OPTIMIZATION CONDITION OF ISOLATION CELLULOSE NANOFIBRILS USING NaOH ALKALINE TREATMENT

Alny Marlynni binti Abd Majid
Faculty of Applied Sciences
Universiti Teknologi MARA Cawangan Negeri Sembilan
Kampus Kuala Pilah,
72000 Kuala Pilah, Negeri Sembilan, Malaysia
Email: alnymarlynni@gmail.com

Maizatul Saadiah binti Mesni
Faculty of Applied Sciences
Universiti Teknologi MARA Cawangan Negeri Sembilan
Kampus Kuala Pilah,
72000 Kuala Pilah, Negeri Sembilan, Malaysia
Email: myzatulmesni@gmail.com

Abdul Hafidz bin Mohd Raffe
Faculty of Applied Sciences
Universiti Teknologi MARA Cawangan Negeri Sembilan
Kampus Kuala Pilah,
72000 Kuala Pilah, Negeri Sembilan, Malaysia
Email: hafidzraffe@gmail.com

Ahmad Husaini bin Mohamed
Faculty of Applied Sciences
Universiti Teknologi MARA Cawangan Negeri Sembilan
Kampus Kuala Pilah,
72000 Kuala Pilah, Negeri Sembilan, Malaysia
Email: ahmadhusaini@ns.uitm.edu.my

Nor Monica binti Ahmad
Faculty of Applied Sciences
Universiti Teknologi MARA Cawangan Negeri Sembilan
Kampus Kuala Pilah,
72000 Kuala Pilah, Negeri Sembilan, Malaysia
Email: normonica@ns.uitm.edu.my

Sheikh Ahmad Izaddin bin Sheikh Mohd Ghazali
Faculty of Applied Sciences
Universiti Teknologi MARA Cawangan Negeri Sembilan
Kampus Kuala Pilah,
72000 Kuala Pilah, Negeri Sembilan, Malaysia
Email: sheikhahmadizaddin@ns.uitm.edu.my

ABSTRACT

Cellulose nanofibers (CNF) possess unique properties such as renewable, biodegradable and low cost which suitable in various applications. However, the alkaline treatment used to isolate CNF contribute several drawbacks such as swollen/shrinkage the fiber, high agglomeration, disrupting the structure and prevent separation of excess non-structural carbohydrates. Thus to overcome this issues, optimization need to be conducted in terms of concentration, isolation time and temperature. In this present work, the oil palm empty fruit bunch (OPEFB) biomass was pre-treated with sodium hydroxide (NaOH) and sodium hypochlorite (NaClO) to remove non-cellulosic component such as lignin and hemicellulose. The treated OPEFB fiber was then isolated using alkaline treatment varied by three parameters such as concentration of NaOH, time and temperature. The CNF was characterized using Fourier transform infrared (FTIR), Ultraviolet visible (UV-Vis) and Thermal gravimetric analysis (TGA). The optimum condition for isolation of CNF was at 0.5 M NaOH, 55 ºC and 2 hours which has been determined based on the percentage yield obtained. From the FTIR spectra, the absence of band at 1700 cm^{-1} and 1200 cm^{-1} indicate efficient removal of hemicellulose and lignin respectively during pre-treatment. Whereas, band at 897 cm^{-1} confirm no rupture of β-glycosidic linkage occurs during isolation of CNF using alkaline treatment process. The optical transparency analysis using UV-Vis shows higher percentage transmittance above 90% indicating CNF obtained smaller diameter in size. TGA analysis displayed three phase CNF degradation curve indicating water loss followed by pyrolysis of lignin, depolymerization and char oxidation. The onset decomposition temperature of CNF isolated using 1 M NaOH at 55 ºC show the initial degradation start at 250.07 °C while the CNF isolated using 0.5 M NaOH at 55 ºC has slightly lower onset decomposition temperature that is 231.70 °C indication that it has low thermal stability.

Keywords: Cellulose nanofiber, oil palm empty fruit bunch, alkaline treatment, degradation.

