THE EFFECTS OF RATTAN FILLER LOADINGS ON PROPERTIES OF RATTAN POWDER-FILLED POLYPROPYLENE COMPOSITES

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This study investigates the effects of filler loading on the properties of rattan powder-filled polypropylene composites. The composites were prepared by incorporating rattan powder of average size 180 µm into polypropylene matrix using a Polydrive Thermo Haake internal mixer. Filler loadings of the rattan powders ranged between 0 and 40 parts per hundred parts of resin (phr). Mechanical, morphological, and thermal properties were studied. The tensile strength, elongation at tensile failure, and impact strength decreased, while stabilization torque, thermal stability, and water absorption increased with increasing filler loading. Tensile modulus increased with addition of rattan powder and eventually decreased at 40 phr filler loading due to the weakening adhesion between the filler and the matrix. The morphological studies of fractured surfaces using SEM confirmed the deterioration in tensile properties.

Keywords:  Rattan filler; Polypropylene; Composite; Processing torque; Mechanical properties; Water absorption; SEM; TGA

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INTRODUCTION

Bio-fibres have long garnered interest as an alternative to synthetic fibres in fibre-reinforced composites. These fibres, mainly of agricultural and forest-produce sources, are known to be comparable in terms of strength per weight of material (John and Thomas 2008). Since natural fibres possess low specific weight and good stiffness, as well as being economical and eco-friendly, they provide a balance between the assets of a polymer matrix and the properties of synthetic fibres. In terms of the economy, cultivation of these natural fibre sources allows for a sustainable market, proving to be cost effective in these times of the ever-rising price of petroleum-derived feedstocks (Bismarck et al. 2006). With regard to commercialization, natural fibre-reinforced composites have been utilized mainly in automotive parts and in the construction sector, which has contributed to its rising reputation (Ashori 2008).

Forest-based materials previously regarded as waste have been penetrating the reinforced-plastic market in order to preserve the depleting source of wood. For example, sawdust has been utilized in producing composites as substitute for timber (Tajvidi and Ebrahimi 2003; Chen et al. 1998). Non-wood forest produce, such as rattan, holds a high value due to its importance and wide usage in the local economy. Due to its flexibility, combined with high strength, it has been traditionally used as binding materials and other household items (Weinstock 1983). Currently, it is extensively used in the furniture industry, and therefore it gives rise to a huge amount of waste in the form of discarded rattan poles. Its disposal is a concern since it may cause adverse effects to the environment. Shaari Balakrishna et al. (2012). “Rattan-PP composites,” BioResources 7(4), 5677-5690. 5677
environment due to its contribution to landfill space and its open burning (Muniandy et al. 2012). The impressive strength and flexibility of rattan has prompted interest in utilizing rattan pole waste as a filler for plastic composites.

As reinforced plastics, thermoplastics have been successfully employed in engineering applications. Natural fibre-reinforced thermoplastics are currently in common use in applications such as building, construction, furniture, and automotive products. Polypropylene (PP), being a polymer of good mechanical properties, is a popular choice as a base for a filled composite. Car manufacturers such as Daimler-Chrysler have opted for composites containing PP and plant fibres for use as interior trim components (Ashori 2008). Currently, numerous researchers have successfully utilized PP in natural or synthetic filler-reinforced composites using conventional methods, due to the ease of its blending operation, as well as versatility (Karmakar et al. 2007; Ansari and Ismail 2009a).

Many researchers have focused on the usability of cultivated fibre crops such as henequen, sisal, flax, jute, and hemp fibres (Tajvidi and Ebrahimi 2003). Meanwhile agro-wastes and agro-forest materials, which are abundantly present in developing countries, are also constantly being researched to provide an alternative to synthetic fibres and reinforcements (Thwe and Liao 2000). Non-wood forest produce, such as bamboo, have gained much attention as reinforcement (Chen et al. 1998; Wang et al. 2008). A few researchers have examined the utilization of rattan powder as filler in rubber composite, where according to the studies, the use of different filler loadings has resulted in deterioration of the tensile properties and fatigue life of the composite (Ismail et al. 2012; Muniandy et al. 2012).

