Structure-Property Relationships in Thermoplastic Cassava Starch/Candelilla Wax Biocomposites Reinforced with *Pennisetum purpureum* Fibers

Mohd Fairus Kayat [a], Ridhwan Jumaidin [b,*] Mastura Mohamad Taha,[a,*] Fahmi Asyadi Md Yusof,[c] Zatil Hafila Kamaruddin,[d] Mohammad Khalid Wahid,[a] and Melbi Mahardika [e]

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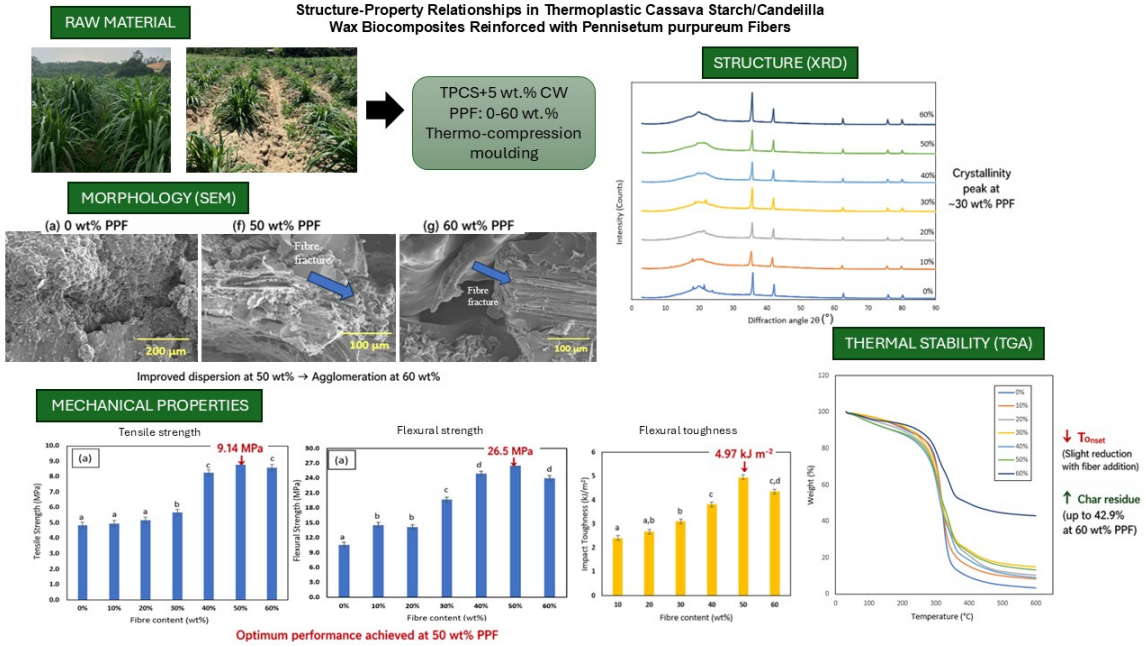
GRAPHICAL ABSTRACT



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Thermoplastic starch-based composites often suffer from poor mechanical performance and high moisture sensitivity. This limits their practical applications. In this study, thermoplastic cassava starch modified with candelilla wax (TPCS/CW) was reinforced with *Pennisetum purpureum* fiber (PPF) at loadings ranging from 0 to 60 wt% using thermo-compression moulding. The structural, morphological, thermal, and mechanical properties of the composites were evaluated using Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), X-ray diffraction (XRD), thermogravimetric analysis (TGA), and mechanical testing. The results showed that fiber incorporation enhanced intermolecular interactions without altering the chemical structure of the starch matrix. Mechanical properties improved significantly with increasing fiber content, with tensile strength and modulus reaching optimum values of 9.14 MPa and 3.28 GPa, respectively, at 50 wt% PPF. Flexural strength and impact toughness also showed maximum values of 26.5 MPa and 4.97 kJ m^{-2} at 50 wt% PPF. Thermogravimetric analysis indicated a slight reduction in onset degradation temperature with fiber addition, accompanied by a notable increase in char residue. However, excessive fiber loading (60 wt%) led to fiber agglomeration and reduced performance. These findings demonstrate that PPF is an effective reinforcement for TPCS/CW composites, with potential applications in biodegradable packaging, disposable products, and low-load structural components.

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Keywords: Pennisetum purpureum fiber; Thermoplastic cassava starch; Candelilla wax; Mechanical properties; Thermogravimetric analysis; Scanning electron microscope

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INTRODUCTION

The extensive use of petroleum-based plastics in packaging and structural applications has led to serious environmental concerns because of their non-biodegradable nature and long-term accumulation in ecosystems. In response, significant research efforts

have been directed toward the development of biodegradable polymers derived from renewable resources as sustainable alternatives (Cataño *et al.* 2023; Arruda *et al.* 2025). Among these materials, starch-based polymers have gained considerable attention owing to their abundance, renewability, biodegradability, low cost, and wide availability from agricultural sources such as cassava, corn, potato, and sago (Syazmini *et al*. 2023; Arruda *et al*. 2025).

Cassava starch is particularly attractive for biodegradable material development in tropical regions due to its high availability and ease of processing. However, native starch exhibits several inherent limitations, including high hydrophilicity, low thermal stability, and poor mechanical strength when compared with conventional petroleum-based plastics (Hazrati *et al.* 2021; Wang *et al.* 2023). These drawbacks restrict its direct application in engineering and packaging fields. Plasticization using agents such as glycerol, sorbitol, or citric acid is commonly employed to convert native starch into thermoplastic starch (TPS), enabling processing under heat and pressure (Hazrati *et al.* 2021; Syazmini *et al.* 2023). Despite this improvement, TPS remains mechanically weak and highly sensitive to moisture, limiting its broader applicability (Wang *et al.* 2023; Arruda *et al.* 2025). Previous studies on thermoplastic starch-based systems have shown that the incorporation of natural components can significantly influence mechanical, thermal, and biodegradation behaviour depending on matrix–filler interactions (Jumaidin *et al.* 2017).

One effective strategy to overcome these limitations is the reinforcement of TPS with natural fibers. Natural fibers are renewable, lightweight, biodegradable, and capable of improving stiffness, strength, and dimensional stability through effective stress transfer and interfacial interactions (Azlin *et al.* 2020; Schutz *et al.* 2024). In starch-based composites, the presence of hydroxyl groups in both starch and lignocellulosic fibers promotes hydrogen bonding, which enhances fiber–matrix adhesion and restricts polymer chain mobility, leading to improved mechanical performance (Sabapathy *et al.* 2025). However, the reinforcing efficiency of natural fibers strongly depends on fiber type, composition, morphology, and loading level. Excessive fiber incorporation often results in agglomeration, poor dispersion, and interfacial voids that compromise structural integrity and mechanical properties (Fazeli *et al.* 2019; Ibrahim *et al.* 2020). The influence of fibre composition, structure, and maturity on the mechanical and thermal performance of natural fibre composites has been widely reported, where variations in fibre characteristics significantly affect reinforcement efficiency (Jawaid *et al.* 2011; Razali *et al.* 2015).

Recent studies have demonstrated the growing potential of grass-based and lignocellulosic fibers in thermoplastic starch composites. Pattnaik *et al.* (2025a) reported that grass fiber-reinforced composites exhibit significant improvements in physico-mechanical properties, particularly in stiffness and dimensional stability, while maintaining environmental sustainability. In another study, Pattnaik *et al.* (2023) showed that the incorporation of waste vetiver root fibers into soy-based biocomposites enhanced mechanical strength, hydrophobicity, and biodegradability, highlighting the importance of fiber–matrix compatibility in improving moisture resistance. Furthermore, Pattnaik *et al.* (2025b) demonstrated that fiber-reinforced composites exhibited improved biodegradation behaviour under different environmental conditions, confirming their suitability for sustainable material applications.

The role of natural fibers in thermoplastic starch systems has also been widely investigated. Behera *et al.* (2021) reported that the incorporation of lignocellulosic fibers into thermoplastic starch significantly enhanced biodegradability under soil and fungal conditions, while maintaining acceptable mechanical properties. More recently, Behera *et*

al. (2025) developed water hyacinth fiber-reinforced thermoplastic starch composites and demonstrated that fiber addition improved mechanical performance and structural integrity. However, they also noted that excessive fiber loading led to reduced interfacial quality and increased structural heterogeneity, which negatively affected composite performance.

