Preferred citation: B. Radvan, C. Dodson and C.G. Skold. Detection and cause of the layered structure of paper. In **Consolidation of the Paper Web**, *Trans. of the IIIrd Fund. Res. Symp. Cambridge, 1965*, (F. Bolam, ed.), pp 189–214, FRC, Manchester, 2018. DOI: 10.15376/frc.1965.1.189.

DETECTION AND CAUSE OF THE LAYERED STRUCTURE OF PAPER

B. RADVAN, and C. DODSON, Wiggins Teape Research and Development Ltd., Butler's Court, Beaconsfield, Bucks. and C. G. SKOLD, Pappersbrukens Centrallaboratorium, Stockholm

---

Synopsis—A method is described of determining the distribution of fibres in the thickness of a sheet of paper. It relies on observing the disposition of a small proportion of dyed fibres in a transparentised sheet. All the samples examined show a highly layered structure.

Density profiles of the boundary of a sheet being formed in a drainage apparatus have been measured and the movement of single fibres was observed in approaching the forming zone in a model of the wire part of a papermachine. Both experiments show that a diffuse zone exists at the boundary of the forming mat, in which thickening of the stock takes place. The length of this zone is only a few millimetres and most of the formation takes place by filtration rather than by thickening. This finding is confirmed by computations based on a theory of formation, which includes both thickening and filtration. An argument is advanced that the layered structure of paper is the inevitable result of this mechanism of formation, which takes place at the usual papermaking consistencies. Much higher consistencies would be needed in order to produce a more felted structure of the sheet.

Nomenclature

h = the mean depth of a fibre within the sheet, see equation (*2*), dimensionless

s = the fractional depth of a point within the sheet, that is, that proportion of total length of fibres that lie above it, see equation (*1*), dimensionless

r = the observed proportion of overcrossings in all crossings of a selected fibre, see equation (*11*), dimensionless

G = the density distribution of h with respect to s, see equation (*3*), dimensionless

G_N = the nth cumulative parameter of G, see equation (*9*), dimensionless

$P_{A,T}$ = the expected proportion of observed random selections of T crossings which contain A overcrossings, see equation (*7*), dimensionless

P_A = the values of $P_{A,T}$ at $T=4$ ($A=0,1,2,3,4$), see equation (*8*), dimensionless

c = the concentration of slurry at time t, position x, g/cm^3
c_0 = the value of c at $t=0$, g/cm^3
n = a constant in equation (*3*), cgs units
t = time from the moment of release of the piston, sec
u = the downward velocity of the stock at position x, time t, cm/sec
x = the distance of a point within the slurry from the closed end of the drainage apparatus, cm
A = a constant in equation (*4*), cgs units
D = a constant in equation (*4*), cgs units
L = the position of the piston face at the moment of release, cm
R = a constant in equation (*3*), cgs units
V = downward velocity of the piston at time t, cm/sec
σ = the superficial compressive stress within the fibre slurry at time t, position x, dyn/cm^2

PART 1—ARRANGEMENT OF FIBRES IN THE THICKNESS OF PAPER

Introduction

IN THIS first part, we describe a method of determining that aspect of the structure of paper that has probably most relevance to its behaviour in consolidation—namely, the arrangement of fibres in the z-direction (the thickness of the sheet). It appears from such determinations that fibres in a sheet of paper lie mainly in layers (we use this term in a sense to be explained later). In the second part, we argue that this result is a necessary consequence of all sheetforming processes known to the authors and therefore applies to all types of paper and non-woven material. It follows from this argument that certain fundamental limitations of the papermaking process must be overcome before a non-layered paper is produced.

Present state of knowledge

AN EARLY formulation of the statistical geometry of fibre networks by Corte & Kallmes(1,2) was further developed in a series of articles by Kallmes and co-workers.(3–6) The problem is approached here by considering a large number of fibres distributed over a plane web, either at random or nearly so. A sheet of paper is then assumed to consist of a number of such 'two-dimensional' layers, piled one on top of another. This latter assumption appears to have served originally as a mathematical simplification, although some indirect support from measurements of the physical properties of paper has been obtained(2) and, indeed, forms the subject of one of the papers at this conference.(7) Publications by Page and Tydeman(8) and Van den Akker(9) treat the paper as essentially a two-dimensional structure, although recent work(10) concerns a method of examining paper structure in depth.

Wahren's analysis of the structure of fibre slurries(11) is also clearly relevant and may yet be extended to treat finished paper.

Most other published work on the structure of paper thickness appears to be mainly empirical and chiefly concerned with the distribution of fines and loading through the sheet and with its two-sidedness. Underhay,(12) Judt(13) and Groen(14) used the techniques of stripping the paper in layers with adhesive tape. A very elegant variant of this method was developed by Parker(16) and much progress may be expected in the near future from its application. Wood(17), Forgacs & Atack(15) and Lehtinen(18) achieved the same object using microtomes.

There is rather less published work about the disposition of the fibres in the thickness of the sheet. Finger & Majewski(19) found several discrete layers in a sheet of newsprint, each with different fibre orientation. They also demonstrated a degree of intermingling—or felting—of fibres between the layers. Sauret,(20) also using the stripping technique, detected the presence of discrete top and bottom layers in light and medium weight sheets. Wrist(21) suggests that fibres in the wire may be oriented more at right angles to the plane of the paper than those in the rest of the sheet.

Thus, it is common ground that fibres in paper lie essentially in the plane of the sheet. This becomes obvious on examining a cross-section through the thickness of paper and very obvious when it is of a non-woven material.(22, 10) Indeed, a fibre that may be some millimetres long, must clearly lie almost parallel with its plane inside a sheet, which is only some hundredths of a millimetre thick. It is by no means clear, however, whether such fibres lie wholly in one part of the sheet—top, middle or wire—or whether they penetrate from one layer to another without departing much from the plane of the sheet—and, if so, to what extent. We know of no published method that could answer this query conclusively, certainly not in a quantitative manner.

Determining the arrangement of fibres

A SMALL proportion of stock (1–5 per cent) is dyed a deep shade and well dispersed in the rest, so as to form a representative sample. The sheet is then made transparent by saturating it with, say, Canada Balsam and the relative dispositions of the dyed fibres are deduced from the following observations under a microscope.

A fibre is selected at random (that is, with the probability of selecting a fibre of a given class proportional to the total length of fibres in that class) and observed along its whole length. The number of other fibres that cross over or under the selected fibre is noted and the observations are repeated for a large number of other selected fibres. The following section demonstrates how it is possible to deduce the arrangement of fibres in the sheet's thickness from the distribution of the ratios of overcrossings to undercrossings. In general, a layered sheet will give a widespread distribution of ratios, whereas a more narrow distribution will result from a felted sheet (see Fig. 1).

The sheet must be sufficiently transparent for even the lowest fibres to be visible with good definition. Apart from Canada balsam and other mounting media, we found some silicone oils to be very effective transparentisers, but inconvenient to use. Tricresyl phosphate is recommended by Vanzio,[23] but probably for industrial rather than laboratory use. Even with the best transparentisers, observations become very difficult with papers of substance exceeding some 100 g/m^2. Besides, we completely failed to transparentise some stocks—notably cotton linters—and some machine-made papers, the latter possibly because of sizing. A magnification of 400–600 appears the most convenient and the decision which of the two crossing fibres lies above the other is made by focusing on each alternately—some practice is required here. Visual assessment of the relative position of two such fibres made with, for example, a stereoscopic microscope we found to be very unreliable.

The method is least time-consuming when each observed fibre crosses not many more than four other fibres, as will be discussed in the next section. The right proportion of dyed fibres to produce this condition may be difficult to gauge, especially in a machine trial. It may be convenient then to add excess dyed fibres of different colours and, when counting, to disregard one or more of the colours.

The chemical woodpulp fibres in a newsprint sheet may be blackened by drying in the presence of hydrochloric acid: a method used by Prober[24] and Forgacs & Atack[15] to observe the orientation of surface fibres. The resulting sheets transparentise well, but the counts of over- and undercrossing are very laborious, because of the large number of crossings per fibre.

