RASĀYAN *J. Chem.*
Vol. 19 | No. 3 | 1299-1306 | July-September | 2026
ISSN: 0974-1496 | e-ISSN: 0976-0083 | CODEN: RJCABP
http://www.rasayanjournal.com
http://www.rasayanjournal.co.in

ENHANCING THE MECHANICAL AND THERMAL PROPERTIES OF ABACA FIBER/GRAPHITE BIOCOMPOSITES THROUGH ALKALI AND SILANE SURFACE MODIFICATION

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ABSTRACT
This study develops and characterizes abaca fiber (AF)/graphite/unsaturated polyester resin (UPR) biocomposites, investigating the effect of vinyltrimethoxysilane (TVS) coupling agent (0–8 wt%) on their physical, mechanical, and thermal properties. After alkaline and silane functionalization, the abaca fibers were prepared for molding. The composites were then held at 50 bar for 24 hours via compression molding. FTIR analysis confirmed successful silane bonding through enhanced Si–O–C absorption at 825 cm^{-1}, while SEM observations revealed a more homogeneous fiber–matrix interface with reduced voids. The application of silane treatment yielded a substantial improvement in composite characteristics. The modified specimens exhibited an increased density of 1.448 g/cm^3, while concurrently demonstrating a reduction in water absorption and thickness swelling to 4.12% and 3.00%, respectively. Characterization results revealed optimal mechanical behavior. The specimens achieved a tensile peak of 35.194 MPa. Flexural performance was also high, measuring 80.264 MPa. TGA results further indicated improved thermal stability in silane-treated composites. These findings demonstrate that surface-engineered natural fibers can overcome interfacial incompatibility-one of the primary limitations in natural fiber composites—while achieving performance comparable to synthetic counterparts. Leveraging graphite as a functional filler allows for the development of resilient materials tailored for the automotive and packaging industries, specifically addressing the need for low water vapor permeability and high load-bearing capacity. From an academic perspective, this study provides a reproducible framework for silane-based interfacial engineering applicable to other agro-industrial fibers, while supporting circular economy strategies through the valorization of agricultural waste and advancing the development of low-carbon structural materials.
Keywords: Abaca Fiber; Silane Treatment; Biocomposite; Interfacial Adhesion; Unsaturated Polyester Resin.
RASĀYAN *J. Chem.*, Vol. 19, No.3, 2026

INTRODUCTION

There is an increasing industrial shift toward high-performance materials that maintain rigorous durability standards while fulfilling stringent sustainability criteria. At the same time, these materials must remain economically competitive.[1] Metallic substrates face significant utility constraints, limited by poor oxidative stability, which high-intensity production cycles further exacerbate. Engineering trends now favor natural fiber biocomposites, which deliver optimized mass efficiency alongside essential biodegradability. Their abundant sourcing and robust mechanical performance make them viable industrial substitutes.[2] Among natural fibers, the abaca-derived pseudostem offers exceptional tensile strength and durability for advanced structural reinforcement.[3] Its utilization in currency paper and marine rigging demonstrates superior mechanical durability and versatility. Significant biomass residuals stem from Indonesian banana cultivation, providing a scalable resource for green engineering. Utilizing these materials supports circular economy objectives within the region. Currently, pseudostems remain underutilized conventional disposal methods include open burning or landfilling. Converting this underutilized feedstock into high-value materials offers a major engineering opportunity.[4] Abaca fiber is a robust sustainable reinforcer that integrates well into polymer matrices to improve mechanical integrity. Its origin from the Musa textilis Nee pseudostem ensures low environmental impact, and engineers increasingly adopt this fiber for eco-friendly material design. Abaca fibers contain 63–64% cellulose and

