

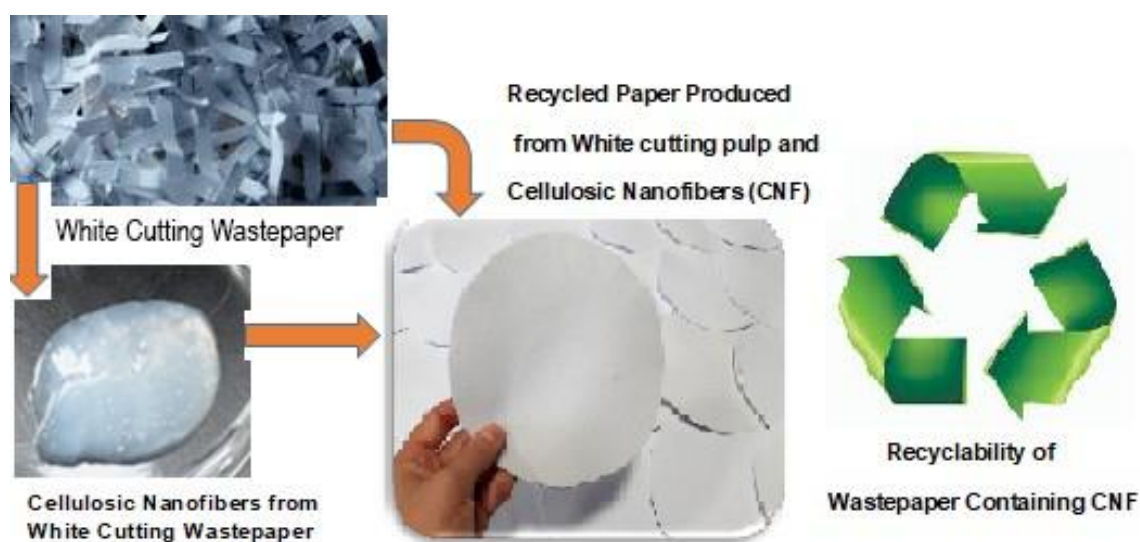
Recyclability of Wastepaper Containing Cellulose Nanofibers

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GRAPHICAL ABSTRACT



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Cellulosic/lignocellulosic nanofibers (CNF/LCNF) are value-added products which recently have been considered widely. This study focused on valorization of waste by making CNF from white cutting wastepaper (WCW), which was confirmed by the transmission electron microscope images. The CNF present in the paper recycling process showed significant effects on the final product and process aspects. Addition of 5% CNF (based on dry weight of pulp) to the recycled fibers improved the strength properties as well as the fines retention. But in contrast, the drainage decreased from 303 to 188 mL CSF. Finally, considering the importance of wastepaper history on the quality of recycling process and final products, as a new vision, this study focused on recyclability of wastepaper which has experienced CNF as an additive in their history of papermaking. In this respect, the papers reinforced with nanofibers in their history, after several recycling, experienced less reduction in the tensile and tear indices and fines retention. In summary, it seems that the presence of CNF in the wastepaper can slow down the reduction of product density and process properties during several recycling cycles and so restore their potential for subsequent papermaking cycles.

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Keywords: Cellulose nanofibers; Wastepaper; Recycling times; Drainage; Fines retention; Mechanical properties

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INTRODUCTION

Nowadays, the papermaking industry has been devoting special attention to recycled fibers. Wood shortages, increasing demands for paper products, increasing environmental concerns, high volume of paper in municipal wastes, and economical aspects have strengthened this emphasis (Viana *et al.* 2018; Amiri *et al.* 2019).

As an advantage, most of paper-based products can be recycled several times. But especially in chemical papers, recycling can affect the product and process properties negatively because of different phenomena such as fibers hornification, pits closure in fiber walls, and fibers shortening (Ek *et al.* 2009).

The history of recycled fibers, *i.e.*, the past phenomena happened for producing a paper sheet (such as applied additives and performed processes, *etc.*) will affect the properties of final recycled paper sheet (Benitez *et al.* 2014; Aliaga *et al.* 2015; Pivnenko *et al.* 2015; Runte *et al.* 2015; Sonmez *et al.* 2022). For example, mechanical treatments such as refining can introduce more hydrogen bonding sites on fibers, producing sheets with more tensile index, but experiencing severe hornification in recycling process (Kermanian *et al.* 2013; Delgado-Aguilar *et al.* 2015; Yilmaz *et al.* 2022). Another

approach is to treat recycled fibers with different additives in papermaking process for various purposes (Sabazoodkhiz *et al.* 2017; Gal *et al.* 2023). These additives can affect the next recycling process negatively or positively. For instance, components of anionic trashes or stickies in recycled pulp have an origin of additives previously added in papermaking process (Hubbe *et al.* 2006; Rahmaninia and Khosravani 2015). Also, in contrast, it has been reported that some of additives in papermaking (such as some of cationic biopolymers) can affect positively the recycled pulp (Zhang *et al.* 2002).

Cellulosic or lignocellulosic nanofiber (CNF/LCNF) is a class of biobased papermaking additive that has opened some alternative approaches for industrial applications (Bardet and Bras 2014; Tarrés *et al.* 2021; Karthika *et al.* 2022; Bastida *et al.* 2023; Liu *et al.* 2023; Milani *et al.* 2024). Cellulose nanofibers are flexible filament-like structures with a width of 1.0 nm to 100 nm and a length of several micrometers (high aspect ratio). They are comprised of alternating crystalline and amorphous regions and can be produced from different lignocellulosic resources (Wu *et al.* 2014). The use of CNF for various applications has received much attention. This product is considered due to various capabilities such as biodegradability, renewability, high specific strength, ability to create abundant hydrogen bonds, *etc.* in various industries and applications (Balea *et al.* 2019). Kajanto and Kosonen (2012) considered the usefulness of nanofibers as a reinforcing agent in the paper. In fact, these nanofibers have relatively high strength and rigidity; they can be used in various sectors such as papermaking, composites, cement, packaging, electronic devices, coatings, and other biomedical or automotive applications. As mentioned, CNF/LCNF, as an effective additive, has received much attention in the papermaking industry. In fact, this product with its high specific surface area and special structure can help increase the strength of the paper fiber network (Merayo *et al.* 2017). Overall, the advantage of CNF has opened significant possibilities for paper reengineering, feature expansion, and the development of a wide range of new products (Rantanen *et al.* 2015).