Fig. 4 shows a number of microphotographs of such transparentised sheets, taken at various focal planes. In many ways, this technique resembles the use of soft X-rays for the study of the internal arrangement of fibres in the sheet —for example, Pelgroms.[25]

Interpretation of the results

WE DEFINE the fractional depth S of a point within the sheet as that fraction of the total length of the dyed fibres that lies above it. Thus—

$$0 \leqslant S \leqslant 1 \qquad . \quad . \quad . \quad . \quad . \quad (1)$$

Then, the mean depth of a dyed fibre, of a projected length L, is—

$$h = \frac{1}{L}\int_0^L S\,dl \qquad . \quad . \quad . \quad . \quad (2)$$

Let also Gdh be the length of all dyed fibres that have a mean depth h, expressed as a fraction of the total length of these fibres—

$$\int_0^1 G\,dh = 1.0 \quad . \quad . \quad . \quad . \quad (3)$$

The distribution of G with respect to h is characteristic of the arrangement of fibres in the sheet thickness—for example, see Fig. 1. It is shown below that certain parameters of this distribution may be calculated from the observed distributions of the proportions of overcrossings to undercrossings, hence the original arrangement of fibres may be deduced.

Thus, the probability of selecting a fibre that has a mean depth h is—

$$G\,dh \quad . \quad . \quad . \quad . \quad . \quad (4)$$

The probability that another fibre, when crossing, is above rather than below the selected fibre is—

$$h \quad . \quad . \quad . \quad . \quad . \quad (5)$$

Therefore, the probability that the selected fibre is overcrossed A times in a random selection of T crossings is—

$$C_A{}^T h^A (1-h)^{T-A} \quad . \quad . \quad . \quad . \quad (6)$$

Combining (*4*) and (*6*), the expected proportion of A overcrossings in random selections of T crossings per fibre is—

$$P_{AT} = C_A{}^T \int_0^1 G h^A (1-h)^{T-A}\,dh \quad . \quad . \quad . \quad (7)$$

In the following, we will always assume $T=4$ for convenience; $A=0$, 1, 2, 3, 4.

Integrating equation (*7*) by parts, we obtain a system of equations—

$$\begin{aligned} P_0 &= 24G_{\mathrm{V}} \\ P_1 &= 24(G_{\mathrm{V}} - 4G_{\mathrm{V}}) \\ P_2 &= 12(G_{\mathrm{III}} - 6G_{\mathrm{IV}} + 12G_{\mathrm{V}}) \\ P_3 &= 4(G_{\mathrm{II}} - 6G_{\mathrm{III}} + 18G_{\mathrm{IV}} - 24G_{\mathrm{V}}) \\ P_4 &= G_{\mathrm{I}} - 4G_{\mathrm{II}} + 12G_{\mathrm{III}} - 24G_{\mathrm{IV}} + 24G_{\mathrm{V}} \\ \hline \sum P &= G_{\mathrm{I}} = 1 \end{aligned} \quad . \quad . \quad . \quad . \quad (8)$$

where

$$\begin{aligned} G_{\mathrm{I}} &= \int_0^1 G\,dh \\ G_{\mathrm{II}} &= \int_0^1 \int_0^h G\,dh\,dh \\ G_{\mathrm{III}} &= \int_0^1 \int_0^h \int_0^h G\,dh\,dh\,dh \quad \text{etc.} \end{aligned} \quad . \quad . \quad . \quad . \quad (9)$$

Thus, if we assume the form of G_{I}, the distribution of mean depths h (the arrangement of fibres in the sheet), we may calculate the expected values of the observed proportions P_A; conversely, we may calculate the values of the cumulative constants $G_{\mathrm{I}}, G_{\mathrm{II}} \ldots G_{\mathrm{V}}$ of the actual distribution of h in a sample from the observed values P_0, P_1, P_4—hence, assess the arrangement of fibres in the thickness of the sample. Thus—

$$\begin{aligned} G_{\mathrm{V}} &= \mathrm{P}_0/24 \\ G_{\mathrm{IV}} &= (P_1+4P_0)/24 \\ G_{\mathrm{III}} &= (P_2+3P_1+6P_0)/12 \\ G_{\mathrm{II}} &= (P_3+2P_2+3P_1+4P_0)/4 \\ G_{\mathrm{I}} &= 1 \end{aligned} \qquad \qquad (10)$$

In practice, random selections of four crossings per fibre are difficult to arrange. Instead, we take note of all the crossings over the selected fibre and calculate the proportion of overcrossings—

$$r = A/T \qquad \qquad (11)$$

until a frequency distribution of r is determined. (Note that the distribution of r, the proportion of overcrossings in different fibres, is in general more diffuse than that of h, the mean depths of fibres, because of the variable number of crossings per fibre.) This distribution cannot, as yet, be directly predicted from the distribution of mean depths (or vice versa), but the expected values of P_0, P_1, P_2, P_3, P_4 and, hence, G_{V}, G_{IV}, G_{III} and G_{II} can be calculated from it in the same way as before.
Thus—

$$\begin{aligned} P_0 &= \textstyle\sum r^4/S \\ P_1 &= 4\textstyle\sum r^3(1-r)/S \\ P_2 &= 6\textstyle\sum r^2(1-r)^2/S \\ P_3 &= 4\textstyle\sum r(1-r)^3/S \\ P_4 &= \textstyle\sum (1-r)^4/S \\ \hline \textstyle\sum P &= S/S = 1 \end{aligned} \qquad \qquad (12)$$

Simplified models of fibre arrangements

THE values of the cumulative parameters $G_{\mathrm{I}} \ldots G_{\mathrm{V}}$ of the distribution of mean fibre depths may be calculated from the observed proportions of $P_0 \ldots P_4$, but it is not immediately obvious how they relate to the arrangement of fibres in the sheet. We interpret them by comparing the experimental values of $G_{\mathrm{I}} \ldots G_{\mathrm{V}}$ of the examined sample with the values calculated for different simplified models of the arrangement of fibres in the sheet.

Two models in particular may be thought to represent two extreme types of arrangement—

(*a*) The layered model (Fig. 1A)—each part of each fibre lies at its mean depth; the mean depths are uniformly distributed over the thickness of the sheet.

(*b*) The felted model (Fig. 1B)—each fibre penetrates from the top to the bottom of the sheet; the mean depth of each fibre is $\frac{1}{2}$.

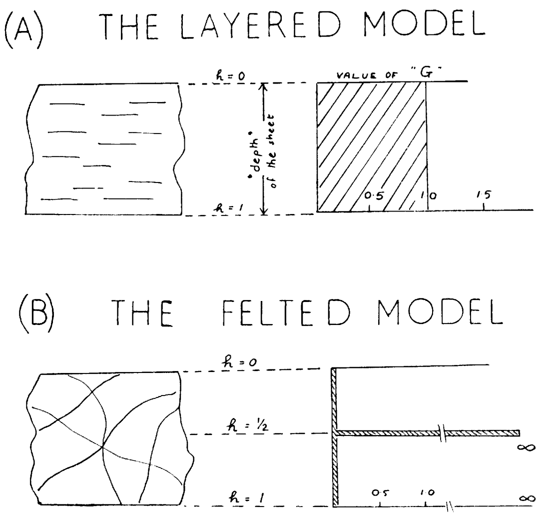


Fig. 1—The two extreme models of the arrangement of fibres in the sheet

A number of intermediate models, partly layered and partly felted, may be conceived. Two such possibilities are represented in Fig. 2—

(*a*) The multi-layers model (Fig. 2A)—the sheet consists of n layers, each fully felted within itself. For a felted sheet, $n = 1$; for a layered sheet, $n = \infty$.

(*b*) The mixed model (Fig. 2B)—the sheet consists of a proportion of fully felted fibres mixed with fully layered ones.

The values of the cumulative parameters G_{V}, G_{IV}, G_{III} are easily calculated for the two extreme models. The corresponding values for the intermediate models depend on the degree of felting and layering as defined for each model and are plotted as solid lines in Fig. 3. The experimentally determined values of G_{V}, G_{IV}, G_{III} are entered on the graphs; if all three correspond to the same degree of layering (that is, the same number of layers in model *A* or the same proportion of felted fibres in model *B*), then it is presumed that the model approximates well to the actual structure of the sample.

