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EFFECT OF PAPER ON THE LOAD–ELONGATION BEHAVIOUR OF FIBRE–TO–FIBRE BONDS

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ABSTRACT

The deformation of paper and its fibre bonds was studied by straining thin paper structures inside the specimen chamber of a scanning electron microscope. The average bond strength values of different sheet structures were characterized by several methods. The handsheet structures were varied by refining and by addition of bond strength chemicals.

The preliminary results obtained show that the structure of the sheet has a pronounced effect on the elongation properties of paper and of its fibres. The coarseness of the fibres had a distinct effect on the loading capacity of fibres and on the simultaneous straining behaviour. The thin wall springwood fibres often became inactive only after the final rupture of the structure. The summerwood fibres tended to become inactive earlier through breakage of the fibre bonds. A blinking light phenomenon was observed during the SEM straining of some paper specimens. The light blinks were interpreted as complete breakages of fibre-to-fibre bonds.

The structural features of the studied handsheets had different effects on the bond strength values obtained by the various methods. These results seemed to indicate that refining produced a sheet structure which could be loaded in a more homogeneous manner. This was also reflected as higher values of bond strength. All bond strength methods used showed that starch increases the bond strength and that the debonding chemical decreases it.

INTRODUCTION

During recent years there has been a trend to lower the grammage of paper and to use less expensive raw materials. This trend will emphasize the need to maintain the strength of paper at a sufficiently high level, while simultaneously other properties - like optical properties - should also remain very good. This need requires a better utilization of the fibre-to-fibre bonds. The objectives of this research have been to experimentally study the load-elongation behaviour of bonds in paper.

General

The relationship between the strength of paper and the strength of fibre-to-fibre bonds was studied intensively in the 1950's and 60's. Many results still of fundamental value were published in those years. In the following those results of research are reviewed, which have been published since the beginning of the 1970's, and deal with the fibre bonding and the load-elongation behaviour of paper. For large general reviews of the deformation and physical behaviour of paper the reader is referred to Ebeling (1) and Dodson and Herdman (2). Robinson (3) has reviewed the bonding between fibres and Uesaka (4) the methods used for characterization of the fibre bonds.

Paper may be considered as an exceptional material because it comprises fibrous elements, which are fairly large in relation to the thickness of the paper. In addition, the fibres are situated heterogeneously. However, the thermodynamic behaviour of paper under deformation is by no means a very special one. On the contrary, the thermodynamic behaviour of paper under straining resembles that of aluminium foil (5). Also, under straining, various grades of paper - in spite of differences in the structure and bonding between the fibres - seem to have a similar thermodynamic response. One may thus conclude that papers made out of natural cellulosic fibres have a similar mechanism of deformation. The apparent plastic behaviour of paper during straining is not caused solely by the breakage of the fibre-to-fibre bonds,

but it is related to the deformation induced structural changes on a molecular and supra-molecular scale. Because of these structural deformations, breakages of fibre bonds may also occur. It has been hypothesized by Ebeling (5) that the irrecoverable work of extension is mainly related to the breakage of intrafibre hydrogen bonds, which are broken because of molecular stress concentration due to shear deformations between the fibrils. Such shear deformations were visualized to take place in the bonded region, i.e. at the periphery of the bonds (wrapping of fibres) and at the micro-compressed regions of the bond plus in the free segments, i.e. at the kinked or "flaw" places of the segments.

Strength of Fibre Bonds

The strength of fibres is usually at least twice the strength of paper made from them. It is because of this that for instance Davison (6) has concluded that the fibre bonds are the weak link of the paper. Individual fibres can be pulled unbroken from the sheet structure although they are bonded to other fibres with numerous bonds. The bonds are not loaded simultaneously. Instead they are loaded one or a few at a time, which also means that the bonds are broken one or few at a time. According to Davison (6), the following reasons might be behind the low strength of the fibre-to-fibre bonds : (a) the hydroxyl groups of the different cellulose chains do not have a good match with each other during the formation of the bond, (b) the uneven surface of fibre and (c) the lateral shrinkage tendency of the cell wall and the subsequent formation of stress concentrations into the bonds. (The last point may lead to partial opening of the bonds by peeling and/or by torsional peeling.)

Of the factors affecting the specific strength of fiber-to-fibre bonds the following have been recently cited:

- pulping method (8, 16, 21)
- amount of refining (9, 12, 13, 21)
- fines (13, 14)
- amount of starch added (13, 14)
- chemical modification of fibre (10)
- drying (16)

No great difference was observed by Mohlin (7) in the bonding strength of springwood and summerwood fibres bonded to cellophane. However, the opening of the bonds differed between these two fibre types. The springwood fibres had undergone axial shrinkage because of the shrinkage of cellophane. Because of this the bond did not open - and most probably - did not become loaded simultaneously but in successive partial breakages. If one sums up these load-elongation responses there seemed to be no difference between the bonding strengths of summerwood and springwood fibres bonded to cellophane. The maximum loads involved were higher for the stiffer summerwood fibres. According to Mohlin (7), the axial compressions in the fibrous cell wall were concentrated to certain positions, which were thought to be less resistant to axial compression.

Distribution of Strain Levels in Microscopic and Submicroscopic Scale

The local strain in a piece of paper undergoing extension varies. A correlation has been observed between variance in the local grammage values and the variances of the corresponding strains (22). Besides the grammage variation also the variation of fibre orientation affects the variation in local strain values (23). Various parts of the fibre have been reported to have different responses to straining. In a light microscope study Giertz and Rodland (15) observed that the fibres strained more in the bonded regions than in the free segments. The straining of the bonded region was not a continuous and homogeneous process. The observed extensions depended on the orientation of fibres in relation to the direction of loading. The extensional strain changed to a compressional one when the angle between the direction of loading and the direction of fibre exceeded 50 to 70°. Thorpe et al.(16) measured a smaller elongation in the bonded region between fibres than in the free segment of a fibre bonded to a shive. The extension was fairly linear and homogeneous up to the rupture. Based on a theoretical analysis they concluded that the highest stresses in a fibre-to-fibre bond take place at the periphery of the bond. The same conclusion was put forward by Van den Akker some 15 years earlier (27). Since load is transferred through the

structure along the periphery of the bonds, the length of the fibre-to-fibre bond should not have a pronounced effect on the rupture load of the bond.

Perkins et al.(17) have analyzed, with a microscope, the structural changes that take place during in-plane straining or compression of paper. They did not find delaminations of cell walls, slip plane occurances or dislocation developments more than what could be observed in undeformed specimens. One could see openings of the fibre bonds due to straining. An analysis of the extension behaviour in a scanning electron microscope showed (18) that most of the charging took place in the bonded regions. (The charging was visualized to originate from straining induced cleavages of the evaporated gold layer on the surface of the fibres.) A homogeneous sheet showed very little variations in local strain values.

Button (19) has also analyzed theoretically the mechanical properties of fibre-to-fibre bonds. He constructed a linear elastic model for a lap joint between cellophane strips. It was possible to predict with the model the drastic decrease in the strength of the bond when the length of the joint was larger than 0.2mm. The difference in the moduli of the strips or in their thicknesses decreased greatly the strength of the bond. Similar conclusions can be made from results concerning joints between fibre reinforced plastics (20). The highest shear stress in a lap joint is usually at the edges of the joint and thus the peel stress is also highest at the edges.

