

Effect of Alkaline treatment on the characteristics of pineapple leaves fibre and PALF/PP biocomposite

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ABSTRACT – Pineapple leaves fibre (PALF) is one of the natural fibre that has high potential to substitute non-renewable synthetic fibre in thermoplastic products. The PALF were alkali treated with different concentrations of NaOH. Untreated and alkali treated PALF were characterized using Thermal Gravimetric Analysis (TGA) and Scanning Electron Microscopy (SEM) to determine the thermal stability and surface morphology of the fibres respectively. Biocomposites were prepared by reinforced alkali treated and untreated PALF with polypropylene (PP) matrix. Tensile properties and water absorption analysis of PALF/PP biocomposites were studied. Biocomposite with 8 wt.% of alkali treated PALF express excellent thermal stability, with maximum degradation temperature at 270 °C which is a 7.17% improvement compared to untreated PALF. This biocomposite also had increased tensile strength (116 MPa) with 43% improvement compared to untreated PALF/PP (66 MPa) biocomposite and had lower water absorption at 6% compared to untreated biocomposite which at 21%. Hence, alkali treated PALF is able to improve the characteristic of PALF and increase the compatibility between fibre and polymer by reducing hemicellulose and lignin components.

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INTRODUCTION

Biomass has a great future as a source of renewable material compare to other resources. The benefit of biomass as a sustainable and abundantly available raw material could help to keep an ecological balance on the environment by utilizing the wastes to be used as a raw material for some value-added products [1]. Natural fibres show many advantages such as renewability, biodegradability, CO₂ neutrality, nontoxicity, wide availability, low cost, low density, low energy consumption during fabrication, and high specific strength compared to synthetic fibres. The main polymer in biomass is cellulose that has semicrystalline with long linear glucose polymer linked by β-1,4 glycosidic bonds [2, 3]. These brand them competitive to glass fibres as a reinforcing agent in applications of composite. Recently, research on biocomposites from natural fibre has gained researcher interest. Studies show that pineapples leaves fibre (PALF) has high specific properties and low-cost fibres which makes it suitable in the production of biocomposites compared with other agro-based fibre. The cellulose content in PALF is higher than that of oil palm empty fruit bunch (OPEFB), mengkuang (screw pine) leaves and water hyacinth [3]. Cherian et al [4] reported that PALF has 81.27% cellulose, 12.31% hemicellulose, 3.46% lignin, and 10.52% moisture content.

Malaysian Pineapple Industry Board (MPIB) reported pineapple is the most important tropical crop in Malaysia with approximately 15,000 hectares of land being planted with pineapples. Pineapple harvest time is twice per year. There are several types of pineapple such as Mois Gajah, Josapine, Sarawak, and N36 that are planted in Malaysia. Josapine type is the finest fibre and could be extracted without causing any damage. Apparently, the main attention of the Malaysian pineapple industry is the fruits and foodstuffs produced from the fruits even though the leaves have been known traditionally to the local people as sources of fibres for textile. Pineapple leaves are currently one of the prime agricultural wastes in Malaysia and massively generated from pineapple cultivation [5]. Hence, utilizing the biomass in the production of value-added product not only reduce the number of wastes generated in the plantation but also decrease the environmental impact to the surrounding.

However, there is a large variation in the properties and characteristics of PALF that became the most challenging part of working with natural fibre reinforced composites [6]. The main drawbacks for natural fibre/polymer composites are the incompatibility of hydrophilic natural fibres, which cause problems of adhesion with hydrophobic thermoplastic matrices. Therefore, it is significant to modify the fibre surface to decrease certain drawbacks of natural fibres such as low thermal degradation, high water absorption, and poor compatibility with polymer.

In recent studies, many researches proposed a few series of treatments and more chemical used in order to improve the surface of fibre. Some of the approach for surface modification treatments such as acetylation, coupling agents, polymer grafting, and mercerization [7]. Currently, the common surface modifications are alkaline [8], peroxide treatment

[9], modification with maleic anhydrous [10], and acetylation [11]. Therefore, in order to utilize PALF for the production of biocomposite, alkali treatment using NaOH is an easier and cost-effective method of pre-treatment.

However, it is crucial to identify the optimum condition for each treatment. The suitable concentration of chemicals used should be determined in order to achieve the maximum level of compatibility of fibre and matrix in producing biocomposite. Moreover, the problems faced at plantation could be manageable as the agriculture residue was utilized efficiently and reduce the burden to the landfill management team. In the past few years, numerous studies have reported the potential use of PALF as reinforcing materials in a wide range of polymers such as poly (hydroxybutyrate-co-valerate), polyester, polyethylene, polylactides (PLAs), polypropylene (PP), and natural rubber. PLAs are biodegradable polymers, are accessible from renewable sources, and degrade to water and carbon dioxide. However, the cost of PLAs is significantly higher than other polymers such as PP. Among the types of plastics, PP is extensively used in industrial products and domestic goods and especially as a matrix material in composites, due to low production cost, design flexibility, and recyclability, compared with other polymers [2]. Heat distortion temperature, flame retardant, transparency, and dimensional stability are several add-on potential properties of PP. Besides, in PALF reinforced polymers, it has been finely documented that the overall properties of composite materials are chiefly governed by adhesion among the PALFs and the matrix, which is a composite material with poor interfaces that have comparatively low stiffness and strength [5].

