MECHANICAL AND STATISTICAL ANALYSIS OF POLYPROPYLENE COMPOSITES DERIVED FROM MIXED CLAY-OIL PALM FRUIT PARTICLES

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ABSTRACT

The work presents a combined experimental and analytical study of the influence of clay mixed with oil palm fruit and oil palm wood particles as reinforcements in polypropylene composites. Sample morphology was characterized via Scanning electron microscopy (SEM), and the mechanical response determined via tensile and compressive testing. Statistical modeling was employed to validate the experimental results. SEM images show the particles are uniformly dispersed in the polypropylene matrix. The polypropylene matrix tensile strength increases with increasing amounts of reinforcement, with highest reinforcement seen for oil palm wood particles of 1.51 MPa. The implications of the results are discussed to enhance the understanding of the clay-oil palm wood particulates as potential reinforcement materials for structural and constructional composites fabrication.

Keywords: clay, mechanical properties, statistical analysis, SEM and composites.

1. INTRODUCTION

The design and fabrication of composites with unique properties has necessitated the search for environmentally sustainable materials which exhibit multiple properties with a wider range of application. These materials have become the holy grail of polymer nanocomposite science [1-3]. Increasingly, the use of single nanofillers or nanofibers is unsatisfactory in meeting today’s multifaceted critical application challenges where low cost, high toughness, weathering resistance, high strength, light weight, flame retarding capability and biodegradability etc., are desired in a single nanocomposite [4-5]. Also, the inclusion of two or more nanofillers of biomass origin in the recycled polymer matrix does not only bring a marked and comprehensive improvement in the properties of polymer nanocomposites due to the synergistic interaction between the individual nanofillers and its host (recycled polymer matrix), but the added advantage of (1) removing from the environment, non-biodegradable and persistent plastic waste and (2) the addition of a biodegradable biomass nanofillers into the recycled polymer matrix increases the biodegradability of the recycled nanocomposite leading to an increase in its environmental appeal [6-9].

Currently, the by-product of oil palm is underutilized and the rapid expansion in the oil palm plantation sector has generated a colossal amount of vegetable waste which creates problems in waste management leading to serious environmental concerns [10]. Therefore, the incorporation of biomass such as oil palm wood dust, rice husk, and oil palm fruit fibers into the recycled polymeric matrix could improve its properties by tailoring them to a wide range of applications such as green furniture, aerospace, building construction, and automotive industries thereby increasing the economic value of these nanocomposites [11, 13].

Oil palm is one of the major cash crops in Ghana. At some point in the production process, lignocellulosic biomass in the form of oil palm trunks (OPT), oil palm fronds (OPF), empty fruit bunches (EFB) and unprocessed fruits etc. are obtained. For every kilogram of obtained palm oil, approximately 4 kg of dry biomass is also produced [14, 15]. Recent studies show that, the empty fruit bunches (EFB), fruit fibers, fronds and even the wood when processed into the form of flour, may be used as reinforcement agent in polymer composites [10].

However an obvious drawback of natural wood fillers or unmodified wood fillers arises from their propensity to absorb or desorb water in relatively humid environments. This tendency to incorporate moisture into their structure stems from the numerous hydrophilic functional groups (hydroxyl groups) which are innate bound to the natural structure of the cellulose fibers as well as the hemicellulose and lignin matrix of the wood [16]. The absorbed water could affect the dimensional stability and mechanical properties of wood reinforced recycled polypropylene nanocomposites, particularly in tropical climates where humidity is high [17, 18]. Chemical modification of wood fibers using various chemical functionalities such as carboxylic anhydrides, quaternary ammonium salts, [19] and acetylation systems have been proposed to overcome the hydrophilicity of the wood fillers [20-23]. However functionalization often requires several steps, coupled with the use of expensive reagents and laborious purification processes which makes the final product expensive to consumers [24]. The use of compatibilizers and other cheap coupling agents such as clay together with wood fillers can reduce the
hydrophilicity and enhance the mechanical properties of wood-clay-polymer composites. It has been reported that, a significant increase in the rigidity, modulus and stiffness of clay reinforced composites are achieved when combined with biomass from palm trees [25, 26].

