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CELLULOSE NANOFIBERS (CNF) AND CELLULOSE NANOCRYSTALS (CNC) AS HIGH-PERFORMANCE FILLERS IMPROVING THE MECHANICAL, OPTICAL, AND SURFACE PROPERTIES OF RECYCLED NEWSPAPER

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ABSTRACT: The increasing focus on sustainability and waste reduction has led to significant interest in recycling paper products. This study investigates the use of two types of nanocellulose (CNF and CNC) as filler materials in the production of recycled newspaper. The aim is to evaluate the effects of these nanocelluloses on the mechanical, optical, and surface properties of the recycled newspaper (ONP). Mechanical properties such as tensile strength, burst strength, and tear resistance were measured to determine the material's durability. Optical properties, including brightness, whiteness, and yellowness, were assessed to understand the aesthetic quality of the recycled paper. Surface roughness values (Ra, Rz, Rq) were also analyzed to evaluate the surface quality, which is essential for printability. The results show that CNF significantly improved the tensile and tear strengths, as well as reduced the yellowness, making the paper more visually appealing. CNC, on the other hand, demonstrated a notable enhancement in burst strength but had a less pronounced effect on other properties. This study demonstrates that nanocellulose, particularly CNF, is a promising additive for improving specific mechanical and optical characteristics of recycled newspaper, making it more suitable for various applications.

Keywords: Nanocellulose, newspaper, mechanical, optical, surface

SELÜLOZ NANOLİFLERİNİN (CNF) VE SELÜLOZ NANOKRİSTALLERİNİN (CNC) GERİ DÖNÜŞTÜRÜLMÜŞ GAZETE KAĞIDININ MEKANİK, OPTİK VE YÜZEY ÖZELLİKLERİNİ İYİLEŞTİRMEDE YÜKSEK PERFORMANSLI DOLGU MALZEMELERİ OLARAK KULLANIMI

ÖZET: Sürdürülebilirlik ve atık azaltma üzerine artan odak, kağıt ürünlerinin geri dönüştürülmesine olan ilgiyi önemli ölçüde artırmıştır. Bu çalışma, geri dönüştürülmüş gazete kağıdı üretiminde iki tür nanoselülozun (CNF ve CNC) dolgu malzemesi olarak kullanımını incelemektedir. Amaç, bu nanoselülozların geri dönüştürülmüş gazete kağıdının (ONP) mekanik, optik ve yüzey özellikleri üzerindeki etkilerini değerlendirmektir. Malzemenin dayanıklılığını belirlemek için çekme mukavemeti, patlama mukavemeti ve yırtılma direnci gibi mekanik özellikler ölçülmüştür. Geri dönüştürülmüş kağıdın estetik kalitesini anlamak için parlaklık, beyazlık ve sarılık gibi optik özellikler değerlendirilmiştir. Baskı kalitesi açısından kritik olan yüzey kalitesini belirlemek amacıyla yüzey pürüzlülüğü değerleri (Ra, Rz, Rq) analiz edilmiştir. Sonuçlar, CNF'nin çekme ve yırtılma mukavemetini önemli ölçüde artırdığını ve sarılığı azaltarak kağıdı görsel olarak daha çekici hale getirdiğini göstermiştir. Diğer yandan CNC, patlama mukavemetinde belirgin bir iyileşme sağlarken diğer özellikler üzerinde daha az etkili olmuştur. Bu çalışma, özellikle CNF'nin, geri dönüştürülmüş gazete kağıdının belirli mekanik ve optik özelliklerini iyileştirmede umut verici bir katkı maddesi olduğunu ortaya koymaktadır ve bu tür kağıtların çeşitli uygulamalar için daha uygun hale getirilmesini sağlamaktadır.

Anahtar kelimeler: Nanoselüloz, gazete kağıdı, mekanik, optik, yüzey

INTRODUCTION

In recent years, sustainability and circular economy practices have become central to the global agenda, driven by growing environmental concerns and the urgent need to reduce waste and resource consumption (Abushammala et al., 2023; Sopelana et al., 2021). The paper and pulp industry, a significant contributor to deforestation, water consumption, and industrial emissions, has increasingly adopted recycling initiatives to mitigate its environmental impact. Recycling paper products, particularly newspapers, has emerged as one of the most effective strategies for reducing raw material use and waste (Abushammala et al., 2023; Dick & Malvessi, 2022; Ozola et al., 2019; Van Ewijk et al., 2020). However, despite these environmental benefits, recycled paper tends to exhibit inferior mechanical, optical, and surface properties compared to its virgin fiber counterparts. This has led researchers to explore ways to enhance the performance of recycled paper, thereby making it suitable for a wider range of applications (Brancato et al., 2007; Małachowska et al., 2023; Okayama, 2002; Wanrosli et al., 2005). Among the many potential solutions, the use of nanocellulose as a filler and reinforcing agent has gained considerable attention for its promising ability to improve the quality of recycled fibers.

Nanocellulose, derived from the breakdown of cellulose into nanometer-sized fibers, possesses unique properties that make it an attractive material for enhancing paper. Its high surface area, excellent mechanical strength, and renewable nature offer significant potential for improving the physical and structural integrity of recycled paper. Numerous studies have explored the impact of nanocellulose on paper properties, particularly in recycled paper applications (Indarti et al., 2023; Li et al., 2021; Sanchez-Salvador et al., 2020; Yi et al., 2023). However, most of these studies have primarily focused on the general reinforcement effects of nanocellulose, without providing a detailed comparative analysis of cellulose nanofibrils (CNF) and cellulose nanocrystals (CNC) in recycled newsprint applications. Additionally, while prior research has demonstrated improvements in mechanical properties, there remains limited understanding of how these nanocelluloses affect the optical and surface characteristics of recycled newspaper, which is crucial for printability and end-use performance.

