



Effects of Filler Content and Particle Size on the Mechanical Properties of Unsaturated Polyester Resin Reinforced with Rice Husk-Coconut Shell Particles

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ABSTRACT

The effects of filler content and particle size on the mechanical properties of unsaturated polyester resin reinforced with rice husk-coconut shell particles have been studied. Rice husks (RH) and coconut shells (CS) particles were ground and particle sizes were obtained using 75, 150 and 300 μ m standard sieves. These particles were mixed in equal proportions by weight to form the filler component of the composite. Percentage by weight fractions of 0, 10, 20, 30 and 40 of the filler were blended with unsaturated polyester resin (matrix) by hand lay-up technique. Thirteen composite samples were produced from different formulations. Tensile, flexural, impact and hardness tests specimens were prepared based on ASTM standards. Tensile fractured surfaces were analysed with the help of Scanning Electron Microscopy (SEM) micrographs. It was observed that Ultimate Tensile Strength (UTS) decreased with filler loading with peak value of 19.691MPa at 10% composition of 150 μ m Rice Husk (RH)/Coconut Shell (CS). Flexural strength decreased with particle content. The filled samples had maximum value of 51.201MPa at 20% composition of 75 μ m RH/CS; the Unsaturated Polyester Resin (UPR) recorded 59.085MPa. Tensile and flexural moduli of elasticity followed irregular patterns. Peak values of 2316.248MPa and 3130.887MPa were obtained respectively. Impact tests did not show a specific pattern across all particle sizes. The closely related values peaked at 12.383J at 10% composition of 300 μ m RH/CS. Hardness values decreased with particle loading. The value of 204.833BHN obtained at 10% composition of 150 μ m RH/CS was the maximum. SEM micrographs showed low surfaces interactions, voids, agglomeration of fillers and shrinkage cavities. The fabricated composite can be used in the automobile and building industries.

Keywords: Rice husk, Coconut shell, Unsaturated polyester resin, Composite, Scanning Electron Microscopy

INTRODUCTION

Resource depletion, environmental pollution, high costs and the health threats posed by hitherto popular reinforcing materials like glass and asbestos fibres have made it inevitable for the development of renewable, biodegradable, cheap and hazard-free engineering materials. The ban on asbestos cement based roofing and the high cost of other light roofing materials such as long span aluminium, aluminium-zinc, etc., calls for the development of a cheaper and hazard-free material that can be produced using abundant, renewable and biodegradable raw materials. Bio-fibres have been known to have low impact strength and high moisture absorption leading to dimensional changes resulting to micro-cracking. From available literature, rice husk has been reported to have the least weight among bio-fibres, it is also naturally flame resistant thus making it being processed at higher temperatures than wood. On the other hand, coconut shell has the highest impact and abrasion resistance and least moisture absorption among bio-fibres.

Particulate composites are designed to produce unusual combinations of properties rather than to improve strength. There has been little development in the use of natural fibres or particles as reinforcement materials for polymeric composites [1], and even more so with agro wastes hybrid particle bio-composites. Studies on the mechanical properties of coconut shell (CS) filled polyester composite showed that the tensile strength and Young's modulus of Polyester/CS composites increased with increasing CS content [2]. Morphology studies indicated that the tendency of filler-matrix interaction improved with increasing filler in the polyester matrix. The mechanical behaviour of coconut shell and groundnut shell reinforced-epoxy composite revealed that the maximum tensile strength was ob-

