Application of Fiber Made from Bark of Salak (*Salacca Zalacca*) Tree as Reinforcement in Polymer Matrix Composites

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*Although Indonesia is abundant supply of salak (*Salacca zalacca*) fruit, bark of salak is not yet advanced utilized. This work therefore chracterized its fiber and process it into polymer matrix composites. The study was conducted using varied chemical treatments. Delignification using NaOH solution with concentration of 1%, 5%, 10% with 3 hours immersion time. Bleaching treatment was using H₂O₂ media with varied immersion time at 30, 60 and 90 minutes. Tensile tests, impact tests, microscopic image test using scanning electron microscopy (SEM) and chemical composition test using fourier-transform infra-red (FTIR) spectroscopy were carried out in this work. This work showed that optimal delignification treatment can be found when 5% NaOH concentration was used. The corresponding ultimate tensile strength and impact strength were 25.47 MPa and 11.95 kJ/m², respectively. The optimal bleaching treatment was 90 minutes immersion. The results of SEM image analysis showed that the salak midrib fiber composite without treatment has a lousy interface. Meanwhile, fiber with delignification treatment only has reasonable good interface and fiber with delignation treatment followed by bleaching treatment has excellent interface. FTIR test results showed that the salak midrib fiber composite without treatment had a cellulose component although hemicellulose and lignin levels still dominated. The delignification treatment had succesfully broken the lignin-specific C = O bond but still could not eliminated hemicellulose and lignin bond. In comparison, bleaching treatment reduced intensity of OH intensity, CH and CO which are typical hemicellulose and lignin functional groups. Based on the results of the study, salak midrib fiber with delignification chemical treatment using 5% NaOH for 3 hours followed by bleaching treatment using 2% H₂O₂ for 90 minutes was the best treatment.*

Keywords: Composite, Salacca Leaf Fiber, Epoxy, SEM

1. INTRODUCTION

Composites can be defined as the assembly of two or more materials, the final assembly having properties superior to the properties of each of the constituent materials. It consists of reinforcement and matrix materials [1]. Polymer, both thermosetting and thermoplastic polymer, metal and ceramic are widely used as matrix materials in composites. Polyamides, polycarbonates, polyesters, phenolic, polyimides, silicones, polyurethanes and polyepoxides belong to thermosetting polymers. Meanwhile, polypropylene (PP), polyoxymethylene (POM), polyesters (TP), polyether sulfone (PES), polyether ether (PEE), polyether ketone (PEK), polyether ether ketone (PEEK) and polyether imide (PIE) are exemplary thermoplastic polymers. Besides depending on mechanical properties of matrix materials, mechanical properties of composites depend on its reinforcement materials. Natural fiber has been widely developed as reinforcement materials due to its benefits, e.g. high strength to weight ratio, reducing bio waste in environment, nonhazardous, inexpensive agriculture by product and abundant supply [2][3]. For example, due to its superior properties, i.e. low bulk density (90–150 kg/m³), abundant supply, good toughness, weather resistance, unique chemical composition, rice husk has been widely used in many engineering composites, e.g. active carbon, panel boards for household and automotive

applications, reinforcement particles in concrete [4]-[7]. Other natural fibers have also been developed as reinforcement materials in composites, e.g. jute [8], leaf of pineapple [8], hemp [9], sisal [10], etc

Of many agriculture products in Indonesia, salak (*Salacca zalacca*) is Indonesian native fruit and can be widely found in the Island of Java and Sumatra. Currently, only its fruit is used and the rest has not yet been used for daily and engineering application. This work then investigates the application of salak's bark as reinforcement materials in polymer matrix composites. According to Raharjo [11], fiber of salak's bark contains up to 42.54% cellulose, up to 34.35% hemicellulose and up to 28.01% lignin. Levels of cellulose can be used more efficiently by using chemical treatment, i.e. delignification followed by bleaching. Meanwhile, Somesi [12] found that addition of 4% CaCO_3 micro particle effectively improves the strength in composite of salak bark fiber and epoxy matrix. Ariawan *et al.* [13] reported increased density of salak bark fiber as NaOH treatment duration increases. In addition, contact angle between fiber and polymer matrix also decreases with longer duration of NaOH treatment. As a result, this increases the wettability of this composite. This study investigates the influence of NaOH concentration used in the delignification process of bark of salak tree. In addition, mechanical properties of epoxy matrix polymer composite specimens reinforced with bark fiber of salak tree are also investigated.

