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SURFACE MODIFICATION OF ABACA FIBERS WITH GLUTARALDEHYDE FOR IMPROVED MECHANICAL PROPERTIES OF INJECTION MOLDED PLA BIOCOMPOSITES

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Abstract. *This research is based on the importance of finding crosslinking agents that are environmentally friendly and able to improve the performance of the interfacial area between natural fibers and their polymer matrix. Poly(lactic acid) PLA composites containing 20 wt.% abaca (Musa Textilis) fibers were manufactured by injection molding. The composite specimens contained surface-modified abaca fibers with 5 wt.% glutaraldehyde (GA) as a cross-linking agent were examined in this experiment. The SEM was applied to assess the morphological surface of the abaca fibers, while FTIR analysis was performed to investigate the chemical components of abaca fibers. The mechanical characteristics of the PLA/Abaca fiber biocomposites were examined using flexural, tensile, and Izod impact tests. The results confirmed that the modulus of elasticity and Young's modulus were observed to increase by 40% and 37% respectively. Meanwhile the tensile strength and the flexural strength increased by 26% and 15% respectively. PLA/Abaca fiber biocomposite that had been treated with glutaraldehyde had better mechanical qualities than pure PLA biocomposite.*

Keywords: *abaca fiber; PLA; alkaline treatment; glutaraldehyde; mechanical properties.*

1. Introduction

Currently, the trend of research on natural fiber composites is being carried out by many researchers. Natural fibers as composite reinforcement is excellent because they are sustainable, easy to obtain, and can reduce environmental pollution. Natural fibers have good mechanical strength, low density, and low price compared to glass fibers and other synthetic fibers [1], [2]. One of the natural fibers widely used in research on composites reinforced with natural fibers is abaca fiber or Musa Textilis fiber. Abaca (Musa Textilis) is a native plant widely cultivated in the Philippines and Indonesia. Abaca fiber contained 56-74 wt.% cellulose, 24-32 wt.% hemicellulose, 10-15 wt.% lignin, and 0.5-5 wt.% pectin [1], [3]–[5]. Some studies showed that abaca fiber as reinforcement in composites has high mechanical properties [5]–[13]. Due to these superior properties, abaca fiber-reinforced composites have been patented by one of the vehicle manufacturers in Europe as a vehicle exterior component [3], [10], [14].

Recently, many researchers have been conducting research on environmentally friendly

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plastics such as PLA because of the growing environmental problems brought on by non-biodegradable plastics. PLA is a type of thermoplastic resin made from plant products, such as cassava, corn, and other plant parts that contain starch. When used as a component of vehicle exterior, PLA-based composites emit less CO_2 than other thermoplastics composites [14]. Although PLA resin has good mechanical properties, it has some disadvantages, such as brittleness, poor thermal deformation, and poor impact resistance. To overcome these drawbacks, PLA is usually strengthened with fiber, one of which could be natural fiber [15]. Nonetheless, most natural fibers show poor adherence to matrix polymers. Poor mechanical strength is a result of inadequate adhesion between the fiber surface and the PLA matrix [16]. To increase interfacial adhesion, natural fibers must be modified before being used as composite reinforcement.

The alkaline treatment is one of the most utilized fiber surface modifications [17]. In alkaline treatment, sodium hydroxide (NaOH) is the most used chemical [18]. Alkaline substances could reduce hydrogen bonding by reacting with hydroxyl groups of fibers. Alkaline treatment can also get rid of hemicellulose, reducing the amount of lignin, reducing water absorption, and increasing tensile strength in natural fibers [3], [10], [19].

Another natural fiber treatment method is to use a coupling agent or cross-linking agent, such as silane. The amount of cross-linking on fiber surfaces can be increased by coupling agents, which is required to optimize fiber-matrix reinforcement [20]–[22]. Glutaraldehyde (GA) is a commonly used chemical cross-linker. In comparison to other aldehyde, glutaraldehyde has the better cross-linking ability [23]. The application of cross-linking agent glutaraldehyde as a surface modifier in abaca fibers has not been investigated in previous studies. The objective of this research is to examine the impact of two stages of fiber treatment, first using alkaline treatment, then continued with treatment using the cross-linking agent glutaraldehyde on the fiber microstructure, chemical composition, and mechanical characteristics of PLA composites with abaca fiber reinforcement.

2. Methods

2.1. Materials

The PLA resin utilized in the present investigation was made from cassava starch under the name PD183577-NV, which has an MFR of 16-24 g/10min and a specific gravity of 1.23-1.36. Abaca fiber is obtained from local agriculture in North Sulawesi, Indonesia. Sodium hydroxide (NaOH) was purchased from Sigma-Aldrich with 100% purity, and the cross-linking agent Glutaraldehyde (GA) 25% aqueous solution from Sigma-Aldrich.

