Materials Research
Express

PAPER • OPEN ACCESS

Enhancing the performance of hybrid bio-composites reinforced with natural fibers by using coupling agents

To cite this article: Alamry Ali *et al* 2025 *Mater. Res. Express* **12** 035504

Materials Research Express

CrossMark

OPEN ACCESS

RECEIVED
5 September 2024

REVISED
17 October 2024

ACCEPTED FOR PUBLICATION
14 November 2024

PUBLISHED
19 March 2025

PAPER

Enhancing the performance of hybrid bio-composites reinforced with natural fibers by using coupling agents

Alamry Ali[1], **Md Saiful Islam**[2], **Sinin Hamdan**[3] **and Masuk Abdullah**[4]

[1] Department of Mechanical Engineering, College of Engineering in Al-Kharj, Prince Sattam Bin Abdulaziz University, Al-Kharj-11942, Saudi Arabia
[2] Department of Chemistry, Bangladesh Army University of Engineering & Technology, Natore-6431, Bangladesh
[3] Department of Mechanical Engineering, Faculty of Engineering. Universiti Malaysia Sarawak, 94300 Kota Samarahan Sarawak, Malaysia
[4] Department of Vehicles Engineering, Institute of Vehicle Engineering and Mechatronics, Faculty of Engineering, University of Debrecen, Ótemető strt. 2-4, 4028 Debrecen, Hungary

E-mail: mdsaifulislam16@bauet.ac.bd

Keywords: natural fiber, hybrid bio-composites, coupling agents, mechanical properties, TGA, SEM, FTIR

Abstract

Natural fiber-reinforced hybrid bio-composites are emerging as sustainable alternatives to traditional composites due to their environmental benefits and desirable properties. However, the interfacial interaction between natural fiber and polymer matrix is very weak. Thus, this study looks into the effect of maleic anhydride (MAH) coupling agents on the performance of natural fiber-reinforced hybrid biocomposites. The bio-composites were prepared using jute fiber, kenaf fiber, and polylactic acid (PLA) through the hot compression method. We treated both natural fibers with MAH coupling agents before using them in the production of biocomposites. Comprehensive characterization techniques, including tensile strength, modulus, and impact strength, were employed to evaluate the mechanical properties of the composites. The mechanical results indicated a significant improvement in mechanical properties for the bio-composites treated with coupling agents. The tensile strength of bio-composites increased by 35%, tensile modulus by 15%, and impact strength by 20% after modification with MAH coupling agents. The surface morphology and chemical interactions between the fiber and polymer matrix were investigated using SEM and FTIR studies. The FTIR result reveled that the intensity of C=O peaks enhanced after MAH treatment. Moreover, SEM images exposed better fiber dispersion and adhesion, corroborated by FTIR spectra showing enhanced chemical bonding where MAH reacted with the cellulose backbone of the fibers and formed fiber cellulose ester. Furthermore, TGA results revealed that adding MAH coupling agent to the fiber increased the thermal stability of biocomposites.

1. Introduction

Natural fibers like jute and kenaf have gained significant attention in the search for sustainable material alternatives [1]. Their inherent properties make them promising candidates for composite materials, particularly in applications where eco-friendliness and performance are essential [2–6]. Despite their potential, the practical use of these fibers in composite materials frequently faces challenges due to compatibility issues with matrices, which in turn limits the ability to achieve the desired mechanical and structural properties [7, 8]. Addressing these challenges necessitates exploring effective strategies to optimize the performance of jute and kenaf fiber in composite materials [9, 10]. Due to their numerous benefits, such as greater strength, lower density, renewability, biodegradability, reduced health hazards, and cost and weight reduction, they are ideal candidates for inclusion in composite materials [11, 12]. The carbon emissions from burning plastics will be decreased by using biodegradable sources in polymer products [13, 14].

PLA is an adherent of the aliphatic polyester family and is thought to be biodegradable and compostable, meaning that it can produce carbon dioxide and biomass when microorganisms operate on it in a humid environment [15]. PLA is a thermoplastic polymer characterized by its notable strength and modulus, which may be derived from annually renewable resources to produce goods for the biocompatible medical device as well as the industrial packaging sector [16, 17]. However, PLA exhibits lower mechanical strength and thermal stability compared to other polymers [18, 19]. These drawback properties can be enhanced by incorporating natural fiber as reinforcement [20].

Despite offering commendable strength and facile processing capabilities in the majority of equipment, this polymer is costly and requires additional modifications to be suitable for numerous practical applications [21]. Therefore, an effective strategy for enhancing and reducing the cost of the material is to incorporate natural fibers, which have acquired popularity as a substitute for synthetic fibers [22]. As a result, it is important to incorporate these inexpensive natural fibers into biodegradable polymer (PLA) so as to effectively decrease manufacturing costs by substituting a portion of the costly PLA with these economical fibers without compromising the matrix polymer's biodegradability [23, 24].

Within the composite sector, the utilization of matured kenaf as fiber reinforcements in different forms has been seen. Research has demonstrated that the incorporation of kenaf fibers has resulted in enhanced mechanical characteristics of pristine polymers [25, 26]. The primary components of kenaf consist of cellulose (45%–57% by weight), hemicelluloses (21.5% by weight), lignin (8%–13% by weight), and pectin (3%–5% by weight). Kenaf fibers exhibit superior thermal and mechanical characteristics in comparison to varying natural fibers when they are combined with appropriate polymers [27]. Kenaf fiber exhibits promise as a filler for reinforcement in polymer matrix composites [28, 29].

Jute and kenaf fibers are increasingly used in the production of natural fiber-reinforced hybrid PLA biocomposites due to their excellent mechanical properties and biodegradability [30]. Jute fibers are known for their high tensile strength and modulus, making them ideal for reinforcing polymer matrices. Similarly, kenaf fibers exhibit high specific strength and stiffness, contributing to the overall structural integrity of bio-composites. The hybridization of jute and kenaf fibers in PLA matrixes can enhance the mechanical performance, providing a balance of strength and flexibility [31]. This synergy between jute and kenaf fibers leads to improved impact resistance and toughness in the resulting biocomposites. Additionally, both fibers have low density, which helps in producing lightweight composites suitable for automotive and construction applications. The integration of jute and kenaf fibers into PLA bio-composites offers a promising route for developing high-performance, sustainable materials [32–34].

The mechanical characteristics of composites composed of natural fiber plastic can be enhanced through hybridization [35]. Composites made of natural fiber and biodegradable polymers can benefit from hybridization in terms of mechanical properties [36]. The natural fiber-reinforced hybrid bio-composites, which combine two or more natural fibers into one matrix, have special qualities that are hard to obtain in polymers reinforced with different kinds of single fibers [37, 38]. Compared to composites made solely of coir or oil palm fiber, hybrids of these fiber-reinforced composites have better tensile and flexural characteristics [39].

