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FORMATION AND PROPERTIES OF FIBRE NETWORKS

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Synopsis—When elongated particles such as fibres are dispersed in water, they form a continuous network, provided the fibre concentration is above a certain level. A measuring technique utilising a concentric cylinder elasto-viscometer has been developed and used for studies of the mechanical properties of such networks.

Networks generally exhibited the same characteristic properties as solid visco-elastic bodies, hence should be characterised by methods used for such materials rather than by hydrodynamic methods. Nevertheless, a close connection was found between the mechanical properties of fibre networks as measured by quasi-static methods and the hydrodynamic behaviour of the same material, then considered as a fibre suspension.

A mechanism for the formation of fibre networks is proposed, in which the network is considered to derive its strength from the energy stored in the fibres when, after being bent in a turbulent shear field—such as is produced during agitation of the fibre suspension—they are prevented from straightening out by their interaction.

A mathematical model of random three-dimensional fibre networks of low concentration has been evolved. The fibre concentration, the length-to-radius ratio and the modulus of elasticity on bending are shown to be the most significant of the parameters determining the rigidity of the network. Experimental study of the shear modulus of model fibre networks substantiated the qualitative validity of the fibre network model.

Introduction

CONSOLIDATION of the paper web is normally thought of as a process that begins towards the dry end of the Fourdrinier wire. It has long been known, however, that the flow behaviour of suspensions of papermaking pulp fibres differs considerably from that expected of suspensions of independent particles and Forgacs, Robertson & Mason[1] demonstrated in 1958 that the fibres in pulp suspensions form coherent networks, the tensile strength of which can be measured. Accordingly, the consolidation of the paper web must be considered to start with the formation of a fibre network even in the head box of the papermachine. The mechanical properties and the mechanism of coherence of such networks have been the subject of an investigation in this laboratory.

The coherence of relatively dense, dried fibre networks such as are encountered in paper is usually ascribed to interfibre chemical bonds that develop during drying. The coherence of wet paper webs has been shown[2] to be due essentially to surface tension in the three-phase system fibres/liquid/air. The coherence of fibre networks that are two-phase systems (fibres and a liquid) cannot be ascribed to chemical bonding or surface tension, but seem to be due primarily to normal forces associated with stresses in the fibres and to frictional forces produced by these normal forces acting on the fibres at the points of contact between them. The mechanism of fibre network formation can then be conceived as follows.

When a fibre suspension is agitated, the fibres are bent by the action of viscous and dynamic forces. When agitation ceases, the fibres tend to recover their original, unstrained shape. In a suspension with many fibres per unit volume and random fibre orientation, however, the fibres will come into contact with each other as they tend to straighten out. Most of the fibres will make contact with so many others that they come to rest in strained configurations and forces will be transmitted from fibre to fibre. These fibres become interlocked by normal and frictional forces and form a network in which forces can be transmitted through the fibres and from one fibre to another. We have here a typical co-operative system in Volkenstein's[3] sense of the term. In other words, fibre networks are coherent because of internal stresses. In real fibre networks, however, in which forces of attraction between the fibres can be neglected, not all the fibres become actively engaged in the network. Hence, there is still room for forces of chemical attraction to influence the properties of the network, if only to a minor extent.

Although the existence of this mechanism of network formation has not been actually proved, there is some evidence of it. If bundles of straight Perlon fibres or any other man-made fibres of a suitable length-to-radius ratio are put into water, they settle to the bottom of the container and do not form a network, even after gentle stirring with a spoon. If, however, the suspension is vigorously agitated—for instance, with a propeller—the fibres are dispersed randomly and form a network that fills the whole available volume and possesses elastic properties. If this process is repeated, using different viscosities of the suspending medium, networks varying in rigidity are formed, as exemplified in Fig. 1. Here, equal amounts of Perlon fibres were dispersed in sugar solutions of different viscosities and the shear modulus was measured. It is seen that the shear modulus decreases as the viscosity of the suspending medium increases. This finding reflects the change in the relationship between viscous forces from the suspending liquid and elastic forces in the fibres. Thus, it takes longer for the fibres to come to rest after agitation in a more viscous medium; hence, the elastic energy of many of them may be dissipated and unstrained configurations will result. Once the fibres have lost their elastic energy, they cannot become actively engaged in the network however much time elapses.

