



# **IMPROVEMENT OF POLYETHYLENE MATRIX COMPOSITES USING COCONUT SHELL AND COW BONE PARTICULATES**

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## **ABSTRACT**

Utilisation of particles of coconut shell and cow bone as reinforcing materials for the production of low density hybrid polyethylene matrix composites by stir casting method was carried out. 50  $\mu\text{m}$  coconut shell and 50  $\mu\text{m}$  cow bone particulates in different proportions (5 – 25 wt. %) were mixed with polyethylene and the microstructural, physical and mechanical characterisations were determined using standardised methods. The hybrid composite exhibited desirable properties in terms of water absorption (0.3 %) indicating reduced pores/voids. It also exhibited ultimate tensile strength (1.78 MPa) and hardness (12.78 HBN) at 15 wt. % filler addition. The uniform dispersion of the reinforcing particles as observed in the SEM microstructure and the strong adhesion of the particles and polyethylene matrix contributed to the enhancement of the tensile strength and hardness of the composites. Increasing the filler concentration beyond 15 wt. % caused a decrease in the average inter-particle distance/spacing thereby increasing the amount of interparticle stress concentration overlap. This led to higher levels of debonding when tensile stress was applied. This ultimately impaired the tensile strength of the composites. The strain energy stored in the matrix which could be equal to the

adhesion/bonding of the particles and polyethylene matrix caused the particle-matrix interface to debond and reduced or impaired the modulus of elasticity of the composites.

Generally, the hybrid composites were better than the mono-reinforced composites and unreinforced polyethylene thus showing the efficacy of added particulates. Hence, the development of this biocomposites will reduce environmental pollution and also has the potential for application in areas where low strength composites are required.

**KEYWORDS:** Polyethylene composites; Coconut shell, Cow bone, Stir casting; Characterisation

## 1. INTRODUCTION

Water sachets and waste nylons are non-biodegradable polymeric materials which constitute environmental pollution in many developing countries due to improper disposal. These pollutants cause sewage blockage, blockage of road drainages causing flooding especially during the raining season and traffic congestion. In severe cases, it can result to accidents with disastrous consequences. Hence, proper disposal and more importantly, conversion of these waste into engineering materials for industrial application is a welcome development. The conversion and utilisation of these polymeric waste will lead to economic benefits and environmental pollution reduction/mitigation.

Polymeric materials are noted for their versatility, high resistance to chemicals, outstanding adhesion to a variety of substrates, toughness, high electrical resistance, durability at high and low temperatures, low shrinkage upon cure, flexibility, and the ease with which they can be poured or cast without forming bubbles (Brostow et al., 2010). Despite these advantages, polymers generally do not possess high impact energy and other mechanical properties thereby limiting their application in many areas. In particular, their strength and stiffness (modulus of elasticity) are low compared to metals and ceramics. In order to overcome these shortcomings, polymer matrix composites (PMCs) have been developed using different kinds of fillers.

The addition of such particulate fillers into polymers is primarily aimed at enhancing their processability, mechanical properties, as well as to reduce materials cost (Singla and Chawla 2010; Hemanth et al., 2014). Addition of fillers is favoured because it is a cheap, effective, and a fast method to modify the properties of the base material. The degree of improvement often depends on the type of filler (synthetic or natural), particle size and shape, filler content, and surface treatment which promotes interaction between the filler and the polymer matrix (Huerta-Martinez et al., 2005; Hemanth et al., 2014). Particulate fillers are used to modify the physical and mechanical properties of polymers in many ways. Various kinds of polymers and polymer-matrix composites reinforced with filler particles have a wide range of industrial applications such as electrical industries, commercial and military aircrafts, heaters, electrodes (Uygunoglu et al., 2015), floor covering, composites with thermal durability at high temperature (Kim et al., 2004).

Hence, reinforcing polymers with natural fillers for the development of PMCs is receiving much attention. The use of natural fillers for reinforcement of composites is receiving much attention. They have significant advantages over synthetic fillers such as low cost and density, comparable

specific tensile properties, ability to reduce abrasion of machinery, non-toxicity, reduced energy consumption, renewability, recyclability and biodegradability ([Malkapuram et al., 2009](#)). The use of particles as fillers is being encouraged because they are economical, effective, and are good for modifying the properties of polymers. PMCs are usually strengthened and hardened as a result of the uniform dispersion of volume or weight fraction of particles in the polymer matrix ([Hassan et al., 2012](#); [Bello et al., 2015](#)).

