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Biochar Filled Polypropylene Composites with Excellent Properties: Towards Maximizing the Utilization of Agricultural Wastes

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Abstract

The high cost and processing challenges associated with inorganic fillers have triggered the quest for alternative and less expensive filler materials from agricultural resources such as waste coir shells. The as-prepared coir shells were carbonized in an electrothermal oven at 200°C for 2 h and later pulverized and sieved into four particles sizes of 63µm, 150µm, 300µm, and 425µm. Furthermore, these carbonized fillers were used as reinforcing fillers in polypropylene matrix at varying filler contents of 0 – 40 wt. %. Composite sheets of PP/CSB were prepared by melt blending of polypropylene and the filler in an injection moulding machine. Our investigations showed that the addition of fillers was found to improve the yield strength, tensile strength, and tensile modulus of polypropylene as these tensile properties increased with increase in filler content. The elongation at break and modulus of resilience of the composites were observed to be inversely related to the filler content. Also, the investigation revealed that the specific gravity, flame propagation rate, water absorption and solvent uptake by PP/CSB composites were found to increase with increase in the filler content. Thus, CSB addition increased the specific gravity but with reduction in the flame, moisture, and solvent resistances.

1. Introduction

Fillers are added to virgin polymers to modify as to achieve theintended properties and to reduced cost. In the past, polymer-based composites were mainly reinforced with fiberglass or filled with minerals. Due to high density, non-degradability and insufficient recyclability of the fiberglass reinforced composites, composites reinforced with botanical fibres are gaining ground for a wide range of technical and lightweight applications [1-3]. Again, greenhouse gases are generated with composites obtained from fossil-based fibres while plant-based composites could reduce this challenge [4, 5]. Furthermore, the high cost associated with the compounding of mineral-based composites and the possible need to expand the use of different fillers in the plastic industry has led to the continued search for filler material that is cheap, renewable and biodegradable. No doubt, advances in technology have made composites obtained from the combination of polymers and natural fillers suitable alternatives to conventional ones, especially for lightweight applications.

Interestingly, fillers from agricultural resources exhibit some excellent properties compared to mineral fillers such as talc, calcium carbonate, kaolin, and mica. They have high specific strength to weight ratio, low density, inexpensive, present a minimal health hazard, less abrasive to processing machinery, renewable, biodegradable and environmentally friendly [6-9]. Also, unconventional natural fibres such as agro-wastes (e.g., wheat and rice straws, coir shells, etc.) and grasses (e.g., miscanthus, switch grass, and bamboo) find application inbiocomposites [10].

However, the hydroxyl groups present on the surfaces of the plant-based fillers tend to affect the bonding strength at the interface with any polymer matrix. These hydroxyl groups are likely to hinder the wetting of the filler surfaces by the matrix [11-13] and are expected to show poor mechanical properties in polymer composites owing to weak interfacial adhesion. Some sort of treatments have been developed and employed to tackle the biofiller drawbacks and improve compatibility with the matrix [10]. Such treatments include (but not limited to) chemical (e.g., silane maleic anhydride), mechanical (e.g., cutting, carding), physical (thermal, plasma, irradiation), and combinations of chemical, mechanical, or biological techniques. The improvement of interfacial adhesion between the matrix and filler via filler surface modification and the use of coupling agents have been reported [14-18].

Coir (Coconut) shells are agricultural by-products that are mostly considered to be hardwood. The lignocelluloses present in these shells have been reported to be tougher than wood; thus their disposal is not only costly but also cause environmental problems [19]. The coir shell powder, coir shell char and coir shell activated carbon are the three main products obtained from the coir shell. Coir shell char is obtained by the burning of fully matured coir shells in a limited supply of air so to reduce burning away to ash but produces only carbonized products — coir shell char which is an essential raw material for the manufacture ofactivated carbon. The activated carbon formed has certain advantages as the raw material and can adsorb certain molecular species [20].

Carbon-based fillers have been widely employed as reinforcements in the polymer matrix composites due to excellent mechanical, thermal and electrical properties [21-24]. Biochar, also known as biocarbon, has seen as a new sustainable material for several applications. It is not limited to filler and reinforcement uses for biocomposites [25-27]; biochar is also useful for the development of next-generation functional carbon materials for potential applications in energy storage and filtration devices [28-30].

The thermochemical conversion of biomass in the absence or limited supply of oxygen generates liquid bio-oil, solid biochar, and syngas. The biochar is an amorphous carbon-rich material with tunable properties such as chemical structure, porosity, size, and intrinsic modulus. The use of biochar natural fillers to improve the properties of polymer composites has also been studied [31-34]. The durability of polypropylene (PP) composites has been reported to be enhanced by the incorporation of biochar (carbon) filler into the matrix [35-37].

In the present study, the coir shell biochar (CSB) were used as reinforcing fillers to reinforce polypropylene matrix. The incorporation of the CSB into the PP matrix gave composites with improved properties. Therefore, the carbonization of the coir shells is apt since this treatment attempted to reconcile the seeming inadequacies of low moisture resistance and less compatibility observed in natural filler/polymer matrix composites. The approach further reduces the need for compatibilizers, without sacrificing the useful properties and biodegradability of the composite material. To the best of our knowledge, no work has been done using CSB to fill PP matrix through an injection molding process.

1. Experimental

1.1.Materials

The virgin polypropylene with a melt flow index of 25g/10 min at 230°C and density of 0.9g/cm^{3}produced by Indorama Eleme Petrochemical Company Limited, Nigeria was used as the matrix. Coir shells used as fillers were obtained from local farmers and traders within Owerri Metropolis, Imo State, Nigeria. The shells were washed with water, sun-dried, carbonized, pulverized, and sieved to particle sizes. Figure 1 shows the dehusked kernel-free coir shells and coir shell biochar (CSB).

