



Article Optimization of Biodiesel Production from Waste Cooking Oil Using a Green Catalyst Prepared from Glass Waste and Animal Bones

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Abstract: Biodiesel as a fuel has been shown to positively impact the environment; replacing or reducing the dependence on fossil fuels while providing a viable alternative. The use of waste oils, such as non-edible or used oils, can reduce competition with food, loss of resources, and the resulting higher prices. In this study, biodiesel was obtained by a transesterification reaction using used cooking oil from fast-food restaurants as the feedstock and catalysts from waste glass and animal bones as the silica and calcium oxide sources, respectively. Utilizing waste or non-edible oils for the production of biodiesel can lessen the competition with food sources while achieving environmental and ethical biofuel standards. Additionally, employing readily available waste oils and catalysts prepared from waste material is an economical and low-cost process compared to the use of conventional expensive feedstock and catalyst. The catalyst characterization for the prepared CaO-SiO₂ catalyst was performed using X-ray diffraction (XRD), scanning electron microscopy (SEM), and Fourier transform infrared spectroscopy (FT-IR). The reaction was optimized using the response surface methodology (RSM) with central composite design (CCD) by varying three parameters: methanol-to-oil ratio, catalyst weight fraction (wt%), and reaction time. The highest biodiesel yield obtained using Design Expert software was 92.3419% at the optimum conditions of a 14.83:1 methanol-to-oil molar ratio, 3.11 wt% catalyst, and 143 min reaction time. This proved that waste cooking oil with CaO-SiO₂ catalyst could be used in the transesterification process to produce a high yield of biodiesel, which was shown in the results obtained from the experimental runs.

Keywords: biodiesel; transesterification; waste cooking oil; heterogeneous catalyst; response surface methodology; optimization

1. Introduction

Energy consumption and demand are increasing worldwide to meet the growing needs of a growing population [1]. As fossil fuels make up most of the energy sector, serious environmental and health problems associated with greenhouse gas emissions are rising [2,3]. Greenhouse gases mainly include carbon dioxide and methane, which are among the main causes of global warming, leading to an increase in temperature, melting of ice, and thus higher sea levels and flooding [4]. The recent floods in Pakistan in 2022 were the deadliest in a series of stunning weather extremes in the Northern Hemisphere that have killed thousands and forced 33 million people from their homes [5]. Other harmful gasses produced by fossil fuel combustion include nitrogen monoxide and dioxide, sulfur



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Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). dioxide, and carbon monoxide. These gasses cause smog that damages human health and plant growth, and acid rain, which is mainly caused by sulfur dioxide [6,7]. Since the emission of these harmful gasses is expected to increase over time with the growth of industry and civilization, renewable energy technologies such as solar, bioenergy, wind, hydropower, etc. [8,9], should be used to reduce the negative effects of greenhouse gas emissions [10].

Bioenergy is a form of renewable energy derived from biomass and leads to biofuels, heat, and electricity production. Biodiesel is widely recognized as a safe, renewable, and non-toxic alternative to conventional fuels [11]. A variety of production methods, feedstocks, and catalyst types (homogeneous acid and alkali catalysts, heterogeneous acid and alkali catalysts, nanocatalysts) can be employed in the production of biodiesel. Biodiesel can be easily produced with edible oils; however, using these oils raises concerns worldwide because they compete with food consumption [12]. Therefore, using waste or non-edible oils in biodiesel production will reduce competition with food consumption while meeting environmental and ethical biofuel standards [13].

For this reason, used cooking oil is used in this study as a low-cost feedstock, which is widely available worldwide. Pyrolysis, supercritical fluid process, and transesterification are well-known processes for biodiesel production [14]. However, transesterification is the most commonly used among all of these techniques, leading toward commercialization [15].