In this paper, the mechanical properties of oil palm wood flour (OPWF), oil palm fruit fiber (OPF) and clay (Abonko & Adawukwa) reinforced polypropylene composites are investigated in order to shed light on their application as biodegradable plastics and as lining for car pickups.

2. MATERIALS AND METHODS

2.1. Materials

Abonko and Adawukwa clays obtained from the central region of Ghana were obtained by grinding samples followed by sieving using 20-40 µm mesh to obtain fine particles.

Oil palm wood flour (OPWF) pieces were ball milled and sieved through 40-70 µm mesh size sieves.

Oil palm fruit fibers (OPF) were cut in the range of 2-5 cm, then soaked in 2% NaOH solution at 100°C for 30 minutes and then oven dried at 60°C for 24 hours.

Recycled polypropylenes (PP) were obtained from Polyplast, a polymer processing industry in Ghana.

2.2. Fabrication of composites

The composite samples were prepared in an open flame within a temperature range of 160-200°C by varying the weight ratios of the reinforcements as shown in (Table 1) below. Neat polypropylene was also prepared. Two metallic molds with dimensions (20 mm x 50mm) and (20 mm x 80 mm) were designed and fabricated for the casting of the composites. The PP and reinforcements were heated for 15 minutes after which the resulting mixture was carefully poured into the molds and allowed to cool to room temperature. 5g of polyvinyl alcohol were used as binder. Table 1 is a list of sample prepared and their mechanical properties.

2.3. Characterization of composites

2.3.1 Mechanical characterization

Pure PP (control) and composite specimens (dog-bone shaped) were characterized for their tensile properties, such as Young’s modulus, tensile stress and elongation at break, using a Force Gauge Model M5-500, with a cross-head speed of 10 mm/min. Tests were carried out at room temperature. Tensile properties were characterized according to ASTM D-638; at least five specimens were tested to obtain the average values (see Table-1).

Compressive test samples were made with cross sections of 900 mm². The Flexural Breaking Load Machine, MOR/5-TS/65 - 800 kg was used for both flexural and compressive tests. Tests were performed at a rate of 0.1N/mm’s. Compressive strength were calculated from the breaking load values and the cross sectional area of the samples.

Water of absorption test was done on composite samples by immersing samples in water bath containing distilled water for 24 hours. Five specimens from each batch with dimensions of 30 cm x 30 cm x 25 cm were cut from composite panels and measured as \( W_o \). Samples were dried for 24 hours and their masses measured as \( W_1 \). The water of absorption \( W_o \) was calculated using the equation (1.1) below;

\[
W_o = \frac{W_1 - W_0}{W_0} \times 100\%
\]  

Where \( W_1 \) is the mass of the sample after the immersion in water and \( W_0 \) is the mass of the dried sample after 24 hour period.

2.3.2 Scanning electron microscopy (SEM)

Morphological features of the composite samples were observed using a field emission gun scanning electron microscope (FEG-SEM), JEOL 5800. A thin layer of gold was evaporated on the samples in order to avoid charging when exposed to the electron beam.

