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Effect of Coconut Fiber Loading on the Morphological, Thermal, and Mechanical Properties of Coconut Fiber Reinforced Thermoplastic Starch/Beeswax Composites

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ABSTRACT

The increasing concern about global warming and the accumulation of non-biodegradable plastic has caused serious environmental issues. Hence, the need to create a more environmentally friendly material such as thermoplastic starch (TPS) has grown. However, the poor properties of TPS, such as high moisture sensitivity and low mechanical properties, have limited the potential application of this biopolymer. This study aims to modify TPS's thermal and mechanical properties by incorporating coconut fiber. The composites were prepared by incorporating various coconut fiber loading (0, 10, 20, 30, 40, and 50 wt.%) into the TPS matrix. The mixture was fabricated using a hot press at 145°C for 1 hour. The sample is then characterized using thermogravimetric analysis and tensile and flexural tests. The results show that the composite with 50 wt.% coconut fiber had higher thermal stability than samples with lower fiber content. A significant increment in tensile strength and modulus of up to 20.7 MPa and 2890 MPa were recorded for samples with 50 wt.% fiber content—the sample with 50wt.% fiber also demonstrated the highest flexural strength and modulus of up to 30.3 MPa and 3266.3 MPa, respectively. These changes are consistent with the FTIR

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and SEM findings, which show good compatibility of TPCS and coconut fiber with a homogeneous structure. Overall, coconut fiber shows good potential as reinforcement for biodegradable-based polymer composites.

Keywords: Coconut fiber, cellulose, mechanical, natural fiber, thermal, thermoplastic starch

INTRODUCTION

Increased awareness of environmental protection has led to vast amounts of studies conducted on biodegradable polymers. Biodegradable polymers are made of sub-components of plants, such as cellulose, protein, and starch. Among the biopolymers, thermoplastic starch (TPS) is one of the most promising materials due to the abundance of resources, low cost, and the fact that the raw material is renewable (Asyraf et al., 2022). However, TPS is often associated with several limitations, such as poor mechanical properties, which limits the widespread application of the end product (Thakur et al., 2019). Hence, several modifications on TPS have been conducted previously to improve the properties of TPS, such as blending with biopolymer or reinforcement with natural fiber (Saepoo et al., 2023; S. Wang et al., 2023).

Moreover, researchers have explored various techniques to overcome the inherent limitations of TPS and enhance its mechanical properties for broader industrial utilization. These efforts have led to significant advancements in the field of bioplastics. Blending TPS with biopolymers, such as starch or cellulose derivatives (Sanyang et al., 2016), has shown promising results in improved tensile strength, impact resistance, and thermal stability. This synergy between TPS and other biopolymers enhances the overall mechanical performance and contributes to the final product's biodegradability.

Another avenue of improvement involves reinforcing TPS with natural fibers or lignocellulosic fiber, such as sugar palm, kenaf, water hyacinth, jute, flax, or hemp. Table 1 shows the lignocellulosic fiber-reinforced starch biocomposites. Incorporating these fibers into the TPS matrix enhances its structural integrity and provides excellent strength-to-weight ratios (Bahloul et al., 2023; Khalili et al., 2023). It addresses the mechanical limitations and offers an environmentally friendly alternative to traditional petroleum-based plastics. The resulting composite materials exhibit enhanced properties, making them suitable for a wider range of applications, from packaging materials to automotive components.

Coconut fiber is a sustainable, renewable, biodegradable, and recyclable material, which makes it a suitable candidate for various applications. Adding coconut fiber to the polymer matrix has been reported to improve the mechanical properties of the composites, such as tensile strength and modulus of elasticity (Meráz-Rivera et al., 2020). Surface treatments, such as pretreating the surfaces of coconut fiber or removing wax, lignin, and hemicellulose from the fiber surface, can enhance the interaction between the fiber and the starch matrix,

