordinates of each point on the curves are the times required to reach a given drainage resistance

in the Lampén mill and Valley beater, respectively. In the earlier stages, the beating rate for the pine pulp, on this criterion, is not very different for the two instruments, although at more advanced stages the Lampén mill beats more slowly. For the eucalypt pulp, the Lampén mill is slower throughout by a factor of 4 or 5. The apparent discontinuity in the curve for the pine pulp suggests that a factor other than the production of fines may also be involved in this behaviour.

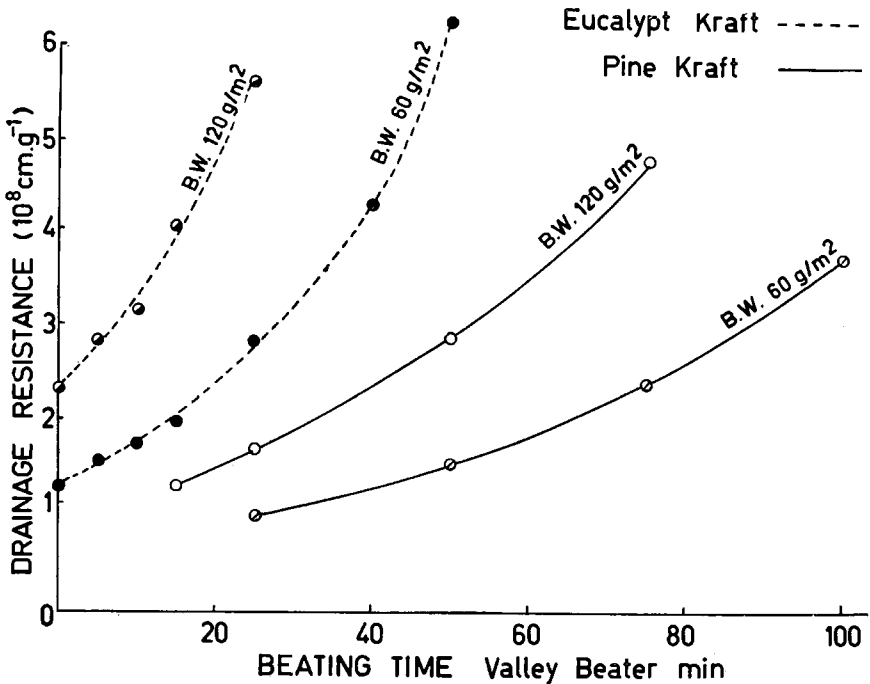


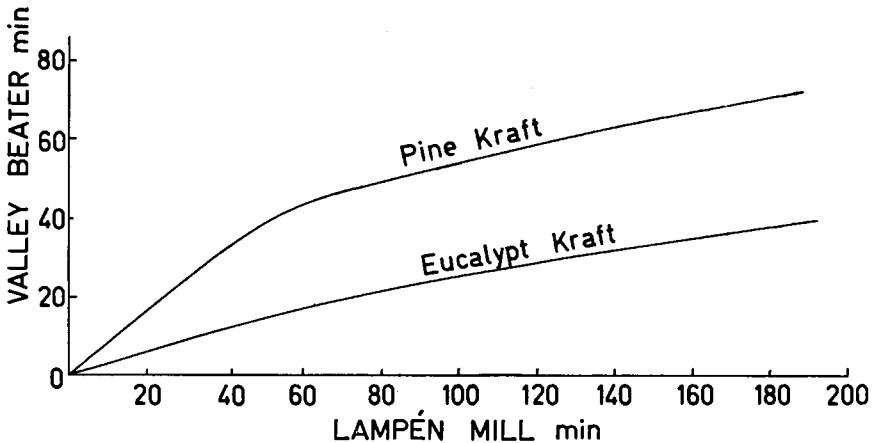
Fig. 9—Change in drainage resistance of eucalypt and pine kraft pulps on prolonged beating in the Valley beater

#### Wet web strength

The strength of the wet web normally increases with beating,<sup>(71)</sup> reaching a fairly constant value for a eucalypt kraft pulp at about 20 000 rev in the Lampén mill, after which, however, extensibility continues to increase up to a value of about 9 per cent at 80 000 rev. In the absence of direct interfibre bonding, the fibres tend to slide past each other when the web is deformed

in tension, this process being resisted by friction and by the Campbell effect.<sup>(72)</sup> Friction will tend to be increased by the external fibrillation brought about by beating and decreased by fibre shortening; surface tensions imparting coherence to the web will tend to be increased both by external fibrillation and by the production of fines during beating.