Despite many advantages, natural fibers exhibit high hydrophilicity, and inherent incompatibility with hydrophobic matrices often leads to interfacial adhesion issues, compromising the mechanical and thermal characteristics of hybrid fiber composites [40]. One promising avenue for enhancing the characteristics of these hybrid composites involves the use of coupling agents. Coupling agents typically contain functional groups that can interact with both fibers and matrix [41]. The fibers contain hydroxyl (–OH) groups, whereas the matrix typically consists of non-polar groups, such as hydrocarbons [42]. Coupling agents with one end that can hydrogen bond with the hydroxyl groups on the fibers and the other end that can cooperate with the non-polar groups in the matrix effectively bridge the gap between the two materials, leading to improved interfacial adhesion. Implementations of coupling agents have shown significant potential in enhancing the mechanical, thermal, and structural characteristics of composite materials. By promoting a stronger bonding between the fibers and polymer matrix, coupling agents have the capacity to bolster the overall performance of jute/kenaf fiber-reinforced hybrid composites. Maleic anhydride (MAH) and silane coupling agents exhibit promising potential as suitable candidates for enhancing the overall characteristics of composites. Maleic anhydride (MAH) coupling agents are widely used to enhance the performance of natural fiber-reinforced biocomposites [43]. MAH acts by grafting onto polymer matrices, creating strong covalent bonds with hydroxyl groups present on the surface of natural fibers [44]. This chemical bonding significantly improves the interfacial adhesion between the fibers and the polymer matrix, leading to better stress transfer. As a result, bio-composites modified with MAH exhibit increased mechanical properties, such as tensile, flexural, and impact strengths. The improved interfacial bonding also enhances the thermal stability of the composites [45].

Additionally, MAH treatment helps in reducing water absorption by the fibers, which is crucial for maintaining the mechanical integrity of bio-composites in humid environments. This reduction in water absorption is due to the hydrophobic nature of the grafted polymer chains, which shield the fibers from moisture. The compatibility between natural fibers and polymer matrices is also enhanced by MAH, enabling the development of composites with more uniform and consistent properties. Consequently, the use of MAH

Table 1. Formulations of natural fibers reinforced hybrid bio-composites.

Types composites	PLA (wt%)	Jute (wt%)	Kenaf (wt%)
PLA	100	—	—
Jute/PLA	50	50	—
Kenaf/PLA	50	—	50
Jute/Kenaf/PLA	50	25	25
Jute/Kenaf/MAH/PLA	50	25	25

Note: Jute/Kenaf/MAH = Maleic anhydride treated Jute and Kenaf fibers.

coupling agents in natural fiber-reinforced bio-composites not only improves their performance but also broadens their applicability in various industrial sectors. This makes MAH play a crucial role in driving the advancement of sustainable, high-performance biocomposites [46].

Natural fibers with polymers are becoming more and more in demand in the market nowadays, which is drawing significant interest from numerous industries. The creation of hybrid natural fiber composites has been the issue of extensive investigation. However, no research has been done on how coupling agents change the properties of natural fiber-reinforced hybrid bio-composites made from PLA, kenaf fiber, and jute fiber [4, 14, 32]. Therefore, this study investigates the critical role of coupling agents in enhancing the characteristics of jute/kenaf fiber reinforcement hybrid bio-composites. The goal of this research is to present a novel, previously unexplored combination of natural fibers composites. It explores the use of MAH coupling agents that improve fiber-matrix interfacial bonding and overall structural integrity efficiently, leading to enhanced mechanical and thermal properties. Unlike previous research, this work targets a unique aspect of comprehensive evaluation, covering mechanical, thermal, water absorption, and morphological properties, providing a more holistic understanding of hybrid biocomposite performance. This research also proposes a pathway to creating green composite with high-performance materials and is significant for its potential to scale for industrial applications, contributing to more sustainable manufacturing practices.

2. Materials and methods

2.1. Materials

The chemicals utilized in this research comprised analytical reagents (AR Grade, 99.9% purity). The pelletized variant of the PLA used in this study, namely NatureWorkTM PLA 3001D, was acquired from NatureWork® LLC, Minnesota, United States. The material demonstrates a particular density of 1.24 g cm^{-3} and a melt flow index (MFI) of g/10 min (190 °C/2.16 kg) when subjected to a temperature of 190 °C and a load of 2.16 kg. The coupling agent utilized in this investigation was maleic anhydride (MAH), and the catalyst was sodium hydroxide (NaOH), obtained from Sigma Aldrich. Two natural fibers, namely jute (Corchorus olitorius) and kenaf (Hibiscus cannabinus L), serve as reinforcement for biodegradable PLA matrixes. Kenaf and jute fibers are collected from Malysia and Bangladesh.

2.2. Fibers treatment with coupling agents

Both fibers were cut to a length of ~2 mm using a Ring Knife Flaker. These chopped fibers were then manually sieved through a sieve (0.5 mm) to obtain a homogeneous size of fiber. The fibers were dried in an oven at 60 °C temperature for 2 h. After that, these dry fibers were soaked and left in a 5% maleic anhydride (MAH) solution (water solvent) for an hour with a catalyst of 0.5% NaOH. The ratio of maleic anhydride to fibers was 1:20 (wt/wt). After 2 h of drying in a hot air oven set at 60 °C, the treated fibers are ready to be used.

2.3. Bio-composites preparation

The fibers and polylactic acid (PLA) particles were thoroughly and uniformly combined using Brabender Mixer Machine. Different weight fractions were designed for the jute and kenaf fibers (treated and untreated) and PLA. The sample formulation is shown in table 1. The mold manufacturing and the desired composite thickness. A hot-pressing technique was utilized to create the composite materials. The mold was heated for five minutes at a temperature of 160 °C on both the upper and lower heaters, and the pressure was 1000 psi. To avoid any shrinkage during the composite extraction process, the mold was allowed to cool for approximately five minutes. The hot press and Brabender machines are shown in (figure 1).

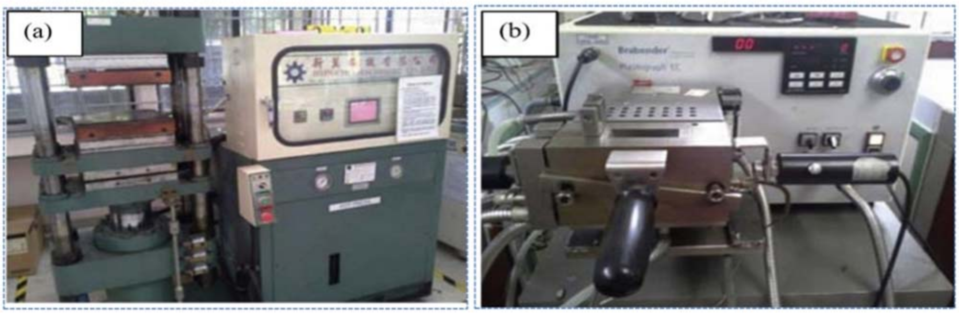

Figure 1. Bio-composites fabrication machine of (a) Hot-press machine and (b) Brabender machine.