In order to achieve good catalytic activity and environmentally friendly behavior, it is imperative to find green and affordable catalysts [16]. Due to the benefits of free fatty acid tolerance and feedstock water content, heterogeneous catalysts are gaining a lot more interest for biodiesel production applications. Because of the distinct phases, recovering the catalyst from the reaction mixture is simple and can be repeated numerous times. The utilization of heterogeneous catalysts reduces the generation of soap and since the solid catalyst can be reused for several cycles, the biodiesel production process becomes more cost-effective [17]. Multiple scholars investigated the use of calcium oxide catalysts in the transesterification of oils. These catalysts were made from calcium carbonate by calcination under temperatures of 800–1000 °C. Calcium carbonate can be found in biological materials such as marine shells, animal shells and bones, and agricultural shells [18]. It was observed that adding calcium oxide to high surface area materials including alumina, zeolite, and silica could speed up the catalytic activity of CaO and increased the production of biodiesel [19]. For instance, waste eggshells and rice husks were successfully employed in the synthesis of CaO catalyst supported with silica. High catalytic activities were demonstrated by the hybrid catalyst during the transesterification process. In comparison to the biodiesel yield using CaO alone, the transesterification process catalyzed by CaO supported by silica catalyst achieved a higher biodiesel yield and higher efficiency [19]. Yaşar [20] investigated the use of calcium oxide in the process of biodiesel production and the findings of the author's study showed that waste eggshells could be utilized several times as a catalyst, and that it greatly improved the product yield and fuel qualities, while significantly lowering the cost of biodiesel. Another research study concluded that commercial CaO could be utilized as a catalyst for biodiesel synthesis on a wider scale due to its regenerative capacity and reusability without requiring substantial adjustments in conversion. Furthermore, the obtained biodiesel using CaO as a catalyst was of good quality, with a high FAME yield. CaO also has a high catalytic activity for biodiesel generation [21]. Different alkali earth metal oxides (calcium oxide, magnesium oxide, and barium oxide) doped with silicon dioxide were studied as catalysts for the biodiesel synthesis process, at several catalyst loading percentages. The results showed that the purity and yield of biodiesel generated with 60% CaO/SiO₂ were greater than with other catalysts, at 97.3% and 82.1%, respectively [22]. In addition, another study revealed that when combining silicon dioxide with biodiesel, the engine's overall performance improved significantly, and this mixture reduced hazardous emissions from the engine [23].

Response surface methodology (RSM) is a statistical tool that can be used to design, optimize, and analyze experiments in any process. Central composite design (CCD), one of

the types of design, is a commonly used design tool in the literature to optimize various processes [24]. RSM is considered a highly valuable tool in statistics for studying how factorial variables impact the response and the link between input factors and output using models. Using the "one factor, one time" technique to optimize the parameters needs a significant number of experiments, whereas RSM can accomplish this with significantly less runs. Furthermore, the "one factor at a time" technique cannot anticipate the combined effect of two or more factors on the output, which is possible using RSM [25].

Several researchers have worked on optimizing various parameters of the biodiesel production process using RSM. For example, Dharma et al. [26] studied the optimization of three parameters: methanol-to-oil ratio, stirring rate, and concentration of KOH catalyst for biodiesel produced from second-generation oil. The resulting optimized biodiesel produced had physiochemical properties that met ASTM D6751 and EN14214 standards. In another study, Silva et al. [27] investigated and optimized the effects of temperature, catalyst concentration, reaction time, and alcohol mole ratio using a factorial design and the response surface method. Numerous other studies were conducted to investigate the effects of a combination of process parameters and optimized the reaction under different conditions using different starting materials and catalysts.

Additionally, some studies examined the properties of alternative biofuels such as biodiesel and its performance in diesel engines compared to conventional fuels. A study by Viswanthan and Wang [28] evaluated the major physical and chemical features of fish oil ethyl ester produced from transesterification reactions based on biodiesel ASTM standards with diesel as a reference fuel for the testing. The fish oil ester fueled a single cylinder direct-injection engine to examine its influence on engine parameters, and its chemical composition was determined using GC-MS analysis. The influence of fuel preheating was also studied at different preheating temperatures, and it was found that high preheating temperatures improved the engine performance. Finally, engine emission findings were presented. Several other studies also reported the effect of biodiesel and biodiesel blends usage in diesel engines in terms of performance, combustion, and emission parameters (such as carbon monoxide, hydrocarbons, NO_x, and smoke emissions) under varying operating conditions [29,30]. In comparison to electric vehicles, electrically powered vehicles are more advantageous compared to biofuels in terms of lowering fuel consumption and mitigation costs, and boosting fuel supply. However, the biofuel route is superior in terms of emissions reduction, the proportion of alternative fuels, and the economic benefits for consumers [31]. There are several advantages of biodiesel, which makes it an interesting option for transportation fuels. The oxygen content in biodiesel fuel is between 10 and 11%, which results in excellent combustion properties. When compared to regular diesel fuel, biodiesel creates 78% less carbon dioxide during its lifespan, as well as less smoke. Moreover, biodiesel production takes less time than petroleum diesel production since no drilling, transporting, or refining are required [32].

