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PRODUCTION OF BIOPLASTIC FROM AVOCADO SEED STARCH REINFORCED WITH MICROCRYSTALLINE CELLULOSE FROM SUGAR PALM FIBERS

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Abstract

The influence of microcrystalline cellulose derived from sugar palm fibers and glycerol on the mechanical properties of bioplastics from avocado seed starch was studied. Sugar palm fibers underwent alkali treatment, bleaching, and hydrolysis with HCl to produce microcrystalline cellulose. Bioplastic was successfully fabricated through solution casting technique. It was prepared from avocado seed starch and reinforced with MCC from sugar palm fibers with composition ratio of 6:4; 7:3, 8:2, and 9:1 (w/w). Glycerol functioned as plasticizer with variation of 0.1; 0.2; 0.3 and 0.4 (v/w of starch). Microcrystalline cellulose was dissolved in NaOH 5% (w/v) before it was mixed with the plasticized starch. Microcrystalline cellulose degree of crystallinity determined by X-Ray Diffraction was 97.5%. Morphological analysis was done with Scanning Electron Microscope given that the isolated microcrystalline cellulose from sugar palm fibers are rod-like shaped with diameter of 5.55-9.44 μm and crystal size of 25.08 nm. Mechanical properties of bioplastic were determined by tensile strength and elongation at break analysis where it is given that the best condition of bioplastics obtained is from 7:3 ratio with 0.2 (v/w) glycerol added which is 2.74 MPa for tensile strength and 3.16% for elongation at break. FT-IR analysis showed the functional groups of bioplastics, majority of O-H groups, that was found from the addition of microcrystalline cellulose that represented substantial hydrogen bonds.

Keywords: Avocado seed, Glycerol, Microcrystalline cellulose, Sugar palm fibers.

Nomenclatures

B	The half-height width of the diffraction band
CrI	Crystalinity index , %
I_{002}	The crystallite height
I_{AM}	Amorphous height
K	The Scherrer constant
L	Crystallite size, nm

Greek Symbols

λ	The X-ray wavelength (Å)
θ	The Bragg angle, deg.

Abbreviations

FTIR	Fourier Transform Infra Red Spectroscopy
MCC	Microcrystalline Cellulose
SEM	Scanning Electron Microscope
XRD	X-ray Diffraction

1. Introduction

Nowadays, demand for plastic products is increasing. The need for such large quantities of conventional plastics and their dominance over other materials is due to their excellent lifelong properties which include resistance to chemical reactions, especially enzymatic reactions [1]. However, the use of conventional plastics for short-life application is not sustainable due to the increase of plastic waste that evokes environmental concern [2]. This is caused by the fact that plastics are derived from crude oil, a non-renewable fossil fuel, subsequently produces high levels of environmental pollution [3]. Biodegradable polymers commonly called bioplastic made from renewable resources as a possible alternative to reduce the number of landfill waste and other environmental issues [4].

Starch is a natural biodegradable polymer that has the potential to replace synthetic polymers used for limited time applications, i.e., disposable packaging and cutlery [2]. It is commonly found in the form of biodegradable films for varied applications as it is a renewable, abundant and inexpensive material [5]. In Indonesia, starch sources are in abundance. Avocado seed for instance, usually discarded [6], has around 30% starch content. Hence it serves as a potential starch source [7, 8].

Researchers and industries have been interested in natural fibres owing to their more specific advantages, i.e., reducing tool wear, low cost and density per unit volume compared to conventional or synthetic fibers [9]. Furthermore, its sustainable, renewable and degradable features are some of the important properties making them suitable as filler in polymer composites [10]. Cellulose has been proven to be the most promising material for reinforcing fillers [11]. Ijuk in Indonesia known as sugar palm fibers from Arenga Pinnata plant is an environmental-friendly material with 52.3% cellulose content [12, 13].

The surface of the natural fibers can be modified by physical, mechanical, and/or chemical means [10]. De Mesquita et al. (2010) reported that the

fabrication of cellulosic materials into micro and nano dimensions gives favorable characteristics such as excellent mechanical properties, high crystallinity, and low molecular weight [14]. Microcrystalline cellulose (MCC) is a pure partially depolymerized non-fibrous cellulose which is a white, odourless and tasteless crystalline powder composed of porous particles [15]. The strong interactions between MCC and starch film matrix play a key role in reinforcing the composites [16].

