LIQUEFIED TROPICAL WOOD/ POLYPROPYLENE COMPOSITES:
PREPARATION AND PHYSICO-MECHANICAL PROPERTIES

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Abstract. In this research, the effects of liquefied tropical wood (LTW) as reinforcing fillers in the thermoplastic polymer composite on the mechanical and physical properties were investigated. The LTW were prepared from tropical softwood species by using phenol as a reagent and sulphuric acid as a catalyst. Polypropylene (PP) as polymer matrix was used with LTW to make LTW/PP composites. The composites were prepared with 10, 20, 30, and 40 wt.% LTW using the hot-compression molding method. The effect of LTW on physico-mechanical properties of LTW/PP composites were evaluated using tensile test, three point bending test and water absorption test. It was found that the flexural modulus of elasticity (MOE) and Young’s modulus (E) increased due to the addition of LTW in the composites. In addition, the water absorption and thickness swelling coefficient increased with the LTW loadings.

1. Introduction
Currently, synthetic polymers are combined with many biodegradable reinforcing types of filler in order to increase the mechanical properties and gained biodegradable and other physical characteristics demanded in actual applications. The demand for materials that is unharneffull to the body and having the appropriate properties has increased due to a lack of resources and increasing environmental pollutions; thus, composites prepared from waste materials are actively being produced now [1]. From previous research it is found that, the use of lignocelluloses materials in the production of thermoplastic composites can give many benefits such as, improving the toughness and strength of virgin polymers, biodegradable, non-toxic and flexible. Furthermore, these materials can be easily obtained from waste products and have a minimal effect on the environment [2]. For these reason, natural fibres and lignocellulusic materials have been used in many application such as furniture, architectural materials, and more recently in the automotive industry [3]. Besides that, these composites can be easily processed through techniques such as injection and compression-molding [4]. Due to this benefit, continuous research development has been conducted in this area.

Liquefaction of wood is regarded as one of the best utilization of waste wood; hence Liquefaction of wood sawdust becomes one of the main thermochemical conversion processes for these materials [5]. It has been globally recognized and a large volume of interest has been applied to the conversion of the lignocelluloses waste into a substance which is soluble in organic solvent, with the addition of liquefying agent [6, 7]. Basically, LTW is produced after removing the liquefying agent and organic solvent [7]. This technique has been studied by many researchers for many years [8].
Biomass such as wood can be liquefied in the presence of phenol or polyhydric alcohols with an acid catalyst at a temperature of 120 °C to 180 °C or without a catalyst at approximately 250 °C [9, 10, 11]. Besides that, other factors that affect the LW such as liquefaction time, temperature, catalyst content, and the amount of reagent solvent on the liquefaction efficiency (i.e., extent of liquefaction) have been investigated. Additionally, some chemical and physical characteristics of the liquefied wood, such as molecular weight and its distribution, and thermal flow property have also been studied [12, 13, 14]. The liquefied products can be used to prepare polyurethane foams, phenolic resin, adhesives [14, 15], and as filler for polymer composites [16]. A broad understanding of the mechanism of wood liquefaction, which is important for improving the physical and mechanical properties of the final products of liquefied wood, has not yet been clearly recognized due to the complex structure and composition of wood. Lignocellulose materials such as LW offer many environmental benefits when used as reinforcing fillers for plastics, some benefits include it being lightweight, decreasing the erosion of the manufacturing equipment and it being relatively low cost [17]. Thus, in recent years, the emphasis has increasingly been placed on these composites that play a major role in solving some of the pressing environmental issues which we faced with in the future [17].

To improve the mechanical properties of plastics, inorganic materials and synthetic fibers were added to plastics as filler. The influence of LW on the mechanical and thermal properties of LTWPC has been investigated by previous research. It is found that the effect of LW was significant on the flexural and tensile properties [16]. The LW use in plastic showed similar effect of filler as wood flour or fiber for thermoplastic polymers [16]. However from the literature review, there are no works has been done on tropical wood as LTW to make composites. Thus in this report, we produced LTW from selected tropical wood and studied the effect on composites based on the loading levels. Now most of the biomass composites have been focusing on the use of PP as a matrix. This is because PP has a number of characteristic such as low price, high toughness and low density. Furthermore, PP can also be easily processed, recycle and improved through the use of glass fillers which have successfully bridged the gap between the commodity polypropylene composites and the engineering thermoplastics [18].

