

Journal of Advanced Research Design

Journal homepage: https://akademiabaru.com/submit/index.php/ard ISSN: 2289-7984



The Effect of Fibre Loadings on the Mechanical and Thermal Properties of Sugar Palm/Waste Tyre Rubber Reinforced Polylactic Acid Hybrid Composites *via* Fused Deposition Modelling

Batrisyia Norhazlin¹, Nadlene Razali^{1,2,*}, Mohd Adrinata Shaharuzaman^{1,2}, Zaleha Mustafa³, Siti Hajar Sheikh Fadzullah^{1,2}, Bushra Rashid⁴

⁴ Institute of Technology, Middle Technical University, Alzafaranya, Baghdad, 29008, Iraq

ARTICLE INFO

ABSTRACT

The growing need for sustainable and recycled material in order to address the Article history: Received 6 January 2025 environmental challenges associated with petroleum-based plastics has led to this Received in revised form 7 February 2025 study. Sugar palm fiber (SPF) is a renewable, biodegradable, and eco-friendly resource, Accepted 23 May 2025 while waste tire rubber (WTR) enhances the properties of hybrid composites which be Available online 2 June 2025 used in this study. Poly (lactic acid) (PLA) serves as a promising bio-based and biodegradable matrix material for green composites. This hybrid composite material is a novel development specifically designed for complex 3D printing designs. Research on this material is still limited, and it has not been applied in the industry, but it holds great potential for advanced 3D printing applications. To improve interfacial adhesion, sugar palm fiber and waste tyre rubber were treated with 6% sodium hydroxide and 3% silane, resulting in a composite formulation of 97.5% PLA and 2.5% SPF/WTR. Three different fiber loadings were assessed, 75% SPF:25% WTR, 50% SPF:50% WTR, and 25% SPF:75% WTR. The filaments produced using a twin-screw extruder were utilized to 3D print tensile specimens according to ASTM D638-14, impact specimens according to ASTM D256-23e01, and for Thermogravimetric Analysis. Additionally, Scanning Electron Microscopy analysis was conducted to evaluate the morphology of the composites. The results indicated that the 75% SPF:25% WTR loading achieved the highest tensile strength of 37.89 MPa, while the 25% SPF:75% WTR loading exhibited the highest impact strength of 4.3 KJ/m². Thermogravimetric Analysis results demonstrated similar thermal degradation patterns across different compositions, suggesting that the component ratios do not significantly affect overall thermal stability. The SEM images improved interfacial adhesion between the treated fibers and the PLA matrix, which is Keywords: critical for enhancing mechanical performance. In short, these findings indicate that 3D printing; mechanical properties; sugar palm and waste tire rubber hybrid composites are viable, high-performance morphology; poly (lactic acid); sugar palm alternatives for filament extrusion and 3D printing applications, offering both fiber; thermal analysis; waste tire rubber mechanical strength and environmental sustainability.

* Corresponding author E-mail address: nadlene@utem.edu.my

¹ Faculty of Mechanical Engineering and Technology (FTKM), Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100, Durian Tunggal, Melaka, Malaysia

² Centre for Advanced Research on Energy (CARe), Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100, Durian Tunggal, Melaka, Malaysia

³ Faculty of Industrial & Manufacturing Technology & Engineering (FTKIP), Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100, Durian Tunggal, Melaka, Malaysia



1. Introduction

The increasing awareness of environmental issues and the growing emphasis on sustainability have sparked a significant interest in the development of biodegradable materials, particularly in the realm of composites. In order to satisfy this increasing trend, renewable and sustainable resources are being utilized [1]. This trend underscores the importance of utilizing polymers sourced from renewable materials that are generally biodegradable [2]. Among these, poly (lactic acid) (PLA), a biopolymer derived from renewable resources like corn starch and sugarcane, has gained attention as a promising matrix for composite materials due to its lower environmental impact and biodegradability compared to traditional petroleum-based polymers [3]. This study centers on a comparative analysis of PLA-based hybrid composites reinforced with Sugar Palm Fiber (SPF) and Waste Tyre Rubber (WTR). Both materials not only enhance the mechanical properties of the composites but also contribute to sustainability and potentially lowering the cost [4].

Natural fibers (Jute, hemp, kenaf etc) serve as an effective reinforcement in composite materials due to their favorable properties. Compared to synthetic fibers like glass fiber, natural fibers offer several advantages that enhance their appeal in sustainable material development. Other than that, the characteristics between natural fibres and synthetic fibres are quite similar, such as low density, high stiffness, and good mechanical properties [5]. As other natural fibers, SPF recognized for its high strength and durability, serves as an effective reinforcement in composites. Its abundance and lightweight nature make it a valuable resource, while its use helps mitigate agricultural waste, aligning with sustainable practices. Each reinforcement type impacts the composite's properties differently for instance. Conversely, WTR is a recycled material that addresses the pressing issue of tire waste, which poses significant environmental challenges [6]. This unique material is characterized by its exceptional elasticity, toughness, and resistance to abrasion, making it highly valuable across various applications by integrating WTR into PLA composites, improvements in flexibility, impact resistance and overall toughness can be achieved.

