Effects of Different Levels of Nanocellulose and Chemical Pulp on the Optical and Mechanical Properties of Money Paper Made with Bottom Combers Pulp

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Simultaneous effects of using cellulose nanocellulose and chemical pulp in paper money were studied with bottom combers cotton pulp. The bottom combers pulp and chemical pulp were prepared in a factory that produced durable paper, and their products were transferred to the laboratory. The nanocellulose was prepared by Nano Novin Polymer Co. and was consumed at 4 levels, *i.e.*, 0%, 0.3%, 0.6%, and 0.9%. After commixing the pulp with the nanocellulose at the identified percentages, handsheets of 90 $g \cdot m^{-2}$ papers were produced. The results showed that by increasing the amount of nanocellulose, up to 0.9%, the tensile strength index, burst strength index, tear strength index, and folding endurance increased 22.7%, 38.9%, 7.7%, and 348%, respectively, when compared with the control sample. Enhancement *via* nanocellulose up to 0.9% increased the surface smoothness up to 7.9% compared with the control sample, while the air resistance and Cobb decreased 69% and 7.9% in comparison with control sample, respectively. By increasing the nanocellulose content up to 0.9% the opacity increased 0.7%; moreover, the brightness and whitening decreased 3.5% and 10.6, respectively. The scanning electron microscopy results indicated that enhancement *via* nanocellulose decreased the air resistance.

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INTRODUCTION

Paper money is an integral part of trade exchange in our lives and is used in all places and all professions. Bottom combers cotton pulp from textile factories (for high pile cottons combing should be performed) has better efficiency compared to raw cotton (without combing fibers). Because of the low consumption amount of chemical materials needed to bleach these fibers, the physical and mechanical resistance of papers made from bottom combers pulp are mostly equal or sometimes more durable than papers that are made from raw cotton; this factor has importance in the production of durable papers.

Nanotechnology is important for global growth. It has novel capabilities for materials, tools, and systems and has revolutionized technologies and industries (Hubbe *et al*. 2008). However, the application of nanotechnology in cellulose industries is an emerging field. Because of the specific properties and biodegradability of nanocrystals, they have been used purely or as a booster to strength characteristics or physical properties and coating materials in paper industry (Luu *et al*. 2011; González *et al*. 2012).

Cellulose nanofibers are structural units composed of nanofibrils and primary fibrils that during various processes have produced nanostructures with a diameter of less than 100 nm, either in singular form or stuck together. These particles reduce the porosity and can increase the paper's strength. Such effects can be attributed to the development of additional joints and the creation of additional hydrogen bonds during paper drying (Charani *et al*. 2013). With the addition of nanocellulose to paper and common cardboards, the production of layered and coverage products will be possible *via* nanostructures. Among these products, bio-based nanomaterials, especially cellulosic based nanomaterials, have specific importance because of their specific resistance, safety properties, and biodegradability.

Henriksson *et al*. (2008) used wood cellulosic nanofibrils to produce porosity cellulosic nanopapers with a high hardness. One type of nanopaper had a tensile strength of 214 MPa; its tensile strength was greater than the tensile strength of cast iron and was similar to the tensile strength of steel (Henriksson *et al*. 2008). The separation and elimination of nanofibrillated cellulosic (NFC) and its effects on paper tensile strength, which was produced by bleached the chemical pulp of hardwoods, have been considered as well. The results of these experiments showed that the addition of NFC particles without high pile fibers considerably increased the tensile strength of the produced paper in comparison to the addition of particles without the NFC treatment (Madani *et al*. 2011).

The effect of the pulp and NFC mixing time duration on paper properties have been studied as well. The results indicated that increasing the mixing time duration up to 1 h increased the dewatering time and decreased the air permeability (Lacani and Afra 2013). The effect of NFC on the mechanical resistance of a paper made by *Brassica napus* stem has been considered. The results showed that the mechanical properties of the nanofibers developed in comparison with microfibers. Moreover, the amount of hydrogen bonds and the involvement of the fibrils with each other had been increased (Yousefi *et al*. 2011).

In a spinning factory, after opening the cotton jersey and separating its seeds, the portions of cotton that have long fibers are mostly used to prepare high quality threads; therefore, they are passed through a needle prong to separate the short and rough fibers from them, which is called combing the cotton fiber. These fibers are not suitable for the textile industry because of their shortness and roughness and their economic value is less than high pile fibers. However, in terms of length and diameter they are categorized in a range that can be used as a proper raw material in the pulp industry, especially for making durable pulp, *e.g.*, for paper money. Use of bottom combers pulp has decreased the harmful damages due to forest over harvesting, and their intrinsic resistance and clean appearance has drawn the attention of paper producers. The fibers used in the factory to produce banknote paper are the fibers obtained by combing cotton fibers in textile factories. In fact, the fibers that remain above the shoulder are used to prepare the fabric and the fibers that are collected under the shoulder are provided to our factory for production. These fibers are shorter than cotton lint and longer than cotton linters and are known as "bottom combers". Therefore, the primary aim of the present study is to consider the effect of using nanocellulose and chemical pulp on paper money that has been made with bottom combers pulp.