In this paper, was used PP as a polymer matrix and LTW as the lignocelluloses material for LTW/PP. The objective of this research is to investigate the mechanical properties of the LTW/PP and the potential of LTW as reinforcing filler at different loading.

2. Experimental

2.1 Materials. Five types of selected tropical softwood were used in this investigation (Eugenia spp, Artocarpus rigidus, Artocarpus elasticus, Koompassia malaccensis, and Xylopia spp). The wood was sawn using a laboratory table saw and the sawdust was collected. The sawdust with 300 mesh size was then oven dried at 70-80 °C to a moisture content of 3 - 5%, then stored in polyethylene bag until needed. The sawdust was blended to make a powder form. Polypropylene’s (PP which was used as polymer matrix, has a melt index of 0.28 g/10 min with a density of 0.938 g/cm³. PP was supplied by Korea Petrochemical Ind. Co., LTD.

2.2 Preparation of Liquefied Tropical Wood (LTW). The sawdust was mixed with phenol solution at a ratio of 1:3, sulphuric acid (H₂SO₄) was used as the catalyst and its content was 5 %, based on the amount of phenol. The mixture of LTW and phenol were heated in an autoclave for 4 hours with a temperature of 150 °C at a pressure of 150 Pa. Dissolute and residues were separated by a centrifuge and filtered through a glass filter and the LTW was recovered. A Rotary-vacuum evaporator was used to remove unreacted phenol
at 170-180 °C and finally the LTW was produced. Fig. 1 shows the physical appearance of LTW.

**Fig. 1.** Physical appearance of Liquefied tropical wood (LTW).

### 2.3 Manufacturing of Liquefied Tropical Wood Polymer Composites (LTW/PP).

The LTW/PP composites were prepared from LTW and PP. The LTW was mixed with PP in a beaker at 4 different ratios; 10 %, 20 %, 30 %, and 40 % by weight. The mixtures were stirred continuously until uniformly mixed with mixer and then pre heated in an oven for 24 hours at 80 °C to ensure the mixing is adequate. The mixture of LTW and polymer were stirred continuously until uniformly mixed and then dried in an oven for 24 hours at 80 °C. The mixture was compression molded into a sheet measuring 270 mm x 270 mm x 5 mm at temperatures 200 ± 5 °C. Then this molding board was cut to the test specimen size appropriate for each test. The molding conditions were as follows; pressure, 6.8 MPa, preheating time, 20 s; heating time, 45 min; and cooling under a slight pressure to ambient temperature [19].

### 2.4 Mechanical Properties.

In order to investigate the mechanical properties of the prepared composites the following (a) tensile (b) flexural test were performed using a Shimadzu Universal Testing machine (capacity of 300 KN) at a crosshead speed of 10 mm/min.

**Tensile test.** The static tensile tests were carried out following ASTM D 638-01 at a crosshead speed of 10 mm/min [20]. The specimen dimension is 148 mm, 10 mm, and 5 mm respectively. For each test, ten replicates samples were tested and the average values were reported.

**Flexural test.** Three points bending test were conducted following ASTM D 790-00 [21] to measure modulus of elasticity (MOE) and flexural strength. Three point flexural test set-up was used to perform this test. The specimen dimension is 79 mm, 10 mm, and 4.1 mm. For each test, ten replicates samples were tested and the average values were reported.

### Dimensional Stability.

The water absorption and thickness swelling test were conducted according to ASTM D 1037-99 [22]. Samples of each composite were soaked in water at ambient temperature. At each testing time, samples were removed from the water, wiped with tissue paper to removed surfaced water and then weighed. Each value obtained represented at the average of five samples. The water absorption and thickness swelling coefficient (TS) were calculated as follows:

Water absorption (%) = \[\left(\frac{W_2-W_1}{W_1}\right) \times 100\%\]  

where \(W_2\) is the specimen weight after soaking and \(W_1\) is the weight of sample before soaking. The thickness swelling coefficient (TS) is calculated as follows:
Thickness swelling coefficient (TS) = \[
\frac{(T_2 - T_1)}{T_1} \times 100 \%
\] (2)

where TS is the percent of thickness swelling; \( T_1 \) and \( T_2 \) are the thicknesses of the specimen before and after the test respectively.