Assessing the mechanical properties of these hybrid composites, particularly tensile and impact strengths is crucial for evaluating their performance [7]. Morphological studies, often performed using scanning electron microscopy (SEM) provide insights into the interfacial bonding characteristics and fiber distribution within the composite, which are vital for understanding material behavior [8]. Nonetheless, the use of plant fibers such as sugar palm in polymers raises some concerns, including moisture absorption and strength properties [9]. Poor thermal stability and poor interfacial adhesion between the polymers and plant fibers resulting poor mechanical properties. When moisture is absorbed by natural fibers, dimensional changes and loosening of interfacial adhesion take place [10]. Incompatibilities and wettability may result from the hydrophilic nature of natural fibers and the hydrophobic nature of the polymer matrix [11]. These issues can be clear up by removing hemicellulose, lignin, pectin, and wax, chemically treated fibers to enhance fiber strength and improve surface roughness. Chemical treatments are the most recommended among all of the treatment (Physical, chemical and biological) because of their excellent efficiency [12]. Further, the combination of alkaline and silane treatments enhanced the cellulose density, resulting in a transcrystalline interphase zone containing tiny crystals. The fiber composites had a stronger adhesive bond between the fibers and the matrix than the untreated fiber composites, resulting in a more effective surround and adhesion of the fibers [13].

As a country that is moving forward with IR4.0 development, Malaysia launched a national strategy in 2018 to drive growth in manufacturing [14]. The use of fusion deposition modeling (FDM) in additive manufacturing (AM) is becoming increasingly popular due to its operational flexibility, time efficiency, and low cost, making it the most widely used 3D printing technology [15,16]. Studies



from Knight *et al.*, [13] in FDM there are three-dimensional objects created by stacking layers of thermoplastic materials with one another. By pulling the filament through rollers, the feedstock material is placed in a heated compartment. In this heated compartment, the filament melts and is extruded onto the preheated bed by means of an extrusion nozzle and subsequently cooled to become firm, providing an accurate 3D model of the finished part. As the part is constructed, the software provides the coordinates necessary to move into the x, y, and z planes [13]. Thermoplastics such as PLA are suitable for filament FDM because they possess the appropriate flexural strength and modulus, making filaments easy to spool and extrude as they may reduce material costs, reduce environmental impact, minimize deformation during processing and potentially preserve the mechanical qualities of the material [17]. In addition to the polymer matrix, PLA has been adopted for use with other fillers such as Cork/PLA [18], Hemp/PLA [19], Flax, bamboo/PLA [20], Bamboo fill, Wood fill, Pine lay wood/PLA [21].

In summary, the investigation of PLA-based hybrid composites reinforced with SPF and WTR not only showcases the mechanical and thermal benefits of combining natural and recycled materials but also highlights the necessity of developing sustainable, biodegradable alternatives in material science. Currently, many engineering areas require the use of a combination of materials with different properties, since one type of material may not meet all the needs [22]. Several studies have examined the individual properties of these materials, however there is a knowledge gap regarding their synergism in hybrid composites SPF/WTR, particularly when it comes to 3D printing. A key aspect of this research is to determine the effect of fiber loading on mechanical properties, interfacial adhesion, and thermal stability of 3D printing materials. The purpose of this research is to provide insight into the mechanical, morphological, and thermal properties of these innovative composites, thus contributing to the ongoing effort to create environmentally friendly materials, specifically Fused Deposition Modeling (FDM) filaments.

2. Methodology

This study employs a systematic methodology to develop and evaluate hybrid composites made from sugar palm fiber (SPF) and waste tire rubber (WTR) within a poly (lactic acid) (PLA) matrix. The methodology encompasses the chemical treatment of fibers, the formulation of composite materials, and the application of various mechanical testing protocols to assess tensile strength, impact resistance, and thermal stability. By integrating these approaches, the research aims to optimize the properties of the composites for enhanced performance in 3D printing applications. Thus, the study of the whole process as shown in the flowchart in Figure 1.





Fig. 1. Flowchart of process methodology

2.1 Fiber Treatment

Sugar Palm Fiber (SPF) and Waste Tyre Rubber (WTR) were used as the reinforcing agents in the hybrid composites, while Poly (lactic acid) (PLA)) with a density of 1.24 g/cm3 Ingeo[™] Biopolymer 2003D pellets (100% PLA pure) served as the matrix material. Both SPF and WTR were sourced locally in Malaysia supplied by Mecha Solve Engineering, ensuring the availability and accessibility of these materials for the study. Sugar palm fiber, obtained from raw material sources, undergoes processing to produce fine sizes (125-250µm) [13] suitable for the composite formulation, as illustrated in Figure 2. Optimizing the interfacial properties of both natural fiber and natural rubber relies heavily on surface treatment [23]. Research has demonstrated the impact of alkaline treatment on the chemical modification of natural fiber surfaces and its effects on the tensile and impact properties of natural fiber composites [24]. In this experiment, sugar palm fiber and waste tire rubber were subjected to a 6% sodium hydroxide (NaOH) alkaline solution to enhance interfacial bonding. The treatment of SPF aimed to eliminate hemicellulose, lignin, and other impurities while treatment of WTR is to remove other contaminated that could negatively affect fiber-matrix adhesion [25]. Following the



treatment, after soaked for 3 hours at room temperature the fibers were rinsed with distilled water until a neutral pH was indicated and subsequently dried in an oven at 60°C for 24 hours [26].



(a) (b) (c) Fig. 2. Process to have fine SPF (a) Raw sugar palm fiber (b) Crushed process (c) Sieve process

To enhance the dispersion of reinforcement and adhesion to the matrix, silanes ((3-Aminopropyl) triethoxysilane) were employed alongside surface modifications following alkaline treatment [27]. After the SPF and WTR underwent NaOH treatment, both materials were immersed in a 3% silane solution to facilitate chemical bonding between the fibers and the PLA matrix [28], as indicated in Table 1. Methanol plays a role in the hydrolysis process of silanes, which is crucial for forming silanol groups that can bond with both organic and inorganic materials. The presence of methanol can help control the reaction conditions, influencing the stability and reactivity of the silane during treatment. The PH of the solution was adjusted to 3.5 using acetic acid and stirred continuously for 10 min Following this treatment, after 3 hours of soaked the fibers were thoroughly rinsed with distilled water to eliminate any leftover chemicals until a neutral pH was indicated and then dried in an oven at 60°C for 72 hours to ensure all moisture was removed [29].

