Printability of variative nanocellulose derived papers

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Research Article

Keywords: Cellulose nanofiber (CNF), Cellulose Nanofiber-Oxidized (CNF-OX), Nanocellulose, Paper coating, Printability

Posted Date: February 24th, 2021

DOI: https://doi.org/10.21203/rs.3.rs-233734/v1

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Version of Record: A version of this preprint was published at Cellulose on April 7th, 2021. See the published version at https://doi.org/10.1007/s10570-021-03861-3.
Abstract

The printability properties of the paper can be increased by some processes applied to the surface. The use of non-recyclable materials derived from petroleum is decreasing day by day, and the demand for recyclable materials obtained from renewable sources is increasing. These materials include cellulose derivatives, starch types and polyvinyl alcohol. The materials ratios, sizes, physical and chemical properties of these materials used in the processes applied to the paper and the content of the paper will affect the strength of the paper as well as change the surface properties and significantly affect the printability.

In this study, fluting and core board papers coated with different amounts of cellulose nanofiber/cellulose nanofiber-oxidized (CNF/CNF-OX) were produced. Surface properties, contact angle, surface energy, color and gloss of the produced papers were measured by optical microscope, goniometer, spectrophotometer and glossmeter. The papers were printed with the IGT C1 offset printability tester. The color and gloss properties of the printed papers were measured. As a result, in terms of printability, it was determined that CNF/CNF-OX coated papers have smoother surfaces and give better results in terms of both gloss properties and printability.

Introduction

In our digital age, despite the increase in communication channels and the increasing interest in digital, there is no decrease in paper production and consumption and the demand for printed materials. Although printing needs seem to decrease in some business areas in the printing industry, the changing community needs and product diversity have brought many opportunities, especially in the packaging industry. Generally, paper and cardboard production is carried out in two steps. These are chemical or mechanical pulp of woody or nonwoody materials to obtain cellulose-rich fibers and the other is the formation of paper sheets from fibers and paper surface treatment. Today, paper-based products are widely used for a variety of applications in packaging, printing, writing, home use and more (Shen et al. 2014). Paper-based packaging materials are expected to protect food, beverage, health and cosmetic products against physical, chemical and microbiological deterioration. Today, it is common to use glass, aluminum and plastic derivative materials other than paper as packaging material. These materials have higher strength and barrier properties compared to paper-based materials. But today, sustainability and accordingly biocompatibility is very important, so the use of paper-based materials will increase even more (Nair et al. 2014; Bayer et al. 2011; Spence et al. 2011). In recent years, worldwide demand for packaging materials from paper and cardboard has been increasing as an alternative to plastic materials that have a long biodegradation time (McCracken and Sadeghian 2018).

In addition to pulp fibers, various chemicals are also used in paper production. These chemicals are used entire in paper production prior to sheet forming or through paper surface treatment. The basic functions of these chemicals in paper production; changing the properties of the fibers, improving the strength of the paper, ensuring the adhesion of the fibers, reducing the penetration of liquids into paper, reducing the
cost, improving the optical properties of paper and adjusting the color of the paper. In this way, better performance can be obtained from paper with its improved properties and better printability properties are obtained. In general, mineral-based chemicals and bio-based chemicals are widely preferred (Arbatan et al. 2012).

The search for renewable, recyclable and biodegradable alternatives has recently increased the interest in cellulose-based materials. Although there is a rapid growth in new green materials and biopolymers, cellulose is the most important renewable material widely used in paper and cardboard, packaging applications today (Hult et al. 2010). Cellulose is the most abundant polymer in nature, and paper-based packaging is light, low-cost, and most importantly sustainable. There are studies in the literature showing that various types of cellulose nanofibers produced from cellulose fibers can be used as environmentally friendly alternatives (Charani et al. 2013; Petroudy et al. 2014; Afra et al. 2014). Another study revealed that when cellulose nanofiber (CNF) is used as a filler, it improves the properties of various composite or paper types (Afra et al. 2016; Mashkour et al. 2015). The use of CNF as a coating material significantly reduces the air permeability of the paper (Aulin et al. 2010), the air, oxygen and water vapor permeability of the paper is significantly reduced after coating (Hult et al. 2010) and it has also been revealed in previous studies that CNF increases the surface and barrier properties of coated papers (Ridgway and Gane 2012).