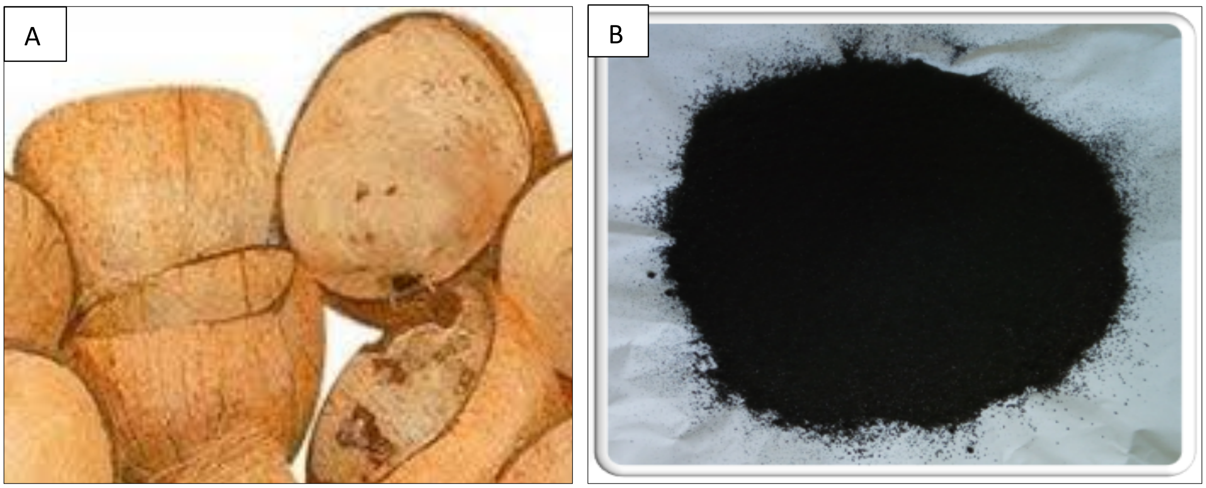

Figure 1. (A) Dehusked Coir shell without the kernels, (B) Coir Shell Biochar

1.2.Preparation of PP/CSB Composites

The cleaned and dried coir shells were carbonized in an electrothermal oven (Model MCEEg DHG) at 200°C for 2 h. The resulting char was later ground to fine particles and sieved into four particle sizes namely; 63µm, 150µm, 300µm, and 425µm. The polypropylene granules were melted and homogenized with the filler in an injection-molding machine at varying CSB content (0 – 40 %) maintained at temperature range of 195° C and 260° C. The resulting composites were extruded as sheets of dimensions 150 x 150 x 3 mm^3. The injection molding operation was carried out at an injection pressure of about ~10 MPa. The nozzle and hopper temperatures maintained at 260℃ and 195℃ respectively. The preparation flow process of composite manufacture is shown in Figure 2.

1.1.Tensile Test

The tensile test was carried out to determine the yield strength, tensile strength, elongation at break, tensile modulus, and modulus of resilience of the PP/CSB composites. The tensile testing of the prepared composites was carried out using a Tensile Testing Kit (MonsantoTensometer, Model

TEK/2/11140) at a uniform rate of 26 mm/minute according to ASTM D638 and ISO 527-1. Three test experiments of each sample were conducted, and average value reported. Three samples were tested for these properties, and the mean values used.

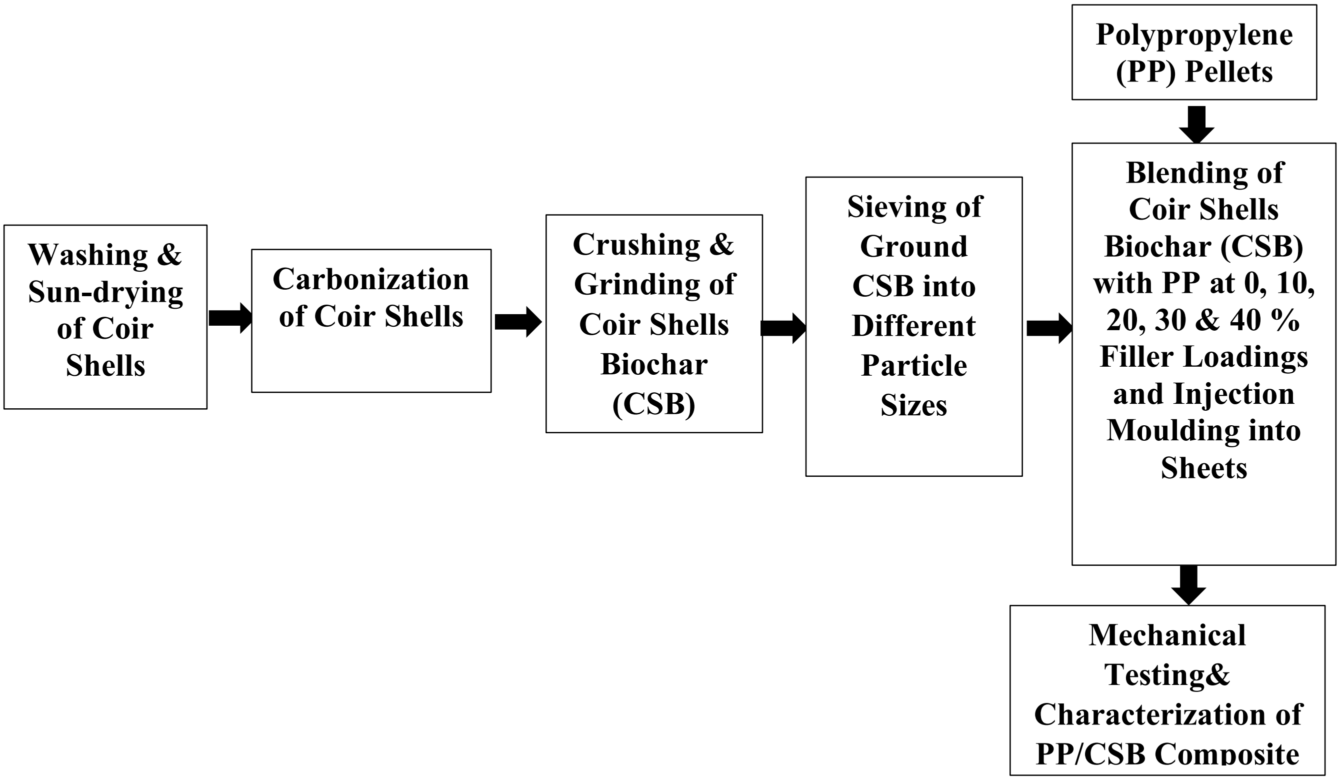

Figure 2. Flow Process for the Preparation of the Polypropylene Composites Sheets

