CELLULOSE CHEMISTRY AND TECHNOLOGY

ANALYSIS OF CELLULOSIC FIBER MORPHOLOGY INFLUENCES ON MASS DISTRIBUTION UNIFORMITY IN TISSUE PAPER THROUGH STATISTICAL GEOMETRY

AFONSO HENRIQUE TEIXEIRA MENDES and SONG WON PARK

Department of Chemical Engineering, University of Sao Paulo, Sao Paulo, Brazil
✉ *Corresponding author: Afonso H. T. Mendes, afonso.mendes@usp.br*

Received August 3, 2023

Quality attributes of tissue products establish important differentials in the current scenario of fierce competition. Although the formation of the tissue paper is not monitored in industrial processes, it is imperative to recognize its importance for developing quality properties. A theoretical analysis of influences of the fiber morphology and furnish composition on the prediction of mass distribution uniformity in the paper forming process is presented. The theory utilizes the Poisson distribution for the grammage probability of points on the sheet, taken as a random fibrous network. The simulations revealed that lower grammage coefficients of variation could be achieved with total or predominant addition of eucalyptus fibers in mixtures with *Pinus* fibers. The results provide meaningful insights into the understanding of how the selection of fiber type and pulp blend could help in achieving suitable structural uniformity of the sheet, in the forming process, in view of the expected paper quality.

Keywords: eucalyptus, fiber morphology, formation, *Pinus* spp., review, statistical geometry, tissue paper

INTRODUCTION

The tissue paper sector represents one of the most promising sectors in the paper industry, in terms of global growth. Tissue paper production has grown steadily for decades[1] and expectations of continued expansion are confirmed by the strong increase registered in the second half of the last decade and in the forecasting of very significant annual growth rates for the coming years.[2-4] Tissue paper is a generic term for papers intended primarily for sanitary purposes, used as a substrate for manufacturing hygiene and cleaning products, destined for consumption in domestic and institutional segments. The industrial portfolio covers, *e.g.*, toilet papers, kitchen and hand towels, facial tissues and table napkins, whose properties may vary depending on the levels required in the application of the finished product.[5] Also included in this category are certain special products for industrial use in clinics and hospitals,[6] in addition to products for decoration and packaging.[7] In a scenario of fierce competition in a market of tens of billions of dollars,[2] with high demand for quality,[8] tissue paper manufacturers are continually dedicated to the development of new products to satisfy the growing quality demands from consumers. For this, the attributes of the tissue product and the optimization of costs, mainly in the selection of fibrous raw materials, represent essential success factors. The tissue paper is designed with a high technological degree, to incorporate particular physical and sensorial properties, such as softness, smoothness, specific volume (bulk), absorbency and strength, that is, primary characteristics that impact the functionality and performance of tissue products and represent key indicators of quality.[9]

Most tissue papers are manufactured from raw materials based on wood cellulosic fibers, especially bleached kraft chemical pulps, called BHKP (bleached hardwood kraft pulp) and BSKP (bleached softwood kraft pulp),[10,11] commonly produced with fibers from broadleaf (hardwood) and coniferous (softwood) trees, such as *Eucalyptus* spp. and *Pinus* spp., which are characterized by the predominance of short fibers (ca. 1.1 mm) and long fibers (ca. 2.8 mm), respectively.[12] High yield mechanical pulps, such as CTMP (chemithermomechanical pulp) and

BCTMP (bleached chemithermomechanical pulp), in addition to pulps produced from recycled fibers from paper waste and pulps from non-wood plant fibers from agricultural residues (*e.g.* wheat straw) or from cultivated plants (*e.g.* sugarcane bagasse), as well as from other sources of fibrous biomass (*e.g.* bamboo), have also been applied to the manufacture of tissue paper.[13-18] The mixture of cellulosic pulps, prepared with different raw materials, is usually adopted, due to specific quality objectives for the tissue product, fiber availability, cultural aspects, and cost control. Tissue products, with superior quality (premium products), incorporate chemical pulps from virgin fibers, in mixtures of short fibers (hardwood) and long fibers (softwood), with a higher proportion of short fibers, which contribute to intensifying the formation, the specific volume (bulk), and softness of tissue paper. Long fibers contribute to strength, which is desired to obtain good efficiency in paper manufacturing and converting operations, as well as the proper tissue product functionality.

Influence of formation on tissue paper properties

Although it is not usual to monitor and control formation in the tissue paper manufacturing process, *i.e.*, the degree of homogeneity of spatial mass distribution in the plane of nascent sheet, in the forming section of the paper machine, it is essential to highlight the relevance of this property to maximize the quality attributes of the paper and, consequently, the properties of finished tissue products. It is widely recognized, in literature, that the properties of paper are strongly influenced by its formation,[19-22] due to important impacts on physical and optical properties.[23] Good formation, which represents one of the fundamental properties for any type of paper, is of great importance for papers of low grammage, such as tissue papers, since, for such papers, an irregular distribution of fibers would critically affect their key properties, which may interfere in correct performance of tissue products during use.[24] Sheet formation influences several structural parameters of the paper, like the free-fiber-length distribution, pore size distribution, absolute contact states and fractional open area of the network.[25]

Strength characteristics, such as tensile, burst and tear strengths, and their interrelationships, represent an important requirement for the utility or functionality of tissue products to guarantee suitable performances, without rupture or disintegration during the use for which they were designed. Strength properties are also determinants of paper machine and converting line good runnability. Studies showing the influence of sheet formation on paper strength parameters have been published since the 1960s, *e.g.*, by Norman.[26] More recently, Nordström and Hermansson[27] published results of experiments carried out in industrial machines, as displayed in Figure 1 (a and b), in which, decreases in the relative tensile index (also called efficiency of tensile strength) were observed in view of the worsening of paper uniformity, as shown by the increase in formation parameter measured by the Ambertec method,[28] obtained by varying the fiber suspension consistency at the headbox, as shown in Figure 1 (b). The typical range of consistencies in the headbox of the tissue machine falls within 0.2-0.5%,[29] which can be associated with Crescent Former and Fourdrinier types of formers, respectively.

