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Nigerian Journal of Textiles (NJT)

A Publication of Textile Researchers Association of Nigeria (TRAN) Volume 5 August 2019

http://tran.org.ng

# Effect of Coupling Agent on the Mechanical Properties of High-Density Polyethylene-Filled Calabash Particles Composites

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# ABSTRACT

In the interest of sustainable development and production of eco-friendly bio-composites, the use of natural fillers with synthetic resins in the production of composites has been the new trend in composite manufacture. In this study, calabash particles (CP) were used as filler in high density polyethylene (HDPE) to produce bio-composites. The calabash which was grinded and sieved into 125µm was introduced at 10% variation up to 50% filler loading followed by compounding and thereafter compression moulded. Two types of composites were prepared, in one, Maleic Anhydride (MA) as coupling agent was introduced at 3% on the weight of the filler together with an initiator Benzoyl Peroxide (BP) at 1% to enhance the interfacial adhesion between the CP and HDPE and in another no coupling agent was used. The result of tensile, flexural, impact strength hand tensile modulus shows increase from 0 to 40% filler loading of composites with coupling agent representing 21.61, 32.11, 63.16 and 42.97% increase respectively, but shows decreasing trend for composites without coupling agent with the exception of tensile modulus and hardness which increased from 0 to 50% filler loading representing 71.9% increase and 76.46% increase respectively. Also, SEM micrographs for the fractured surfaces show even dispersion of the filler particles in the composite with coupling agent and show signs of agglomeration for composites without coupling agent.

Keywords: Calabash Particles, HDPE, Tensile Strength, Flexural Strength, Impact Strength

# **INTRODUCTION**

Calabash (Lagenaria Ciseraria) is simply the dried hollow shell of a gourd used for household utensils [1]. The fresh fruit has a light-green smooth skin and a white flesh. Rounder varieties are called calabash gourds [2]. Thermoplastic polymer has formed a new trend in material development, and it has the advantages of low production cost, great diversity, sufficient sources, light weight, good physical properties, and chemical resistance, as well as various efficient manufacturing process. Therefore, there are a great number of plastic products commercially available [3]. Plant fibres are biodegradable and readily available when compared with glass fibres [4]. Most plastics are not suitable for load bearing applications by themselves due to their insufficient strength, stiffness or dimensional stability [5].

In recent years, wood fibre-reinforced plastic composites or so-called wood-plastic composites (WPCs) have attracted significant interest and they are replacing wood in many fields [6], better stability and favourable mechanical properties has caused WPCs to become a preferred building material [7].

The recent world population growth has been one of the main factors for the technological development and innovation. In fact, exponential growth has taken place in the world population, being expected to reach 9 billion in 2042, compared to 3 billion in 1960 [8].

Contrary to human needs, the earth's resources are limited. In the era of rapidly growing population and global warming, environmental issues cannot be neglected in almost all the fields of engineering. Both the industry and researchers try to utilize renewable resources in more and more products and processes. The idea of the replacement of synthetic fibres with natural-based fillers in polymer composites has been incorporated since 1970s and has recently gained popularity [9].

Eco-friendly bio-composites consisting of agrofillers and bio-based plastics are of great importance to the materials and research world as a feasible solution to growing environmental threats and as a sustainable solution to the uncertainty of the world's petroleum supply [10].

Dissimilarity of the surface chemistry between the filler and polymer usually contribute to the decrease of the tenacity in filler–polymer systems [11] CP are hydrophilic while HDPE is hydrophobic.

Therefore, in order to reduce the nonbiodegradability of synthetic polymers, biodegradable fillers can be incorporated into synthetic polymers to produce composites that are of biodegradable, low cost and eco-friendly.

## MATERIALS AND METHODS

## Materials

High Density Polyethylene (HDPE)(Zayo-Sigma Chemicals Ltd. Jos), Maleic Anhydride (MA) 98% purity (coupling agent), Benzoyl Peroxide 98% purity (initiator), Calabash Particles (CP), Retsch Sieve Shaker Machine (Emdocatt. Model: 7416), Laboratory Mill (Model 4, Arthur H.

Thomas Company, PA., U.S.A.), Vacuum Oven (model: 60648, Cole Parmer), metallic mould, Two-roll Mill (Model: 5183 North Bergen U.S.A), Compression Moulding Machine (model: 0557) Wenzhou Zhiguang Shoes making machines CO., Ltd. China.

