

# Specific Contact Area and the Tensile Strength of Paper

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## Summary

The Page equation for tensile strength of paper is widely and successfully used but has the disadvantage of having a number of variables that cannot easily be estimated or measured. Whilst it is possible to separate tensile strength into fibre strength and bonding strength components, the problem of whether a change in strength is due to increased bonded area or increased bond strength is still difficult to address. This paper suggests a variable change in the Page equation allowing existing measurement methods to be used effectively to separate these effects. A series of experiments using standard papermaking treatments to influence tensile strength are described. Data from these experiments are used to illustrate how the change made to the Page equation can be applied to provide insights into the mechanism of strength development.

**Keywords:** Tensile theory, specific contact area, relative bonded area, bond strength, fibre strength.

## Background and Introduction

Of the many theories relating the tensile strength of paper to sheet structure and fibre properties, perhaps the most used and most useful is that suggested by Page in 1969 [1]. Page showed that the tensile strength of paper could be expressed in a simple and effective way by the equation:

$$\frac{1}{T} = \frac{9}{8Z} + \frac{12A\rho}{bP\lambda RBA} \quad (1)$$

where  $T$  and  $Z$  are the long- and zero-span tensile indices respectively ( $N m kg^{-1}$ );  $A$  is the fibre cross-sectional area ( $m^2$ );  $\rho$  is fibre density ( $kg m^{-3}$ );  $b$  is the shear bond strength per unit area ( $N m^{-2}$ );  $P$  is fibre perimeter ( $m$ );  $\lambda$  is the mean fibre length ( $m$ ), and  $RBA$  is the relative bonded area. The first and second terms on the right hand side of Equation (1) represent the contributions of fibre strength and bond strength to the overall tensile strength of the paper sample, without knowledge of any viscoelastic or de-bonding effects which may occur during straining prior to failure. It is frequently convenient to define a *Bonding Index*,  $B$  ( $N m kg^{-1}$ ) as,

$$B = \frac{bP\lambda RBA}{12A\rho} \quad (2)$$

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which represents the contribution of bonding to tensile strength in units of tensile index and,

$$\frac{1}{T} = \frac{9}{8Z} + \frac{1}{B}. \quad (3)$$

Most of the variables in Equation (2) represent measurable fibre properties; however, as a consequence of their distributed nature, precise definition of appropriate mean values suitable for substitution requires care. The variables  $b$  and  $RBA$  represent the shear bond strength per unit area and the fraction of total fibre surface area for which bonds are present, respectively; these are properties of the sheet, not of the fibres, and consequently are considerably more difficult to determine experimentally.

Indirect estimates of relative bonded area are possible by considering the degree of scattering as a measure of the extent of bonding [2] and attempts to measure bonded area directly using nitrogen adsorption techniques have been investigated [3,4]; for discussion of the limitations of such techniques and mathematical expressions to estimate the structural characteristic analogous to RBA see [5]. A method suggested by Clarke [6] has been used to investigate bonding in a number of investigations in Manchester [7-10] and elsewhere [11,12]. This "Contact Ratio" method involves producing a thin layer of dyed fibres on a filter paper using a British Standard Sheet Former and pressing these fibres into contact with glass microscope slides. The dyed fibres are illuminated by vertical incident illumination such that areas of optical contact appear darker than non-contacting areas in the resultant image. This contact area is measured by image analysis and divided by the total projected area of fibre yielding the contact ratio,  $R$ . The technique has become more accessible with the availability of high-resolution desktop scanners and the use of these is discussed in [13]. The attraction of the contact ratio method is that it has potential to be used, along with other pulp evaluation techniques, to predict the properties of paper to be made from a particular market pulp or source of recycled fibre. However, the precise relationship between contact ratio and the parameters  $b$  and  $RBA$  has not been clear, there being some evidence [10] that contact ratio was a measure of both  $b$  and  $RBA$  and other evidence [9] suggesting that contact ratio gave a measure of  $RBA$  only.