INTRODUCTION

Increasing popularity of renewable and non-toxic nanomaterials such as nanocellulose (NC) has attracted greater attention due to its outstanding properties. This properties and characteristics of NC depend on the types of treatment protocol and sources of origin being used. One of the most studied NC is cellulose nanofibers (CNF) which typically having a length to several micrometer and width less than 100 nm that can be prepared from various treatment such as high-pressure homogenization, grinding, TEMPO mediated oxidation and alkaline treatment (García *et al.*, 2016). The outstanding properties that usually been studied by researchers such as low-cost material, renewability, biodegradable, high aspect ratio, specific surface area, high tensile strength and high stiffness that benefited broad range of application such as biomedical, automotive, packaging *etc*. (Kakroodi *et al*., 2014). Like native cellulose structure, CNF are also consisted of dual polymeric regions which are crystalline and amorphous which composed from repeating unit β-D-glucopyronosyl and linked together by β-glycosidic linkage at C1 and C4 position (Fahma *et al*., 2010; Li *et al*., 2014).

In comparison to other isolating CNF methods, sodium hydroxide (NaOH) treatment is much closer to commercial utilization since it is relatively cheaper, less energy consumption, require less volume of solvent and very effective in further removal of non-cellulosic component (Zuluaga *et al*., 2009). Few studies on chemically isolation of CNF using NaOH found that, in consistency of parameter alkaline treatments will affect the structure behavior of CNF such as swollen/shrinkage the fiber, rate of agglomeration, disrupting the tensile strength and resulting the prevention of separation of excess non-structural carbohydrates (Rambabu *et al*., 2015). The alkaline treatment using potassium hydroxide (KOH) conducted by Andrade-mahecha *et al*., (2015) using two stages of alkaline treatment has caused the fiber structure to swollen resulting in physical changes due to strong alkalinity of KOH compared to NaOH. Furthermore, this swelling effect will permit the dissolution and penetration of solvent into the structure. Apart from that, Abe and Yano (2011) states that the rate of agglomeration of fiber are increasing as the concentration of NaOH increasing.

Malaysia is the world's leading oil palm producers which produced an average of 50 million tons of oil palm residues every year. Therefore, the amount of biomass produced are expected to increase up to 100 million tons by 2020 (Aghamohammadi *et al.,* 2016). Due to this situation, the biomass obtained from the oil palm extraction is currently being disposed through uncontrolled disposable way such as land disposal and open burning activities which are discouraged by the Department of Environment in Malaysia. Furthermore, oil palm biomass can create substantial environmental problems when simply left on the plantation fields without proper disposal management (Khalil *et al*., 2012). Thus, many researches and initiative have been done to convert the biomass into something new. The high percentage of cellulose (44.4%) contained in the OPEFB has made it as a useful source for producing CNF (Fahma *et al*., 2010). The isolation of CNF has previously reported by a few researchers using various treatment protocols (Fahma *et al*., 2010; Rosazley *et al*., 2016; Kargarzadeh *et al*., 2017).

Throughout the decades, the interest in CNF isolated in natural resources has been increasing as an alternative to replace the use of synthetic CNF. This is because the synthetic CNF have several drawbacks such as high energy consumption, longer disposal time, expensive and harmful to the environment (Wang and Sain, 2007). Therefore, it seems a worthwhile exercise to explore the potential of natural CNF. In conjunction with the increasing amount of OPEFB biomass produced and some limitation on alkaline treatment isolation CNF such as agglomeration, it is crucial to study the optimization of alkaline treatment. As there is no previous report on the optimization of sodium hydroxide treatment, therefore, the aims of this study were to optimize the sodium hydroxide treatment for isolation of CNF using oil palm empty fruit bunch (OPEFB) fibers based on three (3) dependent parameters which are concentration of NaOH, isolation time and temperature. The properties of CNF obtained were investigated using Fourier transform infrared spectrometry (FTIR), Optical transparency properties analysis, X-ray diffractometer (XRD) and Thermalgravimetric analyzer (TGA).

EXPERIMENTAL

Materials

Oil palm empty fruit bunch (OPEFB) fibers obtained from Sime Darby Plantation Labu, Negeri Sembilan. Chemicals used to isolate CNF are sodium hypochlorite (NaClO) (not less than 10% active chlorine), 1 M sodium hydroxide (NaOH). All the chemicals are reagent grade and were used as received.

Sample preparation

OPEFB fiber was washed with distilled water to remove any impurities and oil. Then, the fiber was dried at 100 °C overnight. The dried fiber was ground using a crusher (model SY-50) at Forest Research Institute Malaysia (FRIM) Kepong and sieved to reduce the size to 0.5 mm.

