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NANOCELLULOSE ADDITION TO RECYCLED PULPS IN TWO SCENARIOS EMULATING INDUSTRIAL PROCESSES FOR THE PRODUCTION OF PAPERBOARD

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ABSTRACT

This study assesses the incorporation of nanocellulose in a paperboard feedstock emulating two scenarios of industrial processes. It included the production of 170 g/m^2 paperboard, using mixtures of short-fiber and long-fiber fractions from recycled pulps with typical mill additives. In all cases, 3wt.% of nanocellulose was added to the pulp suspensions. The first scenario involved three types of nanocellulose addition in a mixture of 78 % long-fiber/22 % short-fiber pulps. The second scenario included the addition of two types of nanocellulose to an unrefined long fiber pulp to produce a multilayer paperboard. Drainage time and physical-mechanical properties of the handsheets were evaluated. Nanocellulose improved the mechanical properties in all cases. The tensile and burst indexes increased 19 % and 28 % in Scenario 1 and up to 60 % and 43 % in Scenario 2, respectively. The lower values in mechanical properties for Scenario 1 were attributed to the effect of the retention system. A new retention system using a cationic polymer with a high charge density produced decreases up to 79 % in the drainage time.

Keywords: Cellulose nanofibers, industrial processes emulation, microfibrillated cellulose, paperboard, recycled pulps.

1. INTRODUCTION

The application of lignocellulosic pulps to produce newsprint or printing/writing papers continues to struggle against the digital revolution. However, the packaging sector, buoyed by sustainability perceptions, leads to optimistic forecasts for pulp and paperboard. The e-commerce sector is boosting containerboard demand, and the markets will be dominated by recycled paper variants (Taylor 2019).

Recycled cellulosic materials utilization to produce containerboards for packaging involves economic, environmental, and social issues (Tarrés *et al.* 2020). Separation of paperboard wastes, such as food and liquid paperboard packaging, is essential to achieve a circular economy as it increases the quality and volume of materials available for recycling (European Paper Recycling Council 2019).

However, a paper with good strength properties requires refining cycles to promote the bonding ability, to counteract the changes in the fibers which produce their irreversible loss of flexibility (Weise and Paulapuro 1995), and the presence of additives with cationic charges, which influence the retention process of fibers and fillers in paper or paperboard (Hubbe *et al.* 2007). Although chemical additives compensate for the deteriorated quality of recycled fibers, they contribute to higher product costs (Ali 2013).

Nanocellulose additives in papermaking are a reasonable option to reduce the refining cycles (Tarrés *et al.* 2020). The types of nanocellulose used as a papermaking additive to improve the final physical-mechanical properties are microfibrillated cellulose (MFC) and lingo/cellulosic nanofibers (LCNF/CNF) (Boufi *et al.* 2016). The MFC production by purely mechanical treatment (without chemical or enzymatic pretreatments) uses a double disk refiner, PFI mill, Masuko Grinder, or homogenization (Spence *et al.* 2010, Dufresne 2013).

LCNF and CNF production combines chemical and mechanical treatments, where the most common is the application of oxidation followed by mechanical action (Saito and Isogai 2004). Nanocellulose has been widely used as reinforcement in the production of composite materials with great influence on mechanical properties such as tensile strength and elasticity (Poyraz *et al*. 2017, 2018).

In papermaking, the addition of different nanocellulose amounts was screened by other authors (Delgado-Aguilar *et al.* 2015, Espinosa *et al.* 2015, Balea *et al.* 2019, Tanpichai *et al.* 2019), showing that paper strength increases when adding more nanocellulose. Nevertheless, after a 3wt.% addition, drainage is highly compromised.

Several authors studied the effect of nanocellulose addition on recycled pulps for enhancing the final paper and paperboard properties (Saito and Isogai 2004, Balea *et al.* 2016c, Tarrés *et al.* 2020). For example, the addition of nanocellulose in old corrugated container (OCC) pulps showed increases in properties such as tensile index (TI), burst index (BI), short compression span (SCT), and significant decreases in porosity (Sanchez-Salvador *et al.* 2020).

In a mixture between old newspapers and old magazines for newsprint and recycled corrugated board pulps, nanocellulose from corn stalks increased the TI. The highest increment was for the recycled newsprint compared to OCC (Balea *et al.* 2016c). On the contrary, the addition of CNF from broke streams of the paper machine increased the TI of OCC to a greater extent concerning old newsprint paper (Balea *et al.* 2019). On the other hand, NFC and MFC decreased the drainage capacity (Ehman *et al.* 2020), and the evaluation of the retention system is required. The performance of retention agents implies studying cationic and anionic systems, polyelectrolytes, starch, etc. (Tarrés *et al.* 2018).

Despite the numerous studies about the influence of nanocellulose on paper properties, references about the application of cellulose nanofibers in the paper furnish, including the industrial process additives, are limited. This study aimed to assess the influence of nanocellulose addition on final paperboard properties, emulating the papermaking machine processes.

The study included two scenarios for paperboard production using short-fiber and long-fiber fractions of OCC recycled pulps. In all cases, 3wt.% of MFC, CNF, or LCNF from pine pulps, together with the additives used in the current industrial process, was added. The final physical-mechanical properties (density, TI, BI, ring crush test: RCT, SCT, Concora medium test: CMT, and air permeability) were measured. Finally, to ensure nanocellulose retention, different additives systems were evaluated.

