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Effect of liquid smoke on surface morphology and tensile strength of Sago Fiber

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ABSTRACT

This study aims to identify the effect of liquid smoke treatment on surface morphology and tensile strength of sago fiber (SF), including chemical reactions during the treatment. The proposed study is divided into two steps, fiber treatment and property tests. The first treatment, SF was immersion in the liquid smoke solution for 1, 2, 3, 4, and 5 hours, then dried for 1 hour which then characterized SEM, universal machine testing machine, X-Ray diffractometer (XRD), and Fourier transform infrared spectrometer (FTIR). The acetic acid in liquid smoke reacts with fiber to form fiber-liquid and H2O compounds. The heating process degrades H2O content in fibers and decomposes C and C elements to close together forming strong chemical bonds so that the fiber morphology become rough, porous, grooved and increasing the tensile strength of the fiber. But excessive heating treatment makes fibers more fragile because the H2O elements degrade too low. Dominant percentage of SF crystallization occurred after 4 and 5 hours, 64.7 and 66.9 %, respectively among other durations, a significant increase of tensile strength 50.813 MPa was achieved by 1-hour immersion in liquid smoke. Hence, as alternative to improve the mechanical characteristics of SF, immersion in liquid smoke should be applied. Furthermore, studies of similar treatment on other natural fibers can also be considered in the future.

Keywords: Liquid smoke; surface morphology; tensile strength; Sago Fiber.

INTRODUCTION

In recent years, natural fibers have been widely studied by experts as industrial materials such as mengkuang fibers, palm fibers, akaa (*corypha*) fibers, king pineapple leaf fibers, coconut fibers, carica papaya fibers, albizia amara fibers, and juncus effuses L fibers [1-9]. This study discusses the treatment of Sago fiber (SF) using acetic acid in liquid smoke from coconut shell which is more eco-friendly than other alkali treatment. SF is one bio material that has its own uniqueness and also interesting because this material is obtained from midrib of sago trees which is widely found in tropical nations. Since hundreds of years ago, SF has been used as raw material for rope in Luwu regency of South Sulawesi and some other parts of eastern Indonesia. From the best of authors' knowledge, yet no report of SF utilization as engineering material. As its vast and potential source, utilization of SF may not be limited only as traditional ropes but also as composite reinforcing material. SF is relatively cheap, biodegradable, renewable resources, easy handling, and above all, it is environmental friendly.

The chemical composition of SF depends on few unavoidable factors, such as weathers, soil, plant origin, etc. [10,11]. However, it constantly consists of the main component of lignocelluloses (cellulose, hemicelluloses, and lignin). The basic unit of lignocelluloses macromolecules is hydroxyl group (-OH). This hydroxyl forms an intra-molecular hydrogen bond and an inter-molecular and a hydrogen bond with a hydroxyl group of moisture. The major disadvantages of SF as composite materials are its hydrophilic characteristics and water contents, which reduce durability. Modification of SF surfaces to reduce its hydrophilic is possibly improving adhesion properties and increase the hydrophobic part of the fiber surface. These modifications can make it possible to change the orientation of the fiber into an alternative composite amplifier in the future.

Previous studies attempted various treatments on natural fibers including immersion in solutions (NaOH, $KMnO_4$, H_2O_2, and seawater), heating with turmeric solution, and steaming [2, 3, 6, 10-13]. The treatments had altered physical properties of fibers, such as lignin level reduction from coconut husk by immersion in NaOH, cleared and shaped grooves by immersion in $KMnO_4$ and H_2O_2 [3]. Treatment with NaOH removed hemicelluloses and lignin in natural fibers and decreased the moisture content [11]. Seawater immersion of straw fibers also degrades lignin layer while providing harder surfaces [5,12,14]. Heating treatment of akaa (*corypha*) midrib fiber with turmeric solution increased the level of cellulose compound and reduced lignin content, increasing fiber's strength [6]. Steaming of king pineapple leaves had a very significant effect on fiber surface morphology to became harder and grooved surfaces [15]. After fumigation, surface morphology of King pineapple leaf fibers (KPLF) was rough and grooved, fiber density increased, and the single fiber tensile strength was notably increased at the base of the leaf [4]. Chemical treatments are considered in modifying fiber surface properties [16]. When natural fibers soaked in alkali (NaOH 5%), most of the wax and other non-cellulosic substances on the surface were removed, create a cleaner, rough, and porous surfaces due to the peeled outer layer as the chemical components of fiber were found dissolved in alkali [17].

