Mechanical Properties of waste plastic composites reinforced with untreated and alkali treated Coconut Shell Powder as filler.

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Abstract

This study investigated the mechanical properties of a polymer matrix composite from waste plastic (sachet water pouches, SWP) reinforced with untreated and alkali-treated coconut shell powder (CSP). The NaOH concentrations were 4, 6, and 8 % wt. /v; treatment time was 120 minutes and temperature was 70°C. The composites were fabricated using the compression moulding technique and subjected to mechanical testing. The alkali-treated SWP/CSP composites showed an increase in tensile strength and modulus of 19.18% and 20.65% respectively, relative to the untreated composites. Flexural strength and modulus improved by 39.15% and 69.13% respectively, while the impact strength increased by 40.05% when compared with the untreated counterparts. These improvements were attributed to better compatibility between the SWP matrix and alkali-treated CSP filler, confirming the effectiveness of the alkali treatment of lignocellulosic in the production of polymer matrix composites. On the other hand, elongation at break was observed to have decreased by 19.84% compared to composites made from untreated CSP. The untreated SWP/CSP composites recorded poor tensile and flexural properties in contrast to those from treated CSP due to poor interfacial bonding between the polymer matrix and untreated CSP. Overall, the study demonstrated that the use of CSP in polymer composites is a sustainable and environmentally friendly approach to waste management.

Keywords: Alkali treatment, Coconut shell powder, Water sachet matrix, Composites, Mechanical properties.

1.0 Introduction

Polymer composites production involves the reinforcement of a polymer matrix with a filler or fiber. The reinforcement could be in form of fibers, wood sawdust, or particles while the matrix is a polymer. The filler is usually stiff and stronger than the continuous matrix phase and serves as the principal load-carrying member, while the matrix is more ductile and maintains the position and orientation of the reinforcement (Elanchezhian *et al.* 2018). Presently, the major concern in the entire universe is the issue of environmental protection, reuse, and recycling of resources, which had made researchers source novel materials to use as an alternative to man-made fiber-reinforced polymeric composites (Reddy *et al.* 20202). Natural lignocellulosic materials available in the environment include coconut shells, wood sawdust, rice husk, oil palm fiber, coconut coir, bamboo, sugar cane bagasse, peanut shell powder, and many others. They are selected in place of synthetic fibers like nylon, fiberglass, asbestos, carbon, Kevlar and aramid fibers due to their eco-friendliness, low cost, natural wide availability, renewability, light weight and biodegradability characteristics (Munshi *et al.* 2020). Lignocellulosic material is composed of cellulose, lignin and hemicellulose which are the three major constituents (Khan *et al.* 2019). The mechanical properties of polymer composites reinforced with lignocellulosic materials are influenced by factors like, the filler % content, type of treatment used and the composites processing technique (Rajes *et al.* 2017). To improve the interfacial adhesion, there is need to subject natural fiber/filler to either chemical, physical or grafting of short chain molecules to its surface using coupling agents (Chokshi *et al.* 2020). Alkali treatment is one of the most common chemical treatment methods in which NaOH, KOH or LiOH are used as the treatment agent (Samaei *et al.* 2020). The treatment removes non-cellulosic components and surface impurities. It increases the access to active OH groups at the surface of lignocellulosics, improves the filler-matrix interfacial adhesion and compatibility between the filler and the polymer matrix that significantly improves the mechanical properties of the resultant composites (Oushabi *et al.* 2017).

Although, a lot of studies has been carried out on the alkali treatment of lignocellulosic materials (Rajesh & Pitchaimani 2017; Oushabi *et al.* 2017), there is scarcity of existing literature on the NaOH treatment of the abundant indigenous agro waste product like coconut shell powder (CSP) and its utilization in polymer composites production. Researchers have investigated the properties of polymer matric composites from different lignocellulosics. Islam *et al*, (2020), produced coir mat/polyester resin composites using coir mat as reinforcement and polyester resin as matrix. Coconut coir was loaded at 10, 20, 30, 40, 50 and 60% by weight. The composites were fabricated via hand lay-up method. Mechanical properties like tensile strength (TS), flexural strength (FS), tensile modulus (TM), flexural modulus (FM), elongation at break and impact strength (IS) were examined. The authors reported a maximum percentage increase in TS, FS, TM, FM and IS to be 44.44, 128, 17.96, 112.09 and 62.50% respectively for 30% coir content in the composites. They observed that the mechanical strength of the composites increased with increase in filler content up to 30% weight. Subsequent increase in filler content led to decrease in properties.