Strength, optical and surface roughness properties of paper are all recognised to be affected by the coarseness of the fibre furnish used in their manufacture. This is often defined, following established textile industry practice, as the mass per unit length of the fibre. An alternative to this, known as *Clarke coarseness*, has been suggested by Clarke *et al.* [13], who used the mass per unit projected area of fibre as measured by image analysis. The measured quantity is equivalent to the ratio of fibre coarseness and width and may therefore be considered a measure of fibre grammage. This method uses the most straightforward of image analysis measurements and gives a similarly useful measure of the effect of single fibre mass on sheet properties.

Here we propose the introduction of a new variable, *specific contact area*,  $\alpha$  ( $m^2 kg^{-1}$ ), which simplifies the Page equation, removing a number of variables which are difficult to measure. We show that it is possible to estimate the value of  $\alpha$  from measurements of *contact ratio* and *Clarke coarseness*, described above. We have carried out an experimental programme to generate data against which this new form of the Page equation has been compared and demonstrate that the treatment given here provides a technique to decouple the effects of bond strength and RBA when investigating changes in the tensile strength of paper.

## Theory

We define the *specific contact area* as the bonded fraction of fibre surface area per unit fibre mass. It is given by,

$$\begin{aligned}\alpha &= \frac{P\lambda}{A\lambda\rho} RBA \quad , \\ &= \frac{PRBA}{A\rho} \quad .\end{aligned}\tag{4}$$

It is intuitively reasonable to expect the specific contact area to influence tensile strength of a sheet of paper since it takes into account the degree of bonding which occurs between fibres and, through the mass of a fibre, the number of fibres in that sheet with potential to bear load.

On manipulation, substitution of Equation (4) into the Equation (1) yields,

$$\frac{1}{T} = \frac{9}{8Z} + \frac{12}{b\lambda\alpha} \quad .\tag{5}$$

In this form the number of variables in the bonding term has been reduced from six to three, considerably simplifying the Page Equation.

Now, the specific contact area,  $\alpha$  is a property of sheets and the contact ratio,  $R$  is a property of networks formed on slides. If the pressure used to transfer fibres onto slides is the same as that used to produce handsheets from those fibres, then we expect the *contact ratio*,  $R$  to provide an estimate of the fractional area of a given fibre in a handsheet that is available to form bonds with fibres above or below it in the  $z$ -direction. Dividing the contact ratio by the Clarke Coarseness therefore provides the fraction of the projected fibre area available for bonding per unit fibre grammage and it is intuitively reasonable to expect this to provide a sensible estimate of the specific contact area,  $\alpha$  such that,

