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STRUCTURE OF PAPER IN CROSS-SECTION

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Synopsis—Previous work on the sectioning of paper has not fully utilised the power of the light microscope. This paper describes techniques developed to enable the structure of paper to be seen in considerable detail in cross-section. The techniques are illustrated by sections of a wide range of types of paper. The consolidation of the structure of paper during manufacture is revealed by micrographs of the effects of beating, pressing, drying with and without restraint, supercalendering and creping.

Introduction

IN RECENT years, there has been increasing interest in the structure of paper. Kallmes & Corte[1] have described mathematically the random arrangement of fibres in paper; several authors from the Pulp and Paper Research Institute of Canada using the scanning electron microscope[2] have studied its surface; Jayme and his co-workers[3] using the transmission electron microscope have examined the surfaces of paper at somewhat higher resolution; the work at Kenley[4,5] has revealed considerable information on fibre-to-fibre adhesion and the structural changes that take place during drying. In all these approaches, paper structure has been considered from substantially the same aspect, namely, the plan view. It occurred to us that considerable advantage was to be gained from an examination of the structure of paper in cross-section, in somewhat greater detail than has been possible previously. This paper describes the ground that has been covered.

Previous work on the cross-sectioning of paper for light microscopy has used standard microtomes and a variety of embedding media. In the great majority of cases, sections have been cut of the order of 10–50 μ. One of the purposes of this paper is to draw attention to the fact that there are good

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theoretical grounds for believing that, for use in the light microscope, such sections are too thick. In fact, they exceed the depth of field of even a medium power objective and it is not to be expected that such a thick section will be in sharp focus throughout. The image can therefore be considered as a series of superimposed images, the majority of these being out of focus. Such a condition obtains in the majority of applications of microtomy, but it is often acceptable, since generally the contrast in the image comes from absorption of light caused by the differential staining of minute areas of interest. The refractive index of the section does not change appreciably from one gross area to another. In the case of embedded paper, however, we are dealing essentially with a two-phase system, consisting of discrete cellulose fibres, embedded in a matrix. The extensive interfaces that occur between these phases give rise to considerable refraction effects, which in a thick section can seriously impair image quality. Furthermore, a section will contain a great number of fibres that lie at an angle to the direction of illumination, so that the outline of the fibre in a thick section will be blurred. These comments are illustrated by the series shown in Fig. 1 and 2. It is clear that the definition of the image improves as the section becomes thinner, but very thin sections tend to have less contrast and also tend to break during cutting. For optimum information, a section in the region of 4 μ thick appears to be a most satisfactory compromise. The remainder of this paper will deal with the methods by which such sections can be produced and the results that we have obtained so far using this technique.

Technique

THE techniques described here are with some modification substantially those in common use for the cutting of sections for electron microscopy. In electron microscopy, section thicknesses of the order of 1/30th–1/50th of a micron are necessary, thus the less stringent requirements of sectioning for the light microscopy are met with relative ease. Briefly, requirements for cutting thin sections are—

(*a*) The embedding of paper in a suitable matrix.
(*b*) The use of a knife sufficiently sharp, robust and rigid to permit such sections to be cut; it is important to note that the normal metal knives are quite inadequate and, in practice, glass or diamond knives are essential.
(*c*) The use of a microtome sufficiently well engineered to provide small reproducible advances of the specimen and a cutting action free from chatter.

These conditions are most readily met by using an ultra-microtome and specimen techniques as used in electron microscopy, but we have made a

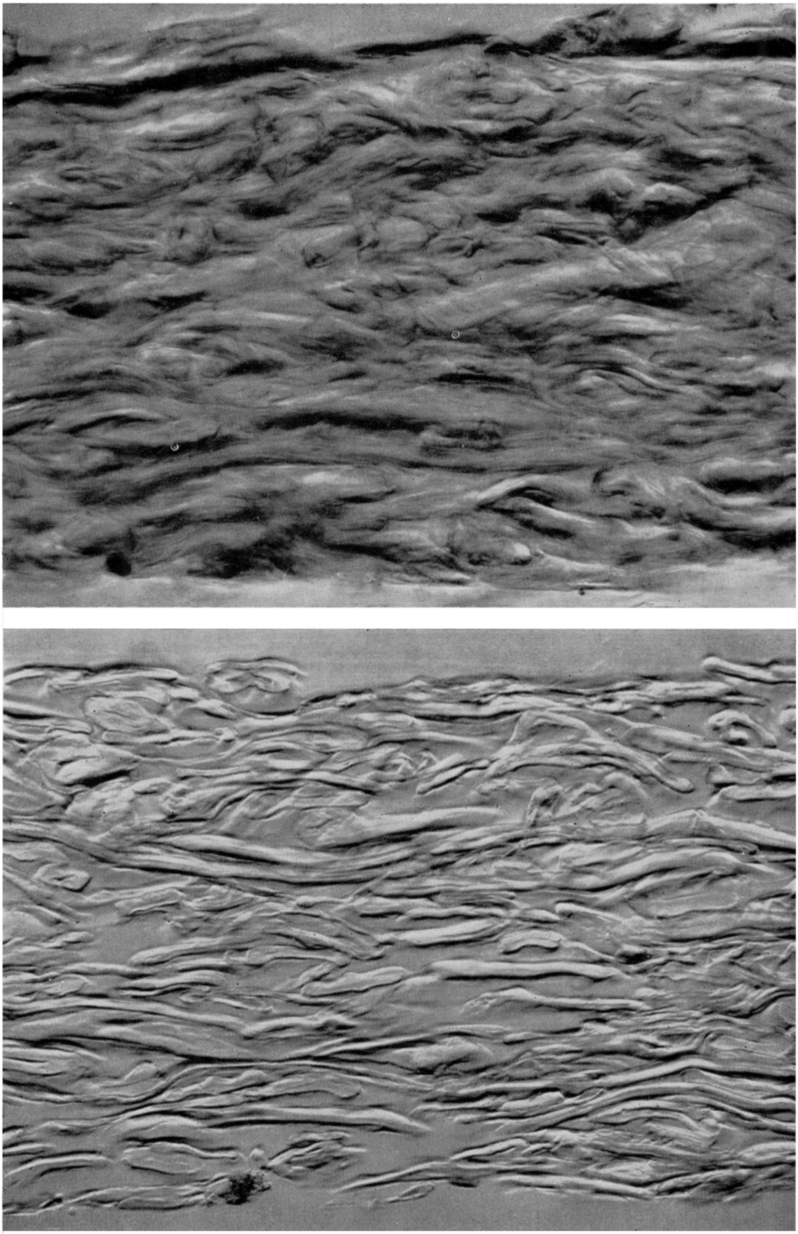

Fig. 1—Softwood sulphite paper—(*a*) 32 μ, (*b*) 16 μ, oblique transmitted light [× 380]

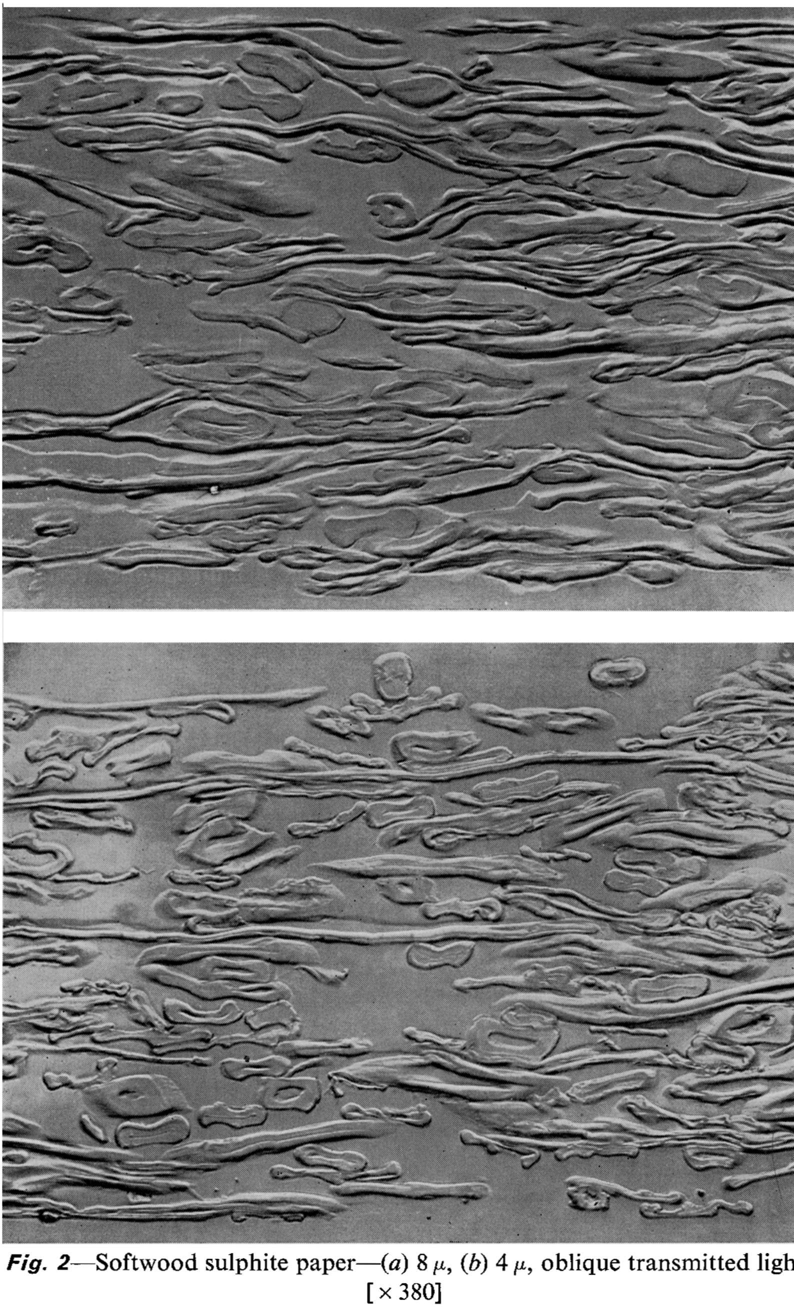

Fig. 2—Softwood sulphite paper—(*a*) 8 μ, (*b*) 4 μ, oblique transmitted light [$\times$ 380]

brief investigation of the possibility of cutting these thin sections with ordinary light microscope microtomes. Provided these are fitted with glass knives, they can under favourable conditions of rigidity of the instrument and care of the operator produce satisfactory results. The techniques described below are those that we have adopted.

Embedding

The embedding medium used throughout this work has been a 4:1 mixture of butyl and methyl methacrylate. This was originally suggested for electron microscopy by Newman *et al.*[6] and has found considerable usage in this field. It has been adopted for light microscope work by several workers and has been used for paper by Jayme.[7] The embedding procedure is briefly as follows.

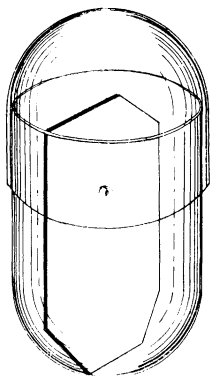

Fig. 3

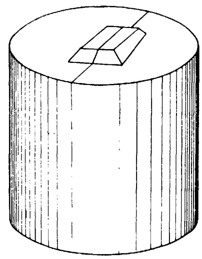

Fig. 4

The acrylic monomers are supplied with stabilisers, which may be removed by a prior treatment with caustic soda. After washing, the methacrylate mixture is dried and 2 per cent weight per volume of benzoyl peroxide is added as a catalyst. After further drying, the monomer may be filtered, when it is ready for use. Embedding of the paper is carried out in small gelatine capsules about 2 cm long and 0.5 cm diameter (Fig. 3). The paper strip is cut to fit the capsule and is inserted before pouring the monomer in. The methacrylate is polymerised under an infra-red lamp. Details of the method can be obtained from textbooks on electron microscopy procedure.[8]

Sectioning

As mentioned earlier, we have used throughout our work diamond or glass knives, since these alone appear to have the necessary properties for cutting thin sections of paper. A diamond knife consists of a single diamond ground to a certain angle and embedded for convenience in metal. It is usual for the knife edge to be 2–3 mm long. Glass knives consist of the fracture edge of a piece of plate glass prepared immediately before usage. The method of preparation can be found in reference books.[8] (Such knives begin to deteriorate after a small number of cuts, but, since they are readily made, they can be regarded as expendable.) The useful length of knife obtained from $\frac{1}{4}$ in plate glass is usually 2–3 mm.

