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NANOFIBRILLATED CELLULOSE AS AN ADDITIVE FOR RECYCLED PAPER

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HIGHLIGHTS

In this work, recycled papers werereinforced with *Eucalyptus* sp. NFC obtained through mechanical defibrillation in a grinder.

Addition of 10 % provided the best results, with improvement of tensile, burst and tear resistance of 97, 133 and 104 %, respectively, in comparison to normal papers.

The papers' apparent density significantly increased with the addition of NFC due to the lower porosity and more compact structure presented when compared to the treatments without the addition of NFC.

The addition of NFC significantly improved the physical and mechanical properties of the recycled papers when compared to normal papers.

ABSTRACT

In this work, we studied the influence on the mechanical and physical properties of paper made of pulp from recycled cardboard and paper (printing/writing and newsprint) by adding different percentages of nanofibrillated cellulose. For each type of recycled pulp, we formed paper with incorporation of 0, 5 and 10 wt% nanofibrillated cellulose. The results showed that addition of nanofibrillated cellulose reduced the paper thickness and increased the density values. Papers with nanofibrillated cellulose presented resistance properties with values statistically superior to the treatments without addition. Addition of 10 % provided the best results, with improvement of tensile, burst and tear resistance of 97, 133 and 104 %, respectively, in comparison to normal papers. The paper produced with the recycled newspaper pulp had lower increase in mechanical properties from the nanofibrillated cellulose in relation to the papers with recycled pulp from cardboard and printing and writing paper. The considerable improvement in the mechanical properties is related to the increase of hydrogen bonds between the fibers and nanofibers, forming a dense network, resulting in greater surface area of nanofibrillated cellulose.

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INTRODUCTION

In recent years there has been an increase in the number of researches on wood in nanotechnology. Most of the studies are application of nanocellulose to improve the properties of the products or creation of new products (Julkapli; Bagheri, 2017; Kargarzadeh et al., 2017; Oliveira et al., 2018). Besides the great potential for application of nanocellulose already reported in studies, be noteworthy it is a renewable, biodegradable, low cost material contributing to a sustainable bioeconomy (Berglund; Burgert, 2018).

Considering the excellent properties and uses of nanocellulose reported in papers (Rajinipriya et al., 2018), pulp and paper industry companies around the world has invested in research and construction of new factories to produce nanocellulose.

In environmentally friendly development context, the use of recycled paper as a source of raw material for new products deserves attention. The recycling process is traditional in the sector, although the use of cellulose from the recycled fibers has been more intensive in the past few years, both for economic and environmental concerns. In the latter case, the use of recycled materials preserves forest resources and reduces the amount of material discarded in dumps and sanitary landfills (Liang et al., 1994; Sixta, 2006).

Recycled paper can become a primary or secondary source of raw material for the paper industry when used as pulp for the production of paper instead of the conventional pulp from species like the *Pinus* sp. and *Eucalyptus* sp., or when combined with conventional raw materials (Dienes et al., 2004).

On the other hand, one of the main disadvantages of reuse is that papers produced from recycled fibers have inferior resistance properties compared to papers with virgin fibers. Recycled fibers are morphologically different from virgin fibers, with shorter average length, lower hydration capacity, less flexibility and lower capacity to form interfiber bonds. These characteristics reduce the quality of certain types of paper (Spangerberg, 1993).

To reduce the effect of the previous production cycles on new products, authors suggest some methods to improve the quality of papers produced from recycled fibers, such as the addition of chemical and other agents, refinement and use of ultrasound (Wistara; Raymond, 1999; Zhang et al., 2002; Heydari; Afra 2013; Manfredi et al., 2013). In particular, the use of nanofibrillated cellulose as an agent in addition to recycled fibers can enable obtaining new nanostructured products with excellent mechanical properties.

Nanocellulose has the ability to generate stronger and more numerous hydrogen bonds between the microfibrils of the cell wall, producing a material with high resistance. Nanofibrillated cellulose or nanocellulose possess singular physical and mechanical properties that along with its low density make it an attractive material for application as coating, in the production of special films and papers, or as additive in the production of paper, to improve mechanical properties such as burst, tear and tensile strength, among others (Henriksson et al., 2008; Siró; Plackett, 2010; Jonoobi et al., 2012; He et al., (2016). NFC improve bonding and can be used as strength enhacement additive in paper and board materials.

