3-D computational simulation of paper handsheet structure and prediction of apparent density

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ABSTRACT: To obtain a better understanding of the influence of fiber morphology on paper properties, a novel 3-D computational simulator of paper structure was developed and validated by experimental work. Depending on the wood species, the dimensions of the fibers may vary by a factor of two or three, which directly influences paper structure and properties. Using known fiber size distributions, the model creates virtual handsheets made of these pulps that can be visualized, manipulated, and qualitatively and quantitatively compared to real handsheets. For a given basis weight, the model calculates the number of fibers in the sample and the main textural properties: thickness, apparent density, porosity, and roughness. It also computes the relative bonded area and the apparent specific surface, which are important factors for assessing mechanical or transport properties. This simulator can be used to predict, in a very short time, the variation of a property as a function of changes in one morphological parameter such as fiber length.

This paper presents the main principles of the simulation and the results for one global textural property, apparent density. A prediction of tensile breaking strength will be presented in a subsequent paper.

Application: This computational simulator enables a better understanding of the influence of fiber morphology on the three-dimensional structure of paper and some of its physical properties by generating, in a very short time, a virtual handsheet that can be manipulated, visualized, and analyzed.

Investigations of fibrous network structures generally aim to obtain a better understanding of process mechanisms or of the effect of raw material on these structures. Various experimental methods have already been used to characterize such structures. In the early 1980s, scanning electron microscopy (SEM) became a widely used technique for examining the surfaces and cross-sections of samples. Use of image analysis in combination with SEM made possible quantitative determinations of, for instance, porosity or pore size. More recently, X-ray microtomography has been successfully used to characterize 3-D structures [1–2].

Another way to study the structure of a fibrous network is to use computational modeling. Some models already exist, but most of them are limited to two dimensions [3–6]. Nowadays, the statistical geometry of two-dimensional random fiber networks is well understood. It provides excellent information on paper formation and local basis-weight distribution. However, because 2-D models do not consider network thickness and fiber conformability, they cannot provide information about the bulk structure or roughness of the material. In fact, it is the three-dimensional structure that directly controls the density, porosity, permeability, and some optical properties. It also plays an indirect role in determining mechanical properties through its influence on relative bonded area. For these reasons, only a 3-D model can predict the properties of a paper sheet. Several models exist that take three dimensions into account [7–9]. These models provide qualitative or relative results for relationships such as the effect of fiber flexibility on porosity. Unfortunately, they are not precise enough. They either allow too great a degree of fiber interpenetration or fail to consider the heterogeneity of fiber morphology. Moreover, no comparison with real samples has been done. Better quantitative modeling will require more elaborate microscopic models [9].

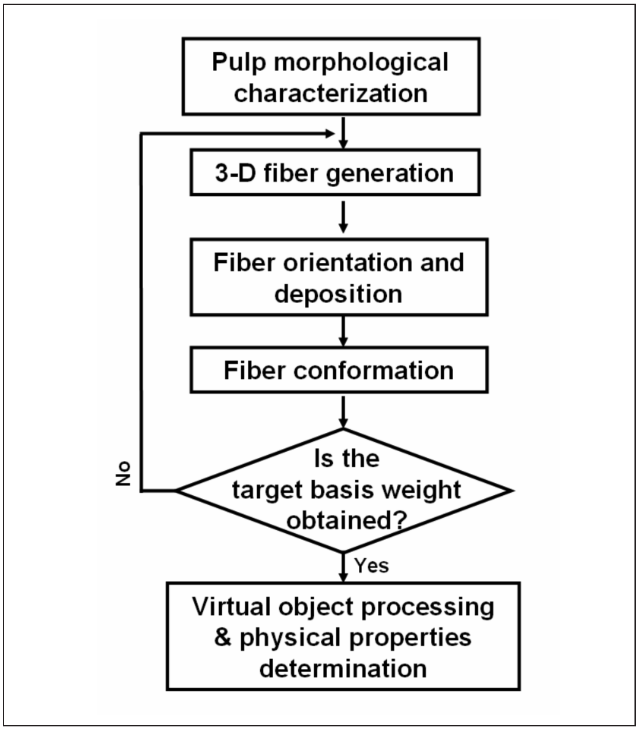
The aim of this article is to present a new 3-D model of a fibrous structure that has been applied to handsheets. This model limits the degree of fiber interpenetration by a precise meshing of the fiber elements. It makes it possible to introduce the real heterogeneity of the fibers in a paper sheet and provides quantitative results that can be compared to physical measurements.

