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Exploring the potential of *Tamarindus Indica* and *Caryota Mitis* (CM) leaf stalk fibers for sustainable biodegradable composites

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ABSTRACT

As a substitute for synthetic materials, researchers studying natural fiber materials are hard at work developing biodegradable composite materials. These natural fiber-infused polymer composites are environmentally friendly and have excellent mechanical qualities. The creation and analysis of epoxy polymer composites reinforced with *Tamarindus indica* and *Caryota mitis* leaf stalk fiber are the main objectives of this work. Five distinct compositions are used in the hand layup procedure to create the composite specimens. An evaluation was conducted on the mechanical qualities, encompassing hardness, tensile strength, and impact resistance. In order to evaluate the physical and chemical properties of the composites, further metallurgical features were investigated using wear, microstructure and thermogravimetric analysis (TGA). Specimen C, which was composed of 20% *Caryota mitis* 20% *Tamarindus indica* powder, and 60% epoxy, showed the best tensile strength (69.12 MPa) and the highest hardness (76.37 VHN) of all the compositions examined. The composite exhibited favourable bonding and temperature adaptability, as demonstrated by the microstructure and TGA studies. All things considered, the research indicates that these hybrid composites have better particular qualities, which makes them a suitable material for use in automotive and aerospace applications.

KEYWORDS

Composites; Natural fibers; *Caryota mitis* leaf stalk fibers; *Tamarindus indica* seed powder; XRD; TGA/DTA

ARTICLE HISTORY

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Introduction

The development of natural composites has been the focus of aircraft industry research in recent years due to the rise in environmental pollutants. Indian researchers are searching for novel applications for residual plant fibers in order to create eco-friendly products [1]. Natural fibers are becoming more and more popular as a result of this green composite and the growing need for sustainable alternatives to conventional materials [2]. Over the past few decades, a lot of researchers have worked to increase awareness of natural fiber-reinforced composites. Accordingly, tests are conducted to determine the qualities of green composites reinforced with *Caryota mitis* leaf sheath fibers and tamarind seed powder mixed with epoxy resin matrix [3]. *Caryota mitis* leaf sheath is easily accessible and has a high tensile strength, which is crucial for preserving the structural integrity of composite materials. Tamarind seed powder is also reasonably priced and is thought to be a waste product in the tamarind industry. Together with epoxy resin, these fillers can be used to prepare sustainable biodegradable composites [4]. Most places underutilize tamarind seeds, but there's plenty that can be done better. Biofillers are utilized because of their exceptional mechanical and thermal properties, as well as their low density, according to numerous authors. A comprehensive analysis of current developments in this sector is required for an evaluation of the effectiveness of renewable sources in composite materials, as several authors have documented. The mechanisms that lead to the degradation of polymeric components from manufacture to eventual failure were the main topic of review [5]. In addition to discussing the many kinds of fibers—such as those found in sugarcane and tamarind— Girimurugan et al. 2022, gave a summary of the current research being done in this field, covering its qualities, processing, and uses investigated the advantages of environmentally friendly composite materials in their study [6]. These advantages included enhanced mechanical performance, excellent electrical insulation, high heat resistance, good fire behavior, high impact resistance, and best abrasion resistance. The creation of composite materials was another area of study. The main applications of fiber-based particulate polymer composites were outlined by Alarifi et al. 2023, who also looked at how reinforcements affected the machinability of polymer composite structures [7]. An overview of recent developments in natural FRP hybrid composites was given by authors Arrakhiz et al. 2013, who also highlighted methods for improving the materials' intrinsic qualities while having a smaller negative environmental impact [8]. Ismail et al. 2022, compared conventional glass fiber composites with biodegradable materials and their mechanical qualities that are reinforced with natural fibers [9]. A novel environmentally friendly composite was presented by Abdollahiparasa et al. 2023, and has the potential to replace synthetic or non-renewable sources [10]. The methodological examination of composites

