

## Effects of bone particle on the properties and microstructure of polypropylene/bone ash particulate composites

F. Asuke<sup>a</sup>, V.S. Aigbodion<sup>b,\*</sup>, M. Abdulwahab<sup>a,c</sup>, O.S.I. Fayomi<sup>c</sup>, A.P.I. Popoola<sup>c</sup>, C.I. Nwoyi<sup>d</sup>, B. Garba<sup>a</sup>

<sup>a</sup> Department of Metallurgical and Materials Engineering, Ahmadu Bello University, Zaria, Nigeria

<sup>b</sup> Department of Metallurgical and Materials Engineering, University of Nigeria, Nsukka, Nigeria

<sup>c</sup> Department of Chemical and Metallurgical Engineering, Tshwane University of Technology, Pretoria, South Africa

<sup>d</sup> Department of Metallurgical and Materials Engineering, Nnamdi Azikwe University, Awka, Anambra State, Nigeria

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### ABSTRACT

The effects of uncarbonized (fresh) and carbonized bone particles on the microstructure and properties of polypropylene composites have been studied. The addition of the bone particles ranges from 5 to 25 wt% in uncarbonized and carbonized forms in the polypropylene matrix. The physical and mechanical properties were used as criteria for the evaluation of the composites. Microstructures of the composites were assessed with scanning electron microscopy (SEM/EDS). The results revealed that the addition of carbonized bone particles reinforcement has superior properties than uncarbonized bone particles composite based materials with an increase in the compressive strength, hardness values, tensile strength and flexural strength by 53%, 45% and 35% respectively, and a decrease in the % elongation and impact energy by 75% and 32%, respectively at 25 wt% carbonized bone, with 28% increase in tensile strength at 15 wt% carbonized bone addition. The increase in strength and hardness values was attributed to the distribution of hard phases of bone particles in the ductile polypropylene matrix.

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### 1. Introduction

Polymer composite materials are being used in a wide range of structural applications in the aerospace, construction and automotive industries due to their lightweight and high specific stiffness and strength [1]. A variety of materials are being used ranging from lower performance glass fiber/polymer, used in small sail boats and domestic products, to high performance carbon fiber epoxy systems used in military aircraft and spacecraft [2]. One sector where the use of composite materials is still evolving is the automotive industry. Composite materials offer great potential in reducing vehicle weight, thus increasing fuel efficiency and reducing CO<sub>2</sub> emissions. In addition to weight reduction, the number of individual parts can be significantly reduced making the high-volume composite car concept cost effective [2,3].

In recent years there is a perceived shortage of wood fiber for composite products due to competition for fiber by pulp mills, reduced harvesting and manufacturing and diminished log quality. Also, there is pressure from environmentalists to reduce forest use and regulatory legislation pending on disposal of agri-fibers [1–5]. For example, any potential to reduce field burning is an environmental benefit and helps address the issue of restricted open burning. There are tremendous quantities of agricultural biomass

available for non-agricultural uses such as paper and composite products [5].

Previous studies have proved that Chicken eggshell (ES) is an agriculture byproduct that has been listed worldwide as one of the worst environmental problems, especially in those countries where the egg product industry is well developed. In the U.S. alone, about 150,000 tons of this material is disposed in landfills [5]. ES contains about 95% calcium carbonate in the form of calcite and 5% organic materials such as type X collagen, sulfated polysaccharides, and other proteins [4,5].

Patricio Toro et al. [5], studied eggshell, a new bio-filler for polypropylene composites. The work proved that ES composites showed lower modulus of elasticity ( $E$ ) values than talc composites, talc filler could be replaced by up to 75% with ES while maintaining a similar stiffness and  $E$  compared to the talc composites. Abdullah et al. [6] studied the water absorption and mechanical properties of high-density polyethylene/egg shell composite. It was found that the addition of egg shell powder to the polymer leads to decrease in the tensile strength, modulus of elasticity, and shore-D hardness on the other hand it increases the % elongation at break and impact strength. Water absorption of the composite behaviors as a function of days was also investigated, and it increases by increasing the exposure time for the same filler content, while the absorbed amount of water increases, by increasing the wt% of egg shell's constant exposure time. Based on the foregoing this present research is looking at the possibility of using

\* Corresponding author. Tel.: +234 8028433576.

E-mail address: [aigbodionv@yahoo.com](mailto:aigbodionv@yahoo.com) (V.S. Aigbodion).

cow bone as a reinforcer in polymer matrix composites, since egg-shell and bone belong to the same calcium group [7].

