Isolation and Characterization of Microcrystalline Cellulose from Coconut Fiber using Acid Hydrolysis Process

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Abstract: The object of this research was to explore the utilization of coconut fiber as a natural source for the production of microcrystalline cellulose. Coconut fiber was treated with alkali at the first time then bleached before the production of microcrystalline cellulose by acid hydrolysis (HCl). The produced materials were characterized using X-ray Diffraction (XRD) and some physicochemical properties such as pH, water soluble substance and loss on drying. XRD showed that acid hydrolysis process increase the crystallinity of microcrystalline cellulose from α-cellulose. Meanwhile the physicochemical properties showed that the produced material was close to British Pharmacopeia standard. The results proved that coconut fiber was a valuable source for the production of microcrystalline cellulose.

1 INTRODUCTION

Natural fibers consist amorphous and semicrystalline structure. Amorphous structure contain lignin and hemicellulose while semicrystalline contain cellulose. Cellulose fiber has better strength, stiffness and thermal stability than natural fiber because natural fiber still contains lignin and hemicellulose (Melbi et al, 2018).

Cellulose is a linear chain of glucose molecules with degree of polymerization between 10,000 to 15,000 and linked together through an oxygen covalently bounded. During biosynthesis, van der Waals and intermolecular hydrogen bonds between hydroxyl groups and oxygen of adjacent molecules. It makes cellulose relatively stable and gives high axial stiffness. In cellulose there are regions where the cellulose chains are arranged in a highly ordered structure. This region is called crystalline region. On the other side there are region that disordered. It is known as amorphous region. Crystalline phase can be extracted using acid catalyst hydrolysis. The result of this extraction is microcrystalline cellulose (Robert et al, 2011).

Microcrystalline cellulose is produced by depolymerization of cellulose materials with solutions of mineral acids at increased temperatures. The acid catalyst destroys glycoside bonds mainly in non crystalline domains. It makes cellulose lose its degree of polymerization from 10,000-15,000 become 120 to 250. Microcrystalline cellulose is used mainly as food products, cosmetic formulation, inactive ingredients of tablets and special additive for some technical applications (Michael and Alex, 2006). There are some natural resources that are used to produce microcrystalline cellulose. That natural resources are cassava bagasse (Panee et al,2015), corn husk (Roshni and Yamini, 2015), rice straw (Chin et al, 2016), empty fruit bunch palm oil (Nasution et al, 2017), cotton stalks (Hassan and El-Sakhawy, 2005). In this study, coconut fiber was used as raw material. The composition of coconut fiber can be seen at Table 1 (Khalil et al, 2006). Table 1 shows that the main constituent of coconut fiber is cellulose.

Table 1: Composition of coconut fiber

Composition	Percentage
Holocellulose	56.3
α – Cellulose	44.2
Lignin	20.5
Ash	2.2

There are some methods to produce microcrystalline cellulose which is ultrasonication

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(Zailani et al, 2016), ultrafiltration membrane (Nguyen and Rajesh, 2016), enzymatic hydrolysis (Herman et al, 2017) and acid hydrolysis (Ravindra et al, 2017). Acid hydrolysis was the conventional method for produce microcrystalline cellulose. Hydrochloric acid and sulfuric acid were common used as mineral acid for acid hydrolysis (Ravindra et al, 2017). With sulfuric acid hydrolysis, the sulphate group are problematic because of the thermal stability if compared to hydrochloric acid hydrolysis. So in this study, we choose hydrochloric acid to hydrolyzed the cellulose because microcrystalline produce with hydrochloric acid has better thermal stability (Birgi and John, 2009).

The main purpose of the present paper is to characterize microcrystalline cellulose that extracted from coconut fiber. The microcrystalline will be used as reinforcement agent for composites.

2 METHODS

2.1 Materials

Coconut fiber was collected from local market in Medan, Sumatera Utara, Indonesia. Other chemicals such as nitric acid, sodium nitric, hydrochloric acid, sodium hydroxide and sodium hypochloride were supplied by Merck.

