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A NEW THEORY OF THE SHRINKAGE, STRUCTURE AND PROPERTIES OF PAPER

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Synopsis

The mechanism of the shrinkage of paper has been investigated microscopically and this has resulted in a better understanding of the structure of paper. It is shown that the shrinkage of paper gives rise to a 'microstructure', in terms of which rheological and other properties of paper, as well as the influence of papermaking variables upon them, can be explained. Although the theory is considered to be comprehensive, it has not been possible to develop it completely in this initial publication.

Une nouvelle théorie de la structure et des propriétés du papier en termes de son retrait

On a étudié au microscope le mécanisme du retrait du papier. Grâce à cet examen, on a acquis une meilleure connaissance de la structure du papier. Il a été démontré que le retrait du papier donne lieu à une 'microstructure', d'après laquelle on explique les propriétés du papier, rhéologiques et autres et l'influence sur celles-ci des variables de la fabrication. Bien qu'elle soit considerée comme couvrant un vaste terrain, cette théorie n'a pu être complètement dévelopée dans cette première communication.

Eine neue Theorie über Schrumpfung, Struktur und Eigenschaften von Papier

Der Mechanismus der Schrumpfung des Papiers wurde mikroskopisch untersucht und damit ein besseres Verständnis der Papierstruktur erreicht. Es zeigte sich, dass die Schrumpfung des Papiers zu *einer 'Mikrostruktur' führt, mit der man die rheologischen und anderen Eigenschaften des Papiers und den Einfluss der verschiedenen bei der Blattbildung auftretenden Variablen erklären kann. Obwohl die Theorie glaubhaft erscheint, war es nicht möglich, sie bis zu der vorliegenden ersten Veröffentlichung vollständig zu entwickeln.*

Introduction

FOR many years, considerable research effort has been devoted to the investigation of the physical properties of paper and the papermaking factors that affect them. All scientists concerned with the properties of materials recognise the need for an explanation of these properties in terms of basic structure and, for the paper industry, it is desirable that this approach be used to explain the physical properties of paper and the complex manner in which they are changed by papermaking processes. Little attention has been given to this fundamental problem (a valuable introduction to which was given by Van den Akker as early as 1950(1)) and this has partly been due to the non-existence of suitable techniques for the examination of the structure of paper and the measurement of the properties of its components. Interest in this approach has recently intensified both theoretically(2) and experimentally by the work on individual fibre rheology(3, 4) and the work on the properties(5–8) and individual strengths of fibre bonding.(4, 9)

The concept of paper now commonly accepted is outlined by the following model. Paper is a network of fibres adhering at their regions of contact. The fibres are limited to an approximately planar orientation: within this plane, their position and orientation have a degree of randomness arising from the disordered state of the suspension from which the sheet is formed. Moreover, in the majority of cases, the fibres are themselves flat, consequently the regions of adhesion lie in the plane of the paper. This model is illustrated by Fig. 1. This structure may be completely open in the case of freely beaten sheets or it may be filled with a matrix of fine material in the extreme case of a highly beaten sheet; generally, the structure will be partially filled with fibrillation.

Recent work(7) has shown that much more emphasis should be given in this model to the regions of contact between fibres. These have been shown to consist of extremely large areas within which adhesive forces must surely operate. Furthermore, they occur very frequently along the fibres, giving a more closely bonded structure than has hitherto been realised. This model is still insufficient and even appears to make it more difficult to explain the effects of some of the papermaking variables. For example, attention has already been drawn to the fact(7) that the large influence of drying constraint upon paper properties cannot be explained by the changes in the size and distribution of fibre-to-fibre bonds. The above model must be deficient in at least one major respect and, at an early stage in this work, it was considered that this was the assumption that fibre segments between bonds were straight. The authors then subscribed to Steenberg's original suggestion(10) that 'the fibres may be creased between the joining points'. It would appear that these micro-crepes, if they exist, are formed as a result of the shrinkage during the drying process and it was therefore decided to make an examination of the structural changes taking place in the sheet while drying, in particular during the shrinkage phase.

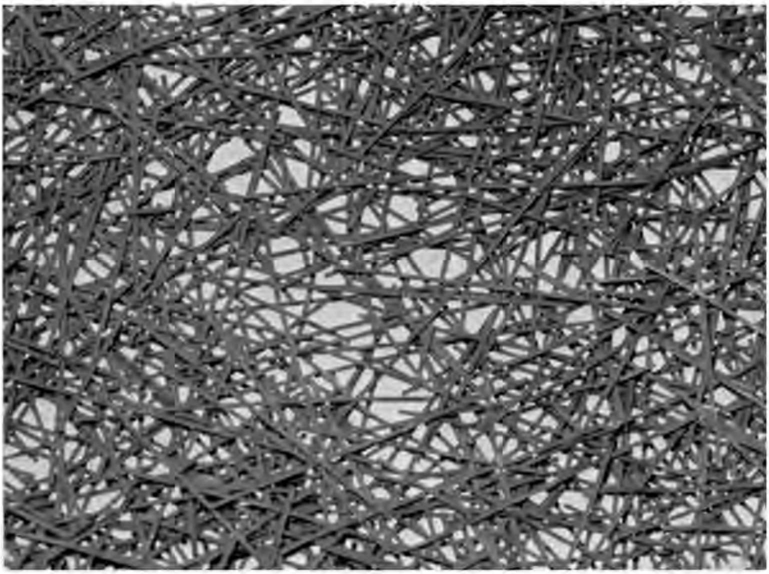

Fig. 1—This model was made by random deposition of strips of board 140 mm × 2·5 mm × 0·33 mm and is an ideal representation at a magnification of about × 70 of a 15 g/m² handsheet

Shrinkage

THE basis of an understanding of sheet shrinkage must originate from the known shrinkage behaviour of individual fibres. It is generally accepted that, owing to the anisotropic structure of fibres, the transverse shrinkage during drying can be very high, perhaps in the region of 20–30 per cent, whereas the longitudinal shrinkage is very small, of the order of 1–2 per cent. It should be observed that, if one considers a network of members fixed at the centre points of their crossings, the shrinkage of the network should be independent of the transverse shrinkage of the members, but equal to their longitudinal shrinkage. This is illustrated by Fig. 2. The anomaly that the shrinkage of paper can be an order of magnitude greater than the natural longitudinal shrinkage of the fibres implies that, during shrinkage, either a relative sliding movement