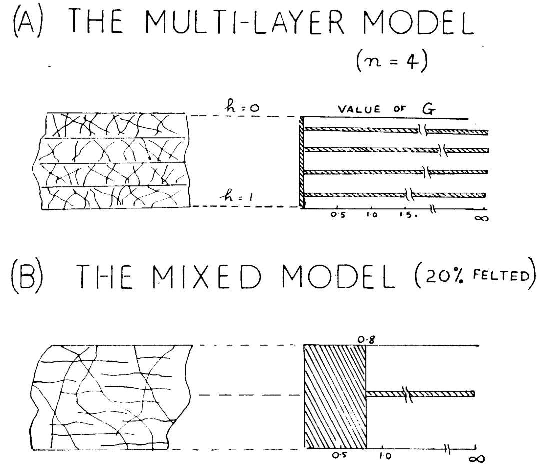


Fig. 2—Two intermediate models of the arrangement of fibres in the sheet

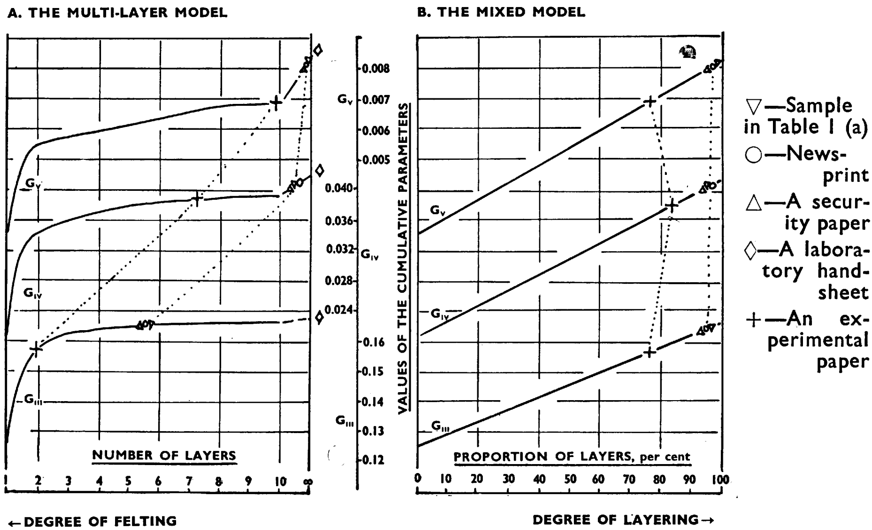


Fig. 3—Estimating the degree of felting from the observed distributions of overcrossings and undercrossings

Experimental results

TABLE 1 shows a record of counts of over- and undercrossings on three samples made from the same stock (free-beaten sulphite woodpulp, 80 g/m^2) in three different ways—on a slow Fourdrinier, on a British sheetmachine and on a model wire part (described in Part 2). The lower part of the table

TABLE 1—DISTRIBUTION OF PROPORTION OF OVERCROSSINGS IN OBSERVED FIBRES OF FREE-BEATEN SULPHITE PAPER, 80 g/m²

Sample	*Machine-made at 240 ft/min*			*Handsheet*			*Simulated machine conditions*		
Observer	*A*	*B*	*Total*	*A*	*B*	*Total*	*A*	*B*	*Total*
Ratio $r = O/(O+U)$	*Number of observed fibres*								
0.00–0.10	9	9	18	10	19	29	17	10	27
0.11–0.20	4	7	11	9	13	22	8	12	20
0.21–0.30	4	7	11	9	17	26	6	12	18
0.31–0.40	9	9	18	8	20	28	13	7	20
0.41–0.50	25	21	46	13	12	25	10	6	16
0.51–0.60	4	4	8	9	11	20	4	4	8
0.61–0.70	4	3	7	7	16	23	4	8	12
0.71–0.80	9	7	16	5	16	21	10	9	19
0.81–0.90	8	5	13	7	14	21	3	6	9
0.91–1.00	11	14	25	9	18	27	8	10	18
			173			242			167

O = Overcrossings U = Undercrossings

CALCULATIONS

Agreement between observers	$\chi_8^2 = 6.1$	$\chi_9^2 = 6.4$	$\chi_7^2 = 8.7$ *Good*
Agreement between samples	$\chi_{18}^2 = 623$		*Poor*
A $\sum r^4$	36.364	47.041	29.257
B $4 \sum r^3(1-r)$	32.768	45.672	27.940
C $6 \sum r^2(1-r)^2$	35.904	46.746	29.670
D $4 \sum r(1-r)^3$	35.784	50.092	36.008
E $\sum (1-r)^4$	31.767	51.816	43.697
F *Sum*	172.587	241.367	166.572
G $P_0 = P_4 = (A+E)/2F$	0.197	0.205	0.219
H $P_1 = P_3 = (B+D)/2F$	0.199	0.198	0.192
I $P_2 = C/F$	0.208	0.194	0.178
	1.000	1.000	1.000
$G_V = P_0/24$	0.0082	0.00855	0.00913
$G_{IV} = (P_1 + 4P_0)/24$	0.0411	0.0422	0.0446
$G_{III} = (P_2 + 3P_1 + 6P_0)/12$	0.165	0.168	0.172

shows the calculations of the three cumulative parameters G_V, G_{IV}, G_{III}, which are then used to characterise the structure of the samples. (Note that, because of the relatively small number of counts, the asymmetry in the distribution was averaged out.)

The resulting values of G_V, G_{IV}, G_{III} are then entered on the graphs in

Fig. 3, together with similar results for several other papers. Note that the estimates of the degree of layering (or felting) based on the multi-layer model are not consistent, each of the three values G_{V}, G_{IV}, G_{III} yielding a different estimate. Better fit was obtained using the mixed model. It is difficult to say at this stage whether valid conclusions may be drawn about the resemblance of the model to the actual structure of the samples—many more results will be required first—but all samples show a high degree of layering, irrespective of which model is used. In fact, two of the samples analysed in Table 1 cannot be represented by any of the models considered here, being more than 100 per cent layered. This is often the case with sheetmachine samples. Again, it is difficult to say at this stage whether this finding corresponds to an actual structure or simply results from error in observations. Note in this connection that random errors of observation would tend to exaggerate the apparent degree of felting and minimise that of layering.

Discussion and conclusions

THE main finding of Part 1 of this paper is to confirm the visual impression that one gains from examining micrographs of cross-sections of paper that the fibres in the sheet are strongly layered and to express quantitatively the degree of layering. The accuracy of the latter depends on carrying out very many visual observations and on choosing a suitable model to represent them. The present suggestion that the sheet consists mainly of strongly layered fibres, with a few felted fibres mixed in, appears attractive. For instance, Lehtinen[18] shows a cross-section of a fibre floc penetrating from top to bottom of a sheet—the flocs may be expected to be highly felted and the dispersed fibres in the rest of the sheet to be highly layered.

The search now continues for samples of papers or non-woven materials that would show a more felted structure, but we are of the opinion that all papers will show a highly layered structure for reasons explained in Part 2.

Finally, one might query whether a felted arrangement of fibres would give a paper with significantly different physical properties from a layered paper. Until such samples are available, the answer is a matter for speculation, but comparison with experience in the field of non-woven materials may be illuminating. There, attempts have been made to produce a felted structure by mechanical means—for example, needling,[26–28] disturbing the sheet with air jets[29] or water jets.[30] According to Buresh,[28] such materials are bulkier, drape better and may be used with less or no binder.

One would expect similar but smaller effects to occur if sheets of felted papers could be made, especially when using free-beaten stocks. The question is how to make them and some of the unavoidable problems involved in this challenge are discussed in Part 2.

