Tensile and impact properties of pulverized oil palm fiber reinforced polypropylene composites: A comparison study with wood fiber reinforced polypropylene composites

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ABSTRACT

The comparison of tensile and impact properties between the fiber extracted from Malaysia’s oil palm plantation wastes and residues and wood fiber that are going to be used as reinforcing agent in polymer composite materials is the main focus of this study. For that purpose, 10 wt.% and 25 wt.% of Malaysian oil palm fiber (OPF) extracted from oil palm empty fruit bunches (OPEFB) was incorporated with polypropylene (PP) and maleated polypropylene (MAPP) to produce injection-molded composite materials. For comparison purposes, virgin PP and wood fiber reinforced polymer composites (WPC) were also fabricated as benchmark samples. Firstly, tensile test was performed for all three samples; virgin PP and wood fiber reinforced polymer composites (WPC) in order to investigate the effect of fiber loading on composite materials. From the tensile test result, 25 wt.% of fiber loading produced higher tensile properties than 10 wt.% of fiber loading. Moreover, the obtained OPF/PP composites showed comparable properties with the WPC material. The equality of properties between OPF/PP composites and WPC indicates the potential of wastes and residual materials such as OPF to be broadly utilized in the production of composite materials.

Keywords: oil palm fiber; polypropylene; wood plastic composites (WPC); tensile strength; impact strength; size effect; failure behavior.
INTRODUCTION

Ongoing research on composite materials is focusing more on natural fiber based composites. Polymer composites reinforced with natural fibers such as kenaf, flax, hemp and ramie show high potential to be further developed and applied. The improvements on mechanical properties of the natural fiber based composites are often reported in the literatures in recent years [1-4]. Poor adhesion between fiber surface and polymer matrix, high moisture absorption due to the nature of the hydrophilic natural fibers, variations of natural fiber’s parameters such as unstable properties due to different origin plant, water uptake during growing process and harvesting time are among the arising issues of using natural fiber as the reinforcement constituent in polymer composites. To some extent, current and ongoing researches have established certain solutions on dealing with the issues mentioned above such as the use of compatibilizer or coupling agents as a bridge. For example, an interaction between the anhydride groups of maleated coupling agents and the hydroxyl groups of natural fibers is used to improve the incompatibility of natural fibers and polymeric resin. T.J Keener et al. [1] in their study on maleated coupling agents for natural fiber suggested that the optimization of 3wt.% of coupling agents in composite materials can greatly enhance the properties of natural fiber based polymer composites.

Wood is one of the natural resources that has been widely used in many applications, ranges from a structural application for buildings to a small kitchenware and home appliances products. It has been used for ages and in modern application, the wood particles are used as filler in many thermoplastic polymers to produce a composite material known as wood plastic composites (WPC). The use of wood filler along with the compatibilizer helps to enhance the mechanical properties of plastic based composites [2,5]. Furthermore, the incorporation of wood fillers gives a wooden look to the WPC products. WPC is broadly used as a material for home furnishings such as kitchen cabinet, fence, cladding, decking, and flooring. It proves to be a competitive material for automotive interior parts and potentially for other non-load bearing applications of wood. Wide applications of WPC signify the utilization of natural fibers for various composite materials appliances.

Recently, the natural fibers that extracted from the plantation residues such as oil palm, bagasse, coconut/coir, pineapple leaf, rice husk and straw, and many others have attracted attention to be utilized as a component of the composite materials. As for the oil palm fiber (OPF), it is extracted from the empty fruit bunch (EFB) after the separation process of oil palm fresh fruit from the bunches. Meanwhile, the oil palm fresh fruits are processed to produce palm oils. The production of palm oil is increasing by years due to the high demand for various products using it as an organic raw ingredient. Palm oil has been used for many applications including personal care products, cosmetics, cleaning liquids, and also as an ingredient in foods and beverages. In 2011, Indonesia and Malaysia accounted for approximately 86% of global palm oil production [6]. Generally, palm oil consists only 10% of the total biomass while the rest are disposed as waste [6]. The increase of palm oil production can potentially escalate the number of agricultural wastes such as fronds and trunks, together with the palm oil residues; EFB, kernels and mesocarp fibers. In total, approximately 91 million metric tons of residues and wastes were produced every year and EFB corresponds to about 20% of this total [7]. The number of wastes and residues from the oil palm plantation are shown in Table 1.
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Table 1. Wastes and residues amount from the oil palm plantation in Malaysia [6].

