Preferred citation: J.R. Parker. Fundamental paper properties in relation to printability. In The Fundamental Properties of Paper Related to its Uses, *Trans. of the Vth Fund. Res. Symp. Cambridge*, 1973, (F. Bolam, ed.), pp 517–543, FRC, Manchester, 2018. DOI: 10.15376/frc.1973.2.517.

FUNDAMENTAL PAPER PROPERTIES IN RELATION TO PRINTABILITY

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Synopsis Printability can be satisfactorily related to fundamental paper properties only if the nature and the consequences of the interactions among paper, ink and printing press during and after impression are well understood.

Our present knowledge of these interactions is critically reviewed with particular reference to wetting, spreading, conformability, roughness, ink transfer, ink setting and print-through. The behaviour of low concentrations of oil in paper is considered in some detail.

Introduction

PAPER is a porous material and, for this reason, it is distinct from other printing substrates such as plastic films and tinplate and has certain properties in common with soils and sedimentary rocks. The characteristic properties of printing paper—its absorbency, permeability, reflectance, opacity, compressibility and roughness—are all to some extent related to its porous structure.

The printing properties of paper include not only these properties, but also such optical and mechanical properties as gloss, surface strength and dimensional stability. This paper is concerned mainly with properties related to the transfer and setting of ink on the paper surface. These are the crucial phases in the manufacture of a print. If the contact of the paper with the ink in image areas is incomplete, the print will be imperfect. If the paper surface is irregular or if the relationship between the porous structure of the paper and the characteristics of the ink is such that too much vehicle is withdrawn from the ink before it can set, then both the gloss and the density of the print will be impaired.⁽¹⁾ If the ink vehicle or solvent remains in the paper, it will link together the light-scattering surfaces of the smaller pores, diminishing the opacity and increasing print-through.

Homogeneous porous materials have a number of well-defined characteristics and it is convenient to refer to them at this point. Porosity ϵ is the

Under the chairmanship of Dr J. A. Van den Akker

ratio of the volume of voids to the bulk volume. The voids in paper are mostly interconnected and it is therefore possible to fill them with liquid, partially or wholly displacing the air present. The fraction θ of the void volume filled with liquid is called the saturation. The specific surface area A of a porous material is the ratio of internal area to bulk volume. The size of a pore of irregular cross-section may be indicated by its hydraulic radius m, the ratio of its cross-sectioned area to circumference. The ratio ϵ/A for a porous material gives the mean hydraulic radius of the pores. For paper of a softwood chemical furnish, the B.E.T. surface area might be 0.4×10^6 m²/m³, the porosity 0.5 and the mean hydraulic radius therefore $1.25 \ \mu m.^{(33)}$ The corresponding pore diameter would be four times this (that is, 5 μ m) comparable to the thickness of collapsed softwood fibres. For newsprint, it is about 2.5 μ m. The intrinsic permeability k of a saturated porous material is defined as the rate of flow of fluid of unit viscosity through a unit cube for unit pressure drop. It must be measured under conditions for which Darcy's law applies, that is, for laminar flow. For unsaturated porous materials, intrinsic permeability may be measured as a function of θ , together with the pressure Δp of the liquid in the partially filled pores and the intrinsic diffusivity D'. The significance of the latter will be explained below and it will merely be noted here that D' controls the imbibition and migration of liquids through unsaturated materials.

All the above properties are related to the structure of the porous material. In the case of paper, however, anisotropy, heterogeneity and surface roughness⁽²⁾ cause practical difficulties that in some cases are best overcome by direct measurement of the appropriate practical property per unit area of the material.

Ordinary pore size distribution measurements do not completely define a porous material, since they do not indicate the orientation, spatial distribution or frequency of interconnection of the pores, nor do they indicate the wetting properties of their surfaces, but they are not without value. The studies made by Chiodi & Silvy⁽³⁾ by a mercury intrusion technique that permitted the effects of clamping pressure and surface roughness to be observed are of particular interest. Distinct groups of pore sizes were identified with surface cavities, spaces between fibres and gaps between particles of coating pigment.

In studying the penetration of thin films of ink into paper, it is necessary to consider the relationship of ink pigment diameter to the size of surface pores, ink thickness in relation to pore diameter and the distance between pore openings in the surface and the depth of penetration in relation to the grain size of the paper structure. Such ratios indicate whether it is reasonable, for example, to neglect resistance caused by lateral flow across the paper surface, whether Darcy's law is strictly applicable to penetration or whether the paper surface may be treated as the termination of a set of parallel capillary tubes, as is assumed in a study by Tollenaar.⁽⁴⁾

One purpose of this paper is to draw attention to work in other fields that seems relevant to the study of printability. In particular, the movement of moisture through unsaturated soil is closely related to the migration of oil through paper, so reference will be made to techniques developed by soil physicists.

It is apparent from the nature of many publications of research on ink/paper interactions and printing phenomenon that too much reliance has been placed on an empirical approach, even in apparently fundamental investigations. To establish sound concepts, it is necessary to test hypotheses by *quantitative* comparisons of experimental results with theoretical predictions. This provides a far more valuable and searching proof of a theory than does qualitative reliance upon linear relationships and statistical correlations. The challenge of research on ink/paper relationships lies in devising experiments in which absolute comparisons are possible, despite the complexity of the materials involved.



Fig. 1—Penetration of liquid of finite contact angle into an irregular capillary; on right, the profile of the walls causes the meniscus to flatten

The fundamental printing properties of paper cannot be identified without a thorough understanding of printing phenomena. For this reason, some aspects of printing are discussed in detail to identify the controlling physical processes. Besides, rather than attempt a comprehensive review of the literature on the printing properties of paper, particular attention has been given to topics that seem of outstanding importance.

