Eksergi
Chemical Engineering Journal
Vol 22, No. 2. 2025

ISSN: 1410-394X
e-ISSN: 2460-8203

Combination Process of Rice Husk Ash Coagulation and Electrocoagulation for Palm Oil Mill Effluent Treatment

Indriana Lestari[a*], Dwi Amalia[a], Widyawanto Prastistho[b], Jeremia Bernadin Perangin Angin[a], Muhammad Haekal Zenatik[a]

[a]*Department of Chemical Engineering, UPN Veteran Yogyakarta, Yogyakarta, 55283, Indonesia*

[b]*Department of Environmental Engineering, UPN Veteran Yogyakarta, Yogyakarta, 55283, Indonesia*

Article history:

Submitted 30 December 2024
Revision 23 May 2025
Accepted 26 June 2025
Online 27 June 2025

ABSTRACT: Palm oil mill effluent (POME) poses a significant environmental threat due to its high organic and inorganic load. This study introduces a novel integration of rice husk ash (RHA) coagulation and electrocoagulation (EC) for sustainable POME remediation. Thermally treated at 500 °C for two hours, RHA was characterized via FTIR, revealing active silica-based functional groups conducive to charge neutralization and adsorption. Treatment experiments employed 9.3 g/L RHA and aluminum electrodes spaced 20 mm apart under varying currents of 10, 15, and 20 A over 15, 30, and 45 minutes. At the highest tested condition (9.3 g/L RHA, 20 A, 45 minutes), the integrated process achieved 78% total solids (TS) and 43% chemical oxygen demand (COD) removal, surpassing individual RHA coagulation removed 34% TS and 17% COD, while EC alone achieved 43% TS and 18% COD removal. The superior performance stems from synergistic flocculation, adsorption, and electroflotation. Compared to conventional methods, the combined RHA–EC system offers faster treatment, lower chemical and energy demands, and improved sustainability. These findings suggest a scalable solution for decentralized POME treatment, particularly in resource-limited palm oil-producing regions.

Keywords: Coagulation; Electrocoagulation; Risce husk ash; Palm oil mill effluent; Combination treatment method

1. Introduction

Palm oil has become one of the most widely used vegetable oils globally due to its high productivity and versatility. It is extensively utilized in food products, cosmetics, biofuels, and industrial applications (Alhaji et al., 2024). Countries such as Indonesia and Malaysia dominate the global market, with Indonesia accounting for more than half of global production. This large-scale production, however, is associated with the generation of significant volumes of highly polluted wastewater, known as POME. As estimated, nearly 50% of the 5.0–7.5 metric tons (MT) of water used in the production of 1 MT of crude palm oil (CPO) is discharged as POME, underscoring the substantial volume of wastewater generated in palm oil processing (Mahmod et al., 2023; Yusof et al., 2023).

POME is typically discharged from multiple stages of palm oil processing, including sterilization, clarification, and hydrocyclone operations (Yusof et al., 2023). It appears as a thick, dark brown fluid containing high concentrations of organic and inorganic pollutants. It is composed primarily of water (95–96%), suspended solids (2–4%), and oils and grease (0.6–0.7%) (Mohammad et al., 2021). Its physicochemical characteristics are extreme: it exhibits an acidic pH ranging from 3.4 to 5.2, extremely high COD levels between 15,000 and 100,000 mg/L, biological oxygen demand (BOD) ranging from 10,250 to 43,750 mg/L, and TS concentrations between 11,500 and 79,000 mg/L, along with elevated turbidity. If discharged without adequate treatment, POME can cause severe ecological problems including oxygen depletion, eutrophication, and disruption of aquatic life (Vijayan et al., 2024).

Traditional treatment methods for POME have focused on biological approaches such as anaerobic and facultative pond systems due to their simplicity and ability to generate biogas as a renewable energy source. However, these systems require large land areas, long retention times, and precise microbial control. Additionally, they are often inefficient in removing recalcitrant compounds and cannot consistently meet environmental discharge standards, particularly in small or decentralized processing units. Physical methods such as sedimentation or filtration alone are inadequate due to the colloidal and emulsified nature of many contaminants in POME. In response to the limitations

*Corresponding Author
Email address: Indriana.lestari@upnyk.ac.id

of conventional systems, recent innovations have advanced thermal, membrane, and integrated zero-liquid-discharge (ZLD) technologies for water reclamation and resource recovery. Thermal techniques such as Multi-Effect Evaporation (MEE) and Mechanical Vapor Recompression (MVR) exploit phase change to concentrate solids and recover clean water, as implemented by industry leaders like Alfa Laval, which couples forced circulation evaporation with decanting to recover residual oil. Membrane technologies, such as Ultrafiltration (UF), Reverse Osmosis (RO), and thermally driven methods like Direct Contact Membrane Distillation (DCMD) and Vacuum Membrane Distillation (VMD) provide high-quality water output but demand extensive pretreatment to mitigate fouling from suspended solids and oil (Mahmod et al., 2023; Mohammad et al., 2021; Saad et al., 2021).

