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CHARACTERIZATION AND TENSILE PROPERTIES OF POLYLACTIC ACID BIOCOMPOSITE FILLED MICROCRYSTALLINE CELLULOSE EXTRACTED FROM KENAF

(Ciri-Ciri Dan Sifat Tensil Poli(Asid Laktik) Biokomposit yang Diisi Selulosa Mikrohablur yang Diekstrak daripada Kenaf)

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Abstract

Microcrystalline cellulose (MCC) was successfully extracted from kenaf bast fiber plant (*Hibiscus cannabinus* L.) and incorporated into PLA biocomposite. The kenaf bast fiber undergone alkali treatment and bleaching prior to acid hydrolysis using 1M HNO_3 in obtaining MCC. Several characterizations on isolated MCC and PLA/MCC biocomposite conducted such as Fourier-transform infrared (FT-IR) spectroscopy and crystallinity index. Further characterization was made on the PLA/MCC biocomposite to study its mechanical and physical properties. FT-IR spectral indicated the successive elimination of non-cellulosic constituents in MCC. Differential scanning calorimetry (DSC) analysis revealed the percentage of crystalline region in the MCC obtained from kenaf bast fiber is higher at 88.53, making it suitable to be used as reinforcement filler in PLA biocomposite. Whilst tensile testing on various loading of PLA/MCC composites showed increment in tensile strength and elastic modulus but decrement in percent elongation. The optimum parameters were found at 9% MCC loading at 18.7 MPa in tensile strength attributed to well mixing of PLA and MCC and 88.53 crystallinity index. The potential application of PLA/MCC biocomposite to be used as food packaging.

Keywords: biocomposite, kenaf, microcrystalline cellulose, polylactic acid

Abstrak

Selulosa mikrohablur (MCC) berjaya diekstrak daripada tumbuhan gentian kulit kenaf (*Hibiscus cannabinus* L.) dan dimasukkan ke dalam biokomposit PLA. Gentian kulit kenaf menjalani rawatan alkali dan pelunturan sebelum dihidrolisis asid menggunakan

1M HNO_3 dalam mendapatkan MCC. Beberapa pencirian pada biokomposit MCC dan PLA/MCC terpencil dijalankan seperti Fourier-transform infrared (FT-IR) dan indeks kehabluran. Pencirian lanjut dibuat pada biokomposit PLA/MCC untuk mengkaji sifat mekanikal dan fizikalnya. Spektrum FT-IR menunjukkan penghapusan berturut-turut juzuk bukan selulosa dalam MCC. Analisis kalorimetri pengimbasan pembezaan (DSC) menunjukkan peratusan kawasan kristal dalam MCC yang diperoleh daripada gentian kulit kenaf adalah lebih tinggi iaitu 88.53, menjadikannya sesuai digunakan sebagai pengisi tetulang dalam biokomposit PLA. Manakala ujian tegangan ke atas pelbagai pemuatan komposit PLA/MCC menunjukkan peningkatan dalam kekuatan tegangan dan modulus keanjalan tetapi penyusutan dalam peratus pemanjangan. Parameter optimum didapati pada 9% pemuatan MCC pada 18.7 MPa dalam kekuatan tegangan yang dikaitkan dengan pencampuran telaga PLA dan MCC dan indeks sistaliniti 88.53. Potensi penggunaan biokomposit PLA adalah untuk digunakan dalam pembungkusan makanan.

Kata kunci: biokomposit, kenaf, selulosa mikrohablur, poli(asid laktik)

Introduction

Poly lactic acid (PLA) is biodegradable and renewable thermoplastic polyesters and one of commercially developed polyester from natural source [1]. It is eco-friendly due to its compostable, biodegradable and consumes carbon dioxide during the production. These advantages make PLA an alternative for other petrochemical-based polymers which mostly non-biodegradable. However, there are many limitations of PLA such as low thermal stability, low impact strength, brittleness and poor elongation. Hence, it is only applicable to products that do not require high performance such as disposable cups, cutlery, plastic bags and food packaging materials [1, 2]. Due to these limitations, many research and studies were done to improve both mechanical and physical properties of composite. Extensive study on the blending between PLA and other biodegradable polymer with reinforcing fillers either natural or even manmade has been carried out. Nano clay, natural fibers, cellulose whiskers and Microcrystalline cellulose (MCC) are among reinforcing filler used in composite preparation. Rice husk, oil palm biomass, kenaf and rice straw are among fibers used as reinforcing filler in composite preparation [2, 3].