1.2.Specific Gravity Test

The hydrostatic weighing method was adopted to determine the specific gravity or relative density of the PP/CSB composite samples. The samples were observed to float in water; thus, a less dense liquid, gasoline was used for this test in place of water. The specimen was suspended in air and in gasoline using a thin string attached to a sensitive spring balance. The sample was initially weighed in air to obtain the weight in air (W_a) and then, immersed in gasoline to get the weight in gasoline (W_g). The specific gravity with respect to gasoline was calculated as follows:

$$\mathbf{SG_g} = \frac{\mathbf{W_a}}{\mathbf{W_a - W_g}} \qquad 1$$

The actual specific gravity of the sample (i.e., relative to water) was calculated by multiplying equation (1) with the specific gravity of gasoline (i.e. 0.74) as shown in equation (2):

$$\mathbf{SG} = \left(\frac{W_a}{\mathbf{W_a - W_g}}\right) \times \mathbf{0.74} \qquad 2$$

1.3.Flame Propagation Test

The flame propagation test was done according to ASTM D 4804. Briefly, a sample specimen of dimensions 150 x 10 x 3 mm^3 with a mark along the length of the sample at 90 mm was clamped horizontally in a retort stand with the marked length ($D_l = 90\ mm$) protruding out of the clamp. The free end of the sample was ignited using a lighter and the initial ignition time (t_1) is recorded. The sample was left to burn to the 90 mm mark before stopping the stopwatch, and the time was noted as the finishing time (t_2). The relative rate of burning for the different samples was determined using the expression:

$$\textbf{Flame Propagation Rate, FPR (mm/s)} = \frac{\mathbf{D_l}}{\mathbf{T_2 - T_1}} \qquad 3$$

In equation (3), D_l = propagation distance of flame measured in mm; and $T_2 - T_1$= flame propagation time measured in seconds. Three experiments were conducted, and the average value was taken.

1.4.Water Absorption Test

Specimens of dimensions 30 x 30 x 3 mm^3 were cut from the prepared composite sheets and weighed to obtain dry weight (W_d). The samples were then placed in a 250 ml beaker containing 150 ml of water, covered with filter paper and put in a safe place. The samples were left in the glass of water for 24 h at room temperature (28°C). At the expiration of 24 h, the samples were carefully removed from the glasses using forceps. The water adhering on the surface of the samples was carefully removed using filter paper; care was taken during the process not to remove the water absorbed by the sample and then, weighed to obtain the wet weight (W_w). The water absorption for each sample after 24 h was calculated using the equation:

$$\textbf{Water absorption, WA}\ (\%) = \left(\frac{\boldsymbol{W_w - W_d}}{\boldsymbol{W_d}}\right) \times \mathbf{100} \qquad 4$$

where $W_w - W_d$ = weight of water sopped up by the specimen after 24 h.

1.5.Solvent Uptake Test

The solvent uptake test was carried out to determine the solvent resistance of the PP/CSB composites using gasoline and some selected aromatic solvents (benzene, toluene, and xylene). Cut samples (30 x 30 x 3 mm^3) of the composites were weighed and put into four glass reagent bottles with a stopper containing about 30 ml of each solvent. The samples were wholly immersed in the solvents and were left at room temperature for 24 h. The samples were removed from the bottles after the expiration of time. The solvents adhering to the surface of the samples were carefully removed using filter papers and then weighed. The molar percentage solvent uptake per gram of the composite was calculated using the formula:

$$\textbf{Molar solvent Uptake, MSU (mol.\%/g)} = \left(\frac{\boldsymbol{W_w - W_d}}{\boldsymbol{W_d}}\right) \times \mathbf{100} = \left[\frac{\left(\frac{\boldsymbol{m_t - m_d}}{\boldsymbol{M_s}}\right)}{\boldsymbol{m_d}}\right] \times \boldsymbol{100} \qquad 5$$

Where m_d = mass of polymer sample in g; m_t = mass of polymer sample in g after soaking in solvent for 24 h; M_s = molar mass of solvent in g/mole.

2. Results and Discussion

2.1. Tensile Tests

2.1.1. Yield Strength

Figure 3 illustrates the effect of filler content on the yield strength of the PP/CSB composites. It is evident from the figure that the yield strength increased from 34.5 MPa for the neat PP (0% filler content) and maintained a steady increase with filler content for all particle sizes studied. The composites filled with 63µm particles had the highest yield strengths for all filler contents, whereas composites filled with 425µm filler particle size gave the lowest values of yield strength for all filler contents. The reason for this observation could be that finer particle sizes are better dispersed within the matrix, thus, forming better interfacial adhesion than the coarser particles. The addition of coir (coconut) shell powder or coir fibre to polymer matrix with a resultant increase in strength has been reported[38-41].