EXPERIMENTAL

Materials

Pulp

The consumed raw cellulosic materials in this study were refined bottom combers pulp and refined chemical pulp, which were both prepared at one factory that produced durable paper; their products were transferred to the laboratory. The obtained pulp was dewatered until it reached a 10% to 15% concentration and then were put in plastic bags and kept in a refrigerator until consumption.

Nano cellulosic fiber

In the present study, 3% nano cellulosic gel, which was produced by Nano Novin Polymer Co. at four different levels (0%, 0.3%, 0.6%, and 0.9%) in relation to the dry weight of pulp, was used. The average diameter of the nanocelluloses was less than 50 nm.

Polyacrylamide

Polyacrylamide used in this study had a molecular weight of 359,000 g/mol and was prepared in a synthesis laboratory using Charani *et al.* (2013) method.

Methods

Preparation of the handsheets

After preparation of the pulp, nanocellulose was added at 4 weight levels, *i.e.*, 0%, 0.3%, 0.6%, and 0.9%, to the chemical pulp and bottom combers pulp at the following ratios: 100% chemical pulp (CHP), 100% bottom combers pulp (BCP), 45% bottom combers pulp + 55% chemical pulp (45% BCP+55% CHP), and 30% bottom combers pulp + 70% chemical pulp (30% BCP+70% CHP); this mixture was made in the presence of 1% polyacrylamide based on the amount of pulp and nanocellulose.

Table 1. Combinations of Different Ratios of Chemical Pulp and Bottom Combers Pulp, Nanocellulose, and Polyacrylamide for Paper Money

Code Treatment	Nanocellulose (phc*)	Bottom combers pulp (%)	Chemical pulp (%)	Polyacrylamide (phc)
100% CHP	0	0	100	1
100% BCP		100	0	1
45% BCP+55% CHP		45	55	1
30% BCP+70% CHP		30	70	1
100% CHP	0.3	0	100	1
100% BCP		100	0	1
45% BCP+55% CHP		45	55	1
30% BCP+70% CHP		30	70	1
100% CHP	0.6	0	100	1
100% BCP		100	0	1
45% BCP+55% CHP		45	55	1
30% BCP+70% CHP		30	70	1
100% CHP	0.9	0	100	1
100% BCP		100	0	1
45% BCP+55% CHP		45	55	1
30% BCP+70% CHP		30	70	1
Phc=Per hundred compound				

The different ratios of chemical pulp and bottom combers pulp, nanocellulose, and polyacrylamide used for handsheet making are shown in Table 1. The obtained mixture was mixed *via* magnetic stirring for 30 min at room temperature. Then, the eight handsheets with a basis weight of 90 g·m^{-2}, based on TAPPI standard T205 sp-06 (2006), were manufactured. The samples were kept in a specific room for paper tests under standard conditions, *i.e.*, a relative humidity (RH) of 50% ± 2% and temperature of 23 °C ± 1 °C, until used in the experiments, according to ISO standard 187 (1990). Eventually, the air resistance, surface smoothness, Cobb, tensile strength index, burst strength index, tear strength index, and folding endurance, as well as the brightness, opacity, and whiteness levels, were measured according to national and international standards which are summarized in Table 2.

Table 2. Tested Physical and Mechanical Characteristics

No.	Properties Tested	Standard
1	Surface smoothness	TAPPI T555 om-04 (2004)
2	Air resistance	TAPPI T460 om-06 (2006)
3	Cobb	ISO 535: 2014
4	Tensile strength index	TAPPI T494 om-06 (2006)
5	Tear strength index	TAPPI T414 om-12 (2012)
6	Burst strength index	TAPPI T403 om-15 (2015)
7	Folding endurance	TAPPI T423 cm-21 (2021)
8	Brightness	TAPPI T452 om-02 (2002)
9	Opacity	TAPPI T425 om-01 (2001)
10	Whiteness	TAPPI T562 om-21 (2021)

Scanning electron microscope (SEM) imaging

The pictorial analysis was performed on the handsheets *via* scanning electron microscope (SEM), model JXA-840, which was located at the Academic Jihad laboratory at the Sharif Industrial University.

Statistical Analysis

This work was analyzed with a completely accidental plot and for processing the results that were obtained from measurements *via* SPSS software (version 22, IBM, Armonk, NY). To perform the statistical analysis, a two-way variance analysis was used and for mean comparison. The Duncan test at a 95% significance level was applied.

RESULTS AND DISCUSSION

The F-value and significance level are shown in Table 3. The direct effects of the paper type and nanocellulose content were significant in terms of the air resistance, surface smoothness, Cobb, tensile strength index, burst strength index, tear strength index, and folding endurance, as well as the brightness, opacity, and whiteness at a 95% significance.

