

Enhanced Physico-Mechanical Properties of LDPE Reinforced Using Agro-Wastes as Hybrid Fillers

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ABSTRACT

The environmental persistence of petroleum-based plastics such as low-density polyethylene (LDPE) has necessitated research into eco-friendly alternatives. This study investigates the incorporation of mixed agro-waste fillers; coconut husk, breadfruit hull, and periwinkle shell into LDPE matrices to assess their mechanical, thermal, morphological, solvent imbibitions, and biodegradation properties. The composites were fabricated using injection moulding at filler loadings of 10-40wt% and evaluated according to ASTM standards. Results revealed significant improvements in tensile, compressive, shear, impact, and hardness strengths at 20-30wt% loadings, after which agglomeration reduced performance. Differential Scanning Calorimetry (DSC) indicated melting transitions between 120-170°C and oxidation stability above 200°C. Scanning Electron Microscopy (SEM) confirmed uniform filler dispersion at lower loadings and voids at higher concentrations. Solvent imbibitions tests showed negligible water absorption but significant uptake in benzene and toluene, while soil burial tests revealed limited biodegradation, with composites showing moderate weight loss compared to neat LDPE. These findings suggest that agro-waste reinforced LDPE composites can serve as cost-effective, sustainable materials for packaging, household, and light construction applications.

Keywords: LDPE composites, agro-waste fillers, mechanical properties, thermal stability, morphology, solvent imbibition, biodegradation

INTRODUCTION

Low-density polyethylene (LDPE) is a versatile thermoplastic widely employed in packaging, films, containers, and insulation due to its toughness, flexibility, and chemical resistance (Ragaert *et al.*, 2017; Khare and Baruah, 2021). However, its non-biodegradability contributes significantly to global plastic pollution (Geyer *et al.*, 2017). Research has shown that agro-waste fillers, particularly lignocellulosic residues, can improve mechanical performance while promoting partial degradability (Ogudo *et al.*, 2021). Previous studies on LDPE composites reinforced with rice husk (Daramola *et al.*, 2022), snail powder (Chris-Okafor *et al.*, 2018), and corn cob (Zhu *et al.*, 2018) demonstrated that filler incorporation improves stiffness, tensile strength, and hardness but reduces elongation at break. The synergistic use of multiple fillers, however, remains underexplored. This study evaluates the effect of mixed agro-wastes (coconut husk, breadfruit hull, and periwinkle shell) on the performance of LDPE composites, emphasizing their mechanical, thermal, solvent resistance, and biodegradation behaviors.

MATERIALS AND METHODS

Sample collection and preparation

LDPE (Indorama Eleme Petrochemicals, grade NGL105FS) was used as matrix. Agro-waste fillers; coconut husk, breadfruit hull, and periwinkle shell were processed to fine powders (<75µm). Composites were

prepared at 10-40wt% filler loadings by injection moulding. Mechanical, thermal, morphological, solvent uptake and biodegradation tests followed ASTM protocols.

Preparation of composites

The fillers were blended in a 1:1:1 ratio and incorporated into LDPE at 10, 20, 30, and 40 wt%. Injection moulding (200 g capacity) produced test specimens, with an average cycle time of 33secs.

Mechanical properties analysis of the composites

The mechanical properties of the composites considered in this work include; tensile strength, compressive strength, shear strength, impact strength, and hardness, which were measured using the American Standard Testing and Measurement method.

Tensile strength

The tensile strength of the composites was measured according to the ASTM standards-ASTM D-638-14, using the universal testing machine Hounsfield tensometer 8889 made in England. The test piece was measured to 160mm x 19mm x 3.2mm dimension.

Compressive strength

The compressive strength is the capacity of the composites to withstand loads tending to reduce its size. The compressive strength of the composites was measured according to the American Standard Testing Method D-695, using the Hounsfield Monsanto Tensometer 8889. The test piece was measured to 40x40mm dimension square shape.

Shear strength

The shear strength of the composites was measured according to the American Standard Testing Method D-732, using the Hounsfield Monsanto Tensometer 8889. The test piece was measured to 20 mm x 20 mm. The readings were automatically recorded and the values computed.

Impact strength

Testing material impact typically refers to evaluating how a material behaves when subjected to a sudden force or shock, commonly known as an impact test. The test piece was measured to 100mm x 19mm x 3.2mm.