The use of carboxymethyl cellulose (CMC) as sizing material is also common. It increases the hydrophobicity and surface strength of the paper (Holik 2006). It reduces paper porosity and provides better printability (Dulany et al. 2011; Shen and Qian 2012). In addition, cellulose-based polymers such as carboxymethyl cellulose, hydroxyethyl cellulose, and methyl cellulose function as density enhancers and binders in pigmented coating formulations. These biopolymers can also function as rheology regulators or water-retaining agents in pigmented coatings (Tang et al. 2013; Klass 2011).

It is known that sizing and coating processes applied to the surface of paper and cardboard have an effect on the gloss of the paper. Cellulose-based biopolymers also function as a good carrier for optical brightening or whitening agents used in papers (Holik 2006).

Although the use of nanocellulose in the paper industry is not very old, it is a subject that has been studied a lot in recent years. In general, nanocellulose, mainly nanofibrillated cellulose (CNF), produced from pulp fibers derived from lignocellulose by mechanical breakdown or chemical hydrolysis, is produced by mechanical process and nanocrystalline cellulose (NCC), produced by the chemical hydrolysis process are common in two types (Aspler et al. 2013; Bardet et al. 2013; Chen et al. 2013; Dimic-Misic et al. 2013; Penttilä et al. 2013; Ridgway and Gane 2013).

In studies where nanofibrillated cellulose was applied to the paper surface with pigment (paper coating) or without pigment (sizing), it was seen that these surface treatments improve the surface strength, barrier properties and printability properties of the paper (Aulin et al. 2010; Richmond et al. 2012; Syverud and Stenius 2009). The improvement in surface strength to the high hydrogen bonding capacity of the large amount of hydroxyl groups. In addition, nano-sized cellulose filling the gaps on the porous surface
of the paper positively affects the barrier properties. Moreover, the surface to be printed is smoother, the filling of the pores means reduced ink penetration and therefore better printability with less ink (Skočaj 2019).

The use of papers with improved barrier properties (due to its resistance to moisture, air, oil, odor, etc.) in the packaging of dry or wet food, cosmetics and pharmaceutical products is important. Due to the barrier properties of nanocellulose in paper-based products, it can be expected to be used much more widely in the future as it is biocompatible compared to other barrier enhancing materials (Tang et al. 2014; Lavoine et al. 2012; Mertaniemi et al. 2012; Aulin and Ström 2013; Martins et al. 2012).

Cellulose is one of the bio-based materials that are widely used in the paper industry and various other industries. Cellulose is also the main material of paper production. In addition to being the main material, cellulose-based polymers are used as additives for both wet and dry surface applications in the paper industry.

Cellulose meets the European Union regulations on materials that come into contact with foodstuffs. It also complies with American food and drug administration legislation on ingredients for food contact materials. Cellulose-based barrier coatings have the potential to provide environmental benefits through recycling or composting as final disposal. In addition, a microfibrillated cellulose coated paper can be burned with high energy recovery and without any health impact (Gatti 2005).

The paper or other packaging materials to be printed on must have better printability properties in order to meet consumer expectations. Therefore, some surface treatments are used to give paper and cardboard better printability properties. These processes are generally sizing, coating and calendering (Ozcan et al. 2020a). With surface treatments, the paper becomes more durable in terms of physical properties, while also improving its optical and printability properties. With the sizing process, substances such as starch types, cellulose types and polyvinyl alcohol are applied on the paper to cover the roughness on the surface; Thus, the ink to be printed on the paper is prevented from penetrating into the paper. In this way, higher printing gloss is obtained with less ink. Coating formulation usually consists of inorganic pigments such as kaolin-clay, calcium carbonate and titanium dioxide, binders, dispersants and some other additives (Morsy et al. 2016; Ozcan et al. 2019).

The aim of this study is to examine the printability properties of different CNF/CNF-OX coated fluting and core board papers and to examine their usability not only without printing but also in printed packaging production.

**Materials And Methods**

**Materials**

Freshly cut wheat straw (*Triticum aestivum* L.) was used as the raw material to produce nanofibrillated cellulose. Recycled fibers were obtained from KMK Paper Co. (Kütahya, Turkey).
Pulping and bleaching chemicals (sodium hydroxide, sodium borohydride, sodium chlorite, sodium acetate, acetic acid, formic acid) and oxidation radical (TEMPO) were provided from Sigma-Aldrich (Taufkirchen, Germany). The hemicellulase enzyme (Pulpzyme HC 2500) was obtained from Novozymes ( Bagsværld, Denmark). All chemicals were used without any further purification.

