

Mechanism of paper tensile-strength development due to pulp beating

Sunil Dasgupta

ABSTRACT: *Pulp refining produces rapid increases in sheet mechanical properties. These increases level off at moderate levels of beating, while prolonged beating reduces tensile strength and elongation. This work presents laboratory data for handsheets prepared from dried pulps beaten to various levels of freeness. The data are analyzed using Page's equation, which relates tensile strength to b , the bond strength per unit of bonded area within the sheet. Calculated values for b increased with pulp beating and showed moderate correlation with increases in specific bond strength (SBS). The increases in b and SBS are attributed to the increase in the number and frequency of fiber-to-fiber bonds per unit optical area. Equivalent increases were obtained with a suitable chemical strength additive. This increase in bonding is postulated to be the main reason for the rise in paper tensile strength due to pulp beating.*

KEYWORDS: *Beating, bonding strength, elongation, paper, pulps, rupture work, tensile strength.*

The effect of pulp beating, or refining, on cellulose fibers is a complex subject. Even a modest understanding of the process requires knowledge of several difficult areas, including fiber structure and uniformity, the stress-strain properties of fibers, and the changes in fiber properties during the beating process. The effort to extrapolate the effects of pulp beating on paper properties

introduces another level of complexity, and this remains a challenging area of study.

The literature on this subject is extensive and includes descriptions of ongoing research and reviews of past research (1-12). In 1985, Page (7) critically analyzed the existing theories describing the mechanism of paper strength development during beating of once-dried pulps. He

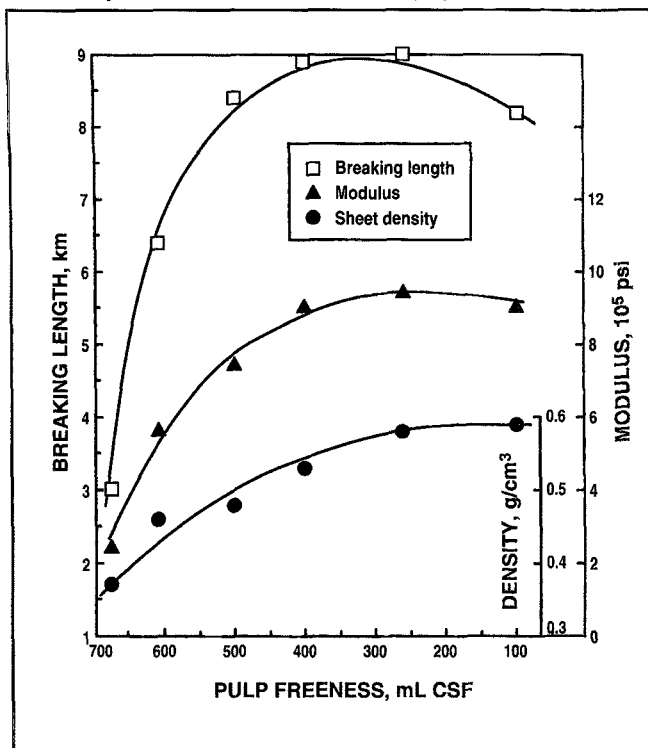
pointed out, with experimental data to support his view, that none of the theories adequately explained all of the observed effects of refining on the mechanical properties of paper.

Of all the existing theories about the development of paper strength during beating, the Page-Tydemann effect (6) offers the most satisfactory explanation of the observed data. This theory proposes that the major effect during beating of dried pulps is the straightening-out of both the long- and short-range crimps and curls that are created in fibers during pulping, pressing, and drying of the pulp. Removal of these crimps and kinks in the fibers greatly improves the stress distribution in the sheet. Page estimates that this mechanism is responsible for at least 50% of the increase in tensile strength properties. An increase in bond strength per unit area in the bonded region and fibrillation of the fiber surface jointly contribute to the rest. Page *et al.* (12) have also evaluated the application of this theory to the mechanism for the increase in strength and elastic modulus.

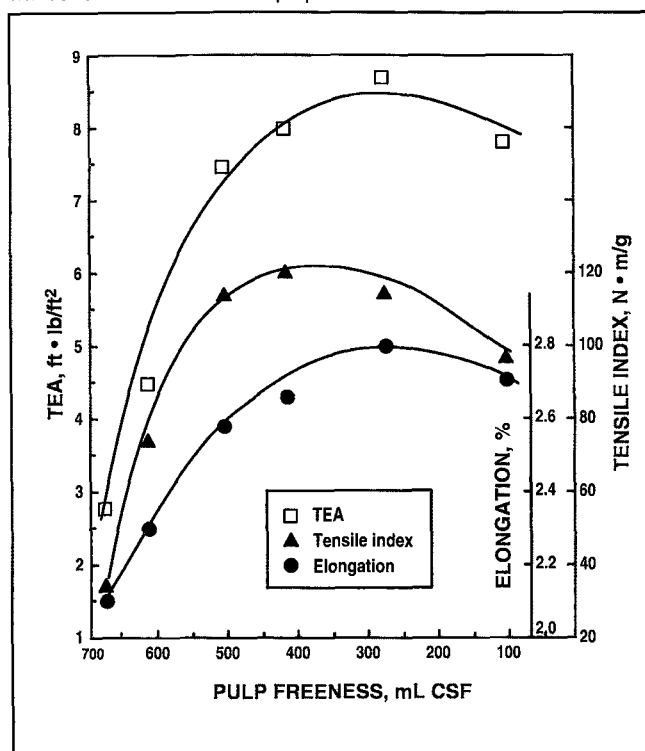
The present study evaluates the effects of pulp beating on paper strength by using Page's equation (13) as a means of analyzing the mechanism of tensile-strength development during refining. The tensile strength, in this case, is analyzed in terms of relative bonded area, fiber length, and other basic parameters for fiber dimensions. To complement this analysis, fiber-to-

Dasgupta is a research scientist in the Paper Technology Group at the Hercules Inc. Research Center, Wilmington, DE 19808-1599.

