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THE EFFECT OF FIBER MILLING ON MECHANICAL PROPERTIES OF POLYPROPYLENE REINFORCED OIL PALM MESOCARP FIBER BIOCOMPOSITE

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ABSTRACT

Oil palm mesocarp fiber (OPMF) is biomass residue abundantly available at palm oil mill. Being a lignocellulose, it can be used as reinforce material in biocomposite. In this study, the effect of OPMF milling on the mechanical properties of polypropylene (PP)/OPMF biocomposite was studied. Two types of mills were used, *i.e.* Wiley mill (WM) and disc mill (DM). Ground OPMF from each milling process was examined for its surface morphology, particle size distribution and aspect ratio by microscopic analyses. Ground OPMF was later sieved and fiber with particle size less than 150 μm was separated and mixed with PP using Thermo Haake mixer at 20 and 50 % (w/w) of fiber loading. Results showed that PP/DM-OPMF biocomposite had higher tensile strength compared to PP/WM-OPMF, with almost two-fold. From the SEM and light microscopic analysis, it can be observed that DM-OPMF had smaller diameter of fiber with almost uniform size of particles. Small diameter and uniform size of fiber may improve stress transfer between the fiber and polymer matrix, improve surface contact between polymer matrix and fiber and cause the well-dispersion of filler throughout the polymer. This explains the better tensile strength of PP/DM-OPMF compared to PP/WM-OPMF biocomposite. Overall, it can be concluded that disc milling could serve as a simple and effective grinding method which is able to improve the mechanical properties of biocomposite.

Keywords : *Oil palm mesocarp fiber (OPMF); disc mill; aspect ratio; biocomposites; mechanical properties.*

INTRODUCTION

Grinding is a common unit process which involves a broad range of application to produce fine particles. Such fine particles is required in converting or utilising natural lignocellulosic materials to enhance the accessibility of reactive agents, improve conversion rates and yields [1], and also to strengthen the interaction between different materials due to higher surface area [2]. Many kinds of grinder are available in which resulting different size of particle such as Wiley mill (WM) and disc mill (DM). Different mode of action occurred in these two mills, where WM able to reduce size by cutting mode and DM involves a combination action includes compression, rubbing action, shearing and cutting.

In the development of biocomposite, mechanical properties of a short-fiber reinforced composite depend on several factors such as fiber particle size, aspect ratio and fiber/matrix adhesion. Fiber with high aspect ratio, *i.e.* long fiber with small diameter will give higher surface area and hence, provides larger contact area between the fiber particles and polymer matrix. High aspect ratio fiber will usually binds better to the polymer than a thick, short fiber. Shinoj et al. [3] reported that better interaction between particles and polymer matrix will improve the mechanical properties of the biocomposites.

Oil palm mesocarp fibers (OPMF) strands which have 100 - 500 mm in diameter and elongated with 30 – 50 mm length, consists of bundle of microfibrils, similarly demonstrated by oil palm empty fruit bunch structure. Natural fibers especially for compounding process by internal mixer or extruder are required to

undergo grinding process to allow better wetting to the filler. From literature [4], tensile strength of biocomposite from wheat by-product

s, where wheat straw fiber size of d_{50} 62 μm, were increased from 32 to 41 MPa as percentage of fiber increased from 1.2 to 11 % (vol), respectively. However, typically most of the researchers chose to use fibers with sizes less than 500 μm [5, 6].

Currently, grinders with cutting as breaking mechanism such as mini chipper, cutting blade, hammer mill and wiley mill are favourable as they are relatively inexpensive and easy to be operated. In this study, we used two different types of milling, *i.e.* Wiley mill (WM) and disc mill (DM) in order to demonstrate the effect of grinding with different breaking mechanism to evaluate the physical changes of OPMF in terms of particle size distribution, aspect ratio and morphological changes. Fibers obtained from both milling techniques were then used for biocomposite preparation and the effects of particle size distribution, aspect ratio and morphological surface on the characteristics and performance of biocomposite were clarified.

