

Effects of Pretreatment on the Dispersibility of Bamboo Pulp Fiber Suspensions for Nanocellulose Preparation

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Industrial production efficiency of nanocellulose by mechanical homogenization was directly affected by dispersibility of pulp suspensions. The bamboo pulp was pretreated by oxidation using 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO), enzymatic hydrolysis, and refining to study dispersibility of the pulp suspensions. Physical morphology and surface charges of the pretreated pulp fibers were analyzed to explain the differences of dispersibility. Multiple light scattering results showed that TEMPO oxidized pulp fibers and refined pulp fibers had good dispersivity, while the pulp fibers treated with cellulase hydrolysis exhibited comparatively lower dispersibility. The TEMPO oxidized pulp fibers had high carboxylate contents and high absolute value of Zeta potential. The dispersibility of the fibers could be improved by dispersants, and the maximum dispersion of fibers from enzymatic hydrolysis was obtained with 0.5% carboxymethylcellulose as dispersant.

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INTRODUCTION

Wood cellulose is a fibrous material composed of fibrils of several microns in length, which are arranged in a spiral pattern and form a hierarchical structure (Chaker and Boufi 2015). Leveraging the hierarchical structure of cellulose fibers, it is possible to isolate nanocellulose through mechanical fibrillation process. Currently, various mechanical processing methods enable nanocellulose isolation. For instance, high-pressure homogenizers disintegrate lignocellulosic fibers *via* high-intensity shear forces, which disintegrates the cell wall and disrupts the hydrogen-bond network, thereby achieving conventional mechanical production of nanocellulose. Unfortunately, so many problems can be found in the preparation of nanocellulose by high-pressure homogenization, such as the high energy consumption (Rol *et al.* 2017; Ang *et al.* 2019) and frequent clogging during the disintegration process (Khalil *et al.* 2014; Nechyporchuk *et al.* 2016; Baati *et al.* 2017). Moreover, the different pretreatments of raw material have an important influence on the preparation of nanocellulose by high-pressure homogenization.

Many scholars are currently focusing on using different pretreatment techniques to improve the efficiency of preparing nanocellulose by mechanical method (Du *et al.* 2016; Zhuo *et al.* 2017; Sharma *et al.* 2019). In the research of Saelee, the steam explosion method and a xylanase-base enzyme were used in the pretreatment process, and the final

cellulose nanofibrils primarily ranged from 5 to 10 nm in diameter were isolated at 15,000 psi for 30 passes by high-pressure homogenization (Saelee *et al.* 2016). Mechanical pretreatment is beneficial to the fibrillation of nanocellulose by high-pressure homogenizer. Ang *et al.* (2019) found that the most heavily treated fibers having a median diameter of 12 nm and aspect ratio of 229 compared to the least treated fibers with a median diameter of 31 nm and aspect ratio of 102. A great number of studies have shown that oxidation by 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO) as pretreatment before homogenization processes were favorable for the preparation of nanocellulose (Gamelas *et al.* 2015; Lu *et al.* 2017; Wu *et al.* 2017).

Furthermore, the clogging of cellulose fibers in high-pressure homogenizer is the key factor to limit the industrial production of nanocellulose, which hasn't been completely solved yet (Nechyporchuk *et al.* 2016). According to Lindström (2017), the clogging phenomena are related to the aggregation of fibers. The bagasse cellulose was initially pretreated with an ionic liquid (1-butyl-3-methylimidazolium chloride) and then the homogeneous solution was passed through a high-pressure homogenizer without any clogging (Li *et al.* 2012). The aggregation of fibers can be observed by the dispersibility of fibers in aqueous solution. Therefore, it is necessary to explore fibers dispersion in aqueous solution to avoid the clogging of high-pressure homogenizer.

Thus, three pretreatment methods (TEMPO oxidation, enzymatic hydrolysis, and refining pretreatment) are commonly used in the preparation of nanocellulose by high-pressure homogenizers were selected as the exploration conditions in this study. Dispersion stability of the pretreated pulp suspensions was observed by multiple light scattering. The microscopic morphology, surface charge, and group of the pretreated pulp fibers were analyzed to explain the differences of fiber dispersions. Finally, the dispersion stability of the enzymatic pulp suspension was improved by adding three kinds of dispersants.