PART 2—MECHANISM OF FORMATION AND ITS EFFECT ON ARRANGEMENT OF FIBRES IN THE THICKNESS OF PAPER

Introduction

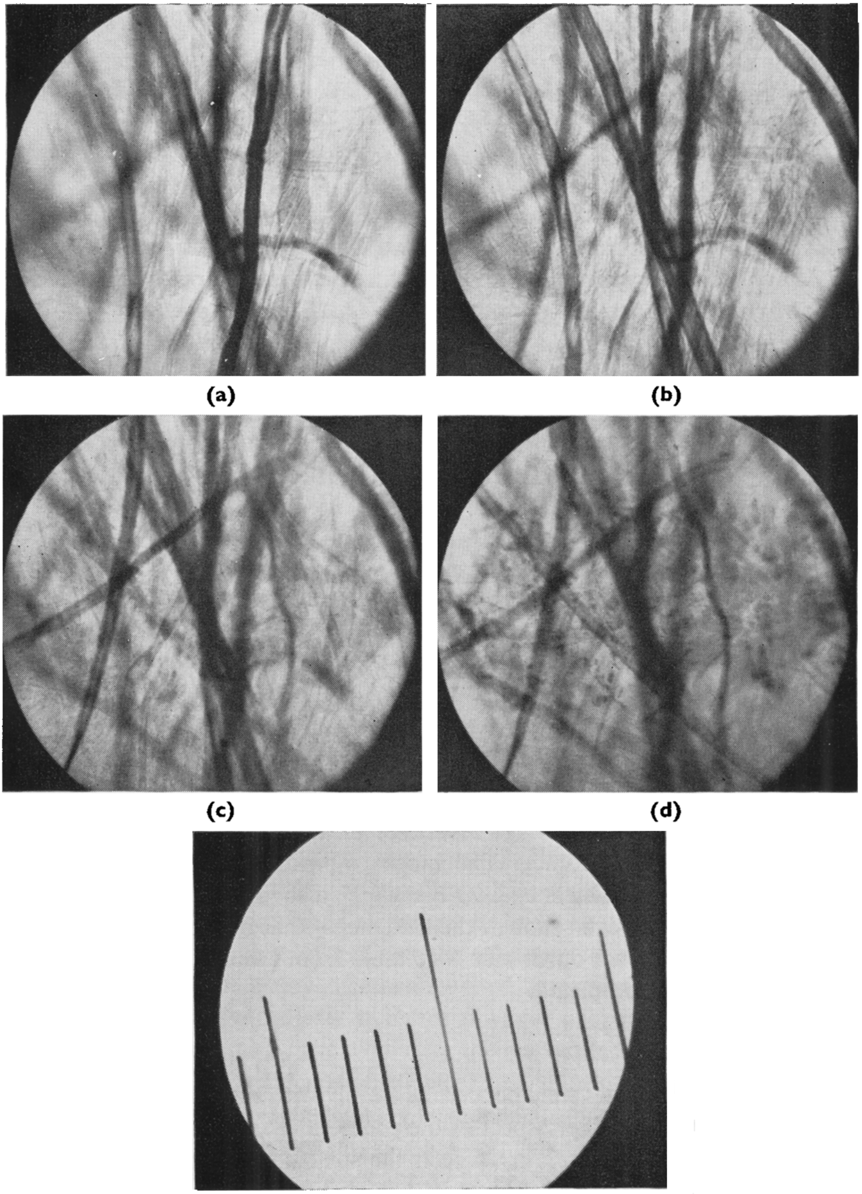
A DISTINCTION is often made between the processes of *formation*, in which a fibrous structure is produced from a structureless slurry and the process of *consolidation*, in which the slurry structure is essentially preserved and only density changes. Formation is represented as a filtration operation and consolidation as thickening and pressing. It is our purpose in this part to establish the extent to which the two processes occur in the manufacture of paper, since each may be expected to result in a different arrangement of fibres in the sheet's thickness.

Studies of rates of drainage

INVESTIGATIONS of the processes of formation and drainage have formed one of the most productive fields of papermaking research in recent years. Burkhard & Wrist,[31] Hendry *et al.*,[32] Taylor[33] and Bergström[34] succeeded in explaining the pumping action of the Fourdrinier table rolls. As a result, the forces that cause drainage became understood and most of the effort since then has been directed towards determination of the resistance to drainage that develops in a forming sheet. A series of investigations by Ingmanson and co-workers,[35-40] Robertson & Mason[41] and, more recently, the work of Meyer,[42] Nelson[43] and Meadley[45] have made it possible to predict the drainage resistance of a given fibre mat with a considerable degree of accuracy. The more empirical studies by Manson & Mardon[44] and Wahlström & O'Blenes[46] also appear to have largely achieved this object, even though the next step—that of predicting the drainage performance of a Fourdrinier wire table from laboratory measurements —is still far from completed.

Filtration versus thickening

MOST OF these studies were concerned simply with the rate of drainage and it seems to us that another aspect of the problem of formation—the question of how the process of formation affects the structure of the resulting sheet—has not attracted much attention. In particular, it has not yet been satisfactorily resolved, which of the two mechanisms of water removal—by filtration or by thickening—accounts for the formation of paper. In the first case, a discrete mat forms on the wire, sharply divided from a top layer of the slurry of the original consistency; in the second, the consistency increases simultaneously throughout the depth of the stock. Clearly, filtration must result in



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

Fig. 4—Microphotographs of sulphite fibres inside a transparentised sheet of newsprint—(*a*) focal plane at the surface of the sheet

(*b*) focal plane 1.3×10^{-2} mm below the surface

(*c*) focal plane 2.5×10^{-2} mm below the surface

(*d*) focal plane 3.5×10^{-2} mm below the surface

(*e*) graticule: 1 division = 0.05 mm

a layered sheet, whereas a sheet formed by thickening may (although need not necessarily) be more felted, in the sense used in Part 1.

The balance of opinion favours filtration, which is indeed often assumed, if only implicitly, to be the only mechanism of formation. Hisey(47) gives perhaps the clearest statement of the distinction between filtration and thickening and devotes most attention to the latter process, which, he states, takes place only towards the end of the drainage, unless the original stock consistency exceeds some 1 per cent. In an earlier paper, Finger & Majewski(19) conclude that paper filters into several layers, corresponding to successive drainage steps on the tube rolls of the Fourdrinier wire table. Ingmanson *et al.* discuss the problem in several papers, but only briefly; the earlier papers(36) estimate the minimum consistency of slurry to produce appreciable strength at approximately 2 per cent; the later estimates reduce this to 1 per cent.(42) In the latest publication,(40) Ingmanson considers that the boundary between the mat and the slurry is diffuse over a distance of one fibre length, but that this region has little effect on the drainage resistance. In contrast, Meyer(42) mentions a mathematical proof of the existence of a discontinuity at that boundary, although he does not produce it. Wahren(11) deduces the existence of a minimum consistency to produce a coherent fibre network by considering the probability of a fibre being interlocked with others at more than two points and finds that slurries of lower consistency show no measurable resistance to shear. Tellvik & Brauns(48) found no evidence of thickening in samples of the top layers of stock on the table of a medium speed experimental papermachine.

Indirect evidence concerning the question of filtration versus thickening is supplied by studies of the distribution of loading and fines in paper. All investigators find that the concentration of these materials decreases towards the wire side of paper,(12–18, 21, 49) but the interpretations differ. Underhay, for example, considers this to be the result of a backwash action of the tube rolls, with consequent disruption of the mat (and, effectively, thickening), but Wrist(21) argues that the results are more consistent with a selective retention effect.

The indications that thickening as well as filtration may also be an important forming mechanism are more scanty. Cowan(50) assumes that thickening occurs from the very beginning of drainage. Mason(51) deduces from the hydrodynamics of very dilute fibre suspensions that coherent networks will form even at very low concentrations. Forgacs, Robertson & Mason(52) succeeded in measuring the tensile strength of suspensions at concentrations down to $\frac{1}{3}$ per cent and demonstrated that the forces of cohesion are of the same nature as at much higher concentrations (up to 20 per cent).

The compressive strength of fibre suspensions has been measured in this laboratory at consistencies below 1 per cent (Fig. 5); it appeared that only the experimental difficulties prevented this measurement at lower consistencies. A very striking demonstration of the strength of dilute fibre networks and of the thickening effect occurs during slow drainage of long nylon fibres; the thickening zone may be observed stretching several centimetres ahead of the forming pad (Fig. 6).

