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A NOTE ON
STRUCTURE OF PAPER AS REVEALED BY THE SCANNING ELECTRON MICROSCOPE

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AN examination has been made of eight paper samples by means of the scanning electron microscope at the Pulp and Paper Research Institute of Canada. This is the first step in a programme of examination of paper surfaces and fractures that is planned for this instrument. The scanning microscope(1, 2) bridges a gap between the capabilities of the light microscope and the conventional transmission electron microscope and the results obtained in this study may therefore be of interest to those attending a symposium on paper structure. The capabilities and limitations of the two more conventional techniques are described in detail in other papers of the symposium and will not be discussed here.

To illustrate the principal findings of the study only three specimens are illustrated here. These are all samples of kraft paper having a basis weight of 60 g/m^2 made in a British sheetmachine under standard conditions of forming, pressing and drying; they have all been examined on the blotter (top) side. These three papers, which were obtained from pulps from the same log of white spruce, are—

1. A 64 per cent yield unbeaten sheet.
2. A 47 per cent yield unbeaten sheet.
3. A 47 per cent yield beaten sheet, 460 CSF.

The physical properties of the paper are given in Table 1.

In addition to the surface of the papers, we have also examined the tensile fractures of the same papers and a few of these pictures, which have a bearing on the structure of paper, are presented.

To prepare a sample for the scanning microscope, it is mounted on a 1 cm diameter stub and coated uniformly with a 200 Å thick layer of metal in a vacuum evaporation unit and examined directly, that is to say, without the

TABLE 1—PHYSICAL PROPERTIES OF THE PAPERS

Specimen	*Yield, %*	*Canadian standard freeness, ml*	*Beating time, min*	*Breaking length, metres*	*Bulk, cc/g*	*MIT fold*
1	64	732	0	4 870	2·23	165
2	47	638	0	10 450	1·57	1 209
3	47	460	46	15 600	1·30	2 400

necessity of making a replica. The specimen is mounted in the microscope with its surface held at an angle of 45° to the electron beam and appears in the pictures as if the top of the picture was tilted at 45° away from the observer. The magnifications given apply to the horizontal direction at the centre of the picture.

The first three pictures are of specimen *1*, the high-yield, unbeaten sheet. A general view is shown in Fig. 1 and higher magnifications of a fibre crossing are shown in Fig. 2 and 3. In comparison with the two specimens to be illustrated later, the following statements can be made—

1. The fibres are relatively stiff and do not conform well to each other.

2. The fibres have not bonded well to each other as evidenced by their complete separation in some cases.

3. There is very little evidence of fibrillation.

The next four pictures are of the unbeaten, low-yield handsheet. Fig. 4 is a low magnification shot of a poorly bonded area and Fig. 5 is a higher magnification of the central area in Fig. 4. Fig. 6 is a low magnification shot of a better bonded area and the central bond crossing is magnified in Fig. 7. By comparison with the high-yield handsheet, it can be said of this specimen that—

1. The fibres conform to one another better and have collapsed more than have the high-yield fibres.

2. There is now some fibrillar attachment between fibres.

The next three pictures are of paper made from the same pulp as the previous sheet except that it had been beaten for 46 min to 460°CSF. Fig. 8 is a low magnification picture and Fig. 9 and 10 are higher magnifications of the central region in Fig. 8. By comparison again, it can be said that—

1. The fibres have conformed very well to one another and have collapsed still further.

2. There is considerable external fibrillar attachment between fibres.

Fig. 1—Unbeaten kraft, 64 per cent yield (×116)

Fig. 2—Unbeaten kraft, 64 per cent yield (×673)

Fig. 3—Unbeaten kraft, 64 per cent yield (×1884)

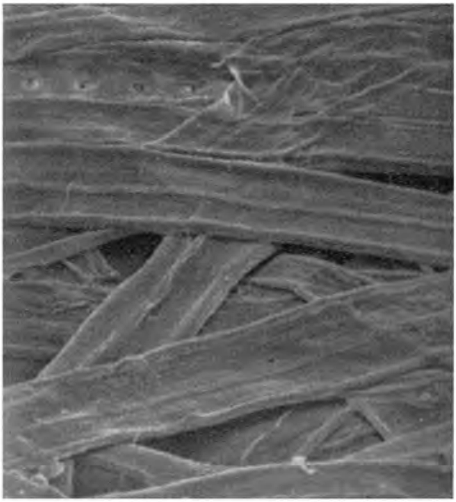

Fig. 4—Unbeaten kraft, 47 per cent yield (×325)