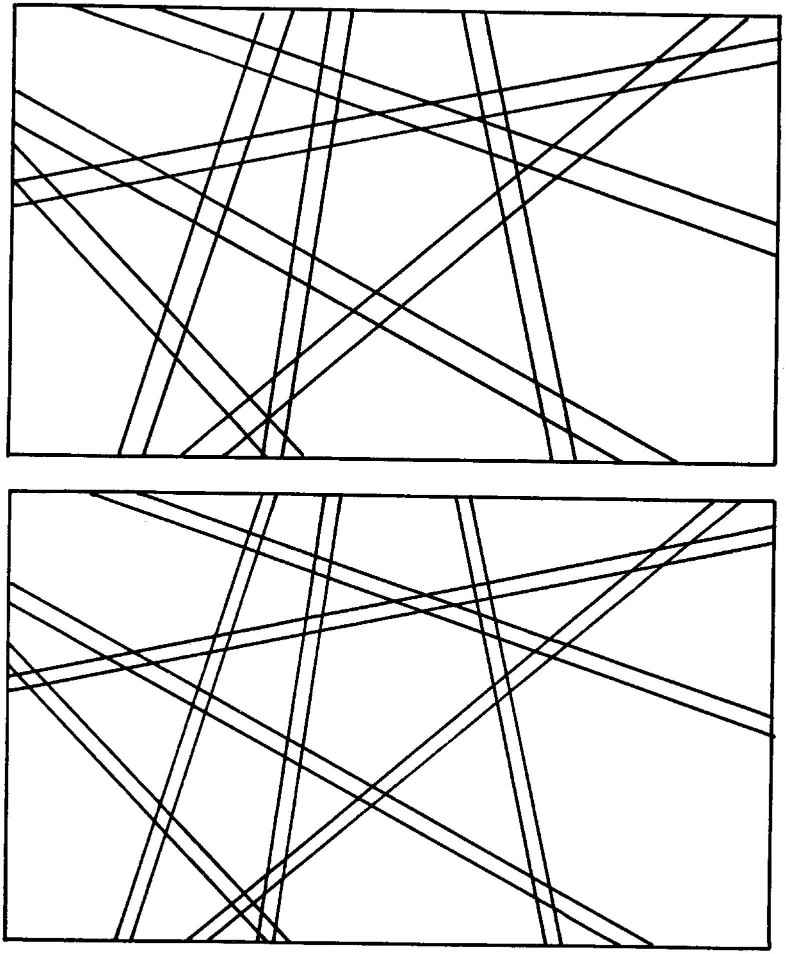

Fig. 2—Schematic plan view of paper illustrating that, if only the centres of bonded crossings are fixed, transverse shrinkage of fibres does not produce sheet shrinkage

occurs at their crossings or the fibres themselves diminish in length in the same proportion as the paper. An experiment was therefore carried out to determine the extent to which these mechanisms occurred.

Pieces of handsheet of bleached spruce sulphite at various beating degrees and having a few of their fibres dyed were photographed in a low power microscope after pressing, but before drying. The sheets were then dried with freedom to shrink by floating them on mercury and their shrinkage measured. The same fields were rephotographed and the longitudinal shortening of the dyed fibres was measured. It was found that, even in the case of shrinkages as high as 12 per cent, the longitudinal shortening of the fibres was the same as the shrinkage of the sheet. Careful examination of the negatives revealed that there had been no relative movement of the fibres at the points where they cross. An example of such a pair of micrographs showing 12 per cent shrinkage is given in the micrographs of Fig. 3. (The transverse shrinkage of the fibres is seen to be high and is in the region of 30 per cent.) The whole of the shrinkage of paper is thus identified with the longitudinal shortening of the fibres, values much larger than the natural longitudinal shrinkage being thus obtained.

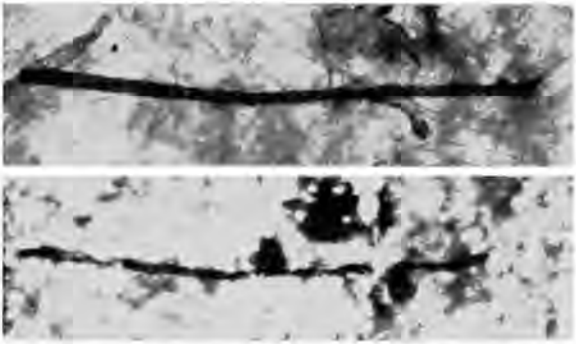

Fig. 3—A dyed fibre in a well-beaten sheet before drying (*above*) and after drying without restraint (*below*) (× 70)

At first sight, it might be concluded that Steenberg's suggestion of microcreping of fibres between bonds accounts for this effect (without bringing in at this point the mechanism of formation of the microcreping), but the largest sheet shrinkages occur in papers that are highly beaten and thus have the smallest fibre segments between bonds. (It has already been shown(7) that beating a sulphite pulp for only 20 min in the Valley beater considerably reduced the fibre length between bonds available for microcreping.) It is evident

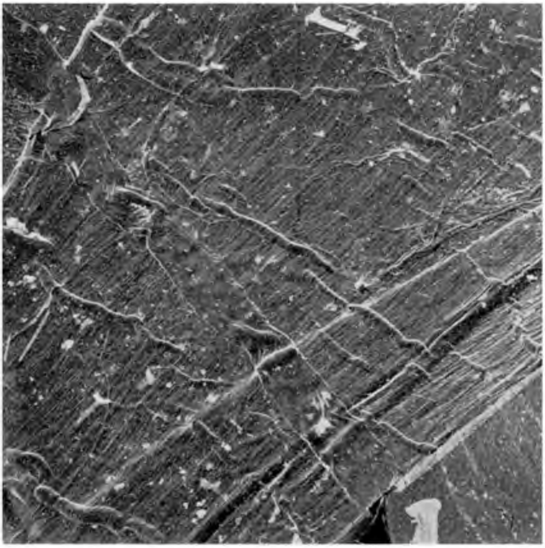

Fig. 4—(*Above*) Wrinkles (microcompressions) in a fibre produced by enforced longitudinal shortening when dried on cellulose film (× 180)

(*Below*) Microcompressions (on a smaller scale) similarly produced on fibrillated material (× 3 000) that in a greaseproof type of paper there is no unbonded length of fibre. This indicates that longitudinal contraction of a fibre is promoted not between bonds, *but at the bond sites themselves.* This can be accounted for only by the assumption that fibre-to-fibre bonding is sufficiently strong before complete shrinkage of the fibres for the transverse shrinkage of one fibre at a crossing to cause enforced longitudinal shrinkage of the other. Certain pieces of macroscopic evidence have been interpreted as supporting the view that fibre-to-fibre bonds are forming while the sheet is still wet and in its swollen state (for example(11)). It was considered of sufficient importance, however, to search for more direct evidence that would provide conclusive proof of this and a series of experiments was carried out to this end.