1. Tensile strength (measured as breaking length), elastic modulus, and density of handsheets as functions of pulp freeness



2. Tensile energy absorption, elongation, and tensile index of handsheets as functions of pulp freeness



fiber bond strength and specific bond strength in paper were also studied using the sheet-delamination technique of Skowronski and Bichard (14). This work is presented in the hope that it will contribute to the goal of understanding how pulp beating affects the mechanism of strength development in paper.

Results and discussion

Once-dried pulps were reslushed to a freeness of 675 mL CSF and beaten to freeness levels ranging from 100 mL CSF to 600 mL CSF. Handsheets prepared from these pulps were tested for strength and other physical properties.

Results for tensile strength (measured as breaking length), sheet density, and elastic modulus are presented in Fig. 1. Results for sheet elongation, tensile index, and tensile energy absorption (TEA) are presented in Fig. 2. Within the freeness range used, all six of the measured paper properties were affected by

the level of beating. Figure 1 shows that tensile strength (measured as breaking length), modulus, and sheet density increase with the level of pulp beating until they level off at moderate levels of pulp freeness. The response of tensile strength to beating is more complex, with strength peaking and then dropping at low pulp freeness. Figure 2 shows that the level of beating has a similar effect on TEA, sheet elongation, and tensile index, with these properties also peaking and dropping at high levels of beating.

Page has theorized (7) that the increases in elastic modulus and in tensile strength are primarily due to improved stress distribution within the sheet, the result of the straightening of crimps and curls in the fibers during the initial stages of refining. This theory is particularly appealing in the case of elastic modulus, where the behavior cannot be attributed to bond strength. The decrease of tensile strength after prolonged beating is possibly the result

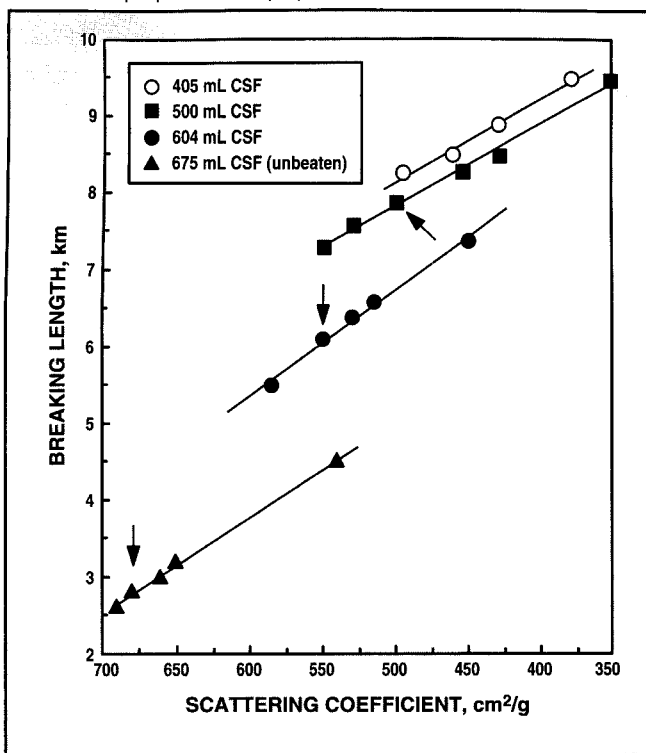
of damage to the fiber structure and the fiber cell wall (4), leading to a reduction in fiber strength.

Sheet properties at various wet pressures and pulp freenesses

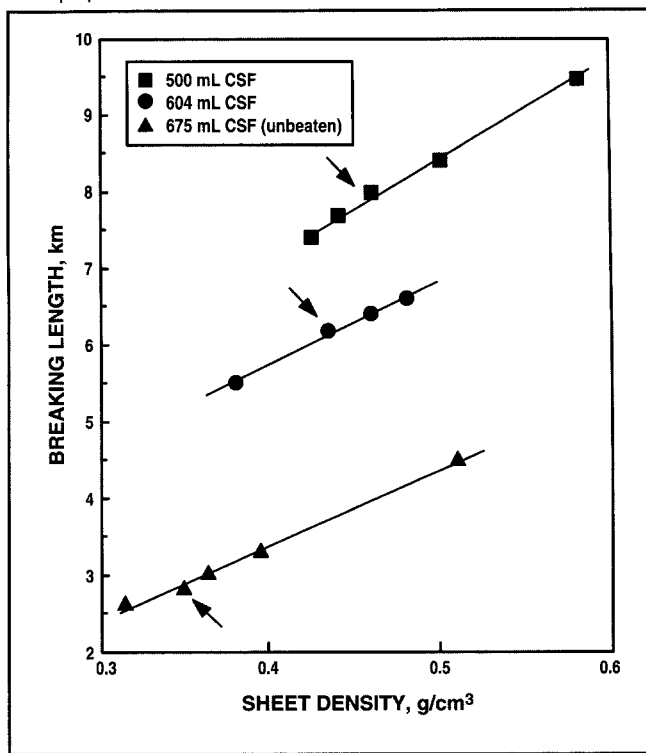
We generated a substantial amount of data for handsheets produced from pulp beaten to various levels of freeness and prepared at several wet pressures. For each set of conditions, handsheets were prepared with and without a strength additive.

