

The physio-mechanical properties of recycled low density polyethylene (RLDPE)/bean pod ash particulate composites



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ABSTRACT

This work presents a systematic approach to evaluate the physio-mechanical properties of bean pod ash particles (BPAP) reinforced recycled polyethylene (RLDPE) polymer based composites. The bean pod ash particles of 75 μm with a weight percentage of 0, 5, 10, 15, 20, 25, 30 (wt%) and recycled polyethylene (RLDPE) were prepared. The surface morphology, physical and the mechanical properties of the composites were examined. The results showed that the fair distribution of the bean pod ash particles in the microstructure of the polymer composites is the major factor responsible for the improvement in the mechanical properties. The bean pod ash particles added to the RLDPE polymer increased the percentage of water absorption and improved its rigidity, modulus and hardness values of the composites. The tensile and flexural strengths increased to a maximum of 20.1 and 39.0 N/mm^2 at 20 wt% BPAP respectively. Based on the results obtained in this study, it is recommended that the composites can be used in the production of indoor and outdoor applications.

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Introduction

Polymer composite materials are being used in a wide range of structural applications in aerospace, construction and automotive industries due to their lightweight nature, high specific stiffness and strength [1]. A variety of materials are being used ranging from lower performance of glass fibre/polyester used in small sail boats and domestic products, to high performance of carbon fibre epoxy systems used in military aircraft and spacecraft [2,3].

In recent years there is a perceived shortage of wood fibre for composite products due to competition for fibre by pulp mills, reduced harvest and diminished log quality. Also, there is pressure from environmentalists to reduce forest use and regulatory legislation pending on the disposal of agro-fibres [4,5]. The use of lignocellulosic natural fibres/particles as fillers or reinforcements has been gaining acceptance in polymer applications in the past few years [2,3]. The natural fillers can be obtained from several sources, both from forestry and agricultural resources. Waste from agriculture delivers renewable, abundant, natural materials that serve as fillers for polymers, with the benefit of lower cost and improved mechanical properties.

Many researches have been reported on natural fillers reinforced thermoplastic composites, which have successfully proved their applicability in various fields [4]. Thermoplastics such as polyethylene (PE), polypropylene (PP) and polyvinylchloride (PVC) have been compounded with natural fillers such as wood, hemp, cotton, coir pineapple leaf, oil palm and banana to prepare composites [4–7].

Shibata et al. [8] studied the biodegradable composites reinforced with bagasse fibre before and after alkali treatments. Mechanical properties of the composites made from alkali treated fibres were superior to the untreated fibres. Approximately 13% improvement in tensile strength, 14% in flexural strength and 30% in impact strength had been found respectively. In alkali treated fibres the aspect ratio of the fibres contributed to the enhancement of the mechanical properties of the composites.

Corbiere-Nicollier et al. [9] reported that kraft pulped banana fibre composite has good flexural strength. In addition, short banana fibre reinforced polyester composite was studied by Pothan et al. [10]; the study concentrated on the effect of fibre length and fibre content. The maximum tensile strength was observed at 30 mm fibre length while maximum impact strength was observed at 40 mm fibre length. Incorporation of 40% untreated fibres provides a 20% increase in the tensile strength and a 34% increase in impact strength.

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Luo and Netravali [11] studied the tensile and flexural properties of the green composites with different pineapple fibre contents and compared them with the virgin resin. Sreekala et al. [12] performed one of the pioneering studies on the mechanical performance of treated oil palm fibre-reinforced composites. They studied the tensile stress–strain behaviour of composites having 40% by weight fibre loading.

Ichazo et al. [13] found, that adding silane treated wood flour to PP produced a sustained increase in the tensile modulus and tensile strength of the composite. Mohanty et al. [14] studied the influence of different surface modifications of jute on the performance of the biocomposites. More than 40% improvement in the tensile strength occurred as a result of reinforcement with alkali treated jute. Jute fibre content also affected the biocomposite performance and about 30% by weight of jute showed optimum properties of the biocomposites.

Agunsoye et al. [15] reported on the effect of particulate cow bone additions on the mechanical properties and tribological behaviour of cow bone reinforced polyethylene. They observed that the tensile strength and the hardness values of the composite increased with an increase in wt% of cow bone particles while the impact strength and rigidity decreased. Also the addition of the particulate cow bone had the most significant effect on the wear behaviour of the composite while, the interactions between load and time had no significant effect.

Beans pod is a waste by product of agricultural processing of bean seeds. Across the globe, much research efforts in recent times are geared towards possible ways of recycling wastes for reuse to keep the environment clean and safe [16]. Bean plant (*Parkia Biglobosa*), is the material resource required for the production of bean pod ash (BPA). The harvested fruits are ripped open while the yellowish pulp and seeds are removed from the pods. Pods make up 39% of the weight of the fruits while the mealy yellowish pulp and seeds make up 61%. The pod ash is used for soap making and for dyeing the traditional indigo clothes [17].