Table-1 is the composition of the various reinforcements. There are variations in the fractional compositions of the OPWF and OPF in this work. This was due to the difficulty in measuring same quantities of the OPWF and OPF to that of the clays. Large quantities of OPWF and OPF will be needed if the results are to be compared with the fractional weights of the clays.
Table-1. Compositions of samples for tensile testing.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Sample code</th>
<th>PP (g)</th>
<th>FILLERS</th>
<th>OPF (g)</th>
<th>OPWF (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP</td>
<td>PP</td>
<td>40</td>
<td>ABONKO Clays (g)</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>PP + ABONKO CLAY</td>
<td>P1</td>
<td>40</td>
<td>15</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>P2</td>
<td>40</td>
<td>10</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>PP + ADAWUKWA CLAY</td>
<td>P3</td>
<td>40</td>
<td>-</td>
<td>15</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>P4</td>
<td>40</td>
<td>-</td>
<td>10</td>
<td>-</td>
</tr>
<tr>
<td>PP + OPF</td>
<td>P5</td>
<td>40</td>
<td>-</td>
<td>0.75</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>P6</td>
<td>40</td>
<td>-</td>
<td>0.5</td>
<td>-</td>
</tr>
<tr>
<td>PP + OPWF</td>
<td>P7</td>
<td>40</td>
<td>-</td>
<td>-</td>
<td>7.5</td>
</tr>
<tr>
<td></td>
<td>P8</td>
<td>40</td>
<td>-</td>
<td>-</td>
<td>5.0</td>
</tr>
<tr>
<td>PP + ABONKO + OPF</td>
<td>P9</td>
<td>40</td>
<td>5.0</td>
<td>-</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>P10</td>
<td>40</td>
<td>2.5</td>
<td>-</td>
<td>0.5</td>
</tr>
<tr>
<td>PP + ADAWUKWA CLAY + OPWF</td>
<td>P11</td>
<td>40</td>
<td>-</td>
<td>5.0</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>P12</td>
<td>40</td>
<td>2.5</td>
<td>-</td>
<td>2.5</td>
</tr>
</tbody>
</table>

2.4. Theoretical model formulation

Mathematical models were developed for the mechanical characterization of the polypropylene and its reinforcements, using multiple linear regression approach based on Statistical Package for Social Sciences (SPSS) software. Independent and dependent variables for the mechanical properties were considered. In analyzing the resulting model, the parameters ‘R,’ R², adjusted R², and F-statistic’ were used. ‘R’ is the multiple correlation coefficient and measures the quality of the prediction of the dependent variable; ‘R²’ is known as the coefficient of determination and is used in measuring the proportion of the variance in the dependent variable that can be explained by the independent variables; ‘Adjusted R²’ is an adjustment of ‘R²’, taking care of the addition of extraneous predictors to the model. The closer the values of these parameters are to 1, the more accurate the model is able to predict the experimental values. The F-statistic and its p-value are used to test whether the overall regression is a good fit for the data. The p-value for F-statistic (with an alpha of 0.05 and 2-tailed decision criteria) is used in testing the null hypothesis that all of the model coefficients are zero (0). Where the p-value for ‘F-statistics’ is less than 0.05, the coefficients of independent variables are significantly different from zero.

3. RESULTS AND DISCUSSIONS

3.1. Tensile test

The tensile behavior and ultimate mechanical properties are very important characteristics of polypropylene and their related composites. The mechanical properties of polymer-reinforced composites are highly correlated to the intrinsic properties, amount, dispersion of the fillers, the properties of the polymer matrix, and the interaction between filler and polymer matrix [10]. Studying the group of composites containing; (PP and clay samples), (PP, OPF and OPWF samples) and lastly (PP, OPF / OPWF and clay samples), it is clear that; there is an increase in tensile stress (Figure-2d). This was much better than the results reported by Zeini et al. [11]. PP had the lowest tensile stress of 0.37 MPa as compared to the composite samples, but the introduction of the reinforcing fibers and clay particles tend to increase the strength of the overall composite (Figure-2(d)).

Interestingly, combination of the three fillers (clays, OPF...
& OPWF) did not show any significant change in tensile stress and elastic modulus characteristics (Figure-2(d)).

In order to explain the behaviour of composites in relation to the tensile properties, we next turn our attention to SEM image characterization of the composite samples (Figure-1).

The composition dependence of stiffness and that of other properties studied, indicate that, the components are separately dispersed in most composites, similar to the work reported in ref. [13]. The micrographs showed rough surfaces. The nature of the morphologies of the dispersed components makes their structural analysis quite difficult. The structure of P1 in the micrograph in Figure-1 is similar to that of PP, suggesting that uniform mixing was achieved with clay nanoparticles, causing fracture at a higher degree of elongation. It is seen that, clay nanoparticles prefer to remain aggregated due to their large surface area, as seen in P4 of Figure-1. This compromises the mechanical integrity of formed composites that contain these nanoparticles or when in combination with other fillers. Even though all the SEM images of the composites were taken, we show only selected images (PP, P1 & P4) in Figure-1.

