Effects of Nanocellulose, Cationic Starch, Cationic Polyacrylamide, and Unbleached Softwood Kraft Fibers on the Properties of Recycled Cardboard

Jafar Ebrahimpour Kasmani , and Ahmad Samariha , b

Separate and combined effects of nanocellulose, cationic polyacrylamide, cationic starch, and bleached softwood kraft fibers were evaluated when producing recycled packaging fluting paper. The focus was on enhancing the structural integrity and performance of this paper product, which is essential for packaging applications. Treatments included 10% refined bleached softwood kraft pulp, 5% cellulose nanofibers, 2% cationic starch, and 0.2% cationic polyacrylamide. Combined treatments involved 5% cellulose nanofibers with 2% starch and 5% cellulose nanofibers with 0.2% cationic polyacrylamide. Handsheets with a grammage of 127 g/m² were produced and tested for physical, mechanical, and microscopic properties. Results showed that these additives, either independently or in combination, improved the properties of the paper. The combination of 5% nanocellulose and 0.2% cationic polyacrylamide yielded the highest density and tensile strength, along with the lowest water absorption. This treatment also enhanced critical strengths in the ring crush and corrugated medium tests, making it optimal for packaging paper production. Electron microscopy revealed reduced porosity in handsheets from combined treatments, which may negatively impact water absorption. Further research is needed to optimize these additives while addressing their effects on water absorption.

DOI: 10.15376/biores.20.3.6599-6614

Keywords: Old corrugated container; Cellulose nanofibers; Cationic starch; Cationic polyacrylamide; Recycled packaging fluting paper; Physical and mechanical properties; Packaging cardboards

Contact information: a: Department of Wood and Paper, Sava.C., Islamic Azad University, Svadkooh, Iran; b: Department of Wood Industry, National University of Skills (NUS), Tehran, Iran; *Corresponding author: kasmani@iau.ac.ir

INTRODUCTION

The recycling of used paper packaging is the most common form of cellulosic product recycling, particularly in Iran. It offers several benefits, including reducing urban waste volume, providing economic and social advantages, and decreasing the need for virgin fibers, which ultimately leads to less exploitation of forests (Afra 2005). However, the use of extreme levels of refining of a minor portion of the fibers, to compensate for the strength loss of recycled paper, has garnered interest over time. While these methods can enhance certain properties, they may also cause severe damage to the fibers and reduce recycled fiber consumption. This poses challenges to dewatering and other production characteristics, including the well-known disadvantages of using nanocellulose (Afra 2005; Afra *et al.* 2015; Ebrahimpour Kasmani *et al.* 2023).

A critical factor in paper production, especially when using recycled fibers, is enhancing fiber bonding to improve the mechanical properties of the final product. Despite

the economic and environmental benefits of using recycled paper, it is important to note that recycled fibers generally have weaker characteristics compared to virgin fibers. The primary issue with recycled paper is the lower mechanical properties resulting from the hornification of fibers in the course of repeated recycling. To address these challenges,, various chemicals, such as starch and polyacrylamide, are often employed to improve fiber bonding and restore the properties of recycled paper (González *et al.* 2012).

Polyacrylamides are one of the most widely used polyelectrolytes in the paper industry. This organic compound forms bonds between fibers through hydrogen bonding between primary amide groups and cellulose molecules, and it also acts as a flocculant and retention aid in papermaking. Cationic polyacrylamide, due to its positive charge and polymeric structure, prevents the loss of fine nanofibers, thereby increasing the amount of nanofibers present in the pulp and enhancing fiber bonding, which improves the properties of the resulting paper. The addition of nanofibers and cationic polyacrylamide has led to increased retention (Tajik *et al.* 2016). Using additives can significantly enhance fiber bonding strength (Hamzeh and Haftkhani-Rostampour 2008).

Typically, the properties of paper pulp obtained from old corrugated containers (OCC) do not provide sufficient strength for creating durable sheets. The average fiber length decreases while fiber softness increases in the course of repteated recyling (Hamzeh et al. 2012). However, the incorporation of specific nanomaterials, such as cellulose nanofibers, in conjunction with cationic additives can significantly enhance the strength, formation, and durability of paper pulp. The unique characteristics of these nanomaterials, including their high surface area and ability to form large numbers of hydrogen bonds, contribute to improved inter-fiber bonding and overall paper quality. This composite approach improved performance and production efficiency, which is increasingly recognized in the paper industry (Hadilam et al. 2013). The integration of nanomaterials with cationic additives enhances the overall durability of the paper pulp, particularly in terms of water resistance and strength retention during the production process.

Recent research highlights the use of cellulose nanofibers as a promising method to improve paper properties (Hassan *et al.* 2011). While the commercial application of these nanomaterials faces challenges—such as high costs and low dispersibility—studies have demonstrated their effectiveness. Specifically, the greater hydrophilicity of cellulose nanofibers contributes to enhanced strength and durability by promoting better inter-fiber bonding and moisture retention. For instance, adding nanofibrillated cellulose (NFC) to eucalyptus bleached pulp can reduce the need for refining without compromising mechanical properties (González *et al.* 2012).