MATERIALS AND METHODS

Material

Oil palm mesocarp fibers (OPMF) were obtained from FELDA Serting Hilir Palm Oil Mill, Negeri Sembilan, Malaysia. OPMF was disintegrated by manual washing and sorting. The OPMF were then sun dried. Samples were stored in sealed plastic bag at room temperature (±24°C) prior to use. Polypropylene (PP) (Titan Ltd) with density of 0.90 g/cm^3 was used as a binding material. Sulfuric acid (90 % purity) and potassium hydroxide were supplied by Merck, Germany and sodium chlorite was supplied by Acros Organics, USA.

Sample preparation

OPMF samples were ground into small particles using two different grinding machines: WM (Taiwan) and DM (AIST, Japan). WM-OPMF and DM-OPMF were then screened using a vibrating screener prior to the particle size analysis at various mesh sizes of 150, 125, 100 and 75 μm. The sieving process was conducted for about 10 min for each sample. The sieved samples were then weighed. Only fibers passed through mesh size of 150 μm were used as biocomposite filler.

Fabrication of PP/OPMF composites

Prior to blending, OPMF were kept in an oven at 50 °C for 24 h. All the composites (PP and OPMF samples) were premixed followed by blended at 170 °C and 50 rpm rotor speed for 10 min using an internal mixer (Haake Rheomix Polydrive). The composites were prepared by using similar mixing procedure with the addition of 20 and 50 % (w/w) of WM-OPMF and DM-OPMF based on weight ratio.

Analytical measurements:

Particle size evaluation

After the screening process, samples of WM-OPMF and DM-OPMF which passed through mesh size of 150 μm were observed for particle size geometry (length and diameter) and aspect ratio study. A light microscope (Motic : Model BA310, China) connected to an image analyzer (Motic Images plus 2.0) was used for measuring the length and diameter of 70 particles of both samples.

Analytical measurements

Thermogravimetric analysis (TGA) was conducted on a TG analyser model EXSTAR6000 TG/DTA6200. OPMF samples (6 – 8 mg) were heated from 50 – 550 °C at a heating rate of 10ºC/min under nitrogen flow of 100 ml/min. The surface morphology of DM-OPMF and WM-OPMF samples was observed under a scanning electron microscopy (SEM) with a model LEO 1455 VPSEM with Oxford Inca EDX. For SEM analysis, dried OPMF samples were mounted on the stub and gold-coated for 180 sec prior to SEM observation.

Tensile test

The composites were compressed with temperature of 170 °C into 1 mm sheets and cut into tensile dump-bell shape following the ASTM method D-638 type-V. Tensile test was carried out using Instron

Universal Testing Machine (Model 4302 Series IX) based on ASTM D638. The test was conducted at a constant crosshead speed of 5 mm/min, load cell of 1 kN and a gauge length of 10 mm. An average of five results was taken as the resultant value.

RESULTS AND DISCUSSION

Particle size distribution

Particle size distribution (PSD) of WM-OPMF and DM-OPMF is shown in Fig. 1. DM-OPMF showed higher percentage of small size particles (<75 μm) compared to WM-OPMF by almost three-fold. From the same figure, it can be seen that disc milling able to reduce the diameter of OPMF, with more than 50% of the OPMF fibers had particle size of <100 μm. This is in contrary with WM, whereby only 20 % of the fibers had particle size of <100 μm. This observation is contributed by the mode of action of each milling process whereby WM only able to shorten the length of the OPMF by cutting knife/teeth without affecting the diameter of the fiber. On the other hand, DM able to reduce the size of OPMF by milling between the rotating disc resulting cutting to short length, imparting and shearing to smaller diameter and even able to expose microfibril of the OPMF.

