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Original Article

Development and Characterization of a Hybrid Macadamia Shell Particle and Sisal Fibre-Reinforced Composite Board

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Abstract *- The use of natural fibres and particulate agro waste in composite fabrication has gained significant importance in recent years due to their environment friendliness and cost-effectiveness. The physical characteristics of macadamia shells make them suitable for producing particleboards. Particleboards tend to absorb water, which compromises their integrity. The use of macadamia shell particulates can alleviate this problem by producing moisture-resistant particle boards. In this study, isophthalic polyester resin, sisal fibres and macadamia nutshells were used for the particleboard fabrication. Different ratios of the reinforcements, which include both macadamia shells and sisal fibre, were used during the fabrication, systematically varying the mass fraction according to the experimental design. From the experimental results it was observed that the tensile strength and flexural strength of the composite increased from 5 % up to 25 % sisal fibre mass fraction loading. The increase in tensile strength is found to be continuous up to 25 % sisal mass fraction with a maximum tensile strength of 146.4 MPa. The compressive strength showed an increase from 5 % to 25 % of macadamia particulate loading with maximum compression strength of 102.3 MPa at 25 % macadamia particulate loading. The composite sample with 15 % wt. macadamia to 15 % wt. ratio of sisal fibre exhibited high mechanical properties of 113.01 MPa, 98.6 MPa, and 106.1 MPa for tensile, compressive, and flexural strengths, respectively. The sample had 4.8 % moisture absorption and a burning rate of 4.8 mm/min.*

Keywords *- Composite, Macadamia shells, Mechanical properties, Sisal fibres.*

1. Introduction

Most food sector operations generate huge amounts of by-products, which are frequently treated as waste and disposed of in landfills. When the nut has been taken from the macadamia shell by the processing industries, the shell is mainly discarded. These waste macadamia shells are largely disposed of by incineration due to landfill disposal being prohibitively expensive due to their large quantity [1]. The physical characteristics of the macadamia shell make it suitable for the manufacture of moisture-resistant particle boards and panels as compared to softwood [2]**.** Boards made from macadamia nut shells can thus be suitable for use in panel furniture in damp settings, sink counters and drawers in kitchens or bathrooms, where dimensional swelling and adhesive issues may arise. The general name for a panel board made of lignocellulosic materials is particleboard [3]. These particleboards are typically made up of dry wood shavings that have been covered with resin and heated up to a wood-based particleboard [4]. A possibly less expensive alternative to solid wood paneling is to use the by-products from the processing of macadamia shells, which has also become a widely used alternative to wood in various applications [5]. Particleboards are commonly used in flooring, ceiling tiles, wall panels, door panel inserts, and for acoustic purposes [6]. Wood shavings are the main lignocellulose components utilized in the particleboard industry [7]. Wood shavings are widely used due to their abundance and low specific weight. Production of high-quality particleboard uses up much resin. Approximately 32% of the cost of manufacturing in the particle board composite sector is attributed to the resin [8]**.** Particleboards are made using a variety of resins, and depending on how they react to moisture and temperature, they are categorized as meeting requirements for internal or exterior use. The most widely used resins are modified formaldehyde condensation polymers, which include Urea-Formaldehyde (UF), Phenol-Formaldehyde (PF), Phenol-Resorcinol Formaldehyde (PRF), and Melamine-Formaldehyde (MF) [8]. The most used resins for composite wood products are urea and phenol-formaldehyde resins due to their low cost and well-established performance. However, they have some negative health impacts. Exposure to formaldehyde condensation polymers can cause chest

constriction, breathing difficulties, and skin and mucous membrane irritation. It is recognized that formaldehyde can potentially lead to the development of cancer cells in humans [9]. One of the most concerning challenges of particleboard use is its low resistance to moisture.

In the presence of moisture, most particle boards tend to swell and warp. Also, on exposure to moisture, particleboard tends to get discolored. If a particleboard is exposed to water, damage can occur due to the water travelling via capillary action inside the particleboard, weakening it.

This phenomenon makes it necessary to design a moisture-resistant board.The purpose of the study is to fabricate a composite board that can be used in high-moisture environments.

Furthermore, the developed composite board will use an alternative resin to the commonly used formaldehyde which is not environmentally friendly and is toxic.

2. Materials and Methods

2.1. Research Framework

The methodology of the study involved crushing the macadamia shells and sieving them for size uniformity. Chemical treatment of the macadamia shells and sisal fibres to be used in the composite was then carried out.

Thereafter, a composite was fabricated from the macadamia shells and sisal fibres in accordance with the experimental design. Lastly, the composite samples were tested to ascertain their mechanical properties. Figure 1 show the research framework used in this study.

