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SCALING, PERCOLATION AND NETWORK THEORIES: NEW INSIGHTS INTO PAPERMAKING?

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ABSTRACT

Recent advances in the theory of condensed matter physics have furnished us with powerful theoretical methods for understanding the structural and dynamical properties of inhomogeneous materials, such as the fibre network which constitutes paper. We discuss the concepts of universality and scaling, on which all the new theoretical arguments are based. In understanding the properties of forming and of paper, the percolation transition, i.e. the unique state where an infinitely connected network of fibres is formed, is of particular interest. The physical aspects and the power of the percolation theory are discussed in the presentation. Applications of these new concepts to paper have so far been few.

We show how the percolation theory yields practical qualitative results explaining the relationship between the consistency of the suspension in the headbox and the formation. The complicated structure of the turbulence on the wire and on the jet, which manifests itself in the residual variations of basis weight, is discussed using some scaling ideas related

to the universality of the nonlinear dynamical systems. The effect of formation on the mechanical properties, especially ultimate strength, can be viewed theoretically with the aid of scaling, percolation and network theories. The reinforcing effect of chemical fibres in newsprint can be judged by a rather simple scaling argument, which, when developed further, gives insight into the nonlinear relationship between strength and the amount of chemical fibres in newsprint.

Scaling and percolation are qualitative methods. When studied and applied properly, these concepts help us get a picture of the inhomogeneous materials and understand the basic principles behind their properties. It is then easier to decide on the right quantities to measure when quantitative information is needed in papermaking.

1 A SHORT INTRODUCTION TO SCALING AND PERCOLATION

When modelling a system it is most important to find out its symmetries. The model is feasible only if it possesses all the symmetries of the system. The model is then solved separately for each representation of the symmetry group(s), which reduces the complexity of the problem considerably. Also, when the problem cannot be solved analytically, but is simulated, the symmetries tell us how to optimise computation and how results obtained for particular parameter values carry over to certain other combinations of the parameter values. The present condensed matter theory which has been very successful in explaining the properties of ordered materials such as metals and insulating crystals, relies heavily on symmetry under two discrete groups: translational and rotational transformations.

Scale transformation is a trivial but less-used operation under which systems can be invariant (and thus "symmetrical").[1] A scale transformation simply changes the "magnification" by which the system is studied. It may at first seem contrary to common sense that a system which consists of any finite size building blocks could be scale invariant: if we magnify the system enough we see the individual blocks which by definition cannot look similar to the inhomogeneous system itself. Also, when studying samples of finite size and using low enough magnification we see the whole system itself in different sizes and the scale invariance appears to be broken. Both these observations are correct. However, there may be an intermediate scale region where the scale invariance applies to the intensive i.e. density-like variables.

Why should we in paper physics be interested in scale invariance? The strength of the scale invariance is in the universality associated to it. We know that close to the point where the system under consideration is scale invariant its properties are similar to those of a large group of other systems which thus constitute a universality class.[1] Solving rather simplified problems yields the universal properties of more complex systems, such as paper.

In this article we discuss the formation and the structure of paper. The scale invariance of this inhomogeneous fibre network cannot be exact because of the randomness. However, scale invariance may apply in the statistical sense; all the statistical distributions of the properties describing the structure and its transport behaviour can be invariant. In fact, the great success of scaling in the theory of second order phase transitions, such as ferromagnetism, and in quantum field theories, such as quantum chromodynamics, is based on statistical asymptotic scale invariance.[1]

The statistical scale invariance can be studied most conveniently by examining the correlation functions of the structure. In general, one needs to know all the N-point correlation functions

$$G_N(\overline{x}_1, ..., \overline{x}_N) = Av(m(\overline{x}_1)...m(\overline{x}_N)) \tag{1}$$

where $m(x)$ is the mass density function of the object considered and Av() denote averaging with respect to all possible realisations. If the stochastic process producing the structure is Gaussian, it is sufficient to know G_1 and G_2 only. The autocorrelation function is defined as

$$R_2(\overline{x}_1, \overline{x}_2) \equiv G_2(\overline{x}_1, \overline{x}_2) - G_1(\overline{x}_1)G_1(\overline{x}_2) = R_2(\overline{x}_1 - \overline{x}_2) \tag{2}$$

where the last equality follows from the assumption of statistical uniformity: all the statistical properties are equal throughout the material. In general $G_2(\bar{r})$ is asymptotically an exponential function

$$R_2(r) \sim e^{-\frac{r}{r_0}} \tag{3}$$

where r_0 is called the correlation length. It is obvious from Eq. (3) that if we change the scale by α ($r \rightarrow \alpha r$), we change the functional form of the R_2. But if the autocorrelation function is of power law form

$$R_2(r) \sim r^{-\gamma} \tag{4}$$

the functional form is scale invariant; only the prefactor changes. This change in prefactor is cancelled by scaling m by a factor $\alpha^{-\frac{\gamma}{2}}$. Hence a system is scale invariant if the corresponding autocorrelation function is a power law.

The scale invariant state of the system is approached through the divergence of the correlation length r_0. The number μ describing the divergence is common for a large group of systems, called a universality class. For the divergence we write

$$r_0 \sim (p-p_c)^{-\mu} \tag{5a}$$

where p is a parameter describing the system and p_c its value at the point where the system is scale invariant.

