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Effect of Nanofibrillated Cellulose Made from Enzyme-pretreated Bamboo Pulp on Paper Strength

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The applicability of bleached bamboo kraft pulp (Ba-BKP) was explored as a raw material for the manufacture of nanofibrillated cellulose (EN-NFC) made of enzyme-pretreated pulps and the effects of the EN-NFC on enhancing paper strength. The Ba-BKP was pretreated using an endo-glucanase enzyme at 50 °C and pH 6, after which the EN-NFC was made by micro-grinding. Bleached hardwood kraft pulp (Hw-BKP) was used as a control, and the non-enzymatic refining pretreatment of BKPs was compared with the enzyme pretreatment. The EN-NFC was incorporated into handsheets, and the sheet strengths were measured. The physical properties of the NFC made from the Ba-BKP were similar to those made from the Hw-BKP. The NFC prepared following enzyme pretreatment were smaller and more uniform than those pretreated with refining. The EN-NFC made from the Ba-BKP was effective at enhancing tensile index by 52.7%, and burst index by 210.2% when 2% of EN-NFC was added in the furnish, and those of handsheets containing the EN-NFC made from Hw-BKP showed the similar improvement. Therefore, Ba-BKP can be used as a raw material for the manufacture of EN-NFC that confers similar physical properties and strength enhancement to paper as those made from Hw-BKP.

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INTRODUCTION

Cellulose is a natural and abundant organic polymer. Recently, with the increasing global climate change and environmental problems, cellulose has attracted increasing attention for replacing fossil-fuel-based materials. Accordingly, the market for cellulose materials is expected to increase steadily (Wood 2020), and the possibility of utilizing non-wood-based resources such as kenaf, bagasse, oil palm trunk, and bamboo to replace wood-based fibers has been evaluated (Dungani *et al*. 2014; An *et al*. 2020). In particular, bamboo fibers are similar to wood-based fibers and have excellent mechanical properties (Chen *et al*. 2020). Thus, bamboo is considered suitable for applications in the paper industry (An *et al*. 2020). Approximately 1,500 species of bamboo are distributed over 22 million hectares worldwide and are most widely distributed in the Pacific region, making it a biomass that is easy to use in Asia (Yu 2018). In addition, bamboo is readily cultivated and is fast growing (Liu *et al*. 2012). Therefore, many countries produce bamboo pulp, which is utilized as a new resource for papermaking (Chen *et al*. 2019). The utilization of bamboo fiber is expected to increase in the future, and further research is required to develop new applications.

Nanocellulose can be manufactured through chemical and mechanical processes starting with cellulose fibers that can be obtained from various species. Unlike conventional pulp fibers, nanocellulose with a large surface area has high reactivity (Dufresne 2013). As a typical chemical process, short rod-shaped cellulose nanocrystals (CNCs) can be manufactured *via* acid hydrolysis, and nanofibrillated cellulose (NFC) can be manufactured by a mechanical process using equipment such as a homogenizer and micro-grinder. Because NFC particles have a high aspect ratio, the strength of paper can be improved by incorporating even a small amount of NFC into the paper (González *et al*. 2013). Park *et al*. (2018) reported that NFC is highly promising for use as an internal additive in the papermaking process. According to previous studies about NFC applications in paper mills, it was reported that NFC can be effective to improve the physical properties of various types of papers, *e.g.* tissue, special paper, printing paper, and coated paper (Gonzáles *et al.* 2012; Hubbe *et al.* 2017; Guan *et al.* 2019; Kim *et al.* 2019).

Despite these advantages, various pretreatment technologies have been performed to complement for the drawbacks such as repetitive mechanical treatment and high energy consumption required to produce NFC. Refining pretreatment (Lee and Mani 2017), carboxymethylation pretreatment (Wågberg *et al*. 2008; Onyianta *et al*. 2018), alkaline treatment (Alemdar and Sain 2008), and other methods of combining mechanical treatment and treatment technologies have been proposed. Among the proposed pretreatments, biological pretreatment, or enzyme pretreatment (Pääkkö *et al*. 2007), is recognized as having a very high potential because it is safe and environmentally friendly (Ribeiro *et al*. 2019), consumes little energy compared to other pretreatments, and does not use organic solvents or chemicals (Trache *et al*. 2020).

Cellulase, an enzyme acting on cellulose, occurs in three types: endo-glucanase, exo-glucanase (cellobiohydrolase), and β-glucosidase, depending on the mode of action (Jayasekara and Ratnayake 2019). Endo-glucanase cleaves the β-1,4 glycosidic bonds of cellulose; thus, the amorphous region is preferentially hydrolyzed, thereby reducing the size of the fibers and increasing the crystallinity (Kim *et al*. 2017). Some studies showed that cellulase reduced the fiber length rapidly and produced fines, thereby lowering the strength characteristics of paper (Lecourt *et al*. 2010; Buzala *et al*. 2016). Buzala *et al* (2016) showed that cellulase was effective at reducing the refining energy. Therefore, the cellulase is advantageous to make the mechanical treatment more efficient, such that the NFC can be manufactured at low energy consumption.

