Engineering Chemistry
ISSN: 2813-6535, Vol. 4, pp 41-49
doi:10.4028/p-ugJQs1

Submitted: 2022-08-02
Revised: 2022-10-30
Accepted: 2023-04-12
Online: 2023-08-11

Depolymerization of Cellulose Components from Oil Palm Empty Fruit Bunches in Bleaching and Delignification Process

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Keywords: OPEFB, cellulose, bleaching, delignification

Abstract. Oil palm empty fruit bunches (OPEFB) are oil palm industry waste which is indeed abundant. OPEFB waste has been explored to make fiber, cellulose, and its derivatives. Extracting cellulose can depolymerize several components, including hemicellulose, lignin, and color elements. The combination of bleaching under acidic conditions and alkaline delignification gave different changes, one of which depends on the type of bleacher, concentration, reaction time, and the number of stages carried out. This study aimed to examine the effect of $NaClO_2$ concentration and bleaching time on the physical and chemical characteristics that indicate the depolymerization process of OPEFB fiber. Depolymerization was shown by cellulose color, purity, and crystallinity changes. The bleacher concentration and bleaching time affected the shift in cellulose mass. High concentrations of bleachers will reduce the cellulose mass significantly. The chromophore elements OPEFB was depolymerized. The change was shown in color from brown to gray-white with different brightness intensities. The XRD test indicated that there was a correlation between low crystallinity with the use of high bleacher concentrations.

Introduction

Chang [1] stated that empty fruit bunches are solid residues from the palm oil processing industry estimated to reach 20 percent of the weight of fresh fruit of *Elaeis guineensis*. Oil palm Empty Fruit Bunches (OPEFB) weigh approximately 3.5 kg, have a thickness of up to 130 mm, a width of 300 mm, and a length of 300 mm. OPEFB is composed of several essential substances that can be utilized and processed into other materials with more economic value. The cellulose content in OPEFB ranges from 23.7 - 65.0%. Although classified as agricultural waste, OPEFB can produce a high-quality final product.

Cellulose is a linear homopolymer composed of *D-anhydroglucopyranose* units consisting of (1-4)-glycosidic bonds. Natural cellulose, also known as cellulose I, is a semi-crystalline polymer arranged in parallel structures and is not the most stable crystalline form [2]. The regenerated cellulose product, commonly known as cellulose II, has the same molecular formula $(C_6H_{10}O_5)n$ as cellulose I but is more stable and can be molded into certain products such as membranes, hydrogels, aerogels, and fibers [3,4].

Getting cellulose from OPEFB, delignification, and bleaching processes were undertaken. In the extraction process, a delignification process is first carried out and followed by bleaching to get the whiteness of cellulose. Based on previous studies, the bleaching process under acidic conditions can be carried out early before alkaline delignification. The bleaching condition will degrade the OPEFB color components. It will also help open the cell wall structure of OPEFB. The release of lignin inside the cell wall in the alkaline delignification process will be more accessible. The acidic bleaching process can depolymerize the structure by reducing hemicellulose, lignin, and extractive impurities.

The cellulose extracted from the fiber was affected by the pretreatment conditions, such as the chemical type, concentration, duration, and temperature of the hydrolysis treatment. Alkaline treatment only partially removed hemicellulose and lignin. Further chemical treatment through a bleaching process can obtain a higher cellulose content. The research applied acidic bleaching before delignification to degrade the brown chromophore element of OPEFB and produce high-purity cellulose. Depolymerization was indicated by changes in color, cellulose content, and crystallinity, which will later become a reference for the derivate product. This study aimed to examine the effect of $NaClO_2$ concentration and bleaching time on the physical and chemical characteristics that indicate the depolymerization process in OPEFB fiber.

Experimental Methods

Materials and Equipment. OPEFB was obtained from PT Nurciptasari Moeda Sentosa, South Kalimantan, NaOH (Merck), $NaClO_2$ (Clover Chemicals Ltd), glacial acetic acid (Merck), H_2SO_4 (Merck), and distilled water. The equipment, such as a beaker glass, hotplate stirrer, spatula, filter paper, filter cloth, X-Ray Diffraction (XRD) (Rigaku MiniFlex HyPix-400 MF (2D HPAD)-USA), and colorimeter CIELAB lab tool apps, were used.