An important example of a system which possesses a scale invariant state is the percolation problem.[2] For example for the two-dimensional square lattice with p being the probability that a bond between two nearest neighbors of lattice is connecting the points, the percolation problem can be phrased as follows:

> "How large a portion p of the bonds of lattice are needed to establish an infinitely large connected geometrical object?"

It can be shown that an infinite cluster of bonds exists if $p > p_c = 0.5$, the threshold value. Furthermore, the autocorrelation function of the infinite cluster is shown to be of the scale invariant form with $\mu = \frac{4}{3}$ and $\gamma = 2 - D$, where $D = 1.73$ is the fractal dimension of the infinite cluster.[2] In the percolation problem the correlation length is the average linear size of the clusters. A third important scaling describing percolation is for the probability that a site is on the infinite cluster:

$$P_\infty \sim (p - p_c)^\nu \tag{5b}$$

The exponents are related to one another through $\gamma = \frac{\nu}{\mu}$. Any network close to its threshold obeys the scaling laws, Eqs. (4-5) with these values of exponents γ and μ.

The transport properties, for example the elastic modulus or strength, of a geometrically scale invariant object also have universal features. The square is not suitable for studying these properties because of its very exceptional symmetry. Instead, one usually replaces the square lattice by the triangular lattice when transport properties are considered. The threshold of the triangular lattice is $p_c = 2sin(\pi/18) \approx 0.3473$, but the geometrical universality class is the same as that of the square lattice with the same γ and μ. The elasticity can be modelled in at least three different ways:

- the Born model which prevents clusters from rotating as a whole;[3]
- the bond-bending model which is the most physical but also the most difficult to use in simulations;[4]
- the central-force model in which there is no energy penalty for the node of the network to move perpendicular to a bond.[5]

The three different models belong to different universality classes of the elasticity. The exponent of interest here is the one describing, how the elastic modulus, E increases from its zero value:

$$E \sim (p - p_c)^\beta \tag{6}$$

The values of β for the three different universality classes mentioned above are: 1.297, 3.8, 3.8.[3-5] The central-force model is peculiar in that the elasticity vanishes before the network becomes disconnected.

The region where universality dominates the network behaviour has been found to be quite wide in the percolation problem. Therefore percolation theory of simple networks helps us to understand the mechanical properties of some real papers, tissue being presumably the most promising material.

When paper is formed out of a suspension, the consistency is close to its three-dimensional percolation threshold value. Thus the flocculation process has some features which are due to the percolation universality.

It is important to know which of the features at the percolation threshold are universal. Basically, all the exponents are universal numbers whereas for example the threshold p_c is not.

In the rest of this presentation we discuss four problems where scaling theory is useful:

- the flocculation at the headbox consistencies;
- the dynamical scaling of turbulence;
- the effect of formation and density on the strength of low-density paper (network theory);[6]
- the optimal use of reinforcement pulp in mechanical printing papers.[7]

The scaling theories being qualitative ones tell us which of the properties and quantities are relevant for the problems under consideration. They help us to attack the problems in the most efficient way rather than solve them. It still remains to be seen whether the scaling guides us towards more practical and useful experiments and simulations or not. Hence the question mark in the title of this presentation.

2 SUSPENSIONS CLOSE TO THE CRITICAL CONCENTRATION

When the volume concentration of fibres in a pulp suspension exceeds a critical value c_{cr}, the fibres form a continuous viscoelastic network. The critical concentration is not a universal number but depends on the width and the length distributions of the fibres. An estimate of this quantity was derived by Thalen and Wahren[8]:

$$c_{cr} = \frac{8\pi A}{[\frac{A}{n} + \frac{n}{n-1}]3(n-1)} \tag{7}$$

where n is the number of contacts per fibre needed for entanglement in the network and A is the ratio of fibre radius to its length. Experimentally, they found that n should be chosen between 3 and 5. The existing Monte Carlo simulations suggest that the critical consistency is given by[9] $2A$. Recently, it has been argued that the critical consistency depends heavily on the history of the sample preparation.[10] One should note that a continuous network is formed at a possibly lower concentration $c_{cr}^{(2)}$, but this does not carry elastic load because of insufficient entanglement.

Below the concentration $c_{cr}^{(2)}$ the suspension consists of individual flocs which are characterised by their size distribution $P(l)$. The percolation theory describes how $P(l)$ scales close to the critical point. In experiments by Smith[11] small scale consistency variations as a function of consistency show rather unexpected behaviour with two maxima and two minima. We repeated these experiments and carried them a step further by studying the spectra of the consistency variations.

The experiment is carried out in the following way. A pulp suspension made in tap water is strongly agitated in order to mix the fibres properly. Then the suspension is poured into a shallow box with a glass floor. At all consistencies the thickness of the suspension layer is the same. The sample is illuminated from below and the pattern of light transmission is recorded with a video camera. With the help of an image analyser the average gray scale and the variations in the gray scales are calculated. The average gray scale vs. consistency is used as the calibration curve for consistency variations. The original image is Fourier-transformed (2-d) with a mainframe and the one-dimensional scanning spectra are determined. The scanning spectrum describes indirectly the floc structure: the position of the maximum is the governing linear floc size and its amplitude roughly estimates how large a portion is in the flocs.