The current research, firstly, has focused on valorization of waste by producing CNF from white cutting wastepaper (WCW). Also, the application of the prepared CNF in upgrading the quality of recycling process and recycled products was studied. Furthermore, as a novel vision, the recyclability of wastepaper containing CNF was investigated. Overall, the current study tried to provide clearer view and information to relevant sections (investigators, paper mills *etc.*) about applying and performance of CNF/LCNF as an internal additive in papermaking and also the effects of its presence in wastepaper during next recycling processes.

EXPERIMENTAL

Materials and Methods

White cutting wastepaper (WCW) was collected locally from Noor city, Iran.

Wastepaper Repulping

After determination of wastepaper moisture content (8%), the repulping process was done using a laboratory Hollander beater at neutral pH according to TAPPI T 200 sp-01 standard. For this purpose, 360 g (based on dry weight) of WCW was soaked one night in 7 L deionized water (conductivity $\approx 1.9 \mu\text{S}/\text{cm}$). The soaked WCW was transferred to the beater, and the final volume of suspension reached 23 L using deionized water. The disintegrating and refining processes were done in 30 min and 2 min, respectively. The

pulp freeness was adjusted to about 300 mL CSF. The freeness of this pulp was determined according to TAPPI T 227 om-99 standard using CSF tester. The prepared pulp was named “1st time recycled pulp (CNF- free)” (Table 1) and was kept in a cold storage room for the next steps.

Preparation of Cellulose Nanofibers

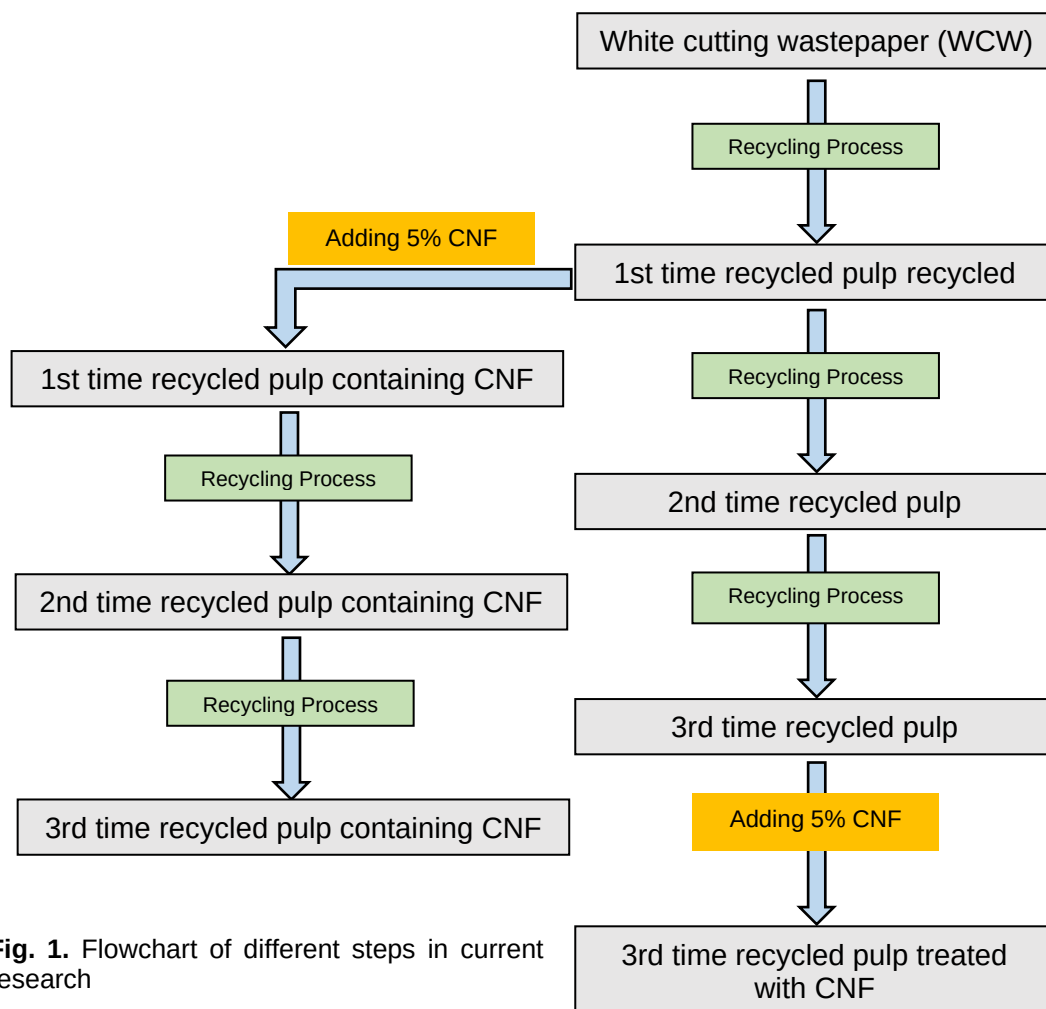
In the current study, white cutting wastepaper was considered for production of CNF. For this purpose, the recycled pulp was washed on a screen with 200 mesh size for removing the fine particles, especially fillers. Then, the obtained pulp with 1% consistency was treated with mild acidic pretreatment applying pure 8% sulfuric acid with a weight ratio of 8% (acid to dry fibers) at a temperature of 70 °C for 1 h. The treated fibers were washed with enough distilled water for reaching neutral pH. This prepared pulp was used for making CNF in Nano Novin Polymer Co. with passing the pretreated pulp 3 times through super grinding disk machine (MKCA6-2; Masuko Sangyo Co., Ltd., Kawaguchi, Japan). The structure of prepared CNF was assessed by transmission electron microscopy (TEM, Philips EM 208S, USA). It should be mentioned that the conditions of preparing CNF were chosen according to the different pre-tests. Furthermore, for better visualizing the nanofibers and fibers, their images with two magnifications (100 and 400) were prepared using light microscope (HP320, Taiwan) and the average fibers length and fibers diameters were determined by using Image tool-2.0 software.

