

# A STUDY OF FIBRE-TO-FIBRE BONDING BY DIRECT OBSERVATION

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## **Synopsis**

*An examination has been made of the size and shape of fibre-to-fibre bonds and their frequency of occurrence in paper sheets. Definitions have been proposed for parameters associated with these bond properties. The effects of beating and drying tension on these parameters have been investigated and the likely effect of other paper-making variables has been considered. The significance of the parameters in controlling the physical properties of paper is discussed.*

## **Une étude par observation directe des liaisons entre fibres**

*Une étude a été faite sur la taille et la forme des liaisons entre fibres, ainsi que sur leur fréquence dans les feuilles. Des définitions ont été proposées pour les paramètres associés à ces liaisons.*

*Les effets sur ces paramètres de l'engraissement et du séchage ont été examinés et on a pris en considération l'effet semblable d'autres variables de la fabrication du papier. La signification de ces paramètres, en ce qui concerne les propriétés physiques des papiers, est exposée.*

## **Studium der Zwischenfaserbindungen durch direkte Beobachtung**

*Die Grösse und Form der Zwischenfaserbindungen und ihre Häufigkeit innerhalb des Papierblattes sind studiert und Definitionen für die von diesen Zwischenfaserbindungen abhängenden Eigenschaften entwickelt worden. Man untersuchte den Einfluss von Mahlung und*

*Trocknungsspannung und stellte Überlegungen über die wahrscheinliche Auswirkung anderer Variablen bei der Blattherstellung auf. Abschliessend wurde die Signifikanz der Parameter bei der Kontrolle der physikalischen Eigenschaften von Papier diskutiert.*

### Introduction

UNTIL recently, evidence concerning fibre-to-fibre bonding in paper sheets has been circumstantial and opinions have been formed about its nature by deduction from a variety of macroscopic data, including mechanical and optical properties of paper sheets. From evidence of the fall in strength characteristics when paper is immersed in solvents with high hydrogen bonding capacities,<sup>(1)</sup> it is clear that hydrogen bonds must play a large part in providing strength; this was first suggested by Huggins in 1936.<sup>(2)</sup> (No evidence exists to the authors' knowledge that hydrogen bonds are solely responsible.) The results do not give any indication, however, of the site of action of the hydrogen bonds responsible for sheet strength, but merely indicate that they are so positioned and sufficiently weak for them to be broken when strained during immersion in a hydrogen-bond-breaking medium. This important issue appears to have been avoided in the past and one can search in vain amongst much of the published literature for an indication of the site at which bonding forces occur.

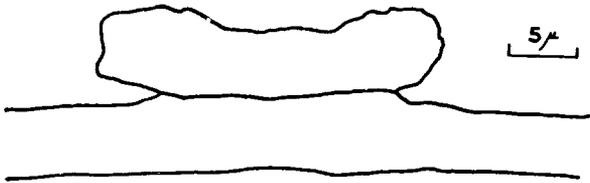
It has been suggested that fibrillation (generally taken to mean fibrillation that can be observed in the light microscope) is responsible for sheet strength by spanning between the fibres,<sup>(3,4)</sup> although the experimental evidence that strength increases rapidly during the early stages of beating without a corresponding increase in fibrillation tends to contradict this. Another school of thought believes that plasticity in fibres when wet governs the strength of the dry sheet by giving a greater opportunity for contact between fibres,<sup>(5-7)</sup> but this theory has been merely qualitative and no indication has been given of the precise meaning of the phrase 'contact between fibres'. Until recently, the only work on this appears to have been by Asunmaa and Steenberg<sup>(8)</sup> who, using the electron microscope, showed that areas of close contact between fibres existed, with a maximum area of around 100  $\mu^2$  in pulps that were not wet pressed.

Over the past two years, we have carried out work that has enabled a completely new approach to be made to the subject of fibre-to-fibre bonding. If discrete areas of bonding exist between fibres, they are, in principle, observable by light microscopy, since bonding implies the replacement of two air-to-cellulose interfaces by a single cellulose-to-cellulose interface that scatters and reflects virtually no light. The practical attainment of this possibility was

shown by Page<sup>(9)</sup> and introductory work has been carried out by Page and Tydeman.<sup>(10)</sup>

Briefly, the technique consists of an examination of the paper specimen at medium power in vertical illumination. The contrast in the image by this simple method is normally much too low; but, by incorporating dyed fibres within the sheet and by using polarised illumination with a crossed analyser, the contrast is enhanced to a useful magnitude. It should be emphasised here that the technique reveals regions of optical contact between two adjacent fibres and that in most papers these regions are large and numerous.

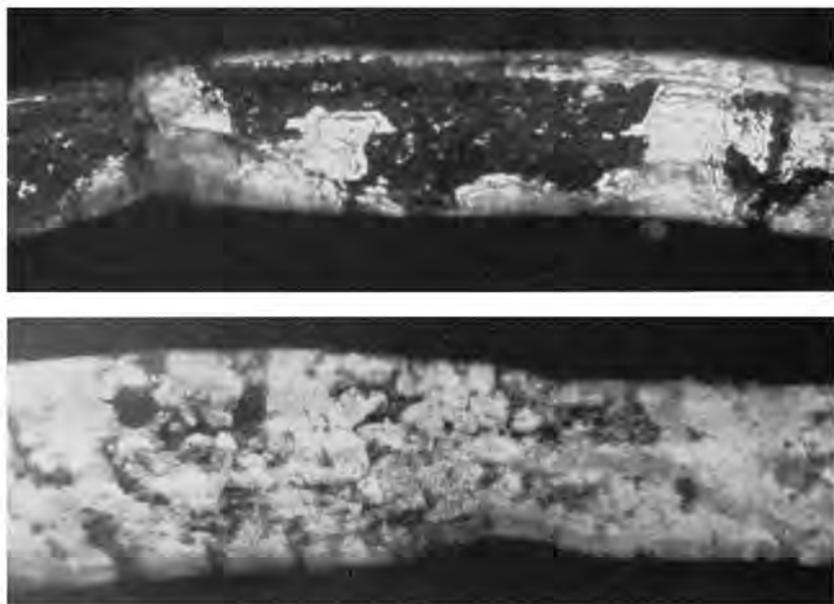
It is important to consider the origin of these areas of optical contact. A dried fibre is normally extremely irregular in shape and for areas of optical contact to form between two adjacent fibres considerable changes in the natural shapes of one or both of the fibres must have occurred. This is evident from Fig. 1, which is a reconstruction of the area of contact between two fibres. The drawing here is to scale, showing the roughness of the fibre



*Fig. 1*

generally, but the line of contact joining the two fibres has necessarily been made 10 times thicker than the maximum separation that can occur between the two fibres if optical contact is to be maintained. The areas of optical contact seen in the microscope are usually perfect; low order interference fringes around them are rare; when they do occur, they are closely spaced, implying that the fibre surfaces move away steeply from the zone of contact as shown in Fig. 1. There are two possible mechanisms by which areas of optical contact may form. Collapse of one fibre on to another may occur because of the large forces of wet pressing and surface tension. In this case, the area of contact will depend on the local plasticity of the fibres. Alternatively, the contact areas may be developed by the accumulation at the regions of crossing, again owing to surface tension, of residual water that, on drying out, will deposit its dissolved and suspended material. In this case, the area of contact will depend on the concentration of this material in the water. Both mechanisms appear to contribute, but the first is thought to be the more important, at least during the early stages of beating.