In another study, Gobikannan *et al* (2020), developed and characterized sisal fiber and wood dust-reinforced polymeric composites. The authors reported highest flexural strength of 7.5 N/mm^2 with 30 wt. % wood dust and 35 wt. % sisal fiber, while the lowest flexural strength of 3.5 N/mm^2 was found with 20 wt. % wood dust and 45 wt. % sisal fiber. The researchers reported that the observed reduction in the flexural strength was appreciable because of increase in the fiber content which resulted in poor wetting of the fiber by the matrix. From the literatures, the mechanical properties of various polymer composites have been studied. However, research on the mechanical behaviour of alkali treated coconut shell powder (CSP reinforced sachet water pouches (SWP) composites is rather rare. Agricultural waste-reinforced polymer matrix composites find applications in construction, building, automobile, household appliances, transportation, furniture, sports and electrical devices (Elanchezhian *et al.* 2018).

Despite the advantages of using lignocellulosic materials as fillers in composites formulation, there are a number of limitations confronting their incorporation into a matrix. This include high moisture absorption, swelling and shrinkage, agglomeration during processing, low durability and poor adhesion with polymer matrix (Barra & Gonzalez 2017; Raghu & Goud, 2020). Also, there is the challenge of the restriction of their processing temperature to 200°C because of the possibility of degradation at higher (Lafia-Araga *et al.* 2020). One of the problems in polymer matrix composites processing with natural fillers is the incompatibility between the fillers and polymer matrix due to the hydrophobic nature of the polymer matrix and hydrophilic character of the filler. This result in non-uniform dispersion and poor mechanical properties (Salleh *et al.* 2018). These challenges are due to the presence of OH^- groups in their chemical structures that accounts for their basic chemical and physical properties. Therefore, the incorporation of CSP as filler in composite fabrication is confronted with the problems of moisture absorption and poor interfacial bonding with a polymer matrix (Gobikannan *et al.* 2020). Hence, there is a need for alkali treatment of CSP. Alkali treatment improves the strength, reduces the moisture absorption capacity, and removes the amorphous components in CSP, thereby making it compatible as a filler in polymer matrix composites.

The use of plastics in developing nations like Nigeria is on the increase, hence the high generation of plastic waste. Plastic pollution is a global environmental problem because plastics are generally non-biodegradable and remain in the environment for a very long period of time (Barra & Gonzalez 2017). Nations are increasingly confronted with the problem of finding lasting solutions to the challenges of plastic pollution. Therefore, it is imperative to source an alternative way of converting waste like sachet water pouches to a useful product. (Oushabi *et al.* 2017; Samaei *et al.* 2020). Thus, this study investigates the mechanical properties of composites from SWP reinforced with untreated and alkali-treated CSP. Using alkali-treated CSP, a renewable agricultural waste as filler in SWP, a common plastic waste, will help alleviate the deleterious effect these wastes have on the environment. This research is aimed at producing SWP/CSP composites with appreciable mechanical properties that will convert these wastes to wealth as a sustainable and environmentally friendly approach to waste management.

2.0 Experimental

2.1 Materials

The Coconut shell was collected from the local coconut fruit traders in Kuje market in the Federal Capital Territory (FCT), Abuja, Nigeria. The sachet water pouches (SWP) was sourced from waste dump sites in the Kuje area council of FCT, Abuja Nigeria.

2.1.1 Preparation of CSP and SWP

The coconut shells were soaked in water for 1-2 hours to remove the fiber, washed with detergent to remove dirt and sand and rinsed severally in distilled water. It was sun-dried for three days and inside an oven at 105°C to constant weight. The dried coconut shell was crushed using mortar and pestle before being pulverized in a locally fabricated grinder and sieved to pass through a 250 µm sieve. The powder was stored in airtight glass bottles for further use. The sachet water pouches were washed with distilled water, sun-dried for 3 days, and placed in an oven at 65°C for 12 hours. It was crushed using a Thomas Wiley laboratory mill to obtain a mesh size of 250 µm and stored in plastic bags.