Applied Treatments

Table 1 shows the treatments used in the current research. In this respect, the 1st time recycled pulp (CNF- free) was used for making 60 g/m² paper according to TAPPI standard, T 205 sp-02. Some of the produced papers were selected for evaluation of different properties and the others were repulped to prepare “2nd time recycled pulp (CNF-free)” (Table 1). The same procedure was applied for 2nd time recycled pulp (CNF- free), i.e., papermaking was done and some of handsheets were selected for determination of properties and the other part was repulped to prepare the “3rd time recycled pulp (CNF-free)”. This pulp was used for preparing handsheets and determination of various paper properties. Furthermore, for considering the recyclability of wastepaper containing CNF, firstly, 5% CNF (based of oven-dried pulp) was added to 1st time recycled pulp (CNF-free). For this purpose, CNF concentration was reduced to 0.5% using distilled water and mixed for 2 min at 2000 rpm by a mechanical mixer. This CNF suspension was then applied to the recycled pulp with 0.5% consistency. The mixture was agitated with mechanical mixer at 1500 rpm for 10 min. This pulp was named “1st time recycled pulp containing CNF”, and the hand-sheets were made of this pulp. Some of handsheets were selected for determination of different properties and the others were repulped for producing “2nd time recycled pulp containing CNF”. The 3rd time recycled pulp containing CNF also was made using the same process. To gather more information about the performance of CNF in improving the properties of recycled paper, 5% CNF was added to the “3rd time recycled pulp (CNF-free)” (step 7). Different steps of current research are shown in Fig. 1.

Table 1. Pulp Treatments Used

Steps	Treatments
1	Recycling of WCW (1 st time recycled pulp (CNF-free))
2	Recycling of step one papers (2 nd time recycled pulp (CNF-free))
3	Recycling of step two papers (3 rd time recycled pulp (CNF-free))
4	Adding 5% CNF to the WCW pulp from step one (1 st time recycled pulp containing CNF)
5	Recycling of step four papers (2 nd time recycled pulp containing CNF)
6	Recycling of step five papers (3 rd time recycled pulp containing CNF)
7	Adding 5% CNF to the pulp from step 3 (3 rd time recycled pulp treated with CNF)

**Fig. 1.** Flowchart of different steps in current research

Handsheet Properties

As mentioned, handsheets with 60 g/m² grammage were prepared according to T 205 sp-02 TAPPI standard. The properties of pulp and prepared papers were determined according TAPPI standards. Table 2 shows the measured properties and their related standards.

Table 2. Handsheet Properties and their Related Standards

Property	Standard
Tensile index	TAPPI T 494 om-01
Tear index	TAPPI T 414 om-98
Drainage of pulp	TAPPI T 227 om-99
Fines content/Fines Retention	TAPPI T 261 cm-00
Ash content of recycled WCW pulp	TAPPI T 211 om-93
Thickness of paper	TAPPI T 411 om-05
Grammage of paper	TAPPI T 410 om-02
Opacity	TAPPI T 425 om-01
Water Absorption (Cobb test-60 seconds)	TAPPI T 441 om-04

Statistical Analysis

For determination of the properties, five replications were used. Completely randomized design (CRD) was applied for statistical analysis and Duncan's multiple range test (DMRT) for categorizing the averages. The Duncan grouping was alphabetically, ascribed. The data with the same alphabetical grouping (same letter) didn't have significant statistical difference (5% error level). SPSS software (IBM, version 16.0, Armonk, USA) was used for statistical analysis.

RESULTS AND DISCUSSION

Ash Content

For better characterization of recycled WCW pulp, the ash content, which is a good indicator for filler or mineral content, was determined. Considering five replications for this experiment, the average ash content of recycled WCW pulp was found to be about 16.28%. Also, the mechanical process of producing CNF is so sensitive to minerals. Hence for removing the minerals present, the recycled pulp was washed on a 200-mesh screen. The ash content of washed recycled pulp was about 4.7%.

Morphological Structure of Recycled WCW Fibers and Nanofibers

The morphological structure and dimensions of recycled fibers and nanofibers obtained from white cutting wastepaper were analyzed using a light microscope. Figure 2 shows the images of the 1st time recycled fibers and nanofibers with 100 x and 400 x magnifications, respectively. The results showed that the average length of fibers in the 1st time recycled pulp (step 1) was about 1.65 mm, and its average diameter was about 32 μ m. Also, the average length of fibers in the 2nd time recycled pulp (step 2) and the 3rd time recycled pulp (step 3) experienced reduction and were about 1.08 mm and 0.94 mm, respectively. This reduction can be attributed to the effect of recycling process and its effect on fiber shortening. Also, TEM technique furnished accurate study of the CNF structure and dimensions.

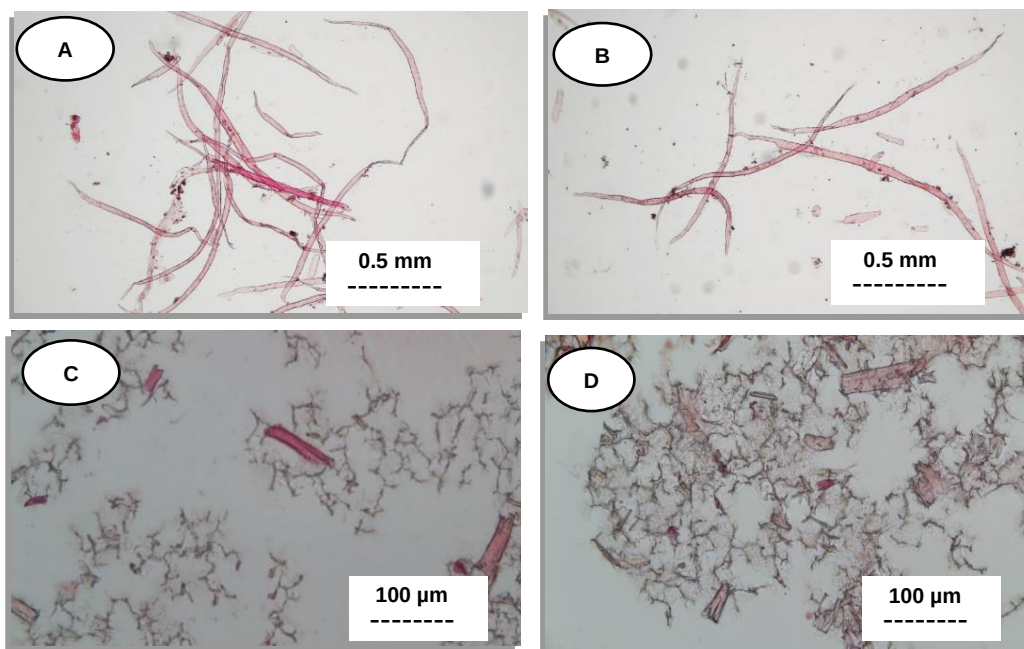


Fig. 2. Light microscopic images of 1st time recycled fibers (A and B with 100x magnification) and recycled nanofibers (C and D with 400x magnification) stained with Safranin

Figure 3 shows the TEM image of CNF from recycled WCW fibers. According to the results, the diameter range of produced nanofibers was in the range of 25 to 50 nm, which indicates the successful process of nanofibers production. As explained previously, the 1st attempt of producing nanofibers from recycled WCW fibers required high energy consumption. In the next round, for reducing energy consumption, mild acid pretreatment was applied. It seems that experiencing high energy consumption for producing CNF from untreated recycled fibers might be due to the happening of shrinkage and hornification in recycled fibers (Tarrés *et al.* 2021; Karthika *et al.* 2022).

