University of Groningen

Optimization of operational and design parameters of a Simultaneous Mixer-Separator for enhanced continuous biodiesel production

Fayazishishvan, Ebi; Ghobadian, Barat; Mousavi, S. Mohamad; Najafi, Gholamhassan; Yue, Jun; Hosseinzadeh Samani, Bahram

Published in:
Chemical product and process modeling

DOI:
[10.1515/cppm-2020-0001](https://doi.org/10.1515/cppm-2020-0001)

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version
Publisher's PDF, also known as Version of record

Publication date:
2021

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Fayazishishvan, E., Ghobadian, B., Mousavi, S. M., Najafi, G., Yue, J., & Hosseinzadeh Samani, B. (2021). Optimization of operational and design parameters of a Simultaneous Mixer-Separator for enhanced continuous biodiesel production. *Chemical product and process modeling*, 16(3), 155–167. <https://doi.org/10.1515/cppm-2020-0001>

Copyright

Other than for strictly personal use, it is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license (like Creative Commons).

The publication may also be distributed here under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license. More information can be found on the University of Groningen website: <https://www.rug.nl/library/open-access/self-archiving-pure/taverne-amendment>.

Take-down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Downloaded from the University of Groningen/UMCG research database (Pure): <http://www.rug.nl/research/portal>. For technical reasons the number of authors shown on this cover page is limited to 10 maximum.

Ebrahim Fayyazi, Barat Ghobadian*, S. Mohamad Mousavi, Gholamhassan Najafi, Jun Yue and Bahram Hosseinzadeh

Optimization of operational and design parameters of a Simultaneous Mixer-Separator for enhanced continuous biodiesel production

<https://doi.org/10.1515/cppm-2020-0001>

Received January 10, 2020; accepted April 28, 2020; published online August 19, 2020

Abstract: Nowadays, biodiesel is promoted as an alternative and renewable fuel. The mass-transfer limited transesterification reaction is commonly used for biodiesel production, but it could benefit from process intensification technologies. The Simultaneous Mixer-Separator (SMS) is a novel process intensification reactor capable of integrating the mixing and separation of reactants within a single unit. The current study aims to determine the ideal parameters for continuous biodiesel production using an SMS setup that was exclusively designed and fabricated in-home for enhanced biodiesel production. The research statistically analyzed the effect of the space between the rotor and the bottom of reactor (h) (0.7, 1.0, 1.3 cm), the diameter ratio between the rotor and the stator (D_r/D_s) (0.5, 0.7, 0.9), and the frequency of the rotor's rotary speed (R_f) (20, 40, 60 Hz) on biodiesel yield using the Response Surface Methodology (RSM). Optimal oil to fatty acid methyl ester (FAME) conversion of 93.2% and the optimal volumetric production rate of 1,980 (kg FAME/m³·h) were obtained by setting the SMS to a rotational frequency of 39 Hz, an h of 0.7 cm, and a D_r/D_s of 0.85.

Keywords: biodiesel; process intensification; response surface methodology; thin layer mixing; transesterification.

1 Introduction

In the past decade, biodiesel has attracted the attention of many researchers as an alternative fuel due to its features of renewability and its capability to be used by currently existing engines. The latter feature, particularly, distinguishes biodiesel from other alternative fuels [1]. According to reports biofuel production was 153 million liters in 2018, which is eight times the amount produced in 2005 [2]. The use of waste cooking oil (WCO) as a resource for the creation of biodiesel fuel is an attractive alternative to other virgin vegetable oils, as the use of WCOs can drastically decrease the capital costs of biodiesel production, thereby enabling biodiesel to become a cost-competitive fuel. There are a variety of ways to derive biodiesel production, and transesterification is among the most common [3]. Transesterification converts vegetable oil to biodiesel in a transformation that occurs in the presence of a catalyst and heat.

However, the process presents a number of challenges, including the immiscibility of oil and methanol (which limits the mass transfer capability) and the sensitivity to conversion as a result of reaction reversibility.

***Corresponding author: Barat Ghobadian**, Department of Mechanics of Biosystems Engineering, Tarbiat Modares University, Tehran, Islamic Republic of Iran, E-mail: ghobadib@modares.ac.ir

Ebrahim Fayyazi and Gholamhassan Najafi: Department of Mechanics of Biosystems Engineering, Tarbiat Modares University, Tehran, Islamic Republic of Iran

S. Mohamad Mousavi: Department of Chemical Engineering, Tarbiat Modares University, Tehran, Islamic Republic of Iran

Jun Yue: Department of Chemical Engineering, Engineering and Technology Institute Groningen, University of Groningen, 9747 AG, Groningen, The Netherlands

Bahram Hosseinzadeh: Department of Biosystems Engineering, Shahrekord University, Shahrekord, Islamic Republic of Iran

Current conventional techniques to overcome these problems include implementing long reaction times, increasing the molar ratio of alcohol to oil, and carrying out several reaction stages. High operating costs and energy consumption are two main issues in the process of purifying crude biodiesel and recovering excess alcohol and catalysts during downstream processing. Furthermore, the downstream purification process can yield significant amounts of toxic waste water. Low production efficiency is primarily affected by long reaction times and the cost of down-stream processing.

