BONDING STRENGTH OF INDIVIDUAL FIBRES

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MICROSCOPIC examination of the nature of paper failure, when tensile stresses are imposed upon the sheet, has shown that two important factors governing paper strength are the strength of individual fibres and the strength of bonding between fibres. A satisfactory method of determining individual fibre tensile strength has been published by this laboratory.[1] Using this method, the changes in strength of individual fibres of Loblolly pine springwood and summerwood holocellulose were followed during extraction with alkali of increasing concentration. The results of this work are reproduced in part in Table 1.[1]

TABLE 1—TENSILE STRENGTH OF UNEXTRACTED AND ALKALI-EXTRACTED
LOBLOLLY PINE HOLOCELLULOSE FIBRES

Treatment	Individual fibre strength, g/fibre	Cross-sectional area, μ^2	Fibre strength per unit area, kg/mm^2
Summerwood			
Chlorite holocellulose	51	504	102
Peracetic acid holocellulose	50	568	88
Dimethylsulphoxide	56	679	84
0·1 N KOH	54	596	92
0·4 N KOH	53	540	97
1·5 N KOH	34	483	70
3·0 N KOH + 0·75 M H_3BO_3	31	464	68
6·0 N NaOH	30	479	64
Springwood			
Chlorite holocellulose	15	322	48
Peracetic acid holocellulose	17	386	46
Dimethylsulphoxide	15	427	34
0·1 N KOH	13	340	39
0·4 N KOH	14	313	47
1·5 N KOH	11	334	34
1·5 N KOH + 0·75 M H_3BO_3	10	274	35
3·0 N KOH + 0·75 M H_3BO_3	7	260	27
6·0 N NaOH	7	277	28

The strength per unit area of summerwood fibres was found to be about twice that of springwood fibres. The reasons for the low strength of spring-wood fibres were thought to be related to the extensive pitting on the fibre walls or to the relatively low proportion of the springwood fibre wall com-posed of the middle layer of the secondary wall (S2 layer).

Comparison of strength data with carbohydrate analyses of the pulps showed that a sharp decrease in strength coincided with an abrupt decrease in xylan content. It was tentatively concluded that the hemicellulose composi-tion of the fibre was of importance to the internal strength of the fibre and
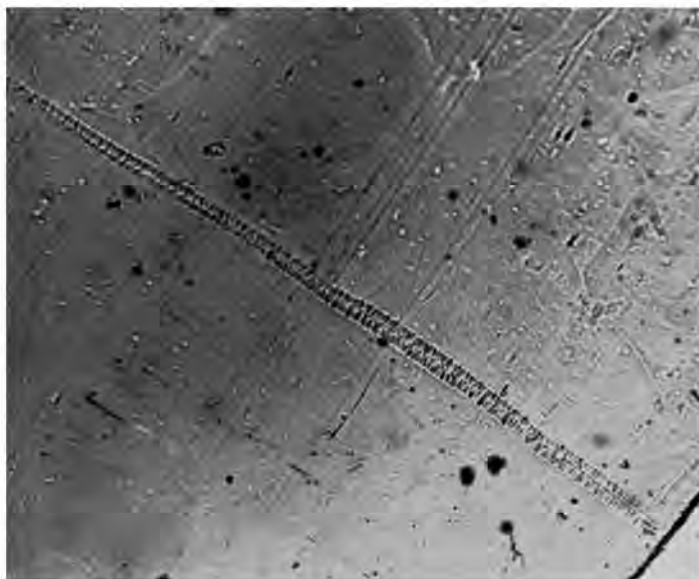

Fig. 1—Replica of summerwood Loblolly pine fibre on cellulose
film following release of the fibre × 50

might well be of equal importance to the fibre–fibre bonding. Work was therefore extended to the development of a method of determining bonding strength by bringing fibres into contact, bonding them under carefully controlled conditions and measuring the strength of the bond.

As an approach to the problem, a method was at first conceived of bonding fibres, under pressure and heat, to unplasticised cellulose film and measuring the force, applied axially, to dislodge the fibre. Measurement of bonded area in this case was determined after fibre release from the replica of the fibre