Many natural fillers have been investigated for use in industries such as flax, hemp, wood, rice husk (RH), snail shell, husks of wheat, barley, and oats ([Bijwe et al., 2001](#)). It has also been proven that the addition of wood dust to recycled polyethylene terephthalate (PET) enhanced the mechanical properties of the PET polymer matrix composites ([Rahman et al., 2016](#)). It has also been proven that reinforcement of recycled high density polyethylene (HDPE) and PET polymer matrix by rice husk particles led to an improvement in the mechanical properties of the composites ([Chen et al., 2016](#)). The addition of cow bone particles to recycled low density polyethylene (LDPE) enhanced the mechanical properties of the LDPE polymer matrix composites ([Agunsoye et al., 2013](#)). Also, reinforcing polyester resin matrix with cow bone particles resulted in improvement of the mechanical properties of the PMCs ([Isiaka and Adewole, 2013](#)). Natural fillers are now fast evolving as potential alternatives to inorganic or synthetic materials in various applications as building materials and automotive components ([Sarki et al., 2011](#)).

This work was aimed at studying the effects of weight percent (wt. %) of coconut shell micro-particles (CSMPs) and cow bone micro-particles (CBMPs) as reinforcing fillers on the water absorption, tensile strength, Young modulus, hardness, impact energy properties and morphology of polyethylene matrix biocomposites.

## 2. MATERIALS AND METHODS

### 2.1. Materials Preparation

The polyethylene (waste nylons) were obtained from the campus of the University of Lagos. The coconut shells were obtained from Oyingbo market in Lagos while the fresh cow bones were obtained from Bariga abattoir in Lagos, Nigeria. The bones were washed with detergent, rinsed with water and sun dried for 6 hours. They were then cleaned with ethanol and finally oven dried at 120 °C for 6 hours. All these were done to remove oil and other contaminants in the bones. The coconut shells and cow bones were separately ground to fine particles using a pulverizer and then sieved to 50 µm. Pictures of these materials are shown in [Fig. 1](#) while the

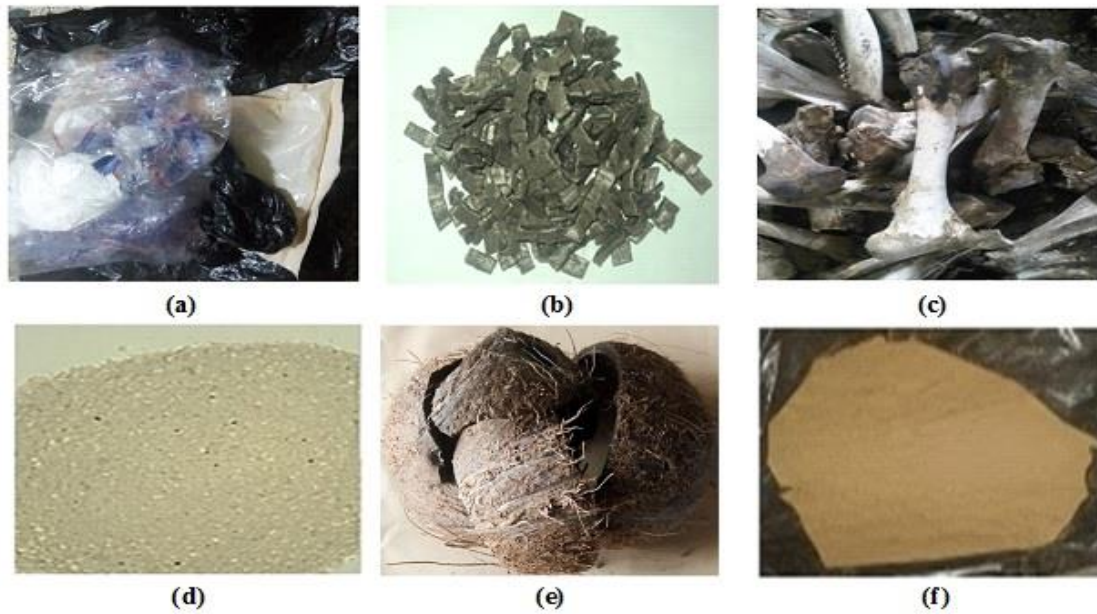
chemical composition of the cow bones obtained using an absorption spectrometer (AAS) is presented in [Table 1](#).

**Table 1. Chemical composition of the cow bone.**

Elements	Ca	Fe	Au	Sr	Sn
Weight (%)	23.298	2.586	0	0	0

## **2.2. Composite Specimens' Production**

The polyethylene (waste nylons) were washed in a detergent solution, rinsed in water, sun dried, shredded into pieces, ground using a pulverizer, put in 5 crucibles and heated in a muffle furnace at 150 °C for 15 mins. They were poured into a rectangular plate, pressed with a presser at 0.33 MPa for 5 mins and cut to small pieces (pellets). 300 g of the pelletized polyethylene was placed in a crucible pot, charged into a muffle furnace and heated to 200 °C until molten form was achieved. Measured proportions (5 – 25 wt. %) of coconut shell particles shown in [Table 2](#) were added to the molten matrix and stirred thoroughly for 10 mins using a long stainless steel tong to avoid clustering and to achieve faster distribution of the particles in the matrix. The composite slurry was steadily poured into the wooden mould to which an aluminium foil had been placed to avoid sticking. The composite specimens were allowed to reach a semi-solid stage by cooling after which they were removed from the moulds and pressed at 0.33 MPa for 5 mins. This was recorded as 1<sup>st</sup> batch. This production method is similar to the one adopted by [Agunsoye et al., \(2013\)](#). The same procedure was used for specimens reinforced with cow bone particles and recorded as 2<sup>nd</sup> batch. The 3<sup>rd</sup> batch were the hybrid specimens which were polyethylene reinforced with equal mixture of particles of coconut shell and cow bone. The reference or control samples were also produced without any reinforcement. A picture of some of the specimens for tensile strength test is presented in [Fig. 2](#).