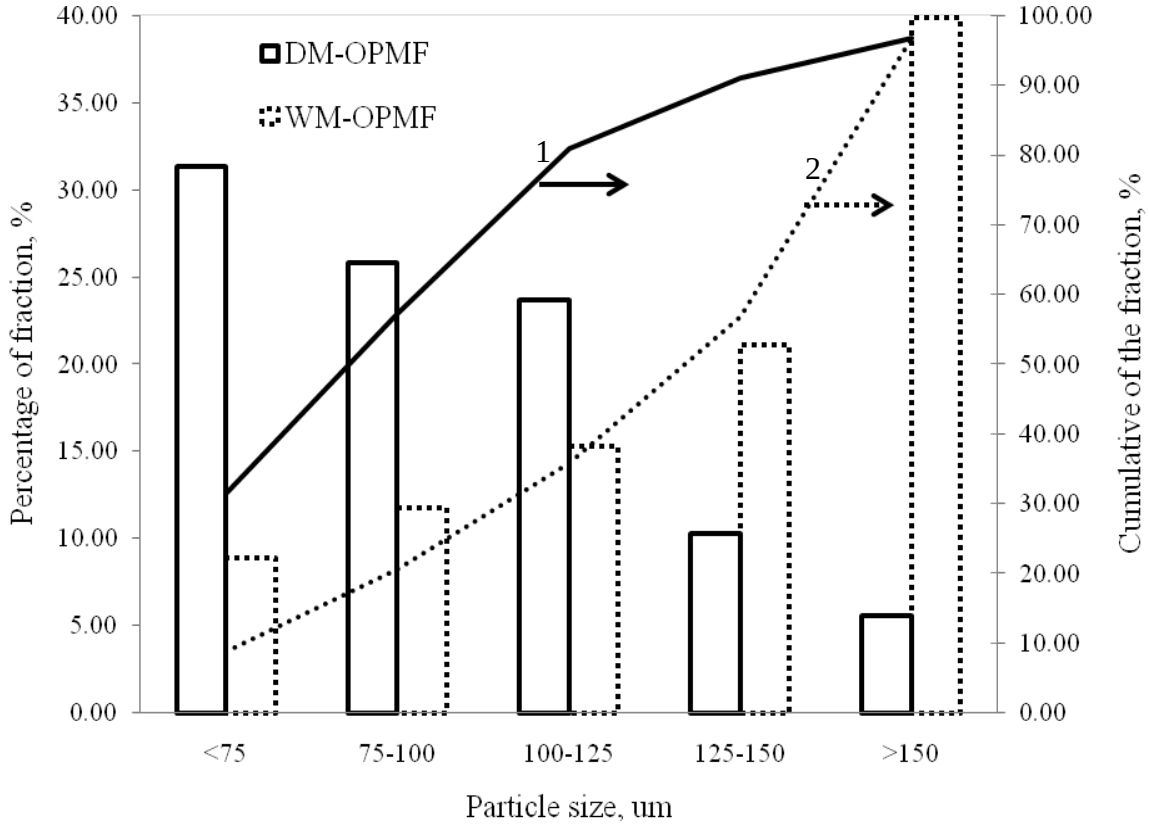

Fig. 1 : Particle size distribution for DM-OPMF and WM-OPMF. Bar graph indicated fraction of particle size (%) and line showed cumulative of the fraction (%), (1) DM-OPMF and (2) WM-OPMF.

The fibers were also measured for their length, L and diameter, D through microscopic analysis. The distribution of the L and D of the milled-OPMF is shown in Fig. 2. Overall, it is seen that both L and D of the DM-OPMF converged towards the lower size with average L and D values of <200 and <50 μm, respectively. WM-OPMF on the other hand has wider distribution of fiber length and diameter. This observation contributes to the aspect ratio values of DM-OPMF and WM-OPMF (Fig. 3). Since DM-OPMF has low L and D values, this contributed to the low aspect ratio of DM-OPMF, with average of aspect ratio of 5. Even though WM-OPMF seemed to have high aspect ratio with average value of ≈10, however the average length and diameter of the fiber is relatively high for biocomposite preparation [4]. Juliana et al. [2] discussed that thin and long particles may increase the contact area between the fiber and polymer matrix, resulting better adhesion with

these two materials. We managed to get thin fiber by using disc milling, however due to the shear effect, the fibers tend to be short. Overall, the particle size of disc milled-OPMF is much smaller compared to that obtained from Wiley mill.

