

# Unified theory for structural statistics of flocculated and random fibre networks

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## **Abstract**

Theory is presented for the influence of sheet uniformity on several structural parameters of paper and general classes of planar stochastic fibrous materials. The theory makes use of the negative binomial distribution for the probability of coverage of points by fibres, allowing expressions to be derived for structural properties of sheets with greater variance of coverage than that observed in a corresponding point Poisson process. The expressions obtained provide insights into the extent that formation influences free-fibre-length distribution, pore size distribution, absolute contact states and fractional contact area.

## **Introduction**

The use of stochastic modelling and statistical geometry to model structural features of paper pertinent to the mechanical, optical and transport properties of the sheet is well established for the special case of random fibre networks. One of the earliest studies of such systems in the context of paper structure is that of Kallmes and Corte [1] who provided a precise definition of a random fibre network:

- The fibres are deposited independently of one another.
- The fibres have an equal probability of landing at all points in the sheet.

- The fibres have an equal probability of making all possible angles with any arbitrarily chosen, fixed axis, *i.e.* the fibres have a random orientation.

For modelling purposes, this definition is satisfied by two statistical distributions. The number of fibres covering a point in the plane of support of the network is a discrete random variable called coverage and the probability that a point has coverage  $c$  is given by the Poisson distribution,

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

with mean  $\bar{c}$ , variance  $var(c) = \bar{c}$  and coefficient of variation,  $cv(c) = 1/\sqrt{\bar{c}}$ . The Poisson distribution for coverage handles the first two of Kallmes and Corte's criteria in their definition of a random fibre network. The third criterion is satisfied by using a uniform probability density function for the angle made by the major axes of fibres to a given direction.

The mean coverage of a fibre network with mean grammage,  $\bar{\beta}$ , depends only on the mass per unit area of the fibres  $\beta_f$ , as given by the ratio of fibre coarseness,  $\delta$ , to fibre width,  $\omega$ :

$$\bar{c} = \frac{\bar{\beta}}{\beta_f} = \frac{\bar{\beta}\omega}{\delta} . \quad (2)$$

Accordingly, the statistics of coverage at points in random fibre networks are independent of fibre length and morphological characteristics of fibres along their length such as curl, kink, *etc.*

Certain properties of random fibrous networks can be determined directly from the Poisson distribution only. For example, the probability of finding a pinhole in a sheet is given by the Poisson probability of coverage zero,

$$P(0) = e^{-\bar{c}} . \quad (3)$$

Similarly, the free-fibre-length distribution arises as the distribution of intervals along the length of a fibre in which there are no fibre crossings [1] and is given by the exponential distribution which has probability density,

$$f(g) = \frac{1}{\bar{g}} e^{-g/\bar{g}} , \quad (4)$$

with mean  $\bar{g}$  and variance  $var(g) = \bar{g}^2$ .

In networks with mean coverage less than about 1, only a small fraction of the network has coverage greater than 3 and wherever fibres cross they can be considered to be in contact. We classify these thin networks as 'two-dimensional' and their fractional contact area  $\Phi_{2D}$ , *i.e.* the structural analogue of relative bonded area, was derived by Kallmes *et al.* [2] by considering the Poisson probabilities of the network having coverage 1, 2 or 3 to give,

$$\Phi_{2D} = 1 - \frac{1}{\bar{c}} + \frac{e^{-\bar{c}}}{\bar{c}} . \quad (5)$$

At higher values of mean coverage, we expect that there will be regions where one fibre lies above another but is separated by some vertical distance through the influence of nearby fibres. The fractional contact area of networks of this type has been shown to depend on mean coverage and porosity only [3], this being consistent with the theoretical approaches of Soszyński [4, 5].

Kallmes *et al.* [2] considered also the configurations of fibre contacts in two-dimensional random networks, *i.e.* the fraction of the network where fibres are in contact with 1, 2 or no other fibres at points along their length, no other configuration being possible. They gave the probability of these *absolute contact states*, zero, 1 and 2 as,

$$B(0) = e^{-\bar{c}} \quad (6)$$

$$B(1) = \frac{2}{\bar{c}} (1 - (1 + \bar{c}) e^{-\bar{c}}) \quad (7)$$

$$B(2) = \frac{1}{\bar{c}} (\bar{c} - 2 + (\bar{c} + 2) e^{-\bar{c}}) \quad , \quad (8)$$

respectively. This treatment was recently extended by Sampson and Sirviö [6] to account for the separation of vertically adjacent fibre surfaces that is characteristic of networks with coverages and porosities more typical of industrially formed papers. Equations (6) to (8) are still valid for such structures, but represent the fractions available for contact with 0, 1 or 2 other fibres.

