A new approach for quantitative analysis of paper structure at the fibre level

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SUMMARY

This paper describes and demonstrates a new method for measuring fibre cross-sectional dimensions, fibre collapse, fibre orientation and relative fibre positions in sheets of paper embedded in resin. Confocal laser scanning microscopy (CLSM) is used to acquire images of paper cross-sections in a manner that avoids artifacts from sample preparation. The measurements of fibre dimensions are validated by comparing the results measured in paper with those measured on freely dried fibres on glass slides. The measurement of fibre orientation is validated by comparing the measured results with a theoretical average value of orientation in a random sheet.

A new term, 'fibre fill factor', is defined and measured with the new technique. The utility of the fill factor for describing the degree of collapse of individual fibres in a sheet is discussed.

Keywords

Confocal laser scanning microscopy, quantitative analysis, fibre dimensions, fibre orientation, fill factor

Many studies have been conducted with the aim of describing the strength properties of paper in terms of the structural properties of the fibre network and the mechanical properties of the fibres (*1-3*). Fibre dimensions and orientation have been shown to be very important for paper strength in these studies. However, no technique is available to obtain quantitative data of fibre dimensions and this 3-dimensional orientation in situ to fully verify these network models. New techniques are required to quantify paper structure at the fibre level.

It is essential to expose and image the internal structure of paper for structure analysis and many techniques have been used for doing this. The most commonly used technique to expose internal structure is thin sectioning (microtomy). Some examples of the successful use of thin sectioning techniques to study paper structure, using both transmitted light and electron microscopy are given in references (*4-7*). The main problem of thin sectioning is that the sample structure can be distorted during mechanical sectioning. Its use for quantitative analysis of paper structure is therefore problematical. Williams et al. (*8*) and Williams and Drummond (*9*) recently developed a new technique in which samples were embedded in resin and the cross-sections of the samples were then exposed by grinding the block surface using progressively finer abrasive papers. In order to get proper topographic contrast for scanning electron microscope (SEM) viewing, chemicals were used to etch away a layer of the supporting resin from the block surface. The Williams et al. technique avoids the effect of the compressive force during the thin sectioning. However, great care must be taken during surface resin removal with chemicals to avoid swelling of the exposed fibres and paper surface. In addition to this, the SEM can only obtain the image of the cross-sectional surface, and this may not be free of artifacts of sample preparation. Using this technique, Forseth and Helle (*10*) quantified the change in fibre lumen area in paper during moistening. In their study, only fibres cut more or less parallel to the fibre length direction were measured to prevent overestimation of the fibre wall thickness.

Confocal laser scanning microscopy has been commonly used in qualitative analysis of paper structure in recent years (*11-13*). The major feature of the CLSM is its optical sectioning capability, with which it is able to obtain three-dimensional images of fibre and paper non-destructively. However, the signal intensity diminishes rapidly with increasing depth in the sample (*14*). Therefore these studies, looking through the in-plane surface of the samples, could not obtain a complete view of a sheet cross-section. (*11-13*).

More recently, Dickson (*15,16*) conducted successful quantitative analysis of paper cross-sections using a combination of the sample preparation technique developed by Williams and Drummond (*9*) and the confocal laser scanning microscopy technique. Complete cross-sections of paper were imaged with CLSM just below the cross-sectional surface to avoid the artifacts on the surface (*15*). Dickson quantified the profile properties of the paper cross-section including the wall area, the total pore area, interfibre pore area and lumen area. The dimensions of individual fibres in the sheet were not measured in any of these studies.

Fibre orientation has often been measured by tracing a fraction of dyed fibres in a sheet (*17,18*). The fibre orientation measured by this technique is the in-plane orientation and the fibres traced are always limited to the paper surface. Another technique is to split a sheet into thin, transparent layers and then measure the fibre orientation in all these layers with image analysis (*19*). This technique allows the in-plane fibre orientation in different layers to be measured but not the out-of-plane fibre orientation. A more accurate and relatively easy technique is still required for measurement of overall fibre orientation.

In this study, we present a new method for quantifying paper structure. The dimensions and orientations of individual fibres in a sheet are measured simultaneously using a combination of resin embedding and confocal laser scanning microscopy. This is the first technique that can simultaneously obtain all this information directly in situ.

MATERIALS AND METHODS

Staining of pulp fibres and handsheet formation

Recycled pulp of commercial plaster linerboard and laboratory made never dried radiata pine kraft pulp (Kappa no. 95) dyed in a stock divider at a concentration of 1.5% prior to handsheet formation. The dye (Acridine Orange) was added at a rate of 0.0005% by mass in water. From each pulp, three sets of square handsheets were made on a Moving Belt Sheet Former (*20*).