This paper first describes the general principles of virtual network generation, then explains the modeling of the individual fibers, which is a key element of the model. Finally, the authors present the computation of the physical properties of the network and the comparison of simulation results with real data.

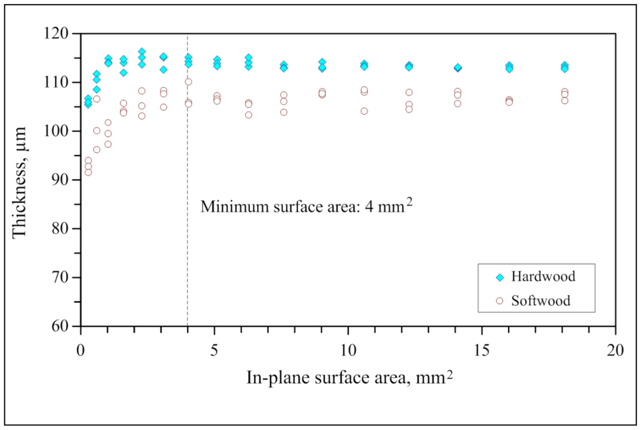
GENERATION OF A 3-D HANDSHEET

General principle of the computational model

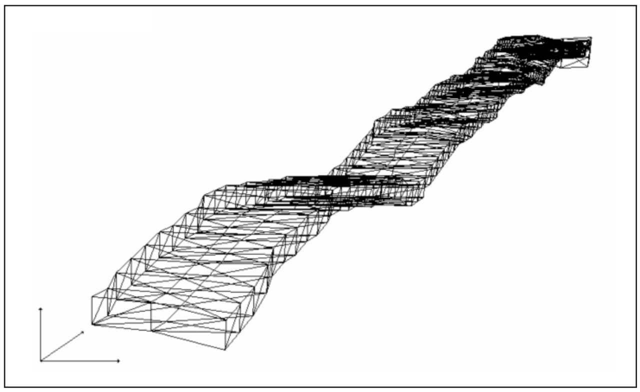
Papermaking is a succession of complex physical operations in which a web is formed by filtration of a dilute suspension of fibers and then consolidated by means of pressing and drying. To simplify the problem, only the final product is taken into account. Considering the filtration of a very dilute suspen-



1. Flowchart of the 3-D computational model of paper.



2. Influence of the in-plane surface area on thickness for a 60 g/m² sample.



3. Meshed fiber in the virtual network.

sion, it can be assumed that each individual fiber is deposited onto the underlying mat and thereafter deformed as a function of its moment of inertia and the underlying network geometry. Therefore, the computational simulation consists of a loop with three steps:

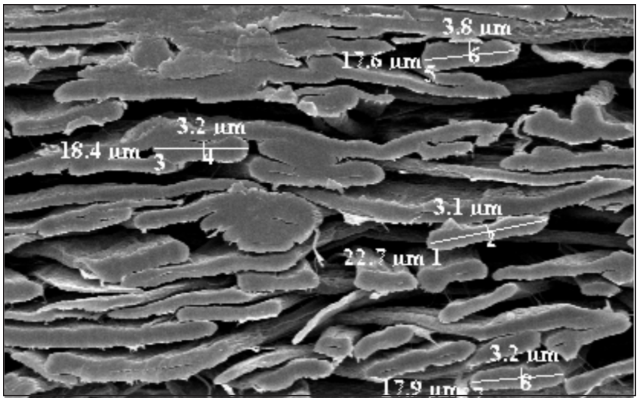
- generation of a fiber,
- fiber deposition,
- and finally, fiber conformation,

as illustrated by the flowchart in **Fig. 1**. The model inputs are the pulp characteristics, the sample size, and the desired basis weight. Its outputs are the structural properties and the virtual handsheet that can be visualized in real time. Thereafter, we can compute other properties, such as mechanical properties.

