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## Investigation into mechanical, absorption and swelling behaviour of hemp/sisal fibre reinforced bioepoxy hybrid composites: Effects of stacking sequences

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#### Abstract

This work focuses on the fabrication of hybrid bio-composites using green epoxy as the matrix material, hemp (H) and sisal (S) fibre mats as the reinforcements. The hybrid composite with sisal/hemp fibres were fabricated by cost effective hand lay-up technique, followed by hot press with different stacking sequences. Static properties of the composites such as tensile, compressive, inter-laminar shear strengths (ILSS) and hardness were examined. The physical properties such as density, void content, water absorption and thickness swelling were also analysed. The experimental results indicate that hybrid composites exhibited minor variation in tensile strength when the stacking sequence was altered. The hybrid composite with the

intercalated arrangement (HSHS) exhibited the highest tensile modulus when compared with the other hybrid counterparts. Hybrid composites (SHHS and HSSH) offered 40% higher values of compressive strength than the other layering arrangements. HHHH sample exhibited the highest ILSS value of 4.08 MPa. Typical failure characteristics of the short beam test such as interlaminar shear cracks in the transverse direction, micro-buckling and fibre rupture were also observed.

**Keywords:** Green epoxy, hemp and sisal fibres, tensile and compressive strengths, inter-laminar shear strength, water absorption, thickness swelling.

#### 1. Introduction

The growing mandate for green products in the field of composites has narrowed the use of synthetic materials in many engineering applications. The development of completely or partially biodegradable composites can be well-defined based on the nature of their ingredients. A major changeover can be witnessed all over the world on the utilisation of biodegradable polymers and natural fibres for the fabrication of composite materials. This could be attributed to the mounting concerns on safeguarding the environment against the ever-increasing pollution by the synthetic non-biodegradable plastic products [1,2]. Furthermore, the disposal of used plastics is unavoidable and their stacked form can create numerous environmental issues. On the other hand, the recycling of used plastics is expensive and time-consuming. Hence, it becomes inevitable to develop environmentally friendly products in all possible means to reduce environmental pollution [3]. In addition to the ecological perspective, the suitability of biodegradable polymers and natural fibres for many applications has increased their demands. They also have ability to perform better than their synthetic counterparts considering many factors: environmental, cost and production, among others. Hence, many industrial sectors such

as automotive, construction, packaging, electronics and even biomedical industries are focusing on employment of environmentally friendly products.

Synthesis of glucose based epoxy resin was carried out by Nierdermann et al. [4] targeting its potential use as composite structures for air crafts. Bio-resin based on furfuryl alcohol and quebracho tannins was synthesized by Lagel et al. [5] for prospective replacement of the automotive brake pads. Similarly, epoxidised hemp oil based bio-resin was prepared by Manthey et al. [6] for structural applications. Novel bio-based resin from the soybean oil was synthesized by Hong et al. [7] for electronics, automotive and aeronautical applications. Lignin-based bio-resin was used in composites applications by Stanzione et al. [8]. Other types of biodegradable polymers matrix systems such as poly(lactic acid) (PLA) [9], poly(propylene carbonate) (PPC) [10], cellulose [11], Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) [12], among others, were also used as matrix systems in bio-composites for different applications.

However, the mechanical, physical and thermal properties of these materials are relatively low compared to their synthetic counterparts and several measures have been taken to enhance their properties. One of the promising methods to enhance the properties is the reinforcement of natural fibres. Natural fibres are used in different forms: short, continuous and randomly oriented, among others. Recently, the woven fabric mats are gaining more interest and are considered as the most attractive due to their outstanding integrity and conformability for advanced structural applications [13]. Some researchers have demonstrated the enhanced functional properties of the composites with woven fabric form of reinforcement from the natural fibres when compared to a short and randomly oriented form of reinforcements [14]. Another method of enhancing the performance is the hybridisation of fibres to form hybrid composites. In

this case, the properties of the hybrid composites mainly depend on some factors, such as the properties of the individual fibres used, the compatibility between the fibres and the matrix as well as the surface characteristics of the fibres [15]. The benefit of hybridisation is that the advantage of one fibre can be used to overcome the shortcoming of the other fibre resulting in balanced performance and sustainability of the composite. Considering the performance of a material, hybridisation of two different woven fabrics with the polymer matrix could be more desirable.