These absolute contact states are important because we expect regions where there is no contact to exhibit different stress-strain behaviours from those with one or two contacts. In particular, fibres at the surfaces of the network can contact other fibres on one side only and as such might be considered less efficient at bearing load than other fibres within the bulk of the network. The fraction of the total fibre surface that is available for contact with other fibres has recently been derived for random fibre networks [7] and is given by,

$$f = 1 + \frac{\gamma - Ei(\bar{c}) + \log(\bar{c})}{e^{\bar{c}} - 1} \quad (9)$$

where  $\gamma$  is Euler's constant and  $Ei(\bar{c})$  is the exponential integral function<sup>1</sup>.

Whereas the structure of handsheets can be considered to approximate that of a random fibre network formed from the same constituent fibres [8, 9], the higher consistencies used in industrial papermaking lead to flocculation of fibres in suspension and worse formation, *i.e.* sheets exhibit a higher variance of local coverage than that arising from an equivalent point Poisson process in two dimensions. When comparing the formation of real sheets with that calculated for a random network formed from the same fibres, it is convenient to use the ratio of their variances. This approach was used by Corte [10] and has subsequently been used extensively by Dodson and coworkers, see *e.g.* [8] who termed the variance ratio the

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<sup>1</sup>Note that Equation (9) is a simplified form of that given in [7]

‘formation number’,  $n_f(x)$ , this parameter being dependent on the scale of inspection,  $x$ . We note that Dodson and coworkers [11–13] report correlations between the formation number and flocculation in suspension, as quantified by the crowding number [14].

Here we derive expressions that account for the influence of formation, as characterised by the formation number at points,  $n_f$ , on the structural properties of the sheet that have been discussed so far. To account for the increased variance of coverage over that observed using the Poisson distribution, we use the negative binomial distribution for the probability of coverage. This distribution has been widely applied in other fields where data exhibit higher degrees of clustering, and hence higher variance, than those arising from equivalent Poisson processes, see *e.g.* [15–17]. It is particularly well suited to modelling flocculation in paper since it allows the variance of coverage at points to be varied independently of the mean, which is precisely the physical manifestation of forming processes, where mean coverage is held constant and flocculation influences its distribution in the plane. Recall that for *random* networks, as modelled using the Poisson distribution, the variance of coverage at points is equal to the mean coverage. We do not concern ourselves here with the mechanisms influencing network uniformity such as suspension crowding and forming dynamics, but constrain our treatment to description of network structure at given level of formation, as parameterised by the formation number.

## Theory

Consider a stochastic fibre network with mean coverage  $\bar{c}$  such that the probability that a point in the plane of support has coverage  $c$  is given by the negative binomial distribution:

$$P^*(c) = \frac{\Gamma(c+m)}{c! \Gamma(m)} (1-p)^c p^m \quad \text{for } c = 0, 1, 2, \dots \quad (10)$$

where  $\Gamma(m)$  is the Euler gamma function and the mean, variance and coefficient of variation of coverage at points are given in terms of parameters  $m$  and  $p$ :

$$\bar{c} = \frac{m(1-p)}{p} \quad (11)$$

$$var^*(c) = \frac{m(1-p)}{p^2} \quad (12)$$

$$cv^*(c) = \sqrt{\frac{1}{m(1-p)}} \quad (13)$$

where the asterisks denote that we are dealing with flocculated networks.

Our interest is flocculated networks with variance of coverage greater than that arising from an equivalent point Poisson process in two dimensions. We quantify the departure from

randomness arising from flocculation by the formation number at points, as given by the ratio of the point variance of a flocculated network to that of a random network,

$$n_f = \frac{var^*(c)}{var(c)} . \quad (14)$$

A property of the Poisson distribution is that  $var(c) = \bar{c}$ , so we have,

$$cv^*(c) = \sqrt{\frac{n_f}{\bar{c}}} . \quad (15)$$

Substituting for  $cv^*(c)$  and solving Equations (11) and (13) simultaneously for  $p$  and  $m$  gives,

$$m = \frac{\bar{c}}{n_f - 1} \quad (16)$$

$$p = \frac{1}{n_f} . \quad (17)$$

Substituting for  $m$  and  $p$  in Equation (10) yields, on manipulation, the negative binomial probability function in terms of  $\bar{c}$  and  $n_f$  only:

$$P^*(c) = \frac{\Gamma\left(c + \frac{\bar{c}}{n_f - 1}\right)}{c! \Gamma\left(\frac{\bar{c}}{n_f - 1}\right)} \left(1 - \frac{1}{n_f}\right)^c n_f^{-\frac{\bar{c}}{n_f - 1}} \quad \text{for } c = 0, 1, 2, \dots \quad (18)$$

We note that as  $n_f \rightarrow 1$ , Equation (18) converges to the Poisson distribution, as given by Equation (1). This is important, since any expressions we derive for structural properties of flocculated networks include the random case as a special case in the limit as  $n_f \rightarrow 1$ .