Pretreatment

OPEFB fibers were treated with ratio (1:10) of 3% NaOH solution for 3 hours at 100 °C with continuously stirs. Then, the treated fiber was filtered and washed with HCl for several times until reached neutrality (pH=7). Then, the treated fiber was further bleaching process with ratio (1:4) NaClO at 80 °C for 2 hours with continuously stir. OPEFB fibers was filtered and washed again with distilled water for several times until reached neutrality (pH=7). The bleaching process was repeated for two (2) times until the color of these fibers changed from dark brown to white fiber. The cellulose obtained was dried in oven at 70 ºC overnight prior for isolation using alkaline treatments (Liu *et al.*, 2016).

Isolation of CNF

The isolation of CNF from OPEFB was done by using alkaline treatment method was conducted according to Zuluaga *et al.* (2009) and Ramli *et al.* (2015) with slightly modification. The bleached OPEFB fiber was mixed with 200 mL at different concentration of NaOH solution (0.5 M, 1 M, 1.5 M) labelled as CNF-C1, CNF-C2, CNF-C3, respectively. The CNF was isolated for 2 (CNF-S1), 4 (CNF-S2) and 6 hours (CNF-S3). Then, the mixture was stirred under constant stirring at different temperature 45 °C (CNF-T1), 55 °C (CNF-T2) 65 °C (CNF-T3). Then, the solution was sonicated at 50 °C for 3 hours. After ultrasonication process, the suspension was washed for several times with distilled water and filtered to obtain the desired product.

Characterization

Fourier transforms infrared spectrometry (FTIR) analysis

The presence of any changes of chemical structure of CNF was recorded by using Fourier Transform Infrared (FTIR) spectrometry (Perkin Elmer 100). Dried CNF were ground and the spectra were recorded in the range 650 to 4 000 cm^{-1}.

Optical transparency analysis

Optical transmission of 0.1% (v/v) CNF was carried out by UV-Vis spectrophotometer (T 80+) in visible range from 400 to 800 nm. UV-Vis spectrophotometer was used in order to determine the light that is absorbed and scattered by CNF suspension.

Thermalgravimetric analysis (TGA)

Thermogravimetric weight loss of CNF in various temperature was carried out by Pyris 1 Thermalgravimetric Analyzer (TGA) (Perkin Elmer) under linear temperature condition. The temperature was set from 25 ºC to 550 ºC at heating rate of 10 ºC/min under nitrogen atmosphere.

X-Ray Diffraction (XRD)

The crystal structure was studied by x-ray powder diffraction measurements using Bruker D8 ADVANCE X-ray diffractometer (Bruker AXS Germany) equipped with Ni-filtered Cu Kα radiation generated at 40 kV and 40 mA. The data were collected at an angular range of 5–80° with exposure time 0.1-0.2s / step. The crystallinity index (CI) was calculated according to the Segal's method (Sunghyun *et al*., 2015):

$$CI = \left(\frac{I_{200} - I_{amorph}}{I_{200}}\right) \times 100\% \qquad \textbf{(Equation 1)}$$

Where I_{200} is the maximum of the diffraction intensity (the 200 plane), and I_{amorph} is the minimum diffraction intensity (between 110 and 200 bands). A comparison of crystallinity of nanocellulose caused by different drying method was evaluated.

RESULTS AND DISCUSSION

Effect of pretreatment

Study by Rambabu *et al*. (2016) and Ching and Ng (2014) show that sodium hydroxide (NaOH) and bleaching with sodium hypochlorite (NaClO) are the common and effective method on removing the non-cellulosic component presence in cellulose structure and to obtain the high purity of cellulose. During pretreatment process, it was observed that both non-cellulosic component was completely delignified with the color changed of the fibers. The color of fibers has changed from brown color to dark brown when added 3% NaOH solution. This process helps on removing hemicellulose presence in the fibers. Then, during chlorite-bleaching, the dark brown color decolorized into white color which helps on removing lignin in the fibers.