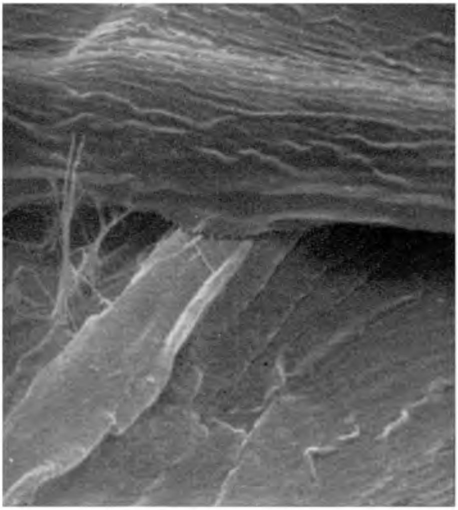

Fig. 5—Unbeaten kraft, 47 per cent yield (×3800)

Fig. 6—Unbeaten kraft, 46 per cent yield (×610)

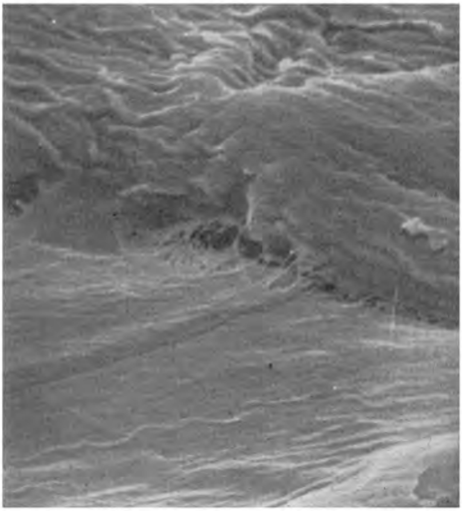

Fig. 7—Unbeaten kraft, 46 per cent yield (×3658)

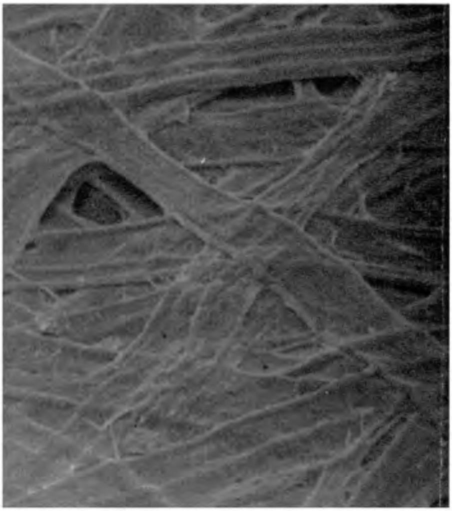

Fig. 8—Beaten kraft, 460 CSF, 47 per cent yield (×293)

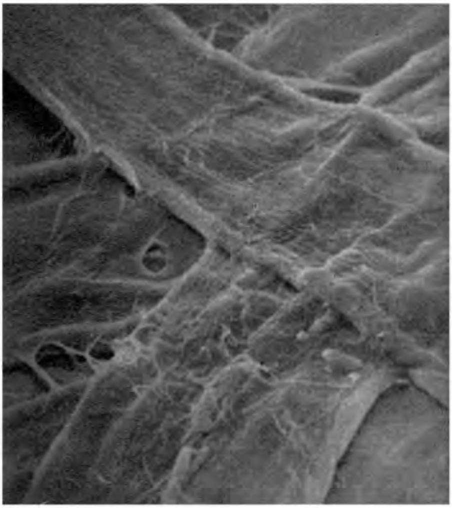

Fig. 9—Beaten kraft, 460 CSF, 47 per cent yield (× 1025)

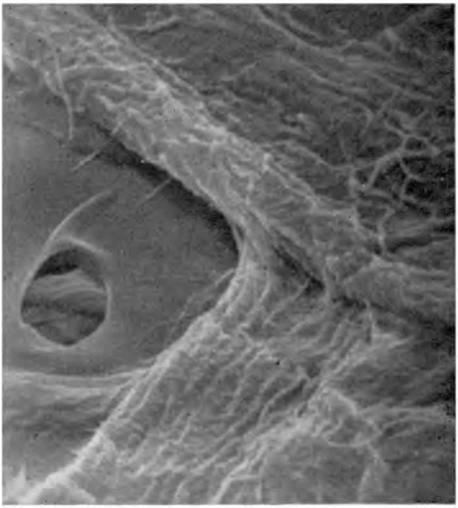

Fig. 10—Beaten kraft, 460 CSF, 47 per cent yield (× 3444)