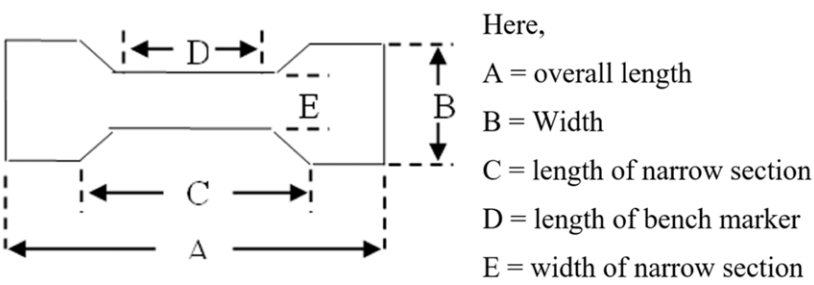

Figure 2. Dumbbell-shape specimen for tensile test.

2.4. Characterization

The morphology, mechanical and thermal properties of green composites were characterized by the following methods. The interaction between fiber and chemical (MAH) were evaluated using Fourier Transform Infrared (FTIR) spectroscopy. FTIR test was conducted within the range of 4000 cm^{-1} to 400 cm^{-1}. SEM analysis examination was conducted on the morphological and structural characteristics of fractured surface composites utilizing a scanning electron microscope (JSM-5510, JEOL, and Japan). For the tensile test was conducted according to ASTM 638 and 790 standards. A dumbbell-shaped sample was prepared using a cutter machine (Machinery Industry Co. Ltd). The thickness and width of the specimen were measured using a digital micrometer. The sample sheets were cut in form of dumbbell specimen for tensile measurement as shown in figure 2.

Each sample was cut into seven pieces for the tensile strength test according to the ASTM. To investigate the mechanical characteristics of the samples, an Instron Tensile Machine type 4032 was employed. The procedure described in ASTM D638 was followed. In order to ascertain the tensile strength measurements of the composites, namely the maximum load, load at break, maximum strain, maximum displacement, maximum percentage of strain, and young modulus, the average value of the five most favourable readings was obtained and computed.

2.4.1. Impact test

The dimension of the specimen is 80 × 15 mm in thickness. The impact energy of the generated data is quantified in Joules (J) by dividing the impact energy in joules by the area under the notch, so establishing the measure of impact strength. The materials are tougher when the resulting number is higher.

2.4.2. Thermogravimetric analysis (TGA)

The thermal stability of developed green composites was examined using a Perkin Elmer Thermogravimetric Analyzer (TGA 7) device. Under a nitrogen environment, sample weights weighing approximately 8 mg were heated with a rate of 10°C min^{-1} from 30 °C to 700 °C. According to ASTM D 3850-2000, the weight loss of the composites was calculated.

2.4.3. Water absorption test

A rectangular size specimen with dimensions of 39 mm × 10 mm × 4.1 mm were considered from each of the composite's samples for the determination of water absorption properties. The samples were properly dried for

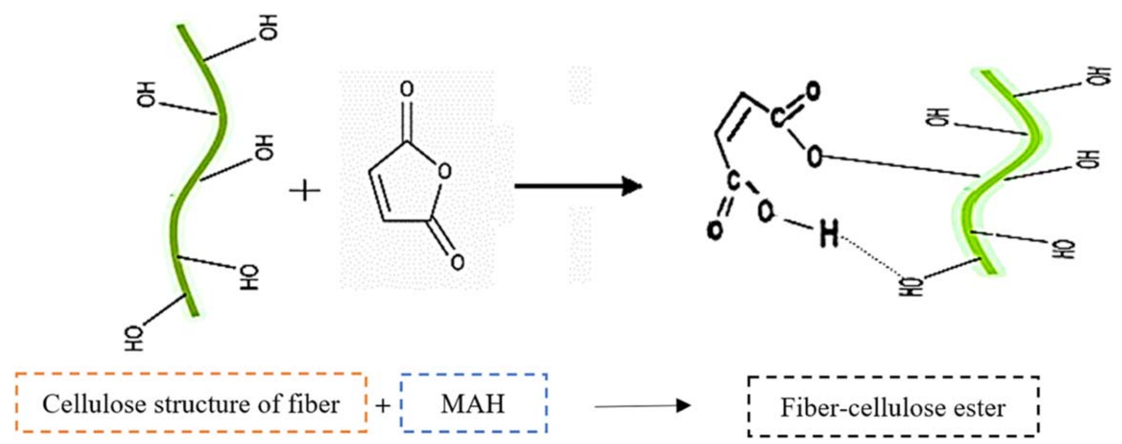

Figure 3. Interfacial bond between the fibers and the polymer matrix.

an hour at 80 °C in an oven, then allowed to cool outside before being promptly weighed. Following the guidelines outlined in ASTM D 570-99, the specimens, which had been weighted and dried, were immersed in distilled water for a duration of two hours. Periodically, the specimens were taken from the water, dried off with tissue paper to eliminate any remaining water, weighed again, and then immersed into the water immediately. The provided formula was utilized to calculate the absorption of water.

$$\text{Water absorption}(\%) = \frac{W_2 - \quad W_1}{W_1} \times 100\% \tag{1}$$

Where, W_1 and W_2 are the specimen weight that taken before and after soaking in water.

The average of five tests of same specimens were taken as the result.

3. Results and discussions

3.1. FTIR result

The FTIR method is employed to investigate the chemical reaction between cellulose fiber and MAH (Maleic anhydride) coupling agent. The FTIR spectra of MAH treated and untreated fibers are depicted in (figure 4). The presence of -OH groups is indicated by the broad peaks observed in the range of 3600–3070 cm^{-1} across all spectra. Additionally, the C–H stretching is responsible for the peak at 2946 cm^{-1} [47, 48]. The absorption peaks at 1740 cm^{-1} and 1622 cm^{-1} indicate the presence of the C=O stretch band and C=C, respectively. It is worth noting that the intensity of C=O peaks enhanced after MAH treatment which proves the addition of carbonyl group in the fiber surface. This band also indicates that the MAH has successfully grafted onto the cellulose chains of the fibers. Moreover, the –OH group peak intensity was also reduced after MAH treatment (figure 4). Other new absorption bands observed in the FTIR spectra of MAH-treated fibers include a peak at around 1490 cm^{-1}, which is related with the C=C stretching vibration of the ester linkage of maleic anhydride, and a peak at approximately 1270 cm^{-1} and 1000 cm^{-1}, which relates to the C–O stretching vibration of the ester linkage of maleic anhydride [47]. The other minor peaks found at around 1454 $cm^{-1}$and 1421 cm^{-1} are ascribed to the C–H bending from lignin. The presence of these new absorption bands confirms that the MAH has successfully reacted with the cellulose backbone of the fibers. This led to the formation of fiber cellulose ester, which significantly improved the interfacial bond between the fibers and the polymer matrix in the composite (figure 3). All of these, as stated early, validate the subsequent reaction (figure 4).

