

Optimizing reinforcement pulps by fracture toughness

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ABSTRACT: *The major use of bleached softwood kraft pulps is to reinforce weaker papermaking furnishes. Reinforcement is needed not only to enhance the strength of the wet web and the dry sheet but also the strength of the sheet when rewetted, such as when sizing, coating, and printing. In-plane fracture toughness was proposed earlier as a criterion to evaluate the reinforcing potential of softwood pulps. At different refining levels, the fracture toughness of a sheet depends strongly on its tensile strength and extensibility. Fiber properties and processes that enhance these two characteristics also tend to enhance wet-web strength and rewetted sheet strength. This suggests a simple strategy for maximizing the reinforcing potential of bleached softwood pulps in papermaking—refine them to a high tensile strength while maintaining acceptable drainage characteristics; this generally results in high sheet extensibility as well. Ways to obtain high sheet tensile strength are discussed.*

KEYWORDS: *Bibliographies, chemical pulps, fiber bonding, fracture, kraft pulps, mechanical properties, mechanical tests, optimization, properties, pulps, refining, reinforcement, softwood pulps, strength tests, stretch, tear tests, tensile strength, test methods, toughness.*

Canada is the world's largest supplier of paper-grade market pulps. Nearly two-thirds of these pulps are bleached softwood kraft (1). The major use of bleached softwood kraft pulps in papermaking is for reinforcement, i.e., to strengthen

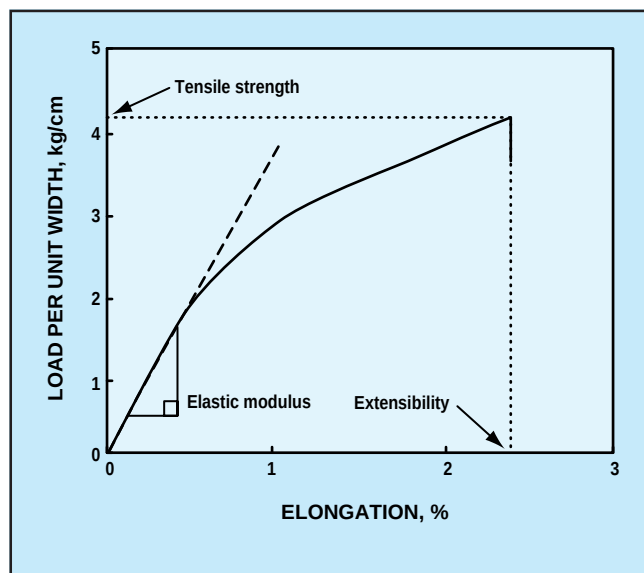
weaker furnishes without significantly impairing their other end use properties. Adding reinforcement fibers to a weak furnish generally improves its drainage and enhances the strength of the wet web (2). These factors control the wet-end

runnability of the paper machine. Reinforcement fibers also enhance the strength of the dry sheet (2); this is required for the runnability of the paper machine and that of the sheet during subsequent converting operations, such as printing. Another important purpose of reinforcement fibers is to enhance the strength of the rewetted sheet. A dry sheet, when wetted, becomes much weaker and is more likely to fail. During papermaking, the sheet is rewetted at the size press. The sheet is also rewetted during coating as well as in certain printing operations. Therefore, a sheet that is to be rewetted requires extra reinforcement.

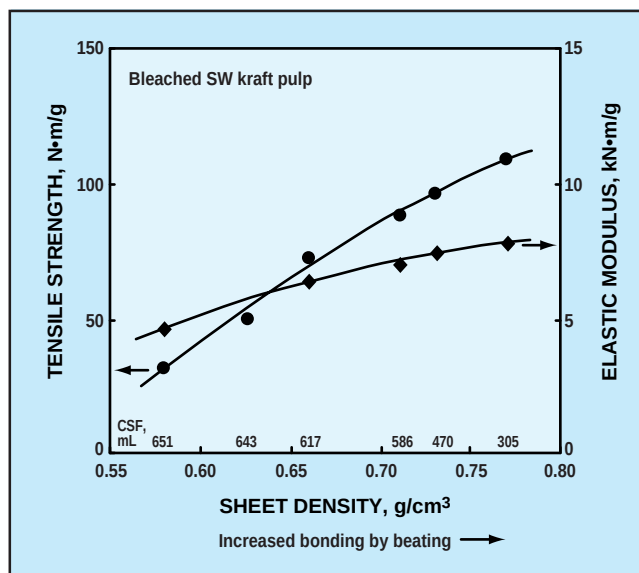
While the term *strength* is used loosely in our industry, the fundamental mechanical properties of a material are its elastic modulus, tensile strength and extensibility, and fracture toughness. **Figure 1** shows the load-elongation curve of a typical paper sheet. The slope of the initial linear part of the curve determines the elastic modulus of the sheet. This property controls sheet stiffness. At a certain load paper yields, and the load-elongation curve deviates from linearity. The endpoint of the curve gives the tensile strength and the extensibility of the sheet. Fracture toughness is the sheet's ability to resist propagation of a pre-existing flaw or crack. It is measured as the work consumed per unit surface area created during crack propagation. Fracture toughness determines the flaw-carrying ability of the sheet.