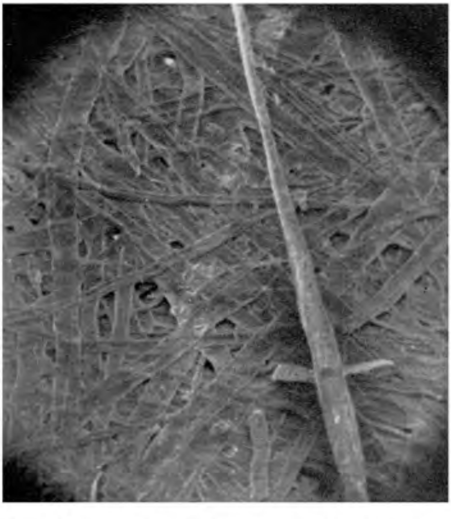

Fig. 11—Tensile failure: unbeaten kraft, 47 per cent yield (× 78)

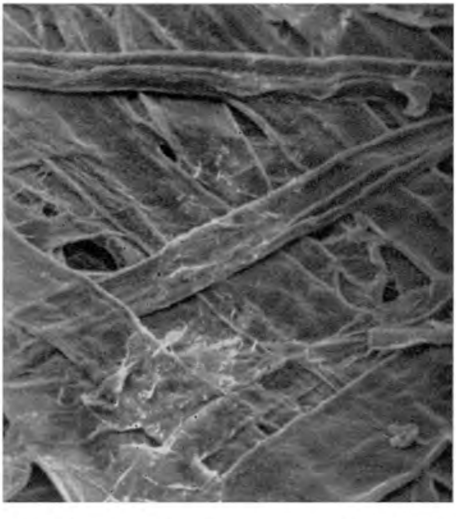

Fig. 12—Tensile failure: unbeaten kraft, 47 per cent yield (× 279)

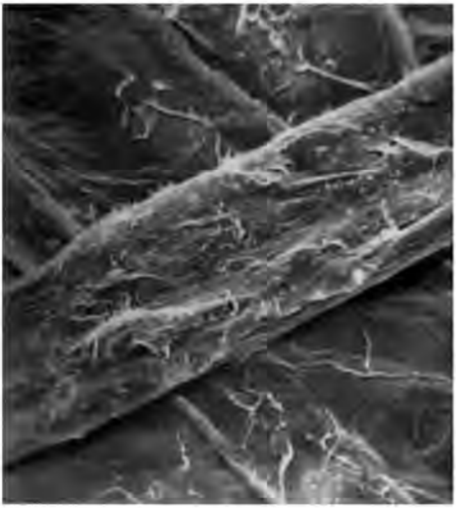

Fig. 13—Tensile failure: unbeaten kraft, 47 per cent yield (×974)

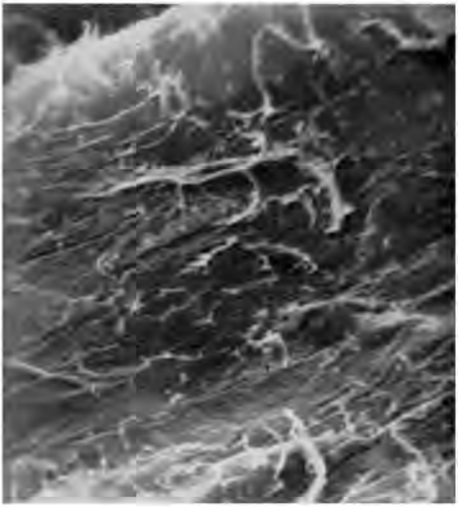

Fig. 14—Tensile failure: unbeaten kraft, 47 per cent yield (×3483)

