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A new method for shear bond strength measurement

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ABSTRACT

Modeling the tensile strength of paper has been a topic of interest over the last few decades. Fibre properties and fibre network properties are the two main sets of inputs required for any such analytical model. Out of these, the most difficult quantity to measure is the "Fibre-Fibre Shear Bond Strength". No reliable method exists for the measurement of this quantity. This paper reports a simple method based on the idea of weakening the fibres independently of the bonds in a sheet of paper, using acid hydrolysis until all the fibres break across the fracture line. The bond strength is then calculated from the fibre strength, as measured by the zero span test at the point where the fibres first are weakened such that they all break. The method was used to calculate the average bond strength of handsheets made out of never dried, 60% yield unbleached Radiata Pine fibres and is found to be 26.9 ± 0.5 MPa. This is considerably higher than most previous estimates for the shear bond strength. The limitations of the methods, which have been used previously used to measure the fibre-fibre shear bond strength, will be discussed in this paper in detail.

INTRODUCTION

Researchers have previously taken five different approaches to designing experiments that measure the shear bond strength of wood pulp fibres. Most of the research has focused on using single fibre joints prepared by various techniques.

The pioneering work in this field was that by McIntosh and Leopold *(1)*. They glued a single fibre (Unextracted and Alkali extracted Loblolly pine holocellulose) across a few shives, holding this assembly in place with cellophane. This assembly was put under heat and pressure and mounted on glass slides and tested. The value of bond strength obtained ranged from 4 to 7 MPa.

Thorpe et al. *(2)* laid a single fibre (Scotch Pine fibre, thermo mechanical pulp) at right angles on a shive of 4-6 fibres, sandwiched between glass microscope slides (previously treated to prevent adhesion of fibres to glass) and clamped at an approximate pressure of 1500 psi. The sandwiched structure was heated at 115°C in a pressure cooker and transferred for overnight treatment to an oven at 105°C. The fibre/shive crossing was then glued onto manila folder stocks and this assembly was used for further testing. The average bond strength measured was 6.9 MPa.

Schniewind et al. *(3)* took a different approach to the design of single fibre joints and mounted the fibres (white fir fibres from an experimental nitric acid pulp) on Teflon blocks and bonded them firmly with the help of heat and pressure. A drop of water was placed on the joint to prevent premature drying of the fibres. The measured bond strength values ranged from 2.4 to 4 MPa. Stratton and Colson *(4)* modified this assembly slightly by using Teflon-rubber discs and cementing the fibres (earlywood and latewood Loblolly pine, 47.5% yield) to the discs using special Mylar mount to reduce the high percentage of mountings lost in handling in the previous arrangement by Schniewind et al. *(3)*. The measured average bond strength values ranged from 1.5 to 4 MPa.

Torgnysdotter and Wagberg *(5)* modified the assembly further and used a dilute fibre suspension (Spruce wood, target Kappa numbers 110, 80 and 16) placed on a Teflon disc allowing the fibres to sink slowly and form a web. This is a better approach as it closely mimics the way a sheet is formed. Average bond strength value ranged from 28 to 32 MPa.

Russell et al. *(6)* focused more on the effect of wet strength additives and sheet strength (St.Regis beaten and unbeaten, unbleached bisulfite softwood pulp), using specially designed clamps. Handsheets were prepared using TAPPI T 205 and after standard drying cycle were oven dried at 120°C for ten minutes. They obtained average bond strength values of 2.6 ± 1 MPa for untreated samples and 2.4 ± 2

MPa and 2.5 ± 1.5 MPa for the samples containing melamine and polyamide resins respectively.

Button *(7)* came up with a novel method of treating fibres like joints using theories in mechanical engineering. He used Loblolly pine trachieds (58% yield) for his studies of bond strength. Fibres were sandwiched between three layers of membranes and placed under a pressure of 100 psi for 24 hours. Average bond strength values of 13.9 MPa and 8.9 MPa were obtained for the latewood and earlywood fibres respectively.

It can be seen that previous measurements have produced a wide range of estimates of shear bond strength. The researchers all used different approaches to design the joints and there are many difficulties with the designs. Most of the joints are prepared using heat and pressure which means that there is going to be a change in the fibre properties during joint preparation itself. The methods also fail to take into account the random nature of fibre stacking during sheet forming. Single fibre joint preparation and representivity of such bonds for a sheet as whole are major issues. For example, Russell et al *(6)* prepared 207 individual joints but just 52% which could be used for testing as the others were damaged during handling. It can be questioned whether so few bonds are representative of a sheet as a whole.

