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RANDOM FIBER NETWORKS AND SPECIAL ORTHOTROPIC ELASTICITY OF PAPER

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ABSTRACT

The objective of this study is to develop an improved understanding of the microscopic (fiber scale) interactions in cellulosic fiber networks, and of their influence on the macroscopic in-plane elastic properties of paper. In particular, the study is motivated by a particular in-plane elastic orthotropy of paper observed experimentally for various types of paper dating back to Horio and Onogi (1951) and Campbell (1961), namely: $S_{1111}+S_{2222}-2S_{1122}-S_{1212}=0$. Here, S_{ijkl} are the components of an in-plane compliance tensor. This is a statement of the invariance of in-plane shear compliance S_{1212}, which has been observed in some studies but contradicted in others.

We present a possible explanation of this "special orthotropic elasticity" of paper, using an analysis in which paper is modeled as a quasi-planar random microstructure of interacting fiber-beams. This model is especially well suited for low basis weight papers. It is shown that geometric disorder in a fiber network is necessary to explain this orthotropy; it is shown analytically that without disorder a periodic fiber network fails this relationship. On the other hand, it is disordered networks with low flocculation that best satisfy the relationship. It also follows from the micromechanical analysis that no special angular distribution function of fibers is required (such as proposed by Schulgasser and Page 1988), and that the uniform strain assumption should not be used.

INTRODUCTION

This study was motivated by a peculiar aspect of in-plane elastic orthotropy of paper (Habeger, 1997), and is an effort to use a fiber network model to reproduce and understand the behavior. The observation, apparently first reported by Horio & Onogi (1951), is that Young's modulus at an arbitrary angle in the plane of a sheet, E_θ, is dependent only on Young's moduli in the principal material directions. The relation given by Horio and Onigi (1951),

$$1/E_\theta = \cos^2\theta/E_1 + \sin^2\theta/E_2 \qquad \text{Eq. 1}$$

was observed by them to be a good predictor of experimental data for a wide variety of papers. This equation obviously differs from the relation derived from proper transformation of coordinates:

$$1/E_\theta = \cos^4\theta/E_1 + \left(2\nu_{12}/E_1 + 1/G_{12}\right)\cos^2\theta\sin^2\theta + \sin^4\theta/E_2 \qquad \text{Eq. 2}$$

Later, Campbell (1961) showed that Eq. 1 is satisfied only if

$$1/G_{12} = (1+\nu_{12})/E_1 + (1+\nu_{21})/E_2 \qquad \text{Eq. 3}$$

and suggested that it is true for all papers. A third expression of the same behavior clearly illustrates the relationship between the in-plane compliances:

$$S_{1111} + S_{2222} - 2\cdot S_{1122} - S_{1212} = 0 \qquad \text{Eq. 4}$$

In Eq. 4, S_{ijkl} are the components of an in-plane compliance tensor. We shall use the term "special orthotropy" to describe the satisfaction of Eqs. 1, 3, or 4.

Over the years, other researchers have reported mixed success in attempts to verify that paper elastic properties do indeed satisfy the requirements for special orthotropy. Craven and Taylor (1965) showed that for a variety of machine made kraft papers Eq. 1 is a very good predictor of E_θ. Jones (1968) noted that one result of special orthotropy is that shear modulus is independent of orientation with respect to material principal directions, but his experimental data showed that G *did* vary with orientation. Jones demonstrated that Eq. 1 was a good predictor of Young's modulus at an angle, but he also noted that Eq. 2 was slightly better. Later, Suhling *et al.* (1989) showed that Eq. 3 gave shear modulus within five percent of their careful experimental measurements.

This special relationship among the elastic properties is not observed in most materials – Liu and Ross (1997) clearly showed that it is not true for wood. The question becomes, therefore, under what conditions can it occur in paper?