NFC improve bonding and can be used as strength enhancement additive in paper and board material (Balea et al., 2016a). Papers produced from nanofibrillated cellulose present higher density and flexibility, can be optically transparent, have lower coefficient of thermal expansion, low porosity and show excellent barrier properties to oxygen (Fukuzumi et al., 2013; Wang et al., 2013; Fall et al., 2014.; Toivonen et al., 2015.; Shimizu et al., 2017).

This paper reports the application of nanocellulose as an additive to improve the properties of paper produced from recycled fibers. Research into the application of nanocellulose in the pulp and paper sector is recent and is still in the initial phase of exploration (Hassan et al., 2011; Luu et al., 2011; Martins et al., 2012; Brodin et al., 2014; Josset et al., 2014; González et al., 2014). Therefore, the aim of this research was to verify the influence of the addition of different percentages of nanofibrillated cellulose as reinforcement on the physical-mechanical properties of paper made with pulp from recycled cardboard, printing/writing paper and newsprint.

MATERIAL AND METHODS

Materials

Eucalyptus sp. Kraft pulp (Kappa n°= 13.0) was used to obtain the nanofibrillated cellulose. The recycled pulp was obtained from cardboard packaging (Kappa n°= 101.8), printing and writing paper (Kappa n°= 3.4) and newsprint (Kappa n°= 134.3).

Obtainment of nanofibrillated cellulose (NFC)

Nanofibrillated cellulose (NFC) was obtained from unbleached *Eucalyptus* sp. Kraft pulp produced in the laboratory. The pulp was first dispersed in water and disintegrated for five minutes, to obtain a suspension of homogenous fibers. The suspension was diluted in water at 1 wt% and submitted to mechanical defibrillation in a Masuko Sangyo Supermasscolloider grinder (MKCA6-3;

Masuko Sangyo Co., Ltd.), where it was submitted to 10 passes at 1500 rpm rotation. The resultant nanocellulose suspension presented a gel aspect, as already observed in other works (Besbes et al., 2011; Kolakovic et al., 2011).

Microscopic characterization

Transmission electron microscopy (TEM) was used to visualize the cellulose's nanofibril dimensions. The nanocellulose suspension was dripped on the surface of the screen for observation under the transmission electron microscope. The samples were left at room temperature for evaporation of the solvent to form a film. The images were acquired by a Joel JEM 1200EXII microscope (600 thousand X).

A FEI Quanta 450 FEG scanning electron microscope was used to visualize the fibers from the recycled materials (cardboard, printing/writing paper, newsprint), previously submitted to metallization.

Crystallinity index (CI)

The crystalline structure of the cellulose was determined using a Shimadzu XRD-7000 diffractometer along with the XRD-6100/7000 v 5.0 software. The scan speed was 1°/min ranging from 3 to 45°, using Cu-Kα radiation with wavelength of 0.15418 nm, voltage of 40 kV and current of 20 mA.

The crystallinity indexes of cellulose in films were calculated with average grammage of 60 g·m^{-2}, produced from the *Eucalyptus* sp. Kraft pulp before and after defibrillation in the grinder. Three repetitions were performed for each material.

Manufacture of paper with addition of nanofibrillated cellulose

The pulps were obtained through disintegration of recycled cardboard, printing/writing paper and newsprint, in the approximate consistency of 10 % in a Bauer disintegrator.

For each type of recycled pulp, paper samples were produced with incorporation of 5 and 10 wt% nanofibrillated cellulose, besides samples without addition of nanocellulose for comparison, making a total of nine treatments (Table 1). Five paper samples were produced for each treatment.

The paper samples were made with a Rapid-Köethen apparatus, with drying temperature of 90±2 °C and pressure of 40 Kpa. Five papers were produced per treatment with dried grammage of 60±3 g·m^{-2}.

After drying, the papers were acclimatized following the T402-om94 standard, at a temperature of 23±2 °C and relative humidity of 50±2 %, before the physical and mechanical tests.

TABLE 1 Treatments used for producing recycled papers.

Recycled Pulp	Treatments	Recycled Fiber (wt%)	NFC[a] (wt%)
Cardboard	E00	100	0
	E05	95	5
	E10	90	10
Printing and writing	I00	100	0
	I05	95	5
	I10	90	10
Newsprint	J00	100	0
	J05	95	5
	J10	90	10

[a] NFC: Nanofibrillated cellulose content.