The interaction effects of the pulp and the nanocellulose were also significant in terms of the air resistance, surface smoothness, Cobb, tensile strength index, burst strength index, tear strength index, and folding endurance, as well as the brightness and opacity at a 95% significance; however, it was not significant for whiteness at this level.

Table 3. Variance Analysis (F-value and Significance Level) of the Effects of Nanocellulose Content and Pulp Type on Paper Properties

Variable Properties	Nanocellulose	Pulp Type	Nanocellulose × Pulp Type
Surface smoothness	5.596*	62.861*	17.221*
Air resistance	70.083*	79.511*	5.326*
Cobb	11.720*	3.037*	10.389*
Tensile strength index	23.962*	127.616*	10.214*
Tear strength index	6.361*	7.247*	6.322*
Burst strength index	62.595*	186.054*	13.708*
Folding endurance	24.539*	92.292*	11.823*
Opacity	4.214*	23.640*	4.346*
Brightness	44.391*	36.432*	2.614*
Whiteness	35.632*	45.934*	1.074*
Note: Significance level: *95% ns: non-significance			

Table 4 shows the effects of the type of pulp and nanocellulose on the physical, mechanical, and optical properties of the handsheet papers. As shown, the lowest surface smoothness was related to 100% CHP during the use of 0.3% nanocellulose, which was equal to 1000 mL/min and the highest surface smoothness was due to the use of 100% BCP along with the use of 0.3% nanocellulose, which was equal to 1580 mL/min.

The results of the variance analysis of the surface smoothness showed that there was a significant difference between these amounts at a 95% significance level. By adding 0.9% nanocellulose, an increase in the surface smoothness has been achieved.

Table 4 indicates that the greatest air resistance was found in 100% CHP without using nanocellulose, which was equal to 2017 mL/min. The lowest air resistance was found in 45% BCP+55% CHP composition along with 0.9% nanocellulose, which was equal to 583 mL/min.

According to the achieved results with combining pulp with 30% BCP+70% CHP and 0.9% nanocellulose, a severe reduction in the air resistance factor was observable.

Table 4 showed that the highest Cobb value was found in 100% CHP without using nanocellulose, which was equal to 210.7 $g \cdot m^{-2}$, while the lowest Cobb value was obtained by using 100% CHP along with 0.9% nanocellulose, which was equal to 157.7 $g \cdot m^{-2}$.

One of the most important properties of paper is its ability to control liquid absorption (Ashori and Raverty 2007). The results of the measurements showed that nanocellulose reduced the Cobb value of paper. This was because nanocellulose has a greater molecular mass and greater viscosity; as such, the water permeation speed was reduced so that a reduction in the Cobb value can improve paper printability (Ashori *et al*. 2005). The results showed that the effect of nanocellulose in the chemical fibers was severe in terms of the Cobb value.

Table 5 showed that the greatest tensile strength index was found in 100% CHP and 0.6% nanocellulose, which was equal to 79.5 $Nm.g^{-1}$, and the lowest tensile strength index was related to the use of 100% BCP without using nanocellulose, which was equal to 36.6 $Nm.g^{-1}$.

Table 4. Effect of Pulp Type and Nanocellulose Addition on Surface Smoothness, Air Resistance, and Cobb Properties of Money Paper

Nanocellulose (%)	Code Treatment	Surface Smoothness (mL/min)	Air Resistance (mL/min)	Cobb (g.m^{-2})
0	100% CHP	1177	2017	210.7
	100% BCP	1537	1560	185.7
	45% BCP+55% CHP	1067	1250	196.7
	30% BCP+70% CHP	1100	1200	180.7
0.3	100% CHP	1000	1770	189.0
	100% BCP	1583	1450	166.0
	45% BCP+55% CHP	1300	1100	174.0
	30% BCP+70% CHP	1250	1200	174.7
0.6	100% CHP	1400	1533	167.0
	100% BCP	1333	1450	190.7
	45% BCP+55% CHP	1167	1167	182.0
	30% BCP+70% CHP	1050	850	183.7
0.9	100% CHP	1433	983	157.7
	100% BCP	1450	1150	173.0
	45% BCP+55% CHP	1083	850	199.0
	30% BCP+70% CHP	1300	583	187.0

Table 5. Effects of Pulp Type and Nanocellulose Addition on Tensile Strength Index, Tear Strength Index, Burst Strength Index, and Folding Endurance Properties of Money Paper