Another approach to the measurement of bond strength has been to try and measure the bond strength indirectly from measurements on the sheet. The first attempt in this area was the work of Nordman *(8)* whose approach was to use irreversible work done during straining of a paper sheet as a measure of bond strength. Bond strength was then given as the energy per unit area of bond failure.

Seth *(9)* modified Nordman's equation and obtained a bond strength value of 8 J/kg (spruce pulp beaten in PFI mill for 5000 revolutions).

Gurnagul et al. *(10)* applied Page's theory *(11)* for indirect sheet measurements and obtained a bond strength value of 3 MPa (black spruce unbleached Kraft pulp, 47.2% yield, Kappa number 30.5). This estimate cannot be considered fully reliable because it is based on Page's theory *(11)* which has never been completely verified.

The core idea of the new method for bond strength measurement presented here is to adjust the fibre strength independently of the bond strength by weakening the fibres. The fibres are weakened using acid vapour exposure for a series of times *(12)*. Sheets are exposed to acid vapours, tested for tensile strength and the exposure time at which all of the fibres appear broken, except those that are not bonded on one side of the fracture line as observed under a confocal microscope is determined. The optimum point is estimated using the confocal microscopy techniques described further. This point is where the fibre strength is equal to the bond strength of the most heavily loaded bond. The fibre strength is obtained at this point for sheets of varying grammages and extrapolated to calculate the shear bond strength.

The approach calculates bond strength from the sheets directly, thus eliminating the problem of forming representative individual bonds for testing as well as measuring.

EXPERIMENTAL METHOD

The moving belt sheet former *(13, 14)* (MBSF) was used for making handsheets from 60% yield, never dried Radiata Pine pulp. The pulp was beaten in a Valley beater for 12 minutes. The initial freeness of the pulp was 720 CSF before beating and 610 CSF after beating. After refining, a 0.2mm slotted screen for shives and 200 mesh screen for fines were used to remove the shives and the fines respectively

Sheets of three grammages 30 gsm, 45 gsm and 60 gsm, were made at 0.05% consistency. Zero span strength shows an upward trend as grammage is reduced and the true strength is obtained by extrapolating the measured values to 0 gsm *(15)*. The sheets were pressed on a dynamic roller press which uses two rollers operated with the help of pneumatic system. Two passes at 0 pressing pressure and two passes at 0.275 MPa were used. The sheets were restrain-dried and conditioned as per ISO 187:1990. Samples were cut and tested as per ISO 1924-2 for tensile strength measurements and ISO 15361 for the Dry Zero span tests.

A chamber was designed to provide uniform acid exposure to the paper sheets and to also have

more control over the acid exposure process. The rapid de-aeration ability of the chambers is one of the major advantages.

The acid exposure is composed of a two chamber assembly (Figure 1) made from plastic (PVC) was connected by a plastic valve. The chamber on the left holds the acid (HCl 32%) and the chamber on the right holds the paper sheet. The small metal protrusion from the right chamber is a pipe connected to the vacuum apparatus for starting a flow of acid gas from the left chamber into the right. During the rapid de-aeration stage the valve is closed and the right chamber is evacuated. Dimensions of each chamber is 220 mm x 220 mm x 40 mm. The papers were exposed to acid gas for times ranging from 50 to 130 minutes. The rapid de-aeration time is not included in the exposure times reported in this paper but was generally less than 5 minutes.

Figure 1: Acid Exposure Chamber Assembly

Confocal Laser Scanning Microscopy (confocal microscopy) was used to measure the fraction of broken fibres after the tensile tests were conducted on the acid exposed samples. Confocal Microscopy was the preferred method because it allowed us to scan into the interior of the sample to image all the fibres. An Optiscan F900e BH2 confocal microscope operating in fluorescence mode with a 20X oil immersion lens was used. After the sheets were acid exposed and tensile tested, they were dyed using a fluorescent acridine orange dye and then allowing them to air dry under a fume hood as per safety procedures. The samples were then put into a Petri dish filled with immersion oil and placed in a vacuum chamber. Vacuum was created in the chamber and the sample was kept there until gentle bubbling could be seen. Cessation of bubbling indicated that the oil had penetrated through the paper pores and impregnated the sample completely. These dyed and oil impregnated samples were examined under the confocal microscope.