Table 1
Lignocellulosic fiber-reinforced starch biocomposites

Starch source (plasticizer)	Polymer/reinforcement (natural fiber dimension)	Fabrication technique	References
Corn (glycerol and natural rubber latex)	Sisal, hemp (10–15 mm)	Melt blending	Gironès et al., 2012
Corn starch (glycerol)	Poly(vinyl alcohol) fiber	Compression molding	Zhou et al., 2020
Maize starch (glycerin)	Flax fiber	Extrusion	Borowski et al., 2020
Rice (glycerol)	Cotton fiber (2.11 and 5.27 mm)	Compression molding	Prachayawarakorn et al., 2011
Arrowroot starch (glycerol)	Arrowroot fiber	Solution casting	Tarique et al., 2022
Cassava (sorbitol and glycerol)	Cellulose nanocrystal from kenaf fiber (70–190 nm)	Solution casting	Zainuddin et al., 2013
Sugar palm starch (sorbitol and glycerol)	Nanocrystalline cellulose from sugar palm	Solution casting	Ilyas et al., 2018
Potato starch (glycerol)	Kaolin clay	Casting method	Kwaśniewska et al., 2020
Cassava starch	Cogon grass fiber	Compression molding	Jumaidin et al., 2020
Corn starch	Kenaf fiber	Casting method	Hazrol et al., 2022
Sugar palm starch	Tin oxide/ Oil Palm Empty Fruit Bunches (OPEFB) nanofibril cellulose	Casting method	Azra et al., 2023
Sugar palm starch (sorbitol and glycerol)	PLA/sugar palm nanocellulose	Compression molding	Nazrin et al., 2020; 2021
Cassava	Durian peel	Compression molding	Jumaidin et al., 2023
Sugar palm starch (sorbitol and glycerol)	Nanofibrillated cellulose from sugar palm	Solution casting	Ilyas et al., 2019, 2020
Pea starch (glycerol and water)	PCL/flax fiber	Compression molding	Fabunmi et al., 2011
Cassava starch (glycerol)	PLA/PBAT/jute fiber	Extrusion	Yokesahachart et al., 2021

leading to better mechanical properties (Gomes et al., 2019; Norfarhana et al., 2022; Norrrahim et al., 2021). Crosslinking agents, such as boric acid, can also affect the density, tensile strength, hardness, water absorption, and biodegradation of the composite (Nansu et al., 2019). Overall, coconut fiber shows promising properties as environmentally friendly reinforcement and biodegradable materials for various applications, including packaging and shockproof materials. Meanwhile, beeswax is a natural hydrophobic agent that can reduce natural materials' moisture sensitivity. Our previous work reported beeswax's effectiveness in reducing TPS's moisture sensitivity (Diyana et al., 2021).

Despite the effectiveness of beeswax and the potential of coconut fiber as reinforcement, to the best of our knowledge, no study was reported on coconut fiber as reinforcement for thermoplastic starch/beeswax composite. Hence, this study aims to develop a thermoplastic

cassava starch/beeswax blend reinforced with coconut fiber to investigate this bio-based material's thermal and mechanical properties.

MATERIALS AND METHODS

Materials

The colorless, odorless, viscous white powder cassava starch for TPS sample preparation obtained for this study was from Antik Sempurna Sdn. Bhd, Shah Alam, Malaysia. Glycerol was purchased from QReC (Asia) Sdn. Bhd., Rawang, Malaysia, with an AR grade of 99.5%. The Aldrich Chemistry Sdn. Bhd., Petaling Jaya, Malaysia, supplied beeswax, and the coconut fiber (CF) was collected from Sungai Petani, Kedah, Malaysia. The retting process was then carried out by soaking the husks for about 2 to 3 weeks. The materials of the husk, which bind together fiber, were degraded, and fibers were loosened during retting. The dried CF was ground to smaller sizes of about 2 to 3 mm.

Sample Preparation

TPS/Beeswax reinforced with coconut fiber was modified by merging different amounts of coconut fiber (0, 10, 20, 30, 40, and 50 wt.%), as shown in Table 2. After the compaction of the composites into the mold was completed, the composite underwent a heating process inside the hot press at 145°C for approximately 1 hour and a cooling process at 30°C for about 15 minutes. Figure 1 shows the sample preparation process, and Figure 2 shows the TPCS/beeswax reinforced with coconut fiber.

Figure 1. TPS/beeswax reinforced with coconut fiber

Figure 2. TPS/beeswax reinforced with coconut fiber

Table 2
Composite formulation

Composite	Modified TPS mixture (%)	Fiber Loading (wt.%)
0%	100	-
10%	90	10
20%	80	20
30%	70	30
40%	60	40
50%	50	50

Thermal Testing

Thermo-Gravimetric Analysis (TGA). TGA was performed using a Mettler-Toledo AG, Analytical (Schwerzenbach, Switzerland) at the Faculty of Mechanical and Manufacturing Engineering Universiti Teknikal Malaysia Melaka, UTeM. The specimen weight amounted to around 10 ± 2 mg. The study was performed in aluminum panels at 25 to 800°C at a fixed heating rate of 10°C min^{-1} in a complex nitrogen atmosphere.