Nanocellulose can be incorporated into paper formulations through various modification techniques, including bulk addition (direct incorporation into the fiber suspension), surface application via size press, and film coating methods (Li et al., 2021; Mazega et al., 2022; Pego et al., 2020; Sharma et al., 2020). Each approach offers unique advantages depending on the intended end-use of the paper. Size press and film coating methods are particularly useful for enhancing surface properties such as printability, water resistance, and barrier performance (Brancato et al., 2007; Tozluoğlu & Fidan, 2023; Van Nguyen & Lee, 2021). However, these techniques generally result in surface modifications rather than bulk reinforcement and may require additional processing steps thereby increasing production complexity and cost. In contrast, bulk addition, which involves the direct incorporation of nanocellulose into the fiber suspension before sheet formation, enables nanoscale fibrils to interact directly with the recycled fibers, reinforcing the fiber network from within. This method has been shown to be particularly effective in improving mechanical strength by enhancing fiber-fiber bonding, as well as influencing optical and surface properties more uniformly throughout the paper matrix (Balea et al., 2020; Sanchez-Salvador et al., 2020). Given that recycled newspaper typically exhibits weakened fiber bonding due to prior mechanical and chemical treatments, the bulk incorporation of nanocellulose was chosen as the most suitable approach for achieving comprehensive reinforcement across multiple paper properties. Furthermore, this method aligns with industrially viable papermaking processes, as it does not require additional post-processing steps and can be seamlessly integrated into conventional recycling operations.

Nanocellulose, in the form of cellulose nanofibrils (CNF) or cellulose nanocrystals (CNC), has been extensively studied for its capacity to increase the tensile strength, burst strength, and tear resistance of paper materials (Bárta et al., 2023; Campano et al., 2018; Indarti et al., 2023; Pego et al., 2020; Perdoch et al., 2022; Yi et al., 2023). This is achieved primarily through its ability to reinforce the fiber matrix, promote hydrogen bonding between fibers, and reduce the porosity of the paper structure. Balea et al., (2020) and Campano et al., (2018) demonstrated that the incorporation of nanocellulose significantly improves the mechanical properties of recycled paper, making it more durable and resistant to mechanical stresses. These studies have laid the groundwork for understanding the role of nanocellulose in reinforcing recycled fibers, particularly in applications involving low-strength papers, such as newsprint and packaging.

However, the advantages of nanocellulose are not limited to mechanical reinforcement. The unique fibrillar network formed by nanocellulose within the paper matrix also has implications for optical properties, which are critical for applications such as printing and packaging where, visual quality is paramount (Lourenço et al., 2020; Pego et al., 2020; Perdoch et al., 2022). Brightness, whiteness, and yellowness are key optical attributes that directly affect the aesthetic and functional quality of recycled paper products. The inherent presence of contaminants, such as ink residues, fillers, and lignin in recycled fibers poses significant challenges for achieving high levels of brightness and whiteness in recycled paper (Jamnicki Hanzer et al., 2021; Radić

Seleš et al., 2020; Zeb et al., 2021). Moreover, aging and the degradation of fiber quality during the recycling process further exacerbate these optical deficiencies. Nanocellulose, by virtue of its nanoscale dimensions and its ability to modify the paper's internal structure, has shown potential to address some of these optical limitations. Li et al., (2021) found that the addition of nanocellulose could improve the light-scattering properties of the paper, thereby enhancing its brightness. Nonetheless, the degree to which different types of nanocellulose influence optical properties in recycled paper remains insufficiently underexplored.

Additionally, surface quality is another crucial aspect of paper products, particularly in contexts where printability and coating adhesion are important. Surface roughness plays a pivotal role in determining the paper’s ability to accept and hold ink during the printing process. A smoother surface typically results in sharper print quality and more efficient ink transfer (Ataeefard, 2014; Aydemir et al., 2021; Havenko et al., 2020). In recycled paper, the surface quality can be compromised due to irregularities introduced during the fiber recovery process, such as fiber entanglement, filler deposition, and fiber breakage (Balea et al., 2018; Wistara et al., 1999). Nanocellulose has been suggested as a potential solution for improving surface properties by filling in surface voids and creating a more uniform and compact fiber network. Imani et al., (2019) and Lourenço et al., (2020b) reported improvements in surface smoothness when nanocellulose was used as a coating material, highlighting its potential to enhance printability and coating performance. However, the interaction between different types of nanocellulose and recycled newspaper fibers, in terms of surface properties, remains a subject that warrants further investigation.

The present study seeks to fill these gaps by systematically exploring the effects of two distinct types of nanocellulose, cellulose nanofibrils (CNF) and cellulose nanocrystals (CNC), as filler materials in the production of recycled newspaper. While both types of nanocellulose have been extensively studied for their ability to improve the mechanical properties of various paper products, previous research has primarily focused on their general reinforcing effects in different paper grades, such as kraft paper and packaging board (Balea et al., 2020; Campano et al., 2018; Indarti et al., 2023). However, comparative studies specifically targeting recycled newspaper remain scarce, particularly with regard to the simultaneous evaluation of both mechanical and optical performance as well as surface roughness, which is a critical parameter for printability.