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approximately 5% lignin. This specific chemical ratio results in a high tensile strength of 600 MPa. Consequently, these fibers serve as an effective reinforcement phase in polymer composites.[5] Applications in currency paper and marine ropes confirm its mechanical versatility across diverse industries. Indonesia, as a top global banana producer, generates a huge volume of pseudostem waste currently discarded or incinerated representing a largely ignored opportunity to transform this biomass into high-value materials.[6] Hydrophilicity in abaca fibers limits their effectiveness in UPR-based composites by preventing strong interfacial adhesion with the hydrophobic polymer phase, negatively affecting the mechanical stability of the final material. To optimize the interface, silane agents are utilized to create durable covalent anchors that bridge the fibers and matrix, improving load transfer while lowering the fiber's hydrophilic nature and protecting against moisture-induced degradation.[7] Graphite serves as a critical secondary filler for matrix refinement. Its inclusion increases the composite's overall stiffness. Simultaneously, it provides superior thermal and electrical transport properties. However, despite individual studies on silane treatment and graphite addition, the synergistic effect of combining both on abaca-reinforced UPR composites remains poorly understood particularly regarding optimal silane concentration and its interaction with graphite fillers under controlled processing conditions, a gap that limits development for high-performance structural applications. This paper analyzes how abaca fiber reinforcement affects the mechanical and structural properties of UPR, with performance systematically assessed using press molding. Graphite filler was fixed at 5 wt%, while silane coupling agent levels ranged from 0 to 8 wt%. Morphological and thermal evaluations determine the most effective treatment for enhanced composite performance. Beyond technical goals, this research promotes SDG 9 via biocomposite development and SDG 12 through a waste-to-wealth model for agricultural residues, minimizing reliance on synthetic polymers and improving industrial sustainability metrics.[8]

EXPERIMENTAL

Material and Methods

Abaca fiber waste with a size of 60–80 mesh was obtained from the banana industry in Aceh Besar Regency, Aceh Province. Graphite came from Semarang, Indonesia. Its particle size was 5 microns. We obtained the UPR and MEKP from Depok, Indonesia. Merck (Germany) supplied the sodium hydroxide (NaOH) and vinyltrimethoxysilane (VTMS) used in this study. Five VTMS concentrations were prepared, ranging from 0 to 8 wt% at 2% increments. Analytical grade acid (p.a.) was obtained from Merck (Germany). Distilled water was obtained from the Environmental Laboratory of Universitas Syiah Kuala.

Biocomposite Manufacturing

A compression molding method was used to create the biocomposites in this study. The specimens were formed using a stainless-steel mould. It measured 20 cm by 20 cm by 0.5 cm. Before use, the mold was coated with glazing wax to prevent the composite from sticking when removed. Then, polyester resin and graphite were mixed evenly. The reaction was initiated via the addition of the catalyst, methyl ethyl ketone peroxide (MEKP). The mixture was poured over the abaca fibers already positioned in the mold, and then compressed with a press machine. Curing was performed at 50 bar for a duration of 24 hours.[9] The experimental setup is depicted in Fig.-1. The composites design variables used in this study are listed in Table-1.

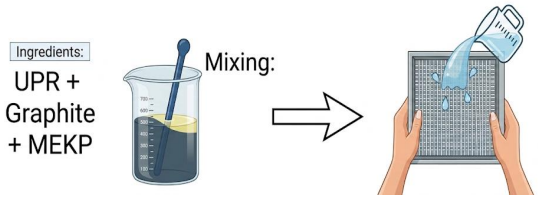

Fig.-1: Manufacturing of FA Composites

Abaca Fiber Preparation

AF were prepared to obtain AF of uniform size. The AF was cleaned by combing until the solids attached to the fiber fell out. The AF was then cut into smaller pieces of approximately 2 – 3 cm, followedby washing with clean water and drying in the sun. The dried AF was thoroughly reduced in size using a dry

blender. To achieve a uniform fiber size, the blended abaca fiber (AF) was subsequently passed through a 60–80 mesh sieve.

Table-1: Variable Desain Serat Abaca (Treated vs Untreated)

Sample	UPR (% wt)	AF Composition (% wt)	Fiber Treatment Method	TVS Concentration (% wt)	Graphite Composition (% wt)
B0	100	0	-	0	0
B1	90	5	AF0	0	5
B2	90	5	AF0	2	5
B3	90	5	AF0	4	5
B4	90	5	AF0	6	5
B5	90	5	AF0	8	5
A1	90	5	AF1	0	5
A2	90	5	AF1	2	5
A3	90	5	AF1	4	5
A4	90	5	AF1	6	5
A5	90	5	AF1	8	5

Note: AF (Abaca Fiber), AF0 (Abaca Fiber Treated Silane), and AF1 (Abaca Fiber Untreated Silane)

Chemical Treatment of Abaca Fiber

The initial step for the abaca fiber is a 4-hour soak in a 5% NaOH solution. Following this, the fiber is washed and then dried for 12 hours in a 60℃ oven. Afterwards, it undergoes a two-hour silane treatment in a solution with an acidic pH, maintained by adding acetic acid. The fiber is then washed using distilled water before being dried at ambient temperature. Direct and Simple: This study created biocomposites using abaca fiber fillers that were either untreated or treated with silane. The key difference is when the silane treatment occurs: untreated fibers are treated with silane after being made into a biocomposite.[10] The AF treatment process is shown in Fig.-2.

