Preparation of Microcrystalline Cellulose from Cotton Yarn Spinning Mills Wastes: Effect of Pretreatment and Hydrolysis Reaction Condition on the Product Characteristics

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Abstract. Cotton wastes from cotton yarn spinning mills has been utilized as an alternative resource for the production of Microcrystalline Cellulose (MCC), an important ingredient in food, cosmetic, pharmaceutical industries, etc. The main processes conducted including cotton wastes sorting, cotton lint pretreatment, and alpha cellulose hydrolysis. In this study, several parameters on pretreatment and hydrolysis were varied in order to produce MCC which characteristics were evaluated comparing to Avicel PH-101, a commercial MCC product. Pretreatment was carried out using 17.5% alkali (w/v) at temperature 80°C for 30 minutes, followed by a bleaching process at 5-30% H_2O_2 (v/v). The hydrolysis reaction was done using 1.25 M sulfuric acid, at temperature 80°C and 96°C for 2-4 hours. Results showed that pretreatment process with alkali for 30 minutes followed by a bleaching process with 5% H_2O_2 (v/v) was able to isolate 87% alpha cellulose with 99.98% of purity. The yields of MCC powders produced from acid hydrolysis of alpha cellulose at 96°C for 2, 3, and 4 hours were 74.6%, 70.2%, and 42.8%, respectively. According to the SEM, infrared spectra, and XRD results, the process conducted was applicable to produce MCC that physically and chemically similar to the characteristics of Avicel PH-101.

1 Introduction

Cotton fiber is still a famous textile material for clothing in tropical regions such as Indonesia. Along with the increasing demand of clothing textile from cotton, the cotton yarn spinning industry was still left out up to 11% of its raw material as wastes in the form of short cotton fibers [1]. Hence, some of them could not be recycled back to the spinning mills due to a down-graded fiber quality in term of fiber length, strength, micronaire, and uniformity. Characterization of cotton fiber wastes from the spinning industry has been carried out in the former study to identify the fiber characteristics as well as potential utilization of these cotton wastes as a raw material for cellulose derivatives. Results showed that according to The Classification of Cotton 2018 (USDA), both wastes obtained from open-end and ring yarn spinning mills mostly classified as very weak to weak in term of fiber strength and low to very low in term of fiber uniformity. Commonly, waste fibers generated in spinning mills can be returned to the same blend from which they came up. However, only up to 5% of wastes can be used in both carded ring-spun yarn and fine rotor-spun yarn, 2.5% of wastes used for combed yarns, and about 10 and 20% of wastes used for medium and coarser rotor yarns, respectively [1]. A study of cotton fiber wastes recycling for both conventional ring-spun and open end-rotor yarn spinning found that blend yarns with pneumafil wastes resulted with better yarn qualities. However, the blend yarns containing blowroom and flat wastes, of which the spinning wastes are mostly coming from, caused a worse yarn quality. Typically, the most deteriorated yarn properties caused by the usage of waste fiber were yarn unevenness in ring-spun yarns and tensile properties in rotor yarns [2].

Meanwhile, the cellulose content in both cotton spinning mills wastes is still very high (more than 90%). Due to the fact that cotton is a preciously high-value imported commodity in Indonesia, therefore it is important to rethink of any other utilization of the wastes into a more valuable product. Besides giving an environmental advantage for overcoming the waste problems, this utilization would also offer an economical benefit for spinning industry.

One way to process the cellulose-rich wastes into a more valuable product is by doing a partial depolymerization of cellulose with an acid hydrolysis which forms powdery fine particles called microcrystalline cellulose. This product has been largely used in food (as stabilizers, emulsifiers, bulking agents, fat substitutes, wall material for encapsulation, reinforcement of edible film) and pharmaceuticals (as binders, adsorbents, and fillers) [3,4]. Small quantities of MCC is also used in oil drilling process, paint, cosmetics, heat shield, and flame-resistant board [5]. Production of microcrystalline cellulose (MCC) utilizes cellulose that can be extracted from both wood and non-wood plants, even from bacteria [3]. Current MCCs available are generally derived from pure cotton and hardwood at very high prices [6]. The need for a cheaper raw material has been trigerred more MCC research from various raw materials, especially the agricultural residues [7-8]. The utilization of diverse agricultural residues such as rice husk, corn stalks, orange mesocarp, oil palm midrib, groundnut husk, etc for MCC extraction has been done using different acids and reaction parameters [9-11]. Most studies showed that raw material origin as well as the pretreatment and hydrolysis reaction condition affects the characteristics of products. In term of material origin, high cellulose materials are