In addition to these studies, recent developments in starch-based composites have emphasized the importance of optimizing interfacial interactions and processing strategies to achieve balanced performance. Advanced research has shown that the mechanical and barrier properties of thermoplastic starch composites can be significantly enhanced through improved fiber dispersion, surface compatibility, and matrix modification (Pattnaik *et al.* 2025a). Other studies have further highlighted that fiber surface characteristics and composite formulation play critical roles in determining mechanical behaviour, moisture resistance, and long-term durability (Pattnaik *et al.* 2023, 2025b). These findings collectively indicate that achieving a balance between mechanical performance, thermal stability, and moisture resistance remains a key challenge in the development of starch-based biocomposites.

In addition to fiber reinforcement, the incorporation of hydrophobic additives has been shown to improve the performance of starch-based systems. Candelilla wax (CW), a plant-derived wax with high hydrophobicity, crystallinity, and thermal stability, has attracted increasing interest in bio-based materials because of its ability to act as a barrier agent and structural modifier (Núñez-García *et al.* 2022; Kowalczyk *et al.* 2024). CW is widely recognized for its regulatory acceptance, being approved as a food additive (E 902) in the European Union and designated as Generally Recognized as Safe (GRAS) in the United States (Aranda-Ledesma *et al.* 2022). Previous studies have demonstrated that CW incorporation into starch and protein matrices can reduce water vapour permeability and enhance structural integrity without compromising biodegradability (Núñez-García *et al.* 2022; Kowalczyk *et al.* 2024).Recent developments in starch modification and wax-based composite systems have further highlighted the role of structural design and material interactions in improving functional and barrier properties (Pawase *et al.* 2025; Qu *et al.* 2024).

Pennisetum purpureum, commonly known as Napier grass or elephant grass, is a fast-growing perennial C4 grass that has gained attention as a lignocellulosic fiber source due to its high biomass yield and favourable chemical composition (Islam *et al.* 2023). Its fibers are rich in cellulose and hemicellulose, with moderate lignin content, making them suitable for reinforcement applications in biodegradable composites (Beber *et al.* 2025). The fiber characteristics of *Pennisetum purpureum* are strongly influenced by harvesting age, which governs cellulose crystallinity, fiber rigidity, and interfacial compatibility with polymer matrices (Dresch *et al.* 2025; Meel *et al.* 2025). At early vegetative stages, approximately 30 days of growth, the fibers exhibit lower lignification and higher chemical reactivity, favouring interaction with hydrophilic matrices such as thermoplastic starch (Meel *et al.* 2025).

Although numerous studies have investigated thermoplastic cassava starch reinforced with various natural fibers, and others have reported the benefits of wax-modified starch systems, the combined incorporation of *Pennisetum purpureum* fiber (PPF) into thermoplastic cassava starch modified with candelilla wax has not been systematically explored. In particular, there is limited understanding of how fiber loading governs the structural evolution, interfacial interactions, thermal behaviour, and mechanical performance of this composite system. Therefore, the present study aims to

investigate the structure–property relationships of thermoplastic cassava starch/candelilla wax (TPCS/CW) composites reinforced with varying loadings of PPF. The effects of fiber content on molecular interactions, morphology, crystallinity, thermal stability, and mechanical properties are systematically evaluated to identify the optimum fiber loading for enhanced performance in fully bio-based composite materials.

EXPERIMENTAL

Materials

Pennisetum purpureum grass was obtained from Ladang Napier Selatan, Rembau, Negeri Sembilan, Malaysia, as shown in Fig. 1. The grass was harvested at 30 days of age, corresponding to the early vegetative stage commonly reported for fiber extraction and composite applications (Zaini *et al.* 2021; Onjai-uea *et al.* 2023). The harvested stems were washed to remove surface contaminants and immersed in fresh water for 14 days to allow retting. After retting, the fibers were manually extracted, rinsed with distilled water, sun-dried for 48 h, and oven-dried at 85 °C for 24 h to remove residual moisture (Huzaifah *et al*. 2017). The dried fibers were then cut and ground into short fibers with an average length of approximately 1.0 cm.

Cassava starch (food-grade) was supplied by THC Sdn. Bhd. (Malaysia). Glycerol (analytical grade, 99.5% purity), used as a plasticiser for thermoplastic starch preparation, was purchased from QReC Chemicals Sdn. Bhd. (Malaysia). Candelilla wax (industrial grade), used as a hydrophobic additive, was obtained from Evergreen Engineering and Resources Sdn. Bhd. (Malaysia).

Thermoplastic cassava starch (TPCS) was prepared by plasticizing cassava starch with 30 wt% glycerol under heat and pressure during thermo-compression moulding. In this system, glycerol acts as the primary plasticiser by disrupting intermolecular hydrogen bonding within starch granules, thereby enabling thermoplastic behaviour. Residual moisture may also contribute as a secondary plasticising agent during processing.

Fig. 1. *Pennisetum purpureum* harvested at 30 days from Ladang Napier Selatan, Rembau, Negeri Sembilan, Malaysia

Preparation of TPCS/CW/PPF Composites

Thermoplastic cassava starch/candelilla wax (TPCS/CW) composites reinforced with *Pennisetum purpureum* fiber (PPF) were prepared using a consistent formulation and processing procedure. The matrix composition consisted of 65 wt% cassava starch, 30 wt% glycerol, and 5 wt% candelilla wax. PPF was incorporated at fiber loadings of 0, 10, 20, 30, 40, 50, and 60 wt% relative to the total composite weight.

All components were pre-mixed using a Panasonic MX-GM1011 dry mixer (Shah Alam, Selangor, Malaysia) at 1200 rpm for 5 min at ambient temperature to ensure homogeneous dispersion. The blended mixtures were then compression moulded using a GOTECH GT7014-P30 C Plastic Hydraulic Moulding Press (GOTECH Testing Inc., Taiwan) at 150 °C for 60 min under a constant load of 10 tonnes. The resulting composite sheets had an average thickness of approximately 3 mm. After fabrication, the specimens were stored in a sealed desiccator containing silica gel to minimize moisture uptake prior to testing.

Analysis of FT-IR

FTIR spectroscopy was performed using a JASCO FTIR-6100 spectrometer (Japan) to examine the functional groups and intermolecular interactions between the TPCS/CW matrix and PPF. Infrared spectra were recorded over a wavenumber range of 4000 to 500 cm^{-1}.

Scanning Electron Microscope

The fracture surfaces of tensile-tested specimens were examined using a Zeiss EVO 18 Research (Germany) scanning electron microscope (SEM) operated at an accelerating voltage of 10 kV. Prior to imaging, the specimens were sputter-coated with a thin layer of gold to improve conductivity. The tensile samples were kept in sealed plastic bags before SEM observation to minimize contamination and moisture uptake.

X-Ray Diffraction

X-ray diffraction (XRD) analysis was conducted using a Rigaku D/max 2500 diffractometer (Rigaku, Japan) equipped with CuKα radiation (λ = 1.5406 nm) operating at 40 kV and 35 mA. As per Eq. 1, the crystallinity index (CI) was calculated based on the intensities of the crystalline (I_{002}) and amorphous (I_{am}) regions using the established method reported in the literature (Reddy *et al*. 2018).

$$Crystalline\ Index, CI\ (\%) = \frac{I002 - Iam}{I002}\ x\ 100 \quad (1)$$

Thermogravimetric Analysis

Thermogravimetric analysis (TGA) was carried out using a Mettler-Toledo Q-series thermal analyzer (Switzerland). Samples were heated from 25 to 600 °C at a heating rate of 10 °C/min under a nitrogen atmosphere. TG and DTG curves were recorded to evaluate thermal degradation behavior.

Tensile Test

Tensile testing was performed in accordance with ASTM D638 (2014) under controlled conditions of 50 ± 5% relative humidity and 23 ± 1 °C. The tests were conducted using a universal testing machine (Instron 5969, USA) at a crosshead speed of 5 mm/min

with a 50 kN load cell. For each composition, five specimens were tested, and the resulting values were averaged to obtain the tensile properties.

Flexural Test

Flexural testing was carried out in accordance with ASTM D790 (2017) under controlled conditions of 50 ± 5% relative humidity and 23 ± 1 °C. The specimens had dimensions of 130 × 13 × 3 mm³ (length × width × thickness) and were tested using a universal testing machine (Instron 5969, USA) at a crosshead speed of 2 mm/min with a 50 kN load cell. For each composition, five specimens were tested and the average values were reported.