tained from the composite prepared with 40% CSP and GSP volume fraction [3]. The maximum flexural strength was obtained from the composite prepared with 50% CSP and GSP filled. Consequently, the composite prepared with 40% to 50% CSP and GSP filled volume fraction is suitable for application in the interior part of an air craft and automobile where materials with good tensile strength characteristics are required. Investigations on the mechanical properties of Tere-Phthalic unsaturated polyester resin reinforced with varying weight fractions of particulate snail shell showed that the flexural strength of the composite with 20wt.% snail shell particulate reinforcement was greatly enhanced and the impact and hardness properties were greatly improved at 5wt.% filler loading [4]. The composite could be considered for applications in areas where high impact strength is a requirement such as in shipping containers. The 20wt.% snail shell reinforced unsaturated polyester can be used in place of pure polyester for applications where flexibility is of utmost importance. The mechanical properties of rice husk filled cashew nut shell liquid resin composites indicated that better mechanical properties (tensile, flexural and Young modulus) were obtained as the filler loading increased but decreased as the particle sizes increased except for impact, the strength increased as the particle sizes increased [5]. A maximum tensile strength of 35.2MPa was recorded at 30% filler and particle size of 400 μ m. Studies on the mechanical properties of hybrid periwinkle and rice husk filled cashew nut shell liquid composite revealed that the maximum tensile and flexural strengths were obtained at 30% filler content and 400 μ m particle size; the maximum tensile modulus and impact strength were obtained at 800 μ m and 400 μ m particle sizes respectively, for same percent of filler content [6]. The flexural strength from the result converged at 30% filler content. They concluded that optimum properties could be obtained at 30% filler content.

In this study, a hybrid particulate composite was fabricated through hand lay-up technique, using rice husks (RH) and coconut shells (CS) particles as fillers and unsaturated polyester resin (UPR) as matrix. The effects of filler content and particle size on the mechanical properties of the composite were studied.

MATERIALS

Rice husks and coconut shells were locally sourced from Makurdi, Benue State, Nigeria. Unsaturated Polyester Resin (matrix), Methyl Ethyl Ketone Peroxide (catalyst) and Cobalt Naphthanate (accelerator) were from obtained from Ojota Chemical Market, Lagos, Nigeria. The equipment used include standard laboratory sieves, Instron 3369 universal testing machine (50 kN, S/No. 3369K1781, USA), Hounsfield balanced impact machine (48 ft-lb, S/No. 0916, England), Monsato tensometer testing machine (Type "W", S/No. 10055, UK), Phenom Pro X scanning electron microscope (Model No. MVEO 16477830, Netherlands).

METHODS

Processing of Rice Husks and Coconut Shells

Rice husks were cleaned by washing with clean tap water - to free it of sand, dust and bran - and dried under the sun for 2 days. The husks were ground with grain milling machine and the resulting particles were oven-dried at 50°C for 24 hours. Standard sieves of 75, 150 and 300 μ m were used to get different particle sizes. Coconut shells were scrubbed with a sharp knife to remove coir fibres and dust on the body, washed with wire brush and clean tap water and dried for 5 days. The dried coconut shells were firstly ground by manual hammering, then through grain milling machine to get them into powdery particle forms. The particles were oven-dried at 50°C for 24 hours. Standard sieves of 75, 150 and 300 μ m were then used to get different particle sizes. The corresponding particle sizes of both rice husks and coconut shells were uniformly mixed at ratios of 1:1 by weight.

Weighing of Unsaturated Polyester Resin, Catalyst and Accelerator

The UPR was weighed by gently pouring it into a plastic container placed on a digital weighing balance until the weight needed for that particular formulation was achieved. The catalyst, MEKP, was weighed by placing a beaker on the digital weighing balance and in it a test tube placed and the catalyst added gradually into the test tube with the help of a syringe, the weight indication was observed as more drops of catalyst were continually added until the desired weight needed for a particular formulation was achieved. The same procedure was used for the CoNap accelerator. The weights of the catalyst and accelerator were determined by the weight of the UPR (Table 1).

Table -1 Formulation of the Composite at Particle Sizes of 75, 150 and 300 Microns

Ingredient	Weight (Grams)				
	(0%)	(10%)	(20%)	(30%)	(40%)
Rice husk and coconut shell particles (1:1 by wt.)	0.00	54.25	108.50	162.75	217.00
Unsaturated Polyester Resin	(100%) 542.50	(90%) 488.25	(80%) 434.00	(70%) 379.75	(60%) 325.50
Methyl Ethyl Ketone Peroxide Catalyst	(1% of UPR) 5.43	(1% of UPR) 4.88	(1% of UPR) 4.34	(1% of UPR) 3.80	(1% of UPR) 3.26
Cobalt Naphthenate Accelerator	(0.5% of UPR) 2.71	(0.5% of UPR) 2.44	(0.5% of UPR) 2.17	(0.5% of UPR) 1.90	(0.5% of UPR) 1.63