Integrated zero-discharge systems combine anaerobic and aerobic reactors with phytoremediation and membrane filtration to remove near-complete pollutants while producing value-added outputs such as biogas and biofertilizers. These advanced systems, often coupled with adsorption and membrane separation units, ensure regulatory compliance and align with sustainable water management goals by converting POME from a pollutant into a reusable resource. However, the deployment of such technologies is often restricted by high operational and capital costs, membrane fouling, and the need for complex system integration. Thus, pretreatment methods such as coagulation-flocculation and electrocoagulation remain essential for reducing turbidity, oil, and organic loads upstream. These approaches improve downstream treatment efficiency and offer simpler, decentralized solutions suitable for small- to medium-scale palm oil mills.

Recent studies have explored natural coagulants and electrochemical methods as more sustainable alternatives to conventional chemical treatments. Plant-based coagulants such as rambutan peel (*Nephelium lappaceum*), *Hylocereus undatus foliage*, and *fenugreek extract* have shown moderate performance in POME treatment. Rambutan achieves relatively low COD removal, approximately 28%, while Hylocereus undatus and fenugreek can reach around 60% removal under optimized conditions (Bunyamin et al., 2023; Som et al., 2023). However, these coagulants often require pH adjustment, are subject to seasonal availability, and may necessitate post-treatment steps to meet discharge standards, limiting their broader applicability in industrial settings.

One promising material is rice husk ash (RHA), a byproduct of rice milling and combustion. As a major rice producer, Indonesia generates over 10 million tons of rice husk annually (Wahyudianto et al., 2024). Controlled thermal treatment produces RHA rich in amorphous silica (~90% SiO_2) with favorable properties for pollutant adsorption and charge neutralization (Santana Costa & Paranhos, 2018; Zuwanna et al., 2023). Previous studies have shown that RHA can achieve 84% TS and 52% COD removal from wastewater through surface adsorption and silanol/siloxane-mediated coagulation mechanisms (Huzir et al., 2019). Nonetheless, RHA alone may not be sufficient for removing soluble organic compounds or ensuring rapid floc settling.

Electrocoagulation (EC) presents a complementary solution, operating through the electrochemical dissolution of metal electrodes to generate coagulant species in situ. In EC, aluminum or iron anodes release cations upon oxidation, while water electrolysis at the cathode produces hydrogen gas and hydroxide ions (Ingelsson et al., 2020; Nasrullah et al., 2022). The core redox reactions are:

Reduction reaction on the cathode:

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(aq)} \quad (1)$$

Oxidation reaction on the anode:

$$2H_2O_{(l)} \rightarrow O_{2(g)} + 4H^+_{(aq)} + 4e^- \quad (2)$$

$$M_{(s)} \rightarrow M^{n+}_{(aq)} + ne^- \quad (3)$$

(Boinpally et al., 2023; Nasrullah et al., 2022)

The resulting metal hydroxides (M) (e.g., $Al(OH)_3$, $Fe(OH)_3$) flocs adsorb and aggregate pollutants, while hydrogen bubbles assist in electroflotation (Bondarenko, 2021). EC is effective for treating various industrial effluents but can be limited by electrode passivation, energy consumption, and metal-rich sludge generation (Boinpally et al., 2023).

Recent studies have explored hybrid systems combining EC with plant-based coagulants to address these challenges. For instance, Garomsa et al. (2024) demonstrated improved pollutant removal and reduced energy input in brewery wastewater by coupling EC with indigenous bio-coagulants. However, applying RHA in synergy with EC for POME treatment remains underexplored, particularly in systematic studies addressing operational optimization.

The integration of RHA and EC called the C-EC system, offers potential synergistic benefits: initial colloid destabilization by RHA, enhanced floc formation and flotation via EC, and improved pollutant adsorption through functional group interactions. Using an agricultural by-product further aligns with circular economy principles and may reduce treatment costs and environmental burdens (Imam et al., 2025).