Hardness strength

The hardness strength of the composite was measured according to the ASTM D2240, using shore scale Durometer hardness tester, made in England. The values were automatically measured and read. The test was measured to (100 x 19 x 3.2) mm dimension.

Thermal Analysis

The thermal properties of the composites were studied using Modulated Differential Scanning Calorimeter MDSC 2920 CE USA. Aluminum pans and lids were used for samples and reference and heating rate of 10°C per minute to determine the glass transition temperature (T_g), crystallization temperature (T_c), fusion temperature (T_m), enthalpy variation and heat capacity.

Surface Morphological study

The microstructural arrangements of the composites were conducted using Scanning Electron Microscope (SEM) model: JEOL-JSM 7600F.

Solvent imbibition Analysis

The solvent absorption of the composites was determined using Standard ASTM D-570-98. The composites cut into 50x50mm dimension were immersed in water, benzene, and toluene respectively for a period of 3days. The moisture absorption by the composite was measured by the weight gain of the material at daily intervals. The percentage moisture absorption capacity was expressed as the ratio of increase in mass of the composite to the initial mass.

Degradation study

This test is done to determine the extent the composites will degrade in the environment. This was determined using soil burial degradation test. Composites were buried in a soil obtained from an automobile mechanic workshop for degradation. Composites were cut into 50x50mm dimension, weighed and buried into the soil at 12cm depth for a three-month period. The composites were weighed at interval of 30 days during the test period to determine the extent of degradation. The degradation rate was calculated using the formula;

$$\text{Percentage degradation} = \frac{W_f - W_i}{W_i} \times 100$$

W_f =Final weight, W_i = Initial weight

RESULTS AND DISCUSSION

The results of the mechanical properties of the low density polyethylene with coconut husk, breadfruit hull and periwinkle shell composites are shown in the figures below.

Mechanical properties

Tensile strength

Tensile strength measures the resistance of a material to breaking under tension. The result of the tensile strength of the composites is shown in Fig. 1.

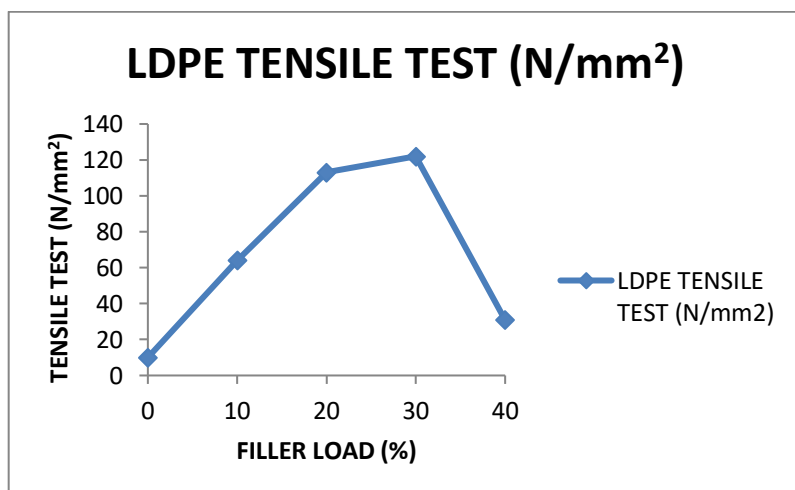


Fig. 1. Effect of filler loading on the Tensile strength of LDPE composites

The tensile strength of LDPE composites increased with filler loading up to an optimum point before declining, as shown in Figure 1. With increasing filler load (10-30wt%), tensile strength improved significantly, reaching a peak of 115N/mm² at 30wt% loading, attributed to effective stress transfer, restricted polymer chain mobility, and strong filler-matrix adhesion (George *et al.*, 2016; Essabir *et al.*, 2013). Beyond this level (40 wt%), tensile strength decreased sharply (<30N/mm²), likely due to filler agglomeration, poor

dispersion, and stress concentration effects, which created weak zones and reduced mechanical integrity (Jawaid and Khalil, 2011).