Mixing

During blending, the mass of the UPR was varied with that of the filler to give a total of 542.50g. For 0, 10, 20, 30 and 40% weight fractions of RH and CS particle reinforcements, there were correspondingly 542.50g, 488.25g, 434.00g, 397.75g and 325.50g respectively of UPR after which 0.5% (wt. of UPR) of accelerator and 1% (wt. of UPR) of catalyst (Table 1) were added to each mixture to give better homogenous interfacial interactions. In general, the ratio of unsaturated polyester resin to accelerator to catalyst was 200g: 1g: 2g respectively.

The mixing was done by first of all emptying gently the beaker containing RH and CS particles into a plastic container containing UPR and stirring the resulting mixture with a small paddle for 2 minutes, followed by addition of 0.5% (wt. of UPR) accelerator from the test tube into the mixture and stirring for 2 minutes and finally adding 1% (wt. of UPR) catalyst from the test tube and stirring for 3 minutes while scrubbing the bottom and walls of the beaker to remove any sticking constituents of the mixture.

Fabrication of Composite

The mixture described above was then poured into thirteen plywood moulds – after being greased with mould release agent (Vaseline) - for casting, using hand lay-up technique. On pouring the mixture into the mould cavity (310 x 140 x 8mm), a roller was rolled on its surface to free it of any bubbles, while a brush was used to dress the surface of the mixture. After casting, the composite was allowed to cure for 24 hours at room temperature. The “green” (not fully cured) composite was then cut and trimmed into required geometrical parts, according to test standards.

Mechanical Properties Test

The mechanical properties of the composite were determined using standard test procedures. Three specimens per formulation were tested to obtain average properties. Tensile test (ASTM D638) was done on dog bone specimens with dimensions of $105 \times 10.04 \times 7.22$ mm and gauge length of 55mm at cross head speed of 5mm/min. Flexural test (ASTM D790) was carried out on specimens measuring $105 \times 25.3 \times 6.71$ mm. A span length of 65mm was used on a 3-point flexure fixture. The test was done at cross head speed of 5mm/min. Izod impact test (ASTM D265) was conducted on notched specimens measuring $65 \times 10 \times 7.5$ mm. Brinell hardness test was done on $30 \times 30 \times 7.5$ mm specimens. Brinell hardness number (*BHN*) was calculated using equation 1 [7]:

$$BHN = \frac{w}{\left(\frac{\pi D}{2}\right)(D - \sqrt{D^2 - d^2})} \quad (1)$$

Where *w* is the load on the indenter (200kg), *D* is diameter of the steel ball (10mm), *d* is the average measured diameter of impression in mm and π equals 3.14159.

Scanning Electron Microscopy (SEM)

SEM was conducted on tensile fractured surfaces to study the morphology of the composite.

RESULTS AND DISCUSSION

Results of mechanical tests are shown in Table 2. Fig. 1 shows variation of ultimate tensile strength with filler content. Along all particle sizes, there was decrease in UTS as filler loading increased. The decrease in UTS is attributed to increase in voids (which coalesce in tension to propagate cracks), poor adhesion at the filler-UPR interface, shrinkage cavities (Fig. 10) as filler loading increased, culminating in quicker failure of the samples. At 10% filler content for 75 μ m and 150 μ m, there was increase in strength because the stress transferred by the matrix was supported by the fillers. The peaking of the strength value at 10% and the decrease as filler loading increased was also reported with rice husk [8]. A similar trend was reported with coconut shell [9]. The mechanical properties of particulate-filled polymer micro and nano-composites are affected by particle size, particle content and particle/matrix interfacial adhesion [10]. Composite strength and toughness are strongly affected by all three factors, especially particle/matrix adhesion. Various trends of the effect of particle loading on composite strength and toughness have been observed due to the interplay between these factors which cannot always be separated. Prediction of the strength of composites is difficult. The difficulty arises because the strength of composites is determined by the fracture behaviours which are associated with the extreme values of such parameters as interfacial adhesion [4]. Thus, the load-bearing capacity of a particulate composite depends on the strength of the weakest path throughout the microstructure, rather than the statistically averaged values of the microstructure parameters.

