Fiber crowding, fiber contacts, and fiber flocculation

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ABSTRACT

The crowding of fibers in the headbox correlates with the uniformity of the paper. Using statistical geometry, we can estimate the numbers of contacts among fibers. Some formulae are derived to help quantify the effects of fiber properties on flocculation in suspensions. An effect recently reported by Kerekes and Schell is explained quantitatively. The objective is to help predict fiber performance better by obtaining better descriptors of flocculated states.

Application:

The tendency of a pulp to flocculate can be estimated in terms of the mean aspect ratio of its fibers.

THE WAY FIBERS CROWD TOGETHER in the headbox suspension correlates with the uniformity of the paper made. Longer, stiffer fibers tend to make larger, stronger flocs than do shorter, more flexible fibers at the same mass density. Using statistical geometry, we can make some estimates about the numbers of contacts and the variability of contacts among fibers in random and flocculated three-dimensional networks.

Here, we derive some formulae that help quantify the effects of fiber properties on flocculation in suspensions. These formulae can have a bearing on paper uniformity. The overall objective is to predict fiber performance more closely by using better descriptors of flocculated states.

FIBER CONTACTS IN SUSPENSION

Consider a random fiber suspension of mass density c_{mass} and volume density c_{vol}, made from fibers with length-weighted dimensions: length l, width w, aspect ratio $A = l/w$, and mass per unit length (coarseness) d. Two apparently important number averages are associated with such a fiber suspension. These two averages are the crowding number and the contact number.

The crowding number, n_{crowd}, is the expected number of fibers in a spherical volume of diameter l. This quantity has been developed from the work of Mason *(1–4)* through Kerekes and Schell *(5, 6)*. The crowding number is given by Eq. 1:

$$n_{crowd} = (2/3)\, A^2 c_{vol} \qquad (1)$$

The contact number, $n_{contacts}$, is the expected number of fiber contacts per fiber. Therefore, $l/n_{contacts}$ is the expected distance between fiber-to-fiber contacts, the mean free fiber length. The appendix shows the derivation of Eq. 2.

$$\begin{aligned} n_{contacts} &= 3\, n_{crowd}/A \\ &= 2\, A\, c_{vol} \end{aligned} \qquad (2)$$

This expression differs from the expression obtained by Meyer and Wahren *(7, 8)*, which was given for $n = n_{contacts} > 1$, in the form of Eq. 3.

$$c_{vol} = \frac{16\,\pi A}{\left(\frac{2A}{n} + \frac{n}{n-1}\right)^3 (n-1)} \qquad (3)$$

Part of the difference may arise from the attempt by Meyer and Wahren to allow for "trapped" fibers *(8)*. The derivation of Eq. A2 in the appendix is quite short, according to a method of van Wyk *(9)* from 1946. The result agrees with a different derivation in the latest work of Pan *(10)* and Ringner *(11)*, which gave:

$$n_{contacts} = (4A\, c_{vol})/(2 + \pi c_{vol}) \quad (4)$$

Results using both Eqs. 2 and 4 are in agreement with results of new computer simulations *(11)*. The discrepancy in using Eq. 3 is greatest at small fiber aspect ratios.

Ringner showed also that the strength of a fiber suspension is not simply related to the volumetric fiber concentration but is strongly influenced by the fiber length distribution. Presumably this influence arises through the potential for making and retaining contacts.

Because paper is sold per unit mass, the number of fiber contacts per unit mass of fiber may be important. This quantity is independent of fiber length, as Eq. 5 substantiates:

$$\begin{aligned} & n_{contacts}/mass_{fiber} = \\ & n_{contacts}/(l\,d) = [\pi\, w/(2d^2)]c_{mass} = \\ & [2/(w\,d)]c_{vol} \end{aligned} \qquad (5)$$

From Eq. 5, we see that reducing the coarseness of fibers will have a substantial effect on flocculation and on the density of bonds in the paper.

Observe that both n_{crowd} and $n_{contacts}$ are linear in suspension concentration and in reciprocal coarseness. They differ, however, in their dependence on fiber dimensions. One of the few scientists who has tried to count the number of contacts in flocs is Soszynski *(12)*. Soszynski locked the structure of nylon fiber flocs by evaporating a sugar solution. Soszynski also showed that the apparent volumetric concentration of wood fibers in suspension is more than double the mass concentration *(13)*. Since contacts are controlled by the sizes of fibers, this finding is important and needs to be borne in mind when one is using the Eq. 2.