**Fig. 1. Photographs of the materials (a) polyethylene (waste nylons) (b) pelletized polyethylene (c) dried cow bones (d) 50  $\mu$ m cow bone particulates (e) coconut shells (f) 50  $\mu$ m coconut shell particulates.**

**Table 2. Quantity of materials.**

Matrix, (wt. %)	Reinforcement, (wt. %)			
Polyethylene	50 $\mu$ m CSP	50 $\mu$ m CBP	50 $\mu$ m hybrid (CSP + CBP)	Total (wt. %)
<b>100 (control)</b>	-	-	-	100
<b>1<sup>st</sup> Batch</b>				
<b>95</b>	5	-	-	100
<b>90</b>	10	-	-	100
<b>85</b>	15	-	-	100
<b>80</b>	20	-	-	100
<b>75</b>	25	-	-	100
<b>2<sup>nd</sup> Batch</b>				
<b>95</b>	-	5	-	100
<b>90</b>	-	10	-	100
<b>85</b>	-	15	-	100
<b>80</b>	-	20	-	100
<b>75</b>	-	25	-	100
<b>3<sup>rd</sup> Batch</b>				
<b>95</b>	-	-	2.5 CSP + 2.5 CBP	100
<b>90</b>	-	-	5 CSP + 5 CBP	100
<b>85</b>	-	-	7.5 CSP + 7.5 CBP	100
<b>80</b>	-	-	10 CSP + 10 CBP	100
<b>75</b>	-	-	12.5 CSP + 12.5 CBP	100

CSP = Coconut Shell Particles

CBP = Cow Bone Particles



**Fig. 2. Picture of some of the composite specimens for tensile strength test.**

### **2.3. Properties Characterisation**

The specimens were etched using Keller's reagent (95 ml water, 2.5 ml HNO<sub>3</sub>, 1.5 ml HCl, 1.0 ml HF) by swabbing manually for 15 secs at room temperature in accordance with ASTM E 407 – 99. Thereafter, a scanning electron microscope (SEM) JOEL JSM – 6480LV was used to examine their microstructure. Initially weighed ( $W_1$ ) dried specimens were placed in a beaker with water and reweighed ( $W_2$ ) at an interval of 24 hours for six days (144 hours). The water absorption ( $W_A$ ) of the composite was determined in accordance with ISO 175:1999 (E) standard using Equation 1 which was also earlier adopted by [Islam et al., \(2013\)](#) and [Mat-Shayuti et al., \(2013\)](#).

$$W_A(\%) = \frac{W_2 - W_1}{W_1} \times 100 \quad 1$$

The tensile test specimens were prepared using QualiLathe-210–CNC lathe machine and an Instron universal testing machine was used in accordance with ASTM D412. The hardness of the specimens of dimension 25 mm x 25 mm x 10 mm was determined in accordance with ASTM D 785 standard using a Brinell hardness machine with ball indenter of diameter 20 mm and maximum load of 4000 N. The impact energy of the specimens of dimension 75 mm x 10 mm x 10 mm with a 2 mm deep V-notch at their centers was determined using an Izod impact tester in accordance with ASTM D256 standard.