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preferred due to a more yield and less pretreatment steps are required to isolate the alpha cellulose. Since cotton has a significantly higher cellulose content than any other agricultural residues, therefore it is a great option of raw materials for the production of MCC. Cotton stalks, cotton rags, cotton wool, cotton fabric wastes, and cotton yarn spinning wastes has been developed as potential non-woody sources of MCC. [12-17]

The MCC preparation from cotton yarn spinning mills showed a result that the prepared MCC is a good candidate as direct compression excipient for tablet formulation and manufacturing. [17] However, the preparation in the study requires more steps and also incorporates more chemicals in order to isolate the alpha cellulose and synthesize the microcrystalline cellulose. This could be a drawback according to industrial point of view and a waste recovery purpose.

This study proposes another method for MCC preparation from cotton yarn spinning mills waste which employs less steps and chemicals. Several conditions on the pretreatment and acid hydrolysis process, such as concentration, temperature, and time was observed in order to understand the effects on the MCC characteristics. In the end, the significance of this study is to propose the utilization of cotton wastes from spinning mills as a raw material for MCC at an optimum pretreatment and hydrolysis condition.

2. Materials

Cotton wastes samples was collected from a cotton yarn spinning mills around Central Java Province, Indonesia. Sodium hydroxide (NaOH) and hydrogen peroxide (H_2O_2) were used for cotton pretreatment and bleaching process. Sulfuric acid (H_2SO_4) was used for acid hydrolysis process. Avicel PH-101 was used as a standard MCC for characterization. All chemicals were purchased from Merck and Sigma Aldrich.

3. Experimental Method

3.1. Sorting of cotton wastes

The cotton wastes from spinning mills was cleaned up from any trash (including leaves, roots, branch) and even dust to get the cotton lint. About 100 gr of cotton wastes was fed into a cotton selector machine (also called Shirley analizer) to separate the lint from the trash. The cotton lint obtained from this process was weighed and the percent yield of cotton lint can be calculated.

3.2. Alkaline pretreatment and bleaching of cotton lint

The alkaline pretreatment of cotton lint was carried out using sodium hydroxide to isolate alpha cellulose. About 10 gram of cotton lint was treated with 400 ml of 17.5% NaOH (w/v) at 80ºC for 30 min, then filtered and rinsed several times with distilled water to remove the solubilized components and neutralize until pH 7 was achieved. The pretreated cotton was then undergone a bleaching treatment using 80 ml of 5-30% H_2O_2 at room temperature for 15 min. The bleached alpha cellulose was filtered and rinsed again using distilled water. Then, it was dried overnight at 70ºC in an oven until constant weight was achieved. The weight of dried alpha cellulose was recorded and the yield was calculated.

$$Percent\ yield\ of\ alpha\ cellulose\ (\%) = \frac{W_B}{W_A}\ x\ 100\%$$

where,

W_A = weight of cotton lint

W_B = weight of alpha cellulose

3.3. Acid hydrolysis

The alpha cellulose was hydrolyzed using 250 ml of 1.25 M H_2SO_4 at 80ºC and 96ºC for 2-4 hour under refluxed condition and continuous stirring. The MCC collected was vacuum filtered and rinsed using distilled water until pH 7 was achieved. Afterwards, there were samples which was fully dried by oven at 70ºC overnight and there were also samples which partly and fully dried by spray drying at 130ºC inlet temperature. The dried MCC from oven and spray dryer were weighed and the weights were recorded for yield calculation. The dried MCC from oven was grounded into fine powder.

$$Percent\ yield\ of\ MCC\ (\%) = \frac{W_C}{W_B}\ x\ 100\%$$

where,

W_B = weight of alpha cellulose

W_C = weight of obtained MCC

3.4. Scanning Electron Microscopy (SEM)

SEM was carried out in order to study the morphology of the obtained MCC. A very small quantity of the MCC powder was deposited on a carbon-coated copper sample holder. A platinum coating was laid on the sample holder with a JEOL JEC-3000FC auto fine coater. The SEM study was performed using a JEOL JSM-6510 instrument.

