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Morphology, Crystallinity and Thermal Properties of Nanocrystalline Cellulose Isolated of Sisal Fiber by Acid Hydrolysis-Ultrasonication

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ABSTRACT

Nanocrystalline cellulose (NCC) from natural Agave sisalana (Sisal) fibers were isolated using a combination of chemical and mechanical processes. The chemical treatment begins with soaking the fiber in a sodium hydroxide (NaOH) solution with a concentration of 5 wt.% at a temperature of 90°C for 60 minutes. Then following by bleaching (fiber refining) using a hydrogen peroxide solution (H_2O_2) with a concentration of 3 wt.% (weight), at a temperature of 60°C, and pH of 10 for 30 minutes. It aims to eliminate the presence of hemicellulose and lignin contained in the fiber. Fibrillation Micro into nano Sisal fibers using sulfuric acid (hydrolysis process). Sulfuric acid (H_2SO_4) with 55 wt.% at temperature 60°C for 30 minutes produced NCC with a diameter of 5±1 nm (D) and a length of 260±10 nm (L), as seen using a TEM (transmission electron microscope). The web-like network structured shape of NCC results in a high aspect ratio (L/D) value is 52. The acid hydrolysis-ultrasonication process produced a high crystallinity index of 78.82% through the XRD (x-ray diffraction) test. The crystallinity and aspect ratio of NCC show that Sisal fiber is a suitable material as a filler for bio-nanocomposite materials. The maximum temperature (T_{max}) of NCC decreased by 10°C due to sulfate ions attached to the cellulose structure, causing the thermal stability to drop from 348°C to 338°C.

Keywords: *Nanocrystalline cellulose, acid hydrolysis, x-ray diffraction, thermal stability*

1. INTRODUCTION

Natural fibers are widely developed as natural resources for the craft industry. The advantages of natural fibers have been directed as an eco-friendly material. The application of natural fiber is not only for arts. Natural fibers have been widely developed to be used as reinforcement in polymers. These past few years, it has been used to fill composite materials. Plant fibers generally consist of three main chemical constituent parts: cellulose, hemicellulose, and lignin. The hydrophilic properties of nature materials cause poor interfacial bonding between fibers and the matrix. The hydrophilic properties were reduced by chemical modifications such as alkalization, bleaching, and acid hydrolysis. In several previous studies, removing amorphous materials (hemicellulose and lignin) increases a crystallinity index, surface morphological structure, and heat resistance stability. The nanocellulose is the most crucial element for reinforcement on the polymer matrix. The fibrillation process into nanoscale gives new material properties to NCC (Nanocrystalline Cellulose) with unique morphology, non-toxicity, high mechanical strength, width surface area, high crystallinity index, bio-compatibility, bio-degradability, and renewability.

The presence of hydroxyl groups and good morphology from NCC gives excellent intermolecular hydrogen bonds between NCC and polymer matrix. According to previous research by Listyanda et al. [1], NCC isolation from ramie fibre using acid hydrolysis (H_2SO_4) with 44 wt.% concentration successfully produced the needle-shaped NCC with an average diameter of ±6.28 nm and a high a crystallinity index (88.12%).

Jamasri & Yudhanto [2] developed research about the effect of microcrystalline cellulose filler as reinforcement of the glass-jute hybrid laminated composite (HLC) for automotive panel bodies. Adding MCC filler into HLC increased the tensile strength and flexural strength of JGJGJG (Jute-Glass- Jute-Glass Jute-Glass) by 11% and 65%, respectively. The MCC inhibits the initial crack and delamination of the HLC. MCC has a high crystallinity index like the NCC, which generally impacts the mechanical properties of HLC because it better creates the intermolecular bonding between resin polymer and fibers. Sisal fiber is derived from one of the cultivated leaf fiber plants, which thrives in barren lands with limited rainfall, such as those found in Madura provinces, Indonesia.

Table 1 Physical properties Acid hydrolysis treatment from various natural sources

Natural sources	Acid concentration	Morphology	Diameter (nm)	Aspect ratio (L/D)	Crystallinity (%)	References
Cotton fiber	H_2SO_4 50% (wt) at room temp for 4 hours + Sonication 120 watts for 60 min.	Spherical shape	20-100	-	81.2	[4]
Ramie fiber	H_3PO_4 60% (wt.) at 150°C for 90 min. in the oil bath	Needle-like structured	21.4	5-20	88.28	[5]
Jute fiber	H_2SO_4 58% (wt.) at 40°C for 45 min. +Sonication 20 min.	Rope like structured	55±10	800±100	90	[6]
Barangan Banana	H_2SO_4 54.5% (wt.) at 100°C for 30 min.	Rod like structured	25-50	10-20	54-57.3	[7]
Agave Americana	HNO_3 70% (v) and CH_3COOH 80% (v); at 100°C for 30 min.	Filament like structured	15	25-33	70.4	[8]
Agave Cantala	H_2SO_4 44% (wt.) at. 60°C for 30 min. + Sonication 180 watt 30 min.	Web-like structure	45	50	78.2	[9]
Agave Tequilana	H_2SO_4 60 % (wt.) at 60°C for 70 min. + HCl 2N at 50°C for 30 min.	Needle-like structured	8.9-9.1	14-47	79.2	[10]
Agave Sisalana	H_2SO_4 55% (wt.) at 60°C for 30 minutes + Sonication 180 watt for 30 min.	Web-like structured	5	52	78.82	This work

The Sisal properties like high strength, salt resistance, weathering resistance, and good dimensional stability make this fiber widely used. The high cellulose content of sisal fiber is 65-76% cellulose, 10-11% hemicellulose, 8-10% lignin, and 3% wax. This fiber has a diameter of about 80-120 μm [3]. The characteristics of species of Agave plants show cellulose content higher than wood which ranges 50 to 65% cellulose, and small amorphous content [11]. Chemical treatment is the best way to remove the amorphous structure from natural fibers, so the cellulose with a semi-crystalline region has high tensile strength and elastic moduli [12]. Chemical treatment of natural fibers includes alkalization, bleaching, and sulfuric acid hydrolysis.