The morphology of P7 composite (PP/OPWF) shows large pores over a rough surface area which indicates that, water will be absorbed into the interstice, thereby confirming the results obtained see Figure-2(c). P10 and P11 showed relative adhesion between the PP, clay and fiber particles with some small pore-like structures and discontinuities (crack) running through the surfaces. This may affect the compressive abilities of the composite formed; hence complete embedding could not be verified. The remaining composites did not show any significant variation in their microstructure.

![SEM morphological images of neat PP with some selected reinforced composite samples. Magnification: 300x and bar = 20µm.](image_url)

**Figure-1.**

3.2. Compressive test

The results for the compressive strength test on the composite samples are shown in Figure.2 (a). The highest compressive strength from the composite samples was that of PP reinforced with Abonko clay and oil palm fruit fiber (P2, & P9). The lowest strength as compared to the neat PP was recorded by that of the fiber reinforced composite. Comparing the neat PP with the reinforced samples, it can be said that, clay, fiber and wood flour fillers improved the compressive strength of the composites.

3.3. Flexural test

Generally, the clay reinforced samples showed the highest modulus of rupture with a peak value of 0.452 MPa compared to other composites. Increase in clay contents in the composites showed an increase in the modulus of rupture. However, when wood flour was added to this composition, the modulus of rupture decreased significantly. This validates the results of the modulus of rupture test, with peak value recorded by the clay samples in Figure-2 (b).

3.4. Water of absorption test

The way in which composite materials absorb water depends upon many factors, such as temperature, fiber volume fraction, fiber orientation, fiber type, area of exposed surfaces, interfacial bonding, diffusivity, the reaction between water and matrix, surface protection and hydrophobic chains of the matrix [9].

Comparing the Polypropylene filled clay composite samples (P1 - P4), it is noted that, the water of absorption percentages are relatively low and comparable to the recycled polypropylene, see Figure-2(c). This may be attributed to the fact that polypropylene is hydrophobic whereas clay is hydrophilic and hence during processing, fewer voids are formed which reduces the volume of water absorbed by the composite material. P1 and P2 have similar composition but a decreasing clay (Abonko clay) content and P3 and P4 (Adawukwa clay) with decreasing clay content from 22.5 g to 15 g respectively.

The introduction of reinforcing fillers such as wood flour tends to increase the sensitivity of polypropylene towards water. Also, for the ranges (P5 - P8) which denote composite samples with polypropylene and oil palm fiber / wood flour, the water of absorption is relatively higher and the highest as shown in Figure-2 (e), is P7, which is 5.9%. The behavior is due to the fact that wood flour is highly hydrophilic [12] and the material processing route creates voids which allow the escape of water into the interior even after drying, giving rise to the higher percentage of water absorbed. Also, with decrease
in the amount of wood flour from P7 to P8 showed a decrease in the water absorbed confirming that wood flour is hydrophilic Figure-2 (c).

The other multi-phase polymer composites made from polypropylene, clay and oil palm fruit fiber / wood flour (P9-P12) also recorded low percentages of water of absorption, Figure-2 (c). The last groups containing all clay and fiber materials with polypropylene are composites with a mixture of hydrophobic and hydrophilic attributes, resulting in a balance in water absorbed.

Figure-2. Graphical plots for pure pp and reinforced composites, with; a) compressive stress, b) breaking strain and load, c) water of absorption and d) tensile stress

3.5. Results of theoretical models

In order to validate the experimental results, mechanical characteristics were tested against a range of values with their limit of application and symbols ‘T, C, F, and W’ with their corresponding values given in Table-2. To estimate accurate dependent variable values from the models above, the independent variable values substituted into the model, must fall within the range of values provided in Table-2. With the exception of models for ‘Elongation (%)’ and ‘Strain’, which had R² values less than 0.5, all other values were close to 1.0 and can therefore predict the dependent variables with a higher degree of certainty. Meanwhile, the p-values for the F-statistics are less than 0.05, confirming that only models for stress, compressive strength, breaking strain, breaking load, breaking period, and water of absorption; have their dependent variable coefficients significantly different from zero, proving the null hypothesis that, all of the model coefficients are zero (0) wrong and hence these models are the successful ones for the prediction of the dependent variables. Additionally, from the F-statistic, models for ‘Elongation’ and ‘Strain’ lack the ability to predict their experimental values accurately, going to confirm their earlier exclusion (from the successful models) using the R² measured. Though the models for ‘Elastic Modulus’ and ‘Modulus of Rapture’ have p-values greater than 0.05 (which is the hypothesis test statistic value), their high R² values suggest they are average models capable of predicting their dependent variables but not as accurate as models for ‘Stress, Compressive Strength, Breaking Strain, Breaking Period, Breaking Period, and Water of Absorption’, with p-values
less than the 0.05 and $R^2$ approximately 1.0 for all. The model results for the mechanical properties are given in Table-2.