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1. Load-elongation curve of paper



2. Elastic modulus and tensile strength of a sheet increase when the degree of bonding between fibers is increased. Increased bonding also increases sheet density.



The reinforcing potential of a pulp, or *pulp strength*, is currently assessed by beating or refining the pulp, and then determining the Elmendorf or Brecht-Imset tearing resistance of handsheets at a given tensile strength. Beating or refining increases the degree of bonding between the fibers and, therefore, increases the density, tensile strength, and elastic modulus of the sheet (**Fig. 2**). However, for most softwood pulps, the tearing resistance decreases with bonding at moderate to high degrees of bonding (**Fig. 3**). This is the range where the tensile strength potential of the pulp is best realized. Thus, a papermaker wanting to increase the tearing resistance must compromise the other important mechanical properties of the sheet. This casts doubt on the usefulness of conventional tearing tests for evaluating the reinforcing potential of pulps.

The difficulty is resolved if the resistance of the sheet to crack propagation is measured for stresses in the plane of the sheet instead of by the Elmendorf or Brecht-Imset tear tests, which apply tearing forces perpendicular to the sheet's plane (**Fig. 4**). The resistance of paper to crack propagation depends on the propa-

gation mode, and it is different for the two modes shown in **Fig. 4**. In-plane fracture toughness increases with bonding (3) like tensile strength and elastic modulus, which are also measured under in-plane stress. During manufacturing, printing, and converting operations, when paper fails by crack propagation, stresses are applied in the plane of the sheet. Therefore, in-plane mechanical properties should be more relevant to end user than the out-of-plane tear tests. Previous work on newsprint showed that pressroom runnability correlated with in-plane fracture toughness of the sheet (4) but not with the Elmendorf tear.

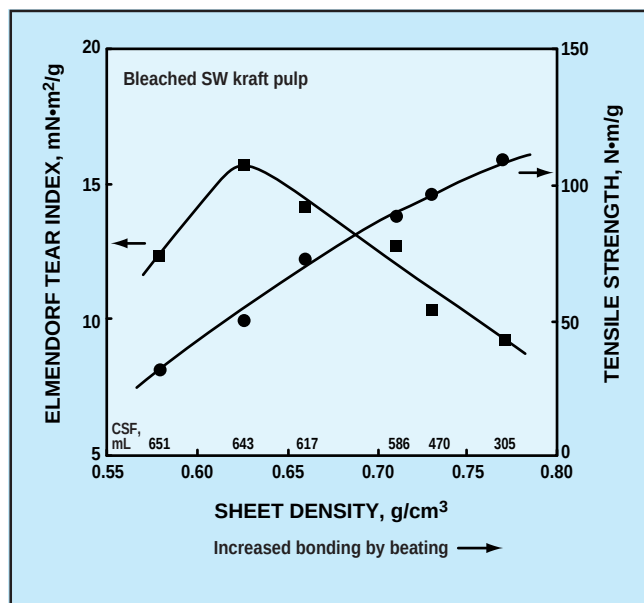
Methods to measure in-plane fracture toughness of tough, ductile sheet materials, such as handsheets of softwood kraft pulps, have been discussed (3, 5, 6). Fracture toughness of several softwood pulps has now been measured following laboratory beating and commercial refining. The effects of some fiber properties and papermaking treatments have also been examined. These results are presented in this report. They suggest a simple strategy to maximize the reinforcing potential of softwood pulps, and this is also discussed.

The effect of bonding on fracture toughness

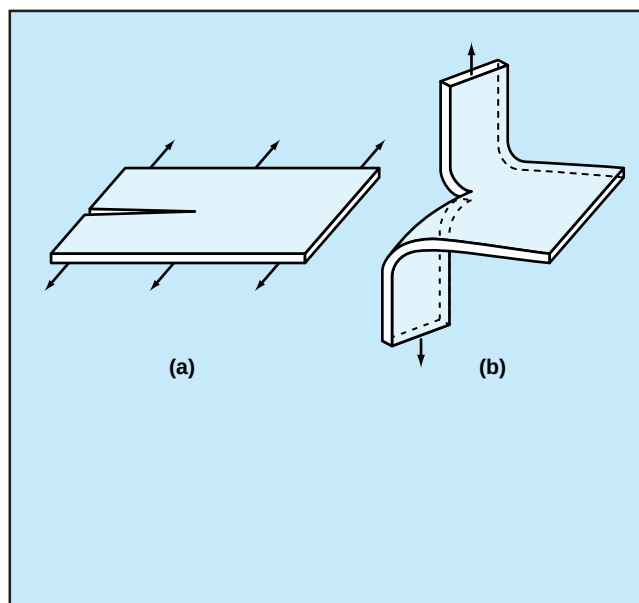
The effect of increased bonding by beating on fracture toughness is shown in **Fig. 5** for a commercial bleached softwood kraft pulp. The pulp was beaten in a PFI mill. The results of elastic modulus, tensile strength, and the Elmendorf tear index for this pulp were shown earlier in **Figs. 2** and **3**. Unlike the Elmendorf tearing resistance, which decreases with bonding after an initial increase, fracture toughness continues to increase before it levels off at high degrees of bonding. The relative changes in elastic modulus, tensile strength, extensibility, fracture toughness, and the Elmendorf tear index with beating from 640 to 470 mL freeness are given in **Table I**. The results in **Figs. 2, 3, and 5** and **Table I** are typical of softwood kraft market pulps when beaten in a PFI mill or in an Escher-Wyss laboratory refiner. [An Escher-Wyss laboratory refiner closely simulates mill refining conditions (7).] They suggest that a papermaker should refine a pulp to optimize tensile strength, extensibility, elastic modulus, and fracture toughness and ig-

