NANOFIBRILLATED CELLULOSE APPLIED AS REINFORCEMENT FOR SHORT-FIBER PAPER

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Resumo

Nanocelulose aplicada como reforço para papel de fibra curta. Este trabalho teve como objetivo avaliar a influência da adição de diferentes porcentagens de celulose nanofibrilada sobre as propriedades mecânicas e físicas do papel feito a partir da polpa de fibras curtas. A celulose nanofibrilada foi obtida a partir de polpa Kraft de *Eucalyptus* sp. branqueada, submetida a três diferentes passes no moinho: 2, 10 e 20 passes. Os papéis foram produzidos com a adição de celulose nanofibrilada nas porcentagens de 3, 6 e 9%. Os resultados mostraram que a adição de celulose nanofibrilada aumentou as propriedades mecânicas: índice de tração, índice de arrebentamento e índice de rasgo. A porosidade e a densidade aparente diminuíram. A adição de 9% de celulose nanofibrilada, obtida a partir de 2 passes, proporcionou os melhores resultados com aumento da resistência à tração, arrebentamento e rasgo de 111, 114 e 70%, respectivamente, em comparação aos papéis normais. A melhoria das propriedades mecânicas do papel está relacionada à rede muito densa de ligações de hidrogênio, resultando em maior área de superfície obtida após a desfibrilação. A nanocelulose apresenta potencial para ser aplicada na melhoria da qualidade do papel e como agente de reforço em compósitos.
Palavras chave: nanofibras, CNF, polpa celulósica, propriedades mecânicas, *Eucalyptus*

Abstract

In this work we studied the influence on the mechanical and physical properties of paper made from short-fiber pulp by adding different percentages of nanofibrillated cellulose. Bleached *Eucalyptus* sp. Kraft pulp was submitted to three different grinding regimes to obtain the nanofibrillated cellulose of 2, 10 and 20 passes through the grinder. Paper was produced by incorporating nanofibrillated cellulose in the proportions of 3, 6 and 9 wt%. The results showed that the addition of nanofibrillated cellulose increased the following mechanical properties: tensile index, tear index, and burst index. The porosity and the apparent density decreased. The addition of 9% of nanofibrillated cellulose obtained from 2 grinding passes provided the best results with improvements in tensile, burst and tear resistance of 111, 114 and 70 %, respectively, in comparison to normal paper. The improvement in the mechanical properties of paper is related to the very dense network of hydrogen bonds, resulting in greater surface area obtained after defibrillation. Nanofibrillated cellulose can be applied to improve paper quality and for reinforcing composites.
Keywords: Nanofibrils, NFC, pulp, mechanical properties, *Eucalyptus*.

INTRODUCTION

Wood is chemically composed of cellulose, hemicellulose, lignin, extractives and ash. The percentages of each component vary according to species, fiber type, tree position, growth condition, age and extraction methods. Cellulose is present in the greatest percentage among these materials, representing about 40-45% of the total wood mass (FENGEL; WEGENER, 1984).

Successful use of cellulose from different sources has been reported to produce new sustainable products or as a substitute for other polymers (CHEN *et al*., 2017). The products obtained from cellulose are biodegradable, low density, environmentally friendly and low cost, and have good physical and mechanical properties (MOHAMMADKAZEMI *et al*., 2015).

Pinus sp. and *Eucalyptus* sp. are the most used species for long- and short-fiber pulping in Brazil, respectively. These species grow rapidly in addition to having good technological properties for pulp and paper production (such as mechanical and physical properties). Brazil's planted forests have the highest productivity and shortest rotation in the world. These high rates are the result of the climate and soil conditions, as well as continuous investments by companies in the country (IBA, 2017). *Eucalyptus* species are mainly used to produce bleached pulp for writing, printing and tissue paper, accounting for about 87% of pulp production in Brazil (IBA, 2017).

In the context of sustainable development, the sector has an advantage in relation to global competition as all pulp and paper production in the country is from planted forests. Considering the global cellulose fiber market, Brazil is the one of the largest suppliers of cellulose fiber and finished products made from it such as packaging, printing and writing paper, and paperboard (IBA, 2017).

The pulp and paper sector has been investing in research with the purposes of creating new products, better use of by-products, reducing inputs in the production process and improving the quality of products. Studies with nanofibrillated cellulose (NFC) indicate the material as an excellent input in the pulp and paper industry (GONZÁLEZ *et al*., 2012; POTULSKI *et al*., 2018). NFC can be used to replace chemical additives, reduce refining time and improve paper properties.