Mechanical Testing

Tensile Testing. The tensile test samples were cut following the ASTM D638 standard. The tests were conducted on three replications using universal testing equipment (INSTRON556) with a 5kN and a crosshead speed of 5 mm/min. The air temperature of 23 ±1°C and relative humidity of 50 ± 5% were used in this study.

Flexural Testing. The flexural test for this study was performed at 23 ± 1°C and a relative humidity of 50 ± 5% according to ASTM D790. The dimension of 130 mm (L) × 13 mm (W) × 3 mm (T) was set for all samples. Crosshead speeds were fixed at 2 mm/min, and the tests were performed on three replications in Universal Test Machines (INSTRON 5556) with a 5kN load cell.

Fourier Transform Infrared Spectroscopy (FTIR)

FTIR was used to detect functional groups in coconut fiber and starch composite. Material spectra were obtained with an IR (Jasco FT/IR 6100) spectrometer. Dimensions of the samples were set to 10 mm (L) × 10 mm (W) × 3 mm (T). The sample FTIR spectrum was collected in a range of 4000 to 400 cm^{-1}.

Scanning Electron Microscope (SEM)

A scanning electron microscope (SEM), Zeiss Evo 18 model, and a 10 kV acceleration voltage were used to analyze the morphology of tensile fractured surfaces. The samples were coated with gold using an argon plasma metallizer (sputter coater K575X) (Edwards Limited, Crawley, United Kingdom) to avoid unwanted charging.

RESULTS AND DISCUSSION

Thermal Analysis

Figure 3 presents the Thermogravimetric Analysis (TGA) curve of thermoplastic cassava starch/beeswax reinforced with coconut fiber from 0 to 50 wt.% fiber, while Figure 4 shows the DTG curve of these materials. The starch began to degrade at approximately 250°C. According to Lomelí-Ramírez et al. (2011), although relatively similar and overlaid, the

TGA curves and composites of the TPS matrix exhibit some differences with the inset (fiber and starch) curves. Besides, there was a small and incremental mass loss of 25–200 °C in the cassava starch matrix and its components related to water losses and possibly glycerol (glycerol boiling point of 198°C). Before the TGA analysis, the composites, as well as the matrix, were dried up at 60°C, indicating a reduction in mass losses of matrix and composites due to most humidity already eliminated. Further dehydration of 25–200°C in this range was minimal. The DTG curve also confirmed no observed inflection point (peak) within this temperature range.

