Preferred citation: J. Silvy. The physical properties of paper in relation to its micro- and macrostructure – prepared contribution. In **Fundamentals of Papermaking**, *Trans. of the IXth Fund. Res. Symp. Cambridge, 1989*, (C.F. Baker & V. Punton, eds), pp 211–218, FRC, Manchester, 2018. DOI: 10.15376/frc.1989.3.211.

THE PHYSICAL PROPERTIES OF PAPER IN RELATION TO ITS MICRO- AND MACROSTRUCTURE

J. Silvy
Professor at l'Ecole Française de Papeterie et de l'Imprimerie, Institut National Polytechnique de Grenoble, URA CNRS No 1100, B.P. 65, F-38402 St Martin d'Hères Cedex

EDITOR'S NOTE: This prepared contribution was received after the Conference - no discussions follow.

ABSTRACT

The properties of paper are defined on two sampling scales, depending on whether the micro- or the macroscopic physical characteristics of the sheet are of interest. Two models are presented for the structure of homogenized paper, based either on the orientational distribution of the fibres or on that of the interfaces of pores and fibres. Orientational anisotropy arises on the scale of the fibre aggregates through shearing between flocs. Accurate tests were performed of the floc anisotropy, which influences the properties of paper on the size scale of a few mm^2.

INTRODUCTION

Paper possesses physical properties on its overall size scale. These are, for example, breaking strength under traction of paper strip intended for printing, bursting strength of paper for bags, tearing strength of a newspaper on unfolding, the degree of whiteness of a poster backing for a wall covering.

Certain properties of paper, in contrast, are measurable on the scale of a few mm^2, or less. This is the case for the properties of printability, where the printing quality depends on the surface topography of the sheet, and the spread from the point of printing to the surface should be less than 1 mm^2.

To investigate the properties of paper relevant to these two types of use, different sampling methods for the macroscopic and the microscopic structural analyses of the sheet must be used. The difference between these two domains is not however as clear-cut as might appear, since the structure of paper, seemingly continuous on the scale of the sheet, is the result of a juxtaposition of microdomains. The latter are flocs, i.e. the fibre aggregates that make up the sheet. To study the sheet of paper itself, one must resort to homogenisation, on account of the random macroscopic nature of paper texture. This process averages the properties that are discrete on the microscopic scale of texture, conferring on the paper an apparently homogeneous behaviour when its overall characteristics are measured.

This investigation describes the method used to homogenize the structure of the paper by means of geometrical models. The microscopic properties of the paper can thereby be transposed to the macroscopic scale.

1. TWO DIMENSIONAL MODEL OF THE PAPER SHEET

The structure of a paper sheet can be described precisely and analytically, if one assumes as a first approximation that the fibres are distributed in superimposed stratified planes in the thickness of the sheet. This hypothesis turns out to be valid in practice [1]. Within each of these planes the fibres are decomposed into a set of linear segments so as to eliminate the effect of their curvature. This arrangement allows one to define a dimensionless density of orientation, n_θ, which is equal to the proportion of segments contained in the angular interval $d\theta$ with respect to some given axis, usually taken in the cross direction of the sheet. This number density is, however, to be weighted by the mean directional length, $<l_\theta>$, of the segments along each direction. A new length-average orientation density is obtained [2]

$$n_{l\theta} = \frac{<l_\theta>}{\lambda} n_\theta , \tag{1}$$

where λ is the mean segment length in the paper, averaged over all angles, and $n_{l\theta}$ is defined such that

$$\int_0^\pi n_{l\theta} d\theta = 1. \tag{2}$$

A simple geometrical interpretation can be given for relations (1) and (2) from the following observation. Within the angular interval $d\theta$, $n_{l\theta}$ is proportional to the arc length ds of a contour in the plane of the paper obtained by cumulatively placing the straight-line segments end to end, while preserving the orientation of each. With this geometrical representation, relations (1) and (2) can be transformed by writing

$$n_{l\theta} = \frac{2}{\pounds} \rho_\theta \tag{3}$$

and

$$2 \int_0^\pi \rho_\theta \, d\theta = \pounds, \tag{4}$$

where $\rho_\theta = ds/d\theta$ is the radius of curvature of the contour at the point where the tangent lies along θ. $\pounds$ is the perimeter of the contour per unit surface area of the sheet, i.e. $2N\lambda$, where N is the total number of segments in all directions per unit surface area.