One may conclude this review by a statement that apparently the bond strength of fibre-to-fibre bonds in ordinary papers is not totally utilized during the loading of papers because the bonds are loaded successively and because the stress concentrations at the periphery of the fibre bonds accelerate the rupture of the bonds.

EXPERIMENTAL

Preliminary results will now be presented about a microscopic and macroscopic evaluation of the load-elongation and strength behaviour or ordinary paper

structures. The objectives of this investigation were to determine how the various structural features of paper affect these behaviours.

Microscopic Analysis

The straining behaviour of the paper structure was analyzed in a scanning electron microscope (Philips SEM 505 with a low voltage attachment). A special straining device was used which could be adapted inside the specimen chamber of the SEM. Simultaneously with the straining, the image output of the SEM was recorded with a video recorder. Table 1 describes the paper specimens that were tested. Appendix I describes in detail how the obtained video information was analyzed and how the samples to be strained were prepared.

Valley beating min	Corresponding degree of refining SR	Sheet structure	Drying method
--	14	isotropic	heated drum, between blotting papers
15	17	isotropic	heated drum, between blotting papers
30	24	isotropic	heated drum, between blotting papers
30	24	isotropic	room air, free shrinkage
30	24	isotropic	room air, glazed against smooth resin surface
45	39	oriented	heated drum, between blotting papers
45	39	oriented	room air, shrinkage
45	39	oriented	room air, frame attachment
75	67	isotropic	heated drum, between blotting papers

Table I. Paper samples strained in SEM (bleached pine kraft pulp, grammage = 15 g/m^2)

Macroscopic Evaluations

The mechanical behaviour of various paper structures (load-elongation) and the macroscopic strength of fibre-to-fibre bonds were measured from 60 g/m^2 laboratory handsheets. These were made from the same bleached pine kraft pulp used in the manufacture of thin handsheets for the microscopic analysis. Table 2 describes the tested paper specimens. In Appendix II a closer description is given of the procedures for the making of the handsheets.

Beating time, Valley, min		-	15	30	45	75
No additives		x	x	x	x	x
Debonding agent	0.15%			x		
	0.50%			x		
Starch	0.5%			x		
	1.5%			x		

Table 2. Paper samples for "macroscopic" analysis

The handsheets were tested at 50% RH and 23°C. The following properties were measured from the handsheets:

- density
- light scattering coefficient
- tear strength
- tensile strength and other pertinent parameters of the load-elongation curve
- relative bonded area (RBA)
- short span tensile and other pertinent parameters (28)
- Internal Scott Bond bond strength
- amount of broken fibres in the rupture line and the average length of fibre pull-out (29)
- "work" of peeling of two ply sheet (Skowronski's specific bonding strength method (13))
- shear strength of two ply handsheet (Clark's cohesiveness method (30))
- bonding strength according to the Nordman method (31)

RESULTS AND DISCUSSION

Special SEM Phenomena During Straining of Paper

Paper is not an ideal material for scanning electron microscope evaluation. This fact has many implications. Some of these implications are reviewed in Appendix III.

When an unmetalled paper specimen undergoing straining was viewed with a low acceleration voltage one could see - as the final rupture was approached, and also during the propagation of the rupture line - tiny light blinks. Some of the light spots showed clearly brighter than their surroundings, some appeared to be darker. These tiny light blinks seemed to be connected with the complete breakage of fibre-to-fibre bonds. It is believed that these complete bond breakages are similar to those reported by Corte and coworkers (25) for acoustic analysis of the prerupture straining behaviour of paper. It is hypothesized that the light blinks are related to charging of the newly created (fibrillated) surfaces. The surface charging phenomenon is believed to be related to the changes in the electrical conductivity of paper, which is affected strongly by the amount of fibre-to-fibre bonds (32,35).

The light blinking was very pronounced in those fibres that were in the middle of the rupture zone. Those fibres that stick out into the rupture zone can be visualized as being the most readily charged ones. It is also possible that those fibre segments, that are carrying a very high tensile load, can turn darker - even if covered by a thin layer of gold. This is so because the force into the segments is transferred through surface layers of the fibre segments. The partial opening of the fibre bonds together with extension induced partial rupture of the gold layer may then create charging in some segments (compare Fig.III-1 in Appendix III).

Semimicroscale SEM Results of Straining Experiments

The straining of thin paper specimens can be divided to four stages, which are:

- apparent elastic straining
- apparent plastic straining
- actual rupture
- postrupture straining

The elongation in the elastic regime is recoverable. During plastic straining the elongation is irrecoverable. At rupture, those fibres will be inactivated that carried the load in the rupture zone prior to the actual rupture. In the postrupture of straining, those fibres and fibre segments, which were slack prior to the actual rupture, will be phase activated. The described division is necessary for such paper specimens in which the stored elastic energy of straining is not high enough to cause a catastrophic rupture.

Based on the semimicroscale SEM experiments the following generalizations may be drawn for the straining phenomena:

1. During the elastic regime no clear deformations can be observed. In very thin papers the pores will enlarge. In the freely dried paper some z-directional deformations will take place.

2. The beginning of the apparent plastic regime cannot be clearly observed from the straining induced structural changes. Only at the end of this phase one starts to observe the already described complete breakages of individual bonds. These breakages will first concentrate on some area at the narrowed edges (Fig. 1).

3. The continuation of bond breakages will eventually form the origin of the actual rupture line. In Fig. 2 the bond breakages at the left hand edge have caused a stress concentration also at the right hand edge of the narrowed specimen. This stress concentration will also cause the beginning of the

rupture line. At the left hand edge one can clearly see as black and white stripes the bond breakages of such fibres, which were lined up parallel to the loading direction. Simultaneously one can see strong charges build up on those fibres situated in the rupture zone. The inactivation of those fibres and fibre segments still supporting the load advances very fast across the specimen. It is highly probable that the load supported also decreases very quickly during this stage.

4. After the first advancement of the rupture line across the specimen (Fig. 3), there are still many fibres which stretch over the rupture line and which now start to carry the load. These fibres have been so far inactive because of their large orientation angle or they have become inactive because of the bond breakages they were involved in earlier. Now these fibres will become active again. The fourth phase ends when all the material extending over the rupture line has finally become inactive. In some special cases the postrupture straining phase may last as long as the three previous phases together.