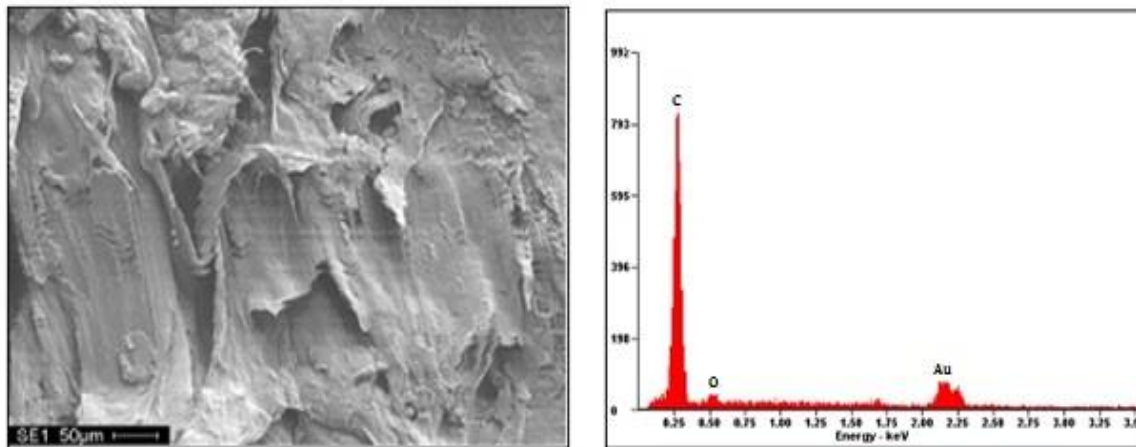
## **3. RESULTS AND DISCUSSION**

### **3.1. Microstructure**

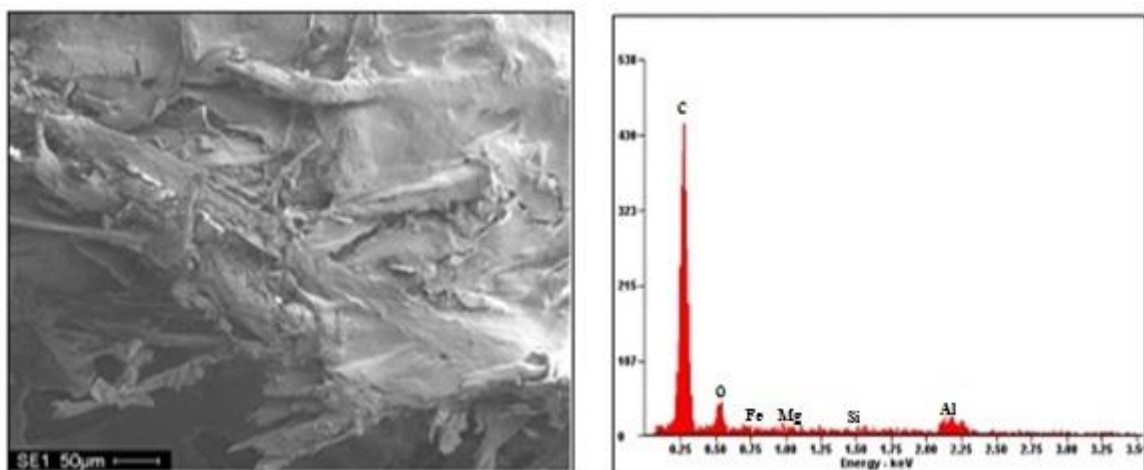
Polyethylene is a branched thermoplastic with many relatively long branches of the molecular chain ([Khanam and Al-Maadeed, 2015](#)). The scanning electron (SEM) micrograph shown in



Fig. 3 reveals uninterrupted interlocking of the polymeric chains. It also shows the partial homogeneity in the microstructure of the unreinforced polyethylene with dendritic (tree like) and oval shapes. This is responsible for the low strength exhibited by the polymer. From Figs. 4 and 5, the micrographs show the presence of reinforcement particles of coconut and cow bone in gray and white patches which are fairly distributed within the polyethylene matrix. In Fig. 6, the SEM micrograph shows a uniform distribution of the reinforcement particles in the microstructure of the hybrid composite. The energy dispersive X-ray spectrographs (EDS) of the reinforced specimens indicate a chemical reaction between constituting elements in the polyethylene matrix and those in coconut shell and cow bone particles leading to the presence of C, O, Fe, Au, Na, Si, Ca, P, and Al in the EDS.