New process intensification technologies have emerged in recent years to improve the mixing and mass/heat transfer procedure between the two liquid phases in the production of biodiesel. These newly emerged technologies make use of novel reactors which have greatly increased the reaction rate, thereby reducing the reaction time. Researchers have identified a number of such technologies that are advantageous in biodiesel production [4]. For example, Thompson and He assessed the use of the stand-alone, closed-loop static mixer system as a continuous-flow reactor with an aim to produce biodiesel from canola oil with methanol, using sodium hydroxide as the catalyst [5]. In a similar vein, Sun et al. [6] put forth a study to examine the KOH-catalyzed transesterification of unrefined rapeseed oil and cottonseed oil with methanol in capillary micro-reactors with inner diameters of 0.25 mm. Harvey et al. aimed to produce saleable biodiesel from rapeseed oil in a pilot-scale plant through their assessment of a continuous oscillatory flow reactor (OFR) [7]. To obtain higher grade ASTM 6751 B100 biodiesel, Arisdyne System, Inc attempted to use flow cavitation (CFC) technology [8]. Gulyurt et al., on the other hand, focused mainly on the transesterification of *Chlorella protothecoides* oil with methanol under microwave irradiation [9]. Gude et al. also assessed the potential of microwave energy for the production of biodiesel [10], as have some other researchers [11, 12]. Fayyazi et al. designed an ultrasonic-assisted setup to examine the intensification of the transesterification reaction. They concluded that the reaction time could be decreased by approximately 87.5%, so long as the biodiesel was obtained from the incorporation of an ultrasonic wave under a conversion percentage condition that was similar to that of the conventional method [4, 14].

Some researchers used other reactor types to perform reaction and separation in the same device which have some advantages like saving energy consumption. Dubè et al. strived to produce biodiesel from canola oil and methanol using a two-phase tubular membrane reactor [15]. In their study on biodiesel production, Cheng et al. tried modeling and simulation using a membrane reactor integrated with a pre-reactor [16]. Many researchers have recently taken notice of the reactive distillation method, which is widely used in both research and commercial applications because it carries fruitful advantage when compared to conventional sequential processes [17–20].

Another process intensification technology that integrates reaction and centrifugal separation into a single unit is the Continuous Centrifugal Contactor Separator (CCCS). The CCCS is composed of a mixing zone and a separating zone, as shown in Figure 1 [21, 22]. The rotation of the rotor within a stationary cylinder (also known as a stator) results in intense mixing and improved mass transfer, as well as quick phase separation via high shear stress and high centrifugal force [4, 23]. It also serves as a mixer-settler for biphasic liquid-liquid systems [1, 6].

There are advantages to using the CCCS over conventional stirred vessels. For one, the crude fatty acid methyl esters (FAME)/FAEE is in-situ separated from the glycerol layer by the action of centrifugal forces, rather than being separated during an excessive post-processing operation. Another advantage is that the volumetric production rates achieved by the CCCS exceed those achieved in stirred tanks. This is likely due to the presence of very fine droplets in the dispersed phase, which gives rise to high volumetric mass transfer coefficients ($k_L a$) [23].

Moreover, the ability to carry out the reaction and separation in a single unit simultaneously leads to much more energy efficient process in this type of reactor. Several researchers have assessed the use and modeling of the CCCS reactor. Some have highlighted studies on enantioselective liquid-liquid extraction, enantio-separation [26–33], liquid hold-up, residence time distribution, phase behavior, and drop size distributions in the CCCS [24, 34] as the major research areas in biodiesel production [1, 23, 24, 35–37]. Kraai et al. used the CCCS reactor for the transesterification of sunflower oil and methanol [35], while Ilmi et al. used the CCCS for liquid-liquid-solid reactions, which were employed to maintain a continuous biodiesel production using an immobilized enzyme. Their study found that it was feasible to improve the performance of the reaction using the Continuous Stirred Tank Reactor (CSTR) and the CCCS. The researchers concluded that the average biodiesel

Figure 1: Schematic representation of the CCCS device [35].

yield at steady state conditions was 86% (without any further purification) for a 9 h run [1]. Abduh et al. evaluated the production of FAME from sunflower oil and methanol in a CCCS using sodium methoxide as the catalyst. The findings clearly showed that (comma hazf mishe) at optimized conditions (oil flow rate of 31 mL/min, rotational speed of 34 Hz, catalyst concentration of 1.2%w/w, and a methanol flow rate of 10 mL/min), the FAME yield was 94 mol% at a productivity of 2470 kg FAME/m³reactor·h [23]. Abduh et al. also used a CCCS setup in order to produce *Jatropha curcas* L. oil ethyl ester [36]. As the most recent research study in this field, Fayyazi et al. focused on using solid calcium oxide (CaO) as a heterogeneous catalyst derived from chicken eggshell for biodiesel production from sunflower oil (containing 5 wt% tetrahydrofuran as co-solvent) in a CCCS setup. The optimal values of reaction parameters determined by the Response Surface Methodology (RSM) and verified experimentally were reported as oil flow rate of 9 mL/min, an alcohol to oil molar ratio of 11:1, and a weight hourly space time (defined as the catalyst weight over the oil mass flow rate) of 0.050 h. In this research, an optimized FAME yield of 83.2% with a productivity of 638 kgFAME/(m³reactor·h) was achieved [37]. Moreover, Schuur et al. investigated the hydrodynamic features of CCCS reactors with a particular focus on experimental studies concerning liquid hold-up, residence time distribution, phase behavior, and drop size distributions [24]. Though an ensemble of research studies perform CFD modeling and study of the hydrodynamic features of CCCS reactors [24, 25], scant attention has been directed toward experimental investigation of the operation and geometrical parameters of biodiesel production in the CCCS reactor.

