Improved Drainage of LNFC-reinforced Recycled Pulp and Mechanical Properties of End Papers by the Zeolite-Chitosan Microparticle Drainage Aid System

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Good drainage of the pulp suspension is vitally important for stable papermaking. Although the addition of lignocellulosic nanofibers (LNFC) in pulp could highly reinforce the end paper sheets, the application of LNFC could diminished the pulp drainage. To solve this problem, the impact of the zeolite-chitosan and bentonite-chitosan microparticle drainage aid systems on the LNFC-reinforced recycled pulp was systematically investigated. Results indicated that the mentioned microparticle systems improved the drainage and retention especially in applying 1% chitosan with 0.3% zeolite. In mechanical properties, applying the microparticle systems, not only did not deteriorate these properties, but also improved most of them, especially in treatment containing zeolite-chitosan. It seems that the improved pulp drainage and the mechanical properties of the end papers was due to successful mission of microparticle system and synergistic interactions of pulp fibres, LNFC, chitosan, and zeolite, which could also lead to denser and more uniform structure of handsheets.

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Keywords: Chitosan; Drainage; Lignocellulosic nanofiber; Recycling; Zeolite

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INTRODUCTION

The increasing demand for cellulosic fibers, along with increasing environmental concerns, has led to the development of products using recycled fibers. Approximately 60% of paper products are now made of recycled fibers (Rahmaninia *et al*. 2008; Khosravani *et al*. 2016; Saxena and Singh 2017). Although recycled fibers can experience negative effects such as hornification and shortening (Delgado-Aguilar *et al*. 2015; Rahmaninia and Khosravani 2015; Campano *et al*. 2018), their portion in papermaking has continued its positive trend (FAO 2021).

Applying different additives is a strategy for improving the process and product properties of recycled paper (Garland *et al*. 2022). Lignocellulosic nanofibers, which also can be called lignin-containing nanofibrillated cellulose (LNFC), have been introduced as novel bio additives (He *et al*. 2018; Wu *et al*. 2021) to improve the mechanical properties of recycled paper (González *et al*. 2013; Osong *et al*. 2016; Yousefhashemi *et al*. 2019). NFC/LNFC, as a natural polymer made from variety of cellulosic/lignocellulosic materials, is a fibrous product with nano-size diameter and micrometer length including both crystalline and amorphous regions. The unique characteristics of NFC/LNFC, such as fine diameter, high aspect ratio, considerable specific surface area, high potential of hydrogen bonding along with its biocompatibility, biodegradability, and renewability (Du 2020), presents it as a high potential product for various applications, especially papermaking (Yousefhashemi *et al*. 2019). It seems that LNFC/NFC can improve the fiber bonding in paper sheets due to its high specific area and the possibility of creating more hydrogen bonds among components in the paper sheet (Ridgway and Gane 2012; Campano *et al.* 2018; Tajik *et al.* 2018). However, adding LNFC to paper pulp reduces the rate of drainage (Rantanen and Maloney 2013; Hollertz *et al*. 2017), which could influence the process stability of papermaking and increase process cost of drying for the paper sheets with more water retention. The reason seems to be the high specific level of nano-cellulose and the possibility of increasing the hydrogen bonding with water, as well as the rapid closure of pores in the fiber sheet formed on the mesh, thus preventing faster and easier removal of water from the sheet (Yousefhashemi *et al.* 2019). Some chemical additives can improve the drainage of wet web fiber (Hubbe 2014; Amiri *et al*. 2019; Leib *et al*. 2022; Barrios *et al*. 2023). These additives are used as single, dual, and multiple systems or as micro- or nanoparticles. Microparticle systems, as one of well-known drainage and retention aid strategy in wet end chemistry, consist of cationic polyelectrolyte and anionic microparticles (Khantayanuwong *et al*. 2017; Rahmaninia *et al*. 2018).

Chitosan, as one of well-known cationic polyelectrolytes, has been considered in different applications. This component is a natural polymer made of chitin (the second most abundant polysaccharide in nature) (Zhang *et al*. 2016). This biopolymer is a linear carbohydrate with high molecular weight that can be dissolved in acid and consists of (1–4) linked 2-amino-2-deoxy-b-D-glucan units (Nicu *et al*. 2011). Chitin and chitosan, as well as their derivatives, are widely used in the production of high value-added products, including cosmetics, food supplements, pharmaceuticals, and semi-permeable membranes (Gavhane *et al*. 2013).

Chitosan has been considered in the paper industry (Rahmaninia *et al*. 2018; Rohi *et al*. 2023). The cationic groups associated with the cationic biopolymers can attract the anionic groups available in the pulp suspension and increase the bonding among fibers and fines. This can involve the mechanisms of bridging, charged patches, and neutralization (Hubbe *et al*. 2009; Rahmaninia and Khosravani 2015). Studies have been conducted on the application of chitosan as a papermaking additive for different purposes such as improving the wet and dry strength of paper, fine-particle retention, the rate of drainage aid, the stabilization of color stabilizer in the production of colored paper, and antimicrobial properties, *etc.* (Sabazoodkhiz *et al*. 2017; Song *et al*. 2018; Rohi *et al*. 2023).

Among the natural anionic microparticles, the aluminosilicate minerals, such as zeolite and bentonite, have found different applications in papermaking. Zeolites as hydrated aluminosilicates, are minerals with octagonal structure. In fact, silicon and aluminium atoms with contribution of oxygen atoms form a tetra-hydrate structure in which aluminium or silicon is at the center and there are four oxygen around it (Montalvo *et al*. 2012). Zeolites are one of the largest categories of known minerals, with more than 40 different natural and nearly 100 synthetic forms (Ko *et al*. 2010). Like zeolite, bentonite or montmorillonite is another well-known aluminosilicate minerals with negative charge in water. Bentonite or montmorillonite is a three-layer mineral consisting of two tetrahedral sheets sandwiched around a central octahedral sheet with a very high surface area (Cui and Chen 2023). Bentonite has different applications in pulp and paper industries such as stickies control (in paper recycling), enhancement of optical properties and printability, as well as improvement of product and process of papermaking (for example as a retention and drainage aid, (Diab *et al*. 2015; Merayo *et al*. 2017). However, using zeolite in different applications of pulp and paper industries is in its early stages. For instance, zeolite has been used in combination with cationic starch or cationic polyacrylamide as a retention aid in pulp (Ko *et al*. 2010) or it was used as a filler in papermaking (Engin and Atik 2018). Therefore, considering the considerable aluminosilicate mineral resources in the world, finding more applications and using these environmental and low-cost additives in pulp and paper industry can be a promising idea.

The purpose of the current study is to assess the performance of zeolite as a low cost anionic microparticle along with chitosan biopolymer for improving the process properties of papermaking especially drainage which has been negatively affected by addition of LNFC in recycled pulp.

EXPERIMENTAL

Materials

Mixed old corrugated containers (OCC) were collected locally, repulped, and named OCC pulp. Medium molecular weight chitosan (400 to 600 kDa) was purchased from Sigma-Aldrich, Germany. Some general specifications of chitosan are in Table 1.

Table 1. Specifications of Chitosan

Degree of Deacetylation (%)	Viscosity at 24–25 °C (cP)	Molecular Weight (kDa)	Powder color
93	200-800	400-600	Yellow

Zeolite in powder form with 400 mesh size was a gift from Afrazand Company, Iran. Bentonite in powder form with 400 mesh size was a gift from Farzan Powder Mashhad Company, Iran.