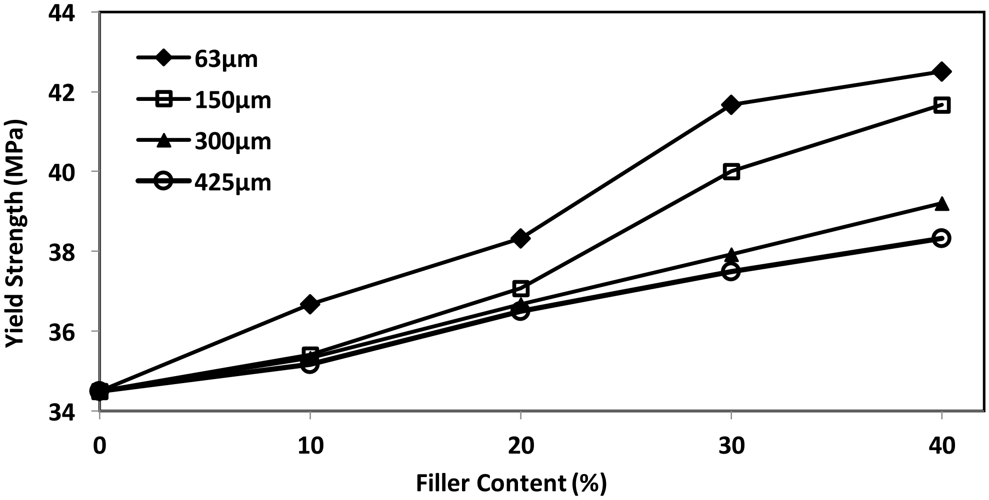

Figure 3. Filler Content Vs Yield Strength of PP/CSB Composites

2.1.2. Tensile Strength

The result of the CSB content on the tensile strength of the polypropylene filled composites is shown in Figure 4. It is clearly seen that the tensile strength of the composites increased with an increase in the filler content for all particle sizes. The figure indicated that are all the filled composites exhibited higher tensile strength than the virgin polypropylene (35.42 MPa). Again, it is noticed that the smaller the particle size of the filler, the higher the tensile strength of the polypropylene composites at any filler content considered. Hence, the 63µm particle size had the optimum tensile strength at all filler loadings. Finer particle sizes are believed to give better dispersion with improved filler-matrix interaction, and such could be the main factor responsible for the observed trend. Investigations to improve the strength of polymer composites using fillers from coir shell have been reported [40-43].

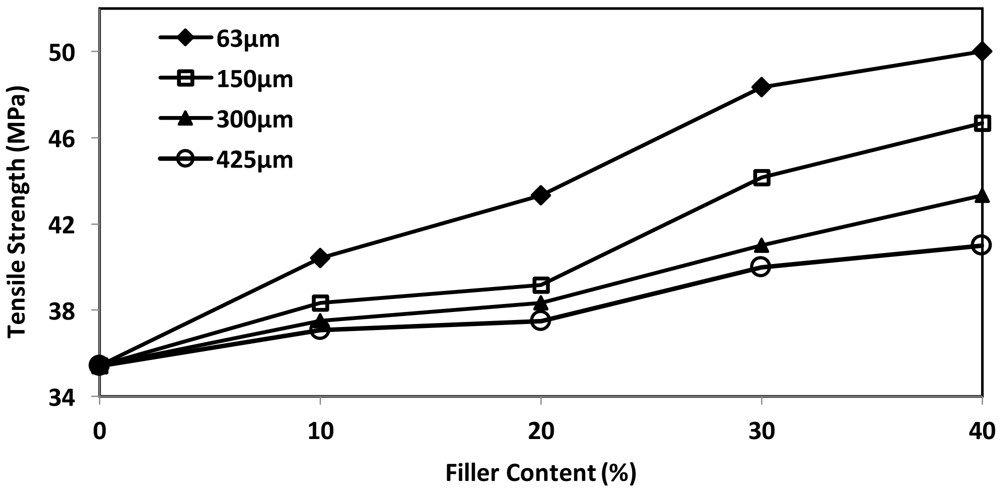

Figure 4. Filler Content Vs Tensile Strength of PP/CSB Composites

2.1.3. Elongation at Break

The effect of filler content on the elongation at break of the PP/CSB composites is presented in Figure 5.It is correct to say that the elongation at break of the filled polypropylene composites decreases with an increase in filler content for any given filler particle size. The elongation at break showed also that the 63µm particle size had the lowest values for all filler contents. The values demonstrate that fine particles and high filler content result in better dispersion and high particle-matrix adhesion, making it difficult for the 'rigid structure' to relax and allow for elongation as the material is being stretched. However, large particle sizes and low filler content would lead to less dispersion and weak particle-matrix adhesion, which result in a more flexible structure that can permit elongation upon stretching.

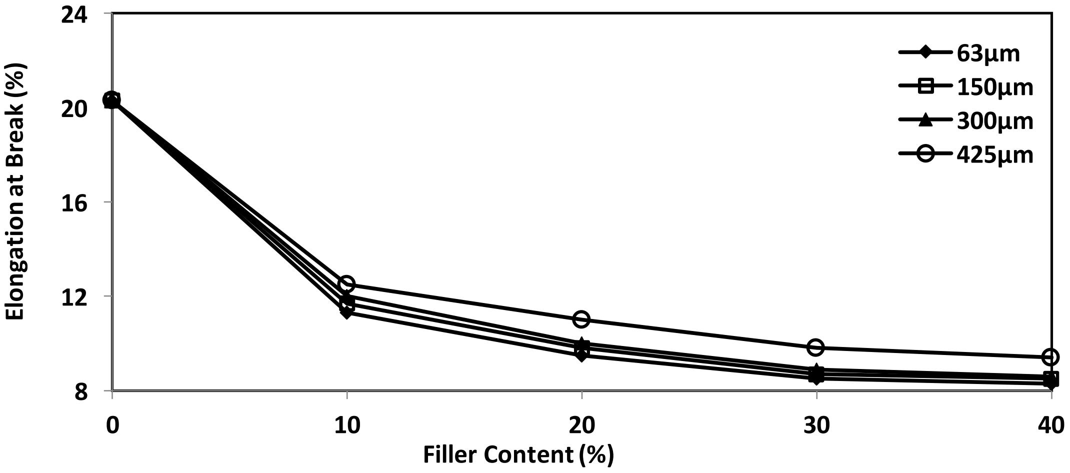

Figure 5. Filler Content Vs Elongation at Break of PP/CSB Composites

The neat PP gave the highest elongation (20.3%) but with a steady reduction as the filler is introduced into the polymer. Moreover, the result indicates that the coir biochar particles added some brittleness in the PP, and this effect increased as strength and hardness increased. A similar response had been reported [44, 45]. The results suggest that CSB filled polypropylene at high filler content would not be suitable for any application where high flexibility is of the essence.