The block of embedded paper must be prepared for sectioning in such a way that the size of area cut is suitable for the length of the cutting edge available. We have found that the best results are obtained by cutting the paper with the knife edge parallel to the plane of the sheet. The length of the paper cut is therefore limited by the length of the knife edge, say, 2 mm. This size of block face is prepared from the original block by extremely careful trimming with razor blades to produce the shape indicated in Fig. 4. In this way, quite a small cutting face is prepared while adequate support for the block face is provided. The block is then mounted in a chuck in such a way that maximum support is given to the somewhat flexible block and the chuck is oriented in the microtome so that the paper is parallel to the knife edge. The arrangement on the Huxley ultra-microtome that we have used is shown in Figs. 5 and 6. The first few sections cut are not collected, since they will be either incomplete or disturbed, owing to the block-trimming process. Subsequent sections are collected from the dry knife with tweezers and are transferred to a slide for mounting.

Mounting

It is the common experience of microtomists that compression of sections can occur during cutting, owing to the action of the knife on the specimen and that this becomes particularly noticeable for thinner sections. Electron microscopists often remove this foreshortening by softening the embedding medium with either a solvent vapour or a weak solution of solvent in water. We have found that, for sections of methacrylate a few microns thick, compression of the section is normally 10–20 per cent; and it is essential for critical work that this compression should be removed in a reproducible way. We have accomplished this by placing the section on the mounting liquid on a glass slide and heating it for 2 h at 35°C. This treatment apparently anneals the strains that are put in during sectioning and generally gives a section that has the same geometry as the face of the block from which it derived.

The choice of mounting medium is somewhat difficult; many non-polar liquids are excluded, because they would swell or dissolve the methacrylate

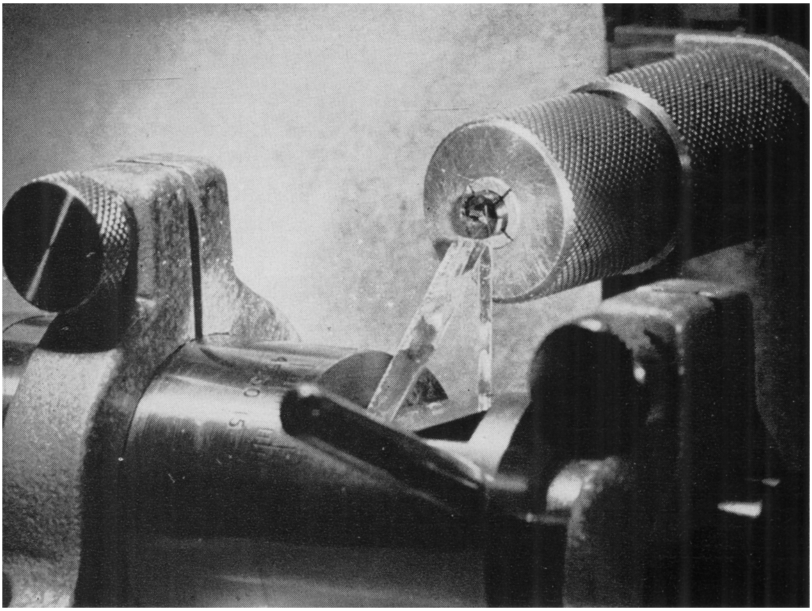

Fig. 5

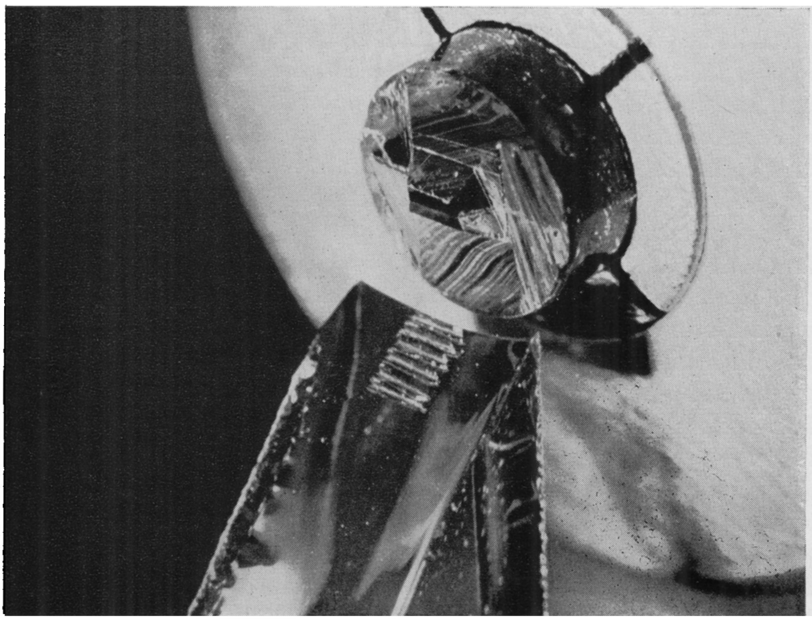

Fig. 6

and many polar liquids would swell the cellulose to give rise to a distortion of the fibre structure. Furthermore, the refractive index of the mounting medium must not be too far removed from that of the cellulose and methacrylate. These considerations eliminate most of the commonly used mountants such as Canada balsam, glycerine, water, gelatine and polyvinyl alcohol.

The work of Britt,[9] who measured the tensile strength of paper in various liquids, led us to try some of the liquids that had little effect on the tensile strength in the belief that these liquids would have the least swelling effect on cellulose. These liquids would of course generally be those most likely to dissolve the methacrylate, but the notable exception in Britt's list is polyethylene glycol, which, besides not dissolving methacrylate, does not swell cellulose, because of its high molecular weight. We have therefore used polyethylene glycol 400 as a mountant throughout our work and have checked its effect on the swelling of paper to satisfy ourselves on this point. Liquid paraffin also seems to be a satisfactory possibility as a mountant.

Such a mount can be regarded as semi-permanent, in that the mountant is sufficiently viscous and non-volatile for the slide to be kept indefinitely, if it is suitably stored.

Micrography

Special consideration must be given to the characteristics of the image produced from sections prepared in this way. At medium to high powers in transmitted light, sections of paper are almost completely invisible, owing to the fact that the only effect on the condensed illumination is refraction of the light, which is not generally sufficient to allow it to deviate from the objective lens. Only in the case of papers containing special fillers (for example, titanium dioxide), for which the refractive index is greatly different from that of methacrylate, is the refraction of light appreciable. Papers containing such fillers give acceptable micrographs in ordinary transmitted illumination.

The most usual method of increasing contrast of transparent sections is by staining and useful results can be obtained by this method, but it must be borne in mind that the staining procedure usually carried out in aqueous solution will involve a swelling of the cellulose and so give rise to some uncertainty in the interpretation of the final results in terms of the original structure.

An alternative method for obtaining contrast, which is particularly useful for outlining the surface of each fibre, is obtained using transmitted illumination with a decentred crescent-shaped aperture in the lower focal plane of the condenser. This form of illumination gives to the image the illusion of a shadowing effect—for example, Fig. 7.

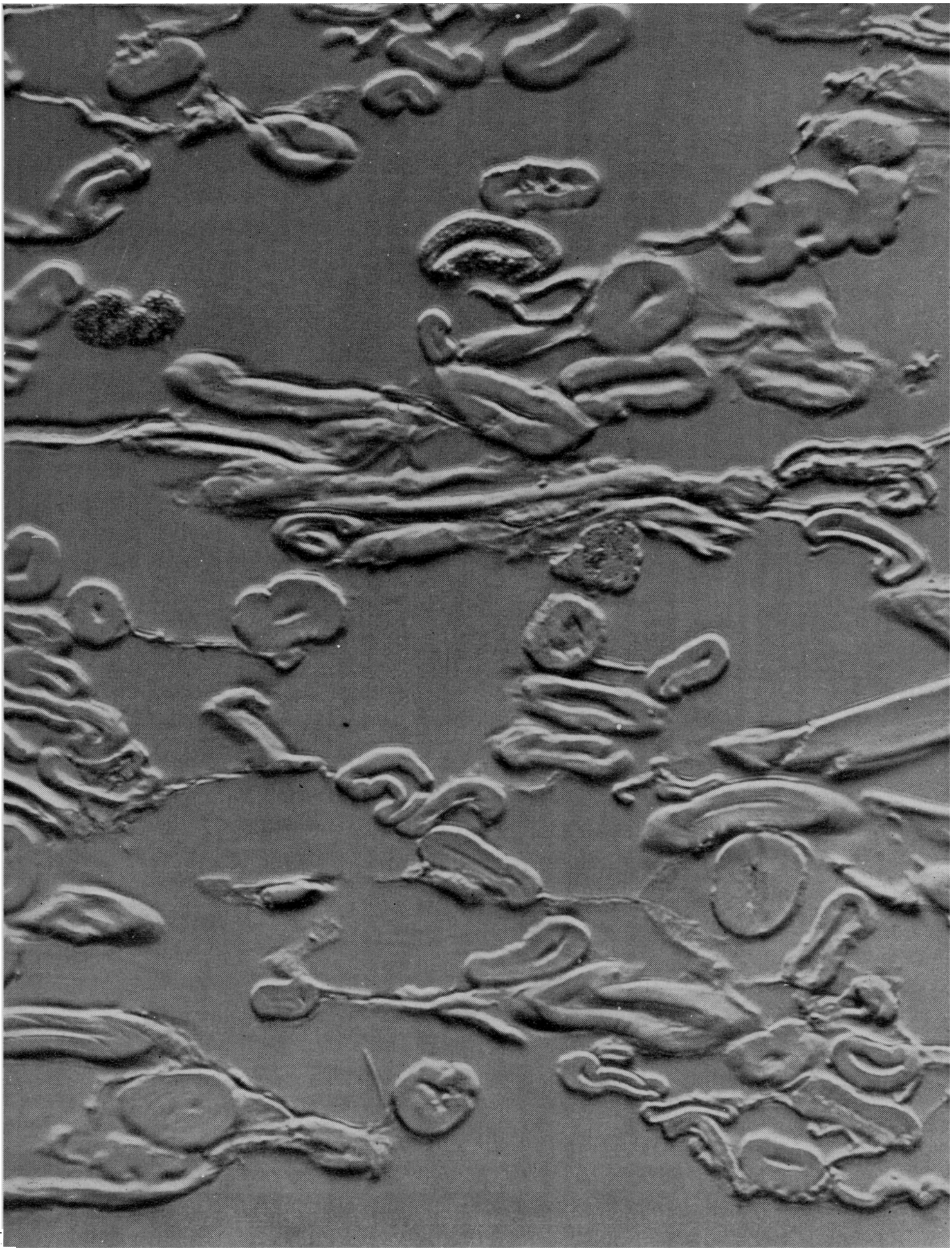

Fig. 7—Blotting paper, oblique transmitted light [× 770]

A further method of obtaining contrast is by the use of transmitted illumination with crossed polariser and analyser. The brightness of a section of a cellulose fibre will depend on three factors—the thickness of the section, the birefringence of the fibre and the direction of the fibre axis both with respect to the plane of the section and the direction of the polarisation. It will be seen that the brightness of any part of a section of paper arises from somewhat complex causes and each image must be interpreted individually. As will be seen later, for certain purposes, this can be an extremely useful method of examination.

Interference microscopy based as it is on revealing areas of differing refractive index is a very good method of examination of sections of paper, but, because a special microscope is needed, no illustrations of this method have been given in this paper.

Applications

A PRELIMINARY examination of the value of this technique applied to a number of problems has been made and these are illustrated by the micrographs (Fig. 7–25).

Blotting paper

The structure of rag blotting paper (Fig. 7) shows marked differences from other papers. The high void volume is apparent, as is the tendency to cylindrical form of the cotton fibres. A few synthetic fibres are recognisable, some of which contain filler particles. Of particular interest is the presence of fibrillar material (which from serial sections can be seen to consist of membranes). These span substantially in the plane of the sheet between fibres and in this type of paper at any rate give the impression of being of significance in contributing to sheet strength.

Pulp laps and effect of sectioning direction

It has been found that the most satisfactory results are obtained by sectioning the block at right angles to the plane of the paper. A comparison between this method and cutting in a direction parallel to the plane of the paper is shown in Fig. 8 and 9. Cutting parallel to the plane produces considerable compression in the fibrous structure. This is made clear by examination in polarised light. Note particularly the highly compressed longitudinal fibres at the bottom of Fig. 9. There is also a greater tendency for the section to break in the plane of the sheet, when it is cut in this way.