## 2. Experimental procedures

### 2.1. Materials/equipment

Polypropylene (matrix), bone (uncarbonized and carbonized), stain remover (water soluble), aluminum foil, polishing agent, grit paper, etchant, water and lubricant. Set of sieves (mesh sizes; 1000, 750, 500, 250, and 100  $\mu\text{m}$ ), hacksaw, Universal tensile testing machine, Digital Rockwell hardness machine, Charpy Impact machine, measuring cylinder, Vernier caliper, bone crusher, two roll mill, hydraulic press, crucible furnace, a pair of tongs, digital weighing balance, polishing machine, optical and scanning electron microscope.

### 2.2. Methods

The flowchart used in this study is shown in Fig. 1: the fresh cow bones (limb bones) were collected from an abattoir in Tudun Wada, Zaria, Nigeria, washed and cleaned to a good physical condition. The bones were then divided into two; one part for carbonization (CB) and the other used as uncarbonized (FB). The former was carbonized in a crucible furnace at about 550  $^{\circ}\text{C}$  for 45 min. Bone crusher was used to crush the bones to a certain particle size ( $\leq 2$  mm) and the latter ground to  $\leq 1000$   $\mu\text{m}$  by replacing the crusher sieve with 1 mm sieve [7]. Sieving was done using a set of sieves having mesh sizes of 1000, 750, 500, 250 and 100  $\mu\text{m}$  diameter to obtain small particle size. After sieving, the under size of 100  $\mu\text{m}$  was used as reinforcement (see Fig. 2) [6,7].

The bone was then degreased so as to get rid of any trace of oil and blood that will inhibit proper bonding between the matrix (polypropylene) and the reinforcement (bone) using a water soluble stain remover. The process utilized the differential weight between the bone and the associating oily substances (gravity separation) by pouring water (mixed with stain remover) and

allowing it to settle for about 30 min. This process was repeated using water without stain remover several times until a clean (shallow) surface was observed on the surface of water. The bone was found negative when tested for the presence of both oil and blood [7].

Compounding or mixing was done not only to mix but also to ensure intimate mixing of composite forming constituents. The two roll mill machines, were switched on and set for preheat to a temperature of 180  $^{\circ}\text{C}$  for 1 h. The polypropylene (Fig. 2c) matrix was introduced when the two rolls are just closed by regulating the gap between them. After 2 min, a measured amount of filler (bone particles); 5, 10, 15, 20, 25 wt% was introduced as carbonated ash bone (Fig. 2a) and uncarbonized (fresh) powdered bone (Fig. 2b). The mixture was left for 8 min to achieve effective homogenization. The mixture was then ejected and allowed to cool.

The blended material was pressed using a hydraulic press at elevated temperature ( $\approx 180$   $^{\circ}\text{C}$ ) to a thickness of the mold. It was pre-pressed prior to melting for 45 min using a Moore hand operated press at high speed (low pressure) to prevent fracture. It was later pressed at a low speed (high pressure) for 3 min. The compact was removed while still hot to obtain a smooth surface as in (Fig. 2d). The same procedure was used for the remaining composition. After, the developed composites were sectioned into test samples for properties' evaluation.

The density of the samples was determined by measuring the weight of the sample ( $m$ ) and then by immersing into a measuring cylinder half filled with water and recording the increase in volume ( $v$ ) as the volume of the sample. The density of each sample ( $\rho$ ) was obtained using equation 1 [8]:

$$\rho = \frac{m}{v} (\text{g/cm}^3) \quad (1)$$

The water absorption was determined by weighing the sample ( $w_1$ ) and placing in a closed container containing water. The sample was then weighed after 24 h as ( $w_2$ ). The percentage weight gained was calculated and recorded for each sample using the following formula [6,7]:

$$\% \text{weight gained} = \frac{w_2 - w_1}{w_1} \times 100\% \quad (2)$$

Tensile test was carried out using a Hounsfield tensometer. The samples were cut into suitable dimensions and a gauge length of 25 mm was marked using a scribe. Each sample was subjected to test by loading to its maximum load carrying capacity, after which the tensile strength was calculated using Eq. (3). The corresponding strain was also computed [8].

$$\sigma = \frac{\text{max.load}(P)}{\text{Initial cross sectional area}(A)} (\text{N/m}^2) \quad (3)$$

The extent to which the material elongates is determined by measuring the greatest separation between the gauge marks just before rupture, and is often expressed as % elongation [8].