It can be shown that these areas of contact occur during the drying of the fibres and are not caused by wet pressing alone. Fibres dried down on to a glass slide without any wet pressing force show quite large areas of contact (Fig. 2*a*). On the other hand, a fibre freeze-dried on to a slide (Fig. 2*b*) shows only very small contact areas in an irregular pattern, presumably in regions where the fibre originally touched the glass. It is clear that these optical contact areas contain the sites of action of the hydrogen bonds—and perhaps other forces of adhesion—that are principally responsible for the mechanical



**Fig. 2**—Fibre-to-glass bonds—  
(*a*) formed during natural drying  
(*b*) formed during freeze-drying  
(Fig. 2*b* taken by A. Simpson)

strength of paper. The distribution of adhesive forces within these optical contact areas may by no means be an even one, but it seems highly unlikely that over appreciable areas within this optical contact zone there should be a total absence of bonding. Work is proceeding with the electron microscope to shed more light on this.

Work has now been carried out to investigate the geometrical arrangement of these fibre-to-fibre bonds in paper sheets and to assess the variables

that will change them. The initial work on the geometry of bonding was confined to an examination of the effect of beating, also the effect of drying sheets under tension, since both these factors are known to affect the load/elongation curve of paper markedly and might thus be expected to affect the degree of bonding between fibres.

### Experimental

HANDSHEETS, of nominal basis weight  $60 \text{ g/m}^2$ , were made from a bleached spruce sulphite pulp with freenesses of 670 and 310 CSF, corresponding respectively to 0 min and 20 min beating time in the Valley

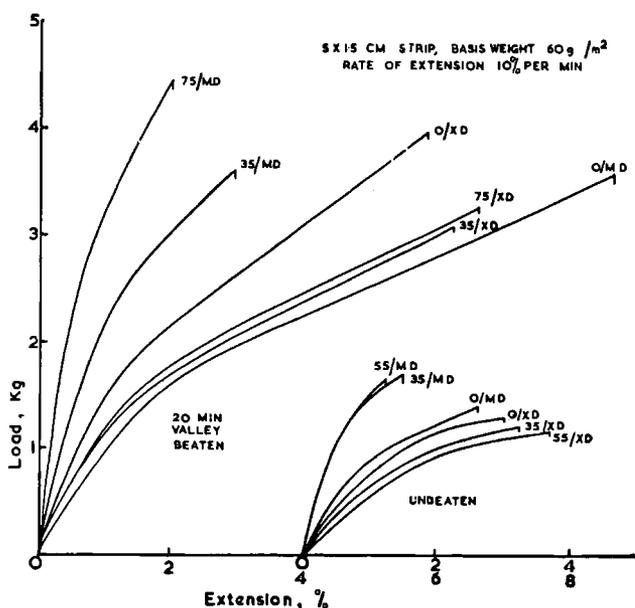


Fig. 3—Load/extension curves of strips used in the experiment—the figures on each curve indicate the applied drying tension in g/cm

beater. Of this, 70 per cent of the fibres were then dyed with Chlorazol Black, a treatment that had earlier been found to have no effect on the mechanical properties of the pulp. The sheets were dried under tension by the method of Burkitt, Kenworthy and Gates,<sup>(11)</sup> there being three levels of tension for each degree of beating. After drying the sheets, strips 5 cm long and 1.5 cm wide were cut from a central area within which it has been shown that the

mechanical properties are constant. The load/elongation curves for individual strips cut parallel (M.D.) and transverse (X.D.) to the direction of applied drying tension are shown in Fig. 3.

The original intention in this work was to obtain a random sample of bonds in the paper sheet. The plan was to use a system of random co-ordinates to choose the field of view in the microscope and to select the undyed fibre nearest to the centre of the field. The bonds contained in a section of this fibre were to be photographed and used for measurement. In practice, it was found that this method was not unbiased. The efficiency of the technique of observation of bonds varies from fibre to fibre and even from bond to bond; for early wood fibres, which collapse into ribbons, the technique is extremely good, but the visibility of bonds deteriorates for fibres that deviate from this form. The bonds in late wood fibres are often ill-defined and it is difficult in some cases to determine whether or not a late wood fibre is undyed. This effect has resulted in a preferred selection of early wood for the overlying fibre, although, of course, the underlying fibres remain random. Because of the tedious nature of this work, there was a tendency to select regions along the fibres that contained several bonds. Much of the early work was affected in this way, but, whenever these biased results are quoted, the implications will be discussed.

A different method of sampling the bonds is now being used and it is hoped that the bias will be reduced to a minimum, although it is considered impossible to eliminate it owing to the above-mentioned imperfections of the technique.

Micrographs of the selected field were then taken on 35 mm film and notes were made of the configuration of fibres responsible for the bonds examined within the field. The micrographs were projected on to paper at a final magnification of  $\times 1\ 060$  and drawings were made accurately of the bonds and the fibres concerned. An example of such a field and its associated drawing is shown in Fig. 4. From these drawings, a large number of measurements were made and the results will now be given together with some important qualitative observations.

## **Results**

### *Configuration of bonds*

As discussed in an earlier publication,<sup>(10)</sup> two essentially different bond configurations exist in paper containing ribbon-like fibres. In addition to the straightforward case of two fibres crossing and bonding, a situation occurs when a third fibre falls partly over such a crossing and the common area of crossing between it and the first fibre is partially obscured. These two cases

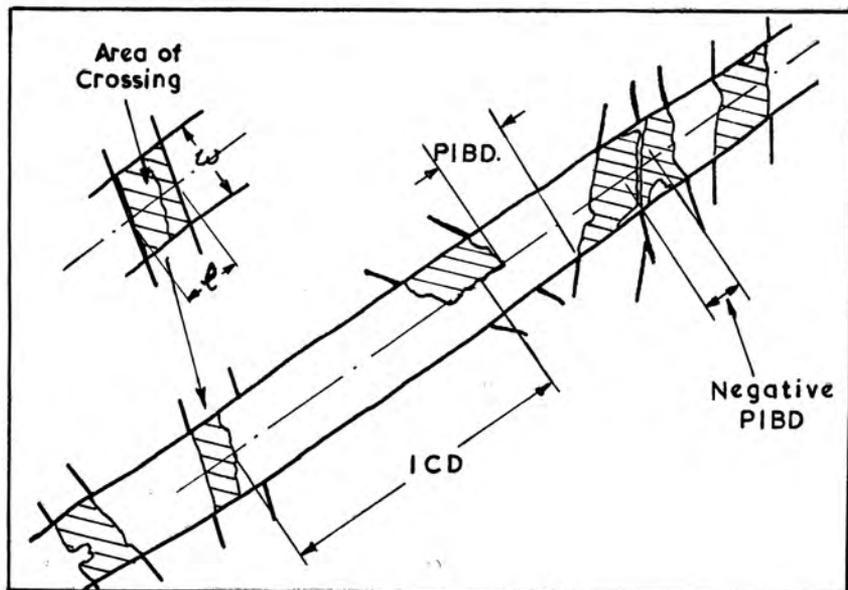
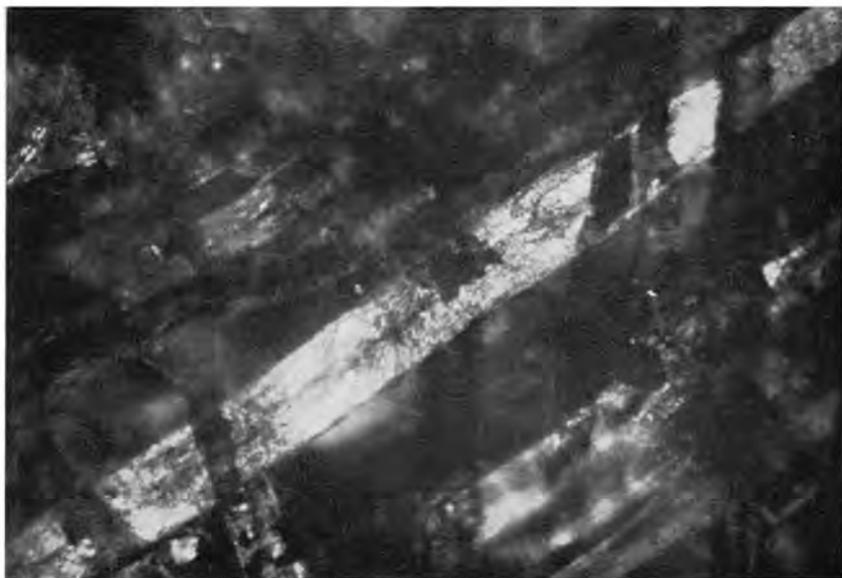
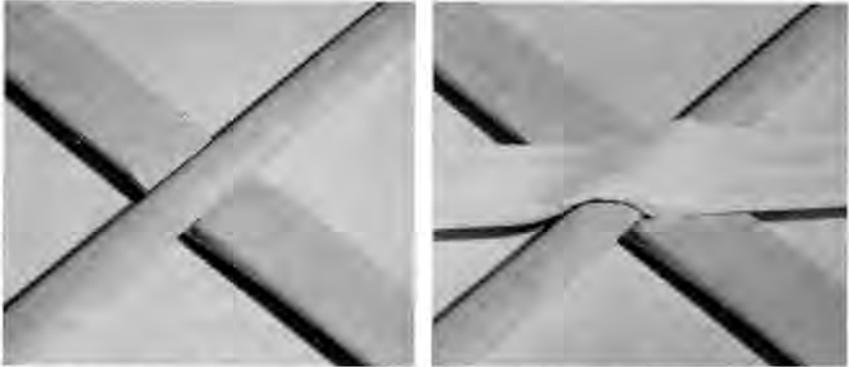