A drop of an aqueous suspension of fibres was placed on a strip of unplasticised regenerated cellulose sheet, which was constrained from shrinking in one direction and free to shrink at rightangles to it. The fibres and cellulose film were then allowed to dry, the resulting lateral shrinkage of the film being of the order of 10 per cent. It was found that, independent of the direction of orientation of the fibre with respect to the cellulose film, it remained bonded to it along its length and took up the shrinkage of the sheet. Along the fibres that were dried with their axes in the direction of large shrinkages, wrinkles were observed (*see* the micrographs of Fig. 4) and these are evidently due to local compressional failures of the fibres, which will in future be termed *microcompressions.*

An attempt was then made to reveal longitudinal shortening of fibres at bonded fibre-to-fibre crossings in paper sheets. While this effect is difficult to observe directly owing to the short spans involved, it was revealed by the examination of metal-shadowed replicas of freely dried sheets (in which the effect might be expected to be at its greatest). The micrographs of Fig. 5*a* and *b* show examples of microcompressions to be seen at bonded crossings.

Microcompressions in fibres were observed also within the body of a sheet of tracing paper. Many microcompressions were found in fibres oriented in the cross-direction, but none were observed in fibres lying in the machine-direction. Several fibres were observed that curled from the machine-direction to the cross-direction, but exhibiting microcompressions only in the cross-direction portion. Typical effects are illustrated in the micrographs of Fig. 6.

The first stage of this theory states then that transverse shrinkage of one fibre causes longitudinal contraction of crossing fibres at the bonds. While it is believed that this is the principal cause of sheet shrinkage, another important aspect must be considered. Fig. 7*a* represents in plan view a paper sheet prior to shrinkage. After shrinkage, the bonded crossings diminish in size. Since we have found that there is no relative movement of the fibres at their crossings, it follows that the *unbonded segments also must shorten to retain the shape of the structure.* This effect is illustrated in Fig. 7*b*, but is perhaps easier to understand from the cross-sectional view given in Fig. 8. It

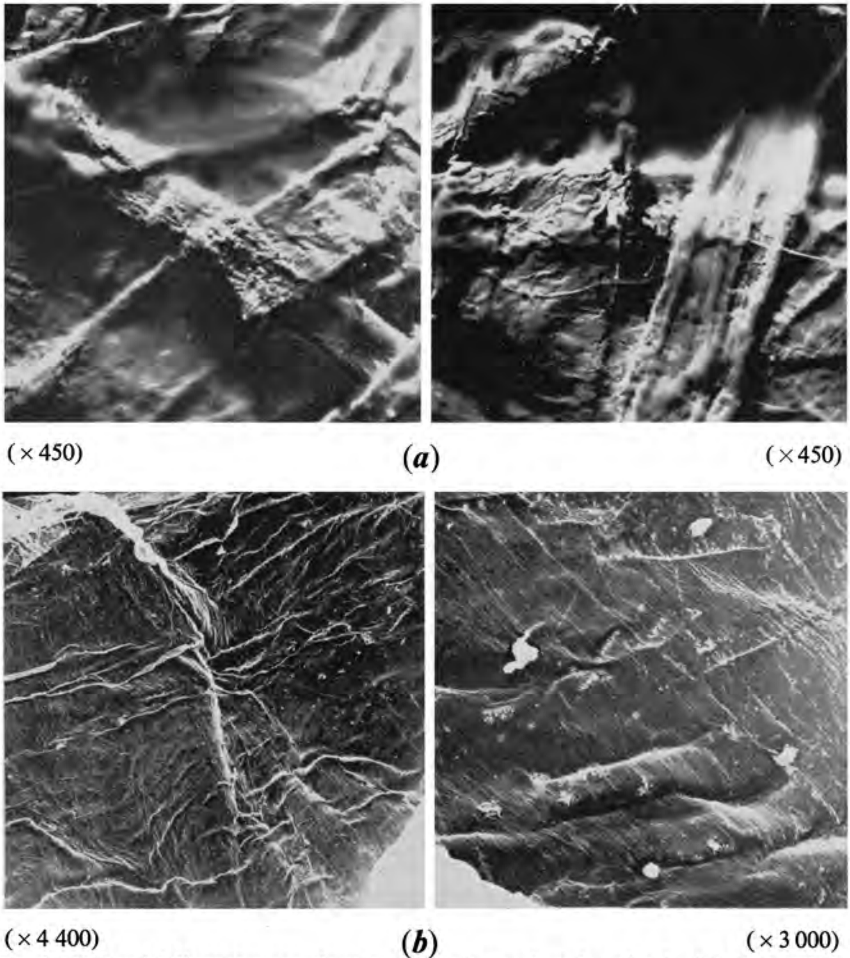

Fig. 5—(*Above*) Microcompressions at bonded crossings in a freely dried handsheet (*Below*) Examples of similar features in freely dried sheets commonly seen in the electron microscope

Fig. 6—(*Opposite*) The uncollapsed lumens of the fibres in the two upper micrographs make the effect more apparent and also serve to show that the microcompressions form throughout the body of the fibre (×525)