MATERIALS AND METHODS

Materials

The base material for this study was old newspaper (ONP), obtained from post-consumer sources. Cellulose nanofibrils (CNF) have a high aspect ratio and excellent mechanical reinforcement properties, while cellulose nanocrystals (CNC) are known for their rigidity and crystalline structure. In this study, both CNF and CNC were obtained from commercial suppliers specializing in nanocellulose production. The raw material used for the nanocellulose was bleached kraft pulp derived from softwood fibers, ensuring high purity and minimal residual lignin content.

The CNF was produced through high-pressure homogenization, in which the pulp fibers were subjected to multiple passes through a mechanical shear system, leading to fibrillation and nanoscale fiber formation. This process results in CNF with long, entangled fibrils, a high surface area, and superior hydrogen bonding capacity, making it ideal for reinforcing paper matrices.

On the other hand, CNC was obtained via sulfuric acid hydrolysis, a process that selectively degrades amorphous cellulose regions while preserving the highly crystalline segments. This method yields rigid, rod-shaped nanocrystals with high crystallinity and enhanced surface charge due to sulfate ester groups, contributing to better dispersion in aqueous suspensions. Both nanocellulose types were delivered as aqueous suspensions with a solid content of 5 wt%.

Additional materials included deionized water, which was used for diluting the nanocellulose suspensions and preparing the pulp slurries. The recycled pulp was prepared by mechanically defibrating the newspaper in a standard laboratory disintegrator (ISO 5263). No chemical treatments were applied to remove ink or other contaminants, as the focus was on the intrinsic effects of nanocellulose on the mechanical, optical, and surface properties of the recycled fibers.

Preparation of Recycled Pulp and Nanocellulose Blends

Recycled newspaper was disintegrated in deionized water at a consistency of 5% (w/w) for 20 minutes at 3000 rpm. The resulting pulp slurry was filtered and washed to remove residual contaminants. The pulp was then divided into four portions, each representing a different CNF and CNC addition level: 0%, 10%, 20%, and 30% by weight of dry recycled fiber. Although these addition levels may seem high compared to conventional papermaking practices, they were selected to systematically evaluate the upper limits of nanocellulose reinforcement in recycled newspaper applications. Previous studies have typically explored nanocellulose dosages in the range of 1–10% (Balea et al., 2020; Indarti et al., 2023); however, our approach aims to provide a comprehensive understanding of both the benefits and the practical limitations of higher dosages. From an industrial and commercial perspective, the feasibility of incorporating nanocellulose at such levels depends on factors such as cost, processing efficiency, and compatibility with existing paper recycling infrastructure. While 30% may not be economically viable for large-scale production, the performance trends observed at different dosage levels can guide optimization efforts towards identifying a commercially practical balance between mechanical improvement and cost-effectiveness. Based on our findings, the most effective concentration that provides substantial improvements while maintaining feasibility is around 10–20%, aligning with the practical application limits reported in prior industrial studies.

Each blend of pulp and nanocellulose fibers was stirred for 10 minutes to ensure uniform dispersion of the nanocellulose within the fiber matrix. Achieving a homogeneous distribution of nanocellulose within recycled fiber suspensions is critical for ensuring consistent improvements in paper properties. Given its high surface area and tendency of nanocellulose to form aggregates, careful mixing and dispersion techniques were employed to promote even incorporation. After mixing, the slurry was diluted to a consistency of 1% and formed into standard handsheets using a Rapid Kothen RK-21 paper machine (ISO 5269-2). The handsheets, each with a basis weight of approximately 60 g/m², were dried and conditioned at 23°C and 50% relative humidity for 24 hours before testing.

Determination of Paper Properties

Mechanical testing was performed to assess the impact of nanocellulose addition on the strength properties of the recycled paper. The tensile strength of the samples was evaluated according to ISO 1924-2 using an Instron Universal Testing Machine (Model 3345) equipped with a 5 kN load cell. The tests were conducted at a crosshead speed of 10 mm/min. Burst strength was measured in accordance with ISO 2758 standards, using a Mullen-type burst tester to determine the burst resistance of the handsheets. Tear resistance, as specified in ISO 1974, was assessed using an Elmendorf-type tear tester. For each mechanical property, five replicates were tested, and the mean values were recorded to ensure accuracy and reliability of the results.

The optical properties of the recycled paper, including brightness, whiteness, and yellowness, were measured to evaluate the effect of nanocellulose on the aesthetic qualities of the paper. All measurements of optical properties were conducted using a Datacolor Elrepho device. ISO Brightness was assessed according to ISO 2470-1, with results reported as the percentage of reflectance at 457 nm. Whiteness was determined based on the CIE whiteness index, following ISO 11475 standards. The yellowness index was evaluated in accordance with ASTM E313, where higher values indicate a greater degree of yellowing in the paper. Each optical property was tested five times to ensure the accuracy of the measurements, with the mean values used for further analysis.

Surface roughness was evaluated using a Surftest SJ-201 roughness tester (Mitutoyo). The Ra, Rz and Rq values were measured over a length of 2 cm with a cut-off length of 0.8 mm. This test was performed to assess the impact of nanocellulose on surface quality, which is critical for printing applications. The rougher surfaces exhibit higher Ra values, indicating lower printability. Five replicates were tested per sample, and the mean Ra values were recorded for analysis.

Statistical Analysis

Pearson correlation analysis was conducted using SPSS to determine the relationships between nanocellulose concentration levels (CNF and CNC) and the mechanical, optical, and surface properties of the recycled paper. Pearson's correlation coefficients (r) were used to evaluate the strength and direction of the linear relationships between the variables, with significance levels set at $p < 0.05$. The analysis focused on identifying whether increasing concentrations of CNF and CNC resulted in significant improvements or detriments in the properties of the recycled newspaper.