#### Methods

## **Collection of Materials**

Calabash bowls (dried seeds and whitish material removed) were obtained from Zurmi, in Zurmi Local Government Area of Zamfara State, Nigeria. All have varying sizes of between 10 to 15 litres in volume. The calabash bowls were broken into smaller pieces which were then fed into a laboratory milling machine and further crushed into smaller particle sizes. The ground calabash particles from the milling machine was removed from the sieved particles in an oven until a constant weight was obtained thereafter stored in a desiccators.

## **Preparation of the Composites**

The composites were prepared from the calabash particles and HDPE at 10% varying filler loading up to 50% while the HDPE was reduced simultaneously from 100% down to 50%, during compounding using a two-roll mill. The composite was prepared in two forms, one with coupling agent (CP/HDPE A) and the other without coupling agent (CP/HDPE B). Two roll mill was used for compounding and thereafter compression moulded and cooling of the composites were carried out for 10 minutes each.

Table 1: Percentage composition of CP (125 $\mu$ m) and HDPE in each of the composite formulation at different filler loadings

		HDPE
S/NO.	CP (%)	(%)
1	0	100
2	10	90
3	20	80
4	30	70
5	40	60
6	50	50

#### **Tests for the Composites**

#### Tensile Strength (ASTM D-638)

Tensile strength test was carried out according to ASTM D-638 standard with multifunctional electric tensile strength tester (YG026D) at Standards Organization of Nigeria in Kaduna. The machine loadcell of 300kg capacity was used for each of the samples. The test sample dimensions were 50mm x 10mm x 4 mm.

#### Flexural Strength (ASTM D-790)

Flexural strength test was carried out according to ASTM D-790 standard with Universal Material Testing Machine (Cat Nr. Model: 26) at the Department of Mechanical Engineering, ABU, Zaria. The machine has a maximum force of 1K, and sample dimensions were 100mm x 25mm x 8mm

## Impact Strength (ASTM D-256)

Impact strength test was carried out according to ASTM D-256 standard with Charpy Impact Testing Machine at The Department of Metallurgical and Materials Engineering, ABU, Zaria. The machine load of 15J capacity was used for each of the samples. The test sample dimensions are of 80mm x 10mm x 10 mm.

## Hardness Test (ASTM E-384)

The hardness test was carried out according to ASTM E-384 standard with a Vickers Hardness Tester Machine Model MVI-PC YADRAN-416145, INDIA, using a diamond indenter at Shell Professorial Building, ABU, Zaria. The machine load of 500gf at 15 sec was used for each of the samples whose dimension is 30mm x 25mm x 8mm (length, width and thickness). The test was repeated three times and an average test result was calculated with its mean value and standard recorded

#### Scanning Electron Microscopy Test (SEM) (ASTM E766-98)

SEM test was carried according to ASTM E766-98 standard using fractured surface of the tensile specimens with Pro: X: Phenom world. Model number 800-07334 at the Department of Biology, Umaru Musa Yaradua University, Katsina. To impart conductivity to the polymer composites, a sputter machine used 5nm gold to coat the sample surface. The samples were inserted into the machine using the sample holder and the morphology of the samples were viewed at 15KV, and Images were obtained at 1000X magnifications.

## **RESULTS AND DISCUSSION**

## **Tensile Strength**

From Fig. 1 the tensile strength of CP/HDPE composite with coupling agent can be seen to be increasing with increased filler loading from 27.54 MPa of the neat polymer to 35.13MPa at 40% filler loading i.e. 21.61%. The increase might be attributed to the interfacial adhesion between the filler and the matrix. Maleic Anhydride was introduced as coupling agent at 3% on the weight of the filler, it might have tailored the properties of the composite by increasing bonding between filler and matrix. The incompatibility of polar wood fibres and non-polar polyolefin leads to difficulties in obtaining uniform dispersion of wood fibres in the matrix, which in turn reduced the efficiency with which the fibres reinforce the polymer, but introduction of coupling agent into a composite system can improve mechanical properties [12].

At 50% filler loading, it can be observed that the tensile strength dropped by about 50%, this might be as a result of the matrix losing its binding ability [13]. The matrix might be approaching its saturation level which occurred at 60% filler loading based on preliminary investigation.

Composites without coupling agent show a decreasing trend from 27.54 MPa of the neat polymer to12.20MPa at 50% filler loading, the decrease might be attributed to poor interfacial adhesion between the filler and matrix [13].

## **Tensile Modulus**

From Fig. 2, it can be observed that tensile modulus of CP/HDPE composite was increasing for both composites with and without coupling agent from 0.88GPa of the neat polymer to 1.21GPa (42.97% increase) and 1.11GPa (37.84% increase) for composites A and B respectively. The increase in tensile modulus might be attributed to the stiffening effect caused by introduction of rigid fillers which might have reduced the polymer chain mobility consequently increasing modulus [14].

