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THE EFFECTS OF $NaClO_2$ AND H_2O_2 AS BLEACHING AGENTS IN THE SYNTHESIS OF CELLULOSE ACETATE FROM OIL PALM EMPTY FRUIT BUNCH

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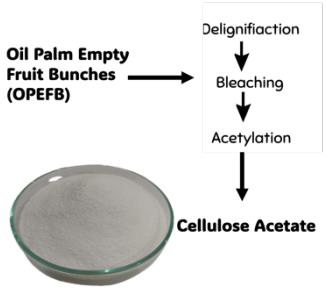
Graphical abstract



Abstract

Indonesia is one of the largest palm oil-producing countries in the world. The results of palm oil processing produce a lot of waste, such as oil palm empty fruit bunches (OPEFB). The utilization of OPEFB in Indonesia is minimal, so it needs further development. OPEFB has a very high α-cellulose content, so that it can be used as raw material for manufacturing cellulose acetate. Making cellulose acetate from OPEFB consists of three main steps: delignification, bleaching, and acetylation. This study aims to compare the effects of the concentration of $NaClO_2$ or H_2O_2 in the bleaching process on the physical properties of cellulose acetate from OPEFB. In the delignification process, 100 mesh-sized OPEFB powder is reacted with 5% sodium hydroxide (NaOH) at 100°C for 2 hours. The residue from deliglinfication was washed using distilled water until pH 7, filtered using filter paper, and dried using an oven at 50°C. The next step is the bleaching process using $NaClO_2$ or H_2O_2 with concentration variations (1%, 2%, 3%, 4% and 5%). The bleaching process was carried out at 90°C for 1.5 hours. After filtration, the bleached residue was washed using distilled water until pH 7, filtered using filter paper, and dried using an oven at 50°C. After the bleaching process, α-cellulose powder was produced. The next step is acetylation, containing three main steps. The first step is the reaction between α-cellulose powder with acetic acid (CH_3COOH) and sulfuric acid (H_2SO_4) at 40°C for 1.5 hours to perform activation. The second step is the addition of anhydrous cellulose in a ratio of 1:10 and mixed at 40°C for 1.5 hours. The third step is the addition of distilled water, acetic acid, and sodium acetate (CH_3COONa) at 40°C for 5 minutes. Then, followed by filtration, the residue was washed using distilled water and methanol until pH 7 and dried at 50°C. The best result is using $NaClO_2$ as a bleaching agent with a concentration of 2%, resulting in cellulose acetate yielding 98.85%, density of 1.954 g/mL, and L value of 91.897 for colorimetric test results. The density and L-value were close to commercial cellulose acetate (Sigma Aldrich). From the results of FTIR analysis, it can be concluded that the acetylation of α-cellulose into cellulose acetate has been successful, as evidenced by the formation of carbonyl groups (C=O).

Keywords: Cellulose Acetate, Oil Palm Empty Fruit Bunches (OPEFB), Bleaching, H_2O_2, $NaClO_2$

1.0 INTRODUCTION

Indonesia is known as an agricultural country because it has diverse and abundant plantation products. Indonesia is also known as the largest palm oil-producing country in the world. In 2018, the area of oil palm plantations in Indonesia reached 14.3 million and produced 40.5 million tons of oil palm [1]. Palm oil processing results in a lot of waste that needs to be utilized, such as OPEFB. OPEFB waste is 25% (12.42 million tonnes) and has only been used 10%. So, the potential for OPEFB waste is abundant at 15% or 11.18 million tonnes [2]. OPEFB comprises 24.65% cellulose, 21.34% hemicellulose, and 14.31% lignin[3]. Previous research reported that OPEFB contained cellulose in amounts of 41.8% [4],36.2% [5], and 32.80% [6]. The high cellulose content in OPEFB can be utilized as a raw material for producing cellulose acetate [7].