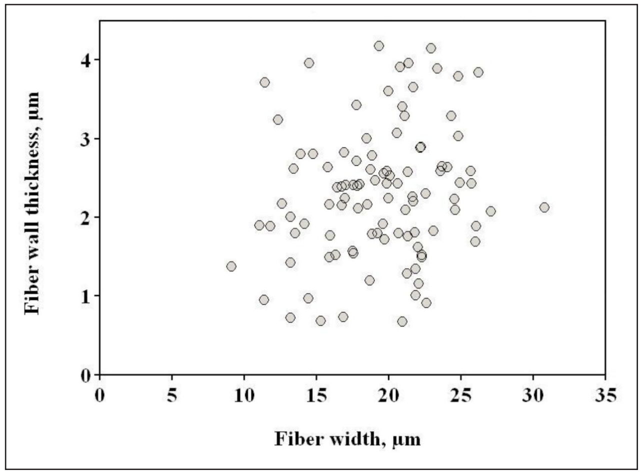
It should be noted that, before validating the model in terms of its capability to predict physical properties, its repeatability and the influence of factors such as sample size had to be tested. This testing is not the focus of this article, and therefore only a brief summary will be given here. Because many input parameters are partly governed by stochastic factors, the model repeatability must be checked. This was done by generating thirty simulated samples of various types of bleached kraft pulps from both hardwoods and softwoods. The worst repeatability error for the density prediction, i.e., the thickness at constant basis weight, was only three percent, which validates the repeatability of the model [10]. Another important factor is the sample size. In fact, the memory requirements for a 15-mm² network are approximately 500 MB. For reasons of computational time and memory size limitations, the size of a virtual sample should be as small as possible. Nevertheless, a minimum size is necessary to overcome the inevitable edge effects and to obtain stable values of the textural parameters, which can vary as a function of the heterogeneities of the structure. To analyze the effect of sample size, several square networks with different areas were generated using two types of pulps. It was found from the simulations that when the edge length is less than the average fiber length, the thickness values are affected, as shown in **Fig. 2**. For this reason, the area for determination of the physical properties of the computed samples was kept constant at 6 mm².

FIBER MODELING

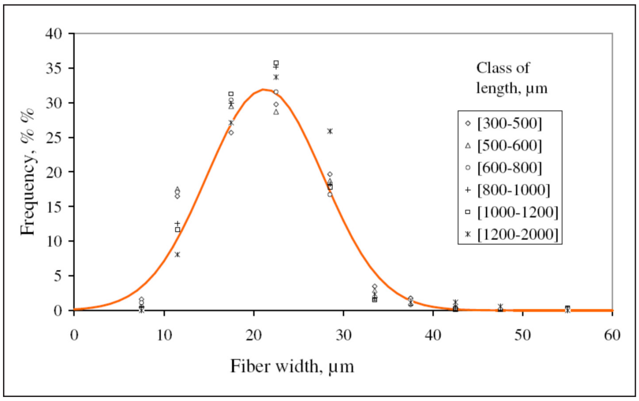
To take into account the heterogeneity of the fiber dimensions and to represent the fibers in a realistic manner in three dimensions, as illustrated in **Fig. 3**, it is essential to know precisely the dimensions of each fiber, i.e., its width, length, and wall thickness. Because the simulator computes the final structure of a handsheet, it is essential to know the dimensions of the dry fibers. Because lengthwise fiber shrinkage during drying is negligible, a MorFi fiber analyzer [11] was used to characterize the fiber length distributions. This apparatus also provides the fiber width distribution, a measurement of fiber curl, and an analysis of the small particles (fines, fiber debris, etc.). Because of the transverse shrinkage of the fibers during drying, the MorFi fiber width distributions are not used in the simulator. However, they will be analyzed, as explained in the next paragraph. The width and wall thick-