Fig. 8—Dry lap of softwood sulphite pulp with direction of cutting along the length of the page, polarised light [× 770]

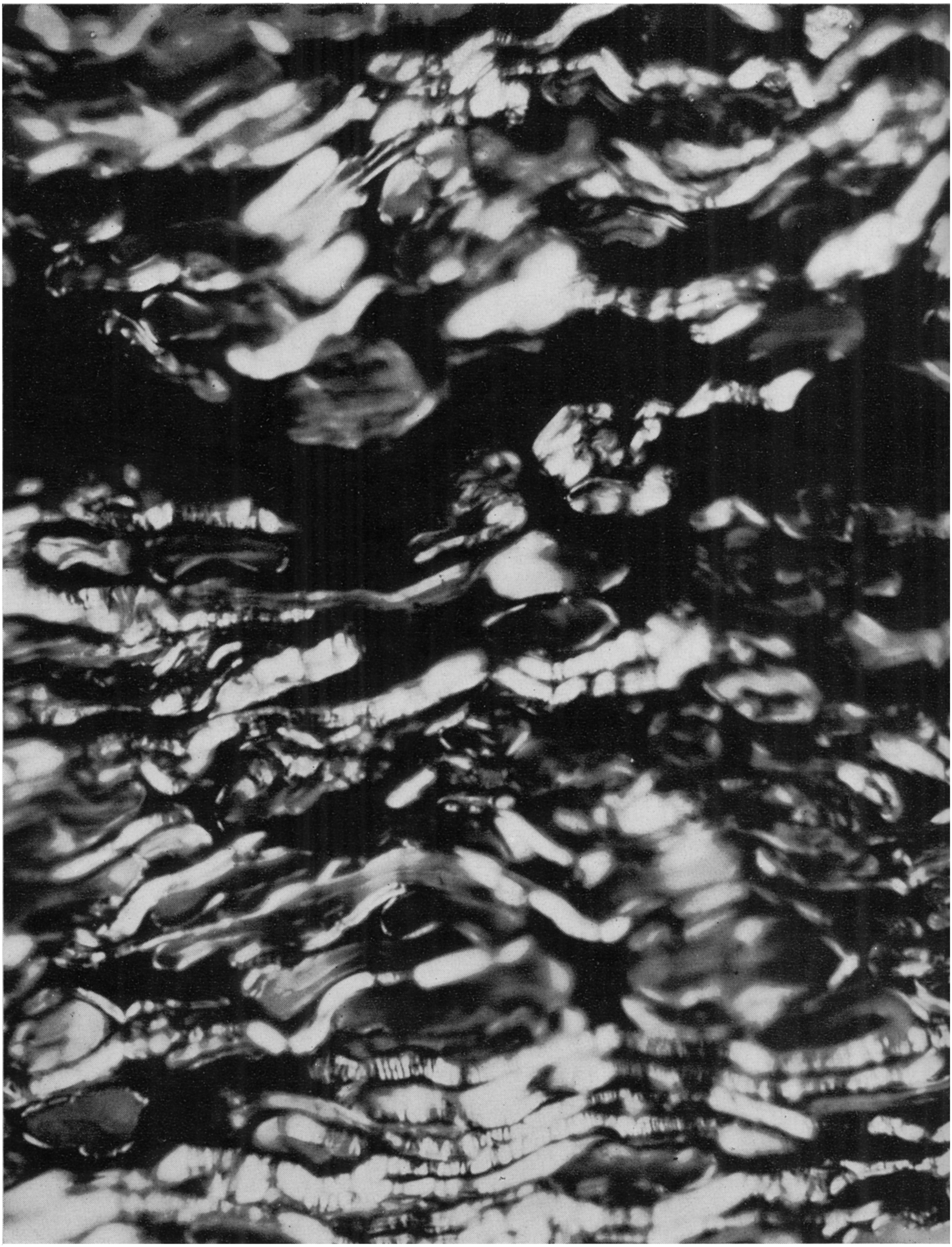

Fig. 9—Dry lap of softwood sulphite pulp with direction of cutting across the page, polarised light [× 770]

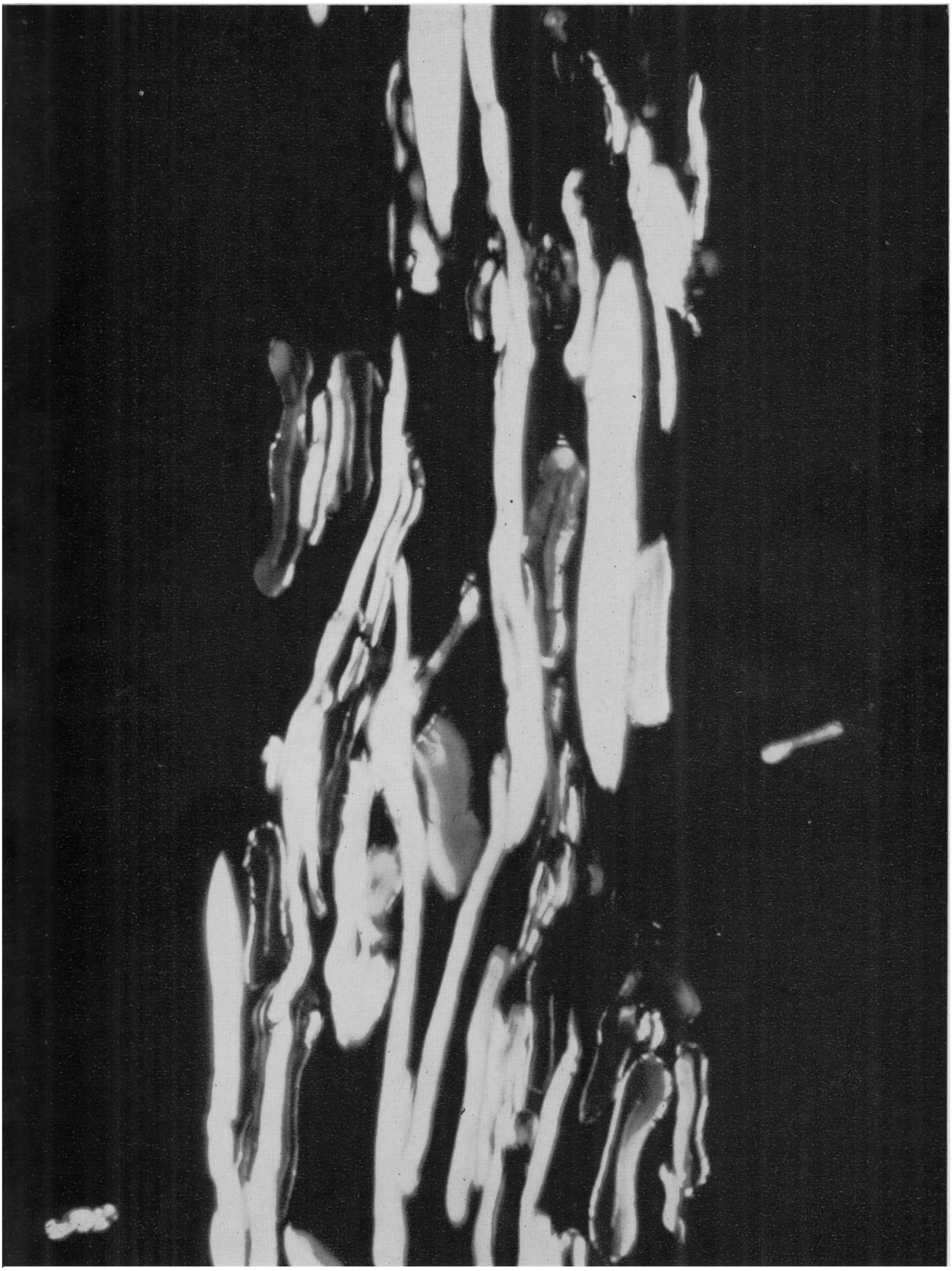

Fig. 10—Spruce sulphite handsheet, unbeaten, polarised transmitted light [× 770]

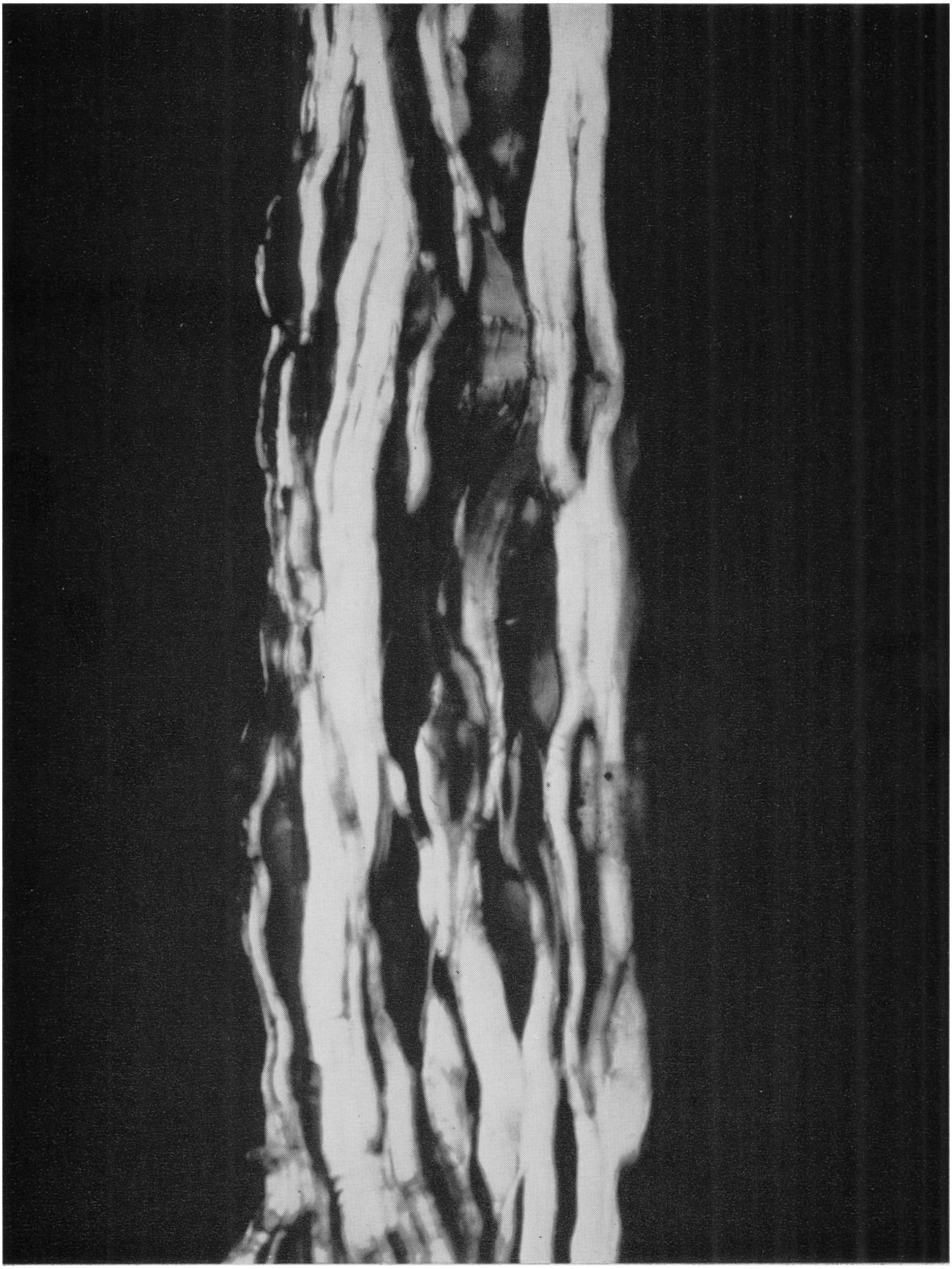

Fig. 11—Spruce sulphite handsheet, Valley beaten 45 min, polarised transmitted light [× 770]