This study explored the applicability of Ba-BKP as a raw material for the production of EN-NFC and the effect of EN-NFC on paper strength. The changes in the Ba-BKP and Hw-BKP fibers pretreated with endo-glucanase were measured, and EN-NFC was subsequently made by micro-grinding. After preparation, the EN-NFC was incorporated into handsheets, and the physical properties were measured to identify the effects of bamboo EN-NFC on the strength of paper.

EXPERIMENTAL

Materials

Bleached bamboo kraft pulp supplied by Jeongho Industrial Co., Ltd., (Seoul, Republic of Korea), as shown in Fig. 1, and Hw-BKP supplied by Moorim Paper Co., Ltd., (Jinju, Republic of Korea) was used to make the NFC and handsheets. Cellulase was used

to prepare EN-NFC. The endo-glucanase type of cellulase, supplied by Novozymes A/S(Copenhagen, Denmark), was used to prepare EN-NFC. The specifications of the enzyme are listed in Table 1.

Table 1. Enzymatic Activity and Optimal Reaction Conditions

Enzyme	* Activity According to Manufacture Data (ECU/g)	* Optimal Reaction Conditions	
		Temperature (°C)	pH
Fiber Care D	9,800	40-60	6-9

* Specifications of enzyme given by the suppliers' data sheets

Fig. 1. Images of Ba-BKP

Methods

Enzyme pretreatment of bleached kraft pulps (BKPs)

The disintegrations of Ba-BKP and Hw-BKP were performed at a consistency of 2% using a laboratory disintegrator (Daeil Machinery Co., Ltd., Daejeon, Republic of Korea) with 30,000 revolutions. Subsequently, the pH and temperature of the pulp slurry were adjusted to the optimum reaction conditions for the enzyme, as shown in Table 1. Thereafter, 0.1%, 0.5%, 1.0%, 2.0%, and 5.0% of enzyme relative to the mass of the oven-dried fibers were added to the pulp slurry. After addition of the enzyme, the pulp slurry was stirred at a rate of 600 rpm at 50 °C and a pH of 6.0 for 6 h. The temperature of the pulp slurry was adjusted to 95 °C for 30 min after reacting for 6 h to denature the enzyme. The Ba-BKP and Hw-BKP were also beaten to 450 ± 5 mL CSF using a laboratory valley beater (FRANK-PTI GmbH, Birkenau, Germany) without the enzyme to compare the beating and enzyme pretreatments for the manufacture of NFC, and the detail specifications of the valley beater is shown in Table 2.

Table 2. Specifications of a Laboratory Hollander Beater for Mechanical Pretreatment of BKPs

Manufacturer	Processing capacity	Operating consistency	Roller speed
FRANK-PTI	23 L	1.57%	500 rpm

Characterization of BKP fibers pretreated by enzyme

The main properties of the fibers were measured to evaluate the effects of enzymatic pretreatment. A fiber analyzer (FQA-360, OpTest Equipment Inc., Hawkesbury, ON, Canada) was used to measure the lengths and widths of the fibers. To analyze the shapes of the pulp fibers after kraft pulping before fractionation, images were obtained at 100× magnification using an optical microscope (BX51, Olympus, Tokyo, Japan). Pulp fiber crystallinity after enzymatic pretreatment was evaluated by X-ray diffractometry (D8 Advance A25, Bruker, Billerica, MA, USA) according to the Segal method (Segal *et al*. 1959).

Manufacture of cellulose nanofibrils made from BKPs

Cellulose nanofibrils prepared by refining without an enzyme (RE-NFC) were manufactured by beating and micro-grinding, and EN-NFC was manufactured by enzyme pretreatment and subsequent micro-grinding, as shown in Table 3. The beaten or enzyme-pretreated pulp slurry was diluted to 1.0% consistency for fibrillation. The pulp slurry with 1.0% solid content was then fibrillated using a Super Mass Colloider (MKZA6-2, 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 minus 150 μm. The NFC samples were ground for 1, 3, 5, 7, and 9 passes (hereafter referred to as the pass number).

Table 3. Manufacturing Conditions of NFC

| NFC Grade | Pretreatment | | Mechanical Treatment |
	Type	Dosage of Enzyme (% on oven-dried fibers)	
RE-NFC	Refining (450 mL CSF)	0.0	Micro-grinding using a Super Mass Colloider (pass number = 9)
EN-NFC0.1	Enzyme (endo-type cellulase)	0.1	
EN-NFC0.5		0.5	
EN-NFC5.0		5.0	

Characterization of NFC

The average particle sizes of the NFC samples were measured to evaluate their main properties depending on the BKP type and the pass number of fibrillation. Laser-scattering-based particle size measurement is not a perfect method for detecting the fiber dimensions because NFC have a high aspect ratio (Gantenbein *et al*. 2011). However, it was thought that the particle size data could be used to indirectly indicate the size differences among the NFC (Park *et al*. 2018). The particle size was measured using a particle analyzer (1090 LD, CILAS, Orléans, France).