We have chosen to study pulp suspensions instead of laboratory sheets because we want to avoid all shear forces which might break up the floc structure after the initial agitation. The constant thickness of the pulp suspension means that we are varying the basis weight of the corresponding sheet. This does not, however, affect the floc size distribution and the amplitude scales can be compared by correct normalization.

When analysing the pulp suspension with Fourier-transform techniques we cannot separate the isolated parts of the network from the infinite network. The spectrum of randomly distributed objects does not behave singularly at the percolation threshold; it is only the spectrum of the infinite cluster which has exceptional properties at the special point. Therefore we must look for indirect evidence of the percolation threshold. One important parameter describing the state of flocculation is the microscale λ defined in terms of the two-dimensional spectrum $S(\bar{k})$ as[12]:

$$\frac{1}{\lambda^2} = \frac{\int d\bar{k} k^2 S(\bar{k})}{\int d\bar{k} S(\bar{k})} \tag{8}$$

Microscale is very sensitive to the cutoff wavelength, i.e. the resolution but when evaluated with equal resolution it is repeatable and a good measure of the variations close to resolution scale. Differences of the order of 0.03 mm in the value microscale are significant when measured from paper.

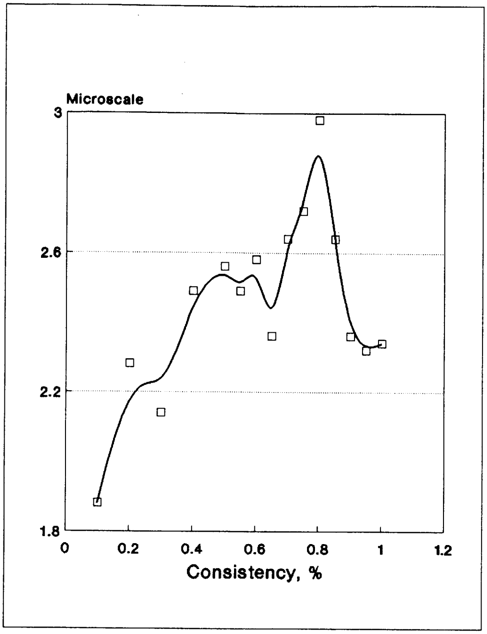

Figure 1. Microscale as a function of the consistency of the pulp suspension.

Fig. 1 shows how the microscale depends on consistency at different cutoff wavelengths in our experiment. We observe that the microscale increases up to consistencies close to 0.7 and then levels off. Qualitatively this can be understood as follows.

When there does not exist an infinite network, the fibres and the flocs can travel long distances during and after the agitation. This allows for phase separation into flocs. The unrestricted movement is especially important for the flocculation after agitation and hence important for the structure if any such movement is strong enough to affect the structure. When the consistency increases, the distances between individual fibres decreases and flocs are more easily produced. Above the threshold consistency the free fibres move in a restricted geometry and separation is therefore also restricted. Thus according to our results on microscale the threshold is 0.7-0.8. This agrees well with the fibre dimensions of our sample pulp.

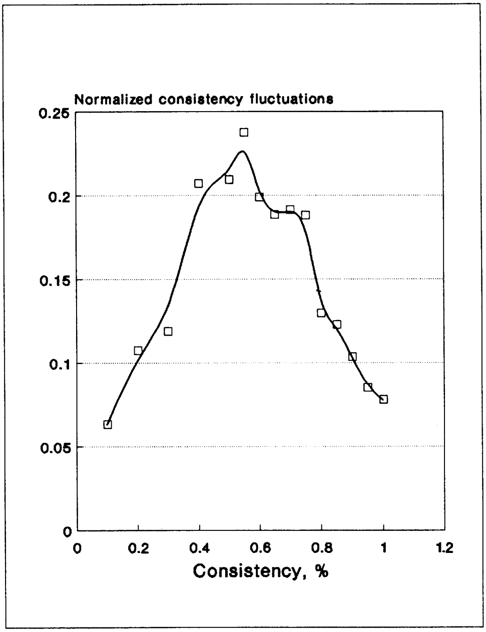

Figure 2. The variance of the consistency fluctuations as a function of consistency scaled to correspond to a sheet of constant grammage.

The standard deviation of the consistency fluctuations was divided by the square root of the consistency to yield grammage independent results. The curve in Fig. 2 shows three regions. At low consistencies the fluctuations increase with increasing consistency. As the flocs are becoming close to one another their movement is restricted. Hence the fluctuations level off. Once the continuous network is formed also during the strong agitation, the infinite cluster restricts the movement of fibres considerably and the fluctuations drop dramatically. The percolation threshold here is around 0.8 % in agreement with the value obtained from microscale, Fig. 1. However, the data are insufficient for deriving any critical exponents.

Above the critical elastic concentration c_{cr} an infinite continuous network which can carry load is formed. This structure is characterised by its elastic properties, such as shear modulus and shear strength. Wahren and coworkers(8) studied these properties both theoretically and experimentally. The theoretical work was based on an approximation which in theoretical physics is known as the effective medium theory:(13) the forces acting on a fibre segment are the ones which the surrounding medium cause on the average and the fluctuations are neglected. Close to the critical concentration the fluctuations are dominating. The percolation theory can be used to study the effects of the fluctuations.

