Liquid Smoke Treatment for Natural Fibers: The Effect on Tensile Properties, Surface Morphology, Crystalline Properties, and Functional Groups of Banana Stem Fibers

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Abstract: This study aims to determine the effect of the treatment of banana stem fibers (BSF) with grade three liquid smoke on changes in the micromechanical properties of the BSF, single fiber tensile strength, morphology, crystal properties, and functional groups. This study used four variations of the specimen model, namely, fiber without treatment and immersion in liquid smoke for 1, 2, and 3 h. BSF with treatment was dried in an oven at 40 °C for 30 min. Several tests were carried out, including the tensile test for single fiber capacity of 50N standard ASTM 3379-02, Scanning Electron Microscope (SEM), X-Ray Diffraction (XRD), and Fourier Transform Infra-Red (FTIR) observation. The results showed that the highest increase in fiber strength occurred in P2J, which was 43.78%, with crystal intensity of 34.97%, compared to TP fiber. Treatment of fiber with liquid smoke can form a strong C-C elemental bond caused by the H2O degradation process in BSF so that the carbon atom (C) becomes solid; under conditions of excessive H2O degradation, the fiber strength will become brittle, however, liquid smoke can increase the fiber tensile strength. The morphology of the fiber changed where the untreated fiber was covered in lignin, while the treated fiber had a rectangular pattern of elongated lines, was porous, and the lignin was eroded. The fiber crystallization index increased due to changes in fiber structure, where the highest peak of TP BSF occurred at point two, while the highest peaks in BSF P1J, P2J, and P3J occurred respectively at points two and three. These results prove that the innovation of BSF treatment with liquid smoke can change the morphology, crystalline, and functional aspects of BSF, so that it becomes the choice of composite reinforcement material in the future, an option that is lightweight and environmentally friendly.

Keywords: liquid smoke; banana stem fiber; tensile strength; morphology; crystalline properties; functional groups; light weight; environmentally friendly

1. Introduction

The importance of utilizing environmentally friendly natural materials nowadays is widely considered worldwide. The environment is currently affected by human activities that cause environmental losses, mainly from processed agricultural waste, and this attracts researchers to convert the waste into composite reinforcement materials containing natural fibers that are environmentally friendly, easy to obtain, and inexpensive [1–3].

Indonesia is a country that has abundant natural wealth. Many natural fibers have not been utilized optimally, one of which is fiber from the banana plant. Almost all agricultural land owned by the community is planted with banana plants. This plant leaves waste in the form of banana stems after the harvest process. Indonesian people generally use fiber from banana stems as a binder.

BSFs are composed of natural fibers derived from banana trees. This material contains high lignin, hemicellulose, and cellulose, so it is widely used by the community as a rigging material because it has unique properties, namely, as a binding material used before the synthetic binder used today. Therefore, banana stem fiber becomes an interesting object.
of experimental study, including examination of several parameters, such as the physical, chemical, and mechanical properties of the fiber [4–6].

Multiple methods have been utilized to increase natural fiber as a composite filler material, such as soaking with NaOH, KMnO₄, H₂O₂, seawater, heating with turmeric, liquid smoke, silane, fumigation, and exposing the micromechanical characteristics of the fiber, such as its physical, chemical and mechanical properties, and by testing and observations such as the single fiber tensile test, SEM, XRD, and FTIR [7–10].

Other fiber processing methods carried out by researchers include the treatment of sago midrib fiber with liquid smoke at a temperature of 40 °C with a treatment time span of 1 to 5 h, which can change the texture and pores of the fiber [11,12]. Treatment of fiber Akaa with turmeric for 3 h, which is the most optimal, has effects on the micro-mechanical properties of the fiber [13]; treatment of king pineapple leaf fiber in a smoke chamber with a temperature of 45 °C effectively increased the physical and mechanical properties of the fiber [14]; treatment of sisal fiber with sodium bicarbonate at a temperature of 40 °C provided an effective improvement in the micro-mechanical properties of the fiber [15]. Other studies have considered changing the morphology and increasing the tensile strength of single fibers [8], changing the thermal properties, and changing the properties of the fiber by means of chemistry [16]. Treatment of king pineapple fiber with liquid smoke increased morphology and tensile strength of single fibers, and prompted changes in functional groups [17]. Treatment of coconut belt fiber with liquid smoke can change the tensile strength and the morphology of the fiber [18].