Statistical significance was set at $p < 0.05$. Additionally, the signal-to-noise ratios (S/N ratios) were calculated for key performance indicators to evaluate the robustness of the results using the Taguchi method, based on the assumption that "larger is better" for mechanical strength properties and "smaller is better" for surface roughness and yellowness.

RESULTS AND DISCUSSION

Mechanical Properties

The mechanical properties, including tensile strength, burst strength, and tear resistance, were evaluated, and the results are presented in Table 1.

Table 1. Effect of CNF and CNC on the mechanical properties of recycled newspaper

Nanocellulose	Tensile Strength (N)		Burst Strength (kPa)		Tear Resistance (gf)	
Cons. (%)	CNF	CNC	CNF	CNC	CNF	CNC
0	51.4 (2.80)		2.44 (0.34)		10.0 (0.00)	
10	59.7 (2.48)	54.8 (2.98)	2.74 (0.55)	2.12 (0.31)	9.50 (0.58)	9.50 (0.56)
20	70.4 (2.54)	61.6 (2.19)	2.38 (0.27)	2.79 (0.25)	11.0 (0.62)	10.0 (0.58)
30	61.1 (2.39)	59.8 (2.32)	2.23 (0.29)	3.09 (0.15)	10.0 (0.59)	9.50 (0.47)

*Values in parentheses represent standard deviations

The mechanical properties of recycled newspaper, as influenced by the addition of CNF and CNC nanocellulose, demonstrate notable differences based on the nanocellulose type and concentration. The tensile strength results, as shown in the table, indicate that CNF nanocellulose provides a more pronounced improvement in mechanical strength, particularly at the 20% concentration level, where tensile strength peaks at 70.36 N. This improvement can be attributed to the ability of cellulose nanofibrils (CNFs) to form a more cohesive network within the paper matrix, enhancing fiber-fiber bonding. Previous studies, such as those by Hsieh et al., (2003), have reported similar findings, noting that CNFs are particularly effective at reinforcing recycled paper due to their high aspect ratio and surface area, which enable for greater interaction with the fibers.

In contrast, cellulose nanocrystals (CNCs) show less significant improvement in tensile strength across all concentrations, with a maximum value of 61.59 N at the 20% concentration level. While CNCs provide increased rigidity due to their high crystallinity, their shorter aspect ratio and lower entanglement capacity limit their ability to reinforce the fiber network as effectively as CNFs. However, beyond crystallinity, the rheological properties of the nanocellulose suspensions also play a crucial role in determining their reinforcement efficiency (Liu et al., 2017; Moberg et al., 2017; J. Xu et al., 2024). CNF suspensions exhibit shear-thinning and gel-like behavior, facilitating better interaction with fibers and leading to stronger inter-fiber hydrogen bonding (Ghosh et al., 2017; Jowkarderis & Van De Ven, 2014). In contrast, CNC dispersions behave more like colloidal suspensions, in which their lower viscosity and reduced interaction with fibers result in weaker stress transfer within the paper matrix (Oguzlu et al., 2017; Y. Xu et al., 2020). The reduced improvement in tensile strength observed with CNCs may therefore be attributed to a combination of lower fiber entanglement, weaker hydrogen bonding, and differences in suspension rheology, all of which influence how nanocellulose integrates into the recycled fiber network. This is consistent with findings from Bai et al., (2019) and Xu et al., (2013), who observed that CNCs, though rigid, do not provide the same level of inter-fiber bonding as CNFs due to their lower surface area and lack of fibrillar structure.

Furthermore, the burst strength values indicate that CNC performs better than CNF at higher concentrations, particularly at 30%, where a burst strength of 3.09 kPa was recorded. This finding aligns with research by Zeng et al., (2021), who suggested that the crystalline nature of CNCs contributes to the paper's resistance to out-of-plane forces, making them more suitable for enhancing burst strength. The tear resistance results, however, are more consistent across both nanocellulose types, with CNF slightly outperforming CNC at 20%, a reflection of the greater flexibility and toughness imparted by CNFs, as noted in Hu et al., (2021), Jele et al., (2022) and Lu et al., (2017).

Table 1 shows that while tensile and burst strength improve, tear resistance does not follow the same trend. This discrepancy arises because tear resistance depends not only on fiber bonding but also on fiber flexibility and energy dissipation (Kärenlampi, 1996; Kärenlampi et al., 1996). CNF forms a dense network that increases stiffness, limiting fiber mobility and reducing energy absorption during tearing, while CNC's high crystallinity further restricts flexibility. Similar trends have been reported in the literature, where excessive nanocellulose addition has been shown to lead to brittle paper structures. To optimize tear resistance while maintaining strength, future studies could explore adjusted CNF concentrations, hybrid formulations, or modified processing techniques that balance bonding and flexibility.

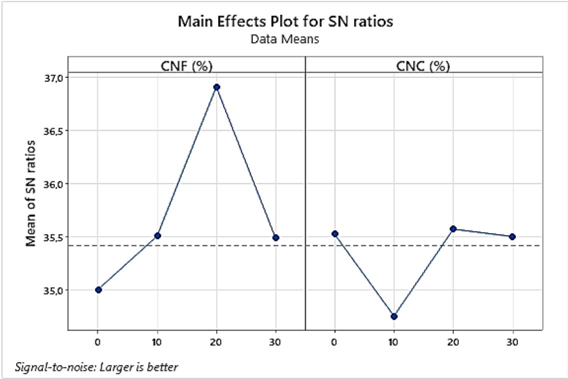
Table 2 presents the results of the signal-to-noise ratio (S/N ratio) analysis for tensile strength, burst strength, and tear resistance, conducted to determine the optimal levels of CNF and CNC nanocellulose for enhancing mechanical properties. This analysis was performed as part of the Taguchi method, which is designed to helps identify the most effective factors and their levels for improving paper strength while minimizing variability.