The relative tensile index (Fig. 1a) refers to the geometric mean of the tensile indexes in the MD and CD directions of paper produced in an industrial machine, in relation to the tensile index of laboratory sheets produced with the same raw material and similar densities. The geometric mean of the tensile strengths, in the MD and CD directions, has also been used to express tissue strength in trade-off studies with other properties.[30] For papers manufactured in industrial machines, the geometric mean represents an invariant with respect to the anisotropy of fiber orientation.[31]

A paper sheet with good basis weight distribution uniformity usually presents higher strength levels.[19] The formation inhomogeneity promotes the creation of areas of low strength in the sheet, which can be more vulnerable to breaks and the consequent detriment of the production efficiency and/or the functionality of the tissue product. In certain circumstances, papermakers resort to seeking chemical or process alternatives to obtain improved formation uniformities.[32] The latest tissue paper manufacturing technologies have supplanted the conventional dry crepe machine and evolved towards configurations with spatial control of mass distribution and sheet density, aiming to obtain greater softness without a severe loss of strength,[30] which reasserts the importance of evaluating the formation uniformity of tissue papers.[33]

The absorbency, which represents a special property of tissue paper, especially for the production of paper towels and napkins, is also influenced by the formation of the sheet,[34] given that the uniformity of the porosity and the arrangement of the pore space affects the performance of fluid absorption, as it depends on the uniformity of pore size distribution.[35] Figure 2 (a and b) shows the results of the work carried out by Dodson and Sampson,[36] in which they reinterpreted data from a previous study developed by Corte and Lloyd,[37] in order to propose a new statistical approach for the effect of the suspension crowding number (*i.e.*, its flocculation propensity) on the distribution of pore size, in which they observed that better levels of formation produce smaller pores and better distribution of pore sizes (lower coefficient of variation).

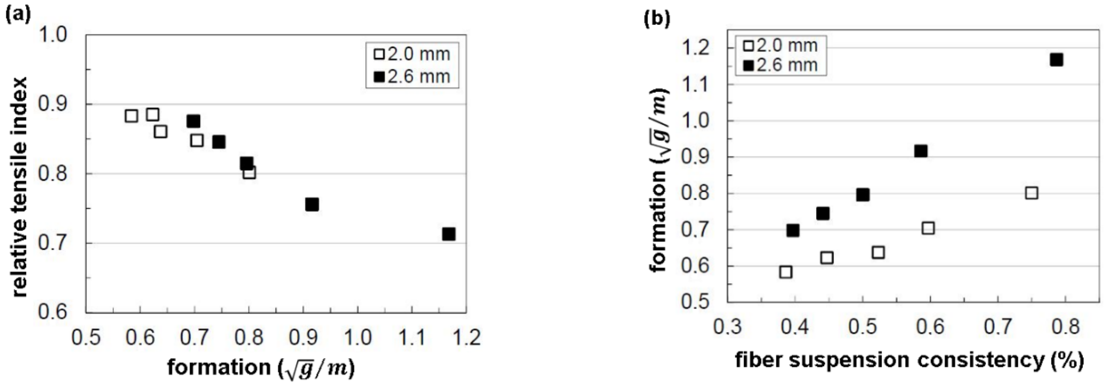


Figure 1: Effects of (a) paper formation on relative tensile index, and (b) of headbox fiber suspension consistency on sheet formation (adapted from Nordström and Hermansson[27])

This is an important feature for the fluid penetration process by capillarity,[36] as shown in Figure 2 (a and b) for raw materials containing long fibers (softwood) and short fibers (hardwood), respectively.

The crowding number values indicated in Figure 2 were determined from parameters reported by Corte and Lloyd,[37] considering fiber suspension consistencies in the range from 0.002% to 1%, to simulate different levels of formation – for details on the calculation of the crowding number, refer to, *e.g.*, Kerekes,[38] and for an approach to the influence of crowding number on formation uniformity of tissue paper, see the work of Mendes and Park.[39]

In addition to strength and absorbency properties, another important key feature of tissue paper is the softness, which is intrinsically associated to the creping operation. Good sheet formation contributes to adequate balance of tensile strength and stretch, and also for improved softness uniformity.[40-42] Raunio *et al.*[40] showed a significant correlation between the CD basis weight profile of tissue paper and the frequency of creping waves (Fig. 3), which has an important effect on tissue paper quality, particularly on softness. Creping frequency depends on the uniformity of the uncreped sheet. Increases in local grammage produce decreases in creping frequency, making tissue paper coarser.[33,43]

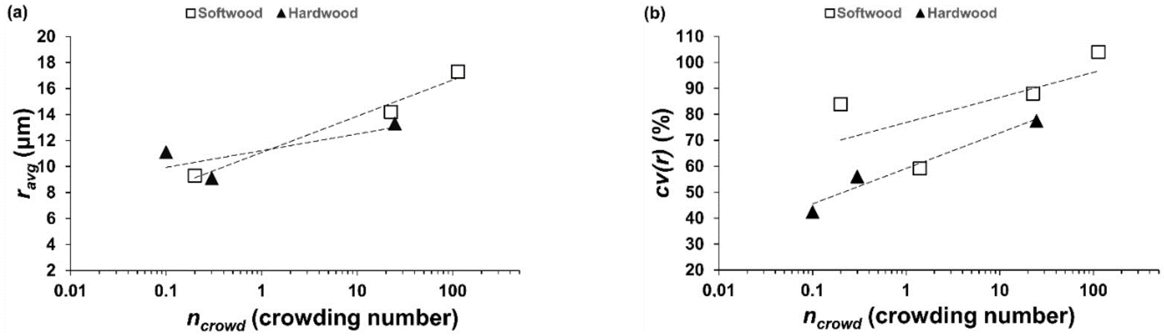


Figure 2: Effect of suspension crowding number on (a) estimated pore size r_{avg}, in the paper structure, and (b) on the coefficient of variation of pore size *cv(r)* (graphics created by authors from data obtained in Dodson and Sampson[36])

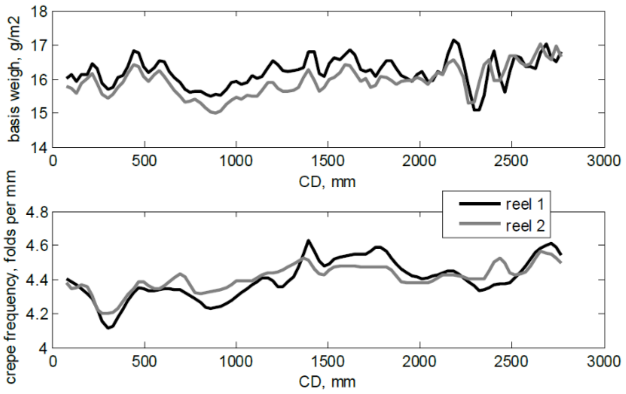


Figure 3: Basis weight and creping frequency profiles of sequential tissue paper reels[40]

Impact of fiber characteristics on paper formation

Among the factors that influence the formation of paper and, therefore, its structural characteristics and properties, there are intrinsic parameters of the cellulosic fibers used in the furnish composition, such as the fiber length, width (or diameter) and specific linear weight (coarseness).[27,44-47] These attributes vary according to the wood species and quantity present in the pulp formulation, which may lead to different coverage of fibers in each point of the sheet.