Toyolife LF - 1600 process magenta commercial offset printing ink was obtained from TOYO ink. Co. (Manisa, Turkey).

Methods

Pulping and Bleaching

Soda-NaBH$_4$ cooking procedure was selected as pulping methodology and pulping and bleaching sequences were conducted as reported by Tozluoglu et al. (2021).

Pretreatments

Bleached soda-NaNB$_4$ fibers were enzymatically and chemically treated before nanofibrillation process. For enzymatic pretreatment, 50 grams of oven dried bleached soda-NaBH$_4$ pulp were hydrolyzed using hemicellulase (Pulpzyme HC 2500) enzyme at 2% solid loading in 2.5 L of phosphate buffer at pH 7. Hemicellulase enzyme was prepared at 25 AXU/g and phosphate buffer was prepared using 11-mM KH$_2$PO$_4$ and 9-mM Na$_2$HPO$_4$. Enzymatic hydrolysis was performed in an incubator (ES-20, Biosan Lab., Riga, Latvia) at 50 °C for 2 h. During hydrolysis, samples were mixed every 30 min and, at the end of 2 h, washed with deionized water. Subsequently, samples were retained in a water bath at 80 °C for 30 min to halt the enzymatic activity. Afterwards, the pulps were washed with deionized water for a second time. For chemical pretreatment, 5 grams of oven dried bleached soda-NaBH$_4$ pulp fiber were oxidized using TEMPO at 1% solid loading in 500 mL of sodium phosphate buffer (0.05 M) containing 250 mg of NaBr and 25 mg of TEMPO at pH 7. Afterwards, sodium chlorite (80%, 1.13 g, 10 mM) and 2 M sodium hypochlorite (0.5 mL, 1.0 mM) were added to the buffer. Oxidization was conducted in a GFL 3033 shaking incubator (GFL Lab., Burgwedel, Germany) at 150 rpm at 60 °C for 72 h. To terminate the reaction, 100 mL of ethanol was added to the suspension, the mixture was filtered and the fibers were washed with deionized water.

CNF/CNF-OX Production

CNF/CNF-OX productions were executed via high-pressure fluidizer (M-110Y Microfluidizer, Microfluidics Corp., Massachusetts, USA) at 2 wt% concentration. For the CNF/CNF-OX productions, enzymatically/chemically pretreated pulp fibers were passed once through a Z-shaped chamber with a diameter of 200 µm at 14000 psi and subsequently passed five times through a chamber with a diameter of 100 µm at 24000 psi.
The material obtained after the homogenization process following the enzymatic pretreatment was coded as CNF and the material obtained after the homogenization process following the oxidation pretreatment was coded as CNF-OX in this study.

**Handsheet Preparation**

Recycled fibers were used as primary raw material for core board and fluting paper production in this study. For the production of fluting papers, mixed old corrugated cardboard fibers (OCC) and for the production of core board papers, old newspaper and old magazine paper fibers (ONP and OMP) were utilized as main fiber sources. Handsheets were prepared using a Rapid Kothen handsheet former (PTI, Vorchdorf, Austria) according to ISO 5269-2 (2004) and then conditioned according to TAPPI T402 sp-13 (2013). Size press coating application was achieved as 40 t/kg starch loading for both core board and fluting paper sheets and produced enzymatically and chemically pretreated CNF/CNF-OX applied at 2% and 4% concentrations to fabricated paper sheets with starch.

The grammage values of the used papers, the types and rates of nano cellulose used are given in Table 1. In addition, L*a*b* color values of all papers were measured and color differences were examined with the control sample. ΔL values were also measured due to the significant change in the L* value in the measurements. Measurement results are given in Table 2.

| Sample No | Grammage (g/m²) | CNF | CNF-OX |
|-----------|-----------------|-----|--------|
| F0        | 119             | -   | -      |
| F1        | 121             | -   | 2%     |
| F2        | 121             | -   | 4%     |
| F3        | 122             | 2%  | -      |
| F4        | 120             | 4%  | -      |
| C0        | 177             | -   | -      |
| C1        | 172             | 2%  | -      |
| C2        | 174             | 4%  | -      |
| C3        | 176             | -   | 2%     |
| C4        | 175             | -   | 4%     |
Color Properties

