Utilization of Cellulose Micro/Nanofibrils as Paper Additive for the Manufacturing of Security Paper

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Effects of cellulose micro/nanofibrils were investigated relative to the strength and drainability of security paper made from cotton lint mixed pulp. Six types of cellulose micro/nanofibrils were made from bleached softwood kraft pulp (SwBKP) and bleached hardwood kraft pulp (HwBKP) using refining and micro-grinding processes, respectively. After their main properties were measured, handsheets were produced by adding micro/nanofibrils into a cotton pulp furnish, and the physical properties of the sheets were measured. The cotton pulp furnish drainage rate was also analyzed with respect to the dosage and the fibrillation degree of the cellulose micro/ nanofibrils. The cellulose micro/nanofibrils made from SwBKP resulted in higher viscosity, lower particle size, and higher zeta-potential than those made from HwBKP. The nanofibrils made from SwBKP were the most effective for enhancing the tensile strength, folding endurance, and sheet density of the security papers made from cotton lint mixed pulp. However, the cellulose micro/nanofibrils made from SwBKP could be limited as a paper additive in papermaking due to its high drainage resistance. Because the cellulose micro/nanofibrils made from HwBKP also reduced the drainage rate, the dosage and the fibrillation degree should be controlled by simultaneously considering the paper strength and drainage.

Keywords: Cellulose microfibril; Cellulose nanofibril; Bleached kraft pulp; Micro-grinding; Strength; Drainage

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INTRODUCTION

As the specialty paper market has grown faster than other paper grades in the Republic of Korea, the production of specialty papers is increasing rapidly. Many grades of specialty papers are produced in the paper industry, such as electric papers, filter papers, release papers, packaging papers, security papers, and various functional papers (Meinander 1997; Patel 2010). Among these specialty papers, the demand for security papers has increased rapidly due to their use as banknotes, checks, share certificates, and lottery and airline tickets (Bobalek *et al*. 2016). These papers require sophisticated security features, high durability, and high strength (Bobalek *et al*. 2016). Among these requirements, the durability and strength of the security paper are most important because security papers are typically used in the harsh and unexpected service environments. Therefore, some security papers, such as banknotes, are made from cotton, linen, or hemp fibers and sized with gelatin or PVA (Hentzschel 1997; Park *et al*. 2017). However, these conventional and natural raw materials are limited in their capacity to enhance the durability and strength of security papers. Some synthetic materials have been utilized to promote the durability and strength of security papers (Varenberg and Tsipenyuk 2014), but their utilization is limited because they are not eco-friendly. Therefore, a functional and eco-friendly material should be developed and

utilized for the enhancement of the durability and strength of the security paper.

Nanocellulose refers to the cellulose particles with at least one dimension in the nanoscale, ranging from 5 to 100 nm (Abdul Khalil *et al.* 2014; Nechyporchuk *et al.* 2016). It has unique properties such as low density, biodegradability, high aspect ratio, and high strength and stiffness (Gong *et al.* 2011; Salas *et al.* 2014; Xu *et al*. 2018). Nanocellulose can be divided into two main categories: (i) cellulose nanocrystal and (ii) cellulose micro/nanofibrils. Cellulose nanocrystal is extracted from cellulosic fibers dispersed in water by sulfuric acid hydrolysis (Valentini *et al.* 2014), and cellulose micro/nanofibril is produced by mechanical fibrillation such as microfluidizers (Grüneberger *et al.* 2014; Taheri and Samyn 2016), homogenizers (Siró *et al.* 2010; Hettrich *et al.* 2014; Baati *et al.* 2018), and micro-grinders (Abe *et al.* 2007; Nair *et al.* 2014; He *et al*. 2018).

Cellulose micro/nanofibrils have been studied intensively for potential use in various applications, such as polymer nanocomposites, magnetics, electronics, biomedical devices, and membranes (Klemm *et al.* 2006; Abitbol *et al.* 2016; Cao 2018; Dufresne 2018). The paper industry has also gained increasing attention in the manufacturing and application of cellulose micro/nanofibrils, and some studies have reported the application of cellulose micro/nanofibrils as a paper additive or coating material (Boufi *et al.* 2016; Johnson *et al.* 2016; Mazhari Mousavi *et al*. 2018).