Impact Test

Izod impact testing was conducted in accordance with ASTM D256 (2023) under controlled conditions of 23 ± 1 °C and 50 ± 5% relative humidity. The specimens had dimensions of 60 × 13 × 3 mm³ (length × width × thickness), with five replicates tested for each composition. A digital pendulum impact tester supplied by Victor Equipment Resources Sdn. Bhd. (Malaysia) was used to perform the measurements. Prior to testing, all specimens were conditioned for two days, followed by an additional two days at 53% relative humidity to ensure consistent moisture content. The impact strength was calculated from the absorbed impact energy and the cross-sectional area of the specimens according to Eq. 2.

$$\text{Impact strength} = \text{Impact energy (J)} / \text{Cross-sectional area (mm}^2\text{)} \quad (2)$$

Statistical Analysis

Statistical analysis was performed using one-way analysis of variance (ANOVA) at a significance level of $P < 0.05$. Duncan's multiple range test was applied as a post-hoc procedure for mean comparison. Results are presented as mean ± standard deviation. In the graphical representations, different lowercase letters (a, b, c, d, *etc*.) denote statistically significant differences between groups at $P < 0.05$, while identical letters indicate no significant difference among the compared means.

RESULTS AND DISCUSSION

Fourier Transform Infrared Analysis

The FTIR spectra of thermoplastic cassava starch/candelilla wax (TPCS/CW) composites reinforced with *Pennisetum purpureum* fiber (PPF) at different fiber loadings are shown in Fig. 2. All spectra exhibited similar absorption profiles, indicating that fiber incorporation did not introduce new chemical bonds or alter the fundamental chemical structure of the starch matrix. This behaviour is consistent with previous studies on starch–cellulose composites, where fiber addition primarily enhances physical interactions rather than chemical modification (Ek *et al.* 2021).

A broad absorbance band observed in the range of 3200 to 3500 cm^{-1} corresponds to O–H stretching vibrations arising from hydroxyl groups present in starch and lignocellulosic fibers. With increasing fiber loading, this peak shifted from 3280 cm^{-1} in neat TPCS to 3329 cm^{-1} at 60 wt% PPF. Such peak shifting is commonly associated with enhanced hydrogen bonding interactions between fiber and matrix, where stronger intermolecular interactions reduce vibrational energy (Novak and Grdadolnik 2021;

Pocheć *et al.* 2022). This enhanced hydrogen bonding restricts polymer chain mobility and contributes to improved mechanical performance (Sabapathy *et al.* 2025).

Additional absorbance peaks in the region of 2850 to 3000 cm^{-1} were attributed to C–H stretching vibrations of cellulose, while bands between 1000 and 1300 cm^{-1} corresponded to C–O stretching of cellulose and hemicellulose structures (Prochoń *et al.* 2019). The persistence of these peaks across all compositions confirms that fiber incorporation enhanced intermolecular interactions without disrupting the polysaccharide backbone.

The absence of new absorption peaks indicates that no covalent chemical bonding was formed between the fibre and the matrix. Instead, the interaction between PPF and the TPCS/CW matrix is primarily governed by physical interactions, particularly intermolecular hydrogen bonding between hydroxyl (–OH) groups present in both starch and cellulose. The observed shift in the O–H stretching band toward higher wavenumbers suggests changes in hydrogen bonding intensity, which is commonly associated with improved interfacial interaction. Therefore, the formation of the composite is validated through these intermolecular interactions, supported by corresponding improvements in mechanical properties and morphological evidence from SEM analysis.

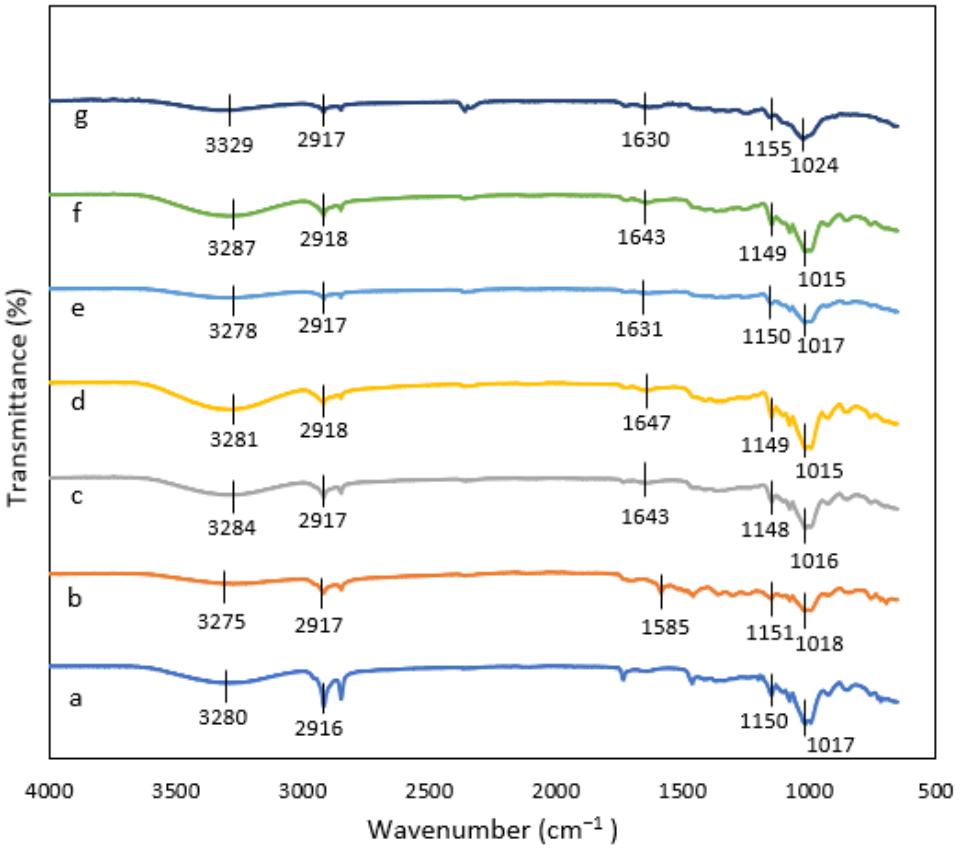

Fig. 2. FTIR spectra of (a) neat TPCS/CW matrix (0% PPF) and TPCS/CW/PPF composites with PPF contents of (b) 10%, (c) 20%, (d) 30%, (e) 40%, (f) 50%, and (g) 60%

Scanning Electron Microscope Analysis

SEM micrographs of tensile-fractured surfaces of neat TPCS/CW and fiber-reinforced composites are presented in Fig. 3. The neat TPCS/CW (Fig. 3a) exhibited a smooth and compact fracture surface, indicative of homogeneous plasticisation by glycerol. Upon fiber incorporation, significant changes in fracture morphology were observed.

At low to moderate fiber contents (10 to 40 wt%) as shown in Figs. 3b-e, fibers were well embedded within the matrix, with evidence of fiber fracture and limited pull-out. These features indicate strong fiber–matrix adhesion and effective stress transfer during mechanical loading, consistent with the hydrogen bonding observed in FTIR analysis (Jagadeesh *et al*. 2021). At 50 wt% PPF (Fig. 3f), fiber fracture dominated over fiber pull-out, suggesting optimal interfacial bonding and reinforcing efficiency. Such fracture patterns are characteristic of composites where hydrogen bonding between fiber and starch matrix enhances adhesion (Jagadeesh *et al*. 2021; Nurazzi *et al.* 2021).

In contrast, composites containing 60 wt% PPF (Fig. 3g) exhibited fiber agglomeration, voids, and extensive fiber pull-out, indicating inadequate wetting of the fibre surfaces by the matrix in its molten state and poor fibre dispersion. Such defects act as stress concentration sites and reduce mechanical performance, a phenomenon widely reported in starch-based composites reinforced with excessive fiber loading (Fazeli *et al.* 2019; Ibrahim *et al.* 2020). These morphological observations directly explain the decline in mechanical properties beyond the optimum fiber loading.

