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RESEARCH ARTICLE

FLEXURAL BEHAVIOUR OF HYBRID EPOXY COMPOSITES REINFORCED BY RAMIE FIBRE AND COFFEE BEAN PARTICULATE

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Abstract. Natural fibre polymer composites (NFRC) have always been the focus of researchers due to their excellent properties and advantages. In recent years, environmental concerns have drawn attention to bio-based material composites. Despite extensive research on natural fibre reinforced polymers, the study of their mechanics is still in its infancy. Ramie fibre is a natural fibre that has been continuously tested to explore its great potential because of its excellent mechanical properties. Biodegradable waste, such as coffee bean particulate waste, has high potential for use as polymer reinforcement to improve the properties of composites. Therefore, in this study, the hybrid epoxy composite of ramie fibre and coffee bean particulate aims to enhance flexural behavior. Ramie fibre epoxy composites were fabricated with different fibre orientations of 0°/0° and 0°/90°, as well as different weight fractions of coffee bean powder at 5 wt%, 10 wt%, and 15 wt%. Fabrication was conducted using the VARI method. Three-point bending tests were carried out to evaluate the flexural properties. The results showed that flexural strength was maximized (29.945 MPa) at a fibre orientation of 0°/0° with 10 wt% of coffee bean powder. The fracture mechanism was investigated using scanning electron microscopy (SEM). The analysis revealed that the fibre-matrix interface exhibited defects such as delamination, matrix microcracks, fibre fracture, voids, and fibre pull-out. These failures indicate poor adhesion between the fibres and the matrix.

Keywords: Ramie fibre, coffee bean powder, flexural properties, natural fibre polymer composites, material performance.

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1. INTRODUCTION

As environmental problems become more serious, bio-based materials have become very important in caring for the environment. Bio-based materials provide an important new market opportunity for domestically grown biomass resources. Due to the "green" advantage of using biomass instead of non-renewable petroleum-based raw materials, bio-based materials have gained momentum in specific market sectors. Another key benefit of switching to bio-based materials is the potential to reduce pollutants, especially greenhouse gases, which are emitted into the environment from fossil chemical processes. Some of these pollutants are dangerous and toxic and have short and long-term effects on human health and safety [1-3]. Recently, scientists have been studying the recycling and processing of bio-based waste into materials with better performance.

Natural fibre is a renewable and sustainable material that is widely considered an environmentally friendly alternative for various applications [4,5] . Among these, ramie fibre is a commercially available natural fibre known for its ability to retain shape, resist wrinkling, and impart a silky luster to fabrics. However, due to its high molecular crystallinity, ramie fibre tends to be stiff and brittle, making it prone to breakage when repeatedly folded at the same location. On the other hand, biodegradable polymers offer a promising solution to reduce the environmental impact caused by conventional synthetic polymers. One such biodegradable material is coffee bean particulate waste, a natural by-product that can be used as a filler in epoxy resin composites. Incorporating this waste into epoxy resins can improve and enhance their mechanical properties [6]. These bio-based composite materials are particularly attractive for engineering applications due to their low weight, high specific strength, stiffness (modulus), and corrosion resistance [7].

A hybrid composite is a material made by combining two or more different types of fibres into a common matrix [8]. The method of manufacturing the hybrid epoxy composite can be done by using a vacuum assisted resin infusion technique. From a scientific point of view, flexural strength indicates the resistance of a material against deformation. The flexural test is an important test method to define the flexural behaviour of composites. The test helps to determine the application suitability and life expectancy of the product as well as the safety of the user. The ability of the material to resist damage when bending and applying a load will be discussed. For this paper, standard 3-point bending tests will be employed to determine the flexural properties of the composites. The primary objective of this study is to investigate the flexural strength of hybrid epoxy composites that are modified with the incorporation of ramie fibre and coffee bean particulate and also, the study will analyse the fracture mechanisms that occur within the composites during flexural testing, aiming to reveal the roles played by ramie fibres and coffee bean particulates in crack initiation, propagation, and failure modes

2. MATERIALS AND METHODS

2.1 Materials

Bisphenol A Diglycidyl Ether (DAGBE) was employed as the epoxy resin, while 4-aminophenyl sulfone (97%) and triethylenetetramine served as curing agents for the fabrication process. Acetone was utilized as the cleaning agent during the study. The reinforcement materials used in the composites were ramie fiber and coffee bean powder, with the coffee powder having a particle size distribution of less than 200µm. To investigate the impact of reinforcement weight fraction on the flexural strength of the composites, samples were prepared with ramie fibers oriented at 0°/0° and 0°/90°. Additionally, coffee bean powder was incorporated in weight fractions of 5 wt%, 10 wt%, and 15 wt%.

2.2 Fabrication of Hybrid Epoxy Composite

In this study, hybrid epoxy composites were fabricated using the vacuum-assisted resin infusion (VARI) technique. This technique is an integrated system that utilizes vacuum pressure to drive epoxy resin into a laminate. To produce the hybrid epoxy composite, the viscous liquid form of epoxy resin, DAGBE, with an epoxide equivalent weight of 172–176, was mixed with the curing agents 4-aminophenyl sulfone and 97% triethylenetetramine. Coffee bean powder was ground into smaller particle size distributions of less than 200μm, and the desired weight fractions of 5 wt%, 10 wt%, and 15wt% were measured and stacked in the vacuum system. Ramie fibers were layered, and the mixture of epoxy resin, curing agent, and coffee bean powder was infused through the resin inlet.