2. MATERIALS AND METHODS

2.1 Materials

The MFC and CNF production was from never-dried bleached pulps. LCNF production was from unbleached commercial kraft pine pulps. Reagents used for CNF and LCNF production were sodium hydroxide (NaOH)(Cicarelli), sodium bromide (NaBr)(Sigma Aldrich), sodium hypochlorite (NaClO) (Sigma Aldrich), and 2,2,6,6-tetramethylpiperidin-1-yl)oxyl (TEMPO) (Sigma Aldrich).

The experiments consisted of two scenarios. The raw material in all cases was OCC. The repulped-OCC was fractionated in long-fiber and short-fiber fractions (corresponding to liner and corrugated medium, respectively).

The first evaluated scenario (Scenario 1) included unrefined and refined fractions, named unrefined and refined long-fiber pulp (ULFP and RLFP, respectively) and unrefined and refined short-fiber pulp (USFP and RSFP, respectively). The second scenario (Scenario 2) consisted only of ULFP pulp.

The used additives (mostly from Nalco and Solenis) were cationic starch; PAC (polyaluminium chloride); anionic flocculant with medium hydrolysis degree and high molecular weight; polyvinyl amine copolymer; copolymers of acrylamide and acrylic acid (dry strength additives); alkenyl Succinic Anhydride, ASA (sizing chemical); high molecular weight cationic latex (flocculant); (3-Chloro-2-hydroxypropyl) trimethyl ammonium chloride; 5-Chloro-2-methyl-4-isothiazolin-3-one (biocides); polyether-modified polysiloxane (defoamer).

2.2 MFC, CNF, and LCNF elaboration

The production of MFC was through a Bauer disk refiner with a recirculation system (200 mm disc diameter, 0,02 mm aperture) and a bleached kraft pine pulp for 60 min, at 1 % consistency.

The CNF and LCNF production was from bleached and unbleached kraft pine pulps. The pulps treatments were TEMPO-mediated oxidation, according to Saito and Isogai (Saito and Isogai 2004): 1 % consistency (1500 mL water), 1,6 % TEMPO on oven-dry pulp (odp), 10 % odp NaBr, and 10 mmol odp NaClO added by dropwise under continuous stirring, at room temperature. 0,5 M NaOH added maintained the pH at 10. The final point of the reaction was when there was no pH variation in the system.

The TEMPO-oxidized pulps were washed using distilled water and then passed through a colloidal grinder (at 1,3 % consistency) to break the fibril bundles. The process finalization was when the recirculation of the suspension stopped because of the material gelling. A firm gel-like suspension was obtained.

2.3 Emulation of two paperboard machine scenarios

Figures 1 and 2 show the industrial Scenarios emulated at the lab for the addition of CNF, LCNF, and MFC to the recycled pulps used as paperboard feedstocks. Scenario 1 (Figure 1) corresponds to the production of a 170 g/m^2 paperboard in a conventional Fourdrinier using different combinations of refined and unrefined short-fiber and long-fiber fractions from OCC. The additives were added to the cleaning system of the headbox circuit, except the control additives, i.e., biocide, and defoamer, which are added before the headbox.

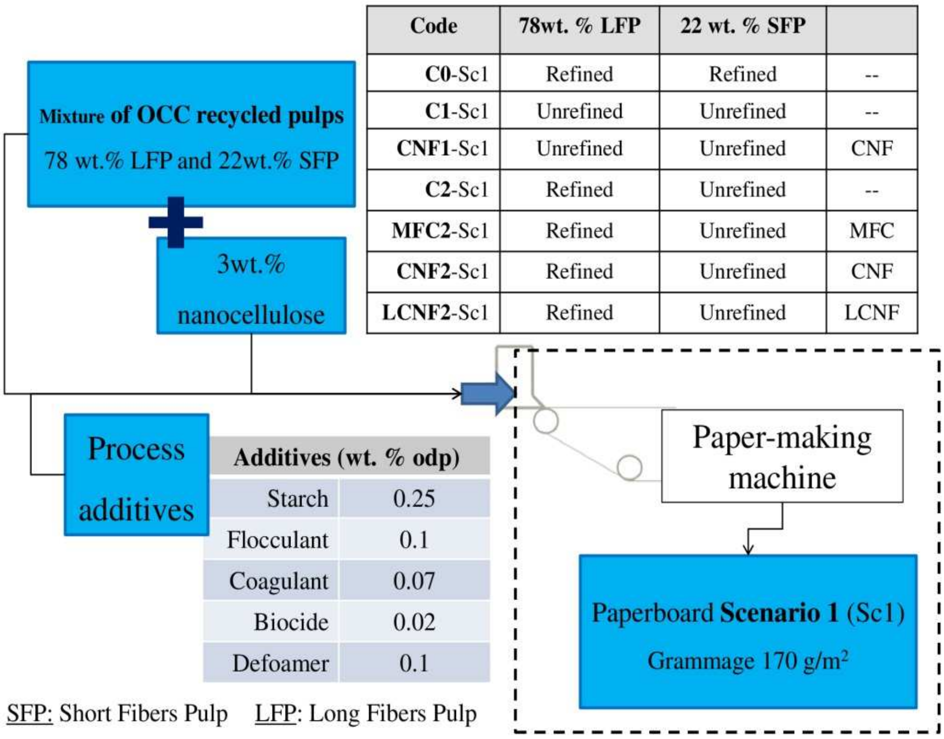

Figure 1: Scenario 1 defined for the production of paperboard using 78wt.% and 22wt.% of the long-fiber and short-fiber refined and unrefined fractions from the OCC recycled pulp.