The color properties of the papers and also printed papers were determined using CIEL*a*b* color values by using X-Rite eXact spectrophotometer according to ISO 13655:2017 standard. The measurement conditions of the spectrophotometer were determined as polarization filter with 0/45° geometry with 2° observer angle with D50 light source in the range of 400-700 nm. The difference between the colors of the different prints were calculated according to the CIE ΔE 2000 color-difference formula ISO 11664-6:2014. Calculations were made by taking the average of five measurements. ΔL*, Δa*, Δb*: Difference in L*, a*, and b* values between specimen color and target color. Lightness is represented by the L* axis which ranges from White to Black. The red area is connected to the green by the a* axis, while the b* axis runs from yellow to blue.

\[
\Delta E_{00} = \sqrt{\left(\frac{\Delta L'}{k_{LsL}}\right)^2 + \left(\frac{\Delta a'}{k_{CSC}}\right)^2 + \left(\frac{\Delta b'}{k_{HSH}}\right)^2 + R_T \frac{\Delta c'}{k_{CSC}} \frac{\Delta h'}{k_{HSH}}} \tag{1}
\]

Gloss

The gloss measurements of coated papers were carried out with BYK Gardner GmbH micro gloss 75° geometry in accordance with ISO 8254-1:2009, gloss measurement results of papers are shown in Figure 1, and the gloss measurements of prints with BYK Gardner GmbH micro Tri-gloss 60° geometry in accordance with ISO 2813:2014. Gloss measurement results of magenta printed papers are shown in Figure 4.

Contact Angle and Surface Energy

The contact angle and total surface energy measurements of the papers were performed by PocketGoniometer PGX+ in accordance with ASTM D5946 standard. Total surface energy values are shown in Table 3 and the contact angle measurement results are shown in Table 4.

| Sample No | L*  | a*  | b*  | ΔE00 | ΔL  |
|-----------|-----|-----|-----|------|-----|
| F0        | 63.25 | 6.36 | 15.01 |      |     |
| F1        | 61.86 | 6.01 | 14.77 | 1.24 | 1.39|
| F2        | 61.45 | 6.26 | 15.53 | 1.59 | 1.80|
| F3        | 61.12 | 6.15 | 14.87 | 1.83 | 2.13|
| F4        | 61.05 | 6.21 | 15.38 | 1.91 | 2.20|
| C0        | 65.85 | 6.42 | 15.20 |      |     |
| C1        | 61.39 | 7.28 | 17.24 | 3.95 | 4.46|
| C2        | 61.84 | 7.31 | 17.36 | 3.61 | 4.01|
| C3        | 61.79 | 6.77 | 16.72 | 3.51 | 4.06|
| C4        | 63.01 | 6.86 | 16.42 | 2.47 | 2.84|
Characterization

Surface images of control papers and CNF/CNF-OX coated fluting and core board papers were taken with a Leica S8 APO microscope. The images obtained are given in Figure 2 and Figure 3.

Printing

Control papers, fluting papers and core board papers were printed with Toyolife LF - 1600 process magenta commercial offset printing ink using an IGT C1 offset printability test device under 400 N/m² pressure printing conditions. After printing, color properties and gloss values of printed papers measured according to standards.

Results And Conclusions

L*a*b* color values of CNF/CNF-OX coated fluting and core board papers are given in Table 2. In the table, after CNF/CNF-OX coated on the fluting papers, the change in ΔE₀₀ in the paper color according to the control sample (F0) was measured as respectively F1-1.24, F2-1.59, F3-1.83 and F4-1.91. After CNF/CNF-OX coated on the core board papers, the change in ΔE₀₀ in the paper color according to the control sample (C0) was measured as respectively C1-3.95, C2-3.61, C3-3.51 and C4-2.47. According to ISO 12647-2:2013 graphic technology standard, if there is no special requirement, the color difference is the acceptable limit up to ΔE₀₀ 5. According to these results, the color difference in the fluting papers is too much to be distinguished by the human eye. The color difference in core board papers can be noticed by the eye, but is still within the acceptable range. The most important point in L*a*b* color measurement of CNF/CNF-OX coated papers is the decrease in the L* value. According to L* value results, the CNF/CNF-OX coated papers are darker than the control sample. When the fluting papers were examined, as the amount of CNF/CNF-OX increased, the difference in the L* value increased, while the L* difference in the core board papers decreased. The reason for this increase in fluting papers is the unique color of CNF/CNF-OX. It caused an extra change in the color of the paper. However, since the color of core board papers is darker than CNF/CNF-OX’s own color, the increase of CNF/CNF-OX caused the color to lighten and the L* difference decreased.

