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Effect of delignification and bleaching stages on cellulose purity of oil palm empty fruit bunches

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Abstract. Oil palm empty fruit bunches (OPEFB) have not been properly utilized by most palm oil mills and communities in Indonesia. The processing and utilization of OPEFB by palm oil mills are still limited. Due to high cellulose contents, it potentially made into various bioproducts, especially as biomaterials. Therefore, a proper extraction technology is needed to obtain a high level of cellulose purity because alkaline treatment can solely remove a part of hemicellulose and lignin. A higher cellulose content can be obtained by further chemical treatments through the bleaching process. However, no information regarding the difference of the isolation performance method to produce cellulose purity by delignification first or vice versa bleaching as an initial stage. The research determined the purity of EFB cellulose with differences in the sequence of bleaching and alkaline delignification stages. The three methods were carried, namely the first method by bleaching using sodium chlorite 3% two times, followed by delignification of NaOH 10% at room temperature, second method with the same conditions but delignification of NaOH 10% at a temperature of 70-80°C and third method by delignification and continued bleaching under the same conditions. The results showed the bleaching method of sodium chlorite of 2 cycles and continued by alkaline delignification (NaOH 10%) at room temperature resulted in higher cellulose purity than other methods. The FTIR test results indicated the detection of the C-O functional group at a wavelength of 1196 cm^{-1} and the C-H functional group at a wavelength of 2967 cm^{-1}.

1. Introduction

The conversion of fresh palm fruit bunches into palm oil in Indonesia produces several wastes, such oil palm empty fruit bunches (OPEFB) which are quite abundant. Indonesia as the producer of palm oil in the world has the largest area of 14.456.611 Ha with a production of 47.12 million tons (2019). It is estimated that in 2021 Indonesia has an area of 15 million Ha with a production of 49.71 million tons[1]. If the OPEFB waste is 25% (12.42 million tons), and only 10% is processed (1.24 million tons) then there are 11.18 million tons of untapped OPEFB waste potential left [2].

Those waste have not been used correctly by most palm oil mills and communities in Indonesia. Processing and utilization of OPEFB by palm oil mills are still very limited. Most of them usually burn OPEFB in incinerators, or other alternatives are open dumping, used as mulch in oil palm plantations, or processed into compost.

OPEFB as lignocellulosic biomass, contains cellulose, hemicellulose, and lignin. The cellulose content of OPEFB is 23.7-65.0%, while the hemicellulose content is 20.6-33.5% and the lignin content is 14.1-30.5% [3]. The high content of cellulose makes it one of the raw materials that have the potential to be developed into various bio products, especially in the field of biomaterials.

Chemical treatment is the most effective way to obtain higher purity cellulose. The combination of chlorite bleaching, alkali treatment, and acid hydrolysis are commonly used to extract cellulose [4]. Chlorite bleaching function as an extracting holocellulose from raw cellulose fibers. The process removes most of the lignin inside the fibercausing defibrillation. Kargarzadeh et al. [5] stated the diameter of the bleached kenaf bark fiber is smaller than the diameter of the raw fiber. Alkali treatment is used to dissolve the lignin and residual pectin and hemicellulose, while acid hydrolysis is used to degrade amorphous cellulose.

Alkali treatment is only available to partially remove hemicellulose and lignin. A higher cellulose content can only be obtained by further chemical treatment with the bleaching process. Therefore, the bleaching process carries the desired cellulose purity. The research examined the effect of the implementation of the bleaching stage at the beginning and after the delignification process on the purity of the cellulose obtained by limiting the concentration of sodium chlorite and sodium hydroxide used.

2. Materials and methods

2.1. Materials and equipment

The OPEFB from PT Nurciptasari Moeda Sentosa Kalimantan Selatan were taken, NaOH, $NaClO_2$, glacial acetic acid, H_2SO_4, and distilled water. Some equipments such as a beaker glass, hotplate stirrer, spatula, filter paper, filter cloth, Fourier transform infrared spectroscopy and colorimeter CIELAB lab tool apps were used.

2.2. Preparation of OPEFB material

OPEFB was treated after the trashing process from the palm oil mill then were washed using hot water and manually separated into stalks and grains. The OPEFB were rinsed up to 4 times using clean water. Furthermore, the fibers were soaked with 2 % soap (1: 4) to remove residual oil and dust for 5 hours and then rinsed with clean water. They were drained and dried in an oven at 60ºC for 48 hours. Dried OPEFB were cut into 5 cm each, then ground and sifted on a 30 mesh sieve.

