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Natural Fiber-Reinforced Composites Using Miswak and Kenaf for Sustainable Food Packaging

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ABSTRACT

This study investigates poly(lactic acid) (PLA) hybrid composites reinforced with miswak fiber (MF) and kenaf fiber (KF) and evaluates how fiber ratio affects performance for food-packaging applications. MF and KF were alkali treated to reduce lignin and hemicellulose, then incorporated into a constant 70% PLA matrix. Composite sheets were fabricated by two-stage hydraulic hot pressing with fiber formulations of 30% KF, 20% KF + 10% MF, 10% KF + 20% MF, and 30% MF. The 10% KF + 20% MF hybrid exhibited the best mechanical properties (tensile strength 51 MPa, tensile modulus 6340 MPa, flexural strength 79 MPa, and flexural modulus 5650 MPa), exceeding common thresholds for packaging materials. The results indicate that an optimal MF/KF balance improves fiber-matrix interfacial adhesion and load transfer. Thermogravimetric analysis showed an onset degradation temperature of ~305°C, about 15°C higher than PLA reinforced solely with kenaf fiber (~290°C). The improved thermal stability is attributed to MF's higher lignin content and enhanced interfacial bonding. Overall, MF/KF-PLA hybrids are promising bio-based alternatives to conventional packaging plastics.

摘要

本研究调查了用米什瓦克纤维 (MF) 和红麻纤维 (KF) 增强的聚乳酸 (PLA) 杂化复合材料, 并评估了纤维比例如何影响食品包装应用的性能。MF 和 KF 经过碱处理以减少木质素和半纤维素, 然后掺入恒定的 70% PLA 基质中。复合板通过两阶段液压热压制, 纤维配方为 30% KF、20% KF + 10% MF、10% KF + 20% MF 和 30% MF。10% KF + 20% MF 杂化材料表现出最佳的机械性能 (拉伸强度 51 MPa, 拉伸模量 6340 MPa, 弯曲强度 79 MPa, 弯曲模量 5650 MPa), 超过了包装材料的常见阈值。结果表明, 最佳的 MF/KF 平衡改善了纤维-基体界面粘附和载荷传递。热重分析显示, 起始降解温度约为 305°C, 比仅用红麻纤维增强的 PLA (约 290°C) 高出约 15°C。提高的热稳定性归因于 MF 的木质素含量更高和界面结合增强。总体而言, MF/KF-PLA 混合材料是传统包装塑料的有前景的生物基替代品。

KEYWORDS

Poly(lactic acid); kenaf fiber; miswak fiber; composites; extrusion

关键词

聚乳酸; 红麻纤维; 纤维; 复合材料; 挤出

Introduction

In recent years, increased concern for food security has led to exponential growth in the food business. The food business was compelled to maintain substantial output levels due to this situation. Synthetic polymers originating from petrochemicals, such as polypropylene and polyethylene, are extensively utilized in food packaging due to their mechanical qualities and inexpensive production costs (Dias et al. 2021). However, the overuse of petroleum-based synthetic polymers can result in issues such as environmental pollution and non-sustainable practice (Jali et al. 2023).

Poly(lactic acid) (PLA) is a biodegradable polymer known for its remarkable mechanical properties, making it versatile for various applications. Studies have demonstrated that PLA exhibits excellent tensile, flexural, and impact strength (Subramaniyan et al. 2023). It is considered a promising, sustainable and biodegradable polymer due to its high molecular weight synthesis capability (Ramezani Dana

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and Ebrahimi 2023). This biopolymer has gained attention for its biocompatibility, sustainability, and processability, making it a preferred material in various industrial fields (Taib et al. 2023).

Research on natural fiber composites has significantly increased attention and advancement. Various facets of natural fiber composites have attracted significant attention from researchers, including processing, characteristics, applications, and cost-effectiveness (Ayu et al. 2020; Nurazzi et al. 2021; Rafiqah et al. 2021). These natural fiber composites have several advantageous individual applications and may potentially replace conventional synthetic materials. A composite is classified as a hybrid composite when it contains two or more reinforcing natural fibers within a single matrix. Research indicates that hybrid composites enhance the qualities typically absent in regular composites by incorporating supplementary reinforcements, hence increasing the versatility of the composite (Nurazzi et al. 2020; Yashas Gowda et al. 2022).

Natural fiber from flax, cotton, hemp, jute, kenaf, sisal, banana, ramie, and miswak are commonly extracted to produce bio-composites, contributing to environmentally friendly product development (Nurazzi et al. 2021). Kenaf (*Hibiscus cannabinus*) has impressive mechanical properties, making it a compelling choice for applications requiring high strength. Meanwhile, miswak (*Salvadora persica*) is well known for its antimicrobial properties, which inhibit the growth of bacteria. Applications such as structural material for building, 3D printing, biomedical, and prosthetic part are examples of the reliability of this natural fiber composite (Alaa et al. 2023; Ayu et al. 2020). This indicates the possibility of using natural fiber composite to compete as an alternative to food packaging materials.

Industries are presently utilizing food packaging composed of plastics and materials containing polymers sourced from petrochemicals. The polymers are predominantly non-biodegradable and require a longer time to decompose. The approach of replacing these polymers with natural fiber reinforced PLA, such as miswak, can serve as a more environmentally sustainable option. Nonetheless, miswak fiber-reinforced PLA has a disadvantage due to its comparatively weaker mechanical qualities compared to other synthetic polymers. Furthermore, PLA has a disadvantage as it has poor thermal stability (Rafiqah et al. 2023).

This study further reinforces miswak fiber composite with kenaf fiber to form a new hybrid composite for food packaging. Kenaf fiber is chosen since it is known for its superior mechanical properties and abundantly available resources in Malaysia. This hybrid composite is expected to not only having a reliable mechanical and thermal properties but also being able to decompose. Therefore, it can be a suitable material for food packaging and able to substitute a portion of synthetic polymer usage. Although kenaf fiber has been extensively studied in polymer composites, the incorporation of miswak (*Salvadora persica*) fiber remains scarcely reported. The unique antimicrobial and lignin-rich properties of miswak distinguish it from conventional plant fibers, offering dual functionality on mechanical reinforcement and potential antimicrobial protection for food packaging applications. This research represents one of the first systematic evaluations of miswak–kenaf hybrid reinforcement in a PLA matrix.