Figure 1 shows the gloss values of control samples and CNF/CNF-OX coated fluting and core board papers. When Figure 1 is examined, gloss values in CNF/CNF-OX coated fluting and core board papers increased compared to control samples. The reason for this is that nano-sized cellulose fills the gaps on the porous surface of the paper and thus creates a smoother surface and the obtained results are consistent with the literature (Sharma et al. 2020). When paper types are compared, while core board paper has lower gloss value than fluting paper, the increase in gloss after CNF/CNF-OX coating is higher than fluting papers.
When Table 3 and Table 4 are examined, it is seen that the surface energy decreases and the contact angle increases with the CNF/CNF-OX coating on both the fluting papers and the core board papers. The reason for this is that the nano-sized cellulose additive has less hydrogen bonding with water than in paper, and therefore wetting is not good. The long cellulose fibers in the paper (control papers / without CNF/CNF-OX) make more hydrogen bonding with water, which increases the adhesion strength between water and cellulose and reduces the surface contact angle, but in shortened nanocelluloses, the number of hydrogen bonds is lower, so the adhesion forces are reduced, the contact angle has been increased and the surface energy has decreased.

**Table 3.** Total surface energy values according to ASTM D5946 method

| Fluting papers | Total surface energy (mJ/m²) | Core board papers | Total surface energy (mJ/m²) |
|----------------|-------------------------------|-------------------|-------------------------------|
| F0             | 43.8                          | C0                | 42.4                          |
| F1             | 38.8                          | C1                | 41.0                          |
| F2             | 39.3                          | C2                | 40.6                          |
| F3             | 40.3                          | C3                | 41.9                          |
| F4             | 43.1                          | C4                | 41.7                          |
When the contact angles are examined in Table 4, the contact angle increased in 2% CNF-OX coated fluting and core board papers (F1 and C1) and 4% in CNF-OX coated fluting and core board papers (F2 and C2). But this increase in fluting papers has increased 2 times compared to core board papers. This is because core board papers have porous structures and therefore the added CNF-OX is less on the surface. For this reason, its effect is less. In the same table, the contact angle of 2% CNF coated fluting paper (F3) and 4% CNF coated fluting paper (F4) remained close to the control sample, almost unchanged. On the other hand, in 2% CNF coated core board (C3) and 4% CNF coated core board (C4) the contact angle decreased, thus increasing the hydrophilicity of the paper.
When the Table 4 is examined, it is more appropriate to print with water-based ink on hydrophilic papers, and it is more appropriate to print with oil-based offset inks on papers with increased contact angle.

When microscopic images are examined in Figure 2 and Figure 3, it is understood that the surfaces of CNF/CNF-OX coated papers are smoother compared to the control samples. In both types of paper, nano-sized celluloses penetrated the pores of the papers and filled them, thus smoother and more uniform surfaces were obtained. Likewise, it is seen that the surfaces of 4% CNF/CNF-OX coated papers have a smoother surface than 2% CNF/CNF-OX coated papers on both fluting papers and core board papers.

Table 5 shows the magenta color printing measurements made on fluting and core board papers. In the table, ΔE_{00} color differences of CNF/CNF-OX coated papers compared to the control samples (F0 and C0) for both paper types are given. The color differences of the prints on all papers are within the tolerance limits according to ISO 12647-2:2013. There is no color difference that can be acceptable for standard (Ozcan et al. 2020b).

Color values in CNF/CNF-OX coated fluting papers differed at most 10% in F2 (3.65) compared to the control sample. This differentiation is compensated by a* and b* values and the ΔE_{00} limit value is 5 according to ISO 12647-2:2013. Color differences between the papers are negligible, since all the prints obtained are below the acceptable limit. The reason for this color difference is that the paper gaps were filled by CNF/CNF-OX, and the ink remained on the surface of the paper instead of penetrating. This caused the smallest difference in color to feel different and to darken the color, causing the L* value to decrease. When the core board papers were evaluated, it was determined that the color differences were within the limits compared to the standard, but they caused more deviation than the fluting papers. This is because the color of the core board paper surface was not fully tolerated by the printed ink film, and the ink progressed towards the paper fibers.