Therefore, this study aims to evaluate the performance of a combined rice husk ash coagulation and electrocoagulation (C-EC) process for treating palm oil mill effluent under varying operational conditions. Specifically, the objectives include (i) characterizing the surface functional groups of RHA through FTIR analysis, (ii) determining the effects of current intensity and treatment duration on the removal of TS and COD, and (iii) comparing the removal efficiencies of the C-EC system with standalone RHA and EC methods.

The hypothesis is that the integrated process will outperform individual treatments in removal efficiency and operational time due to the synergistic effects of physical-chemical–electrochemical interactions. This study contributes to filling the gap in existing literature regarding sustainable, low-cost hybrid treatments for industrial wastewater, particularly in regions with abundant

ISSN: 1410-394X
e-ISSN: 2460-8203

agricultural waste and limited access to centralized treatment facilities.

2. Materials and Methods

2.1. Preparation of RHA Coagulant

Rice husks were collected from a local agricultural supplier in Yogyakarta, Indonesia and thermally treated in an electric furnace (model: ThermoLab Muffle Furnace MF-12) at 500° C for 2 hours to produce an amorphous structure. After calcination, the ash was cooled at room temperature, ground using a ceramic mortar and pestle, and sieved through a 100-mesh screen (150 µm) to obtain uniform particle size distribution suitable for dispersion in water.

2.2. POME Sample

Fresh POME was obtained from the cooling pond of a local Indonesian palm oil company. The wastewater characteristics were analyzed according to the Indonesian National Standard (SNI), with pH measured using SNI 6989.11:2019 and COD following SNI 6989.73:2009. The raw POME was highly polluted, with initial characteristics including a pH of 4.5 and COD of 21,424 mg/L.

POME was diluted in a 1:1 volume ratio with distilled water to reduce viscosity and facilitate uniform mixing, mimicking primary clarification conditions. The dilution step was also necessary to prevent excessive current demand during electrocoagulation.

2.3 Experimental Procedure

All experiments were conducted in a batch reactor using a 1000 mL glass beaker (110 mm diameter × 140 mm height) containing 700 mL of pre-diluted POME. The reactor was equipped with four aluminum electrodes (60 mm × 30 mm × 2 mm), vertically arranged in a monopolar parallel configuration with 20 mm spacing between plates. The electrodes were immersed to a depth of 50 mm and connected to a DC power supply capable of delivering variable currents. A PTFE-coated magnetic stir bar was placed at the base of the beaker and operated continuously to ensure gentle agitation throughout the treatment process, minimizing floc breakage and promoting uniform dispersion. The experimental setup is illustrated in Figure 1.

Three treatment schemes were evaluated: RHA coagulation alone, EC alone, and the combined RHA–EC process (C-EC). In the RHA-only setup, 6.5 g of RHA was added to 700 mL of POME and stirred without electrical input. For EC-only treatment, no RHA was added, and current was applied at 20 A for varying durations. In the C-EC system, 6.5 g of RHA was added first and dispersed via magnetic stirring prior to current application. The combined system was tested under three current intensities (10 A, 15 A, and 20 A) and three time intervals (15, 30, and 45 minutes). A constant RHA dosage of 9.3 g/L was maintained for all combined treatment experiments. Continuous stirring was maintained in all modes to simulate mild flocculation conditions and ensure uniform dispersion of RHA.

2.2. Analytical Methods

Following treatment, samples were allowed to settle briefly and were then filtered prior to analysis. TS were measured using the gravimetric method according to APHA Standard Method. The samples were dried in a hot-air oven at 105 °C until a constant weight was achieved. COD was determined using HACH COD digestion vials, followed by spectrophotometric measurement at a wavelength of 620 nm. Measurements were performed once for each treatment condition due to experimental constraints. The removal efficiencies for TS and COD were calculated using the following equations:

$$\eta_{TS}\,(\%) = \left(1 - \frac{final\ weight}{initial\ weight}\right) \times 100 \quad (4)$$

$$\eta_{COD}\,(\%) = \left(1 - \frac{C_t}{C_o}\right) \times 100 \quad (5)$$

where Co and Ct represent COD concentrations in ppm prior to and following treatment, respectively.