2.3. Cellulose extraction process (delignification and bleaching)

10 grams of OPEFB fibers were bleached using $NaClO_2$ solution in the ratio of 1: 25 (w/v) at 70-80ºC for 2 hours as much as two cycles. The bleached fiber was dried and weighed. The fibers were delignified using 10% NaOH solution with a ratio of 1:20 on a fiber basis at room temperature (Method 1) and 70 – 80°C (Method 2). Then, the cellulose was washed with distilled water and then refluxed for 30 minutes with distilled water for washing and then dried. While in method 3, the delignification process was carried out first and followed by the bleaching process under the same condition as method 1.

2.4. Parameter analysis

The obtained cellulose was measured by its yield after the delignification and bleaching process. The obtained cellulose was analyzed including moisture content, water extractive materials, hemicellulose, cellulose, and lignin content, FTIR test using *Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy* (ATR- FTIR) Thermo Scientific S10, and CIE L*, a*, and b* color test.

2.5. Test method for fiber and cellulose

Analysis of fiber components was carried out using the Chesson method [6]. 1 gram powder sample was added with 150 mL of distilled water and refluxed for 2 hours. The sample was filtered and washed until the pH was neutral and then dried at 105°C, weighed, and calculated using equation (1). The residue 1 obtained was added with 150 mL of 0.5 M H_2SO_4 and refluxed for 2 hours. The sample was then filtered and washed until the pH was neutral, dried at 105°C until dry then weighed and calculated using equation (2). The residue 2 obtained was added with 10 mL 72% H_2SO_4 and macerated for 4 hours at room temperature. The sample was added with 150 mL of 0.5 M H_2SO_4 and refluxed for 2 hours, filtered, and washed until the pH was neutral. The sample was dried at 105°C until dry then weighed and calculated using equation (3). Calculation of lignin content using equation (4) is as follows:

$$\text{water soluble extractive value (\%)} = \frac{\text{initial mass-mass of residue 1}}{\text{initial mass}} \times 100\% \quad (1)$$

$$\text{hemicellulose content (\%)} = \frac{\text{mass of residue 1-mass of residue 2}}{\text{initial mass}} \times 100\% \quad (2)$$

$$\text{cellulose content (\%)} = \frac{\text{mass of residue 2-mass of residue 3}}{\text{initial mass}} \times 100\% \quad (3)$$

$$\text{lignin content (\%)} = \frac{\text{mass of residue 4}}{\text{initial mass}} \times 100\% \quad (4)$$

3. Results and discussion

3.1. Cellulose yield

As the most sources of cellulose, currently OPEFB has not been explored yet for its use. Several studies examined the use of cellulose from OPEFB for fiber, microcrystalline cellulose, and carboxyl methyl cellulose. It should be noted that the cellulose isolation process is diverse, but the chemical methods are quite effective in separating cellulose from its impurities. The combination of the bleaching process, acid hydrolysis, and alkaline delignification can alternatively increase the purity of cellulose.

Figure 1 showed the conversion of oil palm empty fruit bunches to cellulose. The size of the OPEFB was reduced before processing to expand the surface area in order to facilitate the bleaching process and acid hydrolysis to alkaline delignification. The study demonstrated the effectiveness of the cellulose extraction process by comparing the bleaching process at an early stage with the reverse delignification process method. The bleaching-acid hydrolysis was carried out at the beginning of the process with the assumption that the acid hydrolysis process would make it easier to open the cell wall and break down the hemicellulose and lignin noticeably more. Meanwhile, in the cellulose extraction process with the delignification step at the beginning of the process, assume that the alkaline delignification aimed to remove the lignin, therefore the bleaching process would be lighter. The bleaching and delignification process is a pre-treatment process which in turn will affect reducing lignin, hemicellulose extracted components, increasing surface area, porosity, and pore size, reducing cellulose crystallinity, and increasing enzyme accessibility.

Different concentrations of sodium chlorite and bleaching cycles produced different bleaching and delignification yields in the range of 42 – 54% and 15-22% (table 1). The smallest yield was obtained from the third method which the delignification step was early followed by bleaching. The yield is 42.98% after bleaching, and 15.68% after the delignification process, while the moisture content did not differ significantly in the cellulose produced.