3.2. Scanning electron microscopy (SEM)

In order to study the surface morphology of fabricated composites, SEM test was employed in this study. The morphology of each phase can be important in evaluating the overall characteristics of the material. (Figure 5) displays SEM distribution of various splintered surfaces of bio-composites (MAH treated and untreated). (Figures 5(a) and (b)) shows the SEM images of jute/kenaf composites and jute/kenaf/PLA hybrid composites without coupling agent treatment. From these images, it can be found that the fibers are not well dispersed in the matrix, and there are large gaps between the fibers and the matrix for the single fiber and hybrid fiber composites. This observation suggests that there is a lack of strong bond among fibers and polymer matrix, perhaps resulting in the formation of weak composites. However, it is evident from the analysis of (figure 5(c)) that a smooth surface is generated following the MAH treatment. This observation indicates a favorable bond

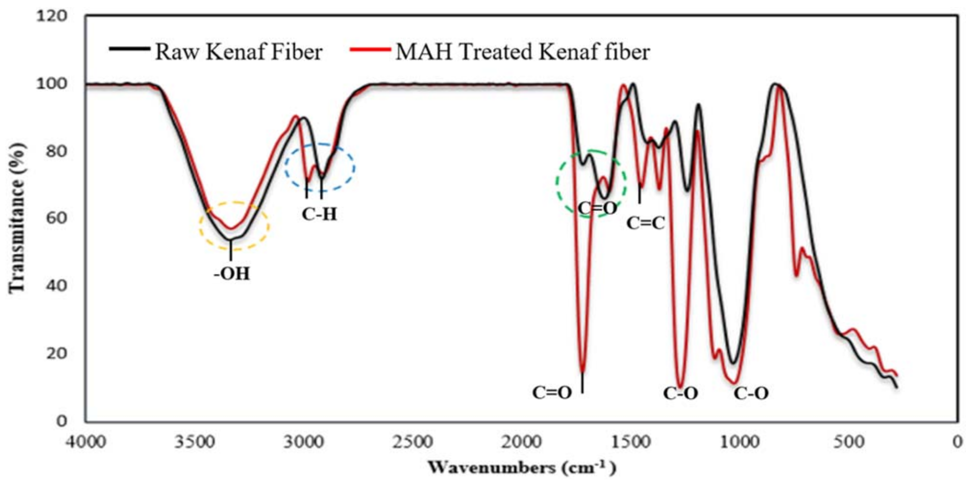

Figure 4. FTIR spectra of MAH treated and untreated jute fiber.

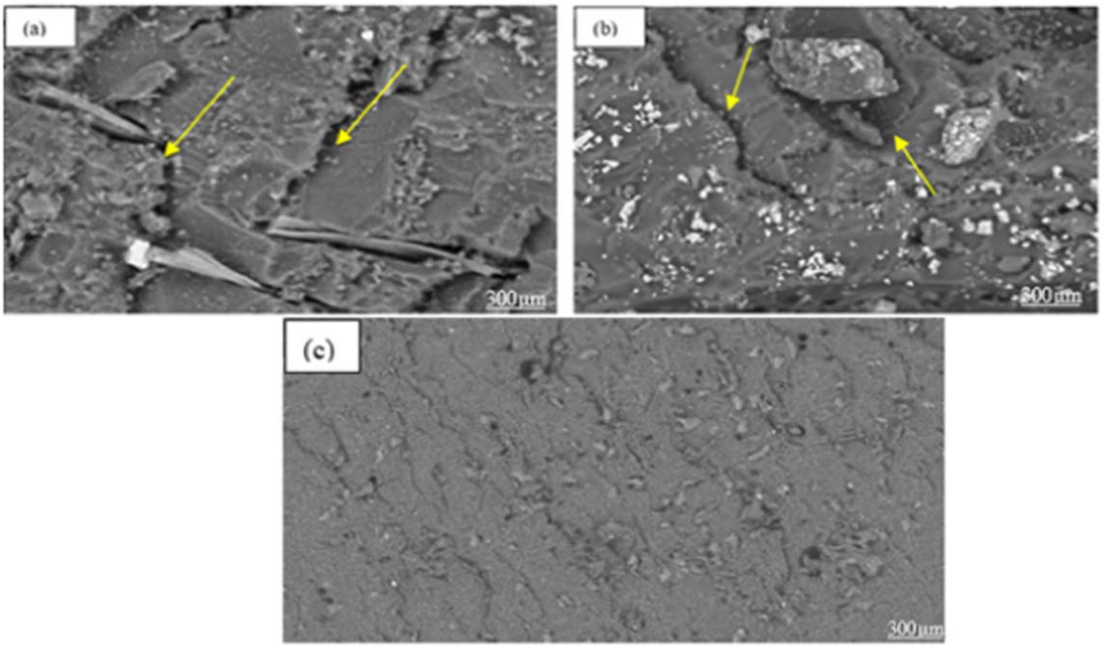

Figure 5. SEM images of bio-composites of (a) jute/PLA, (b) jute/kenaf/PLA and (c) jute/kenaf/MAH/PLA. Arrow indicates micro-cracking and gaps between fiber and PLA of composites.

between jute and kenaf fibers and PLA matrix. The enhancement of mechanical properties in composites is strongly influencing the effective bond among fiber and polymer matrix. Finally, it is worth mentioning that the good wettability features given by the addition of two fibers can limit the void formation at the fiber-polymer interface and result in high stiffness and strength composites, which in turn lead to high impact strength values [2, 47]. All of this indicates that the fibers and PLA matrix were more attracted to each other owing to the coupling agent, which may result in stronger composites.