NFC enables producing paper with higher density, better mechanical properties, low porosity, excellent oxygen barrier properties and less water absorption (JONOOBI *et al*., 2012; POTULSKI, *et al.* 2018). One of the reasons for the increase in the mechanical properties of NFC-produced paper is the improvement of interfibrillar bonding, which reduces porosity and increases the density of these paper products (KHALIL *et al*., 2014). González *et al*. (2012) studied nanofibrillated cellulose obtained by a combination of chemical treatment and mechanical processing and observed an increase in the mechanical properties of the paper.

Recent research shows nanofibrillated cellulose as a reinforcement in composite materials due to its high resistance and rigidity and low weight. The larger contact surface between the adjacent cellulose fibers provides a higher number of hydrogen bonds, forming a denser network. This fact promotes outstanding mechanical properties, biocompatibility, transparency and high reactivity (ZIMMERMAN *et al*., 2010). These have been studied for polymer applications in the packaging, biomedicine, adhesive, fiber, electronics and automotive industries (ZIMMERMAN *et al*. 2010).

Nanofibrillated cellulose is recognized as having potential in the field of pulp and paper technology. There is also research interest on its use in papermaking, coating and films. Many studies have shown that NFC can be applied as strength additives for paper to enhance barrier properties in food packaging, to improve paper gloss, to reduce paper grammage and for smart and sustainable packaging (YOO; HSIEH, 2010). NFC has an advantage in use due to its relative abundance, renewability, biodegradability, high specific surface area, high aspect ratio, high strength and stiffness, and low weight (OSONG *et al.* 2016).

In this context, this work aimed to evaluate the incorporation of different percentages of nanofibrillated cellulose (NFC) in short-fiber paper to reinforce paper sheets.

MATERIAL AND METHODS

Material

Eucalyptus sp. (Kappa no. = 50.9) material was used to obtain the nanofibrillated cellulose and bleached *Eucalyptus* sp. Kraft (Kappa no. = 3.5) commercial pulp was used to make paper.

Obtaining Nanofibrillated Cellulose (NFC)

Nanofibrillated cellulose (NFC) was obtained from *Eucalyptus* sp. cellulose pulp, which was first dispersed in water and disintegrated for five minutes to obtain a homogeneous suspension of fibers with water concentration of 1% by dry mass. This suspension was subjected to mechanical defibrillation in a Masuko Sangyo Super Masscolloider (MKCA6-3), with 2, 10 and 20 passes through a grinder at 1500 rpm rotation.

Microscopic Characterization

The NFC was characterized by transmission electron microscopy analysis to visualize the dimensions and appearance of the fibrillar structures. Each sample was solubilized in water and deposited on a grid with 200 mesh size and then dried to perform the characterization. The samples were kept at room temperature for solvent evaporation to form a film. The images were acquired in a JEOL 1200EXII electron microscope using a magnification of 15,000 X.

Paper manufacture with nanofibrillated cellulose addition

Paper sheets were produced by incorporating nanofibrillated cellulose in the proportions of 3, 6 and 9% by performing a mechanical mixture in a mixer, in addition to the treatment without the addition of nanofibrillated cellulose for subsequent comparison (control), totaling 10 treatments.

The water drainage resistance of the cellulosic pulp was determined with a S*chopper Riegler* tester before producing the sheets, according to ISO 5267/1.

The paper samples were produced with a Rapid-Köethen type forming station with a drying temperature of 90 ± 2 °C and pressure of 40 kPa, according to the ISO 5269/2 and T205 sp-02 standards. Five samples were prepared per treatment, with average grammage of 60 ± 3 g/m².

As specified in the T402-om94 standard, the paper samples were dried and then air-conditioned at a temperature of 23 ± 2 °C and relative humidity of 50 ± 2% before the physical and mechanical tests.

Physical and mechanical tests

The acclimatized paper samples were tested to determine the following physical properties: moisture content (T412-om02), grammage (T410-om02), thickness (T411-om97), water absorption (T441-om98), and apparent density (T220-sp01), as well as the mechanical properties of: tensile strength (T404-om92), bursting strength (T403-om02) and internal tearing strength (T414-om98).