Morphological analysis

SEM analysis was done to support the above findings, as shown in Fig. 4 (a – d). It is seen from the figures that DM-OPMF had smaller size (L/D) as compared to WM-OPMF. Observation can be explained by the mode of action of disc milling which involves crushing, shearing and cutting of the fiber samples [7]. Microfibril structure of DM-OPMF can be clearly observed at higher magnification. Shearing force by rotating disc of DM able to open the pores of the fiber, thus makes PP easily to penetrate deep to OPMF structure.

Thermal stability of milled OPMF

DM-OPMF and WM-OPMF fibers with size <150 μm were blended with PP in the mixer with 20 and 50 % (w/w) of fiber loading. Thermal analysis of the milled fiber and biocomposite samples were done to evaluate the thermal stability of the samples. Fig 5 shows the TG curves of DM-OPMF, WM-OPMF and PP-OPMF biocomposites. From the figure, both DM-OPMF and WM-OPMF had almost similar thermal degradation trend, with three-step degradation indicating the three major components in lignocellulose, *i.e.* hemicellulose, cellulose and lignin.

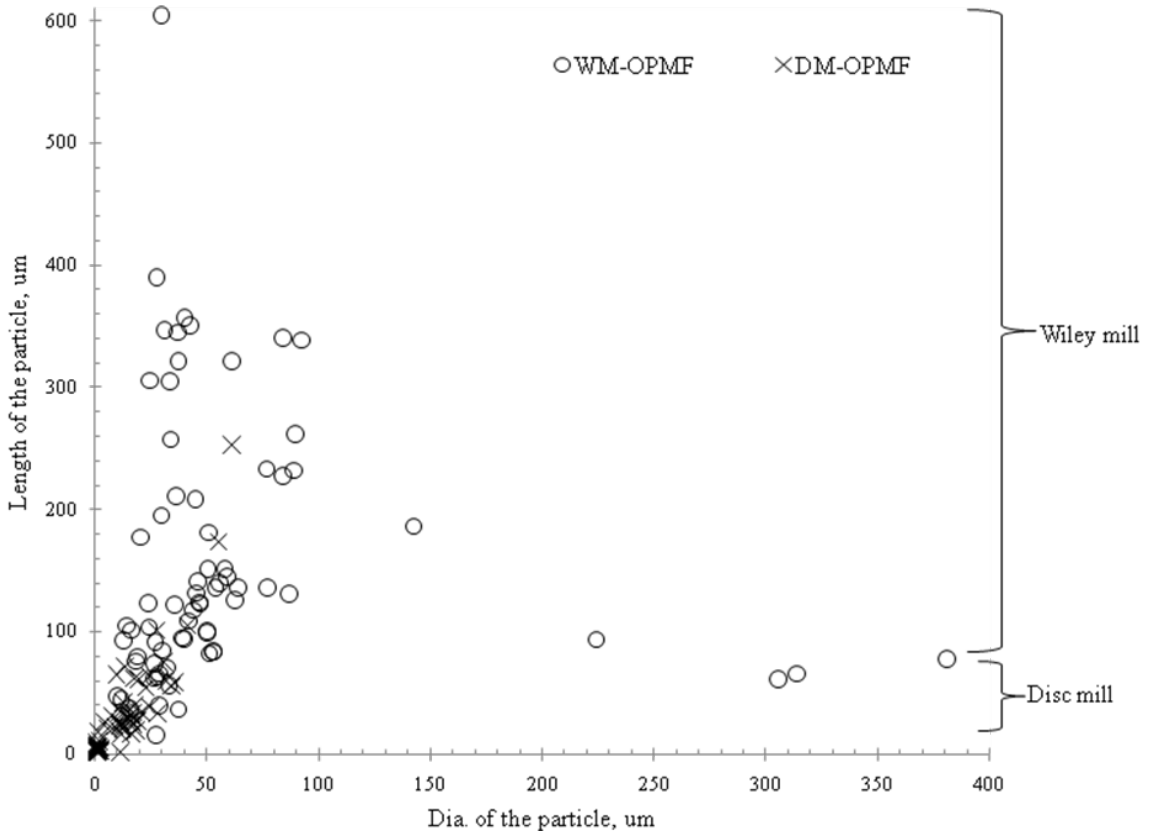

Fig. 2 : Distribution of diameter and length of the particle for disc mill and Wiley mill.