2.2. Fibers Preparation

Fiber washing treatment by distilled water was adapted for washing fibers to remove

impurities by using an ultrasonic cleaner (GT Sonic Professional Ultrasonic Cleaner). The surface of abaca fibers was ultrasonically cleaned in distilled water, and the temperature was kept at 60ºC for 60 min. Abaca fiber was removed from the ultrasonic device when the temperature was low enough at 27ºC. The washed abaca fibers were allowed to dry for 24 h at 60ºC and then heated for 2 h at 120ºC in an oven. The fibers resulted in distilled water-washing treatment referred to as the untreated abaca fibers. The untreated abaca fiber from the earlier washing is subjected to an alkaline treatment in the following stage. Soaking abaca fibers with a 5% NaOH solution at 60°C for 24 h in an ultrasonic cleaner. The abaca fiber was then cleaned three times in an ultrasonic cleaner with distilled water until neutral (pH 7). After washing, the abaca fibers were left to dry for 24 h at 60ºC and then heated for 2 h at 120ºC in an oven. The fibers resulted in NaOH treatment, referred to as the alkaline-treated abaca fibers. Alkaline-treated abaca fibers were then modified with 5% glutaraldehyde (GA) solution at 90ºC for 24 h and then heated at 120ºC for 2 h in an oven. These fibers resulted in GA treatment, referred to as the GA-treated abaca fibers.

2.3. Composites processing

The fabrication of composites consists of two parts: a twin-screw extruder was used to create a filament compound for use in the composite manufacturing process, and the composite material was made using an injection molding machine from the pelletized filament. The PLA resin, abaca fibers were predried in the oven for 2 h at 60°C to eliminate any moisture before being processed into the extruder machine. Then, abaca fibers and PLA resin were mixed with a mixture ratio of 20:80 wt.% for each fiber. This mixture is then fed into the Compounder E Entrance ZK25E series manufactured by COLLIN Lab & Pilot Solutions GmbH Germany with 8 zones of the extruder. The extruder temperature was set to 40ºC, 100ºC, 130ºC, 150ºC, 170ºC, 190ºC, 200ºC, and 220ºC for zone 1 to zone 8 respectively. The screw rate was adjusted to 60 rpm with a PLA resin feed rate of 65 g/min and a fiber feed rate of 1.5 g/min. The filaments produced by the extruder were then chopped into pellets using a COLLIN Granulator type CSG171/1 pelletizer.

The pellets were dried for 2 h at 80ºC to eliminate any moisture left from the cooling process with water during the filament-making process with an extruder machine. The following step is the fabrication of composite specimens using an injection molding machine. The Wittmann Battenfeld type BA400/125 CDC, which has 4 temperature zones, was used for this injection molding technique. The temperatures for the nozzle, zone 2, zone 3, and mold were each set at 210°C, 190°C, 180°C, and 30°C, respectively. The maximum injection speed and maximum pressure were 60 m/s and 140 MPa, respectively. Then, the composite specimens were molded using molds that comply with ASTM D790 testing standards for flexural testing, ASTM D638 for tensile testing and ASTM D256 for IZOD impact testing.

2.4. Measurements

In this experiment, FTIR was utilized to compare the chemical components of untreated and treated abaca fiber. Fourier Transform Infra-Red Spectrophotometer (FTIR) is a laboratory instrument used to determine the molecular structure of compounds by identifying their functional groups in the form of molecular vibration spectra. Each abaca fiber sample was first prepared by pulverizing it into a fine powder before being inserted into the FTIR instrument for analysis. The FTIR instrument used in this experiment is the TENSOR 27 FTIR Spectrometer manufactured by Bruker Optic GmbH Germany with a spectra region of 600 – 10.000 cm^{-1} and 0.9 cm^{-1} spectral resolution. Meanwhile, SEM was utilized to study the microstructure of the surface of each abaca fiber. The Scanning Electron Microscope or SEM is a particular kind of electron microscope that generates images of the surface of a material with magnification up to a certain scale using focused electron beam scanning. The interaction between electrons and atoms in the material generates a series of signals. These signals provide information about the surface topography and composition of the material. Before being analyzed by SEM, each abaca fiber sample was pretreated with gold coating with a thickness of 40 nm. The fiber samples were then placed on the sample holder into the analytical SEM type JSM-6510LA manufactured by JEOL. The surface microstructure of the abaca fiber samples was observed using the secondary electron detector with the accelerating voltage of 15 kV.

In this study, the mechanical properties of biocomposite materials were tested using the Shimadzu AG-50KNXPLUS Universal Testing Machine. UTM is equipment used to perform several standard tests such as tension, compression, adhesion, tensile, and bending tests. Flexural testing is a technique used to assess the mechanical behavior of materials. subjected to bending or flexing. This method involves setting the specimen on a support and applying loads at specific points to bend the specimen. The flexural test used in this study is the three-point bending test. In this method, the specimen is subjected to a load by means of three separate points of support. Two points are placed in the support area, and the other is placed at the center point of the specimen, creating a span between the two points. Then, at the center of the specimen, a load is applied, which gradually increases until the specimen fails or breaks. Flexural tests examine a variety of properties, including flexural strength and modulus. In this study, PLA biocomposite specimens from the injection molding with a size of 125 mm × 12.7 mm × 3.2 mm were previously prepared for flexural testing. The maximum load that was applied was 50 kN at a loading speed of 5 mm per minute.