The initial structure of the tissue paper, generated at the paper machine former, can be modeled from the classic reference of a random network of fibers to evaluate its uniformity. The application of statistical models to estimate the influence of cellulosic fiber properties on the structural characteristics of a fibrous network favors for these properties to be uncoupled in a way that is not easily obtained in the laboratory.[48] The statistical geometry of a stochastic fibrous network, conceived in the works of Corte and Kallmes,[49,50] allows the exact calculation of some attributes of a random network by numerical determination.[51,52] The two-dimensional idealization properly describes the low grammage papers,[53] a condition in which tissue paper is inserted. Given that the thickness of the sheet, generally of the order of 50-100 μm,[54-56] is significantly smaller than its length or width and much smaller than the length of the fibers, the fiber network is flat and practically two-dimensional.[57-59] This type of structure, classified as a special case of random fibrous networks, meets the following conditions, as stated by Corte and Kallmes:[49,50] (a) the fibers are deposited independently of one another; (b) the fibers have an equal probability of being deposited at any point on the sheet; (c) the fibers have an equal probability of orientation, making any angle with respect to an arbitrary fixed axis. For modeling purposes, the above conditions are satisfied by two statistical distributions: (a) the location of the fiber centers on the supporting plane of the fibrous network presents a distribution according to the Poisson point process and (b) the orientation of the fiber axes, with respect to a given direction, shows uniform probability distribution.

Albeit it is known that, to a lesser or greater extent, machine-made papers deviate from a random distribution of fibers, the assumption of a random fiber network model have been used herein as reference structure of the tissue paper, given that even eventual weak deviations from randomness do not harm the central purpose of this work. The properties of a random fiber network provide basis worth of comparison for quantifying the properties of industrial paper. This assumption was also reasoned by: (a) previous theoretical and experimental studies, which have shown that the distribution of local grammage is insensitive to the changes in the fiber orientation, as mentioned by Sampson;[52] (b) the work of Mendes and Park,[39] which discussed the flocculation propensity of fiber suspensions used in tissue manufacturing, emphasizing typical modern paper machine operation conditions at very low consistencies at the headbox, which correspond to low fiber suspensions crowding numbers, particularly when considering *Eucalyptus urograndis* fibers, which are one of the protagonists of the current study, and (c) flocs or clumps break-up governed by proper flow turbulence provided by current headbox technology, combined with high-speed crescent formers and low propensity for flocculation, as mentioned previously, can suggest a minimization of self-healing or hydrodynamic smoothing effects during paper sheet forming.

Several studies show the effects of chemical pulps and mechanical pulps of virgin fibers and recycled fibers, including short fibers (hardwood) and long fibers (softwood), commonly called BHKP, BSHK, BCTMP, DIP (deinked pulp) and mixtures of these pulps, in different proportions, on the properties of tissue paper.[11,17,18,60-63] The studies present results of measurements of important characteristics, with emphasis on softness (superficial and structural), bulk, absorption, porosity and tensile and burst strengths, which represent key performance and quality indicators for tissue products, depending on the pulp composition (furnish) considered in the production of the base paper. However, those works are based on experimental plans that start from the materials (single or blended fibrous raw materials), to investigate their effects on the properties and performance resulting from the papers produced. This method, which traditionally corresponds to the cause-effect model,[64] the intermediate steps of the process – *e.g.*, formation of fiber network structure – are not detailed.

The present work aims to discuss the relationship between raw materials and the tissue paper structure, based on the phenomenology mentioned in the previous paragraphs, by applying the Poisson point process, as a structural uniformity modeling tool.[65] The objective is to provide theoretical insights into the influences of the morphological properties of cellulosic fibers and mixtures of different types of fibers on the perspectives of homogeneity of mass distribution in the fibrous geometric arrangement. The latter is established during the formation process of the nascent sheet of tissue paper, at the wet end of the paper machine, to highlight the need of proper selection of raw materials and mixing proportions of pulps in the production of tissue paper, for improved uniformity and better key quality properties of finished tissue products.

EXPERIMENTAL

Materials and methods

In order to evaluate the perspectives of mass distribution uniformity, in the paper forming process, the central aspect covered in this work was the distribution of probability of local grammages in the plane of the sheet, but focused specifically on tissue paper. To this end, raw materials, and pulp mixtures, commonly used in the manufacture of this paper grade, were considered to assess the influences of fiber morphological parameters and pulp blending ratios on the grammage distribution to, then, evaluate the resulting coefficients of variation. The theoretical procedure utilizes the Poisson distribution and the random fibrous networks concept for the above purposes.

Cellulosic fibers, pulp blends and paper grammages selected for analysis

Different cellulosic pulp formulations containing short fibers (hardwood) or long fibers (softwood) only, as well as other ones formed by mixtures of these fibers, in various proportions, were considered in the present study. Fibers of *Eucalyptus urograndis* and *Pinus taeda* were particularly selected, as they represent raw materials commonly employed in tissue paper production. The characterizing parameters of fibers contained in the reference pulps, were obtained from related literature. The experimental data previously published by Bassa and co-authors[66,67] were used in the simulations presented in this work, but all theoretical calculations involved were carried out by the authors. The average fiber dimensions assessed by those researchers were optically measured in aqueous fiber suspensions, using the Fiber Quality Analyzer - FQA (OpTest, Canada). The fiber length and width were obtained directly through the analyzer, while the fiber coarseness was found by the ratio between dry mass and total length of the fibers existing in the pulp sample analyzed. Mixtures of *E. urograndis* and *P. taeda*, consisting of fractions of 20% to 50% of *P. taeda*, were examined, as they correspond to compositions normally found in the tissue paper industry, which are defined according to the application of finished products intended for different market segments.[68,69] Table 1 summarizes the average parameters of the fibers present in the pulps of interest. In the sequence from A to F, reference pulps correspond to increases in mass fractions of long fibers from *P. taeda*, in mixtures with short fibers of *E. urograndis*. Uncreped papers with grammages of 13 g.m^{-2}, 21 g.m^{-2} and 33 g.m^{-2}, which were used in this work, correspond to those typically found at the Yankee cylinder, prior to the creping operation.

