

Plant Fiber Constituents and Impact of Environmental Degradation on the Physical, Hardness and Thermal Properties of Banana-Sisal Fibre Hybrid Reinforced Epoxy Composites

Abstract

Environmental factors such as moisture, microbial action, UV radiation, and temperature variations can significantly impact the performance and durability of natural fibre-based polymer composites in automobile components. This study investigated the fallout of environmental degradation on the physical, hardness and thermal properties of banana-sisal fibre-reinforced epoxy composites. Composite samples were developed by reinforcing epoxy resin with alkali-treated banana and sisal fibre by the hand lay-up method. Effect of environmental degradation was evaluated by determining the physical, hardness and thermal attributes of the composites subjected to soil burial and atmospheric degradation for a period of 6 and 12 months. Results showed that all the composite samples had specific densities within 1.0 and 1.2 g/cm³. The hardness of the composite samples subjected to atmospheric conditions showed increase with increase in time of exposure while samples that were buried in soil revealed reduction in hardness with time due to enzymatic activity of soil microorganisms. Also, thermal insulating properties of the samples showed high resistance to thermal conductivity except the samples that were subjected to soil degradation within 6 months. Thus, the effects of the studied environments on selected properties are acceptable to the composites within the period of investigation. This implies that, this material will not fail in service if exposed to these adverse environmental conditions within the period of research.

Keywords: Environment, Degradation, Plant fibre constituents, Automobile, UV radiation, thermal property

1.0 Introduction

Polymer composites strengthened with fibres from biodegradable and renewable materials and polymer matrices are of great interest to the automotive industry due to their potential to considerably reduce environmental degradation [1]. However, due to the presence of these natural fibres, the composites are vulnerable to degradation. Consequently, it is necessary to find a poise between the performance of the composite and its biodegradability [2]. As sustainability swiftly ascends as a global priority across various sectors, the composite industry is similarly shifting towards producing sustainable composites. The creation of these composites, which are both lightweight and high-performing at a reduced cost, is primarily driven by the use of biodegradable and sustainable materials like vegetable fibres [3]. Composites materials that comprise of at least one of the matrix or reinforcement gotten from natural sources are grouped as partially biodegradable composites, while those in which the matrix and reinforcement are from natural earth sources are designated fully bio-degradable composites [4, 5]. Natural fibres are therefore demonstrated to be effective materials for creating sustainable composites because of their availability, affordability, non-abrasive quality, lightweight, excellent mechanical properties, as well as their environmental friendliness and biodegradable nature.

Sisal and banana fibres are promising reinforcement materials which have been used in the development of polymeric based composites for different utilizations including automobile parts [6]. Some applications of banana and sisal fibres composites in automobile parts are under-floor protection in passenger cars, door panels, interior panelling and ceiling sheets of automobile [7]. Natural fibre polymer matrix composites are developed using natural fibres which are highly

degradable and can affect the degradability of the matrix in which they are utilised when exposed to certain environments like UV radiation, salt water, road salt, oil, soil and thermal cycling [8,9].

Natural fibre reinforced polymer composites used in automobile parts need to manage heat generated from electronic components on the dashboards like infotainment systems and from external heat sources like sunlight through the windows and also facilitate efficient dissipation of heat generated during braking or due to engine heat near the front of the vehicle [10, 11]. Therefore, there is need for their thermal conductivity to be relatively low and prevent heat build-up, maintain comfort levels inside the vehicle and resist deformation due to heat by absorbing the heat energy.

The thermal conductivity of natural fibre composite materials decreases with increased fibre content fraction and this is due to the presence of lumen which is an air-filled hollow portion, therefore their content of the composite is increased and this leads to heat insulation [2,12]. The thermal conductivity of natural fibre polymer matrix composites can be increased with different modification process as reported by [13]. [14] researched the impact of randomly oriented jute and banana fibre on the heat insulation of epoxy and they observed that the thermal conductivity was reduced at the highest fibre volume fraction [2].

Due to the ability of natural fibres to degrade and deteriorate, the knowledge of how natural fiber composites degrade under different environmental conditions helps to assess the structural integrity of the composites and informs predictions about their performance [15, 16]. This paper therefore reveals the influence of soil burial and atmospheric degradation on the physical, hardness and thermal attributes of banana-sisal fibre hybrid reinforced epoxy composites.

