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AGRO-WASTE FILLERS: IMPACT ON MECHANICAL, MORPHOLOGICAL, SOLVENT IMBIBITION AND DEGRADATION PROPERTIES OF LOW DENSITY POLYETHYLENE.

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Abstract
This research is aimed at studying the impact of agro-waste filler on the mechanical, morphological, solvent imbibition and degradation properties of low density polyethylene. The fine agro waste of cassava peel-cowpea hull of 75μm particle size was blended in a 1:1 ratio then integrated into LDPE matrix at different weight percentages (0, 5, 10, 15, and 20) and composites fabricated via injection moulding technique. The mechanical properties; tensile strength, percentage elongation at break, compressive strength, shear modulus and hardness test were studied in line with ASTM standards. The morphological property was studied with Scanning Electron Microscope. Degradation study was done using soil burial method for 180 days. The mechanical properties results showed that there was an increase in tensile strength, percentage elongation at break, hardness and shear modulus of the composites with increasing filler loading. The compressive strength decreased with increasing filler loading. Morphological study of the composites showed a good adhesion and interfacial bonding between the filler and the polymer matrix due to good dispersion of the fillers in the polymer matrix. Degradation test showed a decrease in the mass of the composites after 180 days burial period indicating that the composite is degradable. The solvent imbibition test showed no increase in the mass of the composites after immersion in water for 72 hours at room temperature, indicating that the composite can be utilized in wet environments. Thus, from the findings, it is suggested that agro-wastes such as cassava peel and cowpea hull be employed as fillers in the manufacture of plastics like packaging containers, shoe soles. This is because the fillers are cheaper and viable and the produced plastics would degrade when discarded.
Keywords: *Cassava peel-cowpea hull, Low Density Polyethylene, Mechanical, Morphological, Degradation Properties.*

Introduction

The world population is greatly increasing as well as human activities and food consumption rate. This increase has led to generation of more agricultural and polymeric products with consequent creation of increased garbage (both agricultural and polymeric) that leads to environmental pollution with resultant health effect. Thus, the necessity to encourage green environment by formulation of technique that will enhance products standard which will be less harmful to life. This can only be achieved by combination of the polymer matrix with cheap, readily available and eco-friendly organic fillers to make bio-composites. The incorporation of agricultural waste as a natural, biodegradable filler in polymer composites has received global attention for more than two decades now. Ogudo, Chris-Okafor, Nwokoye and Anekwe, (2021) worked on mixed agro-waste biocomposites of low density polyethene; impact of fillers on mechanical, morphological, water imbibition and biodegradability properties. From the results, there was a reduction in tensile strength of the composites with increasing filler loading and increase in the percentage elongation at break of the LDPE composite as the filler loading increased. The hardness, compressive strength and shear modulus of the polymer composites increased with increasing filler loading. Morphological study of the composites showed a good adhesion and interfacial bonding between the filler

and the polymer matrix due to good dispersion of the fillers in the polymer matrix. Biodegradation test showed a reduction in the mass of the composites, while the water imbibition test showed no increase in the mass of the composites after immersion in water. Onyenweaku, Nwokoye and Chris-Okafor, (2021) studied the mechanical and surface morphology properties of low density polyethylene composites filled with organic materials. Blends of coconut husk and mango seed shell were used as agricultural wastes. The results showed that the fillers had positive impacts on the flexural strength and hardness, while there were negative impacts on tensile strength and compression strength.

Nwokoye, Okoye and Chris-Okafor, (2024) worked on impact of hybrid biomass fillers on the physico-mechanical and degradation properties of utility polymers. Varying percentage filler load of hybrid cassava peel-rice husk filler on low density polyethylene and polypropylene was adopted. The results showed improved tensile strength, compressive strength and hardness, reduced percentage elongation at break, irregular shear modulus, zero effect on LDPE creep rate, irregular creep behavior for PP composites and improved degradation of the composites.
This research studies the agricultural waste around us, hence the use of cassava peel and cowpea hull mixed agro waste filler with polyethylene matrix.