The prime target of this research is to determine the properties of PALF/PP biocomposite. According to previous studies, there are many types of treatment were applied to natural fibre in order to increase the compatibility of fibre and polymer. Some modifications have been made in alkali treatment on the PALF. Treated PALF and untreated PALF were characterized for their chemical, thermal stability and surface morphology analysis. Preparation of biocomposite from treated and untreated PALF mixed with PP at 30 wt.% of fibre content. Mechanical properties of PALF/PP bio-composite were determined from tensile strength analysis and water absorption analysis which were conducted for 14 days.

METHODS AND MATERIALS

Materials

Raw material PALF were collected from Pekan Pina Plantation located at Sungai Miang, Pekan, Pahang. Sodium hydroxide (NaOH) pellets with 96% purity were purchased from Sigma-Aldrich, while Polypropylene (PP) pellets were supplied by Commercial Plastic Industries Sdn.Bhd, Selangor.

Preparation of Fibre

Fresh Pineapple leaves were taken from the plantation area. The fresh leaves were cleaned with tap water to remove dirt such as soil. Then, the cleaned leaves were dried at ambient temperature overnight to prevent fungus growth on the leaves. Dried leaves were ground and sieved to produce micron-sized PALF using plastic SCV7510 TST Plastic Crusher.

Treatment of Fibres

Alkaline treatment was conducted to modify the surface of fibre with four different concentrations of NaOH which are 1, 2, 4, and 8 wt.%. All the treatments were performed at room temperature for 3 hours. After the treatments, the fibres were washed and dried in a circulating oven at 70 °C.

Characteristic of Fibre

The fibres were analyzed for their chemical, thermal stability and surface morphology using FTIR, TGA and SEM respectively.

Fourier Transform Infrared Spectroscopy (FTIR) Analysis

Untreated and alkali treated PALF fibre were dried at 60 °C until a constant weight was achieved [12]. The FTIR was conducted with Thermo Scientific iD7 ATR FTIR Spectrometer scanned in a range of 500 – 4000 cm⁻¹ at the resolution of 4 cm⁻¹ and the average scans for each sample were taken.

Thermal Stability Analysis

Thermal properties of five PALF samples which are untreated fibre, and alkali treated PALF with different concentrations of NaOH were tested using Thermogravimetry Analysis (TGA) Q500 V6.7 Build 203. About 1–5 mg of samples were placed on the ceramic crucible [13]. Analysis was conducted at 10 °C /min ramping method in a nitrogen atmosphere with gas flowrate at 40mL/min from temperature 26 °C to 700 °C. The derivative TG were obtained by differential of TGA using central difference method as the following Eq. (1)

$$DTG = \frac{W_{t+\Delta t} - W_{t-\Delta t}}{2\Delta t} \quad (1)$$

Moisture Analysis

The moisture content was analyzed using A&D, MS-70 Moisture Analyzer. A sample of 0.1 g was placed on an aluminium pan and heated to temperature of 105 °C. The percentage of moisture content were recorded.

Morphology Analysis

SEM analysis was conducted by using Hitachi TM3030 Plus Tabletop Microscope to obtain the three-dimensional surface morphology of treated and untreated fibres at high magnifications. The samples were mounted on an aluminium specimen stubs using double-sided adhesive carbon tape. Priorly, the samples were gold coated using Quorum machine by sputter technique to minimize sample charging. An accelerating voltage of 20 kV was used to scan the prepared biocomposites [14].

Preparation of PALF/PP Bio-composite

Untreated and treated samples of PALF were ground and sieved to size 180 – 355 µm. Then the samples were mixed with PP at 30 wt% of fibre content using an extruder. The extruder operates from 190 °C (inlet temperature) to 175 °C (temperature of die) with 50 rpm. The compound was compression-moulded by using hot and cold press machine to form a thin film sheet measuring 10 cm x 10 cm with 1 mm thickness [15]. The sample was pre-heated for 3 minutes, before the compound PALF was pressed.

Characteristic of PALF/PP Bio-composite

The biocomposite were analyzed for their mechanical properties and water absorption. Figure 1 shows the summary of overall process to fabricate PALF/PP biocomposite.