Optimization condition of isolation of CNF using alkaline treatment

Three different parameter were used to obtained optimum condition in isolating CNF using NaOH alkaline treatment method which are concentration of NaOH (CNF-C1, CNF-C2, CNF-C3), isolation time (CNF-S1, CNF-S2, CNF-S3) and temperature used during isolation (CNF-T1, CNF-T2, and CNF-T3). Using NaOH treatment, CNF obtained shows white colloidal suspensions and ultrasonication was used to improve the dispersion stability by breaking the inter and intramolecular hydrogen

bonding between the particles producing smaller size of CNF. (Zuluaga *et al.* 2009). Based on the three parameters used to optimized isolation of CNF, CNF-C1, CNF-S1 and CNF-T2 showed the best condition and this was proved based on the highest percentage yield obtained. The percentage yield of CNF-C1, CNF-S1 and CNF-T2 are 23.79 %, 20.40 %, and 28.79 % respectively. Optimization conditions on different concentration of alkali used contribute significant effects on the cellulose content. This was supported by Ariffin *et al*., (2008) and Rambabu *et al*. (2016) indicating that the low concentration of NaOH considered to be the best isolation condition without sacrificing the structure and strength of cellulose itself. This was also to preserve the yield of CNF obtained since repetition of pre-treatment itself has slightly reduced the CNF yield by diminishing the nanocellulose structure (Berglund *et al*. 2016). Furthermore, the optimized condition based on isolation time and temperature was best at 2 hours and 55 ˚C respectively. This condition was concluding by observing better dispersion and high percentage yield compared to others condition.

FTIR spectroscopic analysis

FTIR spectroscopy is a technique used to prove the changes of chemical structure after various treatment were introduced. FTIR spectra in Figure 1 shows that the broad band around 3300 cm^{-1} presented in all spectrum represent the OH-stretching that is equivalent with the intramolecular hydrogen bonding in cellulose (Khawas and Deka, 2016). The band present at wavenumber of 1744.33 cm^{-1} and 1245.56 cm^{-1} in raw OPEFB was contributed by C=O stretching in the acetyl and uronic ester groups of hemicellulose and the ester carbonyl groups in *p*-coumaric acid of lignin (Sundari and Ramesh, 2012). However, these band were completely removed after the pre-treatment and bleaching process. The pre-treatment process using NaOH contributed toward removal of hemicellulose (Zuluaga *et al*., 2009). Whereas Ching and Ng (2014) states that bleaching process using NaClO is an effective method to remove the non-cellulosic material from the fibers.

The band present around 2900 cm^{-1} and 1400 cm^{-1} shows the C-H stretching and CH_2 bending that confirming that there are no rupture or alteration in the cellulose structure (Fahma *et al*., 2010). The ß-glycosidic linkage vibration that linking each monomer in the cellulose structure can be seen around 900 cm^{-1} in all spectrum. From the C-H stretching, CH_2 bending and the ß-glycosidic linkage that present in all spectrum prove that the isolated product is a pure cellulose. The CNF isolated were also proven with the commercial CNF spectra that shows no differences in the spectra.

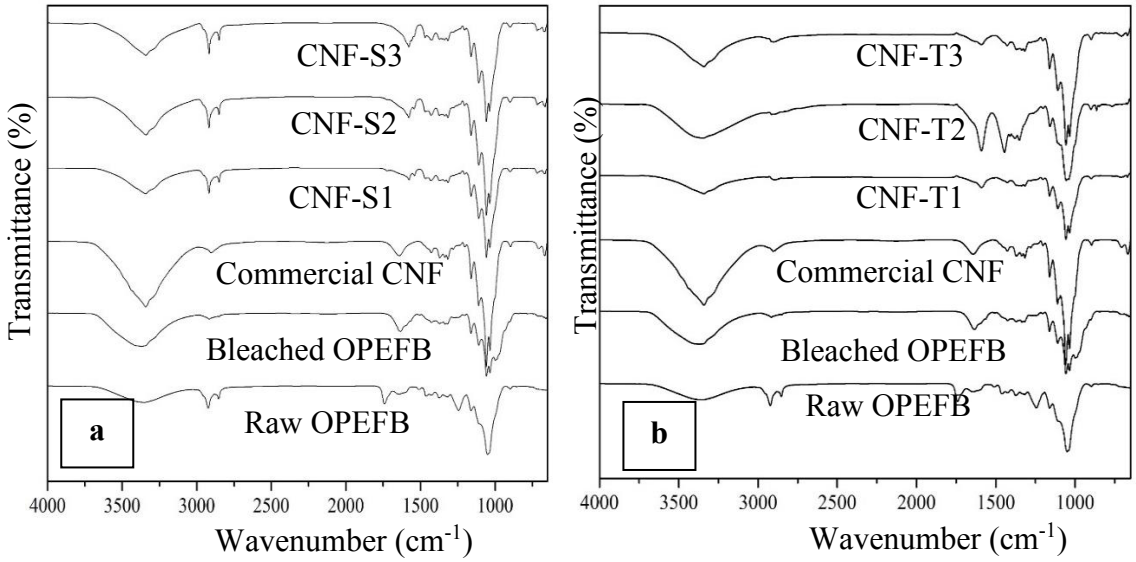


Figure 1: FTIR spectra of Raw OPEFB, Bleached OPEFB, Commercial cellulose with CNF isolated (a) using different NaOH concentration and (b) at different temperature.

