Nanocellulose Preparation from Sugarcane Bagasse and Its Application for Paper Sizing

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ABSTRACT: *The goal of this study was to fully utilize sugarcane bagasse, an abundant residue from the Vietnamese sugar industry that is also known as a potential source of lignocellulose. The biomass material was used as a raw material to produce cellulose pulp followed by a hydrogen peroxide bleaching process. On the one hand, the unbleached cellulose pulp was used to make paper sheets with basis weights of around 100 g/m^2. The bleached cellulose, on the other hand, was chemically and mechanically transformed into nano cellulose via a limited hydrolysis procedure. The obtained nano cellulose possessed a high crystallinity of 80.11% and was used for paper sizing to improve the mechanical and barrier properties of the paper. The ability of sugarcane bagasse nano cellulose-coated paper sheets to produce biodegradable containers for food and beverage applications was investigated.*

KEYWORDS: *Barrier properties; Biodegradable containers; Nanocellulose; Sugarcane bagasse.*

INTRODUCTION

By 2025, global waste generation is estimated to be over 6 million tons per day [1]. Consumer waste has increased more than tenfold over the past century, from 40 kg to 560 kg of waste per person, per year. Currently, up to 5 trillion plastic bags are consumed annually all over the world [2]. Most plastics used in the world today are single-use items causing a massive volume of waste that is disposed to the environment. During the degradation of plastic products, greenhouse gases such as methane and ethylene are released contributing to climate change [3]. According to Plastic Oceans [4], it is estimated that 10 billion tons of plastic are thrown into the oceans. As a consequence, microparticles are found in all mussels tested and plastic pollution has killed 1 million marine animals each year. This has raised a great awareness about how humans treat the environment.

Paper cups, which appear to be environmentally friendly items, are difficult to recycle. The main reason comes from the fact that these cups are mostly coated with plastic, aluminum, or other non-decomposable polymers. Paper and paperboard themselves are biodegradable. However, these coating layers make them more problematic to compost and recycle because they become stickies during the recycling process causing serious operational issues.

The use of renewable biopolymers as an alternative to the common petroleum-based synthetic polymers in temporary food and beverage items has been studied for decades. These biopolymers are primarily derived from natural sources such as polysaccharides, proteins, and lipids. The goal of this work was to create a self-biodegradable paper-based material for food and beverage

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applications that can replace single-use plastic items. In this study, Sugarcane Bagasse (SB) was used as the raw material, both for paperboard and nanocellulose production. This is a plentiful and inexpensive material as it is a by-product of the sugar industry in Vietnam. According to *Brunerová et al.* [5], about 35% of the entire sugarcane stem is bagasse, which is generated during the sugar production process. As a result, approx. 1 million tons of SB is produced every year making this an abundant and popular source for utilization. On the other hand, sugarcane bagasse is a fibrous material with a high content of cellulose (41–55 wt%) and hemicellulose (20.0–27.5 wt%), and relatively low content of lignin (18.0–26.3 wt%) [6–9]. Currently, sugarcane bagasse is primarily burned for energy generation, which has a negative impact on the environment. As a result, the utilization of this residue may benefit the country's long-term development.

Although nanocellulose has been studied for decades, it remains an intriguing material with many promising features. According to its structure and dimensions, nanocellulose is categorized into three types: (i) Cellulose Nanocrystals (CNCs), (ii) Cellulose Nanofibrils (CNFs), and (iii) Bacterial Cellulose (BC) [10].

Compared to traditional materials, nanocellulose possesses distinct properties, such as high surface area, rheological properties, mechanical reinforcement, and barrier properties [11]. This leads to the fact that nanocellulose has a wide range of applications in the paper industry and packaging [12–15], biomedicine [11, 16, 17], environment [18–20], and a variety of other fields. A high-performance nanocellulose-based composite that was resistant to oil and grease was produced by *Tyagi et al*. [21]. In their study, CNC was combined with nano-filler montmorillonite clay and alkyl ketene dimer as a surface-active agent. The formed material demonstrated promise as a coating barrier for packaging. They stated that employing CNC-composite coatings considerably reduced water absorption (up to 71% compared to surfaces without coating and up to 27% for surfaces with a CNC-only coating). Air permeation resistance, on the other hand, was 88% higher when compared to uncoated surfaces. Water-resistant 'nanopaper' was also produced by coating lactic acid-modified cellulose nanofibers [22]. In this study, cellulose nanofibers were modified with lactic acid *via* ultra-sonication and oligomerization. The storage modulus of the 'nanopaper' has been reported to be three times greater than that of the reference paper. Thermal stability was also demonstrated by the modified 'nanopaper' when compared to the reference one.