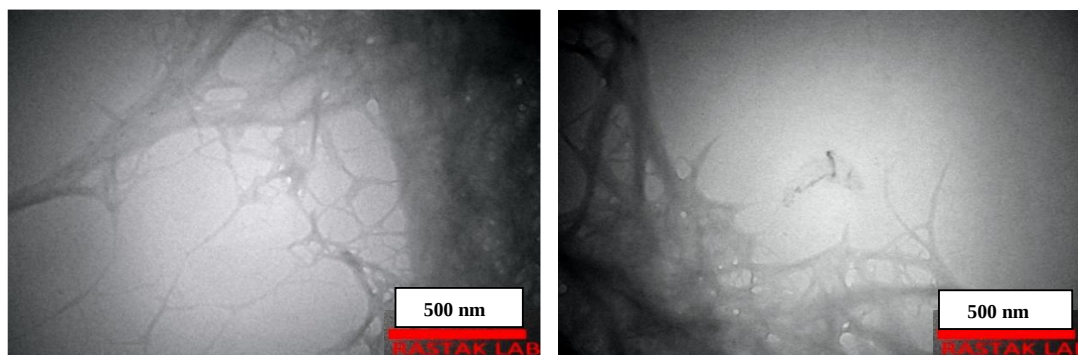


Fig. 3. TEM images of WCW nanofibers

Pulp Drainage

Drainage is an important factor for analysing the papermaking process. This property simulates how water leaves the pulp due to gravity on the wire of papermaking machine. The higher the pulp drainage, the less energy consumption in press and dryer sections. This phenomenon leads to higher paper machine speed, higher productivity, and less production costs.

Figure 4 shows the pulp drainage in different treatments. As can be seen, the amount of drainage in CNF-free treatments decreased from 303 mL CSF in the 1st time recycled pulp to 272 mL CSF in the 3rd time recycled pulp. The reason can be attributed to the shortening of the fiber's length and the corresponding increase in fiber fines during the recycling process.

Although it was expected that the recycling would decrease the fibrillation of the fiber due to the hornification (Kermanian *et al.* 2013) and increase the drainage, it seems that fiber shortening and increasing in fiber fines content, reversely decreased the pulp drainage. Also, addition of 5% nanofibers to the 1st time recycled pulp drastically decreased the drainage (38% reduction). The result can be attributed to nanofibers structure and their high specific surface area, which can increase fines retention and subsequent accelerating in blockage of fiber mat and wire openings. The following results of fines retention in Table 3 confirm this phenomenon. Furthermore, the possibility of more water retention value due to nanofibers shouldn't be neglected (Balea *et al.* 2016; Osong *et al.* 2016). In fact, the possibility of hydrogen bonding with water may cause the drainage reduction (Yousefhashemi *et al.* 2019). In addition, recycling the papers containing CNF (steps 5 and 6) increased the drainage from 188.66 mL CSF to 224 mL CSF and 236 mL CSF, respectively. The observed results can be because of removing some of fiber fines and also nanofibers during recycling process. However, it should be noted that this drainage increment had not reached the level of treatments without CNF.

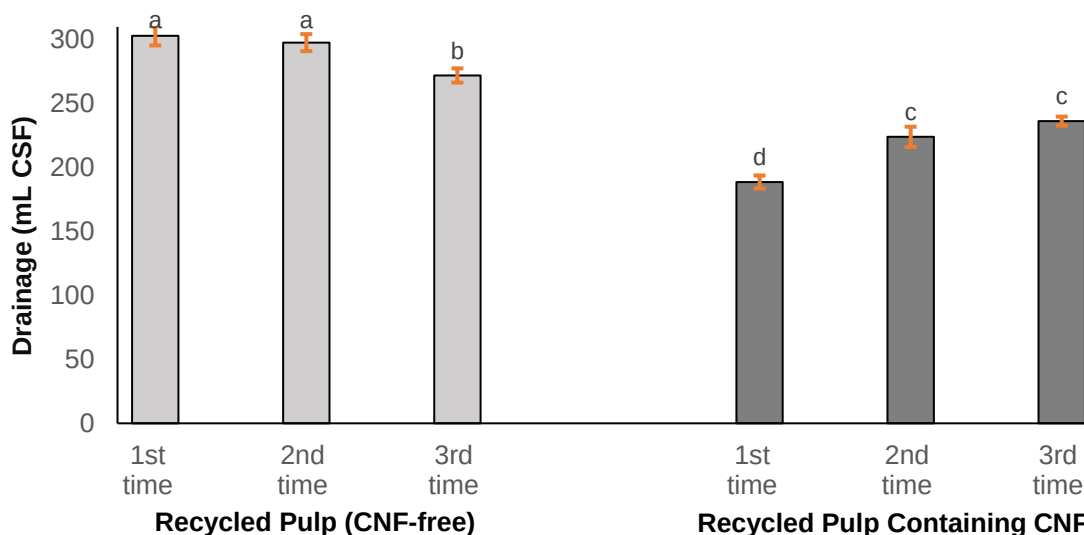


Fig. 4. The effect of applied treatments on the pulp drainage. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

Fines Retention

The retention of fines is an important process outcome in the paper industry that directly can affect other factors such as production efficiency, quality of whitewater and also effluent and final product quality (Sabazoodkhiz *et al.* 2017). Sometimes with increasing fines retention, the pulp drainage and mechanical properties of final paper may be adversely affected, which should also be taken into account (Amiri *et al.* 2019). Usually, in paper structure, especially with high grammage, the fiber fines are physically trapped between the fibers during the formation of the paper sheet, which leads to increasing fines retention. Also, the use of some additives in the pulp suspension causes more fines and

fillers to bind together as well as with fibers and, thus, increasing in the final solid retention (Taheri *et al.* 2022).