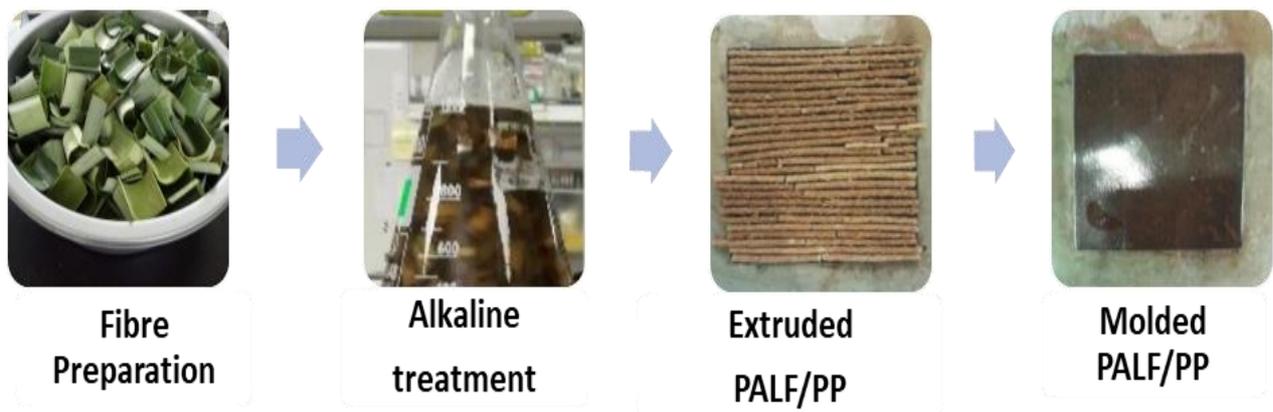


Figure 1. Summary process of PALF/PP biocomposite from raw pineapple leaves

Mechanical Properties

Five specimens were prepared for each condition to analysis with tensile test and the average value was calculated to determine the tensile strength for maximum force and break force of PALF/PP biocomposite. The tensile test was performed using an AG-1 Shimadzu Universal Testing Machine based on ASTM D638-08 Standard. The specimens were fitted with 1 kN load test [15]. The test was operated at a crosshead speed of 10 mm/min. Figure 2 illustrates the detailed experimental setup of the tensile test. Table 1 shows the identification of PALF/PP samples and conditions for each sample.



Figure 2. Experimental setup of tensile test

Table 1. Identification of samples and conditions for each sample

Samples	Condition for each sample
Sample 1	Untreated + PP
Sample 2	Alkali 1 wt.% + PP
Sample 3	Alkali 2 wt.% + PP
Sample 4	Alkali 4 wt.% + PP
Sample 5	Alkali 8 wt.% + PP

Water Absorption Analysis

The weight of the samples was measured before and after the testing. The samples were carefully cut in size 1 cm x 1.5 cm to avoid any cracks and dried at a temperature of 70 °C in an oven. Next, the weight of the dried sample was measured as the initial weight of biocomposite (W_i). A weight, W_f represents the weight of the sample taken daily up to 14 days. Three specimens of each sample were immersed in water for 14 days at room temperature. The average weights were recorded regularly within the time duration [16]. The percentage of water absorption of the biocomposite was calculated using Eq. (2):

$$\text{Water absorption (\%)} = \frac{(W_i - W_f)}{W_i} \quad (2)$$

RESULTS AND DISCUSSION

PALF Analysis

PALF analysis was performed to analyze chemical, thermal stability and morphology of the untreated and alkali treated PALF with different concentrations of NaOH; Alkali 1 wt.%, Alkali 2 wt.%, Alkali 4 wt.%, Alkali 8 wt.%.

Functional Group Analysis of PALF

FTIR supports the identification of changes in natural fibre chemical composition before and after the chemical treatment. Figure 3 shows the comparison result of FTIR for treated and untreated fibres. Alkanes, alcohols, aromatic, and esters were the identified functional group in the chemical structure [17]. As observed, the infrared absorption stretching between 3700–3000 cm^{-1} corresponding to hydroxyl group ($-\text{OH}$) of carboxylic acid and alcohol on lignin [18]. The stretching intensity of alkali treated fibre decreased and become more broader compared with untreated PALF. This pattern indicates most of the lignin was removed gradually as the concentration of NaOH increased [19, 20]. A parallel trend was also visible in the 2930 cm^{-1} (asymmetric $-\text{CH}_2$ stretching) and 2830 cm^{-1} (symmetric $-\text{CH}_2$ stretching) band region. These peaks are basically attributed to C–H stretching of methylene and methyl groups in alkanes [20]. The disappearance of these peaks on alkali treated PALF fibre subsequently reveal the elimination of lignin. Conferring to Song et al [21], infrared absorption peaks at the wavenumbers of 1350, 1710 cm^{-1} are allied with the benzene ring ($\text{C}=\text{C}$) in lignin and acetyl group ($\text{C}=\text{O}$) in hemicellulose respectively. The sharp peak at 1060 cm^{-1} represents pyranose ring skeletal ($\text{C}-\text{O}-\text{C}$) or β -glycosidic linkage between polysaccharides unit in cellulose [14]. These 3 peaks were clearly seen in the untreated PALF and decreased on alkali treated PALF fibre except for peak at wavenumber 1060 cm^{-1} . Hence, increasing alkali concentration (1 to 8wt.% NaOH) during treatments, reduce the lignin and hemicellulose components with less degradation of cellulose [22].