The significance of this study is that it used low-cost waste as feedstock for biodiesel production. In addition, waste glass and animal bones were used as catalysts, which were also economical compared to expensive conventional catalysts. Finally, to improve the economics and productivity of the whole production process, the response surface method (RSM) was used together with central composite design (CCD) to study the effects of catalyst concentration, reaction time, and molar ratio to optimize the transesterification reaction. Although waste oils and green catalysts have been investigated in the literature, there has not been a study concerning biodiesel production using a combination of both waste cooking oil and CaO–SiO₂. The methodology followed to carry out the design of the experiments, experimental runs, and optimization is shown in Figure 1.



Figure 1. Biodiesel production and optimization process methodology.

2. Materials and Methods

2.1. Feedstock

The waste cooking oil was sourced from SS Lootah Biofuels, which specializes in collecting waste cooking oil from numerous fast-food restaurants and chains in Dubai. The properties of the waste cooking oil used for transesterification, such as calorific value, density, and viscosity, were measured as shown in Table 1. The calorific value was measured using a Parr 6400 calorimeter and viscosity was measured using a Brookfield Ametek dv2t viscometer at 40 °C. Comparison with standard values and those in the literature showed that all three properties were in the average range of values for used cooking oils.

Table 1. Physicochemical properties of waste cooking oil.

No.	Properties	Units	Value
1	Heating value	MJ/kg	39.79015
2	Density	kg/m ³	916.73
3	Viscosity @40 °C	cP	39.96

2.2. Catalyst Preparation and Characterization

The catalyst was made from waste glass and animal bones. The waste glass was thoroughly washed, dried, and then ground to a fine powder. The powdered glass was then heated in a muffle furnace at a temperature of 500 °C for 4 h. Then, the powdered glass was washed with warm water to dissolve the water-soluble impurities and subsequently dried in an oven at 120 °C for 12 h. To extract calcium oxide from animal bones, the bones were washed and then dried in an oven at 120 °C for 12 h. The dried bones were ground into powder and heated in an oven at 900 °C for 4 h. Then, the bones were washed thoroughly with warm water (45 °C) and placed in an oven to dry for 12 h at 120 °C. This way, silicon dioxide (SiO₂) was obtained from the waste glass and calcium oxide (CaO) from the animal bones. To prepare the catalyst mixture, 100 g of CaO powder was mixed with 100 g of SiO₂, calcined in a muffle furnace at 500 °C for 4 h, and then cooled. The resulting catalyst was characterized using X-ray diffraction (XRD), scanning electron microscopy (SEM), and Fourier transform infrared spectroscopy (FT-IR).

2.3. Experimental Procedure

The transesterification reaction was carried out with waste cooking oil, methanol, and the catalyst. First, the methanol was preheated to 55 °C, mixing it with a certain amount of catalyst for each run. Once the desired temperature was reached, the used cooking oil was added to the methanol–catalyst mixture and mixed at 300 rpm for the required time of each experiment. After the reaction, the resulting mixture was poured into the separation units and left to separate for 48 h. After 48 h, the biodiesel, catalyst, and glycerol layers were clearly separated. The biodiesel output was weighed, and the yield was measured accordingly to be entered into the RSM software for analysis and optimization. The run

with the highest biodiesel yield was then characterized, and the results were compared and reviewed with the literature.