Treating king pineapple fiber by fumigation increases the morphology of the tensile strength of the single fiber [19,20]. The fumigation of king pineapple leaf fibers affects the contact angle of less than 30° and increases the interlocking capability of the fiber matrix. The interfacial shear stress increases by 282.8% with smoking for 15 h [20,21]. The increase in the strength of the fiber reinforcing composite is due to seawater treatment [22,23]. The morphology and surface roughness of the fibers increased during immersion, strengthening the fiber-matrix bond [24,25]. Fiber will increase mechanical properties due to turmeric treatment [13]. Overall, 4% acetylation treatment is optimum for tensile properties’ enhancement for most of the natural fibers evaluated [26].

Fiber treatment with NaOH increased density, and strengthened fiber’s mechanical properties, with changes in O-H and C-H groups at 3330 cm⁻¹ and 2918 cm⁻¹, reduced lignin, more excellent thermal stability, and a rougher surface after treatment [27]. Corn husk fiber treated with NaOH showed better mechanical properties than fiberglass [28]. Fiber with NaOH treatment changed the mechanical properties for the better, and the absorption increased due to changes in the roughness and lumen structure [29]. Coir fiber’s undergoing NaOH treatment changes the surface of the fiber, which is much cleaner and rougher, improves the tensile and flexural properties of the composite, increases the interfacial adhesion of the fiber and matrix, and effectively increases the compressive strength, flexural strength, and toughness [30]. The alkaline effect is evident from the changes seen on the surface of the Abaca fibers [31]. Pandanus tectorius fiber with alkaline treatment can strengthen the fiber, hence the pandanus tectorius fiber-reinforced composite is an environmentally friendly fiber alternative [32]. The H₂O₂/CH₃COOH and HNO₃ treatment removed lignin, pectin, waxes and also increased cellulose crystallinity in the fibers, especially for HNO₃ treatment [33]. Alkaline treatment of pineapple leaf fiber increases the crystalline properties of the fiber [34].

Chemical treatment harms the environment, so environmentally friendly treatment materials are needed, such as the use of liquid smoke [35], where liquid smoke is a pyrolysis process for lignocellulosic materials and an environmentally friendly antibacterial [36,37]. The use of liquid smoke has the potential as a natural fiber treatment material that protects fibers from bacteria and preserves and improves the micro-mechanical properties of natural fibers.

Many processing materials are not environmentally friendly, in which case environmentally friendly processing materials such as liquid smoke are needed [12]. Liquid smoke contains phenolic compounds, carbonyl compounds, and acids that change the fiber’s
chemical, physical and mechanical properties. Therefore, it is necessary to research natural fibers, especially BSFs, to determine the effect of liquid smoke immersion treatment on physical (morphological), chemical (crystal index and functional groups), and mechanical properties (single fiber tensile strength); BSFs emerge as a composite reinforcement, lightweight and environmentally friendly.

2. Materials and Methods

2.1. Material and Treatment Methods

The material used is gray BSFs, and the treatment method consists of two methods, including the treatment of fiber immersion with grade three liquid smoke with various immersion times of 1, 2, or 3 h, and without treatment. After that, the fiber was dried in a Memmert oven UN 55 Cap 53L at 40 °C for 30 min. The notation of the treatment method can be seen in Table 1.

