Application of Surface-Modified Carboxymethylated Nanofibrillated Cellulose as a Strength Enhancer for Specialty Paper

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Pretreated carboxymethylated nanofibrillated cellulose (CM-NFC) was tested as a strength enhancer for specialty paper, such as banknotes made from cotton linter mixed pulp (CLMP). The pretreatment agent was cationic poly(acrylamide) (C-PAM). The CM-CMF prototype was supplied by a Korean manufacturer. Laboratory tests and pilot trials were performed to evaluate the strength enhancement of banknotes incorporated with surface-modified CM-NFC and determine the process problems encountered in a pilot paper machine. The CM-NFC was surface modified with 0.1% C-PAM without any agglomerates. The prepared laboratory handsheets had high paper strength, which was attributed to the higher nanofibril content of surface-modified CM-NFC compared with that of unmodified CM-NFC. Pilot trials showed that the incorporation of 3% surface-modified CM-NFC was highly effective in promoting the strength of banknote without low retention and drainability on the wet-end part of the pilot paper machine. Therefore, surface-modified CM-NFC at a controlled dosage could be used as a strength enhancer for specialty paper without incurring serious problems in a paper mill.

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INTRODUCTION

Many high-value-added specialty papers have been manufactured, such banknotes, personal identity documents, checks, and share certificates (Bobalek *et al*. 2016; Hubbe 2020). Specialty papers often require high quality because they are utilized in diverse environments. Among the types of specialty papers, banknotes require high durability and strength because they are usually exposed to harsh environments (Kyrychok *et al*. 2014; Hubbe 2020; Wang and Sun 2020; Rafiei *et al*. 2023). A commonly used material for banknotes is cotton linter mixed pulp (CLMP), which has high cellulose content and crystallinity index (Sczostak 2009; Lee *et al*. 2023). Various synthetic additives are utilized to enhance their physical properties (Wang and Sun 2020).

Although diverse synthetic paper strength enhancers have been proposed, their wide usage is limited because most of them are petrochemical products and can be carcinogenic (Bender 2006; Amirabad *et al*. 2018). Therefore, developing an eco-friendly, safe, and functional strength enhancer is necessary for continuously producing eco-friendly banknotes.

Nanofibrillated cellulose (NFC), which also has been called cellulose nanofibril, is derived from the natural polymer cellulose, processed into nanometer dimensions. NFC is produced by mechanical fibrillation using a homogenizer, a microgrinder, or a microfluidizer (Nechyporchuk *et al*. 2016; Djafari Petroudy *et al*. 2021). This material is sustainable, biodegradable, and harmless to the human body because it is made of natural cellulosic resources (Sharma *et al*. 2019; LakshmiBalasubramaniam *et al*. 2021; Mokhena *et al*. 2021). The NFCs have low density and high aspect ratio, strength, and stiffness and thus have various applications, such as in nanocomposites, electronics, biomedical devices, packaging, and papers (Sharma *et al*. 2019; Mokhena *et al*. 2021). In particular, the unique characteristics of NFC have raised great interest in their application in the paper industry (Panchal *et al*. 2018; Yusuf *et al.* 2024). The NFCs are effective for manufacturing high-strength paper and barrier-coated packaging paper. Their utilization as a paper strength enhancer has aroused research interest on the improvement of the physical and mechanical properties of paper (Nechyporchuk *et al*. 2016; Lee *et al*. 2020; Mokhena *et al*. 2021). Therefore, NFC is a promising eco-friendly strength enhancer for banknotes.

The NFC grades can be enhanced by pretreatment prior to mechanical treatment. Various pretreatments have been developed, and the four most common are carboxymethylation, TEMPO oxidation, refining, and enzyme treatments (Fernandes *et al*. 2023). The authors' previous study showed that carboxymethylated NFC (CM-NFC) is more effective in enhancing paper strength than refined NFC (RE-NFC) and enzyme-pretreated NFC (EN-NFC) due to the higher nanofibril content. However, the higher viscosity of CM-NFC compared with other NFC grades reduces drainability on the wet-end and causes a high anionic charge, leading to low retention (Kim *et al*. 2019). Therefore, the electrostatic property of CM-NFC must be modified when it is utilized as a wet-end additive in paper mills.

