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Extracting fiber and network connectivity data using microtomography images of paper

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KEYWORDS: Fiber network, X-ray microtomography, Fibers, Fiber-to-fiber bonds, Free fiber length, Number of contacts, Contact density

SUMMARY: We apply image analysis methods based on micro-computed tomography (μ CT) to extract the parameters that characterize the structure and bonding parameters in the fiber network of paper. The scaling and variational properties of μ CT images are examined by analyzing paper structural properties of two 1×1 mm² test pieces, which have been cut out from a low-grammage handsheet. We demonstrate the applicability of the methods for extracting the free fiber length, fiber cross-sectional data, the distances between the fibers, and the number of fiber-to-fiber bonds, which are the key properties required for the adequate representation of the network in numerical models. We compare the extracted connectivity data with the early reported analytical estimations and conclude that the number of contacts in three-dimensional networks is controlled by the fiber aspect ratio. In addition, we compare the cross-sectional data with those measured by the fiber morphology characterization tools and estimate the fiber shrinkage from completely wet to dry state to be nearly 20%.

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Abbreviations used in this paper

2D = two-dimensional
3D = three-dimensional
FFL = free fiber length
FMA = fiber morphology analyzer
NFC = number of fiber crossings
NCP = number of contact points
 μ CT = micro-computed tomography
WH = width-to-height

Fiber structural properties and network connectivity are the essential characteristics of network materials such as paper and packaging. Having reliable estimations of these characteristics is essential for modeling the networks and

quantifying the impact of the changes introduced by the paper-making process.

The level of inter-fiber bonding correlates with paper strength through the sheet density or the light-scattering coefficient (Skowronski, 1991). However, there is no reliable method of estimating the number of bonds in paper, which creates a hindrance in terms of the ability to describe how different fiber modifications affect the paper structure.

Certain operations in paper-making, such as adding fines, changing the degree of refinement of the pulp, modifying the pressing conditions, and so on, can alter the fiber connectivity, affect the fiber cross-sectional dimensions, and result in densification (Batchelor, He, 2005; Bristow, Kolseth, 1986; Niskanen, 1998; Torgnysdotter *et al.*, 2007). Correct interpretation of the changes in the network structure as well as their impact on the densification and mechanical properties of paper products is particularly vital for connecting laboratory and production scales.

Fiber segments are classified as bonded to other fibers and non-bonded (free). The density of bonds has recently been a subject of intense discussion, in particular with the emergence of new imaging tools. Clearly the number of bonds that the fibers may form is finite. Indeed, the process of fiber conformation during paper creation can be described mathematically. One of the predictions, based on probabilistic and statistical formulation, has been suggested by Komori and Itoh¹ (1994). It is based on the effect of the *steric hindrance* (Ekman, Miettinen, Tallinen and Timonen, 2014), meaning that once two fibers become close enough to form a contact, there is no free space at this particular point in space for other contacts to exist. However, these methods are approximations that disregard the cross-sectional geometry of the fibers and cannot be applied directly on collapsed fibers, that is, fibers without lumens, or where the materials contain a mix of different cross-sections.

Another statistical derivation of the number of contact points (NCP_{pred}) in an isotropic two-dimensional (2D) network showed that when the maximum number of contacts is reached, every contact point is at a distance of one fiber width from the neighboring one (Alava, Niskanen, 2006; Pan, 1993).

Sampson (2004) predicted that the maximum possible number of contacts in a planar fiber network results in the

¹ The original paper by Komori and Makishima (1977) for an idealized network of fibers was criticized by Pan (1993), who suggested his own formulation stating that the mean number of contacts is defined by the fiber aspect ratio, i.e., the ratio of the fiber's length to its diameter.

number of contacts per fiber being limited to $4/\pi$ times the fiber's aspect ratio, the ratio of fiber length to its width.

Estimations of the number of fiber crossings in a three-dimensional (3D) random fiber network were proposed by Corte and Kallmes (1962) and Komori and Makishima (1977). We will test these predictions in the following sections.