In fact, CCCS performance is significantly affected by the geometrical parameters that characterize the dynamics of flow between two cylinders. The flow between two concentric rotating cylinders, which is known as the “Taylor–Couette Flow,” is one of the classic issues in hydrodynamic stability. Taylor–Couette Flow can be generated under the influence of the rotation of one cylinder while the other is fixed or by the simultaneous rotation of two cylinders in the same or opposite directions. There is a critical angular velocity for the inner cylinder if it rotates with a constant speed. In angular speeds smaller than the critical angular velocity, a kind of shear flow exists. This is known as the Couette flow between two cylinders. However, if the angular velocity is increased to the point that it surpasses this critical value, the flow of the fluid becomes unstable and 3D vortices appear. When the flow becomes unstable, the horizontally rotating fluid begins to rotate in a spiral (Figure 2).

The instability of the rotating fluid is caused by mechanical tensions in the inter-cylinder cavity and the instability of centrifugal forces. On the other hand, the fluid viscosity has an inverse effect and can eliminate such instability [38]. Eq. (1) shows the relationship between the Taylor number (Ta) and the involved parameters, where d is the space between two cylinders ($D_s - D_r$) (Figure 3), ν is the kinematic viscosity of the fluid, and ω_1 and r_1 are the angular velocity and the radius of the inner cylinder, respectively.

Figure 2: (a) The horizontal rotating flow at the velocity less than critical angular velocity and (b) the spiral rotating flow once the critical angular velocity has been exceeded.

$$Ta = \frac{r_1 \omega_1 d}{\nu} \quad (1)$$

According to the Taylor–Couette equation Eq. (1), angular speed and inter-cylinder space have significant effects on the flow regime between the two cylinders (rotor and stator) which could make intensive mixing. In addition, the literature indicates that such cylindrical reactors generate effective residence time for the reactants between the rotor and stator [1, 35]. Since researchers have yet to perform an experimental

Figure 3: Independent variables investigated in this research study, reproduced from [35].

investigation of the effects of these factors specially for biodiesel production, this study assesses the following three independent factors: the effect of the space between the rotor and the bottom of the reactor (h), the diameter ratio between the rotor and the stator (D_r/D_s), and the frequency of the rotor's rotary speed (R_f) (Figure 3) in a home-made Simultaneous Mixer-Separator (SMS) reactor which works based on the CCCS reactors. The study applied the RSM through Box–Behnken design as a strong and efficient experimental design tool for a statistical analysis of the above-mentioned operational and design parameters (h , D_r/D_s , R_f) and finding the optimum levels of each independent factor.

The Section 1 of research focused on the statistical analysis of the effects of the independent experimental factors and their interactions on the volumetric production rate and the reaction yield (conversion of oil to methyl ester). The Section 2 aimed to obtain the maximum efficacy of the SMS reactor by discovering the optimal parameters of h , D_r/D_s , R_f in this reactor.

2 Material and methods

2.1 Material and equipment

The WCO collected from restaurants was used in the current study for all experiments. The WCO was filtered and decanted in a container in order to exclude any remaining particles. Methanol (99%) and potassium hydroxide (99.9%) were obtained from Merck, Germany, and Methyl nonadecanoate (>98%) and *n*-heptane for gas chromatography (GC) were obtained from Sigma–Aldrich.

The SMS reactor developed specifically for this study was designed and manufactured at Tarbiat Modares University (TMU), and was based on the operation mechanism used in centrifugal contact reactors (Figure 4). The SMS setup is composed of outer static housing and inner centrifuge. One segment (the space between centrifuge and static housing) performs the reaction and the other (inner centrifuge) performs the process of separation. This system includes a remote rotor frequency controller (0–60 Hz), an inside-reactor temperature monitoring sensor, and an apparatus which enables controlling the raw material flow rate. To set the flow rate of the raw materials (oil and methoxide), the system includes two P566 peristaltic pumps with a low, single channel that rotates 30–120 ml/min and a timer which controls switching the pumps on and off.

