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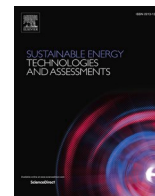
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Biodiesel fuel purification in a continuous centrifugal contactor separator: An environmental-friendly approach

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ABSTRACT

Wet washing is the most common method for biodiesel purification, but higher water consumption, longer purification time, and high expenses bring limitations on the use of this process. One of the suitable methods to remove such limitations is to use new techniques for purification. In this study, biodiesel purification was evaluated in a novel continuous centrifugal contactor separator (CCCS) at 0.5:1, 1:1, and 1.5:1 (V/V%) water to biodiesel ratio; 10 Hz, 20 Hz, and 30 Hz device working frequency; and 25 °C, 40 °C, and 55 °C temperatures. A mathematical model for the wet-washing process of biodiesel and energy consumption of the CCCS device using the response surface method is proposed. A 0.8:1 (V/V%) water to biodiesel ratio, 10 Hz working frequency, and 35 °C temperature were found to be the optimal conditions in the experiment. At this point, the biodiesel yield and the amount of energy consumption were reported to be 96% and 17 kJ, respectively. The results showed that compared to the traditional wet washing method, the biodiesel purification method using CCCS is cost-effective and consumes more than 75% less of water.

Introduction

Considering the decline in fossil fuel resources and increased environmental problems caused by the irregular use of sustainable fuels, particularly in large cities, it has become inevitable to make use of the sustainable energies [1–3]. In recent years, biodiesel has gained growing attention from various researchers due to its sustainability, low pollution combustion, and non-toxicity [4–11]. Biodiesel produced by animal fats and waste oils are highly potential to be mixed with the diesel fuel [1,12–17]. The reason lies in the fact that such raw materials are not only in competition with the food industry, but also result in a global reduction of agriculture wastes through the reuse of wastes [18]. The most common method for biodiesel production is the use of the transesterification process. In this method, two insoluble phases, namely

triglycerides and alcohol, react in the presence of an acid or base catalyst over time, which results in glyceride and alkyl esters production [19–21]. The transesterification process is a reaction limited by the mass transfer, and the reaction rate relies significantly on the alcohol and oil interface as well as on the alkoxide ion density. Also, the quality of biodiesel produced by the transesterification method is affected by various factors, namely the reaction conditions (alcohol to oil molar ratio, temperature, and catalyst), humidity, and free fatty acids [18].

Nowadays, the chemical industries continuously embark on using smaller equipment of high efficiency and increased production rate [22]. Intensification is the process in which the equipment and new processing methods are developed to lessen the production equipment dimensions, increase the energy efficiency and the process safety, and lower the waste production [23]. Accordingly, the biodiesel industry is

Abbreviations: FFA, Free fatty acids; CCCS, Continuous centrifugal contactor separator; RSM, Response surface method; ANOVA, Analysis of variance; Xi, Independent variable; ϵ , Unpredicted error; Y, Response variables; D, Desirability function; N, Number of responses; y_i^{min} , Lower limits; y_i^{max} , Upper limits.

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of no exemption in this regard to reduce the energy consumption of the production process and production costs [24]. In the transesterification reaction, a better contact between the triglycerides and alcohol-catalyst mix can offer an increased reaction rate and result in a more cost-effective reactor [25]. The process intensification technologies can be used to increase the contact between liquid phases, increase the mass transfer rate, and transfer the heat throughout the transesterification process [26–29]. Therefore, new developments have been reported in recent years concerning the use of process intensification technologies in the biodiesel industry. Several intensification methods can be classified as the statistic mixer, micro-channel reactors, oscillatory flow reactors, microwave, hydrodynamic cavitation reactors, rotating/spinning tube reactors, membrane reactors, reactive distillation, and contractor separator [23,30–32]. Among the process intensification technologies, centrifugal contractors perform the reaction process and centrifuge separation simultaneously. Such reactors are composed of stirring and separation parts [33,34].

On the other hand, considering the standard determined by the European Union for alternative biodiesel fuels, the content of free fatty acids, methanol, glycerin, and water has been restricted in biodiesel fuel, where a minimum 96.5% purity has been determined for this type of fuel [35]. However, throughout the transesterification process, impurities such as the alcohol, catalyst, free glycerol, free fatty acids (FFA), water, metals, soap, and non-optimal glycerides are produced as the final product of this reaction [36]. The existence of these impurities has inconvenient effects on a diesel engine, which uses the biodiesel fuel, increases the pollution level, and has impacts on the engine performance [37–41]. Thus, it becomes crucial to consider the purification level in order to produce fuel according to the standards. Glycerol and biodiesel purification is a more complicated task. Although some technologies (enzyme, supercritical, monolithic, resin or acid) can produce biodiesel of high quality even with minimum use of the raw materials, the reaction time becomes lengthier so that the time required for the reaction of acid catalyst and enzyme takes up to 70 h [42]. In addition, the main drawbacks pertain to the use of enzymes, supercritical method, and high technology cost. Currently, the liquid–liquid extraction method, which is already known as the wet washing, is counted as the most common and efficient method for biodiesel purification [43].

The use of common methods requires high levels of biodiesel wet washing process, energy, and water consumption. Besides, water purification followed by this process, aiming to separate the environmentally harmful materials, makes this step of fuel production very expensive. Although dry washing, compared to the wet washing method, is used as compatible with the environment, such a method is not cost-effective for industrial purposes due to the use of absorbents and additional equipment [44–46]. Thus, although a high level of water is required for the total wet washing process, nowadays it is considered as a conventional and applicable method for biodiesel purification when compared to the other methods [47].

Traditionally, researchers have examined new methods for improving the wet washing process and reducing the consumed water. The standard techniques proposed are washing with water, citric acid, sulfur acid, and the use of silica gel [47–49]. Different wet washing methods, namely deionized water and hot water washing [50], acidified water purification [51], and dissolving in organic solvents have been studied to improve the process concerning the separation of impurities from biodiesel [52].

