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Effect of Alkaline Treatment on Mechanical Properties of Pineapple Leaf Fiber-Reinforced Thermoplastic Cassava Starch Composite

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The widespread use of plastics has resulted in a significant accumulation of plastic waste in the environment. Hence, bio-based plastics have emerged as a promising alternative and are gaining commercial traction. Thermoplastic starch (TPS) is one of the materials used in the production of bioplastic films. Despite its environmental advantages, TPS has limitations, such as poor mechanical strength and high moisture absorption. Thus, this study aims to explore the potential of pineapple leaf fiber (PALF) as a reinforcing agent to improve the properties of thermoplastic cassava starch. PALF was treated with NaOH to enhance its characteristics. The physical and mechanical properties of both treated and untreated PALF were measured. The findings show that the treated TPCS/PALF composite with 8% PALF loading exhibited the highest tensile stress (1.14 MPa) and modulus (32.02 MPa) among the tested biocomposites. However, the inclusion of PALF at all filler levels resulted in a decrease in elongation at break. Physical tests also revealed a significant reduction in moisture content in both treated and untreated TPCS/PALF composites, although water absorption increased with higher fiber loadings. Fourier transform infrared spectroscopy (FTIR) analysis indicated notable changes in the physicochemical structure of the treated TPCS/PALF composites compared to the untreated ones.

Keywords: Pineapple leaf fiber; thermoplastic; cassava starch; mechanical properties

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Bioplastic nowadays has proven to be an environmentally friendly alternative to replace conventional plastic. Bioplastics are eco-friendly polymers derived from renewable polymer resources or biomass, such as starch, vegetable oils, fruit waste, lignin, cellulose and animal-derived components like proteins and lipids [1]. Starch-based bioplastics are the most promising because of their environmentally beneficial materials and abundance in nature. Significant studies have been conducted recently on starch-based bioplastics. Marichelvam et al. [2] carried out a study on the use of corn and rice starch-based bioplastics for packaging purposes. The findings indicate that the samples derived from corn and rice starch exhibit superior biodegradability compared to conventional plastic materials. Saiful et al. [3] conducted research on developing a high-performance bioplastic using wheat Janeng starch. Bioplastics that are produced by combining starch with glycerol provide transparent films that exhibit exceptional tensile strength and elongation properties.

Even starch-based bioplastics have promising characteristics, they do have significant drawbacks, such as high production costs and poor mechanical qualities due to high hydrophilicity and affinity to water that limit its application in packaging industries. These disadvantages of high manufacturing costs can be mitigated by employing agricultural wastes. Agro-waste products are often obtained from farming and cultivating activities which are commonly unwanted and are thrown. Thus, using agro-waste may make major contributions to material recovery, landfill reduction and lessen the environmental hazard upon their disposal. Among other types of agro waste, pineapple leaf fiber (PALF) has a relatively good potential due to its high cellulose content but lower hemicellulose. Since, PALF has high specific strength and stiffness and low microfibrillar angle, which is chief responsible factor attributing to increased tensile properties making it's a great candidates as a composite reinforcement [4] .This can be seen from a study conducted by Aji et al. [5] the PALF reinforced composite demonstrates superior mechanical strength. The tensile strength and Young's modulus of the composites rise proportionally with the increase in PALF (wt.%).

An important challenge in utilizing PALF as reinforcement in plastics is the poor adhesion between the natural fiber and the polymer matrix. This issue arises from the low interaction between the fiber and matrix, load transfer from matrix into fiber is not good. To address this, it is essential to modify the fiber's surface to reduce its polarity, thereby enhancing compatibility in the interface region. One

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effective modification method involves alkali treatment using a 5% sodium hydroxide (NaOH) solution, which removes contaminants and improves the fiber's surface characteristics [6]. Therefore, this study investigated the potential of PALF to serve as reinforcement filler in thermoplastic cassava starch. The physical and mechanical characteristics of the PALF, such as moisture content, water absorption and tensile properties were measured to understand the properties of the fiber. The capability of both untreated and treated PALF to disperse in thermoplastic cassava starch were also investigated. Thus, it is worthwhile to examine the effect at various filler loadings on the structural and the mechanical properties of thermoplastic cassava starch. This research not only aims to enhance the performance of bioplastics but also contributes to the sustainable use of agricultural waste, paving the way for innovative applications in environmentally friendly packaging solutions. By reinforcing thermoplastic cassava starch with PALF residues not only adds value to this agricultural byproduct but also enhances the viability of starch-based composites as sustainable materials.