2.0 Materials and Method

The method used in the fabrication of the developed composites is the hand lay-up. Hybrid composites, combining sisal and banana (1:1), and epoxy resin and hardener (2:1) were developed using various mix ratios, 3, 6, 9, 12, 15 and 18 wt. %.

2.1 *Materials Used*

The materials utilized for this work are banana fibre and sisal fibre sourced from farms in Ilaramokin, Ondo State; (NaOH, 97%) manufactured by Molychem, India and procured from Pascal Scientific Ltd., Akure, Ondo State; distilled water was produced in Chemistry Department, Federal University of Technology Akure, Ondo State; epoxy resin and hardener gotten at Adesina Street Off Obafemi Awolowo Road, Lagos State, Nigeria.

2.2 *Plant fibre extraction*

Banana fibre and sisal fibre were extracted using the dew retting and soil retting technique, respectively. These methods have been used by [17, 18] and have produced good fibres with high strength properties [19].

2.3 *Modification of plant fibres*

Banana and sisal fibres were treated using 1M NaOH sodium hydroxide solution at 50°C for 4 hours in a shaker water bath. Then, the fibres were cleansed with distilled water to make the fibres neutral. After this they were dried at 60°C for 24 hours according to [20].

2.4 *Exposure to Degradation*

The developed composites were split into three groups, one part was used as control while the other two groups were subjected to soil burial and atmospheric degradation for degradation for a period of 6 months and 1 year, respectively. For the soil samples, composite samples prepared in

three replicates for each composition were buried in a dug pit of dimension $610 \times 610 \times 610$ mm from the soil surface at Ilara Mokin, Ondo State, Nigeria with GPS coordinates 7.3671° N, 5.1067° E and removed after the experimental period. On the other hand, samples for the atmospheric environmental degradation tests were exposed to normal atmospheric conditions at West African Service Centre on Climate Change and Adapted Land Use (WASCAL) FUTA and the daily observatory data were obtained for the period. The range of weather data obtained for air temperature range, relative humidity, UV radiation, and precipitation for the study were $20.6^\circ\text{C} - 34.88^\circ\text{C}$, $27.69\% - 100\%$, $0.001 - 0.938 \text{ kW/m}^2$, and $0 - 17.7 \text{ mm}$, respectively.

2.5 Chemical content analysis

To ascertain the percentages of cellulose, wax, ash, moisture, lignin, holocellulose and hemicellulose present in the treated banana and sisal fibres used, chemical content analysis was carried out following TAPPI T257 cm-12 standards [21]. The treated fibres were cut to smaller sizes using a scissors and grinded in a laboratory grinder. The grounded fibres were then sieved to a size of $150-300 \mu\text{m}$ particle sizes and then analysed to determine their content. The analysis was carried out according to work of [22,47].

(a) Moisture content

The moisture composition was determined according to TAPPI T412 om-16 standard [23]. For moisture content determination, 2.0 g of milled sample in an evaporating dish was placed in an air oven of Model No. DHG-9101.1SA at 105°C for 3 h, after which it was removed and cooled to ambient temperature in a desiccator and the mass was weighed. This procedure was repeated for another 30 min until the mass was constant. The moisture content was then calculated using Equation (1).

$$\text{Moisture content \%} = \frac{M_1 - M_2}{M_3} \times 100 \quad (1)$$

M_1 = mass of wet sample and beaker; M_2 = mass of the dry sample and beaker; M_3 = mass of wet sample

(b) Ash content

The ash contained in the fibres was determined following TAPPI T211 om-07 standard [24,35] on a moisture-free sample basis. First, an empty crucible with its lid was cleaned by calcination at a temperature 550°C for 30 min and then cooled to room temperature. The mass of the calcined crucible with the lid was then measured. To determine ash content, 2.0 g of milled moisture-free sample was measured and put into the 50 mL porcelain crucible, with the lid removed,

