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Preparation & Characterization of Microcrystalline Cellulose from Agriculture Waste

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Abstract. In this work, microcrystalline cellulose was prepared from oil palm trunk by water treated fibre process and alkali bleaching. The prepared samples were characterized by using Fourier-transform infrared spectroscopy attenuated total reflectance (FTIR-ATR), Scanning Electron Microscopy (SEM) and X-ray diffraction (XRD). FTIR-ATR spectra analysis indicates the presence of the hydroxyl group, alcohol, alkane/alkene and imine group. XRD patterns revealed the amorphous nature of the samples and the crystallinity index for extracted cellulose is 48.7 %. SEM images showed the fibrous structure of the microcrystalline cellulose with a size of 50 µm. This research proved that the synthesized microcrystalline cellulose could be potentially used as reinforcement in bio composite for better performance and ductility.

1. Introduction

In recent years, there has been growing interest in the production of bioplastic from different precursors such as agricultural by-products. In fact, cheap materials with a high cellulose and starch content can be used as a raw material for the production of bioplastic. Agricultural by-product has proven to be

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promising raw material for the production of bioplastic because of their availability at a low price and biocompatible [1].

Cellulose is an organic compound, and it was an essential structural component of the primary cell wall of plants and woods. Cellulose is one of the abundant organic polymers that exist on Earth. Cellulose is a long chain of linked sugar molecule that gives wood its remarkable strength. It is the main component of cell walls. ($C_6H_{10}O_5$) is the chemical formula of the cellulose which polysaccharide that consist of linear chain of several hundred to many thousands of β (1→4) linked D-glucose units.

Oil palm which originates from Africa is widely planted in South-East Asia, especially in Malaysia, Indonesia and Thailand. Without a doubt, it has become the most important agricultural crop in Malaysia and has been the key to the national economic expansion [10]. Oil palm is harvested at age 25-30 years for replanting. Most oil palm trunks are left in the field to rot to be an organic fertilizer or will be burnt down. Thus, this research was intended to implement the waste to wealth concept where the microcrystalline cellulose from oil palm trunk was prepared and characterized.

2. Materials and Methods

2.1. Raw material

Oil palm trunk (OPT) was obtained from a plantation in Jengka, Pahang then was harvested and sawn into discs before the bark was removed. Polyvinyl alcohol (PVA), Mw = 7200, Sodium Chlorite, Mw = 90.44 was purchased from R&M Chemicals. Sulphuric acid, Acetic acid, Potassium Hydroxide, Glycerine, distilled water, deionized water.

2.2. Preparation of OPT Fibre

The oil palm trunk (OPT) was washed repeatedly with distilled water to remove the impurities that are dirt and dust. Then, the OPT was dried in the oven for 24 h that having a temperature at 110 °C to remove all the moisture content. Then the OPT was cut into small pieces and was blended by using a blender obtained at the Material Science Laboratory to get the small but not in a uniform size. Then, to obtain the uniform size, the OPT was sieved using 1.0 mm sieve to obtain granular size [1]. The extraction process was done by water treated fibre.

2.2.1. Preparation of Microcrystalline Cellulose by Water Treated Fibre Process. About 20 g of raw oil palm trunk fibres was weighed. Extractives of raw oil palm trunk fibres were removed by soaking in hot distilled water having a temperature at 60 °C for 6 h before they will be filtered on a Buchner Funnel. This process was called the hot water treated fibre process. This treatment was done because hot water can remove a substantial amount of extractives in oil palm trunk and was reduce the usage of toluene/ethanol in Soxhlet extraction method in producing the cellulose [2]. The extractives of OPT was correctly discarded.

2.2.2 Bleaching Process. Firstly, the extracted fibres were washed four times with interval 30 min for each time in sodium chlorite solution under acidic condition around 4 to 5 pH at 70 ˚C for 1 h. The sodium chlorite was a base, therefore to undergo successfully bleaching process to remove the hemicellulose, the solution must be in acidic condition [3]. Firstly, weighed 1.5 g of sodium chlorite and dissolved it in the 100 mL of distilled water until it dissolves. Then, 20 g of OPT fibre will be placed in the solution. The solution then was mixed with the acetic acid to get the acidic solution. The pH meter was being used to monitor the pH of the solution and was being maintained at 4-5 pH during all the period.

