## Research Article

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# Effect of fiber orientation and elevated temperature on the mechanical properties of unidirectional continuous kenaf reinforced PLA composites

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**Abstract:** Limitation in practical applications of biopolymer–fiber composite is mainly at higher temperatures. Thus, this study highlights the effects of fiber orientation on the durability of polylactic acid (PLA) reinforced with unidirectional (UD) continuous kenaf fibers at elevated temperatures. PLA and long kenaf fiber were fabricated using the hot-pressing method and stacked at fiber orientations of 0°, 45°, or 90°, relative to the tensile force. Dynamic mechanical analysis of the composites shows excellent anti-shock and temperature-resistant properties of the composite. UD PLA–kenaf composites with a 0° fiber orientation showed an ultimate tensile of ~190 MPa and a flexural strength of ~235 MPa, and the strength of the composite was able to retain up to 120°C temperature. The

debonding behavior of the fiber from the matrix (fiber pull-out) supported by microscopy proved that interfacial failure occurs from the local strains, which initiate cracking. Interfacial failure and stress transfer have caused a remarkable reduction in composite strength when fibers were oriented at 90°. Hence, this current improvement in the performance of the UD PLA–kenaf fiber composite may potentially replace conventional synthetic fibers, especially for structural automotive applications.

**Keywords:** natural fiber, fiber orientation, polymer composites, continuous fiber, mechanical properties

## 1 Introduction

Natural fiber composites are widely used in various fields because of their prominent mechanical and thermal properties. The development of complete biocomposites or "green composites" has recently gained increased attention in efforts to create a sustainable environment [1,2]. Alhijazi et al. [3] stated that the momentum of research interest regarding natural fiber composites specifically in finite element analysis has increased from the Year 2004 until 2020. The applications of green composites in the automotive industry, however, have brought about a series of design issues and challenges. Nevertheless, the unique hierarchical microstructures of distinct plant fibers were found as one of the essential elements that control the functionality and the qualities of the end product [4]. The core materials in automotive products made up of plant fibers are not only recyclable but also have outstanding mechanical properties and are easy to manufacture. Fiber composites for automotive applications were not only used in an interior panel, but also in some of the body parts, which require high strength to endure the high impact such as rear or front bumper [5,6]. In

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Table 1: Comparison of the tensile and flexural properties of bast fiber-reinforced biopolymer composites as reported in the literature (\*indicate chemically treated fibers)

Composites	Fiber content and types	Tensile strength (MPa)	Young's modulus (GPa)	Flexural strength (MPa)	Tensile strength (MPa) Young's modulus (GPa) Flexural strength (MPa) Flexural modulus (GPa) Refs	Refs
PLA-Hemp*	35 wt% <sup>*</sup> in UD	85.4	12.6	84.8	6.4	[36]
PLA-Jute*	50 wt% <sup>*</sup> in UD	210	7.5	230	15	
PLA-Flax*	40 vol% woven fabric	72.2	13	ı	1	[30]
Bio-epoxy-Flax*	50 vol% woven fabric	185	13.8	ı	ı	[37]
PP-Kenaf	50 wt% in UD	160	12	110	8.5	[32]
Commercial composites (PP-Kenaf)	50 wt% in random mat	30.5	2.56	39.4	4.5	[11]
PLA-Kenaf	50 wt% in UD	190.92	16.3	235.42	19.01	This study

previous studies, it shows that unidirectional (UD) long fiber could retain greater mechanical strength as compared to short fiber (refer to Table 2). Nevertheless, while using a long UD fiber, insightful research on fiber orientation is needed as it may either improve or deteriorate the whole strength of the structure. During in-service applications, transverse loads may also be present. Therefore, factors that may influence the transverse strength, which include properties of fiber–matrix, fiber–matrix bond strength, and voids, should be further studied. Improving the factors that affect the transverse strength may potentially widen the usage of long fiber in many other parts of exterior automotive components.

The plant-type fibers, kenaf fibers, are commercially used as substitutes for synthetic fibers in automotive interiors. These fibers provide strengths and moduli of as high as 930 and 53 MPa, respectively, while showing a relatively low weight density [7]. In addition to high specific strength and stiffness, composites reinforced with natural fibers may be produced at a lower cost than other synthetic fibers, such as those made from glass and carbon fibers [8]. By having a lower cost of materials, the cost of production as well as the price of any products made up of this composite fiber will be reduced. Thus, the demand of these composites is predicted to increase over the next few years, and current research efforts are aimed at developing components from 100% biocomposites utilizing biosourced polymers as a matrix.

The physical properties, strength, and stiffness of fiber-reinforced polymer composites are governed by many factors, including the fiber/matrix characteristics, treatment [5], volume fraction [9], fiber length [10], fiber orientation [11], fiber-matrix interfacial adhesion [12], and production process [13,14]. Fiber orientation is a crucial element influencing the composite characteristics of long fibers. Continuous and well-aligned fiber composites present highly anisotropic mechanical properties. The mechanical properties of UD, aligned fiber are known to be much higher than those of randomly oriented fibers, depending on the load

**Table 2:** Comparison of mechanical properties of fabricated PLA-kenaf and PP-kenaf composites using the hot press method at 120°C

Mechanical properties	PLA-kenaf**	PP-kenaf** [39]
Tensile strength (MPa)	~73	~90
Flexural strength (MPa)	~55	~50
Young's modulus (GPa)	~7.6	~7.5
Flexural modulus (GPa)	~13	~6.0

<sup>\*\*</sup>Kenaf fibers were chemically untreated and combed using the same parameters [39].

applied to the composite samples. Shah used Ashby plots to prove that UD plant fiber-reinforced polymers have tensile strengths greatly exceeding (by 20 times) those of randomly oriented plant fiber-reinforced polymers [15].