LNFC were manufactured from OCC recycled fibers by using the mechanical process with super disk mill (MKCA6-2; Masuko Co., Japan) in Nano-Novin Polymer Company, Iran. The OCC pulp was washed and screened with 200 mesh hole size to remove contaminants. The obtained pulp with no additional treatment was passed 3 times from super disk mill. The properties of the produced LNFC are in Table 2.

Table 2. The Properties of LNFC

Organic material content (%)		Ash content (%)	Consistency (%)	Suspension Color
94.97		5.03	1.58	Brown

OCC waste paper was repulped and refined with laboratory Hollander beater up to 287 mm CSF according to TAPPI T 200 sp-01 (2007). Reverse osmosis (RO) water was used for the process.

To prepare the bentonite/zeolite suspensions, 0.1 g bentonite/zeolite microparticles were suspended in 100 mL distilled water, and the solution was stirred for 2 h at room temperature (25 °C). Chitosan was dissolved in 1% acetic acid and stirred for 2 h at room temperature.

LNFC was diluted to 0.5% consistency with distilled water and mixed for 5 min at 1000 rpm by a mechanical agitator.

Field Emission Scanning Electron Microscopy (FE-SEM)

The FE-SEM technique (TESCAN MIRA3, Czechia Republic) was used to characterize the microparticles and handsheets.

Atomic Force Microscopy (AFM)

The AFM technique (CP II Veeco, USA) was used to study the size and size distribution of the zeolite/bentonite.

Transmission Electron Microscope (TEM)

Transmission electron microscope images (EM 208S Philips, Netherlands) were used to visualize the structure of LNFC. One drop of the diluted LNFC was put on formvar carbon coated Cu grid, mesh 300 and dried slowly at room temperature. The images were taken and analysed using related software.

Zeolite, Bentonite, and Chitosan Charge Detection with Zeta Meter

The zeta meter (Malvern, UK), was used to check the zeta potential of applied microparticles dispersed in distilled water and also chitosan dissolved in 1% acetic acid.

Applying the Treatment in Pulp Suspension

The recycled OCC pulp was poured into a disintegrator jar. Then, 3% LNFC (based on dried weight of pulp) was added to the pulp and the mixture was diluted with RO water up to 0.5% consistency. The disintegrator speed was set to 25000 rpm for 15 min. The control treatment was a pulp sample without any additives. The treated pulp with LNFC was poured in a beaker. Then, 1% chitosan solution (based oven dried pulp weight) was added to the mentioned pulp and mixed with mechanical agitator in 1000 rpm for 1 min. In the next step, the microparticle was added to the mixture with 800 rpm for 45 s.

Table 3. Research Treatments

Treatment		Additives Material Content (*)			
		Zeolite	Bentonite	LNFC	Chitosan
Control		0	0	0	0
LNFC 3%		0	0	3	0
Chitosan 1%+LNFC 3%		0	0	3	1
Bentonite Content +Chitosan 1% +LNFC 3%	0.1	0	0.1	3	1
	0.2	0	0.2	3	1
	0.3	0	0.3	3	1
Zeolite Content +Chitosan 1% +LNFC 3%	0.1	0.1	0	3	1
	0.2	0.2	0	3	1
	0.3	0.3	0	3	1

*based on oven-dry weight of pulp

Process Properties Determination

Fines content and fines retention were measured with Dynamic Drainage Jar (DDJ) according to TAPPI T 261 cm-00 (2007). Pulp freeness and drainage were determined according to TAPPI T 227 om-99 (2007) using a Canadian Standard Freeness (CSF) Tester.

Measurement of Mechanical Properties of Handsheets

Handsheets with a basis weight of 120 g/m^2 were made in a handsheet maker according to TAPPI T 205 om-02 (2007). After pressing and drying process in a drum dryer, the handsheet samples were prepared for mechanical tests. The tensile (and also tensile energy absorption), tear, and bending resistance indices were measured according to TAPPI T 494 om-01 (2007), TAPPI T 414 om-04 (2007), and SCAN-P 29:95 (1995), respectively. To calculate the apparent density of handsheet samples, their basis weight was divided by their thickness and reported with g /cm^3 unit.

Statistical Analysis

Each treatment was tested with at least three replications. All results were analysed using a completely randomized design (CRD), and the standard deviations were calculated. Duncan's multiple range test (DMRT) was applied to averages grouping. The Duncan results were categorized alphabetically. The data groups that did not share the same letter were significantly different from each other (99% confidence level). The statistical analysis was conducted using SAS software (IBM, version 16.0, Armonk, NY, USA).

RESULTS AND DISCUSSION

Measurement of Fines Content in OCC pulp

The fines content (particles pass through 200 mesh screen) of recycled OCC pulp after repulping was approximately 17%. The fines content in the paper pulp after 15 minutes intense disintegrating process with 25,000 revolutions of the disintegrator was about 30%, which is probably due to the dispersing of cellulosic fines and other particles that had been attached to fiber surfaces.

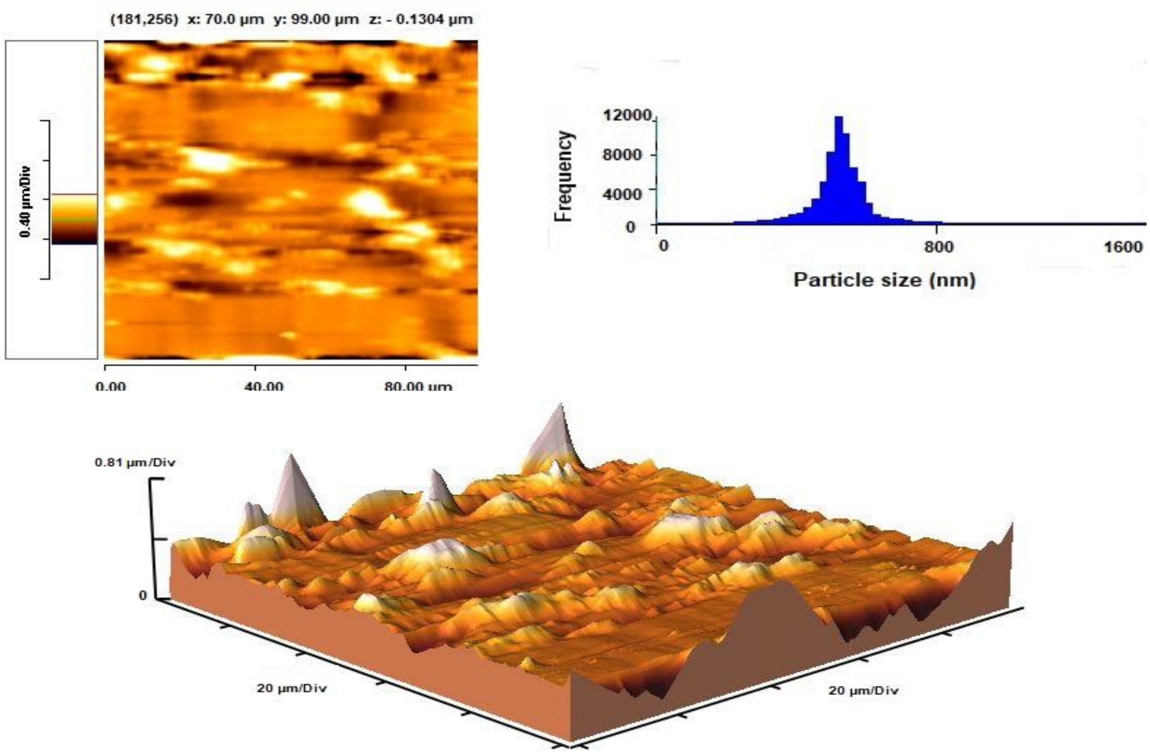

Fig. 1. Topography and size distribution of zeolite

Characterization of Zeolite Microparticles

The topography and size distribution of zeolite microparticles were estimated by AFM (Fig. 1). The range of particles size was 0.4 to 0.8 μm. There was a normal distribution of microparticles with maximum dimension of approximately 0.6 μm. Considering the FESEM images (Fig. 2), the shape and porous structure of these microparticles was in accord with other reports (Jha and Singh 2016).