$$\alpha = \frac{R}{C} \quad .\tag{6}$$

Substitution of Equation (6) into Equation (5), gives,

$$\frac{1}{T} = \frac{9}{8Z} + \frac{12C}{b\lambda R} \quad ,\tag{7}$$

and this allows the bonding index to be written as,

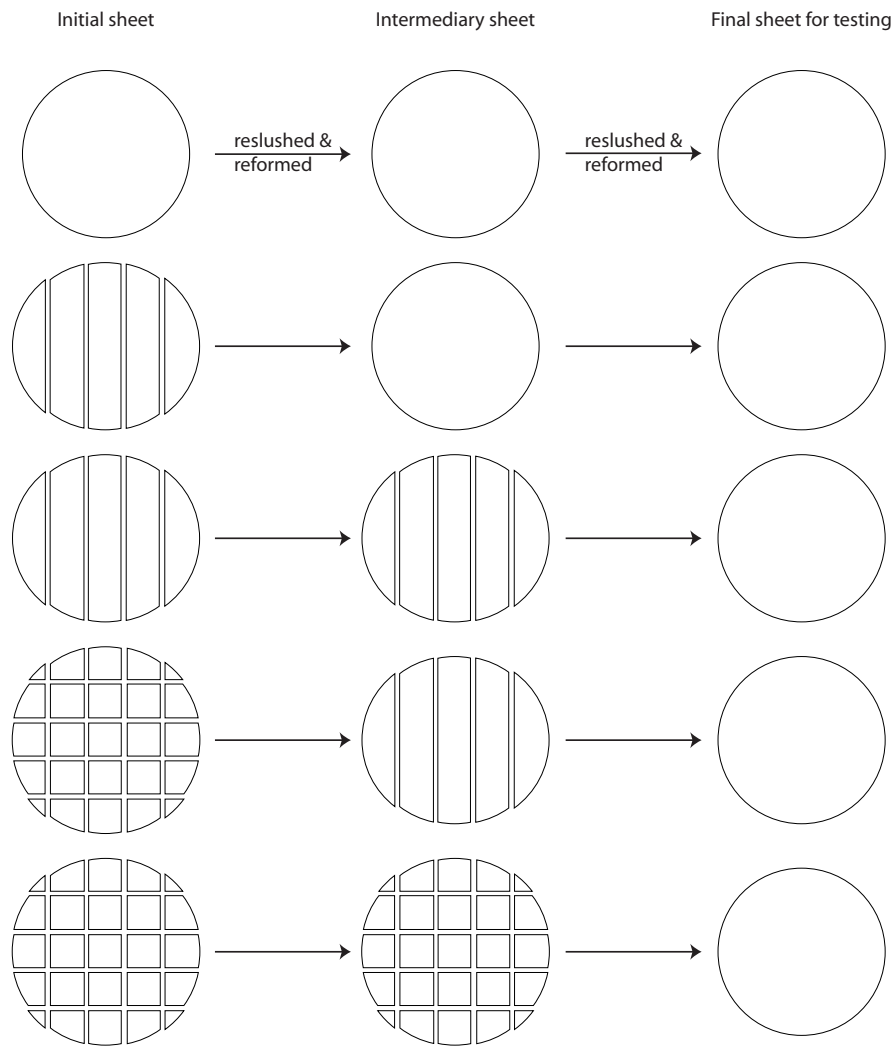
$$B = \frac{b \lambda R}{12 C} \quad (8)$$

## Experimental

A programme of experimental work was planned that allowed tensile strength to be varied in a number of ways. All work was carried out using  $60 \text{ g m}^{-2}$  handsheets formed according to standard ISO 5269-1:2001 using a British Standard Sheet Former from Intercontinental, a commercial unbleached Kraft pine pulp. Handsheets were formed from the unbeaten fibre and from fibre beaten for 56000 and 72000 revolutions using the PFI Mill (ISO 5264-2:2002) such that they had Schopper-Riegler wetness of  $41^\circ\text{SR}$  and  $63^\circ\text{SR}$  respectively. All subsequent treatments were carried out on pulps beaten for 56000 revolutions with the following changes in conditions:

- **Fibre length** was varied by cutting handsheets using a guillotine after couching but before pressing. After cutting handsheets were reslushed in a standard disintegrator for 10,000 revolutions and a new set of handsheets formed. To ensure that each set of handsheets had comparable inter-fibre bonding, all sheets were made from fibre that had been reslushed twice; the forming, cutting and reslushing sequence used to achieve this is represented schematically in Figure 1.
- **Wet Pressing** was carried out at gauge pressures of 0.35, 0.70, 1.41, 2.11 and 2.82 *MPa* (50, 100, 200, 300 and 400 *psi*) using a modified handsheet press.
- **Bonding** was influenced chemically by the addition of a low molecular weight cationic polyacrylamide (DSR 1256 from Ciba Speciality Chemicals) as a dry strength aid and a domestic fabric conditioner consisting of a blend of surfactants as a debonding agent. These were added to thick stock prior to dilution for sheet forming at addition rates of 0.01 %, 0.05 %, 0.1 %, 0.5 % and 1 % w/w active compound on fibre.