Preparation of OPEFB. OPEFB was collected after the threshing process from the Palm Oil Mill. OPEFB was washed using hot water, manually separated into stalks and grains, then rinsed up to 4 times using clean water. Furthermore, the fibers were soaked with a 2% soap solution (1:4) to remove residual oil and dust for 5 h and rinsed with clean water. They were drained and dried in an oven at 60 ºC for 48 h. Dried OPEFB, cut into 5 cm, ground, and sifted on a 30 mesh sieve.

Cellulose Extraction Process (Bleaching and Delignification). Ten grams of OPEFB fiber were bleached using $NaClO_2$ according to concentrations (1.5, 3, and 4.5%) with a ratio of 1: 25 (w/v) at a temperature of 70-80ºC for variations of 1, 2, and 3 h under acidic conditions of pH (4 - 4.5) using acetic acid. The bleached fibers were dried and weighed. The bleached fibers were delignified using a 10% NaOH solution with a ratio of 1:20 at room temperature. Delignified cellulose was washed with distilled water and dried. The cellulose is refluxed for 30 minutes with distilled water for washing and continued by drying at 60 °C for 24 h.

Parameter of Analysis. The cellulose was measured by its yield after the bleaching and delignification process. The cellulose was analyzed, including moisture content, ash content, water extractive material, hemicellulose, cellulose, and lignin content. Color cellulose was analyzed using CIE L*, CIE a*, and CIE b* parameters and crystallinity test using X-Ray Diffraction (XRD).

Test Method for Cellulose. Analysis of fiber components was carried out using the Chesson method [5]. A one-gram powder sample was added with 150 mL of distilled water and refluxed for 2 h. The solution was filtered and washed until the pH was neutral and then dried at 105 °C, weighed, and calculated using Eq. 1. The residue 1 obtained was added with 150 mL of 0.5 M H_2SO_4 and refluxed for 2 h. The sample was then filtered and washed until the pH was neutral, dried at 105°C until dry, then weighed and calculated using Eq. 2. The residue 2 obtained was added with 10 mL of 72% H_2SO_4 and macerated for 4 h in room temperature. The sample was added with 150 mL of 0.5 M H_2SO_4, refluxed for 2 h, filtered, and washed until the pH was neutral. The residue was dried at 105°C until dry, then weighed and calculated using Eq. 3. The calculation of lignin content using Eq. 4. is as follows:

$$water - soluble\ extractive\ value\ (\%) = \frac{initial\ mass - mass\ of\ residue\ 1}{initial\ mass}\ x\ 100\% \qquad \text{(Eq. 1)}$$

$$hemicellulose\ content\ (\%) = \frac{mass\ of\ residue\ 1 - mass\ of\ residue\ 2}{initial\ mass}\ x\ 100\% \qquad \text{(Eq. 2)}$$

$$cellulose\ content\ (\%) = \frac{mass\ of\ residue\ 2 - mass\ of\ residue\ 3}{initial\ mass}\ x\ 100\% \qquad \text{(Eq. 3)}$$

$$lignin\ content\ (\%) = \frac{mass\ of\ residue\ 4}{initial\ mass}\ x\ 100\% \qquad \text{(Eq. 4)}$$

X-Ray Diffraction. X-ray diffraction (XRD) was carried out to study sample crystallinity. The sample patterns of all cellulose samples were pressed to form pellets and recorded on an X'Pert X-ray diffractometer using Ni-filtered Cu Kα radiation (30 kV and 30 mA). The diffraction intensity was measured between the Bragg angle (2θ) of 3-90º. Scan speed of 10 º/min, step width of 0.02º. The crystallinity index (CrI) was calculated by the Segal formula [6] using intensity measurements at 22.5º and 18.5º (amorphous) 2θ:

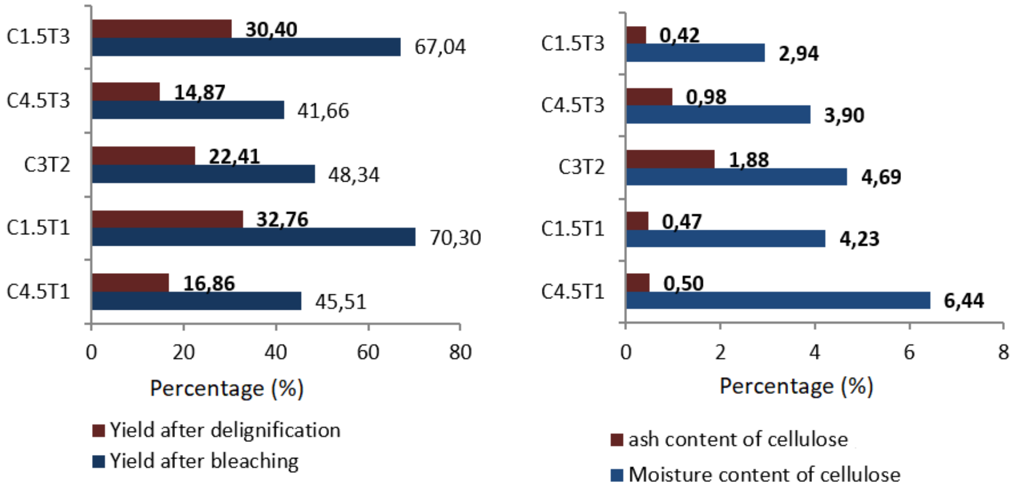
$$CI\ (\%) = (I_{002} - I_{am}/I_{002})\ \text{x}\ 100\% \qquad \text{(Eq. 5)}$$

I_{002} indicates the maximum intensity at 2θ = 22.0-23.00º, I_{am} is the lowest intensity corresponding to the value of 2θ near 18.0º.

CIE-Lab color. CIE-Lab color measurement using colorimeter lab tools apps (modification from Kristanoko et al. [7]) The smartphone camera used is Android Vivo Y 20 SV2029 with a triple macro camera specification of 13MP, +2MP, +2MP. Lighting uses 15 Watt LED lamps. The sample is placed on the cup evenly, the distance between the camera and the sample is 12 cm without flash, and the color scan is carried out in live mode, which is carried out at 25 random points on the sample. The colorimeter will convert the recorded color into CIE *L, CIE *a CIE *b, and export data via email.

Results and Discussion

Yield, Moisture Content, and Ash Content. The bleaching process using $NaClO_2$ is generally intended to remove a material's color components. Acidic bleaching at pH 4 - 4.5 will depolymerize chromophores, lignin, cellulose, hemicellulose, and crystallinity cellulose. The reaction can be observed by decreasing fiber mass or yield after the bleaching and delignification process on the cellulose from OPEFB. The changes in cellulose extracted with variations in bleaching conditions are presented in Fig. 1.



(a) (b)

Figure 1. a) Yield profile bleached and delignified cellulose; b) moisture and ash content profile of cellulose (C = $NaClO_2$ concentration (%); T = bleaching time (hours))

The bleaching process was carried out in the first stage, and Fig. 1a shows that the concentration of $NaClO_2$ will significantly affect the depolymerization. The bleaching process with a higher $NaClO_2$ concentration caused decreasing the fiber mass after bleaching. According to Kim [8], acid hydrolysis of lignocellulosic can depolymerize cellulose and hemicellulose into monomers without pretreatment.

Implementation bleaching at 1.5% $NaClO_2$ for 1 and 3 h in pH 4-4.5 resulted in yields of 70.30% and 67.04%, which were greater than those of 4.5% $NaClO_2$ and 3% $NaClO_2$. A high yield does not indicate a high purity of cellulose. There was still a lot of lignin content left in the fiber. After the alkaline delignification process, the cellulose yield decreased. Yield reduction from bleaching to delignification varied at 1.5% and 3% $NaClO_2$ concentrations in the range of 53.5%, while at 4.5% $NaClO_2$ concentrations, the yield reduction reached 63.6%. $NaClO_2$ concentration significantly causes depolymerization of color, lignin, hemicellulose, and other impurity components. Fig. 1b shows that the bleaching and delignification processes significantly reduced the ash content of cellulose, while the water content of cellulose was not significantly different. Fiber contains minerals such as silica; the bleaching and delignification process can reduce the mineral content in cellulose.

Color of Cellulose. The color of the oil palm empty fruit bunches was originally brown. After the bleaching process will fade to a whitish yellow, and if continued by the delignification, the color will be grayish white. The brightness of the cellulose color after the bleaching and delignification stages can be described by the CIE *L, CIE *a, and CIE *b values. OPEFB has the lightness value of CIE *L shown on a scale of 61.25, and after the bleaching comes to 80.15 – 84.97. The treatment of 1.5% $NaClO_2$ with a bleaching time of 1 h resulted in a lower CIE L* score than the others, especially when compared to the 4.5% concentration at the same bleaching time. A low concentration of $NaClO_2$ needed a longer bleaching process to get a high lightness level. However, it will provide intensive depolymerization at high concentrations and treated at a long bleaching period. However, the higher $NaClO_2$ concentration positively correlated with the reduction of cellulose produced.