Additionally, studies have shown that combining nanocellulose with cationic starch can lead to significant increases in tensile strength and reduced air permeability (Kajanto and Kosonen 2012). Comparisons between cellulose nanofibers and gypsum nanoparticles indicate that cellulose nanofibers outperform in mechanical properties due to their superior hydrophilicity, improving strength, and reducing porosity (Hrabalova *et al.* 2011).

Research focusing on nanofibrillated cellulose has reported substantial improvements in properties such as tensile and tear strength, and density, as well as enhanced durability during paper formation (Pourkarim Dodangeh *et al.* 2016). These improvements can be attributed to the increased inter-fiber bonding and the structural reinforcement provided by the cellulose nanofibers. Currently, the paper industry relies on commercially available dry strength additives, such as cationic starch and polyacrylamide, to compensate for strength deficiencies (Pourkarim Dodangeh *et al.* 2016).

Cationic starch has a positive charge that enhances bonding between fibers and fillers, which in turn improves the overall quality of the paper. Although the addition of cationic starch increases paper strength, it may slightly decreases opacity. Research has shown that cationic starch effectively enhances the physical properties of paper, particularly its strength properties (Hubbe 2014).

Adding unbleached softwood kraft pulp can neutralize the negative impact of shorter recycled fibers on tear resistance, strengthening the paper pulp (Moradian *et al.* 2016).

The recycling industry has become increasingly important in pulp and paper production due to environmental and economic concerns, including the depletion of primary wood resources and the growing demand for paper and cardboard products. Recycling paper and cardboard presents a sustainable solution that conserves resources and reduces waste.

Given these challenges, improving fiber bonding and enhancing strength properties is vital. Additionally, the Iranian pulp and paper industry faces challenges due to reliance on imported pulp, highlighting the need for innovative solutions. Using nanocellulose, either alone or in combination with cationic starch and cationic polyacrylamide, can reduce energy consumption and production costs while improving sustainability and achieving desired strength properties.

The aim of this research is to compare the performance of separate and combined systems —namely, cellulose nanofibers, cationic starch, and cationic polyacrylamide—in improving properties and reducing unbleached softwood kraft pulp usage in the production of recycled packaging cardboard.

EXPERIMENTAL

In this research, the materials were sourced from an industrial production line, and tests were conducted under standard laboratory conditions to ensure that the results could be applied for further research and industrial applications.

Materials

Pulp

The pulp utilized in this study was sourced from Dena Cellulose Factory in Tehran, Iran. In this factory, the waste paper collected from the city is processed through the pulper, followed by the usage of detergents, coarse and fine sieves, disc filter, thickener, and disperser, resulting in recycled pulp. Unbleached softwood kraft pulp was acquired from Irkutsk Oblast Co. located in Ust-Ilimsk, Russia. Prior to any treatments or handsheet production, the pulp underwent refining in a PFI mill to reach a consistency of 400 mL CSF. The pulps were then dewatered to a consistency of 10 to 15%, packaged in plastic bags, and stored in a refrigerator until required.

Cellulose nanofibers

Cellulose nanofibers were sourced from Nano Novin Polymer Company (Gorgan, Iran), produced from unbleached pulp using mechanical forces (up-down mechanism). The average particle diameter of the nanocellulose was around 50 nm.

Cationic starch

Cationic starch was sourced from Lyckeby Amylex, Slovakia, from potato. The starch had a pH of approximately 6, a degree of substitution (D.S.) of approximately 0.035 mol/mol, a protein content of 1.5%, nitrogen content of 0.25%, and moisture content of 10% based on wet weight. The starch solution was prepared by dissolving 0.5 g of pure starch in 100 cm³ of distilled water. While stirring, the temperature was controlled, and the flask was covered with aluminum foil to prevent evaporation. The flask was heated to 90 °C over 30 min and maintained at this temperature for an additional 30 min. The starch solution was freshly prepared daily to avoid changes in viscosity and concentration.

After the initial preparation of old cardboard paper pulp and unbleached softwood kraft pulp with additives, six treatment groups were created, and handsheets were made for each group.

Cationic Polyacrylamide

Cationic polyacrylamide with a molecular mass of 359,000 g/mol and an average electrical charge density was obtained from Degussa (Düsseldorf, Germany). The cationic polyacrylamide was diluted to a concentration of 1.5% with distilled water based on its purity. A total of 10 mL of this solution was added to a volumetric flask, filled with distilled water to a volume of 100 mL. The contents were heated and mixed for 3 h using a stirrer at 500 rpm. The mixture was stored in the refrigerator for 24 h and stirred for 20 min before use (Rezayati Charani *et al.* 2013).

Handsheet Preparation

Recycled cardboard and unbleached CNF, along with other additives depending on the treatment conditions and specified weight percentages, were mixed together in the blender for 10 minutes and then used to produce 127-gram paper.