MCC is a refined wood pulp which derived from cellulose which appears as inert white crystalline powder with no odor and also tasteless [1]. It is generally applied in composite, pharmaceutical, cosmetics, and food industries. Properties of MCC such as its high crystallinity, stiffness, water insolubility, renewability and biodegradability make them an excellent component for various manufacturing purpose. MCC commonly can be obtained from various woody plants and cotton. However, other new potential sources of MCC such as from kenaf, rice husk, oil palm biomass, rice straw and other plant wastes were investigated since the common woody plants and cotton sources make the production costly. It is believed the lignocelluloses contain cellulose (40-55%), hemicellulose (25-50%) and lignin (10-40%) suitable in obtaining the MCC [4]. Enzymatic, mineral acid or microorganism's hydrolysis are among methods used to isolate MCC. Nevertheless, due to high-cost method and lower crystallinity production of MCC using enzymatic hydrolysis becomes less preferable [5]. Previous researchers found that MCC extracted from various sources through acid hydrolysis process has potential to be used in biocomposite preparation [6].

Kenaf or scientifically known as Hibiscus cannabinus L [7] is a common wild plant found in tropical and subtropical countries such as in Asia and Africa. Kenaf is preferred for industrial purposes as an abundantly available renewable annual fiber crop. It is mainly used as high protein animal feed, building materials, in textiles, fuel and also in the composite preparation [7]. This lignocellulosic fiber plant made up of hemicelluloses, lignin and pectin which help cellulose fibrils to be attached together. Presence of both long outer bast (about 25-40%) that provides high quality pulp and inner short core fiber (about 60-75%) of low-quality pulp makes a unique property in kenaf. Research done by Abdul Khalil et al., [8] found that kenaf core fiber contain higher lignin and holocellulose whilst its bast fibers have higher in α-cellulose and ash content. Recently, many researches were done on extraction of microcrystalline cellulose and cellulose nanocrystals from kenaf bast fibers. The potential end usage of biocomposite obtained is in food packaging since it is

biodegradable and food grade PLA. Many biodegradable polymers such as starch, chitosan, polylactic acid (PLA), and chitin commonly used in food packaging. This is due to their low-cost, flexible and see-through properties [8, 9].

Therefore, this study is focusing on the incorporation of MCC extracted from kenaf fiber in PLA biocomposite since less information recorded on usage of kenaf as one of MCC resources. MCC was characterized to determine its morphology using Scanning Electron Microscope (SEM), determination of degree of crystallinity using Differential Scanning Calorimetry (DSC) and Fourier Transform Infrared (FTIR) to identify the molecular bonds exists. The PLA/MCC (kenaf) biocomposite produced undergone characterization using FTIR and tensile mechanical test to determine its tensile strength, elongation at break, and elastic modulus.

Materials and Methods

PLA was obtained from Toray Malaysia while kenaf was collected from Lembaga Kenaf Malaysia in Kelantan. Analytical grade of sodium hydroxide, NaOH (QRec, 99%) and Sodium hypochlorite, NaOCl (QRec, 5%) was used for alkali treatment and bleaching agents respectively whereas 1M HNO_3 was used in acid hydrolysis process. Chloroform was used to dissolve the PLA for solvent casting method.

Extraction of Microcrystalline Cellulose from Kenaf

10 g of dried kenaf was refluxed with 300 mL of NaOH at 80°C for 1 hour and 30 minutes. After alkali treatment, the kenaf pulp was refluxed at 30°C for 18 minutes using 350 mL of 5% sodium hypochlorite (NaOCl). Again, the solution was refluxed in 83 mL of 1M HNO_3 at 80°C for 30 minutes via mechanical stirring. Then the obtained mixture was filtered, washed several times with distilled water and dried at 105°C in oven until constant weight was achieved [2].

Characterization of MCC (Kenaf)

Functional group that presents in MCC was identified using Perkin Elmer FTIR spectrometer 1650 within wave number range of 4000 cm^{-1} to 400 cm^{-1}. As for DSC measurements, approximately 8-10 mg of MCC sample was run through Perkin Elmer Instruments Pyris DSC to determine its crystalline percentage using rate 5 min at 5°C and heated at rate of 10°C/min until 250°C The percent of crystalline region in the MCC was calculated using Equation 1. T_m was determined from the second scan cycle on the melting peak temperature. Scanning Electron Microscopy (SEM) was conducted via Hitachi Regulus 8220 in Universiti Sains Malaysia to analyse the morphological properties of the selected samples.

$$\text{Percent crystallinity, } X_c = (\Delta Hm \text{ (Treated)})/((\Delta H°m \text{ (Untreated)}) \times 100\% \quad (1)$$

Where, ΔH_m is the MCC obtained after acid hydrolysis (treated kenaf) while $\Delta H°_m$ is the untreated kenaf.