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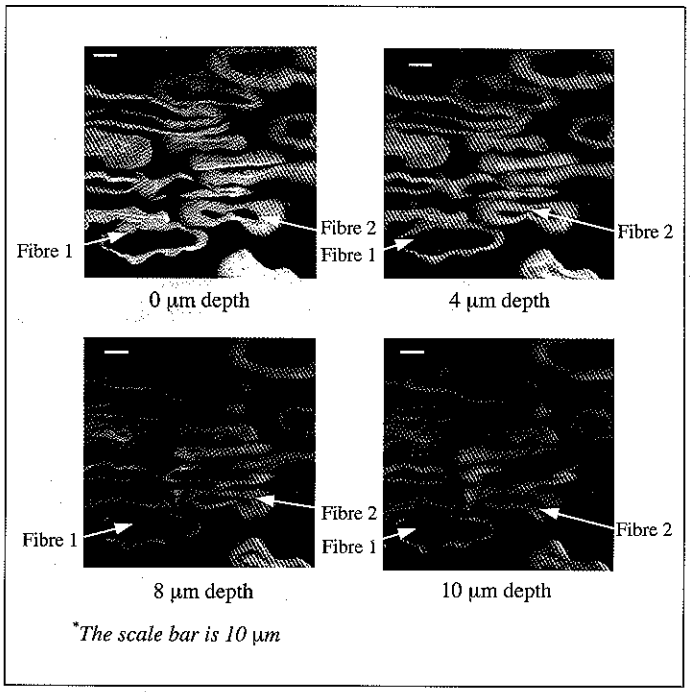

Fig. 1 Cross-sectional images of paper acquired at depths of 0, 4, 8 and 10 μm.

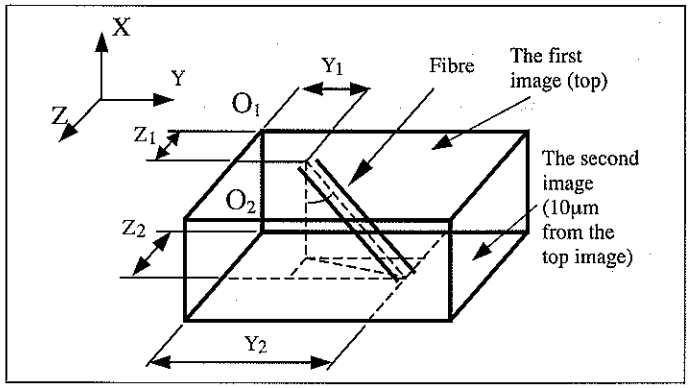

Fig. 2 Depiction of the measurement of fibre orientation in paper.

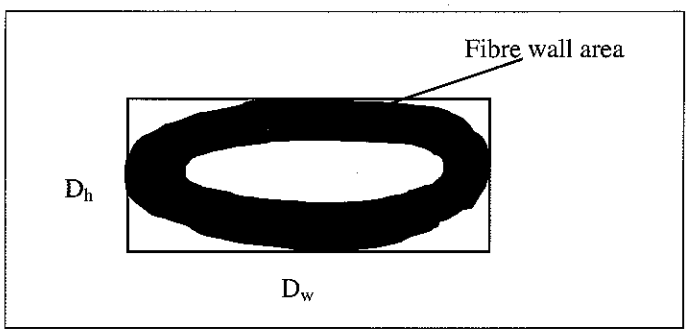

Fig. 3 The bounding box surrounding a model fibre.

Two sets of handsheets from each pulp were pressed using a Sheet Roll Press, one set at low pressure and one set at high pressure. One further set of handsheets was tested without pressing. The handsheets made from the recycled pulp were denoted as R_0, R_L and R_H, and those made from the kraft pulp were denoted correspondingly as Kp_0, Kp_L and Kp_H.

Sample preparation and imaging

The resin used for sample embedding was Epofix (from Struers (*21*)). This resin met the following criteria:

a. No reaction with fibres.
b. Good penetration into the fibres.
c. Refractive index of the resin block is 1.57, which is close to the fibre refractive index 1.53.