Table 1	
Silane Treatment	
Silane concentration	3% of beaker
Methanol	58.5% of beaker
Distilled water	38.5% of beaker

2.2 Composites Preparation

Before compounding, the PLA pellets were dried in an oven at 60°C for 24 hours in order to prevent issues such as brittleness, internal holes, sagging, and other quality and process issues [30]. After drying, the PLA pellets were combined with the treated SPF and WTR fibers using a twin-screw extruder to create hybrid composite filaments with a diameter of 1.75 mm [16]. The composition of these hybrid composites formulation is 97.5% PLA and 2.5% SPF/WTR [26], detailed in Table 2.

Table 2					
Formulation of hybrid composites					
PLA	Hybrid Composites				
97.5%	2.5%				
975g	SPF, wt%	SPF, g	WTR, wt%	WTR, g	
	75	18.75	25	6.25	
	50	12.5	50	12.5	
	25	6.25	75	18.75	

_ . . _



To achieve effective mixing and dispersion of the fibers within the PLA matrix, the extruder was operated according to the temperature profile outlined in Table 3. The extrusion was performed with a twin screw extruder with 26 mm twin screws, co-rotating, 40:1 L/D from Lab Polymer Composite UniKL Ayer Keroh. The samples were loaded into a twin screw extruder barrel. Through the mixing mechanism before the entrance of the twin screw, the SPF/WTR particles and PLA pellets were effectively mixed and transported into the melting compartment where they were extruded through a die. The resulting filaments were then cooled and pelletized for further characterization and processing.

Table 3		
Extrusion setting		
Composites	Melting temperature °C	Screw speed (rpm)
PLA/75%SPF:25%WTR	174.3	20.0
PLA/50%SPF:50%WTR	168.4	20.0
PLA/25%SPF:75%WTR	164.3	18.0

2.3 3D Printing

The prepared filaments were used for 3D printing with a Fused Deposition Modeling (FDM) printer Ender 3. The nozzle temperature was set to 180°C, in accordance with the manufacturer's safety data sheet, while the bed temperature was maintained at 80°C, as PLA does not require high temperatures [31]. Additionally, the printing speed plays a crucial role in the performance of the printed samples in this case, the nozzle speed was set at 60 mm/s, and the travel speed was 80 mm/s. Standard test specimens for tensile and impact testing were created following ASTM D638-14 and ASTM D256-23e01 standards, respectively.

2.4 Mechanical Testing

In this research, five sample of each fiber loading of testing was conducted. For tensile strength it is in accordance with the ASTM D638-14 standard. The Universal Testing Machine model Instron 887, manufactured in Norwood, Massachusetts, United States, was used to determine the tensile properties of the composites. The crosshead speed was set at 1 mm/min, and a load cell with a capacity of 5 kN was employed. To find the average result, five samples were tested for each parameter. The tensile strength of the single fiber can be calculated using the following Eq. (1) where, σ is the tensile strength of the fibre (Pa), F is the maximum force at break (N), and A is the area of the cross section (m²)

$$\sigma = \frac{F}{A} \tag{1}$$

The Izod Pendulum impact ASTM D256-23e01 test is a widely used technique to evaluate the toughness or impact resistance of materials, especially metals. It assesses a material's capacity to withstand fracture and absorb energy from a sudden impact force. To find the average result, five samples were tested for each parameter. The impact strength of composite materials can be calculated using the Knotch Impact Energy Eq. (2) for three-point bending, where KV represents Knotch Impact Energy (kJ), m is the mass of the pendulum (kg), g is acceleration due to gravity (m/s²), H is the height of the pendulum's starting point (m), and h is the height of the pendulum from the first reversal point. The impact characteristics of composite materials are measured using the Charpy-Izod Impact Tester and a standard testing method that includes a hammer weight of 8.8 kg or 58 J.

KV = mgH - mgh



2.5 Morphological Analysis

In this research, detailed morphological studies were conducted on the fracture surface of the tensile test samples using scanning electron microscopy (SEM). Different samples from the tensile specimens were tested to gather comprehensive data. To enhance the resolution of the results, the samples were coated with platinum, which provides excellent electrical conductivity. The micrographs were obtained using a JEC-3000FC SEM Model.

2.6 Thermogravimetric Analysis (TGA) Test

TGA was performed using a Mettler Toledo TGA instrument accordance with the ASTM E1131-20. Samples were heated from 30 to 800 °C at a constant rate of 20 °C/min under a nitrogen atmosphere with a flow rate of 50 mL/min. Each 75% SPF:25% WTR, 50% SPF:50% WTR, and 25% SPF:75% WTR composite sample, weighing between 15 and 20 mg, was analyzed to determine mass loss as a function of increasing temperature and to quantify the final residue yield resulting from composite degradation. The resulting TGA data is presented as a plot of weight percentage versus temperature.

3. Results

3.1 Tensile Strength of The Composition

The Figure 3 below shows a sample that was printed using a fused deposition modelling 3D printer to test the tensile strength. A study by Gao *et al.*, [43] reported that pure PLA exhibits a tensile strength of 39.9 MPa, setting a benchmark for comparison among tested samples, as shown in Figure 4. The incorporation of waste tire rubber (WTR) into PLA has been demonstrated to enhance its tensile strength, reaching approximately 43.55 MPa. This improvement can be attributed to the toughening effect of WTR, which enhances the material's ability to absorb energy under stress. However, the addition of sugar palm fiber (SPF) to PLA tends to decrease the tensile strength significantly, reducing it to 21.9 MPa. This reduction is likely due to weaker interfacial adhesion between the SPF and PLA matrix, which hinders effective stress transfer within the composite.

