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Fiber Properties and Paper Fracture - Fiber Length and Fiber Strength

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ABSTRACT

Phenomenological theories on the effect of pulp fiber properties on the fracture energy of paper are discussed. The effect of fiber length and strength is clarified experimentally. Fiber length appears to affect fiber failure probability only slightly. When fiber strength is changed, the fracture energy decreases greatly with only a small increment in fiber failure probability. This suggests that the fracture energy contribution of a fiber may be correlated between fibers. The effect of fiber length and strength on the cohesive stress - crack widening relationship is clarified.

INTRODUCTION

While considerable financial resources are consumed in adding softwood reinforcement fibers to paper webs, optical and printability properties are simultaneously impaired. Some phenomenological theories have been propounded in order to gain some knowledge of how the reinforcement pulp works. In the first type of theories [1-4], the fracture resistance of paper is assumed to arise from frictional forces when pulling fibers out of the surrounding medium. Alternatively, fiber debonding and fiber failure may be modeled as stochastic, energy-consuming processes [5, 6].

None of these theories has been properly tested experimentally. Further, the probabilistic approach to debonding and fiber failure suffers from serious difficulties in coupling fiber failure work, bond rupture work and fiber failure probability. Particularly, the thermodynamic "rate" equation recently used to illustrate this coupling [6] amounts to the probability of fiber failure in the context of bond failure decreasing with increasing fiber length, which is very unlikely to reflect real paper behavior. In this paper we intend to modify the probabilistic approach, specifically what comes to the effect of fiber and bond properties on the fiber failure probability. We then employ the two types of theory to formulate hypotheses on the effect of fiber length and fiber strength on the proportion of breaking fibers and the specific fracture energy of paper. We concentrate on these fiber properties since they can be changed and measured in experiments, and we discuss the fiber failure probability since the proportion of broken fibers can be observed in fractured sheets. We report experiments by which some of these hypotheses are tested. Finally, we discuss the effect of fiber properties on stress - crack widening relationship, which is likely to control the fracture of real paper webs.

MODIFIED PROBABILISTIC THEORY

The recent probabilistic approach [5, 6] postulates that the average work needed to release one fiber from resisting crack propagation is

$$W_{Fiber} = \left[W_f + \frac{(1 - p_f)}{p_f} W_b \right] \left[1 - (1 - p_f)^n \right] \tag{1}$$

where W_f is the work needed to break a fiber, W_b the work needed to break a bond, p_f the probability of fiber failure in the context of breaking a bond, and n the average number of bonds which fail where the fiber does not break. Assuming that bonds break on average form a quarter of fiber length, we get $n = \frac{l_f}{4\, l_s}$ where l_f is fiber length and l_s is the length of a fiber segment between bond centroids. Naturally $1-(1-p_f)^n$ equals the fiber failure probability, denoted as N_f.

Unfortunately this theory does not provide any information on the interdependence of W_f, W_b, p_f, l_s, and l_f, and thus it is very difficult to formulate hypotheses which could be verified experimentally. Different kinds of arbitrary assumptions about this interaction have been applied in previous studies [5, 6], resulting in more or less unrealistic predictions.

Let us now consider the seminal finding of Cox [7, cf. 8, 9], that the second differential of fiber load P_f with respect to the distance from fiber end x is

$$\frac{P_f^2 x}{dx^2} = 8\ G_f\, RBA^2\ t / w \left(\frac{P_f}{E_f\ t\ w} - \varepsilon \right) \tag{2}$$

where G_f is the shear and E_f the Young's modulus of the fiber, t the thickness and w the width of the fiber, RBA is the relative bonded area and ε is the macroscopic sheet strain in the direction of the fiber. In the circumstances of a fiber bridging the opening crack faces, it is reasonable to assume that the sheet strain ε is small in comparison to the fiber strain. Integrating Eq. (2) and considering the condition for bond failure as the differential of fiber load over the bond exceeding the failure load of the bond, the critical fiber load for bond failure becomes