Cellulose acetate is cellulose in which an acetyl group replaces the hydroxyl group. Cellulose acetate is white solid, non-toxic, tasteless, and odourless. Cellulose acetate is an artificial chemical compound which is a compound derived from

cellulose. Chemically, cellulose acetate is an ester of acetic acid and cellulose [8]. Cellulose acetate is an organic compound of great commercial importance due to its wide application in producing textiles, filters, photographic films, plastic materials, pharmaceuticals, and packaging [9].

There are three main steps to make cellulose acetate from OPEFB. The first step is delignification. This step aims to remove the lignin content in OPEFB so that the purity of cellulose in OPEFB will increase. The second step is the bleaching reaction; this stage seeks to remove the remaining impurities, such as lignin and hemicellulose[9]. The product of the delignification and bleaching stages is a powder with a high content of α-cellulose. The last step is the acetylation reaction. At this step, the activated α-cellulose is reacted with acetic anhydride to produce the final product, cellulose acetate[10].

The novelty of this research is the effect of a concentration of bleaching agent ($NaClO_2$ or H_2O_2) on the bleaching process in the synthesis of cellulose acetate from OPEFB. The effect of bleaching agents on the yield and properties of cellulose acetate, such as density, color, and crystal structure has been studied and then compared with commercial cellulose acetate (Sigma Aldrich).

2.0 MATERIALS AND METHODOLOGY

2.1 Materials

The materials used in this study are 100 mesh-sized OPEFB powder, Sodium Hydroxide (NaOH) from Sigma Aldrich, Hydrogen Peroxide (H_2O_2) 30% (v/v) from Sigma Aldrich, Sodium Chlorite ($NaClO_2$) from Sigma Aldrich, Glacial Acetic Acid (CH_3COOH) from Sigma Aldrich, Sulfuric Acid (H_2SO_4) from Smart-Lab, Acetic Anhydride from Merck, Sodium Acetate (CH_3COONa) from Sigma Aldrich Methanol (CH_3OH) from Sigma Aldrich, and Aquadest.

2.2 Methodology

The preparation of cellulose acetate from OPEFB consists of three steps: delignification, bleaching, and acetylation [11].

2.3 Delignification

Two hundred fifty liters of OPEFB was reacted with 1.5 liters of 5% (m/v) NaOH at 100°C while stirring for 2 hours. After 2 hours, the residue was washed with distilled water until pH seven and filtered using filter paper. The residue was then dried using an oven at 50°C, and the remaining mass was weighed.

2.4 Bleaching

After the delignification reaction stage, the delignification powder will go through a bleaching reaction stage. At this stage, the powder reacts using two bleaching agents, H_2O_2 or $NaClO_2$, to dissolve the remaining lignin after the delignification reaction. At this stage, the concentration variation of the bleaching agent (H_2O_2 or $NaClO_2$) used is 1-5 m/v% with an interval of 1 m/v%.

2.4.1 Bleaching using H_2O_2

Mixing α-cellulose powder and H_2O_2 with the ratio of α-cellulose powder: H_2O_2 is 1:10 (m/v) [12]. Add 0.1 N NaOH into the solution until the pH reaches 9 while stirring. The bleaching reaction lasted for 1.5 hours at 90°C with pH 9. Lignin that dissolves in H_2O_2 is then removed through the washing process using distilled water until the solution reaches pH 7. In this washing process, a separation is made between the filtrate and the residue formed so that the α-cellulose residue and lignin dissolved in H_2O_2 will separate. The α-cellulose residue is then dried in an oven at 50°C and weighed until the mass is constant. The dried α-cellulose was then crushed using a grinder and filtered to a size of 100 mesh.

2.4.2 Bleaching using $NaClO_2$

Mixing α-cellulose powder and $NaClO_2$ with the ratio of α-cellulose powder: $NaClO_2$ is 1:12 (m/v) [1]. Add 0.5 ml CH_3COOH p.a. and heat at 80°C for 1.5 hours with a stirrer. The bleached residue was washed using distilled water until the solution reached pH 7. In this washing process, a separation was made between the filtrate and the precipitate formed so that the α-cellulose and lignin residues dissolved in $NaClO_2$ would separate. The α-cellulose residue is then dried in an oven at 50°C and weighed until the mass is constant. The dried α-cellulose was then crushed using a grinder and sieved to a size of 100 mesh.