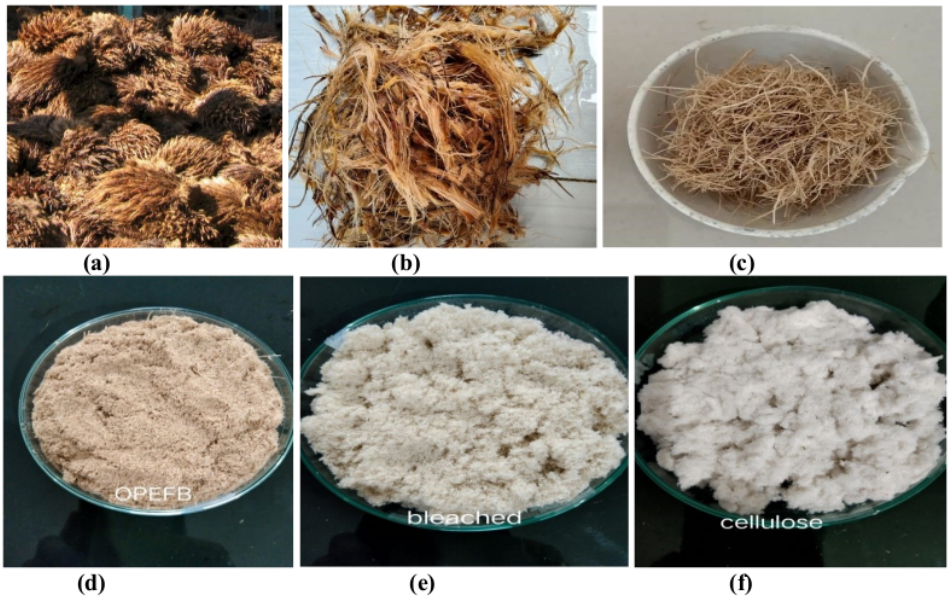


Figure 1. a) OPEFB; b) OPEFB fiber; c) Cuts of OPEFB fiber; d) OPEFB powder; e) after bleaching; f) cellulose from OPEFB.

Table 1. Yield and moisture content of cellulose extracted by various methods from OPEFB.

Method	Bleaching yield (%)	Delignification yield (%)	Cellulose moisture content (%)
Bleaching Na Chlorite 3% 2x, Delignification NaOH 10%, Room Temp.	44.82	18.69	6.60
Bleaching Na Chlorite 3% 2x, Delignification NaOH 10% Temp. of 70 - 80°C	49.64	16.58	7.47
Delignification NaOH 10% Temp. of 70-80° C,Bleaching Na Chlorite 3% 2x	42.98	15.86	6.34
Bleaching Na Chlorite 1.7% 3x, Delignification NaOH 17.5% Room Temp.	54.5	22.25	-
Bleaching Na chlorite 1.7% 4x, Delignification NaOH 17.5%, Room Temp.	49.3	21.4	-

- : not measured

The obtained cellulose from several extraction methods using some variations in sodium chlorite concentration and the cycle (table 2) illustrated the highest cellulose was obtained in the bleaching method with sodium chlorite of 3% for two cycles followed by delignification using 10% NaOH at room temperature. The delignification process at a higher temperature (70-80°C) did not affect enough to get high cellulose. Likewise, the method that applied NaOH delignification first at 70-80°C only produced 68.41% cellulose. It appeared that the application of high temperature in the delignification process did not significantly affect the purity of cellulose.

The bleaching processes using sodium chlorite of 3% by twice cycles and alkaline delignification at room temperature reduced lignin by approximately 87%, while the delignification at 70-80°C decreased lignin up to 94.81%; however, the hemicellulose was relatively higher. According to Kim [7] that the main reaction during alkaline pre-treatment includes dissolution of lignin and hemicellulose and de-esterification (saponification) of intermolecular ester bonds. This reaction changed the degree of polymerization of each component and the physical properties of the solid. These changes can include surface area, porosity, and crystallinity. The change in crystallinity index was caused by the removal of amorphous regions (lignin and hemicellulose) from the biomass, rather than structural changes in the cellulose fibers. The bleaching treatment of sodium chlorite under acidic conditions (pH 4-4.5) will facilitate delignification at room temperature because there has been a change in the structure of the fiber and partial dissolution of lignin and hemicellulose.

