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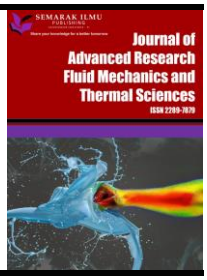

Morphology and Dispersion Stability of Nanocellulose Extracted from Oil Palm Empty Fruit Bunch Fibre by High-Pressure Homogenization

Fatiha Ismail[1,*], Nur Eliyanti Ali Othman[1], Noorshamsiana Abdul Wahab[1]

[1] Biomass Technology Unit, Engineering & Processing Research Division, Malaysian Palm Oil Board, 6, Persiaran Institusi, Bandar Baru Bangi, 43000 Kajang, Selangor, Malaysia

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ABSTRACT

This paper aims to study the morphology and dispersion stability of nanocellulose extracted from oil palm empty fruit bunch (EFB) fibre by high-pressure homogenization method. Microcrystalline cellulose (MCC) from EFB fibre was hydrolysed using sulphuric acid, followed by high-pressure homogenization, to produce nanocellulose. The isolated MCC and nanocellulose was characterized for their microstructure and stability of the particles. Observation with SEM showed that diameter size of MCC from OPEFB were decreased along with the length of the fibres as the concentration of sulphuric acid increased. While the effects of homogenization cycles on the morphology of the obtained nanocelluloses were observed using FESEM. Overall results revealed that an addition of cycle numbers has shown a reduction in diameter size and substantial breakdown of the cell walls of the nanocellulose. In contrast, for the stability test, the MCCs and NCs particles' stability and dispersibility were dependent on the electrostatic interactions, which helped to maintain a more stable suspension. Therefore, combining the chemical and mechanical processes can successfully pave the way for the production of high added-value nanocellulose from a variety of cellulosic sources and has a good potential for nanocellulose production in Malaysia.

1. Introduction

Agricultural waste produced by the oil palm industry during replanting, pruning, and milling operations is referred to as oil palm biomass and is often left to degrade in the fields [1]. Palm oil only makes up 10% of the oil palm tree, despite the fact that the majority of the tree (around 90%) is mostly thought of as biomass [2]. Oil palm trunks and fronds make up the majority of the biomass produced at a plantation; the biomass produced in oil palm processing mills comes from empty fruit bunches, palm kernel shells, mesocarp fibres, and palm oil mill effluent (POME) [3]. Out of 101.02 metric tonnes (MT) of processed oil palm empty fruit bunch fibre (OPEFB), the total oil palm biomass residues (dry weight basis) from replanting, pruning, and milling activities in Malaysia were projected

* *Corresponding author.*
E-mail address: fatihaismail@gmail.com

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to be 51.19 MT in 2017 [4]. In addition to extractives in the cell wall, OPEFB is a lignocellulosic residue that contains 50% cellulose, 25% hemicellulose, and 20% lignin [5].

As one of the most abundant materials on the planet, cellulose is an important component and an appealing renewable feedstock for humanity's energy demands [6]. Cellulosic fibres, on the other hand, contain a complex crystalline structure that is bound together by a network of hydrogen bonds and hydrophobic interactions, making crystal deconstruction difficult. As a result of these factors, the creation of biodegradable materials, especially cellulose-based products, is a major problem [7]. We can increase the applicability of cellulose by shifting the focus from macroscopic qualities to the nanoscale and manipulating or self-assembling molecule by molecule using creative nanotechnology [8].

There are now several feasible techniques for producing nanocellulose, including chemical methods [4,5], biological methods [9], and synthetic methods [10]. However, there are numerous flaws in these methods that limit their applicability. Chemical methods, for example, necessitate a very acidic or alkaline atmosphere, which would contaminate the ecosystem. While, biological treatment takes time, money, and effort. Special equipment and excessive energy consumption [11] are frequently required in the physical processing of nanocellulose. Eco-friendly and energy-saving technology is a highly desirable aspect of a method for producing nanocellulose.