From the best of our knowledge, studies of the incorporation of rattan filler into a thermoplastic matrix have not been reported. This study examines the use of rattan as filler in polypropylene composites, whereby the mechanical properties, fracture surface morphology, water absorption, and thermal stability of rattan powder-filled polypropylene composites were investigated.

**EXPERIMENTAL**

**Materials**

Polypropylene (TITANPRO homopolymer grade 6331) was supplied by Titan Petchem (M) Sdn. Bhd., Johor, Malaysia having a melt flow index of 14 g/10 min. at 230 °C and a density of 0.9 g/cm³. Rattan (Calamus manan) pole wastes were collected from Seng Huat Sdn. Bhd., Nibong Tebal, Penang, Malaysia.

**Preparation of Rattan Powder**

Rattan pole wastes were cleaned and ground into powder, followed by sieving to an average particle size below 180 µm. The rattan powder was then dried in oven at 80 °C for 24 hours to eliminate excess moisture.

**Composites Preparation**

Different contents of rattan powder were used in this study according to the formulation in Table 1. The composites were prepared by melt-mixing using the Haake Rheomix Polydrive R 600/610 internal mixer at 180 °C, with rotor speed of 50 rpm.
Polypropylene (PP) was loaded into the mixing chamber and allowed to melt for 2.5 min., followed by addition of rattan powder within 30 s. Mixing was continued for 4 min., up to a total mixing period of 7 min. The composites were then moulded into a 1 mm-thick sheet using a Go-Tech compression moulding machine type KT-7014-A. The upper and lower moulds were set to heat at 180 °C. Composites were pre-heated for 7 min. and compressed under hydraulic pressure of 1000 psi for 3 minutes, followed by cooling at 1000 psi for 2 min. The samples were cut into shape according to the requirements of tests conducted in this study.

**Table 1. Blend Composition of Rattan Powder-filled Polypropylene Composites**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Designation</th>
<th>PP (phr)</th>
<th>Rattan Powder (phr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>PP</td>
<td>100</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>PP/R10</td>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>3</td>
<td>PP/R20</td>
<td>100</td>
<td>20</td>
</tr>
<tr>
<td>4</td>
<td>PP/R30</td>
<td>100</td>
<td>30</td>
</tr>
<tr>
<td>5</td>
<td>PP/R40</td>
<td>100</td>
<td>40</td>
</tr>
</tbody>
</table>

**Tensile Test**

This test was carried out in accordance with the American Society for Testing Materials (ASTM) D 638 using a Universal Testing Machine (UTM) Instron Model 3366 with a cross-head speed of 50 mm/min. at 25 ± 3 °C. Samples were cut from the moulded sheet into 1 mm-thick dumbbell specimens. Tensile strength, tensile modulus, and elongation at tensile failure were obtained from the average results of five specimens.

**Impact Test**

This test measured the energy absorbed by a standard specimen while fracturing under an impact load. The un-notched Charpy impact test was carried out using an Impact Pendulum Tester (Zwick) under a load of 7.5 J. An average was reported from results of five specimens for each composition.

**Morphological Study**

Examination of the fracture surfaces of tensile specimens was done using Scanning Electron Microscopy (SEM, Zeiss Supra 35vp, Germany). SEM micrographs were taken at various magnifications. The fracture surfaces were sputter-coated with gold prior to SEM observations to avoid electrostatic charging during examinations.

**Water Absorption**

Specimens were dried in an oven for 24 hours at 50 °C until a constant weight was reached. Samples were weighed to the nearest 0.001 g prior to immersion in distilled water. Weighing was repeated periodically using a Radwag balance model AS 310/X with precision of 0.1 mg. Percentage of moisture content at time $t$, $W_t$ (%) was calculated using Eq. 1 as follows,

$$ W_t \, (\%) = 100 \times \frac{w_w - w_i}{w_i} $$  \quad (1)

where $w_i$ and $w_w$ are initial dry weight and wet weight after immersion at time $t$, respectively.
Thermogravimetric Analysis

Thermogravimetric analysis (TGA) was carried out using a Perkin Elmer Pyris 6 TGA analyzer. Thermogravimetric analysis was assessed by heating samples from temperature of 30 °C to 600 °C at a heating rate of 20 °C/min. Tests were done under nitrogen flow of 50 mL/min., and the samples used were of 5 to 10 mg in mass.

