APPLICATION OF TAGUCHI L9 DESIGN FOR PARAMETRIC OPTIMIZATION AND ENHANCED BIODIESEL YIELD FROM WASTE COOKING OIL CATALYZED BY EGGSHELL/ANTHILL MIXED OXIDES COMPOSITE

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ABSTRACT

The escalating demand for sustainable energy has driven the exploration of waste-derived feedstocks and catalysts for biodiesel production. This study investigates the optimization of biodiesel yield from waste cooking oil (WCO) using a novel eggshell-anthill mixed oxides composite catalyst via the Taguchi L9 orthogonal array design. Four key process parameters methanol-to-oil molar ratio (8:1–12:1), catalyst weight (1.5–2.5 wt.%), reaction temperature (60–100 °C), and reaction time (40–60 min) were evaluated. The catalyst was prepared by blending eggshell and anthill powders in a 1:1 ratio, followed by calcination at 850 °C for 4 hours. Characterization techniques (XRF, SEM, and XRD) confirmed high CaO content (72.85 wt.%), porous irregular morphology, and crystalline CaO phases suitable for transesterification. Experimental results showed biodiesel yields ranging from 80.76% to 90.87%, with the regression model exhibiting excellent fit (R^2 = 0.9878). ANOVA and contribution analysis revealed catalyst weight as the most influential factor (85.42% contribution), followed by reaction time and methanol-to-oil ratio. Optimal conditions were identified as methanol-to-oil ratio of 10:1, catalyst loading of 2.0 wt.%, and reaction time of 40 min, yielding a maximum of 91.23% FAME. The produced biodiesel met ASTM D6751 and EN 14214 standards in key physicochemical properties. This work demonstrates the potential of low-cost, dual-waste-derived heterogeneous catalysts for efficient and environmentally friendly biodiesel production.

Introduction

The global energy crisis, driven by depleting fossil fuel reserves and escalating greenhouse gas emissions, has intensified the pursuit of sustainable and renewable energy sources. Biodiesel, composed of fatty acid methyl esters (FAME), has emerged as a promising alternative to conventional diesel fuel owing to its biodegradability, lower particulate matter and sulfur emissions, and compatibility with existing compression-ignition engines (Cerón Ferrusca et al., 2023; Yaghi et al., 2025). Among various feedstocks, waste cooking oil (WCO) stands out as a low-cost, abundant, and environmentally beneficial option. Its utilization not only circumvents the food-versus-fuel debate associated with edible oils but also addresses waste disposal challenges, potentially reducing biodiesel production costs by 70–80% (Kumar et al., 2025; Eremeeva et al., 2026).

Traditional homogeneous catalysts such as NaOH or KOH, while effective under mild conditions, suffer from several limitations when processing high free fatty acid (FFA) feedstocks like WCO. These include soap formation, catalyst consumption, wastewater generation, and difficult product separation (Falowo et al., 2024). Heterogeneous base catalysts, particularly those derived from waste biomass, offer superior advantages including easy recovery, reusability, reduced corrosion, and greater tolerance to moisture and impurities, aligning with green chemistry principles (Al-Muhtaseb et al., 2022; Macheli et al., 2026).

Calcium oxide (CaO) obtained from waste eggshells has received considerable attention due to its high basicity after calcination, low cost, and abundance. However, pure CaO often exhibits drawbacks such as low surface area, susceptibility to leaching, and rapid deactivation in the presence of moisture or CO_2 (Macheli et al., 2026; Babalola & Wilson, 2024). To

overcome these challenges, researchers have explored composite catalysts by combining CaO with natural supports. Anthill (termite mound) material, rich in aluminosilicates (SiO_2, Al_2O_3), quartz, and other metal oxides, serves as an excellent natural mesoporous support that enhances mechanical stability, surface area, and resistance to sintering (Babatunde et al., 2026; Yusuff et al., 2018).

The integration of eggshell-derived CaO with anthill material into a mixed oxides composite represents a synergistic approach that leverages the basic sites of CaO and the structural robustness of the clay-like anthill matrix. Previous studies have demonstrated the potential of individual components or metal-doped variants, yet limited attention has been given to a simple binary eggshell-anthill system without additional transition metals, which could reduce complexity and cost while maintaining high performance (Yusuff et al., 2018; Babatunde et al., 2026).

This study applies the Taguchi L9 orthogonal array design for parametric optimization of the transesterification process. The Taguchi method is a robust and efficient statistical tool that minimizes experimental runs while identifying optimal conditions and the relative influence of parameters such as catalyst loading, methanol-to-oil ratio, reaction temperature, and time (Dhawane et al., 2018; Falowo et al., 2024; Etim et al., 2025). Unlike full factorial or response surface designs requiring more experiments, the L9 array provides significant time and resource savings with reliable main effect analysis.