The tensile strength of PLA hybrid composites, combining both SPF and WTR, varies depending on the fiber loading ratios. Notably, all hybrid blends consistently exhibit higher tensile strength compared to PLA/SPF composites alone, suggesting a synergistic effect between WTR and SPF. Among the hybrid composites, a 75% SPF and 25% WTR ratio achieved the highest tensile strength, approximately 37.89 MPa. Studies from H Anuar *et al.*, [32] state that adding kenaf fiber to thermoplastic natural rubber and glass fiber has lowers the tensile strength, however no significant difference was found between fiber loadings in this study [32]. This result indicates that a higher SPF content in hybrid composite contributes to reinforcing the composite structure, while a moderate WTR content adds toughness without significantly compromising the load-bearing capacity. Parallel with studies from Kumar *et al.*, [7] showed that the increasing of kenaf content in kenaf/coconut epoxy composites enhances the tensile strength of the composites [7], other studies declare by increasing the fiber percentage of sisal/jute/glass fiber-reinforced polyester composites led to a corresponding increase in tensile strength [33]

In contrast, a 25% SPF and 75% WTR blend showed a reduction in tensile strength, emphasizing that excessive WTR content, while beneficial for toughness, can compromise the structural integrity



and load-bearing ability of the composite. The relatively consistent error ranges between ±2-3 MPa suggest good experimental control and measurement precision across different compositions. This highlights the importance of optimizing the fiber loading ratios to achieve a balance between strength and toughness, effectively enhancing the mechanical performance of the composite material. These findings underscore the potential of hybrid composites to tailor material properties for specific applications, leveraging the complementary benefits of SPF and WTR.



Fig. 3. Sample for tensile strength



Fig. 4. Results of tensile strength

3.2 Impact Strength of the Composition

Impact testing is a critical method to evaluate a material's ability to absorb energy during fracture, which provides insights into its toughness and resistance to sudden impacts. This property is especially important in applications where materials are subjected to dynamic or shock-loading conditions. The purpose of the impact test is to assess the material's performance under such scenarios, ensuring safety, durability, and reliability in practical applications. The Figure 5 shows a sample that was printed using a fused deposition modeling 3D printer to test the impact strength.

Figure 6 presents a bar graph illustrating the impact strength of various material compositions. The results reveal that the addition of WTR to PLA significantly enhances impact strength, achieving 6.2 KJ/m², more than double the impact strength of pure PLA, which stands at 2.9 KJ/m² [34]. This



substantial increase can be attributed to the inherent toughness of WTR, which effectively dissipates energy during impact. The elastic nature of WTR likely improves the material's ability to withstand sudden forces, making it an effective toughening agent in the PLA matrix.

In contrast, the combination of PLA with SPF alone reduces the impact strength to 2.32 KJ/m², a decrease compared to pure PLA. This reduction suggests that the introduction of SPF may introduce weaknesses, such as poor interfacial adhesion or stress concentrations, which reduce the composite's ability to absorb energy during impact. These findings highlight the challenges of using natural fibers like SPF, which, while beneficial for certain properties, may compromise toughness if not adequately bonded to the matrix.

Interestingly, hybrid mixtures containing both SPF and WTR exhibit improved impact strength compared to pure PLA, with values consistently ranging between 4.2 and 4.3 KJ/m². Although these values are lower than the PLA/WTR blend, they surpass the impact strength of PLA/SPF composites. Similar with studies from Jawaid *et al.*, [35] research on composites from glass/ kenaf/natural rubber treated with sodium hydroxide and silane, respectively improved impact strength [35]. This indicates that the addition of WTR can counterbalance the reduction in toughness caused by SPF, resulting in a composite with enhanced overall performance. The relatively consistent impact strength across varying ratios of SPF and WTR suggests a balanced interplay between the reinforcing effects of SPF and the toughening effects of WTR.

From these results, it can be concluded that WTR is highly effective in enhancing the impact strength of PLA-based materials, likely due to its energy-absorbing properties. However, the addition of SPF appears to moderate this enhancement, potentially due to its rigidity and weaker interaction with the PLA matrix. This behavior underscores the importance of optimizing the composition of hybrid composites to balance toughness and strength, catering to specific application requirements. A relatively consistent error range between ± 0.4 -0.5 KJ/m2 across various compositions. This consistency of mechanical performance between different formulations of hybrid composites indicates that they can be reliably compared to each other. This strengthens the overall results of the study, as it shows that variations in composition do not introduce significant measurement errors. The study demonstrates the potential of combining SPF and WTR to tailor the mechanical properties of PLA composites, paving the way for the development of materials with improved impact resistance for diverse applications.



Fig. 5. Sample for impact test





Fig. 6. Results of impact strength

3.3 Morphological Properties

To examine the interfacial adhesion between the hybrid fibers and the PLA matrix after the tensile test, morphological analysis was performed using Scanning Electron Microscopy (SEM). The treated SPF and WTR were found to be well-dispersed within the PLA matrix, which is essential for attaining optimal mechanical properties [36]. This uniform fiber distribution reduces voids and improves load transfer efficiency between the fibers and the matrix.