Thus:

$$\% \varepsilon = \frac{\text{gauge length after rupture}}{\text{initial gauge length}} \times 100 \quad (4)$$

The compression test was carried out using a universal tensile testing machine. A compression load was applied and the maximum load was read directly from a digital meter. The compressive strength was then calculated using Eq. (3).

Bend or flexure test was carried out with the same Hounsfield tensometer. Samples were cut into suitable dimensions, each sample was bend tested using an improvised support and a center point load until it breaks. If a load ( $P$ ) is applied over the entire span ( $L$ ), and act on a cross-sectional area ( $A$ ), then flexure strength could be calculated using equation 5 [8].

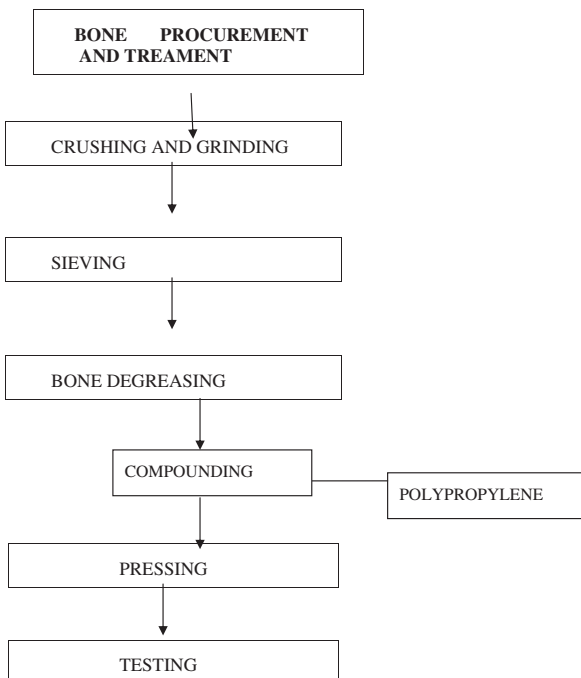


Fig. 1. Schematic diagram of the flow sheet for the development of polypropylene/bone reinforced composite.

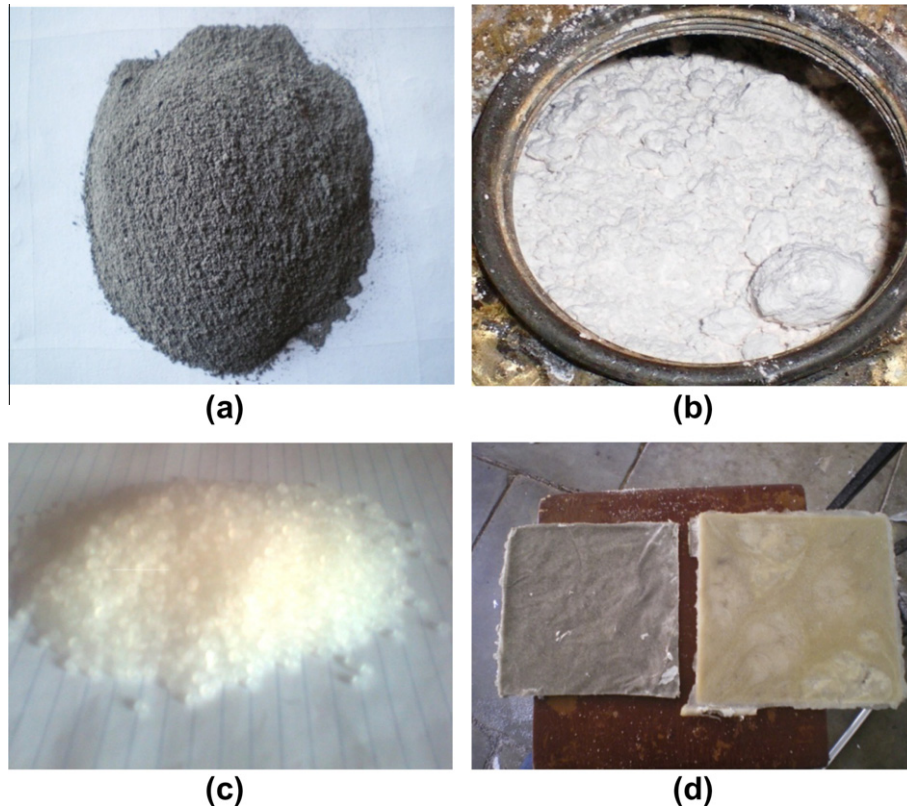


Fig. 2. Micrograph of (a) carbonized bone powder (CB) (b) fresh bone powder (FB) (c) polypropylene (d) pressed PP/bone composite.

$$\text{Flexural strength} = \frac{3(\text{load})(\text{span})}{2(\text{width})(\text{thickness})^2} (\text{MPa}) \quad (5)$$

Rockwell hardness testing machine with 1.56 mm steel ball indenter was used to test the hardness of the samples on a F scale. The hardness of each sample was read directly from the Rockwell tester [8].