During service, composites may be subjected to different temperatures in their environment. Obtaining a better understanding of the uncertainties and complexities of the thermal stability of such materials is essential to predict their durability during the design phase and forming process [16]. Indeed, the performance of green composites exposed to high temperatures is a matter of great concern. The strength and stiffness of these composites are severely deteriorated at moderate temperatures, especially when the matrix polymer features a glass transition temperature  $(T_g)$ [17,18]. Hence, further studies on the behavior of biocomposites over a wide temperature range are necessary because structural failure may occur when the strength or stiffness of the material decreases below a certain value.

The effects of temperature on the mechanical characteristics of fiber-reinforced polymers are still poorly understood, and reports on this subject are relatively limited. Most of the studies, particularly those describing UD fiber reinforcement, refer to non-structural parts and conducted testing under room temperature. Studies addressing structural biocomposites reinforced with fibers with different orientations and under a wide temperature range are relatively scarce [13,16,19].

Therefore, this work will be focusing on variations in the temperature condition of long kenaf fiber-reinforced polylactic acid (PLA) composites as a function of fiber orientation through tensile and flexural tests. Thermal and mechanical properties of PLA-kenaf composites, as well as interactions between fiber and polymer, were studied. Scanning electron microscopy (SEM) was applied to evaluate the effect of interactions between the fibers' orientation and matrix, thus reflecting remarkable changes in the physical and mechanical characteristics of the composite. This combing technique without any chemical treatment was expected to form an enhanced strength of PLA-kenaf composite.

# 2 Materials and methods

## 2.1 Materials

The kenaf fibers used in this work were provided by a local supplier (Innovative Pultrusion Sdn. Bhd., Senawang, Negeri Sembilan, Malaysia). The fibers were obtained from bast (density, 1.29 g·cm<sup>-3</sup>) and used as received without surface treatment. PLA (density, 1.25 g·cm<sup>-3</sup>; melting temperature, 170°C) in the form of pellets and powder was supplied by Shenzhen Esun Ltd (China). The PLA pellets were pressed into slices before stacking for composite fabrication.

## 2.2 Fabrication of composites

Continuous kenaf fibers were subjected to drying for 6 h in an oven set to 80°C prior to sample fabrication to eliminate remnant moisture [19]. The kenaf fibers were combed manually [18], aligned in a single direction, and then cut into lengths of 175 mm according to the size of the hot press mold (175 mm  $\times$  175 mm  $\times$  2 mm). The fibers were sandwiched together with the PLA slices and powder. Misorientation of the fibers in the matrix was minimized by ensuring careful alignment at a single direction of 0°, 45°, or 90° relative to the direction of tensile load applied and stacked in a flash picture-frame mold. The fiber and matrix were pressed between the plates of a 50-ton hot-press machine at a temperature of 200°C and a pressure of 3 MPa, respectively, for 7 min and then cooled [20]. The PLA-kenaf weight ratio was kept at 50:50, which is the optimum composition of PLAkenaf composites [21]. Tholibon et al. observed that this weight ratio results in long PP-kenaf fiber composites with optimal mechanical properties [22]. Schematic illustration of composite arrangement during hot pressing is shown in Figure 1.



Figure 1: Schematic illustration of composite arrangement during hot press process.

## 2.3 Characterizations

#### 2.3.1 Thermogravimetric analysis (TGA)

TGA of all the samples was carried out using a Netzsch TG 200 F3 Tarsus under the nitrogen atmosphere at the flow rate of 50 mL·min<sup>-1</sup>. Samples of mass ranging from 6 to 7 mg were placed on the platinum pan and were heated from room temperature to 500°C at the heating rate of 10°C·min<sup>-1</sup>. This TGA was run based on the ASTM E1131 standard (Standard Test Method for Compositional Analysis by Thermogravimetry) [23] to determine the decomposition temperature of PLA, kenaf, and the composite.

### 2.3.2 Dynamic mechanical analysis (DMA)

DMA test was carried out using Q800 (TA Instruments) instrument equipped with a dual-cantilever fixture. The frequency was set to 1 Hz, and heating was conducted from room temperature to 140°C with a heating rate of  $1^{\circ}\text{C}\cdot\text{min}^{-1}$ . The composites were cut into dimensions of 60 mm  $\times$  12 mm  $\times$  2 mm. In this test, only the UD PLA–kenaf composite with 0° orientation was used as a representative for other variances due to the same fiber composition, and pure PLA was used as a control. The

values of loss modulus, storage modulus, and Tan  $\delta$  obtained were plotted.

#### 2.3.3 Mechanical testing

The composites were cut into specimens of  $115\,\mathrm{mm} \times 20\,\mathrm{mm}$  for tensile and flexural testing according to ASTM D638-99 [24]. The tensile tests were carried out using a Zwick/Roell Z100 universal tensile tester under a 100 kN load. Flexural tests were conducted using an INSTRON 5567 instrument according to ASTM D790-03 [25] with a 30 kN load. The mechanical tests were carried out at room temperature under a crosshead speed of 2 mm·min $^{-1}$ . Five specimens measuring 20 mm wide and 2 mm thick were prepared from each composite and subjected to flexural and tensile testing. The experimental setup and composite specimens are shown in Figure 2.

A thermal chamber was installed on the mechanical tester for thermal exposure studies. The samples were placed in the chamber and subjected to tensile and flexural testing at different temperatures of 30, 60, 90, and 120°C and the same crosshead speed (i.e., 2 mm·min<sup>-1</sup>). These temperatures were selected by adding 30°C increments to the ambient temperature until 70% of the melting temperature of the polymer was reached. Pure PLA specimens were also fabricated and tested as controls.