EXPERIMENTAL

Materials

The bamboo bleached kraft pulp ($11 \pm 1^\circ\text{SR}$) used as the raw material was provided by Fuyang Huabaozhai Co., Ltd. (Hangzhou, China). Celluclast® 1.5 L was produced by Novozymes and purchased from Jiejin Biology Corporation (Suzhou, China). The 2,2,6,6-tetramethylpiperidiny-1-oxyl (TEMPO), sodium bromide (NaBr), and sodium hypochlorite (NaClO) solution with active chlorine content of 14 wt% were purchased from Aladdin Industrial corporation (Shanghai, China). The polyethylene oxide (PEO, average molecular weight 2000000) and the carboxymethylcellulose (CMC, the degree of substitution was 0.7, the molecular weight was 250000 and the viscosity was 600-3000 mPa·s) were dissolved in hot water and prepared for dispersant solution (Jiaxing, China). All other chemicals used were of analytical grade and used without further purification.

Methods

Pretreatment by TEMPO oxidation

The bamboo bleached kraft pulp was oxidized according to the method (Saito *et al.* 2006) with minor modifications. The bamboo pulp (5 g, based on dry pulp) was suspended in the distilled water (375 mL) containing TEMPO (0.0625 g) and sodium bromide (0.625 g) under continuous stirring during the reaction time. The target reaction time was achieved until the constant pH was observed. During the reaction process, pH was maintained at

10.5 by adding 0.5 M NaOH solution. Finally, the TEMPO-oxidized fibers were repeatedly washed by centrifugation with deionized water until the filtrate became neutral and then stored at 4 °C before further treatment and analysis.

Pretreatment by enzymatic hydrolysis

The bamboo pulp was enzymatically hydrolyzed using the method (Akimkulova *et al.* 2016) with minor modifications. The bamboo pulp (4 g) was enzymatically hydrolyzed with a cellulase loading of 6 FPU/g substrate for 16 h in a 250 mL conical flask by a 150-rpm air-bath shaker under the pH and temperature at 4.8 (0.05M sodium acetate buffer) and 50 °C, respectively. After the reaction process was finished, the enzymatic hydrolysate was placed in boiling water for 15 min and the enzymatic fibers were washed and stored at 4 °C for further analysis.

Pretreatment by PFI refining

The bamboo pulp (30 g) was mechanically pretreated using a PFI mill at 10 wt% in accordance with the TAPPI T248 (2021) standards. The PFI mill refiner was stopped when the degree of beating reached 90 °SR. After PFI refining, the bamboo pulp fibers were collected and stored at 4 °C for further analysis.

Dispersion stability of enzymatic pulp fibers

The enzymatic pulp was formulated into a fiber suspension of 1% concentration and stirred uniformly. The PEO and CMC were added into the hot water as dispersants and stirred at 2500 rpm using a magnetic stirrer until the dispersants were completely dissolved. Different concentrations of dispersants solution were added into enzymatic pulp suspensions with even stirring. Experiments without addition of dispersants were used as the control.

Analytical methods

The dispersion stability of the pulp suspensions was determined by using Turbiscan Lab Analyzer (Formulation, France). Surface microscope of pulp fibers was observed with scanning electron microscope (JSM-IT300, JEOL, Japan). Carboxylate content of the pretreated pulp fibers was assayed according to TAPPI T237cm-98 (2006) standard; Chemical functional groups of pulp fibers were analyzed by FTIR spectroscopy (IR Prestige-21, Shimadzu, Japan) in the range of 400 to 4000 cm⁻¹; and Zeta potential of pulp fibers were measured using zeta potentiometer analyzer (B390, AFG, Germany).

RESULTS AND DISCUSSION

The Dispersion Stability of Pretreated Pulp Fibers

Dispersion stability of fibers in solution is an important factor in the effective application of cellulose fibers. Multi-light scattering technique enables the qualitative and quantitative determinations of dispersion. Turbiscan stability index (TSI) is a comprehensive metric used to quantify the stability of dispersion systems (*e.g.*, emulsions, suspensions), where higher TSI values correlate with poorer system stability and lower values indicate enhanced stability (Goscianska *et al.* 2019). Figure 1 presents that the TSI of three pretreated fiber suspensions were changed during 30 min. The TSI of the pulp fiber suspension in the PFI pulping pretreatment process was the lowest, indicating that the