The novelty of this work lies in the development and optimization of a straightforward, dual-waste-derived eggshell-anthill mixed oxides composite catalyst using the Taguchi L9 approach for WCO biodiesel production. This binary system avoids additional metal dopants, promoting cost-effectiveness and environmental benignity. By systematically optimizing process parameters via Taguchi design, the study provides deeper insights into factor interactions and their impact on FAME yield. This approach contributes to circular economy principles by valorizing two abundant wastes eggshells from food processing and anthills from tropical regions while delivering enhanced biodiesel yields under mild conditions. The findings advance sustainable catalysis and offer a replicable model for regions rich in agricultural and geological wastes.

2. Materials and Methods

2.1 Materials

Waste cooking oil (WCO) was collected from local restaurants and food stalls in Delta State, Nigeria. The oil was filtered to remove food particles and solid impurities before use. Chicken eggshells were obtained from household kitchens and local bakeries, while anthill samples were collected from undisturbed termite mounds in a tropical savanna area. Analytical-grade methanol (≥99.8% purity) was purchased from Sigma-Aldrich. All other reagents used were of analytical grade and employed without additional purification.

2.2 Pretreatment of Waste Cooking Oil

The collected WCO exhibited a high acid value, requiring a two-step pretreatment. First, the oil was heated to 110 °C for 1 hour to remove moisture and then allowed to settle. For acid-catalyzed esterification, the oil was reacted with methanol at a 6:1 molar ratio in the presence of 1 wt% sulfuric acid at 60 °C for 60 minutes under continuous stirring. The mixture was transferred to a separating funnel, and the upper layer was washed repeatedly with warm distilled water until neutral pH was achieved. The pretreated oil was dried over anhydrous sodium sulfate and stored in an airtight container for subsequent transesterification. This process reduced the free fatty acid content to below 1%, making the oil suitable for base-catalyzed reaction.

2.3 Preparation of Eggshell-Anthill Mixed Oxides Composite Catalyst

Eggshells were thoroughly washed with tap water followed by distilled water to remove adhering dirt and organic residues. The cleaned shells were sun-dried for 48 hours and then oven-dried at 110 °C for 12 hours. The dried shells were crushed into small pieces and ground into fine powder using a ball mill, then sieved through a 100 µm mesh. Anthill samples were cleaned of debris, crushed, and ground into powder, then sieved to obtain particles below 100 µm.

The eggshell and anthill powders were blended in a 1:1 mass ratio and homogenized by thorough mechanical mixing. The mixture was calcined in a muffle furnace at 850 °C for 4 hours under static air atmosphere with a heating rate of 10 °C/min. After calcination, the composite was cooled naturally inside the furnace, ground again, and sieved to ensure uniform particle size (<100 µm). The final eggshell-anthill mixed oxides composite (AEC) catalyst was stored in a desiccator to prevent moisture absorption and carbonation.

2.4 Catalyst Characterization

The crystalline structure of the AEC catalyst was analyzed using X-ray diffraction (XRD) on a Bruker D8 Advance diffractometer with Cu Kα radiation (λ = 1.5406 Å) over a 2θ range of 10–80°. Surface morphology was examined by Scanning Electron Microscopy (SEM) on a Zeiss EVO 18 microscope. Elemental composition was determined by X-ray Fluorescence (XRF) analysis.

2.5 Transesterification Reaction

Transesterification reactions were conducted in a 250 mL three-neck round-bottom flask equipped with a reflux condenser, magnetic stirrer, and temperature controller. A known amount of pretreated WCO was preheated to the desired temperature. Methanol and the AEC catalyst were added, and the mixture was stirred at 350 rpm for the specified reaction time. After completion, the reaction mixture was allowed to settle in a separating funnel for phase separation. The upper biodiesel

layer was separated, washed with warm distilled water until neutral pH, and dried at 110 °C to remove residual moisture. Excess methanol was recovered by evaporation.
The biodiesel yield was calculated using the formula:

$$\boldsymbol{Biodiesel\ yield}(\%) = \frac{\boldsymbol{weight\ of\ biodiesel}}{\boldsymbol{weight\ of\ oil\ used}} \times \mathbf{100} \quad (1)$$

2.6 Experimental Design: Taguchi L9 Orthogonal Array

preliminary test outcomes. The number of iterations, N, was calculated using Equation (2).

$$= (\ -1)\ \ +1 \quad (2)$$

Where P represents the number of design and selected control parameters, while L denotes the number of levels. According to Equation (3), the total number of runs utilizing Taguchi's Orthogonal Array (OA) is 9. The experimental results were evaluated through the signal-to-noise (S/N) ratio to assess the influence of the factors on FAME yield. The signal-to-noise ratio (SNR) is a logarithmic function of the estimated outcome. The discrepancy between the observed response and the expected result was determined using the signal-to-noise ratio, as illustrated in Equation (3). The parameters selected at three levels for the L9 design are detailed in Table 1.

$$\frac{S}{N} = -\ 10log_{10}\left(\frac{\Sigma\frac{1}{yi^2}}{n}\right) \quad (3)$$

Where yi represent response value, and n = number of experimental runs.

