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Performance of Reinforcement Fibres in Paper

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Paper properties can be controlled by mixing different furnishes. The outcome of the elastic, strength and toughness properties is analyzed in this work using results from other fields of material science, particularly from composites. We discuss the micromechanics of reinforcement fibres, their conformability to the background fibre web and the fracture processes in reinforced paper. Reinforcement fibres should have high ductility and they be similar to the mechanical furnish in their micromechanical stiffness.

I. INTRODUCTION

Reinforcement fibres (RIF) are added to paper to improve its mechanical properties and runnability on the paper machine, in converting or end-use operations. High fibre length, conformability, bonding capacity, strength and perhaps ductility are believed to be essential for a good RIF. They increase the fracture toughness of paper, its ability to resist crack propagation. Much the same fibre properties also increase the tensile strength and elastic modulus of paper.

Reinforcement pulps are relatively expensive and therefore used in low concentrations. High-performance RIFs could offer potential savings in the furnish costs. Unfortunately experiments where the RIF mass fraction is varied are not only tedious but also difficult to interpret. The properties of papers with varying RIF content show often, though not always nonlinear behaviour and the reasons for the nonlinearities are not understood. One does not know if the addition of RIF has the same effect in the machine-made paper as it has in handsheets. Since the RIF performance cannot be accurately characterized , RIFs are usually compared simply by measuring the properties 100-% RIF handsheets. One assumes then that the RIF with the best properties in the pure handsheet also gives the best reinforcement effect when mixed with any mechanical pulp.

Different explanations have been given for the non-linear behaviour of the properties of pulp mixtures as a function of the RIF mass fraction. According to the perhaps most common explanation the bonding properties of mechanical and chemical pulps differ. There are then three kinds of bonds in the mixture. Their relative proportions vary when the RIF fraction is increased and lead to the nonlinear changes in the paper properties. The nonlinearity would be most obvious if the mechanical and chemical pulps formed two nearly independent fibre networks [1]. This is unlikely to happen in reality, among other things because the uniform distribution of fines in all bonds tends to smear out the bonding differences that pure pulps would have.

Another possible explanation is that only the location of the failure point on the load-deformation curve is nontrivial and otherwise the shape of the curve for a pulp mixture can be calculated as the appropriately weighted average of the curves describing the components [3]. However, then the elastic modulus should be linearly related to the RIF content, a postulate that can be easily demonstrated to be invalid. It is also quite possible that the sheet structure itself and particularly the relative bonded area is nonlinearly related to the RIF content. For example, the degree of collapse of the RIF could be a nonlinear function of the RIF content [2].

Published studies of RIF performance in paper ([1,3,2]) have all simplified the real situation by assuming that the network geometry or the mechanical properties of the mechanical pulp fibres do not change when the RIFs are added, or that the mechanical properties of the mixture are given by the average of the two components. In other fields of material science, particularly composites and disordered systems, it is well-known that none of these assumptions is generally valid. In this paper we use physical arguments in an effort to analyze systematically the property requirements of RIF. The treatment is divided into three parts: geometry, mechanical compatibility and toughness mechanisms.

The geometrical aspects of the problem arise from the need that the RIFs should tie together the matrix of the other (mechanical) fibres. Much confusion has arisen from the incorrect assumption that there would be an abrupt change in paper properties precisely at the percolation consistency of the RIF. Furthermore, the performance of different RIFs in a given mechanical pulp network network can be much more dependent on the relative mechanical properties of the fibres and bonds than it is on the geometric percolation of the RIFs. The application of the two-dimensional percolation problem to paper [4] is well-known but it is invalid if delamination of the fibre network affects the reinforcement mechanism. One must then consider the full three-dimensional problem where the RIF's are bonded directly to one another.

We have developed a numerical simulation model [5] that can be used to study this. The model includes fibre conformability as a parameter. We show how the relative area of different types of bonds depends on the RIF content and sheet grammage.

Next we consider the issue of mechanical compatibility. Its significance is demonstrated by the fact that with the addition of a RIF pulp to mechanical pulp it is not uncommon that the tensile properties of paper decrease at first and increase only later. The phenomenon is well-known in composites. What happens at the fibre level is that it is easier for the network to strain more at the "soft" - low-modulus - mechanical fibre matrix and less at the stiff RIFs. As a result the RIF's do not carry "their share" of the external load and the matrix fails at a *smaller* load than it would in the absence of the RIFs. The opposite phenomenon, transfer of strain from RIF to matrix, happens if one adds low-modulus fibres to a proportionately stiff matrix where the network modulus does not decrease in direct proportion to the concentration of the soft fibres.

It is well documented that the conductivity (i.e. electrical analoque of elastic modulus [6]) of disordered binary mixtures differs from the linear 'rule of mixtures' behavior only if the conductivities of the two phases are 'sufficiently' dissimilar [7]. Things become more complicated if one considers strength properties. For instance in the computer simulations of Duxbury et al. [8] of resistor networks, percolation-related scaling behavior was observed only if the elastic moduli *and* strength of the two components were very different. When the components were similar in properties, no such effect could be seen.

Finally we analyze the reinforcement mechanisms related to fracture toughness at the fibre level. Regarding the RIF performance the worst case scenario is the one discussed above where the RIF prematurely trigger the matrix failure. The situation is better when the RIFs pull out of the mechanical fibre matrix. This is analyzed in terms of a stochastic theory for fracture toughness. Perhaps the "best" situation is achieved if the RIF are not too stiff but ductile and stretch plastically at the same time as the bonds fail one after another. The phenomenon one then encounters is called crack-bridging. We also discuss crack deflection that dissipates energy at the fibre level and thereby imbedes crack propagation.

II. CONNECTIVITY

The non-linear behaviour of paper properties that is often observed with RIF addition could arise from geometric factors. Either there may be a transition related to the percolation of the RIF or the bonding degree of the different furnish components may be non-linear. The quantitative consequences of these explanations are considered in the next two chapters.