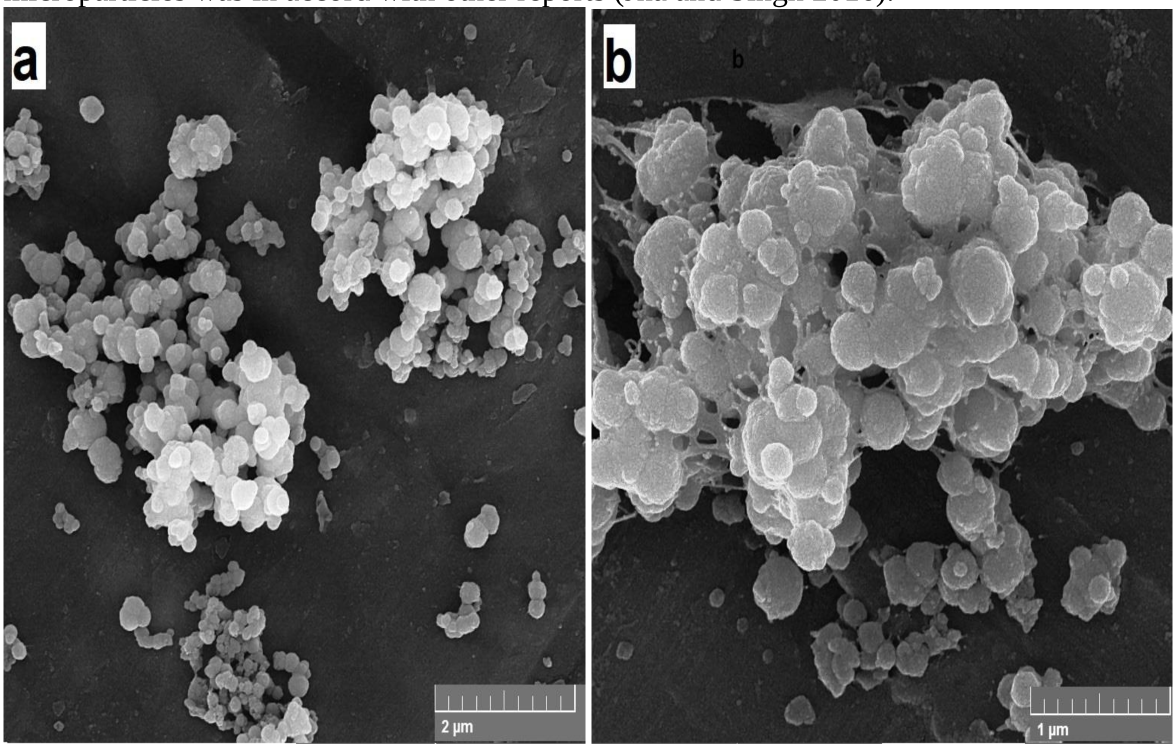

Fig. 2. FESEM images of zeolite microparticles

Characterization of Bentonite Microparticles

In Fig. 3, the topography and size of bentonite microparticles using AFM technique was considered. Accordingly, the range of bentonite particles size was 0.3 to 1 μm. Furthermore, normal distribution of bentonite microparticles with maximum dimension approximately 0.6 μm was determined. The FESEM images (Fig. 4) show the plate-like structure of bentonite microparticles.

Determination of Zeolite, Bentonite and Chitosan Zeta Potential

The results of the zeta potential test using the DLS method showed that the zeolite microparticle suspension (with 10.0 ms / cm conductivity and 0.897 mpa.s viscosity) had a negative charge of -26.8 mV. Also, the zeta potential of bentonite microparticle suspension (with 0.95 mS/cm conductivity and 0.897 mPa·s viscosity) was 32.0 mV. Moreover, the zeta potential for dissolved chitosan in 1% acetic acid (with 1.22 mS/cm conductivity and 2/3 mPa·s viscosity) showed that the charge was positive with 1220 mV.