A tensile test (or tension test) involves exerting pressure on a specimen to measure the material's response to tensile (or pulling) stress. The tensile test was determined according to the ASTM D638 standard. A tensile test specimen with a size of 165 mm × 19 mm × 3.2 mm in the

shape of dogbones was obtained from the injection molding process and then prepared into the UTM machine. From this measurement, the tensile strength and modulus of elasticity or Young's Modulus of the specimen were obtained. The impact strength of the composite specimens was evaluated in this experiment using an Izod impact test. The impact test is a test to determine the energy absorbed by a specimen when it is fractured at high speed. The Izod pendulum impact test was conducted to determine the toughness of the specimen so as not to break when subjected to sudden shocks. The Izod impact test was conducted on a Resil 25 CEAST Italy pendulum testing machine. Specimens produced from injection molding were molded with a size of 65 mm × 12.7 mm × 3.2 mm and given a notch with a depth of 2.7 mm in accordance with ASTM D256 standards. The Instron Motorized Notchvis machine was used to create the notch on the composite specimens. The pendulum hammer for the Izod impact test has an impact energy range from 0.5J to 50J and an impact velocity of 3.5 m/s.

3. Results and Discussion

3.1. FTIR analysis

The FTIR measurement result of untreated abaca fiber is shown in Figure 1. The abaca fiber's FTIR spectrum can be divided into two sections, namely the band between 3400-2800 cm^{-1} and 1700-600 cm^{-1}. The O–H (hydroxyl group) stretching bonds of cellulose and hemicellulose have a peak in the absorption band at 3449 cm^{-1} [1], [3], [5]. Asymmetric C–H stretching bonds of cellulose and hemicellulose occur at 2886 cm^{-1} [1], [3], [5]. The peak at 1735 cm^{-1} corresponds to the aromatic ring of pectin and lignin [1], [3], [5]. The C=C stretching bands of cellulose and hemicellulose are associated with the peak at 1641 cm^{-1} [1], [3], [5]. The C=C bond of the aromatic ring of the lignin is represented by the peak in the band at 1511 cm^{-1} [1], [3], [5]. The =C–H in-plane bending of cellulose and hemicellulose occurs at 1458 cm^{-1} and 1433 cm^{-1} [1], [3], [5]. The cellulose and hemicellulose O–H bending is related to the 1324 cm^{-1} peak [1], [3], [5]. C–H in-plane bending bands of lignin and xylan appear at 1247 cm^{-1} [1], [3], [5]. The =C–H out-of-plane bending of cellulose and hemicellulose appears in the range between 1000 cm^{-1} and 600 cm^{-1} [1], [3], [5].

The abaca fiber that has been treated with NaOH is presented in Figure 2. Alkaline treatment was able to eliminate most of the lignin and pectin in abaca fiber. The peak at 1735 cm^{-1} disappeared after alkaline treatment. Additionally, a decrease in the amount of hydroxyl groups of hemicellulose in abaca fiber resulted in a reduction in the percentage of light transmittance at the 3447 cm^{-1} peak band. The abaca fiber that has been treated with glutaraldehyde is shown in Figure 3. The treatment of glutaraldehyde effectively eliminated a greater quantity of hemicellulose and lignin from the fiber, pointed out by the absence of the peak at 1247 cm^{-1}, which is a representation

of lignin. In addition, the reduction of more hydroxyl groups or hydrogen bonding (O–H) from hemicellulose can be seen from the decrease in light transmittance, which is more than 50% at 3449 cm^{-1} band.