For each condition, the fibre length distribution and fines content were measured using a Kajaani FS-200 fibre length analyser and contact networks of fibres were prepared on clean  $5 \text{ cm} \times 5 \text{ cm}$  glass microscope slides following the technique described by Clarke *et al.* [13]. The networks had target grammage around  $0.5 \text{ g m}^{-2}$  and fibres were stained with Chlorazol black. In each case, the four slides were made and networks were pressed at the same pressure as the corresponding set of handsheets and these pressures were applied for 30 *s*. The contact ratio of the fibres on the slides was measured following the technique described by I'Anson *et al.* [13] using an Agfa DuoScan T2500 desktop scanner to capture reflection and transmission images of the networks and code written using the MatLab Image Processing Toolbox for image processing allowing calculation of the Contact Ratio, *R*. After image analysis, slides were conditioned for 24 hours in a standard atmosphere and weighed ten times using a 5-figure electronic balance. After removing all the fibre, each slide was re-weighed ten times, which allowed the mass of fibre on each slide to be determined. Given the mass of fibre on each



**Figure 1.** Cutting and reslushing scheme to reduce fibre length in handsheets.

slide and the projected area of fibre on each slide, the Clarke Coarseness,  $C$  was calculated for each sample.

Handsheets were conditioned for 24 hours in a standard atmosphere prior to testing for grammage, thickness, tensile strength (ISO 5270:1999), zero-span tensile strength (ISO 15361:2000), and light scattering coefficient (Tappi T 1214 *sp*-02).

## Results and Discussion

Data arising from the physical testing of handsheets is summarised in Table I; the value of mean fibre length reported in this table is the length weighted average and the column headings  $\rho$  and  $s$  represent the apparent density and specific light scattering coefficients respectively.

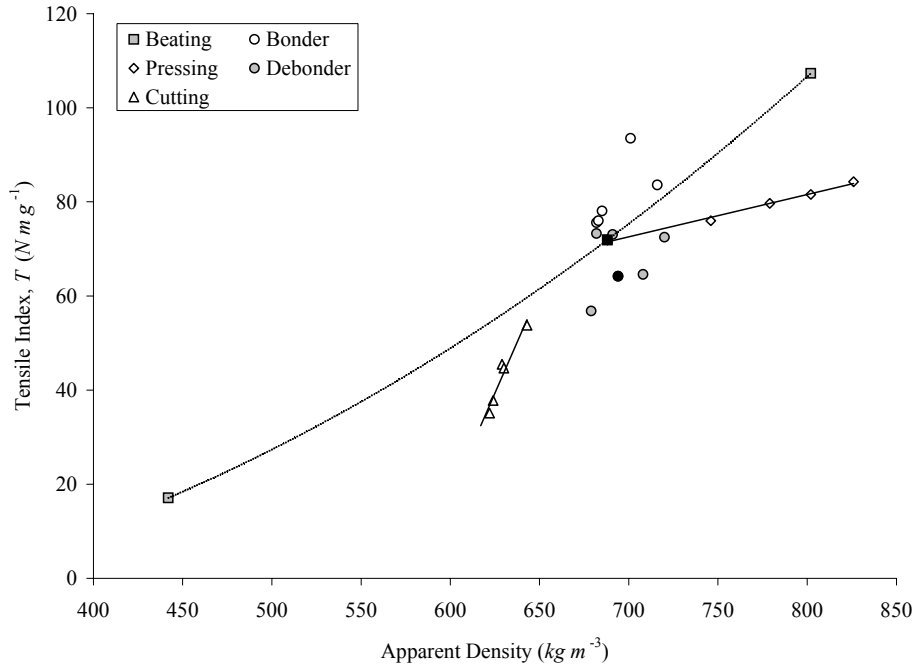
The influence of the various experimental treatments studied upon the tensile strength of the sheet is summarised in Figure 2, which shows Tensile Index plotted against apparent density. In this and all