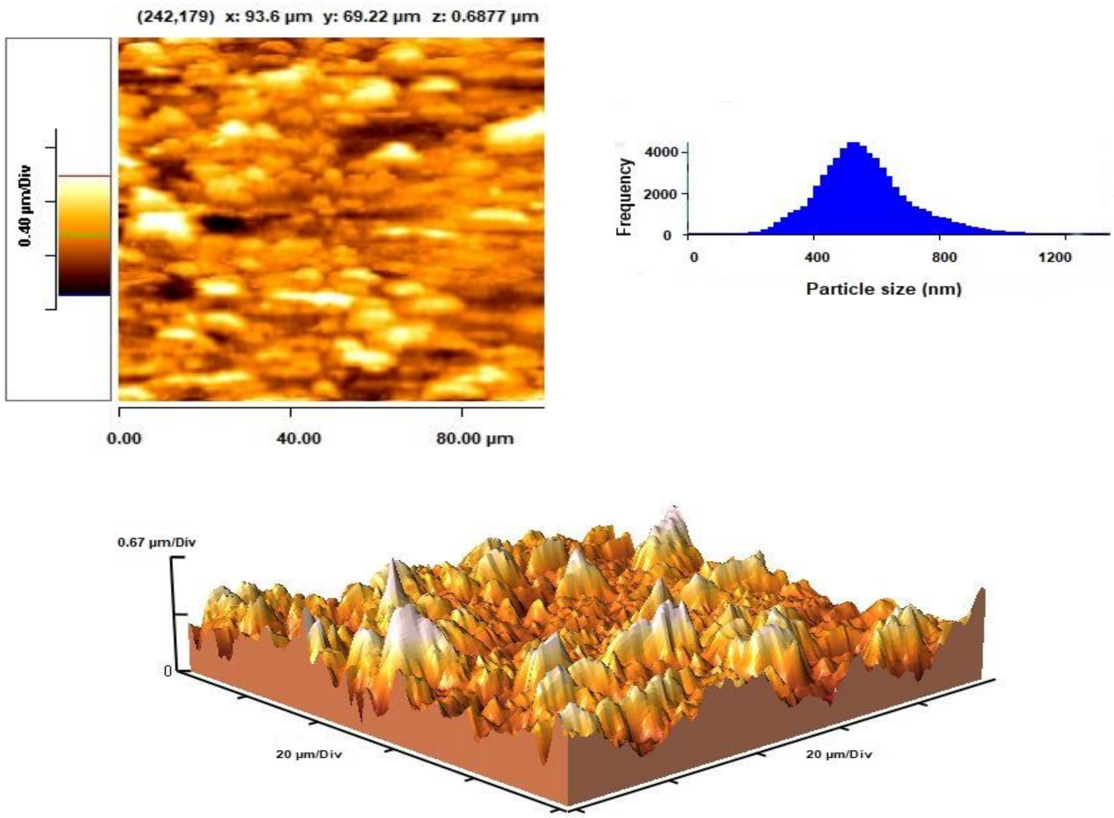

Fig. 3. Topography and dimensions of bentonite microparticle

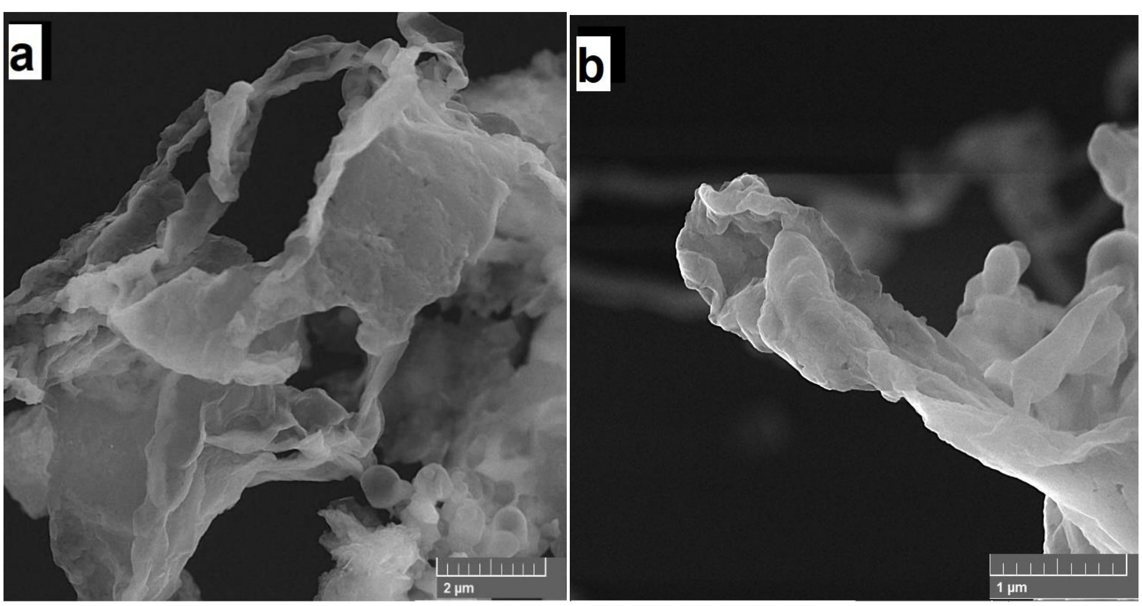

Fig. 4. FESEM images of bentonite microparticles (overlapped bentonite platelets)

Characterization of LNFC by TEM

TEM images of LNFC made from OCC recycled fibers are shown in Fig. 5. Nano-fibers are defined as fibers with nano size diameter and usually micrometer length. The nano sizes of the fibers are completely obvious, which indicated that the production process of LNFC from recycled fibers was successful.

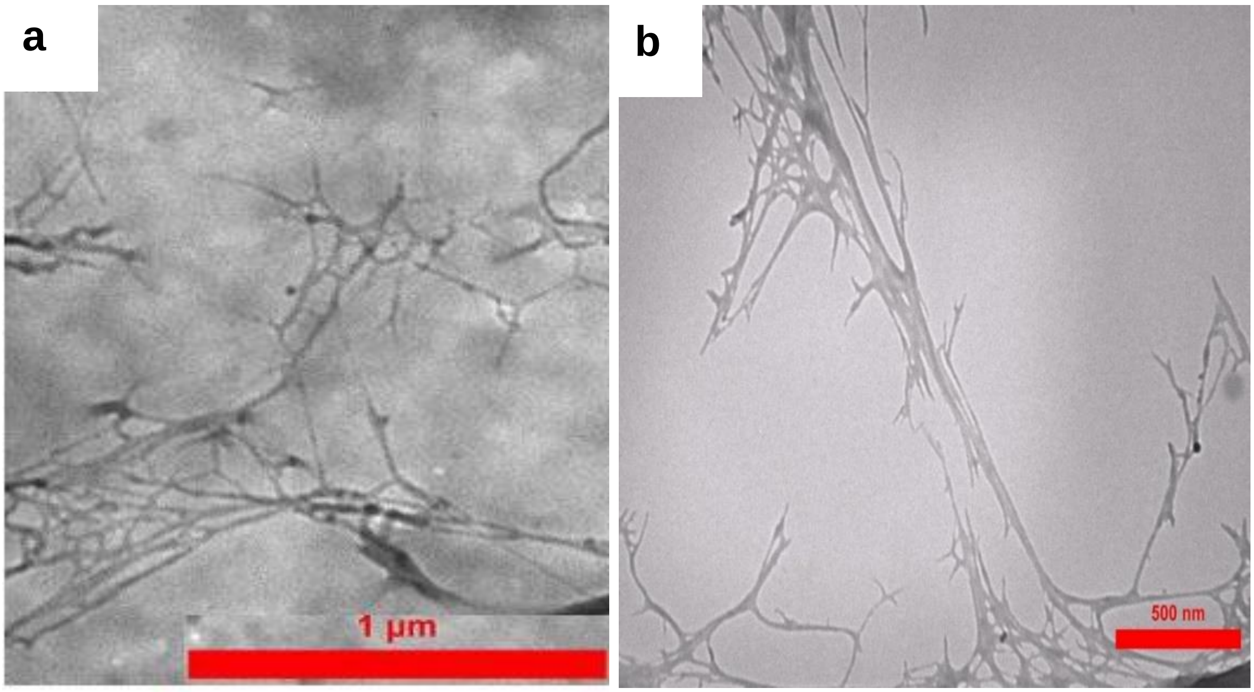

Fig. 5. TEM images of LNFC. a: 1 micrometer, b: 500 nm

Drainage of Recycled Pulp

The drainage of pulp is one of the most important process properties in the paper industry. The more water separated from the wet sheet (before pressing and drying steps), the lower the cost of the product by consuming less energy in the dryers.

Figure 6 shows the results of pulp drainage after treatments. According to the statistical results, there was a significant difference among the mentioned treatments at the 99% confidence level. The addition of LNFC to the control treatment reduced the drainage from 178 mL to 132 mL (about 35% reduction). This decreasing trend has been confirmed in various articles (González *et al*. 2013; Rantanen and Maloney 2013; Osong *et al*. 2016; Hollertz *et al*. 2017; Yousefhashemi *et al*. 2019). The reason seems to be the high specific level of nano-cellulose and the possibility of increasing the hydrogen bonding with water, as well as the rapid closure of pores in the fiber sheet formed on the mesh, blocking the drainage channels, thus preventing faster and easier removal of water from the sheet (Yousefhashemi *et al*. 2019).

Adding 1% of chitosan to the pulp containing LNFC increased the rate of drainage by about 43%. Probably, as a linear biopolymer and polyelectrolyte, chitosan was able to flocculate cellulosic suspension components (lignocellulosic fibers, fine, nano-fibers) with bridging mechanism and electrostatic interactions (Rahmaninia *et al*. 2018). The use of chitosan (at constant level of 1%) along with 0.1, 0.2, and 0.3% dosage of zeolite microparticle to paper pulp containing LNFC, considerably increased the drainage about 42%, 52%, and 66%, respectively. By adding chitosan (at constant level of 1%) along with different levels of bentonite (0.1, 0.2 and 0.3%) to the pulp containing LNFC, the drainage increased properly (about 52%, 35% and 43%, respectively). The high performance of the bentonite at the lowest addition level is consistent with its very high specific surface area. Similar results were reported previously (Rahmaninia *et al*. 2018).