Compressive strength

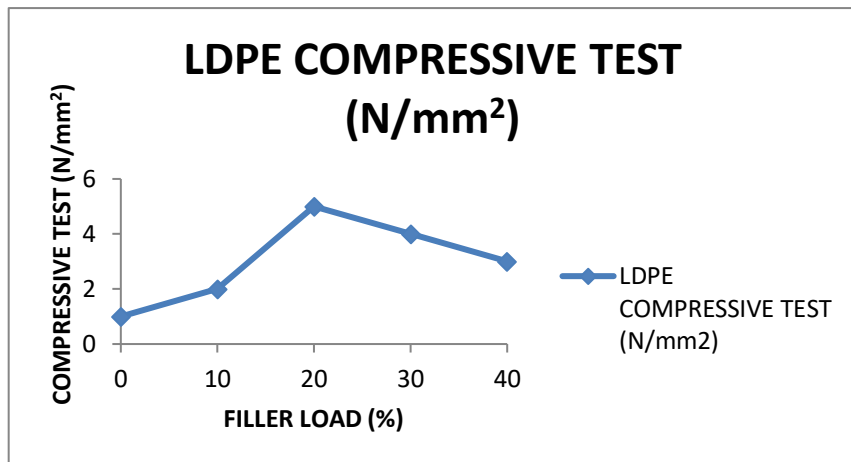


Fig. 2. Effect of filler loading on the Compressive strength of LDPE composites

The compressive strength increases steadily as filler loading rises to 10 wt% and reaches a peak at 20 wt% filler content (5.0 N/mm²). This enhancement can be attributed to improved stress transfer between the matrix and the filler, good interfacial adhesion, and effective dispersion of the filler particles within the LDPE matrix, which restricts polymer chain mobility and enhances load-bearing capacity (Rahman *et al.*, 2019). Beyond 20 wt% filler loading, the compressive strength decreased gradually, dropping about 4.0 N/mm² at 30 wt% and further to 3.0 N/mm² at 40 wt%. This decline is often linked to agglomeration of filler particles at higher concentrations, which introduces voids, micro-cracks, and weak filler-matrix bonding, thereby reducing stress transfer efficiency (Azeez *et al.*, 2020). Such behavior is consistent with the findings of Sanyang *et al.* (2015) that polymer composite where optimum mechanical strength is studied, is typically observed at moderate loadings due to poor dispersion and stress concentration effects.

Shear strength

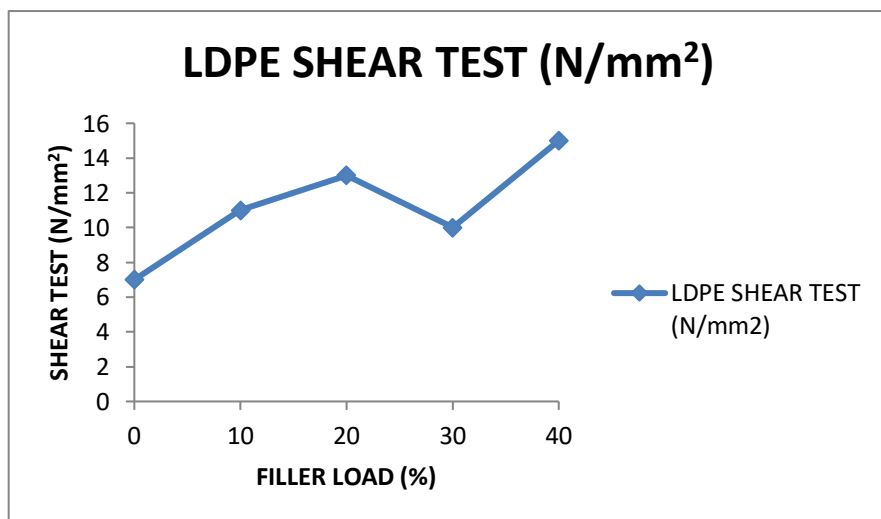


Fig. 3. Effect of filler loading on the Shear strength of LDPE composites

The LDPE shear strength test shows that shear strength increased from about 7 N/mm² at 0% filler to 13 N/mm² at 20 wt% filler, dropped slightly to 10 N/mm² at 30 wt%, and then rose sharply to 15 N/mm² at 40wt% filler load. The initial increase is attributed to good filler dispersion and enhanced interfacial bonding, which improve stress transfer and restrict polymer chain mobility (Callister and Rethwisch, 2020). The decline

at 30 wt% filler suggests possible filler agglomeration and weak adhesion that create stress concentration points. However, the sharp improvement at 40 wt% indicates that higher filler content promoted denser packing and better reinforcement within the matrix, thereby enhancing resistance to shear forces (Ahmed *et al.*, 2012).