CNCs and CNFs, unlike BC, are derived from biomass such as agricultural residues. As previously stated, sugarcane bagasse is a potential lignocellulosic source that can be utilized to produce cellulose. Several methods can be used to achieve nanocellulose materials. While NFCs are commonly produced by mechanical methods such as ultra-sonification, homogenization, and grinding [23, 24], CNCs were synthesized by using a variety of methods, including high-pressure homogenization [25], alkaline-acid sequence hydrolysis [26], chemo-mechanical treatment [27], and, more commonly, concentrated acid hydrolysis [28, 29]. In an acidic environment, the amorphous regions are dissolved, revealing the crystal regions. Sulfuric acid is the most commonly used reagent in the fabrication of CNCs [30]. Aguiar *et al.* [31] produced CNC from sugarcane bagasse and straw via enzymatic hydrolysis. According to their findings, the nanocellulose material had approx. 70% crystallinity and thermal stability up to 300ºC. Another method for obtaining CNC from sugarcane bagasse mentioned in a review of *Khoo et al.* [19] was cellulose purification followed by acid hydrolysis. Compared to other methods, acid hydrolysis results in cellulose nanomaterials with higher crystallinity. CNCs derived from acid hydrolysis of SB have crystallinity indices ranging from 56 to 74% with a yield of up to 58% [30]. A similar procedure for producing CNCs was also performed by *Dien et al.* [32]. In their study, rice straw was subjected to a combined treatment method that included acidic hydrolysis in combination with hydrogen peroxide and a refining process. The procedure resulted in the nanocellulose with fibers diameter distribution mainly in the range of 14–26 nm when performed at 150ºC, for 180 min.

To summarize, current approaches for producing nanocellulose from sugarcane bagasse primarily required concentrated sulfuric acid, making it more difficult to recover and scale-up. In addition, monosaccharides formed during the hydrolysis were degraded further under the strong acid condition. As a result, nanocellulose produced was dark in color and difficult to purify.

This work focused on the total utilization of sugarcane bagasse. To begin, cellulose pulp was produced by regular alkaline pulping and bleaching processes. The bleached cellulose was converted to CNC using the two-stage process

mentioned by *Dien et al*. [32]. The obtained SB nanocellulose was then applied to the surfaces of paper sheets that were made from bleached cellulose pulp of the same source. Mechanical properties and water absorptiveness of the sheets were finally evaluated to assess their ability to contain liquid and food.

EXPERIMENTAL SECTION

Materials

Sugarcane bagasse (SB) used in this study was a byproduct of a sugar extraction process at Lam Son Sugar Joint Stock Corporation (Thanh Hoa, Vietnam). Ten kilograms of sugarcane bagasse were selected at random from the manufacturer's waste pile. The raw material was subjected to a pretreatment procedure that included cleaning, air drying, and sorting before it was used. For chemical composition analysis, the air-dried material was ground to the size of 0.25–0.50 mm.

The chemicals used, including sodium hydroxide, hydrogen peroxide, sulfuric acid, were all analytical grades and originated from Sigma Aldrich.

Determination of the chemical composition of the raw material

The cellulose content of the sugarcane bagasse was determined by Kürschner and Hoffer method [33]. The material was treated with a 1-to-4 volume ratio mixture of concentrated nitric acid (density 1.42 g/mL) and absolute ethanol. Substances other than cellulose were hydrolyzed and dissolved in the strong acid environment. The remaining insoluble residue was weighed to determine the cellulose content using Eq. (1).

$$C_{sb} = \frac{m_{ir}}{m_{sb}} \times 100 \quad (1)$$

Where: C_{sb} (%) is the cellulose content of sugarcane bagasse; m_{ir} (g) is the oven-dried mass of the insoluble residue; m_{sb} (g) is the oven-dried mass of the taken sugarcane bagasse.

Other chemical compositions of SB including lignin, pentosans, and ash contents were determined according to TAPPI standard methods T222 om-11, T223 cm-10, and T211 om-02, respectively.

Cellulose isolation and bleaching

Sugarcane bagasse was subjected to a common alkaline pulping process to produce cellulose pulp, which was then used for the fabrication of paper sheets and nanocellulose. About 150 g (oven-dried mass) of SB was placed in a 3.5-L reactor, together with an appropriate volume of cooking solution to achieve a solid-to-liquid ratio of 1-to-15. The cooking medium was made up of sodium hydroxide and sodium sulfide solutions with an active alkali of 25%. The reaction mixture was heated to approx. 100 ± 5ºC and kept at that temperature for 180 min. The obtained cellulose pulp was thoroughly washed and refined with a laboratory pulper before dewatering and subsequently used for papermaking. The yield of the alkaline pulping, also known as the yield of unbleached cellulose pulp, was calculated according to Eq. (2).

$$Y_p = \frac{m_p}{m_{sb}} \times 100 \quad (2)$$

Where: Y_p (%) is the yield of cellulose pulp; m_p (g) is the oven-dried mass of pulp obtained from the alkaline pulping; m_{sb} (g) is the oven-dried mass of sugarcane bagasse used in the alkaline pulping.

Bleached cellulose pulp with a brightness of approx. 80% ISO was achieved by following a 2-stage hydrogen peroxide bleaching procedure. The bleaching process was carried out at 80ºC for 2 h with a pulp consistency of 10 wt%; H_2O_2 and NaOH dosages were 5 wt% and 1.5 wt% over the oven-dried mass of the pulp, respectively. The bleached cellulose was then used to prepare nanocellulose.