The favourable results of both microparticle systems in improving drainage, especially at the level of 0.3% zeolite, showed the success of the designed systems to improve pulp drainage. In fact, the designed microparticle systems have been able to improve drainage by formation of suitable and fine flocs in suspension. The possible mechanism of this phenomenon is shown in Fig. 7.

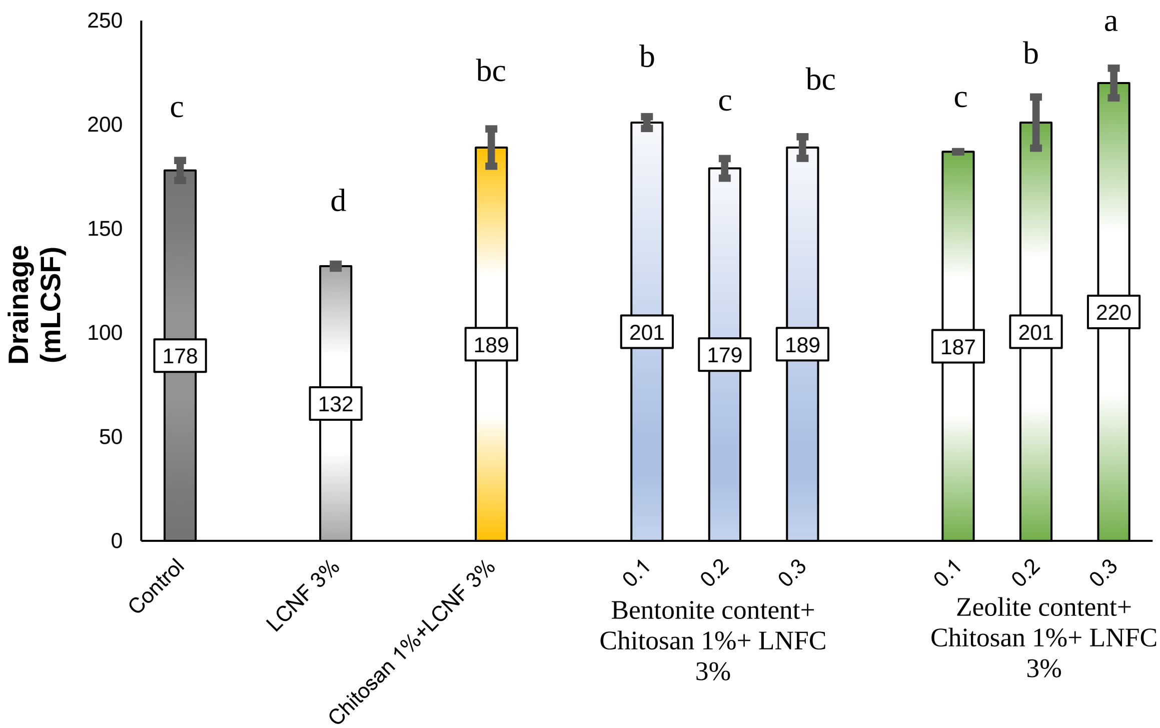

Fig. 6. Effect of treatments on drainage of recycled pulp suspension. Plotted points that do not share the same letter were found to be significantly different from each other (99% confidence level)

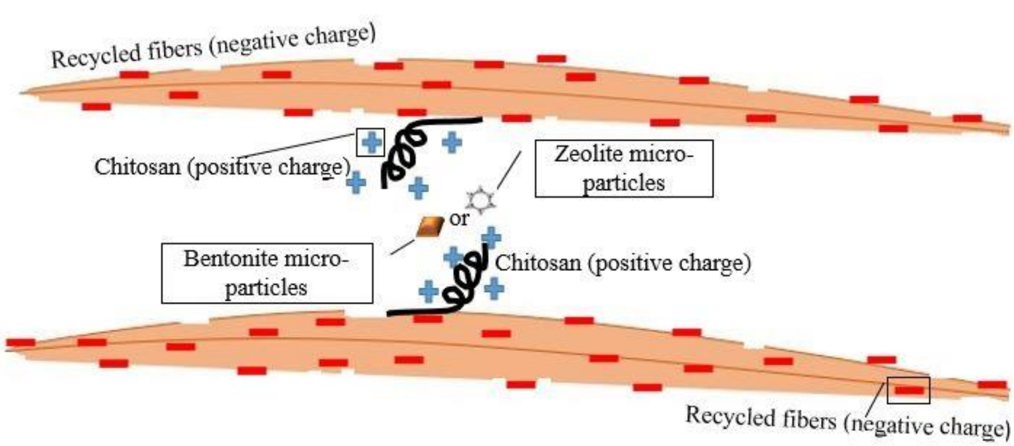

Fig. 7. Probable mechanism of floc formation with microparticle system

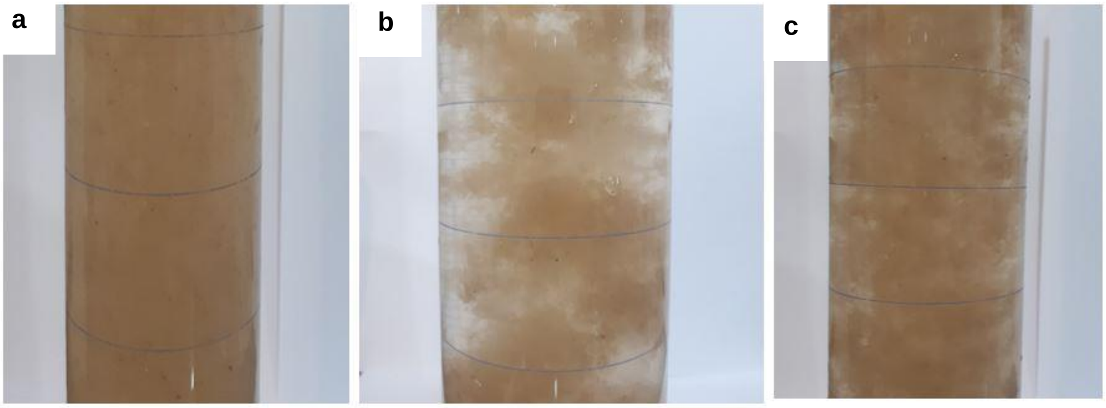

Fig. 8. Visualizing of flocculation phenomenon in: a: pulp containing LNFC, b: pulp containing LNFC only treated with 1% chitosan and c: pulp containing LNFC treated with 1% chitosan + 0.3% microparticle (zeolite).

Figure 8 shows the flocculation phenomenon after introducing the chitosan itself into the pulp with LNFC (Fig. 8b) and also addition of zeolite-chitosan microparticle system in pulp suspension containing LNFC (Fig. 8c). No flocculation happened in the pulp containing LNFC (Fig. 8a). Large non-uniform flocs were formed in sample treated with 1% chitosan (Fig. 8b), as well as uniform and smaller flocculation in sample containing microparticle system.

Fines Retention in Recycled Pulp

One of the most important issues in papermaking is the retention of fine particles in the pulp, which can affect the properties of the final products along with quality of water recirculated in a paper mill. Fines retention, especially in products with higher grammage, happens with physical entanglement of fines in fibrous structure of paper web during sheet formation. Additives can help to retain more fines in sheet structure with more flocculation of fines and fibers in the pulp suspension (Hubbe 2014). The retention of fines in the system has a close relation with drainage. Higher drainage in forming section can remove more fine particles from the forming fabric. However, during the forming process, including the deposition of the first layer of fibers on the fabric, and also plugging the wire holes, the drainage would decrease.