Starch-based bioplastic has poor dimensional stability and mechanical properties. By adding in plasticizer, it improves the workability also suppresses film brittleness [17]. Bioplastic from avocado seed waste and sugar palm fibres can reduce environmental issues from the excess use of conventional plastic [8]. The aim of this research is to obtain the influence of MCC derived from sugar palm fibers and glycerol on the mechanical properties of bioplastic.

2. Materials and Methods

2.1. Materials

Avocado seed was obtained from avocado merchants at Jalan H.M. Joni, Medan, Indonesia. Sugar palm fibers (ijuk) were collected from Binjai, Indonesia. NaOH, NaClO 60%, H_2O_2 10% for delignification and bleaching process, glycerol and HCl were purchased from Rudang Jaya, Medan, Indonesia [8].

2.2. Starch isolation

First, peeled and washed 100 g avocado seeds and shredded into small pieces. Then it was blended in 100 ml water to give starch slurry. Next, it was filtered and placed in a tank for an hour for settling purpose. Starch sediment was formed and separated from the slurry and proceed to washing with distilled water. Settling was done for three times. Starch sediment obtained was dried in an oven at 60^{o}C. Finally it was sieved in 100 mesh strainer to get homogenous size starch [8].

2.3. Extraction of α-cellulose from sugar palm fibers

Sugar palm fibers were cleaned to remove impurity and cut into small pieces. In a beaker glass, put 50 g of cleaned and cut sugar palm fibres into 700 ml HNO_3 3.5% which contains 8 mg of $NaNO_3$. The mixture was heated on a hotplate at 90^{o}C for 2 hours. Then sugar palm fibres were washed repetitively with water until neutral pH was reached. Neutral pH fibers were added into a mixture of NaOH and Na_2SO_3 2%. The second mixture was heated at 50^{o}C for an hour and put into washed until neutral pH was reached. Finally the fibers were heated in 340 ml of NaClO 3.5% to boil for 10 minutes and rinsed with water.

The α-cellulose was purified with 340 ml NaOH 17.5% solution at 80°C for 30 minutes. Sample was washed until pH was neutral and filtered. Then it is bleached with H_2O_2 10% at 60°C for 30 minutes. Last, sample was washed until pH was neutral and filtered [8].

2.4. Isolation of MCC from α-cellulose

5 g α-cellulose was dissolved in 120 ml HCl 2.5 N at boiling temperature for 15 minutes. Then it was put into cold water, strongly stirred with spatula and placed

in an open air for one night until suspension was formed. Suspension formed was washed until pH was neutral. Finally, it is dried in an oven at 60°C for an hour. MCC was put in a desiccator [8].

2.5. Film preparation

10 g of dried starch and MCC were prepared in different ratios, 6 : 4, 7 : 3, 8 : 2, and 9 : 1. Then MCC was dissolved in NaOH 5% (w/v) solution and starch was dissolved in distilled water with starch : distilled water = 1 : 10 (w/v) ratio. Starch solution was heated and stirred on a hotplate for 10 minutes. Glycerol was added after into the starch solution as plasticizer in 0.1; 0.2; 0.3 and 0.4 mL/g of starch mass. The mixture of starch and glycerol was heated. At 70°C, MCC was added into the mixture and kept heated until 85°C. Next, the mixture was cooled, poured onto a flat mold and went to drying at 60°C for 24 hours. The bioplastic obtained from the process was removed from the mold, placed in a desiccator and ready to be analyzed [8].

3. Characterization of Products

3.1. Characterization of starch

Starch was characterized by the contents of starch, amylose and amylopectin, protein, ash and moisture. Chemical compositions of avocado seed starch were shown in Table 1 [8].

3.2. Characterization of MCC

MCC was characterized by its crystallinity and functional groups. It also underwent morphological analysis. The crystallinity was determined by X-Ray diffraction (XRD) (PanAnalytical X'Pr Pro) operating with CuKα radiation (λ= 1,5406 nm) at 35 mA and 40 kV. The crystallite height 002 (I_{002}) and amorphous height (I_{am}) were used to calculate the apparent crystalline index (apparent Cr.I.) following Eq. (1) [18]:

$$CrI = \left[\frac{I_{002} - I_{AM}}{I_{002}} \right] x100 \quad (1)$$

The apparent crystallite size L was calculated following Stokes and Wilson method (Eq. (2)) using integral breadth. Integral breadth determines the volume average of the thickness of the crystallites measured perpendicular to the reflecting plane [19].

$$B(2\theta) = \frac{\lambda}{L.\cos(\theta)} \quad (2)$$

Functional group was analyzed using Fourier Transform Infrared (FT-IR) (Shimadzu IR-Prestige 21). The spectrum was recorded in the range of 4000 cm^{-1} and 400 cm^{-1}. The morphology was analyzed using Scanning Electron Microscope (SEM) (JSM-5310 scanning electron microscope) at an applied accelerating voltage and current, 10 kV and 5 μA respectively [8].