A. RIF percolation

Suppose that the non-linear relationship of paper properties to the RIF content η is caused by a percolation transition, which occurs at a critical value η_c. One would then expect a noticeable change (a bend in the curve etc.) [4,9] at some point well above the transition point so that the concept of 'percolation behavior' in paper properties is rather ill-defined [10,11]. One does not know how many bonds per fibre are needed for the change to occur or what is the critical RBA-value. Naive application of elastic modulus simulations [12,13] suggests that a change in the mechanical properties of paper happens at 2–4 times the percolation threshold but this merely a guess. In any case, on this basis the cross-over in mechanical properties occurs at ca. 10 bonds per a RIF since at the percolation threshold there are, on the average, 2.7 bonds per fibre [12]. Notice that 10 bonds per fibre is far from a 'dilute' limit of non-interacting RIF. The corresponding RBA is given by

$RBA_x = 10w_f^2/2l_f w_f = 5w_f/l_f$ where l_f and w_f are the length and width of the RIF. Scandinavian pine has $l_f = 2.5$ mm and $w_f = 40$ μm, which leads to $RBA_x \cong 0.1$ as a rough order-of-magnitude estimate of the cross-over point.

Regarding the bonding of RIF there are three alternatives when the RIF network becomes stiff:

- assume that the ZD shear stiffness of the entire network is high. Then it is enough that the 2D projection of the RIFs forms a percolating network.
- assume that mechanical fibres prevent strong bonding between the RIFs but that fines mediate or even reinforce the RIF-RIF bonds.
- assume that only the pure RIF-RIF bonds are strong enough. This includes RIF-RIF bonds via chemical fines but not via mechanical fines

The first case is essentially a two-dimensional problem. The presence of the mechanical pulp can be ignored. The percolation threshold [14] can be expressed as a critical grammage m_c:

$$m_c = 5.71C/l_f \tag{1}$$

where C is fibre coarseness (mass per unit length). Using typical values for RIF we find $m_c = 1g/m^2$ so that a change in the mechanical properties of paper might be observed when the RIF grammage is 3 g/m^2 (assuming 10 bonds per fibre at the cross-over). It is important to realize that this threshold is completely independent of the overall grammage of the sheet. In terms of the mass fractions, the threshold means less than a $\eta_x = 5 - 10\%$ RIF content in printing papers. We use the subscript "x" to emphasize the difference from the geometric percolation point.

In the second case of RIF-RIF bonding with intervening fines allowed, there are two factors that complicate the evaluation of RIF connectivity. First, the finite bending stiffness of fibres makes the sheet structure porous. The bending stiffness couples to the thickness of fibres and other particles: if the thickness of all particles were zero then fibre stiffness would be irrelevant and the network would be two-dimensional. The other complication is the *shielding* of RIF-RIF bonds by mechanical fibres that happen to lie between two RIFs that would bond together otherwise. Both of these problems can be dealt with using numerical computer simulations.

We have developed a simulation model [15], "KCL-PAKKA", that "packs" fibres, fines and fillers in to a random structure closely resembling real paper. In the model, fibres are deposited at random and one by one on a two-dimensional square lattice of 10 by 10 μm cells. The discretization does not affect the generic features of the statistical network geometry. The fibres have a given length, width and thickness, l_f, w_f and t_f, as well as "flexibility" T_f. When adjacent cells are compared, fibres can be displaced in the z-direction, up or down, by an amount not exceeding T_f. The fibres are pressed down onto the underlying, already formed network as far as possible while still obeying the flexibility constraint T_f. More details of the simulation model can be found elsewhere in these proceedings [5].

Figure 1 shows the computed RBA of a Scandinavian pine kraft pulp when mixed with a groundwood pulp. It is assumed the GW fines can mediate the RIF-RIF bonds. At grammages above 30 g/m^2, a RIF content of $\eta_x = 10\%$ is enough to reach the cross-over threshold $RBA_x = 0.1$. At lower grammages a higher fraction of RIF would be needed. In our simulations, Fig. 1 would remain practically unchanged if GW were replaced with a Scandinavian TMP or PGW. However, it is quite possible that there are other mechanical furnishes that would lead to different results. In Fig. 1 RBA is essentially linear as a function of η. Hence if the threshold RBA_x were higher, say by a factor of two, then one would need $\eta_x = 20\%$ for the cross-over. A higher threshold would apply to shorter or wider fibres. A higher threshold would also be justified if the above estimate for the location of the cross-over point was simply too low.

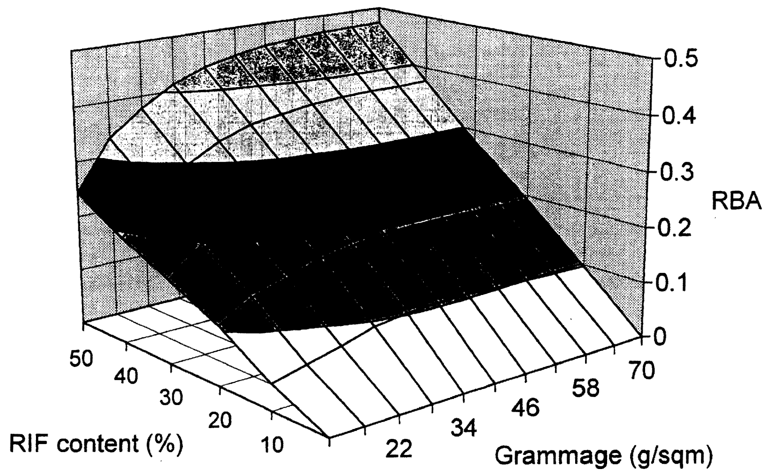

FIG. 1. The relative bonded area computed with the KCL-PAKKA model for a mixture of a Scandinavian pine kraft pulp and a groundwood pulp (CSF42). Both furnishes are modeled with four Bauer-McNett fractions (+28, +48, +200, -200), whose fibre properties have been adjusted in order to match the optical properties and bulk of the model sheets with the values measured from real handsheets made of the corresponding SWK and GW. The computed *RBA* includes the SWK-SWK bonds between three of the fibre fractions either directly or with intervening SWK or GW fines.

In the third case listed above, one accepts only RIF-RIF bonds with no intervening mechanical pulp component. In addition to pure RIF-RIF bonds, bonds mediated by chemical fines are accepted. In this case the corresponding RBA is much smaller than before (Fig. 2) and η_x = 33% is required for the critical level of $RBA_x = 0.1$ almost at all grammages. $RBA_x = 0.2$ would imply η_x = 54%. The crucial difference to the previous case is the shielding of the RIF bonds by the mechanical pulp. This effect depends on the surface area per unit mass of the mechanical pulp. (N.B. fillers would shield bonds in the same way.) In this case there is more variability between mechanical pulps than above. For example, if GW would be replaced with TMP in Fig. 2, $\eta_x \leq 30\%$ would suffice to reach $RBA_x = 0.1$.