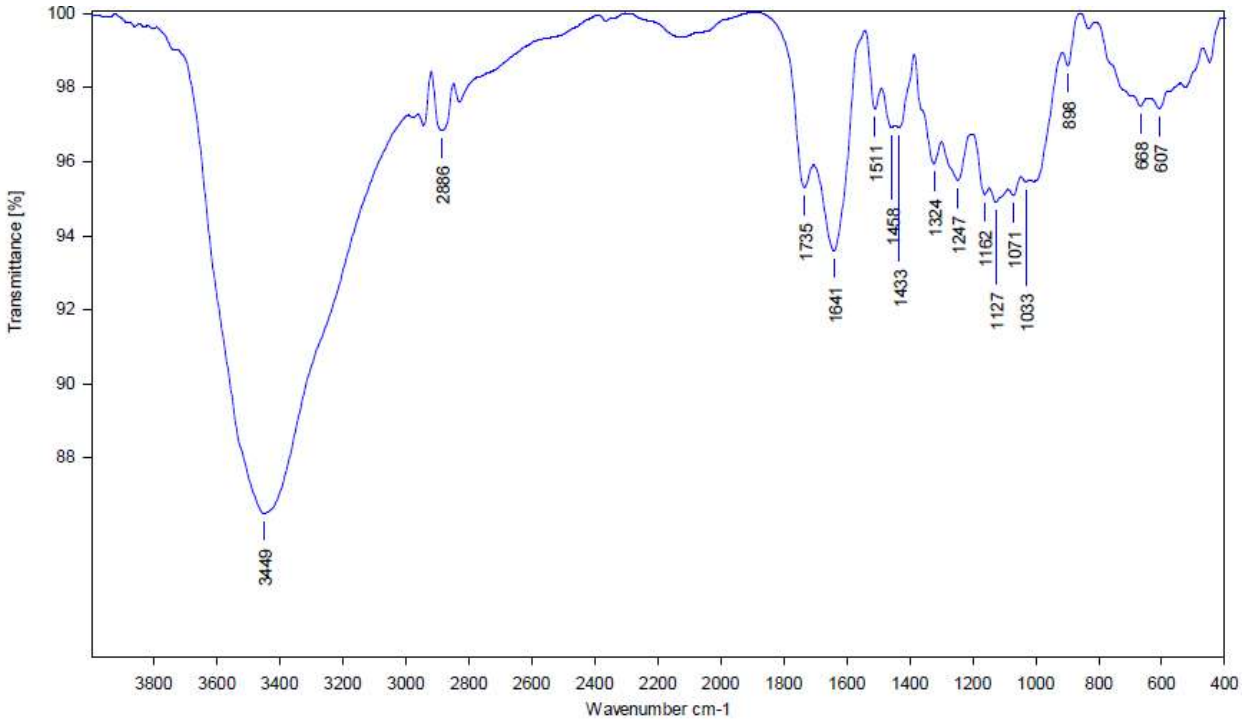


Figure 1. FTIR spectroscopy of untreated abaca fiber

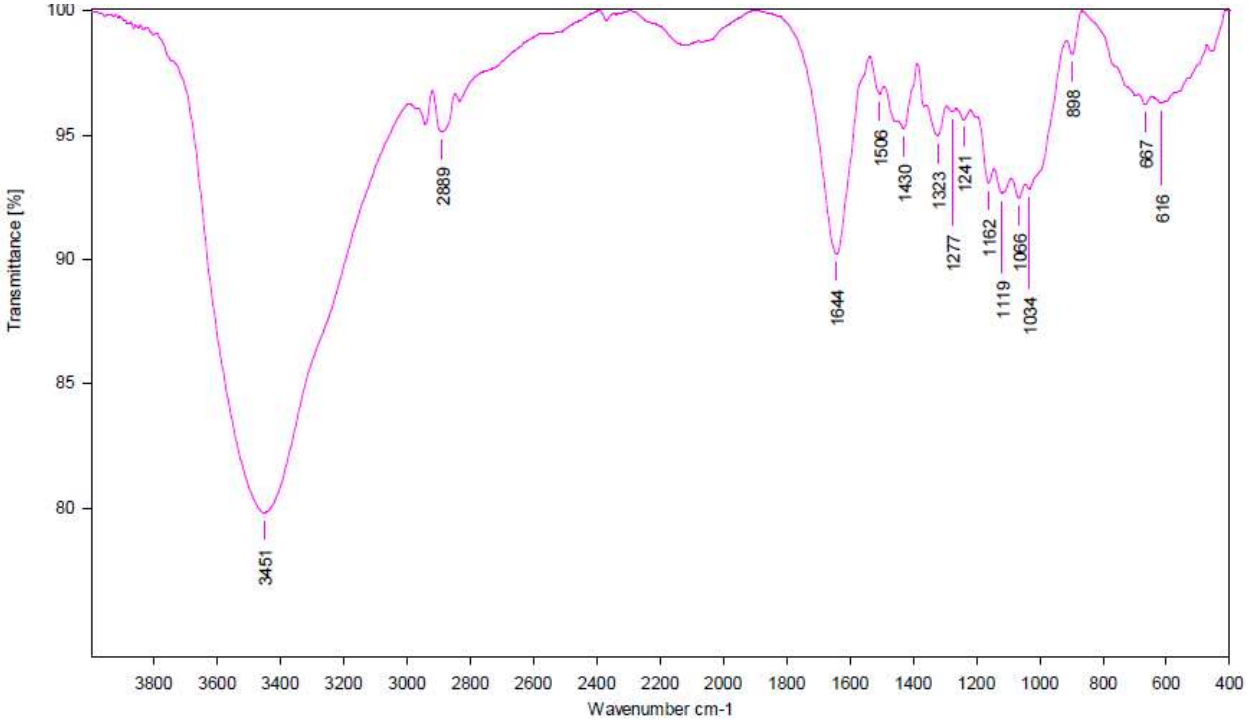