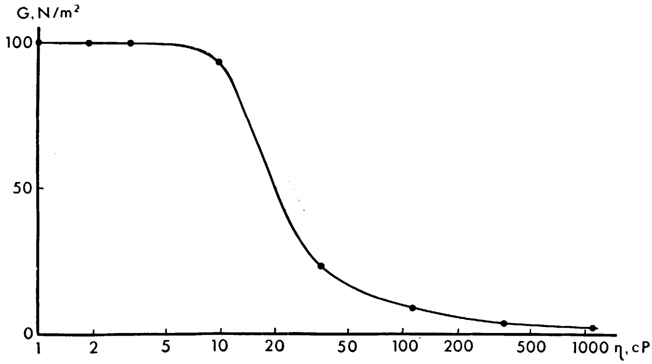

Fig. 1—Measured shear modulus of a Perlon fibre network dispersed in sugar solutions of different viscosities at a fibre volume concentration of 1.5 per cent

A network model

BECAUSE current concepts of fibre distribution have been designed to facilitate description of the arrangement of fibres in paper, most of them are based on two-dimensional or multi-planar models. Examples of these are found in the studies in statistical geometry by Van den Akker[(4)] and Corte & Kallmes.[(5)] A common feature of them is the more or less implicit assumption that three-dimensional assemblies of fibres can be represented by superimposing two-dimensional fibre structures. The sheet formation process would thus be comparable to a sedimentation process in which the fibres settle upon each other. As such a concept cannot fully represent the true state of affairs, a more complete model had to be developed. In 1954, Nimura & Kido[(6)] proposed a truly three-dimensional fibre network model intended to represent the fibre structure in paper and this model was later refined by Onogi & Sasaguri,[(7)] who took account of fibre thickness.

The present model, developed by Meyer & Wahren[(8)] is a further refinement, in which account is taken also of fibre length. The segment length distribution and the total number of segments per unit volume are deduced from the more basic assumption that, for every unit length of fibre, the probability of its receiving a contact point is the same. If the geometric concepts developed by Nimura & Kido[(6)] and Onogi & Sasaguri[(7)] (the latter with some minor modifications) are adopted, it is found that the volume concentration is directly related to the fibre length and radius distributions and the number of contact points per unit volume.

The network model is concerned only with *active fibres*—that is, fibres having three or more points of contact with other active fibres—for, in the absence of attraction forces, at least three points of contact are necessary before a fibre can transmit a force and thus be considered as being entangled in the network. As it has been assumed that every fibre in the population has at least three contact points, the concentration so obtained is a minimum. In practice, there may be fibres that are not entangled in the network (*passive fibres*). An important point is that a continuous three-dimensional network cannot exist below a certain volume concentration. For cylindrical fibres of uniform thickness and length, where A is the length-to-radius ratio, this minimum concentration is—

$$c_v \min = 4\pi A/\left(\frac{A}{3} + \frac{3}{2}\right)^3$$

For large values of A, this expression simplifies to—$c_v \min = 108\pi/A^2$. This expression may be compared with that evolved by Mason[9] for the critical fibre concentration c_0, namely, $c_0 = 6/A^2$; above this concentration, the fibres cannot undergo unimpeded rotation, because they have inadequate space in which to move. Accordingly, at volume concentrations from $6/A^2$ to $108\pi/A^2$, there is an interaction between fibres under dynamic conditions of flow; above $108\pi/A^2$, the fibres can form a continuous network.