The tensile strength was measured through the tensile index, which corresponds to the ratio between the resistance and weight of the sample, expressed in N.m/g. The bursting index is calculated by the ratio between burst strength and sample weight, and is expressed in Kpa.m^2/g. The internal tearing index is calculated through the ratio between tear strength and sample weight, and is expressed in mNm^2/g. Five samples of each treatment were submitted to each physical and mechanical test.

Statistical analysis

The factorial analysis of variance of the physical and mechanical properties was performed considering 95% reliability. This analysis was applied to check if there was a significant influence of the factors corresponding to percentage of nanofibrillated cellulose addition and the number of grinding passes of the nanofibrillated cellulose, and also if there was interaction between these factors. The homogeneity of variances was tested by the Bartlett test, in which all variances of the samples were homogeneous.

RESULTS

Transmission electron microscopy (TEM)

Figure 1 (A, B and C) presents TEM images of the fiber after being submitted to the mechanical defibrillation process with 2, 10 and 20 passes through the grinder, respectively.

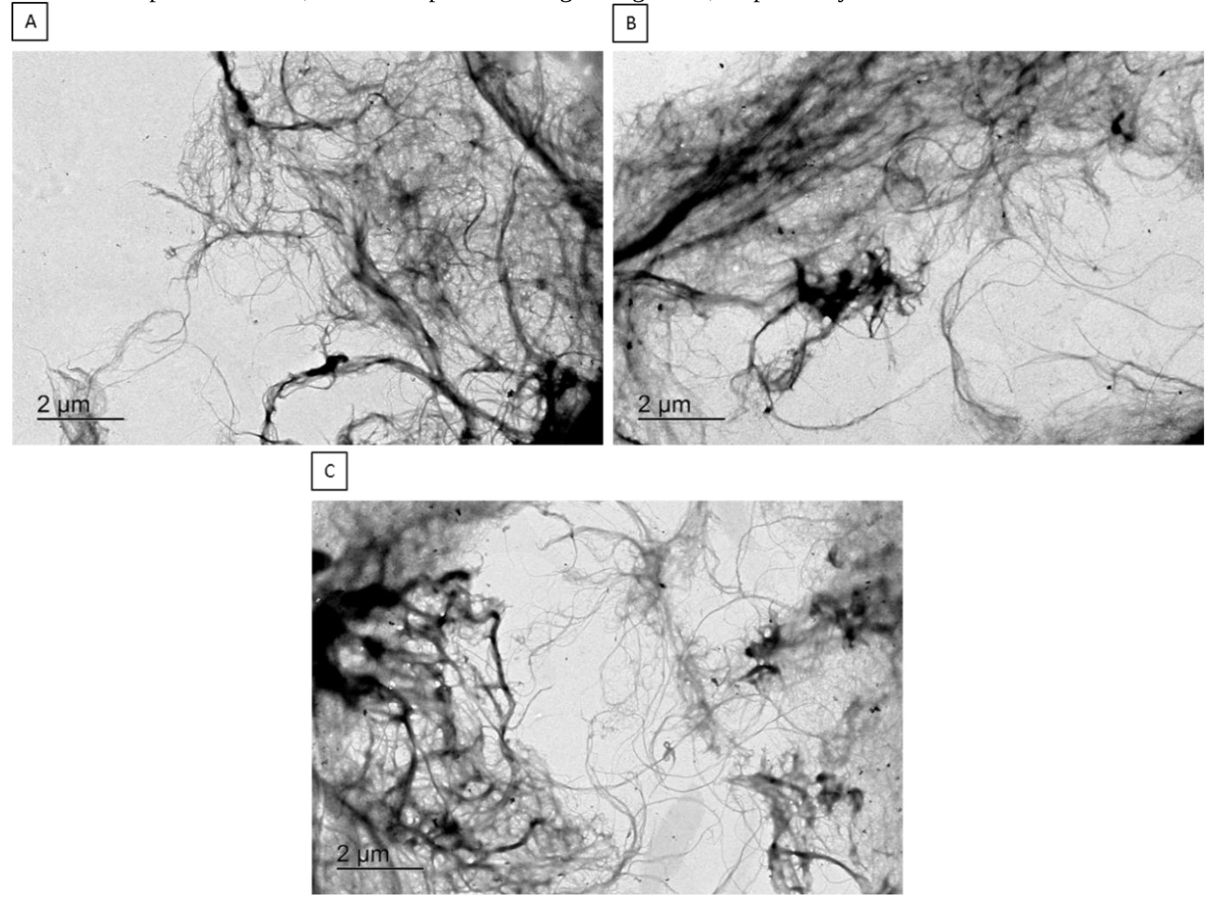

Figure 1. Images of cellulose nanofibrils from *Eucalyptus* sp. obtained by TEM: a) 2 passes; b) 10 passes; c) 20 passes.
Figura 1. Imagens da celulose nanofibrilada de *Eucalyptus* sp. obtidas por meio do MET: a) 2 passes; b) 10 passes; c) 20 passes.