Fig. 7. (a) PLA/75%SPF:25%WTR, (b) PLA/50%SPF:50%WTR, (c) PLA/25%SPF:75%WTR

The analysis clearly indicates the tensile properties of the composites, as illustrated in the accompanying Figure 7(a), 7(b) and 7(c). Sample in Figure 7(a) demonstrates strength compared to the other composites, which can be attributed to the enhanced interfacial bonding between the fibers and the PLA matrix. The irregular shapes in figure 7(a) and potential for interlocking of the phases may contribute to increased toughness and resistance to crack propagation, making the material more durable and resistant to fracture or failure under stress. The effective chemical treatments with sodium hydroxide (NaOH) and silane have significantly improved the compatibility between the hydrophilic fibers and the hydrophobic PLA, resulting in enhanced mechanical performance [37]. In comparison, Sample Figure 7(b) exhibits some fiber pull-out, although it shows



fewer gaps than Sample Figure 7(c). Figure 7(b) shows a different material with more uniform and regular structures. These appear to be well-defined crystalline grains or phases, potentially indicating a more organized and controlled microstructure compared to Figure 7(a). The well-defined, regular crystalline grains or phases observed in this image suggest a highly organized and controlled microstructure. This can contribute to improved mechanical properties, such as strength, stiffness, and dimensional stability, making the material suitable for structural applications. Figure 7(c) displays a material with a varied microstructure. It exhibits a range of different-sized and shaped features due to the 20% of SPF and 75% of WTR, suggesting a potentially heterogeneous composition or a mix of phases within the material. This phenomenon can be attributed to the impurities present in fibers, which lead to weak interfacial bonding between the fibers and the polymer matrix [38]. Consequently, when load is applied, the fibers in Sample Figure 7(c) cannot effectively withstand the stress and detach from the matrix. In other hand, the complex microstructure with its high surface area and varied features may promote increased reactivity, which could be advantageous for applications involving catalysis, adsorption, or energy storage.

3.4 Thermogravimetric Analysis (TGA)

TGA is a standard method to study the overall thermal stability of natural fibers and essential for characterizing the thermal behavior of hybrid composites. This technique provides insights into thermal stability and kinetic parameters by analyzing mass loss as a function of temperature [39]. The resulting data, including degradation temperatures and reaction rates, is critical for predicting material performance in diverse thermal environments on any application [40].

Figure 8 illustrates the thermal degradation behavior of three different SPF/WTR hybrid composites through Thermogravimetric Analysis (TGA). All three compositions (75%SPF:25%WTR, 75%WTR:25%SPF, and 50%SPF:50%WTR) demonstrate similar thermal stability patterns throughout the temperature range of 0-1000°C. The TGA curves reveal a multi-stage degradation process, beginning with the evaporation of water molecules, followed by the decomposition of lignocellulosic components (cellulose and lignin), and culminating in a final residue [41]. Initially, the materials maintain stable weight until reaching approximately 300°C, indicating good thermal resistance at lower temperatures. likely due to the loss of absorbed water, a consequence of its hydrophilic nature. This initial mass loss is consistent with the evaporation or dehydration of weakly bound water and low molecular weight compounds, as reported in previous studies on mixed nano bio-composites [42]. A dramatic weight loss occurs between 350-400°C, where all samples experience rapid thermal decomposition. The decomposition rate slows down after 450°C, and the materials reach a stable residual weight below 20% of their original mass, which continues until the maximum test temperature of 1000°C.

The slight variations in the final residual weights among the different compositions can be attributed to their varying ratios of Sugar Palm Fiber (SPF) and Waste Tire Rubber (WTR) content, suggesting that the composition ratio influences the thermal stability and char formation of these hybrid composites. As shown in Figure 9, cellulose is decomposed at high temperatures, resulting in char residue. As seen in Figure 9, the hybrid composite composite with 75%SPF:25%WTR contains a lower percentage of char residue compared to other hybrid composites, which is probably due to WTR addition. The results of TGA are consistent with previous research that concluded that by adding the WTR as reinforcement material might improve thermal stability. These results provide valuable insights into the thermal behavior and decomposition characteristics of SPF/WTR hybrid composites, which is crucial for understanding their potential applications and limitations in various temperature environments.





4. Conclusions

This study explored the potential of developing sustainable and biodegradable PLA-based hybrid composites reinforced with natural Sugar Palm Fiber (SPF) and recycled Waste Tyre Rubber (WTR). Unlike prior works that primarily assess single-fiber reinforcement or different polymer matrices, this study aims to provide a comprehensive analysis of how varying fiber loadings between two different materials (sugar palm fiber and waste tyre rubber) and treatment methods influence the overall properties of the composite. The goal was to enhance mechanical and thermal properties while promoting environmental sustainability by utilizing renewable resources and reducing waste. Composites with different fiber loading ratios were analyzed, revealing that the 75% SPF:25% WTR combination achieved the highest tensile strength (37.89 MPa), while the 25% SPF:75% WTR composite exhibited the best impact strength (4.3 KJ/m²). These findings highlight the importance of



balancing the stiffness provided by natural fibers and the toughness contributed by rubber to achieve desired mechanical performance. Scanning Electron Microscopy (SEM) analysis validated the positive impact of chemical treatments (6% NaOH and 3% silane) on interfacial adhesion, with treated fibers showing uniform dispersion and enhanced compatibility within the PLA matrix, leading to improved load transfer and overall mechanical performance. The different compositions (75%SPF:25%WTR, 75%WTR:25%SPF, and 50%SPF:50%WTR) displayed similar thermal degradation patterns, suggesting that the ratio of components does not significantly alter the overall thermal stability of the composites. However, slight variations in the final residue content (ranging from 8-12%) indicate that the results demonstrate the potential of PLA-based hybrid composites reinforced with SPF and WTR as sustainable alternatives to conventional petroleum-based materials, offering enhanced properties while contributing to environmental sustainability through the use of renewable resources and waste reduction.

