The Effect of Silane Treatment on the Tensile Properties and Hydrophilicity of Abaca Fiber in a Fiber-Reinforced Polymer Laminate

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Abstract: Natural fiber-reinforced polymer (FRP) composites, specifically Abaca FRPs (AFRP), have gained popularity due to their sustainability and economic viability. However, their widespread application in the construction industry is hindered by the incompatibility of the fibers with various polymer matrices. This study explores the effect of silane treatment on the hydrophilicity and tensile properties of Abaca fibers when incorporated into a FRP composite. It also seeks to examine different fiber loadings for a silane-treated AFRP laminate, specifically at 10%, 15%, 20%, 25%, and 30%, to determine the optimum ratio. The study consists of three methodological phases to establish the tensile properties of Abaca fibers and epoxy resin, evaluate the effect of the silane treatment on the water absorption of untreated and treated fibers and tensile strength of the fibers and AFRP laminates, and determine the optimum fiber loading for a silane-treated AFRP laminate. Test results show that the silane treatment reduced the hydrophilic properties of Abaca fibers by 46.66%. However, the silane treatment decreased the tensile strength of the Abaca fibers by 6.78%, which also led to a decrease in the tensile strength of the AFRP laminate by 7.34%. The findings suggest that the tensile properties of the AFRP laminate are influenced not only by the interfacial adhesion between the fibers and the matrix but also by the individual tensile strength of the fibers. Lastly, the fiber loading for a silane-treated AFRP laminate that obtained the highest tensile strength of 74.50 MPa was experimentally determined to be 24.33%. From plotting a trendline of the experimental values, the actual optimum fiber loading was determined to be 22.65%. The produced laminates are applicable in construction projects that require lightweight, cost-effective, and sustainable retrofitting materials.

Key Words: abaca fiber; silane treatment; fiber-reinforced polymer laminate; fiber hydrophilicity

1. INTRODUCTION

FRPs have been rapidly emerging as a practical retrofitting material for reinforced concrete (RC) structures (Taranu et al., 2008), either made from synthetic or natural fibers. While synthetic fibers offer higher strength, their production contributes to significant greenhouse gas emissions. Abaca fibers, a strong natural fiber widely available in the Philippines, show promise for retrofitting due to their properties comparable to synthetic glass fibers (Barba et al., 2020). However, the hydrophilic nature of Abaca fibers results in weak interfacial adhesion with polymer matrices, limiting their application in the industry. To address this, chemical treatments like silane (3-trimethoxysilyl propyl methacrylate or TMPS) have been explored to enhance the mechanical properties of natural fibers for FRPs like adhesion (Siy et al., 2020) and laminate strength (Seculi et al., 2022). Siy et al. found that the silane treatment with 1 silane:10 solvent mole ratio was able to improve the compatibility of Abaca fibers to polymer matrices by enhancing their hydrophobic properties. To increase the potential of AFRPs in structural applications, the present study evaluates the impact of silane treatment on Abaca fibers and AFRP laminates, through tensile strength tests, water absorption tests, and pull-out adhesion tests. The study also identifies the optimum ratio among 10%, 15%, 20%, 25%, and 30% fiber loadings for the laminates.

2. METHODOLOGY

2.1 Research Design

This study investigated the effect of silane treatment on the tensile properties and hydrophilicity of Abaca fibers in fiber-reinforced polymer laminates and identified the optimum fiber loading (by weight) of silane-treated AFRP laminates, using various test methods conducted in three distinct phases. The first phase established the tensile strength of a single Abaca fiber and epoxy resin through the single fiber tensile test (ASTM D3822) and tensile test for composite laminates (ASTM D3039). Subsequently, untreated and silane-treated Abaca fibers and AFRP laminates underwent a comprehensive comparative analysis, including ASTM D3822, water absorption tests, pull-out tests, and ASTM D3039. Throughout this phase, a two-tailed t-test analysis with a significance level of 0.05 was used. In the final phase, the optimal fiber loading for a silane-treated AFRP laminate was identified through ASTM D3039 for varying fiber loadings. From the obtained parameters, statistical and graphical analyses were performed to evaluate the influence of silane treatment on Abaca fibers and AFRP laminates and assess the variabilities and significance of the difference in results.