2.1.4. Tensile Modulus

Figure 6 presents the effect of filler content on the tensile modulus of PP/CSB composites. It is evident from the figure that the tensile moduli of the composites were higher than that of the neat polypropylene. The increase is in agreement with the findings of Poulose, et al. [34], Li, et al. [44], Nan, et al. [46]. Furthermore, the tensile modulus was found to increase with an increase in the filler content for all particle sizes studied. The composites filled with 63µm filler particles had the highest stiffness. Since stiffness is a function of both strength and elongation within the elastic limit (i.e., the ratio of stress to strain), it infers that tiny filler particles and high filler content produced higher strengths and lower elongations which gave rise to the high stiffness recorded at small filler particle sizes and high filler contents. The high dispersion of small particles within the PP matrix with improved particle-matrix interaction at high filler contents elucidates this observation, which highlights the fact that the addition of CSB filler to polymer matrix enhanced the stiffness of the resulting composites.

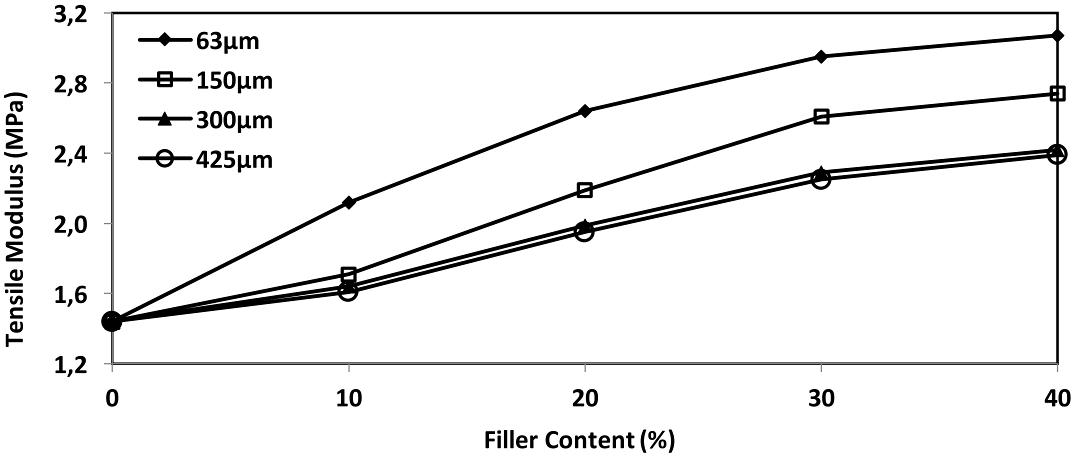

Figure 6. Filler Content Vs Tensile Modulus of PP/CSB Composites

2.1.5. Modulus of Resilience

The plot of modulus of resilience or elastic toughness of the composites versus filler content at different filler particle sizes is displayed in Figure 7. It is seen that the neat PP had the highest observed resilience of 0.414 MJ/m^3 or 0.000414 J/mm^3 implying that the resilience decreased with increase in filler content for all particle sizes. The 63µm particle size produced the lowest values of resilience for all filler contents investigated. Since resilience or toughness within the elastic region depends on the product of strength and strain (elongation that is not expressed in percentage), it means that the strengthening obtained with small filler particle sizes and high filler contents was more than compensated for by the little elongation produced, resulting in the reduction of resilience or elastic toughness. Thus, there is a decrease in the energy required to stretch the composites to the elastic limit as filler content increases and as filler particle size decreases.

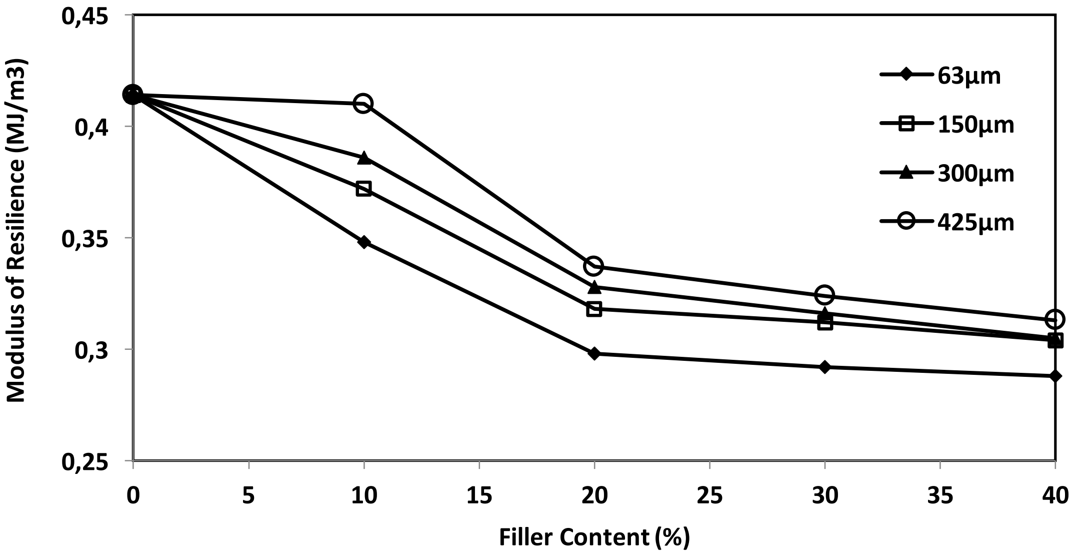

Figure 7. Filler Content Vs Modulus of Resilience of PP/CSB Composites

2.2. Specific Gravity

Figure 8 shows the effect of filler content on the specific gravity of the PP/CSB composites. From the figure, it is observed that the specific gravity increased with filler content for all the composites and higher than neat PP at any given filler particle size considered. The higher specific gravity reported for filled PP could be due to the higher density of carbon (2.267 g/cm^3) compared to 0.905g/cm^3 for PP. Thus, the density of carbonized coir biochar (CSB) used as filler is expected to be greater than that of polypropylene. This behaviour suggests that incorporating CSB into PP matrix could tend to raise the relative density of the PP composites. The increase in the specific gravity or relative density as the filler particle size decreased could be attributed to the improved distribution of the filler particles in the polymer matrix and the improved packing efficiency of these particles within the matrix.