For each level of acid exposure, 150 fibres crossing the fracture line were measured and assigned as either broken or pulled out. The fraction of broken fibres was then calculated. This was later used to determine the optimum exposure time. Image analysis was done using IMAGE PRO 5 software. Fibre width was measured from the images using the built in drawing and distance options for converting pixels into distance.

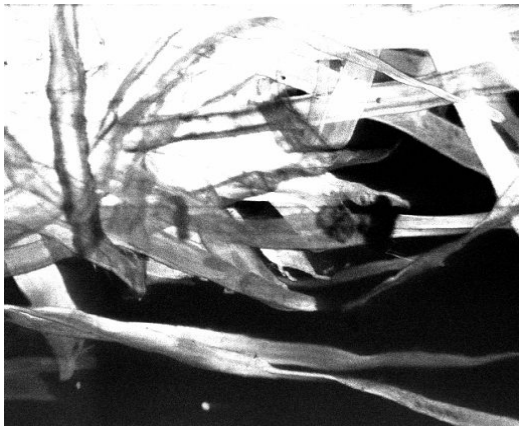

Figure 2: Pulled out fibres (Exposure time 0 minutes)

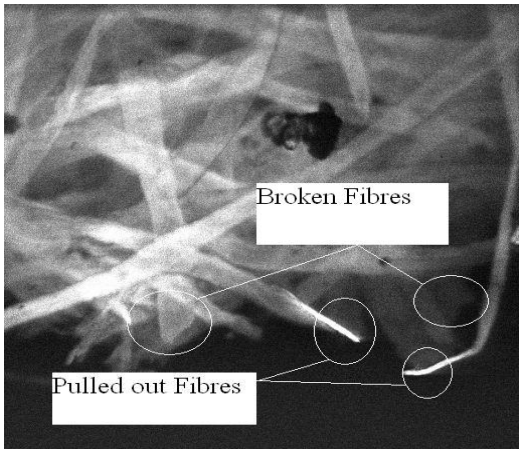

Figure 3: Pulled out fibres and Broken Fibres (Exposure time 105 minutes)

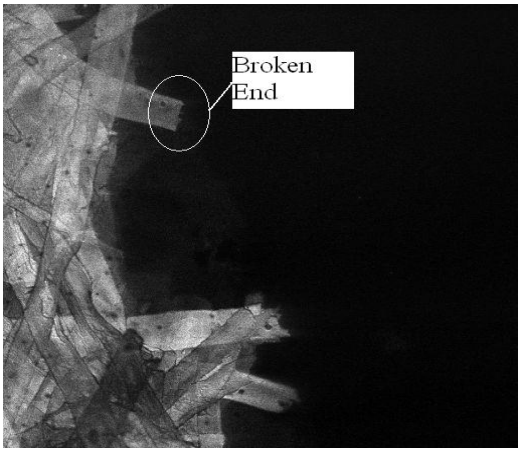

Figure 4: Broken Fibres (Exposure time 120 minutes)

Figure 2, Figure 3, Figure 4 show how the fibres appear when they are pulled out (0 and 105 minutes acid exposure) and when they are broken (105 and 120 minutes).

The Zero span strength measurements were done after the samples were exposed to acid vapours for the optimum time. The clamping pressure used to hold the sample during the zero span tests was 0.41 MPa (60 psi). This particular pressure was chosen because it is the clamping pressure at which the sample is held firmly without crushing the fibres. The clamping pressures of 0.48 MPa (70 psi) and 0.55 MPa (80 psi) were observed to slightly crush the fibres.

RESULTS

Figure 5 shows that the sheet tensile strength falls approximately linearly with acid exposure time.

