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RESEARCH PAPER

Characterization of microcrystalline cellulose from fast-growing species *Artocarpus elasticus*

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Abstract. Microcrystalline cellulose is an important derivative of cellulosic material obtained from wood and non-wood sources, and is used for pharmaceutical, food, cosmetics, and other industries. The aim of this study was to determine the effect of various hydrochloric acid concentrations on the characteristics of cellulose microcrystals isolated from terap wood (*Artocarpus elasticus*). The microcrystalline cellulose was hydrolyzed using hydrochloric acid, at concentrations of 1.5 N, 2.5 N, and 3.5 N for 15 minutes, and within a temperature range of 100-105 °C. The samples were then analyzed for changes in color and functional groups with Fourier Transform Infrared spectroscopy (FTIR), while crystallinity index was evaluated through X-Ray Diffraction Analysis (XRD). The FTIR results showed similarity spectrum patterns between α -cellulose and microcrystalline cellulose, while X-Ray Diffraction confirmed the highest crystallinity index was from 2.5 N of cellulose I (69.395 %) and cellulose II (82.73 %).

Keywords: cellulose; microcrystalline cellulose; fast-growing species; crystallinity index; *Artocarpus elasticus*

1. Introduction

Cellulose is one of the biggest biopolymer in the world, with production rate of about 100 billion ton per year (Hermawan, 2017). In addition, there have been wide applications in various industries, as seen with food, paint, and biopolymers (Naduparambath & Purushothaman, 2016). Cellulose can be hydrolyzed using acids to remove amorphous region and produce microcrystalline cellulose. Microcrystalline cellulose (MCC) is a white crystalline powder that is recognized as one of the widely used cellulose derivatives in cosmetics, food, pharmaceuticals, and filler industries. MCC is generally produced from cellulose by alkali treatment, although acid

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hydrolysis method (Naduparambath & Purushothaman, 2016; Trache et al., 2016) is faster than other methods. Several studies have reported the results of acid hydrolysis method, such as from bagasse (Zulharmita et al., 2017) and palm oil fiber (Xiang et al., 2016) produce high crystallinity of MCC. The solution used in this process is hydrochloride acid (HCl), due to the low price and easy availability. The acid causes more reactive agent for removing amorphous part of cellulose compared to when HNO₃ and H₂SO₄ are used (Nawangsari, 2019).

Although cotton is the main source for the commercial production of MCC, other sources such as sago seed sheel, palm oil fiber, and bagasse have also been reported to produce MCC. The properties of MCC depend not only on the acid type, but also on the source where it is extracted from. Terap wood (*Artocarpus elasticus*) is one of fast growing tree species that is naturally and abundantly distributed in the secondary forest of South Kalimantan, Indonesia (Istikowati et al., 2014). To date, research attention has been mainly focused on physical, mechanical and anatomical characteristics of terap wood (*Artocarpus elasticus*) as a wood and no investigation has been carried out on terap wood (*Artocarpus elasticus*) as a cellulose source for advanced applications. From the previous research, the wood contains 78.0% holocellulose, 50.7% α -cellulose, and 29.7% lignin (Istikowati et al., 2016). The high cellulose content is a potential source for MCC production. Therefore, the main objective of this study is to evaluate the effect of HCl concentration on the properties of MCC produced through acid hydrolysis method.

2. Material and Methods

2.1. Materials

Terap wood (*Artocarpus elasticus* Reinw. ex Blume) were collected from secondary forest in the Education Forest at Lambung Mangkurat University, Mandiangin, South Kalimantan, Indonesia. Hydrochloric acid, acetic acid, ethanol, sodium hydroxide, sodium chlorite were obtained from E. Merck, German, and used without further modification.

2.2. Methods

Terap wood (trunk part) were dried for 3 days, mashed to powder 40-82 mesh size and then dried in the oven at 100-105 °C until a constant weight was obtained. Holocellulose and α -cellulose components were isolated based on the ASTM D1107-96 and the ASTM D 1103-60 standards, respectively. MCC was obtained from the latter. A total of 0.5 grams α -cellulose was hydrolyzed using HCl 1.5; 2.5; and 3.5 N (12 mL) in beaker glasses, within a period of 15 minutes. After that, 25 mL of cold water was added and stirred vigorously to stop the hydrolysis process, and then reserved for 1 night. The MCC obtained were then filtered, washed with distilled water to attain a neutral pH, and subsequently dried in the oven at 60 °C for 30 minutes.