The relevance of the network model of Nimura & Kido and Onogi & Sasaguri in respect of the mechanical properties of paper has been questioned by several authors on the grounds that it is not known how the forces are distributed between the individual segments and that no allowance is made for the deformation of the fibre-to-fibre contact areas or the deformation caused by shearing of the segments. Although this criticism seems to be justified, the basic concepts of Nimura & Kido and Onogi & Sasaguri can still be applied to advantage in calculating the mechanical properties of 'dilute' fibre networks such as are encountered in fibre suspensions of moderate concentration, because the 'bonded area' in this case represents only a small fraction of the total fibre length and the average length-to-radius ratio of the segments is high. It can be shown that, when averaged over all relative directions between force and segment, the deformation caused by elongation, compression or shearing of segments having a high length-to radius ratio is much smaller than the deformation by bending. It was therefore decided to neglect the deformation of segments by elongation, compression and shearing and the deformation of the contact areas. In the case of deformation by bending of segments, only the component in the direction of the original shear stress is of interest in a calculation of the shear modulus. This deformation component is proportional to the shear stress per unit area and the third power of the segment length and inversely proportional to the modulus of elasticity, the number of segments per unit area and the moment of inertia of the segment. The total deformation in the direction of the original shear stress is then obtained by superimposing the deformations of individual segments. A quantity proportional to the shear modulus of the network is then obtained, namely, the ratio of the shear stress to the total deformation of the network in the direction of the shear stress. The network shear modulus can then be calculated as a function of the concentration, the dimensions and the modulus of elasticity on bending of the fibres. It was found that the length and radius of the fibres were involved only as the fibre length-to-radius ratio (higher ratios giving more rigid networks) and that the shear modulus of the network is proportional to the modulus of elasticity on bending.

Investigating the mechanical properties of fibre networks

To ENABLE a closer study to be made of the mechanical properties of fibre networks, an elasto-viscometer was designed, employing a Couette-type arrangement of concentric cylinders.[10] This was used for measurements on networks of cellulose and man-made fibres. Qualitative agreement was found between the mechanical properties predicted by the fibre network model and those obtained by measurements on model fibre networks. The report of this study has been published in a series of articles by Thalén and Wahren.[11–13]

Model fibre networks

IN experiments to determine the influence on the network of pertinent factors such as fibre dimensions, modulus of elasticity and coefficient of friction, networks were formed from fibres of Perlon, Teflon or glass and the network shear modulus was measured as a function of the fibre concentration. The fibre dimensions and elastic properties also were measured, as was the sediment concentration after the fibres had settled to the bottom of a measuring cylinder from a very dilute suspension. The Perlon and Teflon fibres were cut to different but uniform lengths whereas the lengths of the glass fibres varied considerably.

The measurements of the sediment concentration showed the presence of a certain number of passive fibres, but also that the sediment concentration is governed by the fibre length-to-radius ratio in the manner suggested by the network model.

Comparison of the shear modulus with the dependence of this property on concentration, as predicted by the fibre network model, disclosed considerable quantitative deviations, but it was evident that there was qualitative agreement. The measured values of shear modulus were lower than the predicted ones. This was ascribed to the fact that, even when the fibres are uniformly distributed, they are not all actively engaged in the network. The length-to-radius ratio was found to be the most important parameter, higher ratios giving more rigid networks. The concentration limits below which no shear modulus was found were the sediment concentrations, which, as mentioned above, were in qualitative agreement with the predicted values. It was also found that for the three kinds of fibres tested the shear modulus of the network was proportional to the modulus of elasticity on bending of the constituent fibres. It was concluded that, although the fibre network model is not sufficiently complete to give quantitatively correct results, it does give the right order of magnitude for the dependence of network shear modulus on fibre dimensions and concentration.

The shear modulus of the network showed no evidence of a variation because of different coefficients of friction between the fibres, apart from the fact that fibres having a low coefficient of friction (Teflon fibres suspended in paraffin oil) were easier to disperse uniformly and thus gave slightly higher shear modulus values than the corresponding Perlon and glass fibres.