Wetting

IT is important that ink should wet the substrate to which it is applied. Unless the angle of contact is zero or the ink is of high viscosity and sets rapidly, a thin film will inevitably retract and break up into discrete droplets⁽⁵⁾ as observed, for example, by Carlsson & Lindberg.⁽⁶⁾

The penetration of a fluid under capillary action into a porous material is also affected by contact angle, because, whenever a penetrating fluid reaches a point in a capillary at which the bore expands rapidly, a finite contact angle will cause the meniscus to flatten so that capillary action ceases (Fig. 1). Even for zero contact angle, irregularities in the capillaries will diminish the radius of the meniscus and so retard penetration.

The Zisman critical surface energies⁽⁷⁾ of paper fibre constituents has been measured by Larsson,⁽⁸⁾ who found values for cellulose of $35 \cdot 5 - 41 \cdot 5 \text{ erg/cm}^2$ and for hemicelluloses, $33 \cdot 0 - 36 \cdot 5 \text{ erg/cm}^2$, also be Lee,⁽⁹⁾ who observed 36 erg/cm^2 for six samples of lignin. Liquids such as mineral oils for which the surface energy is about 30 ergs/cm² should therefore completely wet cellulose materials. This was confirmed by experiments with single fibres.⁽⁸⁾ Mineral fillers have high energy surfaces and should be wet by inks, but, as suggested by Karttunen,⁽¹⁰⁾ synthetic binders used in coatings may not be readily wettable.

Surface roughness is important. It is known from electron microscopy and from comparisons of the B.E.T. and observed surface areas⁽¹¹⁾ that papermaking fibres have rough surfaces. Larsson⁽⁸⁾ refers to Wenzel's equation (which suggests that contact angles of less than 90° would be lowered by surface roughness), but criticisms of this equation have been made by a number of workers. These are summarised by Johnson & Dettre,⁽¹²⁾ who showed that, although roughness causes hysteresis of the contact angle, it may also (if sufficiently great) lead to surface wicking of fluids with intrinsic contact angles of less than 90°. Surface wicking, which is the flow of liquid through the open-sided capillary channels provided by a rough surface, may well account for the migration of low concentrations of oil from news ink through newsprint.⁽¹³⁾ This view is supported by observations by Bascom⁽¹⁴⁾ of enhanced spreading rates caused by microscratches in polished metal and by Haynes⁽¹⁵⁾ on the flow of fluids over the surface of ground glass beads during the drainage of a packed bed.

Dynamic spreading and gravure printing

DURING the spreading of a drop of viscous liquid on a dry plane surface or the absorption of liquid by a porous material, the line of contact of liquid and air at the solid surface must advance, but the mechanism of this process is obscure. If it is assumed that the vapour pressure of the liquid is negligible and that no slip occurs at the interfaces, then mathematical analysis of the moving system leads to a singularity—that is, the tangential stresses at the liquid/solid interface increase without bound as the triple junction is approached—so that it appears that movement is impossible. This problem, which is relevent to the transfer of ink in gravure printing, has been studied theoretically by Chun Huh & Scriven⁽¹⁶⁾ and experimentally for glass and polymer fibres by Inverarity.⁽¹⁷⁾

Inverarity's results may be summarised for dynamic advancing contact angles ϕ up to $\pi/2$ by the equation⁽¹⁸⁾—

$$V = \frac{\gamma \varphi^3}{\beta \eta}$$
 (1)

where V is the velocity of the line of contact, η and γ the viscosity and surface tension of the liquid respectively and β is a dimensionless parameter that depends on the geometry of the system.

Consider the behaviour of ink in an incompletely filled gravure cell about 10 μ m deep, where initial contact is established between ink and paper only at the edges (Fig. 2). This line of contact will tend to move inwards towards the centre of the paper covering the cell, forming a bubble of air within the cell. Assume arbitrarily that ϕ is $\pi/4$, that β is 70, η is 0.1P and γ is 30 dyne/cm. Equation (1) then gives V = 1.0 cm/s. If the printing speed were 3.3 m/s, the nip width 10 mm and the width of the cell 120 μ m, then the contact line would



Fig. 2—Idealised stages of the contact between paper and ink in a gravure cell

have to advance 60 μ m in 3 ms at a speed of 2 cm/s in order that paper over the cell should be completely inked during contact with the cylinder. Thus, the rate of advance given by Inverarity's data is barely adequate to ensure complete printing of mid-tone cells under practical conditions. This appears to explain the characteristic 'doughnut' appearance of gravure mid-tones. Obviously, the absorbency and roughness of the paper together with the dimensions of the gravure cell would affect ϕ , which would change continuously as the liquid spread over the paper, but it is possible that a realistic model could be developed from this starting point. By incorporating ideas suggested by Banks,⁽¹⁹⁾ this could lead to an adequate theoretical treatment of ink transfer in the gravure process.

Capillary suction

IF A small volume of wetting non-swelling liquid, insufficient to fill the pore volume available, is absorbed by a porous material, it will tend to distribute itself so as to minimise the total surface energy of the system by flowing into the smallest accessible cavities. Flow will continue until the pressure throughout the fluid becomes uniform. Because the liquid wets the material, all its air/liquid interfaces will become concave and its pressure will be less than that of the surrounding air.

This negative pressure, conveniently referred to as capillary suction, is the driving force responsible for the absorption of oil from ink by paper and for the migration of oil within newsprint. Thus, when a non-drying ink film is applied to paper, flow of vehicle will continue until the capillary pressures in



Fig. 3

the two materials become equal. Thus, Larsson⁽¹³⁾ in experiments with newsprint and news ink, observed the retention of oil by the carbon black. In the case of traditional inks based on drying oils, chalking used to occur on coated papers, because vehicle was removed from around pigment particles. It is possible that ink piling also results from the capillary action of the paper.

Means of measuring the capillary suction of moisture in unsaturated soil were devised by an agricultural scientist in 1922⁽²⁰⁾ and, in 1947, a pressure membrane apparatus was described by Richards by which the suction/ moisture content relationships of soils could be explored over a wide range of pressures.⁽²¹⁾ In 1952,⁽²²⁾ a review of this and other techniques was published by Croney. In 1964, Robertson⁽²³⁾ used the pressure membrane method to study water in cellulose and paper.