EXPERIMENTAL

Chemicals and Materials

The pineapple leaf fiber was collected from FGV Chuping Agro Valley, located in Chuping, Perlis while cassava starch was obtained from local market area Perlis. Glycerol was purchased from Sigma Aldrich. **Table 1** shows the composition of the thermoplastic cassava starch composites with varying fiber loading.

Characterization Methods

Moisture Content (MC)

The moisture content test was carried out using the ASTM D2216 method. The moisture content of a material is the quantity of water that can be eliminated from the material without altering its chemical makeup, expressed as a ratio to the initial weight of the material. A film with a known weight was kept in an oven at 105 °C for 24 h. The MC for each sample was calculated using the weight differences before (M1) and after (M2) dehydration. The test was performed triplicates, and each sample was expressed as a percentage using the Equation (1).

$$\mathrm{MC}(\%) = \left(\frac{\mathrm{M1}-\mathrm{M2}}{\mathrm{M1}}\right) \times 100 \quad (1)$$

Where:

M1 = Weight of the sample before dehydration

M2 = Weight of the sample after dehydration

Water Absorption (WA)

The water uptake test was carried out using the ASTM D 570-98 method. A film sample of 15 mm × 15 mm was dried in an oven for 3 h at 105 °C. After that, the sample was allowed to cool and promptly weighed (Mi). After drying, the sample was submerged in 100 ml of distilled water at ambient temperature. Then, the sample was removed from the water, gently dried with a smooth cloth, and weighed (Mf) after a specific immersing period. The assessment of WA was obtained by calculating the mass differences between the dried and saturated conditions of each sample. The results were determined using the Equation (2).

$$\mathrm{WA}(\%) = \left(\frac{\mathrm{Mf}-\mathrm{Mi}}{\mathrm{Mi}}\right) \times 100 \quad (2)$$

Where:

Mi = Weight of the sample before immersion

Mf = Weight of the sample after immersion

Table 1. The composition of the thermoplastic cassava starch composites with varying fiber loading.

Film	Glycerol (%) of dry starch	Cassava starch (CS) g/100 ml distilled water	PALF (%) of dry starch
CS-film	25	5	0
TPCS-UPALF 2%	25	5	2
TPCS-UPALF 4%	25	5	4
TPCS-UPALF 6%	25	5	6
TPCS-UPALF 8%	25	5	8
TPCS-TPALF 2%	25	5	2
TPCS/TPALF 4%	25	5	4
TPCS-TPALF 6%	25	5	6
TPCS-TPALF 8%	25	5	8

Fourier Transform Infrared Spectroscopy (FTIR)

The chemical composition of the fibers was analysed using Attenuated total reflectance Fourier transform (ATR-FTIR) both before and after the surface treatment. A Spotlight 400 Perkin Elmer spectrometer (Waltham, MA, USA) was used to record the FTIR spectra of both the PALF and the treated PALF within a range of 4000–400 cm^{-1}. The scanning was done 10 scans per sample with a resolution of 4 cm^{-1}.

Tensile Properties

The tensile properties are determined using Instron universal testing machine based on the suggestions provided by ASTM D882 at ambient temperature, which are equipped with a 5kN load cell [7]. A film strip measuring 70 mm × 10 mm was placed between the clamps of a tensile machine. It was then pulled at a crosshead speed of 2 mm/min, while maintaining a grip separating 30 mm. The tensile stress, tensile modulus and tensile strain at break was determined by averaging the data of five repetitions for each specimen.

