Preparation and Characterization of Nanocelluloses from Oil Palm Empty Fruit Bunch Cellulose

Dwi Yuni HASTATI※1†, Erliza HAMBALI※2※3, Khaswar SYAMSU※2, and Endang WARSIKI※2※3

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This study aims to prepare nanocelluloses from chemically purified celluloses of oil palm empty fruit bunch (CPC-OPEFB) by using acid hydrolysis. The nanocelluloses from CPC-OPEFB was prepared with sulfuric acid treatment at the concentration of 67 wt% and temperature of 40 ± 1 °C, for 10, 20, 30, and 40 min. As a comparison, nanocelluloses from commercial microcrystalline cellulose (C-MCC) were also prepared with the same condition. The effects of hydrolysis time on the morphology and physical properties of the obtained nanocelluloses were investigated. Observation with TEM showed that nanocelluloses from CPC-OPEFB were long and fibril, whereas the ones from C-MCC showed a rod-like structure and crystalline. This observation was in agreement with the DSC analysis, i.e. the endothermic peaks were not present on the DSC curve of nanocelluloses from CPC-OPEFB, whereas it was clearly observed on the DSC curve of nanocelluloses from C-MCC, indicating that the nanocelluloses from CPC-OPEFB were amorph, and the ones from C-MCC were crystalline. The particle size analysis revealed that the diameter of the obtained nanocelluloses was affected by hydrolysis time. The best hydrolysis time to obtain the smallest diameter of NFCs from CPC-OPEFB and NCCs from C-MCC was 30 and 40 min, respectively.

Key Words
Nanocellulose, Chemically purified celluloses, Nanofibrillated celluloses, Oil palm empty fruit bunch, Acid hydrolysis

1. Introduction
The utilization of biodegradable materials from biomass has been increased because of its widespread applicability, sustainability, and minimization of dependence on fossil fuel. The production of oil palm biomass in Indonesia is approximately 40–60% from total production, including oil palm kernel shell (OPKS), oil palm mesocarp (OPM), and oil palm empty fruit bunch (OPEFB) fibers. OPEFB fiber consists of 44.4 % of cellulose, 30.9% of hemicellulose, and 14.2 % of lignin. Without using chemical pretreatment on the cellulose extraction from OPEFB fiber, hemicellulose, lignin, and other extractives materials are still remained.

Biomass from OPEFB with high cellulose content is one source of nanocellulose as the filler in nanocomposite and beneficial for nanocomposite reinforcing agents. The utilization of this cellulose is a clear advantage in terms of environmental impact. Preparations of nanocelluloses from cellulose fiber have been studied by some researches including chemical treatment, mechanical treatment, and chemo-mechanical treatment processes. The type of nanocelluloses can be classified in two main categories, i.e., nanofibrillated celluloses (NFCs) and nanocrystalline celluloses (NCCs), based on their morphology and crystallinity. A typical shape of NFCs is long and flexible with the diameter is less than 100 nm and the length ranges from 100 nm to 2 µm. The crystallinity of NFCs is quite low, less than 50%. On the other hand, a typical shape of NCCs is a rod-like with less than 20 nm width and the length varies between 100 and 500 nm. The crystallinity of NCCs are generally high, more than 88%. The preparation of nanocelluloses with acid hydrolysis...
treatment has been extensively studied by using different cellulotic sources [10,11]. Some important parameters to the acid hydrolysis are acid to pulp ratio, reaction time, temperature, and acid type used [12]. Sulphuric acid (H2SO4), hydrochloric acid (HCl), and phosphoric acid (H3PO4) are recognized as the most effective to remove the amorphous regions in the isolation process of nanocelluloses [10,11]. The advantage of using sulphuric acid is that it can produce a negative surface charge on the particles leading to more stable suspensions [13]. This processing route of nanocelluloses influence the characteristics of nanocelluloses properties obtained.

Isolated nanocelluloses from OPEFB showed excellent properties such as low density, superior mechanical strength, renewability, biodegradability [14]. They have good potential as nano-filler that can improve the mechanical properties of polymer with an only small addition. Many potential applications of nanocelluloses have been reported for nano-composite based packaging [15]. The use of nanocellulose could improve the mechanical properties and oxygen barrier properties, while decreasing water vapor permeability [16,17].

