RASĀYAN *J. Chem.*
Vol. 18 | No. 1 |151-157| January - March | 2025
ISSN: 0974-1496 | e-ISSN: 0976-0083 | CODEN: RJCABP
http://www.rasayanjournal.com
http://www.rasayanjournal.co.in

INFLUENCE OF SILANE COUPLING AGENT CONCENTRATION ON THE CHARACTERISTICS OF COCONUT FIBER-REINFORCED UNSATURATED POLYESTER RESIN BIOCOMPOSITES

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ABSTRACT

This study explores biocomposites fabricated from coconut fiber (CF) and unsaturated polyester resin, enhanced using silane coupling agents. CF was treated in silane solution at various concentrations (0–10 wt%) to improve compatibility with the resin matrix. Biocomposites were prepared by press molding and characterized by using FTIR, SEM, TGA, and UTM to assess their morphological, mechanical, thermal, and physical properties. The results show that 10 wt% silane-treated fibers (PE-CF-TVS10) exhibited the best performance, achieving superior tensile (56.1 MPa) and flexural strength (155 MPa), enhanced thermal stability, and reduced water absorption and swelling. Morphological analysis confirmed improved fiber-matrix integration due to silane treatment, reducing porosity, and increasing material density. These findings highlight the potential of CF-based biocomposites as eco-friendly, high-performance alternatives for applications such as project helmets, offering reduced dependence on synthetic materials.

Keywords: Biocomposite, Coconut Fiber, Silane Coupling Agent, Silane Treatment, Unsaturated Polyester Resin.

RASĀYAN *J. Chem.*, Vol. 18, No.1, 2025

INTRODUCTION

Increasing environmental challenges associated with synthetic materials have driven the demand for eco-friendly alternatives.[1] Biocomposites, which combine natural fibers with polymer matrices, present a sustainable solution owing to their biodegradability and potential to replace conventional materials in various applications.[2] Coconut fiber (CF), a renewable resource abundant in Indonesia, the world's largest producer of coconuts, is a promising raw material for biocomposites because of its high lignin and cellulose content, which confers excellent tensile strength, durability, and rot resistance.[3] However, the inherent hydrophilicity and limited compatibility of CF with polymer matrices, such as unsaturated polyester resin (UPR), necessitate surface modifications to improve adhesion and material performance. Among the strategies to enhance fiber-matrix compatibility, the utilization of silane coupling agents has garnered significant attention. These agents chemically modify the fiber surface and reduce hydrophilicity while enhancing interfacial bonding through covalent interactions.[4] Despite promising findings, the optimal conditions for silane treatment, including the concentration and application methods, remain under-explored for CF-reinforced UPR biocomposites. Furthermore, although biocomposites have been extensively studied, their potential for high-performance applications, such as helmet materials, has not been sufficiently demonstrated in the context of standardized physical, mechanical, and thermal properties. This study addresses these knowledge gaps by investigating the effect of silane coupling agent concentration (0–10 wt%) on the properties of CF-reinforced UPR biocomposites. This study aims to optimize the fiber treatment process to produce biocomposites with superior performance, meeting industrial standards for applications such as protective helmets. By examining the physical, mechanical, thermal, and morphological properties of the resulting biocomposites, this study contributes to advancing sustainable material development and reducing the dependence on non-biodegradable resources.

EXPERIMENTAL

Material and Methods

CF waste with a size of 60–80 mesh was obtained from home industries in the Aceh Besar Regency, Aceh Province. UPR and methyl ethyl ketone peroxide (MEKP) with technical standards were obtained from Depok, Indonesia. Vinyltrimethoxysilane (TVS) and sodium hydroxide (NaOH) were obtained from Merck, Germany. P. A was obtained from Merck (Germany). Distilled water was obtained from the Environmental Laboratory of Universitas Syiah Kuala.

Coconut Fiber Preparation

CF were prepared to obtain CF of uniform size. The CF was cleaned by combing until the solids attached to the fiber fell out. The CF was then cut into smaller pieces of approximately 2 – 3 cm, followed by washing with clean water and drying in the sun. The dried CF was thoroughly reduced in size using a dry blender. The blended CF was then sieved using a 60 - 80 mesh sieve to obtain a uniform fiber size.

Chemical Treatment of Coconut Fiber

The chemical treatment procedure was adapted from a previous study with several modifications.[5] As part of the alkaline treatment procedure, the CF was soaked in a 5% NaOH solution for 6 h and then washed thoroughly with distilled water. Subsequently, the CF that underwent alkali treatment were subjected to silane treatment. Silane treatment in different concentrations of TVS was carried out by soaking the CF in a silane solution for 1 h, to which acetic acid was added to keep the pH of the silane solution acidic. Subsequently, the CF was washed with distilled water and dried at room temperature. The CF treatment process is shown in Fig.-1.