Nanocellulose is the smallest part of the fiber structure obtained from the fibrillation process. Mechanical and chemical processes are utilized for the fibrillization of fibers from the micro to the nano level. It aims to remove the amorphous region and increase the quantity of crystalline regions. Nanocellulose has many benefits in terms of properties, including high crystallinity, aspect ratio, surface area, and excellent dispersion capabilities, and continues to be developed as a polymer reinforcing filler, additive for biodegradable products, packaging, membrane reinforcement, hybrid nanocomposite reinforcement, drug carrier media, medical implants, and semiconductor materials [13, 14].

Nanocellulose is produced from mechanical processes using tools such as high-speed grinder, cryo-crusher, ultrasonic homogenizer, ultrafine grinder, and high-pressure homogenizer. This automatic process requires a high energy and long-time process, and high costs. Furthermore, another alternative is to use a chemical treatment process that is easy to carry out and less costly. In addition, the chemical treatment process is easier to control the crystallinity index value than the mechanical process. The isolated process of natural fibers must be monitored on crystallinity index (CI), which indicates that the cellulose structure is not damaged due to the treatments. Combining chemical and mechanical isolation processes aims to efficiently process, cost, and get nanocellulose with excellent physical properties [15]. Table 1 shows the report from previous research to

produce the NCC by acid hydrolysis from various natural sources.

2. MATERIALS AND METHODS

2.1. Materials

The Sisal fibers were obtained from Madura province, Indonesia. Sodium hydroxide (NaOH), Hydrogen peroxide (H_2O_2), and sulfuric acid (H_2SO_4), which is a chemical solution for isolation was obtained from Sigma-Aldrich (CV. Wahana Hi Lab) Yogyakarta, Indonesia.

2.2. Extraction of Cellulose from Sisal Fiber

The Sisal fiber was cut into 10 mm pieces, then the extraction process started by immersing the Sisal fiber (micro-cellulose) in NaOH and H_2O_2 solution to reduce the hemicellulose and lignin content in the Sisal fiber. It was immersed in an alkaline solution using NaOH with a concentration of 5 wt.% for 4 hours at room temperature. The fibers were rinsed clean, and the pH returned to normal: pH = 7. This alkaline process continues by soaking the sisal fiber in 3 wt.% H_2O_2 solution for purified cellulose fiber.

2.3. Isolation of Microcrystal Cellulose (MCC)

The isolation of sisal micro cellulose fibers involves a hydrolysis process using sulfuric acid with a concentration of 55 wt.% at a pre-heat temperature of 60°C. The sulfuric acid was dropped by a burette tube with a clamping jig. The sulfuric acid was added drop by drop until the pre-heat temperature was reached. The purified sisal fiber was put into a beaker glass with a fiber suspension ratio of 1:50. The acid hydrolysis ran for 30 minutes and then stopped by immersing the NCC suspension in the ice bath (cold water at 4°C). The NCC suspension was rinsed by a centrifuge at 3000 rpm for 15 minutes repeatedly two times to eliminate the sulfate ion in the suspension. After rinsing the NCC's suspension, still, acid and then it was giving sodium hydroxide at 20 wt.% by dropping slowly gradually, and became neutral (pH of 7), and then rinsed again with a centrifuge at 3000 rpm for 10 minutes repeatedly (four times) to eliminate the Natrium chloride (NaCl) in the suspension. The neutral NCC suspension was homogenized by an ultrasonic homogenizer with a power of 240 watts, for 15 minutes. The scheme procedures of fibrillation of MCC to NCC can be seen in Figure 1.

2.4. Morphology Analysis

Photo Scanning Electron Microscopy (SEM) was performed using JCM-7000 JEOL/MP to observe the purified fiber after pre-treatment. It was coated with platinum in a vacuum bath using an electron beam for five minutes. The sample must be a solid material, and it was operated at 5 kV to achieve excellent image contrast. The micro-scale of the fiber was observed by 50 to 1,000 times magnification. Ten bundle fibers were analyzed to determine the distribution of diameter and length. Photo Transmission Electron Microscopy (TEM) was conducted using JEM-1400 to observe NCC (Nanocrystalline cellulose). The sample must be liquid or gel-like NCC's suspension; it was operated at 100kV. The NCC shape was observed by 10,000- 80,000 times magnification.

2.5. XRD Analysis

The scattering angle peaks and intensities of natural fibers (organic materials) were analyzed using the Rigaku Miniflex-600 X-ray diffractometer, operating at 40 kW, 15 mA, and CuKα radiation (λ=1.54060Å). The micro and nano cellulose samples were scanned in 2θ range from 5° to 40° with a scan speed of 5° min^{-1} and sampling pitch of 0.02°. The powder structure of MCC and NCC have crystal and amorphous structures which can be quantified using the crystallinity index (CI), calculated according to Equation 1.

$$CI = (I_{002} - I_{am})/I_{002} \quad (1)$$

where I_{002} is a crystalline structure plane at a scattering angle on the 2θ=22°, and I_{am} is an amorphous structure at a scattering angle on the 2θ=18°.