Specifically, the use of cellulose micro/nanofibrils as an additive to enhance the physical and mechanical properties of paper has recently generated much interest (Missoum *et al.* 2013; Johnson *et al.* 2016). Cellulose micro/nanofibrils effectively enhance dry strength by increasing the bonded area, sheet density, and filler particle binding in the sheet while retaining the strength properties; however, the addition of cellulose micro/nanofibrils adversely affects the pulp suspension draining rate (Taipale *et al.* 2010; Brodin *et al.* 2013). Therefore, cellulose micro/nanofibrils could effectively improve the durability and strength of security paper if the pulp drainage was well-controlled. No studies have examined the effect of cellulose micro/nanofibrils on the strength and drainage of security paper made from cotton pulp.

This study investigated the effects of cellulose micro/nanofibrils on the physical properties and drainage rate of security paper made from cotton lint mixed pulp. Many types of cellulose micro/nanofibrils were prepared in a laboratory using a Hollander beater and a micro-grinder, depending on the pulp type and the pass number of fibrillation. The micro/nanofibrils characteristics were determined. Handsheets were made by adding cellulose micro/nanofibrils into a cotton lint mixed pulp furnish, and the sheets' physical properties were measured. The drainability of the cotton lint mixed pulp furnish was also analyzed with respect to the dosage of cellulose micro/nanofibrils.

EXPERIMENTAL

Materials

Bleached softwood kraft pulp (SwBKP) and bleached hardwood kraft pulp (HwBKP), supplied by Moorim Paper (Jinju, Republic of Korea), were used to make the cellulose micro/nanofibrils. Cotton lint mixed pulp (CLMP) was obtained from KOMSCO Co. Ltd. (Daejeon, Republic of Korea) and used to prepare handsheets as well as to measure the drainage rate in the laboratory. Ethyl alcohol (C_2H_5OH, 95.0%, Daejung, Republic of Korea), acetone (CH_3COCH_3, 99.9%, Fisher, USA), and n-hexane (C_6H_{14}, 95.0%, Daejung, Republic of Korea) were used for the solvent exchange of micro/nanofibril pads.

Methods

Manufacturing of cellulose micro/nanofibrils made from bleached kraft pulps

Cellulose micro/nanofibrils were manufactured *via* beating and micro-grinding. SwBKP and HwBKP at 1.57% solids were soaked in tap water and then beaten to 450 ± 5 mL CSF using a laboratory Hollander beater. The beaten pulp slurry was diluted to 1.0% consistency for fibrillation. The pulp slurry at 1.0% solids was then fibrillated using a Super Mass Colloider (MKCA6-2-J, Masuko Sangyo Co., Ltd., Kawaguchi, Japan) at 1,500 rpm. The pulp slurry was fed continuously to the grinder consisting of two stone grinding disks positioned on top of each other. The gap between the two disks was adjusted to -150 μm. The cellulose micro/nanofibril samples were corrected at pass numbers of 3, 5, and 7, respectively.

Characterization of cellulose micro/nanofibrils

The fiber width of the cellulose micro/nanofibrils was analyzed using a FE-SEM (JSM-7610F, JEOL, Tokyo, Japan) to ensure the material was ground to the nano-scale after micro-grinding. Wet micro/nanofibril pads were prepared as test specimens for the measurement of fiber width using the vacuum filtration system. The wet micro/nanofibril pads were dried using the solvent exchange method with ethyl alcohol, acetone, and n-hexane to complete the specimens. Then FE-SEM images of the pads were captured, and the fiber width was measured with image analysis using 3D image software (MP-45030TDI, JEOL, Osaka, Japan).

The viscosity, average particle size, and zeta-potential of the cellulose micro/nanofibril samples were measured to evaluate their main properties depending on the pulp type and the pass number of fibrillation. The viscosity for 1.0% CNF slurry was determined using a low-shear viscometer (DV-IP, Brookfield Engineering Laboratories, Middleborough, USA) at 25 °C and the zeta-potential for 0.01% CNF slurry was measured using a zeta-potential analyzer (Nano ZS, Malvern Panalytical, Malvern, UK). The laser scattering-based particle size measurement is not a perfect method for detecting the fiber dimensions because micro/nanofibrils have a high aspect ratio (Gantenbein *et al.* 2011). However, particle size data were used to indicate indirectly the size differences between the micro/nanofibrils. The particle size for 0.2% CNF slurry was measured using a particle analyzer (1090 LD, CILAS, Orléans, France).

Handsheet manufacturing and physical property measurement

The CLMP at 1.57% solids were soaked in tap water and then beaten to 170 ± 5 mL CSF using a laboratory Hollander beater because the security paper is usually made from the CLMP furnish at the freeness of 170 mL CSF in Korean paper mills. The beaten pulp slurry was then diluted to 0.7% consistency for handsheet preparation. Handsheets, with a grammage of 90 ± 5 g/m^2, were produced according to TAPPI T205 sp-06 (2006). Then the cellulose micro/ nanofibril suspension was added to the pulp slurry and mixed for 5 min at 600 rpm. The cellulose micro/nanofibril suspension dosages were 3, 6, and 10% of the oven-dried fibers. 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 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), and bulk (TAPPI T411 om-10 2010) were measured to identify the effect of the cellulose micro/nanofibrils on the physical properties of the sheets.