3.3. Mechanical properties of bioplastic

Mechanical properties of bioplastic were analyzed through tensile strength and elongation at break. Tensile strength test was based on ASTM D638 standard where product was chosen and cut to form specimen [8].

3.4. Characterization of bioplastic

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4. Results and Discussion

4.1. Characterization of avocado seed starch

100 g of avocado seed could produce 16% of starch or 16 g of starch. It gave brown starch with particle in the size of 100 mesh. Chandra et al. (2013) reported that the phenolic compounds namely dopamine (3,4-dihydroxyphenylalanine) in avocado seed can cause enzymatic browning reactions because of oxygen [20].

Characterization of avocado seed starch was conducted to know the percentage of each component contained in the starch produced including starch, amylose, amylopectin, water, ash, and protein content to determine the quality of starch produced [8, 21]. Composition of isolated starch is shown in Table 1.

Table 1. Composition of isolated starch from avocado seeds.

Component of avocado seed starch	Percentage (%)
Water	16.6
Ash	0.23
Lipid	1.09
Protein	2.16
Amylose	0.07
Amylopectin	73.55

4.2. MCC yield

MCC derived from sugar palm fibers made gave 60% of yield [8]. In this research, the MCC produced was a good material as it was white odorless granular powder.

4.3. Crystalinity and crystallite size of MCC

Crystallinity and crystallite size of MCC was determined with X-ray Diffraction (XRD). The XRD pattern of MCC sample prepared from sugar palm fibers is shown in Fig. 1.

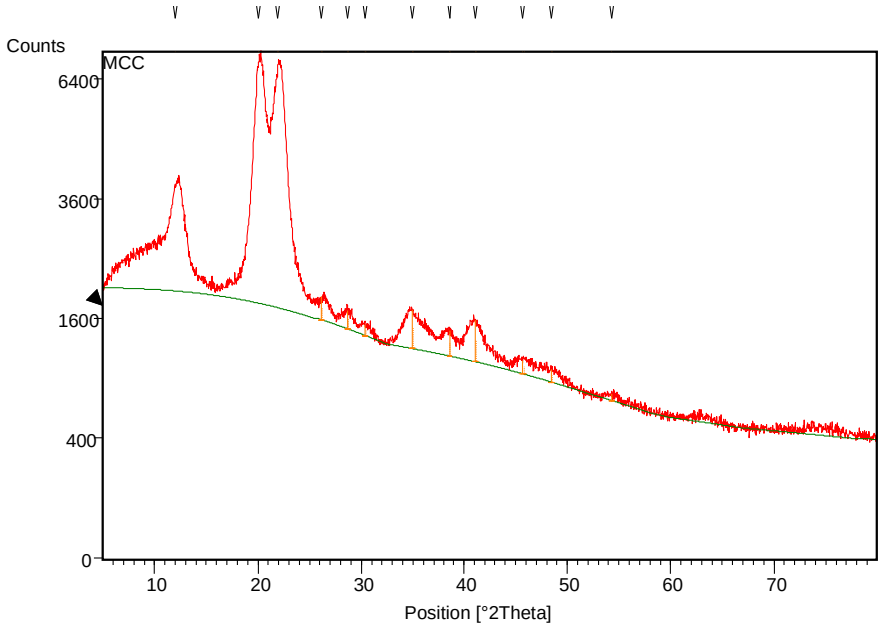

Fig. 1. XRD patterns of MCC samples derived from sugar palm fibers.

The absorption peaks that occurred at 2-theta (2θ) were 12.01^{o} (I_{am}) and 20.06^{o} (I_{002}) [8]. The sharp peaks indicate high crystallinity degree in MCC structure [8, 22].

High crystallinity index of MCC obtained was 97.5% [8]. It is consider high, as other non-wood indexes are at 52 - 53% [22]. High crystallinity is related to acid hydrolysis process in MCC preparation where in cellulosic materials, amorphous regions are disintegrated, resulting in highly crystalline substrate with differen degrees of crystallinity index [23]. Karim et al. (2014) reported for the treated sample, disordered amorphous regions decrease with the increase of hydrogen bond crystalline region in cellulosic matrix, which might be due to the partial breaking up of glycosidic linkages inside the amorphous region while the crystalline region almost unaltered [8, 24].