Materials and methods

Materials

Miswak fiber were purchased from Al-Imtinaan Sdn Bhd and kenaf fiber were purchased from Polycomposite Sdn Bhd. The pellets of polylactic acid and NaOH utilized in this research came from Evergreen Sdn. Bhd. of Selangor. According to supplier data, kenaf fiber (Polycomposite Sdn. Bhd.) contains approximately 72–75 wt% cellulose, 14–16 wt% hemicellulose, and 8–9 wt% lignin. Miswak fiber (Al-Imtinaan Sdn. Bhd.) comprises 45–50 wt% cellulose, 20–25 wt% hemicellulose, 18–20 wt% lignin, and minor traces of silica and calcium salts. The PLA (Evergreen Sdn. Bhd.) used has a melt flow index of 6 g/10 min (190°C, 2.16 kg) and a density of 1.24 g/cm³.

Fiber preparation

The fibers were crushed and sieved into 250 µm for miswak and 300 µm for kenaf.

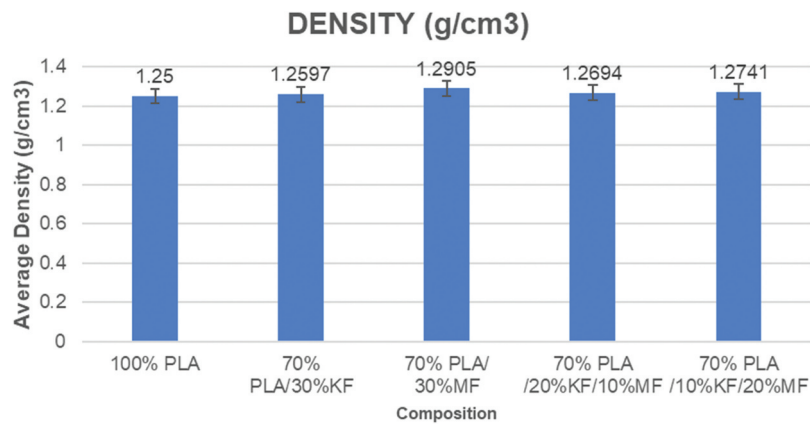


Figure 1. Density of hybrid composites with different compositions.

Fiber alkaline treatment

Figure 1 shows the fiber treatment process with alkaline treatment. The concentrations of NaOH solutions at 1% were determined by weight per volume percentage (w/v %). 20 grams of sodium hydroxide pellets were measured using a digital balance. 2 liters of distilled water were poured into a beaker and then heated using a heating plate. The sodium hydroxide was poured into the beaker to form a solution. The solution was stirred until all the pellets dissolved. Next, fiber was weighed accordingly before mixing with the solution. The 1 wt% sodium hydroxide solution was poured into the container, ensuring all the fiber was immersed for 1 h. After that, the treated fibers were thoroughly rinsed with tap water to remove any remaining NaOH and maintain a pH of 7 (± 0.5). The fibers were air-dried for 48 h and oven dried at 60°C for 24 h.

Preparation of composite

The composite was prepared using a Brabender internal mixer. Table 1 shows the PLA, kenaf, and miswak composite composition loaded in the Brabender. The mixture was blended at 160°C and 50 rpm for 15 min. After obtaining the composite blend, it was pressed into a specimen shape according to the ASTM standard. The composite compound were subjected to hot pressing by the Vechno Vation compression molding technique for 8 min at 160°C, having been pre-heated for 4 min before applying full pressure for 4 min. The sheet was subsequently cold pressed between two plates for 4 min at 15°C. The composite sheets were sectioned into designated standard dimensions for subsequent testing. The total fiber loading of 30 wt% was selected based on previous studies reporting optimal mechanical performance for natural fiber/PLA composites at fiber contents between 25 and 35 wt%. Higher fiber loading beyond 35% typically leads to agglomeration and poor fiber dispersion, reducing composite strength. Preliminary mixing trials confirmed that 30 wt % total reinforcement provided good fiber dispersion and homogeneous mixing without excessive viscosity during compounding. All fiber ratios in this study are expressed as weight percentages (wt%).

Table 1. Ratio of hybrid composite sample.

Sample	Composition (PLA : miswak : kenaf)		
	PLA (%)	Miswak fiber (%)	Kenaf fiber (%)
1	70	30	0
2	70	0	30
3	70	20	10
4	70	10	20

Characterization of treated kenaf fiber

Physical properties (density)

Density was determined according to ASTM D 1895-17 (2017) standard. The density of the sample was calculated by using the following equation:

$$\text{Density}(\text{g}/\text{cm}^3) = m/v(\text{Where } m \text{ represents mass and } v \text{ for volume}) \quad (1)$$

Physical properties (moisture content)

Seven samples were prepared for moisture content evaluation. The samples were placed in normal climatic conditions at room temperature ($27 \pm 2^\circ\text{C}$) with 65% relative air humidity for 24 h before being weighed. The percentage of moisture content was determined by using Equation 2. The samples were heated in the oven for 24 h at 105°C . Before heating, the samples were measured as M_0 . After 24 h in the oven, the material was weighed again as M_1 . Therefore:

$$\text{Moisture Content (\%)} = (M_1 - M_0/M_0) \times 100 \quad (2)$$

Physical properties (water absorption)

Water absorption analysis was performed by immersion of the samples for 24 h in distilled water by ASTM D570. Samples with dimensions of $76.2 \text{ mm} \times 25.4 \text{ mm} \times 3 \text{ mm}$ were oven-dried for 24 h and weighed (M_0) before being immersed in distilled water. After 24 h, Each sample was wiped to eliminate surface moisture, promptly weighed (M_t), and then immersed in water again. The M_t of the samples were recorded every day for 5 days. Five samples for each composition were used to obtain the average % water absorption (WA).

$$\text{WA} = M_t - M_0/M_0 \times 100\% \quad (3)$$

Mechanical characterization

The experiment adhered to ASTM D638 standards, conducting tensile testing on the composites with a 5 kN Bluehill INSTRON Universal Testing Machine to determine their tensile strength and tensile modulus. Before testing, the materials were cut into Type I dumbbell-shaped specimens. The testing was conducted under controlled settings with a relative humidity of $23 \pm 2^\circ\text{C}$ and $50\% \pm 5\%$. The strain rate was established at 2 mm per minute, and the gauge length measured 30 mm. Statistical significance was determined by replicating five samples.