In Scenario 1, two mixture pulps were used as controls to compare the influence of nanocellulose addition with the effect of refining. The control C0-Sc1 (both refined pulps) is the base state of the scenario, which allows us to compare the impact of adding micro and nanofibrillated cellulose as a substitute for refining. The control pulp C1-Sc1 was a mixture of unrefined pulps, and the control C2-Sc1 was a mixture of RLFP and USFP.

Figure 2 corresponds to the 170 g/m^2 paperboard produced by ULFP in a two-headboxes paper machine (Scenario 2). PAC was added before the cleaners, and the slurry containing the coagulant and the other additives was diluted with whitewater and added to the fan pump.

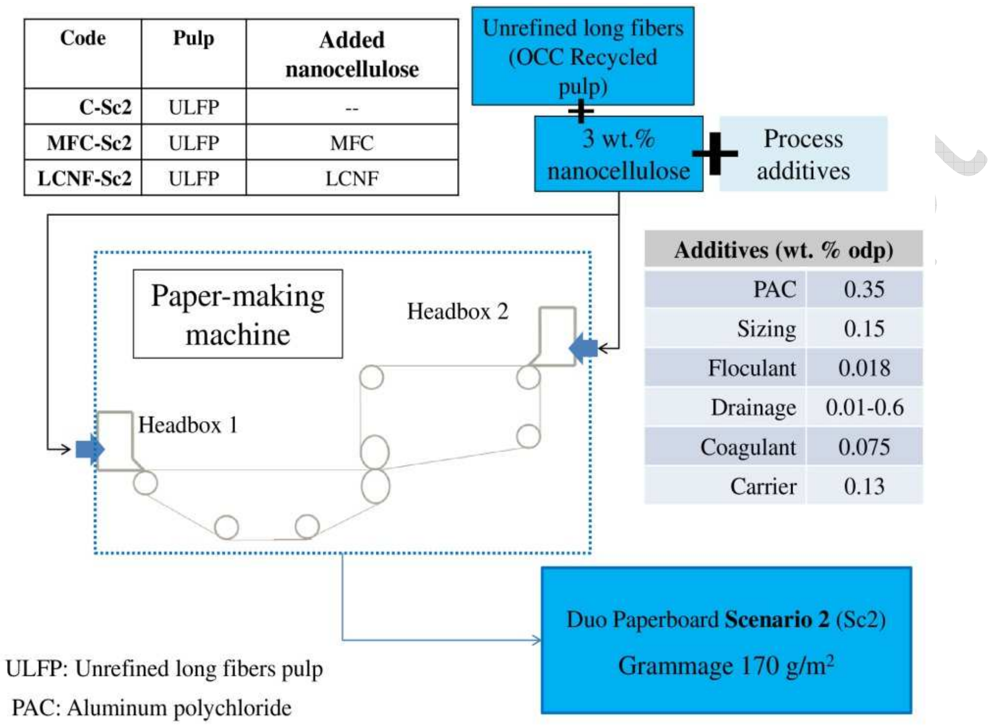

Figure 2: Scenario 2 defined for the production of 170 g/m^2 paperboard from ULFP and nanocellulose

Table 1 shows a summary of the main differences for each studied scenario. Each scenario included the OCC pulp and 3wt.% of MFC or nanocellulose suspensions in a high turbulence system for optimal mixing. Then, as shown in the figure, each additive was added in the order and doses of the respective paper machine. The scenarios also show differences in the formation stage of the papermaking machine.

The influence of nanocellulose characteristics on the properties of the corrugating medium was laboratory-evaluated in both scenarios. For it, the pulps with 3wt.% of MFC, CNF, or LCNF were dispersed for 15 min. Additives were added to each sheet using a micropipette, considering the doses per gram of pulp reported by the mills, emulating the industrial order of addition, and guaranteeing the same time of action for each one. The sheet former was adapted for white-water recirculation.

The experiments shown in Figure 2 included unrefined long-fiber pulps (C-Sc2) to produce 170 g/m^2 two-layer paperboard. LCNF and MFC from an unbleached and bleached commercial pine pulp were added. The two-layer sheets were formed by overlapping two wet sheets before pressing.

Table 1: Summary of the scenarios to be emulated in this study.

	Pulps studied	Control pulp/Type of nanocellulose	Additives applied	Type of papermaking machine
Scenario 1	**22wt.%** Refined short-fiber pulp and **78wt.%** Refined long-fiber pulp	C0	Starch, flocculants, coagulant, defoamer, and biocide	Fourdrinier (1 headbox)
	22wt.% Unrefined short-fiber pulp and **78wt.%** Unrefined long-fiber pulp	C1/CNF		
	22wt.% Unrefined short-fiber pulp **78wt.%** Refined long-fiber pulp	C2/CNF, MFC, or LCNF		
Scenario 2	**100wt.%** Unrefined long-fibers pulp	C/MFC or LCNF	PAC, ASA, flocculants, drainage aids, coagulant, and carriers	Double layer forming (2 headboxes)

During the preparation of the handsheets, the drainage time was measured according to TAPPI T221 cm-09 (TAPPI 2009). The handsheets were dried and conditioned for 24 h at 23°C and 50 %RH. Ten specimens were assembled for each property. The average values were used for the properties' increase and decrease determinations. In all cases, relative standard deviation % was less than 10%.

The measured physical properties were grammage following TAPPI T410 om-19 (TAPPI 2019) using a digital electronic scale with 0,001 g precision and air permeability according to TAPPI 460 om-16 (TAPPI 2016) by Gurley porosimeter. The measured mechanical properties were: TI according to TAPPI 494 om-13 (TAPPI 2013) using a universal testing machine (Adamel Lomargy) equipped with a 1kN load cell, BI in a Mullen tester (Perkins) according to TAPPI 403 om-15 (TAPPI 2015), bending stiffness according to TAPPI 489 om-15 (TAPPI 2015) using a Taber tester (Regmed).