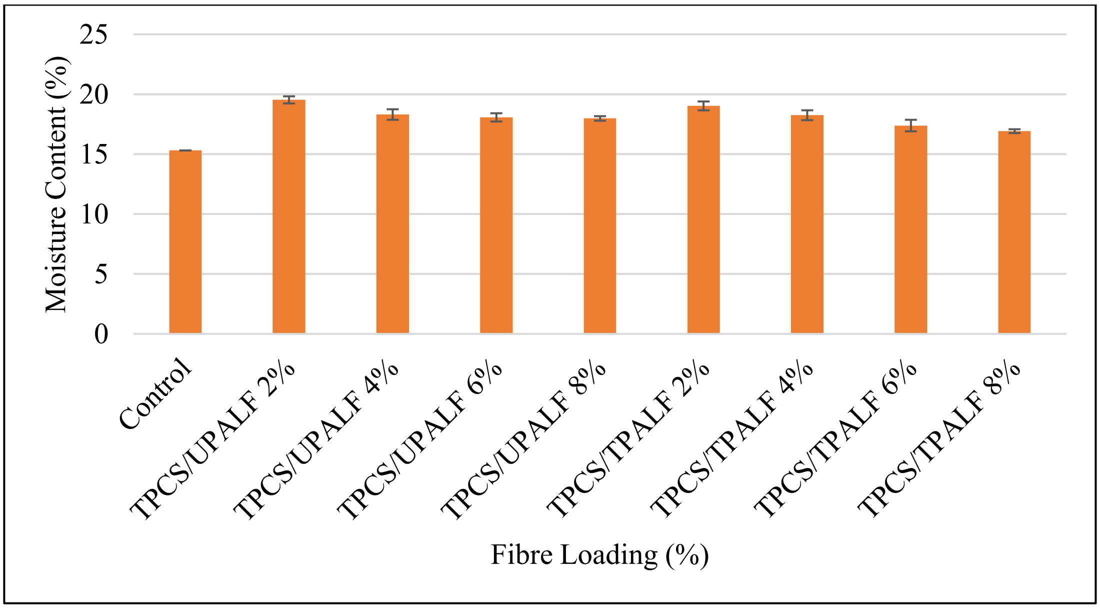

Figure 1. Moisture content of control, TPCS/UPALF and TPCS/TPALF composite films.

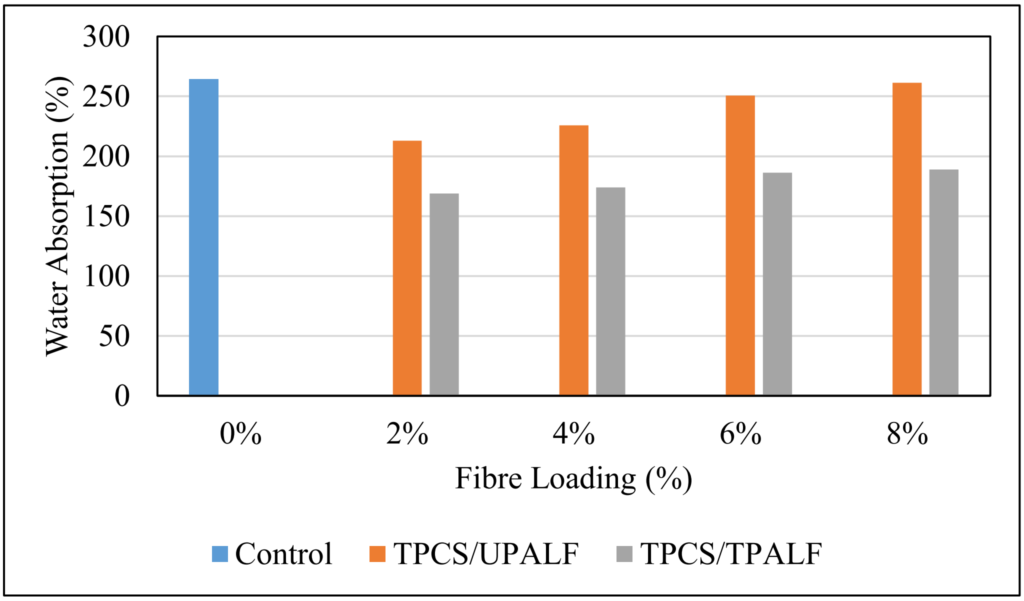

Figure 2. Water absorption of control, TPCS/UPALF and TPCS/TPALF composite films for 180 min time immersion.

RESULTS AND DISCUSSION

Moisture Content (MC)

Moisture content is an important factor to consider when choosing natural fibers as reinforcement materials to produce new composite materials. A low water content is necessary to prevent any negative impact on the dimensional stability of the composite materials, especially in terms of mechanical performance, creation of porosity, and water holding capacity [8]. **Figure 1** shows moisture content of control, untreated and treated TPCS/PALF composite films.