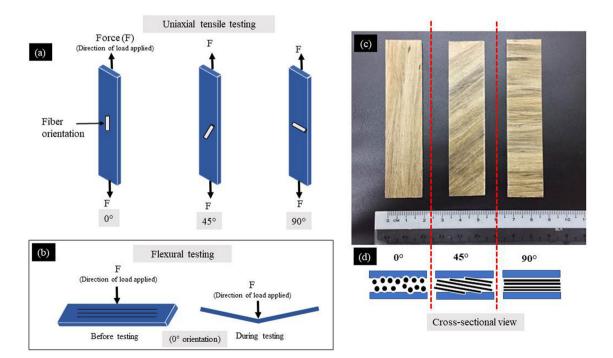


Figure 2: Schematic of samples in the (a) tensile and (b) flexural tests. (c) Images of PLA-kenaf composites with different fiber orientations. (d) Cross-sectional illustration of the composite samples.

#### 2.3.4 Morphology

Field emission scanning electron microscopy (FESEM; SUPRA 55 VP; acceleration voltage, 3 kV) was carried out to observe the morphology of the cross-sectional tensile fracture surfaces of all PLA–kenaf composites. Charging during sample observation was avoided by sputter-coating the specimens with gold.

# 3 Results and discussion

# 3.1 Thermal stability

TGA curves in Figure 3 demonstrated the thermal stability of PLA, kenaf fibers, and PLA-kenaf composite. The decomposition of PLA components was indicated by the high weight loss when the temperature reached 331.9°C. Meanwhile, early weight loss of kenaf fiber up to 120°C was due to the moisture evaporation and the decomposition of the fiber components such as hemicelluloses, waxes, and pectin, especially for untreated fibers that have greater moisture absorption [26]. This is followed by the glycosidic linkages of cellulose starting at 250°C and lignin oxidation at temperatures after 300°C (Figure 3). For PLA-kenaf composite, the weight loss of about 4% at 80°C can be observed from the TG curve PLA-kenaf composite, which is essentially due to moisture evaporation that causes the hygroscopic behavior of PLA. A high decrease in the second onset decomposition of PLA begins in the temperature range from 319.2 to 331.9°C consequent to the decomposition of 50 wt% kenaf fibers. Overall, the maximum degradation rate is between 347 and 365°C. A similar decrease in PLA's maximum degradation temperature with the existence of kenaf fibers was also reported by Ridzuan et al. [27]. These results indicate that PLA–kenaf composite is stable to be mechanically tested at different temperatures up to 120°C.

The dynamic mechanical properties of PLA composites under the influence of temperature were studied using DMA. The load-bearing capacity of the composites, which refers to their ability to store applied energy, was examined by observing changes in their storage modulus (E'). Figure 4 compares the elastic part through E' value of pure PLA and the PLA-kenaf composite with a fiber orientation of 0°. The E' of the PLA-kenaf composite was fourfold greater than that of pure PLA at 40°C. Moreover, the E' of the composite (6,700 MPa) remained higher than that of pure PLA (2,100 MPa) at the glass transition region (~60°C) and 120°C (Figure 4). A high E' reflects the extent and effectiveness with which stress is transferred along the kenaf fibers and PLA matrix [28]. In the composite, the kenaf fibers act as sponges that prevent energy in the composite from dissipating. The large reduction in E' of the composite at higher temperatures is initially due to the loss of stiffness in matrix, followed by the reduction of fiber reinforcement stiffness. This may consequently cause reinforcement-matrix load transmission failure, which then deteriorates the whole structure of the composite.

The viscous properties of a material could be described by its loss modulus (E'') and loss factor (tan  $\delta$ ). This loss modulus (E'') determines the ability of a material to dissipate

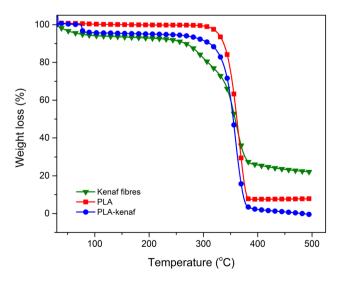


Figure 3: TGA curves for PLA matrix, kenaf fibers, and PLA-kenaf composite.

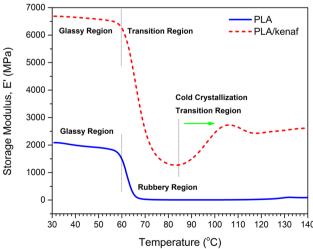


Figure 4: Storage modulus of pure PLA and the UD PLA-kenaf composite with a 0° fiber orientation.

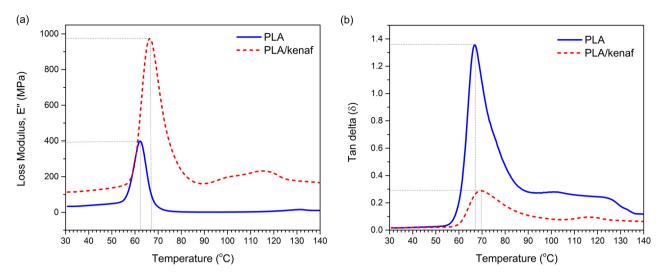


Figure 5: (a) Loss modulus and (b) loss factor (tan  $\delta$ ) of pure PLA and the UD PLA-kenaf composite with a 0° fiber orientation.

some applied energy. As shown in Figure 5a, the lower peak E" of pure PLA (~398 MPa) compared with that of the PLAkenaf composite was due to changes in the molecular motion of polymer particles under an applied energy [29]. The addition of UD kenaf fibers as reinforcements improved the peak E" of the PLA-kenaf composite to ~978 MPa, which may be attributed to an increase in molecular friction in the sample.  $T_g$  is defined as the temperature at which damping and E'' attain their maximum values. The addition of UD kenaf fibers to PLA caused a slight improvement in the  $T_{\sigma}$ of the resulting composites. These results confirm that kenaf fibers can reduce the molecular mobility of the matrix chains and improves the anti-shock properties of the composite [30]. In addition, it shows that UD kenaf fibers and PLA have good compatibility, which will enhance interfacial filler-matrix adhesion and improves stress transfer that could strengthen the whole structure of the composite [28,31].