Three bending flexural tests were conducted on the composites following the ASTM D790 standard. The experiments were performed utilizing a 5 kN Instron Universal Testing equipment. The testing samples measured 127 mm in length, 12.7 mm in width, and 3 mm in thickness. The trials were performed at a temperature of $23 \pm 2^\circ\text{C}$, a relative humidity of 50%, and a strain rate of 2 mm min^{-1} . Five samples were assessed to determine average and statistical significance.

Fourier transform infrared spectroscopy

The composite board of size $10 \text{ mm} \times 2 \text{ mm} \times 3.2 \text{ mm}$ was prepared for Fourier transform infrared spectroscopy (FTIR) analysis. The FTIR test was conducted by using Spectrum 1000 series spectrometer. The spectra were recorded in the range of $500\text{--}4000 \text{ cm}^{-1}$ with the resolution of 4 cm^{-1} and 16 scan.

Morphological characterization

A Hitachi S-3400N scanning electron microscope (SEM) was used to analyze fractured specimens from the tensile tests. The samples were examined under a 15 kV accelerated voltage to separate the fibers from the matrix. The samples were gold-sputtered before observation to avoid the charging effect.

Chemical analysis

Several TAPPI standard procedures were used to carry out the chemical composition analysis of the treated kenaf fiber. These procedures included the analysis of ethyl benzene extractive (TAPPI T 204 CM97), the measurement of lignin content (TAPPI 222 CM-98), and the analysis of holocellulose and alpha-cellulose content (TAPPI T 203 CM-99).

Results and discussion

Physical properties of specimen density

Figure 1 shows the density of hybrid composites with different fiber compositions. From the graph, it was found that the density increment increases with an increase in fiber content. This finding is aligned with a previous study where it states that as the weight fraction of natural fibers increases, there is an increase in the bulk density of the composites (Akter et al. 2021). The 30% MF sample was found to have the highest density value. However, the increment in density is not significant as the increase is low, between 2 and 3%, compared with pure PLA, which has a density of 1.25 g/cm³.

Physical properties of specimen moisture content

Moisture content of hybrid composites is represented in Figure 2. The highest moisture content is observed in the 70% PLA/30% KF composition at 1.466%, indicating that kenaf fibers absorb more moisture due to their more open, porous structure and higher hydrophilicity (Lee et al. 2021; Rafiqah et al. 2023). In contrast, the 70% PLA/30% MF composition shows the lowest moisture content at 0.354%, suggesting that miswak fiber has a more compact structure or higher lignin content, which reduces water absorption. The hybrid composites mixed KF and MF display intermediate moisture content values. For hybrid composite of 70% PLA/20% KF/10% MF shows 1.076%, closer to the kenaf dominant blend. Meanwhile, for 70% PLA/10% KF/20% MF shows 0.676%, reflecting the increased proportion of miswak, which helps reduce moisture uptake.

Physical properties of the specimen water absorption

The water absorption behavior of pure PLA and PLA-based hybrid composites reinforced with kenaf fiber (KF) and miswak fiber (MF) was evaluated over a period of 120 h, as presented in Figure 3. The results indicate that all samples exhibited an increasing trend in water uptake with immersion time, which is consistent with the hygroscopic nature of natural fibers. Pure PLA displayed the lowest water absorption, starting at 3.2% after 24 h and gradually increasing to 6.5% after 120 h. The low water absorption is attributed to the hydrophobic nature of PLA, which limits the penetration of water molecules into the polymer matrix. The incorporation of natural fibers significantly influenced the water absorption of the composites. Among the single-fiber composites, 70% PLA/30% KF showed the highest water absorption, reaching 8.62% after 120 h. This can be explained by the hydrophilic nature of kenaf fibers, which readily absorb water due to the presence of hydroxyl groups in cellulose and hemicellulose. Conversely, the 70% PLA/30% MF composite exhibited lower water uptake (6.23% at 120 h), suggesting that miswak fibers may possess a comparatively lower hydrophilicity or higher compatibility with the PLA matrix. For the hybrid composites, the water absorption was dependent on the fiber ratio. The 70% PLA/20% KF/10% MF composite exhibited intermediate water uptake (8.38% at 120 h), while the 70% PLA/10% KF/20% MF

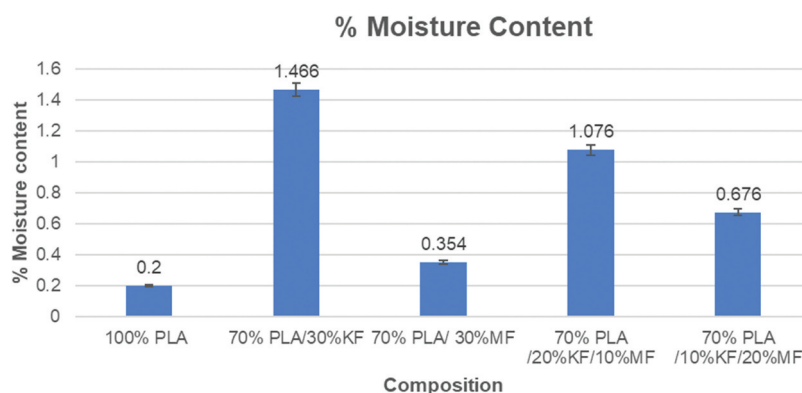


Figure 2. Moisture content of hybrid composites with different composition.