As an example, wet washing method was compared with the hot water, silica gel, and phosphoric acid methods. The results indicated that washing with silica gel and phosphoric acid yields above 92% conversion percentage while the use of the hot water method yields only 89% conversion percentage [45]. In another study, the effect of wet washing through the use of hydrochloric acid, washing with distilled water, and dissolving with a solvent such as hexane and its extraction was investigated. Findings of this study emphasized that biodiesel with above 97% purification is achievable using these three methods [53]. In

a similar experiment, three parameters affecting the wet washing process were examined [53]. In that study, different water and biodiesel ratios were explored at three levels (0.5:1, 1:1, and 1.5:1 (V/V%)), different temperatures (30 °C, 45 °C and 60 °C, and three types of water, namely the tap water, distilled water, and water with 3% phosphoric acid. The results of optimization using the RSM introduced water to biodiesel ratio (1.5), temperature (60 °C), and water with 3% phosphoric acid as the optimal conditions for the reaction [53]. Also, in another research work, Bashir et al. investigated the effect of water pre-wash on both the quality and yield of biodiesel. Outcomes of their study showed that adding 5% water pre-wash results in a 60% reduction of water required for the purification process. Furthermore, their study suggested that with this method, lower levels of wastewater are produced compared to the common wet washing method [43]. The review of literature emphasizes that employed parameters such as the reactor type and wet washing process conditions have much effect on efficiency. Since the separation process in centrifugal reactors is based on density between liquids, these reactors can play a vital role in the wet washing process (liquid phase-biodiesel) [42,54,55].

Recently, the continuous centrifugal contactor separator (CCCS) device has received a lot of attention because of integrating both subsequent separation and intense mixing time of two immiscible liquids. The CCCS technology could enhance the efficiency of the transesterification reaction and reduce the energy input/molar ratio of alcohol to oil. Also, the CCCS is easy to scale up. Therefore, it has a high commercial potential compared with the other biodiesel production methods [54–56]. Moreover, the ability to carry out mixing and separation in a single unit simultaneously leads to a much more energy-efficient process in this type of reactor.

A number of investigations have been performed on enhancing the efficiency of the purification process of biodiesel using the CCCS technology. However, these studies focus on the simultaneous biodiesel and separation process. The main challenge in CCCS devices is their low residence time, resulting in an uncontrollable and uncomplete transesterification reaction [23]. This drawback can be addressed by using the CCCS devices for biodiesel production and separation separately for continuous production of biodiesel. To the best of our knowledge, most of the published papers did not consider CCCS device parameters for the washing process in their study. Besides, to make the biodiesel production process more environmentally friendly and economical, reducing the amount of water used in the wet-washing process is essential. In addition, according to the statements above, due to the uncontrollable reaction in the CCCS, modeling and optimization of such device is essential. Moreover, to the best of the authors' knowledge, investigating the effective parameters of the wet-washing process using the CCCS, i.e., frequency, water to biodiesel ratio, and temperature, and multi-objective optimization technique to achieve a better understanding of the wet-washing process and energy consumption in the CCCS reactor have remained contentious. Accordingly, the novelties of present work are: (1) mathematical modeling of the continuous wet-washing process in the CCCS using the RSM, (2) introducing an optimal point for the CCCS working condition in wet-washing process, (3) modeling of the energy required in the wet-washing process using the RSM technique. Therefore, the focus of the current study is to examine the effect of parameters on the wet washing process, including the frequency, wet washing temperature, and water to biodiesel ratio on the purification of crude biodiesel by investigating the purified biodiesel yield and energy consumption of the wet-washing process parameters. Since the optimization process is complicated and obtaining optimal conditions or the reactor performance is of great importance, in the current study, the RSM was used to attain the optimal conditions for the wet washing of crude biodiesel using the CCCS.

Materials and method

In this study, waste cooking oil, KOH catalyst of high purity (90%),

and methanol (99%) produced by the Merck company were used to produce biodiesel. Transesterification reaction was used to convert triglyceride in the waste cooking oil to biodiesel. The catalyst with 1% wt. concentration and 6:1 oil to alcohol molar ratio were used. The catalyst and methanol preheated waste oil were entered into the CCCS reactor as the light and heavy phases, respectively. Also, to determine the methyl ester content of fatty acids in produced biodiesel and the oil to biodiesel conversion percentage, the BS EN 14103 standard was used. The gas chromatography device used in this study was from Perkin Elmer Company (Clarus 580 model). The device is equipped with a FID detector, biodiesel capillary column of Varian Company (CP 9080 model) of 30 m length, 0.32 mm inner diameter, and 250 μm constant phase thickness. The initial yield of the produced biodiesel was found to be 85%. In this study, three critical factors, namely the CCCS working frequency (10 Hz, 20 Hz, and 30 Hz), device temperature (25 $^{\circ}\text{C}$, 40 $^{\circ}\text{C}$, and 55 $^{\circ}\text{C}$), and water to biodiesel ratio (0.5:1, 1:1, and 1.5:1 (V/V%)) were used to evaluate the reactor performance concerning the biodiesel wet washing. The reactor which was designed and fabricated at Tarbiat Modares University (TMU) was used to run the experiments of the present study. The device shares the same operating mechanism with the CCCS (Fig. 1). Features of the CCCS device used in this study are presented in Table 1.

Since obtaining optimal points of these parameters was of immense significance in this study, the RSM was employed to model and analyze the data. RSM is known to be an efficient method at present, which can model the chemical reactions using a set of mathematical and statistical techniques. This method not only results in the reduction of expensive simulation efforts, but also predicts the normal trend of wet washing process optimization, which is often non-linear and provides a multi-variate experimental model.

In this study, the Box-Behnken method was used to determine the relationship between a dependent (input) and independent (output) variables. In this method, all variables were evaluated individually on three levels. The range of studied variables was selected based on the literature review, which were studied to find the frequency in the CCCS device for optimum mixing. Also, the other researches drew their attention toward the purification of biodiesel through the wet washing method so as to find the best temperature as well as determination of the volume of the water needed to be used for washing the contaminations of crude biodiesel [53,56]. Table 2 illustrates the independent variables range considered in this study.

Eq. (1) represents the statistical model used for y (Yield) input modeling based on x_1, x_1, \dots, x_k input variables [57]

$$y = f(x_1, x_1, \dots, x_k) + \varepsilon \quad (1)$$

Table 1

CCCS reactor properties used in the present research study.

Feature	Value	Unit
Maximum power consumption	60	W
Working frequency	0–100	Hz
Diameter of house	62	mm
Outer diameter centrifuge	50.75	mm
Inner diameter centrifuge	50	mm
Maximum output flow	1.9	L/min

Table 2

Experimental range of the independent parameters of the current study.