**Density Determination.** All specimens were kept in the oven at 103 °C for 72 hours before density determination. Oven-dry density of each sample was then determined by using the Water Immersion Method.

**2.5 Micro Structural Analysis**

**Fourier Transform Infrared Spectroscopy (FTIR).** The infrared spectra of the raw and LTWPC specimens were recorded on a Shimadzu Fourier Transform Infrared Spectroscopy (FTIR) 81001 Spectrophotometer. The transmittance range of the scan was 4000 to 400 cm\(^{-1}\). The obtained spectra are described in the results and discussions section.

**Scanning Electron Microscopy.** The surfaces of the LTWPC were examined by a scanning electron microscopy (JSM-5510, JEOL Co. Ltd., Japan). The samples were sputter coated with gold and observed under the SEM using a voltage of 15 kV.

**3. Results and Discussion**

**3.1 Fourier Transform Infrared Spectroscopy (FTIR).** The formation of LTW with

![Fig. 2. Typical FTIR spectra of raw sawdust and LTW.](image)

Phenol was confirmed by FTIR spectroscopy. The first row shows the characteristic absorption band of raw sawdust. It was found that in the region show the existing of O-H absorption band at 3403 cm\(^{-1}\), C-H at 2903 cm\(^{-1}\) and C=O stretching carbonyl at 1735 cm\(^{-1}\) [23]. The absorption band at LTW shows increase at O-H which shifted towards 3349 cm\(^{-1}\) and at C-H the absorbance divides into two which at 3046 cm\(^{-1}\) and 2959 cm\(^{-1}\). It can be seen that, the carbonyl peak C=O at 1735 cm\(^{-1}\) was disappear in the spectra of LTW because the ester carbonyl bonds in the hemicellulose was break due to the liquefaction process [5]. This
phenomena were the peak at 1735 cm\(^{-1}\) disappear in the LTW was very similar with the result of wood or natural fiber being treated with sodium hydroxide (NAOH) [24, 25]. The region between 1800 to 1100 cm\(^{-1}\) comprises band assigned to the main components of wood which is cellulose, hemicellulose and lignin. Clear difference can be observed in the absorbance spectra both in the absorbance values and shape of the peak in their location. In addition, peak in the region 1229 cm\(^{-1}\) exists is due to the –OH plane deformation [26] indicating that this peak might related to the phenol-lignin complex bonding. However this peak was not appear in the raw wood. The spectra were found significantly changes and exist in the regions 1596 cm\(^{-1}\), 1510 cm\(^{-1}\), and 1229 cm\(^{-1}\) were related to the mechanism and characteristic of LTW. All these result confirmed the formation of LTW by the liquefaction process with the phenol and raw sawdust.

3.2 Scanning electron micrographs (SEM). Figure 3 shows the surface morphology of the composites at different LTW loadings respectively. From the observation, all composites at a different LTW loading of 10 wt.% up to 40 wt.% showed a few particles on the surface which are filler from the LTW. The surface morphology view of all composites clearly shows that the LTW is evenly dispersed in the PP matrix. The composites surface is covered with a layer of the uneven substance, which is probably LTW. This layer was also observed by other researcher [27]. At 10 wt.% LTW loading (Fig. 3a), a few filler particles are seen. Meanwhile at 20 wt.% LTW loading (Fig. 3b), the filler particles slightly increased and uneven layers also are seen. Further, at 30 wt.% LTW loading (Fig. 3c) more traces is seen. At 40 wt.% LTW loading (Fig. 3d), many voids are seen on the surface which can contribute to the weak mechanical properties. The large amount of poor bonded interfacial area between filler and matrix polymer increased at 40 wt.% LTW loadings. These voids are called as a defect in hot press moulding composite. The uneven layer and voids exist is due to the poor interfacial bonding between LTW and PP [28]. Figure 4 shows the voids exist on the surface on the LTWPC at 40 wt.% LTW. At 40 wt.% LTW, there are clear many voids or empty spaces the interface, means that LTW/PP composites adhesion is not good.
3.3 Mechanical Properties of LTW/PP Composites. The flexural properties for LTW/PP composites with various LTW loading levels were reported in Fig. 5. Tensile properties are presented in Fig. 6.