2.5 Acetylation

Acetylation is the formation of cellulose acetate by reacting α-cellulose and acetic anhydride with a sulfuric acid catalyst, which aims to replace hydroxyl groups with acetyl groups. Sulfuric acid reacts with acetic anhydride to form acetyl sulfate, then reacts with α-cellulose to form cellulose acetate. Reacted 5 grams of α-cellulose powder with 125 ml of Glacial Acetic Acid (CH_3COOH) and added ten drops of Sulfuric Acid (H_2SO_4) at 40°C while stirring for 1.5 hours. Reacted the solution with Acetic Anhydride at a ratio of α-cellulose: acetic anhydride of 1:10 and added ten drops of H_2SO_4 as a catalyst at 40°C. Adding 5 ml of distilled water, 12 ml of CH_3COOH, and 2 grams of CH_3COONa at 40°C while stirring for 5 minutes. Then, followed by filtration, the residue was washed using distilled water and methanol for 10 minutes. The precipitate formed was then dried using an oven at 50°C and weighed to a fixed weight.

2.6 Chemical And Physical Analysis

Yield and density are analyzed using gravimetry methods by Cao et al. [13] and Blake et al. [14]. Hemicellulose, cellulose, and lignin contents are analyzed by using the Chesson method [15]. The colorimetry test was done by using the colorimeter WR-10 QC Color Meter[16].

The Fourier Transform Infrared Spectroscopy (FTIR) using (Agilent Cary 630 FTIR, United States) has been used to observe the functional groups [17]. To identify the crystal structures, X-ray diffraction (X'Pert PRO PANalytical, Holland) has been used[18].

3.0 RESULTS AND DISCUSSION

3.1 Delignification

Before entering the bleaching process, OPEFB has been delignified using NaOH to separate cellulose from lignin and hemicellulose content. The delignified pulp will be analyzed using the Chesson method to determine the levels of lignin, cellulose, and hemicellulose remaining in the OPEFB. Table 1 shows the analytical results of OPEFB before and after the delignification reaction. The delignification response has successfully increased the cellulose content of OPEFB from 40% to 75.09%, decreased the lignin content from 23.74% to 8.07%, and decreased the hemicellulose content from 35.43% to 16.84%.

Table 1. Content Analysis of OPEFB Before and After Delignification (wt%)

Sample	Hemicellulose Content (wt%)	Cellulose Content (wt%)	Lignin Content (wt%)
OPEFB Before Delignification	35.43	40.83	23.74
OPEFB After Delignification	16.84	75.09	8.07

3.2 Bleaching

3.2.1 Xrd Analysis

XRD analysis is performed to characterize the crystal structure of a solid material using X-rays. XRD analysis is performed to describe the crystal structure of a solid material using X-rays. The X-ray diffraction method can explain the lattice parameters, type of structure, differences in atomic arrangement in the crystal, the presence of imperfections in the crystal, orientation, grain, and grain size of a material.

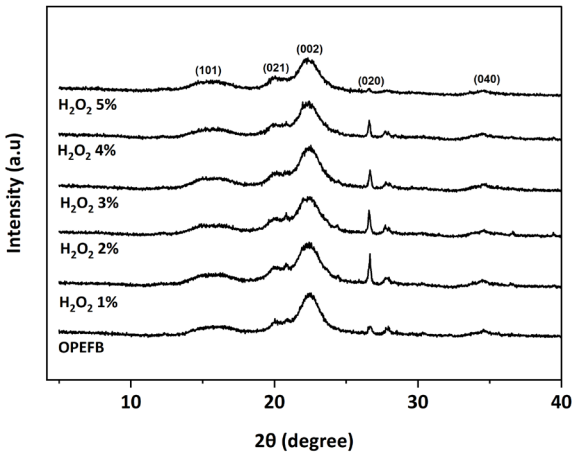


Figure 1 XRD results of OPEFB after bleaching process using variations concentrations of H2O2