Fig. 4—Example of a field with its associated drawing from which measurements were made

are illustrated in Fig. 5. On the left, a simple fibre-to-fibre bond is shown and on the right an obstructed bond between the lower and upper fibre is formed when the third fibre is laid down. In future, the terms *simple* and *obstructed* will be used to describe bonds having these configurations (these correspond respectively to types I and III and type II defined in the previous publication). It has not been found possible to classify every bond into these categories owing to the difficulty of observing the configuration of the underlying fibre and approximately one fifth of the total bonds were unclassified.



**Fig. 5**—Models illustrating, on the left, simple and, on the right, obstructed fibre-to-fibre bonds

Fig. 6, 7 and 8 are line drawings traced from photographs of typical bonds in softwood pulps, the black areas representing optical contact between the fibres outlined. The first seven drawings of Fig. 6 illustrate the formation of obstructed bonds, whilst the remainder of the drawings illustrate simple bonds. Even in this latter case, the whole of the common area of crossing is not usually bonded and a whole range of degrees of bonding exists from virtually 100 per cent as exemplified by the last five drawings of Fig. 6, down to quite a small percentage.

#### *Size and shape of the fibre-to-fibre contact areas*

The extent of bonding occurring at a crossing is clearly of prime importance in determining both the mechanical and optical properties of a sheet. It has already been explained<sup>(10)</sup> how the surface topography of the fibres and their inflexibility can be responsible for incomplete bonding. Furthermore, it was suggested that the extent of bonding varies with beating.