3.3. Thermogravimetric analysis (TGA)

The thermal stability of composites consisting of jute/PLA, jute/kenaf/PLA, and jute/kenaf/MAH/PLA was evaluated and presented in (figure 6). A three-step deterioration process was visible in the TGA thermograms of the jute/PLA and jute/kenaf hybrid composites without coupling agent treatment. This first step starts at about 100 °C and leads to the loss of water. When hemicellulose breaks down at about 250 °C, the second step takes place. The third step occurs at around 350 °C and is due to the degradation of cellulose and lignin [49]. The TGA thermogram for the jute/kenaf hybrid composites with coupling agent shows a similar three-step degradation process. However, the temperatures at which the degradation steps occur are slightly higher for the composites

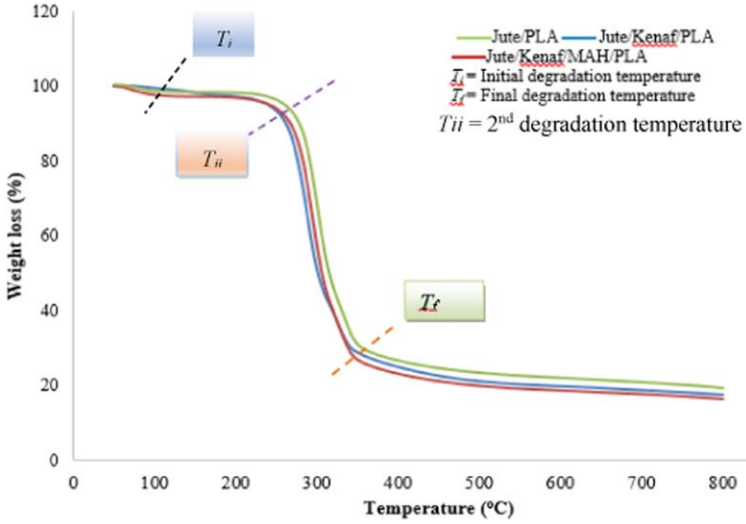

Figure 6. TGA thermograms of bio-composites of (a) Jute/PLA, (b) Jute/Kenaf/PLA and (c) Jute/Kenaf/MAH/PLA.

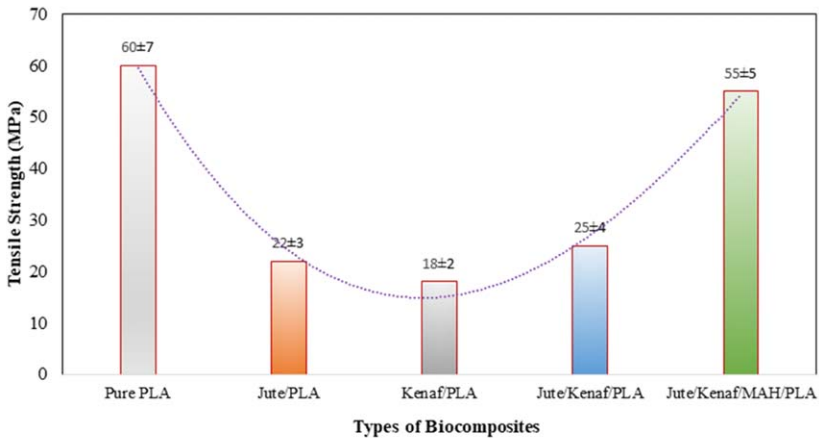

Figure 7. Tensile strength of various composites with and without coupling agent treatment.

with coupling agent. This means that coupling agent has made composites more stable at high temperatures. Additionally, the TGA thermograms reveal that at 800 °C, the composites with coupling agent have a sophisticated residual weight than the composites without coupling agent. This suggests that the coupling agent has decreased the char formation of the composites [49].

3.4. Mechanical properties

Bio-composites made from MAH-treated and raw jute and kenaf fibers are shown in (figures 7 and 8) along with their tensile strength and tensile modulus. Natural fiber-reinforced hybrid composites can have higher tensile strength and modulus when MAH coupling agents are used to modify the surfaces of the fibers [50]. MAH coupling agent enhances the adhesion and interaction between two distinct materials [51]. In the context of jute/kenaf hybrid composites, the utilization of MAH coupling agent serves to enhance the interfacial bonding and adhesion between fibers and PLA-matrix. (figure 7) clearly depicts the influence of MAH coupling agents on the tensile strength of jute/kenaf hybrid bio-composites. The modification of natural fibers by MAH coupling agents led to a notable increase in their tensile characteristics compared to unmodified ones. The addition of a coupling agent improves adhesion between fibers and the PLA-matrix, resulting in composites with increased strength. The tensile strength of bio-composite increased by 35% after modification with MAH.

The effect of MAH coupling agent on the tensile modulus of jute/kenaf hybrid bio-composites is seen in (figure 8). The tensile modulus of the composites treated with a coupling agent exhibits a notably greater value compared to tensile modulus of the composites without treatment. The observed trend in (figure 8) shows that the composites' tensile modulus increases (15%) with the coupling agent modification. Furthermore, we can

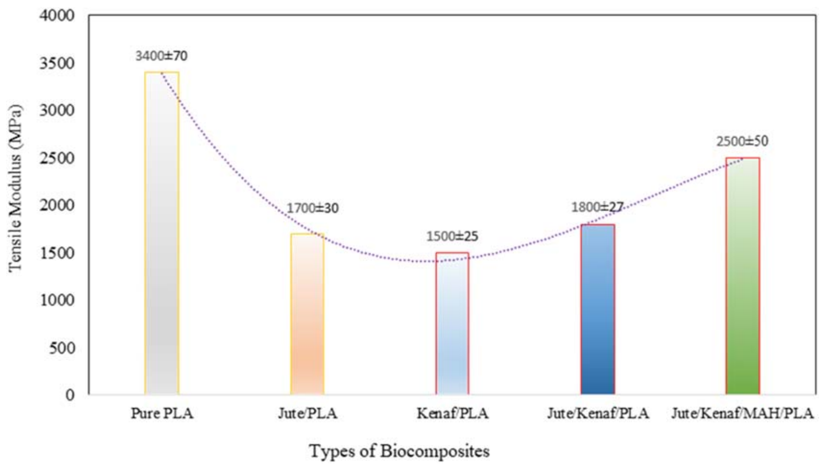

Figure 8. Tensile modulus of various composites treated with and without coupling agents.

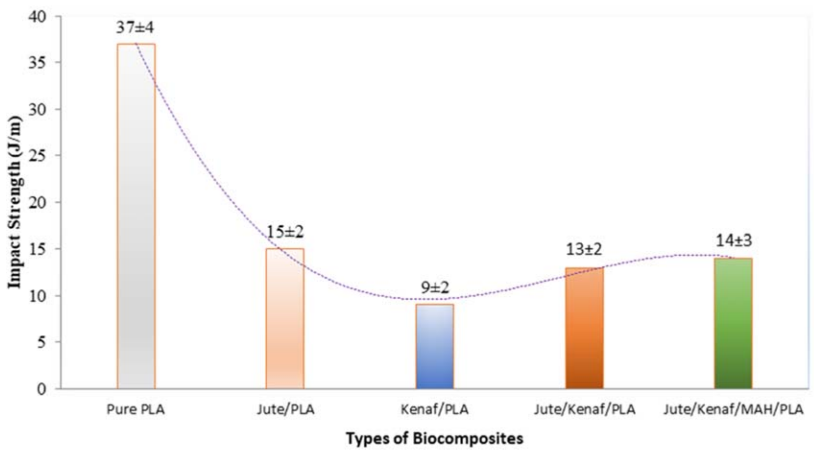

Figure 9. Impact strength of various composites treated with and without coupling agents.

attribute the enhancement in stiffness of the composites to the effect of the coupling agent treatment. MAH coupling agents can modify fiber surfaces to improve jute/kenaf hybrid composites' tensile strength and tensile modulus. The aforementioned attribute makes composites a more suitable choice for applications that require heightened degrees of strength and stiffness [47, 52].