The tension fractures of the three varieties of paper above have also been examined microscopically. The fracture of the high-yield specimen is not illustrated here: it was characterised by few broken fibres and very little disturbance of the surface at the debonded areas of fibres. The unbonding that takes place in the low-yield, unbeaten paper (specimen *2*) is illustrated in the next four pictures. Fig. 11 is a low magnification view, just above the tension fracture that had been produced by stressing in the vertical direction and illustrates the unbonding that occurred when a single fibre that had lain on the upper surface of the sheet was pulled off. The former path of the fibre shows up as a whitened band running almost vertically. Fig. 12, 13 and 14 are successively higher magnifications of the central area in Fig. 11. There is considerable variation in the nature of the debonded surface, but Fig. 13 might be considered as average. Examination of this picture and Fig. 14 reveals fibrils protruding above the base of the fibre, similar to those shown by Jayme.(3) It is notable also that, in the central area of this bond, the characteristic longitudinal wrinkling apparent on the free fibre surface is absent. Therefore, it could be deduced that there was fibrillar contact over the entire apparent area of bond crossing, but that close contact between the bodies of the fibres was limited to the central region. Presumably, the close contact prevented the lateral shrinkage that is responsible for wrinkling.

Finally, Fig. 15 illustrates a debonded fibre crossing in a beaten sheet that had been fractured in tension. In this picture, the fracture lies above the field and the stress direction was vertical. In this debonded area, there are fibrils protruding above the base of the fibre, but there are more, larger fragments (probably bundles of fibrils) than was the case in the unbeaten sheet. Moreover, it appears that the entire area of fibre crossing had been in close contact, the transition from the wrinkled free surface to the flatter debonded area being sharper.

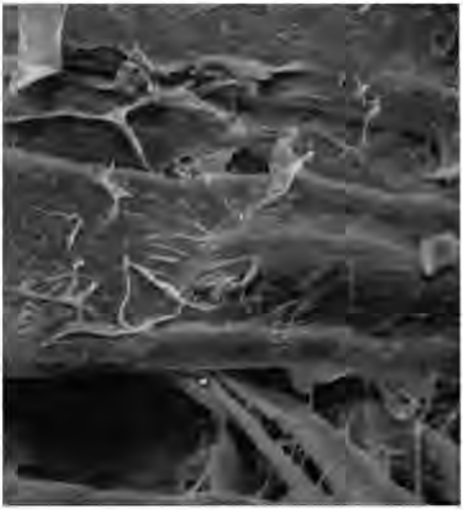

Fig. 15—Tensile failure: beaten kraft, 460 CSF, 46 per cent yield (×974)

Summary

THIS note gives an example of how the scanning electron microscope reveals the structure of paper. A comparative study of three specimens of kraft paper has given visual evidence that (*a*) lowering the yield and (*b*) beating increase the conformability of fibres to one another, hence the amount of close contact between them. The conformability (or flexibility) of the fibres appears to be closely related to their degree of collapse.(4) The amount of fibrillar connection between fibres also is increased by the two means above. From examination of the unbonding produced by tensile failure in the paper sheets, it is observed that the disturbance of the debonded

areas and presumably the amount of material transferred at a debonded junction increases with beating and with lower yields.

Acknowledgement

We would like to thank Mr. M. P. Foley who prepared the specimens and operated the microscope during this examination.

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DISCUSSION

MR. G. F. UNDERHAY: It was a joy to hear Emerton start off his paper by telling us he is taking an area 1 mm^2 as representative of the paper and saying that any 1 mm^2 is the same as any other for this purpose. Thus, the uniformity problem that we heard about a little earlier on is almost solved by him straight off!

There is some uncertainty in the discussion this morning and perhaps there will be an opportunity in one of the later papers to raise the subject of uniformity and distribution again. Meanwhile, has Corte any information about the relationship of the length of the fibres to the work that he was doing?

MR. H. W. EMERTON: In case there is any misapprehension, may I say that my comments on the representativeness of 1 mm^2 referred to the phenomena to which we drew particular attention.

MR. D. ATTWOOD: There are many excellent photographs in this paper and the wire side surface photograph of newsprint (Fig. 4) is interesting in that there is obviously a cavity of about 0·008 in. across. It is not apparent from the photograph how this hole was formed, whether by a random process or by the indentation of a wire knuckle.

We have found that it is very difficult in studying surface photographs at this magnification to distinguish between wire mark and random variations. Random variations can be reduced by averaging and we have produced a photographic technique to do this.*

Of two illustrations shown, the first is a typical surface photograph of newsprint about $\frac{1}{5}$ in. across produced by oblique illumination (Fig. 8*a*). Some indication of wire mark is seen relative to the machine-direction. When the randomness is reduced by the averaging technique, an extremely clear picture of the wire mark is obtained (Fig. 8*b*). It should be stressed that the second illustration was produced directly from the surface photograph: it is not a true photograph of a particular piece of paper, but a photographic average.

