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THE ROLE OF FIBRE COLLAPSE IN PAPERMAKING

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THE role of fibre collapse in determining the extent of bonding and the density and strength of paper has been discussed in several of the papers presented at this symposium.[1–5] The effect appears to be twofold: firstly, fibre flattening permits larger contact areas between fibres and, secondly, fibre collapse results in a greater flexibility of the fibres.

Fig. 1—Cross-sections of wet hollow filament rayon fibres: the fibres were mounted for sectioning by embedding an oriented wet web in methyl methacrylate containing a water soluble plasticiser (methyl phthalyl ethyl glycollate) and carrying out a polymerisation according to Moore *et al.*[6] (× 245)

Both fibre collapse and fibrillation affect the bonding and strength of paper. The two effects may be demonstrated separately by the use of model cellulose fibres—hollow filament rayon and fibrillating rayon—both of which produce sheets of reasonable strength.

Fig. 2—Electron micrograph of the surface of a sheet formed from hollow filament rayon fibres (× 146)

The hollow filament fibres when wet are more or less tubular in form (Fig. 1), but are almost completely collapsed when formed into a sheet and dried (Fig. 2). The sheet is strong (4 100 m breaking length) despite the absence of fibrillation and hemicelluloses. The strength derives from the large areas of intimate fibre contact that develop during water removal.

A portion of a fibrillated rayon sheet is shown in Fig. 3. The strength of a standard handsheet formed from fibrillating rayon fibres beaten in a homogeniser for 30 min is 2 200 m. The strength here is derived from the bonding of fibrils and lamellae on contiguous fibres. The fibres themselves are solid and uncollapsed.

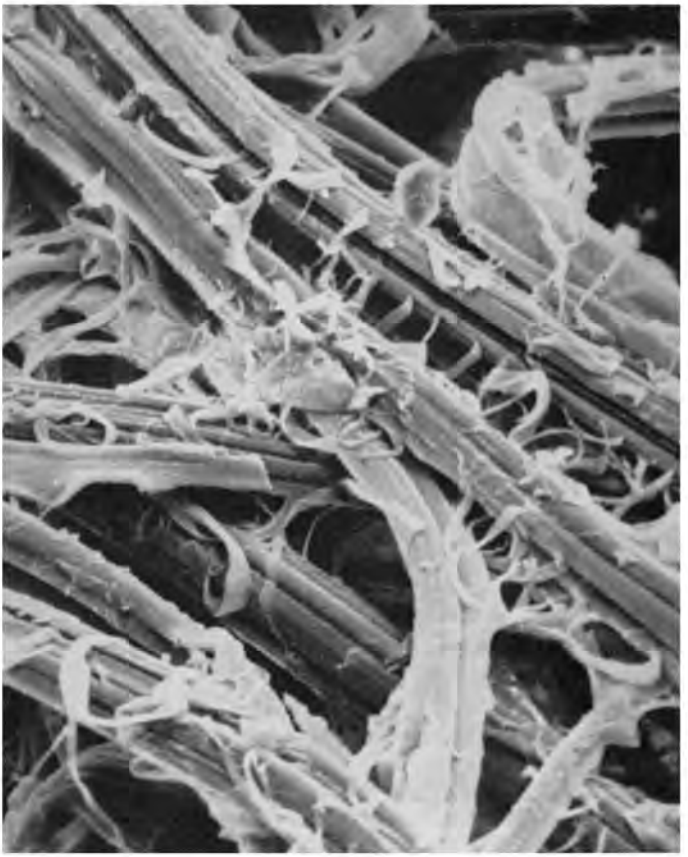

Fig. 3—Electron micrograph of the surface of a sheet formed from fibrillated rayon (× 1 260)

Electron micrographs of a sheet formed from a beaten sulphate pulp show that both fibre collapse and fibrillation may play a part in strength development (Fig. 4).