Tensile strength is plotted as a function of light-scattering coefficient and density in Figs. 3 and 4, respectively. Elastic modulus is plotted as a function of light-scattering coefficient and density in Figs. 5 and 6, respectively. As Page has shown (7), the tensile strength of a sheet produced from unbeaten pulp at any specific density or light-scattering coefficient is much lower than that of a sheet produced from beaten pulp at the same density or scattering coefficient. From results in Figs. 3 and

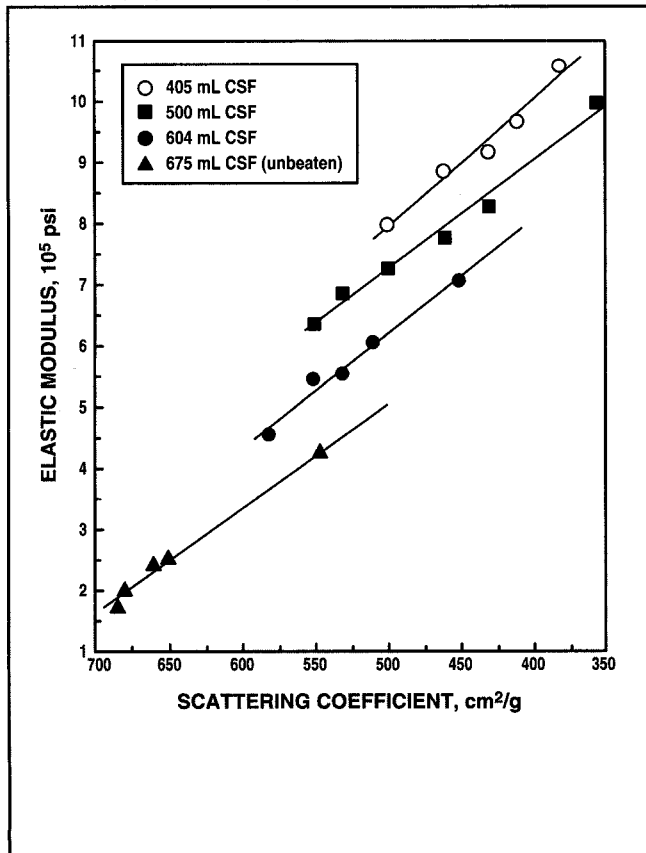
3. Breaking length as a function of light-scattering coefficient for handsheets prepared from pulps at various levels of freeness



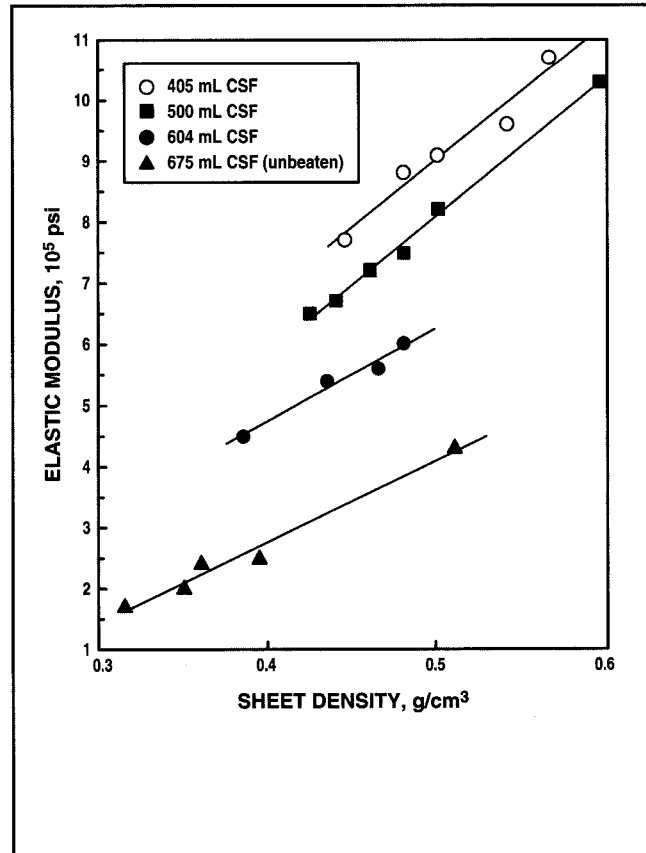
4. Breaking length as a function of density for handsheets prepared from pulps at various levels of freeness



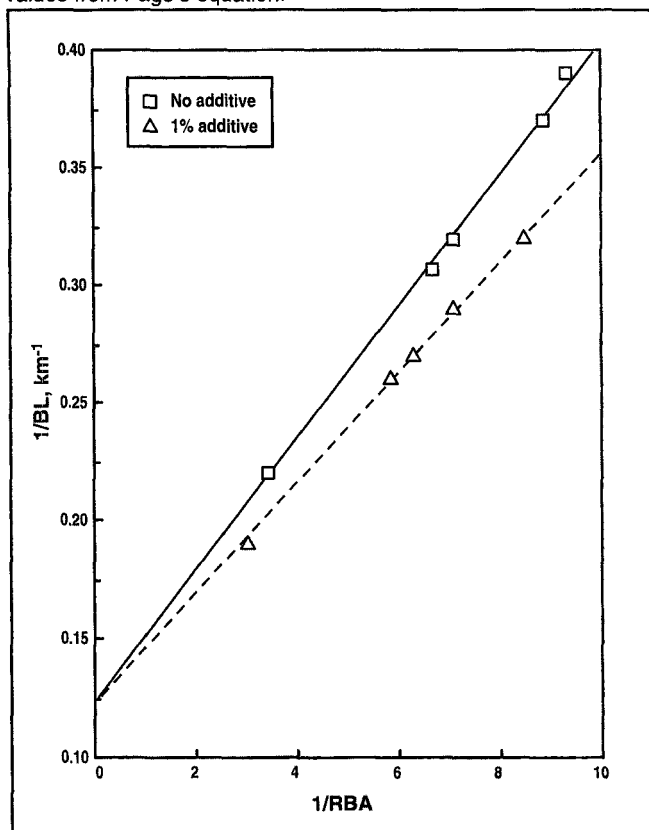
5. Elastic modulus as a function of light-scattering coefficient for handsheets prepared from pulps at various levels of freeness