To clarify the composition of the handsheet treatments, the recycled cardboard served as the primary pulp source for all formulations. Subsequently, the following treatments were carried out, concentrating on the effects of the additional components:

Table 1. Composition of the Treatments and the Amounts of Nanocellulose,
Cationic Starch, and Polyacrylamide

No.	Code	OCC Pulp	Unbleached softwood kraft pulp	Nanocellulose	Cationic Polyacrylamide	Cationic Starch
1	0LP	100	0	0	0	0
2	10LP	90	10	0	0	0
3	5NC	95	0	5	0	0
4	0.2CPAM	99.8	0	0	0.2	0
5	5NC +0.2CPAM	94.8	0	5	0.2	0
6	2CS	98	0	0	0	2
7	5NC+2CS	93	0	5	0	2

Preparation of Handsheets Papers

Handsheets with a basis weight of 127 g/m² were prepared according to the TAPPI T205 sp-02 (2002) standard using a KCL handsheet making machine (model KCL 2000; KCL, Stockholm, Sweden). Ten handsheets were produced for each treatment.

Measurement of Paper Properties

The physical and mechanical properties of the handsheets were measured according to TAPPI standards, as presented in Table 2. A laboratory scale with a precision of 0.01 g and a thickness gauge with a precision of 1 µm were used for physical property measurements. The tensile strength was measured using a machine from L&W Company (model L&W 900; Stockholm, Sweden), burst strength was assessed with a machine from Drick Company (model DBK 300; Hangzhou, China), and tear strength was measured using another machine from L&W Company (model L&W 900; Stockholm, Sweden). The resistance to the ring crush test (RCT) and corrugated medium test (CMT) were evaluated using a device manufactured by Hangzhou Zhibang (model ZBJ-200; Hangzhou, China).

Properties	Standard No.		
Density	TAPPI T410 om-02 (2002)		
Water absorption	TAPPI T441 om-09 (2009)		
Tensile strength	TAPPI T494om-01 (2001)		
Burst strength	TAPPI T403om-02 (2002)		
Tear strength	TAPPI T414om-04 (2004)		
RCT	TAPPI T818 cm-97 (1997)		
CMT	TAPPLT809 om-99 (2006)		

 Table 2. Properties of the Tested Handsheets Papers

FE-SEM Analysis

To analyze the microscopic structure of the control and treated handsheet samples, a MIRA3 TESCAN (TESCAN, Brno, Czech Republic) scanning electron microscope with a magnification of 200x was utilized. This equipment allowed for detailed examination of the surface morphology and fiber arrangement in the handsheets.

Statistical Calculations

The experimental design employed in this research was completely randomized. Data analysis was performed using one-way analysis of variance (ANOVA), with Duncan's test applied to group the averages at a confidence level of 95%. Statistical analyses were conducted using SPSS software (IBM, version 23.0, Armonk, NY, USA) to ensure the reliability and validity of the results.

RESULTS AND DISCUSSION

The ANOVA table (Table 3) demonstrates the significant differences between the examined properties of the seven handsheet groups, which include the control sample and samples with additives, both independently and in combination. The results indicate that the effects of additives on various properties—such as density, water absorption, tensile strength, burst strength, tear strength, resistance to the RCT, and resistance to the CMT—were significant at a confidence level of 95%. This suggests that the incorporation of additives has a meaningful impact on the performance characteristics of the handsheets, supporting the hypothesis that these materials enhance the properties of recycled packaging paper.

Property	Density (g/m³)	Water Absorption (g/m²)	Tensile Strength Index (Nm/g)	Tear Strength Index (mN.m²/g)	Burst Strength Index (Kpa/g)	RCT (kN/m)	CMT (kN/m)	
F value	21.941*	7178.0*	8.936*	15.853*	24.913*	57.596*	93.883*	
Sig.	0.000	0.000	0.000	0.000	0.000	0.000	0.000	
*Significant at the 95% confidence level: ns not significant at the 95% confidence level								

Table 3. Analysis of Variance (F-Value and Significance Level) of the Effect of Treatments on the Properties of Handsheets

Effect of Treatments on Handsheets Density

The impact of treatments on the density of the seven handsheet groups, created from old cardboard pulp and various additives, is presented in Fig. 1. The average densities were analyzed using Duncan's test, which identified five distinct groups among the samples. Notably, the highest density was recorded for the treatment with 5% nanocellulose and 0.2% cationic polyacrylamide, measuring 0.587 g/m³. Conversely, the lowest density was found in the treatment containing 10% unbleached softwood kraft pulp, which measured 0.503 g/m³.

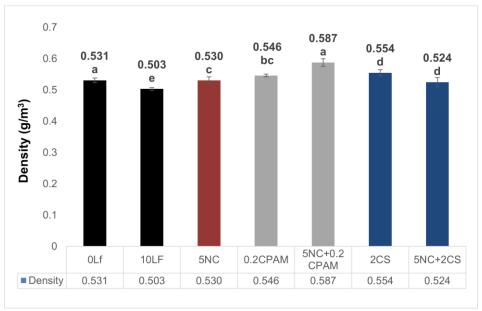


Fig. 1. Average density of handsheets papers

Density is a critical structural property of paper that affects nearly all mechanical, physical, and electrical properties, especially in packaging applications. It influences properties such as modulus of elasticity, tensile strength, and compression strength. In a dense structure minimizes the twisting and warping of fibers, thereby enhancing resistance to bending stresses. Apparent density is crucial for predicting paper strength and typically increase with improved inter-fiber bonding (Main *et al.* 2015).

The incorporation of cationic polyacrylamide into the handsheet significantly increased its density due to the enhanced contact surface and inter-fiber bonding, thereby strengthening the network between cellulose fibers (Afra *et al.* 2015; Gharehkhani *et al.* 2015).