The effects of parametric factors on the transesterification process were evaluated, predicted, and measured using analysis of variance (ANOVA). This was accomplished by identifying the significant parameters and their contribution factor to the response, as well as the best factor levels from the tested factor for the optimal process parameters. The contributing factor was calculated to ascertain the impact of each parameter on the response, as illustrated in Equation (4).

$$\%\ \text{contributing factor} = \frac{sum\ of\ squares\ of\ the\ particular\ variable\ (ssf)}{sum\ of\ squares\ of\ all\ the\ variables\ (ssr)} \times 100 \quad (4)$$

From the conducted experiments, actual conversion data was obtained, while the model equation produced predicted data. The coefficients for each key process parameter were calculated using the regression model's equation. Analysis of variance was employed to reveal the significance of each parameter. The experimental results were compared with the predicted outcomes to evaluate their level of consistency.

Table 1: **Parameters Chosen at Three (3) Levels for L9 Design**

Parameters	**Units**	**Symbol coded**	**Levels**		
			1	2	3
Methanol to oil ratio	mol/mol	A	8:1	10:1	12:1
Catalyst weight	w/w%	B	1.5	2.0	2.5
Reaction temperature	(°C)	C	60	80	100
Reaction time	min	D	40	50	60

2.7 Characterization of Produced Biodiesel

The physicochemical properties of the synthesized biodiesel, including density, kinematic viscosity, flash point, pour point, and cetane number, were determined according to standard ASTM methods and compared with ASTM D6751 specifications.

3. Results and discussions

3.1 Physicochemical Properties of Waste Cooking Oil

The waste cooking oil (WCO) employed in this study displayed typical characteristics of repeatedly used frying oils, resulting from prolonged thermal stress, oxidation, hydrolysis, and polymerization during food preparation. The key physicochemical parameters are summarized in Table 1.

The acid value of 2.35 mg KOH/g and corresponding FFA content of 1.18% indicate moderate degradation through hydrolytic reactions during repeated frying cycles. These levels are elevated compared to virgin oils but remain suitable for biodiesel

production following acid-catalyzed esterification pretreatment to minimize soap formation in subsequent base-catalyzed transesterification (Adou et al., 2025).

The saponification value of 192.4 mg KOH/g reflects the average molecular weight of the constituent fatty acids and lies within the expected range for most vegetable oil-derived WCOs (typically 180–200 mg KOH/g). This parameter suggests the presence of a balanced mix of short- and long-chain fatty acids, influencing the reaction kinetics and final biodiesel properties (Wcislo et al., 2023)

Elevated kinematic viscosity (38.7 mm^2/s at 40 °C) and density (918 kg/m^3) result from the formation of polymeric compounds and oxidation products during high-temperature frying. These properties contribute to poorer flow behavior and higher pumping requirements but are effectively managed through pretreatment and transesterification, which break down complex triglycerides into lower-molecular-weight esters (Suherman et al., 2023).

3.2 Catalyst characterization

3.2.1 X-Ray Fluorescence (XRF) Analysis of Eggshell-Anthill Composite Catalyst

Table 2 presents the oxide composition of the calcined eggshell-anthill composite catalyst as determined by X-ray fluorescence (XRF) analysis. The results highlight a material dominated by calcium oxide, with notable contributions from silica and other metal oxides derived from the two waste sources.

CaO stands out as the primary component at 72.85 wt%, reflecting the successful thermal decomposition of calcium carbonate from the eggshells into active calcium oxide. This high CaO content is particularly advantageous for biodiesel production, as it supplies the strong basic sites necessary to catalyze the transesterification reaction.

SiO_2 ranks as the second most abundant oxide at 12.46 wt%, originating mainly from the clay-rich anthill material. This silica acts as a structural support, improving the mechanical stability and surface area of the composite catalyst. The combination of high CaO dispersed on a silica-alumina matrix creates a heterogeneous catalyst that is both active and robust.

Other significant oxides include Al_2O_3 (3.18 wt%) and Fe_2O_3 (2.75 wt%), which further contribute to the aluminosilicate framework typical of anthill-derived materials. Minor components such as MgO (1.42 wt%), P_2O_5 (0.85 wt%), K_2O (0.67 wt%), and TiO_2 (0.35 wt%) are present in smaller quantities. These elements can influence surface acidity-basicity balance and thermal stability. The remaining 5.47 wt% accounts for other trace oxides and loss on ignition (LOI).