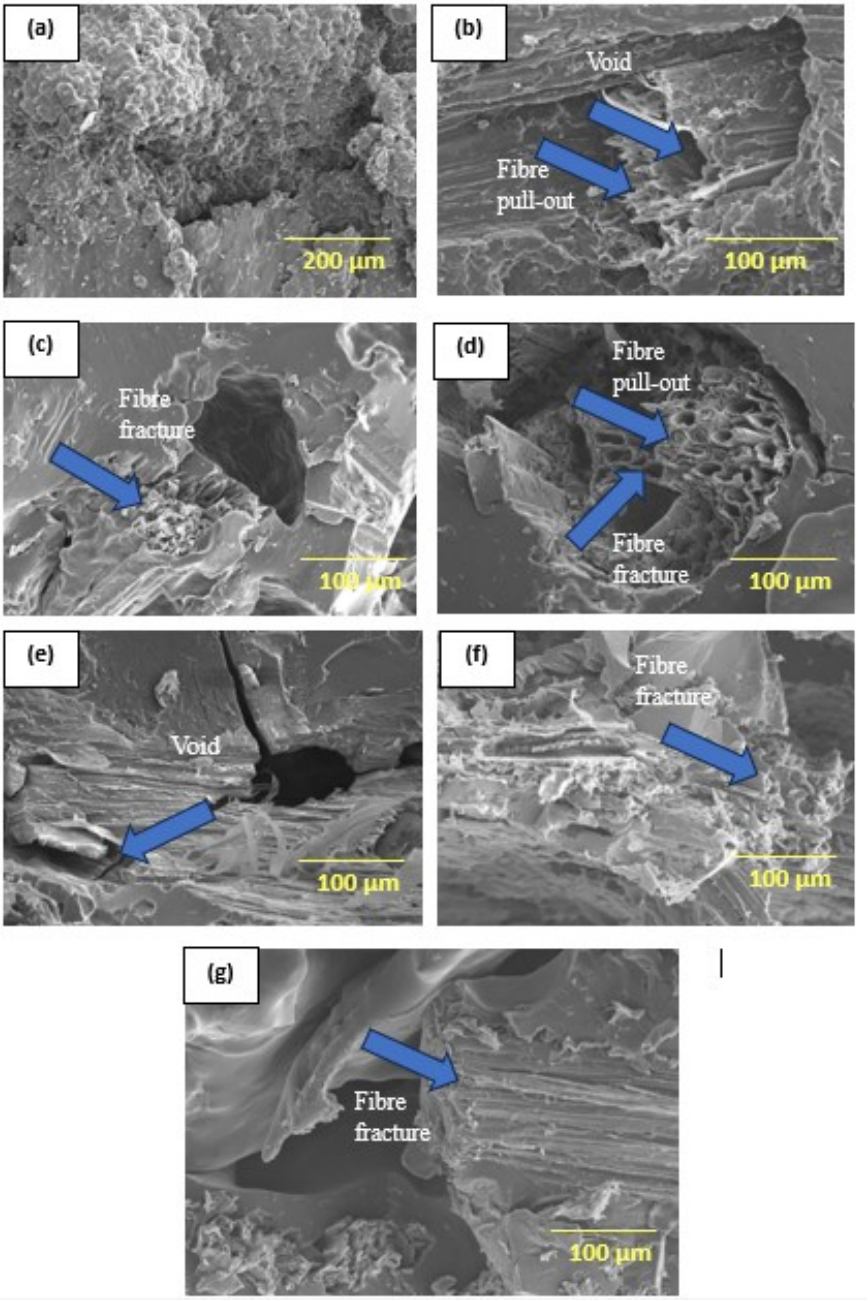

Fig. 3. SEM micrographs of the fractured surfaces of TPCS/CW composites reinforced with different *Pennisetum purpureum* fiber (PPF) contents: (a) neat TPCS/CW, (b) TPCS/CW/PPF with 10 wt%, (c) 20 wt%, (d) 30 wt%, (e) 40 wt%, (f) 50 wt%, and (g) 60 wt%

X-ray Diffraction

The crystalline structure of TPCS/CW composites reinforced with PPF was analysed using XRD in order to evaluate the influence of fiber loading on molecular ordering within the composite system. The XRD diffractograms of neat TPCS and fiber-reinforced composites are presented in Fig. 4, while the corresponding crystallinity index (CI) values are summarised in Table 2.

All samples exhibited characteristic diffraction peaks associated with starch-based materials, confirming the retention of the starch crystalline structure after composite fabrication. The broad diffraction peaks observed are primarily associated with semi-

crystalline starch structures, typically corresponding to A-type crystallinity formed during retrogradation after thermo-compression processing. In fiber-reinforced samples, an increase in peak intensity was observed in the 2θ range of approximately 20° to 23°, which is commonly attributed to cellulose I crystalline reflections originating from the PPF. Therefore, the diffraction patterns represent a combined contribution from both the starch matrix and the cellulose structure of the fibres. The increase in peak intensity with fiber loading reflects the increasing contribution of cellulose crystallinity rather than changes in the starch crystal structure alone (Aaliya *et al.* 2021; Arruda *et al.* 2025).

The neat TPCS sample exhibited a CI of 62.3%, reflecting the partially crystalline nature of plasticized cassava starch. This crystallinity is typically associated with an A-type starch structure, which forms as a result of starch retrogradation during cooling following thermo-compression processing (Edhirej *et al.* 2017; Kamaruddin *et al.* 2023; Wang *et al.* 2025). The presence of glycerol as a plasticiser disrupts native starch crystallites during processing, but partial recrystallisation occurs upon cooling, leading to the observed semi-crystalline structure.

Upon incorporation of PPF, a slight reduction in CI was observed at low fiber loadings. At 10 wt% and 20 wt% PPF, CI values decreased to 59.9% and 59.6%, respectively. This initial reduction in crystallinity can be attributed to the disruption of starch crystalline regions caused by fiber dispersion within the matrix. At these fiber contents, the presence of randomly distributed fibers may interfere with starch chain alignment and retrogradation, thereby increasing the amorphous fraction of the composite (Arruda *et al*. 2025).

Interestingly, a notable increase in crystallinity was recorded at 30 wt% PPF, where the CI reached 63.7%, exceeding that of the neat TPCS matrix. This increase suggests a synergistic interaction between the starch matrix and the crystalline cellulose domains of the PPF. At this fiber loading, the cellulose crystals may act as nucleating sites that promote more ordered packing and alignment of adjacent starch chains, thereby enhancing overall molecular ordering within the composite (Ibrahim *et al.* 2020; Liu *et al.* 2022; Kibet *et al*. 2025). Such behaviour has been reported in other starch–fiber systems, where moderate fiber contents facilitate crystallite organisation rather than disrupting it.

However, further increases in fiber loading beyond 30 wt% resulted in a progressive reduction in CI. At 40, 50, and 60 wt% PPF, CI values decreased to 56.7%, 56.2%, and 57.7%, respectively. This decline in crystallinity at higher fiber contents is likely due to fiber agglomeration, reduced matrix continuity, and limited interfacial interaction between starch chains and fiber surfaces. Excessive fiber loading restricts the mobility of starch chains and suppresses effective retrogradation, leading to a more disordered composite structure (Kabir *et al.* 2012; Arruda *et al.* 2025).

The observed crystallinity trends are consistent with findings reported for other starch-based biocomposites. For instance, Lomelí-Ramírez *et al.* (2014) reported an increase in crystallinity with fiber incorporation in cassava starch reinforced with green coconut fiber, followed by structural heterogeneity at higher fiber contents. Importantly, the peak crystallinity observed at 30 wt% PPF in the present study correlates with changes in mechanical behaviour, including increased stiffness and reduced ductility. This relationship highlights the critical role of crystallinity in governing tensile and flexural properties, as increased molecular ordering generally enhances rigidity while limiting chain mobility (Ilyas *et al.* 2019; Kibet *et al.* 2025).

Overall, the XRD results demonstrate that fiber loading plays a significant role in controlling the crystalline organisation of TPCS/CW/PPF composites. Moderate fiber

incorporation promotes molecular ordering through synergistic starch–cellulose interactions, while excessive fiber content disrupts crystallinity due to agglomeration and restricted starch chain rearrangement. These findings further support the importance of optimizing fiber loading to achieve a balanced combination of structural order and mechanical performance in starch-based biocomposites.