<table>
<thead>
<tr>
<th>No</th>
<th>Notation</th>
<th>Code</th>
<th>Treatment</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Without treatment</td>
<td>TP</td>
<td>Without</td>
</tr>
<tr>
<td>2</td>
<td>1 h treatment</td>
<td>P1J</td>
<td>Liquid smoke grade (III)</td>
</tr>
<tr>
<td>3</td>
<td>2 h treatment</td>
<td>P2J</td>
<td>Liquid smoke grade (III)</td>
</tr>
<tr>
<td>4</td>
<td>3 h treatment</td>
<td>P3J</td>
<td>Liquid smoke grade (III)</td>
</tr>
</tbody>
</table>

Table 1 shows treatment notations of the BSF. TP is the notation of BSF without treatment while P1J, P2J, and P3J are the notation of liquid smoke grade 3 treatment by 1, 2, and 3 h.

Two stages of fiber treatment were conducted, namely, immersion and heating process. The immersion process of the fiber with liquid smoke is a compounding process between the lignin compounds in the fiber and the acid in the liquid smoke so that liquid smoke fiber compounds are formed. The next stage is the heating process for 30 min at a temperature of 40 °C to facilitate the decomposition of lignin compounds and the degradation of H₂O compounds. Figure 1 shows the reaction process of banana stem fiber with liquid smoke for immersion and heating:

![Figure 1. The reaction process of banana stem fiber with liquid smoke.](attachment:image.png)

The reaction process above causes an increase in the tensile strength of the fiber, a change to the crystalline nature, the functional group properties change to C-C bonds, and...
the morphological properties of the fiber become coarser and more porous. Changes in these properties are needed for the use of fiber in composite reinforcement because new fibers can be used as composites not only to be strong but also to have coarse, porous, crystalline morphology and changes in functional groups. This change in properties will make banana stem fiber potentially an environmentally friendly composite reinforcement.

2.2. Testing and Observation

Before testing, the specimen was made according to ASTM 3379-0 standard and tested with a single fiber tensile tester with a capacity of 50 N. For morphological observations, the coating process was carried out and then observed with the JEOL JCM 6000 SEM tool. In the crystalline and functional group tests, the fibers were shaped into folders and then tested with XRD type Rigaku mini flex ll and FTIR type Shimadzu prestige-21 model 8400S tolls.

2.3. Single Fiber Tensile Test

In the single fiber testing stage, the specimen is made according to the ASTM 3379-02 standard, as shown in Figure 2. Then the diameter of the specimen is measured, and a tensile test obtains the tensile strength of the single banana stem fiber.

![Figure 2. A tensile test specimen ASTM 3379-02.](image)

2.4. SEM Observation

The SEM observation test with the JEOL JCM 6000 SEM tool attempts to see the morphology of the coated test material by placing the test object on the preparation. The SEM test equipment is operated and observed until the surface of the test object is visible, which is then photographed and stored.

2.5. FTIR and XRD Test

In the test of crystals and functional groups, the fibers were formed into powder and then tested with Rigaku mini flex ll type XRD and Shimadzu prestige-21 model 8400S type FTIR. The result of this test is a graph that describes whether the fiber is amorphous or crystalline and that identifies the functional groups of the fiber.