FTIR and XRD tests had complement some previous studies on natural fiber treatments. For example, akaa (*corypha*) midrid with turmeric solution treatment greatly affect the ability of C=C and O=H absorption[6],the lignin and hemicelluloses content in the KPLF (King pineapple leaf fibers) were reduced after fumigation [4]. In an XRD test of sugar palm fiber, a consistent emergence phenomenon of crystal peak diffraction occurred as reaction to seawater [5]. Chemical treatment leads to crystallization on the surface of the fiber, where alkali treatment improves the crystallinity and exhibits new chemical bond formation in the sponge-gourd fiber (SGF) [18].

From various treatments of natural fibers so far, just a few are considered as environmental friendly. Environmental friendly material such as liquid smoke is a unique material and potentially used for fiber treatment. Liquid smoke is product of pyrolysis process of natural substances from coconut shell supplies, reasonably cheap, and degraded naturally. It has similar content to other natural fibers consist of lignin, hemicelluloses, and cellulose. Liquid smoke has also potential uses as an all-natural antimicrobial [19, 20]. In current applications, liquid smoke is utilized as natural preservatives due to its content of phenol and acid compounds, functioned as antibacterial, and antioxidant [21]. The main products of wood pyrolysis are phenols, carbonyls, and organic acids which are responsible for the flavor, color, and antimicrobial properties of liquid smoke [20]. The compounds of liquid smoke have hydrophilic properties due to their OH group molecules, classified into the polar compound with negative electrons; therefore easily bind to fiber lignin to form another compound.

Through extensive experiments of this study, mechanical characteristics of SF can be analyzed to reveal its potential as natural fiber for green composite applications. Alterations of tensile strength of SF specimen will be clarified by chemical reactions of C-O-H elements during immersion in liquid smoke. It is expected to have clear understanding on the correlation of liquid smoke treatment and the alteration of tensile strength of SF, including its potential as green fiber alternative.

METHODS AND MATERIALS

Materials

The raw materials of this research are Sago Fiber (SF) from Luwu regency, South Sulawesi province, Indonesia and liquid smoke made of coconut shells.

Table 1. Chemical components of liquid smoke.

No	Component	Percentage
1	L-Alanine, ethyl ester, 2-propanamine, 1-methoxy 2-Pentanamine	33.37
2	Acetic acid, methyl ester	07.39
3	2,3-Butanedione, Acetic acid ethyl ester	02.60
4	Acetic acid	56.65

Liquid smoke treatments

SF were separated from midrib leaf and cambium; then be immersed in the liquid smoke solution for certain durations, as shown in Table 2.

Table 2. Study notation.

No	Treatment	Notation	Remarks
1	No Treatment	TP	No immersion, no drying
2	1 hour immersion	P1J	All dried at 43^0C for 1 hour
3	2 hours immersion	P2J	
4	3 hours immersion	P3J	
5	4 hours immersion	P4J	
6	5 hours immersion	P5J	

Groups of the specimen were treated and coded as listed in Table 1. After being immersed, fibers were dried in an oven at temperature 43^0C for one hour and then took out for slow cooling at room temperature.

Morphology of SF

The morphology of fiber surface of each group then is observed using electron microscope Vega 3 Tescan Scanning Electron Microscope (SEM) at the 5kV voltage.