Nanocellulose preparation

The procedure for nanocellulose preparation from sugarcane bagasse is illustrated in Fig. 1. Each experiment was carried out with approx. 50 g of the bleached SB pulp in a 1000-mL autoclave that was placed in a glycerol bath equipped with a temperature control unit. The material was first hydrolyzed with an aqueous mixture of 0.25 wt% sulfuric acid and 0.1 wt% hydrogen peroxide solutions. In all cases, the pulp consistency was held constant at 10 wt%, while the volume composition of the H_2SO_4 and H_2O_2 solutions in the mixture was varied to determine the most suitable reaction conditions. The reaction temperature and time were set at 140ºC and 90–180 min, respectively. After that, the product was purified with a dilute solution mixture of 0.1 wt% hydrogen peroxide and 0.05 wt% sodium hydroxide. The mixture was then washed with distilled water before being gently refined for 2 min with a laboratory OSAKA® multifunctional cutter (model DH-807, 350W, fixed rotation of 2500 rpm).

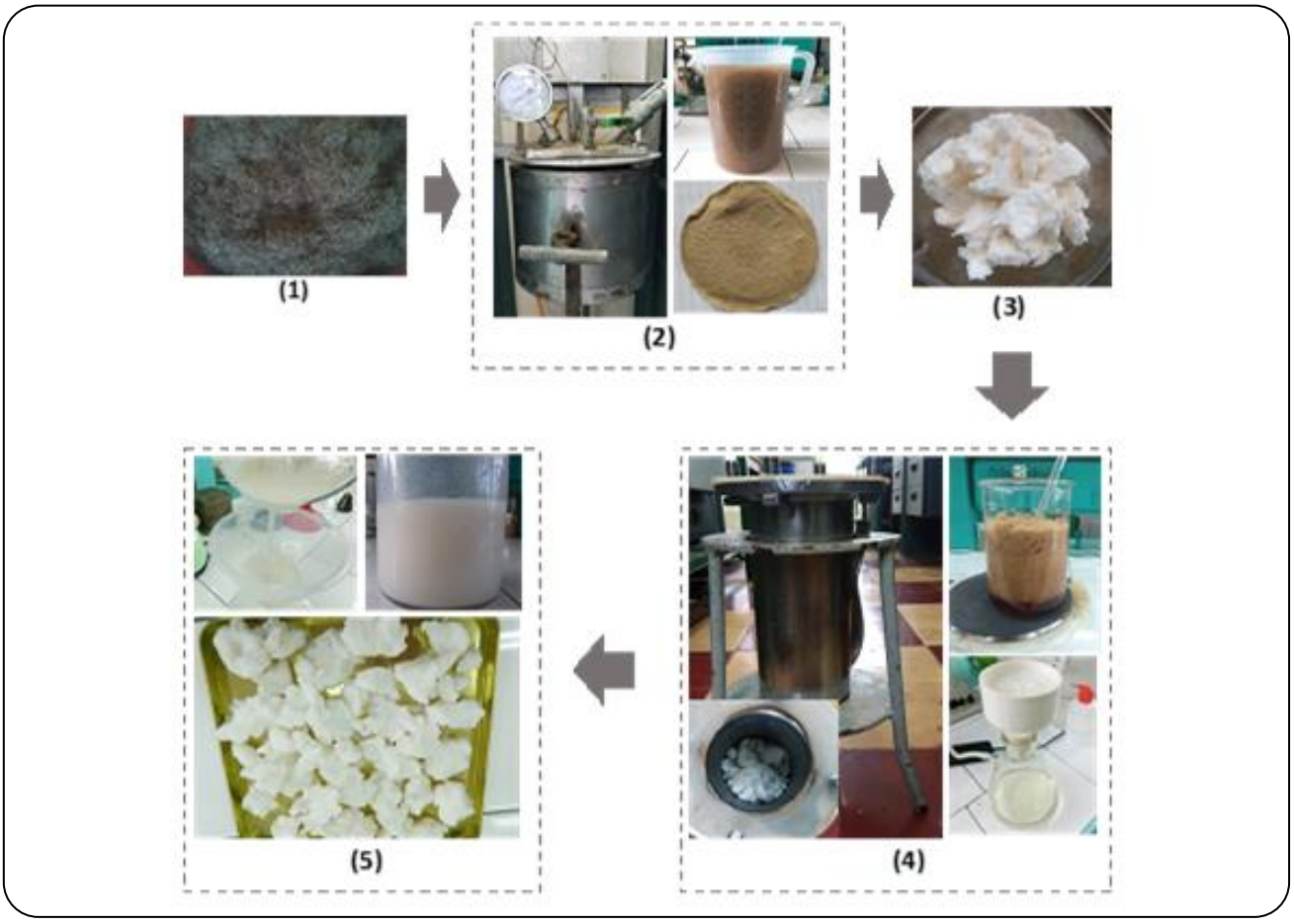


Fig. 1: Production of nanocellulose from sugarcane bagasse; (1) Sugarcane bagasse, (2) Cellulose isolation and unbleached cellulose pulp, (3) Bleached cellulose, (4) Cellulose hydrolysis and purification, (5) Nanocellulose refining and sample preparation

The treatment was carried out to gain a homogeneous suspension of nanocellulose that did not precipitate after a long period of storage (i.e., more than 48 h).