Wahren and coworkers(8) fitted their experimental results for shear modulus and shear strength according to formulas:

$$G = G_0(c - c_{cr})^{\delta} \tag{9a}$$

and

$$\tau = \tau_{u0}(c - c_{cr})^{\nu} \tag{9b}$$

The five numbers G_0, τ_{u0}, δ, ν and c_{cr} were considered as the characteristic numbers of a pulp. (In fact, Wahren and coworkers used the sedimentation concentration instead of c_{cr}. However, by definition it is the c_{cr} below which the shear modulus and the shear strength vanish. Sedimentation consistency is easier to measure but leads to some conceptual difficulties as we show below).

A power law behaviour is also predicted by the percolation theory. The universality at c_{cr} means that the exponents δ and ν are the same numbers for all pulps and therefore only G_0, τ_{u0} and c_{cr} are needed to describe a pulp. This is apparently contradictory to the experimental results by Wahren and coworkers.[8] We suggest the differences in the exponents of various pulps in their experiments to be due to the replacement of c_{cr} by the sedimentation consistency. Mechanical pulps, which roughly speaking consist of two fractions, a fibrous one and the fines which cannot carry load, sediment rather differently from chemical pulps. The relationship between c_{cr} and sedimentation consistency is different for these two types of pulps and this difference produces differences in exponents in curve fitting.

The fact that δ and ν are universal numbers is extremely helpful when classifying pulps. Not only is the number of parameters reduced from five to three but also the remaining parameters can be estimated more accurately from the data. The values of δ and ν can be estimated either numerically or experimentally in simplified systems (for example, consisting of fibres with equal length and radius). The critical consistency is then estimated by extrapolation of both τ and G data and finally the prefactors are obtained by linear regression analysis of $logG$ and $log\tau$ vs. $log(c - c_{cr})$, respectively. We also note that if the functional forms of fibre length L and radius W distributions of two different pulps are equal and the distributions are described by one parameter, the parameter A = W/L is equally good for classification as c_{cr} which is simply a monotonously decreasing function of A.

The analysis above suggests that the considerable amount of work needed to estimate δ and ν accurately would be rewarded.

3 TURBULENCE - ANOTHER SCALING BEHAVIOUR

In Section 2 we considered the properties of a fibre suspension at rest after agitation. During the papermaking process this structure is subject to turbulent shearing which breaks the floc structure. The final structure of the paper is a combination of these two processes.

The modern chaos theory has showed that at the onset (or offset) of turbulence the system shows universal dynamic scaling. Three universality classes exist: chaos via period-doubling, intermittency or Ruelle-Takens scenario.[14] All three are known to exist in Newtonian fluids. During dewatering on the wire section of a paper machine the turbulence decays and reaches the offset point.

The dynamic scale invariance and the corresponding fractal structure in the phase space means that even though the dynamics is unpredictable, the statistical features of the dynamics have a well defined structure. Such a fractal should introduce scaling behaviour in the machine direction as well. It is clear that the closer the point where turbulence is offset is to the wet line the more pronounced is the effect of this scaling. No attempts have been made to detect such a correlation. We suggest the study of the correlations of the basis weight variations in the scale intermediate to the formation scale and the scale where external disturbances such as pressure pulsations dominate.

We also note that in the case of fully developed turbulence there exists asymptotically scale invariance at small distances, which is due to the decay of large externally created eddies into smaller ones. Presumably this happens on the jet or on the forming board but the fibres in the suspension there are still so mobile at this point that the structure, *i.e.* the basis weight variations, does not show this scaling. The expected formation spectrum should behave as[15]

$$E(\lambda) \sim \lambda^{-\frac{1}{3}} \tag{10}$$

at small values of the wavelength λ.

4 ELASTICITY AND STRENGTH - NETWORK SIMULATIONS

4.1 Model

We have simulated the elasticity and strength of random networks in order to study the effects of disorder and inhomogeneity on the mechanical properties of the fibre network constituting the paper. Nissan and Batten[16] have suggested similarly the use of percolation ideas to connect the theory of H-bonded solids to the structural theories. Our network is constructed on an underlying triangular network according to following rules:

- the endpoint and the direction of a "fibre", a linear object with fixed length, is uniformly random on the lattice
- the fibres are independent of each other
- if two or more fibres occupy the same segment of the underlying lattice they are assumed to operate mechanically parallel
- all the fibre segments have the same elastic properties
- the fibres bond to each other with infinite strength
- the segments have a maximum elongation which is common to all the fibres; when the local elongation exceeds the maximum value the segment breaks

The elastic energy to be minimised is the Born Hamiltonian[3]

$$H = \frac{1}{2}\sum_{i,j}[\alpha(\overline{n}_{i,j}\cdot(\overline{u}_i - \overline{u}_j))^2 + \beta(\overline{m}_{i,j}\cdot(\overline{u}_i - \overline{u}_j))^2] \qquad (11)$$

where the sum is over the nearest neighbours of the underlying lattice, $\overline{n}_{i,j}$ is the unit vector connecting the neighbouring points multiplied by the number of fibres occupying the segment i,j and $\overline{m}_{i,j}$ a vector perpendicular to $\overline{n}_{i,j}$ and having the same length. $\overline{u}_i$ is the displacement of the node in the underlying network. Therefore the ratio of α to β describes the relative strength of the extensional stiffness to transverse stiffness. When $\beta = 0$ the network consists of simple elastic strings and is called the central force model.[4] When $\alpha = \beta$ the problem is a scalar one and is called the random resistor network.[2]