2.2 Methods

2.2.1 Reactant preparation and related properties: Water loss capability and the presence of additional materials (such as food particles) were the two important elements to be considered in the oil. The presence of water and additional materials in the waste oil resulted in failure to improve the efficiency of the procedure, even when the amount present was low. In the current research, the WCO was filtered and decanted for 24 h in a container, with the aim of removing any particles remaining in the oil. Following this, the purified oil was methylated using the Metcalfe method [39]. The prepared sample was then injected into the GC instrument in order to determine the used oil's fatty acids profile and its molecular weight. According to Eq. (2), the titration method [14] was used to determine the FFAs. In order to determine the water content of the oil, the distillation method was used (ASTM, D4006-7).

Figure 4: The SMS system used for biodiesel production.

Table 1: Coded and actual values of the variables that were used for the experimental design.

Independent variable	Unit	Coded variables levels		
		−1	0	+1
Rotational frequency(R_f)	Hz	20	40	60
D_r/D_s	–	0.5	0.7	0.9
h	cm	0.7	1.0	1.3

$$\%FFA = \frac{0.5 \times A \times N \times W_{cat}}{W} \quad (2)$$

where A is the catalyst volume for the oil titration (mL); W is the sample value (g); N is the normality; and W_{cat} is the molecular mass of the catalyst (g), which is 56.1 for KOH.

Two corresponding reactants (methanol and KOH) were mixed prior to the initiation of the transesterification reaction without further purification; the methoxide solution was thereafter imported to the reaction.

2.2.2 Analytical methods: Three parameters were taken into consideration in the present study for use as the independent variables: the diameter ratio between the rotor and the stator (D_r/D_s), the space between the rotor and the bottom of reactor (h), and the frequency of the rotor's rotary speed (R_f).

For the other effective parameters on the reaction, considering the research studies in the literature, the alcohol to oil molar ratio (6:1), the catalyst concentration (1% w/w%), the reaction temperature (60 °C), and the flow rate of oil (16 ml/min) remained constant in all the experiments [23, 35, 40]. The biodiesel yield and volumetric productivity were measured and calculated for each experiment as the dependent parameters of the study.

At the end of each experiment, a sample of the product, which is a mixture of FAMES and glycerol, was put into a refrigerator to stop the reaction and kept for the GC analysis. A GC unit of the Perkin Elmer clarus-580 model (made in USA), which was based on the BS-EN 14103 standard, was used for calculation of the reaction yield Eq. (3) [13, 14].

$$Y_{FAME} (\%) = \frac{W_{FAME}/M_{FAME}}{3W_{WCO}/M_{WCO}} \times 100 \quad (3)$$

where W_{FAME} is the produced biodiesel (mg); W_{WCO} is the mass of oil (mg); M_{FAME} is the mean molecular mass for the biodiesel; and M_{WCO} is the mean molecular mass of the WCO.

Another dependent variable in the present study is the volumetric production rate of FAME, which is defined as the amount of FAME produced per volume per time Eq. (4) [1, 3].

$$\text{Volumetric production rate } (V_r) = \frac{3\Phi_{oil}Y\left(\frac{MW_{FAME}}{MW_{oil}}\right)\rho_{oil}}{V} \left(\frac{kg_{FAME}}{m^3h}\right) \quad (4)$$

where Φ_{oil} is the volumetric flow rate of the waste cooking oil (m^3/h); ρ_{oil} is the oil density (kg/m^3); Y is the FAME yield (mol %); V is the reactor volume (m^3); MW_{FAME} is the molecular weight of FAME (kg/mol); and MW_{oil} is the molecular weight of the oil (kg/mol).

2.2.3 Statistical analysis: The RSM and the Box–Behnken design method were used to conduct the statistical analysis in the present research study.

First, the dependent variables' levels, according to Table 1, were coded based on the selected method. Second, after running the experiments determined by Design Expert version 7.0 software, the effect of each independent variable and their interaction were investigated by analysis of variance (ANOVA) and using the depicted diagrams. All the experiments were replicated three times to determine the variability of the results and to assess the experimental errors. Finally, the optimal values predicted by the software were evaluated by further experimentation. Table 1 represents the outcomes of this procedure.

3 Results and discussion

The data presented in Table 2 shows the relevant physical and chemical properties of the WCO and also shows the results of the chromatogram of the oil used. As the table suggests, because the level of free fatty acids in the used oil is less than 1%, the base-catalyzed transesterification reaction can be performed in one step, and esterification is not needed.

The SMS reactor for biodiesel production was operated at a reaction temperature of 60 °C, catalyst concentration 1%(w/w) and alcohol to oil molar ratio of 6:1. Table 3 list all the experimental runs and the obtained results.

The Design Expert 7.0 software selected the quadratic polynomial equation as the most suitable regression model for this study. The statistical analysis (ANOVA) based on this equation indicated that there was a significant effect of individual factors and the interactions among the independent parameters on the effectiveness of biodiesel production using SMS reactor. The results are shown in Table 4.

Eq. (5) shows the regression equation used to process the experimental data and the selected model in the RSM. The algebraic sign in this equation indicates the increase or decrease effect. In addition, the coded value of coefficients in this equation emphasized the importance of the corresponding parameter when considering the biodiesel conversion percentage.

Table 2: The chemical and physical properties of the WCO used in this research.