Preparation of PLA/MCC composite

Ten g of PLA was added into 100 mL chloroform to obtain 10% w/v solution in water bath at 60 °C for 2 hours until PLA was completely dissolved. Different amount of MCC (0, 1, 3, 5, 7 and 9 wt.%) was added into 10% w/v solution of PLA and sonicated for 5 minutes to ensure homogenous mixing. Then, casted on glass plates with dimension of 16 cm x 13 cm x 0.5 cm and left to dry at room temperature in 24 hours for complete removal of chloroform.

Characterization and mechanical testing of PLA/MCC biocomposite

The functional groups present in PLA/MCC was identified using Perkin Elmer FTIR spectrometer 1650 within wave number range of 4000 cm^{-1} to 400 cm^{-1}. Tensile testing was carried out using ASTM D882 type V standard test method using Instron Tensile Tester with 5 replicates for each formulation.

Water absorption test for PLA/MCC biocomposite.

ASTM D570 standard test method was used to determine the water absorption properties of PLA/MCC biocomposite. Five replicates of each composite with dimension of 1 cm x 3 cm were made using Equation 2.

Percent water absorption = ([Wet weight - Dry weight])/ (Dry weight) x 100 (2)

Results and Discussion

Fourier transform infrared (FTIR) analysis on isolated MCC

The FTIR spectral obtained were shown in Figure 1. The broad absorption band in at 3409 and 3407 cm^{-1} for untreated kenaf and MCC respectively indicate the presence of –OH group. The –OH groups of untreated kenaf slightly shifted toward lower wavelength after the treatment. This is probably due to weakening of hydrogen bonding in cellulose during acid hydrolysis process [10]. Absorption peaks at 2922 cm^{-1} for untreated kenaf and 2896 cm^{-1} for the MCC is related to CH_2 stretching frequency. The peaks that appeared due to water absorption between 1637 until 1654 cm^{-1} indicates the water presence in the sample [10]. Previous research by Rosa et al. [10] stated that bending modes of water molecules arise due to great interaction between water and cellulose molecules. On the other hand, peaks that appeared within 1035 to 1112 cm^{-1} are due to C-O stretching vibration within the ring of anhydroglucose in cellulose [11, 12]. The absorption peak presents at 1739 cm^{-1} for untreated kenaf is in line with Abdul Khalil et al. [8] and approximate to 1735cm^{-1} [13]. This signifies the presence of C=O stretching vibration of carbonyl group and acetyl group in the hemicelluloses [8]. Whilst for MCC, the absent peak for the C=O stretching indicates a complete removal of hemicelluloses during alkali treatment. The peak at 1424 cm^{-1} in untreated kenaf indicates the presence of lignin (1509-1609 cm^{-1}) whereas it is absent from MCC spectrum proves that lignin had been completely extracted out of the MCC [10].

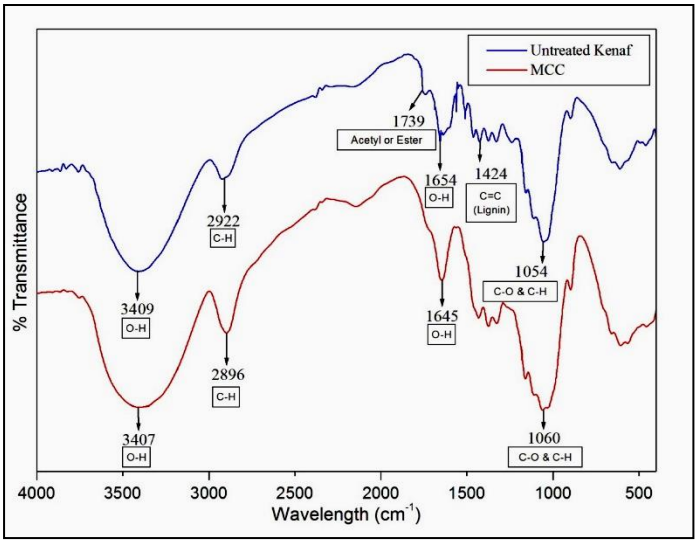

Figure 1. FT-IR spectroscopy for untreated kenaf and MCC from kenaf

FT-IR analysis was performed on pure PLA and PLA/MCC biocomposite in order to determine their interaction and phase behavior. Figure 2 shows the typical FT-IR spectra obtained. The characteristics peak for pure PLA is shown in Figure 2(a). The –OH stretching vibration is apparent at 3316-3843 cm^{-1}while the band observed in between 2850-3000 cm^{-1} is the asymmetric stretching of C-H. The peaks that attributes to C=O stretching vibration of carbonyl appears at 1630-1820 cm^{-1}. Peak that appears in between 1450-1475 cm^{-1} resulted from C-H bending of the -CH_3 group. The C-C bonding of alkane appears at 820-871 cm^{-1} almost similar with peak at 871 cm^{-1} by Haafiz *et* al. [9].