We concluded that the question of the relative importance of thickening against filtration had not been settled and that direct observations of the

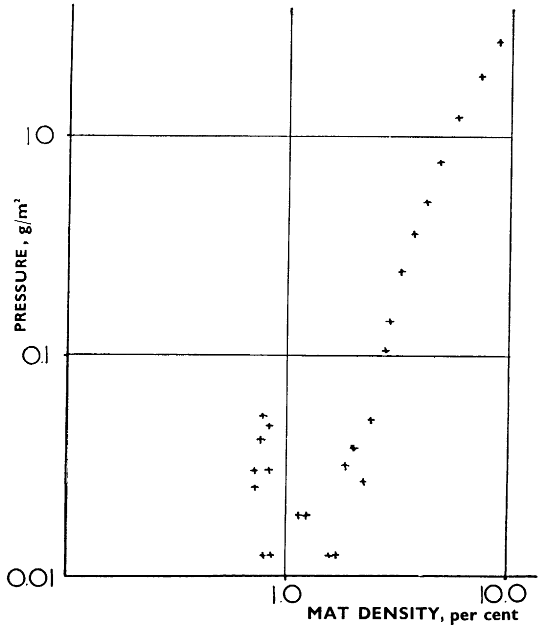


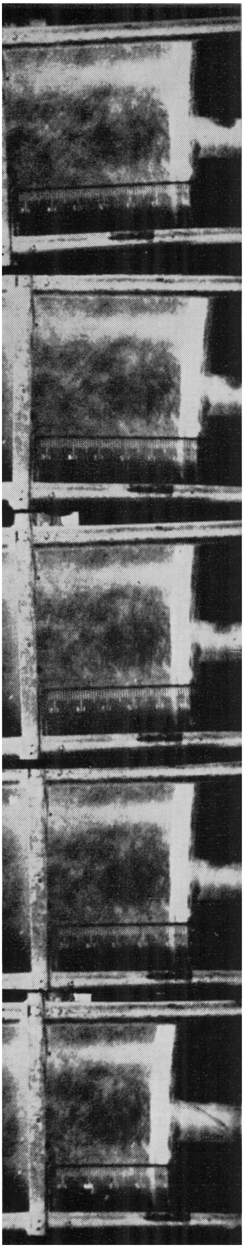
Fig. 5—Compression curve of the NSSC pulp at very low consistencies

manner in which fibres settle to form a sheet were needed. This was done in two ways—

1. By measuring the density profile at the boundary of a mat forming in a drainage apparatus.

2. By observing the motion of single fibres in the stock on the wire under machine conditions.

In the drainage experiment, if the mat was observed to have a sharp boundary or if the boundary was diffuse over only a short distance, then we would



conclude that thickening was of little importance. In the second experiment, the same conclusion would be reached, if fibres were observed preserving their relative positions in the slurry while approaching the forming surface.

Profile of a forming pad

Apparatus—The principle of the apparatus is shown in Fig. 7: a permeable piston rests on top of a volume of stock contained in a cylinder. When released, the loaded piston descends through the slurry, forming a mat of fibre on its face. Thus, the arrangement is a reversal of the normal drainage apparatus, with the resulting advantage of ease of pressure application, but also with the disadvantage of unknown friction between the piston and the cylinder wall and, of course, with the consequent neglect of the effects of fibre buoyancy on the process of formation (probably negligible).

A thin collimated beam of light shines through a layer of stock 2 cm wide and on to a light-detecting element, the response of which is calibrated in terms of consistency. As the piston approaches the beam, both the position of the piston and the change in consistency inside the light beam are recorded simultaneously on a fast recorder. When the experiment is repeated at different initial separations between the piston and the light beam, a series of density profiles of the boundary of the pad at various stages of formation is obtained. Only the thinner part of the pad can be profiled in this way, because the response of the photosensitive element falls off sharply with increasing consistency.

Fig. 16 and 17 show more detail of the apparatus and of the calibration.

Fig. 6—Appearance of thickening during the drainage of nylon fibre slurry at 0.6 per cent consistency (note the movement of flocs several centimetres ahead of the porous piston)

Theory—An attempt has been made to predict these profiles from knowledge of the compressibility and permeability as functions of slurry concentration. The process of drainage is assumed to be similar to the progress of a compression wave in a deformable solid, which results from the balance of the forces of elasticity against inertia and friction. In this case, elasticity is replaced by the compression resistance of a fibre network; inertia is neglected and friction replaced by the viscous resistance of water that permeates the network. Consider again Fig. 7.

At time $t=0$, let the piston be at height $x=L$ above the closed end of the cylinder; the consistency is then c_0 throughout. The piston is released and reaches a velocity V at time t_0. Let then a small element of the slurry at a

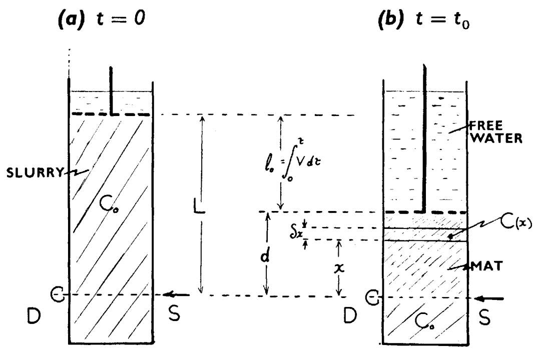


Fig. 7—Principle of the drainage apparatus

height x to $(x+\mathrm{d}x)$ above the closed end have a consistency c and move downwards with a velocity u. The condition of continuity yields—

$$\frac{\partial c}{\partial t} = \frac{\partial}{x}(cu) \qquad . \quad . \quad . \quad . \quad . \quad (1)$$

and the balance of compression gradient versus viscous drag—

$$d\sigma = ku \cdot dx \qquad . \quad . \quad . \quad . \quad . \quad (2)$$

where σ is the compressive stress and k the coefficient of permeability. In general, k and σ will depend on the concentration (neglecting time effects in a fast process). Say—

$$k = Rc^{n+1} \qquad . \quad . \quad . \quad (3) \qquad R, n \text{ constant}$$

$$\sigma = Ac^{D} \qquad . \quad . \quad . \quad (4) \qquad A, D, \text{ constant}$$

Differentiating equation (*4*) with respect to x and substituting together with equation (*3*) into equations (*1*) and (*2*) yields a partial differential equation of the parabolic type—

$$\frac{\partial c}{\partial t} = \frac{AD}{R(D-n)} \; \frac{\partial^2 c^{D-n}}{\partial x^2} \qquad . \quad . \quad . \quad . \quad (5)$$

The three boundary conditions necessary for the solution of equation (*5*) are as follows—

1. The concentration is constant throughout at the start—

$$c = c_0 \text{ at } t = 0 \quad . \quad . \quad . \quad . \quad (6)$$

2. The cylinder is closed at its lower end—

$$u = 0 \text{ at } x = 0 \quad . \quad . \quad . \quad . \quad (7)$$

3. No fibres pass through the permeable piston—

$$\int_{x=0}^{x=L=\int v\,dt} c\,dx = c_0 L \quad . \quad . \quad . \quad (8)$$

An equation similar to (*5*) has been derived more rigorously by Hisey,[47] but for a different set of boundary conditions. Hisey's treatment has recently been extended by Nelson[43] to include the effects of expression as well as of permeability—these we have neglected here.

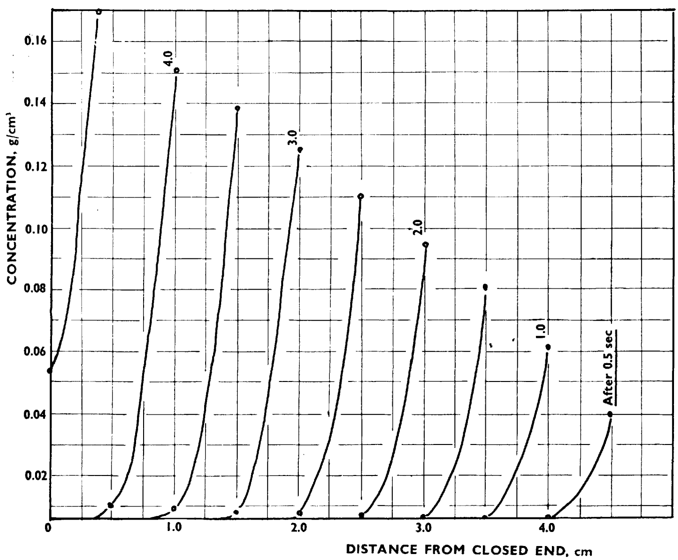


Fig. 8—Computed density profiles of the forming pad (piston moves with a constant velocity of 1cm/sec)

A numerical solution for the case of constant piston velocity was obtained by Burton[53] and Wallis;[54] more recently, Parker[55] has produced a solution by assuming the movement of the piston as actually observed during the experiment. The values of the constants A and D in equation (*4*) were extrapolated from measurements in Fig. 5; those of R and n in equation (*3*) were estimated from measurements of permeability.[41] The computed results are shown in Fig. 8 and 9 in the form of a series of concentration profiles at different times.