Samples were first placed directly in 100% resin and degassed overnight so that the resin could penetrate into the paper structure and expel gas in the paper. The samples were then taken out of the resin and put on a plate for about 5 minutes to allow the resin at the sample surfaces to drain away. The samples were then put into a plastic mold, and a pre-degassed mixture of resin and hardener (15 parts of resin and 2 parts of hardener) was poured into the mold and the whole sample was put into a vacuum apparatus and left until the resin cured into a block. It is very important to ensure that all gas is removed from the samples and the internal structure of the paper is thoroughly penetrated by the mixture of resin and hardener. The cross-sections of the samples were then exposed by abrading the block surface with a range of abrasive papers of decreasing roughness.

Imaging was performed with a HBH Fibrescan C900 confocal laser scanning microscope using a 60× oil immersion lens. The inspection area of each image frame was 100×100 μm with a resolution of 512×512 pixel.

For each region of interest, a group of images of the region was captured at different depths from the cross-sectional surface. Fifty groups of cross-sectional images of each sample were captured and processed for measurement of fibre orientation and fibre cross-sectional dimensions. A typical group of cross- sectional images of the paper is given in Figure 1. The image with 0-depth was captured just below the exposed surface of the sample. The depths of the other images are measured with respect to the 0-depth image. It can be seen that the image brightness decreases with increasing depth. The maximum scanning depth used in this study was 10 μm. Scanning deeper led to a rapid deterioration of image quality.

Image analysis

The image analysis was conducted using commercial image analysis software (OPTIMAS 6.1). The analysis sequence used was: 1) select a region of interest; 2) smooth the image with a 3×3 Gaussian filter; 3) adjust the threshold level; and 4) extract features. A macro was written in the software for performing the above process automatically. The boundary of a fibre was identified by manually adjusting the threshold. For fibres that touched each other, a line was drawn between them to separate the fibres but keep the features of the fibre of interest unchanged.

Fibre orientation was measured by following the positions of individual fibres in sheet cross-sections scanned at 0 μm depth and 10 μm depth. The position of the centre of mass of the fibre cross-section was determined by the distance from the centre of mass to a reference point in the Y and Z directions. As shown in Figure 2, the centre of mass of the fibre cross-section in the 0-depth image is (Y_1, Z_1), and in the 10 μm depth image is (Y_2, Z_2). The angle θ that the fibre makes to the X direction (the plane of the paper) can then be calculated from geometry. For example, in Figure 1, the position of fibre 1 has no apparent change when scanning down from 0-depth inside the sample to 10μm deep. This indicates fibre 1 is almost perpendicular to the exposed surface. However, the position of fibre 2 shifts markedly indicating that fibre 2 sits at an angle other than 90° to the exposed surface of the sample.

Table 1
Summary of the experimental results.

Pulp sample	Handsheet sample code	Wet press level	Apparent density (kg/m³)	Tensile index (kNm/kg)	Fibre wall area (μm²) (at 0 μm depth)	Fibre wall area (μm²) at 4 μm (depth)	Fill factor (at cross-sectional surface)	Fill factor (at 4 μm depth)	Fibre angle to X-direction (degree)	Number of fibres measured
Recycled pulp	R_0	Not pressed	234	17.3	220±9*	181±9*	0.56±0.01*	0.54±0.01*	25.8	147
	R_L	Lightly pressed	334	22.9	223±9	179±9	0.53±0.01	0.55±0.01	24.6	146
	R_H	Highly pressed	380	22.3	209±10	172±9	0.56±0.01	0.57±0.01	26.0	147
Recycled pulp fibres dried on slides	/	/	/	/	170±8	170±8	/	/	/	319
Kraft pulp	Kp_0	Not pressed	112	13.1	312±9	266±9	0.45±0.01	0.46±0.01	28.1	176
	Kp_L	Lightly pressed	215	29.2	295±10	256±9	0.50±0.01	0.48±0.01	25.2	192
	Kp_H	Highly pressed	393	51.0	294±10	260±9	0.55±0.01	0.56±0.01	26.9	180
Kraft pulp fibres dried on slides	/	/	/	/	249±9	249±9	/	/	/	338

* ± 95% confidence interval.

The wall areas of fibres from both pulps were measured from both the 0 μm and 4 μm depth images. If the fibre is not perpendicular to the paper cross-section, the wall area will be enlarged by a factor of cosθ. The measured wall area of each individual fibre has been corrected by this factor.

As shown in Figure 3, the fill factor, f_h, is defined as the ratio of the fibre wall area, A_f, to the area of the smallest rectangular bounding box, A_b, that can completely enclose the irregular shape of the fibre and with one side parallel to the paper plane. The fill factor was measured and used to describe the degree of collapse of the fibre. For fibres from the same pulp, the greater the fill factors, the higher the degree of collapse of the fibres.