The fiber widths of the NFC were analyzed using field-emission scanning electron microscopy (FE-SEM) (JSM-7610F, JEOL, Tokyo, Japan) to confirm that the NFC were ground to the nano-scale by micro-grinding. Wet NFC pads were prepared as test specimens for the fiber width measurements using a vacuum filtration system. The wet NFC pads were dried using the solvent exchange method with ethyl alcohol, acetone, and n-hexane in order to prevent deformation of specimens during drying (Stelte and Sanadi 2009; Kim *et al*. 2019). After capturing FE-SEM images of the pads, the fiber width of 100

individual nanofibers for each NFC sample were measured by image analysis using 3D image software (MP-45030TDI, JEOL, Osaka, Japan).

Handsheet manufacture and physical property measurement

The Hw-BKP at 1.57% solid content was soaked in tap water and then beaten to 450 mL ± 5 mL CSF using a laboratory valley beater. The beaten pulp slurry was then diluted to 0.7% consistency for handsheet preparation. Handsheets with a grammage of 60 g/m^2 ± 5 g/m^2 were produced according to the TAPPI T205 sp-06 (2006) protocol after adding the NFC suspension to the pulp slurry and mixing for 2 min at 600 rpm. The NFC loadings in the suspension were 1.0%, 1.5%, and 2.0% relative to the mass 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, Japan) and a cylinder dryer (Daeil Machinery Co., Ltd., Daejeon, South Korea), respectively.

The sheets were conditioned at 23 °C and 50% relative humidity to maintain their moisture content at 8%. Bulk (TAPPI T411 om-10 2010), tensile strength (TAPPI T494 om-06 2006), and burst strength (TAPPI T403 om-10 2010) were measured to identify the effects of the NFC on the physical properties of the sheets. The light scattering behaviors of the sheets were measured using a spectrophotometer (Elrepho, Lorentzen & Wettre, Stockholm, Sweden) to determine the bonding area between the cellulosic fibers.

RESULTS AND DISCUSSION

Characteristics of BKP Fibers Pretreated by Enzyme

The EN-NFC were prepared by enzyme pretreatment and micro-grinding. To perform the enzyme pretreatment using an endo-glucanase, the effect of the enzyme on the physical properties of the Ba-BKP and Hw-BKP fibers should be analyzed, and comparison of the enzyme pretreatment process with the refining counterpart is necessary, with both methods employing the valley beater as a pretreatment device before micro-grinding at the lab scale (Lee and Mani 2017; Lee *et al*. 2018).

Table 4. Effect of Pretreatment on the Characteristics of BKPs

| Pulp	Measurement	Refining without Enzyme		Enzyme Addition (% of oven-dried fibers)				
		Control	Beaten	0.1	0.5	1.0	2.0	5.0
Ba-BKP	Fiber length (mm)	1.39 (±0.01)	1.03 (±0.04)	1.31 (±0.03)	0.61 (±0.03)	0.51 (±0.01)	0.39 (±0.01)	0.36 (±0.01)
	Fiber width (μm)	17.7 (±0.07)	17.6 (±0.21)	17.3 (±0.07)	18.2 (±0.21)	18.9 (±0.21)	19.3 (±0.31)	19.4 (±0.35)
	Fines (%)	15.6	19.3	14.1	22.4	26.7	31.5	33.5
	Crystallinity (%)	75.9	75.7	79.6	80.6	81.5	81.8	81.9
Hw-BKP	Fiber length (mm)	0.67 (±0.01)	0.59 (±0.01)	0.61 (±0.01)	0.37 (±0.01)	0.34 (±0.00)	0.32 (±0.00)	0.27 (±0.00)
	Fiber width (μm)	18.9 (±0.07)	18.8 (±0.14)	18.9 (±0.07)	19.0 (±0.21)	19.2 (±0.42)	19.3 (±0.49)	20.0 (±0.76)
	Fines (%)	8.9	14.9	12.0	27.1	28.8	31.9	39.8
	Crystallinity (%)	80.2	80.7	82.5	82.6	83.6	84.1	84.6