Figure 5b shows variations in the mechanical loss factor ( $\tan \delta$ ) of neat PLA and the UD PLA-kenaf composite with a 0° fiber orientation. These dynamic parameters not only illustrate the compliance loss or dynamic viscosity but also demonstrate the dynamic fragility of the material. Compared with that of the PLA-kenaf composite, the peak  $\tan \delta$  of neat PLA was higher. Compared with the composites, PLA featured greater molecular mobility and higher viscosity. The tight adhesion between the polymer matrix and kenaf fibers dramatically reduces the  $\tan \delta$  of the composite and, hence, its molecular mobility [28]. This reduction of the maximum value of  $\tan \delta$  is associated with the increment in storage and loss modulus, which signifies the enhancement of PLA-based composite stiffness due to the presence of fiber. A lower peak

tan  $\delta$  also reflects a reduction in damping capacity. The results reveal that the composites developed in this work have good load-bearing capacity. Previous researchers demonstrated similar reductions in the damping capacity of PLA-fiber composites as a function of temperature [30]. Taken together, the results of DMA indicate that PLA-kenaf fiber composites may have great potential applications as load bearing over the temperature range of 60–70°C.

## 3.2 Tensile properties

The effects of fiber orientation and temperature on the composites were assessed by examining their tensile behavior. Figure 6a plots the variations in tensile strength of pure PLA and the PLA-kenaf composites. Among the samples obtained, composites bearing fibers oriented at 0° showed the highest tensile strength (191 MPa) and modulus (16.3 GPa) at 30°C. The results reveal that incorporating long kenaf fibers effectively improves the strength of the PLA composites under a tensile load. At ambient temperature, the strength of the PLA-kenaf composite with fibers oriented at 45° was 65.79% lower than that of pure PLA. This decrease in strength was even more evident in composites with fibers oriented at 45° and 90° (Figure 6a and b). Such results indicate that fibers oriented at 0° can better resist applied stress and transfer the same to other fibers in the matrix compared with fibers oriented at 45° and 90°. Composites with fibers aligned diagonally or perpendicularly (i.e., 45° and 90°) to the applied force easily split along their longitudinal axis. Thus, fibers with these orientations fail as a reinforcing agent because load transfer between the fiber and PLA matrix is restricted. In this case, the stress transfer

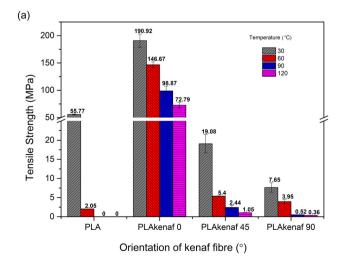
will not successfully happen; thus, the strength of the composite depends entirely on the polymer [32]. This supposition is supported by Figure 6b, which clearly shows that PLA–kenaf composites with a fiber orientation of 0° are 3.5 times stronger than pure PLA. Therefore, failure occurs because the strength of the polymer is much lower than that of composites reinforced with kenaf fibers, especially those with fibers oriented parallel to the direction of the applied forces.

The PLA-kenaf composites with fibers oriented at 45° and 90°, respectively, demonstrated approximately 48 and 27% higher Young's moduli than pure PLA, as shown in Figure 6b. This finding indicates that these composites have greater stiffness compared with pure PLA. A better stiffness may be explained by the structure–property relationship, which is influenced by cellulose contents and interfacial actions between fiber and polymer that allow the transmission of load throughout the matrix [33]. Some other researchers found that damage to the inner layer of fibers oriented at different angles may be attributed to a reduction in the composite strength [22,29].

The tensile failure mode of composites reinforced with kenaf fibers of different orientations is illustrated in Figure 7. Different fiber orientations exhibited distinct stress transfer behaviors. Load transfer occurs along the direction of fiber orientation. Composites with fibers oriented in the 0° direction experience low shear stress at the fiber–matrix interface. Hence, this fiber orientation results in random tensile breakage and causes debonding cracks across the fibers. The debonding cracks grow, propagate into neighboring fibers, and eventually combine into a single large crack (Figure 7a). This failure type is usually a consequence of the high strength of UD composites. As the fiber orientation is changed to 45° or 90° (i.e.,

perpendicular to the direction of the applied load), the interfacial compatibility between the fibers and matrix decreases, and a failure in stress transfer occurs. Under applied forces, stress is propagated along the fiber direction, resulting in tensile failure, as shown in Figure 7b and c. In this case, weakness at the fiber surface and matrix interface is the main cause of failure. Fractures occurring at the fiber–matrix interface may be attributed to matrix shear failure. The interlaminar shear strength and fracture toughness indicate the interlaminar properties of the composite. Stress transmission along the fiber direction is disrupted and causes significant reductions in the strength of the composite, as shown in Figure 6a. Similar phenomena of the tensile fiber composite breakage have been reported in the literature [29].