Schulgasser (1981) tried to explain Eq. 4, using the so-called Cox model (Cox, 1952). This model provides an analytical derivation of the in-plane compliance of a mat of infinitely long fibers, laid in a plane according to probability density function

$$f(\theta) = \frac{1}{\pi}(1 + a_1 \cos 2\theta + a_2 \cos 4\theta + ... + a_n \cos 2n\theta)$$

Eq. 5

In Eq. 5, θ is the angle a fiber makes with respect to the x-axis and it must be between zero and π. The Cox model involves an assumption that all the fibers carry axial forces only, which necessarily implies that they interact via frictionless pivots. This, combined with the fact that they are infinite, results in the entire fiber network deforming by a uniform strain. The Cox model generally leads to good estimates of effective Young's moduli but underestimates the shear modulus, both for isotropic and orthotropic systems. Schulgasser (1981) showed that the Cox model results in Eq. 4 when Eq. 5 is used with

$$a_2 = \frac{(a_1)^2}{2}$$

Eq. 6

or when a wrapped Cauchy distribution is used in place of Eq. 5:

$$f(\theta) = \frac{1}{2\pi}\left(\frac{1-\rho^2}{1+\rho^2 - 2\rho\cos\theta}\right)$$

Eq. 7

where ρ is a constant between 0 and 1. Schulgasser & Page (1988) showed that a model of paper treated as a laminate composite could yield the special orthotropic relations as well, providing certain values of fiber moduli were used; this model, too, relied on the uniform strain assumption.

In the present work we show that an analysis which simulates the heterogeneous three-dimensional network of fibers in paper, avoiding several of the assumptions that made the Cox model attractive for its simplicity, can reproduce the conditions of special orthotropy. In this computationally intensive procedure, fibers can transmit axial and shear forces and bending and torsional moments between rigid bonds (an extension to the current work, which allows bonds to deform under load, is in progress). The present form of analysis is justified on three counts: (i) Hydrogen bonds between fibers are not perfect hinges; mechanically we have a frame rather than a truss. Researchers since Page *et al.* (1962) have described paper fibers as carrying shear and moment in addition to axial loads. (ii) It is shown that periodic beam networks can only satisfy Eq. 4 under a specific geometric condition (which renders them isotropic, so the point is moot), so *disordered* networks are necessary if we are to explore special orthotropy. (iii) Once we have made the leap to analysis of disordered networks, it is not difficult to include important geometric features such as flocs. We show that any disorder, especially non-uniform areal distribution of fibers into flocs, produces strain fields which are not uniform.

PLANAR PERIODIC FIBER NETWORKS

We introduce an idealized, albeit exactly solvable model of a network of fiber-beams: a triangular lattice, whose perspective view is given in Fig. 1. For simplicity the lattice nodes are assumed to have the same thickness as a single fiber. Each fiber is taken to have a rectangular cross section of height t and width w, with the distance between nodes equal to s. Such a beam lattice was analyzed by Wozniak (1970), but given the presence of short fiber segments in the present case, we use the Timoshenko beam model to admit beam shear deformation. Much of the following development follows the work of Wozniak (1970) and Ostoja-Starzewski *et al.* (1996).

The strain energy density in each unit cell (defined by hexagons in Fig. 1) is given as:

$$U_o = \frac{1}{2}\bar{\varepsilon}_{ij} C^1_{ijkm} \bar{\varepsilon}_{km} + \frac{1}{2}\bar{\kappa}_i C^2_{ij} \bar{\kappa}_j$$

Eq. 8

where $\bar{\varepsilon}$ and $\bar{\kappa}$ are the strain and curvature on the boundary of the unit cell (this is a micropolar elastic problem). The constitutive tensors, C^1_{ijkm} and C^2_{ij}, are found as