Figure 2. FTIR spectroscopy of alkaline-treated abaca fiber

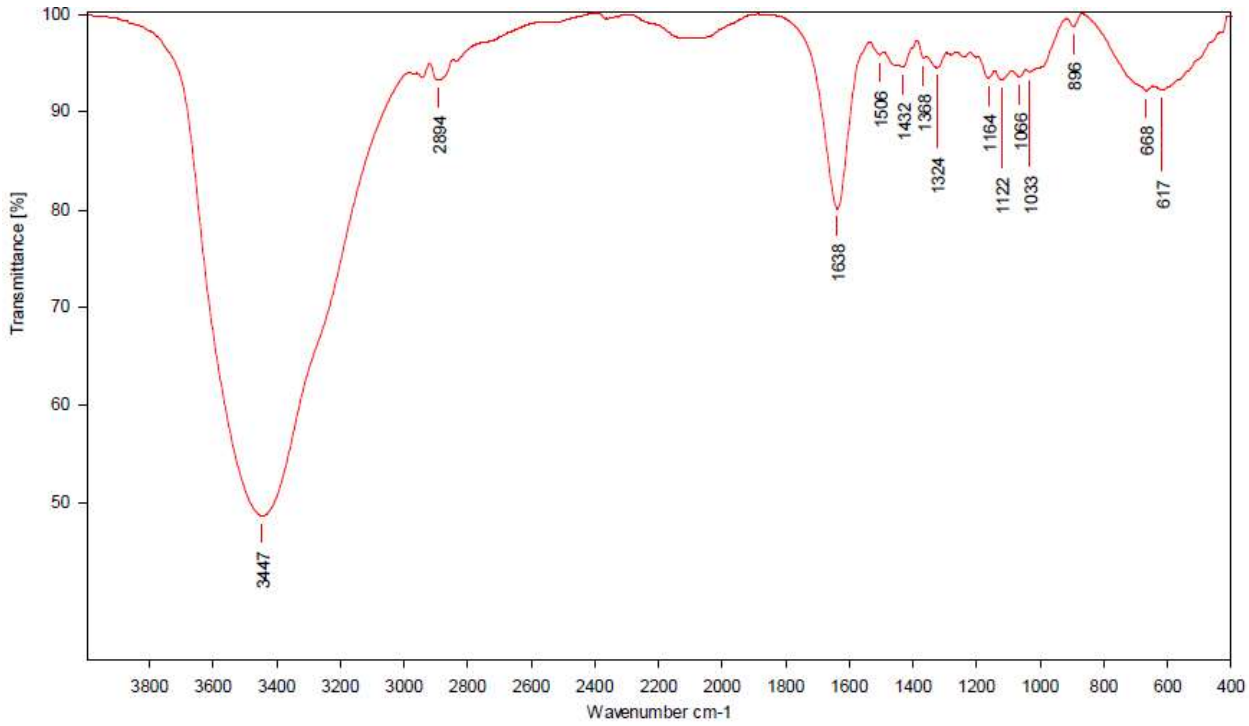
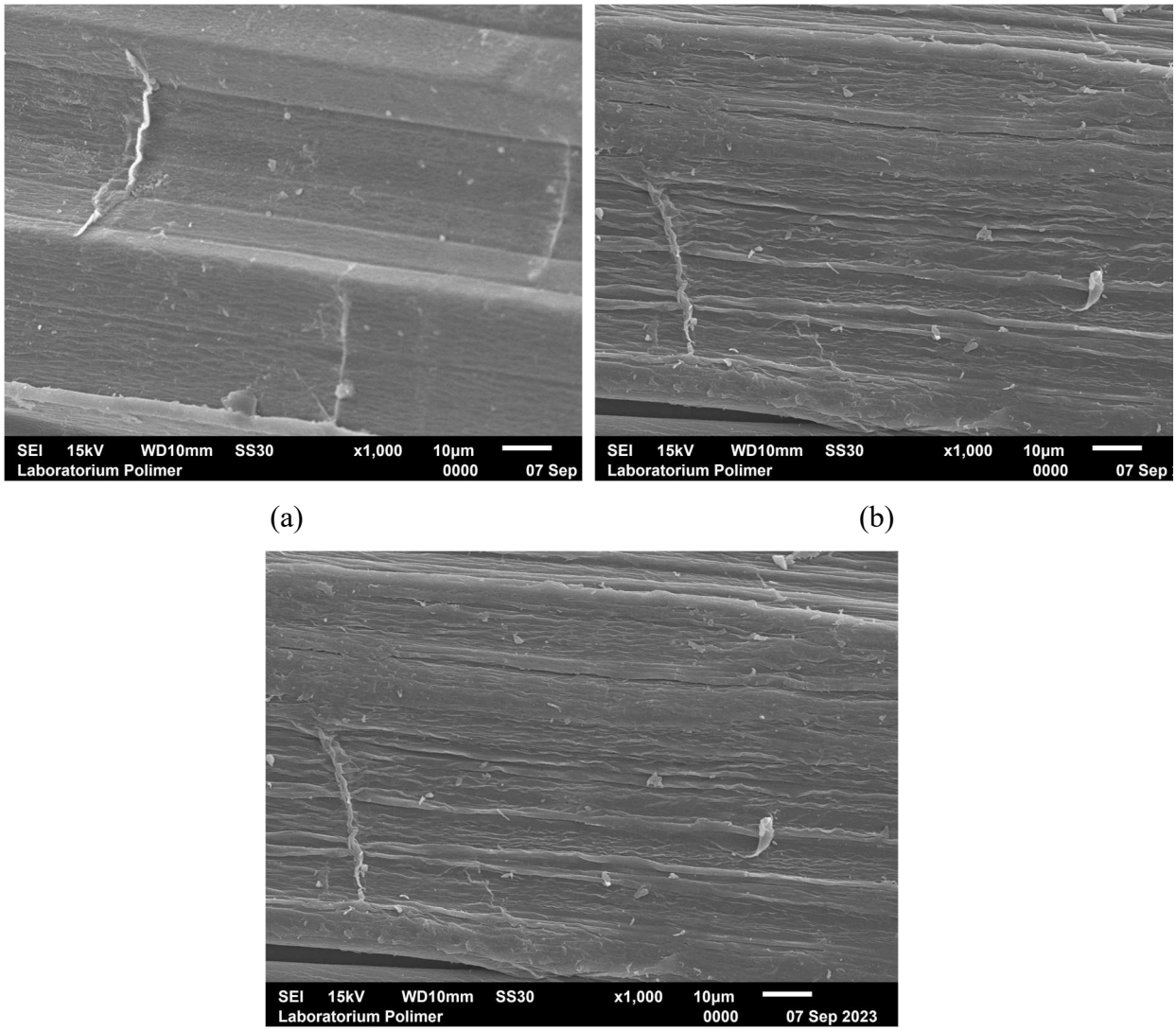