Figure 9 contains the results of fines retention after applying different treatments. By adding LNFC to the pulp, the fines retention experienced a 14% increase (changing retention from 41% to 48%), confirming the results of previous studies (Hollertz *et al*. 2017; Yousefhashemi *et al*. 2019). This increase can be related to drainage decrement of pulp suspension because of plugging the wires holes and also blockage of fiber web pores.

Adding 1% chitosan to paper pulp containing LNFC did not significantly change the fines retention. This interesting result can be because of chitosan’s dual function. Considering the result of drainage in previous section, although chitosan increased the drainage successfully (which could help to release more fines with filtrate), this linear biopolymer with high positive charge has the ability to make bridges between different components of recycled pulp suspension, especially LNFC. Hence, less fine particles would be removed by this higher flocculation.

With the addition of microparticles, the retention of fine particles increased compared to the treatment containing only LNFC. This retention gain is indicative of the successful performance of the system. The micro-particle system usually has the ability to create small and stable flocs that help to retain more fine particles. The best retention results achieved in the treatment containing 1% chitosan with 0.3% zeolite. It appears that the electrostatic interaction between chitosan polyelectrolyte and zeolite micro particle played the main role (Sabazoodkhiz *et al*. 2017).

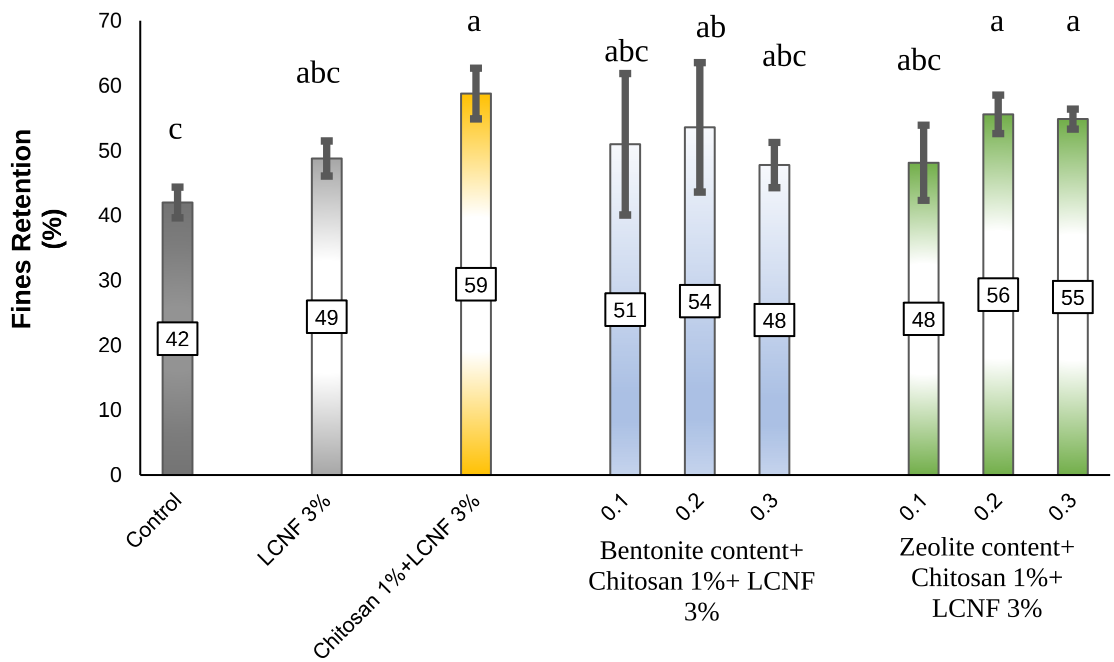

Fig. 9. The effect of treatments on fines retention of recycled pulp suspension. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

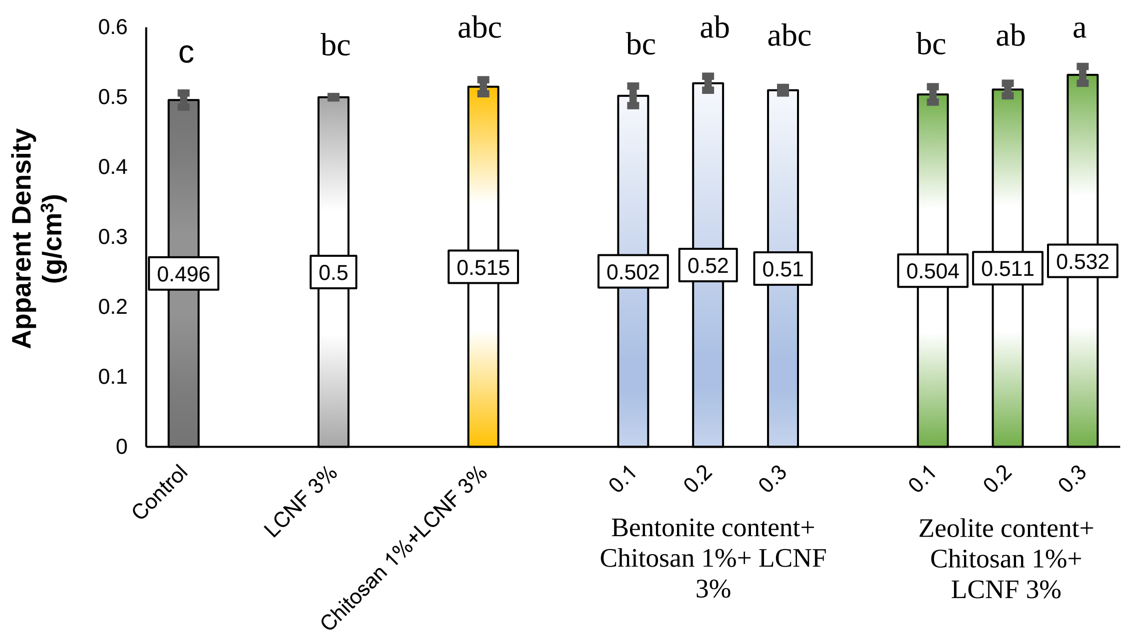

Fig. 10. The effect of treatments on apparent density of recycled paper. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

Apparent Density of Paper Sheet

Figure 10 shows the changes in the apparent density of papers produced by applying different treatments. There was not a significant difference in apparent density among the papers made by applying additives in the pulp, but all the treated paper showed significant increment compared to control sample (with no additive). The highest density was achieved in samples prepared by addition of 1% chitosan and 0.3% zeolite which showed the proper performance of treatment containing micro particle system. The comparison of FE-SEM images related to control papers and papers prepared with consumption of 1% chitosan and 0.3% zeolite (Fig. 11) confirmed this density result. Applying the micro particle system could make a proper fine floc in the pulp suspension which led to denser handsheets with less thickness, as shown by the thickness results in Table 4.

Table 4. The Effect of Treatments on Thickness of Recycled Paper

Treatment		Thickness Average (mm)	Standard Deviation	Duncan Grouping*	Grammage Average (g/m^2)
Control		0.250	0.011	Ab	130.03
LNFC 3%		0.237	0.006	Bc	115.4
Chitosan 1%+LNFC 3%		0.255	0.011	A	129.81
Bentonite Content +Chitosan 1% +LNFC 3%	0.1	0.244	0.013	Ab	127.06
	0.2	0.249	0.013	Ab	127.36
	0.3	0.249	0.006	Ab	128.05
Zeolite Content +Chitosan 1% +LNFC 3%	0.1	0.258	0.006	A	130.36
	0.2	0.234	0.008	Bc	115.85
	0.3	0.225	0.013	C	116.13

* The treatments that do not share the same letters were found to be significantly different from each other (99% confidence level).

