Influence of sawdust particles reinforcement on physical and mechanical properties of High-Density Polyethylene (HDPE) matrix composites

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Abstract
The influence of sawdust particles reinforcement on the physical and mechanical characteristics of high-density polyethylene (HDPE) matrix composites was studied for application as sustainable wood plastic composites (WPCs) for housing. The WPCs developed by compression moulding method were characterised. The results revealed web-like structures/cross-linking in the microstructure of the samples, which is a characteristic of polymers. The microstructure revealed a good dispersion of sawdust particles and compatibilizer in the HDPE matrix and bonding, which enhanced the properties of the composites. The control sample C exhibited water absorption of 0.22 % whereas sample S8 having 1.1 to 1.4 mm sawdust particles, 30 wt. % of sawdust particles content, and 3 wt. % of compatibilizer exhibited the least water absorption of 0.14 %. The unreinforced HDPE control sample exhibited a tensile strength of 12.53 MPa while sample S7 with the smallest size (less than 1 mm) and 30 wt. % of sawdust particles content, and 7 wt. % of compatibilizer exhibited the highest tensile strength of 16.22 MPa. This is 29.5 % higher than that of the control sample. The control sample exhibited a flexural strength of 10.2 MPa while sample S7 exhibited the highest flexural strength of 14.85 MPa, which is 45.6 % higher than that of the control sample. The control sample exhibited a hardness value of 13.93 HV while sample S7 exhibited the highest hardness value of 19.17 HV, which is 37.6 % greater than that of the control sample. Samples S5, S7, S8, and S9, which contained high content of sawdust particles demonstrated impact energy values of 34.27, 33.14, 35.17, and 36.46 J respectively. The unreinforced control sample demonstrated a low wear rate value of 0.35 g/Nm. However, sample S7 demonstrated the least wear rate of 0.23 g/Nm, which is 34.3 % lower than that of the control sample. In view of these characteristics, the composites especially sample S7, has the potential for application as a sustainable building material.

Keywords: High-density polyethylene; Sawdust particles; Compression moulding; Characterization

1. Introduction
The increasing environmental concern and depletion of petroleum resources have motivated researchers and industries to consider using sustainable natural fillers, which could be particles or fibres rather than synthetic fillers. In addition to being environmentally-friendly, natural particles/fibres can exhibit better properties than synthetic fillers [1]. Besides good physical and mechanical properties, they are economical, recyclable, nontoxic, and widely available. Therefore, natural fillers have evolved as suitable materials for applications as reinforcement in various composite products [2, 3]. Thus, the worldwide market of natural fillers reinforced polymer composites reached $5.3 billion in 2019, with an estimated growth of 11.4 % between 2000 and 2017 [4], and the expectation of steady global growth in the future [3-6].

With the global increase in population and a high percentage of millennials, there is a high demand for housing. For example, Nigerians have forced their government to invest heavily in housing in the last decade [7-9]. However, no significant improvement has been recorded as the number of houses is primarily limited by the high cost of materials and construction [9], making it impossible to meet even greater population growth and increase in demand. A promising path forward is the development of low-cost, available, and sustainable materials that do not add to the existing pollution and climate change challenges. In this context, sustainable polymer matrix composites made of natural reinforcements and possibly recycled plastics have emerged as potential materials of choice.

Polymer matrix composites generally consist of a thermoplastic or thermoset matrix with organic (e.g., wood flour, chicken feather) or inorganic (mineral or glass materials) fillers (particulates or fibres) [10]. The fibre/particle-reinforced polymer matrix composites have attracted considerable attention in numerous applications, with natural fillers being advantageous compared to their synthetic counterparts due to superior properties and outstanding sustainability [11]. Specifically, wood-plastic composites (WPCs) are made by blending plastic matrix with agricultural or forestry waste wood fibres (WFs), which give the composite material a wood facade with significant environmental benefits and mechanical performance advantages [5, 12].