Nano-cellulose (%)	Code Treatment	Tensile Strength Index (Nm.g^{-1})	Tear Strength Index (mN.g^{-1})	Burst Strength Index (Kpam2.g^{-1})	Folding Endurance (N)
0	100% CHP	46.8	13.4	2.6	139.3
	100% BCP	36.6	10.6	1.9	42.3
	45% BCP+55% CHP	52.2	12.9	3.1	187.7
	30% BCP+70% CHP	52.6	11.4	2.9	304.0
0.3	100% CHP	72.5	12.8	4.5	2829.3
	100% BCP	40.7	12.9	2.5	65.0
	45% BCP+55% CHP	58.8	12.6	3.5	649.0
	30% BCP+70% CHP	54.1	12.4	3.3	866.7
0.6	100% CHP	79.5	13.2	4.7	2743.7
	100% BCP	38.7	12.3	2.4	67.7
	45% BCP+55% CHP	61.7	12.0	3.6	723.3
	30% BCP+70% CHP	53.1	13.1	3.4	390.0
0.9	100% CHP	75.4	12.5	5.1	1960.3
	100% BCP	42.1	12.3	2.4	120.0
	45% BCP+55% CHP	64.1	13.6	3.9	551.3
	30% BCP+70% CHP	49.4	13.4	3.4	384.3

Tensile strength is the most fitted index for all inter-fiber bonds that is a mix of alternative strengths. Tensile strength is an index for paper tensile potential sturdiness. The most effective factors on strength of a paper measures the variety and quality of fiber bonds. Enhancement of fiber joints will increase paper tensile strength (Kasmani and Samariha 2019). While the durability of a paper invariably is a smaller amount than a fiber (Afra *et al*. 2013). Tensile strength in the machine direction is more than that in crosswise direction, because of fiber becoming aligned in the longitudinal direction more than in the crosswise direction. In machine direction two completely different sets of bonds are stretched: chemical bonds (C-C, C-O), which are inter- and intra-aldohexose units in cellulose chains, and H-bonds between fibers. In sum, there are a lot of valence bonds in the machine direction, and there are fewer valence bonds in a transversal direction. As in handsheet paper, the fibers are settled at varied directions accidentally. As the dimensions become smaller up to the nanometer scale, the precise surface of cellulose fibers will increase. This means that there will be a lot of functional groups at the nanofiber level that will be able to produce H-bonding with adjacent nanofibers and eventually, thus building a network of nanofibers (Yousefi *et al*. 2011) that will increase its strength.

Table 5 indicates that the lowest tear strength index was found in 100% BCP and 0% nanocellulose, which was equal to 950 $mN.g^{-1}$, and the highest tear strength index was found in 30% BCP+70% CHP composition along with 0.9% nanocellulose, which was equal to 1230 $mN.g^{-1}$.

In that report, the effective factors of such changes were identified as the intrinsic strength of the fibers and the level of hydrogen bonds. To explain the behavior of the tearing strength of combining papers, it can be said that the shortening of the fibers during the refining action causes a reduction in the tearing strength. However, an enhancement of their bonds level causes an improvement in strength. By combining papers, the effective factor on strength reduction under the effect of super grinding surpasses the further factor of resistance improvement. However, the addition of NFC and its establishment between the fiber and the creation of an obstacle for direct bonds between fibers can all cause a reduction in the resistance of the fiber network in comparison to the tearing stress (Afra *et al*. 2013).

Table 5 showed that the highest burst strength index was found in 100% CHP and 0.9% nanocellulose, which was equal to 456 $KPam^2g^{-1}$ and the lowest burst strength index was found with the use of 100% BCP without using nanocellulose, which was equal to 175 $KPam^2g^{-1}$.

Table 5 shows that the greatest folding endurance was found in 100% CHP and 0.3% nanocellulose, which was equal to 2830 N, and the lowest folding endurance was related to the use of 100% BCP without using nanocellulose, which was equal to 42 N.

The results of the variance analysis of the folding endurance showed that there was a significant difference among the different treatments at a 95% significance level. The results indicated that the effect of nanocellulose was severe on the chemical fibers and combining pulp had the greatest effect with 0.3% nanocellulose.

Table 6 indicated that the lowest opacity value was found in 100% CHP and 0.9% nanocellulose, which was equal to 90%, and the highest opacity value was found in 30% BCP+70% CHP composition along with 0.3% nanocellulose, which was equal to 92.4%.

The results of the variance analysis on the opacity treatments showed that there was a significant difference between these amounts at a 95% significance level. By adding 0.9% nanocellulose, the opacity increased. The highest level of opacity was obtained in the papers that were made by combining 30% BCP+70% CHP and 0.3% nanocellulose.

Table 6 showed that the highest brightness value was found in 100% CHP without using nanocellulose, which was equal to 83% and the lowest brightness value was found in 30% BCP+70% CHP composition with 0.3% nanocellulose, which that was equal to 75%. The results of the variance analysis on the brightness treatments showed that there was a significant difference between these amounts at a 95% significance level. The brightness was reduced with the addition of 0.9% nanocellulose.