**Flexural Properties.** Figure 5 shows the flexural strength and flexural modulus of raw polymer and composite with contains liquefied tropical wood (LTW) at different loadings. The results show that the flexural strength decreased with the addition of LTW from 10 to 40 wt.%. The flexural strength of composite at 10 wt.% LTW loadings is found at 46.97 MPa which is the highest among all ratios, which is about 11.7 % from the raw polymer. The decrease of flexural strength from 10 - 40 wt.% LTW loadings is in the range of 11 - 23%, respectively. It is observed that, the flexural strength depends on the strength properties of polymer itself, instead of LTW. This shows that, with the small incorporation of small amount of LTW, at 10 wt.% loading levels, the flexural strength of the composite is lowest than the raw PP. When the LTW loading increases, the flexural strength gradually decreases. This is due to the reduction in resistance to shearing in the composites structure probably because the presence of the LTW.

The flexural modulus of composites increases as LTW loading level increases from 10% to 40% (Fig. 5). The increase of flexural modulus of composites by the addition of LTW is in the range of 16.5 – 47 %, due to the improvement of the interfacial bonding between the filler and the matrix. Similar results have also been reported by other researchers with different reinforcement materials [29, 30, 31]. Composite with 40 % LTW loading shows the highest flexural modulus respectively. It means the increase of flexural modulus is related to the increase of stiffness of LTWPC, due to the addition of liquefied tropical wood [30].

**Tensile Strength.** Tensile properties for LTW/PP composites are reported in Fig. 6. From Fig. 6, the maximum tensile Strength (29.85 MPa) and Young’s modulus (996.5 MPa) of composites are observed at 10 % and 40 % LTW loading level. In Fig. 6 the histogram shows that the tensile strength decreases as the LTW loading increases respectively. As the LTW loadings increase, therefore increasing the interfacial area, the falling interfacial bonding between filler (hydrophilic) and matrix polymer (hydrophobic) decrease the tensile strength, which nevertheless within acceptable levels of strength [32]. The decreasing in tensile strengths of the composites at high LTW loadings especially at 40 % LTW because at these particular compositions, the composites cannot effectively transfer the load during the test. This is in agreement with other researcher [4], where the decrease of tensile strength shows that the LTW gives no significant reinforcement with the PP [29]. Figure 6 shows that, the Young’s modulus of LTW/PP composites increases respectively when 10 % to 40 % LTW loading is added. It is observed that the increased start from 2.63 % to 5.5 %. Raw polymer shows the lowest Young’s modulus. The LTW improves the Young’s modulus of composites.
The resistance of the materials to a deformation has increased. The Young’s modulus increase with LTW loadings is in accordance with other reported work [28-30, 33]. During tensile loading, partially separated micro-spaces are created, which block the stress propagation between the LTW and the matrix. As the fibre loading increases, the degree of obstruction increases, which in turns increases the stiffness. Young modulus improves with increasing filler loading [34, 35].

![Graph](image1)

**Fig. 5.** Flexural properties of composites at different LTW loadings.

![Graph](image2)

**Fig. 6.** Tensile Properties of composite at different loading of LTW.

### 3.4 Physical Properties of LTW/PP composites

The effect of LTW loading level on water absorption and thickness swelling of the composites are listed in Table 1. Water absorption and thickness swelling increase with increase of LTW loadings.
Table 1. Physical performance of composites.