### Table-2. Applicability limit for model independent variables.

<table>
<thead>
<tr>
<th></th>
<th>$X_1$</th>
<th>$X_2$</th>
<th>$X_4$</th>
<th>$X_5$</th>
<th>$X_6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Range</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>22-91 (T)</td>
<td>542.54-2364.52 (C)</td>
<td>0.776 (T)</td>
<td>0.139-0.403 (F)</td>
<td>5.48-16.45 (W)</td>
</tr>
<tr>
<td></td>
<td>0-15 (T)</td>
<td>0-22.5 (C)</td>
<td>0-15 (T)</td>
<td>0-22.5 (C)</td>
<td>0-0.75 (T)</td>
</tr>
</tbody>
</table>

### Table-3. Model results of multiple linear regression analysis.

<table>
<thead>
<tr>
<th>Mechanical characterization</th>
<th>Dependent variable</th>
<th>$R$</th>
<th>$R^2$</th>
<th>Adjusted $R^2$</th>
<th>F-statistic</th>
<th>F-value</th>
<th>p-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Elongation (%)</td>
<td>$y_e$</td>
<td>0.683</td>
<td>0.467</td>
<td>0.086</td>
<td>1.227</td>
<td>0.388</td>
<td></td>
</tr>
<tr>
<td>Stress (MPa)</td>
<td>$y_\sigma$</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>102917.3</td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>Strain</td>
<td>$y_\varepsilon$</td>
<td>0.466</td>
<td>0.217</td>
<td>-0.342</td>
<td>0.388</td>
<td>0.843</td>
<td></td>
</tr>
<tr>
<td>Elastic Modulus (GPa)</td>
<td>$y_E$</td>
<td>0.776</td>
<td>0.602</td>
<td>0.318</td>
<td>2.121</td>
<td>0.177</td>
<td></td>
</tr>
<tr>
<td>Compressive Strength (MPa)</td>
<td>$y_{cs}$</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>102629.9</td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>Modulus of Rapture (MPa)</td>
<td>$y_g$</td>
<td>0.747</td>
<td>0.558</td>
<td>0.242</td>
<td>1.767</td>
<td>0.238</td>
<td></td>
</tr>
<tr>
<td>Breaking Strain</td>
<td>$y_{b}\varepsilon$</td>
<td>0.968</td>
<td>0.937</td>
<td>0.892</td>
<td>20.763</td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>Breaking Load (N)</td>
<td>$y_{BP}$</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>0.0</td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>Breaking Period (s)</td>
<td>$y_{BP}$</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>0.0</td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>Water Absorption (%)</td>
<td>$y_w$</td>
<td>0.962</td>
<td>0.926</td>
<td>0.873</td>
<td>17.57</td>
<td>0.001</td>
<td></td>
</tr>
</tbody>
</table>

### 4. CONCLUSIONS

In this study, we investigated the effect of adding clay, oil palm fruit and oil palm wood particulates into polypropylene matrix. The results showed an increase in tensile and compressive strengths for the composite materials as against the raw polymer. Flexural tests on clay samples recorded higher values; however, an increase in clay content in reinforced composites resulted in a higher modulus of rupture values. The results from SEM suggest, a further improvement is needed in the adhesion between phases. Also, out of the ten (10) multiple linear regression models used for the prediction of mechanical property values, six models were found to predict accurately the experimental data on the mechanical property values for the range of independent variable values shown in Table 2.

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