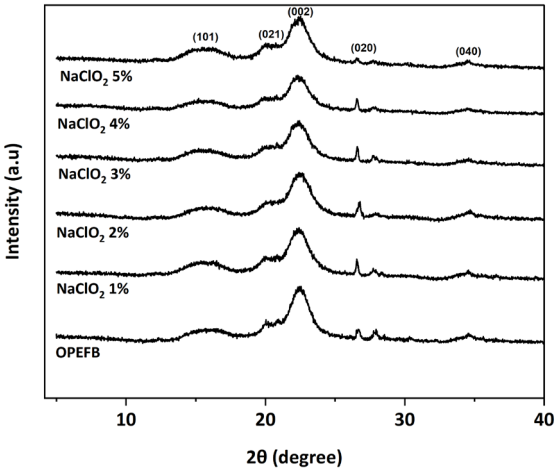


Figure 2 XRD results of OPEFB after bleaching process using variations concentrations of $NaClO_2$

The delignified OPEFB is then bleached using H_2O_2 or $NaClO_2$. The bleaching process serves to make the OPEFB color brighter. The XRD tests were then carried out for samples after the bleaching process to analyze the crystal structure of cellulose. Figures 1 and 2 show the XRD results of OPEFB after the bleaching process using H_2O_2 and $NaClO_2$, respectively. The main peaks of α-cellulose are at crystal planes of (110), (100), (002), and (040). The XRD results show the crystal structure of cellulose is alpha crystal. This result indicates the formation of α-cellulose in OPEFB after bleaching[19][20][21].

3.2.2 Ftir Analysis

FTIR analysis aims to qualitatively determine the functional groups in a chemical compound contained in OPEFB raw materials and OPEFB resulting from bleaching.

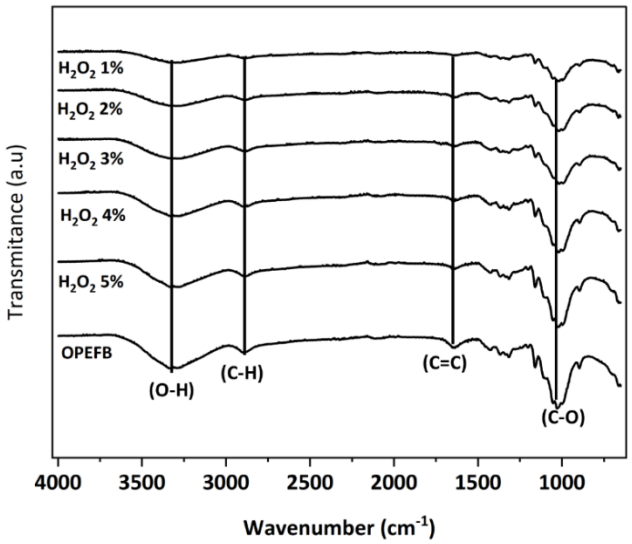


Figure 3 FTIR results of OPEFB after bleaching process using variations concentrations of H_2O_2

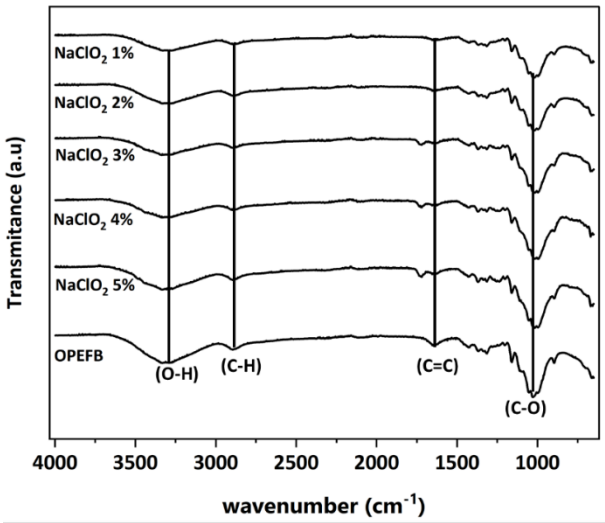


Figure 4 FTIR results of OPEFB after the bleaching process using variations of $NaClO_2$