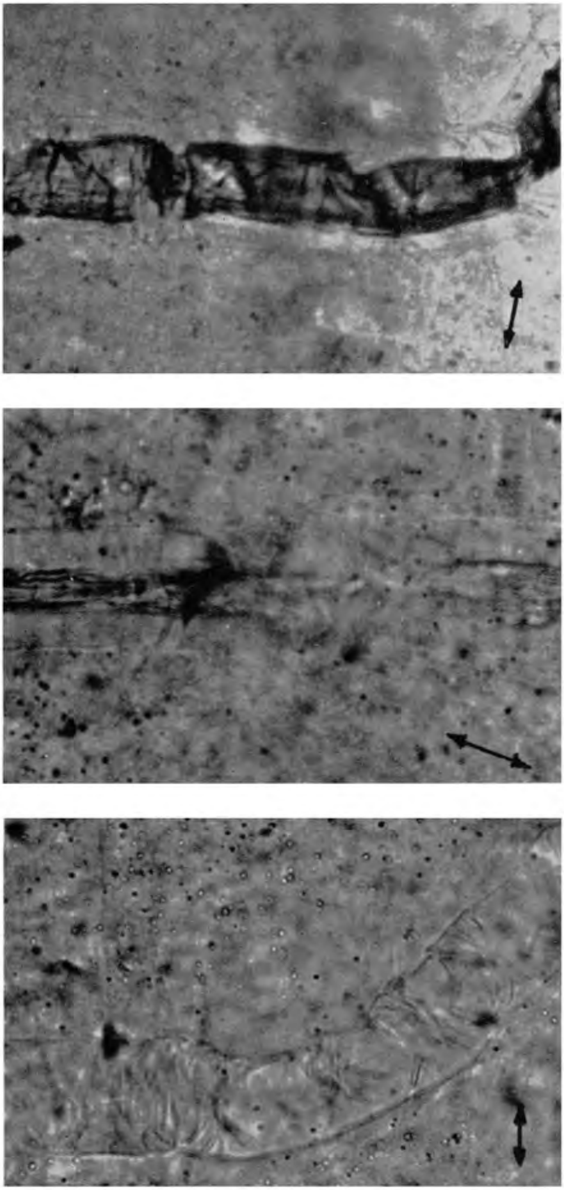

Fig. 6—Tracing paper, showing the preferred formation of microcompressions in fibres lying in the cross-direction (arrows indicate machine-direction) (*see also opposite*)

will be realised from this treatment that the mean shrinkage of the fibres within the sites of bonding is equal to the longitudinal shortening of the fibre segments between the bonds. This enforced shortening can manifest itself either as kinks or as microcompressions. It is likely that kinks form preferentially in the larger fibre segments, which occur more frequently in unbeaten sheets and in the surface layers of more highly beaten sheets, whereas microcompressions will form in short segments. (It is possible that formation of kinks may be aided by surface tension forces.)

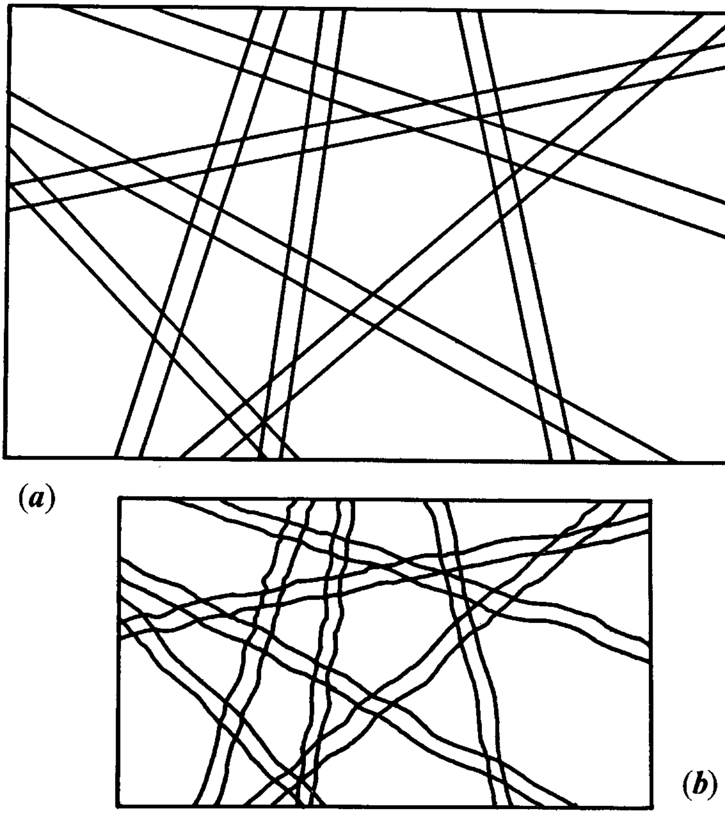

Fig. 7—Schematic plan view of paper illustrating that shrinkage of bonded crossings enforces longitudinal shortening of the fibres to retain the structure

So far, we have considered only the component of shrinkage that is derived from the shrinkage of whole fibres and no consideration has been given to the effects due to fibrillation. Fibrillation is visualised as a material contained within the structure and is capable of large shrinkage (owing both to physical effects of surface tension and chemical effects). Although the physical effects occur at higher moisture contents, it is thought that bonding has reached a sufficient level to impress the shrinkage of fibrillation in part on to the longitudinal shortening of fibres.

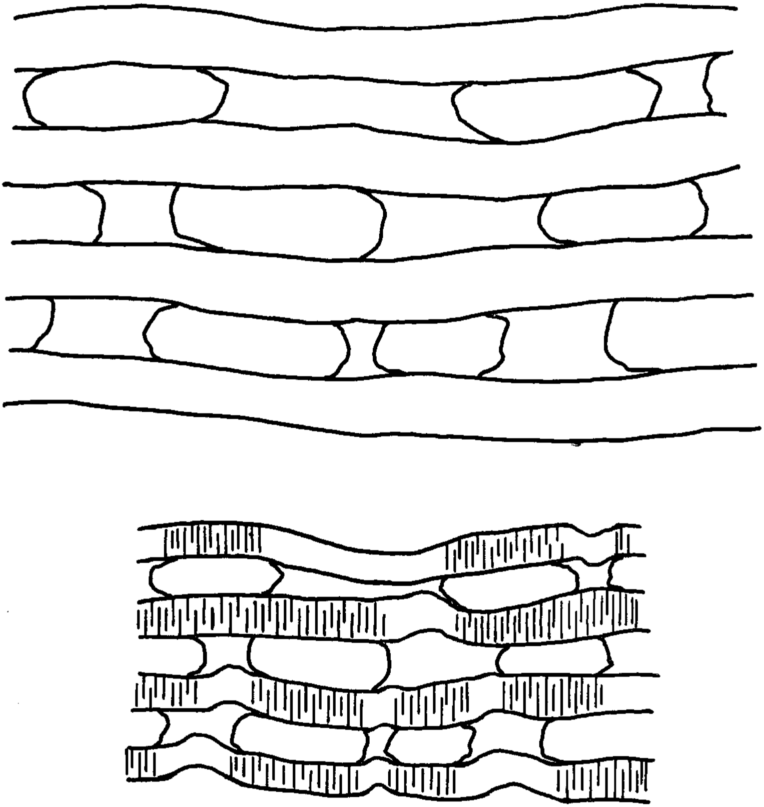

Fig. 8—Schematic cross-section of paper illustrating same point as Fig. 7: microcompressed regions of the fibres are represented by hatching

In summary, the theory presented here for the shrinkage of paper during drying is that fibre-to-fibre bonds become sufficiently strong to allow subsequent transverse shrinkage of the fibres and fibrillation to be imposed on the fibres to which they bond, causing them to shorten longitudinally, by forming microcompressions *at the bond sites*. Structurally, this enforces longitudinal shortening of the fibres in their unbonded regions. The mean shrinkage of the paper is equal to the total longitudinal shortening of the fibres, which is equal to the mean shortening of the fibres at the bonded regions, which in turn is equal to the mean shortening of the free fibre segments.