Thermalgravimetric analysis (TGA)

The TG curves (Figure 2) shows that all samples undergoes three phases of degradation. From the TG curve, it shows that all samples undergo an initial drop of weight (Phase 1) below 130 ºC with weight loss of 5.65 to 11.58%. According to Sundari and Ramesh (2012), the initial weight loss that occurs at temperature below 150 ºC indicates the weight loss of adsorbed moisture in the sample. The degradation of hemicellulose and cellulose occurs at second phase at 220 to 320 ºC (Ida *et al*., 2013). The onset degradation temperature for raw OPEFB is higher than the other sample due to the high amount of hemicellulose and lignin that lowered the thermal stability of the raw OPEFB (Ching and Ng, 2014). The degradation of the lignin occurs at higher temperature because lignin is the least reactive component of the fibers (Pelissari *et al*., 2014).

The onset degradation temperature of CNF-C2 that was isolated using 1 M NaOH is higher compared to the CNF-C1 that was isolated using 0.5 M NaOH with 247.73 and 231.70 ºC respectively. This shows that the thermal stability of CNF-C1 is higher than CNF-C2. The increasement of isolation time and the different temperature used to isolate the CNF also decrease the onset degradation temperature. The residues remaining in the untreated and treated fibers after heating to 550 ºC constitutes the

presence of carbonaceous materials (Sundari and Ramesh, 2012). The char produced by bleached OPEFB was the highest with 69.36% due to the NaOH pre-treatment. Alkaline pre-treatment will cause the char produced to be higher due to the crystalline cellulose type I which is more flame resistance (Ching and Ng, 2014).

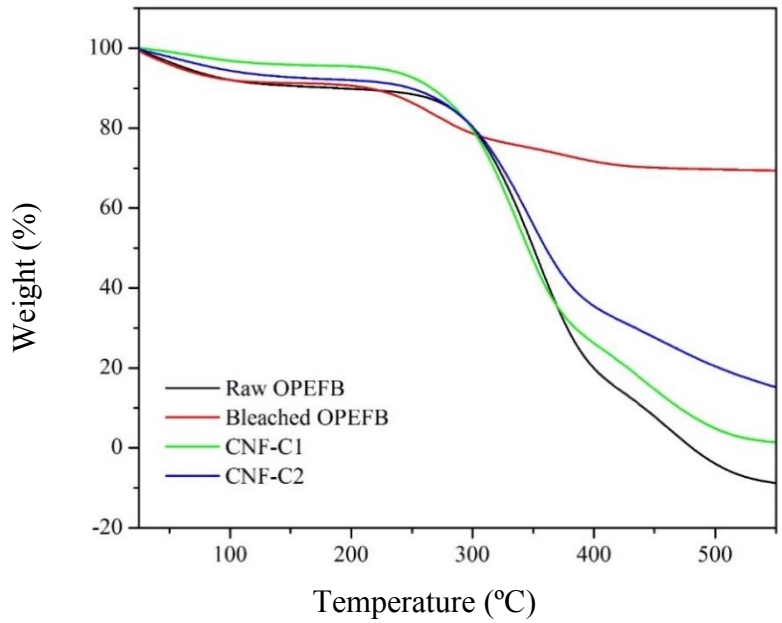
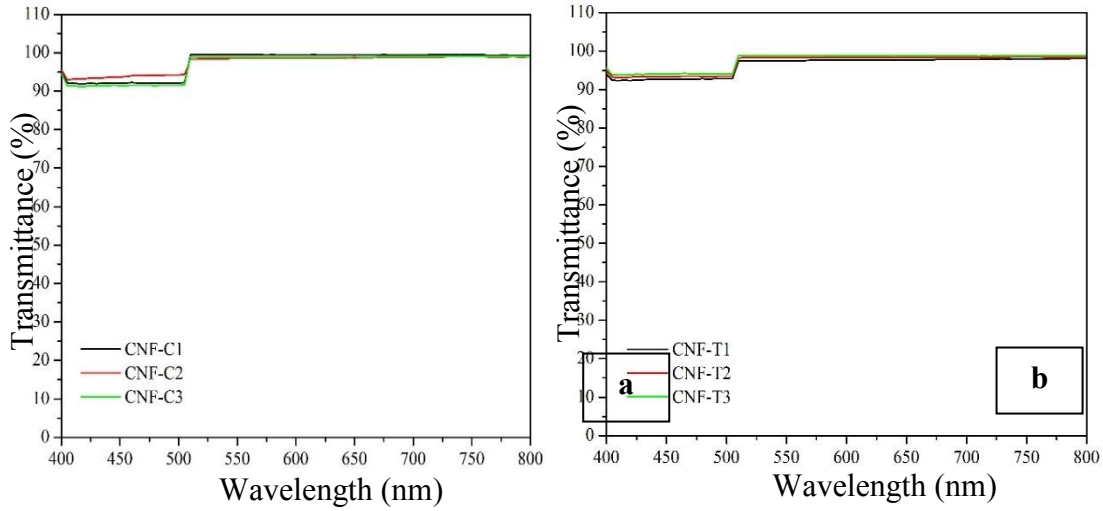