Crystallite size can be calculated from XRD result where the absorption peak of the spectra produced was at $2\theta = 20.06^{o}$. By calculating the crystallite size of MCC at the absorption peak $2\theta = 20.06^{o}$, we can get crystals diameter which is equal to 25.08 nm. Crystallite size affects the compatibility or the compactness of a material where the smaller the size, the more its compatibility of a material improved [25].

4.4. Tensile strength of bioplastics

The effects of microcrystalline cellulose derived from sugar palm fiber and glycerol to bioplastic tensile strength are shown in Fig. 2.

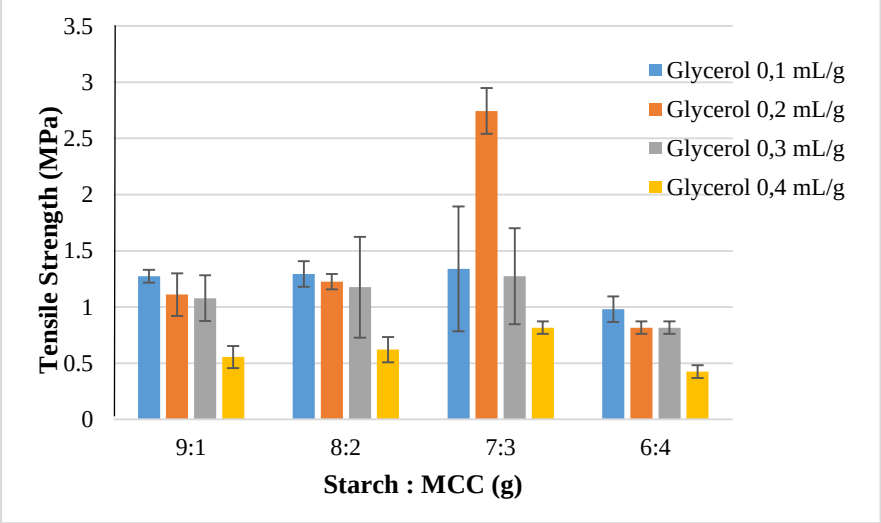

Fig. 1. The effect of MCC from sugar palm fibers and glycerol addition on tensile strength of bioplastics.

Figure 2 shows that with the increase mass of MCC, tensile strength for bioplastics increases. 7 g of MCC and 0.2 v/w glycerol gave the maximum tensile strength at 2.74 MPa. The addition of MCC on gelatinized starch films resulted in intermolecular hydrogen bonding to group which cause molecular bonds between amylose to be more compact [26].

A decrease in tensile strength value is shown for bioplastic with 4g MCC for all variation of glycerol. According to Wittaya (2009) this deviation is possibly due to the higher content of MCC that retard the intermolecular interaction which hasten the formations of aggregates and heterogeneous film [16].

Figure 2 also shows that more glycerol caused decreases in bioplastic tensile strength. The high tensile strength values can be attributed to the number of hydrogen bonds between the starch chains that contribute to cohesiveness and low flexibility. When glycerol was incorporated in the starch network, competition for the formation of hydrogen bonding started. As a result, direct interactions between starch chains were partly reduced due to hydrogen bond formation with glycerol, allowing the polymer chains to have more freedom of motion [27]. Abdorreza et al. (2011) reported that the flexibility of the film could be increased by increasing the plasticizer content, but this can lead to plasticizer crystallization in the film [8, 28].

4.5. Elongation at break of bioplastics

The effects of increasing MCC and glycerol content to bioplastic for bioplastic elongation at break are shown in Fig. 3.