The changes in the BKP fibers pretreated with endo-glucanase were measured, and the results are summarized in Table 4. When the enzyme was added to the BKP fibers, the length of the BKP fibers decreased. Even at 0.1% enzyme loading, which was the lowest addition, the fiber length decreased by 5.7% for Ba-BKP and 9.0% for Hw-BKP. When the enzyme loading was increased to 5.0%, the fiber length was ultimately reduced by 74.1% for the Ba-BKP and 59.7% for the Hw-BKP. In contrast, as the enzyme loading was increased, the fiber width increased slightly and ultimately increased by 9.6% for the Ba-BKP and 5.8% for the Hw-BKP. It has been reported that enzymatic hydrolysis proceeds at or in between the layers of the cell wall of fibers and leads to swelling and peeling (Arantes *et al.* 2014). Increase in the average value and standard deviation of the fiber width of BKPs pretreated with endo-glucanase indicate that the swelling process which is the stage before peeling might have proceeded. The Ba-BKP fibers appeared more swollen than the Hw-BKP fibers because the Ba-BKP fibers were composed of nine or more microfibril layers (Huang *et al.* 2016; Lian *et al.* 2020). Fiber swelling is advantageous for fiber collapse during nanonization by the micro-grinder (Seo *et al.* 2016). These changes in the fiber length and fiber width could also be confirmed visually, as in Figs. 2 and 3.
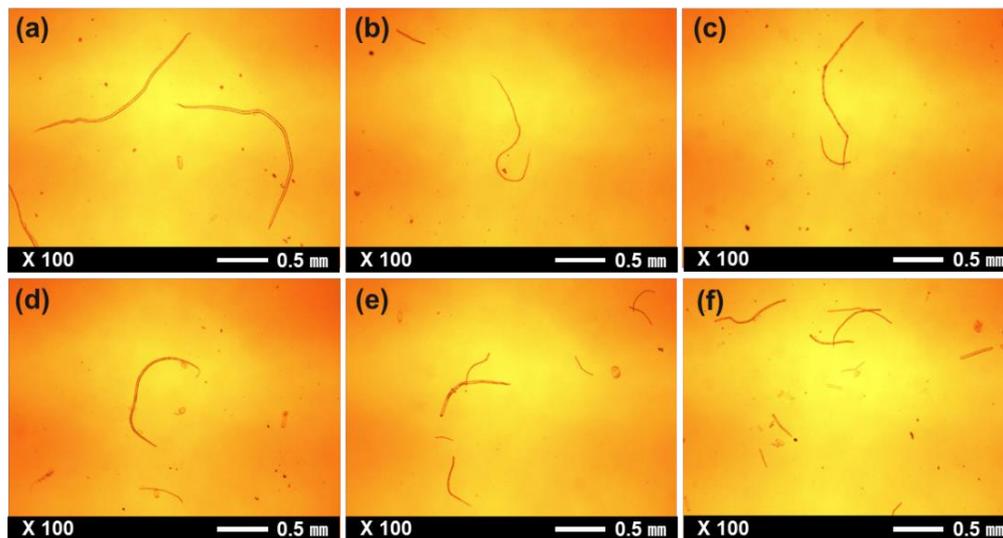
Fig. 2. Microscope images of Ba-BKP fibers pretreated with (a) beater and (b) 0.1%, (c) 0.5%, (d) 1.0%, (e) 2.0%, and (f) 5.0% of endo-glucanase of oven-dried fibers

As the enzyme loading was increased, the fiber length decreased, and many short fibers were generated. The proportion of fine fibers of 0.2 mm or less increased from 15.6% to 33.5% for the Ba-BKP and from 8.9% to 39.8% for the Hw-BKP. The short fibers produced by enzyme hydrolysis had almost no fibrillation and had a smooth surface, unlike the fibers treated with the beater. The crystallinity of the enzyme-pretreated BKP fibers increased depending on the enzyme loading because cellulase attacks and removes amorphous regions in the fibers (Kumar and Wyman 2010; Kim *et al.* 2017). When the BKPs were beaten to 450 mL CSF, the fiber length of the Ba-BKP decreased by 25.9%, and that of the Hw-BKP decreased by 11.9%. The fines contents of the BKPs increased according to the beating treatment, but the fiber widths and crystallinities of the BKPs did not change appreciably. During enzyme pretreatment, the length of the BKP fibers decreased, but their width, fines content, and crystallinity increased. As beating proceeded,