The sum of these effects is likely to lead to an increased tenacity of the wet web on beating; but, in a process in which fibre cutting predominates, it is possible that the reduction in friction between individual segments would overwhelm the other effects and so reduce the strength.



The co-ordinates of each point on the curves represent the times taken to reach the same drainage resistance

**Fig. 10**—Relative effects of a ball and a bar type beater on the drainage resistance of a softwood and a hardwood kraft pulp

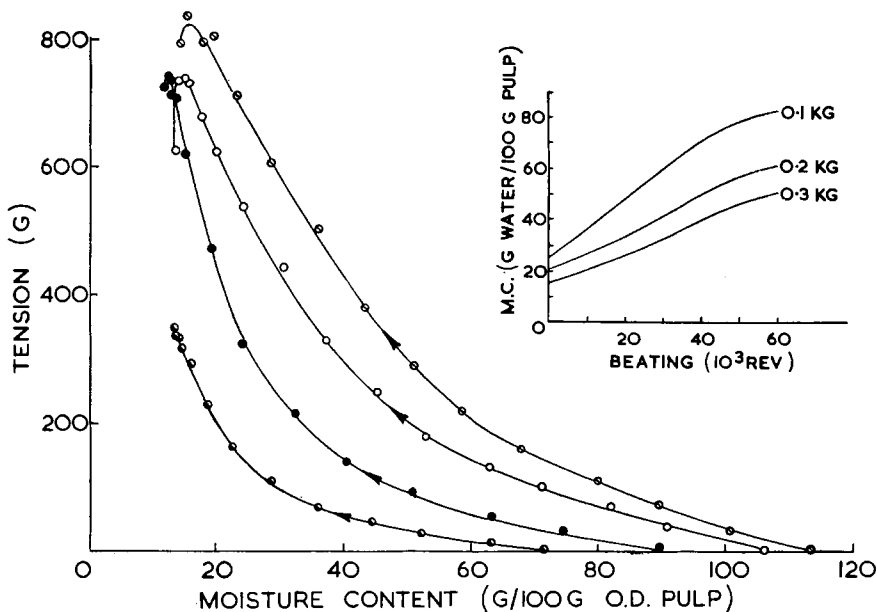
Intrafibre bond breaking with its consequent increase in fibre flexibility and plasticity probably has only a minor effect on the wet web, despite its prime importance for the properties of the dried sheet; however, this point requires further clarification.

#### *Drying tensions*

Beating increases the tension that develops in the sheet upon drying to a given moisture content, as shown in Fig. 11 and in the work of Ivarsson.<sup>(73)</sup> As in the case of wet web strength, perhaps even more so, the surface tension effect of Campbell is of prime importance, particularly in the earlier stages of

drying. Thus, external fibrillation and fines contribute greatly to the increase in drying tensions with beating in paper maintained at constant length or to the enhanced shrinkage on beating in the case of paper kept under constant load.<sup>(71)</sup>

Plasticisation of the fibre on beating, resulting from internal bond-breaking, also plays a part in the development of drying tensions, particularly in the later stages of drying when direct fibre-to-fibre hydrogen bonds are



Numbers on curves refer to Lampén mill rev; the insert shows the effect of beating on the moisture content to which the paper must be reduced to reach the various arbitrary tensions shown on the curves

