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Sustainable and Light Weight Cellulose-Based Hybrid Reinforced Epoxy Composites for Automotive Application

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ABSTRACT

This study focuses on the development and investigation of sustainable and lightweight cellulosebased hybrid reinforced epoxy composites. The research contributes to the ongoing efforts to create durable and biodegradable composite materials for automotive applications. The hybrid composites were fabricated using a hand layup approach, combining sisal/dombeya fiber with paper particles as reinforcements in an epoxy matrix. Prior to incorporation, the fibers underwent mercerization to reduce hydrophilicity. Hybrid composites with 3-15 wt% reinforcements were produced. Mechanical properties, including tensile, flexural, impact, and hardness, were evaluated, and scanning electron microscopy (SEM) was used to examine the surface morphology of fractured composites. Wear resistance, density, and water absorption were also studied. Results demonstrated significant improvements in all properties compared to the unreinforced epoxy matrix. Notably, composites with 9-12 wt% sisal fiber-paper particles (SF-PP) exhibited optimal mechanical properties. Flexural modulus, hardness, tensile and impact strengths were 721 MPa, 67 HS, 32.94 MPa and 46.24 kJ/m2, respectively from 9 wt.% while flexural strength and tensile modulus were of 57.30 MPa and 438.21 MPa, respectively from 12 wt.%. On the other hand, the composite reinforced with 12 wt% dombeya fiber-paper particles (DF-PP) demonstrated superior wear resistance. DF-PP-based composites exhibited low moisture absorptivity and density compared to SF-PP. Conclusively, the study recommends epoxy-based composites reinforced with hybrid sisal fiber and paper particles for automotive components like bumpers and dashboards, while composites reinforced with hybrid dombeya fiber and paper particles are suitable for battery enclosures and wheel covers.

Key words: Plant fibers; Sisal fiber; Dombeya fiber; Paper particles; Hybrid composites; Sustainable; Lightweight.

1. INTRODUCTION

Green composites have been the focus of many researchers in recent times due to environmental concerns. Attentions have been given to composite materials based on its inherent benefit which are being derived from the flexibility in getting products developed for diverse areas of applications [1]. Composites have a wide range of applications in our day-to-day activities in areas such as automobiles, construction, electronic and households where glass fibers have recently become the most commonly used reinforcement material in polymer matrices [1, 2]. Glass fibers have been seen to be incorporated into approximately 95% of composite materials, and the extensive utilization of these synthetic fibers for reinforcement in the composite industry has

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raised concerns as they possess high manufacturing costs, non-biodegradability, and high toxicity levels, making them environmentally unfriendly and relatively unsafe for human health [1, 3-5]. As a result, researchers have shifted their focus towards natural fibers. The use of natural fibers as reinforcements in composite materials has been a significant area of research for the last couple of decades. These fibers are less dense (1.2-1.6 g/cm³), readily available, non-toxic, non-abrasive, and environmentally acceptable when compared to synthetic fibers most especially that of glass fibers (2.4 g/cm^3) [6-11] and with the exploration and development of green resources to create green products. The use of these fibers can serve as reinforcement for matrix material in the production of composite materials for various applications such as internal parts of automotive and building structures, making them eco-friendly and sustainable [1, 6, 12-14]. These lightweight and inexpensive materials aid researchers in developing high-performance products for various applications [1, 15-16]. Natural fiber integration can improve composite properties while using less polymer and lowering production costs. The automotive industry now uses natural fiber-reinforced polymer composites for a variety of interior and exterior car parts. Minerals, plants, and animals are the three main sources of natural fibers. Plant fibers including sisal, jute, hemp, and kenaf are widely used as reinforcement in composite goods. These fibers are readily available with good mechanical properties and recycling capacity [1, 17].

Plant fibers are already widely used in automotive applications as filler materials. The engine and gearbox covers of a Mercedes-Benz Travego are areas where they are currently been used in various exterior parts of automobiles. Banana fibers have also been used in the car industry to make body parts, such as the Mercedes A-Class's underfloor protective trim, which is a composite reinforced with banana fiber [6, 18]. Mercedes-Benz began utilizing an epoxy matrix to include jute in the door panels of their E-class vehicles in 1996 [6, 19]. In 2000, Audi unveiled the A2, a mid-range vehicle with door trim panels composed of polyurethane reinforced with sisal and flax fibers [6, 20]. Toyota has also made progress in this area by creating a sugar cane-based eco-plastic that will be used to line the interiors of its vehicles [6, 19]. The prospective uses of cellulosic fiber composites in different areas, including aircraft, biomedical and pharmaceutical industries, electrical components, packaging, and electromagnetic applications, have also been thoroughly investigated [6, 21-22]. Although natural fibers offer numerous benefits, their processing temperature and high cellulose content cause them to absorb a larger volume of water. The presence of lignocellulose presents challenges in achieving optimal strength properties and seamless compatibility between the fiber and polymer matrix [6, 23-25]. In order to determine the physical characteristics of cellulosic fiber-reinforced polymer composites, several researchers have studied the effects of water absorption. According to Atiqah et al. [26] water absorption results in a buildup of moisture in the cell walls of the fiber, which promotes fiber swelling and a decrease in dimensional