Referring to Seo et al. [8], alkaline reagent used in pretreatment are very selective for lignin separation. They can be used to recover most lignin with relatively high purity. Base pretreatment retained more hemicellulose than acid or neutral pretreatment. Pretreatment using dilute acid or hot water is effective for hemicellulose recovery. With proper selection of the pre-treatment and its sequence of operations, effective fractionation can be achieved. If alkaline treatment is applied in the first step, low lignin biomass and lignin are obtained as products.

Table 2. Composition of cellulose extracted by various methods from OPEFB.

Method	Water Extractive Material	Hemicellulose	Cellulose	Lignin	% Decrease in Hemicellulose	% Decrease in Lignin	% Increase in Cellulose
OPEFB fiber	15.73	31.51	32.97	18.79	-	-	-
Bleaching Na Chlorite 3% 2x, Delignification NaOH 10% Room Temp.	7.45	9.86	80.32	2.37	68.71	87.39	143.62
Bleaching Na Chlorite 3% 2x, Delignification NaOH 10% Temp.of 70-80 °C	6.53	24.01	68.41	1.05	23.80	94.41	107.49
Delignification NaOH 10% Temp of 70-80° C, Bleaching Na Chlorite 3% 2x	21.22	11.47	65.26	2.06	63.59	89.04	97.94
Bleaching Na Chlorite 1.7% 3x, Delignification NaOH 17.5% Room Temp.	10.18	16.83	64.84	8.15	46.59	56.63	96.66
Bleaching Na Chlorite 1.7% 4x, Delignification NaOH 17.5% Room Temp	10.56	11.16	70.96	7.32	64.58	62.00	115.22

In using small sodium chlorite, namely at 1.7% concentration, a more intensive bleaching cycle is required to obtain high purity cellulose. Bleaching at 1.7% sodium chlorite for four cycles and followed by the delignification process at a higher concentration of 17.5% was only able to obtain 70.96% cellulose. According to Yimlamai et al. [9], the cellulose extraction process with 17.5% NaOH delignification and two cycles of delignification using sodium chlorite resulted in 83.7% purity of cellulose, hemicellulose 5.6%, and lignin 1.7%. This phenomena indicated that a process is needed optimization to obtain high purity cellulose but with efficient sodium chlorite concentration and bleaching cycle. The bleaching process with acid combination will also allow continued

depolymerization, on the other hand, if it is too short then the impurities are still high, especially the lignin content is still relatively high.

Hemicellulose with a less regular structure is more amorphous than cellulose, thus consequently is easily hydrolyzed by acids or enzymes into monomers. With acid pretreatment, hemicellulose can be easily degraded to decomposition products including furfural. Hemicellulose has a highly branched structure, responsible for a high soluble in water [7].

The components of cellulose, hemicellulose, and lignin biomass are degraded at different temperatures. In contrast to cellulose and hemicellulose, lignin contains many phenyl rings which are relatively strong in structure, although they begin to degrade at about the same temperature as cellulose and hemicellulose (<200°C), lignin is not completely degraded until temperatures over 700°C [9].

3.2. Cellulose color

Using sodium chlorite as a bleacher affects the removal of color components in OPEFB fiber. OPEFB was originally yellowish-brown in color and the bleaching process will result in a color fade to white. Characteristics of CIE L, a*, and b* (Table 3) can give an idea of the white intensity of the cellulose produced. The cellulose produced tends to be a bright gray color, in the bleaching process of 3% sodium chlorite 2 cycles resulted the whiteness level (CIE L*) is 81.3 on average. The complete color description is presented in figure 2.

The cellulose resulted from the extraction process by bleaching with 3% sodium chlorite 2 cycles before delignification had a higher brightness level than other methods. Meanwhile, CIE a* is greener and CIE b* points to blue. At the CIE a* value, the positive value tends to be red and the negative value tends to be green. While at the CIE b* value, the positive value tends to be yellow and the negative value tends to be blue.

Table 3. CIE L*, a*, b*, Chroma and HUE of cellulose extracted by various methods from OPEFB.

Method	CIE L*	CIE a*	CIE b*	Chroma	HUE
OPEFB Fiber	61.25	8.40	23.38	24.88	29.9
Bleaching Na Chlorite 3% 2x, Delignification NaOH 10% Room Temp.	81.3	-0.5	-0.9	1.9	180.8
Bleaching Na Chlorite 3% 2x, Delignification NaOH 10% Temp. of 70 °C	79.1	-0.8	1.02	1.43	108.2
Delignification NaOH 10% Temp. of 80° C, Bleaching Na Chlorite 3% 2x	77.28	-0.23	2.64	2.77	65.3

If the chroma value increases, the color will be lighter. While if the chroma value decreases, the color fades. Chroma is a color level based on sharpness which serves to define the color of a product that tends to be shiny or tends to be dull. Chroma follows a percentage that ranges from 0 to 100. The higher the chroma value, the duller the product is, while the lower the chroma value, the shinier it is.