Table 2. Signal-to-Noise ratios for mechanical properties of recycled paper with CNF and CNC nanocellulose at different levels

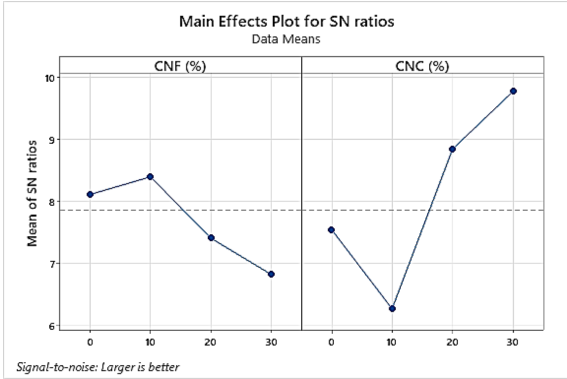
	Tensile Strength (N)		Burst Strength (kPa)		Tear Resistance (gf)	
Level	CNF (%)	CNC (%)	CNF (%)	CNC (%)	CNF (%)	CNC (%)
1	35.00	35.52	8.109	7.545	19.76	20.09
2	35.51	34.75	8.392	6.268	19.52	19.52
3	36.91	35.57	7.406	8.839	20.83	20.00
4	35.49	35.50	6.821	9.768	20.00	19.52
Delta	1.91	0.82	1.571	3.501	1.31	0.57
Rank	1	2	2	1	1	2

The Taguchi analysis of tensile strength, burst strength, and tear resistance demonstrates that CNF nanocellulose consistently outperforms CNC in enhancing mechanical properties. For tensile strength, CNF shows the highest improvement at level 3 (36.91 N), while CNC's impact is more limited across all levels, peaking at 35.57 N at level 3. In burst strength, CNC exhibits a stronger effect at level 4 (9.768 kPa), whereas CNF peaks at level 2 (8.392 kPa), indicating that higher concentrations of CNC are more effective for burst strength. For tear resistance, both CNF and CNC perform similarly at level 3, but CNF slightly outperforms CNC with a maximum tear resistance of 20.83 gf at the same level. The ranking based on delta values confirms that CNF has a greater overall influence on tensile and tear resistance, while CNC has a stronger effect on burst strength.

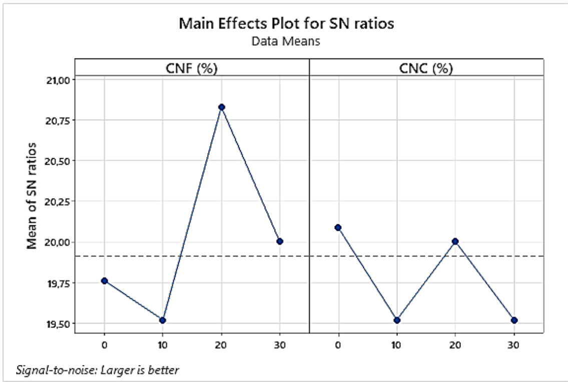
Figure 1 presents the main effects plots for the Signal-to-Noise ratios (S/N ratio) derived from the Taguchi analysis, conducted to assess the influence of CNF and CNC nanocelluloses on tensile strength, burst strength, and tear resistance across different concentration levels. This analysis was performed to determine the optimal levels of nanocellulose for maximizing paper strength properties while minimizing variability.



Tensile Strength (N)



Burst Strength (kPa)



Tear Resistance (gf)

Figure 1. Mechanical properties: Main effects plots for Signal-to-Noise ratios of CNF and CNC nanocellulose at different concentration levels

The Taguchi analysis on tensile, burst, and tear strengths revealed distinct contributions from CNF and CNC nanocelluloses to paper strength. CNF showed a more significant effect, particularly at the 20% concentration level, where it achieved the highest improvements in tensile strength, as indicated by optimal signal-to-noise (S/N) ratios. CNC had a more limited impact, with moderate improvements at 20%, suggesting its effectiveness at lower levels. For burst strength, CNF again performed best at 20%, while CNC showed a stronger effect at 30%. The optimal combination for burst strength was found to be 20% CNF and 30% CNC. In tear resistance, CNF at 20% provided the highest improvement, while CNC's contribution was less significant. CNF outperformed CNC across all three strength parameters, particularly at 20%. While CNC's effect was more modest, concentrations between 20% and 30% still showed improvements in tensile and burst strength. The ideal combination to optimize strength properties is 20% CNF with 20-30% CNC.

The correlation analysis reveals that CNF has a moderate positive effect on both tensile strength (r = 0.427) and tear resistance (r = 0.413), indicating its role in enhancing fiber bonding and overall toughness. In contrast, CNC shows little impact on tensile strength (r = -0.028) and a negative correlation tear resistance (r = -0.336), but has a stronger positive correlation with burst strength (r = 0.511), suggesting its ability to improve the paper's out-of-plane resistance

Optical Properties

The optical properties, including whiteness, brightness, and yellowness, were evaluated, and the results are summarized in Table 3. The optical performance of recycled newspaper, as influenced by CNF and CNC nanocelluloses, shows that both types of nanocellulose types affect these properties to varying degrees, exhibiting several notable trends.