Some researchers have investigated the combining effect of reinforcing different natural fibres to form hybridized natural fibre composites. Alavudeen et al. [1] studied the effect of hybridisation on the mechanical properties of banana/kenaf hybrid composites. They reported that the hybridisation of kenaf with banana fibres enhanced the mechanical strength when compared with the individual fibre based composites. Similarly, another report says that the hybridisation of sisal with oil palm improved the mechanical properties of the composites [16]. Some researchers have investigated the effects of hybridisation on mechanical properties of banana/sisal composites. They reported that an increase in sisal content in the composite increased the mechanical properties up to 50% [17]. Boopalan et al. [18] studied the effects of different weight ratios of jute and banana fibres on mechanical and moisture absorption properties of jute/banana fibre reinforced epoxy composites. Their study revealed that the tensile, flexural and impact strengths were found to be maximum for 50:50 weight ratio of jute and banana fibres in the hybrid composites. Moisture absorption study of hybrid composite showed a minimum moisture uptake for the same 50:50 hybrid composites. Effects of stacking sequence of woven jute/glass fabric reinforced isophthalic polyester composites were experimentally

investigated. It was reported that altering the position of the glass plies greatly affected the flexural and inter-laminar shear properties [19].

Moreover, many natural fibres such as kenaf, hemp, flax, jute, sisal, banana, coir and pineapple leaf fibres, among others, are drawing more considerable attention as a green reinforcement in the formulation of composite materials [20]. Sisal is the most widely used fibre in the automotive and construction industry. This could be attributed to the exceptional mechanical characteristics and hence, it is used to manufacture automotive car parts and concrete structure [21]. Similarly, hemp is a plant-based natural fibre available as lignocellulosic reinforcing material in composites. It is used for different structural applications and its importance in the biocomposite applications can be evinced through the European Industrial Hemp Association (EIHA) Hemp conference organised every year to exchange information on the recent developments in automotive and construction, textiles, food, food supplements and pharmaceuticals industries using hemp fibres.

Hemp and sisal fibre mats have been used separately with different polymer matrices in the formulation of composites, but the combination of both hemp and sisal fibre mats in a bio-resin based hybrid composites has not been explored till date. Hence, this research aims to fabricate bio-hybrid composites using green epoxy as the polymer matrix and reinforcing hemp and sisal fibre mats in different layering sequences. The effect of different layering sequences on the mechanical properties such as tensile, compression, inter-laminar shear strength and hardness were investigated. Furthermore, the thickness swelling, water absorption behaviour, density, void content and percentage (%) weight reduction of the hybrid composites with different stacking sequences were also studied.

#### 2. Materials and methods

### 2.1 Materials

Bidirectional woven hemp and sisal fibre mats were supplied by Nirmala Industries, Hyderabad, India. Green epoxy (SR Greenpoxy 56<sup>®</sup>) produced with a high content of carbon from plant origin. The hardener (SD Surf Clear<sup>®</sup>) was supplied by Sicomin Epoxy Systems, France. The properties of the green epoxy resin and both hemp and sisal fibre mats are presented in Tables 1 and 2.

## Table 1

Properties of the green epoxy matrix and the hardener

Properties	Green Epoxy resin	Hardener
Aspect/colour	<u>Clear liquid</u>	Clear liquid
Density @ 20 $^{\circ}$ C (g/cm <sup>3</sup> )	1 108	0.958
$V_{i}$ as a size (m $P_{i}$ s) (20 $^{\circ}C$	1.178	60
Viscosity (mPa.s) @ 20 C	1400	60
Refractive Index @ 25 °C	1.535	-
Bio based carbon content (%)	50-58	0

## Table 2

Properties and composition of hemp and sisal fibre mats [22–24]