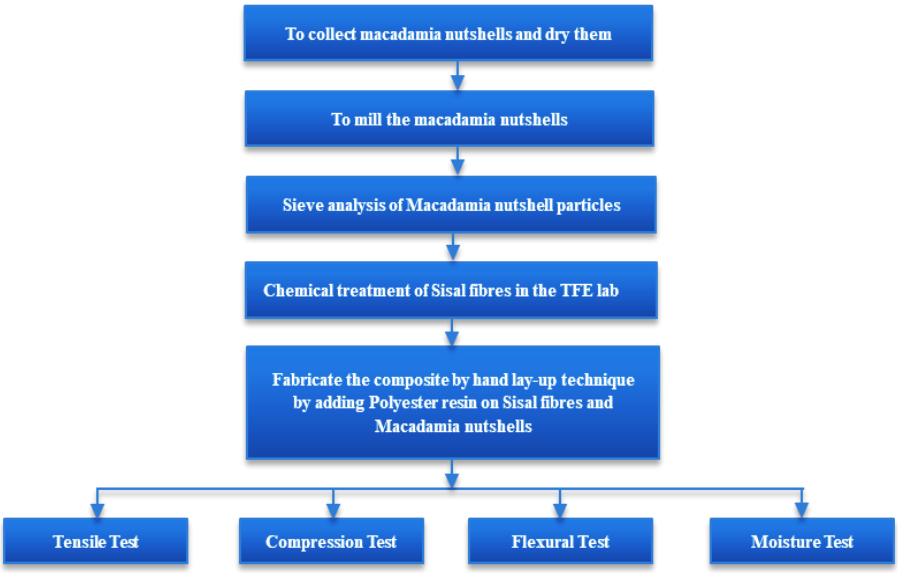

Fig. 1 Research framework

2.2. Raw Materials

The raw materials used in this study included macadamia nutshells and sisal fibres.

2.2.1. Macadamia Shells

The macadamia shells were ground and oven-dried at 100ºC to constant weight, then sieved in a 35-mesh sieve, as shown in Figure 2. The macadamia particles were then washed with warm hot until the wash water was clear. The nutshell particles were then oven-dried for 24 hours at 100°C.

The macadamia nutshell particles were treated with NaOH (4% w/v) for an hour under constant stirring at 70°C. Thereafter, the macadamia particles were washed with distilled water to neutralise the pH. The chemical treatment of the macadamia shells improves their dimensional stability, reduces water absorption, and increases resistance to microbial attack [10].

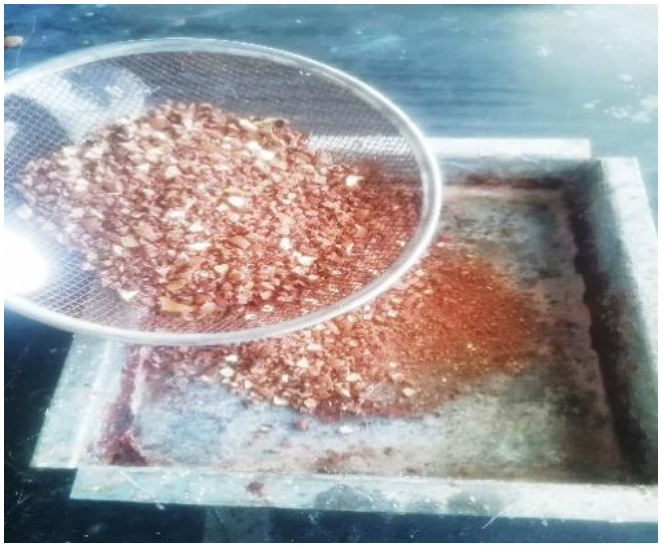

Fig. 2 Sieving of the macadamia shells

Fig. 3 Mercerized sisal fibres

2.2.2. Sisal Fibre Surface Treatment

Modification of the sisal fibre surface and internal structure was carried out by chemical treatment. The modification was done through the process of mercerization. 4% NaOH solution was used to soak the sisal fibres, activating the hydroxide (OH) groups of lignin and cellulose in the fibres.

The treatment with NaOH was carried out at room temperature for 24 hours. Thereafter, the sisal fibres were thoroughly rinsed with distilled water and left to sun dry for 24 hours. The dry mercerized fibres are shown in Figure 3.

2.2.3. Polyester Resin

The polyester resin NCS 901 PA was used as the matrix for the fabrication of a macadamia-sisal fibre-polyester composite.

2.3. Experimental Design

The composite was fabricated according to the experimental design shown in Table 1. The composite fabrication process was done through hand layup.

2.4. Characterization of Composite

Mechanical tests carried out on the fabricated composites include tensile, compression and flexural tests. Moisture absorption and flammability tests were also carried out.

Table 1. Experimental design followed in composite fabrication

Sample	Macadamia Nutshells Mass Fraction (%)	Sisal Fibre Mass Fraction (%)
1	15%	15%
2	5%	25%
3	25%	5%
4	10%	20%

2.4.1. Tensile Strength Test

The tensile test of the samples was done using the Testomeric Micro 500 machine in accordance with ASTM D638. The crosshead speed used was 3 mm/min.

2.4.2. Flexural Strength Test

The flexural strength of the hybrid composite samples was determined by subjecting the specimens under load in a three-point bending set-up in accordance with ASTM D790M standard with a dimension of 125 mm × 15 mm × 3 mm. The rate of the crosshead was 3 mm/min. The flexural strength was reported in MPa. The flexural strength of the samples was calculated using the formula below:

$$\sigma = \frac{3FL}{2bd^2} \quad (1)$$

Where F is the load at the fracture point, L is the length of the support, b is the width of the sample, and d is the thickness of the sample.