Histograms of the coverage at points as given by the Equation (18) for some flocculated cases ( $n_f > 1$ ), and by Equation (1) for the random case ( $n_f = 1$ ), are shown in Figure 1 for networks with mean coverage 5 and 10. As expected, increasing  $n_f$  increases the probabilities of the highest and lowest coverages in the network and reduces the fraction with coverage close to the mean. Inevitably, this influences the skewness of the distribution, defined as the third central moment divided by the cube of the standard deviation and given by,

$$S_k = \frac{2n_f - 1}{\sqrt{\bar{c}n_f}} . \quad (19)$$

So, skewness increases with flocculation, decreases with increasing coverage and when  $n_f = 1$  we recover the skewness of the Poisson distribution,  $1/\sqrt{\bar{c}}$ .

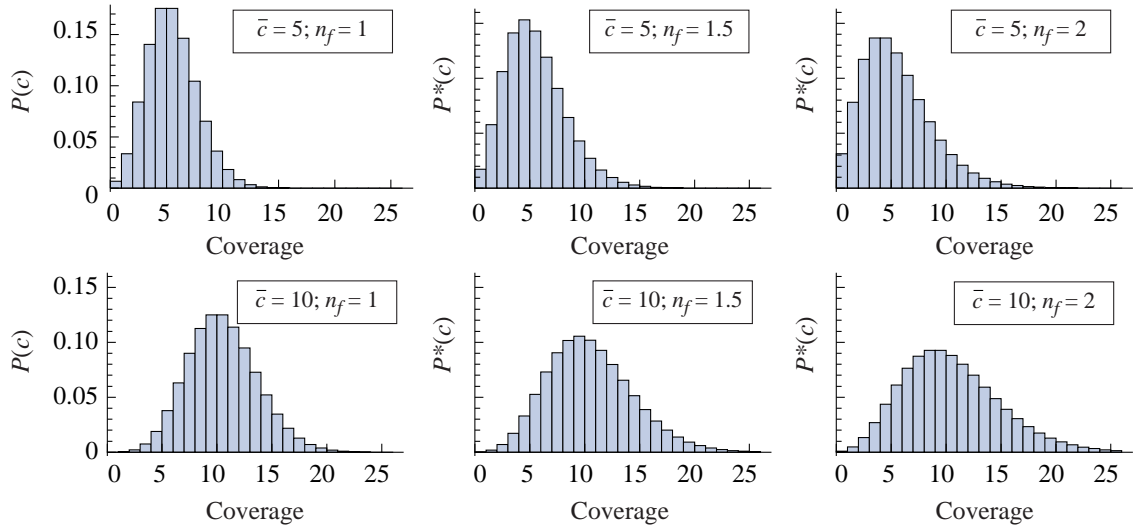


Figure 1: Histograms of coverage for networks with mean coverage  $\bar{c} = 5$  (top row) and  $\bar{c} = 10$  (bottom row). Histograms on the left represent the random case as given by the Poisson distribution; other histograms represent flocculated cases as given by the negative binomial distribution with variance over random quantified by  $n_f$ .

## Pinholes

The probability of a pinhole occurring in the network is given by the probability of coverage zero, *i.e.*,

$$P^*(0) = n_f^{-\frac{\bar{c}}{n_f-1}}, \quad (20)$$

and  $\lim_{n_f \rightarrow 1} P^*(0) = e^{-\bar{c}}$  recovering the result for the random case.

Equation (20) is plotted against mean coverage in Figure 2 where  $n_f = 1$  corresponds to the random case and the probability of pinholes increases with flocculation. Direct comparison of Equation (20) with experimental data reported in the literature is difficult, since the formation number at points,  $n_f$  is rarely reported. It is illustrative however to consider how the incidence of individual pinholes is related to the probabilities plotted in Figure 2. Wiseman [18] suggests that pinholes in newsprint have equivalent circular diameters of order  $30 \mu m$ , so we may estimate the area of an individual pinhole as being around  $7 \times 10^{-10} m^2$ . Accordingly, a probability of pinholes of  $10^{-6}$  corresponds to about 1500 pinholes per square meter. Thus, although the probabilities plotted are relatively low, Figure 2 suggests that small changes in formation can have a significant influence on the occurrence of pinholes. This is consistent with Wiseman's observations for newsprint and directory grades made on twin-wire and Fourdrinier formers. Similarly, Mauranen [19] reports a probability of pin-