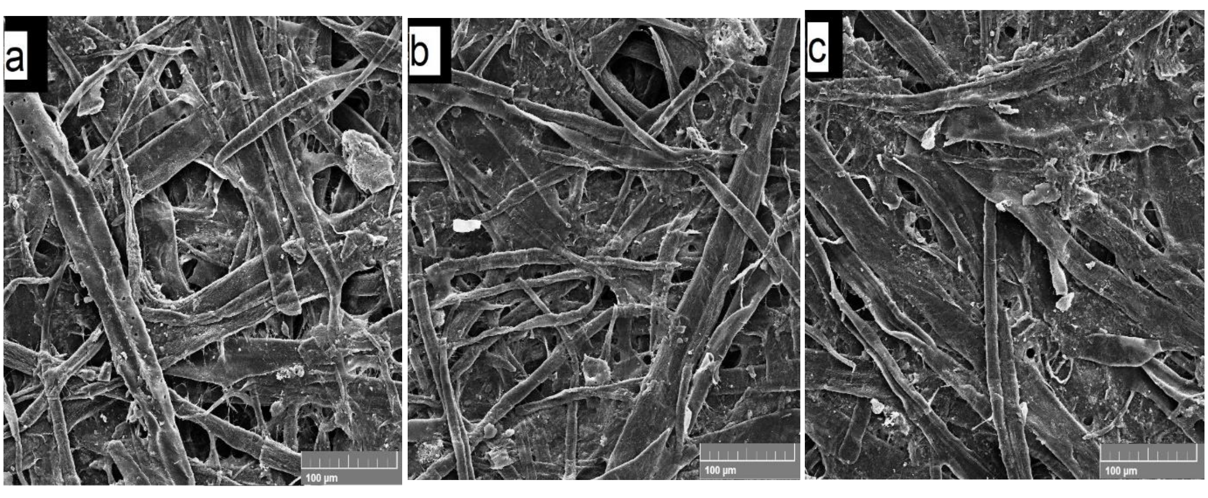

Fig. 11. FE-SEM images of made papers with: a) no additive (control), b) only 3% LNFC, c) 3% LNFC+1% chitosan +0.3% zeolite.

FE-SEM Images of Handsheets

Figures 11 shows FE-SEM images of handsheets made of control pulp, pulp containing only 3% LNFC and pulp with 3% LNFC + 1% chitosan + 0.3% zeolite. The handsheets with LNFC had more uniform and denser structure compared to control sample without any additives. The presence of LNFC in the paper structure improved the fibrous components bonding along with more fines retention, leading to more uniform, denser sheets. The same results were reported in another study (Yousefhashemi *et al*. 2019). Even by applying the microparticle system and forming the flocs in the pulp suspension, the final structure of paper made of this treatment did not show considerable difference compared with the sample containing only LNFC. As mentioned previously, there may be an advantage to forming paper with a fine scale of flocs in the pulp. The drainage and retention as main process properties improved without negative effect on formation of final paper made by using these components, which can be a great achievement in this study.

Tensile Strength of Prepared Papers

Figure 12 contains the results of the tensile index in different treatments. Tensile strength is one of the important properties in papermaking, which is strongly influenced by the fibers bonding in the fiber sheets. The tensile index of control samples was 27 Nm/g, which was the lowest among all treatments. By adding LNFC (3% dry weight of pulp), the tensile index increased about 12% compared to control, which could be related to the improvement of the fiber bonding in paper sheets due to the high specific area of LNFC and the possibility of creating more hydrogen bonds among components in the paper sheet. Various studies have reported the same results (Ridgway and Gane 2012; Campano *et al*. 2018; Tajik *et al*. 2018; Yousefhashemi *et al*. 2019).

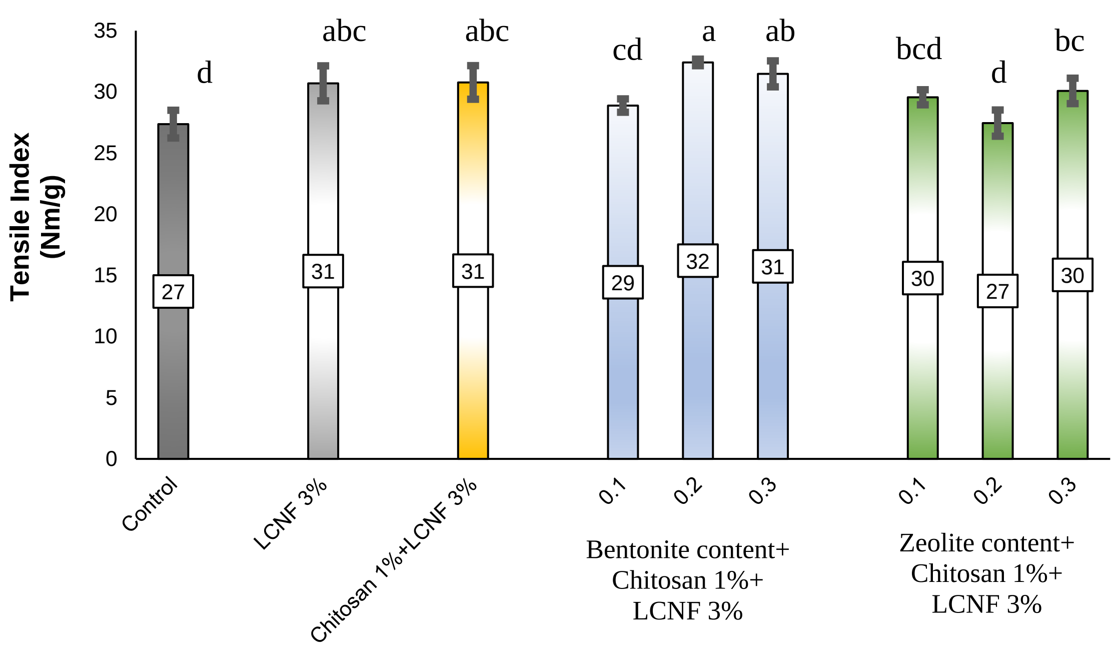

Fig. 12. The effect of treatments on tensile index of recycled paper. Plotted points that do not share the same letter were found to be significantly different from each other (99% confidence level).

By introducing chitosan (1% constant consumption) to the control pulp, the tensile index was improved. The addition of chitosan in the sample containing LNFC did not change the strength significantly. This result was contrary to some investigations that reported tensile enhancement by accompanying chitosan with nanofibers (Merayo *et al*. 2017; Campano *et al*. 2018).

Applying treatments containing the microparticle system increased the tensile strength index by about 9% compared to the control treatment. Also, the addition of microparticle system in sample containing LNFC did not change the tensile index by much. Considering this result along with the mentioned results of drainage and retention showed the successful mission of microparticle system. In fact, according to our previous knowledge, application of additives in pulp suspension can deteriorate the formation of final paper sheets. The microparticle could improve the process properties without negative impact on tensile strength which can be referred to the proper impact of this system (especially system containing zeolite particle) on formation of suitable fine flocs in pulp suspension as mentioned before.

The amount of tensile energy absorption in the paper samples is presented in Table 5. The tensile energy absorption (TEA) is the amount of work done when the paper is stretched until it breaks. Considering the results, the proper coordination of TEA and tensile index can be observed.