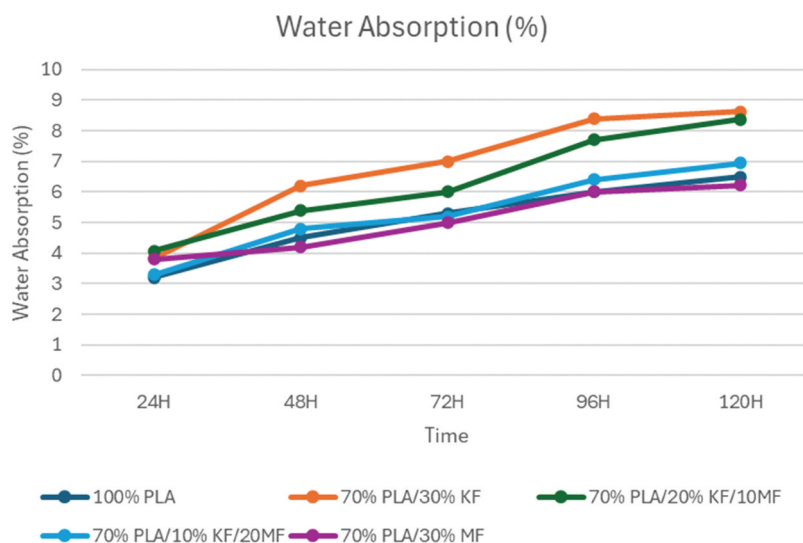


Figure 3. Water absorption of hybrid composites with different composition.

composite showed reduced water absorption (6.95% at 120 h), indicating that increasing the proportion of MF can mitigate the water absorption induced by KF. These findings highlight the influence of fiber type and composition on the water absorption behavior of PLA-based composites, which is critical for applications where moisture resistance is required. Composite with more miswak fiber loading was expected to record the lowest water absorption compared with the composite with more kenaf fiber as it is directly related to their structural differences and the specific chemical composition. This result was correlated with the moisture content findings. Hydroxyl group is mainly hydrophilic and largely exist in cellulose of a fiber. Since kenaf fiber is well known for its high cellulose, this proves the reason why composite with more kenaf has tendency to absorb more water (Hu et al. 2024). In addition, composite with more miswak fiber in its matrix have a better packing arrangement, resulting less void. There will be less spaces for water to penetrate into the matrix (Sosiati et al. 2022). Previous researchers have reported that the hydrophilicity of the fibers can be alleviated through an appropriate chemical treatment that enhances the bond strength between the fibers and the matrix. This improvement in bond strength results in a reduction in water absorption (Abd Halip et al. 2019; Jawaid, Sultan, and Saba 2018). The highest water absorption observed at 8.62% corresponds to the kenaf-rich composite (70% PLA/30% KF). The values are significantly lower than those typically reported for uncoated natural fiber composites used in structural or construction materials, which may range from 10–20% depending on fiber type and matrix hydrophilicity (Jawaid, Sultan, and Saba 2018). Although construction composites demand much lower moisture uptake to prevent dimensional instability, the water absorption values obtained here are within the acceptable range for bio-based packaging applications, particularly considering that the composites can be further improved through surface coating or compatibilizer addition. The relatively higher water absorption of kenaf fiber composites is attributed to the high cellulose content and porous microstructure of kenaf fiber, whereas increasing miswak fiber content reduces moisture uptake due to its denser structure and higher lignin content.

Mechanical properties (tensile testing)

Figure 4 shows the tensile strength and tensile modulus for miswak and kenaf-reinforced hybrid composite. The pure PLA sample exhibits a tensile strength of 45 MPa and the lowest tensile modulus of 3800 MPa. By observing the results, by increasing 30% of KF and 30% MF showed decrease in tensile strength by 21.7% and 12.2%, respectively. However, for 70% PLA/10%KF/20%MF hybrid composite shows increase in tensile strength by 13.3%. Meanwhile, the tensile modulus shows an increasing value for all compositions. By increasing 30% of KF shows an increase in modulus by 55.5% and addition of 30% MF increased by 61.8%. The combination samples (20% KF/10% MF and 10% KF/20% MF) continue this trend. The extraction of lignin from kenaf fiber and miswak fiber markedly improves its mechanical properties, especially tensile

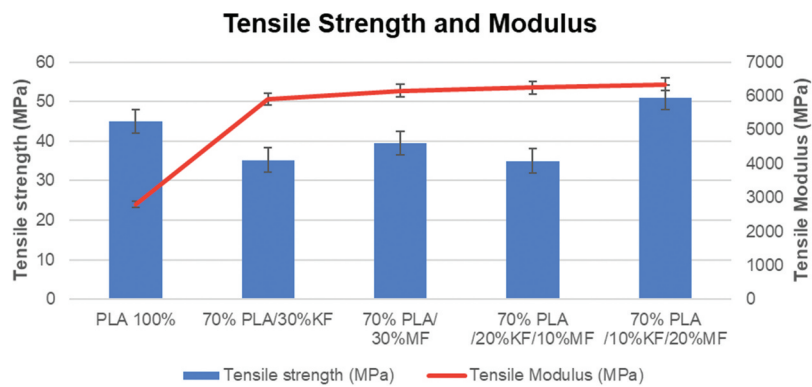


Figure 4. Tensile strength and modulus of hybrid composites with different composition.

strength and tensile modulus. The surface of the treated fiber becomes cleaner and more reactive, enhancing interfacial bonding in composite applications. The improved fiber–matrix adhesion further contributes to better mechanical performance, especially in composite reinforcement (Hamidon et al. 2019). The tensile modulus increases progressively from 6250 MPa to 6340 MPa, while tensile strength fluctuates. Notably, the 70% PLA/10% KF/20% MF sample shows the highest tensile modulus (6340 MPa) and highest tensile strength (51 MPa) among the composites, even surpassing pure PLA in tensile strength. This suggests a synergistic effect of combining the two fiber types in optimal ratios, which may enhance fiber dispersion and interfacial bonding, leading to better mechanical performance. The tensile modulus of a composite material increases with the incorporation of a greater percentage of fibers, as fibers typically possess significantly higher stiffness than the polymer matrix. As additional fibers are integrated into the matrix, they substitute the softer, flexible polymer with a more durable and rigid substance, therefore enhancing the composite’s overall resistance to deformation under tensile stress (Saba, Paridah, and Jawaid 2015)