3.5. Fourier Transform Infrared (FTIR) Spectroscopy

FTIR analysis of untreated cotton lint, alpha cellulose, and obtained MCC was performed using FTIR Prestige 21 Shimadzu, Japan within the spectral range of 500 to 4500 cm^{-1}. FTIR spectroscopy was performed on the obtained MCC to determine the presence of functional groups which are relevant to the characteristic of MCC, also in comparison with Avicel PH-101.

3.6. X-Ray Diffraction (XRD)

The crystallinity of cotton lint, alpha cellulose, and the obtained MCC was studied by X-ray diffraction. Different patterns were obtained using a PANalytical-X'Pert HighScore.Multi-Purpose Diffractometer with Cu Kα radiation. The diffractometer was operated at 30 mA and 40 kV with scanning region of the diffraction angle (2θ) was 10º to 80º.

4. Results and discussions

4.1. Percent yield of alpha cellulose and MCC

MCC obtained in this study was extracted from the lint of cotton wastes from one of the ring spinning mills. The lint was pretreated with sodium hydroxide in order to remove lignin and hemicellulose substances. Subsequently, the bleaching process was carried out using hydrogen peroxide to remove any residual traces of lignin and hemicellulose providing a white fibrous alpha cellulose. The fine MCC powder was obtained as a result of acid hydrolysis reaction of alpha cellulose followed by filtration, washing, and finally a drying process with oven and spray dryer. The percent yield of alpha cellulose and MCC obtained at different reaction condition are presented in Table 1 and Table 2, respectively.

Tabel 1. Alpha cellulose content and yield at 80ºC, 30 min pretreatment and various bleaching agent concentrations

H_2O_2 concentration (%)	Alpha cellulose content (%)	Yield of alpha cellulose (%)
30	99.72	90
10	99.96	87
5	99.98	87

Tabel 2. Yield of MCC at several hydrolysis conditions

Temperature (ºC)	Reaction time (hour)	MCC Yield (%)
80	2	80
80	3	69
96	2	75
96	3	70
96	4	43

Table 1 shows the highest alpha cellulose yield was 90% which was obtained at 30% H_2O_2 as the bleaching agent. At higher concentration of H_2O_2, more lignin and hemicellulose was solubilized hence increasing the yield of alpha cellulose. However at high concentration of H_2O_2, alpha cellulose tends to undergo an overoxidation indicated by the turning colour of alpha cellulose from white to yellowish during storage. This can be happened especially when the H_2O_2 is not completely removed during the washing step. At lower concentration of H_2O_2 (5%), the yield of alpha cellulose was decreased to 87%. However, the alpha cellulose content was the highest and more importantly the white colour of alpha cellulose still remained during storage which indicates that the overoxidation was not happened. The importance of using less chemicals while producing a good quality of alpha cellulose surpassed the need of a higher yield of it. Therefore, the alkaline pretreament followed by bleaching process at 5% bleaching agent was chosen as the optimum pretreatment condition in this study.

Table 2 shows the yield of MCC obtained at hydrolysis reaction temperature of 80 and 96ºC and at different hydrolysis reaction times of 2-4 hr. The yield of MCC tends to decrease with the increasing of both hydrolysis reaction temperature and time. The highest yield was obtained at reaction temperature of 80ºC and the reaction time of 2 hr. However, at this temperature, MCC was not able to be dried by a complete spray drying process due to a blockage of some bigger MCC particles in the nozzle. At higher temperature of 96ºC, the MCC powder was entirely obtained by a complete spray drying process. However, the longer the hydrolysis reaction time, the less yield of MCC obtained. This could be due to a further hydrolysis of cellulose chains at higher temperature which leads to the cleavage of the β-1,4-glycosidic bonds by acids resulting in the formation of sugar molecule of glucose or oligosaccharides.