the length of the BKP fibers decreased, and the fines content increased, whereas the fiber width and crystallinity were not appreciably affected. If EN-NFC are used to replace those prepared by beating pretreatment, it was estimated that an enzyme-to-fiber ratio of 0.5% or greater is required to reduce the fiber length for efficient mechanical treatment.
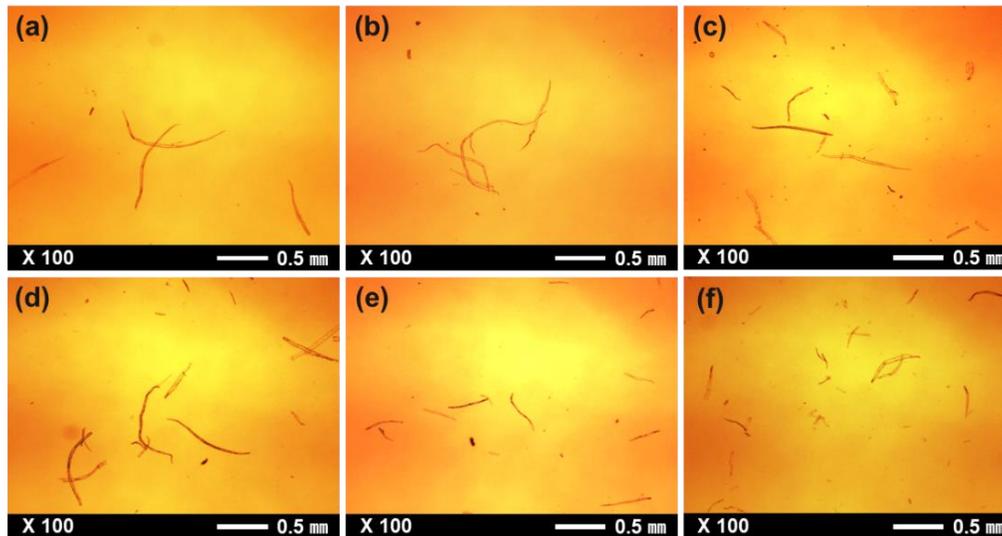

Fig. 3. Microscope images of Hw-BKP fibers pretreated with (a) beater and (b) 0.1%, (c) 0.5%, (d) 1.0%, (e) 2.0%, and (f) 5.0% of endo-glucanase of oven-dried fibers

Characteristics of the NFC Made of Ba-BKP and Hw-BKP

The particle sizes of the NFC were measured to determine the final pass number for micro-grinding to make real nanofibrils from the BKPs using a laser scattering-based particle size analyzer. The particle sizes of the NFC were not direct indicators of the fiber length or fiber width, but the changes in the sizes of the NFC with mechanical treatment could be indirectly inferred from the particle size measurements. Figures 4 and 5 show the average particle sizes of the NFC made from Ba-BKP and Hw-BKP depending on the enzyme loading and the pass number for micro-grinding.
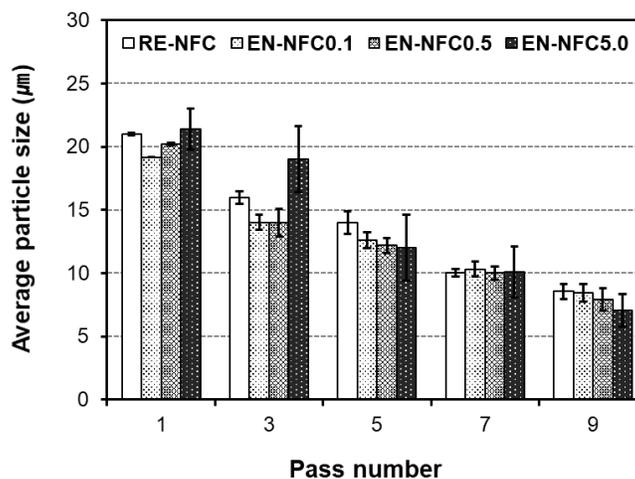
Fig. 4. Average particle sizes of NFC made from Ba-BKP depending on the enzyme loading and the pass number for micro-grinding
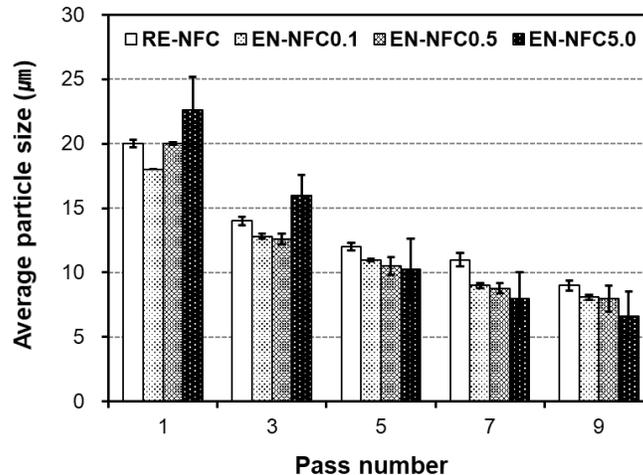
Fig. 5. Average particle sizes of NFC made from Hw-BKP depending on the enzyme loading and the pass number for micro-grinding

As the pass number of fibrillation increased, the average particle size of the NFC decreased. When the pass number was 5 or greater, most of the EN-NFC had an average particle size less than that of RE-NFC, irrespective of the pulp type. When the pass number was 9, the average particle size of all NFC was less than 10 μm, and the differences in the particle sizes of all NFC prepared for each condition were not significant. Therefore, it is reasonable that a final pass number for fibrillation of 9 is adequate for producing nano-sized fibrils from Ba-BKP and Hw-BKP.