Physical-mechanical tests

From the acclimatized papers were tested to determine the physical properties, moisture content (T412-om02), grammage (T410-om02), thickness (T411-om97), apparent density (T220-sp01) and mechanical properties: tensile resistance (T404-om92), burst resistance (T403-om02) and tear resistance (T414-om98).

The tensile resistance was measured through the tensile index, which corresponds to the ratio between the resistance and weight of the sample, expressed in N.m·g^{-1}. The burst index, calculated by the ratio between burst resistance and sample weight, is expressed in Kpa.m^{2}·g^{-1}. The tear index, calculated through the ratio between tear resistance and sample weight, is expressed in mNm^{2}·g^{-1}. In the realization of each physical and mechanical tests, five samples were evaluated per treatment.

Statistical analysis

The values of the physical property (apparent density) and mechanical properties (tensile, burst and tear indexes) were submitted to analysis of variance and the means were compared by the Tukey test at 5 % probability. The Bartlett test was previously performed to test the homogeneity of variances, indicating homogeneous samples.

RESULTS AND DISCUSSION

Transmission and scanning electron microscopy (TEM and SEM)

Figure 1 (A and B) presents TEM images of the fiber after being submitted to the mechanical defibrillation process with 10 passes through the grinder. The process resulted in the fibrillation of the cell walls of the fibers, producing structures that consist of clusters of microfibrils with diameters lower than 100 nm and lengths in the micrometer range (Missoum et al., 2013; Stelte; Sanadi, 2009). The fibers' cell wall is an aggregate structure of microfibrils that form bigger structures, the fibrils. During defibrillation, liberation and individualization of the surfaces previously situated inside the fibers occurs, producing microfibrils through the action of shear force on the fibrils.

The defibrillation process causes substantial alterations in the morphology of the cellulose fibers, which before the process had diameters in the dozens of micrometers, as reported by other authors who have studied species of the genus *Eucalyptus* (Oliveira et al., 2012). The TEM investigations show that after the defibrillation, the nanofibrils present average diameter varying between 20 and 30 nm, an approximate thousand-fold decrease.

The images acquired through scanning electron microscopy (SEM) referring to the recycled fibers from cardboard (A, B and C), printing and writing paper (D, E and F) and newsprint (G, H and I) are presented in Figure 2.

The recycled cardboard is formed mostly of long unbleached fibers, but it is also possible to see fibers with shorter lengths (short fibers). The recycled pulp comes from the chemical process of Kraft pulping, so it can also contain a lesser amount of high yield pulp. It is possible to observe the presence of fine and damaged fibers of different lengths, possibly resulting from the repulping process as well as the mechanical processing.

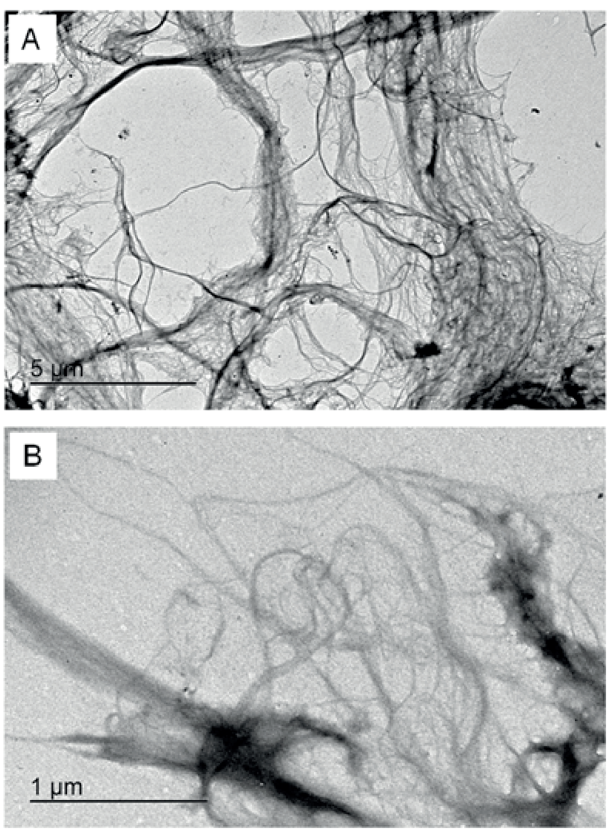

FIGURE 1 Images of cellulose nanofibrils obtained by TEM.