In the light of this theory, it appears that there are four major structural factors that control the shrinkage of paper—

1. The intrinsic potential shrinkage of the fibres.
2. The resistance of the fibres to axial compression during shrinkage.
3. The strength during shrinkage and the extent of the fibre-to-fibre bonding.
4. Fibrillation, which, when present, has shrinkage forces associated with it.

The effect of papermaking variables on shrinkage can be explained in terms of their effect on the above basic factors. It is intended to expound this aspect of the theory in a further publication.

Dimensional stability and wet expansion

THE field of Fig. 3 was rephotographed after immersion of the dried sheet in water (*see* micrographs in Fig. 9). The longitudinal expansion of the fibres was found to be the same as the expansion of the sheet with no relative movement of the fibres at their crossing points. It must thus be concluded that

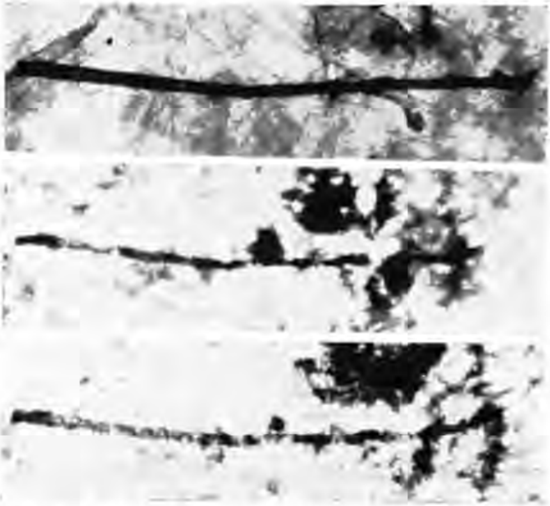

Fig. 9—The field of Fig. 3, but with (*below*) the sheet rewetted (× 70)

the expansion of the paper is due to the reversible part of the shrinkage of the fibres. Interfibre bond breakage plays no part in the expansion process. The transverse expansion of the fibres at their crossings is transferred to the longitudinal direction of the crossing fibres. The kinked and microcompressed fibres may themselves be dimensionally unstable as they contain a component of the transverse swelling in the longitudinal direction. The original transverse shrinkage of the fibres may not be completely reversible (Fig. 9) and this, together with the irreversibility of any surface tension contractions,(13) results in the incomplete recovery of the initial wet sheet dimensions.

The dimensional instability with respect to relative humidity follows the same pattern as wet expansion, but at a lower magnitude.

Rheological properties

THERE is no complete theory, even of a qualitative nature, for the rheological properties of paper. Attempts have been made to explain the elastic modulus of paper not by a stress analysis of its structure, but by using oversimplified assumptions. Onogi and Sasaguri(14) and later Litt(15) make the assumption that the fibre segments can be considered in isolation. The former authors assume all the fibre segments to suffer the same strain and the latter the same force. The extension of the sheet is then visualised as originating from the extension and bending of each segment. Implicit in these assumptions is that the lengths of fibre that are bonded do not contribute to the extension of the sheet. It would seem that this type of approach is incompatible with our own observations that paper is in general very well bonded.(7) Greaseproof paper, which is almost entirely bonded, does not have the infinite modulus that is predicted by this approach. It is apparent then that the extension of paper (particularly when it has a high density) is partly due to *deformation of fibres within the bonded regions.* At lower densities, it would appear that not only should elongation and bending of the segments be considered, but also shear in the plane of the sheet and shear in the plane perpendicular to the sheet lying on the axis of the fibre when bonds are staggered above and below it (Fig. 8). It should be emphasised then that the modulus of the sheet depends not only on the Young's modulus of the fibres, but also upon other moduli not yet determined.

The theory of shrinkage can now be applied to a qualitative discussion of the part played by shrinkage in determining the Young's modulus of the sheet. The microcompressed and kinked fibres will have a lower modulus than those restricted from shrinking by applied tensions and the fibres permitted to shrink transversely may have a lower transverse modulus. Thus, the theory explains the well-known sensitivity of modulus to sheet shrinkage.

The theory to be presented for the load/elongation properties of paper (for example, Fig. 10) is based on the new model in which shrinkage introduces microcompressed fibres at bonds and kinked or microcompressed fibre segments between the bonds. Stretching such a structure can be visualised to some extent as the reversal of shrinkage, the applied forces tend to pull out the kinks and microcompressions both between and within the bonds. It is most important to realise that *these cannot occur independently*. Just as the structure demands that the enforced longitudinal shortening of fibres at bond sites produces longitudinal shortening of fibre segments, so the pulling out of segments must be associated with the extension of the fibres at the bond sites.

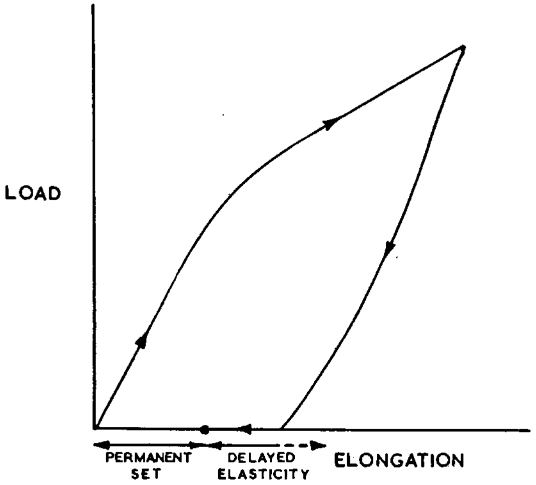

Fig. 10—Tensile behaviour of paper

There is no reason to assume that the structure of paper as we now visualise it (containing kinks and microcompressions to a degree depending on the shrinkage) should not have an initial linear elastic region. Fig. 11 shows the stress/strain curves of strips of brass 'kinked' to various degrees, but of constant cross-section. It will be seen that the elastic modulus decreases with increasing kink amplitude, but a 'toe' is not introduced at the beginning of the curve. The curve would display a 'toe' only if the kinks had a large amplitude and wavelength compared with the thickness of the member. The curves also show that yield for the highly kinked member occurs at an earlier stage and this is due to the stress concentrations introduced by the kink. It is reasonable to believe that the microcompressed fibres in paper display a similar trend in their stress/strain curves.