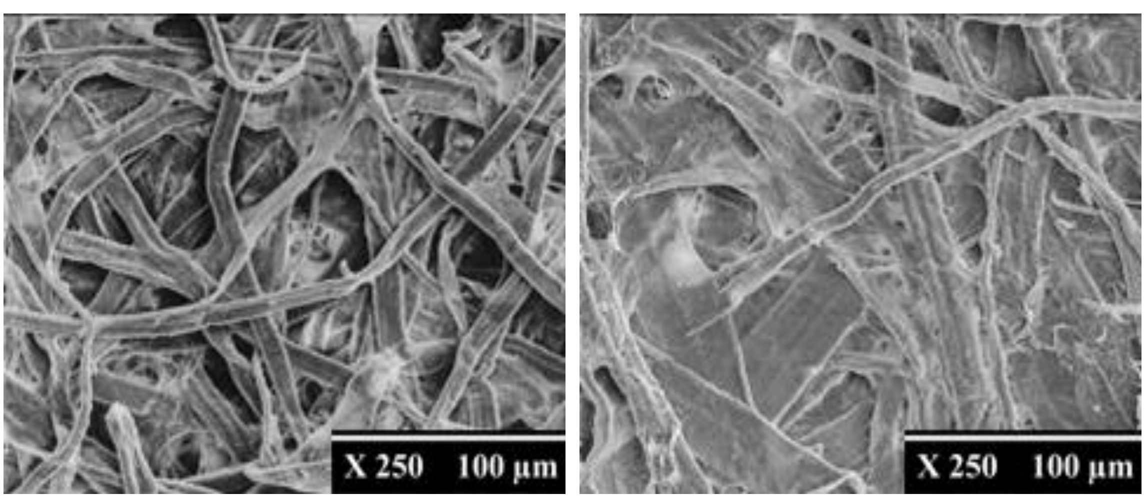
Table 6. Effects of Pulp Type and Nanocellulose Addition on the Opacity, Brightness, and Whiteness Properties of Money Paper

Nanocellulose (%)	Code Treatment	Opacity (%)	Brightness (%)	Whiteness (%)
0	100% CHP	90.3	81.9	62.0
	100% BCP	90.8	82.5	67.7
	45% BCP+55% CHP	91.2	81.1	62.3
	30% BCP+70% CHP	90.2	82.4	67.6
0.3	100% CHP	90.2	78.7	57.8
	100% BCP	90.5	81.1	63.2
	45% BCP+55% CHP	92.4	75.3	61.3
	30% BCP+70% CHP	91.3	78.3	65.5
0.6	100% CHP	90.4	79.5	56.7
	100% BCP	90.6	80.6	62.7
	45% BCP+55% CHP	91.8	76.6	59.3
	30% BCP+70% CHP	91.3	78.7	63.2
0.9	100% CHP	90.0	79.2	56.4
	100% BCP	91.4	80.5	61.4
	45% BCP+55% CHP	91.4	77.5	56.3
	30% BCP+70% CHP	92.2	79.6	60.8

Table 6 showed that the highest whiteness value was found in 100% BCP and 0% nanocellulose, which was equal to 67.7%, and the lowest whiteness value was found in 30% BCP+70% CHP composition with 0.9% nanocellulose, which was equal to 56.3%.

The results of the variance analysis on the whiteness treatments showed that there was a significant difference between these amounts at a 95% significance level. The whiteness value was reduced with the addition of 0.9% nanocellulose. When chemical pulp and bottom combers pulp were combined, increasing the chemical pulp percentage reduced the paper brightness and whiteness.

Figures 1 through 4 show the surface fibers of the treated handsheet papers with pulp and nanocellulose. It is observable in these figures that the pulp type did not affect the fiber surface, but the addition of nanocellulose can cover the fiber surface in a way that during the use of 0.9% nanocellulose lower air permeability can be observed.



(a) (b)