The coffee bean powder was dried in an oven at 60 °C for 8 hours, then stored in a container with silicone gel to prevent moisture absorption. It was then ground with a pestle and mortar to reduce particle size further. Bisphenol A diglycidyl ether (DAGBE) was used as the epoxy resin to prepare the igecomposite materials, while 4-aminophenyl sulfone and 97% triethylenetetramine were employed as hardeners [9] . The epoxy resin and curing agent were mixed in a ratio of 6:4. Coffee bean powder was added to the epoxy resin according to specified weight fractions. The epoxy resin mixture was stirred slowly and at a consistent speed to avoid gas bubbles forming in the mixture. Figure 1 shows the sample preparation of epoxy-ramie fibre.

Figure 1: Epoxy-ramie fibre with fibre orientation 0°/90°

Ramie fibers with dimensions of 30 cm × 30 cm were utilized as reinforcements for the composite. Two layers of ramie fiber were fabricated using the VARI technique with varying fiber orientations and weight fractions of coffee bean powder. During this process, it was essential to ensure the entire system was vacuumed. Leaks could allow air to enter the vacuum system, resulting in gas bubbles within the composite. To address air leakage, the vacuum pump was activated for a few minutes after the system was fully set up. The pointer on the pressure gauge was checked to ensure it was at zero; if not, the vacuum bagging film and sealant were inspected to confirm a proper fit. The location of leaks could also be identified by listening for gas leakage sounds. The epoxy-ramie fiber composite was cured in the vacuum system for 24 hours. After curing, the composite was removed, and the countertop was cleaned with acetone.

2.3 Flexural Testing

The three-point bending test was conducted to measure the force required for a beam under three-point loading conditions. A SHIMADZU AG-10KN X-PLUS Universal Testing Machine was used to perform the bending test on the hybrid epoxy composites. The test followed the ASTM D7264 standard, which is the standard test method for determining the flexural properties of polymer matrix composite materials. A variety of specimen shapes could be used for this test, but the standard specimen thickness was 4 mm, and the specimen length was approximately 20% longer than the support span. Before testing, the specimens were stored in a conditioned environment, and the span was measured accurately to within 0.1 mm for spans less than 63 mm and within 0.3 mm for spans greater than 63 mm. The testing speed was set at a rate of 1.0 mm/min. During the test, each specimen was supported by a support span, and the load was applied at the centre of the specimen by a loading nose, which potentially caused the specimen to bend. The support span, loading speed, and maximum deflection were the primary parameters for the test. If a specimen did not break, the test was continued. The mode, area, and location of failure were recorded for each specimen. The maximum flexural stress, flexural stress at break, and flexural modulus were all determined through the three-point bending test [10].

In this study, two categories of test specimens with different fibre orientations and varying reinforcing filler loadings were tested. The first category included two types of specimens: Type-A, with a fibre orientation of 0°/0°, and Type-B, with a fibre orientation of 0°/90°. This category did not include any reinforcing filler. Table 1 presented the types of specimens prepared with different fibre orientations. For the second category involving reinforcing filler loading, three types of specimens and one control specimen were prepared. Each specimen consisted of two layers of ramie fibre and coffee bean powder at 5 wt%, 10 wt%, and 15 wt%, respectively. All specimens in this category were fabricated with a ramie fibre orientation of 0°/90°. Table 2 presented the types of specimens prepared with different weight fractions.

Table 1: Type of specimens of different fibre orientation

Specimen	Matrix	Ramie Fibre (layer)	Fibre Orientation	Number of specimens
Type-A	Epoxy	2	0°/0°	3
Type-B	Epoxy	2	0°/90°	3

Table 2: Type of specimens of different weight fractions of coffee bean powder

Specimen	Matrix	Ramie Fibre (layer)	Coffee Bean Powder (wt.%)	Number of specimens
Control	Epoxy	2	0	3
Type-1	Epoxy	2	5	5
Type-2	Epoxy	2	10	5
Type-3	Epoxy	2	15	5

The specimen sizes of the composites were cut according to the ASTM D7264 standard. For flexural strength testing, the standard support span-to-thickness ratio was selected to ensure that failure occurred at the outer surface of the specimens due solely to the bending moment. The standard span-to-thickness ratio was 32:1, with the specimen length being approximately 20% longer than the support span. Since maintaining a standard thickness of 4 mm using the Vacuum-Assisted Resin Infusion (VARI) technique was challenging, alternative specimens were used while preserving the 32:1 support span-to-thickness ratio and the standard specimen width. The standard specimen width was 13 mm. The average thickness of the composites was measured using a Vernier Caliper with a systematic error of ±0.02 mm, and the specimen length was calculated based on the 32:1 ratio.

2.4 Characterization Methods

To study the fracture behaviour and fracture mechanism of hybrid epoxy composites reinforced with ramie fibres and coffee bean particles, microstructural analysis was used as an effective and accurate method to determine the characteristics of the material. An Olympus BX53M optical microscope was used to observe the particle size of the coffee bean powder, which was ensured not to exceed 200 µm. The microscopic images obtained from the optical microscope were processed and analysed using ImageJ software to calculate the mean particle size of the coffee grind. The fracture mechanism of the hybrid epoxy composites was examined using scanning electron microscopy (SEM). Features such as fibre or particle breakage, fibre pull-out, and resin microcracks were observed through SEM to analyse the microstructural changes on the surface of the specimens.