The collapsibility of fibres is logically a function of their cross-sectional

geometry. The ratio of lumen to wall diameter has been used to assess the relative papermaking potential of various hardwood species,[4, 7] to differentiate the behaviour of springwood, summerwood and reaction wood[2] and to explain the papermaking properties of cotton linters and esparto.[5] In

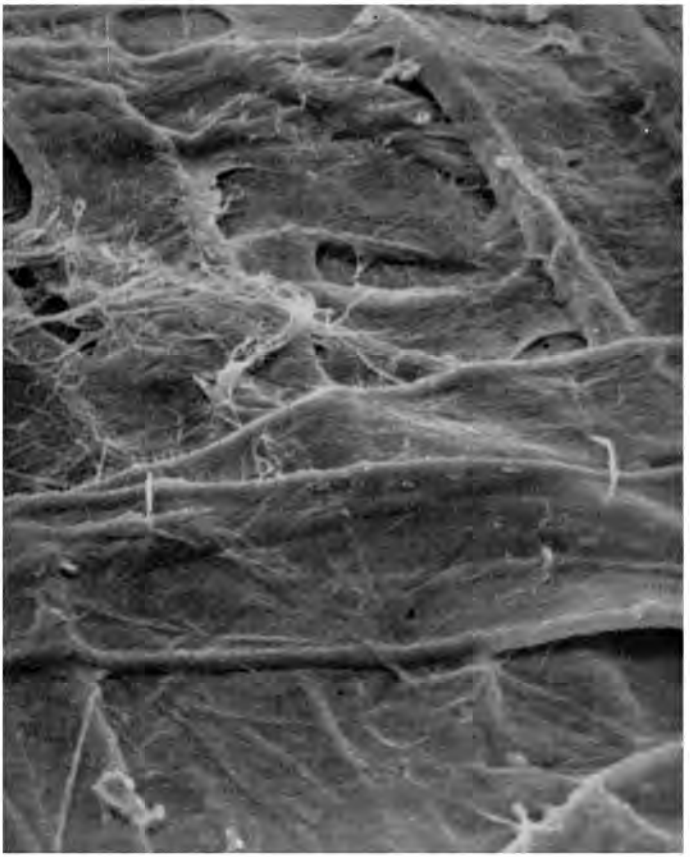

Fig. 4—Electron micrograph of the surface of a sheet formed from a beaten spruce sulphate pulp (× 715)

addition, one would expect that collapsibility would be a function of the degree of delignification, the method of cooking and the extent of mechanical treatment.

The effect of delignification on collapse may be shown pictorially by comparing the cross-sectional aspect of two wet spruce pulps of different

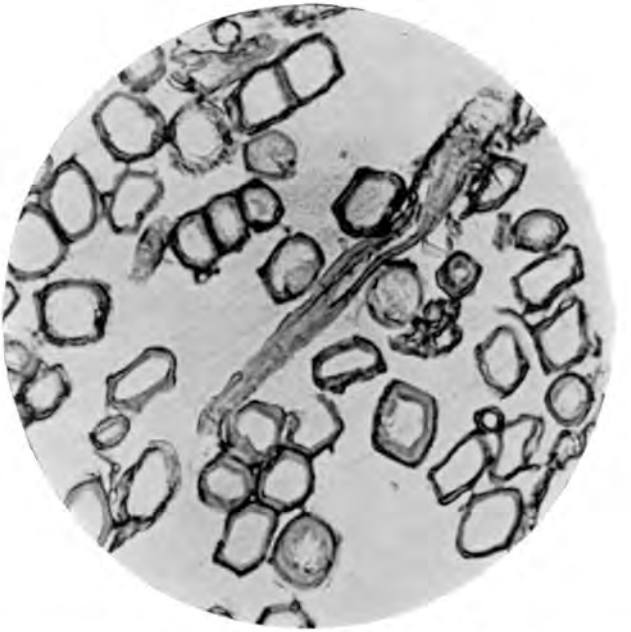

Fig. 5—Cross-sections of wet spruce sulphite fibres at a 70 per cent yield (×245)

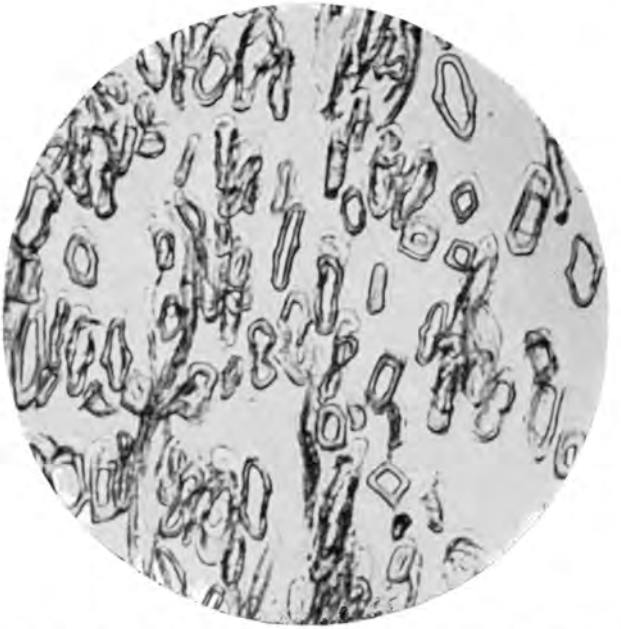

Fig. 6—Cross-sections of wet spruce sulphate fibres at 50 per cent yield (×245)