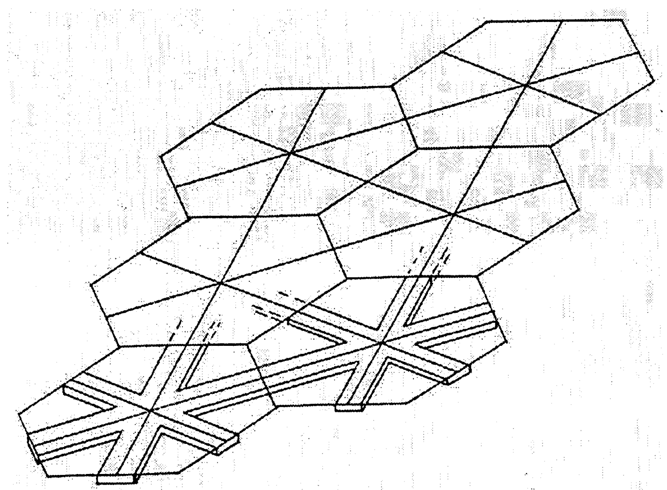


Figure 1. Periodic network.

$$C^1_{ijkm} = \sum_{\text{fibers}} n_i n_k (n_j n_m R + \tilde{n}_j \tilde{n}_m \tilde{R}) \qquad C^2_{ij} = \sum_{\text{fibers}} n_i n_j S$$

Eq. 9

where n_i and $\tilde{n}_i$ are components of the unit vectors along and normal to a fiber, and

$$R = \frac{2EA}{s\sqrt{3}} \qquad \tilde{R} = \frac{24EI}{s^3\sqrt{3}}\frac{1}{1+\beta} \qquad S = \frac{2EI}{s\sqrt{3}}$$

Eq. 10

In Eq. 10 E, A, and I are the fiber modulus of elasticity, cross sectional area, and moment of inertia. As noted earlier, s is the length. In Eq. 9 it is assumed that all fibers are identical. The quantity β is the dimensionless ratio of bending to shear stiffness:

$$\beta = \frac{12EI}{GAs^2} = \frac{E}{G}\left(\frac{w}{s}\right)^2$$

Eq. 11

Working with Eq. 9 and Eq. 10 for a periodic network similar to that of Fig. 1, but with the beam orientation angles α at arbitrary values, we find the following expressions for terms in the network's compliance tensor:

$$S_{1111} = \frac{-R + \left(R - \tilde{R}\right)\cos^2\alpha}{R\left(-R + \left(R - 3\tilde{R}\right)\cos^2\alpha\right)}$$

$$S_{2222} = \frac{R + 2\left(R - \tilde{R}\right)\cos^4\alpha + 2\tilde{R}\cos^2\alpha}{2R\left(R - \left(2R - 3\tilde{R}\right)\cos^2\alpha + \left(R - 3\tilde{R}\right)\cos^4\alpha\right)}$$

$$S_{1122} = S_{2211} = \frac{\left(R - \tilde{R}\right)\cos^2\alpha}{R\left(-R + \left(R - 3\tilde{R}\right)\cos^2\alpha\right)}$$

$$S_{1212} = \frac{2}{-\tilde{R} + \left(2R - \tilde{R}\right)\cos^2\alpha + \left(R - \tilde{R}\right)\cos^4\alpha}$$

Eq. 12

The compliance terms of Eq. 12 can only satisfy the special orthotropy condition of Eq. 4 for angle α equal to 60°; which creates an *isotropic* network so the point is moot. If we cannot devise a periodic fiber network that satisfies the special orthotropy conditions, can we satisfy those conditions with a disordered network?

COMPUTATIONAL MECHANICS OF RANDOM FIBER NETWORKS

Our modeling of the mechanics of fiber networks is based on the following assumptions and steps (Stahl and Cramer 1998, Stahl and Saliklis 1997):

(i) Generate a system of finite-length straight fibers according to probability density functions controlling the spatial distribution of fibers and distribution of fiber orientations. The fibers are placed in three dimensions with possible non-zero angles to control out-of-plane orientation of the fiber axis and the "roll" of the fiber about its own axis.

(ii) Fibers may have different dimensions and mechanical properties, sampled from any prescribed statistical distribution.