X-Ray Diffraction (XRD) of SF

Rigaku Miniflex ll was used to get X-Ray diffraction pattern (XRD). In this machine, a high-voltage supply (30 kV, 15 mA) is used to produce X-ray radiation. The specimens were step-wise scanned by the XRD over the operational range of scattering angle (2θ) between 10^0 and 60^0 by steps of 0.02^0. Percentage crystallinity (%Cr) was calculated as follow:

$$\%\mathrm{Cr} = \frac{I_{10}}{I_{10}+I_{60}}\ x\ 100 \tag{1}$$

I_{10} and I_{60} are the crystallines amorphous intensities at 2θ scale close to 10^0 to 60^0, respectively.

Fourier Transforms Infrared Spectroscopy of SF

Fourier Transform Infrared (FTIR) spectra of the samples were recorded at room temperature using Shimadzu prestige-21 model 8400S in the wave number range of 250-4000 cm^{-1}. For these measurements, the samples were crushed for recording the FTIR spectra in the transmittance (%) mode.

Tensile Testing of SF

Preparation of SF specimen is started by separation of fibers off the midrib leaf and cambium then grouped in of type of treatments, coded (Table 2) and isolated as single fibers based on ASTM D3379-75 standard. Codes of treatments (TP, P1J, P2J, P3J, P4J, and P5J) were labeled to each group of the 5 treatments. Total 25 specimens then underwent the tensile strength (σ) tests by Universal Material Testing Machine type LR10K.

RESULTS AND DISCUSSION

Morphology Observation

Figure 1 shows surface morphology of SF. Figure 1(a) is the untreated fiber surface, while Figure1(b) to (f) indicate surfaces of SF with liquid smoke treatments, resulted in grooves, porous, and increased rough of the fiber surface.