The basis of the pressure membrane apparatus is illustrated in Fig. 3. A pressure cell is divided by a diaphragm or plate of porous material. This material must have a low contact angle with the liquid to be used and a pore size less than that of the material to be tested. One side of the cell is filled with the test fluid and the sample is placed in the other side in contact with the diaphragm. A pressure difference $(P_1 - P_2)$ is established across the diaphragm, sufficient to displace some liquid from the sample. Air will not pass through the diaphragm if its pore size is suitable.

The saturation of the sample changes until the capillary suction becomes equal to (P_1-P_2) . The liquid content of the sample is then measured. In practice, the results depend upon whether the liquid content is increasing or decreasing, therefore the saturation of the sample is usually taken through a cycle so that a hysteresis loop can be plotted.

Few, if any, capillary suction characteristics have been published for oil in paper. The results shown in Fig. 4–6 were obtained by the author using a low viscosity silicone fluid of 5 centistokes nominal viscosity and surface tension 19.7 dyne/cm. The results are restricted to differential pressures of less than 1 atmosphere because an open cell was used. Although hysteresis loops are given, it is the lower limb, for increasing oil content, that is of greatest interest in relation to the setting of inks. The upper limb of the hysteresis loop is related to the mercury intrusion curve. Similar hysteresis loops are found in sorption isotherms for porous materials in which capillary condensation occurs; their cause and interpretation have been widely discussed in relation capillary structure^(24, 25) and the behaviour of liquids in capillaries.⁽²⁶⁾

Scattering power, oil content and pore size

As THE oil content of paper is increased, both its scattering power and capillary suction fall.

Data on the scattering coefficient/oil content characteristics of newsprint



Fig. 4—Capillary suction characteristics for 53.8 g/m² newsprint with a bulk of 1.49 cm³/g; surface tension of fluid is 19.7 dyne/cm (The broken line shows the capillary suction estimated from optical measurements)



Fig. 5— Capiliary suction characteristic for a bond of 72.8 g/m² bulk 1.48 cm³/g



ig. 6—The capitary suction characteristic of a blade-coat supercalendered paper of 86.5 g/m²

has been given by Levlin & Nordman⁽²⁷⁾ and Aneliunas.⁽²⁸⁾ Levlin & Nordman noted the rapid fall in scattering coefficient on the addition of small volumes of oil to dry paper and found that the logarithm of scattering power was linearly related to oil content. They connected this fact with the number and size of pores filled with oil. Aneliunas found that, by plotting the logarithm of the ratio of scattering powers of oily and dry paper against saturation, a common relationship was obtained that applied to handsheets of groundwood fractions as well as to newsprint subjected to varying degrees of calendering. This, in view of Levlin's comment, suggests that such materials have closely related pore size distributions.

The influence of oil content on scattering power is of obvious importance in relation to print-through and this was the prime reason for the studies made by Levlin,⁽²⁷⁾ Larsson⁽¹³⁾ and Aneliunas.⁽²⁸⁾ The specific scattering coefficient of the surface of printing paper has been shown by Yule⁽²⁹⁾ to have a fundamental influence on the apparent sharpness of half-tone dots and lines, though Bergström⁽³⁰⁾ found functional relationships among scattering power and such printing properties of newsprint as its ink transfer characteristics. He suggested that these occurred because scattering power is a measure of the porous structure of paper. This idea has been further developed by Scallan & Borch.^(31, 32) It is therefore of interest to examine quantitatively the theoretical basis of some of the above observations.

Consider unit volume of material of porosity ϵ and specific surface A_o per unit volume containing a volume v of liquid that wets the material, but does not cause swelling. The saturation θ will then be given by the ratio v/ϵ . Let the pressure of the oil in the material be Δp above ambient and assume that all accessible pores with hydraulic radii less than m_{θ} are filled with liquid. Let A_{θ} be the total area of the solid/air and liquid/air interfaces. The quantity A_{θ} will lie between A_{θ} and zero.

Suppose that a further small volume of oil δv is added to the porous

material, causing the liquid in each pore to advance slightly, thus decreasing A_{θ} by an amount δA . If the diameters of the pores into which the liquid moves are large compared with the wavelength of light and if the refractive index of the liquid is the same as the porous material, then the specific scattering coefficient S_{θ} per unit volume will fall in proportion to A_{θ} . For paper, the proportionality constant F is roughly 20.⁽³³⁾ By definition⁽³⁴⁾—

$$m = -\frac{dv}{dA} \qquad . \qquad . \qquad . \qquad . \qquad (2)$$

and therefore

or

If γ is the surface tension of the liquid, then energy considerations lead to Carman's approximation⁽³⁴⁾ to the equation of Laplace⁽³⁵⁾—

$$-\Delta p = \gamma/m_{ heta}$$
 (5)

These equations are strictly valid only for pores with relatively smooth walls and constant cross-sectional areas.

Equation (4) may be used to estimate the cumulative pore size distribution of paper from the scattering coefficient/saturation characteristic; some results obtained by the author are given in Fig. 7. Equation (5) permits Δp to be estimated; the broken line in Fig. 4 was obtained in this way.