*This technique is described in *Paper Tech.*, 1961, **2** (5), T 191–T 198 in a paper by J. R. Parker and D. Attwood and the illustrations referred to appear in the article by the figure numbers quoted.

The effect of the impression of the twill wire can be clearly seen, particularly the indentation of the warp knuckles forming the diagonal pattern.

This technique is applicable also to beta-ray transmission photographs. A further figure (Fig. 1) shows a typical beta-ray photograph, the light areas corresponding to low substance. When the technique is applied to the beta-ray negative, the randomness is reduced and shows a very regular pattern of

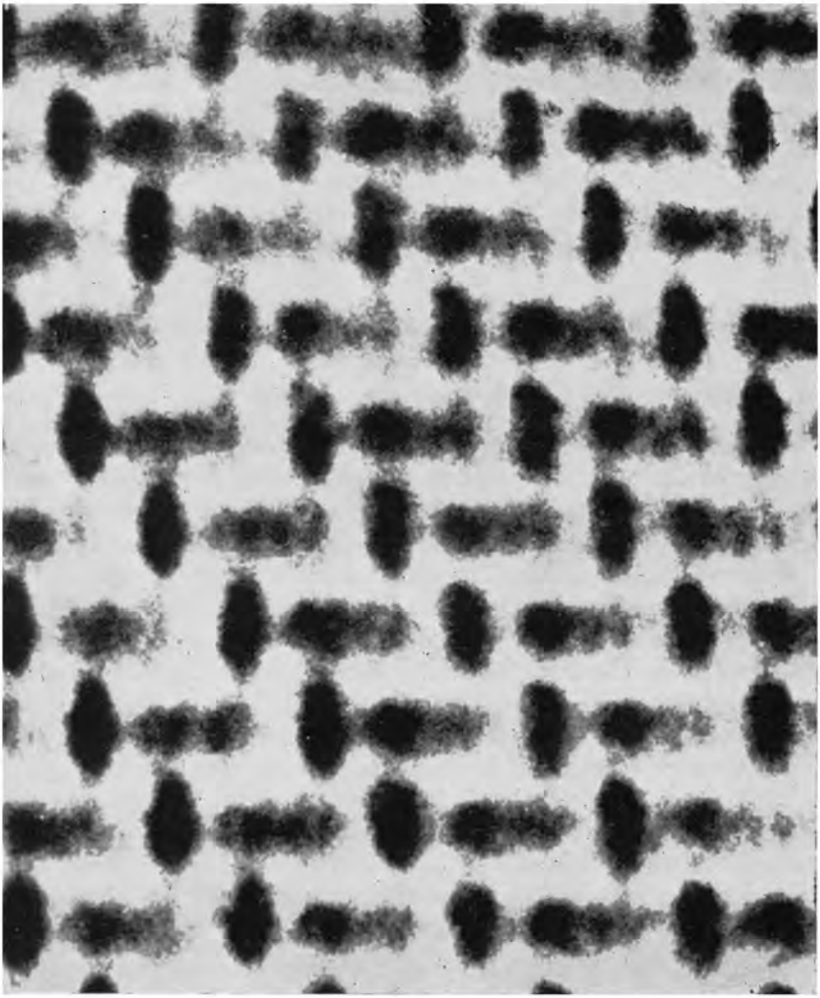

Fig. D1—Multiple print of beta-ray print negative basis weight variation corresponding to the wire pattern (Fig. 6*b*). The regular twill pattern then shows up—one up, two down—and exceptional agreement even in the small scale can be seen. For instance, in the light areas corresponding to the weft knuckles, these are seen to be lighter at the left side and, in fact, an examination of a twill wire shows that the weft knuckle does stick up more at this point (Fig. D1).

A DELEGATE: Would Mr. Emerton comment on the kind of microscope he used?

MR. EMERTON: This is not a particularly special microscope. The specimen had been prepared by making a replica in clear plastic material.* One ends up with a substantially plain specimen of metal with a tone variation sandwiched between clear plastic. There is no confusion of the image.