Figure 1 The scheme procedure of extraction and isolation on Sisal fiber

2.6. FTIR Analysis

FTIR is an analytical test method used to identify organic polymeric materials. The material composition analysis method by FTIR uses infrared light to scan the sample test and observe its chemical properties. It is used in the analysis to transmit infrared radiation from the sample test by absorbing and passing some of it through radiation. It measured wavelength range from 4000-400 cm^{-1} with a Shimadzu 8400S Spectrometer. Thin pellet samples were prepared with the help of KBr (potassium bromide).

2.7. Thermal Analysis

The TGA (Thermogravimetric Analysis) Mettler Toledo can run on a single temperature program (a constant heat rate at 10 °C/min which ranges from 30 to 600°C through a nitrogen gas flow rate of 60 mL/minute). The 10 mg powder of MCC and NCC was tested for thermal degradation to generate the polymer's degradation at different temperatures. Some examples of mass change processes are decompositions and oxidations. On the other hand, the TGA curve data are coupled with a derivative thermogravimetric (DTG) curve to improve the results.

3. RESULT AND DISCUSSION

3.1. Morphological

The purified Sisal fiber was observed with SEM, which produces a stacked micro-cellulose image (Figure 2a). The purified Sisal is clean and smooth, indicating pure microcrystal cellulose (MCC) without impurities (hemicellulose and lignin). Figure 2b illustrates the dimensions of MCC, showing a diameter (D) of 11.5 µm and a length (L) of 950 µm.

Isolation with a combination of chemical (acid hydrolysis) and mechanical (ultrasonication) processes effectively produced NCC. The NCC has been observed with TEM, which makes the web-like network structure (Figure 3a). The dimension of NCC is 5 ± 1 nm in diameter and 260 ± 20 nm in length, and the aspect ratio (L/D) is 52 (Figure 3b). A high aspect ratio of NCC Sisal is improve mechanical properties of the hydrophilic matrix such as PVA (polyvinyl alcohol), PLA (polylactic acid) and PEO polyethylene oxide use to bio-plastic composite, bio-film, membrane, and hydrogel.

Yudhanto et al. [16] isolated Agave Cantala fiber into NCC through acid hydrolysis-ultrasonication. The NCC with the highest aspect ratio (43.8) reinforces the PVA matrix for bio-plastic application. Adding 8 wt.% NCC to the PVA film increases the tensile strength by 79% and the elongation at the break by 138%, respectively. The addition of NCC can barrier 82% of ultraviolet rays at a low wavelength (350 nm). Another study using fiber from the same genus, namely Agave Americana by Krishnadev et al. [17], isolated this fiber by an alkalization process of 4wt.% at temperature 80°C for two hours, and then bleached using 2wt.% $NaClO_2$ (Sodium Chlorite) at temperature 80°C for four hours. This chemical treatment process was followed by an acid hydrolysis process using 80% nitric acid (HNO_3) and 70% acetic acid (CH_3COOH) at 100°C for 30 minutes. It continuous by ultrasonication for 30 minutes. It is procedure produces NCC with a diameter of 18.2 ± 10.14 nm. The crystallinity index increases from 50.1% (raw fiber) to 64.1% (NCC). The thermal stability of fiber increases from 331˚C (raw fiber) to 351°C (NCC). Rosli et al. [18] investigated NCC from another species of Agave, namely Agave Angustifolia, with the same pre-treatment chemical process and then it is continuous with fibrillation by acid hydrolysis using 60 wt.% H_2SO_4 at 45°C for 45 minutes without ultrasonication can produce NCC with a diameter of 15 nm and a crystallinity index of 82 %.

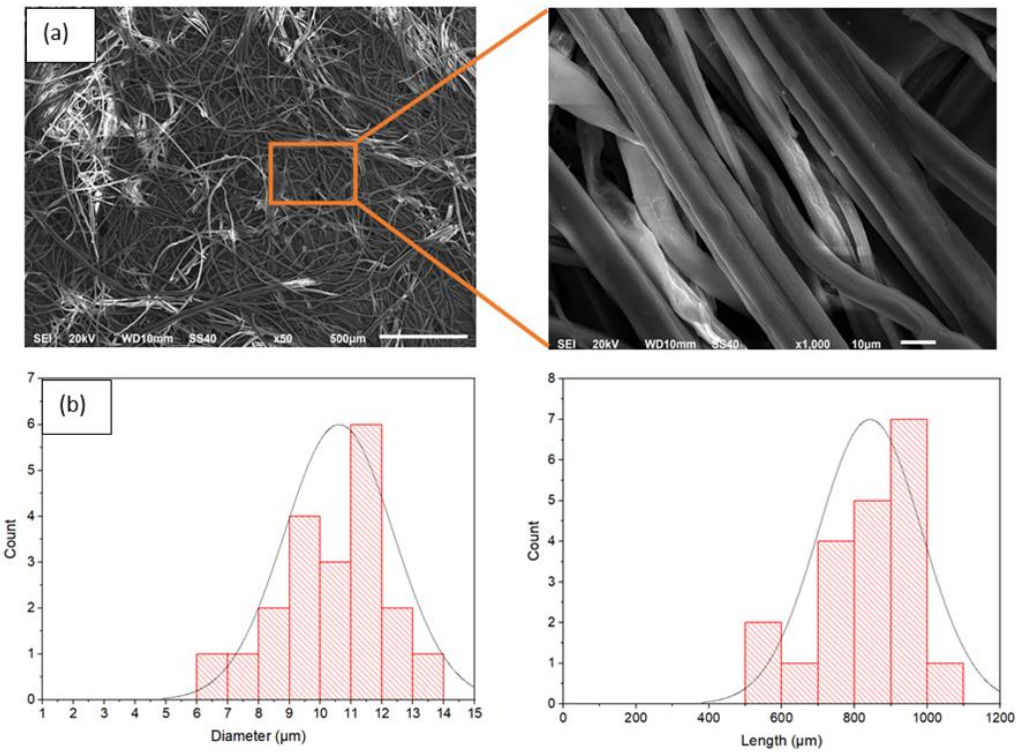

Figure 2 Image of micro-cellulose of Sisal (a) surfaces roughness, and (b) distribution of diameter and length of MCC