After the nanofabrication steps, nanocellulose was centrifuged to determine the yield. The yield of nanocellulose was simply determined by drying a certain volume of the nanocellulose suspension at 105 ± 5ºC until the net weight remained unchanged.

Papermaking and sizing

Paper sheets with a basis weight of around 100 g/m^2 were first made from the bleached SB cellulose on a laboratory paper-making machine. Sizing solutions with a concentration of 6.0 wt% were prepared from starch and the synthesized nanocellulose in various ratios of dry solid. The sizing solutions were applied to both sides of the paper using a laboratory-scale sizing machine.

Characterization of sugarcane bagasse cellulose and nanocellulose

The morphology structure of SB cellulose was characterized using an FEI 450 NANO SEM scanning electron microscope. The structure of SB nanocellulose was investigated with a JEOL JSM-7600F field emission scanning electron microscope, an X-Ray Diffractometer Bruker D5005, and a SHIMADZU 1S Fourier Transform InfraRed (FT-IR) spectrometer. Statistical size distribution of nanocellulose was generated based on its Scanning Electron Microscopy (SEM) images using NIS-Elements BR (Nikon) software.

Based on the X-Ray Diffraction (XRD) pattern, the crystalline index (I_C) of cellulose and nanocellulose was calculated according to Eq. (3) [28].

$$I_C(\%) = \left[\frac{I_{200}-I_{110}}{I_{200}}\right] \times 100 \quad (3)$$

where: I_{200}, I_{110} are the intensity values of the peaks at $2\theta = 22.5^{o}$ and 18.7^{o}, respectively.

Evaluation of mechanical and barrier properties of paper sheets

The basis weight of each paper sheet was determined, and the sizing weight was calculated accordingly. Physical properties of the sheets, including tensile strength and burst index, were measured according to TAPPI standard T 220 sp-01. Water absorptiveness via Cobb value was tested and calculated using ISO 535:2014 standard.

RESULTS AND DISCUSSION

Chemical compositions of sugarcane bagasse

The composition of sugarcane bagasse varies slightly depending on species and pedological features but ranges from 35-50% cellulose, 20-30% hemicellulose, 15-29% lignin, and around 2-7% ash [34]. Major chemical components of SB used in this work are listed in Table together with their contents. The material contains 44.02% cellulose, 18.70% pentosan, and 25.49% lignin. Around 5.2% ash, which comes from inorganic compounds, is typical for annual plants such as rice or sugarcane. With a relatively high cellulose content, SB is a promising source for producing many value-added products.

The material also has a hollow structure with many pores as shown in Fig. 2A below. Because of this feature, sugarcane bagasse is easily treated with NaOH and then with acid to remove amorphous regions and reveal crystalline regions.

Production of sugarcane bagasse cellulose and nanocellulose

Under the cooking conditions mentioned in the section *"Cellulose isolation and bleaching"*, the yield of cellulose pulp from SB was 63.6%. The obtained pulp had the Kappa number of 10.4 (residue lignin content is 1.46%). After the bleaching process, the cellulose got its brightness of 80.9 % ISO with a yield of 57.4% over the oven-dried mass of SB.

Microstructures of SB raw material, SB-derived unbleached, and bleached cellulose were characterized through their SEM images, which are presented in Fig. 2.

As seen from Fig. 2, the structure of SB has been changed significantly after being processed at a low temperature. According to Fig. 2A and B, the cell walls are connected to each other but still show hollow structure due to the presence of typical pith in its stalk. After the alkaline treatment, components other than fibers have been decomposed, revealing a fibrous structure (Fig. 2C). In addition, almost all of the lignin was dissolved after the cooking process, resulting in thinner and more unconsolidated fibers. However, there is almost no difference in the surface between unbleached and bleached SB cellulose (Fig. 2C, D).

Table 1: Chemical compositions of sugarcane bagasse.

No.	Component	Content (%)
1	Cellulose	44.02
2	Lignin	25.49
3	Pentosans	18.70
4	Ash	5.19
5	Others	6.60

Additional information about cellulose before and after bleaching, as well as the resulting nanocellulose, is presented in Fig. 3. A broad peak at a wavelength of 3410 and 2950 cm^{-1} are assigned to the stretching vibration of the OH and CH group in cellulose chains [28, 29, 35, 36]. Furthermore, the presence of lignin is indicated by peaks observed at 1620 and 1260 cm^{-1}, which are associated with the aromatic ring and the C-O stretching vibration of the aryl group, respectively [29, 37]. Peaks in the range of 1350-1250 cm^{-1} are typical for chemical groups in hemicellulose [35, 36]. Typically, peaks at 900-890 cm^{-1} signal the CH_2 groups of cellulose [28, 35, 36]. This suggests that the alkaline cooking and hydrolysis, which were both performed under mild conditions, did not completely dissolve lignin and hemicellulose. As a result, the resulting nanocellulose has a similar structure to the bleached cellulose and the initial cellulose.