This latter experiment illustrates how the introduction of kinks can increase the permanent set of the members and this is believed to be the effect of shrinkage on the magnitude of the permanent set of paper. However, a further important factor must be considered. When the sheet is stretched, the microcompressions at the bond sites tend to be pulled out to an extent that depends on the orientation of the microcompressed fibre with respect to the direction of straining. At the same time, the crossing fibres are stretched transversely. A point is reached when the shear bond strength is exceeded and bond breakage occurs at the perimeter. It is reasonable to assume then that bond strength partially controls the shape of the load/elongation curve, in particular its second slope. As the bond slowly breaks, the microcompressed regions are released, allowing a greater extension and greater permanent set. This concept can thus explain satisfactorily not only the common occurrence of partial bond breakage, but the observation that cross-direction strips display greater partial bond breakage than do machine-direction strips.(8)

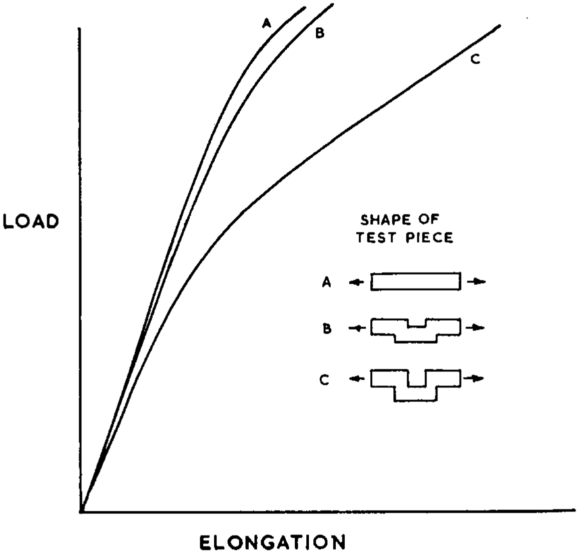

Fig. 11—Tensile behaviour of kinked structures

It will be apparent that the total separation of fibre-to-fibre bonds does not play a significant part in this theory of the stress/strain curve. We visualise that, if total bond breakage were sufficiently widespread to produce a significant degree of permanent set by pulling out kinked fibres, it would produce a much slacker structure, with a lower elastic modulus and this is not observed. Our observation of the infrequency of occurrence of total bond breakage supports the view that it should not be considered as a main factor contributing to the stress/strain curve.

Paper exhibits rheologically not only elasticity and permanent set, but also delayed elasticity (or primary creep). Although it has not been possible to consider this property in detail in the light of this theory, it must be considered to be a purely intrafibre phenomenon. It is not to be associated with frictional effects between fibres. (In fact, friction must be completely dismissed, since virtually every site at which it could operate is bonded.)

Summary

IT has been put forward here that the simple model of paper as a random three-dimensional network must be refined to incorporate the structural effects of shrinkage. It is shown that the transverse shrinkage of fibres (and the shrinkage of fibrillation, if present) gives rise to kinks and microcompressions throughout the structure to a degree controlled by drying restraints. It is this microstructure that principally controls the rheological properties of the sheet and many of the effects of papermaking variables upon the rheology can be explained through their influence upon the tendency to form this structure. The theory is applicable to the great majority of papers and can explain why the change of rheological properties from one extreme of beating to the other is a gradual one.

In conclusion, it will be observed that this structural theory predicts that many until now unmeasured properties of the structural components of paper contribute to its mechanical properties. These include the properties governing shrinkage—for example, the bond strength and the resistance of fibres to axial compression over the whole range of moisture contents. After formation, there are the shear moduli and transverse and longitudinal Young's moduli of the fibres and the effect of kinks and microcompressions on these moduli. A realistic quantitative prediction of the mechanical properties of paper from first principles must take account of these factors.

It is regretted that this paper has been prepared hurriedly, owing to the shortage of time between the conception of the theory and its presentation. Accordingly, no discussion has been given of previous theories nor has it been possible to acknowledge the origin of certain concepts that have been incorporated. It will be apparent, however, that we are deeply indebted to the many previous investigators in the field of paper properties. Our own work and the derivation of the theory has been inspired particularly by the published papers of Nordman, Steenberg and Rance, (e.g. 10, 12, 16, 17), certain of whose suggestions based on macroscopic data are now borne out by direct observation. It is intended to publish a fuller account of the theory elsewhere.

Acknowledgements

We are indebted to our colleagues Mr. D. R. Wembridge for carrying out the shrinkage experiments and Mr. J. W. Sargent for the electron microscope work.

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DISCUSSION

DR. W. GALLAY: I would like to ask Page or Nissan (or both) for their views on one key question about the relationship between bond breakage and rheology, on which there has been some difference of opinion in the literature. I refer to the chances of and the conditions for re-formation of interfibre bonds after the removal of the stress in stress/strain cycling. If I remember correctly, at least two authors have said that these bonds do in fact re-form on relaxation, several others have said they do not; others have said that it depends on the relative humidity.

MR. D. H. PAGE: We do not believe that fibre-to-fibre bonds re-form after optical contact is broken. We have observed a very small number of cases in which increases in optical contact after straining have occurred and we have been mystified by this, but we do not believe that it is bond re-formation. Within the body of the fibre, it is a totally different story.

MR. Z. J. MAJEWSKI: Craven and I are working on the effects of shrinkage on sheet properties. We are trying to relate stress behaviour to the length of paper, taking the formed length as 100 per cent.

A wet strip of paper is clamped in the jaws of a stress/strain recorder and is dried by means of infra-red radiation for a predetermined time. The paper is allowed to shrink without any restraint until any chosen length is reached. This is done by moving one clamp manually. After reaching the chosen length, development of drying stress is allowed to take its natural course. When dried, the paper is destrained, cooled off and a stress/strain—or rather a stress/length—curve is determined (Fig. D11).

For a free-to-shrink paper, it can be seen that the stress/length curve embraces all the other curves. Probably, some additional bonds have been allowed to form as a result of freedom to shrink. In fact, when this paper is subjected to a destraining cycle at any given length, its behaviour is very similar to the behaviour of paper dried to this length only.

