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Bivariate Normal Thickness-Density Structure in Real Near-Planar Stochastic Fiber Networks

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We present the analysis of experimental data that supports the recently presented hypothesis that the relationship between local areal density and local thickness in planar stochastic fiber networks may be described by the bivariate normal distribution. Measurements of the local averages of areal density and thickness have been made on experimental fiber networks with differing degrees of structural uniformity. The experimentally determined variance of local density at the 1 mm scale is in excellent agreement with that calculated from the theory. Also, the use of the bivariate normal distribution to describe the relationship between local areal density and local thickness measured in complete sampling schemes is appropriate for both near-random and clumped networks.

KEY WORDS: Stochastic fiber networks; density; porosity; clumping; network geometry; paper.

1. INTRODUCTION

The structure of near-planar stochastic fiber networks and their planar projections has been the subject of many studies, a comprehensive review of which is given by Deng and Dodson.[1] Much of this work has considered the special case of planar random fiber networks. Analytic expressions for the distribution of local averages of areal density, in networks with fiber centres distributed according to a two dimensional Poisson process and

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uniformly distributed fiber axis orientations, were derived by Dodson.[2] Miles[3] obtained the results that the distance between fiber crossings in such networks is described by the negative exponential distribution and the mean number of sides per polygon is four; this result was used by Corte and Lloyd[4] to derive the probability density function for equivalent pore radii in random fiber networks. Typical commercial examples of near-planar stochastic fiber networks such as paper, nonwoven fabrics, glass fiber filter mats, etc. have thickness of the order of one tenth or less of a mean fiber length; so though we deal with a 3-dimensional structure, many features of importance are accessible though projections onto a plane.

Real fiber networks have nonrandom structures, largely as a consequence of interactions between fibers in the suspensions from which the network is formed. The resulting near-planar structures can be studied by a complete sampling scheme of uniform square zones and this gives rise for each choice of zone size to a bivariate distribution of the local averages in zones for areal density and thickness, which in the sequel we shall represent by the random variables $\tilde{\beta}$ and $\tilde{z}$, respectively. In the random case fibers are by definition placed independently; then the parameters of the bivariate distribution are known analytically for arbitrary rectangular fibers[2] and so provide reference cases for real data. The alternatives to random are on the one hand a "clumped" structure where fibers have aggregated together during the forming process as if mutually attracted somewhat, and on the other hand a "smoothed" structure wherein fibers have acted as if mutually repelled somewhat. In a density bitmap representation of the structure a clumped case has more evident contrast than a smoothed case; this is because the variance of local averages is greater than in the random case when clumping occurs and less than in the random case when smoothing occurs.[1,4] We consider the variability of the *local average* values of given properties since these are observable and dependent on the scale of inspection or *zone size*; typically, the variance of local average density and thickness is a monotonically decreasing function of zone size.

The probability density function for equivalent pore radii in clumped or *flocculated* networks was derived by Dodson and Sampson[5,6] extending the result for Poisson processes obtained by Corte and Lloyd.[4] The degree of clumping is important in commercial fiber networks as it affects, for example, the deformation of a network under strain[7,8] and the flow of fluids through the network.[9] The properties of random fiber networks, as discussed above, are widely used to provide reference structures against which the structural characteristics of real structures may be compared, and their differences quantified.

Recently, we presented a model for the distribution of local porosity in two and three dimensional fiber networks;[10] if local areal density, $\tilde{\beta}$ and local thickness, $\tilde{z}$ are distributed according to the bivariate normal distribution then the variance of local porosity, $\mathrm{Var}_x(\tilde{\varepsilon})$ is given by,

$$\mathrm{Var}_x(\tilde{\varepsilon}) = \frac{1}{\rho^2}\left(\frac{\bar{\beta}}{\bar{z}}\right)^2 \left(\frac{\mathrm{Var}_x(\tilde{\beta})}{\bar{\beta}^2} - \frac{2\,\mathrm{Cov}_x(\tilde{\beta},\tilde{z})}{\bar{\beta}\bar{z}} + \frac{\mathrm{Var}_x(\tilde{z})}{\bar{z}^2}\right) \tag{1}$$

where $\bar{\beta}$ is the average network areal density, $\bar{z}$ is the average network thickness, ρ is the fiber density and $CV_x(\tilde{\beta})$ and $CV_x(\tilde{z})$ are the coefficients of variation at a scale of inspection x of areal density and thickness respectively. Now, the local and mean porosities are defined by Eqs. (2) and (3) respectively:

$$\tilde{\varepsilon} = 1 - \frac{\tilde{\beta}}{\tilde{z}\rho} \tag{2}$$

$$\bar{\varepsilon} = 1 - \frac{\bar{\beta}}{\bar{z}\rho} \tag{3}$$

and for $\tilde{\varepsilon} \approx \bar{\varepsilon}$ we have the approximate expression

$$\mathrm{Var}_x(\tilde{\varepsilon}) \approx \frac{1}{\rho^2}\left(\frac{\bar{\beta}}{\bar{z}}\right)^2 |CV_x(\tilde{\beta})^2 - CV_x(\tilde{z})^2| \tag{4}$$