Properties	Hemp fibre	Sisal fibre
Density (g/cm <sup>3</sup> )	1.48	1.50
Tensile Strength (MPa)	690	511-635
Young's Modulus (GPa)	70	9.4 - 22.0
Elongation at break (%)	1.6	2.0 - 2.5
Cellulose content (%)	74.4	65.8
Hemicellulose (%)	17.9	12.0
Lignin (%)	3.7	9.9
Microfibrillar angle (°)	6.2	20.0

2.2 Preparation of hybrid composites

The green epoxy matrix was made by mixing the green epoxy (SR Greenpoxy 56<sup>®</sup>) resin and the hardener (SD Surf Clear<sup>®</sup>) in the ratio of 100:37 by weight. The hemp and sisal fibre mats were layered according to the designated layering sequence shown in Fig. 1. They were impregnated with green epoxy resin in the mould with dimensions of 20 x 20 cm<sup>2</sup>. The mould was left to cure at 100 °C for about 1 h in a hot press at a constant pressure of 275 bars. Post curing was performed at 100 °C for about 10 minutes after removing from the mould. In addition to the pure green epoxy matrix, composites with pure hemp and sisal fibre mats were prepared for comparison of their properties. The different layering sequences of the hemp and sisal fibre mats used for the fabrication of the hybrid composites and abbreviations for the designated layering sequences are presented in Fig. 1.

Fig. 1. Different layering sequences of hemp and sisal fibre mats.

#### 2.3 *Experimentation*

#### 2.3.1 Tensile test

The tensile strength and modulus of the composites were measured using a universal testing machine (INSTRON 3382). The test was carried out in accordance to the ASTM D3039 standards. The crosshead speed and gauge length were set to 1.27 mm/min and 50 mm, respectively. For each layering sequence, five identical specimens with dimensions of 120 x 20 x  $3 \text{ mm}^3$  were tested, and the average values are reported. For instance, and better understanding, the specimen sample before and after the tensile test is presented in Fig. 2.

**Fig. 2.** Tensile specimen (a) before testing; (b) after testing, (c) tensile test set-up.

## 2.3.2 Hardness test

Shore D hardness tests were performed using a Rex Durometers, Model OS-1. It was supplied by Total Innology Co., Ltd, Rayong, Thailand. The composite specimens were fabricated according to the ASTM D 2240-86. The indenter has the diameter of 1.27 mm, and the

maximum applied load of 5 kg. The test was carried out in an ambient temperature condition. The hardness values were taken 10 seconds after ensured indenter had a firm contact with specimens. For each configuration, the hardness values are taken at ten different locations on the upper surface of the composites, and the average values were reported.

#### 2.3.3 Compressive test

The compressive test was performed in accordance to the ASTM 695 standard, using a universal testing machine (INSTRON 3382). For each layering sequence, five identical specimens with dimensions of  $20 \times 20 \times 3 \text{ mm}^3$  each were tested with a crosshead speed of 1.5 mm/min and the average results were reported.

#### 2.3.4 Inter-laminar shear stress test

Inter-laminar shear strength (ILSS) test was carried out with a small beam of 24 mm length loaded under three-point bending at a rate of 1.27 mm/min. The maximum load recorded from the beam was used to calculate the ILSS, using Eq. (1).

$$\sigma = \frac{3P}{4bh} \tag{1}$$

where,  $\sigma$  represents the inter-laminar shear strength, *P* denotes the maximum load in kN, *b* and *h* are the width and height of the specimen in mm. For each stacking sequence, five identical specimens were tested, and average results were reported.

#### 2.3.5 Water absorption

The water absorption test was performed in accordance to the ASTM D570 standard, and the percentage of water absorption was calculated from the Eq. (2).

Water absorption (%) = 
$$\frac{W_n - W_d}{W_d}$$
 (2)

where  $W_n$  stands for the weight of the composite samples after immersion and  $W_d$  represents the weight of the composite samples before immersion. The hybrid composite samples were immersed in de-ionised water at room temperature. The initial weights of the samples were recorded before immersion into the water, and the weights of the samples immersed in the water were recorded at different time intervals. After the samples were removed, they were gently rubbed with filter paper to remove the excess water on the surface, and the weights of the samples of the samples were recorded using 4 digits weighing balance. The water absorption test was continued for several days until a constant weight of the samples were obtained.