<table>
<thead>
<tr>
<th>Wastes/residues</th>
<th>Amount (Million tonnes)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Empty fruit bunches</td>
<td>7.0</td>
</tr>
<tr>
<td>Mesocarp fibers &amp; shells</td>
<td>11.6</td>
</tr>
<tr>
<td>Trunks</td>
<td>13.9</td>
</tr>
<tr>
<td>Fronds</td>
<td>44.8</td>
</tr>
</tbody>
</table>

Because of the arising awareness of environmental problems due to the abundance of agricultural waste and maximizing the use of EFB, OPF appears to be an attractive option to be used as a reinforcement in fiber-filler polymer composite materials. On top of that, the cellulose composition in OPF is comparable with other natural fibers that are used widely in various composites products nowadays. Table 2 summarizes the average properties and compositions of wood fiber, OPF, and other natural fibers. Additionally, the use of OPF in composite materials production is expected to diversify the sources other than wood in strengthening the environmentally-friendly-polymer composite materials.

Table 2. Properties and compositions of natural fibers [4,8-9].

<table>
<thead>
<tr>
<th>Properties</th>
<th>OPF</th>
<th>Abaca</th>
<th>Bamboo</th>
<th>Sisal</th>
<th>Softwood</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tensile strength (MPa)</td>
<td>248</td>
<td>400</td>
<td>140-230</td>
<td>511-635</td>
<td>-</td>
</tr>
<tr>
<td>Density (g/cm³)</td>
<td>0.7-1.55</td>
<td>1.5</td>
<td>0.6-1.1</td>
<td>1.5</td>
<td>-</td>
</tr>
<tr>
<td>Cellulose (%)</td>
<td>42.7-65</td>
<td>56-63</td>
<td>26-43</td>
<td>65</td>
<td>40-45</td>
</tr>
<tr>
<td>Hemicellulose (%)</td>
<td>17.1-33.5</td>
<td>20-25</td>
<td>30</td>
<td>12</td>
<td>7-14</td>
</tr>
<tr>
<td>Lignin (%)</td>
<td>13.2-25.31</td>
<td>7-9</td>
<td>21-31</td>
<td>9.9</td>
<td>26-34</td>
</tr>
<tr>
<td>Moisture (%)</td>
<td>-</td>
<td>1.5</td>
<td>8.9</td>
<td>11</td>
<td>-</td>
</tr>
<tr>
<td>Waxes (%)</td>
<td>4.5</td>
<td>3</td>
<td>-</td>
<td>2</td>
<td>-</td>
</tr>
</tbody>
</table>

Azman Hassan et al. [10] in their review article on OPF-polymer composites mentioned several issues related to natural fiber composites such as the compatibility between OPF and matrix, and the moisture absorption. The optimization of OPF fiber and compatibilizer might resolve some of the mentioned issues and led to several consequences on the physical and mechanical properties of the composite materials. On the other hand, fiber length or the aspect ratio of the fiber as described in certain literatures is one of the parameters that significantly contributes to the performance of the composite materials. Amir Nourbakhsh et al. [2] suggested in their study on the effect of wood fiber particle size on the mechanical properties of the particulate-filled polymer composite, that smaller particle size by approximately 0.25mm produced excellent mechanical properties of the wood fiber-PP composites. Moreover, Sébastien Migneault et al. [3] studied the effect of fiber length on processing and properties of extruded wood fiber/HDPE (high-density polyethylene) composites. The results showed improvement in both tensile and flexural properties of the WPC by increasing the fiber length. As above mentioned, WPC was successfully used in various applications. Thus, this study aims to investigate and identify the possibility of OPF to be utilized as structural material in the same way as wood fiber utilization in WPC application. For this purpose, two types composites specimens; wood fiber reinforced PP and OPF reinforced PP composites, hence referred as WPC and OPF/PP composites respectively, were prepared through a kneading and injection molding process. The mechanical properties of both composites made with
different loads and fiber sizes were investigated. Further analysis on the fracture surface of the specimen was carried out by using a scanning electron microscope and a laser microscope.