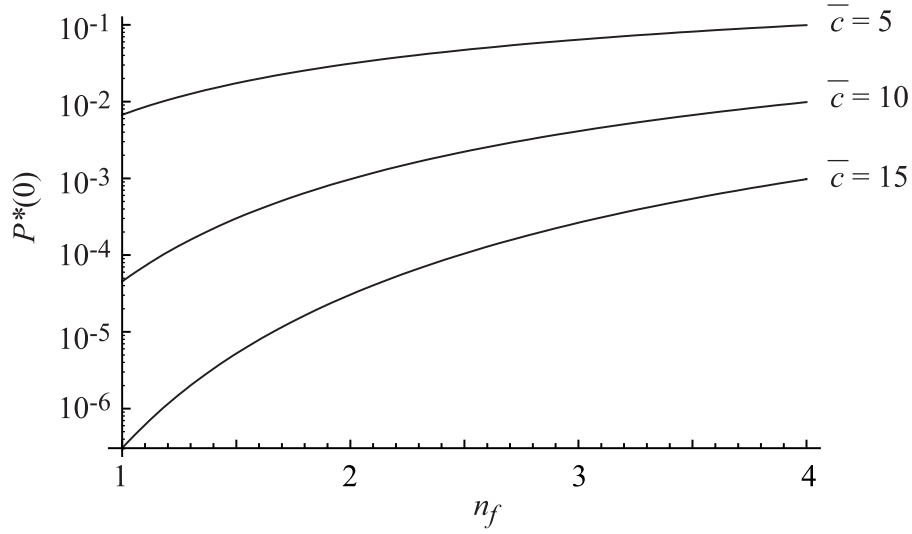


Figure 2: Probability of coverage zero plotted against formation number at points,  $n_f$ .

holes of order  $10^{-3}$  for  $35 \text{ g m}^{-2}$  directory grade sheets formed on a Fourdrinier compared with  $10^{-5}$  for the same grade formed on a roll-blade former and having better formation. Reduced incidence of pinholes with improved formation was observed also by Harwood [20].

### Free-fibre-length distribution

To derive the free-fibre-length distribution, we use the negative binomial distribution to describe the frequency of crossings along a line with  $\mu$  crossings per unit length. The expected number of crossings in an interval of length  $g$  is  $\mu g$  and the probability of there being no crossings in a gap of length  $g$  is,

$$P^*(0) = n_f^{-\frac{\mu g}{n_f - 1}}. \quad (21)$$

The probability density of  $g$  is given by,

$$\begin{aligned} f^*(g) &= \frac{P^*(0)}{\int_0^\infty P^*(0) dg} \\ &= \frac{\mu \log(n_f)}{n_f - 1} n_f^{-\frac{\mu g}{n_f - 1}}, \end{aligned} \quad (22)$$

with mean,

$$\bar{g}^* = \int_0^\infty g f^*(g) dg \quad (23)$$

$$= \frac{n_f - 1}{\mu \log(1/n_f)} \quad (24)$$

Solving Equation (24) for  $\mu$  and substituting into Equation (22) yields on manipulation,

$$f^*(g) = \frac{1}{g^*} e^{-g/g^*} , \quad (25)$$

which is, of course, the probability density function for the exponential distribution. Since clustering of points along a line has no influence on the mean distance between adjacent points,  $\bar{g}^* = \bar{g}$  and we recover Equation (4) for random networks.

As crossings can occur on both sides of a given fibre, intervals containing a small integer number of crossings may be more appropriate for characterisation of the free-fibre-lengths that form the in-plane perimeter of pores within the sheet [21]. The distribution of intervals containing  $n$  crossings in a *random* fibre network is,

$$f(g, n) = \frac{P(n)}{\int_0^\infty P(n) dg} \quad (26)$$

$$= \frac{\mu (\mu g)^n e^{-\mu g}}{n!} . \quad (27)$$

The expected length of an interval containing  $n$  crossings is  $\bar{g}_n = (n + 1) \bar{g}$ , so we have,

$$\bar{g}_n = (n + 1) \bar{g} = \int_0^\infty g f(g, n) dg \quad (28)$$

$$= \frac{n + 1}{\mu} , \quad (29)$$

such that  $\bar{g} = 1/\mu$  and,

$$f(g, n) = \frac{g^n e^{-g/\bar{g}}}{\bar{g}^{(n+1)} n!} , \quad (30)$$

which is the probability density function for the gamma distribution with mean,  $\bar{g}_n = (n + 1) \bar{g}$  and coefficient of variation,  $1/\sqrt{n + 1}$ .

It has not been possible to obtain the corresponding closed-form expression for the probability density of intervals containing  $n$  crossings in a flocculated fibre network as given by,

$$f^*(g, n) = \frac{P^*(n)}{\int_0^\infty P^*(n) dg} . \quad (31)$$



The probability densities for some small  $n$  can be obtained in closed form however, and for  $n = 1$  we recover the gamma distribution that is known to describe the intervals for the random case:

$$f^*(g, 1) = \frac{g e^{-g/\bar{g}}}{\bar{g}^2} \quad , \quad (32)$$

with  $\bar{g}_1 = 2\bar{g}$  and coefficient of variation,  $cv^*(g_1) = 1/\sqrt{2}$ .