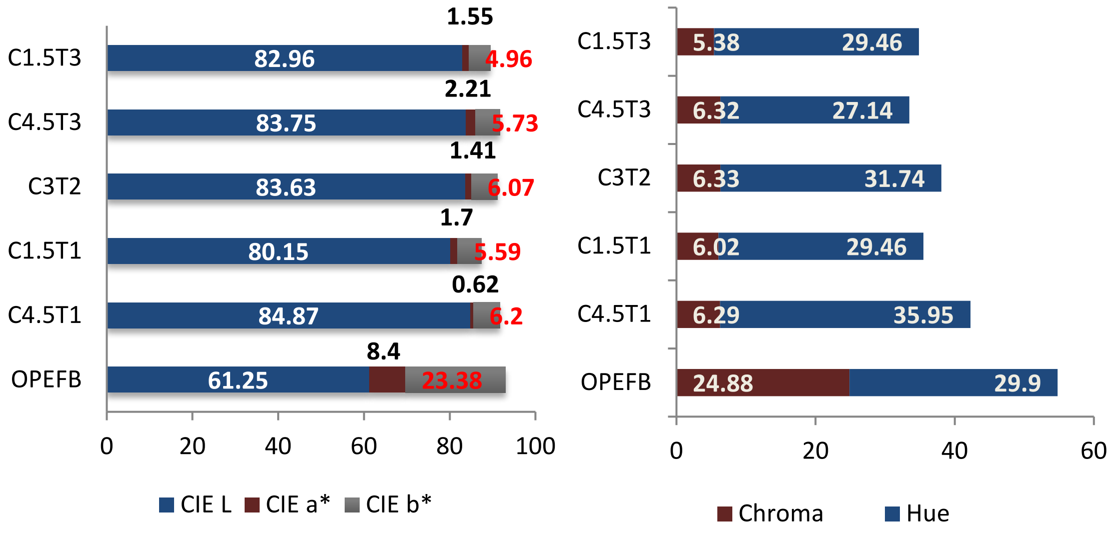


Figure 2. a) Pattern of CIE L, CIE a*, and CIE b*; b) Chroma and Hue pattern of cellulose from OPEFB (C = $NaClO_2$ concentration (%); T = bleaching time (hours))

The CIE a* value (Fig. 2a) indicated the color intensity in the red-green range, and the CIE b* value indicated the yellow-blue range. Different bleaching treatments showed that the CIE a* and CIE b* values were relatively not significantly different. However, when compared with OPEFB, it showed a significant color reduction. The value of CIE a* of OPEFB is greater than cellulose and indicated the red tendency in OPEFB is more visible. Similarly, the value of CIE b* on OPEFB was 23.38, it was higher than that of cellulose in the range of 5-6. a higher positive CIE b* value leads to a stronger yellowish color.

The chroma and hue values among bleaching treatments did not significantly differ (Fig. 2b). The chroma value of cellulose averages 6.00, it’s different from the chroma’s OPEFB is 24.88. and the hue value of OPEFB and its cellulose were 27.14 – 35.95. In the context of color, hue refers to purity, hue is the main identity of color so that it can be easily distinguished from other colors. chroma is the degree of color strength or intensity of the color. The strongest colors are those with high chroma while the less intense ones are those with low chroma.

Brightening effect that can be caused by the presence of hexanuronic acid groups in the bleaching process. Hexenuronic acid is sensitive to acidic conditions, the hydrolytic stage can be carried out either as part of the long-term chlorine stage at high temperatures to produce high brightness. This step aims to achieve selective hydrolytic degradation of hexenuronic acid groups into 5-formylfuroic acid and furoic acid [9].

The color change in OPEFB fiber (Fig. 3) shows a chromophores gradation after the bleaching process, which was naturally from brown to white in a color range of light brown to yellowish white. This degradation is strongly affected by the concentration of bleach and the duration of bleaching. In the bleaching process, some lignin removal occurs, and decreasing lignin will be continued in the alkaline delignification process (Fig. 3) so that the color changes in the cellulose are more visible than the OPEFB color. The success of the bleaching process determines the rate of color change from brown to white under acidic conditions before delignification. The main purpose of chemical pulp bleaching is to remove residual lignin and chromophore in the pulp without affecting the strength of the pulp. It's achieved through a multistage process by delignifying and further brightening the pulp [10].