$$P_{bf}(x) = F_b \sqrt{\frac{E_f}{2G_f}} \tanh\left(\sqrt{\frac{2G_f}{E_f}} \frac{2\ RBA}{w} x \right) \tag{3}$$

where F_b is the (shear) failure load of a single bond. Fibers being much longer than their width, the value of the $\tan$ -expression is close to unity and thus the critical fiber load is not sensitive to the distance from the fiber end. The Young's modulus of a fiber being roughly 30 times the shear modulus [cf. 10-15] we find that the fiber load needed to break a bond is roughly 4 times the failure load of the bond, which agrees with experiments where single fibers have been extracted from a handsheet [16]. Thus the transition from bond failure to fiber failure takes place around

$$F_f \approx 4 F_b \tag{4}$$

where F_f is the fiber tensile failure load.

Provided that the fiber and bond failure work W_f and W_b are proportional to the square of fiber and bond failure load F_f and F_b respectively, the transition from bond failure to fiber failure should occur around

$$\sqrt{W_f} \approx k \sqrt{W_b} \tag{5}$$

where k is a constant or presumably only a weak function of bond and fiber properties. Classical fracture mechanics suggests that the fracture work of a brittle object is proportional to the square of its failure load as long as Young's modulus is constant. The definition of Eq. (5) is somewhat arbitrary, amounting to the relationship between bond failure load and bond failure work. However, the thermodynamic "rate" equation discussed above [6] giving biased predictions, this is so far the most reasonable way we have found to couple bond and fiber failure work to fiber failure probability.

Now we should formulate a continuous function for fiber failure probability in the context of bond failure p_f as a function of the quantities given in Eq. (5); we assume that p_f does not depend on the original length of fiber segments between bond centroids since the length of the highly stressed fiber portion bridging the crack faces is not determined by segment length. Unfortunately we do not know what kind of function of the quantities of Eq. (5) p_f is. All we can do is introduce a simple statistical distribution function which has seemed useful for a wide range of distributions [17, 18]:

$$p_f = 1 - e^{-\left(\frac{k\sqrt{W_b}}{\sqrt{W_f}}\right)^m} \tag{6}$$

Eq. (6) defines the coefficient k as the square root of the particular ratio of W_f and W_b at which p_f equals 0.63. The Weibull exponent m reflects the variability of fiber and bond properties: with a large m Eq. (6) is a step function, while a smaller m reflects greater variability in fiber and bond properties. Quite a few simplifications and assumptions have been made in this treatment. The advantage of Eq. (6) in comparison to previous approaches [cf. 5, 6] is that now we are using the argument derived above for the criterion defining the transition between bond failure and fiber failure hoping to avoid the unphysical consequences which were found previously.

HYPOTHESES DERIVED FROM THE THEORIES

The Effect of Fiber Length

Eq. (6) implies that the fiber failure probability when breaking a bond p_f is independent of fiber length. In such circumstances, fiber length affects the estimate of fracture energy given by Eq. (1) only through the fiber failure probability N_f being affected by the number of bonds. Both of these quantities increase monotonically with fiber length, but their second fiber length differential is below zero (Figure 1a). The fracture energy being estimated as the fiber failure probability multiplied by a form which is independent of fiber length (Eq. (1)), the specific fracture energy R should be linearly proportional to the fiber failure probability N_f (Figure 1b; please note that this prediction applies only when N_f is affected by changing fiber length). These results differ slightly from the predictions of earlier probabilistic models [5, 6].

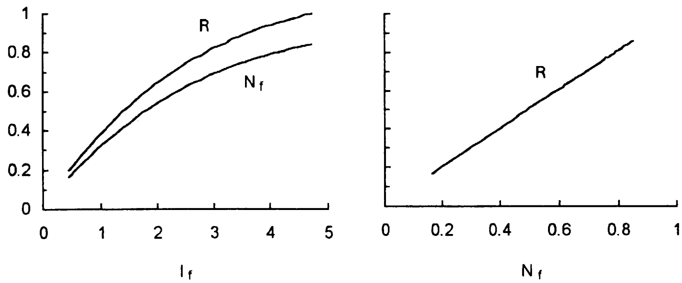

Figure 1: The effect of fiber length on the proportion of breaking fibers and the fracture energy of paper according to the modified probabilistic theory.