As a result of this research, the hybrid composites can be used for a wide range of consumer goods, including furniture, packaging materials, and household items fabricated by FDM. Their biodegradable nature is consistent with consumer demand for sustainable products. However, hybrid composites may be slow to gain acceptance in traditional industries due to established preferences for synthetic materials that provide proven performance metrics. In order to overcome this barrier, education and awareness are both essential. For further potential research, investigating into various chemical and physical treatments might be enhance the interfacial adhesion and overall performance of natural fibers in composites at a time improve mechanical properties and moisture resistance.

Acknowledgement

The authors gratefully acknowledge the Universiti Teknikal Malaysia Melaka (UTeM) for funding the research work with grant number PJP/2023/TD/FKM/S01976 and Kesidang Scholarship Universiti Teknikal Malaysia Melaka (UTeM).

References

- [1] Razali, Siti Aisyah, Nor Azwadi Che Sidik, and Hasan Koten. "Cellulose nanocrystals: a brief review on properties and general applications." *Journal of Advanced Research Design* 60, no. 1 (2019): 1-15.
- Mazani, N., S. M. Sapuan, M. L. Sanyang, A. Atiqah, and R. A. Ilyas. "Design and fabrication of a shoe shelf from kenaf fiber reinforced unsaturated polyester composites." In *Lignocellulose for future bioeconomy*, pp. 315-332. Elsevier, 2019. <u>https://doi.org/10.1016/B978-0-12-816354-2.00017-7</u>
- [3] Sanyang, Muhammed Lamin, R. A. Ilyas, S. M. Sapuan, and Ridhwan Jumaidin. "Sugar palm starch-based composites for packaging applications." *Bionanocomposites for packaging applications* (2018): 125-147. <u>https://doi.org/10.1007/978-3-319-67319-6_7</u>
- [4] Khashi'le, Najiyah Safwa, and Khairum Hamzah. "Mechanical properties of jute fiber polyester hybrid composite filled with eggshell." *Semarak Eng J* 6, no. 1 (2024): 20-8. <u>https://doi.org/10.37934/sej.6.1.2028</u>
- [5] Bambach, Mike R. "Direct comparison of the structural compression characteristics of natural and synthetic fiberepoxy composites: Flax, jute, hemp, glass and carbon fibers." *Fibers* 8, no. 10 (2020): 62. <u>https://doi.org/10.3390/fib8100062</u>
- [6] Sienkiewicz, Maciej, Helena Janik, Kaja Borzędowska-Labuda, and Justyna Kucińska-Lipka. "Environmentally friendly polymer-rubber composites obtained from waste tyres: A review." *Journal of cleaner production* 147 (2017): 560-571. <u>https://doi.org/10.1016/j.jclepro.2017.01.121</u>
- [7] Sinha, Agnivesh Kumar, Harendra Kumar Narang, and Somnath Bhattacharya. "Mechanical properties of hybrid polymer composites: a review." *Journal of the Brazilian Society of Mechanical Sciences and Engineering* 42, no. 8 (2020): 431. <u>https://doi.org/10.1007/s40430-020-02517-w</u>
- [8] Lee, Ching Hao, Abdan Khalina, and Seng Hua Lee. "Importance of interfacial adhesion condition on characterization of plant-fiber-reinforced polymer composites: a review." *Polymers* 13, no. 3 (2021): 438. <u>https://doi.org/10.3390/polym13030438</u>