Whilst, PLA/MCC spectra for 1, 3, 5, 7 and 9 wt. % MCC is shown in the same Figure 2. From analysis, there is no new peak present upon gradual addition of MCC into PLA matrix. Haafiz et al. [9] suggest that the

interaction between PLA matrix and MCC most probably involve only physical interaction instead of chemical. Relatively small peaks of pure PLA for –OH stretching and bending that appear at 3200-3400 cm^{-1} gradually absent upon increasing MCC content. Identical result was described by Haafiz et al. [9] in PLA/MCC biocomposite extracted from rice husk.

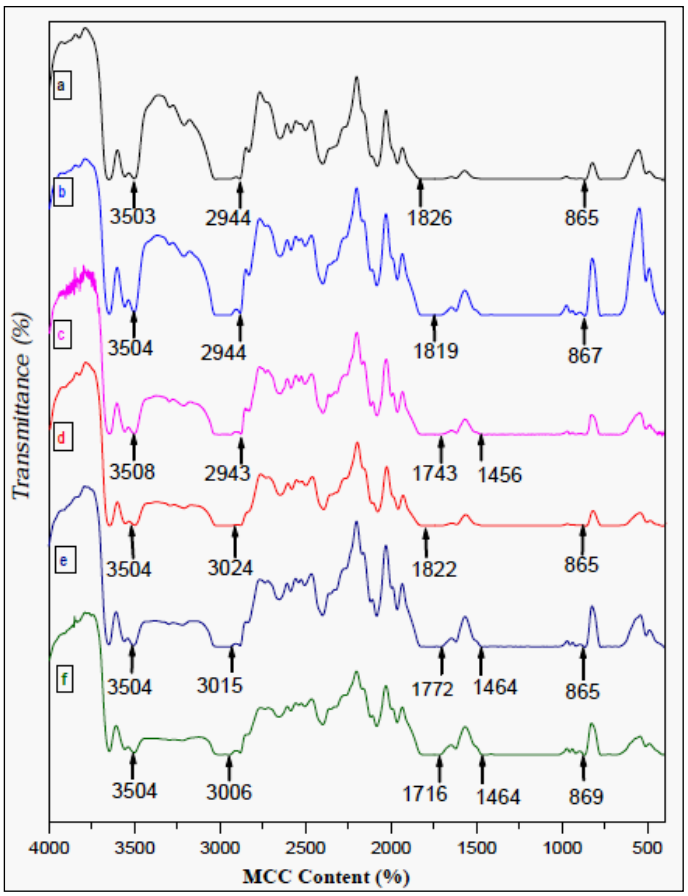

Figure 2. FT-IR spectroscopy for (a) PLA and PLA/MCC composite with different MCC loadings ((b) 1%, (c) 3%, (d) 5%, (e) 7% and (f) 9%)

Differential scanning calorimetry analysis on isolated MCC

Table 1 indicates some thermal characteristics obtained for kenaf, PLA, PLA/MCC. It is shown that melting enthalpies (ΔH_m) obtained for both untreated kenaf bast fiber and MCC extracted from kenaf is 492.38 and 435.89 J/g respectively. The T_m for PLA/MCC composite of 1, 3, 5, 7 and 9 wt. % MCC is in between 152-153°C which is not very different with T_m for pure PLA (152.9°C) due to MCC having nucleating effect on PLA [2]. The degradation of PLA composite lowers its melting point since the length of molecular chains is reduced [14]. It is believed the addition of chemical additives to enhance the PLA for other purpose especially in manufacturing industries decreased the melting point. The percent crystallinity was measured according to formula in Equation. 1. The percentage for crystalline region in MCC calculated is 88.53%. The effect of higher crystallinity in MCC obtained is manifested in increment of tensile and young modulus properties. This indicated physical crosslinking taken place and hindered the mobility of polymer matrix. It is believed highly crystalline MCC acted as nucleating agents and restricted the mobility of the polymer chains

by arranging the crystal lattice structure [1]. The results are in good agreement with SEM images Figure 3(b) showing rough surfaces found at 9 wt.% of PLA/MCC. This is in agreement with Nur Aimi et al. reported that higher crystallinity of the fiber improves the mechanical properties of the composites [15].