Fig. 2 shows that tensile modulus of elasticity did not follow a specific pattern and was not significantly affected by particle size. Similar observations were made while studying UPR/Coconut fibre and UPR/Bone ash [11-12]. This may be as a result of unevenness in voids density and agglomeration of fillers arising from poor dispersion of fillers in the matrix. The variation of flexural strength with filler content is shown on Fig. 3. Flexural strength, which was greatly affected by particle size, decreased with filler loading. Similar submissions were made on UPR/CS/Groundnut shell particles, UPR/Carbonised eggshell and Epoxy/CS [3, 13-14]. The decrease in flexural

strengths was attributed to voids and weak interfacial bonding as the particles increased which resulted to weak bending load carrying capacity by the matrix.

Fig. 4 represents variation of flexural modulus with filler content. Just like in tensile test, the modulus did not follow a specific pattern and was not markedly affected by particle size. Similar reports were made on UPR/Palm fruit particles; UPR/Bone ash and UPR/CS [1, 12]. Agglomeration of fillers was cited to be responsible for this behaviour. Fig. 5 illustrates variation of impact strength with filler content. Although impact strength was not significantly influenced by filler content and particle size, it was the revelation of the composite because of the high values recorded. There was no specific pattern to the response of the composite to energy absorbed prior to fracture. This fluctuating pattern was also reported while investigating the impact strengths of UPR/Palm fruit particles, UPR/Snail shells and UPR/CS and UPR/Palm kernel shells [1, 4, 15]. Uneven distribution/concentration of micro-cracks resulting from poor interfacial bonding between matrix and filler, shrinkage cavities, voids and agglomeration of fillers are factors responsible for this unpredictability.

Except for some isolated cases, the general trend showed decrease in hardness values with particle loading (Fig. 6). Hardness was also markedly affected by particle size. Similar trends were also reported [1,4,15]. Increase in voids and micro-cracks with filler loading must have made it easier for the indenter to make bigger impressions on the samples. The 204.833BHN obtained at 10% of 150µm compares with peak values of 208BHN and 182.3BHN reported on UPR/CS and UPR/Palm fruit particles respectively [1].

Table -2 Mechanical Properties of the Composite

Property	Filler Content (wt.%)					Particle Size
	0	10	20	30	40	
Ultimate Tensile Strength (MPa)	17.779	18.455	11.891	9.413	9.017	75 µm RH/CS
	17.779	19.691	18.102	12.504	11.037	150 µm RH/CS
	17.779	12.326	7.240	7.095	9.036	300 µm RH/CS
Tensile Modulus (MPa)	1670.656	1851.010	1367.009	1400.446	1516.478	75 µm RH/CS
	1670.656	1938.745	2316.248	1851.742	1742.537	150 µm RH/CS
	1670.656	1698.157	1396.877	1670.356	1780.361	300 µm RH/CS
Flexural Strength (MPa)	59.085	37.221	51.201	41.921	31.171	75 µm RH/CS
	59.085	41.775	47.480	38.086	36.774	150 µm RH/CS
	59.085	41.717	38.148	34.521	27.559	300 µm RH/CS
Flexural Modulus (MPa)	2336.762	1498.901	3130.887	2272.416	1833.498	75 µm RH/CS
	2336.762	1877.914	2267.535	2535.029	2357.755	150 µm RH/CS
	2336.762	2365.729	2554.155	2319.742	1812.246	300 µm RH/CS
Impact Strength (J)	11.570	11.186	11.167	11.660	11.389	75 µm RH/CS
	11.570	12.162	11.665	11.660	11.841	150 µm RH/CS
	11.570	12.383	10.214	10.440	11.778	300 µm RH/CS
Hardness (BHN)	168.741	147.266	181.027	141.134	134.245	75 µm RH/CS
	168.741	204.833	177.671	169.666	135.359	150 µm RH/CS
	168.741	152.450	202.415	156.511	152.716	300 µm RH/CS