3. RESULTS AND DISCUSSION

3.1 Coffee Beans Particle Size Distribution

The fabrication of the hybrid epoxy composites reinforced by ramie fibre and coffee bean particulate was successful. The average size of the coffee bean powder was 27.5 µm. Figure 2(a) shows an example of the image used in the determination of the coffee bean powder size distribution. Figure 2(b) shows the size distribution of coffee bean powder fabricated in this study.

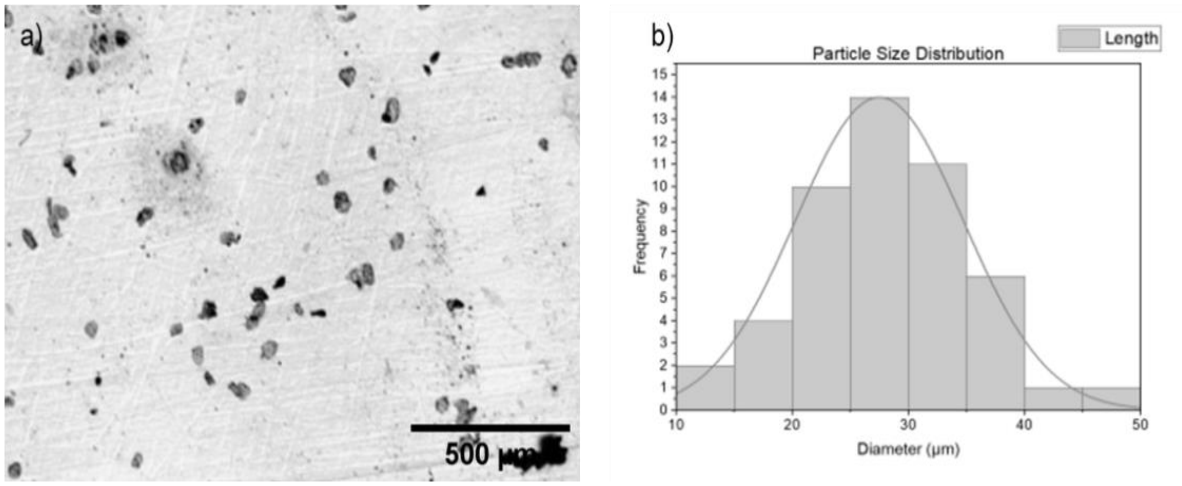

Figure 2: (a) Coffee bean powder size distribution with optical microscopy image at 100x magnification and (b) Particle size distribution of coffee bean particulate

3.2 Effect of Ramie Fibre Orientation

The flexural strength values from experimental results of epoxy ramie fibre composites with different fibre orientations are shown in Table 3. From herein, the ramie fibre composite with 0°/0° (unidirectional) fibre orientation will be referred to as the Type-A specimen, and the ramie fibre composite with 0°/90° (bidirectional) fibre orientation will be referred to as Type-B. The results show that ramie fibre composite with 0°/0° (unidirectional) fibre orientation has higher flexural strength and flexural modulus compared to 0°/90° (bidirectional) fibre orientation.

Table 3: Flexural strength and flexural modulus of ramie fibre composites of different orientation

Specimens	Fibre Orientation	Flexural Strength (MPa)	Flexural Modulus (MPa)
Type-A	0º/0º	97.4906	2749.37
Type-B	0º/90º	27.05	1075.04

The flexural strength and flexural modulus of ramie fibre composites with different orientations are plotted into bar charts as shown in Figure 3(a) and (b), respectively. The result shows that the flexural strength of ramie fibre composite of 0°/0° fibre orientation is 3 times higher than that of 0°/90° fibre orientation. Likewise, the 0°/0° oriented ramie fibre composite also showed a higher flexural modulus than the 0°/90° oriented, almost 2.5 times higher. The results obtained were similar to the previous study where the fibre orientation of 0°/0° shows higher flexural strength as compared to 0°/90° [11]. According to Muller et, al 0°/0° fibre orientation composites are attributed to the propagation of the crack being perpendicular to the orientation of fibres, while 0°/0° fibre orientation composites are attributed to the propagation of the crack being parallel to the orientation of fibres and load-bearing by only matrix material. This is because, in a 0°/0° fibre orientation specimen, the crack propagation is arrested by fibres during the fracture process. In a 0°/90° fibre orientation composite, there is a minimum contribution of fibres during the fracture process [12].