			Fibre Properties			Sheet properties				
			°SR	<i>C</i>	<i>R</i>	$\lambda$	$\rho$	<i>T</i>	<i>Z</i>	<i>s</i>
				$g\ m^{-2}$		$mm$	$kg\ m^{-3}$	$N\ m\ g^{-1}$	$N\ m\ g^{-1}$	$m^2\ kg^{-1}$
Beating	Unbeaten		14	5.40	0.097	2.52	442	17.1	186	32.1
	56000	revs*	41	5.45	0.370	2.50	688	71.9	190	21.7
	72000	revs	63	5.63	0.585	2.37	802	107.3	209	13.8
Pressing	100	psi	43	5.56	0.435	2.44	746	76.0	197	19.9
	200	psi	43	4.87	0.498	2.49	779	79.7	196	18.4
	300	psi	43	5.15	0.468	2.48	802	81.6	194	17.6
	400	psi	43	5.35	0.542	2.45	826	84.3	189	15.9
Cutting	Uncut		39	5.85	0.387	2.50	643	53.8	181	24.5
	1	cut		5.30	0.403	2.14	629	45.5	164	25.0
	2	cuts		5.91	0.409	2.09	630	44.7	159	25.2
	3	cuts		5.97	0.405	1.62	622	35.1	160	25.6
	4	cuts		5.62	0.405	1.70	624	37.8	160	26.1
Dry Strength	0	%*	43	5.61	0.376	2.53	694	64.2	183	21.9
	0.01	%		5.39	0.410		682	75.6	188	21.3
	0.05	%		6.03	0.346		683	76.0	188	20.6
	0.1	%		5.95	0.372		685	78.1	184	20.5
	0.5	%		5.75	0.366		716	83.6	190	18.4
	1.0	%		6.07	0.405		701	93.5	187	18.4
Debonder	0.01	%	43	5.66	0.334	2.47	691	73.1	178	21.3
	0.05	%		5.72	0.331		682	73.3	190	21.4
	0.1	%		5.97	0.302		720	72.5	175	21.2
	0.5	%		5.85	0.268		708	64.6	174	21.8
	1.0	%		5.48	0.298		679	56.8	180	24.5

**Table I.** Fibre and handsheet data for all conditions. Conditions identified with an asterisk are nominally identical.

subsequent figures, data represented by solid black markers are those marked with an asterisk in Table I and represent control conditions against which other data with the same shape marker should be compared.

The relationships observed in Figure 2 are broadly in line with expectation. Beating and pressing have increased the density and strength of sheets formed from this once-dried fibre. The dry strength aid and the debonding agent have had minimal effect on density and the dry strength aid has had the expected effect; the debonding agent has only decreased strength at the highest addition rate used and has, in fact, increased strength at lower addition rates. We observe also that systematic reduction of fibre length has resulted in a corresponding decrease in Tensile Index; it is important to note however