According to the images acquired through TEM, there were no significant differences between 2, 10 and 20 grinding passes (Figure 1 a, b and c). The TEM investigations show that the nanofibrils present average diameter after the defibrillation varying between approximately 20 and 45nm with 2, 10 and 20 passes.

Physical and mechanical properties of the paper

Figure 2 shows the effect of the relationship between the increase in the number of passes and the percentage addition of nanofibrillated cellulose on the physical properties of the paper samples.

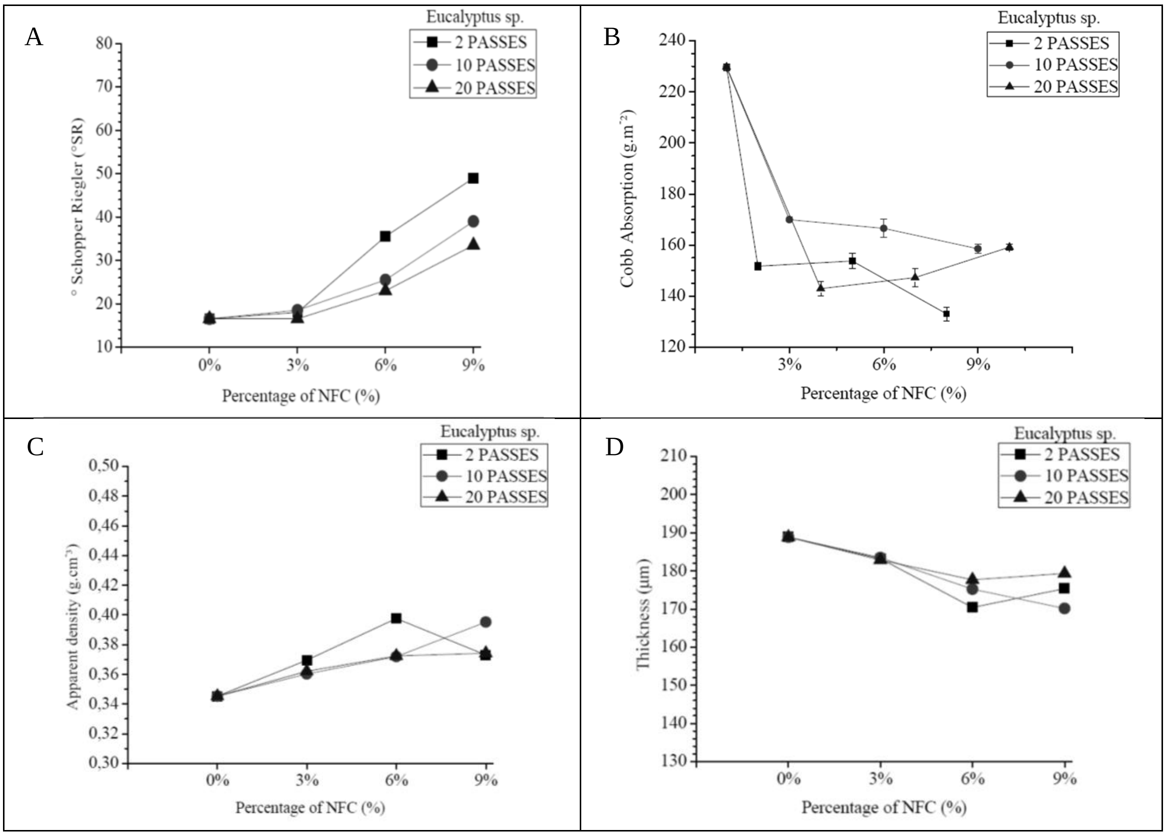

Figure 2. Physical properties of paper produced with NFC after different passes in relation to NFC percentage. In which: A. *Schopper Riegler*; B. Cobb absorption; C. apparent density; D. thickness.
Figure 2. Propriedades físicas dos papéis produzidos com CNF após diferentes passes em relação a porcentagem de CNF. Em que A. *Schopper Riegler*; B. absorção Cobb; C. densidade aparente; D. espessura.

