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Effect of Nano-Structured Bambara Nut Shell (Vigna Subterranea (L.) Verdc) As Filler on the Physical, Mechanical and Morphological Properties of Epoxy Matrix

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Abstract
In this study, the physical, mechanical and morphological properties of epoxy/bambara nut shell (BNS) nano composite were studied. Different samples were produced with (5 wt%, 10 wt%, 15 wt%, 20 wt%, 25 wt%, 30 wt%, 35 wt%) of the nano-structured filler while 100% epoxy resin without any filler was used as the control sample A. The fabrication of the composite was done by casting on a glass mould and cold curing for 24 hours at 25^{0}C. The fabricated composites were tested based on ASTM standards for tensile strength, hardness, impact strength, density, flexural strength and morphological properties. Results showed that Sample D with 15% filler loading gave superior properties in most cases. Tensile strength result shows that sample with 15% of filler had high tensile strength of 80.20MPa when compared to 37.87MPa of unfilled (control) sample. Similarly, Impact strength result shows that the sample with 15wt% of filler had highest Impact strength of 0.45J/mm while 35% of filler loading had least Impact strength of 0.15J/mm. Generally, the inclusion of nano-structured bambara nut shell not more than 15 wt% into epoxy matrix improved most of the composite properties.

1. Introduction
Volumes of agricultural wastes are generated annually during harvesting season of crops, in agro-processing industries and also from live stocks farming. These wastes could be solid, liquid or slurries and their composition are largely dependent on their various sources [1,2]. In most developed countries, significant volumes of these wastes are utilized as raw materials by other industries thereby reducing their negative impact on man and environment [3,4]. Contrary, in underdeveloped and most developing countries the use of these wastes for value added products is still very low. While agricultural activities are on the increase in most developing countries to meet the target self sufficiency in food production,

more wastes arising from these intensive farming has not received adequate attention particularly in Africa. Consequently, the risk due to hazardous wastes and poor hygiene is inevitable.

Estimates of wastes arising from agricultural activities are rare, but they are generally thought of as contributing a significant proportion of the total waste matter in most cities around the world [5]. However, researches on utilization of solid agro wastes are gradually receiving attention from the scientific community with little or no attention from the industries in developing nations. The use of these wastes as fillers in polymer composites [4], nano fillers in reinforcing plastics and rubber [3, 7, 8], lignin-containing cellulose nanomaterials [9, 10] biodegradable polymer films [11] and many other functional materials have been reported. The excellent mechanical properties and biodegradability of most crop residue/waste have positioned them as choice reinforcing materials in polymer composites [11, 12] as well as in tissue engineering [13].

Bambara nut shells are major agro waste in Africa. They are by-products obtained during the processing of Bambara seeds into flour for human consumption. The shell is produced after splitting the seeds in an attrition mill to remove the shells, winnowing to remove loosened testa and converting the cotyledons into fine flour by milling several times followed by sieving. In Nigeria and indeed most African countries, large amounts of this nut are cultivated annually (Table 1). The shells are used in the feeding of some livestock [14] thereby limiting its use while large volume of it is discarded and they cause hazards to the environment [15]. Utilizing this agro waste as reinforcement for plastics (thermoplastics and thermoset) is a valorization route that will solve the twin problem of environmental and economic sustainability.

Table 1: Estimated production of Bambara nut in some African countries

S/No	Country	Estimated production (metric tonnes)
1.	Nigeria	100,000
2.	Bostwana	6,000
3.	Togo	4,400
4.	Burkina Faso	44,712
5.	Cameroun	24,000
6.	DR Congo	10,000
7.	Mali	23,165
8.	Zambia	18,750
9.	Zimbabbwe	750
10.	Ghana	20,000
11.	Niger	30.000
12.	Cote d'ivoire	7,000
13.	Chad	20,000

(Source: Aviara *et al*., [16])

Epoxy resins are one of such thermosets that possesses excellent design flexibility, good mechanical, thermal, chemical properties and also as a thermoset it does not require chemical compatibiliser to synergies with reinforcing materials. However, excellent properties could be achieved by reinforcing epoxy with most agro fillers, thus making it choice material for most high-performance engineering applications. These excellent properties have shown to be profound with nano fillers because the important structural characteristics which give unique properties to nanocomposites are the nanosize and a huge interfacial surface area of the nanofillers. Even though dispersion of nano fillers in thermoset poses a challenge during polymer/nano composite fabrication, it is less so with crop nano fillers because of their relative low bulk density which is an edge for most agro waste polymer composites. In recent years researchers have focused on the study of epoxy nano composites filled with mineral fillers such as clay [17, 18, 19], glass [20, 21, 22], carbon nano tubes [23], grapheme [24, 25] and many other functional materials, with less emphasis on agro wastes in their nano structured form.

This study is therefore aimed at reducing the volume of bambara nut shell wastes by utilizing it as a potential light weight reinforcing nano-structured filler to possibly improve the properties of epoxy for most engineered application especially were light weight and excellent strength and cost effectiveness are priority.

2. Materials and method

Local untreated bambara nut were obtained from Dawano market in Kano State, Nigeria (Figure 1). The epoxy resin is composed of; a diglycidyl ether of bisphenol A epoxide (DGEBA) and the mixture of cycloaliphatic amines both a product of PubChem, were purchased from local dealer and used as received.

2.1. Preparation of Nano bambara nut shell

Bambara nut shell was gotten from Dawano market in Kano State, washed and sun dried to remove moisture/water to between 8-12% to ease processing. It was sieved to remove impurities such as sand, stone or any foreign material that may disturb the process. It was grinded to reduce its size, and then followed by high energy ball milling at an estimated speed of 300 rpm at ball to particle ratio (BTP) of 1:20 on eight bases with an average porcelain ball diameter of 2cm (Figure 2).