2.1.2 Modification of coconut shell powder (CSP)

The alkali treatment was carried out by soaking about 120 g of CSP in 1000 cm^3 of various concentrations of NaOH solutions (4, 6 and 8 w/v %) for 120 minutes duration and at a temperature of 70°C respectively. Treated samples were neutralized with 2% acetic acid and washed several times with deionized water to remove the remaining trace of NaOH. The samples were dried in the open air for 48 hours and in an oven at 105°C until constant weight and stored in plastic bags for compounding.

2.2 Physicochemical characterization of Coconut Shell Powder

The chemical compositions of untreated and treated CSP were determined using standard test methods. The moisture content was determined according to ASTM D-4442 (2016) standard. The extractive content was determined using ASTM D-1105 (2016). Acid-insoluble lignin was ascertained using ASTM D-1106 (2016) standard. The holocellulose (cellulose and hemicellulose) content was prepared according to ASTM D-1104-(2016) from which the cellulose content and hemicellulose content of CSP sample was estimated using the difference between the values of holocellulose residue. The ash content was determined using the ASTM-E-1755-(2016) standard procedure. All determinations were done in three replicates and the average value was recorded.

2.3 Composite manufacturing

Untreated and alkali-treated CSP and SWP were compounded at a weight percentage ratio of 40:60 in 100 g portions using a Brabender two roll mill (Germany) at the Nigeria Institute for Leather Science and Technology Zaria, Nigeria. The shredded sachet of water plastic was added while the roller was in a counter clockwise motion for a period of 10 minutes at a temperature of 170°C. Upon melting, the CSP was gently introduced manually at a speed of 100 rpm. Each compounded composite was subjected to compression moulding using a compression moulding machine, Moore model (Kaohsiung, Taiwan). A mould of dimension 150 x 150 x 3 mm was sprayed with a release agent before loading the sample. The upper platen was lowered so that the mould just closed. The composite was preheated for 2 min and compressed at a temperature of 170 °C and 3.4 MPa pressure for 4 min (Barra & Gonzalez 2017; Ejiogu *et al.* 2019). The mould was cooled for 20 min with water and the composites were removed. Tensile, flexural, and impact test specimens were cut from the moulded samples.

2.4 Measurement of Mechanical Properties

The tensile test was performed according to ASTM D-638 (2016), type IV using a universal testing machine (MONSANTO Tensometer; UK) with a load of 600 N, a gauge length of 40 mm at a crosshead speed of 5 mm/min. Five samples of each composite were tested and the mean value was recorded. The results obtained were used to calculate the tensile strength, tensile modulus and percentage elongation. The flexural test was done in a three-point bending mode using Monsanto tensometer Model 9875, UK, following ASTM D790-00 (2016) standard method. The sample support span (L) was 40 mm with maximum deflections of 10 mm. A load of 100 KN was applied at a temperature of 25±2°C, at a crosshead speed of 2 mm/min till fracture occurred or the maximum deflection was reached. A total of five samples from each composite were tested and the average readings were recorded.

ASTM E23 (2007) standard testing procedure was used in carrying out the impact properties in Charpy mode using an impact testing machine (Norwood instrument, model No: 412-07-0715269C). Samples were notched at the center of one edge to produce a single edge notch (SEN) impact test specimen. The notch angle was set at 45° at 2 mm depth and 0.25 mm radius along the base. The pendulum hammer of velocity 2.87 m/s was released downward from an initial height of 425 mm towards the specimen. For each sample, a minimum of five specimens were tested and the average result was recorded.

3.0 Results and Discussion

3.1 Chemical Composition

The percentage chemical composition of untreated and treated CSP are presented in Table 1.

The observed increase in cellulose content could be attributed to the partial removal of impurities and the non-cellulosic components like hemicellulose, pectin, and wax that are present in coconut shell powder surface (Ejiogu *et al.* 2019). Also, it could be due to the conversion of amorphous cellulose 1 to crystalline cellulose II will lead to higher crystallinity index values (Agunsoye *et al.* 2018; Rajeshkumar 2020). This shows the potentiality of utilizing coconut shell powder as filler in polymer matrix composites because the increase in cellulose content will enhance the mechanical strength of the resultant composite (Manimaran *et al.* 2018).