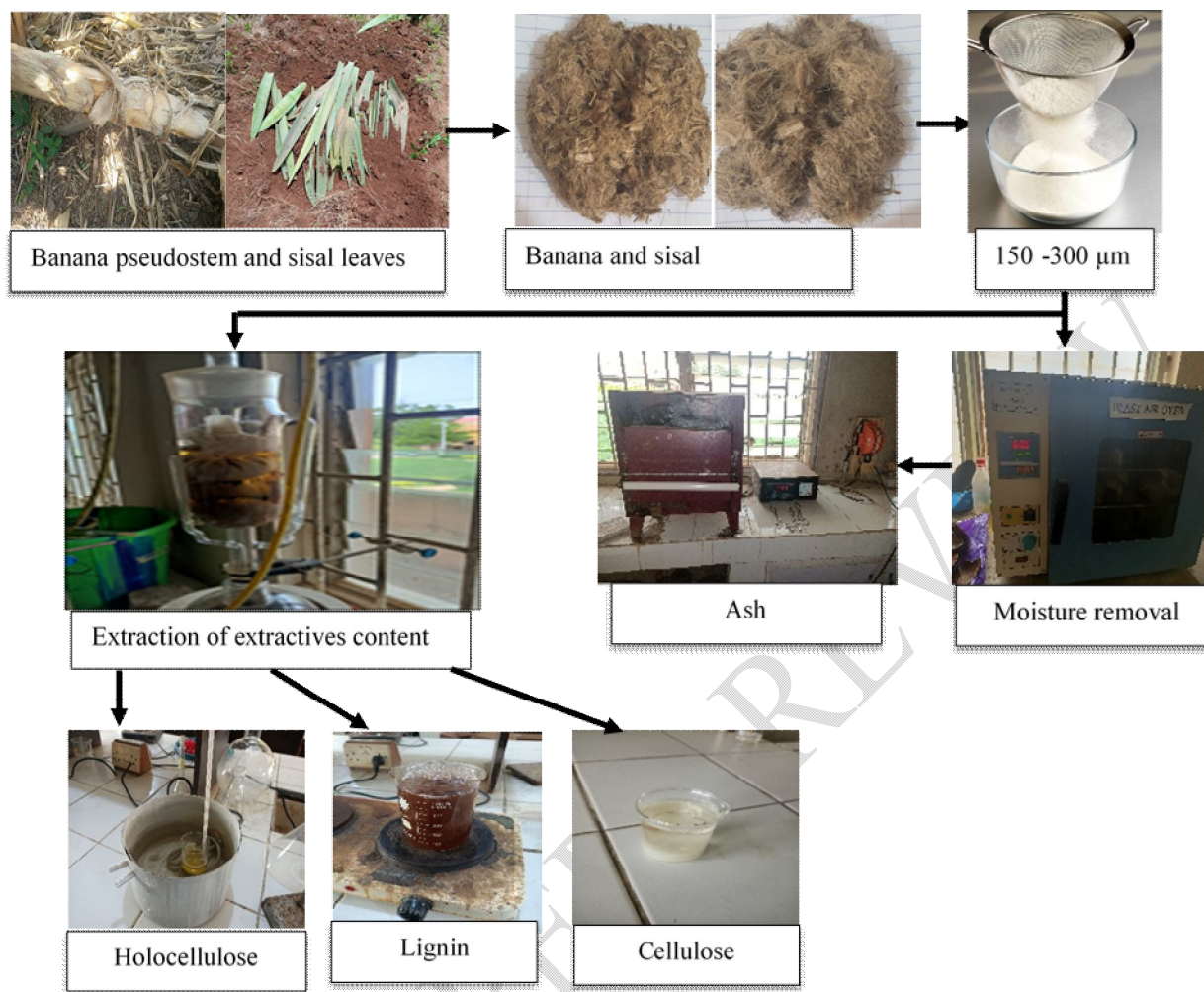


Figure 1: Quantitative Analysis for Chemical Composition Determination

which was placed in a muffle furnace (Techmel and Techmel USA Model No. 05210100P) at a temperature of approximately 100°C. The temperature was then slowly raised to 550°C to carbonize the sample without flaming. The system was maintained at this temperature for 3 hours. After complete combustion of the sample the crucible was brought out from the furnace, covered and allowed to cool to room temperature. The mass of the crucible (with cover) with ash was measured. The process was repeated until the mass was constant.