stability. Cellulosic fibers have a significant disadvantage due to fiber swelling brought on by moisture absorption, which leads to weak connections when the fiber interacts with the matrix in composites [27]. Akash et al. [28] observed that the water absorption patterns of these composites exhibited conformity to Fickian behavior when evaluated at room temperature, but Fick's law was not found to be applicable for water absorption properties at elevated temperatures. It can be noticed that water absorption increases as the proportion of cellulose fibers within the sisal/coir fiber-reinforced composites increases. Sherwani et al. [29] recently presented findings regarding an augmentation in the saturation of water absorption after a 15-day period of water immersion for composites composed of polylactic acid (PLA) reinforced with sugar palm fiber. Nevertheless, no further absorption was detected beyond the 5th day. The introduction of sugar palm fiber (SPF) into PLA resulted in a decrease in the value of the diffusion coefficient. Numerous distinct mechanisms are present in polymer composites that elucidate the diffusion of moisture within them. However, in order to enhance the interfacial bonding between hydrophilic natural fibers and hydrophobic polymer matrix, it is suggested to employ fiber pretreatment. From chemical standpoint, fiber pretreatment, such as alkali, silane, and acetylation treatment, works towards reducing the hydrophilicity of the fiber. Simultaneously, it physically eliminates waxy materials that cover the fiber surface, thereby, modifying the fiber texture [10, 17].

Among the polymeric materials utilized in the formation of the composite matrix, one that has been thoroughly researched by numerous researchers is epoxy. With exceptional mechanical, chemical, and thermal properties, it is a thermosetting polymer that is frequently utilized as matrices, adhesives, or impregnating materials. High adhesion, light weight, good mechanical and tribological capabilities, adequate chemical and corrosion resistance, little shrinkage after curing (good dimensional stability), and less rigorous processing are all characteristics of epoxy-matrix composites [30-31]. However, epoxy resins for engineering applications have lower stiffness and strength than metals. Also, the inability of epoxy resins to be recycled, reprocessed, or dispersed because of their permanent cross-linked structures makes them environmentally unfriendly compared to their thermoplastic counterparts. Resolving these environmental issues orchestrated by the thermoset material which is a significant concern [30-31], researchers have adopted diverse techniques to obtain and integrate additives into the polymer matrix with the aim of improving their properties and also produced sustainable and ecofriendly composites. One significant illustration of these additives is the utilization of plant fibers. Their integration into the polymer was commended as a prosperous recycling mechanism that yields eco-friendly products. This facilitates the production of environmentally conscious composites, thereby, reducing the ecological consequences of the initial waste [32]. In addition, this also makes the material a bio-composite material.

In a research conducted by Padmavathi et al. [33] on

mechanical behavior of surface modified sisal fibre epoxy composites. Studies were conducted on sisal-epoxy composites made by utilizing a novel thermogravimetric analyzer technique and ideally treated (18% NaOH) sisal fibers. Improvements in the mechanical properties of the composite ranged between 18% and 158% when the tensile property of the ideally treated fiber was increased by 110%. To measure the success of converting the improved fiber tensile property into the composite tensile property, a novel fiber treatment effectiveness criterion was devised. Furthermore, Oladele et al. [34] studied the mechanical and wear behaviour of pulverized poultry eggshell/sisal fiber hybrid reinforced epoxy composites. The results showed that composite reinforced with pulverized poultry eggshell/sisal fiber exhibited improved properties than the unreinforced epoxy. Also as seen in the results where dombeya fiber has been researched by Oladele et al. [35] by considering the influence of designated properties on the characteristics of dombeya buettneri fiber/graphite hybrid reinforced polypropylene composites. The findings indicated that the hybrid composite demonstrated superior properties, particularly the composite comprising of 12 wt% of dombeya buettneri fiber (DBF) and 8 wt% graphite particles (GP).