Figure 3. Color of a) OPEFB fiber; after the bleaching process b) C1.5T3, c) C3T2, d) C4.5T3; after the delignification process e) C1.5T1, f) C1.5T3, g) C3T2 and h) C4.5T3 (C = $NaClO_2$ concentration (%); T = bleaching time (hours))

The leaching of color components in OPEFB can be seen in the bleaching process, where the liquid changes from yellowish white to brownish yellow to dark brown (Fig. 4). The oxidation of the pulp with chlorine dioxide takes place at a temperature of about 65 - 75 °C and with reaction time 30 - 45 minutes. The initial consumption of chlorine dioxide by the pulp is very fast and within seconds, subsequently, the chlorine dioxide concentration will decrease to a considerable extent [11]. Changes in the color intensity of the bleach solution correlated with the duration of bleaching. The change to dark brown will begin after 45 minutes of the bleaching process at a temperature of 80°C. The color change is a cumulative element of chromophore and lignin from OPEFB fiber.

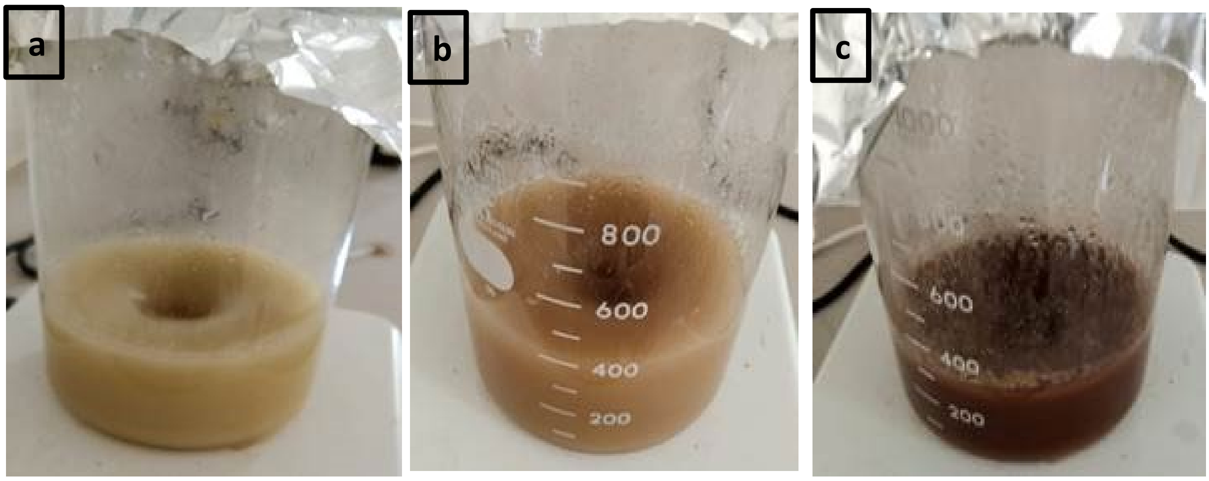


Figure 4. Changes in the color of the bleaching liquid at a) the beginning; b) the middle and c) the end of the bleaching process

Cellulose and Lignin. The alkaline delignification process was less optimal in reducing the lignin content in cellulose, so it must be combined with a bleaching process under acidic conditions in the early stages of extraction. In the cellulose extraction process, reducing lignin content required reactivity to open the cell wall. Lignin is covalently bonded to cellulose, so it requires high reactivity to break the bond, making it easier to reduce the lignin content.