Despite the availability of these studies, no investigation has been conducted on the application of the bean pod ash particles (BPAP) in polymer composite materials. This paper therefore reports for the first time the potential of using bean pod ash particles in reinforcing polymer composites. Based on the above-mentioned situation, the study described in this work intends to investigate physio-mechanical properties of RLDPE composites reinforced with bean pod ash particles.

Method

The uncrushed bean pods were alkali treated using 2.5% NaOH solution. After treatment, bean pods were packed in a graphite crucible and fired in a control atmosphere muffle electric furnace at a temperature of 1300 °C for 5 h to form carbonized bean pod ash particles (BPAP). The particle size analysis of the BPAP was carried out in accordance with ASTM-60. 100 g of the BPAP was placed on a set of sieves arranged in descending order of fineness and shaken for 15 min which is the recommended time to achieve complete classification. The weight retained on 75 µm was used in this research [18].

The RLDPE matrix and the BPAP were pre-dried prior to compounding. The mixture was compounded using a co-rotating twin extruder (APV Baker Ltd. England, Model: MP19PC) with an L/D ratio of the screw of 25:1. Mixing speed of 60 rpm and 140 °C was maintained for all the compositions [18].

Metal moulds were used in the production of the composite samples. The compounded material was placed in a 350 mm × 350 mm rectangular mould and pressed to a thickness of 4 mm with a pressure of 10 MPa and a temperature of 160 °C. At the

end of the press cycle the composites were removed from the press for cooling. BPAP varied from 5 to 30 wt% with a 5 wt% interval.

A scanning electron microscope (SEM) JEOL JSM-6480LV was used to examine the surface morphology of the BPAP and the composite samples. Samples were washed, cleaned thoroughly, air-dried and were coated with 100 Å thick platinum in a JEOL sputter ion coater and observed in SEM at 10 kV [7].

Test samples were cut from the composites for the physical and mechanical test according to the recommended standards. Prior to the test, all the samples were conditioned at a temperature of 23 ± 2 °C and relative humidity of 65% according to ATM D618-08 [18].

Density was measured using the gravimetric method g/cm³ by Geopyc 1360, Micromeritics Germany. A known inner volume of glass cylinder was taken which is directly connected to the precious balance for measurement. The glass cylinder was equipped with two automatic sensors which control the cylinder full or empty. Composite samples were dried at 80 °C for 48 h before measurement. The dry sample mass was taken while the glass cylinder was full. The density was calculated by dividing the dry mass of each sample by the known volume of glass cylinder [4].

Water absorption test was performed according to IS: 2380 (PART XVI) standard. Test specimens of 75 mm × 50 mm and thickness 4 mm were prepared for the water absorption test. This test covers the method of determination of water absorption of composites from lignocellulosic materials. The specimens were immersed in water for 24 h. The water absorption of composites was measured by the weight gain of the material at regular intervals. The percent of water absorption is expressed as the ratio of increase in the mass of the specimen to the initial mass.

The hardness values of the 'cast' specimens were determined using the Rockwell hardness tester on "B" scale (Frank Well test Rockwell Hardness Tester, model 38506) with a 1.56 mm steel ball indenter, minor load of 10 kg, and major load of 100 kg and a hardness value of 101.2HRB as the standard block [19].

The tensile test was performed according to ASTM D638 standard using a Universal Testing Machine at a crosshead speed of 5 mm/min. Six specimens for each sample were tested and the tensile strength and tensile modulus were expressed as [19]:

$$\text{Tensile strength (MPa)} = \frac{P}{bh} \quad (1)$$

$$\text{Tensile modulus (MPa)} = \frac{\sigma}{\epsilon} \quad (2)$$

The flexural test was performed according to ASTM D790 using the Universal Testing Machine at a constant rate of 2 mm/min. Six test specimens with span to thickness ratio of 16 ($L/h = 16$) were used. The flexural strength and flexural modulus were calculated using the following equations [19].

$$\text{Flexural strength} = \frac{3PL}{2bh^2} \quad (3)$$

$$\text{Flexural modulus} = \frac{mL^3}{4bh^3} \quad (4)$$

where P = maximum load applied on test specimen (N), L = support span (mm), b = width of specimen tested (mm), m = slope of tangent to the initial straight line portion of load deflection curve (N/mm), ϵ = strain and σ = stress.

The impact test was performed according to ASTM D256 standard using the impact testing machine. The test method determines the charpy impact strength of the polymer composites. Charpy type test in which the specimen is held as a cantilever beam (usually vertical) and is broken by a blow delivered at a fixed distance from the edge of the specimen clamp was adopted. Eight specimens for each sample having a size of 63.5 mm by 10 mm and

4 mm thickness were prepared and tested. The V notch was made at a distance of 31.75 mm from the top at a depth of 2.0 mm. Impact strength was calculated by [20]:

$$\text{Impact strength} = \frac{J}{A} \quad (\text{kJ/m}^2) \quad (5)$$

where J = Energy absorbed (kJ), A = Area of cross section of the specimen below the notch (m^2).

Results and discussion

Fig. 1 showed SEM/EDS of BPAP, from the SEM it was observed that the BPA particles are roundish with some angular in shape and a small amount of particles are longitudinal in shape. The BPAP surface morphology plays a vital role in the case of composite materials. External surface features of particles such as contours, defects and damage and surface layer were not observed in the SEM. The particles surface layers play an important role in wettability and surface tension [20].