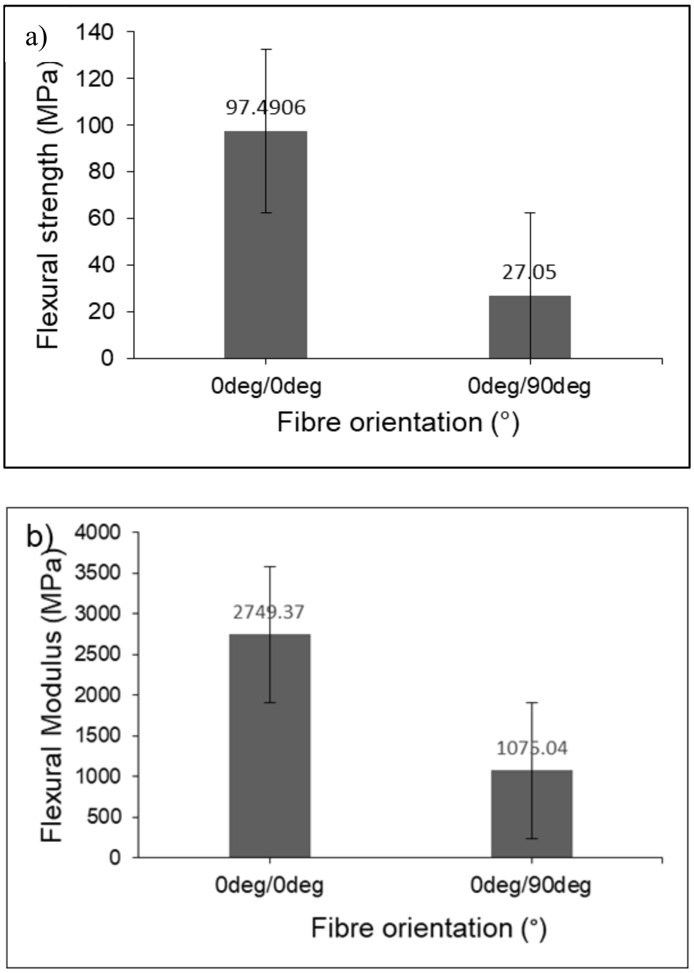

Figure 3: Bar graph for (a) Flexural strength of ramie fibre composite of different orientations and (b) Flexural modulus of ramie fibre composites of different orientation

Figure 4 shows the load vs. displacement curves of ramie fibre composites of different orientations.

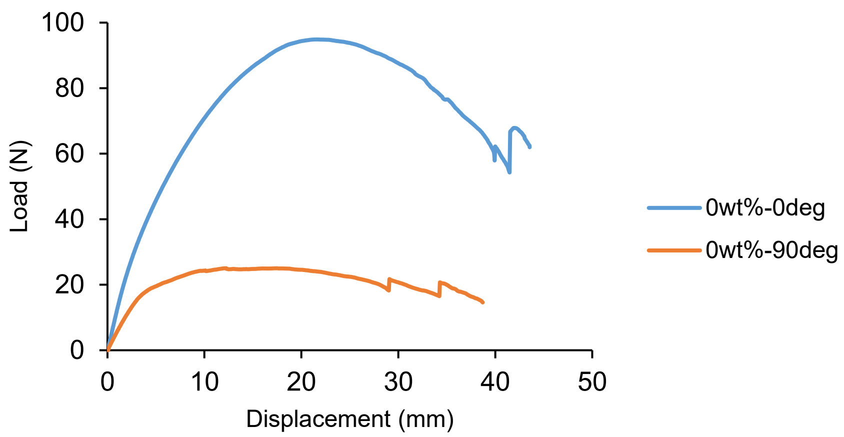

Figure 4: Load-displacement curves of ramie fibre composites of different orientations

It can be observed that the ultimate load for a ramie fibre composite of 0°/0° fibre orientation is at 94.86 N, and the maximum failure displacement is at 41.48 mm. Whereas for ramie fibre composite of 0°/90° fibre orientation, the ultimate load is only at 25.03 N and the maximum displacement at the failure point is 29.02 mm. From the load-deflection curve, the ramie fibre composite of 0º/0º fibre orientation shows better flexural behaviour as compared to 0°/90° fibre orientation.

3.3 Effect of Coffee Bean Particulate Weight Fraction

The flexural strength values from experimental results of reinforced hybrid epoxy ramie fibre composites with different weight fractions of coffee bean particulate are shown in Table 4. Ramie fibre composite with 0wt% of coffee bean powder as the control specimen for the comparison. The reinforced hybrid epoxy ramie fibre composite with 5 wt%, 10 wt%, and 15 wt% of coffee bean powder is referred to as Type-1, Type-2, and Type-3, respectively. The results show that reinforced ramie fibre composite with 10 wt% coffee bean powder has the highest flexural strength at 29.945 Mpa. However, among the reinforced ramie fibre composites with different weight fractions, the flexural modulus value of the ramie fibre composite containing 15 wt% coffee bean powder was the highest.

Table 4: Flexural strength and flexural modulus of reinforced hybrid epoxy ramie fibre composites with different weight fractions of coffee bean powder

Specimens	Coffee Bean Powder (wt.%)	Flexural Strength (MPa)	Flexural Modulus (MPa)
Control	0	27.05	1075.04
Type-1	5	28.8803	1417.54
Type-2	10	29.945	1347.64
Type-3	15	25.9785	1441.66

The flexural strength and flexural modulus of ramie fibre composites with different weight fractions are plotted into bar charts as shown in Figure 5(a) and (b), respectively. The flexural strength of the control specimen (0wt% coffee bean powder) was 27.05 MPa, which was lower than the 47.58 MPa of the previous study [7]. It can be observed that as the weight fraction of coffee bean powder increases, the flexural strength also increases. However, after reaching the optimal weight fraction at 10 wt%, the flexural strength starts to decrease. As shown in Figure 5, the flexural strength of the ramie fibre composite containing 15 wt% coffee bean powder was the lowest, even lower than that of the control specimen. As the filler loadings were increased to a significant amount, the flexural properties started to drop, which was attributed to the increase in viscosity in the matrix and, in turn, the increase in porosity and decrease in wettability in the composites. The study by Liu et al. [13] shows the weakness of the lignocellulosic phase in stress transition to polymer matrix will decrease the flexural strength as the filler loading increases.