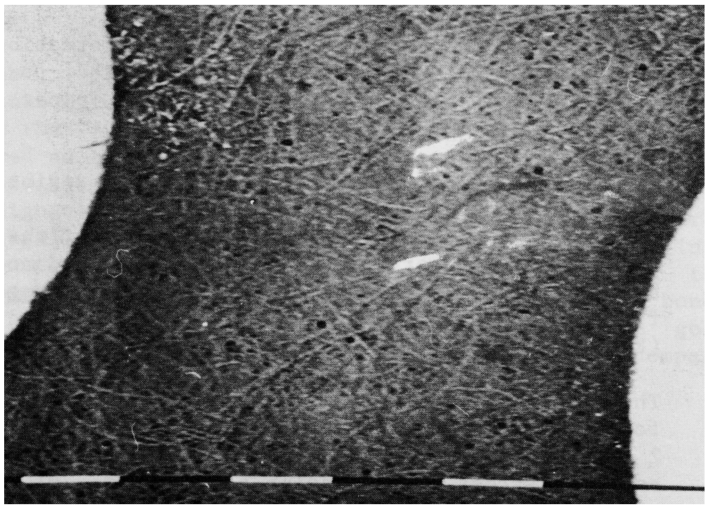


Fig 1—The rupture line is just beginning at the left hand corner of this 35 g/m² bleached kraft handsheet. (Observe the concentration of bond breakage induced light blinking in this area.) The length of the bars in the picture is 1 mm.

There were large differences between the rupture behaviour of the various specimens. In many cases the rupture line did not appear in the narrowed part of the specimen. This was due to large variations in the grammage of these thin paper samples. The freely dried specimens exhibited plenty of complete breakages of fibre-to-fibre bonds during the prerupture straining. The specimen dried against the glossy resin plate showed complete breakages of fibre-to-fibre bonds only at the beginning of the actual rupture phase. In the freely dried handsheet - with mild beating treatment (24 °SR) - the rupture line progressed stepwise across the specimen. After considerable refining of the fibres (39 °SR), the rupture line of the freely dried samples propagated in one step across the specimen.

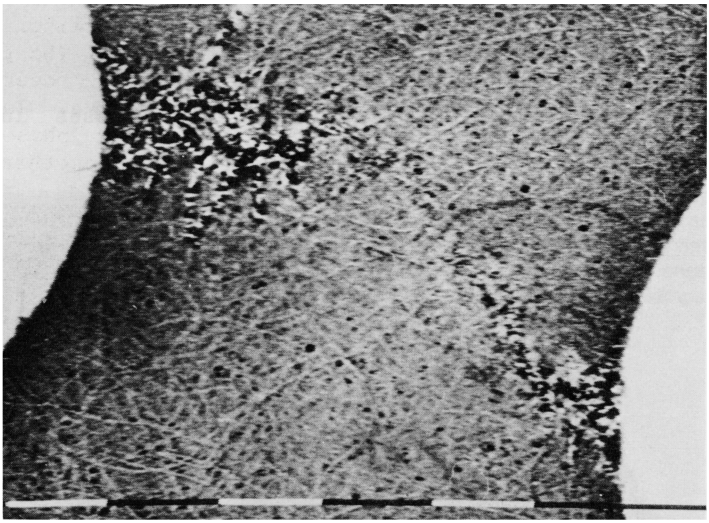


Fig 2—Also the right hand edge of the specimen exhibits a beginning of the rupture line. Bar length is 1 mm.

It is believed that the described variations in bevahiour were due to differences in stored elastic energy, which was attributed to differences (at the onset of rupture) in the number of the active load bearing elements.

Microscale SEM Results During In-Plane Tear Experiments

A 2mm long cut was made to one of the edges of the paper specimens. The straining induced deformation around the tip of the cut was analyzed inside an area of 300 by 300μ m^2. Results in Fig. 4 show that the local deformation varied considerably. A distinctive difference was observed between the local deformations in the straining direction and in the direction perpendicular to it. A typical value for the average of the straining direction elongations was 3.2% and that for deformations in the perpendicular direction -0.2%. The perpendicular deformation values seemed to react sensitively to the microscale evenness of the loading. As a function of time the deformations did not take place linearly or evenly. These variations probably reflected the redistributions of stresses in the segments and bonds.

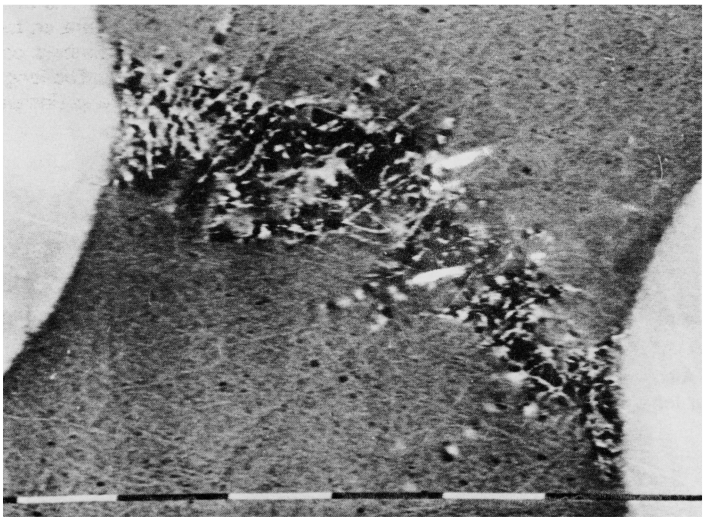


Fig 3—The inactivation of the original load carrying fibres has taken place across the specimen. The length of the bars in the picture is 1 mm.

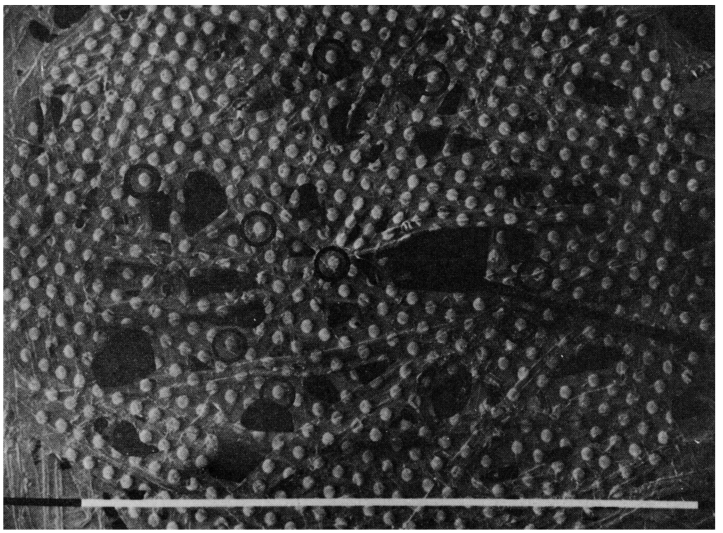
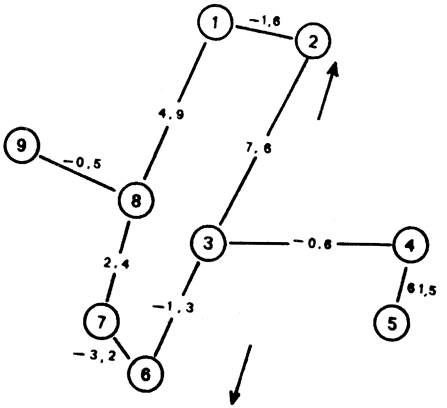


Fig 4—Example of deformations taking place during in-plane tearing. The evaporated grid pattern of gold has been used as co-ordinate system. (Shown here on the unstrained specimen. Note also the original cut for the tear line.) Numbers between the points indicate a deformation in percentage between the points. The length of white bar equals to 1 mm. The overall elongation betwen the straining clamps was 3%



The results indicated that those fibres that were oriented into the stretching direction showed a predictable high probability for tensile rupture. In the mildly refined paper structure (24 °SR), the opening of the fibre bonds was the main mechanism for the advancement of the in-plane tear line. However, springwood fibres that were completely collapsed and in the straining direction showed a tendency to tensile rupture. This might be so because of the high ratio between the bonded area (of such fibres) and the area of fibre cross section. The observed tensile rupture of such fibres was related to places, where the fibre was bonded almost completely across its width to a perpendicular fibre - and in many cases - on both sides.