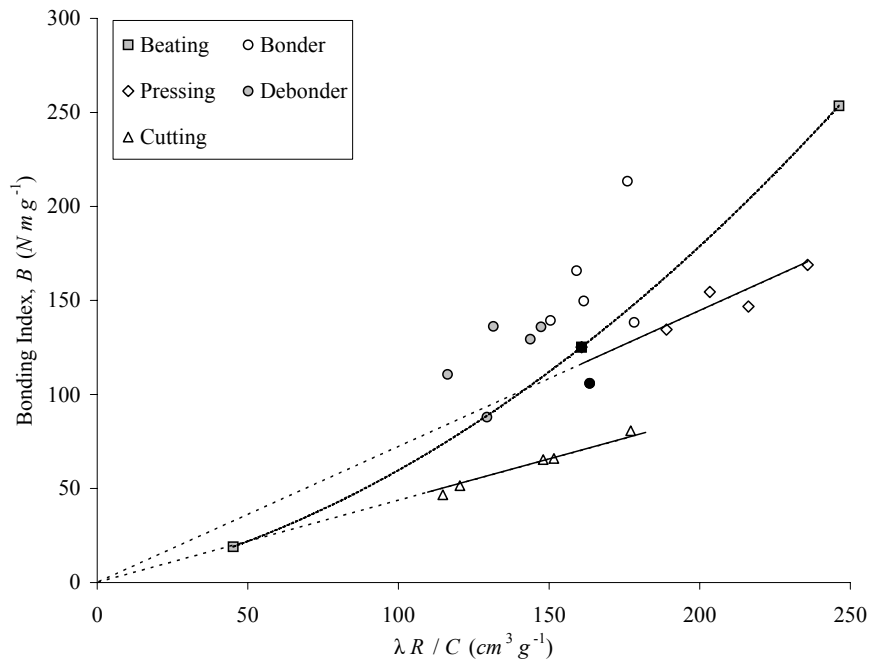


**Figure 2.** Tensile Index plotted against apparent density.

that the cutting procedure used has introduced a significant shift in density and hence tensile strength. We bear in mind that the average fibre length of the uncut sample is nominally identical to that of the middle point of the beating data and the first point of the pressing data; the experimental conditions for the preparation of this sample differ only in the repeated forming and reslushing. We expect any direct influence of fibre length on sheet density to be weak and this is consistent with the range of densities for the samples formed in the cutting experiment being within around 3 % of their mean. This suggests that the delay between cutting and reslushing has been sufficient to influence inter-fibre bonding, and hence density (or *vice versa*), to some extent. In what follows we shall see that the theoretical treatment given earlier allows us to obtain some insights into the nature of this change.

In order to test Equation (7) we sought to plot the contribution of bonding to tensile index,  $B$  against the product of specific contact area and mean fibre length, *i.e.*  $(\lambda R/C)$ ; from Equation (8) we expect this to yield a line with gradient  $b/12$  such that if the shear bond strength per unit area,  $b$  is unaffected by a given treatment then the line will be straight and otherwise we will observe a non-linear relationship where the non-linearity indicates the nature of the change in  $b$ .

For some of the parameters measured, there should be no effect of the treatment applied and a better estimate of the value of this parameter is obtained by averaging across the groups. Thus, for all conditions involving fibre beaten for 56000 *revs* in the PFI mill, the average value of Clarke coarseness for these data was used to calculate the value of  $(\lambda R/C)$ . For the experiments involving pressing and the addition of dry strength aid or debonding agent, an average fibre length was

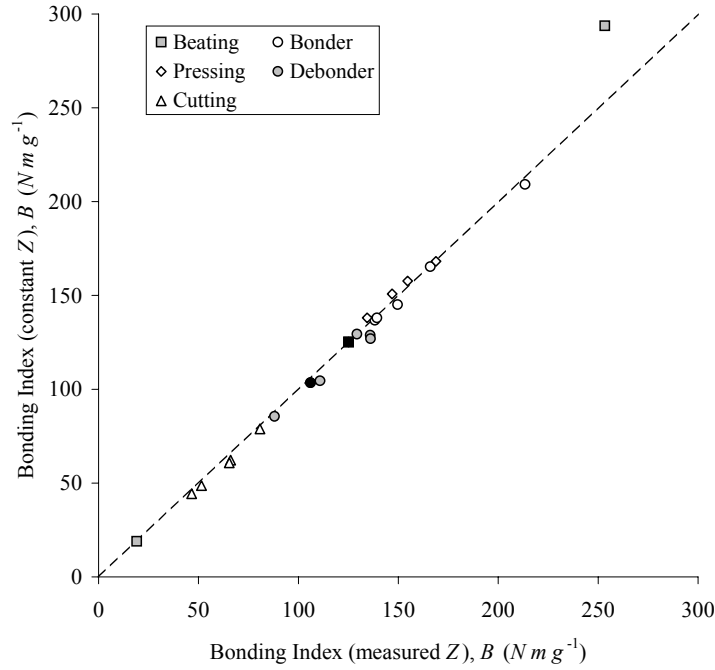


**Figure 3.** Bonding Index plotted against the product of specific contact area and fibre length.

calculated from the data across these groups and for the cutting experiments, the average contact ratio,  $R$  was calculated from the data within the group. Figure 3 shows the bonding Index,  $B$  plotted against the parameter  $(\lambda R/C)$  calculated in this way. We observe that for the pressing and cutting experiments the data yield linear relationships that pass through the origin; a small increase in gradient is observed for the points arising from the beating experiment and the influence of the dry strength aid and the debonder is seemingly less systematic. We shall discuss these families of data in turn in due course; before doing this, however, and in order that the observed differences can be discussed with confidence, it is worth considering the effect of the measurement of zero-span tensile strength on the calculated values of the Bonding Index,  $B$ .