The addition of hydrogen peroxide to the reaction promotes nano-fibrillation and makes the resulting nanocellulose easier to purify and bleach after the hydrolysis. This was demonstrated by running an experiment at 140ºC for 90 min with a 0.5 wt% H_2SO_4 solution only (i.e., without H_2O_2). The results show that the nano-fibrillation was successful, but the resulting fibers are large and precipitated after only a short time in storage (data not shown). In addition, because contaminants were not removed, the obtained nanocellulose was yellowish. This issue also appeared

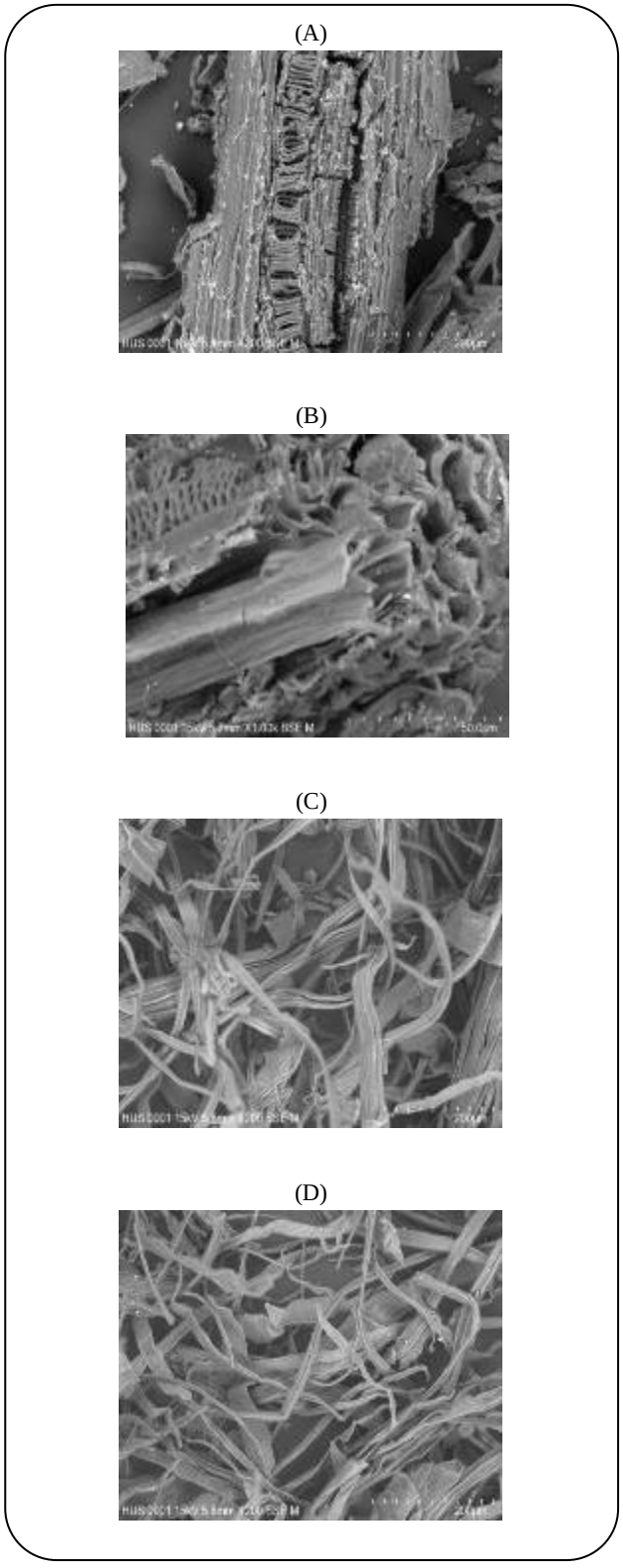


Fig. 2: Scanning electron micrographs of sugarcane bagasse (SB) raw material and cellulose; (A) and (B) Sugarcane bagasse (200x and 1.000x magnification, respectively), (C) Unbleached SB cellulose; (D) Bleached SB cellulose.

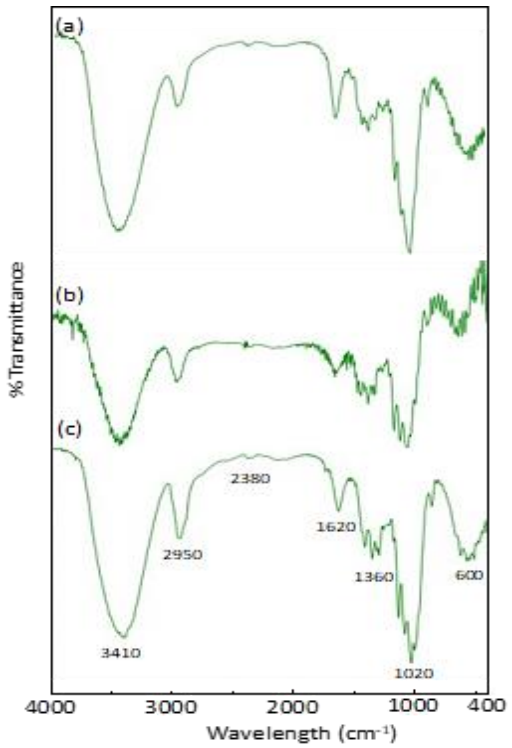


Fig. 3: FT-IR spectra of (a) unbleached cellulose; (b) bleached cellulose; and (c) nanocellulose from sugarcane bagasse.