2.4. Optimization with Response Surface Methodology (RSM)

The analysis and optimization of the biodiesel production process were performed using the Response Surface Methodology (RSM) software (version 13.0) from StatEase. The software is a tool for designing experiments with different experimental designs. For this study, the Central Composite Design Tool (CCD) was used to study the response. Three parameters: Catalyst Concentration (A), Methanol-to-Oil Ratio (B), and Time (C) were varied in a range of 2 to 8 wt% for catalyst concentration, 6:1 to 20:1 for methanolto-oil ratio, and 60 to 240 min for reaction time. The reaction selected was the percent biodiesel yield. RSM software was used to design 20 experiments with the input parameters shown in Table 2. The biodiesel yield obtained from the experiments was calculated by dividing the mass of biodiesel obtained by the mass of oil used in each run as represented in Equation (1) below:

Biodiesel yield (%) =
$$\frac{Mass \ of \ biodiesel \ obtained \ (g)}{Mass \ of \ oil \ used \ in \ each \ run \ (g)}$$
 (1)

Table 2. Design of experimental runs obtained using RSM.

Run	Factor 1: A: Catalyst Concentration (wt%)	Factor 2: B: Methanol-to-Oil Ratio	Factor 3: C: Time (min)	Response: Biodiesel (%)
1	2	6	240	72.17
2	5	13	150	91.36
3	2	13	150	92.14
4	5	13	150	91.84
5	5	20	150	82.25
6	2	20	60	84.21
7	5	13	150	91.15
8	8	6	60	75.62
9	2	20	240	81.28
10	8	20	60	80.89
11	5	13	60	88.54
12	2	6	60	77.67
13	8	6	240	70.68
14	8	20	240	79.36
15	5	13	240	86.67
16	8	13	150	90.25
17	5	13	150	91.54
18	5	6	150	74.37
19	5	13	150	91.29
20	5	13	150	91.79

3. Results and Discussion

3.1. Catalyst Characterization

The CaO–SiO₂ catalyst was characterized using X-ray diffraction (XRD), scanning electron microscopy (SEM), and Fourier transform infrared spectroscopy (FT-IR). The presence of calcium oxide was detected at 20 values of about 32.145° , 37.313° , 53.795° , 63.99° , 67.928° , and 79.50° , while silica was observed at 20.737° , 26.557° , and 50.07° (see Figure 2). Figure 3a,b shows SEM images of the catalysts at 10 µm and 5 µm scales. The images show a flour-like structure and uniform morphology over the entire surface.



Figure 2. X-ray diffraction of CaO-SiO₂ heterogeneous catalyst.



Figure 3. Scanning electron microscopy images for CaO–SiO₂ catalyst: (a) At 10 µm scale; (b) at 5 µm scale.

The FT-IR spectra of the catalyst are shown in Figure 4. CaO was responsible for the broad absorption in the 500–700 cm⁻¹ range, while the wavenumber of the SiO region was about 1200 cm⁻¹.



Wavenumber (cm⁻¹)

Figure 4. Fourier transform infrared spectrum of CaO–SiO₂ heterogeneous catalyst.

3.2. ANOVA Analysis

Among the different models, including linear, quadratic, and cubic models, the model proposed by the software with a p-value of less than 0.0001, which is the quadratic model, was selected for its significance (see Table 3). The equation below presents the model equation and coefficients:

Biodiesel yield = $+45.64164 - 1.48750 \times A + 6.70666 \times B + 0.056792 \times C - 0.010119 \times AB + 0.000907 \times AC + 0.001187 \times BC + 0.112727 \times A2 - 0.242254 B2$ (2) $- 0.000318 \times C2$

where A = catalyst weight percent (wt%), B = methanol-to-oil ratio, and C = time. The results of the analysis of variance (ANOVA) as well as the correlation coefficients presented in the table of fit statistics (Table 4) indicate that the model was significant and that there was a strong and direct relationship between the three parameters chosen for the study and the response identified as biodiesel yield. The ANOVA results (Table 5) also show that the parameters and interactions with the greatest significance are all three factors: A—Catalyst Concentration, B—Methanol-to-Oil Ratio, C—Time, and the interaction, BC, between the methanol-to-oil ratio and time.

Table 3. Fit summary for response.

Source	Sequential p-Value	Lack of Fit p-Value	Adjusted R ²	Predicted R ²	
Linear 2FI	0.3675 0.9936	<0.0001 <0.0001	$0.0197 \\ -0.1989$	-0.3876 -3.9577	
Quadratic Cubic	<0.0001 0.8077	0.0029 0.0004	0.9858 0.9813	0.9631 - 5.7944	Suggested Aliased

Table 4. Fit statistics and regression coefficients.

