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LETTER

Inter-fibre contacts in random fibrous materials: experimental verification of theoretical dependence on porosity and fibre width

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The mechanical, optical and transport properties of industrially manufactured stochastic fibrous networks, such as paper, non-woven fabrics and fibrous filters, are strongly influenced by the extent of contact between fibres. The classical reference structures for modelling these materials are random fibre networks with fibre centres distributed according to a two-dimensional point Poisson process and with uniformly distributed fibre axes. The number of fibres covering a point in the plane of support of the network is a discrete random variable called *coverage*, c, with Poisson probability,

$$P(c) = \frac{\bar{c}^c e^{-\bar{c}}}{c!} \quad \text{for } c = 0, 1, 2, 3 \ldots \tag{1}$$

The earliest work considering the statistical geometry of random fibre networks was derived for paper [1] and this has given rise to a significant body of literature, which has been recently reviewed [2, 3]. The development of new heterogeneous fibrous materials for use in fuel cells and as scaffolds for tissue engineering has provided new applications for structural models of stochastic fibrous materials, see e.g. [4, 5], and we note that the theory is applicable to networks with arbitrary scale features [6].

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Our interest here is the extent of contact between fibres. When the mean coverage, $\bar{c}$, is less than about 1, the number of inter-fibre crossings per fibre is a function of fibre length λ (m), fibre width ω (m) and mean coverage [1]:

$$n = \frac{2}{\pi}\frac{\lambda}{\omega}\bar{c} \quad \text{for } \bar{c} \le 1 \tag{2}$$

The fraction of the fibre surface in contact with other fibres, termed the *fractional contact area* (FCA), is a function of mean coverage only [7]:

$$\Phi = 1 - \frac{1 - e^{-\bar{c}}}{\bar{c}} \quad \text{for } \bar{c} \le 1 \tag{3}$$

Real networks typically have mean coverage of 10 or more and hence a given pair of fibres may be obscured from contact by the influence of nearby fibres. For such networks, Kallmes et al. [8] derived expressions for the expected number of contacts per fibre by considering the superposition of several two-dimensional networks and including fibre flexibility as a variable to weight the probability of contact between fibres in nearby layers. The theory was difficult to verify however since the number of layers was not well-defined and meaningful measures of fibre flexibility are experimentally difficult. Recently, Sampson [9] noted that for modelling purposes the mean coverage of a layer could be defined by the porosity of the network, ε, such that the number of layers in a network of a given mean coverage was well defined and the influence of fibre flexibility was implicitly accounted for. The FCA of a network with infinite coverage is [9],

$$\Phi^{\infty} = \frac{2}{\bar{c}} \sum_{c=1}^{\infty} \frac{c-1}{c} P(c) + (1-\varepsilon)^2 \sum_{c=1}^{\infty} \frac{1}{c} P(c), \quad (4)$$

where $\bar{c} = \log(1/\varepsilon)$ such that Φ^{∞} is a function of porosity only. Note that the second term on the right hand side of Eq. (4) differs from that given in [9] by a factor of 2; the influence on the predicted FCA is typically small though Eq. (4) makes the theory precise.

For a network of finite mean coverage $\overline{c^*}$, the fibres in the outer surfaces can contact others on one side only, so FCA is given by [10],

$$\Phi = \left(1 - \frac{1}{\overline{c^*}}\right) \Phi^{\infty}, \quad (5)$$

where for networks of mean mass per unit area, $\bar{\beta}$ (g m^{-2}), formed from fibres of mass per unit length δ (g m^{-1}), we have $\overline{c^*} = \bar{\beta}\,\omega/\delta$.

Sampson proceeded to show that if FCA was known, then for fibres of known length and width, the expected number of contacts per fibre was given by

$$n = \frac{\Phi}{\Phi^1} \quad (6)$$

where Φ^1 is the FCA of a thin network calculated from Eq. (3) with mean coverage, $\overline{c^*}$, determined by setting $n = 1$ in Eq. (2).