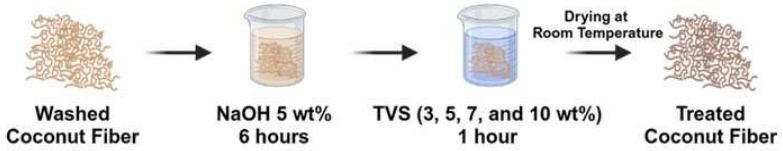

Fig.-1: CF Treatment Scheme with Alkali and Silane Solutions

Biocomposite Manufacturing

Biocomposites were prepared by the compression molding method using a stainless mold measuring 20 × 10 x 0.5 cm. The mold was first prepared and then coated with glazing wax. The CF was then spread evenly on the surface of the mold, which was glazed wax. The polyester resin was mixed with MEKP as the catalyst. The mixture was poured into a prepared mold and then forged using a press with a pressure of 250 bar and a curing time of 24 h (Fig.-2). The biocomposite design variables used in this study are listed in Table-1.

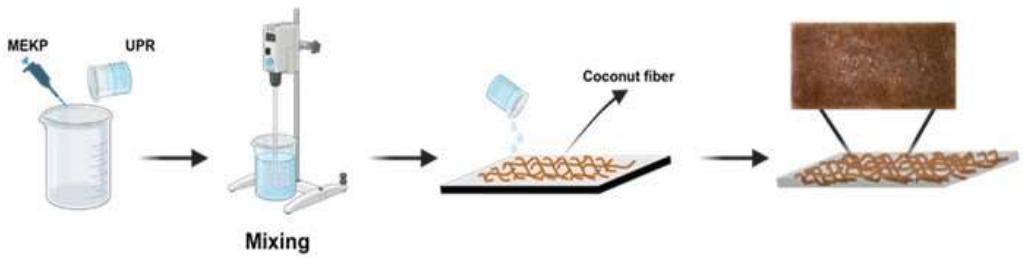

Fig.-2: Manufacturing of CF Biocomposite

Characteristics and Properties of Biocomposite Coconut Fiber

The characteristics of the CF biocomposite were determined using Fourier Transform Infrared (FTIR) spectroscopy and Scanning Electron Microscopy (SEM) analyses to determine the functional groups before and after modification using TVS and morphology. The biocomposite property test was carried out to determine its performance and compare it with SNI standards as a material for making helmets. Several biocomposite performance tests were conducted to determine the physical, mechanical, and thermal properties.

Table-1: Variable design of biocomposites

Sample	UPR (wt%)	CF (wt%)	TVS Concentration (wt%)
PE	100	0	0
PE-CF	95	5	0
PE-CF-TVS3	95	5	3
PE-CF-TVS5	95	5	5
PE-CF-TVS7	95	5	7
PE-CF-TVS10	95	5	10

The physical properties evaluated were the density, water absorption, and swelling thickness.[6,7] The mechanical properties evaluated were tensile strength and flexural strength. Thermal properties were evaluated using Thermogravimetric Analysis (TGA).

RESULTS AND DISCUSSION

Functional Group of Coconut Fiber

Functional group analysis of the CF before and after chemical treatment was performed to determine the success of the treatment. Alkali (NaOH) and silane (TVS) treatment was confirmed to be successful based on the appearance of new peak intensities in the FTIR spectrum. Figure-3 shows the FTIR spectra of pure CF and CF after alkali and silane treatments. The hydroxyl group (O-H) at a wave number of approximately 3600 cm^{-1} increased in intensity after chemical treatment because the lignin that coats the internal structure of CF was reduced by alkali treatment. As a result, the O-H groups in the CF were more exposed in intensity. However, the increase in the intensity of the O-H group can also be caused by the alkali and silane treatment itself. The NaOH and TVS solutions (which have been hydrolyzed) also contain O-H groups, which may also affect the hydrogen bonds between the O-H groups and the structure of the CF.[8] The C-H group at a wave number of 910 cm^{-1} indicated the presence of cellulose in the CF structure. The intensity of the C-H group is more visible in the spectrum of the CF after alkali treatment compared to pure CF and after silane treatment. This indicates that alkali treatment can remove lignin protection so that the presence of cellulose is more easily detected in the spectrum. The absorption of Si-O-Si groups from silane treatment at wave numbers 1000-600 cm^{-1} widened, causing the C-H groups of cellulose to be closed. Meanwhile, the decrease in lignin concentration can be confirmed by the C=O and C-O-C groups, whose intensity decreased after alkali and silane treatment. Notably, the functionalization of the CF as a coupling agent was successfully confirmed through the appearance of the Si-O-Si group.[9]