Impact strength

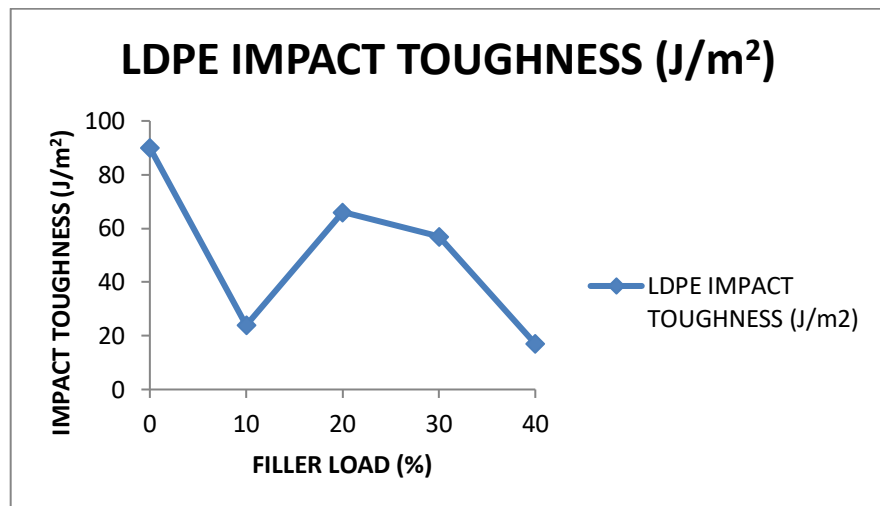


Fig. 4. Effect of filler loading on the Impact strength of LDPE composites

A significant decline was observed at 10wt% filler loading, where the impact toughness dropped drastically to around 25 J/m². This reduction can be attributed to poor interfacial adhesion between the filler and the polymer matrix, which promotes stress concentration and reduces energy absorption capacity (Idris *et al.*, 2020). An improvement in impact toughness was recorded at 20 wt% filler loading, suggesting better filler dispersion and interfacial bonding at this concentration, which could enhance energy dissipation during impact. Beyond 20 wt%, a gradual decline in impact strength was observed up to 40 wt%, likely due to filler agglomeration and matrix embrittlement, which hinder stress transfer efficiency and create microvoids that act as crack initiation sites (Owonubi *et al.*, 2020). Overall, the result indicates that moderate filler loading (around 20 wt%) enhances impact performance, while excessive filler addition deteriorates the toughness of LDPE composites. This is in line with the observations of Raj *et al.* (2023) that optimal filler concentration enhances polymer toughness through improved filler-matrix compatibility, while higher loadings lead to brittleness and reduced ductility.