Independent variable	X_i	Codes factor levels		
		-1	0	+1
Water to biodiesel ratio (V/V %)	X_1	0.5:1	1:1	1.5:1
Frequency (Hz)	X_2	10	20	30
Temperature ($^{\circ}\text{C}$)	X_3	25	40	55

F is the target function optimized by the software. In this equation, ε shows the variables (error level), which can affect the y parameter value but is considered in the f function. The equation can be presented differently for determining the regression problem. The general representation of a quadratic polynomial function in the current study is shown in Eq. (2) [58]:

$$\text{Yield}(\%) = \beta_0 + \sum_{i=1}^k \beta_i X_i + \sum_{i=1}^k \beta_{ii} X_i^2 + \sum_{i=1}^k \sum_{j=i+1}^k \beta_{ij} X_i X_j + \varepsilon \quad (2)$$

where Y is the response variable i.e., purified biodiesel yield. $\beta_0, \beta_i, \beta_{ii}$, and β_{ij} are the constant coefficient, linear coefficient, square coefficient, and interactive effect of parameters, respectively. Also, X_i and X_j are the independent variables, and ε is the unpredicted error. For multipurpose optimization, all parameters were evaluated using the optimization function presented in Eq. (3):

$$D = ((d_1)^{p_1} (d_2)^{p_2} \dots (d_n)^{p_n})^{\sum p_i} = \left(\prod_{i=1}^n d_i^{p_i} \right)^{\sum p_i} \quad (3)$$

where d_1, d_2, \dots, d_n are the output (response) variables and n is the number of responses in this experiment. Since the goal of this test is to achieve the maximum purity percentage and minimum consumed energy, the following functions were used for response maximizing and minimizing, respectively. In addition, d_i is the desirability function of the defined responses in this study. Furthermore, y_i^{\min} and y_i^{\max} are the lower and

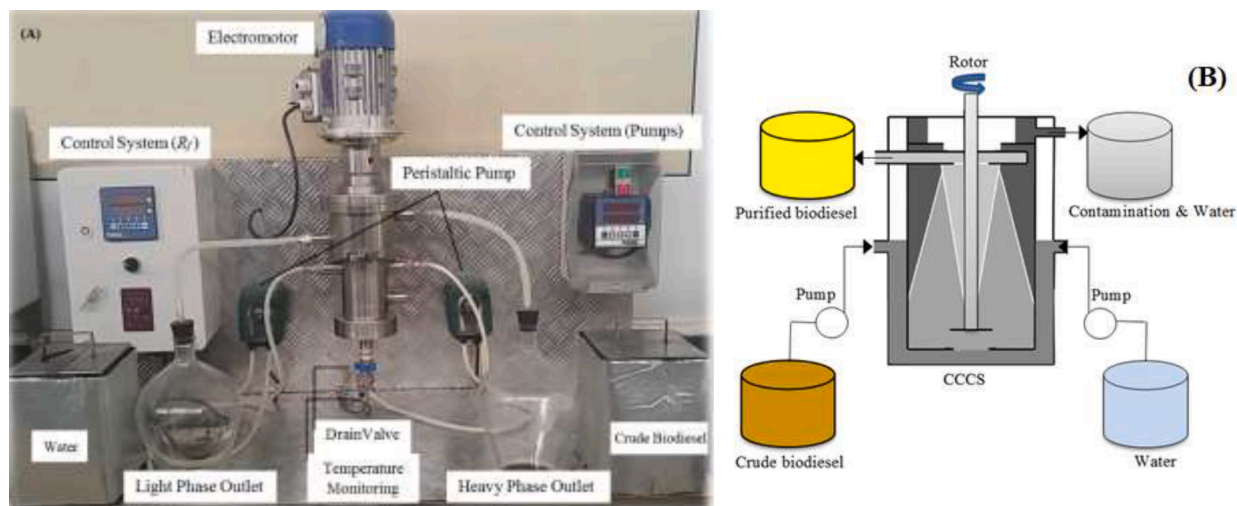


Fig. 1. (A) The illustration and (B) schematic representation of the CCCS device used for purification of crude biodiesel designed and fabricated in-house.

upper limits for the response y_i , respectively. P_1 , P_2 , and P_n are the number of experimental runs [59].

$$d_i = \begin{cases} 0 & \text{if } : y_i \leq y_i^{min} \\ \left(\frac{y_i - y_i^{min}}{y_i^{max} - y_i^{min}}\right)^{w_i} & \text{if } : y_i^{min} \leq y_i \leq y_i^{max} \\ 1 & \text{if } : y_i \geq y_i^{max} \end{cases} \quad (4)$$

$$d_i = \begin{cases} 1 & \text{if } : y_i \leq y_i^{min} \\ \left(\frac{y_i^{max} - y_i}{y_i^{max} - y_i^{min}}\right)^{w_i} & \text{if } : y_i^{min} \leq y_i \leq y_i^{max} \\ 0 & \text{if } : y_i \geq y_i^{max} \end{cases} \quad (5)$$

In general, based on the RSM for running Box-Behnken design using three independent variables (each variable in three levels), 17 experiments were listed by the software, and the response variables, namely, biodiesel purity percentage and energy consumption were calculated for each of the tests [60]. The regression models were fit to determine the relationship between response parameters and independent variables. Besides, the multi-objective optimization was performed by employing the desirability function. In this study, optimization was performed to minimize the water consumption and consumed energy as well as to achieve acceptable yield of purified biodiesel according to the biodiesel standard (e.g. ASTM D-6751).

Result and discussion

The effect of frequency, water ratio, and temperature on the purified biodiesel yield

Using the RSM, the statistical analysis results showed that all parameters examined in this study have a significant effect on the response parameter (purified biodiesel yield) (Table 3). Also, the proper regression equation for data fitting is the quadratic type.

The fit regression equation concerning the experiment data and chosen model using the RSM are presented in Eq. (6). The algebraic mark indicates the increasing or decreasing effect, and the numerical values of coefficients show the importance of parameter considered in the biodiesel conversion percentage level. In this equation, the negative mark of the coefficient means an antagonistic effect, while the positive sign represents a synergistic effect.

$$\text{Yield of purified biodiesel}(\%) = 96.38 + 1.59X_1 - 2.63X_2 + 1.01X_3 - 0.43X_1X_2 + 0.2X_1X_3 - 1.13X_2X_3 - 2.07X_1^2 - 1.29X_2^2 - 0.046X_3^2 \quad (6)$$

According to the statistical analysis results of Table 3, the water to biodiesel ratio has a significant effect on the produced biodiesel purity. According to F-value shown in Table 3, among the different independent variables, the effect of parameters, namely the device working frequency and water to biodiesel ratio, was reported to be two times and seven

Table 3
ANOVA table for purified biodiesel yield.