formed on the cellulose film during bonding (Fig. 1). Later, work was extended to determining the stress required to separate single fibres bonded to shives (5–15 fibres wide) and other single fibres. In this method, bonded
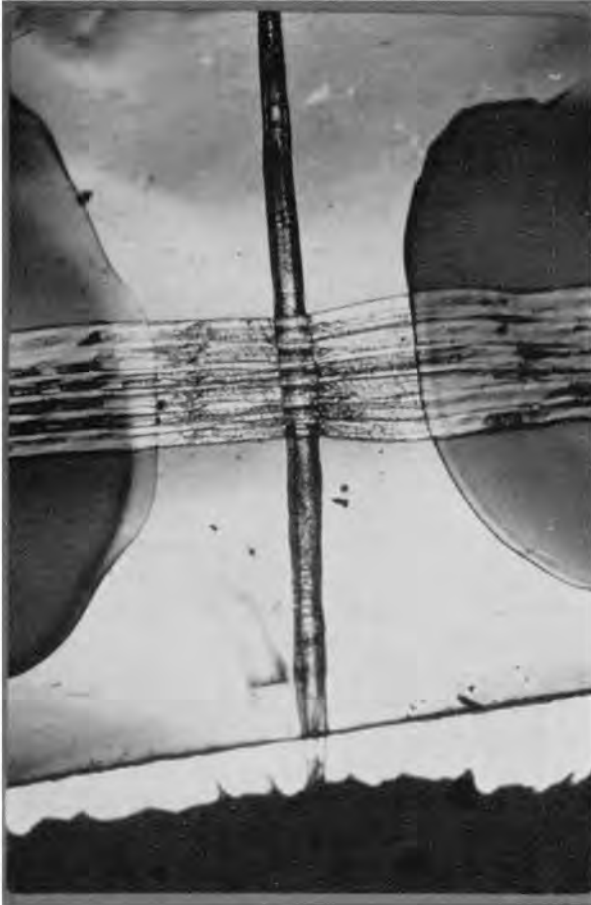

Fig. 2—Loblolly pine springwood holocellulose fibre bonded to springwood fibre shive—shive is glued to cellulose film ready for testing × 50

area was measured in polarised transmitted light before tension was applied to separate the fibres (Fig. 2 and 3). Details of the method will be published shortly elsewhere.

Investigations were carried out on unrefined springwood and summer-wood fibres of Loblolly pine cooked separately by the peracetic acid holo-cellulose method. Results so far obtained are given in Table 2.

TABLE 2—BOND STRENGTH OF LOBLOLLY PINE HOLOCELLULOSE FIBRES

Fibre type	Average load required to release fibre, g	Average area of fibre contact,* μ^2	Bond strength, kg/mm^2	No. of fibres tested
	Fibre/cellulose film			
Springwood	4·7	149 000	0·03	30
Summerwood	11·3	111 000	0·10	17
	Fibre/shive			
Springwood (range)	8·4	30 900	0·27 (0·12–0·45)	14
Summerwood (range)	26·5	37 400	0·71 (0·34–1·33)	15

* Area is dependent upon length of the fibre end contacting the cellulose film or the width of the fibre shive and is an experimental variable.

These data show that the strength of fibre/shive bonding was about 7 times the strength of fibre/cellulose film bonding. The reason for this differ-ence, whether due to the poor bonding potential of the cellulose film, the destruction of bonds during shrinkage of the cellulose film or to the non-conformability of the cellulose film surface in decreasing contact area is not certain. Observation of the process of release of the fibres bonded to the cellulose film, however, suggests that bonding of the fibre/cellulose film surfaces takes place before the moisture content is reduced to a point that the cellulose film shrinks. This is substantiated by the abnormally large amount of stretch of the fibre—approaching 10 per cent—as it is being released from the cellulose film. Fig. 4a shows a pine springwood fibre bonded to cellulose film and Fig. 4b shows the same fibre as it is being re-leased from the cellulose film. Note the disappearance of cross striations in part of the fibre in response to the tension applied.

Studies in which the length of the portion of the fibre bonded to the cellulose film has been varied have shown that the bonding strength of the fibre surface varies along its length with the greatest bonding strength per unit area at the tip of the fibre.

Probably the most interesting result is that for both fibre/cellulose film and fibre/shive bonding, the bond strength of summerwood fibres is about three times that of springwood fibres. The reason for this difference is as yet not clear, but statistical analysis of the data shows that the difference is highly significant. It is possible that chemical and physical differences exist