In this research sustainable and light weight hybrid reinforced epoxy-based composites for automotive application was developed using sisal fiber-paper particles and dombeya fiber-paper particles as hybrid reinforcements in epoxy matrix, respectively. Although several researchers have developed and studied hybrid reinforced epoxy-based composite with either sisal fiber or dombeya fiber and other types of fibers or particles, but paper particles have not been used. Consequently, this study investigates the utilization of sisal fiber and paper particles, as well as dombeya fiber and paper particles, as reinforcement materials for automotive applications. Its uniqueness lies in the incorporation of paper particles, which are expected to enhance lightweight and other properties when integrated into the epoxy matrix. The anticipated improvements was attributed to the cellulose content found in paper particles and both sisal and dombeya fibers. Part of the innovation was the re-use of papers which are often regarded as waste for secondary applications, thereby establishing a pioneering approach in the field of automotive materials.

2. Experimental

2.1 Materials and method

The present study employed various materials, including commercially available Bisphenol A diglycidyl ether epoxy resin and diethylene triamine curative, commonly known as hardener. These two liquids were mixed in ratio 2:1 and cured at ambient temperature of $24 \pm 1^{\circ}$ C within 5 hours. Also, waste papers, Agave sisalana leaves, Dombeya buettneri stems, sodium hydroxide, and distilled water were used. The epoxy resin and triamine curative were acquired from Pascal Scientific Akure in Ondo State, Nigeria. The Agave sisalana leaves were sourced from a farmland, Dombeya buettneri stems were cut from the plant sourced from pinkball trees in a shrub plantation while the waste paper was obtained from the school environment all been located at longitude 5.159440 and latitude 7.296990 in Akure, Ondo State, Nigeria.

2.2 Extraction and treatment of fibers

The extraction of sisal fiber from the plant leaves was carried out using the soil retting method while dombeya fiber was obtained by manually removing the bark from the stem and separating the strands. The fibers were sun-dried for a period of 5 days after extraction. Both the sisal and dombeva fibres were respectively, subjected to chemical treatment using 1 M sodium hydroxide solution at 50°C for 4 hours in a shaking water bath (Uniscope Surgifriend Medicals, England; Model No.: XMTD-8222). This was followed by thorough washing using tap water and rinsing with distilled water until neutral status was confirmed by the digital pH meter. This was done to ensure complete removal of any sodium hydroxide present within the fibres. Following this, the mercerized fibers were dried under the sun for a period of 5 days during the dry season and cut into small pieces measuring 10 mm in lengths before oven dried at 60°C for 4 hours to remove moisture content and stored in airtight zip-lock polybags for further use. These methods of extractions and alkaline treatment were carried out in accordance with Oladele et al. [34] and Oladele et al. [35] Fig. 1 (a-b) showed the sun-dried sisal and dombeya fibers, respectively.

2.3 Production of paper particles

Waste papers were first reduced in size using a paper cutter, and subsequently immersed in water within a plastic bucket for a period of 3 days. The soaked papers were then grinded with laboratory grinding machine to form pulp. The paper pulp was manually squeezed, and later sun dried for 10 days to obtain paper particles.

2.4 Fabrication of composite

The composites were fabricated through open mould hand lay-up technique which involved the incorporation of both particles and fibres, within the range of 3 to 15 wt% into the epoxy matrix while the epoxy resin and hardener were added in a ratio of 2:1. The fibers and paper particles were mixed in ratio 1:1 as shown in Table 1. The epoxy resin, hardener, and sisal/dombeya-paper particles were mixed manually with a glass rod stirrer for 5 minutes within a plastic container to get a proper mix. The compounded mixtures were then introduced into moulds specifically designed for each property to be investigated. The samples were allowed to cure in the moulds at ambient temperature of $24 \pm 1^{\circ}$ C before extraction from the moulds. Details regarding the formulation and constituent amounts utilized are provided in Table 1.



Fig.1 (a) Extracted sisal fiber



Fig. 1 (b) Extracted dombeya fiber

2.5 Evaluation and characterization of the developed composite

2.5.1 Flexural test

Flexural properties were evaluated using Three Points Bending Test in accordance with the standards established by ASTM D790 [36]. The tests were conducted through the utilization of a universal tensile testing machine of the Instron Series 3369 model. The dimensions of the specimens used were $120 \times 15 \times 3$ mm for length, width, and thickness, respectively. The execution of the test was carried out under a displacement control rate of 10 mm/min. The test speed was set at 5 mm/min over a span measuring 65 mm. The representative values were calculated from the average of the results obtained from three samples tested for each composition.

2.5.2 Tensile test

The examination of tensile properties of the fabricated samples was executed in accordance with the ASTM D638-14 standard. [37] The procedure was carried out utilizing a Universal Testing Machine (Model: Instron series 3369). Dumbbell-shaped specimens measuring 90 \times 10 \times 3 mm were employed for experimentation, and the trial was executed with a crosshead speed of 5 mm/min utilizing a 10 kN load cell.