Networks of papermaking fibres

As the networks of different cellulose papermaking fibres have different mechanical properties, the elasto-viscometer can be used to characterise pulps by their mechanical properties in the wet state. Yet all pulps have some features in common—for instance, the shear modulus G and the ultimate shear strength of a wide range of papermaking fibre networks can be expressed as a function of the fibre concentration, using a formula of the type $G = G_0(c-c_s)^k$, where c is the fibre weight concentration, c_s is the final sediment concentration reached when fibres settle to the bottom of a measuring cylinder from a very dilute suspension, G_0 and k are parameters characteristic of the particular pulp.

Pulps that are normally beaten to give high paper strength such as kraft and sulphite pulps generally show an increase in network strength on beating, whereas groundwood and high alpha-cellulose pulps show a decrease in this property. These results may be compared with the measurements on Perlon and glass fibres of the same thickness, but cut to different lengths: as these fibres are shortened at constant thickness, there is a fall in the shear modulus and ultimate shear strength at any given fibre concentration. The combined results find ready explanation if it is assumed that, on beating, fibres are to a large extent split into particles having at least as high a length-to-radius ratio as the original fibres. It should be noted here that a comparison of, on the one hand, the values predicted by the fibre network model and the values obtained by measurements on model fibre networks and, on the other, the values for papermaking fibre networks indicates that, if a fibre length-to-radius or length-to-diameter ratio is to be measured for ribbon-like native fibres, it is the thinnest cross-sectional dimension that should be used.

Further measurements on networks of native cellulose and man-made fibres in the elasto-viscometer showed that the fibre networks had much the same properties as those normally encountered in solid visco-elastic materials. The shear modulus of the networks is determined by the concentration and network structure, together with the properties of the fibres, whereas time-dependent effects and damping are determined by the properties of the individual fibres and the conditions at the fibre-to-fibre contact points. The ultimate shear strength is closely related to the shear modulus, but, as measurement of the former involves considerable deformation of the network, the ultimate shear strength may also be dependent on the conditions at the fibre-to-fibre contact points. These conditions were not investigated.

Fibre networks and the formation of paper

THE measurements reported above were carried out under quasi-static conditions, where the possible influences of viscous drag, dynamic fibre-to-fibre interaction, etc. were eliminated. Examining the extent to which a network exists under conditions of flow, Raij & Wahren[14] found that the shear strength of the fibre network, measured under these quasi-static conditions, is a factor of primary importance to the flow properties of pulps in pipes. Resistance to flow in a straight pipe could be conveniently represented by curves in which the ratio of the wall shear stress to the ultimate shear strength τ_u of the network is plotted against the dimensionless factor $(\rho v^2/\tau_u)^{\frac{1}{2}}$, where ρ is the density of the suspending liquid and v is the average flow velocity. Thus, with the network strength as a normalising parameter, the flow data measured at varying fibre concentrations fell within rather narrow limits. As the head box consistency is frequently above the sediment concentration or this concentration is reached in the early part of the wire section, it is reasonable to assume that fibre suspensions exhibit solid body properties already in the above sections of the papermachine and that consolidation of the paper web commences shortly after the slice.

ADDENDUM

I WOULD like to illustrate the active and passive members of a fibre network with two examples.

The first concerns the shear strength of mixtures of groundwood and sulphite pulps. In Fig. 2, the shear strength is on the vertical axis and the curves refer to different average concentrations of the mixtures.

At 2.4 per cent consistency, the mixing curve is almost straight. At higher concentrations, it is S-shaped and the network strength of the mixture can be up to 30 per cent higher than it is for either of the pure pulps alone. The explanation of this phenomenon is probably that the sulphite pulp consists of long slender fibres, whereas the groundwood pulp fibres are shorter, thicker and *stiffer*. At low concentrations, the long sulphite fibres form a coarse meshed network in which the shorter groundwood pulp fibres cannot partici-