(iii) Bonds are identified where the prismatic volumes of two fibers intersect. Bonds are rigid, but because fiber elements extend to the center of a bond, the end of the fiber element in what really is a bonded zone with finite dimension offers some simulation of bond flexibility.

(iv) Each fiber is a series of linear elastic three-dimensional Timoshenko beam elements (shear deformation is permitted) between bonds.

(v) One type of analysis establishes the network's effective stiffness tensor from a postulate of strain energy equivalence (Ostoja-Starzewski and Wang 1990). Equivalent continuum elastic properties are extracted from the inverse of the stiffness tensor.

(vi) A progressive failure analysis is possible, in which a prescribed displacement at one or more edges is incremented and fiber and/or bond failures are serially identified. This analysis will not be discussed further in the present paper.

Prior to presenting results of the elastic analysis, it is worth noting additional details of the network geometries that this procedure can produce. When placing fibers in a network, we begin by defining the volume with dimensions LMD, LCD, and t (actually a larger network is generated, and then its boundary layers are trimmed off to create a spatially homogenous field). We define the "density" d of the network as the total fiber length per MD-CD square mm area. Coverage and sheet grammage are obviously directly proportional to d. By keeping d and t independent, we can simulate papers with the same coverage but different degrees of compaction – corresponding to different degrees of pressing during papermaking. The degree of compaction is measured by the relative bonded area RBA, which is the ratio of total area of all bonds to the total projected area of all fibers. This methodology reflects our understanding that the fiber network model is likely to be realistic only for papers that have relatively low RBA – so their primary load-carrying mechanism is transfer of forces along fiber axes between nodes.

Flocculation in the network is modeled through a two step process: First floc centers are generated in the volume – the number is controlled by a parameter n equal to the number of flocs per MD-CD square mm. Then fibers are placed into the network, first by randomly assigning each fiber to a floc and then by locating each fiber with respect to the floc's center. The latter is done by placing the fiber's center a distance r from the floc's center according to the probability density function

$$f(r) = -\frac{b^2}{2} r + b \qquad r > 0$$

Eq. 13

As b increases, fibers are clustered into tight flocs, and as b decreases they are scattered. This is apparent in Fig. 4. A fiber's in-plane orientation angle is controlled with Eq. 5, and the flocs are dilated in the MD direction according to

$$r_{MD} = r(1 + a_1)\cos\theta \qquad r_{CD} = r\sin\theta$$

Eq. 14

This analysis has the capability to handle displacements and forces in three dimensions, but the work reported here concerns only planar deformation. This is obviously a computationally intensive procedure, and to allow analysis of larger networks we have disabled the out-of-plane degrees of freedom at each node. The network geometry, however, remains truly three-dimensional.

ANALYSIS RESULTS: SPECIAL ORTHOTROPY

Network analysis was used to explore the relationship between variables controlling network geometry and the idea of special orthotropy. The variables considered were network thickness t, floc parameter b, and fiber orientation with the parameters a_i in Eq. 5. As noted earlier, changing t for a given coverage affects the network's RBA. All networks were 4 mm square, with 35 mm of fiber per mm^2 area. All fibers were 1 mm long, .05 mm wide, and .015 mm thick. The resulting coverage is approximately 1.75.

Pior to discussing this data directly, we show some of the differences between the predictions of the present model and the Cox model. Example networks with each of five thicknesses and each of two degrees of scatter were analyzed. The networks were nominally isotropic, with all a_i of Eq. 5 equal to zero. Results shown in Fig. 2 are the ratio of network modulus from the present analysis to the predictions of the Cox model applied to these geometries. Each data point shown represents the mean modulus for ten example networks; variability was low with standard deviations between 3 and 8 percent of means. The data show that the present model is "softer" than the Cox model, as one should expect due to the present model's short fibers. The softness increases as thickness increases, because the networks become less well-connected (RBA decreases). Also, networks with tighter flocs are softer, as the sparse areas between flocs present little resistance to deformation.