2.4.3. Compression Strength Test

The ASTM D3410 test method was used to determine the compressive properties of the macadamia/sisal/polyester composites. The compressive strength of composite materials was calculated using Equation 2.

$$\sigma c = \frac{Fc}{A} \quad (2)$$

Where σ_c is the compressive strength of the material, F_c is the maximum force or load sustained during a compression test, and A is the cross-sectional area of the sample.

2.4.4. Moisture Absorption Test

The water absorption test was carried out in accordance with ASTM 570 [11]. To measure the water absorption, the sample was submerged in distilled water. The percentage water absorption was calculated from the difference of final and initial weights before and after immersion in the water bath for 24 hrs, 48 hrs and 72 hrs. The testing was performed until the percentage of water absorptive reached equilibrium, as shown in Equation 3 [12].

$$Water\ absorption = \frac{Final\ weight - Initial\ weight}{Initial\ weight} * 100 \quad (3)$$

2.4.5. Flammability Test

The flammability test was carried out according to ASTM D635. The bar samples were supported horizontally at one end. The free end was exposed to a specified gas flame for 30 seconds. The burning time and extent of burning were measured and reported. The flame propagation speed and dripping properties were observed and noted. The horizontal burning test involved applying a 20 mm blue flame to the sample end clamped horizontally. The flame was applied for 30 seconds and removed if ignition resulted. The time taken by the flame to reach the second mark from the first mark (25 mm from the front end) was noted. The flame propagation speed of the sample was then calculated using Equation 4.

Fig. 4 Samples completely submerged in NaOH solution for chemical resistance test

$$V = (60L)/t \qquad (4)$$

Where V is the flame propagation speed (mm/min), L is the burned length (mm), and t is the time taken by the flame to reach the second mark.

2.4.6. Chemical Resistance Test

A chemical resistance test was carried out in accordance with ASTM D543. The resilience of the composites to the chemicals was examined in terms of weight loss after they were exposed to the chemicals. The composite samples were submerged, as shown in Figure 4. Three separate chemicals, an acid HCL, an alkali NaOH, and solvent benzene, were utilized for the current work. The composites were originally weighted during the experiment and submerged in the prepared chemical solutions for 24 hours at room temperature.

The specimens were removed from the water after 24 hours, dried, pressed, and then weighed. The proportion of weight loss/gain was by the following Equation 6.

$$Wc = \left(\frac{W1-Wo}{Wo}\right) * 100 \qquad (5)$$

Where W_c is the weight gain/loss percentage, and W_0 and W_1 are the weights of the specimen before and after immersion, respectively.

3. Results and Discussion

3.1. Sisal Fibre Mercerization

The mercerisation process of sisal fibres resulted in a colour change of the sisal fibres from white to yellow. The colour change is an indication of the removal of lignin and other contaminants on the sisal fibres. The treatment ultimately removed the impurities present at the surface of the fibre and enhanced the interfacial bond strength between the fibre and the matrix which ultimately improved the mechanical properties of the composites.

3.2 Effect of Macadamia Shell and Sisal Fibre on Tensile Strength

The results of the tensile strength testing are shown in Figure 5. The control, which is the Polyester resin NCS 901 PA composite, had a tensile strength of 76 MPa. There is an increase in the tensile strength value of the macadamia shell (M)/sisal fibre (S) reinforced polyester composite samples as compared to unreinforced polyester composite. An increase in values of tensile strength was recorded for the reinforced samples with a minimum tensile strength of 81.72 MPa and a maximum of 146.4 MPa for samples 3 and 2, respectively.