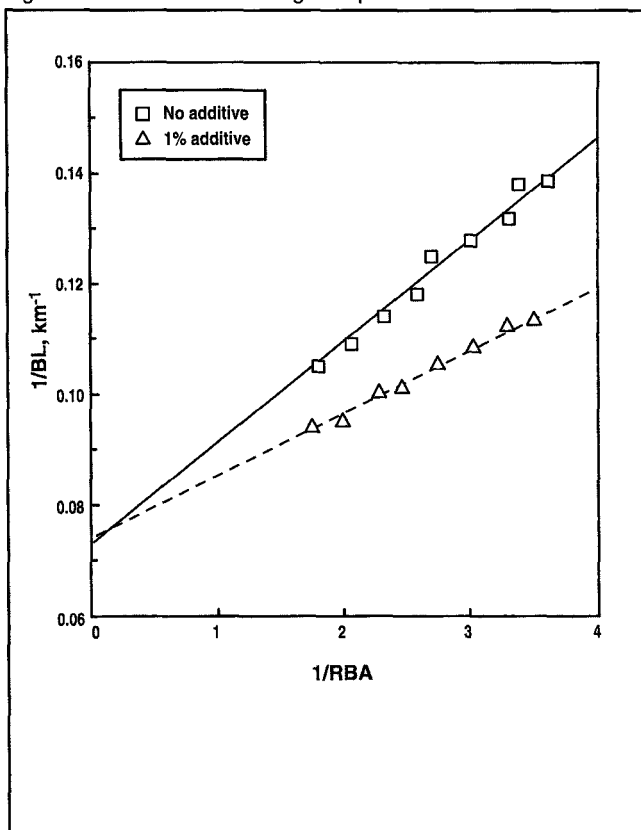
6. Elastic modulus as a function of density for handsheets prepared from pulps at various levels of freeness



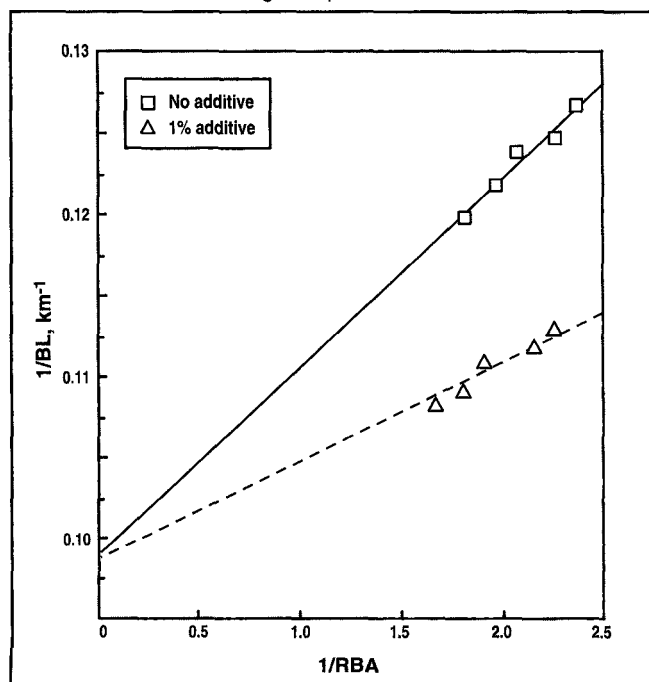
7. Inverse of breaking length vs. inverse of RBA for handsheets prepared from unbeaten pulp (675 mL CSF) with and without strength additive. Data points coincide with the lines predicting theoretical values from Page's equation.



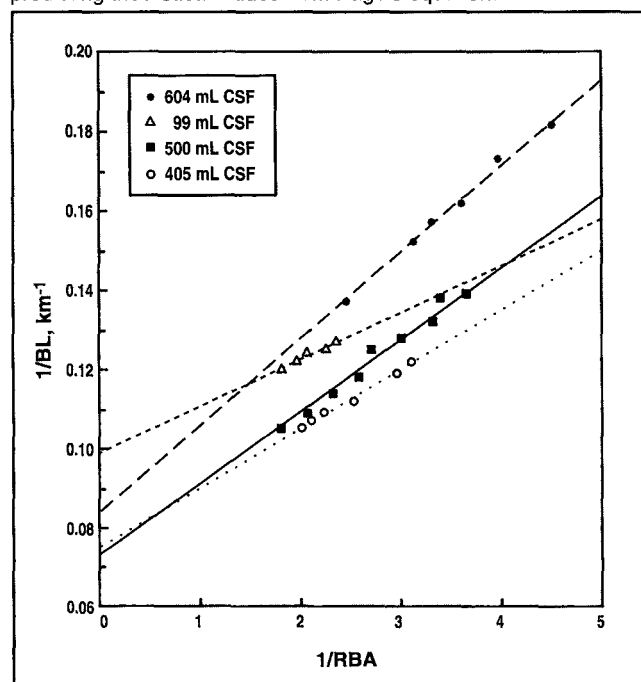
8. Inverse of breaking length vs. inverse of RBA for handsheets prepared from moderately beaten pulp (500 mL CSF) with and without strength additive. Data points coincide with the lines predicting theoretical values from Page's equation.



9. Inverse of breaking length vs. inverse of RBA for handsheets prepared from heavily beaten pulp (99 mL CSF) with and without strength additive. Data points coincide with the lines predicting theoretical values from Page's equation.



10. Inverse of breaking length vs. inverse of RBA for handsheets prepared from pulps beaten to various levels of freeness and containing no strength additive. Data points coincide with the lines predicting theoretical values from Page's equation.



Paper Strength

4, by comparing the data points indicated by arrows, only 30–35% of the increase in tensile strength for the beaten handsheets (604 mL CSF) can be accounted for by densifying sheets produced from unbeaten pulp. Modulus data (Figs. 5 and 6) show similar results. These results qualitatively agree very well with those of Page (7).

Analysis of results using Page's theory

The Page equation (13) was used to analyze the tensile-strength data obtained on handsheets prepared at various wet pressures from pulps beaten to various levels of freeness. In each case, duplicate sheets were prepared with 1% of a strength additive (guar gum) based on dry pulp weight. The objective was to generate information on zero-span tensile strength Z , which is the effective fiber strength, and on bond strength per unit bonded area, b . The analysis was carried out in an effort to understand the relative contribution of fiber-to-fiber bonding to the increase in tensile strength obtained by beating of pulp.

The Page equation (13) can be written as follows:

$$1/T = (9/8Z) + [(12 Apg)/(b P L RBA)] \quad (1)$$

where

T = tensile strength, expressed as breaking length (BL)

A = average fiber cross-sectional area

ρ = density of fiber material

g = acceleration due to gravity

P = perimeter of fiber cross section

L = average fiber length

RBA = relative bonded area.