In its simplest form, the Page Equation can be considered as a series model of sheet strength with the contribution of fibre strength to the tensile strength of the sheet being inferred from the zero-span tensile strength. Inspection of the data in Table I reveals some sensitivity of the zero-span Tensile Index to the treatments applied. For example, there is a weak systematic decrease in the zero-span tensile index as the degree of wet pressing increases and over the first two cutting operations; similarly there is some increase in zero-span tensile index with beating and some variation within the data for the networks treated with dry strength aid or debonder. Now, there is no reason to expect these treatments to have any significant effect on fibre strength, yet it is clear that bonding and density effects are influencing the zero-span tensile index; this is not a new observation but in the context of this study, it means that we should quantify the influence of variations in the measurement of zero span upon the calculated values of bonding index. To achieve this, we calculated  $B$  from the measured



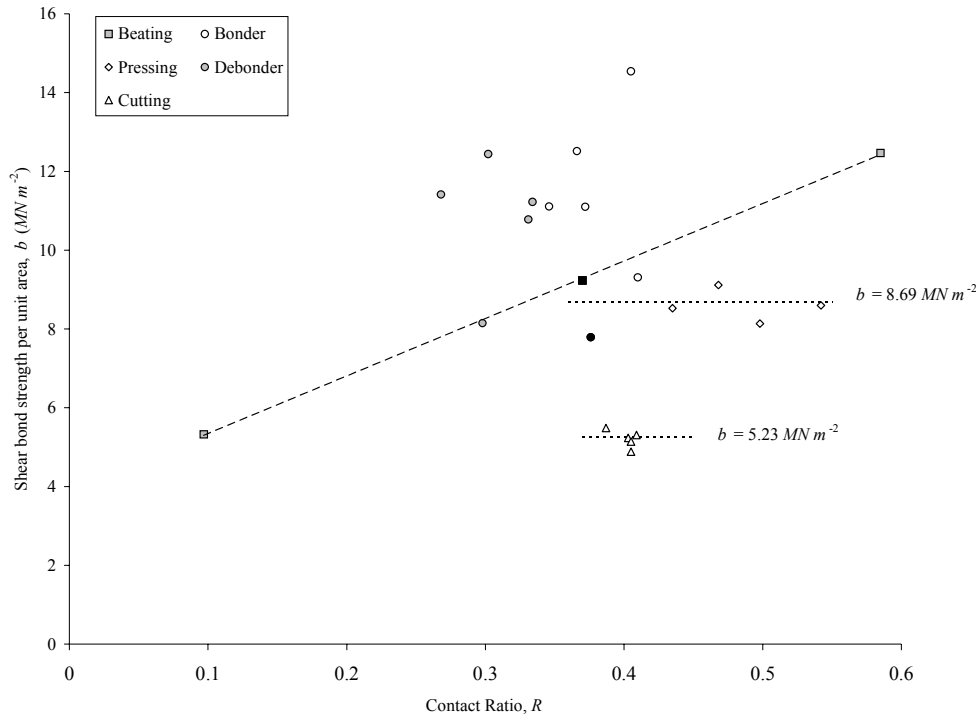


**Figure 4.** Comparison of Bonding Index calculated using a single value of  $Z$  against that calculated using the value of  $Z$  measured for each sample.

value of tensile index,  $T$  for each condition and the zero-span tensile index for the standard handsheets formed from the pulp beaten to 41°SR in the beating experiment, *i.e.*  $Z = 190 \text{ Nm g}^{-1}$ . The value of  $B$  calculated in this way is compared with that calculated using the value of  $Z$  measured for each sample in Figure 4 where the broken line has unit gradient. The only significant difference in the two estimates of  $B$  is observed for the sample beaten to 63°SR such that when Figure 2 is replotted using the estimate of  $B$  calculated using constant  $Z$ , the observed trends and clusters are not affected significantly. Accordingly, in what follows, we base our discussion and subsequent calculations on the estimates of  $B$  plotted in Figure 3.