**Fig. 11**—Influence of beating on the development of tension during drying<sup>(71)</sup>

forming. If the fibres are rendered less resistant to deformation, they are, in the apt phrase of Thode (*personal communication*), “more conformable one to another under the influence of a given level of the Campbell effect. . . . The principal factor involved in making these fibres more conformable is the opening up of the internal structure of the fibre in the beating operation.” A greater number of interfibre hydrogen bonds, rather than intrafibre bonds, will form in the regions of fibre contact. The elimination from the incipient bonded areas of water molecules bridging hydroxyl groups, ring or

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glycosidic oxygen atoms, etc. will result in a dispersion of shrinkage forces or drying tensions over the whole web; with unbeaten pulp, by contrast, local fibre shrinkage takes place, with consequent internal dissipation of tension without its being communicated to the fibre network. Thus, in the case of the highly beaten pulp, the drying sheet tends to behave as a single thermodynamic system; in the case of the unbeaten pulp, equilibrium is approached in a multiplicity of more or less discrete systems with marginal interaction only.

### *Secondary effects: paper*

#### *Bonded area*

AN interesting conclusion from the work of Nordman<sup>(24)</sup> is that beating does not affect the bond strength and that the increase in tensile strength of the paper depends on the increased bonded area and, in certain cases, improved formation only. Ingmanson and Thode<sup>(74)</sup> concluded that at the same total bonded area the tensile strength of a number of pulps, classified and unclassified, was the same, irrespective of degree of refining, amount of fines or degree of wet pressing. When they plotted beating time against various paper properties (tensile strength, z-tensile, tear, fold, apparent density), distinct curves were obtained for classified and unclassified pulps; when these properties were plotted against bonded area, as measured by a modification of the official method, the results were reduced in each case to a common curve. The picture of strength development that emerged from these studies is as follows: "As a pulp is beaten, the fibre surface becomes fibrillated, fines and fibre debris are formed and the degree of swelling of the fibre increases. The only major role of fibrillation and fines in producing strength is to provide greater surface tension forces (the Campbell effect) to draw fibre surfaces into close enough proximity for bonds to be established. The potential area available for final bonding in the dry paper is the original, unbeaten fibre surface and hence is fixed by the original fibre diameter. By increasing the fibre swelling during beating, the fibre flexibility or conformability of the surfaces is increased and hence given surface tension forces during drying will produce more bonded area."

Thus, in Fig. 1, the bonded area is depicted as being mainly influenced by the breaking of intrafibre bonds; fines and fibrillation contribute through surface tension effects.

#### *Density*

Density is closely related to bonded area, but the production of fines during beating has a direct effect on density by filling up the interstices



between fibres. This is shown experimentally by the very large effect of fines on drainage resistance and, as surface tension is not involved at this stage, we may speculate that the direct packing effect may be of the same order of importance as the Campbell effect. The experiments of Marchessault, Lodge and Mason<sup>(20)</sup> showed a decrease in bulk with beating when sheets were freeze-dried, but it was considerably smaller than the decrease with air-dried sheets. In any case, fines may influence density by way of two mechanisms and the effect of fines is therefore shown in Fig. 1 as stronger than that of external fibrillation; both fines and fibrillation, however, normally exert weaker influences than swelling, since the fibres occupy the main bulk of the sheet and their volumetric conformability under pressure therefore becomes the overriding factor.

### *Tenacity*

We may accept the general correlations between tensile strength (tenacity) and swollen specific volume<sup>(33)</sup> and bonded area.<sup>(74)</sup> Again intra-fibre bond-breaking, with its associated fibre swelling, is the primary beating effect of major significance. As sheets made from pulps that have been highly acetylated after beating<sup>(14)</sup> and beaten air-dried sheets<sup>(20)</sup> are considerably stronger than the corresponding sheets made from unbeaten pulps, however, the fibre interactions sponsored by beating appear to have some effect, even in the virtual absence of hydrogen bonding at the fibre surfaces and with low surface tensions. Thus, while inclining strongly towards the general view of strength development on beating favoured by Thode and Ingmanson, we believe that external fibrillation may play a direct part (although a minor one) in this process, one that is distinct from its influence on the level of the Campbell effect. The effect of fines on strength, discussed earlier, is likely to be operative through the increase in bonded area.