Properties	Unit	Amount
Density	kgm^{-3}	0.905×10^3
Kinematic viscosity	mPa.s	25.580
Acid value	mg KOH/g oil	0.91
Iodine value	$\text{g I}_2 / 100 \text{ g oil}$	67.5
Water content	mg g^{-1}	0.11
Palmitic acid (C16:0)	wt. %	10.71
Stearic acid (C18:0)	wt. %	4.45
Oleic acid (C18:1) ^a	wt. %	26.90
Linoleic acid (C18:2) ^a	wt. %	53.01
Linolenic acid (C18:3) ^a	wt. %	2.67
Other fatty acids	wt. %	2.26
Mean molecular weight of WCO	g mol^{-1}	876.60

^a Carbon atoms number: double bond number.

Table 3: Experimental coded conditions for biodiesel production using the SMS reactor.

Random	Run	Rotational frequency(R_f) (Hz)	h (mm)	D_r/D_s	Biodiesel Yield(%)	$V_r(\text{kg}_{\text{FAME}}/(\text{m}^3_{\text{reactor}} \cdot h))$
5	1	20	1	0.5	79.0	593.93
12	2	40	1.3	0.9	91.0	1447.25
1	3	20	0.7	0.7	84.0	1068.7
6	4	60	1	0.5	79.7	599.19
15	5	40	1	0.7	91.0	1003.42
3	6	20	1.3	0.7	82.0	797.8
2	7	60	0.7	0.7	85.9	1092.91
7	8	20	1	0.9	80.0	1575.238
13	9	40	1	0.7	91.5	1008.94
14	10	40	1	0.7	91.7	1011.14
11	11	40	0.7	0.9	92.8	2398.3
4	12	60	1.3	0.7	83.0	807.54
8	13	60	1	0.9	82.0	1614.61
10	14	40	1.3	0.5	87.9	605.7
9	15	40	0.7	0.5	92.0	776.4
16	16	40	1	0.7	90.8	1001.22
17	17	40	1	0.7	91.4	1007.83

Table 4: Analysis of variance (ANOVA) for biodiesel production using the SMS reactor.

F-value	Mean square	df	Source
439.6934**	0.004541	9	Model
37.95297**	0.000392	1	A- R_f
141.1618**	0.001458	1	B-h
62.73859**	0.000648	1	C- D_r/D_s
1.960581 ^{n.s.}	2.02E-05	1	AB
4.090595*	4.22E-05	1	AC
12.80429**	0.000132	1	BC
3414.881**	0.035271	1	A^2
104.0346**	0.001075	1	B^2
155.4097**	0.001605	1	C^2
	1.03E-05	7	Residual
0.425791 ^{n.s.}	5.83E-06	3	Lack of Fit
	1.37E-05	4	Pure Error
		16	Cor Total

** significant on 1% level of probability.

* significant on 5% level of probability.

^{n.s.} not significant.

$$\begin{aligned} \text{Yield} = & +0.91 + 7E - 3.R_f - 0.013*h + 9E - 3*D_r/D_s - 2.25E - 3*R_f*h + 3.25E - 3*R_f*D_r/D_s + 5.75E \\ & - 3*h*D_r/D_s - 0.092*R_f^2 + 0.016*h^2 - 0.02*(D_r/D_s)^2 \end{aligned} \quad (5)$$

As Figure 5 indicates, the scattering of data derived from the experiment with respect to the data obtained from the model shows the accuracy of the proposed model.

3.1 The effects of R_f and h on biodiesel yield

Figure 6a and 6b shows the interaction between rotational frequency (R_f) and the space between the rotor and the bottom of the reactor (h). The increasing trend of R_f from 20 to 40 Hz resulted in an increase of the biodiesel yield; what follows is an increase of the R_f to more than 40 Hz, which caused a decline in the conversion percentage. In simpler terms, the increase of R_f from 20 to 40 Hz resulted in a 10% increase of the biodiesel yield, which can be attributed to the more intensified mixing caused by the higher R_f . The increase of the R_f improved the mass transfer between the oil and methoxide phases, and this caused the increased conversion of

Figure 5: The experimental data versus predicted points by quadratic model for reaction yield.

Figure 6: (a),(b):The effect of R_f and h on the biodiesel conversion percentage when $D_r/D_s = 0.7$, (c),(d): The effect of R_f and D_r/D_s on the biodiesel conversion percentage when $h = 1$ cm. (e),(f): The effect of h and D_r/D_s on the biodiesel conversion percentage when $R_f = 40$ Hz.

oil to methyl ester. Exceeding the R_f of 40 Hz, however, resulted in decreased efficiency in conversion. This effect can be attributed to the extreme mixing of the two phases and lack of sufficient time for interaction between the two phases. In addition, the increase of the R_f caused decreased residence time for the reactants in the space between the rotor and the stator. Previous studies report similar results. For example, Kraai et al. conducted an experiment on biodiesel production from sunflower oil on a CINC reactor of V-02 model. As inferred from Kraai et al.'s research, the increase of the R_f has a beneficial effect up to the 35 Hz level, which is followed by decreasing conversion when exceeding the 40 Hz level [35].