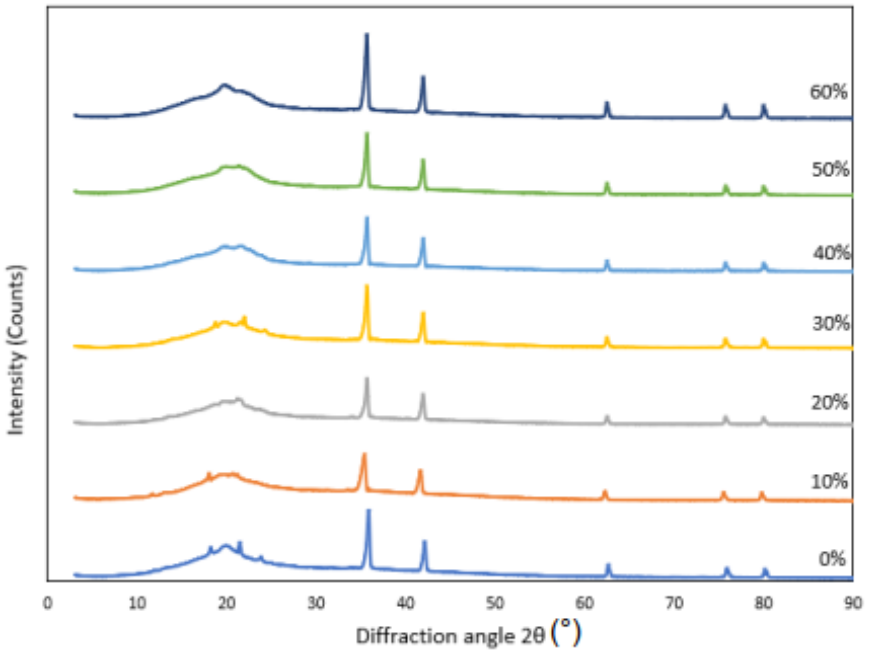

Fig. 4. X-ray diffraction patterns of TPCS/CW/PPF composites with fiber contents ranging from 0 to 60 wt%

Table 1. Crystallinity Index Values of TPCS/CW/PPF Composites

Samples	Crystallinity Index, CI (%)
TPCS	62.3
TPCS/PPF-10%	59.9
TPCS/PPF-20%	59.6
TPCS/PPF-30%	63.7
TPCS/PPF-40%	56.7
TPCS/PPF-50%	56.2
TPCS/PPF-60%	57.7

Thermogravimetric Analysis

The thermal degradation behaviour of TPCS/CW composites reinforced with PPF was investigated using thermogravimetric analysis (TGA) and derivative thermogravimetric (DTG) curves, as presented in Fig. 5 and summarised in Table 2. TGA provides critical insight into the thermal stability and degradation mechanisms of starch-based composites, which is essential for evaluating their processing window and potential service temperature range (Monteiro *et al.* 2012). Lignocellulosic fibres typically begin thermal degradation above 200 °C; therefore, processing at 150 °C does not induce thermal degradation but is sufficient for starch plasticization (Kabir *et al*. 2012; Monteiro *et al*. 2012).

All samples, including neat TPCS/CW and PPF-reinforced composites, exhibited three distinct stages of thermal degradation. The first stage occurred in the temperature range of approximately 30 to 180 °C and was attributed to the evaporation of physically absorbed moisture and low-molecular-weight volatiles. The moisture-related behaviour of the composites indicates that fiber incorporation influences water absorption due to the hydrophilic nature of starch and lignocellulosic components. This behaviour is consistent with the presence of hydroxyl groups in both starch and cellulose, which promote moisture uptake. This initial mass loss ranged between 3% and 8% and is characteristic of starch-based materials due to the presence of hydrophilic hydroxyl groups that readily bind water molecules (Prachayawarakorn *et al*. 2013). The slightly higher moisture loss observed in PPF-reinforced composites is associated with the hygroscopic nature of lignocellulosic fibers, which retain bound water within their amorphous regions.

The second and most prominent degradation stage occurred between 200 and 380 °C and corresponded to the thermal decomposition of the starch matrix and the polysaccharide components of the fiber, primarily hemicellulose and cellulose. In this region, the breakdown of glycosidic linkages and depolymerisation of starch chains resulted in rapid mass loss. Neat TPCS/CW exhibited an onset degradation temperature (T_{on}) of approximately 282 °C and a maximum degradation temperature (T_{max}) at around 336 °C. Upon incorporation of PPF, a slight reduction in both T_{on} and T_{max} was observed, with values shifting to the range of 268 to 276 °C and 308 to 324 °C, respectively. This downward shift can be attributed to the earlier degradation of hemicellulose present in PPF, which decomposes at lower temperatures than starch and cellulose (Kabir *et al*. 2012; Monteiro *et al*. 2012).

Despite the reduction in onset degradation temperature, fiber incorporation significantly altered the high-temperature degradation behaviour of the composites. The third degradation stage, extending beyond 380 °C up to approximately 500 °C, was associated with the gradual decomposition of lignin. Lignin degrades over a broad temperature range due to its complex aromatic structure, resulting in slower mass loss and the formation of thermally stable char (Monteiro *et al.* 2012; Ullah *et al.* 2025). As a result, fiber-reinforced composites exhibited substantially higher char residues compared to neat TPCS/CW. At 60 wt% PPF loading, the char yield reached approximately 42.9%, nearly three times higher than that of neat TPCS/CW (6.1%).

The increased char residue observed with increasing fiber content indicates improved thermal endurance at elevated temperatures. This behaviour is attributed to the higher lignin content and inorganic constituents present in *Pennisetum purpureum* fibers, which promote char formation and act as a protective barrier that slows down further thermal degradation (Lomelí-Ramírez *et al.* 2014; Edhirej *et al.* 2017). Similar trends have been reported for starch-based composites reinforced with natural fibers such as banana leaf, green coconut, and sayote fibers, where fiber addition led to increased char formation despite a slight reduction in onset degradation temperature (Lomelí-Ramírez *et al.* 2014; Antonio and Díaz 2025).

The DTG curves further support these observations, showing that the main degradation peak of neat TPCS/CW occurred at approximately 336 °C, while fiber-reinforced composites exhibited lower peak temperatures in the range of 318 to 324 °C. Although the maximum degradation rate shifted to lower temperatures with fiber incorporation, the overall degradation process became more gradual at higher temperatures due to the stabilising effect of lignin-rich fibers. This phenomenon highlights the dual role of PPF in the composite system: while the presence of hemicellulose slightly reduces

thermal stability at the initial decomposition stage, lignin contributes to enhanced resistance against complete thermal breakdown at elevated temperatures.

The variation in char residue at higher fiber loadings, particularly at 60 wt% PPF, may appear non-linear due to the heterogeneous distribution of natural fibers within the composite. Such behaviour is commonly observed in lignocellulosic systems, where localised differences in fiber dispersion, lignin content, and microstructural heterogeneity influence thermal degradation pathways. Therefore, the apparent irregularity does not indicate experimental inconsistency but reflects the inherent variability of natural fiber-reinforced composites.

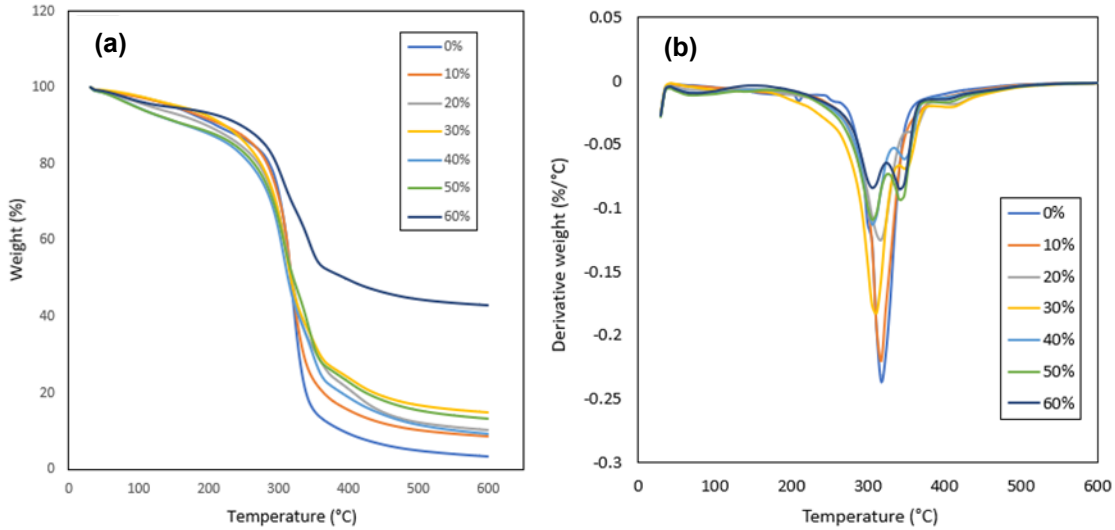

Fig. 5. Thermogravimetric analysis results of TPCS/CW/PPF composites with different fibre loadings (0, 10, 20, 30, 40, 50, and 60 wt%), showing (a) TG curves and (b) DTG curves

Table 2. Summary of TGA Parameters for TPCS/CW/PPF Composites

Samples	T_{on} (°C)	T_{max} (°C)	Weight loss at T_{max} (wt%)	Char at 600°C (wt%)
TPCS	278	318	50.7	3.4
TPCS/PPF-10%	273	318	44.6	9.7
TPCS/PPF-20%	271	318	42.3	12.5
TPCS/PPF-30%	265	312	41.2	15.1
TPCS/PPF-40%	262	306	39.5	18.8
TPCS/PPF-50%	259	306	39.2	20.3
TPCS/PPF-60%	259	342	39.5	42.9

Overall, the TGA results demonstrate that the incorporation of PPF modifies the thermal degradation behaviour of TPCS/CW composites. While fiber addition slightly lowers the onset of thermal degradation, it significantly enhances char formation and thermal resistance at higher temperatures.