Ash content was then calculated using Equation (2).

$$\text{Ash content \%} = \frac{M_{ccs} - M_c}{M} \times 100 \quad (2)$$

M_{ccs} = mass of calcined crucible with the sample; M_c = mass of crucible; M = mass of moisture-free sample.

(c) Determination of extractives content

The content of extractives was determined following TAPPI T204 cm-07 standards [25], with toluene-ethanol as the extraction media. The extractives removed from lignocellulosic sources include low molecular weight carbohydrates, non-volatile hydrocarbons, waxes, salts, fats and resins. To determine extractives content, 2.0 g of moisture-free milled, sieved, dried, powdered sample was placed in an extraction thimble. 2:1 (v/v) toluene/ethanol mixture solution was added to the extraction thimble and the mixture refluxed for 24 hours, 4 hours per day at a boiling temperature in a Soxhlet apparatus. The dewaxed sample was dried at 105°C for 1 hour in an oven and the mass was measured. The extractive content was calculated using Equation (3).

$$\text{Extractables, \%} = \frac{M_{re} - M_{df}}{M} \times 100 \quad (3)$$

M_{re} = mass of container with the sample; M_{df} = mass of container and dry extractive-free sample; M = mass of moisture-free sample

(d) Acid-insoluble lignin content

Acid-insoluble lignin was content of the fibres were determined according to the TAPPI T222 om-06 standard [26]. Lignin which is also called Klason lignin is the wood constituent insoluble in 72% sulphuric acid. To determine this, 15 mL of 72% (w/w) sulphuric acid with a temperature of between 10 and 15°C was gradually added to the beaker containing 1.0 g of sample remaining

in (c) above while stirring with a glass rod. The mixture was left to rest for 2 hours at ambient temperature and later transferred to a flask ensued by the addition of 575 mL of distilled water to bring the acid concentration to 3%. The resultant solution was boiled for 4 hours in a hotplate while maintaining the volume by adding hot water frequently. Afterwards, it was left to settle for 24 h with the flask kept in an inclined position to allow insoluble lignin to decant; which was filtered and washed with hot water to get rid of acid residuals until neutrality (pH 6-7). Afterwards, the insoluble residual was dried at 105°C in an oven for 4 h, cooled in a desiccator, and its mass was measured. The insoluble lignin content was calculated using Equation (4).

$$\text{Lignin \%} = \frac{M_L}{M} \times 100 \quad (4)$$

M_L = mass of dry insoluble lignin residual; M = mass of moisture and extractive-free sample

(e) Determination of holocellulose

The percent content of holocellulose in the fibres was determined using a protocol established by [27] with slight modifications using moisture-free and extractive-free samples. 2.5 g of the sample was mixed with 80 mL of hot water and the temperature of the mixture was maintained at 70°C in a hot water bath. 2.6 mL of 25% (w/v) NaClO₂ and 0.5 mL of glacial acetic acid were added after each hour for 6 hours. The resultant mixture was filtered and cleansed with distilled water until neutrality (pH 6-7). The white residual was dried at 105°C in an oven for 4 hours and allowed to cool in a desiccator for about 10 minutes. The mass of obtained white material which is holocellulose comprising of cellulose and hemicellulose was weighed. The percentage of holocellulose content was calculated using Equation (5).

$$\text{Holocellulose \%} = \frac{M_{hc}}{M} \times 100 \quad (5)$$

M_{hc} = mass of holocellulose; M = mass of moisture and extractive-free sample

(f) Determination of alpha-cellulose content

The percentage of alpha-cellulose content was determined as per TAPPI standard T203 cm-99 [28]. The standard defined alpha-cellulose as a fraction of wood that is resistant to 17.5% sodium hydroxide solution under testing conditions. To determine the percentage of alpha-cellulose content, 12 mL of 17.5% (w/v) NaOH was slowly added to 1.0 g of moisture and extractive-free holocellulose sample and the mixture was allowed to stand for 3 minutes followed by the addition of 10 mL of the solution and left to stand for 30 minutes. Thereafter, 30 mL of distilled water was added to the solution followed by magnetic stirring for 30 minutes to homogenize the solution. The resultant suspension was allowed to settle and filtered followed by washing using 8.5% (w/v) NaOH solution and 15 mL of 10% (v/v) glacial acetic acid and then twice with 300 mL of distilled water to eliminate any residual base present. The residual mass was dried at 105°C for 4 hours and left in a desiccator for about 10 minutes. Alpha-cellulose content was calculated using Equation (6).