The summerwood fibres were probably carrying the load right from the beginning of the straining. These fibres also seemed to become inactive due to bond breakages, in many cases before the springwood fibres and already in the early stages of the propagation of the tear line. This behaviour might be understood because of the small available bond area (less fibre collapse) and because of a smaller number of bond sites (stiffer fibres).

From the fibre and bond strength literature, one can deduce that straining of a wide (collapsed) springwood fibres may cause a tensile rupture, when that fibre is well bonded to two perpendicular fibres and that these "two bonds" are strained in opposite directions. A threefold bonded area is needed for the springwood fibre for the tensile rupture to take place. Unfortunately, it seems that summerwood fibres, which need a larger bonded area (in order to utilize their strength best), form smaller and thus weaker bonds.

In the moderatively refined paper structure (39 °SR) one could also observe breakages of summerwood fibres. Beating, which increased the bonded area of summerwood fibres, also decreased their extensional stiffness. Thus beating contributed to a more homogeneous sheet structure.

The straining of paper samples - the surfaces of which were covered with a thin layer of sputtered gold - induced cracks into the gold layer. The freely dried specimens exhibited the greatest amount of such cracking. Specimens dried without shrinkage showed very little of such cracking. Only after a considerable amount of straining had taken place could some cracks be observed.

Based on the detailed analysis of such images it was concluded that the cracking of the sputtered gold layer was related to the local telescopic extensions. It was hypothesized that cracking would appear in such places of the fibrous cell wall where there were disturbances in the orientation of the microfibrils of the S_2 layer. Such disturbance areas would be frequent in the freely dried paper structure especially in the areas of fibre-to-fibre bonds.

Fig. 5 shows a fibre-to-fibre bond, which exhibits cracking after straining (Fig. 6). By far the largest deformations have taken place at the bond area (Fig. 6). The straightening of the fibre wrap at the periphery of the bond (springwood fibre around summerwood fibre) was responsible for the major part of the observed extension. Simultaneously part of the bonded area has been opened up. At a later stage, when the bond was completely broken due to straining, the stresses in the fibre were relieved to a large extent and 20 to 30% of the longitudinal elongation was recovered.

Figs. 7 and 8 exhibit the other typical type of fibre bond. There is perhaps less fibre wrap in this type of bond (collapsed springwood fibres). In the depicted area the largest local deformation took place as an elongation of the segment between the bond sites and as a partial opening of the bond periphery due to straightening of the wrapped fibre segment. The actual bond itself, however, exhibited only an elongation of about 2%. Inside the bonded area one could not see any opening of the bonded structure. The transversely oriented fibre exhibited also some straining into the direction of loading, i.e. in transverse direction of cell wall. This deformation is visible from the small longitudinal cracks in the gold spots.

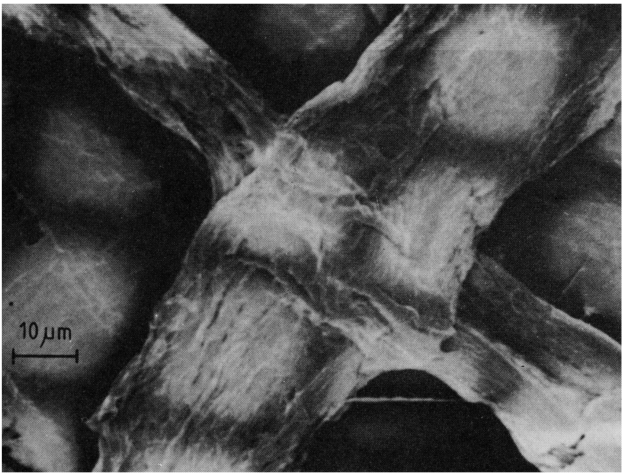


Fig 5—Example of a fibre bond (with considerable wrap) in a freely dried paper structure before straining (39°SR)

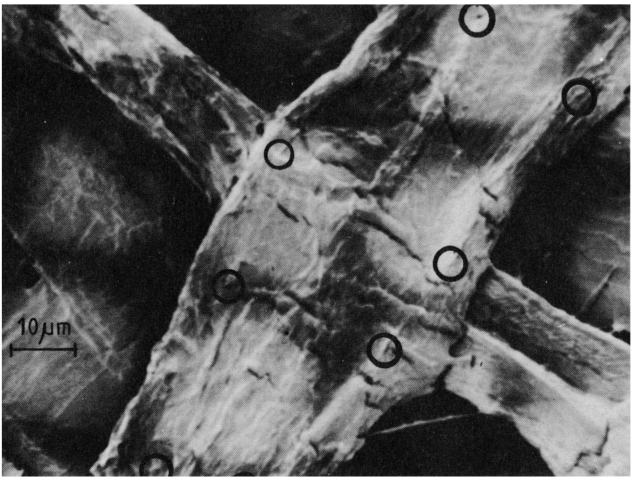
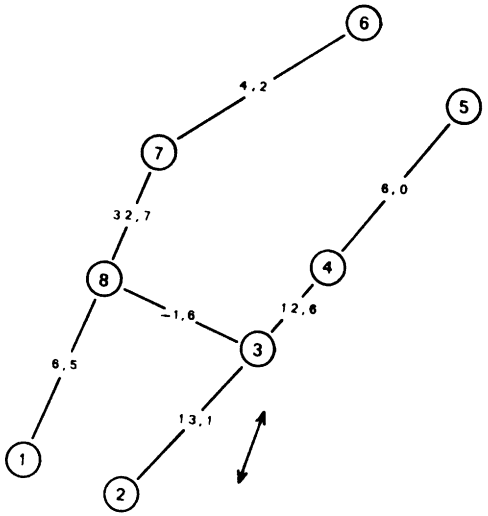


Fig 6—Same area (under load) after extensive straining. Observe the cracks formed into the sputtered gold layer and the attached local deformations (%)



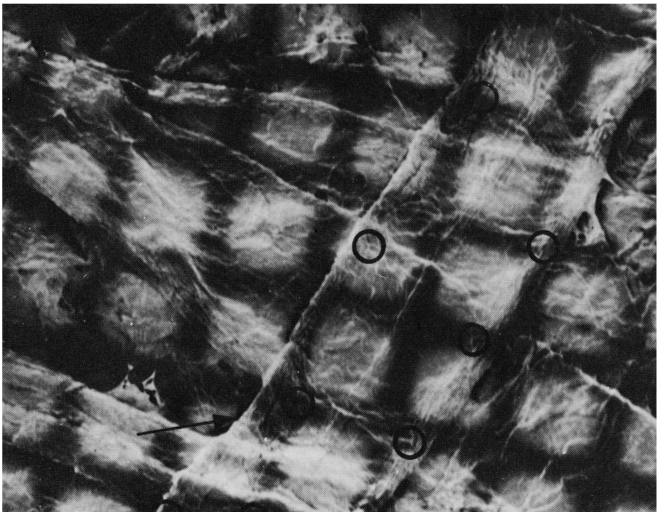


Fig 7—A loose segment in a freely dried sheet structure before straining (39 SR). Arrow points to a bent segment.