Tensile Test

The influence of PPF loading on the tensile behaviour of TPCS/CW composites is illustrated in Fig. 6. Statistical analysis using one-way ANOVA confirmed that variations in fiber content resulted in significant differences in tensile properties ($P < 0.05$). Tensile

performance was evaluated in terms of tensile strength, tensile modulus, and elongation at break to assess the reinforcing efficiency of PPF within the starch-based matrix.

The neat TPCS matrix exhibited the lowest tensile strength, with a value of 4.85 MPa, reflecting the inherently weak load-bearing capability of plasticized starch. Upon incorporation of PPF, a progressive improvement in tensile strength was observed, indicating the effective reinforcing role of the fibers. The tensile strength increased steadily with fiber loading and reached a maximum value of 9.14 MPa at 50 wt% PPF, corresponding to an enhancement of approximately 88.3% relative to the neat matrix. This improvement can be primarily attributed to efficient stress transfer from the starch matrix to the fibers, facilitated by good interfacial adhesion. SEM observations of fractured tensile specimens revealed fiber fracture and partial fiber pull-out at moderate fiber loadings, confirming that the fibers actively participated in load bearing rather than debonding prematurely. The continuous improvement in tensile properties up to 50 wt% PPF indicates effective wetting of fibre surfaces by the thermoplastic starch matrix in its molten state, resulting in strong interfacial adhesion and efficient stress transfer. This behaviour is notable, as many natural fiber composites exhibit reduced performance at lower fiber loadings due to incomplete fiber wetting and agglomeration.

The enhancement in tensile strength is also closely related to the inherent chemical compatibility between starch and PPF, as per Fig. 6a. Both components contain abundant hydroxyl groups, which promote the formation of intermolecular hydrogen bonding at the fiber–matrix interface. Such interactions restrict polymer chain mobility and improve stress transfer efficiency, as reported in earlier biopolymer systems (Phan *et al.* 2009) and more recent reports of starch–cellulose and natural fiber composites (Asyraf *et al.* 2022; Jayarathna *et al.* 2022; Sabapathy *et al.* 2025). However, when the fiber content was increased to 60 wt%, a slight reduction in tensile strength to 8.61 MPa was observed. This decline suggests that excessive fiber loading exceeded the optimal reinforcement threshold, leading to fiber agglomeration and wetting of fibre surfaces by the matrix in its molten state that acted as stress concentration sites and weakened the composite structure (Fazeli *et al.* 2019; Ibrahim *et al.* 2020; Schutz *et al.* 2024).

A similar trend was evident in the tensile modulus results, as illustrated in Fig. 6b. The neat TPCS sample exhibited a relatively low modulus of 1.34 GPa, reflecting its flexible but mechanically weak nature. With increasing PPF content, the tensile modulus increased markedly, reaching a maximum value of 3.28 GPa at 50 wt% fiber loading, which represents an improvement of approximately 144%. This increase in stiffness is attributed to the rigid nature of lignocellulosic fibers and the strong intermolecular interactions between fiber cellulose and starch hydroxyl groups. These interactions restrict molecular motion within the matrix and enable more efficient stress transfer, thereby enhancing the stiffness of the composites (Chen *et al.* 2020; Sabapathy *et al.* 2025). Nevertheless, a slight decrease in modulus to 3.09 GPa was recorded at 60 wt% PPF. This behaviour is commonly observed in starch–fiber composites, where excessive fiber loading results in poor fiber dispersion and inadequate interfacial bonding, creating weak regions that limit stiffness enhancement (Prachayawarakorn *et al.* 2013; Kamaruddin *et al.* 2023). In contrast to strength and stiffness, the elongation at break of the TPCS/PPF composites remained relatively low across all compositions, indicating predominantly brittle tensile behaviour. The neat TPCS exhibited an elongation at break of 0.76%, while fiber incorporation led to a gradual increase, reaching 1.88% at 60 wt% PPF, as shown in Fig. 6c. This modest increase suggests that fiber addition slightly improved energy absorption and delayed crack propagation through mechanisms such as fiber pull-out and interfacial debonding.

However, the rigid and cellulose-rich nature of PPF significantly restricted the mobility of starch chains, thereby limiting the overall ductility of the composites. Similar tensile behaviour has been reported in other natural fiber-reinforced starch systems, where improvements in strength and stiffness are accompanied by relatively low elongation at break (Ilyas *et al.* 2019; Kamaruddin *et al.* 2023; Schutz *et al.* 2024).

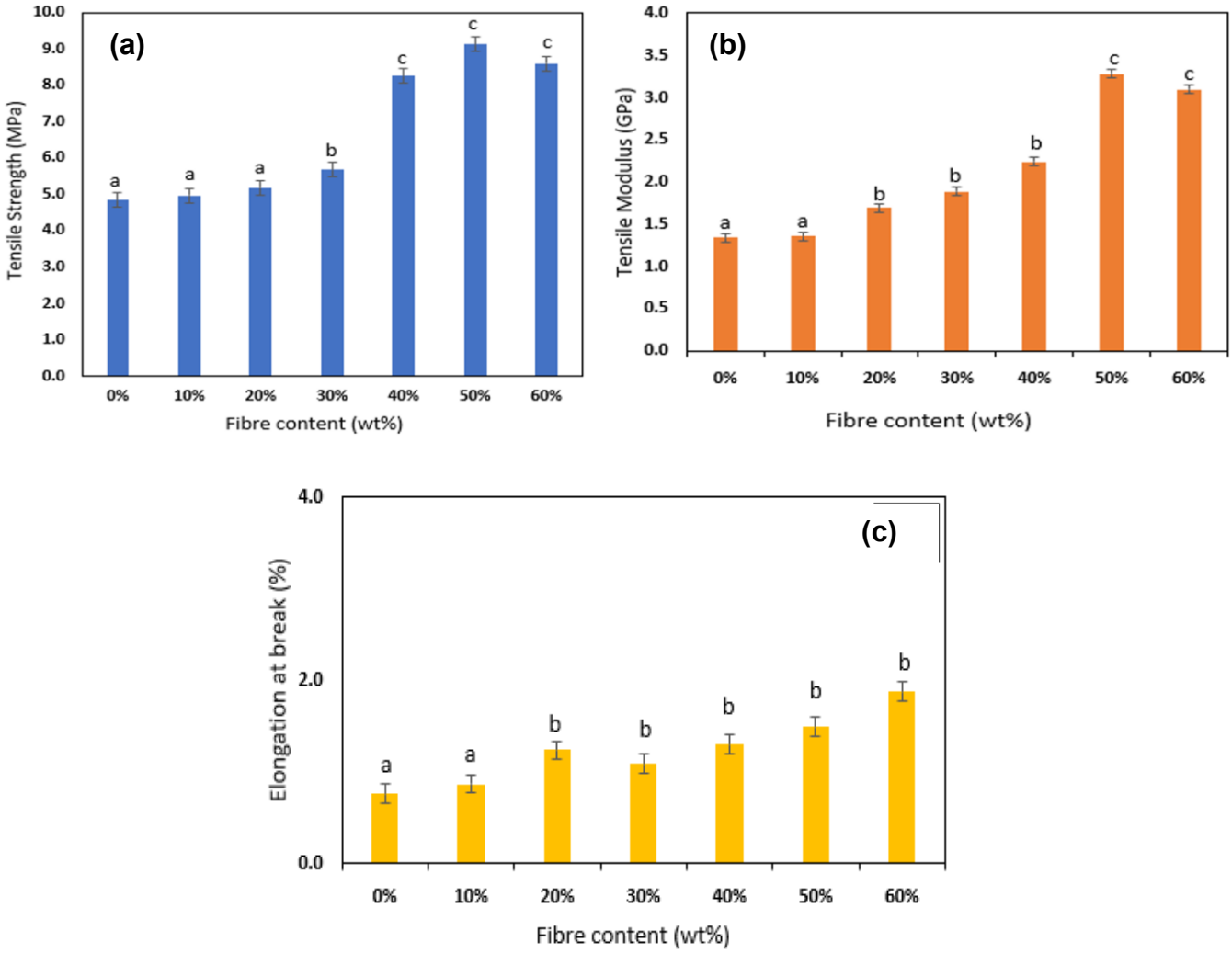

Fig. 6. Effect of PPF loading on (a) tensile strength, (b) tensile modulus, and (c) elongation at break of TPCS/CW/PPF composites