2.5.3 Impact test

Impact test was conducted to evaluate the impact resistance of the samples using Charpy impact testing device per ISO 179 regulations. The samples with dimensions $64 \times 11 \times 3$ mm and centrally notched were placed flat on the equipment, separated by 60 mm between each point of support. The initial reading of the gauge was documented prior to releasing the suspended handle that swings and fractures the sample. Once the sample was fractured, the final reading was taken. Each composition was tested 3 times, and the calculated average was considered as the representative value.

2.5.4 Hardness test

Hardness test was executed on the samples through implementation of a Shore D hardness tester according to ASTM D2240-00. [38] In order to accomplish this, a force of 15 kgf was exerted upon each composite sample with a 15 s pause. To obtain a comprehensive understanding of the composite's hardness, 5 discrete values were procured by indenting the material at 5 distinct locations. The average value acquired from these measurements was subsequently employed for analytical purposes.

2.5.5 Wear test

For wear test, a CS-10 calibrator was employed using the Taber Abraser (Model: ISE AO16). A normal load of 750 g was applied, and 76 revolutions per minute (rpm) were used. To attach the test piece to the apparatus, an 8-mm center hole was bored into the sample. The instrument platform, which is propelled by a fixed-speed motor, was secured with the sample. Wear resistance (weight loss technique) was calculated using the difference in weight between the two weights before and after abrasion. Before testing, loose particles that were stuck to the samples were thoroughly removed. Using Equation (1), the wear index of each sample was determined. Where; W_1 is initial weight, W_2 is final weight and RPM is revolution per minutes number of test cycles.

Wear Index =
$$\frac{W1-W2}{RPM} * 100$$
 (1)

2.5.6 Water Absorption

Water absorption tests were conducted in accordance with the guidelines set by ASTM D5229M-12 [39]. To execute the test, 250 cm³ of water medium was dispensed into uncontaminated plastic containers. The original weight of each sample was measured via chemical weighing, and readings were noted every 24 hours for a duration of 4 days. Similar number of durations was done in Bekele et al. [48]. The samples were taken out, cleansed with a fresh cloth before each weighing session. The data collected during the experiment were employed to determine the weight gained through utilization of Eq.2. Where, %W is percentage water absorption, Wo and Wt are the oven-dry weight, and the weight of the sample after time t, respectively. Similar formula was used in Gurunathan et al. [49]

$$\%W = \frac{Wt - Wo}{Wo} * 100 \tag{2}$$

2.5.7 Density Measurement

The samples were weighed, and their volume was estimated after taking the volume and mass of each sample into consideration. Densities were then computed using Eq.3.

$$Density = \frac{Mass}{Volume}$$
(3)

2.5.8 Microstructural Examination

The surfaces of the fractured fabricated composites from the enhanced samples were examined using Scanning Electron Microscope (Model: JEOL JSM-6480LV). The samples were securely affixed to stubs with the aid of silver paste. To heighten the conductivity of the composite specimens, a slender layer of platinum was subjected to vacuum evaporation onto the surface before photomicrographs were captured at a voltage of 15 kV.

Table 1	- Formulation of	f composite
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Fiber Composition	Epoxy	Hardener	Sisal/Dombeya fibers	Paper particles
(wt %)	(g)	(g)	(g)	(g)
Control	133.3	66.6	-	-
3	116.4	58.2	2.7	2.7
6	112.8	56.4	5.4	5.4
9	109.2	54.6	8.1	8.1
12	105.6	52.8	10.8	10.8
15	102.0	51.0	13.5	13.5