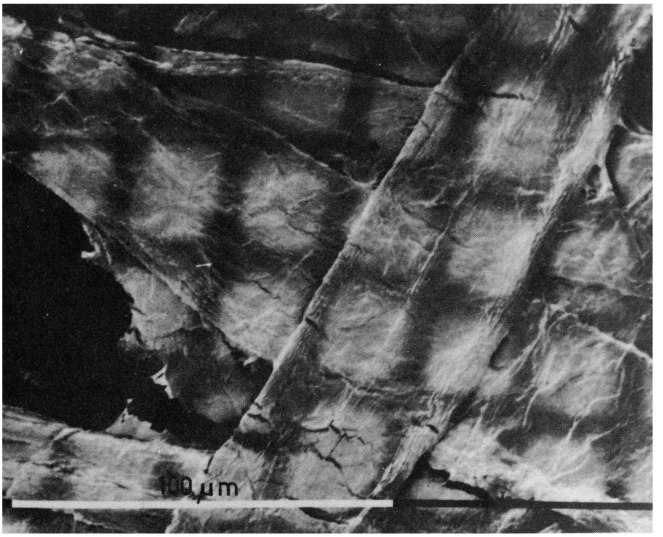
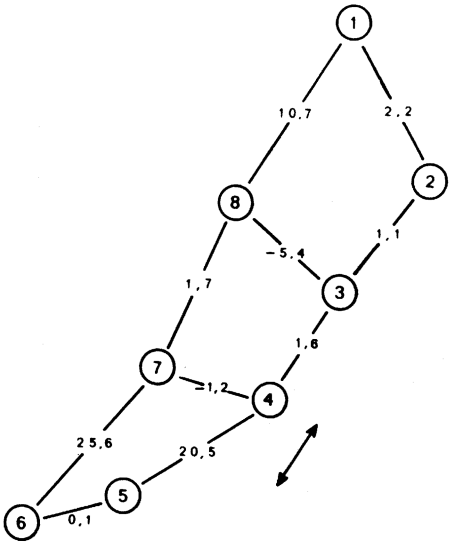


Fig 8—The same area (under load) after extensive straining. Observe the cracks formed into the gold layer and the attached local deformations.



Macroscopic Analysis of Bond Strength

The bond strength values obtained by the various macroscopic methods from the tested handsheet specimens are presented in Table 3. The tensile strength values of the tested specimens are given in Fig. 9.

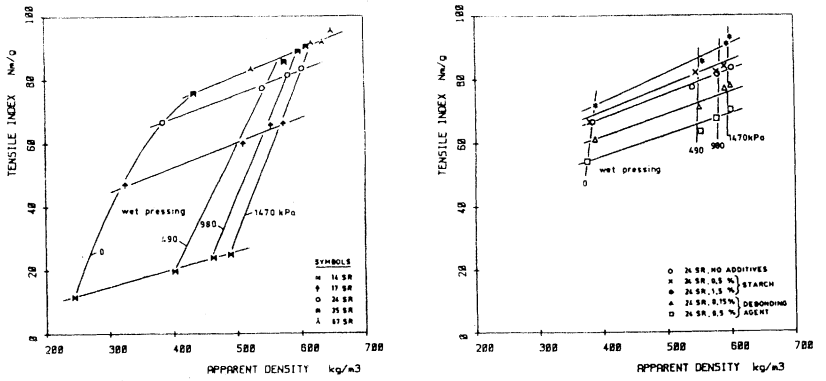


Fig 9—The effect of beating, wet pressing and dry strength chemicals on the tensile strength of bleached pine kraft handsheet.

The effect of debonding chemicals and starch can be noticed clearly. Also the other parameters describing the mechanical behaviour of paper structure - like the primary and secondary extension modulus, yield force and tensile energy absorption - indicated a similar response. The various values obtained for the bond strengths were normalized by dividing the appropriate force or energy with the related bond area parameter. The normalizing factor was usually based on the relative bonded area derived from light scattering results. The normalizing of the shear strength of the two-ply lap joint formed an exception; it was normalized with the density of the sheet.

	SR	Fiber length mm	Density kg/m3	Tear index mNm2/g	Scatt. coeff. m2/g	Zero Span Nm/g	Tensile index Nm/g	Prim. modul. MNm/kg	Rupture modul. MNm/kg	Yield stgth. Nm/g	TEA J/kg
1. No additives	14	1.95	482	12.7	37.3	128	19.8	3.88	0.20	9.0	314
2. No additives	17	1.95	509	13.9	29.2	133	59.9	5.11	0.93	18.8	1629
3. No additives	24	1.85	540	11.3	25.8	139	77.4	5.82	1.32	23.5	2088
4. No additives	35	1.76	575	9.8	23.2	150	85.6	6.20	1.53	27.3	2246
5. No additives	67	1.67	617	8.8	19.7	146	91.8	7.57	1.62	30.9	2313
6. 0.50% starch	24	1.85	546	10.2	25.1	143	82.0	6.28	1.38	24.5	2275
7. 1.50% starch	24	1.85	557	9.6	25.5	144	85.6	5.77	1.48	27.2	2301
8. 0.15% debonder	24	1.85	553	11.9	26.6	148	71.3	5.74	1.23	25.9	1512
9. 0.50% debonder	24	1.85	551	12.9	26.1	144	63.6	5.57	1.02	24.9	1479

	SR	RBA %	Bonding index * %	Nordman BS J/m2	Cohesiveness** kNm/kg	Specific BS*** J/m2	Scott Bond J/m2	BS from Page**** MN/m2	Length pulled out %	Broken fibres %
1. No additives	14	22.8	32.5	315	1.09	9.2	52.3	2.69	44.9	0.8
2. No additives	17	41.3	67.8	435	1.80	8.0	154.5	7.54	37.3	9.0
3. No additives	24	52.4	78.2	459	2.40	8.5	280.6	10.69	38.4	11.0
4. No additives	35	57.8	80.0	449	2.68	8.4	368.0	11.68	34.6	21.4
5. No additives	67	67.1	84.6	422	2.87	8.4	508.6	13.94	30.7	26.5
6. 0.50% starch	24	52.8	90.1	593	2.49	11.0	351.6	11.82	29.0	26.6
7. 1.50% starch	24	50.2	98.3	734	3.17	12.2	527.5	13.73	26.3	34.9
8. 0.15% debonder	24	49.0	69.3	441	2.20	7.5	216.9	8.57	32.6	14.8
9. 0.50% debonder	24	47.2	67.4	338	2.19	7.3	122.6	7.24	36.8	6.8

* from Cowan & Cowdrey (28)
** from Clark (30)
*** from Skowronski (13)
**** from Page's equation (33)

Table 3. Two sets of results of macroscopic tests for the studied handsheets (bleached pine kraft, drum dried, 60 g/m^2).