Table 3. Effect of CNF and CNC nanocellulose on the optical properties of recycled newspaper at different concentrations

Nanocellulose	Whiteness (ISO%)		Brightness (ISO%)		Yellowness (E313)	
Cons. (%)	CNF	CNC	CNF	CNC	CNF	CNC
0	39.54 (0.35)		35.46 (0.40)		14.03 (0.31)	
10	38.26 (0.23)	37.76 (0.03)	34.25 (0.03)	33.87 (0.12)	13.66 (0.23)	14.00 (0.35)
20	36.89 (0.69)	36.86 (0.12)	33.25 (0.77)	33.10 (0.19)	13.43 (0.53)	13.62 (0.37)
30	36.56 (0.76)	36.29 (0.42)	32.94 (0.74)	32.58 (0.38)	13.50 (0.36)	13.68 (0.05)

*Values in parentheses represent standard deviations

For whiteness, the results in Table 3 show that the addition of both CNF and CNC nanocelluloses led to a reduction in the ISO whiteness values compared to the control sample. As the concentration of nanocellulose increased, the whiteness values decreased progressively. This effect can be attributed to the fact that nanocellulose, particularly at higher concentrations, tends to fill the paper's voids and reduce light scattering, which contributes to high whiteness in paper. However, the degree of whiteness reduction differs between CNF and CNC, indicating that their influence on optical properties is governed by distinct underlying mechanisms.

CNF exhibited a more pronounced decrease in whiteness compared to CNC, which can be explained by its gel-like nature and high aspect ratio, leading to the formation of a denser and more compact fiber network that further limits light scattering. The entangled structure of CNF also increases fiber bonding, which may contribute to a more uniform but less reflective paper surface. In contrast, CNC, due to its rigid, crystalline nature and lower tendency to form an extensive fiber network, results in a relatively smaller reduction in whiteness. CNC particles, being discrete and rod-like, do not contribute to network densification to the same extent as CNF, allowing for slightly better light diffusion. Previous studies (He et al., 2016; Tajik et al., 2018; Toivonen et al., 2018) have also reported that while CNFs improve mechanical properties, their compact structure leads to reduced whiteness, whereas CNC's impact on whiteness is less severe but still noticeable at higher dosages. These findings highlight the importance of optimizing nanocellulose concentration to balance mechanical improvements with acceptable optical performance, particularly for applications where brightness and whiteness are critical.

In terms of brightness, both CNF and CNC resulted in decreased brightness values as concentrations increased, with CNC showing a slightly better performance at the 30% concentration level. Brightness is closely linked to the reflectance of light, and the findings align with those of Sun et al., (2018), Toivonen et al., (2018) and Xu et al., (2013), who observed that cellulose nanocrystals (CNCs) generally exhibit higher transparency than CNFs, potentially allowing for a slight improvement in brightness retention. However, the reduction in brightness at higher nanocellulose levels suggests that while CNC may slightly outperform CNF in this aspect, both types of nanocellulose can reduce brightness due to their impact on the paper's surface uniformity and light reflection.

When examining yellowness, both nanocelluloses demonstrated a reduction in yellowness index, especially at the 20% CNF concentration, which exhibited the lowest yellowness value. This can be seen as a positive outcome, as lower yellowness is desirable for maintaining the paper's aesthetic quality. This finding aligns with studies by Campano et al., (2018), Li et al., (2021) and Sanchez-Salvador et al., (2020), which reported that nanocellulose could help minimize the degradation effects (such as yellowing) typically associated with recycled paper by improving the uniformity of the fiber matrix. The slightly higher yellowness observed with CNC at the 30% concentration level might be due to the greater crystalline structure of CNCs, which can result in less effective light diffusion and a more yellow appearance under certain conditions.

The analysis reveals that both CNF and CNC nanocelluloses significantly influence the optical properties of recycled paper, with CNF at a 20% concentration demonstrating the most balanced effect by effectively reducing yellowness while maintaining adequate levels of whiteness and brightness. These observations are consistent with established literature, which suggests that cellulose nanofibrils (CNFs) excel in applications where enhanced fiber bonding and minimized optical degradation are essential. In comparison, cellulose nanocrystals (CNCs) show a modest advantage in preserving brightness but exhibit less efficacy in controlling yellowness. This may be attributed to their highly crystalline structure, which improves transparency but results in reduced optical uniformity.

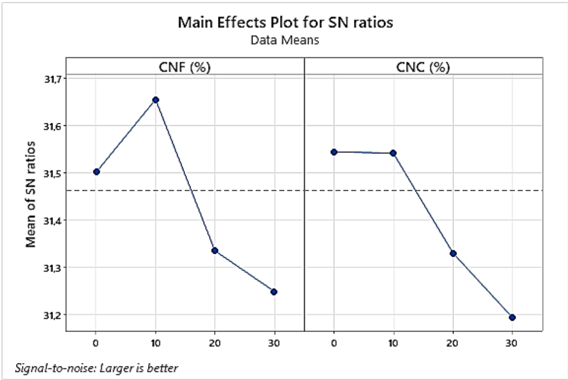
Table 4 presents the results of the signal-to-noise ratio (S/N ratio) analysis for the optical properties, including whiteness, brightness, and yellowness, of recycled paper containing varying concentrations of CNF and CNC nanocellulose.