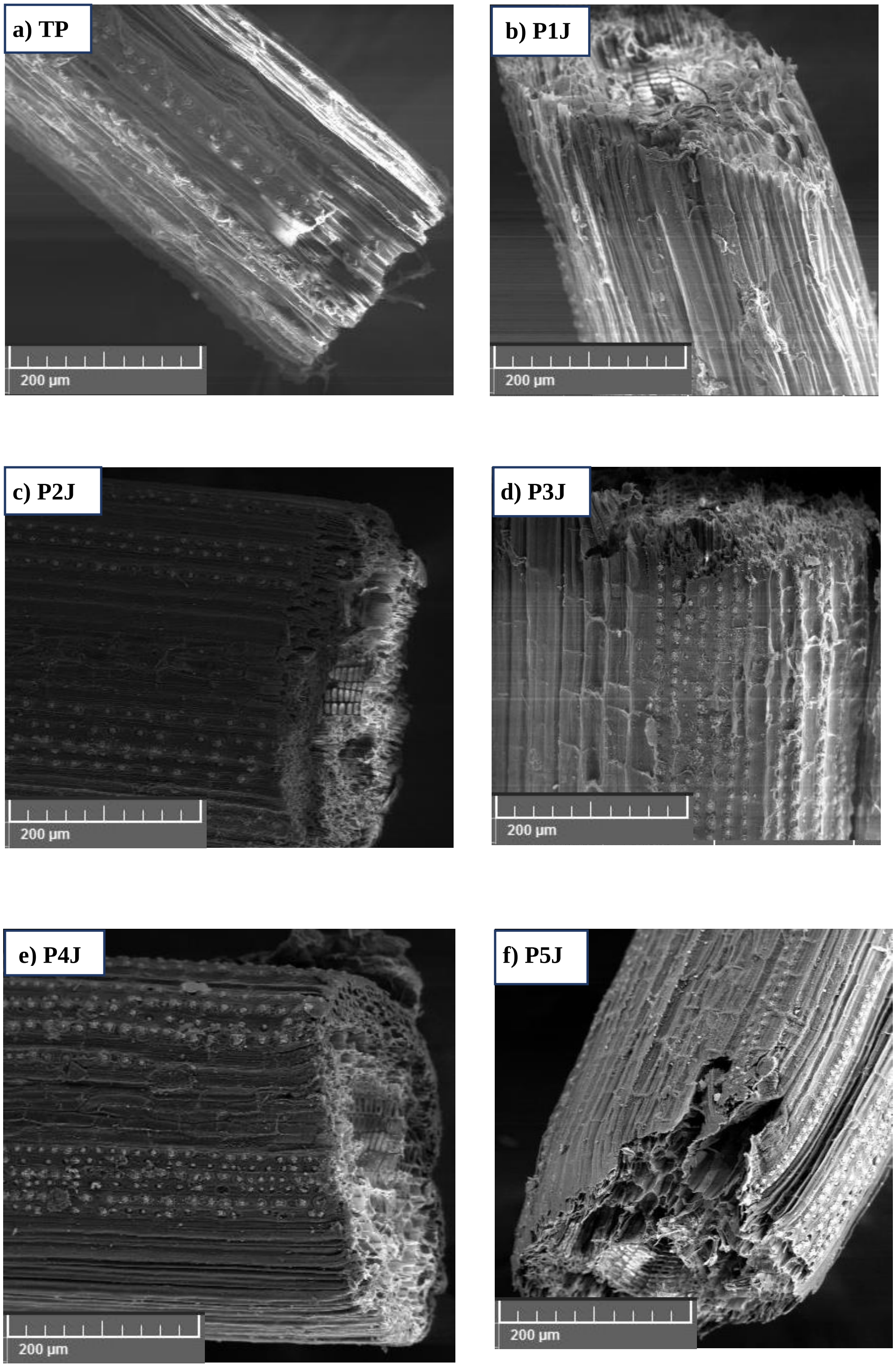


Figure 1. Surface morphologies of specimen with: (a) no treatment, (b) 1 hour immersion, (c) 2 hours immersion, (d) 3 hours immersion, (e) 4 hours immersion, and (f) 5 hours immersion.

Figure 1(a) shows the surface of SF with no treatment is fairly smooth. This indicates that SF surface with lignin content is still abundant aligning on fibers surface. The figure shows the regular rectangular pattern and which is smooth on the surface of the fiber. However, after being treated with liquid smoke for 1, 2, 3, 4, and 5 hours, as shown in Figures. 1(b-f), respectively, the rectangular patterns of the surfaces are gradually rougher compared to the untreated one. The roughness depends on the duration of the liquid smoke treatments.

Figure 1(b) of 1 hour treatment shows a new pattern on the surface of SF. The surface starts to change into groove. After 2 hours immersion, the rectangular groove patterns become more arranged, visibly rough, and porous as shown in Figure 1(c). The morphology of the fiber surface with 3 hours of immersion indicates a rectangular, rough, and pore groove patterns and increased as shown in Figure 1(d).

Figure 1(e-f) show rougher patterns and clearer pores during the 4 and 5 hours treatments. In these cases, the plot is clearer, coarser and the pores clearer. Larger grooves, rough and pore patterns are expected to interact with fiber matrix compounds to increase the bond between fiber and matrix.

Figure 1(b – f) denotes a rough surface because the OH group content of the fiber has been degraded, and the surface becomes coarse, grooved, and porous by the effects of liquid smoke. The treatment of liquid smoke makes the surface roughness are distinguished from the fiber surface without treatment. The increased coarseness resulted from growing lignin on fibers surface, enriched by the composition of H and O elements in lignin compound along with the density of C bonds on the fibers, which also found in other studies [8].

Crystalline Structure Observation

In the XRD test results, visible differences between SF curve of TP andP1J, P2J, P3J, P4J, and P5J as shown in Figure 2