DR. J. GRANT: Is there any special significance that the fibres from the early stages of beating show an increase in transparency in the case of the unglazed sheet, but not in the glazed sheet? Is this fortuitous?

MR. D. PAGE: There were no fibres in the pictures shown: all you saw were pictures of metal.

MR. EMERTON: There is no question of transparency: the fibres were not there. The replica technique used reveals only the surface form.

DR. J. A. VAN DEN AKKER: It is known that the gloss of paper is irreversibly diminished by exposure of the sheet to high humidity—an effect that might be related to changes in the configuration of the fibres dried under restraint. Have you used your technique to observe the effect of high humidities on the surface of fibres that have been dried in contact with a plate?

MR. EMERTON: We have not done this. It is clearly a valuable thing to do.

PROF. J. D'A. CLARK: A much greater area of fibres in contact with the polished plate after only 5 min beating does not necessarily denote greater fibre flexibility gained in that short beating time. It was probably due rather to the development of fibrils, which drew the structure closer together and hence caused greater pressure on the plate. An excess of tension so developed with longer beating was finally large enough to cause the sheet to part from the plate after the compacting effect of this tension had passed its maximum.

***Svensk Papperstidn.*, 1959, **62** (9), 318–332

MR. T. H. FAREBROTHER: I should like to show one or two slides that relate to the supercalendering effect Mr. Emerton has already illustrated.

The first three slides are photomicrographs of the topside surface of a rotogravure magazine paper taken by oblique top illumination. They show an identical area of the paper in three conditions, the first 'off-dryer' (Fig. D2*a*), the second after laboratory supercalendering (Fig. D3*a*) and the third after 40 min soaking in water and redrying (Fig. D4*a*). The increased smoothness and glossiness of surface produced by the supercalendering is very marked, but more interesting is the degree to which the surface returns to the 'off-dryer' condition after the soaking and redrying. The only noticeable difference from the original condition is a slight reduction in the surface contours.

The reversible nature of supercalendering

Mechanical printing (top side) in three states

All six micrographs record one identical field

The machine-direction of the specimen is aligned parallel to the side of the page

Surface configuration (Fig. D2*a*, D3*a* and D4*a*) was recorded by top illumination in the machine-direction and at 20° to the paper surface

Look-through (Fig. D2*b*, D3*b* and D4*b*) was recorded by transmitted illumination

Magnification × *15*

The other three pictures (Fig. D2*b*–D4*b*) show the same three conditions of the paper, but by transmitted light. The supercalendering is seen to suppress most of the visible detail of the look-through such as the rather spotty appearance and the dark fibres and some new features appear, notably the bright translucent fibres. By soaking and redrying, the original appearance is again restored in great detail, although with some loss of contrast.

The main interest in these experiments is the finding that the visible effects of supercalendering may be reversed almost completely by soaking and redrying, though it should be mentioned that some measured physical properties such as bulk and smoothness showed quite appreciable residual effects from the supercalendering.

MR. P. E. WRIST: In Buchanan's micrographs of strained paper, was the shadowing done before straining or after straining?