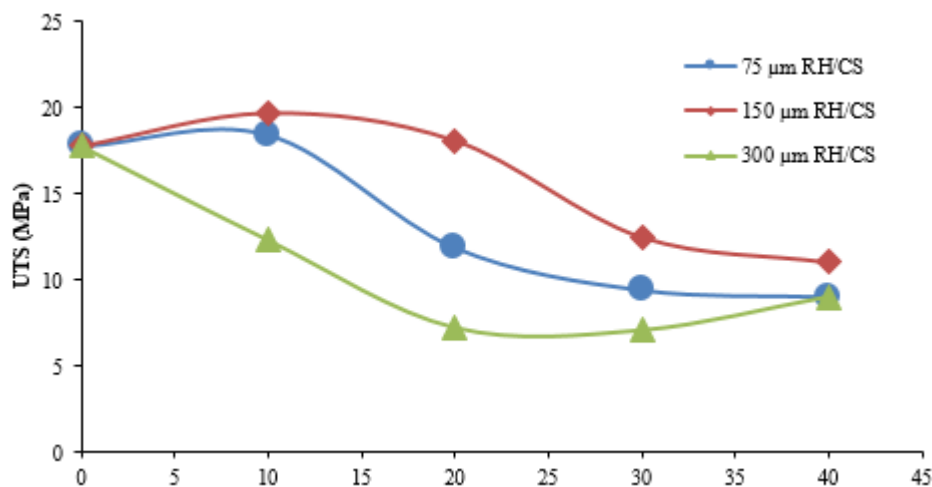


Fig. 1 Variation of Ultimate Tensile Strength with Filler Content

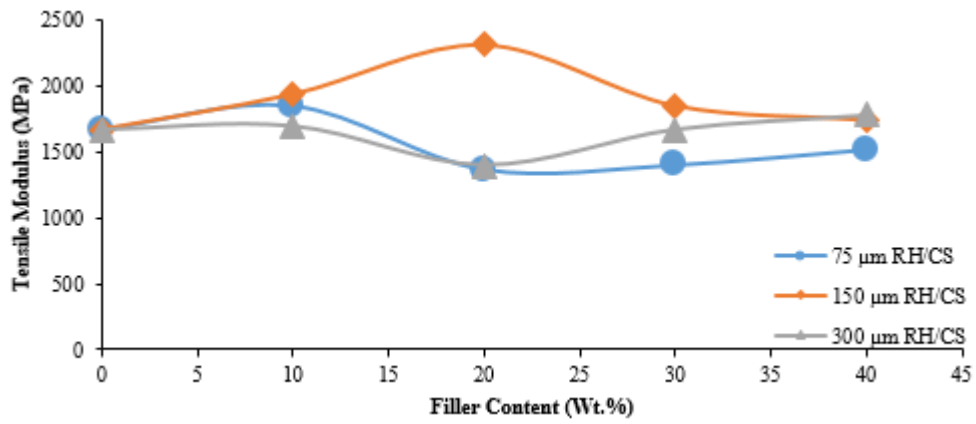


Fig. 2 Variation of Tensile Modulus with Filler Content

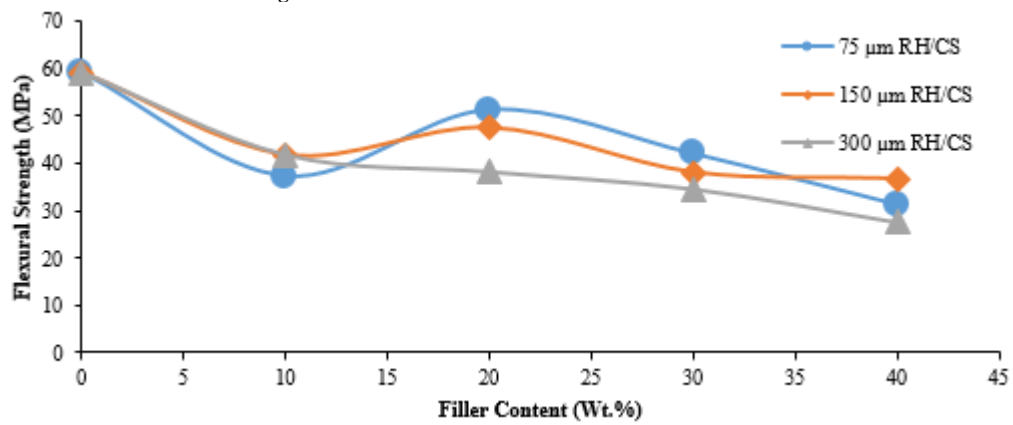


Fig. 3 Variation of Flexural Strength with Filler Content

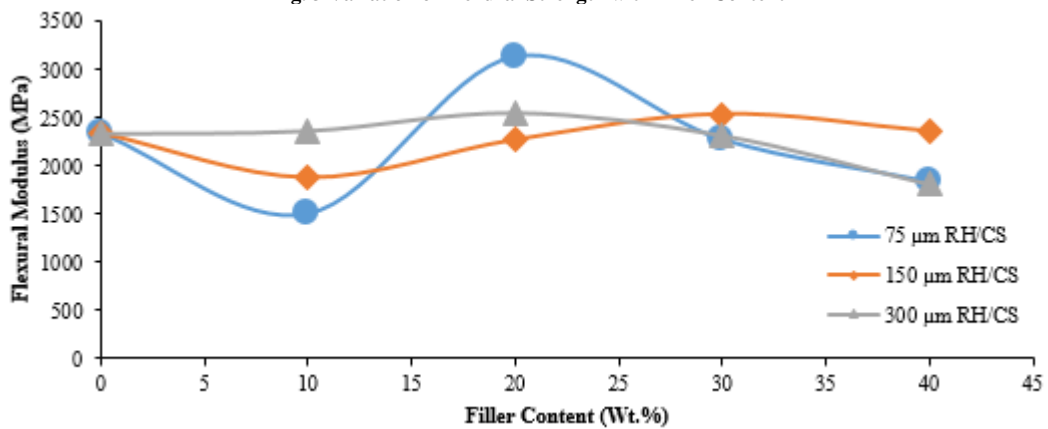


Fig. 4 Variation of Flexural Modulus with Filler Content

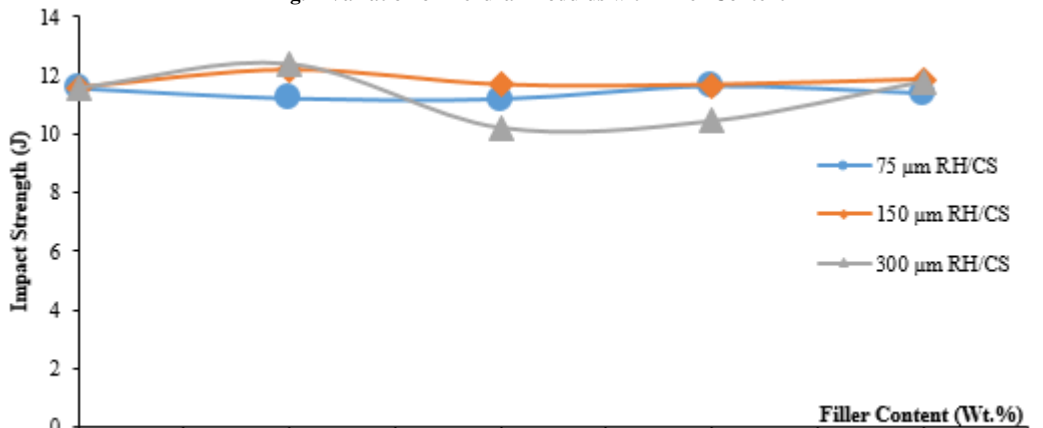


Fig. 5 Variation of Impact Strength with Filler Content

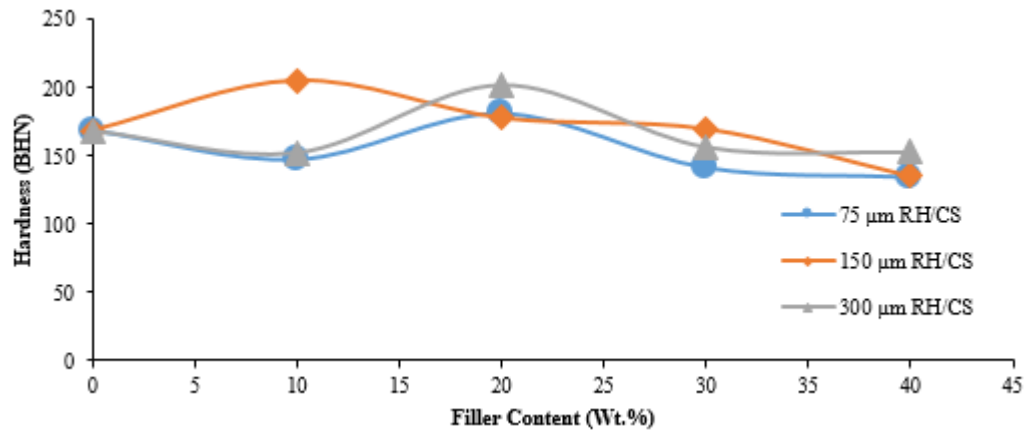


Fig. 6 Variation of Hardness Value with Filler Content

Scanning Electron Microscopy (SEM)

Figures 7-10 are micrographs of tensile fractured surfaces revealing the extent of interaction of matrix with fillers using scanning electron microscope (SEM). Fig. 7 shows a plastic fracture surface of the UPR. This must have accounted for higher flexural strengths compared to other filled, more brittle samples. The impurities observed on the surface can act as crack initiators, thereby lessening the strength of the matrix. Fig. 8 is SEM micrographs of 10% of 75μm. A fracture plane which must have initiated the failure was seen on the surface. This was caused by agglomeration of fillers. Agglomerates create stress concentration zones which might act as crack initiators [16]. On Fig. 9, voids, agglomeration of fillers, low surfaces interaction and detachment of agglomerates were observed on the surface. These were responsible for irregular moduli of elasticity and impact behaviours as well as reduced strengths. Voids, filler chip out, micro-cracks and shrinkage cavity were observed on Fig. 10. These are majorly accountable for reduced strengths and some fluctuating performances of the composite. Figures 8-10 show brittle fracture surfaces due to presence of brittle particles (RH and CS) used as fillers.

