Structural Characteristic and Mechanical Behaviour of Polypropylene Composites Reinforced with Entada Mannii Fibre

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Abstract: The mechanical behaviour of polypropylene based thermoplastic composites reinforced with Entada mannii fibre suitable for light weight panel in automobile part was investigated. The composites were fabricated by mixing the Entada mannii fibre of 1 %, 3 %, 5 % and 7 % of alkaline-treated and untreated and the polypropylene matrix in a twin screw extruder followed by compression moulding. Tensile properties, impact strength and flexural properties of the composites were determined while the surface morphology of the composite fracture surface was examined using scanning electron microscopy (SEM). The results show that NaOH treatment modified the surface characteristics and physical constituents of the fibres. Tensile strength and elastic modulus of the composites were improved with the treated and untreated fibre reinforced composites with increasing fibre loading. Reduction in the percentage elongation was observed as the fibre loading increases for all the treated and untreated composites. A remarkable improvement in impact strength was observed for the alkaline-treated composites than the unreinforced and untreated fibre reinforced composites. However, flexural strength and flexural modulus of the alkaline-treated composites improved significantly than that of the untreated and unreinforced composites. The fracture surface morphology of the untreated composites revealed fibre pullout, fibre debonding, and deposition of pores/holes indicated a weak adhesion between the fibre and the matrix. Finally, alkaline-treated fibre reinforced composites revealed significant improvement in the bonding of the fibre to the matrix.

Keywords: Entada mannii; interfacial adhesion; bonding; composites; pores, holes

1. Introduction

In recent years, the use of biodegradable plant derived fibres in reinforcements of polymer based composites had raised interest and awareness among researchers and industries. Several plant fibres have been explored in this regards with varied degrees of success. Among natural fibres for composites are the bast fibres, extracted from the stems of plants such as jute, kenaf, flax, ramie and hemp. They are widely used due to their excellent properties such as low cost, weight reduction, good dimensional stability and good mechanical strength (Mohanty et al., 2000). Natural fibres as potential reinforcement of composites are emerging rapidly as substitute to the counterpart synthetic fibres in applications such as automotive, aerospace, marine, sport and electronic equipment (Monteiro et al., 2009). The automobile industries are successfully applying natural fibres in the interiors and exteriors panels (Thakur, 2014; Ashori et al., 2008). The goal of using natural fibres in automotive exterior and interior components is essential to recover eco-efficiency, renewability and also trim down the production cost (Jeyathi and Janci, 2012).

Research works on the use of both long and short fibres in composites production are widely used for automobile applications. The efficiency and performance of the short fibre in reinforcement of composites is less as compared with long fibre due to orientation and distribution (Jeyanthi and Janci, 2012). However, the same short natural fibre can be used in automotive components by considering automotive safety legislations, crashworthiness and safety especially in selection of light weight panels (Feng and Feng, 2002). While fibre reinforced composites have already proven their worth as weight-saving materials, the current challenge is to improve the interfacial adhesion between the fibre and matrix, physical properties and make them cost effective.

Interfacial adhesion between fibre and matrix plays an important role in the production of fibre reinforced composites (Kabir et al., 2013). A good interfacial adhesion between fibre and the matrix enhanced stiffness and strength but brittle in nature (Beckermann and Pickering, 2008). On the other hand, a weak interfacial adhesion between the fibre and matrix leads to a lower strength and stiffness with increase toughness (Aruan and Pickering, 2014). On this account, poor fibre-matrix
adhesion which impoverishes mechanical properties of the composite has been a major concern of natural fibres as reinforcements in polymer composites (Arrakhiz et al., 2013). Many attempts have been made by researchers towards improving the mechanical properties with the aim of improving the interfacial adhesion between the fibre and matrix.

Ahmed et al. (2006) adopted the use of chemical modification to improve the fibre–matrix interfacial adhesion. This improved the mechanical properties of the treated composites such as tensile strength and flexural properties of the composites, respectively.