The impact test was carried out on the samples using a charpy impact testing machine to determine the impact energy. A v-notch of 2 mm deep was created at 45° into a 10 × 3.3 × 55 mm standard specimen for charpy impact test which is intended to provide the stress concentration during the impact test [1,8]. The sample was then placed on the machine and the pendulum was allowed to hit the specimen when it swings under gravity. The impact energy was obtained by reading energy loss of the pendulum as a result of hitting the sample directly from the machine.

Samples were sectioned from each composition for metallographic examination. The samples were ground with grit papers (80–600 grits) using water as coolant. Polishing was followed using alumina polishing powder mixed with distilled water and later etched with Keller's reagent. The microstructures obtained were photographically captured using an optical microscope with built-in camera [10]. The surface of developed composites with 0%, 15% and 25% from both uncarbonized and carbonized samples were observed using a scanning electron microscope equipped with energy dispersive spectroscopy (SEM-EDS) and optical microscope (OPM).

### 3. Results and discussion

SEM is used to study the morphology of the composites produced. Plate 1 shows the SEM/EDS micrograph of the polymer matrix, while Plates 2–5 show the SEM/EDS of the composites.

Morphological analysis using SEM clearly shows the difference in the morphology of the polymer matrix and its composites (see Plates 1–5). The microstructure clearly shows that when the bone particle was added to the polymer, morphological changes in the structure took place.

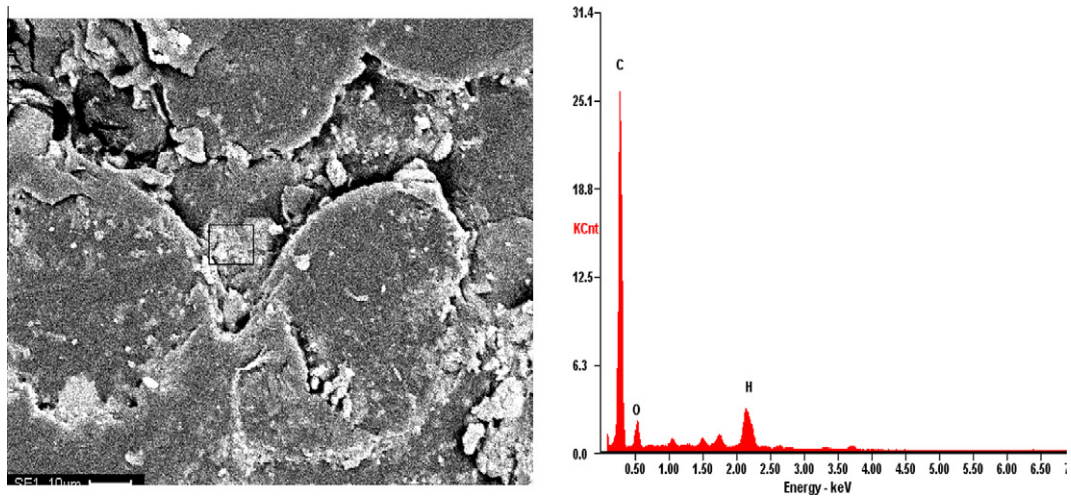
The density of the composites increases with an increase in weight percent of bone (Fig. 3) owing to the fact that bone is denser than PP. It can also be seen that relatively higher values of densities are recorded for the composites with carbonized bone. This can be explained by the fact that during carbonization, the spongy portion of the bone is being decomposed and densified, resulting in the elimination of volatile matters [7].

In Fig. 4, it can be seen that there is an increase in the amount of water absorption. This is because, the matrix (PP) and the reinforcement (bone) are not in any way soluble in one another and interfacial bonding between the bone and the matrix phases may be not be perfect. This results in inter-phase and inter-particle spaces (porosity). As the wt% of bone increases, the level of porosity also increases hence the amount of water absorption. Composites of uncarbonized bone (FB) particles showed a higher level of porosity by absorbing relatively large amount of water than composites with carbonized bone (CB). This is because any separation in the bone as a result of loose particles is removed when carbonizing the bone to obtain carbonized particles. This result is in agreement with the outcome obtained when polyester was reinforced with iron fillings [6,7,9].