One of the consequences of the surface roughness and fibre inflexibility is

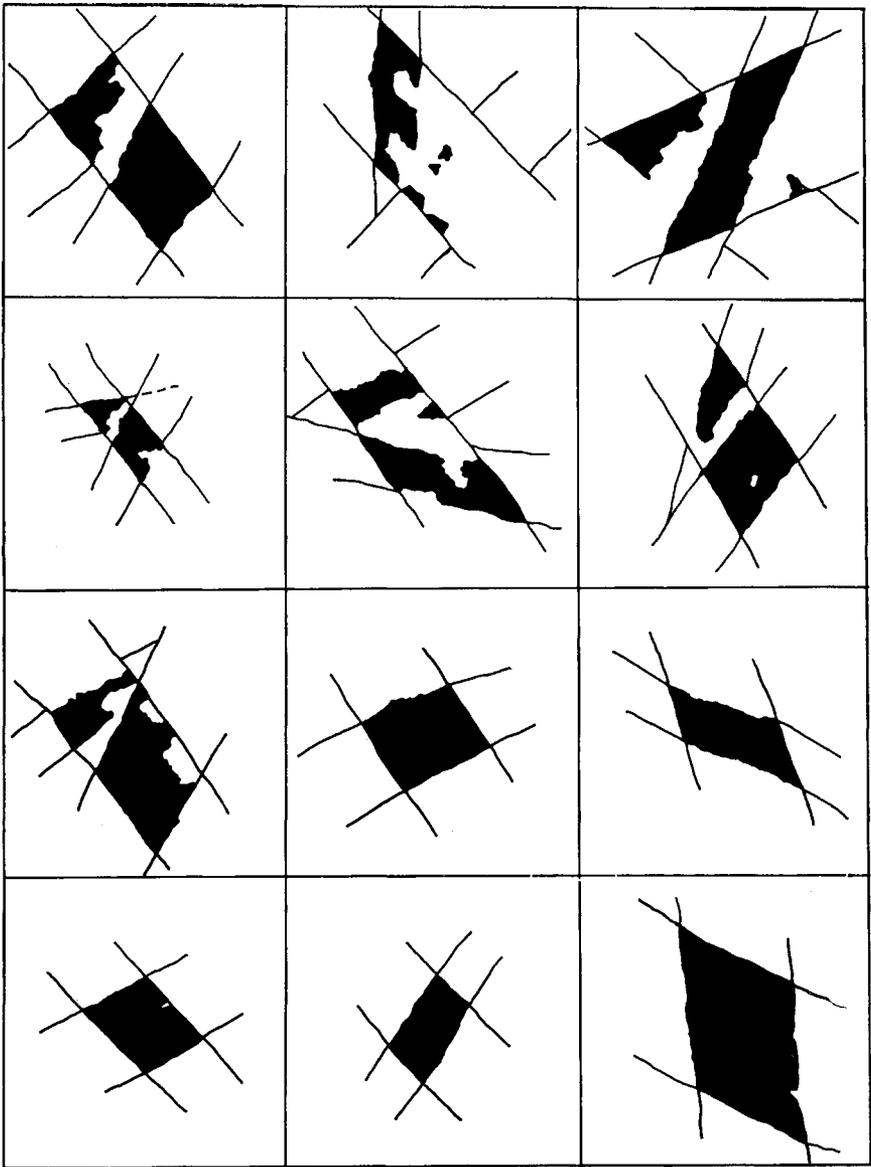


Fig. 6

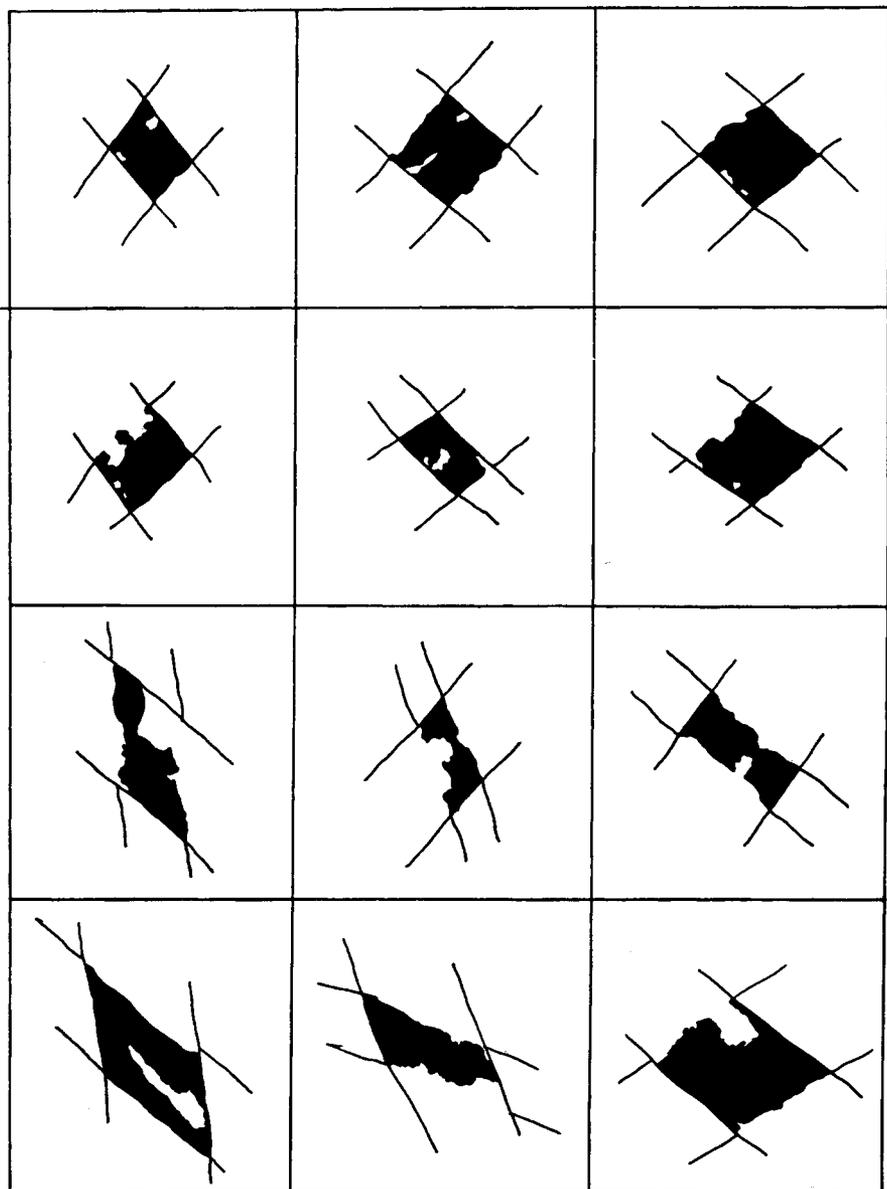


Fig. 7

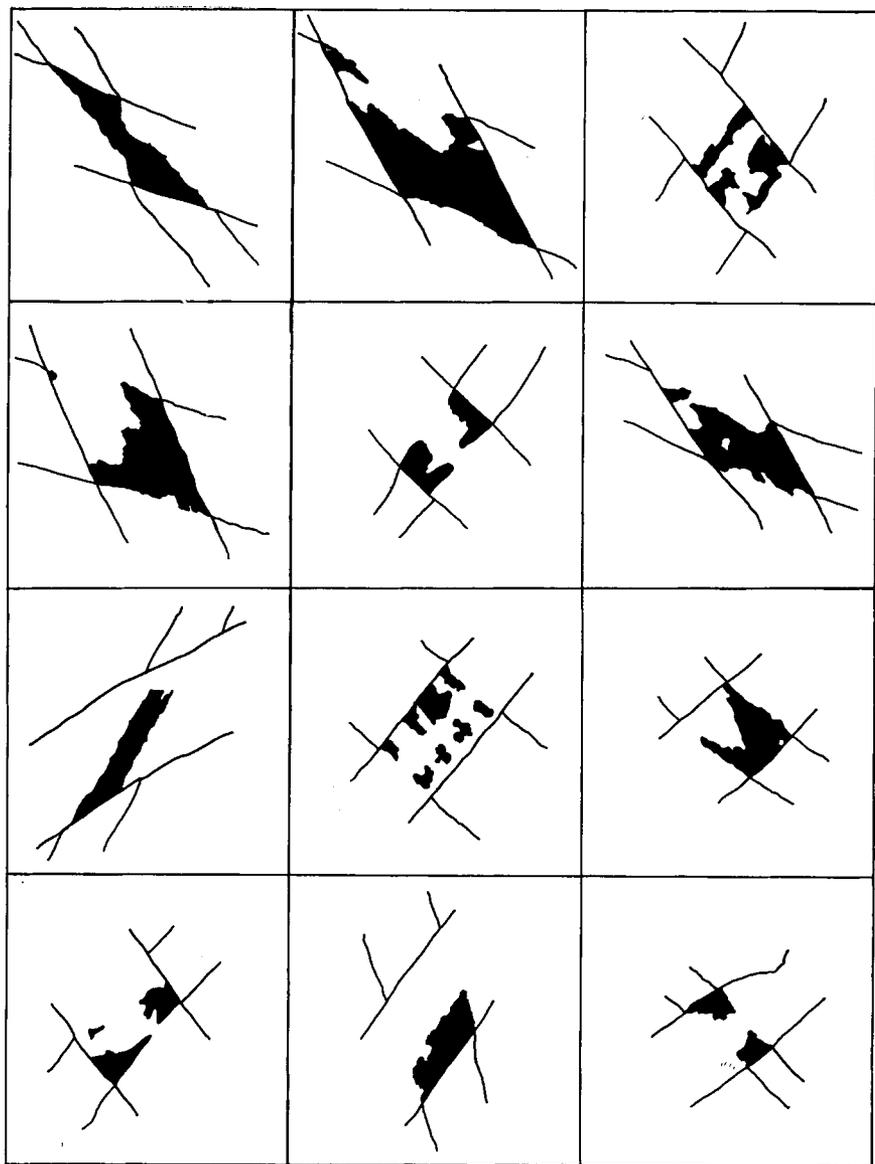


Fig. 8

that a bond may consist of more than one area of contact. Examples of this phenomenon are shown in some of the fields of Fig. 6, 7 and 8. Quantitative determinations have been made of the frequency of occurrence of such *multi-point bonds* and the results are shown in Fig. 9. In the unbeaten sheets, 45 per cent of the bonds show more than one area of contact; a reduction in frequency occurs with increasing number of points of contact, although two bonds with seven points of contact were observed in this survey. In the beaten sheets, only 12 per cent of bonds show multi-point contact and of these 9 per cent had only two areas. It thus seems likely that beating affects the local plasticity of fibres sufficiently to allow the individual areas of contact to merge

