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Effect of MAPP Addition on Impact Strength, Water Absorption, and Filler Volume Fraction of the Waste Polypropylene/Modified Coconut Wood Flour/Glass Fiber Hybrid Composite

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ABSTRACT

In this study, hybrid composite materials were prepared from combination of modified coconut wood flour (MCWF) and glass fiber (GF) as fillers, and waste polypropylene (WPP) as matrix. The coconut wood flour (CWF) was modified by 18% sodium hydroxyde solution to reduce its adhesion between polypropylene matrix and the polar fillers could be enhanced by using maleic anhydride-g-polypropylene (MAPP) which was synthesized from the refluxed anhydride, polypropylene, xylene and benzoyl peroxide at 135°C. MAPP and GF composition were made constant at 2 wt.% and 10 wt.%, respectively and MCWF composition was varied from 10 – 40 wt.%. Mixing process was carried out in an extruder at 180°C and the samples were then molded in a hot press machine. The impact strength, water absorption, and filler volume fraction analysis were investigated. The results showed that addition 30 wt.% of MCWF and 10 wt.% of MAPP can elevate the impact strength of the composite. The study also revealed that water absorption of the composite has reduced with the utilization of MAPP. As comparison, composite without MAPP, no fillers and using one type of filler were also prepared. These results were also supported by Fourier Transform Infra-Red (FTIR) and Scanning Electron Microscopy (SEM) analysis.

KEYWORDS: Coconut Wood Flour, Hybrid Composite, Waste Polypropylene.

INTRODUCTION

Hybrid composite is a system constituted by two different reinforcements incorporated into a single matrix or establishment of a single reinforcement into two different matrices. The properties exhibited by the hybrid composite is generally the combination of the individual properties of the individual materials.
constituents which would give convenience in balancing each of their superior and inferior properties. Introduction of extra reinforcement or matrix are conducted to overcome the weakness of the other matrix, and this will eventually lead to cost and perform an appropriate material design. Using a hybrid composite that contains two or more types of different fibers, the advantages of one type of fiber could complement deficiencies in the other \[^{[1,2]}\]. Environmental issues which has become major concern is the main reason to develop replacement of synthetic fibers with natural fibers. However, the fact if the natural fibers reinforced composites provide lower mechanical properties and resistance when compared to synthetic fibers (such as glass fibers) cannot be neglected. Therefore, in order to weaken weaknesses, glass fiber (GF) and natural fiber can be integrated in a matrix to formulate a hybrid composite that demonstrates the properties of its constituents, and thereby, an economical and friendly composite. Coconut wood flour (CWF) was selected as the natural fibers in this research due to its abundant supply \[^{[3]}\]. CWF is also inexpensive, biodegradable and is not abrasive during the process \[^{[1,4]}\]. CWF also have high content of cellulose (40.99\%) which indication to be used in polymeric composites, as a material that has high content of cellulose would exhibit stiff and strong material \[^{[5]}\]. The dispersion and adhesion between the non-polar PP matrix and the polar CWF might be difficult. Therefore, chemical modification to CWF is needed to reduce its polarity. Compatibilizer, such as maleic anhydride-g-polypolypropylene (MAPP) could also be applied to improve the compatibility of the matrix with the reinforcement. In Indonesia, the production of plastic waste was second ranked as domestic waste producer at 5.4 million tons per annum (14\% of total waste production in Indonesia). As a matrix, polypropylene has good physical, mechanical, and thermal properties at room temperature. Its stiffness, low density, high impact strength and lower cost make it widely use in many applications \[^{[8]}\]. Lee et al. \[^{[9]}\] have studied hybrid composite polypropylene/wood flour/clay with MAPP with improved tensile strength and stiffness. In this study, the effect of MAPP addition on impact strength, water absorption and filler volume fraction of waste polypropylene (WPP) / Modified coconut wood flour (MCWF) / Glass Fiber (GF) Hybrid Composite was studied. Fourier Transform Infra-Red (FTIR) characterization and Scanning Electron Microscopy (SEM) analysis were used to show the morphology of the samples after modification.

EXPERIMENTAL CONDITION

Materials Preparation

Wasted Polypropylene (WPP) was collected from wasted mineral water plastic cup disposal. The WPP were cut into smaller pieces (5 mm x 5 mm). Coconut wood flour (CWF) was supplied from furniture store waste. CWF was dried under the sunlight and milled in a ball mill into 100 mesh particles. Chopped strand E-type glass fibers (GF) were supplied by PT. Justus Kimiaraya (Medan, Indonesia). The GF were milled in a ball mill into 100 mesh particles. Commercial xylene supplied by CV. Rudang Jaya (Medan, Indonesia) was used as received.
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To produce MAPP, isotactic polypropylene (melt flow index 14 g/10 min) supplied by Titan PP Polymers (TitanPro 6331 grade) (Johor, Malaysia), maleic anhydride, acetone and benzoyl peroxide (supplied by Merck Indonesia) were used as received.

The coconut wood flour was immersed in 18% solution of NaOH, heated at 90°C for half an hour. Followed by washing with distilled water to remove all traces of alkali until neutral, and then dried in tray dryer to diminish water absorption.