$$\text{Alpha - cellulose \%} = \frac{M_{ac}}{M} \times 100 \quad (6)$$

M_{ac} = mass of alpha-cellulose; M = mass of moisture and extractive-free holocellulose sample

(g) Determination of hemicellulose content

The percent content of hemicellulose was calculated using Equation (7).

$$\text{Hemicellulose \%} = \text{holocellulose \%} - \text{alpha cellulose \%} \quad (7)$$

2.6 Evaluation of developed composites properties

Physical properties (specific gravity), thermal and hardness attributes of the composites were evaluated. Specific gravity of the samples was obtained using AU-300R Rock Density Tester. Hardness test was conducted on the specimen using a Shore D hardness tester. A load of 15 kgf was applied to each specimen with 25 s dwell time. Three values were obtained by indenting the samples in three distinct points and the average value was used for analysis. Thermal conductivity was examined to determine the flow of heat from one side of the polymeric material to the other using Holmarc Lee Disc Apparatus and in accordance with [18]. The thermal conductivity was calculated using Equation (8)

$$K = \frac{M c_p (T_1 - T_2) 4x}{\pi D^2} \quad (8)$$

where, K is the thermal conductivity; T₁, T₂ is the temperature of disk A and B in Kelvin; X thickness of the sample; D diameter of the sample; T₁ - T₂ initial and final temperature of disk; M is mass of the disk; C_p is the specific heat capacity of the disk;

3.0 Result and Discussion

3.1 Quantitative chemical analysis

Vegetable fibres are composed mainly of three main components: hemicellulose, cellulose and lignin [7, 29]. Each of the compositions of these components varies with each type of fibre. Cellulose is the most important component in plant fibres present in the cell wall. It provides strength to the fibres. Hemicellulose polymers are amorphous branched polymers that possess a considerably lower molecular weight compared to cellulose. They are connected to cellulose fibrils through hydrogen bonds. Due to their open structure with numerous hydroxyl and acetyl groups, they are partially water-soluble and hygroscopic and act as a compatibilizer between

cellulose and lignin [30, 31]. Lignin serves as a natural adhesive that holds the other components of plant fibres such as cellulose, hemicellulose, pectin and waxy materials together. The amorphous lignin matrix assists in binding the helically arranged cellulose microfibrils together, leading to the formation of composite fibres. It has the least water absorption ability among the natural fibre components and it helps to strengthen the stem of the plant against gravity forces and wind and provides protection against biological attacks [30]. However, for use as reinforcement materials, a high amount of cellulose is considered to enhance tensile strength, resistance to hydrolysis, crystallinity index, stiffness and thermal attributes[32]. Figure 2 shows the quantitative chemical analysis for banana and sisal fibres.