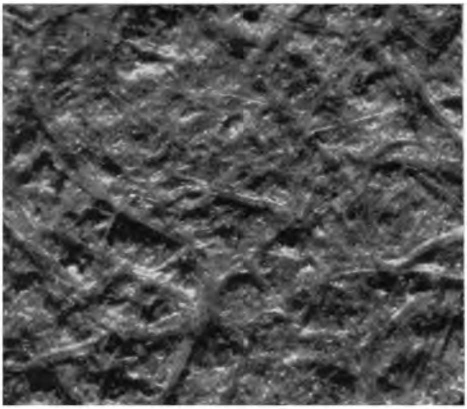

Fig. D2a—Surface detail in the off-dryer condition

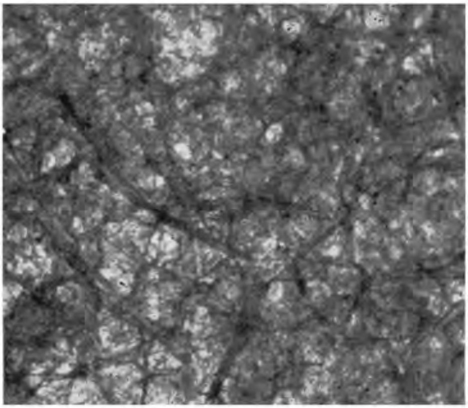

Fig. D2b—Look-through in the off-dryer condition

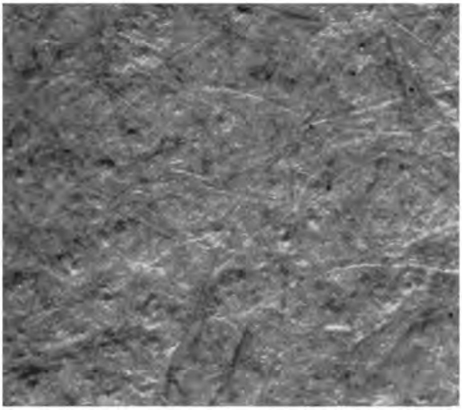

Fig. D3a—Surface detail after moderate laboratory supercalendering

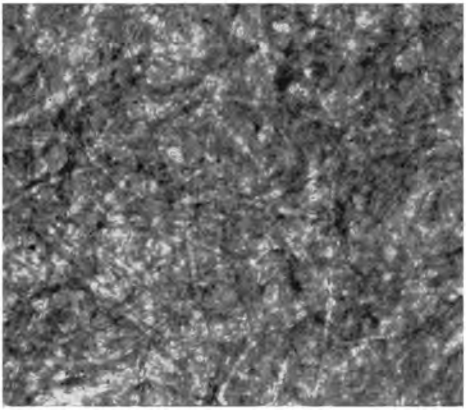

Fig. D3b—Look-through after moderate laboratory supercalendering

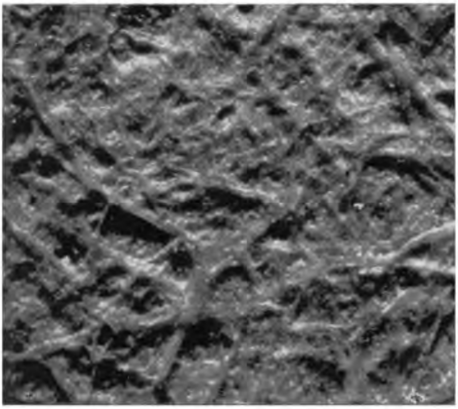

Fig. D4a—Surface detail after soaking and redrying

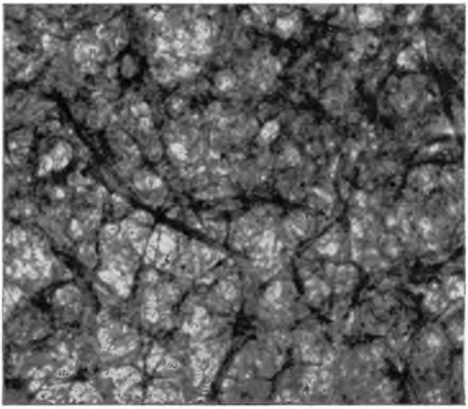

Fig. D4b—Look-through after soaking and redrying

MR. J. D. BUCHANAN: It was done after straining. The specimens were not completely ruptured. We applied the arresting mechanism on the Instron tester, which prevented the full separation of the specimen, though the maximum load had been exceeded.

A DELEGATE: How would you estimate the length of the fibrils forming the bond?

MR. BUCHANAN: In Fig. 14 (p. 106), they are 1–2 microns long.

THE CHAIRMAN: Is it right to say they form the bond or part of it? One has only observed them after the event.

MR. BUCHANAN: Agreed. In the central section of the debonded area in question, the surface is not wrinkled and was probably in close contact with the debonded fibre. Here, the fibrils must have been flattened, but towards the edges of the bond they were free.

MR. J. A. S. NEWMAN: The micrographs show fibrils between fibres at bonded points. More fibrils appear on the wetter beaten fibres. Could these fibrils be formed by the action of the bonds being strained—that is, the fibrils are more easily torn from the surface of beaten fibres, because of the internal damage done to the fibres by beating?

DR. W. GALLAY: It seems to me that we have been presented with strong evidence that failure of the union between fibres very frequently takes place, not in the bond, but rather in the intrafibre structure away from the bond. This is not surprising, since, from analogies with other systems, as I pointed out some years ago, one would expect the intrafibre bond to be stronger than faulted regions with the fibres. Whether we obtain lamellae or fibrils of different lengths or diameter hanging on after two adjacent fibres have parted at the crossover region could merely reflect the state of the structure adjacent to the bonds.