Many treatments have been introduced to modify the anionic charge of NFCs. Glycidyl-trimethyl-ammonium chloride, Girard's reagent T, and 3-chloro-2-hydroxy-propyl trimethyl-ammonium chloride have been used to introduce quaternary amine groups to NFC molecules (Song *et al*. 2010; Chaker and Boufi 2015). Surface modification with cationic polyelectrolytes has been proposed to reverse the totally or partially anionic charge of NFCs (Garland *et al.* 2022; Barrios *et al*. 2023). Among the many types of polyelectrolytes used for this purpose, cationic polyacrylamide (C-PAM) is the most effective in modifying the charge of RE-NFC (Lee *et al*. 2018). Given that organic solvents are not easy to handle and can be toxic to human health, surface modification with cationic polyelectrolytes appears to be a promising solution for neutralizing of NFC (Bourganis *et al*. 2017; Hennecke *et al*. 2018; Henschen 2019). However, previous surface modifications were carried out only on RE-NFC, and the majority of the results were derived only from laboratory tests.

This study analyzed the effect of surface-modified CM-NFC with C-PAM on the strength and process parameters of banknotes, a specialty paper made from CLMP, through laboratory tests and pilot trials.

EXPERIMENTAL

Materials

The CLMP obtained from KOMSCO Co., Ltd. (Daejeon, Republic of Korea) was used to prepare handsheets for laboratory tests and test sheets for pilot trials. CLMP is composed of 70% lint and 30% noil. The CM-NFC prototype was supplied by Moorim P&P (Ulsan, Republic of Korea), and Table 1 shows its manufacturing conditions. The CM-NFC prototype was manufactured by carboxymethylation and high-pressure homogenization using hardwood bleached kraft pulp (HwBKP). The C-PAM was supplied by Kemira Chemicals Korea Corp. (Gunsan, Republic of Korea), as shown in Table 2. Potassium hydroxide flakes (KOH, 93.00%), ethyl alcohol (C_2H_5OH, 95.00%), and n-hexane (C_6H_4, 86.18%) provided by Daejung were used to measure the fiber width of CM-NFC. Titanium dioxide (TiO_2) and epoxidized polyamide (PAE) resin provided by Komsco Co., Ltd. were used as a filler and a wet strength agent, respectively.

Table 1. Manufacturing Conditions for the CM-NFC Prototype Used in this Study

Pretreatment	Carboxylate Group	Mechanical Isolation	Supplier
Carboxymethylation	400 μmol/g	High-pressure homogenizer	Moorim P&P

Table 2. Properties of the C-PAM Used in this Study

Molecular Weight	Charge Density	Supplier
5,000,000 g/mol	1.63 meq/g	Kemira Chemicals Korea Corp

Methods

Characterization and surface modification of CM-NFC

The fiber width and low-shear viscosity of CM-NFC prototype were measured to determine whether it was fibrillated to the nanoscale. The viscosity of NFC has been reported to be proportional to the nanofibril content in the suspension (Lasseuguette *et al*. 2008). The fiber width was analyzed using a field emission scanning electron microscope (FE-SEM; JSM-7610F, JEOL, Tokyo, Japan). Wet CM-NFC pads were prepared as test specimens to measure the fiber width using a vacuum filtration system and then dried by the solvent exchange method using ethyl alcohol and n-hexane. The FE-SEM images of the pads were captured, and the fiber width was measured with image analysis using a 3D imaging software (MP-45030TDI, JEOL, Osaka, Japan). The low-shear viscosity of CM-NFC slurry with 1.0% solids was determined using a low-shear viscometer (DV-IP, Brookfield Engineering Laboratories, Inc., Middleborough, MA, USA) with a spindle number of 64 and a speed of 60 rpm. The temperature of CM-NFC slurries was maintained at 25 °C during the viscosity measurement. The average zeta potential of CM-NFC slurry with 0.01% solids was measured using a zeta potential analyzer (Nano ZS, Malvern Panalytical, Malvern, UK).