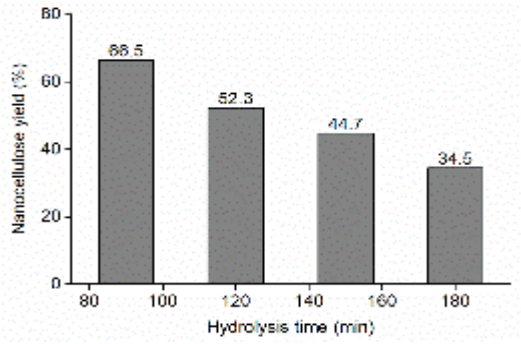


Fig. 4: Influences of hydrolysis time on the yield of sugarcane bagasse nanocellulose

when we tried to produce nanocellulose from other raw materials such as *Acacia mangium* wood, rice straw, bamboo, or tapioca residue without using hydrogen peroxide.

The influences of hydrolysis time on SB nanocellulose yield are illustrated in Fig. 4. The yield of nanocellulose after 90 min of hydrolysis was 66.5% over the oven-dried

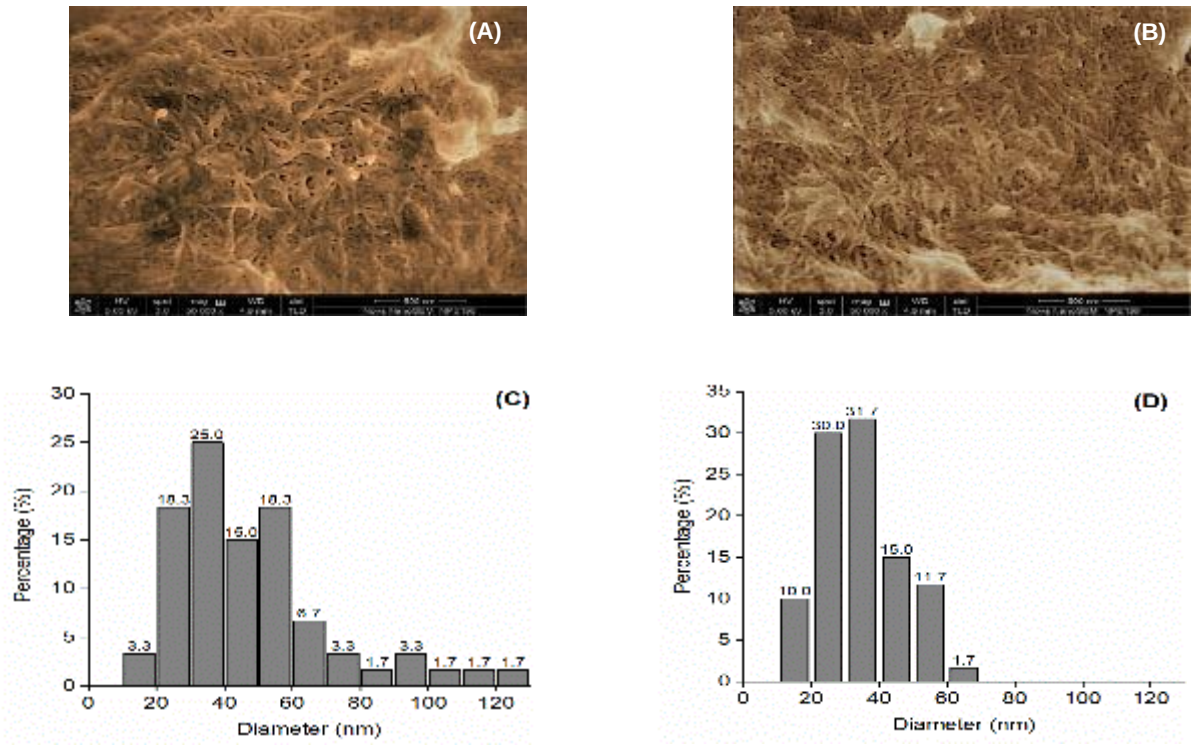


Fig. 5: Scanning electron micrographs of sugarcane bagasse nanocellulose obtained after 90 min (A), 120 min (B) of hydrolysis and the corresponding size distribution diagrams (C&D)

mass of the bleached cellulose, or 38.2 % over the oven-dried mass of SB. This indicates that cellulose is highly hydrolyzed, and this value is comparable to that of rice straw [32]. Obviously, increasing the reaction time reduces the nanocellulose yield significantly. After 180 min, the yield was only 34.5 %. The drop in nanocellulose yield is thought to be due to the emergence of a plateau in the cellulose conversion process. When the acid hydrolysis is prolonged, the created nanocellulose is further hydrolyzed, producing several by-products such as glucose, levulinic acid, and formic acid [38, 39]. The nanocellulose yield can be improved by conducting the hydrolysis at lower temperatures or for a shorter period of reaction time, depending on the expected dimension of nanocellulose.