The EDS spectra of BPAP (Fig. 1) show that the particles exhibited spectra containing carbon, oxygen and a small amount of silicon, potassium, calcium, phosphorous, magnesium and aluminium. The relative atomic percent of the atoms were obtained from the peak area and corrected with an appropriate sensitivity factor. The BPAP showed a higher proportion of carbon atom. The higher proportion of carbon in the particles can be attributed to the presence of carbon in the BPAP.

Morphological analyses of the composites using SEM clearly showed a difference in the morphology of the RLDPE matrix and the composites (Figs. 2–5). The microstructure showed that when the BPAP was added to the RLDPE matrix, morphological change in the microstructure occurred. The microstructure of the RLDPE matrix revealed chains of amorphous structure (Fig. 2).

The morphology of bean pod ash particle composite shows a smooth spherical surface having more surface area for interaction. There is good dispersion of BPAP in the polymer matrix. Particles-matrix interface plays an important role in composite properties. A strong particles-matrix interface bond is critical for high mechanical properties of composites [19]. Pulling out of the particles from the matrix, delamination between the particles and the polymer matrix was not observed in the study. All the composites contain polymer fibrils attached to the BPAP an indication of improved adhesion between the phases. The RLDPE-BPAP based composite

particles pull-out as well as holes resulting from particles delamination were not observed. Observations of improved adhesion are consistent with the mechanical performance improvement reported below. As the BPAP was increased beyond 20 wt% there was some agglomeration, sedimentation and cluster of the particles (see Fig. 5).

Density is one of the most important physical properties of a material. The density of reinforced composites is presented in Fig. 6. The results revealed that the presence of bean pod ash particulates slightly decreased the density of the polymer composites.

The decrease in density can be related to the fact that the BPAP is light in weight and occupies a substantial amount of space. Hence, it can be said that with an increase of percentage of BPAP the material becomes light in weight. For example, the density of the reinforced RLDPE particle composites decreased from 0.95 g/cm^3 at 0 wt% BPAP addition to 0.76 g/cm^3 at 30 wt% BPAP. Hence, composites with lower weight can be produced with BPAP. This work is in line with the earlier works carried out by Abdullah [20] and Patricio et al. [21].

In Fig. 7, it can be seen that % of water absorption slightly increased with increasing wt% BPAP. Composites with higher BPAP loading showed more water absorption. This is due to the higher content of filler loading in the composites that absorbed more water. As the filler loading increased, there is the difficulty of achieving a homogeneous dispersion of filler which resulted in agglomeration filler. The agglomeration of the filler in composites increased the water absorption of the composites. The water absorption in case of BPAP is minimum compared to other agro based composite material [1,4,18]. This may be attributed to lower void spaces in composites arising due to fair bonding between particles and matrix. Water absorption is an important property to select appropriate materials for outdoor application. The water absorption results obtained in this work are within the recommended standard for general purpose applications [12,14].

The mechanical properties of particulate-polymer composites depend strongly on the particle-matrix interface adhesion, stress-strain behaviour of the filler or the matrix, and volume of particle loading. The results of hardness values are showed in Fig. 8. It can be seen that with increase in wt% BPAP in the matrix the hardness values of the composites increased.

The increments in hardness values were attributed to an increase in the percentage of the hard BPAP in the RLDPE matrix. Also the differences in coefficient of thermal expansion (CTE)

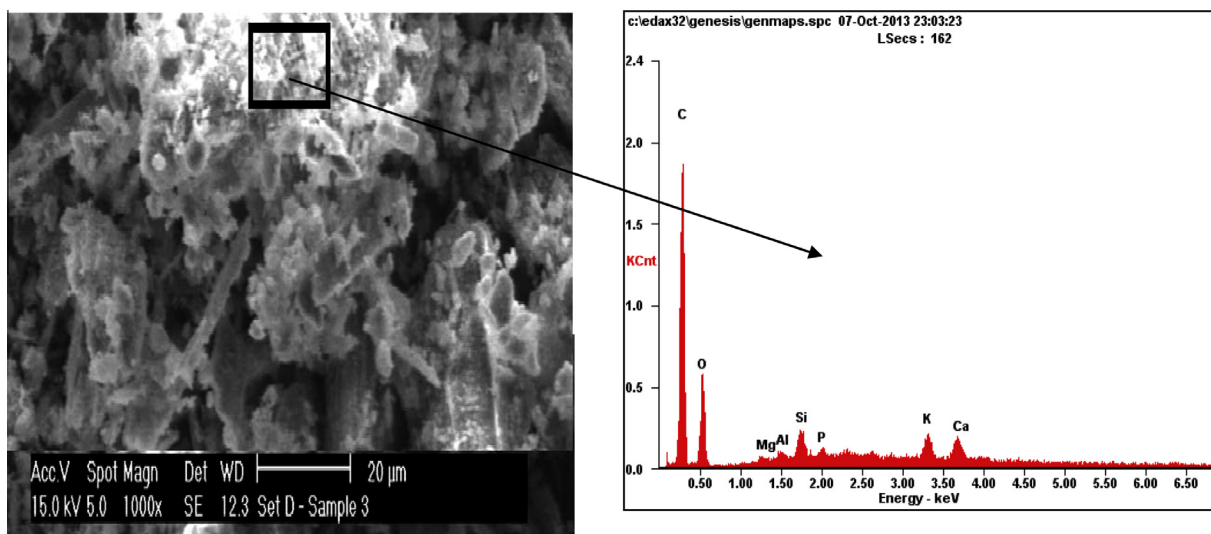


Fig. 1. SEM/EDS of the bean pod ash particles.