2.2 Experimental Setup

The methodology used Streaky Two (S2) Abaca fibers due to their superior tensile strength (27.67 to 32.08 kg/gm-m) (Moreno et al. 2010, as cited in Hirondo et al., 2020). Defective fibers were removed, while the selected ones were cleaned, oven-dried at 80°C for 24 hours and trimmed as required per phase. The resin matrix was from a clear laminating epoxy resin which has a tensile strength of 33.78 MPa (4,900 psi) and specific gravities of 1.15 for the resin and 1.02 for the hardener (Pioneer Adhesives Inc., n.d.). The treatment of the fibers made use of a silane coupling agent called 3-(trimethoxysilyl) propyl methacrylate 98%. It has a chemical composition of $C_{10}H_{20}O_5Si$ and is non-hazardous. A summary of the number of samples for each test per phase is listed accordingly in Table 1. The percentages represent the fiber loading of the laminates by weight.

Table 1. Summary of Specimens.

No. of Samples	Specimen (Dimensions, in mm)	Condition of Fiber
	Phase 1	
5	Abaca Fiber Strand (40 mm length)	Untreated
5	Epoxy Resin Matrix (310x50x2)	N/A
	Phase 2	
5	Abaca Fiber Strand (40 mm length)	Untreated
5		Treated
5	5 grams Abaca Fiber Bundle	Untreated
5		Treated
5	Abaca Fiber Strand embedded in a Polymer Matrix (25x25x10)	Untreated
5		Treated
5	20.54% AFRP Laminate (310x50x3.55)	Untreated
5	22.22% AFRP Laminate (310x50x3.13)	Treated
	Phase 3	
5	11.50% AFRP Laminate (310x50x1.90)	Treated
5	17.39% AFRP Laminate (310x50x2.51)	Treated
5	24.33% AFRP Laminate (310x50x3.77)	Treated
5	28.13% AFRP Laminate (310x50x4.53)	Treated

2.3 Silane Treatment of Abaca Fibers

The Abaca fibers underwent surface modification through a silane treatment patterned after the method of Siy et al. (2020). The treatment process began with the preparation of the silane solution dipping bath with a 1:10 silane-solvent ratio. The solvent was created by combining 4 parts of ethanol (3,200 mL) and 1 part of distilled water (800 mL). Ten parts of this solvent were then mixed with 1 part silane coupling agent (400 mL) to produce a mixture with a 9% concentration. After 2 hours of constant stirring, the pH of the dipping bath was adjusted to 4.00 by adding acetic acid. Subsequently, the Abaca fibers were soaked in the silane solution dipping bath and four plastic bottles filled with water were placed on top as weights to ensure complete submersion. Following a 3-hour immersion, the fibers were removed from the dipping bath, placed on the surface at room temperature, rinsed with an ethanol and water solution to eliminate residual silane, and dried in an oven at 80 °C for 24 hours.

2.4 Fabrication of AFRP Laminates

AFRP laminates were fabricated using the hand lay-up method, patterned from the studies of Iqbal et al. (2020) and Alam & Al Riyami (2018). In this process, Abaca fibers were manually placed longitudinally in a 310x310x2 mm acrylic glass casting mold and saturated with epoxy resin. The samples were then cured at room temperature for 24 hours, removed from the mold, and cured for an additional 14 days. Afterward, the batch sample was cut into 5 smaller specimens of 310x50 mm and shaped into dumbbell geometry in preparation for tensile testing as shown in Figure 1.

(a) (b)

Fig. 1. AFRP Laminate Specimens: (a) Division of Individual Samples from Batch Sample, (b) Shape of FRP Laminate

Preliminary fabrication of laminates was conducted before the fabrication of the actual samples to determine the required weights of Abaca fibers and epoxy resin per ratio. Three mock-up laminates were fabricated with target fiber loadings of 10%, 30%, and 50%. Additional epoxy resin was required during the fabrication process to ensure complete fiber coverage, as the fibers absorbed the resin. The actual fiber loadings for the 10%, 30%, and 50% AFRP laminates were 9.80%, 23.17%, and 29.45%, respectively. These findings suggest that the maximum fiber loading achievable using this method was 30%. This value was used as the basis for establishing the target fiber loadings of 0%, 10%, 15%, 20%, 25%, and 30% for the tested samples. Table 2 summarizes the actual weights for the fabricated AFRP laminates.

Table 2. Actual Weights of the AFRP Laminates.