The bond strength values are shown in figs. 10-14. The obtained results can be divided into three groups of beating response. The various bond strength methods are situated in the three groups as follows:

1st group:

- peeling "work" of two-ply handsheet
- bond strength estimated from the pull-out length of fibres in the rupture zone of the tensile test.

2nd group:

- bond strength according to the Nordman method (NBS)
- relative bonding index of the short span tensile test (BI/RBA)

3rd group:

- tensile strength at constant density level
- relative Scott-Bond strength, i.e.Scott Bond value/RBA
- shear strength of the lap joint of a two ply handsheet
- bond strength calculated from Page's equation for tensile strength (33)

The results in group 1 were characterized by a decreasing bond strength as beating increased. For the results in group 2 it was found that there was a small maximum in the strength as beating progressed. The results of group 3 indicated that the bond strength values increased continuously as beating was advanced.

The rupture zone of a tensile test specimen can be used to estimate the bond strength if the specimen contains a small amount of dyed fibres. The average pull-out length of the fibres gives an estimate for the bond strength assuming that the combined strength of all bonds broken during the pull-out equals the average tensile strength of the fibres. In this study the dyed fibres were always unrefined fractionated long fibres. The bond strengths obtained with this method are shown in Fig. 10a. The measurement of the pull-out length of fibres was not very accurate and therefore there was scatter in the results.

However, it should be kept in mind that this method perhaps is the one that measures the actual bond strength in a relatively straightforward manner. The effect of refining seems to be to decrease the bond strength of this method.

The method developed by Skowronski (work of peeling) gave bond strength values, which did not seem to react very much to refining although there was a tendency to smaller values as refining was increased (Fig. 10b). The addition of starch or of debonding agent had a distinct effect on the bond strength. However, the stress geometry of this method is fairly complicated. The results obtained are affected - in addition to the actual bond strength - also by the bending properties of the sample. If one would account for the effect of bending stiffness, the points corresponding to the lowest levels of refining would be moved up. Besides, it has been observed in the case of adhesives research that the peeling method is a questionable one (26).

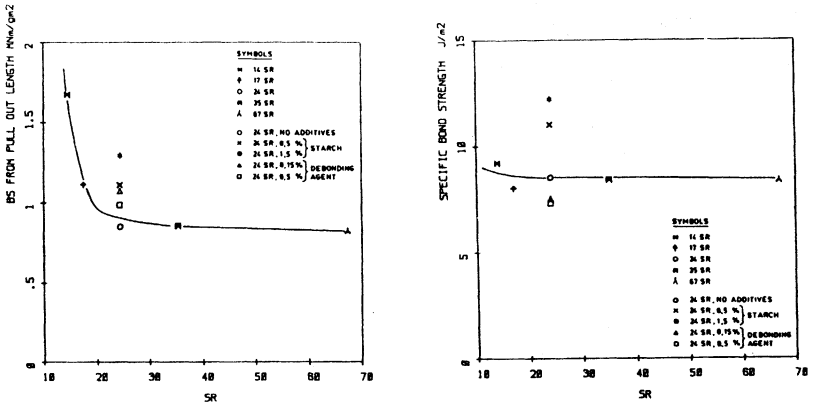


Fig 10—(a) Bond strength based on pull-out of fibres in the tensile rupture zone and (b) bond strength based on splitting of a two-ply sheet structure for the tested handsheets.

The bonding strength according to Nordman's method contained a small maximum (Fig. 11a) as a function of refining. This response was similar - as a function of refining - to the ratio of the primary and secondary slopes of the load-elongation curve and to the ratio of the tensile energy absorption and the primary elastic modulus Fig. 12). If we assume that the primary elastic modulus is related to the number of the active load-bearing material, then the Nordman's bonding strength value is related to the yielding of these load-bearing fibres. If it could be assumed that the described yielding of the active fibres depends on those fibril disturbances - that the S_2 layers of these fibres contain - then one could relate the number and severity of such disturbances to changes in NBS-values upon refining.

Also the relative bonding index of the short span tensile test gave a response to beating, where there was a small maximum (Fig. 11b). However, this result was obtained through division with two uncertain numbers because of the uncertainty involved in the basic measurements. The nominator represented the normalized strength value of the bonds and the denominator the surface area of the bonds. The obtained trend seemed to be, however, that prolonged refining decreased the bond strength values obtained with this method.

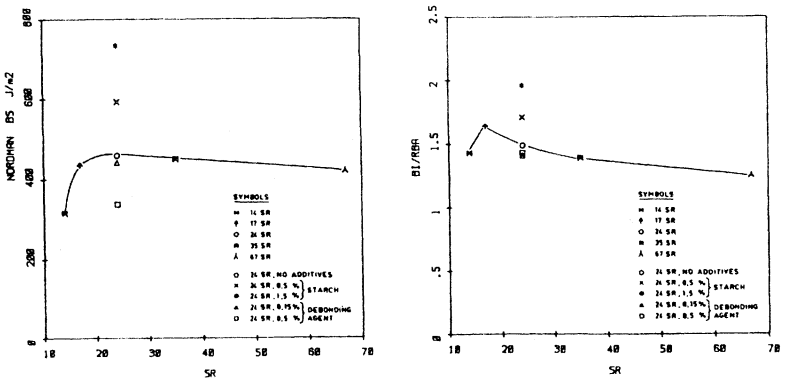


Fig 11—(a) Norman's bonding strength and (b) the relative short span bonding index (BI/RBA) for the tested handsheets.

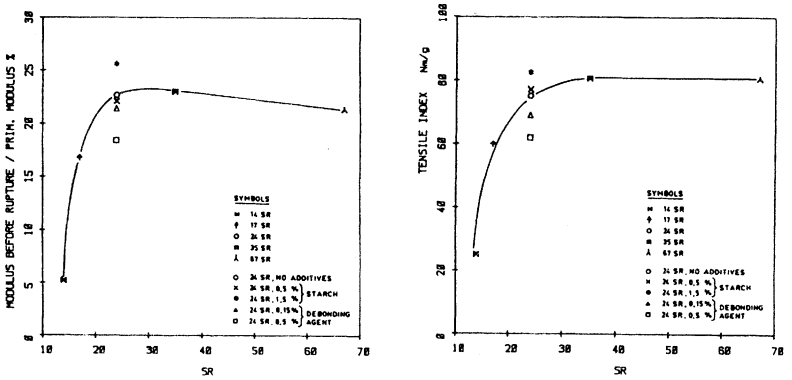


Fig 12—(a) The ratio of the elastic modulus before rupture to the primary elastic modulus of the tested handsheets (b) tensile strength at constant density level (500 kg/m^3).

The beating responses of the bond strength methods belonging to group 3 were quite similar to the response of the primary elastic modulus to refining (Figs. 13 and 14). Based on this observation one could speculate that the difference in the refining responses of the bond strengths of group 3 vs. bond strengths of group 2 was due to the fact that in group 3 the bond strength is an average value for all the fibrous material or at least for a large number of fibrous elements, unlike group 2 methods. This interpretation would imply that refining increases the amount of active fibrous material in binding the kink bands, dislocated zones and curl induced inactive segments ot the more active fibrous material.