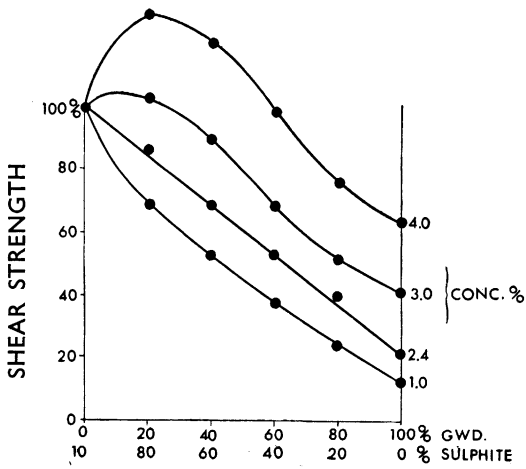

Fig. 2

pate. As the concentration is increased, the sulphite fibre network becomes denser and then the groundwood fibres, which are not very much shorter than the sulphite fibres, become *activated*. As the groundwood fibres are *stiffer*, they will in co-operation with the sulphite fibres give a higher network strength than that of the pure sulphite pulp. The composite network strength will also

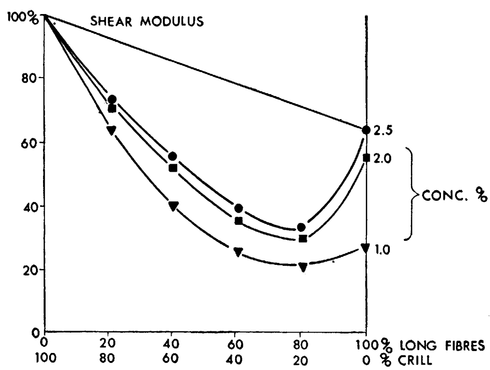

Fig. 3

be higher than that of the pure groundwood pulp, because of the presence of the long, *activating* sulphite fibres.

In Fig. 3, it is apparent that mixtures of long kraft fibres and small crill particles behave differently. The network strength is much lower for the mixture than it is for any of the constituents. We believe that the reason for this is that a coarse meshed fibre network cannot activate the very small crill particles and, conversely, a very finely meshed crill network cannot be considerably

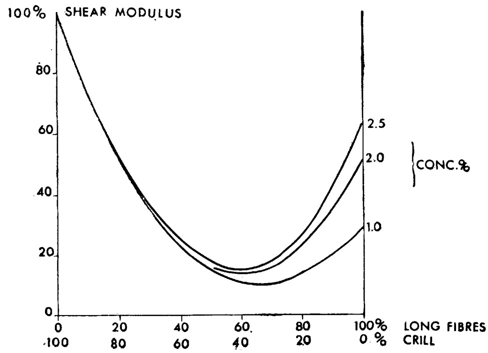

Fig. 4

influenced by a few coarse fibres. Actually, assuming that the crill and the fibre networks are quite independent and that their network strengths are additive, the resulting mixing curves for the network strength can be calculated and are found to give at least qualitative agreement with the measurements. In Fig. 4, these calculated curves are shown.

These examples are to demonstrate the rather intricate interactions taking place. In the first example, the long fibres activated the shorter groundwood fibres; in the second example, particles of incompatible sizes formed independent networks.

During the past few years, there has been a growing interest in measurements of wet fibre flexibility or stiffness. So far, the measurements have been performed on individual fibres and the principle or mechanism of selection may give rise to large systematic errors. The methods and network theories developed at the Paper Technology Dept. have been extended so that the average fibre stiffness can now be measured and evaluated as a bulk property.

Takamura & Bergman, then working at the Wood Research Centre in Stockholm, empirically found a good correlation between the network shear modulus, the concentration and the fibre stiffness measured by Samuelsson's method.

Through dimensional analysis, using Wahren's original assumptions, Almin, Biel & Wahren worked out a better basis for correlating network shear modulus and fibre stiffness. This approach makes it possible to collect in one single graph (Fig. 5) data from measurements on cellulose fibres of different kinds, as well as results of measurements on networks of various model fibres such as nylon, Teflon and glass. (The terms appearing on the horizontal axis result from the analysis.)

Based on this general correlation, wet fibre stiffness can be determined as a bulk property, taking duly into consideration the influence of the fibre length distribution and the distribution of fibre stiffness.