3. Results and Discussion

3.1. Effect of Liquid Smoke Treatment on Tensile Strength of Single Fiber

The study of BSF treatment with liquid smoke showed that the micromechanical properties of BSF were increased, and there was a tendency of fiber strength to be different without and with treatment. The tensile strength of TP fiber was 148.54 MPa, while P1J, P2J, and P3J increased this to 219.19 MPa, 264.21 MPa, and 239.46 MPa. The percentage change in the tensile strength of the fiber compared to TP fiber for P1J, P2J, and P3J was 32.23%, 43.78%, and 37.97%. A significant increase occurred in P2J, which was 43.78%. This change is influenced by changes in fiber composition, whereas in previous studies conducted by [8,17], the changes in fiber composition could increase fiber strength.
Figure 3 shows that BSFs with 2 h of treatment have a higher tensile strength per single fiber, where the shape of the cross-section does not break as brittle or change the shape of the fiber before breaking, as shown in Figure 4. Following previous research conducted by [16,17], fiber treatment with liquid smoke can form strong C-C elemental bonds caused by the H₂O degradation process in BSFs, hence carbon atoms (C) are dense, and in conditions of excessive H₂O degradation, the fiber strength will become brittle and the liquid smoke can increase the tensile strength of the fiber. Based on these data, the tensile strength of the fiber with treatment for 2 h with grade three liquid smoke is the most significant, and in line with previous research, which stated that the treatment of liquid smoke on the fiber increased the tensile strength of a single fiber [8,17].

Figure 3. Single fiber strength test (TP, P1J, P2J, and P3J).

Figure 4. The surface of BSFs with SEM Photo of TP, P1J, P2J and P3J.
3.2. Effect of Liquid Smoke Treatment on Fiber Morphology

The surface shape of BSFs from SEM observations is shown in Figure 4. Figure 4a is the morphology without treatment, Figure 4b–d is the morphology of the fiber with treatment.

In addition, there is also a difference in the cross-sectional pattern of breaking during the single-fiber tensile test where the BSFs fiber at TP, P1J, and P3J shows a brittle fracture pattern, while the BSFs fiber at P2J shows that the cross-sectional surface is not brittle.

The result shows that liquid smoke affects changes in tensile strength and morphology and improves the micromechanical properties of the fiber so that the fiber treated with liquid smoke for 2 h is the most significantly improved. Previous studies stated that the fiber treated with liquid smoke increased the tensile strength of the single fiber and changed the morphology of the fiber [8,11,17], hence it can be used as a composite reinforcement.

3.3. Effect of Liquid Smoke Treatment on Fiber Crystalline Properties

The XRD test was conducted to see the effect of liquid smoke treatment on the percentage of compound content, peak crystals, and intensity of BSFs. The XRD test generates graphic information on different treatments of BSFs, as shown in Table 2.

Table 2. Percentage of compound changes in fiber.

<table>
<thead>
<tr>
<th>No</th>
<th>Compounds</th>
<th>TP (%)</th>
<th>P1J (%)</th>
<th>P2J (%)</th>
<th>P3J (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>SiO$_2$</td>
<td>87</td>
<td>47</td>
<td>90</td>
<td>48</td>
</tr>
<tr>
<td>2</td>
<td>C$_{12}$H$_8$</td>
<td>7.5</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>3</td>
<td>H$_2$O</td>
<td>5.1</td>
<td>53</td>
<td>10</td>
<td>52</td>
</tr>
<tr>
<td>4</td>
<td>Unknown</td>
<td>0.4</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 2 shows the differences in compounds between the untreated fiber and the treated fiber for 1, 2, and 3 h. Where in TP, there are compounds (SiO$_2$ 87%, C$_{12}$H$_8$ 7.5%, and H$_2$O 5.1%), in P1J, there are compounds (SiO$_2$ 47% and H$_2$O 53%). In P2J, there are compounds (SiO$_2$ 90% and H$_2$O 10%), and P3J contains compounds (SiO$_2$ 48% and H$_2$O 52%). The location points of the different compounds are shown in Figure 5.

Figure 5 shows the percentage of C$_{12}$H$_8$ compounds found in TP fibers. On the other hand, P1J, P2J, and P3J fibers do not contain C$_{12}$H$_8$ compounds, which theoretically
means that the liquid smoke fiber compounds change after treatment. Furthermore, H$_2$O compounds are found in all fibers, as related to the results of tensile and morphological tests, where the water content is minor in P2J fiber. This result proves that the theoretical concept of the treatment method is original, and the H$_2$O degradation process occurs in the fiber so that the remaining percentage is only 10%, and confirms that the highest fiber crystalline index occurs in P2J fiber and in line with previous research on the effect of liquid smoke on the crystalline properties of fibers [8,17].