Pulp Strength

3. With increased bonding, while the sheet tensile strength increases, for softwood pulps, the tear index first increases and then decreases for high degrees of bonding.



4. Modes of crack propagation in paper under (a) in-plane and (b) out-of-plane tearing forces



nore any loss in the out-of-plane Elmdorf tearing resistance.

The effect of increased bonding by increased wet-pressing pressure during sheetmaking is shown in **Table II**, and that from the addition of a bonding agent in the furnish is shown in **Table III**. The pulp in both cases was a laboratory-made, never-dried, unbleached kraft pulp of western hemlock. The bonding agent was locust bean gum. These results show that while an increase in fiber-fiber bonding increased all in-plane mechanical properties of the sheet—elastic modulus, tensile strength, extensibility, and fracture toughness—only the out-of-plane Elmdorf tear index decreased. They once again demonstrate the inadequacy of the Elmdorf tear for evaluating the reinforcing potential of softwood pulps.

The effect of intrinsic fiber properties on fracture toughness

To better understand in-plane fracture toughness, its dependence on intrinsic fiber properties—strength, length, and fineness—was investi-

gated and compared with that of the Elmdorf tearing resistance.

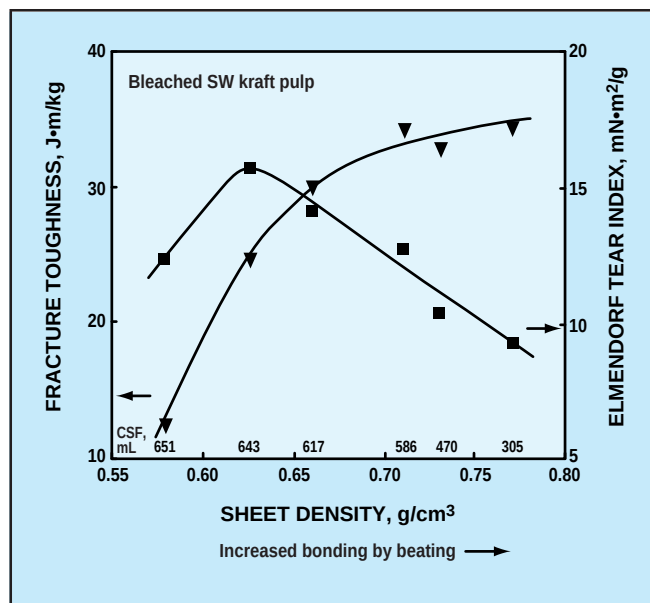
For well-bonded sheets, the Elmdorf tearing resistance of a sheet depends strongly on fiber strength (8). The effect of fiber strength on fracture toughness and the Elmdorf tear index for well-bonded sheets is compared in **Fig. 6**. Standard handsheets were made from a laboratory-made, never-dried, unbeaten, unbleached kraft pulp of western red cedar. The strength of fibers in the sheet was decreased by exposing the sheets to vapors of concentrated hydrochloric acid. This treatment allows the strength of fibers in the sheet to be decreased without affecting other fiber properties and the sheet structure (8). Fiber strength was measured as the zero-span tensile strength of the sheet. Sheets of different zero-span tensile strengths were obtained by varying the exposure time to the acid vapor. The results in **Fig. 6**, which are logarithmic plots of tearing resistance vs. zero-span tensile strength, show that, for a given level of bonding, both fracture toughness and the Elmdorf tear index depend similarly on the zero-span ten-

sile strength; this is indicated by the similar slopes of the logarithmic plots.