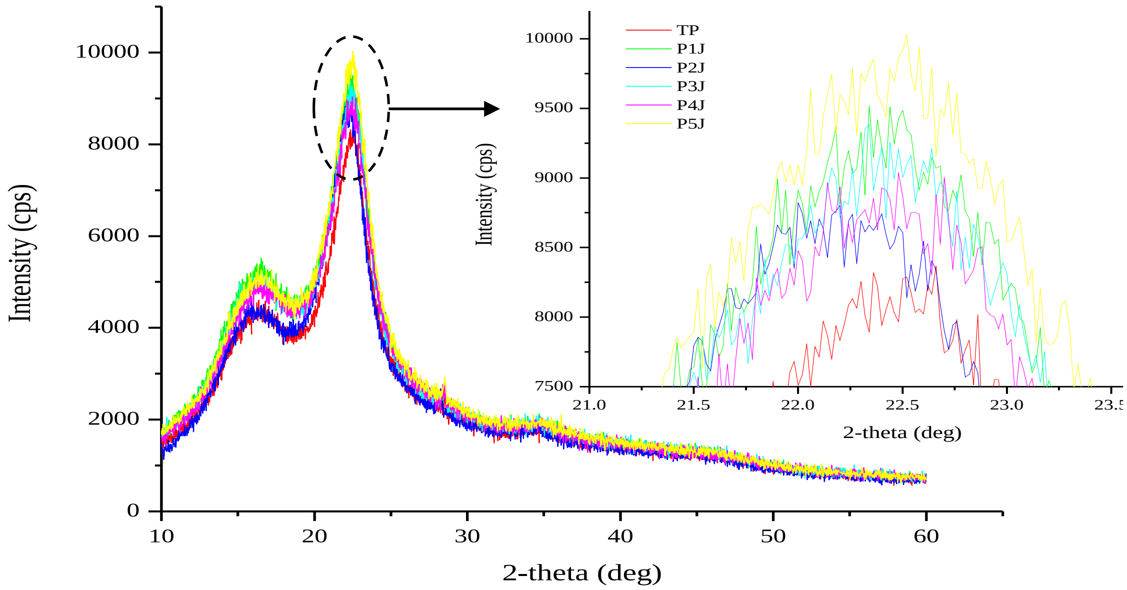


Figure 2. The result of SF for no treatment (TP), 1 hours immersion (P1J), 2 hours immersion (P2J), 3 hours immersion (P3J), 4 hours immersion (P4J), 5 hours immersion (P5J).

It is evident from Table 2 that relative intensities of SF at 2θ scale were 4280 (I_{10}) and 8276.7 (I_{60}). The percentage crystallinity (%Cr) of SF has been found to be 65.9. Whereas SF treated with liquid smoke for 1 hour showed relative intensities of 5203.3 (I_{10}) and 9486.7 (I_{60}). Relative intensities of SF treated with liquid smoke for 2 hours were 4336.7 (I_{10}) and 8690 (I_{60}). The percentage crystallinity of SF treated with 1 hours and 2 hours were 64.5 and 66.7, respectively. The relative intensities of SF treated with 3 hours were 4883.3 (I_{10}) and 9003.3 (I_{60}). The percentage crystallinity of SF was found to be 64.8.Whereas SF treated with liquid smoke for 4 hours showed relative intensities of 4846.7 (I_{10}) and 8913.3 (I_{60}). Relative intensities of SF treated with liquid smoke for 5 hours were 4946.7 (I_{10}) and 10033.3 (I_{60}). The percentage crystallinity of SF treated with 4 hours and 5 hours were 64.7 and 66.9, respectively.

Table 3. The percentage of crystalline of SF specimen.

Notation	**I_{10} (cps)**	**I_{60} (cps)**	**Cr (%)**
TP	4280	8276.7	65.9
P1J	5203.3	9486.7	64.5
P2J	4336.7	8690	66.7
P3J	4883.3	9003.3	64.8
P4J	4846.7	8913.3	64.7
P5J	4946.7	10033.3	66.9

FTIR Structure Analyses

The change of the compound was proved by FTIR (Shimadzu prestige-21 model 8400S). Figure 9 shows the difference of fiber compounds composition before and after treatments. Analysis using FTIR to find out the changes of the functional group of SF compound due to the treatment of liquid smoke in various timeframe. The graph of FTIR transmittance incorporation patterns as shown in the following Figure 3:

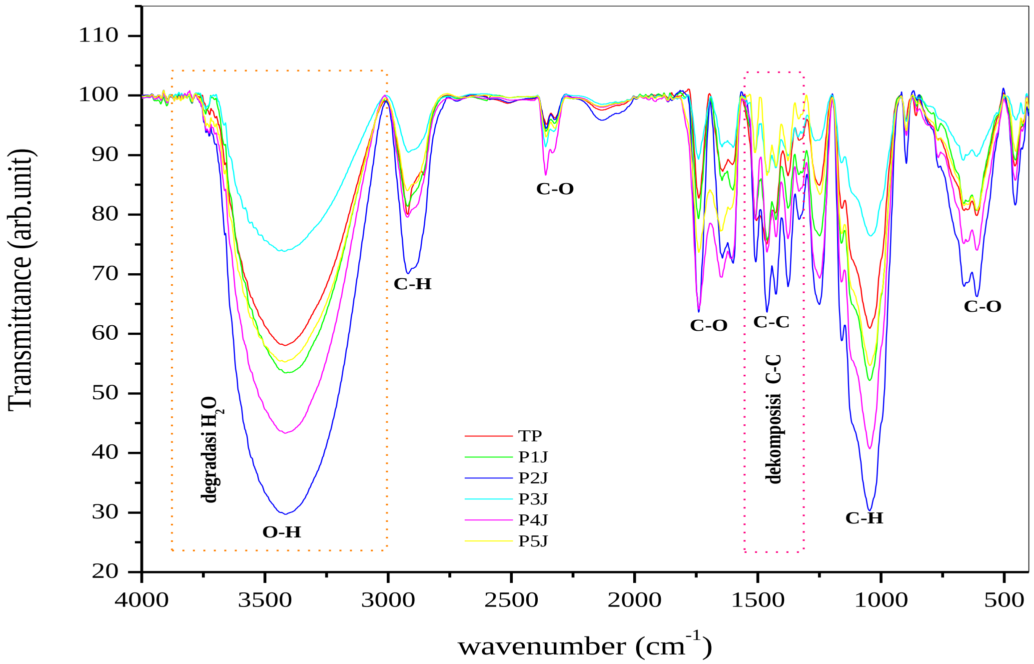


Figure 3. FTIR test results for non-treated (TP) and 1 – 5 hours immersion (P1J-P5J) of SF.

Figure 3 shows the incorporation of the FTIR transmittance patterns of no treatment and with treatment of SF. Where the peak shape changes, explain that treatment with liquid smoke affects the wave crest, wherein the liquid smoke compound acetic acid can break the chain of fiber compounds, an acetic acid was able to break the chain of fiber compounds such as lignin and hemicelluloses [9].

The molecule of SF composed of OH groups with a range of 3000-4000 cm^{-1}, the hydrogen bonding (OH) group in the fiber undergoes changes after treatment. In the range of 2850-3000 cm^{-1} wave range associated with the C=H bonds observed in the alkenes group. The peak of the C=O group in the ketene is observed in the peak range 1715-1765 cm^{-1} and C=O in the carbonyl in the peak range 2000-2500 cm^{-1}. Peak cluster C=C in the range 1450-1650 cm^{-1}.

The degradation process after 1-5 hours treatment process decreased the OH group in the range 3000-4000 cm^{-1} and the decomposition process of C=C group also occurred in the range 1450-1650 cm^{-1}. The change of the OH group on the degraded fiber becomes free released water so that the fiber surface becomes coarse, grooved, and porous where diameter of fiber decreased. Meanwhile, the decomposition of the C=C group forms a tighter bond increasing the tensile strength of the fiber.