Drainage determination

The CLMP was beaten and CLSM furnish was diluted in the same manner as handsheet manufacturing and physical property measurement section. Drainage tests were carried out on a dynamic filtration system (DFS, BTG, Herrsching, Germany). The stirring chamber was filled with 860 g of 0.7% CLMP furnish, and then the furnish was stirred for 30 s at 600 rpm before adding a cellulose micro/nanofibril suspension. After adding the cellulose micro/nanofibrils into the CLMP furnish, the mixture was stirred for 5 min at 600 rpm. The dosages of the cellulose micro/nanofibril suspension were 3, 6, and 10% of oven-dried fibers. Filtration through a 60-mesh screen began immediately after the final stirring for 5 min, and the filtrate was continuously measured for 10 min. The drainage curve was obtained as the amount of drained water as a function of drainage time, and the drainage rate was calculated, as shown in Fig. 1.

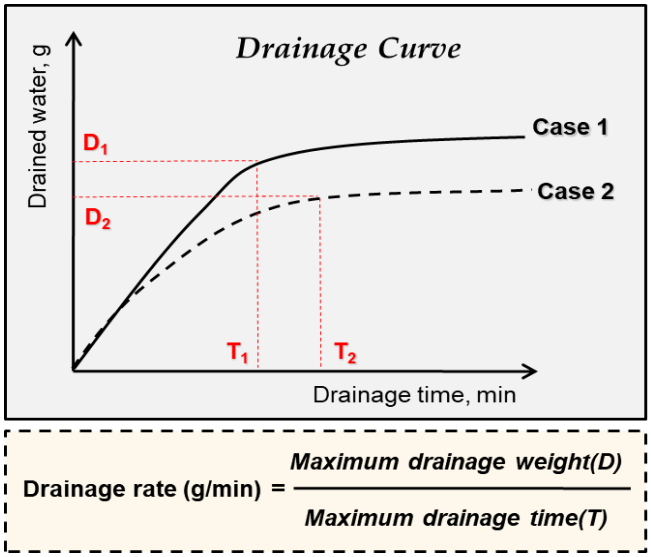

Fig. 1. Drainage rate determination of the CLMP furnish in the presence of cellulose micro/nanofibrils

RESULTS AND DISCUSSION

Characterization of Cellulose Micro/Nanofibrils made from Bleached Kraft Pulps

To determine the cellulose micro/nanofibrils fiber width, FE-SEM images of micro/nanofibril pads were captured, and the fiber width was measured using 3D image software. Figures 2 and 3 show the FE-SEM images of cellulose micro/nanofibrils made from SwBKP and HwBKP, respectively. The fiber width of micro/nanofibrils made from SwBKP decreased as the pass number increased. When the pass number was three, micro/nanofibrils fiber widths of > 100 nm were observed. When the pass number increased more than five times, micro/nanofibrils with fiber widths of > 100 nm were not observed. HwBKP showed a similar trend to that of SwBKP. As the pass number increased more than five times, micro/nanofibrils with fiber widths > 100 nm were not observed any more in the FE-SEM images. However, the widths of all fibers could not be measured. As nanofibrils are defined as nano-sized fibers < 100 nm wide or micron-sized fibers with nano-dimension cross-sectional structures (Chinga-Carrasco 2011; Isogai *et al.* 2011), fibers that were treated less than three times can be defined as cellulose microfibrils, and those treated more than five times can be defined as cellulose nanofibrils.

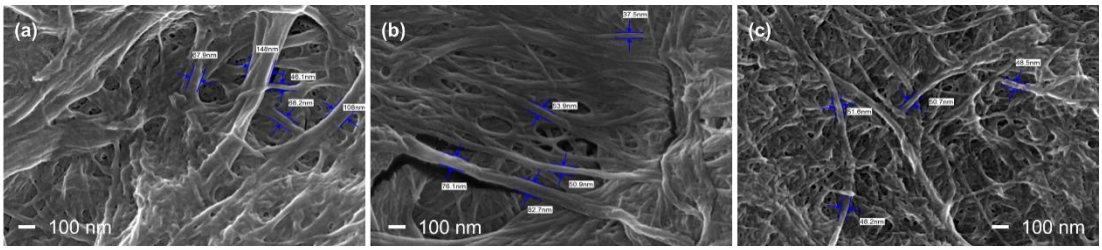

Fig. 2. FE-SEM images of cellulose micro/nanofibrils made from SwBKP depending on the pass number of micro-grinding, (a) pass number 3, (b) pass number 5, and (c) pass number 7.