Arun and Pickering (2014) worked on Comparison of harakeke with hemp fibre as a potential reinforcement in composites. The result revealed that tensile strength of treated Harakeke and hemp fibre was improved after the chemical treatment using 5 wt% NaOH/ 2 wt% Na₂SO₃ and 5 wt% NaOH removing surface constituents from the fibre. This shows a potential use of the fibres in reinforcement of polymer composites. Incompatibility of the natural fibre exists due to the hydrophobic nature and the hydrophobic nature of polymer matrix which creates a weak interface between the fibre and the matrix.

Fattai et al. (2009) worked on evaluation of the effect of fibre volume fraction on mechanical properties of a polymer matrix composite. Bagass fibre of various formulations of 0%, 2%, 4%, 6%, 8%, 10%, 15% and 20% were prepared as the reinforcement of composites. Investigation revealed that, there is possibility of using the bagass fibre as reinforcement with improvements in the UTS, modulus of elasticity and extension at break of the composites attaining a maximum reinforcement at 10%.

Al Maadeed et al. (2012) investigated the mechanical and thermal properties of use of date palm wood/glass fibre reinforced composites of recycled polypropylene. Addition of 5% glass fibre to the wood flour would increase the tensile strength by 18% relative to the wood flour alone. The hardness properties of the glass fibre composites was improved and then other composites.

Ayrilmis et al. (2011) evaluated the physical, mechanical and flammability properties of coconut fibre-reinforced polypropylene (PP) composite panels. Fibre volume of (40, 50, 60 and 70%) weight were selected and compounded with polypropylene matrix. The results show that, addition of coconut fibre into the matrix would increase the flexural strength, tensile strength and hardness of the composites with increasing fibre loading up to 60%wt.

Islam and Haque (2013) investigated the influence of fibre surface treatments on the mechanical properties of coir fibre-reinforced polypropylene composites. Sodium hydroxide (NaOH) was used for the chemical treatment of the fibre to remove fibre constituents and wax from the fibre surface and enhanced fibre-matrix interfacial. The result revealed that the mechanical properties of the composites were improved than untreated fibre reinforced composites owing to the chemical treatment which helped to remove fibre constituents that could be detrimental to the fibre-matrix bonding.

Recently, there has been interest in investigating the potentials of Entada mannii plant stem fibre as reinforcement in PMCs (Balogun et al., 2015). Entada mannii belongs to the family (Oliv.) Tisser - Leguminous mermosaesae, and liana plant. The plant is about 5 to 10m high semi-climber which grows in the tropical forest of Nigeria, Gabon and Madagascar. The Entada mannii plant which is traditionally acclaimed for its competence as binding and rope making material in several indigenous communities in Africa is yet to receive attention as a potential source of fibres for reinforcing polymers. This natural fibre can be easily modified by chemicals to improve their mechanical and physical properties (Balogun et al., 2016). In spite of lots of research done on the use of natural fibre in reinforcement of polypropylene composites for automobile applications, no research work has been reported on the use of randomly distributed Entada mannii fibre of lower weights for light components in automobile applications. The aim of this study investigates the mechanical behaviour of thermoplastic composites reinforced with short Entada mannii fibre suitable for light weights in automobile applications.

2. Materials and Methods

2.1 Materials

Entada mannii bast was obtained from Ikare-Akoko, Ondo State, Nigeria; Polypropylene-Homopolymer was supplied by Safripol, South Africa; Teflon sheet was used as the releasing agent; while a 5% of Maleic anhydride polypropylene (MAPP) served as the coupling agent to improve the fibre-matrix interfacial bonding.

2.2 Methods

2.2.1 Extraction of fibre

The Entada mannii fibre was obtained from the bark stem plant (see Figure 1) by hand stripping method. The extracted fibres were dried in an oven at 65 °C for two (2) days to remove moistures and other fibre constituents that could be detrimental to the fibre matrix bonding.

Figure. 1. (a) Distribution of Entada mannii plant containing the fibre (b) Extracted Entada mannii fibres.
2.2.2 Fibre surface treatment
A 5 % NaOH solution was used for the fibre surface treatment of the Entada mannii fibre. The fibres were immersed in the NaOH solution and placed in a water shaker bath at 50 °C for 5 hr. The insoluble residue was delignified at pH3 and washed with distilled water in order to remove mineral traces. The treated fibre obtained was later dried in an oven at 60 °C for 2 days to remove moistures from the fibre while some untreated Entada mannii fibres were also retained for control experimentation.