Another important parameter in this table is the density (ink density) values in prints made with the same amount of ink. Density values have increased in both fluting and core board CNF/CNF-OX coated papers. This is because the CNF/CNF-OX coating smoothes the paper surface, filling pores and thus the ink remains on the surface by preventing it from moving into the pores of the paper. This means better
printability with less ink and the results obtained are consistent with the literature (Reshmy et al. 2020; Rautkoski et al. 2015).

When the gloss values of the printed papers were examined (Figure 4), the printing gloss of the CNF/CNF-OX coated fluting and core board papers was higher than the control papers.

**Conclusions**

The use of CNF/CNF-OX in the paper and cardboard industry not only mitigated environmental impacts, but also enabled the use of low thickness, higher strength, durable and better printable paper and cardboard.

It has been determined that CNF/CNF-OX coated papers have better surface properties and thus improve both paper gloss and printing gloss values.

When the produced papers are examined, it is more convenient to print on hydrophilic papers (all papers except C3 and C4) with water-based ink, and on the papers with increased contact angle (core board papers containing CNF) with oil-based offset inks.

The color differences of the prints on all papers are within the tolerance limits according to ISO 12647-2:2013. There is no color difference that can be acceptable for standard

When the gloss values of printed and unprinted papers were examined, it was determined that the gloss of CNF/CNF-OX coated fluting and core board papers increased.

As a result, the use of CNF/CNF-OX in the production or coating of paper and cardboard is expected to provide an important advantage in the production of food packaging, especially due to its high strength resistance, improved barrier properties, and especially its better printability and also its biocompatible polymer.

**Declarations**

**Compliance with Ethical Standards**

This article does not contain any studies with animals performed by any of the authors.

**Conflict of Interest**

The authors declare that they have no conflict of interest.

**References**

Afra E, Yousefi H, Lakani SA (2014) Properties of chemi-mechanical pulp filled with nanofibrillated and microcrystalline cellulose. J. Biobased Mater. 8:489-494. https://doi.org/10.1166/jbmb.2014.1462
Afra E, Mohammadnejad S, Saraeyan A (2016) Cellulose nanofibils as coating material and its effects on paper properties. Prog. Org. Coat. 101:455-460. https://doi.org/10.1016/j.porgcoat.2016.09.018

Arbatan T, Zhang L, Fang XY, Shen W (2012) Cellulose nanofibers as binder for fabrication of superhydrophobic paper. Chem. Eng. J. 210:74-79. https://doi.org/10.1016/j.cej.2012.08.074

Aspler J, Bouchard J, Hamad W, Berry R, Beck S, Drolet F, Zou X (2013) Review of nanocellulosic products and their applications. In: Dufresne A, Thomas S, Pothen LA (Eds) Biopolymer Nanocomposites: Processing, Properties, and Applications, pp 461-508. https://doi.org/10.1002/9781118609958.ch20

Aulin C, Gällstedt M, Lindström T (2010) Oxygen and oil barrier properties of microfibrillated cellulose films and coatings. Cellulose, 17:559-574. https://doi.org/10.1007/s10570-009-9393-y

Aulin C, Ström G (2013) Multilayered alkyd resin/nanocellulose coatings for use in renewable packaging solutions with a high level of moisture resistance. Ind. Eng. Chem. Res. 52:2582-2589. https://doi.org/10.1021/ie301785a

Bardet R, Belgacem MN, Bras J (2013) Different strategies for obtaining high opacity films of MFC with TiO2 pigments. Cellulose, 20:3025-3037. https://doi.org/10.1007/s10570-013-0025-1

Bayer IS, Fragouli D, Attanasio A, Sorce B, Bertoni G, Brescia R, Corato RD, Pellegrino T, Kalyva M, Sabella S, Pompa PP, Cingolani R, Athanassiu A (2011) Water-repellent cellulose fiber networks with multifunctional properties. ACS Appl. Mater. Interfaces. 3:4024-4031. https://doi.org/10.1021/am102089f

Charani PR, Dehghani-Firouzabadi M, Afra E, Blademo Å, Naderi A, 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. https://doi.org/10.1007/s10570-013-9998-z