Source	df	Sum of square	Mean square	F value	P-value
Model	9	117.44	13.05	10.22	0.0029
X₁	1	20.16	20.16	15.79	0.0054
X₂	1	55.13	55.13	43.16	0.0003
X₃	1	8.20	8.20	6.42	0.0390
X₂ × X₁	1	0.72	0.72	0.57	0.4765
X₁ × X₃	1	0.16	0.16	0.13	0.7338
X₂ × X₃	1	5.06	5.06	3.96	0.0868
X₁²	1	17.95	17.95	14.06	0.0072
X₂²	1	7.01	7.01	5.49	0.0517
X₃²	1	0.91	0.91	0.71	0.4264
Residual	7	8.94	1.28		

times of the temperature effect. As presented in Fig. 2, the increase of water ratio in the wet washing process increases the biodiesel purity. The increase in water to biodiesel ratio from 0.5:1 to 1:1 (V/V%) causes a 4% increase in biodiesel purity. An increase in water concerning the water to biodiesel ratio (1:1) causes a decreasing trend. With the increased water content, the catalyst existing in biodiesel showed a decreasing trend due to its increased dissolution in water. Although a higher amount of the consumed water can have significant effects on wet washing costs, the use of lower water amount can yield increased time required for wet washing and increase the consumed energy level for the production process. Thus, obtaining the optimal water to biodiesel ratio is of practical significance, which is one purpose of the current study. The increase in water to biodiesel ratio (greater than 1:1 V/V) causes a decrease in the biodiesel purity percentage. This can be attributed to the increase in the emulsion of soap in water, thus causing the problem in the separation process.

In addition, the increase in device working frequency leads to a reverse effect on the wet washed biodiesel purity content. Based on the results shown in Fig. 2a, the increase in frequency from 10 Hz to 30 Hz leads to a reduction in biodiesel percentage by 5.3%. In other words, the increase in device working frequency and extreme mixing of two phases, as well as lack of enough time or separation of two phases, cause a reduction in wet washing percentage. Putting into simpler terms, the increased frequency yields reduction of the residence time of the reactants between the rotator and stator. Some studies conducted on this type of reactor for biodiesel production have also reported similar results. Kraai et al. (2009) led a study to examine the biodiesel production from sunflower oil related to the CCCS reactor (V-02 model). They suggested that the increase in frequency up to 35 Hz causes an increasing effect on the efficiency of biodiesel production, and followed by 40 Hz this trend decreases [56].

As the results of Fig. 3 represent, the increase in temperature causes an increase in biodiesel purity content. Accordingly, the increase in temperature from 25 °C to 55 °C leads to a 2% increase in the wet washing level. The amount of catalyst remained in biodiesel is significantly dependent on the biodiesel temperature in wet washing status: the increase in temperature leads to increased catalyst concentration due to higher solubility in water. Results obtained in this regard are confirmed by Abbaszadeh et al. [53].

The effect of frequency, water ratio, and temperature on the energy consumption

The fit quadratic model or the consumed energy of the CCCS was obtained using the surface response method based on the independent input parameters as follows:

$$\text{Energy (kJ)} = 23.62 + 0.47X_1 + 3.63X_2 + 10.34X_3 - 0.1X_1X_2 + 0.7X_3X_2 + 0.99X_2^2 + 0.59X_3^2 \quad (7)$$

Table 4 presents the statistical analyses result for the three independent variables on energy consumption. According to the results, water to biodiesel ratio is significant at 5% level, and it has the lowest effect on the consumed energy level among the independent input parameters. Temperature and working frequency parameters are significant at 1% level concerning the consumed energy. Also, the effect of the temperature of water is higher than that of the working frequency of the CCCS. The increase in water temperature leads to a significant increase in the consumed energy level. As can be seen in Fig. 4, the increase in device working frequency from 10 Hz to 30 Hz leads to a 33% increase in the consumed energy, while the increase in water to biodiesel ratio from 0.5:1 to 1.5:1 causes a 4.3% increase in the energy consumption.

Fig. 4a and b illustrate the interactive effect of the system working frequency, water to biodiesel ratio, and temperature on the consumed energy of the wet washing process.

According to the results presented in Table 4 and considering the

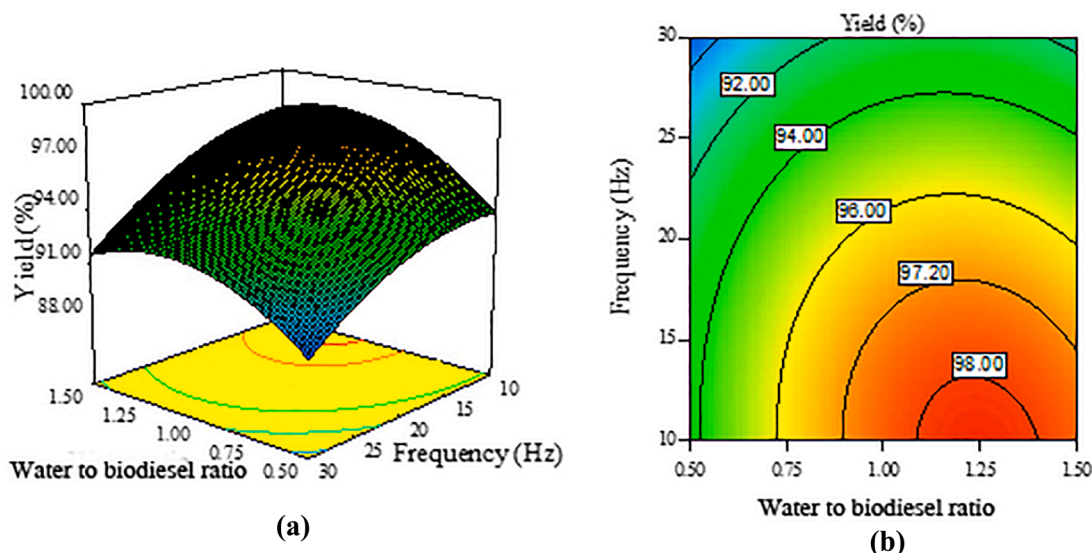


Fig. 2. The interactive effect of the CCCS working frequency and water to biodiesel ratio on the yield of purified biodiesel after wet washing process.