Table 1 shows that the *Schopper Riegler* degree (°SR), water absorption (Cobb 60) and apparent density of the papers were influenced by the different numbers of passes, NFC percentages and the combination of these factors. The paper thickness was only influenced by the NFC percentage.

Table 1. Summary of factorial analysis of variance.
Tabela 1. Resumo da análise de variância fatorial.

Anova	**SR**	**Cobb$_{60}$**	**AD**	**Thickness**
	P	***P***	***P***	***P***
(NFC) wt%	0.0001*	0.0004*	0.0001*	0.0001*
Number of passes	0.0001*	0.0001*	0.0130*	0.0623^{ns}
NFC wt% x passes	0.0001*	0.0001*	0.0001*	0.0646^{ns}

SR = *Schopper Riegler* degree; AD = Apparent Density; * corresponds to significant differences ($p<0.05$), and ns corresponds to non-significant ($p>0.05$).

An increase of °SR in the paper was observed with the addition of nanofibrillated cellulose with 2, 10 and 20 passes. The water absorption by the paper decreased with nanofibrillated cellulose addition with 2, 10 and 20 passes (Figure 2). The density values increased with increasing NFC percentage, except for paper with the addition of 9% NFC with 2 passes. The values ranged from 0.35 to 0.40 ($g.cm^{-3}$) for treatments E00 and E109, respectively. The paper thickness formed by cellulose nanofibers presented a reduction when compared to the paper without NFC. This behavior was not observed for paper with the incorporation of 9% nanofibrillated cellulose with 2 and 20 passes.

Table 2 presents the physical properties of the paper in relation to the number of passes through the grinder and the nanofibrillated cellulose percentage addition.

Table 2. Average physical property values of paper.
Tabela 2. Valores médios das propriedades físicas dos papéis produzidos.

Treatment *	**SR (°)**		**Cobb$_{60}$ ($g.m^{-2}$)**		**AD ($g.cm^{-3}$)**		**Thickness (μm)**	
	$\overline{X}$	**Σ**	$\overline{X}$	**Σ**	$\overline{X}$	**Σ**	$\overline{X}$	**Σ**
E000	16.5 A	0.1	229.7 G	1.3	0.35 A	0.01	188.9 C	4.2
E023	18.0 B	0.3	151.7 CD	1.5	0.37 B	0.02	183.2 BC	9.5
E026	35.5 F	0.5	153.7 CDE	3.0	0.40 C	0.01	170.4 A	9.6
E029	49.0 H	1.0	133.0 A	2.7	0.37 B	0.02	175.3 AB	9.0
E103	18.5 B	0.2	169.9 F	1.0	0.36 AB	0.01	183.5 BC	6.7
E106	25.5 D	0.5	166.5 F	3.6	0.37 B	0.02	175.2 AB	8.1
E109	39.0 G	1.0	158.5 DE	1.7	0.40 C	0.02	170.1 A	8.2
E203	16.5 A	0.3	142.9 B	3.0	0.36 AB	0.01	182.9 BC	5.5
E206	23.0 C	1.0	147.3 BC	3.5	0.37 B	0.01	177.7 AB	6.1
E209	33.5 E	0.5	159.2 E	1.3	0.37 B	0.01	179.3 B	7.0

SR = *Schopper Riegler* degree; AD = Apparent density; *The first and second numbers after the letter represent the number of passes through the grinder and the third number the different NFC percentages. Averages followed by the same lower-case letter in the column are not different by the *Tukey* test, with a significance of 5%.

The ANOVA summary shows that the tensile, burst and tear indices (Table 3) were significantly influenced by the number of passes (2, 10 and 20 passes) ($p = 0.0001$), by the nanofibrillated cellulose percentage incorporation (3, 6 and 9%) ($p = 0.0001$) and the combination of these factors.

Table 3. Summary of factorial analysis of variance.
Tabela 3. Resumo da análise de variância fatorial.

Anova	**Tensile index**	**Burst index**	**Tear index**
	p	***P***	***p***
%wt Nanofibrillated celulose (NFC)	0.0001*	0.0001*	0.0001*
Number of passes	0.0001*	0.0001*	0.0001*
% wt NFC X no. of passes	0.0125*	0.0026*	0.0020*

* corresponds to significant differences ($p<0.05$) and ns corresponds to non-significant ($p>0.05$).