METHODS AND MATERIALS

Production method
To investigate the influence of different fiber sizes and loading amount on the mechanical properties of composite materials, the wood fiber and OPF were screened into several ranges by using a vibration sieve machine. Firstly, they were screened into two different size ranges; less than 90 µm and larger than 90 µm by using a 90 µm mesh opening sieve. However, due to the longer raw fiber, the raw OPF was cut into approximately 3cm long, then 2mm via a cutting machine, cutting mill P-15 (Fritsch Japan Co. Ltd.) and finally pulverized to 0.2mm by using a cutter mill P-14 (Fritsch Japan Co. Ltd.). Then, a medium size vibration sieve machine was employed to screen the pulverized OPF with the mesh opening size of 45µm, 90µm, and 180µm. As the result, the fibers were screened into three sizes; less than 45µm, 45µm to 90µm and larger than 90µm, which were denoted as S, M, and L, respectively.

Table 3. Composition of wood, palm, PP and MAPP in Neat PP and composite specimens.

<table>
<thead>
<tr>
<th>Specimens</th>
<th>Wood (wt.%)</th>
<th>Palm (wt.%)</th>
<th>PP (wt.%)</th>
<th>MAPP (wt.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neat PP</td>
<td>0</td>
<td>0</td>
<td>100.0</td>
<td>0</td>
</tr>
<tr>
<td>W-10%</td>
<td>10</td>
<td>0</td>
<td>88.7</td>
<td>1.3</td>
</tr>
<tr>
<td>W-25%</td>
<td>25</td>
<td>0</td>
<td>74.0</td>
<td>1.0</td>
</tr>
<tr>
<td>P-10%</td>
<td>0</td>
<td>10</td>
<td>88.7</td>
<td>1.3</td>
</tr>
<tr>
<td>P-25%</td>
<td>0</td>
<td>25</td>
<td>74.0</td>
<td>1.0</td>
</tr>
</tbody>
</table>

The composites specimens were prepared by mixing the PP with the screened fibers respectively and the compatibilizer MAPP in a twin screw internal mixer, Laboplasto Mill (Toyo Seiki Seisaku-Sho Ltd.) at 30rpm and 190°C for 10 minutes. The compositions of reinforcement fibers and matrix are shown in Table 3. Mixed compound was then granulated and stored at 80°C for at least 4 hours. By using an injection molding machine BabyPlast 6/10P (Rambaldi Group), the granulated compound was then molded at 200°C into a 60mm×10mm×2mm dumbbell-shaped and 60mm×10mm×3mm rectangular-shaped of tensile and impact test specimen, respectively. Figure 1 shows the shape and dimension of tensile and impact tests specimens and the notch condition on impact specimen as referred to the ASTM D638 and JIS K7111.
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Figure 1. Shape and dimension of (a) tensile and (b) impact test specimens, and (c) notch on impact specimen.

Tensile and impact tests
The tensile test was conducted by using Servopulser hydraulic type EHF-FB-4LB (Shimadzu Corporation) at room temperature with three different cross-head speeds; 1.5 mm/min, 10 mm/min and 100 mm/min. Five specimens were tested for each type of the composites and virgin PP. A strain gauge (Kyowa Electronic Instruments Co., Ltd.) was installed at the middle of gauge length to measure the strain as the specimens were pulled. In addition, Charpy impact test was carried out at room temperature for Neat PP, WPC and OPF/PP composites by using an Izod-Charpy impact test machine, CIT-25J-CI (A&D Company, Limited). Five specimens for each type of composites and virgin PP were tested. Both notched and unnotched specimens were tested for the Charpy impact test. The V-notch with a radius of 0.25mm±0.05mm was made at the center of the specimens by using a PFA plastic forming machine (Yasuda Seiki Seisakusho Ltd.).

Fracture surface observation
In order to understand the fracture behaviour of the specimen, the fractured surface of each type of tensile and impact specimens were observed by using a 3D measuring laser microscope, OLS4000 (Olympus Corporation). The notched impact test specimen was conducted by using a field emission scanning electron microscopy, JAMP-9500F FE-SEM (JEOL, Ltd.).