RESULTS AND DISCUSSION

Processing Torque

Figure 1 displays the mixing torque versus time data for the rattan powder filled-PP composites with various filler loadings. The initial mixing torque rapidly increased, which can be attributed to the shearing action toward the solid PP. The continuous shearing and high temperature due to mixing friction caused the PP granules to melt and provided less resistance, resulting in a lower torque reading. Another peak was observed at 3 min. into mixing, at which point the rattan powder was added to the molten PP. This caused the viscosity of the blend to increase due to the increasing dispersive resistance provided by rattan filler. It is also observed that higher loading of rattan powder displayed higher torque value readings due to the increasing viscosity. Torque values decreased gradually as the dispersion of the filler increased in the homogenized mixture, causing lower resistance of flow.

![Figure 1](image)

**Fig. 1.** Mixing torque development for rattan powder-filled PP composites with increasing filler loading during processing

Figure 2 shows the stabilization torque at 7 min., which clearly displayed an increase in torque value as soon as filler is added into the composite. This trend was also observed in a study by Othman et al. (2006). The increase in stabilization torque with filler loading can be attributed to the reducing ability of the matrix to flow due to decrease in PP polymer chain mobility, which increased the viscosity of the composite.
Fig. 2. Stabilization torque at 7 min. for rattan powder-filled PP composites with increasing filler loading

**Tensile Properties**

Figure 3 shows the effect of rattan filler loading on the tensile strength of rattan powder-filled PP composites. The addition of filler reduced the tensile strength, which gradually decreased with increasing filler loading. Similar observations were reported by other studies using natural fillers (Demir et al. 2006; Yang et al. 2004). The hydrophilic nature of the rattan powder, in contrast with the hydrophobic nature of the PP matrix, caused incompatibility between the matrix and the filler. Therefore, stress cannot be efficiently transferred across the matrix-filler interphase.

Fig. 3. Effects of different filler loadings on tensile strength of rattan powder-filled PP composites. Error bars represent the standard deviation of the measurements.

The presence of hydroxyl groups in the amorphous regions of the fibre hindered its ability to develop adhesion with non-polar materials (Mwaikambo and Ansell 2002). Also, higher filler content resulted in higher filler-filler interaction, leading to the agglomeration of rattan filler within the PP matrix. These agglomerates indicated that the wettability of rattan powder by the matrix was reduced, resulting in poor tensile stress transfer. This will be shown later in the Morphological Study section with the SEM micrographs (e.g., Figs. 6(d) and 6(e)). Furthermore, the interaction between rattan-rattan particles became more pronounced than the rattan-PP interaction with increasing filler content (Yew et al. 2005).

Figure 4 shows the tensile modulus of the rattan powder filled-PP composites with different filler loadings. Incorporation of filler increased the tensile modulus of the composite by increasing the composite’s stiffness. The presence of fillers restricted the polymer chain mobility of PP matrix, adding to the rigidity of the composite (Santiagoo et al. 2011; Sam et al. 2009). The stiffness of the composite can also be attributed to the cellulose content of the rattan filler (John and Thomas 2008). However, at 40 phr of filler loading, the tensile modulus showed no significant change in value. This may be due to poor wetting of the fillers with increasing filler content, therefore causing the inability of the filler to impart its inherent stiffness to the PP matrix. The Young’s modulus of the composite corresponded to the intrinsic properties of the filler, where the filler may exhibit its high stiffness (Supri et al. 2011). The effect of poor fibre wetting by the matrix was demonstrated by the easy detachment of the fibres from matrix (see Morphological Study Section (SEM micrograph of Fig. 6(e))).

![Fig. 4](image_url). Effects of different filler loadings on tensile modulus of rattan powder-filled PP composites. Error bars represent the standard deviation of the measurements.