The degree of bonding is an important factor in the micro-mechanical models of fibrous materials (Heyden, 2000; Hägglund, Isaksson, 2008; Kulachenko, Uesaka, 2012; Niskanen *et al.*, 1999; Strömbro, Gudmundson, 2008). The number of bonds in a given network structure can influence both the stiffness and the strength of the network (Borodulina *et al.*, 2012). Erroneous estimation of the number of contacts can hinder the analysis and potentially lead to wrong conclusions.

Not only fiber connectivity data but also the properties of the ultrastructure of paper have attracted growing interest in characterizing fiber networks. In turn, this has created a niche for developing image analysis methods for non-destructive wood fiber segmentation applicable to fiber-based materials. A number of partially and fully automated methods have been proposed (Dokládal, Jeulin, 2009; Marulier *et al.*, 2012; Wernersson *et al.*, 2009; Vigié *et al.*, 2013; Yamakawa, Chinga-Carrasco, 2010). Previously, we presented the methods of describing the internal characteristics of fiber networks in paper using X-ray tomography, adapted for estimation of the number of bonds in paper (see Wernersson *et al.*, 2014). In that work, we developed a fast semi-automatic method for extracting important properties from 3D micro-computed tomography (μ CT) images of sparse handsheets. The methods are based on manual demarcation of fiber centerlines. The demarcation procedure takes on average 1 minute per fiber using an intuitive graphical interface. Our approach is distinct from these studies, with the main advantage being the possibility of extracting the information from several measurements without a complete segmentation of the fiber network. Moreover, the developed algorithms work equally well for the complex fiber geometries – collapsed (without the lumen) and non-collapsed fibers – and for the fibers with pores.

In this paper, we extract and analyze the microstructure of two $1 \times 1 \text{ mm}^2$ low-grammage fiber networks in terms of fiber and network connectivity data. The work is based on the previously developed algorithms (Wernersson *et al.*, 2014). We merge the data from two analyzed samples to create a comprehensive set with 250 demarcated fibers. A few independent studies by different research groups have been presented lately (Malmberg *et al.*, 2011; Tan *et al.*, 2006; Vigié *et al.*, 2013). In the recent study by Marulier *et al.*, (2015), the influence of papermaking conditions in terms of fiber orientation, pressing levels, and drying conditions was analysed regarding the geometry of fibers and fiber-to-fiber bonds. We will compare the reported data with the derived quantities. To the best of our knowledge, our work is the first large-scale (regarding the number of fibers) study to investigate the structural characteristics of paper networks,

including estimation of fiber cross-sectional shapes, distances between fiber-to-fiber contacts, and relative contact areas, and to discuss the representativeness of the studied samples. The results presented in this paper are to be compared with the existing statistics on the number of contacts (Alava, Niskanen, 2006; Komori, Makishima, 1977; Sampson, 2009). The tube model (Toll, 1993), which accounts for the number of fiber crossings based on the orientation distribution, was excluded from the analysis since it is known to underestimate the experimental data (Marulier *et al.*, 2015). Furthermore, we propose a methodology for the density of contacts in 3D fiber networks. The results include geometrical characterization of fiber networks in terms of distributions and/or mean values with the intention of a direct data transfer to fiber network mechanical modeling and data verification.

Material and Methods

We worked with a laboratory (isotropic) handsheet with a basic weight of 27 g/m^2 that was made of never-dried, unbleached softwood sulfate pulp with removed fines (for details, see Borodulina *et al.*, 2012). The ultrastructural characteristics of the sample were acquired with Xradia MicroXCT-200 X-ray tomography equipment at Innventia, Stockholm, Sweden. We operated on two samples of $2 \times 2 \text{ mm}^2$ that were randomly sampled by cutting the handsheet using a sharp die. The two test pieces were mounted as a stack and scanned simultaneously, with a captured area of approximately $1 \times 1 \text{ mm}^2$ from each paper sample, shown in *Fig 1a* and *b*. The voxel size was set to $0.55^3 \mu\text{m}^3$.

Methodology

To analyze the computed tomograms of paper, we used an open-source software tool written for Matlab (MathWorks, Natick, MA, USA) that we had developed previously (Wernersson *et al.*, 2014). The code as well as a portion of our data set is available online (Wernersson, 2015).