A limited set of experimental data from the literature[11] was used in ref. 10 to test the assumption that the relationship between the local thickness and local areal density of the network could be described by a bivariate normal distribution and hence allow, through estimation of the variance of local porosity, estimation of the variance of local density. Agreement was satisfactory, but the original data did not include values for the covariance of areal density and thickness and hence only Eq. (4) could be tested.

Here we discuss the use of the bivariate normal distribution to describe the relationship between the local averages of thickness and areal density. Data is presented for a range of experimental near-planar fiber networks with differing degrees of clumping; the data is compared with the model presented in ref. 10 via expressions following directly from Eqs. (1) and (4) and developed in the sequel.

2. VARIANCE OF DENSITY

Consider the coverage of points by fibers. The thickness of the network at a given point is determined by the number of fibers covering that point, their thickness and their vertical separation. For a given zone, it is the local averages of these values which determines the local average thickness. The relationships between the coverage of points, the distribution of local averages of areal density of zones and their dependence on fiber morphologies are well established.[1,2]

From the Central Limit Theorem, we expect the distributions of the local averages of areal density and thickness to be Normal and this is confirmed by experimental measurements made on paper.[1,11] Denoting the local and global averages of variables by placing a tilde ~ and a bar ¯ respectively over the variables, we note, that density, porosity, thickness and areal density are related, by definition, by the expressions:

$$\tilde{c} = (1 - \tilde{\varepsilon})\,\rho \qquad \text{and} \qquad \tilde{\beta} = \tilde{c}\tilde{z} \tag{5}$$

$$\bar{c} = (1 - \bar{\varepsilon})\,\rho \qquad \text{and} \qquad \bar{\beta} = \bar{c}\bar{z} \tag{6}$$

where $\tilde{c}$ and $\bar{c}$ are the local and global averages of density respectively. Thus, as density is the complement of porosity, it follows directly from Eq. (1) that at a given inspection zone size x, the variance of local density, $\mathrm{Var}_x(\tilde{c})$ is given by $\rho^2\,\mathrm{Var}_x(\varepsilon)$ using (5) directly, or equivalently

$$\mathrm{Var}_x(\tilde{c}) = \left(\frac{\bar{\beta}}{\bar{z}}\right)^2 \left(\frac{\mathrm{Var}_x(\tilde{\beta})}{\bar{\beta}^2} - \frac{2\,\mathrm{Cov}_x(\tilde{\beta}, \tilde{z})}{\bar{\beta}\bar{z}} + \frac{\mathrm{Var}_x(\tilde{z})}{\bar{z}^2}\right) \tag{7}$$

Following ref. 10, if $\tilde{c} \approx \bar{c}$, then

$$\mathrm{Cov}(\tilde{\beta}, \tilde{z}) \approx \frac{\bar{\beta}}{\bar{z}}\,\mathrm{Var}(\tilde{z}) \tag{8}$$

$$\approx \frac{\bar{z}}{\bar{\beta}}\,\mathrm{Var}(\tilde{\beta}) \tag{9}$$

and substitution of Eqs. (8) and (9) in (7) yields the approximate expression

$$\mathrm{Var}_x(\tilde{c}) \approx \left(\frac{\bar{\beta}}{\bar{z}}\right)^2 |CV_x(\tilde{\beta})^2 - CV_x(\tilde{z})^2| = \rho^2\,\mathrm{Var}_x(\tilde{\varepsilon}) \tag{10}$$

in agreement with the case for (4).

The use of the bivariate normal distribution to describe the relationship between the local averages of thickness and areal density is seemingly appropriate as these are Normally distributed and proportional to each other; the constant of proportionality being the local network density, which we expect to be distributed according to some random process.