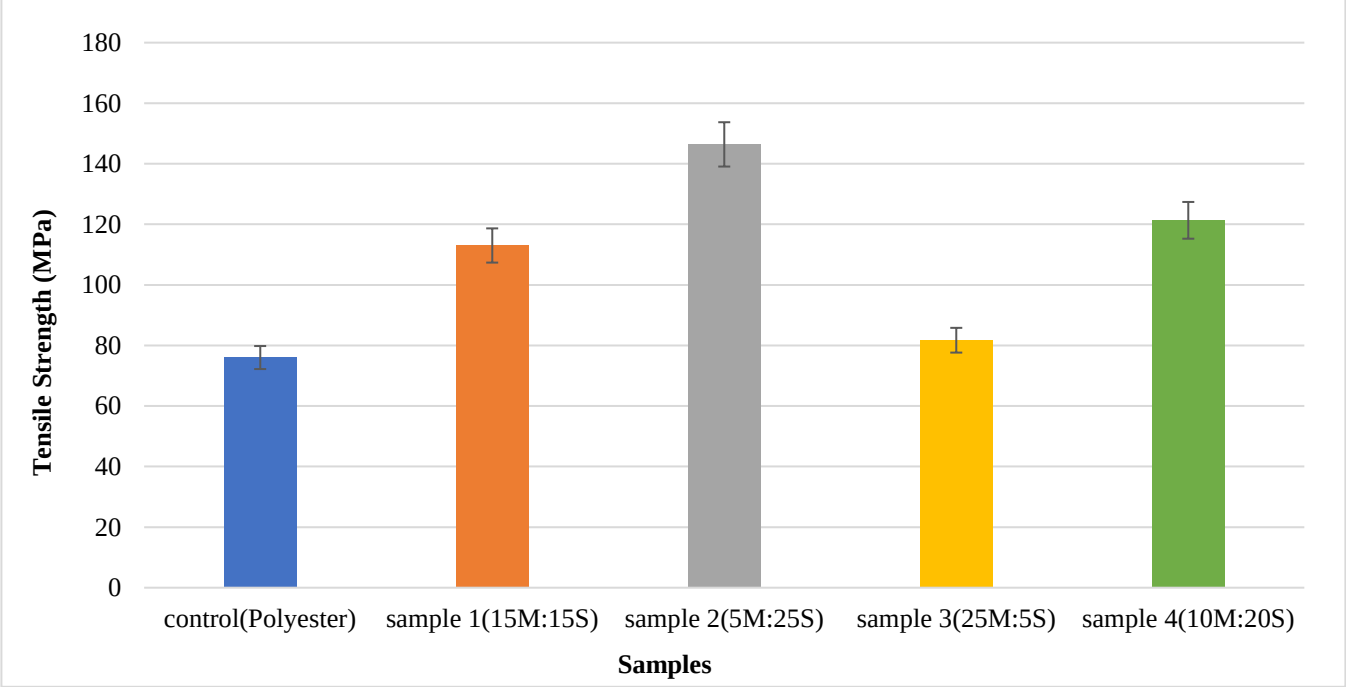

Fig. 5 Tensile **strength of macadamia/sisal/polyester composite samples**

Sample 2, with the ratio of most sisal fibres to macadamia particles (5M:25S), showed a greater tensile strength of 156.4 MPa, followed by sample 4, sample 1 and sample 3. The lower tensile strength of sample 3 as compared to other samples, could be attributed to the higher macadamia particle loading [13].

3.3. Effect of Macadamia Shell and Sisal Fibre on Compressive Strength

The bar graph in Figure 6 shows the effect on the compressive strength of macadamia shell particles and sisal fibre. It can be seen from the graph in Figure 6 that an increase in macadamia particulate content has a positive relationship with the compressive strength of the composites.

The compressive strength of unreinforced polyester resin NCS 901 is 76 MPa. Sample 3 had the highest compressive strength, up to 102.3 MPa, due to the higher macadamia particulate loading as compared to other samples. The findings show that the addition of the macadamia shell particulate significantly increases the compressive strength of the composites [14].

Figure 6 demonstrates that adding incremental amounts of sisal fibre has the effect of lowering the compressive strength of the composite [15]. In composites with high fibre loading, the failure mode of the composite was predominantly fibre kinking and buckling [16] [17].

3.4 Effect of Macadamia Shell and Sisal Fibre on Flexural Strength

Figure 7 shows the effect on the flexural strength of incremental amounts of macadamia shell particles and sisal fibre. The flexural strength of the fibre-reinforced polyester composite samples was higher than that of the pure polyester composite. The maximum value of flexural strength for reinforced polyester composite samples was reported to be 121.3 MPa for sample 2, with 5% wt. of macadamia particulates to 25 % wt. of sisal fibre.

This is an increment of 44.4 % over neat polyester resin. According to a study by [18], treated sisal fibre-reinforced polyester composites have better-bending properties and lower water absorption than untreated fibres.

Sample 3, with lower sisal fibre loading of 5 % wt., showed a lower flexural strength of 88.2 MPa, followed by sample 4, exhibiting 15 % wt sisal fibres, with 94.6 MPa and lastly, sample 2 exhibiting 25 % wt. sisal fibres, showing a high flexural strength of 121.3 MPa.

The increase in the values of flexural strengths of the composite samples was influenced by the increase in sisal fibre loading in the polyester matrix. The sisal fibres act as load bearing phases, transferring stresses from the matrix to the fibres, which have a higher strength and stiffness than the polymer matrix [19]. Sisal fibres can also improve the flexural strength of the polymer composites by providing a mechanism for crack deflection. When a crack propagates through the composite, it is defected by fibres, which prevent the crack from propagating further. This agrees with a study by Chauhan et al. (2016), which showed that crack bridging is particularly effective in composites with long and continuous fibres.

3.5. Water Absorption

Figure 8 shows the results of the water absorption test for samples that were soaked for 24 hrs, 48 hrs, and 72 hrs. The hydrophilic nature of the macadamia particles is what causes the weight gain to be higher during the first 24 hours [21]. However, the rate of weight gain tends to decrease with an increase in the immersion time of the samples, as shown in Figure 8.

During the 48-hour and 72-hour periods, slight changes in moisture absorption of samples are observed. The increase in moisture absorption with an immersion time of macadamia-reinforced composites has been supported by reports from the Ministry of Environment, Water and Natural Resources in Kenya and the US International Trade Commission [22] [23]; however, decrease in moisture absorption rate was observed for all the composite samples between 48 hrs and 72 hrs.