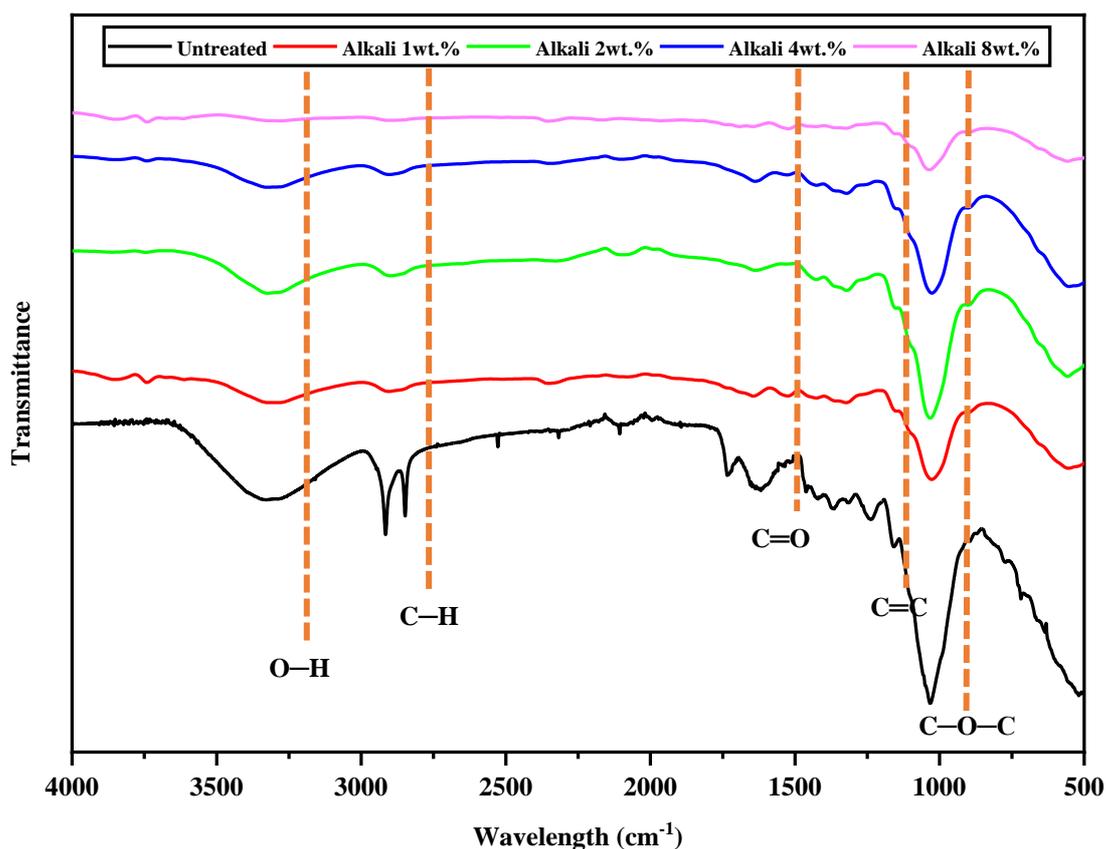


Figure 3. FTIR pattern of PALF

Thermal Stability Analysis of PALF

The degradation of the chemical composition of PALF was analyzed by TGA as shown in Table 2 by indicating the weight loss of the sample at 20% and 50% weight loss. Table 2 also showed the residue weight of PALF at maximum temperature of 700 °C. Figure 4 showed weight loss of PALF samples for untreated and alkali treated from room temperature to 700 °C. The Differential Thermal Gravimetry (DTG) thermograms is presented in Figures 5. The thermal degradation of natural fibre is a three-stage process [23]. All the treated fibre had almost similar thermal behaviors.

Table 2. Thermal stability analysis for PALF

PALF sample	Temperature for 20% Weight Loss (°C)	Temperature for 50% Weight Loss (°C)	Main thermal degradation temperature (°C)	Residue at 700 °C (%)
Untreated	146.68	401.94	251.06	39.82
Alkali 1 wt. %	151.68	379.82	255.33	36.86
Alkali 2 wt. %	213.26	352.38	258.33	33.46
Alkali 4 wt. %	216.78	288.38	253.71	27.08
Alkali 8 wt. %	232.71	292.95	270.46	25.00

According to Neto [24], the degradation temperature of hemicellulose and cellulose ranged between 150 to 350 °C, and 275 to 350 °C, respectively. Meanwhile, the degradation temperature of lignin occurs in a range of 250 to 500 °C. Figure 4 shows similar trend which had three regions where the weight loss occurred: (i) temperature less 100 °C, (ii) 150 °C – 250 °C, and (iii) 250 °C – 360 °C. The first region where the weight loss at a temperature less than 100 °C was due to moisture content. Hemicellulose degradation starts at 150 °C. As shown in Figure 4, multi-step degradation of untreated PALF gradually changed to one-step degradation for alkali PALF at a temperature between 500 °C to 700 °C. This indicates that the lignin and hemicellulose components had been removed partially after the treatment [25].