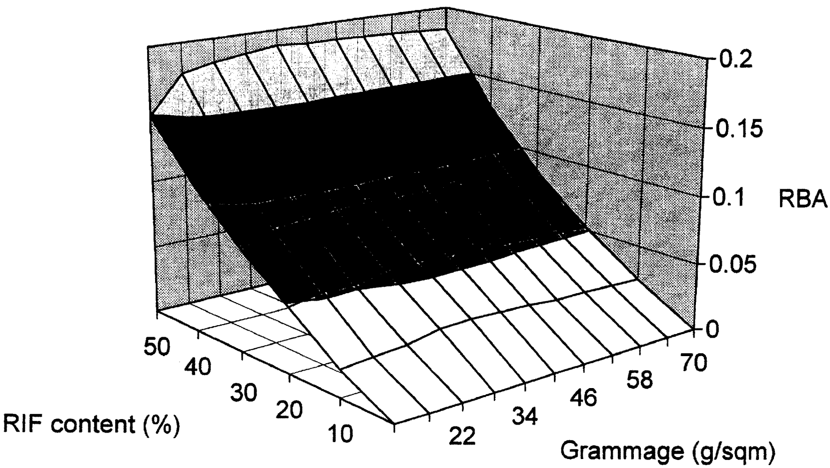

FIG. 2. The same as Fig. 1 except that now the SWK-SWK bonds with intervening GW fines are excluded from *RBA*.

To summarize, the stiffness threshold η_x of the RIF network depends on nature of the bonding between the RIFs. The quess of $RBA_x = 0.1$ leads to $\eta_x = 30\%$ when pure RIF-RIF bonds are necessary and to $\eta_x = 10\%$ if intervening fines (of any type) do not weaken te bonds too much. In the other extreme is the case where percolation the in-plane projection of the RIF suffices. The stiffness threshold is then expected at the RIF grammage of 3 g/m^2; for any overall grammage of the sheet. It must be understood that these estimates are based on a guess about the distance from the percolation point to the concentration at which a non-linear change in paper properties occurs.

B. Relative bonded area against RIF content

In this section we demonstrate that changes in the RIF content η can lead to non-linear behaviour in the bonding degree of the RIF and the mechanical fibre furnish. Since the *RBA* of furnish components cannot be readily measured we use the KCL-PAKKA model for demonstration. The computed *RBA* of RIFs and mechanical matrix against η at 52 g/m^2 are shown in Fig. 3. The bonding degree of each components includes all bonds of the fibre to other fibres, independent of the type of the fibre with which the bond is formed. Bonds with intervening fines particles are included but the bonded area of fines is not included in the calculation of *RBA*. Also shown in the figure is the *RBA* -curve for pure RIF-RIF bonds.

It is interesting to observe that the *RBA* of both the RIFs and the mechanical fibres increases almost in the same manner when the the mass fraction of the former increases. The bonding degree of the mechanical fibres increases because they form bonds more easily with the flexible RIF than with other mechanical fibres. Hence there is a synergistic effect in the *geometry* of the network when η increases. Only in the pure RIF-RIF bonding the shielding by the mechanical furnish causes the antagonistic effects with increasing η.

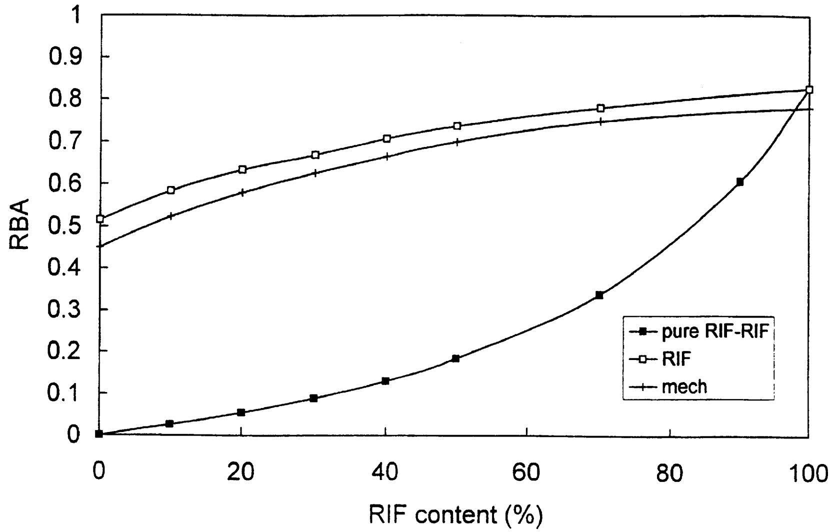

FIG. 3. The relative bonded area computed with the KCL-PAKKA model for the same SWK and GW furnishes as in Fig. 1, at grammage 52 g/m^2. The total bonded area of the SWK fibres and GW fibres to any fibre type is given by the open squares and crosses, respectively. Bonds mediated by fines are included in both. The black squares show the same case as Fig. 2.

The RBA curves of the RIFs and the matrix could be used to calculate the mechanical properties of paper as a function of η. For example, the shear-lag equation [16] could be applied to the elastic modulus of both components and the Page equation to the tensile strength. These would then be weighted by the mass fractions η and $1 - \eta$. The net result of such an exercise would give the same convex curvature for the η-dependence of the mechanical properties as is shown in Fig. 3 for the RBA components. Only the overall trend would vary depending on the properties of the pure one-component sheets. The situation would be different if the pure RIF-

RIF bonding had an additional, separate effect on the mechanical properties of the mixture.

The above results have been calculated for mixtures of a softwood kraft and a groundwood pulp. The quantitative details and even the curvature of the curves in Fig. 3 could change if different furnishes were considered. In particular the curvature would change if the collapse of RIF would occur only above a critical RIF content, as Fernandez and Young [2] propose happens in mixtures of TMP and springwood SWK. In that case an antagonistic η-dependence would be observed in RBA. However, in the absence of such a co-operative effect in the network geometry it seems probable that the synergistic effect displayed in Fig. 3 is common to mixtures of slender flexible fibres and coarse stiff fibres.

In summary, the relative bonded area of pulp mixtures is probably always somewhat nonlinear as a function of the mixing ratio and this causes deviations from the linear "rule-of-mixture" behaviour of paper properties. Unless there are abrupt changes in the network geometry at some critical η, the changes in RBA alone seem to imply smooth and *synergistic* deviations from the rule-of-mixture behaviour of the mechanical properties of paper.