Fig. 7—Cumulative pore size distributions for three papers estimated from scattering power/saturation curves
A—Cast-coated B—Blade-coated C—Newsprint

Paper properties and printability

Aneliunas' correlation⁽²⁸⁾ between specific scattering coefficient and saturation is as follows—

From this and equation (4), it follows that—

$$m_{\theta} = \frac{\epsilon}{2.64 \ FS_{\theta}} \quad . \qquad . \qquad . \qquad (7)$$

or

$$m_{\theta} = \frac{\epsilon e^{2\cdot 64\theta}}{2.64 \ FS_{\theta}} \qquad (8)$$

Equation (7) is remarkable in its implication that the smallest size of pores in newsprint available to take up further oil is merely a function of porosity and the scattering coefficient by volume, whether or not the material contains oil already. If the probability density distribution of pore size by volume is defined by—

then from equation (8)—

$$\varphi(m_{\theta}) = 1/2.64 \, \mathrm{m}_{\theta}$$
 (10a)

for $z < m_{\theta} < z e^{2 \cdot 64}$ and

$$\varphi(m_{ heta})=0$$
 (10b)

for m < z or $m > z e^{2 \cdot 64}$ where $z = \epsilon/(2 \cdot 64 F S_{\circ})$

Fig. 8 shows a pore size distribution calculated for newsprint for which S_{a} is 600 cm²/g, the apparent density 0.67 g/cm³ and the density of solid matter 1.50 g/cm³. The distribution is highly skew, but similar to some pore size distributions determined by Levlin & Nordman⁽²⁷⁾ for calendered groundwood handsheets by mercury intrusion. The absence of pores with hydraulic radii less than 0.26 μ m (corresponding diameter 1.04 μ m) may be related to the wavelength of light. According to the elementary theory of multiple reflections between parallel plane surfaces.⁽³⁶⁾ however, the reflectance should first reach a maximum for normal incidence when the film thickness is only one quarter of a wavelength, that is, about $0.12 \ \mu m$. For papers containing synthetic silicate fillers of ultimate particle diameter much less than the wavelength of light such as those described by Mays⁽³⁷⁾ and Roehr⁽³⁸⁾ with diameters of about 0.03 μ m, the scattering power/oil content characteristic deviates from the form observed by Aneliunas. Small additions of oil to the dry paper have been found by the author to cause the scattering power to increase, although once the small pores provided by the filler have been saturated the scattering power falls in the normal way. The scattering of light by materials containing pores of size similar or less than the wavelength of light has been recently discussed by Gate,⁽³⁹⁾ who has shown that for suitable materials mean pore size may be estimated from the change in scattering with wavelength.



Fig. 8—A pore size distribution calculated for newsprint using Aneliunas' equation

Oil absorption tests and ink setting

MANY methods have been proposed for measuring the oil absorption characteristics of paper surfaces. The earlier methods⁽⁴⁰⁻⁴²⁾ permit measurement of the thickness of oil absorbed as a function of time, whereas later methods⁽⁴³⁻⁴⁵⁾ are concerned with the observation of the absorption of oil films of insufficient thickness to saturate surface coatings. In every case, the sample surface is flooded with liquid so that all pores communicating with the paper surface contribute to the absorption process, regardless of their relative sizes. This may not be the case, however, when oil is absorbed from an ink film after a print has been made.

For uncoated papers, the thickness of ink used in letterpress or offset printing is similar to the mean diameter of the pores in the paper. During impression, some ink is forced unchanged into the larger pores;⁽⁴⁶⁾ after impression, vehicle tends to drain into the smaller pores,⁽⁴⁷⁾ leaving the large pores open. Vehicle from ink on the paper surface can then drain only relatively slowly into the body of the paper by way of small pores or rough-ened fibre surfaces. It is this process, rather than imbibition as from a flooded surface with all the large pores full, that probably controls the diminution of set-off with time.

With coated papers, the retention of ink pigment and high molecular weight resins at the paper surface is an accepted part of the setting of an ink. This retention implies, however, that the flow of low viscosity components from the body of the ink into the paper will be restricted by the permeability ink film itself and, as drainage proceeds, the capillary or gel characteristics of the ink will also tend to retain vehicle. Thus, the pressure of ink vehicle at the paper surface must fall so that, even for coated papers, only the smaller pores may contribute to ink setting.

It is suggested therefore that, to obtain realistic results from oil absorption tests, it is necessary to make measurements under conditions that restrict the saturation at the paper surface. Alternatively, the rate of flow of oil through unsaturated paper in a direction normal to the plane of the sheets should be studied.

Imbibition and unsaturated flow

THE penetration of liquids into individual capillaries of uniform bore was considered in detail by Washburn,⁽⁴⁸⁾ who thus extended the earlier work of Lucas.⁽⁴⁹⁾ By assuming that porous materials could be treated as bundles of unconnected capillaries of differing diameter but of individually uniform radius, he deduced that the volume of liquid v penetrating into such a material in time t would be given by an equation of the form—

Although Washburn considered that this relationship would probably not hold when the cross-sections of the capillaries changed with length, it has been established by many workers, including Lucas,⁽⁴⁹⁾ that it holds with remarkable precision for the imbibition of non-aqueous organic liquids by strips of paper. Bickerman⁽⁵⁰⁾ has also commented on difficulties in applying simple capillary models to practical materials. Hsu⁽⁵¹⁾ found that the ratio of pore sizes estimated from the rates of penetration of varnish into paper under pressure and under capillary action differed by a factor of 20. He concluded that this was caused by the complexity of the paper structure. Tollenaar ^(47, 52) concluded that the change in liquid concentration along a strip of paper during imbibition could not be accounted for by Washburn's model and postulated one based on interconnected capillaries. Accounts of other capillary models may also be found in the literature, but it is evident that, unless a model is very detailed, it will be valid only in limited circumstances.

An alternative approach of considerable interest is used in soil physics. This dates from a publication by Buckingham in 1907 (see Swartzendruber⁽⁵³⁾), in which a quantity $D(\theta)$, later identified as a diffusion coefficient,⁽⁵⁴⁾ was introduced. The variation in this parameter with saturation was shown to

account for the concentration changes such as those observed by Tollenaar. The situation was summarised in 1965 by Denton⁽⁵⁵⁾ as follows.

"In any homogeneous porous material in which Darcy's law and the equation of continuity are obeyed, liquids will be absorbed at a rate determined by a square root relationship and the shape of the concentration profile will be independent of time. No assumption need be made as to the structure of the porous material and no particular model for a capillary system need be adopted. It is only necessary to assume that the capillary pressure and permeability are dependent only on the structure of the material and the liquid concentration."