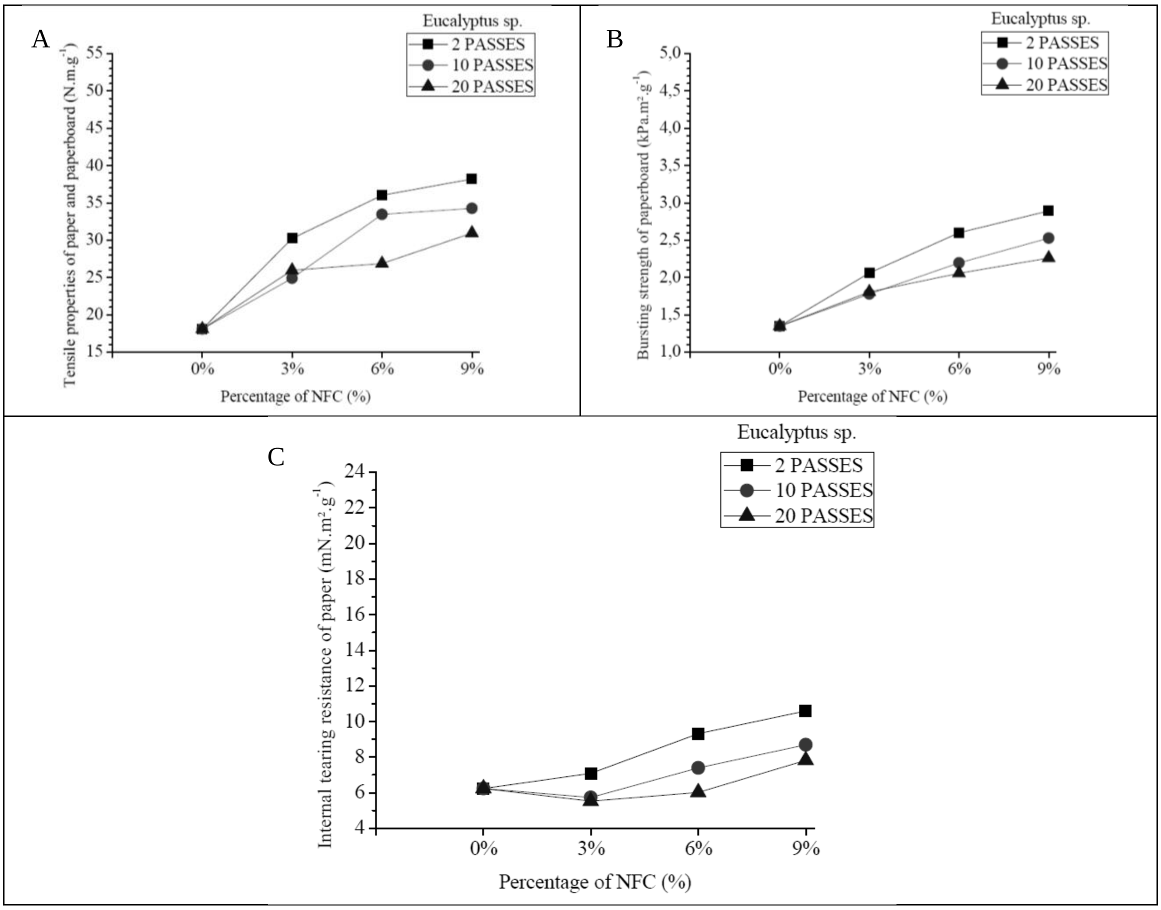

Figure 3. Mechanical properties of paper produced with NFC and different passes in relation to the NFC percentage. In which: A. Tensileindex; B. Burst index; C. Tear index.

Figura 3. Propriedades mecânicas dos papéis produzidos com CNF e diferentes passes em relação a porcentagem de CNF. Onde A. índice de tração; B. Índice de arrebentamento; C. Índice de rasgo.

The highest values of tensile, burst and tear indices were observed for the paper produced with 2 passes. A gradual increase occurred with the increase in the nanofibrillated cellulose percentage (Figure 3). The highest values were obtained for 9% NFC (Table 4).

Table 4. Average mechanical properties of paper.

Tabela 4. Valores médios para as propriedades mecânicas dos papéis.