Fibre wall areas of the two pulps were also measured on freely dried fibres on glass sides using a confocal laser scanning microscope and following a procedure described by (*22*). These results were compared with the fibre wall areas measured in situ on fibres in paper sheets.

RESULTS AND DISCUSSION

Fibre wall area

Table 1 summarises the fibre dimensions and the sheet properties measured in this study. As can been seen from Table 1, the average fibre wall areas of the recycled pulp samples R_0, R_L and R_H are very close to each other, especially those measurements acquired 4 μm below the sample surface. A similar situation is shown for the kraft pulp samples Kp_0, Kp_L and Kp_H. This is as expected because wet pressing should not change the wall areas of fibres in the sheets.

Comparisons were made between the fibre wall areas measured in sheets and those measured on dried fibres on glass sides for the purpose of validating the measurements made by the new method. As shown in Table 1, for both of the pulps used in this study, when the fibre wall areas were measured in images of paper cross-sections scanned 4 μm below the sample surface, the results were very close to those measured on the same pulp fibres dried on glass sides. The maximum difference between them is 11 μm² for the recycled fibres and 17 μm² for the kraft fibres. The frequency distributions of fibre wall areas of these measurements are given in Figure 4 for the recycled pulp and Figure 5 for the kraft pulp. From these figures, it can be seen that the distributions of fibre wall areas of samples made from the same pulp fibres, but pressed at different pressing levels, are not only in close agreement with each other but also in

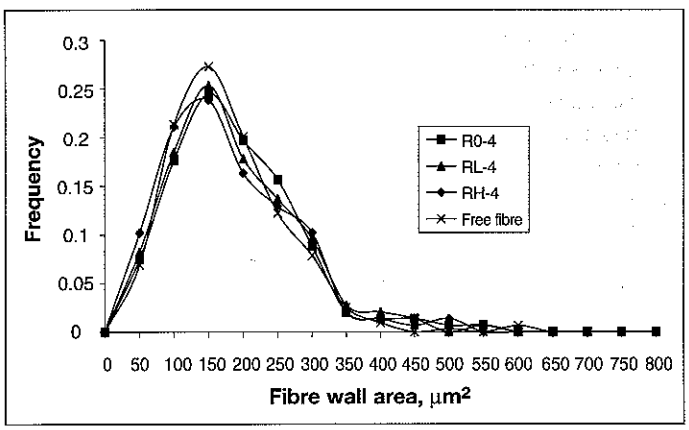

Fig. 4 Distribution of fibre wall areas for the recycled fibres measured in paper and on a glass slide. The fibre wall areas measured in paper were determined from images taken at 4 μm depth.

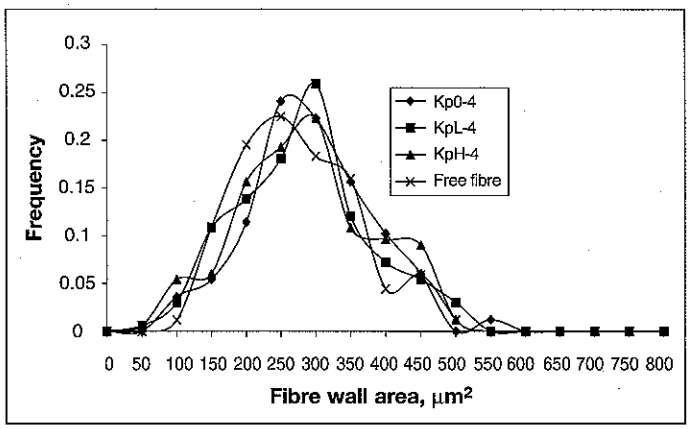

Fig. 5 Distribution of fibre wall areas for the kraft fibres measured in sheet and on a glass slide. The fibre wall areas measured in paper were determined from images taken at 4 μm depth.