OPEFB after the bleaching process using $NaClO_2$ or H_2O_2 were analyzed using Fourier Transform Infrared Ray Spectroscopy (FTIR), as shown in Figure 3 and Figure 4, which aims to analyze functional group changes before and after the bleaching reaction. Based on Figure 3 and Figure 4, FTIR results show that in OPEFB, there is a C = C double bond, characteristic of lignin. After going through the bleaching process, the greater the concentration of H_2O_2 or $NaClO_2$, the lower the transmittance value, indicating the presence of C = C bonds. It can be concluded that the lignin content is lost after the delignification and bleaching process. And some peaks show O-H and C-H bonds. The two bonds refer to the CH_2-OH bond, which is characteristic of hemicellulose and cellulose. In Figure 3 and Figure 4, it can be seen that the higher the concentration of H_2O_2 or $NaClO_2$, the lower the transmittance value, which indicates the presence of O-H and C-H bonds, which refer to the cellulose content in the sample [22][23].

3.3 Acetylation

3.3.1 Yields

Yield is the ratio between the mass of cellulose acetate resulting from acetylation (gr) and the initial mass of α-cellulose entering the acetylation process (gr)[24].

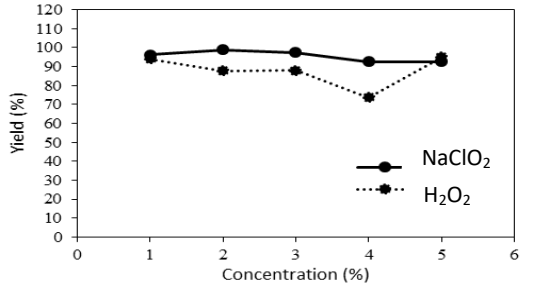


Figure 5 Comparison Between Yield and Concentration of H_2O_2 and $NaClO_2$

Based on Figure 5, the concentration of H_2O_2 or $NaClO_2$ affects the yield value of cellulose acetate produced from the acetylation reaction. The best yield value was obtained in the bleaching reaction using NaClO2 with a concentration of 2% and a yield of 98.85%.

3.4 Ftir Analysis

FTIR analysis aims to qualitatively determine the functional groups in a chemical compound contained in cellulose acetate from OPEFB and commercial cellulose acetate (Sigma Aldrich).

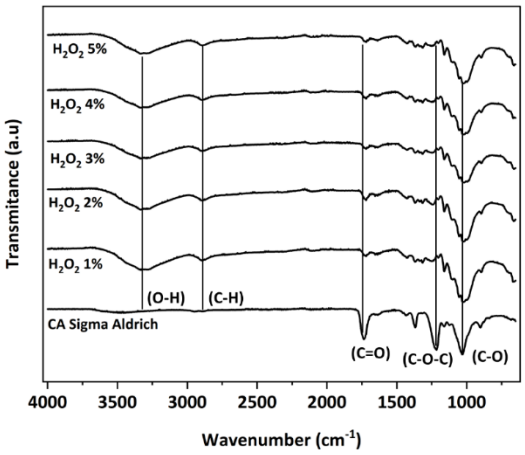


Figure 6 FTIR results of OPEFB after acetylation process using H_2O_2 with variations concentrations of 1-5 m/v with interval 1 m/v %

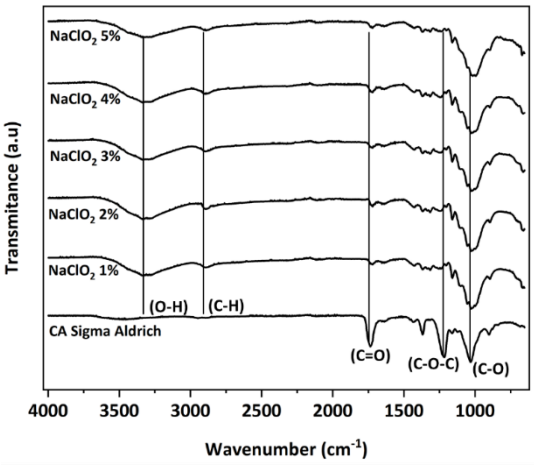


Figure 7 FTIR results of OPEFB after acetylation process using variations concentrations of $NaClO_2$