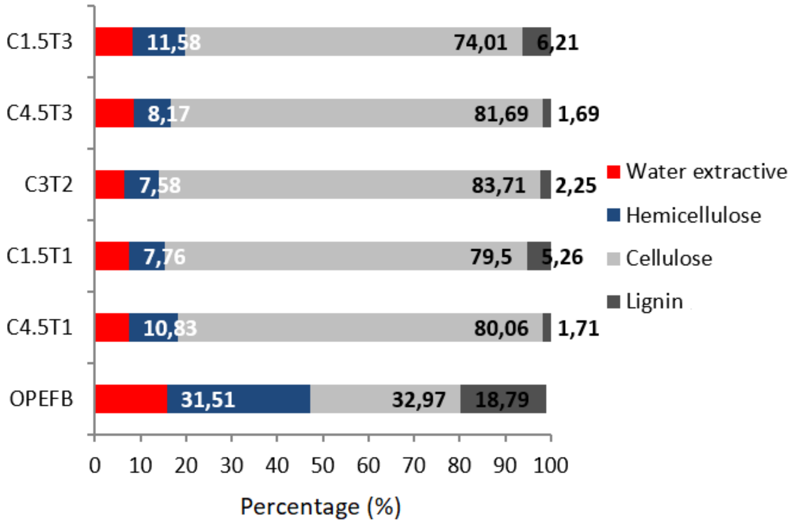


Figure 5. Content of water extractive, hemicellulose, cellulose, and lignin of OPEFB cellulose ($C = NaClO_2$ concentration (%); T = bleaching time (hours))

The cellulose content of OPEFB is 32.97%. Fig. 6 describes that the concentration of $NaClO_2$ in the bleaching process affects the increase in cellulose. Treatment of bleaching in 3% $NaClO_2$ for 2 h at 70 - 80 °C resulted in cellulose being 83.71%. At concentrations of 3% and 4.5% tended to produce a significant reduction in lignin in the range of 1-2%, while the hemicellulose relatively decreased from 31.51% to a range of 7.76-15.77% Hemicellulose had an irregular structure, it is more amorphous than cellulose, so more readily hydrolyzed by acids to monomers. On the other hand, under acid conditions, it can be easily degraded to decomposition products, including furfural.

The extraction process with acid will cause hemicellulose to degrade, which is indicated by a decrease in the chain length. Acidic conditions will boost the autohydrolysis of hemicellulose glycosidic bonds, producing hemicellulose with low molecular weight and water-soluble [12]. Acidic conditions using acetic acid will contribute to hydronium ions that enhance the extraction kinetics. Chio et al. [13] stated that lignin is a complex structure and has low reactivity. The separation of lignin from the cellulose structure can be carried out by mechanical, thermochemical, chemical, and biological catalysis. It is known that acid hydrolysis is widely used for depolymerization. The use of acetic acid to lower the pH can increase the depolymerization of lignin at the O-4 bond.

This research carried out the bleaching process under acidic conditions, followed by alkaline delignification. Lignin depolymerization can also occur in alkaline hydrolysis using NaOH or KOH. Using NaOH was proven to be able to depolymerize four times compared to without alkaline. The research was conducted treatment of 10% NaOH as a base catalyst in the delignification process and carried out at room temperature. Evans et al. [14] stated that strong bases such as NaOH and KOH were able to convert and produce depolymerization products compared to weak bases. Alkaline delignification at low concentrations could not catalyze the depolymerization of lignin in water [15]. At the same concentration of 4.5% $NaClO_2$ with a bleaching time of 1 h and 3 h, cellulose still has lignin content ranging from 1.70%, while at a concentration of 3% for 2 h, there remains 2.25% lignin residue.

Bleaching at 4.5% $NaClO_2$ for 3 h tends to give higher cellulose and lower hemicellulose than bleaching for 1 h. This phenomenon is not in line with bleaching at 1.5% $NaClO_2$, where the cellulose content of 1 h of bleaching time is higher, and hemicellulose is lower than 3 h of bleaching time at the same concentration. According to Karagöz et al. [16], the formation of carboxylic acids during depolymerization can lower pH and enhance repolymerization.

X-ray diffraction. OPEFB is a type of wood fiber that is quite sturdy. Intensive bleaching and delignification treatment must be carried out to depolymerize it so high-purity cellulose can be obtained. Impurities in OPEFB are not only components of hemicellulose and lignin but also a waxy layer or wax on the outer wall. The grinding process in the initial treatment will help facilitate the depolymerization reaction by chemical components, both acids, and bases.