#### 2.4 Characterisation

#### 2.4.1 Microscopic analysis

The morphological analysis of the hybrid composites was carried out using a scanning electron microscope (SEM). For this study, a FEI, Quanta 450 SEM was used.

#### 2.4.2 Void content

Void contents for the composites were calculated according to the ASTM D 2734-70. Then, Eq. (3) was used to calculate the theoretical density  $(T_d)$ , while measured densities  $(M_d)$  were used to calculate the void content by using the Eq. (4).

$$T_d = \frac{100}{\left(\frac{R}{d_m} + \frac{r}{d}\right)} \tag{3}$$

where,  $T_d$  represents the theoretical density (g/cm<sup>3</sup>), R denotes the weight % of the green epoxy matrix in the composite,  $d_m$  stands for the density of the green epoxy matrix (g/cm<sup>3</sup>), r is the weight % of the reinforcement in the composite and d denotes the density of the reinforcement (g/cm<sup>3</sup>).

$$V = \frac{100 (T_d - M_d)}{T_d}$$
(4)

where, V is the void content (%),  $T_d$  represents the theoretical density (g/cm<sup>3</sup>), and  $M_d$  stands for the measured density (g/cm<sup>3</sup>). The percentage weight reductions of the matrix, all the composites include same fibre composite and hybrid samples with different stacking sequences were also reported.

#### 2.4.3 One-way ANOVA analysis

Statistical analysis of the measured tensile, compressive and inter-laminar shear strength properties of the different composite configurations were carried out with the one-way analysis of variance (ANOVA) using Origin Pro 8 software. The analysis was performed at a 5% significance level.

#### 2.4.4 Thickness swelling

In order to measure the swelling of the composites, thickness swelling test was performed in accordance to the ASTM D570 standard. The thickness of each sample was measured before their soaking in distilled water. Thickness of the specimens was measured at different time intervals, and the test was continued for several days until constant thickness values of the composites were obtained. The percentage of thickness swelling was calculated from the Eq. (5).

Thickness swelling (%) = 
$$\frac{T_1 - T_0}{T_0}$$
 (5)

where,  $T_o$  and  $T_1$  represent the thicknesses before and after soaking, respectively.

#### 3. Results and discussion

#### 3.1 Mechanical properties

#### 3.1.1 Tensile properties

In order to investigate the influence of the different layering sequences on the tensile properties, the tensile test was carried out and the results obtained are presented in Table 3.

#### Table 3

Pattern of composites	Tensile strength (MPa)	Tensile modulus (MPa)	Elongation at break (%)	Shore D hardness
Green epoxy	25.67±1.53	904.82±04.03	2.20±0.00	81.62±1.12
HHHH	32.00±1.39	1158.95±46.22	2.25±0.09	80.13±0.89
SSSS	30.70±0.60	968.97±70.37	5.28±1.45	80.77±0.70
HSSH	30.24±0.73	1095.51±26.64	3.28±1.03	79.28±0.82
SHHS	30.76±1.17	1095.00±43.64	3.63±2.44	81.01±1.02
HHSS	30.00±1.23	1028.41±60.19	6.85±2.47	81.42±0.81
HSHS	31.76±0.88	1173.47±32.97	3.26±0.41	80.56±0.83

Tensile properties of sisal/hemp green epoxy hybrid bio-composites.

Tensile strengths of the hybrid composites varied from 30 - 32 MPa and did not show a regular pattern with the alteration of stacking sequence. Hybrid composites have equal number of hemp and sisal fibre layers and were subjected to uniform load and equal deformation during the tensile test. In hybrid composites, the merits of one type of fibre can be balanced with what is deficient in the other fibre providing them with the enhanced properties [22,24]. However, in this study, there were no significant changes in the tensile strength due to hybridization. Similar observations were reported in a recent study on hybrid composites with the banana/coir, sisal/coir and sisal/banana and their tensile strength ranged between 38 – 43 MPa [25]. In another study, tensile properties of polypropylene based hybrid composites with coir and jute fibres were analyzed by varying the fibre weight in the ratio of 50:50 and 25:75. The hybrid composites had tensile strength in the range of 20 - 22MPa and the changes in tensile strength between different configurations were insignificant [26].