- [9] Tengsuthiwat, Jiratti, Vijay Raghunathan, Vinod Ayyappan, Laongdaw Techawinyutham, Rapeeporn Srisuk, Krittirash Yorseng, Sanjay Mavinkere Rangappa, and Suchart Siengchin. "Lignocellulose sustainable composites from agro-waste Asparagus bean stem fiber for polymer casting applications: Effect of fiber treatment." International Journal of Biological Macromolecules 278 (2024): 134884. https://doi.org/10.1016/j.ijbiomac.2024.134884
- [10] Atiqah, A., M. Jawaid, S. M. Sapuan, and M. R. Ishak. "Physical properties of silane-treated sugar palm fiber reinforced thermoplastic polyurethane composites." In *IOP conference series: materials science and engineering*, vol. 368, no. 1, p. 012047. IOP Publishing, 2018. <u>https://doi.org/10.1088/1757-899X/368/1/012047</u>
- [11] Nurazzi, NorizanMohd, M. R. M. Asyraf, M. Rayung, M. N. F. Norrrahim, S. S. Shazleen, M. S. A. Rani, A. R. Shafi et al. "Thermogravimetric analysis properties of cellulosic natural fiber polymer composites: A review on influence of chemical treatments." *Polymers* 13, no. 16 (2021): 2710. <u>https://doi.org/10.3390/polym13162710</u>
- [12] Raghunathan, Vijay, G. Sathyamoorthy, Vinod Ayyappan, D. Lenin Singaravelu, Sanjay Mavinkere Rangappa, and Suchart Siengchin. "Effective utilization of surface-processed/untreated Cardiospermum halicababum agro-waste fiber for automobile brake pads and its tribological performance." *Tribology International* 197 (2024): 109776. <u>https://doi.org/10.1016/j.triboint.2024.109776</u>
- [13] Nasir, Mohd Hakim Mohd, Mastura Mohammad Taha, Nadlene Razali, Rushdan Ahmad Ilyas, Victor Feizal Knight, and Mohd Nor Faiz Norrrahim. "Effect of chemical treatment of sugar palm fibre on rheological and thermal properties of the PLA composites filament for FDM 3D printing." *Materials* 15, no. 22 (2022): 8082. https://doi.org/10.3390/ma15228082
- [14] Mazlan, Muhammad Haziq, Saifullizam Puteh, Zunuwanas Mohamad, Nor Lisa Sulaiman, Kahirol Mohd Salleh, Wan Rosemehah Wan Omar, Rosnawati Buhari, and Hartoyo Mp. "Crafting the future workforce: a Fleiss Kappa exploration of Industry 4.0 talent perspectives." *Higher Education, Skills and Work-Based Learning* (2024). https://doi.org/10.1108/HESWBL-05-2024-0145
- [15] Dilberoglu, Ugur M., Bahar Gharehpapagh, Ulas Yaman, and Melik Dolen. "The role of additive manufacturing in the era of industry 4.0." *Procedia manufacturing* 11 (2017): 545-554. <u>https://doi.org/10.1016/j.promfg.2017.07.148</u>
- [16] Rahim, Tuan Noraihan Azila Tuan, Abdul Manaf Abdullah, and Hazizan Md Akil. "Recent developments in fused deposition modeling-based 3D printing of polymers and their composites." *Polymer Reviews* 59, no. 4 (2019): 589-624. <u>https://doi.org/10.1080/15583724.2019.1597883</u>
- [17] Mazzanti, Valentina, Lorenzo Malagutti, and Francesco Mollica. "FDM 3D printing of polymers containing natural fillers: A review of their mechanical properties." *Polymers* 11, no. 7 (2019): 1094. <u>https://doi.org/10.3390/polym11071094</u>
- [18] Daver, Fugen, Kok Peng Marcian Lee, Milan Brandt, and Robert Shanks. "Cork–PLA composite filaments for fused deposition modelling." *Composites Science and Technology* 168 (2018): 230-237. <u>https://doi.org/10.1016/j.compscitech.2018.10.008</u>
- [19] Lee, Ching Hao, Farah Nadia Binti Mohammad Padzil, Seng Hua Lee, Zuriyati Mohamed Asa'ari Ainun, and Luqman Chuah Abdullah. "Potential for natural fiber reinforcement in PLA polymer filaments for fused deposition modeling (FDM) additive manufacturing: A review." *Polymers* 13, no. 9 (2021): 1407. <u>https://doi.org/10.3390/polym13091407</u>
- [20] P. Kumar, A. Robins, and H. Apsimon, "Production and characterization of bambo," vol. 27, no. March, pp. 327– 331, 2010. <u>https://doi.org/10.1002/asl.307</u>
- [21] Krapež Tomec, Daša, and Mirko Kariž. "Use of wood in additive manufacturing: review and future prospects." *Polymers* 14, no. 6 (2022): 1174. <u>https://doi.org/10.3390/polym14061174</u>
- [22] sabri Abbas, Zainab, Mustafa M. Kadhim, Ahmed Mahdi Rheima, Alaa dhari jawad al-bayati, Zainab Talib Abed, Firas mohamed dashoor Al-Jaafari, Asala Salam Jaber et al. "Preparing hybrid nanocomposites on the basis of resole/graphene/carbon fibers for investigating mechanical and thermal properties." *BioNanoScience* 13, no. 3 (2023): 983-1011. <u>https://doi.org/10.1007/s12668-023-01119-9</u>
- [23] Imraan, M., R. A. Ilyas, A. S. Norfarhana, Sneh Punia Bangar, Victor Feizal Knight, and M. N. F. Norrrahim. "Sugar palm (Arenga pinnata) fibers: new emerging natural fibre and its relevant properties, treatments and potential applications." *Journal of Materials Research and Technology* 24 (2023): 4551-4572. https://doi.org/10.1016/j.jmrt.2023.04.056
- [24] Huzaifah, M. R. M., S. M. Sapuan, Z. Leman, M. R. Ishak, and M. A. Maleque. "A review of sugar palm (Arenga pinnata): Application, fibre characterisation and composites." *Multidiscipline Modeling in Materials and Structures* 13, no. 4 (2017): 678-698. <u>https://doi.org/10.1108/MMMS-12-2016-0064</u>