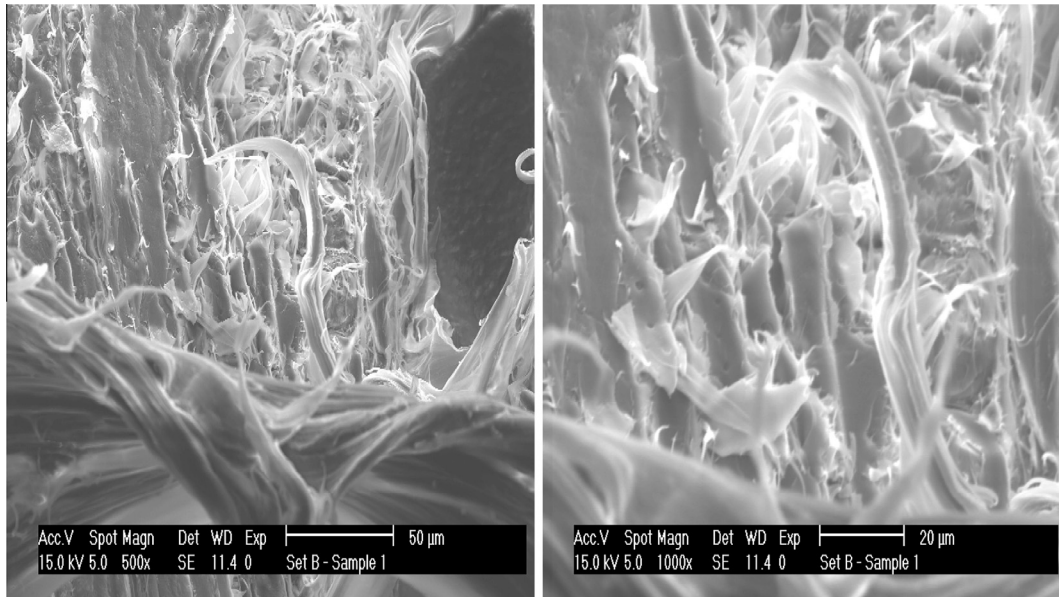


Fig. 2. SEM of the microstructure of the RLDPE matrix.

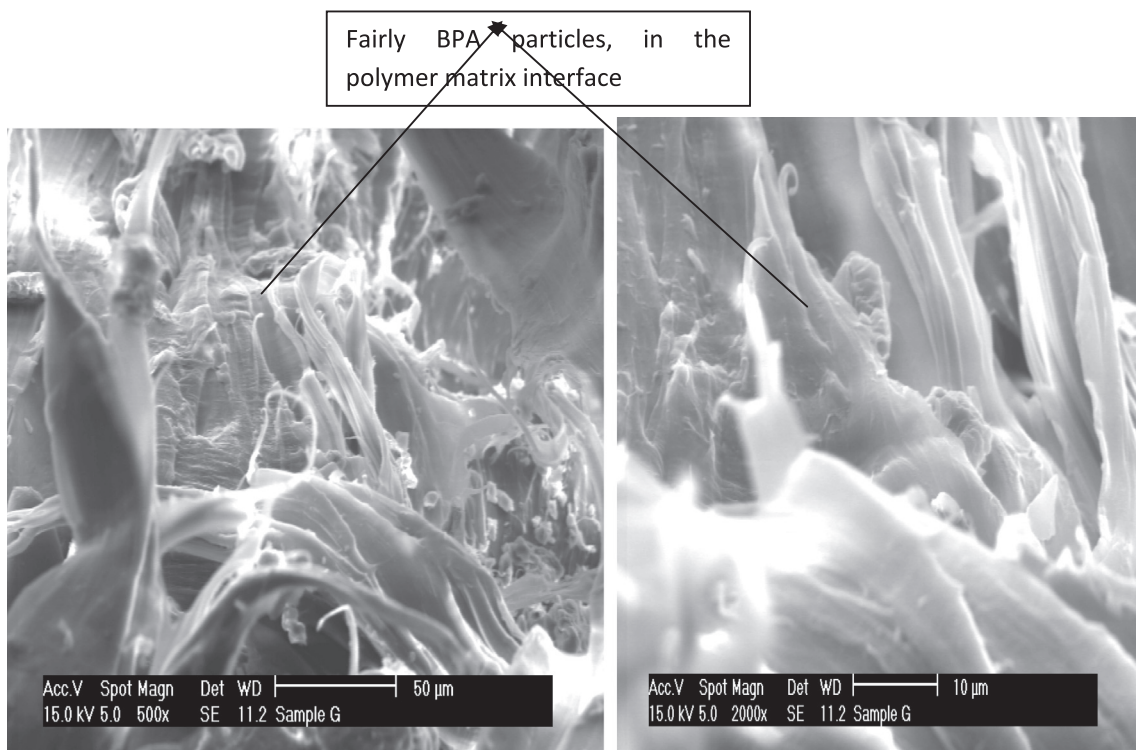


Fig. 3. SEM of the microstructure of the RLDPE with 10 wt% BPAp.