III. FIBRE COMPATIBILITY

When a network containing mechanical fibres and reinforcement fibres is strained, the stress and strain state of the two components are different. Also the state of the mechanical fibres in the vicinity of the RIFs is different from what it would be in a homogeneous mechanical fibre network. These effects cause an additional and antagonistic deviation of paper properties from the RBA-dependence discussed above. The first effect determines the RIF contribution to the modulus and the second the contribution to the strength and toughness of paper. We consider first the phenomena at the dilute limit $\eta \approx 0$ and then at higher concentrations.

A. Micromechanics in the Dilute Limit

The standard model for fibre composites and paper is the Cox shear-lag theory [17,18]. In the theory stress is transferred to fibres through shear deformations of crossing fibres, inter-fibre bonds or a continuous matrix. The shear-lag calculations predict that the contribution of a fibre to the elastic modulus of a sheet is proportional to the fibre modulus multiplied by a geometric orientation factor and a correction factor for the inactive fibre ends. In composite mechanics the inactive fraction of fibres is characterized by a critical length [18]. Long RIFs increase the network modulus more than short RIFs because the critical length is 'relatively' shorter in the first case.

The shear-lag theory and the critical length are convenient if one wants to explain qualitatively why or how the elastic properties of fibre composites depend on the fibre and matrix moduli, fibre length and fibre content (mass fraction). In random fibre networks such as paper the theory predicts qualitatively, but not quantitatively [12], how the elastic modulus depends on density. The theory even gives a roughly correct form for the variation of *average* axial stress along the fibre axis [12,19].

Shear- lag theories also predict high shear stresses at fibre ends if either the fibres are long or RBA is high. The high shear stress arise from that the stress transfer occurs only at the fibre ends. However, recent numerical calculations have revealed that this picture is incorrect in random fibre networks [13]. Because of the disordered geometry stress transfer to and from a fibre occurs in random increments all along the fibre. In addition, direct transfer of axial stress between crossing fibres – instead of shear deformation of the crossing fibres – seems to be the dominant mechanism [13]. The failure of the shear-lag theory has also been demonstrated experimentally and theoretically in fibre-reinforced composites when the fibre content is high [20,21]. Again, axial stress transfer has been put forward as the explanation for the breakdown of the shear-lag theory [22].

In the case of paper the failure of the shear-lag theory would perhaps not be too serious if one concentrated on the elastic properties. However, the strength properties of paper and particularly reinforced paper cannot be derived from the shear-lag theory. This is because the failure of inter-fibre bonds is directly coupled to the stress transfer between fibres. There is an upper limit on the load that a bond can transfer from one fibre to another without failing. Hence shear-lag theories would predict that breaking strain goes to zero when RBA increases.

We construct next a simplistic model in order to demonstrate the axial stress transfer from the mechanical fibre matrix to the RIF. A more careful analysis of will be given elsewhere [23]. Consider, for simplicity, a RIF and two crossing matrix fibres all parallel to external elongation (Fig. 4). The disturbance in the matrix is strongest in this case. Only the fibre crossings at the ends of the RIF are included. This is a resonable approximation at high RBA [13].

We assume that the outer end points of the crossing segments (marked with P in Fig. 4) are displaced homogeneously. This is the same boundary condition as used in the Cox theory. The axial strains in the P1-P1 chain in Fig. 4 are coupled through

$$(l_f + 2l_s)\epsilon_x = \epsilon_f l_f + 2\epsilon_m l_s \qquad (2)$$

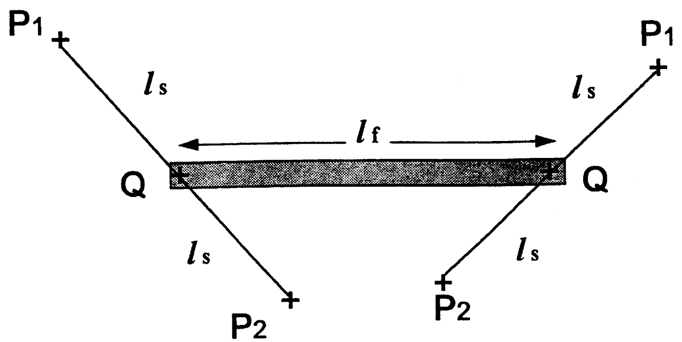

FIG. 4. Illustration for the axial stress transfer calculation. The reinforcement fibre is shown by the thick bar Q-Q and the two crossing mechanical pulp fibres by the lines P1-P2. For clarity the latter two are drawn at an angle to the RIF even though in the calculation they are taken parallel to the RIF and hence to the external elongation.

where l_f is the length of the RIF (assuming high RBA), l_s is the length of segment Q-P1, ϵ_f and ϵ_m are the corresponding strains and ϵ_x is the external strain. The axial strain of the segments Q-P2 is $2\epsilon_x - \epsilon_m$ since the average strain of Q-P1 and Q-P2 must equal to ϵ_x for the homogeneity assumption to hold. The force balance equation reads

$$2(\epsilon_m - \epsilon_x) = S\epsilon_f, \tag{3}$$

where S is the RIF-to-matrix stiffness ratio

$$S = E_f A_f / E_m A_m \tag{4}$$

with $E_{m,f}$ denoting the elastic moduli of the matrix fibres and RIF, and $A_{m,f}$ their cross-sections.

The strains in the RIF and matrix are now given by

$$\epsilon_f = \epsilon_x(1 + Sl_s/l_f)^{-1} \approx \epsilon_x(1 - S\frac{l_s}{l_f}) \tag{5}$$

and

$$\epsilon_m = \epsilon_x(1 + \frac{1}{2}\frac{S}{1 + Sl_s/l_f}) \approx \epsilon_x[1 + \frac{S}{2}(1 - S\frac{l_s}{l_f})] \tag{6}$$

where the approximations apply for $l_s/l_f \ll 1$, which is valid for all ordinary papers. The stiffness ratio S must be reasonably close to unity when ordinary wood pulps are considered and thus also $l_s/Sl_f \ll 1$.

In the simplest approximation the relative contribution of a RIF to the elastic modulus of paper is proportional to $S\epsilon_f/\epsilon_x = S(1-S\frac{l_s}{l_f})$. The result would of course change quantitatively if fibre orientations and RBA were included. The modulus benefit is proportionately smaller if the RIF is very stiff (S high). Notice also that in this approximation the matrix disturbance does not affect paper modulus because the average strain in the two segments Q-P1 and Q-P2 equals ϵ_x.