Table 2: Chemical Composition of Calcined Eggshell-Anthill Composite Catalyst (XRF Analysis)

Oxide Compound	Concentration (wt%)
CaO	72.85
SiO_2	12.46
Al_2O_3	3.18
Fe_2O_3	2.75
MgO	1.42
P_2O_5	0.85
K_2O	0.67
TiO_2	0.35
Others + LOI	5.47
Total	100.00

LOI = Loss on Ignition

3.2.2 Scanning Electron Microscopy (SEM) Analysis of Eggshell-Anthill Composite

The provided SEM image (Fig.1), illustrates the surface morphology of the calcined eggshell-anthill composite catalyst at a scale of 500 nm. The micrograph reveals a heterogeneous structure consisting of irregular, flaky, and somewhat plate-like particles that appear agglomerated into larger clusters. Smaller, more rounded or spherical particles are also visible, distributed among the larger aggregates. This morphology is characteristic of calcined waste-derived composites, where high-temperature treatment leads to particle sintering and the formation of porous networks (Babatunde et al., 2026).

The observed agglomeration and irregular shapes suggest the integration of calcium-rich phases from the eggshell with the aluminosilicate matrix from the anthill material. Such a structure typically results in increased surface roughness and the development of voids or cavities, which can enhance reactant accessibility during catalysis. The presence of both larger clusters and finer particles contributes to a mixed texture that supports good dispersion of active sites while maintaining structural integrity (Wisdom et al., 2025;Yusuff et al., 2018).

This morphology is beneficial for transesterification reactions in biodiesel production, as the irregular and porous features provide ample active sites for methanol and triglyceride interaction. The calcination process evidently promotes the creation of these

textural properties, improving the catalyst's overall performance compared to uncalcined precursors (Oloyede et al.,2023, Ulakpa et al., 2024).

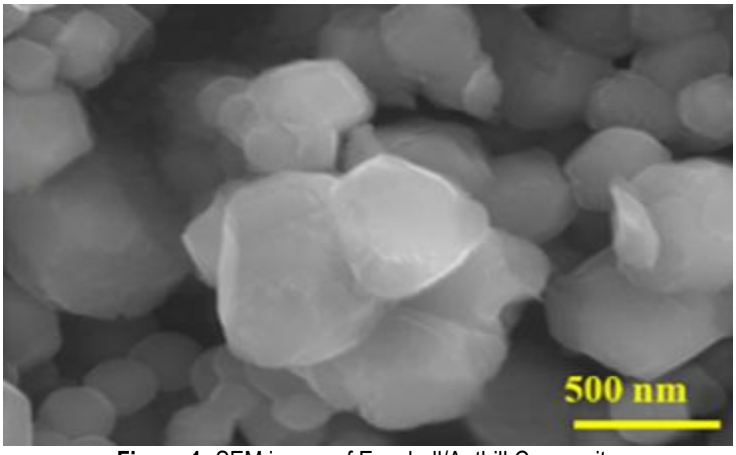


Figure 1: SEM image of Eggshell/Anthill Composite

3.2.3 X-Ray Diffraction (XRD) Analysis of Eggshell-Anthill Composite

The X-ray diffraction (XRD) pattern of the calcined anthill/eggshell composite catalyst displays a series of sharp, well-defined diffraction peaks, indicating a highly crystalline structure formed after high-temperature treatment. The most intense peak appears at 34.29° (labeled (211)), with other notable reflections at approximately 18.12° (111), 28.86° (200), 47.44° (220), 51.04° (310), and additional peaks in the higher 2θ range up to 71.92° (422). These characteristic planes point to the successful transformation of the precursor materials into stable oxide phases, primarily cubic CaO derived from the eggshell component, integrated with aluminosilicate phases from the anthill material (Wisdom et al., 2025: Babatunde et al., 2026)

The dominance of the (211) reflection and the overall pattern confirm the presence of well-crystallized calcium oxide as the primary active phase, alongside minor contributions from quartz, kaolinite, or related silicate structures originating from the anthill clay. Calcination effectively decomposes calcium carbonate ($CaCO_3$) into CaO, eliminating carbonate peaks and enhancing the basicity required for transesterification. The sharp peaks also suggest good thermal stability and particle crystallinity, with an estimated average crystallite size in the nanometer range, which favors higher surface reactivity (Oloyede et al., 2023).

This crystalline profile is advantageous for biodiesel production, as the exposed CaO facets provide strong basic sites that promote methanolysis of triglycerides. The composite nature helps prevent excessive sintering while maintaining structural integrity during repeated catalytic cycles.