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reinforced with jute fabric and green polyurethane resin was the authors' main focus. Loganathan et al. 2020, examined the compressive and flexural properties of a composite material made of tamarind seed powder, sugarcane fibers, and an epoxy resin matrix [11]. The procedure for extracting the fiber from the areca nut's sheath, preparing the composite, arranging the fiber naturally, determining the orientation of the fiber, and analyzing experimental results on mechanical properties were all covered in detail in the paper. The impacts of structural, thermal, and elemental investigations on epoxy polymer composites reinforced with short areca nut fibers that have been treated with NaOH were examined by Sankarathil et al. 2023 [12]. The mechanical characteristics of fiber/matrix glass-reinforced epoxy composites with varying fiber/matrix magnitudes were the subject of study [13]. The use of bioplastics and bioresources as viable remedies to environmental issues resulting from single-use plastic waste food packaging was examined by Pulikkalparambil et al. 2023 [14]. The survey underlined how popular natural fibers are becoming because of their affordability, environmental friendliness, and biodegradability. The improvement in the tensile characteristics of hybrid bio-composites reinforced with sugarcane fiber and tamarind seed powder and epoxy resin matrix was reported by Jayaraman et al. 2022 [15]. The production of biodegradable composites employing areca nut frond fibers for low-strength applications was studied by Padmaraj et al. 2013, who emphasized the areca fibers excellent tensile strength. The effects of tamarind seed gum and banana fiber on the produced composites characteristics were investigated by Kiruthika et al. 2012 [16,17]. A wear study of banana fly ash (BA) combined with a hybrid natural fiber mixture of sisal and pineapple was presented by Keerthiveettil et al. 2022 [18]. They noticed that favorable tribological qualities were produced by a particular combination of loading, sliding distance, and hybrid fiber content. The creation of naturally fiber-reinforced polymer composites that are environmentally sustainable was covered by Mishra S et al. 2023 [19]. The technological difficulties, processing methods, characterization, characteristics, and possible uses of these composites were all compiled in their paper. One of the main drawbacks of natural fibers is that they are prone to absorbing moisture, which can lead to damage or degradation because of the hydroxyl groups present [1,20]. Natural fiber's mechanical characteristics are mostly determined by the amount of cellulose present, which affects the state of the cell geometry. However, it has been discovered by researchers that adding small amounts of nanoclay and nanosilica carbide can lessen the hydrophilicity of natural fiber-reinforced polymer composites introducing a novel approach to developing bio-composites from natural sources, advancing the promotion of green composites for structural applications [6,21]. The study demonstrates the potential of using a blend of plant-based and animal-derived reinforcements, offering an innovative alternative to the conventional practice of using these materials individually.

The review article is designed to provide a comprehensive overview of recent literature on epoxy structures, synthesis, modified epoxy resins, bio-based epoxy resins, and their diverse applications. It also aims to cover the latest advancements in natural fiber-reinforced epoxy composites and nanocomposites, including manufacturing techniques and various industrial applications [22].

The study aims to fabricate an epoxy resin-based polymer matrix composite incorporating alkali-treated tamarind fruit fiber at varying weight percentages (0 wt%, 10 wt%, 20 wt%, and 30 wt%) using the resin mould technique [23]. The study aims to provide essential insights into natural fibers and their composites, supporting future research, fostering the development of innovative polymer composite frameworks, and contributing to the achievement of Sustainable Development Goals [24]. Seyda Eyupoglu et.al. 2024 study goals to evaluate the effects of enzymatic and alkali treatments on Sambucus ebulus L. stem fibers, to achieve this, the fibers were treated with alkali, cellulose, and pectinase enzymes [25]. To determine the fiber characteristic properties, physical, chemical, and instrumental tests were performed.

This study contributes to the growing field of sustainable composite materials by investigating the potential of a hybrid composite made from *Caryota mitis* leaf stalk fibers and *Tamarindus indica* seed powder as a reinforcement in an epoxy resin matrix. A novel approach has been adopted by treating the areca fibers with alkali (NaOH) treatment, improving the interfacial adhesion between the fibers and the matrix, thus enhancing the mechanical properties of the composite. The research explores the varying ratios of fiber and seed powder in the composite to optimize its performance across a range of mechanical, chemical, and thermal properties. By conducting detailed measurements such as hardness, tensile strength, impact strength, XRD analysis, microstructural examination, and TGA-DTA testing, this work aims to provide comprehensive insight into the composite's characteristics.