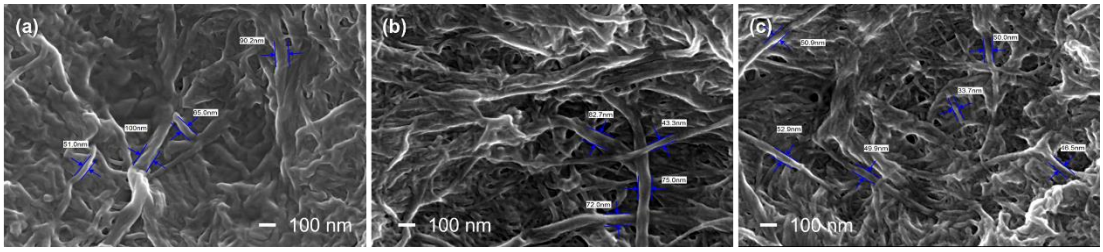

Fig. 3. FE-SEM images of cellulose micro/nanofibrils made from HwBKP depending on the pass number of micro-grinding, (a) pass number 3, (b) pass number 5, and (c) pass number 7.

Table 1 shows the main properties of cellulose micro/nanofibrils in relation to the pass number of micro-grinding. As the pass number increased, viscosity increased, and average particle size decreased. Micro/nanofibrils made from SwBKP showed higher viscosity and lower average particle size than those made from HwBKP. The viscosity of the cellulose micro/nanofibrils has been reported to be proportional to the nanofibril concentration in the suspension (Lasseuguette *et al.* 2008). More intense mechanical treatment reduced the amount of microscopic fibers and increased the nanofibril concentration in the suspensions (Shogren *et al.* 2011) according to FE-SEM image analysis and average particle size in this study. Especially, it was reported that SwBKP was found to be faster and more easily fibrillated compared to HwBKP (He *et al*. 2018). Therefore, micro/nanofibril suspensions made from SwBKP had relatively higher viscosity compared to those made from HwBKP. The zeta-potential decreased slightly as the pass number increased, and the micro/nanofibrils made from SwBKP showed slightly higher zeta-potential than those made from HwBKP because HwBKP generally has higher hemicellulose content (Park *et al.* 2017). Additionally, it could be concluded that micro/nanofibrils from SwBKP had higher specific surface area than those from HwBKP (Spence *et al.* 2010) because the specific surface area increased as the particle size decreased.

Table 1. Main Properties of CNFs made from SwBKP and HwBKP in Relation to the Pass Number of Micro-Grinding

Pass Number	Viscosity (cPs)		Average Particle Size (μm)		Zeta Potential (mV)	
	SwBKP	HwBKP	SwBKP	HwBKP	SwBKP	HwBKP
3	1,430	700	17.3	20.3	-21.0	-22.0
5	2,140	1,550	14.5	19.0	-21.1	-22.8
7	3,500	2,770	13.4	15.3	-21.5	-24.0

Effect of Cellulose Micro/Nanofibrils on the Physical Properties of Sheets

The sheets were made with CLMP to prepare the security papers prototypes in the laboratory and investigate the effect of SwBKP and HwBKP cellulose micro/nanofibrils on the physical properties of the sheets, such as tensile index, folding endurance, and bulk. Figures 4 and 5 show the effect of cellulose micro/nanofibrils on the tensile index and folding endurance of handsheets.