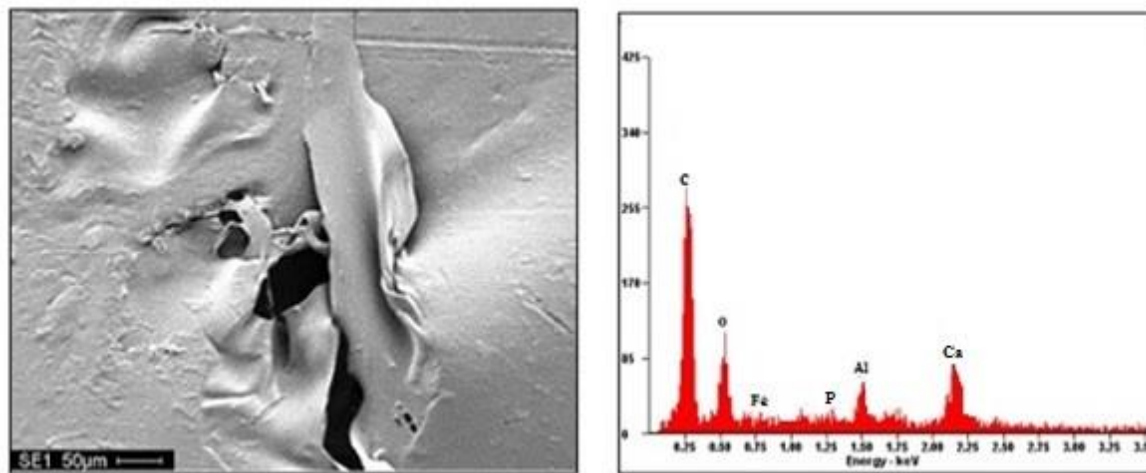


**Fig. 3. The SEM and EDS microstructure of the unreinforced polyethylene.**

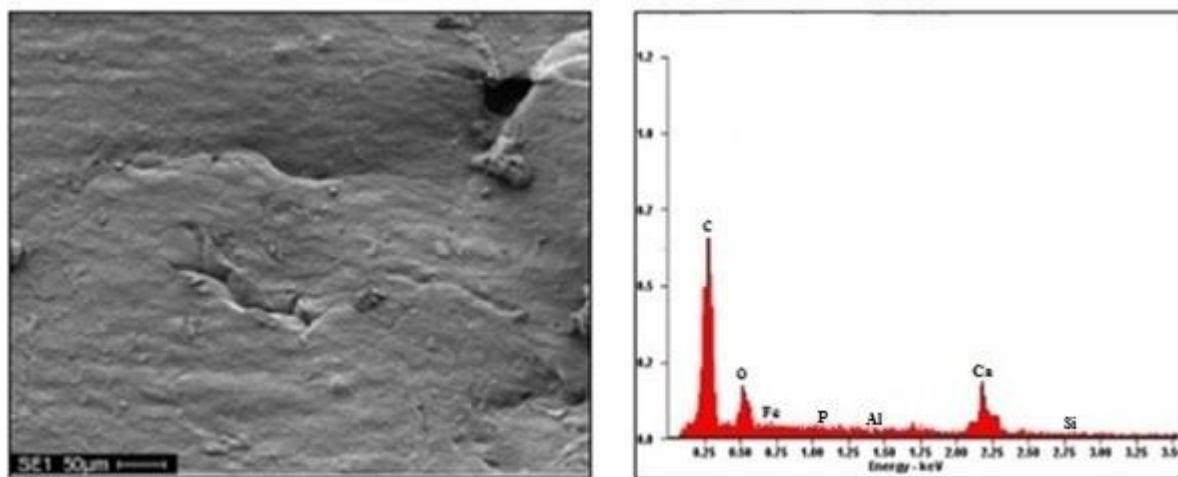


**Fig. 4. The SEM and EDS microstructure of the 15 wt. % coconut shell particles reinforced polyethylene composite.**





**Fig. 5.** The SEM and EDS microstructure of the 15 wt. % cow bone particles reinforced polyethylene composite.



**Fig. 6.** The SEM and EDS microstructure of the 15 wt. % hybrid composite.

### 3.2. Water Absorption

Water/moisture absorption could be detrimental to the mechanical, physical, chemical and dimensional properties of polymers (H'ng et al., 2011; Mat-Shayuti et al., 2013). Some polymers swell and soften in water such as nylon and polyvinyl alcohol (Mat-Shayuti et al., 2013). In the case of swelling and softening, molecular mobility is increased through the absorption of water. By the crowding of solvent molecules, polymer structure will open up and swell leading to increase in spacing between polymer molecules. This will lower the bonding and result in less resistance to applied stress from the decrease in intermolecular friction, allowing for easier translational motion (Mat-Shayuti et al., 2013).

As presented in Fig. 7, the value of water absorbed by the composites over a period of 144 hours at an interval of 24 hours increased. This is an indication of the presence of pores/voids

in the microstructure. Water absorption of the specimens can be explained by the presence of voids that have been occupied by water (Pantyukhov et al., 2016). This is also similar to the earlier report of Tewari et al., (2012).

Water is an essential component to support vital activity of microorganisms and one of their metabolites. The penetration of water through the surface layers and diffusing deep into the specimens can cause both plasticization and wedging effects (Pantyukhov et al., 2016). Washing of water-soluble substances out of the fillers on the one hand can also lead to the formation of ruts and voids that promote degradation (Pantyukhov et al., 2016).

The maximum water absorption level (0.3 %) exhibited by the hybrid composite is lower than that of the unreinforced and mono-reinforced specimens. This was due to the fairly strong interfacial bonding of the reinforcing particles with the polyethylene matrix indicating reduced pores in the microstructure.