The novelty of this study lies in its innovative combination of *Caryota mitis* fibers and tamarind seed powder as bio-fillers in a sustainable, lightweight composite material. This hybrid composite not only offers superior mechanical properties but also contributes to environmental sustainability by utilizing natural, readily available agricultural waste products.

The primary aim of this research is to develop a cost-effective, eco-friendly composite material with potential applications in aircraft structures and other industries requiring lightweight and high-strength materials. The outcomes of this study will contribute to the development of bio-based composites that can replace synthetic materials in advanced engineering applications.

Materials and Methods

Caryota mitis leaf sheath fiber extraction

The fiber material utilized in this piece is the sheath of *Caryota mitis* leaves. The *Caryota mitis* leaf stalk is one of the main raw materials that is taken from the areca-nut palm. The stiff, fibrous, biodegradable covering on leaves. It carries out a variety of biological functions, including carrying water, giving mechanical support, and enduring a range of stresses [4]. The technique of extracting the *Caryota mitis* leaf sheath is depicted in Figure. 1(a-d).

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(a) *Caryota mitis* leaf stalk

(c) Extracted *Caryota mitis* leaf stalk

(b) *Caryota mitis* leaf stalk soaked in NaOH

(d) Final *Caryota mitis* reinforcement

Figure 1. (a-d) Extraction process of *Caryota mitis* leaf sheath natural fiber.

Long staple fibers could be extracted from the *Caryota mitis* leaf stalk by applying an alkali treatment. 40 grams of fibers could be extracted from 100 grams of *Caryota mitis* leaf sheath [5]. Alkali treatment, which alters and cleanses natural fiber's surface to improve interfacial bonding and lower surface tension, is one of the most widely used chemical treatment techniques [6,26]. *Caryota mitis* fibers are submerged in a 5% NaOH solution for two hours at room temperature. The alkali-treated fibers were immersed in distilled water for an entire hour in order to remove any leftover NaOH. Lastly, sun drying was done after washing in distilled water. Table 1 shows the mechanical properties of Areca catechu leaf stalk fiber.

Table 1. *Caryota mitis* leaf stalk fiber mechanical properties.

Specific Strength (Pa/kg/mm^{2})	Transverse Young's Modulus (GPa)	Tensile Strength (MPa)	Specific Modulus (Pa/kg/mm^3)	Longitudinal Young's Modulus (GPa)	Density (kg/mm^3)
96.457×10^{6}	0.114	34.65	7.373×10^{9}	2.660	0.3439×10^{-6}

Tamarindus indica seed powder extraction

Tamarindus indica seed powder is the second fiber utilized in this investigation. *Tamarindus indica* seed, which is produced in tropical countries including Thailand, Sri Lanka, Malaysia, Indonesia, and India, is one of the most important natural fillers. Because of its great strength and modulus properties as well as its high lignin content, *Tamarindus indica* seed filler is a promising material for the production of natural fiber composites [6]. Seed kernels decorated with almonds have 46–48% of a gel-forming ingredient. Polyose exhibits good gelling and film-forming qualities. Polyose, another name for *Tamarindus indica* seed, has been shown to provide better bonding. Many efforts have been focused on using different natural fillers in composites in recent years. The high lignin content of the composites made with these fillers makes them more suitable and boosts their resilience to weathering [15]. The seeds also absorb less water as a result of their decreased cellulose content. When the *Tamarindus indica* seeds arrived, they were first washed with water. After that, they had three or four days to spend in the sun drying. The seed powder was ground and prepared locally using milling, and sun drying is necessary to reduce the moisture content [19]. The image of tamarind seeds and powder that has been extracted is displayed in (Figure 2).

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(a) *tamarindus indica* seeds

(b) *tamarindus indica* seed powder

Figure 2 (a & b). Extracted *tamarindus indica* seed powder.

Preparation of composites

The weight percentage composition of the fiber and reinforcement composite specimens is displayed in Table 2. Numerous composite goods in a range of sizes can be produced using the open moulding method of hand lay-up [1]. It is one of the most often used techniques for creating composite materials and has the benefits of being simple, affordable, and having little technical knowledge [2]. It involves the draperies and the fibers being impregnated with resin.

Table 2. Composition of *Caryota mitis* and tamarind composite specimens.