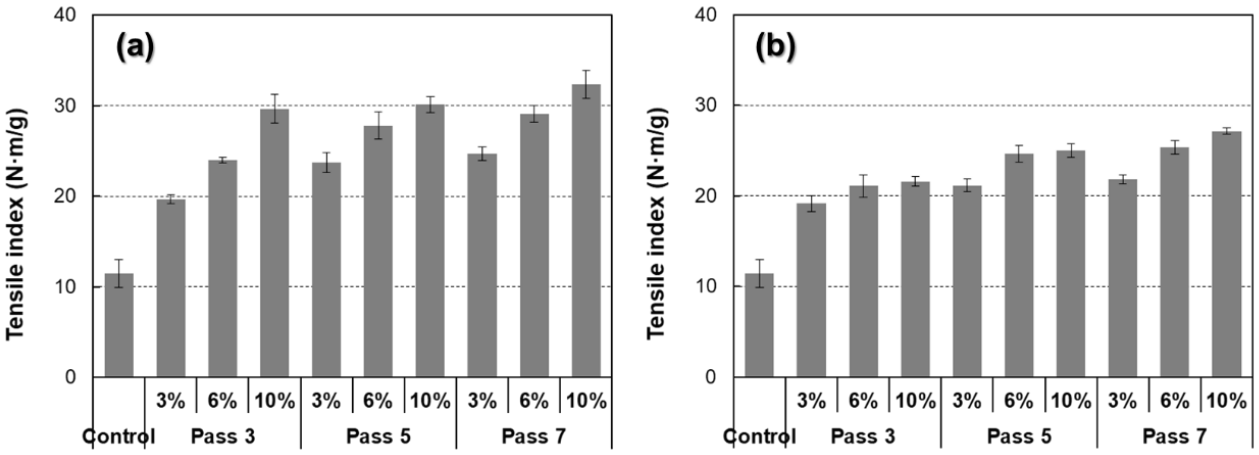

Fig. 4. Effect of cellulose micro/nanofibrils made from (a) SwBKP and (b) HwBKP on the tensile index of sheets

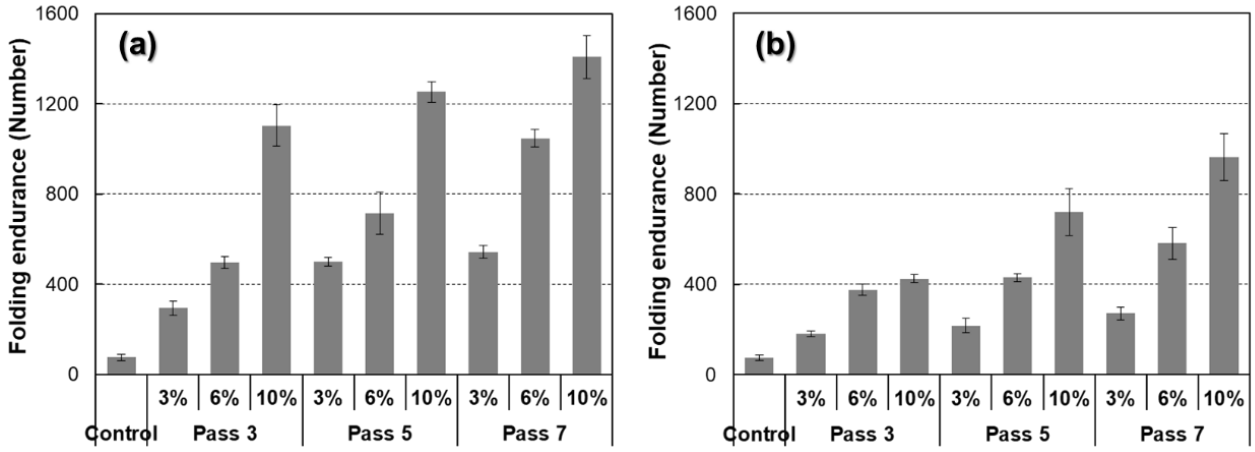

Fig. 5. Effect of cellulose micro/nanofibrils made from (a) SwBKP and (b) HwBKP on the folding endurance of sheets

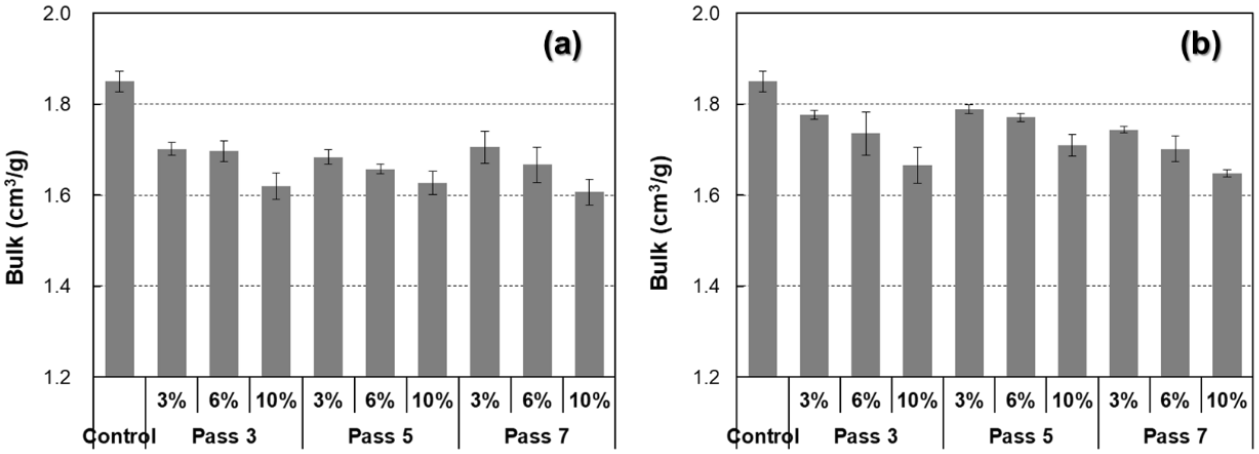

Fig. 6. Effect of cellulose micro/nanofibrils made from (a) SwBKP and (b) HwBKP on sheet bulk