As can be observed in Table 3, the addition of 5% CNF in 1st time recycled pulp increased fines retention by up to 18.5% (Changing the retention from 39.22% to 48.15%). These results are consistent with the results of some other studies (Balea *et al.* 2016; Hollertz *et al.* 2017).

It was also observed that with increasing the recycling times in CNF-free papers, the fines retention experienced a reduction. The results can be attributed to decreasing in the physical trapping of fines (as one of the main mechanisms of fines retention) in the fibers mat structure phenomenon (Milani *et al.* 2024).

A similar phenomenon was observed in the recycling of papers containing CNF (Steps 5 and 6). Here, in addition to the previous reason, removing some of nanofibers from pulp during the recycling process and so less CNF retention in produced papers might be another reason for the mentioned results. However, it should be noted that even after removing some of nanofibers in steps 5 and 6, the remained ones could positively affect the fines retention compared to CNF-free treatments (Steps 2 and 3). Furthermore, as mentioned for gathering more data about the performance of nanofibers in improving the properties of recycled paper, 5% CNF was added to 3rd time CNF-free recycled pulp (pulp produced in Step 3). The results showed that CNF surprisingly could improve the fines retention of pulp that experienced three times of recycling.

Table 3. Fines Retention in Different Treatments

Steps	Treatments	Fines Retention (%)
1	Recycling of WCW (1 st time recycled pulp (CNF-free))	39.2
2	Recycling of step one papers (2 nd time recycled pulp (CNF-free))	25.3
3	Recycling of step two papers (3 rd time recycled pulp (CNF-free))	13.3
4	Adding 5% CNF to the WCW pulp from step one (1 st time recycled pulp containing CNF)	48.2
5	Recycling of step four papers (2 nd time recycled pulp containing CNF)	37.6
6	Recycling of step five papers (3 rd time recycled pulp containing CNF)	25.0
7	Adding 5% CNF to the pulp from step 3 (3 rd time recycled pulp treated with CNF)	39.2

FE-SEM Images of Handsheets

Figure 5 presents the FE-SEM images of end papers prepared from different recycled fibers (1st time CNF-free recycled pulp (Step 1), 1st time recycled paper treated with 5% CNF (Step 4), 3rd time recycled paper containing CNF (Step 6), and 3rd time recycled pulp treated with CNF (Step 7)).

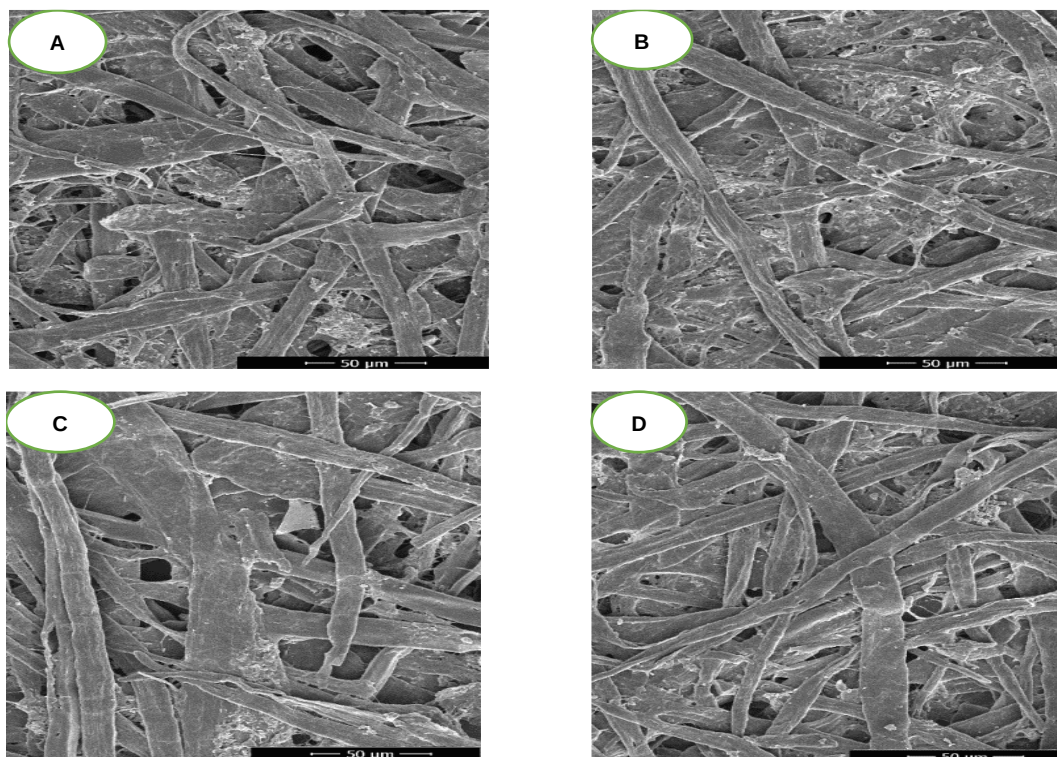


Fig. 5. The FE-SEM images of end papers: 1st time CNF-free recycled pulp (step 1) (A), 1st time recycled paper treated with 5% CNF (step 4) (B), 3rd time recycled paper containing CNF (step 6) (C), and 3rd time recycled pulp treated with CNF (step 7) (D)

As can be seen, addition of 5% CNF to 1st time recycled pulp (Step 4) (Fig. 5B) made papers with denser structure (compared to 1st time CNF-free recycled paper (Fig. 5A)) which might be because of creating more bonding in paper structure and even more retention in final mat structure (Kajanto *et al.* 2012; Merayo *et al.* 2017; Balea *et al.* 2019). Also, the open structure of end papers (Fig. 5C) produced from 3rd time recycled paper containing CNF (Step 6) shows the obvious effect of recycling times on bonding reduction among fibers and decreasing of density compared to paper produced from 1st time recycled pulp treated with CNF (Fig. 5B) (Kermanian *et al.* 2013). Furthermore, according to Fig. 5D, addition of 5% CNF to 3rd time CNF-free recycled pulp (Step 3) produced papers that had a relatively denser and uniform structure compared papers produced in step 6 (Fig. 5C).