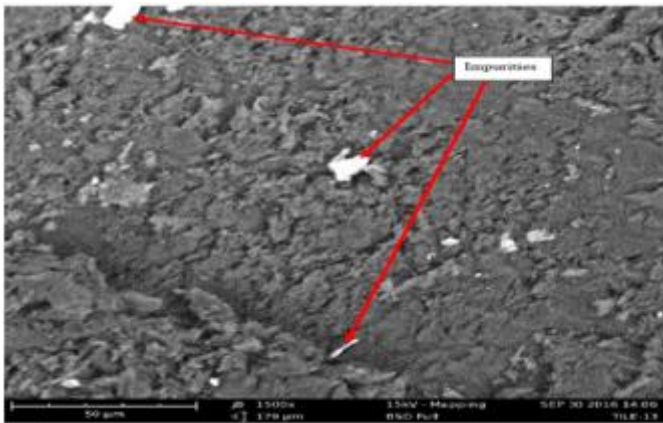


Fig. 7 SEM Micrograph of 100 % UPR

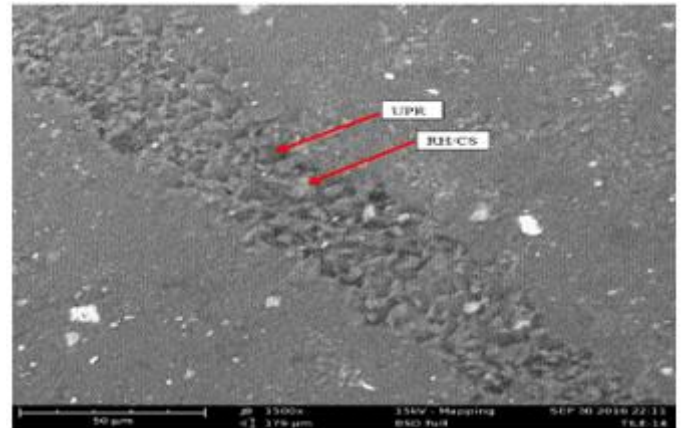


Fig. 8 SEM Micrograph of 10 % 75 μm RH/CS Particles

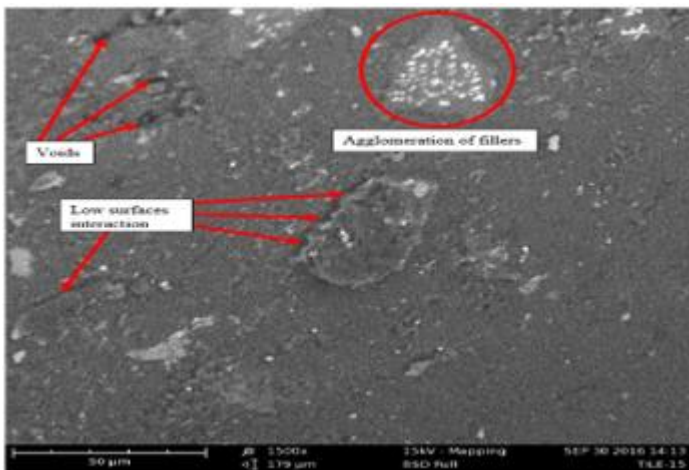


Fig. 9 SEM Micrograph of 10 % 150 μm RH/CS Particles

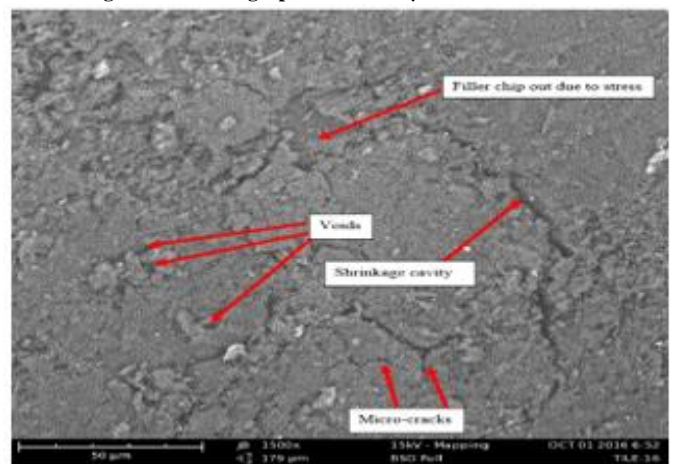


Fig. 10 SEM Micrograph of 10 % 300 μm RH/CS Particles

CONCLUSION

A hybrid particulate composite using rice husk and coconut shell particles as fillers and unsaturated polyester resin as matrix has been developed. It was observed that filler loading had more impact on the mechanical properties of the composite than particle size. Ultimate tensile and flexural strengths decreased with filler content. The strengths were also greatly affected by particle size. The variation of tensile and flexural moduli with filler loading followed irregular patterns. The moduli of elasticity were not markedly affected by particle size. Impact test recorded the highest values when compared with other similar literatures. Bar 150µm particle size, impact strength did not follow a specific pattern. Impact strength was not markedly influenced by filler content and particle size. There was decrease in hardness values with particle loading. The effect of particle size on hardness was significant. Although, filler loading gave rise to better composite properties, the virgin matrix exhibited better flexural strength. Morphological tests show plastic fracture of the control sample and brittle fracture of the loaded samples. It also showed voids, low surfaces interactions, agglomeration of fillers, filler chip out, detachment of agglomerates, micro-cracks and shrinkage cavities. These were responsible for the behaviours of the composite samples during tests. The fabricated composite can be used in making interior car components (dashboard, door claddings, central consoles and seat carriers). Applications in the building industry include ceiling and roofing sheets.

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