After 2 hours of the process, the bleached process was completed, and the solution was being cooled. Next, the solution was being filtered using the Buchner funnel set and also a vacuum pump. The OPT fibre was washed with distilled water and also acetone to remove the excessive sodium chlorite solution in the fibre. Moreover, the OPT fibre was changed its colour from yellow to white after the process was done. Last but not least, the OPT fibre was being dried in the oven for 24 h having 50 °C temperature to

ensure maximum drying. Then after being dried the OPT fibre was weighed on the weighing scale and was recorded in Table 1 below. The W_o indicated the weight of initial OPT fibre before process and W_i for the weight of OPT fibre after the process.

Table 1. The weight recorded of OPT fibre

Sample	W_o (g)	W_i (g)
OPT fibre	20	15.34

2.2.3 Lignin Removal by Kraft Process. Potassium hydroxide (KOH) was used to dissolve the lignin from the cellulose. The solution was prepared with 6 wt. % to remove the lignin from the extracted fibres by soaking it in KOH [4]. Firstly, 15 g of OPT fibre was weighed and placed in a conical flask that had 15 mL of 20 wt. % of KOH solution. Then, the solution was stirred for 20 min and at each interval 5 min the 10 mL of 20 wt.% KOH was added in the solution for four times. Moreover, after 20 min stirred, the conical flask was being closed and was let being stirred for another 30 min without using heat. Next, after 30 min stirred, 100 mL of distilled water was added in the solution and was being stirred for another 30 min.

After that, the solution was being filtered using the Buchner funnel and vacuum pump set. Then, the OPT fibre was washed with distilled water to remove the alkali residue on the fibre and was again undergo filtration with the Buchner funnel and vacuum pump set. Last but not least, 30 wt. % of sulphuric acid solution was prepared to soak the fibre. The soaking process was ongoing for 20 min. The OPT fibre was being filtered and dried in the oven for 24 h at 50 °C. Then, the weight after and before the process of the OPT fibre was being recorded in Table 2 below. The OPT fibre that we gain after this process was the extracted cellulose [5]. Moreover, the yield per cent of the extracted cellulose from the OPT fibre was also calculated to be 41.75 %.

Table 2. The weight recorded OPT fibre

Sample	w_o (g)	w_i (g)
OPT fibre	15	8.35

3. Results and Discussion

3.1 Fourier-Transform Infrared Spectroscopy (FTIR-ATR)

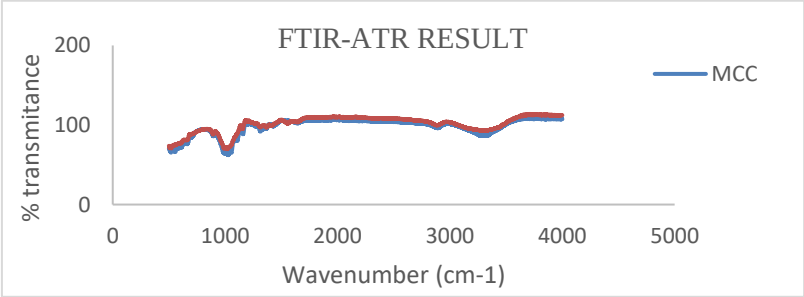

Figure 1. FTIR-ATR analysis for extracted cellulose and MCC

In order to gain more insights into the effects of hot water treated fibre process, studies on the chemical and structural characteristics of the extracted cellulose and microcrystalline cellulose (MCC) were essential.

The FTIR-ATR spectra of extracted cellulose from the oil palm trunk (OPT) and pure microcrystalline cellulose (MCC) as shown in Figure 1. In this experiment, we want to compare the functional group present between these two samples. In case of extracted cellulose, the peak at 3316.68 cm^{-1} and at 1557.34 cm^{-1}, was of O-H stretching vibration and C=O stretching vibration, respectively. Moreover, a strong peak at 1021.09 cm^{-1} and 992.55 cm^{-1} corresponds to C-N stretching vibrations C=C bending respectively. Next, it also shows a strong peaks C-H stretching at 2888.83 cm^{-1} that owe to the methylene group in cellulose [6].