Apparent Density and Thickness

Apparent density and thickness are important physical properties that directly or indirectly affect different mechanical, physical, and optical properties of papers (such as their tensile strength, modulus of elasticity, and light scattering coefficient) and depend on the conditions of the papermaking process (such as fiber type, refining, pressing, *etc.*) (Ek *et al.* 2009). Figures 6 and 7 show the apparent density and thickness of different papers. As can be seen, by increasing the recycling times, the apparent density and thickness in both CNF-free and CNF-treated papers (Steps 4, 5, and 6) decreased and then increased. As mentioned before, increasing the recycling times would negatively affect the fiber flexibility and fiber bonding due to hornification phenomenon and lead to forming the thick and porous sheet of fibers with lower density and higher thickness (Kermanian *et al.* 2013).

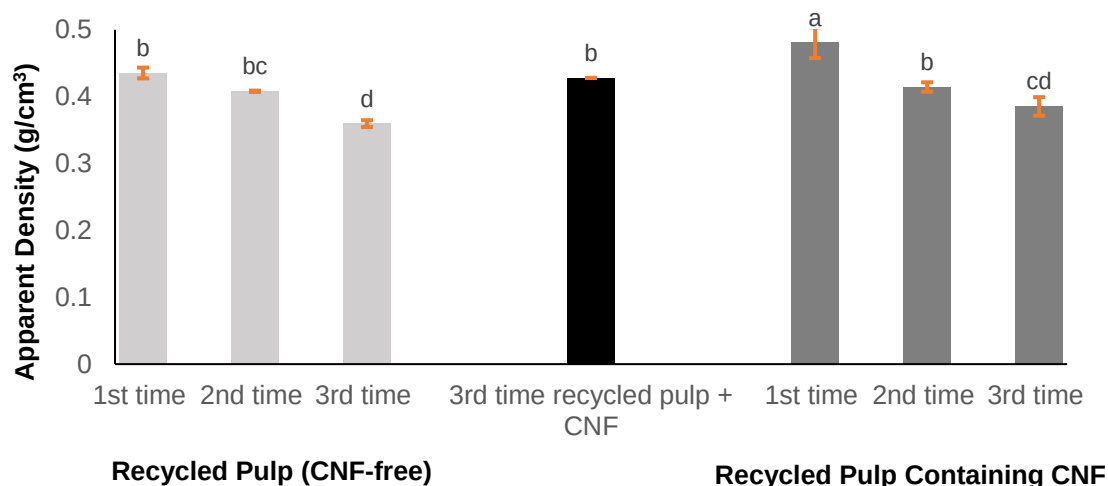


Fig. 6. Changes in the apparent density of papers made under different treatments. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

Also, the presence of CNF in the structure of 1st time recycled paper (Step 4) improved its bonding and therefore its density even more than papers made of basic pulp (Step 1). The same results were reported by Viana *et al.* (2018). Also, as a new achievement, the presence of CNF in wastepaper had a slightly beneficial effect on restoration of bonding after experiencing several times recycling. However, by increasing the recycling times (Steps 5 and 6), the effect of CNF in restoring the bonding ability was decreased, but the papers reinforced with nanofibers in their history, after several recycling, experienced less reduction in the density comparing the CNF-free papers.

It seems that removal of CNF from pulp (because of its nano-size) after each recycling process and the negative effects of drying phenomenon on CNF bonding ability are the reasons for the above results. Furthermore, adding the CNF directly to pulp which underwent various recycling processes (Step 7) made better bonding restoration in end paper structure. Moreover, as a comparison, adding 5% CNF to the 3rd time recycled pulp significantly increased the density and improved it up to 1st time recycled CNF-free pulp (Yousefhashemi *et al.* 2019). The reduction of thickness showed a good coordination with density increment. The results showed that CNF successfully improved the density of papers produced from recycled fibers (Steps 4 and 7). Considering the inverse relationship between density and thickness, the thickness results obtained are consistent with the above relation and formula.

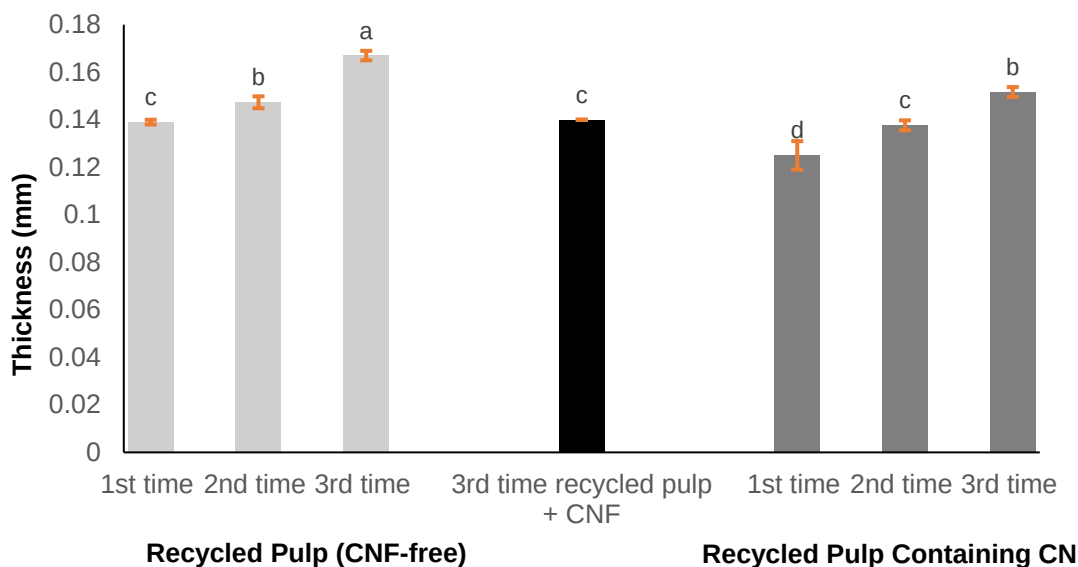


Fig. 7. Effect of applied treatments on the thickness of handmade paper. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

Tensile Index

Tensile strength is one of the important mechanical properties of paper sheets. Its value can be significantly affected by fiber-fiber and fiber-fines bonding. Figure 8 shows the tensile index of handsheets made in this study. According to the results, by increasing recycling steps in CNF-free treatments (Steps 1, 2, and 3), the tensile index decreased. The recorded reduction can be attributed to the fibers' hornification, blocking the hydroxyl groups (important cellulose functional groups), reducing the flexibility of mentioned fibers, and increasing the fiber fines content in the recycled pulps (Kermanian *et al.* 2013). However, adding 5% CNF to 1st time recycled pulp (Step 4) improved the tensile index significantly (28.8% increase). The result can be attributed to increment of hydrogen bonding in the paper structure by applying CNF (González *et al.* 2013; Viana *et al.* 2018).