Hybrid composite Specimens	Fiber %		Reinforcement %
	Caryota mitis leaf stalk fiber in wt%	*Tamarindus indica* seed powder in wt%	Epoxy in wt%
Sampling - A	10	30	60
Sampling - B	15	25	60
Sampling - C	20	20	60
Sampling - D	25	15	60
Sampling - E	30	10	60

This technique can be applied to both large and small items. As a result, part size is not restricted. Better methods can be used to reduce the quantity of these manufacturing-related errors. It's among the easiest and most effective methods. Next, a coating made of epoxy resin polymer was applied. Correct rolling was done after one type of particle fiber was layered uniformly [3]. We mixed some of the resin hardener mixture with powdered *Tamarindus indica* (10:1 Ratio). This mixture was rolled and then applied to the fibers. This technique was continued until the required thickness was reached. A high-quality surface finish was achieved by applying a final layer of epoxy coating. To level the fibers over the reinforcing material, a little rolling was done. For the testing procedure, samples were sliced in accordance with ASTM standards [4,27]. All of the prepared tamarind and *Caryota mitis* leaf stalk fiber-reinforced epoxy composite specimens are displayed in Figure 3(a–c).

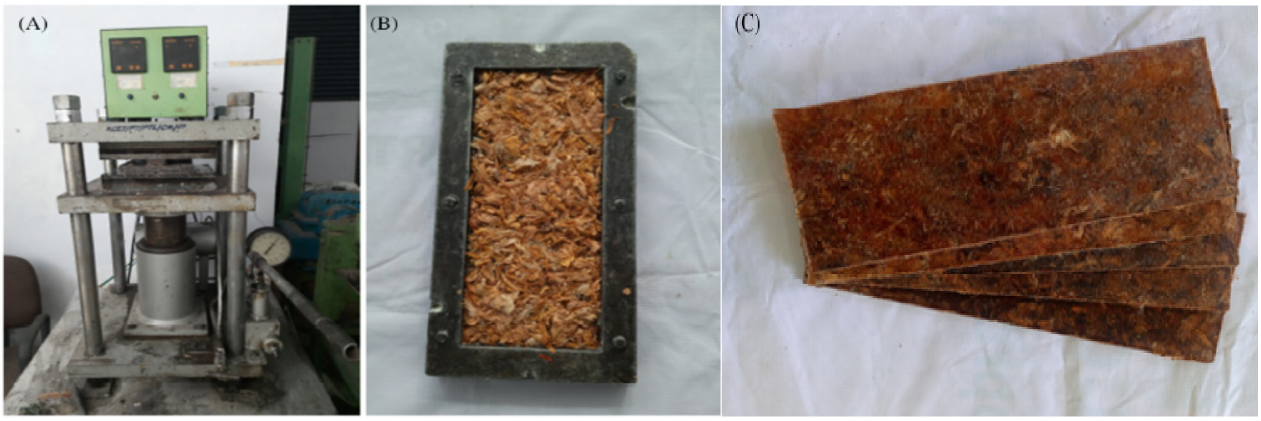

Figure 3(a-c). Prepared *Caryota mitis* leaf stalk fiber and tamarind-reinforced composite specimens.

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Wear test using pin on disc

A data acquisition system-equipped pin-on-disc friction and wear monitor was used to evaluate the wear behavior of the composite materials. This specialized apparatus is intended to investigate wear in sliding situations using a stationary pin in contact with a revolving disc. The wear machine and the revolving disc with the specimen A. D.C. motor with a speed range of 100-1500 rev/min rotates the disc to create sliding conditions, resulting in a sliding speed of 0 to 10 m/sec. With a maximum loading capacity of 100 N, the load is applied to the pin (specimen) via a pulley string arrangement employing deadweight. Precise assessment of the wear properties of the composites is possible with this configuration.