The Ba-BKP and Hw-BKP pretreated with enzymes at loadings of 0.1%, 0.5%, and 5.0% of oven-dried fibers were ground nine times using a micro-grinder, and the fiber width of NFC was measured from FE-SEM images of NFC using 3D imaging software. Because nanofibrils are defined as nano-sized fibers that are less than 100 nm wide or micro-size fibers with nano-dimensional cross-sectional structures (Chinga-Carrasco 2011; Isogai *et al*. 2011), the fiber width measurement is the first step for confirming complete conversion of the BKPs to NFC through enzyme pretreatment and micro-grinding processes. Figure 6 shows the average fiber width of NFC depending on the enzyme loading and the pulp type. The fiber widths of all the NFC prepared in this study were less than 100 nm, which showed that nine micro-grinding passes were sufficient for preparing the nano-sized fibrils. Meanwhile, there were no notable differences in the fiber widths of the NFC according to the pulp type. Figures 7 and 8 show the fiber width distributions of the NFC made from the Ba-BKP and Hw-BKP, respectively. The RE-NFC had a greater fiber width than the EN-NFC due to the larger portion of nanofibrils in the range of 50 nm to 100 nm. As the enzyme loading increased, smaller nanofibrils were produced. A distribution curve expressed as mean value and standard deviation of the fiber width measured supported this result. As the amount of enzyme increased, the x-axis of the graph moved to the left and showed a narrow shape. Notably, the portion of EN-NFC5.0, having the lowest average fiber width, was the highest of those in the range of 10 nm to 30 nm. The NFC prepared with Ba-BKP and Hw-NFC showed the same fiber width distributions under the same pretreatment conditions and micro-grinding. Figure 9 shows FE-SEM images of the RE-NFC and EN-NFC. All NFC showed lower fiber width than 100 nm, and there was no significant difference in morphology according to pretreatment and pulp type.
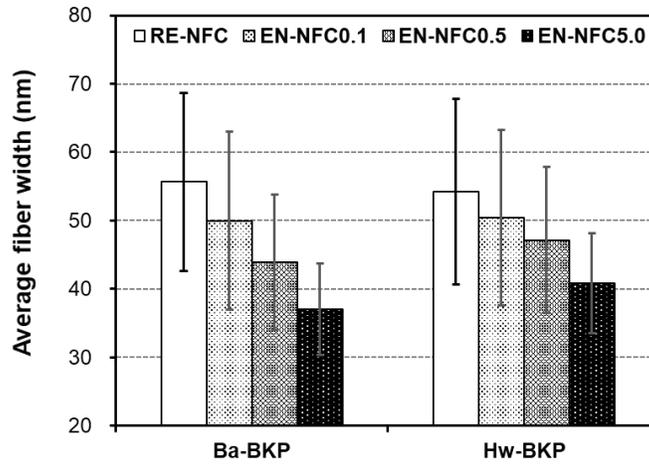
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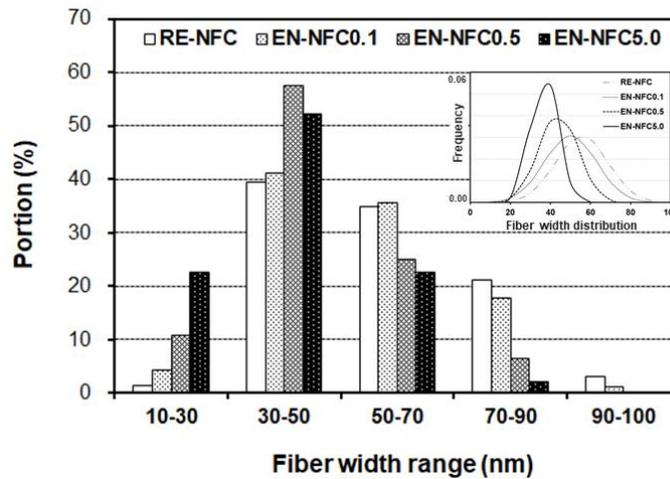**Fig. 6.** Average fiber widths of NFC made from BKPs depending on the enzyme loading