Before proceeding it is instructive to apply the above equations to the homogeneous case where all fibres are alike, or $S = 1$. According to Eq. (6), the highest axial strain in the network is $\epsilon_m = 1.5\epsilon_x$ as long as $l_s/l_f \ll 1$. In the Cox model the highest axial strains are roughly equal to ϵ_x. The strain difference between the segments Q-P1 and Q-P2 (Fig. 4) is equal to ϵ_x. This implies that the first inter-fibre bonds in the network would fail at

$$\epsilon_x = \epsilon_c = A_b\tau_{crit}/A_mE_m \tag{7}$$

where A_b is the average bond area and τ_c the bond shear strength. The critical strain would even be somewhat smaller if the stress transfer were shared by more bonds than just the two at fibre ends. The important point is that ϵ_c does not go to zero when RBA increases as it would if the Cox model was applied.

Returning to the dilute RIF mixture, the above analysis implies that the matrix can fail prematurely because the matrix strain ϵ_m around the ends of RIFs increases with increasing S (Eq. (6)). The change would be observed as a decrease in the ratio of tensile strength to elastic modulus which is the elastic breaking strain of paper ϵ_{el}. This is similar to reinforcement-induced matrix failure in composites [24]. In composites stiff fibres may also debond prematurely from the matrix. The same may happen in paper if the RIF-to-matrix bonding is weak. From the above analysis it follows that the external strain at which the debonding would occur is inversely proportional to the RIF stiffness, i.e. stiff fibres debond first.

When the matrix fails prematurely at the ends of a RIF the change $\Delta\epsilon_{el}$ in the elastic breaking strain of paper can be assumed to be linearly proportional to the relative number of RIF in the sheet in the dilute limit $\eta \ll 1$. At the same time the addition of the RIF raises the RBA of the network (cf. Fig. 3) and consequently the elastic breaking strain of paper. The deviation that the premature matrix failure causes a strain reduction given by

$$\Delta\epsilon_{el} = const \times \eta(1 - S)/(2 + S) \tag{8}$$

where the constant takes care of statistical geometry and the relative weights of the RIF and mechanical fibres. If debonding of the RIF would contribute to the sheet failure, the factor $(1 - S)/(2 + S)$ should be replaced with $(1 - S)/S$. Both these factors are displayed in Fig. 5(a). The combination of the strain reduction $\Delta\epsilon_{el}$ with a smooth RBA dependence is illustrated in Fig. 5(b). Here $\epsilon_{el}(S = 0)$ is assumed to be linearly proportional to $RBA(\eta)$ as given by Fig. 3 and typical values, ϵ_{el} =0.8% and 1.2% are used for the pure GW and SWK, respectively. $\Delta\epsilon_{el}$ is assumed to be constant, $\Delta\epsilon_{el} = \eta \times 0.6\%$.

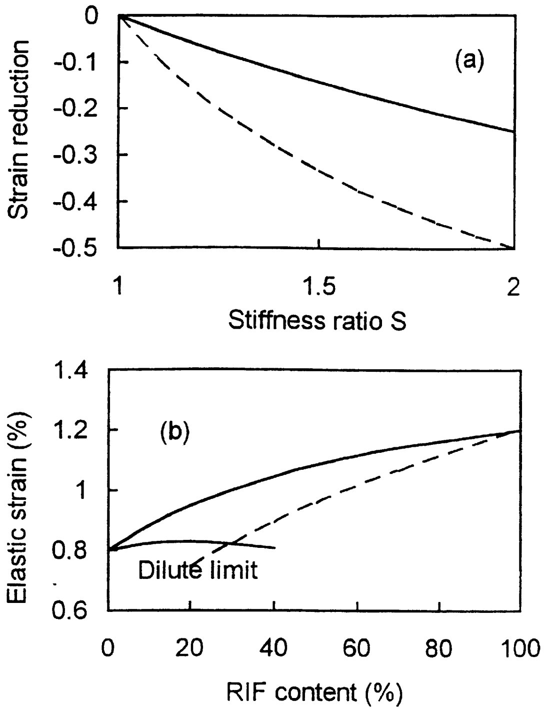

FIG. 5. In (a), the relative change in the strain reduction $\Delta\varepsilon_{el}$ against the modulus ratio S, (1 - S)/(2 + S) and (1 - S)/S, for matrix failure (solid line) and RIF debonding (dashed line), respectively. In (b) a schematic illustration of the elastic breaking strain of the SWK-GW mixture of Fig. 3. The solid line is calculated from the RBA-curves of Fig. 3, as explained in the text; the dilute limit result results from adding $\Delta\varepsilon_{el}$, and the dashed lines shows the cross-over from the dilute limit to full matrix confinement at high η.

It must be emphasized that the behaviour plotted in Fig. 5(b) is a qualitative illustration. The magnitude of the strain reduction $\Delta\epsilon_{el}$ cannot be evaluated quantitatively from the simple model presented above. On the other hand, if the elastic breaking strain of paper against the RIF mass fraction η is measured, the result can be used to assess the mechanical compatibility of the RIF with the mechanical fibre matrix. Additional information can be obtained from the elastic modulus of paper E. According to the simple model above, poor mechanical compatibility has only a weak effect (of the order of $1 - Sl_s/l_f$) on the elastic modulus. Therefore the behaviour of the elastic modulus of paper as a function of η should be linearly related to $RBA(\eta)$. In the absence of premature matrix failure or RIF debonding, the elastic breaking strain of paper should follow a similar dependence on η with the same curvature. If in contrast ϵ_{el} falls in the dilute limit below the trend of elastic modulus then a reduction in the axial stiffness of the RIFs could help.

B. Finite RIF concentrations

When the concentration of RIF in the mechanical fibre matrix increases, the dilute limit behaviour changes and matrix *confinement* by the RIF starts to play a role. Confinement simply means that the strains of the matrix and RIFs become similar and eventually equal in magnitude when η increases [24,22]. The confinement also reduces the strain enhancement at RIF ends. The relevance of matrix confinement at finite RIF concentrations has been demonstrated for fibre reinforced composites. Early work had already proved that the mechanical properties of composites are not linearly related to the RIF content [25]. Figure 6 shows an example of local stresses in a 2D uniaxial, random fibre composite. The matrix stresses vary spatially in a complicated manner because of interactions between fibres at random locations. Similar random fluctuations would also occur in paper and any random fiber network. Computer simulations have shown that random fibre networks have very broad stress distributions [13,12].