The types of plastics commonly used in WPCs production are polyvinyl chloride (PVC), polyethylene (PE) and polypropylene (PP) due to their low melting points and good thermoplasticity [13]. Among them, high-density polyethylene (HDPE) exhibits the advantages of high shrinkage, high melt strength, and relatively easy processability, recyclability, good chemical resistance and bio-compatibility at a relatively low cost [14]. The properties of HDPE can be further improved by the incorporation of various inorganic or organic nanofillers into HDPE matrix [14]. That is why HDPE
Based WPCs are the most widely used in the market. As a result, WPCs have witnessed a rapid development in recent years [4]. Furthermore, their superior efficiency, lightweight, abundance, and low moisture absorption with excellent dimensional stability are attractive characteristics [15]. They are gaining applications in diverse fields from household to office appliances, garden architecture, building construction, business equipment, automotive, decorative material, and furniture [16].

Studies have shown that green materials have become more mainstream with considerable awareness of preserving the environment across the globe [3]. In improving the mechanical properties of WPCs, it has been shown that both matrix and fibre properties are essential. The tensile strength has shown to be more sensitive to the matrix characteristics while the elastic modulus has shown to be mainly dependent on the fibre properties [6, 17]. As such, fibres/particles are essential for the structural performance of WPCs for use in building construction. The modulus of rupture (MOR) is used for the derivation of appropriate composition values, which in conjunction with the modulus of elasticity (MOE) build the basis for the determination of strength and serviceability [18].

Many studies have been conducted on WPCs. For instance, Ramesh et al [19] investigated the tensile strength and hardness of phenol formaldehyde (PF) based WPCs. It was reported that increased wood flour content increased the tensile strength of the composites. However, above 50 vol. % wood flour, a deterioration in interfacial bonding between the matrix and fibres and a decrease in the tensile strength were observed. Rahman et al [20] investigated the physical and mechanical properties of WPCs using recycled polyethylene terephthalate (PET) as matrix. The production was done using different mixing ratios and flat-pressed method. It was reported that the MOR and MOE of the composites increased with reinforcement content at lower contents but decreased with further increasing sawdust content above 40 wt. %. Tabarsa et al [2] also investigated the physical and mechanical properties of WPCs using wood flour particles, polypropylene (PP) matrix, and polypropylene grafted maleic anhydride (coupling agent). The blends were compounded in a twin-screw extruder at a controlled temperature. The results showed that with increase of wood flour up to 35 wt. %, MOR, MOE, water absorption, and thickness swelling increased but beyond 35 wt. % particles reinforcement, these properties decreased. However, increase of wood flour up to 40 wt. % increased the hardness of the composites.

Increasing global awareness of environmental challenges and the need for sustainable development have increased interest in using natural fibres/particles as substitutes for their synthetic counterparts in the reinforcement of polymer composites. The approach is further motivated by the potential to achieve improved composite properties that can be readily tailored to the desired final products [21]. However, one potential weakness of WPCs is their degradation when exposed to sunlight and weather conditions. However, some additives have been found to reduce or even eliminate this risk effectively. Hence, this challenge is drastically reduced using additives.

Many research groups have directed their works toward defining numerous combinations of biodegradable matrix/natural fillers. Among the emerging biodegradable composites is wood reinforcement of polypropylene or polyethylene [22-27]. Therefore, the need to continuously improve the characteristics of polymers through particles/fibres reinforcement to develop polymer matrix composites with enhanced functional characteristics is a welcome development.

Because bio-based materials are indispensable reality for a future sustainable society, their application in many areas will continue to increase. Because of the availability of trees on earth, wood is one of the most abundant renewable/sustainable materials. Trees are biological materials with long life span and are biodegradable. Hence, they are considered as important building materials for sustainable development. A promising path forward is the development of low-cost, available, and sustainable materials that do not add to the existing pollution and climate change challenges. In this context, sustainable wood-plastic composites (WPCs) made of natural reinforcement (sawdust) and possibly recycled plastics have emerged as potential materials of choice as building materials. Hence, the aim of this study is geared toward the utilization of plastics waste for WPCs production and investigation of their properties for possible application as building construction materials using sawdust particles and recycled HDPE as input materials. This will reduce pollution and other environmental challenges caused by the growing amounts of plastics waste and their improper disposal, particularly in developing countries.