Std. Dev.	0.8766	R ²	0.9925
Mean	84.25	Adjusted R ²	0.9858
CV. %	1.04	Predicted R ²	0.9631
		Adeq. Precision	35.9693

Table 5. Analysis of variance for quadratic model.

Source	Sum of Squares	Diff.	Mean Square	F-Value	p-Value	
Model	1023.64	9	113.74	148.02	< 0.0001	Significant
A-	11.38	1	11.38	14.82	0.0032	0
B-	140.48	1	140.48	182.82	< 0.0001	
C-	28.12	1	28.12	36.60	0.0001	
AB	0.3613	1	0.3613	0.4701	0.5085	
AC	0.4802	1	0.4802	0.6249	0.4476	
BC	4.47	1	4.47	5.82	0.0366	
A2	2.83	1	2.83	3.68	0.0839	
B2	387.50	1	387.50	504.30	< 0.00001	
C2	18.24	1	18.24	23.74	0.0006	
Residual	7.68	10	0.7684			
Lack of fit	7.30	5	1.46	18.84	0.0029	Significant
Pure error	0.3874	5	0.0775			0
Cor total	1031.32	19				

3.3. Parametric Study

The plot of predicted and actual values of biodiesel yield (Figure 5) shows that the model is a good estimate for predicting the actual values of the yield, since the predicted and actual values are very close. In addition, the analysis of the residuals (Figure 6), such as the plot of normal probability and the plot of residuals versus predicted values, shows no significant deviations from normality or evidence that the model shows signs of dependence or inadequacy. Since there were no discernible patterns in the plot, it could be assumed that the residuals have a constant variance. The different colors in the plots in Figures 5 and 6 represent the values of the yield of biodiesel ranging from 70.68% to 92.14%, where blue is the lowest yield and red is the highest.



Figure 5. Predicted versus actual values.



Figure 6. (a) Normal probability plot; (b) Residuals plot.

Figure 7 shows the 3D contour of the interactions between the different parameters chosen. The first plot (Figure 7a) for the interaction between parameters A (Catalyst Concentration) and B (Methanol-to-Oil Ratio) shows that the value of the methanol-to-oil ratio leading to the highest yield is somewhere in the middle of the selected range of values, while the change in catalyst concentration is not very significant. Plotting the interaction of

parameters B (Methanol-to-Oil Ratio) and C (Time) in Figure 7b showed that the highest biodiesel yields are obtained somewhere in the middle of the ranges considered for both parameters, and that further increasing either parameter does not contribute to increasing the biodiesel yield. Figure 7c represents the interaction between parameters A (Catalyst Concentration) and C (Time), which does not appear to be significant, as shown by both the 3D contour plot and the results of the ANOVA.



Figure 7. 3D interaction between: (a) A and B; (b) B and C; (c) A and C.

Optimization Results

To obtain the optimal parameters and optimize the biodiesel production process, the desired reaction was set to the maximum in the software to obtain the highest yield. The software generated the optimal values for the three parameters as a catalyst concentration of 3.11 wt%, a methanol-to-oil ratio of 14.83:1, and a reaction time of 143 min, which resulted in the optimal yield percentage of 92.3419%, as shown in Figure 8. These values coincided with the highest biodiesel yield obtained experimentally (92.14%), corresponding to the third run using 2 wt% catalyst, a 13:1 methanol-to-oil ratio, and a 150 min reaction time. The red-colored dots represent the optimum values for the input parameters whereas the blue color represents the optimized output value. Table 6 provides a comparison between the optimum biodiesel yield under optimal conditions in previous research and the present study.



Figure 8. Optimization of experimental parameters.