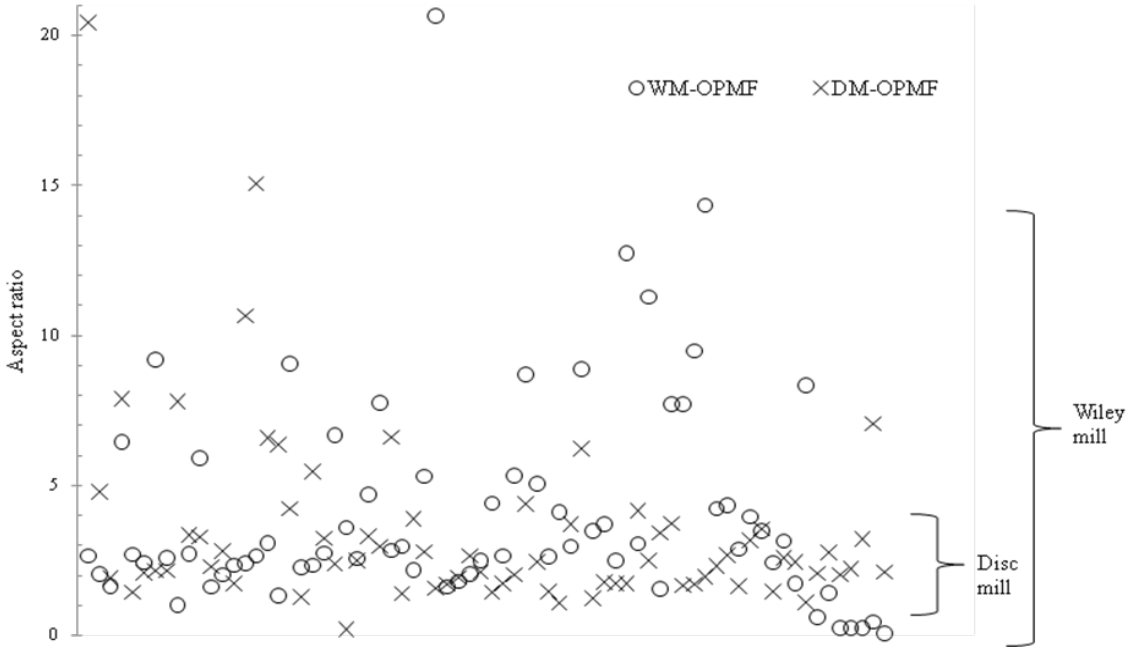

Fig. 3 : Distribution of aspect ratio in disc mill and Wiley mill particles.