Other points of interest are—

(*a*) All papers dried with varying degrees of shrinkage (but not necessarily when extended before or during drying) break at about the same absolute length.

(*b*) All papers show about the same breaking tensile strength (the differences in width caused by differences in cross-direction shrinkage are neglected in this technique).

(*c*) The part of the stress/length curve below the level of drying stress is a straight line, indicating a purely elastic behaviour up to that point.

(*d*) The strain at which the elastic limit is reached shows remarkable constancy. This may represent an 'elastic strain' of a group of bonds carrying the load in a purely elastic manner.

The curve of drying stresses can be assumed to represent the number of bonds formed at any given length of paper. During straining, these bonds are being gradually engaged in carrying the load. The area above this curve of elastic limit represents a 'post-elastic' behaviour of bonds stressed already past this elastic limit. Stress/length behaviour of paper can, therefore, be considered as composed of an elastic and post-elastic behaviour during the whole process of straining.

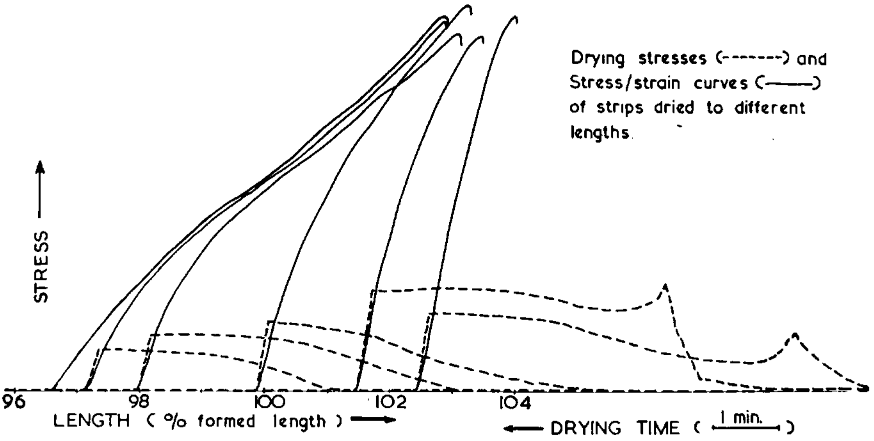

Fig. D11

When the above reasoning is taken a step further, it is possible to obtain a stress/strain curve for a single group of bonds. After having reached the elastic limit, the curve falls rather sharply.

MR. B. RADVAN: One of Wrist's earlier slides showed a single fibre bonded across some six others, which prevented lateral shrinkage at the points of bonding and these fibres showed strong 'necking'. Is such necking visible as well in photographs of real papers in which lateral shrinkage is also prevented—for example, the cross-direction fibres in a tracing paper? In fact, what proportion of the shrinkage takes place after (rather than before) the bonding?

MR. PAGE AND MR. P. A. TYDEMAN (*written reply*): It is part of our theory, of course, that fibres in paper dried under tension are prevented from shortening in length and this restricts the crossing fibres from transverse shrinkage in the bonded regions. This will give rise to the 'necking' phenomenon, but the observation of this is difficult, owing to the fact that fibres are bonded in regions along much of their length, leaving only small regions for free shrinkage. The effect was particularly evident, however, in one of the slides that we showed of a sheet of self-bonding rayon fibres, also in some of the scanning electron micrographs taken at the Canadian Pulp and Paper Research Institute.

We do not know the proportion of shrinkage, if any, that takes place before bonding. We are only able to say at present that sufficient bonding is present while appreciable potential shrinkage of the fibres remains.

MR. P. E. WRIST: The evidence presented by the authors for substantial longitudinal shrinkage of the fibre is very convincing, but I would like them to clarify exactly what they consider to occur in the local compressional failure that they call a microcompression. My impression is that the wrinkles or folds we have seen on the surface layer of several photographs are indicative of a fold extending all the way through the fibre. I very much doubt that this is true in the case of thick-walled summerwood fibres.

The primary wall with its interlaced structure is unable to shrink on drying and therefore, if constrained by bonds to a section of S1 or S2 layer, which normally shrinks orthogonally to its fibril direction, the primary wall will be thrown into wrinkles. The S1 and S2 layers, on the other hand, are structurally able to shrink on drying and, in the absence of any restraining force, they do so principally at rightangles to their fibril direction. Their ability to shrink arises from the near parallelism of their fibrils and their reduction in volume as water is withdrawn from their open swollen structure. Such a structure subjected to a longitudinal compression during drying would be able to compress, however, without the need to produce a coherent wrinkle across the entire fibre. Minor curvilinear bending along individual fibrils would be the most probable method and could produce compressions as high as 10 per cent. Only in thin fragmentary layers of S2 would wrinkling be more likely to occur and, if I recall correctly, the photographic evidence of wrinkles in S2 layers were of this nature.

Wrinkling of the P layer is not restricted to fibres in web form, but occurs also in freely dried single fibres. Here, a ready distinction of springwood and summerwood is evident. In springwood, the shrinkage is largely controlled by the S1 layers and wrinkles form in the P layer lying along the S1 spiral angles. With summerwood, the larger bulk of the S2 layer controls and wrinkles are predominantly axial in direction. The free fibre also becomes highly twisted.

My second question concerns the progress of strength development with beating. It has been demonstrated by several of the authors that during the shrinkage phase of drying many of the already formed bonded areas are subjected to high strains, resulting in bond breakages. Is it possible therefore that the explanation for the maximum and then decrease in many of the strength properties with beating time is explained by the increased shrinkage strains and increased bond breakage that accompany beating, rather than by the more customary explanation of fibre damage and shortening? Do you have any comments on this suggestion?

MR. PAGE: The work described by Tydeman and myself says little about the nature of the longitudinal shortening of the fibres throughout the cell wall, but merely draws attention to its existence. Electron microscope observations by my colleague Sargent are aimed at clarifying the situation, but it would be premature to report results here. I do not agree that the micrographs we showed are indicative of a *fold* extending all the way through the fibre, although they are sometimes indicative of a *fault* throughout the fibre wall, perhaps similar to compression failure of fibres in wood. This occurs in quite thick-walled cells in tracing paper. Curvilinear bending along individual microfibrils in an incoherent manner could surely not give compressions as high as 10 per cent without considerable transverse expansion of the structure. If bending of the microfibrils occurs coherently, it becomes a fault throughout the structure.