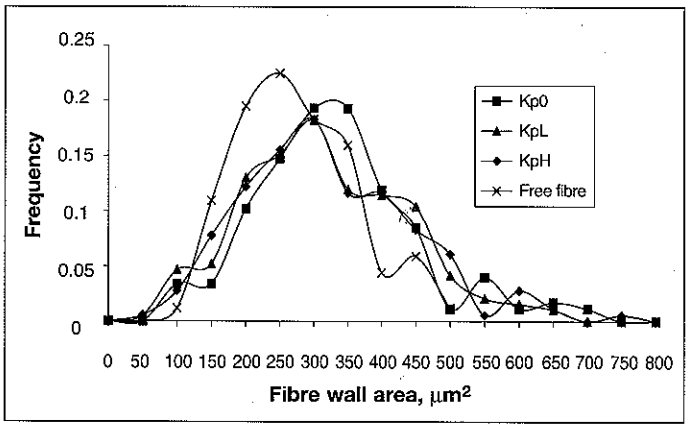

Fig. 6 Distribution of fibre wall areas for the kraft fibres measured in sheet and on a glass slide. The fibre wall areas measured in paper were determined from the surface images.

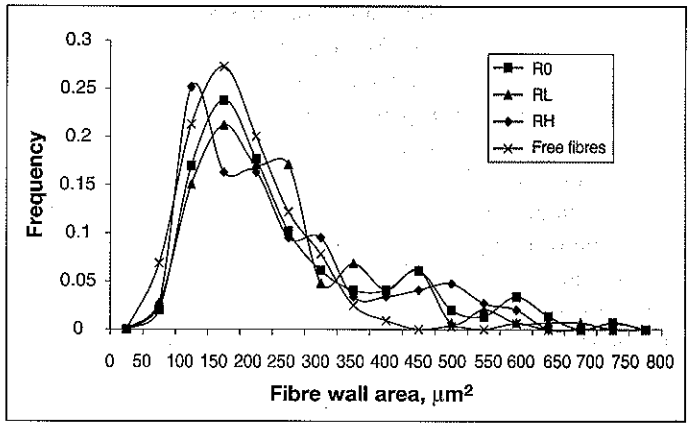

Fig. 7 Distribution of fibre wall areas for the recycled fibres measured in sheet and on a glass slide. The fibre wall areas measured in paper were determined from the surface images.

close agreement with those measured on the freely dried fibres on glass. The fibre wall areas of recycled fibres were mostly distributed between 100 μm² and 200 μm², and between 200 μm² and 350 μm² for the kraft fibres. These comparisons indicate that the measurements of fibre wall areas using the proposed new method and taking measurements from images of paper cross-sections acquired 4 μm below the surface of the sample cross-sections are valid.

When the measurements were made on the 0-depth images of the paper cross-sections, the mean values of the fibre wall areas of both pulps were greater than those of their corresponding freely dried fibres (see Table 1). The average difference is 50.9 μm² for the kraft fibres and 47.2 μm² for the recycled fibres. Despite these relatively large differences in average fibre wall areas, the frequency distributions are still quite similar (see Fig. 6 and Fig. 7). It is very interesting to note that the results measured in sheets showed some large values (greater than 500 μm²), which could not be seen in the results measured on free fibres from the same pulp. It is believed that these large fibre wall area values result from artifacts produced by sample preparation. A careful observation of the images of the paper cross-sections found that there were a few fibres, especially large fibres, which seemed to have been 'smeared' during grinding and polishing. The 'smeared' fibres obviously created larger images and this effect would cause an overestimation of fibre wall areas when these fibres are included in the measurements. The 'smeared' effect implies that the sample structure had not been supported sufficiently by the resin block. A possible reason for this could be that the resin that penetrated into the paper structure during the degassing treatment was not replaced sufficiently by the mixture of resin and hardener in the subsequent step. Another possibility is that the resin and hardener mixture may not have penetrated into the fibre walls. This indicates that the sample surface was not free of artifacts although great care was taken during sample preparation. When measurements were taken 4 μm below the sample surface, there were only a few large values (as shown in Fig. 4 and Fig. 5) and the measured mean values of fibre wall areas were close to the results obtained for the free fibres. It appears that taking measurements at 4 μm below the sample cross-section eliminates any artifacts at the sample cross-sectional surface.

Fibre orientation

The average values of the angles of fibres to the x-direction (Fig. 2) are given in Table 1. The fibre orientation measured in this study was actually the orientation of fibre segments. In fact it is almost impossible to properly define the fibre orientation because fibres are not straight in a sheet. However, the orientation of fibre segments might be more important for paper properties than the average orientation of entire fibres.

In this study the measurements for fibre orientation calculation included any position shift of the fibre, in any direction, between the surface and 10 μm depth into the paper cross-section, and the fibres measured were sampled randomly through the whole thickness of the paper. Therefore, the fibre orientation measured is an overall fibre orientation. Although the data of the position shift of fibres in sheets are not given in this paper, in fact the positions of fibres were most likely to shift in both y- and z-directions (see Fig. 2), although the shift in the z-direction was usually very small. If the position shift in the z-direction was zero, it indicates that the fibre is in-plane with respect to the paper surface. Otherwise the fibre is out-of-plane. The small shift in the z-direction indicates that the out-of-plane movement of the fibres in the handsheets used in this study was small.