between the BPAp and RLDPE matrix resulted in elastic and plastic incompatibility between the matrix and the particles [18]. For example the hardness values of 3.5HRB and 8.9HRB were obtained for the RLDPE matrix and 30 wt% BPAp respectively. In comparison with the unreinforced RLDPE matrix, a substantial improvement in hardness values was obtained in the reinforced polymer matrix. This is in line with the earlier researches of [8,12].

The tensile stress–strain curves for unfilled RLDPE matrix and composite materials containing 5, 10, 15, 20, 25 and 30 wt% of BPAp are shown in Fig. 9. A remarkable difference can be seen in the stress–strain behaviour, due to addition of BPAp in the RLDPE

matrix. The RLDPE matrix has a large area under the stress–strain curve than the composite materials.

Figs. 10–13 show the variation of tensile modulus, flexural modulus, tensile and flexural strengths with wt% BPAp.

There is a significant increment in both the tensile modulus and flexural modulus as the BPAp loading increased. The increase of tensile modulus from 256 N/mm² for the RLDPE matrix to 789 N/mm² at 30 wt% BPAp and flexural modulus from 1.2 KN/mm² for the RLDPE matrix to 6.8 KN/mm² at 30 wt% BPAp may be attributed to the fair distribution of BPAp in the RLDPE matrix which efficiently hinders chain movement during deformation leading

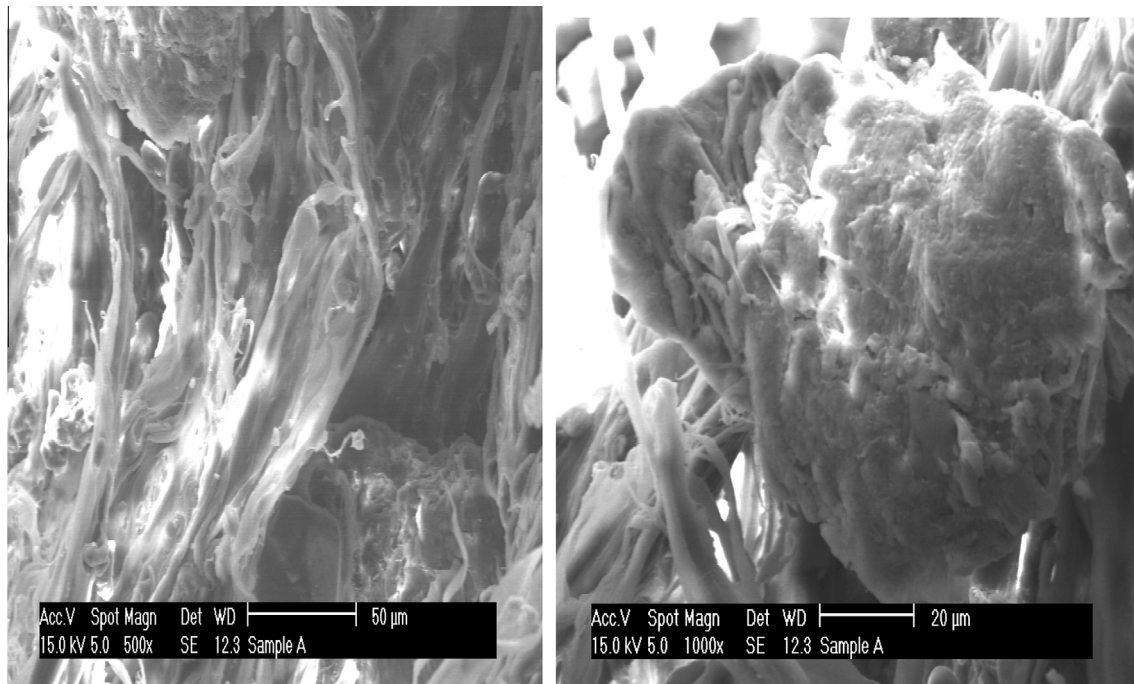


Fig. 4. SEM of the microstructure of the RLDPE with 20 wt% BPAP.