Subject to the above assumption the flow q of liquid under capillary action per unit cross-sectional area is given by—

From the equation of continuity----

where

For boundary conditions appropriate to one-dimensional imbibition experiments, θ may be expressed as a function of a single variable $\lambda = x/t^{\frac{1}{2}}$. This is known as the Boltzmann (1894) transformation ^(53, 56) and it leads to the result—

$$\frac{\lambda}{2} = -\frac{d}{d\theta} \left[\mathbf{D}(\theta) \frac{\mathrm{d}\theta}{\mathrm{d}\lambda} \right] \qquad . \qquad . \qquad . \qquad (15)$$

which on integration gives-

$$\int_{\theta_0}^{\theta_1} \lambda d\theta = -2D \left[\frac{d\theta}{d\lambda} \right]_{\theta = \theta_1} \quad . \qquad . \qquad . \qquad (16)$$

Here, θ_0 is the initial uniform saturation of the sample, usually zero in capillary rise experiments and θ_1 is the saturation at any distance x from the fluid source at time t. Thus, from a plot of θ against λ , by integration of λ with respect to θ between the limits θ_0 and θ_1 and measurement of the slope at θ_1 , values of $D(\theta_1)$ may be found for the range of values of θ_1 given by the experiment.

The quantity $D(\theta_1)$ is generally referred to as the diffusivity of a given liquid/porous material combination. Although it is mathematically of the nature of a diffusion coefficient, this is not meant to imply that random

molecular processes are involved. Diffusivity depends on the laws of hydrodynamics and should therefore be proportional to $\gamma D'(\theta)/\eta$, where $D'(\theta)$ depends solely on θ and the structure of the porous material and will be referred to as the *intrinsic diffusivity*. It is interesting to note that this quantity



Fig. 9—Results of an experiment (duration 148 h) on the imbibition of a silicone fluid by newsprint in the machine-direction

The properties of the fluid were $\eta = 9.4$ cP, $\rho = 0.94$ cm³/g and $\gamma = 20.2$ dyne/cm



Fig. 10

has length as its sole dimension. Thus, it may be thought of as a measure of the effective size of the pores through which unsaturated flow takes place, just as an effective pore radius may be calculated from a form of the Lucas-Washburn equation.

Fig. 9 shows the results of a capillary rise experiment for the machinedirection of newsprint of which the capillary suction characteristic was given in Fig. 4. Using the method described, the intrinsic diffusivity was calculated and the results are shown in Fig. 10. This diagram also shows data for unsaturated flow in the Z-direction of the same paper. The latter results were obtained from two experiments in which about 10 discs of newsprint were pressed together at 200 kPa against a porous material in which fluid was maintained at a steady concentration. The rapid increase in diffusivity with saturation will be noted, as well as its lower value than that for the paper's machine-direction. The Washburn radii estimated from imbibition in the machine-direction and Z-direction were approximately 2×10^{-7} m and 3×10^{-8} m, respectively. They are apparently of the same order of magnitude as the intrinsic diffusivity of saturated paper.

Roughness

PRINTING roughness refers to the dimensions and the spatial distribution of gaps and contact areas occurring between paper and the surface from which the ink is transferred. It is affected both by the printing pressure and by the manner in which the pressure is applied and should therefore be measured under appropriate conditions. Roughness cannot be completely described by a single parameter, although appropriate single parameter measurements are of practical value for specific purposes. The 'cube root mean cube' gap measured under printing conditions by an airleak technique gives a useful indication of letterpress printability^(57, 58) and the frequency of non-contacting areas in areas in excess of the size of a gravure cell is claimed to correlate well with gravure speckle.⁽⁵⁹⁾

An adequate description of paper roughness requires the use of statistical distributions to indicate both the depth and width of depressions in the surface by considering the association between pairs of observations made short distances apart. The problem is similar to that of describing small-scale grammage variations.⁽⁶⁰⁾ It is possible that such information could be obtained by the magnetic scanning technique developed by Lyne⁽⁶¹⁾ or by measurement of the sizes of individual inked and uninked areas in prints made at a series of ink levels. Hsu ^(62, 63) used the observed relationships between inked area fraction and ink thickness on a letterpress plate to deduce the probability density distributions of the depths of surface depressions for a number of papers. All were found to be log-normal distributions, a fact probably

connected with Corte's observations ^(64, 65) that pore radii also tend to be distributed in this manner. As indicated under ink transfer, it was necessary to make some assumption to permit the maximum depth reached by the ink to be deduced from the ink thickness on the printing plate. Possible uses of such information include the calculation of the effect of roughness on the nonuniformity of ink film thickness, hence on print density, ⁽⁶⁶⁾ also the study of factors influencing gloss.

The Chapman instrument ^(67, 68) has been used in various forms to study the distribution and size of areas of paper in contact with a plane surface and has been elaborated by Bliesner⁽⁶⁹⁾ to permit such observations under dynamic loading conditions. The proximity of the paper surface to the glass prism required to bring about optical contact is usually quoted as half a wavelength of light, but a more precise consideration of the nature of such so-called contact is highly desirable.

Conformability

CONVENTIONAL printing processes all rely on adequate contact of the paper with the ink on the plate or blanket to affect transfer of the image. It is accepted that roughness of the paper can have a crucial effect on the perfection of the transferred image, especially with letterpress and gravure printing. The use of a deformable material on one side or other of the paper by which to apply pressure and so diminish the effective paper roughness seems a fundamental requirement for successful printing, although the significance of this feature of printing processes has received inadequate attention.