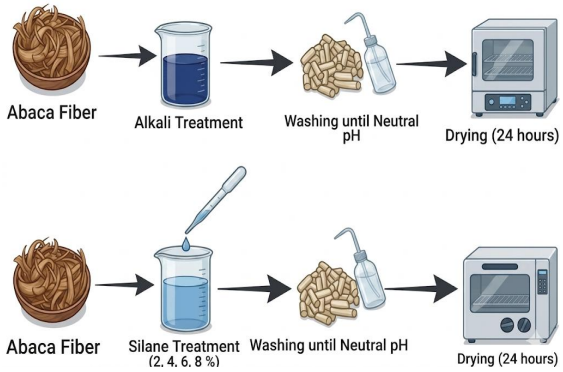

Fig.-2: AF Treatment Scheme with Alkali and Silane Solutions

Analysis Data

The analysis in this study was conducted using FTIR, SEM, TGA, and UTM instruments. The obtained data were processed descriptively by presenting them in the form of tables and graphs to identify the samples that are potentially for use.

RESULTS AND DISCUSSION

Functional Group of Abaca Fiber

FTIR was employed for the functional group identification of abaca fibers. Untreated samples were assessed alongside alkali and alkali-silane hybrid treatments. This comparison identifies the chemical shifts in the abaca fibers. This approach identifies chemical alterations in the fiber structure. Untreated abaca shows a significant O–H absorption peak at 3430–3450 cm^{-1}, characteristic of cellulose and hemicellulose in unmodified lignocellulosic fibers. Additional peaks confirm C=O groups (1750 cm^{-1}), C≡N nitrile groups (2260–2200 cm^{-1}) in both untreated and alkali-treated fibers, and C–N amine groups (1190 cm^{-1}) in raw abaca. Silane-treated fibers exhibit a more pronounced C=O peak and notably higher

Si–O–C peak intensity at 825cm^{-1}, confirming covalent bonding and validating the silane coupling mechanism at the fiber-matrix interface. Post-treatment, abaca mirrors the functional modification observed in sisal and jute, distinguished primarily by its stronger Si–O–C peak intensity- reflecting its high surface reactivity. Silane treatment effectively optimizes the abaca-matrix interface. This modification improves chemical compatibility between components. Consequently, the biocomposite exhibits superior mechanical integrity for high-performance use.[11]

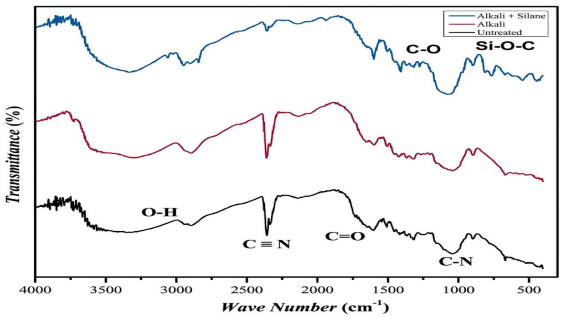

Fig.-3: Functional Group of AF Before and After Silane Treatment

Morphology of Composites

The SEM technique generates high-fidelity surface imagery via electron beam interaction. This method is essential for analyzing complex surface architectures. It also provides critical data regarding the distribution of material compositions. Interfacial interactions govern composite strength. Therefore, precise bonding observations are necessary for engineering analysis. Fig.-4(a) observations on the resin show a smooth and uniform surface without any fiber structure, but is susceptible to micro cracks when stressed-consistent with reported limitations of unreinforced polymer matrices. SEM imaging indicates superior interfacial adhesion in the treated samples. Fibers show seamless integration with the surrounding resin. Small voids still occur, however, creating detrimental stress points. These sites reduce the composite's load-carrying capacity. Silane treatment enhanced fiber homogeneity, as illustrated in Fig.-4(c). The interface is stable and void-free. This indicates robust chemical bonding across all phases. This aligns with established findings regarding surface modification via silane. Improved interfacial adhesion remains a key driver for performance in bio-composites. Micrographs of the untreated composite reveal interfacial cavities. These defects result from the hydrophilic behavior of abaca fibers in a hydrophobic matrix. The results justify the requirement for fiber surface modification to ensure structural integrity.[12]