The strength of the PLA-kenaf composites was much higher than that of pure PLA at temperatures ≥60°C (Figure 6a). The transition of PLA from brittle to rubbery take place as the temperature increases to 60°C. PLA softened and started to deform at 60°C as some portion of the amorphous part of PLA began to slide each other, but the chain was still highly entangled. Disentanglement of the PLA chains occurs as the temperature increases to 90°C and diminished its strength. The results reflect the temperature dependence of some mechanical properties of PLA-kenaf composites. Between tensile strength and Young's modulus, the former appears to be more sensitive to temperature changes than the latter, mainly because the nature of the material under deformation influences its tensile strength under high-loading conditions. At a high temperature of 120°C, Young's modulus of the PLAkenaf composites remained higher (~72 MPa) than that of pure PLA (~56 MPa). According to the literature, this range



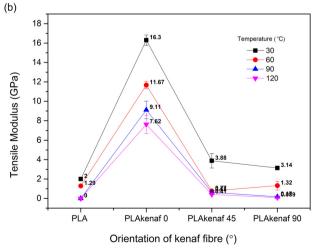


Figure 6: (a) Tensile strength and (b) Young's modulus of pure PLA and PLA-kenaf composites at different temperatures.

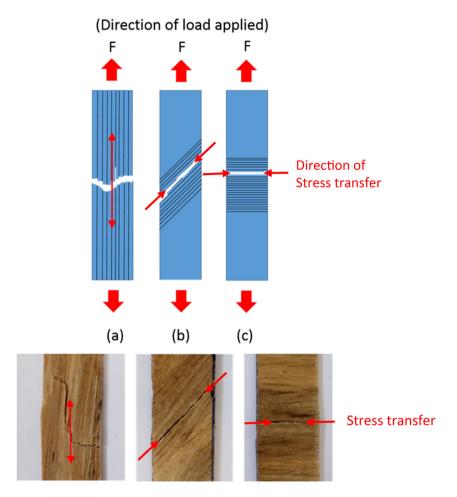


Figure 7: Failure of the PLA-kenaf composites with fiber orientations of (a) 0°, (b) 45°, and (c) 90° during tensile testing.

of tensile strength is sufficient for interior automotive parts, such as dashboards or trunk covers, which require a tensile strength of approximately 30 MPa [22].

## 3.3 Flexural properties

The flexural strength of UD PLA-kenaf fiber composites showed significantly increased strength (by 232%) and a high Young's modulus (19.01 GPa) when the fibers are oriented at 0° (Figure 8). Interestingly, the strength of all other composites with different orientations was much lower than that of PLA. The stiffness of the composite with fibers oriented at 0° was mostly retained at 60°C and showed only a 6.05% reduction in modulus compared with that obtained at 30°C. The strength of the PLA-kenaf composites was 70× times higher than that of pure PLA even at a temperature of 60°C (Figure 8a).

In Figure 8b, for example, the flexural moduli of the PLA-kenaf composite with a 0° fiber orientation were 9.11 and 7.42 GPa at 90 and 120°C, respectively. This finding

reflects the ability of long kenaf fibers to strengthen the PLA composite structure. However, at an ambient temperature of 30°C, the UD PLA-kenaf composites showed an 84.5% reduction in flexural strength when the fibers are at a 45° orientation and nearly fail (4.83 MPa) when the fibers are at a 90° orientation. These findings may be attributed to the inability of the fibers to withstand stress given their alignment along the direction parallel to the flexural load.

The composite failure is not catastrophic as the fiber will not fracture at the same time. In this case, cracks were initiated at the fiber-matrix interface and quickly propagated along the fiber direction, separating the components [34]. Obviously, the fiber breakage between the layers of UD kenaf causes strength degradation. Wu et al. [11] reported that this fiber breakage is usually initiated from the upper side that experiences compression properties and usually will have lower flexural strength than the tension side (lower side), particularly if it comprises many layers of fiber-polymer. This is the reason for the variance in fracture strength of brittle fiber materials.

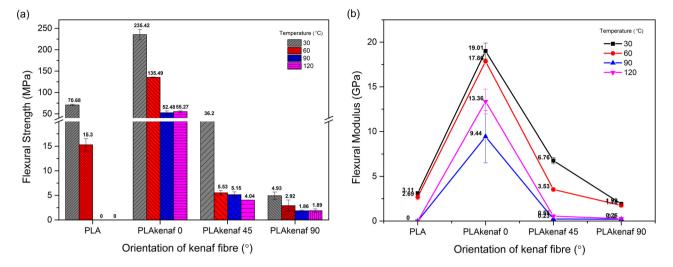


Figure 8: (a) Flexural strength and (b) modulus of pure PLA and PLA-kenaf composites at different temperatures.

Kenaf fibers with 0° orientation show high stiffness, which could be attributed to their excellent uniformity and beneficial for load distribution. As the plastic deformation occurred, the fractured fiber was still embedded in the matrix and remained supporting the diminished load.

The flexural strength of a fiber-reinforced composite is remarkably influenced by the presence of defects and fiber strength; thus, in the absence of the former, the flexural strength may only be attributed to the latter. By comparison, under a tensile load, all of the composite fibers equally experience the load; in this case, the weakest fibers are the first to fail when their stress limit is exceeded. These differences explain why the flexural strength of a material is typically higher than its tensile strength. A narrative review done by Alkbir et al. [35] stated that variations in fiber strength of natural fiber-reinforced polymer composite may be influenced by factors such as fiber aspect ratio, extraction method, fiber treatment, fiber orientation, and the dimensional stability of the fibers used.

Table 1 compares the present composite with other bast fiber-reinforced biopolymer composites, including kenaf, hemp, flax, and jute, at different fiber ratios. The tensile and flexural properties of the PLA-kenaf composites obtained in this study are better than those of some composites. The mechanical strength of the composites developed in this study was much higher than those of commercial kenaf composites used for automotive products [11]. The chemically untreated kenaf composites in this study demonstrated higher Young's modulus and flexural properties compared with treated PLA-jute composites with the same fiber content. Interestingly, the comparison also showed that PLA composites reinforced with combed kenaf fibers possess good

mechanical properties even without any chemical surface treatment. This translates to lower costs during composite fabrication.