3. RESULTS AND DISCUSSION

3.1 Flexural properties

The flexural examination evaluates the magnitude of force necessary to flex a beam under specific three-point loading conditions. Frequently, the results of this test are employed to determine the most suitable materials for manufacturing components that can endure loads without yielding to flexure [40]. The findings are presented in Fig. 2, which delineates the correlation between maximum flexural strength at various sisal/dombeya fiber-paper particles loadings and the control. A notable elevation in the flexural strength of composites reinforced with sisal fiber/paper particles was observed within the range of 6 to 15 wt. %, with an optimal strength of 57.30 MPa at 12 wt. %, as compared to the control's 32.52 MPa of epoxy material which is a 76% increment observed. Also an increase was observed from 3-9 wt% dombeya fiber/paper particles with 9 wt% dombey a fiber/paper particles having the optimum flexural strength at 35.70 MPa as compared to control having an increment of approximately 10%. It is plausible that the alkaline treatment contributed to the enhancement of mechanical properties of the polymer composites [10]. Also, it was observed that the flexural strength exhibited an upward trend upon the inclusion of reinforcement (SF-PP) within the range of 3-12 wt% and within the range of 3-9 wt% DF-PP. This occurrence could potentially be ascribed to the homogeneous distribution of stress and the interlocking configuration of the fibers in the matrix, which served as influential factors in the refinement of the material's attributes. Likewise, the suitable blend of paper particle in the epoxy matrix played a pivotal role in augmenting the material's characteristics. At 15 wt. % SF-PP, a reduction in the flexural strength of the reinforced composite sample was detected likewise at 12-15 wt% DF-PP. This decline could be ascribed to insufficient blending of paper-particles with epoxy or the dispersion of sisal fiber/dombeya fiber in the matrix [41]. In addition, SF-PP reinforcement from 6-15 wt% all had higher flexural strength compared to that of DF-PP reinforcements which can be attributed to higher strength of sisal fiber compared to dombeya fiber. However, at 3wt% DF-PP the flexural strength (34.30 MPa) was higher than at 3 wt% SF-PP reinforcement (22.32 MPa) which can be due to proper dispersion of DF-PP in the matrix at this content. Similar results were observed in Oladele et al. [34] and by Oladele et al. [35] when sisal fiber and eggshell were reinforced in epoxy matrix and when dombeya buettneri fiber/graphite were added as reinforcements in polypropylene matrix, respectively. Also, the result was in agreement with Madgule et al. [42] when banana fiber and sugarcane bagasse powder were used in epoxy-based composite.

In a similar vein, Fig. 3 displays the correlation between the maximum flexural modulus at various sisal/dombeya fiber-paper particles loadings and the control. It exhibits a comparable trend with Figure 2, albeit with a shift in the optimal value from 12 wt% SF-PP of reinforced epoxy composites for flexural strengths to 9 wt% reinforcements in flexural modulus. Thus, the optimal values from the sisal fiber/paper particles hybrid-based composite was 721 MPa, compared to the unreinforced epoxy with a value of 410 MPa giving a 76% increment. While for dombeya fiber/paper particles reinforced matrix there was no change in the optimum value among the weight of reinforcement used, 9 wt% DF-PP exhibited the highest flexural modulus across the composites having 502 MPa in its flexural modulus which delignates a 22% increment in the flexural modulus. This enhancement can be attributed to the robust interconnectivity between the bio fillers and epoxy matrix. However, the addition of further reinforcement subsequently led to a marginal decrease in the flexural modulus at 12 and 15 wt. % SF-PP and DF-PP, respectively. This phenomenon can be ascribed to an elevated fiber loading in the matrix. The aforementioned reduction can also be attributed to the entanglement and agglomeration of fibers, resulting in the initiation of cracks and stress concentration in the materials [10]. Similar to these findings was the work of Panneerdhassa et al. [43] When comparing the two types of reinforcement applied in the matrix, it was observed that sisal fiber-paper particles reinforcement exhibited a higher flexural modulus as opposed to dombeya fiber-paper particles reinforcement.



Fig.2 Maximum flexural strength of hybrid reinforced sisal/dombeya fiber-paper particles composites and unreinforced epoxy matrix



Fig.3 Maximum flexural modulus of hybrid reinforced sisal/dombeya fiber-paper particles composites and unreinforced epoxy matrix

3.2 Tensile properties

The result in Fig. 4 features the stress strain curve of the reinforced and unreinforced epoxy matrix while Fig. 5 presents the plots of the ultimate tensile strength results. It was observed that there was an increase in the maximum

tensile strength of the composite materials from 3 wt. % to 9 wt. % SF-PP and DF-PP for reinforced composites followed by gradual reduction. This enhancement in strength with an increase in reinforcement content followed by a decrease was in agreement with the report of the findings in Oladele et al. [10]. The outcomes of the study reveal that the composite that incorporated 9wt% SF-PP and DF-PP reinforcement manifested a tensile strength of 32.94MPa and 22.82MPa, respectively, which is significantly higher in comparison to the 17.64 MPa observed in the control sample culminating to 87% enhancement for SF-PP and 29% for DF-PP based composites, respectively. These results imply that the amalgamation of hybrid reinforcements and epoxy matrix was successful in achieving proficient adhesion and stress distribution. Nevertheless, the inclusion of sisal fiber/dombeya fiber and paper particles beyond the abovementioned reinforcement loadings was found to cause a gradual decrease in tensile strength. This decrease could be ascribed to inadequate bonding at the fiber/matrix interface, ultimately leading to a reduction in the bonding strength of the composite [10]. As illustrated in Figure 5, it was evident that the tensile strength of the epoxy reinforced with sisal fiber-paper particles was superior to that of the dombeya fiber-paper particles reinforced matrix. This disparity may be attributed to the dissimilarity in the tensile strength characteristics of sisal and dombeya fibers.