Materials and Methods

Materials

Low Density Polyethylene, LDPE (NGL015FS) produced by Indorama Eleme Petrochemicals, Nigeria. Cassava tubers and Cowpea were obtained from the family farm at Ebenator Ezeoye, Nibo, Awka South LGA. Anambra State, Nigeria. The tubers were manually peeled to detach the peel from the flesh, while the hulls were detached manually from the seed. The agricultural waste materials were cut into smaller fragments, washed and air dried for fourteen days before being crushed and sieved locally and repeatedly using a grain mill machine M6FFC-270 and muslin cloth respectively to a fine powder of 75μm mesh size.

Preparation of Composites

The fine powders of cassava peel and cowpea hull were homogeneously mixed at the ratio of 50:50 to form the hybrid filler. Varying weight percentages of the hybrid fillers (0,5, 10, 15 and 20) corresponding to 0g,10g, 20g, 30g and 40g were mixed with virgin crystalline pellets of LDPE of corresponding weights 200g,190g, 180g, 170g and 160g respectively. The polymer matrix and filler mixture was put into the hopper of a TU150 200gram injection moulding machine. Production of the composites at 288^{o}C took average of 33 seconds.

Mechanical Properties Analysis of Composites

The mechanical properties were studied according to American Society for Testing and Materials (ASTM) standard and include; tensile strength, hardness, shear modulus and compressive strength.

Tensile Strength

This is the measure of the resistance of a material to breaking under tension. The tensile properties of the composites were measured according to the ASTM D-638-14, using Hounsfield Monsanto Tensometer 8889, Made in England. The test piece was measured to 160mm x 19mm x 3.2mm dimension.

Hardness Test

Hardness explains the resistance of material to permanent indentation or scratching. The surface hardness of the polymer composites were measured according to the ASTM D2240 using Shore Scale Durometer Hardness Tester, Made in England.

Shear Modulus

This measure the material's rigidity as it shows the amount of force that can tear the composite apart. The shear modulus of the composites was measured according to the ASTM D-732, using the Hounsfield Monsanto Tensometer 8889 Made in England. The test piece was measured to 20mm x 20mm.

Compressive Strength

Compressive strength refers to composite's ability to withstand compression or pressing. The compressive strengths of the composites were measured according to the ASTM D-695 using the Hounsfield Monsanto Tensometer 8889 Made in England. The test piece was measured to 20mm x 20mm. The readings were automatically recorded and the values computed with the formula;

$$Compressive\ Strength, \frac{N}{mm^2} = \frac{maximum\ force, P(N)}{cross\ sectional\ area, A_0(mm^2)} \qquad Eqn.\ 2.1$$

Morphological Analysis

This is used to determine the arrangement or compatibility of the blends with the matrix**.** Morphological studies of the composites were conducted using Scanning Electron Microscope (SEM) model: JEOL-JSM 7600F.

Degradation Test

This test determines the rate of degradation of the composites in the environment. It was determined by burying the composites 10cm depth into soil obtained from an automobile mechanic workshop mixed with poultry waste. The composites were weighed at 30 days interval to see the extent of degradation till it completed 180 days. Degradation was measured from the mass reduction of composites buried.

Solvent Absorption Test

This illustrates the rate of solvent absorption of the composites as determined by ASTM D-570-98. The composites were cut into 40mm x 40mm x 3.2mm dimension, dried and weighed before being immersed in deionized water for seventy-two hours (3 days) at room temperature. Then, the respective weights of the composites were checked after a period of 24 hours to ascertain weight gain/loss.

Result and Discussion

Mechanical Properties

The results of the mechanical properties of the LDPE composites with cassava peel-cowpea hull fillers are shown in the figures below.

Tensile Strength

The impact of filler content on the tensile property of the composite is shown in Figure 1.

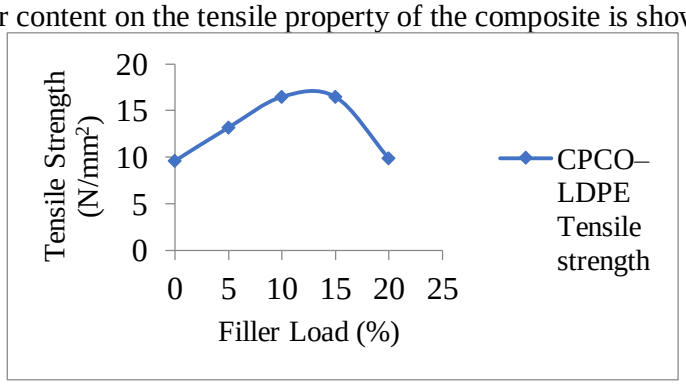


Figure 1. Impact of filler content on the tensile strength of LDPE composites.