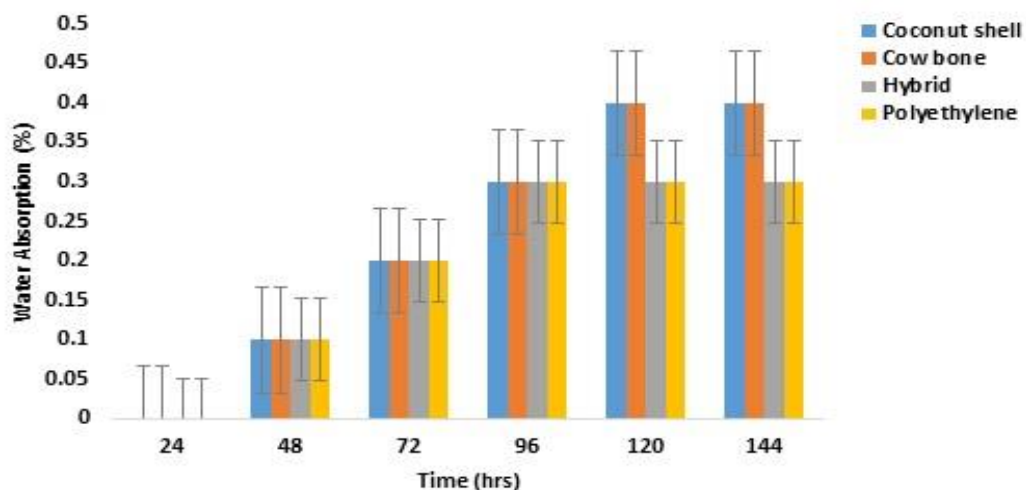


Fig. 7. Graph of water absorption against time of the composites.

### 3.3. Tensile Strength

The tensile property provides information about the behaviour of the specimens when they were subjected to stretching or pulling force before failure. The ultimate tensile strengths of the reinforced composite specimens are higher than the unreinforced polyethylene specimen as shown in Fig. 8. At 15 wt. %, the hybrid specimen demonstrated the highest ultimate tensile strength value of 1.78 MPa. This shows the ability of the blend of particles of coconut shell and the cow bone in enhancing the strength of the composite.

The uniform dispersion of the hybrid reinforcing particles as observed in the microstructure and strong adhesion of the particulates and polyethylene matrix must have contributed to the enhancement of the tensile strength of the composite which is similar to the earlier

report/postulation of Hassan et al., (2012) and Bello et al., (2015). This is predicated on the fact that composites properties depend critically on the level and nature of the adhesion/bonding between the reinforcement and the bulk polymer matrix because it is through this medium that stresses are transmitted to the reinforcements (Renner et al., 2005).

Increasing the filler concentration beyond 15 wt. % caused a decrease in the average inter-particle distance/spacing thereby increasing the amount of inter-particle stress concentration overlap. This led to higher levels of debonding when tensile stress was applied. This ultimately impaired the tensile strength of the composites which is similar to the earlier postulation of Rutz, (2014).

The decrease in the tensile strength of the composites at filler concentration beyond 15 wt. % was due to the non-uniform dispersion of particulates in the matrix. This generated a distribution of local stress which created a wider distribution of stress that caused debonding as a result of stress concentrations in the matrix thereby reducing/impairing the tensile strength of the composites. This is similar to the earlier postulation of Rutz, (2014).

The reduction in tensile strength beyond 15 wt. % filler addition was also due to improper filler distribution in the matrix. This led to weak adhesion of the particles and polyethylene which adversely affected load distribution. This is also similar to the earlier report/postulation of Agunsoye et al., (2013) and Durowaye et al., (2018).

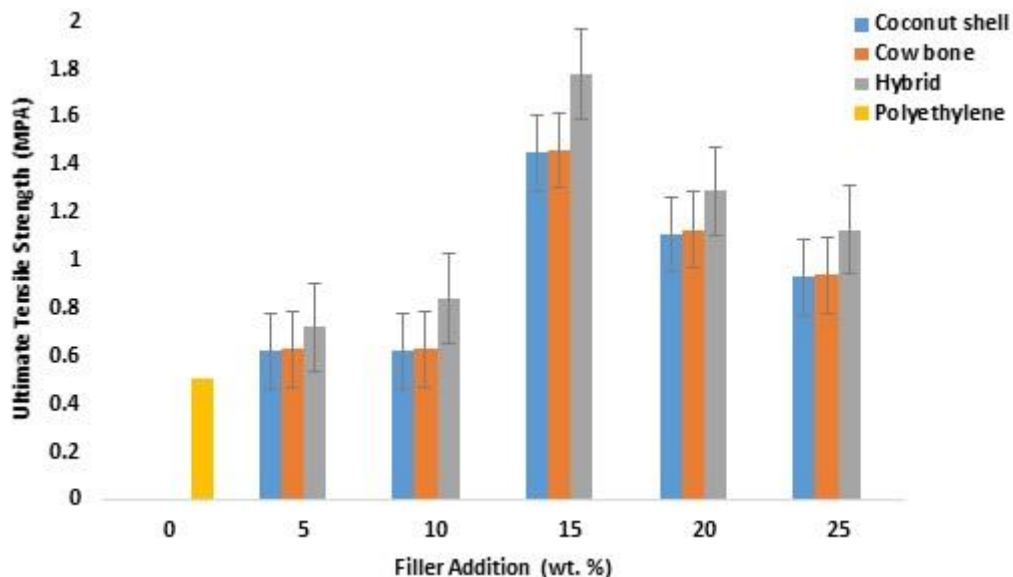


Fig. 8. Effect of filler addition on the tensile strength of the composites.