Due to its high efficiency, reproducibility, and ease of scaling up in industry [12], high pressure homogenization is a common technique in many technical applications. Examples include the creation of emulsions to extend shelf life [13], a cutting-edge method for creating co-crystals [14], and others. In this procedure, samples are moved at a fast rate of speed and high pressure via a small homogenization gap [15]. By using shearing forces during high-pressure homogenization, which results in the breakup of big particles into thinner ones, molecular refinement was accomplished [16]. It can get through the strong barrier to modify macromolecules' molar mass distribution and functioning [17]. This necessity, however, restricts the production's potential scaling-up and leads to low efficiency and limited capacity [18].

This study aims to provide a comparative analysis for more economical and environmentally sustainable methods of generating nanocellulose. In this work, combination of chemical and mechanical (acid hydrolysis and high-pressure homogenization) were used as the methodology. Investigating how the amount of high-pressure homogenization treatments and the acid-to-MCC ratio affected the final nanocellulose was one of the objectives in particular. The structural and physicochemical features of the nanocellulose was examined using field emission scanning electron microscopy (FESEM). We assessed the physicochemical characteristics and dispersion stability of cellulose, MCCs and NCs, respectively. This article refers to the groundbreaking research on separating MCC and NCC from OPEFB so they can be used as bio-based fillers for composites with sustainable manufacturing processes.

2. Methodology

2.1 Materials

Oil palm empty fruit bunch (OPEFB) fibre samples were obtained from the palm oil mill located in Perak. The OPEFB The OPEFB shredded fibres were then transported to MPOB head office, Selangor, Malaysia for further processing. Other reagents that were used in the experiment are analytical grade sodium hydroxide, monochloroacetic acid, and glacial acetic acid (Merck Sdn. Bhd, Malaysia). ethanol, methanol, and isopropanol (Systerm), sulphuric acid (95-98% purity, AR/ACS was purchased from R&M), and sodium chlorite (80% purity, Acros Belgium). OPEFB cellulose was

produced in a lab setting utilising ASTM recommended practises (ASTM D 1104-56 and ASTM D 1103-60).

2.2 Preparation of Microcrystalline Cellulose

The preparation of microcrystalline cellulose (MCC) was carried out using published procedure of Ismail *et al.,* [19]. Acid hydrolysis process of MCC was conducted with several concentrations of sulphuric acid (5%, 15%, and 25%). The mixture was treated with an ultrasonicator at 50 °C for three hours after being hydrolyzed in an autoclave for roughly an hour. After the ultrasonication procedure, the suspension was repeatedly rinsed with distilled water to remove all traces of acid (the filtrate displayed a pH that was almost neutral) before being filtered to remove the MCC. The MCC was then dried for 24 hours in an oven to achieve a constant weight before being ground into a fine powder. Three different sulphuric acid concentrations were used to make the MCC: MCC A (5% sulphuric acid), MCC B (15% sulphuric acid), and MCC C (25% sulphuric acid).

2.3 High Pressure Homogenization

Isolation of nanocellulose via high-pressure homogenization (HPH) was carried out based on the work of Ismail *et al.,* [20] There are two mechanical steps involved which were the mechanical disintegration and a high-pressure homogenization method in order to isolate nanocellulose. The treated fibres were mechanically disintegrated to release and soften them as well as to use high shear to reduce the MCC diameter to micron size. The final step in the isolation process for nanocellulose (NC) is high-pressure homogenization (HPH). The MCC suspension was run through a high-pressure homogenizer (Model: APV Model 1000 Homogenizer) with various passing periods of 10, 20, and 30 cycles at a constant pressure of 800 bar. A freeze dryer was used to dry the NC after it had been successfully separated from the MCC suspension at the optimal HPH passing time.

2.4 Characterization Methods

Scanning Electron Microscopy (SEM, Hitachi S2700) analysis was used to evaluate MCC's specific properties and morphology. The samples were examined with a 1000x magnification. Nanocellulose samples were coated with a thin layer of gold using an ion sputter coater, and their morphology were analyzed with a Field Emission Scanning Electron Microscope (FESEM, JSM7600F Tokyo, Japan) with field emission gun operated at 5 kV.