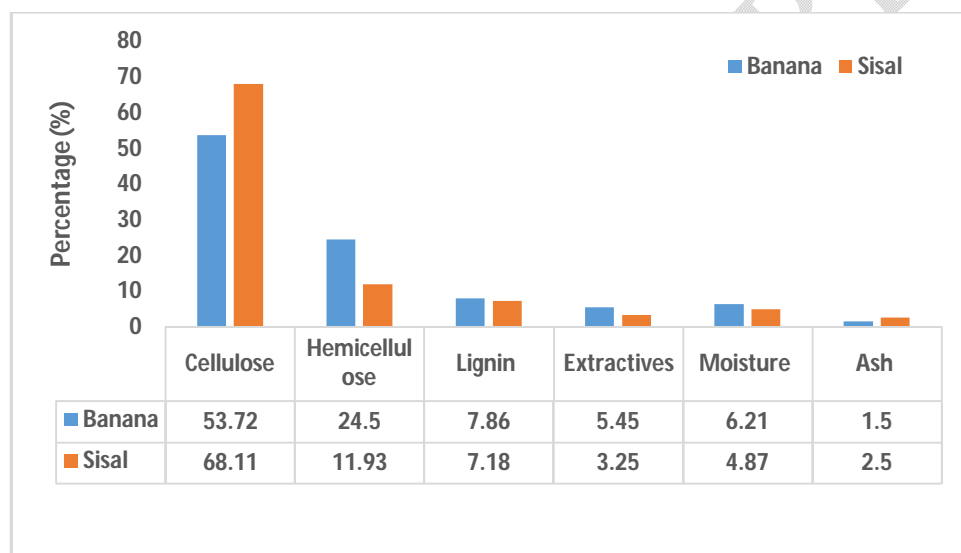


Figure 2: Quantitative chemical analysis for banana and sisal fibres.

From the result it was observed that the chemical constituents (cellulose, hemicellulose, lignin, extractives, moisture and ash for the banana fibres were 53.72, 24.5, 7.86, 5.45, 6.21 and 1.5% respectively while that of the sisal fibre was 68.11, 11.93, 7.18, 3.25, 4.87 and 2.5% respectively. It was seen that the cellulose content which is mainly responsible for strengthening is higher in

sisal compared to banana fibre. This result is similar to the reports in some earlier works [22,33,34].

3.2 Specific density

Figure 4 shows the result of specific density of the banana-sisal fibre hybrid reinforcement subjected to both soil and atmospheric degradation. From the results it was observed that the specific density of all the tested composites samples was between 1.0 – 1.2 g/cm³. The samples that were not subjected to any form of degradation had relatively higher density of 1.12-1.17 g/cm³ compared to those that were subjected to soil and atmospheric degradation of values 1.0 - 1.15 and 1.0 – 1.14g/cm³, respectively. The densities of polymer composites are related to porosity [35], therefore the actions of microbes in the soil, UV radiation, change in temperature, rainfall and relative humidity could have created pores in the composites which led to reduced density. From the results, it was also observed that the densities of both the composite samples subjected to soil and atmospheric degradation increased with increase in the time of exposure.

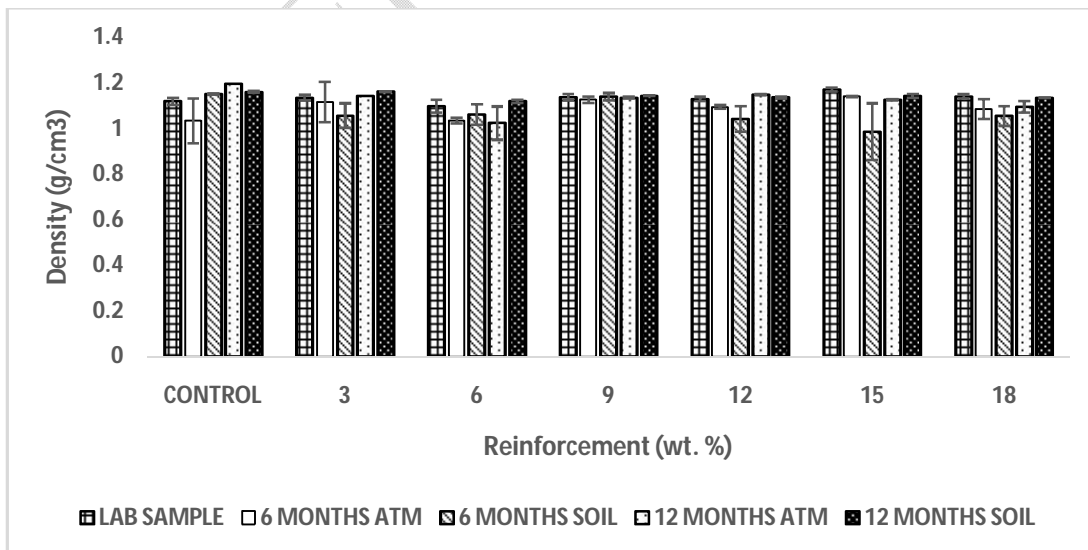


Figure 3: Variation of specific density of banana-sisal fibre reinforced epoxy composites subjected to degradation for 6 and 12 months