Samples with higher macadamia particle loading and less sisal fibres loading reported the highest water absorption rates. This is because they had more water residence sites in macadamia particles than in mercerised sisal fibres. Ashori (2009) [24] obtained similar results for Wood particle-based Plastic Composites (WPS).

On the other hand, the water absorption of treated sisal fibre composites is reduced. This could be attributed to the higher surface contact area of fibre with the matrix, which has less permeability of water than untreated fibre.

There is a notable transition of interest that can be clearly observed in Figure 8, where there is a great increase of 0.94 % in the moisture absorption of sample 3 at 25 % macadamia loading to 5 % sisal fibre loading for 24 24-hour immersion time. Thamae (2010) [25] observed this behaviour using a thermoplastic matrix and concluded that at higher filler loadings, there is a greater likelihood of particles creating agglomerates, which in turn increases moisture absorption.

The idea has since been advanced that there is a "critical macadamia fibre content" at which agglomerates form into clusters that act as channels for water to travel through the particles [26]. It is at this critical point that the composite water uptake increases rapidly to levels higher than those observed for all other samples. This behaviour could be due to the increased difficulty in mixing at higher filler loadings leading to agglomerates and fibre exposure.

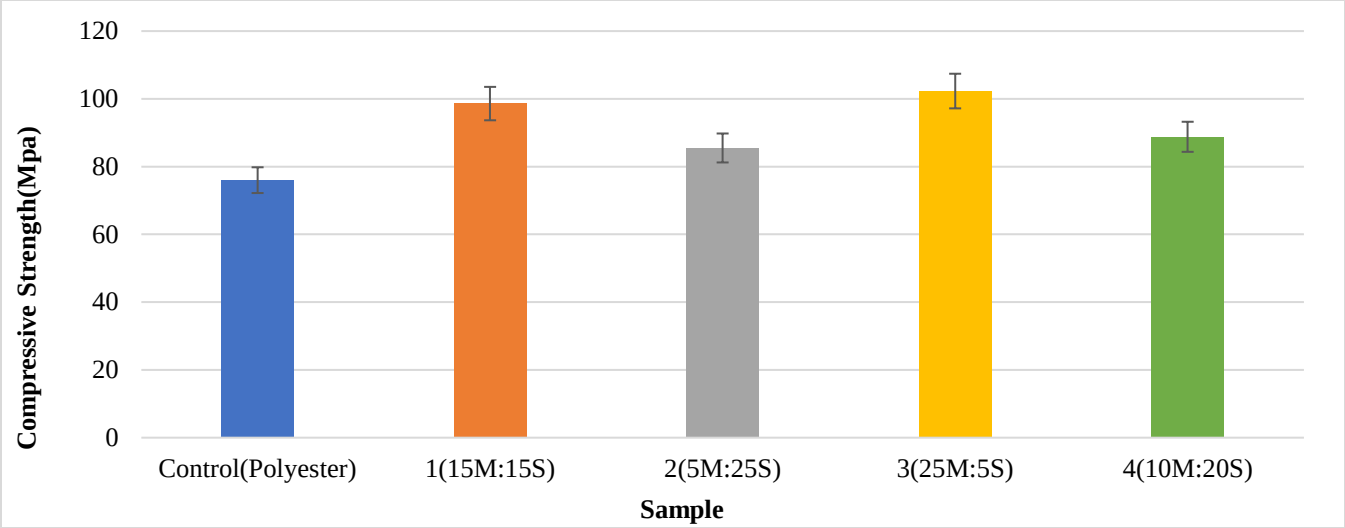

Fig. 6 Compressive strength of samples

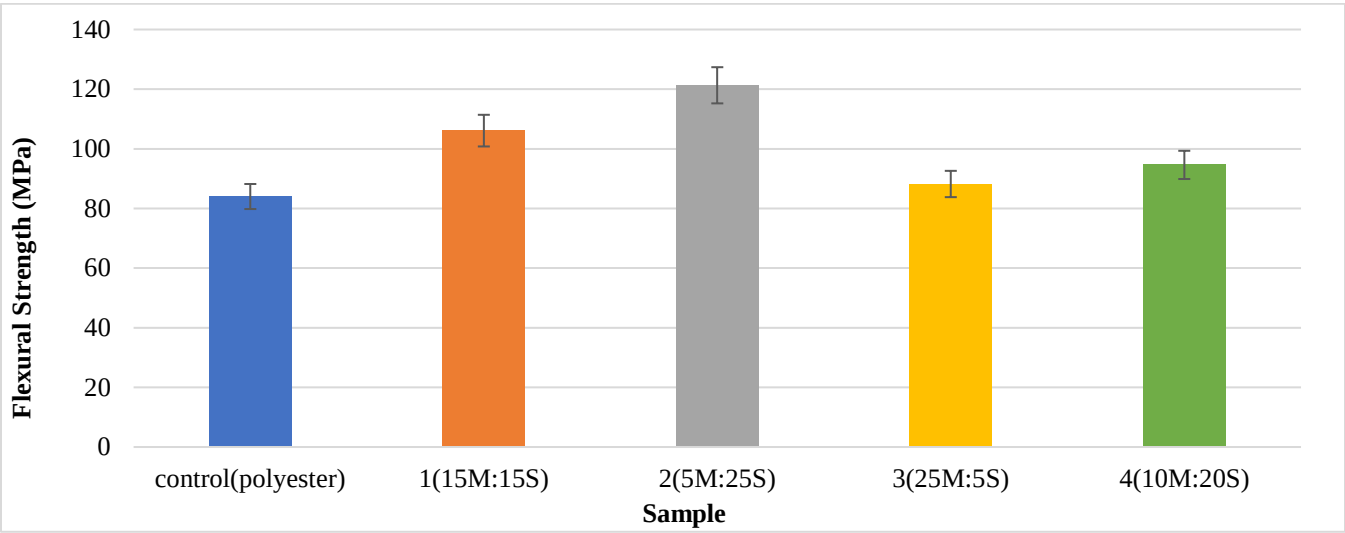

Fig. 7 The flexural strengths of fabricated samples

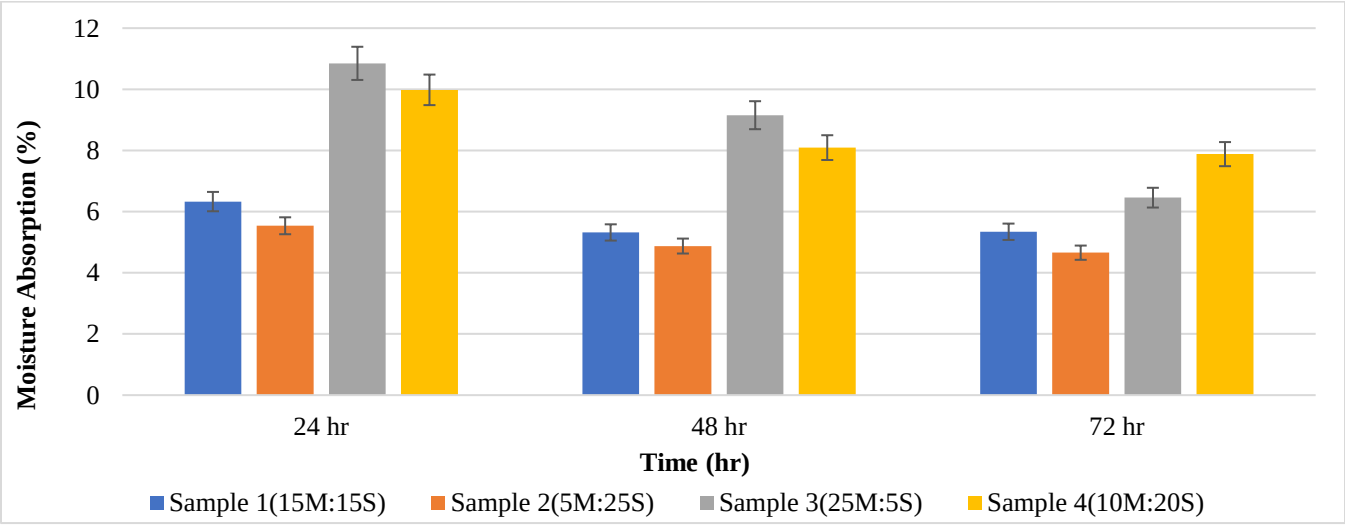

Fig. 8 Moisture absorption percentage for macadamia/sisal/polyester composites for 24 hrs, 48 hrs and 72 hrs.

3.6. Flammability