After the acetylation process using $NaClO_2$ or H_2O_2, OPEFB was analyzed using FTIR, as shown in Figure 6 and Figure 7, which aims to analyze functional group changes before and after the acetylation reaction. Cellulose acetate comprises three functional groups: C-O, O-H, and C-H. FTIR analysis results differ (C=O), and peaks show acetyl groups (C-O). The carbonyl (C=O) functional group is the group that distinguishes cellulose from cellulose acetate. It shows that an acetyl group has substituted the O-H functional group on cellulose to produce a C=O carbonyl functional group. A carbonyl group is a constituent group that characterizes cellulose acetate. Thus, the FTIR results can prove

that the acetylation process of OPEFB into cellulose acetate has been successful [22],[25],[26].

3.5 Density

The density test aims to determine the density of cellulose acetate from OPEFB, which will then be compared with the density of commercial cellulose acetate from Sigma Aldrich.

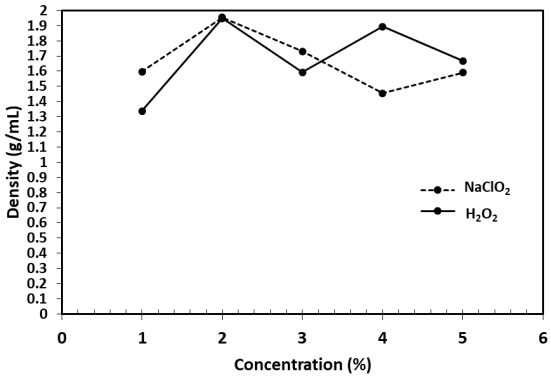


Figure 8 Density results of cellulose acetate using H_2O_2 or $NaClO_2$

Based on Figure 8 density test results, it can be seen that of all variables, $NaClO_2$ or H_2O_2 has a better density value when compared to the density of commercial cellulose acetate (Sigma Aldrich), which is 1.3 g/mL. It can be seen that the best density value uses a $NaClO_2$ bleaching agent with a concentration of 2%, which is 1.954 g/mL.

3.6 Colorimetry

The colorimetric test measured the color of the cellulose acetate product produced from OPEFB. Based on Table 2, the color of cellulose acetate products using $NaClO_2$ has a higher L value than those using H_2O_2. The L value of cellulose acetate from OPEFB is close to the L value of commercial cellulose acetate using $NaClO_2$ with a concentration of 4% and an L value of 93.327.

Table 2 Colorimetric Test Result

Variable	Concentration	L	a	b
Cellulose Acetate Commercial*		100	**27.44**	**-11.9**
Cellulose Acetate using $NaClO_2$	1%	90.703	1.667	8.947
	2%	91.897	1.827	9.427
	3%	92.573	2.613	10.680
	4%	93.327	2.927	10.143
	5%	93.157	2.773	10.497
Cellulose Acetate using H_2O_2	1%	91.833	3.367	12.870
	2%	90.363	2.603	11.077
	3%	83.103	4.257	13.260
	4%	89.010	4.113	12.737
	5%	88.970	3.720	11.393

*Cellulose Acetate Commercial (Sigma Aldrich)

4.0 CONCLUSION

Based on the results of the study, it can be concluded that the best cellulose acetate produced from OPEFB using NaClO2 bleaching agent with a concentration of 2%, by creating cellulose acetate yield of 98.85%, density of 1.954 g/mL, and a L value of 91.897 for colorimetric test results. The density and L-value were close to commercial cellulose acetate (Sigma Aldrich). The acetylation reaction of α-cellulose to cellulose acetate has been successfully proven by carbonyl group (C=O) formation in the cellulose acetate product.

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