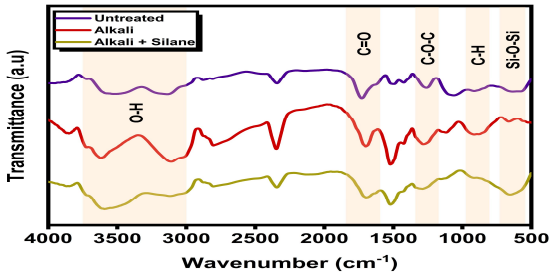

Fig.-3: Functional Group of CF Before and After Silane Treatment

Morphology of Biocomposites

Biocomposite surface analysis was carried out to show the effect of the silane coupling agent on the interaction of the CF with the UPR matrix. Fig.-4(a) shows the surface of the PE biocomposite without the addition of the CF filler. The surface of the PE biocomposite appeared smooth because there was no interaction with the filler, which caused an increase in surface roughness. However, bubbles were still present in the PE biocomposite structure because of the mixing process of UPR with MEKP and printing, which trapped the bubbles. This is related to the results of the PE biocomposite density being lower than that after adding filler and the use of CF filler, which was treated with a s coupling agent. Fig.-4(b) and 4(c) show that the surface structures of the PE-CF and PE-CF-TVS10 biocomposites appear rougher after the addition of the CF filler to the UPR matrix. From this morphological analysis, it can be seen that the UPR cannot be integrated with the CF and resembles two materials that are separated from each other. This is

possibly caused by the absence of reaction interactions between the matrix and the filler. CF is an organic material while UPR is an inorganic material. This makes sense when different types of materials cannot form strong bonds in the material matrix structure. However, after the CF was treated with a silane coupling agent, the CF seemed to be integrated into the matrix. It can also be seen in Fig.-4(c) that there is no gap between the matrix and CF, as shown in Fig.-4(b). This analysis confirms that the silane coupling agent functionalized in the CF can improve the interaction between organic and inorganic materials.

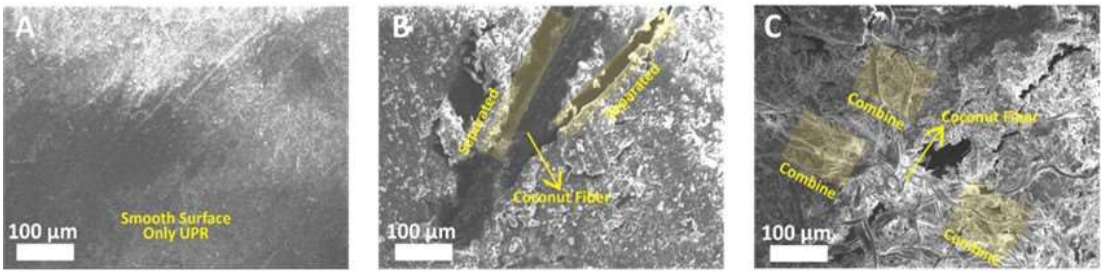

Fig.-4: Morphology of Biocomposite: (a) PE, (b) PE-CF, and (c) PE-CF-TVS10

Physical Properties of Biocomposite

The addition of CF and the application of a silane coupling agent affect the density of the biocomposite by reducing the empty space in the matrix (voids). Fig.-5(a) shows the effect of adding CF and different concentrations of silane coupling agent on the density of the biocomposite. The increase in the density value of the biocomposite is caused by the addition of a new material, namely CF so that the biocomposite matrix of the UPR is filled with CF and causes its mass to increase. The increasing mass of the material causes the ratio with the volume of the material to increase so that the value of the material density also increases. However, the addition of a silane coupling agent also increases the material density. This is because the amount of CF added was the same for each type of biocomposite PE-CF, PE-CF-TVS3, PE-CF-TVS5, PE-CF-TVS7, and PE-CF-TVS10, but the silane coupling agent created cavities. empty cavities in the biocomposite matrix were reduced. As a result of the reduced voids, the weight of the material also increased and the density value increased.