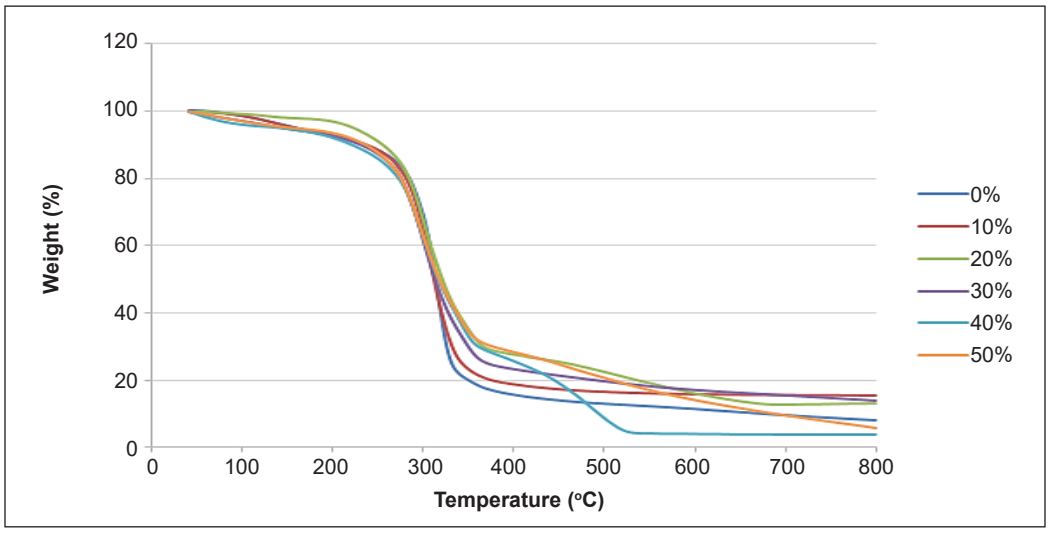


Figure 3. TGA curve of TPS/Beeswax + coconut fiber composite

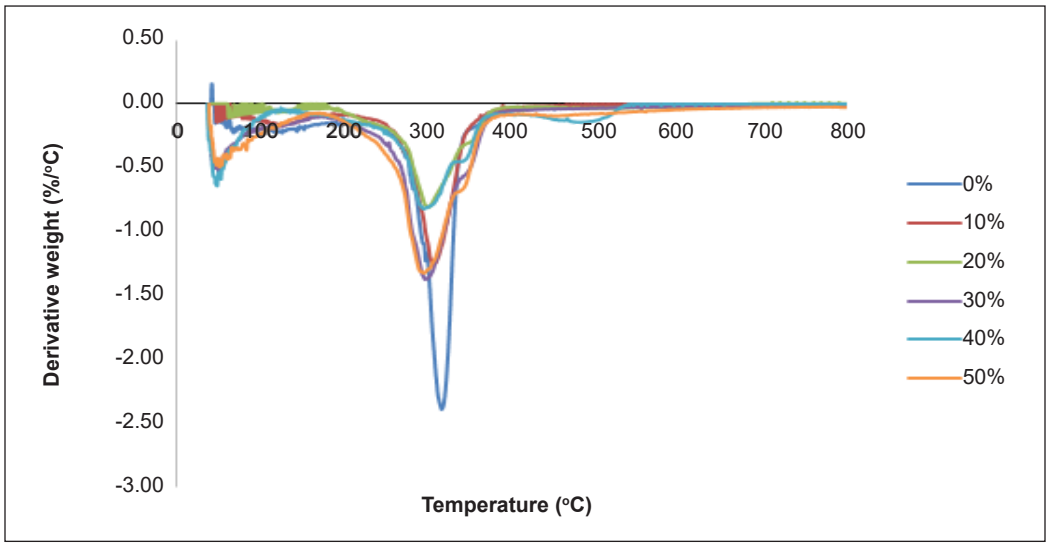


Figure 4. DTG curve of TPS/Beeswax reinforced with coconut fiber composite

This study parallels Gómez et al. (2006), who noted no mass loss associated with humidity and ascribed to the processing conditions. The dehydration mass losses observed for coir fibers and native starch in the 20 to 200℃ range were still greater. The fiber additions to the starch matrix have been reported to increase their thermal stability if the fiber and the matrix are well adhered to and thus reduce the mass loss in the sample (Ilyas et al., 2018; Ma et al., 2009). Figure 3 demonstrates that cassava starch degradation commenced after 300℃, while coconut fiber thermal degradation starts with hemicellulose degradation (200–260℃), lignin (280–500℃), and cellulose (240–350℃) (Ilyas, Sapuan, & Ishak, 2017; Ilyas, Sapuan, Ishak, & Zainudin, 2017; Lomelí-Ramírez et al., 2014).

Alemdar and Sain (2008) conducted a prior investigation wherein they examined the thermal stability of biocomposites derived from wheat straw nanofibres through TGA analysis—the TGA thermo grams with 5 wt.% nanofibers demonstrated starch degradation began at around 275℃ with thermoplastic starch (TPS) and nanocomposite. The temperature of degradation was about 296℃ for the nanofibers. Evaporation of glycerol contributed to the lesser weight losses of TPS and nanocomposites. Additionally, the results indicated that the temperature at which the polymer matrix and nanocomposites degrade was comparable to and less than that of each component.

In terms of fiber loading, adding coconut fiber improved the material's thermal stability. It can be seen through the higher residue content upon degradation for samples with coconut fiber as compared to the TPS matrix. The residue content was increased from 5.9% to 14.1% as the coconut fiber content increased from 0 to 30 wt.%. This finding can be attributed to higher carbonate content in the fiber than the TPS matrix (Kamaruddin et al., 2022).

Mechanical Testing

Tensile Properties. Table 3 presents the findings of analysis of variance (ANOVA) of the tensile testing. Since the P-value was less than 0.05, the average tensile strength and modulus varying from one level of polymer mixture to another is statistically important. Figure 5 displays the tensile and tensile modulus of starch composite and coconut fiber with 0 to 50 wt.% of coconut fiber obtained in the present study. From the data, we can observe a clear trend of increasing tensile strength as the loading percentage of coconut fiber increases. At 0% coconut fiber loading, the composite material exhibits a tensile strength of 5.42 MPa. As the percentage of coconut fiber loading increases to 10%, a notable improvement in tensile strength can be observed, which rises to 8.12 MPa. The trend continues with further increases

Table 3
Summary of the analysis of variance (ANOVA) of TPS/ Beeswax reinforced with coconut fiber

Variable	df	Strength	Modulus
Mixture	5	.000	.000

Note. *Significantly different at $p \leq 0.05$

in tensile strength as the coconut fiber loading increases to 20%, 30%, and 40%, resulting in a value of 9.26 MPa, 13.57 MPa, and 14.95 MPa, respectively.