Table 5. The Effect of Lignocellulosic Nano-fiber and Chitosan-bentonite / zeolite System on Tensile Energy Absorption in Papers

Treatment		Tensile Energy Absorption (J/m^2)	Standard Deviation	Duncan Grouping*
Control		87611.70	24317.37	ab
LNFC 3%		78001.91	17173	ab
Chitosan 1%+LNFC 3%		84861.76	16992.61	ab
Bentonite Content +Chitosan 1% +LNFC 3%	0.1	81848.89	4822.53	a
	0.2	98290.28	6742.36	a
	0.3	102856.56	14470.68	a
Zeolite Content +Chitosan 1% +LNFC 3%	0.1	100666.41	18929.61	a
	0.2	69358.17	16477.79	b
	0.3	95357.13	15309.17	ab

* The treatments that do not share the same letters were found to be significantly different from each other (99% confidence level).

Tear Strength of the Paper

The effect of different treatments on tear index of handsheets is presented in Fig. 13. The tear strength depends on the fiber length, strength, and bonding (Ek *et al*. 2009). The tear index of control was 5.84 mN/g, which was the lowest value among others. Different mechanical forces applied on waste fibers in recycling process (especially in repulper) usually have a negative impact on the fibers length, which can reduce tear strength (Kermanian *et al*. 2013). Addition of LNFC to control pulp increased the tear index by 19%. This strength depends primarily on the length of the fibers and the strength of the fibers. These two factors were consistent in this study. The increased tear strength was a result of improved the bonding in paper sheets and the high surface area of LNFC.

Applying the chitosan-bentonite and zeolite-chitosan to pulps containing LNFC did not make any significant change in tear index. Thus, the microparticle system improved process properties (drainage and retention), prevented deterioration of final papers, and preserved the strengths of the final products.

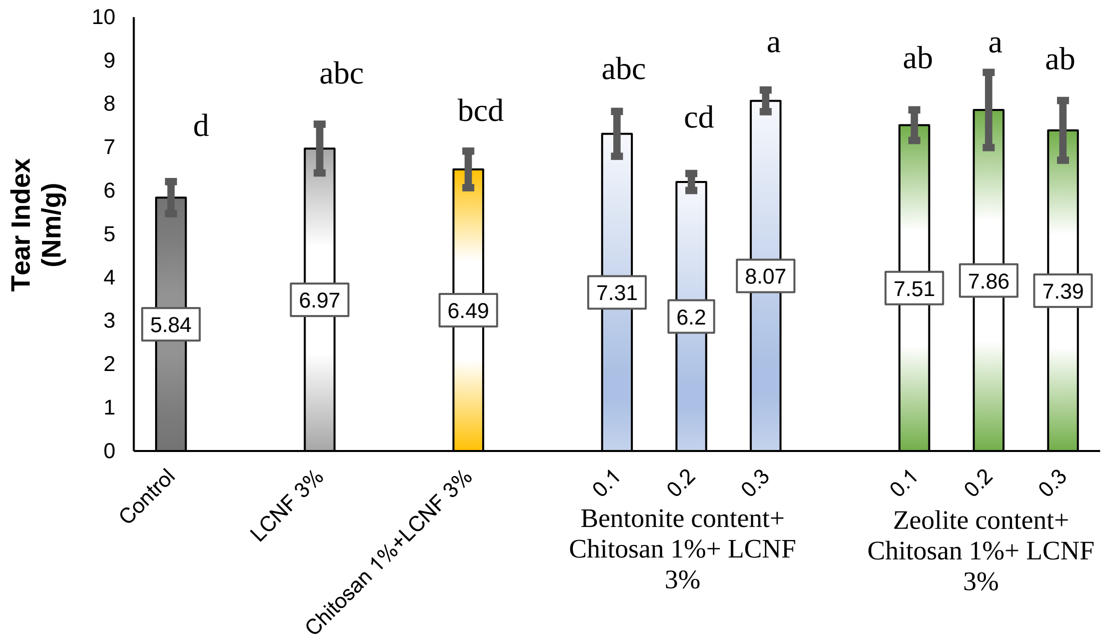

Fig. 13. The effect of treatments on tear index of recycled paper. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

Bending Resistance Index

The bending resistance index of recycled paper is shown in Fig. 14. Although most of the applied treatments showed no significant difference in bending strength, microparticle system containing 0.3% zeolite showed the highest result. This was attributed to appropriate performance of microparticle system in improving or preserving the paper sheets strengths along with enhancing the process properties (drainage and retention).

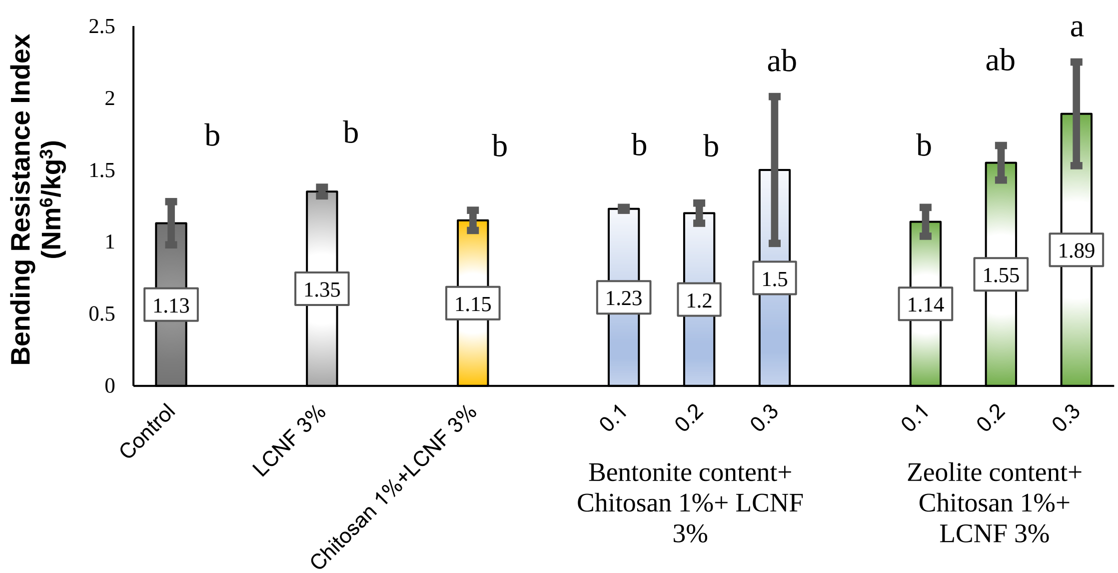

Fig. 14. The effect of treatments on bending resistance index of recycled paper. Plotted points that do not share the same letter were significantly different from each other (99% confidence level).

CONCLUSIONS

1. The addition of lignin-containing nanofibrillated cellulose (LNFC) prepared from recycled old corrugated container (OCC) pulp improved the fines retention and mechanical properties of recycled papers, but reduced the pulp drainage. These behaviors can be related to that the high specific surface area of LNFC, their ability to improve of hydrogen bonding, and also to the rapid closure of pores in the fiber sheet formed on the mesh, blocking the drainage channels.
2. Applying chitosan with bentonite/zeolite microparticles enhanced the fines retention, drainage, and mechanical strengths compared to samples with and without LNFC.
3. The best performance was achieved by applying 1% chitosan along with 0.3% zeolite. This treatment improved the drainage and fines retention and made paper sheets with higher density. These results reflect the performance of applied microparticle system (especially containing 0.3% zeolite) in producing fines and stable flocs in the pulp suspension. Visualizing the flocculation phenomenon in experiments confirmed this conclusion.

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