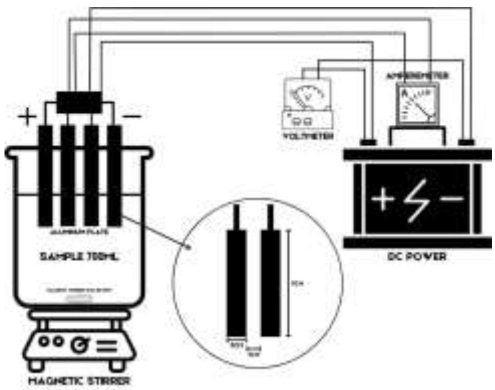

Figure 1. Schematic of electrocoagulation setup.

3. Results and Discussion

3.1. Functional Characterization of RHA

The surface chemistry of rice husk ash (RHA) was characterized using Fourier Transform Infrared (FTIR) spectroscopy to identify functional groups associated with coagulation activity.

As shown in Figure 2, broad absorption bands at 3409 cm⁻¹ and 3855 cm⁻¹ are attributed to O–H stretching vibrations, indicating the presence of surface hydroxyl groups, particularly silanol (Si–OH), as well as adsorbed water molecules. A distinct band at 1699 cm⁻¹ corresponds to carbonyl stretching, likely derived from residual lignin components. Additionally, a sharp peak at 1596 cm⁻¹ is associated with H_2O bending vibrations within the silica network.

The dominant peak at 1099 cm⁻¹ is indicative of asymmetric Si–O–Si stretching, characteristic of siloxane bridges in amorphous silica structures. This is further supported by the presence of a peak at 797 cm⁻¹, which reflects symmetric Si–O stretching vibrations. At lower wavenumbers, the peaks observed at 504, 461, and 413 cm⁻¹

are assigned to Si–O–Si bending modes, confirming the structural complexity of the silica matrix. Collectively, these spectral features confirm the high siliceous content and the abundance of reactive surface groups in RHA. These functional groups—particularly Si–OH and Si–O–Si—are known to facilitate electrostatic interactions, charge neutralization, and pollutant adsorption, thereby reinforcing the role of RHA as an effective and sustainable natural coagulant. (Santana Costa & Paranhos, 2018; Zuwanna et al., 2023).

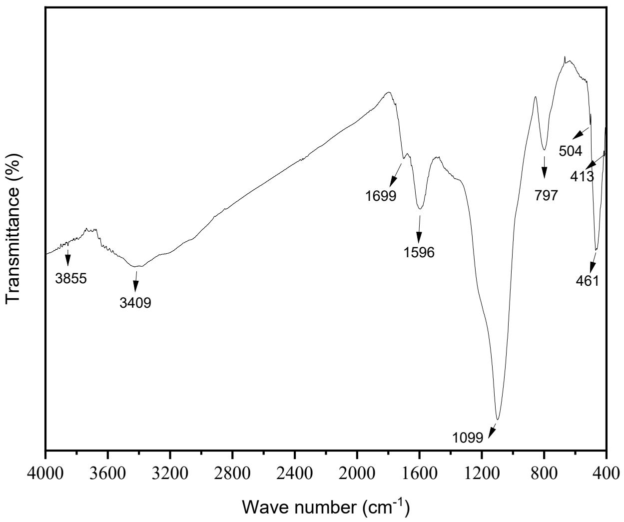

Figure 2. FTIR spectra of RHA

3.2. Performance of Individual and Combined Treatments

POME contains a substantial amount of negatively charged colloidal particles that resist natural sedimentation due to electrostatic repulsion.

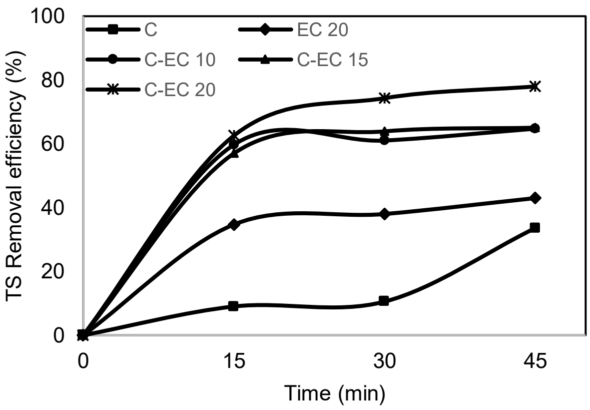

Figure 3. The effect of coagulation and combine coagulation-electrocoagulation on TS removal efficiency

Coagulants are used to neutralize these surface charges, allowing particles to aggregate into larger flocs that settle or float more easily. RHA has emerged as a sustainable alternative coagulant due to its silica content and surface-active functional groups that enhance charge neutralization and adsorption (Huzir et al., 2019). As shown in Figures 3 and 4, RHA coagulation alone achieved 34% TS and 17% COD removal at 9.3 g/L dosage and 45 minutes of settling. This moderate performance is attributed to surface interactions between negatively charged pollutants and the cationic or polar groups on the RHA surface.