The addition of starch separately has also increased the density, primarily due to the absorption of starch and the development of bonds between fibers, as well as the replacement of bonds with stronger energies, such as electrostatic bonds with hydrogen bonds (Ashori 2006). As a result, the paper had become denser; however, nanocellulose was not been able to enhance the fiber-to-fiber bonding like starch due to its negative surface charge. This increase in density plays a crucial role in improving the overall performance and durability of the paper, rendering it more appropriate for packaging applications.

Effect of Treatments on Handsheets Water Absorption

The effect of treatments on the water absorption of the handsheets are illustrated in Fig. 2. The comparison of average water absorption capacities, analyzed *via* Duncan's test, categorized the samples into six distinct groups. As shown in Fig. 2, the lowest water absorption was observed in the handsheet containing 5% nanocellulose and 2% starch, measuring 197 g/m². Conversely, the highest water absorption was noted in the handsheet with 0.2% cationic polyacrylamide, at 245 g/m².

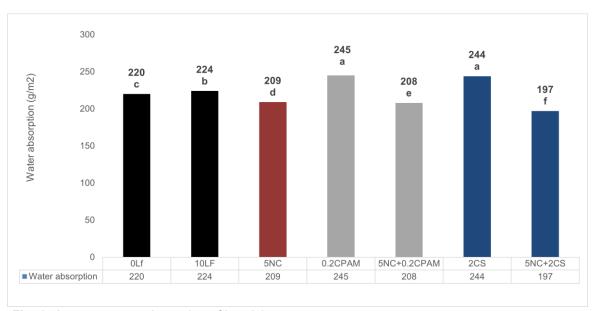


Fig. 2. Average water absorption of handsheet papers

In treatments where additives were used separately, water absorption increased. However, when a combination of additives was used, this parameter decreased.

These results indicate that the incorporation of certain additives can significantly influence the water absorption characteristics of the paper. The lower water absorption of the handsheet with nanocellulose and starch suggests improved inter-fiber bonding and a denser structure, which can effectively limit the penetration of water.

This includes theories about the bonding of positive charges in starch and negative charges on the cellulose fiber surface, where the reduction of certain hydroxyl groups and water attractants leads to decreased water absorption in the resulting paper (Song *et al.* 2018).

Effect of Treatments on Tensile Strength Index

The effect of treatments on the tensile strength index of the handsheets is shown in Fig. 3. All additives, both independently and in combination, significantly increased the tensile strength index of the handsheets compared to the control sample. Duncan's grouping categorized the averages of the tensile strength index into three independent groups. The highest tensile strength index, associated with the treatment of 5% nanocellulose and 0.2% cationic polyacrylamide, reached 71.6 Nm/g, placing it in group C.

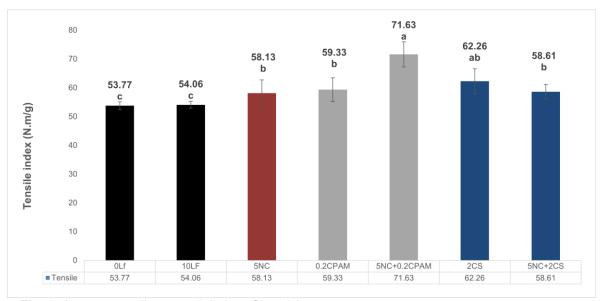


Fig. 3. Average tensile strength index of handsheets papers

The number and quality of fiber bonding connections are crucial factors affecting the tensile strength of paper (Darstan *et al.* 2013).

As a long-chain polymer with cationic charge density, cationic polyacrylamide bonds to fiber surfaces, creating bridges between the components of pulp and paper slurry. This results in better formation and durability. The density, proximity, and quality of these bonds ultimately enhance the tensile strength of the paper (Pourkarim Dodangeh *et al.* 2021).

By adding starch and cationic polyacrylamide, especially in combination with nanocellulose, these materials fill the spaces between the fibers. This increases the bonding surface area in the fibers and the bonding interface, resulting in enhanced tensile strength (Nogi *et al.* 2009).

Nanocellulose can also contribute to the tensile strength by increasing the number of hydrogen bonds and enhancing the bonding between fibers due to their higher specific surface area and physical interactions with pulp fibers. This leads to a stronger fiber network. The uniform distribution of stress, facilitated by the high specific surface area of nanocellulose, promotes the expansion of the inter-fibrous bond network, increasing the fiber-to-fiber contact surface and accessibility of hydroxyl groups.

Studies have reported improvements in both dry and wet tensile strength of paper when using micro/nanofibrillated cellulose in paper pulp (Hassan *et al.* 2011; Hadilam *et al.* 2013). Nanomaterials not only enhance the bonding capability to fibers but also improve the tensile strength of the paper layer. Recent advancements have focused on applying

anionic micro/nanomaterials alongside long-chain cationic polymers, a strategy known as complex flocculation, which enhances both processing and product properties. The anionic nature and high specific surface area of cellulose nanofibers have shown positive effects when combined with biopolymers such as starch in paper pulp (Pourkarim Dodangeh *et al.* 2021).