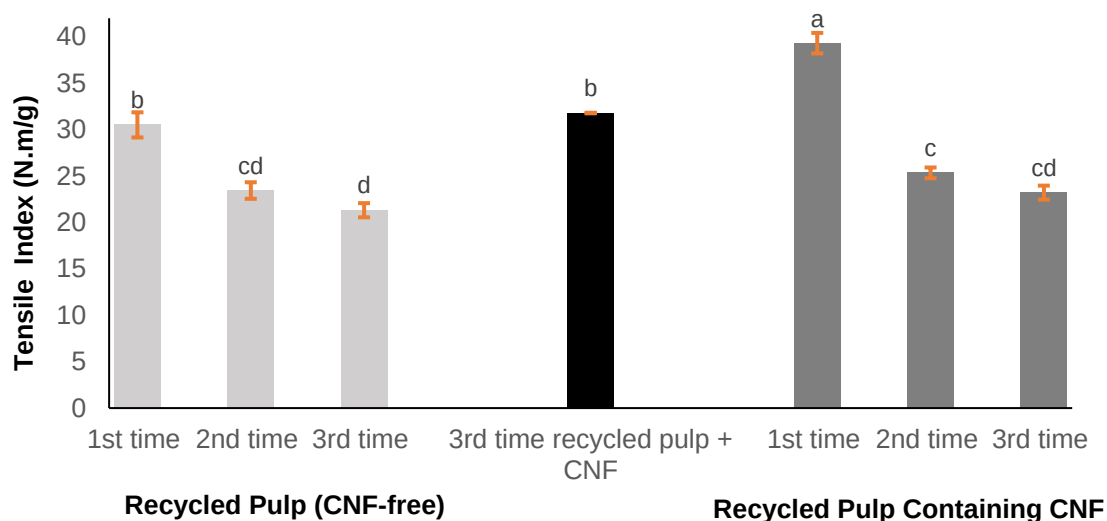


Fig. 8. The effect of applied treatments on tensile index of produced papers. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

Although subsequent recycling of this paper (Steps 5 and 6), experienced the reduction of tensile index similar to CSF-free papers, the papers reinforced with CNF in their history, numerically experienced less reduction in the tensile index after several recyclings.

Also, in step 7, addition of 5% CNF to pulp that experienced 3 times recycling, improved tensile index significantly (about 10 Nm/g increase in tensile index). As a result, it can be concluded that the direct addition of CNF to recycled pulps also, can be an effective strategy in controlling the strength decrement of recycled paper.

Tear Index

Tear strength is one of important mechanical properties. Its value can be affected by fibers length, inherent strength of fibers, and bonding status in the paper structure (Ek *et al.* 2009).

Figure 9 shows the effect of recycling process on the tear strength of CNF-free papers and papers containing CNF. According to the results, the tear index was reduced significantly in both paper sheets (with and without CNF) by experiencing the recycling process. The most important reason for mentioned results can be attributed to the shortening of the fibers length during several recycling process. Also, as stated for tensile strength, other phenomena (such as fibers hornification, blocking some of cellulose hydroxyl groups, reduction of fibers flexibility, and decreasing of fibers bonding ability) may affect the tear strength negatively (Kermanian *et al.* 2013). Furthermore, in step 4, addition of 5% CNF to 1st time recycled pulp, increased the tear index significantly (about 28.4% gain). It seems that CNF with its unique characteristics (high hydroxyl groups content, high specific surface area, and nano-fibrous structure) improved the bonding in the end paper structure, which in turn can compensate the shortening of recycled fibers (Yousefhashemi *et al.* 2019). Furthermore, the presence of CNF in paper structure had significant effect on the preservation of tear strength in several recycling times, which is an important achievement in current research.

Also, applying 5% CNF in 3rd time recycled fiber in step 3 increased the tear index of end papers perfectly (increment from 2.61 to 4.26 mNm²/g).

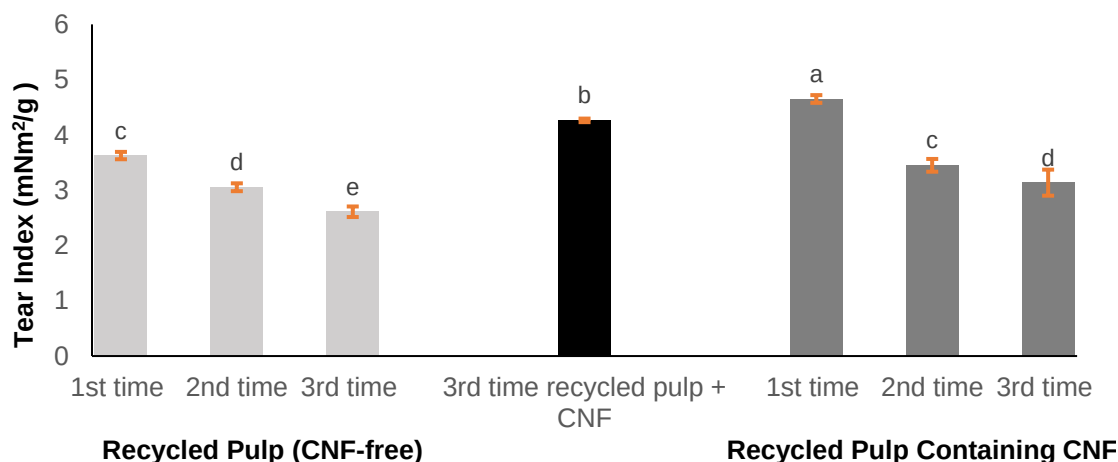


Fig. 9. Effect of applied treatments on tear index of papers. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

Opacity of Papers

Opacity, as one of the important optical properties of paper. Its value can be affected by various factors such as filler, fiber fine percentage, sheet bonding status, *etc.*

As can be seen in Fig. 10, in all treatments there was no significant difference in opacity. The results were in contrary to expectations. It was expected that the recycling process might increase the opacity because of the hornification phenomenon (reduction of fibers flexibility, decreasing fibers bonding, increase in structure porosity ...). In contrast, as an expectation, the presence of CNF in paper structure would increase the bonding status in paper structure and might decrease the opacity. Although other physical properties such as thickness and density showed significant changes in different treatments, no obvious change was found in opacity.