Table 4. Signal-to-Noise ratios for optical properties of recycled paper with CNF and CNC nanocellulose at different levels

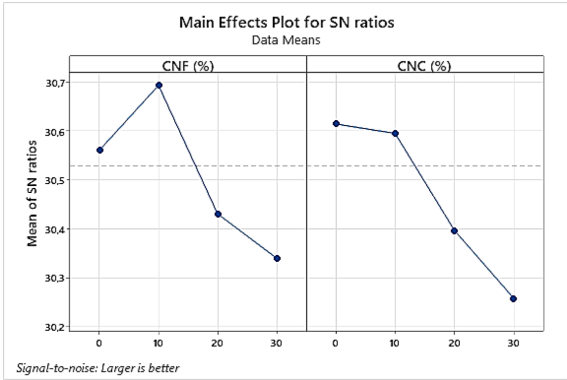
	Whiteness (ISO%)		Brightness (ISO%)		Yellowness (E313)	
Level	CNF (%)	CNC (%)	CNF (%)	CNC (%)	CNF (%)	CNC (%)
1	31.50	31.54	30.56	30.61	-22.82	-22.71
2	31.65	31.54	30.69	30.59	-22.71	-22.92
3	31.33	31.33	30.43	30.40	-22.57	-22.69
4	31.25	31.19	30.34	30.26	-22.61	-22.72
Delta	0.41	0.35	0.35	0.36	0.25	0.24
Rank	1	2	2	1	1	2

The analysis of the optical properties reveals that CNF nanocellulose outperforms CNC in enhancing both whiteness and reducing yellowness, with the highest S/N ratio for whiteness observed at level 2 (31.65) and the lowest yellowness at level 3 (-22.57). This suggests that CNF's fine fibrillar structure improves light scattering and minimizes yellowing, consistent with findings by Aydemir et al., (2021), Havenko et al., (2020), Lourenço et al., (2020a), Radić Seleš et al., (2020) and Toivonen et al., (2018). While CNC demonstrates comparable but slightly lower effectiveness in whiteness, its crystalline structure limits light scattering, and it shows a stronger effect on maintaining brightness, with the highest S/N ratio for brightness observed at level 2 (30.69). Overall, CNF has a greater impact on improving whiteness and reducing yellowing, while CNC performs moderately in maintaining brightness, aligning with previous studies on CNFs and CNCs.

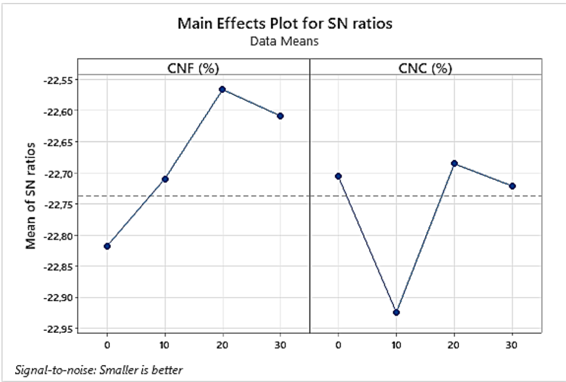
Figure 2 presents the main effects plots for the Signal-to-Noise ratios (S/N ratio) derived from the Taguchi analysis, conducted to evaluate the influence of CNF and CNC nanocelluloses on whiteness, brightness, and yellowness across different concentration levels.



Whiteness (ISO%)



Brightness (ISO%)



Yellowness (E313)

Figure 2. Optical properties: Main effects plots for Signal-to-Noise ratios of CNF and CNC nanocellulose at different concentration levels

The Taguchi analysis on the optical properties, including whiteness, brightness, and yellowness, demonstrated varying effects of CNF and CNC nanocelluloses at different concentrations. CNF exhibited a more significant influence on whiteness, particularly at 20% concentration level, where the highest S/N ratios were observed, indicating that CNF improves whiteness more effectively at this level. In contrast, CNC showed slightly lower but comparable improvements in whiteness, with the best performance at 10% concentration. For brightness, CNF and CNC demonstrated relatively similar trends, although CNC achieved the highest S/N ratios at 20%, suggesting its effectiveness in maintaining brightness. In terms of yellowness, CNF showed a stronger effect in reducing yellowness, especially at 20%, where the lowest values were recorded, whereas CNC had a more modest impact. Overall, CNF at 20% concentration provided the best balance across all optical properties, while CNC showed moderate effectiveness in maintaining brightness, particularly at lower concentrations.

The correlation analysis shows that CNF has a moderate negative impact on whiteness (r = -0.321) and brightness (r = -0.274), while also significantly reducing yellowness (r = -0.416), indicating its role in improving the visual quality by decreasing yellowness as its concentration increases. In contrast, CNC shows a stronger negative correlation with whiteness (r = -0.471) and brightness (r = -0.487), but exerts little effect on yellowness (r = 0.023). This suggests that while CNC tends to reduce optical clarity, CNF is more effective in controlling yellowness, thus presenting a trade-off between different optical properties based on the nanocellulose type.

Surface Properties

Surface roughness, a critical parameter for printability, was analyzed to evaluate the effects of nanocellulose addition. Table 5 presents the surface roughness values (Ra, Rq, Rz) for recycled paper samples treated with varying concentrations (0%, 10%, 20%, and 30%) of CNF and CNC nanocelluloses.