Figure 2: TG curve of Raw OPEFB, Bleached OPEFB and CNF isolated with different NaOH concentration.

Optical transparency properties analysis

The visible light transmission of CNF is shown in Figure 3. The suspended of CNF isolated using different concentration, temperature and time were analyzed by using UV-Vis in visible range of 400 to 700 nm to study the light transmission properties. Saito *et al*. (2007) mention that the transmittance is wavelength-dependent because the light is scattered more when the wavelength is close to the diameter of the CNF.

Figure 3a shows the UV-Vis spectrum for the CNF isolated with different concentration of NaOH. From the figure, it shows that the percentage transmittance for CNF-C1, CNF-C2 and CNF-C3 are 99.6%, 99.0% and 99.1% respectively. The size property of CNF is directly proportional to the percentage transmittance. There is no different in the average size of the CNF when isolated using different concentration of NaOH. The average size of CNF isolated with different temperature increase with the increase of temperature (Figure 3b). The average size of CNF isolated are 730 nm, 740 nm and 745 nm with percentage transmittance of 98.0%, 98.5% and 98.9% for CNF-T1, CNF-T2 and CNF-T3 respectively. However, the average size of the CNF isolated with different isolation time shows that the average size is decreasing with the increasing time (Figure 3c). The percentage transmittance for CNF-S1, CNF-S2, CNF-S3 are 97.3%, 96.3% and 96.4% respectively.



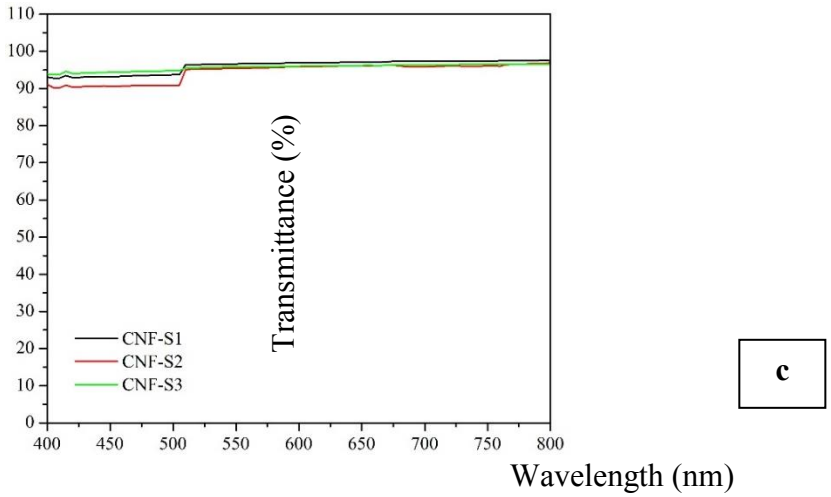


Figure 3: UV-Vis spectrum for CNF isolated (a) different NaOH concentration, (b) at different temperature and (c) at different time.