Identification of fiber-to-fiber contacts is performed automatically in the reconstructed micro-computed tomography (μ CT) images. Fiber cross-sections (CS) are first identified in the images perpendicular to the fiber's centerline. The CS is then classified as free or bonded, mainly based on the cross-sectional area. If free, further statistics on the geometry of the CS are calculated.

Fiber cross-sectional geometry is quantified in the form of the width-to-height (WH) ratio. Most of the fibers have a collapsed cross-section that can be represented as a solid rectangle. For each CS image of the free fiber, we defined the orientation of the CS by the following procedure. We maximized the ratio between geometrical moments of inertia I_{xx} and I_{yy} , computed with respect to the center of mass of the individual CS. By using both geometrical moments of inertia and CS area, we computed the fiber width, height, and wall thickness for fibers that are not in contact.

Free fiber length. In our measurements we define the free fiber length (FFL), l , as a part of the fiber length or fiber

segment that is enclosed between two fiber contacts, also illustrated in Fig 2. There also exists another nomenclature, defining FFL as the distance b between the *centers* of fiber contacts, so that $b \geq l$. Thereby, the average distance between the adjacent contacts, b , can be determined by dividing the length of the fiber by NCP:

$$b = \frac{L}{NCP}, \quad [1]$$

where L is the fiber length.

The reciprocal of b gives us the number of contacts per unit fiber length (No./mm):

$$n_l = \frac{NCP}{L}. \quad [2]$$

c)

Fig 1 a) and b) Extracted 3D representation of the scanned samples. Each sample has a size of 2 mm × 2 mm with thickness, t , scanned parallel with a slight separation. Mean thicknesses are $t = 44.9$ and $44.5 \mu\text{m}$ for a) and b), respectively. c) Top view of one of the 2D slices from X-ray tomography scanner showing a random distribution of fibers; the bar indicates $500 \mu\text{m}$.

The total number of contacts for a sample is:

$$N = \frac{1}{2} \sum_{i=1}^{n_f} NCP_i. \quad [3]$$

where n_f is the number of fibers.

Fig 2 The two ways of defining the free fiber length: either as the distance between the middle of subsequent contact regions, b , or as the distance between the edges of the contact regions, l . We have used the latter definition in this paper.

Thickness of the paper sample. To calculate the paper thickness, t , from the 3D images, first, the volumetric image was separated into three cut samples. Then morphological closing (Soille, 2004) was applied on each sample to define its surfaces using a probe of radius $30 \mu\text{m}$. We define the thickness as the average distance between the upper and lower surfaces of each sample, also shown in Fig 3.

Fig 3 Sample thickness is defined using morphological closing presented in a 2D illustration of the cut sample with the cyan line defining the border of the sample. The scale is presented in pixels with 1 pixel being equal to 1.11 μm .

Results and Discussion

Measured characteristic properties

From the two test pieces (Fig 1a and b), denoted as “first” and “second” throughout the text, we have manually demarcated the centerlines of 250 fibers. Marked fiber centerlines are the only measure required for the automated post-processing methods presented in the first part of our work (Wernersson *et al.*, 2014). Representation of the sample in terms of a few basic measurements allows us to demonstrate the fiber distribution in space, as illustrated in Fig 4. The reconstructed 3D fiber network is used for visualization purposes only as the circular shape of the CS used in it differs from the actual one.

Fig 4 a) and b) Three-dimensional visualization of the demarcated fibers from two pieces of the test sample corresponding to Fig 1a and b. In the visualization, each fiber is defined by a fiber axis and the median of its inner and outer radii and shown as hollow cylinders of constant radii, based on Wernersson *et al.* (2014).

Variation between the test pieces

Fiber dimensional properties such as length and width are subject to natural variation. We investigated whether the extracted sample size is appropriate for quantifying the mean values of fiber properties by comparing the measures extracted from the two samples.