The C-PAM with 0.5% concentration was used for the surface modification of CM-NFC. The CM-NFC slurry was diluted to 0.5% consistency using distilled water, and C-PAM was added into this CM-NFC slurry. The dosages of C-PAM were 0.1%, 0.3%, 0.5%, and 0.7% of the oven-dried CM-NFC amount as previously described (Lee *et al.* 2023). After C-PAM was added, the CM-NFC slurry was mixed at 1,000 rpm for 20 min. Agglomeration was observed to determine the minimum dosage of C-PAM needed for the surface modification of CM-NFC.

Handsheet preparation at laboratory scale and measurement of sheet strength

Laboratory tests were performed to identify the effect of surface-modified CM-NFC on the physical properties of the sheets. The CLMP with 1.57% solids was soaked in tap water and then beaten to 425 ± 5 mL CSF using a laboratory-scale Hollander beater. The beaten pulp suspension was then diluted to 0.7% consistency for handsheet manufacturing. The surface-modified CM-NFC slurry was added to the diluted pulp suspension, and the mixture was blended for 5 min at 600 rpm. The dosage of surface-modified CM-NFC slurry was 1%, 3%, and 5% of the oven-dried CLMP. Unmodified CM-NFC was used as a control. Handsheets with a grammage of 170 ± 5 g/m^2 were then prepared in accordance with TAPPI T205 sp-06 (2006). Afterward, the chemical oxygen demand (COD) of the white water was measured using a COD reactor (HI 839800; HANNA Instruments, Smithfield, RI, USA) and a COD detector (DR890; HACH Company, Loveland, CO, USA) was used to qualitatively analyze the retention of CM-NFCs.

The handsheets were wet-pressed at 345 kPa for 5 min and dried at 120 °C using a laboratory wet press (model 326; Wintree Corporation, Osaka, Japan) and a cylinder dryer (Daeil Machinery Co., Ltd., Daejeon, Republic of Korea), respectively.

The handsheets were conditioned at 23 °C and 50% relative humidity (RH) to maintain their moisture content at 8%. Tensile strength (TAPPI T494 om-06 (2006)), folding endurance (TAPPI T511 om-08 (2008)), and bulk (TAPPI T411 om-10 (2010)) were measured. For the analysis of the change in the bonding area in the sheets, the light scattering coefficient was measured using a spectrophotometer (Elrepho, Lorentzen and Wettre, Kista, Sweden) and calculated with Eq. 1,

$$S = \frac{s_W}{w} \times 10 \quad (1)$$

where S is light scattering coefficient, s_W is scattering power of the material, and w is grammage of the sheet.

Pilot trial

Pilot trials were carried out at Komsco Co., Ltd. in Daejeon to determine the effect of surface-modified CM-NFC on the process parameters and physical properties of the test sheets. A pilot paper machine was operated at the R&D center of Komsco Co., Ltd. as shown in Fig. 1. This vat machine has a 300 mm web, runs at a speed of 2 m/min, and produces a sheet in basis weight from 70 to 100 g/m^2.

The surface-modified CM-NFC slurry was prepared with C-PAM. The CM-NFC with 2% solids was diluted to 0.5%, followed by the addition of C-PAM at 0.1% of the oven-dried NFC amount and mixing for 5 min at 600 rpm. Unmodified CM-NFC was used as a control.

Fig. 1. Pilot paper-machine installed in Komsco Co., Ltd.