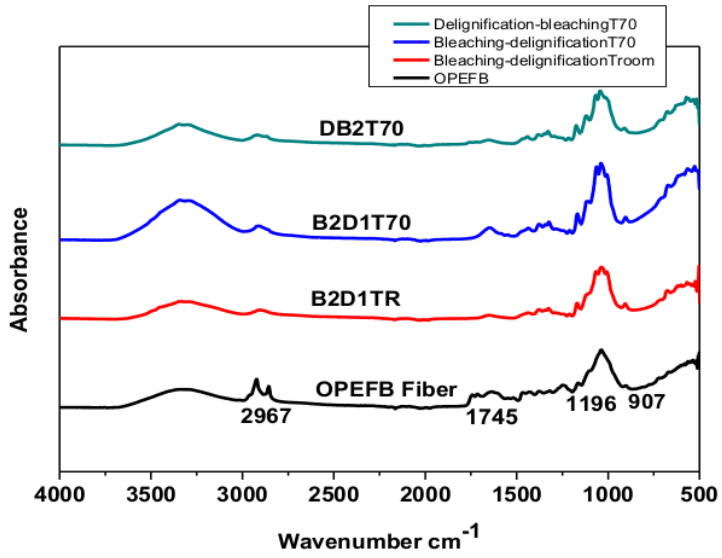
Figure 2. Color appearance a) OPEFB fiber; b) Cellulose of bleaching 2x-delignification; c) Cellulose of bleaching 2x-delignification at temp of 70-80°C; d) Cellulose of delignification-bleaching 2x.

Cellulose with a bleaching process of two cycles is still whitish yellow, this is due to the residual lignin content in the remaining structure. Balaji and Nagarajan [10] stated the alkaline delignification process allows the dispersion of NaOH in the amorphous region which causes intermolecular disturbances thereby promoting the removal of non-cellulose parts, especially lignin, from OPEFB fibers. With the washing process, a whiter color and a smoother surface structure will be obtained on the cellulose powder.

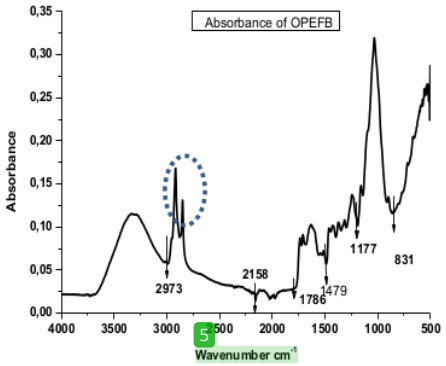
In the chemical bleaching process, the oxidation reaction will form a carboxyl group of lignin which in alkaline conditions will be ionized which in turn will increase the solubility. The principle of this reaction is not sufficient to completely remove lignin from the fiber and in the fragmentation reaction, depolymerization is required to produce small molecules that can escape from the structure of the swollen fiber network. The initial removal of lignin will take place quickly and then slow down so that one oxidative step is not sufficient to completely remove lignin [11].

3.3. Fourier transform infrared spectroscopy analysis

The peak at wave number 907 cm^{-1} indicated the deformation of C1-H which are cellulose and hemicellulose, as well as at 1196 cm^{-1} a stretching C-O-C which indicates cellulose. Hemicellulose was identified at the peak of wave number 1745 cm^{-1} where stretching of C=O occurred unconjugated to ketones, carbonyls, and aliphatic groups. Similarly, at the peak, extractive material was detected, namely the presence of stretching C=O in the carbonyl ester. At the peak of wave number 2967 cm^{-1} indicates the presence of lignin, cellulose, and hemicellulose through symmetrical methyl and methylene stretching.