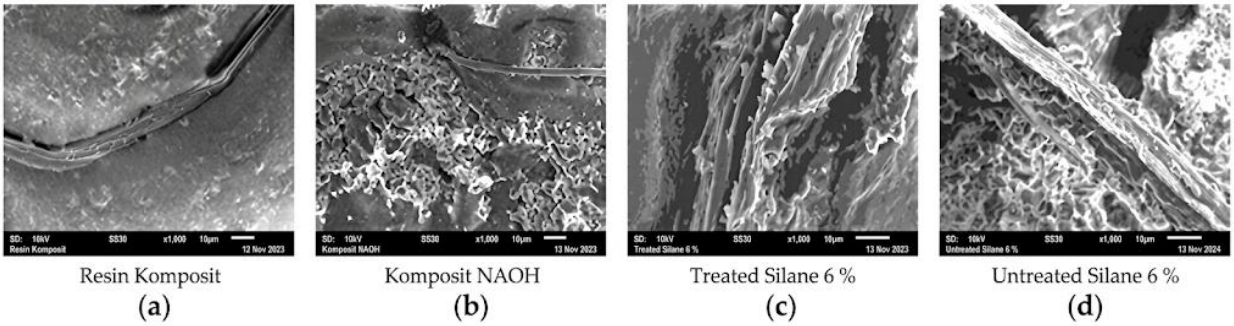

Fig.-4: Morphology of Composites: (a) Resin Magnification 100× to 1000×, (b) NaOH Composite Magnification 100× to 1000×, (c) Treated Composite Magnification 100× to 1500×, (d) Untreated Composite Magnification 100× to 1500×

Physical Properties of Composites

The addition of AF and the application of a silane coupling agent affect the density of the biocomposite by reducing the empty space in the matrix (voids). In contrast to the increasing density value, Fig.-5(a) illustrates the effect of silane coupling agent on the density of biocomposites that have been treated, untreated, resin only, and composites treated with NaOH. The B0-AF0 sample used as a control has the lowest density, which is 0.924 g/cm³ because there are no fibers or fillers that increase its density. Density

values rose incrementally across the samples. A1-AF0 measured 1.208 g/cm^3, while B5-AF0 and A5-AF1 reached 1.328 and 1.448 g/cm^3, respectively. All measurements align with the expected theoretical range. Silane treatment increased these values by enhancing fiber–matrix adhesion.[13] This modification improves stress distribution and effectively minimises porosity within the composite structure. Adding graphite as a filler further increases density by filling the empty spaces within the matrix.[10] These findings align with prior studies on natural fiber-reinforced composites and contribute to the body of knowledge by demonstrating that the combined use of abaca fiber, graphite filler, and silane treatment yields density improvements comparable to synthetic fiber composites, reinforcing the viability of agro-based biocomposites as sustainable structural materials. Figure-5(b) shows water absorption values in samples B0-AF0, A1-AF1, A5-AF1, and B5-AF0 at constant time, namely 2.73; 6.81; 4.12; and 6.77%, respectively. Silane treatment enhances the matrix-filler interfacial bond. This modification increases fiber hydrophobicity. Consequently, the composite exhibits significantly lower water absorption. Adding graphite closes the pores, preventing water penetration and improving dimensional stability. These results extend existing understanding by confirming that the synergistic effect of silane treatment and graphite addition can reduce water absorption to levels approaching those of synthetic composites, supporting the use of these biocomposites in moisture-prone environments.[14]

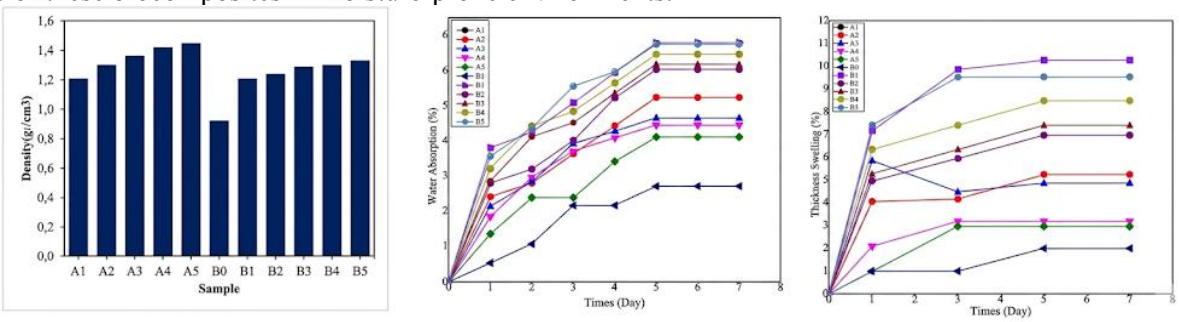

Fig.-5: Silane Coupling Agent Effect on Composites Density (a), Water Absorption (b), Thickness Swelling (c)