Finally, medium and liner compression tests: Ring crush test (RCT), Concora medium test (CMT), and SCT according to TAPPI 822 om-16 (TAPPI 2016), TAPPI 809 om-17 (TAPPI 2017), and TAPPI 826 om-13 (TAPPI 2013), respectively. The properties are expressed as increments compared with the control pulps (C0, refined mixture; C1, unrefined mixture; C2, RLFP, and USFP mixture) to better visualize the effect of nanocellulose addition.

2.4 Evaluation of retention systems

In Scenario 1, different retention systems were tested using the CNF2 sample to improve drainage performance. The reagents and conditions are shown in Table 2.

Table 2: Evaluated retention systems for CNF2 addition (Scenario 1).

Code	**Additives**	**Dose** (Kg/t)
RS-0	Cationic polymer with high molecular weight (Reference)	1,6
RS-1	Cationic polymer with a medium charge density	1,06
RS-2	Cationic polymer with a medium charge density	2,12
RS-3	Cationic polymer with a high charge density + Colloidal silica	1,06 + 3,8
RS-4	Cationic polymer with a high charge density	1,06
RS-5	Cationic polymer with a high charge density	2,12

A dual cationic starch-colloidal silica system and a cationic polymer with a medium charge density were tested. In all cases, 2,55 kg/t starch, 0,69 kg/t coagulant, and control additives (sodium hypochlorite and a defoamer) were added to emulate the industrial process. The sample named RS-0 corresponds to the reference retention system.

Statistical analyses were performed using the Statgraphics software at a significance level of p<0,05.

3. RESULTS AND DISCUSSION

3.1 Drainage measurements and physical properties of the suspensions

The addition of nanocellulose increased the drainage time (p<0,05) in Scenario 1. The increase was similar for all types of nanocellulose added in both pulps. The °SR increased in comparison with the control in 64,6 % (MFC), 73,1 % (CNF), and 70,7 % (LCNF) when nanocelluloses were added to a mixture of unrefined short-fiber pulp and refined long-fiber pulp. However, the highest °SR value, with an increase of 82,3 %, was obtained when CNF was incorporated into the unrefined pulps mixture. In Scenario 2, the addition of nanocellulose increased the drainage time by 57,1 % in both cases.

Nanocellulose addition has a similar effect to refining concerning the drainage of pulp suspensions. The refining process generates high internal and external fibrillation, increasing bonding points between the fibers and reducing the number of pores. Also, during pulps refining, fines are produced and have a large specific surface area that increases the bonding between fibers. Besides, fines fill the spaces between fibers during handsheets dewatering (Joutsimo and Asikainen 2013; Motamedian *et al.* 2019). This effect is an undesired feature during the papermaking process since it retards paper drying and increases production costs (Ehman *et al.* 2020).

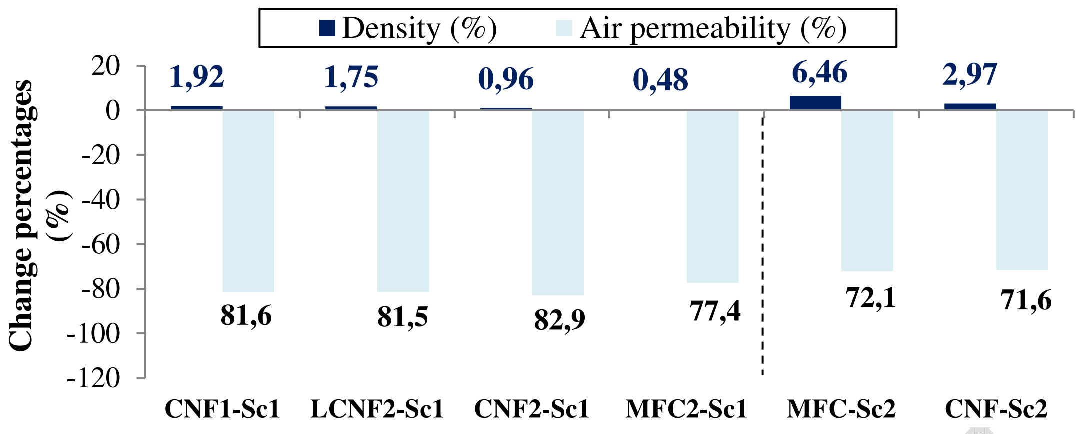

Figure 3: Changes in the density and air permeability with the addition of nanocellulose in both scenarios.

Paper density is an indirect indication of its number of pores and is expected to increase with the addition of CNF, MFC, or LCNF (Tanpichai *et al.* 2019). Nanocellulose incorporation into the furnish increases the interaction between fibers and provides a uniform and compact paper structure by filling the void spaces (Dufresne 2013). The addition of all types of nanocellulose (CNF, LCNF, and MFC) increased the handsheets density in all cases (Figure 3).

For Scenario 1, the addition of CNF to the unrefined pulps mixture (C1) produced the highest increment in density value, whereas it generated the lowest increases in the unrefined and refined pulps mixture (C2). In Scenario 2, densities reached higher increases than in the unrefined pulps mixture in Scenario 1 (Figure 3). These results agree with previous studies (Balea *et al.* 2016a, Sánchez *et al.* 2016, Lourenço *et al.* 2017, Tanpichai *et al.* 2019).