Modeling the structural uniformity of paper

The 'coverage' concept allows evaluating the mass distribution in the plane of the sheet of paper.[57,70] Coverage c is a stochastic variable, defined as the number of fibers covering a point on the sheet support plane. The average coverage $\bar{c}$ at any point in the plane, for a given total number of fibers N_f,, with length λ and width ω, over an area A, is expressed by:

$$\bar{c} = N_f \lambda \omega \,/A \qquad (1)$$

Since $N_f/A = \bar{\beta}/\lambda\,\delta$, where $\bar{\beta}$ corresponds to the average mass per unit area, *i.e.*, the nominal basis weight of the sheet, and δ represents the coarseness of fibers, Equation (1) can be rewritten as:

$$\bar{c} = \bar{\beta}\omega/\delta \qquad (2)$$

As the individual basis weight of fibers is given by $\beta_f = \delta/\omega$, it follows that:

$$\bar{c} = \bar{\beta}/\beta_f \quad (3)$$

The average coverage $\bar{c}$ can assume any real value, but the coverage in points c is a discrete stochastic variable and the probability *Pr* (*c*) that a random point, in the plane that supports the fiber structure, contains a coverage *c*, is defined according to the Poisson distribution.[51,71] In terms of the dimensional parameters of the fibers and a random fibrous network, *Pr* (*c*) can be expressed as follows:

$$Pr(c) = e^{-(\bar{\beta}\omega/\delta)}(\bar{\beta}\omega/\delta)^{c}/c! \text{ for } c = 0, 1, 2, 3 \,.. \quad (4)$$

The Poisson distribution has a specific property with respect to its variance, whose value is identical to its mean. Accordingly, for the coverage *c* distribution, $\sigma^2(c) = \bar{c}$, or:

$$\sigma^2(c) = \bar{\beta}\omega/\delta \quad (5)$$

The distribution of basis weight β can be found from *Pr* (*c*), given that:[51,65]

$$\beta = c\beta_f \quad (6)$$

Therefore, knowing from the variance properties, that $\sigma^2(c) = \sigma^2(\beta/\beta_f) = (\beta_f)^{-2}\sigma^2(\beta)$,[72] Equation (5) can be modified to the form below:

$$\sigma^2(\beta) = \bar{\beta}\,\delta/\omega \quad (7)$$

and then,

$$\sigma(\beta) = \sqrt{\bar{\beta}\,\delta/\omega} \quad (8)$$

Table 1
Fiber characteristics in cellulosic pulps obtained by mixing *E. urograndis* fibers with *P. taeda*, in different mass ratios[66,67]

Reference pulp	*E. urograndis* (%)	*P. taeda* (%)	λ (mm)	ω (µm)	δ (mg.m^{-1})	Population (10^6 fibres.g^{-1})
A	100	0	0.832	18.15	0.062	23.80
B	80	20	0.918	17.73	0.064	22.17
C	70	30	0.974	17.58	0.072	19.73
D	60	40	1.044	18.55	0.074	18.60
E	50	50	1.137	19.08	0.083	16.06
F	0	100	1.975	32.00	0.148	6.29

Symbols: λ = average length; ω = average width; δ = average coarseness

Statistical mass distribution

The coefficient of variation of the Poisson probability distribution for the basis weights in points over the paper sheet plane was used for a comparative analysis between the dispersions found in pulps containing different types of fibers and different proportions of mixture of these fibers. Equation (8) shows the expression used to characterize the uniformity of the intensity of mass distribution in points of the sheet. This statistical descriptor is also widely used as an index of formation of paper produced in industrial machines:[19,28,52]

$$cv(\beta) = \sigma(\beta)/\bar{\beta} \quad (9)$$

where $cv(\beta)$ is the coefficient of variation of the basis weight probability distribution in points β; $\sigma(\beta)$ is the respective standard deviation and $\bar{\beta}$ corresponds to the average mass of fibers per unit area, that is, the nominal basis weight of the paper sheet.

RESULTS AND DISCUSSION

Cellulose fibers have a wide range of dimensions, which depend on the wood species. Table 1 shows fibers of *E. urograndis* (hardwood), with an average length of around 0.8 mm and an average width of 18 µm, and fibers of *P. taeda* (softwood) with an average length close to 2 mm and an average width of 32 µm and coarseness of the order of 0.06 mg.m^{-1} and 0.15 mg.m^{-1}, respectively. The morphological differences between these fiber types cause distinct influences on the structure of the tissue paper sheet, during the forming process in the wet end of the machine, which were then evaluated by modeling the structural uniformity through the mass distribution in the plane of the random fiber network, which provides important insights into the structure of paper produced industrially. The analysis of the average coverages, as well as the probabilistic distribution of coverages and basis weights in points, allows the evaluation of the interference factors of fiber morphological and structural parameters with the uniformity of the sheet formation.

Fiber network coverage

Figure 4 (a) shows the mean values obtained for fiber mass and basis weight for different mixtures of cellulosic pulps containing *E. urograndis* and *P. taeda* fibers, correlated with the *P. taeda* mass fraction. Fiber mass values ranged from 0.5 x 10^{-7} g to 2.9 x 10^{-7} g, while basis weights were found to range from 3.4 g.m^{-2} to 4.6 g.m^{-2}, respectively.

Fiber basis weight directly determines the average value of coverages, as indicated in Figure 4 (b), which more broadly depend on intrinsic parameters of the fibers, *i.e.*, on the width and coarseness, which define the basis weight of each type of fiber or the resulting average basis weight for each fiber mix ratio. The average coverage variation also follows a proportionality with respect to the basis weight of the sheet. For the cases studied, a minimum coverage value of 2.81 was found for tissue paper of 13 g.m^{-2}/100% *P. taeda* and a maximum of 9.66 for tissue of 33 g.m^{-2}/100% *E. urograndis*.

Fiber mass defines the fiber population (number of fibers/grams of paper), which represents an important parameter for obtaining uniformity in paper formation. An increase in population resulting from the use of fibers with low basis weight – as with short fibers – determines an increase in the total length of fibers and consequent increase in the uniformity of the fiber network.[70] A comparative analysis between types of fibers and mixtures is presented in Table 3.

The data presented in Figure 4 (b) show typically lower coverage values for tissue papers when compared to other types of paper, such as printing and writing papers and newsprint. For the latter ones, the literature mentions usual coverages of the order of 5-20 and 10-20, respectively, considering fibers with basis weights in the range of 5-10 g.m^{-2}.[55,57] In this study, the total range of fiber weight variation (3.4 - 4.6 g.m^{-2}) is below the lower limit of typical weights indicated in Niskanen *et al.*[55] and Raunio and Ritala,[57] as a consequence of the characteristics of the raw materials considered. The low coverage levels (2.81-9.66), which represent the "equivalent number" of fiber layers in the paper sheet, corroborate the validity of the two-dimensional planar model of the tissue sheet, taken as a premise for the purposes of this work.