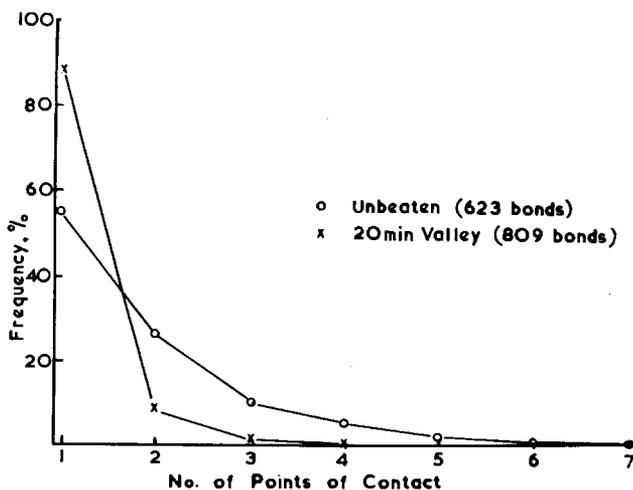


Fig. 9

together, increasing the degree of bonding of a crossing and it was thought desirable to measure this in a more quantitative manner. The *degree of bonding* was adopted as a basis of measurement and is defined as the ratio of the bonded area to the projected area of the crossing between the two fibres and expressed as a percentage. The bonded areas in the drawings made from the micrographs were measured by planimetry and the projected areas of the crossing calculated from the dimensions  $w$  and  $l$  indicated in Fig. 4. The results of this work are given in Table 1. The effect of drying tension at constant beating time is not consistent, although some significant differences are observed. In the case of simple bonds, the mean degree of bonding of unbeaten, freely dried sheets is significantly greater than both the tension dried sheets

(at the 1 per cent and 5 per cent levels, respectively). On the other hand, for the beaten sheets, the mean for the sheet dried at 35 g/cm was higher than that dried at 75 g/cm at the 5 per cent significance level. For the obstructed bonds, no significant differences were detected. It is concluded that the effect of drying tension on degree of bonding is small and, if significant, tends to give lower bonding at higher drying tensions.

TABLE 1

Beating time, min	Drying tension, g/cm	Degree of bonding, %	
		Simple bonds	Obstructed bonds
0	0	46.6	27.8
	35	38.0	22.0
	55	40.3	25.3
20	0	71.6	48.4
	35	75.1	42.5
	75	69.0	41.0
0	All	41.7	25.1
20	All	71.8	43.7

In view of this, the results have been compounded within the two beating times between which there is clearly a highly significant difference. For clarity of interpretation, a division is made between simple and obstructed bonds. The upper diagram of Fig. 10 shows the effect of beating on the degree of bonding of simple bonds. It is clear from the change in distribution that the effect of beating is to develop the degree of bonding. Whereas in the unbeaten pulp half of the bonds have a degree of bonding lower than 40 per cent, in the beaten pulp, three quarters of the bonds are more than 60 per cent bonded. For the obstructed bonds, there is a similar upward trend with beating. However, as the degree of bonding is defined as the fraction of the geometrically projected common area, obstructed bonds are never capable of 100 per cent contact and thus, for each bond, there is a maximum degree of bonding that lies between 0 and 100 per cent, depending upon the geometry of the three-point crossing. The effect of this is apparent from the second diagram of Fig. 10.

A further parameter of importance is the absolute size of each area of contact between fibres. The results obtained from the planimeter measurements of the drawings are given in Table 2. It is apparent that there is a highly

significant increase in the size of both simple and obstructed bonds with beating. The effect of drying tension is again not marked, the only statistically significant results being that the simple bonds in the beaten sheets are larger

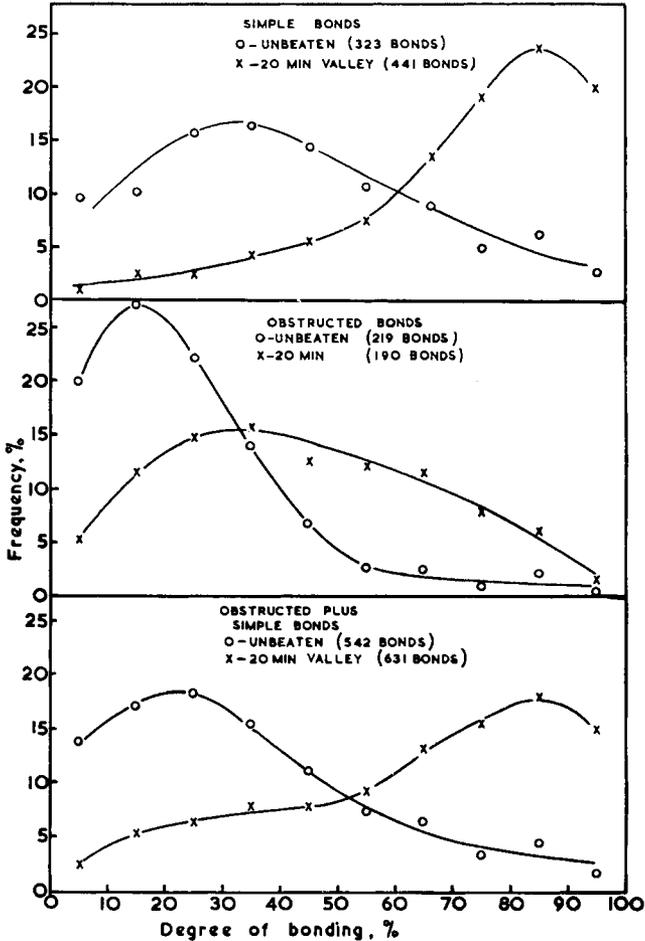


Fig. 10

at 35 g/cm drying tension than those at 75 g/cm and that the obstructed bonds in unbeaten sheets freely dried are larger than in sheets dried under 35 g/cm. If there is an effect of drying tension, it thus appears to decrease the bond size.

The frequency of occurrence of bond sizes is shown in Fig. 11 for all bonds including unclassified.

The results of this examination of the size and shape of fibre-to-fibre bonds are clearly affected by the inevitable preferential selection, mentioned

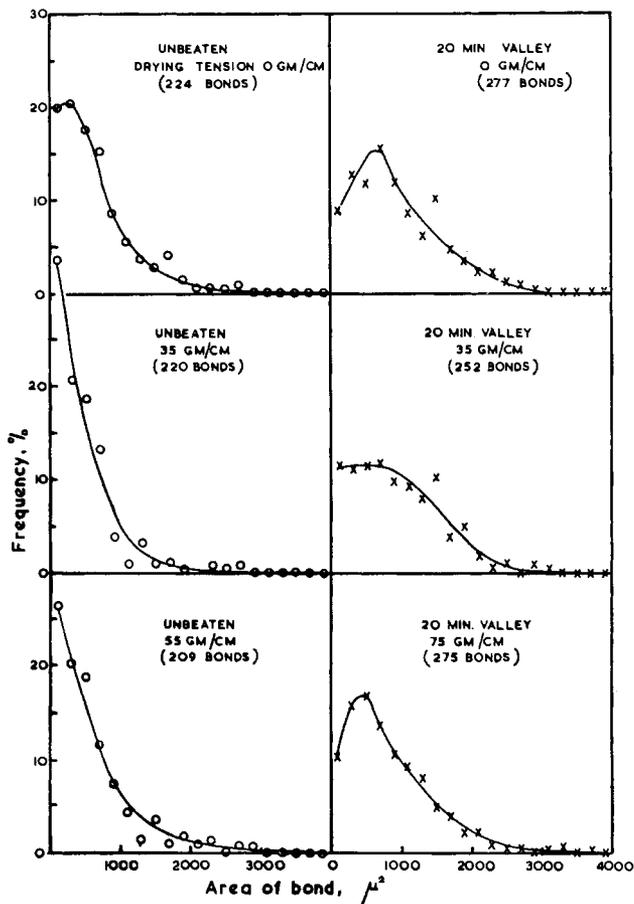


Fig. 11

earlier, of fibres that give bonds with a greater visibility. These are, of course, usually those fibres of early wood that, having thinner cell walls, collapse to form wide ribbons. The mean width ( $44 \mu$ ) of the fibres selected for observation was 40 per cent higher than that for the underlying fibres ( $31 \mu$ ). Although this bias will seriously affect the absolute magnitude of the above variables,

TABLE 2

Beating time, min	Drying tension, g/cm	Mean bond area, $\mu^2$		
		Simple bonds	Obstructed bonds	All bonds*
0	0	772	536	643
	35	656	331	483
	55	708	455	572
20	0	1 102	715	956
	35	1 199	610	989
	75	1 003	651	856
0	All	712	440	567
20	All	1 099	656	932

\* Including unclassified

their comparison at different beating times and drying tensions is unlikely to be affected to any great extent, since the same bias operates on all specimens.