2.5 Dispersion Characteristics of Cellulose and MCC

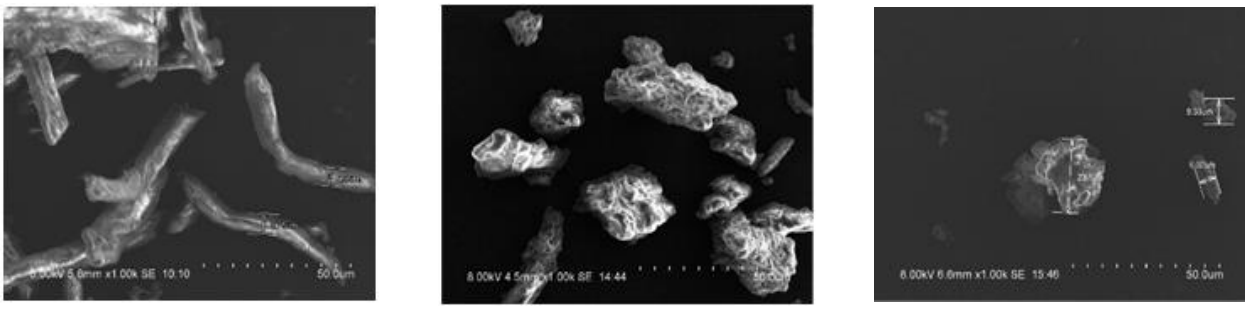
The dried cellulose and MCC were dispersed in deionized water and each dispersion was sonicated using a ultrasonicator at an amplitude of 40 for 1 hour. The dispersion samples were then allowed to stand at room temperature for three days and the observation and photographing process will be starting from the first day.

3. Results

3.1 SEM Analysis of MCC

The morphological structures of MCC after hydrolysis are depicted in Figure 1. The photos show that the morphology of MCC changed after therapy. The morphology of MCC showed that the

diameter size of MCC decreased along with the length of the fibres as the concentration of sulphuric acid rose. Before treatment, OPEFBs' raw fibres are composed up of bundles bound together by lignin and hemicellulose, and their surface morphology is uneven and rough [21]. Due to the depolymerization of cellulose polymers, a shorter chained MCC was produced, which had a different structure from that of cellulose [22]. The results demonstrate that the acid treatment significantly decreased the fibre width, and their shape was comparable to commercial MCC (Figure 1). When compared to untreated fibres, the total diameter of the acid-treated MCC fibres had been drastically reduced to roughly 8 m, and the fibres' length had been decreased to a few microns. This discovery is comparable to those made by Fahma *et al.,* [23], who found that the acid concentration and the acid-to-fibre ratio have a significant impact on the shape and diameter of the MCC formed. The elimination of lignin, hemicellulose, and silica left the MCCs' exterior surfaces with both smooth and irregular forms.



(a) (b) (c)

Fig. 1. The SEM micrograph of MCC (after acid hydrolysis) (a) MMC A (b) MCC B (c) MCC C

3.2 Turbidity Changes of Nanocellulose Solution After the HPH Process

At a constant pressure of 800 bar, MCC A, MCC B, and MCC C solutions undergo 10, 20, and 30 cycles in a high-pressure homogenizer, respectively. Figure 2 shows that from 10 to 30 cycles, the MCC solution at 0 cycle became cloudier. The particle size of cellulose decreased as cycle periods increased from 10 to 30. As the particles are reduced in size and become less entangled, it has been demonstrated that applying additional shearing force could damage the connection between the cellulose and the creation of NC [24]. As the number of homogenization cycles increased, it indicates that the particle sizes decreased. The temperature of the solution would rise with more cycles or at higher pressure, which would result in cellulose breakdown [25].