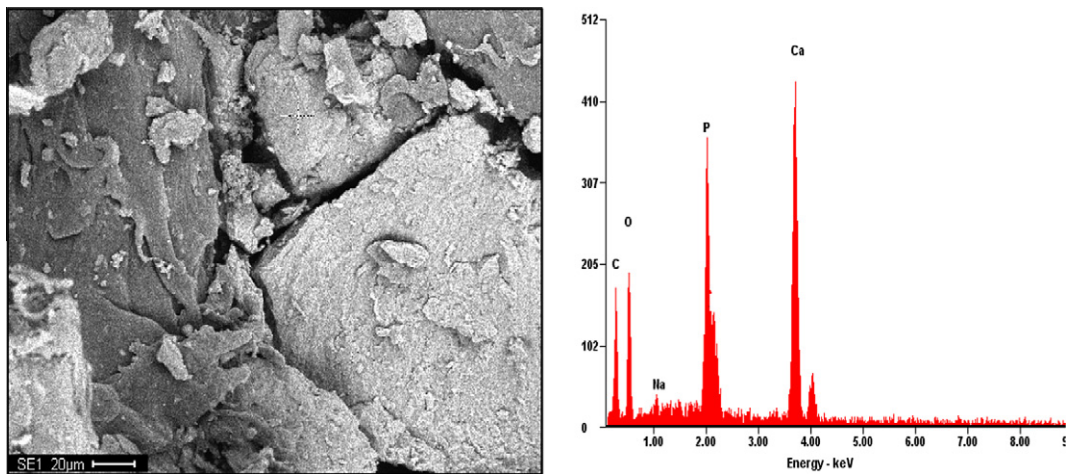
As the wt% of bone increase, there is a corresponding increase in tensile strength (Fig. 5) owing to the fact that as the volume of reinforcement increases, its strengthening effect also increases.

A superior property was recorded while testing PP reinforced with CB particles this is due to the removal of spongy portion and subsequent elimination of volatile matters during carbonization [10–12]. From the results of EDS, it was found that there