4. Measurement of the width and wall thickness of fibers (SEM image).



6. Wall thickness as a function of width for individual eucalypt pulp fibers.



5. Width distribution of eucalypt pulp fibers for various length classes and the average distribution.

ness distributions were determined from SEM images of the surface and from cross-sections observed at high magnification. The fiber cross-section of each individual fiber is represented by an equivalent rectangular area, which will be deformed during the fiber conformation process. **Figure 4** shows an example of fiber width and thickness measurement in a cross-section. In this picture, the thickness and width values are averaged values for each fiber. For these measurements, only the fibers that were perpendicular to the cross-section plane, or nearly so, were selected to avoid width overestimation. The agreement between the width distributions given by the measurements from the sample faces only and those obtained for the cross-sections was also verified. This study focused on the case of bleached kraft pulps, and therefore we assumed that the fibers were collapsed, as was observed on the images of the cross-sections. We further assumed that the fiber wall thickness was equal to half the fiber thickness.

To generate the fibers, it is essential to know whether these distributions are independent of each other. In other words, it is necessary to know whether the longest fibers are wider than the other fibers, and whether the widest fibers are thicker than the others. **Figure 5** shows the fiber width frequency distribution for various length classes, as obtained from the fiber analyzer for a eucalypt pulp. In this graph, the solid line represents the average frequency distribution of fiber width. It shows that the differences between the individual distributions and the average distribution are relatively small. The assumption that the width distribution for any length class is identical to the average distribution was further validated by a χ^2 statistical test for all the pulps used. It may therefore be concluded that there is no correlation between the length and the width of the individual fibers in a paper pulp.

The same type of investigation were also performed for the width and wall thickness distributions as determined by SEM image analysis. These analyses led to similar conclusions as those illustrated in **Fig. 6**, where the wall thickness of the individual fibers of the eucalypt pulp is plotted as a function of fiber width. There is obviously no correlation between fiber width and fiber wall thickness. Statistical tests, namely the calculation of the correlation coefficient and of the statistical significance of the correlation coefficient, made it possible to validate the assumption that there is no correlation between the width and the wall thickness of individual fibers for all the pulps used in this study.

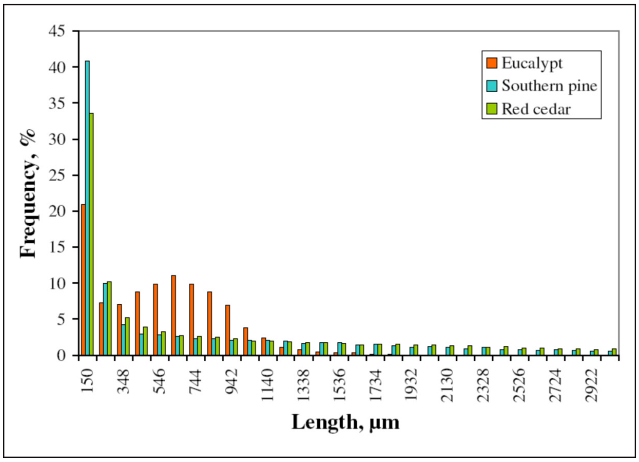
Figure 5 shows that the width distribution is approximately normal, which is in agreement with the results obtained by Deng and Dodson [6]. This is also the case for the wall thickness distributions. As a result, it was possible to replace the discrete distributions by continuous distributions in the model. In this case, the probability density of a given parameter can be expressed as:

$$f(x) = \frac{1}{\sqrt{2\pi} \times \sigma} \times \exp\left[-\frac{1}{2}\left(\frac{x-\sigma}{\mu}\right)^2\right] \quad , \qquad (1)$$

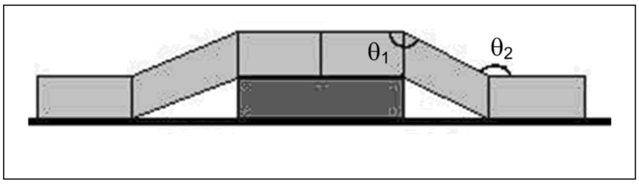
where μ is the mean value and σ the standard deviation of the parameter.

Conversely, it was found that the fiber length distributions (**Fig. 7**) could not be fitted with a simple mathematical function. Therefore, point values of the discrete fiber length distributions were introduced into the model.

The dimensions of each fiber were obtained by three random selections, the first from the table of the length distribution, the second and third from the continuous width and wall thickness distributions respectively, using the Box-Muller method [12].