The exact values of nanocellulose retention are difficult to assess. No technique has been found to visualize the retained amount. For example, measuring nanocellulose retention after pressing could involve weighing errors. However, it is well documented that the decrease in air permeability of the handsheets compared with a control without nanocellulose addition is a good indication of its retention (Tanpichai *et al.* 2019). The effect of nanocellulose on the porosity of the paper structure is related to its high aspect ratio, leading to the formation of a stiff and homogeneous network (Lavoine *et al.* 2012, Viana *et al.* 2018). Nanocellulose incorporation into pulp suspensions decreased the air permeability of the handsheets in all cases (Figure 3).

The air permeability results measured for the reference pulp handsheets in Scenario 1 (C0, C1, and C2) showed significant differences between samples ($p<0,05$). The refined pulps mixture reached the lowest permeability. Besides, nanocellulose addition significantly decreased the permeability ($p<0,05$). For the 170 g/m^2 handsheets in Scenario 2, significant differences were observed with LCNF or MFC addition ($p<0,05$). Similar permeability values were reached when 3wt.% of CNF and MFC were added to an OCC pulp to produce recycled cardboard (Sanchez-Salvador *et al.* 2020).

3.2 Tensile, burst, and stiffness

Figure 4 shows the gain in tensile and burst indexes. In all cases, the tensile index increased with the addition of nanocellulose ($p<0,05$).

For both short-fiber and long-fiber pulps, the effect of nanocellulose addition on tensile properties was similar to that of refining. The addition of CNF or MFC in a mixture of unrefined short and refined long pulp mixture emulates the refining of the pulp mixtures (tensile indexes about 36,0 Nm/g).

The increases in mechanical properties produced by the addition of nanocellulose avoid numerous refining cycles. Refining cycles change the morphology of the fibers, i.e., decrease the fiber length by cutting and the fiber width by external fibrillation and changes in the curl and kink values because of the mechanical shear. The fibers became brittle with weak points. These morphological changes could produce decreases in strength properties (Ali 2013). Delgado Aguilar *et al.* 2015 (Delgado-Aguilar *et al.* 2015) found that the evolution of mechanical properties by adding CNF in bulk represents an alternative to classic refining. As refining progresses, the mechanical properties reach an inflection point and begin to descend, whereas, with the addition of CNF, the properties remain increasing.

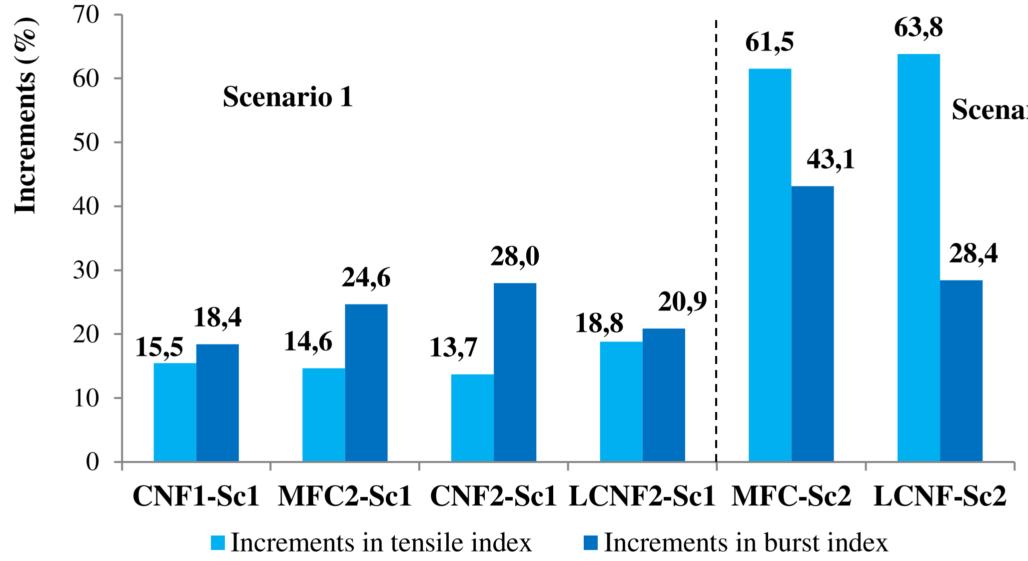

Figure 4: Increments in tensile and burst indexes with the incorporation of nanocellulose in both scenarios.

The tensile index results for samples in Scenario 1 (Sc1) were slightly lower than those obtained by Sanchéz-Salvador *et al.* (Sanchez-Salvador *et al.* 2020), where a 19,2 % increment was achieved by adding 3wt.% of nanocellulose (from northern bleached softwood kraft pulp) in OCC pulp. However, the increases were similar to that of applying 3wt.% of CNF (obtained in similar conditions during TEMPO-oxidation) in the old newspaper (ONP) (increments around 17 %) (Balea *et al.* 2019).

The elongation values varied according to the type of pulp studied. The unrefined mixture of short and long fiber (C1-Sc1) showed no significant differences when adding the CNF. However, the addition of MFC and CNF produced statistically significant increases

($p<0.05$) in the pulp mixture of unrefined short and refined long fiber (C2-Sc1). The increase in the sample when adding MFC was 25.3% concerning the control, with a similar elongation value of the short fiber and long refined fiber mixture (C0-Sc1: elongation value of 3,60%). The addition of CNF produced the highest increase in elongation values concerning the C2-Sc1 pulp (increment of 67,4%), exceeding the elongation value of refining both fiber fractions.