### *Extensibility*

Stretch or extensibility must be regarded as one of the most important criteria of the satisfactory behaviour of paper in use. An appreciation of this fact, largely as an outcome of studies on paper rheology, has led to the development of papers with very high extensibility. In reviewing factors influencing extensibility, one is struck by the sensitivity of this property towards tension or compression applied to the paper itself, either in the latter stages of drying or in the dried state. In the simplest case, the breaking of a few bonds at critical points in the fibre network can lead to a 'taking up of the slack' in the rest of the structure. Thus, extensibility is more a network

property than a fibre property. To a certain extent this applies also to the strength properties of paper, but it is more so for extensibility. No single primary effect of beating, taken individually, has an overwhelming influence on extensibility, but beating usually leads to an increase.

Fibres that have been 'opened up' by light acetylation<sup>(14)</sup> or by treatment with dilute alkali<sup>(69)</sup> yield papers with slightly greater extensibility and it is probable that the swelling brought about by beating would have a similar effect. When fibres are heavily acetylated after beating the extensibility of the paper still rises with the degree of beating, possibly because of the effects of external fibrillation. The magnitude of the effect of both intrafibre bond breaking and external fibrillation on extensibility is uncertain, however. The addition of fines has been shown to increase extensibility in some cases<sup>(37)</sup> and it is reasonable to believe that the production of fines on beating would have a similar effect. On prolonged beating, a point is often reached beyond which a large increase in post-yield extensibility occurs: the production of a well-bonded, quasi-homogeneous matrix, in which well-plasticised fibres are embedded permits large deformations to take place without failure.

It appears that fibre cutting, as distinct from fines production, reduces extensibility. This is shown by experiments in which handsheets cut into narrow strips were disintegrated and remade into sheets.<sup>(49)</sup> If one of the factors governing extensibility is the slackness in the fibre network, then fibre shortening, by providing less flexible segments, might be expected to have a deleterious effect on extensibility. Normally, this would be overwhelmed by the positive beating effects; in the case of a long-fibred pulp subjected to a strong cutting action, extensibility might be reduced.

### *Young's modulus*

In Fig. 1, the quantitative relationship predicted by Nissan<sup>(75)</sup> between Young's modulus  $E$  and the number  $n$  of hydrogen bonds per unit volume of material subjected to strain is emphasised by depicting intrafibre bond breaking as having a very strong influence. According to this view, the hydrogen bonds in the amorphous regions of the fibre contribute to the stress/strain relationship and evidence from experiments in which hydrogen bond density was varied by chemical means is consistent with this basic hypothesis.<sup>(76)</sup> Even so, there are indications that supermolecular factors intervene to modify the predicted relationship ( $Ean^3$ ). In particular stress concentrations in the region of bonding between fibres may render the behaviour of hydrogen bonds so located of more significance during deformation than that of the bonds within the fibre. It is difficult to assign

a quantitative measure to the influence on  $E$  of external fibrillation and the production of fines; the absence in Fig. 1 of connections between these quantities should not be taken as meaning that the influences are negligible.

### *Rupture energy*

The area under the stress/strain curve of a strip of paper tested in tension is partly defined by the tenacity and extensibility and to a lesser extent by Young's modulus. Since extensibility is the least conservative of these properties, the factors that determine it largely determine rupture energy also (Fig. 1).

The measured rupture energy is not proportional to strip length,<sup>(77)</sup> although a linear relationship is usually found that allows extrapolation to zero length. Comparisons are often made at a fixed initial length: when this is done, the rupture energy can only be referred to the amount of material between the clamps under the conditions used (basis weight, width, length).

When hydrogen bond density  $n$  is varied by hydroxyl substitution within cellulose fibres, one would expect from Nissan's theory that, to a first approximation,  $n$  would be proportional to the rupture energy of the paper. Owing to supermolecular factors, a linear relationship is not observed experimentally,<sup>(76,78)</sup> but another requirement of the theory—a similarity between the dependence of the predicted rupture energy and Young's modulus functions on the degree of substitution—is fulfilled.