The tensile modulus of pure hemp and pure sisal fibre composites were recorded to be 1158.9 MPa and 968.97 MPa, respectively. It can be noticed that the pure hemp fibre reinforced composites (HHHH) showed higher values than the pure sisal fibre reinforced composites (Table 3). This could be attributed to the superior stiffness of the hemp fibre and effective load transfer between the fibre and matrix through the strong interfacial bonding. According to Baley [27], the elasticity of a fibre decreases with higher micro-fibrillar angle, while it progresses linearly with higher cellulose content. It is clear from Table 2, that the hemp fibre possesses lower micro-fibrillar angle of 6.2° and higher cellulose content of 74.4% when compared to the sisal fibre. Hence, HHHH possessed higher tensile modulus.

It can be observed from Table 3 that the pure sisal fibre reinforced composites (SSSS) exhibited the least tensile modulus values. However, hybridizing the sisal fibre with the hemp fibre (HSSH, SHHS, HHSS and HSHS) resulted in a slight increase in the tensile modulus. Among the hybrid composites, HSHS showed highest tensile modulus. The difference in fibre diameter between the hemp and sisal fibre is believed to have promoted better compatibility between the fibres, hence slightly higher tensile modulus for the hybrid composites.

#### Fig. 3. Tensile stress-strain plot of the green epoxy hybrid-composites

Fig. 3 illustrates the tensile stress-strain behavior by varying the fibre stacking sequences on sisal/hemp green epoxy hybrid bio-composites and the results were shown in Table 3. The brittle nature of green epoxy is evident from the lowest value of the percentage elongation at break. Furthermore, it can be observed from Fig. 3 that an improvement in the percentage elongation at break can be observed after reinforcing the matrix with the fibres. This enhancement could be ascribed to reduced brittleness of the green epoxy matrix with the fibre reinforcement. Owing to

the intrinsic high elongation property of the sisal fibre, sisal fibre reinforced composites exhibited higher values than the pure hemp fibre composites [28]. On the other hand, hybrid composites produced improved values of percentage elongation at break This is in accordance with the fact that the lower elongation property of one of the fibres is compensated by the presence of other fibre with greater elongation capability in a hybrid composite [29].

The mean values of shore D hardness of fibre reinforced composites with different stacking sequences are given in Table 3. It can be observed that the hardness of the green epoxy matrix was higher than the rest of the composites. This could be attributed to the hard and brittle nature of green epoxy. When the fibre mats are reinforced into the green epoxy, the moisture present in the fibre cell wall could have acted as a plasticizer, decreasing the hardness of the fibre reinforced composites. The decreasing hardness values with the reinforcement of fibres into the matrix was also observed in a recent study on the kapok/sisal hybrid composites [30]. In another study, shore A hardness of hybrid composite with coir and jute fibres was found to between 97 - 99 and no significant changes in magnitude was observed from the hardness test [26].

## 3.1.2 Compression properties

To study the effect of stacking sequences on the compressive properties of the hybrid composites, the compressive test was carried out. The compressive stress-strain plot obtained is presented in Fig. 4a. It is evident from Fig. 4 that the composites exhibited relatively higher compressive strength and strain than the pure green epoxy matrix. This is attributed to the presence of fibres in the composites that offered resistance to the applied compression load and imparted greater load bearing capability. The fibres also help in provide ductility to the composite as observed by the increased strain values on failure compared to the green epoxy matrix.