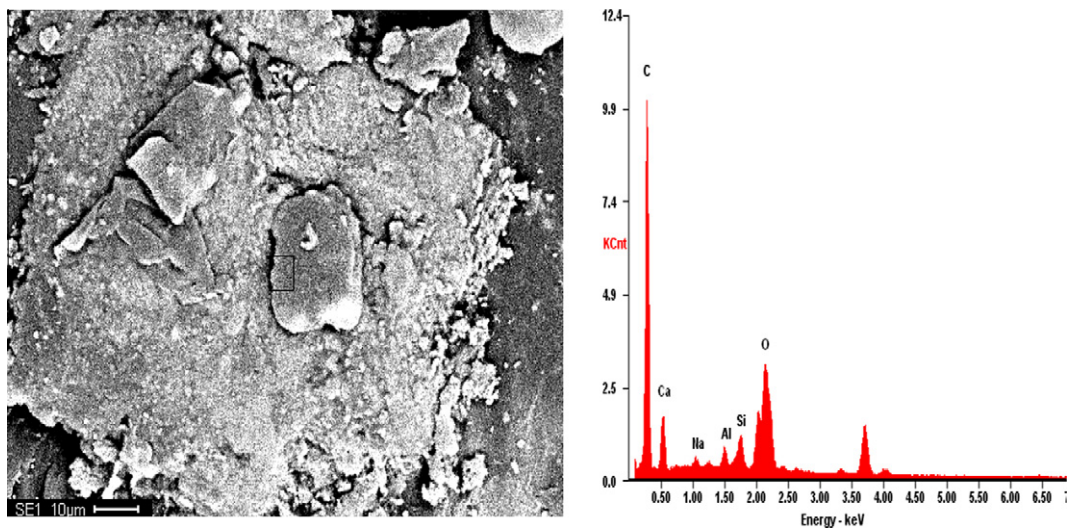




**Plate 1.** SEM image of the polypropylene matrix with the ED ( $\times 500$ ).



**Plate 2.** SEM image of the polypropylene matrix with 15% FB ( $\times 500$ ).



**Plate 3.** SEM image of the polypropylene matrix with 15% CB ( $\times 500$ ).

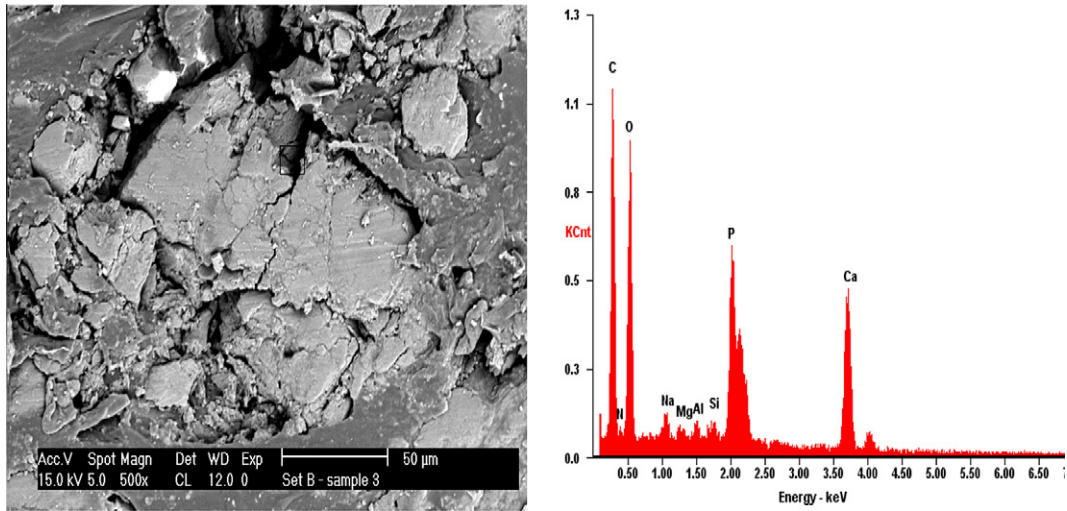


Plate 4. SEM image of the polypropylene matrix with 25% FB (×500).

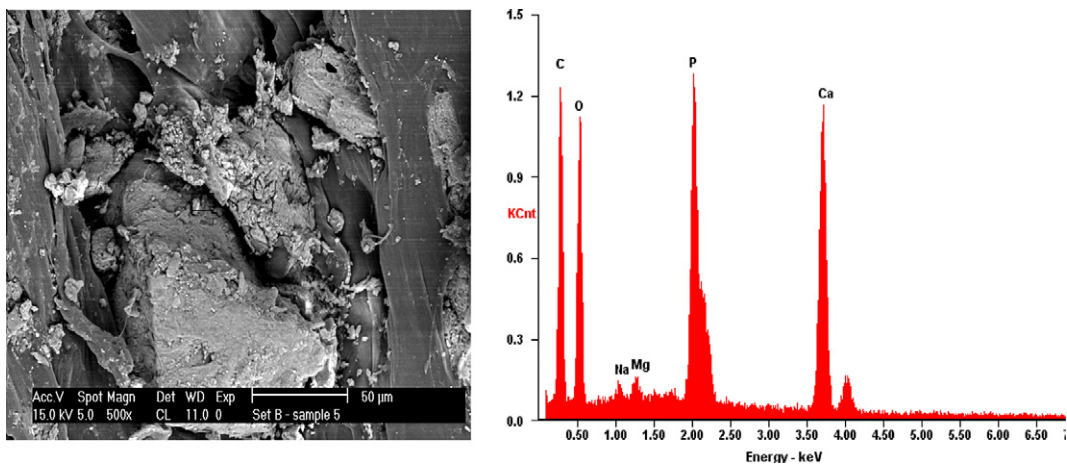


Plate 5. SEM image of the polypropylene matrix with 25% CB reinforcement (×500).



Fig. 3. Variation of density with increase in bone percentage.

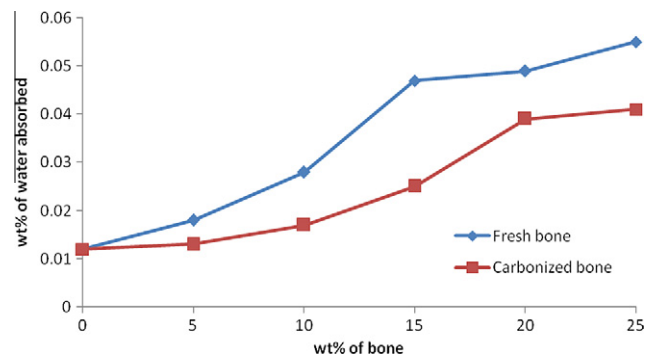


Fig. 4. Variation of water absorbed with wt% of bone particle after 24 h.

was high level of carbon as against that of composites with uncarbonized bone particles which could result in higher strength values when PP was reinforced with CB. The tensile strength recorded for CB and FB composites are: 31.73 MPa and 29.88 MPa respectively at 15 wt% of the reinforcement which can be attributed to proper bonding dispersion of reinforcing phase as seen in Plates 2 and 3. Meanwhile, from the micrographs, it can also be observed that clustering of reinforcing phase gave rise to a fall in tensile strength

above 15 wt%. This is also in agreement with recent research [9–15].