Target Fiber Loading	Abaca Fiber Weight (g)	Epoxy Resin Weight Wet (g)	Epoxy Resin Weight Dry (g)	Total Weight (g)	Actual Fiber Loading (%)
0%	0.00	228.06	195.80	195.80	0.00
10%	22.16	229.71	170.54	192.70	11.50
15%	45.55	250.25	216.35	261.90	17.39
20%	70.28	306.04	246.02	316.30	22.22
25%	96.47	317.64	300.03	396.50	24.33
30%	124.26	320.10	317.44	441.70	28.13
20% Untreated	70.28	306.04	271.82	342.10	20.54

2.5 Determination of Tensile Strength Properties of AFRP Laminate Materials

Before assessing the individual strengths of the AFRP laminate materials, the diameters of untreated and silane-treated Abaca fibers were measured using a digital caliper. Thirty random trials were conducted for each condition, resulting in an average diameter of 0.17 mm for both untreated and treated Abaca fibers. This diameter was used to calculate the cross-sectional area of the fibers. Meanwhile, the cross-sectional area of the epoxy resin was determined by measuring the average thickness and width of the sample at the 100 mm gauge length. The tensile properties of the Abaca fibers and the epoxy resin were then determined using the universal testing machine (UTM). Both the fibers and the epoxy resin were subjected to tensile loads until failure for five trials each. The tensile strength was calculated by dividing the maximum load by their respective cross-sectional areas.

2.6 Evaluation of Silane Treatment on Abaca Fibers and AFRP Laminates

In evaluating the silane treatment, four test methods were employed. ASTM D3822 was conducted on the silane-treated Abaca fibers to compare the results with those of untreated fibers. Water absorption tests by Begum et al. (2021) were then performed to assess the influence of silane treatment on the hydrophilic properties of Abaca fibers. In this test, 5 grams of untreated and silane-treated Abaca fibers were immersed in distilled water for 10-minute intervals within one hour. After each interval, the fibers were placed between paper towels to facilitate surface moisture absorption and subsequently weighed. The water absorption was quantified by computing the difference between the fiber weights after and before immersion, divided by the initial weight before immersion.

Pull-out tests based on Cai et al. (2016) were also conducted to examine the interfacial adhesion of the Abaca fibers to the polymer matrix. Each specimen had one fiber embedded in the epoxy resin with an embedment length ranging from 0.5 to 1 mm. The free end of the fiber was then subjected to tensile load until detachment from the epoxy resin occurred. Lastly, the impact of silane treatment on AFRP was investigated by subjecting the AFRP laminate samples with 20% fiber loading to tensile strength tests following ASTM D3922. The maximum load until failure was then recorded and divided by the cross-sectional area to determine their tensile strengths.

2.7 Determination of Optimum Silane-Treated Abaca Fiber to Epoxy Resin Weight Ratio

The optimum fiber-to-epoxy-resin ratio was determined through a series of tensile strength tests performed on the AFRP laminates of varying ratios of 10%, 15%, 20%, 25%, and 30%. The silane-treated AFRP laminates underwent ASTM D3922, with five trials for each weight ratio. In these trials, the maximum loads before failure were recorded, and tensile strengths were calculated. The highest tensile strength observed among the fiber loadings was identified as the optimal ratio.

3. RESULTS AND DISCUSSION

3.1 Tensile Strength Properties of AFRP Laminate Materials

Tensile strength tests on single Abaca fibers showed an average tensile strength of 1,031.41 MPa with a standard deviation of 255.76 MPa. This high variability suggests variations in fiber diameters and tensile strengths. The epoxy resin had slight variations in thickness (ranging from 1.42 to 2.47 mm) and width (ranging from 27.0 to 27.9 mm) due to fabrication. The epoxy resin sample elongated by an average of 3.96 mm, with an average tensile strength of 33.76 MPa and a low standard deviation of 8.67 MPa. This tensile strength closely matched the specifications of the manufacturer (Pioneer Adhesives Inc.) with a 0.09% difference, validating the experimental results.

3.2 Effect of Silane Treatment on Abaca Fibers and AFRP Laminates

3.2.1 Effect of Silane Treatment on the Tensile Strength of Abaca Fibers

The silane-treated single fibers upon subjection to the tensile test showed an average maximum tensile strength of 961.45 MPa with a standard deviation of 272.82 MPa. This was found to be lower than the untreated fibers by a percent decrease of 6.78% and a percentage difference of 7.02%. The t-test initially suggested no significant difference due to a p-value of 0.5443, which is higher than the significance level of 0.05. However, this may be explained by the box-and-whisker plot in Figure 2 revealing high variability and comparable ranges between the tensile strengths of the treatment conditions. The median and mean for the untreated fibers were also higher than the silane-treated fibers. Therefore, this showed that the silane treatment significantly caused a decrease in the tensile strength of the fibers.