Mechanical testing (flexural test)

Figure 5 shows the flexural strength and modulus of the composite with different fiber percentage. From the testing, the flexural strength of 100% PLA is 65 MPa. The addition of 30% kenaf fiber (KF) resulted in a notable increase in flexural strength to 75.11 MPa, reflecting an enhancement of roughly 15.6% compared to pure PLA. This improvement is due to the significant stiffness and reinforcing properties of kenaf fibers, which efficiently transmit stress throughout the composite matrix. Meanwhile, 20% KF + 10% MF yielded a flexural strength of 65 MPa, equal to pure PLA but lower than the 30% KF blend. This indicates that substituting 10% of KF with MF compromises the reinforcing efficiency. Next for sample with 10% KF +

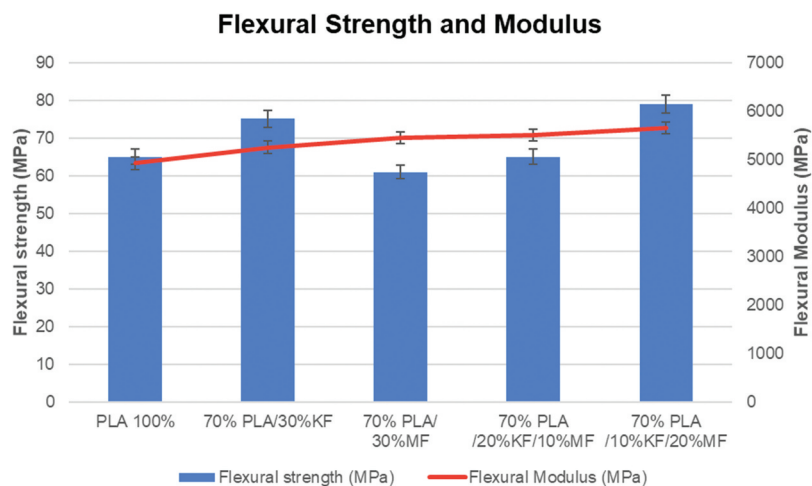


Figure 5. Flexural strength and modulus of hybrid composites with different composition.

20% MF significantly improved the flexural strength to 79 MPa, the highest among all samples. This suggests that a higher MF content in the presence of some KF may optimize the interfacial bonding and stress distribution, leading to synergistic reinforcement effects. The modulus of pure PLA was 4920 MPa. Incorporation of 30% KF increased the modulus to 5250 MPa, while 30% MF further increased it to 5450 MPa. The modulus continued to rise slightly with the hybrid composites, reaching 5500 MPa for the 20% KF/10% MF blend, and 5650 MPa for the 10% KF/20% MF blend. This ascending trend signifies that both KF and MF enhance the stiffness of PLA, with MF seemingly more successful in increasing the modulus. The maximum modulus was recorded in the 70% PLA/10% KF/20% MF blend, supporting the concept of a synergistic effect when MF and KF are combined (Yusuff, Sarifuddin, and Ali 2021).

Scanning electron microscope

Figure 6 is a SEM image at 50× magnification for a hybrid composite with different compositions. The image distinctly illustrates the incorporation of reinforcing fibers into the PLA matrix. The elongated structures of kenaf fibers and Miswak fiber (MF), appearing partially extracted or fractured, suggesting fiber–matrix interaction and load transfer processes under mechanical stress. The observable spaces and interstices among some fibers and the adjacent matrix indicate regions of inadequate interfacial adhesion, which may act as stress concentrators and may result in early failure. Nevertheless, certain fibers seem to be firmly embedded and oriented inside the matrix, signifying areas of strong interfacial bonding, which enhances mechanical performance. The coarse surface roughness and the occurrence of fiber pull-outs indicate energy absorption during fracture, a beneficial trait for enhancing composite toughness. The strong interfacial bonding supports the result in mechanical testing where the strength and modulus increase with the addition of fiber. In terms of fiber distribution, Figure 6(b) shows the better homogeneity compared to Figure 6(a). This is due to the miswak fiber have a smaller fiber length compared to kenaf fiber (Shafi et al. 2024). Since the amount of miswak fiber is more at composition, 10% KF 20% MF shows good distribution of MF along the matrix. This enables the miswak fiber to fill more voids in the matrix, thus improving the mechanical properties.

Figure 7 shows SEM image for hybrid composite at 100× magnification. Fracture analysis will be analyze based on the cross-section of the hybrid composite after failure. Figure 7(a) indicates that in composition 20% KF 10% MF. The image shows many voids present in the matrix after failure. These voids were formed because the lack of miswak fiber with short fiber length to fill the gap between those voids. As for comparison in Figure 7(b), there were sufficient miswak fiber in composition 10% KF 20% MF to fill the gap, thus producing matrix with less void. Shorter fibers are easier to integrate into the matrix, leading to better mechanical performance due to reduced fiber breakage and improved stress distribution. Moreover, studies have shown that shorter fibers can reduce the likelihood of fiber pull-out during loading, thus improving the load transfer efficiency within the composite (Bin Abdollah, Amiruddin, and Nordin 2021). Enhancing interfacial adhesion through fiber treatment or compatibilizers could significantly improve the structural integrity and performance of such composites.