2.3.Flame Propagation Rate

The flame propagation rate (FPR) is a measure at which flame moves along a specified length of a material with time. The FPR of PP/CSB composites is displayed in Figure 9. It was observed that at any given filler particle size, the flame propagation rate increased with increase in filler content and with a decrease in particle size. The enhancement of flame propagation in lieu of flame-retardant property by the filler could be attributed to the combustibility of carbon and the coir shell used. Thus, the presence of coir shell-derived-carbon enhanced the combustibility of PP, a material that is already combustible. At high filler content, more carbon particles present and higher flame propagation rate. Furthermore, the 63µm filler particles increased flame propagation rate because more carbon particles could be accommodated within the matrix due to better dispersion and packing efficiency.

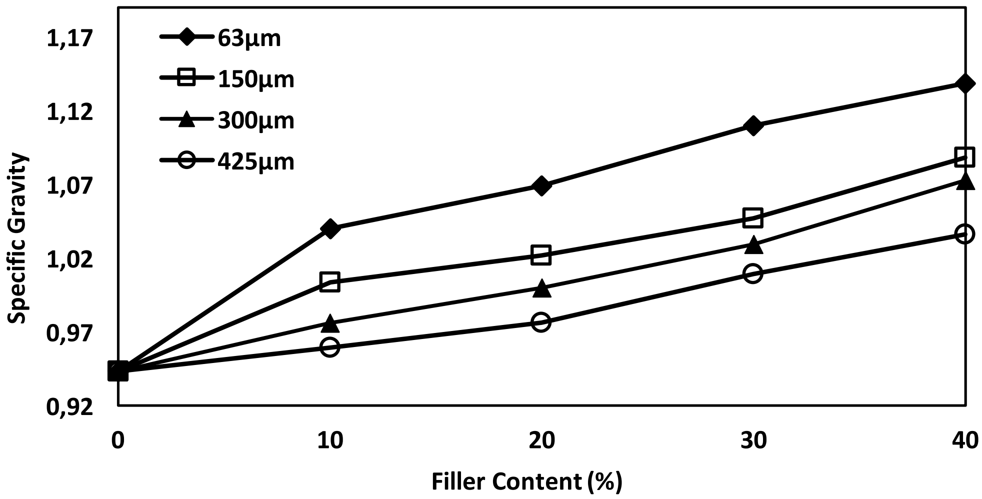

Figure 8. Filler Content Vs Specific Gravity of PP/CSB Composites

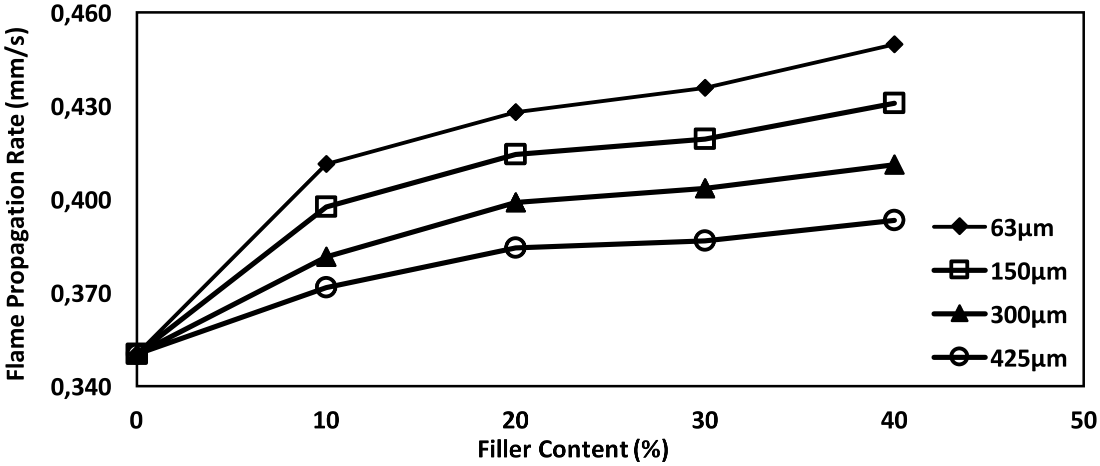

Figure 9. Filler Content Vs Flame Propagation Rate of PP/CSB Composites

2.4.Water Absorption

Water absorption is vital for biodegradability of polymer composites as pure synthetic matrices are inert to water molecules. This demonstrates the critical cause of their extended period of degradation. It is expected that these composites degrade after their useful lives. Hence, microorganisms are attracted to the natural fibre filled composites because of the ability to absorb water which aids in the degradation. The water absorption capacity of the PP/CSB composites after 24 h exposure to water is presented in Figure 10. The water absorption increases as the filler content increases for all particle sizes.

Furthermore, it is observed that water absorption increased with decrease in particle size and even more significant at high filler content. The figure indicates the neat PP matrix had the least water absorption behaviour. The behaviour could be linked to the high surface area – to – volume ratio present in tiny particles. High filler content suggests incorporating more particles in the PP matrix which ultimately increases the total surface area for water absorption. The enhanced water absorption capacity of carbonized coir shell particles in this study could also be that the carbon acts as activated carbon, which has high adsorption tendency [20].