In all cases the micro/nanofibrils dramatically increased the tensile index and folding endurance. Specifically, as the addition level of micro/nanofibrils increased, the strengths of the sheets increased linearly, and the nanofibrils exhibited a greater ability to improve strengths than the microfibrils.

The micro/nanofibrils from SwBKP improved the tensile index and folding endurance more than those from HwBKP. This enhancement of sheet strengths is a direct consequence of the increase in specific surface area (Brodin and Eriksen 2015). The boost in specific surface area promotes the formation of fiber-fiber bonds, which consolidate the paper structure and increase sheet density (Brodin and Eriksen 2015; Boufi *et al.* 2016).

SwBKP showed higher tensile strength, higher folding endurance, and low sheet bulk, as shown in Fig. 6, because the suspensions made from SwBKP had a higher viscosity, which indicated high nanofibril content. It was also noticeable that the promotion in folding endurance by cellulose micro/nanofibrils was much higher than that in the tensile index, and the highest promotion in paper strengths was observed in the presence of nanofibrils that were treated seven times.

It was concluded that the nanofibrils from SwBKP were the most effective for promoting the tensile strength, folding endurance, and density of the security papers made from CLMP furnish.

Effect of Cellulose Micro/Nanofibrils on the Drainage of CLMP Furnish

It has been reported that one of the major disadvantages of using CNF as a paper additive is the detrimental effect on stock drainage (Taipale *et al.* 2010). Drainage is a critical parameter in the papermaking process because it limits the production efficiency of the paper machine (Boufi *et al.* 2016). Therefore, the effect of cellulose micro/nanofibrils on stock drainage should be analyzed before utilizing the material in the papermaking process.

Figures 7 and 8 show the drainage curves of CLMP furnish with respect to the cellulose micro/nanofibrils dosage made from SwBKP and HwBKP, respectively. CLMP drainage in the presence of cellulose micro/nanofibrils made from SwBKP resulted in the dramatic deterioration of drainage, even in the case of low dosage. However, HwBKP showed a different drainage rate from that of SwBKP. The drained water of the CLMP furnish in the presence of the cellulose micro/nanofibrils from HwBKP demonstrated a linear decrease with respect to the dosage and the pass number. The drainage rate was calculated in order to objectively evaluate the effect of the cellulose micro/nanofibrils on the pulp slurry drainage, as shown in Fig. 9. A significant reduction in the drainage rate was observed when SwBKP cellulose micro/nanofibril was added into CLMP furnish.

In the case of HwBKP, the CLMP furnish drainage rate resulted in a linear decrease with respect to the dosage and the pass number. The deterioration in drainage can be explained by the increase in the specific surface area, which caused the drainage resistance (Hubbe and Heitmann 2007), in the presence of the cellulose micro/nanofibrils in the CLMP furnish.

Therefore, the cellulose micro/nanofibrils made from SwBKP are limited in their use as a paper additive because they are highly resistant to drainage. As the cellulose micro/nanofibrils made from HwBKP also reduces drainability, the dosage and the fibrillation degree should be controlled considering the paper strengths and drainage simultaneously.