Fourier transform infrared spectroscopy

FTIR spectroscopy revealed the structural properties of PLA with hybrid fiber composite and the effect of different fiber loading in composite. Figure 8 shows the FTIR spectra of kenaf and miswak fiber-reinforced PLA hybrid composite with different composition. The analysis of this hybrid composite will be focused on low-range wavelength between 500 cm^{-1} and 1800 cm^{-1} , where the characteristic of fiber can be observed. In the 30% KF sample, a broad band near 3300 cm^{-1} indicates O–H stretching vibrations typical of cellulose. As KF content decreases and MF content increases, this peak reduces in intensity, suggesting reduced cellulose content. At $1510\text{--}1600\text{ cm}^{-1}$ shows the reducing intensity of the peak associated with aromatic ring in lignin. Peaks in this region reduce significantly, indicating degradation or removal of lignin during alkaline treatment (Manral and Bajpai 2021). Meanwhile, the PLA's characteristic is located between 2800 cm^{-1} and 3000 cm^{-1} . The peak at 2994 cm^{-1} was identified as O–H stretching from carboxylic acid, and the peak at 2944 cm^{-1} was identified as C–H stretching from the alkane. The peak at 1746 cm^{-1}

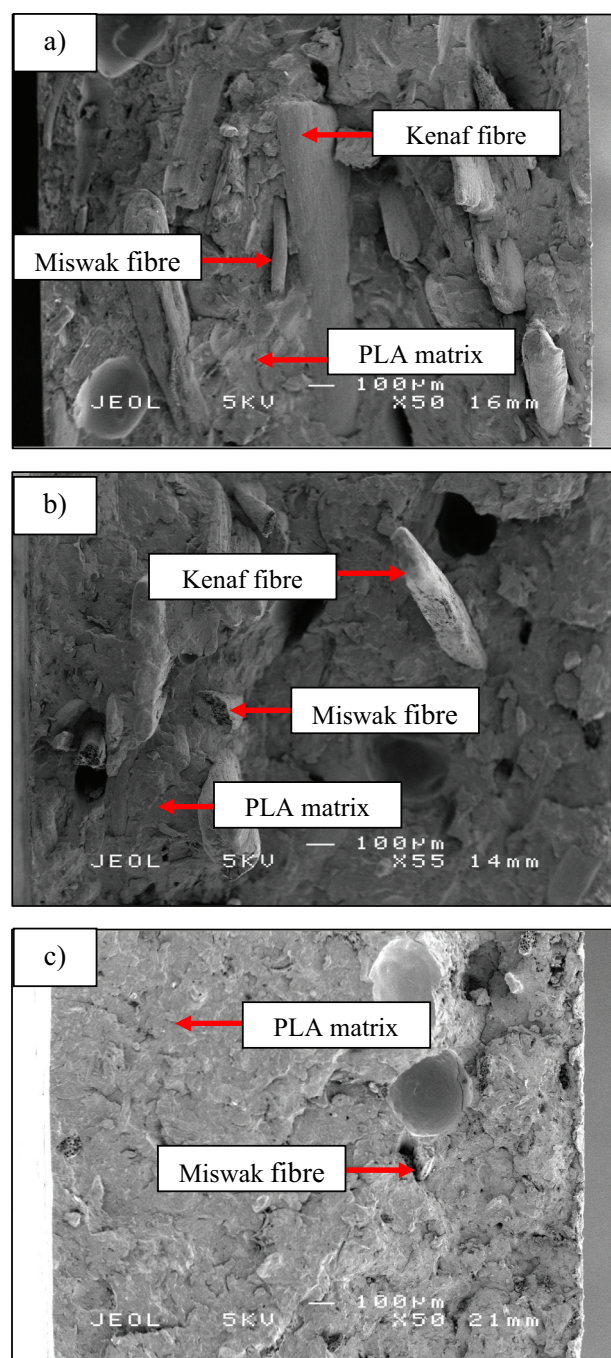


Figure 6. Scanning electron microscope image at 50 \times magnification of (a) 20% KF 10% MF, (b) 10% KF 20% MF, and (c) 30% MF.

identified as C=O stretching from lignin, peak at 1451 cm^{-1} identified as C-H bending from cellulose and peak at 1180 cm^{-1} identified as C-O stretching from aliphatic ether groups of hemicelluloses (Karimi et al. 2014). Peaks in the region 1000–1200 cm^{-1} are attributed to C-O-C stretching and C-O stretching vibrations, typically found in cellulose and hemicellulose structures. The peak near 2900 cm^{-1} corresponds to C-H stretching vibrations in aliphatic $-\text{CH}_2$ and $-\text{CH}_3$ groups, typically arising from the cellulose and hemicellulose backbone. Another notable absorption is observed around 1730 cm^{-1} , which is associated with C=O stretching vibrations of the ester or carboxylic groups in hemicellulose or waxy substances (Abdul Razak et al. 2014).