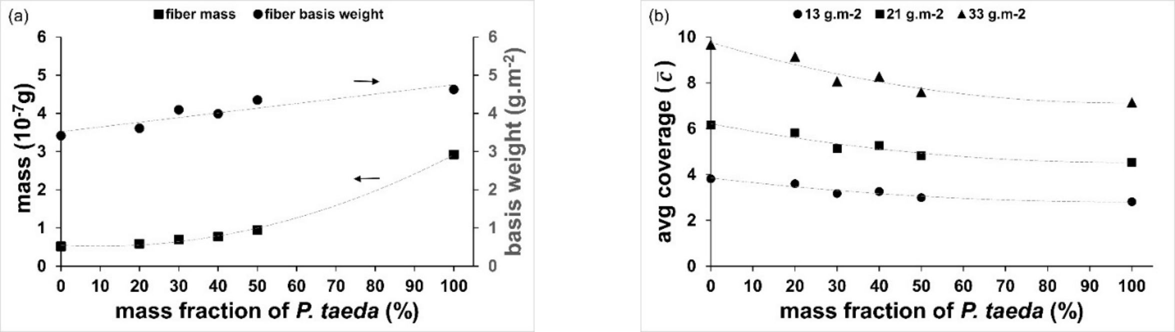


Figure 4: Influence of (a) the fraction of *P. taeda*, in the mixture with *E. urograndis*, on average basis weight and mass of individual fibers, and (b) on average coverage for different tissue grammages

Distribution of local basis weight

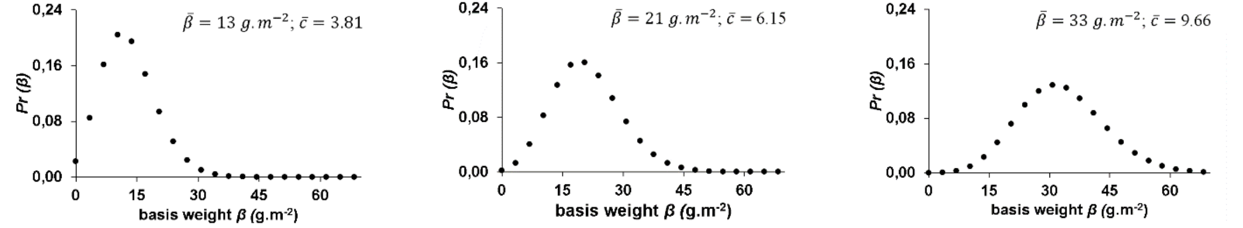
The Poisson distributions of basis weights in points, as given by Equation (4) combined with Equation (6), are displayed graphically in Figure 5 (a-c), for tissue paper grammages of 13 g.m^{-2}, 21 g.m^{-2} and 33 g.m^{-2}, having fiber basis weights of 3.42 g.m^{-2}, 4.35 g.m^{-2} and 4.63 g.m^{-2}, which correspond to the conditions of pulp compositions containing 100% *E. urograndis* (reference pulp A), 50% *E. urograndis* + 50% *P. taeda* (reference pulp E) and 100% *P. taeda* (reference pulp F), respectively.

The graphs disposed horizontally from left to right indicate that increases in the nominal grammage (or average basis weight) of the paper sheet determine reductions in the asymmetry (*skewness*) of the distribution. In the vertical direction, from top to bottom, increases in the mass fraction of long fibers (*P. taeda*) lead to increases in the peak and skewness of the probability distribution.

The probability distributions shown in Figure 5 (a-c) indicate that, for each grammage of the paper, the addition of long fibers (*P. taeda*) in the mixture with short fibers (*E. urograndis*) increases the probability of the lowest and highest basis weight values in the fiber network, as expected, and also produces an increase in the height (peak), asymmetry and variance of the distribution. In turn, lower degrees of asymmetry and variance are observed with the increase in the proportion of short fibers (*E. urograndis*) in the mixture. As for the increase in paper grammage, the result shows a probability distribution with less asymmetry, but with greater basis weight variability.

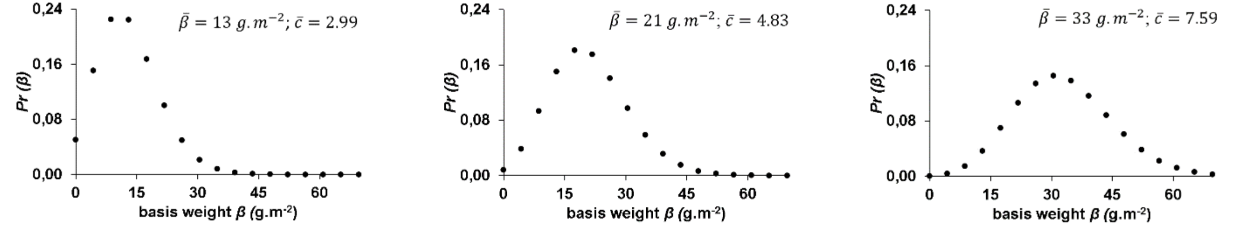
Table 2 gathers additional statistical information of the basis weight distributions shown in Figure 5 (a-c), in aid of interpreting the characteristics of the graphs. The values of skewness coefficients found for different paper compositions and grammages characterize positive asymmetries and, consequently, distributions with data variability towards the lower values of local basis weight, whose effect is more pronounced for the paper with the lowest grammage, *i.e.*, for 13 g.m^{-2}. The data also confirm increases in skewness and dispersion, produced by the increase in the mass fraction of long fiber in the composition of the pulp.

(a)



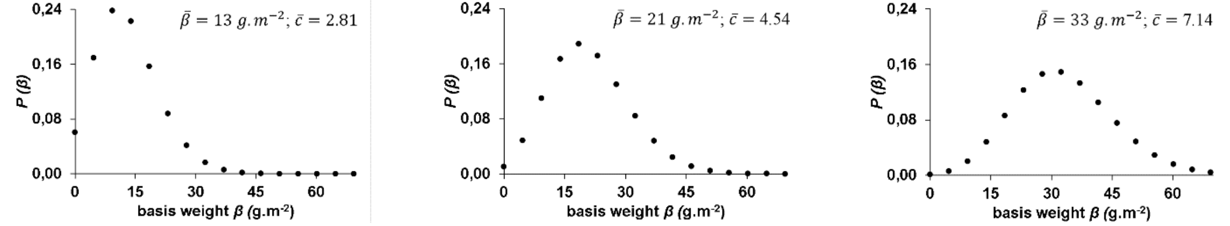
reference pulp A (β_f = 3.42 *g.m^{-2}*)

(b)



reference pulp E (β_f = 4.35 *g.m^{-2}*)

(c)



reference pulp F (β_f = 4.63 *g.m^{-2}*)

Figure 5: Probability distributions of local basis weight for different paper grammages and furnish compositions