Table 5. Surface properties (Ra, Rq, Rz) of recycled paper with different nanocellulose types and concentrations

Nanocellulose	Ra		Rq		Rz	
Cons. (%)	CNF	CNC	CNF	CNC	CNF	CNC
0	1.855 (0.04)		2.402 (0.14)		11.952 (0.58)	
10	1.864 (0.17)	2.177 (0.34)	2.425 (0.20)	2.701 (0.44)	12.715 (0.78)	13.584 (2.67)
20	2.028 (0.10)	2.113 (0.19)	2.591 (0.07)	2.642 (0.27)	12.686 (0.60)	12.517 (1.34)
30	1.691 (0.20)	1.780 (0.03)	2.196 (0.22)	2.379 (0.14)	12.198 (1.47)	13.221 (1.34)

*Values in parentheses represent standard deviations

The surface roughness parameters (Ra, Rq, and Rz) highlight the influence of CNF and CNC nanocellulose types and concentrations on recycled paper surfaces. In this study, CNF exhibited a reduction in surface roughness, particularly at the 30% concentration, where Ra and Rz values were lower than those for CNC. This suggests that CNF, at higher concentrations, promotes smoother surfaces by enhancing fiber bonding and distribution without significant agglomeration. These findings are in line with previous studies, such as that of Hu et al., (2021), which demonstrated that the long fibrillar structure of CNF helps bridge gaps between fibers, thereby reducing surface roughness (Ataeefard, 2014; Bai et al., 2019; Havenko et al., 2020; Lourenço et al., 2020b). CNC, while effective in other applications, showed higher roughness values in this context, indicating that it may not be as suitable for applications where surface smoothness is critical. This observation is consistent with the works of Aydemir et al., (2021), Li et al., (2021), Perdoch et al., (2022), Radić Seleš et al., (2020) and Sanchez-Salvador et al., (2020), who noted that CNC's rigid and crystalline nature can lead to a less uniform surface, especially at higher concentrations.

The Rz values further support this trend, with CNF displaying more stable and lower extreme surface variations compared to CNC. At higher concentrations (30%), CNC resulted in greater peaks and valleys on the surface, as evidenced by its higher Rz value. These results align with the study by Lourenço et al., (2019), which found that CNC can create a rougher surface due to its inability to conform to the fibrous structure as readily as CNF. In contrast, CNF's flexibility and ability to entangle with paper fibers allow for a more uniform and smooth surface finish, a characteristic emphasized by Guan et al., (2019) in their analysis of nanocellulose applications in paper coatings.

In line with these observations, CNF appears to be the more appropriate choice when minimizing surface roughness is the primary objective, particularly for applications like packaging or printing, where smooth surfaces are essential. CNC, while valuable in reinforcing mechanical properties, may not perform as well in reducing roughness, especially at higher concentrations. These results emphasize the need to carefully select nanocellulose both the types and concentrations based on the specific performance requirements of the end product. The Pearson correlation analysis reveals a weak negative correlation between CNF concentration and Ra (-0.325), suggesting that increasing CNF concentration slightly reduces surface roughness, which supports findings by Perdoch et al., (2022), who observed similar reductions in roughness with increased CNF content.

The Taguchi analysis confirmed that 30% CNF yielded the lowest S/N ratios for surface roughness, indicating optimal printability at this concentration. The Ra values were reduced by 20% at 30% CNF compared to the control, showing that CNF effectively fills micro-voids and improves surface uniformity, an essential factor for applications requiring high-quality printing. This observation aligns with the work of Ozcan et al., (2021), who found that CNF's ability to reduce surface roughness makes it an ideal additive for improving printability in coated papers. CNC, on the other hand, exhibited a weaker negative correlation with Ra values ($r = -0.325$, $p > 0.05$), and the Taguchi analysis indicated that its effect on surface roughness was minimal across all concentration levels. The S/N ratios did not show significant improvement with increasing CNC concentrations, reinforcing the conclusion that CNF is the superior additive for improving surface smoothness and printability.

CONCLUSIONS

This study systematically evaluated the effects of cellulose nanofibrils (CNF) and cellulose nanocrystals (CNC) on the mechanical, optical, and surface properties of recycled newspaper, identifying the most effective concentrations for enhancing specific performance metrics. The findings highlight that CNF at a 20% concentration provides the most balanced improvement by significantly enhancing tensile strength, tear resistance, brightness, and reducing yellowness, making it a promising additive for applications requiring both durability and visual quality. Additionally, CNF substantially improved surface smoothness, which is particularly beneficial for printing and high-quality packaging applications. In contrast, CNC was most effective in improving burst strength at a 10% concentration but exhibited adverse effects on whiteness and brightness, limiting its suitability for applications where optical and surface quality are critical. While both nanocellulose types improved mechanical properties, CNF demonstrated a greater overall impact, positioning it as the more versatile and industrially relevant option for enhancing recycled newspaper quality.

From an industrial perspective, the economic feasibility of incorporating nanocellulose at high concentrations is a critical factor, as large-scale implementation requires a balance between performance gains and cost-effectiveness. While the study examined concentrations up to 30%, a more practical range for industrial implementation would be 5–15%, particularly for CNF, which provided notable improvements even at lower dosages. Furthermore, nanocellulose retention within the fiber network plays a crucial role in maximizing its reinforcing potential, and future studies should explore strategies such as optimized pulp chemistry, modifications to retention aids, or hybrid reinforcement approaches to enhance efficiency. Overall, CNF emerged as the superior nanocellulose type for improving mechanical, optical, and surface properties, while CNC, despite its benefits for burst strength, is better suited for applications where mechanical resilience is the primary concern. These findings provide valuable insights into the scalable and cost-effective use of nanocellulose in recycled paper production, contributing to the advancement of sustainable and high-performance paper materials.

AUTHOR CONTRIBUTIONS

Mustafa Çiçekler: Conceptualization, Methodology, Data curation, Formal analysis, Writing - original draft. **Ahmet Tutuş:** Conceptualization, Methodology, Data curation, Formal analysis, Writing - original draft.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

ETHICS COMMITTEE APPROVAL

This study does not require any ethics committee approval.

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