between the surfaces of the two types of fibres, which could affect their bonding potential. Morphological differences such as well-developed bordered pits and cross-field pits in springwood fibres, however, could be a factor in that they decrease the actual area of contact between bonded surfaces.
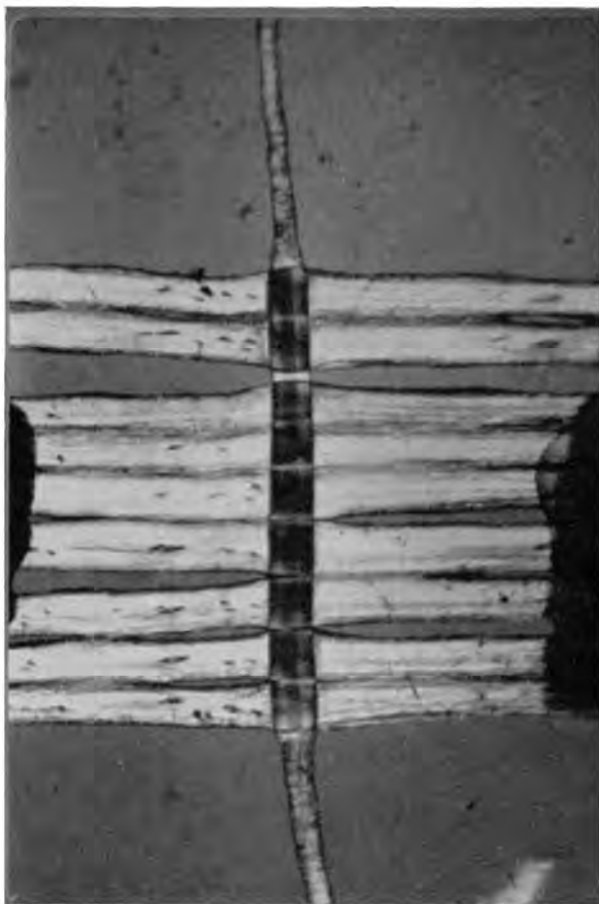

Fig. 3—Loblolly pine summerwood fibre bonded to summerwood fibre shive × 125

Tests so far conducted on the bonding strength of two single fibres arranged at 90° have given results of the same order of magnitude as those for fibre/shive bonding. The advantage of using shives is that the bonded area

(hence bond strength) is substantially greater, which makes manipulation of the bonded fibres easier.

It is of interest that, although springwood fibres are weaker in tensile strength per unit cross-sectional area and in bond strength than summerwood fibres, they will, in the unrefined state, produce sheets with much higher tensile strength than will summerwood sheets. This is due to the much larger
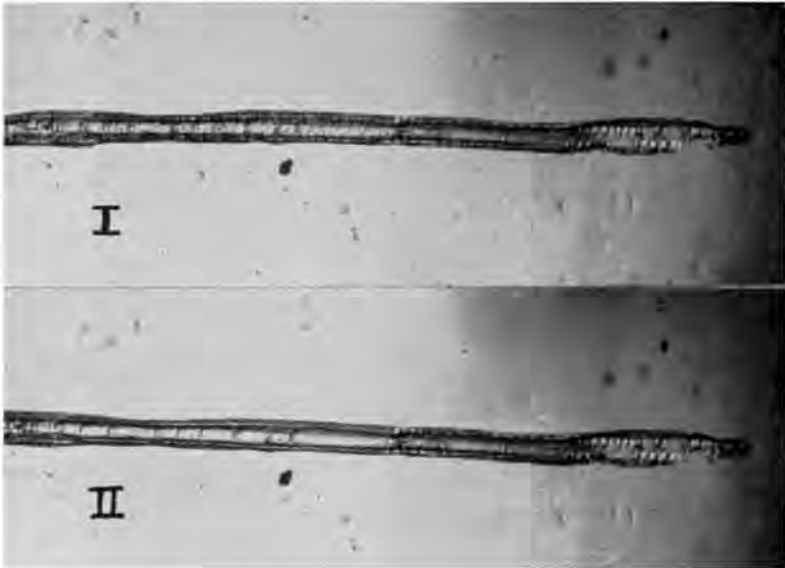

Fig. 4(*a*)— Springwood Loblolly pine holocellulose fibre bonded to
cellulose film × 50 (I)
(*b*)—Same fibre under tension with part of the fibre released from
the cellulose film × 50 (II)

number of bonded fibre crossing points in springwood sheets, which in turn is a result of greater fibre flexibility and conformability.

Investigations into the possible causes for the difference in bond strength between springwood and summerwood fibres, along with determinations on fibres from different pulps, are continuing.

REFERENCE
1. Leopold, B. and McIntosh, D. C., *Tappi*, 1961, **44** (3), 235–240

DISCUSSION

DR. H. CORTE: Stresses in the centre of the sheet are different from those in or near the surface. How far are the optical results representative of what happens inside the sheet?—or do they show merely edge effects?

MR. P. A. TYDEMAN: We think that qualitatively they are closely representative.

