Preparation and Characterization of Durian Husk Fiber Filled Polylactic Acid Biocomposites

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Abstract. Polylactic acid (PLA) is biodegradable thermoplastic that made from renewable raw material, but its high cost limited the application. Thus, addition of natural fiber can be effectively reduced the cost of PLA. This research is utilised natural fiber extracted from durian husk to made PLA biocomposites. This paper is focus on the effect of fiber content on tensile and thermal properties of PLA/durian husk fiber (DHF) biocomposites. The results found that the tensile strength and modulus of this biocomposites increased with increase of fiber content, but the strength still lower compared to neat PLA. Then, the elongation at break of biocomposites was expected decreased at higher fiber content. The PLA/DHF biocomposites with 60 phr fiber content exhibited tensile strength of 11 MPa, but it is too brittle yet for any application. The addition of DHF caused an early thermal degradation on PLA biocomposites. Then, the thermal stability of PLA biocomposites was decreased with more fiber content.

1 Introduction

In 2015, researchers found 8.3 billion metric tons of plastics waste were produced globally and 6.3 billion tons of this plastic products are turning into waste. As well known, not all the plastic can be recycle, thus only 9% of that waste total was recycled, 12% was used for energy recover and left were sent to landfills. The amount of plastics waste for land fill are expected to achieve 12 billion metric tons, if the current trends is keep going on [1]. Plastic material made from petroleum resources are chemically stable which hardly to biodegrade in environment. In present, part of the plastic product has replace by bioplastic that able to biodegradable after disposal. Polylactic acid (PLA) is a type of bioplastic that synthesized from natural material, such as corn starch, tapioca root and sugarcane [2-3]. Besides, PLA offers a biodegradable properties and also good mechanical strength, non-toxic and sustainable since it made from renewable resources [4-5]. Nowadays, the production of bioplastic over the world is about 750,000 tons per year. Unfortunately, this

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amount still very small compare to conventional plastic that produced 200 million tons per year. The main reason why PLA is seldom choose by industrial as their main choice of material, because the price of PLA resin is more expensive than conventional plastic [5-6].

In order to promote the usage of PLA material, natural filler can be introduced to PLA and make it into a biocomposites. The natural filler can be easily obtained from any agricultural crop and waste, for instance, corn cob [7], cocoa pod [8], coconut shell [3] and palm kernel shell [9]. The main benefit of natural filler very low cost, biodegradable, less abrasive to machine, and inexhaustible resources [10-11]. Thus, by making the PLA in to biocomposite, the addition of natural filler can be partially replace the PLA without influences its biodegradability and highly reduced the cost of final product. *Durio zibethinus Murray*, also called as durian, the famous fruit among the Southest Asia countries especially in Malaysia, Thailand, Indonesia and Philippine [12]. The durian fruit is made up of 40% of flesh and 60% of husk. Usually, the durian husk was discarded and end up on landfills or burnt which poses an environmental issues. In Malaysia, about 3,800 metric tons of durian fruits are produced in year 2016. Assuming every 60% of total amount produced durian fruits is husk, it estimated the durian husks waste produced in year 2016 was achieved 2,280 metric tons. The production of durian fruits are predicted to achieve 22,000 metric tons at year 2020 [13]. This meaning the amount durian husks waste will be increased. The durian husk consists 60.45% of cellulose, 15.45% of lignin and 13.09% of hemicellulose, and the content are similar to wood fiber [14]. For this reason, this research is underway to utilize the fiber obtained from durian husk and combined with PLA to produce biocomposites.

This present research is focus on the effect of fiber content on the tensile and thermal properties of durian husk fiber filled polylactic acid biocomposites.

2 Methodology

2.1 Research Materials

PLA used in this experiment was supplied by TT Biotechnologies Sdn. Bhd. (Malaysia) and durian husk was collected from durian fruit stall at SS2, Petaling Jaya (Selangor).

2.2 Preparation of Durian Husk Fiber

The durian husks obtained from durian fruit stall were washed with tap water and cut into small pieces. Then, the durian husks were dried using circulated air oven at 70^{o}C until completely dried. The dried durian husks were ground into short fiber using mechanical grinder. Next, the short durian husk fiber (DHF) was passed through a sieve with mesh size of 600 micron to get the homogenous size of fibers. The DHF further dried using oven before compounding process.