Fig. 7. Fiber width distributions of NFC made from Ba-BKP depending on the enzyme loading
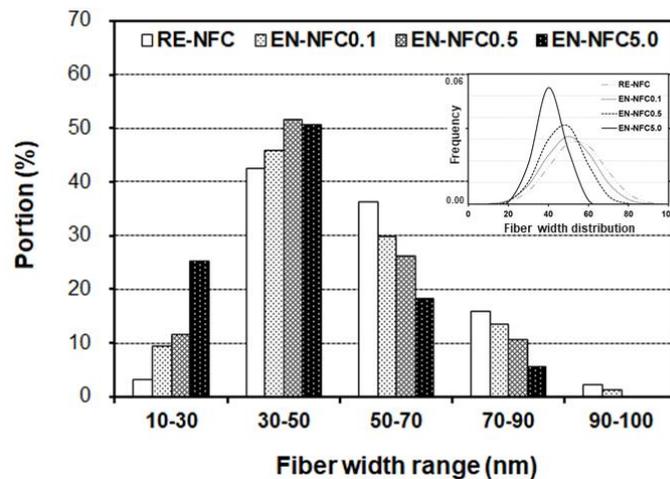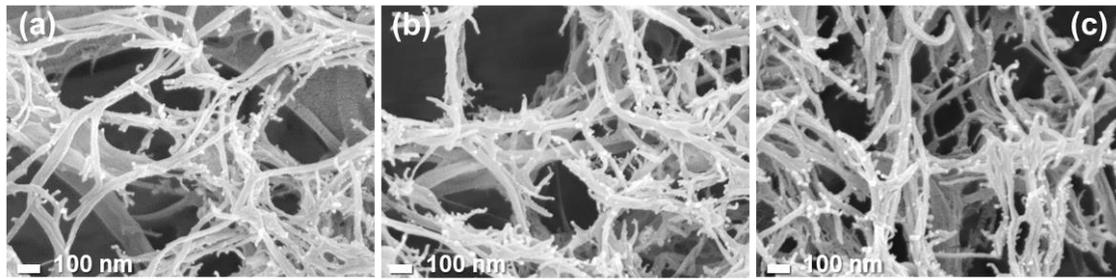

Fig. 8. Fiber width distributions of NFC made from Hw-BKP depending on the enzyme loading

Fig. 9. FE-SEM images of (a) RE-NFC and EN-NFC made from (b) Ba-BKP and (c) Hw-BKP

Therefore, enzyme pretreatment was more effective for the preparation of small and uniform NFC from Ba-BKP and Hw-BKP than the refining pretreatment, and the physical properties of the NFC made from Ba-BKP were similar to those of the NFC made from Hw-BKP.

Evaluation of the Physical Properties of Sheets Containing NFC

Sheets were made with the Hw-BKP for lab-scale preparation of model paper containing the NFC and to investigate the effects of the EN-NFC, made from the Ba-BKP and Hw-BKP, on the physical properties of the sheets, including tensile index, burst index, bulk, and light scattering. Figures 10 and 11 show the effects of EN-NFC0.5 on the tensile indices and the burst strengths of the handsheets. As the loading of EN-NFC0.5 increased, the tensile and burst indices of the sheets linearly increased. The improvement in the strength of the handsheets is a direct consequence of the increases in specific surface area, sheet density, and fiber-fiber bonds (Brodin and Eriksen 2015; Kim *et al*. 2019). The bulk values and light-scattering coefficients of the handsheets decreased depending on the loading of EN-NFC0.5, as shown in Figs. 12 and 13, which supported the mechanism of strength enhancement by EN-NFC0.5.