Some direct indications of the primary beating effects influencing rupture energy are given<sup>(14)</sup> by comparing beating curves on (a) normal pulps, (b) heavily acetylated pulps and (c) pulps beaten before heavy acetylation. In the absence of intrafibre hydrogen bond breaking and swelling (b), there is only a small increase in rupture energy initially, which tends towards a constant value, until after prolonged beating another small rise occurs that is probably due to erosion of the more highly acetylated exterior of the fibre. In the case of pulps beaten *before* heavy acetylation (c), there is a much greater initial increase in rupture energy (still far less marked than that for a normal pulp) and a subsequent decline. Complete surface acetylation after beating would be expected to eliminate interfibre hydrogen bonding and most of the swelling effect (in water), so that the initial increase can be accounted for by fibrillation, the subsequent decline by fibre shortening. The normal pulp (a) shows a very rapid initial increase in rupture energy, with three positive effects operating (Fig. 1) and a decline in beating rate as beating proceeds. With some theoretical justification,<sup>(14)</sup> the curve can be approximated by an exponential function, from which a general beating rate can be derived.

Arlov<sup>(79,80)</sup> has carried out a series of interesting experiments on the *shape* of stress/strain curves of fractions derived from a sulphite pulp beaten in four types of laboratory beater. The rupture energy, relative to the whole pulp, was found to depend on the type of beater and it was concluded that the fibre fragments in a pulp beaten in the Lampén mill have a particularly strong influence on the bonding capacity of the pulp as a whole.

### *Creep*

Brezinski<sup>(81)</sup> found that, at constant solid fraction, increased beating resulted in decreased deformation in tests of the same duration and initial stress. The first increments of beating appeared to produce the greatest effect. Similar results were obtained by van Royen.<sup>(82)</sup> It was found also<sup>(81)</sup> that increases in the solid fraction by wet pressing were very effective in decreasing the creep response at the lower solid fractions, but that similar changes were much less effective at higher values of solid fraction.

These observations suggest that the dominant creep mechanism may involve movement between fibres rather than distension of the matrix. Creep is reduced by improved bonding resulting from intrafibre bond breaking, presumably by external fibrillation, which would increase interfibre friction. The influence of fines production on creep is not clear and it has been omitted from Fig. 1. On the one hand, fines might increase density and improve bonding, thus reducing creep; on the other hand, they give rise to paper with a more homogeneous structure, which extends irreversibly in the post-yield region to a much greater extent than does fines-free paper.<sup>(37)</sup> A long-fibred bleached kraft pulp beaten for 40 000 rev in the Lampén mill, however, showed a much lower creep rate than the unbeaten pulp, even under a higher load.<sup>(83)</sup>

Data on the effect of fibre length on creep are limited, but it was found<sup>(83)</sup> that the creep rates of a series of six eucalypt/pine kraft blends beaten together for 3 500 rev in the Lampén mill showed little variation between the two extremes of composition.

### *Tear factor*

Consider the following facts about internal tearing strength—(a) structurally homogeneous materials such as cellulose film have very little resistance to tear compared to paper of the same tensile strength; (b) tear rises with increasing fibre length; (c) tear rises continuously with beating when the sheets are freeze-dried;<sup>(20)</sup> (d) when fibres are highly acetylated after beating,<sup>(84, Fig. 11)</sup> the tear rises more slowly in the early stages of beating,

but then surpasses the value for normally made paper (which reaches an early maximum) and on prolonged beating follows much the same trend as that of the normal paper.

From (a), it would appear that the increased homogeneity resulting from beating tends to decrease tear. Improved bonding in the sheet is brought about by intrafibre bond breaking during beating and the fines produced lead directly to greater homogeneity, as well as increasing the forces of compaction during drying. In the tearing test, stresses are no longer distributed over such a large area and their concentration leads to local failure at lower loads.

From (b), fibre shortening on beating also tends to reduce the tear, partly, again, by increasing stress concentration at the point of test and partly by reducing interfibre friction

In the partial or complete absence of interfibre bonding, as in cases (c) and (d), one of the negative factors is eliminated and tear continues to rise with beating. With acetylated fibres, however, it does not reach the same value as with normal fibres, possibly because a *small* amount of interfibre bonding is advantageous both directly and by bringing the fibres close enough for friction to be fully effective. Eventually, as beating proceeds, the other negative effects (shortening and fines) decrease tear.