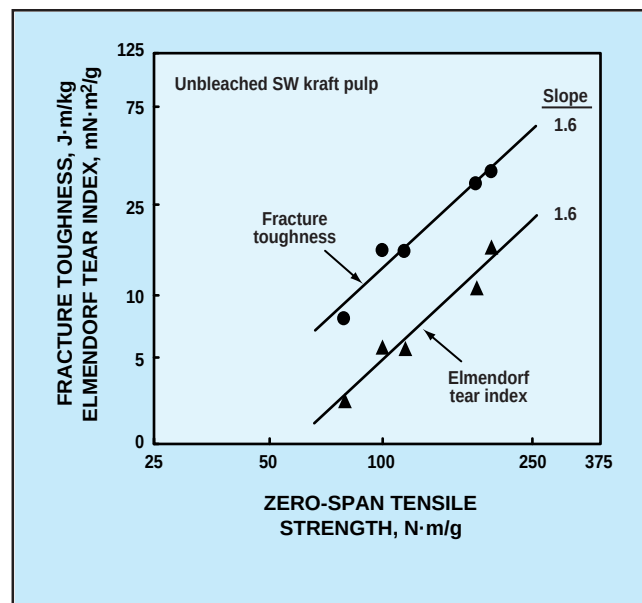
The Elmdorf tearing resistance of a sheet depends on fiber length, and for softwood pulps, the dependence is stronger for poor-to-moderately bonded fibers than for well-bonded fibers in the sheet (8). **Figure 7** shows the effect of fiber length on fracture toughness and the Elmdorf tear index for moderately bonded sheets. Fibers of different length but similar other fiber properties (e.g., strength, coarseness, curl, etc.) were obtained by guillotining dried handsheets of a laboratory-made, never-dried, unbeaten, unbleached kraft pulp of western red cedar. Standard handsheets were then made from guillotined fibers. The results in **Fig. 7**, which are logarithmic plots of tearing resistance vs. fiber length, show that, for a given moderate level of bonding, fracture toughness depends more strongly on fiber length than does the Elmdorf tear index; this is indicated by the slopes of the two lines.

The effect of fiber fineness on tearing resistance vs. tensile strength plots for two pulps is shown in **Fig. 8**.

5. While the out-of-plane Elmendorf tear index for softwood pulps decreases with increased bonding in the sheet, the in-plane fracture toughness continues to increase.



6. Dependence of in-plane and out-of-plane tearing resistance on fiber strength



I. The effect of increased bonding by beating on the mechanical properties of a commercial, dried, bleached softwood kraft pulp.

Parameter	Elastic modulus, kN-m/g	Tensile strength, N-m/g	Extensibility, %	Fracture toughness, J-m/kg	Elmendorf tear index, mN-m²/g
Lightly beaten (500 revs. in PFI mill, 640-mL CSF)	5.39	50.6	3.11	24.8	15.7
Well beaten (6000 revs. in PFI mill, 470-mL CSF)	7.43	96.3	3.55	32.8	10.3
Change, %	+38	+90	+14	+32	-34

The pulps were laboratory-made, never-dried, unbleached kraft of Douglas-fir (SWK1) and western red cedar (SWK2). Their fiber properties are given in **Table IV**. The pulps were beaten in a PFI mill to obtain a range of bonding levels in the fibers. The results in Fig. 8 show that, while coarse fibers have a high Elmendorf tear index, fine fibers have superior fracture toughness. Irrespective of how the comparison is made, at similar beating levels (**Table V**) or at similar drainage (**Table VI**), fine fibers have superior in-plane properties—elastic modulus, tensile strength, extensibility, and fracture toughness. This is because fine softwood fibers have thinner walls and, therefore, are more conformable;

they form larger contact areas. Fine fibers are more numerous per unit mass and have a higher surface area per unit mass. Therefore, they make more contacts per unit pulp mass. It is because of the better bonding that they produce sheets that are denser and less porous; they have superior in-plane strength properties but poor out-of-plane tearing resistance. Finer fibers also have a lower propensity to flocculate (9); they give sheets with better formation, smoothness, and opacity (10) and retain more filler (11).

It seems, therefore, that for softwood chemical pulps, the major difference between the two modes of crack propagation in Fig. 4 is in their response to increased fiber-fiber

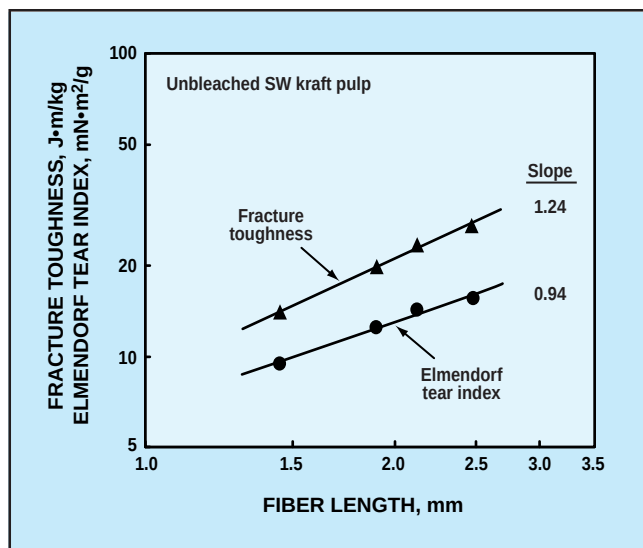
bonding. While the resistance to propagation under out-of-plane forces decreases with bonding, the resistance under in-plane forces increases.