Figure-5(c) shows thickness swelling values of B0-AF0, A1-AF1, A5-AF1, and B5-AF0 at 2.02; 10.31; 3.00; and 9.57%, respectively. Silane treatment reduced thickness swelling by forming strong covalent bonds between the silanol and hydroxyl groups, increasing fiber–matrix compatibility and reducing –OH groups available to interact with water.[12] This contributes empirical evidence to the broader literature, affirming that silane coupling agents are a practical solution for controlling dimensional instability-a key barrier to the wider adoption of natural fiber biocomposites in engineering applications.[15]

Mechanical Properties of Composite

Tensile strength is the maximum pulling stress a material can withstand before failure. Abaca-PE biocomposites outperform traditional natural fiber alternatives in tensile testing. Data indicates significantly higher stress resistance and modulus values. Reinforcement efficiency is governed by the inherent strength of the Musa textilis fiber. However, final composite stiffness is contingent upon fiber orientation and chemical surface treatments. As shown in Fig.-6a, the tensile strength values of samples B0-AF0, A1-AF1, A5-AF1, and B5-AF0 were 12.434, 17.571, 35.194, and 29.958 MPa, respectively. Reinforcing the resin with abaca and graphite fibers significantly improved tensile performance. The observed enhancement is attributed to strong interfacial bonding.

This mechanism effectively linked the fibers and the matrix. Among all treatments, silane-treated samples demonstrated the greatest improvement, achieving the highest tensile strength and elastic modulus at optimal concentration, indicating enhanced strength and stiffness.[13] Flexural strength is another crucial mechanical property for evaluating biocomposite modifications. As shown in Fig.-6(b), flexural strength values for samples B0-AF0, A1-AF1, A5-AF1, and B5-AF0 were 17.112, 38.665, 70.543, and 80.264 MPa, respectively. Chemical bonding from silane treatment significantly improves the composite's flexural response. It ensures uniform stress dissipation throughout the matrix. Graphite further reinforces the structure, balancing high strength with essential flexibility.[16]

