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An analytical model for number of fibre–fibre contacts in paper and expressions for relative bonded area (RBA)

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Abstract This paper proposes an analytical model for calculating the number of fibre–fibre contacts per unit fibre length from the cross-sectional dimensions of the fibres in a sheet and sheet density. This model has been verified with data of the number of fibre–fibre contacts measured directly in handsheets. The measured fibre–fibre contacts in the handsheets were classified into full and partial contacts. The model best fits this data when 1.5 partial contacts are equated to one full contact. A plot of measured versus predicted equivalent full contacts produced a linear correlation with a slope of 0.99 and correlation coefficient of $R^2 = 0.93$.

The model was used to derive expressions for the fraction of the available fibre surface, which is bonded to other fibres, a quantity called the Relative Bonded Area (RBA). The validity of the model was checked using experimental data for RBA. RBA was determined both by nitrogen adsorption (RBA_{N_2}) and by scattering coefficient (RBA_{sc}). The extrapolation method of Ingmanson and Thode to determine S_0, the scattering coefficient of an unbonded sheet, proved to be inaccurate. We estimated S_0 for some samples from their linear relationship between scattering coefficient and nitrogen adsorption. The new model accurately predicted both RBA_{N_2} and RBA_{sc}.

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Introduction

Fibre–fibre contacts have long been an interesting topic in the study of paper physics with numerous attempts to develop analytical models for the number of fibre–fibre contacts. Corte and Kallmes [1] presented the first important model for the number of fibre crossings in a three dimensional random fibre network. An expression equivalent to this model was proposed some years later by Komori and Makishima [2]. Komori and Makishima's model was later modified by Pan [3] by allowing for the reduction in free fibre length due to existing contacts. Pan's work was criticized and corrected by Komori and Itoh [4]. More recently, a similar model was also derived independently by Dodson [5]. These models use statistical analysis to predict the possible number of fibre–fibre contacts that a certain number of fibres could make in a given volume. They ignore the effects of fibre cross-sectional properties on the fibre–fibre contacts, so cannot model the effects of fibre collapse. However, the fibre width, fibre height and the degree of fibre collapse all affect the fibre–fibre contacts and it is necessary to include these parameters in any model of fibre–fibre contacts.

Because the measurement of the number of fibre–fibre contacts is usually very difficult, it has previously been difficult to obtain effective data to verify models for the number of fibre–fibre contacts. In this paper, we first present a new analytical model for the number of fibre–fibre contacts, which relates the cross-sectional properties of the fibres in the sheet to the number of fibre–fibre contacts per unit length of fibre. This model is then verified with data of the number of fibre–fibre contacts that had been previously measured directly in handsheets using a new combined technique of resin embedding and confocal microscopy [6]. The model is further used to derive expressions for the Relative Bonded Area (RBA). RBA is defined as the fraction of available fibre surface area that is bonded to other fibres and is given by the expression RBA = $(A_t - A_u)/A_t$, where A_t is the total fibre surface area available bonding and A_u is the total unbonded fibre surface area. RBA is one of the most important parameters determining the paper mechanical properties [7]. It is possible to calculate RBA directly from the area measured using nitrogen adsorption isotherms or indirectly by measuring light scattering coefficient, S, and by assuming that $S \propto A$. For this paper, both methods were used to measure RBA and the measured values will be compared with those predicted from the model for the number of fibre–fibre contacts as an additional validation of the model.

Theory

Number of fibre–fibre contacts per unit fibre length, N_c

We start with the model fibre cross-section shown in Fig. 1. A fill factor, f_h, is defined as the ratio of the fibre wall area, A_f, to the area, A_b, of the smallest rectangular bounding box that completely encloses the irregular shape of the fibre and has one side parallel to the paper plane. D_h and D_w are the dimensions of this bounding box.