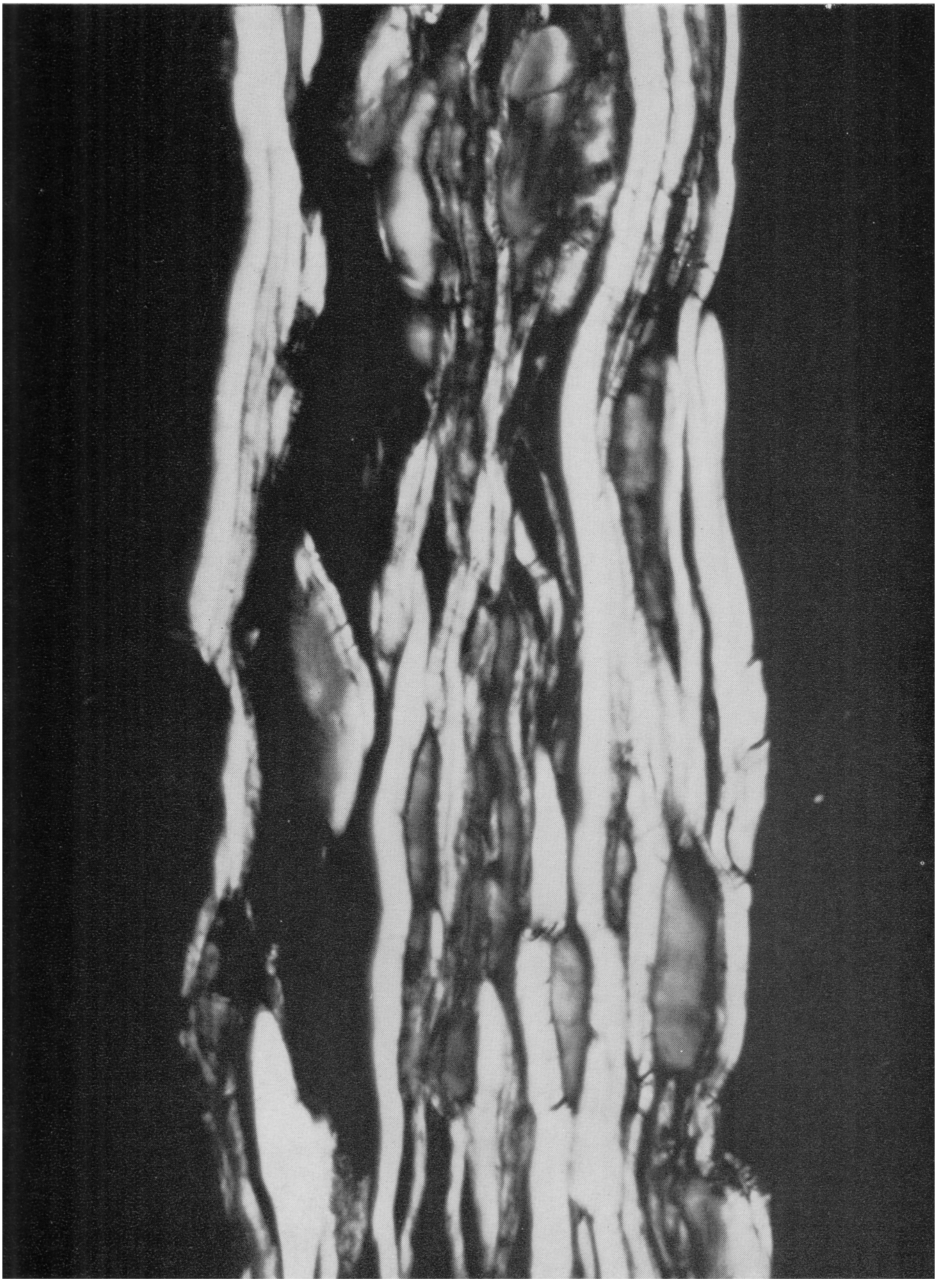

Fig. 12—Highly beaten sulphite sheet with tension applied to the sheet along the page, polarised transmitted light [× 770]

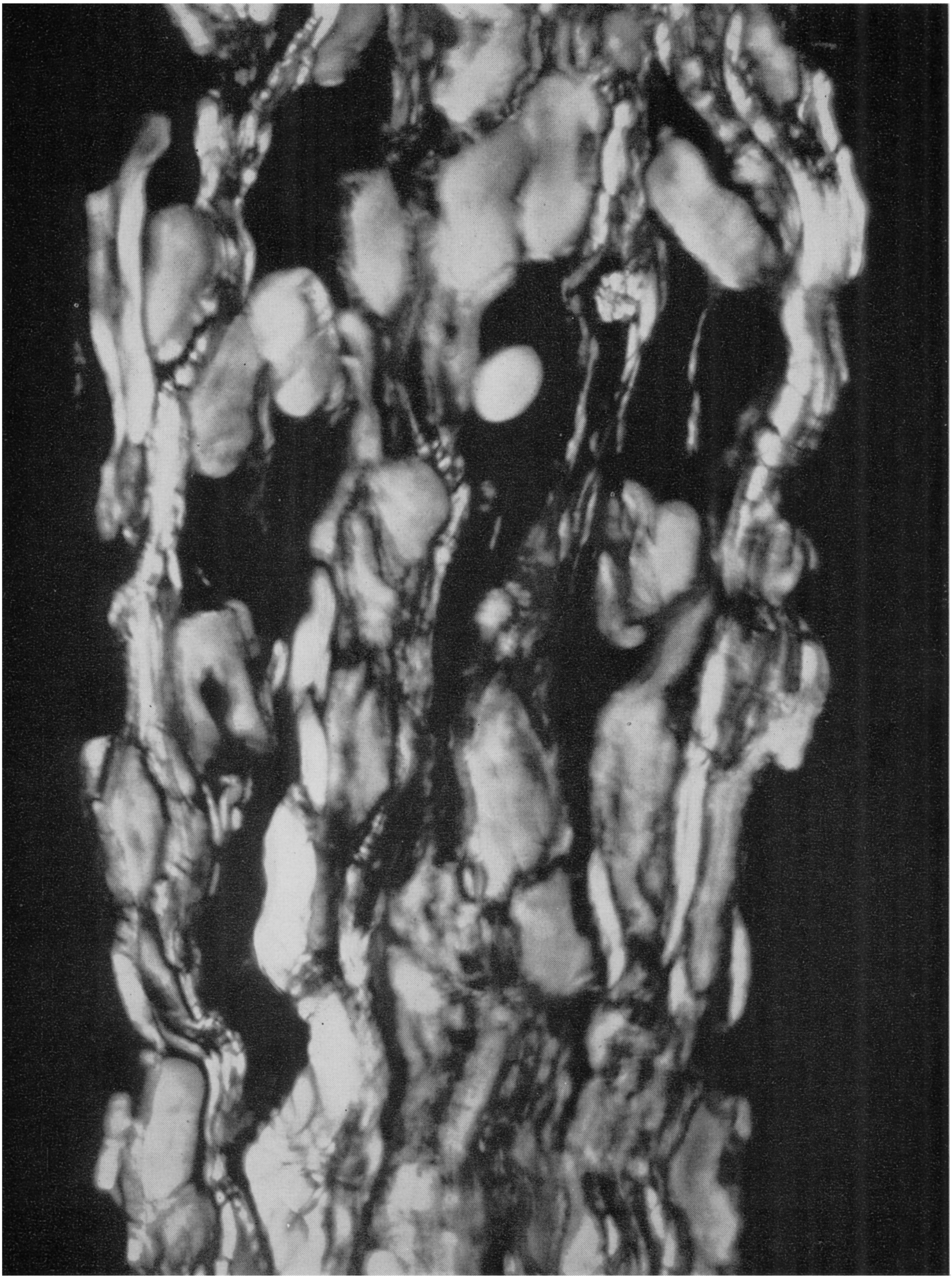

Fig. 13—Highly beaten sulphite sheet with tension applied normal to the plane of the page, polarised transmitted light [× 770]

Fig. 14—Melamine-impregnated paper, transmitted light [× 640]

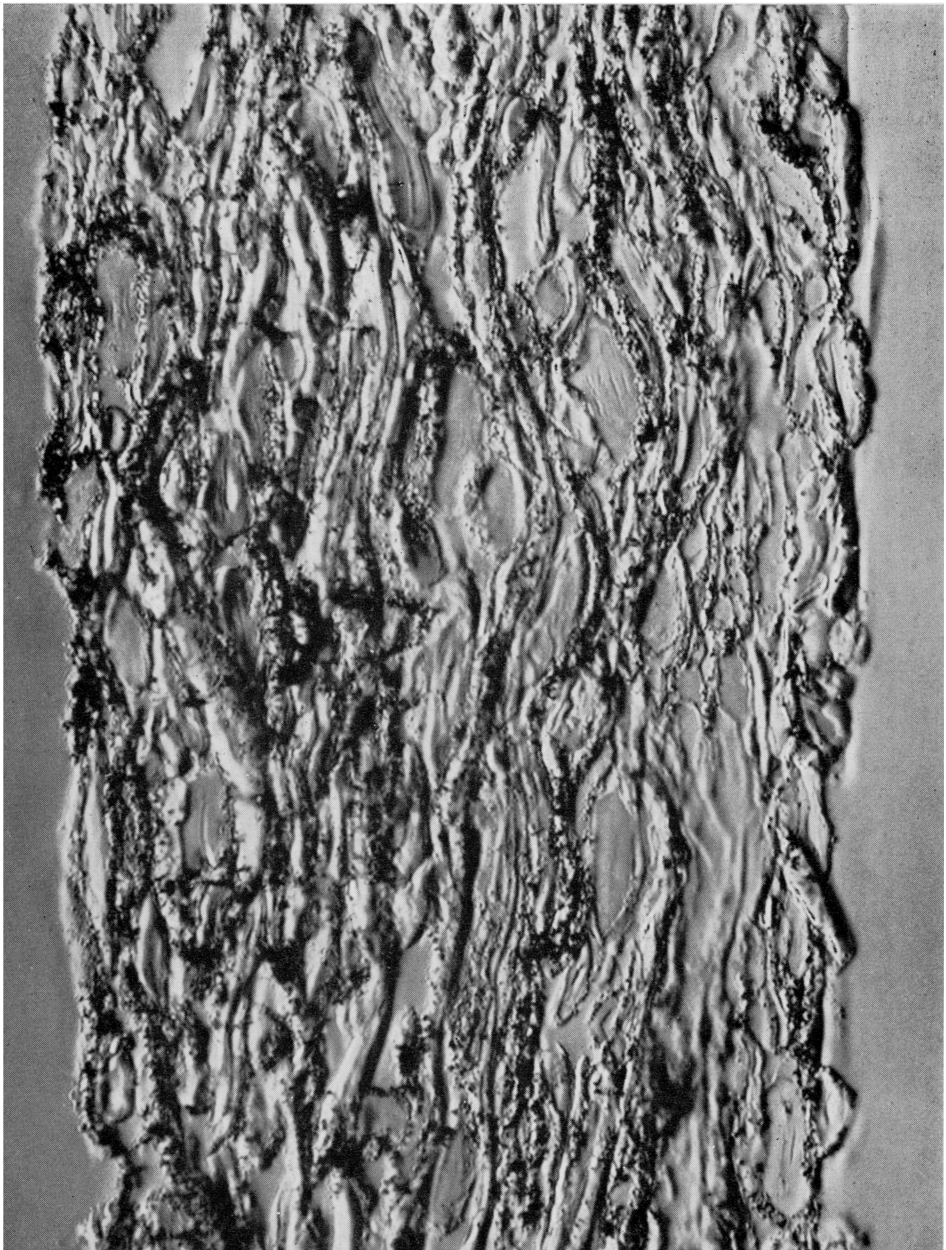

Fig. 15—Titanium dioxide filled paper, oblique transmitted light [× 770]

Fig. 16—Titanium dioxide filled paper, oblique transmitted light [× 770]