2. Materials and method

2.1. Materials, preparation and samples production

The materials used are high-density polyethylene (covers of water and beverage plastic bottles), which were collected from food vendors, home waste bins, and waste management center inside the campus of the University of Lagos, sawdust, and maleic anhydride grafted polyethylene as a compatibilizer. The sawdust was obtained from the sawmill and wood markets in Lagos while maleic anhydride grafted polyethylene was obtained from a registered vendor in Lagos. The plastic bottle covers were washed to remove dirt and other impurities and sun-dried. Thereafter, they were ground to smaller pieces using a shredding machine. The sawdust was sun-dried at a temperature between 27-32 °C for six days at 12 hrs stirring intervals to ensure proper drying. The sawdust was ground and sieved into three particle size ranges of 0-1 mm, 1.1 to 1.4 mm, and 1.5 to 2.8 mm respectively using British standardised sieves (BSS). Weighed ground plastics were put in a stainless container, charged into an industrial muffle furnace of temperature between 170 and 190 °C, and was held for about 45 mins. Thereafter, the melt was removed from the furnace and weighed quantities of the sawdust and maleic anhydride compatibilizer were added to it, and stirred for proper blending. After a semi-solid of the blend was formed, it was poured into the bottom half of a wooden mould of cavities of size 17 x 165 mm. The top half of the mould was then placed to cover the set-up. Finally, a mass of 10 kg was placed on the mould for 15 mins to compress the composite. The composite was then removed from the mould to cool at room temperature. The formulation for each of the samples is presented in Table 1 with a total mass of 100 g. Sixty-five samples were produced and designated as C (control sample), S1, S2, S3, S4, S5, S6, S7, S8 and, S9 accordingly and were characterized. The photographs of the input materials, apparatus, equipment, and samples are shown in Fig. 1 and 2.
Figure 2: Input materials and apparatus used (a) 0 – 1 mm sawdust (b) 1.1 – 1.4 mm sawdust (c) 1.5 – 2.8 mm sawdust (d) maleic anhydride grafted polyethylene (e) sieves (f) some of the samples produced (g) a machined sample for tensile test.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Sawdust Particle Sizes (mm)</th>
<th>Sawdust (wt. %)</th>
<th>Maleic anhydride grafted polyethylene (wt. %)</th>
<th>HDPE (wt. %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C (control)</td>
<td>Nil</td>
<td>Nil</td>
<td>Nil</td>
<td>100</td>
</tr>
<tr>
<td>S1</td>
<td>&lt;1</td>
<td>10</td>
<td>3</td>
<td>87</td>
</tr>
<tr>
<td>S2</td>
<td>1.1 – 1.4</td>
<td>10</td>
<td>5</td>
<td>85</td>
</tr>
<tr>
<td>S3</td>
<td>1.5 – 2.8</td>
<td>10</td>
<td>7</td>
<td>83</td>
</tr>
<tr>
<td>S4</td>
<td>&lt;1</td>
<td>20</td>
<td>5</td>
<td>75</td>
</tr>
<tr>
<td>S5</td>
<td>1.1 – 1.4</td>
<td>20</td>
<td>7</td>
<td>73</td>
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<tr>
<td>S6</td>
<td>1.5 – 2.8</td>
<td>20</td>
<td>3</td>
<td>77</td>
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<tr>
<td>S7</td>
<td>&lt;1</td>
<td>30</td>
<td>7</td>
<td>63</td>
</tr>
<tr>
<td>S8</td>
<td>1.1 – 1.4</td>
<td>30</td>
<td>3</td>
<td>67</td>
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<tr>
<td>S9</td>
<td>1.5 – 2.8</td>
<td>30</td>
<td>5</td>
<td>65</td>
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</tbody>
</table>