Fig. 7—Cross-sections of high yield fibres (Fig. 5), after forming a sheet and drying (× 245)

Fig. 8—Cross-sections of low yield fibres (Fig. 6) after forming a sheet and drying (× 245)

yields. Collapse is almost absent in the high-yield pulp (Fig. 5) and is only partial in the low-yield fibres (Fig. 6). In dry sheets made from the same pulps, the differences in degree of collapse are more apparent (Fig. 7 and 8). Electron micrographs of paper made from fibres of varying delignification have shown similar differences in collapse and have been presented in a previous discussion (Buchanan, p. 101).

In connection with an investigation of the relation of fibre properties to wet web properties,[8] we have become interested in the subject of fibre collapse—its extent, the factors affecting collapsibility and determination of the point in the papermaking process at which collapse occurs. This work is in progress.

Some evidence relating to fibre collapse may be inferred from a measurement of critical moisture contents by two separate methods. Firstly, the water retention of a pad of fibres is measured by application[8] of a technique due to Barkas,[9] in which a wet pad or web is placed on a sintered glass plate and subjected to a standard hydrostatic tension. The equilibrium water retention of the fibres is determined. Secondly, a method due to Preston[10] is applied to wet webs[8] to determine the moisture content in a drying web at which liquid water can just no longer migrate from fibre to fibre as indicated by dye migration.

The critical moisture content measured by the first technique is never less than that measured by the second and the difference is interpreted as a measure of the ability of the fibre to collapse. The hydrostatic tension method measures the volume of water occluded in the lumen, water involved in wall swelling plus an envelope of water surrounding the fibres, which is a function of the surface conformation. The dye migration technique measures the water held by the fibres when the fibre surfaces are 'dry'. A principal component in the difference between the two results is whether, at the dye migration end point, the fibre is collapsed—that is, whether water is held in the lumen. Cotton linters and rayon fibres (which are non-collapsing) show the same critical moisture content by both methods.

The direct measurement of fibre collapse in moist fibres is inconvenient; however, the ability of a fibre to collapse is closely related to its flexibility,[11] since deformability of the cell wall is the governing factor in both cases. The relationship may be imperfect when flexibility results from local mechanical damage to the fibre, local points of weakness[12] or if the fibre is already collapsed.

A graph of the flexibility index plotted against the difference in critical moisture contents (Fig. 9) gives a fair correlation for unbeaten, undried kraft and sulphite spruce fibres of varying yields.

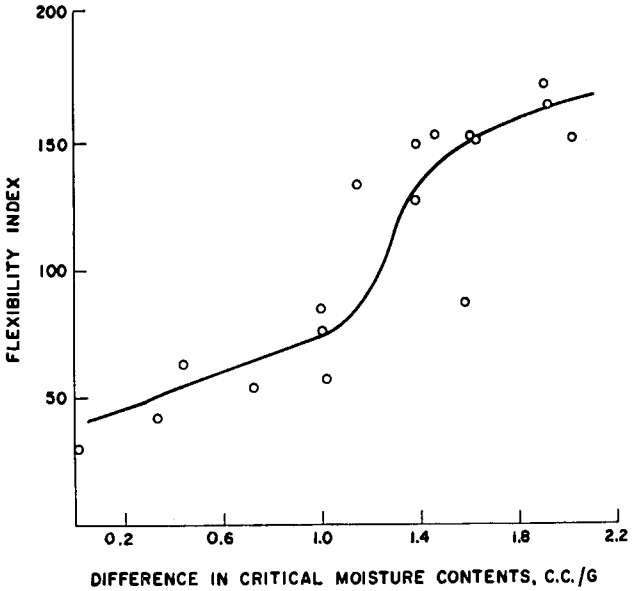

Fig. 9—Fibre flexibility as a function of the difference between critical moisture contents determined by the hydrostatic tension and dye migration techniques respectively: the pulps are all spruce, but include both sulphites and krafts