Figure 3. FTIR spectroscopy of Alkaline-GA-treated abaca fiber

3.2. SEM Analysis

Figure 4(a) showed the surface morphology of the untreated abaca fiber. A strong lignin structure and a waxy layer of abaca fibers are seen in the microstructure of untreated abaca fibers. While in both alkaline (Figure 4b) and alkaline-GA treated abaca fibers (Figure 4c), the rigid structure is no longer visible. This is supported by the FTIR examination results, which show that the lignin, pectin, and hemicellulose content of the abaca fiber has been removed. Abaca fibers also fibrillate when treated with alkaline and glutaraldehyde treatments, leading to the formation of finer elementary fibers. Several studies have reported that alkali treatment of abaca fibers can reduce fiber diameter and increase fiber surface roughness [1]. This was also confirmed in the appearance of alkali-treated and alkali-GA-treated abaca fiber, showing improved roughness of fiber surface in comparison to untreated fiber. The increased surface roughness of abaca fibers resulting from the alkaline-GA treatment provides an improvement in the mechanical properties of fibers and their composites. This is because the roughness of the fiber interface can increase the adhesion of the fiber with PLA matrix.



(a) (b)

(c)

Figure 4. (a) Untreated abaca fiber, (b) Alkaline treated abaca fiber, (c) Alkaline-GA treated abaca fiber

3.3. PLA/fiber flexural properties Materials

Flexural testing measurements show that the addition of abaca fibers, both untreated and treated fibers, can significantly increase the elastic modulus of PLA/fiber biocomposites compared to the neat PLA biocomposites (Figure 5). The modulus of elasticity of biocomposites reinforced with abaca fibers treated with alkaline GA was observed to increase by 40%, which have a modulus of elasticity of 5.2 GPa. The flexural strength of the PLA/fiber biocomposite also increased compared to the pure PLA biocomposite. There was a 15% increase from 63.9 MPa for the neat PLA biocomposite to 74.9 MPa for the alkaline-GA treated fiber-reinforced PLA biocomposite. The increased modulus of the biocomposite is attributed to the fibers being embedded and aligned in the PLA matrix. Meanwhile, the modification of the fiber surface improves the adhesion of the fiber surface to the PLA matrix, resulting in increased flexural strength of PLA biocomposite.