2.2. Microscopy, physical and mechanical testing

Microstructural characterisation was conducted on five samples using a scanning electron microscope (SEM). The samples are flat and square-shaped with 20 mm dimensions. Surface preparation was done, followed by photographic imaging of the surfaces at 10,000 magnification. The water absorption test was conducted by immersing the samples in water at room temperature for 2 and 24 hrs in accordance with ASTM D570-98 (2018) [28] standard. The water absorption was calculated using equation (1).

Water Absorption (%)

\[
\text{Water Absorption} = \frac{W_1 - W_0}{W_0} \times 100
\]

where,

- \(W_0\) = weight before immersion
- \(W_1\) = weight after immersion

Ten tensile test samples were fabricated according to ASTM D638 (2014) [29] standard dimension of the flat sample with a reduced section of 12.5 mm and tensile strength was obtained using a digital XLC universal tester. The sample was placed in the center. The ultimate tensile strength was recorded on loads application until fracture. The raw data was generated and used to plot a graph.

Impact testing was conducted on ten samples of size 55 mm \(\times\) 10 mm \(\times\) 10 mm with a 2 mm deep V-notch at the center using an izod impact-testing machine according to ASTM D256-10 (2018) [30] standard. The striking pendulum was released from a height of 1.5 m, hitting the sample with a velocity of 5 m\(s\)\(^{-1}\). The energy absorbed to break each sample was read from the dynamometer. The flexural test was performed using a digital flexural testing machine with the sample specification according to ASTM D7264 (2015)[31] standard. Each of the samples was laid on a support span while the load was applied to the centre by the landing nose producing three points at a specified rate before permanent deformation allowing accurate measurement of flexural modulus and strength.

For microhardness testing, ten samples were polished using a surface grinder-polisher. Microhardness testing was then done according to ASTM E384-17 (2022) [32] standard using a Vickers hardness tester with a test load of 1.91N. The specific abrasive wear rate of ten samples was measured using a pin-on-disc set up. The counter surface was a grounding paper made of aluminum oxide abrasive. A custom-made pin on disc testing machine was used according to ASTM G99-05 (2010) [33] standard with a load of 11.25 N and a rotating speed of 125 rpm for two minutes. Before the test, initial weights of samples were obtained and their weights after the test using an electronic weighing balance of 0.01 mg accuracy. The difference between initial and final weight was obtained using equation 1 [34].

Where, \(W\) is wear rate (g/N\(\text{m}\)), \(\Delta m\) is mass loss (mg), \(\rho\) is applied load (N) and \(L\) is sliding distance (m).

3. Results and discussion

3.1. Microstructure

The SEM micrographs of Fig. 3 to 7 reveal web-like structures/cross-linking in the microstructure of the samples, which is a characteristic of polymers. Cross-linking or network of polymer molecules makes polymers to be strong in the solid state [35]. The energy-dispersive spectroscopy (EDS) elemental analysis shows the elemental composition of the samples. The presence of carbon (C) in the EDS spectrographs confirms the organic nature of the samples. The microstructure of the reinforced samples reveal some degrees of inhomogeneity between the plastics and sawdust particles. However, the EDS spectrograph of sample S7, which shows the presence of elements such as Fe, S, and Al originating from the high 30 wt. % sawdust content in the composite, suggests a good interfacial interaction/bonding between particles and HDPE matrix [36]. The SEM micrographs of reinforced samples...
also show a good dispersion of sawdust particles in the matrix without clustering or agglomeration. This enhanced load transmission between the matrix and the particles thereby enabling the particles to bear load until fracture occurred. The white phases present in the reinforced samples could be from the maleic anhydride grafted polyethylene. The microstructures show little pores, which is an indication of effective stirring of the melt of the composite mixture prior to pouring and solidification/cooling of the composite melt.