Probability densities and hence the variance and coefficient of variation of  $g_n$  have been derived for  $n > 1$ , though these become increasingly cumbersome with increasing  $n$ . We find however that plots of these are well approximated by gamma distributions with the same mean and coefficients of variation. *i.e.*  $\bar{g}_n = (n + 1)\bar{g}$  and

$$cv^*(g_2) = \sqrt{\frac{1}{2} - \frac{3}{2(3 + \log^2(n_f))}} \quad (33)$$

$$cv^*(g_3) = \sqrt{\frac{1}{2} + \frac{9(4 - \log(n_f))}{4(12 + 9\log(n_f) + 2\log^2(n_f))^2} - \frac{15}{4(12 + 9\log(n_f) + 2\log^2(n_f))}} \quad (34)$$

such that there is a dependence of the coefficient of variation on uniformity, as quantified by the parameter  $n_f$ . This is illustrated in Figure 3 where the coefficient of variation of the length of intervals containing  $n$  crossings is plotted against  $n_f$ .

Recall that free-fibre-lengths represent the boundaries of voids in the plane of the sheet which have been modelled using the product of independent and identical gamma distributions [22]. We note however that despite an expectation that pore size will be influenced by formation [8, 22, 23], experimental evidence suggests that any dependence is rather weak [24]. Recent theoretical treatments suggest two reasons for this, firstly that the adjacent sides of polygons representing voids in the plane are correlated, and that this correlation is expected to be rather insensitive to fibre orientation and flocculation [25], and secondly that measurements of pore size are strongly influenced by the out-of-plane dimensions of voids [26], these being rather insensitive to formation [27, 28]. The insensitivity of the free-fibre-length distribution and the distributions of intervals containing  $n$  crossings to the parameter  $n_f$ , which quantifies formation, provides a third contributing factor to the observed insensitivity of pore size to formation. Interestingly, we observe that the dependence of the coefficient of variation of free-fibre-lengths is most sensitive to  $n_f$  when it is close to 1; as such any influence in near-random structures would be rather difficult to isolate experimentally.

## Absolute Contact States

The absolute contact states derived by Kallmes *et al.* [2] for random networks and given by Equations (6) to (8) represent the fractions of the fibre surface in the network that is available for contact with no other fibres, one other fibre or two other fibres. For flocculated networks,

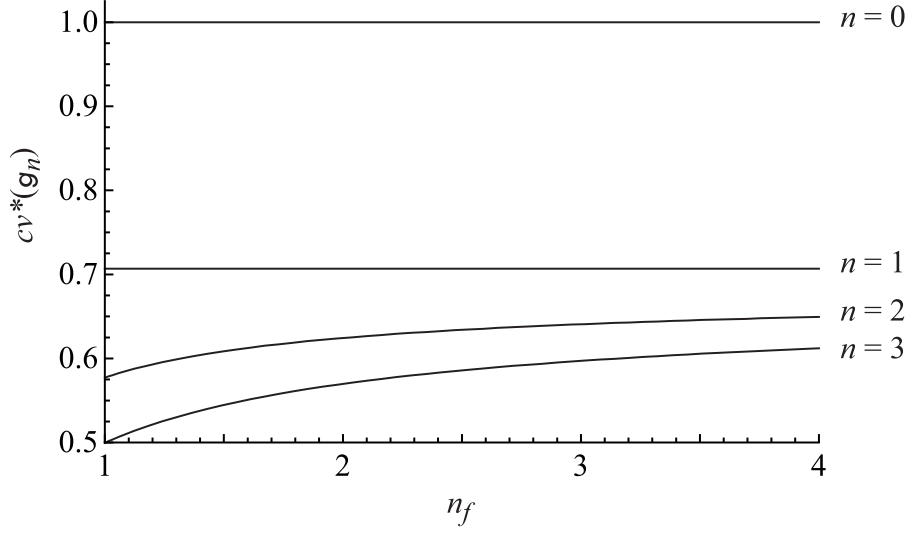


Figure 3: Coefficient of variation of intervals contain  $n$  crossings.  $cv^*(g_n)$  is independent of formation for  $n = 0$  and  $n = 1$  and exhibits a weak dependence for higher  $n$ .

we repeat their derivation using the negative binomial distribution for the probability of coverage  $c$  and we have,

$$B^*(0) = \frac{P^*(1)}{\bar{c}} \quad (35)$$

$$B^*(1) = \frac{2(1 - P^*(0) - P^*(1))}{\bar{c}} \quad (36)$$

$$B^*(2) = \frac{1}{\bar{c}} \sum_{c=3}^{\infty} (c-2) P^*(c) . \quad (37)$$

On simplification, these yield,

$$B^*(0) = \left( \frac{1}{n_f} \right)^{1 + \frac{\bar{c}}{n_f - 1}} , \quad (38)$$

$$B^*(1) = \frac{2}{\bar{c}} \left( 1 - (\bar{c} + n_f) \left( \frac{1}{n_f} \right)^{1 + \frac{\bar{c}}{n_f - 1}} \right) , \quad (39)$$

$$B^*(2) = \frac{1}{\bar{c}} \left( \bar{c} - 2 + (\bar{c} + 2n_f) \left( \frac{1}{n_f} \right)^{1 + \frac{\bar{c}}{n_f - 1}} \right) . \quad (40)$$