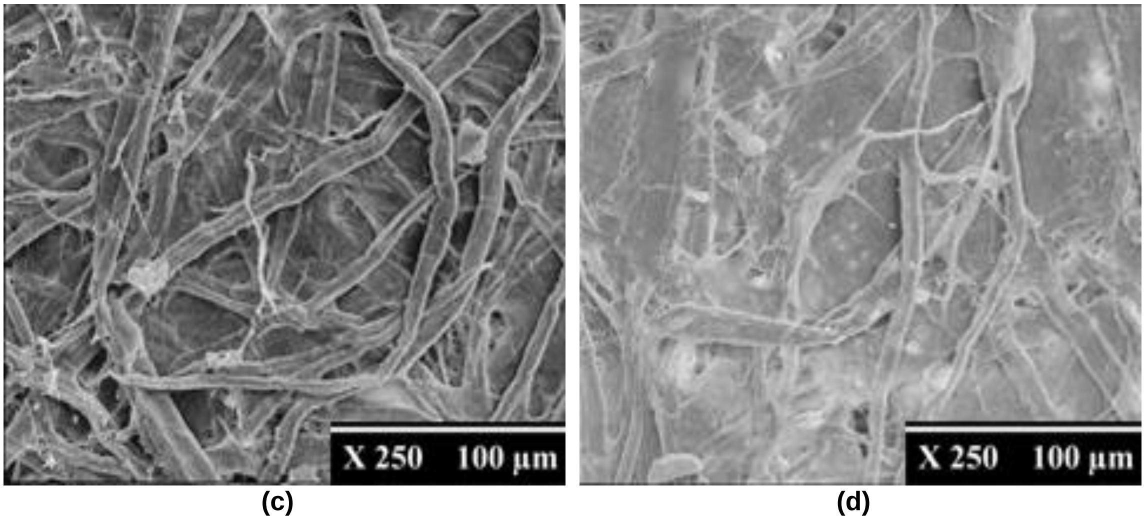


Fig. 1. The surface of paper made by 100% CHP: a) 0% nanocellulose; b) 0.3% nanocellulose; c) 0.6% nanocellulose; and d) 0.9% nanocellulose (Fathi and Kasmani 2019)

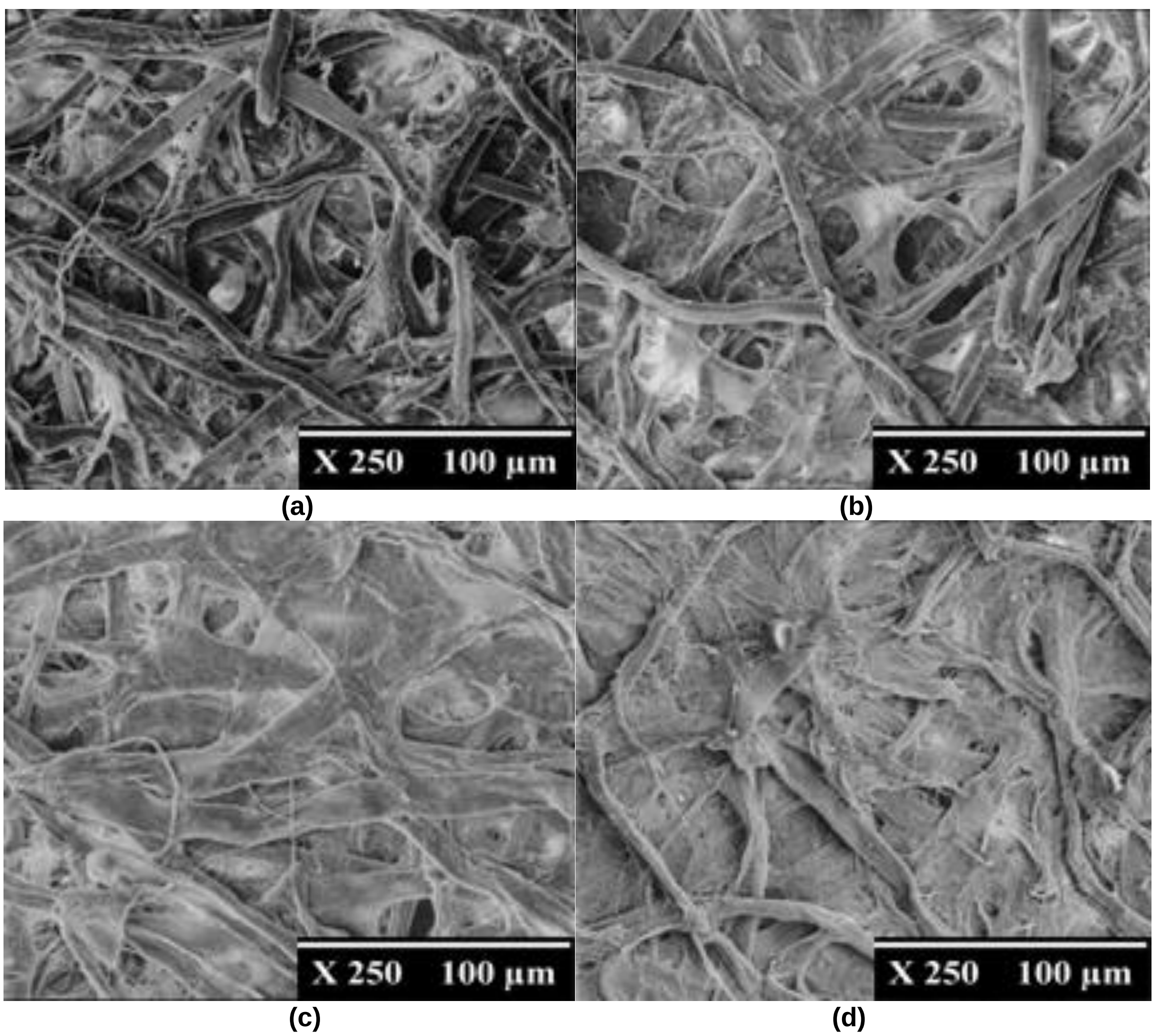


Fig. 2. The surface of paper made by 100% BCP: a) 0% nanocellulose; b) 0.3% nanocellulose; c) 0.6% nanocellulose; and d) 0.9% nanocellulose