Table 3 and Fig. 2 show the dosage of additives and flow diagram of the pilot trials, which were performed twice depending on whether the surface-modified CM-NFC was added or not. The CM-NFC prototype was treated with 0.1% C-PAM prior to the pilot trials. The CLMP at 1.50% solids was soaked in tap water and then beaten to 425 ± 10 mL CSF using a pilot-scale Hollander beater. The beaten CLMP furnish was transferred to a beater chest. Then 6% titanium dioxide was added into a beater chest, and 3% PAE resin was added into a vat machine chest in both pilot trials. The surface-modified CM-NFC was added into the beater chest following titanium dioxide addition. Finally, the prepared stock was delivered to the pilot paper machine, and test sheets with basis weight from 70 g/m^2 to 80 g/m^2 were produced.

Table 3. Dosage of Additives in Pilot Trials

Experiment	NFC Dosage (on o.d. CLMP)	Filler Dosage (on o.d. CLMP)	Wet Strength Agent Dosage (on o.d. CLMP)
		TiO_2	PAE resin
No CM-NFC (control)	0%	6%	3%
Surface-modified; CM-NFC (0.1% C-PAM on o.d. CM-NFC)	3%	6%	3%

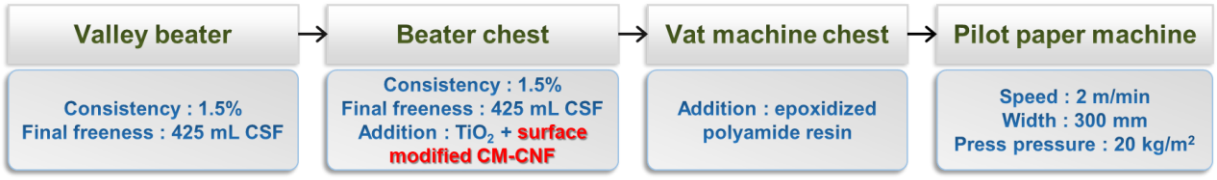

Fig. 2. Flow diagram of pilot trials

During the pilot trials, the process parameters of the pilot paper machine were analyzed. Stock in vat and white water in wire tray were collected to calculate the first pass retention (FPR) with Eq. 2 (Krogerus 1997) by measuring their consistencies (TAPPI T240 om-93 (1993)). The dryness (TAPPI T412 om-94 (1997)) of the test sheets after press was also measured to analyze the drainability of stock containing surface-modified CM-NFC.

The sheets were conditioned at 23 °C and 50% RH to maintain their moisture content at 8%. Tensile strength (TAPPI T494 om-06 (2006)), folding endurance (TAPPI T511 om-08 (2008)), bulk (TAPPI T411 om-10 (2010)), and ash content (TAPPI T211 om-93 (2007)) were measured.

$$\text{First pass retention (FPR)} = \frac{C_h - C_w}{C_h} \times 100 \quad (2)$$

where C_h and C_h refer to headbox and wire tray consistencies (%), respectively.

RESULTS AND DISCUSSION

Characteristics and Surface Modification of CM-NFC

Table 4 shows the CM-NFCs characteristics. The fiber width was 13.5 nm, which was lower than the 100 nm standard that defines nanofibers. According to the fiber width, the CM-NFC used in this study was sufficiently nanosized. The low-shear viscosity was approximately 2,300 cPs, which was relatively higher than that of other NFCs. The viscosity of the NFC slurry was proportional to its nanofibril content, and a higher nanofibril content can effectively improve paper strength (Saarikoski *et al*. 2012; Grüneberger *et al*. 2014). Thus, the CM-NFC qualified as a paper strength enhancer. The zeta potential was −36.0 mV due to the introduction of carboxymethyl groups on the CM-NFC molecules. Therefore, the CM-NFC used in this study can be used as a paper strength enhancer because it contains many nanofibrils. However, surface modification using a cationic polyelectrolyte is necessary to improve its wet web retention.

The C-PAM was used to change the electrostatic properties of CM-NFC. However, CM-NFC aggregates were formed as a consequence of the C-PAM addition. Aggregates are unfavorable to the strength enhancement resulting from the addition of CM-NFC because they can cause irregularities in paper sheet. Therefore, it is necessary to select the minimum dosage of C-PAM in which no aggregates are formed and the charge of CM-NFC is partially changed.