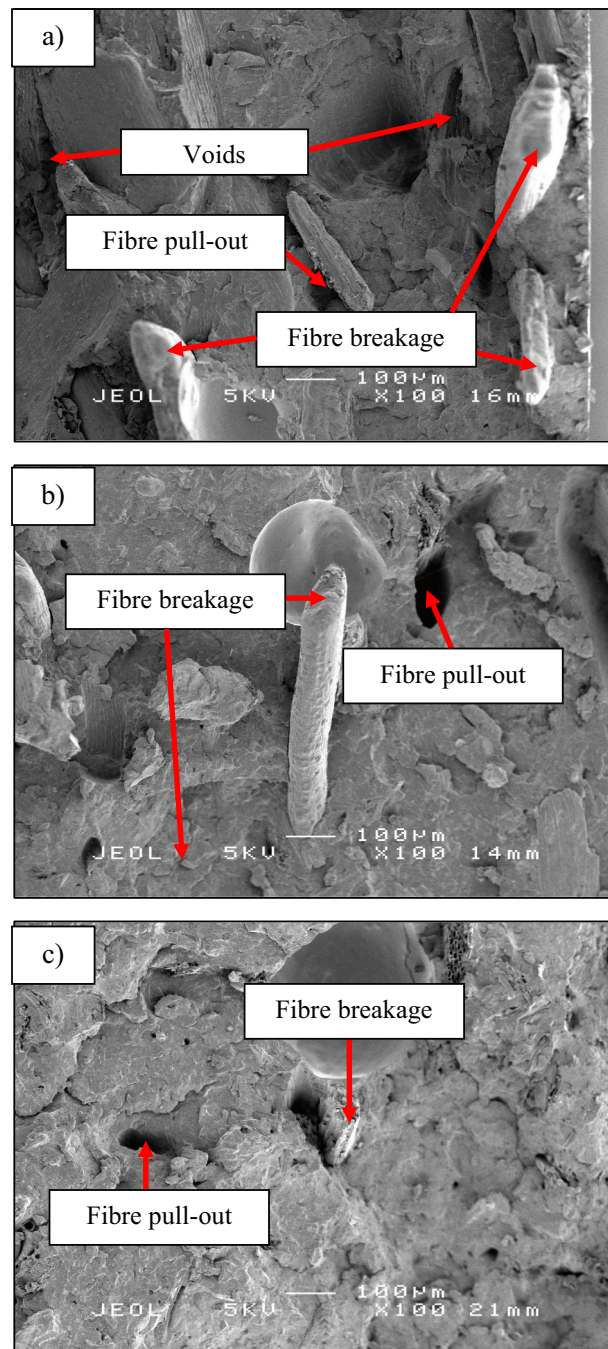


Figure 7. Scanning electron microscope image at 100× magnification of (a) 20% KF 10% MF, (b) 10% KF 20% MF, and (c) 30% MF.

Thermal gravimetric analysis

Figure 9 shows the thermogravimetric analysis (TGA) profiles, and Figure 10 is DTG profiles of miswak and kenaf fiber hybrid composite. The different stage of mass loss along the change in temperature, with the residual mass at 600°C. The first step, there is a small but noticeable step in the first stages of the profiles of the composites between 70–100°C that is due to the loss of volatiles water in the composite (Khiari et al. 2020). Next thermal degradation pattern, beginning with a stable region up to around 270–300°C, followed by a sharp weight loss indicating major decomposition. During this temperature multiple thermal degradation of complex chemical structures such C–H and C–O bonds broken, mainly from the cellulose of the reinforcing fibers (Zakaria et al. 2014). The 30% MF and 10% KF/20% MF composites exhibit slightly higher

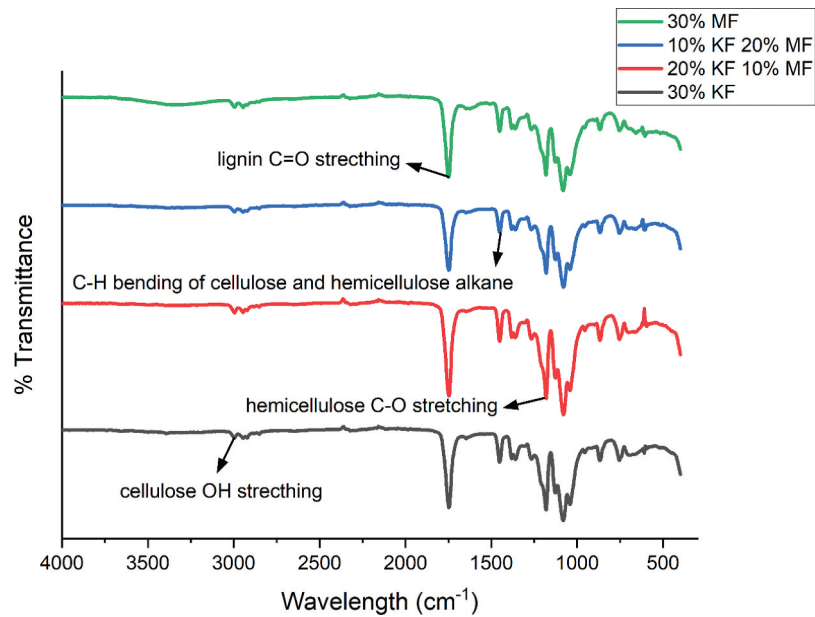


Figure 8. FTIR spectra of hybrid composites with different composition.

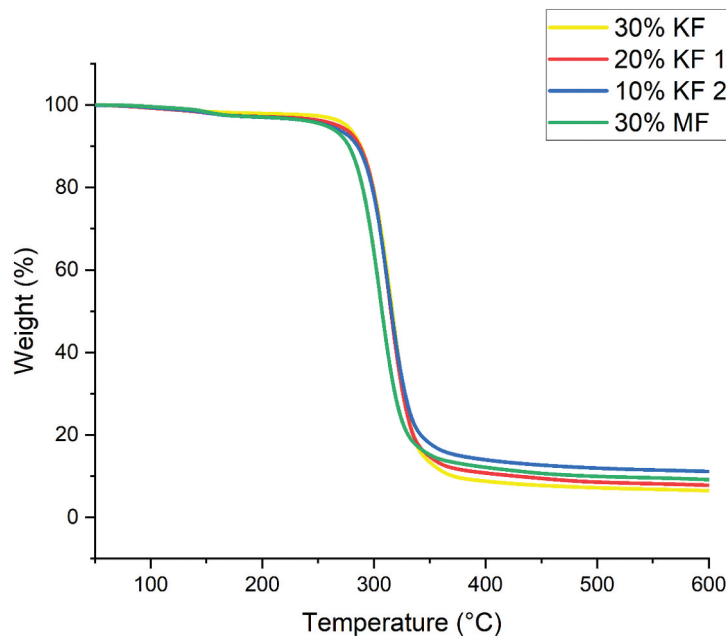


Figure 9. TGA analysis of hybrid composites with different composition.

thermal stability, as their weight loss onset appears at a slightly higher temperature compared to the others. The 30% KF composite degrades earlier, indicating lower thermal stability, likely due to the higher hemicellulose content in kenaf which decomposes at lower temperatures. Increasing the ratio of miswak fiber improves the thermal stability of the PLA composite. This may be related to MF's higher lignin concentration and more compact structure, which delay heat breakdown. Consequently, miswak fiber-reinforced PLA composites may be more appropriate for applications requiring enhanced thermal resistance (Nur Diyana et al. 2022). The DTG analysis reveals clear trends corresponding to filler matrix composition. Increasing MF content results in higher decomposition temperatures and reduced mass-loss rates, indicating improved thermal stability due to the thermoset nature of MF. Conversely, higher KF content lowers the DTG peak temperature because the lignocellulosic fibers degrade at lower temperatures.