### 3.4. Modulus of Elasticity

As shown in Fig. 9, the unreinforced polyethylene matrix exhibited the highest Young modulus of elasticity of 57 MPa while the reinforced composites exhibited lower values. The modulus

of elasticity of the composites decreased with increasing filler addition. When tensile stress was applied, strain energy was stored in the specimens. The strain energy stored in the matrix which could be equal to the adhesion/bonding of the particles and polyethylene matrix caused the particle-matrix interface to debond and reduced or impaired the modulus of elasticity of the composites which is similar to the earlier postulation of [Rutz, \(2014\)](#).

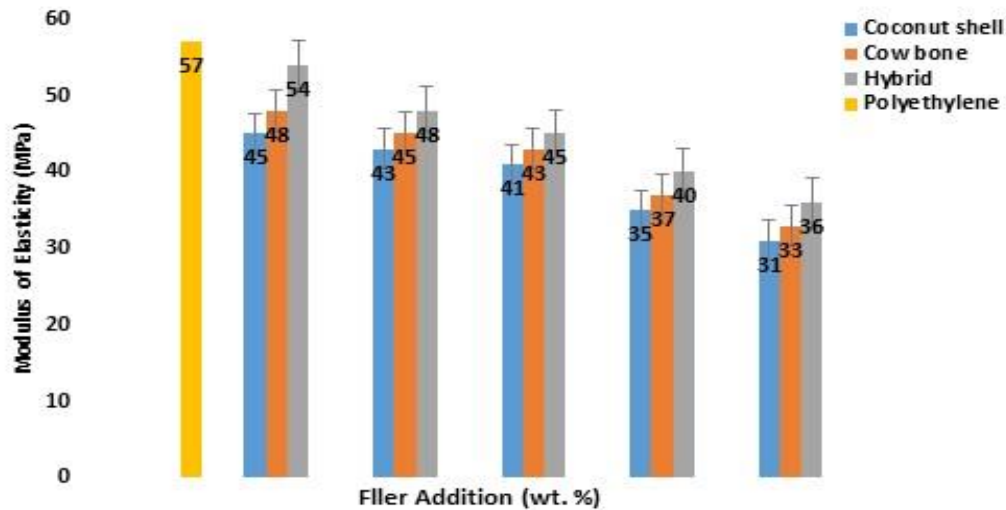
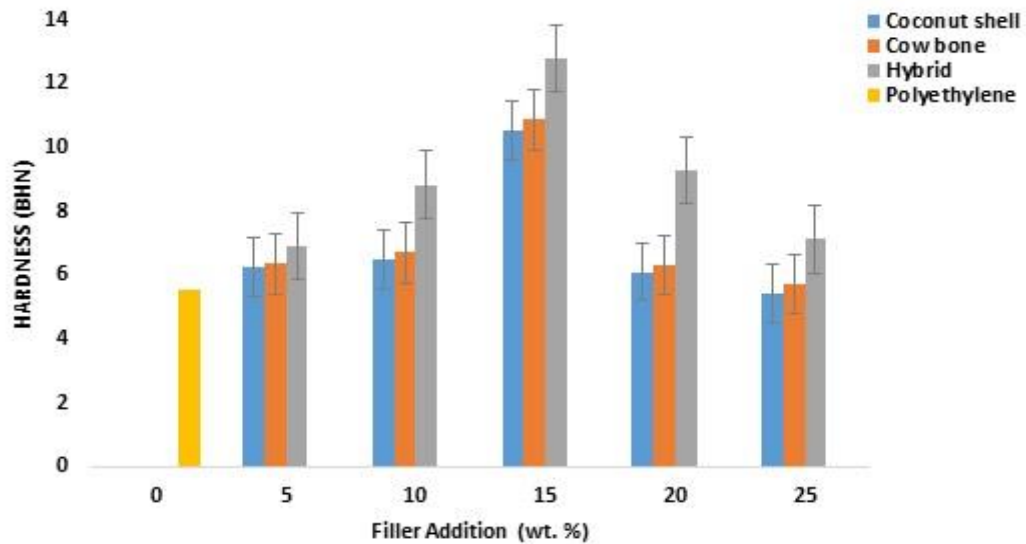


Fig. 9. Effect of filler addition on the modulus of elasticity of the composites.