The size and characteristics of nanocelluloses depend on the source of the original cellulose material and on the acid hydrolysis process such as the type of acid and its concentration, temperature and time of hydrolysis [18]. Some studies reported successful isolation of nanocelluloses from OPEFB fibers with varied of acid hydrolysis concentration [19,20], or with pre-treatment process [21], or even by using the stronger acid concentration [22]. Nevertheless, using chemically purified celluloses of OPEFB (CPC-OPEFB) as a source of nanocellulose was still limited. The objectives of this study were to prepare nanocelluloses from chemically purified celluloses of OPEFB (CPC-OPEFB) by using sulfuric acid hydrolysis and characterize the morphology and physical properties of the nanocelluloses obtained by varying the time of acid hydrolysis and the source of cellulose.

2. Experimental

2.1 Sample preparations

Chemically purified celluloses of OPEFB (CPC-OPEFB) were supplied by Balai Penelitian Kelapa Sawit Medan (the Research Center of Palm Oil, Medan) and were used as raw materials in this study. Commercial micro crystalline cellulose, Avicel® PH 101, Fluka, supplied by Sigma Aldrich, USA, with a size of ~ 51 µm, was used as a reference. The chemical used for acid hydrolysis was sulfuric acid (H2SO4) 95-97 %, purchased from Merck, Indonesia and all of materials were used as received.

Isolation of nanocelluloses was conducted by hydrolysis of sulfuric acid solution (67 wt %) on CPC-OPEFB under moderate cycle agitation (260 rpm) at 40 °C for 10, 20, 30, and 40 min. The cellulose acid ratio was 1:12 (dry base). As soon as the hydrolysis treatment was complete, the suspension was added by cold distillate water as much as 4x the initial volume of the sample to stop the reaction. Subsequently, the diluted suspension was centrifuged at 4600 rpm for 30 min to obtain a precipitate. The precipitate was re-suspended in water with moderate agitation, followed by centrifugation. This process was repeated many times until the pH of the suspension reached 5 – 6 and the supernatant became cloudy. After that, the suspensions were sonicated for 20 minutes to disperse the nanocelluloses in water using an ultrasonic homogenizer at 42 kHz and 70 W output power (Branson Ultrasonics Corporation, Danbury, USA) and stored in a refrigerator. During the sonicated treatment, the suspension was cooled by an ice water bath to avoid overheating. The same treatment was also conducted to commercial MCC (C-MCC).

A series of nanocellulose suspensions were coded as S-t and M-t, where S stands for CPC-OPEFB, M stands for C-MCC and t stands for the hydrolysis time. Thus S-10, for example, signifies that CPC-OPEFB was hydrolyzed with sulfuric acid for 10 min.

2.2 Characterization methods

2.2.1 Fourier transform infrared spectroscopy

Fourier transform infrared spectroscopy (FT-IR) was performed on a Perkin Elmer 1600 infrared spectrometer using the KBr method in the ratio of 1:100 and made to a pellet. FT-IR spectra of the coated pellet were recorded by using a Nicolet avatar 360 at 32 scans with a resolution of 4 cm⁻¹ and within the wave number ranges from 370 to 4000 cm⁻¹. The positions of significant transmittance peaks were determined by using the find peak tool provided by the Nicolet omnic 5.01 software.

2.2.2 Transmission electron microscopy (TEM)

The morphology of nanocelluloses was examined under a transmission electron microscope (JEC 560, JEOL Japan). A droplet of an aqueous suspension of nanocelluloses was deposited using a sputtering coater on the surface of a carbon-coated copper grid and allowed to settle for about 3 min to dry. The sample was then analyzed by using a TEM accompanied by selected area electron diffraction (SAED). The analysis was operated at 120 kV with magnifications of 50.0 nm and 200.0 nm.

2.2.3 Particle size distribution

The particle size distribution of nanocelluloses was investigated using a particle size analyzer (PSA)-MALVERN
Zetasizer. Before measurement, 1% (v/v) supernatant of nanocelluloses was diluted in 20 mL aqua distillate and put in a cuvette. The cuvette passed through the visible light that diffraction occurred. Measurement of particle size utilizes the principle of visible light scattering. The output produced was a graph of particle size distribution.

2.2.4 Differential scanning calorimetric (DSC)

Thermal properties analysis was conducted by using DSC (Perkin Elmer Thermal Analysis instrument). Samples (5-10 mg) were placed in DSC pan. The study is carried out by heating the sample at temperature 10.0 °C to 400.0 °C with heating speed 10.0 °C/min in a nitrogen atmosphere. Blank pan is used as a reference.