For the flexural modulus, different weight fractions of coffee bean powder of reinforced epoxy ramie fibre composites show different flexural modulus. Although the reinforced ramie fibre composite with 10 wt% coffee bean powder has the highest flexural strength among the reinforced ramie composites composed of various compositions of coffee bean powder, its flexural modulus is slightly lower than 5 wt.% and 15 wt%. The previous study showed that the flexural modulus does not have a significant change as the filler content increases [9]. However, the flexural modulus of the hybrid epoxy ramie fibre composite reinforced with coffee bean powder was higher than that of the control specimen without any reinforcing filler. Among them, 15 wt.% reaches the highest flexural modulus at 1441.66 MPa, followed by 5 wt% at 1417.54 MPa and 10 wt% at 1347.64 MPa. The slight increase in flexural modulus as the filler loading increased most properly is due to the increase in volume fraction of high-modulus lignocellulosic in composite panels [14].

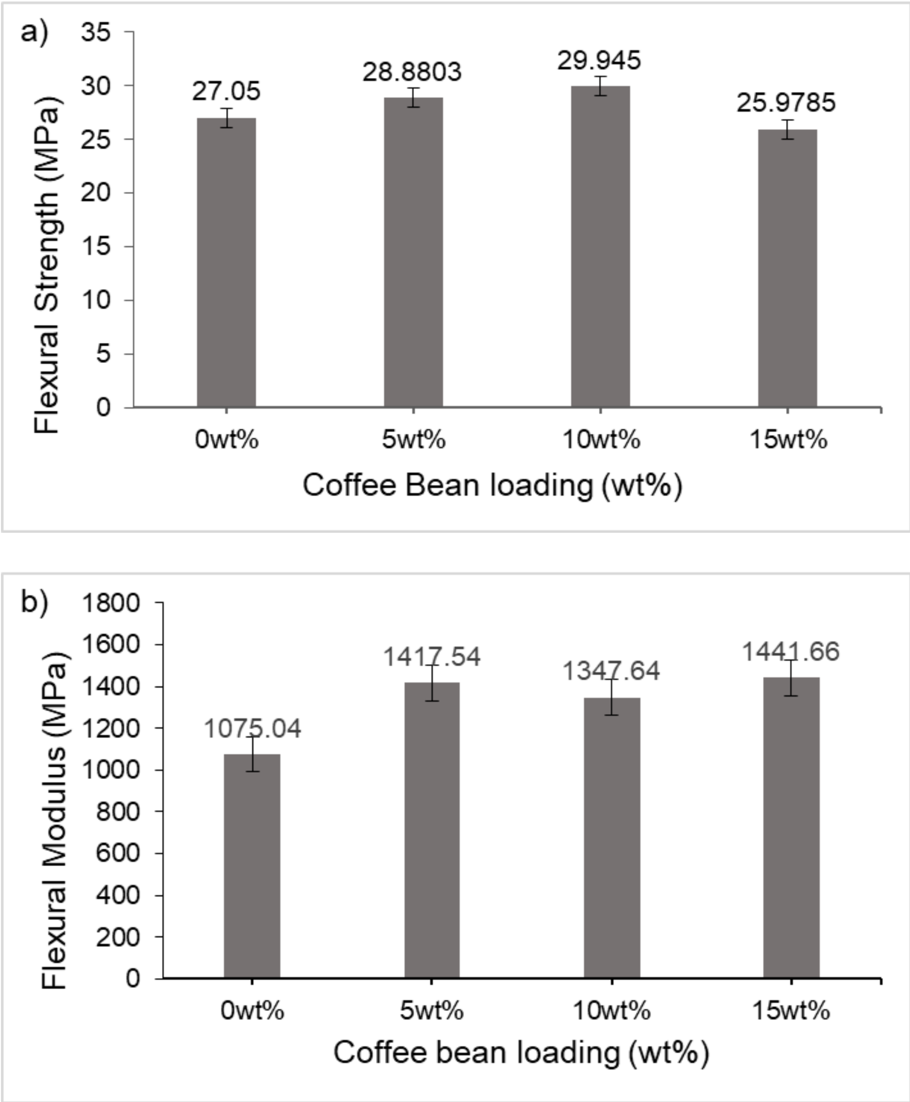

Figure 5: (a) Flexural strength of reinforced hybrid epoxy ramie fibre composites with different weight fractions of coffee bean powder and (b) Flexural modulus of reinforced hybrid epoxy ramie fibre composites with different weight fractions of coffee bean powder

Figure 6(a) shows the load vs. displacement curves of ramie fibre composites of various compositions of coffee bean powder. It can be observed that the loading increases with the weight fraction of coffee bean powder up to 10 wt% and then decreases to 15 wt%. The load-displacement curves show that the reinforced ramie composite with 5 wt% coffee bean powder behaves similarly to 10 wt%. Figure 6(b) shows the initial straight-line portion of the load deflection curve. From the graph, it can be seen that as the weight fraction of coffee bean powder increases, the slope of the curve also increases. However, there was only a slight increase in flexural modulus. The flexural modulus refers to the tendency of a material to resist bending, which is related to the slope of the load-deflection curve. Hence, the flexural modulus for reinforced ramie composite with 15 wt% coffee bean powder has the highest flexural modulus as compared to other weight fractions.