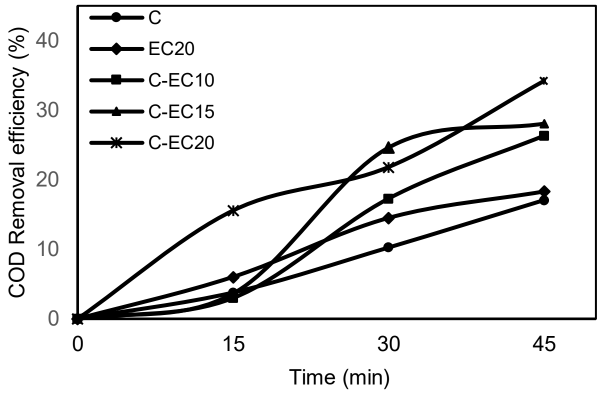

Figure 4. The effect of coagulation and combine coagulation-electrocoagulation on COD removal efficiency

In comparison, EC alone, performed at 20 A for 45 minutes, achieved slightly higher removal 43% for TS and 18% for COD. The increased performance stems from the electrochemical generation of Al^{3+} ions, which hydrolyze into amorphous $Al(OH)_3$ flocs that effectively destabilize and bind contaminants. Simultaneously, gas bubbles formed at the electrodes enhance pollutant separation via flotation. When RHA and EC were combined in the C-EC system (20 A, 45 minutes), removal efficiencies improved markedly to 78% for TS and 43% for COD. The significant increase in removal is attributed to the synergy between physical adsorption by RHA, chemical destabilization by Al^{3+}, and flotation facilitated by hydrogen bubbles. This combination allows more stable and denser flocs to form, which are more efficiently separated from the effluent.

The improved removal performance of the C-EC system is supported by the electrochemical and physicochemical mechanisms involved. During electrocoagulation, aluminum at the anode undergoes oxidation:

$$\text{Anode: } Al \rightarrow Al^{3+} + 3e^- \quad (6)$$

Simultaneously, water is reduced at the cathode, releasing hydrogen gas and hydroxide ions:

$$\text{Cathode: } 2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(aq)} \quad (7)$$

In solution, Al^{3+} ions hydrolyze:

$$Al^{3+} + 3OH^- \rightarrow Al(OH)_3 \quad (8)$$

The resulting $Al(OH)_3$ flocs act as primary coagulants by destabilizing colloidal particles and adsorbing organic matter. Meanwhile, hydrogen gas bubbles attach to the flocs, promoting flotation and separation.

In the presence of RHA, additional interactions occur. The silica surface (with Si–OH and Si–O–Si groups) offers

ISSN: 1410-394X
e-ISSN: 2460-8203

nucleation sites for floc growth and promotes bridging with aluminum hydroxides. Moreover, under the alkaline microenvironments created near the cathode, hydroxyl ions can initiate further polymerization of aluminum hydroxides into poly-hydrolytic species (Ingelsson et al., 2020). These polymers enhance entrapment of suspended solids and soluble organics.

Gas bubbles not only promote flotation but also provide oxidation potential. Oxygen generated at the anode may oxidize organic molecules either directly or indirectly, contributing further to COD reduction (Nasrullah et al., 2019). These interactions are illustrated in Figure 5, which schematically represents the synergistic mechanisms of pollutant destabilization, floc formation, and electroflotation in the combined RHA–EC system.

3.3. Effect of Current Intensity and Contact Time

Figure 3 and Figure 4 demonstrate the effects of varying current intensity and reaction time on TS and COD removal. Increasing current from 10 A to 20 A enhanced removal efficiency, consistent with higher generation rates of Al^{3+} ions and microbubbles. Similarly, longer contact times (up to 45 minutes) allowed more complete floc growth and interaction with coagulants, resulting in better sedimentation and flotation.

However, energy input and electrode degradation increase with current intensity and time, indicating that beyond a certain threshold, the process becomes less efficient. Future optimization should balance removal performance with operational sustainability, especially in full-scale applications.