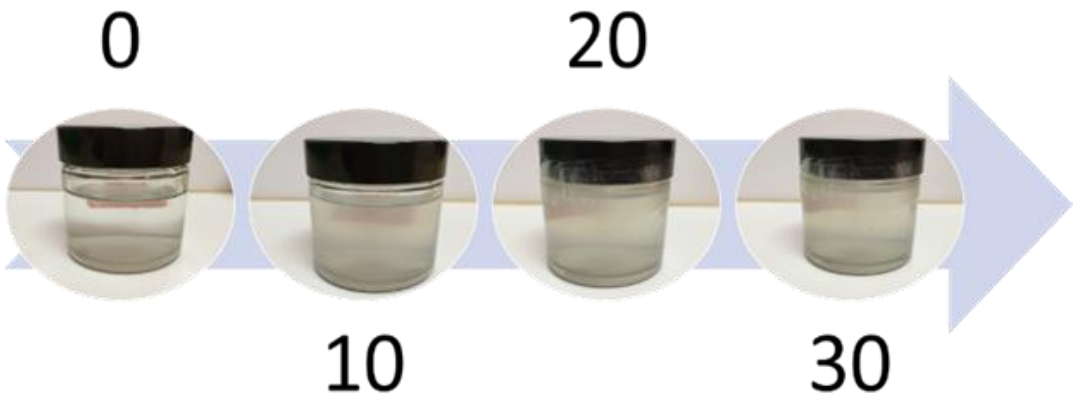
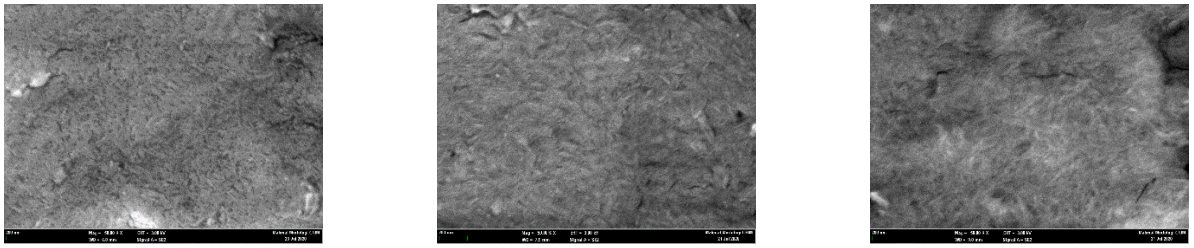


Fig. 2. Observation of turbidity changes of OPEFB particles solution from zero to 30 cycles during the HPH process

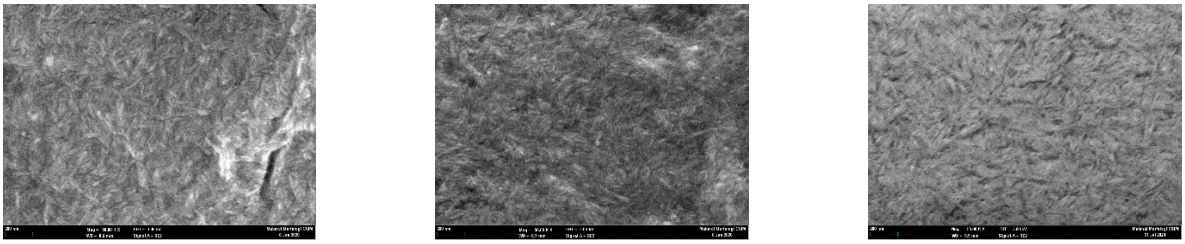
3.3 FESEM Micrographs of Nanocellulose

FESEM was used to verify that nanocellulose (NC) was successfully extracted. Figure 3, Figure 4, and Figure 5 displays FESEM micrographs of the NC for MCC A, MCC B, and MCC C with three distinct passage times (10, 20, and 30). After the HPH treatment, the nanocellulose's diameter had approached the nanometre scale, as evidenced by the measurement of nanocellulose particles at a size of roughly 20 nm. The FESEM micrographs illustrated the structure of nanocellulose to be primarily agglomerated and showed the effectiveness of the combination of chemical and mechanical treatments. The similar phenomena, in which various agglomeration mechanisms took place among the crystal nanocellulose particles during each drying procedure, was described by Michael *et al.,* [26]. All of the NC typically showed equivalent modifications after homogenization, which entails a decrease in diameter size and significant breakage of the cell walls. There were very little changes between the treated NC at various passage periods when the fibres had vanished, and only short rod-like/whisker-shaped structures in the nanometer dimension were observed.