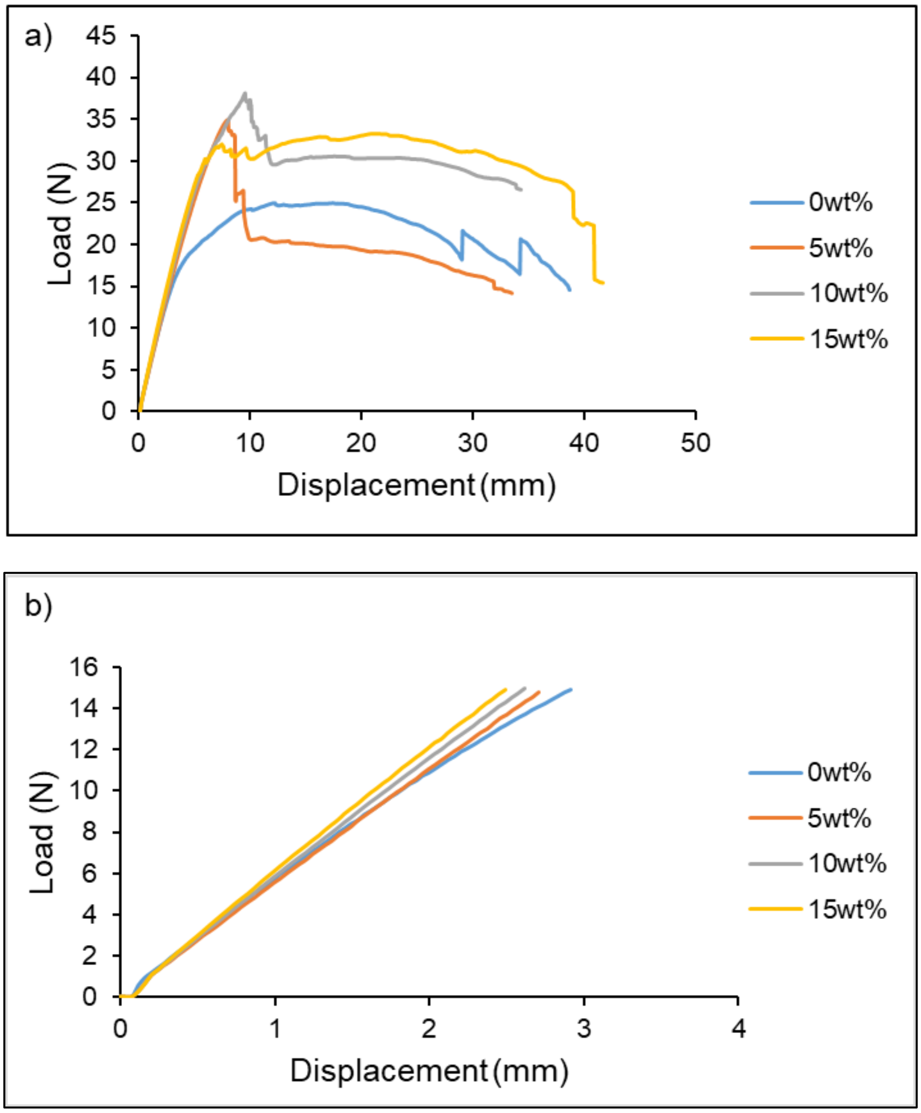

Figure 6: (a) Load-displacement curves of reinforced hybrid epoxy ramie fibre composites with different weight fractions of coffee bean powder and (b) Load-displacement curves of reinforced hybrid epoxy ramie fibre composites with different weight fractions of coffee bean powder

3.4 SEM Analysis

Figure 7 shows the SEM image of fibre-matrix interfacial bonding in a ramie fibre composite. The ramie fibres were observed to be unidirectionally oriented in the composite. However, some improper interfacial adhesion was observed on composites, as shown in Figure 7. This indicates that there is poor adhesion between the fibres and matrix. This contributes to the failure of composites and reduces the flexural strength and modulus.