The composite shows an increase in the tensile strength as filler content increases, having the maximum value observed at 15wt% filler load. . This increase in the tensile strength of the composite could be attributed to stronger adhesion between filler and matrix interface which leads to better stress transfer from the matrix to the filler. The decrease after attainment of peak shows weakening of the interfacial attraction of the constituent composition as the fraction of the matrix is reduced with increasing weight fraction of reinforcement which tries to alter the inherent properties of the matrix. This is in agreement with the works of Chris-Okafor *et al.*, (2018) and Nwokoye *et al*., (2024).

Percentage Elongation At Break

This measures the degree of ductility of the composite materials. The impact of filler content on the percentage elongation at break of the composite is shown in Figure 2.

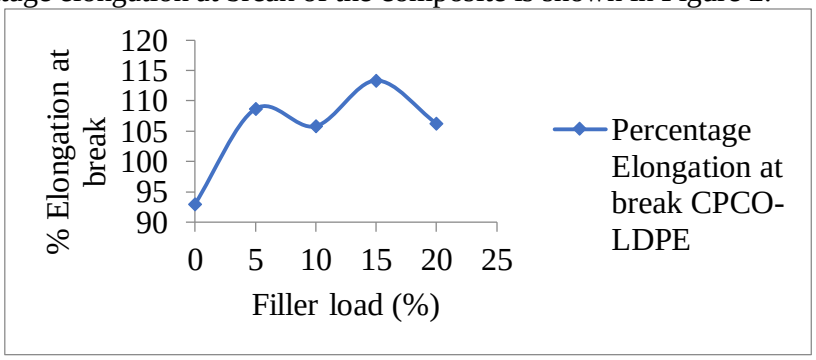


Figure 2. *Impact of filler content on percentage elongation at break.*

From Figure 2, the percentage elongation at break increased with an increase in filler loading. This indicates that increasing filler loading increased the ductility/ elasticity of the polymer composites. This increase indicates the filler ability to facilitate stress transfer from filler to matrix. This corroborates the work of Ogudo *et al.*, (2021).

Hardness

The impact of filler content on the hardness of the composite is shown in Figure 3.

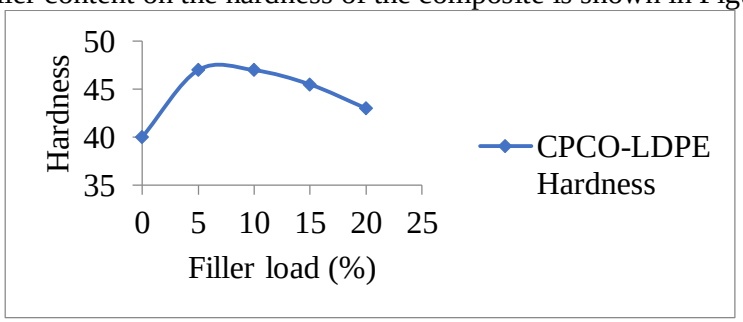


Figure 3. *Impact of filler content on the hardness of LDPE composites.*

From Figure 3, the hardness value as observed exhibited an overall increase as the filler loading increases. This is an indication that the fillers enhanced the hardness of the composites with the composite exhibiting the highest hardness value at 5wt% filler loading. The increase in hardness could be attributed to the filler's strengthening activity in the polymer matrix. Fillers are mostly used to improve the stiffness and strength of polymeric materials. This could possibly be due to the fact that adding fillers tightened the composites' elasticity and enhanced the matrix surface resistance to indentation which is in line with the work of Chris-Okafor *et al.*, (2018) and Onyenweaku *et al.*, (2021).

Compressive Strength

The impact of filler content on the compressive strength of the composite is shown in Figure 4.