Figure 1: Bambara nuts

Figure 2: Bambara nut shell Nano Particles

The particles were subjected to particle size distribution measurement with the aid of Malvern master-sizer 3000 (Malvern instrument, UK). The machine uses the principle of light scattering to measure the diffusion of particles under Brownian motion using methanol as the dispersant at 60 seconds under a temperature of 25^0C.

2.2. Fabrication of the Composite samples

Bambara nut shell particles of varying weight percent of (0 wt%, 5 wt%, 10 wt%, 15 wt%, 20 wt%, 25 wt%, 30 wt%, 35 wt%)) were mixed with epoxy and hardener at ratio (2:1) respectively then poured into a glass mould of 180x180x3mm dimensions. The mixing was done according to the formulation presented below. An aluminum foil was neatly placed on the mould to avoid the composite from sticking on the surface of the mould. The composites are kept at room temperature and allowed to cure for 24 hours.

Table 2: Formulation for the composite preparation

Sample ID	Filler (wt%)	Epoxy and hardener (wt%)
A	0	100
B	5	95
C	10	90
D	15	85
E	20	80
F	25	85
G	30	70
H	40	60

3. Physico-mechanical and morphological properties of bambara/epoxy nano composite

3.1 Physical Properties

3.1.1. Density

The density of the composites was obtained from the mass and volume. The mass was obtained using an electric weighing balance and the volume was obtained by direct measurement using a vernier caliper. Samples of dimensions 50 mm × 40 mm × 3 mm; length x width x thickness respectively were used for the density measurement.

$$Density\ (\rho) = M/V$$ Eqn. 1

Where, m is the mass of the composites (g), v is the volume of composites (m^3). Three samples of each different types of composites were used and the average value was reported.

Mass determination was carried out by weighing the composites to four decimal places on an analytical balance (Mettler 5000). Volume was determined by using a digital veneer calliper (Mitutoyo).

3.1.2. Hardness

Hardness of each composite sample was determined according to ASTM- D2240 [26] using Durometer hardness tester model no. 5019 on Shore A scale.

3.1.3. Water absorption

Water absorption was carried out in water at 25 °C according to ASTM D570 [27] using samples of dimensions 50 × 40 × 3 mm (figure 3). The water absorption was calculated as follows:

$$\% \textbf{ Water absorption} = (Wf - Wo/W)\, x\, 100$$ Eqn. 2

Where *Wf* is the final weight after immersion in water and *Wo* is the initial weight before immersion in water for 24 hours.

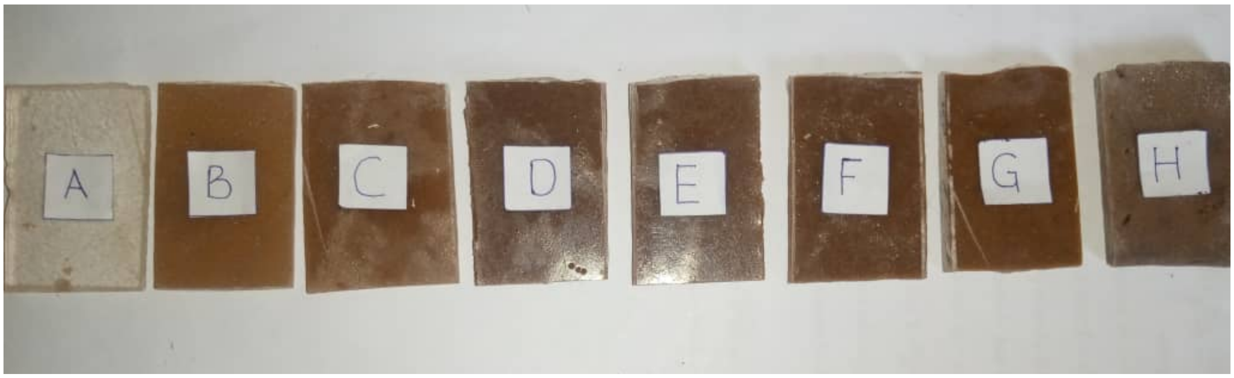

Figure 3: *Hardness and Density Test samples*

3.2 Mechanical Properties

3.2.1. Tensile Strength

Tensile Strength was measured according to ASTM D3029M [28] using Hounsfield Monsato Tensometer at a cross speed of 10 mm/min. Dumbbell shaped samples with the following dimensions; overall length of 100mm, thickness of 3 mm, width of grip section of 15 mm, width of reduced section of 8 mm, distance between shoulders of 60 mm and gauge length of 40 mm were used Figure 4 and Figure 5.

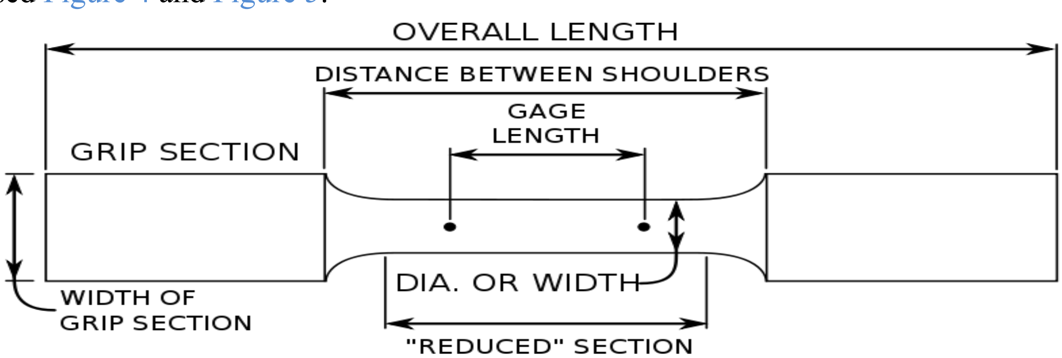

Figure 4: Dimensions of the tensile specimen

$$\textbf{Tensile Strength}\ (6s) = \frac{Fm}{A}$$ Eqn. 3

Where *F*m is the maximum force before the sample breaks and *A* the cross sectional area