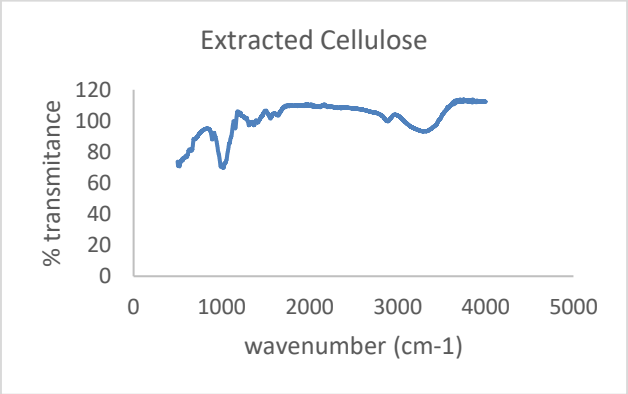

Figure 2. Extracted cellulose FTIR spectra

Cellulose, hemicelluloses, and lignin are the major constituents of the natural fibres. Cellulose is a linear polymeric compound has some important functional group within the cellulose units. In Figure 2 and Table 3, clearly shows bands between 2840 cm^{-1} and 3000 cm^{-1}, which indicates the aliphatic saturated C-H stretching associated with methylene groups in cellulose [7].

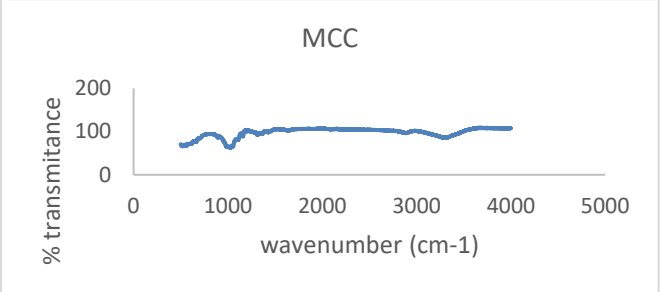

Figure 3. MCC FTIR spectral

The pure microcrystalline cellulose (MCC) that shown in Figure 3 showed peaks at 3330.45 cm^{-1} and 2890.18 cm^{-1}, was O-H stretching vibration and C-H stretching vibration respectively. The strong peaks of O-H vibration, indicating free hydroxyl groups owing to the strong intermolecular and intramolecular bonds [8]. Moreover, the extracted cellulose from the OPT had the match at 63.11% with the pure cellulose.

Based on Figure 3, it shows the result MCC that has been used for reference and also the comparison with extracted cellulose. From the spectral above, the result shows no high difference with the MCC. As can be seen, the strong absorption between 3400 cm^{-1} and 3600 cm^{-1} in all FTIR spectra is caused by the remaining OH groups of the fibre constituents. Besides, all the absorption bands that occurred in the extracted cellulose were present in the MCC spectra, indicating that both have similar compositions of the functional group.

Table 3. FTIR band for extracted cellulose and MCC

Sample	O-H stretching	C=O stretching	C-N stretching	C-H stretching
MCC	3330.45 cm^{-1}	1645.41 cm^{-1}	1027.24 cm^{-1}	2890.18 cm^{-1}
Extracted cellulose	3316.68 cm^{-1}	1557.34 cm^{-1}	1021.09 cm^{-1}	2888.83 cm^{-1}

From Table 3, the intensity of O-H bonds in the region 3500 cm^{-1} and 3100 cm^{-1} has a slight difference between the extracted cellulose and MCC. This implies that the hydrophilic nature of extracted cellulose has significantly reduced after the extraction process.

3.2 X-ray Diffraction (XRD)

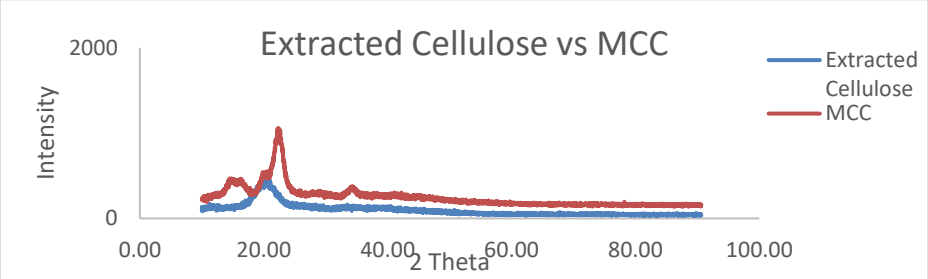

Figure 4. The different pattern results for extracted cellulose and MCC

The X-ray diffraction analysis is used to determine the percentage of crystallinity of the extracted cellulose that was isolated from OPT fibres and microcrystalline cellulose (MCC). Basically, cellulose compounds are composed of lignin, hemicellulose, and cellulose. Cellulose is known as crystalline in nature, while lignin is amorphous in nature. Figure 4 exhibits the XRD thermograms of all samples tested. The crystallinity index for extracted cellulose from OPT fibre and MCC are 48.7 % and 51.6 %, respectively.