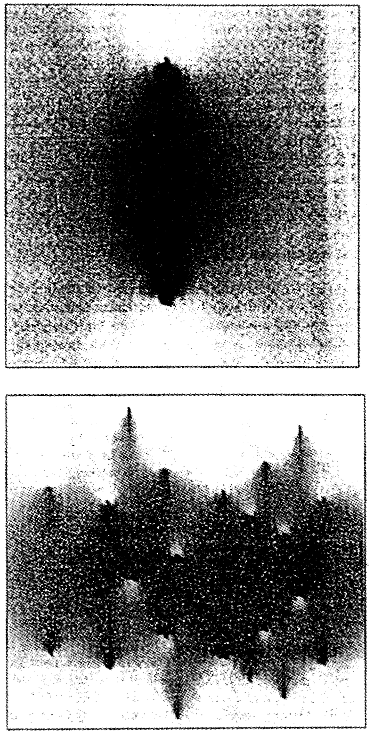

FIG. 6. Strain field plots for a uniaxial 2D fibre reinforced composite from the computer simulation by Monette et al. (J. Appl. Phys. **75**, 1155 (1994)). Dilute case, η = 0.005 (top) and semi-dilute case, η = 0.055 (bottom). White corresponds to high stress and black to low stress.

The percolation effects discussed in Chapter IIA might play a role in matrix confinement. There should be a cross-over concentration above which the strains in the matrix and RIFs are indistinguishable. Above that point even the strength properties of paper should follow the smooth behaviour given by $RBA(\eta)$, as illustrated in Fig. 5(b) for the elastic breaking strain. The non-linear behaviour arises from the competition between the elastic strain reduction in the dilute limit and the matrix confinement.

From the discussions above it is clear that the eventual cross-over concentration η_x depends on the RIF length, the nature of the RIF-to-matrix coupling and on the mechanical compatibility in the dilute limit. The simplified analysis of Chapter IIA suggests that at η_x every RIF would on the average bond to ten other RIFs. Hence the mesh formed by the RIFs is already quite tight. If the RIFs are ductile they should then be able to prevent microcracks in the matrix from growing into macroscopic failure [24]. The phenomenon is called crack arrest.

In Fig. 6 sharp peaks are seen in the matrix stress even when there is an adjagent confining fibre. One can guess that it would be difficult to model or predict the strength properties of fibre composites and fibre mixtures. Direct numerical simulations have been applied to that problem by Duxbury et al. [8]. Figure 7 shows the analoque of tensile strength for different values of a stiffness ratio analogous to S and a corresponding ratio for the elastic breaking strains of the two components in pure form. The composite strength drops from the one-component value immeadiately when the other component is added and varies nonmonotonically for all mixing ratios. The tensile strength behaviour is determined by the elastic breaking strains of the components. It is smooth if the elastic breaking strains are equal and a kink occurs if they are different. The kink resembles a percolation-induced cross-over point. For two-dimensional model systes it occurs at the 40-60 mixing ratio, whereas the percolation threshold is at the 50-50 ratio. For 3D systems the corresponding 'cross-over' takes place at a much lower RIF fraction. This might correspond to paper in the case when the sheet has a low ZD shear stiffness (cf. Chapter IIA). A s a final note such simulations show, that if one defines "toughness" as the energy consumption prior to tensile failure/maximum stress the components should have both differing breaking strains and stiffnesses. The right combination depends unfortunately on the volume fraction and other details.

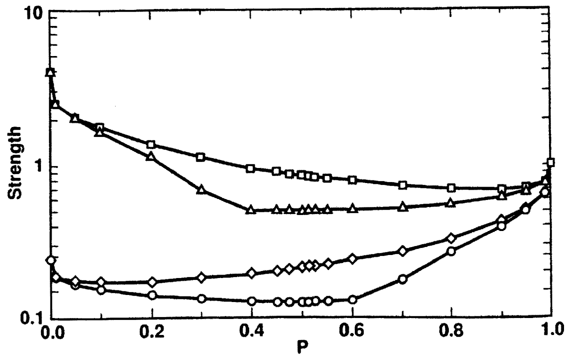

FIG. 7. The strength of a binary mixture against the mixing ratio P. Adopted from the square lattice simulations of Duxbury et al. (Phys. Rev. **B51**, 3476 (1995)). The stiffness ratio S and the tensile strength ratio T of the two components are S = T = 4 (squares); S=1/4, T= 4 (triangles); S=T=1/4 (diamonds) and S=4, T=1/4 (triangles);

In summary, mechanical compatibility has a complicated effect on the mechanical properties of paper against the RIF content η. The dilute limit behaviour can be qualitatively understood from the local strain enhancement at RIF ends. The cross-over to the RBA-controlled behaviour occurs when the RIFs are sufficiently well connected. The geometric factors that control the cross-over content η_x were discussed in Chapter II. One expects naturally that non-linearities in the mechanical properties are smallest when η_x and the reduction in elastic breaking strain $\Delta\epsilon_{el}$ are small.

IV. FRACTURE TOUGHNESS

A. Microscopic mechanisms

One of the main motivations for using reinforcement pulp in paper is to improve the mechanical runnability or fracture toughness of paper. It is believed that the fracture toughness of paper can be characterized by the rupture energy per unit length of the growing crack. On the other hand, in tough materials the energy consumption takes place in a large area around the actual crack tip called the fracture process zone (FPZ). Paper is often so tough that the FPZ is large compared to segment length and standard fracture mechanics cannot be applied [26]. The spatial distribution of the energy consumption can be seen in e.g. the IR images of Yamauchi [27]. However, one can still study the micromechanical processes in the FPZ. In the following we shall briefly discuss the microscopic mechanisms that may affect the fracture toughness of paper. In the absence of experimental material it is practically impossible to evaluate their relative importance in practice.

The effect of a RIF addition on paper toughness can be divided in two parts: the change in the energy spent on matrix failure, and the additional energy spent on RIF debonding and failure. The positive effect of RIFs on fracture toughness arises from their ability to hold together one way or another the network and prevent crack propagation that would otherwise take place. Unless matrix delamination is relevant, the mass fraction η of RIF is in practice always so high that the crack path can not avoid crossing RIFs (cf. Chapter IIA). Then the matrix confinement by the RIFs usually reduces the stress concentration at the crack tip and thereby makes paper tougher.

In the dilute limit the positive confinement effects of RIFs may be partly offset by the premature matrix failure discussed in Chapter IIIA. Tensile strength is reduced but toughness may still increase. This is because the premature microcracks ahead of the actual crack tip may increase the true crack length by crack deflections [28]. Similarly, poor formation may lower tensile strength but increase toughness, though no experimental verification of this is available. In general terms the best toughness with a microcracking material is achieved if the microcracks in front of the crack tip do not coalescence easily. In the case of reinforced paper this can be achieved if the RIFs are ductile.