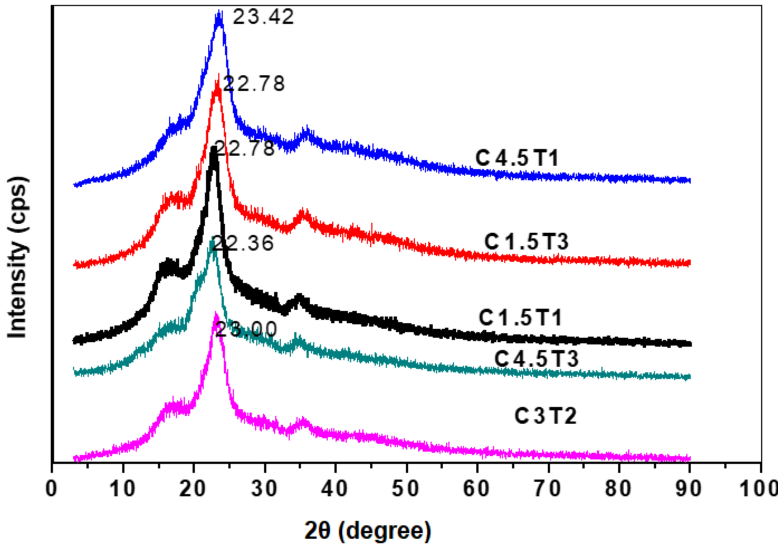


Figure 6. Correlation of 2θ and intensity on OPEFB cellulose

Based on Fig. 6, the 2θ peaks resulted in the range of 22 - 23°, while the amorphous was in the range of 16-18°. The low crystallinity indicates that the amorphous structure is high. The effect of $NaClO_2$ concentration as a bleacher on crystallinity has not been fully explained. Based on Table 1, bleaching at $NaClO_2$ of 3% for 2 h resulted in crystallinity of 82.29%, which is higher than at a concentration of 1.5%. Bleaching at 1.5% and 4.5% $NaClO_2$ concentration for 3 h gave lower crystallinity. Bleaching at $NaClO_2$ of 4.5% in excess time (3 h) will not only reduce amorphous lignin and hemicellulose but will be able to erode the crystalline structure. On the other hand, the crystallinity index can be related to the phenomenon of crystalline polymorphism of cellulose. According to Nam et al. [17], irreversible polymorphic can changes in the crystalline structure of cellulose but the mechanism has not been fully explained. The bleaching process under acidic conditions and alkaline delignification will affect the rearrangement of the cellulose chain, which will later change cellulose I to cellulose II and have specific thermodynamic properties [18].

The crystallinity of cellulose can be increased by acid hydrolysis treatment. In this study, the crystallinity of cellulose becomes important to support the quality of microcrystalline cellulose (MCC). The crystallinity of cellulose is important in determining thermal stability, elasticity, absorption efficiency, and increasing stiffness [14]. Furthermore, high crystallinity MCC will also be able to support the mechanical strength of the hydrogel formulation.

Table 1. Crystallinity Index of cellulose from oil palm empty fruit bunches

Treatments	2θ I_{002} [°]	2θ I_{am} [°]	Intensity [cps °]		FWHM [°]	Crystallinity Index [%]
			I_{002}	I_{am}		
C4.5T1	23.58	17.1	9699	1999	3.67	79.39
C1.5T1	23.09	16.16	11520	2282	2.94	80.19
C3T2	23.01	17.47	4942	875	2.19	82.29
C4.5T3	22.55	18.19	6789	3006	3.69	55.72
C1.5T3	23.18	16.25	7229	2828	3.10	60.88

Note: C = $NaClO_2$ concentration T = bleaching time; FWHM : Full Width at Half Maximum

Table 1 explains that the FWHM of cellulose resulting from 4.5% $NaClO_2$ has a higher value than the others, followed by 1.5% $NaClO_2$ and the lowest at 3% $NaClO_2$. The FWHM value also shows a non-linear relationship with the crystallinity index. Adjacent peaks at positions (110) and (200) even overlap to a greater extent, increasing the intensity between them and resulting in smaller crystallinity values. The data showed that a smaller FWHM value indicates a higher CI. FWHM effect on CI was more significant for cellulose Iβ than cellulose II [17].

Summary

Extracting cellulose by applying bleaching under acidic conditions and delignification provides OPEFB'cellulose purity by reducing some components were hemicellulose, lignin, and other impurities. Depolymerization occurs in the color element (chromophores), where the color of the fiber fades from brown to grayish white with the intensity of brightness depending on the bleacher concentration and the bleaching time. It will be positively correlated with the recovery of cellulose in it. From the results of the XRD test, the effect of bleacher concentration in acidic conditions and the duration of bleaching did not show a completely linear relationship, the bleaching period of 3 h gave a lower degree of cellulose crystallinity effect than the 1 and 2 h bleaching time.

Acknowledgments

The authors would like to thank Gadjah Mada University for its financial support for this research. This research was conducted using the Research Grant Fund in 2022 based on the Letter of Assignment No. 3550/UN1.P.III/Dit-Lit/PT.01.05/2022.

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