The recycled printing and writing paper fibers presented smaller quantities of fines when compared to other fibers with more homogeneous lengths and less structural damage in relation to the cardboard pulp. Printing and writing paper pulp is mostly formed by short and bleached fibers obtained through the Kraft process.

The newsprint pulp (Figure 2H and I) comes from the high yield pulp where the defibrillation occurs through a mechanical process. As can be observed, especially in Figures 2h and 2i, this pulp is characterized by the presence of high quantities of fines. There may also be fragments of cell walls or vessels. The fibers are not complete, and are highly damaged, meaning the material has low quality in terms of physical and mechanical resistance.

Crystallinity index (CI)

The crystallinity index was calculated by the difference in intensities between the high intensity peak (crystalline peak), located between the angles of $22° \leq 2\theta \leq 23°$, and the low intensity peak (amorphous region), located between $18° \leq 2\theta \leq 19°$ (Figure 3). The method adopted was that suggested by Segal *et al.* (1959).

The average value obtained for the crystallinity index of the nanofibrillated cellulose was 70.7 %, a value lower to that observed for the crystallinity index (CI= 81.0 %) of the cellulose in films produced before the mechanical defibrillation process. These values indicate that the defibrillation process or nanofibrillation caused damage to the crystalline structure of the cellulose, causing degradation to parts of this region (Iwamoto et al., 2008; Kalia et al., 2014).

The crystallinity degree is important, because it indicates the behavior and properties of a material. The crystalline region corresponds to the region of the fiber with greatest tensile and stretch resistance, so that higher values of the crystallinity and the polymerization degree are related to better nanocomposite resistance (Chun et al., 2011; Kulachenko et al., 2012; Lavoine et al., 2012).

Iwamoto *et al.* (2007) concluded that the degree of crystallinity of nanofibrillated cellulose produced by grinding decreases with increase in the number of passes, which can be explained by the hornification of the cellulose nanofibrils when subjected to high-speed cutting.

Physical and mechanical properties of the paper

Figure 4 (A and B) shows the behavior of physical properties thickness and apparent density for the papers with addition of 0, 5 and 10 % NFC. It can be noted that the three types of papers made from recycled pulp are thinner with increased percentage of nanofibrillated cellulose addition. The values for the treatment with 0