Fig. 4a. Compression stress-strain plot of the green epoxy hybrid bio-composites

Fig. 4b. Compression properties of the sisal/hemp green epoxy hybrid bio-composites

The compressive strength and modulus extracted from the plot is illustrated in Fig. 4b. Pure hemp and pure sisal reinforced composites recorded superior compressive strength and modulus than other hybrid counterparts. The inferior compressive properties for the hybrid composites could be due to the poor compatibility between the sisal and hemp fibres. Hybrid composites exhibited substantial difference in the strength and modulus when the hemp and sisal fabric was arranged in the different stacking sequences. Among the hybrid composites, sisal/hemp in the outer layer/core arrangement (SHHS) and vice-versa (HSSH) had nearly 40% higher compressive strength than the composites with intercalated arrangements (HHSS and HSHS). The decline in compressive strength for the intercalated arrangement could be attributed to the inefficient stress transfer between the sisal and hemp due to the fibre-fibre interaction [31].

#### 3.1.3 Inter-laminar shear strength properties

Table 4 summarizes the ILSS results obtained from the short beam test for the hybrid and non-hybrid composites. It can be observed that the composites with the fibre reinforcement exhibited nearly 2 to 3 times higher magnitude of ILSS than the pure matrix.

#### Table 4

Inter-laminar shear properties of sisal/hemp green epoxy hybrid bio-composites

Composite	ILSS (MPa)
Green epoxy	1.40 <u>+</u> 1.34
НННН	4.08±0.33

SSSS	3.39 <u>+</u> 0.44
HSSH	3.37 <u>±</u> 0.22
SHHS	3.15 <u>±</u> 0.14
HHSS	3.27 <u>±</u> 0.18
HSHS	3.48±0.26

This indicates that sisal and hemp fibres acted as a stress transfer medium and played a significant role in resisting the shear load within the matrix. It can be observed from Table 4 that HHHH possessed the maximum ILSS value of 4.08 MPa. Some lower ILSS values were obtained for the HSSH, SHHS and HHSS hybrid composites when compared with HHHH sample. This suggests that sisal and hemp fabric had less compatibility within the matrix and varying the stacking sequence did not improve the interfacial bonding characteristics. Decrease in ILSS due to the differences in interfacial bonding characteristic between the flax and sugar palm fibres was highlighted by Chandrasekar et al [32]. In their study, they highlighted that the pure flax fibre reinforced epoxy composite outperformed the hybrid composites.

Figs. 5 (a)-(d) present the SEM micrographs of the fractured specimens from the short beam test. Both the hybrid and non-hybrid composite specimens showed typical failure characteristics, such as inter-laminar shear cracks in the transverse direction, micro-buckling and fibre rupture. In addition to these failure modes, normal cracks can be observed in Figs. 5(b) and 5(c). Composites subjected to short beam tests are expected to have normal cracks, since the inter-laminar shear failure does not always occur at the mid-plane of the laminate [19].

Table 5 presents the results obtained on the statistical analysis from the one-way ANOVA. P-value for the tensile, compressive and inter-laminar shear strength properties were less than 0.05 at 5% significance level. This signifies that null hypothesis is invalid and there is a statistically

significant difference in the population means between the different composite configurations or stacking sequences.

#### Table 5

Properties	Degree of Freedom	Sum of Squares	Mean Square	F-value	P-value
Tensile strength	5	16.24124	3.24825	2.98131	0.03123
Tensile modulus	5	150766.43954	30153.28791	12.55111	0.00000
Compressive strength	5	2553.88614	510.77723	22.21634	0.00000
Compressive modulus	5	1.16210	0.23242	4.52677	0.00477
Inter-laminar	5	2.69230	0.53846	6.92634	0.00039

One-way ANOVA analysis of the mechanical properties of hybrid bio-composites.

3.1.4 Void content, density and weight reduction

It is well known that the trapped air or volatiles exist in the composites during the impregnation of fibre into the matrix or during the fabrication of composites [33,34]. During the fabrication and curing process, resin flow through the fibre layer influences: (i) void content, (ii) fibre distribution, and (iii) fibre wetting in the composite. These factors could affect the properties and have consequence on performance of the composites [35]. Therefore, the mechanical and physical properties of the composites are expected to decrease due to the presence of the voids. The values of measured density, theoretical density, % weight reduction of the composite from the matrix and % of voids of composites with different stacking sequences are shown in Table 6. Among the investigated composites, SSSS presented the lowest void percentage.