## **Flexural Strength**

Fig. 3 shows similar trend with tensile strength, the flexural strength of CP/HDPE composite A was increasing with increase in filler loading from Nigerian Journal of Textiles (NJT) Vol. 5: 11 - 16

43.47MPa of the neat polymer to 64.03MPa at 40% filler loading representing 32.11% increase. The increase in flexural strength might be attributed to the incorporation of maleic anhydride as coupling agent which might have improved the bonding between the CP and HDPE. Increase in interfacial bonding between filler and matrix improved mechanical properties [15], because improved filler matrix interaction provides effective transfer of stress from matrix to filler [16]. For composites without coupling agent there is a decreasing trend from 43.47MPa of the neat polymer to 26.42MPa (39.22% decrease) at 50% filler loading, the decrease might be attributed to poor interfacial adhesion between the filler and matrix [15].

## **Impact Strength**

The result of impact strength is shown in Fig. 4, there is increase in impact strength with increase in filler loading of the composite for CP/HDPE composites A from 35J/m of the neat polymer to 95J/m at 40% filler loading representing 63.16% increase. This might be attributed to improved interfacial interaction between the filler and matrix when coupling agent was introduced in the composite system, maleic anhydride has the property of covalent bonding on to another polymer chain [17]. Introduction of coupling agent into a composite system with dissimilar surface chemistry between the filler and matrix improves interfacial adhesion and consequently improves mechanical properties, increase in impact strength has been reported by several researchers due to the incorporation of coupling agent into a composite system [18].

Meanwhile, for composites without coupling agent, impact strength was found to be decreasing with increased filler loading from 35J/m of the neat polymer to 12J/m (65.71% decrease) at 50% filler loading. The dissimilarity between surface chemistry between filler and matrice brings about reduction in the tenacity of composites [16], decrease in impact strength my result from the noninclusion of coupling agent in the composite.

## Hardness

Hardness of CP/HDPE composites A and Bwas found to be increasing with increased filler loading from 9.37Hv of the neat polymer to 44.50Hv (71.9% increase) and39.80Hv (76.46%) increase for composites with and without coupling agent respectively. Hardness of a material is dependent on stiffness of that material, introduction of rigid fillers impart stiffness to synthetic polymer materials consequently increasing hardness [19].



Fig. 1: Tensile strength of CP/HDPE compositesA and B at varying filler loading



Fig. 2: Tensile modulus of CP/HDPE compositesA and B at varying filler loading



Fig. 3: Flexural strength of CP/HDPE composites A and B at varying filler loading



Fig. 4: Impact strength of CP/HDPE composite A and B at varying filler loading



Fig. 5: Hardness of CP/HDPE compositesA and B at varying filler loading

#### Scanning Electron Microscopy



Fig. 5: SEM image of CP/HDPE composite A at 40% filler loading with coupling agent



Fig. 6: SEM image of CP/HDPE composite B at 40% filler loading without coupling agent

Fig. 5 and 6 shows the SEM images of CP/HDPE composite at 40% filler loading with and without coupling agent, since 40% filler loading gave the best mechanical property, fractured surface of the tensile test specimen was used for SEM determination.

From the micrograph in Fig. 5, it can be observed that there is an even distribution and dispersion of the CP in the composite with coupling agent. One advantage of using coupling agent in composites is that it helps in making the reinforcing fillers to be fully embedded in the matrix thereby preventing debonding which leads to plastic deformation of composites [19]. Meanwhile, in Fig. 6, the SEM image shows areas of agglomeration and uneven dispersion of the filler in the composite without coupling agent which might be the reason why there is reduction in the mechanical properties of composites without coupling agent.

#### CONCLUSION

The effect of coupling agent on the mechanical properties of CP/HDPE composites was studied to determine the viability of maleic anhydride in improving the mechanical properties of composites. The result of tensile, flexural, impact strength, show increasing trend from 0 to 40% filler loading which might be due to incorporation of maleic anhydride as coupling agent but shows reducing trend with composites without coupling agent. Meanwhile, tensile modulus and hardness where increasing with increased filler loading for composites with and without coupling agent. The SEM image of CP/HDPE composite A at 40% filler loading shows even distribution of fillers in the matrix which is a sign of proper bonding between the filler and matrix preventing debonding, while, CP/HDPE composite B shows uneven dispersion of fillers with areas of agglomeration. From the result of the mechanical properties obtained, it can be deduced that the composites can be used for applications where low to moderate mechanical properties are required.

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