Table 1. DSC Characteristic of kenaf, MCC, PLA and PLA/MCC biocomposite

Characteristics	Kenaf	MCC	PLA	PLA/MCC (9%)
Melting Temperature T_m(0c)	80.38	74.83	168	153
Glass transition Temperature T_g(0c)	45	55	58	63

Scanning electron microscope image analysis

Fracture surfaces of pure PLA and PLA/MCC composite for 9 wt.% were analyzed using SEM to investigate the morphology of microcrystalline cellulose embedded in the PLA matrix. Smooth fracture surface can be observed on the pure PLA as shown in Figure 3(a) which can be categorized as ductile fracture due to amorphous property of PLA itself. Whilst, uneven fracture shown for both composite filled MCC (Figure 3b). Smooth fracture surface can be observed on the pure PLA as shown in Figure 3a. Mukherjee et al. [16] suggested that uneven fracture of composites possibly take place after the addition of MCC cause a significant deformation on PLA matrix as compared. It is also indicating that more force is needed to fail the samples during testing.

Through observation, the diameter of 5 and 9 wt.% MCC content measured is approximately 25.55 μm and 17.69 μm respectively (Figure 3(c) and (d)) and there is no significant filler agglomeration observed. This is the evidence of good interaction between MCC and the PLA matrix that helps to reinforce the composite as it manifested in tensile strength and modulus. In addition, only a little irregularity in the MCC shape found in both samples which is a good feature for filler. According to Yussuf et al. [17], the irregularities shape of filler in polymer matrix lowers the mechanical strength of the composite because it reduces the ability of filler to transfer the applied stress from the polymer matrix [16]. Since the size of MCC is small enough to lower the irregularities shape in the composite, mechanical properties of the PLA/MCC composite are enhanced.