DR. VAN DEN AKKER: In the case of the fibre pulled from the web, could it have been directly involved in the rupture or was it remote from the fracture?

MR. BUCHANAN: It extended across the fracture and could be traced right over the broken zone, with its end still bonded to the other side. It is a very long fibre.

DR. VAN DEN AKKER: Did you see this in any regions removed from the fracture?

MR. BUCHANAN: This was an unusual event. We purposely chose it, because it gave a very good range of types of debonding. Close to the fracture in this sheet, many debonded areas similar to this could be found, but none of them extending back as far as this particular one did. It was only because this was one of the last fibres to be deposited in the sheet mould that the debonding extended back so far.

PROF. A. H. NISSAN: If the length of these little struts and columns between the two structural elements were, say, of the order of 0·1 micron, would you not expect that area to be transparent, even though it is so thoroughly interconnected by strings as to constitute almost, but not quite complete 'bonding'?

MR. BUCHANAN: I think Page might answer that better than I could.

MR. PAGE: I am reserving my comments about the whole question of the fine structure of bonding until this afternoon.

MR. C. A. CHESTER: I would like to refer back to the effects of calendering, whether machine calendering or supercalendering, to the damage done to the surface bonds during that process. There is an increase in the smoothness, but undoubtedly damage to the surface bond. Has Emerton come across this effect? Were his specimens completely untouched after calendering, because I believe that, although the increase in smoothness may be beneficial for some printing processes, the damage done to the surface bonding may be very detrimental and may not show up in smoothness tests. The damaged bonds may be flat under static conditions, but they may cause fibre lifting during printing, when a vertical stress is applied.

MR. EMERTON: The answer is that this particular method does not give sufficiently high results to resolve the detail of bonded areas, so our pictures shed no light on bonding. Clearly Buchanan's technique would throw light on it.

MR. UNDERHAY: Farebrother showed in his slides the reversion of the calendered state. Has he gone further, calendered again and reproduced the same effect—and is that sort of thing capable of being produced indefinitely?

MR. FAREBROTHER: No, we have not examined this matter further.

MR. J. MARDON: Has Chester detailed experimental evidence upon the reduction of surface bonds by calendering? I would suggest one side of the sheet stays about the same and the other side is improved.

MR. CHESTER: Yes, we have a certain amount of evidence and I am afraid it is rather conflicting—to be quite honest, it is still a complete puzzle. We found there is an effect that probably varies with the type of machine on which the paper is made. Some evidence shows that, on one particular Fourdrinier machine, the damage during calendering is apparent on both sides of the sheet; on another Fourdrinier machine that is apparently very similar, the damage occurs much more on the top side of the sheet. I think that the effect is pronounced enough to be worth study. One certain fact is that the number of fibres anchored at one end only to the surface of the paper increases progressively with increasing smoothness obtained by calendering. Of course, this applies only to uncoated paper.

PROF. G. JAYME: I am going to show this afternoon that calendering has a double effect. One is the pressing of microfibrils together to a fairly coherent mass; the other is that some very fine fibrils are at the same time torn out of the surface and lifted from it.

DR. S. W. KINGSNORTH: From our experience, we could expect the erratic changes in paper properties following supercalendering, observed by a previous speaker, to be associated with variability in moisture content in the sheet.

Referring to Emerton's interesting photograph of a fibre forming a spiral as it dried and straightening out again as it re-absorbed moisture, could he tell us the approximate moisture content at which the fibre resumed its straight form?

MR. EMERTON: It was impracticable to measure the humidity as the method of moisture gain was very crude here—sucking acid drops, then breathing over it! You will appreciate that we had little control over the pH value!

Written contributions

MR. F. M. CROOK: The photomicrographs are excellent and the technique most elegant. It has been our experience in microscopic examination of paper surfaces that they are very far from homogeneous and that the variations in surface texture within a single surface of one paper may be as great as the differences between either the back and wire side of a single sheet or those between the surfaces of two sheets that have been given different gradings for some surface property.

To what extent are the micrographs representative fields? How much personal choice was involved in selecting the areas photographed as being typical of the surface of the specimen?

MR. H. W. EMERTON: The fields were not selected by co-ordinates taken from tables of random numbers. Nevertheless, we believe that, so far as the phenomena described in our text and presentation are concerned, the fields are representative. Experienced microscopists are well aware of the dangers of generalising from the particular!