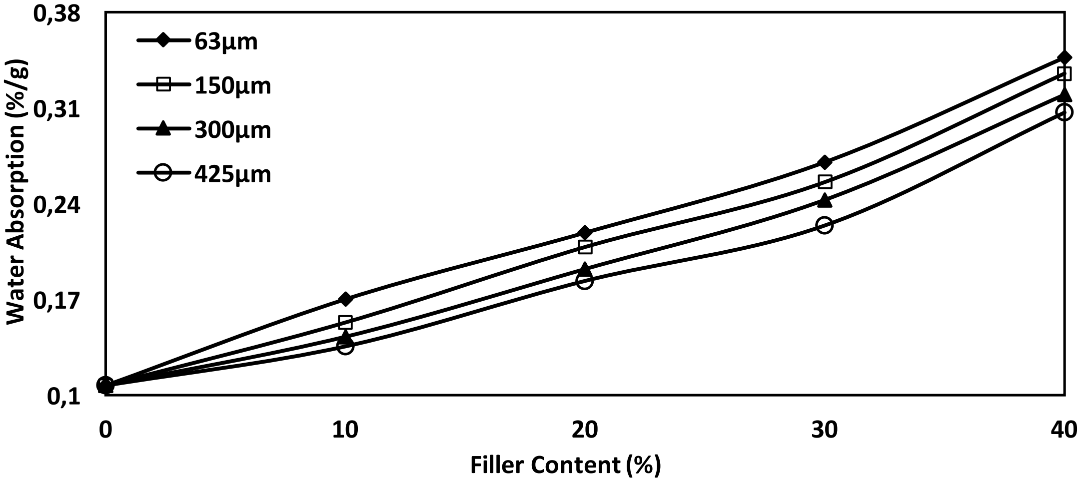

Figure 10. **Filler Content Vs Water Absorption of PP/CSB Composites**

2.5.Molar Solvent Uptake Behaviour

The molar solvent uptake of benzene, toluene, xylene, and gasoline plotted against filler content for all the PP/CSB composites filled 63, 150, 300, and 425µm filler particles are shown in Figures 11(a – d). It was observed that molar benzene sorption at all filler particle size studied was the highest, followed by toluene, xylene, and gasoline in that order. It is reasonable to say that this trend could be linked to the proximity of the solubility parameters of the solvents relative to that of polypropylene.Furthermore, the difference in solubility parameter is like a driving force for solvent sorption [47]. From the literature, polypropylene has a solubility parameter of 9.3 $(cal/cm^3)^{1/2}$, benzene (9.1) $(cal/cm^3)^{1/2}$, toluene (8.9) $(cal/cm^3)^{1/2}$, xylene (8.8) $(cal/cm^3)^{1/2}$ and gasoline (7.6) $(cal/cm^3)^{1/2}$. That is, the decreasing order of difference in solubility parameters between each solvent and PP is 1.7 (gasoline, octane), 0.5 (xylene), 0.4 (toluene), and 0.2 (benzene). Therefore, it is expected that benzene should be absorbed most by polypropylene, or polypropylene should swell most in benzene. Another possible factor could be the molar masses of the solvents. The effect of molar mass of solvent as observed is such that as the molar mass of solvent decreases, the molar solvent uptake increases. Thus, benzene with a molar mass of 78.11 g/mole, had the highest molar solvent uptake by polypropylene, followed by toluene (92.14), xylene (106.16), and gasoline (114.23) in that order. A possible explanation could be that the composites experienced a reduction in interaction between the solvent molecules and the matrices as the solvent molecular size increases, particularly in the case of the filled matrices.