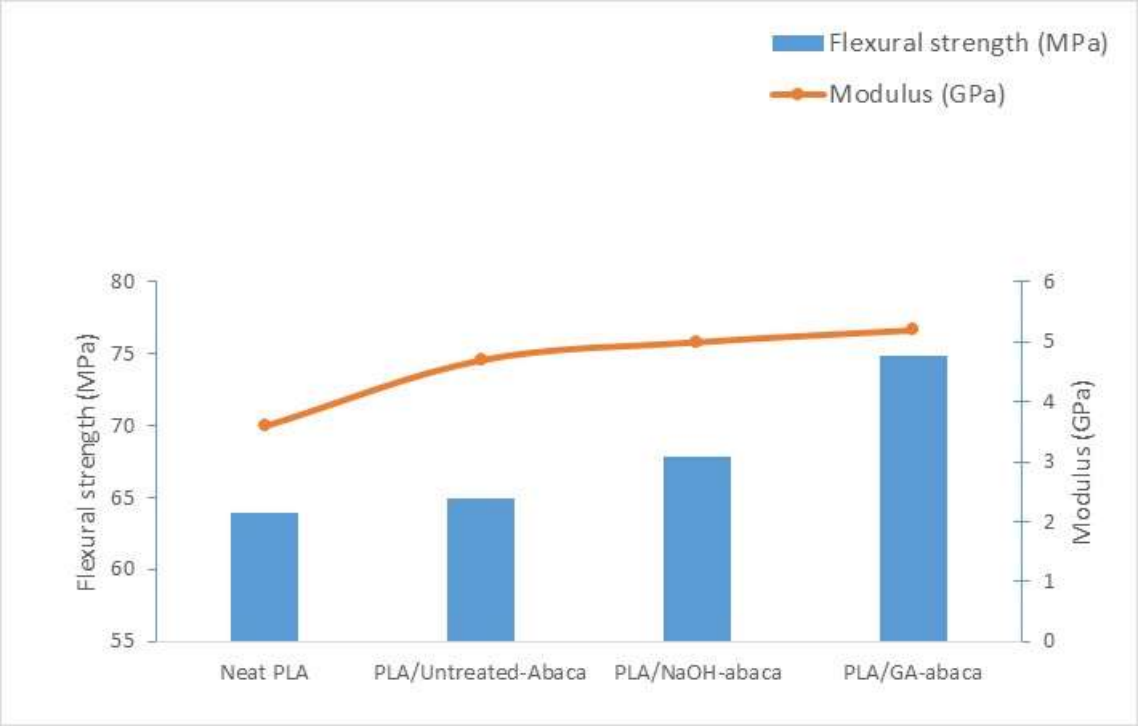


Figure 5. Flexural properties of the PLA/abaca fiber biocomposites

3.4. PLA/fiber tensile propertiesMaterials

The tensile test findings are presented as the PLA biocomposite specimens' tensile strength and Young's modulus (Figure 6) The results of these measurements show that The introduction of abaca fiber can improve the modulus of elasticity of PLA biocomposites. It is known that cellulose contained in fibers increases the modulus of materials [21]. Young's modulus increases by 37% in biocomposites reinforced with alkaline-GA-treated abaca fibers, which have Young's modulus of 3.3 GPa. However, there is a decrease in tensile strength in PLA biocomposites reinforced with untreated abaca fibers and alkali-treated abaca fibers in comparison to pure PLA biocomposites. This is likely due to the weak adhesiveness between the fiber surface and PLA matrix. In contrast, there was an increase in tensile strength in PLA biocomposites reinforced with abaca fiber treated with alkaline-GA. This is due to a cross-linking mechanism between the fiber surface and the PLA matrix. The tensile strength of the PLA/alkaline-GA treated fiber biocomposite increased by 15% compared to the PLA/untreated fiber biocomposite from 42.1 MPa to 52.1 MPa. Meanwhile the alkaline-GA treated fiber-reinforced PLA biocomposite exhibited a 26% increase in strength, from 63.9 MPa to 74.9 MPa, in comparison to the pure PLA biocomposite.