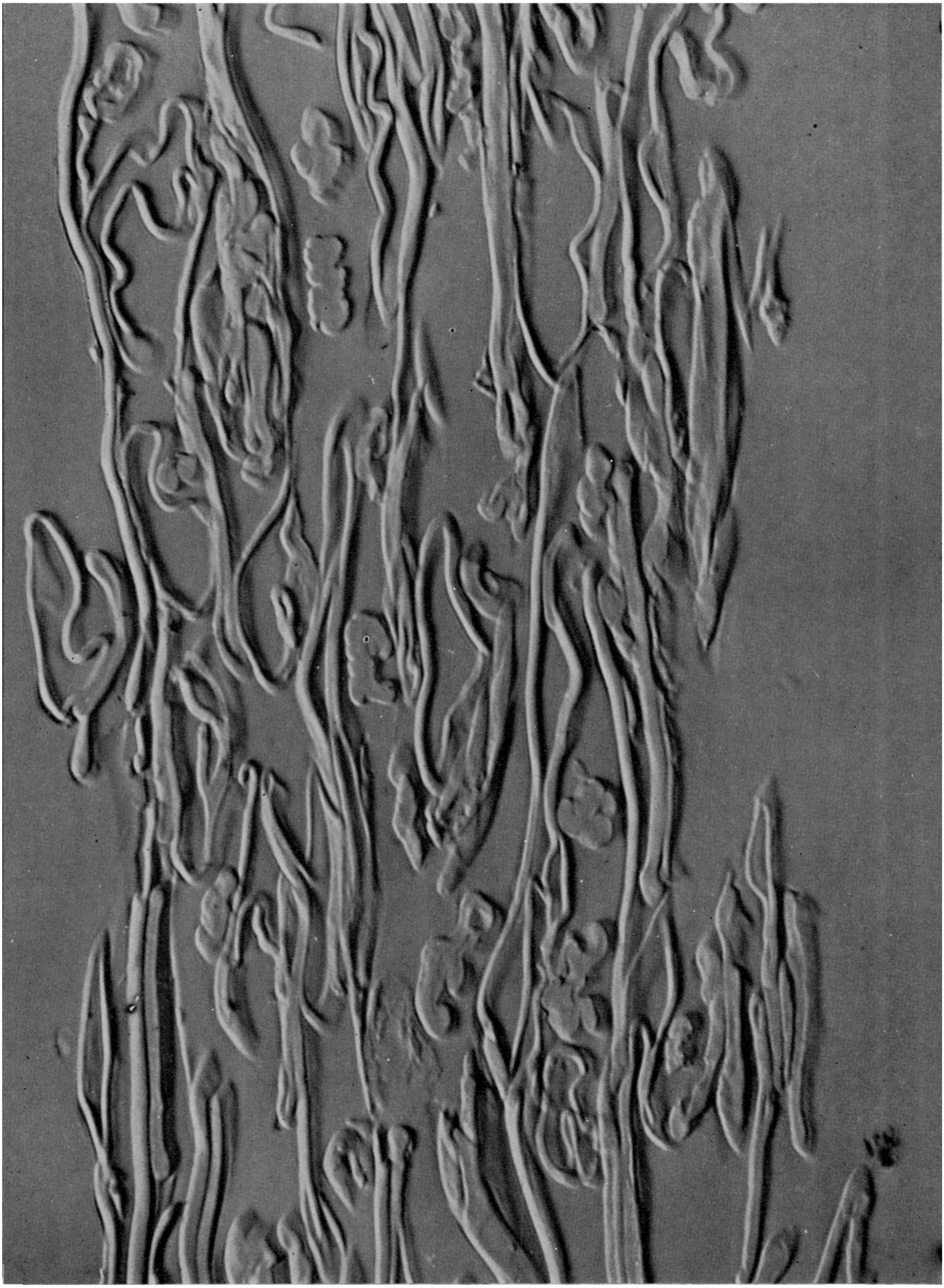

Fig. 17—Self-bonding rayon, oblique transmitted light [× 770]

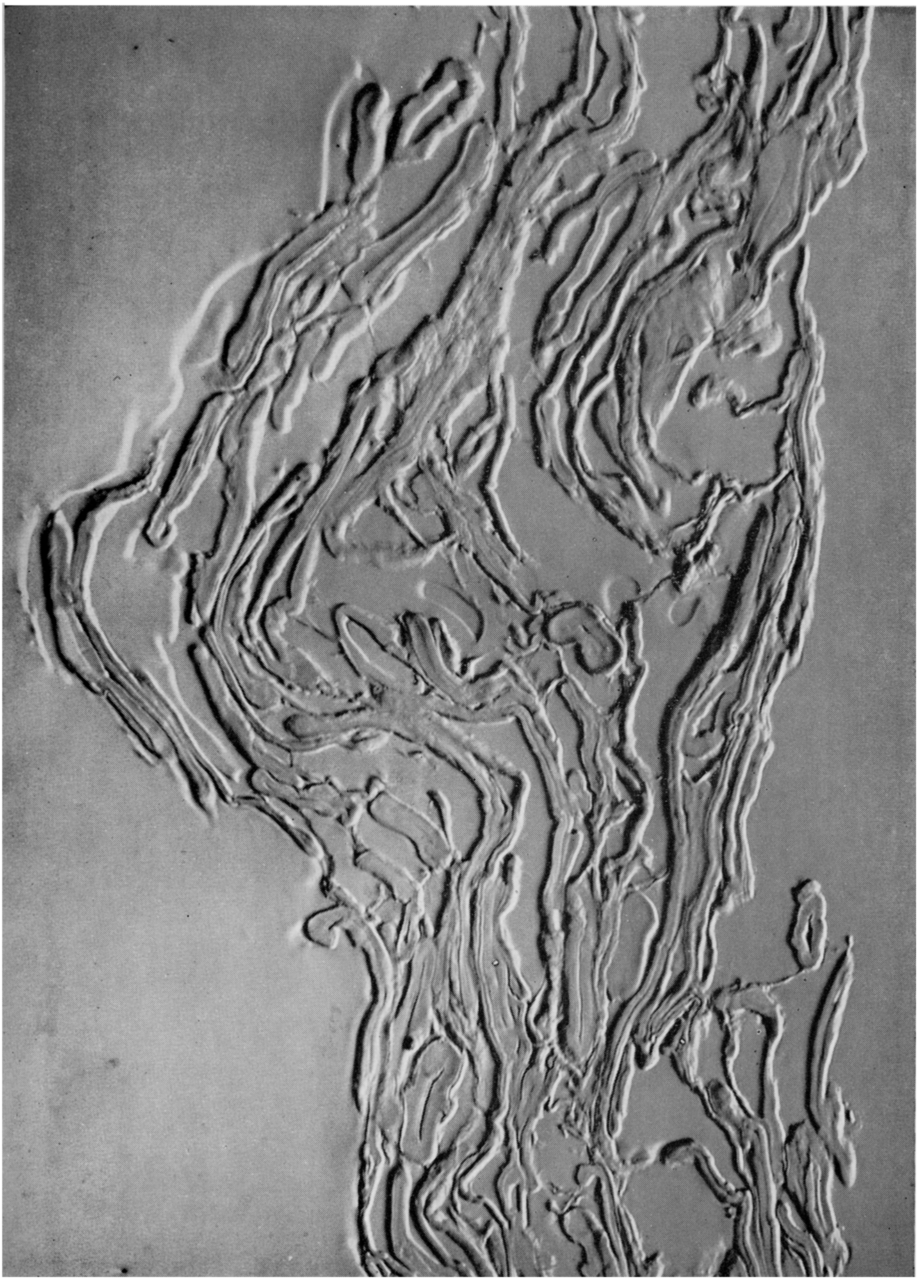

Fig. 18—Fine creped paper, oblique transmitted light [× 590]

Fig. 19—Fine creped paper, polarised transmitted light [× 590]

Softwood sulphite handsheets and effect of beating

Beating has clearly the major effect of reducing the bulk of the sheet, but other effects are also observed (Fig. 10 and 11). The cross-sectional shape of the fibre appears to change, all lumens being collapsed after beating. Furthermore, there appears to be a tendency for fibres to adopt a lenticular shape in cross-section. This probably arises from the effect of pressure on the plasticised fibre.

Softwood sulphite handsheet, effects of free and tension drying

The change in the structure of paper caused by tensions on the sheet during drying can be readily studied by the use of thin sections and the

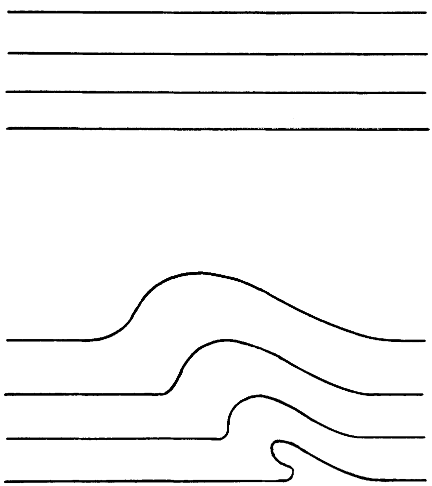

Fig. 20

results of an extensive study of this effect will form part of a later publication. Fig. 12 and 13 show micrographs of handsheets of paper formed from heavily beaten sulphite pulp and dried under appreciable tension in one direction while being allowed to shrink in the other, the extension in the two directions being +20 per cent and −30 per cent. The micrographs reveal the anisotropy of structure produced in this way. The preferred fibre orientation in the tensioning direction is clear from both micrographs. The compression of fibres in the cross-direction is apparent. It appears to be accommodated partly by the undulation of fibres, but there is also a second effect, namely, the local change in orientation of crystalline regions of the fibre (as revealed by

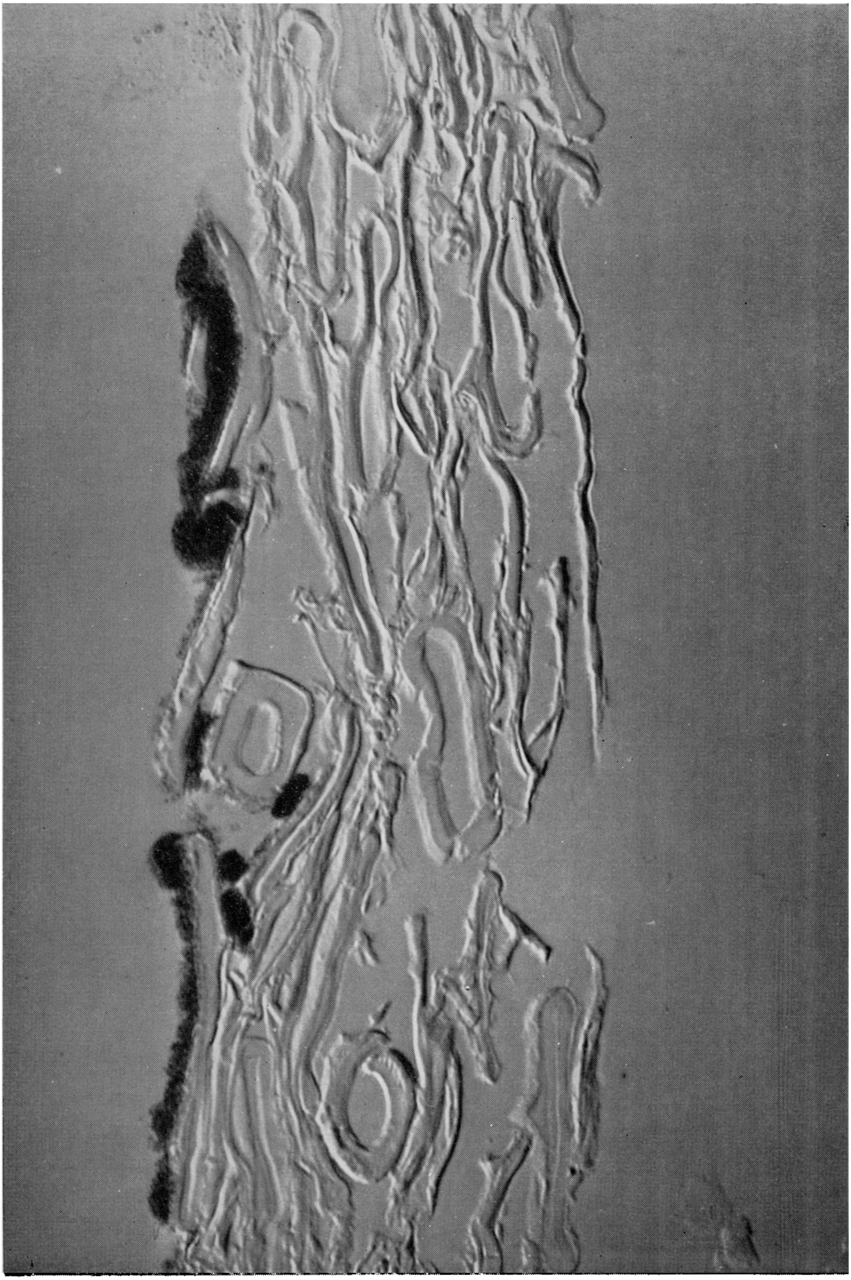

Fig. 21—Printed newsprint, oblique transmitted light [× 770]