Prior to handsheet making, the pulp properties were characterized using image analysis tools applied to a highly diluted fiber suspension (Hirn, Bauer, 2006). Such tools are often called fiber morphology analyzers (FMAs), since they are frequently used for extracting basic pulp morphologies. However, FMAs are applied to the swollen fibers. Consequently, the reported fiber wall thickness and fiber width are larger than those in the final sheet. Furthermore, these tools do not report the fiber WH ratio, which is an important factor that can be either inherited from or affected by the papermaking process.

Fiber dimensional properties – both those from the FMA characterization and those calculated from the μCT images – are summarized in Table 1 in terms of mean values and standard deviations.

The distributions of the measured fiber length from μCT are presented in Fig 5, which reflects the dimensions of the cut samples of $1 \times 1 \text{ mm}^2$. This implies that the maximum length of straight fibers cannot exceed $\sqrt{2} \text{ mm}$ for the considered samples.

Fig 5 The distribution of fiber lengths measured from the μCT images.

The extracted CSs along the free segments of the fibers were analyzed in order to compute fiber width and height data. The examples of this analysis are presented in Fig 6, showing the extracted CS and the computed fit. The WH ratio is one of the parameters that control the structural thickness of the fiber network. We based our fit on the assumption of having a hollow rectangle that is geometrically identical to the original CS in terms of CS area and geometrical moments of inertia (I_{xx} and I_{yy}). We assume a fiber to be collapsed if its calculated wall thickness is greater than or equal to half of the fiber's height. In our sample, 46% of fibers had a collapsed CS. The algorithm works equally well for fibers with and without the lumen.

Fig 6 Data from fiber CSs: initial image (a) and a rectangle (width w and height h) fitted to it (b). The bar indicates 20 μm .

Table 1: Fiber properties (mean $\pm$ standard deviation) extracted from μCT images compared with those measured directly from the diluted pulp used for handsheets preparation.

	Directly from FMA	From μCT images
Fiber length (mm)	2.19 ± 1.68	0.61 ± 0.29
Fiber width (μm)	27.82 ± 9.63	21.73 ± 6.69
Fiber wall thickness (μm)	6.46 ± 3.70	3.41 ± 1.21
Fiber WH ratio	N/A	2.90 ± 1.78

Obviously, the fiber length measurements performed on the scanned images cannot be used directly due to the limited size of the considered samples. The algorithms for extracting length distributions of the full-length fibers from the limited size of the network are presented in the literature (Miettinen, Hendriks, Chinga-Carrasco, Gamstedt, & Kataja, 2012; Wernersson, 2014). However, the length statistics can be sampled directly from FMA measurements. The length distribution does not change significantly from wet to dry state, considering that most of the shrinkage occurs in the transverse direction of the fiber. Hence, the primary focus of the developed methodology was the extraction of the complementary information about fiber CS properties in the dry sheet from μCT . The difference between width data measured from fiber suspension and those calculated from μCT is demonstrated in Fig 7.

One expects a transversal shrinkage of the fiber CS at a level of 15–40% (Alava, Niskanen, 2006; Niskanen, 1998). Assuming that FMA measures true fiber width, we conclude that our unbleached softwood sulfate pulp CS shrinkage was about 20%, as indicated by the results in Table 1. The calculated WH ratio and shrinkage factor can be readily used for data rescaling in numerical simulations of the networks in 3D. This information is also valuable for changes in the network structure, for example, due to refining.

Fig 7 Distribution of fiber widths measured by fiber analyzer (FiberMaster) and computed from μCT images.

A common limitation, when imaging samples for further analysis, is the size of a test piece (see, for example, He, Batchelor, & Johnston, 2004; Viguié *et al.*, 2013). It is natural to expect variations between the test samples, and therefore the quality of statistical data that is able to be represented is questionable. We checked the variability between the two samples in terms of extracted quantities.

The level of variation between the two cut samples (with mean thicknesses of 44.9 and 44.5 μm) is presented in Fig 8. While the results show discrepancies in measured fiber length values, the fiber width statistics are repeatable and thus representative, and therefore they are more relevant for the presented methods.

a)

b)

Fig 8 Effect of the variation in measured parameters: a) fiber length and b) fiber width, between the two corresponding test samples, denoted as "first" and "second".