4.2 Elasticity

We solved numerically the elastic modulus of the network as a function of the number of fibre segments per number of segments in the underlying lattice, ρ. The size of the underlying lattice varied from 10 x 10 to 40 x 40. The results for the largest lattice size are shown in Fig.3. Two β-to-α ratios and four fibre lengths, l = 1, 2 and 4 were studied. The following conclusions can be drawn:

- the density ρ needed to establish a connected network is lower for higher fibre lengths;
- at fibre length $l = 1$ the scaling behaviour predicted by the percolation theory is extending to fairly high densities;
- at higher fibre lengths the scaling behaviour is less pronounced and the curves appear fairly linear down to the threshold density; however, we also plotted the elastic modulus vs. the probability that a segment is occupied by one or more fibre segments and noticed that this curve shows an equally clear scaling region; all this means that scaling holds at all fibre lengths but the scaling theory is valid in a smaller region around the threshold the longer the fibres; dimensionality arguments support this conclusion;
- if αs are chosen such that the complete singly connected network has a constant elastic modulus at different β to α ratios the curves with a higher value of the ratio rise more steeply; this is in agreement with the fact that if β vanishes the elastic threshold density is lower than the threshold for connectivity;

Elastic modulus is easy to solve in simulations because of the linearity of the fibres. Such simulations could be easily extended to incorporate variations in the elastic modulus of the fibre segments. However, this would not change the qualitative behaviour discussed above. It would be straightforward but much more time consuming to simulate the true

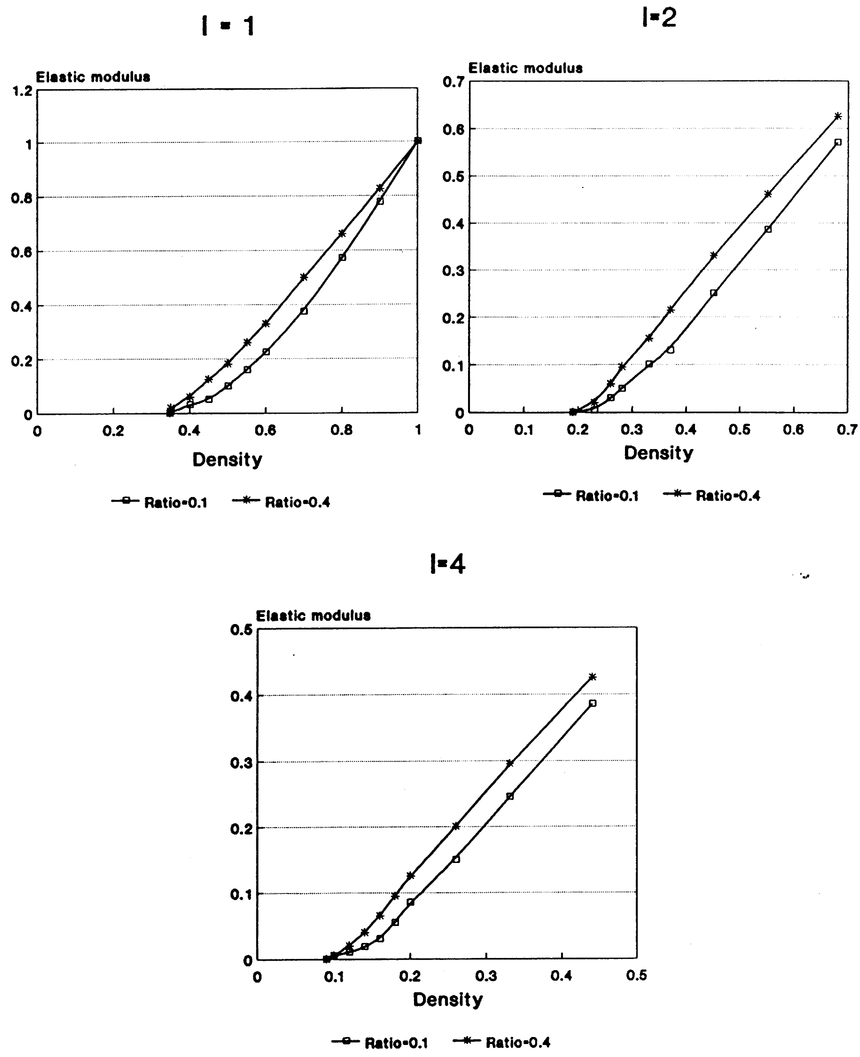

Figure 3. The elastic modulus of the network as a function of density. The β-to-α ratios and fibre lengths are indicated.

layered structure of the fibre network where fibre segments laying on top of each other in an unstrained network can move differently from each other during straining.

Our results show that in order to understand the elastic behaviour of a fibre network the inhomogeneity fluctuations are not relevant except at the very lowest densities. These densities may include the practically important case of tissue. However, even at higher densities one must take into account that the fibre network gets disconnected at finite ρ: the elasticity index is the elasticity divided by the $\rho - \rho_{cr}$ not just by ρ.