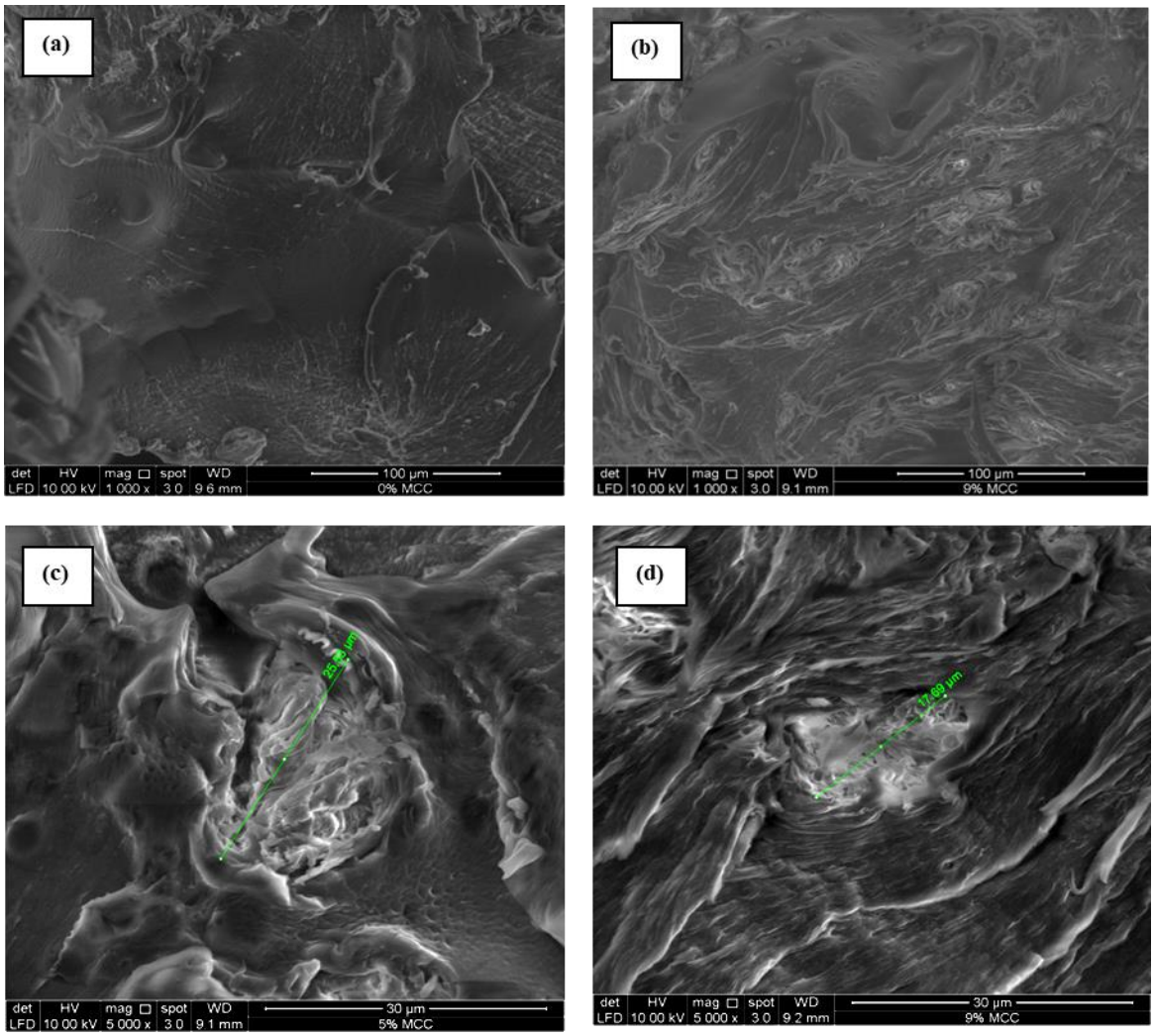

Figure 3. (a) Scanning electron micrographs of fractured surface of pure PLA at 1000, (b) 9 wt.% PLA/MCC at 1000x, (c) 5 wt.% PLA/MCC biocomposite at 5000x, and (d) 9 wt.% PLA/MCC biocomposite at 5000x

Tensile properties of PLA/MCC (kenaf) biocomposite

The tensile strength of PLA/MCC blends is higher than that of pure PLA as shown in Figure 4. For 1 and 3 wt. % composites, slight increment of tensile strength can be observed compared to the pure PLA. Whereas, at 5 to 9 wt.% MCC contents in PLA composite shows higher tensile strength which is 1.2 to 1.4-fold greater than the pure PLA due to good dispersion between MCC and PLA resulting in good interaction between them. Previous study by Haafiz et. al. indicates that MCC have higher tendency to agglomerate which makes it interact and distributed poorly in PLA matrix [9]. As proven by analysis under SEM, no significant agglomeration observed in all the composites thus it can be suggested that the interaction and distribution of MCC in the PLA matrix is good. Furthermore, it can be seen from the SEM fractography there is good interaction between PLA and MCC. This is because of the MCC possess a high aspect ratio giving way to better interaction and finally increased the tensile strength. Figure 5 illustrate the result for elastic modulus. PLA/MCC blends of 1, 3 and 5 wt.% MCC in PLA show improvement compared to the pure PLA with order of 217.94, 226.83 and 236.52 MPa correspondingly. Nevertheless, as MCC contents increased more than 5 wt.%, the elastic modulus

maintained at average value approximately at 237 MPa. According to Haafiz et al. the increasing of MCC loading may be attributed by common filler/polymer features such as increasing rigidity effect, hydrogen bonding and also the percentage of crystalline region in the filler [9].

On contrary, PLA/MCC blend shows a gradual reduction for percent elongation when more MCC added as shown in Figure 6. The values then retained between 39 to 40% elongation for 5, 7 and 9 wt.% MCC loading with average of 3.2-fold lower than pure PLA. This might due to properties of MCC itself which possess strong hydrophilic properties compared to PLA that slightly hydrophilic cause poor interfacial adhesion between them [14]. Stiffening action by MCC may cause the restriction on chain movement of PLA molecules during testing [17]. In contrast, PLA/MCC blend shows a gradual reduction for percent elongation when more MCC added as shown in Figure 6. The values then retained between 39 to 40% elongation for 5, 7 and 9 wt.% MCC loading with average of 3.2-fold lower than pure PLA. This might due to properties of MCC itself which possess strong hydrophilic properties compared to PLA that slightly hydrophilic cause poor interfacial adhesion between them [12]. Stiffening action by MCC may cause the restriction on chain movement of PLA molecules during testing [15].