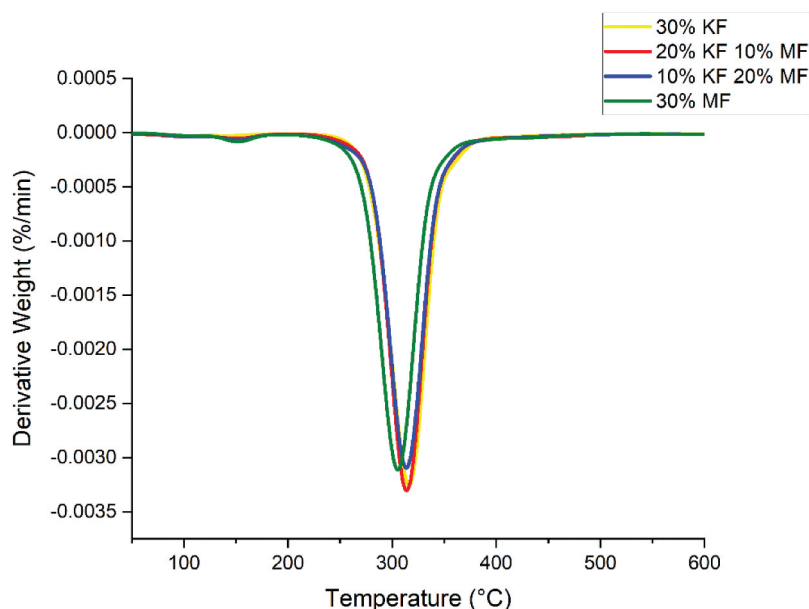


Figure 10. DTG analysis of hybrid composites with different composition.

Table 2. Chemical composition of fiber.

Fiber type	Treatment	Cellulose (wt%)	Hemicellulose (wt%)	Lignin (wt%)
Kenaf (KF)	Untreated	75	16	9
Kenaf (KF)	NaOH treated (1 wt%)	84	9	7
Miswak (MF)	Untreated	50	30	20
Miswak (MF)	NaOH treated (1 wt%)	68	18	14

Overall, the sample containing 30% MF exhibits the highest thermal stability, while 30% KF shows the lowest, confirming the dominant influence of matrix composition on thermal degradation characteristics.

Chemical analysis

Table 2 presents the chemical composition of untreated and NaOH-treated kenaf (KF) and miswak fibers (MF), highlighting clear differences in their lignocellulosic structures and the effect of alkali modification on fiber performance. Both fibers are composed of cellulose, hemicellulose, and lignin. However, kenaf exhibits a markedly higher cellulose content than miswak in both untreated (75 wt% and 50 wt%) and treated states (84 wt% and 68 wt%), indicating superior intrinsic stiffness, crystallinity, and load-bearing capability. In contrast, miswak contains substantially higher amounts of hemicellulose and lignin, which explains its greater hydrophilicity, lower thermal stability, and earlier thermal degradation onset. Alkali treatment significantly increases the relative cellulose content in both fibers by removing amorphous constituents such as hemicellulose, lignin, and surface impurities, while simultaneously reducing hemicellulose content for (KF: 16 to 9 wt%; MF: 30 to 18 wt%) and lignin (KF: 9 to 7 wt%; MF: 20 to 14 wt%). This compositional shift enhances fiber rigidity, thermal stability, moisture resistance, and fiber–matrix interfacial adhesion due to increased surface roughness and exposure of reactive hydroxyl groups. From a composite performance perspective, kenaf fiber is expected to provide superior mechanical reinforcement and thermal resistance owing to its higher cellulose and lower hemicellulose contents, whereas miswak fiber, although initially more hydrophilic and less crystalline, shows substantial improvement after NaOH treatment. Overall, the results demonstrate that alkali treatment effectively upgrades fiber quality, with kenaf showing inherently higher reinforcement potential and miswak benefiting significantly from chemical modification, thereby influencing the fibers' thermal behavior, moisture sensitivity, and interfacial interactions within composite systems.

Conclusion

A hybrid composite reinforced with natural fibers has been successfully developed using miswak (MF) and kenaf (KF) fibers in a PLA matrix. Among all formulations, the composite containing 10% kenaf fiber and 20% miswak fiber demonstrated the most favorable overall performance. This composition achieved the highest tensile strength (51 MPa), flexural strength (79 MPa), and modulus values, while also exhibiting improved thermal stability with an onset degradation temperature of approximately 305°C. These findings indicate that combining miswak and kenaf fibers creates compatibility, enhancing interfacial bonding and mechanical stiffness beyond what can be achieved by either fiber alone. The alkaline treatment applied to kenaf fiber effectively removed amorphous non-cellulosic components, particularly lignin and hemicellulose, as confirmed by the reduction in FTIR peaks at 1730 cm^{-1} (C=O stretching) and $1510\text{--}1600\text{ cm}^{-1}$ (aromatic skeletal vibrations). This chemical modification enhanced the exposure of cellulose microfibrils and increased the surface roughness of the fiber, thereby improving interfacial adhesion between the fiber and PLA matrix. As a result, tensile strength and tensile modulus of the treated fiber-reinforced composites significantly improved.

In terms of moisture behavior, kenaf fiber composites displayed higher water absorption (up to 8.62%), whereas miswak fiber composites showed reduced water uptake (approximately 6.23%) due to their higher lignin content and denser fiber structure. For food packaging applications, this moderate water absorption level is considered acceptable, especially when the composite surface is coated or laminated with a thin biodegradable barrier layer to prevent direct liquid contact. Both miswak and kenaf are naturally derived, nontoxic plant materials, and miswak's well-documented antimicrobial properties may further reduce microbial contamination risks at the food – packaging interface.

Overall, the 70% PLA/10% KF/20% MF hybrid composite presents the most balanced combination of mechanical strength, thermal stability, and moisture resistance, confirming its potential as a sustainable alternative to petroleum-based polymers in biodegradable food packaging.

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Author contributions

All the authors have agreed to be fully accountable for the submitted manuscript's content and have approved submission.

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Data availability statement

The data that support the findings of this study are available to publish and share in any relevant platform

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