7. Length distributions of eucalypt, southern pine, and red cedar pulp fibers.



8. Schematic illustration of the significance of fiber conformation parameters.

Negative or excessively large values were rejected by considering only the values in the range of plus or minus 3·σ.

To reproduce the real appearance of the fibers in a paper sheet, the fibers were given a wavy longitudinal shape by means of a polynomial function. For this purpose, researchers used the individual fiber curl measurements made with the fiber analyzer (MorFi apparatus). What is more important is that a dense meshing of the fibers is required for several reasons. On the one hand, it should be recalled that the fibers have widths generally ranging between 15 and 30 µm and that the fibers overlap in the network. On the other hand, it is desirable to avoid fiber-fiber interpenetration, which was one of the critical problems with simulations previously published in the literature. Therefore, the cross-section of each fiber is represented by a mesh with six points, which is repeated every three micrometers along the length of the fiber. Using six points in each cross-section makes it possible to deform the fibers across their width. Three micrometers is the lowest acceptable value for which a correct overlapping could be produced. It must also be pointed out that, for predicting some physical properties, it is necessary to determine accurately the value of the contact area, a problem which will not be discussed in this paper. Consequently, each fiber is modeled with two thousand points per millimeter, as shown in Fig. 3. This is acceptable in terms of memory capacity requirements and computational duration.

Papermaking pulps contain fines, i.e., elements that are not retained on a 200-mesh screen [13,14], of various origins. It is well known that the anatomical makeup of hardwood pulps is much more complex than that of softwood pulps. Generally, most hardwoods contain many more short parenchyma cells than do softwoods; they also contain vessel elements. Nevertheless, the percentage by weight of these components in hardwood pulps is extremely low. In addition, the pulp manufacturing process tends to cut some fibers, and therefore some fiber fragments are present in the fines fraction. Finally, the pulp preparation process, and more specifically the beating process, liberates secondary fines from the fiber walls. This type of fines will not be considered in the present paper, because only unbeaten pulps were used. The fraction by weight of fines and their geometrical dimensions were also determined. By extension, the term "fiber" will be used here for any pulp component, whether fiber or fine.

After being generated, each fiber must be oriented and deposited onto the network. Because the fiber suspension is sufficiently dilute to yield a crowding factor of approximately unity, as defined by Kerekes and Schell [15], no flocculation occurs. Therefore, the position of the center of gravity of each fiber and its in-plane orientation should follow random uniform processes. Nevertheless, it is well known that, during the forming and consolidation processes, the particles, and especially the fines, have a tendency to seal the most porous regions, leading to a gradient in the z-direction [16]. These concepts are included in the simulator, and therefore the process is not totally stochastic. The fiber deposition process consists of two steps: first, the position and orientation of the fiber are randomly determined, and second, the fiber is slightly moved, by a maximum of 100 µm, to fill the voids underneath most effectively. To reduce edge effects, the center of gravity of each fiber was allowed to lie in a larger square than that where its properties were computed, that is to say, with an edge length 1 mm longer (500 µm on each side).

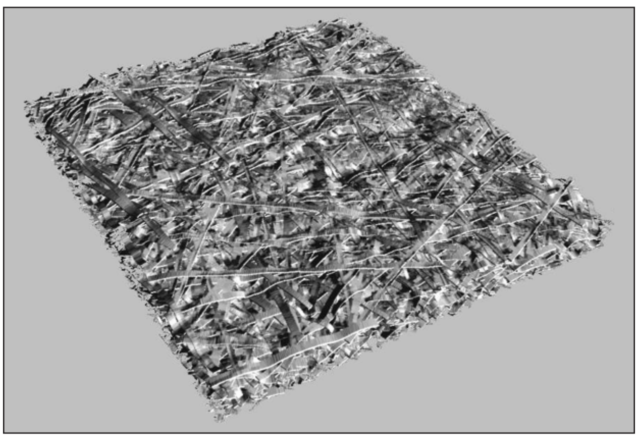
Then the fiber is deformed onto the surface of the underlying network. The conformation algorithm, developed for bleached kraft pulps, takes into account only the moment of inertia of the fiber, i.e., its morphology, and not the wood species. It should be noted that, because the forces acting on the network are unknown during the forming, pressing, and drying processes, two empirical coefficients, independent of wood species, were fitted to obtain correct density values for characterized laboratory samples. These coefficients represent the maximum deformation angle of a fiber at the crossing areas, θ_1, and in the free regions, θ_2, as illustrated in **Fig. 8**. In addition, it was verified that the pictures of the simulated networks resembled SEM images of plane surfaces and cross-sections (**Figs. 9–12**). The influence of fiber morphology on paper formation is illustrated in **Figs. 13–14**, which look like images of real handsheets.