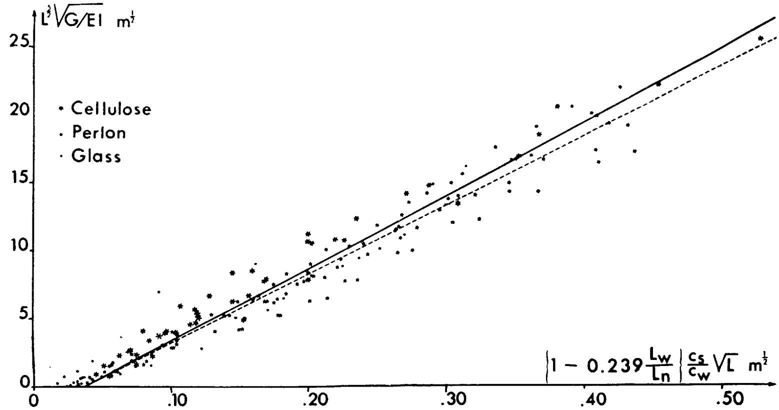

Fig. 5

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Transcription of Discussion

Discussion

Chairman—One of the important properties of the fibre network about which we have heard is the stiffness of the fibre. I would like to ask our speakers to comment a little further on the measurement of stiffness. In the case of a ribbon-like softwood fibre, the stiffness about the two axes of symmetry of the fibre cross-section are quite dissimilar. I suspect that, in most cases, we are concerned with bending about the broad axis of the fibre's cross-section.

Another feature of softwood fibres is the occurrence of structurally weak points at intervals along the fibre's length, which give rise to knee-like joints at these positions under flexural strain. When we talk about the stiffness of such a fibre, we are undoubtedly measuring the stiffness of the knee joints or nodes. In a concentrated network, however, in which the mean free fibre length is less than the node spacing, we are mostly concerned with the stiffness of the fibre segment between the nodes. This distinction could lead to quite different stiffness behaviour for the same pulp at different fibre consistencies.

Prof. B. Steenberg—Stiffness experiments used in the series of data given here are partially made on synthetic fibres, which have no nodes. Diameter and modulus are determined with the fibres in a moist state. All the single pulp fibre stiffness experiments were made on specially prepared pulp and the least possible damage was done to them during defibration, washing and handling. These undamaged fibres were than used for stiffness determinations by Samuelsson's hydrodynamic method.

In the microscope, during stressing, you can see if the fibre bends at any angle to the stream, also whether the fibres are bent. I believe that all sorts of stiffness measurement of single fibres that have been subjected to any mechanical action whatever—such as beating, ultrasonics and even ordinary screening—are useless for this purpose. Such fibres are broken and stiffness measurements cannot be interpreted using theories of elastic beam behaviour. I note that single fibre stiffness is coming up in a number of papers later on.

Dr D. Atack—Would Wahren discuss the behaviour of the networks he has described under dynamic conditions?

Dr D. Wahren—All of our measurements so far have been performed under static or quasi-static conditions. Measurement in flow is something that I would like to start on now, but we know very little. Some measurements we made indicate that, in conventional head boxes running at fairly high speeds, there is no network at the slice; whereas it is very hard in pipe or channel flow to avoid network fragment formation at papermaking consistencies.

Mr J. Mardon—When one examines fibre networks at head box consistencies, there is a break-point, depending on the length of the fibres, the speed of flow and concentration at which the inherent flocs that correspond with the network disappear and this is very sharply defined. When flocs are present, they can be destroyed by the acceleration going down to the slice; but, if they are formed by some element of the head box, such as a perforated roll, it is actually possible with the Edgerton flash to photograph the network of fibres as they go through the slice, if you have a transparent slice.

Dr H. K. Corte—How can you be certain that the fibres maintain the same contacts with each other and with the cylinder walls when the network is sheared?

Dr Wahren—During our measurements, deformations have been small and there is no possibility of telling from the recordings what is actually happening. In turbulent flow, the network is subjected to large deformations. Contacts are being broken. New contacts may be formed, especially, of course, if the turbulence is allowed to decay.