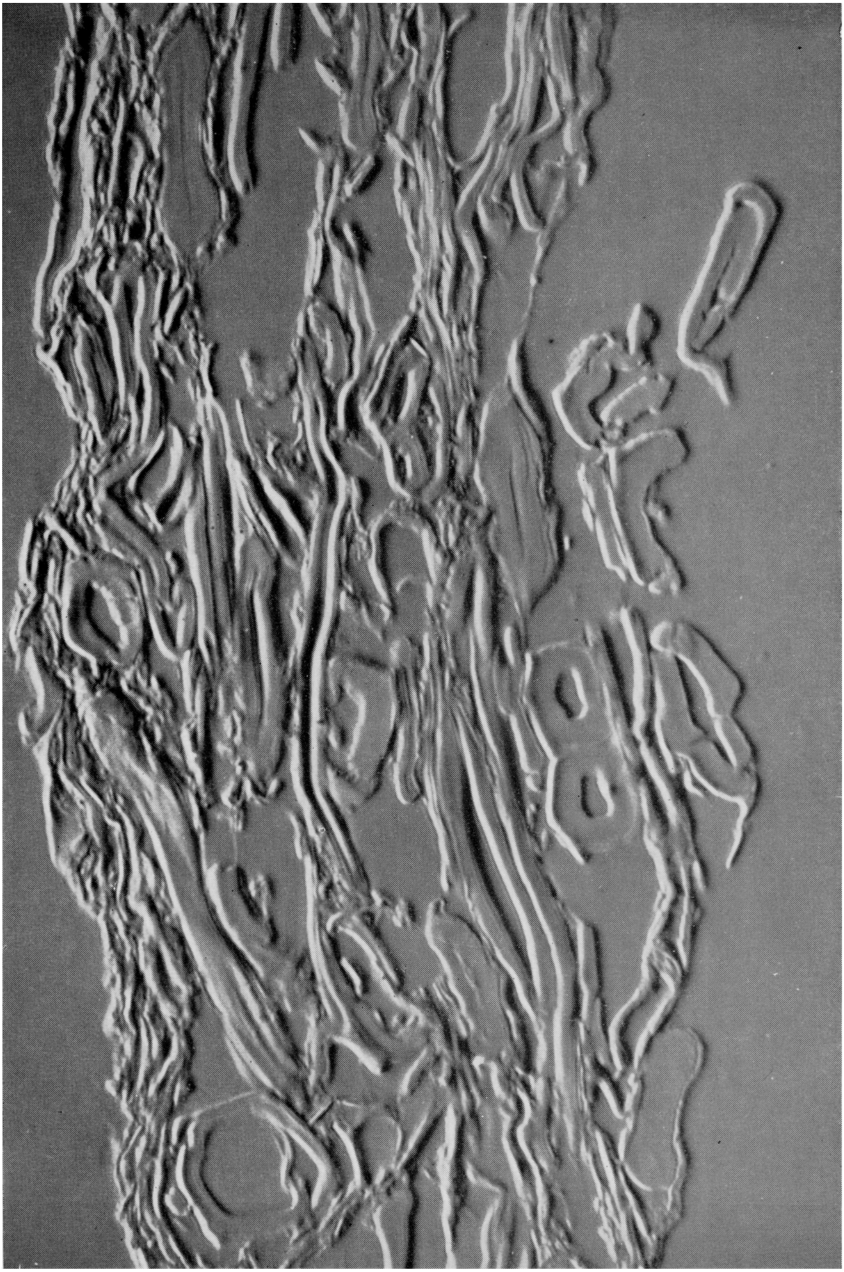

Fig. 22—Mechanical printing before supercalendering, oblique transmitted light [× 870]

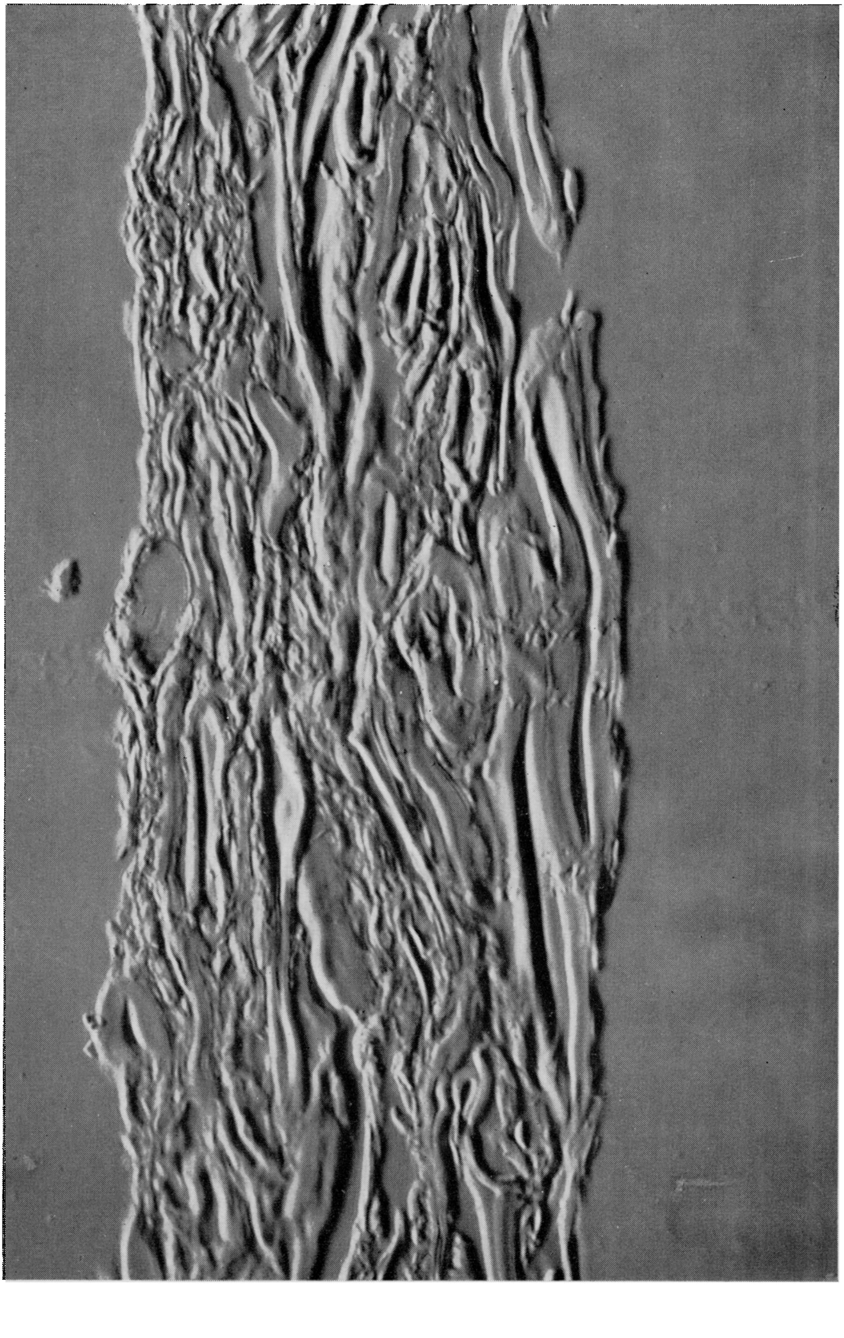

Fig. 23—Mechanical printing after supercalendering, oblique transmitted light [× 870]

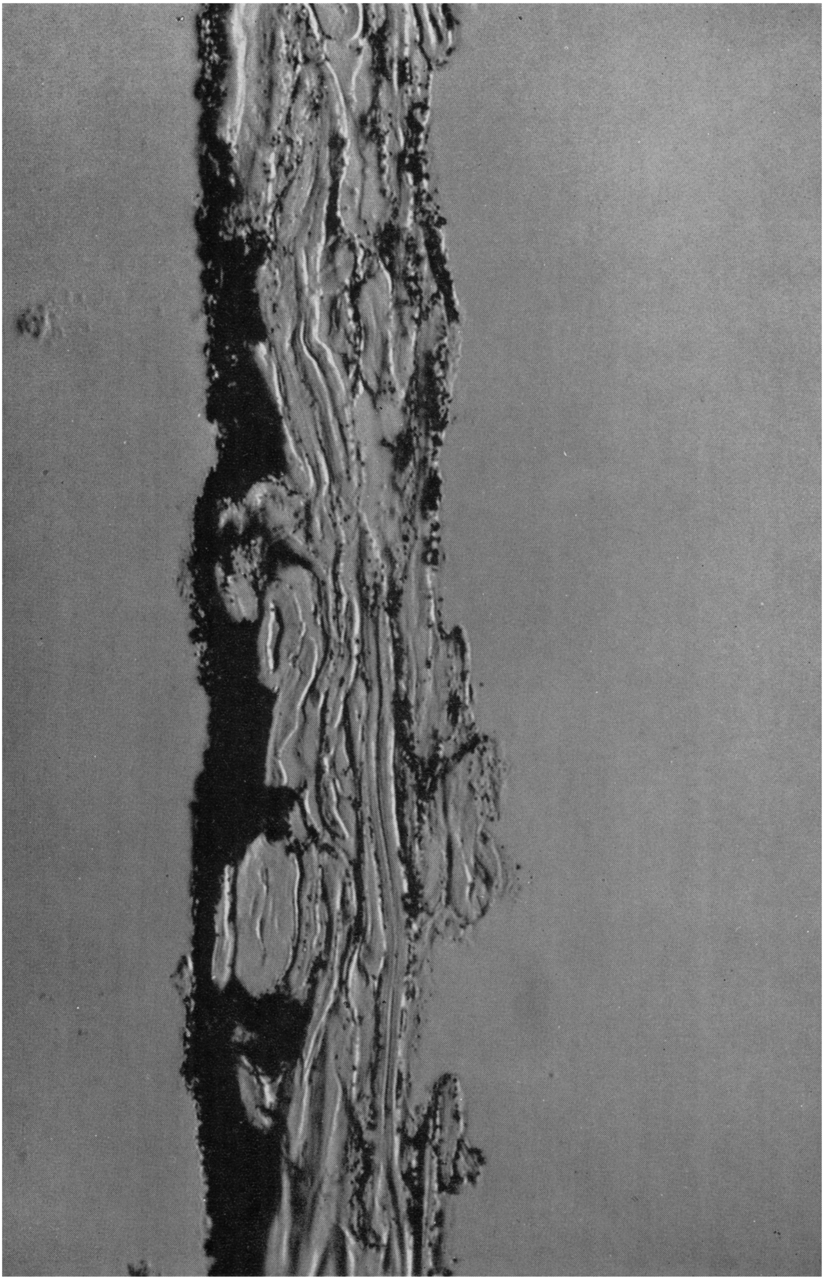

Fig. 24—Bread wrap, titanium dioxide filled coating, oblique transmitted light [× 675]