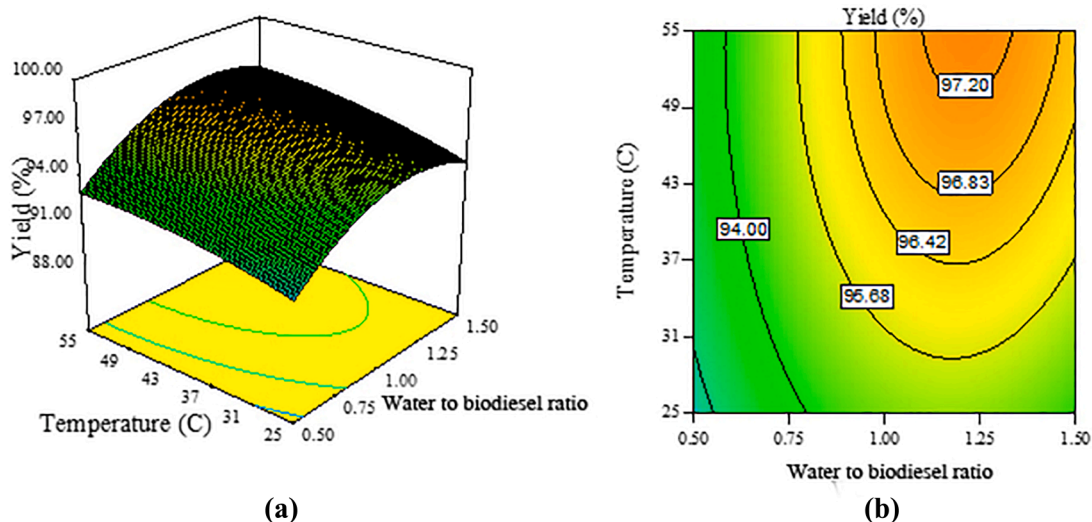


Fig. 3. The interactive effect of water ratio and temperature on the yield of purified biodiesel after wet washing process.

Table 4
ANOVA results for energy consumption.

Source	df	Sum of square	Mean square	F value	P-value
Model	9	970.56	107.84	410.20	< 0.0001
X ₁	1	1.76	1.76	6.69	0.0362
X ₂	1	105.20	105.20	400.14	< 0.0001
X ₃	1	855.74	855.74	3255.00	< 0.0001
X ₂ × X ₁	1	0.042	0.042	0.16	0.7012
X ₁ × X ₃	1	0.040	0.040	0.15	0.7081
X ₂ × X ₃	1	2.02	2.02	7.67	0.0277
X ₁ ²	1	0.037	0.037	0.14	0.7201
X ₂ ²	1	4.14	4.14	15.75	0.0054
X ₃ ²	1	1.36	1.36	5.19	0.0568
Residual	7	1.84	107.84		

effect of temperature as well as working frequency, these two parameters were chosen at their minimum level studied. Thus, considering the energy as the dependent variable, water to biodiesel ratio (0.5:1), temperature (25 °C), and working frequency (10 Hz) have the minimum energy level (9.9 kJ) among the test data. Moreover, at the experimental condition of water to biodiesel ratio (1.2:1), working frequency (10 Hz),

and temperature (25 °C the minimum energy level (10.6 kJ) and maximum biodiesel purity level (95.6%) can be obtained (Fig. 5). As can be seen in Fig. 5, the best yield was obtained for optimal input values.

Optimization

In the optimization process, the criteria for each parameter (i.e., lower limit and upper limit) were defined according to the defined levels in Table 2. The results of the optimization emphasized that the model could predict the trend of experimental data with the maximum R² = 93% value. Fig. 6 represents the estimated values vs. the experimental data.

In addition, results of the optimization illustrated that water to biodiesel ratio (0.8:1), working frequency (10 Hz), and temperature (35 °C) are the best device performance conditions. At this optimized point, the energy level of 17 kJ and purified biodiesel yield of 96% were obtained. The desirability function level was obtained as 0.89 for this optimized point. Fig. 7 shows the desirability function for the suggested optimal point. As the figure pinpoints, a large range was obtained for achieving greater than 96% efficiency, which is based on the ASTM

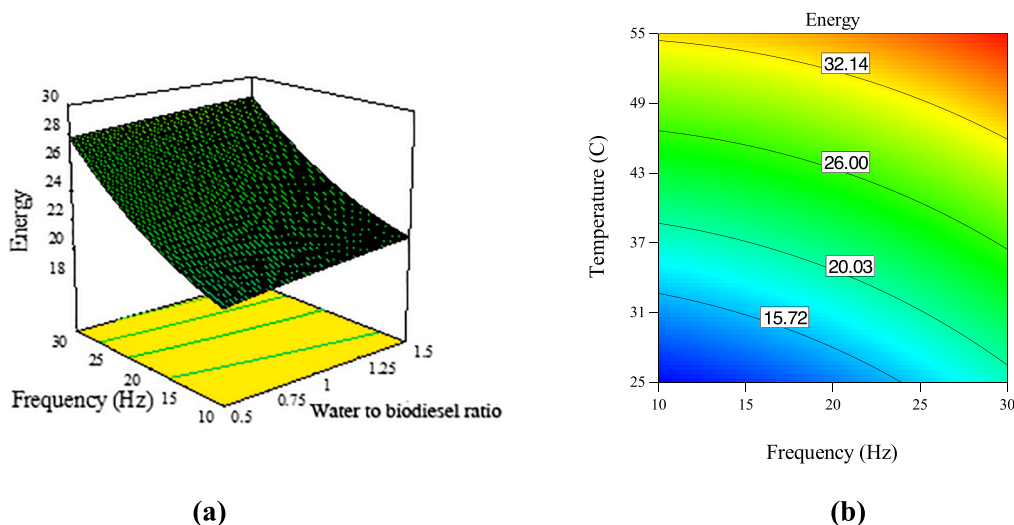


Fig. 4. The interactive effect of frequency, water to biodiesel ratio, and temperature on biodiesel consumed energy level.

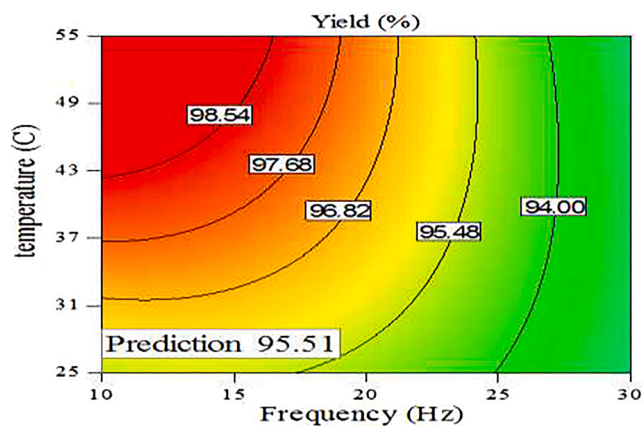


Fig. 5. Energy consumption based on the studied input parameters.

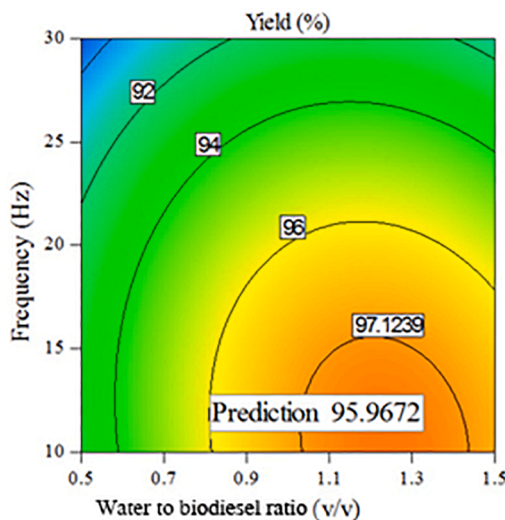


Fig. 7. The desirability function level based on the frequency and water to biodiesel ratio variables.