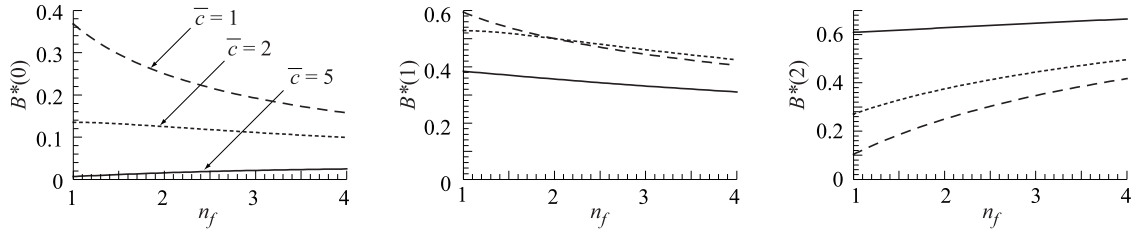


Figure 4: Absolute contact states. A weak dependence of the fractions  $B^*(0)$  and  $B^*(1)$  on flocculation is observed for networks with mean coverage 5, though the fraction  $B^*(2)$  is less sensitive. At lower coverages, sensitivity is greater.

Equations (38) to (40) are plotted against the formation number,  $n_f$  in Figure 4 for mean coverages up to 5. At mean coverage  $\bar{c} = 5$  and greater, we observe only a weak dependence on  $n_f$  and obtain the expected approximations,

$$B^*(0) \approx 0 ; \quad B^*(1) \approx \frac{2}{\bar{c}} ; \quad B^*(2) \approx 1 - \frac{2}{\bar{c}} .$$

Note that in the limit as  $n_f \rightarrow 1$ , Equations (38) to (40) recover Equations (6) to (8).

## Fractional contact area

### Two dimensional networks

Consider first a network of fibres of infinitesimal thickness such that they make contact with each other at all regions where they cross. Each fibre has an upper and a lower surface and those parts of the fibres that constitute the surfaces of the sheet can make contact with other fibres on one side only.

At a point in a fibre network with coverage  $c$ , the fraction of the fibre surface covering that point which is in contact with other fibres is,

$$\phi(c) = \frac{c-1}{c} , \quad (41)$$

such that the fractional contact area of a random two-dimensional network is,

$$\begin{aligned} \Phi_{2D} &= \frac{1}{\bar{c}} \sum_{c=1}^{\infty} c \phi(c) P(c) \\ &= 1 - \frac{1}{\bar{c}} + \frac{e^{-\bar{c}}}{\bar{c}} , \end{aligned} \quad (42)$$

which is Equation (5) as derived by Kallmes *et al.* [2] by considering the Poisson probabilities of coverage up to 3.

The fractional open area,  $\varepsilon$  of a two dimensional fibre network is given by the probability of coverage  $c = 0$  and is the two-dimensional equivalent of porosity in networks of finite thickness. From Equation (3) we have, for the random case

$$\bar{c} = \log(1/\varepsilon), \quad (43)$$

so the fractional contact area of a two dimensional random fibre network can be given in terms of its fractional open area only:

$$\Phi_{2D} = 1 + \frac{1 - \varepsilon}{\log(\varepsilon)} . \quad (44)$$

For the flocculated case we use the negative binomial distribution for coverage, and we have,

$$\begin{aligned} \Phi_{2D}^* &= \frac{1}{\bar{c}} \sum_{c=1}^{\infty} c \phi(c) P^*(c) \\ &= 1 - \frac{1}{\bar{c}} + \frac{n_f^{-\frac{\bar{c}}{n_f-1}}}{\bar{c}} . \end{aligned} \quad (45)$$

and, since  $\varepsilon = P^*(0) = n_f^{-\frac{\bar{c}}{n_f-1}}$ , we have the following unified expression for the fractional contact area of random and flocculated two-dimensional networks:

$$\Phi_{2D}^* = 1 - \frac{1}{\bar{c}} + \frac{\varepsilon}{\bar{c}} . \quad (46)$$

For two dimensional networks of equal coverage, the free parameter  $\varepsilon$  in Equation (46) increases with flocculation such that the fractional contact area increases also. Such behaviour is consistent with expectation, since increasing flocculation will increase the fraction of the network with coverage greater than 2 and hence the proportion of the total fibre length that contacts other fibres on both sides. An alternative scenario is that both coverage and flocculation vary and we note from Equation (20) that these variables are coupled such that,

$$\bar{c} = \frac{(n_f - 1)}{\log(n_f)} \log(1/\varepsilon) , \quad (47)$$

and  $\lim_{n_f \rightarrow 1} \bar{c} = \log(1/\varepsilon)$ . In the random case,  $\varepsilon = e^{-\bar{c}}$ ; it follows therefore that the fractional contact area of a flocculated network with mean coverage  $\bar{c}$  is the same as that of a random network with the same fractional open area and mean coverage  $\bar{c} \log(n_f)/(n_f - 1)$ .