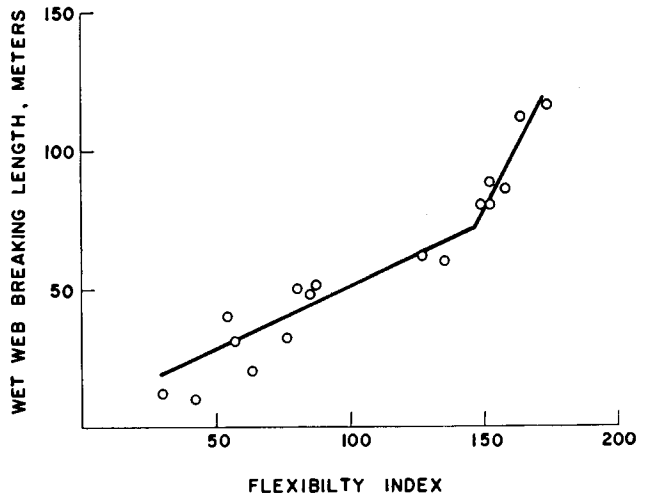

Fig. 10—Wet web tensile strengths measured in the plateau region[8] as a function of the flexibility index: the pulps are similar to those in Fig. 9

Points on the extreme left of Fig. 9 represent fibres that do not collapse in the initial stages of drying and, on the right, flexible fibres that collapse almost completely as they are dried. The remainder show intermediate behaviour. Some evidence in verification has been found in cross-section studies and in wet web and paper properties. Examination by microscope after drying and reslushing pulps reveals air occlusions in the non-collapsing fibres.

The following consequences of the variation of flexibility and collapse have been observed—

(*a*) The increase in the density and strength of handsheets by controlled wet pressing is relatively greater for pulps represented in the centre of Fig. 9, presumably because collapse is assisted mechanically.

(*b*) All pulps when beaten increase their flexibility and the difference between the two critical moisture contents. The pulps to the right of the graph do so by increasing the hydrostatic tension value presumably by fibrillation effects, whereas pulps to the left do so by also *decreasing* the dye migration value owing to facilitated fibre collapse.

(*c*) The wet web strength properties—both breaking length and stretch—are strong functions of the fibre flexibility (Fig. 10). A similar relation between dry sheet properties, flexibility and collapse (other variables being equal) is anticipated, but remains to be fully demonstrated.

Acknowledgements

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REFERENCES

1. Emerton, H. W., Page, D. H. and Hale, W. H., this vol., 53–99
2. Jayme, G. and Hunger, G., this vol., 135–170
3. Page, D. H., Tydeman, P. A. and Hunt, M., this vol., 249–263
4. Dadswell, H. E. and Watson, A. J., this vol., 537–564
5. Grant, J., this vol., 573–591
6. Moore, A. T., Scott, L. W., de Gruy, I. V. and Rollins, M. L., *Textile Res. J.*, 1950, **20** (1), 620–630
7. Tamolang, F. N. and Wangaard, F. F., *Tappi*, 1961, **44** (3), 201–216
8. Robertson, A. A., *Tappi*, 1959, **42** (12), 969–978
9. Barkas, W. W. and Hallan, R., *Proc. Tech. Sect. B.P. & B.M.A.*, 1953, **34** (2), 289–310
10. Preston, J. M. and Bennett, A., *J. Soc. Dyers Colourists*, 1951, **67** (3), 101–103
11. Robertson, A. A., Meindersma, E. and Mason, S. G., *Pulp & Paper Mag. Can.*, 1961 **62** (1), T3–T10
12. Forgacs, O. L., *Tappi*, 1961, **44** (2), 112–119

DISCUSSION

MR. P. E. WRIST: One statement made during the presentation of this paper was of great interest to me—that beating has the effect of stiffening a fibre. I think this is an important statement and worthy of more discussion.

It is often stated in the literature that one of the effects of beating is to make the fibre more flexible. In our studies of fibre networks in suspensions, however, we have always had difficulty in explaining why the network strength of a beaten pulp was lower than that of the unbeaten pulp. The two effects of fibrillation and of increased fibre flexibility (which are usually attributed to beating) should both have made the flocs stronger and the only factor by which we could explain this weakening of the floc structure was the shortening of the fibres. Having measured fibre lengths, we found that in the cases we studied we had not shortened the fibres very much. An enormous dependence of strength on fibre length would therefore offset the other two effects. If we can accept, however, that beating stiffens the fibres in suspension, then our experimental data can be explained more reasonably.

The distinction you have made between increased stiffness in the wet swollen stage and increased flexibility during the drying phase is not adequately appreciated in the literature.