The effect of filler loading onto the elongation at tensile failure of the rattan powder filled-PP composites is shown in Fig. 5. Due to the increasing filler content, stress transfer became increasingly poor as the adhesion between the filler and the matrix...
decreased. The tendency of the formation of agglomerates was much higher due to the filler-filler interaction. These agglomerates can develop into stress-concentrated areas, resulting in a catastrophic failure of the composite. Agglomeration may lead to stress-concentrated points, which are able to initiate cracks in the composite (Ansari and Ismail 2009b). Also, the poor adhesion between rattan filler and PP matrix can cause poor stress transfer, resulting in a brittle behaviour of the composite. This caused the elongation at tensile failure of the composites to decrease with increasing filler loading. Similar trends were observed by other researchers (Zaini et al. 1996; Tajvidi and Ebrahimi 2003).

![Fig. 5. Effects of different filler loadings on elongation at tensile failure of rattan powder-filled PP composites. Error bars represent the standard deviation of the measurements.](image)

**Morphological Study**

Figures 6(a) and 6(b) show the SEM micrographs of the tensile fracture surfaces of PP matrix, while Figs. 6(c) through 6(e) show the tensile fracture surfaces of rattan powder-filled PP composites. The area of interest would be the presence of fibre pull-outs in the composites. In Fig. 6(a), the fracture surface of pure PP showed no presence of voids due to absence of rattan filler. Figure 6(b) shows a higher magnification of the PP matrix. It was apparent that there were fibrous ligaments on the surface of the fracture. This implied that the stress was efficiently transferred throughout the matrix.

Figure 6(c) shows the tensile fracture surface of the PP/R10 composites, where minimum voids were present due to the low fibre content. It also showed the presence of tearing in fibre, which indicated that the stress was successfully absorbed by the fibre, signifying a fibre failure occurrence. As a result, the performance of PP/R10 composites in tensile properties was better than composites with higher filler loading.

Figures 6(d) and 6(e) shows the tensile fracture surfaces of PP/R30 and PP/R40 composites, respectively. It can be seen that the presence of rattan filler was greater in these composites, as compared to Fig. 6(c). Therefore, the occurrences of fibre detacments and voids were more frequent in PP/R30 and PP/R40 composites compared to
PP/R10 composites. Fibre pull-outs occurred due to the weak adhesion between the filler and the matrix.

Fig. 6. SEM micrographs of tensile fracture surfaces of: (a) PP matrix at magnification of 100x; (b) PP matrix at magnification of 500x; (c) PP/R10 composite; (d) PP/R30 composite; (e) PP/R40 composite; and (f) non-uniformity of rattan particles.
The increasing presence of fibre pull-out voids with increasing filler content indicated that higher filler content caused poor wetting of the fibres. The larger voids indicated that filler particles of larger sizes created bigger non-bonding areas, so the gaps were much more visible and were more able to serve as points of initiation for failure. Small cellulosic particles created small non-bonding areas, so the gaps were not very visible. Poorly bonded interfacial areas between filler and matrix, in the form of gaps, were present as a result of the difference of polarities between the filler and the PP matrix. The non-uniformity of filler particle sizes is illustrated in Fig. 6(f).

Impact Strength

Effects of rattan filler loadings on the impact strength of rattan powder-filled PP composites are shown in Fig. 7. The addition of rattan filler caused a sudden decrease in the impact strength of the composites. This can be attributed to the poor interfacial adhesion of the filler and the matrix, causing poor distribution of stress throughout the composite. Poor interfacial bonding induced micro-spaces between the filler and the matrix polymer, therefore causing numerous micro-cracks when impacted; these micro-fractures propagated cracks easily and decreased the impact strength of the composites (Yang et al. 2004). Higher filler content reduced the impact resistance of the composites. The fillers act as stress concentrators, which can initiate the fracture of the composite while a load is being applied. Bledzki and Faruk (2004) observed a similar trend with wood fibre-reinforced polypropylene composites.

![Impact Strength Graph](image)

**Fig. 7.** Effects of different filler loadings on impact strength of rattan powder-filled polypropylene composites. Error bars represent the standard deviation of the impact strength measurements.