3. Results and Discussion

3.1 Visual Appearance of Nanocellulose

Fig. 1 shows photographs of the visual appearance of the aqueous suspension of nanocelluloses, isolated from CPC-OPEFB (1a) and isolated from C-MCC (1b). All nanocellulose samples were dispersed in water at the concentration (0.1%- 0.2% w/v). They formed turbid suspensions, like milk, and stable. The yield of aqueous suspension of nanocelluloses, isolated from CPC-OPEFB and C-MCC was approximately in the range 78.69 - 94.40%. For the obtained nanocelluloses from CPC-OPEFB, with longer hydrolysis time, the number of dispersed nanocelluloses was increased and reached 94.40% when acid hydrolysis was performed for 40 min. The previous study has reported the same phenomena.

The stability of nanocellulose suspensions was good and remained in a milky colloidal suspension. That can be visibly observed during the research (image not presented). It can be achieved because the hydrolysis treatment by sulfuric acid has introduced sulfate half-ester groups which yield a negative surface charge on nanocelluloses surfaces. These negative sulfate charges (SO₄²⁻) were able to create a strong electrostatic repulsion between the nanocellulose surfaces and limit their ability to flocculate.

3.2 Morphological analysis

The morphology of the obtained nanocelluloses was investigated using transmission electron microscope (TEM). The TEM image showed that the nanocelluloses were well-isolated. Fig. 2 shows the different size of nanocellulose from CPC-OPEFB and C-MCC. Crystallographic analysis of SAED pattern was generated by TEM. The isolated nanocelluloses from CPC-OPEFB exhibited long and flexible structure which is of a typical shape of nano fibrillated celluloses (NFCs). This individual of NFCs had a diameter of less than 50 nm and were several micrometers in lengths (Fig. 2 h). These images were in agreement with those that have been reported by previous researches. For comparison, the isolated nanocelluloses from C-MCC exhibited a rod-like structure (wider in the middle than at the ends), which is a common observation of well-dispersed nanocrystalline celluloses (NCCs) in water.

In the preparation of nanocelluloses from CPC-OPEFB, sulfuric acid hydrolysis cleaved the amorphous region of microfibrils cellulose transversely, and resulting reduction in fiber diameter from microns to nanometers. This is the reason why the acid hydrolysis process in CPC-OPEFB just yielded the NFCs, as can be seen at the TEM image (Fig. 2 b, e). Contrariwise, the isolated nanocelluloses from C-MCC showed the shape of NCCs at TEM image (Fig. 2 h). It could be explained that the process of getting C-MCC is by dissolving the amorphous region of cellulose. This is the first region that is attacked easily by the acid in the process of cellulose hydrolysis. Thus, the next sulfuric acid attack, just like in the isolation process to generate nanocellulose in this work, the sulfuric acid could penetrate easily and release the crystalline part of C-MCC and yield the NCCs.

It was found that some self-aggregations in irregular shapes still occurred on the obtained nanocelluloses in this work. They displayed with dark color and seen non-homogenous distribution in distilled water (Fig. 2 a, d, g at a TEM magnification of 200.0 nm and Fig. 2 b, e, h at a TEM magnification of 50.0 nm). These aggregations...
occurred due to the strong hydrogen bonding between the nanocelluloses and van der Waals forces. Besides that, another reason for re-aggregation of the particles forms high molecular dimensions due to the presence of reactive sites on the surface of supramolecular that associated relatively with small heterogeneous molecules. This association also supported by weak dispersive forces, e.g., low intensity of ultrasonication. Thus, it could be explained why the nanocelluloses obtained in this research still formed self-aggregation in distilled water even though they had sonicated. It is presumably due to the power level of sonication which was too low and was not enough to disperse the nanocelluloses in distilled water optimally.

The results on the crystallographic analysis of SAED pattern generated by TEM revealed a bright spot on the images (Fig. 2 c, f, i). SAED spectra indicated the disappearance of the crystallinity of NFCs and NCCs due to no periodicities and regularities of the crystal. It was signified as an amorphous region. This diffraction pattern of the SAED spectra was similar to that of previous amorphous studies that NFCs contain a higher amount of non-crystalline part. The existence of the amorphous part could be from amorphous cellulose region and lignin.

This observation was in agreement with the differential scanning calorimetry (DSC) analysis. In the DSC thermograms (Fig. 3), the endothermic peaks were not observed on the DSC curve of nanocelluloses from CPC-OPEFB, whereas it was clearly seen on the DSC curve of nanocelluloses from C-MCC. Thus, TEM and DSC analysis revealed that nanocelluloses made from CPC-OPEFB were amorph, and nanocelluloses from C-MCC were crystalline. The existence of the crystalline parts influenced the stability on the thermal effect of nanocelluloses. The introduction of active sulfate ester groups on the obtained nanocelluloses surfaces after sulfuric acid hydrolysis, however, will potentially contribute to a detrimental effect on thermal properties of the nanocelluloses.