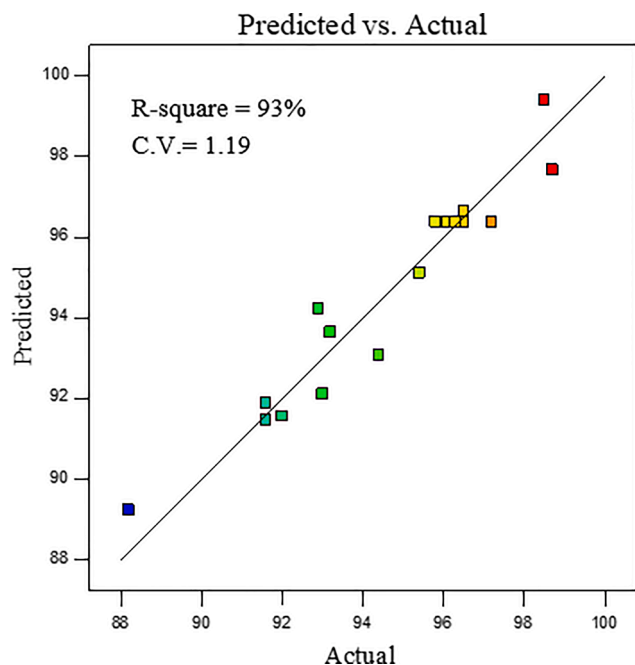


Fig. 6. Predicted vs experimental data.

standard. These findings emphasize that the studied system has an efficient performance in increasing the biodiesel purity [54].

As the results suggest, the temperature of water was obtained lower when compared to the common methods, which affect the consumption level of wet washing [61,62]. Considering that the minimum water to biodiesel ratio required for the biodiesel wet washing using conventional method is 3:1 (V/V %), it is concluded that results obtained in this study can affect the consumed water content for biodiesel wet washing up to 75%. From an economic perspective, the biodiesel producing costs can be reduced, and the production of this fuel can be more cost-effective [63]. Also, the values suggested by the software were experimentally tested in the current study, and the obtained 4% difference can validate the predictability power of the model.

Conclusion

In recent years, one of the most challenging issue regarding the continuous production of biodiesel is its relatively high manufacturing costs. Therefore, eco-friendly and cost-effective biodiesel production technologies need to be developed. The traditional purification methods such as the wet washing are one of the most common methods used

nowadays for biodiesel purification.

In the present study, a mathematical model based on the RSM techniques for the wet washing process of the biodiesel and energy consumption of the CCCS device is proposed. Also, an optimal condition for the CCCS is obtained. Water to biodiesel ratio of 0.8:1 (V/V %), working frequency of 10 Hz, and temperature of 35°C were introduced as the best performance conditions of the CCCS for the wet washing process. At this optimized point, the energy level of 17 kJ and purified biodiesel yield of 96% were obtained. Considering that the consumed water content was lower when compared to the common wet washing methods, this method can be more cost-effective compared to the traditional methods.

CRedit authorship contribution statement

Ebrahim Fayyazi: Data curation, Investigation, Methodology, Writing - review & editing. **Barat Ghobadian:** Conceptualization, Project administration, Writing - review & editing. **Seyed Mohammad Safieddin Ardebili:** Investigation, Software, Writing - original draft. **Gholamhassan Najafi:** Writing - review & editing, Investigation. **Seyed Mohamad Mousavi:** Writing - review & editing, Investigation. **Bahram Hosseinzadeh Samani:** Data curation, Investigation. **Jun Yue:** Writing - review & editing, Investigation.

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.