We evaluated the impact strength of jute/kenaf hybrid bio-composites in relation to the surface modification of fibers using MAH coupling agents. (Figure 9) illustrates the impact strength of the composites treated and untreated treated with coupling agents. The chemical modification of fibers surface with MAH coupling agent results in a significant enhancement of their impact strength in comparison to the composites without modification. Treatment of fibers with MAH coupling agents increased the impact strength of bio-composites by 20%. The enhancement of impact strength in jute/kenaf-reinforced hybrid composites could be achieved through the surface modification of fibers using a coupling agent [52–54]. MAH is an effective coupling agent that enhances the adhesion between two substances. Within utilization of a coupling agent serves to augment the adhesive interaction between the jute fibers and kenaf fibers, as well as between these fibers and the matrix, within the context of jute/kenaf-reinforced hybrid composites. The enhanced adhesion between the matrix and the fibers, a direct result of the coupling agent, facilitated the significant improvement in impact strength of the composites. By enhancing adhesion between fibers and matrix, the coupling agent enhances the impact resistance of jute/kenaf-reinforced hybrid bio-composites. This increases the fibers' stress-transfer capability to the matrix, thereby preventing composite failure. Furthermore, the coupling agent has the potential to enhance the matrix's resilience. This increases the matrix's resistance to fracture, thereby contributing to the overall improvement of the composite's impact strength.

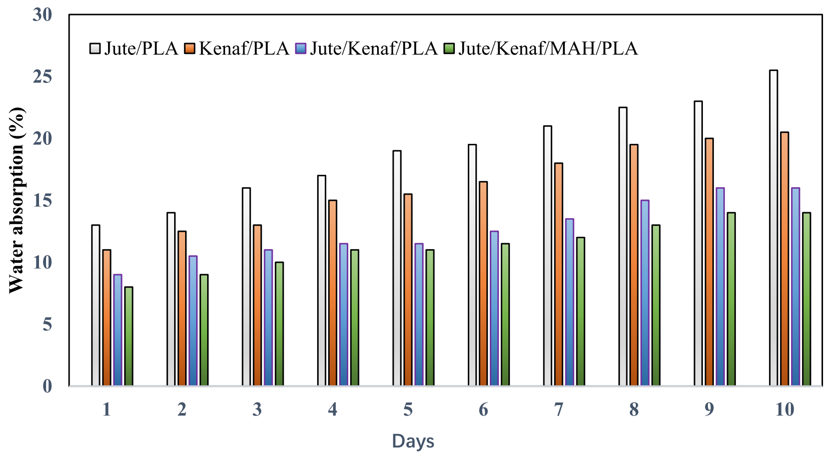

Figure 10. Water absorption of various composites with and without coupling agent treatment.

3.5. Water absorption analysis

The results of water absorption for a variety of bio-composites, both treated and untreated with MAH, are depicted in (figure 10). According to the data in the (figure 10), water absorption increased during the first few days before progressively decreasing. However, the modification of maleic anhydride (MAH) as a coupling agent greatly reduces water absorption in hybrid composites.

In order to adequately seal the interface and stop water intrusion, the MAH molecules make covalent interaction with the active sites on the matrix as well as the hydroxyl groups on the fibers. Furthermore, coupling agents have the ability to improve the interfacial tensions and uniform fiber dispersion within the matrix by enhancing the compatibility among the fibers and matrix. This adds even more to the decreased absorption of water [6, 55]. With this it can be assumed that coupling agent modification offers a promising approach to mitigating water absorption in jute/kenaf fiber reinforced hybrid bio-composites. By improving interfacial adhesion, imparting hydrophobicity, reducing fiber swelling, and enhancing matrix compatibility, coupling agents can significantly improve the water resistance and long-term performance of eco-friendly composites.

4. Conclusions

A Promising advance in the field of sustainable composite materials have been demonstrated by the effective preparation of natural fiber hybrid biocomposites by coupling agent modification. The reaction between fibers and coupling agents was confirmed through FTIR results. In addition, there were significant changes observed in the SEM micrograph of coupling agent-treated composites in contrast to composites made without treatment. The smoother surface observed upon the MAH used in the hybrid fiber composites. The use of MAH coupling agent to jute/kenaf/PLA hybrid bio-composites resulted in an increase in mechanical characteristics, including by means of tensile strength, modulus, and impact strength, when compared to both single composites and hybrid composites. The tensile strength increased by 35%, while the tensile modulus and impact strengths improved by 15% and 20%, respectively. For the coupling agent treatment, the thermal stability of composites was further improved. When compared to a single composite, water absorption of the composites treated with MAH was dramatically decreased. Because of the reduction of hydrophilic behavior of natural fiber. FTIR and SEM confirmed that the strategic use of coupling agents significantly enhanced the interfacial bond between the fibers and polymer matrix. The observed enhancements in mechanical and thermal stability signify the efficacy of MAH coupling agent modification in enhancing the composites performance. The successful integration of jute and kenaf fibers not only demonstrates the feasibility of employing agricultural waste in composite manufacturing but also underscores the potential for creating durable, eco-friendly composite materials. The results advocate for further exploration and optimization of this hybridization technique to meet the multifaceted demands of various industries. The findings also highlight the potential of MAH-treated natural fibers to enhance the mechanical and thermal properties of PLA-based composites, opening avenues for broader applications in industries like automotive, construction, and packaging. Despite many property improvements, the coupling agent may degrade the fiber structure for higher concentrations. Therefore, future research directions might investigate deeper into optimizing the coupling agent's composition, refining manufacturing processes, and exploring wider applications across industries. Finally, this study demonstrates that MAH coupling agent modification is an effective method to enhance the performance of natural fiber-reinforced hybrid bio-composites, potentially expanding their application in various industries seeking sustainable material solutions.

Acknowledgments

This research was supported by the 'University of Debrecen Program for Scientific Publication'.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Conflicts of interest

The authors declare that they have no conflicts of interest to report regarding the present study.