Finally, at 50% coconut fiber loading, the composite material exhibits its highest tensile strength of 20.47 MPa. The phenomenon behind this trend lies in the reinforcing nature of the coconut fibers. Coconut fibers are known for their high strength and stiffness, which can enhance the mechanical properties of composite materials when incorporated as a reinforcement. The interfacial bonding between the coconut fibers and the TPS/Beeswax matrix also contributes to the improved tensile properties. Good bonding at the fiber-matrix interface allows efficient load transfer and minimizes stress concentration, enhancing tensile strength. These values demonstrated that the tensile strength and modulus were greater. Incorporating coconut fiber into the starch composite increased the tensile properties of the composite material.

According to Lomeli et al. (2014), these increments in Young's Modulus and Ultimate Tensile Strength may be associated with the strong fiber-matrix bonding, which resulted in increased adhesion between the fibers, thereby leading to a higher matrix-fiber stress transmission. Besides, an optimum fraction of the fiber volume is necessary to achieve better mechanical performance. In this study, the highest fiber content of 50 wt.% resulted in the highest strength. The increment in coconut fiber loading would increase the composite's tensile modulus. Figure 6 shows the tensile modulus increasing rapidly from 10 to 20 wt.% with increased fiber content and the tensile modulus values.

Since natural fiber is very hydrophilic, it helps to absorb moisture for a longer period using natural fiber composites (Dhakal et al., 2007). The increase in percentage length for both composites was lessened by increasing fiber content (treated and untreated) (Lai et al., 2005). The effect of matrix/composite treatment on the findings can be demonstrated

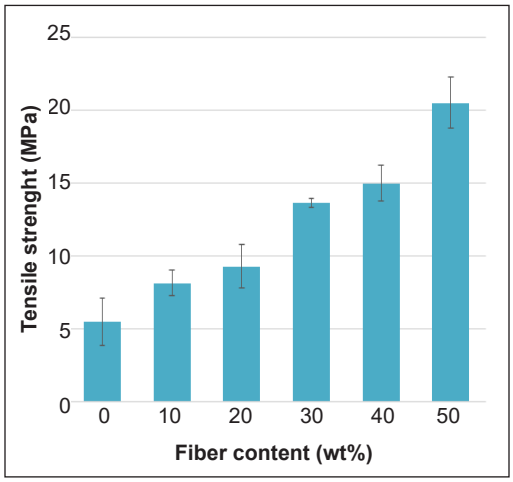


Figure 5. Tensile strength of TPS/Beeswax + coconut fiber

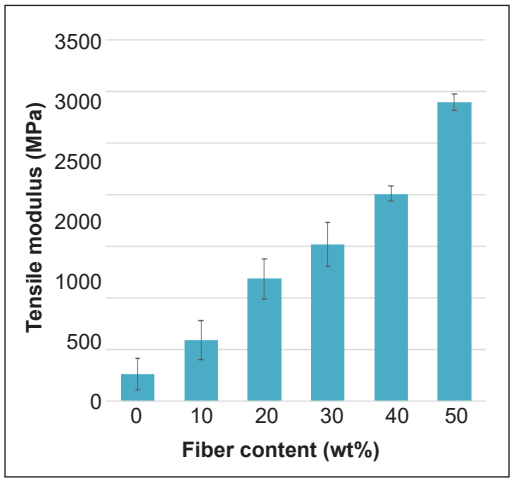


Figure 6. Tensile modulus of TPS/Beeswax + coconut fiber

based on a discrete molecular organization, which created a more ordered structure with less free energy (Lomelí-Ramírez et al., 2011).

According to Lai et al. (2005), with the incorporation of natural lignocellulose fibers into the polymer matrix, the strength of the composite may increase or decrease. Typically, lignocellulose fibers like coconut fiber can withstand the stress of polymer transfer as they are able to improve strength. However, the strength of the composites is impacted by some fibers that are inconsistent or irregularly created. The strength decreased as a result of the fiber's inability to withstand stress transferred from the polymer matrix.

Flexural Properties. Table 4 presents the summarized findings of the analysis of variance (ANOVA) of flexural testing. The finding shows that the statistically significant difference ($p<0.05$) between the mean flexural strength and modulus from one composite-to-one stage is statistically important. In general, with tensile testing, the effects of flexural testing meet a similar pattern. The flexural properties of a composite incorporated with coconut fiber were demonstrated by significant improvements ($p<0.05$). The obtained flexural strength data showed a consistent and significant improvement with increasing coconut fiber loading. At 0% coconut fiber loading, the composite exhibited a flexural strength of 2.49 MPa.