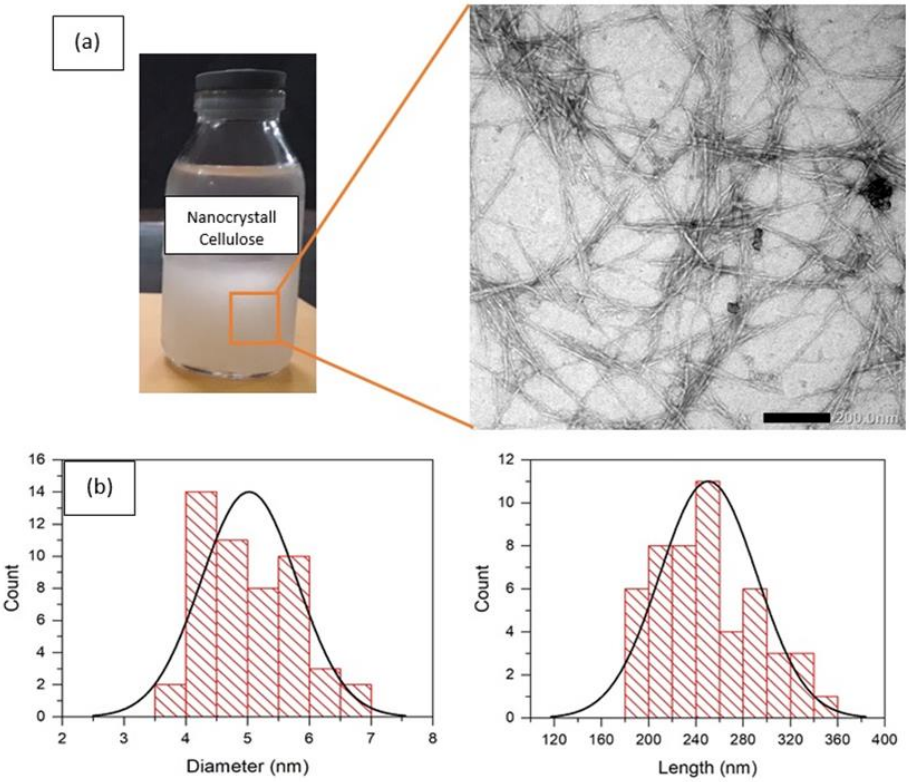

Figure 3 Image of NCC Sisal (a) web-like network structured, and (b) distribution of diameter and length of NCC

Sosiati et al. [19] investigated NCC, which can be observed using a conventional SEM tool (JSM-6510LA). The extracted NCC was obtained from Sisal fiber using alkali treatment in the 6 wt.% NaOH solution at temperature 100°C, for 3 hours. The alkalization process is then followed by bleaching 3 wt.% H_2O_2 (hydrogen peroxide) at temperature 70°C, for 2 hours. The fiber fibrillization process was carried out by 20 wt.% acid hydrolysis (H_2SO_4) solution at a temperature of 60°C for 5 hours using a magnetic stirrer, the hydrolysis process was stopped by ice cooling. The morphology is like a nano-whisker shape with a diameter (D) of 50 nm, a length (L) of 500 nm, and a high crystallinity index of 90.7%.

3.2. XRD Result

The data obtained from the XRD analysis in a graph in Figure 4 shows the relationship between the X-ray diffraction angle in the sample and the light diffracted by the sample where the x-axis is 2θ, and the y-axis is intensity. Each compound has a different angle, 2θ as its identity. The diffraction pattern of the XRD test results for each raw, purified, and NCC sample from Sisal fiber are different intensity peaks. Based on the XRD results, the presence of cellulose was known to have the highest peak for raw fiber and purified fiber samples at an angle of 2θ = 21.8, and for example, the highest NCC peak is at the angle 2θ = 22.5, which is the crystalline phase. Cellulose is a crystalline structure, while hemicellulose and lignin are amorphous structures. The content of amorphous substances in fiber can affect its crystallinity index (CI) value [20, 21, 22, 23]. The right concentration and time parameters can increase the value of the CI. The uncontrol concentration, time, and temperature are causing the CI of natural fiber to decrease [24]. The crystallinity index values of this work can be seen in Figure 4 and Table 2.

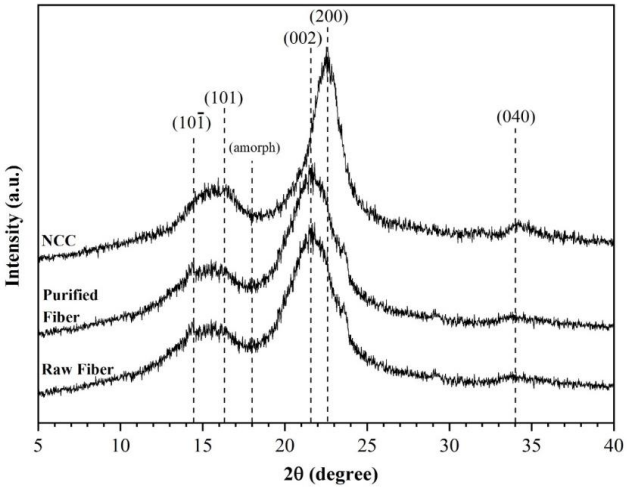

Figure 4 XRD spectra of raw, purified fiber and nanocrystalline cellulose

Table 2 Intensity value of crystalline and amorphous value of Sisal fiber

Sample	I_{002}	I_{am}	Crystallinity index (%)
Raw Fiber	224	764	70.68
Purified Fiber	233	900	74.11
NCC	201	949	78.82