Table 4. Characteristics of CM-NFC Prototype

NFC Type	Fiber Width	Low-shear Viscosity (1.0%, 25 ℃)	Zeta-potential
Carboxymethylated NFC (CM-NFC)	13.5 nm	2,300 cPs	-36.0 mV

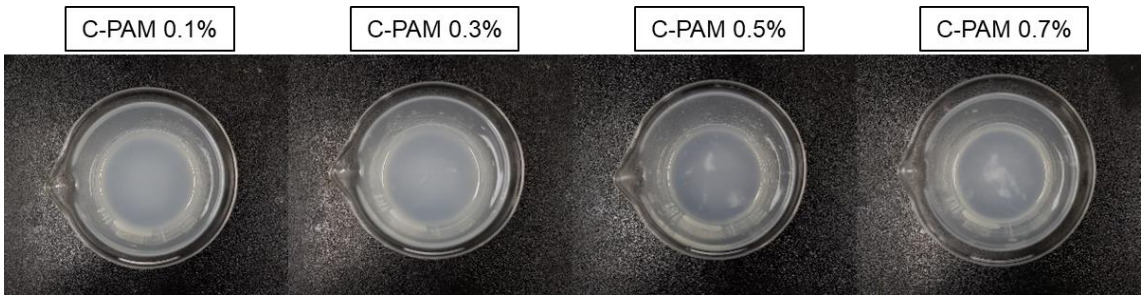

Fig. 3. Images of surface-modified CM-NFC depending on the dosage of C-PAM

Figure 3 shows images captured from the top view of 200 mL beakers containing CM-NFC slurries with varying C-PAM dosages. When the dosage of C-PAM was 0.1% of the oven-dried CM-NFC amount, aggregation was not observed. However, aggregates appeared when the dosage of C-PAM was increased from 0.3% to 0.7%. The dosage of C-PAM for CM-NFC surface modification was determined to be 0.1%.

Laboratory Handsheet Strength

Handsheets were prepared with CLMP to investigate the effect of surface-modified CM-NFC on their physical properties. Figure 4 shows the effect of CM-NFCs on the tensile index and folding endurance of the handsheets, and Fig. 5 shows the influence of CM-NFCs on the light scattering coefficient and bulk of the handsheets.

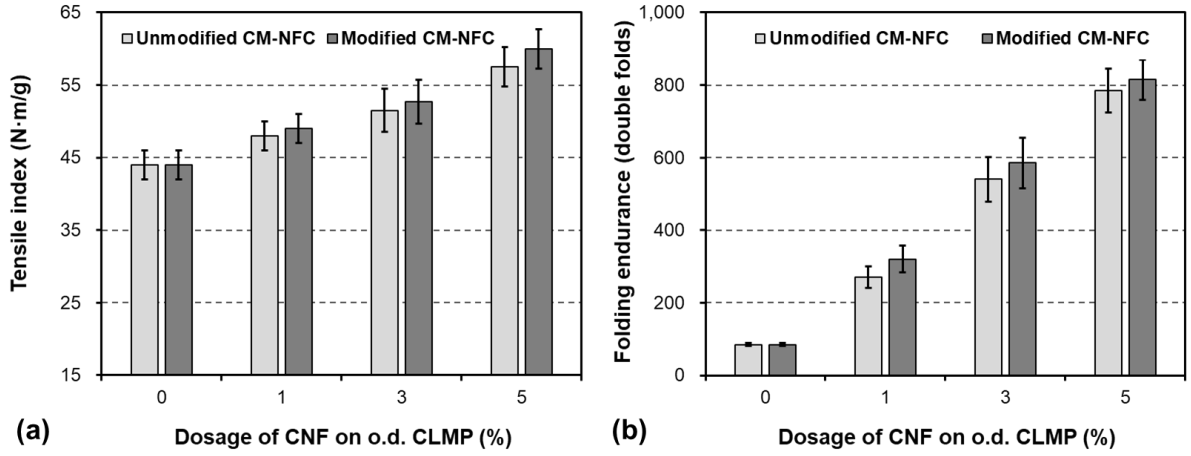

Fig. 4. Effect of CM-NFCs on the (a) tensile index and (b) folding endurance of handsheets