This analysis leaves us with external fibrillation and foliation as the primary beating effect exerting the major positive influence on tear (Fig. 1). It is easily understandable that this effect reaches its maximum at an earlier stage of beating than the others. The decline in tear after moderate beating appears usually to be the result of greatly improved bonding, but the negative effects may vary in their relative predominance according to the type of pulp—for example, for a very long-fibred pulp, tear may decrease almost from the very outset of beating as a result of fibre shortening.

Kane<sup>(85)</sup> has approached the effect of beating on tearing strength in a theoretical way. By considering the cases (a)—in which all fibres are pulled intact from the web—and (b)—in which some fibres are not pulled from the web—he concludes that the tearing strength contributed by fibre slippage decreases with beating, but that the tear contributed by elongation increases to some extent.

### *Folding endurance*

Studies on structural factors influencing folding endurance<sup>(37)</sup> have indicated that the presence of fines plays a most significant role. The effect of homogeneity on fold is quite the opposite to the effect on tear: as homogeneity is approached, folding endurance becomes very large rather

than very small. Intrafibre bond breaking leads to improved bonding and external fibrillation probably also improves homogeneity. We thus have three of the primary beating effects exerting a positive influence on fold, with one effect (fines) predominating (Fig. 1).

Fibre length appears to be related directly to folding endurance under some conditions. When the properties of the paper are principally determined by the fibre network (as in papers from lightly or moderately beaten pulps or in those made from pulps deficient in fines), fold is related to tensile strength and to fibre length. Thus, fibre shortening during beating would tend to retard the rate of increase in folding endurance.

In highly beaten papers or in papers made from fine fractions, the predominant structure is that of a more or less continuous matrix, with discrete fibres or an imperfect network making a subsidiary contribution. The high fold exhibited by such materials may be related to high post-yield extensibility.<sup>(37)</sup>

The mechanisms underlying folding endurance have been discussed by Rance<sup>(11)</sup> and Emerton;<sup>(9)</sup> reference should be made also to the work of Andersson<sup>(86)</sup> and of van Nederveen and van Royen<sup>(87)</sup> on this subject.

### Conclusion

THE relationships depicted in Fig. 1 are tentative and will probably be amended eventually. Nevertheless, an attempt has been made to sort out some of the complexities in the light of recent work and it is hoped the result may provide a basis for discussion of the primary effects of beating and the way in which they influence pulp and paper properties.

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## Transcription of Discussion

### DISCUSSION

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PROF. B. STEENBERG: I am going to suggest a mechanism whereby a fibre can be shortened without being cut. A fibre can be divided into two parts either by cutting or by being pulled apart.

A pulp fibre under the influence of viscous drag on a sheared liquid is stressed or compressed, depending on its position as demonstrated by Forgacs, Robertson and Mason (*Fundamentals of Papermaking Fibres*, p. 466). Very strong shear fields indeed must exist to stress a pulp fibre to its breaking load, especially since the fibre moves in the field and the time element, when it is stressed, is rather short. In beating, however, we do not treat suspended fibres individually. In real systems, the fibre under study is always part of a network of fibres. If this network is stressed—for instance, by drag in a shear field—stresses may be transferred from several fibres and somewhere the stress concentration may be large enough to make a fibre break. With this picture, not very strong shear fields should be needed to accumulate stresses in a fibre to exceed its breaking strength.

We have here a case of interaction between the fibres, which could be a possible explanation of the observed shortening of fibres.

It is interesting to note that this hypothesis can also make it understandable why fibres are straightened out when the pulp is beaten. You may assume that the forces that act on a fibre in the network are not strong enough to pull the fibre apart, but large enough to exceed its yield point and straighten the fibre.