DR. CORTE: I can imagine that stresses in the sheet are higher. The fibres lying on top certainly have fewer bonds than those inside the sheet.

MR. D. H. PAGE: The change in optical contact area we get does correspond very well with the change in scattering coefficient that Nordman got and it indicates that there is the same area loss in the body of the sheet as there is on the surface. Furthermore, we must remember that, as we said, many of the fibres we observed are almost in the body of the sheet, in this sense, that we look at the surface and we examine only a small part of a fibre, the remainder of which may be completely covered by other fibres.

MR. J. MARDON: It would be interesting to know the rate at which you were straining. At, say, 2 in/sec, the actual breakage observed under normal light occurs at something less than 9 000 frames a second. On one frame you see nothing, on the next frame the paper will be broken.

Do you say that the work is carried out on bond breakage after tensile rupture? On rupture, surely all the bonds in the area of rupture must be broken, so we need to know the area you were investigating behind the rupture.

MR. TYDEMAN: We sampled bonds over the whole of the area of the tensile specimen as explained in the paper.

MR. MARDON: It would be interesting to know how the distribution of broken bonds varies on the specimen about the area of your rupture.

MR. TYDEMAN: We have been unable to detect any variation over the surface area of the specimen in terms of the total number of bonds on the surface. The samples are quite small.

MR. PAGE: I think we lean very heavily on Nordman's work here—and we are right to do so, because ours is a difficult technique and involves a tremendous amount of work, since we are looking each time at a single fibre-to-fibre bond. Therefore, we feel justified in using Nordman's observations on change in scattering coefficient to give a broader and more integrative picture. He showed quite conclusively that the scattering coefficient change occurs all over the sheet, not just in the region of the fracture. We must bear his very good work in mind when we are thinking of this.

DR. H. F. RANCE: On average, how many fibres thick were the sheets represented by these particular frequency distributions and does the frequency distribution vary according to the thickness of the sheet or according to how many fibres thick it is?

MR. TYDEMAN: I cannot say offhand how many fibres thick the sheets were, but they were 60 g/m² handsheets. We have not investigated whether or not there is any effect of thickness.

DR. RANCE: Would you not expect some variation quantitatively in the nature of these distributions with different thicknesses?

MR. TYDEMAN: No, not over the range of normal basis weights.

DR. J. A. VAN DEN AKKER: I wish to comment on the interesting experiments of Kallmes, McIntosh and Leopold. Mr. Truman raised a very interesting question in an earlier discussion. The matter of stress concentration involved in the discussion of his question would inevitably be involved here, but before mentioning this I would like to recall the classic work of deBruyne. He showed the importance of stress concentration in glued joints and the incorrectness of arriving at failing stress by taking the quotient of shear force and area of the joint. In experiments on fibre-to-fibre bond strength of the kind we have just heard about, one would expect severe stress concentration. Accordingly, the apparent shear strength of the bond would be much too low—possibly by a large factor.

MR. PAGE: I would like to follow on from there, if I may. I think there is even more to it than this, particularly when we start considering materials like cellulose film, because the stress concentration is then not only dependent on the fibre, but on the elasticity and thickness of the material to which it is bonded. If we change the thickness of the cellulose film, we might get different

results for the force required to break this bond, owing to the change in stress concentration. We did this experiment ourselves (I think I mentioned yesterday), using glass as a substrate—which obviously is extremely inextensible—and we have tended to get lower values of the force required to remove the fibre in shear, because of the very high stress concentration that occurs here. If we make the glass thinner or use something less rigid than glass, we could get higher values. This whole question of the shear strength of bonds is extremely complicated and needs to be looked into in rather more detail.

MR. P. E. WRIST: Van den Akker's point is well taken and must be considered when attempting to explain the differences between the fibre-cellulose film and fibre-fibre bundle strength results. The same uncertainties of geometry during tensioning that cloud the interpretation of these experiments must occur also during actual straining of fibrous webs, therefore we feel there may still be value in the results. To me, of more significance than the difference between the two methods is the consistent and highly significant difference between the bond strength per unit optical bonded area for springwood and summerwood. We had expected that a difference would occur, but had guessed that the greater conformability of the springwood fibres would have made them the stronger bond formers. This was not so.

DR. L. NORDMAN: Have any tests been carried out in which the loading has been less than that required to produce rupture and, in that case, was it observed that the frequency of occurrence of 100 per cent bond breakage increased with increased straining?