Chen P, Yu H, Liu Y, Chen W, Wang X, Ouyang M (2013) Concentration effects on the isolation and dynamic rheological behavior of cellulose nanofibers via ultrasonic processing. Cellulose, 20:149-157. https://doi.org/10.1007/s10570-012-9829-7

Dimic-Misic K, Puisto A, Gane P, Nieminen K, Alava M, Paltakari J, Maloney T (2013) The role of MFC/NFC swelling in the rheological behavior and dewatering of high consistency furnishes. Cellulose, 20:2847-2861. https://doi.org/10.1007/s10570-013-0076-3

Holik H (2006). Handbook of paper and board. John Wiley & Sons.

Dulany MA, Batten Jr GL, Peck MC, Farley CE (2011) Papermaking additives. Kirk-Othmer Encyclopedia of Chemical Technology, 1-28. https://doi.org/10.1002/0471238961.1601160504211201.a01.pub2
Hult EL, Iotti M, Lenes M (2010) Efficient approach to high barrier packaging using microfibrillar cellulose and shellac. Cellulose, 17:575-586. https://doi.org/10.1007/s10570-010-9408-8

Gatti A (2005) Risk assessment of micro and nanoparticles and the human health. Handbook of Nanostructured Biomaterials, American Scientific Publisher.

Klass CP (2011, May) Biobased materials for paper coating. In Proceedings of the Papercon Conference, Covington, KY, USA (pp. 2249-2081).

Lavoine N, Desloges I, Dufresne A, Bras J (2012) Microfibrillated cellulose - its barrier properties and applications in cellulosic materials: a review. Carbohydr Polym 90:735-764. https://doi.org/10.1016/j.carbpol.2012.05.026

Martins NCT, Freire CSR, Pinto RJB, Fernandes SCM, Neto CP, Silvestre AJD, Causio J, Baldi G, Sadocco P, Trindade T (2012) Electrostatic assembly of Ag nanoparticles onto nanofibrillated cellulose for antibacterial paper products. Cellulose 19:1425–1436. https://doi.org/10.1007/s10570-012-9713-5

Mashkour M, Afra E, Resalati H, Mashkour M (2015) Moderate surface acetylation of nanofibrillated cellulose for the improvement of paper strength and barrier properties. RSC Adv. 5:60179-60187. https://doi.org/10.1039/C5RA08161K

McCracken A, Sadeghian P (2018) Corrugated cardboard core sandwich beams with bio-based flax fiber composite skins. J. Build. Eng. 20:114-122. https://doi.org/10.1016/j.jobe.2018.07.009

Mertaniemi H, Laukkanen A, Teirfolk J-E, Ikkala O, Ras RHA (2012) Functionalized porous microparticles of nanofibrillated cellulose for biomimetic hierarchically structured superhydrophobic surfaces. RSC Adv. 2:2882-2886. https://doi.org/10.1039/C2RA00020B

Morsy FA, El-Sherbiny S, Samir M, Fouad OA (2016) Application of nanostructured titanium dioxide pigments in paper coating: a comparison between prepared and commercially available ones. J Coat Technol Res. 13:307-316. https://doi.org/10.1007/s11998-015-9735-7

Nair SS, Zhu JY, Deng Y, Ragauskas AJ (2014) High performance green barriers based on nanocellulose. Sustain. Chem. Process. 2:1-7. https://doi.org/10.1186/s40508-014-0023-0

Ozcan A, Kandirmaz EA, Hayta P, Mutlu B (2019) Examination of the effect of melamine as a filler in paper coatings on print quality. Cellul. Chem. Technol. 53:307-313. https://doi.org/10.35812/CelluloseChemTechnol.2019.53.30

Ozcan A, Arman Kandirmaz E, Zelzele OB (2020) The effect of deinking and binder type on inkjet print quality, Proceedings of the 10th International Symposium on Graphic Engineering and Design, GRID2020, Novi Sad, (pp 387-394). https://doi.org/10.24867/GRID-2020-p43
Ozcan A, Kasikovic N, Arman Kandirmaz E, Durdevic S, Petrovic S (2020) Highly flame retardant photocured paper coatings and printability behavior. Polym Adv Technol. 31:2647-2658. https://doi.org/10.1002/pat.4991