The tensile indexes for the 170 g/m^2 handsheets (Scenario 2-Figure 4) significantly increased with the addition of nanocellulose ($p<0,05$). The increases were similar for LCNF and MFC (increment of about 60,0 %). Increases in tensile indexes for Scenario 2 were higher than those obtained in already mentioned studies (Balea *et al*. 2019; Sanchez-Salvador *et al*. 2020). Tensile index values were comparable to increases when 3wt.% of CNF is applied to reinforce virgin eucalyptus pulp (González *et al*. 2012). The elongation value increases were 44,6 % with MFC addition to the mixture pulp in Scenario 2 and 27,1 % with the incorporation of LCNF.

Bursting strength property is relevant for packaging grade boards, especially in containerboards (Kainulainen and Söderhjelm 1999). The addition of nanocellulose and MFC in the pulp mixture increased the burst indexes in all cases in Scenario 1 ($p<0,05$), as shown in Figure 4. The values were higher, up to 10 % more than when refining the pulps mixture. The increases in burst indexes reached for the rest of the samples in this study (32,3 % of increment with 3wt.% addition of nanocellulose) were lower than those obtained by the mentioned authors. The incorporation of 3wt.% CNF obtained from recycled OCC pulp increased this property by up to 15 % when added to a mixture of OCC/ONP (Balea *et al.* 2019). In Scenario 2, the increment was higher with the addition of MFC (MFC-Sc2). The sample reached around a 43 % increment in burst index (MFC-

Sc2), in the range of the increases achieved by the mentioned authors (Sanchez-Salvador *et al.* 2020) using 4,5wt.% and 6wt.% of CNF.

The differences between the tensile and burst indexes increments obtained by the previously mentioned authors adding the same percentage of nanocellulose may be due to the slurry mixture (long and short fiber fractions), the size of the nanoparticles, and the wet end chemistry of the paper machine.

Paperboard producers commonly seek to achieve greater bending stiffness with less fiber consumption. With this objective, multilayer cardboard is produced with dense and rigid outer layers and a weaker and bulkier medium (Hagman *et al.* 2013). Bending stiffness is an indicator of the cardboard's ability to resist bending forces when a perpendicular force is applied to the free end of a strip held on one side.

The handsheets bending stiffness decreased with the addition of nanocellulose in Scenario 1 (Figure 5), whereas it did not produce any significant changes in Scenario 2. In Scenario 1, the decrease was 10 % when adding CNF to the suspension of unrefined mixture pulp (C1-Sc1).

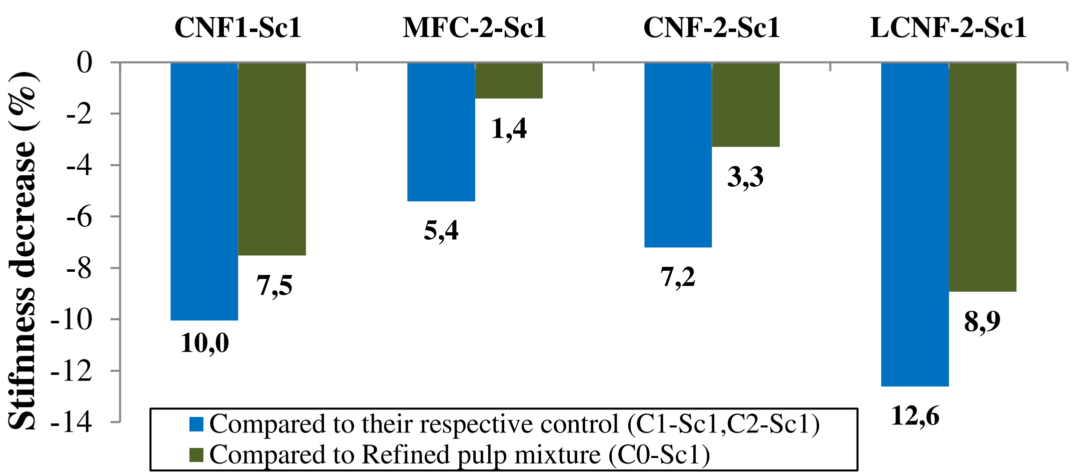

Figure 5: Changes in bending stiffness with the addition of nanocellulose in Scenario 1.

The bending stiffness in handsheets prepared with the mixture of unrefined short/refined long pulp (C2-Sc1) reached the highest value with the LCNF addition. However, the incorporation of CNF (CNF2-Sc1) and MFC (MFC2-Sc1) made it decrease by less than 10 %. The bending stiffness of samples with nanocellulose in Scenario 1 was up to 10 % lower than that of a mixture of refined pulps.

Nanocellulose and MFC act by forming bridges connecting fibers. As was previously demonstrated, this improves tensile strength and increases the fiber-fiber bond. However, in bending stiffness, bridging reduces fiber mobility (stiffening of the bonds), reducing bending energy. It has been demonstrated that bond stiffening is produced when adding 3 % fines to a chemi-thermomechanical pulp, causing bending energy reductions, even though it increases the elongation energy. The authors also highlight that the length of fines significantly influences the bending stiffness (Motamedian *et al*. 2019). In this study, MFC produces less reduction in bending stiffness. It seems that it can form longer bridges as the fibers are more distant, improving their mobility concerning the application of CNF or LCNF. It is to consider the significance of these additives' effect on the collective contribution of tensile and bending energies.

3.3 Compression strength measurements

The increments values in compression indexes are shown in Figure 6. The effect of the grammage value on RCT, CMT, and SCT properties is significant (Popil 2009). Therefore the indexes of measured properties were used. The compressive strength values represent the crushing behavior of the box and evaluate the resistance in the liner and medium layers. Specific paperboard tests were applied, namely RCT and SCT compression strength for liner and CMT for corrugated medium. Nanocellulose or MFC addition can be compared to pulp refining's effect on bonding increase, which also straightens the fibers, improving stress distribution under compressive strength and the axial compressive strength of the fibers (Ju *et al*. 2005).