The parameter most seriously affected by this selection is the bond size. Since the mean area of the crossings is 40 per cent higher than would be obtained from a truly random selection, it would be expected that the values quoted for bond size are too high by this effect alone. In addition, there is the possibility that the crossings between wider fibres, being thinner walled, are capable of a greater degree of bonding, which would result in a further over-estimation of the bond size.

The effect of bias on frequency of multi-point contacts is obviously complex. The wide early wood fibres usually develop larger numbers of ridges giving more points of contact within the bond, but this might be offset by their greater potential flexibility. These factors contribute also to a bias in the degree of bonding.

### Interbond distances

The distance between bonds along a fibre is very important in determining sheet properties, since, together with the degree of bonding, it governs the optical properties as well as influencing all the mechanical properties. The term *free fibre length* has been used elsewhere in this connection<sup>(12)</sup> to mean the distance between the centres of fibre crossings. This would not now seem to be a satisfactory term for general use in the consideration of paper structure, as the size of the bonds is often appreciable compared with the distance between them, therefore the distance between crossing centres is not entirely

'free'. The distance between the centres of bonded crossings (equivalent to the *free fibre length* of Corte and Kallmes only when all crossings are bonded) is a fundamental dimension and has been termed the *intercrossing distance* (I.C.D.). However, owing to the size and complicated pattern of the bonded areas, the unbonded length of the fibre available for deformation will clearly be in general much less than this. Moreover, it is not possible to make any one definition of *equivalent free fibre length* that is universally useful in relating the mechanical properties of the sheet to its geometrical structure, since the dimension required will depend on the type of deformation undergone by the fibre whether it be extension, bending, torsion or shearing. It was necessary to make some choice of parameter to be measured in this work that would be in some way representative of a free fibre length. The measurement chosen has been defined as the *projected interbond distance* (P.I.B.D.) and is the distance, measured in the direction of the fibre, between the projection of the bonds on the fibre axis. It should be realised that P.I.B.D. is the smallest reasonable measure that can be made and, in general, equivalent free fibre lengths will be greater than this. The definitions of I.C.D. and P.I.B.D. are illustrated in Fig. 4.

Initially, measurement of I.C.D. and P.I.B.D. were made from the drawings, but it was felt that these measurements were much more sensitive to bias in the choice of fields. The work was therefore repeated employing direct measurement with an eyepiece graticule, using a better method of selection of the fibres and by measuring along the whole of the visible length of each fibre. So far, sufficient results have not been obtained to separate the effects of drying tension, but results for I.C.D. at the two beating degrees are shown in Fig. 12. The mean values are  $81.0 \pm 2.3 \mu$  and  $59.0 \pm 1.3 \mu$  respectively, for unbeaten and 20 min Valley beaten pulp (95 per cent confidence limits of the means). The reduction in I.C.D. represents an increase in the number of bonds per unit length of the fibres, a result that strongly supports the theory of the increase in non-local flexibility of the fibres during beating. It is thus possible to define another important parameter, *the frequency of bonding*, which is the number of bonds per unit length along the fibre. The frequency of bonding for these pulps is, respectively, 12.3 and 16.9 per mm, implying that the effect of 20 min Valley beating is to increase the total number of fibre-to-fibre bonds in the sheet by about 37 per cent.

Different furnishes, having different fibre lengths, would be expected to have different frequencies of bonding, even if they had geometrically similar structures. The frequency of bonding for geometrically similar structures is inversely proportional to the fibre width, implying that a more fundamental definition can be made. The mean number of bonds per length along the fibre

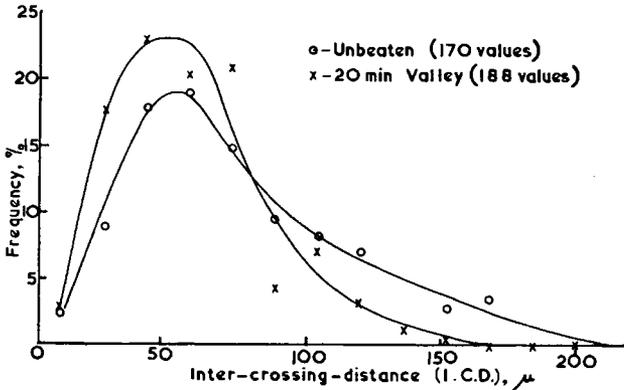


Fig. 12

equal to the mean fibre width can be defined as the *specific frequency of bonding*. This number is dimensionless and is a basic geometric property of the structure. Furthermore, it can be shown to have a maximum, for a random arrangement of fibres, in the region of 1.4 if every fibre crossing is bonded.

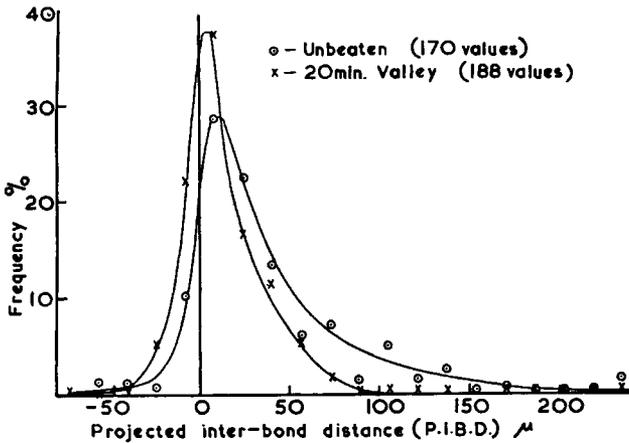
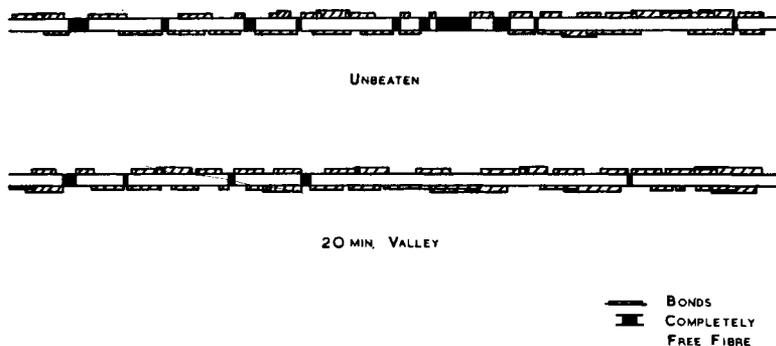


Fig. 13

Converting the above results for the unbeaten and beaten sheets, the specific frequency of bonding is 0.38 and 0.52, respectively. (It is interesting to note that esparto with its very loose structure gives a value of about 0.2.)

The results for similar work on projected interbond distance for the two beating times are shown in Fig. 13, the means being  $34.0 \pm 2.1 \mu$  for unbeaten and  $13.2 \pm 1.0 \mu$  for the beaten pulps. The P.I.B.D. is negative when the projections of adjacent bonds overlap and this occurs more frequently in the beaten sheets because of the higher degree of bonding. Beating also considerably decreases the incidence of very large values of P.I.B.D., as would be expected from its observed effect on I.C.D.