Treatment	**Tensile Index (N.m.g^{-1})**		**Burst Index (Kpa.m^2.g^{-1})**		**Tear Index (mn.m^2.g^{-1})**	
	$\overline{X}$	**Σ**	$\overline{X}$	**Σ**	$\overline{X}$	**σ**
E000	18.1 A	1.9	1.3 A	0.1	6.2 AB	0.3
E023	30.2 CDE	2.2	2.1 C	0.1	7.1 BC	0.4
E026	36.0 FG	4.7	2.6 E	0.2	9.3 E	0.4
E029	38.2 G	4.6	2.9 F	0.1	10.6 F	0.5
E103	24.9 B	1.9	1.8 B	0.1	5.7 A	0.5
E106	33.5 EF	3.2	2.2 CD	0.2	7.4 C	0.6
E109	34.3 EFG	3.6	2.5 E	0.1	8.7 DE	0.5
E203	26.0 BC	2.9	1.8 B	0.1	5.5 A	0.4
E206	26.9 BCD	3.2	2.1 C	0.1	6.0 A	0.1
E209	31.0 DE	2.7	2.3 D	0.2	7.8 CD	0.6

*The first and second numbers after the letter represent the number of passes through the grinder and the third number the different NFC percentages. Averages followed by the same lower-case letter in the column are not different by the *Tukey* test, with a significance of 5%

Table 5 shows the percentage change in the mechanical properties of the paper with the addition of nanofibrillated cellulose. The incorporation of 9% nanofibrillated cellulose obtained with 2 passes through the Masuko grinder was notable for increasing the tensile index (111.35%), burst index (114.53%) and tear index (70.13%) of the paper made with virgin *Eucalyptus* sp. fibers.

The smallest percentages obtained were for the incorporation of 3% of nanofibrillated cellulose with 10 and 20 passes through the grinder.

Table 5. Percentage variation of mechanical properties of paper with the addition of NFC in relation to treatment without addition.
Tabela 5. Variação em porcentagem das propriedades mecânicas dos papéis com adição de celulose nanofibrilada em relação ao sem adição.

Treatment	Tensile Index (%)	Burst Index (%)	Tear Index(%)
E023	67.28	52.85	13.79
E026	99.29	92.90	49.52
E029	111.35	114.53	70.13
E023	37.76	31.85	-7.90
E106	85.11	63.01	18.64
E109	89.63	87.58	39.51
E103	43.63	34.16	-11.30
E206	48.58	52.36	-3.42
E209	71.26	67.62	25.63

*The first and second numbers after the letter represent the number of passes through the grinder and the third number the different NFC percentages.

DISCUSSION

Transmission electron microscopy (TEM)

In considering nanofibrillated cellulose to be nanostructures with smaller diameter than 100 nm (VIANA *et al*., 2017), the mechanical defibrillation process with 2 passes through the mill was sufficient to obtain nanometric scale materials.

Other authors have also observed the same tendency, such as Wang *et al.* (2013), who obtained cellulose nanofibrils with diameter between 30 and 100 nm after 20 mill passes at 1,600 rpm and consistency of 1%. Viana *et al.* (2017) did not observe significant differences between nanofibrillated cellulose diameters obtained with 2, 5, 10, 20, 30 and 40 passes through the grinder at 1% consistency. Fonseca *et al*. (2016) obtained diameters of less than 40 nm. The authors observed that it was possible to increase the swelling capacity with the reduction of the fibril dimensions, since the fiber surface was larger in relation to volume.

The ratio between the fiber length and diameter in NFC is high, enabling better capacity to form a stiff and homogeneous network which produces a paper with lower porosity, high density and resistance properties (CAMPANO *et al*., 2018).

Physical and mechanical properties of the paper

NFC promotes an increase in the number of hydrogen bonds, which in turn increases the drainage resistance. The reason is that the cell wall surface is more exposed, meaning that the hydration rate is higher, and the flexibility of the fibers forming the sheet. The results show that the addition of nanofibrillated cellulose can be used to reduce the refining time, since resistance to drainage increased (KALIA *et al*., 2014).

The increase in °SR has also been reported in other studies. González *et al.* (2012) observed an increase from 18 to 54°SR in paper produced with short-fiber cellulose and with the incorporation of 9% nanofibrillated cellulose.

The water absorption of the paper in this study decreased, as reported by other authors such as VIANA *et al*. (2017). This can be explained by the very compact structure and low porosity of nanostructured films which decrease the water penetration.