Table 1: Chemical composition of untreated and treated coconut shell powder (CSP)

Concentration (%)	Duration (min)	Temperature (°C)	Cellulose (%)	Ash (%)	Hemicellulose (%)	Lignin (%)	Moisture (%)	Extractives (%)
Untreated	-	-	46.70±0.10	2.50±0.03	14.30±0.20	26.50±0.10	5.80±0.05	4.20±0.20
4	120	70	69.20±0.10	3.10±0.30	7.40±0.40	15.10±0.30	2.30±0.10	1.10±0.60
6	120	70	73.00±0.41	3.50±0.08	6.10±0.33	12.10±0.12	2.40±0.20	1.20±0.06
8	120	70	73.80±0.42	3.10±0.03	6.20±0.31	14.10±0.22	1.70±0.01	1.20±0.02

Results are mean values, ± standard deviations where n=5

The hemicellulose content decreased from 14.30% in untreated CSP to 6.10% and was attributed to the dissolution of hemicellulose and lignin in an alkali solution. High hemicellulose reduces the thermal stability and increases the hydrophilic nature hence, the need for treatment of CSP to enhance its suitability for high-temperature applications in composites. The highest moisture content of 5.80% was recorded in the untreated sample which decreased to 1.70% in CSP soaked in 8% NaOH for 120 minutes at 70°C. The reduction in moisture content in CSP was attributed to the possibility of compression of the cell wall lumen present in the structure of CSP. The reduction could also be because of the fact that the –OH group present in crystalline cellulose is not easily accessible to water molecules. The implication of these observations is a reduction in the moisture absorption capacity of CSP that will lead to improvement in dimensional stability as well as mechanical properties of the resultant composite.

The extractive content decreased from 4.20% in untreated to 1.10% in CSP treated in 4% NaOH for 120 minutes at 70°C. This may improve interfacial bonding between the filler and polymer matrix. The Also, amount of ash increased from 2.50% in untreated CSP to 3.50% in sample treated in 6% NaOH for 120 minutes at 70°C. From the chemical composition analysis of CSP, it can be inferred that polymer matric composites manufactured with such an agricultural waste will give low moisture absorption capacity, better reinforcement and improved mechanical properties. The mechanical, thermal, moisture absorption, morphological, biodegradability and bonding properties of agrowaste products are usually dependent on the chemical composition of the natural plant based resources when employed as reinforcement in the manufacture of polymer composites (Kathirselvam *et al.* 2019).

3.2 Mechanical Properties

3.2.1Tensile Properties

The tensile properties of unfilled matrix, untreated SWP/CSP and alkali treated SWP/CSP composites are shown in Table 2.

Table 2: Tensile Properties of SWP/CSP Composites

S/No	Composites ID	Tensile strength (MPa)	Tensile Modulus (MPa)	Elongation at break (%)
1	Matrix	14.38 ±1.94	103.84 ±5.82	835.33 ±334.21
2	Untreated	7.82 ±0.19	123.25 ±5.71	9.18 ±0.48
3	4% NaOH/120 min/70°C	8.99 ±0.37	147.09 ±7.27	8.54 ±0.45
4	6% NaOH/120 min/70°C	8.43 ±0.34	137.32 ±6.92	9.47 ±0.19
5	8% NaOH/120 min/70°C	9.32 ±0.33	148.70 ±4.91	7.66 ±0.68