fibers were dispersed most uniformly in the solution. The stability of the enzymatic fibers was much lower than refining and TEMPO oxidation pretreatment. The dispersion stability of TEMPO-oxidized fibers was slightly lower than that of the refining pretreatment fibers, while the enzymatic fibers had the worst dispersibility. The TSI curve of the enzymatic fibers rises very fast while the upward trend became slow after 400 s. It indicates that the sedimentation of enzymatic fibers mainly occurs in the initial 400 s and the remaining fine fibers in the upper solution begin to settle slowly with time going on. The dispersion stability of colloidal systems depends on a number of factors, including the particle size, surface area, surface charge, Van der Waals force and electrostatic force (Goscianska *et al.* 2019). In order to explain the difference of the dispersion stability of the pulp fibers after various pretreatments, the fiber morphology and surface charge were explored in the subsequent work.

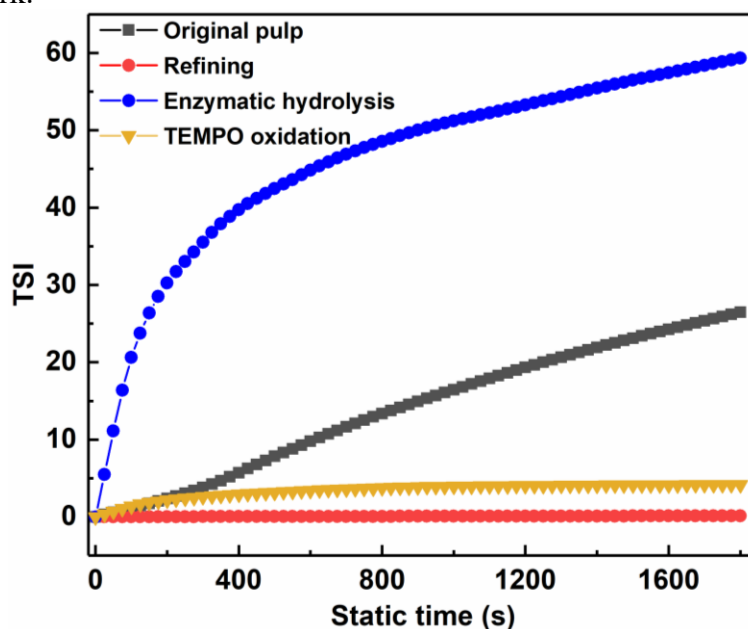


Fig. 1. Dispersion stability of the pretreated pulp fibers by multiple light scattering method

Morphology Analysis of Fibers

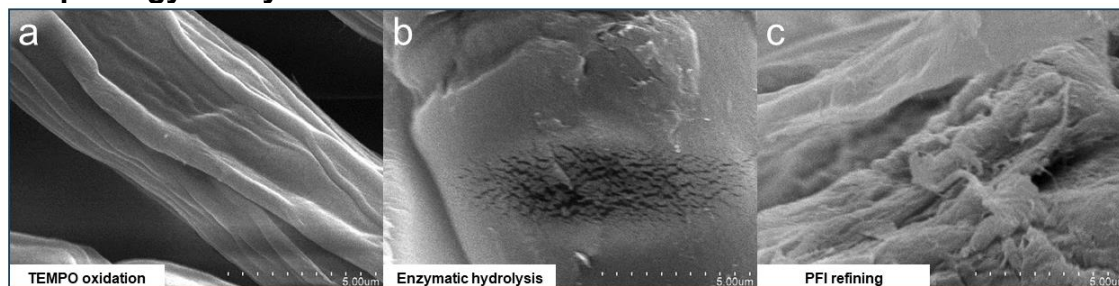


Fig. 2. SEM images of the pulp fibers with different pretreatments: (a) TEMPO oxidation, (b) enzymatic hydrolysis, and (c) PFI refining

In Fig. 2, smooth wrinkles on the TEMPO-oxidized pulp fibers surface, cracks on the enzymatic pulp fibers surface, fibrillations on the refined pulp fibers surface were observed, respectively. The TEMPO oxidation only modified the surface of fibers, both fibrous forms and microfibrillar nature of the original native celluloses were maintained after the TEMPO-mediated oxidation (Saito *et al.* 2006; Saito and Isogai 2006). Previous

works had shown that the cutting of enzyme-mediated fibers predominantly occurred at the disordered regions of cellulosic fibers (Clarke *et al.* 2011; Thygesen *et al.* 2011). Hence, the long fibers were cut into short fibers by enzymatically hydrolysis. However, the mechanical refining could lead to fiber splitting by the mechanical friction between two disks in the grinder, which were shown in Fig. 2c. Furthermore, the action of fibrillation involved loosening of the fibrils and raising of the finer microfibrils on the surface of the fibers, which resulted in a large increase in surface area for the refined pulp (Bhardwaj *et al.* 2004). The dispersion stability of the fibers in aqueous solution might be closely related to fibers morphology. Large surface area of the pulp fibers facilitated to reduce the precipitation in water caused by gravity.