1 g of maleic anhydride, 10 g of polypropylene and 90 ml xylene were refluxed at 135°C for 20 minutes. 0.1 g of benzoyl peroxide which had been dissolved in 10 ml xylene were added into the solution and reacted for 10 minutes. The product were immersed in acetone to precipitate the MAPP. The filtered MAPP were then rinsed with distilled water until neutral pH was obtained. Finally, the MAPP were dried in a tray dryer for 24 hours, milled into 100 mesh particles and characterized by FTIR spectroscopy.

Sample Preparation
Composites were manufactured in a two-stage process. In the first stage, WPP, MCWF, GF and MAPP were premixed mechanically at certain formulations. The mixtures were then fed into a single-screwed extruder at a screw speed of 50 rpm and temperature profile of 180°C. In the second stage, the stranded extrudates were granulated manually and cooled in a water bath. The procured granules of extrudates were then dried at 105°C for 24 hours before being molded in a hot press machine at 180°C. The hybrid composites were produced with various contents of MCWF from 10 to 40 wt. %.

The GF loadings and MAPP addition were remained constant at 10 wt. % and 2% respectively. All formulations of the composite components and abbreviations used for the respective formulations were given based on the codes in Table 1.

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>WPP (%)</th>
<th>MCWF (%)</th>
<th>GF (%)</th>
<th>MAPP (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>100</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>8</td>
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<td>0</td>
<td>30</td>
<td>10</td>
<td>0</td>
</tr>
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<td>8</td>
<td>30</td>
<td>10</td>
<td>2</td>
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<tr>
<td>10</td>
<td>50</td>
<td>40</td>
<td>10</td>
<td>0</td>
</tr>
<tr>
<td>11</td>
<td>48</td>
<td>40</td>
<td>10</td>
<td>2</td>
</tr>
</tbody>
</table>

### Impact Strength
Izod impact strength for the specimens was determined having dimensions 60 x10 x 3 mm as per ASTM D-4812 - 11, with “V” notch depth of 2.54 mm and notch angle of 45°, using Impact Testing Machine. The conventional V notched specimens were according to ASTM D-256.

### Water Absorption Analysis
The specimens of dimension 25 x 25 mm (ASTM D 570) were tested. The samples were dried in an oven at 50±5°C for 24 hour, cooled in desiccators for 24 hours. Then, all the samples were immersed in a container of distilled water. At the end of 24 hour, the specimen...
were removed from water one at a time, all surface water was wiped off with a dry cloth, and they were immediately weighed to the nearest 0.01 g. The percentage of composite mass accretion during water absorption was determined. The water absorption of the test was determined with the following equation:

\[ \text{Percentage of mass accretion} = \frac{w - w_0}{w_0} \times 100 \]  

where \( w \) is the composite mass after \( t \) immersion days, \( w_0 \) is the composite mass before immersion.

### Filler Volume Fraction

Consider a composite material that consists of fibers and matrix material. The volume of the composite material is equal to the sum of the volume of the fillers and the matrix.

\[ V_c = V_f + V_m \]  

where \( V_c \) is the composite volume, \( V_f \) is the filler volume and \( V_m \) is the matrix volume.

The matrix volume fraction (Mvf) was measured with the formula:

\[ \text{Mvf} = \frac{V_m}{V_c} \]  

The filler volume fraction (Fvf) can be calculated by this equation:

\[ \text{Fvf} = 1 - \text{Mvf} \]  

Fourier Transform Infra-Red (FTIR) characterization and Scanning Electron Microscopy (SEM) analysis were considered to identify to show structural changes and morphology of fractured samples, respectively.

### RESULTS AND DISCUSSION

#### Fourier Transform Infra-Red (FTIR) Analysis

Figure 1. FTIR spectra of the synthesized compatibilizer, maleic anhydride-g-polypropylene (MAPP). It revealed the presence of absorbance in the wavelength number of 2955 cm\(^{-1}\) which indicated the existence of alkanes groups (C–H). Ketones groups (C=O) and C–O from anhydrides were also observed at 1712 cm\(^{-1}\) and 1165 cm\(^{-1}\) respectively. Moreover, the appearance of C–H from –CH\(_2\) (methylene) and C–H from –CH\(_3\) (methyl) were also determined in the peak.
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absorbance at 1454 cm\(^{-1}\) and 1369 cm\(^{-1}\). All of these peak absorbances numbers strongly supported the chemical structure of maleic anhydride-g-polypropylene presented in Figure 2.

Fig. 2. Chemical structure of maleic anhydride-g-polypropylene (MAPP)

From Figure 3, it is observed that alkaly modification had successfully decreasing the peak absorbance in the wavelength number of 3444 cm\(^{-1}\) and 2357 cm\(^{-1}\) (for –OH) and 1593 cm\(^{-1}\) (for N–H). It is also indicated the presence
of absorbance in the wavelength number of 2878 cm\(^{-1}\) and 2357 cm\(^{-1}\) which indicated the presence of –OH and N–H (the main functional group of cellulose) had been dispersed well into the matrix (WPP).