2.2 Extraction of α-Cellulose from Coconut Fiber

Coconut fibers were cleaned from impurities and cut into small pieces. 50 gram of coconut fiber were added with 700 ml of 3,5% nictric acid that contained 8 mg of sodium nitric, then heated on temperature 90 °C using hotplate for 2 hours. Then, coconut fiber was washed with distilled water until its filtrate neutral. 2% of sodium hydroxide solution were add to the fiber and heated on temperature 50 °C for 1 hour. Clean the filtrate using distilled water till its filtrate neutral. Then the cellulose were added with 3,5% sodium hypochloride and then heated until boiled for 10 minutes then cleaned the filtrate using distilled water until its filtrate neutral.

Purified the cellulose using 340 ml sodium hydroxide solution 17,5% for 30 munites on temperature 80 °C. Cellulose was washed using distilled water until its filtrate neutral then bleached again for 30 minutes using sodium hypochlorite on temperature 60 °C then washed with water until filtrate neutral.

2.3 Isolation of Microcrystalline Cellulose from α-Cellulose

The α-cellulose hydrolysed with hydrochloric acid 2,5 N at temperature 75 °C for 15 minutes. Cold water were added and stirred strongly then placed at free air area for one night until the solution formed a suspension. Distilled water was used to washed the suspension until neutral and then dried using oven for 1 hour at temperature 60 °C. Then Microcrystalline Cellulose was saved in desiccator.

2.4 Yields of α-Cellulose and Microcrystalline Cellulose

Yields of α-cellulose and microcrystalline cellulose were determined by the following equations:

$$\alpha\text{-cellulose yield} = \frac{\alpha\text{-Cellulose weight}}{\text{Coconut fiber weight}} \times 100\ \% \quad (1)$$

$$\text{Microcrystalline cellulose yield} = \frac{\text{microcrystalline cellulose weight}}{\alpha\text{-cellulose weight}} \times 100\ \% \quad (2)$$

3 CHARACTERIZATION OF COCONUT HUSK FIBER, A-CELLULOSE AND MICROCRYSTALLINE CELLULOSE

3.1 Crystallinity

Crystalinity of coconut husk fiber, α-cellulose and microcrystalline cellulose was determined by X-Ray Diffraction (XRD) and recorded in 6100 Shimadzu. The crystallinity index was determined with Segal's empirical method (Larissa et al, 2015):

$$C_{IR}(\%) = (I_{200} - I_{am})/I_{200} \times 100 \quad (3)$$

3.2 Determination of Phsycochemical Properties of the Microcrystalline Cellulose

Some of the properties were pH, water-soluble substance and moisture content identification.

3.2.1 pH

Shake 1 g of microcrystalline cellulose for 5 minutes in 50 ml of distilled water the pH was determined with a pH meter (Achor et al, 2014).

3.2.2 Water Soluble Substances

5 g of microcrystalline cellulose was shook in 80 ml of distilled for 10 minutes, then filtered using filter paper Whatman no.1. Then dried at 100-105 °C for 1 hour to evaporate the water and weight the microcrystalline cellulose (Paul, 2008).

3.2.3 Loss on Drying

5 g of microcrystalline cellulose was dried at 100-105 °C for 1 hour then cooled in a desiccator. After then the microcrystalline cellulose was weighed. The % loss on drying was then determined as the ratio of weight of mass loss to weight of sample expressed as a percentage (Achor et al, 2014).

4 RESULT AND DISCUSSION

4.1 Yield of Process

From the process, the yield of α-cellulose was 45.44% and the yield of microcrystalline cellulose was 42.74%. From 50 g of coconut fiber, 22.72 g of cellulose was obtained. Then from 22.72 g of cellulose, 9.71g of microcrystalline cellulose was obtained. Here the results show concluded that coconut fiber contains a lot of amorphous phase in cellulose.