From Table 2, pure matrix had the highest tensile strength of 14.38 ±1.94 MPa while the untreated SWP/CSP composite had the lowest tensile strength of 7.82 ±0.19 MPa. This could be because of the high hemicellulose and lignin content that created a barrier against the formation of bond between strong OH group of hemicellulose and the matrix (Binoj, 2018). Also, the high OH groups in hemicellulose increases the moisture absorption capacity of untreated CSP. This is responsible for poor adhesion between the hydrophilic untreated CSP and the hydrophobic water sachet matrix hence, the reduction in tensile strength in untreated SWP/CSP composite. The tensile strength of alkali treated SWP/CSP composites had values that ranged from 8.43±0.34 MPa to 9.32±0.33 MPa. The tensile strength of 9.32±0.33 MPa corresponds to composite reinforced with CSP sample treated in 8% NaOH concentration for 120 minute at 70°C. The percentage increase in tensile strength for this sample was 19.18% compared with the untreated SWP/CSP composites. This increase could probably be because of increase in surface roughness and enhanced compatibility which lead to good dispersion of treated CSP within the SWP. Hence, alkali treatment is necessary for obtaining better tensile property (Binoj 2018; Zin *et al.* 2018). This result is similar to the findings of Samaei *et al.* (2020), who investigated the tensile strength of Gloriosa fiber and reported 29.58% increase in the tensile strength of optimally alkali treated Gloriosa fiber, which was attributed to reduction in moisture absorption, increase in crystal size and high percentage removal of amorphous components from the fiber.

From Table 2, the pure matrix had the lowest tensile modulus of 103.84±5.82 MPa which is an indication of its flexibility. The composite containing untreated CSP had a tensile modulus of 123.25±5.71 MPa. The tensile modulus of alkali treated SWP/CSP composites is higher and is in the range of 137.32 ±6.92 to 148.70±4.91 MPa. The composite made from CSP sample soaked in 8% NaOH concentration for 120 minutes at 70°C had tensile modulus of 148.70 ±4.91 MPa. This showed a percentage increase of about 20.65% when compared to the untreated CSP composites and about 41.22% increase in contrast to the unreinforced matrix. The percentage increase in tensile modulus observed in this study is high compared to the work of Islam *et al.* (2020), who reported 17.96% increase. The increase could be attributed to the removal of hemicellulose, lignin and impurities that lead to better interlocking of CSP within the matrix (Lafia-Araga *et al.* 2020). This result agrees with the finding of Shanmugasundaram *et al.* (2018), who concluded that the removal of non-cellulosic constituents by alkali treatment increases the tensile modulus of natural fibers and attributed it to increase in close packing structure of the alkali-treated natural fiber.

Table 2 presents the elongation at break of untreated and alkali-treated SWP/CSP composites. The SWP matrix had the highest elongation at a break value of 835.33 ±334.21%. This is an indication of the ductility of the polymer matrix. The untreated SWP/CSP composite had 9.18±0.48%. This value decreased to 7.66 ±0.68 in composites loaded with alkali-treated CSP. The observed decrease in elongation at the break of untreated and alkali-treated SWP/CSP composites may be due to the proper dispersion of CSP into the plastic matrix, which provided stiffness and suppressed the deformation of the matrix. This result is similar to the finding of Prabhakar *et al.* (2019), who investigated the impact of modification with alkali (NaOH) on Peanut Shell Powder (PSP) and reported that the treatment offered a substantial enhancement in filler/matrix adhesion along with overall elongation at break properties of the composites.

3.1.2 Flexural properties

The flexural strength and modulus of the matrix, untreated and alkali-treated SWP/CSP composites are shown in Table 3.

Table 3: Flexural properties of untreated and alkali-treated SWP/CSP Composites

S/No	Sample ID	Flexural strength (MPa)	Flexural modulus (MPa)	Impact strength (KJ/m^2)
1	Matrix	18.54 ±0.92	197.90 ±28.35	2860 ±106.77
2	Untreated	12.44 ±0.62	165.62 ±4.64	1518 ±18.33
3	4% NaOH/120 min/70°C	16.98 ±0.49	259.71 ±26.61	1914 ±19.60
4	6% NaOH/120 min/70°C	15.35 ±0.47	224.44 ±7.37	1840 ±20.00
5	8% NaOH/120 min/70°C	17.31 ±0.97	278.04 ± 14.64	2126 ±33.23

The unfilled matrix had a maximum flexural strength of 18.54±0.92 MPa compared to the filled CSP composites. The untreated SWP/CSP composites exhibited a flexural strength of 12.44 ±0.62 MPa. The flexural strength of alkali-treated SWP/CSP composites is about 17 MPa.