Anisotropic networks were analyzed to evaluate special orthotropy. Two degrees of anisotropy were evaluated; some networks had a_1 of Eq. 5 equal to 0.5 and other a_i equal to zero, and some had a_1 equal to 0.5 and a_2 equal to 0.05. Closeness of fit to the condition of special orthotropy is given by a non-dimensional parameter we call the "Campbell number"

$$n_C = \frac{S_{1111} + S_{2222} - 2 \cdot S_{1122} - S_{1212}}{S_{1111} + S_{2222}}$$

Eq. 15

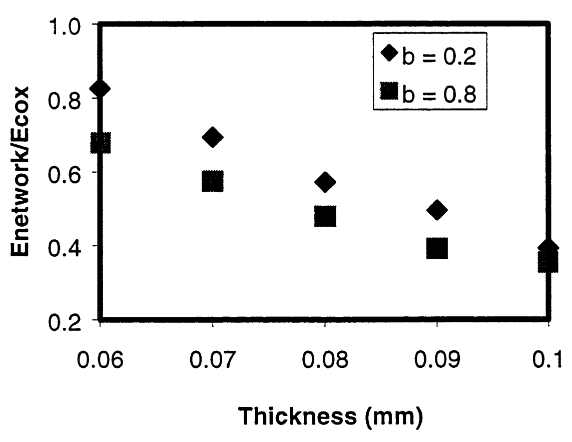


Figure 2. Network effective modulus from present analysis compared to Cox model, as affected by sheet thickness t (RBA decreases as t increases) and flocculation parameter b (flocs tighten as b increases).

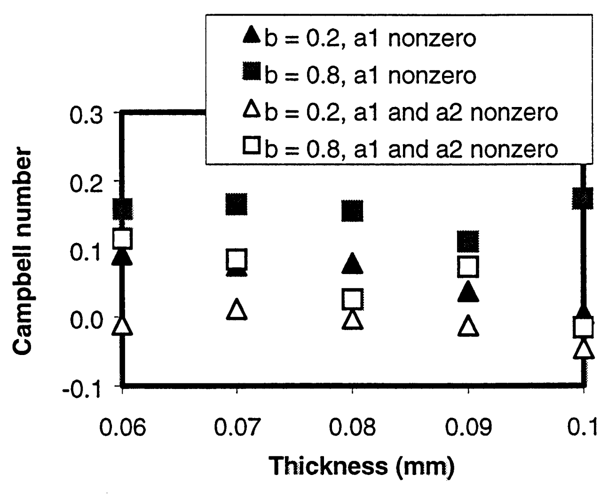
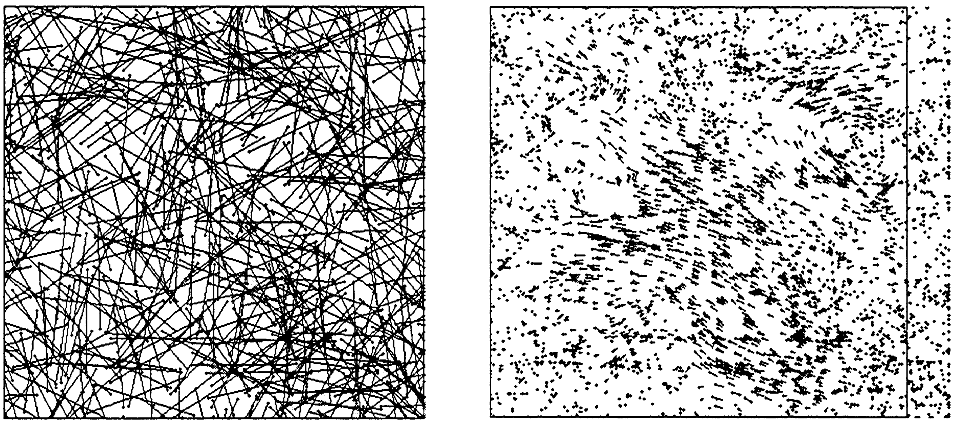
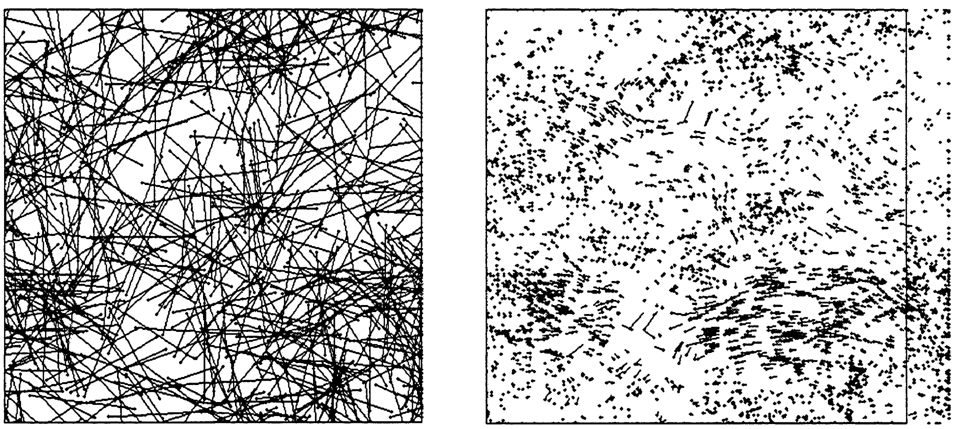