The mechanical properties of this kenaf reinforced composites outperformed those of bio-thermoset polymer even though bio-thermoset matrix polymers, such as bio-epoxy, are generally more favored than bio-thermoplastics because the former have better thermal resistance and this epoxy could potentially produce high-performance composites [38]. Moreover, bio-thermoplastics, such as PLA, are the most suitable choice for producing green composites because they do not require curing and combine the benefits of fast processing and biodegradability.

Table 2 reveals that the strength of PLA-kenaf composites with 0° orientation was comparable with that of PP-kenaf composites at 120°C. The PLA-kenaf composites developed in this study showed better mechanical strength and a higher modulus than PP-kenaf composites manufactured via the same method. This finding may be attributed to the higher fiber-matrix compatibility of PLA compared with that of PP. Thus, PLA-kenaf composites appear to be an excellent alternative to PP-kenaf. The composites developed in this study were completely biodegradable but demonstrated mechanical properties similar to those of petroleum-based composites.

Interestingly, the comparison also showed that PLA composites reinforced with combed kenaf fibers possess good mechanical properties even without any chemical surface treatment. This translates to lower costs during composite fabrication. The mechanical properties of the kenaf-reinforced composites outperformed those of biothermoset polymer.

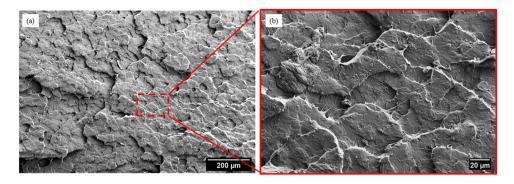


Figure 9: Micrograph of PLA failure sections at (a) low and (b) high magnification.

# 3.4 Morphological analysis

FESEM was employed to collect micrographs of the fracture surfaces of PLA (Figure 9) and the PLA–kenaf composites following tensile testing (Figures 9 and 10). Figure 9a under its magnification in Figure 9b shows that pure PLA undergoes brittle fractures with a rough surface. PLA–kenaf composites with fibers oriented at 0° revealed fiber breakage and pull-out, as shown in Figure 10a–c. By comparison, the kenaf fibers were dispersed and well adhered to the PLA matrix where the kenaf fiber was appropriately imprinted into the PLA matrix (Figure 10b). These features contribute to the high strength of the composites. The results indicate

that the fibers, especially those oriented parallel to the direction of force, were the main load carriers in the composites. Closer examination revealed brittle fractures in the matrix, fiber pull-out, shear-out, extensive fiber breakage, and matrix fractures. Good adhesion between the fibers and the PLA matrix contributed to the superior strength of the composites (Figure 10c).

A dramatic decrease in tensile properties was observed in composites with fiber orientations of 45° and 90°, as illustrated in Figure 11. The failure modes of exposed kenaf fiber bundles reflect brittle fractures, which indicates that interfacial failure is related to fiber delamination without matrix separation. Weak adhesion between the fibers and

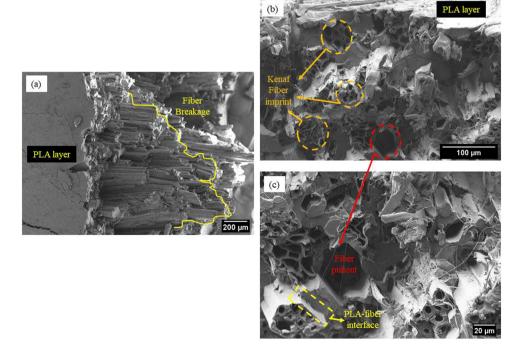
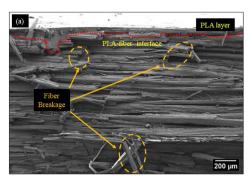


Figure 10: SEM micrographs of the tensile fractures of PLA-kenaf composites with a fiber orientation of 0°: (a) Top view, (b) cross-sectional view, and (c) detailed view.



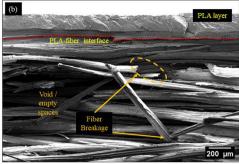


Figure 11: SEM micrographs of the tensile fractures of PLA-kenaf fiber composites with fiber orientations of (a) 45° and (b) 90°.

the matrix causes interfacial failure. Some factors contributing to this type of failure include short fiber lengths, low adhesion strength, and the absence of fiber interlocking [40]. In this case, the PLA matrix, rather than the kenaf fibers, functions as the main load carrier in the composite. A high degree of cracking was observed in composites with fibers oriented at 90°. This indicated that debonding of fibers from the matrix (fiber pull-out) occurs when the interfacial stresses exceed the interfacial strength of fiber-matrix composites. Local strain produced by the interfacial failure may initiate cracking at the fiber-matrix interface. Fibers laid perpendicular to the load easily split along their longitudinal axis, separating them from the matrix [37]. In this case, the fibers cannot provide reinforcement functions, and load transfer is restricted and controlled by the matrix. Among the samples prepared, PLA-fiber composites with a fiber orientation of 90° showed the greatest fiber misorientation and fiber breakage, accompanied by the appearance of numerous voids (Figure 9b); these features demonstrate the low tensile properties of this composite. During the fabrication process, sometimes the polymer matrix was unable to expel the existence of air in the kenaf fiber that creates the void. Thus, the rolling process of the wet composite was suggested to assist the air pocket removal [35].