## Multiplanar networks

The fractional contact area of networks with finite thickness can be modelled by considering the superposition of several two-dimensional networks. Our approach is guided by that of recent theory [3], but it has the advantage that the resultant expressions are simpler and applicable over the full range of network porosities, whereas those presented in [3] were applicable only to networks with porosity greater than about 0.3.

When two structures with fractional open area,  $\varepsilon$ , are brought together, the fraction of their projected solid area that makes contact is  $(1 - \varepsilon)^2$ . For a given two dimensional network, with fractional contact area,  $\Phi_{2D}$ , the fraction of the fibre surface that is available for additional contacts is  $(1 - \Phi_{2D})$ . Accordingly, for a network with infinite coverage formed from the superposition of an infinite number of layers, we have,

$$\Phi_{\infty} = \Phi_{2D} + (1 - \varepsilon)^2 (1 - \Phi_{2D}) \quad . \quad (48)$$

We have seen earlier that the fractional contact area of a flocculated two dimensional network with mean coverage  $\bar{c}$  is equivalent to that of a random network with the same fractional open area but with mean coverage  $\bar{c} \log(n_f)/(n_f - 1)$ . This is convenient, as it allows us to use Equation (44), which arises from consideration of random networks, to compute  $\Phi_{2D}$  and hence  $\Phi_{\infty}$  using Equation (48) such that  $\Phi_{\infty}$  is independent of  $n_f$ .

When we consider multiplanar networks of finite thickness we must take account of the fraction of the total fibre length which is located at the surfaces of the network and can be in contact with other fibres on one side only. At points with coverage  $c$ , the fraction of fibre surfaces available for contact with other fibres is  $(c - 1)/c$ . The fraction of the network covered by fibres is  $(1 - P^*(0))$  so the fraction of the flocculated network as a whole that is available for contact is,

$$f^* = \frac{1}{1 - P^*(0)} \sum_{c=2}^{\infty} \frac{c-1}{c} P^*(c) \quad (49)$$

$$= 1 - \frac{\bar{c} F(\bar{c}, n_f)}{n_f \left( n_f^{\frac{\bar{c}}{n_f-1}} - 1 \right)} \quad (50)$$

where  $F(\bar{c}, n_f)$  is the hypergeometric function,  ${}_3F_2 \left( 1, 1, \frac{\bar{c}}{n_f-1} + 1; 2, 2; 1 - \frac{1}{n_f} \right)$ . For the random case,  $f = \lim_{n_f \rightarrow 1} f^*$  and we recover Equation (9).

The fractional contact area of a network of finite coverage is given by,

$$\Phi_c^* = f^* \Phi_{\infty} \quad . \quad (51)$$

The influence of coverage and formation on the fractional contact area as given by Equation (51) is plotted against normalised density  $(1 - \varepsilon)$  in Figure 5; for clarity, the only

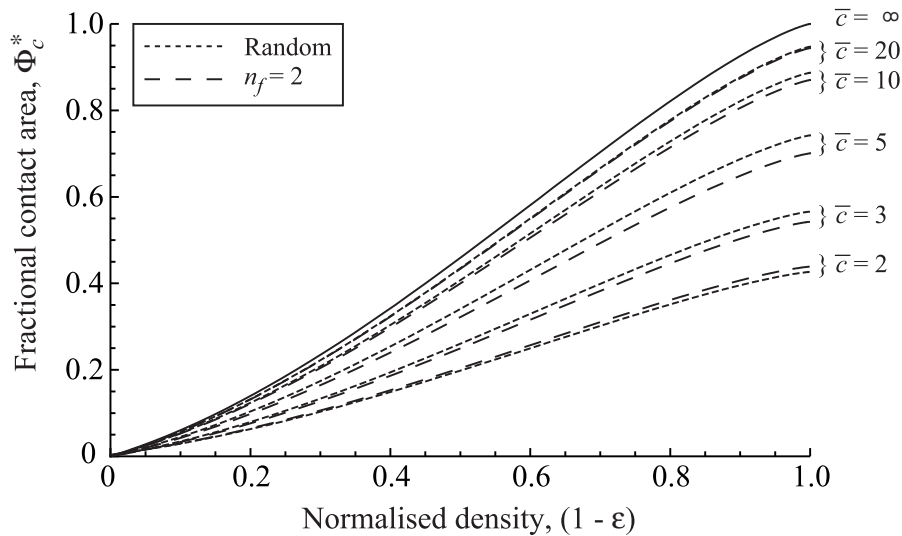


Figure 5: Fractional contact area plotted against normalised density. The influence of formation on fractional contact area decreases with increasing coverage and at a given density depends only on the fraction of fibre surface available for contact.