RBA was determined by light-scattering measurements according to the method of Ingmanson and Thode (15).

Figures 7–10 plot the inverse of breaking length ($1/BL$ or $1/T$)

I. Average fiber length and calculated values (Page's equation) of Z and b for handsheets prepared from pulps at various levels of freeness

Pulp freeness, mL CSF	Average fiber length, mm	Zero-span tensile strength, km	Bond strength per unit of bonded area (b), N/cm ²	
			No strength additive	1% strength additive*
675 (unbeaten)	2.20	9.0	600	840
604	2.09	13.4	790	1120
500	2.04	15.6	1060	1600
405	1.94	15.2	1420	2110
266	1.84	15.5	...	...
99	1.63	11.3	2250	3480

*Guar gum, based on dry pulp weight

against the inverse of RBA. The whole subject of paper strength properties is complex, and there are several uncertain factors involved in the process of evaluating paper strength. Despite these difficulties, the "Page plots" in Figs. 7–10 show an almost perfect fit, as predicted in the theory.

Table I shows the zero-span tensile strength, Z , and bond strength per unit of bonded area, b , as calculated from the Page equation using the slopes of these plots and their intersections on the $1/BL$ axes. Figure 11 shows the near-perfect linearity of calculated values of b with pulp freeness. Figure 12 shows the calculated values of Z and average fiber length, L , as functions of pulp freeness. The plot of Z vs. pulp freeness shows good correlation with the results in Fig. 1 for tensile strength vs. freeness. The decrease in Z at high beating levels reflects a degradation of fiber strength. Measured fiber length possibly has little influence on tensile strength in this case.

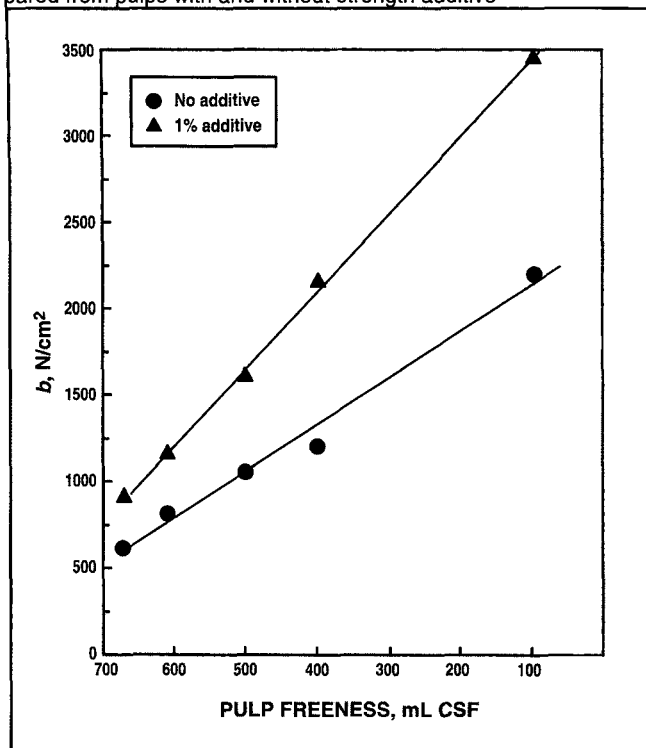
As mentioned earlier, the main emphasis of this study was to evaluate the relationship between b (bond strength per unit of bonded area) and pulp beating. The results in both Table I and Fig. 11 show that b increased by 30–45% from one beating level to the next. This is the range

of increase we also observed in tensile strength (BL) with increased beating. Figure 11 also demonstrates that a suitable strength additive enhances b to a comparable extent in sheets independent of the effect of beating. Thus, fiber-to-fiber bonding is the major contributor to the increase of tensile strength in paper due to pulp beating.

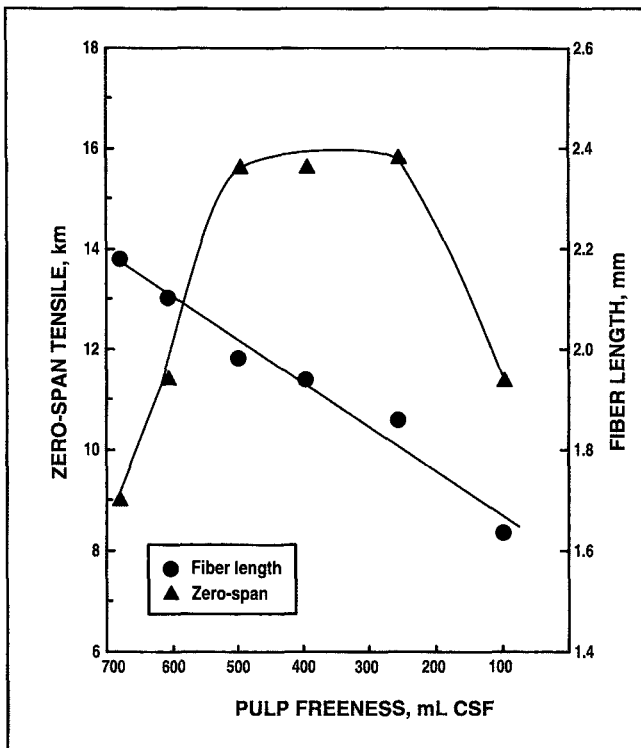
How to explain the increase in b due to beating of pulp? Results obtained by Page (16) and Page *et al.* (17) have revealed a great deal about the nature of fiber-to-fiber bonds in paper. Their work has provided substantial insight into the effects of pulp beating on the size, shape, and the abundance or frequency of these bonds. Beating increases the "degree of bonding"—the ratio of bonded area to the projected area of crossing between two fibers—as well as the "frequency of bonding."