3.2.2 Modulus of elasticity (MOE)

Modulus is a measurement of the material's stiffness. MOE test was carried out according to ASTM D638 [29].

$$\textbf{Tensile Modulus}\ (Et) = \frac{6s}{\varepsilon}$$ Eqn. 4

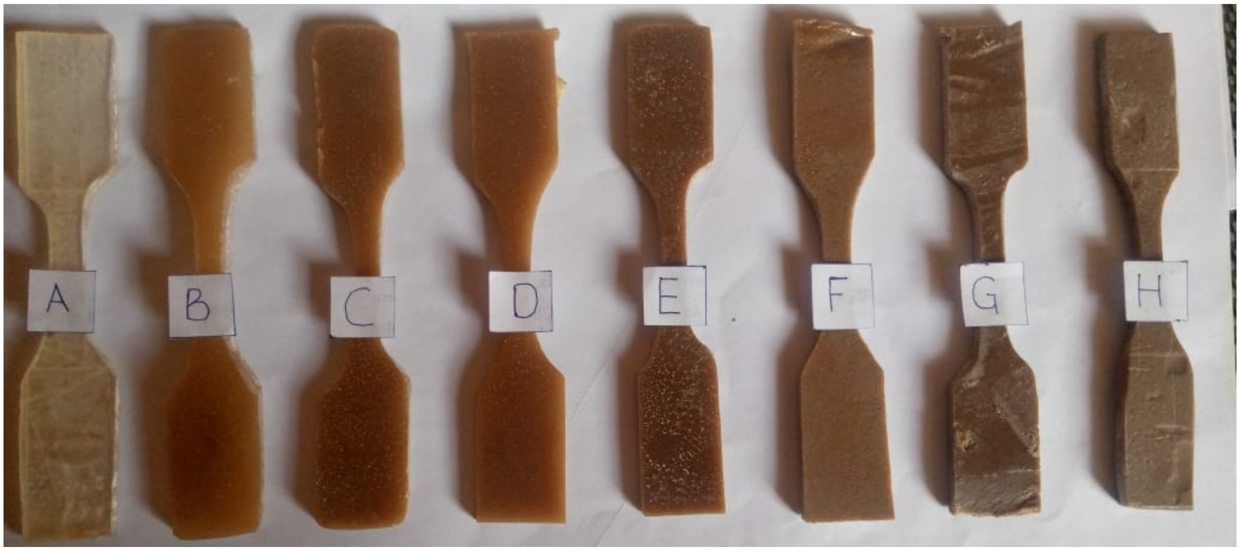

Figure 5: *Tensile strength Test samples*

3.2.3 Flexural Test strength

Flexural tests were performed in accordance with ASTM D7264/D7264M-07 [30] standards using a SHIMATSU AGS-X test machine having a load cell of 10 kN. Three point bending tests performed to examine bending behavior of epoxy/bambara nut nanocomposite samples which were carried out on samples (3 mm×15 mm×80 mm) (figure 3). The span length equal to 65 mm and the crosshead speed to 1 mm/min.

Figure 6: *Flexural Test samples*

3.2.4 Impact strength

The impact properties were studied on notched specimens to determine impact strength using a pendulum hammer impact test machine in accordance with ASTM D256 [31]. Izod impact testing machine (type 6957) with a pendulum arm of 4 J capacities was used to obtain the impact strength of each composite sample. The samples used for the impact test had the dimensions; thickness of 3mm, width of 10mm, length of 55 mm and v-notched at mid length (Figure 4).

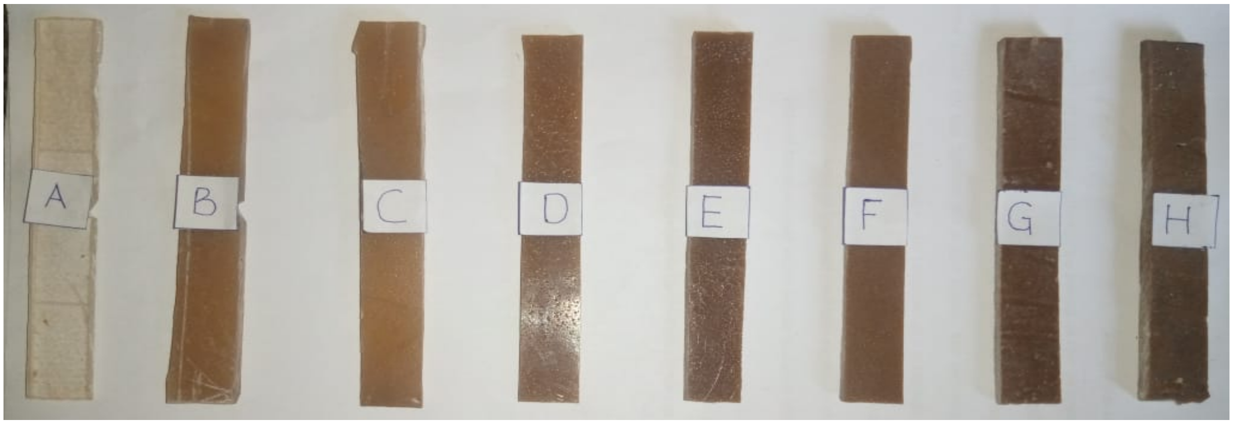

Figure 7: *Impact strength Test samples*

3.3 Morphology

SEM micrographs of fractured impact test specimens of neat epoxy and also various nanocomposites were studied using Scanning Electron Microscope; Phenom world model ProX with acceleration voltage of 15 KV.