## Table 6

Pattern of composites	Measured density (g/cm <sup>3</sup> ) M <sub>d</sub>	Theoretical density (g/cm <sup>3</sup> ) T <sub>d</sub>	% Weight reduction of composite from the matrix ((d <sub>m</sub> - M <sub>d</sub> )/d <sub>m</sub> )×100	% void content ((T <sub>d</sub> - M <sub>d</sub> )/T <sub>d</sub> )×100
Green epoxy	1.194	-	19.334	-
НННН	1.150	1.271	22.267	9.46
SSSS	1.177	1.264	20.443	6.84
HSSH	1.087	1.267	26.586	14.26
SHHS	1.129	1.267	23.701	10.89
HHSS	1.060	1.267	28.378	16.36
HSHS	1.113	1.267	24.786	12.16

Density, void and % weight reduction of sisal/hemp green epoxy hybrid bio-composites.

Density of green epoxy  $d_{m=} 1.198 (g/cm^3)$ 

It can be observed from Table 6 that hybrid composite with sisal fibre on the skin presented comparatively lower void content than the HSSH. The variation in void content with respect to the stacking sequence of fibre layers in the hybrid composite could be associated to the higher percentage of weight reduction of composite from the matrix. Hence, it is clear that void content varied between 10 -16% for the hybrid composite due to the differences in geometry (diameter and lumen size) and morphology of the fibre reinforcements.

#### 3.2 Water absorption

The natural fibre reinforced composites are susceptible to moisture absorption for numerous reasons: (i) viscosity of the matrix, (ii) fibre fraction, (iii) voids and (iv) humidity and temperature [36]. The water absorption characteristic of sisal/hemp hybrid composites presented in Fig. 6 shows a linear increase in the initial days of soaking in water. This is because the water

molecules quickly diffused into the micro-voids existing in the composites. Upon increasing the soaking period, the water uptake by the sisal/hemp fibre composites reduced and finally reached a state of saturation. Among the fibre reinforced composites, SSSS composite had the lowest water absorption of nearly 7% after 35 days followed by the HHHH composites. This could be ascribed to the lower percentage of cellulose and hemicellulose contents of sisal fibre than the hemp fibre. Lesser the cellulose and hemicellulose contents of a natural fibre, higher is their resistance to water absorption [37,38]. Furthermore, the water absorption of a natural fibre reinforced composite is mainly influenced by their porosity, fibre-matrix adhesion, void percentage and lumen size [39]. Besides these factors, hydroxyl groups present in the fibres tend to absorb moisture due to their hydrophilic nature.

HSHS, SHHS, HHSS and HSSH composite specimens showed higher water absorption than the pure fibre composites, especially at the saturation stage. It is evident from Table 6 that hybrid composites have more voids and porous surface than the pure fibre reinforced composites. Therefore, the water molecules might have quickly diffused into the voids and led to weight gain. However, the water absorption of all the hybrid composite specimens was below 12% until saturation. Higher weight gains due to the moisture absorption for the hybrid composites compared to the composites without hybridization or pure fibre reinforced composites were reported in the literature. The weight gains due to water absorption increased in the order: pure sugar palm fibre composite < roselle/sugar palm-based hybrid composite [40] and pure sugar palm/thermoplastic sugar palm starch agar matrix composites < seaweed/sugar palm fibre hybrid [41].

#### 3.3 Thickness swelling

In an attempt to examine the effects of moisture diffusion on the thickness of the composite specimens, thickness swelling test was conducted. The results obtained are presented in Fig. 7. It can be noticed from Fig. 7 that HHHH and SSSS exhibited higher dimensional stability, while the hybrid composites showed a higher rate of thickness swelling. However, the rate of thickness swelling reduced when the stacking sequences of hybrid composites were changed.