Fig.4 Stress-strain curve of hybrid reinforced sisal/dombeya fiber-paper particles composites and unreinforced epoxy matrix

The effect of hybrid reinforcement consisting of sisal/dombeya fiber-paper particles on the epoxy matrix was demonstrated by analyzing the tensile modulus, as presented in Fig. 6. The results indicated that the tensile modulus of the composite samples, which were fabricated using various reinforcement ratios ranging from 3-12 wt% SF-PP, exhibited an overall improvement than the DF-PP based composites and the control. While for DF-PP based reinforcements, the tensile modulus of the composite samples which were fabricated using various reinforcement ratios ranging from 3-15 wt% DF-PP, exhibited an overall improvement than the control. Hence, all the composite samples that were reinforced either with SF-PP or DF-PP exhibited a higher tensile modulus than the control sample except for 15 wt% SF-PP. Thus, the

optimal content of the reinforced SF-PP hybrid composite was found to be 12 wt%, yielding a tensile modulus of 438.21 MPa while that of DF-PP was found to be 9 wt% having 415 MPa. It can be inferred that the significant increase in the tensile modulus from 3-12wt%SF-PP and 3-15 wt% DF-PP was due to the excellent bonding between each reinforcement and the epoxy matrix [34].



Fig.5 Ultimate tensile strength of hybrid reinforced sisal/dombeya fiberpaper particles composites and unreinforced epoxy matrix



Fig.6 Tensile modulus of hybrid reinforced sisal/dombeya fiber-paper particles composites and unreinforced epoxy matrix

The reduction in tensile modulus when the concentration reaches 15 weight percent SF-PP can be ascribed to insufficient improper adhesion at interface as the concentrations increases or the clustering of fibers [44]. This observation emphasizes the concept that the integration of fillers in the polymer matrix amplifies the stiffness of the composites within a specific range of optimal values [34]. Similar trend was observed in Fig. 5 and in Oladele et al. [34]. A reduction in the tensile modulus was detected in the composites reinforced with dombeva fiber/paper particles at 12-15wt%. However, the reduction did not fall below the control. This observation may be attributed to the same issue with SF-PP based composites. Thus, the increase observed in the tensile modulus of the bio-composites aligns with the findings of Abare et al. [45], Ismail et al. [46] and Vigneshwaran et al. [47], which stated that the tensile modulus of the developed composites improves as the filler addition increases up to a specific point. However, it decreases when the reinforcement content reaches its maximum level.

3.3 Impact strength

The impact of hybrid reinforcement employing sisal/dombeya fiber-paper particles on the epoxy matrix was demonstrated by means of Fig.7, and a congruent tendency was noted much like the mechanical properties indicated in Fig. 2-6.

The composites that were produced possess significantly enhanced impact strengths when compared to the control. with the optimal value being 9 wt. % for both SF-PP and DF-PP exhibiting an impact strength of 46.24 kJ/m2 and 23.74 kJ/m2, respectively when compared to the control, which had an impact strength of 13.82 kJ/m2. Nevertheless, a decline was observed with further addition of sisal/dombeya fiber-paper particles reinforcement from 12-15 wt%, which could be attributed to the accumulation of particles/higher fiber loading in the composites, thereby reducing their energy-absorbing capacity [48]. However, in comparison to the control sample, the reinforced samples exhibited adequate interfacial adhesion between the bio fibers subjected to treatment and the epoxy matrix. This improvement can be attributed to the surface modification of the bio fibers [25]. Similar findings have been reported by Oladele et al. [34], Adekomaya et al. [49], Teboho et al. [50], Gudayu et al. [51] as the addition of reinforcement led to a significant increase in the impact strength of the composite. By comparing the two types of reinforcements, it was observed that 3-6 wt. % DF-PP reinforcement exhibited higher impact strength when compared to SF-PP reinforcement. Conversely, 9-15 wt.% SF-PP reinforcement showed higher impact strength when compared to DF-PP reinforcement.