Overall, the tensile results demonstrate that PPF is an effective reinforcement for TPCS/CW composites, particularly at moderate fiber loadings. An optimum fiber content of 50 wt% was identified, where the composites achieved the best balance between tensile strength and stiffness. Beyond this level, further fiber addition compromised tensile performance due to agglomeration and insufficient fiber–matrix bonding. These findings are consistent with previously reported behaviour in lignocellulosic fiber-reinforced starch composites and emphasise the importance of optimising fiber loading to maximise tensile performance in fully bio-based composite systems (Prachayawarakorn *et al.* 2013; Fazeli *et al.* 2019; Ilyas *et al.* 2019; Kamaruddin *et al.* 2023; Schutz *et al.* 2024).

Flexural Test

The effect of PPF loading on the flexural performance of TPCS/CW composites is presented in Figs. 7(a-b), which shows the variation in flexural strength and flexural modulus as a function of fiber content. Statistical analysis using one-way ANOVA confirmed that fiber loading exerted a statistically significant influence on flexural behaviour ($P < 0.05$), highlighting the critical role of fiber reinforcement in governing the bending performance of the composites. The neat TPCS/CW matrix exhibited relatively low flexural strength and modulus, reflecting the limited load-bearing capacity and stiffness of plasticised starch under bending deformation. With the incorporation of PPF, a clear and progressive improvement in flexural performance was observed. The flexural strength increased steadily with fiber content, rising from 10.54 MPa for the neat matrix to a maximum value of 26.51 MPa at 50 wt% PPF, corresponding to an enhancement of approximately 151.5%. This pronounced improvement indicates that PPF effectively reinforces the starch matrix under flexural loading, where both tensile and compressive stresses act simultaneously across the specimen thickness.

A similar trend was observed for the flexural modulus. The modulus increased markedly from 0.29 GPa for neat TPCS/CW to 3.52 GPa at 50 wt% fiber loading, representing an improvement of more than one order of magnitude. The substantial increase in stiffness can be attributed to the rigid nature of lignocellulosic fibers and their ability to restrict deformation of the starch matrix during bending. Efficient stress transfer from the matrix to the fibers, combined with strong interfacial adhesion, enables the fibers to act as effective load-bearing elements, thereby enhancing resistance to flexural deformation. Comparable improvements in flexural properties have been widely reported for starch-based composites reinforced with natural fibers (Elsayed 2012; Ibrahim *et al.* 2020; Vignesh *et al.* 2021).

The enhanced flexural behaviour at intermediate fiber loadings is closely linked to favourable fiber–matrix interactions. Both starch and cellulose-rich PPF contain abundant hydroxyl groups, which promote hydrogen bonding at the interface. These intermolecular interactions improve fiber wetting and interfacial adhesion, allowing applied bending stresses to be transferred efficiently from the matrix to the reinforcing fibers. Similar mechanisms have been reported in other starch–cellulose systems, where improved chemical compatibility contributes to increased flexural resistance (Müller *et al.* 2009; Ilyas *et al.,* 2019).

However, when the fiber content was increased to 60 wt%, a slight reduction in flexural performance was observed. The flexural strength decreased to 24.0 MPa, while the flexural modulus declined marginally to 3.37 GPa. This reduction suggests that excessive fiber loading exceeded the optimal reinforcement threshold. At high fiber contents, fiber agglomeration, wetting of fibre surfaces by the matrix in its molten state, and reduced matrix continuity become more pronounced, leading to stress concentration sites that limit effective load transfer under bending (Radzi *et al.* 2019; Kamaruddin *et al.* 2023). Similar behaviour has been reported in other starch–fiber composite systems, where excessive fiber addition disrupts the structural integrity of the matrix and results in diminished flexural properties (Sanyang *et al.* 2016; Ayu *et al.* 2020; Kamaruddin *et al.* 2023).

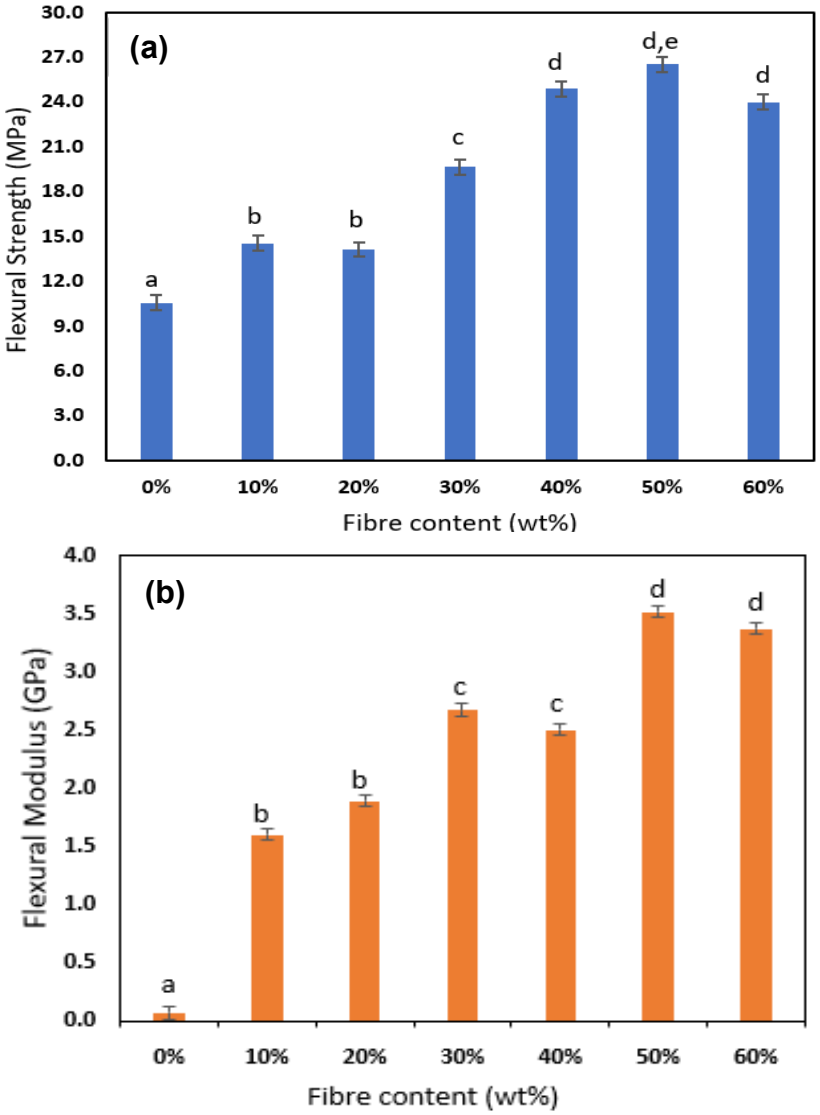

Fig. 7. Effect of PPF loading on (a) flexural strength, and (b) flexural modulus of TPCS/CW/PPF composite

Recent literature further supports the trends observed in the present study. Elfaleh *et al.* (2023) reported that polysaccharide-based composites reinforced with lignocellulosic fibers typically achieve maximum flexural strength at intermediate fiber loadings, while further fiber addition promotes agglomeration and reduces mechanical reliability. Likewise, Kibet *et al.* (2025) emphasised that optimal flexural performance in starch-based biocomposites is commonly achieved within the range of 30 wt% to 50 wt% fiber loadings, beyond which interfacial and dispersion limitations dominate. These observations are in good agreement with the current results, confirming that 50 wt% PPF represents the most effective reinforcement level for enhancing flexural properties in TPCS/CW composites. Overall, the flexural results demonstrate that the incorporation of PPF significantly improves both flexural strength and stiffness of TPCS composites up to an optimal fiber loading of 50 wt%. Beyond this level, excessive fiber content negatively affects matrix continuity and interfacial quality, leading to a slight deterioration in flexural performance.