Hardness strength

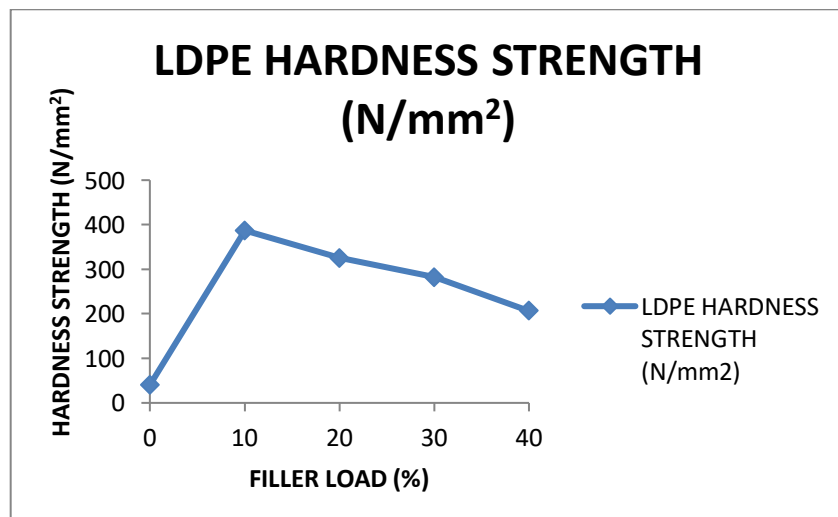


Fig. 5. Effect of filler loading on the Hardness strength of LDPE composites

As observed, the hardness strength initially increased sharply from the neat LDPE value (0% filler) to its peak at 10 wt% filler loading, after which it gradually declined with further increases in filler content up to 40 wt%. The initial increase in hardness strength can be attributed to the efficient dispersion of filler particles within the LDPE matrix, which enhances rigidity and resistance to deformation (Abdul Khalil *et al.*, 2012). At this stage, good interfacial adhesion between the filler and polymer matrix likely restricted the movement of polymer chains, leading to improved surface hardness (Nwabanne *et al.*, 2017). However, beyond 10 wt% filler loading, the hardness strength decreased progressively, indicating that excess filler led to particle agglomeration and poor stress transfer efficiency within the matrix (Osei *et al.*, 2020). Such agglomeration creates weak points that act as stress concentrators, thereby reducing the overall resistance of the composite to indentation. This is in line with the works of Eze *et al.* (2019), who reported that excessive filler content often results in poor matrix-filler interfacial bonding and increased void formation, thereby lowering hardness.