Even in the absence of RIF there is a clear difference in the shape of the crack path in MD and CD (Fig. 8), reflecting the different nature of the fracture process in the two in-plane directions of machine-made paper. In MD crack deflections increase the true crack length significantly. At least superficially the crack path is suggestive of crack deflections generated by stiff fibres in MD. Addition of *stiff* RIFs to the furnish may enhance the crack deflections.

In CD the crack path (Fig. 8) is shorter than in MD but on the other hand energy is consumed in ductile yielding. Hence the MD/CD ratio of paper toughness is usually much closer to unity than is the MD/CD ratio of elastic modulus or tensile strength. The ductile yielding of paper improves toughness by "blunting" the shape of stress field ahead the crack tip. A blunt stress field spreads the damage to a wide area and energy consumption is high. In composites a weak matrix-fibre interface is also known to lead to crack blunting, as damage develops ahead the crack [29].

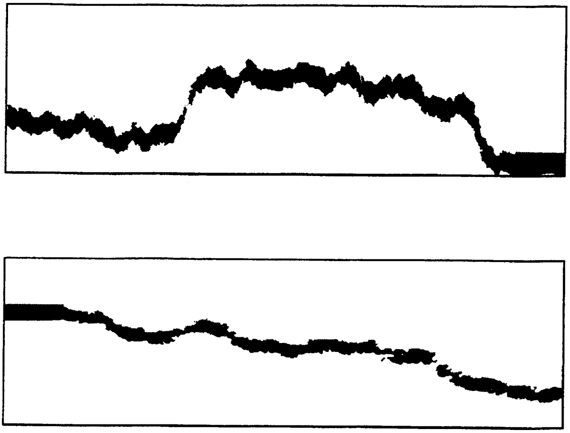

FIG. 8. Typical crack path in an edge-cut newsprint specimen when loaded in MD (top) and CD (bottom).

If *ductile* RIFs (i.e. fibres with high breaking strain) are added to mechanical furnish, the stress field at crack tips may remain unchanged. Instead the RIFs can increase paper toughness by bridging [30,31] slightly opened cracks in the brittle mechanical fibre matrix. Bridging is quite common in paper cracks (Fig. 9). Marshall and Cox [30] have analyzed in detail various scenarios of matrix reinforcement with uniaxial fibres. They demonstrate that bridging can make a brittle material ductile. Linear fracture mechanics is then valid only for cracks that are smaller than a critical size [31,32] below which the cracks are 'linearly elastic'. In other words, linear fracture mechanics applies to the onset of crack growth but not to crack propagation. It is conceivable that the same could be true for reinforced paper.

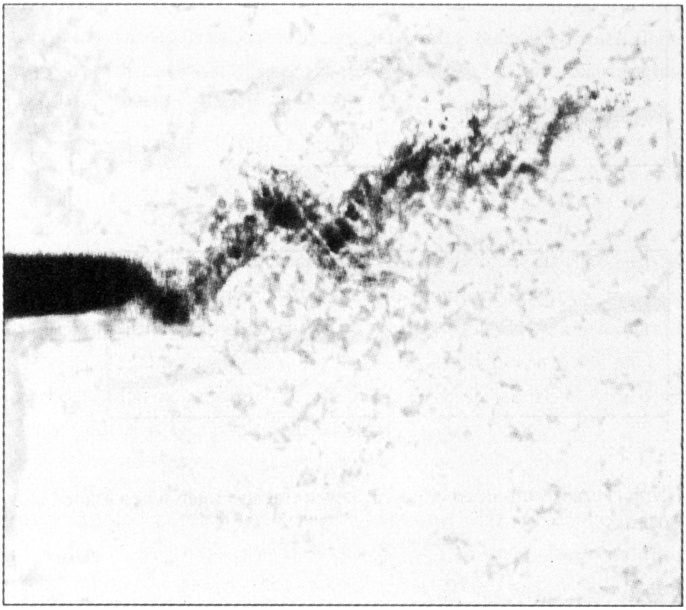

FIG. 9. A propagating crack in a newsprint specimen loaded in MD. The crack tip is on the right and fibres bridging the crack can be seen behind the tip.

The reinforcement action of stiff high modulus fibres can now be compared with ductile, high stretch fibres. High stiffness may be beneficial at high RIF contents where matrix confinement dominates. However, at low RIF contents the stress of stiff fibres increases rapidly as the crack begins to open. Either the matrix break prematurely at the RIF ends or the RIFs debond from the matrix. This does not happen if the RIFs yield plastically. Then the RIFs can bridge cracks in the brittle mechanical fibre matrix and thereby increase paper toughness. In the square lattice simulations of Duxbury et al. [8] the highest toughness for the binary mixture was obtained when the two components had different stiffness and different strength.

The bonding strength of fibres also affects paper toughness. It is a well-known phenomenon in fibre reinforced composites as well as in paper that there is an optimal bonding strength that maximizes the fracture work per RIF [33,34]. Too strong a bond leads to matrix rupture or RIF failure and too weak a bond leads to low energy consumption in fibre pull-out. However, care must be excercised in how the bond strength is varied. Increasing the bond stiffness should generally increase the probability of fibre failure. If, instead the plasticity of the bond is increased, fibre failure need not become more frequent even if bond strength as such increases.

The orientation of RIFs can also have an effect on the debonding and toughness contribution. Fibres at off-axis angles seem to be most effective in increasing toughness. In fibre reinforced concrete the optimum angle has been found to be 30 to 40 degrees depending on the properties of the matrix and RIFs. For long or weak fibres the optimum angle is smaller [35]. The optimum angle arises because the energy consumption in RIF debonding and rupture is different in bending, shearing and axial elongation [36–38,35].

B. Quantitative analysis

The toughness contribution of a RIF addition can be estimated from a simplified model [39] where the fracture energy per fibre is a sum of bonding energy, including the plastic yielding per bond, and fibre failure energy. It is assumed that the RIF pull-out is a gradual process where bonds open one after another. The process need not proceed systematically along the fibre. The model is similar to Friedrich's analysis of the toughness of brittle, fibre reinforced composites [40] and it can be also compared to studies of weak fibre pull-out in composites [41,38]. The difference between the present model and traditional shear-lag models [42,43] is that the latter assume all the bonds to be loaded simultaneously. Then fibres break if the cumulative bond contribution exceeds fibre strength. The two approaches are compared with experiments on paper elsewhere in these proceedings [44]. If fibre breaks are included in composites, gradual debonding takes place and the shear-lag theories seem to be wrong [45,21].