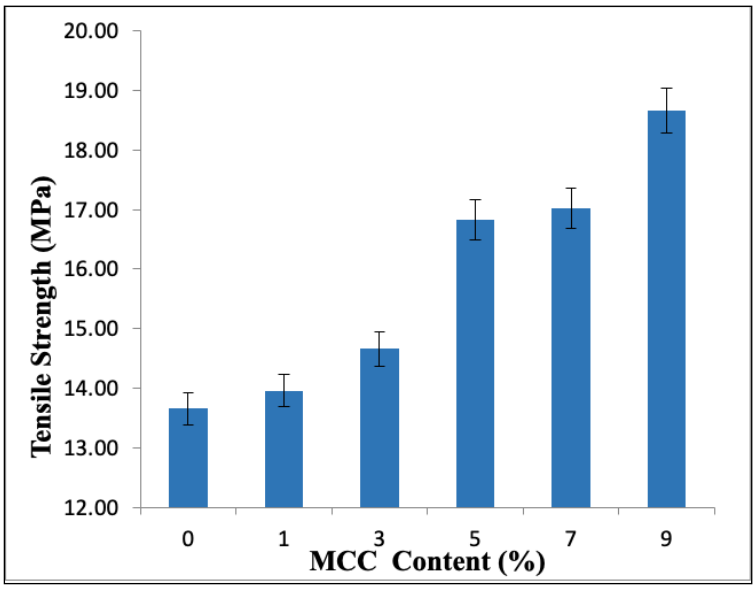

Figure 4. The effect of MCC content on the Tensile strength of PLA biocomposite

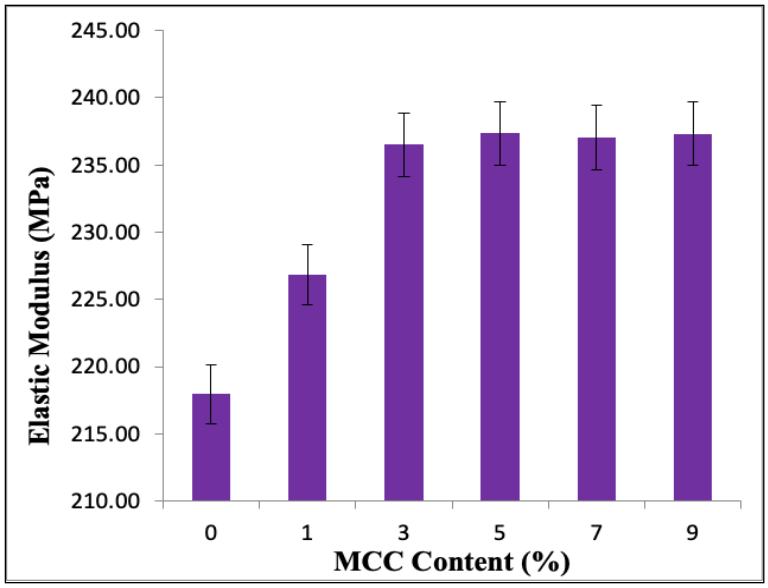

Figure 5. The effect of MCC content on the tensile modulus of PLA biocomposite

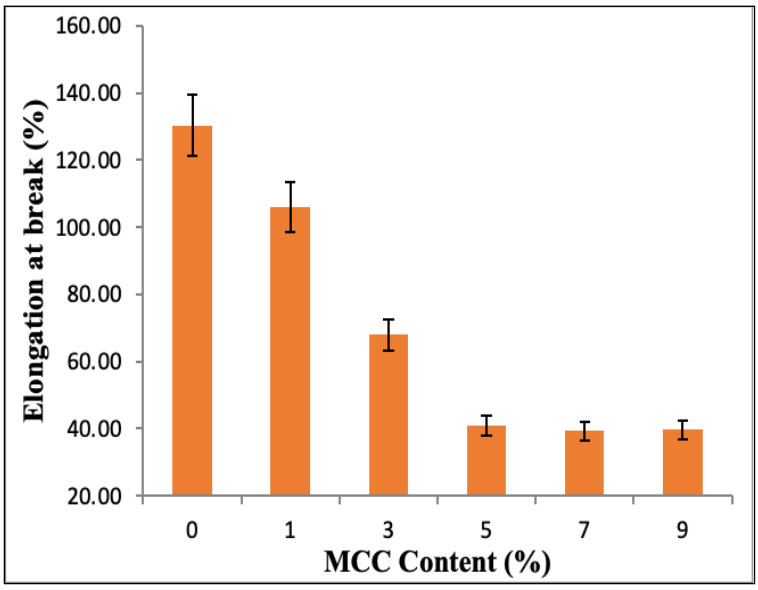

Figure 6. The effect of MCC content on the elongation at break of PLA biocomposite

Water immersion testing

The percentage of water absorption and thickness swelling of PLA/MCC composites increases upon MCC addition as shown in Figure 7 and 8. However, the water absorption does not linearly increase and might due to dispersion of MCC in the matrix not good enough for some of the composite. For an example the percentage of water absorption for 1% MCC is closer to pure PLA compared to 3 wt. % MCC. Based on the result obtained, it shows significant increment in the water absorption and thickness swelling with the increasing of MCC content after 1 % loading. It gives a better picture that

addition of MCC in the composite will help to boost the degradation of PLA since water is one of the important aspects in hydrolysis process as mentioned by Rocca-Smith et al. [18]. The process is the hydrolysis induced by water penetration and diffusion is where the penetration water into the PLA is driving force the biodegradability of the PLA [19]. From the result obtained also show that pure PLA have low water absorption capability. So that beside the mechanical improvement, addition of MCC into the PLA also can contributed to the degradability of the material which significantly important aspect as bio-based thermoplastic.