Table 2
Skewness and excess kurtosis of the basis weight Poisson distributions

Sheet basis weight ($\bar{\beta}$) (g.m^{-2})	Coefficient of skewness [a] $s_k = (\beta_f/\bar{\beta})^{1/2}$			Coefficient of ex. kurtosis [b] $k_t = \beta_f/\bar{\beta}$		
	ref. pulp A	ref. pulp E	ref. pulp F	ref. pulp A	ref. pulp E	ref. pulp F
13	0.51	0.58	0.60	0.26	0.16	0.10
21	0.40	0.46	0.47	0.33	0.21	0.13
33	0.32	0.36	0.37	0.36	0.22	0.14

[a, b] corresponds to $s_k = 1/\sqrt{\bar{c}}$ and $k_t = 1/\bar{c}$, respectively, according to Fazal and Bashir[73]

The excess kurtosis values found confirm the increase in the peak of the distributions observed in the graphs, for additions of long fibers in the mixture. Similarly, these values also confirm a greater flattening of the distributions, for conditions of higher paper grammages, which produce greater data dispersion and, therefore, determine higher variance magnitudes. As for the Gaussianity of basis weight distributions in points, by the decrease of the skewness and excess of kurtosis coefficients, a tendency of approximation of the Poisson distribution to the Gaussian distribution is observed, in conditions of *coverage* increase, that is, of increase of paper grammage, as discussed by Niskanen *et al.*,[55] remembering that the Gaussian distribution has null skewness and excess of kurtosis coefficients.

The morphological characteristics of the fibers, associated with the basis weight of the sheet, also show impacts on the probabilities of occurrence of holes in the paper sheet, from Equation (4), considering zero coverages [$Pr\ (0) \mid c = 0$] and various proportions of mixtures, as shown in Figure 6 (a). Papers with lower basis weight show greater probability of the occurrence of holes, which increase with the increase in the fraction of long fibers (*P. taeda*). The first fact is justified by the lower number of equivalent layers of fibers in sheets of lower basis weight, which tends to limit the uniformity of coverage, increasing the possibility of the appearance of uncovered spots in the projection on the plane of the sheet of paper. The second fact is explained by the smaller amount, or population of fibers (no. of fibers. g^{-1}), which similarly also induces coverage failures, given that in a random network, fibers have an equal probability of orientation, establishing any angle in relation to any arbitrary fixed axis.[26]

The effects of the morphological characteristics of the fibers on the variances of the probability distributions are reflected in the coefficients of variation of the basis weight probability distributions [$cv\ (\beta) = (\beta_f/\bar{\beta})^{-1/2}$], for each mixture of fibers, as shown in Figure 6 (b), where the coefficient of variation increases with the increase in the fraction of *P. taeda* in the pulp mixture and with the decrease in basis weight of the tissue sheet.

Increased short fiber content in tissue paper composition

The trend towards the predominant use of hardwood short fibers (*e.g.*, *E. urograndis*) in tissue paper composition has been remarkable due to the objectives of leveraging the key quality properties of tissue products. This trend can be theoretically supported by the approach of geometric statistics, which is the focus of this work. The lowest levels of the coefficient of variation of the basis weight probability distribution in points found by decreasing the *P. taeda* fraction or even by adoption of 100% of *E. urograndis* in the pulp composition, as shown in Figure 6 (b), support the propensity for better sheet formation when short fibers are considered. In addition to the effects found by analyzing the probability distribution of coverage and basis weight, Table 3 shows other impacts of cellulose fibers morphological properties on the structural characteristics of the paper.

The uniformity of the fibrous network is understood as being proportional to the total length of the fibers present per unit area of paper sheet.[70] It is influenced by the properties of mentioned fibers, such as length, width, and coarseness, and therefore by the choice of high proportions of *E. urograndis*, (short fibers) in the composition of the cellulosic pulp used in the production of tissue paper. This leads to greater total length of fiber per unit area, as shown in Table 3, for 13 g.m^{-2} paper, as an example, since trends are similarly valid for grammages other than those addressed in this work.

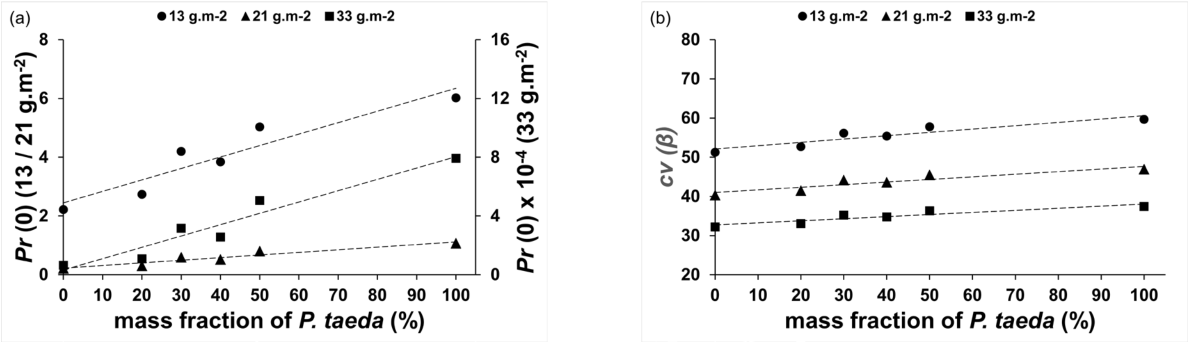


Figure 6: Influence of *P. taeda* fraction in the mixture with *E. urograndis* on (a) the probability of holes occurring in the tissue sheet and (b) on the coefficient of variation of the basis weight distribution in points

Table 3
Structural properties for tissue paper of 13 g.m^{-2}, containing the pulp blends indicated in Table 1

Parameters		Reference pulp					
		A	B	C	D	E	F
Mass fraction of *P. taeda*	%	0	20	30	40	50	100
Mass fraction of *E. urograndis*	%	100	80	70	60	50	0
Paper grammage ($\bar{\beta}$)	g.m^{-2}	. . . . 13					
Average coverage ($\bar{c}$)	-	3.81	3.60	3.17	3.26	2.99	2.81
Fiber centers / unit area	mm^{-2}	252	221	185	168	138	44
Total fiber length / unit area	cm. mm^{-2}	21.0	20.3	18.1	17.6	15.7	8.8

CONCLUSION

The present article addresses, through a specific theoretical study, the influence of intrinsic morphological properties of cellulosic fibers on the structural aspects of typical tissue papers and, thus, provides insights into the selection of fibers and pulp blends for manufacturing these papers. For modeling purposes, the classic reference structure for a paper web was adopted. It consists of a stochastic fibrous network, in which the fibers are randomly arranged on the support plane during the paper sheet forming process, according to the Poisson distribution function.