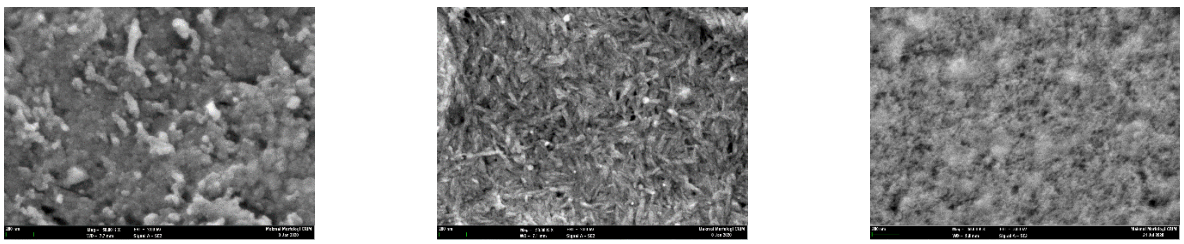
(a) (b) (c)

Fig. 3. FESEM micrographs of the nanocellulose at 5% H_2SO_4 for MCC A at (a) 10 cycles (b) 20 cycles (c) 30 cycles



(a) (b) (c)

Fig. 4. FESEM micrographs of the nanocellulose at 15% H_2SO_4 for MCC B at (a) 10 cycles (b) 20 cycles (c) 30 cycles



(a) (b) (c)

Fig. 5. FESEM micrographs of the nanocellulose at 25% H_2SO_4 for MCC C at (a) 10 cycles (b) 20 cycles (c) 30 cycles

Microscopic observations revealed that the nanocellulose has a smooth surface and the rigid and rod-like shape of nanocrystalline cellulose (NCC) with mostly agglomerates. This effect happens as a result of the NCC chemical structure's hydrophilic activity, which is caused by the presence of hydroxyl (OH-) and hydrogen (H+) groups that form intra-chain and inter-chain hydrogen bonds at the end of the structure, as illustrated in Figure 6 [27]. Abraham *et al.,* [28] revealed that several hydroxyl groups can bind with each of the NCC interfaces because of cellulose's nanosize, thus raising its hydrophilic properties.

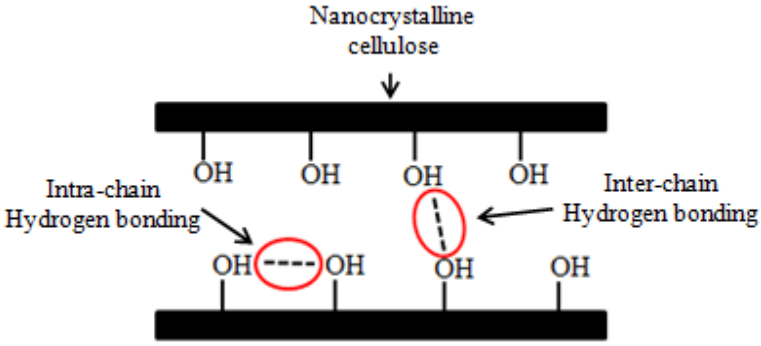


Fig. 6. Inter- and intra-chain of NC bonding chemical structure [27]

As the passing time is increased, all of the NC exhibited decreasing dimensions. Due to the above-mentioned agglomerated network structure, it was impossible to identify the diameter of the NC from microscopic measurements; as a result, it was determined to be below 20 nm using FESEM measurement (Figure 3, Figure 4, and Figure 5). In comparison to previous studies on the isolation of nanocellulose, these findings are the first to produce nanocellulose from the MCC of OPEFB fibres using the chemo-mechanical process. Another related study on the combination of chemical and mechanical HPH processes was done by Pan *et al.,* [29] on the isolation of cellulose nanowhiskers from commercial MCC. Their investigation of different sulphuric acid concentrations effectively leads to a narrower production of cellulose nanowhiskers with less polydispersity and low crystallinity as the acid concentration increased from 20 to 60 wt %. According to Abitbol *et al.,* [30] altering the surface charge or the extent of replacement of sulphate groups on the surface of cellulose nanocrystal suspensions in water may change their stability.