Results and Discussion

Hardness testing

A 1/16-inch-diameter ball intender was used in the Vikers hardness test to determine the material's resistance to indentation. Their thinness and softness led to the adoption of the ASTM standard of 64 mm × 12.5 mm [28]. The hardness test results of the epoxy hybrid composite specimens reinforced with *Tamarindus indica* and *Caryota mitis* leaf stalk fiber is displayed in Figure 4. The samples are tested at two distinct weights, 60 kg and 100 kg, and the findings are combined from the three trials. It is discovered that sampling C and D have greater hardness under both 60 kg and 100 kg loads. Among them, sampling C has the highest HRA values, measuring 76.37 under 60 kg and 72.85 under 100 kg. Because, in comparison to the other specimens, sampling C contains a higher concentration of fibers and reinforcing. Higher fiber and epoxy concentrations improve the connection between the fiber and the filler material, increasing the material's hardness [8,29].

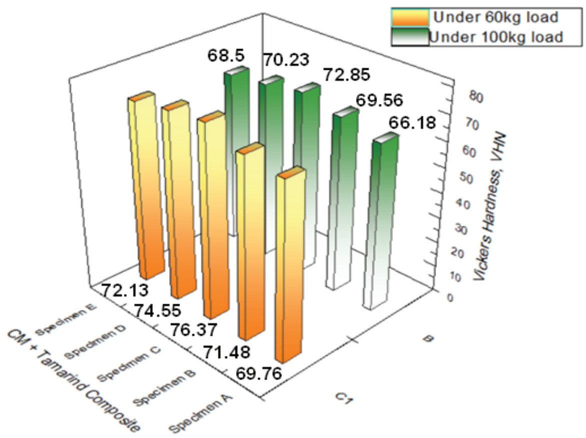

Figure 4. Hardness test result of composite specimens under 60 kg and 100 kg load.

Tensile strength analysis

The composite specimens measured 120 x 20 x 3 mm after they were made and sliced. They were conditioned for 48 hours at 50% humidity and 22°C in accordance with ASTM standard D3039 before being tested. For every composite, tensile strength and modulus were computed for each of the five samples, and the outcomes were examined. The tensile characteristics of epoxy hybrid composites reinforced with *Tamarindus indica* and *Caryota mitis* leaf stalk fiber is shown in Figure 5. Significant differences in the tensile characteristics were observed across different fiber ratios at 60% fiber loadings. The tensile strength and modulus of the hybrid composites reinforced with *Caryota mitis* leaf stalk natural fibers were noticeably higher. The specimen with the highest values, specimen C, had a modulus and tensile strength of 66.32 MPa. On the other hand, the values of the first two composite specimens, A and B, were significantly lower. Specimen E had the lowest tensile strength, measuring 60.68 MPa, while specimen A of the composite had a tensile strength and modulus of 58.15 MPa. The tensile strength and modulus of composites specimen C and D were found to be improved by increasing the proportion of *Caryota mitis* leaf stalk fiber and *Tamarindus indica* powder, with the least desirable mechanical attributes being produced by the highest ratio of tamarind powder. These variations show how important reinforcing is in determining the characteristics of the composite [14,30].

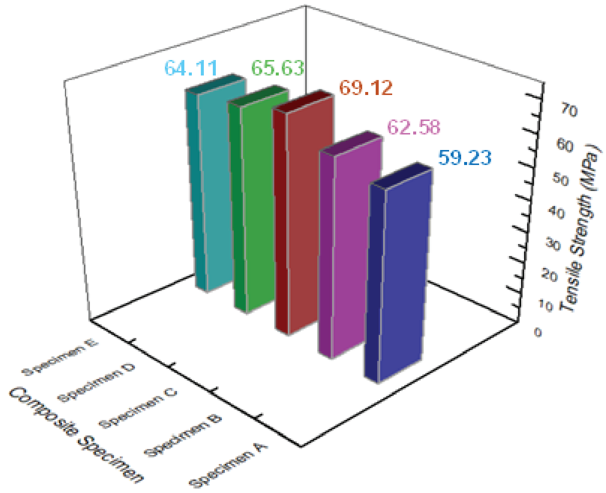

Figure 5. Tensile test result of *Caryota mitis* leaf stalk fiber and tamarind reinforced epoxy composite specimens.