ORCID iDs

Md Saiful Islam https://orcid.org/0000-0003-1579-9452
Masuk Abdullah https://orcid.org/0000-0002-9330-1026

References

[1] Soni A, Das P K, Gupta S K, Saha A, Rajendran S, Kamyab H and Yusuf M 2024 An overview of recent trends and future prospects of sustainable natural fiber-reinforced polymeric composites for tribological applications *Ind. Crops Prod.* **222** 119501
[2] Ramesh M, Rajeshkumar L N, Srinivasan N, Kumar D V and Balaji D 2022 Influence of filler material on properties of fiber-reinforced polymer composites: a review *e-Polymers* **22** 898–916
[3] Birat K, Panthapulakkal S, Kronka A, Agnelli J A M, Tjong J and Sain M 2015 Hybrid biocomposites with enhanced thermal and mechanical properties for structural applications *J. Appl. Polym. Sci.* **132** 42452
[4] Abraha Guangul H, Prabhakar M N, Lee D W and Song J 2024 Development of hybrid green composites by dual natural fibers (Jute/Bamboo) reinforcement and investigation of their mechanical behavior *Polym. Compos.* **45** 8225–35
[5] Haris N I N, Hassan M Z, Ilyas R A, Suhot M A, Sapuan S M, Dolah R, Mohammad R and Asyraf M R M 2022 Dynamic mechanical properties of natural fiber reinforced hybrid polymer composites: a review *J. Mater. Res. Technol.* **19** 167–82
[6] Karthi N, Kumaresan K, Sathish S, Gokulkumar S, Prabhu L and Vigneshkumar N 2020 An overview: natural fiber reinforced hybrid composites, chemical treatments and application areas *Mater. Today Proc.* **27** 2828–34
[7] Shui Y, Deng N, Wang Y, Wang G, Chi H, Zeng Q, Peng Z, Cheng B and Kang W 2024 Recent progress on MOFs and their derivative-carbon fiber composite materials for oxygen electrocatalysis *J. Mater. Chem.* A **12** 20655–90
[8] Andrew J J and Dhakal H N 2022 Sustainable biobased composites for advanced applications: recent trends and future opportunities—a critical review *Compos. Part C Open Access* **7** 100220
[9] Majeed K, Jawaid M, Hassan A, Abu Bakar A, Abdul Khalil H P S, Salema A A and Inuwa I 2013 Potential materials for food packaging from nanoclay/natural fibres filled hybrid composites *Mater. Des.* **46** 391–410
[10] Feng Y, Hao H, Lu H, Chow C L and Lau D 2024 Exploring the development and applications of sustainable natural fiber composites: a review from a nanoscale perspective *Compos. Part B Eng.* **276** 111369
[11] Faheed N K 2024 Advantages of natural fiber composites for biomedical applications: a review of recent advances *Emergent Mater.* **7** 63–75
[12] Zhu S *et al* 2024 Recent advances on bast fiber composites: engineering innovations, applications and perspectives *Compos. Part B Eng.* **284** 111738
[13] Samir A, Ashour F H, Hakim A A A and Bassyouni M 2022 Recent advances in biodegradable polymers for sustainable applications *npj Mater. Degrad.* **6** 68
[14] Krishnasamy P, G R, Belaadi A and R S 2024 Dynamic mechanical characteristics of natural fiber hybrid composites, bio composites and nano composites –a review *Eng. Res. Express* **6** 012503
[15] Wu Y, Gao X, Wu J, Zhou T, Nguyen T T and Wang Y 2023 Biodegradable polylactic acid and its composites: characteristics, processing, and sustainable applications in sports *Polymers (Basel).* **15** 3096
[16] Raj A, Yousfi M, Prashantha K and Samuel C 2024 Morphologies, compatibilization and properties of immiscible PLA-based blends with engineering polymers: an overview of recent works *Polymers (Basel).* **16** 1776
[17] Islam M S, Hasbullah N A B, Hasan M, Talib Z A, Jawaid M and Haafiz M K M 2015 Physical, mechanical and biodegradable properties of Kenaf/Coir hybrid fiber reinforced polymer nanocomposites *Mater. Today Commun.* **4** 69–76
[18] Shanmugam V, Babu K, Kannan G, Mensah R A, Samantaray S K and Das O 2024 The thermal properties of FDM printed polymeric materials: a review *Polym. Degrad. Stab.* **228** 110902
[19] Khouri N G, Bahú J O, Blanco-Llamero C, Severino P, Concha V O C and Souto E B 2024 Polylactic acid (PLA): properties, synthesis, and biomedical applications—a review of the literature *J. Mol. Struct.* **1309** 138243
[20] Saiful Islam M, Hamdan S, Jusoh I, Rezaur Rahman M and Ahmed A S 2012 The effect of alkali pretreatment on mechanical and morphological properties of tropical wood polymer composites *Mater. Des.* **33** 419–24
[21] National Research Council and Division on Engineering and Physical Sciences and Commission on Physical Sciences and Mathematics and Applications and Committee on Polymer Science 1994 *Polymer Science and Engineering: The Shifting Research Frontiers* (National Academies Press)