Soszynski actually found fewer contacts than expected from the formulae for isotropic random networks *(12)*. In some cases, he found fewer than three contacts per fiber, whereas $n_{contacts} \geq 3$ is thought to be necessary to hold them in a coherent network. A value of $n_{contacts}$ of approximately 4 seemed to correspond to the experimental sedimentation concentration from the work of Thalen and Wahren *(8, 14)*. In our statistical geometrical analysis above, we are neglecting the physical features of fiber shape and flexibility, which are known to be important in the flocculation behavior of fibers. [For discussions of the effects of fiber dimensions and flexibility on floc strength, see Ringner *(11)*, Thalen and Wahren *(14)*, Almin *et al.* *(15)*, and Farnood *et al.* *(16)*.]

SEDIMENTATION CONCENTRATION

A random suspension of fibers is formed when the fibers are allowed to settle freely and independently. To start with an idealized model of a sedimentation experiment, let us take a dilute, random suspension of fibers with three contacts per fiber on average. Like Wahren, we can use this model to obtain a relationship between the fiber aspect ratio and the sedimentation concentration, $c_{vol,sed}$. Let us set $n_{contacts,sed}$ equal to 3. Then, from our Eq. 2, the critical concentrations for coherent networks are:

$$n_{crowd,\ sed} = A$$

$$c_{vol,\ sed} = 3/2A$$

$$c_{mass,\ sed} = 6d/(\pi\ w\ \ l) \qquad (6)$$

Ringner's formula gives Eq. 7:

$$c_{vol,\ sed} = 6/(4A - 3\pi) \qquad (7)$$

Both Eqs. 6 and 7 are essentially the same and have $c_{vol,\ sed}$ inversely proportional to A, whereas Wahren's formula is more like $c_{vol,\ sed}$ being inversely proportional to A^2 *(8)*. Equations 6 and 7 are in better agreement with Soszynski's measured data *(12)* than would be found for $n_{contacts} = 4$. Figure 3.3 in Ringner's thesis shows the differences, which are actually rather small, in the range of $A > 30$ *(11)*. Note that Wahren and Ringner used A to represent the ratio of fiber length to radius.

Both the crowding number and contact number in random suspensions would be expected to vary locally from region to region, probably following a Poisson distribution, which in large enough regions will resemble a normal distribution. Now, cellulose fibers usually have aspect ratios on the order of $20 \leq A \leq 70$, and headboxes usually have crowding numbers on the order of $10 \leq n_{crowd} \leq 40$. The expected number of contacts per fiber in headboxes is up to about 4; it will be perhaps 30 times this value just before the dry line on a fourdrinier table.

FLOCS IN SUSPENSION

So far, our discussion has been restricted to random fiber suspensions. However, the concepts of crowding number and contact number are still valid if we know something about how the flocculated suspensions depart from ideal random suspensions.

It is tempting to try to estimate the size of flocs in terms of the mean length of fibers and the mean number of contacts per fiber. To be held in a floc, a fiber needs at least three contacts with other fibers. Also, within a reasonable range of concentrations, we expect the mean floc diameter $\overline{D}$ to increase with the mean fiber length l This relationship suggests a heuristic estimate of the form:

$$D = (l/2)\ [1 + (n_{contacts} / 3)] \qquad (8)$$

This equation is in approximate numerical agreement with the observations of Kerekes and Schell *(6)*. For flocs of diameters of about 2.5 fiber lengths, which are ordinarily observed, we expect them to average internally about 12 fiber contacts per fiber.

The reciprocal of the expected number of contacts per unit length of fiber is the expected distance between contacts. In random paper or in a not-too-dilute random suspension of fibers, this distance has a negative exponential distribution. In a nonrandom arrangement of fibers such as flocculated paper, we expect that the distance between contacts resembles a gamma distribution *(17, 18)*. The negative exponential distribution is then a special case, and it corresponds to a random arrangement of fibers.

This consideration allows us to estimate the distribution of the inter-contact distances in a flocculated suspension, which in turn permits us to estimate the corresponding effect of flocculation on the variation of contacts per fiber. At an inter-contact distance g, the number of contacts per fiber is l/g. For the random case, we expect the variance of the number of contacts per fiber to coincide with the mean value $(3w\ \ n_{crowd})/l$ For the flocculated case, the variance of the number of contacts per fiber will exceed this value.