Fig.7 Impact strength of hybrid reinforced sisal/dombeya fiber-paper particles composites and unreinforced epoxy matrix

3.4 Hardness

Fig. 8 illustrates the hardness of the composites and the control. It was observed that, with the exception of 6-12

wt% SF-PP and 3-12 wt% DF-PP reinforced samples, the hardness of all other composite samples was lower than that of the control sample. This could potentially be attributed to the reasons stated for the response of other mechanical properties from Fig. 2-7. It has further been noted that an increasing trend was observed between the 3-9 wt% ranges of sisal/dombeya fiber-paper particles hybrid reinforcement, with the optimum value being recorded at 9 wt% reinforcement for both SF-PP and DF-PP with hardness value of 67 HS and 64 HS that culminated to 16% and 10% enhancements, respectively. However, at higher addition within 12-15 wt% reinforcements from both fibers, the hardness value decreased. From the result, the enhancement in rigidity could potentially be attributed to the effective interfacial adhesion between the reinforcing agents and the matrix, which fosters the property of hardness. Additionally, this phenomenon may also be linked to an increase in stiffness and the interlocking of fibers [52]. It was also observed that with the exception of the 3 wt% SF-PP reinforcement, all other weight percentages of SF-PP displayed a greater degree of hardness when compared to that of the DF-PP reinforcement. This observed trend was also in agreement with the findings of Özturk et al. [53].



Fig. 8 Hardness of hybrid reinforced sisal/dombeya fiber-paper particles composites and unreinforced epoxy matrix

3.5 Wear Property

This investigation was carried out to compare the wear behavior of various materials with the control as illustrated in Fig. 9. An abrasion wear test was executed to examine the impact of reinforcements on the wear loss of material surfaces. The findings revealed that these reinforcements resulted in variances in the abrasive properties of the produced composites in comparison to the epoxy resin, which functions as the control. All the generated composites exhibited lower wear index, indicating a higher level of wear resistance than the control sample. Thus, it can be inferred that the addition of these reinforcements can enhance the wear resistance of epoxy materials in all areas of application where surface abrasion is prevalent [40]. The findings from Fig. 9 indicate that composites from DF-PP exhibited the best wear resistance than SF-PP based composites. Moreover, composites that are reinforced with DF-PP had lower level of wear index when compared to those reinforced with SF-PP. This indicates that the composite reinforced with DF-PP will demonstrate greater wear resistance than the one reinforced with SF-PP. Composites containing 9-12 wt% DF-PP reinforcements exhibited low wear index of 0.14 and 0.11 mg while 3-6 wt% SF-PP reinforcement exhibited the lowest wear index of 0.49 and 0.46 mg, respectively. This finding can be supported by the similar results observed in the findings of Ramesh et al. [54] and Shalwan and Yousif [55].



Fig. 9 Wear Index of hybrid reinforced sisal/dombeya fiber-paper particles composites and unreinforced epoxy matrix

3.6 Water absorption

The water absorption characteristics of all the composites and the control samples under study as influenced by immersion time of 96 hours was presented in Fig. 10. The data depicted in the plots clearly indicate that a rise in the content of reinforcements is directly proportional to an increase in the percentage of water absorption. This observation could be attributed to the inherent hydrophilic properties of the natural fibers [56]. It was discovered that the reinforced composite having 3 wt% SF-PP and DF-PP exhibited low absorption like the unreinforced matrix sample compared to the other reinforced composite that exhibited rapid and linear water absorption during the initial phase, specifically within the first 24 hours. Similar trend was observed for composites that were reinforced with other natural reinforcements like eggshell and sisal fiber [34] as well as plantain fibers [10]. It was revealed from the plots that further increase in immersion period can lead to increase in the absorption rate of the composite as there was no saturation observed after 96 hours. Hence, further work can be done to investigate water absorption rate beyond 96 hours. As seen in the research done by Oladele et al. [57] where moringa oleifera fruit waste pod was used as reinforcement to develop a biocomposite for structural application. The water absorption rate was studied for duration of 168 h likewise in the research conducted by Akash et al. [58] where the water absorption rate was also studied for 627 h using sisal/coir fibers as reinforcements. Though, the process of diffusion can be

delineated into three stages which are in accordance with Fick's law of diffusion and was noticed in the previous research outputs. The process consists of the initial stage in which water molecules permeate the micro-gaps within the polymer chains followed by capillary action that facilitates the transportation of water through cracks. Thus, failure occurs at the interface between the fibers and the matrix which is attributed to inadequate wetting and impregnation during the initial phase of water molecule diffusion. Finally, the swelling of the fibers that induces the propagation of microcracks throughout the matrix [27, 59]. To prevent this failure mode, fiber treatment is usually carried out on natural fibers. If treatments are not carried out on these fibers, the absorption may likely be more than this, hence, further study should also investigate comparatively, the influence of water absorption on both treated and untreated fibers beyond this period in this study. This can aid the use of the materials in moist environments. It was noticed from Figure 10 that 15 wt% SF-PP reinforced composite had the highest water absorption rate and composite reinforced with SF-PP exhibit high water absorption rate compared to that of DF-PP composites. This good water resistance can also be attributed to part of the reasons for good wear resistance achieved in Fig. 9.