The measurements of interbond distances reported above were made between bonds on one side of the fibre only, since it is essential for observation that the upper surface of the fibre should be free. To a good approximation,



*Fig. 14*

however, it can be assumed that fibres in the body of the sheet display on both sides bonding similar to that measured for the single side of the observed fibres. An estimation of the interbond distances within the sheet has been made by the superimposition of two sets of measurements, one set representing the upper surface of the fibre and the other the lower surface. This is illustrated in Fig. 14, in which the hatched regions represent bonds drawn from the measured values of I.C.D. and P.I.B.D. Values of the 'double-sided' interbond distances were measured from these drawings (Fig. 14 shows only a small section of the constructions for the unbeaten and beaten values). The double-sided results are shown in Fig. 15 and 16; the mean values of I.C.D. being  $39.6 \mu$  and  $30.4 \mu$  for unbeaten and beaten, respectively and, for the P.I.B.D.,  $-6.8 \mu$  and  $-15.2 \mu$ , respectively. It is evident from Fig. 14, 15 and 16 that fibres in the body of the paper sheet are bonded at frequent intervals along their lengths and indeed that an overlap of bonds occurs to such an

extent that there is hardly any length of fibre that is not bonded on one side or the other.

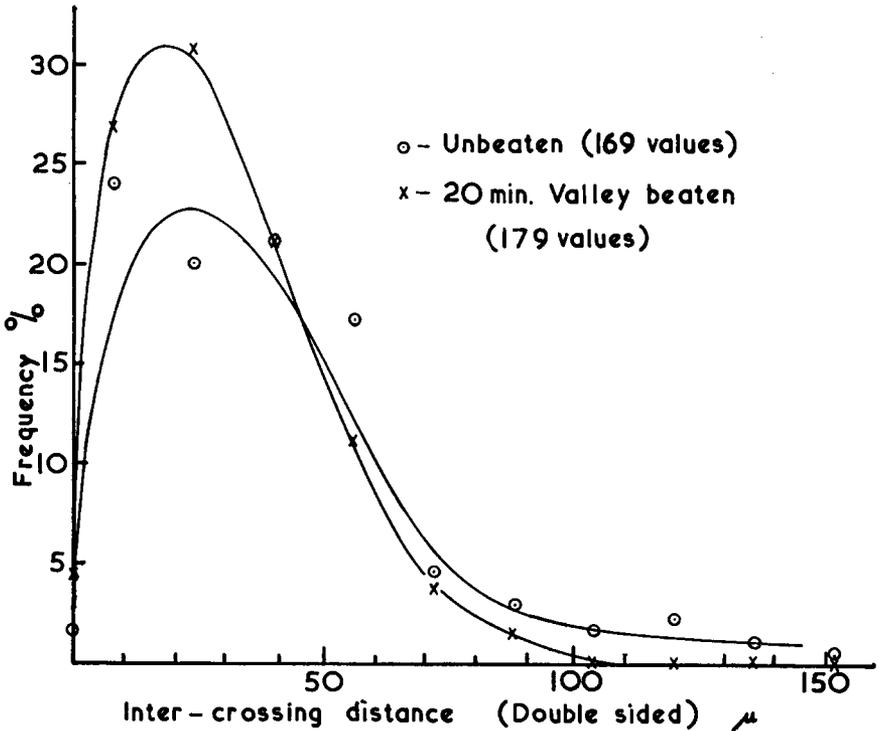


Fig. 15

### Discussion

THE variables available to the papermaker wishing to change the physical properties of his sheet consist mainly of furnish, beating, pressing and drying conditions: it is of value to consider the effect of these variables on the parameters previously defined in this work. Two of these variables have been examined, but it is worthwhile indicating at this stage the likely effect of the others.

There is obviously an overriding effect of furnish on the absolute bond size, since fibre width is a controlling factor—for example, the mean bond size in esparto would be a fraction of that in a softwood furnish. Furthermore,

the surface topography and plasticity of fibres in different furnishes will have a major effect. It has been shown that beating increases the bond size of a softwood furnish and it is to be expected that wet pressing would have a

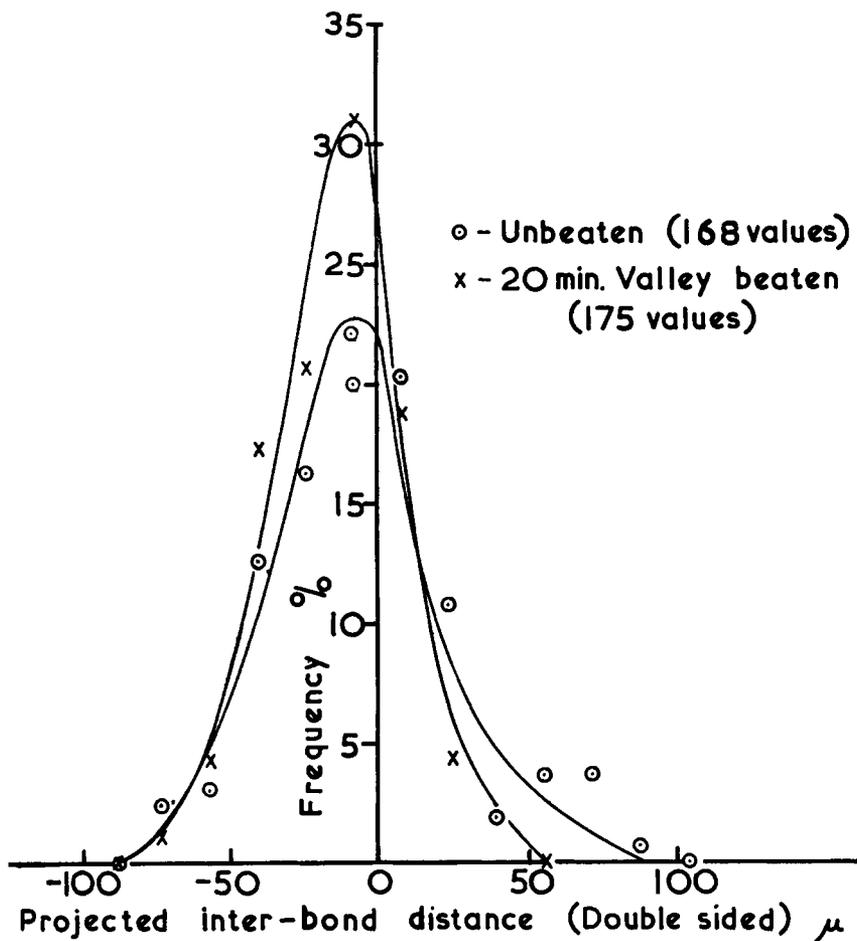


Fig. 16

similar action. On the other hand, drying tension, which has a major effect on the physical properties of the sheet, appears to have only a small effect on the bond size and, if anything, tends to reduce it.

The absolute values of interbond distances are also highly sensitive to

furnish, being dependent not only on the fibre width, but also on the fibre flexibility. Beating decreases the distances between bonds (because of the increase in fibre plasticity) and the action of wet pressing might be expected to give a similar result. Any effect of drying tension appears to be smaller than we have been able to detect in the limited results obtained so far.