In laboratory studies of flocculated fiber suspensions in narrow chambers, researchers use transmitted light to estimate the state of flocculation. [Some representative photographs may be seen in a paper by

Kerekes and Schell *(5, 6)*.] Transmission images are projections onto a plane, so statistical geometry may be used to model these states.

Suppose that the mean floc diameter is $\overline{D}$ and the mean projected floc grammage is $\overline{G}$. If the flocs are distributed at random, and the total grammage of fibers (not fines or filler) is $\overline{\beta}$, then the variance of local grammage measured using inspection squares of side length x is given by Eq. 9:

For $x \ll \overline{D}$,

$$\mathrm{Var}(x) \approx \overline{\beta}\ \ \overline{G}\ \ [1 - (2x / \pi\overline{D}\ \ )] \quad (9)$$

Chatterjee reported measurements of the effects of polymeric retention aids on the size of flocs *(19)*.

Kerekes and Schell found that for short fiber fractions (mean length l = 1.8 mm), the mean floc diameter was about 2.3 mm. For long fiber fractions (mean length l = 2.7 mm), the mean floc diameter was about 11 mm, at a similar n_{crowd} of about 75 *(6)*. Assuming that the mean floc grammage is the same for both length fractions, we would expect the ratio of variances to be from Eq. 9 the ratio expressed in Eq. 10:

For $x \ll \overline{D}$ mm,

$$\mathrm{Var}(x)_{long}/\mathrm{Var}(x)_{short} = 1 + (x / 5) \quad (10)$$

This ratio explains quantitatively the results reported earlier by Kerekes and Schell *(6)*: fractions of longer fibers display greater variances than do fractions of shorter fibers, at the same crowding number. Moreover, there is no conflict with the regression equation reported by Dodson and Schaffnit *(20)*, because they measured the formation in units of the random variance. The increase of the variance of grammage with fiber length is known analytically for random networks and is shown in the graph on p. 101 in *Paper: An Engineered Stochastic Structure*, a book published by TAPPI PRESS *(17)*. Work is in progress to use Eq. 9 for analysis of the transmission images of suspensions reported by Kerekes and Schell *(5, 6)*.

The practical application of these results depends on a knowledge of what constitutes a floc and on a knowledge of what range of sizes they have. There is evidence that in fiber suspensions flocs have a log-normal size distribution *(21, 22)*. If this is so, it would mean that the logarithm of the floc diameter follows a normal, or Gaussian, distribution. In consequence, the standard deviation of floc diameters will increase with the mean floc diameter, which is just like the case for pore size distributions in flocculated paper *(18)*. In commercial papers and handsheets, a log-normal distribution of floc diameters apparently gives a good fit for the observed structures, and the flocs are rather loose or open structures *(23, 24)*.

Presumably, flocs in a tank of fiber suspension are roughly spherical, so that their mass is proportional to the cube of their diameter. However, when projected onto a plane, a floc mass is spread over an area proportional to the square of the diameter. It follows that mean floc grammages in transmission images will increase in direct proportion to their diameters and will probably have a log-normal distribution. The phenomenon of flocculation in suspensions of natural cellulose fibers continues to elude deterministic physical models, but the methodology presented here may help represent in measurable terms the different states of flocculation that can occur.

On a paper machine, the situation is different because the fiber suspension is delivered in a stratified form. So we expect flocs there to arise by fibers jamming together, more or less in the same plane. To have structural coherence, a floc would need to have not less than about three contacts per fiber among its constituent fibers.

To obtain an estimate of the implications of this consideration, we shall use the formula for random networks, Eq. 2. Let's say that a typical floc has diameter D and grammage G. Suppose it occupies a region bounded by a cylinder of thickness T, so the floc mean density is $c = G / T$. Then we must have from Eq. 2,

$$n_{\text{contacts, floc}} = (\pi\ w\ \ l G) / (2 d\ T) \geq 3 \quad (11)$$

From studies of radiographs of paper, we have typical values of G = 0.5 g/m^2, w = 40 µm, l = 2 mm, and d = 0.0002 g/m *(25)*. Using these values, we can estimate the thickness T of the floc on the forming fabric to be bounded by

$$T \leq (\pi\ w\ \ l G) / (6 d) \approx 100\ \mu m \quad (12)$$

So we arrive at a mental picture of a floc on a paper machine forming section as occupying a thin cylindrical region a few fiber lengths in diameter and a few fiber widths in thickness. It remains to be seen, so to speak, if this is observable!