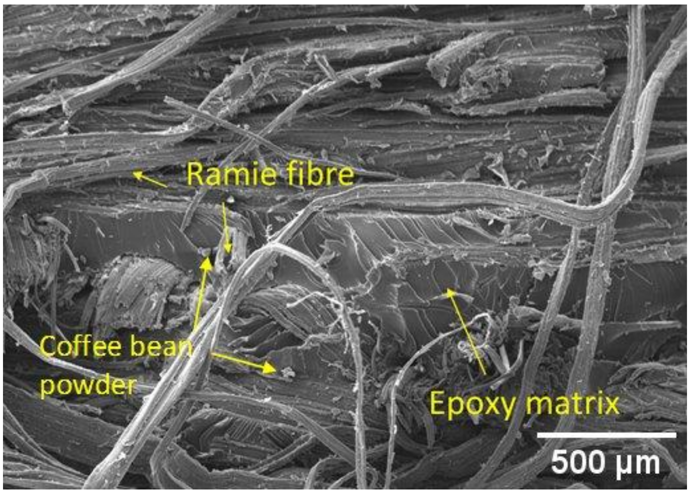

Figure 7: SEM image of fibre-matrix interface

The ramie fibre composites were made up of 2 layers of ramie with different orientations 0°/90° as shown in Figure 8. Referring to the images, the boundary between the two orientations of ramie fibre and the fibre alignment can be identified. The fracture of composites only occurred at the area where ramie fibre was 90° orientated, which is parallel to the load cell and impact roller when conducting the three-point bending tests. Figure 9 shows the fracture pattern of ramie fibre composites reinforced by various compositions of coffee bean powder after completion of the three-point bending tests. The crack pattern for all the composites is similar under SEM micrography, which is mainly due to the breakage of the matrix. The presence of coffee bean powder as a filler may have introduced interfacial incompatibility or hindered proper matrix infiltration, especially in regions where agglomeration or poor wetting occurred. This could further explain the observed interfacial voids and the reduced mechanical performance [15].

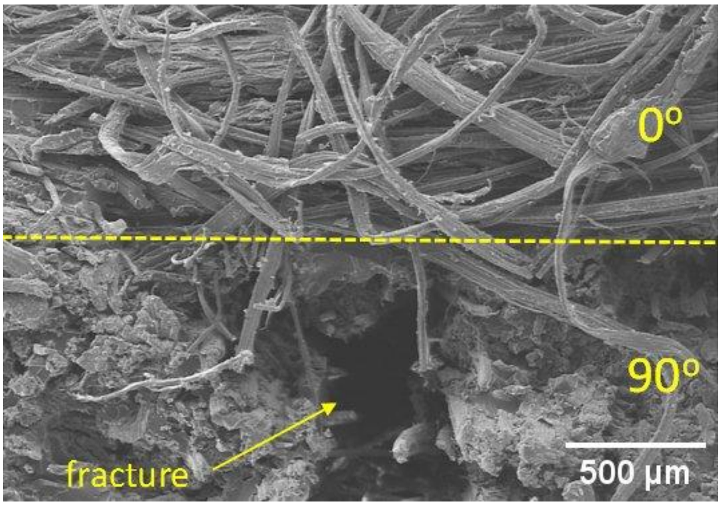

Figure 8: SEM images of fibre alignment of different fibre orientations in ramie fibre composite

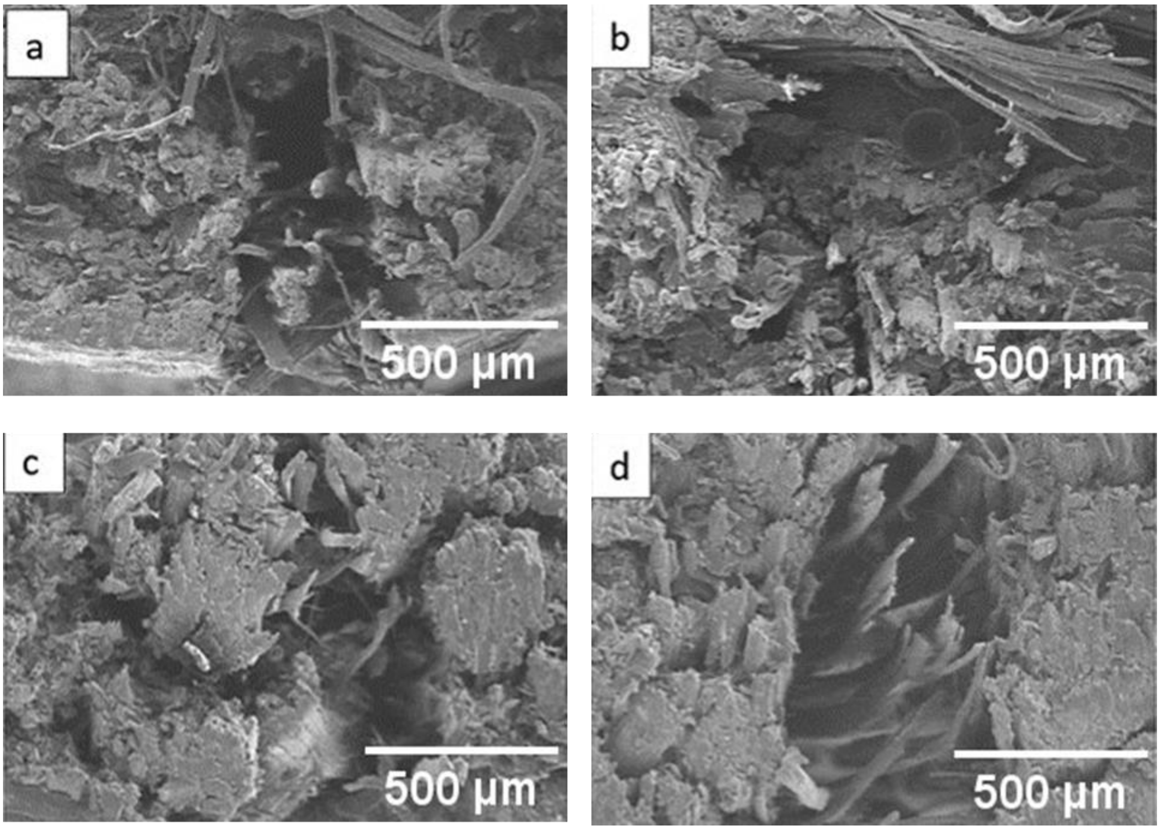

Figure 9: SEM images of the fracture surface of ramie fibre composites reinforced with (a) 0 wt%, (b) 5 wt%, (c) 10 wt% and (d) 15 wt% of coffee bean powder