The chemical process of immersing the fibers in liquid smoke forms a liquid-fiber-smoke compound. Thus, the formation of the compound will increase the composition of C, H, and O on the fiber compound. Liquid smoke of SF took place as per the following schemes:

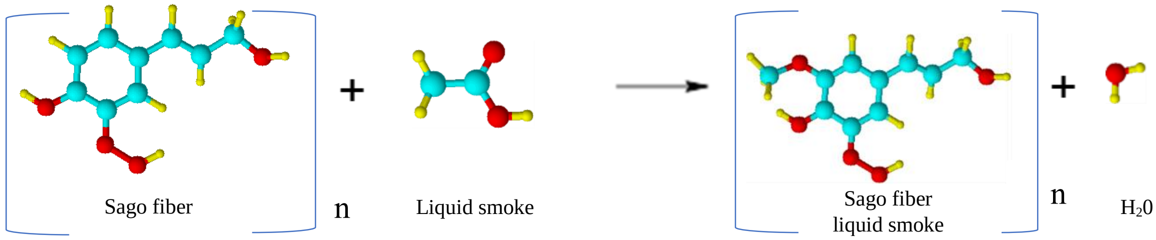


Figure 4. The process of SFand liquid smoke compounds formation.

SF and liquid smoke compounds are compounds containing OH radicals, the polarity of OH group in fiber and liquid smoke molecules can form a relatively strong hydrogen bridge between molecules in the liquid phase. During heating process with temperature 43°C, degradation process occurred between element O and H as H_2O, while C element were decomposed and diameter of fiber becomes smaller, porous, grooved, and coarser on fiber surface. The degradation process on the fiber as sketched in the following figure:

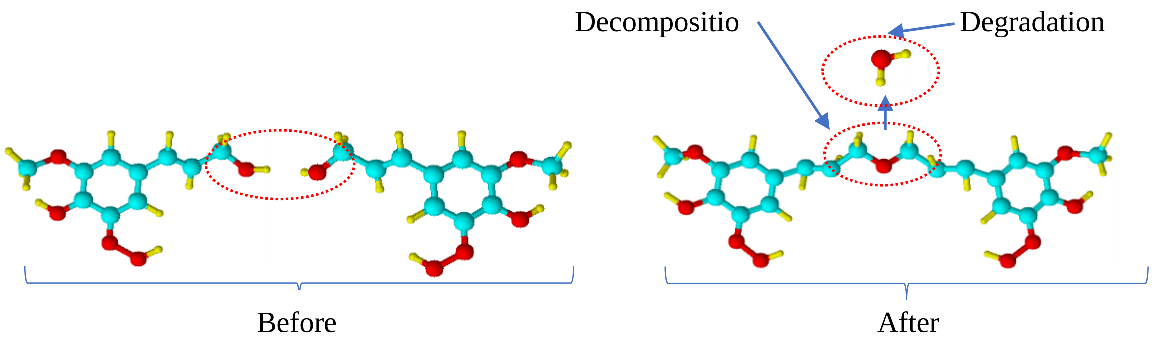
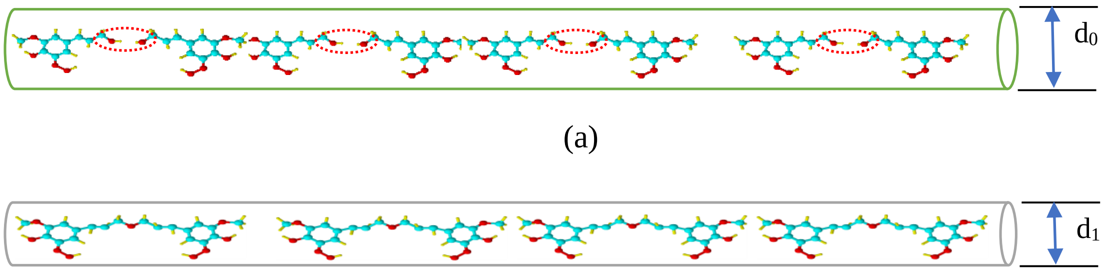


Figure 5. Degradation process of H, O elements and decomposition of C.

The degradation process forms the SF cross-sectional area to become smaller ($A_1 < A_0$). Here, A_0 is the initial cross-sectional area while A_1 is the cross-sectional area after heating, as shown in Figure 6.



(a)

(b)

Figure 6. Changes in cross-sectional area of SF: (a) Fiber diameter before heating (d_0) and (b) Fiber diameter after heating (d_1).