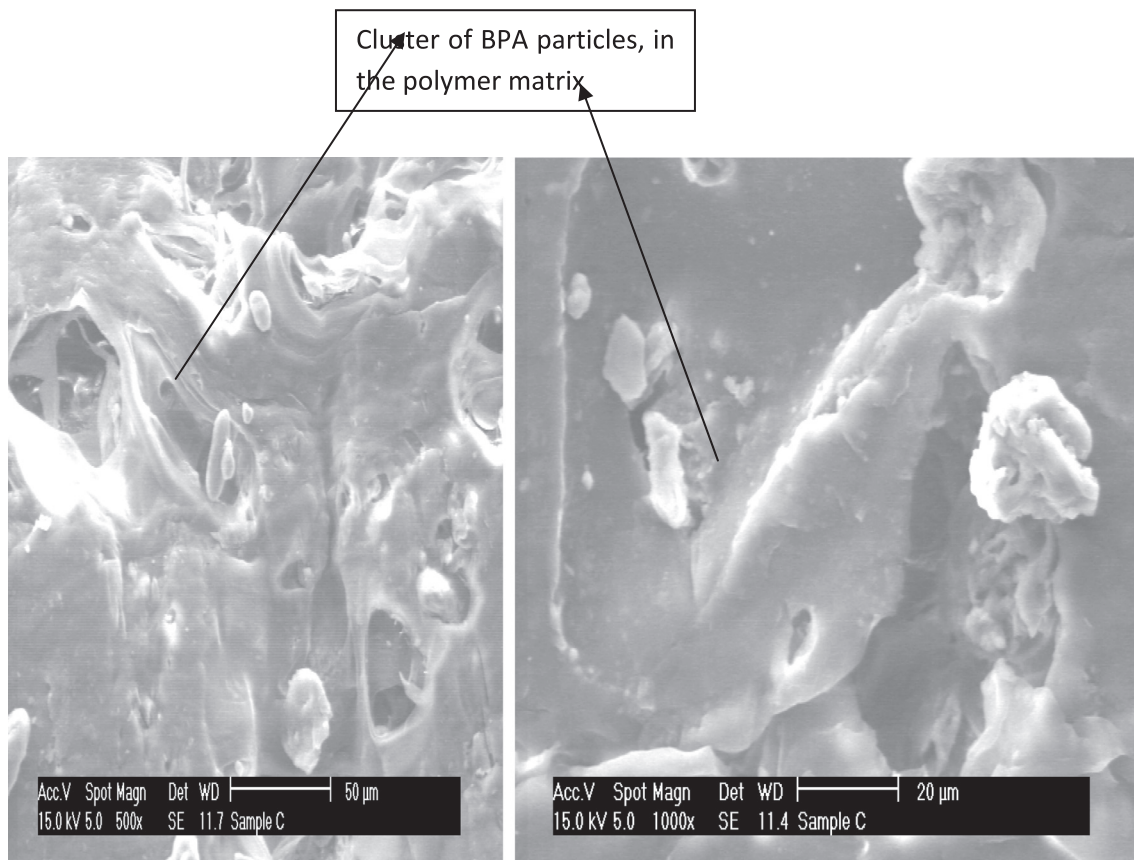


Fig. 5. SEM of the microstructure of the RLDPE with 30 wt% BPAP.

to high particle orientation. This mechanism will increase the stiffness of the composites as well as the modulus.

The increase in modulus of the BPAP-filled composites indicates an increase in the rigidity of RLDPE related to the restriction of

mobility in the RLDPE matrix due to the presence of BPAP. The modulus of these composites increased with increasing BPAP loading. This suggests stress transfer across the polymer–particles interface. BPAP in the matrix prevented movement in the area

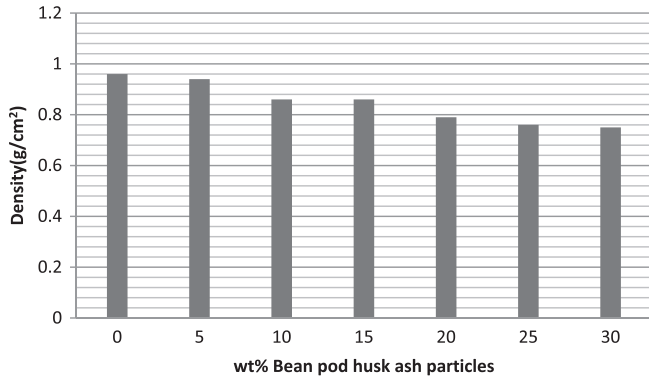


Fig. 6. Variation of density with wt% of bean pod ash particles.

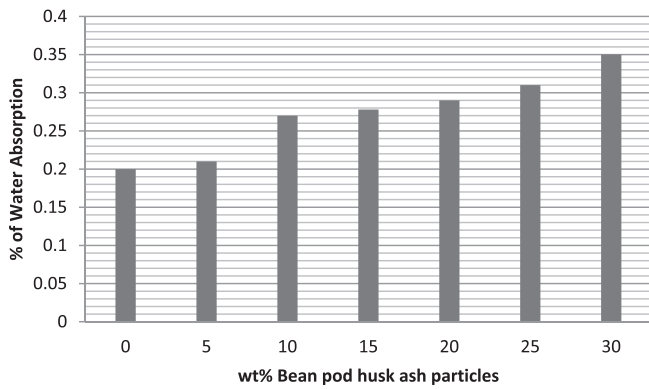


Fig. 7. Variation of % of water absorption with wt% of bean pod ash particles.