Figure 3. Relation of Campbell number to sheet thickness t (RBA decreases as t increases), flocculation parameter b (flocs tighten as b increases), and fiber orientation function (Eq. 5).

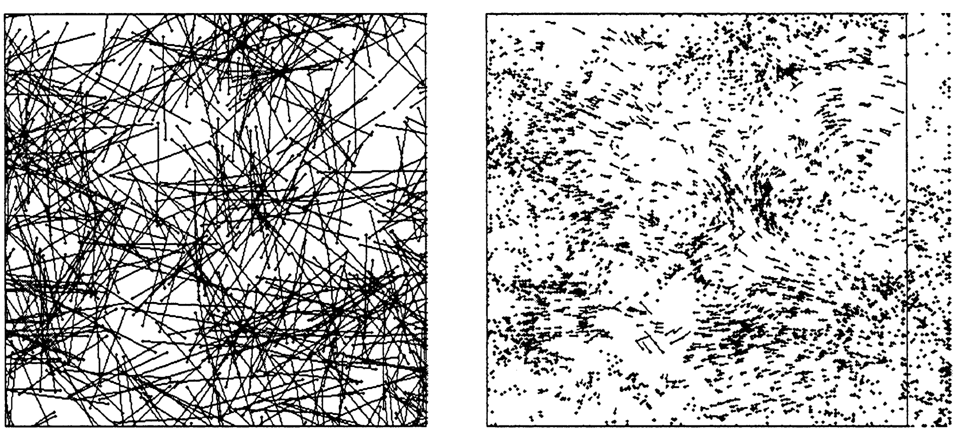
Each point in Fig. 3 represents the mean value for five example networks. The variability is high enough that we cannot make significant conclusions regarding the trends as network thickness increases, but we can make qualitative observations regarding the effect of flocculation and fiber orientation: The data show that networks with little flocculation (low parameter b) can come closer to satisfying the Campbell relation than networks with tighter flocs (compare solid triangles and squares, or outlined triangles and squares). Also, networks with slightly less orientation (a_1 *and* a_2 nonzero) are closer to satisfying the Campbell relation than strongly oriented networks (compare solid and outlined shapes).



a) floc parameter b = 0.2



b) floc parameter b = 1.6



c) floc parameter b = 2.4

Figure 4. Undeformed network geometry (left) and comparison of node displacements to uniform displacement field (right)

ANALYSIS RESULTS: NON-UNIFORMITY OF DISPLACEMENT FIELDS

Having made the argument that disorder plays a central role in establishing the relationships between the effective elastic properties, we now consider the effect of disorder on the uniformity of the displacement field. As a starting point we have the assumption inherent in closed-form solutions such as the Cox model, and in most meso-scale or cellular models, that the displacement of any point within the network is determined by a uniform strain field. Under this scenario, measures of non-uniformity such as, for example, the fiber's proximity to a floc do not affect its deformation. Clearly this assumption is appropriate for *homogeneous* materials.