Fig. 10 Absorption rate of hybrid reinforced sisal/dombeya fiber-paper particles composites and unreinforced epoxy matrix

3.7 Density

Fig.11 displays the response of an epoxy composite reinforced with sisal/dombeya fiber-paper particles as a function of density. It was noticed that an upward trend was evident from 3-9 wt% when reinforcing materials were added to the epoxy matrix with 9 wt% exhibiting the highest density of 1.194 gcm⁻³ and 1.162 gcm⁻³ for SF-PP and DF-PP composites based, respectively. It was discovered that all the samples with reinforcements had lower densities compared to the reinforcement epoxy (1.237gcm⁻³) which can be attributed to the inherent density of both the matrix and the reinforcements. By incorporating sisal/dombeya fiber and paper particles into the epoxy resin, the overall density of the composite

materials is decreased. This occurs because the fibers and paper particles displace some of the epoxy resin, which has a higher density, thereby lowering the overall density of the composite material. Also, it was discovered that DF-PP based composites possess less density compared to SF-PP based composites but 3 wt.% SF-PP reinforced composite was the sample with the least value of 1.107gcm⁻³, Since all the developed composites possess low densities compared to epoxy matrix, they are therefore, suitable materials for automotive application as a light-weight.



Fig. 11 Density of hybrid reinforced sisal/dombeya fiber-paper particles composites and unreinforced epoxy matrix

3.8 Microstructural analysis

Scanning electron microscope (SEM) was used to examine the fractured surface morphology of the developed composites. From Fig. 12 and 13 (a-b), the image depicts the various compositions of 3 wt. % and 12 wt. % from sisal fiber-paper particles hybrid reinforced epoxy composite and 3 wt% and 9 wt% dombeya fiber-paper particles hybrid reinforced epoxy composites. The images revealed the intrinsic coalescence and adhesion of the epoxy polymer to the reinforcements as shown in Fig. 12 and 13 (a-b). The surface pattern further indicates the creation of homogeneous mix in the host resin by the reinforcements, with both the paper particles, sisal fiber and dombeya fiber being embedded in the polymer matrix. No observation of fiber debonding due to the chemical treatment given to the fiber as shown in Fig. 12 and 13 (b). This may contribute to the reasons why there was an improvement in the mechanical properties at this high reinforcement contents.

4. CONCLUSIONS

Based on the findings from this research, composites from the selected plant fibers were suitable for polymer composite development with epoxy resin. It was discovered that sisal fiber-paper particles-based composites demonstrated superior properties in terms of mechanical properties where 9-12 wt% sisal fiber-paper particles (SF-PP) reinforcements exhibited the optima values. Flexural modulus, hardness, tensile and impact strengths were 721 MPa, 67 HS, 32.94 MPa and 46.24

kJ/m2, respectively from 9 wt.% while flexural strength and tensile modulus were of 57.30 MPa and 438.21 MPa, respectively from 12 wt.%.



Fig. 12 Scanning Electron Micrograph (SEM) of epoxy composites reinforced with: (a) 3wt %; (b) 12wt % sisal fiber- paper particles



Fig. 13 Scanning Electron Micrograph (SEM) of epoxy composites reinforced with: (a) 3wt %; (b) 9wt % dombeya fiber- paper particles

Combining the good wear resistance with low density properties, these composites are potential materials for automotive applications where they can be used majorly for interior applications like dashboards, covers and seat parts. While composites comprising dombeya fiber-paper particles that exhibited higher properties in terms of wear index and hydrophobicity as well as low density can be used in battery enclosures, glove box and wheel covers. The work has presented the use of recycled wastepaper particles as fillers in thermosets for light-weight materials development in addition to fiber reinforcement where both reinforcement shapes (particles and fibers) will play synergetic roles. Also, this research has advanced the use of natural resources as sustainable materials for automotive application. Since recycled waste papers has not been widely used for composite development for most engineering application, this materials is now being recommended for use for sustainable low technology and light-weight material development as alternative to other fillers. Recycled wastepaper-based polymer composites are also suggested for transportation and electronic industries. Due to the presence of paper and other lignocellulose materials, more moisture absorption test research should be carried out to determine the suitability in various moisture environments.

DECLARATION OF CONFLICTING INTEREST

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