### 3.3 Particle size distribution

The geometrical dimensions for nanocelluloses depend on the origin of cellulose fibers, the width is generally tenths of nanometers and the length is hundredths of nanometers. In this study, the various width of nanocelluloses from CPC-OPEFB (marked by S-10, S-20, S-30, S-40) and nanocelluloses from C-MCC (marked by M-10, M-20, M-30, M-40) under 100 nm were successfully isolated. Table 1 shows the particle size distribution of these nanocelluloses analyzed using cumulants method. Based on the result, generally, the increased sulfuric acid hydrolysis time resulted in a decreased of nanocellulose...
size, including z average (particle size distribution), D mean intensity, volume, and number. However, it was observed that some self-aggregations still occurred in the suspension. The aggregates were approximately in the range 159.90 to 308.20 nm for nanocelluloses from CPC-OPEFB and from 127.50 to 250.80 nm for nanocelluloses from C-MCC based on PSA results. This might be due to the strong hydrogen bonding and van der Waals forces in nanocelluloses.

It was found that with increasing reaction time, the length of nanocelluloses from CPC-OPEFB decreased quite significantly, whereas the diameter remained almost unchanged. This condition was the same as the previous report that the size of nanocelluloses from OPEFB, as to signify as NFCs, decreased as the hydrolysis time was increased. It was observed that the diameter of the obtained nanocelluloses from CPC-OPEFB, signify with NFCs, ranges from 26.51 to 40.29 nm; from 17.92 to 45.74 nm; from 13.34 to 35.28 nm; and from 13.59 to 111.77 nm for 10, 20, 30 and 40 minutes hydrolysis time, respectively. On the other hand, the diameter of the obtained nanocelluloses from C-MCC, signify as NCCs, ranges from 22.62 to 61.90 nm; from 18.74 to 46.64 nm; from 27.08 to 48.78 nm; and from 12.81 to 35.51 nm with acid hydrolysis for 10, 20, 30 and 40 minutes, respectively (Fig. 4). These results showed that acid hydrolysis for 30 min was the best treatment to prepare NFCs from CPC-OPEFB, while 40 min treatment was the best condition for preparing NCCs from C-MCC. The previous research reported that the longer reaction time of acid hydrolysis (64 wt% and 90 min) has succeeded to isolate NFCs from OPEFB cellulose with the diameter in the range 1 - 3.5 nm and estimated length between 100 nm and 2 µm.

### 3.4 FT-IR analysis

Evaluation of chemical composition due to acid hydrolysis treatment on nanocelluloses obtained was carried out through FT-IR spectroscopy (Fig. 5). The wavenumber of the transmittance band observed was in the range 400 to 4000 cm$^{-1}$. The FTIR spectra displayed all samples which consisted of CPC-OPEFB (S-0), C-MCC (M-0), and the obtained nanocelluloses (isolated from CPC-OPEFB: marked by S-10, S-20, S-30, and S-40; and isolated from C-MCC: marked by M-10, M-20, M-30, and M-40). This spectra showed two main absorbance regions at high wavenumbers (3000-3500 cm$^{-1}$) and low wavenumbers (1500-1700 cm$^{-1}$) respectively, consistent with the previous studies.

A dominant and broad absorption band

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**Table 1** Particle size distribution of oil palm empty fruit bunch NFCs based on cumulants method

| Hydrolysis time (min) | Particle size by number (nm) | Z average (nm) | D mean intensity (nm) | D mean volume (nm) | D mean number (nm) | PDI |
|-----------------------|-------------------------------|----------------|-----------------------|-------------------|-------------------|-----|
| 10                    | 26.51-40.29                   | 308.20         | 77.33                 | 55.65             | 33.40             | 0.75|
| 20                    | 17.92-85.74                   | 178.50         | 295.20                | 62.80             | 51.83             | 0.44|
| 30                    | 13.34-35.28                   | 159.90         | 225.10                | 45.84             | 24.31             | 0.39|
| 40                    | 13.59-111.77                  | 220.00         | 275.60                | 50.92             | 62.68             | 0.29|

**Fig. 3** DSC image of: (a) isolated nanocellulose from CPC-OPEFB treated for 20 min and (b) isolated nanocelluloses from C-MCC treated for 20 min.
located at peak in the region 3300 cm\(^{-1}\) and peak at approximately 2900 cm\(^{-1}\) which were observed in the entire spectra are attributed to O–H groups stretching in cellulose and contributed to the aliphatic saturated C-H stretching vibration in cellulose, hemicellulose, and lignin\(^{28}\). A transmittance band at around 1600 for CPC-OPEFB, C-MCC, nanocelluloses from CPC-OPEFB, and nanocelluloses from C-MCC, was assigned to absorb water due to the strong interaction between cellulose and water\(^{25}\).