The fibrous structure (rod-like) of the nanocellulose obtained from sugarcane bagasse is visible in SEM images shown in Fig. 5A and B. Additional information obtained from statistical data presented in Fig. 5C and D reveals that the average diameter of SB nanocellulose is in the nano-range, with the majority of the diameter distributed around 20-60 nm. According to *Pereira et al*. [30], the material can thus be classified as CNCs. This indicates that the hydrolysis process successfully converted sugarcane bagasse cellulose into nanomaterial.

Fig. 6 depicts XRD patterns of the bleached SB cellulose and the corresponding nanocellulose. The typical structure of cellulose-I is indicated by the two strong peaks observed at $2\theta = 17.8^{o}$, 22.5^{o}, which correspond to (110) and (200) planes, respectively [40–43]. It is important to note that the (110) plane represents the amorphous region in the cellulose/nanocellulose material, while the (200) plane represents the whole region, including both crystalline and amorphous areas. A moderate peak located near 35^{o} corresponds to (400) plane, which is also typical of cellulose I structure but is not the dominant contributor [40].

According to Eq. 3, the Crystalline Index (CI) increases from 72.44% for the bleached SB cellulose to 80.11% for the SB nanocellulose, which is in agreement with previous works [28, 44, 45]. This demonstrates that the acid hydrolysis has destroyed the amorphous areas in cellulose, revealing more crystalline regions. The CI of SB nanocellulose obtained in this work is comparable to that of the rice straw nanocellulose produced by *Dien et al.* [32] (80.11% vs. 82.3%). The SB nanocellulose has a relatively

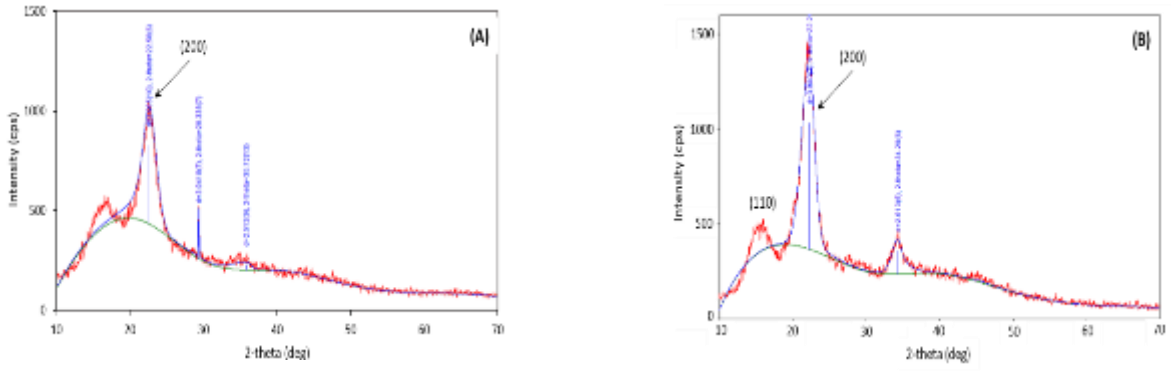


Fig. 6: X-ray diffractograms of bleached cellulose (A) and corresponding nanocellulose (B) from sugarcane bagasse.

high CI, which is typical for nanocellulose derived from annual plants and is expected to be advantageous in the production of materials with high tensile strength and thermal stability [46].

Application of sugarcane bagasse nanocellulose for paper sizing

Paper sheets coated with SB nanocellulose that were prepared as mentioned in the section "*Paper making and sizing*", have sizing mass ranging from 0.9 to 1.1 g/m^2. Tensile strength and burst index of the sheets were evaluated in comparison with control samples, which were sized solely with starch, i.e., without the addition of SB nanocellulose.

Cobb sizing test was used to determine the sheets' water resistance. A high Cobb value means that the material can absorb and retain moisture. A low Cobb value, on the other hand, indicates that the material can withstand moisture penetration and retention. Changing the ratio between nanocellulose and starch in the sizing solution affects the Cobb value and mechanical properties of the sheets. According to the data on water absorption shown in Fig. 7 and mechanical properties presented in Table 2, the more nanocellulose used in the sizing solution, the higher tensile strength and higher Cobb value are obtained. This suggests that nanocellulose improves the strength of paper but decreases the water resistance ability. This is confirmed by many published works as reviewed by *Sharma et. al.* [14].