These values had lower crystallinity characteristics as compared to those of cellulose isolated from oil palm empty fruit bunches having 59 % and 69 % carried out in two previous works, respectively [9]. In Table 4 above shows the value that we got for the crystalline and amorphous percentage for the extracted cellulose and MCC.

Table 4. Percentage of amorphous and crystallinity between extracted cellulose and MCC

Sample	Crystallinity (%)	Amorphous (%)
MCC	51.6	48.4
Extracted cellulose	48.7	51.3

After the hot water treated fibres were exposed to chemical treatment in order to produce the cellulose, it was found that the crystallinity of the samples was decreased as shown in Figure 5 below. This may be due to the low concentration of sulphuric acid attacking the amorphous region, thus does not penetrate the region that can cause hydrolytic cleavage of glycosidic bonds and releasing individual crystallites [10]. Efficient removal of non-cellulosic polysaccharides such as hemicelluloses and lignin matrix that attached to the celluloses fibres would also contribute to an increase in crystallinity.

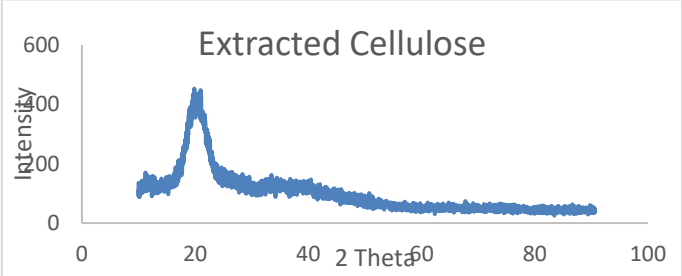

Figure 5. The XRD spectrum for extracted cellulose

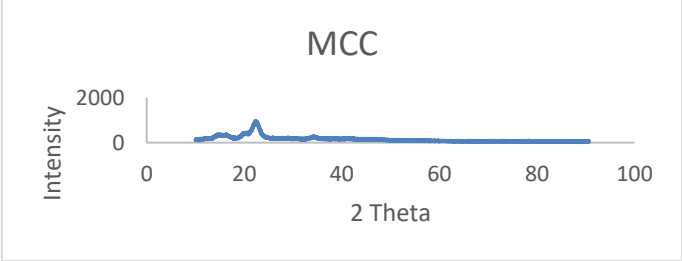

Figure 6. The XRD spectrum for MCC

These changes of diffraction peaks indicate the significant cleavage of hydrogen bonds in cellulose, leading to the significant recrystallization of lignocellulose after dissolution, chemical modification and regeneration. The peak of amorphous and crystalline are losing its shape because the composition of extracted cellulose had degraded. According to [11] under drastic conditions of acid hydrolysis, amorphous and crystalline regions may be only partially destroyed because of erosion caused by the high concentration of strong acid such as sulphuric acid. Then, the study also said that the concentration of sulphuric acid must be above 50 wt. % in order to undergo sufficient acid hydrolysed.

The crystalline structure of cellulose is very important to determine the elasticity, rigidity, thermal stability and absorption for the thin film sample. From the results, it can be said that the low concentration sulphuric acid does not fully and partially attack the amorphous region of the cellulose and then can increase the crystallinity index of the extracted cellulose. The sufficient acid hydrolysis also removed the non-celluloses polysaccharides such as hemicelluloses and lignin matrix, which was surrounded by the cellulose fibres.

4. Conclusion

In this research, microcrystalline cellulose was extracted from agriculture waste, namely oil palm trunk and was compared to the commercial microcrystalline cellulose. FTIR-ATR spectra revealed that the extracted cellulose from the OPT and commercial micro cellulose crystal have similar compositions of the functional group. XRD patterns revealed the amorphous nature of the samples and the crystallinity index for extracted cellulose is 48.7 %. Finally, SEM images showed the fibrous structure of the microcrystalline cellulose with a size of 50 µm.

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