2.2.3 Composite fabrication
NaOH treated and untreated dried Entada mannii fibres of 1,3,5 and 7 wt % of the polypropylene based composites were chopped into 2 mm length and mixed with 5% Maleic anhydride polypropylene (MAPP) and homo polypropylene as the matrix. The mixtures were fed into a Jones high speed mixer for proper mixing of the fibre and the matrix. The mixtures were dried in an air circulated oven at 60 °C for 8 hr to remove moisture before they were extruded using a Twin-Screw extruder. The barrel temperature was in the range of 130-190 °C and the screw speed was fixed at 60 rpm. The extrudate was granulated in an industrial granulator into pellets dimensioning 3 to 5 mm and randomly oriented and distributed in a metallic mould. The composites were compounded for 10 minutes at a temperature of 190 °C under a constant pressure allowing thorough penetration and dispersion of the fibre into the matrix. Afterwards, the mould was transferred to another compression moulding machine and cold-pressed at 100 MPa for 12 min. The composites sheets produced were approximately 150 mm by 150 mm by 2 mm in thickness for both untreated and treated composites (see Figure 2). Polypropylene sheets of same dimensions were also prepared for control experimentation.

2.2.4 Tensile testing
Tensile test were performed on the composites produced using a universal tensile testing machine operated at a strain rate of 10mm/min with 10 KN load cell. The sample preparation, testing procedure and determination of the tensile strength and tensile modulus were in accordance with ASTM D638 (Al Maadeed et al., 2013). Six repeat tensile tests were carried out to guarantee the reliability of the results obtained.

2.2.5 Impact Strength
The impact strength of the Entada mannii fibre composite was evaluated using an Izod impact test machine. The sample preparation and testing procedure were in accordance with ISO 179 standard 10. All composite specimens were notched and the test specimen supported by a cantilever beam. Hammer head of 7.5 J was released with impact velocity of 2m/s to strike and break the notch specimens. Six specimens were tested at room temperature and the values were recorded.

2.2.6 Flexural strength
Flexural testing commonly known as three-point bending test was carried out on the composites using a universal tensile testing machine. The sample preparation and testing procedure were in accordance with ASTM D790. The test was performed by supporting the composite specimens on a beam and load was applied at the center. The test was carried out at temperature of 23 °C with a cross head speed of 2 mm/min. 6 samples were tested and the results were documented.

2.2.7 Morphology analysis (Scanning Electron Microscope analysis)
The surface morphology and fracture morphology of composites after tensile test were examined using a JEOL JSM-7600F model scanning electron microscope. Secondary electron was used while sample was placed in vacuum chamber, air dried and coated with 100A thick irradium in JEOL sputter ion coater at 15Kev.

3. Results and Discussion
3.1 Chemical treatment
Table 1 shows the fibre constituents of the NaOH treated and untreated Entada mannii fibre. The untreated fibre consists of cellulose (41.18%), hemicellulose (46.79%), lignin (8.12%), Ash content (5.81%) and moisture content (7.83%). It is evident that the cellulose content (54.79%) increases after the alkaline treatment as compared to that of untreated fibres (41.18%). This was due to the removal of hemicellulose and lignin contents from the fibre surfaces which increases the relative amount of the cellulose contents on the treated fibre. These changes in fibre properties were due to the alkaline treatment (Kabir et al., 2013).

The results reveal that after the treatment of Entada mannii fibre, NaOH removes the fibre constituents (Lignin and hemicellulose from the fibre surface and thereby reduces the hemicellulose and lignin contents). Similar work was reported by Balogun et al., (2016) on the effect of chemical treatment of tensile properties of soil retted Entada mannii fibre and found that, alkal...
treatments have higher reactivity in removing hemicellulose and lignin constituents from the fibre.