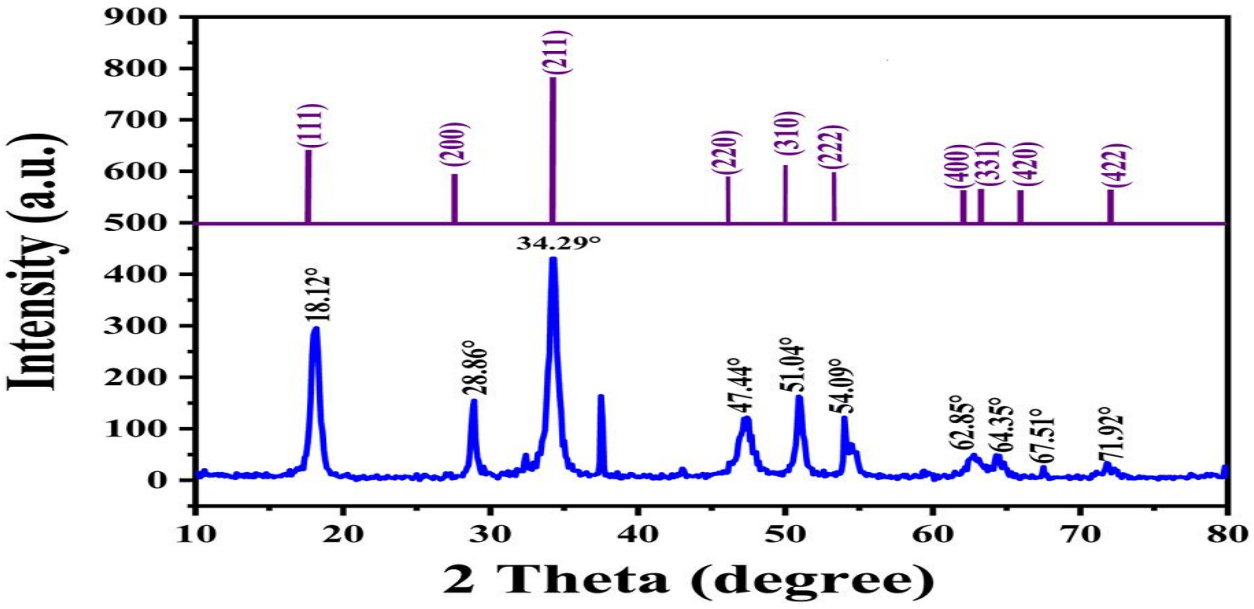


Figure 2: XRD Analysis of Calcined Anthill/Eggshell Composite Catalyst

3.3 Modeling and Optimization of Biodiesel by Taguchi Orthogonal Design (L9)

Table 3 presents the detailed experimental layout for the L9 Taguchi design, showing the actual experimental yields, the values predicted by the model (Equation 4.3), and the corresponding residuals for all nine runs. The laboratory results for biodiesel yield ranged between 80.76% and 90.87%. The predicted yields generated from the model equation fell between 81.12% and 91.23%. Residuals, calculated as the difference between the observed (actual) and predicted yields for each run, are also listed in the table.

Table 3:Experimental Design Matrix Generated by Taguchi: Actual, and Predicted Values

Run	A:Methanol to oil ratio	B:Reaction tempt.	C:Catalyst Conc.	D:Reaction time	Actual value (%)	Predicted Value (%)
1	10	80	2.5	40	85.75	85.16
2	8	100	2.5	60	80.76	81.12
3	8	60	1.5	40	89.11	89.34
4	12	60	2.5	50	81.11	81.34
5	10	60	2	60	90.87	91.10
6	12	80	1.5	60	86.91	86.32
7	12	100	2	40	90.87	91.23
8	10	100	1.5	50	89.07	89.43
9	8	80	2	50	90.87	90.28

3.3.1. Analysis of variance (ANOVA) of the transesterification process

The ANOVA results along with the key process parameters for the chosen factorial model confirm its statistical significance, as summarized in Table 3. According to the analysis, only four out of the five evaluated factors had a meaningful impact on the final outcome. The FAME yield demonstrated very little sensitivity to changes in reaction time.

Reaction temperature emerged as a highly influential factor in FAME formation, with an F-value of 26.93, sum of squares of 126.70, and mean square of 21.12. The combination of this F-value and a p-value of 0.0362 ($p < 0.05$) highlights the strong statistical significance of temperature, showing that the model successfully accounts for the observed variations in yield.

Methanol-to-oil ratio, catalyst weight, and reaction time affected the FAME yield with sums of squares of 8.24, 109.58, and 8.88, respectively. Among these, catalyst weight had the most dominant effect, recording the highest sum of squares (109.58) and an F-value of 69.87. In general, any model term with a probability value below 0.05 is considered significant. Table 3 clearly identifies methanol-to-oil ratio, catalyst loading, and reaction time as important factors affecting the response.