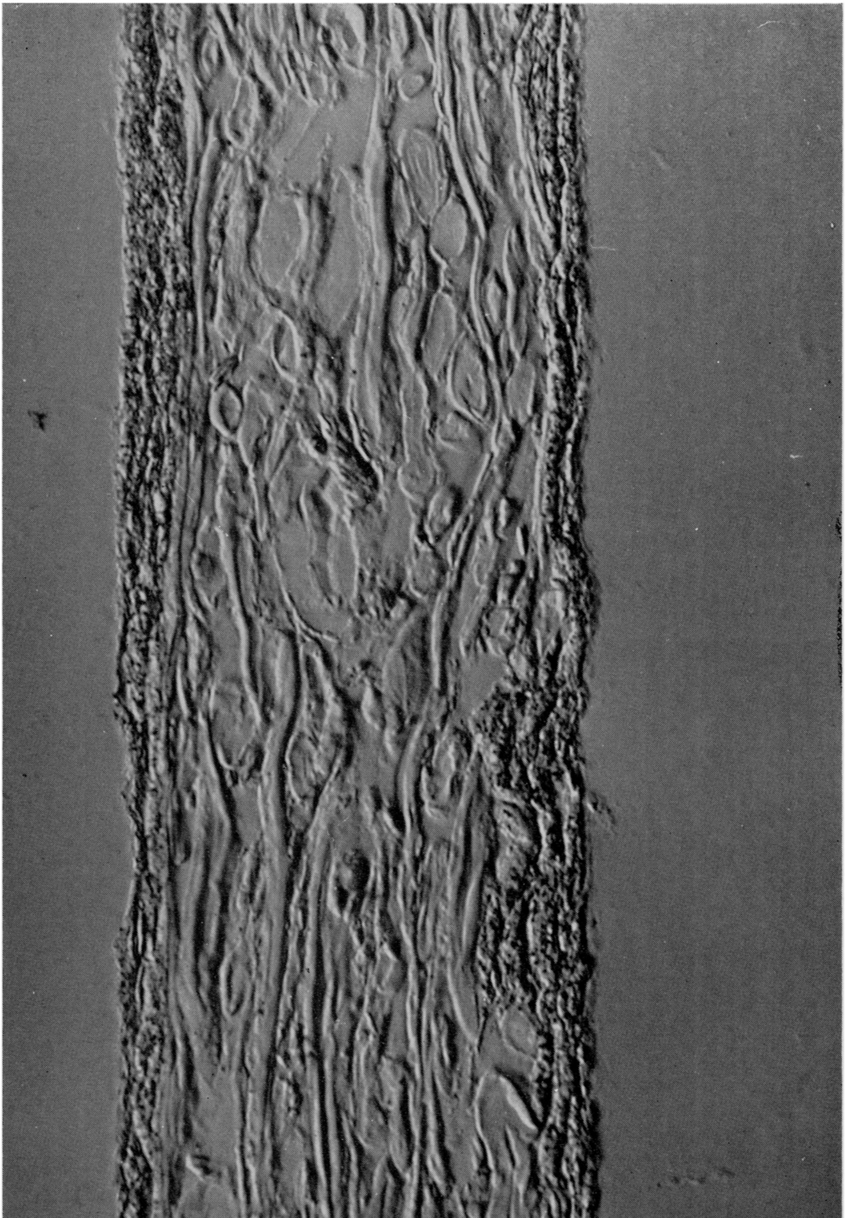

Fig. 25—Two-sided art, oblique transmitted light [× 770]

the polarising microscope). The relative significances of the two effects is being studied. It must be borne in mind that the transverse shrinkage of the paper in this case arises from two causes—the natural shrinkage of the paper and the contraction caused by the longitudinal tension at rightangles. The two effects may well produce dissimilar structures.

Melamine-impregnated paper

This heavily pigmented paper (Fig. 14) shows the presence of pigment granules both in the pores of the paper and occasionally in the lumens of the fibres. A fine acicular growth, presumably of melamine, can be seen on the surfaces of certain fibres, particularly in the regions of large voids.

Filled papers

The presence of fillers in papers can be detected in these sections (Fig. 15 and 16), but, owing to the similarity of refractive index between filler particles and the embedding medium, it is not usual for low refractive index fillers to be revealed with very great contrast or clarity. With titanium dioxide (which has a very high refractive index), each particle scatters light to an appreciable extent and it is possible to obtain an image that clearly reveals each individual particle.

Fig. 15 and 16 show two papers both titanium dioxide filled—Fig. 15 a paper with a considerable degree of two-sidedness and Fig. 16 a paper with a much more even distribution of filler.

Self-bonding rayon

A sheet of self-bonding rayon shows in Fig. 17 the presence of two fibre types—the thin-walled hollow fibre and the solid fibre with the convoluted outline characteristic of rayon.

The hollow fibre tends to collapse to a ribbon-like form in some cases, but, rather than collapse on to themselves, it appears that opposite walls of the same fibre can be drawn by surface tension forces on to other fibres. This presumably occurs because of the bulking effect of the solid fibre component on the structure of the sheet. This may well be what happens in paper made from slush pulps, whereas it may not occur in dried pulps in which the lumens are already collapsed.

Fine creped paper

The mechanism of creping is shown by Fig. 18 and 19. It appears that, on the lower face of the paper, compression consists of a severe local folding over of the surface layers and this folding extends over a larger region with less severity towards the upper layer. Delamination occurs within the creped region. Fig. 20 indicates the mode of deformation that is believed to occur.

Newsprint

The ink shows very little penetration into the structure of the low-grade newsprint in Fig. 21. The splits apparent in the cell walls are characteristic of mechanical pulps and are probably associated with the action of the grinder as suggested by Forgacs.[10]

Mechanical printing and effect of calendering

The effect of calendering on the structure of mechanical printing paper is clearly shown in Fig. 22 and 23. Not only does calendering reduce the inter-fibre void space, but it collapses the lumens of the mechanical wood fibres. Note the marked presence of the filler and fines on the top side of the sheet.

Coated bread wrap

This paper (Fig. 24) has been machine-glazed on the wire side and coated by the trailing blade method on that side using a titanium dioxide coating mix. The titanium dioxide filling in the paper stems from the use of coated broke.

Two-sided art paper

In this paper, which has a furnish of hardwood, softwood and esparto (Fig. 25), the cylindrical form of the esparto fibres is apparent. The coating consists substantially of clay, casein and latex, but with a small amount of titanium dioxide, which is apparent as dark particles in the coating layer.

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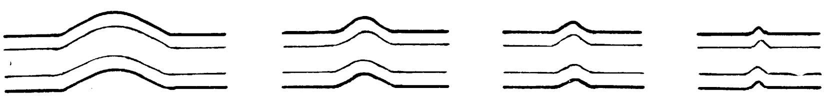
ADDENDUM 1—*The undulatory structure of fibres in paper*

In the article *The Theory of Shrinkage of Paper*, presented at the Fundamental Research Symposium in 1961, it was shown that fibres in paper that had been allowed to shrink became shorter by an amount exactly equal to the shrinkage of the paper and that no relative movement of fibres occurred at their crossing points. The term microcompression was adopted to describe the longitudinal compression of the fibres that was believed to take place at the bonded areas. With the availability of a technique that enabled high resolution micrographs of paper to be obtained in cross-section, it has become possible to examine in more detail the structural changes taking place due to shrinkage. The longitudinal shortening of fibres in shrunken sheets may clearly be accommodated by deformation modes that may be considered in terms of wavelength. This is made clear from Fig. 26. The wavelength over which undulations in the fibre occur can range from several fibre thicknesses to molecular dimensions. By the term microcompression, we wish to indicate that shortening of fibres is due substantially to deformations of wavelengths lower than that of the fibre width, for example, (*c*) and (*d*), rather than those that are greater, for example, (*a*).



(a) (b) (c) (d)

Fig. 26

The resolution of the micrographs now obtained is sufficient to deduce values of fibre shortening for the (*a*) and (*b*) types, though compressional deformations smaller than this would be lost. A factorial experiment was carried out on two woodpulps, a bleached sulphite and an unbleached sulphate both obtained in the form of dry pulp laps and disintegrated in a laboratory disintegrator. Each was beaten to three different degrees and wet pressed under three different levels of pressure. The B.P. & B.M.A. sheetmaking procedure was adopted up to the pressing stage, for which only one pressing at the appropriate pressure was applied. The sheets were dried either on a drying plate or by freely drying on a wire. The values of shrinkage of the paper sheets are tabulated overleaf. From each sheet of paper, a section was produced and a micrograph obtained from each section.

A replicate experiment was carried out, but was not completed, since it became apparent that adequate results were being obtained from the initial experiment. The shortening of fibres due to undulation was measured in two

TABLE 1—PERCENTAGE SHRINKAGE OF SHEETS

Beating pressure, lb/in²	*Bleached sulphite woodpulp beating time (Valley)*			*Unbleached sulphate woodpulp beating time (Valley)*		
	0 *min*	20 *min*	60 *min*	0 *min*	30 *min*	90 *min*
0	1.4	4.7	15.5	0.6	2.5	8.1
20	1.25	4.6	14.1	0.8	2.6	8.1
100	1.0	3.7	11.5	0.3	2.2	8.1

ways. Firstly, it was done by superimposing a grid over a micrograph and measuring at each grid bar along the length of the fibre the angle that the fibre made with the bar. The shortening of the fibre was then computed from this angle. The grid spacing in this case was 13 μ (referred to the original specimen), but a check was made that reducing the spacing to 6.5 μ did not make a significant difference. Secondly, the undulatory shortening was obtained from a large projection of the micrograph using an opisometer to measure the curved length. This gave shortening values significantly, but not appreciably greater than the first method. Values obtained from the first method on all fibres in a section are given in Table 2.

It is immediately apparent that these values of undulatory shortening are completely inadequate to explain the shrinkage of paper and that the suggestion of microcompressions of a lower order of undulation is confirmed. An

TABLE 2—PERCENTAGE SHORTENING OF FIBRES BY UNDULATION

Beating pressure, lb/in²	*Bleached sulphite woodpulp beating time (Valley)*			*Unbleached sulphate woodpulp beating time (Valley)*		
	0 *min*	20 *min*	60 *min*	0 *min*	30 *min*	90 *min*
Plate-dried sheets						
0	1.6	1.6	1.4	1.4	1.4	1.6
20	1.3	1.2	1.3	1.2	1.3	1.2
100	0.9	0.7	0.1	1.9	2.3	1.4
Freely dried sheets						
0	1.8	2.0	1.7	2.0	1.8	2.9
20	2.0	1.7	3.3	2.8	1.8	1.5
100	2.0	1.8	2.3	1.6	1.4	2.3

analysis of variance on these results indicates that the only major effect on undulatory shortening is whether the sheet was plate-dried or freely dried. Beating and pressing surprisingly have no effects, although there is a barely significant effect of pressing for the tension-dried sheets. The results have therefore been compounded in Table 3.

Further experiments were carried out on two handsheets that had been dried under uniaxial tension, the results for shrinkage being shown in Table 4.

The corresponding undulatory shortening of the fibres in these sheets was as indicated in Table 5.

TABLE 3—PERCENTAGE SHORTENING OF FIBRES BY UNDULATION

Drying	*Mean of all beating times and pressing pressures*	
	Bleached sulphite	*Unbleached sulphate*
Plate-dried	1.3	1.5
Freely dried	1.8	2.0

TABLE 4—SHRINKAGE OF 60 MIN VALLEY BEATEN BLEACHED SULPHITE WOODPULP DRIED UNDER UNIAXIAL TENSION

Drying direction	*Dried under high tension*	*Dried under low tension*
Tensioning direction	−24 per cent*	+10 per cent
Cross-direction	+30 per cent	+15 per cent

*The minus sign indicates extension in the tensioning direction

TABLE 5—PERCENTAGE SHORTENING OF FIBRES BY UNDULATION

Drying direction	*Dried under high tension*	*Dried under low tension*
Tensioning direction	1.3 per cent	2.9 per cent
Cross-direction	3.5 per cent	1.5 per cent

It must be noted here that this very large value of shrinkage in the cross-direction of the sheet dried under high tension (namely, 30 per cent) arises from two causes—firstly, the natural shrinkage of this highly beaten paper and, secondly, the compressive force in the cross-direction of the sheet arising from the tensioning stress at rightangles. It will be seen that, even in the case of this very high shrinkage, only a small proportion can be explained in terms of undulatory shortening.