### 3.5. Hardness

Generally, polymers exhibit low hardness ([Al-Mosaw et al., 2012](#)) and hardness implies resistance to indentation, permanent or plastic deformation of materials. The hardness value of the unreinforced specimen is 5.57 BHN as shown in [Fig. 10](#). The reinforced specimens exhibited higher hardness than the unreinforced polyethylene. The hybrid composite exhibited the highest hardness value of 12.78 BHN at 15 wt. % reinforcement. This indicates the ability of the blend of particles of coconut shell and cow bone in enhancing the hardness of the specimen. The uniform dispersion of the hybrid reinforcing particles as observed in the microstructure and strong adhesion of particles and polyethylene must have also contributed to the enhancement of the hardness. The reduction observed in hardness above 15 wt.% reinforcement may be due to improper dispersion of the filler in the matrix resulting to weak adhesion of particles and polyethylene matrix which is similar to the earlier report/postulation of [Agunsoye et al., \(2013\)](#).



**Fig. 10. Effect of filler addition on the hardness of the composites.**

### 3.6. Impact Energy

The unreinforced polyethylene specimen exhibited an impact energy (IE) of 6 J. The IE of the composites decreased with increasing filler addition as presented in Fig. 11. The decrease in IE may be attributable to the hardness of reinforcing particles which imparted brittleness to the polyethylene matrix.

During impact loading, the formation and propagation of cracks and micro-voids (Manikandan and Rajkumar, 2016) within the composite led to a reduction in the impact energy. Increasing filler concentration also led to an increase in the surface area available for filler-matrix interaction. This ultimately led to an increase in mobility of the matrix molecules. As a result of increase in the mobility of matrix molecules the particle sizes tended to accumulate and increase which resulted into weakening of the interfacial bonding between the matrix and the filler. It is therefore expected that as the filler content and particle size increased there will be a decrease in impact energy which is similar to the earlier report/postulation of Agunsoye et al., (2013).

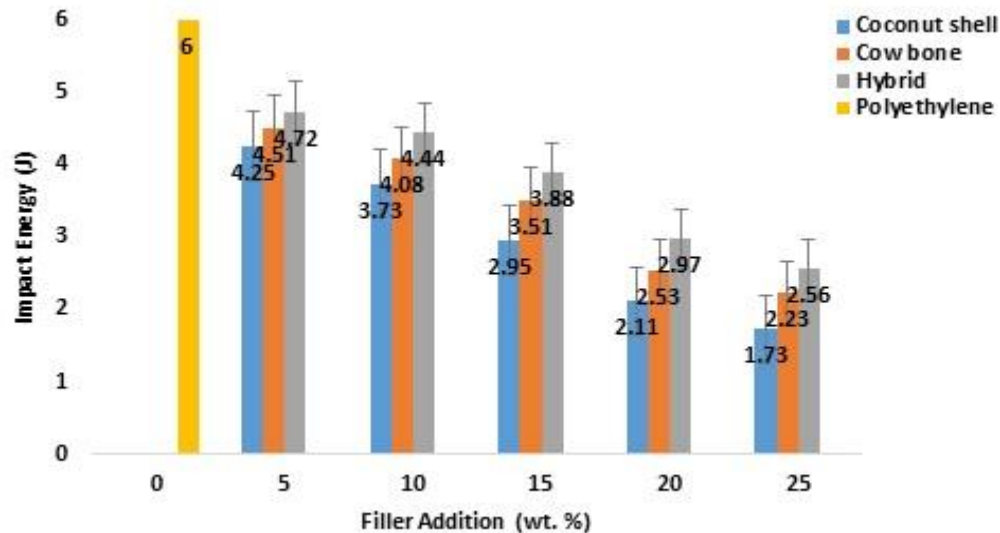


Fig. 11. Effect of filler addition on the impact energy of the composites.

#### 4. CONCLUSIONS

The development and characterisation of coconut shell and cow bone particulate polyethylene matrix biocomposites have been successfully carried out and the following conclusions can be drawn.

1. The hybrid composite exhibited desirable properties in terms of water absorption (0.3 %) indicating reduced pores/voids. It also exhibited ultimate tensile strength (1.78 MPa) and hardness (12.78 HBN) at 15 wt. % filler addition.
2. The uniform dispersion of the hybrid reinforcing particles as observed in the SEM microstructure and strong adhesion of the particles and polyethylene matrix contributed to the enhancement of the tensile strength and hardness of the composites.
3. The reduction in tensile strength and hardness beyond 15 wt. % filler addition could be due to weak adhesion/bonding which adversely affected load distribution.
4. Increasing the filler concentration beyond 15 wt. % caused a decrease in the average inter-particle distance/spacing thereby increasing the amount of interparticle stress concentration overlap. This led to higher levels of debonding when tensile stress was applied. This ultimately impaired the tensile strength of the composites.
5. The strain energy stored in the matrix which could be equal to the adhesion/bonding of the particles and polyethylene matrix caused the particle-matrix interface to debond and reduced or impaired the modulus of elasticity of the composites.

6. Morphological analysis using SEM and EDS clearly showed the difference in morphology of the developed polymer matrix biocomposites.
7. The development of this biocomposites will reduce environmental pollution and also has the potential for application in areas where low strength is required.

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