The results indicate that the control film has the lowest moisture content, as it lacks hydrophilic fibers that absorb water due to their hydroxyl groups [9]. In contrast, films with fibers show higher moisture content, with untreated TPCS/UPALF absorbing more water than treated TPCS/TPALF. This difference is due to alkali treatment, which reduces hydroxyl groups on the fibers, lowering their water absorption capacity [10]. The reduced moisture content in treated composites helps improve stability and prevent premature failure. Similarly, Vijay et al. [11] observed a decrease in moisture content for NaOH-treated Pennisetum oriental grass fibers compared to untreated fibers, attributed to reduced hydrophilic groups. These findings confirm that adding and treating PALF significantly influences the moisture properties of TPCS films.

Water Absorption (WA)

WA tests were performed for both treated and untreated TPCS/PALF biocomposites. The composites' water absorption increases as the immersion period increases. The impact of immersion time was restricted to 180 min as the film samples started to disintegrate beyond this duration. **Figure 2** shows water absorption of untreated and treated TPCS/PALF composite films at 180 min immersion time.

The control film shows the highest water absorption rates, with values of 264.59% at 180 minutes. By comparing between treated and untreated PALF composites, the treated samples absorb less water, showing that the treatment helps reduce water absorption. Alkaline treatment reduces the hydrophilicity of natural fibers and eliminates waxes and contaminants from their surface, hence improving the fiber-matrix adhesion in composites [12]. Among the untreated composites, TPCS/UPALF 8% had the highest water absorption (261.36%), followed by TPCS/UPALF 6% (250.67%), TPCS/UPALF 4% (225.89%) and TPCS/UPALF 2% (213.07%). For treated composites, TPCS/TPALF 8% absorbed the most water (189.02%), with TPCS/TPALF 6%, 4%, and 2% showing absorption rates of 186.32%, 173.89%, and 168.87%, respectively. Overall, all TPCS/PALF composites demonstrated high water absorption, exceeding 100%, consistent with findings by Tarique et al. [13].

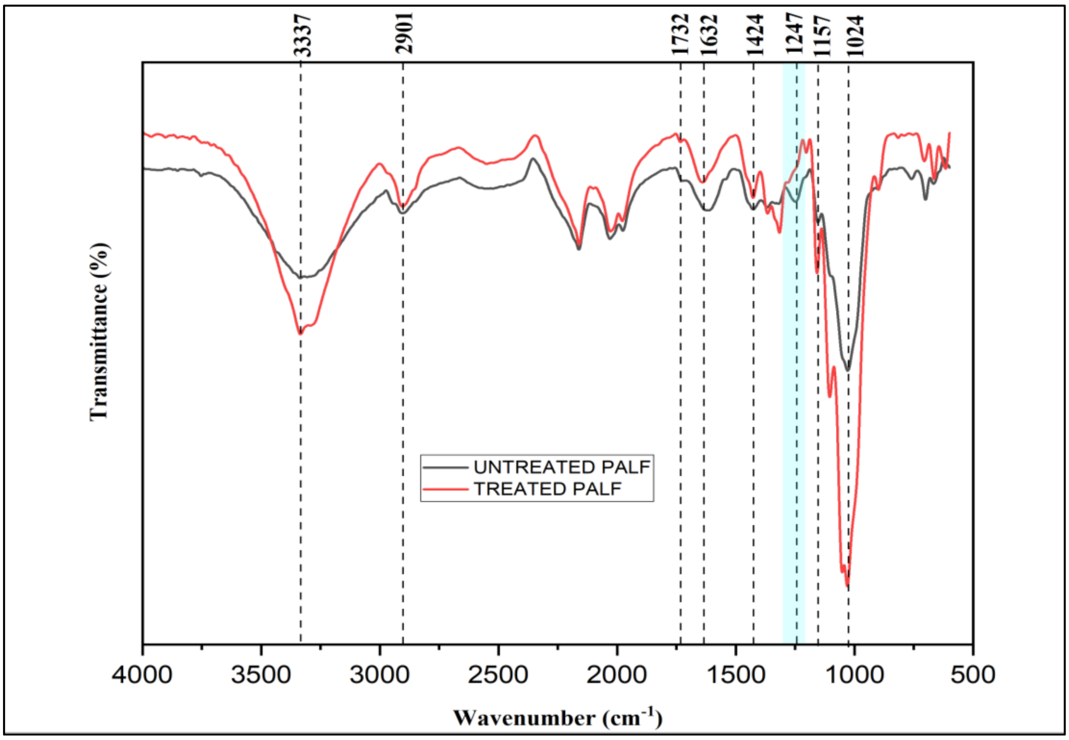

Figure 3. FTIR analysis of untreated and NaOH treated PALF.