Thermal analysis

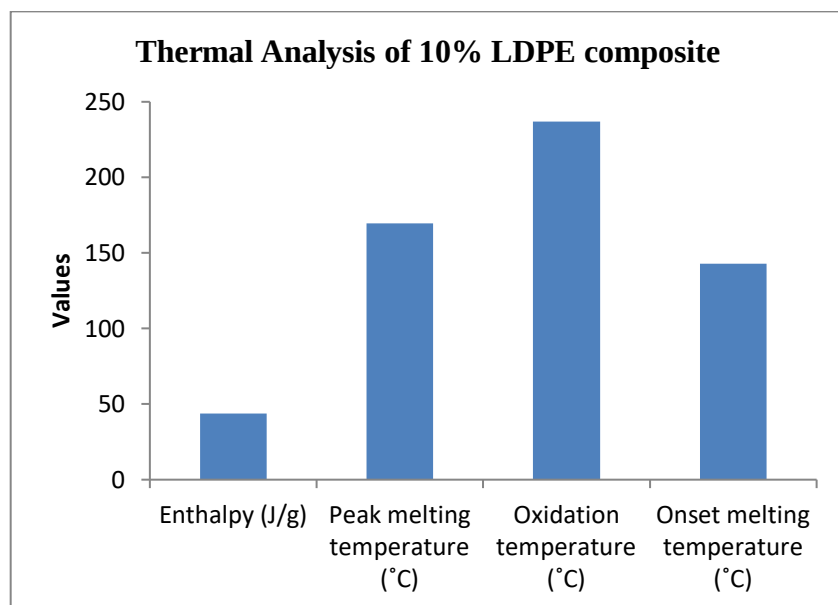


Fig. 6a: DSC thermogram of 10wt% LDPE composite

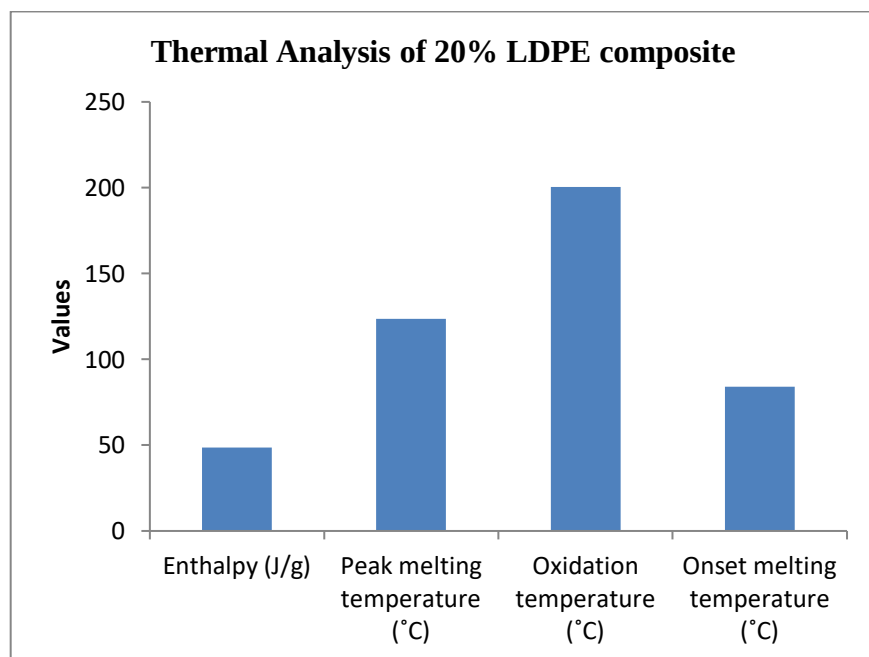


Fig. 6b: DSC thermogram of 20wt% LDPE composite

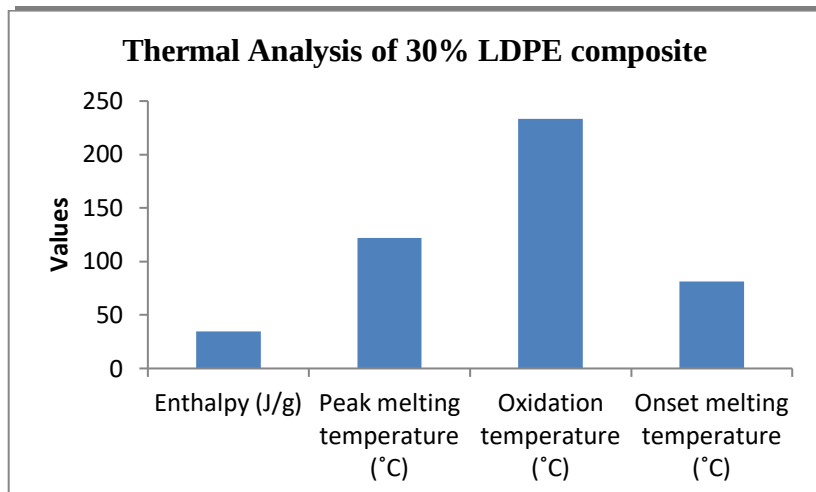


Fig. 6c: DSC thermogram of 30wt% LDPE composite

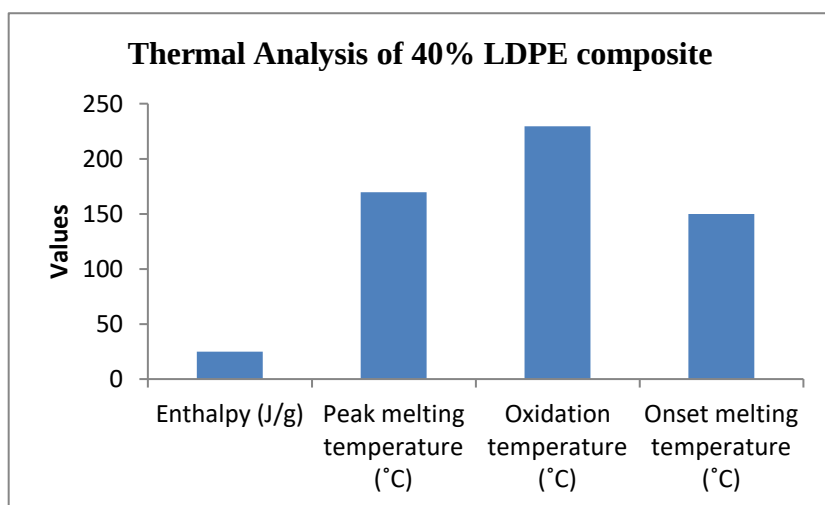


Fig. 6d: DSC thermogram of 40wt% LDPE composite

From Figures 6a-d, the DSC results showed that the LDPE composites generally exhibited melting transitions within 120-170°C and oxidation stability in the range of 200-237°C. This is in agreement with the works of Zhang *et al.*, (2018) that LDPE composites typically display melting peaks around 120-135°C and oxidation temperatures above 200°C depending on filler interaction. The observed enthalpy values reflect variations in crystallinity, which is consistent with findings that filler loading can either promote or restrict chain packing (Joseph *et al.*, 2020). At low filler loading (10wt%), higher melting (169.53°C) and oxidation temperature (236.95°C) suggest enhanced stability and this is in agreement with Essabir *et al.*, (2013), that reported improved thermal resistance at low filler levels. However, the sharp reduction in melting temperatures at 20-30wt% contrast with George *et al.* (2016), who found more gradual changes with increasing filler. The decline in enthalpy at 30-40wt% indicates imperfect crystallization, aligning with Singh *et al.* (2017), who observed reduced crystallinity in overloaded composites.