Relationship between fracture toughness, tensile strength, and extensibility

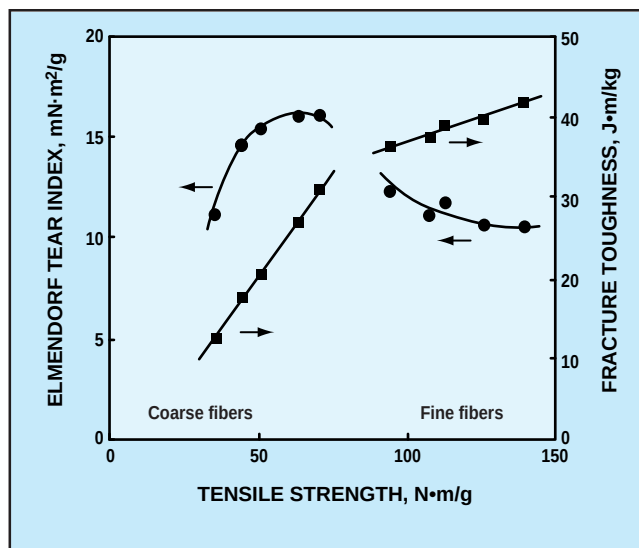
Fracture involves the creation of new surfaces at the expense of work. When a paper sheet containing a crack is strained, fracture processes (e.g., fiber breakage and bond breakage) are concentrated in the region just ahead of the crack tip. This region is called the *fracture process zone*. The work consumed in this

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7. Dependence of in-plane and out-of-plane tearing resistance on fiber length



8. Elmendorf tear index and fracture toughness as a function of sheet tensile strength for coarse- and fine-fibered softwood pulps



II. The effect of increased bonding by wet pressing on the mechanical properties of a never-dried, unbleached softwood kraft pulp

Parameter	Elastic modulus, kN-m/g	Tensile strength, N-m/g	Extensibility, %	Fracture toughness, J-m/kg	Elmendorf tear index, mN-m²/g
Sheets made at 138 kPa	4.55	39.9	2.25	17.9	14.0
Sheets made at 1.38 MPa	7.11	73.4	2.89	29.7	13.3
Change, %	+56	+84	+28	+66	-5

zone per unit crack area is the fracture toughness of the sheet. This work is supplied by the strain energy in the sheet and the applied external forces. Fracture toughness is high if the work consumed in the fracture process zone is high.

If it is assumed that the fracture process zone offers a high opposing stress that tends to prevent the crack from propagating (**Fig. 9**) and the strain associated with this stress is also high, then the resistance to crack propagation is high. It is difficult to precisely determine the stress and strain at the crack tip at propagation, but it can be assumed that they will be high if the unflawed material is strong under tensile load and has a high extensibility (3). In other words, the in-plane fracture toughness of the sheet will be high if it has high (in-plane) tensile strength and extensibility. Analyses on two sets

of results for softwood pulps show the following relationship:

$$F = 1.08T^{0.63} E^{0.52} \quad (1)$$

where

F = fracture toughness

T = tensile strength

E = extensibility

with $R^2 = 0.89$ for Set 1 containing 93 different handsheet samples, and

$$F = 0.60T^{0.74} E^{0.58} \quad (2)$$

with $R^2 = 0.91$ for Set 2 containing 123 different handsheet samples.

While fracture toughness measurements for samples of Set 1 were obtained by several operators, results for Set 2 were obtained by a single operator. In each set, the pulps included commercial and laboratory-made, bleached and unbleached kraft and were beaten or refined in a PFI mill, Escher-Wyss laboratory re-

finer, or commercial refiners. The strong dependence of fracture toughness on sheet tensile strength and extensibility suggests that fiber properties and papermaking treatments that enhance these two sheet properties are likely to enhance its fracture toughness as well.

The tensile strength of a sheet depends on the strength of the fibers and the extent of bonding between them. The extensibility of a sheet is a function of the extensibility potential of the fibers and the ability of the fiber network to utilize that potential, i.e., it also depends on the tensile strength of the sheet. If the sheet is weak and fails prematurely, the extensibility of the fibers is not utilized. For this reason, the sheet extensibility generally increases with an increase in tensile strength when the degree of bonding between fibers is increased (**Fig.**

III. The effect of increased bonding with a bonding agent on the mechanical properties of a never-dried, unbleached softwood kraft pulp.

Parameter	Elastic modulus, kN·m/g	Tensile strength, N·m/g	Extensibility, %	Fracture toughness, J·m/kg	Elmendorf tear index, mN·m/g
Without bonding agent	5.67	53.0	2.50	20.8	14.9
With bonding agent	5.62	61.0	2.85	27.7	14.4
Change, %	None	+15	+14	+33	-3

IV. Fiber properties of coarse- and fine-fibered, never-dried, unbleached softwood kraft pulps.

Parameter	SWK 1 Coarse	SWK 2 Fine
Fiber coarseness, µg/m	241	125
Number of fibers/g·10 ⁶	2.28	4.76
Mean fiber length, mm	3.36	2.86

10). Therefore, having a high sheet tensile strength is important for high fracture toughness. This may be the reason for the higher exponents of tensile strength in Eqs. 1 and 2 compared with those of the extensibility.

The extensibility potential of the fibers is enhanced by the induction of microcompression (12).