4.2. Scanning Electron Microscopy (SEM)

The electron micrographs from the SEM study are presented in Figure 1. Figure 1(a) shows the morphology of Avicel PH-101. While on Figure 1(b), (c), and (d), the effect of hydrolysis reaction temperature and time on the size and morphology of MCC particles is illustrated.

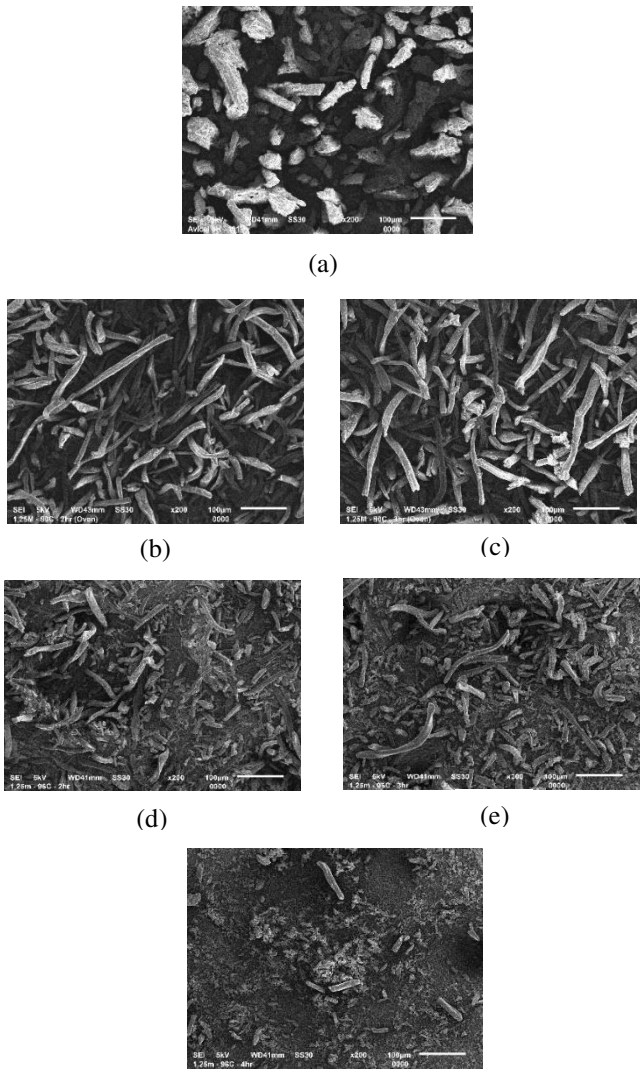

Figure 1. SEM images of (a) Avicel PH 101 and MCC at different temperature and time of acid hydrolysis reaction: (b) 80ºC, 2 hr; (c) 80ºC, 3 hr; (d) 96ºC, 2 hr; (e) 96ºC, 3 hr; (f) 96ºC, 4 hr

It is clear that the initial elongated fiber like structure of cotton is preserved after hydrolysis with sulphuric acid, which is also in accordance with the previous study [14]. During the hydrolysis at the same acid concentration, the increased reaction temperature and time tends to break and further shorten the fibrous structure of MCC particles. As can be seen in Figure 1(f), the higher temperature (96ºC) and longer time (4 hour) of hydrolysis reaction results in a finer MCC particles. This is an evident that both reaction temperature and time have an effect on the morphology and size of MCC particles. However, the morphology and size of MCC particles at 2 and 3 hours of reaction is comparable with Avicel PH-101 though it seems more fibrous and has a broader range of particle size than the Avicel PH-101. This might happen due to the fibrous origin of cotton itself as the raw material of MCC utilized in this study.