FTIR Analysis

Fourier transform infra-red spectroscopy is a useful tool in elucidating the functional groups of organic macromolecules, as well as understanding the bonding interactions of these functional groups (Wong *et al.* 2009). Figure 3 depicts the FTIR spectra of pulp fibers pretreated by TEMPO oxidation, refining, and enzymatic hydrolysis. The FTIR spectra of the pulp fibers subjected to TEMPO oxidation show similar bands assigned to others pretreated pulp fibers, which indicates that the functional groups of pulp fibers after the three pretreatments were the same. The broad absorption band observed in the 3433 cm^{-1} is related to O-H stretching vibrations of the adsorbed water. It also indicates the vibrations of -COOH groups (Goscianska *et al.* 2016; Goscianska *et al.* 2017). The bands at 1625 cm^{-1} are assigned to C=O stretching vibration of carboxyl and ketone groups (Álvarez *et al.* 2015; Tanaka *et al.* 2015; Goscianska *et al.* 2016). The presence of the carboxyl group was illustrated by infrared spectra. Although the detection process resulted in slightly different peak strengths for the three fibers, the peak intensity at 1625 cm^{-1} in the spectrum of TEMPO oxidized sample is significantly higher than that of the other fibers. It indicates that the oxidation of TEMPO introduced a large number of carboxyl groups on the fibers surface.