4.3 Strength

The strength of the network can be simulated according to the rules presented above. This is much more tedious because we have to solve for the elastic problem between all the breaks of fibre segments. The size of the system was here restricted to 20 x 20 and we studied only three different densities, the ones which had the probability 0.9, 0.7, 0.5 for a segment to be occupied by one or more fibres.

We have chosen all the fibre segments with equal properties, i.e. equal values of α and β and the maximum elongation so that all the deviations from linear elastic behaviour followed by a sudden rupture are due to the geometrical inhomogeneity. Figure 4. shows the generic behaviour of the force-elongation curves we obtained, and a real sample (averaged over 10 networks).

The generic behaviour consists of the following five regions:

- elastic region where no fibres break;
- some weak segments break but they are surrounded by such strong areas that the broken segments do not serve as nuclei for cracks; the broken segments are uniformly distributed over the sample;

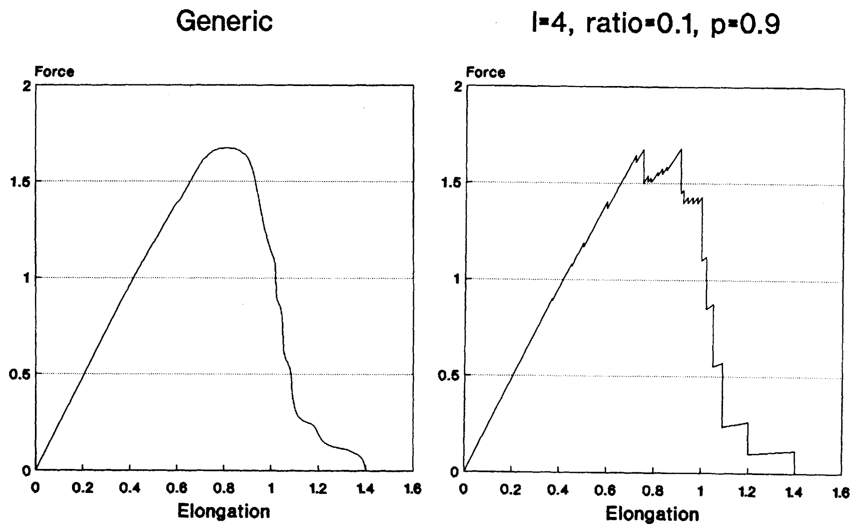

Figure 4. The generic force-elongation curve of the inhomogeneous network.

- fibres which break are so close to each others that the load carried by the network is rerouted in a larger scale, the force-carrying capability of the network stays constant but the network can still be further elongated;

- the final rupture which is characterised by large areas of the network becoming connected to the rest of the system through a single segment and thus unable to carry load;

- a tail which is due to the small size effects and thus unphysical.

The main result of this simulation is that networks in the medium density region have a large "plastic" region even though the fibres themselves are linear up to their point of rupture. This shows that some features of the force-elongation curves of paper can be due to the inhomogeneity of the network.

The force-elongation curve does not predict correct behaviour when the load is decreased: the curve always returns to zero elongation at zero force. The irreversibility is created in the simulations only if the fibre segments themselves behave irreversibly. A simple way to incorporate this is by adding some fibre segments which need a finite elongation before they start to carry load. Once such fibres are activated they are always active.

A drawback of the model we simulated is that bonds between fibres are infinitely strong. In practise, the breaking of bonds is a relevant part of the rupture mechanism. Again it is straightforward to incorporate the breaking of bonds into the model but as this creates interactions between nodes which are not between the nearest neighbours of the underlying lattice, the solution of the elasticity problem becomes more complex. Note that irreversibility can also be built into the model by allowing the bonds to open and the system originally to have internal stresses.

This simulation is a first step towards understanding the relationship between the inhomogeneity and the strength of paper. The model is certainly oversimplified, but serves as a natural starting point. As we have studied networks away from the threshold density, we cannot rely on universality. However, we expect that the behaviour described in Fig.4 is generic.

5 REINFORCEMENT PROBLEM[7]

Printing papers consisting of weak mechanical fibres are reinforced by strong chemical fibres; the most typical example is newsprint. When the consistency of chemical fibres is low enough these fibres do not form a continuous network and thus cannot carry the load without the help of the mechanical fibres. Therefore the strengthening effect in these consistencies is a small one. When a continuous network is formed the strength of the paper is greatly enhanced. We discuss this problem in two extreme approximations:

- below the threshold consistency of chemical fibres the chemical fibres are infinitely strong and the mechanical fibres constitute a continuous medium with finite strength

- above the threshold the medium of mechanical fibres does not have any strength; the chemical fibres have finite strength

We choose these approximations because they are exact percolation problems. In fact, they are the vector versions of the random superconductor network and the random resistor network problems, respectively. Finally we shall relax the assumption that the strength ratio of chemical fibres to mechanical ones is infinite. When this ratio is large enough, the dependence of the strength on the consistency of chemical fibres displays features of these extreme cases.