DR. O. J. KALLMES: With regard to discussions on the flexibility of fibres, we must keep in mind the axis of reference of the fibre's cross-section. Fibres that have undergone mechanical treatment tend increasingly to collapse, making them more flexible around their y axis, but less around their z axis. The former effect enhances the bonding ability of fibres, the latter makes them more rigid within the sheet when the sheet is strained.

PROF. G. JAYME: Giertz has pointed out the difficulties of removing lignin from plant materials without removing hemicelluloses. There are two approaches. Maass showed about 30 years ago that a sulphite cook of wood at 50°C will yield a pulp that may be regarded as a holocellulose containing practically all of the hemicelluloses originally present. Another way has recently been used by us—a cold soda pulp was prepared from poplar wood at about 93 per cent, which could be delignified easily with sodium chlorite practically without removal of hemicelluloses. These could be removed by treatment with caustic soda solutions of various concentrations. The strength data on the pulp prepared in this way indicated that strength increased very

considerably with lignin removal, whereas the influence of the hemicelluloses was less pronounced. The highest strength data were obtained when all the lignin and part of the hemicelluloses were removed.

DR. G. N. RICHARDS: To what extent is fibrillation affected by hemicelluloses alone? For instance, what is the fibrillation behaviour of a holocellulose in which only the lignin has been removed and all the hemicelluloses remain? —are the fibrils held together by a hemicellulose glue?

PROF. H. W. GIERTZ: It is quite possible to fibrillate holocellulose fibres, but, because of the slippery consistency, it must be done in the right machine. The Lampén ball mill, for instance, is unsuitable.

DR. S. G. MASON: Giertz has stated that lignin is hydrophobic. In view of the fact that water is sorbed by lignin (as shown by our own work and that of Christensen in Australia) and that water acts as a plasticiser for lignin, I find this concept doubtful. What is the experimental evidence for this claim?

PROF. GIERTZ: For small molecules, the terms *hydrophilic* and *hydrophobic* are well-defined. A problem occurs when dealing with large molecules and macro-molecules. Propanol, glycerol and phenol are hydrophilic and anisole (methoxybenzene) is hydrophobic—but what about lignin? Many substances are neither typically hydrophilic nor typically hydrophobic, they are something in-between and may be graded in this respect over a very wide spectrum.

From a practical point of view, the words should in each case be used appropriate to the conditions concerned. Let me take an example. Secondary cellulose acetate with a D.S. of 2·0–2·5 is less hydrophilic than cotton and viscose rayon are, because it absorbs less moisture and is plasticised less in laundry operations. As a textile, secondary cellulose acetate belongs to the hydrophobic filaments. This is still truer for cellulose acetate with a D.S. of 2·8–2·9. On the other hand, a cellulose acetate film with a D.S. of 2·9 is considered to be hydrophilic when used as a loudspeaker membrane, because it becomes slightly plasticised by the moisture in the air. For this purpose, it has to be a true triacetate.

Hemicellulose is hydrophilic, but a sol of cellulose micelles, prepared by hydrolytic degradation of wood cellulose is classed by Rånby as hydrophobic, despite the fact that the precipitated sol material absorbs moisture. There is such an important difference in behaviour towards water between a groundwood fibre and a holocellulose fibre or between precipitated gamma-cellulose and precipitated alkali lignin that to my mind this difference is expressed most simply and accurately by using the words hydrophilic and hydrophobic.

MR. J. W. SARGENT: I should like some comment on the fact that patterns of microfibrils similar to those Giertz has visualised for the S2 cell wall layer are to be found in such two completely different fibres as bleached sulphite and unbleached kraft.

PROF. B. G. RÅNBY: It is not a question of the strength of the individual hydrogen bonds and we know that the hydrogen bond energies are between 3 and 10 kcal/mol. Under certain conditions, you can form many ordered hydrogen bonds (as in crystalline regions), in ordered adsorption of hemicellulose chains on to cellulose microfibrils or in ordered aggregation (association) of a bundle of cellulose microfibrils. The resistance of such systems, say, to swelling with water is because the swelling reagent must give a simultaneous opening of a whole sequence of identical bonds and this is thermodynamically unfavourable, because it is unlikely. The insolubility of cellulose in water can be interpreted along these lines.

PROF. JAYME: The influence of hemicelluloses on light scattering in handsheets is very marked. Jayme and Pommer proved many years ago, using the Kubelka–Munk method and formula, that the scattering coefficient of handsheets closely followed the alpha-cellulose content changes obtained by the caustic soda treatment of pulps—with progressive removal of hemicelluloses, the handsheets became more opaque.