Earlier theories considering fiber pull-out friction as the main mechanism of paper fracture energy consumption [1-4] yield quite different predictions. Shallhorn [3, 4] proposes that a fiber fails rather than being pulled out if the total bond shear strength within the shorter fiber end embedded in the matrix exceeds fiber failure load. This results in a critical fiber length where the probability of fiber failure first differs from zero:

$$l_c = \frac{2\,F_f}{\tau_l} \tag{7}$$

where τ_l is total bond shear strength per fiber length unit. This results in a fiber failure probability of

$$N_f = 1 - \frac{l_c}{l_f} \tag{8}$$

provided that $l_f \geq l_c$; otherwise $N_f = 0$.

We further find that the fracture energy equations derived in references [3, 4] become

$$R = \frac{F_f^2}{6\tau_l}\left(\frac{l_f}{l_c}\right)^2 \tag{9}$$

if $l_f \leq l_c$, and

$$R = \frac{F_f^2}{6\tau_l}\frac{l_c}{l_f} \tag{10}$$

if $l_f \geq l_c$.

Shallhorn's theory has been derived assuming that all fibers are uniform, which is quite restrictive in a deterministic theory. However, we can easily illustrate the effect of scattered fiber properties by numerically introducing a distribution of fiber length. This has been done in Figure 2, where the fiber length l_f has been given a coefficient of variation of 0.15, other quantities remaining constant.

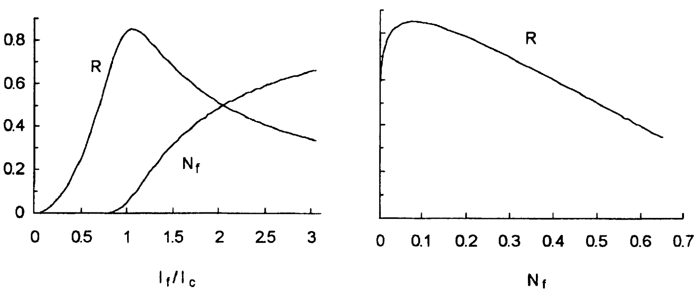

Figure 2: The effect of mean fiber length on the proportion of breaking fibers and the fracture energy of paper according to the deterministic pull-out theory.

The difference between the consequences of the probabilistic theory (Figure 1) and the deterministic pull-out theory (Figure 2) are striking. In Figure 1 we find a monotonic increase in fracture energy as a function of fiber length, whereas in Figure 2 the fracture energy is a nonmonotonic function of fiber length. When the proportion of failing fibers is increased by increasing fiber length, Figure 1 predicts increasing fracture energy while Figure 2 predicts a decrease.

The Effect of Fiber Strength

We find from Eq. (5) that the transition from bond failure to fiber failure when breaking a single bond is defined by the ratio $\frac{\sqrt{W_f}}{k\sqrt{W_b}}$ approaching unity. This dimensionless ratio being linearly proportional to fiber failure load, it can be used to show the effect of fiber strength on the probability of fiber failure and fracture energy. Having a small Weibull's exponent m which reflects large variability in fiber and bond properties, p_f and N_f decrease smoothly as a function of $\frac{\sqrt{W_f}}{k\sqrt{W_b}}$ (Figure 3). The fracture energy appears to increase monotonically with fiber strength and thus decrease monotonically as a function of the proportion of failing fibers N_f.