[22] Voutetaki M E and Mpalaskas A C 2024 Natural fiber-reinforced mycelium composite for innovative and sustainable construction materials *Fibers* **12** 57
[23] Kamarudin S H, Abdullah L C, Aung M M and Ratnam C T 2018 Jusoh @ Talib, E.R. A study of mechanical and morphological properties of PLA based biocomposites prepared with EJO vegetable oil based plasticiser and kenaf fibres *Mater. Res. Express* **5** 085314
[24] Deshpande A R, Kulkarni A P, Wasatkar N, Gajalkar V and Abdullah M 2024 Prediction of wear rate of glass-filled PTFE composites based on machine learning approaches *Polymers (Basel).* **16** 2666
[25] Eslami-Farsani R, Reza Khalili S M, Hedayatnasab Z and Soleimani N 2014 Influence of thermal conditions on the tensile properties of basalt fiber reinforced polypropylene–clay nanocomposites *Mater. Des.* **53** 540–9
[26] Bezazi A, Boumediri H, Garcia del Pino G, Bezzazi B, Scarpa F, Reis P N B and Dufresne A 2022 Alkali treatment effect on physicochemical and tensile properties of date palm rachis fibers *J. Nat. Fibers* **19** 3770–87
[27] Yusuff I, Sarifuddin N and Ali A M 2021 A review on kenaf fiber hybrid composites: mechanical properties, potentials, and challenges in engineering applications *Prog. Rubber, Plast. Recycl. Technol.* **37** 66–83
[28] Hayeemasae N, Viet C X, Masa A, Shuib R K, Ismail H and Surya I 2023 The use of kenaf fibre as a natural anti-degradant in recycled high-density polyethylene and natural rubber-based thermoplastic elastomers *Polymers (Basel).* **15** 1237
[29] Boumediri H, Bezazi A, Del Pino G G, Haddad A, Scarpa F and Dufresne A 2019 Extraction and characterization of vascular bundle and fiber strand from date palm rachis as potential bio-reinforcement in composite *Carbohydr. Polym.* **222** 114997
[30] Tabrej K, Sultan M T H, Shah A U M, Safri S N A and Jawaid M 2022 The mechanical properties of Kenaf/Jute hybrid composites in different angles of orientation *Repair of Advanced Composites for Aerospace Applications* (CRC Press) 37–45
[31] Ju Z, Brosse N, Hoppe S, Wang Z, Ziegler-Devin I, Zhang H and Shu B 2024 Thermal and mechanical Properties of Polyethylene Glycol (PEG)-Modified Lignin/Polylactic Acid (PLA) biocomposites *Int. J. Biol. Macromol.* **262** 129997
[32] Anand P, Rajesh D, Senthil Kumar M and Saran Raj I 2018 Investigations on the performances of treated Jute/Kenaf hybrid natural fiber reinforced epoxy composite *J. Polym. Res.* **25** 94
[33] Morreale M, Scaffaro R, Maio A and La Mantia F P 2008 Effect of adding wood flour to the physical properties of a biodegradable polymer *Compos. Part A Appl. Sci. Manuf.* **39** 503–13
[34] Kharate N, Anerao P, Kulkarni A and Abdullah M 2024 Explainable AI techniques for comprehensive analysis of the relationship between process parameters and material properties in FDM-Based 3D-Printed biocomposites *J. Manuf. Mater. Process.* **8** 171
[35] Cavalcanti D K K, Banea M D, Neto J S S, Lima R A A, da Silva L F M and Carbas R J C 2019 Mechanical characterization of intralaminar natural fibre-reinforced hybrid composites *Compos. Part B Eng.* **175** 107149
[36] de Oliveira Filho G C, de Sousa Mota R C, da Conceicao A C R, Leao M A and de Araujo Filho O O 2019 Effects of hybridization on the mechanical properties of composites reinforced by piassava fibers tissue *Compos. Part B Eng.* **162** 73–9
[37] Estrada R G, Santiuste C and Barbero E 2021 Failure maps of biocomposites mechanical joints reinforced with natural fibres *Compos. Part C Open Access* **5** 100159
[38] Karthik K, Rajamanikkam R kumar, Venkatesan E P, Bishwakarma S, Krishnaiah R, Saleel C A, Soudagar M E M, Kalam M A, Ali M M and Bashir M N 2024 State of the art: natural fibre-reinforced composites in advanced development and their Physical/Chemical/Mechanical properties *Chinese J. Anal. Chem.* **52** 100415
[39] Prasad N, Agarwal V K and Sinha S 2018 Hybridization effect of coir fiber on Physico-Mechanical properties of Polyethylene-Banana/Coir fiber hybrid composites *Sci. Eng. Compos. Mater.* **25** 133–41
[40] Ismail S O, Akpan E and Dhakal H N 2022 Review on natural plant fibres and their hybrid composites for structural applications: recent trends and future perspectives *Compos. Part C Open Access* **9** 100322
[41] Fu Y and Yao X 2022 A review on manufacturing defects and their detection of fiber reinforced resin matrix composites *Compos. Part C Open Access* **8** 100276
[42] Romanowska B, Różańska W and Zimniewska M 2024 The influence of the chemical composition of flax and hemp fibers on the value of surface free energy *Materials (Basel).* **17** 1104
[43] Qian S, Mao H, Zarei E and Sheng K 2015 Preparation and Characterization of maleic anhydride compatibilized Poly(Lactic Acid)/Bamboo particles biocomposites *J. Polym. Environ.* **23** 341–7
[44] Whba R, Su'ait M S, Whba F, Sahinbay S, Altin S and Ahmad A 2024 Intrinsic challenges and strategic approaches for enhancing the potential of natural rubber and its derivatives: a review *Int. J. Biol. Macromol.* **276** 133796
[45] Jiang X, Wang C, Li G, Yu Y and Yang X 2024 Enhanced interfacial property and thermal conductivity of pitch-based carbon fiber/epoxy composites via three-layer assembly of PDI/GN/PDI interphase *Compos. Part B Eng.* **273** 111238
[46] Ghernaout D, Belaadi A, Boumaaza M, Chai B X, Jawaid M, Abdullah M M S, Krishnasamy P and Al-Khawlani A 2024 Effects of incorporating cellulose fibers from yucca treculeana L. on the thermal characteristics of green composites based on high-density polyethylene: an eco-friendly material for cleaner production *J. Mater. Res. Technol.* **31** 787–98
[47] Gorgun E, Ali A and Islam M S 2024 Biocomposites of Poly(Lactic Acid) and microcrystalline cellulose: influence of the coupling agent on thermomechanical and absorption characteristics *ACS Omega* **9** 11523–33
[48] Azeez S and Shenbagaraman R 2025 Fourier transform infrared spectroscopy in characterization of bionanocomposites *Characterization Techniques in Bionanocomposites* (Elsevier) 209–27
[49] Neto J S S, de Queiroz H F M, Aguiar R A A and Banea M D 2021 A review on the thermal characterisation of natural and hybrid fiber composites *Polymers (Basel).* **13** 4425
[50] Jawaid M, Abdul Khalil H P S and Alattas O S 2012 Woven hybrid biocomposites: dynamic mechanical and thermal properties *Compos. Part A Appl. Sci. Manuf.* **43** 288–93
[51] Zhu H, Dong B, Cai X, Xi L, Zhang P, Hao Y, Chen S and Zeng R 2024 *In situ* covalent crosslinking strategy to construct highly stable composite separators for lithium-ion batteries *Chem. Eng. J.* **488** 151120
[52] Jawaid M, Chee S S, Asim M, Saba N and Kalia S 2022 Sustainable Kenaf/Bamboo Fibers/Clay hybrid nanocomposites: properties, environmental aspects and applications *J. Clean. Prod.* **330** 129938
[53] Krishnasamy S, Thiagamani S M K, Muthu Kumar C, Nagarajan R, R.M. S, Siengchin S, Ismail S O and M.P I D 2019 Recent advances in thermal properties of hybrid cellulosic fiber reinforced polymer composites *Int. J. Biol. Macromol.* **141** 1–13
[54] Huang M, Liu H, Xiang Q and Gao H 2024 Improving dispersion of basalt fiber with polyethylene glycol and effect of coupling agent *Compos. Interfaces* **31** 1309–30
[55] Ayrilmis N, Jarusombuti S, Fueangvivat V, Bauchongkol P and White R H 2011 Coir fiber reinforced polypropylene composite panel for automotive interior applications *Fibers Polym.* **12** 919–26