Fourier Transform Infrared Spectroscopy (FTIR) for Untreated and Treated PALF

FTIR spectroscopy was used to examine the chemical structure of untreated and treated PALF fibers, aiming to identify changes caused by alkali treatment. These changes are expected to improve the adhesion between the fiber components. **Figure 3** shows the FTIR spectra of untreated and treated PALF.

The FTIR spectra of both treated and untreated PALF show absorption peaks around 3337 cm⁻¹, corresponding to the stretching vibrations of OH groups. A smaller peak around 2901 cm⁻¹ is attributed to CH bond stretching in cellulose and hemicellulose. The peak at 1424 cm⁻¹, with higher intensity, represents CH deformation in the cellulose structure. Both treated and untreated PALF show an absorption peak at 1732 cm⁻¹, associated with the stretching vibrations of C=O in hemicellulose. A similar peak at 1747 cm⁻¹ was observed in Miswak composites in a study by Rafiqah et al.[14], attributed to C=O bond stretching in carbonyl groups. The prominent peak at 1632 cm⁻¹, indicating carboxyl groups in hemicelluloses, is present in both untreated and treated PALF. Peaks at 1157 cm⁻¹ and 1024 cm⁻¹ represent the C-O-C stretching of β-1,4 glycosidic bonds in cellulose and C-H and C=O bonds in cellulose, respectively.

Following treatment, several FTIR bands weakened due to the removal of hemicellulose and lignin. The intensity of the OH peak at 3337 cm⁻¹ increased in treated PALF, likely due to the exposure of OH-rich fibrils after the removal of hemicellulose and lignin [15]. Similarly, the absorption peak at 1024 cm⁻¹, associated with C-H and C=O bonds in cellulose, showed increased intensity in treated PALF, reflecting the removal of non-cellulosic materials bound to cellulose [16]. In contrast, the peak at 1247 cm⁻¹, related to C-O stretching in hemicelluloses (xylan), appeared in the untreated PALF spectrum but was absent in the treated PALF, confirming the successful removal of hemicellulose, extractives, and lignin [17]. These FTIR results confirm that alkali treatment effectively alters the chemical composition, particularly reducing hemicellulose, lignin, and cellulose content. **Figure 4** shows the characteristic peaks of the composites, with most of them reflecting the control sample after adding 8 wt.% of UPALF and TPALF to the TPCS composite system.

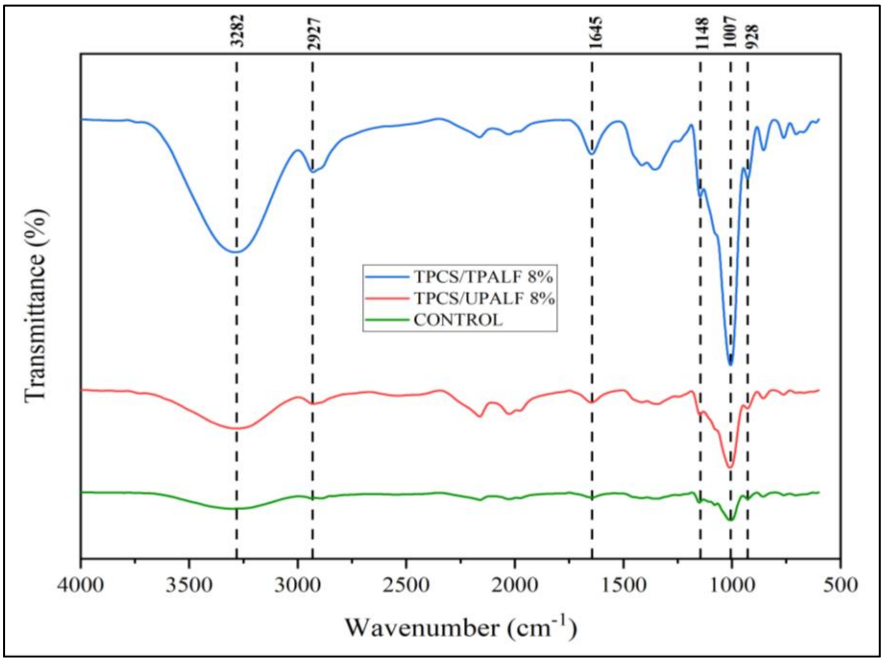

Figure 4. Comparison of FTIR analysis between control, untreated and treated TPCS/PALF 8% composites film.