Implications for reinforcement and runnability

The purpose of adding reinforcement fibers into a furnish is to enhance its wet-web strength, the dry-sheet mechanical properties, and the strength of the dry sheet when rewetted. Previous work has shown that the strength of the wet web, i.e., its tensile strength and extensibility, is controlled by fiber-fiber interaction and the presence of curl and microcompression in the fibers (13). While curl and microcompression in fibers increase wet-web extensibility, factors such as fiber length, fiber fineness, conformability, fibrillation, and fines contribute to fiber-fiber interaction and therefore increase the tensile strength of the wet web (10, 13). It is only when the fiber-fiber interaction is high

that the extensibility potential of the fibers (provided by curl and microcompression) is fully utilized (13). It has also been shown that high wet-web tensile strength and extensibility are essential for good wet-end runnability (14–17).

Palsanen (18) showed that the mechanical properties of the base paper considered important for coater runnability are tensile strength and extensibility; the Elmendorf tearing resistance was not found to be important. It is apparent, therefore, that those fiber properties of the reinforcement pulps (e.g., fineness, conformability, fibrillation, etc.) and treatments (e.g., beating and refining) that increase fiber-fiber interaction in the wet-web or bonding in the dry sheet are likely to decrease the Elmendorf tearing resistance of the dry sheet. Consequently, basing the evaluation and use of reinforcement pulps on the out-of-plane tear index is flawed and may have hindered their effective use. The results reported here suggest that, paying little regard to the decrease in the Elmendorf or Brecht-Imset tearing resistance with increased bonding, the reinforcement pulps should be refined to a high tensile strength while maintain-

ing optimum drainage. Furthermore, instead of using the (out-of-plane) tear index at a given (in-plane) tensile strength as the criterion of “pulp strength,” criteria such as tensile strength at a given refining energy and drainage should be considered. These may prove more relevant for reinforcement and runnability. In addition to having the desired reinforcement in the furnish, expending minimum energy in refining and obtaining optimum drainage on the wire are some of the important goals of the papermaker using reinforcement pulps.

The various arbitrary strength criteria of pulp assessment, such as (tensile strength x Elmendorf tear index) or (tensile strength + 0.5 Elmendorf tear index) are simply attempts to downgrade the out-of-plane tear in favor of the in-plane tensile strength. This is because tear index has not proved useful for assessing the quality of reinforcement pulps. Since the tear index decreases only about 50% with increased beating or refining while the tensile strength increases 3- to 4-fold (Fig. 3), the above combinations of the two properties therefore increase, thus giving the semblance of a more relevant index for end use.

High tensile strength and extensibility of the reinforcement fibers translate into the high tensile strength and extensibility of the reinforced furnish (2).

V. Comparison of the mechanical properties of coarse- and fine-fibered, never-dried, unbleached softwood kraft pulps beaten for 6000 revolutions in a PFI mill.

Parameter	Elastic modulus, kNm/g	Tensile strength, Nmg	Extensibility, %	Fracture toughness, Jmg	Elmendorf tear index, mNm^2/g
Softwood kraft, coarse	6.96	69.6	2.30	31.1	16.1
Softwood kraft, fine	8.73	137	3.60	41.8	10.5
Change, %	+25	+97	+56	+34	-35

VI. Comparison of the mechanical properties of coarse- and fine-fibered, never-dried, unbleached softwood kraft pulps beaten to 500 mL CSF.

Parameter	Elastic modulus, kNm/g	Tensile strength, Nmg	Extensibility, %	Fracture toughness, Jmg	Elmendorf tear index, mNm^2/g
Softwood kraft, coarse	6.96	69.2	2.30	30.7	16.1
Softwood kraft, fine	8.53	129	3.35	40.5	10.8
Change, %	+23	+86	+46	+32	-33

Obtaining high sheet tensile strength

The tensile strength of paper is related to fiber properties and sheet structure through the following semiempirical equation (19):

$$\frac{1}{T} = \frac{9}{8Z} + \frac{12A \rho g}{PLb (RBA)} \quad (3)$$

where

- T = tensile strength of the sheet
- Z = zero-span tensile strength of the sheet
- A = fiber cross-sectional area
- ρ = density of the fiber wall material
- g = acceleration due to gravity
- P = perimeter of the fiber cross section
- L = fiber length
- b = bond shear strength per unit bonded area
- RBA = relative bonded area of the sheet.

It is assumed that Z reflects the tensile strength of the fibers. In Eq. 3, $A\rho$ = fiber coarseness, and $P/\rho A \approx$

SSA, the specific surface area of the fibers. It, therefore, is clear from Eq. 3 that the tensile strength of the sheet is high if the fibers are strong, long, and fine and have a high SSA. Fiber strength is controlled by fiber morphology and composition and pulping, bleaching, and pulp processing conditions (20).

The sheet tensile strength is also high if the bonding potential of fibers, $b \cdot RBA$, is high. The fiber-fiber bond shear strength b is high if the pulps are rich in hemicellulose content (21). Shear strength b is also increased by the presence of bonding agents such as starch or gum in the furnish (22). Shear strength b is adversely affected by pulp drying (23), and the loss in b is expected to increase with the degree of drying and the number of drying cycles (such as in recycling). The shear strength b is also expected to be adversely affected by the presence of debonding agents in the furnish.