3.4 Stability Test

Figure 7 depicts the dispersion image following three days of storage at 25°C. After three days of observation, cellulose displayed a distinct stratification phenomenon, with the cellulose particles separating into two phases: a transparent phase at the top and a bottom phase that is opaque and rich in cellulose. All of the original MCC and NC samples were disseminated uniformly in water, and their suspensions were stable and homogenous, with no stratification, but the cellulose particle dispersion displayed very low stability in water and no interaction on the first day. The MCCs and NCs experienced precipitation and flocculation after 3 days at 25°C, demonstrating that the particles were relatively unstable in water.

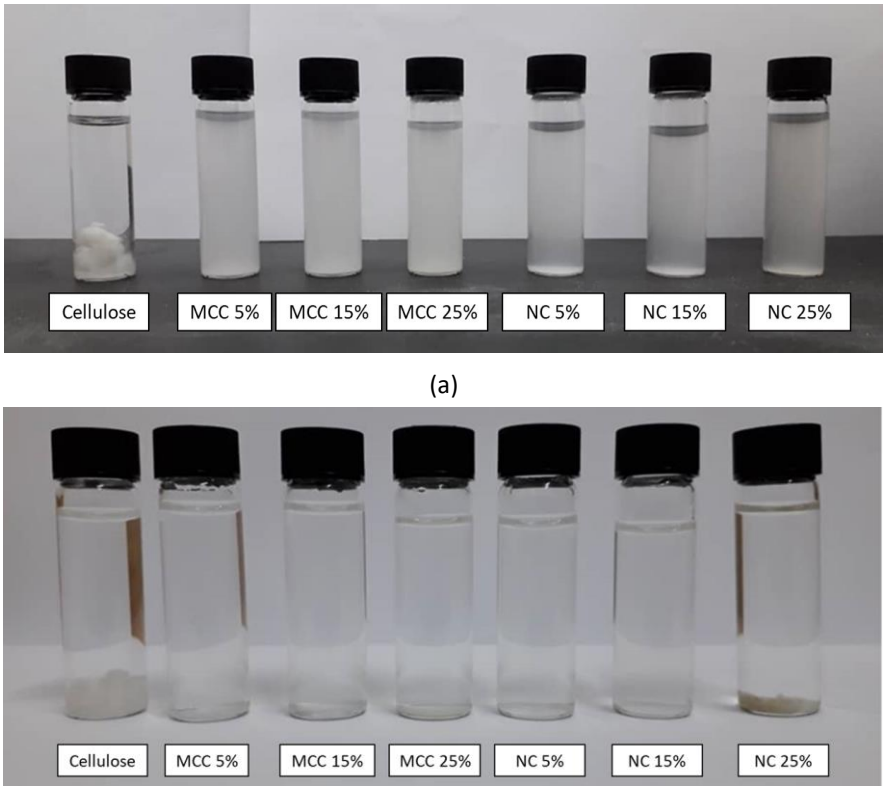


Fig. 7. The visual images of cellulose, MCC and NC dispersion in distilled water were taken after storage from (a) 0 days (b) three days

The net charge is a significant component that affects the stability of MCC and NC particles. In general, better dispersion and stability are correlated with larger absolute zeta potential levels [31]. According to the study's findings, adding sulphate groups to MCC through acid alteration enhanced the MCC and NC particle stability and dispersion.

4. Conclusions

In this work, oil palm empty fruit bunches were successfully processed by a chemo-mechanical method that combined high-pressure homogenization (HPH), mechanical disintegration, and sulphuric acid hydrolysis to extract and isolate nanocellulose. The agglomerated network structure made determining the NC diameter difficult, thus it was assumed to be less than 20 nm based on FESEM micrograph analysis. The sulphate groups were added to MCC through acid modification, which increased the particles' stability and water dispersion. The more stable solution was produced by the comparatively small cellulose particles, indicating that electrostatic interactions were crucial in preserving the nanocellulose particles' stability and dispersibility. Chemo-mechanical treatment was found to be superior in terms of improving the physical qualities of nanocellulose powder and modified cellulose powder as well as reducing the fibre size. This recent discovery offered a significant prospect for the production of nanocellulose from plentiful agricultural waste, which may be profitably used in the creation of new nanocomposites for a variety of industries, such as packaging, automobiles, precast concrete, cosmetics, aerogel, and water filtration.

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