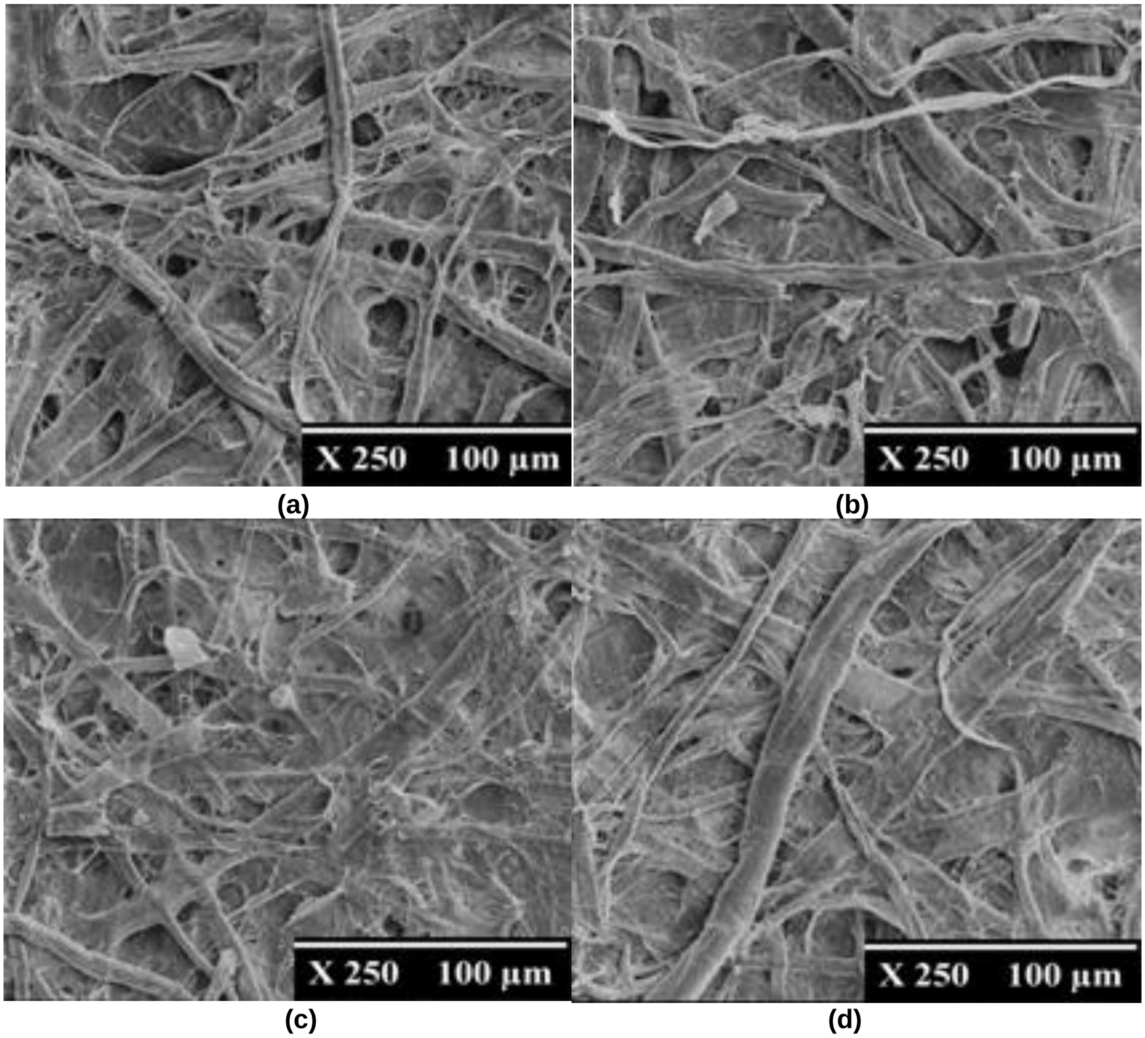
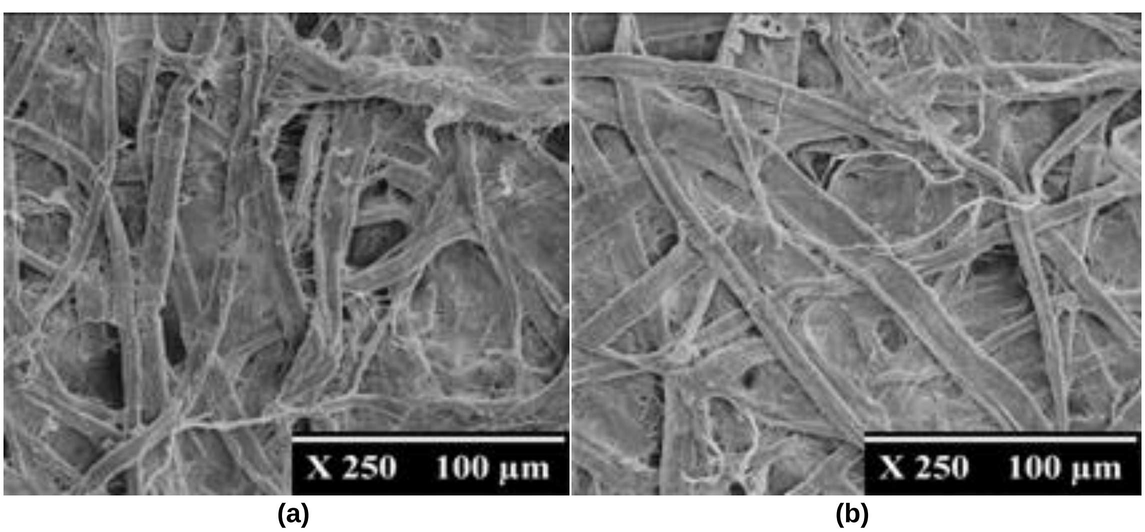