Equation (6) is not easily verified because the extent of inter-fibre contact is difficult to measure in anything but the thinnest networks. Elias [11] counted the number of contacts per fibre in low-weight networks of glass fibres and showed correlations between the data obtained and the solid fraction of the network, $(1-\varepsilon)$. Experimental estimates of FCA obtained from image analysis of cross-sections of paper embedded in resin [12, 13] exhibit a dependence on porosity consistent with Eq. (4) [9]. These techniques are not readily applicable to the quantification of the number of contacts per fibre because it is difficult to obtain a section along the full length of any fibre. This was recently addressed by He et al. [14] using confocal scanning laser microscopy (CLSM) and we extend their analysis here.

It is easy to show that for a network of randomly oriented, rectangular fibres of width ω and infinite length, crossings have expected area $\pi\omega^2/2$. For a network with FCA, Φ, formed from fibres with width, ω (m) and length, λ (m), the expected total area of contacts on a given fibre is

$$A = 2\Phi\lambda\omega, \quad (7)$$

where the coefficient 2 accounts for crossings above and below that fibre. The expected number of crossings per fibre is therefore,

$$n = \frac{2A}{\pi\omega^2} = \frac{4}{\pi}\frac{\lambda}{\omega}\Phi \quad (8)$$

Equation (8) provides an approximation to Eq. (6) and is more open to intuitive interpretation. We note the very reasonable dependencies suggested, i.e. the number of contacts per fibre is directly proportional to the length of a fibre and to its FCA; the inverse proportionality to fibre width arises through the influence of fibre width on the coverage of a network with a given number of fibre centres per unit area.

We have analysed the data of He et al. [14] to obtain values for the number of crossings per fibre for fibres with a range of known widths and lengths formed into networks with known porosity. Full experimental details are provided in [14], so here we give only a summary. Sheets weighing 60 g m^{-2} were formed using a laboratory device designed to produce paper samples with structures similar to those obtained industrially [15]. All sheets were made from radiata pine fibres extracted in a laboratory pulping process and processed to yield fibres with different lengths and widths; some sheets were pressed to reduce porosity independently of fibre dimensions.

Prior to forming sheets, the fibres were stained with Acridine Orange so that they would fluoresce under the CLSM. Samples were embedded in Struers Epofix resin and cross-sections were exposed by polishing the block with abrasive papers. A 14 mm length of each cross-section, consisting of 70 consecutive frames, was examined under a CLSM using a 60× oil-immersion lens. Fibres that were imaged along their major axis were identified and the number of fibres in contact with each of these was counted. To aid identification of contacts, images were thresholded and binarised. A contact was counted between each pair of fibre surfaces not separated by black pixels and classified as either a full or partial contact. This is illustrated by example in Fig. 1 where fibres 2 and 3 make full contacts with the fibre of interest, fibre 1 makes a partial contact, and fibre 4 makes no contact. For each of the samples, fibre cross-sectional area and shape were also measured. This involved selecting a minimum of 200 individual fibres for imaging at the cross-section surface and 10 μm under the surface. The angle at which each fibre cut the surface was then calculated from the change in position of the centre of mass of the fibre. Each of the measured cross-sections was then corrected by this angle to obtain the true cross-section

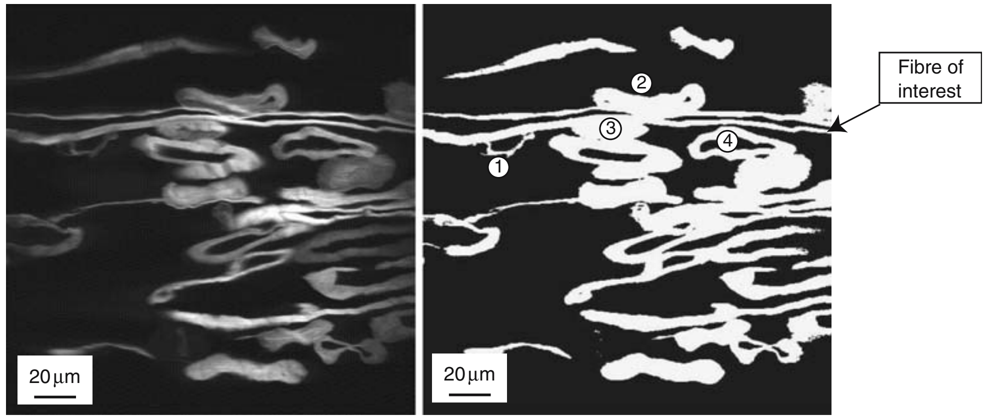

Fig. 1 Cross-section image before (left) and after thresholding and binarisation (right). Fibre 2 and 3 in (**B**) make two full contacts, fibre 1 makes a partial contact, and fibre 4 is not in contact with the fibre of interest