FLOCS IN PAPER

We have used video-beta-radiography to measure Var*(x)* over $0.2 \leq x \leq 8$ mm *(25)*. From the small scale data, we estimated the mean floc grammage and the mean floc diameter. Neither quantity seemed very sensitive to the mean fiber length, though the longest fibers did generate the largest and heaviest flocs.

Presumably, physical factors like fiber shape and flexibility (not included here) would contribute to this lack of direct sensitivity. More-

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over, experienced machine operators modify their control parameters to achieve target formation in different ways, the whole forming process being highly nonlinear. From measurements on 60 papers *(26)*, we found the following regression equation for the mean floc grammage in paper, with $R^2 = 0.7$:

For $1 \le \bar{D} \le 4$ mm,

$$\bar{G} \approx 0.28\bar{D} - 0.12 \text{ g/m}^2 \quad (13)$$

So, a rough rule of thumb seems to be $\bar{G} \approx (\bar{D}/4)$ g/m², for $\bar{D}$ in mm, though it is easy to find exceptions. Then, using Eq. 12, we find that the average floc thickness $\bar{T}$ satisfies $\bar{D} / \bar{T} \ge 20$. Farnood's work indicates that the floc diameter D does follow approximately a log-normal distribution with standard deviation increasing with the mean *(23, 27)*. From his algorithm to find the optimal mean and standard deviation of disk diameters, Farnood obtained for most papers a relation like

$$s_D \approx 1.2\bar{D}^{2/3} \quad (14)$$

The plot of $\bar{G}$ against $\bar{D}$ provides a so-called "formation diagram" of intensity vs. scale for flocculation *(27)*, which is convenient for interpreting the effects of changes in the process and furnish during formation trials. Chatterjee reports the effects of suspension concentration, settling time, polymeric additives, and shear on the size and grammage of flocs in paper *(19)*.

Corresponding to Eq. 9 for discs of diameter $\bar{D}$, the equation for the variance of small-scale grammage in random paper made from fibers of mean width w and mean aspect ratio A is *(17)* given by Eq. 15.

For $x \le$ w,

$$\mathrm{Var}(x) \approx \bar{\beta}\,\frac{\delta}{\omega}\left[1 - \frac{x}{\pi\,\omega}\left(1 + \frac{1}{A}\right)\right] \quad (15)$$

Now, the formation number n_{form} *(x)*, perhaps better called the "variance number" of paper, is the ratio of measured variance of local grammage to the variance expected for a random network made from the same fibers, using inspection zones of size x. So, if we suppose that our fibers form, on average, flocs of diameter $\bar{D}$ and grammage $\bar{G}$, then the formation number for small zone sizes is given by the ratio of Eq. 9 to Eq. 15. For $x \le$ w $<< \bar{D}$, this ratio is

$$n_{\text{form}}(x) \approx \frac{\omega\,G}{\delta}\left[1 + \frac{x}{\pi\,\omega} - \frac{2x}{\pi D}\right] \quad (16)$$

For typical papers *(24)*, the ratio of floc grammage to fiber grammage, w$\bar{G}$/d, seems to be about 1/10. From a structural standpoint, this relationship helps us understand the observation first made by Norman and Wahren *(28)* that even flocculated papers can have variances smaller than that for random networks, at very small scales *(29)*.

Interestingly, Eq. 16 shows that for $x \le$ w $<< \bar{D}$, we have n_{form} *(x) independent* of floc diameter, though experimentally we know that floc grammage tends to increase with floc diameter according to Eq. 13. Furthermore, n_{form} *(x)* increases linearly with zone size x, which is often observed even at larger scales *(17, 24)*, which is a powerful argument for using this parameter. Farnood has derived an analytical form for Eq. 16 valid for all x *(23)*, which explains fully the experimental findings at large scale *(24)*.

The gamma distribution for free fiber lengths in flocculated paper was applied to typical paper samples in a study of pore size distribution by Dodson and Sampson *(18)*. They found that the standard deviations of both free fiber length and pore radius *increase* with their respective means, with the slope depending on the fiber type. Mean free fiber length and hence mean pore size both decrease with increasing grammage, but both increase with increasing flocculation.