To evaluate the effect of disordered network geometry on displacement fields, a series of networks with three degrees of flocculation were analyzed. Example networks are shown in the left column of Fig. 4; they are 4 mm square with 30 mm of fiber length per square mm MD-CD area. The fibers are 0.05 mm wide, and this combined with the total fiber length gives a coverage of 1.5. Using a typical value for fiber coarseness this is equivalent to approximately 8 gram/m^2 basis weight. The sheet thickness was 0.03 mm, and the resulting RBA varied from 0.56 to 0.66. There is no preferred direction in these networks – all coefficients in Eq. 5 are zero. The only parameter that was varied was the floc parameter, with the three values shown in Fig. 4.

The networks were analyzed subject to a prescribed x-strain *on the boundaries*, and the figures in the right column of Fig. 4 show the difference between the resulting displacement of each node and what the displacements would be if the strain field *in the interior of the network* were uniform. If the displacement field were actually uniform, the figures would consist simply of dots; the lines represent deviation from uniformity. Two qualitative observations can be made: First, the deviation from uniform displacements is certainly apparent in all three networks, even the one with low degree of flocculation. Second, there seems to be a combination of two main effects – groups of well connected fibers are held back or pulled along depending on whether their connections to the left or right edge are stiffer (apparent at the lower right and lower left of the middle figure); and there are swirls where the fibers on opposite sides of open areas are pulled more or less severely (apparent near the upper right corner of the top figure and also at several spots in the bottom figure). To quantitatively describe the non-uniformity in the displacement fields we calculate the root-mean-square difference between each node's actual displacement and what it would be if displacements were uniform. The average rms displacement difference for sets of ten networks is 1.66 for floc parameter b = 0.2, 2.61 for b = 1.6, and 2.86 for b = 2.4. While the absolute magnitudes of these numbers are not worth discussing, they clearly show a trend toward less uniformity in the displacement fields as the network geometry becomes less homogeneous.

SUMMARY

The literature does not unambiguously answer the question of whether or not "special orthotropy" as proposed by Campbell is common in paper. The work presented in this paper was undertaken with the view that we should consider under what conditions special orthotropy *could* occur. The Cox model, with its neglect of fiber bending stiffness, can only produce special othotropy with a few specific fiber orientation distributions. On the other hand, closed form solutions for the effective compliances of a *periodic* network were used to show that such a network, if anisotropic, cannot satisfy the required conditions. Given the need to consider disordered networks, an analysis for random networks which may contain flocs and other non-homogeneities was used.

The random network analysis showed that under certain conditions, namely low degree of flocculation and weak fiber orientation, special orthotropy can be achieved. These results are dependent on taking into account the flexural and shear deformation of fibers as well as their axial deformation. The evaluation of parameter space was not comprehensive, so there likely are other means to achieve the same relation among the effective material properties. Such potentially important parameters as fiber curl, ratio of fiber flexibility to bond flexibility, and ratio of fiber flexural stiffness to fiber axial stiffness were not evaluated.

Finally, a demonstration was made regarding the non-uniformity of the displacement fields in these networks. It was shown that decreasing the degree of geometric homogeneity decreases the uniformity of the displacement field. Non-uniformity of the displacement field is likely to have a significant effect on the progression of microfailures that leads to a network's stiffness degradation and failure. Fiber and/or bond failures are likely to occur in areas of relatively intense deformation before they would occur if the deformation were uniform. Network effective strengths should be lower than they would be if the strain were uniform. This is the subject of continuing work.

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