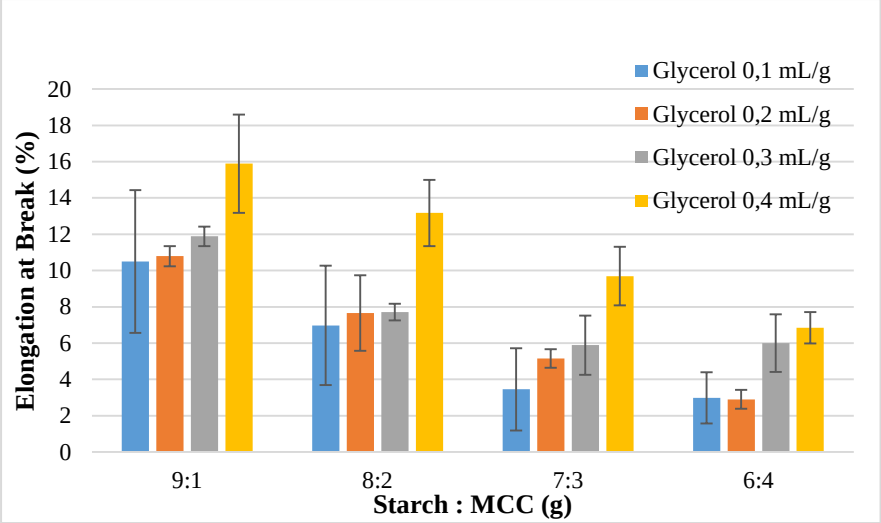

Fig. 3. The effect of MCC from sugar palm fiber and glycerol addition on elongation at break of bioplastics.

Figure 3 shows that elongation at break value decreased with the addition of MCC content. Meanwhile, addition of plasticizers to bioplastics have the opposite effect. Addition MCC fillers from 1 to 4 g caused decrease in elongation at break value from 15,88% to 2,90%. Bioplastic with MCC content 1 g and 0.4 (v/w) glycerol gave the maximum elongation at break value at 15,88%. The best mechanical properties of bioplastic obtained (Table 2) when MCC and glycerol were at 3 g and 0.2 (v/w) which gave 3.16% elongation at break. The desired flexibility of bio packaging films is dependant on their intended application and subsequent packaged food transportation, handling and storage [17].

The increasing elongation in film is due to decrease in plasticizers in the intermolecular bonds between amylose, amylopectin and amylose-amylopectin in the starch matrix and thus substituted by hydrogen bonds formed between plasticizer and starch molecules. Such disruption and reconstruction in starch molecular chains reduce the rigidity and promotes flexibility of film by allowing more chain mobility [17]. Reduction in elongation at break value is as a result of the addition of MCC mass and reduction of glycerol roles as plasticizer. MCC tends to be more interactive with hydrogen and other monomers in bioplastics [8, 29]. These results clearly show that proper amount of MCC in starch solution increases ductility [30].

Table 2. Mechanical properties of bioplastic from avocado seed starch with varying amount of MCC and glycerol

Treatment (starch:MCC)	Tensile strength (MPa)				Elongation at break (%)			
	0.1	0.2	0.3	0.4	0.1	0.2	0.3	0.4
9:1	1.27	1.11	1.07	0.55	10.49	10.78	11.88	15.88
8:2	1.29	1.22	1.17	0.62	6.97	7.65	7.70	13.17
7:3	1.33	2.74	1.27	0.81	3.45	5.15	5.88	9.69
6:4	0.98	0.81	0.81	0.42	2.97	2.90	6.00	6.84

4.6. Functional group analysis

The FTIR spectra of MCC, sugar palm fibers, avocado seeds starch, bioplastic from avocado seeds starch without MCC and bioplastic from avocado seeds starch with MCC and glycerol are shown in Fig. 4.

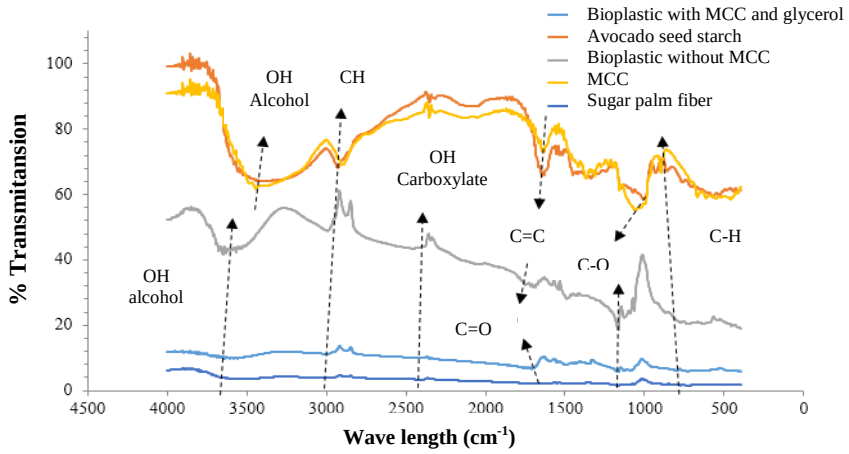

Fig. 4. The FTIR analysis results of avocado seeds starch, MCC, sugar palm fiber, bioplastic from avocado seeds starch without MCC, and bioplastic from avocado seeds starch with MCC and glycerol.