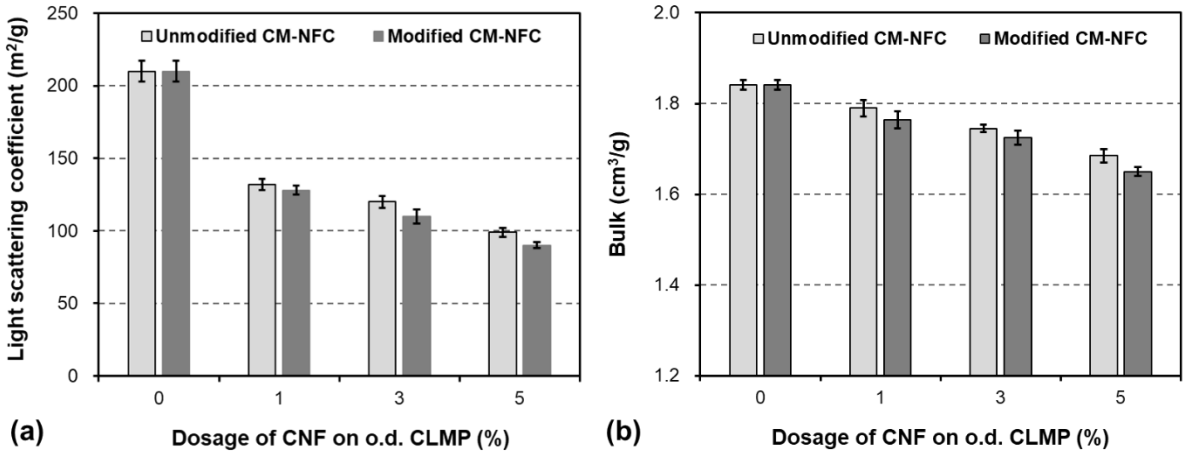

Fig. 5. Effect of CM-NFCs on the (a) light scattering coefficient and (b) bulk of handsheets

As the dosage of CM-NFCs increased, the tensile index and folding endurance increased linearly irrespective of surface modification. Meanwhile, the bulk of the sheets decreased as the strengths increased, indicating the consolidation of the paper structure by

the addition of CM-NFCs (Boufia *et al*. 2016; Pego *et al* 2020; Zeng *et al*. 2021). The nanofibrils are located in the interfiber voids and effectively promote the paper strength by increasing the bonded number and area between fibers (Ahadian *et al*. 2023; Andze *et al*. 2024). The increase in bonded number and area can also affect the decrease in light scattering coefficient, because the number of the voids that cause light scattering decreases when the bonded area between fibers becomes enlarged (Hirn and Schennach 2017).

Comparison of paper strength enhancement revealed that the surface-modified CM-NFC gave relatively higher strengths than the unmodified CM-NFC because the partially charge-reversed CM-NFC could be retained on the handsheets more than the anionic unmodified CM-NFC. The retention trend of CM-NFCs can be confirmed by the COD measurement of the stock supernatant as shown in Table 5. The COD of the stock containing the surface-modified CM-NFC was relatively lower than that of the stock containing the unmodified CM-NFC, indicating that the retention of surface-modified CM-NFC was higher than that of unmodified CM-NFC.

Therefore, the surface-modified CM-NFC was more effective in promoting the strength of the specialty paper made from CLMP compared with the unmodified CM-NFC.