2. MATERIALS AND METHODS

Preparation of Bark of Salak Tree

This research was conducted in several steps, i.e. preparation of salak bark, fiber treatment, fabrication of composites, testing of properties, data processing and analysis. In the beginning, fiber was extracted from the bark of Salak tree (**Figure 1**). After that, the bark was cleaned to remove any remaining cambium in the fiber. The process was continued by drying the fiber using an oven at 80 °C for approximately 1.5 hours. This was to reduce water content in the fiber.

Many strategies are applied to improve mechanical properties of composites reinforced with natural fiber. Of many strategies, chemical treatments of fiber is applied to increase the strength of interfacial bond which then improves mechanical properties [14]. Exemplary chemical substances used for this treatment are alkaline, silane, peroxide (H_2O_2) and per-manganate [15][16]. For this reason, delignation treatment using NaOH was applied in this study following the drying process. Delignation treatment in current study was varied for 2 conditions, i.e. only delignification, and delignification followed by bleaching using peroxide (H_2O_2). The properties of bark fiber after both treatments were then compared with the fiber without any delignation treatment. All delignation treatments, either followed by bleaching or not, were carried out by immersing the bark fiber into NaOH solution with different concentrations, i.e. NaOH 1% (2 gr NaOH in 200 ml distilled water), 5% (10 gr NaOH in 200 ml distilled water) and 10% (20 gr NaOH in 200 ml distilled water) (**Table 2**). Temperature and immersion duration for all delignation treatments were constant at 30 °C and 3 hours, respectively. As comparison, Ariawan *et al.* [13] used 5% NaOH for delignation treatment of salak bark fiber.

For bark fiber without any bleaching process, the bark fiber was washed after NaOH treatment. For bark fiber with bleaching treatment, bark fiber was then immersed in 2% H_2O_2 solution (10 ml H_2O_2 in 240 ml distilled water), at varied immersion time of 30, 60 and 90 minutes. Temperature was constant at 80 °C for these three immersion duration. After bleaching process, the bark fiber was immersed in distilled water having pH of 7 at 30 °C for 1 hour. After that, the fibers were then dried again using the oven at 80 °C. In the end, Fourier-transform infrared spectroscopy (FTIR) spectroscopy test on the bark fiber was then carried out to investigate the chemical composition of bark fiber from three conditions, i.e. without any treatment, delignation treatment only and delignation treatment followed by bleaching.

Figure 1. Salak (*Salacca zalacca*): a) tree, b) fruit

After that, the fiber for all varied conditions was mixed with epoxy as composites matrix using volume fraction ratio of 40% fiber and 60% epoxy resin. When the mixture was cast into mold of tensile test specimen, fiber has 0 degree orientation to tensile test, i.e. parallel with tensile test direction. Specimen of tensile test and impact test follow ASTM D638 [16] and ASTM D256 [18], respectively. After tensile and impact test were carried out, an interface morphology analysis was carried out using SEM to observe the morphology and interactions between the epoxy matrix on the surface of the bark fiber. Table shows the specimen code

Table 1. Variation of bark fiber composite specimen

Specimen code	Treatment
A	Original bark fiber without delignation and bleaching treatment
B1	Bark fiber with delignation at 1% NaOH solution without any bleaching treatment
B2	Bark fiber with delignation at 5% NaOH solution without any bleaching treatment
B3	Bark fiber with delignation at 10% NaOH solution without any bleaching treatment
C1	Bark fiber with delignation at 5% NaOH solution and bleaching using 2% H ₂ O ₂ for 30 minutes
C2	Bark fiber with delignation at 5% NaOH solution and bleaching using 2% H ₂ O ₂ for 60 minutes
C3	Bark fiber with delignation at 5% NaOH solution and bleaching using 2% H ₂ O ₂ for 90 minutes

3. RESULT AND DISCUSSIONS

Tensile and Impact Test

In general, ultimate tensile strength (UTS) and impact strength of bark fiber after NaOH treatment are higher (specimen B1-B3) than original bark fiber (specimen A). UTS and impact strength of original fiber are 17.00 MPa and 10.72 kJ/m², respectively. UTS after delignation ranges from 22.77 to 25.47 Mpa, i.e. an increase by 33.94% to 49.82% compared than that of the original bark fiber. Meanwhile, impact strength slightly increases by 11% for specimen B2 compared than that of original bark fiber. A discrepancy however can be observed for specimen B3 where both UTS and impact strength decrease when NaOH concentration increase. SEM and FTIR spectroscopy analysis were then carried out to further investigate this circumstance.