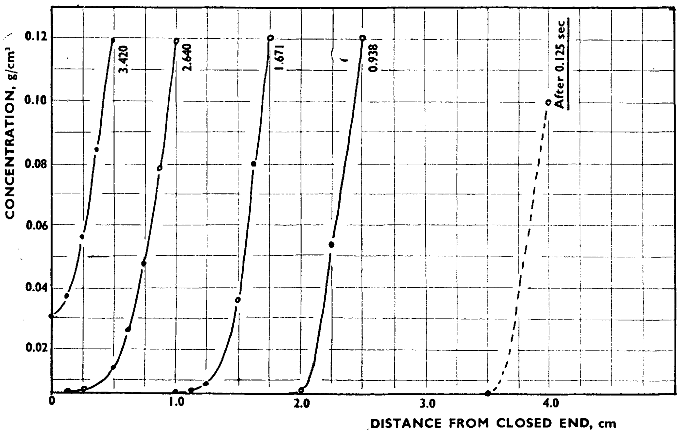


Fig. 9—Computed density profiles of the forming pad (piston movement as observed in the drainage experiment under a constant pressure of 10 g/cm²)

Results of drainage experiment—The main experiment used an NSSC pulp disintegrated to 19° SR. This pulp disperses easily, contains reasonably long fibres (Table 2) and is reputed to produce bulky sheets, indicating stiff fibres. We thought therefore that such a pulp would be the most likely to exhibit thickening effects, if any. Eleven series of runs were made, with piston loads of 10, 21, 44, 65, 114 and 229 g/cm² at a consistency of approximately 0.6 per cent and of 21, 44, 65, 114 and 229 g/cm² loads at 1 per cent consistency. Within each series, runs were made from five different starting positions of the piston, giving total piston travels of 1.7–7 cm. Thus, each run of a series was intended to represent the conditions at different stages of formation of the same pad.

The record of each run (see Fig. 12) contains the following information—(*1*) the position of the piston at the time when an increase in the consistency inside the light beam is first detected (hence the total length of the pad, as well as the mean pad consistency); (*2*) the consistency profile at the boundary between the pad and the slurry up to a point that the consistency exceeds some 1.5 per cent; (*3*) the position of the piston as a function of time. Only

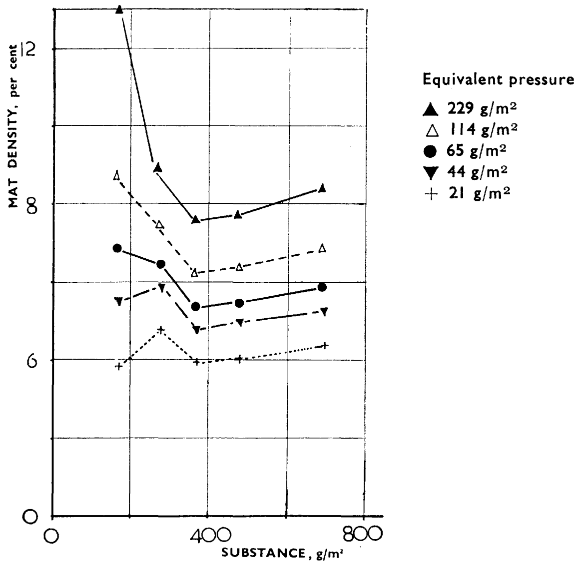


Fig. 10—Observed mean densities of the forming pad, NSSC pulp at 1 per cent consistency

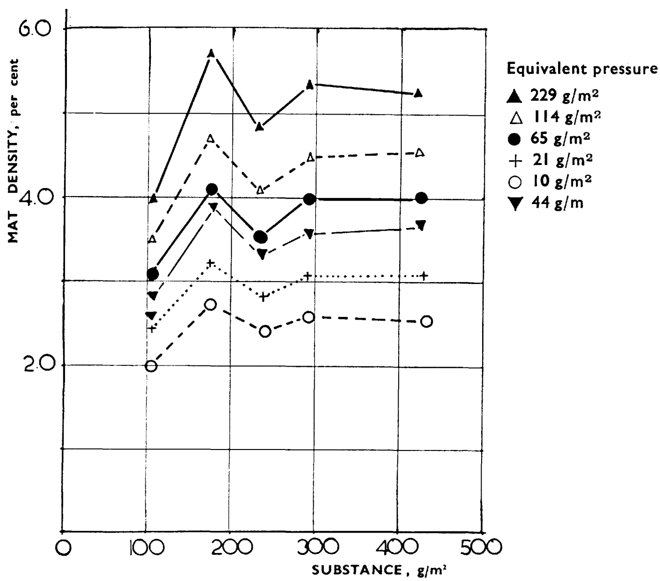


Fig. 11—Observed mean densities of the forming pad, NSSC pulp at 0.6 per cent consistency

the values of (*1*)—the lengths and mean consistencies of the pads—were calculated for each run. Fig. 11 and 10 show the averaged results of some 120 runs.

The computation of the profiles were very lengthy and only some 20 were made. The lengths of these 'tails' of the profiles varied 3–5 cm, without any apparent effect of the experimental conditions. Typical plots are shown in Fig. 13 and 14. Fig. 15 shows a plot of piston travel against time, as in a usual determination of the drainage rate.

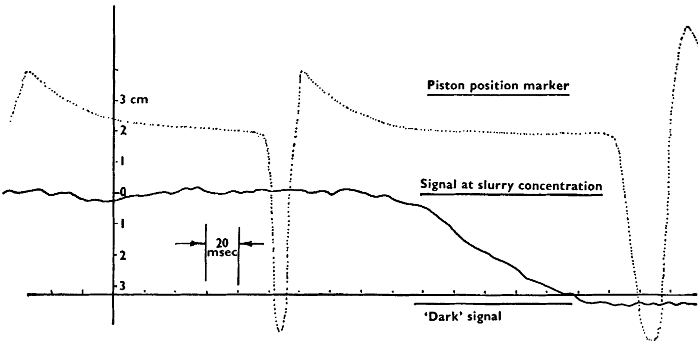


Fig. 12—Typical record of the drainage experiment

Visual observations of deposition of fibres on a papermachine wire—It would be extremely difficult to observe this phenomenon on a running papermachine. Instead, a model of the forming section of a Fourdrinier was constructed, the details of which will be published elsewhere. The principle* of it was to reverse the operation of the papermachine; instead of an endless wire belt passing over stationary rotating tube rolls, we had a set of rollers mounted on two chains rolling in contact with the underside of a length of wire cloth, stretched tightly on a fixed frame. The two systems, the machine and its model, differ only by a constant speed and should therefore faithfully reproduce the balance of forces.

A layer of cotton linters stock, containing a few dyed rayon fibres, was confined in a vertical cylinder having the fixed wire as the base and drained by means of the roller system at a simulated machine speed of 300 ft/min. The movement of the dyed fibres was filmed through a periscope-like device of small dimensions,[56] inserted in the stock just above the wire. Again, little evidence of thickening was observed, even with this stock; the fibres approach the mat with little disturbance, then collapse on to it.