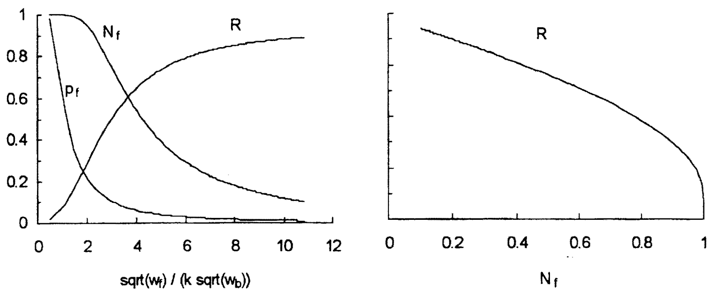

Figure 3: The effect of fiber strength on the proportion of breaking fibers and the fracture energy of paper according to the modified probabilistic theory. $k = m = 2$.

Less variability in terms of a larger exponent m makes the model to predict that the fracture energy is a non-monotonic function of fiber strength (Figure 4). A larger value for k makes this effect more pronounced, the energy consumed in fiber failure being considerable compared with the energy needed to break bonds. Thus, when some of the fibers fail, the total energy consumed may be greater than where bonds only fail. Such a complementarity of fiber and bond failure work has been previously argued to be expectable: one of these quantities becoming much greater than the other, the fracture energy should decrease [6]. However, we see in Figure 3 that this effect may not be found if the variability in fiber and bond properties is large. The real-life situation should be clarified by a physical experiment.

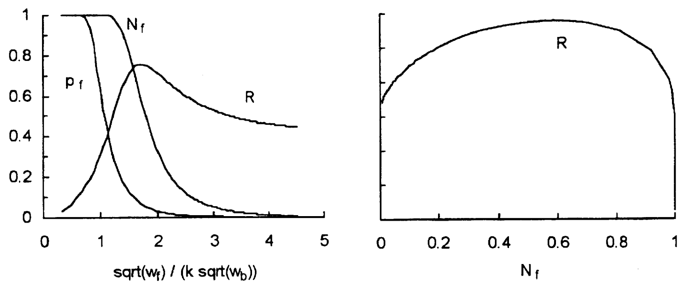

Figure 4: The effect of fiber strength on the proportion of breaking fibers and the fracture energy of paper according to the modified probabilistic theory. $k = 3$, $m = 5$.

Shallhorn's approach [3, 4] can be readily modified to address the effect of fiber strength. The critical fiber failure load where the probability of fiber failure first differs from zero is

$$F_c = \frac{l_f \tau_l}{2} \tag{11}$$

The fiber failure probability is

$$N_f = 1 - \frac{F_f}{F_c} \tag{12}$$

provided that $F_f \leq F_c$, otherwise $N_f = 0$.

The fracture energy equations derived in references [3, 4] become

$$R = \frac{\tau_l \, l_f^2}{24} \tag{13}$$

if $F_f \geq F_c$, and

$$R = \frac{\tau_l \, l_f^2}{24} \left(\frac{F_f}{F_c} \right)^3 \tag{14}$$

if $F_f \leq F_c$.

Let us then introduce a distribution of fiber failure load with a coefficient of variation of 0.15, other quantities remaining as constants, and consider the proportion of breaking fibers and fracture energy as a function of fiber failure load in Figure 5.

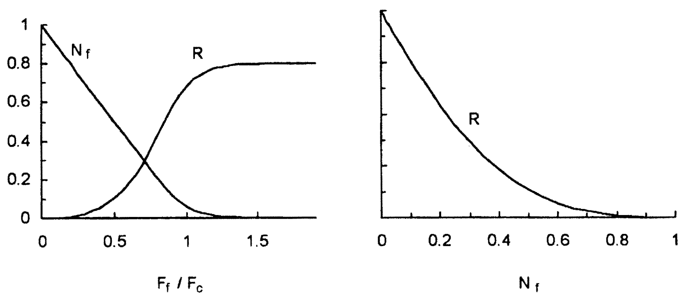

Figure 5: The effect of mean fiber failure load on the proportion of breaking fibers and the fracture energy of paper according to the deterministic pull-out theory.

Comparing Figures 3 and 5 we find the predictions of the two theories quite similar as to the effect of fiber strength, particularly with small values of m (Figure 3). As long as m is small, the only difference which may be distinguished in experiments is the much stronger decrement of fracture energy as a function of the proportion of breaking fibers in Figure 5.