3.2. Water Absorption

The water absorption level of the samples increased after 24 hrs of immersion compared to that of 2 hrs as illustrated in Fig. 8 indicating that there are pores in their microstructures. The control sample C exhibited water absorption of 0.22 % whereas sample S8 having 1.1 to 1.4 mm sawdust particles, 30 wt. % of sawdust fibers content, and 3 wt. % of compatibilizer exhibited the least water absorption of 0.14 %. However, sample S5 that contains 20 wt. % of sawdust particles exhibited the highest water absorption of 1.016 % after 24 hours of immersion. As shown in Fig. 5, there are pores in the microstructure of sample 5. Furthermore, wood particles contain hemicelluloses, which are highly hydrophilic. The hydrophilic hydroxyl (-OH) group is responsible for water absorption in the sawdust particles of sample 5 and other samples containing sawdust particles. Hence, much presence of pores in the microstructure could be responsible for the high water absorption exhibited by sample 5 in Fig. 8. The diffusion of water into the microstructure of the samples can cause changes in the structure and increase flexibility and break up, which could have an adverse effect on the mechanical properties [37]. This can cause an increase in the space of the HDPE molecules, which can reduce their bonds and can cause a reduction in resistance to applied stress [38]. However, the strong bonding of the sawdust particles to the HDPE matrix, which was facilitated by the compatibilizer, enhanced the resistance of the composites to water absorption. This led to the reduction in the water absorbed by the composites especially samples S5, S7 and S8. This agrees with the report by [2, 22].

3.3. Tensile strength

The unreinforced HDPE (control sample C) exhibited a tensile strength of 12.53 MPa as presented in Fig. 9. The tensile strength of the composites improved with increased sawdust particles concentration in the composites. Generally, increase in the concentration of particles resulted to increased tensile strength except sample S3 that exhibited a lower tensile strength. However, sample S7 with the smallest size (less than 1 mm) and 30 wt. % of sawdust particles content, and 7 wt. % of compatibilizer exhibited the highest tensile strength of 16.22 MPa. This is 29.5 % higher than that of the control sample, which is due to the strong bonding of the particles to the HDPE matrix that was facilitated by the compatibilizer. This agrees with the report by [22]. Generally, higher concentration of maleic anhydride compatibilizer must have facilitated the bonding of the particles with the HDPE polymer matrix, which improved the properties of the composites especially the tensile strength, flexural strength, and hardness compared to lower concentration of compatibilizer.

3.4. Flexural strength

The unreinforced HDPE (control sample C) exhibited a flexural strength of 10.2 MPa as presented in Fig. 10. The reinforced composites exhibited higher flexural strength than the control sample except samples S2, S6, and S9, which contained large sawdust par-
particles of 1.1 to 1.4 mm, 1.5 to 2.8 mm, and 1.5 to 2.8 mm respectively. The decrease in flexural strength could be due to the largeness of the particles size. Small size particles enhance densification, which in turn enhance the mechanical properties better than coarse/large particles of the same concentration [39]. Sample S7 with the smallest size (less than 1 mm) and 30 wt. % of sawdust particles content, and 7 wt. % of compatibilizer content exhibited the highest flexural strength of 14.85 MPa. This is 45.6 % higher than that of the control sample, which is due to the strong bonding of the sawdust particles to the HDPE matrix that was facilitated by the compatibilizer. This agrees with the report by [22, 40].

3.5. Hardness and impact energy

The control sample C exhibited a hardness value of 13.93 HV as presented in Fig. 11. The reinforced composites exhibited higher hardness values than the control sample. Sample S6 that contained 20 wt. % of sawdust particles exhibited hardness value of 18.67 HV. Sample S7, which contained 30 wt. % of sawdust particles exhibited the highest hardness value of 19.17 HV. This is 37.6 % greater than the hardness of control sample. As revealed by the EDS spectra in Fig. 2 to 8, the composites contained calcium (Ca) from the matrix, which is hard. The hardness of composite materials is directly proportional to the quantity of integrated hard constituents [41]. The hard and well-bonded composites impeded or restricted the movement of dislocations, which increased the hardness of the composites. This agrees with the report by [42].