If θ is the in-plane fibre angle to any given line within the plane in the direction perpendicular to the exposed surface (the x-direction in Fig. 2), then from simple geometry the average value for a randomly oriented sheet, θ_{av}, is given by equation 1. Integrating equation 1 gives an average value of π/2-1, or 32.7°.

$$\theta_{av} = \frac{\int_0^{\pi/2} \theta\cos\theta \, d\theta}{\int_0^{\pi/2} \cos\theta \, d\theta} \qquad [1]$$

From Table 1, there is about an 8° difference between the average measured value and this theoretical value of fibre orientation. This difference is considered to be reasonable because the theoretical value includes all fibres with angles between 0° and 90° but in this study fibres with high angles were excluded. After examining the raw data of fibre angles carefully, it was found that the measured values seldom included fibres with angles over 80°. The reason for this is that the 'high angle fibres' are likely to be sectioned along the fibre axis and were treated as 90° fibres and not included in the measurements. This difference between the measured average fibre angle and the theoretical average value does not affect the validity of the angle measurements using the new technique. It appears that the effect of the out-of-plane movements of fibres

in the sheets used in this study on the overall fibre orientation is also negligible.

This study made no attempt at measuring in-plane and out-of-plane orientation separately, however the new technique could be used as needed to measure in-plane fibre orientation as well as out-of-plane movement of fibres in a sheet. It can also be used to measure the distribution of fibre orientation through the thickness of a sheet.

Application of the new technique

One possible application of the new technique is to quantify the behaviour of individual fibres in a sheet after wet pressing. Consolidation of the paper structure during pressing will occur through a combination of compressing fibres closer together and collapse of the fibres. How much the fibres are collapsed and brought closer together in wet pressing is still not clear. It is only possible to quantify these changes when the fibre dimensions in the sheet can be measured.

The new term, fill factor of fibre, as defined by equation 1 was used to quantify the degree of collapse of fibres in sheets. As shown in Table 1, the fill factors of the recycled fibres in sheets show no significant change with increasing intensity of wet pressing. Additionally, the value of the fill factor measured in the 0-depth images and that measured in the 4 µm depth images shows no significant difference. These results indicate that the degree of collapse of the recycled fibres is not increased in wet pressing.

In the case of the kraft pulp fibres, the fill factor increases steadily as the pressing intensity is increased. This means that wet pressing has increased the degree of collapse of the kraft fibres in the sheets. The collapse behaviour of the kraft fibres in wet pressing is different to that of the recycled fibres. A possible explanation for this difference is that the recycled fibres have already been collapsed to a certain degree during the previous papermaking process. It is therefore not easy to collapse these fibres further by wet pressing. The kraft pulp fibres were never dried and most of the fibres were uncollapsed after pulping. Therefore they are more likely to be collapsed in wet pressing.

The fibres in a sheet move closer together in wet pressing, forming more bonds. However other types of movement of fibres in wet pressing must still be clarified. One additional type of movement that was observed when the sheet was pressed was that the fibres were twisted so that they lay flat and became closer to each other. The new technique presented here can be used to quantify this twist of fibres in wet pressing. This will be done in a future study.

CONCLUSIONS

Fibre dimensions, fibre orientation and fibre collapse have been measured directly in paper using a combined technique of resin embedding and confocal laser scanning microscopy. The images of the paper cross-sections have to be acquired several microns below the cross-sectional surface to avoid the artifacts produced by sample preparation. This is the first time a technique has been demonstrated for quantifying paper structure at the fibre level. Comparisons between the measured values of fibre wall areas by the new technique in paper with those measured by the routine microscopy technique on free fibres show close agreement. The fibre orientation measured in the handsheets compares reasonably well with the theoretical value of fibre orientation of a random sheet. These results show that the measurements made by the new technique are valid.

A fill factor has been defined and used to characterise the collapse behaviour of individual fibres in paper in wet pressing. The preliminary results show that the recycled fibres behave differently in wet pressing compared to the never-dried kraft fibres. The degree of collapse of the recycled fibres was not changed significantly by wet pressing, while the degree of collapse of the kraft fibres was increased as the intensity of wet pressing increased.

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