For the second question, Wrist's suggestion could be tested experimentally, since it predicts that the scattering coefficient of sheets should rise during the final stages of beating.

MR. H. W. EMERTON: Page illustrated phenomena along the length of the fibres lying in the surface of a paper sheet, which he has called microcompressions. Surely, we have both observed very similar phenomena along the length of single fibres dried on to glass. In this case, the mechanism that you put forward for fibres in paper does not operate.

MR. PAGE: No, we are convinced that these microcompressions are formed on freely shrunk paper and that they are caused by the mechanism we have stated. We are happy that we have observed real effects.

Written afterthought—The observations referred to by Emerton were made on fibres resuspended from dried pulp sheets and, in this drying process, microcompressions will have formed.

DR. H. CORTE: I am most impressed by this new piece of experimental microscope evidence and detailed observation. The danger begins, of course, when it comes to connecting microscope observations of details of single fibre crossings with the physical behaviour of a large number of fibres in a network. This is because the individual behaviour of a single fibre or fibre bond can be completely superseded by secondary effects resulting from the simultaneous (or nearly simultaneous) action of many of them.

To take an example—one can say the shape of a microscopic stress/strain curve is always *concave* towards the abscissa, whereas the mechanism for the pulling out of kinks would lead to a convex curvature. Is that right? (It was one of the points you made in favour of it.) If you stress a two-dimensional (that is, very thin) sheet, the stress/strain curve consists of a large number of peaks each marking the end of a short *convex* curve and it is only because of the inertia of the tensile tester that the curve is not the same in thicker sheets. With increasing basis weight of the sample and the same rate of elongation, the convex-shaped discontinuous character of the stress/strain curve gradually converges into the well-known steady concave character. This does not necessarily prove that the pulling out or ironing out of kinks between crossings does not occur, but can be due merely to too fast a rate of elongation or that the tensile tester is not sensitive enough to detect events on the scale of individual fibres or fibre crossings.

MR. J. G. BUCHANAN: How do fibres at fibre crossings shrink in the sheet interior, where they are bound on both sides? Do they kink or do they compress axially?

MR. PAGE: The experimental evidence is that in the body of sheets fibres do shrink in length to the extent of the shrinkage of the sheet itself and this is independent of beating. We are compressing the whole of the fibre. We do not yet understand the mechanism of the shortening. We might be shortening the microfibrils!

DR. H. F. RANCE: I think the paper presented by Page and Tydeman is a most interesting and valuable contribution that definitely advances our understanding of wet web shrinkage and allied phenomena. In some respects, I do not fully share the views of the authors, especially in the more speculative parts of their paper. For example, their rejection of the possible significance of frictional factors in the deformation of paper seems unwise, since currently available techniques can give no evidence either way. One must question also the wisdom of extrapolating as boldly as they have done in building a comprehensive theory claimed to be applicable to almost every type and thickness of paper upon limited observations of the surface layers of one type of paper.

On the other hand, I must express full agreement with and approval of the authors' theories when they are firmly based upon unequivocal experimental observation. Their micro-studies of the visible characteristics of bonded areas and their arguments concerning the interdependence of the two types of microcompression are both convincing and significant, especially when related to their observations on the incidence of partial bond breaking.

This is excellent work and I am delighted that at long last my call for 'more precise researches directly designed for investigation of the fundamental aspects of this technologically important subject' (quoted in the paper(12)) is now being answered.

THE CHAIRMAN: I am quoted here as being also partially responsible. I cannot see very much difference now to my ideas of 15 or 16 years ago. The concept that I introduced was micro-creping and this was only a name for secondary creep that made the idea more easily understandable.

DR. J. A. VAN DEN AKKER: The authors' remarks on the effect of anisotropic shrinkage are very interesting and valuable. A number of years ago, when we did our first work on the hygroexpansivity of paper, we wondered why equilibration of the sheet required so much more time (following a change in relative humidity) than equilibration in moisture content determination. For the same degree of approach to equilibrium, the former required time intervals several times those of the latter. It occurred to us that some of the effect might be due to anisotropic shrinkage of the fibres at the fibre-fibre bonds. This would produce mechanical stresses in the bonds (as discussed in my present paper) and the consequent creep effects, which involve notoriously long intervals of time, might well account for the long equilibration periods, as well as for the dimensional instability itself. With regard to the latter, we are not at all sure that anisotropic shrinkage of the fibres at the bonds is the major factor in the hygroexpansivity of paper, as certain experimental data are not consistent with this assumption. We believe, with Page, that it is an important factor.

PROF. G. JAYME: Two electron micrographs that I showed (Fig. 16 and 17 in my paper) proved that in caustic-treated pulp the fibre surface appears wrinkled and that this wrinkling takes place practically in the S1 layer only. Fig. 16 shows this effect as such. In Fig. 17, part of this wrinkled S1 layer has been shorn off and the practically unwrinkled S2 layer can be seen clearly.

MR. PAGE: This is an interesting and significant observation. It shows that the shrinkage of one part of a cell wall can enforce the wrinkling of another that is bonded to it. It is exactly in this way that we picture the shrinkage of one fibre microcompressing another bonded to it.

DR. O. L. FORGACS: I have often seen on the scanning electron microscope the type of shrinkage described by Jayme.

Written contributions

DR. J. D. BROATCH: Could the authors comment on the fact that a small percentage of glass fibres in the furnish can bring about a considerable decrease in the shrinkage that occurs during drying?

MR. D. H. PAGE AND MR. P. A. TYDEMAN: We had hoped to deal with this question in our delivery text as an illustration of the application of our theory, but time did not permit this.

During shrinkage, each glass fibre suffers forces of axial compression owing to the transverse shrinkage of fibres bonded to it. Glass fibres are, however, sufficiently rigid to resist these forces and cannot suffer microcompressional shortening at the bond sites. Moreover, to retain the local structure, which is virtually a triangulated network, this prohibition of shrinkage in the longitudinal direction of each glass fibre is transmitted to a region surrounding it and so a relatively small number of glass fibres produces a considerable reduction in the shrinkage of the paper and an improvement in its dimensional stability.

Thus, the effect on the dimensional stability of paper of the inclusion of glass fibres is seen to rely on the combination of two properties—their high resistance to axial compression and their appreciable strength of bonding to swollen cellulosic fibres. The dimensional stability of glass, while an essential feature, is not considered to be the main reason for its inclusion and it is predicted that fibres having perfect dimensional stability would be of little value for the stabilisation of paper, if their axial compressive resistance were poor.