2.3 Preparation of PLA/DHF Biocomposites

The PLA/DHF biocomposites were compounded using Haake Rheomix 600p at SIRIM Malaysia (Shah Alam). The biocomposites were prepared with fiber various from 15, 30, 45, and 60 phr (part per hundred resin). The processing temperature was fixed at 180^{o}C and rotor speed of 80 rpm. The compounding procedures included: i) addition of PLA resin in

to mixing chamfer for 1 minute to fully melt the PLA resin; ii) Then, the DHF was added into melted PLA and compounded for 5 minutes; iii) Last, the compound was removed from mixing chamber.

The PLA/DHF biocomposites were further molded into thin sheet with thickness of 1 mm. The hotpress machine (model Moore) was used in this molding process and temperature was set at 180^{o}C. The molding procedures included: i) preheated to soften the compound for 2 min; ii) fully compressed the softened compound under 100 MPa pressure for 1 min; and iii) Last, cooled specimen under same pressure for 20 minutes. All the biocomposite sheets were cut into tensile specimen and dimension following ASTM D638.

2.4 Testing and Characterization

Tensile test was performance using Instron Universal Testing Machine (Model 5569). The tensile test was referring to ASTM D638. The cross-head speed of machine was set at 5 mm/min and a 15kN load cell was used. The tensile strength, modulus and elongation at break of the samples were recorded by Bluehill 3 software. A minimum of 7 specimens were tested for every formulated biocomposites.

Thermogravimetric analysis (TGA) was carried out using Pyris Diamond TGA (Perkin-Elmer). The specimen was prepared in small size with weight from 6 to 8 mg and placed in a ceramic pan. The specimen was heated from 30^{o}C to 600^{o}C at hearing rate of 10^{o}C/min. The TGA analysis was run under nitrogen atmosphere with gas flow rate of 20 ml/mm.

3 Results and Discussion

3.1 Tensile Properties

Figure 1 shows the tensile strength of PLA/DHF biocomposites with different DHF content. The tensile strength of PLA/DHF biocomposite exhibits an increasing trend as the DHF content increased. Usually, natural fiber displays a higher mechanical strength compared to most plastic material [15]. Thus, the addition of DHF fiber might share part of the tensile load which subjected PLA matrix and increased the tensile strength of biocomposite. From the previous study found that the tensile strength of neat PLA was about 54 MPa [3]. The biocomposites exhibited lower strength compared to neat PLA. This is because the DHF used in this experiment was short fiber and the reinforcing ability of the fiber is highly depending on the fiber orientation. The short fibers usually is randomly orientated. The presence of fibers that orientated perpendicular to direct of applied load might unable to share the load which caused the strength of biocomposite to decrease. However, at higher fiber content, the presence of more fibers that parallel to direction of load and it will share load that subjected to biocomposite. Thus, the strength of biocomposite was raised when more fiber content was added. Similar findings have also been reported by Yu et al. [16].

The effect of different fiber content on PLA/DHF biocomposites is illustrated in Figure 2. As expected, the tensile modulus of the biocomposites gradually increases with increasing amount of DHF. It is because the modulus of DHF is usually higher than plastic [12]. Therefore, the presence of higher amount of DHF contributed to higher modulus of the PLA/DHF biocomposite. For this reason, the tensile modulus of PLA/DHF biocomposite increases due to addition of DHF. Moreover, Gunti et al. [17] also agreed that

the modulus of biocomposite is highly depending on the amount of filler. Hence, the biocomposite with higher fiber content showed higher tensile modulus.