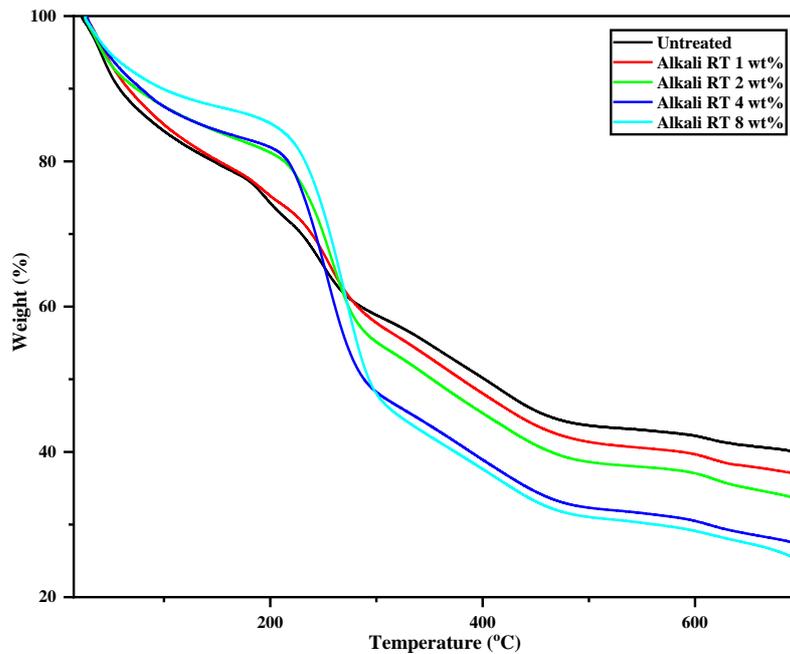


Figure 4. Weight loss of PALF

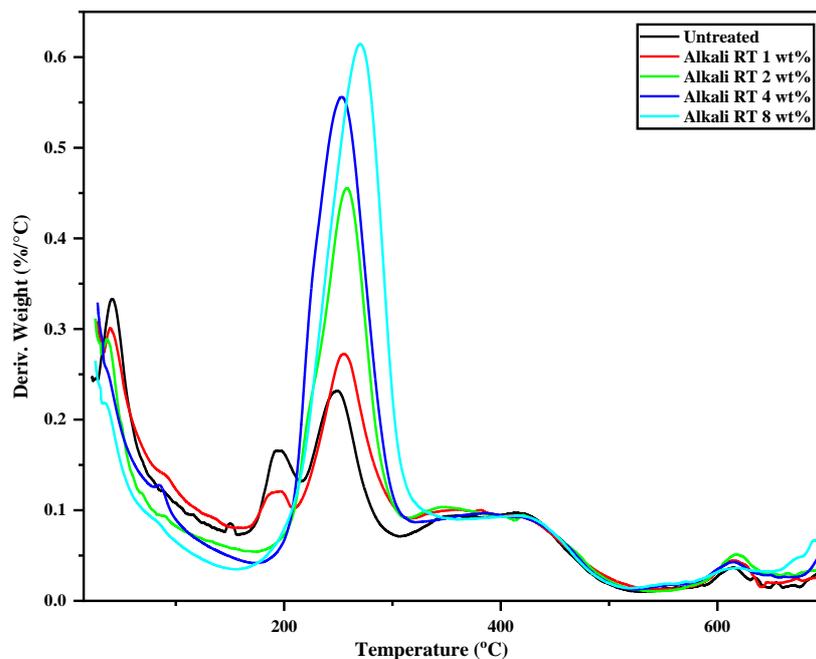


Figure 5. Differential Thermal Gravimetry of PALF

In Figure 5, it can be clearly observed that there are five peaks plotted where the weight loss occurred at; (i) temperature less than 100 °C, (ii) 180 °C to 220 °C, (iii) 220 °C to 300 °C, (iv) 300 °C to 500 °C and (v) 500 °C to 700 °C.

The first and second regions indicated weight loss due to moisture content, hemicellulose and low molecular weight of lignin. The weight loss at the third and fourth regions has corresponded to the formation of volatile products which arose from random chain intermolecular and scission transfer involving tertiary hydrogen abstractions from the cellulose and lignin. The weight loss in the fifth region was due to high molecular of lignin. The DTG of alkali treated PALF has low peaks compared to untreated fibre at temperature less than 200 °C as it shows the removal hemicellulose and low molecular weight of lignin in the samples. Furthermore, it also shows that the treated PALF is more thermally stable compared to the untreated PALF due to the removal of lignin and higher percentage of cellulose content. In addition, Wan Nadirah et al., [26] reported that an increase in the amount of cellulose may increase the percentage crystallinity of fibre. The increasing percentage of crystallinity influenced its materials properties such as thermal, physical, and mechanical properties.