In practically all the literature on beating, the relationship between the bars and the fibres is stressed. The interaction between the fibres seems to me a commoner event than an interaction between a bar and a fibre. Using the network concept of fibres in a pulp suspension, we can easily understand a mechanism when this 'rag' is pulled apart, producing both fibre shortening and fibre bruising. If fibres are pulled apart, it is quite reasonable that the ends will be frayed and show fibrillation.

DR. O. ANDERSSON: In a suspension of beaten fibres with the ions removed by ion exchange, the supernatant liquid after the suspension has stood for 5 days at 5°C is still slightly light scattering because of the microfibrils it contains. On hydrolysis, it analyses roughly to 50 per cent glucose and 50 per cent xylose.

### *Primary beating effects*

DR. H. CORTE: I should like to correct a remark by Steenberg. My published data were histograms with a  $\frac{1}{3}$  mm class width. The frequency of the shortest fibres ( $0\text{--}\frac{1}{3}$  mm) increased steadily with beating—the experiments did *not* have the results he said they had.

MR. D. H. PAGE: I would like to mention here, in connection with Steenberg's suggestion, some work that we have carried out and is as yet unpublished. We have attempted by high speed flash microscopy the direct observation of fibres in the beating zone of a beater. A hole has been drilled radially through one of the beater bars and plugged with a transparent plastic. It has been possible to obtain pictures of the beating zone showing clearly the fibres and the faces and edges of the rotating bars. Two facts of interest have emerged—the leading edges of the bars do not have the fibrage associated with them that Sigurd Smith predicted and, secondly, in normal operation, there are very few fibres in the beating zone at all. It seems therefore that the interaction mechanism in the shear field proposed by Steenberg cannot account for fibre shortening in this case.

*Written afterthought:* I assumed that Steenberg's suggestion referred to the interaction on fibres in the shear fields that occur when two bars face each other and it is this possibility only that our experimental results would not support. There is nothing in the experimental results to suggest that Steenberg's mechanism might not be operating because of shear fields outside the region that we can see and, in particular, when the bar edges are approaching.

MR. E. J. JUSTUS: Measurements of beater bar clearance in a Jordan when running have been made and found to be considerably greater than when the Jordan was stationary. Presumably, any refining action on the fibre was due to fluid shear rather than to mechanical working by the beater bars.

DR. J. A. VAN DEN AKKER: A number of years ago, Sigurd Smith did some careful study work on the passage of fibres between the bars of a beater and, in his very interesting book, *The Action of the Beater*, describes the phenomena that might account for the so-called cutting or fibre shortening resulting from the high compressive and shear stresses existing between the bars. I was very interested in Page's remarks: what was the consistency in these experiments?

MR. PAGE: About 3 per cent. If you work out the consistency in the beating zone, it would be very much lower than that.

## Discussion

MR. P. H. DIXON: To try to tie up the questions of Justus and Van den Akker to Page's statement, we must know four things—consistency, bar width, gap between bars and speed—before we can deny Steenberg's theory.

PROF. J. D'A. CLARK: At the risk of oversimplification, may I say that I feel that, if two metal bars come together in water with fibres present, only three possible things can happen—the fibres can be shortened, the fibres can be split and the fibres can be bruised. Nothing else can happen and it is up to us technologists to relate all the beating phenomena to those three factors. I am old enough to have been at the meeting when Sigurd Smith gave his paper on fibrages at the Royal Society of Arts. In more recent years, I have grave doubts whether we really do or can get a fibrage on the bar edges at the speeds at which they go through the stock, because of turbulence.

MR. T. TREVOR POTTS: I, too, was at that meeting and there was a fourth thing that can happen—nothing! This is the reason for the beating process being so little more efficient than when Sigurd Smith addressed us at the Royal Society of Arts.

PROF. STEENBERG: There is all this talk about the relationship between bars and the fibres, but there is actually a fifth thing that can happen. The relationship among the fibres themselves, which has been neglected.

PROF. CLARK: It is three *effects* that I detailed, not three causes.