The load-displacement curves from bending tests on composite specimens with the labels A, C, and E are shown in Figure 6. The findings show that as the composite's fiber content rises, so do the bending stress and resistance. Higher percentages of tamarind powder and *Caryota mitis* leaf stalk fiber also result in a decrease in the displacement value at which failure onset occurs. Because of the better mechanical qualities of *Caryota mitis* leaf stalk fiber-reinforced epoxy and its lower powder content, the composite specimen with a higher percentage of *Caryota mitis* reinforcement shows the greatest improvement [16]. Compared to the other specimens, the composite specimen with 20% caryota content exhibits notably earlier failure. The fiber *Tamarindus indica* powder-reinforced composite's solid structure and the *Caryota mitis* leaf stalk fiber layers' ability to absorb energy allow for an improvement in the strength of between 25% and 30%. The hybrid composite shows ductile behavior and fails at larger displacements because it has *Caryota mitis* sheath layers on the exterior. When compared to other fibers, *Caryota mitis* leaf stalk fiber has a higher tensile strength and elongation capabilities, which is why it exhibits this behavior [17,20].

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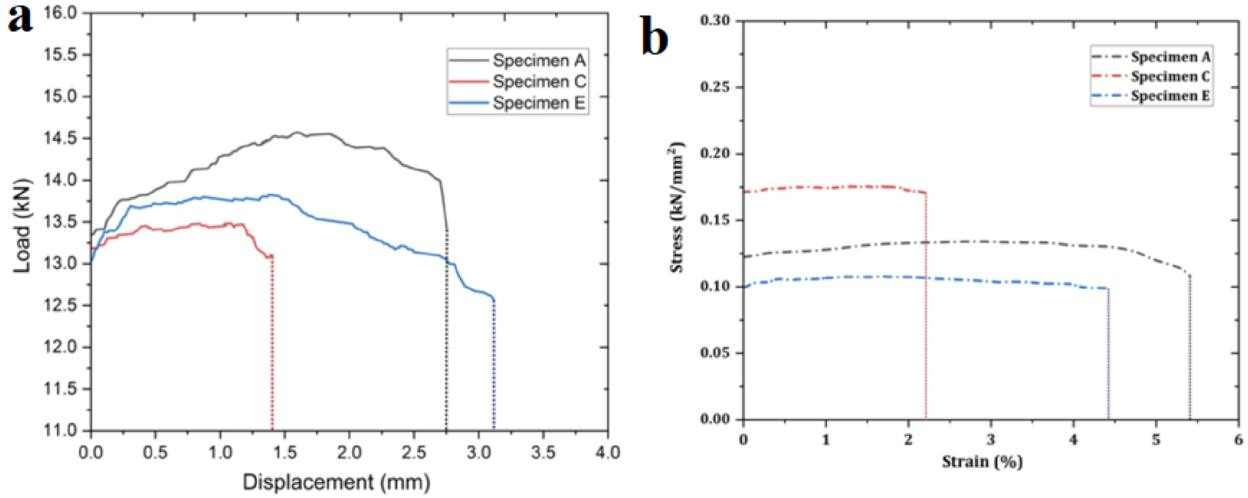

Figure 6. Tensile test result of *Caryota mitis* leaf stalk fiber and tamarind reinforced epoxy composite specimens.

Three chosen manufactured composite specimens' tensile behavior was investigated. The stress-strain behavior of epoxy composite specimens reinforced with *Tamarindus indica* and *Caryota mitis* leaf stalk fiber is shown in Figure 6 (b). Critical details on the material's strength and elasticity, as well as its behavior under various loads, were revealed by the tensile test. The tensile capabilities of composites with a higher proportion of *Caryota mitis* leaf stalk fiber reinforcement was clearly superior to those with a lower percentage of *Caryota mitis* leaf stalk fiber, as can be observed in Figure. 6 [31]. Tensile stress increased by 55% in specimens with concentrated *Tamarindus indica* and *Caryota mitis* leaf stalk fiber content from 5%, and it increased even more in specimens with 25% *Caryota mitis* leaf stalk fiber reinforcement. The specimens containing 20% *Tamarindus indica* and 20% *Caryota mitis* leaf stalk fiber exhibited the highest tensile stress, which was explained by the better surface conditions attained by the NaOH treatment. It is important to remember that natural fibers treated with alkali at high concentrations may degrade and fail, while lesser concentrations would leave behind amorphous hemicellulose on the fiber's surface, which would decrease its tensile strength [12].