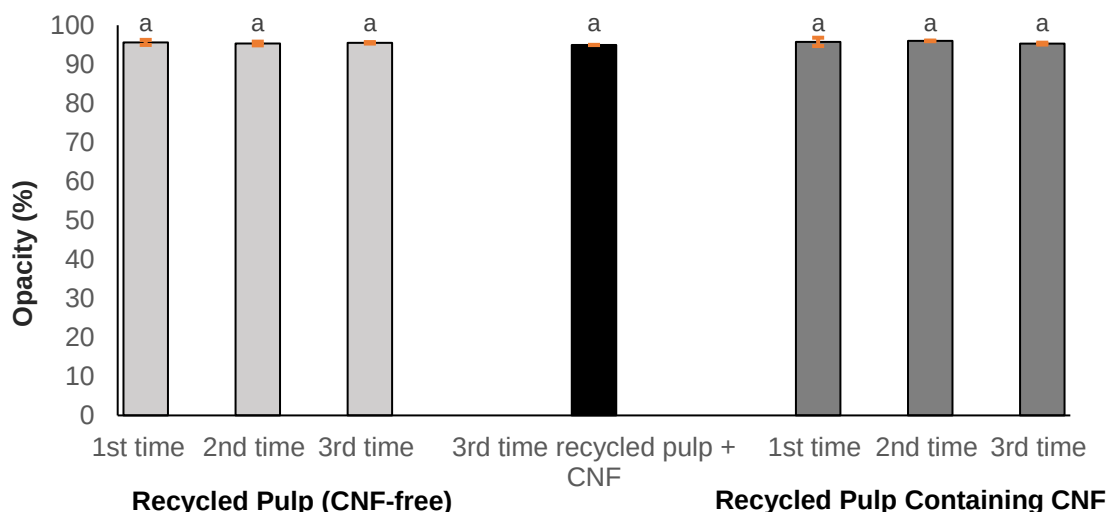


Fig. 10. Effect of treatments on the opacity of produced papers. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

Water Absorption Capacity (Cobb 60)

Figure 11 shows the water absorption of papers (Cobb 60) made from different treatments. The Cobb test describes the amount of water absorbed per unit area of paper. at a specified time under standard conditions. Various factors, especially porosity and sizing treatments, can affect water absorption of papers (Zawawi *et al.* 2013).

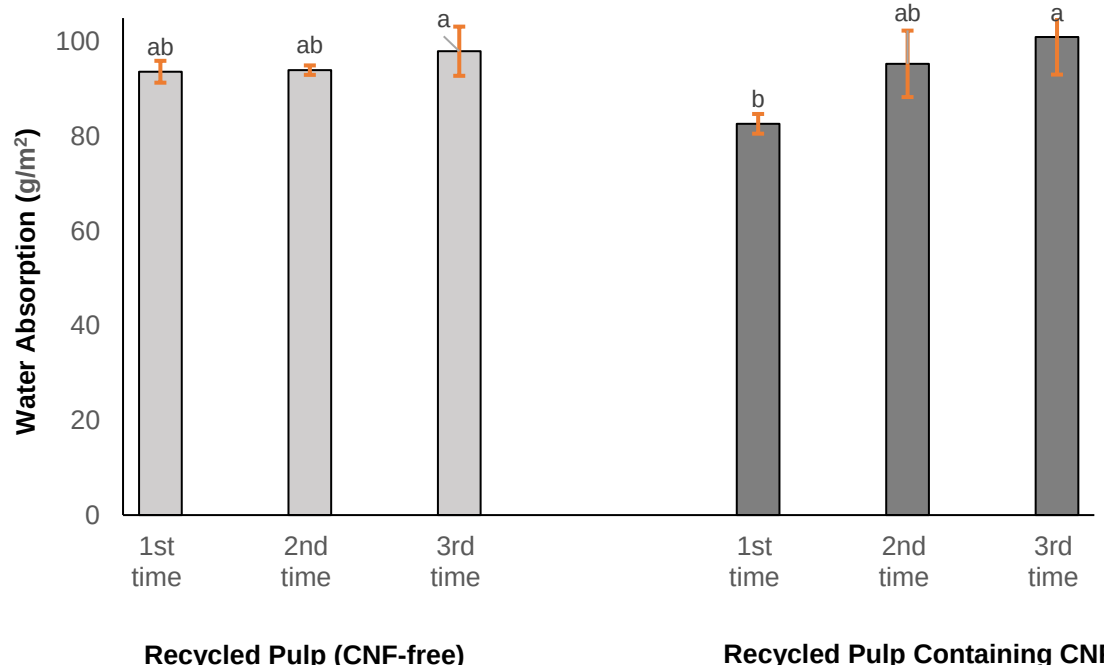


Fig. 11. The Effect of applied treatments on water absorption. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

The results obtained suggest that, with increasing recycling times in both CNF-free pulp and pulp containing CNF, the Cobb test value increased. However, this increment was not statistically significant in CNF-free papers. In general, increasing the porosity and the presence of more pores in the paper structure can increase the water absorption in the paper product. It seems that increasing in recycling times can increase the porosity of the papers due to the occurrence of the fibers' hornification and reduction of bonding (Kermanian *et al.* 2013). This can lead to increasing the water absorption. The results of thickness and density (Figs. 6 and 7) agree with the results of the Cobb test.

CONCLUSIONS

1. This study analysed the recyclability of wastepaper with or without cellulose nanofibers (CNF) as an additive in their history of papermaking.
2. The CNF was successfully produced from the white cutting wastepaper (WCW) fibers, applying mild acidic pretreatment followed by mechanical treatment with super grinding disk machine.
3. Applying the produced CNF in WCW pulp showed successful improvement of the strength properties as well as the fines retention. But in contrast, the pulp drainage rate was drastically decreased.
4. Applying CNF didn't have significant effect on opacity and Cobb as physical properties of produced products.
5. Increasing the recycling times in both pulps (with and without CNF) caused reduction in density, mechanical properties, and fines retention. It should be noted that the drainage in pulps containing CNF increased after experiencing recycling times.

6. As the most important achievement, the papers reinforced with CNF in their history experienced less reduction in mechanical strength and retention.

It should be noted that the wide range of variables such as kind of recycled fibers and the history of processes applied on fibers (mechanical and chemical process) can affect the results which should be considered widely in future investigations.

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