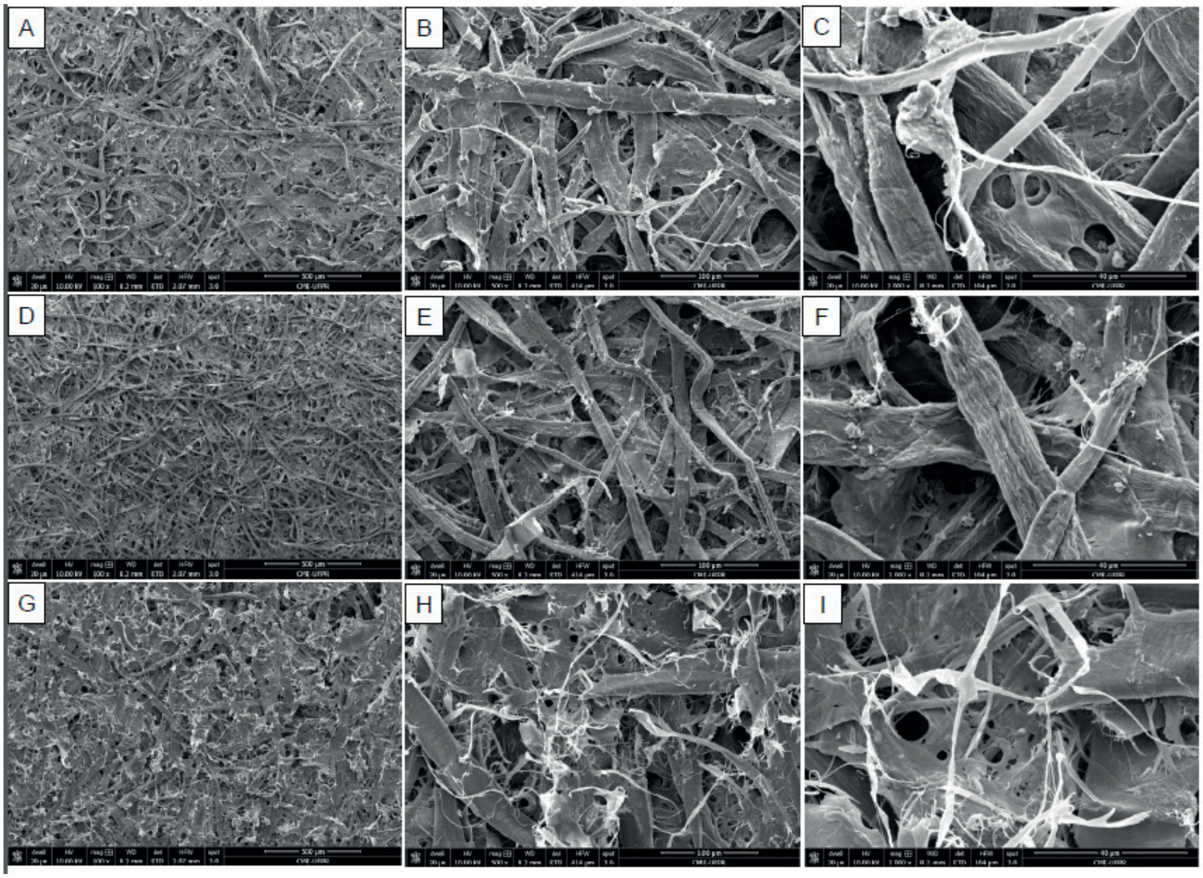

FIGURE 2 SEM images of recycled fibers from cardboard (A, B and C), printing and writing paper (D, E and F) and newsprint (G, H and I) at magnification of 100, 500 and 2000 x (left to right).

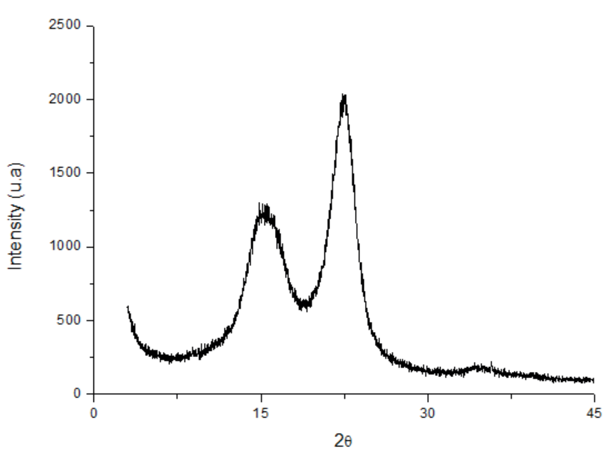

FIGURE 3 Crystallinity curve of nanofibrillated cellulose.

and 10 % NFC, respectively, were 165 and 154 μm for the cardboard, 158 and 134 μm for printing and writing paper, and 188 and 167 μm for newsprint. The highest thickness reduction observed was for papers belonging to the 110 treatment, which presented a decrease of approximately 15 % when compared to the papers treated without addition of NFC.

The apparent density, which is given by the ratio between grammage and thickness of the paper, presented an inverse tendency becoming higher with increase of NFC quantities in the papers, as also reported by González et al. (2014). The average values varied from 0.40 to 0.43 g·cm^{-3} for the cardboard, 0.40 to 0.50 g·cm^{-3}for printing and writing paper, and from 0.40 to 0.43 g·cm^{-3} for newsprint.

Papers produced from the recycled newsprint pulp presented the lowest average apparent density values. This can be explained by the presence of a greater quantity of lignin in the fiber, which provides more stiffness to the cell wall. This way, smaller accommodation of the fiber elements occurs during formation of the paper, generating a less dense and more porous structure, as indicated by the greater thickness of the papers made from recycled newsprint in relation to the cardboard, and especially the printing/writing paper.