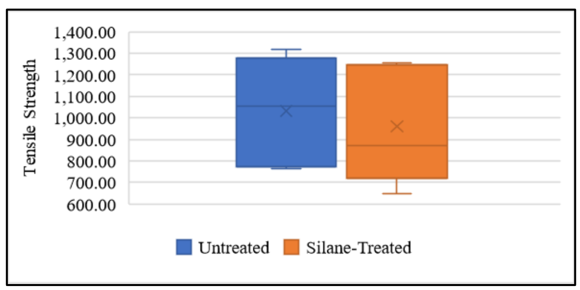

Fig. 2. Tensile Strength of Untreated and Silane-Treated Single Abaca Fibers

3.2.2 Effect of Silane Treatment on the Hydrophilicity of Abaca Fibers

The tests showed that the untreated fibers had a 219% water absorption rate with a 24% standard deviation. In comparison, the silane-treated fibers had a water absorption of 117% with a standard deviation of 14%, resulting in a decrease of 46.66% and a percentage difference of 60.85%. The t-test p-value of 1.64×10^{-5} being less than the significance level and the low variability in data (exhibited in Figure 3) proved the effectiveness of the silane treatment in reducing the water absorption, and therefore decreasing the hydrophilicity of the fibers and potentially enhancing fiber-resin compatibility.

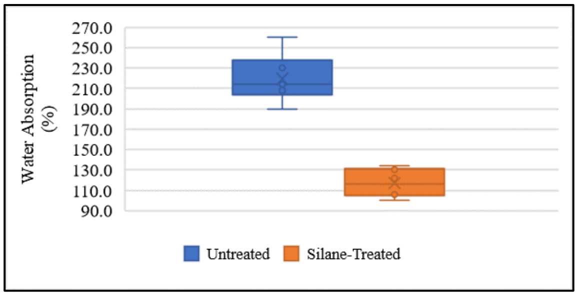

Fig. 3. Water Absorption of Untreated and Silane-Treated Abaca Fibers

3.2.3 Effect of Silane Treatment on the Interfacial Adhesion of Abaca Fibers

The pull-out test for both the untreated and silane-treated single fibers showed failure at the fiber on all the samples. The inability to extract fibers from the epoxy resin matrix may suggest a higher required pull-out force or adhesion than the single fiber tensile strength. This may indicate the presence of good interfacial adhesion between the fiber and the epoxy resin but may need further investigation to quantify the exact interfacial adhesion strength of these fibers.

3.2.4 Effect of Silane Treatment on the Tensile Strength of AFRP Laminates

The tensile strength of the AFRP laminates with 20% fiber loading averaged at 77.31 MPa (standard deviation of 10.24 MPa) for the untreated AFRP and 71.63 MPa (standard deviation of 9.28 MPa) for the silane-treated, resulting in a decrease of 7.34%. The t-test showed a p-value of 0.5658, exceeding the significance level due to the high data variability and close mean values as seen in Figure 4. However, the median, mean, and range of the tensile strengths for the silane-treated AFRP are lower. As such, the silane treatment decreased the tensile strength of the laminates despite reducing the hydrophilicity of the fibers. Additionally, there may be a factor of excessive removal of cellulose in the fibers due to potentially high silane concentration and the soaking time (Orue et al., 2016; Rongpipi et al., 2019). This cumulative tensile strength reduction of the individual silane-treated fibers may have also led to a more pronounced decrease in the tensile strength of the AFRP laminate than the single fibers. These findings suggest that the tensile properties of the AFRP laminates are influenced by both the interfacial adhesion and the tensile strength of the individual fibers.

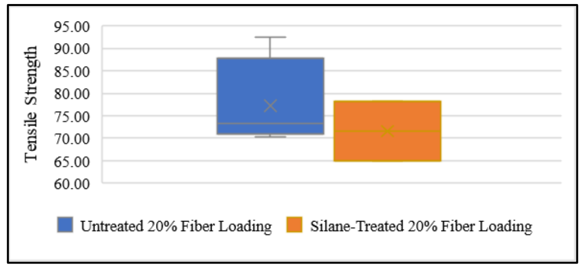

Fig. 4. Tensile Strength of Untreated and Silane-Treated AFRP Laminates with 20% Fiber Loading

3.3 Optimum Silane-Treated Abaca Fiber to Epoxy Resin Weight Ratio

Due to slippage and failure beyond gauge length during ASTM D3922, the 15% fiber loading was excluded in the comparison of the different weight ratios. The remaining test results showed that increasing the fiber loading resulted in thicker laminates with higher fiber volume and density, demonstrating greater resistance to breakage. The resulting average tensile strengths of the 10%, 20%, 25%, and 30% fiber loadings were calculated to be 54.57 MPa, 71.63 MPa, 74.50 MPa, and 57.94 MPa, respectively.