We employ light-scattering measurements to determine the bonded area, which is used to calculate specific bond strength. The area of optical contact represents a region where two fibers are separated by less than 150 Å (18). Since the bonded area is generally smaller than the crossover area in the case of unbeaten pulp, it is likely that the optical-contact area includes a substantial portion of unbonded area in this case. With the increase of the extent of beating, the

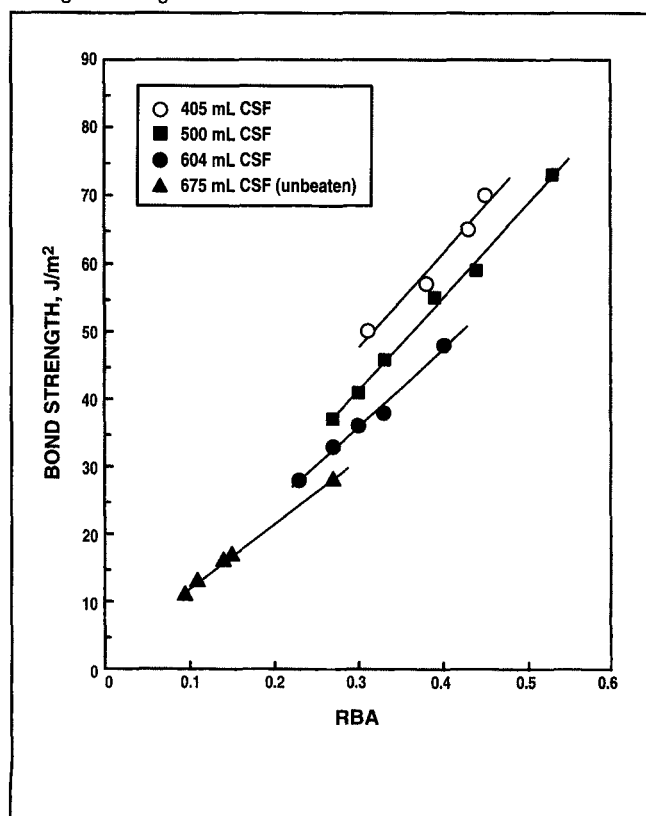
11. Calculated values (Page's equation) of b (bond strength per unit bonded area) as a function of pulp freeness for handsheets prepared from pulps with and without strength additive



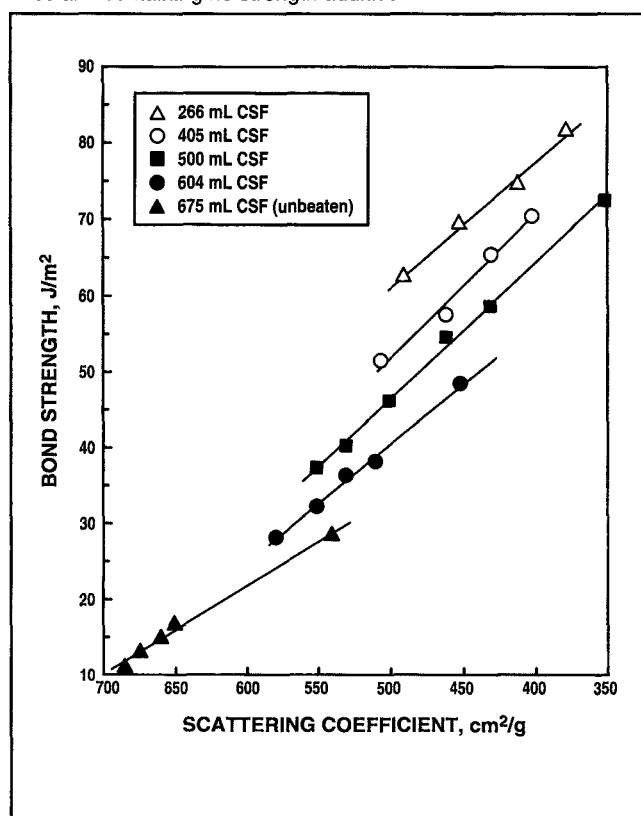
12. Calculated values (Page's equation) of zero-span tensile strength and average fiber length as functions of pulp freeness



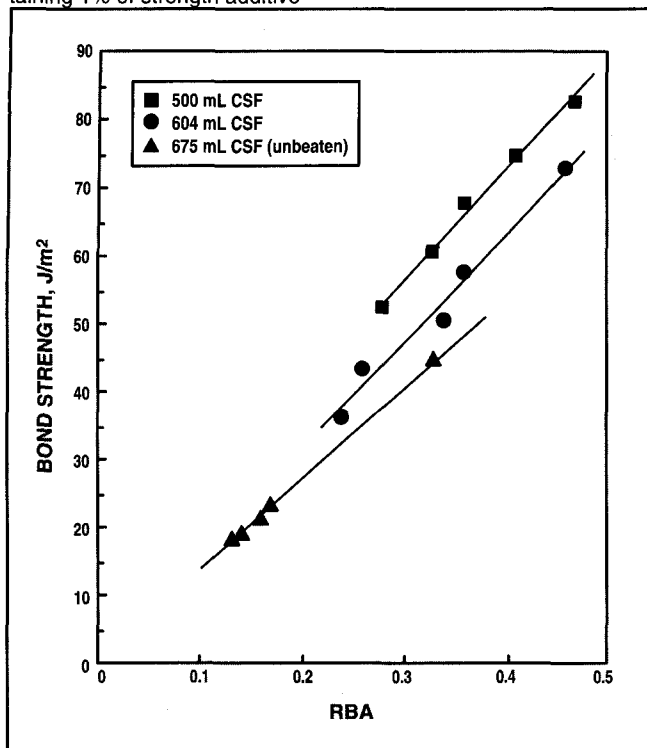
13. Bond strength as a function of relative bonded area for handsheets prepared from pulps beaten to various levels of freeness and containing no strength additive