PROF. B. G. RÅNBY: I would mention that there is considerable support for the following views. We can look upon fibre cutting in a shear gradient as a molecular phenomenon by comparison with polymer solutions. If we force a polymer solution through a small orifice we need a certain rate of shear gradient, a certain minimum length of the chains and a certain minimum concentration to obtain breakage of the chains. The breakage of fibres in a suspension being refined would be a complete analogy to these concepts and I think there is nothing to prevent us from at least accepting the possibility that fibres break owing to shear in suspension. To what extent it happens is another matter.

DR. CORTE: How does Steenberg think the probability of such a cutting mechanism would depend on the fibre length? Experimentally, one can determine the probability of a cut occurring in a given time and how it depends on the fibre length. This is technically important. With a purely cutting

### *Primary beating effects*

mechanism, apart from this mechanism (which I have never thought of), it is quite logical that the cutting probability is proportional to the length of the fibre, which means that the *longest* fibres are in the greatest danger. With any other mechanism, it may be that the cutting probability is independent of the fibre length. In this case, the *most frequent* fibres are in the greatest danger, which in natural fibre length distributions are the shortest.

MR. H. G. HIGGINS: Could this pulling apart of the fibre be detected microscopically?

PROF. STEENBERG: I am limiting myself here to the exposition of a hypothesis. We are working experimentally on this approach and the results will be published.

DR. M. KROFTA: In this discussion of the beating process, the main talk has been about the influence of steel bars and knives. The shear action of the water has hardly been mentioned. The highly turbulent movement of the stock suspension produces strong shear forces in the water that have an important influence in the beating process.

### *Written contributions*

MR. E. J. HOWARD: Concerning the beating process, there is one factor that has been overlooked, namely, the probability that under hydraulic shear *colloidal* particles of hemicellulose and cellulose can be torn from the fibres and subsequently redeposited and thus act as bonding agents. Andersson pointed out that he had found xylan-glucan micelles of 150 D.P. in the supernatant water after beating. Other workers have shown that wood hemicelluloses can go into solution and later be re-adsorbed by fibres during cooking and caustic refining processing of wood.

MR. H. G. HIGGINS: Certainly, colloidal particles can be removed from the fibre, but the extent to which they are redeposited is problematical. Re-adsorption of hemicelluloses during alkali cooking is rather different in that mechanisms exist whereby they can be forced out of solution—for example, change in alkali concentration, reduction in solubility resulting from rupture of xylan-uronic acid linkages (*see* Yllner, Hamilton and others). We have found that extraction of the water-soluble colloids from a highly beaten eucalypt kraft pulp by repeated washing has only a small effect on the mechanical properties of the paper—much less than the effect of removing fine fibrous material. Although treatment with alkali produces larger effects,

## Discussion

these are in part dependent on physical changes in the cellulose (*see* Centola and Borruso, McKenzie and Higgins and others). It is also possible for the removal of hemicelluloses to have a greater effect on the bonding capacity of a fibre than their redeposition on either an alkali-extracted or a normal fibre. In the native fibre, they can assist in holding the fibrillar elements apart, which reduces the tendency of the cellulose to crystallise, increases the swelling capacity and plasticity of the fibre and permits interfibre hydrogen bonding at points close to the covalent skeleton of the larger cellulose molecules. Redeposited material, on the other hand, cannot restore or impart an amorphous character to the more crystalline regions of the cellulose and, where it occurs in multi-molecular layers, its cohesiveness would be impaired by the lack of reinforcement that cellulose chains of high D.P. would provide.

On this view, the effectiveness of an added bonding agent would depend on the extent to which an even and thin spread could be attained in the areas of contact—a condition quite analogous to that required in glueing surfaces of wood and other materials and one unlikely to be easily fulfilled when dealing with papermaking fibres. The recognition of hydrogen bonding as the main source of interfibre bonding leads to the conclusion that, provided sufficient lateral conformability of the fibres is assured by beating or other means, added bonding agents are of secondary importance. When the conformability is insufficient to lead to a high bonded area, however, additives may be of value in reducing stress concentrations at fibre junctions. Some materials often regarded as strengthening agents may depend for their efficacy more on their effect on flocculation than on their direct bonding action. The point that Howard has raised is an interesting one that warrants further study.