The Effect of Treatments on Burst Strength Index

The effect of treatments on the burst strength index of the handsheets is shown in Fig. 4. All additives notably increased the burst strength index compared to the control sample. Duncan's grouping categorized the averages of the burst strength index data into five independent groups. The highest burst strength index was associated with the treatment of 5% nanocellulose and 2% cationic starch, achieving a value of 2.37 kPa/g, which placed it in group E.

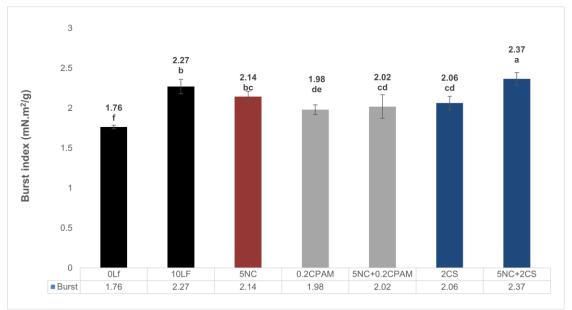


Fig. 4. The average Burst strength index of handsheets

The bursting strength of paper is primarily influenced by the degree of bonding between fibers, with a significant impact from the inter-fiber connections (Kiaei *et al.* 2016). The highest burst strength observed with the combination of 5% cellulose nanofibers and 2% starch has been corroborated by several studies (Kiaei *et al.* 2016; Amani *et al.* 2021). Starch, due to its cationic charge, demonstrates good compatibility with the anionic surfaces of cellulose fibers, facilitating the formation of electrostatic bonds (Pourkarim Dodangeh *et al.* 2021).

The strong bonding capability of starch with cellulose fibers enables the establishment of amide, ionic, and hydrogen covalent bonds, enhancing the overall strength of the paper. Additionally, cellulose nanofibers contribute to increased burst strength due to their high aspect ratio and the physical contact they establish with other fibers within the paper structure (Ebrahimpour Kasmani and Mahdavi 2020). This combination of factors leads to improved structural integrity and performance in applications requiring burst resistance.

The Effect of Treatments on Tear Strength Index

The effects of treatments on the tear strength index of the handsheets are shown in Fig. 5. Duncan's grouping categorized the tear strength index averages of different treatments into four separate groups. The highest tear strength index, placed in group D, corresponded to the 10% unbleached softwood kraft pulp, while the lowest tear strength index, found in group A, corresponded to the combination of 5% nanocellulose and 0.2% cationic polyacrylamide.

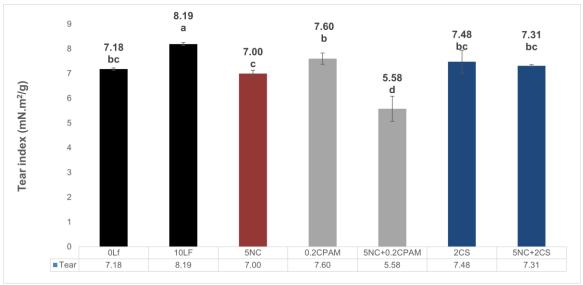


Fig. 5. The average tear strength index of handsheets

The tear strength index is a critical characteristic used to evaluate paper quality. This index is influenced by several factors, including the average length of the fibers, their inherent resistance, the degree of bonding between fibers, and the orientation of the fibers (Yousefhashemi *et al.* 2019). It is important to note that anything that makes the paper more brittle will tend to lower the tear strength, including increased inter-fiber bonding. As a result, tear strength often shows discrepancies with other strength measures, such as tensile strength, particularly when refining processes or bonding agents are applied. This aspect is particularly significant for the qualitative assessment of paper and cardboard, especially in packaging applications.

Recycled paper pulp typically exhibits weaker bonding abilities and fibers, which underscores the importance of inter-fiber bonding in achieving higher tear strength with the addition of certain additives. The inclusion of 10% unbleached softwood kraft pulp demonstrated a positive impact on improving the tear strength index by increasing both the average length and diameter of the fibers, thereby enhancing the overall structural integrity of the paper. This improvement is vital for applications where durability and resistance to tearing are essential.

The Effect of Treatments on the Ring Crush Test

Duncan's grouping categorized the averages of the RCT index into four independent groups, as shown in Fig. 6. The lowest RCT value, averaging 0.8 kN/m, corresponded to the treatment with 10% unbleached softwood kraft pulp, placed in group

0.2

■ RCT

1.6

1.31

1.10

1.10

1.12

1.18

1.18

1.18

1.18

1.10

1.10

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1.11

1

A. In contrast, the highest RCT value, averaging 1.38 kN/m, was associated with the combination of 5% nanocellulose and 0.2% cationic polyacrylamide, placed in group D.

Fig. 6. Average index of RCT of handsheets

0.80

1.10

The ring crush test is a critical strength indicator for cardboard used in packaging applications. This strength is influenced by various factors, including fiber refinement, the type of resin used, chemical additives, and the characteristics of the fibers themselves. The addition of positively charged cationic polyacrylamide enhances the cationic charge on the surface of the anionic cellulose fibers in the paper, effectively absorbing negatively charged particles when combined with cellulose nanofibers.