The composite formed with CSP sample soaked in 8% NaOH concentration, for 120 minutes at 70°C temperature had high flexural strength of 17.31±0.97 MPa. The percentage increase in flexural strength at this treatment condition was 39.15% and 49.04% when compared to the untreated SWP/CSP composite and the unfilled matrix respectively. A higher flexural strength was achieved in this research relative to the study of Gobikannan *et al.* (2020), who reported a flexural strength of 7.5 N/mm^2 with 30 wt. % wood dust and 35 wt. % sisal fiber. The observed increase in flexural strength could be due to a measure of compatibility between treated CSP and SWP that imparted a resistance to the applied force in flexure (Vinayagamoorthy *et al.* 2020). The efficient removal of non-cellulosic amorphous components from the cell wall surface of CSP that lead to better interaction with the matrix could be another reason for enhanced flexural strength in alkali treated composites (Lafia-Araga *et al.* 2020). In a similar study Vinayagamoorthy *et al.* (2020), reported that the immersion of CSP in 8% NaOH concentration for 90 minutes enhanced the tensile and flexural strength of the fabricated epoxy composites than the untreated counterpart.

The flexural modulus of alkali treated and untreated SWP/CSP composites is presented in Table 3. From Table 3, the matrix had flexural modulus of 197.90 ±28.35 MPa. The untreated SWP/CSP composite had flexural modulus of 165.62 ±4.64 MPa, while the alkali treated SWP/CSP composites had increased flexural modulus in the range of 224.44 ±7.37 to 278.04 ±14.64 MPa. This resulted in 69.13% increase which could be because of better interfacial bonding between the filler and the matrix. In a similar study Ahlawat, *et al*. (2019), observed that at 8% alkali concentration, walnut shell particles (WSP) reinforced polyester resin recorded an increase in the flexural modulus of the composites which was attributed to increase in uniform distribution of polyester resin in WSP. However, the percentage increase in flexural modulus in this study is less than those of Islam *et al.* (2020), who reported about 112.09 % increase. This could be attributed to differences in filler volume % content used in the two studies.

3.1.3 Impact strength

The mean impact strength for the various SWP/CSP composites are reported in Table 3. From the Table 3, the unfilled matrix had the highest impact strength of 2860 ±106.77 KJ/m^2. The alkali treated CSP reinforced composites had reduction compared to the matrix though an enhancement in impact strength in the range of 1840 ±20.00 to 2126 ±33.23 KJ/ m^2 when compared to the untreated SWP/CSP composite that had impact strength of 1518 ±18.33 KJ/ m^2. The SWP/CSP composite reinforced with CSP sample treated in 8% NaOH soaked for 120 minutes at 70°C, recorded the second highest impact strength of 2126 ±33.23 KJ/m^2. This gave a percentage increase in impact strength of about 40.05% compared to the untreated counterpart. This could be attributed to improvement in interfacial adhesion between the treated CSP and used SWP matrix. This is an indication that alkali treated SWP/CSP composites have the capacity to absorb energy from the applied force. The result obtained in this study is higher compared to that of Manimaran *et al.* (2018), who investigated the impact strength of alkali treated red banana peduncle fiber reinforced (RBPF) and red banana wood flour (RBWFs) reinforced composites. The authors reported a decrease in impact strength of 4.6 KJ/m^2 value in alkali treated fiber, which was attributed to improper distribution and alignment of fibers. In addition, the impact properties of the composites studied (1518 to 2126 kJ/m^2) are appreciably higher than the Nigeria Industrial Standards (NIS) requirement for plastic tables and chairs (28.7 J/m^2) (NIS 970-2017; NIS 799-2020).

Conclusion

The untreated and alkali treated CSP samples were used as the reinforcing fillers for used SW plastic and different SWP/CSP composites were produced. The results of the mechanical testing showed that alkali treated SWP/CSP composites had increase in tensile strength, tensile modulus, flexural strength, flexural modulus, impact strength and decrease in elongation at break, when compared to the untreated SWP/CSP counterpart. The improvement was attributed to better CSP-matrix interface and compatibility after alkali treatment via the removal of impurities/non-cellulosic material, increase in cellulose content, reduction in hydrophilicity and improved surface roughness. The study demonstrated that the alkali treatment of CSP at appropriate concentration, time and temperature could result in CSP that can improve the mechanical properties of polymer composites. This presents a sustainable and an environmentally friendly approach to waste management.

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