The shape of the curve defined by (3) and (4) is homothetic with that of the average pore in the paper. This can be defined by projection on the plane of the sheet if, for each direction in the plane of the sheet, the mean chord half-length of the pores $\langle g_\theta \rangle$ is drawn on either side of a centre O.

On account of this similarity with the average pore, the geometrical contour depicted in the structural model is called the "2D equivalent pore". Its shape is sufficient to define the orientational distribution $n_{l\theta}$ of the fibres in the plane of the sheet, starting with a knowledge of the radius of curvature ρ_θ over the contour of the equivalent pore.

For all porous structures it is found that

$$\langle g_\theta \rangle P_{l\theta} = \varepsilon = \text{constant} \tag{5}$$

and

$$\langle P_l \rangle = \pounds/\pi. \tag{6}$$

ε is the porosity of the sheet, and $P_{l\theta}$ the number of fibres intersected per unit distance along the direction θ in the plane of the sheet. $\langle P_l \rangle$ is the value of $P_{l\theta}$ averaged over all directions in the plane.

From relation (5) it can be seen that the polar diagram of $P_{l\theta}$ is the inverse of the equivalent pore curve. A simple practical method can thus be devised to identify the equivalent pore, starting from measurements of the number of fibres intersected per unit length, $P_{l\theta}$. This can be performed by image analysis of

the sheet of paper, or by using coloured fibres dispersed throughout the sheet.
Our observations are that ellipses represent the equivalent pore with good accuracy for a large number of industrial papers. This allows one to define the fibre distribution in the paper in terms of a single ellipticity variable, a/b.
This concept can be further generalized, since one can decompose any continuous periodic distribution $n_{l\theta}$ into a weighted sum of a finite number of modes. These are equivalent elliptic pores, with axes rotated with respect to a central mode [3].

2 APPLICATIONS OF THE 2 DIMENSIONAL MODEL OF PAPER

We have tested the use of this geometrical model of paper structure in investigating the rupture resistance under traction with zero span jaws, $R_{o\gamma}$, for different directions γ in the plane of the sheet.

The traction measurements provide a knowledge of the rupture force f of the paper fibres, as well as of the ellipticity, which gives an overall characterization of the orientation distribution $n_{l\theta}$ of the fibres in the sheet. One has [2]

$$R_{o\gamma} = \frac{\pounds}{\pi} f \int_{\gamma-\pi/2}^{\gamma+\pi/2} n_{l\theta} \, |\cos^3(\theta-\gamma)| \, d\theta \qquad (7)$$

and

$$\frac{a}{b} = 1.08 \, (R_{oSM}/R_{oST})^{0.45} . \qquad (8)$$

To apply formula 8, the resistance R_o is measured in the machine and the cross directions on paper samples. These are saturated with water so as to remove prestressing in the fibres induced during the drying process [2,4].

Another application is the evaluation of the elastic modulus under traction, E_γ, calculated for an arbitrary direction γ in the plane of the sheet, with the assumption that the paper is orthotropic [5]. E_γ is calculated from the following formula

$$E_\gamma = E_o^f \int_{\theta=-\gamma}^{\pi-\gamma} n_{l\theta} \cos^2(\theta+\gamma) \left[\cos^2(\theta+\gamma) + \frac{S_{12}}{S_{11}} \sin^2(\theta+\gamma) + \frac{S_{16}}{S_{11}} \sin(\theta+\gamma)\cos(\theta+\gamma) \right] [1 + \alpha\cos^2\theta + \beta\sin^2\theta] \, d\theta, \qquad (9)$$

where $n_{l\theta}$ is the length-average orientation density of the fibres, E_o^f the elastic modulus of a fibre that has been dried without constraint, S_{ij} are the flexibility matrix coefficients, and α and β are coefficients that describe the effects of the preconstraints induced in the fibres upon drying [6].
Further applications of this geometrical two dimensional model are possible for investigations of the properties of paper, in particular the dimensional stability of the sheet.