A paper sheet cross-section is then idealized as a regular matrix as shown in Fig. 2, in which only the bounding boxes of the fibres are shown. The factor, b, here is an angle factor and accounts for the fibres in

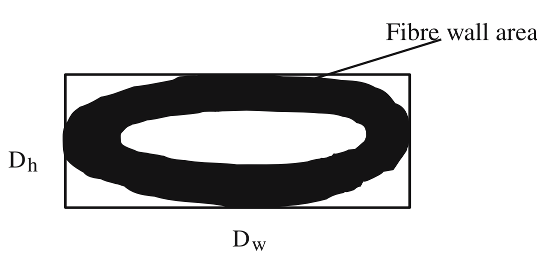

Fig. 1 The bounding box surrounding a model fibre

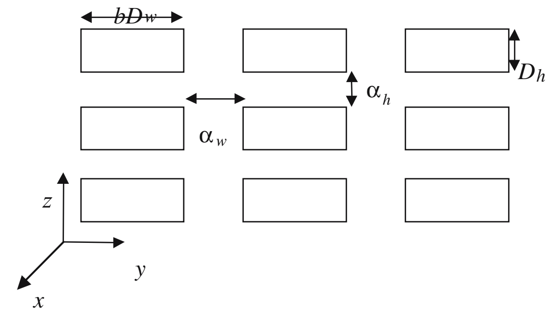

Fig. 2 Idealized cross-sectional matrix of fibres

general not cutting the y–z plane at right angles. In Fig. 2, z is the direction through the sheet thickness and the y-axis can be selected to be any direction within the plane of the sheet. The density of the sheet, ρ_a, is (ignoring the effect of surfaces)

$$\rho_a = \frac{\rho f_h D_h b D_w}{(D_h + \alpha_h)(bD_w + \alpha_w)} \tag{1}$$

where ρ is the density of the cell wall material, and α_h and α_w are the packing variables giving the spacing of the fibres within each layer and between the layers, respectively. In equation 1, f_h, D_h and D_w can be relatively readily measured by confocal microscopy, leaving b, α_w and α_h to be estimated theoretically.

To determine b, we need to determine the average angle, θ_{av}, that a fibre makes in crossing the y-axis. If the fibres are randomly oriented, as in a standard handsheet, then θ_{av} is $\pi/2 - 1$, or 32.7° [8]. b is then given by $b = 1/\cos\theta_{av} = 1.19$.

The matrix presented in the previous section represents the fibres sitting in their idealized ''equilibrium'' positions and not actually in contact with each other. However, as each fibre crosses the y–z plane shown in Fig. 2 at an angle, fibre–fibre contacts will occur either in front of or behind the plane. This is shown in Figs. 3

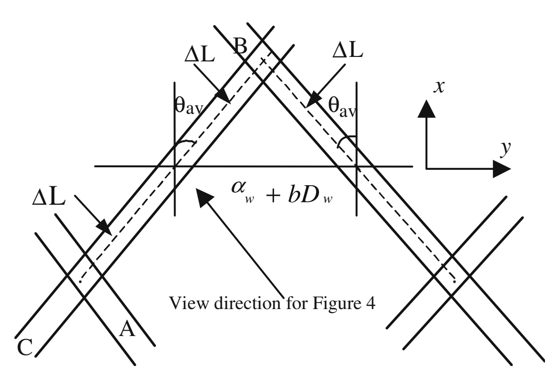

Fig. 3 x–y projection of one layer of fibres from the y–z projection shown in Fig. 2. The position of the plane from Fig. 2 is indicated by the horizontal line

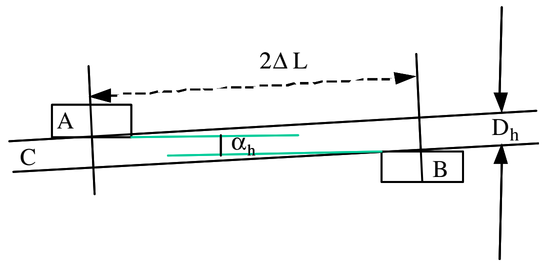

Fig. 4 Side view of the fibre–fibre crossings of fibres A, B and C from Fig. 3

and 4. Figure 3 depicts the x–y projection of three fibres (labelled A, B and C) from one layer from Fig. 2. Figure 4 shows the side view of the fibre–fibre crossings of fibres A, B and C from Fig. 3. Each fibre has been assumed to cross the plane in Fig. 2 at θ_{av}. If we define ΔL as the distance along the fibre from the plane in Fig. 2 to the fibre–fibre crossing, then $2\Delta L$ is the distance between fibre crossing midpoints and from geometry the following relationship can be derived.