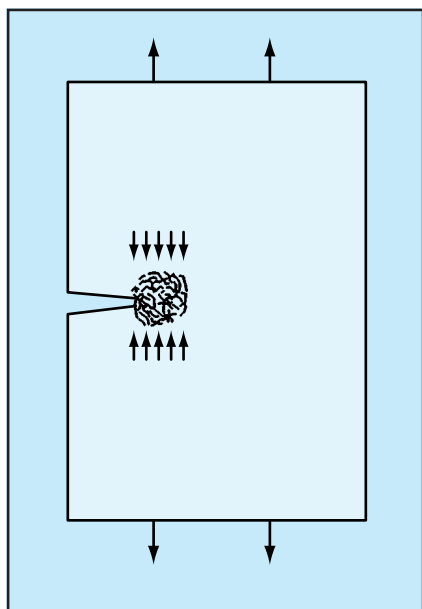
The sheet tensile strength is high if the relative bonded area, RBA, is high. High RBA also results in high sheet density. RBA depends on fiber conformability. Since finer softwood fibers have thinner walls (10), they are more conformable and

therefore require less refining energy to reach a high tensile strength. Pulps rich in hemicellulose content are more conformable (24) and therefore reach a high tensile strength with less refining energy. Fiber conformability is decreased when pulps are dried (25), and the loss in conformability is expected to increase with the degree of drying and the number of drying cycles. The relative bonded area can also decrease if chemicals that decrease the surface tension of water are present in the furnish.

It is assumed in Eq. 3 that fibers are straight and that the sheets are well formed. The presence of curl, kinks, and crimps in fibers (12) and poor sheet formation (26) adversely affect the tensile strength of the sheet. This is because these factors cause a poor stress distribution in the sheet, leading to its premature failure.

Curl, kinks, and crimps are induced in fibers by incidental mechanical treatment imparted during pulping and bleaching and are set in during pulp drying. Although fibers are partly straightened by swelling and mechanical stressing in subsequent low-consistency refining dur-

9. Stress at the crack-tip fracture process zone opposing crack propagation



ing papermaking (27), sufficient curl remains to give adequate wet-web stretch provided the fiber-fiber interaction in the web is high. If the fibers remain severely curled, crimped, and kinked, the tensile strength of both the wet web and the dry sheet is impaired (12).

One of the primary objectives of refining is to restore fiber conformability that is lost during drying and to enhance it. For a given refining energy input or freeness drop, low-intensity refining, which inflicts more impacts on fibers but of less severity (28), increases sheet tensile strength more than does high-intensity refining (29, 30), as also demonstrated in **Fig. 11**. Low-intensity refining preserves fiber length as well. Compared to high-intensity refining, it is likely that low-intensity refining is more effective in making fibers conformable and straighter through internal delamination and swelling, and it produces less external fibrillation and, therefore, flocculation.

Refining intensity can be altered by changing the plate design and the operating conditions of the refiner (28).

Concluding remarks

For softwood chemical pulps, the Elmendorf or Brecht-Imset tearing resistance of the dry sheet decreases with increased fiber-fiber bonding. Obtaining high out-of-plane tearing resistance contradicts optimizing the in-plane elastic modulus, tensile strength, extensibility, and fracture toughness of the sheet, which increase with bonding. In-plane mechanical properties of the sheet are more relevant to end uses of paper than the out-of-plane tear. Based on the out-of-plane tear criteria, the reinforcing potential of softwood pulps may not be fully utilized. A papermaker should refine a reinforcement pulp to obtain high tensile strength, extensibility, elastic modulus, and fracture toughness and ignore the loss of out-of-plane tearing resistance. Out-of-plane tearing resistance is relevant only where paper fails in that mode [Fig. 4(b)].

Measurement of fracture toughness of softwood chemical pulps at different refining levels shows that this property strongly depends on the tensile strength and the extensibility of the sheet. Fiber properties and treatments that enhance sheet tensile strength and extensibility also tend to enhance wet-web strength and the strength of the rewetted sheet. This suggests a simple strategy to best utilize the reinforcing potential of softwood market pulps in papermaking—refine them to a high tensile strength while maintaining acceptable drainage. Refining to a high tensile strength generally results in a high sheet extensibility as well.

A mill producing lightweight coated papers is already taking advantage of these findings (31). By refining its softwood pulp to a high sheet tensile strength using a lower refining intensity, it is able to increase its production speed and decrease the number of breaks.