ESTIMATING FIBER LENGTH, WIDTH, AND COARSENESS

The simplest experiment that should be done with any pulp is to measure the sedimentation concentration $c_{\text{vol, sed}}$. This concentration gives an estimate of the mean fiber aspect ratio A, from Eq. 6.

We can also use statistical geometry in conjunction with modern image analysis methods to estimate the principal fiber characteristics. In a three-dimensional random suspension, the mean number of contacts per fiber is (3wn_{crowd})/l. Now, by forming a thin sheet from a dilute suspension or by projecting a transmission image onto a plane, we can create a structure that we can study with image analysis devices.

In particular, it is easy to count fibers, taken as half the number of ends. It is also easy to count the number of contacts, $n_{\text{contacts, proj}}$, for different grammages projected. Then, we have for projected grammage $\bar{\beta}$:

$$n_{\text{contacts, proj}} =$$

$$[4l/(\pi\,\mathrm{w})]\,[1 - (1 - e^{\bar{c}})/\,\bar{c}\,] \quad (17)$$

Equation 17 reduces to approximately $4l\bar{c}/(3\pi w) = 4l\bar{\beta}/(3\pi d)$ for $\bar{c} \leq 3$, which allows us to estimate l/d.

For planar random fiber networks, the total variance of the local grammage is $(\bar{\beta} d)/w$, where $\bar{\beta}$ is the mean grammage. Therefore, by extrapolating measured variances to zero zone size, we can estimate d/w. (Total variance means measured at all points, using zones of zero size).

In principle, Eq. 15 allows us to estimate the fiber properties w and d from the intercept and slope of the variance plot. In practice, it is easier to extrapolate to inspection zones of size zero the logarithm of the variance. However, it is still difficult to get a reliable estimate unless accurate data are available for zones of size similar to one fiber width. If we do have good enough data, for independent measurements of $2wl/d$, d/w, we can deduce l. Another estimate of ld from asymptotic variances of grammage in random structures is well known:

$$x^2\, Var(x) \rightarrow l\, d\, \bar{b}, \text{ as } x \rightarrow \infty \quad (18)$$

A different method of estimating w and d from the gradient at small scale is given by Farnood *(23)*. He provided an approximate analytic form of Var*(x)* for the whole range of zone sizes in random networks. TJ

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APPENDIX: DERIVATION OF $n_{contacts}$

This derivation is based on the methods of van Wyk *(9)*. Consider a fiber suspension represented by a random arrangement in space of cylindrical fibers of length l and diameter w. The number of fibers, n_{cube} expected to be contained in a cube of side length l is

$$n_{cube} = n_{crowd} \frac{6\,\lambda^3}{\pi\,\lambda^3}$$

$$= \frac{6}{\pi}\, n_{crowd} \qquad \text{(A1)}$$

where n_{crowd} is the expected number of fibers in a spherical volume of diameter l.

The expected number of fibers in the cube that intersect a central vertical cylinder of diameter w and height l is found by considering the projection of the cylinder and the fibers onto the base of the cube. The cylinder becomes a circle of diameter w, and the fibers become rectangles of width w. The expected number of intersections is given by the product of the number present, n_{cube}, and a ratio, which is the ratio of the mean projected length of one fiber times twice the width to the area of the base of the cube. The denominator is l^2. We obtain the numerator as follows.

A fiber inclined at angle q to the vertical has a projected length of (l sinq), and we need the average value of this length for all orientations in space. This average is given by:

$$l\,\overline{\sin\,\theta} = (\pi\, l)/4 \qquad \text{(A2)}$$

Hence, the expected number of contacts per fiber in a random three-dimensional suspension is

$$n_{contacts} = \frac{6}{\pi}\, n_{crowd} \frac{\pi\,\omega}{2\,\lambda}$$

$$= \frac{3\,\omega}{\lambda}\, n_{crowd}$$

$$= \frac{3}{A}\, n_{crowd} \qquad \text{(3A)}$$

where *A* is the aspect ratio of fibers.

Intuitively, we see that this value for $n_{contacts}$ is reasonable by considering a regular stack of layers of $n \leq A$ fibers, in which the *n* fibers in each layer are perpendicular to those in the next layer. In that case, a typical fiber in a central layer has 2*n* contacts, *n* above and *n* below, and the crowding number is $(\pi\, n\, A)/6$. So here we get

$$n_{contacts} = 12\, n_{crowd} / (\pi\, A).$$