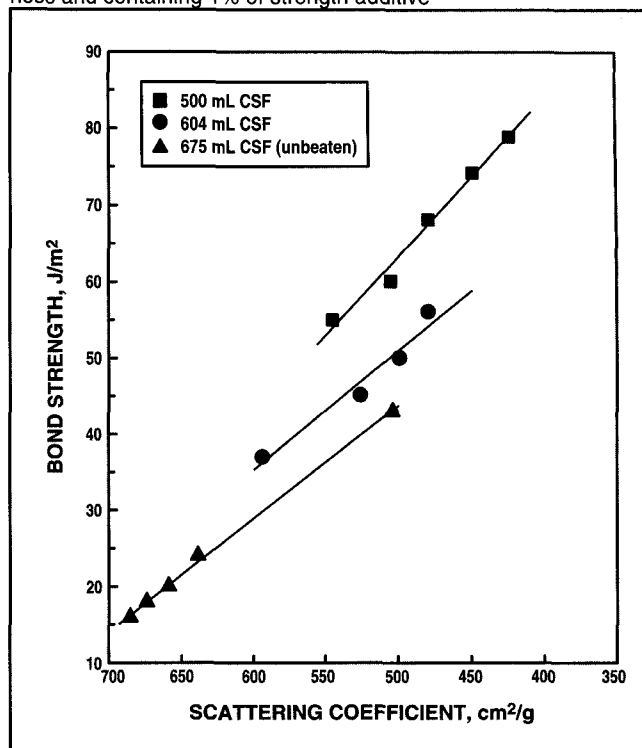
14. Bond strength as a function of light-scattering coefficient for handsheets prepared from pulps beaten to various levels of freeness and containing no strength additive



15. Bond strength as a function of relative bonded area for handsheets prepared from pulps beaten to various levels of freeness and containing 1% of strength additive



16. Bond strength as a function of light-scattering coefficient for handsheets prepared from pulps beaten to various levels of freeness and containing 1% of strength additive



optical-contact area includes an increasing amount of actually bonded area. Thus beating of pulp increases the number of bonds while also reducing the distance between bonds.

Bond strength as measured by sheet-delamination technique

Bond strength (BS) and specific bond strength (SBS) were determined by delaminating handsheets using the technique of Skowronski and Bichard (14). The paper sample was delaminated using an Instron tester equipped with a free-rotating wheel assembly. The force required to delaminate the sample was continuously recorded on the Instron chart recorder. The bond breaking energy was determined from the area under the curve.

The change in optical-contact area as a result of delamination was calculated on the basis of the Kubelka-Munk definition (19). The change in the area of optical contact was assumed to be equivalent to the area created during bond failure.

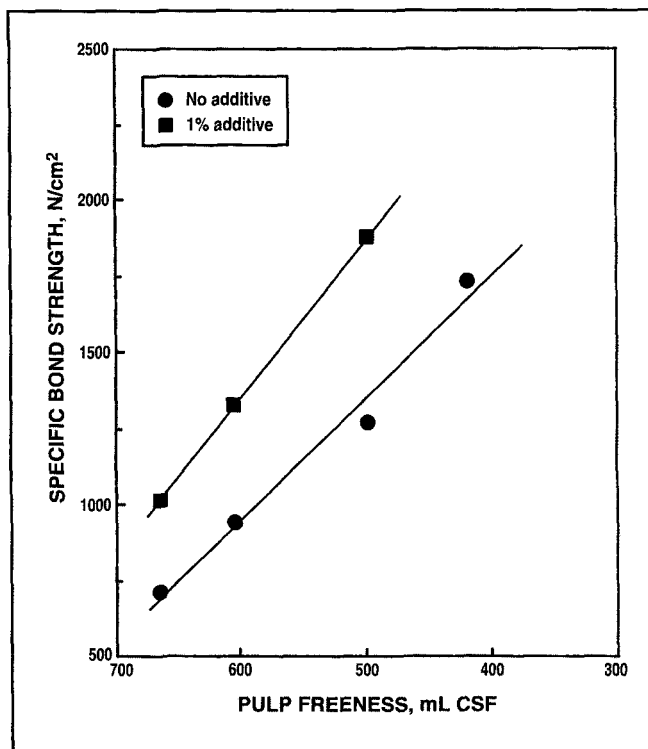
Bond strength is defined as the average bond breaking energy per unit area of sample. Specific bond strength is defined as the bond breaking energy per unit of optical surface area created by broken bonds. Results for bond strength are plotted against relative bonded area and light-scattering coefficient in Figs. 13 and 14, respectively. Similar plots of bond strength obtained with sheets containing 1% of a strength additive are presented in Figs. 15 and 16, respectively. In each case, it is obvious that at any specific value of scattering coefficient or relative bonded area, increased beating of pulp produces a significant increase in the amount of fiber-to-fiber bonds within a sheet. Stratton (20) has observed z -direction paper properties and reported that refining improved bonding.

Table II and Fig. 17 present results for the specific bond strength. SBS was determined as follows: At each level of wet pressing, an SBS value was calculated according to the

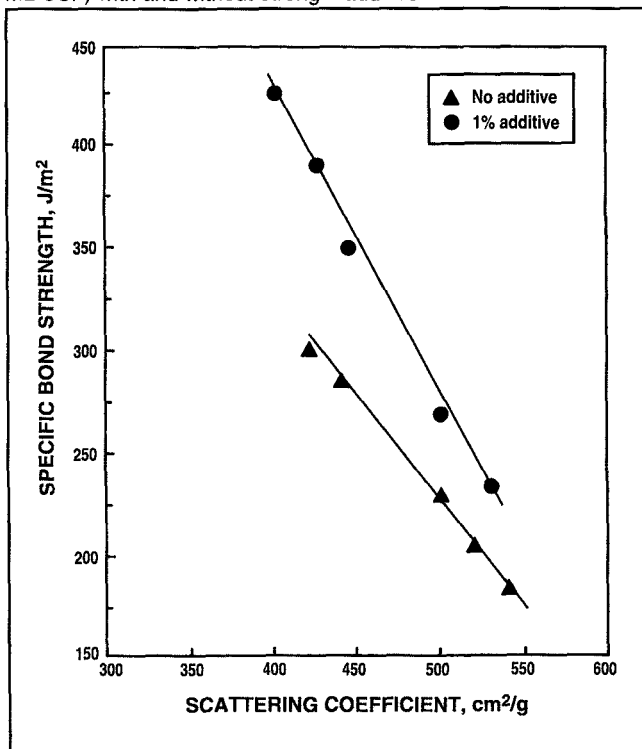
method outlined by Skowronski and Bichard (14). Obviously, the SBS values thus obtained are dependent on the degree of wet pressure. Consequently, these SBS values were plotted and extrapolated to zero scattering coefficient. Table II and Fig. 17 are based on these extrapolated SBS values.