However, as the percentage of coconut fiber loading increased to 10%, the flexural strength improved significantly to 9.66 MPa. This initial increase in flexural strength can be attributed to the inherent mechanical properties of the coconut fibers, such as their high strength and stiffness (Khalil et al., 2010; Martinelli et al., 2023; Surnam & Imrith, 2023). As the loading percentage increased to 20%, a further enhancement in flexural strength to 11.65 MPa was observed. This improvement can be explained by the increased distribution and alignment of the coconut fibers within the matrix, resulting in improved load transfer and resistance to bending forces.

Furthermore, the interfacial bonding between the coconut fibers and the TPS/Beeswax matrix played a crucial role in enhancing the flexural strength. At 30% coconut fiber loading, a substantial jump in flexural strength to 18.16 MPa was observed. This increase can be attributed to the effective stress distribution and load-bearing capacity of the coconut fibers, along with the improved interfacial bonding. The progressive increase in flexural strength continued with 40% and 50% coconut fiber loading, resulting in values of 28.15 MPa and 30.25 MPa, respectively. The trend indicates that the higher the loading percentage of coconut fiber, the stronger the composite becomes in terms of flexural properties.

Figure 7 demonstrates that the introduction of fiber loading had an equivalent influence on the enhancement of flexural strength as it did on the improvement

Table 4
Summary of the analysis of variance (ANOVA) of TPS/ Beeswax reinforced with coconut fiber

Variable	df	Strength	Modulus
Mixture	5	.001	.000

Note. *Significantly different at $p\leq0.05$

of tensile strength, thereby augmenting the overall flexural strength property. Figure 8 shows the flexural modulus of TPS/Beeswax + coconut fiber. It was observed that the flexural strength and modulus were increasing gradually from 10 to 50 wt.%. The finding might be due to the polymer chain having a strong interaction with the fiber, which overcame the weak adhesive of the fiber matrix with increased fiber content. According to Lai et al. (2005), the higher aspect ratio might be why higher bending strength resulted from greater fibers. Other factors that influenced the mechanical strength of the composites include the fiber aspect ratio.

The previous study of mechanical properties of the TPS, TPS/jute fiber, and TPS/kapok fiber composites noted that adding jute or kapok fibers increased the flexural strength and Young's TPS modulus. The flexural strength and Young's TPS modulus were found to be significantly increasing by adding jute or kapok fibers. Besides, the increased content of the two cellulosic fibers significantly improved the flexural strength at full load and the TPS Young's modulus (Prachayawarakorn et al., 2013).

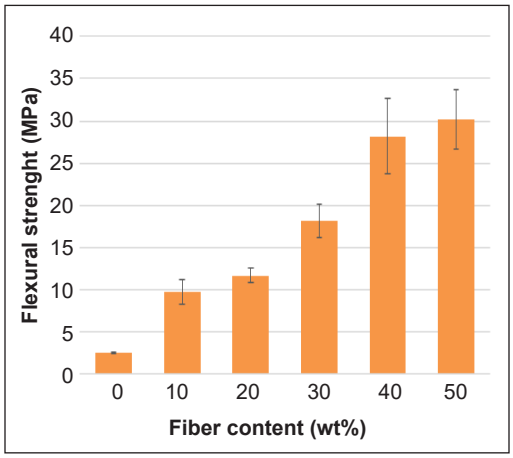


Figure 7. Flexural strength of TPS/Beeswax + coconut fiber

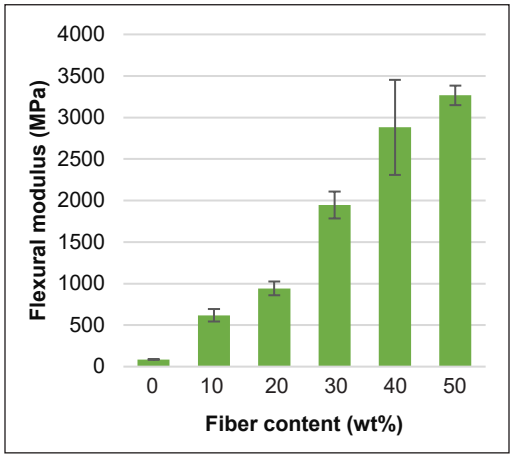


Figure 8. Flexural modulus of TPS/Beeswax + coconut fiber

Fourier Transform Infrared Spectroscopy (FTIR)

Figure 9 shows that the IR spectrums were identical for both the TPS and the composites, provided the main starch composite and coconut fiber were based on the structure of cellulose. In general, both the TPS and the composite spectrums showed the same band pattern. The chemical composition of starch remained unaltered during the plasticization process, regardless of the presence of glycerol or the quantity of fiber added. The observed relationship between these factors can be attributed solely to molecular interactions.