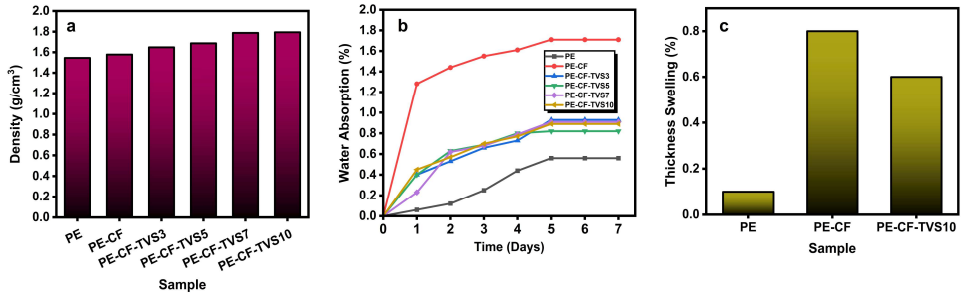

Fig.-5: Silane Coupling Agent Effect on Biocomposite Density (a), Water Absorption (b),Thickness Swelling (c)

In contrast to the increasing density value, Fig.-5(b) shows a decrease in the water absorption value of the biocomposite owing to the application of the silane coupling agent. The addition of CF without silane treatment produced a biocomposite with the highest water absorption value. This is because of the natural properties of CF, which is an organic material that easily absorbs water because it is rich in hydroxyl groups. Meanwhile, after silane treatment, the water absorption value of the CF biocomposite decreased because of several factors, such as a decrease in voids, which could cause water to penetrate the biocomposite structure, and a decrease in the number of hydroxyl groups due to the formation of hydrogen bonds with the hydroxyl groups of TVS. Zhu *et al.*[10] used KH550 and KH590 as coupling agents for epoxy resins and hollow glass microspheres (HGM). The decrease in water absorption after modification also showed significant results compared with the case without a coupling agent. Fig.-5(c) shows a comparison of the thickness swelling between PE and PE-CF and PE-CF-TVS10 biocomposites owing to the influence of the application of the silane coupling agent to CF. Changes in the swelling thickness of a material provide information that there is a change in the internal structure of the matrix. The presence of the silane coupling agent in this case

reduced the swelling thickness of the PE-CF-TVS10 biocomposite because the adhesion and condensation reactions between the PE and CF matrices became stronger. In their research, Saidi *et al.*[11] explained the role of coupling agents in rice husks, where it is difficult for water molecules to penetrate between the rice husk and the PVC matrix owing to the decrease in interfacial gaps and voids, thereby also reducing the opportunity for an increase in matrix volume, which is related to thickness swelling.

Mechanical Properties of Biocomposite

The tensile strength can be defined as the maximum stress that a material can withstand before breaking when stretched or pulled.[12] This tensile strength measurement can be used to evaluate the mechanical strength of a material so that the field of application can be adjusted. There was an increase in the tensile strength of the CF biocomposite after treatment with a silane coupling agent at several different concentrations. Biocomposites PE-CF, PE-CF-TVS3, PE-CF-TVS5, PE-CF-TVS7, and PE-CF-TVS10 obtained tensile strength values of 34.7, 47.9, 53.6, 55.7, and 56.1 MPa respectively (Fig.-6(a)). The insignificant increase starting from TVS concentrations of 5, 7, and 10 wt% may be caused by the achievement of the optimum conditions in the silane coupling agent treatment. The silane coupling agent interacts with the available CF active groups to form hydrogen bonds and condenses to form stronger chemical bonds. The greater the TVS concentration, the more the CF reactive side was affected. Thus, the maximum possible conditions were almost reached. Pisanu *et al.*[13] reported the role of coupling agents in increasing the tensile strength of polymer materials. They used CF as a filler in composites with a polypropylene (PP) matrix and maleic anhydride (MA) as a coupling agent. As a coupling agent, MA increased the interfacial adhesion between CF and PP. According to Liu *et al.*[14], silane coupling agents can also increase tensile strength because the amount of energy that can be absorbed by the material increases with a strong bond between the filler and the matrix. The stronger the interfacial bonding, the greater the force that the biocomposite can accept before breaking.