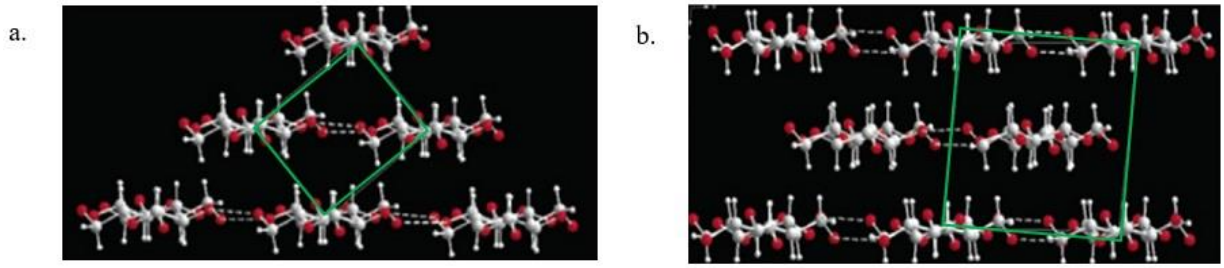

Figure 5 Schematic representation of the unit cells for cellulose structures; (a) Iα monoclinic structure, I_β triclinic structure

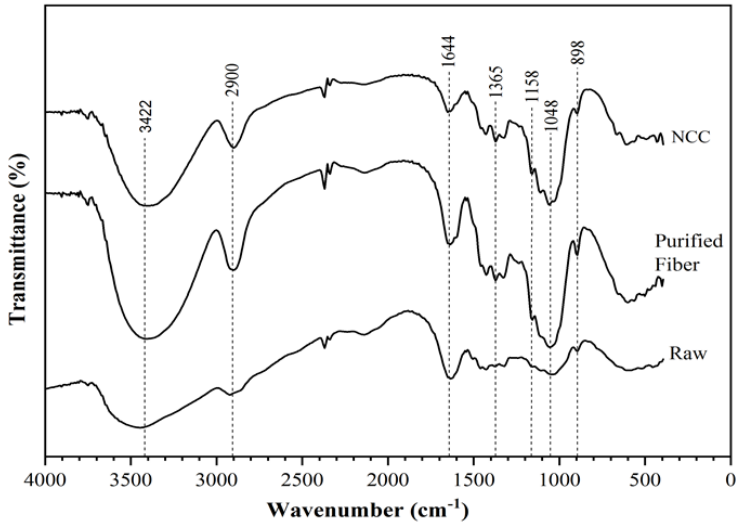

Figure 6 FTIR spectrum on the raw, purified, and NCC Sisal

The efficiency of ultrasonication in cellulose dissolution can be determined by the ultrasonic energy provided during the process, typically ranging from approximately 10 to 100 kJ/mol, indicative of the hydrogen bond energy scale. Consequently, ultrasonication can gradually disintegrate micro-crystalline cellulose (MCC) into nano-crystalline cellulose (NCC) rapidly. Through the stages of pre-treatment, acid hydrolysis, and ultrasonication isolation, NCC with a high crystallinity index of 78.82% can be produced.

Cellulose has two polymorphs: Iα (Alpha-cellulose) and I_β (Beta-cellulose). Figure 5a shows that Iα is a monoclinic structure containing one cellulose chain, and Figure 5b shows that I_β is a triclinic structure with two cellulose chains. The main difference between Iα and I_β is the relative displacement of cellulose along the (101) lattice plane in the triclinic structure and the (200) monoclinic structure, called the hydrogen bond [25, 40].

Insulation using the same fiber, namely sisal from Brazil by Mooran et al. [26], using pre-treatment de-waxing followed by soaking in NaOH and bleaching using H_2O_2 (hydrogen peroxide). The fibrillation process used the 60 wt.% sulfuric acid hydrolysis method, temperature 45°C for 30 minutes. This process produces NCC with a diameter of 30.9 ± 12 nm.

3.3. FTIR Result

Figure 6 shows a graph of the chemical structure of raw sisal fiber, purified fiber, and NCC. Absorption around 3422 cm^{-1}, 1400 cm^{-1}, 1365 cm^{-1}, and 890 cm^{-1} is characteristic of cellulose [27]. The broad absorption at 3400–3600 cm^{-1}, is associated with the stretching vibration of the OH group, and the absorption at 2920 cm^{-1} is associated with the stretching vibration of C-H [28]. The peak at 1644 cm^{-1} is related to the bending mode of the water molecule resulting from the strong interaction between water and cellulose. The other adsorption peaks can mainly be determined for the intermolecular hydrogen attraction at the C-O group at 1425 cm^{-1}, the stretching vibration of the C-O-C glycosidic band at 1158 cm^{-1}, and the rock vibration C-H at 898 cm^{-1}.

3.4. Thermal Properties Result

The TGA (thermalgravimetric Analysis) and DTG (derivative thermal gravimetry) tests aim to determine the heat resistance of the organic material of agave sisalana fiber. The TGA/DTG curve shows the degradation of the material due to the gradual increase in heat temperature, which causes the fiber material to lose weight (weight loss). Figure 7 shows the three central regions in the TGA/DTG test results named Region 1 and Region 2. Weight loss in the fiber of 10 wt.% begins at temperatures of 30-100°C. This is due to the evaporation of the natural fiber content, which contains water vapor and impurities.