First we note that if we can assume that the width of the chemical fibres is irrelevant, the only geometric parameter to be considered is

$$\Gamma = cl^2 = l\omega^{-1}m \tag{12}$$

where c is the number of fibres per unit area, l the fibre length, m the grammage of the chemical fibres at fixed grammage of the sheet and ω the coarseness. This is a simple dimensional argument, but can also be derived from a scaling argument. The critical value of Γ, Γ_c is known to be approximately 5.7. This corresponds to a grammage of 0.5 g/m^2. This grammage of chemical fibres must coexist within one layer of fibres in the network, i.e. in 5-15 g/m^2. Hence the critical concentration is 3-10 %, which agrees well with experimental evidence.

In the first case the chemical fibres block the curve of rupture. However, as the network is not infinite, the curve of rupture can find its way through the sheet. When the number of chemical fibres is increased the curve of rupture becomes more and more twisted. The energy required to break the web is proportional to the length of the curve of rupture as the mechanical fibres were assumed to form a continuous homogeneous medium. The work to rupture, T, scales as

$$T \sim (\Gamma_c - \Gamma)^{-\alpha} \tag{13}$$

where the exponent α is determined by the fractal dimensionality of the network of chemical fibres at the threshold Γ_c.

In the second case the network strength vanishes below the threshold. When chemical fibres form a continuous network, a finite amount of energy is needed to break the network. The better connected the network the higher the work to rupture. Percolation theory and a simple dimensionality argument shows that the work to rupture scales as

$$T \sim \frac{(\Gamma - \Gamma_c)^{\beta}}{l} \tag{14}$$

The exponent β is determined by the fractal dimensionality of the backbone of the network at the threshold. The backbone of the network is the part which consists of fibres which are connected at least twice to the infinite cluster.

The two scaling behaviours are shown in Fig. 5a, 5b. When the strength of the mechanical fibres and reinforcing fibres are finite there is a point close to the threshold at which the curve of rupture undergoes no further twisting. Slightly above the threshold it is energetically unfavourable to seek especially weak spots in the network chemical fibres. These conditions can be expressed more exactly in terms of the fractal dimensionalities and relative strength ratios. A practical situation of crossover is illustrated in Fig. 5c. The nonlinearity at the threshold is a sign of the percolation universality. The higher the strength ratio the more pronounced the transition.

In practice, curves like Fig. 5c are observed for both the dry and wet strength of newsprint. We suggest that this behaviour to be explained by the threshold and the universality around it.

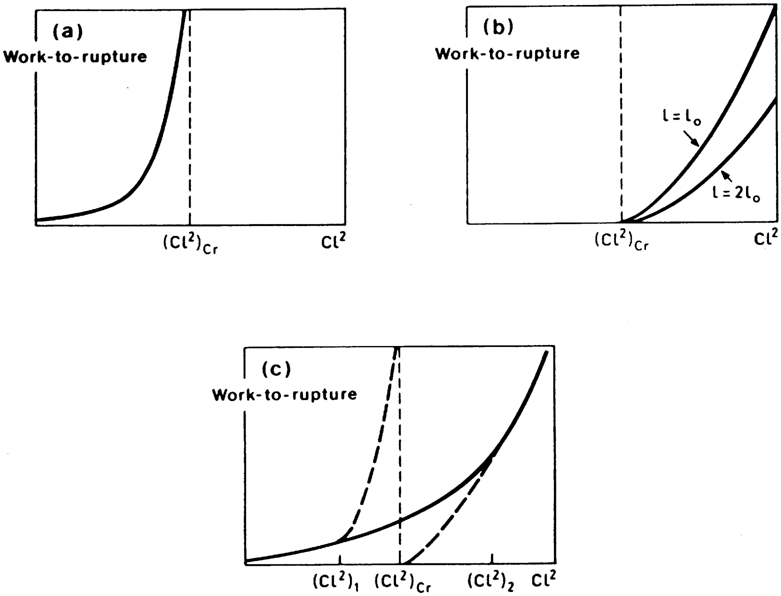

Figure 5. **The two extreme cases, (a), (b) of the reinforcement problem and the crossover (c), which corresponds to the reinforcement of newsprint.**

6 CONCLUDING REMARKS

We have demonstrated in this paper that scaling arguments, percolation theory and network models appear useful when studying the structure of paper theoretically. All the methods used in our presentation are qualitative and hence they do not give us exact techniques for making better paper. Instead, we find them to be useful methods in optimising the simulation and analysis work on the structure of paper.

In this paper we have suggested several problems to be attacked by these methods. We expect network theories to grow into an active field of research in paper physics. The transport properties of networks is a hot topic in theoretical physics. Many of the results obtained there carry over quite directly into paper physics.

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SCALING, PERCOLATION AND NETWORK THEORIES; NEW INSIGHTS INTO PAPERMAKING

R. Ritala and M. Huiku

Dr. B.D. Jordan, PAPRICAN

The world view you are discussing may or may not fit some of the realities. The physics of critical phenomena is a sub field of physics for a very good reason. It only deals with those phenomena that are unique to the situation. When a system moves exceedingly close to a critical point, very large scale fluctuations in the system cause separate phenomena like critical opalescence. Only within this very limited domain near the critical point do the interactions between the fluctuations or clusters grab control of the physics away from the interparticle interactions In our context this would correspond to the interactions between flocs dominating the physics instead of intrafibre or interfibre interactions. Such a situation gives rise to power law scaling. Most things in condensed matter physics take place far enough away from the critical points that they are dominated by rules of their own rather than the interactions between critical fluctuations. I am wondering to what extent we can use this world view of having a picture of the forming process, where adding a few more fibres to a slurry will cause a single massive floc. Instead I suggest a world view where clusters which are far from a critical point quench straight into the paper structure.