$$\sin\theta_{av} = \frac{bD_w + \alpha_w}{2\Delta L} \tag{2}$$

The layer–layer separation is determined by α_h. Figure 4 shows fibres within a single layer crossing each other. As a strong theoretical basis for determining layer–layer separation is lacking, we assume that $\alpha_h = \beta\ D_h$, where β is a packing factor that will be determined experimentally.

Substituting $b = 1/\cos\theta_{av}$ and $\alpha_h = \beta\ D_h$ into Eq. 1 and using Eq. 2 yields the following expression for the sheet density

$$\rho_a = \frac{\rho f_h D_w}{(1+\beta)\Delta L \sin 2\theta_{av}} \tag{3}$$

Now if we assume that an equal number of fibre–fibre contacts on a fibre come from the layers above and below a given layer then there are three fibre–fibre contacts on $2\Delta L$ length of fibre so that the number of fibre contacts per unit length, N_c, is given by:

$$N_c = \frac{3}{2\Delta L} = \frac{3(1+\beta)}{2}\frac{\rho_a}{\rho f_h}\frac{\sin 2\theta_{av}}{D_w} \tag{4}$$

which can be rearranged to yield

$$\beta = \frac{2}{3}\frac{N_c \rho f_h D_w}{\rho_a \sin 2\theta_{av}} - 1 \tag{5}$$

Equation 5 was checked [9] against the data of Elias [10], in which the number of contacts per unit length of fibre was measured for mats of glass fibres with a diameter of 7.22 μm and varying lengths pressed at different pressures. β was found to be –0.6 [9]. The negative value of β implies that fibres from one layer have deflected into the neighbouring layers. It was also found that the calculated value of β was independent of the network density.

Expressions for RBA

Equation 4 can be used to provide theoretical expressions for the RBA. If the RBA is measured by nitrogen adsorption, then the total surface area of a unit length of fibre can be assumed to be $2(D_w + D_h)$. If the area of each fibre contact is A_c, then the RBA for nitrogen adsorption, RBA_{N_2}, can be written as: $RBA_{N_2} = N_c A_c/(2D_w + 2D_h)$, provided that the sheets are thick enough that the contribution of the sheet surfaces can be neglected. We assume that $A_c = R_c(D_w^2/\sin 2\theta_{av})$, where the factor R_c includes any deviation from the idealized model of two ribbon-like fibres crossing each other at an angle 2θ. Using Eq. 4, it can be shown that

$$RBA_{N_2} = \frac{3(1+\beta)}{2}\frac{R_c}{2}\frac{1}{(1+\delta)}\frac{\rho_a}{\rho f_h} \tag{6}$$

where $\delta = D_h/D_w$.

For the RBA measured by scattering coefficient we assume that only the top and bottom surfaces contribute to the measured scattering coefficient, from which it can be shown that

$$RBA_{sc} = \frac{3(1+\beta)}{2}\frac{R_c}{2}\frac{\rho_a}{\rho f_h}. \tag{7}$$

Materials and methods

Handsheet preparation

A laboratory made unbleached never-dried radiata pine kraft pulp, cooked to 45.6% yield with kappa number 30.0, was used in this study. This pulp was used for the subsequent fractionation experiments and for the experiments in which the fibre length was varied by cutting wet handsheets.