Since the energy consumption in crack propagation is an averaging property, the fracture energy per unit length against the RIF mass fraction η can be written as

$$W = N_m W_m + N_f W_{RIF} \tag{9}$$

where W_m and W_{RIF} are the fracture energy per fibre of the pure matrix and the RIF embedded in the matrix; and N_f and N_m the average numbers of the two types of fibres per unit length of the crack path. At constant basis weight m of an isotropic sheet we have $N_f = \eta m/2\pi C_f$ and $N_m = (1-\eta)m/2\pi C_m$ where C_m and C_f are the coarsenessses of the matrix and RIFs, respectively. Crack deflections and microcracking away from the crack (crack blunting) are ignored.

The model gives

$$W_{RIF} = N_{fail} w_{fail} + L_{pull} w_l. \tag{10}$$

where N_{fail} is the fraction of RIF that fail, w_f the energy needed to break a fibre, and w_l is the energy needed to pull out a unit length of RIF and the average pull-out length is

$$L_{pull} = -\frac{N_{fail} l_f/4}{\log(1 - N_{fail})}. \tag{11}$$

Thus we finally obtain

$$2\pi W/m = (1-\eta) W_{ave,m}/C_m + \eta (N_{fail} w_{fail} + L_{pull} w_l)/C_f. \tag{12}$$

A few remarks are in order. The fracture energy W_{RIF} has a non-trivial dependence on the fraction of broken fibres, N_{fail}. The precise dependence of N_{fail} on fibre and bond properties is unknown. The pull-out energy per unit fibre length, w_l, arises from the opening of inter-fibre bonds and from the plastic elongation of bonded fibre segments. The latter component, the energy of plastic yielding of the fibres, is believed to be much larger than the pure bonding energy [46,47]. Also, the matrix fracture energy, $W_{ave,m}$, may depend on the RIF addition through the stress enhancement and confinement effects discussed in Chapter IIIB.

Given the uncertainty related to N_{fail} we consider only the $N_{fail} \to 0$. The opposite case, $N_{fail} \to 1$, is not an interesting case for a RIF. In the first case the fracture toughness of paper becomes

$$2\pi W/m = (1-\eta)W_{ave,m}/C_m + \eta w_l l_f/4C_f. \tag{13}$$

The energy consumed in opening the bonds, w_l, should be linearly proportional to RBA and should also increase with the drying shrinkage of paper. Well-bonded long RIFs improve paper toughness. If the RIFs are ductile, have a high breaking strain, their failure probability in the opening crack is low and also w_l is high. Again the ductility of the RIF and the RIF-to-matrix bonds improves paper toughness. The increased ductility may be obtained by increasing the plastic strain component of the fibres and bonds.

In contrast, increasing the strength of the RIFs through either the stiffness or elastic breaking strain of the fibres is not beneficial even though it might reduce N_{fail} to zero. At the same time the probability of premature matrix failure increases and may dramatically decrease the energy consumed in the matrix failure, $W_{ave,m}$. The situation is of course different if almost all of the RIF across the crack path fail, but such a furnish will hardly be used as a reinforcement pulp in the first place.

V. CONCLUSIONS

We have analyzed the factors affecting the performance of reinforcement fibres in paper. The criteria for a good RIF depend on the paper property that the end-user is most interested in and, to some extent, on the RIF content that one is using.

The effect of RIFs on the mechanical properties of paper depends on their geometric connectivity, mechanical compatibility with the matrix and ability to resist crack propagation. To improve the tensile strength and toughness of paper, the reinforcement fibres should be chosen to match the stiffness of the mechanical fibres. In this way the risk of premature matrix failure at low RIF contents is minimized. At high RIF contents the risk of triggered matrix failure is lower and particularly the tensile strength of paper can be improved by adding stiff and strong reinforcement fibres. At all RIF contents the elastic modulus benefits most from stiff reinforcement fibres.

Long, ductile and low-stiffness fibres with many ductile bonds should enhance the fracture toughness of paper at all concentrations. If on the other hand high strength and modulus are required, well bonded high stiffness fibers are the best. Mixtures of more than one kind of a RIF pulp [48], 'hybrids' in composite language, may also improve paper properties.

The geometric percolation of the RIF network may or may not be of importance. That depends crucially on the bonding properties of the reinforcement fibres. The mass fraction of RIF necessary for the 'percolation effects' to appear can be quite high, 30 % or even more.

The effect of distributed RIF properties has been ignored above. One could consider variable fibre length and strength, to name but two. In the first approximation, assuming that all the RIFs bond in a roughly similar manner, variations in fibre length should not play any crucial role. On the other hand a wide distribution of fibre strengths and moduli may reduce or even increase [8] the fracture toughness of paper. However, since the weakest spot of a fibre may be shielded behind bonds that have to opened first, it is also possible that the width of the fibre strength distribution does not affect toughness.

Finally, the mechanical compatibility of the reinforcement pulp with a given mechanical pulp can be evaluated from the mechanical properties of paper measured at small RIF contents η. The asymptotic dependence of strength, elastic modulus and fracture toughness on $\eta \to 0$ gives information on both the fibre compatibility and the RIF-to-matrix bonding.

Acknowledgements

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Transcription of Discussion

Performance of Reinforcement Fibres in Paper

Mikko J Alava, Helsinki University of Technology, Finland

Petri Kärenlampi, Champion International Corp, USA

Your last two slides showed the effect of fibre strength and bond strength on fracture energy. Did you really have strength or failure load or did you actually have failure energy of these constituent fibres and bonds in these figures?

Mikko Alava

The figures were based on a simple argument which we had in our paper which basically related to the fibre failure probability to the strength of bonds and the fibres. This exercise on calculating fracture toughness is based on simple arguments and it has one largish hole in it in that it uses the quantity which is the probability of a fibre to fail in crack propagation which is rather hard to calculate from first principles. The figures shown include an argument of how that probability should actually depend on the strengths of the bonds and fibres, respectively.

Petri Kärenlampi

So, have we solved the problem between the failure energies and the fibre failure probability, the answer is No - we know hardly anything about that.

Mikko Alava

That statement is accurate to my mind.

Dr John Parker, Messmer Büchel, UK

It has been assumed during this discussion that the structure of paper is random, whereas the presence of flocs suggests that it is not. I have made observations which indicate that if the stock is moving relative to the wire of a Fourdrinier the flocs tend to rotate. In doing so they would be expected to gather up longer reinforcing fibres into their surfaces. This would decrease the number of such fibres between the flocs, simultaneously decreasing the strength of the web.