Table 6. Comparison of optimized biodiesely	viel	d	l.
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Feedstock	Catalyst	Optimization Method	Optimized Conditions	Biodiesel Yield	Reference
Waste cotton	KOH• 7:1 methanol-to-oil ratioste cotton vd cooking oil• 0.65 (w/w) % catalyst loadingCaO• 9.6:1 methanol-to-oil ratio• 1.33 (w/w) % catalyst loading• 9.7 min		 7:1 methanol-to-oil ratio 0.65 (w/w) % catalyst loading 9.6 min 	96.44% (Model), 96.55 \pm 0.23% (Experimental)	[00]
seed cooking oil			89.94% (Model), 90.41 \pm 0.02% (Experimental)	[33]	
Waste cooking oil	КОН	RSM	 1.4 wt.% catalyst 7.5:1 methanol-to-oil ratio 65 °C 500 rpm 60 min 	99.38 wt.%	[34]
Castor oil	H ₂ SO ₄) or heterogeneous sulfonated carbon	Taguchi approach	 50 °C 1 h 1% w/w H₂SO₄ 20:1 methanol-to-oil ratio 700 rpm 	90.83%	[35]
Cotton seed oil	КОН	RSM	 6:1 methanol-to-oil molar ratio 55 °C 60 min 0.6% catalyst concentration 	96%	[36]
Flax seed oil	КОН	RSM	• methanol-to-oil molar ratio (5.9:1), catalyst weight (0.51%), temperature (59 °C), reaction time (33 min).	98.6%	[37]
Scum oil	КОН	RSM	 4.5:1 methanol-to-oil molar ratio 75 min 1.20% catalyst concentration 62 °C 	93%	[38]
Fish waste oil	NaOH	RSM	 60 °C 9:1 methanol-to-oil ratio 90 min 	95.39%	[39]

Feedstock	Catalyst	Optimization Method	Optimized Conditions	Biodiesel Yield	Reference
Edible and nonedible vegetable oils	КОН	RSM	 43.50 °C 8.8:1 methanol-to-oil molar ratio catalyst concentration of 1.9 g/100 cc feed 58.4 min 	97.02%	[40]
Waste cooking oil	CaO	One parameter at a time	 50 °C 8:1 methanol-to-oil ratio 1% by weight of catalyst loading rate 90 min 	Around 96%	[41]
Microalge	Chicken- eggshell waste	RSM	 1.7% (w/w) catalyst ratio 3.6 h 140.6 rpm 	86.41%	[42]
Waste cooking oil	CaO–SiO ₂	RSM	 14.83:1 methanol-to-oil molar ratio 3.11 wt% catalyst 143 min 	92.34%	Present work

Table 6. Cont.

3.4. Properties of Biodiesel

The biodiesel heating value, density, viscosity, and flash point were measured for the run with the highest obtained biodiesel yield as per the analysis from the RSM software and experimental results (Table 7). The heating value and viscosity were measured using a Parr 6400 calorimeter and the Brookfield Ametek dv2t viscometer. The flash point was measured using the Normalab NPM 450 device. The higher heating value, density, and flash point values all fell in a reasonable range. Additionally, the viscosity values were compared to ASTM D6751 standard values and proved to comply with the standard.

Table 7. Properties of biodiesel.

No.	Properties	Units	Value
1	Heating Value	MJ/kg	39.71617
2	Density	kg/m ³	920.32
3	Viscosity @40 °C	cP	41.26
4	Flash point	°C	226

4. Conclusions

This study successfully carried out transesterification and optimization processes using a combination of waste cooking oil as feedstock and a catalyst composed of animal bone waste (CaO) and silicon dioxide (SiO₂). Even though research has been conducted on waste cooking oil and green catalysts, previous studies have not yet investigated the combination of both waste cooking oil and CaO–SiO₂ heterogeneous catalyst utilization for biodiesel production. The process was analyzed and optimized using the Response Surface Methodology (RSM) and the Central Composite Design (CCD) tool. The RSM allowed for the analysis of the model's correlation, significance, and adequacy, as well as visual representation of all interactions between the parameters. All three parameters were significant, in addition to the interaction between the methanol-to-oil ratio and time. By optimizing the parameters using RSM, it was found that the highest biodiesel yield was 92.34% using a methanol-to-oil molar ratio of 14.83:1, 3.11 wt% catalyst, and a reaction time of 143 min. Experimentally, these values matched the values obtained that correspond to the yield of 92.14% using a 13:1 methanol-to-oil ratio and 2 wt% catalyst in a 150 min duration. The optimum biodiesel yield under optimal conditions found in this study and the optimized

yield reported in previous research were compared. The run with the highest biodiesel yield was finally characterized, and the results were compared and verified with the literature. The values of the higher heating value, density, viscosity, and flash point for the obtained biodiesel were 39.71617 MJ/kg, 920.32 kg/m^3 , 41.26 cP, and $226 ^\circ\text{C}$, respectively.

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