On the other hand, the presence of NFC in papers increases the interaction between the cellulose fibers and promotes a better rearrangement, filling the empty spaces between the fibers during the production of the paper, and providing a more uniform and compact structure (Dufresne, 2012; González et al., 2012). This explains the formation of thinner papers with greater densities for the

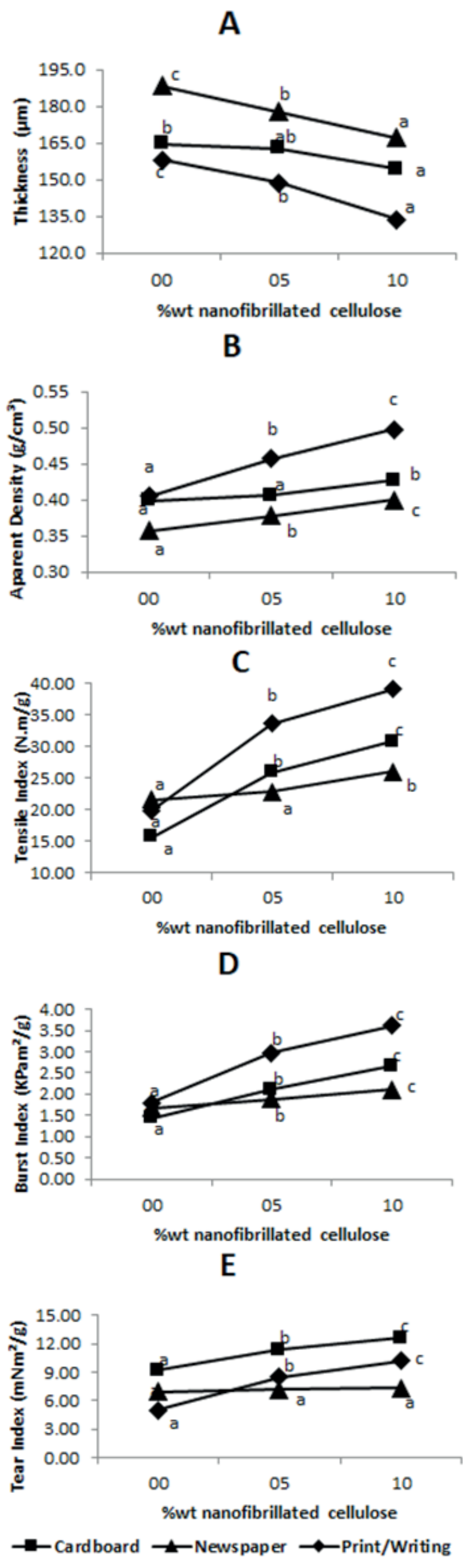

FIGURE 4 Physical and mechanical properties of recycled cardboard, printing and writing paper and newsprint in relation to the percentage of NFC. Averages followed by the same lower case letter do not differ from each other according to the Tukey test at 5% significance.

same grammage (Balea et al., 2016). The density is related to the diameter of fibers: the smaller the fiber dimensions are, the better their conformation is, producing denser papers. The increase in density and consequent reduction in porosity are important, because these aspects are closely related to improvement of the mechanical properties. The larger contact surface between the adjacent cellulose fibers provides a higher number of hydrogen bridge bonds, forming a denser network, resulting in more strength and stiffness of the paper.

Figure 4 (C, D e E) shows the tensile, burst and tear resistances in relation to the percentage of NFC. It is possible to observe that the mechanical properties of the papers, for the three types of pulps, present a considerable gain after the addition of NFC, an effect that became stronger with increased NFC percentage.

In relation to treatment E00, the tensile index increased approximately 97 % with the addition of 10 % NFC to the recycled cardboard. The values went from 15.66 to 30.83 $N.m{\cdot}g^{-1}$ and were statistically different from each other. The burst and tear indexes for the E10 treatment reached values of 2.65 $KPam^2{\cdot}g^{-1}$ and 12.56 $mNm^2{\cdot}g^{-1}$, respectively, which correspond to increases of 133 % and 38 % in resistance. Authors report increased tensile strength in 15% with addition of 3 wt% of CNFs from pine residues into the recycled paper (Balea et al., 2018).

The recycled printing and writing paper also stood out for its high average tensile and burst indexes. Furthermore, the mechanical properties also improved more markedly than the paper samples made from recycled cardboard and newsprint. With the presence of 10 % NFC in the papers, the increases were 96, 101 and 104 % for the tensile, burst and tear indexes, respectively. The values varied from 19.93 to 39.00 $Nm{\cdot}g^{-1}$ for the tensile index; 1.80 to 3.61 $KPam^2{\cdot}g^{-1}$ for the burst index and 5.00 to 10.22 $mNm^2{\cdot}g^{-1}$ for the tear index.

The papers produced from the recycled newsprint pulp presented smaller improvements in mechanical properties with the addition of NFC. The tensile, burst and tear resistances increased by 20, 26 and 5 %, respectively, with the addition of 10 % NFC in relation to the J00 treatment.