FT-IR analysis for avocado seed starch shows O-H group bonded with hydrogen, C-H alkanes, C=O and C-O ethers [8]. They represented the composition of avocado seed starch, amylose-amylopectin and reducing glucose $(C_6H_{10}O_5)n$ [31]. This result is the same as Ginting's and Tarigan's (2015) functional group analysis of avocado seed starch [21].

The FTIR analysis results for MCC and sugar palm fibers give C-O stretch peak as part of hemicellulose, pectin and lignin compound in 1300-1000 cm^{-1} wavenumber and 1500-1400 cm^{-1} peaks are related to C-C stretch in lignin ring [13]. At 1489 cm^{-1} absorption peak indicates the existence of lignin ring in the fibers. However, MCC FTIR analysis results does not show the peak with wavenumber that refers to the existence of lignin. According to Dufresne, et al (1997) in Lubis (2016) the loss of absorption peaks at wavenumber in the range of 1500-1400 cm^{-1} shows that the existence of lignin and hemicellulose has been terminated properly due to delignification process with acid and bleaching treatment that have removed lignin and hemicellulose from lignocellulosic material [32, 33].

From Fig. 4 can also be seen the comparison of bioplastic without MCC and bioplastic with MCC and glycerol have the same functional groups with no new cluster formed. But, there is an increase in wavenumber for O-H functional groups for starch and MCC, from 3394.72 cm^{-1} to 3603.03 cm^{-1}, in bioplastics from 3437.15 cm^{-1} to 3603.03 cm^{-1} [8]. The increase in O-H group value is due to hydrogen interaction when starch and MCC were combined in bioplastics,

hydrogen bonds are in between amylose-amylose, amylose-amylopectin, MCC-MCC and amylose-MCC-amylopectin chains [8, 34].

4.7. Morphological analysis using scanning electron microscope (SEM)

SEM micrographs of fractures bioplastics with MCC filler and glycerol as plasticizer also MCC derived from sugar palm fibersin 5000x magnification are shown in Fig. 5.

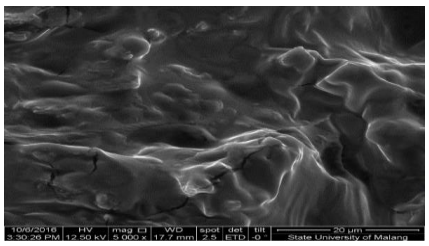

Fig. 5(a). Fractures Bioplastics with Filler MCC and Plasticizer Glycerol Using a Magnification of 5000x.

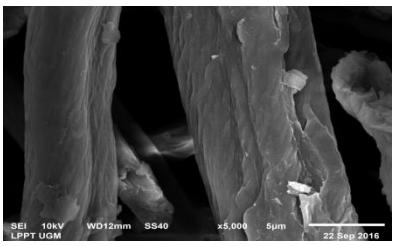

Fig. 5(b). MCC from Sugar Palm Fiber Using a Magnification of 5000x.

It can be seen from Fig. 5(a) that bioplastic film has jagged surface and less compact structure as void presence was found in the film. It was caused by microcrystalline cellulose which did not dissolved completely in NaOH 5% [8]. Wang et al (2008) reported that solubility of microcrystalline cellulose in NaOH is only about 25-30% [35]. Less compact structure from these fibers resulting in more water absorption. The figure also shows less smooth surfaces with cavities as a result of non-homogenous film [29].

Figure 5(b) shows MCC derived from sugar palm fibers are rod shaped with average diameter of 5.55-9.44 µm [8]. It is the same as MCC obtained from sisal with needle or rod shapes [36, 37]. Shapes of particle should be a critical determinant to determine density. It also reflected porosity where particles with larger size have lower porosity and spesific surface area [8, 38]. The particle size of this MCC had a bigger particle size when compared to other non-wood MCC sources, such as rice straw and cotton stalks [39].

5. Conclusions

Microcrystalline cellulose can be isolated from sugar palm fiber through alkali treatment and acid hydrolysis. Rod-shaped microcrystalline cellulose with an average diameter of 5.55-9.44 µm, crystallinity of 97.5% and crystallite size of 25.08 nm was obtained from sugar palm fiber. Starch-based bioplastic films can be prepared by polymers solution casting with isolated MCC as reinforcing filler. The best bioplastics obtained is at mass starch : MCC = 7 : 3 ratio with 0.2 (v/w) glycerol giving 2.74 MPa tensile strength and 3.16% elongation at break.

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