While decomposition of C element on SF compounds occurred, consequently the distances between C compounds become closer, produces stronger interconnection force so that the tensile strength of a single fiber increases as fiber surface becomes rough, porous, and grooved [4][3]. Decomposition can be caused by loss of moistures or due to cellulose and lignin degradations [22], as shown in Figure 7.

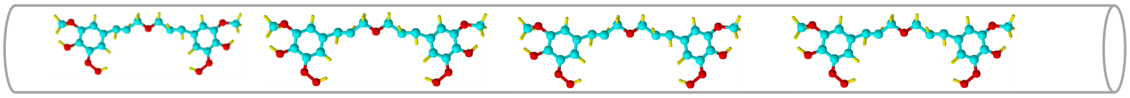


Figure 7. The form of fiber compound due to decomposition.

Tensile Strength Analysis

Tensile strength of natural fibers is important characteristic for bio composite applications, thus certain treatments are required to improve the characteristic. As one of natural fibers, SF samples were treated with liquid smoke and showed significant results on the increasing tensile strength of the SF.

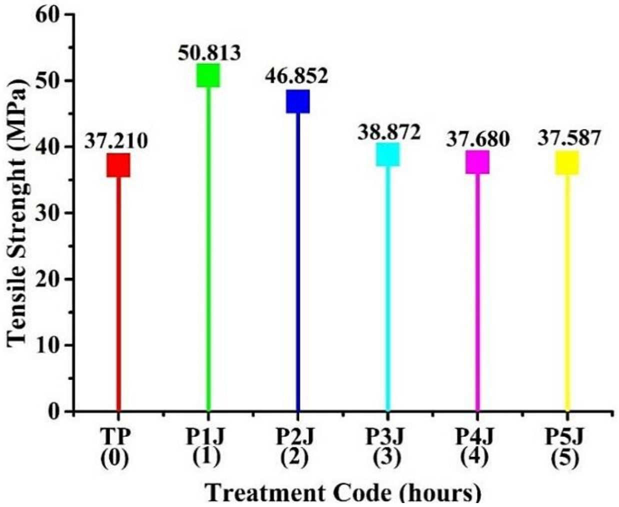


Figure 8. Tensile strengths of SF with no treatment (TP) and 1-5 hours immersion (P1J - P5J).

As shown on Figure 8, in comparison to SF without treatment (TP) by tensile strength of 37.210 MPa, improvement of tensile strengths (Δσ) were occurred to all treated SF, labeled as P1J, P2J, P3J, P4J, and P5J by 50.813, 46.852, 38.872, 37.680 and 37.587 MPa, respectively. The highest tensile strength among these treated SF was achieved by 1 hour immersion in liquid smoke with almost 40% increased strength, from 37.210 to 50.813 MPa. From this point, continuing the immersion to another 2 to 5 hours had degraded the SF tensile strengths, gradually down to only 37.587 MPa. The last treatment of 5 hours immersion (P5J) only managed to improve tensile strength of untreated SF by just 1% higher. After 1 hour treatment, the decreased trends of tensile strengths assume that duration of immersions should determine the level of SF deterioration along with weaker tensile strengths. Hence, this study recommends maximum 1 hour immersion in order to improve the SF tensile strength due to longer durations should alter the tensile strength in a negative state.

CONCLUSIONS

The effects of SF treatment in liquid smoke can be explained as follows:

1. The acetic acid in liquid smoke reacts with fiber in the immersion process to form fiber-liquid and H_2O compounds. Furthermore, the heating process facilitates the degradation process of H_2O content in fibers and makes C and C elements to close together to form strong chemical bonds so that the tensile strength increases and fiber morphology become rough, porous, and grooved.
2. The significant increase of tensile strength of 50.813 MPa was achieved by 1-hour immersion in liquid smoke, so that may be considered as composite enforcement fiber alternative. However, longer than 1 hour treatment should make the fibers become dense and brittle due to degradation of H_2O elements is too excessive.
3. Among other durations, dominant percentage of SF crystallization occurred after 4 and 5 hours, by 64.7 and 66.9 %, respectively.

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