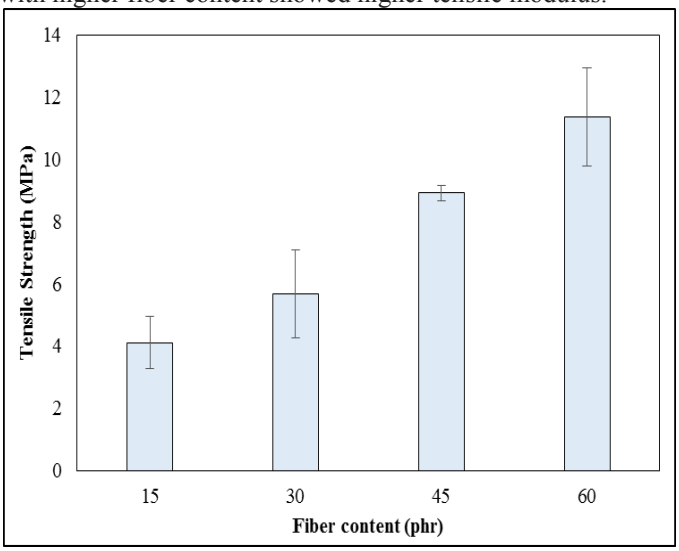

Fig. 1. Tensile strength of PLA/DHF biocomposites with different fiber content.

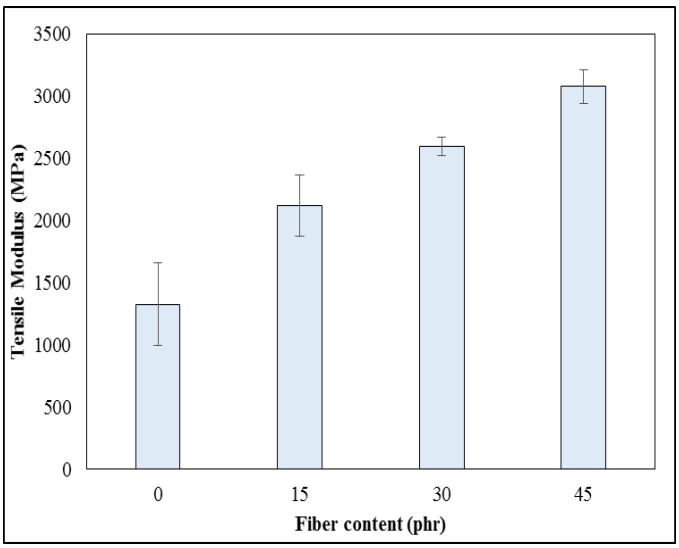

Fig. 2. Tensile modulus of PLA/DHF biocomposites with different fiber content.

Figure 3 displays the elongation at break of the PLA/DHF biocomposite with different DHF content. The result trend of tensile modulus of biocomposite typically opposite with elongation at break. The neat PLA is well known brittle material and its elongation at break was only 4.2% [3]. The addition of DHF had dramatically reduced the elongation at break of PLA. The result found the fiber content increased, the elongation at break of PLA/DHF biocomposite decreased. In general, natural fiber is rigid material that not elongate much [18]. Thus, the addition of DHF would not contribute on the elongation of PLA matrix. In addition, the friction presents between PLA matrix and DHF fiber will significantly reduce the mobility of the polymer chains. For this reason, PLA/DHF biocomposite with more fiber content became more rigid and brittle. This observation is in agreement with findings of many others researcher [19].

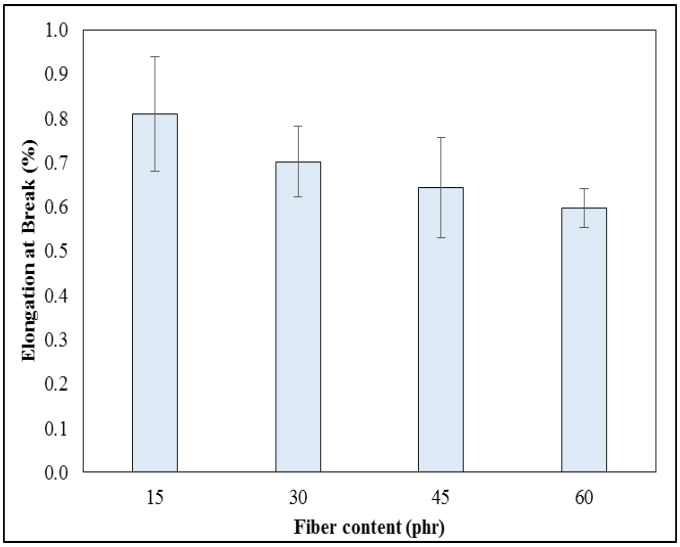

Fig. 3. Elongation at break of PLA/DHF biocomposites with different fiber content.