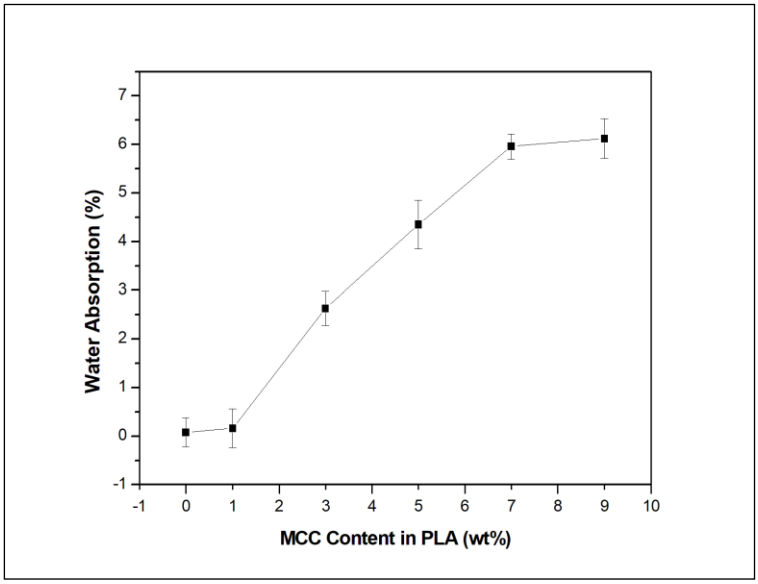

Figure 7. Water absorption of various MMC PLA biocomposite

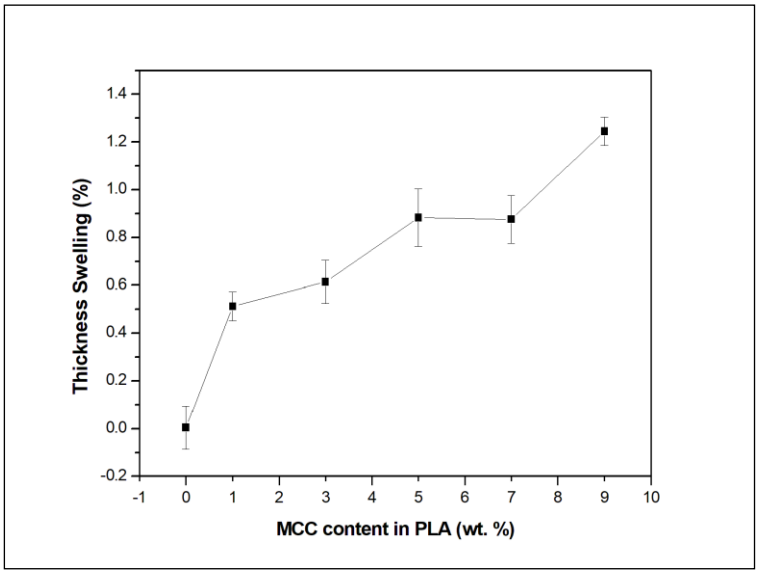

Figure 8. Thickness Swelling of Various MMC PLA Biocomposite

Conclusion

The finding from the research shows that MCC from kenaf bast fiber has been successfully isolated and characterized. The uniqueness of the research is the combination of MCC isolated from kenaf and PLA in biocomposite preparation since it possesses the biodegradability and renewability properties. In addition, Cellulose was purified using general chemical treatments which are alkali treatment and bleaching. The alkali treatment was used to remove hemicelluloses whilst bleaching was done for delignification purpose. Extraction of MCC from the treated kenaf fiber was done via acid hydrolysis 1M HNO_3. Characterization using FT-IR spectrum obtained for both kenaf and MCC proved a successful elimination of hemicelluloses and lignin from MCC. The analysis under DSC shows that percent crystallinity of MCC is high which validated that acid hydrolysis does not alter the crystal structure of cellulose in the kenaf fiber. The isolation of MCC from kenaf by means of acid hydrolysis is sufficient to produce high crystalline MCC. PLA/MCC biocomposite was successfully fabricated with addition of 1, 3, 5, 7, and 9 wt.% MCC through solvent casting technique. FTIR spectra of showed insignificant changes in the peak position of PLA/MCC biocomposite suggest that PLA matrix and MCC only physically interacted. Mechanical properties of PLA/MCC biocomposite proved an increasing tensile strength and elastic modulus due to stiffening action of MCC upon increasing MCC content. Conversely, elongation at break decreased when MCC content increased. This caused by brittleness property of crystalline region in MCC causing subsequent restriction on the biocomposite chain movement. Observation under SEM revealed a good dispersion of MCC in PLA matrix which gave reason for the increasing in tensile strength of the biocomposite. Physical test was done upon PLA/MCC to determine the water absorption properties of the biocomposite. Upon addition of MCC, water absorption of the composite showed increment as MCC content added due to hydrophilic properties of MCC which important for degradability of PLA. Thus, it can be concluded that addition of 1 to 9 wt. % MCC into PLA matrix gave significant effect.

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