2.3. Characterization

The MCC obtained was analyzed using colorimeter, Fourier Transform Infrared (FTIR), and X-Ray Diffraction Analysis (XRD). A total of 100 mg MCC was analyzed using colorimeter and the result was compared to the Avicel PH 102 standard, while the color was compared with the Ditjen POM 1979 standard. In addition, the sample pellet (2-5 mg) was combined with KBr (200-250 mg) and was measured with FTIR in the region of 4000-400 cm⁻¹ wave numbers. The infrared spectra was used for estimation of the Total Crystallinity Index (TCI) (Nelson & O'Connor, 1964). MCC was also mashed into powder and was analyzed with XRD to determine the crystallinity index (CrI) from cellulose, based on the Segal formula (Segal et al., 1959).

3. Results and Discussion

3.1. Holocellulose and α -Cellulose Isolation

Terap (*Artocarpus elasticus*) is one of unutilized native tree species from South Kalimantan. The species is naturally distributed and found abundantly in the secondary forest in South Kalimantan, Indonesia (Istikowati *et al.* 2014). Fig. 1 shows the photograph of terap (*Artocarpus elasticus*) tree species.



Figure 1. Photograph of terap tree (*Artocarpus elasticus*)

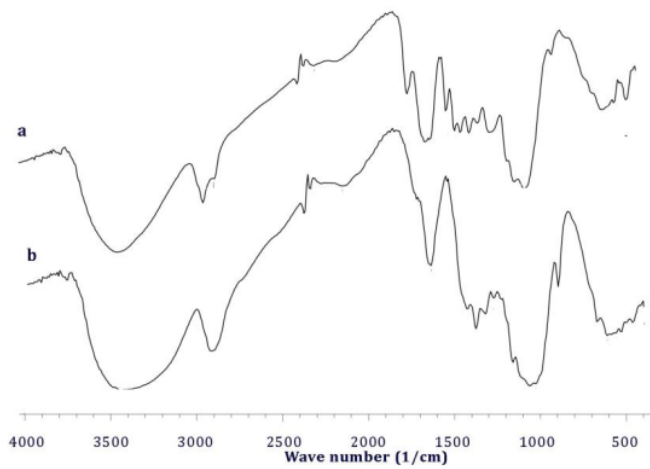


Figure 2. FTIR spectrum of (a) holocellulose and (b) α -cellulose of terap wood (*Artocarpus elasticus*)

Holocellulose and α -cellulose of terap wood (*Artocarpus elasticus*) were analyzed with FTIR to determine the functional group present (Fig. 2), and different absorption patterns were observed in wave number with a shift because the absence of hemicellulose. For example, the area of 3400-2900 cm^{-1} is typical for stretching O-H group and C-H. Table 1 shows the holocellulose absorption at a wave number of 1512.19 cm^{-1} which features the stretching vibration of C=C, indicating lignin compounds. In addition, wave number vibration 1735.93 cm^{-1} also appeared because of the amorphous parts (Sunardi et al., 2018).

Table 1. Holocellulose dan α -cellulose specific peaks of FTIR of terap wood (*Artocarpus elasticus*)

Wave number (cm^{-1})		Functional group
Holocellulose	α -cellulose	
3425.58	3448.72 3410.15	stretching O-H
2924.09	2916.37	stretching C-H
1735.93	-	hemicellulose stretching C-H
1635.64	1635.64	Water absorption O-H bonding
1512.19	-	C=C in lignin
1373.32 1327.03	1373.32 1319.31	-O-in cellulose
894.97, 1064.77 1033.85	894.97 1064.71	C-O stretching glycoside bonds

3.2. MCC Isolation

The percentage of MCC obtained from HCl hydrolysis of 1.5 N, 2.5 N, and 3.5 N was 71.05%, 57.64%, and 53.23%, respectively (Fig. 3). Based on Tukey test, significant differences were recognized at the three different formulations. According to Sumiati (Sumiati et al., 2016), higher HCl concentration in the hydrolysis process will remove not only amorphous phase, but also crystalline part of cellulose. This result also similar with the result from previous research (Sunardi et al., 2019). Also, there is an increase in glucose monomer formation, leading to improved dissolution in the washing process.

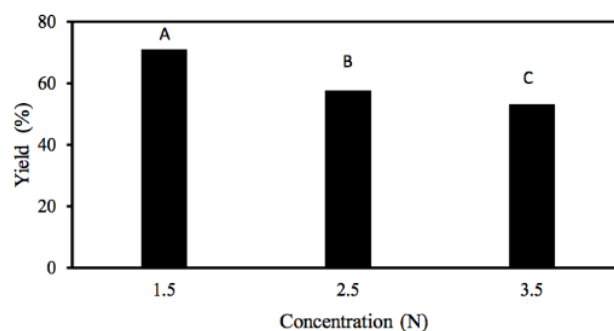


Figure 3. MCC yield in various concentration of HCl (A) 1.5 N; (B) 2.5 N; (C) 3.5 N)

3.3. Characteristic of MCC

3.3.1. MCC color analysis

Table 2 shows that the color of MCC in 1.5 N HCl concentration has L* value which is similar with L* standard (87.01). Meanwhile the value of a*, b*, and ΔE increased with the increase of acid concentration. In addition, the indicators used to determine good MCC color was L* value or the brightness level, and significant differences were reported between each group in this study.