It is vital to take into account the fiber orientation distribution, since it affects the number of contacts in a given volume (Toll, 1993). In our handsheets, the fiber orientation was close to isotropic in both samples. The orientation was estimated as follows. For each fiber, i , the end points $\mathbf{p} = \mathbf{f}_i(0)$ and $\mathbf{q} = \mathbf{f}_i(l_i)$ were chosen. In this manner, the vector $\mathbf{c} = \mathbf{p} - \mathbf{q}$ is a linear approximation of the fiber between the end points. From the linear approximation we obtain a unit length direction vector, $\mathbf{d} = \mathbf{c} / \|\mathbf{c}\|$. Since fibers have no specific direction (no head or tail), the inverse direction $-\mathbf{d}$ is also plotted for each fiber (see Fig 9). The isotropic fiber orientation means that the ratio between the radii of the outer and inner semi-axes is reasonably close to 1, and this assumption holds for both samples. We expect no influence of fiber orientation on the statistics concerning the number of contacts that we computed.

Fig 9 Distribution of radial fiber orientations in-plane for the "first" (shown in red) and "second" (shown in blue) test samples, normalized by the number of fibers in each sample. The numbering of the test samples corresponds to Fig 8.

Distance between the fibers crossing in-plane

Since we use optical measurement methods, restricted to resolving the distances of half the wavelength of light, one should bear in mind that any derived quantities lie within that limit. Hence, the resolution of microtomography images is far from the range of molecular contact or van der Waals force, which becomes important at sub-nano levels. In view of the ongoing discussion about the mechanisms holding fibers together in a dry state (Hirn, Schennach, 2015), it is therefore impossible to use current X-ray tomography technology for evaluation of the quality of the bonds. Therefore, the information about whether the overlapping fibers are indeed bonded, just in contact, or at a distance within the resolution is inaccessible.

One of the parameters that characterize the fiber network is the distance between the fibers that appear to cross in-plane, that is, that cross in projections on the XY-plane, represented by the top view, as in Fig 1c. The statistics of these distances provide information about the spatial arrangement of the fibers in a 3D network. In the calculation procedure, the distance between the fibers is estimated as the distance between the outer surfaces of the fibers (fiber cell walls) (for details see Wernersson *et al.*, 2014). We evaluated the shortest distance d between the pair of fibers crossing in-plane. The distribution of the distances is illustrated in Fig 10.

We estimate that any legitimate fit to the given distribution, extrapolated up to the point of the intersection with the Y-axis, referring to Fig 10, will provide us with information about fiber connectivity below resolution. This is true since bonded fibers have zero distance between each other.

The generalized Pareto fit (Eq 4) was chosen among several distributions based on the minimum of the Akaike values (Akaike, 1974) that satisfy the goodness of fit to the data. The other three distributions that we tried with the corresponding increasing Akaike values are the gamma, Weibull, and exponential distributions respectively.

$$f(\mathbf{x}|k, \sigma, \theta) = \left(\frac{1}{\sigma}\right) \left(1 + k \frac{\mathbf{x} - \theta}{\sigma}\right)^{-1-1/k} \quad [4]$$

We now depict the slope of the function $f(\mathbf{x}=0)$, which gives a pdf of 0.13 at the intersection with the Y-axis in Fig 10. This value gives us the *probability of a contact* assuming bonded fibers with zero distance between them.

It is interesting to observe that the calculated probability of a contact underestimates the number of estimated contacts (represented by the gray bar). The explanation for that is the fact that the fibers are pressed and the CSs effectively deform, which can be interpreted as having a negative distance between the fibers. Therefore, extrapolating the data to zero will lead to underestimation of the number of contacts. This distribution of the distances can be used, however, in verifying the artificially generated networks to ensure a proper spatial arrangement of the fibers together with the contact estimations.

Free fiber length (FFL)

The free fiber length (cf. Methodology section) is yet another parameter that can be used for characterizing the differences between the networks (Alava, Niskanen, 2006; Borodulina *et al.*, 2012; Hawinkels, 2009; Marulier *et al.*, 2015; Sampson, 2009; Strömbro, Gudmundson, 2008). It depends on the density, fiber dimensions, and the thickness of the network, as both free surfaces affect the FFL statistics, also known as the free span distribution. We calculate FFL, denoted by l , as the segment of the fiber enclosed between two contact regions (as in Fig 2), which means that the zero distance corresponds to the case when fibers are stuck right besides each other.