QUANTITATIVE DETERMINATION OF PHYSICAL PROPERTIES OF VIRTUAL HANDSHEETS

All the model results are numerically calculated. The 3-D visualization of the virtual samples is used mainly for qualitative validation purposes. It can also be used to generate cross-sections that can be analyzed using stereometry concepts [17] to determine pore sizes or to compute transport properties of the network [18,19].

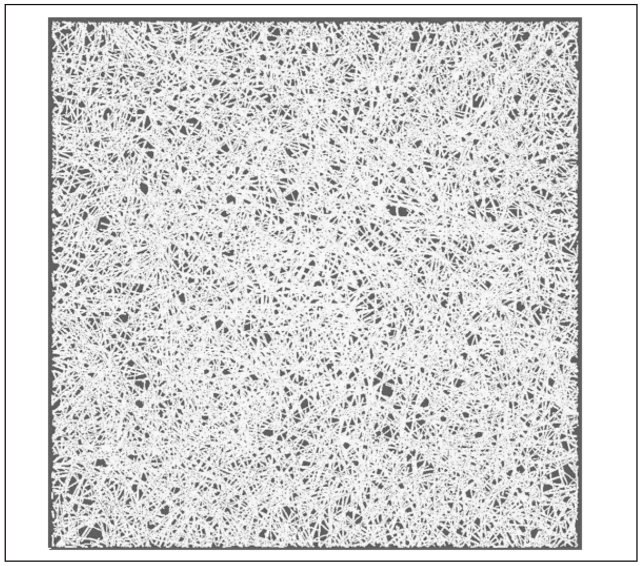
9. View of the surface of a simulated network.

10. SEM picture of a handsheet surface.

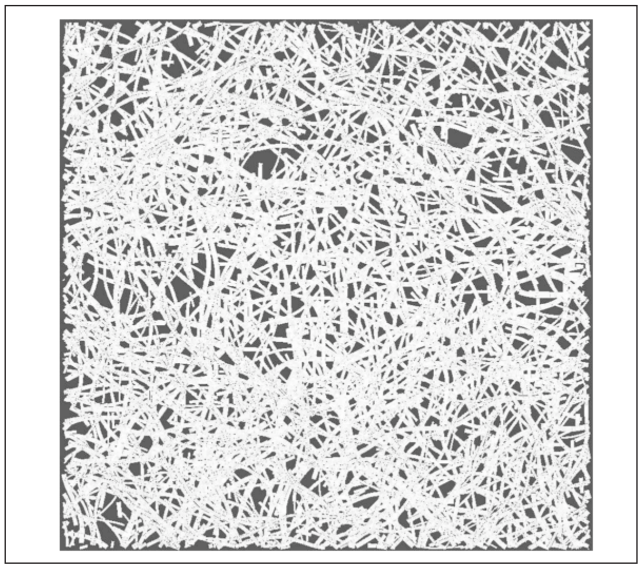


11. 3-D view of a 60-g/m² virtual handsheet, 2.5 mm x 2.5 mm.

12. View of a cross-section of a 60-g/m² virtual handsheet.



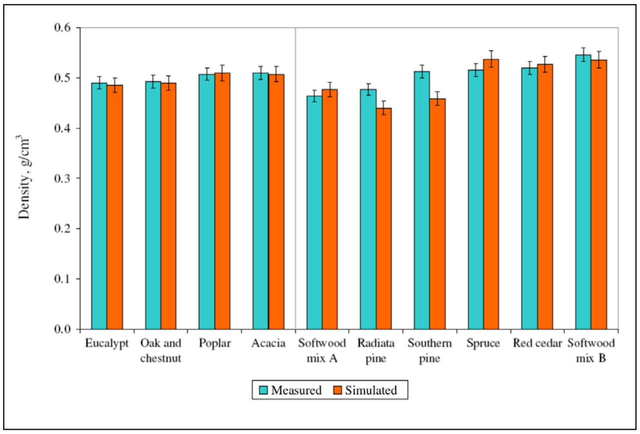
13. View of a 10-g/m² virtual handsheet sample made of hardwood pulp, 5 mm x 5 mm.