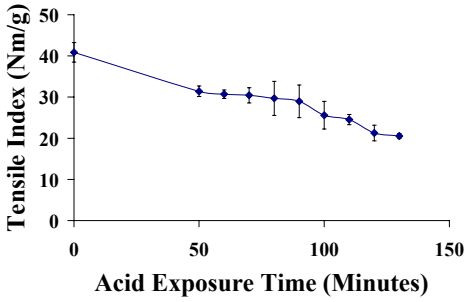

Figure 5: Tensile Index vs. Acid Exposure Time

Figure 6 shows the fraction of broken fibres crossing the fracture line as a function of acid exposure time. The fraction of broken fibres can be seen to increase with increasing acid exposure until it reaches a plateau of 0.90. The fraction of broken fibres never reaches 100% because there are always a small fraction of fibres that are bonded only on one side of the fracture line. The optimum point where the fraction of pulled out fibres first reaches the plateau value was determined by fitting straight lines to the data from 80 to 100 minutes exposure and to the data in the plateau region and determining the intersection point.

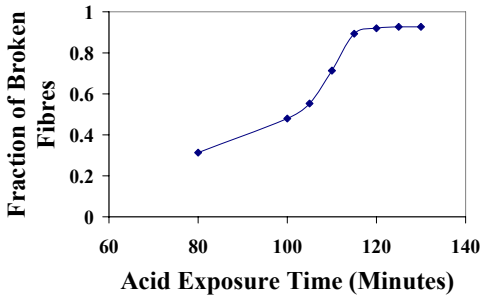

Figure 6: Fraction of Broken fibres vs. Acid Exposure Time

For the data shown in Figure 6, the optimum point was found to be 115 minutes.

The Zero span strength values obtained for the varying grammages sheets at the optimum exposure time of 115 minutes can be seen in Figure 7.

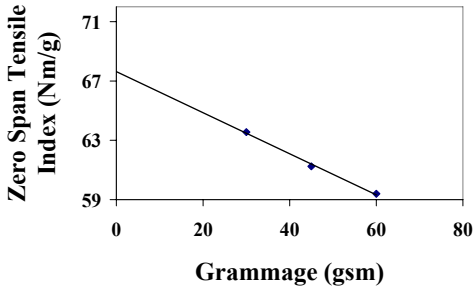

Figure 7: Zero Span Tensile Index vs. Grammages

As expected we see a slight increase in the values of zero span strength as the grammages decrease.

It has been argued that the shear bond strength at zero grammages can be calculated by extrapolating this graph in the direction of the y-axis *(15)*. The value of the zero span strength obtained from the extrapolated graph (Figure 7)

is 67.6 MPa. This value of the zero span strength can then be used to calculate the shear bond strength.

The shear bond strength is given as:

$$\sigma_s = \frac{\sigma \times C_s}{A} \qquad \textbf{.....Equation 1}$$

Where,
σ_s = Shear Bond strength
σ = Fibre strength
C_s = Cross sectional area of one fibre wall
A = Area of the bond

The fibre strength is calculated as:

$$\sigma = \frac{F_{total}}{A_{cs}} \qquad \textbf{.....Equation 2}$$

Where,
F_{total} = Force on the sample
A_{cs} = Total cross sectional area of all fibres

The breaking force on the sample in the zero span test is given by:

$$F_{total} = F \times w \qquad \textbf{.....Equation 3}$$

Where,
F = The breaking force for a random sheet per unit width
w = width of the sample sheet

Van den Akker *(16)* found that the zero span breaking force for a random sheet is 3/8 the zero span breaking force for a sheet in which all the fibres are oriented in the direction of the stress.

$$\therefore \frac{3}{8} F = F_o \qquad \textbf{.....Equation 4}$$

Where,
F_o = breaking force for an oriented sheet

There fore the breaking force for a random sheet is given by:

$$F = \frac{8}{3} \times Z \times M \qquad \textbf{.....Equation 5}$$

Where,
Z = Zero span tensile index
M = Mass per unit area of the sheet (i.e.: grammage) assumed to be 1500 kg/m^3

The mass per unit are of the sheet, i.e.: grammage is given by:

$$M = \rho \times t \qquad \textbf{.....Equation 6}$$

Where,
ρ = Fibre wall density
t = thickness of sheet if it was compressed to a solid mass of fibres