All physical and mechanical tests were carried out three times and the average was taken to minimize human and instrument error.

4. Results and discussion

4.1. Particle size

The particle size distribution of the Bambara nut shell nanoparticles sample by volume is shown in Figure 8 and Table 3; the results showed that the Bambara nut shell nanoparticles have three peaks with the first peak having 89.4% of particles size of 57.82 (d.nm), the second peak having 9.8% of the particle between the size of 1-10.38 (d.nm) while the last peak having 0.5% of the particle between the size of 1-4950 (d.nm).

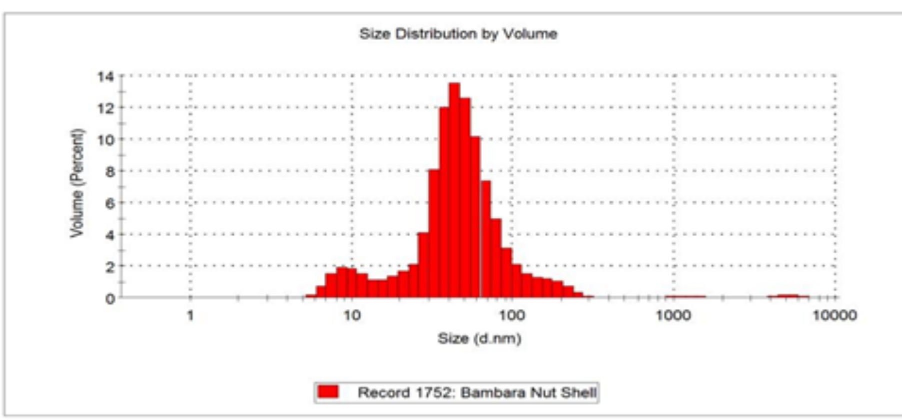

Figure 8: Particle size distribution of bambara nut fibre powder by volume

The volumetric quantity of Bambara nut shell particles between 1nm and 100nm is 89.4% indicating that the ball milled fiber falls within the acceptable nano standard of 100nm according to Uzochukwu *et al.,* [3].

Table 3: Particle size distribution for the bambara nut fibre powder

		Size (d.n…)	% Volume	St Dev (d.n..)
Z-Average (d.nm): 74.20 PdI: 0.197 Intercept: 0.944	Peak 1 Peak 2 Peak 3	57.82 10.38 4950	89.4 9.8 0.5	37.03 2.816 912.8

4.2. Physical properties

4.2.1 Density

The maximum density recorded for the composite sample is 0.82g/cm^3 representing sample B with 5 wt% of bambara shell nano filler. This value is significantly lower than the value for unfilled epoxy sample (1.18g/cm^3), suggesting that the bambara shell nano filler is light in weight but tough as will be seen in the subsequent properties investigated. Figure 9 shows gradual decrease in the density of the samples as filler loading increased. This is expected because the average density of the bambara nut shell is 0.87g/cm^3. Sample H with 35% bambara nano filler had the least density of 0.62 g/cm^3 indicating a higher fraction of the light weight component in the composite formulation. Furthermore, the presence of voids and agglomeration as filler loading increases will create free volumes which will also contribute to decrease in density. The presence of voids in polymer composites is inevitable and can only be minimized; they arise from sources such as air bubble entrapped within epoxy matrix, residual solvent, moisture in the filler and volatile arises during curing of the resin [32].

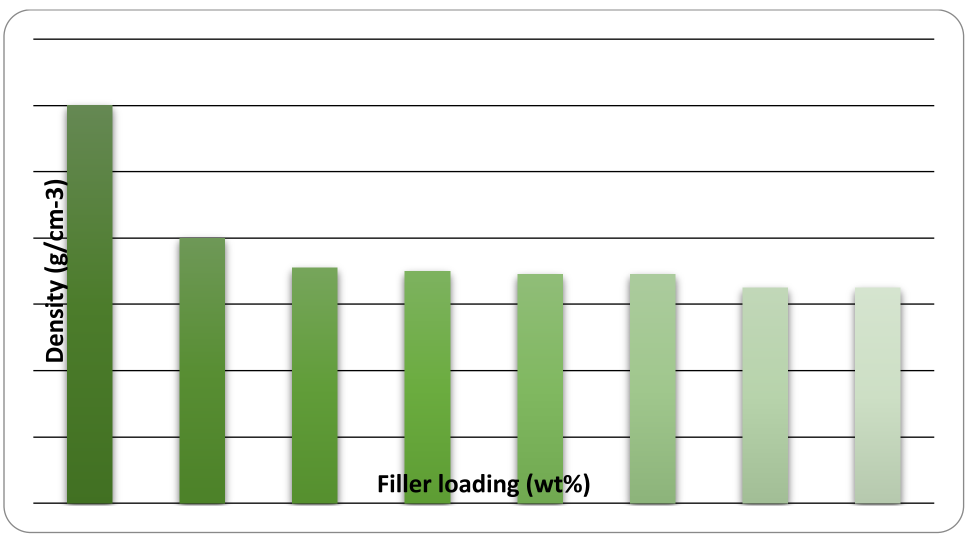

Figure 9: Effect of filler loading on *density of bambara nut shell/epoxy nano composite*

4.2.2 Hardness

Hardness is a measure of how hard the surface of a substance is. The harder the surface of a material is, the more abrasion-resistant it is. Generally, the shore A hardness of the composites increases as the filler loading. Figure 10 shows that 35 wt% filler loading (sample H) has the highest hardness value of 131. This could be attributed to the increasing volume of the bambara nano fillers which is harder than neat epoxy matrix. This is consistent with the report of Moorthy, *et al.,*[33]. Furthermore, the agglomerated nano particles at higher filler loading have resisted the indentation of the composites consequently leading to higher hardness of the composites surface. Similar trend was reported by Vinay [34], in his study on Mechanical behavior of walnut (Juglans L.) shell particles reinforced bio-composite. They observed that the hardness of the composite samples increased from 56.4 MHR for unfilled epoxy resin Y-230+8 wt% of HY-951 to 89.8MHR for 25% filler loading on ockwell M-scale.