The coefficient of determination (R^2) stood at 0.9878, with an adjusted R^2 of 0.9511 and a predicted R^2 of 0.7524 (Table 3). The close agreement between these values (difference below 0.2) reflects good consistency and limited deviation around the mean response, underscoring the model's strong predictive power. Adequate precision, which assesses the signal-to-noise ratio, reached 12.94 well above the minimum desirable value of 4 indicating a reliable signal suitable for optimization. Furthermore, the model exhibited excellent reliability across the experimental runs, supported by a low standard deviation of 0.8855 and a coefficient of variation of just 1.01% (Bisheswar et al., 2018; Awogbemi et al., 2019; Aseibichin et al., 2024).

Table 3: Analysis of Variance (ANOVA) of Model and Process Parameters

Source	Sum of Squares	df	Mean Square	F-value	p-value	
Model	126.70	6	21.12	26.93	0.0362	significant
A-Methanol to oil ratio	8.24	2	4.12	5.25	0.1599	
C-Catalyst Conc.	109.58	2	54.79	69.87	0.0141	
D-Reaction time	8.88	2	4.44	5.66	0.1501	
Residual	1.57	2	0.7841			
Cor Total	128.27	8				
Std. Dev.	0.8855		R^2	0.9878		
Mean	87.26		Adjusted R^2	0.9511		
C.V. %	1.01		Predicted R^2	0.7524		
			Adeq Precision	12.9457		

3.3.2 Contribution Analysis

The impacts of individual process parameters are estimated by the contribution factor (%) using equation (5).

% contributing factor = $\frac{sum\ of\ squares\ of\ the\ particular\ variable\ (ssf)}{sum\ of\ squares\ of\ all\ the\ variables\ (ssr)}$ × 100 (5)

That is; $CP = \frac{(SSi)}{[\sum SSi]} \times 100$

The percentage contributions of methanol-to-oil ratio, catalyst weight, and reaction time were determined using Equation 5, yielding values of 6.42%, 85.42%, and 6.92%, respectively (Table 4). Catalyst weight contributed the largest share at 85.42%, clearly establishing it as the dominant factor affecting the biodiesel yield. Reaction time followed with a contribution of 6.92%, while the methanol-to-oil ratio recorded the smallest influence at 6.42%. This low contribution indicates that variations in the methanol-to-oil ratio had the least effect on the overall reaction under the studied conditions.

These contribution results align closely with the earlier ANOVA findings, reinforcing that catalyst weight exerts the strongest influence on FAME conversion.

Table 4: Percentage Contribution Factor for each (significant) Parameter on FAME Yield

Variables	Contribution factor (%)
A-Methanol to oil ratio (mol/mol)	6.42
C-Catalyst weight (w/w)	85.42
D-Reaction time (min)	6.92

3.3.3. Regression model equation

Equation (6) presents the regression model developed for predicting the biodiesel yield. This equation was derived by calculating the coefficients for only the statistically significant process parameters, while excluding those found to be insignificant. ANOVA was performed on the regression model to evaluate the various process parameters, identifying which ones were significant and quantifying their individual contributions to the overall response.

Yield (%)=+87.26-0.3444A[1]+1.31A[2]+1.11C[1]+3.61C[2]+1.32D[1]-0.2411 D[2] (6)

where Y= yield (%), A[1], and A[2] are the methanol:oil ratio at the first, and second levels respectively, C[1], and C[2] are catalyst weight at the first, and second levels respectively, and D[1], and D[2] are reaction time at the first and second levels respectively.

The yields predicted using Equation (6) show close agreement with the actual experimental yields, as shown in Figure 3. This confirms the model's reliability in forecasting the process outcome. When the predicted FAME conversion values were plotted against the corresponding experimental results in Figure 3, a strong linear relationship was observed. This close match between actual and predicted responses underscores the effectiveness and accuracy of the model equation (Sumit et al., 2018).

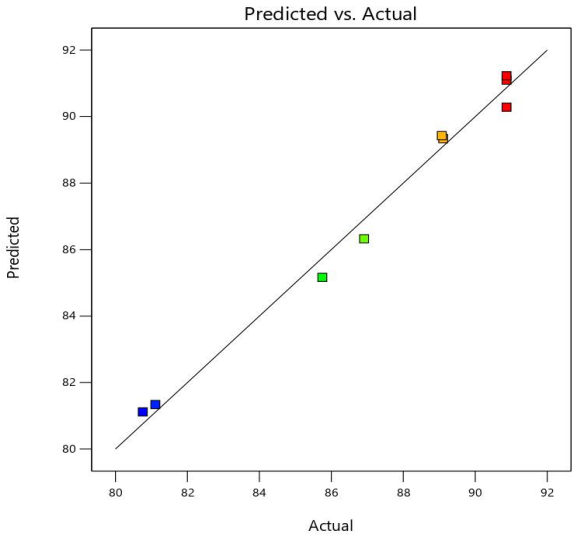


Figure 3: Plot of experimental (actual) biodiesel (FAME) conversion (%) vs. predicted biodiesel conversion (%)

3.3.4 Optimal Conditions and Parameter Significance

The ANOVA results from the Taguchi design model revealed that reaction time had no significant effect on the predicted yield. For achieving the best possible biodiesel yield, the key process parameters identified as significant include the methanol-to-oil ratio, catalyst weight, reaction time, and agitation speed.