Several questions arise:If you have something which is scaling by the fibre length, how does one deal with the fact that you have a distribution of fibre length and therefore your critical point would be, in a sense, a whole ensemble of critical points? Secondly, to what degree does the non-linearity of individual fibres come into this model which is completely dominated by the inter particle topology of the situation. Thirdly, in section three of your manuscript you seem to imply that formation would be fractal. As we discussed before your presentation, I would contend that the formation would not be fractal in reality, and if the power spectrum should exhibit a power law in wavelength over any wavelength range, it would occur only at long wavelength rather than at short wavelength as stated in your manuscript.

Dr. R. Ritala The Finnish Pulp and Paper Research Institute

I did not mean to say that percolation solves everything, but what I meant was that scaling and percolation are very much the reality in fibre suspensions at headbox consistencies. We have a very powerful method to analyze problems, and I think that we can use it in real life applications. I know that we can use it for very low density fibre networks, but there is little real application for this. We can use the same methods to study the reinforcement problems which I did not have time to discuss, but this work was completed and reported about 2 years ago. In that case, there is a percolation transition and we can operate close to that situation. In suspensions we need heavy mixing in fibre suspensions to assure that we are in an equilibrium state, we are not doing any quenching but, in fact, something more like annealing. Distribution in fibre length does not lead to an example of fixed points but is still desirable by a single fixed point.

Dr. R.E. Mark, ESPRI

You stated that "Percolation and network theories yield no new results on elasticity which are of a practical nature in paper physics". Since we work in network theory at Syracuse, I feel constrained to give you 6 examples I have jotted down where new results on elasticity are flowing from network theory.

Firstly, network theory allows you to take into account 2- and 3-dimensional anisotropy, which is not allowed for in your examples of scaling. We offer references (1 - 3) in support of this point. Secondly, we have had a lot of success in matching theoretical Poisson ratios with the experimentation very accurately and we can predict Poisson ratios under a wide variety of circumstances (4). Thirdly, we have a good explanation for the behaviour of lightweight and/or low density sheets -which are very differently, particularly in their strain behaviour in comparison with medium and high density sheets - using network theory (5). By

lightweight and low density, we mean around 25 g/m^2 and/or 150 kg/m^3 or below.
The fourth example is that network theory gives you a better picture of the location of the safe zone of stress, due to the fact that network theory can account for compressive behaviour of the fibres as well as tensile and in fact does so taking into account the orientation of the fibre within the anisotropic sheet (6-8). The fifth example is that network theory has assisted us in devising a new method for the derivation of the in-plane shear modulus of rigidity without the necessity of a difficult experiments (9).

A sixth example relates to the effects of non-linear fibres (10). My question, then, is how can you justify your statement when a lot of useful findings are coming from this area of work?

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Dr. R. Ritala

All this is very well, but what I am saying is that all those properties which you mentioned are model dependent, you do not have universality. Percolation universality does not give us any new insight except in a very a low density case; but I am not sure if anyone is interested. I did not mean that network theories are useless. I simply meant that the universal percolation properties of elasticity in network theories appear in such a narrow region that it is most probably of no practical use. I apologise if I have expressed myself inaccurately in my overhead.

Prof. D. Wahren, Stora Technology

Dr. Ritala, you have provided a new way to look at many things and phenomena. I think I can finally see the forest in spite of all the trees. You appear to have given us a broad outline of basic phenomena which, although not complete, gives a very good start.

Referring to Figure 4 on page 212 of Volume 1, the last line on that page says that the generic curve has a tail which is due to the small size effects and thus unphysical. That tail is exactly what one would see in practice. It looks just like some stress strain curves on short paper strips published by Goldsmith and myself in Svensk Papperstidning around 1968.

Dr. R. Ritala

The reason for my scepticism is that to obtain these curves, we take fairly small systems, study their loading curves and average them out. In this case, that tail is coming from only one or two of those samples. That is why I could not be sure from my simulations that this was real. The averaging means that you have these networks in parallel, and you have the loading curve for this system, but I am not sure about the tail.

Prof. D. Wahren

In physical reality, 4 out of 5 short span tensile measurements would show this behaviour. It is a physical reality.

Dr. A. Nissan, Westvaco Corporation

I am delighted to see that you are bringing this subject to the forefront of paper science. Many of us find difficulty in understanding many of the terms, e.g. scaling, universality, an infinite network or cluster in a finite space or terms such as fractal, fractal dimensions. In your paper, you also talk about period doubling, chaos, and although you do not mention attractors, you make implicit reference to them and again they are infinite in a confined space. Because the books you refer to, and there are many being published, start well into the subject, many of us need a simpler introduction. Would it be possible for you to write an extended appendix for Volume 3 which gives a more detailed explanation of your terms with practical examples we can relate to? I believe that, although the paper industry is a "smoke stack" industry, it is a high tech one and the papers so far demonstrate this, but I cannot use these papers as a demonstration until I can understand all of their concepts.