Figure 9 shows the results of the burning rate test on the composite samples. The control polyester sample burned rapidly at 12.1 mm/min, and it was observed to drip as it burnt. The sample burnt smoke-free; this could be attributed to the aliphatic hydrocarbon structure. The fabricated samples showed a decrease in the burning rate when compared to the control polyester, as shown in Figure 10. The lowest burning rate of 4.8 mm/min was shown by sample 3 with higher macadamia particulate loading as compared to the other samples. The macadamia particles have a high lignin content [27] that produces a dense char layer when burnt. This dense char assists in fire resistance. In this case, during the burning process, the high macadamia loading produces a dense char layer that becomes the flame barrier between the fire and the polymer matrix [28]. This developed layer is an eco-friendly way of reducing combustion with low smoke production.

Char formation is the key to achieving low flammability; this is because char is formed at the expense of possible flammable fuel. Furthermore, due to the char locking in the available carbon, less smoke is produced, and the char acts as a barrier to its release [29]. Additionally, the char acts as an insulating layer and protects the composite minimising the loss in tensile properties during fire exposure [30].

It was observed that sample 3, with higher macadamia particulates to sisal fibre ratio, took more time to ignite and reach the 100 mm mark. The higher the distribution of the macadamia particulates in the composite, the more it inhibits the rapid burning of the polyester resin. Hence more time is needed for ignition and to reach the 100 mm mark. No dripping behaviour was noted during the burning of the composites; yellow flames and black soot were observed during the burning of all the reinforced samples.