3.3 Hardness

The variation of hardness with the banana and sisal fibre reinforced composites for the individual fibres is shown in Figure 3. It was observed that the hardness of the composite samples increased for the samples exposed to 6 and 12 months environmental conditions compared to the ones that were not subjected to any form of degradation. However, for the composite samples subjected to soil burial, the hardness was lower compared to the reference i.e. the samples that were not subjected to degradation. About 6 – 10 % reduction in hardness was recorded. This indicated that the soil burial contributed to the reduction in hardness and this can be due to enzymatic action involving microorganisms [36] and the absorption of moisture present in the soil as stated by [37] that the water that surrounds the epoxy in a humid environment can lead to a reduction in its main characteristics. Equally, with reference to the time of exposure for the samples subjected to atmospheric degradation, the hardness of the composite samples increased with increase in time. Here, 3.4 – 5 % increment was observed. The 12 wt. % B/S composites subjected to atmospheric degradation for 12 months showed the highest hardness value of 71.67 HS. It can be inferred from this result that exposure to atmospheric degradation increased the hardness of the B/S composites and this is as a result of combined influence of temperature, humidity, UV radiation and pressure of the environment. Various studies have shown that exposure of polymer composites to UV and varying temperatures can cause polymer chains to rearrange and crystallize thereby inducing additional cross-linking within the epoxy matrix. This cross-linking process enhances the network structure of the polymer, leading to an increase in hardness [38,39].

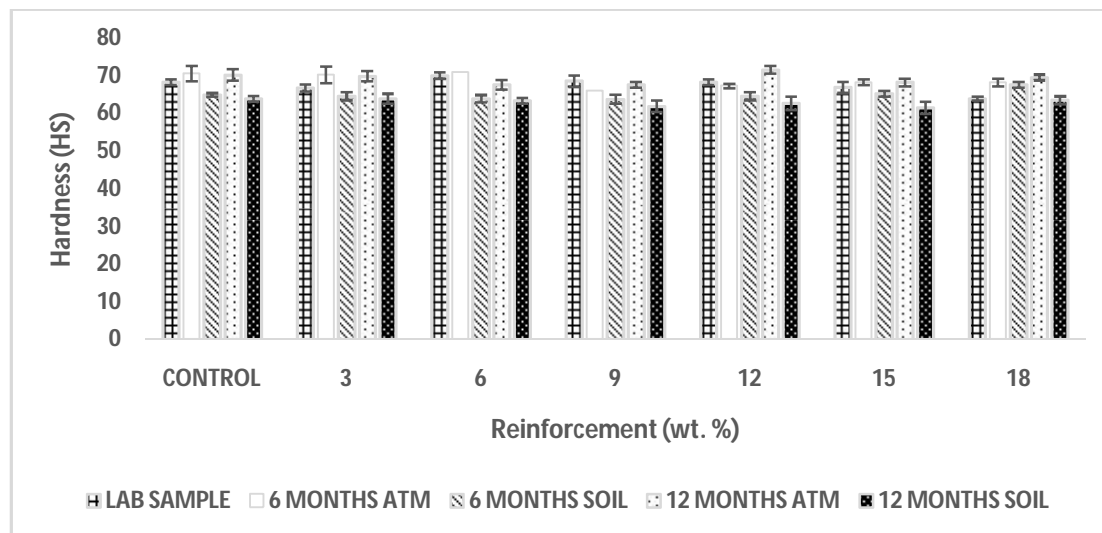


Figure 4: Variation of hardness of banana-sisal fibre reinforced epoxy composites subjected to degradation for 6 and 12 months

3.4 Thermal conductivity

The result of the variation of thermal conductivity with B/S reinforcement for composites subjected to degradation for 6 months and 12 months is shown in Figure 5 and the representative average values is shown in Figure 5. From the results, it was seen that the incorporation of banana-sisal fibre reinforcement into epoxy increased its thermal conductivity. The thermal conductivity of the reference sample increased with increase in the banana-sisal fibre content from 3 – 6 wt. % with values of 0.69 W/mK and 1.18 W/mK respectively and reduces gradually from 9 – 18 wt. %. A similar observation was noticed in work of [40] where they observed an increase in the thermal stability of the coir fibre-epoxy composite from 10 to 30 % weight loading. [41] also stated that the incorporation of fibre into epoxy matrix can improve its thermal conductivity. [13] also stated that the treatment of fibres could enhance their thermal conductivity when used as reinforcements. It was also observed that after exposure to degradation for the given period the thermal conductivity is within 0.1 to 0.8 W/m.K which has been reported