3 THREE DIMENSIONAL MODEL OF PAPER

In addition to the forces that are exerted purely in the plane of the sheet, certain properties of paper depend on the characteristics of the texture in relation to the thickness. A three dimensional model is therefore required that no longer relies on the orientation density of the fibres, as if these were infinitely thin lines lying in the plane of the sheet. The model must be based instead on the density of the surface elements of the fibre walls, which constitute a set of microfacets at the limit of the pores and fibres in the sheet.

Stereological analysis of thin slices of paper, obtained by isotropic sampling, gives the number of directional intercepts $P_L(l,m,n)$ between the pore interfaces and the fibres traversed per unit distance in a direction (l,m,n) in space. This in turn gives the projection of the specific area S_V of the porous texture on each plane in space . One has

$$P_L(l,m,n) = S_V, \tag{10}$$

projected on to the plane normal to the direction (l,m,n). Also

$$\langle P_L \rangle = S_V/2 \tag{11}$$

where $\langle P_L \rangle$ denotes the mean value for all directions in space of the intercepts $P_L(l,m,n)$ [7].
The left-hand surface, whose projections in all the isotropic planes of space obey eqs. 10 and 11, characterizes in a one-to-one fashion the orientation density distribution of the pore interfaces and of the fibres in the texture. This surface is homothetic with that defined by the volume of the mean pore of the texture, just as described in paragraph 1 above, but in the present case for three dimensional space. For this reason this geometrical model is also designated "3D equivalent pore".
It was checked that ellipsoidal equivalent pores provide a satisfactory characterization for several porous textures, including paper. The only parameters required in defining the model are the ellipticities

a/b and c/b and the surface area of the ellipsoidal model, which is taken to be equal to the specific surface area S_V of the sheet.

4 APPLICATIONS OF THE THREE DIMENSIONAL MODEL OF PAPER

The three dimensional model allows investigation of a large number of properties that depend on the porosity and the anisotropy of the sheet. Examples are: surface roughness of the sheet, optical properties of the paper using transmitted and scattered light in the fibrous texture, pore size distribution, and directional permeability to fluid flow through the sheet.

5 MICROSTRUCTURE OF THE SHEET: ANALYSIS OF THE CAUSES OF ORIENTATION OF THE FIBRES IN THE PAPER

In the manufacture of the sheet, the pulp concentration upon impact with the wire of the machine is too high for the fibres to move freely in the suspension. The fibre movements are thus confined to the interior of the flocs. These are composed of fibre aggregates whose size depends on the pulp and on the turbulence in the head box.

If there is a difference between the speed of the pulp jet at the outlet of the head box and the speed of the draining wire(s), then a velocity gradient dV/dz is established in the direction of the thickness Oz at the beginning of the formation of the sheet. This gives rise to shear which deforms the flocs in the suspension, at the same time as causing them to be stretched along the direction of motion of the paper machine. The deformation of the flocs alters the orientation of the fibres inside the aggregates and generates the overall anisotropy of the paper sheet.

It follows from this analysis that the difference in velocity between the two sides of the sheet is one of the principal factors causing fibre orientation in the paper. The anisotropy of the paper is in fact determined by the absolute value of the velocity gradient in the thickness of the sheet at the initial formation stage. This explains the observed symmetry of fibre orientation in a sheet of paper, compared with an isotropic paper, for values of the pulp jet speed either greater or less than that of the wire of the paper machine.

Structural isotropy on a paper machine is, however, not obtained when the jet speed equals that of the wire, except in the case of a laboratory arrangement in which the components of velocity in the plane of the sheet are zero. It should be recalled that the flocs are already subjected to shear in the outlet lips of the head box. Furthermore, for double wire machines, backflow reduces the speed of the pulp jet in the formation zone compared with the values attained at the head box outlet.