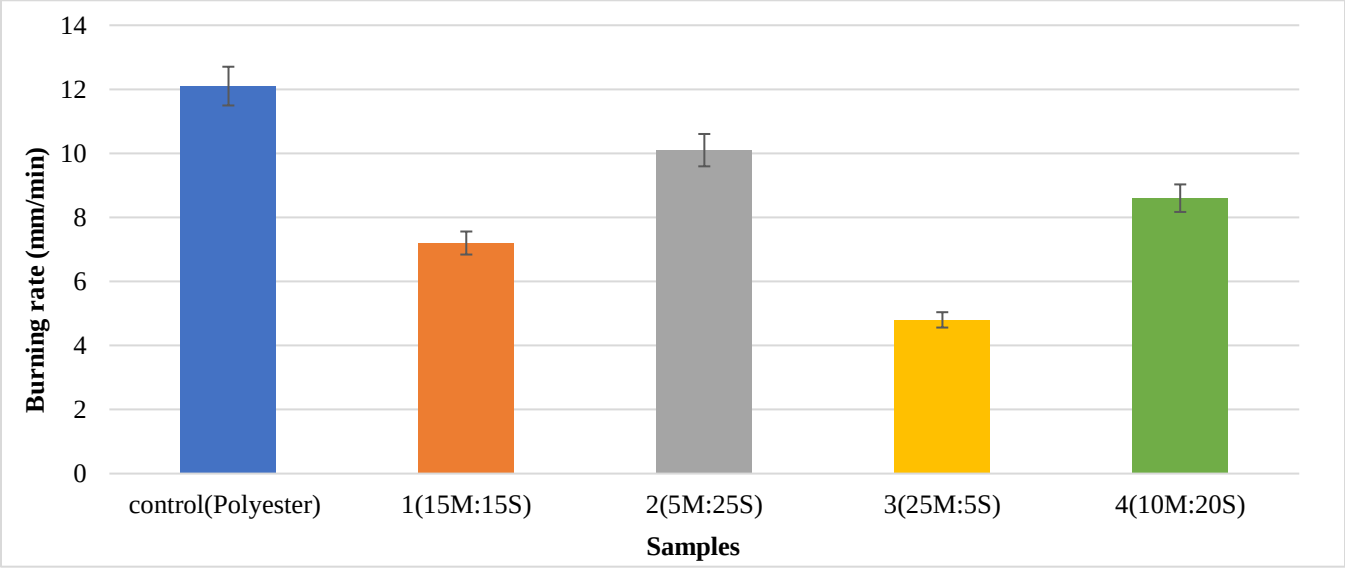

Fig. 9 Burning rate of samples in mm/min

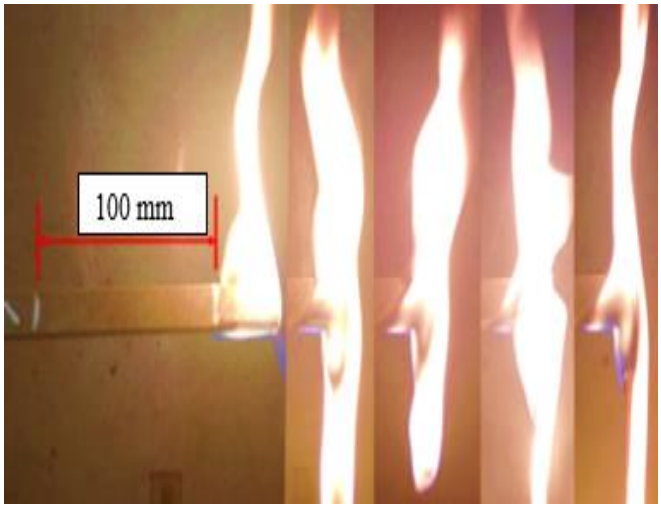

Fig. 10 The combustion process of macadamia/sisal/polyester sample during UL-94 horizontal burning test.

3.7. Chemical Resistance

The results of the chemical-resistant test are shown in Figure 11. All the composite samples gained weight after the chemical resistance test and no weight loss was noted on any samples. This indicated that these composites had resistance to chemical attacks. Higher weight gain was recorded on the benzene solvent, followed by the acid, HCL and lastly NaOH. The increase in the weight can be attributed to the hydrophilicity nature of the macadamia particulate. Compared to the benzene treatments on the samples, HCL treatments and NaOH treatment composites showed lower weight gain percentages on all samples. Sample 2, with the lowest particulate loading, showed the lowest percentage weight gain of 4.38 % when exposed to NaOH reagent for 24 hours. The maximum percentage weight gain was observed for sample 3, with 7.59 % weight gain; this is because of the highest macadamia particulate ratio loading of 25 % wt. as compared to the 5 % wt. of sisal fibres.