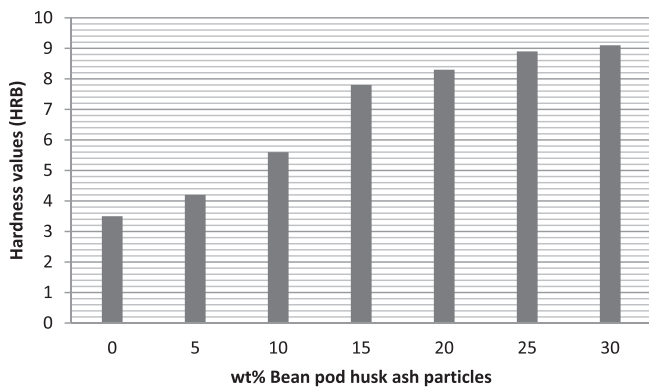


Fig. 8. Variation of hardness values with wt% of bean pod ash particles.

around each particle, contributing to an overall increase in the modulus. The high modulus values also support the use of the developed composites in general purpose applications [14].

The tensile strength increases from 8.2 N/mm² for the RLDPE matrix to a maximum of 20.1 N/mm² at 20 wt% BPAP and a flexural strength from 23.4 N/mm² for the RLDPE matrix to a maximum of 39.0 N/mm² at 20 wt% BPAP (Figs. 12 and 13). The increment may be due to the platy structure of the BPAP filler providing good reinforcement [15,18].

The high values of strength observed in this work may be due to the fair distribution of the BPAP in the RLDPE matrix resulting in strong particle-RLDPE matrix interaction. The particles dispersion improved the particle-RLDPE matrix interaction and consequently increases the ability of the BPAP to restrain gross deformation of

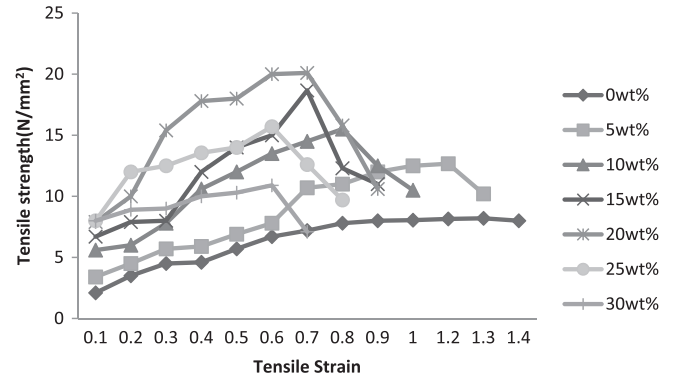


Fig. 9. Variation of tensile stress–strain curves with wt% of bean pod ash particles.

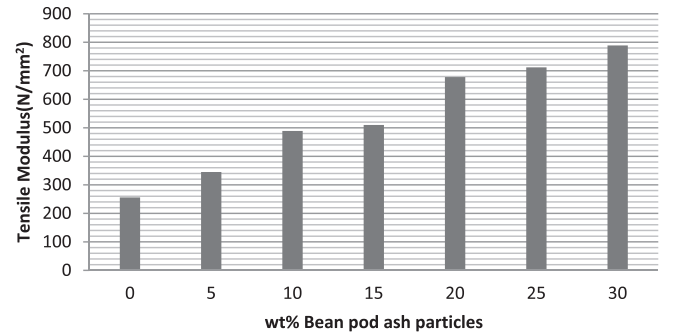


Fig. 10. Variation of tensile modulus with wt% of bean pod ash particles.

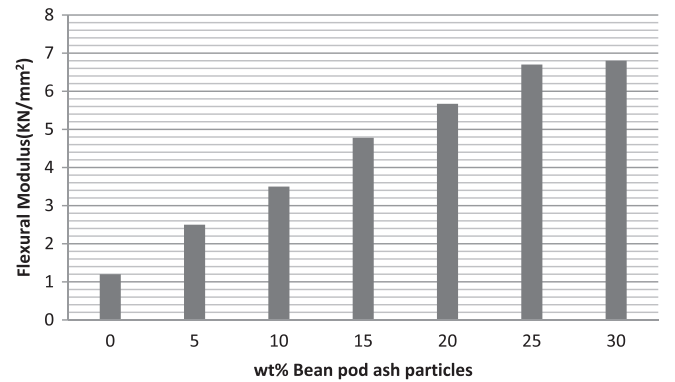


Fig. 11. Variation of flexural modulus with wt% of bean pod ash particles.

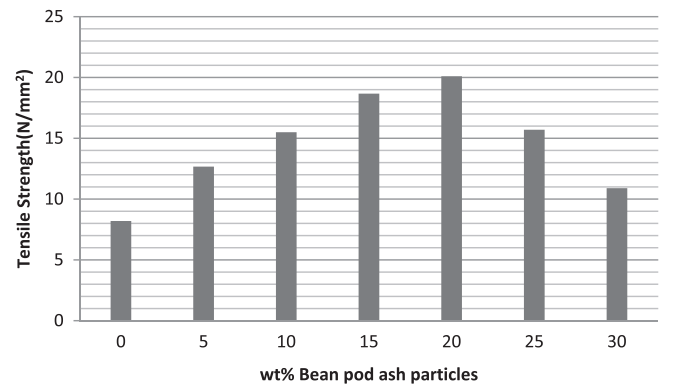


Fig. 12. Variation of tensile strength with wt% of bean pod ash particles.