XRD test results also showed that the intensity of the crystals in the fiber experienced a change where P1J, P2J, and P3J fibers increased compared to TP fibers, as shown in Table 3 and Figure 6.

Table 3. Peak list 2-theta(deg) and crystal intensity on Banana Stem Fiber.

<table>
<thead>
<tr>
<th>No</th>
<th>TP</th>
<th>2-Theta(deg)</th>
<th>Int. I(cps deg)</th>
<th>P1J</th>
<th>2-Theta(deg)</th>
<th>Int. I(cps deg)</th>
<th>P2J</th>
<th>2-Theta(deg)</th>
<th>Int. I(cps deg)</th>
<th>P3J</th>
<th>2-Theta(deg)</th>
<th>Int. I(cps deg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>15.65(7)</td>
<td>7267(156)</td>
<td>15.60(5)</td>
<td>373(109)</td>
<td>15.82(5)</td>
<td>7884(302)</td>
<td>15.76(13)</td>
<td>4110(1348)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>22.09(5)</td>
<td>9074(111)</td>
<td>21.20(4)</td>
<td>489(420)</td>
<td>21.63(9)</td>
<td>7351(4626)</td>
<td>21.85(5)</td>
<td>7510(703)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>26.72(3)</td>
<td>86(9)</td>
<td>22.21(6)</td>
<td>525(401)</td>
<td>22.28(8)</td>
<td>8134(269)</td>
<td>23.30(7)</td>
<td>4435(1797)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>34.80(3)</td>
<td>665(39)</td>
<td>26.96(4)</td>
<td>247(20)</td>
<td>26.72(13)</td>
<td>271(17)</td>
<td>26.77(16)</td>
<td>122(16)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>28.91(6)</td>
<td>68(15)</td>
<td>28.91(6)</td>
<td>68(15)</td>
<td>28.57(10)</td>
<td>1387(68)</td>
<td>28.71(2)</td>
<td>101(6)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>35.56(6)</td>
<td>95(14)</td>
<td>29.53(16)</td>
<td>21(7)</td>
<td>35.43(3)</td>
<td>149(11)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>47.04(15)</td>
<td>390(38)</td>
<td>35.26(7)</td>
<td>65(127)</td>
<td>52.06(5)</td>
<td>93(5)</td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
</tbody>
</table>

Figure 6. The intensity comparison of the BSF XRD test (TP, P1J, P2J, and P3J).

Table 3 shows that TP fiber only formed 4 peaks 2-theta, and P1J, P2J, and P3J fibers formed 7 peaks 2-theta, and additionally shows that the fiber treated with third-grade liquid smoke formed crystalline on the fiber. The crystal peak points are shown in the following graph:

Figure 6 shows the fiber crystallization index increased due to changes in fiber structure, where the highest peak TP BSFs occur at point two, while the highest peak BSFs P1J, P2J, and P3J occur at points two and three, respectively. This result proves that the innovation of BSFs treatment with liquid smoke can change the crystalline properties of fibers, so it has become the latest innovative method in changing the crystalline properties of fibers.
The XRD test also confirmed the suitability of the morphological data, where the crystalline level in the treated fiber increased and the fiber morphology also became more coarse and porous, as shown in Figure 7.