Other materials in the fiber that have not been degraded at 100°C are lignin and hemicellulose. This compound is a thermoplastic material, so it can only be degraded at 260-360°C in Region 1. Hemicellulose, lignin, and cellulose experience degradation up to 60 wt.%. The last degradation occurred in Region 2, where the remaining fiber material lost another 26-29 wt.% active coil, so only 1-4 wt.% is residue.

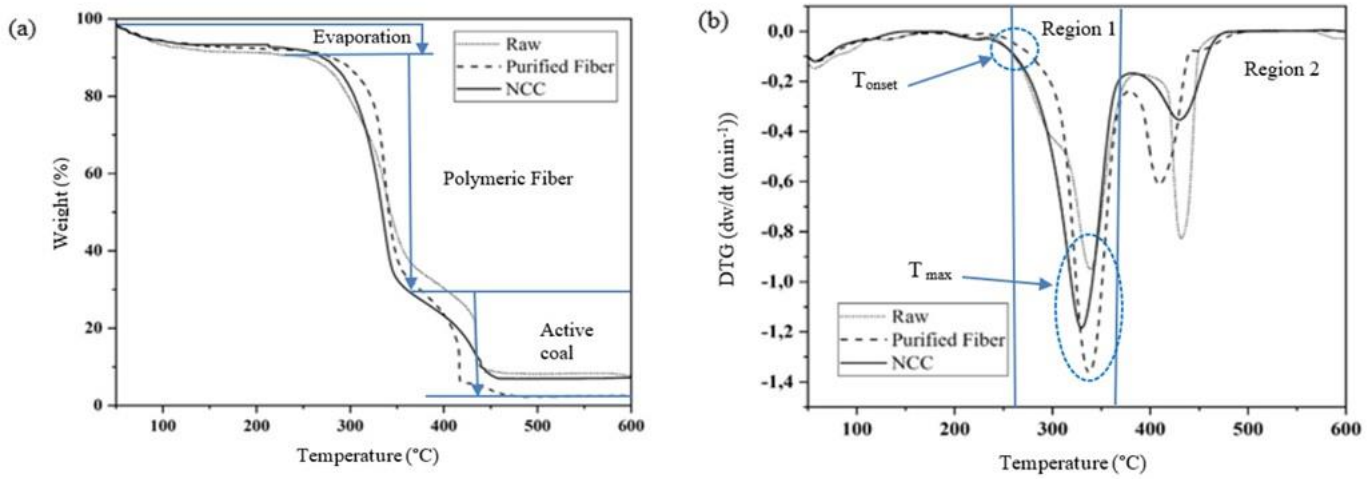

Figure 7 Thermal stability of raw, purified fiber, NCC (a) TGA curve, (b) DTG curve

The initial temperature degradation (T_{onset}) and the maximum temperature degradation (T_{max}), occur at a sharp peak on the DTG curve. Active coal formed results from the remaining combustion of natural fibers in Region 2 [29, 30, 31, 32, 33]. Purified fiber and NCC have initial temperatures (T_{onset}) higher than raw fiber, there are 260°C and 265°C. However, it differs from the maximum temperature (T_{max}), slightly lower than raw fiber (348°C), namely purified fiber at temp. 343°C, and NCC at temp. 338°C. It causes the ion-sulfate group to adhere and attach hydroxyl group in the NCC. Table 3 shows the comparative thermal stability of the other raw sources of natural fiber.

Table 3 Thermal analysis by initial (T_{onset}) and maximum (T_{max}) temperature of Sisal fiber, compare the other natural sources

Fiber Type	T_{onset}	T_{max}	reference
Agave *Sisalana**	246°C	348°C	This work
Agave *Americana*	203°C	332°C	[8]
Agave *Angustifolia*	210°C	312°C	[18]
Curaua	317°C	344°C	[34]
Bamboo	330°C	364°C	[35]
Ramie	258°C	346°C	[36]
Coir Fiber	271°C	331°C	[37]
Kenaf	230°C	330°C	[38]
Rice husk	198°C	353°C	[39]

4. CONCLUSION

NCC has successfully been produced from Sisal fibers by acid hydrolysis combined with ultrasonic treatment and it has produced a web-like network structured form with a diameter of 5±1 nm length of 260±20, and a high aspect ratio of 52. FTIR and XRD curve analysis showed that lignin and hemicellulose functional groups are eliminated after pre-chemical treatment, acid hydrolysis, and ultrasonication. It impacted the crystallinity index by 78.82%. The thermogravimetric analysis by TGA/DTG shows that NCC has good thermal stability (246-348°C), so NCC is suitable for filler polymers like bioplastic, thin film, filter membrane, wound healing membrane, and bridging interlayer of laminate composite.

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REFERENCES

[1] R.F. Listyanda, K. Kusmono, M.W. Wildan, and M.N. Ilman, "Extraction and Characterization of Nanocrystalline Cellulose (NCC) from Ramie Fiber by Sulphuric Acid Hydrolysis," *AIP Conference Proceedings*, vol. 2217, no. 1, April 2020. DOI: 10.1063/5.0000579

[2] F. Yudhanto, "Effect of Addition Microcrystalline Cellulose on Mechanical Properties of Jute/Glass Fibers Hybrid Laminated Composite," *Int. J. Automot. Eng.*, vol. 12, no. 1, pp. 1–8, 2021.

[3] K.V. Sabarish, K. Dhanasekar, R. Manikandan, R. Ancil, R. Venkat Raman, and P. Selva Surender, "Strength and Durability Evaluation of Sisal Fiber Reinforced Concrete," *Int. J. Civil Eng. Technol.*, vol. 8, no. 9, pp. 741–748, 2017.