An AKW (Amberger Kaolinwerke Gmbh) 40 mm hydrocyclone was used to fractionate the pulp fibres into low density fibres (the accepts) and high density fibres (the rejects). The fibre apparent density factors, defined as the ratio of fibre wall area to the lumen area plus wall area [11] for the accepts and the rejects are 49.9 ± 2.1 and 56.8 ± 2.3, respectively (where the errors give the 95% confidence intervals). The fibre length was also changed by cutting wet handsheets using a specifically designed die. Four fractions of fibres with different fibre length were created and are labelled L_0 to L_3. The average length weighted fibre lengths of L_0, L_1, L_2 and L_3 are 3.14, 2.53, 2.10 and 1.80 mm, respectively. For each fraction, the cut wet sheets were then reslushed and formed into handsheets. Details of this experiment have been reported previously [6]. When sheets are cut and reslushed in this way, the fibre length is changed without affecting the cross-sectional shape of the fibre.

Handsheets of 60 g/m^2 from different fractions of fibres generated by fractionation or cutting wet handsheets were made on a Moving Belt Sheet Former [12]. Handsheets for examination in the confocal microscope were made from fibres pre-dyed with Acridine Orange. Handsheets made from fractionated fibres were pressed dynamically using a Sheet Roller Press at one of five pressing levels, while the handsheets made from the four fractions with different fibre lengths were pressed statically for 2 min using a hydraulic press, at one of five pressing levels. The pressing load or pressure of each pressing level is given in Table 1. Each set of handsheets was denoted by the pulp name followed by the pressing level. For example, $AccP_1$ represents handsheets made from the accepts and pressed at pressing level P_1. All of the handsheets used in the experiments were dried under restraint at 23 °C and 50%RH.

Measurement of RBA

The free surface areas of the handsheets were calculated using the Brunauer, Emmett and Teller (B.E.T.) theory [13]. The isotherm for this calculation was obtained by nitrogen adsorption using a Micromeritics ASAP (Accelerated Surface Area and Porosimetry) 2010. Each data point given here was the average of two duplicate measurements. The scattering coefficient was determined using a Colortouch Brightness tester at 700 nm wavelength.

Table 1 Pressing load and pressure for dynamic and static pressing

Press level	Dynamic press load (kN/m)	Static pressing pressure (kPa)
P_1	0[a]	100
P_2	3.0	200
P_3	7.0	500
P_4	3.0 + 10 × 2 passes	2000
P_5	3.0 + 10 + 20 × 10 passes	4000

[a] "0" represents pressing with no additional force applied to the rollers

For each sample, a fibre suspension with very low consistency (0.04%) was sprayed on a teflon surface and dried in air. The dried fibres were then collected carefully, avoiding losing fines. Fibres dried in this way are rarely in contact with one another and should have an equivalent free fibre surface to an unbonded sheet. The free fibre surface area of these spray dried fibres were measured by the nitrogen adsorption method and used to calculate RBA by RBA = $(A_u - A)/A_u$, where A_u is the free surface area of the unbonded fibres and A is the free surface area of the normal sheets.

Microscopic measurement of the number of fibre–fibre contacts and the fill factor

The number of fibre–fibre contacts was measured directly in paper cross-sectional images generated by using a combined technique of resin embedding and confocal microscopy. The method involved choosing fibres of interest that had been cut longitudinally by the cross-section and counting the number of fibres in contact with each of fibre of interest. Fibre–fibre contacts were classified into full contacts and partial contacts. The cross-section of a fibre is assumed to have two long sides and two short sides. When one of the long sides is totally in contact with the fibre of interest, this contact is a full contact. Otherwise, the contact is a partial contact. The details of the measurement were reported in [6].

The fill factor was measured on fibres that were cut through their cross-section by the paper sample cross-section. Fibre positions were measured at the surface of and 10 μm under the sheet cross-section. The shift in position of the fibre between the two measurements was used to determine the angle of orientation of the fibre with respect to the sample cross-section. The measured fibre dimensions in the sheet cross-section were then corrected for the orientation of the fibres, before being fitted to calculate the bounding box. Further details can be found in [8].

Due to the time required for these measurements only a selection of the available samples was measured for the number of contacts.

Model verification

Determination of β in Eq. 4

The packing factor, β, required for Eq. 4 was determined by using the measured results of the number of fibre–fibre contacts in Eq. 5.