As a result, an ideal concentration was estimated using the fibers tensile characteristics at various concentrations. Specimen C, as seen in Figure 6 (b), demonstrated the maximum tensile strength, increasing from 0.125 kN/mm² to 0.175 kN/mm². This result is in line with predictions because the tested specimens performed better due to the reduction in diameter, increased crystalline packing, and removal of hemicelluloses and surface contaminants. Specimen C's compressive strength was 0.182 kN/mm2, while specimens D and E had compressive values that were much lower than specimen C at 0.177 kN/mm2 and 0.180 kN/mm2, respectively. Because of this, composite specimen C has the greatest value for compressive strength.

The specimen 3 has a higher compressive strength than the specimens 4 and 5, according to the results. Because, in comparison to the other two specimens, specimen C has a higher concentration of fiber and epoxy. A higher concentration of epoxy and *Caryota mitis* leaf stalk fiber results in a stronger connection between the filler material and fiber. Consequently, specimen 3 performs better than specimens 4 and 5 because specimen 3 contains less tamarind seed powder, which has increased the composite material's strength relative to the strength of the *Caryota mitis* fibers [8,32,33].

Impact properties

According to the ASTM D638 standard, Figure 7 displays the different ratio-reinforced epoxy composites made of *Tamarindus indica* and *Caryota mitis* leaf stalk fiber that were employed for impact analysis. The test was carried out at a temperature of 25 degrees Celsius with the specimens clamped vertically and the pendulum released after a predetermined height. The energy absorbed by the entire specimens during fracture is 18 to 21 joules. When there is no stress concentration and the biodegradable composite material is more ductile, it can deform without breaking, the unnotched Charpy impact test usually yields average impact energy values [9,34].

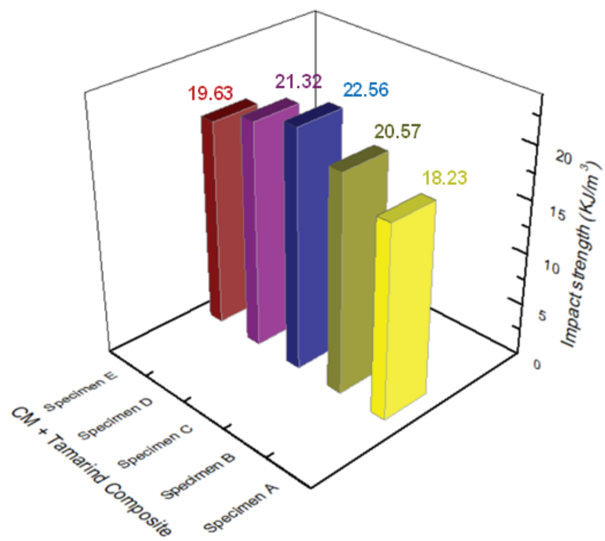

Figure 7. Charpy impact results of *Caryota mitis* leaf stalk fiber and tamarind reinforced composite specimens.

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Specimen C, which had a 25% *Caryota mitis* leaf stalk fiber component and the optimal ratio for impact properties, had the highest impact strength of all the hybrid composites. Because of the chemical bonding that forms from *Tamarindus indica* powder, which functions as a shock absorber and preserves the basic structure of the fibers and composites, *Caryota mitis* leaf stalk fiber and *Tamarindus indica* concentrated fibers showed greater impact resistance when compared to other composite fibers [10,35]. Nevertheless, the impact characteristics decreased as the tamarind % rose, most likely as a result of asymmetric fiber dispersion or inadequate interfacial bonding. When compared to *Tamarindus indica* alone, *Caryota mitis* leaf stalk fiber alone showed superior mechanical qualities and interfacial bonding.

For the shock to be adequately absorbed during impact, it must travel from the matrix to the fibers with efficiency. Lower impact strength can arise from inefficient transmission due to inadequate interfacial bonding [33,36]. Because of their combined tamarind and *Caryota mitis* leaf stalk fiber and their improved interfacial bonding, specimens C and D showed quality impact qualities. However, because the fiber content in the matrix had a smaller impact on impact properties, specimen E displayed somewhat lower impact properties, comparable to specimen A. Furthermore, it was shown that strain energy was linked to fiber debonding and that the fiber fracture was directly proportional to the length of fiber debonding [17,37].