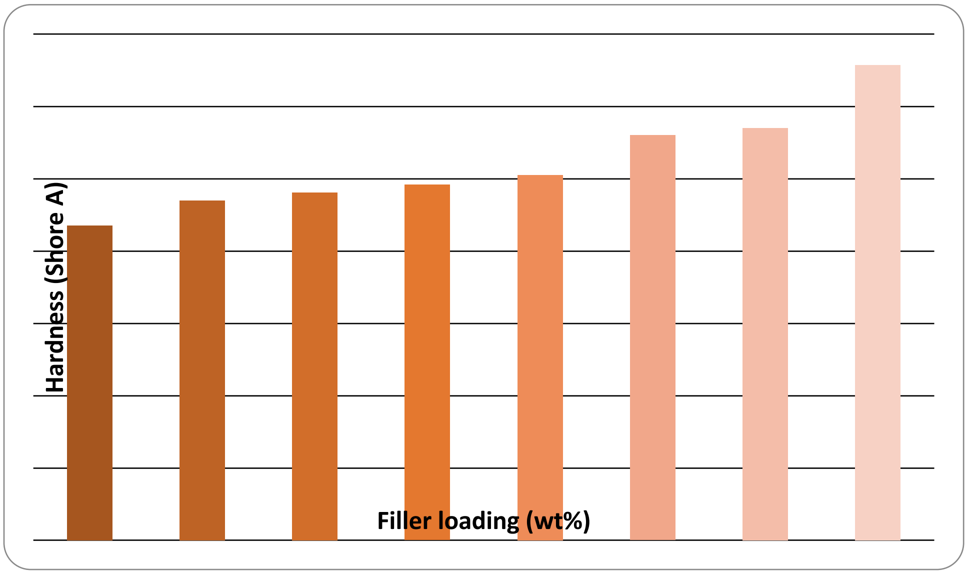

Figure 10: Effect of filler loading on *the Hardness of bambara nut shell/epoxy nano composite Hardness*

4.2.3 Water absorption

Water absorption behavior of nano-bambara nut shell filled epoxy composites are shown in Fig. 11. The water absorption property of polymer-filled composites is enfluenced by factors, such as processing techniques, type of matrix, type of filler, filler/matrix composition of the composites, and cotact time in water [35]. Water absorption was observed to continual increase with increase in filler loading from 5 wt% to 35 wt%. The water absorption for 5% filler loading was found to be 1.2 % with steady increase filler loading to 35 wt% the water absorbtion was recorded to be as high as 3.5 % after 24 hours immersion in water. The water absorbtion of the composite is attributed to the hydophilic property of the bambara shell filler while the significant increase in percentage water absorbtion as the volume of the filler increased from sample B to sample H is due to high presence of void on the composites which traps water within its free volume. In addition, the hydrophilic property of the bambara nut shell fillers promotes the water uptake by the formation of hydrogen bonds between filler and water molecules. Thus, increase in filler loading will increase the formation of hydrogen bond between filler and water molecules [36]. Several researchers have reported that the water absorption of natural fibre polymer composite increases with increase in filler loading; Yang *et al* [37] reported linear relationship between fibre loading and water absorption while Väisänen *et al* [38] and Uzochukwu *et al*.[3], also showed that water absorbtion of most natural fiber in polymer matrix could be up to 20%.

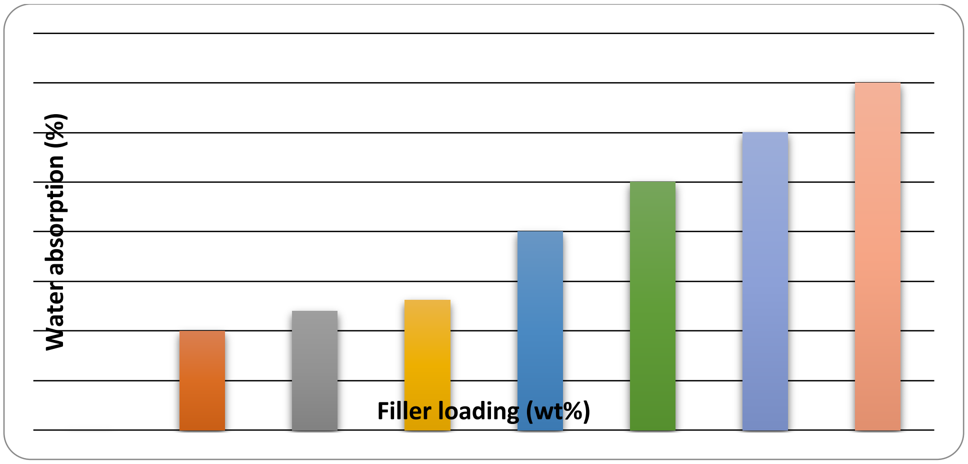

Figure 10: Effect of filler loading on water absorption of *bambara nut shell/epoxy nano composite*

4.3 Mechanical properties

4.3.1 Tensile strength

The results of the tensile property of the composites developed at various filler loadings are presented in figure 11. It was observed that there was significant increased in tensile strength between unfilled epoxy (sample A) and the filled composite samples. A gradual increase in tensile strength was recorded from 5wt% filler loading up to 35 wt%. The inclusion of the bambara shell nano-structured filler into the epoxy matrix significantly improved its tensile strength from about 39 MPa to 80 MPa representing 5wt% (sample B) and 15 wt% (sample D) respectively. This observed increase is due to reinforcing ability of the filler and good filler- matrix interaction. It could be deduced therefore that at 15 wt% filler loading there was a good transfer of load between the filler and the matrix.