Multiplying both sides of **Equation 6** by **w**, we get:

$$w \times M = \rho \times t \times w \qquad \textbf{.....Equation 7}$$

The cross sectional area of fibre wall material in a sheet of paper is

$$A_{cs} = t \times w \qquad \textbf{.....Equation 8}$$

$$\therefore A_{cs} = w \times \frac{M}{\rho} \qquad \textbf{.....Equation 9}$$

Equations 2, 3, 5 and **9** can be used to calculate the fibre strength as:

$$\sigma = \frac{8}{3} \times Z \times \rho \qquad \textbf{.....Equation 10}$$

It is worth noting that using the assumed fibre wall density of 1500 kg/m^3, **Equation 10** simplifies to:

$$\sigma \textbf{ (MPa)} = 4Z \textbf{ (kNm/kg)} \qquad \textbf{.....Equation 11}$$

The area of one bond is given by:

$$A = R \times \frac{D_w^2}{\sin 2\theta_{avg}} \qquad \textbf{.....Equation 12}$$

Where,
A = Area of one bond
R = Correction factor to account for the spreading of the fibre when it is bonded; found to be 1.35 *(17)*

θ_{avg} = Average crossing angle of fibres, which has been derived to be 32.7° *(17)*
D_w = Width of the fibre (40 microns)

Substituting **Equation 10, 11**, **12** and the values of θ_{avg} and **R** into **Equation 1** we get the complete expression for the shear bond strength as:

$$\sigma_s = \frac{\frac{8}{3} \times Z \times \rho \times C_s}{R \frac{D_w^2}{\sin 2\theta_{avg}}} \qquad \text{.....Equation 13}$$

This simplifies to

$$\sigma_s \text{ (MPa)} = \frac{4Z \times C_s \times \frac{2}{3}}{D_w^2} \qquad \text{.....Equation 14}$$

as $\frac{R}{\sin 2\theta_{avg}} = \frac{3}{2}$

$$\therefore \sigma_s \text{ (MPa)} = \frac{8Z \times C_s}{3D_w^2} \qquad \text{.....Equation 15}$$

The average tensile index obtained from the zero span test was 67.63Nm/g after an optimum exposure time of 120 minutes and when extrapolated back to zero gsm. When all the other variables were substituted into **Equation 13,** we obtained the average bond strength of 26.9 ± 0.5 MPa.

DISCUSSION

The value of the bond strength seems significantly higher than the published previous measurements. The only similar values are those by Torgnysdotter and Wagberg *(5)*, ranged from 22 MPa to 32 MPa.

The methods used previously didn't concentrate on testing the bond strength in the sheet itself and most of them focused on designing single fibre bonds which would be representative of the whole sheet. Unfortunately no number of single fibre bonds can correctly represent what happens in a sheet. Also the testing procedures become complicated and there are significant difficulties with joint creation, positioning etc. Most of the previous test methods also use heat and pressure as a means of bond formation and this changes the nature of the joint and affects the properties of fibres in individual joints. Glues and adhesives also do not help much because they may interfere with the actual bonding process in itself and subsequently in the testing procedures. The same criticism may apply to this new method as well because the zero span strength and other parameters are also averages.

The new method does have some potential weaknesses. One of the questions arising is what if the bonds are weakened due to HCl exposure? HCl exposure seems to cause polymer chain scission and no reports have been found which indicate that HCl affects surface bonding. Hence we assume that the HCl doesn't affect the bonds. In addition, the bonded areas of the paper are likely to be amongst the least accessible areas, thus least prone to being affected by HCl. However if we assume that HCl does affect the bonds then the measured value would be an underestimate and thus even further away from most previously measured values.

Paper is a variable structure due to the non uniform nature of the raw material and the non uniform pulping process. One may question the utility of a technique which gives a single value of bond strength. Mechanical properties of paper are directly related to the bond strength and the Shear bond strength was the last unmeasured variable required to validate and develop models and theories for paper mechanical properties. Shear bond strength has been measured in terms of in-plane tensile strength and zero span tensile strength as a network parameter. Other factors that influence tensile strength are formation, pressing pressure, types of fibre used, pulping techniques, quantity of fines etc. The experiments performed here illustrate that if all the other variables are known then this new technique can be used to measure the shear bond strength directly. The value of shear bond strength can be plugged into the different models to predict the tensile and other mechanical properties of paper. Experiments have also been conducted to measure the shear bond strength of recycled fibres and heat treated fibres, but those results will be presented in later work.

CONCLUSION

The average shear bond strength value of sheets made of never dried unbleached Radiata pine

fibres (60% yield) is 26.9 ± 0.5 MPa. This value is much higher than most previously published values and takes into account the entire fibre network instead of just single fibre joints.

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