Microstructure study

Through microstructure testing, the mechanical characteristics of the tested specimens are further investigated in order to assess fracture patterns and the interfacial bonding between the generated composites' fibers and matrix. Scanning electron microscopy (SEM) data obtained from tensile testing on composites reinforced with 25% and 30% *Caryota mitis* leaf stalk fiber is presented in Figure 8. There is more fiber withdrawal in these examples. In a similar vein, the composites containing 20% *Tamarindus indica* and 20% *Caryota mitis* leaf stalk fiber exhibit matrix breaking and void gap formation. Because of the non-homogeneous mixing of fibers in the matrix brought about by the insertion of hybrid reinforcement at lower weight percentages, these observations show reduced adhesion between the fiber and matrix. The composite containing 10% *Tamarindus indica* and 25% *Caryota mitis* leaf stalk fiber, on the other hand, exhibits shrinkage formation at the fiber/matrix interface, improving the tensile strength of the composites made of natural fibers. Because of its rougher fiber surface than tamarind, *Caryota mitis* leaf stalk fiber also shows greater physical bonding with the matrix in this investigation. It can be deduced that fiber pullouts during failure are less common in composites with strong interfacial adhesion [17,38].

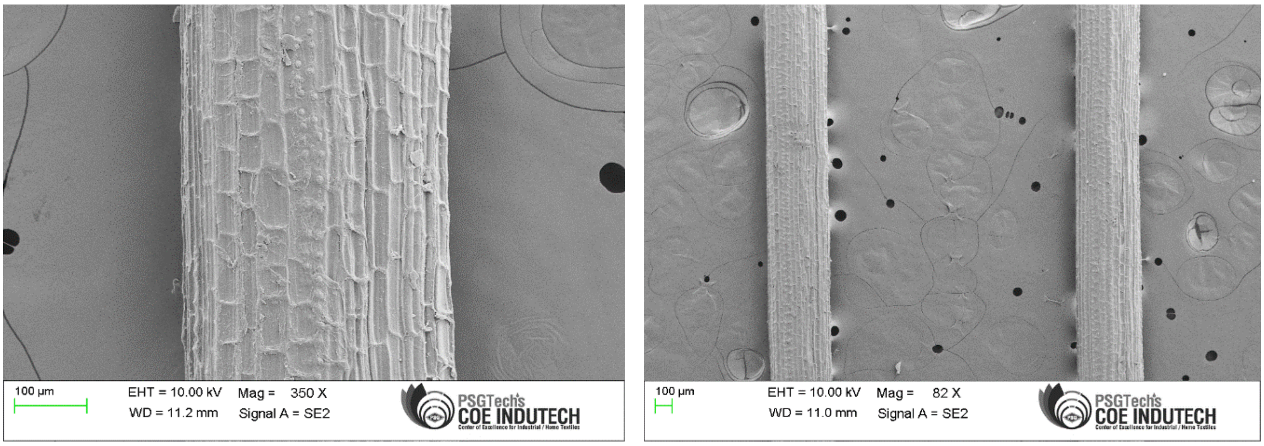

Figure 8. SEM images of *Caryota mitis* and *tamarindus indica* reinforced epoxy composites.

Conclusions

This study compares the tensile strength, impact strength, hardness, microstructure, TGA-DTA, and wear properties of epoxy hybrid composites reinforced with *Tamarindus indica* powder and *Tamarindus indica* leaf stalk fiber. Composites with 25% *Tamarindus indica* and 20% *Caryota mitis* fiber showed the highest tensile strength. Increasing *Caryota mitis* content enhanced the hardness, reaching the highest value at 25%. Microstructure studies revealed stronger interfacial bonding with higher *Tamarindus indica* content, uniform fiber distribution, and fiber pull-out dominating over fiber breakage. The composite with 20% *Caryota mitis* fiber exhibited the best impact strength. Wear tests showed that the wear rate decreased with increased load. TGA revealed weight loss at various temperature stages, corresponding to different composite constituents. These findings suggest that *Caryota mitis* and *Tamarindus indica* fiber composites have superior properties compared to glass and other common fibers, making them potential alternatives in aircraft applications.

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Disclosure Statement

No potential conflict of interest was reported by the authors.

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