Fig 10 Distribution of the distances between the fibers. The influence of bonded fibers – corresponding to zero fiber-to-fiber distance – is shown as a gray bar.

Fig 11 Calculated distribution of the free fiber length from μ CT images of two samples.

The measured distribution of FFL from μ CT of our samples is presented in Fig 11.

A negative exponential distribution fit for the frequency of free fiber lengths, presented in Fig 11, was chosen based on Kallmes and Bernier (1963). The quality of the fit is similar to that presented for the data of Kallmes *et al.*, who performed

both optical measurements of the FFL on surface layer fibers of a multi-planar sheet and numerous Monte Carlo simulations and concluded that a negative exponential distribution of the FFL was preferable. Furthermore, this distribution holds for a 2D, almost completely bonded sheet; this 2D assumption defines the probability of any point placed on a line of fiber length being uncovered by the contact with other fibers. Additionally, the exponential distribution has been shown to hold for generated 3D networks (Ekman *et al.*, 2012).

Similarly, He *et al.* (2004) performed an experimental study of the microstructure of paper with the help of confocal microscopy and showed a shape of the FFL distribution comparable to the one presented in Fig 11. Free fiber length is a local property since it is very unlikely to be above 20 μ m (being only a few percent of the fiber length); therefore our sample size of $1 \times 1 \text{ mm}^2$ is large enough not to cause any considerable bias.

The results presented in Fig 11 can be explicitly used in formulas, for example, in estimation of the average strain of the fiber, based on FFL and total fiber length, as described in Strömbro and Gudmundson (2008). The distribution of FFL can also be integrated into any micro-mechanical models of paper, regardless of the implied definition of FFL, and serve as yet another verification parameter for reconstructing the fiber networks. Free fiber length can be calculated as a distance between the *contacts* (l) or between the *centers* of the contact regions (b) as illustrated in Fig 2. While the interpretation of FFL is essentially different in these two cases (depending on whether the contact is modeled as an area or as a point), the distributions (of either l or b) have been shown to be unaffected by their definition (Kallmes, Bernier, 1963). Likewise, the distribution of the free fiber length l would give a similar FFL distribution of b .

Since the connectivity of a network affects FFL, which is also relevant for the strength of paper, it is vital to have a quantitative measure of how many inter-fiber bonds a sample contains.

Number of fiber-to-fiber bonds

The total number of contacts in a handsheet sample depends (among other things) on its size and basic weight (or fiber network density). It is also a function of the volumetric concentration of the network and the aspect ratio of the fibers (Sampson, 2009). However, the mean number of contacts *per fiber* is not a unique measure with cropped fibers in contrast to the number of bonds *per fiber length*.

When comparing any estimation with theoretical predictions, it is essential to distinguish between 2D random networks of fibers and 3D networks, such as our samples. For example, the equation for density in 3D is straightforward and is characterized by the mass of fibers (m) per unit network area (S) and network thickness t (Eq 5a). The volume of an individual fiber is based on its length and the shape of its cross-section (either collapsed or with as a hollow rectangle,

as shown in Fig 6b) with area A_{fiber} . We assume fiber density (ρ_{fiber}) of 1442 kg/m³. In contrast, there is no “true” thickness for a 2D network; it can be defined only by the mean fiber diameter. The density in a 2D network can instead be substituted by a linear density (Eq 5b), with L_{tot} being the total length of the fibers.

$$\rho_{3D} = \frac{\sum_{i=1}^{n_f} m_{fiber}^i}{St} = \frac{\rho_{fiber} \sum_{i=1}^{n_f} L_{fiber}^i A_{fiber}^i}{St} \quad [5a],$$

$$q_{2D} = \frac{L_{tot}}{S} = \frac{\sum_{i=1}^{n_f} L_{fiber}^i}{S} \quad [5b].$$

Table 2 Comparison of the calculated density values (ρ_{3D}) for 3D networks and their linear equivalent (q_{2D}) for 2D networks estimated by μ CT methods and using Eq 5 a and b.