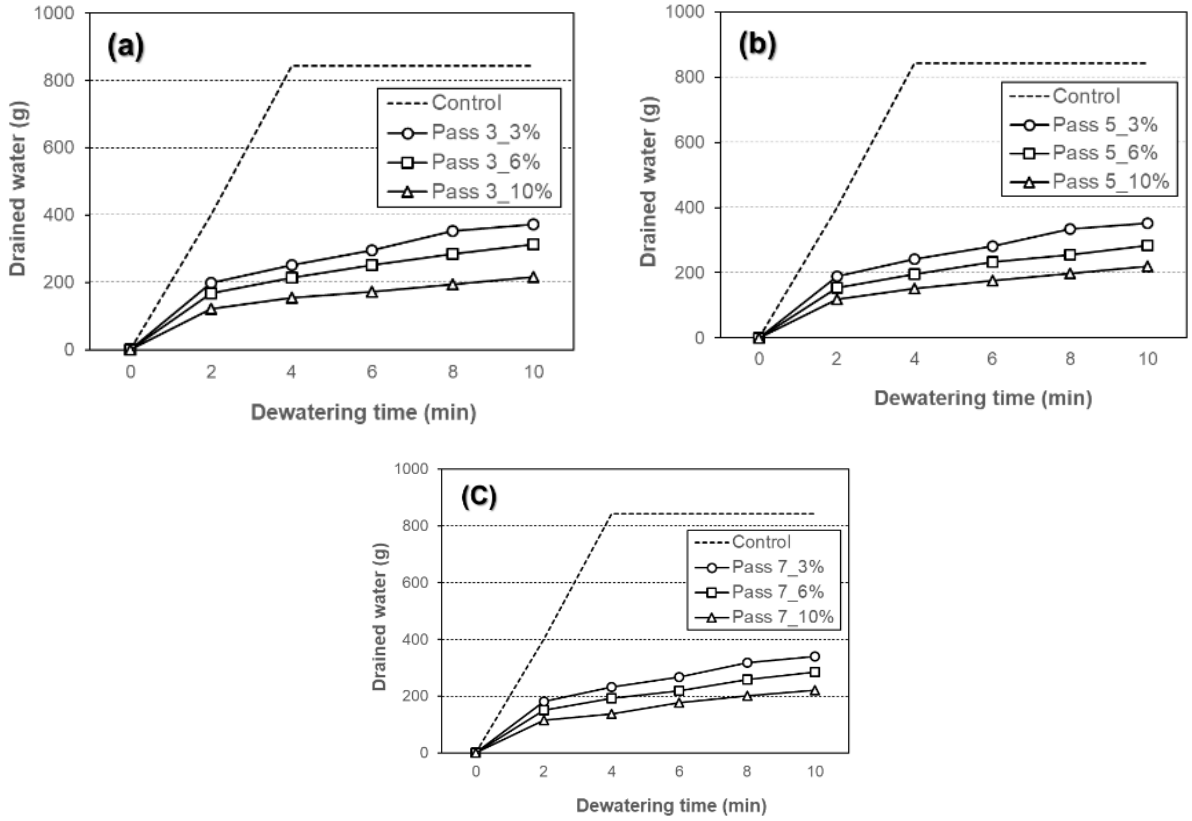

Fig. 7. Effect of cellulose (a) microfibril (pass number 3), (b) nanofibril (pass number 5), and (c) nanofibril (pass number 7) made from SwBKP on drainage curves of CLMP furnish