**Impact Strength**

Impact strength is a parameter that shows the maximum number of pressures caused by the high speed could be received by a material before it undergoes cracking. Results of this testing indicate the toughness, it is strongly influenced by the strength of the bond interphase and the characteristic of the matrix and fillers.\(^{10}\)

From Figure 4, the drop of impact properties is occurred to composites without MAPP (sample code 4, 6, 8, 10). It might be due to poor interfacial adhesion between the matrix and the fillers, which is a general phenomenon in incompatible composites with characteristics, such as non-polar of the PP and GF and the polar of CWF.\(^{6}\)

![Impact Strength Graph]

**Fig. 4.** Impact strength properties of hybrid composites as functions of the fiber loadings

However, addition of GF gave a significant effect in raising the impact strength of the composite, from 37.9 J/cm\(^2\) to 40.5 J/cm\(^2\) (for sample codes 2 to 5). Abdul Khalil et al.\(^{11}\) also reported a similar trend. This result is due to the high energy absorption capability of the glass fiber, indicating that the impact strength was observed when MCWF was added into composite from 49.5 J/cm\(^2\) to 40.5 J/cm\(^2\) (sample code 3 to 5). This 21%
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The rate of water absorption depends on the internal material status, nature of reinforcements, fiber-matrix interface, as well as environmental factors such as temperature and applied stress. The material shrinks or swells when incorporated wood absorbs water; this lack of dimensional stability is a major drawback of such materials. In this study, after the test period, the samples were dried with a cotton cloth, and reweighed.

Figure 5 shows the water absorption of hybrid composites. With the increased of MCWF loadings, the water absorption increased from 9.7% to 13.5% (sample codes 8 to 10). Sample code 2 (with formulation of 88/10/0/2)
and sample code 5 (with formulation of 78/10/10/2) resulted the same amount of water absorption due to negligible water absorption capacity of water impermeable glass fiber [13]. However, addition of MAPP had significantly reduced the water absorption (sample codes 10 to 11) since there is more compatibility and better adherence to the polymer matrix which covers more wood's surface and reduces the direct contact wood-water. It also increases the ester linkages between the hydroxyl groups of MCWF and the anhydride part of MAPP. Therefore, the amount of free OH– in the wood cellulose is reduced because some of them are interacting with succinic anhydride, so this change generates less water absorption, compared to the composite without MAPP [14].

**Table 2. Filler Volume Fraction of Hybrid Composite**

<table>
<thead>
<tr>
<th>Sample code</th>
<th>Composite ratio (WP/MCWF/GF/MAPP)</th>
<th>Filler Volume</th>
<th>Filler Volume Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>100/0/0/0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>88/10/0/2</td>
<td>12</td>
<td>0.125</td>
</tr>
<tr>
<td>3</td>
<td>88/0/10/2</td>
<td>12</td>
<td>0.126</td>
</tr>
<tr>
<td>4</td>
<td>80/10/10/0</td>
<td>20</td>
<td>0.224</td>
</tr>
<tr>
<td>5</td>
<td>78/10/10/2</td>
<td>22</td>
<td>0.203</td>
</tr>
<tr>
<td>6</td>
<td>70/20/10/0</td>
<td>30</td>
<td>0.318</td>
</tr>
<tr>
<td>7</td>
<td>68/20/10/2</td>
<td>32</td>
<td>0.303</td>
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<tr>
<td>8</td>
<td>60/30/10/0</td>
<td>40</td>
<td>0.421</td>
</tr>
<tr>
<td>9</td>
<td>58/30/10/2</td>
<td>42</td>
<td>0.413</td>
</tr>
<tr>
<td>10</td>
<td>50/40/10/0</td>
<td>50</td>
<td>0.522</td>
</tr>
<tr>
<td>11</td>
<td>48/40/10/2</td>
<td>52</td>
<td>0.502</td>
</tr>
</tbody>
</table>

This was due to the improved interfacial adhesion between the matrix and the fillers that causes the composite to have less void and higher density.

**Fracture Sample Analysis**

Figure 6 displays SEM micrographs of fractured composite which had the highest impact strength. In addition, the sample without MAPP had more voids in the composite. The increasing amount of MCWF which has lower density than the WPP and GF, resulting in a lower density of the composites also indicated composite with MAPP has lower fiber volume fraction due to the improved interfacial adhesion between the matrix and the fillers that causes the composite to have less void and higher density.
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It was clearly showed the uniform distribution of filler in WPP matrix, was due to the improved interfacial adhesion created by the MAPP. Without MAPP, fillers pull-out was observed. This was due to the improved interfacial adhesion that makes the stress transfer from the fillers to the matrix and consequently failed.

Adding MAPP had significantly helped in enhance the interfacial adhesion of the composite.

CONCLUSION

The utilization of MAPP as compatibilizer improved the impact properties and reduced water absorption of the composite. The highest impact strength was achieved in WPP/MCW/GF/MAPP ratio of 58/30/10/2.

Moreover, SEM micrographs showed that MAPP had significantly enhanced the interfacial adhesion between the matrix and the fillers.

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