Cellulose nanofibrils were found to be able to compensate for the reduction of paper strength. Nanocellulose together with cationic starch in coating formulations is believed to increase the mechanical properties of paper and printing properties. This is

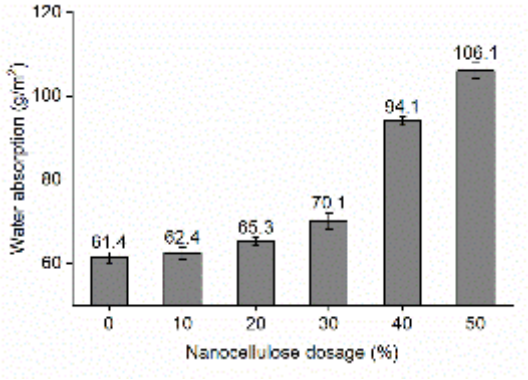


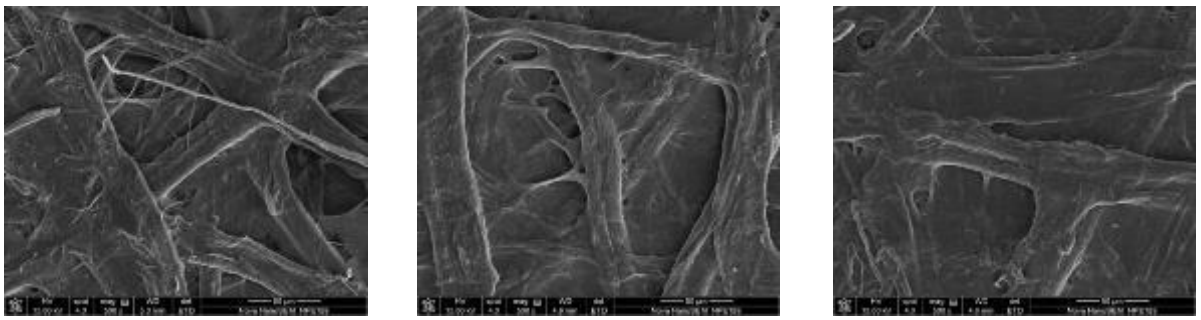
Fig. 7: Water absorption of paper sheets coated with different percentages of sugarcane bagasse nanocellulose in the sizing solution.

demonstrated by the tensile and bursting strength of the paper sheets coated with sizing mixtures containing different amounts of SB nanocellulose (Table 2). Obviously, the addition of SB nanocellulose improves tensile and bursting strength when the dosages of nanocellulose range from 10 to 40 wt%. When the amount of SB nanocellulose in the sizing mixture is increased to 40-50 wt%, the tensile strength tends to decrease, while the burst index is still raising slightly but insignificantly. This could be because SB nanocellulose has a lower retention capacity than starch. Considering the water resistance ability and mechanical properties of the paper sheets, the suitable percentage of nanocellulose in the sizing solution is recommended to be from 20 to 30 wt%.

Fig. 8 presents SEM images of paper sheets with and without SB nanocellulose coating, revealing some intriguing changes in the paper structure. After being sized, spaces in the uncoated paper were filled with the nanomaterial, making the sheets smoother and flatter.

Table 1: Tensile strength and bursting index of sugarcane bagasse nanocellulose coated paper sheets.

No	SB nanocellulose (%)	Basis weight (g/m^2)	Tensile strength (N/m)	Burst index (kPa)
1	Control	101.5	4926.4 ± 40.5	407.2 ± 6.1
2	10	102.4	4942.5 ± 40.5	430.0 ± 7.1
3	20	101.4	4954.5 ± 48.4	417.5 ± 6.4
4	30	101.9	5257.0 ± 47.6	443.0 ± 7.1
5	40	102.6	5752.5 ± 43.3	448.5 ± 5.0
6	50	101.7	5612.5 ± 48.9	450.0 ± 7.1



(A) (B) (C)

Fig. 8: Scanning electron micrographs of (A) uncoated paper, (B) paper coated with 10% of SB nanocellulose, (C) paper coated with 50% of SB nanocellulose

The more SB nanocellulose used (i.e., 50 wt%), the smoother the surface of the sheet.

CONCLUSIONS

- Nanocellulose with a crystallinity of 80.11% and an average diameter of 20-60 nm has been successfully produced from sugarcane bagasse. This high crystalline index indicates that the obtained nanomaterial belongs to the category of cellulose nanocrystals.
- When being used as a component of an external sizing solution, sugarcane bagasse nanocellulose has the potential to improve the paper tensile strength and barrier properties, which are essential for packaging papers. The amount of nanocellulose in the sizing solution has a significant impact on the water absorption capacity and mechanical properties of the paper sheets.
- Considering the paper's water-resistance and tensile strength, the appropriate percentage of nanocellulose in the sizing suspension of 6 wt% concentration is recommended to be 20-30 wt%. Sugarcane bagasse nanocellulose-sized sheets show good tensile strength of around 5200 N/m, burst index of around 440 kPa, and Cobb values of 65-70 g/m^2. With these properties, the paper sheets demonstrate great potential for use in food and beverage applications.

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