The coefficient of variation of the grammage probability distribution in points, taken as predictor of the tissue sheet structural uniformity, was found to decrease (better formation) with the increase of the proportion of short fibers (hardwood) in the furnish composition. Conversely, an increase in the coefficient of variation (worse formation) was observed with the increase of the proportion of long fibers (softwood). The expression derived from the Poisson distribution for the probability of holes in the paper agreed with the results above, indicating greater expectations of pin holes to exist with the increase of the proportion of long fibers (softwood). Additionally, the present study reconciled the effects of fiber properties with the sheet grammage, indicating better possibilities of formation uniformity for higher grammages, due to the greater number of equivalent layers of fibers in the paper sheet. The increased coverage, found in papers with higher grammages, also led to a greater total fiber length, when the proportion of short fibers (hardwood) is increased, then contributing to an enhanced formation.

The current tendency to use short fibers (hardwood) in high proportions for manufacturing tissue paper is thus legitimate. This consideration supports not only the attempts to reduce raw material cost in specific geographic regions, but also contributes to greater levels of structural uniformity of the paper sheet, which represents an essential requirement for achieving proper key quality properties of tissue end products, such as softness, absorption, and strength.

The theoretical approach presented in this study, based on statistical geometry, brings meaningful insights for practical considerations related to the selection and optimization of papermaking furnishes intended to industrial production, when the focus on the uniformity of tissue paper is of prime importance.

ACKNOWLEDGEMENTS: The support by the Chemical Engineering Department Research Associate Programme of the Polytechnic School of the University of Sao Paulo is thankfully acknowledged by A.H.T. Mendes.

REFERENCES

[1] Food and Agriculture Organization of the United Nations (FAO), "Forestry Production and Trade" (2022), https://www.fao.org/faostat/en/#data/FO/

[2] Research and Markets, "Tissue Paper Market: Global Industry Trends, Share, Size, Growth, Opportunity and Forecast 2021-2026" (2021), https://www.researchandmarkets.com/reports/5353327/tissue-paper-market-global-industry-trends/

[3] Food and Agriculture Organization of the United Nations (FAO), "Pulp and Paper Capacities Survey 2020-2025" (2022), https://www.fao.org/3/cb7300t/cb7300t.pdf/

[4] C. A. Farinha e Silva, M. R. Neves and M. Porto, in "ABTCP Guide of Pulp and Paper Manufacturers and Suppliers 2021/22", edited by ABTCP, 2002, pp. 18-29

[5] J. Kullander, Licentiate Thesis, Karlstad University, 2012

[6] W. E. Scott, J. C. Abbott and S. Trosset, "Properties of Paper: An Introduction", 2nd ed., Tappi Press, 1995, Chapter 11, pp. 179-185

[7] Packaging Specialties, Specialty Tissue Paper (2021), https://www.packagingspecialtiesonline.com/collections/specialty-tissue-paper/

[8] J.-P. Raunio, T. Loyttyniemi and R. Ritala, *Nord. Pulp Pap. Res. J.*, **33**, 133 (2018)

[9] X. Zou, in *Procs. PaperCon 2017 Conference*, Minneapolis, April 23-26, 2017, pp. 1-28

[10] H. Morris, *Tissue World Magazine*, April 21 (2012), https://www.tissueworldmagazine.com/technical-theme/optimising-the-tissue-fibre-furnish/

[11] M. Fiserová, J. Gigac, M. Stankovská and E. Opálená, *Cellulose Chem. Technol.*, **53**, 469 (2019), https://doi.org/10.35812/CelluloseChemTechnol.2019.53.47

[12] E. Retulainen, K. Niskanen and N. Nilsen, in "Paper Physics", edited by K. Niskanen, Fapet Oy, Papermaking Science and Technology Series, Book 16, Chapter 2, 1998, pp. 54-87

[13] W. R. Hurter, Bagasse Pulp Uses in Papermaking, Hurter Consult Incorporated, 2020, www.HurterConsult.com

[14] M. Byrd and R. Hurter, in *Procs. Tissue 360° Forum at PaperCon 2013 Conference*, April 27-May 1, Atlanta, 2013, pp. 1-60

[15] M. Guan, X. An and H. Liu, *Cellulose*, **26**, 2613 (2019), https://doi.org/10.1007/s10570-018-2212-6

[16] X. An, J. Liu, L. Liu, H. Zhang, S. Nie *et al.*, *Ind. Crop. Prod.*, **150**, 112410 (2020), https://doi.org/10.1016/j.indcrop.2020.112410

[17] M. Stankovská, M. Fiserová, J. Gigac and E. Opálená, *Wood Res.*, **65**, 447 (2020)

[18] R. Kumar, F. Zambrano, I. Peszlen, R. Venditti, J. Pawlak *et al.*, *Cellulose*, **29**, 6907 (2022), https://doi.org/10.1007/s10570-022-04687-3

[19] B. Norman and D. Wahren, in "Fundamental Properties of Paper Related to its Uses", edited by F. Bolam, FRC, 1973, Manchester, pp. 7-70

[20] M. M. Nashad, E. J. Harris, C. T. J. Dodson and R. Kerekes, *Tappi J.*, **83**, 1 (2000)

[21] M. Alzweighi, R. Mansour, J. Lahti, U. Hirni and A. Kulachenko, *Acta Mater.*, **203**, 116460 (2021), https://doi.org/10.1016/j.actamat.2020.11.003

[22] W. Karnchanapoo, A. Palokangas and M. M. Nashad, in *Procs. 55th Appita Annual Conference*, Hobart, April 30 - May 2, 2001, pp. 259-265

[23] H. Lee, H. Youn, S. G. Lee and Y. B. Jeong, *Tappi J.* (Korea), **39**, 1 (2007)

[24] A. J. Kiviranta, in *Procs. 1996 Papermakers Conference*, Philadelphia, March 24-27, 1996, pp. 239-245

[25] W. W. Sampson, *J. Pulp Pap. Sci.*, **34**, 91 (2008)

[26] R. J. Norman, in "Consolidation of the Paper Web", edited by F. Bolam, FRC, Manchester, pp. 269-298

[27] B. Nordström and L. Hermansson, *Nord. Pulp Pap. Res. J.*, **32**, 119 (2017)

[28] B. Norman, in "Stock Preparation and Wet End", edited by H. Paulapuro, Fapet Oy, 2000, Book 8, Papermaking Science and Technology Series, pp. 191-250

[29] S. Adanur, "Paper Machine Clothing", Technomic Publishing Co. Inc., 1997, pp. 33-152