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References

- [1] Ansari Samani M, Hosseinzadeh Samani B, Lotfalian A, Rostami S, Najafi G, Fayyazi E, et al. The feasibility and optimization of biodiesel production from *Celtis australis* L. oil using chicken bone catalyst and ultrasonic waves. *Biofuels* 2020;11(4):513–21.
- [2] Mehrpooya M, Ghorbani B, Abedi H. Biodiesel production integrated with glycerol steam reforming process, solid oxide fuel cell (SOFC) power plant. *Energy Convers Manag* 2020;206:112467.
- [3] Mehrpooya M, Ghorbani B, Bahnamiri FK, Marefati M. Solar fuel production by developing an integrated biodiesel production process and solar thermal energy system. *Appl Therm Eng* 2020;167:114701.
- [4] Abbaszadeh A, Ghobadian B, Omidkhah MR, Najafi G. Current biodiesel production technologies: A comparative review. *Energy Convers Manag* 2012;63:138–48.
- [5] Haghghi Mood S, Hossein Golpheshan A, Tabatabaei M, Salehi Jouzani G, Najafi GH, Gholami M, et al. Lignocellulosic biomass to bioethanol, a comprehensive review with a focus on pretreatment. *Renew Sustain Energy Rev* 2013;27:77–93.
- [6] Wan Ghazali WNM, Mamat R, Masjuki HH, Najafi G. Effects of biodiesel from different feedstocks on engine performance and emissions: a review. *Renew Sustain Energy Rev* 2015;51:585–602.
- [7] Othman MF, Adam A, Najafi G, Mamat R. Green fuel as alternative fuel for diesel engine: A review. *Renew Sustain Energy Rev* 2017;80:694–709.
- [8] Samuel OD, Gulum M. Mechanical and corrosion properties of brass exposed to waste sunflower oil biodiesel-diesel fuel blends. *Chem Eng Commun* 2019;206(5):682–94.
- [9] Shrivastava P, Salam S, Verma TN, Samuel OD. Experimental and empirical analysis of an IC engine operating with ternary blends of diesel, karanja and roselle biodiesel. *Fuel* 2020;262:116608.
- [10] Samuel OD, Okwu MO, Amosun ST, Verma TN, Afolalu SA. Production of fatty acid ethyl esters from rubber seed oil in hydrodynamic cavitation reactor: study of reaction parameters and some fuel properties. *Ind Crops Prod* 2019;141:111658.
- [11] Giwa SO, SAMUEL OD, Okwu MO, Lagouge SK, Sharifpur M, Jagun ZO. Modeling of *Nicotiana Tabacum* L. Oil Biodiesel Production: Comparison of Artificial Neural Network and Adaptive Neuro-Fuzzy Inference System. *Front Energy Res* 2020;8:377.
- [12] Safieddin Ardebili SM. Green electricity generation potential from biogas produced by anaerobic digestion of farm animal waste and agriculture residues in Iran. *Renew Energy* 2020;154:29–37.
- [13] Moosavi SA, Aghaalikhani M, Ghobadian B, Fayyazi E. Okra: a potential future bioenergy crop in Iran. *Renew Sustain Energy Rev* 2018;93:517–24.
- [14] Yusaf T, Hamawand I, Baker P, Najafi G. The effect of methanol-diesel blended ratio on CI engine performance. *Int J Automotive Mech Eng* 2013;8:1385–95.
- [15] Najafi G, Ghobadian B, Yusaf TF, Rahimi H. Combustion analysis of a CI engine performance using waste cooking biodiesel fuel with an artificial neural network aid. *American J Appl Sci* 2007;4(10):759–67.
- [16] Etefaghi E, Ghobadian B, Rashidi A, Najafi G, Khoshtaghaza MH, Rashtchi M, et al. A novel bio-nano emulsion fuel based on biodegradable nanoparticles to improve diesel engines performance and reduce exhaust emissions. *Renewable Energy* 2018;125:64–72.
- [17] Soudagar MEM, Afzal A, Safaei MR, Manokar AM, EL-Seesy AI, Mujtaba MA, et al. Investigation on the effect of cottonseed oil blended with different percentages of octanol and suspended MWCNT nanoparticles on diesel engine characteristics. *J Therm Anal Calorim* 2020:1–18.
- [18] Saraf S, Thomas B. Influence of feedstock and process chemistry on biodiesel quality. *Process Saf Environ Prot* 2007;85(5):360–4.
- [19] Safieddin Ardebili SM, Ge X, Cravotto G. Flow-mode biodiesel production from palm oil using a pressurized microwave reactor 2019.
- [20] Farvardin M, Hosseinzadeh Samani B, Rostami S, Abbaszadeh-Mayvan A, Najafi G, Fayyazi E. Enhancement of biodiesel production from waste cooking oil: ultrasonic-hydrodynamic combined cavitation system. *Energy Sources, Part A Recover Util Environ Eff* 2019:1–15.
- [21] Hosseinzadeh Samani B, Ansari Samani M, Shirmeshan A, Fayyazi E, Najafi G, Rostami S. Evaluation of an enhanced ultrasonic-assisted biodiesel synthesized using safflower oil in a diesel power generator. *Biofuels* 2020;11(4):523–32.
- [22] Budzaki S, Miljic G, Tisma M, Sundaram S, Hessel V. Is there a future for enzymatic biodiesel industrial production in microreactors? *Appl Energy* 2017;201:124–34.
- [23] Chuah LF, Klemes JJ, Yusup S, Bokhari A, Akbar MM, Klemes JJ, et al. A review of cleaner intensification technologies in biodiesel production. *J Clean Prod* 2017;146:181–93. DOI:10.1016/j.jclepro.2016.05.017.
- [24] Budzaki S, Miljic G, Sundaram S, Tisma M, Hessel V. Cost analysis of enzymatic biodiesel production in small-scaled packed-bed reactors. *Appl Energy* 2018;210:268–78.
- [25] Samani BH, Zareiforoush H, Lorigooini Z, Ghobadian B, Rostami S, Fayyazi E. Ultrasonic-assisted production of biodiesel from *Pistacia atlantica* Desf. oil. *Fuel* 2016;168:22–6.
- [26] Ardebili SMS, Hashjin TT, Ghobadian B, Najafi G, Mantegna S, Cravotto G. Optimization of biodiesel synthesis under simultaneous ultrasound-microwave irradiation using response surface methodology (RSM). *Green Process Synth* 2015;4:259–67.
- [27] Fayyazi E, Ghobadian B, Najafi G, Hosseinzadeh B, Mamat R, Hosseinzadeh J. An ultrasound-assisted system for the optimization of biodiesel production from chicken fat oil using a genetic algorithm and response surface methodology. *Ultrason Sonochem* 2015;26:312–20.
- [28] Mostafaei M, Ghobadian B, Barzegar M, Banakar A. Optimization of ultrasonic assisted continuous production of biodiesel using response surface methodology. *Ultrason Sonochem* 2015;27:54–61.
- [29] Samuel OD, Okwu MO. Comparison of Response Surface Methodology (RSM) and Artificial Neural Network (ANN) in modelling of waste coconut oil ethyl esters production. *Energy Sources, Part A Recover Util Environ Eff* 2019;41(9):1049–61.
- [30] Oh PP, Lau HLN, Chen J, Chong MF, Choo YM. A review on conventional technologies and emerging process intensification (PI) methods for biodiesel production. *Renew Sustain Energy Rev* 2012;16(7):5131–45.
- [31] Tasdemir A, Cengiz Ibrahim, Yildiz E, Bayhan YalcinK. Investigation of ammonia stripping with a hydrodynamic cavitation reactor. *Ultrason Sonochem* 2020;60:104741.
- [32] Ong HC, Milano J, Silitonga AS, Hassan MH, Shamsuddin AH, Wang C-T, et al. Biodiesel production from *Calophyllum inophyllum*-*Ceiba pentandra* oil mixture: optimization and characterization. *J Clean Prod* 2019;219:183–98.
- [33] Fayyazi E, Ghobadian B, Moosavi SM, Najafi G. Intensification of continuous biodiesel production process using a simultaneous mixer-separator reactor. *Energy Sources, Part A Recover Util Environ Eff* 2018;40(9):1125–36.
- [34] Fayyazi E, Ghobadian B, Najafi G, Hosseinzadeh B. Genetic algorithm approach to optimize biodiesel production by ultrasonic system. *Chem Prod Process Model* 2014;9:59–70.
- [35] Silitonga AS, Masjuki HH, Mahlia TMI, Ong HC, Chong WT, Boosroh MH. Overview properties of biodiesel diesel blends from edible and non-edible feedstock. *Renew Sustain Energy Rev* 2013;22:346–60.
- [36] de Farias BS, Vidal EvelinM, Ribeiro NatáliaT, da Silveira N, da Silva Vaz B, Kuntzler SG, et al. Electrospun chitosan/poly (ethylene oxide) nanofibers applied for the removal of glycerol impurities from biodiesel production by biosorption. *J Mol Liq* 2018;268:365–70.
- [37] Chen X, Wang Z, Pan S, Pan H. Improvement of engine performance and emissions by biomass oil filter in diesel engine. *Fuel* 2019;235:603–9.
- [38] Naja G, Ghobadian B, Yusaf T, Mohammad S, Mamat R. Optimization of performance and exhaust emission parameters of a SI (spark ignition) engine with gasoline e ethanol blended fuels using response surface methodology 2015;i. DOI: 10.1016/j.energy.2015.07.004.