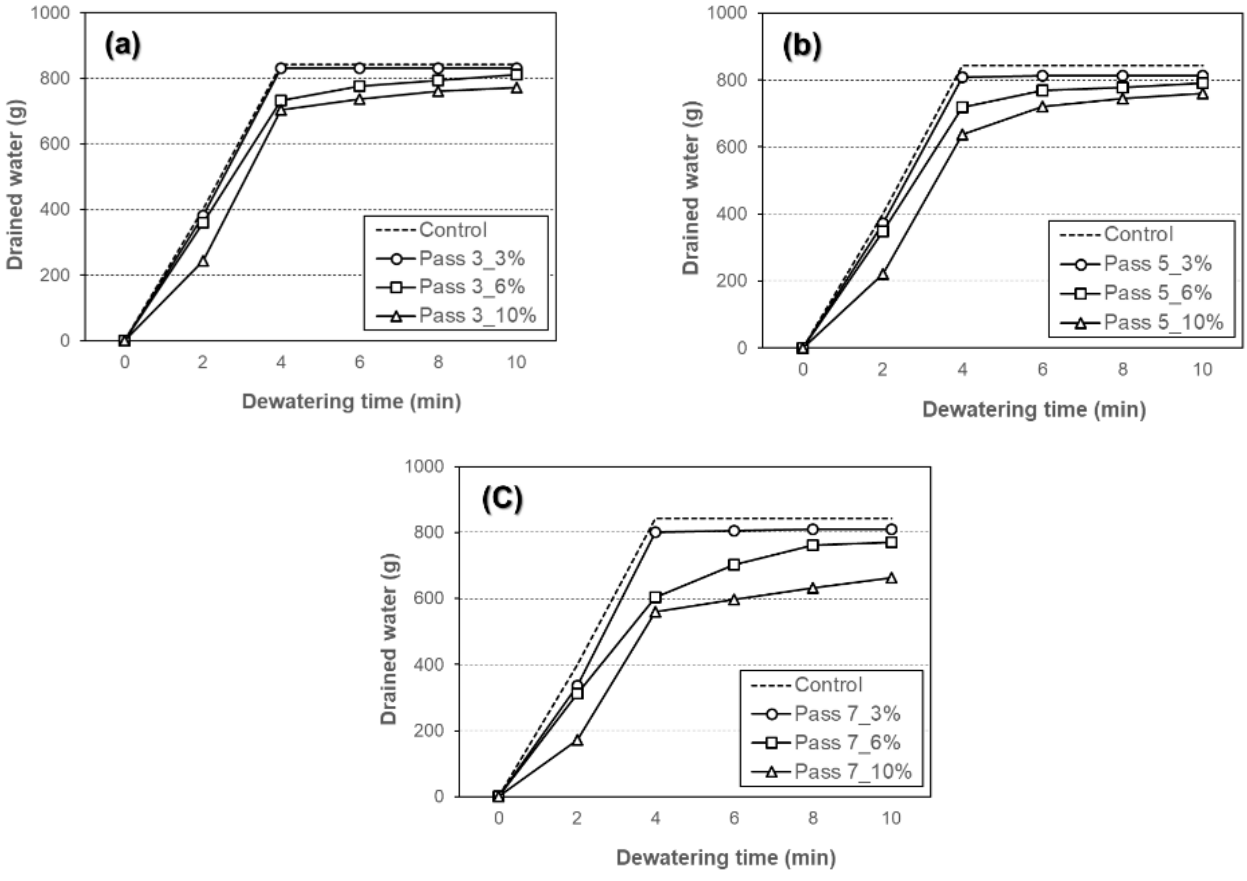

Fig. 8. Effect of cellulose (a) microfibril (pass number 3), (b) nanofibril (pass number 5), and (c) nanofibril (pass number 7) made from HwBKP on drainage curves of CLMP furnish

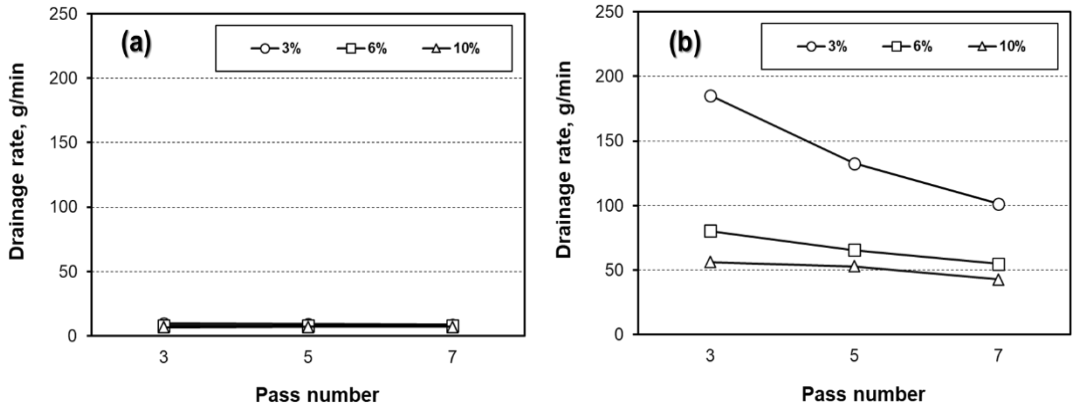

Fig. 9. Effect of cellulose micro/nanofibrils made from (a) SwBKP and (b) HwBKP on the drainage rate of CLMP furnish

CONCLUSIONS

1. Cellulose micro/nanofibrils were manufactured by a combination of refining and micro-grinding processes with softwood bleached kraft pulp (SwBKP) and hardwood bleached kraft pulp (HwBKP). The micro/nanofibrils made from SwBKP at the same mechanical treatment levels resulted in higher viscosity, lower particle size, and higher zeta-potential than those made from HwBKP.
2. The cellulose nanofibrils made from SwBKP were the most effective for promoting tensile strength, folding endurance, and sheet density of the security papers made from cotton lint mixed pulp (CLMP) furnish.
3. A significant reduction in the drainage rate was observed when cellulose micro/nanofibril made from SwBKP was added into the CLMP furnish. In the case of HwBKP, the CLMP furnish drainage rate resulted in a linear decrease with respect to the dosage and the pass number. Therefore, the cellulose micro/ nanofibrils made from SwBKP are limited in their use as a paper additive because they are highly resistant to drainage. As the cellulose micro/nanofibrils made from HwBKP also reduce drainability, the dosage and the fibrillation degree should be controlled considering paper strengths and drainage simultaneously.

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