EXPERIMENTS

Laboratory handsheets from bleached softwood kraft pulp, length-weighted mean value of fiber length 2.17 mm and coarseness 0.126 mg/m, were prepared. Wet handsheets from beaten fibers were guillotined, reslushed and then fractionated in order to achieve a variation in fiber length. On the other hand, handsheets from uncut, unfractionated, beaten fibers were exposed to HCl vapor in a closed glass container in order to hydrolyze cellulose and reduce fiber strength [cf. 19-22].

The mechanical properties of the handsheets were examined by the short-span tensile test [23-25]. With a testing span of 8 mm the elastic energy stored in the specimen was small enough to achieve stable fracture. Strips of 15 mm wide were elongated at a rate of 10%/min. Load-displacement curves were recorded, and the fracture energy was determined as a sum of the elastic energy stored at the moment of greatest load and the additional external work needed to tear the specimen. Tensile stiffness at the moment of maximum load was assumed to be equal to initial stiffness [cf. 26-33]. Confidence limits of 95% for measured quantities varied between +-3% and +-7%.

Two percent of the fibers were dyed with Congo Red prior to sheetmaking. In each sample, 200-400 dyed fibers crossing the failure path were inspected for fiber failure under a microscope. Tensile loading was terminated at 95% load decrement from the maximum load, which allowed relatively convenient detection of the failure path, there being a clearly observable displacement, but some cohesion was still holding the crack faces together.

Fiber Length

It is difficult to change the distribution of only one property within a population of fibers and leave others unaffected. We find from Table 1 that in addition to a very considerable variation in the length-weighted mean fiber length, there is a small variation in coarseness. The variation in the apparent density of the handsheets implies that there is a small variation in fiber flexibility as well. However, these variations being small in comparison to the variation in fiber length, it is likely that the effect of fiber length can be deduced from this experiment.

Mean Fiber Length, mm	Mean Fiber Coarseness, mg/m	Apparent density, kg/m^3	Tensile Stiffness Index, kNm/g	Tensile Strength Index, Nm/g	Rupture Strain, %	Total Work to Fracture, Jm/kg	Work to Maximum Load, Jm/kg	Fracture Energy, Jm/kg	Breaking Fibers, %
0.8	0.127	560	4.5	39	3.4	13	8	6	2
1.4	0.125	560	5.4	54	4.2	21	14	9	6
2.2	0.125	520	5.5	63	4.4	26	16	13	8
3.1	0.138	500	5.0	60	4.1	30	14	19	8

Table 1: Properties of fractionated fiber populations and handsheets made from them.

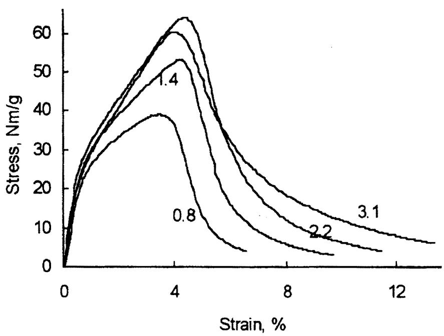

Figure 6: Stress-strain curves of handsheets of different fiber length.

We see in Table 1 and Figure 6 that the tensile stiffness is not sensitive to fiber length. This agrees with the seminal theoretical approaches in that as long as the fibers are long in comparison to their width, stress transfer efficiency between fibers does not depend drastically on fiber length [7, 8]. Micromechanical models have not been successful in explaining the effect of fiber properties on the tensile strength and rupture strain of paper [34]. We see in Table 1 and Figure 6 that the rupture strain is a weak function of fiber length, and tensile strength not much affected either unless the fiber length is very low. This agrees with previous observations with softwood kraft pulp handsheets [20, 35].

The tensile energy absorption being the integral of load over displacement, the weak effect of fiber length naturally applies to the energy absorption up to maximum load as well (Table 1, Figure 6). Instead, the effect of fiber length on the fracture energy is very considerable (Table 1). This is further illustrated in Figure 7, where we find that the fracture energy appears to be linearly proportional to the fiber length.