Water Absorption

Figure 8 presents the water absorption of rattan powder-filled PP composites with different filler loadings. Results showed that water uptake gradually increased with increasing filler content, reaching a saturation point where the moisture content remained...
constant. The hygroscopic nature of rattan filler was due to the presence of hydroxyl groups. These hydroxyl groups increased with increasing rattan filler content due to increasing cellulosic content; therefore formation of hydrogen bonds between the fillers and the water molecules increased. Also, the poor adhesion associated with high filler content may have caused an increase in water uptake. As reported by Demir et al. (2006), poor adhesion caused cracks and voids between the matrix and fibre, which resulted in easy penetration and storage of water through the voids.

**Fig. 8.** Percent increase in weight after water absorption as a function of time of immersion, for different filler loadings

**Thermogravimetric Analysis**

Figure 9 displays TGA thermograms of rattan powder filled-PP composites with varying filler loadings, while Table 2 shows the detailed variation of TGA experimental data of rattan powder filled-PP composites. Samples of the lowest and highest filler content in this study, designated as PP/R10 and PP/R40, respectively, were tested and evaluated against neat PP.

The derivative thermogravimetric (DTG) curves in Fig. 10 showed single-stage degradation for neat PP, whereas three-stage degradation was exhibited by PP/R composites. Between 250 °C to 450 °C, a distinctive weight loss was observed in PP/R composites, corresponding to the thermal degradation of rattan filler. The first and second peak of the PP/R composites (Fig. 10), observed at 310 °C and 358 °C, respectively, can be attributed to the decomposition of hemicellulosic and cellulosic constituents. Panthapulakkal and Sain (2007) indicated in a study that decomposition of cellulosic and hemicellulosic components of natural fibres occurred around 250 to 400 °C. Due to its filler content, PP/R10 showed a less evident weight loss when compared to PP/R40. The final decomposition peak corresponded to the decomposition of PP.
It was observed that the thermal decomposition of PP/R10 and PP/R40 shifted to higher temperatures as compared to neat PP, indicating an increase in thermal stability. As the composites reach the temperature at maximum rate of weight loss ($T_{\text{max}}$), the thermal stability showed an increasing trend with filled composites, as shown in Table 2. The decomposition of cellulose at high temperatures produced carbonaceous residues known as char, which forms a barrier between the heat source and the polymer material (Beyler and Hirschler 2001). As the char layer thickened, thermal decomposition of PP/R
composites was most likely reduced. From Table 2, the percent char residue of the composites increased with increasing filler loading. The char residue may correspond to the cellulosic and lignin content of the composites, which was higher in PP/R40 than PP/R10. According to Abdullah et al. (2010), lignin decomposed at a low rate from 250 °C to 500 °C due to its thermally stable nature, therefore contributing to the percent char residue. The thermal degradation for pure PP showed no char residue, which indicated that PP decomposed completely at high temperatures. Rosa et al. (2009) reported that thermal decomposition of PP leaves a very low residual mass due to formation of volatile compounds by the hydrogen and carbon atoms present in PP chains.

Table 2. TGA Data for Rattan Powder-filled Polypropylene Composites

<table>
<thead>
<tr>
<th>Sample Designation</th>
<th>$T_{max}$ (°C)</th>
<th>Char Residue (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP</td>
<td>455</td>
<td>0</td>
</tr>
<tr>
<td>PP/R10</td>
<td>474</td>
<td>0.241</td>
</tr>
<tr>
<td>PP/R40</td>
<td>480</td>
<td>5.958</td>
</tr>
</tbody>
</table>

CONCLUSIONS

The main conclusions drawn from this study were as follows:

1. Increasing the amount of rattan powder content in polypropylene resulted in higher mixing and stabilization torque due to an increase in viscosity.
2. Higher rattan powder content in polypropylene lowered the tensile strength, the elongation at tensile failure, and the impact strength of the composite due to the poor interfacial adhesion between the filler and the matrix.
3. The tensile modulus increased with increasing filler content, and it decreased at 40 phr of filler as the wettability of filler by PP matrix was reduced.
4. Deterioration of tensile properties was confirmed by the SEM images.
5. Water absorption by the composite increased with increasing filler loading due to increasing presence of hydroxyl groups in the composite.
6. Thermal stability of the rattan powder-filled PP composites increased with increasing filler loading due to formation of char residue.

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REFERENCES CITED


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