Comparing the strength enhancement effects of the EN-NFC made of the two types of pulp, the physical properties of the handsheets did not show any significant differences at the same EN-NFC0.5 loading.
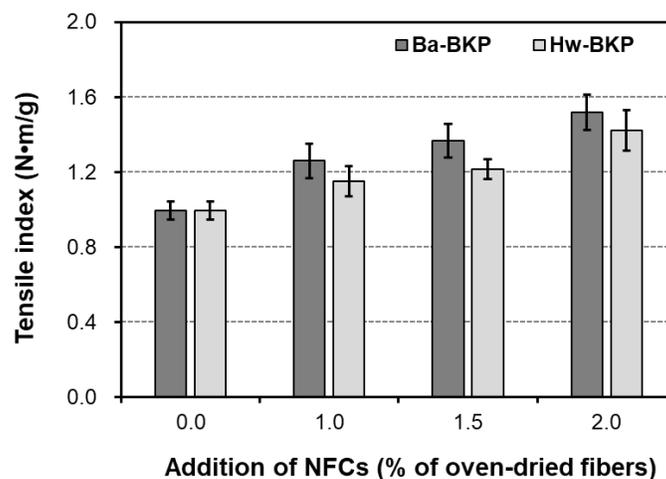
Fig. 10. Effect of EN-NFC0.5 made from Ba-BKP and Hw-BKP on the tensile index of sheets
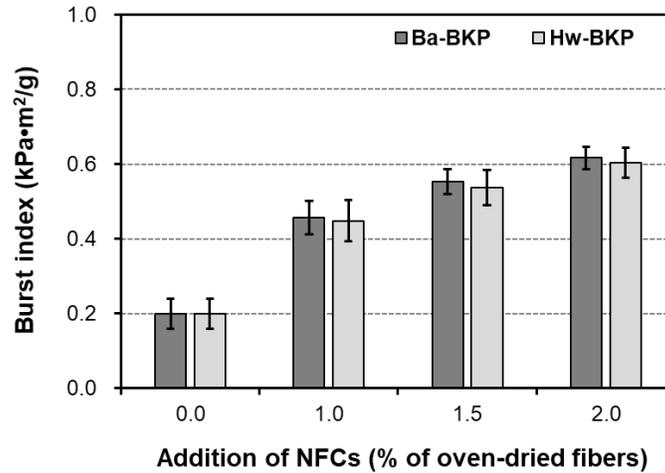
Fig. 11. Effect of EN-NFC0.5 made from Ba-BKP and Hw-BKP on the burst indices of sheets
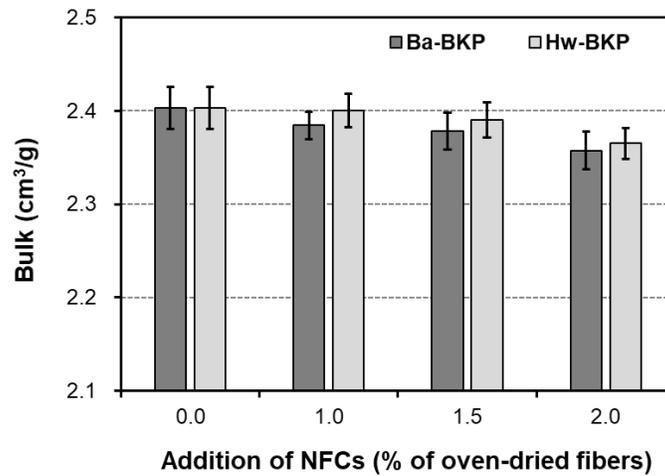

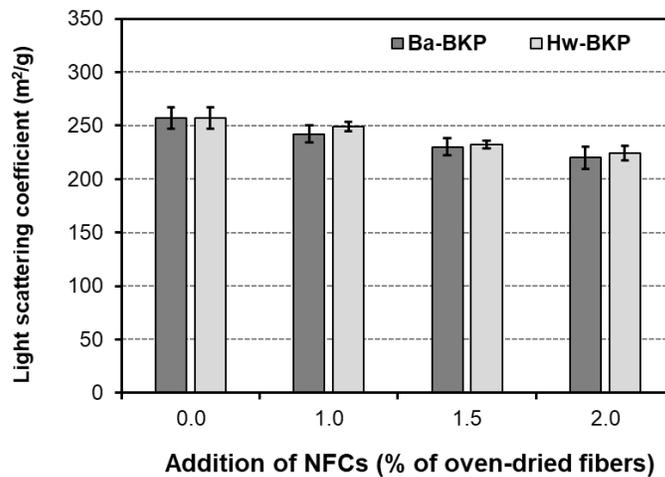**Fig. 12.** Effect of EN-NFC0.5 made from Ba-BKP and Hw-BKP on the bulk values of sheets

Fig. 13. Effect of EN-NFC0.5 made from Ba-BKP and Hw-BKP on the light-scattering coefficients of sheets

The physical properties of the EN-NFC from the Ba-BKP and the Hw-BKP, subjected to the same enzyme pretreatment and micro-grinding, were the same. Thus, the physical properties and strengths of the sheets containing the EN-NFC made from the Ba-BKP and Hw-BKP were also similar.

Therefore, Ba-BKP can be used for the production of NFC having similar physical properties to NFC made from Hw-BKP. The NFC made from Ba-BKP or Hw-BKP are expected to confer similar levels of strength promotion when used as paper strength enhancers.

CONCLUSIONS

1. Endo-glucanase was effective for reducing the fiber length of Ba-BKP for manufacturing EN-NFC. If enzyme pretreatment is used to replace refining pretreatment, 0.5% or greater of endo-glucanase should be added to the Ba-BKP slurry to reduce the fiber length to the level of the Ba-BKP fibers treated by refining.

2. Enzyme pretreatment was more efficient in the preparation of small and uniform NFC from Ba-BKP and Hw-BKP than the refining pretreatment, and the average fiber widths and distributions of the EN-NFC made from Ba-BKP were similar to those of the EN-NFC made from Hw-BKP.

3. The strength of the sheets increased in proportion to the loading of the EN-NFC made from Ba-BKP and Hw-BKP. The bulk values and light scattering coefficients of the sheets decreased depending on the loading of the EN-NFC, which supported the increases of fiber-fiber bonds and sheet density. The sheets containing the EN-NFC made of Ba-BKP and Hw-BKP had similar physical properties.

4. Therefore, Ba-BKP can be used as a raw material for the manufacture of EN-NFC with similar physical properties and paper strength enhancement properties at those made from Hw-BKP.

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