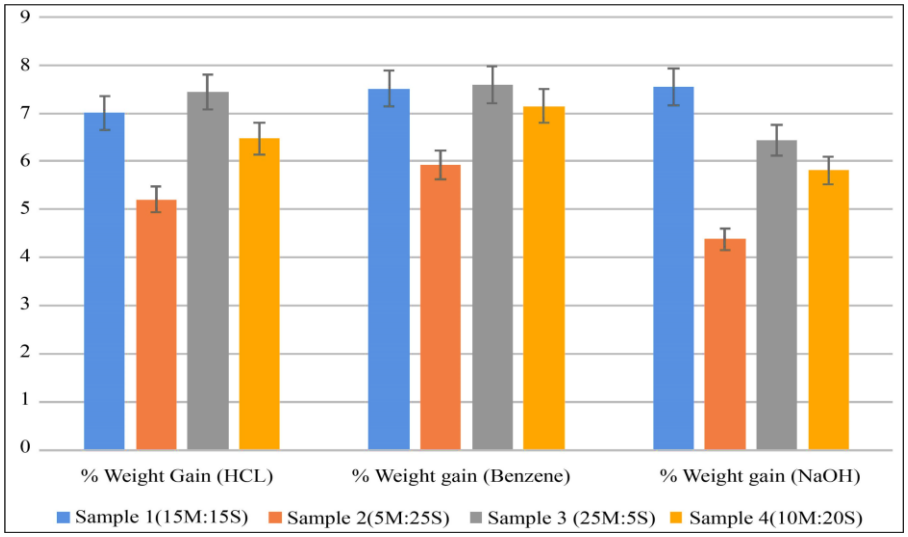

Fig. 11 The weight gain % of samples for the chemical test for different reagents

The hydrophilic nature of the macadamia particles is what caused the weight gain to be higher during the first 24 hours. A study done by Cipriano (2019) [21], supports the weight gain percentage of polymer composites to be directly related to the hydrophilic nature of the macadamia particles. This is because they had more water residence sites in macadamia particles than in mercerised sisal fibres. Ashori (2009) [24] obtained similar results on Wood Plastic Composites (WPS). Overall, all the developed composites showed better resistance to the chemical attack.

4. Conclusion

The developed composite material exhibited good mechanical properties and showed favourable moisture absorption properties and flammability properties. The conclusions reached in this study are:

- The macadamia/sisal/polyester composite samples exhibited substantially lower water absorption as compared to conventional wood particleboards, with maximum water absorption of 11.84 % shown by sample 3 during the 24 hrs, whilst conventional particle boards had water absorption of 34 % [20]. Mercerized sisal fibres 'natural hydrophobic qualities, the hydrophobic nature of macadamia nut particles, and the water resistance of polyester resin combine to improve dimensional stability and reduce moisture-induced damage.
- The increase in macadamia particle loading increased the compressive strength and decreased the tensile strength of the composite. Sample 3 showed the maximum increase in compressive strength up to 102.3 MPa due to the higher macadamia particulate loading as compared to other samples. An appreciable increment of 34.6 % when compared with the unreinforced neat polyester composite sample. An increase in values of tensile strength was recorded for the reinforced samples with a minimum tensile strength of 81.72 MPa and a maximum of 146.4 MPa, for samples 3 and 2, respectively, with sample 2, with the ratio with most sisal fibres and lowest macadamia particulate loading (5M:25S) showed a greater tensile strength of 146.4 MPa.
- A great increase of 10.8 % moisture absorption of sample 3 at 25 % wt. macadamia loading to 5 % wt. sisal fibre loading for 24-hour immersion time was clearly noted. A slight increase of 5.5 % moisture absorption was observed for sample 3 with a ratio of 5 % wt. macadamia particulate to 25 % wt. sisal fibres. The slight increase of 5.5 % in moisture absorption, shown by sample 3, implies that the composite can be used in applications with high moisture and can replace wood particleboards with moisture absorption ranging from 5 to 10 % moisture absorption.
- The composite samples showed good chemical resistance. This chemical resistance test showed that the composites are highly resistant to all the tested chemicals, and a maximum weight gain of 7.59 % was noted on benzene treatment. These observations suggest that these composites can be used in chemical erosion resistance applications.
- Flexural strength maximum value of 121. 3 MPa was achieved by sample 2 with a ratio of 25 % wt. sisal fibres to 5 % macadamia particles. It is shown from experimental results and calculation that flexural modulus increases with the weight fraction of macadamia nutshell particles. Adding macadamia nutshell particles does not improve the flexural strength of polyester, but the addition of sisal fibre does. The flexural strength shown by all the samples exhibited good properties, which are suitable for use in particleboard construction. The minimum flexural strength for standard-grade particleboard is 31 MPa (4500 psi) for boards with a density of 0.50 g/cm³ (31.2 lb/ft³) or greater and 27 MPa (3900 psi) for boards with a density less than 0.50 g/cm³ (31.2 lb/ft³) and greater than or equal to 0.42 g/cm³ (26.2 lb/ft³).

- Fabrication of the macadamia/sisal/polyester composite showed the possibility of utilization of macadamia nutshell waste, which is underutilized locally by macadamia processing companies; hence, their disposal to the environment is mitigated. A favorable macadamia/sisal/polyester composite particleboard, of 15 % wt. macadamia to 15 % wt. sisal fibre ratio was obtained with the optimum requirements of both the mechanical and physical properties needed for use in high moisture environments.

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