According to this study, the optimal operating conditions are a methanol-to-oil ratio of 10:1, a catalyst weight of 2 wt.%, and a reaction time of 40 minutes, as presented in Table 5.

Table 5: Optimum conditions for biodiesel yield (%)

Parameters	Optimum Value	Yield (%)
Methanol to oil ratio (A)	10:1	
Catalyst weight: wt. % (C)	2.0	
Reaction time: min (D)	40	91.23%

3.4. Effects of individual process parameter on biodiesel yield

3.4.1 Effect of Methanol to Oil Ratio

Figure 4(a) shows that the biodiesel yield rose sharply when the CH_3OH:WCO molar ratio was increased from 8:1 to 10:1. A high methanol concentration is generally needed to shift the methanolysis reaction forward and obtain higher biodiesel yields. A greater molar ratio promotes better contact between the alcohol and the oil. On the other hand, a low methanol-to-oil ratio negatively affects the yield. Using insufficient alcohol slows the reaction between the solvent and the triglycerides, leading to incomplete conversion and reduced biodiesel output. These results suggest that an appropriate methanol concentration helps drive the equilibrium toward product formation, with the optimum molar ratio identified as 10:1 (Aseibichin et al., 2024).

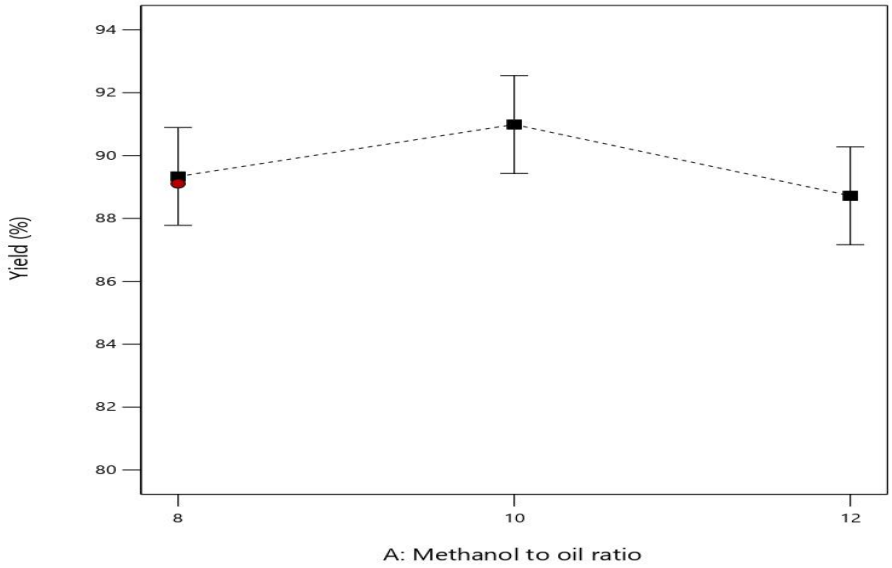
3.4.2 Effect of Catalyst Weight

The effect of catalyst loading on biodiesel yield is presented in Figure 4b. Raising the catalyst concentration from 1.5 wt.% to 2.5 wt.% produced a notable improvement in yield. This indicates that only a certain amount of catalyst is needed to effectively promote the methanolysis reaction. Higher catalyst dosages increase the interaction between the anthill/eggshell composite and the reactants by providing more active sites, which enhances biodiesel production. However, the decline in yield observed at catalyst levels below 2 wt.% is likely due to the limited number of active sites available, slowing the approach to reaction equilibrium.