0.2CPAM

5NC+0.2CPAM

1.38

5NC+2CS

0.94

5NC

The sequential use of positive and negative polyelectrolytes improves the stability of cationic polyacrylamide and cellulose nanofibers in the paper matrix, resulting in enhanced RCT performance. Main *et al.* (2015) reported a 17% increase in RCT strength in paper pulp prepared from fibers with the addition of 1.5% cationic polyacrylamide. However, it is noteworthy that increasing the amount of this additive to 2% resulted in a decrease in strength (Main *et al.* 2015). This highlights the importance of optimizing additive concentrations to achieve the desired mechanical properties in paper products.

The addition of polyacrylamide, whether separately or in combination with cationic starch and nanocellulose, enhances the cationic charge on the surface of the anionic cellulose fibers in the paper. As a result, the anionic nanocellulose is attracted and, in fact, the sequential use of positive and negative polyelectrolytes retains more fines in the wet paper, leading to greater RCT resistance in the resulting paper (Wagberg *et al.* 2002).

The Effect of Treatments On Corrugated Medium Test

Duncan's grouping categorized the averages of the CMT into five different groups, as illustrated in Fig. 7. The rate of changes in CMT for the handsheet papers showed a pattern similar to that observed in the RCT. The lowest CMT value, averaging 145 $N \cdot m^2/g$, corresponded to the treatment with 10% unbleached softwood kraft pulp, placed in group A. In contrast, the highest crushing resistance, measured at 223 $N \cdot m^2/g$, corresponded to the combination of 5% nanocellulose and 0.2% cationic polyacrylamide, placed in group E.

The corrugated medium test is a critical strength indicator for cardboard used in packaging applications. As previously noted, polyacrylamide, both individually and in

combination with other additives, has been shown to enhance CMT strength. Main *et al.* (2015) reported a 26% increase in CMT strength in paper pulp prepared from fibers when incorporating 1.5% cationic polyacrylamide. However, it is important to note that increasing the amount of this additive to 2% resulted in a decrease in strength (Main *et al.* 2015). This underscores the necessity of carefully optimizing additive concentrations to maximize the mechanical properties of paper products while avoiding potential negative effects from overuse.

Cationic polyacrylamide enhances strength through two main mechanisms, both separately and in combination with other additives. The first mechanism involves increasing the bonding between fibers, typically enhancing the number of hydrogen bonds that occur at the fiber junctions. This process acts as a form of chemical hydration of the fibers. The second mechanism is that cationic polyacrylamide, with its positive charge, effectively connects to negatively charged recycled fibers, functioning as an electronic bridge that improves the bonding strength between fibers. Additionally, it helps restore lost surface points of the fibers and enhances inter-fiber adhesion (Darvishzadeh *et al.* 2014).

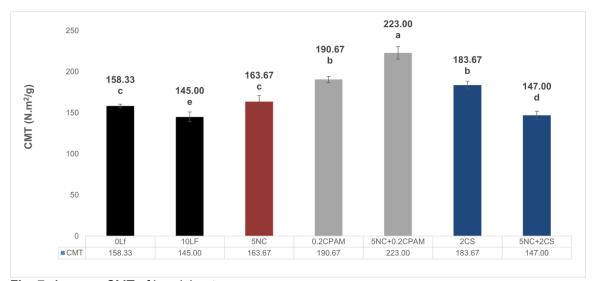


Fig. 7. Average CMT of handsheets

FE-SEM Analysis

Figure 8 presents micrographs obtained from a field emission scanning electron microscope (FE-SEM) for various treatments: 0% unbleached softwood kraft pulp (a), 10% unbleached softwood kraft pulp (b), 5% cellulose nanofibers and 2% starch (c), and 5% cellulose nanofibers with 0.2% cationic polyacrylamide (d).

This observation aligns with findings reported by Sanchez-Salvador *et al.* (2020), Trinh *et al.* (2023), Ebrahimpour Kasmani *et al.* (2022), highlighting the role of nanocellulose in enhancing the structural integrity of paper by minimizing pore sizes.

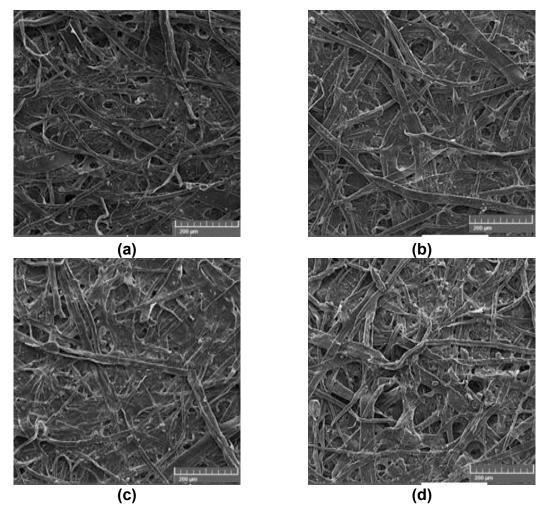


Fig. 8. Microscopic structure of handsheets papers: (a) 0% unbleached softwood kraft pulp, (b) 10% unbleached softwood kraft pulp, (c) 5% cellulose nanofibers and 2% starch, and (d) 5% cellulose nanofibers and 0.20% poly cationic acrylamide

CONCLUSIONS

In conclusion, the use of various additives can significantly enhance the properties of recycled paper made from old cardboard pulp, yielding technical, economic, and environmental benefits. Through improving the characteristics of packaging papers (liner and fluting) through the incorporation of additives, the added value of the final product increases, making it suitable for producing strong and exportable cartons. Additionally, enhancing the quality of paper reduces the reliance on virgin unbleached softwood kraft pulp, thereby decreasing the consumption of coniferous wood and contributing to environmental preservation.