The acid hydrolysis temperature and time plays a significant role on the morphology of the obtained MCC. The fibrous structure of cotton cellulose is degraded more rapidly at a higher acid hydrolysis temperature and reaction time. This is in accordance with previous study which recommends the acid hydrolysis reaction for manufacturing MCC at boiling temperature using a mineral acid solution (sulphuric acid) 15 wt. % for 1 hour [18].

4.3. Fourier Transform Infrared (FTIR) Spectroscopy

The effect of hydrolysis reaction time on the presence of functional groups is studied by using FTIR spectra. Figure 2 below shows the FTIR spectra of MCC obtained at acid hydrolysis reaction temperature of 96ºC and reaction time of 2, 3, and 4 hour represented by a black, red, and blue curve, respectively. The green curve corresponds to the FTIR spectra of Avicel PH-101 as the standard MCC for characterization purposes.

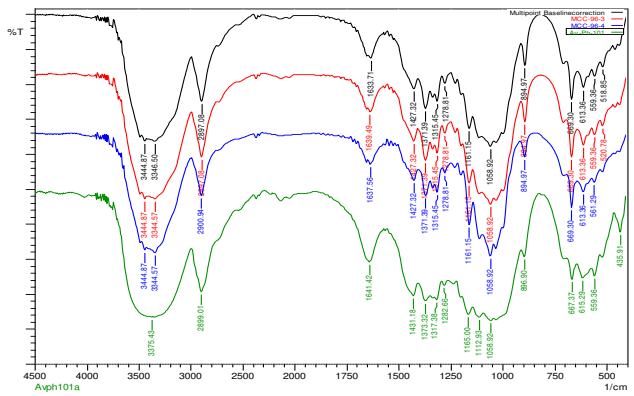

Figure 2. FTIR spectra of MCCs obtained at different hydrolysis reaction time: 2 hour (black), 3 hour (red), and 4 hour (blue), and Avicel PH-101 (green)

From Figure 2, we can see that all the observed MCC samples has similar spectra and nearly the same absorption bands with Avicel PH-101. This means the same functional groups as in Avicel PH-101 are also emerged in all MCC samples. In Avicel PH-101, the absorption bands appear at 3375 cm^{-1} which corresponds to the stretching vibration of –OH groups in cellulose and also at 2999 cm^{-1} which represents C-H stretching vibrations of $–CH_2–$ groups. The absorption band at 1641 cm^{-1} corresponds to the –OH bending of adsorbed water mingled with C=O in the aldehyde on the terminal anhydroglucose unit [14]. This is related to the water molecules bending modes due to a strong interaction between cellulose and water [9]. As the hydrolysis time was differed from 2 to 4 hours, there is no significance different on the FTIR spectra of all MCC samples. The typical cellulose functional groups are still maintained in between 2-4 hour hydrolysis reaction time. In addition, there are no absorption bands arised at 1700-1740 cm^{-1} and 1509-1609 cm^{-1} on the spectra, which means the hemicellulose and lignin, respectively, are completely removed from the cellulose during the treatment leading to the pure MCC product [9]. The IR spectra result also in accordance with the MCC results prepared from waste cotton fabric and cotton wool with different hydrolysis method and acid hydrolysis condition, respectively. [14,16]

4.4. X-Ray Diffraction (XRD)

The diffraction patterns of MCC obtained at different reaction time are presented in Figure 3. The XRD studies were performed to investigate the crystalline structure of of pre-treated cellulose, MCC, and commercial MCC (Avicel PH-101). Due to chemical treatments, the noncellulosic moieties and amorphous region of

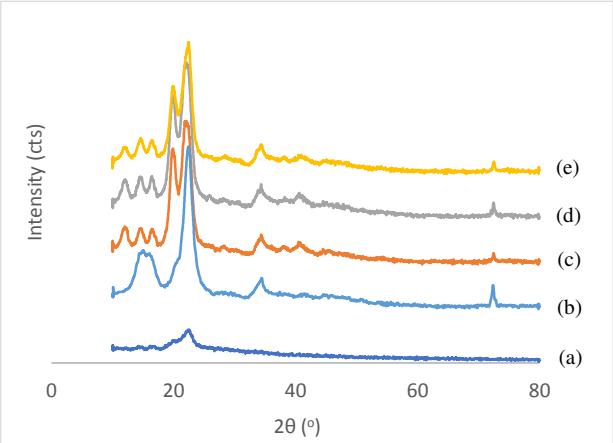

Figure 3. X-ray diffraction patterns of (a) Alpha cellulose of cotton waste, (b) Avicel PH-101, and MCC from cotton waste at different time of hydrolysis reaction: (c) 2 hour, (b) 3 hour, (c) 4 hour