For specimen C1, both UTS and impact strength decrease compared than specimen without any bleaching treatment (B2). In general, bleaching process with H₂O₂ solution (specimen C2 and C3) increases both UTS and impact strength compared than specimen without any bleaching treatment (specimen B2). After bleaching treatment, UTS increases for specimen C2 and C3 by 32% and 54%, respectively. Meanwhile, an increase of impact strength can be observed only for specimen C3 where impact strength increases by 15% from 11.95 kJ/m² to 13.77 kJ/m². As immersion duration increases from 60 minutes (specimen C2) to 90 minutes (specimen C3), UTS increases by 17% from 29.98 MPa to 35.09 MPa. CaCO₃ can be added to further improve the strength. Somei [12] reported the highest UTS at 134.49 MPa was obtained for a epoxy matrix composite containing 40 vol% of salak bark fiber and 4 vol% CaCO₃ micro particles.

Table 2 Result of tensile and impact test of salak midrib fiber composite

Specimen code	UTS [MPa]	Impact strength [kJ/m ²]
A	17.00	10.72
B1	22.77	10.71
B2	25.47	11.95
B3	23.15	6.46
C1	15.27	11.46
C2	29.98	10.21
C3	35.09	13.77

SEM Analysis

Scanning electron microscopy (SEM) analysis was carried out to investigate the interface between reinforcement fiber and polymer matrix. When fiber has no delignation nor bleaching treatment, interfacial gap exists (**Figure 2**). This shows not good adhesion between fiber and matrix. Lumen in the structure of fiber exists to maintain the structure of fiber. This prevents more voids formation in the fiber. Lumen presence also informs presence of lignin and hemicellulose in the fibers. Specimen A shows debonding at the interface of

matrix and filler. In summary, presence of lumen, debonding, void and interfacial gap contribute to lower mechanical composites.

After delignation treatment, debonding occurs at the interface between matrix and filler (**Figure 3**). Fiber pull-out can also be observed. The presence of fiber pull-out in specimen B2 indirectly indicates good adhesion between fiber and matrix since failure does not only occur at the interface of matrix and filler but also occurs inside the fiber. In this case, mechanical properties depends on mechanical properties of fiber. Thus, stronger fiber contributes also to higher composites strength. However, micro void still presents which can initiate composite failure and decreases mechanical properties of composite. Meanwhile, lumen and cell walls starts to degrade. This indicates less lignin, hemicellulose and voids in the fiber. This circumstance leads to higher fiber density and better mechanical properties.

Figure 2. SEM analysis on composite without any treatment (specimen A)

Figure 3. SEM analysis on composite after delignification treatment only (specimen B2)

SEM analysis on composite with bleaching treatment shows that fiber pull-out as the main composite failure (**Figure 4**). In addition, good adhesion between fiber and matrix in specimen C3 can be observed. Many previous works, e.g. [16][19], also reported that improved mechanical properties in polymer matrix composites (PMCs) reinforced with natural fiber are due to removal of hemicellulose, pectin and lignin. Limited debonding and many fiber pull-out can be observed in specimen C3. This indicates very good interface between fiber and matrix. Under this circumstance, failure at the interface between fiber and matrix can be prevented as more failure occur inside the fiber as fiber pull-out. The structure of fiber also does not show the presence of cell walls between microfibrils of the fiber indicating significant decrease of hemicellulose and lignin. The size of the fiber is also much smaller due to the loss of lumen. With less presence of lumen, the microfibril fiber

deforms into more flattened and huddle form. This circumstance reduces the number of voids contained in the fiber resulting in denser fiber and improved mechanical properties.