A unique peak at 1736 cm\(^{-1}\) could be observed in the spectra of CPC-OPEFB and its nanocelluloses obtained (meant by NFCs), but this peak was not seen in the other spectra (C-MCC and its nanocelluloses obtained (signified by NCCs). This peak indicated the presence of the acetyl
and uronic ester groups of the hemicelluloses or to the ester linkage of the carboxylic groups of the ferulic and p-coumaric acids of lignin and hemicellulose). It could be assumed that there were residual lignin and hemicellulose in CPC-OPEFB and nanocelluloses from CPC-OPEFB. Besides, the transmittance peaks at around 1400 cm⁻¹ and about 1100 cm⁻¹ were the other signals for tracking the existence of residual lignin component. In this work, they could be seen slightly on the transmittance of CPC-OPEFB and their nanocelluloses. However, these characteristic peaks disappeared on the transmittance of nanocelluloses from C-MCC. The disappearance of this peak indicated that most of the hemicelluloses and lignin were removed in C-MCC due to the chemical treatments during the synthesis of C-MCC, such as an alkali and bleaching treatments. In addition, using sulfuric acid on this work has made deformation effectively on the structure of lignin and hemicellulose on nanocelluloses from C-MCC.

The transmittance peaks observed at around 1640 cm⁻¹ in the spectra of all samples were recognized by the O-H bending of the adsorbed water. This moisture content may be related to the absorption of moisture in the spaces left vacant from the removal of hemicellulose and lignin. The open surfaces resulted from the alkali treatment of cellulose helps absorb moisture. Besides that, sulfuric acid hydrolysis generates some hydroxyl groups on the nanocellulose surfaces.

The vibration peak detected at 1365 cm⁻¹ in nanocelluloses from CPC-OPEFB and nanocelluloses from C-MCC samples is related to the bending vibration of the C-H and C-O bonds in the polysaccharide aromatic rings. The presence of cellulose and C-MCC can be seen at around the peak 900 cm⁻¹ which is known as β-glycosidic linkages between the sugar units.

The spectra of hydrolysis samples (the obtained nanocelluloses) at peaks around 3300–3500 cm⁻¹ became shallower than the unhydrolysis samples (S-0 and M-0) as hydrolysis time was increased. It indicates that hydrogen bonds were broken due to sulfuric acid hydrolysis. This pretreatment attached OH with the sulfate group, resulting in a reduction in the amount of OH, evident from the decreased intensity of the peaks between 3300–3500 cm⁻¹ band in all hydrolysis samples obtained. The longer hydrolysis reaction time reduced the intensity of these peaks.

In general, it can be seen that the peaks of nanocelluloses from C-MCC were shallower than those of nanocelluloses from CPC-OPEFB. This is caused by the fact that a decrease in hydroxyl groups was not only induced by solvent hydrolysis but also by different chemo-mechanical treatments including delignification, bleaching, and ultrasonication. Presumably, nanocelluloses from CPC-OPEFB passed fewer chemo-mechanical processing routes than nanocelluloses from C-MCC. And the CPC-OPEFB was categorized as cellulose I with the presence of signals at around 3300 cm⁻¹, 1100 cm⁻¹, and 800 cm⁻¹.

4. Conclusion

Nanocelluloses from CPC-OPEFB were successfully obtained by using acid hydrolysis. The best condition for preparing nanocelluloses from CPC-OPEFB was with a sulfuric acid concentration of 67 wt%, under moderate cycle agitation (260 rpm) at 40 ± 1 °C, for 30 min of the acid hydrolysis process. Using this method, the obtained nanocelluloses from CPC-OPEFB had the diameter in the range 13.34 to 35.28 nm. TEM and DSC analysis revealed that the nanocelluloses made from CPC-OPEFB were fibrillated and amorph, so it is classified as NFCs. This study found that to obtain nanocrystalline cellulose, it is better to use cellulose crystalline as starting material.

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