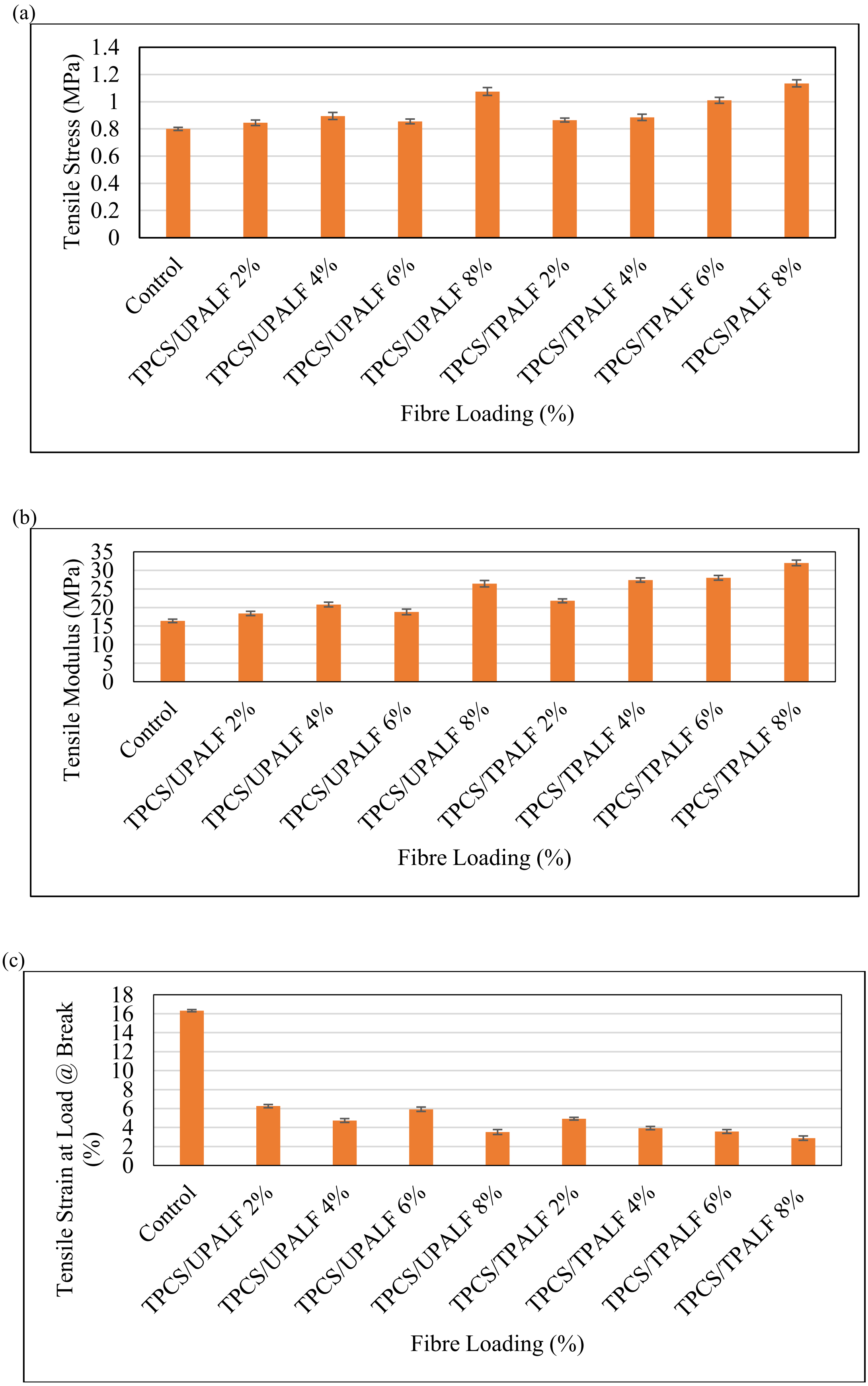

Figure 5. Tensile properties of the control and the thermoplastic cassava starch/PALF bioplastic at 2, 4, 6, and 8 wt % of untreated and treated: (a) tensile strength, (b) tensile modulus, and (c) elongation at break.