MR. TYDEMAN: We have not carried out the exact experiment described in this question, but we have done a similar test. One tensile specimen was strained in successive cycles that, in most cases, progressively increased in magnitude. The first cycle consisted of straining the specimen by an amount slightly less than that corresponding to the yield point, then releasing it and examining the behaviour of the bonds. Only one out of the fifty seven examined showed any loss of area, its degree of breakage being 23 per cent. Successive cycles were carried out on the same specimen and the bond breakage after each cycle was determined and is summarised in the table below.

Thus, even after the sixth cycle, still only 5 per cent of the bonds had completely broken. Each cycle did produce further bond breakage, sometimes by the partial breakage of previously intact bonds, sometimes by an increase of the breakage during a previous cycle. Eventually, this recycling

10 min Valley beaten spruce sulphite handsheet

Cycle No.	Total strain of specimen after each cycle, %	Number of bonds		
		Unbroken	Partially broken	Completely broken
1	0·4	56	1	0
2	0·6	54	3	0
3	0·7	29	28	0
4	0·6	25	31	1
5	1·5	17	37	3
6	2·6	8	46	3

produced more total loss in bonded area than results from a single straining to rupture; moreover, the breakage is still mostly in the form of partial loss in the area of the bonds. After the sixth cycle, only 14 per cent of the bonds had not suffered some degree of loss. It should finally be pointed out that, although these figures were the result of a single experiment, they are considered to have considerable qualitative value.

MR. MARDON: I wish to draw attention to one thing that is relevant to picking at the printing press. Without going into any details, the mathematics of peeling are entirely different from the mathematics of direct separation.

PROF. G. JAYME: On the very interesting results obtained by McIntosh and Leopold on the strength of individual bonds between early and late wood fibres, I would refer to my slides shown yesterday, especially the cross-section of sheets made from pure early and late wood pulps. There can be no doubt that the former form a denser sheet, with higher figures for tensile, burst and fold, but a lower tear value. It is difficult to reconcile this well-known fact with the data of McIntosh and Leopold. One explanation may be of course that the total bonded area is greater in early wood pulp, even if the bonds themselves should be weaker. In addition, the fibres' own strength has to be taken into consideration and the packing density of the cell wall is greater in the late wood fibres as we have proved by staining and other techniques. This should explain the higher value for tearing strength obtained with late wood pulp, together with the fact that the total bonded area in late wood pulp sheets is smaller.

MR. WRIST: There is not necessarily any difficulty of reconciliation between the facts as stated by Prof. Jayme and the results of Leopold and

McIntosh. The latter have shown that individual summerwood fibres have a greater strength per unit cross-section than springwood and that summerwood fibres form stronger bond densities with one another than springwood fibres do. They have shown, moreover, that summerwood sheets are weaker than springwood sheets (in agreement with Jayme) and we must therefore conclude that this is explained by the differences in packing density and by a lower total bonded area in the summerwood fibre sheets. The figures of bonded area reported by Leopold relate only to the bonds they formed and broke experimentally and bear no connection to the number or to the average area of bonds that would be formed within a sheet.

DR. CORTE: At what approximate stresses (as percentage of the breaking stress) did areas of optical contact begin to disappear and at what stresses did the first 'optical bonds' disappear completely?

MR. PAGE: This is a question you should ask Dr. Nordman. We are looking at only a small sample of the bonds; he has looked at the whole lot at once.

Written contributions

DR. O. J. KALLMES (*for C. Mayhood, O. J. Kallmes and M. M. Cauley*): In the film just shown, we demonstrated the quantitative shear rupture of a single fibre-fibre contact. The shear forces on four softwood pulps were about the same, about 30 000 g/cm² per unit area of *optical* contact. This finding indirectly emphasises that optical contact is somehow linearly related to bonded area, but so long as we talk in terms of shear forces per unit optical contact area, this relationship is irrelevant *for now*.

MR. A. W. O'SULLIVAN: Some work that may be of help in a discussion on the differences in behaviour between handsheets dried (*a*) under tension and (*b*) free to shrink is being carried out at the present.

Using the optical method and a modification of Ingmanson and Thode's extrapolation technique (Young's modulus instead of breaking length)* on similar handsheets dried (*a*) and (*b*), results have shown that, in the case of (*b*), increased beating (Lampén mill) was followed by increased shrinkage on drying and decreased dried fibre surface area when compared with that of (*a*). Values of percentage bonded area (percentage of the total fibre surface) at any given freeness were about the same for the two cases: but, since the

* Ingmanson, W. L. and Thode, E. F., *Tappi*, 1959, **42** (1), 74–83; 83–93

surface area of (*b*) was less than that of (*a*), it follows that fewer optical bonds were present in (*b*).