Petroudy SRD, Syverud K, Chinga-Carrasco G, Ghasemain A, Resalati H (2014) Effects of bagasse microfibrillated cellulose and cationic polyacrylamide on key properties of bagasse paper. Carbohydr. Polym. 99:311-318. https://doi.org/10.1016/j.carbpol.2013.07.073

Penttilä A, Sievänen J, Torvinen K, Ojanperä K, Ketoja JA (2013) Filler-nanocellulose substrate for printed electronics: experiments and model approach to structure and conductivity. Cellulose, 20:1413-1424. https://doi.org/10.1007/s10570-013-9883-9

Rautkoski H, Pajari H, Koskela H, Sneck A, Moilanen P (2015) Use of cellulose nanofibrils (CNF) in coating colors. Nord Pulp Pap Res J. 30:511-518. https://doi.org/10.3183/NPPRJ-2015-30-03-p511-518

Reshmy R, Philip E, Paul SA, Madhavan A, Sindhu R, Binod P, Pandey A, Sirohi R (2020) Nanocellulose-based products for sustainable applications-recent trends and possibilities. Rev Environ Sci Biotechnol. 19:779-806. https://doi.org/10.1007/s11157-020-09551-z

Richmond F, Co A, Bousfield D (2012) The coating of nanobrillated cellulose onto paper using flooded and metered size press methods. Paper conference and trade show 2012: growing the future, PaperCon 2012—co-located with control systems 2012. United States, New Orleans, LA

Ridgway CJ, Gane PA (2012) Constructing NFC-pigment composite surface treatment for enhanced paper stiffness and surface properties. Cellulose, 19:547-560. https://doi.org/10.1007/s10570-011-9634-8

Ridgway CJ, Gane PA (2013) Size-selective absorption and adsorption in anionic pigmented porous coating structures: case study cationic starch polymer versus nanofibrillated cellulose. Cellulose, 20:933-951. https://doi.org/10.1007/s10570-013-9878-6

Sharma M, Aguado R, Murtinho D, Valente AJ, De Sousa APM, Ferreira PJ (2020) A review on cationic starch and nanocellulose as paper coating components. Int. J. Biol. Macromol. 162:578-598. https://doi.org/10.1016/j.ijbiomac.2020.06.131

Shen J, Fatehi P, Ni Y (2014) Biopolymers for surface engineering of paper-based products. Cellulose, 21:3145-3160. https://doi.org/10.1007/s10570-014-0380-6

Shen J, Qian X (2012) Application of fillers in cellulosic paper by surface filling: An interesting alternative or supplement to wet-end addition. BioResources, 7:1385-1388.

Skočaj M (2019) Bacterial nanocellulose in papermaking. Cellulose, 26:6477-6488. https://doi.org/10.1007/s10570-019-02566-y
Spence KL, Venditti RA, Rojas OJ, Pawlak JJ, Hubbe MA (2011). Water vapor barrier properties of coated and filled microfibrillated cellulose composite films. BioResources, 6:4370-4388.

Syverud K, Stenius P (2009) Strength and barrier properties of MFC films. Cellulose, 16:75-85. https://doi.org/10.1007/s10570-008-9244-2

Tang Y, Yang S, Zhang N, Zhang J (2014) Preparation and characterization of nanocrystalline cellulose via low-intensity ultrasonic-assisted sulfuric acid hydrolysis. Cellulose, 21:335-346. https://doi.org/10.1007/s10570-013-0158-2

Tang Y, Zhou D, Zhang J, Zhu X (2013) Fabrication and properties of paper coatings with the incorporation of nanoparticle pigments: rheological behavior. Dig J Nanomater Biostruct 8:1699-1710.

Tozlouglu A, Fidan H, Tutuş A, Arslan R, Sertkaya S, Poyraz B, Küçük SD, Sozbir T, Yemsen B, Gucus MO (2021) Reinforcement potential of modified nanofibrillated cellulose in recycled paper production. BioResources 16:911-941.

**Figures**

![Gloss values of papers according to ISO 8254-1:2009 – Part 1](image)

**Figure 1**

Gloss values of papers according to ISO 8254-1:2009 – Part 1
Figure 2

Microscopic images of CNF/CNF-OX coated fluting papers
Figure 3

Microscopic images of CNF/CNF-OX coated core board papers
Figure 4

Gloss values of magenta ink printed papers accordance with ISO 2813:2014.