14. View of a 10-g/m² virtual handsheet sample made of softwood pulp, 5 mm x 5 mm.

This research focuses on two global properties of the paper, thickness, and density. The basis weight of the network is an input parameter to the computational program. When the fibers are generated, their volumes are known. Assuming that all the fibers are collapsed, which is valid for a bleached kraft pulp, and assuming that the density of the fibers is close to that of cellulose, i.e., 1.54 g/cm³, it is easy to calculate the weight of the fibers and to stop the program when the basis weight target has been reached. Moreover, knowing the position of every point on the fibers, it is possible to create a matrix containing the coordinates of the points of the top surface, which is linked to the roughness of the sample. Then the height of the highest point in the network, h_{max}, and the average value of the top surface height, $h_{topsurf}$, can be determined. Finally, the thickness of the sample, t_{sample}, can be calculated as follows:

$$t_{sample} = \frac{h_{topsurf} + 2 \times h_{max}}{3} \quad . \qquad (2)$$



15. Comparison of the densities of real and simulated handsheets.

This empirical formula implicitly includes the compaction of the network during standard caliper measurement. Moreover, neither the maximum height nor the average top surface height is individually representative of the average sample thickness. In fact, the maximum height overestimates the thickness and the average top surface value underestimates it, because of the asymmetry of the surface topography, which is revealed by study of the skewness of the surface roughness. Therefore, although it is imperfect, this empirical formula makes it possible to fit experimental results.

By definition, the apparent density, d, is the ratio of the basis weight of the sample, W, to its thickness:

$$d = \frac{t_{\text{sample}}}{W} \quad . \tag{3}$$

VALIDATION OF THE MODEL

Materials and methods

To validate the model and to analyze the effect of fiber morphology on paper structural properties, ten commercial bleached kraft pulps from both softwoods or hardwoods were used, which represent a wide variety of morphologies (long fibers, short fibers, fibers with thin walls, fibers with thick walls, etc.). The pulp samples were disintegrated and diluted, and then 60-g.m^{-2} laboratory papers were prepared using standard procedures [20]. Special attention was paid to the repulping process. The main properties of these pulps are summarized in **Table I**, where the values in parentheses are standard deviations.

RESULTS AND DISCUSSION

Figure 15 compares, for various density values, the results obtained for the real handsheets made of unbeaten pulp and for the virtual handsheets. In this figure, the error bars represent, on the one hand, the standard deviation of the experimental results, and on the other hand, the repeatability error of the simulation. As Fig. 15 shows, the density of the real samples varies within a narrow range from 0.46 to 0.55 g/cm^3, and there is no significant difference between the average results obtained for hardwoods and softwoods. These are not surprising results and can be proven experimentally. Figure 15 also shows that the density values are less scattered for the hardwood pulps than for the softwood pulps. This is most likely due to the narrower range of variation of the morphological properties of the hardwood species compared to those of softwoods.

What is more important is a good match between the simulated and experimental values. Indeed, the values are very close to each other. For instance, the average density of the real samples is 0.503 g/cm^3, with a standard deviation of 0.023 g/cm^3. For the virtual samples, values of 0.497 g/cm^3 for the average density and 0.033 g/cm^3 for the standard deviation were obtained, which are quite similar to those for the real samples. Moreover, the average relative difference between experimental and simulated values is 3.1%, with most values having less than 4% relative difference. Only one pulp (the Southern pine pulp) gives a slightly worse result (10.6% relative difference), which could not be explained. In fact, this pulp, which has thick wall fibers, produced handsheets with a relatively high density, contrary to mix A or the radiata pine fibers. The model developed here takes into account the moment of inertia of the fibers, and therefore it is