XRD analysis

To calculate and determine the crystallinity index (CI) for the isolated CNF samples from the NaOH alkaline treatment, x-ray diffractogram and Equation 1 were used. According to Tibolla *et al*., (2016), wide domains in the diffractogram refer to the amorphous region whereas thin and sharp band correspond to the crystalline portion. The x-ray diffractogram patterns (Figure 4) for CNF-C1, CNF-C2 and CNF-T2 shows sharper diffraction band at 2θ = 22.39˚, 2θ = 22.59˚, 2θ = 22.54˚ respectively that correspond to the (200) crystallographic plane of cellulose type-1. The 2θ angles at about 16.20º and 16.15º were assigned to the reflection plane with respect to cellulose type-1 (110). The raw OPEFB, CNF-C1 and CNF-S2 show no distinct patterns with calculated CI are 48.13%, 74.98% and 73.69% respectively. The raw OPEFB shows less crystalline nature due to no alteration in the origin structure. Furthermore, the diffractogram patterns for raw OPEFB conform to the previous report stated that the origin cellulose sources exhibit higher intensities at (200) that at (110) (Ching and Ng, 2014). The CNF-C1 and CNF-S2 patterns demonstrate that by increase the isolation time and the concentration of NaOH does not affect the origin structure of cellulose.

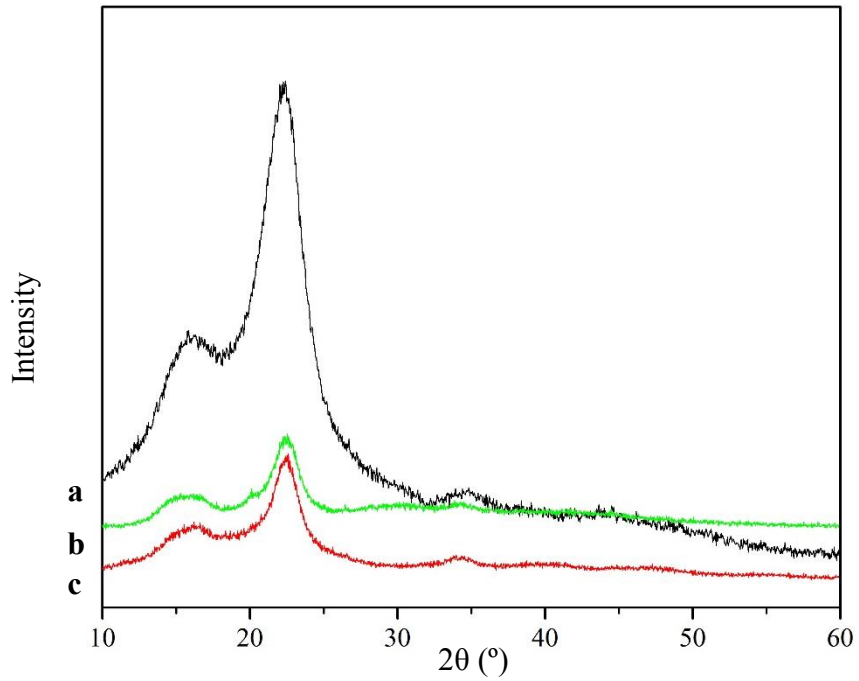


Figure 4: XRD pattern for (a) Raw OPEFB, (b) CNF-S2 and (c) CNF-C1.

CONCLUSION

The present work proved that the pre-treatment using NaOH and bleaching process using NaClO is an effective method to remove hemicellulose and lignin. The CNF was successfully isolated from OPEFB biomass sources with optimum condition at 0.5 M NaOH, 55 ºC and 2 hours. The FTIR shows that, all spectra for CNF isolated resemble to the commercial CNF spectrum. The thermal degradation and the XRD analysis show that the CNF isolated using 0.5 M NaOH has low degradation temperature and has a high crystallinity index. The percentage transmittance for all of the isolated CNF is more than 95% with wavelength ranging from 675 to 745 nm. CNF isolated using 0.5 M NaOH has the highest percentage transmittance that is 99.6%. For further improvement, the isolated CNF can be characterized using Field Emission Scanning Electron Microscope (FESEM) and Transmission Electron Microscope (TEM) to study the change in the morphology of the nanofiber when isolate at different condition. The high number of reactive OH groups on the surface of CNF also provides the possibility for fabricating a wide range of functionalized CNF-based materials for future advanced applications to add value of OPEFB to Malaysia's oil palm industry. Some limitation in this study is the high amount of NaOH used to isolate the small amount of CNF. The information presented in this study reveals the potential of CNF isolated from oil palm biomass for future research. The production of CNF is expected to enhance in nanomaterial field around the globe as it can be used as the reinforcement material, packaging, biomedical and production of nanocomposite, *etc*.

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