flocculated case shown is that with  $n_f = 2$ . For all mean coverages greater than 2, we observe that flocculation reduces the fractional contact area, though the effect is negligible at mean coverage 20 or higher. Interestingly, when the mean coverage is 2, flocculation increases the fractional contact area. The cause of these behaviours is rather subtle; we have identified that  $\Phi_\infty$  is independent of formation, so the observed dependence on formation of the fractional contact area of networks with finite coverage arises only from the fraction of the fibre network that is available for contact with other fibres. This is primarily influenced by the fraction of the network with coverage 1,  $P^*(1)$ . The surface showing how this depends on  $\bar{c}$  and  $n_f$  is shown in Figure 6. At mean coverages below about 2, increasing  $n_f$  decreases the fraction of the network with coverage 1, increasing the fractional contact area; the opposite effect is observed at higher coverages.

## Summary

Theory has been presented that provides for the first time analytic expressions for the influence of formation on several structural parameters of paper and general classes of planar stochastic fibrous materials. The expressions derived arise from consideration of the probability of coverage of points by fibres as modelled by the negative binomial distribution. This distribution permits greater variance of coverage in a network with mean coverage  $\bar{c}$  than that

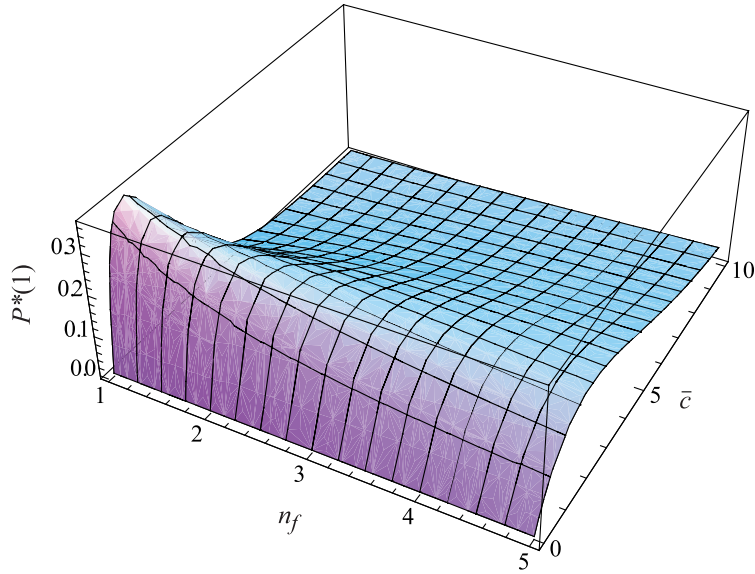


Figure 6: Surface showing the fraction of a network with coverage 1 as a function of mean coverage and formation number,  $n_f$ . At mean coverages below about 2, increasing  $n_f$  decreases the fraction of the network with coverage 1, increasing the fractional contact area; at higher coverages,  $P^*(1)$  decreases as  $n_f$  increases.

of a classical random fibre network, as modelled using the Poisson distribution for coverage. Importantly, the expressions derived recover the established results for random networks, as given using the Poisson distribution for point coverage, in the limit as  $n_f \rightarrow 1$ .

Having stated the probability function for the negative binomial distribution in terms of the mean coverage and the formation number at points—a measure of the additional variance when compared to a random network—expressions have been derived for the probability of pinholes, the free-fibre-length distribution, the distribution of absolute contact states and the fractional open area of the network. Importantly, the derivations involve no assumptions of fibre geometry, so the resultant expressions are applicable to straight, curled or kinked fibres.

The theory predicts that the skewness of the distribution of coverage increases with flocculation; a consequence of this is that the incidence of pinholes increases, in line with expectation and observations reported in the literature [18–20]. Surprisingly, the free-fibre-length distribution, *i.e.* the distribution of intervals between fibre crossings, is found to be independent of network uniformity. The distribution of intervals containing a small number of crossings, which can be considered to represent the perimeters of in-plane voids, is found to be well approximated by the gamma distribution and to exhibit a weak dependence on network uniformity. Data characterising the dependence of the free-fibre-length distribution

on formation that would allow confirmation of such a dependence have not been identified in the literature, and are probably difficult to obtain. Nonetheless, the finding is consistent with the experimentally observations that pore size distribution is well approximated by the gamma distribution and is rather insensitive to formation [24].

Expressions characterising the extent and configuration of contacts between fibres have been derived. We find that the configuration of fibre contacts, as characterised by the absolute contact states, are very insensitive to formation once mean coverage exceeds 5. This is reflected in the influence of formation on the fractional contact area; for very thin networks of a given coverage, we predict some dependence of this parameter on formation, though for networks of coverage more representative of most paper products, our models predict that the influence of formation is weak and decreases with increasing coverage.

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