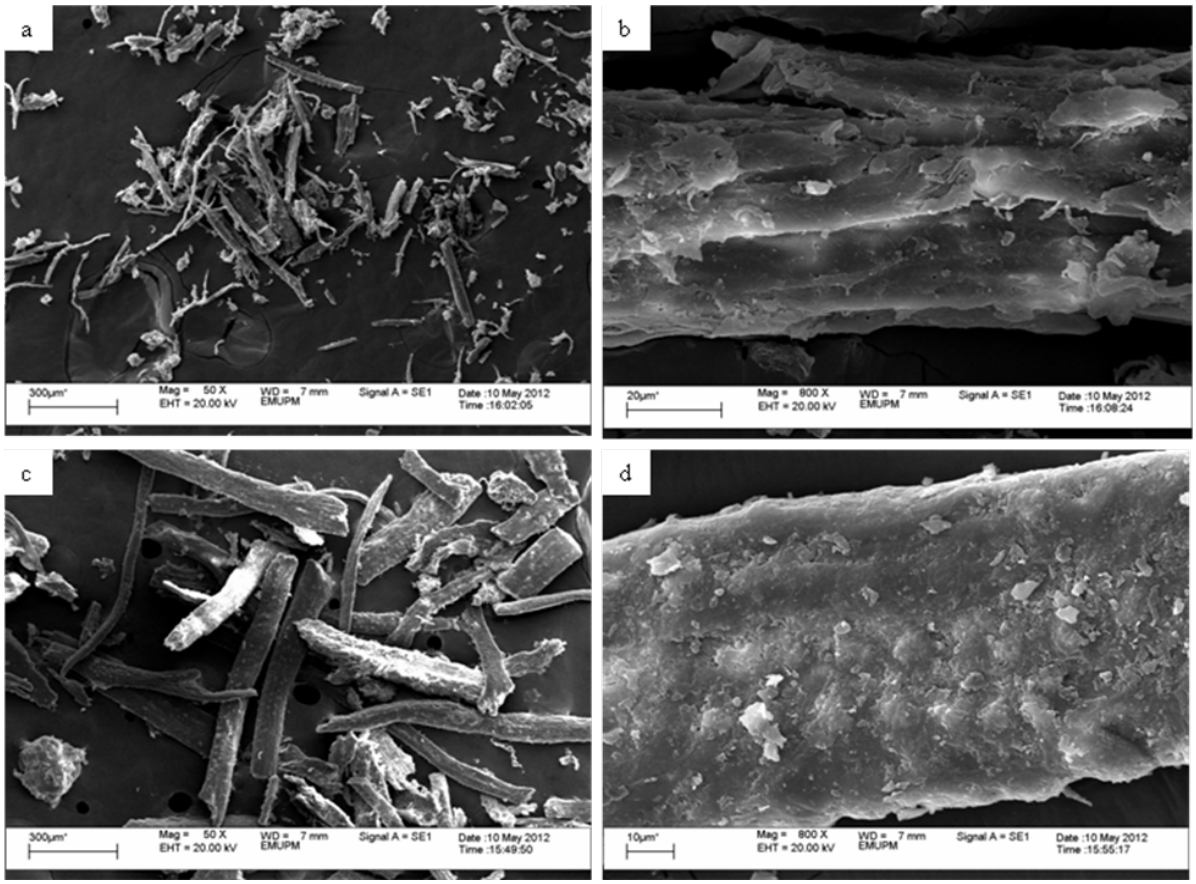

Fig. 4 : Morphological difference between samples : a , b disc mill; c , d Wiley mill.