Experimental

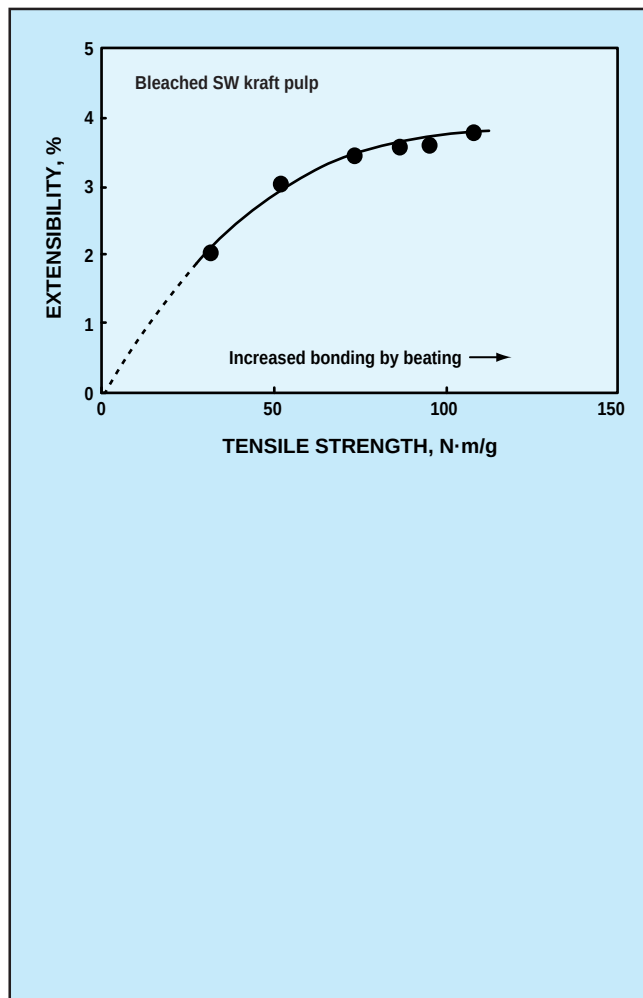
The results in Figs. 2, 3, and 5 and Table I were obtained on the NIST reference pulp (32). The laboratory-made pulps of Figs. 6–8 were cooked to kappa no. 30. Standard procedures of the Technical Section, CPPA were generally followed for handling pulps, for specimen preparation, and for measuring physical properties. Fiber length and coarseness of pulps were measured with a Kajaani FS-100 fiber analyzer. The sheet properties were measured at 23°C and 50% relative humidity following prolonged conditioning of the samples. All results are based on air-dry grammages. The Elmendorf tear index was obtained on single-ply samples. This is because multi-ply test results depend on the number of plies torn together and, therefore, do not measure a material property (33). ■

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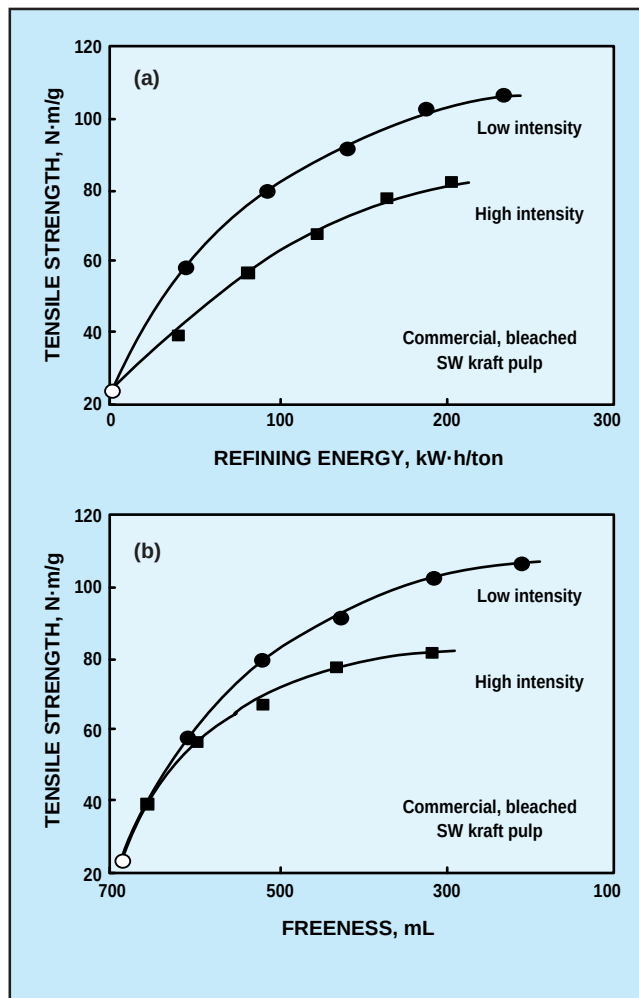
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Pulp Strength

10. Extensibility of the sheet increases as the tensile strength increases with increased degree of bonding. The pulp is the same as that shown in Figs. 2, 3, and 5.



11. At a given (a) refining energy or (b) freeness, low-intensity refining gives a higher tensile strength. Pulp was refined in an Escher-Wyss laboratory refiner. Specific edge loads, denoting refining intensities, were 1 and 4 W·s/m, respectively.



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I thank Yuxiu Peng of Nanjing Forestry University for the results presented in Tables II and III and Fig. 7, which she obtained while at Paprican as a visiting scientist during 1991–92. Jim McPhee of Paprican's Pointe Claire laboratory is thanked for preparing pulps. John Hoffmann, Chengbao Wu, and David Lo are thanked for their skilled technical assistance in experimental work; Ben Chan for statistical analysis; and Kelly Wadham for preparing the manuscript and illustrations. David Lo was employed during the summer of 1992 on an NSERC Industrial Undergraduate Student Award.

Received for review Jan. 25, 1995.

Accepted April 10, 1995.