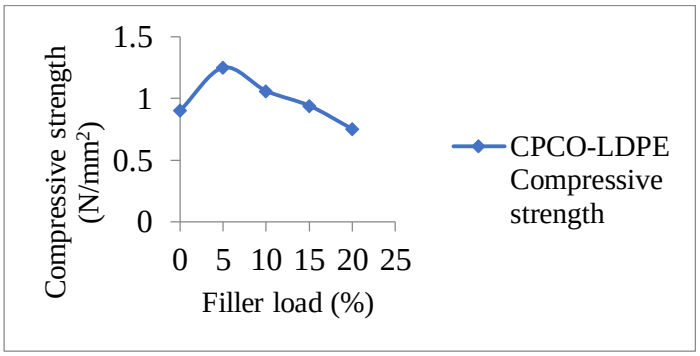


Figure 4. *Impact of filler content on the compressive strength of LDPE composites.*

Figure 4 shows that the composites' compressive strength increased at 5wt% before gradually and continuously decreasing. This decrease could be attributed to the filler poor interaction with the matrix which affected its reinforcing characteristics. This agrees with the work of Onyenweaku *et al.,* (2021).

Shear Modulus

Figure 5 shows the influence of filler content on the composites' shear modulus.

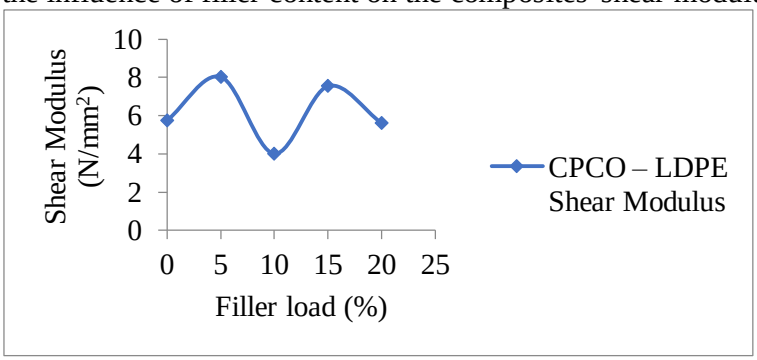


Figure 5. Impact of filler content on the Shear modulus of LDPE composites.

The shear modulus fluctuates as the filler loading increases but there was increased property. This increase suggests that the fillers increased the rigidity of the composites; a larger force is required to deform the composites along the plane of the direction of the force. This is in line with the work of Ogudo *et al.*, (2021) and Nwokoye *et al.,* (2024).

Morphological Analysis

Scanning electron microscopy (SEM) is an effective media for the investigation of the surface morphology of the composites as it studies the dispersion and compatibility of the filler and the polymer matrix. It also relates the differences in the mechanical properties of the composites. The results for the analysis of the morphology of the composites were obtained as follows. Figures 6(a), 6(b), 6(c), 6(d) are the micrograph of CPCO-LDPE 0wt%, 5wt%, 10wt%, and 15t%, respectively.