Some recent work by the author on the effect of backing hardness and paper caliper on measurements of Print-Surf roughness suggests that the term compressibility is ill chosen. It appears that the local compression of the paper is probably less important than its local flexing. The Print-Surf instrument^(57, 58) is provided with two standard backings-a hard backing made from granulated cork covered with polyester film and a soft backing, which is a litho blanket. The roughness of a wide range of thicknesses of a number of classes of paper were measured under a pressure of 2.0 MPa using each backing in turn. The ratios of the roughnesses found with the hard and soft backings were calculated for each paper and the results were plotted as shown in Fig. 11. For each class of paper, the reduction in roughness caused by the use of the soft backing was greatest for the samples of lowest caliper. If compression of the paper at its points of contact with the roughness measuring head were important, then a *decrease* in roughness would have occurred when a *hard* backing was used because of the concentration of pressure at these contact points. Moreover, this decrease in roughness would have been greatest for the thickest papers. Both deductions are, however, the contrary of what was

observed. It is suggested therefore that the soft backing improves the contact between the paper and the sensing head, because it distributes the clamping pressure more uniformly over the back of the paper. This causes local flexing of the paper around cavities between the paper and the roughness sensing head, which in turn considerably reduces their depth and therefore decreases the paper roughness (as illustrated in Fig. 12).



F g. 11—Effect of backing hardness and caliper of paper on Print-Surf roughness



Fig. 12—The flexing of printing paper induced by a soft backing

The same arguments may be applied to the effects of using a softer backing or greater impression pressure in a printing press, since the sensing head of the Print-Surf instrument is equivalent to a printing plate. The rigidity of paper is therefore important in the printing process. An appropriate empirical test for measuring the flexing of small areas of paper has been suggested by Paskiewicz.⁽⁷⁰⁾ It is suggested that the term *conformability* rather than compressibility be employed to describe the paper property that has been discussed.

Ink transfer theory and conformability

CONFORMABILITY is connected with ink transfer theory. For letterpress printing, the simplest form of ink transfer equation may be written—

This implies that, when a print is made using thickness x of ink on the printing plate, a constant quantity b is immobilised in or on the paper surface and a fraction f of the remaining ink also splits so as to remain on the paper, giving total transfer y. This holds only for films of ink that are thick enough completely to separate the plate from the paper throughout impression. Under such conditions, the entire impression pressure is transmitted to the paper by the ink film. This not only renders the immobilisation independent of the ink film thickness, but also ensures that the impression pressure is applied uniformly to both the troughs and crests of the paper surface.

For ink films that are of insufficient thickness to fill the surface cavities of paper, it is unlikely that the ink film itself would offer any appreciable support to the paper. In this regime, the asperities of the paper surface therefore tend to be flattened against the printing plate, as suggested by Parker⁽⁷²⁾ and Karttunen.⁽⁷³⁾ The surface volume of the voids is therefore less than for the thick ink regime described above. For the thin ink regime, up to the point at which coverage of the paper surface becomes substantially complete, the ink transfer relationship may be represented by the equation—

Here F(x) is called the coverage function. It is related to the probability density function of roughness g(h) as follows—

$$F(x) = \frac{1}{x} \int^{x'} h g h' dh \qquad . \qquad . \qquad . \qquad (19)$$

where x' is the depth reached by the ink in contact with the paper during impression. By making assumptions to relate x' to x, this equation was used by Hsu^(62, 63) to determine g(h).

A third transitional ink thickness regime, characterised by increasing pressure in the ink film, must be postulated to link the thin and thick ink film situations. In this regime, one must consider two factors—

- 1. The change in volume of paper surface cavities caused by the increasingly uniform support provided by the ink.
- 2. The increasing thickness of ink immobilised at the paper surface.

The general form of the ink transfer equation is usually written-

$$y = F(x)[b\varphi(x) + f\{x - b\varphi(x)\}]$$
 . . . (20)

Fetsko & Walker suggested that the transition function in this equation could be taken as-

thus implying that factor I above was negligible. Ichikawa *et al.*⁽⁷¹⁾ also make this assumption in their calculation of $\phi(x)$ from mercury intrusion data. In an experiment by Parker,⁽⁷²⁾ however, the thickness of free ink between plate and paper in the transitional regime was observed and found to increase somewhat with increasing ink thickness on the printing plate. Although it was not fully appreciated at the time, this is direct evidence of a change in paper roughness as postulated above, the omission of factor I by Ichikawa *et al.* may explain why their experimental results for $\phi(x)$ usually fell below their calculated curves.

Ink penetration under applied pressure

THE penetration of ink into paper during impression is important for several reasons. It has, in theory at least, a strong connection with the ink immobilisation parameter calculated from ink transfer theory. Although penetration removes ink from the paper surface and therefore diminishes set-off, it increases the ink requirement as well and detracts from print gloss. Ink penetration is also of importance in the printing of newspaper and other uncoated papers, because ink that penetrates into such papers during impression may do so as a whole,⁽⁴⁶⁾ carrying with it pigment that contributes significantly to print-through.⁽⁷⁴⁾ Ink penetration during impression is caused mainly by the printing pressure, Assuming an adequate supply of ink at the paper surface and viscous flow, the depth of penetration through individual pores would increase rapidly with diameter. Thus, the irregularity of penetration from place to place⁽⁷⁵⁾ may be explained by the pressure of occasional large pores.

The penetration of inks into uncoated paper was studied by $Hsu^{(46)}$ using a platen printing device, by which ink was forced against paper with pressures up to 10^8 dyne/cm² and dwell times down to 0.01 s. Hsu found that the penetration changed with viscosity and dwell time, as would be expected for laminar flow. The effect of pressure was similarly explained, except that a correction for compression of the paper was required. If Darcy's law were obeyed, then for a rolling nip it should be found that—

where p is the local pressure in the ink film over any point on the paper surface at any instant t during its passage through the nip, k and ϵ are the intrinsic permeability and porosity of the paper surface, η is the viscosity of the ink and B is the penetration. The line load L in the nip is given by—

where 1 is distance measured through the nip and, since we may write dp. dp/dt for dt, then—

where u is the printing speed.