According to Oushabi et al. [27], alkali treatments at high concentration results in disintegration of fibres and removal of lignin and hemicellulose thus the degradation of temperature increases as the concentration of NaOH increases.

Generally, alkali treatment is able to improve the thermal stability of PALF due to the removal of hemicellulose, lignin and impurities from the fibre.

Moisture Content of PALF

The moisture content of PALF is presented in Table 3. Untreated PALF owns the highest moisture content (16%) compare with treated fibre due to its intrinsic hydrophilic properties [28]. Besides, all the treated fibre depicted low moisture content as the concentration of NaOH increased. The hydrophilic nature of the PALF corresponds to the high of free hydroxyl groups bonding between the $-OH$ functional groups and surrounding water molecules and amorphous region [29]. However hydrophilic nature may cause poor interfacial adhesion between polymer and filler, which generally reduce mechanical properties of composites [30]. Sample treated with 8 wt.% alkali posses the lowest moisture content which is 10%, expresses great potential to be reinforced in the hydrophobic polymer matrix to form the composite with good compatibility.

Table 3. The moisture content of untreated and treated PALF

PALF sample	Moisture content (dry basis)
Untreated	16.36%
Alkali 1 wt.%	15.54%
Alkali 2 wt.%	12.89%
Alkali 4 wt.%	12.84%
Alkali 8 wt.%	10.41%

Morphology Analysis of PALF

Scanning electron microscopy was used to distinguish the difference between the surface morphology of the untreated and alkali treated PALF samples. Prior to the SEM analysis, the samples were coated with gold using a vacuum sputter coater to improve their conductivity and the quality of the obtained images [31]. Figure 6 shows the morphology of untreated and alkali treated. Figure 6;b-e reveal different morphology of alkali treated PALF. Interestingly, Figure 6(e), 8 wt.% of alkali treated PALF at room temperature shows that the fibre produced skeletal string-like microfibril bundle structures.

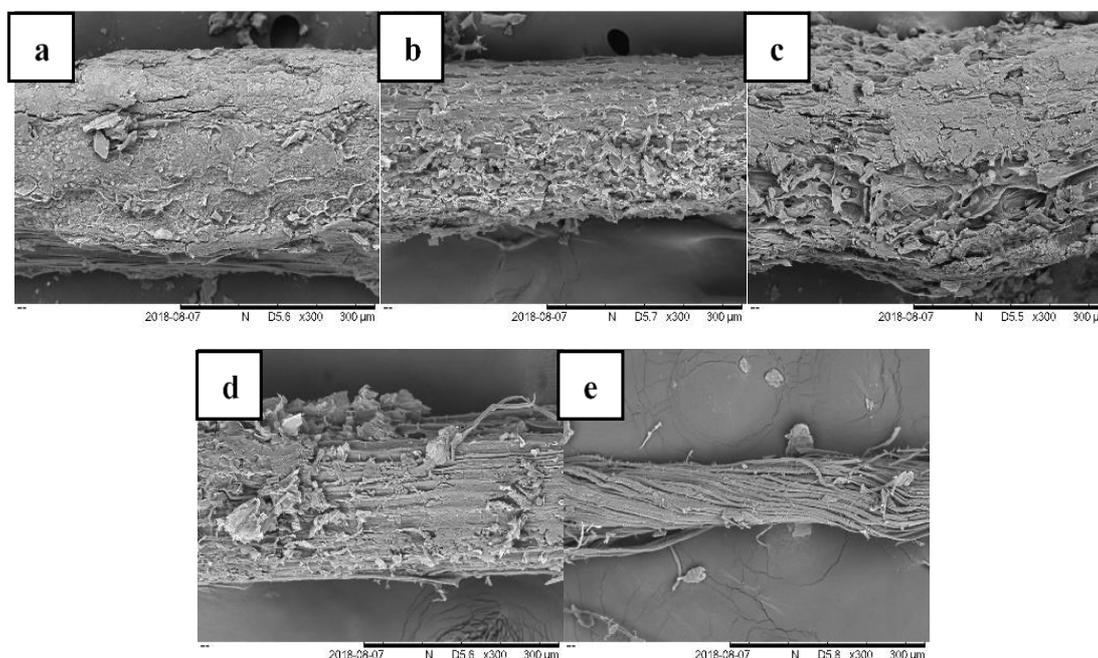


Figure 6. SEM image of (a) Untreated, (b) Alkali 1 wt.%, (c) Alkali 2 wt.%, (d) Alkali 4 wt.%, (e) Alkali 8 wt.% PALF under magnification of $\times 300$

Theoretically, alkaline treatment is one of the common chemical treatments used to solubilize hemicellulose lignin, oil, and waxy components and pectin in order to remove them from the natural fibre surface. The mechanism of alkaline hydrolysis is interrelated to the saponification of intermolecular ester bonds cross-linking lignin and part of the

hemicellulose [32]. The reduction of this cross-linking tends to increase the material porosity [33]. Apparently, the higher concentration of NaOH induces the conversion of alkoxide by ionization of hydroxyl group. It causes the defibrillation of fiber by loosening the cellulosic bundle and provides high aspect ratio for further surface modification.