4.2 Crystallinity

Figure 1, Figure 2 and Figure 3 show the XRD patterns of microcrystalline cellulose, α-cellulose and coconut fiber. The sharp peak indicative the crystallinity (Larissa et al, 2015). From fig 1(a), the amorphous peak still high and the difference between amorphous peak and crystalline peak still small and it means the crystallinity still low. From fig 1(b), the amorphous peak of cellulose was lower than coconut fiber, it means the most of the lignin and hemicellulose was removed from coconut fiber and the crystallinity increased. From fig 1(c) the amorphous peak already low and the difference peak between amorphous and crystal peak was large, it means most of the amorphous region already removed from the crystal region.

From the for coconut fiber XRD characteristics, a sharp peak is resolved which is indicative of crystalline at 2θ = 21.80°. The α-cellulose XRD characteristics, a sharp peak is resolved which is indicative of crystalline at 2θ = 22.80°. Then microcrystalline cellulose XRD characteristics, a sharp peak is resolved which is indicative of crystalline at 2θ = 22.80°. The index crystallinity of coconut fiber, α-cellulose and microcrystalline cellulose was determined by Segal empirical. The results show that the crystalinity of coconut fiber, α-cellulose and microcrystalline cellulose were 43.45%, 46.54% and 75%. The results show that the crystallinity increases in the series of transformation from coconut fiber to microcrystalline cellulose. Some studies about microcrystalline cellullose percent crystallinity by hydrochloric acid hydrolysis have been report such as empty fruit bunch palm oil is 73% (Nasution et al, 2017), cotton stalks is 77%, bagasse is 76% and rice straw is 77% (Hassan and El-Sakhawy, 2005).

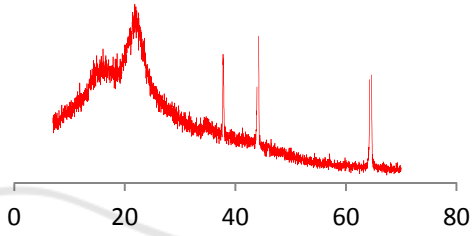

Figure 1: XRD patterns of coconut fiber.

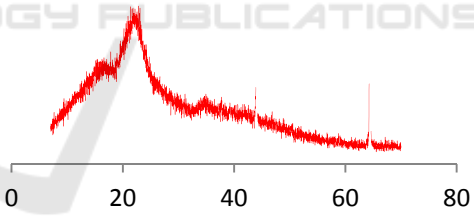

Figure 2: XRD patterns of α-cellulose.

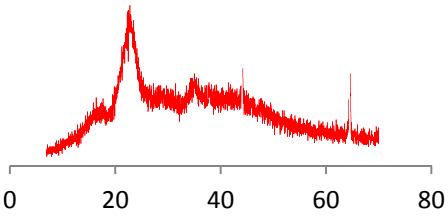

Figure 3: XRD patterns of microcrystalline cellulose.

4.3 Physicochemical Properties of Microcrystalline Cellulose

By using universal pH indicator, the pH of the microcrystalline cellulose was 7. The water soluble substance was 0.2% and the loss on drying was 5%. From British Pharmacopeia, the standard pH for microcrystalline cellulose was 5-7, the water soluble substance was <0.25% and the loss on drying was <7% (British Pharmacopoeia, 2009). From the results can be concluded that the microcrystalline cellulose in accordance with British Pharmacopeia's standard.

5 CONCLUSIONS

Microcrystalline cellulose has been isolated from coconut fiber by acid hydrolysis, after coconut fiber treated by alkali and bleaching process. it showed that the crystallinity of microcrystalline cellulose has increased because the exposure of crystalline phase after the removal of lignin via alkaline treatment and removal amorphous region of cellulose via acid hydrolysis. The physicochemical properties of microcrystalline cellulose showed that the produced material is close to British Pharmacopeia standard. Meanwhile XRD showed that increasing crystallinity of microcrystalline cellulose from α-cellulose after acid hydrolysis. The results obtained here suggest that coconut fiber is capable of being a source for production microcrystalline cellulose which can be used as reinforcing fillers in various industries.

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