![XRD Test Results](image)

**Figure 7.** Intensity comparison of the XRD test of BSFs (TP, P1J, P2J, and P3J).

The changing trend of TP, P1J, P2J, and P3J fibers increased the crystallization index in the fiber along with treatment time. A longer treatment time makes the fiber crystalline index higher. The fiber crystallization index increases due to changes in the structure of the fiber, and it can change the micromechanics properties of the fiber. SEM results in Figure 4 can be analyzed for crystal size using ImageJ software as shown in Figure 8:

![Average Crystal Size](image)

**Figure 8.** Average Crystal Size of BSFs (TP, P1J, P2J, and P3J).

From Figure 8, it can be seen that there was a change in the crystal size of the treated fiber when compared to the crystal size of TP fiber; the largest crystal size occurred in P1J.
while the lowest occurred in P2J, this indicates that P2J experienced the most significant change in crystal size, reaching 34.97%, as in previous studies, which stated that the effect of fiber treatment with liquid smoke had an impact on the crystalline properties of the fiber [8,17]. Changes in the crystal size of the fiber impact when the fiber becomes a composite reinforcement, given that the composite becomes stronger.

3.4. Effect of Liquid Smoke Treatment on Fiber Functional Groups

Changes in fiber functional groups with liquid smoke treatment can be analyzed by performing FTIR testing. Figure 9 shows the result of fiber FTIR testing on TP, P1J, P2J, and P3J.

The graph of the FTIR test results shows differences in the composition of fiber compounds where the functional groups O-H, C-H, C=O, C=C, and CH₃ look different; this indicates that the treatment of fiber with liquid smoke affects the change in wave crest. Figure 10 is a combination of the FTIR BSFs transmittance pattern TP, P1J, P2J, and P3J, where the shape of the peak on the graph changes due to the liquid smoke treatment. The constituent molecules in the FTIR Spectrum show that the wave peaks numbered from 2500–4000 cm⁻¹ are in the O-H and C-H spectra and caused a significant addition of P2J BSFs composition. The peak wave number 1500–2000 cm⁻¹ is in the spectrum C=O and C=C, which indicates that the BSFs compound increased in intensity after treatment, and the compound content was more significant than BSFs without treatment. Therefore the liquid smoke can add C=O and C=C compounds to the chain of BSFs compounds such as lignin and hemicellulose, and the high carbon composition of BSFs can increase their strength of BSFs. While at the peak of the wave number 500–1500 cm⁻¹ is in the CH₃ spectrum, which indicates the addition of CH₃ composition, P2J BSFs become the most significant increase in functional groups.

Figure 9. FTIR test of BSFs (TP, P1J, P2J, and P3J).
Figure 10. Transmittance comparison of FTIR test (TP, P1J, P2J, and P3J).

As the result of liquid smoke treatment from this study, P2J fiber was the most optimal fiber with a significant increase in tensile strength, crystalline intensity, functional groups of BSFs, and changes in morphology. This result is in line with previous research, which states that liquid smoke can change the functional groups of fiber [8,17]. Therefore, BSF fiber with P2J liquid smoke treatment becomes new knowledge about the characteristics and properties of BSF and the latter becomes one of the solutions as lightweight and environmentally friendly composite reinforcement for the future.

4. Conclusions

Treatment of BSF with liquid smoke formed a fiber morphology with an elongated, porous pattern, and the tensile strength of single fibers increased, changed the crystalline properties, and changed the functional groups. P2J fiber is a fiber that shows morphological changes that look coarser, more porous, and there is an elongated pattern. The fiber also has a significant increase in strength of 264.21 MPa and increased tensile strength by 43.78% compared to TP fiber. The crystal intensity also increases by 34, 97% of the TP fiber, and a change of functional groups occurs in the fiber. Thus, liquid smoke is an essential ingredient for BSF treatment by changing the physical, mechanical, and chemical properties, making for lightweight and environmentally friendly options for composite reinforcement in the future.

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References


20. Palungan, M.B.; Soenoko, R.; Gapsari, F. The effect of king pineapple leaf fiber (Agave Cantala Roxb) fumigated toward the fiber wettability and the matrix epoxy interlocking ability. *EnvironmentAsia* 2019, 12, 129–139. [CrossRef]


33. Soatthiyanon, N.; Crosky, A. Characterisation of Elementary Kenaf Fibres Extracted Using HNO3 and H2O2/CH3COOH. Fibers 2022, 10, 63. [CrossRef]