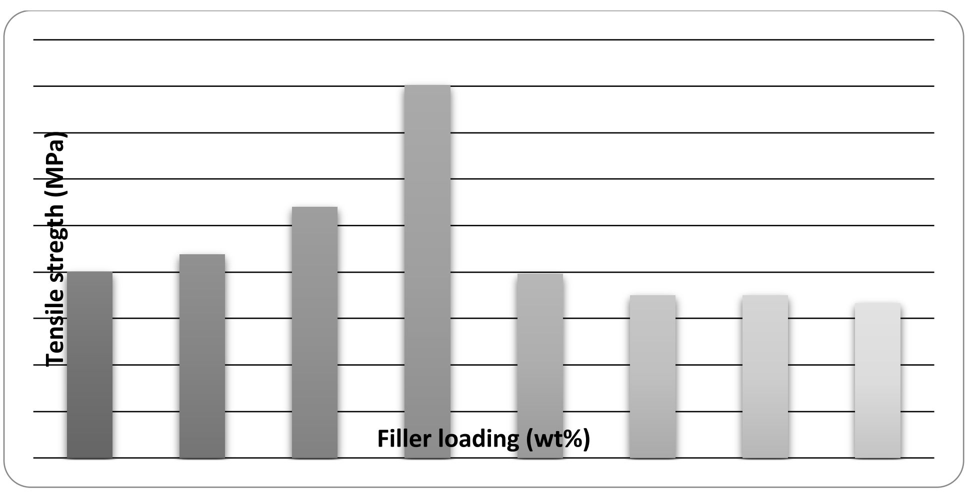

Figure 11: Effect of filler loading on *Tensile strength of bambara nut shell/epoxy nano composite*

It can also be seen clearly that at filler loading above 15 wt% up to 35 wt% the tensile strength dropped very significantly due to inability of the matrix to wet the filler at higher loading. Consequently, the matrix is not able to transfer load effectively to the filler. Furthermore, at higher filler loading fiber agglomeration becomes almost inevitable due to poor wetability, during loading the composite is bound to fail at the regions of high stress concentration created by agglomerated fillers. Several reports on epoxy nano fiber reinforced composites have reported similar observation [2, 3, 39]. It can be seen that beyond the optimum level of dispersion i.e 15 wt% the composites are unable to support high load. This is true not only for nano fillers but also for micro size fillers [6, 40] While it is true that nature of fillers and filler types affect the tensile strength to great extent, the trend is generally the same that tensile strength will decrease beyond a volume of filler that the matrix can no longer wet.

4.3.2 Modulus of Elasticity

The effect of filler loading on the Modulus of elasticity of the composites is presented in the figure 12, 15 wt% filler loading (sample D) exhibited the highest elastic modulus of 131.71MPa this is an indication of optimum filler loading. This further suggests that up to 15 wt% filler loading, the filler exerts maximum stiffening effect on the epoxy chain leading to high stiffness of the composite. The increase in filler loading from 15 wt% to 35 wt% caused gradual decrease in the modulus of elastic of composite samples which might be attributed to poor dispersion of filler in the epoxy matrix at higher loading. Similar trend was reported by Majeed *et al.* [36] and Uzochukwu *et al.* [3].

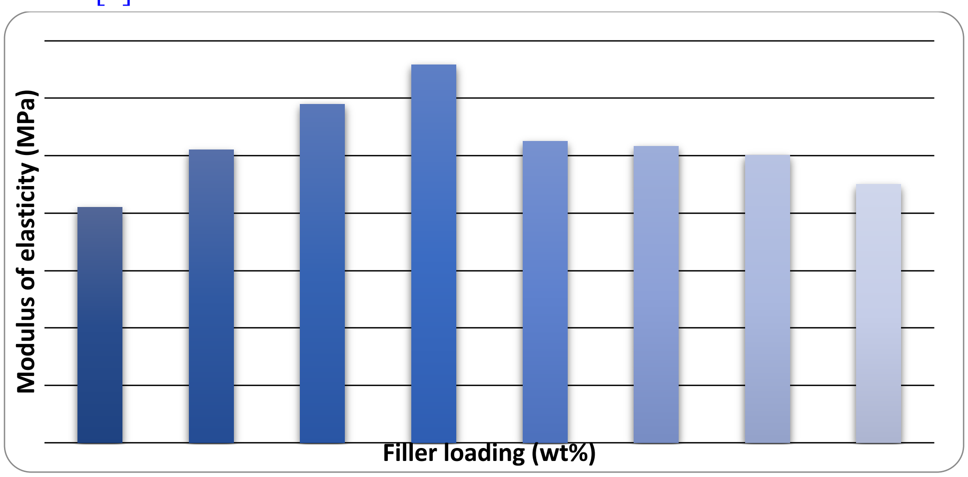

Figure 12: Effect of filler loading on *modulus of Elasticity of bambara nut shell/epoxy nano composite*

4.3.3 Flexural strength

Flexural strength, also known as bent strength is a material property that is defined as the strength of a material just before it yields in a flexure test. Figure 13 shows the effect of filler loading on the flexural strength of bambara shell nano filled epoxy composites. It is clear from the graph that the flexural strength initially increased up to 15 wt% of the nano-structured filler and then decreased with increasing of filler loading. The flexural strength reached the maximum value when the filler loading was about 15 wt% which is 53.90 MPa. The least flexural strength was observed with 35 wt% filler loading. Several research on micro size filler polymer composites supports that flexural strength decreases with increasing filler loading above optimum filler/matrix loading [41, 42]. However, the advantage of nano size filled composite is in the improved properties due to better interfacial adhesion and improve surface area between filler and matix.