Among the investigated composites, HHHH exhibited the highest dimensional stability and the HSHS showed the highest thickness swelling. Similar to water absorption (Fig. 7), the hybrid fibre composites showed more thickness swelling. High porosity and presence of voids on the surfaces of hybrid composites could be responsible for the changes in their dimensional stability. Contrary to the hybrid composites, HHHH composite was assumed to have lesser pores, fewer voids and better fibre-matrix bonding which in turn led to reduced thickness swelling. This finding is in agreement with the previous work by Radzi et al. [40]. In their study, they showed that pure roselle/polyurethane composites exhibited better dimensional stability than the roselle/sugar palm fibre/polyurethane hybrid composites.

#### 4. Conclusions

The effect of stacking sequences of sisal and hemp in the green epoxy matrix on the mechanical, physical and water absorption properties have been experimentally investigated. The concluding remarks of this study are as follows:

• The least tensile strength was recorded for the green epoxy matrix at 25.66 MPa and the highest value was found to be 31.997 MPa for HHHH, followed by 31.762 MPa for HSHS composite. A similar trend was also observed in case of the tensile modulus. The lowest value was recorded with the matrix at 904.8 MPa and the highest at 1173.47 MPa for HSHS, followed by HHHH hybrid composite.

- For the compressive strength, the green epoxy matrix possessed the lowest value of 22.46 MPa, and the highest value of 47.47 MPa was recorded for HHHH followed by SSSS with 41.088 MPa, SHHS with 37.30 MPa and HSSH with 36.78 MPa. Similar trend was seen in the compressive modulus. The least compressive modulus of 0.94 GPa was recorded for green epoxy matrix and the highest modulus of 1.892 GPa for HHHH followed by other configurations.
- Green epoxy matrix had the lowest ILSS value of 1.4 MPa and HHHH exhibited the highest value of 4.08 MPa followed by HSHS, SSSS and HSSH. It can be established that stacking sequences of the hybrid composites influenced their ILSS properties.
- Even though, there were some improvements in the mechanical properties of the hybrid composites when compared to the green epoxy matrix, there were no great significant effects on varying the layering sequence of the fibre mats. The pure fibre or non-hybrid composites exhibited superior mechanical properties than the hybrid counterparts. This could be due to the poor compatibility between sisal and hemp fibres as well as poor interfacial bond/strength of the hybrid sisal-epoxy-hemp fibres reinforced composite samples.
- The ANOVA analysis showed that the null hypothesis is invalid and there is a statistically significant difference in the population means between the different composite configurations or stacking sequences.
- The pure fibre or non-hybrid composites exhibited better resistance to water absorption and thickness swelling when compared to the hybrid counterparts. Similarly, this can be attributed to poor fibre compatibility and interfacial bond strength of the hybrid composite. However, the weight gain of all the hybrid composites was below 12% during the entire test

period and the rate of thickness swelling reduced when the stacking sequences of hybrid composites were changed.

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## **Figures and Captions**

Fig. 1. Different layering sequences of hemp and sisal fibre mats.

Fig. 2. Tensile specimen (a) before testing; (b) after testing, (c) tensile test set-up.

Fig. 3. Tensile stress-strain plot of the green epoxy hybrid-composites

Fig. 4a. Compression stress-strain plot of the green epoxy hybrid bio-composites

Fig. 4b. Compression properties of sisal/hemp green epoxy hybrid bio-composites.

**Fig. 5.** SEM Micrographs of the fractured composite specimens from the short beam test performed on (a) HSHS, (b) HHSS, (c) SSSS and (d) HSSH samples.

Fig. 6. Water absorption of sisal/hemp green epoxy hybrid biocomposites.

Fig. 7. Thickness swelling of sisal/hemp green epoxy hybrid biocomposites.

Sont

## **Highlights**

- Hybrid biocomposites with varying the stacking sequences of hemp and sisal fibre mats in green epoxy matrix was fabricated.
- Effect of varying sequences on the static mechanical properties such as tensile, compression and inter-laminar shear strength were investigated.
- Significant improvements were seen on the tensile and compressive modulus of the composites when compared with the matrix.
- Statistical analysis was performed by One-way ANOVA method.

Sont of Contraction o



(d)





(C)



- b SSSS
- c HSSH
- d SHHS
- e HHSS
- f HSHS



(f)

Figure 1





Figure 3





(b)





Figure 6



Figure 7