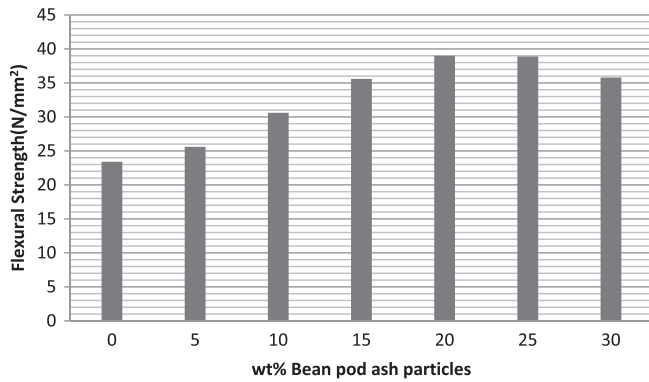


Fig. 13. Variation of flexural strength with wt% of bean pod ash particles.

the RLDPE matrix. The decrease in both the tensile and flexural strengths as the BPAp particles increased beyond 20wt% is due to the interference of particles in the mobility or deformability of the matrix. This interference was created through the physical interaction and immobilisation of the polymer matrix in the presence of mechanical restraints, thereby reducing the strength [9,10].

At a high filler loading, the composite will not be able to withstand greater loads, the formation of the microfiller (agglomeration) is found due to the difficulties in achieving a homogeneous dispersion of the filler at high filler loading and BPAp agglomerate and remain confined in the polymer matrix (see Fig. 5). At the particles/matrix boundary region, stress concentrates around the reinforcement particle. Consequently, the particles-matrix interaction weakens up thus, leading to debonding at the boundary region at higher BPAp loading. These observations are in agreement with the low strength values reported for these materials at higher BPAp loading.

The impact energy of a composite is influenced by many factors, including the toughness properties of the reinforcement, the nature of interfacial region and frictional work involved in pulling out the particles from the matrix. The nature of the interface region is of extreme importance in determining the toughness of the composite. The Charpy impact test is a standardized high strain-rate test which determines the amount of energy absorbed by a material during fracture. This absorbed energy is a measure of a given material's toughness and acts as a tool to study brittle-ductile transition. Fig. 14 showed the effect of filler loading on the impact energy of RLDPE and RLDPE-BPAp filled composites. It can be seen that the impact energy for the composites slightly decreases with increasing filler loading. Increased filler loading in the RLDPE matrix resulted in the stiffening and hardening of the composite. This reduced its resilience and toughness, and led to lower impact energy.

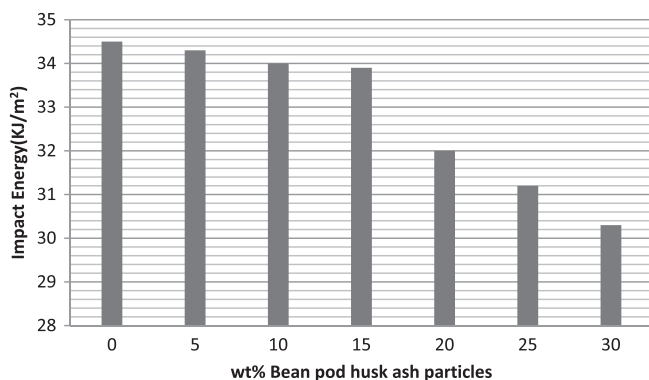


Fig. 14. Variation of impact energy with wt% of bean pod ash particles.

However, the decrease in impact energy for an increase in filler content, may be attributed to the weak interfacial interaction between the filler and matrix material for higher filler content, similar behaviour of the composite specimens was also observed by Joseph et al. [1] and Godwin et al. [3]. As the loading of BPA particles increases, the ability of the composites to absorb impact energy decreases since there is lower ratio of the RLDPE matrix to particles. However, the results obtained are within the standard level for bio-composites for general purpose applications [14].

Conclusions

In this present work, physio-mechanical properties of RLDPE bio-composites reinforced with bean pod ash particles have been investigated. From the results and discussion presented, the following conclusions are made;

1. This work showed that successful production of RLDPE and the bean pod ash particle composite by compounding and compressive moulding was realizable.
2. The fair distribution of the bean pod ash particles in the microstructure of the polymer composites is the major factor responsible for the improvement in the mechanical properties.
3. The bean pod ash particles added to the RLDPE polymer increased the percentage of water absorption and improved its rigidity, modulus and hardness values of the composites.
4. The tensile and flexural strengths increased to a maximum of 20.1 and 39.0 N/mm² at 20 wt% BPAp respectively.
5. Based on the results obtained in this study, it is recommended that the composites can be used in the production of indoor and outdoor applications.

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