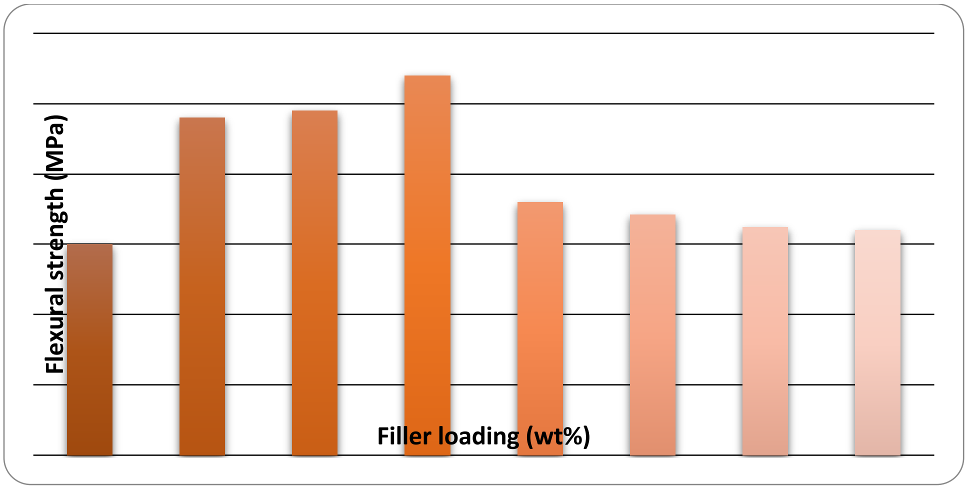

Figure 13: Effect of filler loading on *Flexural strength of bambara nut shell/epoxy nano composite*

4.3.4 Impact strength

From the Figure 14, sample B (D wt%) and sample H (60/ wt%) had the highest and least impact strength of 0.45J/mm and 0.13 J/mm for filled sample respectively. it is also clear that the inclusion of the nano filler up to 15 wt% caused a significant improvement on the impact energy of unfilled epoxy sample A.

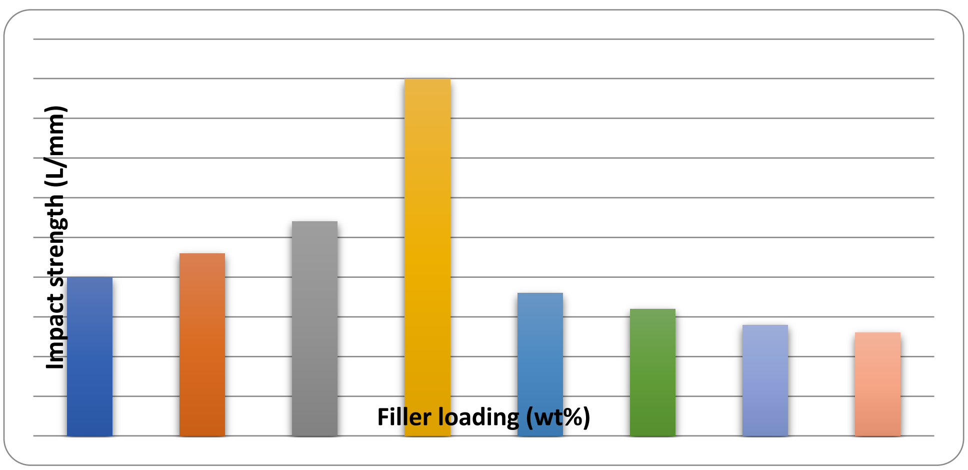

Figure 14: Effect of filler loading on *Impact Strength of bambara nut shell/epoxy nano composite*

However, above 15 wt% of the filler the matrix could no longer wet the filler which led to decline in impact strength of the samples. Furthermore, the possible presence of agglomerated fillers could create weak interfacial bonding between fillers and matrix consequently given room for weak regions for cracks propagation.

Scanning Electron Microscopy (SEM) :

The tensile fracture surfaces of nano filled composites were examined using SEM and typical micrographs at X 300 magnification are presented in Figures 15b–15d. Unfilled epoxy after impact testing at high X300 magnification is produced in figure 15a.