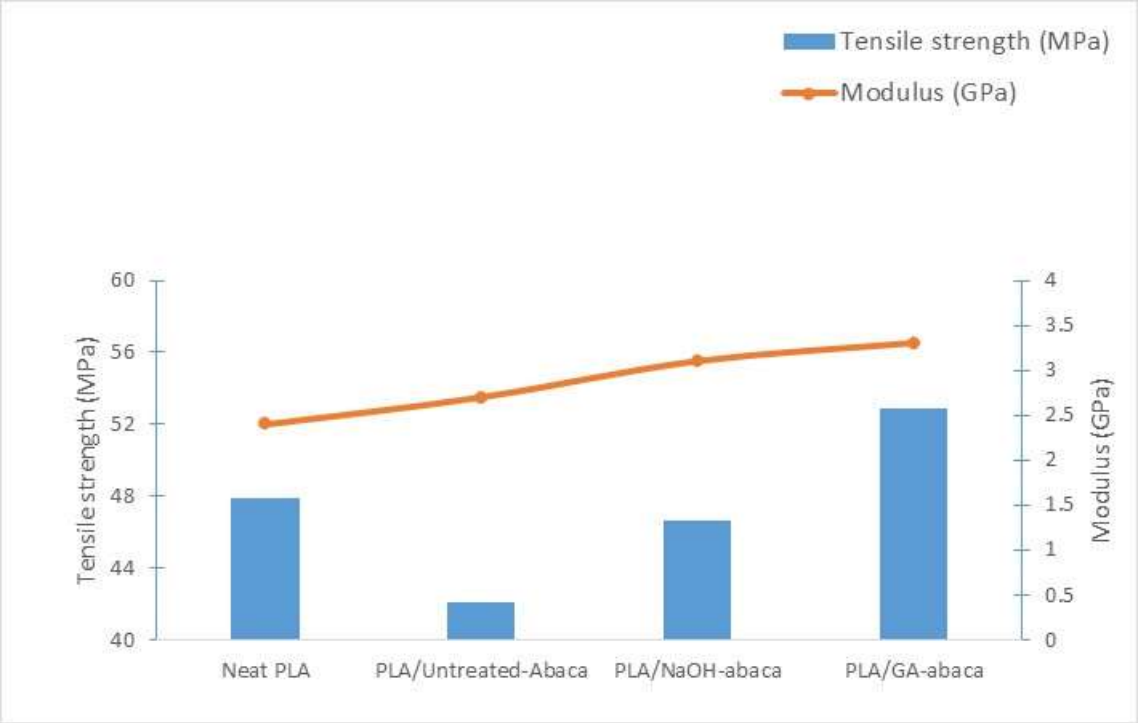


Figure 6. Tensile properties of the PLA/abaca fiber biocomposites

3.5. PLA/fiber Izod impact properties

Figure 7 shows the impact strength of PLA biocomposites as determined by Izod testing. The impact strength of the PLA/alkaline-GA treated fiber composite increased by 63% compared to the pure PLA composite. Surface treatment with alkaline-GA was able to increase the impact strength of PLA biocomposites. Impact strength for the pure PLA composite is 11.5 J/m, and after adding alkaline-GA treated abaca fiber the impact strength of the composite was significantly increased to 18.7J/m. The increase in the impact strength of PLA biocomposites is influenced by the adhesion of the fiber surface with the PLA matrix. This tends to be influenced by the amount of celluose in the fiber due to alkaline-GA treatment of abaca fiber.

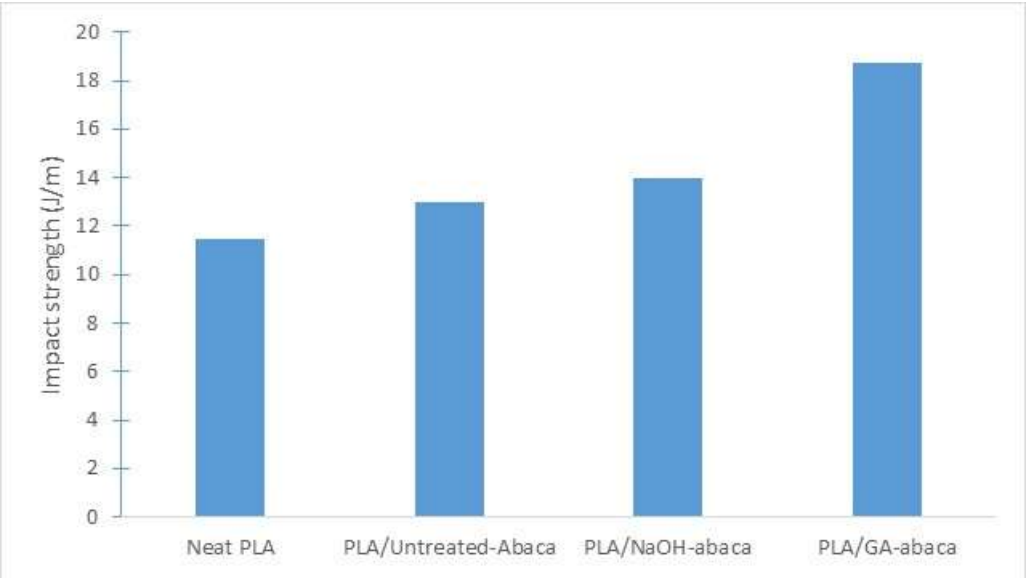


Figure 7. Izod impact properties of the PLA/abaca fiber biocomposites

4. Conclusions

This study successfully investigated the influence of NaOH treatment followed by glutaraldehyde treatment on the mechanical properties of abaca fiber used as reinforcement in PLA biocomposite. The results showed that after alkaline treatment and glutaraldehyde treatment, the cellulose content of abaca fibers increased significantly. This was indicated by the loss of functional groups of lignin, pectin, and hemicellulose in the fiber, which was confirmed through the FTIR spectrum. From the results of the fiber surface morphology through SEM analysis, abaca fibers were fibrillated due to alkaline and glutaraldehyde treatment, which was indicated by the visible elementary structure of the fiber. The introduction of fibers into the PLA matrix improved the mechanical properties of the PLA biocomposites, in particular the elastic modulus and Young's modulus values. The addition of alkaline-treated fibers and 5 wt.% glutaraldehyde significantly increased the tensile and flexural strength of the PLA biocomposites.

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