In this study, 8 wt.% of alkali treated PALF (high concentration of NaOH) lead the major removal of impurities and amorphous region, from the fibre surface [34]. Moreover, the loose structure of the fibrils can be clearly seen.

PALF/PP Bio-composite Analysis

Figure 7 shows the physical appearance of PALF/PP biocomposite. Analysis of PALF/PP biocomposite is to determine the best treatment that improved the fibre matrix adhesion of PALF with PP matrix by comparing the tensile strength of PALF/PP biocomposite and the water absorption analysis on each treatment.

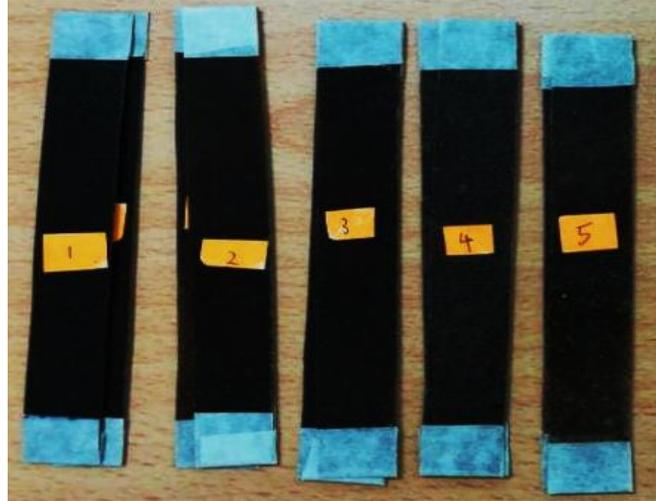


Figure 7. Physical appearance of PALF/PP bio-composite

Mechanical Properties of PALF/PP Bio-composites

Following mechanical tests were performed to evaluate the strengthness of the PALF/PP biocomposite. Result for tensile strength of PALF/PP biocomposite with different treatment conditions at 30wt% of PALF loading is shown in Figure 8. The highest tensile strength of the PALF/PP biocomposite was sample Alkali 8wt.% resulted at 116.17 MPa followed by Alkali 4wt.% and Alkali 2wt.% having 99.85 MPa and 90.56 MPa, respectively. It represented that, the higher the concentration of NaOH induce more removal of hemicellulose and lignin from PALF surface. Significantly, it will disrupt the hydrogen bonding network, thus increasing surface roughness fibre resulting in better physical interlocking [35]. Alkali treated PALF has a loss structure especially with 8wt.% alkali treatment, may increase penetration of PP hence give good tensile strength of the composite [36].

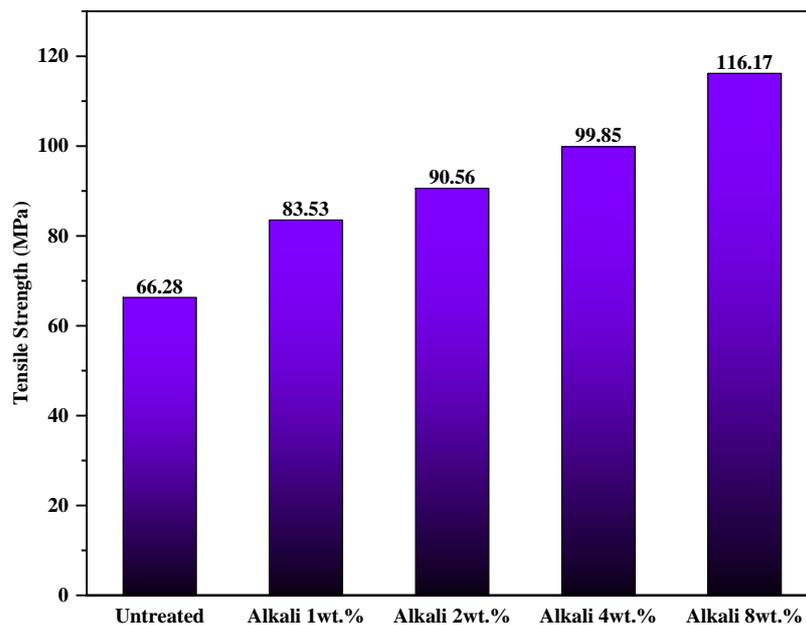


Figure 8. Summary of the mechanical analysis for PALF/PP bio-composite

Water Absorption Analysis on PALF/PP Bio-composite

Reinforcing PALF in composite materials is desirable because it can reduce the cost and usage of nondegradable PP. However, PALF might not be fit for numerous application fields because of their high water absorption. Therefore, water absorption is one of the significant aspects to assess on the properties of biocomposite. To limit the absorption of water, the surface of PALF is modified via several approaches including the alkaline treatment method [37,38]. Maximum percent water absorption for the samples was tabulated in Table 4.