These findings reinforce the importance of optimising fiber loading to achieve a balanced enhancement of bending strength and stiffness in fully bio-based starch composites.

Impact Test

The variation in impact toughness of TPCS/CW composites reinforced with PPF is shown in Fig. 8. The results demonstrate a clear dependence of impact performance on fiber loading, highlighting the role of PPF in governing the energy absorption behaviour of the composites under sudden loading conditions.

As the fiber content increased from 10 wt% to 50 wt%, the impact toughness rose progressively from 2.42 to 4.97 kJ m^{-2}. This continuous improvement indicates that fiber incorporation effectively enhanced the ability of the composites to absorb and dissipate impact energy. At low to moderate fiber contents, the presence of well-dispersed fibers introduced multiple energy-dissipation mechanisms during fracture, including fiber pull-out, fiber bridging, crack deflection, and interfacial debonding. These mechanisms delayed crack initiation and propagation, allowing greater energy absorption prior to catastrophic failure (Shah *et al.* 2013; López-Alba *et al.* 2018; Asim *et al.* 2020; Kamaruddin *et al.* 2023).

The progressive increase in impact toughness with fiber loading also suggests improved stress transfer efficiency between the starch matrix and the reinforcing fibers. Strong interfacial interactions, promoted by hydrogen bonding between hydroxyl groups in starch and cellulose-rich PPF, enabled effective load sharing and enhanced fracture resistance (Ilyas *et al.* 2019; Vignesh *et al.* 2021). At 50 wt% fiber content, these mechanisms appeared to be optimally balanced, resulting in the highest recorded impact toughness of 4.97 kJ m^{-2}.

However, when the fiber loading was further increased to 60 wt%, the impact toughness decreased slightly to 4.37 kJ m^{-2}. This reduction indicates that excessive fiber incorporation exceeded the optimal reinforcement threshold. At high fiber contents, fiber agglomeration and lack of complete wetting of fibre surfaces by the matrix in its molten state become more pronounced, leading to the formation of stress concentration regions that promote brittle fracture behaviour. In addition, the reduced matrix volume limits the capacity for plastic deformation, thereby diminishing the composite's ability to absorb impact energy effectively (Thomason and Rudeiros-Fernández 2018; Radzi *et al.* 2019; Kamaruddin *et al.* 2022).

The observed impact behaviour is consistent with trends reported in other natural fiber-reinforced starch composites. Several studies have shown that impact toughness typically increases with fiber loading up to an intermediate level, after which further fiber addition compromises toughness due to dispersion and interfacial limitations (Sanyang *et al.* 2016; Ayu *et al.* 2020). More recent work by Elfaleh *et al.* (2023) similarly demonstrated that polysaccharide-based composites reinforced with lignocellulosic fibers achieve maximum impact resistance at moderate fiber contents, while excessive fiber loading reduces mechanical reliability. Kibet *et al.* (2025) further highlighted that an optimal balance between strength and toughness in starch–fiber composites is commonly achieved within the 40 wt% to 50 wt% fiber range, which aligns well with the present findings.

Overall, the impact results confirm that PPF effectively enhances the impact resistance of TPCS/CW composites up to an optimal loading of 50 wt%. The improvement is primarily attributed to strong fiber–matrix adhesion and the activation of multiple energy-absorption mechanisms during fracture. Beyond this fiber content, the adverse

effects of fiber crowding and reduced matrix continuity outweigh the reinforcing benefits, leading to a slight decline in impact toughness.

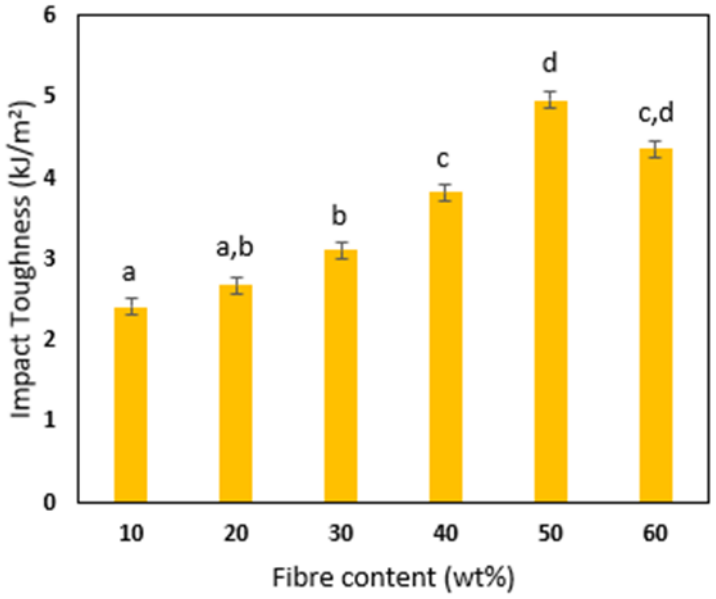

Fig. 8. Effect of PPF loading on the impact toughness of TPCS/CW/PPF composite

Table 3. Analysis of Variance Summary for TPCS/CW/PPF Composite

Variables	df	Tensile Strength	Tensile Modulus	Flexural Strength	Flexural Modulus	Elongation at Break	Impact Strength
Mixture	4	0.00*	0.00*	0.00*	0.00*	0.00*	0.00*

*Note: Differences are statistically significant at $P < 0.05$

CONCLUSIONS

1. Fourier transform infrared (FT-IR) analysis confirmed that the incorporation of *Pennisetum purpureum* fiber (PPF) into thermoplastic cassava starch/candelilla wax composites enhanced intermolecular hydrogen bonding without altering the chemical structure of the starch matrix. This indicates that fiber reinforcement primarily promotes physical interactions at the fiber-matrix interface rather than the formation of new chemical bonds.
2. Scanning electron microscope (SEM) observations revealed that composites with low to moderate PPF loadings exhibited good fiber dispersion and effective interfacial adhesion, as evidenced by fiber fracture and limited pull-out. In contrast, excessive fibre incorporation resulted in fiber agglomeration, void formation, and reduced matrix continuity, which negatively affected stress transfer efficiency.
3. X-ray diffraction (XRD) analysis demonstrated that fiber loading significantly influenced the crystalline organisation of the composites. Moderate PPF incorporation (30 wt%) promoted molecular ordering through synergistic interactions between starch and cellulose crystalline domains, resulting in a maximum crystallinity index of 63.7%. However, higher fiber loadings reduced crystallinity due to restricted chain mobility and increased structural heterogeneity.

4. Thermogravimetric analysis indicated that fiber addition slightly reduced the onset degradation temperature of the composites due to the presence of hemicellulose. Nevertheless, fiber incorporation significantly increased char residue, reaching up to 42.9% at 60 wt% PPF, indicating enhanced thermal resistance at elevated temperatures due to the lignin-rich structure of the fibers.
5. Mechanical testing demonstrated that PPF effectively enhanced the tensile, flexural, and impact properties of thermoplastic cassava starch/candelilla wax (TPCS/CW) composites. The optimum performance was achieved at 50 wt% PPF, with tensile strength, tensile modulus, flexural strength, and impact toughness reaching 9.14 MPa, 3.28 GPa, 26.51 MPa, and 4.97 kJ m^{-2}, respectively. Beyond this level, mechanical performance declined slightly due to fiber agglomeration and insufficient fiber–matrix wetting.
6. Overall, the results confirm that PPF is an effective and sustainable reinforcement for thermoplastic cassava starch/candelilla wax composites. The developed materials, composed entirely of renewable resources, exhibit improved mechanical performance, enhanced thermal resistance, and potential biodegradability. These characteristics highlight their suitability for applications such as biodegradable packaging, disposable products, and low-load structural components. The results also demonstrate that optimizing fiber loading is essential to achieving a balanced combination of structural integrity, durability, and environmental performance in starch-based biocomposites.

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