	°SR	Length µm	Width µm		Fiber thickness µm	
Hardwood pulps						
Acacia	15	737	17.5	(4.2)	4.1	(0.8)
Eucalypt	13	783	19.1	(4.2)	4.5	(1.6)
Oak and chestnut	14	709	20.4	(4.5)	4.7	(1.5)
Poplar	17	766	22.8	(5.2)	4.6	(1.6)
Softwood pulps						
Mix A	15	1868	37.8	(11.3)	5.9	(2.2)
Mix B	14	1925	33.6	(9.2)	4.6	(1.6)
Radiata pine	12	2214	34.5	(12.8)	7.1	(2.5)
Red cedar	15	2003	35.2	(9.2)	4.7	(1.6)
Southern pine	15	2056	37.9	(11.5)	6.5	(2.4)
Spruce	15	1558	35.0	(8.8)	4.7	(2.0)

I. Properties of the different pulps used (values in parentheses are standard deviations).

to be expected that thick wall fibers will generate low-density networks.

In summary, a comparison between the experimental results and the simulated results generated by the models implemented in the simulator developed here, in particular the fiber arrangement model and the fiber conformation model, reveals that it is possible to generate virtual networks that not only look like real samples, but also have similar properties. Recall that, in the models, only the fiber morphology is considered, and that the same conformation algorithm is used for all pulps with constant coefficient values for bleached kraft pulps.

CONCLUSIONS

Paper has a complex structure, and its physical properties depend on the properties of the raw material used. Assuming that the morphological properties of the fibers play an important role in paper properties, the authors developed a novel 3-D computational simulator of paper structure with the final objectives of predicting paper properties knowing the morphological properties of the pulp, of analyzing the influence of morphological parameters, and of optimizing fiber composition. This simulator makes it possible to generate a virtual object made of real fibers and assess some of its physical properties in a short computation time. In this simulator, the same algorithms were used for all fiber morphologies (long fiber, short fiber, etc.). Because of the dense meshing of the fibers, the accuracy obtained was much better than that of the models already reported in the literature. Problems such as fiber-fiber interpenetration were also avoided. Thus, it was possible to create virtual samples that resemble real handsheet samples.

To achieve the final objective, the simulator was also validated in terms of its capability to predict physical properties on the basis of experimental work. This article presents the first step of the validation process. This first step focused on a macroscopic structural property, sheet density. The validation consisted of comparing the physical results obtained for laboratory handsheets and the simulation results for unbeaten bleached kraft pulps for a large variety of fiber morphologies. A highly satisfactory level of agreement was obtained between experimental and simulated values. This made it possible to continue this study for other properties which involve microstructural parameters that are difficult to measure, such as relative bonded area. The results for breaking strength will be presented in another paper [22]. **TJ**

INSIGHTS FROM THE AUTHORS

Many fiber resources are available to the paper industry, and it is very well known that fiber morphology has a great influence on the structure and properties of the end products. Conventional methods for evaluating the papermaking potential of a pulp or a pulp mix or for optimizing fiber composition require the manufacture of laboratory samples and the execution of physical tests. Although it cannot completely replace experimental work, computational simulation may be a rapid method to assess properties and then to focus on specific experiments.

Investigations of fibrous network structures have been undertaken to obtain a better understanding of the process mechanisms or of the effect of the raw material upon these structures and their physical properties. Many studies have been carried out over many years on this subject.

The objective of this research was to develop a simple 3-D computational model that could be used on the one hand to visualize structure, and on the other hand to predict physical properties, taking into account the real heterogeneity of the fibers in a paper sheet. One specification was to be able to run the simulations on a personal computer and to obtain results in a short time. We validated all the development stages by experimental work.

The most difficult aspect of this research was certainly the development of a model that describes fiber arrangement and deflection in the paper structure. An accurate characterization of 3-D fiber morphology was also a challenge to achieve.

It was interesting to discover that macroscopic physical properties of handsheets of the tested pulps, which represented a wide range of fiber morphologies, could be predicted using a single model with a single set of coefficient values.

Future research will need to address the effect of refining or chemicals on property prediction, to generate a more realistic simulation of fiber orientation, and to predict additional properties.

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