# 4 Conclusion

The present study investigates the physical and mechanical characteristics of UD continuous PLA–kenaf composites with fiber orientations of  $0^{\circ}$ ,  $45^{\circ}$ , and  $90^{\circ}$  under elevated temperatures. DMA, mechanical test under variant temperatures, and surface morphologies were carried out to determine the durability of the composite. The DMA results showed that the kenaf fibers positively affect the thermal responses of the composites by endowing the latter with a high E' and E'', shifting the  $T_g$ , and providing

great potential applications as load-bearing capacity over temperature range of 60-70°C. The optimal tensile and flexural strength were obtained at 0° orientation with ~190 and ~235 MPa, respectively, at room temperature. The temperatures remarkably affect the properties of the PLA-kenaf composites; specifically, great reductions in strength and modulus were observed at higher temperatures with the orientations of 45° and 90°. The failure of fiber reinforcement within the composite was due to the existence of voids and debonding of fibers during the mechanical test. This has caused reinforcement-matrix load transmission failure that deteriorates the whole structure of the composite. Nevertheless, PLA-kenaf composites with a 0° fiber orientation have sufficiently good mechanical properties at temperatures up to 120°C compared to those of commercial PP-kenaf composites. Therefore, a PLA-kenaf composite with UD alignment is ideal for use in structural interior components of automotive products. Future studies may be focusing on the moisture absorption and weathering resistance of these composites to assess their potential as alternative materials for exterior automotive components.

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## References

- Yildirim, A. and H. Acay. Applications of biodegradable green composites. *Green Composites*. Springer, Singapore, 2021, pp. 373–392.
- [2] Rafiee, K., H. Schritt, D. Pleissner, G. Kaur, and S. K. Brar. Biodegradable green composites: It's never too late to mend. Current Opinion in Green and Sustainable Chemistry, Vol. 30, 2021, id. 100482.
- [3] Alhijazi, M., Q. Zeeshan, Z. Qin, B. Safaei, and M. Asmael. Finite element analysis of natural fibers composites: A review. Nanotechnology Reviews, Vol. 9, 2020, pp. 853–875.
- [4] Koronis, G., A. Silva, and M. Fontul. Green composites: A review of adequate materials for automotive applications. *Composite Part B: Engineering*, Vol. 44, 2013, pp. 120–127.
- [5] Mei, C., Y. Fan, L. Mei, and G. Luo. Surface characteristics of modified poplar wood-flour and its effect on the mechanical properties of wood polymer composites. *Reviews on Advance Materials Science*, Vol. 33, 2013, pp. 203–210.
- [6] Wu, Y., B. Tang, K. Liu, X. Zeng, J. Lu, T. Zhang, et al. Enhanced flexural properties of aramid fiber/epoxy composites by graphene oxide. *Nanotechnology Reviews*, Vol. 8, 2019, pp. 484–492.
- [7] Akil, H. M., M. F. Omar, A. A. M. Mazuki, S. Safiee, Z. A. M. Ishak, and A. A. Bakar. Kenaf fiber reinforced composites: A review. *Materials and Design*, Vol. 32, 2011, pp. 4107–4121.
- [8] Xie, H., H. Fang, W. Cai, L. Wan, R. Huo, and D. Hui. Development of an innovative composite sandwich matting with GFRP facesheets and wood core. *Reviews on Advance Materials Science*, Vol. 60, 2021, pp. 80–91.
- [9] Ku, H., H. Wang, N. Pattarachaiyakoop, and M. Trada. A review on the tensile properties of natural fiber reinforced polymer composites. *Composite Part B: Engineering*, Vol. 42, 2011, pp. 856–873.
- [10] Graupner, N., G. Ziegmann, F. Wilde, F. Beckmann, and J. Müssig. Procedural influences on compression and injection moulded cellulose fibre-reinforced polylactide (PLA) composites: Influence of fibre loading, fibre length, fibre orientation

- and voids. Composites Part A: Applied Science and Manufacturing, Vol. 81, 2016, pp. 158-171.
- [11] Ma, Y., L. Wu, L. Yu, E. A. Elbadry, W. Yang, X. Huang, et al. Effect of fiber breakage position on the mechanical performance of unidirectional carbon fiber/epoxy composites. *Reviews on Advanced Materials Science*, Vol. 60, 2021, pp. 352–364.
- [12] Chanda, S. and D. S. Bajwa. A review of current physical techniques for dispersion of cellulose nanomaterials in polymer matrices. *Reviews on Advanced Materials Science*, Vol. 60, 2021, pp. 325–341.
- [13] Sulong, A. B. S., N. A. Mohd Radzuan, M. R. Mamat, I. Tharazi, D. Tholibon, R. Dweiri, et al. Kenaf reinforced PLA composite thermoforming: A numerical simulation. *International Journal of Integrated Engineering*, Vol. 10, 2018, pp. 15–20.
- [14] Mohd Radzuan, N. A., N. F. Ismail, M. K. Fadzly Md Radzi, Z. B. Razak, I. B. Tharizi, A. B. Sulong, et al. Kenaf composites for automotive components: Enhancement in machinability and moldability. *Polymers*, Vol. 22, 2019, pp. 1–10.
- [15] Shah, D. U. Developing plant fibre composites for structural applications by optimising composite parameters: A critical review. *Journal of Materials Science*, Vol. 48, 2013, pp. 6083–6107.
- [16] Mukherjee, T. and N. Kao. PLA based biopolymer reinforced with natural fibre: A review. *Journal of Polymers and the Environment*, Vol. 19, 2011, pp. 714–725.
- [17] Pantani, R. and L. S. Turng. Manufacturing of advanced biodegradable polymeric components. *Journal of Applied Polymer Science*, Vol. 132, 2015, pp. 1–15.
- [18] Tholibon, D. Hot pressing of unidirectional kenaf fibres reinforced polypropylene composite. Universiti Kebangsaan Malaysia; 2018.
- [19] Murariu, M. and P. Dubois. PLA composites: From production to properties. Advance Drug Delivery Reviews, Vol. 107, 2016, pp. 17-46.
- [20] Tharazi, I., A. B. Sulong, F. M. Salleh, A. H. Abdullah, and N. F. Ismail. Application of response surface methodology for parameters optimization in hot pressing kenaf reinforced biocomposites. *Journal of Mechanical Engineering*, Vol. 17, 2020, pp. 131–144.
- [21] Tharazi, I., N. Muhamad, N. F. Ismail, M. K. F. M. Radzi, Z. Razak, D. Tholibon, et al. Effects of fiber content and processing parameters on tensile properties of unidirectional long kenaf fiber reinforced polylactic-acid composite. *Journal of Mechanical Engineering*, Vol. SI 3, 2017, pp. 65-76.
- [22] Radzuan, N. A. M., D. Tholibon, A. B. Sulong, N. Muhamad, and C. H. C. Haron. Effects of high-temperature exposure on the mechanical properties of kenaf composites. *Polymers*, Vol. 12, 2020, pp. 1–13.
- [23] ASTM-E1131-08. Standard Test Method for Compositional Analysis by Thermogravimetry, ASTM International, West Conshohocken, PA, USA, 2008. https://www.astm.org/ DATABASE.CART/HISTORICAL/E1131-08.htm (accessed on 9 August 2020).
- [24] ASTM-D638-99. Standard Test Method for Tensile Properties of Plastics, ASTM International, West Conshohocken, PA, USA, 2000. https://www.astm.org/d0638-99.html (accessed on 4th October 2022).