From these values, the 10% fiber-loading AFRP laminate exhibits a 61.66% increase in tensile strength compared to the epoxy resin alone. This indicates that the incorporation of Abaca fibers has shown an improvement in the tensile properties of the epoxy resin. The tensile strength was also observed to increase steadily until the 25% fiber loading. Therefore, the optimum fiber loading is 24.33% (labeled as 25% fiber loading) with a tensile strength of 74.50 MPa. The exact optimum fiber loading was found to be 22.65% using the generated trendline from the graph in Figure 5.

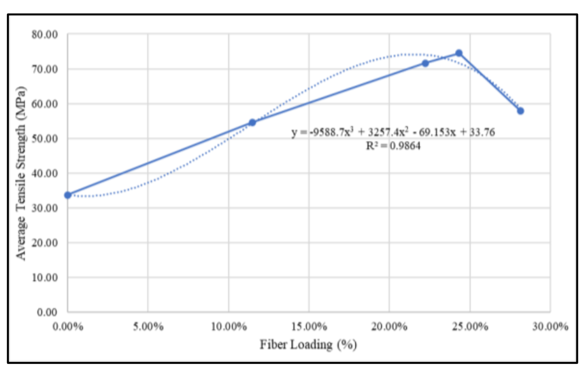

Fig. 5. Average Tensile Strength vs. Actual Fiber Loading

The results indicate a positive correlation between the fiber loading and the tensile strength, with an upward trend until a certain point, beyond which the tensile strength decreases. This decline is attributed to insufficient epoxy resin coverage, which exposes fibers and creates air voids, weakening the laminate. Additionally, inadequate fiber dispersion prevented proper epoxy resin coating, increasing fiber-fiber interactions like entanglement and clustering.

3.6 Application of Silane-Treated Abaca Fibers and AFRP Laminates in Retrofitting

Several studies have explored the potential of natural FRPs (NFRPs) for retrofitting. In one study by Saidi et al. (2021), single-layer AFRP sheets exhibited a tensile strength of 54.58 MPa, lower than those produced in this study. Its application to RC beams contributed to 11% of the total maximum shear load, increasing the shear capacity. Another study by Yinh et al. (2021) explored sisal FRPs with a tensile strength of 80 MPa, which was slightly higher than the silane-treated AFRP laminates of this study. Their sisal FRPs increased the ultimate load of RC beams by 14% to 36% and the cracking load by 45% to 73%. Given the similar tensile strengths with previous research, the AFRP laminates of the present research show the potential to retrofit RC beams effectively. Moreover, upon assessing the grammage of the silane-treated AFRP laminates which ranged from 200.52 g to 459.63 g, they were deemed lightweight than their counterparts and suitable for cost-effective, sustainable construction projects.

4. CONCLUSIONS

The study reached the following conclusions:

The silane treatment reduced the hydrophilicity of the fibers by 46.66%. However, it decreased the tensile strength of Abaca fibers by 6.78% and the AFRP laminates by 7.34%. This indicates that lower water absorption of fibers does not necessarily result in higher tensile strength for FRPs.

The decrease in tensile strength of the AFRP laminates is more pronounced than that in the single Abaca fibers. Thus, the tensile properties of the AFRP laminates not only depend on the tensile strength of the individual fibers but also their interfacial adhesion with the epoxy resin matrix.

There is a positive correlation between fiber load and tensile strength where an upward trend was evident until the 25% fiber loading then declined the 30% fiber loading.

The optimum fiber loading for silane-treated AFRP laminate is 24.33%, exhibiting the highest tensile strength averaging 74.50 MPa.

To further improve the study, future research is recommended to perform Scanning Electron Microscopy (SEM) analysis, utilize pre-fabricated Abaca fiber sheets, consider alternative fabrication methods and similar matrices, quantify interfacial shear stress, and test different silane concentrations.

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