	First sample	Second sample
ρ_{3D} , kg/m ³	300.8	327.9
q_{2D} , mm/mm ²	61.8	71.1

We will first relate our μ CT measurements with currently available knowledge on the number of bonds in paper for 3D networks and then compare them with 2D networks (Alava, Niskanen, 2006). We retain the existing notation and elaborate upon our results accordingly. The total number of fiber crossings (NFC) in volume V for a 3D random fiber network in equivalent form has been independently derived by Corte and Kallmes (1962) and Komori and Makishima (1977) (Eq 6):

$$NFC = \frac{\pi}{2} n_f^2 \frac{r_f \bar{\lambda}^2}{V} \text{ for } V \gg \bar{\lambda}^3, \quad [6]$$

where r_f and $\bar{\lambda}$ are the mean values of fiber radius and length respectively. The results of the calculation are presented in Table 3.

Table 3 The total number of contacts in a 3D sample estimated by μ CT methods and using Eq 6.

Number of contacts	First sample (96 fibers)	Second sample (145 fibers)
3D (Eq 6)	1421	1979
3D (μ CT)	1220	1133

Since 2D models of the network structure are sufficient in many applications, we would like to couple our measurements to planar networks as well. Taking the

arithmetic mean of the μ CT calculated number of contact points for each fiber (NCP_i), the average number of bonds estimated per fiber is 22.6. Interestingly, the number of free fiber segments can be less than the number of contacts as in some cases parts of the fibers were continuously bonded, leaving no free distance between the detected bonds (see Fig 2).

Among the results obtained from μ CT, besides NCP_i , the fiber length and width can be used to predict the number of contact points in 2D (NCP_{pred}) directly from the fiber aspect ratio. A comparison between NCP_{pred} and the values of NCP_i is shown in Fig 12.

Fig 12 Theoretical prediction of the number of contact points NCP_{pred} , estimated as the ratio of fiber length to its width (fiber aspect ratio) plotted against the calculated NCP_i acquired from μ CT results. The agreement between two quantities (NCP_{pred} and NCP_i) is found by the linear fit with the calculated Bravais-Pearson's correlation coefficient (r) as $0.9530 \leq r \leq 0.9575$ (95% confidence level).

As can be seen in Fig 12, the average number of fiber-to-fiber contacts is around 30. The value of the correlation coefficient r indicates a nearly perfect positive linear relationship (with the 95% confidence level being $0.9530 \leq r \leq 0.9575$). Since the calculated values of NCP_i correspond well with the number of contacts predicted by the fiber aspect ratio (NCP_{pred}), Fig 12 signifies that almost a complete association between the calculated (3D) and theoretical predictions (2D) has been reached. This implies that the number of fiber-to-fiber contacts in 3D networks is controlled by fiber geometrical parameters assuming isotropic orientation of the fibers. For example, experimental observations of the bundle of fibers under compression (Latil *et al.*, 2011) confirm that fiber-to-fiber contact occurs at a distance of $145 \pm 7.5 \mu\text{m}$ with a fiber diameter of $150 \mu\text{m}$, which is also similar to the reported data of Marulier *et al.* (2015) for the mean distance between the contacts, which they found to be $35.8 \mu\text{m}$ with a mean fiber width of $30 \mu\text{m}$ in isotropic specimens of 40 g/m^2 . Furthermore, our results correspond well to the range of the number of fiber-to-fiber contacts reported by Page (1969), who predicted it to be between 10 and 40 for paper sheets of medium strength.

If, instead, we make a transition from a real 3D sample to a 2D network by analyzing the projections of the fibers into a plane, we can estimate the total *potential* number of contacts. In such a case, we get an expected overestimation of the number of contacts (since all the fibers whose projections cross in-plane do not necessarily connect with each other in 3D). The numbers of calculated contacts in 2D are 1260 and 1687 for the first and second samples respectively (cf. the results for 3D in Table 3).