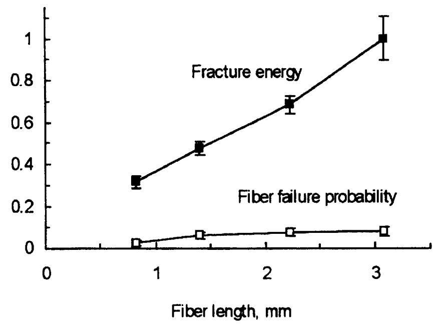

Figure 7: The effect of fiber length on fracture energy and fiber failure probability. The fracture energy has been normalized by 19 Jm/kg which was the greatest value achieved in this experiment.

From the point of view of testing the hypotheses given in Figures 1 and 2 one might consider it unfortunate that the proportion of failing fibers in the experiment was quite low and hardly affected by fiber length (except in the case of very short fiber length). However, the small effect of fiber length on fiber failure probability is in agreement with Eq. (4) and in striking conflict with Eq. (7).

Another issue supporting the probabilistic bond failure theory rather than the deterministic theory based on fiber pull-out friction is that as long as the fiber failure probability is small, Eq. (1) predicts the fracture energy will be linearly proportional to fiber length, which is seen in Figure 7. On the other hand, the pull-out theory predicts a quadratic fiber length dependency, of which no signs are apparent.

Fiber Strength

The vaporization experiment decreased the zero-span tensile index effectively, and retained sheet density as well as tensile stiffness except in the case of most intensive vapor treatment where the fibers became very brittle (Table 2). All mechanical properties except tensile stiffness (and possibly yield strain) seem to be strongly affected by fiber strength, both strength and rupture strain decreasing considerably with increased vaporization (Figure 8). Both pre-failure tensile energy absorption and fracture energy depend strongly on fiber strength.

Zero-span Tensile index, Nm/g	Apparent Density, kg/m^3	Tensile Stiffness Index, kNm/g	Tensile Strength Index, Nm/g	Rupture Strain, %	Total Work to Fracture, Jm/kg	Work to Maximum Load, Jm/kg	Fracture Energy, Jm/kg	Breaking Fibers, %
33	520	4.0	20	0.6	1	0.8	0.5	100
56	510	5.0	36	2.2	7	5	3	58
78	510	5.0	50	3.9	17	12	7	20
114	520	5.0	70	5.8	38	23	19	8

Table 2: Properties of vaporized handsheets.

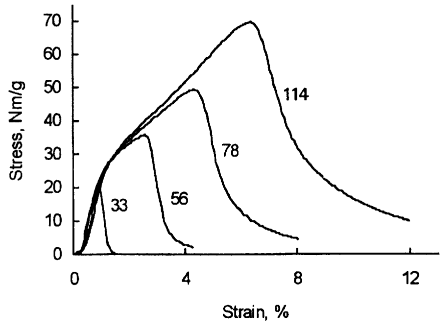

Figure 8: Stress-strain curves of vaporized handsheets.

The fiber strength experiment resulted in a very considerable variation in the proportion of breaking fibers. Both the fracture energy and the proportion of breaking fibers appear to be somewhat nonlinear functions of the zero-span tensile index as was indicated in Figures. 3 and 5 (Figure 9)). No non-monotonic fiber strength effect on fracture energy, as was hypothesized recently [6] and illustrated in Figure 4, is apparent.

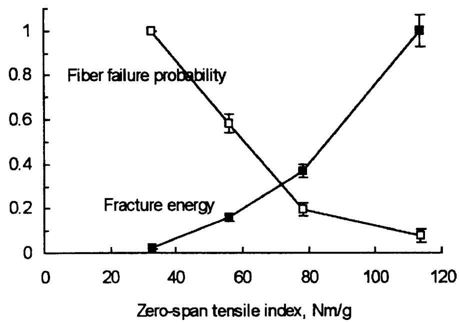

Figure 9: The effect of fiber strength on fracture energy and fiber failure probability. The fracture energy has been normalized by 19 Jm/kg which was the greatest value achieved in this experiment.