Fig. **13**—The immobilisation parameter in relation to the velocity-viscosity product from data by Karttunen⁽⁷⁶⁾

(The parameter given here has been calculated using Karttunen's second modification of the Fetsko-Walker equation)



Fig. 14—The ink-splitting parameter as a function of the velocity-viscosity product from data by Karttunen⁽⁷⁶⁾

These results contrast strangely with those calculated from the Walker-Fetsko transfer equation or its modifications. Karttunen⁽⁷⁶⁾ recently published ink transfer parameters for four approximately Newtonian inks printed at 1 m/s and 4 m/s on to newsprint. Fig. 13 & 14 show the ink splitting factor fand the immobilisation b plotted against log ηu . Although b would be expected to change by a factor of six on the basis of equation (24), its variations are barely significant, but f changes progressively from 0.116 to 0.522. Similar results have been obtained in other studies.^(77, 78)

Various explanations have been put forward for this anomalous behaviour. That given by Taylor⁽⁷⁷⁾ may well be valid for ink transfer to coated stock. For uncoated papers that are of higher permeability and roughness, however, the complexity of the explanations required suggests that the validity of the transfer equation may be in doubt. In ink transfer theory, it has been assumed that the ink splitting factor does not change with the thickness of the ink on the printing plate, even though it changes dramatically with every other printing variable. This assumption will now be questioned.

An obvious justification of the assumption that the splitting factor is constant is that, particularly in the case of coated papers, the ink transfer curve is straight at high ink levels. For uncoated papers, however, the linear part of the curve tends to be ill-defined. Schaeffer,⁽⁷⁸⁾ for example, found it necessary to use ink film thicknesses as high as $35 \,\mu$ m in order to obtain linear sections of the curves where equation (17) was applicable.





Fig. 15—The suggested effect of air from porous paper on the splitting of ink films on exit from a press nip: the thin film above splits almost symmetrically, but the thicker film below splits near the paper surface

It has been pointed out by Truman⁽⁷⁹⁾ and by Hull⁽⁸⁰⁾ that the permeability of paper surfaces affects the magnitude of tack forces and, by implication, also

affects the manner in which ink films split. Fetsko⁽⁸¹⁾ suggested that air bubbles entering an ink film from a porous paper would encourage splitting to occur close to the paper surface. The distance between pores would be similar to the width of woodpulp fibres, say, 30 μ m. Such pores would have only a small effect on the splitting of a thin ink film, since they would be widely spaced in relation to the ink film thickness.



Fig. 16—Estimated variation of the ink-splitting factor with ink hickness on the printing plate for four inks of which the viscosities η are given, based on assumed values of the immobilisation constant b^{i} chosen to be consistent with Darcy's law

Moreover, the temperature gradients postulated by Taylor⁽⁷⁷⁾ as one of the causes of asymmetric splitting would die out more quickly if the films were thin, even though the ink films, used for determining ink transfer parameters often exceed 20 μ m. Under these circumstances, ruptures of the ink film originating from pores or surface cavities might be expected to affect the symmetry of splitting by air from pores would be influenced by the quantity of ink that has been forced into the paper by the impression pressure. If the expected penetration was high, that is, if the velocity-viscosity product was low or the printing pressure high, then the initiation of splitting by bubbles might be inhibited and the splitting factor might remain reasonably constant regardless of the changing ink film thickness.

If the assumption of a constant splitting factor is dispensed with, it becomes possible to interpret ink transfer curves in a manner that is consistent with Darcy's law. Curves given by Karttunen⁽⁷⁶⁾ for the above inks printed at 4 m/s will now be considered. It is necessary to assume a set of tentative values b' for the immobilisation constant that vary inversely as the square

roots of the ink viscosities. The values taken were rather less than those estimated in the usual way from the original Walker-Fetsko transfer equation. For any point on a transfer curve, it is then possible to calculate a value of f' by substitution of b' into equation (17). By trial, a set of values of b' can be found to give results as shown in Fig. 16 for the four inks in the thick film regime. Values for the ink viscosity b' and the standard ink transfer parameters are shown against each curve. It will be noted that f' falls as x increases and, as expected, the slope increases with viscosity. For very thin ink films, the splitting factors tend towards a constant value of about 0.43.

It is possible to estimate the intrinsic permeability k in equation (24) from the Bendtsen air permeability.⁽⁸²⁾ Karttunen did not quote this, but it is interesting to find by working back from the apparent density of the paper (0.753 g/cm³), the grammage (60.2 g/m^2) and the values assumed in the above analysis that the Bendtsen permeability is roughly 42 ml/min, which is low, but not unreasonable.

Thus, it seems possible to interpret ink transfer curves for uncoated papers satisfactorily in a manner that is compatible with elementary hydrodynamic principles if it is supposed that the ink splitting factor varies with ink thickness and the inks are Newtonian.

Values of b calculated from ink transfer curves are very sensitive to errors in f; if the assumption that f is constant is abandoned, therefore, present methods of interpreting ink transfer curves would require drastic revision. This may be necessary if ink transfer parameters are to be satisfactorily correlated with fundamental paper properties.

Acknowledgements

My grateful thanks are due to Dr J. M. Haynes, University of Bristol School of Chemistry and to Dr E. G. Youngs, Agricultural Research Council Unit of Soil Physics at Cambridge for their willing help and valuable comments on my inquiries into their fields of study, also to my colleague Mr J. B. A. Epton for his assistance at many stages during the preparation of this paper.

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Transcription of Discussion

Discussion

The Chairman The next contribution is rather unusual. It is the presentation of illustrations by Dr J. Mardon, who has studied the interrelationship between a number of factors that relate to the things we are talking about this morning and to other areas covered by the symposium. He has offered to supply copies of the charts to anyone who approaches him for them.