3. EXPERIMENTAL

Samples of stochastic fiber networks were formed using natural cellulose fibers obtained from different woods treated by two different processes, thermo-mechanical (TMP) and chemical (CS); samples were made also from a 50:50 blend of the two types. Samples were formed by filtration of a suspension over a standard woven wire fabric in a British Standard Handsheet Former; this equipment conforms to international standards for forming paper in the laboratory and is described in ref. 12. The fibers were chosen for their different morphologies and these are summarised in Table I. The linear density of a fiber is defined as its expected mass per unit length, so at a given mean areal density, networks formed from the CS fibers will have more constituent fibers per unit area than those formed from the TMP fibers.

The degree of clumping was altered by forming at different mass concentrations in the suspension and by allowing time for the fibers in suspension partially to sediment before filtration. Both mechanisms allowed increased potential for fiber interaction in suspension and hence increased nonuniformity in the formed network. It should be noted that one set of sheets formed from each fiber type and the blend were formed using the mass concentrations and sedimentation times described in ref. 12; these conditions are known to produce networks with a distribution of mass density at the 1 mm scale close to that of a random fiber network formed from the same constituent fibers.[1] The degree of fiber clumping in suspension, induced through the range of experimental conditions, therefore produced manifestly non-random networks with a broader distribution of local areal densities than their corresponding random networks.

For each sample, the local averages of thickness and areal density of 1 mm square zones were measured within a 50 mm × 50 mm area and the samples marked to allow zone by zone comparison. Thickness was measured using a laser triangulation device[13] and areal density using a calibrated β-radiation device. Full experimental details have been presented in a recent PhD thesis[14] and will be reported fully elsewhere.

Table I. Properties of Fibers Used to Prepare Sheets

	Mean width, $\bar{\omega}$ μm	Mean length, λ mm	Linear density, δ g m^{-1} × 10^4
TMP	36.5	1.98	2.22
CS	38.7	2.41	1.16

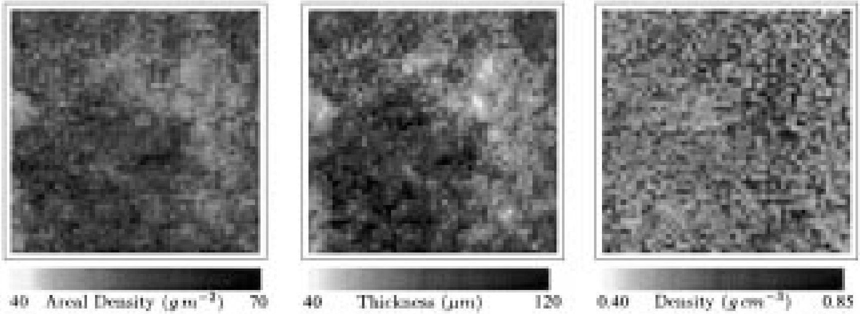

Fig. 1. In-plane distributions of areal density, thickness and density. Example shown is for a network formed from CS fibers with a mean areal density of $45\,g\,m^{-2}$. Each image represents the same $50\,mm \times 50\,mm$ zone.

4. RESULTS

Knowledge of the local averages of areal density and thickness for each 1 mm square zone in a given sample allowed direct determination of the mean and variance of each property and their covariance for each sample. Also, the local average density of each zone could be calculated directly. Sample density plots for local averages of areal density, thickness and density are shown in Fig. 1; the data shown is for a $45\,g\,m^{-2}$ network formed from CS fibers with an intermediate level of structural nonuniformity. A plot of the local averages of areal density and thickness for the same sample are shown in Fig. 2; the coefficient of determination for a linear regression on these data was 0.763 and this is typical of all the

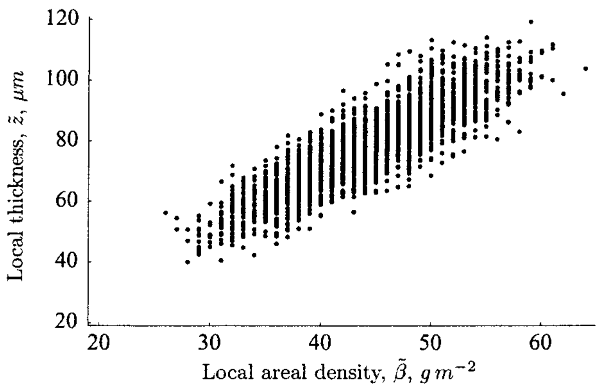

Fig. 2. Example of relationship between local averages of areal density and thickness. The bivariate normal distribution is justified by a linear regression between local thickness and local areal density.

samples tested. The use of a bivariate normal distribution in our model is justified by this good linear regression found between local thickness and local areal density.