Several points need to be clarified here. Equation 4 assumed that all of the contacts in paper are identical and are full contacts. However, fibre–fibre contacts measured in the handsheets can be divided into two types, viz. full contacts and partial contacts [6]. There is no theoretical basis for determining how many partial contacts are equivalent to one full contact. However, the "conversion factor" is most likely to be within the range 1–2. In this range, the value that gave the best fit between the measured number of fibre–fibre contacts per unit fibre length (N_{cm}) and the predicted number of fibre–fibre contacts per unit fibre length (N_{cp}) was 1.5. This was used to calculate the equivalent number of full contacts for each sample. Table 2 then shows the value of β for each sample required to exactly match the measured number of equivalent full fibre–fibre contacts with the prediction made by Eq. 4. This was done by substituting the measured value of the number of contacts into Eq. 5. It can be seen that the calculated value of β is constant, within errors, for all samples. In all following calculations, we used an average value of $\beta = -0.29$.

A comparison between the measured number of fibre–fibre contacts and the prediction made by Eq. 4 using $\beta = -0.29$ are shown in Fig. 5. When the trend line is forced to pass the origin, the slope (0.99) is very close to 1 and the correlation coefficient R^2 is 0.93. The closeness of the predictions and the measurements indicates that the model, with best-fit parameters, predicts the number of fibre–fibre contacts with reasonable accuracy.

Verifying the expressions for RBA

The measurement for RBA is always problematic. In this study, the nitrogen adsorption method was used to measure the free surface area for all samples. As discussed before, the surface area of completely unbonded sheets was determined by measuring the free surface of spray-dried fibres. The measured surface areas of unbonded sheets for samples L_0, L_1, L_2 and L_3 are 905, 920, 921 and 927 m^2/kg, respectively, and for the accepts and the rejects are 993 m^2/kg and 1,063 m^2/kg, respectively. The RBA was then calculated from RBA = $(A_u - A)/A_u$, where A_u is the free surface area of the unbonded fibres and A is the free surface area of the normal sheets.

Table 2 Determination of β using the measured results of fibre–fibre contacts in paper and assuming that 1 full contact equals 1.5 partial contacts

Sample	D_w (μm)	f_h	N_{cm} (No/m)	β
L_0P_3	30.68	0.55	19,375 ± 4,008	–0.31 ± 0.14[a]
L_1P_3	34.45	0.52	18,710 ± 4,215	–0.31 ± 0.15
L_2P_3	33.11	0.55	18,725 ± 4,074	–0.33 ± 0.15
$AccP_1$	30.19	0.43	9,743 ± 1,710	–0.36 ± 0.17
$AccP_3$	34.08	0.45	16,309 ± 2,509	–0.31 ± 0.12
$AccP_5$	36.38	0.51	22,784 ± 3,521	–0.29 ± 0.12
$RejP_1$	28.03	0.46	9,377 ± 1,992	–0.31 ± 0.16
$RejP_3$	32.22	0.49	15,757 ± 3,141	–0.16 ± 0.20

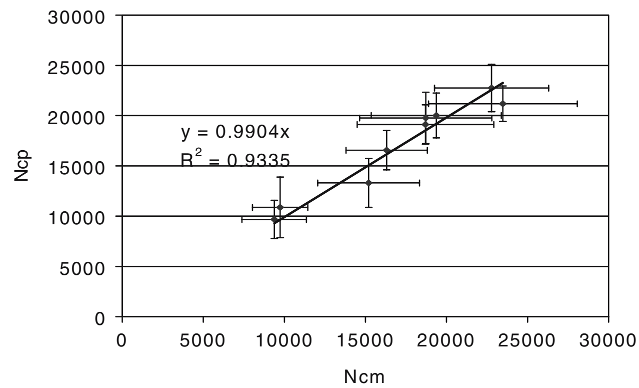

Fig. 5 Correlation of measured number of fibre–fibre contacts per unit fibre length against the predictions made by Eq. 5