A peak at 928 cm^{-1} corresponds to the stretching of the C-O bond in the glycosidic ring of starch and lignin, specifically in the C-O-C and C-O-H groups. Sharp peaks at 1007 cm^{-1} and 1148 cm^{-1} result from the vibrational stretching of the C-O-H group, indicating changes in the starch crystal structure and the interaction of C-C and C-O groups. The peak at 1645 cm^{-1} in the second region for both untreated and treated composites is attributed to the bending mode of water molecules within the starch. The strong peak at 2927 cm^{-1} is associated with C-H bond stretching vibrations, while the peak at 3282 cm^{-1} corresponds to the elongation and oscillation of O-H groups in both the matrix and reinforcement, suggesting the composite films' affinity for water due to hydroxyl groups [7]. The shifts in band positions after fiber loading indicate interactions between the matrix and reinforcement. However, the untreated and treated PALF show similar characteristics, in line with findings from Mohammed et al. [9].

Tensile Properties

Figure 5 shows the effects of fiber loading on the mechanical properties of TPCS/PALF composites, specifically tensile strength, modulus, and elongation at break. The results show that treated composites (TPCS/UPALF and TPCS/TPALF) exhibit a significant improvement in tensile strength due to better adhesion between the cassava starch matrix and treated PALF fibers. For TPCS/PALF composites, tensile stress increases as fiber content rises, with the highest tensile stress (1.08 MPa for UPALF and 1.14 MPa for TPALF) achieved at 8% fiber loading. The treated composites demonstrate up to 42% enhancement in tensile strength compared to the control sample, which had a tensile stress of 0.80 MPa. This improvement is attributed to better fiber-matrix bonding, allowing effective stress transfer during loading. Similar results were observed in a study by Zubairi et al. [18], which also found that increasing PALF fiber content in PLA composites enhances tensile strength. An important factor determining the tensile strength is the efficacy of the bonding and stress transfer between the matrix and the fibre. When a load is applied to a fiber-reinforced composite, the load is carried from the matrix along the fibers, resulting in a uniform and effective distribution of stress. The tensile strength of the composites is significantly improved by the active participation of the fibers in stress transfer activity at the optimal fiber loading. This is the reason for the excellent mechanical strength of 8 wt.% TPCS/UPALF and TPCS/TPALF composites.

Figure 5(b) shows the tensile modulus for TPCS/UPALF increased between 18.40 MPa at 2% and 20.80 MPa at 4%. However, it declined to 18.81 MPa at 6% and then experienced a substantial increase to 26.42 MPa at 8% fiber loading. In contrast, the tensile modulus for TPCS/TPALF varied from 21.80 MPa at 2% to 27.80 MPa at 4%. Subsequently, it increased to 28.0 MPa at a concentration of 6% and experienced a substantial increase to 32.02 MPa at 8 wt.% of fiber loading. The control film has a tensile modulus of 16.40 MPa, which is significantly lower than that of both untreated and treated composites. Specifically, the untreated TPCS/PALF composites exhibited an increase in tensile modulus up to 61% while the treated TPCS/PALF composites demonstrated an increase of 95% at 8% filler. This increase may be attributed to the improvement in the stiffness and brittleness of the matrix. The adhesion at the interface between the fibers and the matrix limited the movement of polymer chains when subjected to reduced strain, allowing the transfer of stress from the matrix to the fibers [18].

Figure 5(c) depicted the tensile strain at load at break for the TPCS/UPALF composites showed variation, with a decrease from 6.26% at a 2 wt.% to 4.74 % at a 4 wt.% of fiber loading. Conversely, in the case of TPCS/TPALF, the tensile strain reduced from 4.93% at 2% to 3.94% at 4wt.%. The control film exhibits a much higher tensile strain of 16.33%. This indicates the presence of natural fibers may have restricted the mobility of the polymer chains inside the matrix. As a result, increasing the fiber content replaced the ratio of elasticity matrix phases, making the composites stiffer [18].

CONCLUSION

In this study, bioplastic films made from cassava starch and pineapple leaf fiber (PALF) were successfully prepared through solution casting. FTIR analysis confirmed the removal of lignin and hemicellulose in NaOH-treated PALF, as indicated by the absence of the absorption peak at 1247 cm^{-1}. This contributed to reduced water absorption in treated samples. Mechanical tests indicated that tensile stress and modulus increased with increased filler loading, with treated samples exhibiting a 42% enhancement in tensile stress and a 95% enhancement in modulus relative to the control. The bioplastic with 8% filler demonstrated superior mechanical performance. The integration of PALF as a filler improves the durability of TPCS films and presents a viable alternative to petroleum-based polymers.

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