Figure 10 shows the failures that occurred in the composites. The ramie fibre composite begins to delaminate as the load is transferred to the ramie fibre composite. Due to the high tensile strength properties of natural fibres, the matrix tends to delaminate first, as shown in Figure 10(a). The microcracking in the matrix is shown in Figure 10(b). Due to the brittleness, epoxies are susceptible to damage in the form of microcracks. Once microcracks, the cracks are easy to propagate. The same behaviour [12] and in the literature [5].

It can be seen that voids and fibre pull-out were observed in Figure 10(d) and (e) respectively, which indicates poor adhesion in the fibre-matrix interface. Poor adhesion at the fibre-matrix interface, voids, and debonding were observed, which may have initiated cracks to fail the composite during three-point bending tests. The three-point bending test sample from SEM analysis concludes that the three-point bending test sample has significant damage in reinforced ramie fibre composites. The breakage of the fibre and matrix indicates that the failures are due to the flexural tests as shown in Figures 10(a) and (c). The increased crack density at the fibre-matrix interface further confirms the detrimental effect of poor adhesion and filler-induced defects [16,17].

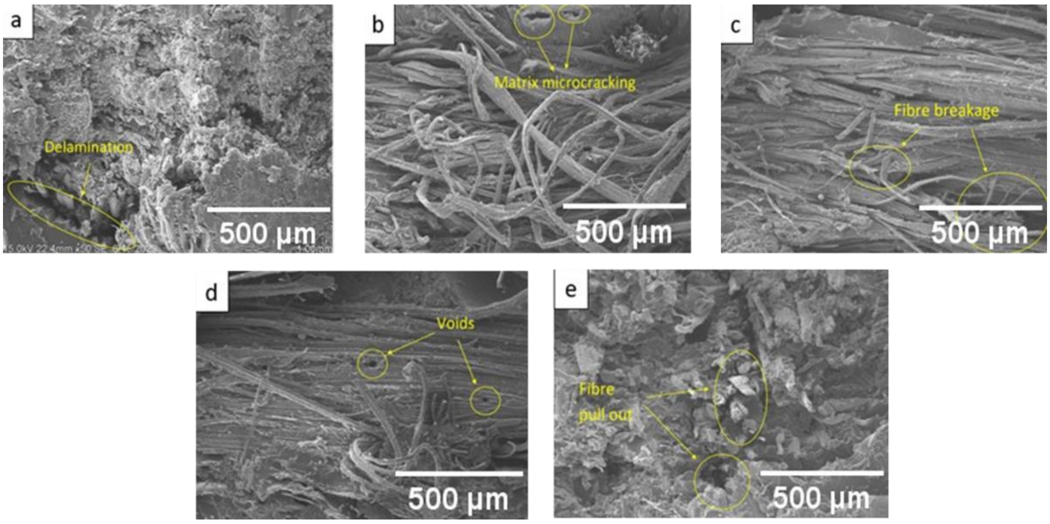

Figure 10: SEM fractography of failure in hybrid epoxy ramie fibre composites which are (a) delamination, (b) matrix microcracking, (c) fibre breakage, (d) voids and (e) fibre pull out

4. CONCLUSIONS

The present investigation revealed that fibre orientation and filler weight fraction significantly influence the flexural properties of composites. The current study shows that fibre orientation and filler weight fraction significantly affect the flexural properties of composites. Ramie fibre composites with 0°/0° (unidirectional) fibre orientation achieved higher flexural strength and flexural modulus compared to 0°/90° (bidirectional) fibre orientation. Bidirectional fibre orientation had a tendency to line up perpendicular to the load cell and impact roller of the Universal Testing Machine during three-point bending tests and hence increased the total number of fibres the radial cracks formed during testing.

The flexural strength for ramie fibre composites of various weight fraction of coffee bean powder was found to be maximum at 10 wt.%. The flexural strength of ramie fibre composites increased with the increase of the weight fraction of coffee bean powder, but after reaching the optimal weight percentage of coffee bean powder, the flexural strength began to decrease. The flexural modulus for the hybrid epoxy composite reinforced ramie fibre and coffee bean powder was found to be maximum at 15 wt.% of coffee bean powder. The flexural modulus is referred to the tendency of a material to resist bending, which is related to the slope of the load deflection curve.

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Author Contributions

All authors contributed toward data analysis, drafting and critically revising the paper and agree to be accountable for all aspects of the work.

Disclosure of Conflict of Interest

The authors have no disclosures to declare.

Compliance with Ethical Standards

The work is compliant with ethical standards.

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