The variance of local density obtained using Eq. (7) is plotted against the experimentally determined value in Fig. 3; the broken line has unit gradient. Regression analysis on the data yields a slope of 0.92, an intercept of 0.98 and a coefficient of determination of 0.974; the regression is represented by the dotted line in Fig. 3. The excellent agreement between the two sets of data suggests strongly that the relationship between local areal density and thickness is well described by the bivariate normal distribution.

The same experimental data is presented in Fig. 4, though the values on the ordinate are calculated from the approximate expression given by Eq. (10); again, the broken line has unit gradient. The dotted line represents a linear regression on the data; the gradient is approximately one, the intercept on the ordinate is -5×10^{-4} g^2 cm^{-6} and the coefficient of determination is 0.727. The underestimation of the variance of local density using Eq. (10) results from the assumption that $\tilde{c}\approx\bar{c}$ and is therefore expected. Remarkably, the linear regression between local thickness and local areal density persists even to clumped cases of very much higher variances than for a Poisson process. This means that the bivariate normal model applies well over a broad range of real structures, not only to the random network case.

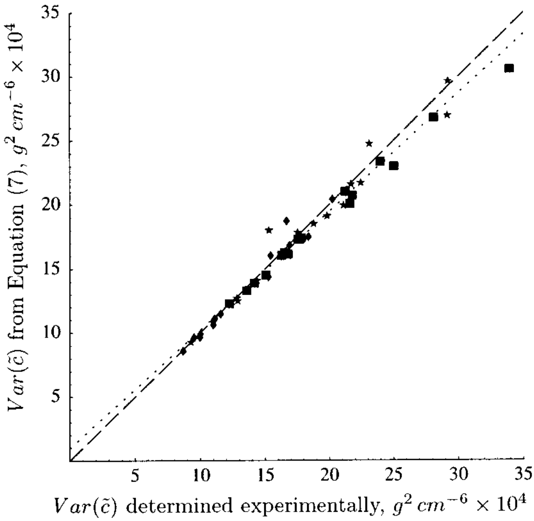

Fig. 3. Variance of local density as given by Eq. (7) plotted against that determined experimentally. Key: Diamonds—TMP; squares—CS; stars—50:50 blend. The broken line has unit gradient; the dotted line represents a linear regression on the data.

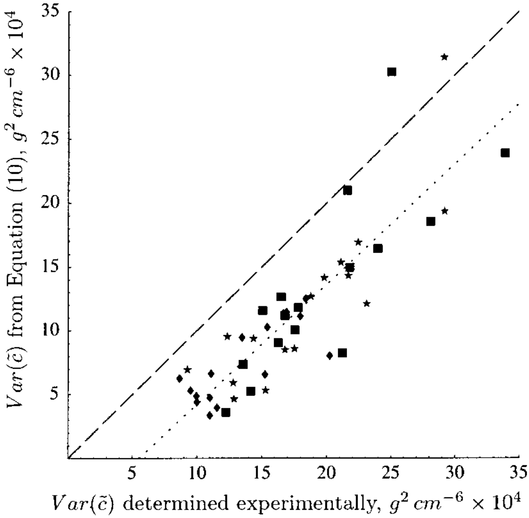

Fig. 4. Variance of local density as given by Eq. (10) plotted against that determined experimentally. Key: Diamonds—TMP; squares—CS; stars—50:50 blend. The broken line has unit gradient; the dotted line represents a linear regression on the data.

5. CONCLUSIONS

Our results suggest that the relationship between the local areal density and local thickness in these near-planar stochastic fiber networks indicates a linear regression that is well described by the bivariate normal distribution. Such a property would be expected for any Poisson process of extended objects, but we have shown that it persists in the important non-random case of clumped structures.

If local areal density and local thickness are measured independently in a structure and their covariance is unknown, then we provide an approximate expression for the variance of local density.

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NOMENCLATURE

$\tilde{\beta}$	Local areal density	$g\,m^{-2}$
$\bar{\beta}$	Mean areal density	$g\,m^{-2}$

$\tilde{c}$	Local mass density	g cm^{-3}
$\bar{c}$	Mean mass density	g cm^{-3}
δ	Fibre linear density	g m^{-1}
$\tilde{\varepsilon}$	Local porosity	{ }
$\bar{\varepsilon}$	Mean porosity	{ }
λ	Fibre length	m
ρ	Fibre density	g m^{-3}
ω	Fibre width	m
x	Zone size	m
$\tilde{z}$	Local network thickness	m
$\bar{z}$	Mean network thickness	m

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