and this corrected cross-section was then fitted with a rectangular bounding box, as shown in Fig. 2. The ratio of the fibre wall area to the bounding box area was called the fill factor. Further details are given in [16]. The 95% confidence interval of each measured value was usually less than ±4%.

To calculate the number of contacts using Eq. (8), we must calculate first the FCA using Eqs. (4) and (6) and so require a measure of the inter-fibre porosity. From inspection of Fig. 1 we observe that a fraction of the network voids exists within fibres. Accordingly the total porosity, as estimated from

$$\varepsilon = 1 - \frac{\rho_{\mathrm{network}}}{\rho_{\mathrm{solid}}}, \tag{9}$$

where ρ_{network} and ρ_{solid} are the densities of the network and solid phases, respectively, would overestimate the porosity. Recently, Batchelor and He [17] presented a linear correlation between an experimental estimate of the FCA of paper and the sheet density weighted by the fill factor, f, given by the ratio of the cross sectional area of the cell wall to the area of the bounding rectangle with sides ω and t, and illustrated in Fig. 2. This weighting accounts for the volume of the network rendered inaccessible by the presence of a fibre so the accessible porosity is

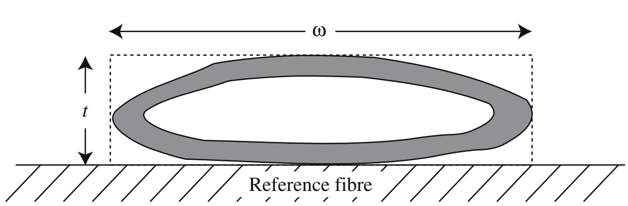

Fig. 2 Determination of the fill factor, f, which is given by the ratio of the cross sectional area of the cell wall to that of the bounding rectangle with sides ω and t [16]

$$\varepsilon^* = 1 - \frac{\rho_{\mathrm{network}}}{f\rho_{\mathrm{solid}}}. \tag{10}$$

We use this value of porosity to calculate Φ using Eqs. (4) and (5) and hence the expected number of contacts per fibre via Eq. (8).

Equations (6) and (8) are arrived at by dividing the expected total area of contact on a fibre by that of a full contact. By definition, partial contacts exhibit a smaller area than full contacts so the theory will underestimate the total number of contacts observed in experiments. If the fraction of a full contact represented by a partial contact is a non-skewed random variable then, on average, two partial contacts are equivalent to a full one. Accordingly, we determine the number of equivalent contacts from our experiments as the sum of the number of full contacts and half the number of partial contacts.

The experimental data are summarised in Table 1 and the calculated and measured contacts are compared in Fig. 3; these include also the data of Elias [11]. Since the networks Elias studied were very thin, only the first term on the right hand side of Eq. (4) was used to calculate the FCA and hence the number of contacts; similarly, each contact was classified as a full contact. Clearly there is good agreement between theory and measurement, as indicated by the broken line with unit gradient. From Table 1, it can be seen also that the total number of contacts does not fall linearly with paper sheet density; we attribute this to the fact that increasing density not only increases the number of full and partial contacts but converts some of the partial contacts to full ones also. The lowest density paper tested (192 kg m^{-3}) still exhibited around 40 contacts per fibre, but almost all of these were partial. Any further reduction in sheet density would rapidly reduce the total number of contacts since almost no full contacts would remain; accordingly,