### Table 1. Entada mannii fibre constituents for treated and untreated fibre

<table>
<thead>
<tr>
<th>Samples</th>
<th>%Lignin</th>
<th>%Ash Content</th>
<th>%Cellulose</th>
<th>%Hemicellulose</th>
</tr>
</thead>
<tbody>
<tr>
<td>Treated (NaOH) (Untreated)</td>
<td>6.29</td>
<td>4.35</td>
<td>54.79</td>
<td>40.88</td>
</tr>
</tbody>
</table>

### 3.2 Tensile Strength

The variation in the tensile strength of the polypropylene based composites reinforced with NaOH treated and untreated Entada mannii fibres that are presented in Figure 3a. It is evident that the tensile strength varies with fibre wt % for both treated and untreated fibres. It is observed that on the average, the tensile strength of the composites reinforced with NaOH treated Entada mannii fibre improves significantly over the composites reinforced with untreated fibres and PURE PP composites.

This improvement is largely due to the appreciable removal of lignin and hemicellulose from the fibre surface which has been reported to contribute to poor fibre/matrix adhesion (Ashori et al., 2008). The tensile strength of 5wt% (56.8 MPa) NaOH treated composites is also observed to increase by 20% and 12% than PURE PP and untreated composites due to good adhesion between fibre and the matrix. This is consistent with the research work of Asumani et al. (2012) who attributed that the increase in tensile strength of the composites was due to the removal fibre surface constituents such as lignin and hemicellulose that could be detrimental to the fibre-matrix interfacial bonding and enhances better fibre adhesion.

It is also noted that the NaOH treated fibre reinforced composites increases significantly in stiffness for the 1%, 3% and 5% wt fibre than the untreated fibre due to better dispersion of the fibre into the matrix. Arrakhiz et al. (2013) reported that treated fibre exhibited good interaction between the fibre and the matrix which makes better improvements in the stiffness imparted from the fibre to the composites. Hence, this is an indication that the NaOH treatment promotes evenly distribution and better fibre-matrix adhesion, allowing efficient stress transfer between the fibre and the matrix.

The modulus of the Entada mannii fibre reinforced composites is shown in the Figure 3b. It is observed that the variation in tensile modulus with the composite is similar to that tensile strength in Figure 3a. The tensile modulus of the fibre reinforced composite for treated and untreated composites improves over that of the unreinforced composites with increase in the fibre wt %.

This improvement was due to the removal of the fibre constituents from the fibre surface which improves the fibre-matrix adhesion of the treated composites than the untreated and PURE PP. Bledzki et al. (2012) reported that increase in tensile modulus of the kenaf treated fibre reinforced composites improves over the untreated fibre reinforced composites due to alkaline treatment attributed to improved bonding between the fibre and matrix.

### 3.3 Percentage Elongation

Figure 4 shows the percent elongation for the NaOH treated and untreated Entada mannii fibre reinforced composites. It is observed that the percent elongation reduces with increase in the fibre wt% which is in contrast to the trend observed for tensile strength and tensile modulus. Generally, for the polymer composites, the percentage elongation at break decreases with the addition of fibres to polymer matrix despite the interface between the polymer and the fibre (Morsyleide et al., 2009). Almaadeed et al. (2013) reported that increase in fibre
loading would translate into higher tensile strength and lower elongation due to higher crystallinity of the polypropylene composites.

3.4 Impact properties

The notched Charpy impact strength of the (treated and untreated) *Entada mannii* fibre reinforced composites is presented in Figure 5. It is observed that the treated *Entada mannii* fibre reinforced composites increased with increase of 5wt% (7.88kJ/m²) fibre composites by 70% than PURE PP. This progressive improvement in the strength was due to the modification of the fibre which led to an increase in the toughness of the composites compared to PURE PP and untreated composites (Arrakhiz et al., 2013; Bledzki et al., 2012; Morsyleide et al., 2009). A slight drop in the impact properties was observed for the 7% wt fibre composites. The reduction in the impact strength was due to poor fibre-fibre interface as a result of increase fibre density that leads to a poor interaction of the fibre with the matrix.