Surface morphology study

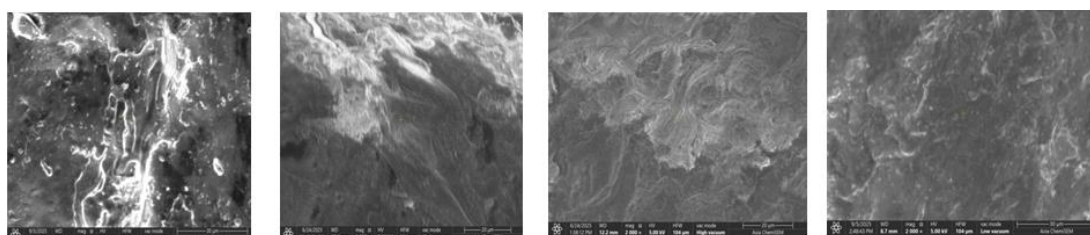


Fig. 7a LDPE 10% LDPE 20% LDPE 30% LDPE 40%

From Figure 7a, 10-20wt% showed visible voids, micro cracks distributed across the matrix. Such features can be due to inadequate filler-matrix adhesion and phase separation at lower filler loadings (AlMaadeed *et al.*, 2012). The elongated streaks and uneven dispersion suggest partial agglomeration of fillers, which may hinder uniform stress transfer during mechanical loading. The moderate roughness observed in these micrographs is advantageous for biodegradation. Surface irregularities and voids increase the accessible surface area for microbial attack and enzymatic penetration, facilitating gradual breakdown of the polymer composite (Ojijo and Ray, 2013). This aligns with the work of Chiellini *et al.* (2003) that composites with discontinuous morphologies tend to degrade faster due to increased hydrophilicity and moisture uptake pathways created by interfacial defects. The higher filler loadings (30-40 wt%) showed good dispersion and this can be due to matrix-filler compatibility which improved the interfacial adhesion and minimized agglomeration.

Solvent imbibition study

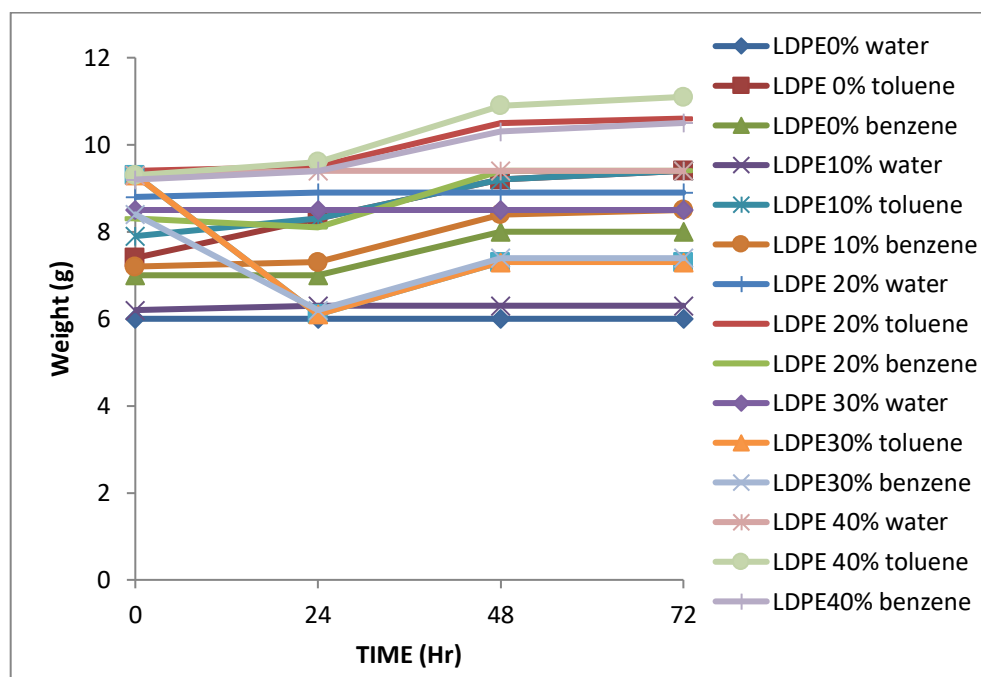


Fig. 8: Effect of filler loading on the solvent imbibition properties of LDPE composites.