- [25] Nur Saadah Zainal, Zaleha Mohamad, Mohammad Sukri Mustapa, Fazimah Mat Noor, and Nur Azam Badarulzaman. 2023. "Effect of Thermal and Alkali Treatment on Morphological Analysis of Natural Bamboo Fibre". Journal of Advanced Research in Fluid Mechanics and Thermal Sciences 110 (1):157-71. https://doi.org/10.37934/arfmts.110.1.157171
- [26] Jamadi, Aida Haryati, Nadlene Razali, Michal Petrů, Mastura Mohammad Taha, Noryani Muhammad, and Rushdan Ahmad Ilyas. "Effect of chemically treated kenaf fibre on mechanical and thermal properties of PLA composites prepared through fused deposition modeling (FDM)." *Polymers* 13, no. 19 (2021): 3299. <u>https://doi.org/10.3390/polym13193299</u>
- [27] Torrado Perez, Angel R., David A. Roberson, and Ryan B. Wicker. "Fracture surface analysis of 3D-printed tensile specimens of novel ABS-based materials." *Journal of Failure Analysis and Prevention* 14 (2014): 343-353. https://doi.org/10.1007/s11668-014-9803-9
- [28] Georgiopoulos, Panayiotis, Evagelia Kontou, and George Georgousis. "Effect of silane treatment loading on the flexural properties of PLA/flax unidirectional composites." *Composites Communications* 10 (2018): 6-10. <u>https://doi.org/10.1016/j.coco.2018.05.002</u>
- [29] Jamadi, Aida Haryati, Nadlene Razali, Michal Petrů, Mastura Mohammad Taha, Noryani Muhammad, and Rushdan Ahmad Ilyas. "Effect of chemically treated kenaf fibre on mechanical and thermal properties of PLA composites prepared through fused deposition modeling (FDM)." *Polymers* 13, no. 19 (2021): 3299. <u>https://doi.org/10.3390/polym13193299</u>
- [30] Suharjanto, G., and J. P. Adi. "Design and manufacture of polylacticacid (PLA) filament storage for 3-dimensional printing with composite material." In *IOP Conference Series: Earth and Environmental Science*, vol. 998, no. 1, p. 012028. IOP Publishing, 2022. <u>https://doi.org/10.1088/1755-1315/998/1/012028</u>
- [31] MorCh, Nine. "Analysis of the Effects of Basic Printer Settings on PLA 3D Prints." PhD diss., Massachusetts Institute of Technology, 2023.
- [32] Anuar, H., W. N. Wan Busu, S. H. Ahmad, and R. Rasid. "Reinforced thermoplastic natural rubber hybrid composites with Hibiscus cannabinus, I and short glass fiber—part I: processing parameters and tensile properties." *Journal of Composite Materials* 42, no. 11 (2008): 1075-1087. <u>https://doi.org/10.1177/0021998308090450</u>
- [33] Kistaiah, N., C. Udaya Kiran, G. Ramachandra Reddy, and M. Sreenivasa Rao. "Mechanical characterization of hybrid composites: A review." Journal of Reinforced Plastics and Composites 33, no. 14 (2014): 1364-1372. <u>https://doi.org/10.1177/0731684413513050</u>
- [34] Sherwani, S. F. K., S. M. Sapuan, Z. Leman, E. S. Zainudin, and A. Khalina. "Physical, mechanical and morphological properties of sugar palm fiber reinforced polylactic acid composites." *Fibers and Polymers* 22, no. 11 (2021): 3095-3105. <u>https://doi.org/10.1007/s12221-021-0407-1</u>
- [35] Jawaid, M., HPS Abdul Khalil, and A. Abu Bakar. "Mechanical performance of oil palm empty fruit bunches/jute fibres reinforced epoxy hybrid composites." *Materials Science and Engineering: A* 527, no. 29-30 (2010): 7944-7949. <u>https://doi.org/10.1016/j.msea.2010.09.005</u>
- [36] Sherwani, S. F. K., S. M. Sapuan, Z. Leman, E. S. Zainudin, and A. Khalina. "Physical, mechanical and morphological properties of sugar palm fiber reinforced polylactic acid composites." *Fibers and Polymers* 22, no. 11 (2021): 3095-3105. <u>https://doi.org/10.1007/s12221-021-0407-1</u>
- [37] Aisyah, H. A., Mohamad Thariq Paridah, S. M. Sapuan, Raden Asyraf Ilyas, Ahmad Khalina, N. M. Nurazzi, Seng Hua Lee, and Chien Hsin Lee. "A comprehensive review on advanced sustainable woven natural fibre polymer composites." *Polymers* 13, no. 3 (2021): 471. <u>https://doi.org/10.3390/polym13030471</u>
- [38] Fiore, V., G. Di Bella, and A. J. C. P. B. E. Valenza. "The effect of alkaline treatment on mechanical properties of kenaf fibers and their epoxy composites." *Composites Part B: Engineering* 68 (2015): 14-21. <u>https://doi.org/10.1016/j.compositesb.2014.08.025</u>
- [39] Asim, Mohammad, Mohd T. Paridah, M. Chandrasekar, Rao M. Shahroze, Mohammad Jawaid, Mohammed Nasir, and Ramengmawii Siakeng. "Thermal stability of natural fibers and their polymer composites." *Iranian Polymer Journal* 29 (2020): 625-648. <u>https://doi.org/10.1007/s13726-020-00824-6</u>
- [40] Sahari, J., S. M. Sapuan, E. S. Zainudin, and M. A. Maleque. "Thermo-mechanical behaviors of thermoplastic starch derived from sugar palm tree (Arenga pinnata)." *Carbohydrate Polymers* 92, no. 2 (2013): 1711-1716. <u>https://doi.org/10.1016/j.carbpol.2012.11.031</u>
- [41] Radzi, A. M., S. M. Sapuan, M. Jawaid, and M. R. Mansor. "Effect of alkaline treatment on mechanical, physical and thermal properties of roselle/sugar palm fiber reinforced thermoplastic polyurethane hybrid composites." *Fibers* and Polymers 20 (2019): 847-855. <u>https://doi.org/10.1007/s12221-019-1061-8</u>
- [42] Ilyas, Rushdan Ahmad, Salit Mohd Sapuan, Rushdan Ibrahim, Hairul Abral, M. R. Ishak, E. S. Zainudin, Mochamad Asrofi et al. "Sugar palm (Arenga pinnata (Wurmb.) Merr) cellulosic fibre hierarchy: A comprehensive approach from macro to nano scale." *Journal of Materials Research and Technology* 8, no. 3 (2019): 2753-2766. <u>https://doi.org/10.1016/j.jmrt.2019.04.011</u>



- [43] Gao, Ge, Fan Xu, Jiangmin Xu, and Zhenyu Liu. "Study of material color influences on mechanical characteristics of fused deposition modeling parts." Materials 15, no. 19 (2022): 7039. <u>https://doi.org/10.3390/ma15197039</u>
- [44] "Standard Test Method for Tensile Properties of Plastics," n.d. Retrieved from: <u>https://store.astm.org/d0638-</u> <u>14.html</u>
- [45] "Standard Test Methods for Determining the IZOD Pendulum Impact Resistance of Plastics," n.d. Retrieved from: https://store.astm.org/d0256-10r18.html
- [46] "Standard Test Method for Compositional Analysis by Thermogravimetry," n.d. Retrieved from: https://store.astm.org/e1131-20.html