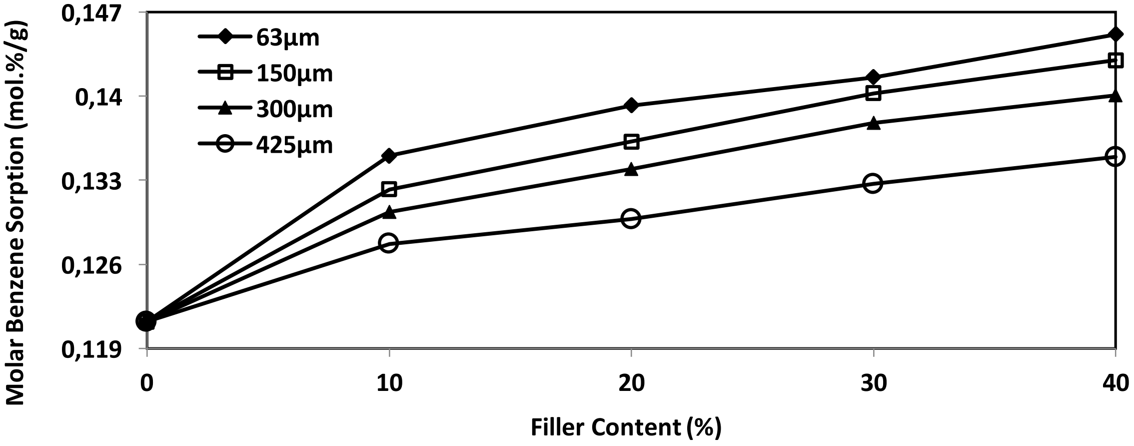

Figure 11a. Filler Content Vs Molar Benzene Sorption of PP/CSB Composites

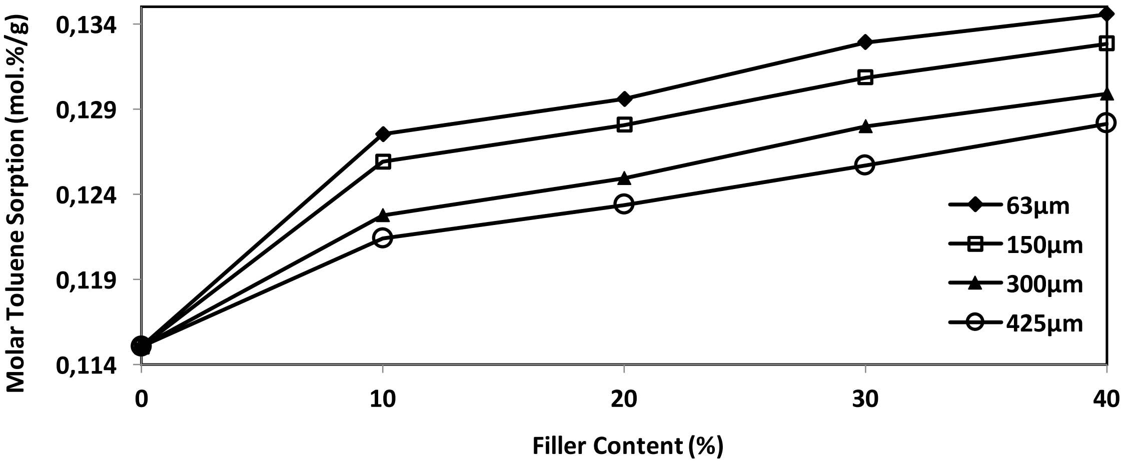

Figure 11b. Filler Content Vs Molar Toluene Sorption of PP/CSB Composites

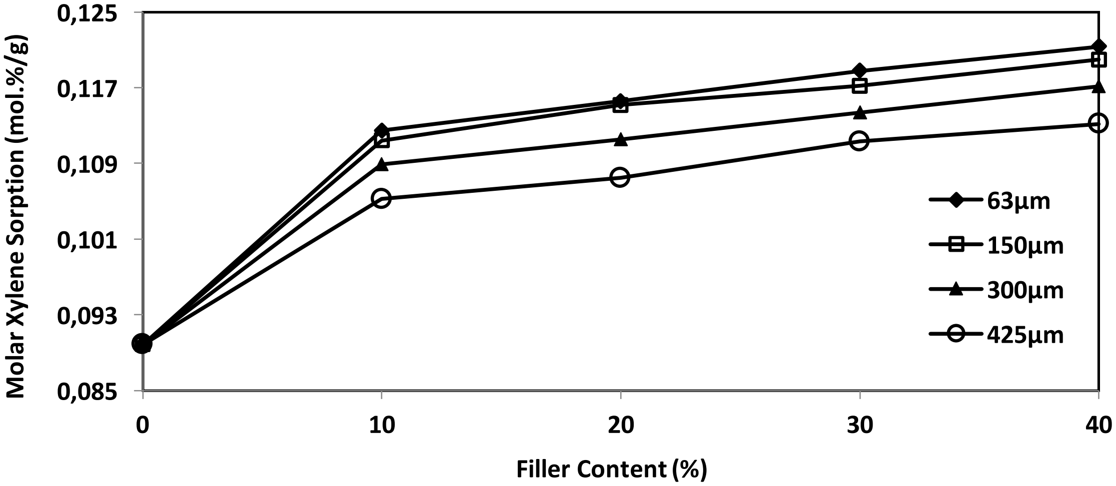

Figure 11c. Filler Content Vs Molar Xylene Sorption of PP/CSB Composites

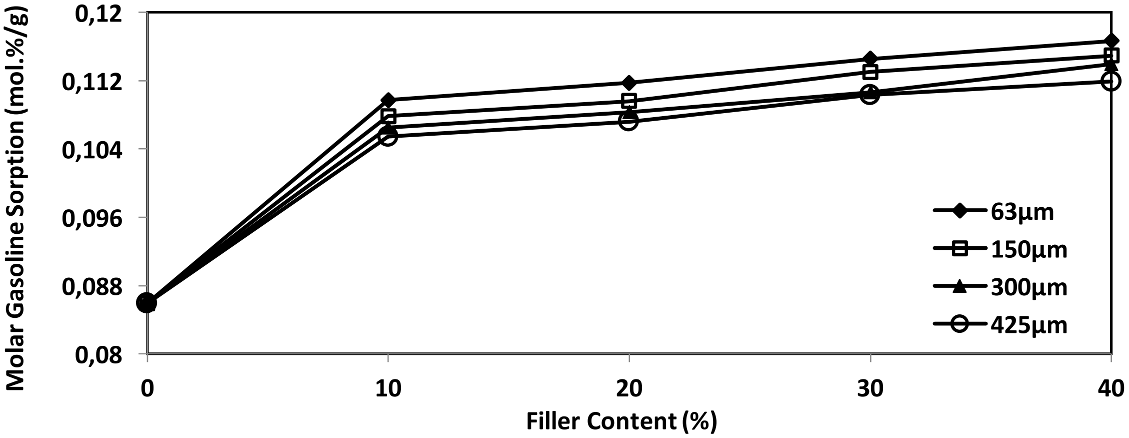

Figure 11d. **Filler Content Vs Molar Gasoline Sorption of PP/CSB Composites**

Conclusion

The tensile and physical properties of polypropylene (PP) filled with coir shell biochar (CSB) were investigated at filler contents of 0, 10, 20, 30 and 40% using filler particles sizes of 63, 150, 300 and 425 μm. The results of tensile properties showed that the addition of CSB filler to the PP matrix enhanced the yield strength, tensile strength, and tensile modulus when compared to the control (neat PP). These properties were found to increase with an increase in filler content at all particle sizes studied. However, elongation at break and modulus of resilience were found to decrease with increase in filler content, thus, justifying the strengthening and hardening that accompany the corresponding loss of flexibility and toughness.The physical properties evaluated indicated that the use of CSB as filler for PP matrix improved the specific gravity, flame propagation rate, water absorption, and solvent uptake in comparison with the virgin PP matrix. These properties were found to increase with increase in filler content at all particle sizes studied. However, the filler imparted some weight to the PP but could not have the needed flame retardant and solvent resistant properties. Therefore, the CSB filled PP can be used mostly for indoor applications, especially where weight and rigidity are essential but not fire or solvent resistance such as plastic seats, cabinets, storage boxes, wastebaskets, etc.

Conflict of Interest: The authors declare that there are no conflicts of interest.

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