<table>
<thead>
<tr>
<th>LTW (%)</th>
<th>Density (g/cm³)</th>
<th>Water absorption (%)</th>
<th>Thickness swelling (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.841</td>
<td>0.08</td>
<td>0.00</td>
</tr>
<tr>
<td>10</td>
<td>0.784</td>
<td>2.80</td>
<td>0.24</td>
</tr>
<tr>
<td>20</td>
<td>0.782</td>
<td>3.20</td>
<td>0.93</td>
</tr>
<tr>
<td>30</td>
<td>0.759</td>
<td>5.13</td>
<td>1.48</td>
</tr>
<tr>
<td>40</td>
<td>0.727</td>
<td>5.68</td>
<td>1.66</td>
</tr>
</tbody>
</table>

**Thickness swelling test.** Figure 7 shows the values of the thickness swelling coefficient for the composites which vary depending upon the LTW loading. The value increases as the LTW loading level increases. The raw PP shows no change in thickness swelling because the nature of the materials which is hydrophobic.

Fig. 7. Thickness swelling of the LTWPC at different filler loadings.

From Table 1 and Fig. 7 it is observed that the thickness swelling coefficient for the composites at 40% LTW loadings exhibit the highest values i.e. 1.66%. It is observed that at 20 - 40 wt.% LTW loadings the values are closed each other. It is suspected that the composite is already saturated (optimum) at 20 wt.% loadings level. This fact indicates that there is high porosity or the present of voids on the surface of the composites at 20 - 40 wt.% LTW loadings level. Composites containing 10 wt.% LTW exhibit less thickness swelling at 0.24%.

**Water absorption.** Figure 8 shows the water absorption characteristic for the composites against filler loading. The water absorption (%) for the raw polymer and composites, were varies depending on the LTW loading. The water absorption of composites increased with LTW loading [36]. This is due to the higher contents of filler loading in the composites that can absorb more water [35]. When LTW loading increased, the formation of agglomerations between the matrix PP and LTW increased. This is due to the difficulties of achieving a homogeneous dispersion of filler at high filler loading. The agglomeration of the filler in composites increases the water absorption of the composites. It is expected because cell wall with hydrophilic hydroxyl groups will absorb water to its surface through the
formation of hydrogen bonding [37]. LTW is very hydrophilic material due to the large amount of hydroxyl groups in the major wood components. Hence it can be assumed that the LTW retained the hydrophilic characteristic of wood. From the result, composites with 10 % LTW had least water absorption, whereas at 40 % LTW the composite achieved the highest water absorption. Composite with high LTW contents (40 %) also exhibit highest water absorption because of the increase number of micro voids on the surface which is caused by the bigger amount of poor bonded area between the hydrophilic LTW and hydrophobic matrix polymer. Thus water is easily entered through these voids [17].

![Fig. 8. Water Absorption (wt.%) by LTWPC after 16 days soaking in water.](image)

**Density of LTWPC.** The density of composites decreases with the LTW loading (Table1). The density of raw polymer PP is higher (0.841 g/cm³) than other composites. Whereas at the same time the densities of LTW/PP composites at 10 wt.%, 20 wt.%, 30 wt.%, and 40 wt.%. LTW are at 0.784, 0.782, 0.759, and 0.727 g/cm³ respectively which are lower than raw polymer. This shows that there is voids present inside the composites, thus the LTW may leak out during the processing step [38]. Further when the LTW loading level increases, the numbers of voids increase as more LTW leak out and create void inside the composites. The SEM (Fig. 4) shows many voids inside the composites respectively.

**4. Conclusions**

In the present study, LTW was prepared from selected tropical wood species using phenol as a reagent and sulphuric acid as the catalyst. LTW/PP composites were manufactured by hot-compression moulding method. The LTW inside the polymer composites had improved the mechanical properties of LTW/PP composites. The result showed increased in flexural modulus and Young’s modulus of LTW/PP due to the addition of LTW. However, the flexural strength and tensile strength of the composites decrease slightly as the LTW increases. The composites have an acceptable strength up to a filler loading at 40 wt.%. The water absorption and thickness swelling coefficient of the composites increase gradually with the LTW loading but also to a negligible extent. LTW has a potential to be utilized as biodegradable filler for end user in polymeric materials, to minimize environmental pollution, rather than produce strong reinforcing filler.
Acknowledgements
The authors would like to thank the University Malaysia Sarawak (UNIMAS) for providing
the sponsorship under research FRGS grant (UNIMAS/ FGRS/02(20)/741/2010(27) that has
made this work possible.

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