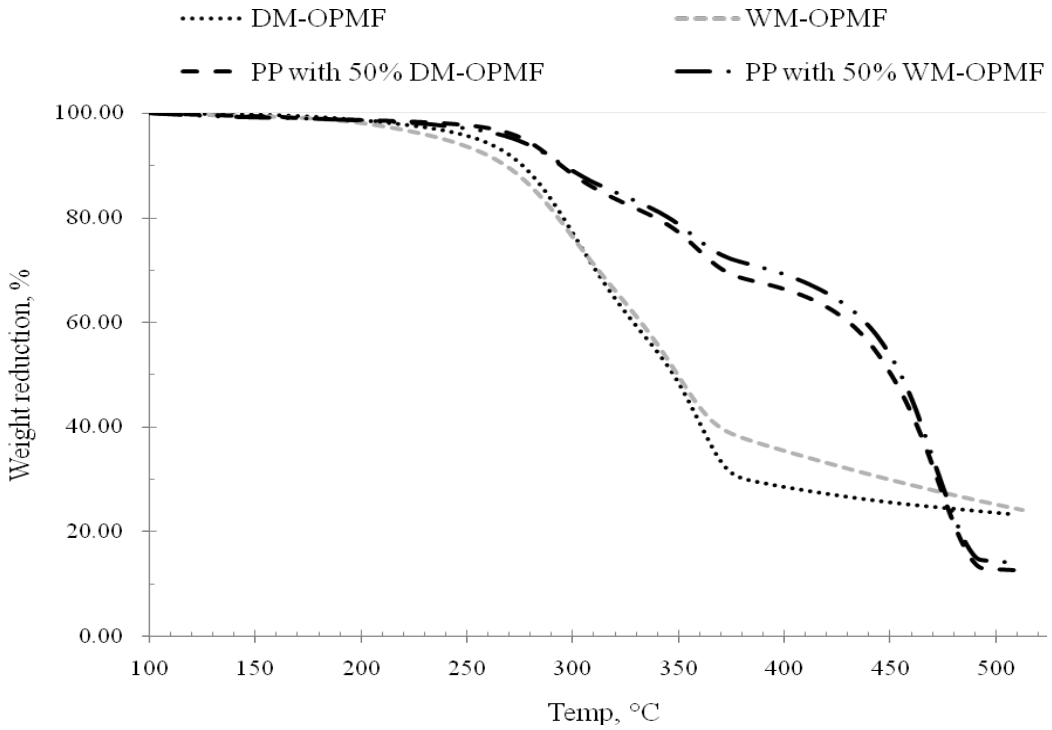

Fig. 5 : TG curves of DM-OPMF, WM-OPMF and PP reinforced OPMF biocomposite

Nevertheless, the DM-OPMF degraded faster at higher temperature range, above 350 ºC. The summary of thermal decomposition profile of each sample is presented in Table 1. From the table, it is seen that the total weight loss for DM-OPMF at temperature range of 300 – 450 ºC was 6% higher compared to that of WM-OPMF. The higher thermal stability of WM-OPMF at high temperature range can be contributed by lignin component in the sample [8]. WM-OPMF may contain higher lignin content compared to DM-OPMF. This can be explained by lignin loss due to shear effect during disc milling. The structure of lignin component in DM-OPMF may be disrupted by the shear and friction during disc milling and hence, the lower lignin content in DM-OPMF. This is supported by our morphological analysis by SEM (Fig. 4) which shown that DM-OPMF have smaller fiber size with lower diameter value. Meanwhile, from Fig. 2 we can see that the diameter of DM-OPMF is generally much lower (<50 μm) compared to that of WM-OPMF (up to 400 μm). The small diameter of fiber indicates that the cellulose macrofibrils have been ripped into microfibrils due to mechanical action. In lignocellulose material, lignin acts as glue in between the cellulose fibrils [9] causes them to be arranged in stacks and hence give strength to the material. The disruption of lignin causes the cellulose fibrils to tear apart and hence the formation of microfibrils. Moreover, Lundquist [10] highlighted that milling is expected to cause chemical changes in lignin.

Table 1 : Summary of degradation temperature of DM-OPMF and WM-OPMF.

Sample	$T_{5\%}$ (°C)	Weight loss (%)		Residue at 550 °C (%)
		Temp region (°C)		
		150-250	300-450	
DM-OPMF	256	4	52	23
WM-OPMF	240	7	46	24

PP-reinforced OPMF biocomposite

PP-reinforced milled OPMF biocomposites were tested for thermal stability and tensile strength. Generally PP has higher thermal stability compared to lignocellulose and hence the biocomposite samples had better thermal stability compared to OPMF, as seen in Fig. 5. Overall, biocomposites prepared from DM-OPMF

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and WM-OPMF showed almost similar trend in thermal degradation from beginning to completion indicating that alteration in lignin content in DM-OPMF did not really affect the biocomposite thermal property. The tensile strength of WM-OPMF and DM-OPMF biocomposites were tested in order to evaluate the performance of the biocomposites. Fig. 6 showed the tensile strength of PP/WM-OPMF and PP/DM-OPMF biocomposites at 20 and 50 % (w/w) fiber loading. There was marked different in tensile strength value for biocomposite prepared from DM-OPMF with that from WM-OPMF, especially for sample with 20 % (w/w) of fiber loading. Biocomposite with 20 % (w/w) DM-OPMF had tensile strength of 23.6 MPa, which is 83 % stronger compared to the biocomposite with 20 % (w/w) WM-OPMF. The same observation can be seen for biocomposite prepared with 50 % (w/w) fiber, even though the difference in tensile strength value was not as high as biocomposite with 20 % (w/w) fiber. The higher tensile strength demonstrated by biocomposite prepared from DM-OPMF could be contributed by the smaller size of fiber particles as illustrated in Fig. 1, 2 and 4. Even though OPMF used in this study were obtained after sieving to pass through 150 μm mesh, however microscopic analyses by light microscope and SEM revealed that fibers with larger particle size (long fiber) can pass through the mesh since the fibers passed through the mesh vertically. Based from the observation, it can be concluded that DM-OPMF had smaller fiber size and hence gives higher surface area per volume which can improve the surface contact between fiber and polymer matrix. Higher surface contact between fiber and polymer may improve the physical adhesion between the two materials. This explains why PP/DM-OPMF had better tensile strength compared to PP/WM-OPMF. Moreover, the small size of DM-OPMF may improve the stress transfer between polymer matrix and fiber compared to longer fiber. Small diameter fiber also makes it easily arrange in the polymer matrix and hence, caused the tensile strength to be improved in PP/DM-OPMF biocomposite.