The control sample demonstrated an impact energy value of 26.28 J as shown in Fig. 12. The reinforced composites demonstrated higher impact energy values than the control sample. Specifically, samples S5, S7, S8, and S9, which contained high content of sawdust particles, demonstrated impact energy values of 34.27, 33.14, 35.17, and 36.46 J respectively. The sawdust addition improved the energy absorption of the composites before fracture occurred compared to the control sample. The increase in impact energy is due to the strong bonding of the sawdust particles to the HDPE matrix that was facilitated by the compatibilizer.

3.6. Wear rate and its correlation with hardness

The wear rate graph presented in Fig. 13 shows that the samples demonstrated some degrees of deformation in the friction surface due to ploughing force of asperities when load was applied. This agrees with the reported by [43]. The unreinforced control sample C demonstrated a low wear rate value of 0.35 g/Nm. However, the reinforced composites demonstrated lower wear rate values than the control sample. Specifically, samples S6 and S7 that contained 20 wt. % and 30 wt. % of sawdust particles respectively, exhibited lowest wear rate values of 0.25 g/Nm and 0.23 g/Nm respectively. The wear rate demonstrated by sample S7 is 34.3 % lower than that of the control sample. The decrease in wear rate, which implies high wear resistance, could be due to strong interfacial bonding between the HDPE matrix and the particles, which was facilitated by the coupling agent (compatibilizer). Among the samples, sample S7, which contained 30 wt. % of sawdust particles exhibited the highest hardness and lowest wear rate. The high hardness increases the composite’s resistance to indentation, resulting in a lower wear rate [44, 45].
4. Conclusion

In this study, high-density polyethylene (HDPE) plastic composites reinforced by varied weight percentage of sawdust particles and maleic anhydride (coupling agent or compatibilizer) were developed by compression moulding method and were characterised. From the results of investigation and discussion of the study, the following inferences can be drawn:

i. The SEM micrographs revealed web-like structures/cross-linking in the microstructure of the samples, which is a characteristic of polymers.

ii. The reinforced samples revealed a good dispersion of sawdust particles and compatibilizer in the HDPE matrix and bonding, which enhanced load transmission between the matrix and the particles thereby enabling the particles to bear load until fracture occurred.

iii. The control sample C exhibited water absorption of 0.22 % whereas sample S8 having 1.1 to 1.4 mm sawdust particles, 30 wt. % of sawdust fibers content, and 3 wt. % of compatibilizer exhibited the least water absorption of 0.14 %.

iv. The unreinforced HDPE control sample exhibited a tensile strength of 12.53 MPa while sample S7 with the smallest size (less than 1 mm) and 30 wt. % of sawdust particles content, and 7 wt. % of compatibilizer exhibited the highest tensile strength of 16.22 MPa. This is 29.5 % higher than that of the control sample.

v. The control sample exhibited a flexural strength of 10.2 MPa while sample S7 exhibited the highest flexural strength of 14.85 MPa, which is 45.6 % higher than that of the control sample.

vi. The control sample exhibited a hardness value of 13.93 HV while sample S7 exhibited the highest hardness value of 19.17 HV, which is 37.6 % greater than that of the control sample.

vii. Samples S5, S7, S8, and S9, which contained high content of sawdust particles demonstrated impact energy values of 34.27, 33.14, 35.17, and 36.46 J respectively. The sawdust addition improved the energy absorption of the composites before fracture occurred compared to the control sample.

viii. The unreinforced control sample demonstrated a low wear rate value of 0.35 g/Nm. However, sample 7 demonstrated the least wear rate of 0.23 g/Nm, which is 34.3 % lower than that of the control sample.

ix. In view of these characteristics, the composites especially sample 7, has the potentials for application as a sustainable building material.

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References