- [25] ASTM-D790-03. Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials, ASTM International, West Conshohocken, PA, USA, 2003. https://www.astm.org/d0790-03.html (accessed on 13 January 2020).
- [26] Pietro, R., C. Cosimo, C. Francesca, and P. Paola. Biodegradable composites reinforced with kenaf fibers: Thermal, mechanical, and morphological issues. Advances in Polymer Technology, Vol. 10, 2012, pp. 1–10.
- [27] Ridzuan, M. J. M., M. S. A. Majid, M. Afendi, M. N. Mazlee, and A. G. Gibson. Thermal behaviour and dynamic mechanical analysis of *Pennisetum purpureum*/glass-reinforced epoxy hybrid composites. *Composite Structures*, Vol. 152, 2016, pp. 850-859.
- [28] Chee, S. S., M. Jawaid, M. T. H. Sultan, O. Y. Alothman, and L. C. Abdullah. Thermomechanical and dynamic mechanical properties of bamboo/woven kenaf mat reinforced epoxy hybrid composites. *Composites Part B: Engineering*, Vol. 163, 2019, pp. 165–174.
- [29] Lee, S., S. Q. Shi, L. H. Groom, and Y. Xue. Properties of unidirectional kenaf fiber-polyolefin laminates. *Polymer Composites*, Vol. 16, 2010, pp. 1067-1074.
- [30] Nassiopoulos, E. and J. Njuguna. Thermo-mechanical performance of poly(lactic acid)/flax fibre-reinforced biocomposites. Materials and Design, Vol. 66, 2015, pp. 473-485.
- [31] Chee, S. S., M. Jawaid, and M. T. H. Sultan. Thermal stability and dynamic mechanical properties of kenaf/bamboo fibre reinforced epoxy composites. *BioResources*, Vol. 12, 2017, pp. 7118–732.
- [32] Lau., K., H. Pui-yan, Z. Minhao, and D. Hui. Properties of natural fibre composites for structural engineering applications. *Composites Part B-Engineering*, Vol. 316, 2018, pp. 222–223.

- [33] Perić, M., R. Putz, and C. Paulik. Influence of nanofibrillated cellulose on the mechanical and thermal properties of poly(lactic acid). European Polymer Journal, Vol. 114, 2019, pp. 426–433.
- [34] Yong, C. K., Y. C. Ching, C. H. Chuah, and N. S. Liou. Effect of fiber orientation on mechanical properties of kenaf-reinforced polymer composite. *BioResources*, Vol. 10, 2015, pp. 2597–2608.
- [35] Alkbir, M. F. M., S. M. Sapuan, A. A. Nuraini, and M. R. Ishak. Fiber properties and crashworthiness parameters of natural fiber-reinforced composite structure: A literature review. *Composite Structure*, Vol. 149, 2016, pp. 59–73.
- [36] Sawpan, M. A., K. L. Pickering, and A. Fernyhough. Improvement of mechanical performance of industrial hemp fibre reinforced polylactide biocomposites. *Composites Part A: Applied Science and Manufacturing*, Vol. 42, 2011, pp. 310–319.
- [37] Romhány, G., J. Karger-Kocsis, and T. Czigány. Tensile fracture and failure behavior of thermoplastic starch with unidirectional and cross-ply flax fiber reinforcements. Macromolecular Materials and Engineering, Vol. 288, 2003, pp. 699-707.
- [38] Yu, T., S. A. Soomro, F. Huang, W. Wei, B. Wang, Z. Zhou, et al. Naturally or artificially constructed nanocellulose architectures for epoxy composites: A review. *Nanotechnology Reviews*, Vol. 9, 2020, pp. 1643–1659.
- [39] Mohd Radzuan, N. A., D. Tholibon, A. B. Sulong, N. Muhamad, and C. H. C. Haron. New processing technique for biodegradable kenaf composites: A simple alternative to commercial automotive parts. *Composite Part B: Engineering*, Vol. 184, 2020, id. 107644.
- [40] Csikós, Á., G. Faludi, A. Domján, K. Renner, J. Móczó, and B. Pukánszky. Modification of interfacial adhesion with a functionalized polymer in PLA/wood composites. *European Polymer Journal*, Vol. 68, 2015, pp. 592–600.