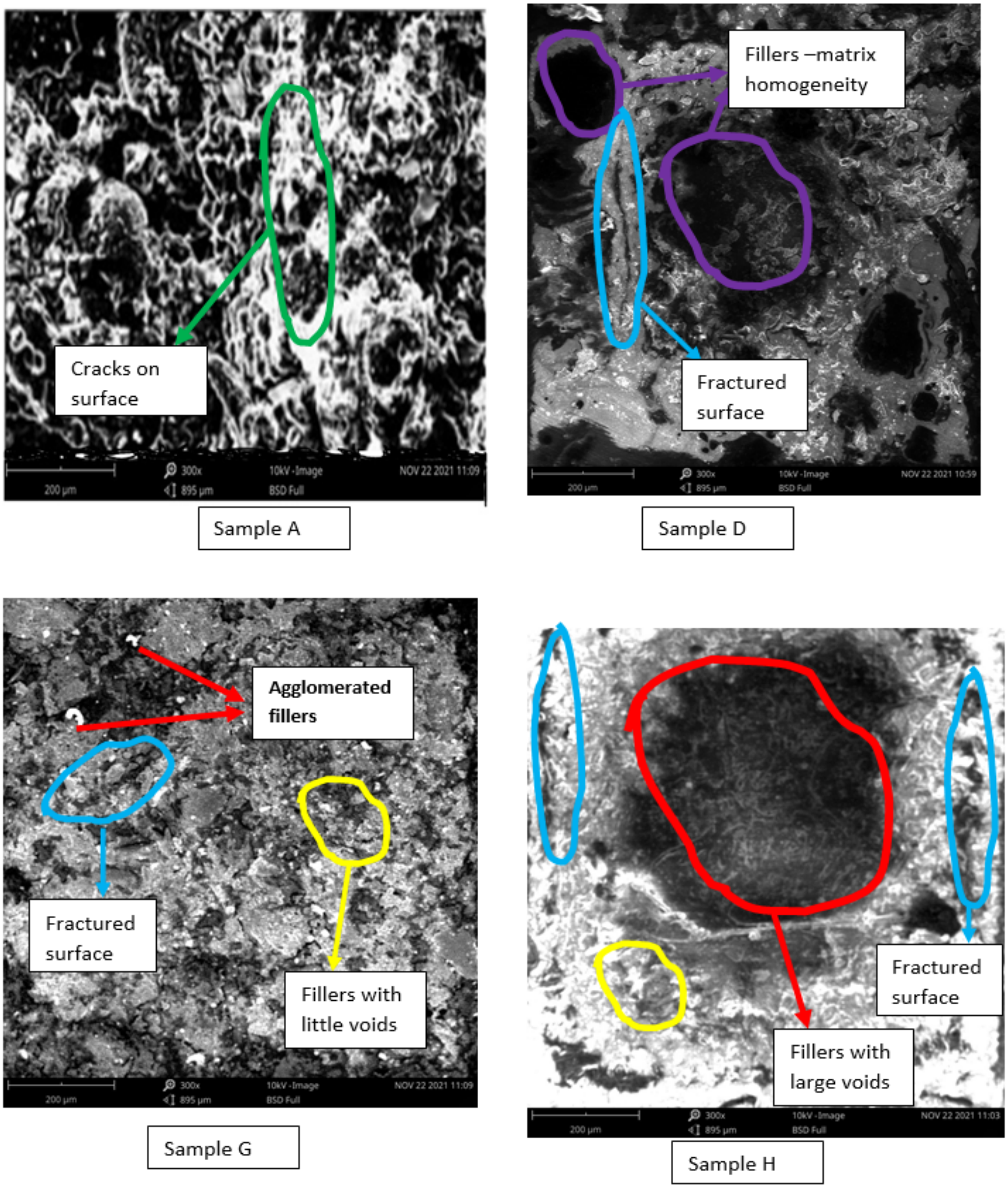

Figure 15a-d: *SEM micrographs of tensile fractured specimens for unfilled sample and different filler loading of bambara nut shell at 300X magnification: (a) 0 wt % (b) 15wt% (c) 30wt% (d) 40wt%*

From these figures, it was observed that the fracture surface of unfilled epoxy (sample A) 0 wt% filler loading had obvious crack propagations in running through the entire surface, which clearly indicates that the material could not withstand reasonable tensile stress. Hence, the resistance to crack propagation is less and leads to brittle failure. There were near absence voids because of homogeneity of the resin [43]. The investigation of the fractured surface of the epoxy composite reinforced with Bambara but shell nano-structured filler shows that Sample D (15 wt%) of filler loading had a better nano filler- matrix dispersion with less voids, less particle aggregation and better adhesion of the epoxy matrix which accounted for its improved properties and performance compared with sample G and sample H. Large voids were seen in sample H suggesting poor that there were areas of poor adhesion between fillers and matrix [36, 44]. Less filler agglomerations were also found in sample D (5 wt% filler) compared to sample G (30 wt% filler) and sample H (35 wt% filler) indicating good wetability at lower filler volume. Finally, the fracture surfaces of the bambara nut shell/ epoxy composites showed that there were no prominent gaps on the surfaces, indicating a good mix during processing [6].

Conclusion

The aim of this study is to utilize the bio-agricultural waste as filler material for composite production which are abundantly available and low cost compared to mineral materials such as; silica, alumina etc. The effects of abundant bambara nut shell in Africa on the physical, mechanical and morphological properties of epoxy composite have been reported. The results obtained confirm that mechanical properties and physical properties are all largely influenced by the amount of bambara nut shell fillers in the epoxy matrix. Within the range of the variations in this study, the optimal properties of the composites were obtained at filler loading up to 15 wt%. Above this filler thresh load most properties were seen to decrease.

The optimum tensile strength of 80.20 MPa was obtained with 15 wt% of the nano filler, increasing the filler loading to 20 wt% led to a significant decrease in the strength performance to as low as 39.5 MPa. Similarly, modulus of elasticity also dropped from 138.5 MPa to 118 MPa for 15 wt% and 10 wt% respectively.

Other mechanical properties studied in this research such as impact energy and flexural strength shows clearly the optimum bambara nut shell nano filler for epoxy matrix is not more than 15 wt%.

Physical properties such as density of the composite recorded highest value with 5 wt% (0.8g/cm^3) filler loading, increase in the volume of the filler above 5 wt% gave rise to composites with lesser density, confirming that the bambara shell nano filler is a light weight agro material suitable for applications were light weight and high strength is desired.

This suggests that; at optimum filler loading, the composite can find application in industrial as well as domestic purposes, especially where light weight, good tensile strength, and high impact energy are key priority.

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Conflict of interest statement: The authors declare that there are no conflicting interest regarding this work.

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