- 1. The results of this research demonstrated that different additives, whether used independently or in combination, can have varying effects on the properties of old cardboard paper pulp. Most additives, either alone or in combination, were shown to improve the properties of the cardboard.
- 2. Notably, the combined treatment of 5% nanocellulose and 0.2% cationic

- polyacrylamide emerged as the optimal treatment for producing packaging paper, enhancing both the RCT and the CMT—key properties in packaging cardboard manufacturing.
- 3. However, microscopic images revealed a reduction in the porosity of handsheet papers when cellulose nanofibers and polyacrylamide were added compared to the control sample. This reduction in porosity could adversely affect the wet-end dewatering capabilities of the paper machine.
- 4. Therefore, further research is necessary to optimize the use of these additives, ensuring that the benefits are maximized while minimizing any negative impacts on paper machine operations.

REFERENCES CITED

- Afra, E. (2005). Fundamental of Paper Properties, Aeej Publication, Tehran, Iran.
- Afra, E., Mohammadi, M., Imani, R., Narchin, P., and Roshani, S. (2015). "Improvement of the antibacterial properties of sanitary papers using by silver nano particles," *Journal of Wood and Forest Science and Technology Research* 22(2), 119-136.
- Amani, N., Vaysi, R., Najafi, A., and Ebadi, S. E. (2021). "Investigation of the effect of using activated sludge treated with acetic acid on physical and strength properties of recycled pulp," *Sadraj* 72(3), article 298. DOI: 10.5552/drvind.2021.2112
- Ashori, A. (2006). "Pulp and paper from kenaf bast fibers," *Fibers and Polymers* 7, 26-29. DOI: 10.1007/BF02933598
- Darstan, R., Zabihzadeh, M., and Nazarnezhad, N. (2013). "Papermaking properties of *Carpinus betulus* with kraft, soda and soda-urea pulping processes," *Iranian Journal of Wood and Paper Industries* 3(2), 105-117.
- Darvishzadeh, O., Jahan Latibari, A., Sepidehdam, S. M. J., and Tajdini, A. (2014). "Effect of simultaneous addition of cationic starch and bentonite on some properties of writing and printing paper," *Forest and Wood Products* 66(4), 519-527.
- Ebrahimpour Kasmani, J., and Mahdavi, S. (2020). "Utilization of nano-ligno-cellulose to be replaced with the imported long-fiber pulp in durable paper made from waste lint pulp," *Forest and Wood Products* 73(2), 151-162.
- Ebrahimpour Kasmani, J., Samariha, A., and Khakifirooz, A. (2023). "Comparison of separate and combined effect of nanosilica and cationic additives on the optical properties of the white packaging liner," *Iranian Journal of Wood and Paper Science Research* 38(4), 266-280.
- Ebrahimpour Kasmani, J., Samariha, A., and Mahdavi, S. (2022). "Investigation of the independent and combined effect of different additives on the properties of liner and fluting paper made from OCC," *Iranian Journal of Wood and Paper Science Research* 37(3), 204-216.
- Gharehkhani, S., Sadeghinezhad, E., Kazi, S. N., Yarmand, H., Badarudin, A., Safaei, M. R., and Zubir, M. N. M. (2015). "Basic effects of pulp refining on fiber properties—A review," *Carbohydrate Polymers* 115, 785-803. DOI: 10.1016/j.carbpol.2014.08.047
- Hadilam, M. M., Afra, E., and Yousefi, H. (2013). "Effect of cellulose nanofibers on the properties of bagasse paper," *Forest and Wood Products* 66(3), 351-366.
- Hamzeh, Y., Najafi, S. M. H., Hubbe, M. A., Salehi, K., and Firouzabadi, M. R. D. (2012). "Recycling potential of unbleached and bleached chemical pulps from