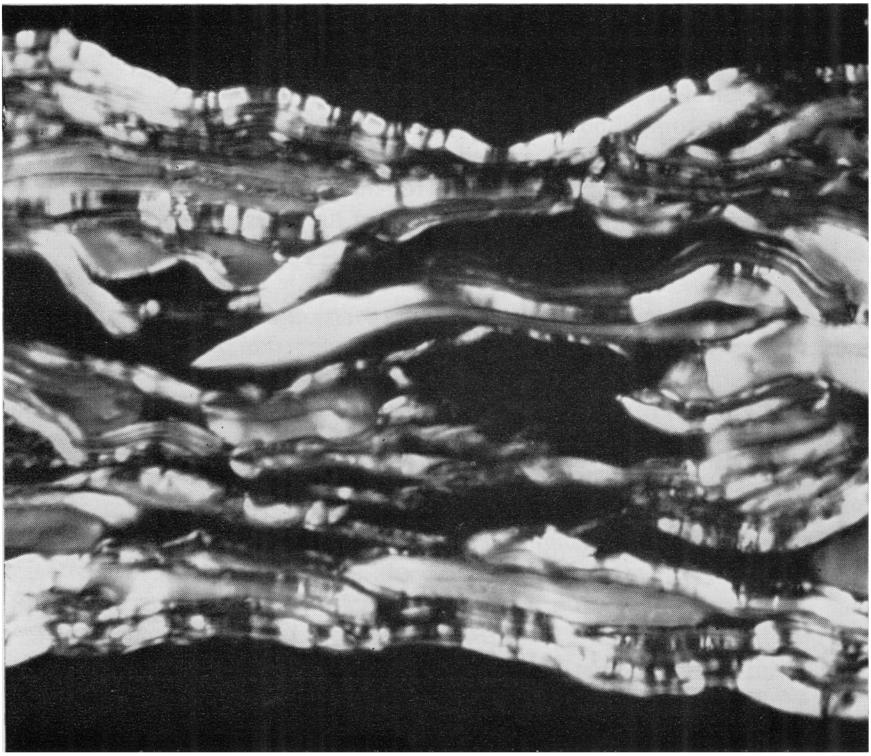


Fig. 27

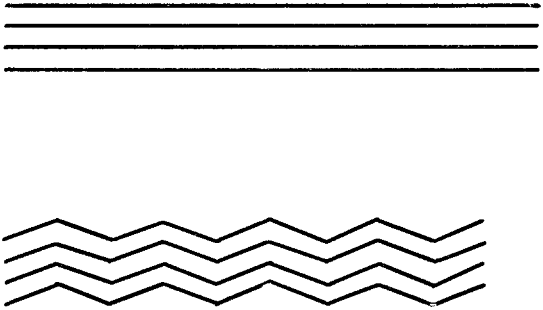


Fig. 28

It is clear then from these results that the undulation of fibres cannot contribute more than a fraction of the shrinkage of paper when the shrinkage is appreciable and that the term microcompression, although it is not intended to describe the actual molecular rearrangement that has taken place, is a reasonable term for the effect of paper shrinkage. Evidence for the compression of the structure of the fibre wall has been sought using polarised light and indeed it is quite apparent that sections of fibres do show a disarrangement of the fibrillar orientation on a small scale. Fig. 27 shows this effect. The alternating light and dark bands along the length of many of the fibres must be interpreted as changes in direction of the fibrils as indicated schematically in Fig. 28. This effect is observable, however, both in tension-dried and freely dried papers and it is not possible to conclude that it is present to a greater extent in a freely dried sheet. The sectioning procedure itself may disarrange the fibrillar orientation sufficiently to mask such an effect, if it exists. It must always be borne in mind that microcompressions may be on an even smaller scale still—namely, the molecular scale—and that no techniques of light or even electron microscopy may be capable of revealing them.

ADDENDUM 2—*Collapse of cell lumens in paper*

It is possible using the thin sectioning technique to obtain quantitative data on the proportion of fibres in a sheet that have their lumens collapsed. For the purpose of this work, a fibre is considered collapsed when the inner walls of the fibre are touching along more than half the length of the lumen in the fibre section.

TABLE 6—PERCENTAGE OF UNCOLLAPSED FIBRES IN SECTIONS OF PAPER

Plate-dried and freely dried data combined

Wet pressure, lb/in²	*Bleached sulphite woodpulp beating time (Valley)*			*Unbleached kraft woodpulp beating time (Valley)*		
	0 *min*	20 *min*	60 *min*	0 *min*	30 *min*	90 *min*
0	7.8	2.4	2.9	14.5	14.9	4.3
20	8.1	2.5	3.8	13.0	12.7	1.9
100	1.3	0	0	9.6	9.2	4.2

The results obtained from an analysis of the micrographs of the previous work are given in Table 6.

An analysis of variance of the data (after an arc sine transformation) reveals significant effects of beating and pressing for the kraft pulp and of beating for the sulphite pulp. The number of sulphite pulp fibres uncollapsed is very low and a real effect of pressing may have been missed.

Three conclusions may evidently be drawn. The unbleached sulphate pulp differs from the bleached sulphite pulp in showing less collapse, approximately 14 per cent of fibres in the sulphate pulp being uncollapsed in the unbeaten unpressed state compared with 8 per cent for the sulphite pulp. Beating reduces the number of uncollapsed fibres in the sheet until in the highly beaten state perhaps only 1 or 2 per cent are uncollapsed. Wet pressing also increases the proportion of collapsed fibres, certainly for the kraft pulp. Collapse does not appear to be affected by free or plate drying. These findings agree well with the established concepts of the increase in plasticity of the cell wall with beating.

The large degree of collapse observed in these sheets even in the unbeaten state was found to agree with values obtained directly from the dry pulp lap and it would appear that most of the collapse has arisen from pressure during the lap forming process. To avoid this complicating factor, some work was carried out on standard handsheets made from sulphate-pulped *Pinus radiata* that had not been dried before sheet formation. This gave much larger values for the number of uncollapsed fibres, 134 out of 350 (or 38.3 per cent) being uncollapsed. The sheet was disintegrated and reformed under the same conditions, when 62 out of 291 fibres (or 21.3 per cent) were found to be collapsed. A second sheet that had been formed once only was disintegrated and reformed, *but without applying pressure*. This gave 117 out of 327 fibres uncollapsed (or 35.7 per cent) and this is not significantly different from the first result.

It would appear from these experiments that the main factor controlling the degree of collapse in twice-dried pulps is not the second drying itself, but the wet pressure that has been applied during the second formation of the sheet. The hypothesis put forward to explain this result is that, during the first pressing of the sheet, fibres are collapsed at regions where they bear the load and, except for very thin-walled cells, they remain open between these regions. (The regions will correspond to fibre crossings especially in local areas of high sheet density.) During the second sheet formation and pressing, previously uncollapsed parts of fibres will become collapsed when they happen to occur in load-bearing regions. Previously collapsed parts of the fibre either remain collapsed during this process or, if they reopen on wetting, they collapse again on subsequent drying. This hypothesis would predict as an approximation that the proportion of uncollapsed fibre after the second pressing should be equal to the square of the proportion after the first and this is indeed roughly true. This hypothesis would further suggest that fibres are not either collapsed or uncollapsed down their entire length, but rather go through regions of collapse down their lengths, depending on the local pressure that they have experienced. The preferential collapse of fibres at regions where they have evidently borne the load during wet pressing has been observed in serial sections and this is illustrated in Fig. 29.

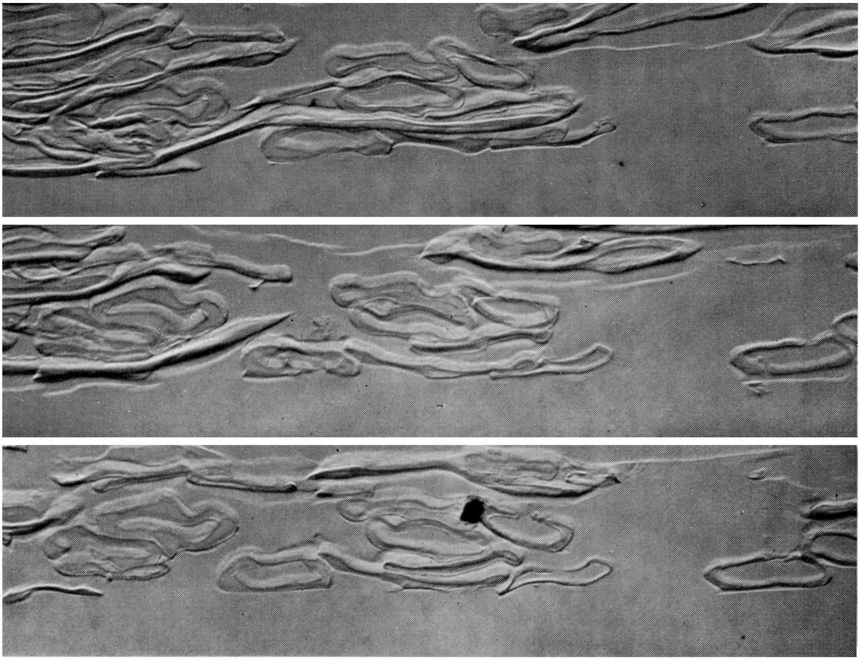


Fig. 29

This work raises the interesting possibility that, contrary to general opinion, the strongest papers may not be those that are made from collapsed ribbon-like fibres. A greater interfibre bonded area can be produced for the same sheet density, if the fibres are initially uncollapsed and collapse only at the regions of high pressure. At regions of low pressure, the fibres will then be better able to contact each other in their uncollapsed state. This effect may be in part responsible for the losses in pulp strengths on drying.

Discussion

Mr H. W. Emerton—It probably sounds like a statement of the obvious to say there have not been many scientists of eminence who were blind. We have had deaf musicians and artists who were armless, perhaps, but not blind scientists. I think the reason for this is that, if one wishes to examine or describe a material or a process, often the very first thing one does (or should do) is to look at it and, if the process or material is beyond the limit of resolution of the unaided eye or proceeds too quickly or too slowly, then one has recourse to various aids—the light microscope, the conventional transmission electron microscope, the powerful scanning electron microscope, the high-speed camera, the ciné camera and time-lapse photography. In the past, I think we have been rather slow in our industry in exploiting these methods and to me one of the significant things about the Oxford meeting was that more than a quarter of the papers based their results on work that employed such direct methods. These should, of course, whenever possible, be linked to careful and meaningful measurements and this has been characteristic of this paper and indeed most of the work carried out by Page and his colleagues.

The importance of this type of work has been recognised in the planning of this symposium and we have drawn together several papers using direct observational methods into one session. We have heard this morning from Radvan, who used direct observation in the work he reported and I have no doubt that there will be several other examples during the week.

There are two points that I would like to take up with Page. The first concerns his Addendum 1 and his reference to undulation: in Table 1, he quotes figures from which he concludes that as beating proceeds the amount of undulation in fibres is unaffected. We are dealing here with undulation in what in a sheet of paper we call the *z*-direction. At Oxford, micrographs were shown in which the surface of the paper was observed and one of the conclusions there was that, so far as fibres lying near the surface were concerned and looking not at a plane containing the *z*-direction, but one at rightangles to this, the tendency as beating proceeds is for the sinuosities and kinks to straighten out. I do not know whether Page would think these two observations—that with beating the fibres show no increased undulation in the *z*-direction and that they tend to straighten out in a plane at rightangles to this—should be linked.

My other comment is on Addendum 2, Table 6, in which it is shown that for unbleached kraft pulp, in sharp contrast to the bleached sulphite pulp, the first 30 min of beating at each of the three pressures has little effect on the proportion of fibres that have collapsed. It is believed by many that one of the most immediate effects in the early stages of the beating is to make the fibres more flexible. This is not revealed by the results for kraft fibres reported here and perhaps Page or somebody else here would like to comment on this.

Finally, may I just reiterate my belief in the importance of this kind of work, particularly when, as I say, it is linked with careful and meaningful measurement.

Mr D. H. Page—We should not pay too much attention to those percentages. They were worked out from extremely small numbers of fibres and, although one can draw conclusions from an analysis of variance of the data, it is dangerous to draw conclusions from individual values.

Dr H. F. Rance—A very small point on semantics. I wonder if we should not reconsider our use of the word collapse. To my mind, collapse implies a self-induced effect, whereas some of these examples involve a squeezing up or crushing of the fibre tube. We ought to use different words to describe surface tension collapse on the one hand and crushing by application of external force on the other hand.

Mr Page—I should emphasise that both things happen; perhaps Pye will have something to say about this later on.

Dr E. L. Back—If we look in these photomicrographs at the space between the fibres, we find a considerable pore anisotropy. There are many more bottlenecks for flow at rightangles to the sheet than in the plane of the sheet. This pore anisotropy can be evaluated separately by capillary flow measurements.* We have recently measured the effective pore radius according to the Lucas-Washburn equation for a number of papers and found the pore radius to be between 5 and 50 times larger in the plane of the sheet than at rightangles to it. With the powerful equipment developed by Page and his co-workers, someone has to analyse the space between the fibres.

Mr Page—One point I did not make in that connection is that we are now

* Osterberg, L., Brauns, O. and Back, E. L., *Svensk Papperstidn.*, 1960, **63** (19), 658–664

able to define the complete cellulose/air interface throughout the entire sheet. This gives the possibility of looking at the whole problem of porosity and even of the scattering of light in a new way. One could use Monte Carlo methods on the structure defined by serial sections to get a more complete understanding of the two factors affecting porosity and light scattering.

Dr O. J. Kallmes—I think this point will add to Page's derivation. He maintains that large undulations are not of importance to paper shrinkage. By the same token, the large undulations cannot explain the stretch of paper as demonstrated by the following example. For a large and a small undulation that in fact forms a microcompression, assuming that it is part of a circle, the ratio of its circumferential length to the distance between its ends is $r\theta/(\sin\theta/z)$. For small values of θ, this ratio approaches unity. For large undulations, the angle is relatively small, hence the ratio is almost unity; for small undulations, the radius is small, but the angle is very large, so the ratio is much greater than unity. Thus, on stretching a microcompression, its length can be increased appreciably.