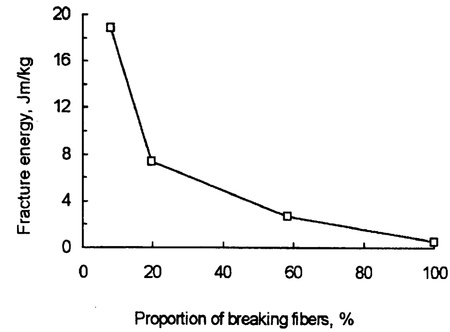

Figure 10: The relationship between fracture energy and fiber failure probability with changed fiber strength.

An interesting and significant observation is that the fracture energy as a function of fiber failure probability seems to display a concave rather than convex shape (Figure 10). This agrees with Figure 5 which is calculated from the deterministic fiber pull-out theory, and disagrees with Figures 3 and 4 which have been calculated from the theory of stochastic debonding. However, the initial decrement in fracture resistance in Figure 10 is so steep that it is difficult to explain just by assuming that fibers failing fail without bond failure and that the fiber failure work is negligible (cf. Figure 5). It is possible that even a moderate proportion of fiber failure decreases the width of the fracture process zone and possibly reduces the work consumed in debonding other fibers.

Conclusions

The fiber length experiment (Figure 7) demonstrates that the fiber failure probability is a weak function of fiber length. Thus it is very questionable to assume that a fiber fails rather than gets pulled out if the total bond shear strength within the shorter fiber end embedded in the matrix exceeds fiber failure load. This discourages applications based on Eq. (7) or (11) [3, 4].

On the other hand, the fiber strength experiment (Figure 10) shows a very considerable decrement in the fracture energy with a relatively small increment of fiber failure probability. This can hardly be explained in terms of a theory where the work consumed by releasing any fiber is calculated independently and the contribution of any fiber on the fracture energy is then totalled. It is rather likely that the energy-consuming contributions of fibers are correlated, possibly because of variations in the width of the fracture process zone.

DISCUSSION

The fracture toughness of paper has been discussed above in terms of critical energy release rate reaching the autonomous end region of crack propagation [36-38]. This equals the critical value of the J-integral provided that the integration path is taken around the end region [39-42]. It was assumed that traction-free crack surfaces are opened. In the experiments, the straining was terminated at 95% load decrement (from the maximum) which means that the crack surfaces were nearly traction-free and the remaining work needed to separate the surfaces was a negligible part of the total fracture energy.

However, it is questionable whether the specific essential work of fracture really is critical for the durability of real paper webs. Mode 1 fracture properties of a material are mainly determined by the cohesive stress appearing between the surfaces of an opening crack, and this attraction is a function of the crack widening [43, 44, 25]. The specific essential work of fracture (or the critical value of the J-integral as defined above) equals the integral of the cohesive stress over the crack widening (crack opening displacement).

It has been shown that if the cohesive stress is a decreasing function of the crack opening displacement, the fracture process zone does not develop fully prior to failure, *i.e.* completely traction-free surfaces are not created before instability [45, 43]. This means that even if the specific essential work of fracture is a material property, it is not the factor determining the durability of a real paper web. The material property directly contributing to the fracture behavior of an object is the cohesive stress - crack widening curve as such. Let us now look at the effect of fiber length and fiber strength on the stress - crack widening behavior.

On the assumptions that the crack widening is uniform and that the Young's modulus of the paper outside the fracture process region remains constant, the crack widening can be calculated from the total specimen displacement by subtracting the uniform displacement remote from the fracture process zone. Crack widening then becomes

$$w = \Delta - \Delta_c + \frac{\sigma_c - \sigma}{E} L \tag{15}$$

if $\Delta \geq \Delta_c$, where Δ is total displacement, Δ_c is displacement at maximum stress σ_c, E is Young's modulus and L specimen length.