- [39] Ong HC, Masjuki HH, Mahlia TMI, Silitonga AS, Chong WT, Yusaf T. Engine performance and emissions using *Jatropha curcas*, *Ceiba pentandra* and *Calophyllum inophyllum* biodiesels in a CI diesel engine. *Energy* 2014;69:427–45.
- [40] Samuel OD, Okwu MO, Oyejide OJ, Taghinezhad E, Afzal A, Kaveh M. Optimizing biodiesel production from abundant waste oils through empirical method and grey wolf optimizer. *Fuel* 2020;281:118701.
- [41] David Samuel O, Adekojo Waheed M, Taheri-Garavand A, Verma TN, Dairo OU, Bolaji BO, et al. Prandtl number of optimum biodiesel from food industrial waste oil and diesel fuel blend for diesel engine. *Fuel* 2021;285:119049.
- [42] Sander A, Antonije Koščak M, Kosir D, Milosavljević N, Parlov Vuković J, Magić L. The influence of animal fat type and purification conditions on biodiesel quality. *Renew Energy* 2018;118:752–60.
- [43] Bashir MA, Thiri M, Yang X, Yang Y, Safdar AM. Purification of biodiesel via pre-washing of transesterified waste oil to produce less contaminated wastewater. *J Clean Prod* 2018;180:466–71.
- [44] Atadashi IM, Aroua MK, Aziz ARA, Sulaiman NMN. Refining technologies for the purification of crude biodiesel. *Appl Energy* 2011;88(12):4239–51.
- [45] Manuale DL, Greco E, Clementz A, Torres GC, Vera CR, Yori JC. Biodiesel purification in one single stage using silica as adsorbent. *Chem Eng J* 2014;256:372–9.
- [46] Stojković LJ, Stamenković OS, Povrenović DS, Veljković VB. Purification technologies for crude biodiesel obtained by alkali-catalyzed transesterification. *Renew Sustain Energy Rev* 2014;32:1–15.
- [47] Okumuş ZÇelik, Doğan TH, Temur H. Removal of water by using cationic resin during biodiesel purification. *Renew Energy* 2019;143:47–51.
- [48] Bateni H, Saraeian A, Able C, Karimi K. Biodiesel purification and upgrading technologies. *Biodiesel*, Springer 2019:57–100.
- [49] Hemmat Y, Ghobadian B, Loghavi M, Kamgar S, Fayyazi E. Biodiesel fuel production from residual animal fat as an inedible and inexpensive feedstock. *Int Res J Appl Basic Sci* 2013;5:84–91.
- [50] Squizzato AL, Fernandes DM, Sousa RMF, Cunha RR, Serqueira DS, Richter EM, et al. Eucalyptus pulp as an adsorbent for biodiesel purification. *Cellulose* 2015;22:1263–74.
- [51] Huerga IR, Zanuttini MaríaS, Gross MartínS, Querini CA. Biodiesel production from *Jatropha curcas*: integrated process optimization. *Energy Convers Manag* 2014;80:1–9.
- [52] Gomes MCSérgi, Arroyo PA, Pereira NC. Influence of oil quality on biodiesel purification by ultrafiltration. *J Memb Sci* 2015;496:242–9.
- [53] Abbaszadeh A, Ghobadian B, Najafi G, Yusaf T. An experimental investigation of the effective parameters on wet washing of biodiesel purification. *Int J Automot Mech Eng* 2014;9:1525–37.
- [54] Fayyazi E, Ghobadian B, van de Bovenkamp HH, Najafi G, Hosseinzadehsamani B, Heeres HJ, et al. Optimization of biodiesel production over chicken eggshell-derived CaO catalyst in a continuous centrifugal contactor separator. *Ind Eng Chem Res* 2018;57(38):12742–55.
- [55] Ilmi M, Kloekhorst A, Winkelman JGM, Euverink GJW, Hidayat C, Heeres HJ. Process intensification of catalytic liquid-liquid solid processes: continuous biodiesel production using an immobilized lipase in a centrifugal contactor separator. *Chem Eng J* 2017;321:76–85.
- [56] Kraai GN, Schuur B, van Zwol F, van de Bovenkamp HH, Heeres HJ. Novel highly integrated biodiesel production technology in a centrifugal contactor separator device. *Chem Eng J* 2009;154(1-3):384–9. <https://doi.org/10.1016/j.cej.2009.04.047>.
- [57] Safieddin Ardebili SM, Solmaz H, Mostafaei M. Optimization of fusel oil – Gasoline blend ratio to enhance the performance and reduce emissions 2019;148:1334–45. DOI:10.1016/j.applthermaleng.2018.12.005.
- [58] Solmaz H, Mohammad S, Aksoy F, Calam A, Emre Y, Arslan M. Optimization of the operating conditions of a beta-type rhombic drive stirling engine by using response surface method 2020;198. DOI:10.1016/j.energy.2020.117377.
- [59] Khoobakht G, Najafi G, Karimi M, Akram A. Optimization of operating factors and blended levels of diesel, biodiesel and ethanol fuels to minimize exhaust emissions of diesel engine using response surface methodology. *Appl Therm Eng* 2016;99:1006–17. <https://doi.org/10.1016/j.applthermaleng.2015.12.143>.
- [60] Khoobakht G, Karimi M, Kheiralipour K. Effects of biodiesel-ethanol-diesel blends on the performance indicators of a diesel engine: a study by response surface modeling. *Appl Therm Eng* 2018. [j.applthermaleng.2018.08.025](https://doi.org/10.1016/j.applthermaleng.2018.08.025).
- [61] Iglesias J, Melero JA, Bautista LF, Morales G, Sánchez-Vázquez R. Continuous production of biodiesel from low grade feedstock in presence of Zr-SBA-15: Catalyst performance and resistance against deactivation. *Catal Today* 2014;234:174–81. <https://doi.org/10.1016/j.cattod.2014.01.004>.
- [62] Jitputti J, Kitiyanan B, Rangsunvigit P, Bunyakiat K, Attanatho L, Jenvanitpanjakul P. Transesterification of crude palm kernel oil and crude coconut oil by different solid catalysts. *Chem Eng J* 2006;116(1):61–6. <https://doi.org/10.1016/j.cej.2005.09.025>.
- [63] Fayyazi E, Ghobadian B, Mousavi SM, Najafi G, Yue J, Hosseinzadeh B. Optimization of operational and design parameters of a Simultaneous Mixer-Separator for enhanced continuous biodiesel production. *Chem Prod Process Modeling* 2020. <https://doi.org/10.1515/cppm-2020-0001> (ahead-of-print).