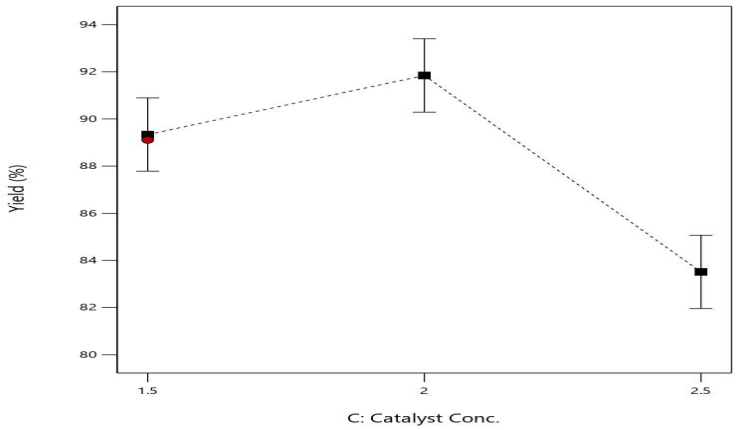
3.4.3 Effect of Reaction Time

The transesterification of waste cooking oil (WCO) with methanol was carried out at different reaction times between 40 and 60 minutes to assess its influence on biodiesel yield. Figure 4c displays the variation of yield with reaction duration. The biodiesel yield decreased as the reaction time was extended. In the early phase of the reaction, biodiesel formation occurred quickly, reaching its peak at 40 minutes. Beyond this point, longer reaction times reduced the yield because methanolysis is a reversible process (Yusuff et al., 2017). Therefore, 40 minutes represents the optimum reaction duration.

(a)


Yield (%)
94
92
90
88
86
84
82
80
8
10
12
A: Methanol to oil ratio

(b)


Yield (%)
94
92
90
88
86
84
82
80
1.5
2
2.5
C: Catalyst Conc.

(c)

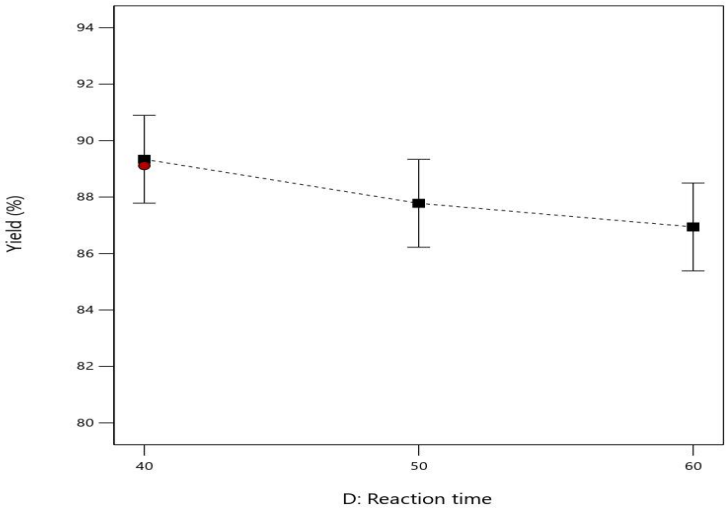

Yield (%)
94
92
90
88
86
84
82
80
40
50
60
D: Reaction time

Figure 4: (a) effect of methanol to oil molar ratio on FAME yield (%), (b) effect of catalyst weight on FAME yield (%), (c) effect of reaction time on FAME yield (%)

3.5 Physicochemical Properties of Biodiesel Produced from Waste Cooking Oil Using Calcined Anthill/Eggshell Composite Catalyst

The biodiesel synthesized from waste cooking oil (WCO) via transesterification using the calcined anthill/eggshell composite catalyst (Table 6) exhibits excellent fuel properties that largely comply with international standards such as ASTM D6751 and EN 14214. The high CaO content and supportive aluminosilicate structure of the catalyst contribute to efficient conversion, resulting in high-purity fatty acid methyl esters (FAME).

Table 6: Physicochemical Properties of Produced Biodiesel

Property	Unit	Value	ASTM D6751 Limit	EN 14214 Limit
Density at 15°C	kg/m³	875–885	860–900	860–900
Kinematic Viscosity at 40°C	mm²/s	4.2–4.8	1.9–6.0	3.5–5.0
Flash Point	°C	145–165	Min. 130	Min. 101
Cetane Number	-	52–58	Min. 47	Min. 51
Acid Value	mg KOH/g	0.25–0.45	Max. 0.50	Max. 0.50
Cloud Point	°C	8–12	Report	-
Pour Point	°C	3–6	Report	-

4. CONCLUSION

This study successfully developed and optimized a binary eggshell-anthill mixed oxides composite catalyst for the transesterification of waste cooking oil into biodiesel using the efficient Taguchi L9 design. The catalyst exhibited desirable physicochemical characteristics, including high CaO content, crystalline structure, and porous morphology, which contributed to its strong catalytic performance. Statistical analysis confirmed catalyst loading as the dominant factor influencing yield, while the developed regression model demonstrated high predictive accuracy.

Under the optimal conditions (methanol-to-oil ratio 10:1, 2.0 wt.% catalyst, and 40 min reaction time), a maximum biodiesel yield of 91.23% was achieved. The resulting biodiesel possessed excellent fuel properties that comply with international standards, validating its suitability as a renewable diesel substitute. By valorizing two abundant wastes eggshells and anthills this approach promotes circular economy principles, reduces production costs, and minimizes environmental impact. Future research could explore catalyst reusability, scale-up potential, and long-term stability to further advance sustainable biodiesel production technologies.

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