Table 1 Fibre and network properties

	Fibre properties				Network properties				Number of contacts per fibre			
	Width (μm)	Length (mm)	Coarseness ($\times10^7$ kg m^{-1})	Fill factor	Mean coverage	Density (kg m^{-3})	Porosity[a]	FCA	Experiment: Full	Experiment: Partial	Experiment: Effective	Model
This study	30.7	3.1	3.0	0.548	6.2	509	0.38	0.396	35.5	38.1	54.5	51.6
	34.5	2.5	3.1	0.524	6.7	541	0.31	0.470	29.8	26.3	42.9	43.9
	33.1	2.1	3.0	0.550	6.7	434	0.47	0.314	23.5	23.8	35.4	25.4
	30.2	3.1	2.9	0.428	6.3	218	0.66	0.169	9.9	31.0	25.4	22.3
	34.1	3.1	2.9	0.448	7.2	392	0.42	0.371	22.7	42.8	44.1	43.6
	36.4	3.1	3.0	0.507	7.3	651	0.14	0.621	40.8	46.1	63.9	68.2
	28.0	3.1	3.2	0.459	5.3	193	0.72	0.128	7.2	33.4	23.9	18.2
	32.2	3.1	3.4	0.490	5.7	306	0.58	0.217	20.8	40.4	41.0	26.9
	34.2	3.1	3.4	0.535	6.2	574	0.28	0.490	40.1	50.5	65.3	56.4
Elias [11]	7.2	2.3	–	–	–	–	0.97	0.016	7.3	–	7.3	6.2
	7.2	2.3	–	–	–	–	0.95	0.024	11.4	–	11.4	9.7
	7.2	2.3	–	–	–	–	0.95	0.027	12.6	–	12.6	10.9
	7.2	1.1	–	–	–	–	0.94	0.030	6.5	–	6.5	5.8
	7.2	1.1	–	–	–	–	0.93	0.037	6.5	–	6.5	7.1
	7.2	4.6	–	–	–	–	0.94	0.029	21.6	–	21.6	23.1
	12.9	2.3	–	–	–	–	0.93	0.033	7.2	–	7.2	7.6

[a] For this study, porosity calculated using Eq. (10) assuming ρ_{solid} = 1.5 kg m^{-3}; for data of Elias, porosity calculated from solid fraction given in [11]

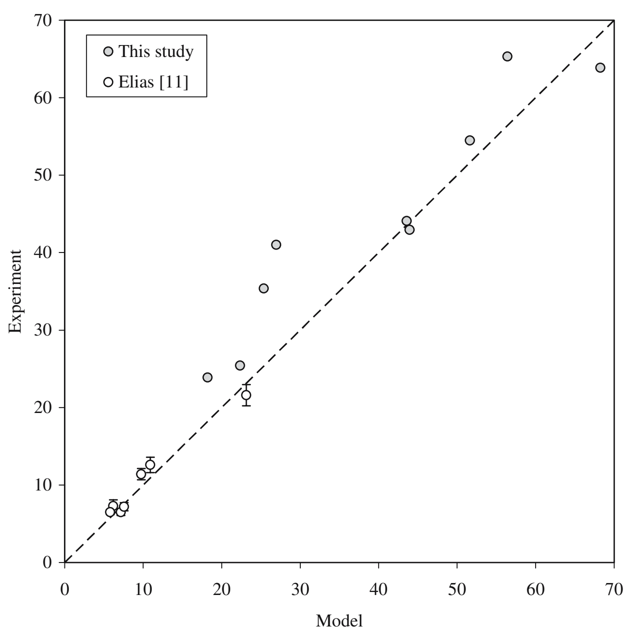

Fig. 3 Comparison of number of equivalent contacts per fibre measured experimentally with that calculated using the model. Error bars representing 95% confidence intervals

no contacts would be expected when the sheet density is zero.

Although we have considered experimental data for paper and glass fibre mats only, it is important to note that the theory is derived for general classes of stochastic fibrous materials and therefore are applicable to networks formed from fibres with arbitrary dimensions. This is important because it follows directly from Eq. (8) that the expected interval between crossings, i.e. the mean ligament length, is a function of accessible porosity—through FCA—and fibre width only. Since ligaments in fibrous structures represent the boundaries of voids and provide the available surface of the structure, then the characteristic pore size and specific surface area also depend only on fibre width and network accessible porosity. Such dependencies should aid targeted experimentation in the development of fibrous filters, non-woven components in fuel cells and nanofibrous scaffolds for tissue engineering.

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