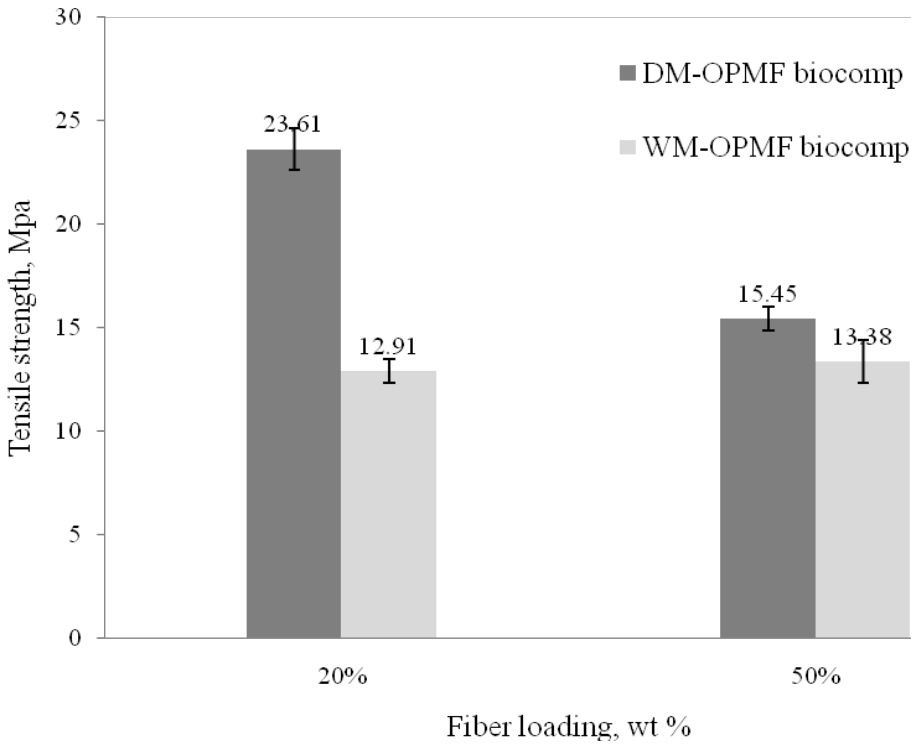

Fig. 6 : Tensile strength of DM-OPMF and WM-OPMF biocomposite prepared from fiber with particle size <150 μm with 20 and 50 % (w/w) of fiber loading.

CONCLUSIONS

Two different grinding techniques were used to grind OPMF for biocomposite production. Ground DM-OPMF and WM-OPMF were sieved to pass through different sieve size to determine the particle size distribution and aspect ratio of the particles. DM-OPMF showed higher percentage of small size particles (<75

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μm) compared to WM-OPMF by almost three-fold. Disc milling acted on OPMF by crushing, shearing, ripping and cutting the OPMF samples, resulted in disruption in lignin structure, and hence unmasked the macrofibrils to form microfibrils. . This affected the tensile strength of the PP/OPMF biocomposites. PP/DM-OPMF biocomposite with 20 % (w/w) fiber had tensile strength of 23.6 MPa, which is 83 % higher compared to PP/WM-OPMF biocomposite. This study revealed that milling has pronounced effect in improving mechanical property of biocomposite. Disc milling provides smaller size fiber particles compared to Wiley mill and hence, enhances surface contact between fiber and polymer matrix.

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