Figure 6 compares, for all the samples used in this study, the measured RBA_{N_2} and the RBA_{N_2} predicted by Eq. 6. The factor R_c in Eq. 6 was first determined by finding the value of R_c that maximized the R^2 statistic of the correlation between measured and predicted values of RBA_{N_2} by Eq. 6. The value of R_c determined in this manner was 1.35, which is greater than 1, indicating that the actual bond width is larger than D_w. This is reasonable as fibres tend to flatten when they are bonded together, forming a skirt-shaped bond [14]. The best-fit line, shown in Fig. 6, shows an intercept close to zero and slope close to 1.0 with a reasonably good correlation ($R^2 = 0.88$) between measurements and predictions. Overall this indicates that Eq. 6 can

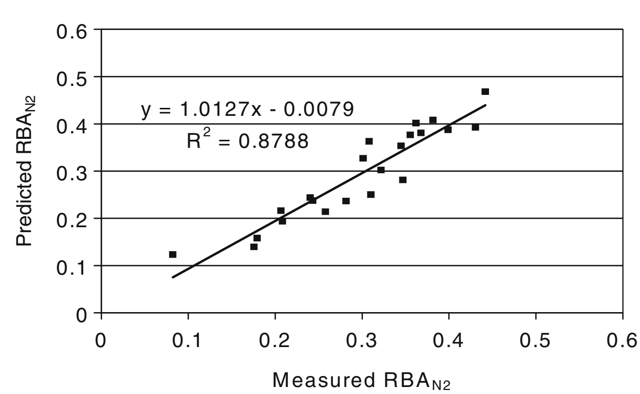

Fig. 6 The correlation between the measured RBA by N_2 adsorption and the predicted RBA by Eq. 6

predict RBA_{N_2} well. This also indirectly proves that the model for the number of fibre–fibre contacts, Eq. 5, is reasonable.

Traditionally, the Ingmanson and Thode pressing/beating extrapolation method [15] has been the most widely used method for RBA measurement. In this method the light scattering coefficient for an unbonded sheet is calculated by extrapolating a plot of tensile strength versus scattering coefficient to determine the *y*-axis (zero-strength) intercept. This intercept (called S_0) is believed to be equivalent to the scattering coefficient of an unbonded sheet. The RBA_{sc} for a sheet with scattering coefficient, S, is then calculated by assuming that the bonded area is proportional to scattering coefficient, from which $RBA_{sc} = (S_0 - S)/S_0$.

The Ingmanson and Thode extrapolations are shown in Fig. 7 for the samples with varying fibre lengths. The values of S_0 determined by this method of extrapolation for L_0, L_1, L_2 and L_3 are 42.0, 34.1, 33.1 and 28.6 m^2/kg. These results are unreasonable because these fibre fractions only differ in fibre length and should have almost the same S_0. It is possible that cutting the fibres might have caused a small increase in the fibre area, as the cutting process creates new surfaces. However, it is inconceivable that the cutting process would have caused the surface area (and therefore S_0) to decrease and accordingly the extrapolation method is unreliable. Obviously, an alternative method needs to be found to estimate S_0.

A consistent linear relationship was found between the light scattering coefficient and the BET area for samples made from fibre fractions generated by cutting wet handsheets (see Fig. 8). The best-fit linear equation is also shown in Fig. 8. Similar relationships have previously been reported [16, 17]. S_0 was calculated by taking the values of BET area of the spray dried (unbonded) sheets into the linear equation in Fig. 8. The S_0 obtained using this method was 32.4 m^2/kg for L_0, 33.1 m^2/kg for L_1, 33.2 m^2/kg for L_2 and 33.5 m^2/kg for L_3. These values were then averaged and used as S_0 for RBA calculation for all of the four different fibre length fractions. It is important to note that, as expected, S_0, determined in this manner, does not change as the fibre length is decreased. Unfortunately, the relationship between BET area and light scattering coefficient was highly non-linear for samples made from the rejects or from the accepts (Data is shown in [9]) and so this procedure was not able to be used for these samples. Currently, we have no good explanation for this non-linear relationship.