cellulosic materials were removed resulting in an increased degree of crystallinity. Figure 3 (a) shows the XRD pattern of alpha cellulose that is highly amorphous as exhibited by the low intensity of peak at 22.5º angular position of 2θ and it represents cellulose I structure. This amorphous region defines more disordered structure which means low crystal region as well as low degree of crystallinity [19, 20]. Figure 3 (b) represents the XRD pattern of cellulose I from Avicel PH-101 with high intensity of crystal region as indicated by a high and sharp peak appeared at angular positions of 2θ 22.5º. The sharp peaks indicating the high crystalline regions also present

in MCC samples obtained in this study. However, as seen in Figure 3 (c), (d), and (e), the MCC samples show doublet intensity of the main peaks around 2θ 20° - 22.5° which confirms the presence of both cellulose I and cellulose II allomorphs [19]. These XRD patterns represent different crystal lattice type of cellulose compared to the previous MCC studies from cotton plant sources, which mainly showed cellulose I type [12-16].

The transformation of cellulose I to cellulose II can be happened due to dissolution or regeneration and swelling of cellulose in a concentrated alkali [20]. In this study, the cellulose fiber swelled during the pre-treatment process using a concentrated alkali, but the cellulose chains were still maintained in a parallel direction. This is confirmed by the XRD pattern of alpha cellulose which shows a typical cellulose I diffractogram (Figure 3 (a)) and also in accordance with previous study of MCC extraction from fodder grass [6]. However, due to a subsequent hydrolysis reaction, the cellulose crystal in parallel microfibrils were re-arranged in opposite direction which leads to an anti-parallel arrangement. This arrangement corresponds to cellulose II chain structure which is thermodynamically more favourable than the parallel arrangement and also has more stable structure. These properties make it preferable for various application [20].

From the XRD patterns, it is clear that there is a significant difference between the main peak intensity of alpha cellulose and MCC samples. This difference implies an increasing of crystalline structure in MCC as a result of successful removal of amorphous region through an acid hydrolysis reaction. The tightly packed cellulose chains are existed in the crystalline regions of cellulose and this is also stabilized by strong and complex hydrogen-bond network. Therefore, it is more resistance to an acid hydrolysing agent. As a result, the amorphous regions are preferentially attacked by the acid and undergo hydrolysis reaction first, resulting in a removal of amorphous regions and a formation of high crystalline cellulose [20]. The high crystallinity property of a material is promising to be used for a better reinforcement agent for composite materials [21].

5. Conclusions

Microcrystalline cellulose (MCC) powder from various process conditions was successfully obtained. Of the process variations that have been carried out, alpha cellulose has been successfully isolated from clean cotton fibers (cotton lint) through the pretreatment process at 17.5% NaOH concentration for 30 minutes and bleaching process at 5% H_2O_2 concentration for 15 minutes. This shorter pretreatment time (30 minutes) as well as less bleaching agent (H_2O_2 5%) concentration has been considered as the best process condition for cotton pretreatment. From this pretreatment process, 87% alpha cellulose yield was successfully achieved with the highest purity of alpha cellulose (99.98%). According to the SEM results, the alpha cellulose hydrolysis at reaction temperature of 96°C and sulfuric acid concentration of 1.25 M for 2 hour was able to generate MCC powders which has rod fibrous morphology comparable with Avicel PH-101. The results of the FTIR showed the emergence of functional groups which are similar to Avicel PH-101. The XRD pattern analysis shows the increasing of crystalline region in the MCC samples indicated by a pronounced peak at around angle position of 2 theta 22.5° which is around the main peak of Avicel PH-101.

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