In Scenario 1, RCT, SCT, and CMT were similar for all pulps without nanocellulose (including the sample of RSFP and RLFP mixture). However, the addition of CNF to the unrefined pulps mixture (CNF1-Sc1) increased the RCT, CMT, and SCT with increments of 12,3 %, 23,0 %, and 27,2 %, respectively, compared to the control (the mixture with pulp refined, C1-Sc1).

In the case of nanocellulose addition in mixtures of USFP and RLFP, the RCT varied between the types of nanocellulose. CNF2-Sc1 produced similar values as the control (C2-Sc1), whereas MFC2-Sc1 and LCNF2-Sc1 increased RCT by about 10 %. On the contrary, the addition of nanocellulose increased SCT and CMT values in all cases ($p<0,05$) compared to the control (C2-Sc1), being less than 10 % for SCT but 15,7 % for CMT with MFC (MFC2-Sc1), 22,8 % with CNF (CNF2-Sc1), and 20,3 % with LCNF (LCNF2-Sc1). However, no significant differences were found in SCT and CMT values when adding any nanocellulose type.

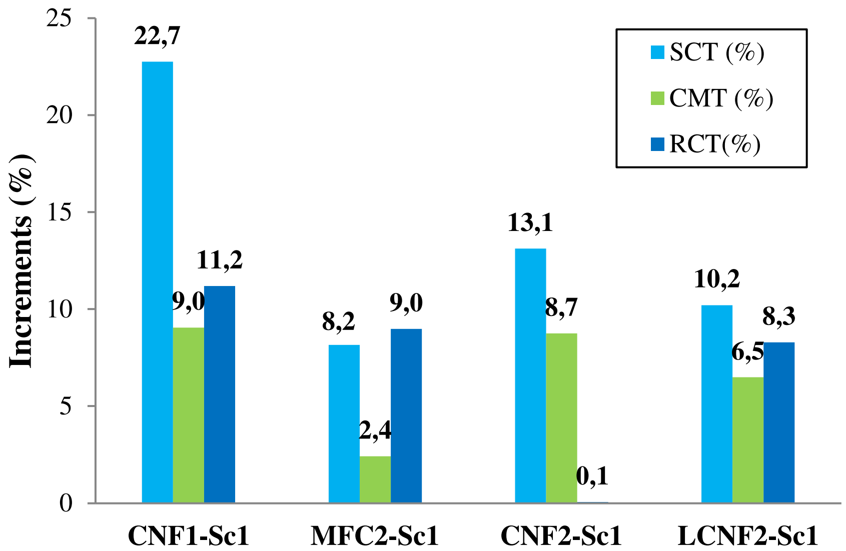

Figure 6: Increments of SCT, CMT, and RCT for the unrefined pulps mixtures as compared to the refined pulp mixture (C0-Sc1) in Scenario 1.

Figure 6 shows the differences in RCT and CMT with the addition of nanocellulose in Scenario 1, compared to a refined pulps mixture (C0-Sc1). In all cases, the values of the SCT and CMT were higher than those of a mixture of short-fiber and long-fiber refined pulps.

RCT, SCT, and CMT significantly increased with the addition of nanocellulose in Scenario 2 ($p<0,05$), with increments of 15,1 %, 22,9 %, and 36,7 %, respectively, for MFC, and 29,1 %, 10,3 %, and 23,5 %, respectively for LCNF. The increases in SCT values were similar to those obtained when 3wt.% of MFC was added to OCC pulp suspensions (Sanchez-Salvador *et al.* 2020) and higher than that of eucalyptus pulp recycled fluting paper with CNF. CMT values were similar to those of chemimechanical pulp with 3wt.% CNF (Ehman *et al.* 2020).

3.4 Evaluation of different retention systems

The increase in mechanical properties in Scenario 1 was lower than expected, possibly because of the nanocellulose loss. The traditional systems for particle retention in papermaking machines (filters in the formation section or chemical retention) are not sufficient for the complete retention of the micro/nanoparticles. Consequently, CNF or MFC may be lost, passing directly to the white waters. In addition, retention in the paper web is more difficult in the case of recycled slurries due to the anionic trash (Tarrés *et al.* 2018). So, new retention systems must be considered to maintain the nanocellulose in the paper web.

The efficiency of the drainage time during the forming stage is of utmost importance in the papermaking machine. A suitable drainage time allows for optimizing the water elimination in the forming section retaining the maximum amount of fibers, nanocellulose, and paper fillers. The strategy implemented by various authors to reduce drainage time and °SR after nanocellulose addition in pulp slurries is the use of different chemical retention systems (Ehman *et al.* 2020). They include cationic starch (González *et al.* 2012, Balea *et al.* 2016b, Sanchez-Salvador *et al.* 2020), polyDADMAC (Lenze *et al.* 2016), and polyacrylamide (PAM) (Merayo *et al.* 2017). In some cases, the combination of retention reagents also leads to a complex catching system that reduces the anionic trash (Tarrés *et al.* 2018).

The retention systems were tested on the sample CNF2 from Scenario 1 (Figure 7). In all cases, 2,55 kg/t of cationic starch was added. A biocide and a defoamer, auxiliary additives usually used in industrial processes, were also included to take account of eventual interactions.