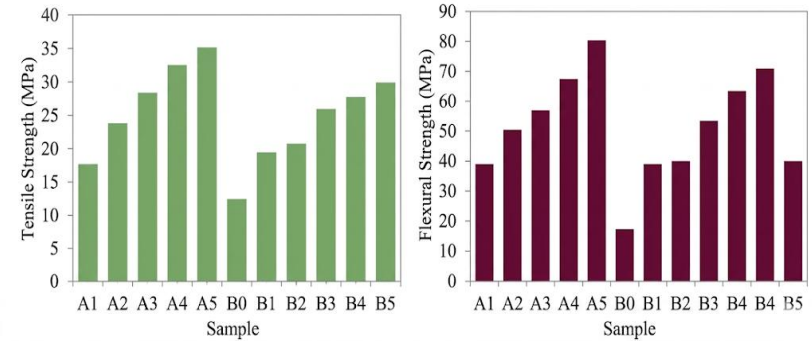

Fig.-6: Tensile (a) and Flexural (b) Strength of Biocomposite-Treated Silane

Thermal Properties of Composites

Thermal degradation in natural fiber biocomposites proceeds through three distinct stages visible in Figure 7. The first involves evaporation of bound water below 100°C. The second most significant occurs between 240 and 500°C, where hemicellulose fully degrades below 300°C and cellulose undergoes thermal decomposition. The third reflects the gradual decomposition of lignin between 280 and 520°C, consistent with lignin's well-documented thermal resistance in the literature. As shown in Fig.-7, the TGA curves reveal that the A5-AF1 sample exhibited the highest thermal stability among the four samples tested (B0-SA0, A1-SA1, A5-SA1, and B5-AF0), evidenced by its minimal weight loss at elevated temperatures, consistent with previous studies confirming that chemically modified natural fibers significantly enhance the thermal resistance of biocomposites. The Tonset parameter further confirms this superiority, suggesting that the optimum fiber-to-matrix ratio critically determines degradation onset, an aspect still underexplored in literature on locally sourced natural fiber biocomposites. The application of a silane coupling agent significantly enhanced thermal stability. This improvement is evidenced by higher final weight loss and midpoint temperatures. Additionally, both the decomposition setpoint and endpoint increased. These results extend beyond previous studies by demonstrating that silane treatment enhances not only mechanical interfacial compatibility but also thermal performance, offering empirical contributions to the body of knowledge on surface modification strategies for high-performance biocomposite applications.[17,18]

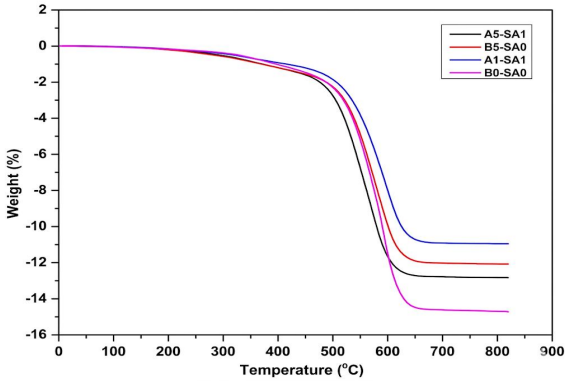

Fig.-7: Weight Loss of Biocomposite-Treated Silane

Guided by SDGs 9, 12, and 13, this study harmonises industrial output with environmental preservation by developing abaca (Musa textilis) fiber-based biocomposites through alkali-silane treatment and graphite reinforcement — producing high-performance materials from renewable agro-industrial resources that reduce dependence on fossil-based synthetic fibers. The A5-AF1 sample demonstrated competitive mechanical and thermal performance, with tensile strength of 35.194 MPa, flexural strength of 70.543 MPa, along with enhanced thermal stability and dimensional resistance, confirming its viability for structural applications while supporting local economic empowerment and reducing the carbon footprint of engineering material production-consistent with global efforts toward sustainable and climate-resilient industry.[19]

CONCLUSION

This study developed high-performance abaca fibre biocomposites. The process utilised alkali-silane treatment and graphite reinforcement. The fabrication involved dual chemical treatments: alkali-silane and graphite reinforcement. This method enhances material performance significantly. Furthermore, the development supports global sustainability goals for industry and climate. FTIR data verified the covalent bonding between the fiber and matrix through a peak at 825 cm^{-1}. This chemical modification led to a refined microstructure. SEM observations revealed a unified interface in treated samples, whereas untreated samples showed clear cavities. These defects are attributed to the inherent incompatibility of the hydrophilic fiber and hydrophobic matrix. Across all evaluated properties, A5-AF1 showed the highest performance. It recorded a maximum density of 1.448 g/cm^3. Water absorption and thickness swelling were minimised at 4.12% and 3.00%, respectively. These findings highlight a significant reduction in porosity and enhanced stability. Strong interfacial adhesion, driven by silane treatment, yielded the highest mechanical values. Tensile strength reached 35.194 MPa. Simultaneously, flexural strength peaked at 70.543 MPa. TGA results likewise confirmed its greatest thermal stability with minimal weight loss at elevated temperatures. Overall, abaca biocomposites with alkali-silane and graphite reinforcement represent a viable sustainable structural alternative to synthetic fiber composites, supporting agro-industrial resource utilization and carbon footprint reduction.

ACKNOWLEDGMENTS

I'd like to thank the Department of Chemical Engineering, Faculty of Engineering, Universitas Syiah Kuala for their support of this research. Special thanks also go to the Institute for Research and Community Service (LPPM) at Universitas Syiah Kuala for providing research funding through the Professor Candidate's Research program (grant number: 170/UN11.2.1/PG.01.03/SPK/PTNBH/2024).

CONFLICT OF INTERESTS

The authors certify that they have no competing interests, either personal or professional, that could be perceived to bias the outcomes or interpretation of this study.

AUTHOR CONTRIBUTIONS

The present work is related to *SDG-12: Responsible Consumption and Production*. 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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Cite this article as: F. Mulana *et al.*, *Rasayan J. Chem.*, 19(3), 1299-1306(2026), https://doi.org/10.31788/RJC.2026.1939595

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