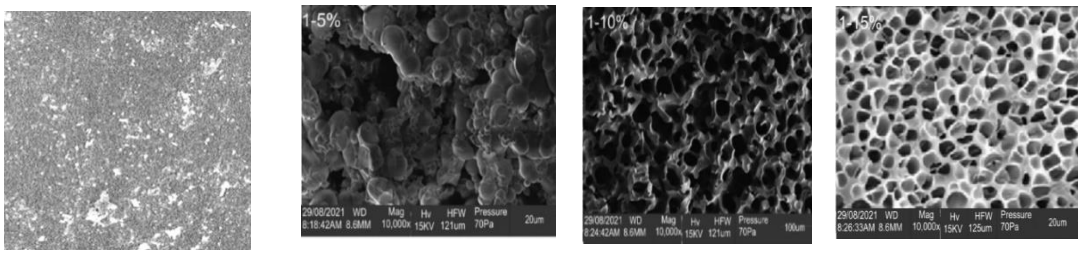


Figure 6(a) 0% LDP (b) 5% CPCO-LDPE (c)10% CPCO-LDPE (d)15% CPCO-LDPE

Figure 6a showed the morphology of LDPE without any filler as homogeneous surface, while Figures 6b-d showed the morphology of the composite as the filler load increased from 5wt% to 15wt%. From the Figures, there was observed presence of globular shaped filler in the composite with pronounced voids at 5wt% filler loading which indicates that the interfacial bonding between the fibre and the matrix polymer was weak, but improved as the filler loading increased to 10wt% and gave the best result at 15wt%. At 15wt% filler load, it could be observed that the fillers dispersed well in the matrix and formed a good network. Generally, as the filler loading increased, the filler dispersion improved and perfectly bonded to the matrix with the formation of an interphase. The high interfacial contact between the hybrid fillers and the polymer matrix was attributed to the high mixing efficiency of the cassava peel and cowpea hull. The even dispersion of the fillers in the polymer matrix improved the compatibility of the composites. Thus, CPCO-LDPE composite that showed higher tensile strength, was because the filler particles were well dispersed in the polymer matrix as both had excellent adhesion. This is in agreement with the works of Chan *et al.*, (2016); Jumaidin *et al.*, (2017).

Biodegradation Test

This was carried out to determine the extent the composites will degrade in the environment over a period. Figure 7 reports the degradation test results of LDPE composites.

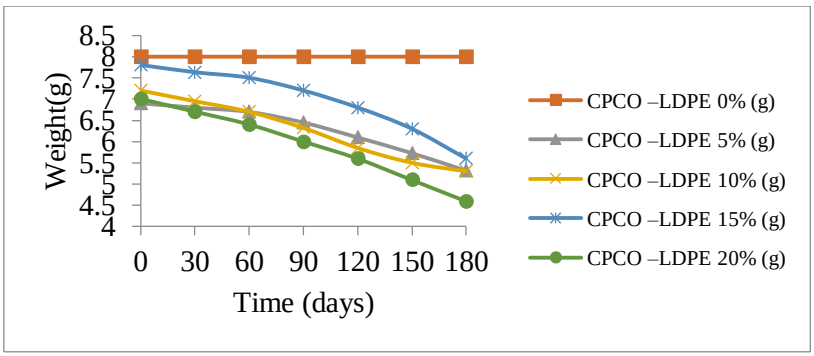


Figure 7. Impact of filler loading on the degradation of LDPE composites.

From Figure 7, there was no reduction in weight for 0% CPCO-LDPE during the 180 days test period, but there was a general reduction in weight with other composites as the burial time increased. The initial weight loss was noticed after 30 days test period for all the composites. It was also observed from the Figure that as the filler content increases, the degradation rate also increases. This observation corroborates the research findings of Ogudo *et al.*, (2021); Nwokoye *et al.*, (2024). These authors opined that biodegradability of composite is more enhanced and pronounced if the composite contains higher percentage of the natural fillers. The highest degradation rate was observed at 20wt% filler loading giving a 15% reduction after 180 days.

Solvent Imbibition Test

There was no weight gain for all the LDPE composites in water during the 72 hours test period. This corroborates with works of Ogudo *et al.,* (2021) and Nwokoye *et al.,* (2024).

Conclusion

Cassava peel and cowpea hulls were successfully incorporated as hybrid fillers into low density polyethylene (LDPE) polymer matrix to make polymer composites. The incorporation of the filler enhanced the tensile strength, percentage elongation at break, hardness, and shear modulus of the composite. However, the compressive strength increased before decreasing. This finding is in agreement with existing literature and shows that this is a common occurrence with lignocellulosic fillers which is in agreement with Nguyen *et al.,* (2017) that observed hybrid composites of all natural fibres to generally exhibit satisfactory strength and can be used for various applications. The synergy between the polymer matrix and the filler, as well as the particle size and dispersion of the fillers within the matrix, was discovered to influence the mechanical properties of the composites. The composites degradation rate improved as the filler load increase as evidenced in their weight loss during the one hundred and eighty (180) days test period. The composites were found to resist water absorption over the testing period. Thus the composite is ideal for use as in production of water storage tanks as well as pipes and fittings.

From the result of the findings, it is suggested that agro-wastes such as cassava peel and cowpea hulls be employed as fillers in the manufacture of plastics like water storage tanks, pipe and fittings, because they are cheap, eco-friendly and readily available.

Recommendations

1. Surface modifications of the agro-waste is recommended to enhance performance and promote better adhesion between the natural reinforcement and the polymeric matrix.
2. Study on varying the biomass filler ratio with constant matrix percentage is recommended.
3. Study of this research topic with other techniques such as compression moulding.

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