Let us then plot the stress-widening curves from the fiber length experiment in Figure 11. We find a concave form of curve as has been previously demonstrated for paper samples [25]. The maximum stress equals tensile strength, and the critical crack widening where the cohesive stress approaches zero is obviously determined by the length of the longest fibers. The front end of the curve is more critical for the failure of an object than the rear end [25, 43, 45].

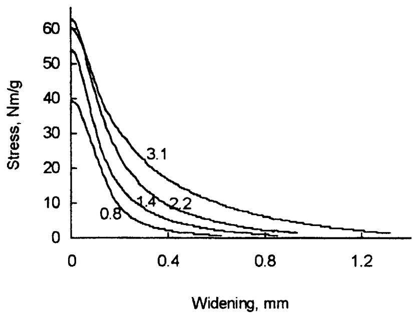

Figure 11: Stress - crack widening curves for softwood kraft pulp handsheets of different fiber length.

We find in the stress-widening curves of the fiber strength experiment that both the maximum stress and the critical widening are strong functions of fiber strength. The reason for the critical widening being strongly affected by fiber strength is that with increasing fiber failure probability the relevant length scale turns from the length of fibers towards the length of fiber segments between bonds.

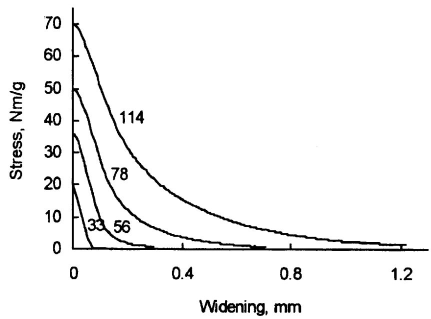

Figure 12: Stress-crack widening curves for softwood kraft pulp handsheets of different fiber strength.

There are strong geometry and size dependencies in the fracture behavior of objects with a decreasing stress-widening curve [43]. Thus it is hardly possible to draw conclusions on the fracture of real paper webs on the basis of material properties directly. The authors are looking forward for numerical treatments [cf. 25, 43, 46].

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Transcription of Discussion

Fibre Properties and Paper Fracture - Fibre Length and Fibre Strength

Yongzhong Yu, Consultant, University of Helsinki, Finland

Professor C T J (Kit) Dodson, UMIST, UK

Can I just clarify whether you have addressed a mixture of two theories - pull out and fracture?

Yongzhong Yu

No I haven't yet.

Professor Jacques Silvy, Universidade de Beira Interior, Portugal

Don't you think that the stress concentration near the crack propagation has something to do with the length of the fibres. I mean that the effect of the length could provide a factor for the stress - only depending on a geometrical effect involved in the stress concentration phenomena?

Yongzhong Yu

Sorry I did not catch the question

Jacques Silvy

What I mean usually in crack propagation there is a geometrical effect of the stress concentration as paper is not homogeneous. This factor could change in respect of length of the fibres that are involved in the propagation of the crack.

Yongzhong Yu

Yes, the effect of the fibre length on paper fracture will be affected by the stress concentration near the crack propagation. I mean in addition to the fibre properties, the geometry of the tested samples and the loading will influence the fracture behaviour of paper as well. However, keeping the geometry and loading conditions unchanged for all samples, then the effect of fibre lengths can be detected. From the experimental results, we found that the fibre length affected fracture process zone, and further affected the fracture energy of paper.

Jacques Silvy

Could you verify the value of the fibre load that you need to assume in respect of your results.

Yongzhong Yu

No I cannot comment.

Steve Eichhorn, Student, UMIST, UK

Do you think that it's possible in the future to find the energy release in the sheet during fracture from measurable parameters?

Yongzhong Yu

I think it is very likely.

Steve Eichhorn

That's very interesting because of our own probabilistic theory of bond failure which requires the energy release in the sheets to prove whether they are actually correct.

Yongzhong Yu

In this paper we use short span tensile test which seems to be a very convenient method for measuring the fracture energy of paper.