The peak in the 3400–3200 cm^{-1} range was attributed to the hydroxyl hydrogen-bonded group as a result of the free, inter, and intra-molecular bounded hydroxyl groups—axial

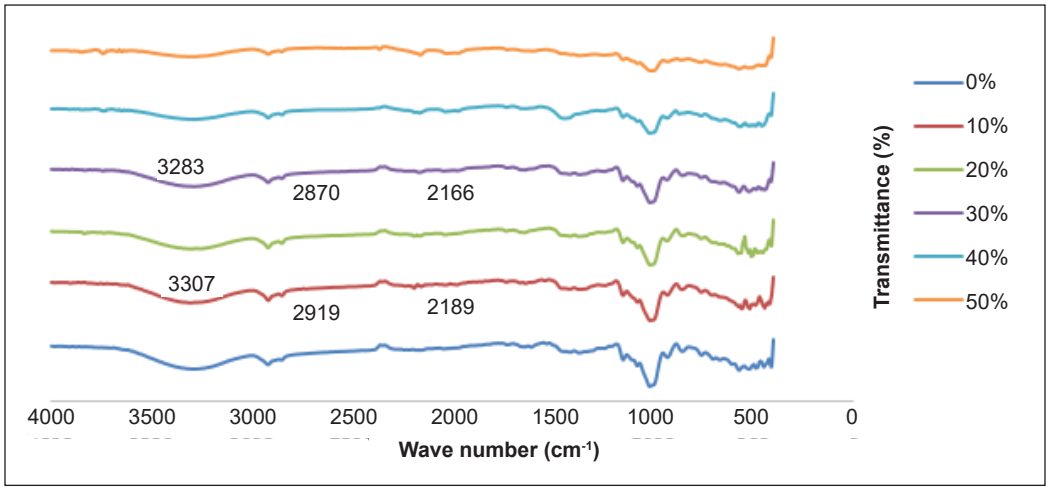


Figure 9. FT-IR spectra of the TPS/Beeswax reinforced with coconut fiber composites

deformations of the O–H group attributed to the large band. From 10 to 30% of coconut fiber, the wavenumber started to drop slowly from 3307 to 3283 cm^{-1}, respectively. The finding from Prachayawarakom et al. (2013) is strong evidence that new hydrogen relations were formed between the cellulosic fibers and the TPS matrix. The change in the IR peak position of O–H detention was also recorded and believed to occur between two compatibility polymers because of the hydrogen bonding. It was also reported that the plasticizing process may have been attributed to this improvement (Ma et al., 2005). It means that glycerol is affected by intra- and intermolecular hydrogen bonding networks between starch molecules.

The band of approximately 2936–2916 cm^{-1} is related to C–H aliphatic group vibrations of hydrocarbon starch, glycerol-starch (matrix), and TPS composites observed in all the spectra. Inversely, the effect of raising fiber content in composites is represented by this band (less defined band) (Lomelí-Ramírez et al., 2014). Tongdeesoontorn et al. (2011) reported that the TPS matrix sample and its biocomposites decreased this band. Besides, the band's intensity gradually changed (decreased) when the amount of fibers was added in certain thermoplastic samples. This finding indicates that starch is primarily responsible for this water belt, and thus, because of its low moisture content, coconut fibers have no detectable contribution.