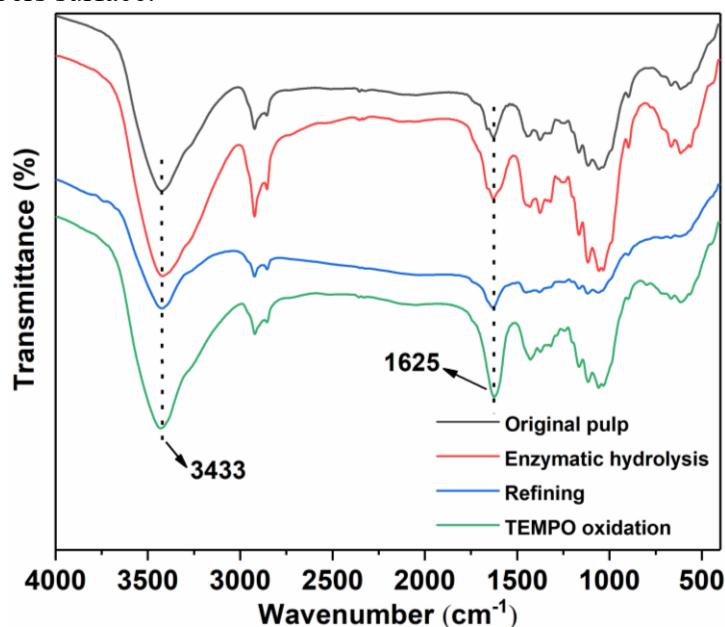


Fig. 3. FT-IR spectra of fibers following three pretreatment methods

Surface Charges Analysis of Fibers

The surface charge of the pulp fibers after different pretreatments were determined using zeta potentiometer analyzer and conductometric titration. Table 1 shows the zeta potential and carboxylate contents of the fibers after three pretreatment methods. The relative levels of zeta potential for different pulp were similar, and the higher the zeta potential meant the higher was the carboxyl group contents. The zeta potential of the TEMPO oxidation pulp was -77.9 mV, and the carboxyl group contents was 13.15 mmol/g. It is mainly because the C6 primary hydroxyl group of the cellulose fibers could be selectively oxidized and converted into carboxyl groups in the TEMPO/NaClO/NaBr oxidation system (Song *et al.* 2015). After TEMPO oxidation pretreatment, the presence of carboxyl groups on the surface of fibers leads to a certain repulsive effect among fibers, which can achieve better fiber dispersion. The zeta potential of the fibers after refining pretreatment was -31.6 mV and the carboxylate contents was 2.69 mmol/g. Refining is a process of physical disintegration that changed the morphology and structure of the fibers, which increased the surface area of the fibers. It caused more carboxylate contents to be revealed and more negative charges (Bhardwaj *et al.* 2004). For PFI pulping pretreatment, the absolute values of zeta potential and the content of carboxyl groups are moderate, and the dispersion stability of fibers in the suspension solution is the best. It can be seen that the quantity of charge is not the only the factor affecting the fiber dispersion performance; the surface morphology and structure of fibers are also important factors affecting fiber dispersion. It can be speculated that the microfibrillation and fibrillization on the surface of PFI pulping pretreated fibers enhance the dispersion stability of fibers in the suspension solution. After the biological enzyme pretreatment of fibers, the absolute value of surface charge was low (zeta potential = -0.7 mV; carboxyl content = 0.79 mmol/g), and the surface morphology is relatively dense, which led to the dispersion stability of fibers in the suspension solution was the worst.

Table 1. Zeta Potential and Carboxyl Content of the Pretreated Pulp Fibers

Pretreatment Methods	Zeta Potential (mV)	Carboxyl Content (mmol/g)
TEMPO oxidation	-77.9±2.21	13.15±1.27
Refining	-31.6±1.98	2.69±1.52
Enzymatic hydrolysis	-0.71±0.17	0.79±0.19

Improvement of the Enzymatic Pulp Dispersion

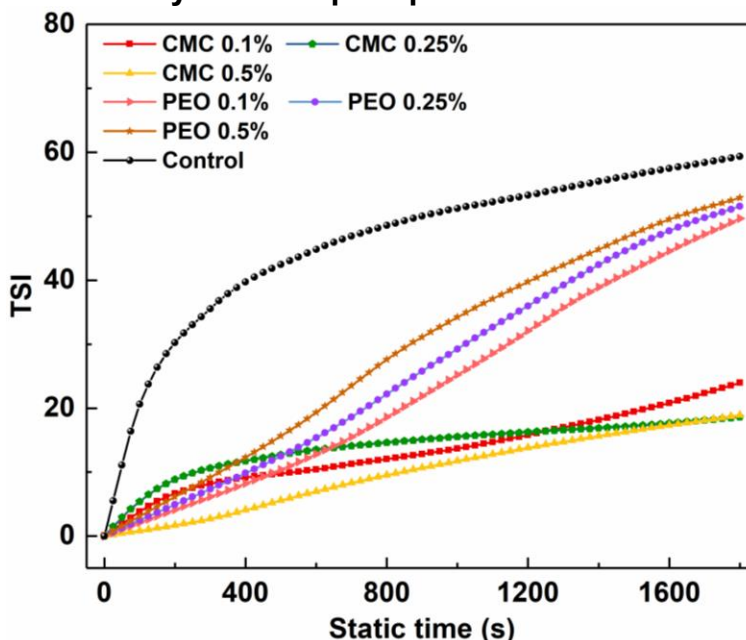


Fig. 4. Effect of different dispersants on the dispersion stability of the enzymatic pulp

The poor dispersion stability of the enzymatic fiber suspension had a great negative impact on its application. So, CMC and PEO were used to improve the dispersion stability in this study. Figure 4 shows the trend of TSI values of the enzymatic fibers over time after adding different dispersants. The effectiveness of dispersants has been proved by the determination of the dispersion stability. The good dispersion stability of enzymatic fibers suspensions was obtained with addition of CMC and PEO. The effect of CMC was better than that of PEO, it may be due to the excellent adsorption ability of CMC on the enzymatic fibers surface (Lin *et al.* 2015).

CONCLUSIONS

1. The dispersion stabilities of pretreated pulp fibers were studied. The refined fiber had the best dispersion stability, followed by the TEMPO oxidized fiber, and the worst is the enzymatic fibers.
2. Fibrillations on the refined fibers were observed while the TEMPO oxidized fiber and the enzymatic fibers were smooth. Moreover, the TEMPO oxidized fibers had the most carboxylate contents of 13.15 mmol/g and the highest zeta potential of -77.9 mV, while that of enzymatic fibers were 0.79 mmol/g and -0.7 mV, respectively.
3. CMC and PEO could improve the stability of enzymatic fibers suspensions, and the effect of CMC was better than that of PEO.

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