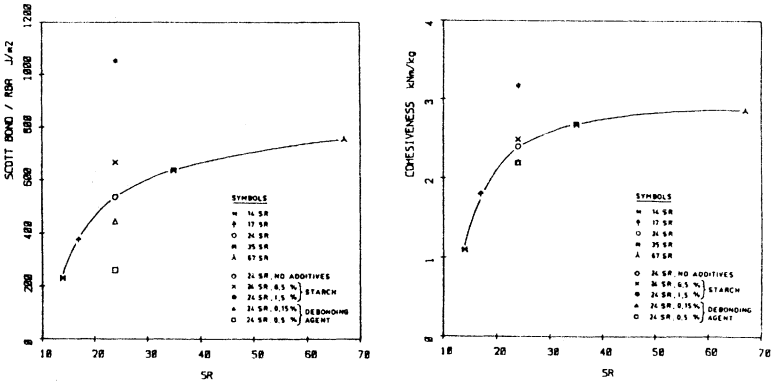


Fig 13—(a) Relative Scott Bond strength and (b) shear strength of two-ply lap joint for the tested handsheets.

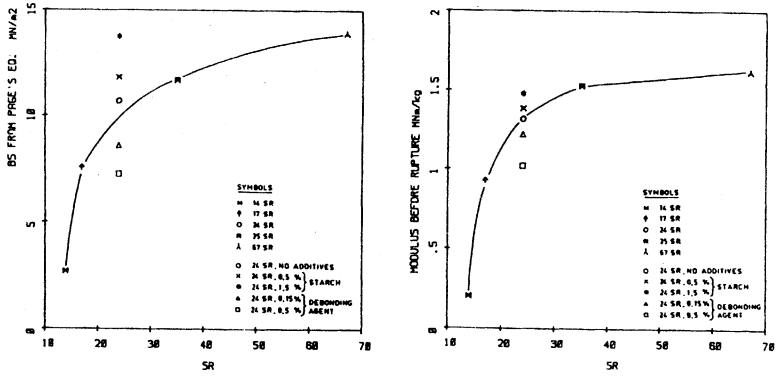


Fig 14—(a) Bond strength calculated from Page's equation and (b) the "final" apparent modulus of just before rupture straining response for the tested handsheets.

Implications of Macroscopic Bond Strength Results

The macroscopic bond strengths presented above were somewhat contradictory for the characterization of the effect of refining on the true bond strength. It has generally been accepted that refining increases the strength of interfibre bonding. The results of this study show that only some of the methods recommended for bond strength measurement comply with the accepted trend. Contradictory results have also been published earlier. Mayhood et al. (34) have reported that the bond strength between individual fibres changes very little due to beating. Mohlin (7), on the other hand, has observed a small decrease in the bond strength of kraft pulp fibres due to refining. If one accepts the results of Mayhood and coworkers as well as those of Mohlin one could then conclude that the NBS values must depend on the RBA of the sheet. This explains also the small maximum observed in the NBS-method.

One should keep in mind that the NBS value is a ratio between the irreversible straining energy and the created new surface area due to bond breakages. Therefore it is possible to understand that the irreversible straining energy absorption cannot get very high, when the fibres have not had a chance to bond strongly to each other. This is so because weakly bonded fibres cannot support such high loads, which are needed to get large scale deformations inside the cell walls of the fibres. This reasoning explains the small increase in NBS at the beginning of beating.

When the bonded area between the fibres is very small as it is in a sheet made from unrefined fibres, it is highly probable that the other factors involved will bury the effect of bond strength. Besides, there is no easy and accurate method for measuring the bonded area. A similar effect is caused by the uncertainty in the measurement of the pull-out length of fibres in the tensile rupture zone. When the pull-out length approached 50%, the obtained result will become biased towards weak fibres because, no matter how strong the fibres may be, their average length of pull-out will not become larger than 50%.

CONCLUSIONS

Considerable local and time distributed variations could be observed in the microscale deformations of thin paper structures. The springwood and summerwood fibres seemed to behave differently during the straining. The thick wall springwood fibres differed in extensional behaviour from the rest of the fibre population. Indications were observed - based on the preliminary studies - that the deformation of paper was related to discontinuous deformations in the cell wall of fibres. These deformations are believed to be related to disturbances in the microfibril orientation of the S_2 layer. In addition to this mechanism, it was observed in freely dried paper structures that straightening of the wrapped springwood fibres and of simultaneous peeling of the bond periphery and stretching of the loose segments contributed to the straining. The elongation of the bonded segments was observed to be considerably less than the extension of the free segments for bonds involving springwood fibres.

All the macroscopic bond strength measurements yielded average values, which reflected the heterogeneities of the paper. Besides, many methods gave results, which were not in agreement with generally accepted concepts of refining. Such measurements can never give values of pure bond strength, i.e. describe only the average adhesion between the bonded surfaces. The breakage of individual fibre bonds is affected by the load distribution in the sheet structure, i.e. how the neighbouring bonds, the neighbouring fibres and the whole sheet are loaded. Therefore, there exists no unambiguous concept for macroscopic evaluation of the bond strength. It can be concluded that the bond strength is a property related to the structure of the bond. The various existing methods for measuring the macroscopic bond strength seem to differ in how the effect of paper structure is affecting the obtained value for the bond strength.

ACKNOWLEDGEMENTS

One of us (ER) would like to thank the Foundation for Research of Natural Resources in Finland for sponsoring part of this research.

APPENDIX I - Preparation of paper specimens for SEM straining and analysis of obtained video information.

The paper specimens were tested both with a cool sputtered gold coating and without coating. The acceleration voltage of the electron microscope was varied between 0.9 and 3.5 kV.

The straining speed was 1%/min. The specimens were 15mm wide and their span length was 15mm. Two types of specimen geometries were used. For analysis of deformations in individual fibres the specimen contained a 2mm long razor blade cut from the edge (Fig. I-1a). For analysis of slightly larger scale of deformation a type of specimen was used in which the specimen was narrowed to 5mm width by semicircle cuts (Fig. I-1b).

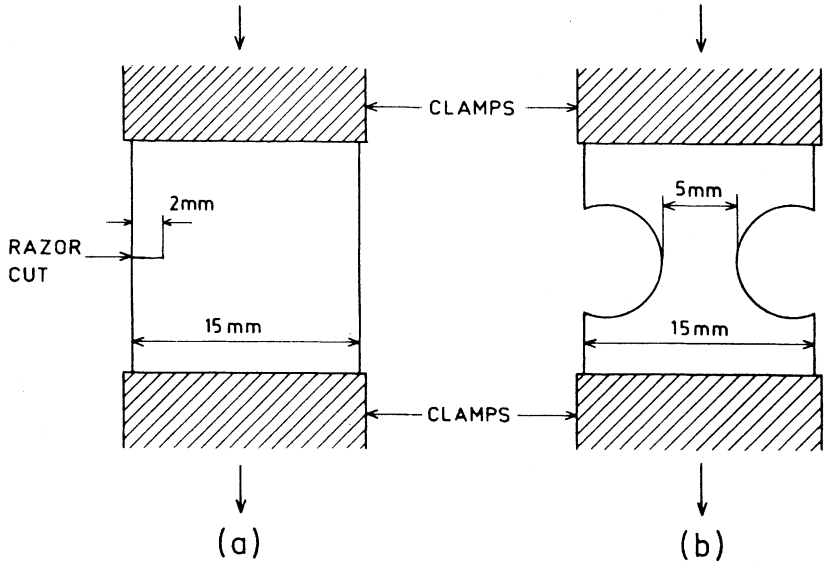


Fig I - 1—Specimen geometry for (a) analysis of fibre scale deformations and (b) larger area deformations.