RESULTS AND DISCUSSION

Tensile test
Figures 2(a), (b) and (c) show the average tensile strength values of Neat PP, OPF/PP composites and WPC specimens at different fiber loading amount and cross-head speeds. As illustrated, the incorporation of wood and palm fibers into PP matrix enhance the tensile strength of the virgin PP at any cross-head speed. The fillers help to transfer the high load and the use of MAPP enhances the interfacial bonding between the fibers and the PP matrix, thus improves the tensile properties of the composites. Moreover, there is a significant increase of tensile strength by increasing the amount of the fibers to 25wt.%.

Unlike wood, the classification of OPF brings significant results for PP-based composites at 25wt.% of loading amount. This can be seen by comparing the unclassified OPF/PP composites; P-25%-MIX, with the classified OPF/PP composites; P-25%-S, -M, and -L, in Figure 2(b). There is comparatively similar tensile strength obtained for classified and unclassified WPC. Classified OPF of -L size has the greatest influence on the performance of OPF/PP composite, while the classification of wood fibers leads to almost no significant improvement on WPC performance. Figures 3(a) to (c) show the
different fiber dimensions of classified OPF and Figures 3(d) to (f) show the different fiber dimensions of wood fibers. It is clearly proved that the pulverizing process lowering the aspect ratio of OPF, which is classified into -M or -S sizes. For the dimension of wood fibers, the pulverizing process also reduces the aspect ratio. However, several high aspect ratio of wood fibers still remain mixed with the low aspect ratio as shown in Figure 3(d). By comparing to OPF, the wood fibers show less disparity in fiber aspect ratio.

The effectiveness of OPF as compared to wood fiber was quantitatively evaluated by dividing the average tensile result of composites with classified OPF of -L size by the tensile strength of WPC. As the result, at any loading rate, the OPF/PP composites exhibit a comparable strength at 96.7% or higher of the WPC material at 10wt% of fiber loading amount. Meanwhile, at 25wt.% of OPF loading amount indicates 95% to 97% of comparable strength as compared to WPC. In addition, for the tensile strength performance, as mentioned above, it is understood that the larger fiber aspect ratio is conceived to transfer the load more efficiently. Regarding the comparison between -M and -S sizes, the latter one has higher tendency to form a cluster, so-called agglomeration, as shown in Figure 4. Such an agglomeration often increases the stress concentration, thus inhibits the stress transfer and accelerates the fracture behavior of the system.
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Figure 2. Average tensile strength value of Neat PP, OPF/PP composites and WPC at different fiber loading amount and cross-head speed; (a) 1.5mm/min, (b) 10mm/min and (c) 100mm/min, respectively.

Figure 3. Dimension of wood fiber and OPF between each of the classifications; (a) OPF less than 45 µm, S, (b) OPF 45 µm to 90 µm, M, (c) OPF larger than 90 µm, L, (d) wood fiber less than 90 µm, SM, (e) wood fiber larger than 90 µm, L, and (f) unclassified wood fiber, MIX, respectively.

From Figures 2(a), (b) and (c), it is understood that the tensile strength of both virgin PP and OPF/PP composites increases as the cross-head speed increases. In other words, the obtained results reflect the dependency of strain-rate on the tensile properties for both PP and composites materials. This behavior can also be observed on the fracture surfaces. As shown in Figures 5(a) to (c), the whitened areas caused by PP fibrillation at the fracture surface denote the region where the crack grows along with the increase of tensile load [11, 12]. The higher cross-head speed of tensile test causes smaller crack growth region
(Figure 5(c)) as compared to the whitened area of the tested specimen at lower cross-head speed (Figure 5(a) and 5(b)). It is customary to relate this phenomenon to the viscoelastic nature of PP, where it behaves as hard and brittle at a higher tensile speed that resulting in lower fibrillation process (whitening). Thus, it reduces the size of whitened area.

![Figure 4](image_url)

Figure 4. Fracture surface of P-10%-S specimen indicates the agglomeration of smaller size OPF.

![Figure 5](image_url)

Figure 5. Fracture surface observation on specimen tested at different tensile speed; (a) 1.5mm/min, (b) 10mm/min and (c) 100mm/min.