Table 2. MCC colour of terap wood (*Artocarpus elasticus*)

No	HCl Concentration	L*	a*	b*	ΔE
1	Standard (white paper)	88.51	1.94	3.60	
2	1.5 N	87.01	5.16	16.24	4.76
3	2.5 N	82.15	6.66	16.58	15.66
4	3.5 N	80.83	6.76	17.39	16.95

3.3.2. Fourier Transform Infrared Spectroscopy Analysis of MCC

The infrared spectrum of α-cellulose and MCC obtained from terap wood (*Artocarpus elasticus*) in this study are shown in Fig. 3. The aim of the infrared spectroscopy analysis was to determine the functional groups of the α-cellulose and MCC as a α-cellulose derived. FTIR spectroscopy results of the α-cellulose and MCC with various HCl concentrations shows similarity in the main absorption pattern functional group area. This was indicated by a shift in the number and transmittance value of O-H stretching group, observed to be lower at 1.5 N and 2.5 N, followed by a subsequent increase at 3.5 N HCl (Fig. 4).

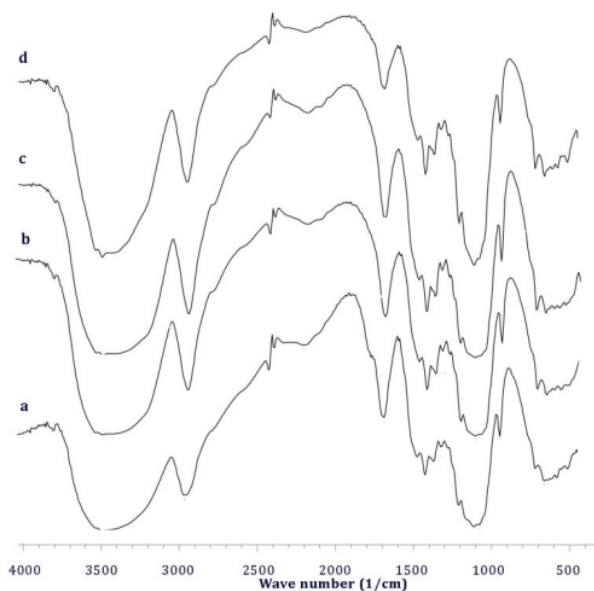


Figure 4. FTIR spectrum of cellulose and MCC obtained from terap wood (*Artocarpus elasticus*) with various HCl concentration (a) α-cellulose; (b) 1.5 N; (c) 2.5 N; (d) 3.5 N

The wave number pattern of α -cellulose and MCC has similarities with commercial cellulose at 894.97, 1035-1060 cm^{-1} indicating the presence of C-O-C stretching (glycosidic bond). Also, the region of 1374 cm^{-1} and 1373 cm^{-1} shows the presence of -O- group in cellulose (Naduparambath & Purushothaman, 2016). The O-H and C-H stretching on α -cellulose and MCC treated with different HCl concentrations denote a shift in wave number because of hydrolysis process.

The O-H stretching groups of α -cellulose identified in this study are formed at wave numbers of 3448.72 and 3410.15, while C-H was at 2916.37 cm^{-1} . Furthermore, MCC treated with 1.5 N HCl showed O-H at 3441.01 and 3371.57 cm^{-1} , while C-H was at 2900.94 cm^{-1} . The samples with 2.5 N treatment exhibited stretching O-H groups at 3441.01 and 3363.86 cm^{-1} , while C-H was at 2893.22 cm^{-1} . In addition, MCC with 3.5 N HCl showed O-H groups at 3441.01 and 3387.00 cm^{-1} , while C-H was 2893.22 cm^{-1} .

The FTIR peak in MCC with wave number of 3441.01 – 3387 cm^{-1} indicates a decline in O-H stretching group, while the absorption pattern widens, using the 1.5 and 2.5 N HCl treatments. In addition, lower peak pattern cause an increase in the functional group absorbance, thus indicating the existence of intermolecular hydrogen bonds in the cellulose molecule.

Nelson and O'Connor method was used to determine the Total Crystallinity Index (TCI). This refers to the ratio of absorbance in wave number of 1373-1375 cm^{-1} indicating -O- functional group, and also 2800-2900 cm^{-1} for C-H and CH_2 (A_{1373}/A_{2800}) stretching (Nelson & O'Connor, 1964; Sunardi et al., 2018). Table 3 shows the TCI value of MCC.