Fig. 3. The surface of paper made by 45% BCP+55% CHP composition: a) 0% nanocellulose; b) 0.3% nanocellulose; c) 0.6% nanocellulose; and d) 0.9% nanocellulose



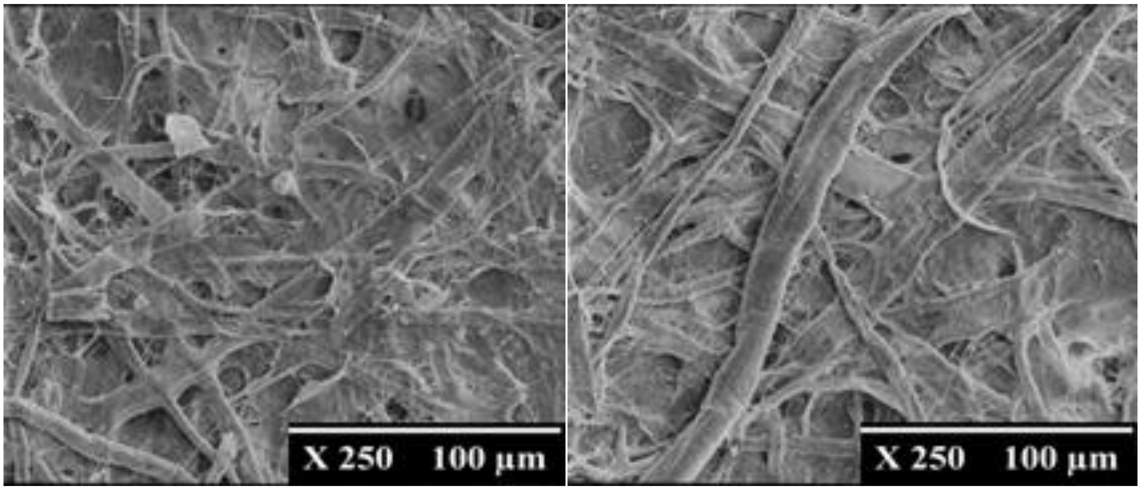


Fig. 4. The surface of paper made by 30% BCP+70% CHP composition: a) 0% nanocellulose; b) 0.3% nanocellulose; c) 0.6% nanocellulose; and d) 0.9% nanocellulose

CONCLUSIONS

1. The papers that were made with 100% chemical pulp (CHP) along with 0.6% nanocellulose, 45% bottom combers pulp (BCP)+55% CHP and 0.9% nanocellulose, 100% CHP with 0.9% using nanocellulose, or 100% CHP with 0.3% using nanocellulose, had greater tensile strength index, burst strength index, tear strength index, and folding endurance, comparing to the other pulps respectively.
2. With the addition of up to 0.9% nanocellulose to 100% BCP, the tensile strength index, burst strength index, tear strength index, and folding endurance increased 15%, 16.4%, 21.7%, and 183.7% respectively, when compared to the 100% BCP.
3. By increasing the nanocellulose content up to 0.9% in BCP, the surface smoothness, and the air resistance and Cobb decreased by 6%, 35.7%, and 7.3%, respectively, when compared to the 100% BCP.
4. By increasing the nanocellulose content up to 0.9% in BCP, the opacity increased by 0.7% when compared to the 100% BCP. The brightness and whiteness decreased by 2.5% and 10.3%, respectively, when compared to the 100% BCP.
5. The scanning electron microscopy (SEM) results showed that increasing the nanocellulose percentage reduced the air resistance.
6. Regarding the results of the production of durable papers based on the components, a combination of 45% BCP+55% CHP composition with 0.9% nanocellulose yielded an improvement in most of the tested parameters.

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