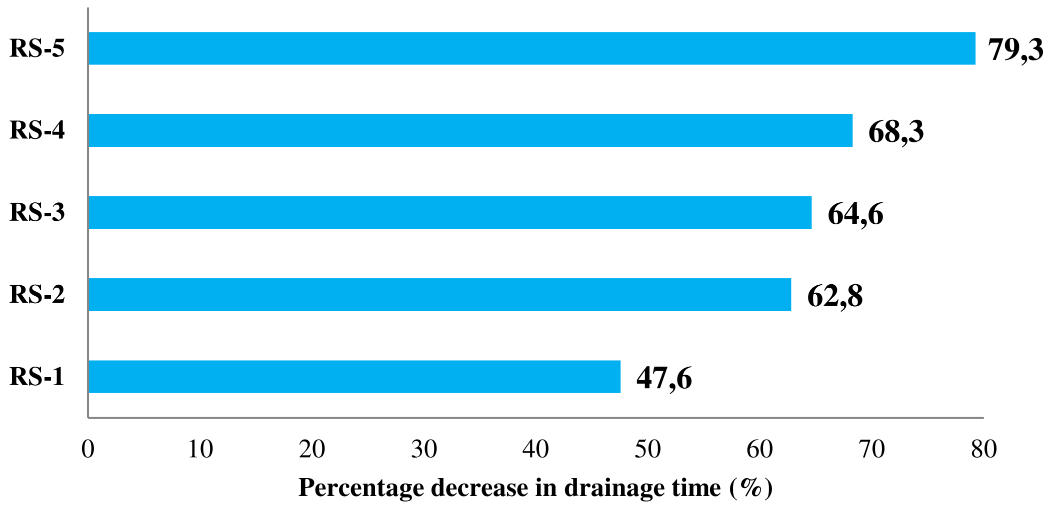

Figure 7: Changes in drainage time produced by the different retention systems using CNF2 in Scenario 1.

The drainage time decreased with the new systems tested. The highest decreases corresponded to the cationic polymer with a high charge density. The cationic polymer with a medium charge density produced significant but lower changes in retention time than the high-grade polymer.

A high dose of the high charge density cationic polymer two-folded the drainage. The addition of colloidal silica did not improve the results. The decreases in drainage time are similar to those obtained by (Merayo *et al.* 2017) when medium charge density cationic polymers were used. However, the highest drainage time decrease in this work was 10 % less than the maximum achieved by the authors with the use of poly-quaternary ammonium chloride and polyacrylamide system or polyvinilamide.

Nanocellulose efficiently retains cationic polymers because of its high surface area. Besides the improved retention, they are used as dry-strength additives, generating a higher increase in paperboard strength.

4. CONCLUSIONS

In all cases, the addition of all studied nanocellulose types (CNF, MFC, and LCNF) to a recycled OCC pulp enhanced strength properties like tensile index (> 14 %), burst index (> 18 %), RCT (< 11 %), SCT (< 22,7 %), and CMT (< 9 %). The most noticeable effect occurred when added to the short-fiber fraction. Besides, any nanocellulose or MFC improved properties, obtaining higher values than the completely refined mixture.

All types of nanofibers incorporated in papermaking furnish allow the elimination of the recycled short-fiber pulp refining. This effect enables the reduction of the long-fiber pulp in paper furnishes, a sheet grammage decrease, and an increase in the number of recycles.

The addition of CNF, MFC, and LCNF impaired the drainage of the slurries and the air permeability of the handsheets more than refining both pulps. The utilization of complex systems composed of a high-density charge cationic polymer, cationic starch, and coagulants, which can be applied at the industrial level, is recommended to decrease the drainage time.

The nanocellulose/MFC addition presents numerous benefits when applied in recycled slurries for paperboard production. The choice of the type of nanocellulose or MFC to use in a papermaking machine is associated with the production costs (water, energy consumption) and costs related to its retention on the paper web system. One solution currently proposed by mills is the on-site manufacturing of MFC using modified disc refiners, which will be evaluated in future studies.

5. AUTHORSHIP CONTRIBUTIONS

N. V. E.: Data curation, Formal analysis, Investigation,Visualization, Writing original draft, review and editing. **Y. S. A.:** Investigation, Methodology, Writing review and editing. **F. E. F.:** Conceptualization, Investigation, Methodology, Writing review and editing. **M. E. V.:** Conceptualization, Funding acquisition, Investigation, Writing review and editing. **M. C. A.:** Conceptualization, Funding acquisition, Investigation, Project administration, Resources, Writing review and editing.

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ABBREVIATIONS

BI: Burst Index
C-Sc2: Control pulp in Scenario 2
C0-Sc1: Refined mixture pulps in Scenario 1
C1-Sc1: Unrefined mixture pulps in Scenario 1
C2-Sc1: 78wt.% refined long-fibers and 22wt.% unrefined short-fibers mixture pulps in Scenario 1
CMT: Cóncora Medium Test
CNF: Cellulose Nanofibers
CNF1-Sc1: Sample C1 with CNF added in Scenario 1
CNF2-Sc1: Sample C2 with CNF added in Scenario 1
LCNF: Lignocellulose Nanofibers
LCNF-Sc2: Sample with LCNF added in Scenario 2
LCNF2-Sc1: Sample C2 with LCNF added in Scenario 1
MFC: Microfibrillated Cellulose
MFC2-Sc1: Sample C2 with MFC added in Scenario 1

OCC: Old Corrugated Container
RCT: Ring Crush Test
RLFP: Refined Long fiber Pulp
RSFP: Refined short Fiber Pulp
Sc1: Scenario 1
Sc2: Scenario 2
SCT: Short Compression Test
TI: Tensile Index
ULFP: Unrefined Long Fiber Pulp
USFP: Unrefined Short Fiber Pulp