Table 3. TCI MCC of terap (*A. elasticus*) wood

HCl Concentration (N)	TCI
1.5	0.992
2.5	0.993
3.5	0.963

The Total Crystallinity Index (TCI) estimates the influence of varied HCl concentrations, which increased from 1.5 to 2.5 N, followed by a decline at 3.5. The data obtained showed that the treatment with 2.5 N gave the highest TCI value. Thus, the treatment is concluded as the optimum parameter for hydrolysis, due to the adequate loss of amorphous cellulose (Klemm et al., 1998).

3.3.3. X-Ray Diffraction Analysis

Fig 5 shows the XRD analysis of MCC, and there are similarities in the patterns obtained from three different HCl concentrations. In addition, all samples showed two diffractogram peaks in the crystalline area in the (002) and (200) Miller index with an angle of $(2\theta) = 21-22^\circ$. These indicate the presence of two types of crystal cellulose, interpreted as cellulose I and II (Klemm et al., 1998; Xiang et al., 2016). Moreover, treatment with 1.5 N HCl shows the crystallinity index of cellulose I and II at (2θ) angle = 21.95° and 20.05° with intensity value of 549 and 596, while the amorphous area appears at (2θ) angle 12.15° and 14.05°, with 170 and 119 intensity. Hence, the crystallinity indexes were computed as 69.03% and 80.03%, respectively. Conversely, 69.39% and 82.73% was recorded for MCC with 2.5 N treatment, which appeared at (2θ) angle = 21.895° and 20.03° with intensity value 549 and 608, while the amorphous area was at (2θ) angle = 12.26° and 14.05° with intensity of 168 and 105. In addition, the crystallinity index of 67.25% and 75.86% were recorded in 3.5 N treatments for cellulose I and II. This appeared at (2θ) angle = 22.57° and 20.00

° with intensity value of 455 and 414, while the amorphous area was at (2θ) angle 12.10° and 14.05° with intensity of 149 and 100, respectively.

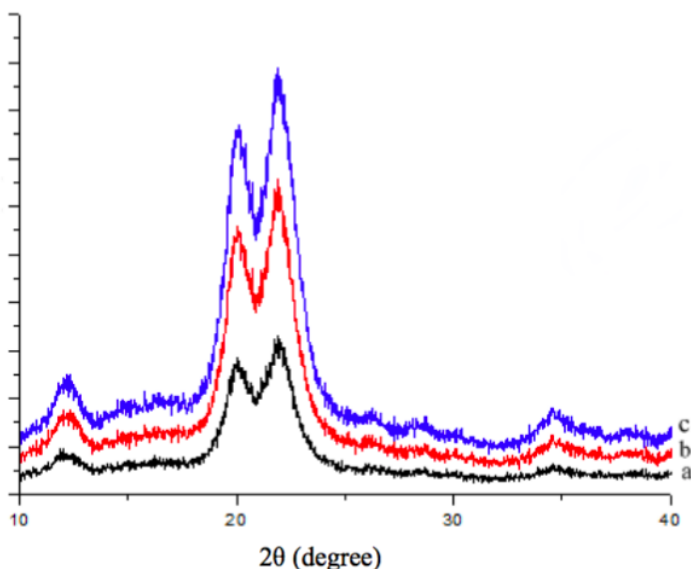


Figure 5. Diffractogram of MCC from terap (*A. elasticus*) wood in various HCl concentration (a) 1.5 N; (b) 2.5 N; and (c) 3.5 N

Table 4 shows the CrI of MCC where it can be seen that the value of cellulose II is higher than the value of cellulose I due to the relatively better stability. Furthermore, the CrI was reported to have increased from HCl concentration of 1.5 N to 2.5 N, followed by a decline at 3.5 N. This phenomenon occurs due to the cutting speed of amorphous areas in cellulose during hydrolysis (Steven et al., 2014). Based on the characterization result, 2.5 N was confirmed to be the optimum HCl concentration needed to isolate MCC from terap wood.

Table 4. Crystallinity index of MCC obtained from terap wood (*Artocarpus elasticus*)

No	HCl concentration (N)	Crystallinity index cellulose I (%)	Crystallinity index cellulose II (%)
1	1.5	69.03	80.03
2	2.5	69.39	82.73
3	3.5	67.25	75.86

4. Conclusion

FTIR analysis shows the similarity peak patterns between α -cellulose and MCC from terap wood. In addition, XRD identified the highest crystallinity index in cellulose I (69.395 %) and cellulose II (82.73 %), using 2.5 N HCl. This is further recommended as the optimum acid concentration to isolate MCC from terap wood through the hydrolysis process.

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