Considering the ANOVA results of biodiesel yield to be presented in Table 4, it is observed that while increasing of h leads to decreased biodiesel yield, the interaction of two independent variables (AB terms in Table 4) does not have a significant effect on response (biodiesel yield), making such a decrease not to be significant statistically. The change range of the biodiesel yield was 82–84% in the case of the change range of h . However, as the increase of h increased the volume of the reactor (as shown in Eq. (5)) and the increase from 0.7 to 1.3 cm caused a 25% decrease of the volumetric production rate, it could be expected that the minimum level of h is much more desirable.

It is observed in Figure 6a and b that the changing trend of the biodiesel yields with the changes of both the h and R_f factors was of a quadratic nature; however, the trend was shuffle down and shuffle up in R_f and h factors, respectively. This indicates that the optimal points for R_f and h were the interval, mid, and interval sides, respectively.

3.2 The effects of R_f and D_r/D_s on biodiesel yield

Figure 6c and d shows the interaction between R_f and the diameter ratio between the rotor and the stator (D_r/D_s). The increase of R_f had a trend corresponding to the quadratic equation when considered in terms of its

interaction with the diameter ratio between the rotor and the stator (D_r/D_s). It followed an increasing trend followed by a decreasing trend in biodiesel yield. It is worth noting that the increase of D_r/D_s with a medium trend resulted in an increased biodiesel yield in the chosen interval. In this parameter, the increase of D_r/D_s resulted in a reduction of the mixing zone volume, which caused a 2.65 times increase of the volumetric production rate on all R_f levels when considering the minimum to maximum level of the afore-mentioned parameter. In their study on standard and modified CINC[®] reactors for biodiesel production, Abduh et al. used sunflower oil and reported similar results [23]. If the D_r/D_s parameter is dimensionless, then the authors believe that commercial producers of biodiesel will benefit from finding its optimal points for scaling up, as well as building reactors to attain semi-industrial and industrial scales.

3.3 The effects of h and D_r/D_s on biodiesel yield

The interactions between the space of the rotor and the bottom of the reactor (h) and the diameter ratio between the rotor and the stator (D_r/D_s) are illustrated in Figure 6e and f.

As is evident from the figure, as well as Table 4, the interactive effects of these parameters were significant. On each level of h , the biodiesel yield reached its maximum value when D_r/D_s falls into the 0.65–0.85 range. The increase of D_r/D_s meant the reduction of the distance between the two cylinders. The 85% increase of the biodiesel yield as a result of the D_r/D_s increase could be attributed to the formation of a thin layer of reactants between the rotor and the stator, which maximized the shear stress between the oil and methoxide layers and resulted in an accelerated reaction. The reduction of the biodiesel yield in the higher D_r/D_s values could be attributed to the extreme reduction of the distance between the two cylinders, which restricted the formation of a 3D vortex (shown in Figure 2), as well as the lack of residence time for reactants in the mixing zone. The study on the hydrodynamic features of CCCS reactors yielded the dimensions of the experimental standard reactor. The reported values for the rotor's outer dimension and the stator's inner dimension were reported to be 50.75 and 62 mm, respectively. Consideration of these values shows that the D_r/D_s value in this reactor is 0.82 [24]. The values reported by Schuur et al. are consistent with the ones reported in the present study.

In addition, based on the regression model Eq.(5), the biodiesel yield reduces as a result of the h value, and the reduction is of the quadratic type. The minimum biodiesel yield of h was obtained at 0.7 cm. The value reported in Schurr et al.'s research for determining the distance of the rotor from the bottom of the reactor was 7 mm [24].

3.4 The optimization of effective parameters on the production process

The optimal parameters for this process, based on the regression model Eq. (5) and the boundary conditions are illustrated in Table 5. The goal of optimization was to reach the maximum yield and maximum volumetric production rate.

These conditions indicate that the optimal points for the three factors (R_f , h , and D_r/D_s) were determined to be 39 Hz, 0.7 cm, and 0.85, respectively. At these optimal parameters, RSM predicted that the yield and

Table 5: The boundry conditions for reaction optimization using RSM.

Parameter	Goal	Minimum	Maximum	Importance
R_f	In range	20	60	–
h	In range	0.7	1	–
D_r/D_s	In range	0.5	0.9	–
Conversion	Maximize	79	93	*****
Volumetric production rate	Maximize	594	2340	***

Table 6: The produced biodiesel properties in comparison with the ASTM D6751 Standard.

Property	Test method	Limits	Units	Measured property
Water and sediment	ASTM D 2709	<0.05	% volume	0.05
Kinematic viscosity at 40 °C	ASTM D 445	1.9–6.0	mm ² /s	3.7
Sulfated ash	ASTM D 874	<0.02	% mass	0.014
Methanol content	EN 14110	<0.20	% volume	0.16
Flash point, closed Cup	D 93	>130	°C	174
Acid number	ASTM D 664	<0.50	mgKOH/g	0.36
Total glycerin	ASTM D 6584	0.026	% mass	0.017
Oxidative stability	EN 14112	>3	hours	5.3

volumetric production rate would be 93.2% and 1,980 (kg FAME/m³·h), respectively. The optimal values were experimentally evaluated through an extra experimental run in the condition suggested by the software. The value of biodiesel yield and volumetric production rate in the validation experiment were 92.5% and 1,785(kg FAME/m³·h), respectively. The ignorable differences (less than 1%) related to the obtained responses validated the model's predictability, both theoretically and experimentally.