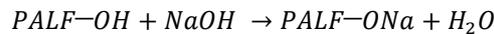
Table 4. Water absorption for all samples of PALF/PP biocomposites

PALF/PP bio-composite sample	Maximum water absorption (%)
Untreated	21.20
Alkali 1 wt.%	14.43
Alkali 2 wt.%	9.67
Alkali 4 wt.%	7.53
Alkali 8 wt.%	6.07

Based on Table 4, maximum percent water absorption for untreated, alkali 1 wt.%, alkali 2 wt.%, alkali 4 wt.%, and alkali 8 wt.% at room temperature of PALF/PP biocomposites were 21.2%, 14.43%, 9.67%, 7.53%, and 6.07% respectively. Figure 9 showed that all samples absorb water after being immersed in water until reached the saturation point at a duration of 14 days.

Untreated PALF has the highest water absorption compared to alkali treated due to the presence of lignin and hemicellulose which has higher OH group in the structure [38]. Alkali treated PALF at 8 wt.% showed that the treatment was successfully removed some percentage of hemicellulose and lignin content and decreased the water absorption of its PALF/PP biocomposite.

A finding by Beg et al., [39] stated that the hemicellulose contains hydroxyl group that has ability to absorb and create a hydrogen bond to form a water molecule lead to increase the weight of biocomposite. When PALF is treated with alkaline, the hydrophilic —OH groups in the fibre structure are converted to hydrophobic —ONa groups as the following reaction [37]:



According to Khalil et al., [40] removing hydrophilic hydroxyl groups from the fibre structure through fibre treatments is an essential step for the preparation of biocomposite. Apart from hydroxyl group content (hemicellulose), an additional reason for water absorption could be due to poor adhesion between fibre and a hydrophobic polymer, causing the biocomposite sample to have voids. These voids may have contributed to water penetration and higher water absorption, especially for less compatible biocomposite [16].

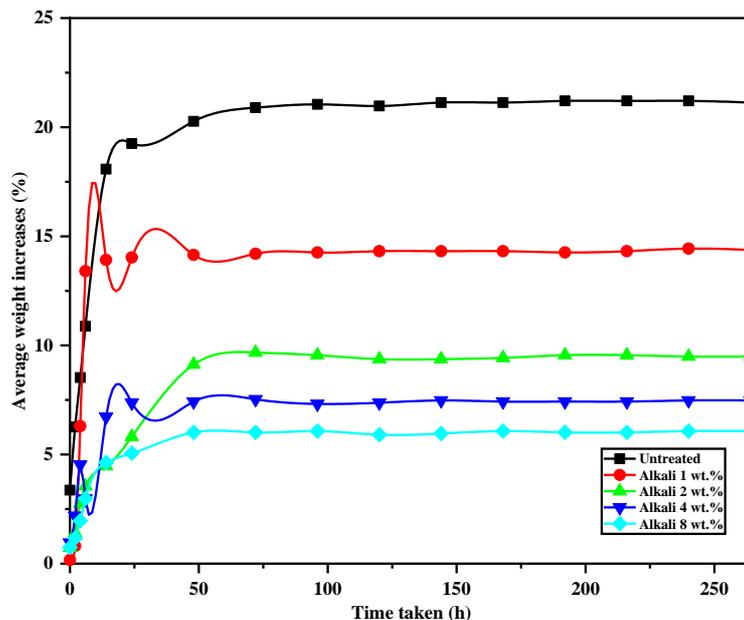


Figure 9. Water absorption for untreated, alkali 1 wt.%, alkali 2 wt.%, alkali 4 wt.%, alkali 8 wt.% PALF/PP biocomposites

CONCLUSIONS

The characteristics of PALF biocomposite such as tensile strength and water absorption were studied in this research. It was found that the characteristics of PALF were improved through alkali treatments. Significantly, PALF which is treated with a high concentration of alkali (NaOH) induces removal of impurities and promotes defibrillation by removing lignin and hemicellulose. In this study, 8 wt.% alkali treated PALF showed the best performance for thermal stability by increased the maximum thermal degradation temperature from 251.06 °C of untreated PALF to 270.46 °C. It had the lowest water absorption which was 71% lower than untreated PALF/PP bio-composite and tensile strength of 116 MPa.

To overcome the hydrophilic properties of the natural fiber, surface modification is necessary to improve the interfacial adhesion and mechanical properties of biocomposite. Natural fiber composites can be used in an automobile industries such as parcel shelves, door panels, instrument panels, armrests, headrests, and seat shells. Plastic/wood fiber composites also are being used in a large number of applications such as in construction and good packaging.

In conclusion, alkali treatment is able to improve the characteristics of PALF and able to promote a better adhesion of PALF with polymer matrix.

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