**Impact test**

The effect of fiber loading amount and size on impact properties of WPC and OPF/PP composites were investigated. Figure 6 illustrates the plotted data obtained from the notched specimen of Charpy impact test results. Based on the plotted data, it is indicated that the virgin PP has a higher value of impact strength as compared to the composites materials. However, at 25wt.% of fiber amount in the WPC and OPF/PP composites provide a nearly equal impact resistance performance of the virgin PP. This is expected as the increasing number of fiber loading amount creates fiber pull-out mechanism and increases the energy absorption. The result from the Charpy impact test of notched specimens indicates that the higher loading amount of OPF might further increases the impact strength, as achieved in notched Charpy tests of injection-molded glass/PP [13], abaca/PP [14] and cellulose/PP [14] composites. The holes from the fiber pull-out mechanism can be identified and qualitatively confirmed from Figure 7. Figure 7 shows the scanning electron microscopy (SEM) images of the fractured surface of notched Neat PP, 10wt.% and 25wt.% of OPF/PP composites specimens. From the observation, the number of holes rises as the amount of OPF increases, which means an increase of energy.
absorption during the occurrence of specimen's failure. On the other hand, as for the notched specimens, there is no clear trend can be observed on the impact strength with the varying sizes of OPF.

![Graph showing Charpy impact test results](image1)

**Figure 6.** Charpy impact test result of notched WPC and OPF/PP specimens.

![Scanning electron microscopy (SEM) results](image2)

**Figure 7.** Scanning electron microscopy (SEM) result of notched Charpy impact test specimen. A number of holes indicates fiber pull-out at the fracture surface.

Figure 8 shows the Charpy impact test results of unnotched specimens. The impact strength decreases with increasing of OPF loading amount and Neat PP has the highest impact strength value. This trend agrees with the previous literatures regarding bamboo/PVC (polyvinyl chloride) [15], flax/PP [16], ramie/PP [17], and abaca/PP [18], where the incorporation of fillers leads to decreasing of impact strength value due to the reduction of the material's ability to absorb energy during crack propagation. Based on Figure 8, the impact strength values of the medium, -M size fiber; P-10%-M and P-25%-M are slightly higher than that of smaller, -S and larger, -L size fibers. From the Charpy impact test results in Figures 7 and 8, the effect of the fiber's classification can be clearly seen from the unnotched specimen than that of the notched specimen results. It can be observed from SEM photograph that, the impact fracture surface of -S size of OPF/PP composites contains several agglomerations, as in the tensile fracture surface.
Due to only one specimen was successfully tested for the P-10%-L condition, whereas the results for -L and -M are almost similar, the -M size of OPF is used as a comparison to WPC material in this study. As compared to similar range size of wood fiber, -SM, the OPF/PP composites obtained 98.7% or higher impact strength, which is comparable to WPC for both notched and un-notched Charpy impact test specimens. In some cases, for example, 10wt.% of OPF indicates higher impact strength than that of W-10%-MIX specimen. As for the real production of the material is not accompanied by a notch, the Izod impact test for unnotched specimen was additionally conducted. Figure 9 shows the Izod impact test results of unnotched specimens where similar trends to Charpy unnotched specimen were confirmed. As for Izod test, the impact strength of 25wt.% of OPF/PP composites is approximately 93% as compared to the impact strength of WPC materials with the same filler content.
CONCLUSIONS

In this study, Japanese type cedar thinned wood and Malaysian oil palm fiber (OPF) were used as the reinforcements for PP matrix composite materials. The fibers were respectively classified into different sizes in order to investigate the effect of fiber loading amount and size on mechanical properties of the composite materials. The obtained results in this study are summarized as follows:

1) From the experimental results, it is understood that the tensile strength of the composites increases with the increment of OPF amount. The larger size of OPF slightly increases the tensile properties of the OPF/PP composites. Moreover, the incorporation of OPF into PP enhances the tensile strength and stiffness properties of virgin PP specimens.

2) The tensile strength of OPF/PP composites is 95% or higher level of that of wood/PP composites (WPC). In other words, the utilization of OPF into PP matrix demonstrates almost similar strength of WPC materials.

3) From the impact test results of the notched specimens, there is no significant trend of the different fiber size of the OPF/PP composites can be observed. The level of impact strength is almost equal to WPC materials. It can be observed from the un-notched specimens that the impact resistance of the OPF/PP composite material decreases with the increment of OPF amount. The OPF/PP composites exhibit almost the same level of Charpy impact strength as WPC, while shows approximately 93% of WPC impact strength as for the un-notched Izod test.

4) It is concluded that OPF has high tendency to be used as the reinforcement in the composite materials as the same way as wood fiber by choosing an appropriate classification of OPF.

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