3.5 The physico-chemical properties of the produced biodiesel

In order to be useful, the produced biodiesel must meet the fuel standards for alternative pure and diesel-biodiesel blended fuel in diesel engines. Therefore, following the statistical analysis, biodiesel was produced in the SMS reactor under optimal conditions for conversion. After washing and purifying the produced biodiesel, some of its important properties were measured based on ASTM D6751 (Table 6).

4 Conclusions

This research investigated what design parameters for continuous biodiesel production are most effective when using an SMS reactor. Data analysis showed that there were significant individual and interaction effects of the variables on biodiesel production. The increasing trend of (R_f) from 20 to 40 Hz caused an increase in the biodiesel yield. The increase of (R_f) more than 40 Hz led to a decline in the biodiesel yield. The increase of h resulted in the increased volume of the reactor; increases from 0.7 to 1.3 cm led to the decrease of the volumetric production rate. It should be noted that, on each level of h , the biodiesel conversion percent reached its maximum value when D_r/D_s fell into the 0.65–0.85 range. The optimal points for the three factors (R_f , h , and D_r/D_s) were determined to be 39 Hz, 0.7 cm, and 0.85, respectively. The major properties of the produced biodiesel in this study met the requirements of EN 14214 and ASTM D6751 biodiesel standards.

Acknowledgments: The authors are grateful to the Tarbiat Modares University (<http://www.modares.ac.ir>) for financial supports given under IG/39705 grant for Renewable Energies of Modares research group. Also, the authors express their thankful regards for TMU Renewable Energies Research Institute who provided insight and expertise that greatly assisted this research.

Author contribution: All the authors have accepted responsibility for the entire content of this submitted manuscript and approved submission.

Research funding: The research was funded by Tarbiat Modares University.

Conflict of interest statement: The authors declare no conflicts of interest regarding this article.

References

1. 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.
2. Renewables Global Status Report; 2019:1–335 p. Available from: https://www.ren21.net/wp-content/uploads/2019/05/gsr_2019_full_report_en.pdf.
3. Abbaszaadeh A, Ghobadian B, Omidkhan MR, Najafi G. Current biodiesel production technologies: a comparative review. *Energy Convers Manag* 2012;63:138–48.
4. Qiu Z, Zhao L, Weatherley L. Process intensification technologies in continuous biodiesel production. *Chem Eng Process Process Intensif* 2010;49:323–30.
5. Thompson JC, He BB. Biodiesel production using static mixers. *Trans ASABE* 2007;50:161–6.
6. Sun J, Ju J, Ji L, Zhang L, Xu N. Synthesis of biodiesel in capillary microreactors. *Ind Eng Chem Res* 2008;47:1398–403.
7. Harvey AP, Mackley MR, Seliger T. Process intensification of biodiesel production using a continuous oscillatory flow reactor. *J Chem Technol Biotechnol* 2003;78:338–41.
8. Available at: <https://www.arisdyne.com/content/controlled-flow-cavitation-cfc>.
9. Gülyurt MÖ, Özçimen D, İnan B. Biodiesel production from *Chlorella protothecoides* oil by microwave-assisted transesterification. *Int J Mol Sci* 2016;17:579.
10. Gude V, Patil P, Martinez-Guerra E, Deng S, Nirmalakhandan N. Microwave energy potential for biodiesel production. *Sustain Chem Process* 2013;1:5.
11. Lin JJ, Chen YW. Production of biodiesel by transesterification of *Jatropha* oil with microwave heating. *J Taiwan Inst Chem Eng* 2017;75:43–50.
12. Chee Loong T, Idris A. One step transesterification of biodiesel production using simultaneous cooling and microwave heating. *J Clean Prod* 2017;146:57–62.
13. 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.
14. 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.
15. Dube MA, Tremblay AY, Liu J. Biodiesel production using a membrane reactor. *Bioresour Technol* 2007;98:639–47.
16. Cheng LH, Yen SY, Chen ZS, Chen J. Modeling and simulation of biodiesel production using a membrane reactor integrated with a prereactor. *Chem Eng Sci* 2012;69:81–92.
17. Poddar T, Jagannath A, Almansoori A. Use of reactive distillation in biodiesel production: a simulation-based comparison of energy requirements and profitability indicators. *Appl Energy* 2017;185:985–97.
18. Boon-anuwat N, Kiatkittipong W, Aiouache F, Assabumrungrat S. Process design of continuous biodiesel production by reactive distillation: comparison between homogeneous and heterogeneous catalysts. *Chem Eng Process Process Intensif* 2015;92:33–44.
19. Poddar T, Jagannath A, Almansoori A. Biodiesel production using reactive distillation: a comparative simulation study. *Energy Procedia* 2015;75:17–22.
20. Perez-Cisneros ES, Mena-Espino X, Rodríguez-López V, Sales-Cruz M, Viveros-García T, Lobo-Oehmichen R. An integrated reactive distillation process for biodiesel production. *Comput Chem Eng* 2016;91:233–46.
21. Vedantam S, Joshi JB. Annular centrifugal contactors—a review. *Chem Eng Res Des* 2006;84:522–42.
22. Schuur B, Jansma WJ, Winkelman JGM, Heeres HJ. Determination of the interfacial area of a continuous integrated mixer/separator (CINC) using a chemical reaction method. *Chem Eng Process Process Intensif* 2008;47:1484–91.
23. Abduh MY, Van Ulden W, Van De Bovenkamp HH, Buntara T, Picchioni F, Manurung R, et al. Synthesis and refining of sunflower biodiesel in a cascade of continuous centrifugal contactor separators. *Eur J Lipid Sci Technol* 2015;117:242–54.
24. Schuur B, Kraai GN, Winkelman JGM, Heeres HJ. Hydrodynamic features of centrifugal contactor separators: experimental studies on liquid hold-up, residence time distribution, phase behavior and drop size distributions. *Chem Eng Process Process Intensif* 2012;55:8–19.
25. Vedantam S, Wardle KE, Tamhane TV, Ranade VV, Joshi JB. CFD simulation of annular centrifugal extractors. *Int J Chem Eng* 2012; 2012:1–31.
26. Tang K, Wang Y, Zhang P, Huang Y, Hua J. Optimization study on continuous separation of equol enantiomers using enantioselective liquid-liquid extraction in centrifugal contactor separators. *Process Biochem* 2016;51:113–23.
27. Schuur B, Verkuil BJV, Bokhove J, Minnaard AJ, De Vries JG, Heeres HJ, et al. Enantioselective liquid-liquid extraction of (R,S)-phenylglycinol using a bisnaphthyl phosphoric acid derivative as chiral extractant. *Tetrahedron* 2011;67:462–70.
28. Zhang P, Zhang H, Tang K, Yi J, Huang Y. Influence of pH on enantioselective extraction of aromatic acid enantiomers in centrifugal contactor separators: experiments and simulation. *Separ Purif Technol* 2015;141:68–75.