From Fig. 6, hardness value of the composite increased with an increase in weight percent bone addition. The increase in hardness is a result of increase in the hard and brittle phase of bone particles in the PP matrix. Similarly, due to the absence of loose constituents of bone (spongy) and volatile matters in CB, its addition resulted in

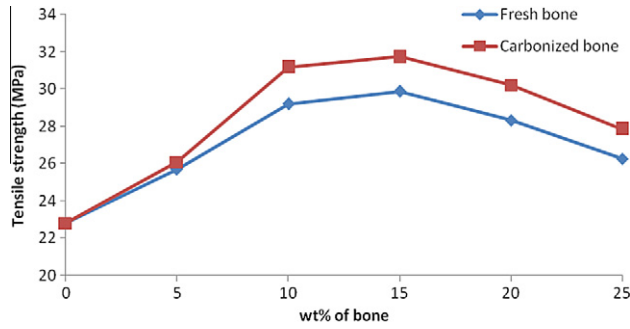


Fig. 5. Variation of tensile strength with wt% of bone particle.

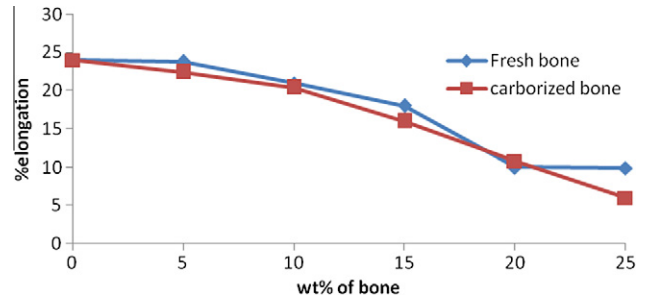


Fig. 7. Variation of % elongation with wt% of bone.

a superior hardness value than FB. This result is in agreement with many recent researches [9–15].

From Fig. 7, as the percentage of bone increases, % elongation decreases; this is because there is a loss of ductility in the composite with increase in wt% bone. This can be justified by the change in fracture surface (visual examination) from cup and cone (for ductile) to relatively flat and smooth surface, which is an indicative feature of embrittlement. The decrease in elongation is more prominent in CB this can be justified by the fact that its fracture surface revealed more flatness which comes as another brittle aspect [9].

From the result of impact test (see Fig. 8), it was found that as the wt% of bone increases, the energy absorbed on impact decreases with the exception of 0–5 wt% and 5–10 wt% in FB and CB respectively. The rise in those ranges are believed to be a result of inter-particle spacing which often tends to slowdown the nucleation of cracks by absorbing some fraction of energy. This is in agreement with findings of recent research [9–15].

It was observed that (see Fig. 9) as the weight of bone increases in the composite, the flexure strength also increases. Bearing in mind that CB is denser than FB, CB shows superior flexure property than FB. The drop and unsteady increase in FB and subsequent drop is attributed to uneven dispersion and poor bonding between the matrices and reinforcing phase (Plates 4 and 5). This result is in agreement with the recent result obtained in recent research [9].

From the data obtained in Fig. 10, it can be seen that as the weight of bone increases there is an increase in compression properties. The increase in compression strength is attributed to the good compression properties of bone. However, reinforcement with CB gives superior properties over FB and this may be a result of proper interfacial bonding between CB and PP matrix as revealed by micrograph. This result is in agreement with other researches [9,10].

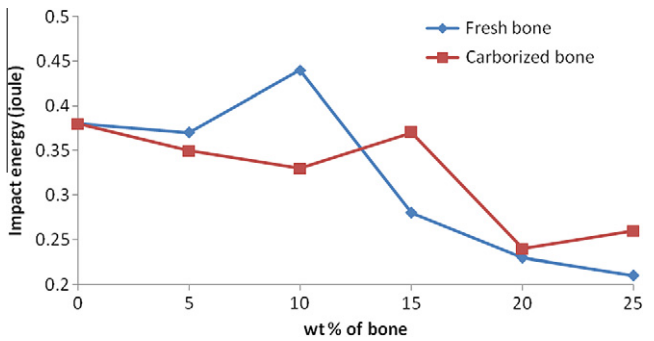


Fig. 8. Variation of impact energy with wt% of bone.