[4] N. Pandi, S.H. Sonawane, and K.A. Kishore, "Synthesis of Cellulose Nanocrystals (CNCs) from Cotton Using Ultrasound-Assisted Acid Hydrolysis," *Ultrasonics Sonochemistry*, vol. 70, p. 105353, 2021.

[5] Kusmono and M.N. Affan, "Isolation and Characterization of Nanocrystalline Cellulose from Ramie Fibers via Phosphoric Acid Hydrolysis," *J. Nat. Fibers*, vol. 19, no. 7, pp. 2744–2755, 2022.

[6] M.S. Rana, M.A. Rahim, M.P. Mosharraf, M.F.K. Tipu, J.A. Chowdhury, M.R. Haque, and A.A. Chowdhury, "Morphological, Spectroscopic and Thermal Analysis of Cellulose Nanocrystals Extracted from Waste Jute Fiber by Acid Hydrolysis," *Polymers*, vol. 15, no. 6, p. 1530, 2023.

[7] R. Ratna, N. Arahman, A.A. Munawar, and S. Aprilia, "Extraction, Isolation, and Characterization of Nanocrystalline Cellulose from Barangan Banana (*Musa acuminata* L.) Peduncles Waste," *Indonesian J. Chem.*.

[8] P. Krishnadev, K.S. Subramanian, G.J. Janavi, S. Ganapathy, and A. Lakshmanan, "Synthesis and Characterization of Nano-Fibrillated Cellulose Derived from Green *Agave americana* L. Fiber," *BioResources*, vol. 15, no. 2, pp. 2442–2458, 2020.

[9] F. Yudhanto, J. Jamasri, H.S.B. Rochardjo, and A. Kusumaatmaja, "Experimental Study of Polyvinyl Alcohol Nanocomposite Film Reinforced by Cellulose Nanofibers from *Agave cantala*," *Int. J. Eng.*, vol. 34, no. 4, pp. 987–998, 2021.

[10] M.A. Gallardo-Sánchez, T. Diaz-Vidal, A.B. Navarro-Hermosillo, E.B. Figueroa-Ochoa, R. Ramirez Casillas, J. Anzaldo Hernández, and E.R. Macías-Balleza, "Optimization of the Obtaining of Cellulose Nanocrystals from Agave Tequilana Weber var. Azul Bagasse by Acid Hydrolysis," *Nanomaterials*, vol. 11, no. 2, p. 520, 2021.

[11] S.K. Singh, S. Khan, R.K. Mishra, and J. Karloopia, "Fabrication and Evaluation of Mechanical Properties of Polymer Matrix Composite Using Nano Fibers as a Reinforcement," *Materials Today: Proceedings*, vol. 46, pp. 1376–1383, 2021.

[12] A. Tozluoglu, S. Ates, E. Durmaz, S. Sertkaya, R. Arslan, O. Ozcelik, and Z. Candan, "Nanocellulose in Paper and Board Coating," in *Emerging Nanomaterials: Opportunities and Challenges in Forestry Sectors*, Springer, 2022. DOI: 10.1007/978-3-031-17378-3

[13] P.F. Muñoz-Gimena, V. Oliver-Cuenca, L. Peponi, and D. López, "A Review on Reinforcements and Additives in Starch-Based Composites for Food Packaging," *Polymers*, vol. 15, no. 13, p. 2972, 2023.

[14] A.O.C. Iroegbu and S.S. Ray, "Recent Developments and Future Perspectives of Biorenewable Nanocomposites for Advanced Applications," *Nanotechnology Reviews*, vol. 11, no. 1, pp. 1696–1721, 2022.

[15] F. Yudhanto, "Komposit Polimer-Nanoselulosa dari Serat Agave Cantala dengan Ekstraksi Kimia dan Proses Mekanis," Doctoral dissertation, Universitas Gadjah Mada, 2021.

[16] F. Yudhanto, J. Jamasri, H.S.B. Rochardjo, and A. Kusumaatmaja, "Experimental Study of Polyvinyl Alcohol Nanocomposite Film Reinforced by Cellulose Nanofibers from Agave Cantala," *Int. J. Eng.*, vol. 34, no. 4, pp. 987–998, 2021.

[17] N.A. Rosli, I. Ahmad, and I. Abdullah, "Isolation and Characterization of Cellulose," *2013 Tech Science Press*, 363.

[18] H. Sosiati, M. Muhaimin, Wijayanti, D.A., and K. Triyana, "Microscopic Characterization of Cellulose Nanocrystals Isolated from Sisal Fibers," in *Materials Science Forum*, vol. 827.

[19] R.A. Ilyas, S.M. Sapuan, M.S.N. Atikah, M.R.M. Asyraf, S.A. Rafiqah, H.A. Aisyah, and M.N.F. Norrrahim, "Effect of Hydrolysis Time on the Morphological, Physical, Chemical, and Thermal Behavior of Sugar Palm Nanocrystalline Cellulose (*Arenga pinnata* (Wurmb.) Merr)," *Textile Res. J.*, vol. 91, no. 1–2, pp. 152–167, 2021.

[20] N. Halib, F. Perrone, M. Cemazar, B. Dapas, R. Farra, M. Abrami, G. Chiarappa, G. Forte, F. Zanconati, G. Pozzato, and L. Murena, "Potential Applications of Nanocellulose-Containing Materials in the Biomedical Field," *Materials*, vol. 10, no. 8, p. 977, 2017.

[21] A. Oushabi, S. Sair, F.O. Hassani, Y. Abboud, O. Tanane, and A. El Bouari, "The Effect of Alkali Treatment on Mechanical, Morphological and Thermal Properties of Date Palm Fibers (DPFs): Study of the Interface of DPF–Polyurethane Composite," *South African J. Chem. Eng.*, vol. 23, pp. 116–123, 2017.