3.5 Flexural Strength and Modulus

Flexural strength of the composites reinforced with treated and untreated *Entada mannii* fibre is presented in Figure 6a. It can be seen that the flexural strength of the NaOH treated composites has a significantly higher flexural strength than the untreated and PURE PP. The peak in flexural strength is observed for the 7wt% NaOH treated composites of 73.64MPa of 71% increase than PURE PP. The increase in the flexural strength was due to the NaOH treatment of the fibre which removes fibre constituents such as lignin and hemicellulose that reduce the fibre-matrix interfacial bonding. Carvalho et al. (2010) reported that the use of alkaline treatment is a greatly beneficial in the removal of dirts and fibre constituents which improves the better fibre-matrix adhesion. Figure 6b shows the flexural modulus comparison for the NaOH treated and untreated *Entada mannii* fibre reinforced polypropylene composites. The result also indicates that the flexural strength of alkaline-treated composites improves significantly over the untreated and pure PP.
Flexural modulus of *Entada mannii* fibre reinforced composites increases with the increase in the fibre weight percent for both treated and untreated fibre reinforced composites. This indicated that *Entada mannii* fibre provided good reinforcement to the pure PP matrix. Mohammed *et al.* (2011) worked on the effect of removing polypropylene fibre surface finishes on mechanical performance of kenaf/polypropylene composites that reported that the average chemical treatment of the fibre provided a significant improvement over the untreated fibre reinforced composites with considerable increase in flexural modulus.

### 3.6 Fracture Analysis

Figure 7 shows the fracture surface of the pure polypropylene polymer. The image shows that the matrix phase has a complete homogeneous phase. Figure 8 shows the fracture images of the untreated *Entada mannii* fibre reinforced composites. It can be seen that the tensile rupture is accompanied by fibre debonding, fibre pullout and deposition of pores/holes which is indicative of weak adhesion between the fibre and the matrix. The fibre pull out was due to poor interfacial adhesion between the fibre and the matrix.

![Figure 7. SEM micrographs for the unreinforced polypropylene](image)

Figure 9 shows the SEM fracture images of the NaOH treated *Entada mannii* fibre reinforced composites. Significant improvement in the fibre/matrix adhesion is observed between the fibre and the matrix, indicated a better fibre-matrix bonding. This might be due to the removal of some fatty substance and impurities from the fibre surfaces deposited pores on the fibre. These pores enhanced bonding characteristics of the fibre during lamination (Sherely *et al.*, 2008; Beckermann and Pickering, 2008). It shows that there is reduction in fibre pullout, suggesting that polypropylene matrix and the fibre adhere strongly to the fibre. Very few fibres can be observed on the fracture surfaces due to fibre concealment by the matrix material, although a few severely damaged fibres can be seen protruding out of the matrix (Beckermann and Pickering, 2008). This also indicated the higher interfacial shear strength for treated fibres and good interfacial adhesion between the fibres and matrix (Morsyleide *et al.*, 2009).

![Figure 9. SEM micrographs for (a) 1%, (b) 3%, (c) 5% and (d) 7% Alkaline-treated Entada mannii composite](image)

### 4. Conclusion

The mechanical behaviour of the thermoplastic composites reinforced with *Entada mannii* fibre was investigated.

- Alkaline treatment improved the surface modification of the fibre and enhanced better fibre-matrix interfacial adhesion than the untreated fibres.
- Tensile strength and elastic modulus of the composites were improved with the treated and untreated fibre reinforced composites than unreinforced composites as the fibre loading increases.
- Reduction in the % elongation was observed as the fibre loading increases for all the treated and untreated composites.
• A remarkable improvement in impact strength was observed for the Alkaline-treated composites than the unrefinced and untreated fibre reinforced composites.

• Flexural strength and flexural modulus of the alkaline-treated composites improved significantly than the untreated and unrefinced composites. The peak in flexural strength and flexural modulus for both the treated and untreated composites increases with increase in fibre loading at 7%wt.

• The fracture surface morphology of the unrefinced composites revealed fibre pullout, fibre debonding, and deposition of pores/holes indicted a weak adhesion between the fibre and the matrix. Alkaline-treated fibre reinforced composites revealed significant improvement in the bonding of the fibre to the matrix.

• Incorporating Entada mannii fibre into polypropylene matrix has shown an improvement in mechanical properties of the composites and this could be utilised where lightweight materials is required in automobile application.

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References:


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