by several researchers [42,43,44,45] to be well suited for automobile parts made from natural fibre polymer matrix composite except 6 months SOIL. The thermal conductivity of the banana-sisal hybrid reinforced epoxy composites exposed to environmental degradation decreases with increasing exposure time. It was also observed that the thermal conductivity of the developed laboratory samples increased with increase in the weight % of banana-sisal fibres from 3 to 6 wt. % and thereafter decreases from 9 to 18 wt. %.

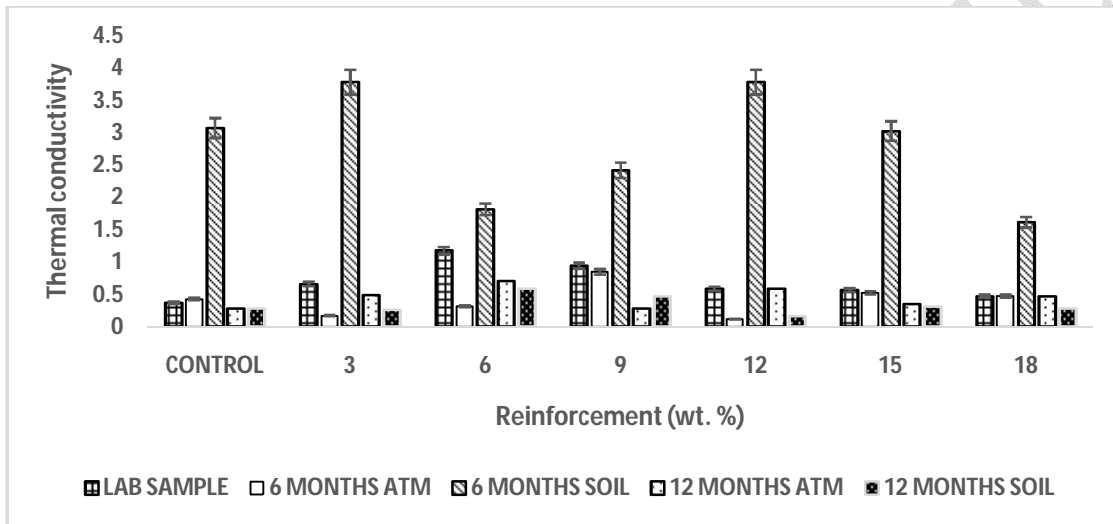


Figure 5: Variation of thermal conductivity of banana-sisal fibre reinforced epoxy composites subjected to degradation for 6 and 12 months

Conclusion

From the results, the following conclusions were made;

- a. The chemical constituents (cellulose, hemicellulose, lignin, extractives, moisture and ash for banana fibres was 53.72, 24.5, 7.86, 5.45, 6.21 and 1.5%, respectively while that of sisal fibre is 68.11, 11.93, 7.18, 3.25, 4.87 and 2.5%, respectively.

- b. The specific densities of all the composite samples subjected to environmental degradation were between 1.0 – 1.2 g/cm³ which were lower than the density of the reference sample. This reduction was as a result of the microbial action and the atmosphere on the composite samples, hence, the materials possess biodegradation characteristics.
- c. The hardness of the composite samples subjected to atmospheric conditions showed increase with increase in time of exposure while samples that were immersed in soil revealed reduction in hardness with time due to enzymatic activity in the soil and this implies that the composites subjected to soil burial showed more degradation.
- d. It was also discovered that most of the composite samples showed high resistance to thermal conductivity which is within the allowable thermal conductivity values for natural fibre polymer matrix composites automobile parts except the samples that was subjected to soil degradation within 6 months.

Ethical issues: None

Abbreviations

Lab Sample- Composite samples used as reference. They were not subjected to any form of degradation; **6 months ATM-** Composite samples subjected to atmospheric degradation for 6 months; **12 months ATM-** Composite samples subjected to atmospheric degradation for 12 months; **6 months SOIL-** Composite samples buried in soil for 6 months; **12 months SOIL-** Composite samples buried in soil for 12 months.

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