In order to be able to measure accurately the local deformations several methods were tested. The best methods seemed to be to utilize structural details of the fibre surfaces or to evaporate a regular grid pattern of gold on the fibre surfaces.

Because of the dynamic behaviour of the straining it was deemed necessary to utilize TV-rate monitoring in SEM. This will create noise into the SEM images especially with uncoated specimens and with a very low acceleration voltage. Because of the noise problem in many cases, the video recorded SEM output was treated with a noise reduction image processing unit (Microconsultants, Crystal Image Processing Unit). This way the video recorded straining event could be developed to a set of extremely high quality (low noise) photographs. In some cases normal SEM photographs were also taken by stopping the straining for the picture taking.

It should be observed that the straining of paper specimens inside the specimen chamber of the SEM implied that there was no moisture present in the specimens. It is well known that the moisture content of paper has a considerable effect on its load-elongation properties. As the moisture content is decreased, the tensile strength, the strength of the fibre bonds and the extension modulus increase, while the amount of apparent plastic elongation and elongation to rupture decrease. Experimental results have, however, shown that the mechanism of elongation deformation is the same for straining of dry paper and of paper containing some moisture (1). Thus it is believed that the results obtained in this investigation have a bearing also to the behaviour of fibre bonds when normal paper is being strained.

APPENDIX II - Preparation of paper specimens for "macroscopic analysis of mechanical properties and of bond strength"

The ordinary handsheets were wet pressed for 4 min at 490 kPa (5 kp/cm^2) and they were then dried on a rotating drum between blotting papers. The purpose of adding slightly cationic starch (Finnamyl/Cat 015) was to enhance

the bonding strength. The purpose of adding the softener (Quaker 2003 BS) to the stock was to decrease the bond strength between fibres. In order to measure the relative bonded area (RBA) of these papers, handsheets were also made with and without 980 and 1470 kPa (10 and 15 kp/cm^2).

APPENDIX III - Background for scanning electron microscope studies

The image in the scanning electron microscope is based on the emission of electrons from the surface of the specimen to the detector. The emission is caused by the incoming electron flux. Because the electrical conductivity of the paper is low, the surface of paper tends to collect electrical charges, which disturbs the image formation. There are several ways to alleviate these problems, e.g. one may increase the electrical conductivity by evaporating (sputtering) a thin layer of conductive material on the fibre surfaces, or one may expose the paper specimen with a very low acceleration voltage so that an equilibrium is reached between the absorbed and emitted electrons. Both of these methods can be used to study the deformation of paper.

When one elongates a gold coated paper structure, new surfaces are revealed because of extension and movement of fibre segments and because of breakage of the bonded area between fibres. These local elongations will be to high for the gold to yield without rupture. These new surfaces will show up either as light blinks (shiny or dark spots) or as a darkened area (Fig. III-1). The first case is emphasized by the very low electrical conductivity of the new surfaces. The second case is probable when the acceleration voltage is very low. The darkening of the newly formed surface is due to general atomic number induced grey scale difference.

Interaction between the Electron Beam and Paper

During straining the paper specimen was exposed to the electron beam for no more than 10 min. The beam current was less than 10^{-9}A and acceleration voltage less than 3.5kV.

This amount of radiation is not likely to cause damage to a significant degree. In a study of Grauer et al. (36) much more energetic radiation (100kV, 5mA) was used. It had only a small effect on the tensile strength of paper. Smaller currents seemed to increase the tensile strength of paper made of softwood kraft pulp.

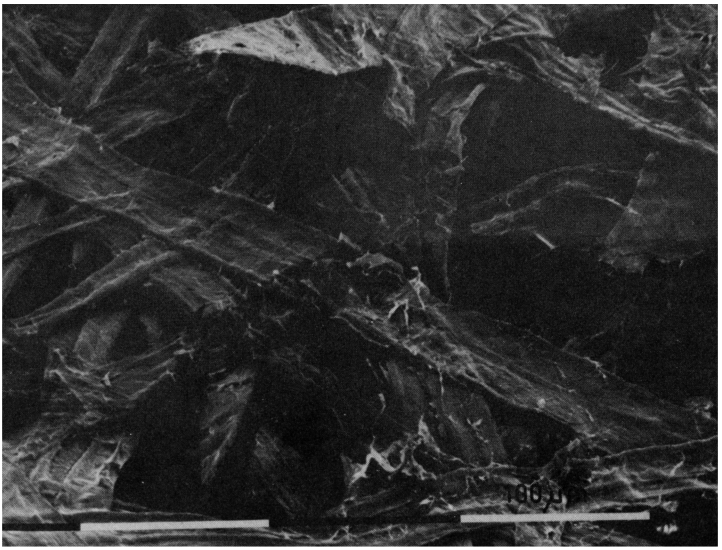


Fig III - 1—Deformed fibre structure in the neighbourhood of the rupture zone. The new surface area due to breakage of bonds will show up darker.

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Transcription of Discussion

Effect of Paper Structure on the Load-Elongation Behaviour of Fibre-to-Fibre Bonds

by E. Retulainen and K. Ebeling

Prof. J. Silvy Ecole Francaise Papeteries, Cedex, France

Do you not think we should place great emphasis on the effects caused by restraint during drying of the sheet in the interpretation of results where fibre orientation is involved?

Prof. K. Ebeling If you refer to the printed text you would see that is what we have done. We have studied sheets which have been dried with and without restraint. What we observed was that the more we allowed the shrinkage to take place during drying, the more we observed this "cracking phenomena" of the sputtered gold layer.

Page May I congratulate you on this very interesting piece of work and your experimental technique. There are two problems which I can see in the interpretation of this data. Firstly, in the scanning electron microscope the sample is being held under vacuum at zero moisture content which could affect the stress-strain behaviour of paper. You could overcome this by straining outside the chamber first, so that the paper is responding to straining under normal moisture conditions. The second problem is that of the beam damage to the cellulose, which you cannot really overcome in the SEM. Do you have any comments to make on whether these two factors influence the results you have presented?

Ebeling That is a very important question. We believe that both the elastic and plastic regions of straining do exist in the SEM but the elongation behaviour is reduced due to the very low moisture content of the sample. As regards beam damage we are in the fortunate position of being able to use very low acceleration voltages, i.e. below 1 kV. We were able to see that during the duration of the experiment, about 3 to 5 minutes, no visible damage was evident, (see Appendix III, p. 259 in Volume 1).