3.2 Thermal Properties

The TGA curves of DHF, neat PLA, and PLA/DHF biocomposites at selected fiber content is shown in Figure 4. All the data obtained from TGA curves are tabulated in Table 1. The TGA result shows the DHF was thermal degraded earlier than neat PLA and yielded high char residue at 600^{o}C. The earlier thermal degradation of DHF was due to the removing of moisture and volatile compounds that found in DHF. Then, the thermal degradation of DHF was following by degradation of hemicellulose at temperature above 250^{o}C. Furthermore, the thermal degradation of lignin and cellulose from DHF were found beyond 350^{o}C [20]. The thermal degradation of lignin and cellulose were contributed to char formation that caused high amount of char residue. From Table 1, the temperature at 5% ($T_{d5\%}$) and 50 % ($T_{d50\%}$) weight loss of PLA/DHF biocomposites were shifted to lower temperature when fiber content increased from 30 to 60 phr. The PLA/DHF biocomposites also exhibited lower $T_{d5\%}$ and $T_{d50\%}$ compared to neat PLA. This indicated the PLA/DHF biocomposites have lower thermal stability compared to neat PLA. The biocomposite had lower thermal stability was due to components in DHF that decomposed when subjected to high temperature. The char residue of PLA/DHF biocomposites also increased with the increases of fiber content. As mentioned early, the formation of more char residue was due to thermal degradation of lignin and cellulose from DHF. The similar observation also found by other researchers [21-22].

4 Conclusion

The increase of fiber content increased the tensile strength and modulus of PLA/DHF biocomposites, but decrease in elongation at break. The PLA/DHF biocomposites exhibited lower strength compared to neat PLA, but the average tensile strength of PLA/DHF biocomposites with 60 phr of fiber still have 11 MPa. The strength is comparable to low density polyethylene, but its to brittle to make any application. Besides, the addition of DHF had caused an early thermal degradation on PLA/DHF biocomposites and also lower in thermal stability. The TGA results also found the biocomposite with more fiber content exhibited high char residue after decomposed at temperature 600^{o}C.

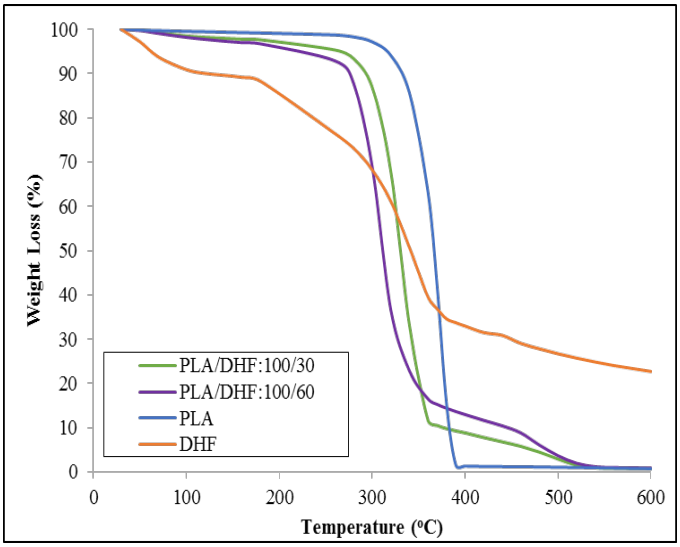

Fig. 4. TGA curves of DHF, neat PLA and PLA/DHF biocomposites at selected fiber content.

Table 1. Formatting sections, subsections and subsubsections.

Samples	Temperature at 5% weight loss (°C)	Temperature at 50% weight loss (°C)	Char residue at 600°C (%)
Neat PLA	313.4	365.1	0.7
DHF	55.5	339.3	22.7
PLA/DHF: 100/30	260.9	331.5	0.8
PLA/DHF: 100/60	232.6	307.7	0.9

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