Dr Mardon is associated with Mr George Williams in describing these results as important. They state that it is not accurate to discuss the sheets produced by two-wire formers as if they were identical. Each two-wire former has its own characteristics. Results obtained during a paper structure investigation carried out over a long period are shown in Fig. L. It should be noted that the results of the Bel Baie former are for the Bel Baie mark 1.

Fig. M relates papermaking technology and those papermaking characteristics important in printing to sheet structure.

Fig. N illustrates uniformity of sheet strength and printing quality as related to furnish, system and papermachine characteristics.

Fig. O illustrates the interrelationship of surface and internal structure characteristics, which together determine the total sheet structure. The vital areas of study of sheet structure are given.

Fig. P-T illustrate typical sheet structural characteristics as described from sheet cross-sections.

Dr J. Marton My one comment is on the basic difficulty in printability assessment of fine papers and boards, namely, that the customer may have minimum requirements in most properties, but they have a preference. Their variety may make it very difficult (if not impossible) to express printability of a paper in one given number. Nonetheless, certain properties might be more generally sought after than others—one group of customers will prefer matt paper, others glossy paper. It seems usual for one common demand to be for high printed gloss. A good understanding of the printed gloss measurements or the development of printed gloss would be quite useful and generally applicable: it necessarily depends *inter alia* on the gloss of the unprinted

Under the chairmanship of Dr J. A. Van den Akker

Discussion



Fig. L—Table and drainage resistance for four types of two-wire former for four successive layers of the sheet

paper, on the smoothness of the surface and on the ink absorbency. Taking a homogeneous group of samples of coated board, our study indicated that, in a homogeneous group in which the variance comes mainly from production, both from changing formulation or conditions, 60–70 per cent of the printed gloss variance was caused by the variance of the unprinted gloss of the board and the rest by variance of surface roughness. This means that, although the level of expected printed gloss cannot be predicted from the unprinted gloss alone, it would be a challenging task—to keep production more uniform and it would be very useful if one could measure the unprinted gloss of the paper or the smoothness or both, continuously and directly on-line. Our greatest challenge is to maintain consistency of product quality.



Fig. M—Sheet structure as the focal point in a study of papermaking technology

Mr J. R. Parker Just before this conference, I spoke to Dr Larsson by phone and his comment on my paper was about the term printability. He said, 'It's very misleading, for it conveys the idea of just one number.' Printability is a term that we realise embraces a number of very different properties. It is impossible to express the idea of printability in any one number and paper must quite clearly be tailored to the requirements of certain customers. With newsprint, those running fast presses want one thing and those with old slow presses want another.

Dr H. G. Higgins I should like to refer to your conclusion or thesis that it is not merely the compression of the projecting parts of the paper surface, but also the flexing of the paper that is responsible for the decrease of roughness with pressure. Just before I left Melbourne, Dr Colley completed some Print-Surf experiments on a range of hardwood kraft handsheets beaten to 2 000, 4 000 and 8 000 rev in the PFT mill. We were prepared for the effect of roughness either to decrease or to remain constant (based on our experience with de Yong's profiler). What happened in fact was that the roughness decreased with beating in some cases, but there was a highly significant increase in the Print-Surf measurement with beating in others. I think this to

Discussion

be consistent with your thesis. We have not yet measured the elastic modulus of these papers, but it may be interesting to see how the trends with beating in relation to the roughness changes fit in with your hypothesis. I should mention also that compressibility measurements were taken as the ratio of the Print-Surf readings at 10 kgf/cm^2 to that at 20 kgf/cm^2 (soft backing, gloss side of the sheet). The compressibility figures ranged $1\cdot03-1\cdot08$.



Fig. N—Relationship of papermachine system characteristics to uniformity of sheet strength and printing quality

(Submitted addendum The variation of stiffness with beating—an initial increase arising from increase in Young's modulus, followed by a decrease because of reduction in thickness—was discussed by Gallay in his symposium contribution. Different positions of the stiffness maximum, with beating, could help to determine whether there is a rise or fall in Print-Surf roughness.)

Dr S. Karttunen I would like to speak about Joe Marton's comment on print gloss and paper gloss. It is quite natural when trying to explain the variations in print gloss to use the paper gloss as the first argument. It is

because paper gloss measures the paper smoothness indirectly in the same manner as print gloss is measured, since paper gloss is normally measured without using any pressure. Most current roughness or smoothness methods use pressure, which is another thing. Pressure comes into the picture at the printing nip, but it has nothing to do with paper or print gloss in its relaxed state after the ink film has been set and dried.



Fig. O—A breakdown of vital study areas when examining sheet structure

Mr J. A. S. Newman The oil penetration test measures the time it takes for an oil drop to penetrate the sheet completely. This time can be very different, depending on which side of the paper is tested; furthermore, it can be altered drastically by the action of the wet presses on the papermachine for instance, a straight press increases the time that the oil takes to go from the wire side of the sheet to the top side. Thus, this change may occur through a change in the structure of the paper and particularly of the surface of the paper.

Has this change in structure ever been observed physically in any other tests on the porous structure of the sheet or of the surface of the sheet? Secondly, has it ever been considered as a possible reason for differences in printability on the two sides of the sheet?

Prof. D. Wahren Mr Graeme Robertson at STF, Stockholm has performed a long series of experiments that relate to this question. In general, the more wet pressing used and the higher the temperature in drying and the higher the drying wire tension, the more dense the paper and the rougher its surface measured with a Bendtsen surface roughness tester and a Print-Surf tester using various pressures.



Mr B. Radvan It was suggested that conformability is concerned rather than compressibility of the paper. This takes place over very small distances comparable with the thickness of the paper, so it is not really stiffness as measured by modulus or by any flexural test, but something rather more complicated.

Mr Parker I was talking about flexing over distances of the order of 100 microns. I agree with you, but I do not know what is the appropriate property. There is an empirical test by Paszkiewicz (referred to in my paper) in which the paper is pushed through small holes to find how far it has gone. This looks relevant, but it is of course not at all fundamental.