3.4. Comparison with Prior Studies

The results obtained align with and improve upon previously reported performances. Huzir et al. (2019) achieved 52% COD and 84% TS removal using RHA alone, while Mohamad et al. (2022) reported 91.7% COD removal using EC alone, but only after 8 hours of treatment. In contrast, this study achieved 43% COD and 78% TS removal in just 45 minutes using the combined C-EC system, without requiring chemical additives or pH adjustments.

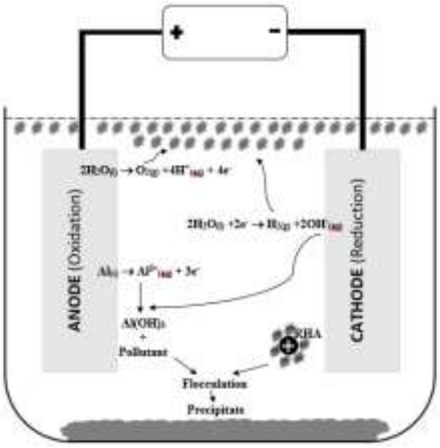

Figure 5. A diagram illustrating the integrated coagulation–electrocoagulation process within an electrochemical cell

Other plant-based coagulants such as *Nephelium lappaceum* and *Hylocereus undatus* showed 28–60% COD removal, often requiring pH correction and exhibiting batch-to-batch variability (Bunyamin et al., 2023; Som et al., 2023). RHA, being a stable industrial byproduct with high silica content, offers greater consistency and lower cost, making it a promising alternative when integrated with EC.

3.5. Limitations and Practical Considerations

While the C-EC system shows great promise for POME treatment, several limitations must be addressed before real-world application. The use of aluminum electrodes leads to sludge generation that must be properly managed to prevent secondary pollution. Although bench-scale energy consumption was moderate, future work must assess full-scale energy efficiency and electrode replacement costs.

This study also did not consider seasonal variation in POME composition, which could affect treatment efficiency. Moreover, as only a single trial was conducted for each condition, the results should be interpreted with caution. Further work is needed to statistically validate findings and evaluate system robustness under variable influent loads.

Nonetheless, the strong synergy observed between RHA and EC supports the potential scalability of the combined process. With further optimization, the C-EC system could serve as a sustainable and decentralized solution for POME treatment in palm oil-producing regions.

4. Conclusions

This study demonstrated the effectiveness of a combined rice husk ash (RHA) coagulation and electrocoagulation (EC) process for treating palm oil mill effluent (POME). RHA, characterized by its silanol and siloxane surface groups, contributed to charge neutralization and adsorption, while EC generated in situ aluminum hydroxide flocs and hydrogen gas bubbles that facilitated coagulation and flotation. Under optimal conditions (9.3 g/L RHA, 20 A, 45 minutes), the integrated RHA–EC (C-EC) system achieved 78% total solids (TS) and 43% chemical oxygen demand (COD) removal—surpassing standalone treatments. These improvements were attributed to synergistic mechanisms, including silica-based bridging, electrochemical destabilization, and flotation-enhanced separation.

Compared with conventional methods and other natural coagulants, the C-EC process offers a more time-efficient, sustainable, and scalable solution for POME pretreatment. The use of RHA, an abundant agricultural byproduct, supports circular economy principles by converting waste into a functional treatment agent. However, challenges remain related to sludge management, long-term electrode stability, energy use, and influent variability. Future studies should explore replicability, scale-up potential, and life-cycle impacts. Nevertheless, this method presents a viable option for decentralized palm oil wastewater management with reduced chemical inputs and enhanced treatment performance.

Acknowledgments
We are grateful to the Institute for Lembaga Penelitian dan Pengabdian kepada Masyarakat (LPPM) UPN Veteran Yogyakarta for providing financial support for this study No. 120/UN62.21/DT.07.00/2024

Statement
During the preparation of this work, the authors used ChatGPT-4o in order to improve the English language and proofread the text. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

CRediT authorship contribution statement
Indriana Lestari: Writing – review & editing, Writing – original draft, Resources, Methodology, Investigation, Formal analysis, Conceptualization.
Dwi Amalia: Writing – original draft, Methodology, Formal analysis, Conceptualization.
Widyawanto Prastistho: Writing – original draft, Methodology, Formal analysis, Conceptualization.
Jeremia Bernadin Perangin Angin: Investigation, Formal analysis, Conceptualization.
Muhammad Haekal Zenatik: Investigation, Formal analysis, Conceptualization.

Declaration of Competing Interest
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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