The data in Figure 3 show linear relationships between  $B$  and  $(\lambda R/C)$  for the pressing and cutting experiments. For the cutting experiment, the observed range of  $(\lambda R/C)$  is attributable entirely to the change in fibre length; for the pressing experiment however, it is attributable entirely to the change in contact ratio. This is important because the fact that this linear relationship passes through the origin tells us that there is proportionality between our measurement of contact ratio and the RBA of fibres in the sheet. As such, any estimate of the shear bond strength per unit area,  $b$  based upon the measured value of contact ratio will itself be proportional to the real value of  $b$  in the sheet.

Linear regressions on the data shown in Figure 3 for pressing and cutting yielded coefficients of determination of 0.800 and 0.965 respectively. From the gradients of these regressions, we calculate the shear bond strength per unit area,  $b$  as being  $8.69 \text{ MN m}^{-2}$  for the pressed sheets and  $5.23 \text{ MN m}^{-2}$



**Figure 5.** Shear bond strength per unit area plotted against contact ratio

for the sheets formed from cut fibres. Whilst these gradients provide a good estimate of the shear bond strength per unit area,  $b$  for these two families of data, it is possible to calculate  $b$  also from the values of  $T$  and  $Z$  measured for each experimental condition. These data are plotted against Contact Ratio,  $R$  in Figure 5; the horizontal broken lines represent the values of shear bond strength per unit area determined from the regressions shown in Figure 3. Since we consider Contact Ratio to provide an estimate of RBA, the plot allows identification of whether a given treatment has influenced bond strength, bonded area, both or neither. Thus in Figure 5 we see that pressing increases the contact ratio of fibres on the slides without influencing the shear bond strength per unit area of fibres in the sheet suggesting that pressing increases tensile strength by increasing relative bonded area, but has no appreciable effect on the strength of bonds over these areas. Whilst factors such as removal of curl and latency will influence the tensile strength of the unbeaten sample in a manner not readily detectable by the treatment given here, we may state that beating has increased both the strength and the area of bonds. Also, after the initial reduction in shear bond strength per unit area arising from the cutting procedure, the treatment shows, as expected, that reducing fibre length has no influence on either bonded area or bond strength. This initial reduction was unexpected and it may be that the repeated reslushing and forming required to prepare these fibres has resulted in some removal of fines and a general deterioration of the surface condition of the fibres in a manner that has influence sheet density and hence light scattering coefficient but not contact ratio.

Whilst it is clear that the dry strength aid has increased bond strength without affecting bonded area, the action of the debonding agent is a little more complex. With only a small amount of agent added, the bonded area has been reduced but the strength of bonds increases for all but the case

with 1 % w/w addition. This point lies close to the line through the data for the beating experiment and is arguably the same as that for the sample with no chemical treatment. The chemistry behind this effect does not concern us here, though we speculate that this may be due to charge effects at fibre surfaces. What is important though is that the theoretical treatment given here provides a relatively simple mechanism for identification of such effects where, in its traditional form, the Page Equation does not.

## Conclusions

We have suggested a simplifying variable change for the Page equation describing tensile strength. This change improves the usefulness of the equation because it reduces the second term to 3 variables, 2 of which can be estimated experimentally, allowing calculation of the third. This means that it is possible to separate the contribution to tensile strength of the area of bonds between fibres from the shear strength of the bonds.

A family of experiments has been presented to test the validity of the approach and the data arising from these have been used to identify changes in the shear bond strength per unit area arising from the treatments applied.

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