(a)



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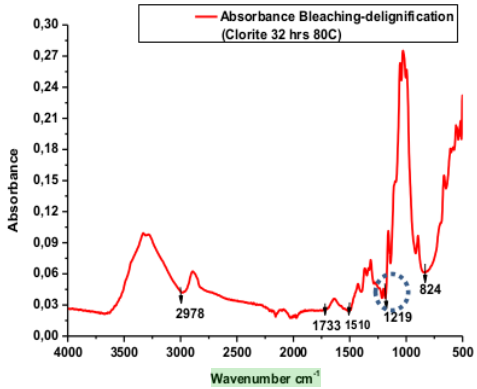

(c)

Figure 3. FTIR analysis a) on cellulose from several methods; b) OPEFB fiber c) cellulose with bleaching 2 times and delignification.

Figure 3 indicated a change in the wave number area of 1479 – 1768 cm^{-1} in OPEFB fiber where the cellulose from the bleaching-delignification method was sloping as well as at the peak of 2857 cm^{-1} sloping. In the wave number area of 1400 cm^{-1} was an area of lignin reduction (C=O stretching), while at a wavelength of 1700 cm^{-1} was an area of hemicellulose reduction (C=O stretching occurred unconjugated to ketones, carbonyls, and aliphatic groups) and extractive materials, namely the presence of stretching C=O in the carbonyl ester. Likewise, the slope at wave number 2857 cm^{-1} indicated a

decrease in hemicellulose and lignin (asymmetric stretching of methyl and methylene). A sharp peak in Figure 3c pointed at 1219 cm^{-1}, correlated with an increase of the purity of cellulose after the bleaching and delignification process. The research of Balaji and Narajanan [10] showed that after alkaline delignification treatment, the peak intensity at wave numbers 3340 cm^{-1}, 1750 cm^{-1}, 1250 cm^{-1}, 1663 cm^{-1}, and 1600 cm^{-1} was significantly reduced. This illustrated a lower percentage of hemicellulose content and hydroxyl groups.

4. Conclusions

The bleaching process in the early stage using 3 % sodium chlorite in 2 cycles and followed by delignification using 10% NaOH can produce more than 80% cellulose purity. The bleaching process using sodium chlorite can support delignification by reducing hemicellulose and lignin Indicated the identification of functional groups using FTIR showing a sharp peak at a wave number of 1219 cm^{-1}.

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References

[1] Direktorat Jenderal Perkebunan 2021 Statistik Perkebunan Unggulan Nasional 2019-2021 *Angew. Chemie Int. Ed. 6(11), 951–952.*
[2] Dewanti D P 2018 Cellulose potential of empty fruit bunches waste as the raw material of bioplastics *J. Teknol. Lingkung.* **19** 81
[3] Chang S H 2014 An overview of empty fruit bunch from oil palm as feedstock for bio-oil production *Biomass and Bioenergy* **62** 174–81
[4] Chen D, Lawton D, Thompson M R and Liu Q 2012 Biocomposites reinforced with cellulose nanocrystals derived from potato peel waste *Carbohydr. Polym.* **90** 709–16
[5] Kargarzadeh H, Ahmad I, Abdullah I, Dufresne A, Zainudin S Y and Sheltami R M 2012 Effects of hydrolysis conditions on the morphology, crystallinity, and thermal stability of cellulose nanocrystals extracted from kenaf bast fibers *Cellulose* **19** 855–66
[6] Datta R 1981 Acidogenic fermentation of lignocellulose–acid yield and conversion of components *Biotechnol. Bioeng.* **23** 2167–70
[7] Kim T H 2013 Pretreatment of Lignocellulosic Biomass *Bioprocess. Technol. Biorefinery Sustain. Prod. Fuels, Chem. Polym.* 91–110
[8] Seo M, Seo M, Choi S E, Shin K, Lee J B, Yang D Y and Kim J W 2020 Cellulose nanofiber-multilayered fruit peel-mimetic gelatin hydrogel microcapsules for micropackaging of bioactive ingredients *Carbohydr. Polym.* **229** 115559
[9] Yimlamai B, Choorit W, Chisti Y and Prasertsan P 2021 Cellulose from oil palm empty fruit bunch fiber and its conversion to carboxymethylcellulose *J. Chem. Technol. Biotechnol.* **96** 1656–66
[10] Balaji A N and Nagarajan K J 2017 Characterization of alkali treated and untreated new cellulosic fiber from Saharan aloe vera cactus leaves *Carbohydr. Polym.* **174** 200–8
[11] Castellan A and Grelier S 2016 Color and Color Reversion of Cellulosic and Lignocellulosic Fibers *Lignocellul. Fibers Wood Handb. Renew. Mater. Today's Environ.* 531–51

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