Figure 4. SEM analysis on composite after delignation and bleaching treatment (specimen C3)

FTIR Spectroscopy Analysis

Composite without any treatment (specimen A) shows lower UTS compared to others. FTIR spectroscopy analysis shows that group functions chemistry of lignin still dominates the bark fiber. Group function chemicals for cellulose are OH, CH, and CO followed by the presence of several groups function chemical from lignin in the form of chemical functional groups C = O, C = C and C = C aromatic. Since specimen A is a control variable and does not receive any treatment, the intensity of the C = O functional group and the functional groups of other lignins in the salak midrib without treatment is still high and used as control value.

Fourier transform infra red (FTIR) spectroscopy analysis was carried out to characterize chemical functional groups that occur in the specimen A, B2 and C3. FTIR was performed using a FTIR Spectrophotometer (Perkin-Elmer) with the KBr pellet method. FTIR spectroscopy analysis is based on wavelength. According to Nandiyanto *et al.* [20], functional groups of OH is at $3260\text{--}3280\text{ cm}^{-1}$, while 2900 cm^{-1} is CH, 1500 cm^{-1} is C = C and 1040 cm^{-1} is CO. **Figure 5** shows the comparative efficiency of hemicellulose and lignin removal identified by each treatment. Since the structure and binding are similar to cellulose, removal of hemicellulose and lignin can reduce the absorption intensity at wavenumbers around $3260\text{--}3280\text{ cm}^{-1}$ (stretching OH), 2900 cm^{-1} (stretching CH), 1500 cm^{-1} (stretching C = C), and 1040 cm^{-1} (stretching CO). FTIR analysis of bark fiber after delignification shows higher OH stretching and CH stretches. This can be due to breaking of C = O lignin bond at 1700 cm^{-1} wavelength. Thus, hemicellulose level in the fiber increases. After bleaching treatment, the intensity of O-H stretching and C-H stretching becomes smaller which shows good removal of hemicellulose and lignin.

The highest tensile test occurs delignification with NaOH with a concentration of 5% (specimen B2). Based on the results of tests FTIR, group function chemicals in the composites are OH, CH and CO followed by the presence of groups function chemistry of lignin as chemical C = C and C = C aromatic. Spike intensity in OH functional groups and CH functional groups show presence of cellulose or hemicellulose structure.

Composite with delignation treatment followed by bleaching (specimen C3) shows the highest UTS and impact strength. Based on the results of FTIR spectroscopy analysis, chemical functional groups presence are OH, CH and CO followed by several groups function chemistry of lignin in the form of group function of chemical C = O, C = C, and C = C aromatic. The result of FTIR spectroscopy analysis show intensity decrease in the OH and CH chemical functional groups. Removal of hemicellulose and lignin is shown by decrease of OH functional groups at $3260\text{--}3280\text{ cm}^{-1}$ (stalling OH), 2900 cm^{-1} (overextend CH), 1500 cm^{-1} (stretching C = C) and 1040 cm^{-1} (stretching CO). This shows significant decreases of hemicellulose and lignin during the delignation followed by bleaching treatment. However, since group functions of C = O, C = C and C = C aromatic belong to a group function typical of lignin, it can be concluded that removal of lignin did not perfectly occur.

Figure 3.8 Result of FTIR spectroscopy analysis

4. CONCLUSIONS

Bark of salak (*Salacca zaiacca*) has not yet widely used in daily and engineering application. This work then investigates the use of bark of salak as reinforcement fiber in polymer matrix composite (PMC). Fiber treatment in this research was carried out in 3 conditions, i.e. without any treatment, delignation treatment with various NaOH concentrations and delignation treatment followed by bleaching treatment. When bark fiber experiences not treatment, the UTS and impact strength are 17.00 MPa and 10.72 kJ/m², respectively. Meanwhile, 5% is the best NaOH concentration which can result in UTS of 25.47 MPa and impact strength of 11.95 kJ/m². Duration of bleaching treatment, using 2% H₂O₂, for 90 minutes results in UTS and impact strength of 35.09 MPa and 13.77 kJ/m², respectively. SEM image and FTIR analysis shows that the highest UTS and impact strength occurs due to good adhesion between fiber and polymer matrix. Thus, composite failure occurs inside the fiber, not at the interface between fiber and polymer matrix.

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