It would be expected that the combined effect of bond sizes and distances between them would control many of the physical properties of paper. Opacity (more exactly scattering coefficient) is the most striking example of a property almost entirely controlled by these parameters, since they determine the fraction of the fibre surface bonded. The percentage of fibre surface bonded is given by—

$$\text{Percentage bonded} = \frac{\text{Mean bond area} \times 100}{\text{Mean fibre width} \times \text{Mean I.C.D.}}$$

and this can be rewritten in the dimensionless form—

Percentage bonded =  $\phi \times$  degree of bonding  $\times$  specific frequency of bonding where  $\phi$  is the ratio of the mean area of crossing to the square of the mean fibre width and is in the region of 1.1 for the pulp examined. The value of  $\phi$  for random sheets of other pulps will not deviate significantly from this. The high opacity of esparto can thus be attributed to the low specific frequency of bonding (it seems likely also that the degree of bonding is low, although the technique of observation cannot be used to obtain a reliable measure of this for cylindrical fibres). Beating the softwood pulp increased the fraction of fibre surface bonded from 16 per cent to 36 per cent and this increase is attributable to increases in both the degree of bonding (by 65 per cent) and the specific frequency of bonding (by 37 per cent). These higher values indicate the lower opacity of such pulps particularly at higher degrees of beating.

The specific frequency of bonding is closely correlated with the density of the sheet; the bulky esparto sheet exhibits a low specific frequency of bonding, whereas a high specific frequency of bonding is found in the dense sheet of a well-beaten softwood pulp. Wet pressing, by increasing the density, would be expected to increase the specific frequency of bonding in a similar way to the effect of beating.

Although the increases in bonded area and number of bonds with beating could reasonably account for the associated change in the stress/strain curve, no similar effect on bonding has been observed with drying tension, which also markedly changes the shape of the stress/strain curve. It must therefore be concluded that the stress/strain curve derives its shape from parameters that have not so far been included in this geometrical approach. The significance of this result will be discussed in a later piece of work.

It is envisaged that work along these lines will proceed with the object of a fundamental elucidation of the structure and physical properties of paper and it is hoped that the structural parameters defined in this work have provided a basis for future theories.

### Glossary

<i>Fibre-to-fibre bond</i>	The total area of optical contact between two fibres
<i>Multipoint bond</i>	A fibre-to-fibre bond consisting of more than one discrete area of contact
<i>Area of crossing</i>	The common projected area of two fibres
<i>Obstructed bond</i>	A fibre-to-fibre bond in which the total geometrical area of crossing is not available for bonding due to the material intervention of a third fibre
<i>Simple bond</i>	A fibre-to-fibre bond in which the area of crossing is entirely available for bonding
<i>Degree of bonding</i>	The ratio of the bonded area to the area of crossing, expressed as a percentage
<i>Intercrossing distance</i>	The distance between the centres of the areas of crossing of two adjacent bonds
<i>Projected interbond distance</i>	The distance measured in the direction of the fibre between the projection of adjacent bonds on to the fibre axis
<i>Frequency of bonding</i>	The number of bonds per mm of fibre
<i>Specific frequency of bonding</i>	The number of bonds per length (along the fibre) equal to the mean fibre width

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## Transcription of Discussion

### DISCUSSION

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PROF. G. JAYME: Hunger and myself wish to congratulate Page and his co-workers on the very interesting results they have obtained. However, on account of the evidence obtained by us, it would appear that the 'optical contact areas' shown by Page contain numerous parts where the contact is not 'optical'. The areas are not black throughout, but show grey and even white spots. We are inclined to believe therefore that all types of bond including bonds by microfibril lamellae and 'strings' are present in these areas.

MR. D. H. PAGE: The light microscope technique itself is not easy. There are many reasons that these areas should not appear black, simply because of the local optical conditions. Light is scattered and reflected within the fibre throughout which the observation is made and from fibres below and around it. I have dealt with all these contrast effects in my first publication on this subject.

When we have a springwood fibre collapsed, however, we have the ideal optical condition for looking at bonded areas; then we find that the areas are uniformly black in spite of the surface roughness of the fibres. I agree there is room for disagreement to some extent here; but, because there is light and shading, it does not disprove our contention. I agree the onus is on us to prove that it is good contact and our electron microscope work has done that.

MR. H. W. EMERTON: I would add to this remark that at every interface there will be a measure of depolarisation. Some of the incident plane polarised light will become elliptically polarised and this will not be extinguished by the analysers.

PROF. H. W. GIERTZ: When we speak about the adhesion between fibres in the contact area, we should also pay attention to the quality of the surface material. We have spoken mainly about microfibrils on the surface, but there may also be some hemicellulosic material there that could drastically change the behaviour at the contact surface. If these hemicelluloses are swollen and partly dissolved, they will give rise to very strong adhesion forces when the water is removed by drying and they will act as an adhesive. The amount of such surface hemicellulose will depend on the way of preparing the fibres (*see p. 605*).

## *Fibre bonding*

MR. P. H. J. ABBOTT: What is the effect of moisture content on optical contact? One would imagine that, as the moisture content went up, the area in optical contact would increase and that this would be associated with a loss of mechanical strength. On this basis, the use of area of optical contact as a measure of bonded area would appear doubtful.

MR. PAGE: It depends on what you mean by moisture content; but, considering normal ranges of moisture content due to relative humidity changes, no effect has been observed. There is no basis therefore for any doubt.

PROF. J. D'A. CLARK: Because of surface tension forces, surfaces fibrillated when wet will appear smooth after drying. There is thus danger in observing photomicrographs of dried fibre surfaces as smooth when in reality, before drying, they were covered with fuzz and fibrillae that contributed to the bonding. There appears no evidence of fibrillae on the outer surface of a primary wall in water with a silvering technique, but this becomes apparent on the secondary wall immediately the primary wall is removed. This explains the rapid increase in strength of unbeaten fibres with beating.

A DELEGATE: Surprisingly, the same weight of a high yield pulp (about 70 per cent yield) has the same wet web strength as has a normal (perhaps 50 per cent) yield pulp on a papermachine in the same admixture with other pulps. Since there are far fewer fibres in the 70 per cent yield pulps for the same percentage weight in a mixture, some reason must be found that these fibres, present in far fewer numbers for the same percentage weight, should give such a very high proportion of wet strength. The only way to explain it is by some such supposition as Giertz has advanced that these pulps are commonly used unbeaten.

PROF. A. H. NISSAN: I wonder whether we do not sometimes create problems for ourselves by insisting on one mechanism to the exclusion of all others. Do we not really get all the different types of bond discussed today contributing to papermaking? Molecules are on all surfaces and when, through any agency, they come within certain distances from other surfaces, there will be a number of energies of interactions—electrostatic, hydrogen bonds, van der Waals' bonds, even some covalent bonds. In other words, there may be a large number of mechanisms. Sometimes, when arguing this among ourselves and each bringing evidence in support of his point of view, are we not simply unconsciously choosing evidence that illustrates the mechanism we favour?

## *Discussion*

There was a note about hemicelluloses in *Nature* this last August by Prof. Preston of Leeds University: he analyses the microfibrils and finds some differences in chemical composition between what one might call the amorphous portion and the crystallite. It is worth studying in connection with this discussion, because once again there is the possibility of different 'bonds', even between 'pure' cellulose.

PROF. B. G. RÅNBY: There is no justification for calling a hydrogen bond of the type now under discussion an ionic bond: it is not. It is actually a dipole bond. It can be argued in the case of carboxylic acids, which form two very strong hydrogen bonds. The strongest hydrogen bond we know of—for example, in hydrogen fluoride—can be ionic in nature, but this is another matter. We should call the hydrogen bonds in cellulose dipole bonds.