Table 5. Effect of CM-NFCs on the COD of Stock

CM-NFC Dosage (on o.d. CLMP)	1%	3%	5%
Unmodified CM-NFC	50.5 mg/L	133.0 mg/L	202.0 mg/L
Surface-modified CM-NFC	41.0 mg/L	116.0 mg/L	184.5 mg/L

Pilot Trial

The strength of the laboratory handsheets increased linearly as the dosage of surface-modified CM-NFC increased to 5%. Thus, the dosage of surface-modified CM-NFC was selected as 5% in the pilot trials. However, the commercialization of CM-NFC did not proceed at the time of the authors' pilot trials. Consequently, the supply of CM-NFC prototype was not sufficient. Therefore, the dosage of surface-modified CM-NFC was lowered to 3%, which was found to be an intermediate dosage in the laboratory tests.

Table 6 shows the effect of surface-modified CM-NFC on the process parameters in pilot trials. When 3% of surface-modified CM-NFC was added, the consistencies of vat stock and white water increased slightly. The FPR was 55.2% under the control conditions, but the FPR decreased slightly to 51.7% when 3% of surface-modified CM-NFC was added. Given that the difference in retention was less than 5%, surface modification using 0.1% C-PAM was considered effective for retaining CM-NFCs. The dryness of wet web was 29.0% under the control conditions but it increased to 32.4% when 3% of surface-modified CM-NFC was added. The difference in dryness was approximately 3.4%, which did not reach statistical significance. Therefore, the incorporation of 3% surface-modified CM-NFC did not adversely affect the drainability and retention on the wet-end part of the pilot paper machine.

Table 7 shows the effect of surface-modified CM-NFC on the physical properties of the test sheets produced in pilot trials. The ash content showed a difference of approximately 1%, but this value did not reach statistical significance. Strength comparison revealed the tensile strength increased 24.5% in machine direction (MD) and 42.9% in cross directions (CD), and the wet tensile strength increased 13.9% in MD and 23.4% in

CD when surface-modified CM-NFC was added. In particular, a relatively high increase in folding endurance was observed upon the addition of surface-modified CM-NFC, with increase percentages of 356% in MD and 506% in CD. The improvement of folding endurance by CM-NFC was much higher than that of tensile index, which was consistent with previous studies (Kim *et al.* 2019).

The pilot trials showed that 3% surface-modified CM-NFC was effective in promoting the strength of banknotes without causing low retention and drainage on the wet-end part of the pilot paper machine.

Table 6. Effect of Surface-modified CM-NFC on the Process Parameters in Pilot Trials

Experiment	Consistency (%)		Retention (%)	Dryness (%)
	Vat stock	White water		
No NFC (control)	0.067	0.030	55.2	29.0
Surface-modified CM-NFC	0.092	0.045	51.1	32.4

Table 7. Effect of Surface-Modified CM-NFC on the Physical Properties of Sheets in Pilot Trials

Experiment	Grammage (g/m^2)	Thickness (μm)	Density (cm^3/g)	Ash Content (%)	Tensile Index (N·m/g)		Wet Tensile Index (N·m/g)		Folding Endurance (Double Folds)	
					MD	CD	MD	CD	MD	CD
No NFC (control)	71.4	100.4	0.68	3.6	46.5	22.6	10.8	6.4	281	94
Surface-modified CM-NFC	79.6	111.8	0.73	2.6	57.9	32.3	12.3	7.9	1,282	570

CONCLUSIONS

1. The carboxymethylated nanofibrillated cellulose (CM-NFC) did not agglomerate when incorporated with 0.1% of cationic acrylamide (C-PAM). However, agglomerates were observed with a C-PAM dosage of 0.3% and higher. Therefore, 0.1% C-PAM was needed to modify CM-NFC surface without inducing agglomeration.
2. Compared with unmodified CM-NFC, surface-modified CM-NFC was more effective as a strength enhancer of banknotes made from CLMP due to its higher retention during sheet formation.
3. Pilot trials showed that 3% surface-modified CM-NFC was effective in promoting the strength of banknotes without causing serious problems on the wet-end part of the pilot paper machine.
4. These results are expected to be applicable not only to banknote papers but also to products such as security paper, maps, and wallpaper, where high strength characteristics are required.

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