The presence of NFC in paper increases the interaction between the cellulose fibers and promotes better rearrangement, filling the empty spaces between the fibers during paper production, and providing a more uniform and compact structure (GONZÁLEZ *et al*., 2012). This explains the formation of thinner paper with greater density with the same grammage.

This behavior was reported by Sehaqui *et al*. (2011) in studying the properties of nanostructured paper with the addition of 0 to 12% of cellulose micro and nanofibers. Potulski *et al.* (2018) verified an increase in the density of paperboard delignified from 0.43 to 0.46 g.cm^{-3} with the addition of 6% of nanofibrillated cellulose.

Greater paper density can occur due to the formation of a large amount of fiber-nanofiber bonds because the defibrillation process exposes microfibrils. Tensile and burst properties directly depend on the interfiber bonds and the formation and structure of the paper. Fibers with smaller dimension and/or nanofibrillated fibers have increased specific area and more contact points, increasing the number of bonds. The increase of these bonds raises the apparent density, as well as the tensile and burst strengths, although to a limited extent in all cases.

In a study on the potential application of nanofibrillated cellulose to improve paper strength, Sehaqui *et al.* (2011) observed a 64% increase in tensile index with the addition of 10% nanofibers. González *et al*. (2012) studied the incorporation of nanofibrillated cellulose in unrefined *Eucalyptus* cellulosic pulp. The authors found average values for the tensile index of 31.9 N.m.g^{-1} with the addition of 3% of nanofibrillated cellulose and 42.8 N.m.g^{-1} with the addition of 6%. Potulski *et al*. (2014) also found an increase in the tensile index in bleached paper of *Eucalyptus* sp. with 5% nanofibrillated cellulose.

Torres *et al.* (2005) obtained a tensile index near 50 N.m.g^{-1} for non-bleached long-fiber paper with *Schopper Riegler* grade 25º, while in this study we obtained a tensile index close to 46.8 N.m.g^{-1} with addition of 9% nanofibrillated cellulose and 2 passes through the grinder. Thus, it would be possible to maintain the same tensile strength of the paper by replacing the packaging produced with long fibers subjected to a refining process by recycled short fibers with the addition of nanofibrillated cellulose.

The tensile index found by Wistara and Young (1999) was higher than in the present study, ranging from 79.99 to 97.77 (N.m.g^{-1}). Campano *et al*. (2018) reported that the addition of nanofibrillated cellulose increased the mechanical strength characteristics of paper.

The trend of increased burst index with the addition of nanofibrillated cellulose has also been reported by other authors. González *et al*. (2012) observed mean burst indices of 2.2 kPa.m².g^{-1} and 2.9 kPa.m².g^{-1} for delignified *Eucalyptus* pulp with 3% and 6% addition of nanofibrillated cellulose, respectively. The results reported by the authors are similar to those found in this study, considering possible intrinsic differences of materials and processes.

Regarding the tear index, a significant influence of the incorporation of the nanofibrillated cellulose was observed (Table 4). The study by Manfredi *et al.* (2012) showed inferior performance to the results found for this index of *Eucalyptus* sp.

According to Viana *et al*. (2017), the process of obtaining nanofibrillated cellulose by mechanical defibrillation separates the microfibrils, but also reduces its length, which can negatively influence some properties, especially the tear resistance. Stelte and Sanadi (2009) also observed a reduction in the length of the nanofibrils due to high shear forces in treatments involving more than 10 passes through the mill Masuko grinder. Iwamoto *et al*. (2007) also noted that the increase in the number of passes caused degradation of the cellulose in nanofibrils due to the reduced degree of crystallinity and polymerization of the cellulose, causing a significant decrease in the mechanical properties.

CONCLUSION

- The physical and mechanical properties of the paper sheets were influenced by the number of passes to produce nanofibrillated cellulose by mechanical defibrillation, the nanofibrillated cellulose percentage incorporation and the combination of these two factors.
- For the physical properties, the addition of nanofibrillated cellulose in the paper reduced the thickness, increased the apparent density and decreased the water absorption.
- For the mechanical properties, the addition of nanofibrillated cellulose in the paper caused an increase in the tensile, burst and tear strength indices.
- The greatest increase in paper strength properties was observed with the addition of 9% nanofibrillated cellulose and 2 passes through the grinder.
- Nanofibrillated cellulose obtained with 10 and 20 passes for 3, 6 and 9 wt % added to the paper presented less significant increments in the strength properties.

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