6 FIBRE AGGREGATE ANISOTROPY IN THE PAPER SHEET

On order to prove that the orientational anisotropy originates in the flocs, the microscopic structure of different papers was analysed, using samples of a few mm^2 or less in surface area. The papers were made in a Fourdinier machine with different degrees of orientation. The pulp consisted of 100% bleached coniferous Kraft, refined at 25°SR, and the average paper basis weight was 50 $g.m^{-2}$.
Various analysis techniques were tried. Firstly, samples of diameter 3.3 mm were selected in more and less flocculated areas of the paper, by visual observation in transmitted light. The fibre orientation of these samples was measured directly by image analysis of photomicrographs of the structure, enlarged 183 times by a scanning electron microscope. Also, the traction strength using zero span jaws was measured for paper micro test-strips, each of width 5 mm [8].
The interpretation of these tests by the two different methods confirms that anisotropy in the paper is already clearly present on the size scale of the flocs. Occasionally the value of the ellipticity a/b is different for the flocculated and less flocculated areas, but it is, on average, equal to that of the sheet, estimated by using coloured fibres with samples separated by several cm in the paper. Hasuike et al., using image analysis, have also shown that orientation differences exist between fibres in flocculated and non flocculated areas [9].
The above result was confirmed by optical measurements performed using the fibre orientation test equipment belonging to the Paul Lippke GmbH company at Neuwid, F.R.G. [10]. This apparatus, which detects laser light scattering from paper, directly measures the ellipticity a/b of the 2D equivalent pore and characterizes the fibre orientation, following the model expounded in paragraph 1 [11].
The impact area of the laser beam on the paper sheet has a cross section of 0.2 mm, hence allowing direct sampling of ellipticity in zones of differing flocculation. A sliding average of seven successive measurements was made at intervals of 0.1 mm. In these conditions the surface being tested by laser scattering appears elliptical, with a surface area of approximately 1.8 mm^2, i.e. an average diameter of 1.5 mm. These measurements confirm that paper already displays its orientational anisotropy on the microscopic scale of

the flocs, and that the mean value of the ellipticity fluctuations detected at this microscopic level is the same as that observed at large scale using coloured fibres, or that calculated using the zero span jaw traction measurements of water-saturated test strips.

7 CONCLUSION

The texture of paper can be described by geometrical models: the 2D and 3D equivalent pores which account for the properties of the sheet, either in its plane or in three dimensions.
The properties of paper arise from its microstructure. The so-called "formation factor" of the sheet must allow the transfer efficiency in the physical properties of the sheet to be established between the microscopic and the macroscopic level.

8 ACKNOWLEDGEMENT

The author thanks the Lippke GmbH company, its director Dr A. KOHL and his collaborators for their kind permission to use the laser apparatus for fibre orientation measurements.

9 REFERENCES

1. MARK,R.E. Handbook of Physical and Mechanical Testing of Paper and Paperboard, Vol.2 p.306-310.
2. SILVY,J. Thèse de Doctorat d'Etat, Université de Grenoble and Institut National Polytechnique de Grenoble 1980.
3. SILVY,J., HERVE,H. ATIP vol.36 N°6 7 June-Sept. 1982, p.343-359.
4. KORAN,Z., SILVY,J. PRUDHOMME,R.E. TAPPI Jnl. vol 69 n°5 May 1986 p.126-128.
5. MORIN,V. Thèse nouveau Doctorat. Institut National Polytechnique de Grenoble, Dec. 1988.
6. PERKIN,R.W. Handbook of Physical and Mechanical Testing of Paper and Paperboard Vol.1 p.65.
7. UNDERWOOD,E.E. Quantitative Stereology Addison Wesley Pub. Co. 1970.
8. ARCONDARA, E. Project study: diploma of Ingénieur de l'Ecole Française de Papeterie 1984.
9. HASUIKE,M, JOHANSSON,P.A., FELLERS,C and TERLAND,O. International Paper Physics Conference Preprint, Mont Rolland, Quebec, Sept. 14-18 1987.
10. JOENSBERG,CH., KOHL,A., KLEPP,E., LANDMARK,P. Preprints 22nd EUCEPA Conference, Florence, 6-10 Oct. 1986.
11. COUTY, D. Project study: diploma of Ingénieur de l'Ecole Française de Papeterie 1988.