[22] M. Jonoobi, R. Oladi, Y. Davoudpour, K. Oksman, A. Dufresne, Y. Hamzeh, and R. Davoodi, "Different Preparation Methods and Properties of Nanostructured Cellulose from Various Natural Resources and Residues: A Review," *Cellulose*, vol. 22, no. 2, pp. 935–969, 2015.

[23] S.M. Shahril, M.J.M. Ridzuan, M.A. Majid, A.M.N. Bariah, M.T.A. Rahman, and P. Narayanasamy, "Alkali Treatment Influence on Cellulosic Fiber from *Furcraea foetida* Leaves as Potential Reinforcement of Polymeric Composites," *J. Mater. Res. Technol.*, vol. 19, pp. 2567–2583, 2022.

[24] M. Poletto, V. Pistor, and A.J. Zattera, "Structural Characteristics and Thermal Properties of Native Cellulose," in *Cellulose-Fundamental Aspects*, vol. 2, pp. 45–68, 2013.

[25] J.I. Morán, V.A. Alvarez, V.P. Cyras, and A. Vázquez, "Extraction of Cellulose and Preparation of Nanocellulose from Sisal Fibers."

[26] Z. Liu, X. Li, W. Xie, and H. Deng, "Extraction, Isolation and Characterization of Nanocrystalline Cellulose from Industrial Kelp (*Laminaria japonica*) Waste," *Carbohydrate Polymers*, vol. 173, pp. 353–359, 2017.

[27] W. Xie, Z. Song, Z. Liu, and X. Qian, "Surface Modification of PCC with Guar Gum Using Organic Titanium Ionic Crosslinking Agent and Its Application as Papermaking Filler," *Carbohydrate Polymers*, vol. 150, pp. 114–120, 2016.

[28] Y. Guo, S. Zhu, Y. Chen, and D. Li, "Thermal Properties of Wood-Plastic Composites with Different Compositions," *Materials*, vol. 12, no. 6, p. 881, 2019.

[29] M. Jawaid, H.A. Khalil, A.A. Bakar, A. Hassan, and R. Dungani, "Effect of Jute Fibre Loading on the Mechanical and Thermal Properties of Oil Palm–Epoxy Composites," *J. Composite Mater.*, vol. 47, no. 13, pp. 1633–1641, 2013.

[30] V. Dalla Libera Junior, R.M. Leão, V. Franco Steier, and S.M. da Luz, "Influence of Cure Agent, Treatment and Fibre Content on the Thermal Behaviour of a Curaua/Epoxy Prepreg," *Plastics, Rubber and Composites*, vol. 49, no. 5, pp. 214–221, 2020.

[31] C. Li, X. Yin, Y. Wang, L. Zhang, Z. Zhang, Y. Liu, and G. Xian, "Mechanical Property Evolution and Service Life Prediction of Pultruded Carbon/Glass Hybrid Rod Exposed in Harsh Oil-Well

Condition," *Composite Structures*, vol. 246, p. 112418, 2020.

[32] M. El Boustani, G. Lebrun, F. Brouillette, and A. Belfkira, "Effect of a Solvent-Free Acetylation Treatment on Reinforcements Permeability and Tensile Behaviour of Flax/Epoxy and Flax/Wood Fibre/Epoxy Composites," *Can. J. Chem. Eng.*, vol. 95, no. 6, pp. 1082–1092, 2017.

[33] L.A. Teixeira, L. Vilson Dalla Junior, and S.M. Luz, "Chemical Treatment of Curaua Fibres and Its Effect on the Mechanical Performance of Fibre/Polyester Composites," *Plastics, Rubber and Composites*, vol. 50, no. 4, pp. 189–199, 2021.

[34] S.C. Chin, K.F. Tee, F.S. Tong, H.R. Ong, and J. Gimbun, "Thermal and Mechanical Properties of Bamboo Fiber Reinforced Composites," *Materials Today Communications*, vol. 23, p. 100876, 2020.

[35] K. Kusmono and D.A. Akbar, "Influence of Hydrolysis Conditions on Characteristics of Nanocrystalline Cellulose Extracted from Ramie Fibers by Hydrochloric Acid Hydrolysis," 2020.

[36] Y. Dong, A. Ghataura, H. Takagi, H.J. Haroosh, A.N. Nakagaito, and K.T. Lau, "Polylactic Acid (PLA) Biocomposites Reinforced with Coir Fibres: Evaluation of Mechanical Performance and Multifunctional Properties," *Composites Part A: Appl. Sci. Manuf.*, vol. 63, pp. 76–84, 2014.

[37] A.A. Yussuf, I. Massoumi, and A. Hassan, "Comparison of Polylactic Acid/Kenaf and Polylactic Acid/Rice Husk Composites: The Influence of the Natural Fibers on the Mechanical, Thermal and Biodegradability Properties," *J. Polym. Environ.*, vol. 18, pp. 422–429, 2010.

[38] B. Rashid, Z. Leman, M. Jawaid, M.J. Ghazali, and M.R. Ishak, "Influence of Treatments on the Mechanical and Thermal Properties of Sugar Palm Fibre Reinforced Phenolic Composites," *BioResources*, vol. 12, no. 1, pp. 1447–1462, 2017.

[39] F. Fatkhurrohman, H.S.B. Rochardjo, A. Kusumaatmaja, and F. Yudhanto, "Extraction and Effect of Vibration Duration in Ultrasonic Process of Cellulose Nanocrystal (CNC) from Ramie Fiber," in *AIP Conference Proceedings*, vol. 2262, no. 1, 2020.