29. Schuur B, Winkelman JGM, de Vries JG, Heeres HJ. Experimental and modeling studies on the enantio-separation of 3,5-dinitrobenzoyl-(R),(S)-leucine by continuous liquid-liquid extraction in a cascade of centrifugal contactor separators. *Chem Eng Sci* 2010;65:4682–90.
30. Schuur B, Hallett AJ, Winkelman JGM, De Vries JG, Heeres HJ. Scalable enantioseparation of amino acid derivatives using continuous liquid-liquid extraction in a cascade of centrifugal contactor separators. *Org Process Res Dev* 2009;13:911–4.
31. Xu W, Dai G, Tang K, Zhang P, Xiong B, Liu Y. Continuous chiral separation of 2-phenylbutyric acid by liquid-liquid extraction in centrifugal contactor separators. *Separ Purif Technol* 2017;179:53–60.
32. Zhang P, Feng X, Tang K, Xu W. Study on enantioseparation of α -cyclopentyl-mandelic acid enantiomers using continuous liquid-liquid extraction in centrifugal contactor separators: experiments and modeling. *Chem Eng Process Process Intensif* 2016;107:168–76.
33. Tang K, Wang Y, Zhang P, Huang Y, Dai G. Process optimization of continuous liquid-liquid extraction in centrifugal contactor separators for separation of oxybutynin enantiomers. *Separ Purif Technol* 2015;150:170–8.
34. Schuur B, Kraai GN, Winkelman JGM, Heeres HJ. Chemical engineering and processing: process intensification hydrodynamic features of centrifugal contactor separators: experimental studies on liquid hold-up, residence time distribution, phase behavior and drop size distributions. *Chem Eng Process Process Intensif* 2012;55:8–19.
35. 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;154:384–9.
36. Abduh MY, van Ulden W, Kalpoe V, van de Bovenkamp HH, Manurung R, Heeres HJ. Biodiesel synthesis from *Jatropha curcas* L. oil and ethanol in a continuous centrifugal contactor separator. *Eur J Lipid Sci Technol* 2013;115:123–31.
37. 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;57:12742–55.
38. Taylor GI. Stability of a viscous liquid contained between two rotating cylinders. *Philosophical Trans Royal Soc A* 1923;223:289–343.
39. Metcalfe LD, Schmitz AA, Pelka JR. Rapid preparation of fatty acid esters from lipids for gas chromatographic analysis. *Anal Chem* 1966;38:514–5.
40. Fayyazi E, Ghobadian B, Mousavi SM, Najafi G. Intensification of continuous biodiesel production process using a simultaneous mixer- separator reactor. *Energy Sources, Part A Recover Util Environ Eff* 2018;40:1125–36.