* It was suggested to us by Mr Forgacs, then at PPRIC, Montreal

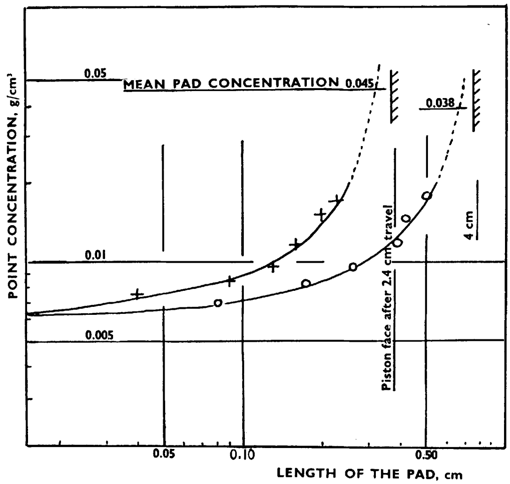


Fig. 13—Observed density profiles at the boundaries of the forming pad, NSSC pulp at 0.62 per cent consistency, piston pressure 70 g/cm^2

Discussion of the results

THE MAIN findings concern the existence and the size of the diffuse thickening zone at the boundary of the mat. From Fig. 13 and 14 it appears that such a zone existed, but was only 3–5 mm long. This is considerably longer than a fibre length or indeed a floc size. A few runs with a long-fibred, low-yield soft sulphite pulp, which tended to flocculate badly, produced even shorter depths of the diffuse zone, suggesting that the effect was genuine thickening, not flocculation or edge effects. Theoretical calculations (Fig. 9) also predict the existence of a diffuse zone of approximately the same length.

It may be concluded that, even at these forming pressures, the thickening zone, although it clearly exists, is considerably smaller than the usual depth of stock at the beginning of drainage on a Fourdrinier table. It follows that, even under such conditions, paper would form mainly by filtration until some three quarters of the water had drained.

The other results serve mainly to check the validity of the experiment and/or of the theory. Thus, Fig. 15 shows that the time for the piston to travel a given distance is proportional to a power of that distance—this relationship was used in Parker's computations[(55)]—and to a negative power of the pressure, as found by Wahlström & O'Blenes.[(46)] The mean pad density remains sensibly constant throughout the run (Fig. 10 and 11), again as

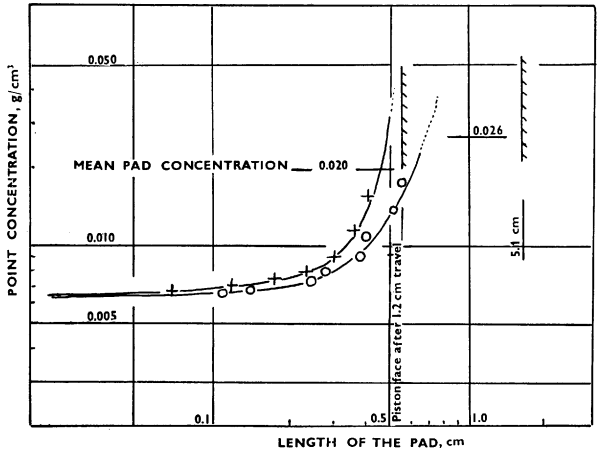


Fig. 14—Observed density profile at the boundaries of the forming pad, NSSC pulp at 0.6 per cent consistency, piston pressure 15 g/cm²

stated[46] and is calculated in Fig. 9. The large spread of these results is probably due to treating different runs as if they were stages of the same run —in fact, owing to the small volume of the apparatus and small size of the light beam, large variations in both the average and the recorded local consistency would occur from run to run.

Significance of the findings

PART 1 demonstrates that many different papers have an essentially layered

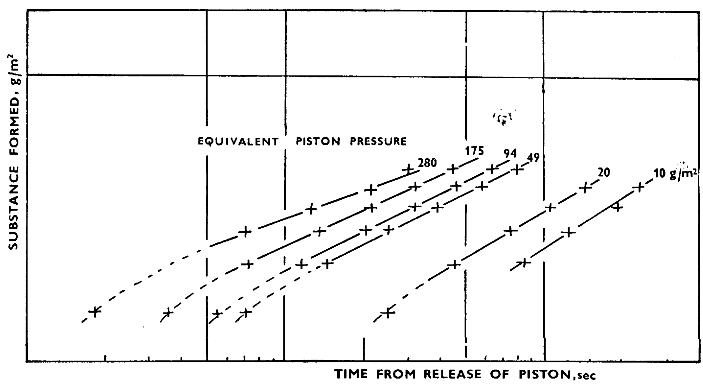


Fig. 15—Drainage record of the NSSC pulp

structure, Part 2 that they are formed mainly by a filtration-like process. In our view, the first finding is a necessary consequence of the second, given the fact that all papermaking operations are limited to a consistency range of, say, 0.05–1.0 per cent. Consider the formation of 100 g/m^2 paper at 0.5 per cent consistency, with fibre length of 0.1 cm. The depth of stock on the wire at the beginning of formation is then 2 cm. This means that, even if all fibres were vertically oriented at the moment of formation, the sheet could still contain some 20 layers of fibres, piled up on top of each other, provided drainage takes place by filtration. If, instead, thickening occurs to a large extent, then possibly (though not necessarily) the fibres in the top layer of the

TABLE 2—FIBRE LENGTH DISTRIBUTION, NSSC PULP AT 19°S.R.

Length of the observed fibre (× 94), cm	0–2	2–4	4–6	6–8	8–10	10–12	12–14	14–16
Number of such fibres	69	50	74	116	85	57	33	23

Mean fibre length = 0.075 cm

slurry could become intermingled with those at the bottom, resulting in a felted structure. We find that this is not the case in any papermaking process known to us; consequently, one would expect all papers, however made, to have a layered structure.

One would expect filtration to predominate even more strongly and layering be more pronounced in long-fibred papers and non-woven materials, because of disproportionately low consistencies necessary to ensure dispersion. Conversely, in order to produce an unlayered or felted sheet, one would have to form at very high consistencies; in the above example of 100 g/m^2 paper with 0.1 cm long fibres, a consistency of some 10 per cent would be needed!

Thus, the filtration mechanism of the formation of paper supplies both a simple explanation and a sufficient cause of the observed layered structure of paper and non-woven materials. When stated so, this finding appears so obvious as hardly to need experimental verification. Yet, such an explicit statement is necessary, we think, because it draws attention to a fundamental limitation of the papermaking process (or of any of its known variations) that must be overcome before a completely new kind of material is produced.

Perhaps the clearest statement of this challenge may be quoted from Prober[24] in a special issue devoted to problems of the technology of non-wovens, 'These materials have a layered structure and are easily delaminated.

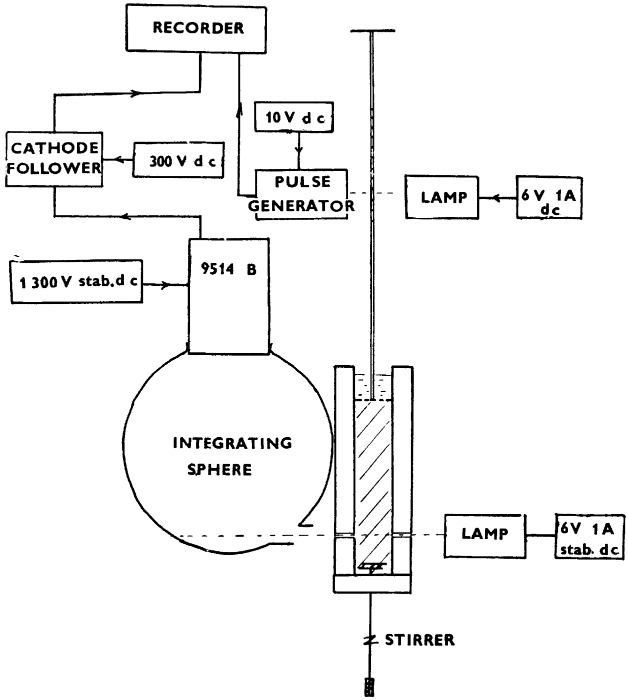


Fig. 16—Instrumentation of the drainage apparatus

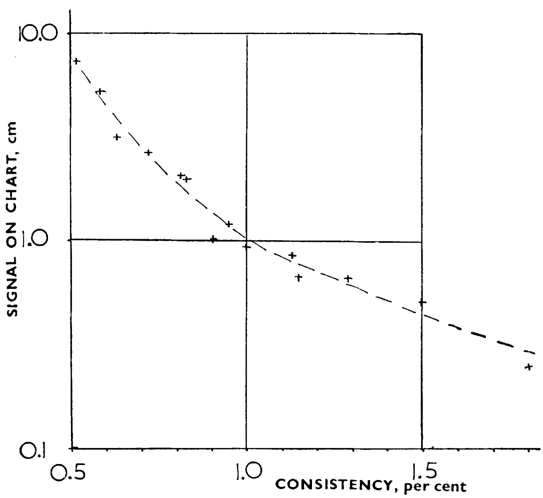


Fig. 17—Calibration curve of the drainage apparatus using NSSC pulp

The main task of their technology is to produce structures disoriented in their thickness.' At least, we know now a necessary, if not sufficient condition of success—to form the paper web at very high consistencies.