The extrapolated SBS data show a reasonable agreement with the values of b (Fig. 11), particularly in view of the possibility that the measured fiber length, L , used in calculating b may have been incorrect. Note that the increase in SBS was 30–40% when the unbeaten pulp was beaten to 604 mL CSF and again when the pulp was further beaten from 604 mL CSF to 500 mL CSF. This agrees quite well with the behavior observed for b . Moreover, SBS values were similarly enhanced by the addition of a suitable strength additive. The arguments presented to explain the increase in b with pulp beating can also be applied to explain the increase in SBS with beating.

17. Specific bond strength as a function of pulp freeness for handsheets with and without strength additive



18. Specific bond strength as a function of light-scattering coefficient for handsheets prepared from moderately beaten pulp (500 mL CSF) with and without strength additive



II. Specific bond strength (SBS)^a of handsheets prepared from pulps at various levels of freeness

Pulp freeness, mL CSF	SBS, N/cm ²	
	No strength additive	1% strength additive ^b
675 (unbeaten)	710	1000
604	940	1330
500	1270	1880
405	1730	...

^aAs determined by sheet-delamination technique of Skowronski and Richard (14)
^bGuar gum

These results contradict the conclusions of Skowronski and Richard (14) and Nordman *et al.* (21). Skowronski and Richard (14), from a very limited set of data, concluded that SBS was not dependent on pulp beating. Nordman *et al.* (21), on the other hand, chose not to employ the rupture strength and totally formed bonds. Instead, they generated straight lines by

plotting a "consumed" energy to break "a certain number of bonds" against the corresponding increases in scattering coefficients. They found the slopes of these straight lines, called "bond strength" (BS), to be independent of the degree of pulp beating.

Conclusion

The mechanism by which tensile strength is affected by pulp refining was examined by focusing on the strength of bonds within handsheets prepared from once-dried pulp. Data on bond strength per unit bonded area, *b*, were analyzed using the Page theory (13). A sheet-delamination technique was used to obtain additional data for bond strength and for specific bond strength. The study included sheets with and without a strength additive. The results indicate that increases in the number and the frequency of fiber-to-fiber bonds play the major role in the enhancement of tensile strength of paper resulting from pulp beating. The adverse effect of high levels of

pulp beating on tensile strength possibly reflects damage to the fiber and subsequent degradation of fiber strength.

Experimental

The pulp used in this work was a 70/30 combination of once-dried northern softwood kraft and chemithermomechanical pulp produced from softwood.

Handsheet preparation

The pulp was refined in a Valley beater to specific levels of freeness. The pulp slurry (2.50% consistency) was diluted to 0.266% solids with normal tap water in a proportioner. For handsheets produced with a strength additive, the chemical was added to the pulp at this point while stirring. An aliquot of this pulp slurry was further diluted to about 0.023% consistency in a deckle box for molding handsheets. Both refining and papermaking were done at pH 7.5–8.0.

Paper Strength

Measurement of handsheet properties

Tensile strength of the handsheets (1-in.-wide samples) was determined in an Instron at a drawing rate of 0.5 in./min and a span of 4 in. The dimensional parameters of the fibers, such as radii in the case of an elliptical cross-section, were determined using a microscope as described by Van den Akker (22). Many of the fibers examined were in collapsed form. Light-reflectance values R_0 and R_∞ of the handsheets were measured in a Diano S-4 brightness tester according to a procedure described in TAPPI Test Method T 220 om-88. Average fiber length was determined in a Kajaani fiber analyzer (model FS-100).

Bond strength was measured by the Skowronski-Richard sheet-delamination technique (14). The free rotating wheel, built according to their description, was attached to a tensile tester (TH-1-AB Lorentzen & Wettre). The calculations of bond strength (BS) and specific bond strength (SBS) were made according to the following relations.

$$BS = W/LR$$

where

W = bond breaking energy measured during delamination

L = length of delamination

R = sample width

and

$$SBS = W/\Delta C$$

where

ΔC = change in optical contact area due to breakage of fiber-to-fiber bonds, calculated from light-reflectance values (19).

The highest value of bond strength to be measured by this method is obviously limited by the adhesion strength of the tape to the paper surface. This method for measuring bond strength (in particular) and for calculating SBS (when high accuracy in measuring light scattering is pos-

sible) was found to be a dependable tool in this study.

Conceptually, SBS and b should be the same, since "unit area" is measured optically in both cases. However, the methods used to measure SBS and b are very different. The following procedure was used to compare the results from the two methods: Figure 18 shows a linear relationship between SBS and the sheet's light-scattering coefficient. The scattering coefficient of paper in this case is controlled by the degree of beating and by wet pressure used during sheet formation (and by the presence of strength additive). Hence, to obtain a representative value of SBS for paper made with pulp having a specific degree of beating, the linear plots of SBS against scattering coefficient were extrapolated to S_0 . The SBS values thus obtained are expressed as energy per unit of area (Joule/m²), while the values of b are usually calculated as force per unit of area (Newton/cm²). Thus, the final values of SBS were converted to force units by normalizing the SBS values to unit sample thickness. The agreement between the SBS values thus obtained and the values of b is reasonable given the complexity of the method used to derive values for both b and SBS.

Additive

Guar was the strength additive employed in this work. Guar gum is a natural alternating copolymer consisting of galactose and mannose, usually in the ratio of one to two. The accepted structure of guar is as a linear chain of β -D-manopyranosyl with α -D-galactopyranosyl units as side branches (23). **[7]**

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