- juvenile and mature wood of Populus deltoides," *Holzforschung* 66, 155-161. DOI: 10.1515/HF.2011.141
- Hamzeh, Y., and Rostampurhafthkani, A., (2008). *Principles of Papermaking Chemistry*, University of Tehran Press, 424 pp.
- Hassan, E. A., Hassan, M. L., and Oksman, K. (2011). "Improving bagasse pulp paper sheet properties with microfibrillated cellulose isolated from xylanase-treated bagasse," *Wood and Fiber Science* 43(1), 76-82.
- Hrabalova, M., Schwanninger, M., Wimmer, R., Gregorova, A., Zimmermann, T., and Mundigler, N. (2011). "Fibrillation of flax and wheat straw cellulose: Effects on thermal, morphological, and viscoelastic properties of poly (vinylalcohol)/fibre composites," *BioResources* 6(2), 1631-1647. DOI: 10.15376/6/2/1631-1647
- Hubbe, M. A. (2014). "Prospects for maintaining strength of paper and paperboard products while using less forest resources: A review," *BioResources* 9(1), 1634-1763. DOI: 10.15376/biores.9.1.1634-1763
- Kajanto, I., and Kosonen, M. (2012). "The potential use of micro-and nanofibrillated cellulose as a reinforcing element in paper," *Journal of Science & Technology for Forest Products and Processes* 2(6), 42-48.
- Kiaei, M., Samariha, A., and Farsi, M. (2016). "Effects of montmorillonite clay on mechanical and morphological properties of papers made with cationic starch and neutral sulfite semichemical or old corrugated container pulps," *BioResources* 11(2), 4990-5002. DOI: 10.15376/biores.11.2.4990-5002
- Main, N. M., Talib, R. A., Rahman, R. A., Mohamed, A. Z., Ibrahim, R., and Adnan, S. (2015). "Effect of amphoteric and cationic polyacrylamide on the structural and strength properties of coir paper," *Procedia Manufacturing* 2, 28-34. DOI: 10.1016/j.promfg.2015.07.006
- Moradian, M. H., Rezayati Charani, P., and Saadat Niya, M. (2016). "Improving paper breaking length using cellulosic nanofibers in bagasse pulp," *Forest and Wood Products* 69(3), 603-614.
- Nogi, M., Iwamoto, S., Nakagaito, A. N., and Yano, H. (2009). "Optically transparent nanofiber paper," *Advanced Materials* 21(16), 1595-1598. DOI: 10.1002/adma.200803174
- Pourkarim Dodangeh, H., Jalali Torshizi, H., and Rudi, H. (2021). "Cationic polyacrylamide/ cellulose nanofibril polyelectrolytes effect on suspension and network properties of packaging recycled fibers," *Journal of Applied Research of Chemical-Polymer Engineering* 5(1), 3-15.
- Pourkarim Dodangeh, H., Jalali Torshizi, H., Rudi, H., and Ramezani, O. (2016). "Performance of nano fibrillated cellulose (NFC) and chitosan bio-polymeric system on recycled pulp and paper properties of old corrugated containers (OCC)," *Iranian Journal of Wood and Paper Industries* 7(2), 297-309.
- Rezayati Charani, P., Dehghani-Firouzabadi, M., Afra, E., Blademo, Å., Naderi, A., and Lindström, T. (2013). "Production of microfibrillated cellulose from unbleached kraft pulp of kenaf and Scotch pine and its effect on the properties of hardwood kraft: Microfibrillated cellulose paper," *Cellulose* 20, 2559-2567. DOI: 10.1007/s10570-013-9998-z
- Sanchez-Salvador, J. L., Balea, A., Monte, M. C., Negro, C., Miller, M., Olson, J., and Blanco, A. (2020). "Comparison of mechanical and chemical nanocellulose as additives to reinforce recycled cardboard," *Scientific Reports* 10(1), article 3778. DOI: 10.1038/s41598-020-60507-3

- Song, Z., Li, G., Guan, F., and Liu, W. (2018). "Application of chitin/chitosan and their derivatives in the papermaking industry," *Polymers* 10(4), article 389. DOI: 10.3390/polym10040389
- TAPPI T205 sp-02 (2002). "Forming handsheets for physical tests of pulp," TAPPI Press, Atlanta, GA, USA.
- TAPPI T403 om-02 (2002). "Bursting strength of paper," TAPPI Press, Atlanta, GA, USA.
- TAPPI T410 om-02 (2002). "Grammage of paper and paperboard (weight per unit area)," TAPPI Press, Atlanta, GA, USA.
- TAPPI T414 om-04 (2004). "Internal tearing resistance of paper," TAPPI Press, Atlanta, GA, USA.
- TAPPI T441 om-09 (2009). "Water absorptiveness of sized (non-bibulous) paper, paperboard, and corrugated fiberboard (Cobb test)," TAPPI Press, Atlanta, GA,USA.
- TAPPI T494 om-01 (2001). "Tensile breaking properties of paper and paperboard," TAPPI Press, Atlanta, GA, USA.
- TAPPI T809 om-99 (2006). "Flat crush of corrugating medium (CMT test)," TAPPI Press, Atlanta, GA, USA.
- TAPPI T818 cm-97 (1997). "Ring crush of paperboard (RCT test)," TAPPI Press, Atlanta, GA, USA.
- Trinh, B. M., Chang, B. P., and Mekonnen, T. H. (2023). "The barrier properties of sustainable multiphase and multicomponent packaging materials: A review," *Progress in Materials Science* 133, article 101071. DOI: 10.1016/j.pmatsci.2023.101071
- Wagberg, L., Forsberg, S., Johansson, A., and Juntti, P. (2002). "Engineering of fibre surface properties by application of the polyelectrolyte multilayer concept. Part I: Modification of paper strength," *Journal of Pulp and Paper Science* 28(7), 222-228.
- Yousefhashemi, S. M., Khosravani, A., and Yousefi, H. (2019). "The effect of addition of lignocellulose nanofiber produced from old corrugated container pulp on recycled paperboard properties," *Iranian Journal of Wood and Paper Industries* 9(4), 575-584.

Article submitted: April 11, 2025; Peer review completed: May 18, 2025; Revised version received and accepted: June 12, 2025; Published: June 24, 2025. DOI: 10.15376/biores.20.3.6599-6614