The results indicate that the assumption that a contact will appear every width of the fiber is a reasonable expectation for a given fiber length and width in 3D. The number of contacts in the 3D sample per 1 mm of fiber length fluctuates between 23 and 36. Moreover, the calculated mean NCP in our sample (for both 2D and 3D) does not exceed the upper limit of $4/\pi$ times the fiber's aspect ratio estimated by Sampson (2004).

Contact density. The structure of fiber networks can be characterized by the density of contacts, that is, the number of contact points per unit volume or unit area of the sample. We calculate the contact density of the two test layers by gradually increasing the area from the center to the corners. The results presented in Fig 13 show that the results from both measurements approach the same values, having less than 1% difference between the two data sets, and show a relatively small downward trend. This suggests that the selected size can be representative of the density of contacts.

Fig 13 Contact density, calculated as the total number of contacts (Eq 4) per area of the sample. The two test samples (denoted as "first" and "second") correspond to the numbering presented in Fig 8.

Contact area. A total of 2710 contact areas have been calculated with Algorithm 3 (Wernersson *et al.*, 2014). The algorithm is based on continuous graph cuts with the following calculation of its minimal area, which separates two centerlines of the bonded fibers built by the max-flow algorithm, where a continuous maximum flow from the source location s to the sink location t is simulated (Appleton, Talbot, 2006). The surface area is defined as the minimal cut between the center lines and is found using the marching

cubes algorithm (Lorensen, Cline, 1987). We use the term "relative contact area (RCA)" as the fibers may only form a partial contact due to the fact that the surfaces with a distance of less than half of the resolution appear to be bonded. The distribution of the results is presented in Fig 14.

Fig 14 Distribution of the calculated bonded areas.

For the never-dried unbleached softwood sulfate pulp used in this work, the calculated RCAs are in the range of 50–2000 μm^2 . Similar results of 500 to 3000 μm^2 have been reported for unbleached softwood kraft pulp (Kappel *et al.*, 2010; Hirn *et al.*, 2010), while a range of 1000 to 3000 μm^2 has been reported for unrefined pulp (Schmied *et al.*, 2013). Such a spread of data is known to be due to the differences in the fiber morphology. Moreover, the RCA data presented in Fig 14 are probably the highest estimate for the contact area and will decrease with increased resolution of the method. The actual area that might be in contact is significantly lower due to several factors including the final roughness of the fiber surface. At the same time, the entanglement due to external fibrillation cannot be assessed at the resolution used.

Conclusions

Methods for the analysis of μCT images, presented in the first part of this work (Wernersson *et al.*, 2014), have been applied to collect information about fiber morphologies and connectivity data in an isotropic handsheet made of never-dried unbleached softwood sulfate pulp with fines removed by considering two test pieces with nearly 250 manually marked fibers. The results obtained from μCT include the fiber length, width, height, wall thickness, network density, estimated number of contact points, and free fiber length statistics. The control of fiber orientations demonstrated the isotropy of the considered samples. The main conclusions of this study are as follows:

- The transversal cross-sectional shrinkage of the unbleached softwood sulfate pulp from completely wet state to the dry condition in the handsheet was about 20% as estimated by comparing the fiber morphology data in the wet state with microtomography data acquired from the dry sheets.

- The size of $1 \times 1 \text{ mm}^2$ is representative for the evaluation of fiber cross-sectional dimensions; moreover, the contact density does not vary significantly between the samples of this size.
- The number of contacts is controlled by the fiber aspect ratio, indicating that a contact occurs every width of the fiber for a given fiber length and width. This has been confirmed by the number of contacts calculated from μCT measurements of isotropic low-grammage handsheets and is consistent with analytical expressions based on the aspect ratio of the fibers. Furthermore, the results are in good agreement with reported experimental data on the distances between the contacts. The influence of fiber orientation and network density is beyond the scope of this work.
- The presented methods can be used for collecting all the required information needed for reconstruction of the fiber network using, for example, deposition techniques (Kulachenko, Uesaka, 2012) or for verifying the methods based on physical modeling of the paper-making process (Lavrykov *et al.*, 2012; Lindström *et al.*, 2009; Lindström, Uesaka, 2008)

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