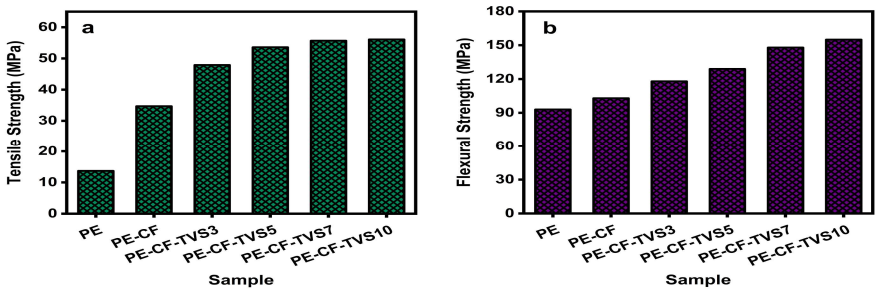

Fig.-6: Tensile (a) and Flexural (b) Strength of Biocomposite-Treated Silane

Another mechanical property of biocomposite materials, which is also important for evaluating changes due to modification, is flexural strength. Fig.-6(b) shows the increase in the flexural strength of the biocomposite without the coupling agent and after the addition of the coupling agent. In addition, increasing the concentration of the coupling agent, TVS, on CF can also increase the flexural strength of the biocomposite. The highest flexural strength was obtained by the PE-CF-TVS10 biocomposite with a value of 155 MPa, whereas the lowest was obtained by PE-CF without a coupling agent at 103 MPa. The PE matrix itself had a flexural strength of 93 MPa, which was 40% lower than that of PE-CF-TVS10. Similar results with the same trend were also obtained by Tissandier *et al.*[15], where there was an increase in flexural strength between wood fiber and polyethylene as a matrix. Under dry conditions, applying pressure to the body of the biocomposite material with a coupling agent makes the force that the biocomposite can withstand higher, owing to better stress transfer and interface bonding.[16]

Thermal Properties of Biocomposite

The thermal properties of the biocomposites were evaluated using thermogravimetric analysis (TGA). This evaluation was also intended to determine how the silane coupling agent using TVS improves the thermal stability of the CF biocomposite. Fig.-7 shows the relationship between the biocomposite weight loss and the increasing temperature of the biocomposite. Natural fibers typically undergo thermal degradation in three stages, with most of the decomposition occurring in the temperature range of 240–500°C. In the first

stage, weight loss is caused by moisture loss. The second stage involved the thermal degradation of cellulose, while the third stage included an initial peak indicating hemicellulose decomposition and a later peak corresponding to lignin decomposition. Hemicellulose decomposes maximally at 300°C, whereas lignin shows peak thermal decomposition between 280 and 520°C. The reduction in molecular weight is influenced by the composition of the filler and matrix. T onset is the temperature at which the biocomposite begins to decompose.[17] It can also be seen that the final weight loss of the biocomposite with the silane coupling agent treatment increased slightly. This is because the application of the TVS coupling agent adds an ethinyl group (−CH=CH_2) to the CF biocomposite structure, which directly increases the existing bonding energy. The increasing bonding energy also increases the thermal stability of the CF biocomposite, as indicated by the heat required to degrade the material, which also increases.[18]

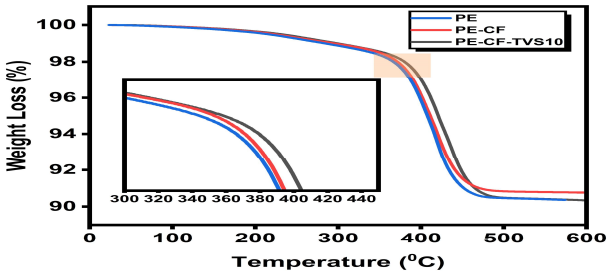

Fig.-7: Weight Loss of Biocomposite-Treated Silane

CONCLUSION

This study demonstrated that incorporating a silane coupling agent enhances the properties of CF-reinforced unsaturated polyester resin biocomposites. The PE-CF-TVS10 sample, with 10% silane-treated fibers, showed the best performance. Chemical modification improved the fiber and matrix interaction, confirmed by functional group analysis. Morphological analysis revealed a more integrated and less porous structure in silane-treated biocomposites, leading to superior mechanical properties. The PE-CF-TVS10 sample exhibited increased tensile strength (56.1 MPa) and flexural strength (155 MPa), outperforming untreated samples. Thermal stability also improved, as indicated by higher decomposition temperatures and reduced weight loss in thermogravimetric analysis. Physical properties, including density and water absorption, benefited from silane treatment. Increased density, reduced water absorption, and swelling thickness indicated a more compact, hydrophobic material structure. These findings advance sustainable materials by utilizing natural fibers and enhancing their compatibility with polymer matrices through chemical treatments. Further research is recommended to explore these biocomposites' long-term durability and performance in practical applications.

ACKNOWLEDGMENTS

The author would like to thank the Department of Chemical Engineering, Faculty of Engineering, Universitas Syiah Kuala for its support for this research. The author also would like to thank the Institute for Research and Community Service (LPPM) at Universitas Syiah Kuala for providing research funding under the program Penelitian Calon Professor (170/UN11.2.1/PG.01.03/SPK/PTNBH/2024).

CONFLICT OF INTERESTS

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

All the authors contributed significantly to this manuscript, participated in reviewing/editing, and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

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