[30] B. W. Janda, in *Procs. PaperCon 2014 Conference*, Nashville, April 27-30, 2014, pp. 3231-3269

[31] M. Htun and C. Fellers, *Tappi J.*, **65**, 113 (1982)

[32] M. A. Hubbe, *BioResourses*, **1**, 281 (2006)

[33] M. K. Ramasubramanian, in "Handbook of Physical Testing of Paper", edited by R. E. Mark, C. C. Habeger Jr., J. Borchi and M. B. Lyne, 2nd ed., Vol. 1, Marcel Derker Inc., 2002, pp. 661-698

[34] https://www.amazon-papyrus.com/getimage/index/action/news/name/4.pdf

[35] I. Olson and L. Pihl, *Svenk Paperstidn.*, **55**, 233 (1952)

[36] C. T. J. Dodson and W. W. Sampson, *J. Pulp Pap. Sci.*, **22**, J165 (1996)

[37] H. K. Corte and E. H. Lloyd, in "Consolidation of the Paper Web", edited by F. Bolam, B. P. & B. M. A., 1965, pp. 981-1009

[38] R. J. Kerekes, *Nord. Pulp Pap. Res. J.*, **21**, 100 (2006)

[39] A. H. T. Mendes and S. W. Park, *Nord. Pulp Pap. Res. J.*, **38**, 253 (2023), https://doi.org/10.1515/npprj-2023-0011

[40] J.-P. Raunio, R. Ritala and M. Mäkinen, in *Procs. Control System 2012 Conference*, New Orleans, April 22-25, pp. 23-41

[41] D. Pettman and I. Padley, *Pap. Technol. Int.*, **56**, 12 (2015)

[42] G. S. Furman, "Tissue Story", 2017, Chapter 4-2, https://www.tissuestory.com/wiki-tissue/

[43] H. Hollmark, "Study of the Creping Process on an Experimental Paper Machine", Stockholm, Sweden, Swedish Forest Products Laboratory, STFI-Meddelande, Series B, Nr. 144

[44] R. S. Seth, *Tappi J.*, **78**, 99 (1995)

[45] L. Paavialainen, in "Handbook of Physical Testing of Paper", edited by R. E. Mark, C. C. Habeger Jr, J. Borchi and M. B. Lyne, 2nd ed., 2002, Vol. 1, Marcel Derker Inc., pp. 699-725

[46] B. Nordström and L. Hermansson, *Nord. Pulp Pap. Res. J.*, **33**, 237 (2018)

[47] Y. J. Park, L. Melani, H. Lee and H. J. Kim, *Holzforschumg*, **74**, 497 (2019)

[48] S. J. Eichhorn and W. W. Sampson, *J. R. Soc. Interface*, **2**, 309 (2005)

[49] H. Corte and O. J. Kallmes, in "Formation and Structure of Paper", edited by F. Bolam, B. P. & B. M. A., 1962, pp. 13-46

[50] H. Corte and O. J. Kallmes, *Tappi J.*, **43**, 737 (1960)

[51] M. Deng and C. T. J. Dodson, "Paper: An Engineered Stochastic Structure", Tappi Press, 1994

[52] W. W. Sampson, in "The Science of Papermaking", edited by C. F. Baker, FRC, 2001, pp. 1205-1288

[53] K. Niskanen, N. Nilsen, E. Hellen and M. Alava, in "The Fundamentals of Papermaking Materials", edited by C. F. Baker, FRC, pp. 1273-1291

[54] R. Das, K. Pan, S. Green and A. S. Pani, *Adv. Eng. Mater.*, **23**, 2000777 https://doi.org/10.1002/adem.202000777

[55] J.-P. Raunio and R. Ritala, *Meas. Sci. Technol.*, **24**, 1 (2013), https://doi:10.1088/0957-0233/24/12/125206

[56] J.-P. Raunio and R. Ritala, *Nord. Pulp Pap. Res. J.*, **27**, 375 (2012)

[57] K. Niskanen, I. Kajanto and P. Pekarinen in "Paper Physics", edited by K. Niskanen, Fapet Oy, 1998, Book 16, Papermaking Science and Technology Series, pp. 13-53

[58] C. Bronkhorst, *Int. J. Solids Struct.*, **40**, 5441 (2003)

[59] J.-P. Raunio, Doctoral Thesis, Tampere University of Technology, 2014

[60] C. H. Chang, S. T. Yu and Y. S. Perng, *Cellulose Chem. Technol.*, **52**, 433 (2018), https://www.cellulosechemtechnol.ro/pdf/CCT5-6(2018)/p.433-440.pdf

[61] F. P. Morais, R. A. C. Bértolo, J. M. R. Curto, M. E. C. C. Amaral, A. M. M. S. Carta *et al.*, *Mater. Lett. X*, **4**, 1 (2019), https://doi.org/10.1016/j.mlblux.2019.100028

[62] M. Stankovská, M. Fiserová, J. Gigac and E. Opálená, *Wood Res.*, **63**, 505 (2021), https://doi:/10.37763/wr.1336-4561/66.3.505516

[63] M. Debnath, K. S. Salem, V. Naithani, E. Musten, M. A. Hubbe *et al.*, *Cellulose*, **28**, 7981 (2021), https://doi.org/10.1007/s10570-021-04024-0

[64] G. B. Olson, *Science*, **277**, 1237 (1997)

[65] W. W. Sampson, *Int. Mater. Rev.*, **54**, 134 (2009)

[66] A. G. M. C. Bassa, Master's Thesis, University of Sao Paulo, 2006

[67] F. G. Silva Jr., A. G. M. Bassa, V. M. Sacon and E. Patelli, in *Procs. Tappi 2007 Engineering, Pulping and Environmental Conference*, Jacksonville, October 21-24, 2007, pp. 1-11

[68] A. C. F. Vidal and A. B. Hora, "Panorama de mercado: papéis sanitários", BNDES Setorial 37, 2013, pp. 273-332, http://web.bndes.gov.br/bib/jspui/handle/1408/1495

[69] Voith Paper Technology Center, Tissue Paper and Fibers, 2019, pp. 1-22

[70] W. W. Sampson, "Modelling Stochastic Fibrous Materials with Mathematica®", Engineering Materials and Processes Series, edited by B. Derby, Springer, 2009, pp. 1-277

[71] O. J. Kallmes, H. Corte and G. Bernier, *Tappi J.*, **144**, 519 (1961)

[72] M. H. DeGroot and M. J. Schervish, in "Probability and Statistics", 3rd ed., Addison Wesley, 2002, Chapter 4, pp. 181-245

[73] A. Fazal and S. Bashir, *Int. J. Appl. Math. Stat. Sci.*, **6**, 1 (2017)