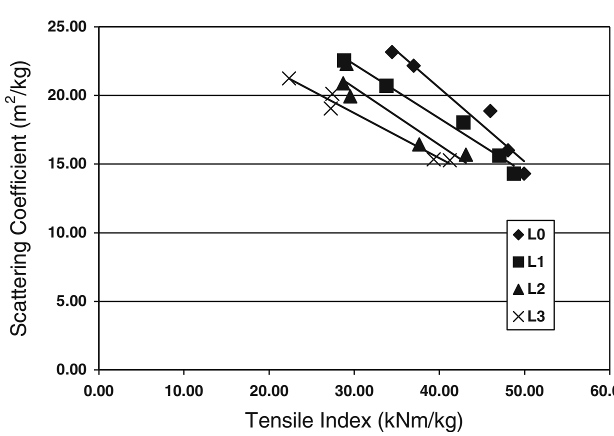

Fig. 7 Plot between tensile index and light scattering coefficient for the sheets made from the cut fibres

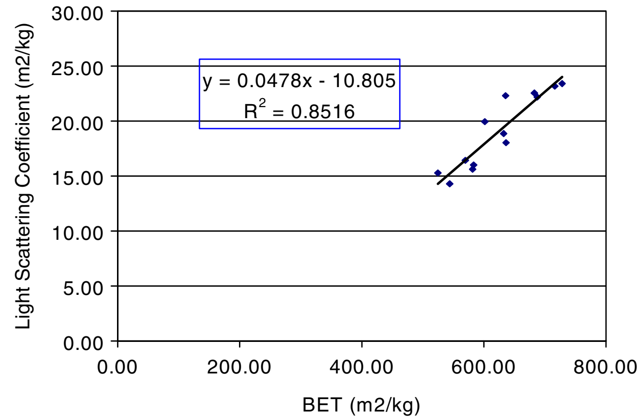

Fig. 8 The relationship between the BET area and the light scattering coefficient for samples made from fibres with different length and pressed at different pressing levels

A comparison, for the sheets with different fibre length, between the measured RBA_{sc} and the predicted RBA_{sc} by Eq. 7 is shown in Fig. 9. The value of R in Eq. 7 was found by minimising the errors between measured and calculated RBA_{sc}, in a similar manner to Fig. 6 for RBA_{N_2}. The value of R_c found in this way was 1.39, which is consistent with the value of R_c determined using RBA_{N_2}. This is to be expected, given that R_c represents the spreading of the bonded area and this should not depend on the method by which it is measured. The line of best fit in Fig. 9 shows an intercept very close to zero and a slope close to 1 and very high R^2 (=0.94) indicating that Eq. 7 can predict RBA very well. This further proves that the new model for number of fibre–fibre contacts is valid.

Conclusions

A new model that relates the fibre cross-sectional dimensions and the density of paper to the number of fibre–fibre contacts per unit length of fibre has been

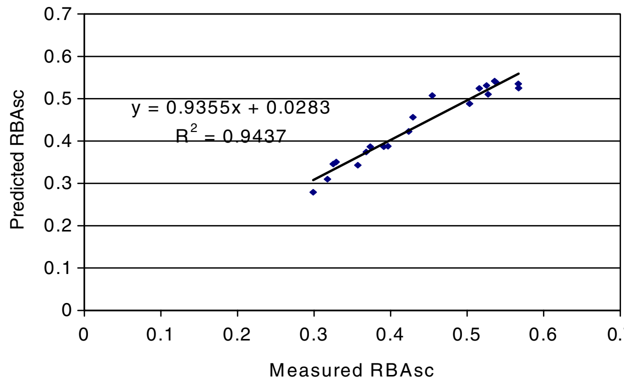

Fig. 9 Correlation between measured RBA_{sc} and RBA_{sc} predicted by Eq. 7 for the sheets with fibres of different fibre lengths generated cutting

developed. The model has been verified directly by using data of fibre–fibre contacts measured in paper. The comparison between the measurements and the predictions made by the new model shows a very good correlation, indicating that the model is valid.

The model has also been converted into two expressions for RBA, which have been verified against RBA measured by nitrogen adsorption and scattering coefficient. It has been shown that both expressions can predict RBA well. This further proves that the new model for fibre–fibre contacts is valid. The data showed that the Ingmanson and Thode extrapolation to determine S_0 is questionable.

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