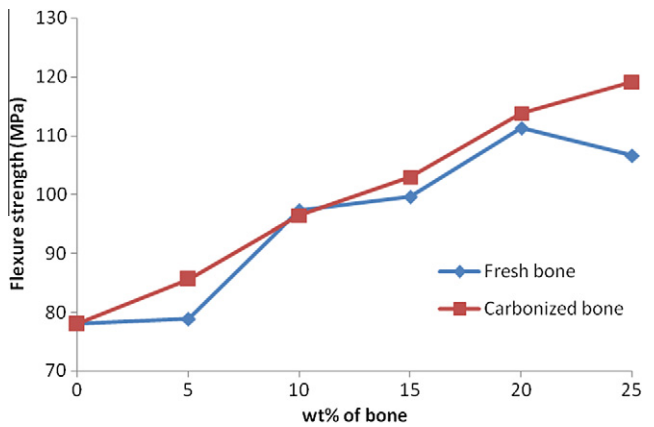


Fig. 9. Variation of flexural strength with wt% bone.

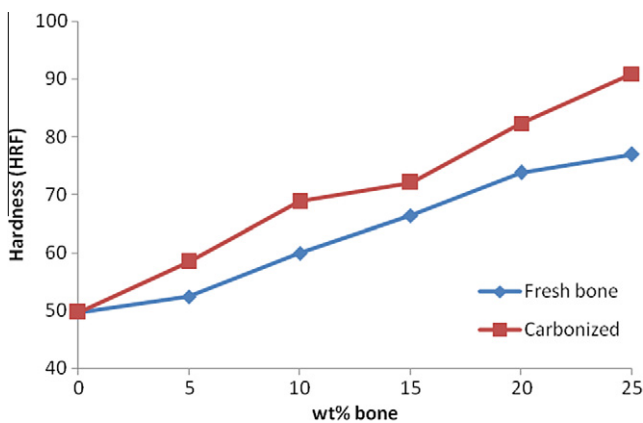


Fig. 6. Variation of hardness with wt% of bone particle.

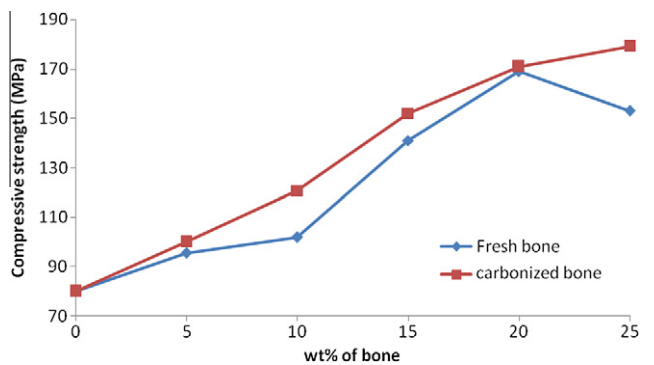


Fig. 10. Variation of compressive strength with wt% of bone.

#### 4. Conclusions

In the present research, different experimental techniques have been used to characterize the microstructure and properties of PP and its composites containing different volume fractions of bone particles. From the results of the investigations and discussions in the preceding section, the following conclusions have been made.

- 1) This work shows that successful fabrication of PP and the bone particles composite by compounding and compression molding.
- 2) The uniform distribution of the bone particles in the microstructure of the polymer composites is the major factor responsible for the improvement in the mechanical properties.
- 3) There is not much difference between the density of unreinforced PP and that reinforced with bone particles.
- 4) The hardness values obtained from PP reinforced with bone particles increased with an increase in the weight fraction of bone particles.
- 5) Absence of volatile matters, high carbon content and high density of carbonized bone are believed to be responsible for its relatively superior properties.
- 6) The developed composites have better properties at the ranges of 5–15 wt% bone particle additions, and for optimum service condition, bone particle addition should not be exceeding 15 wt% in order to have better properties.
- 7) The work provides a unique combination and a wide range of both physical and mechanical properties of polypropylene, a thermoplastic polymer without much sacrifice to its density through reinforcement with bone which is light in weight, yet hard and strong.

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