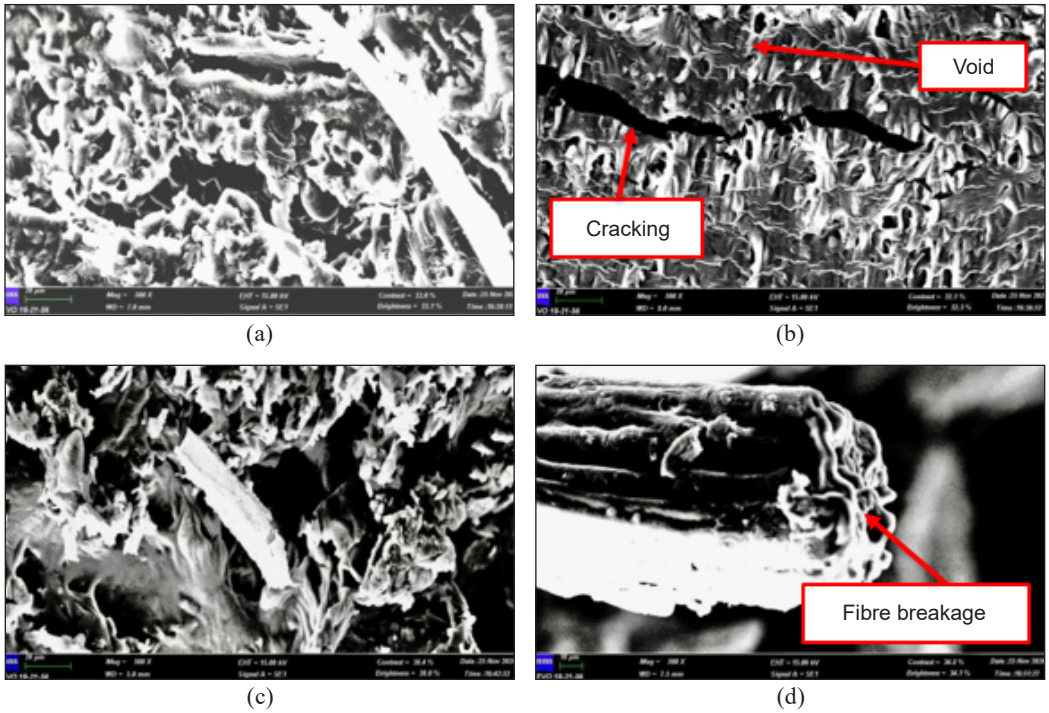
The other band is N–H stretching vibration at 2800–2000 cm^{-1}. According to F. C. Wang et al. (1994), for the results that attributed to N–H bonding and strength or variation on the integrated absorption coefficient with the change from the linked N–H band limit, there was a significant decrease in the integrated absorbing of the N–H band with increasing temperatures. These results were comparable to the N–H region. The decline in integrated absorption with increased temperature indicated the association of certain aromatic rings.

Scanning Electron Microscope (SEM)

Figure 10 shows the SEM micrograph of a fracture surface of TPS/beeswax reinforced with coconut fiber composites. The microstructures of the specimens were different due to the amount of fiber in the samples. Based on the results obtained, for coconut fiber content increasing from 10 to 50 wt.%, a cleavage structure was found on the surface of the fracture, which may be due to the polymer-polymer-bonding, which resisted loading deformations before being fractured (Jumaidin, 2016).

Additionally, Figure 10(b) shows that the cracks propagating through the brittle TPS matrix were avoided by the coconut fibers sticking out of the matrix. It facilitated premature failure and decreased the strength and rigidity of the composite of up to 20% of coconut fibers. The amount of coconut fibers was a strong barrier to crack propagation, and both tensile performances of the composites were improved for 50 wt.% composites.

According to Santafé Júnior et al. (2010), the amount of coconut fibers in composites of 40 wt.% is, by itself, was sufficient to withstand tensile loads and, thus, in contrast to the pure polyester matrix, increased both composite resistance and rigidity. Also, a similar finding was reported by Ma et al. (Ma et al., 2005) for thermoplastic processing of TPS by showing that the fiber in the granular starch fusion was affected by the increased fiber content. The fiber content was decreased during processing by high viscosity or inadequate dispersion.



(a) (b)

(c) (d)

Figure 10. SEM micrograph of a fracture surface of TPCS/Beeswax + coconut fiber composites: (a) 10% fiber; (b) 20% fiber; (c) 30% fiber; and (d) 50% fiber

CONCLUSION

Thermoplastic cassava starch/beeswax reinforced with coconut fiber has been developed using the hot press method. The thermal and mechanical properties of the composites were evaluated using TGA, tensile, and flexural tests, as well as FT-IR and SEM. TGA results demonstrated a major improvement in the composite's thermal stability by incorporating coconut fiber. The residue content was increased from 5.9% to 14.1% as the coconut fiber content increased from 0 to 30 wt.%. A significant increment in tensile strength and modulus of up to 20.7 MPa and 2890 MPa were recorded for samples with 50 wt.% fiber content. The sample with 50 wt.% fiber also demonstrated the highest flexural strength and modulus of up to 30.3 MPa and 3266.3 MPa, respectively. These changes were consistent with the findings of FTIR and SEM, proving the compatibility of TPS and coconut fiber with a homogeneous structure. Overall, coconut fiber has shown promising characteristics as a natural reinforcement material, which could benefit the composites manufacturing sector.

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