The newsprint pulp, obtained by the mechanical process, contains stiffer fibers due to the presence of a large amount of lignin (Kappa n°= 134.3). Consequently, they are less hydrated due to the hydrophobic nature of lignin and less exposure to the cellulose's hydroxyl groups, reducing the possibility of interfiber bonds between the fiber and nanofibrils. On the other hand, fibers with lower levels of lignin, which is the case of the recycled printing and writing paper, are more flexible, which provides a larger contact surface and increases the bonds between fibers and nanofibrils, promoting an increase of density and the mechanical properties (Biermann, 1996). Thus, a possible explanation for the lower gains in the resistance properties of the paper made from recycled newsprint with the addition of NFC, in comparison to the cardboard and printing and writing paper, can be the weaker bond between cellulose fibers and nanofibrils and a possible loss of nanofibrillated cellulose because of the smaller interaction between the fibrous elements. Further, the inferior quality in terms of physical and mechanical resistances of the fibers obtained through mechanical processing and the loss of hemicellulose during the repulping reduce the potential bonds of the fibers.

As discussed regarding Figure 2, the recycled newsprint pulp presents a larger amount of fines characteristics of mechanical pulps. In the recycling process, the fines lose their properties by the hornification process and behave like fillers, not contributing to the increase of the mechanical properties. Hornification is an irreversible process that causes reduction of flexibility, hygroscopicity and the bond strength between the recycled fibers (Howard, 1991).

The nanofibrillated cellulose presents high specific area, at least ten-fold that of the untreated cellulose fibers, as well as good capacity to form hydrogen bonds. Furthermore, the ratio between the fiber length and diameter is high in NFC, enabling better capacity to form a stiff and homogeneous network with lower porosity (Carrasco et al., 1996; Lavoine et al., 2012.; Campano et al., 2018). According to Spence et al. (2010a and 2010b), the improvement of paper's mechanical properties is related to the very dense network of hydrogen bonds, resulting in greater surface area obtained after defibrillation. Gonzalez et al. (2014) also highlights that porosity is an important characteristic and is closely relate to mechanical properties. Papers or films with lower porosities are more resistant due to an increase in the number of hydrogen bridge bonds.

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 resistances, although to a limited extent in all cases.

In relation to the tear resistance, it is known that this property is also influenced, among other factors, by the degree of the bond between the fibers, but mostly by the individual resistance of the fiber elements such as length and thickness of the wall. Length reduction in fibers can negatively influence the tear resistance. This could explain the lower increases observed in general for the tear indexes in relation to the tensile and burst indexes with the addition of NFC. Therefore, the addition of NFC in paper can in certain situations decrease the tear resistance, as observed by Hassan et al. (2011). Authors reported a decrease in tear strength index with the addition of nanofibrillated cellulose (Balea et al. 2016c) whereas other authors observed a increase in this property with the increase of NFC (Potulski et al., 2014). According to Balea et al. (2018) that not only the amount and the type of CNFs, but also the furnish, have influence on the tear index.

Excessive defibrillation to obtain nanofibrillated cellulose, besides reducing the fiber diameters, can cause more cutting, meaning shorter nanofibrils. Higher tensile, burst and tear index values can also be related to the length of the fiber. Furthermore, the defibrillation in the grinder can negatively affect the properties of papers by decreasing the degree of polymerization of the cellulose chains and the attack of crystalline regions in this molecule (Gomide et al., 2005). According to Stelte and Sanadi (2009), more than 10 passes to obtain nanofibrillated cellulose in the grinder can lead to reduction in the length of nanofibrils, negatively influencing the resistance properties of papers.

CONCLUSION

The addition of NFC significantly improved the physical and mechanical properties of the recycled papers when compared to normal papers. For the three types of recycled pulp, there was a reduction of thickness with the addition nanofibrillated cellulose. The papers' apparent density significantly increased with the addition of NFC.

The mechanical properties presented a considerable gain in resistance after the addition of NFC (10 wt%). In general, the highest values observed were from recycled printing and writing papers, since the tensile, burst and tear resistances were improved by 96, 101 and 104%, respectively, in comparison to normal papers. The presence of nanofibrillated cellulose provides a better conformation of the fibers occurs, forming a more compact, homogeneous and resistant structure.

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