The solvent imbibition behavior of low density polyethylene composites varied with solvent type, filler loading, and immersion time. Low density polyethylene showed very low affinity for water at room temperature for seventy-two hours. This is in line with the works of Ogudo *et al.*, (2021); Nwokoye *et al.*, (2024) that observed no water adsorption by the hybrid filler. Thus, the non-absorption of water by the composites could be due to its hydrophobic nature, with only slight weight change attributed to surface adsorption. In contrast, higher absorption was observed in toluene and benzene, consistent with their non-polar character and closer solubility parameters to low density polyethylene (Sivakumar and Rajini, 2016). Benzene exhibited the highest uptake, indicating stronger polymer-solvent interaction. On the other hand, filler content also influenced solvent uptake. At lower loading (0-10 wt%), absorption was relatively low due to reduced free volume and better polymer-filler adhesion. Intermediate loadings (20-30wt%) showed higher uptake, likely from microvoids and weak interfacial bonding that facilitated solvent diffusion (Thakur *et al.*, 2014). At higher filler content (40 wt%), solvent absorption stabilized, suggesting restricted chain mobility and limited penetration. Moreover, weight gain increased with immersion time and tended towards equilibrium after 48-72 hours, characteristic of fickian-type diffusion (Sreekumar *et al.*, 2007). Overall, low density polyethylene composites demonstrated good resistance to water but significant swelling in organic solvents, implying suitability for wet environments but reduced stability in hydrocarbon-rich conditions.

Biodegradation test

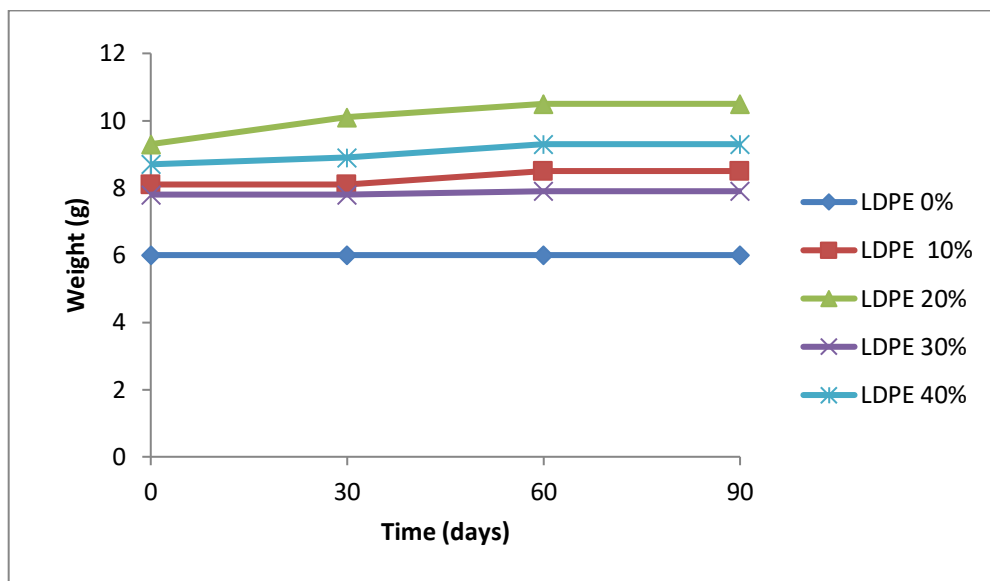


Fig. 9: Effect of filler loading on the degradation of LDPE composites

From Figure 9, there was no reduction in weight for 0% LDPE during the 90 days test periods. For 30wt% composites, minimal weight loss was noticed indicating limited microbial attack and surface oxidation. This is in line with the works of Arutchelvi *et al.*, (2008) that pure polypropylene undergoes very slow biodegradation due to its hydrophobic backbone and high crystallinity. This observation corroborates the research findings of Narancic *et al.*, (2018) that incorporating LDPE into biodegradable matrices slows the overall degradation. The minimal degradation exhibited as the filler loading increased could be attributed to the organic nature of the fillers, which attracted the presence of micro organisms. These microbes will usually attack the sites of fillers interaction thereby creating voids in the composites framework, hence a collapse of the framework after sometime.

CONCLUSION

Mixed agro-waste fillers effectively reinforced LDPE composites, improving mechanical and thermal properties at moderate loadings (20-30 wt%). However, high filler loadings compromised performance due to agglomeration. Limited solvent resistance and biodegradation highlight the need for compatibilizers and further modification.

RECOMMENDATIONS

1. Employ coupling agents (e.g., maleic anhydride grafted polyethylene) to enhance interfacial adhesion.
2. Long-term biodegradation under real environmental conditions should be evaluated.

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