Acknowledgements

The work of C. Dodson formed the industrial training part of the Dip. Tech. course at the Brunel College, London; that of C. G. Skold was supported by a Wiggins Teape research student grant.

All three authors wish to express their thanks to the directors of Wiggins Teape Research & Development Ltd. for permission to publish this paper.

References

1. Kallmes, O. and Corte, H., *Tappi*, 1960, **43** (9), 737–752
2. Corte, H. and Kallmes, O., *Formation and Structure of Paper*, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 13–46; 351–368
3. Kallmes, O., Corte, H. and Bernier, G., *Tappi*, 1961, **44** (7), 519–528
4. Kallmes, O. and Bernier, G., *Tappi*, 1962, **45** (11), 867–872
5. Kallmes, O. and Bernier, G., *Tappi*, 1963, **46** (2), 108–114
6. Kallmes, O., Corte, H. and Bernier, G., *Tappi*, 1963, **46** (8), 493–502
7. Corte, H., *Consolidation of the Paper Web*, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1966)
8. Page, D. H. and Tydeman, P. A., *Formation and Structure of Paper*, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 397–413
9. Van den Akker, J. A., *Formation and Structure of Paper*, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 205–241
10. Page, D. H. and Sargent, J. W., RA/T/112 (B.P. & B.I.R.A., Kenley, 1964)
11. Meyer, R. and Wahren, D., *Svensk Papperstidn.*, 1964, **67** (10), 432–436
12. Underhay, G. F., *Tappi*, 1954, **37** (11), 547–553
13. Judt, M., *Das Papier*, 1958, **12** (21/22), 568–578
14. Groen, L. J., *Formation and Structure of Paper*, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 697–720
15. Forgacs, O. and Atack, D., *Formation and Structure of Paper*, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 721–747
16. Parker, J. and Mihi, W. C., *Tappi*, 1964, **47** (5), 254–263
17. Wood, R. L., *Tappi*, 1959, **42** (1), 14–17
18. Lehtinen, A., *Pulp & Paper Mag. Can.*, 1964, **65** (2), T61–T64
19. Finger, E. R. and Majewski, Z. J., *Tappi*, 1954, **37** (5), 216–224
20. Sauret, G., *ATIP Bull.*, 1961, (5), 395–403
21. Wrist, P. E., *Formation and Structure of Paper*, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 839–888
22. Ovechenko, N. G., *et al.*, *Text. Prom.*, 1963, **23** (9), 27–30
23. Vanzio, F. V. E., *Tappi*, 1960, **43** (1), 18–24
24. Prober, P. V., *Bumazh. Prom.*, 1959, **34** (9), 19–20

25. Pelgroms, J. D., *Papeterie*, 1951, **73** (1), 2, 5, 7, 9, 11, 13
26. Canadian Patent 656 145
27. British Patent 920 848
28. Buresh, F. M., Papers of American Assoc. for Textile Technology, (New York, 4th September 1963)
29. US Patent 3 120 463
30. British Patent 896 002
31. Burkhard, J. and Wrist, P. E., *Pulp & Paper Mag. Can.*, 1956, **57** (4), 100–118
32. Hendry, I. F., Mardon, J., Scruton, H. and Wilkinson, D. M., *Proc. Tech. Sect. B.P. & B.M.A.*, 1952, **33** (3), 603–653
33. Taylor, G. I., *Pulp & Paper Mag. Can.*, 1956, **57** (3), 267–273, 276
34. Bergström, J., *Svensk Papperstidn.*, 1957, **60** (1), 1–9
35. Ingmanson, W. L., *Tappi*, 1952, **35** (10), 439–448
36. Ingmanson, W. L. and Whitney, R. P., *Tappi*, 1954, **37** (11), 523–534
37. Ingmanson, W. L., Andrews, B. D. and Johnson, R. C., *Tappi*, 1959, **42** (10), 840–849
38. Ingmanson, W. L., *Tappi*, 1959, **42** (6), 449–454
39. Ingmanson, W. L. and Andrews, B. D., *Tappi*, 1963, **46** (3), 150–155
40. Ingmanson, W. L., *Tappi*, 1964, **47** (12), 742–750
41. Robertson, A. A. and Mason, S. G., *Pulp & Paper Mag. Can.*, 1949, **50** (13), 103–110
42. Meyer, H., *Tappi*, 1962, **45** (4), 296–310
43. Nelson, R. W., *Tappi*, 1964, **47** (12), 752–764
44. Manson, W. D. and Mardon, J., *Pulp & Paper Mag. Can.*, 1962, **63** (5), T259–T266
45. Meadley, C. K., *Can. J. Chem. Eng.*, 1962, **40** (6), 256–263; 1963, **41** (3), 95–103
46. Wahlström, B. and O'Blenes, G., *Pulp & Paper Mag. Can.*, 1962, **63** (8), T405–T417
47. Hisey, R. W., *Tappi*, 1956, **39** (10), 690–696
48. Tellvik, A. and Brauns, O., *Svensk Papperstidn.*, 1960, **63** (22), 803–812
49. Majewski, Z. J., *Formation and Structure of Paper*, Ed. F. Bolam (Technical Section, B.P. & B.M.A., London, 1962), 749–766
50. Cowan, E., *Pulp & Paper Mag. Can.*, 1937, **38** (2), 85–91, 199–200
51. Mason, S. G., *Tappi*, 1950, **33** (8), 403–409
52. Forgacs, O. L., Robertson, A. A. and Mason, S. C., *Fundamentals of Papermaking Fibres*, Ed. F. Bolam (Technical Section, B.P. & B.M.A., Kenley, 1958), 447–473
53. Burton, A., Project Report (Brunel College of Advanced Technology, London, 1963)
54. Wallis, J., Project Reports (Brunel College of Advanced Technology, London, 1964)
55. Parker, I. B., Maths. Dept., Brunel College of Advanced Technology, Private communication (January 1965)
56. Optec Ltd., 54 Upper Montagu Street, London, W.1

Discussion

Chairman—To me, one of the interesting things raised by this paper is the concept that the type of sheet produced depends to a very large extent on the consistency of the suspension from which it is produced in a way that is more fundamental than simply the general worsening of the degree of flocculation or non-uniformity that frequently arises from the use of high consistency. In other words, a sheet made from a very dilute suspension has a very highly layered structure; but, as the starting consistency rises, the final structure has an increasingly third dimensional component to fibre orientation and, in Radvan's terminology, becomes more felted. This changing character of web structure should affect both the strength and porosity of the sheet.

Mr D. H. Page—I would just like to go back to the 1957 symposium, when Frey-Wyssling drew on the board what he had never seen in fibrils to show that fibrils were not entangled (Fig. A). The point is that you cannot pick up any one of the fibrils, because one is over the other.

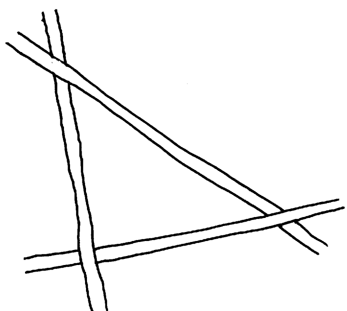


Fig. A

Since then, I have looked at a very large number of paper surfaces under the microscope and have often looked for this in fibres. I have never seen it on paper surfaces, which indicates that the fibres do not entangle in this way. The implication is, I think, that we can add to the list of properties that Radvan suggests would arise from an unlayered structure and we now come full circle to Prof. Cherry's address. If we could produce a surface structure like this, it would have a high abrasion resistance.