

Optimization of Ultrasonic Reactor Geometry for Biodiesel Production using Response Surface Methodology

B. Mostafaei¹, B. Ghobadian^{1*}, M. Barzegar², and A. Banakar¹

ABSTRACT

Biodiesel fuel, which is produced by transesterification reaction between alcohol and vegetable oil/animal fat is proposed as a clean alternative to petro diesel fuel. Today, one of the new technologies to produce biodiesel is using ultrasonic energy that makes production faster, with improved quality and less expensive. Various factors that affect the design of ultrasonic reactors are ratio of vibrating rod diameter to reactor diameter, reactor height, depth of horn penetration into fluid and chamber characteristics (material and shape). In this paper, two parameters namely the ratio of vibrating rod diameter to reactor diameter and reactor height were studied in order to increase the reaction efficiency. In all performed tests, the horn diameter of 14 mm, molar ratio of alcohol to oil of 5 to 1, catalyst concentration of 0.7% wt?? oil, depth of horn penetration into fluid of 15 mm and a cylindrical reactor were used. Experimental design involved the use of Central Composite Design (CCD) and the statistical Response Surface Methodology (RSM). Considering the empirical model, a significant relationship was found between independent and dependent variables with a regression coefficient of 0.99. Taking into account the desirability of increasing the efficiency, the optimal function of reactor diameter and reactor height were 63 and 110 mm, respectively with a reaction yield of 87%. In order to verify the model, function responses in the defined area were tested with five replicates and the average efficiency of the reaction was 87.2%. The obtained model suggests the simultaneous reverse effects of reactor diameter and height on the reaction efficiency.

Keywords: Biodiesel production, Optimization, RSM, Transesterification reaction efficiency, Ultrasonic reactor.

INTRODUCTION

Today, researchers are looking to find a reliable source to replace fossil fuels due to environmental considerations, increase in petrol prices and gradual oil reserves decline. One substitute which has attracted much attention is biodiesel fuel that is produced from a transesterification reaction between vegetable oil/animal fat and alcohol in the presence of an alkaline or acidic catalyst. Biodiesel has several advantages over petrodiesel as it is renewable, has a closed cycle,

reduces emission pollutants such as CO₂, CO, SO₂, smoke, etc. (Murugesan *et al.*, 2009). The main environmental advantages of biodiesel fuel are related to OH chain in its chemical structure. Oxygen present in biodiesel fuel results in a complete combustion process and therefore less unburned hydrocarbons are emitted from the engine exhaust (Carraretto *et al.*, 2004).

The most common method for producing biodiesel is the transesterification of triglycerides or fatty acids with an alcohol in the presence of a strong catalyst (acid, base, or

¹ Department of Agricultural Machinery Engineering, Faculty of Agriculture, Tarbiat Modares University, Islamic Republic of Iran.

* Corresponding author; e-mail: ghobadib@modares.ac.ir

² Department of Food Science and Technology, Faculty of Agriculture, Tarbiat Modares University, Islamic Republic of Iran.



enzymatic). Selection of the catalyst depends on the amount of free fatty acid (FFA). The alkali-catalyzed reaction gives a better conversion in a short time with lower amounts of FFA, while with higher amounts of FFAs, acid catalyzed esterification followed by transesterification is more suited. Industrial processes usually favor base catalysts because alkaline catalysts are less corrosive than acidic compounds (Singh *et al.*, 2007).

Alcohol and oil are immiscible together, so transesterification reaction is limited in terms of mass transfer and hence requires mixing (Ji *et al.*, 2006). Use of ultrasonic wave as a new technology to speed up transesterification reaction, increase reaction efficiency and reduce the amount of catalyst and energy required to produce biodiesel has been reported by several researchers (Hingu *et al.*, 2010; Mootabadi *et al.*, 2010 and Thanh *et al.*, 2010). This would be due to the collapse of cavitation bubbles and the ultrasonic jet that impinges one liquid to another, disrupting the phase boundary thereby causing emulsification. The ultrasound in the chemical processing enhances both the mass transfer and chemical reactions. It offers the potential for shorter reaction times, cheaper reagents and less extreme physical conditions leading to less expensive and smaller chemical plants (Hanh *et al.*, 2008). Transfer of ultrasonic energy by a probe processor into the reaction medium, which is mainly made of titanium, has been used in laboratory environments with more efficiency than a bath processor. Various factors that affect the design of ultrasonic reactors are ratio of vibrating rod diameter to reactor diameter, reactor height, depth of probe penetration into fluid and chamber characteristics (material and shape) (Sutker and Gogate 2009; Gogate *et al.*, 2011).

On the other hand, use of the statistical response surface method in the optimization problems is an effective method for reducing the number of the tests and saving time and materials as well as offering a complete evaluation of the experimental process through creating regression models between dependent and independent variables (Worapun *et al.*, 2011 and Rashid *et al.*, 2009).

In this study, among factors affecting the design of ultrasonic reactors, keeping alcohol to oil molar ratio of 5 to 1, catalyst concentration of 0.7 wt% oil, sonication time of 3 minutes, probe penetration into the reaction solution of 15 mm, probe diameter of 14 mm, ultrasonic power of 55 watts (vibrating amplitude 40%) and temperature of 35 Celsius degree constant during the experiments, both height and ratio of probe to reactor diameter were evaluated to produce biodiesel by ultrasonic energy and to select the most suitable reactor volume. Evaluation of the other parameters change effects on the dependent variable (yield response) was avoided due to the extent and complexity of the issues which would ultimately increase the content volume that was outside the scope of this article.

MATERIALS AND METHODS

Potassium hydroxide and methanol were purchased from Merck and waste oil was obtained from the Faculty of Agriculture, Tarbiat Modares University restaurants. The necessary standard materials for using gas chromatography (such as methyl fatty acids the C18: 2, C18: 1, C18: 0, C17, C16: 1, C16:0) and *n*-heptane were obtained from Sigma Aldrich.

Preparation of Waste Oil

Presence of water in oil has negative effects on the efficiency of conversion of methyl esters. Also free fatty acids (FFA) react with base catalysts and produce soaps that prevent biodiesel separation from glycerin and water. For completing the reaction in the presence of a base catalyst, free fatty acid and water content should be less than 3 and 1 wt %, respectively (Demirbas, 2006). Kitchen waste oil after passing through filter was placed in a cylindrical tank for 24 hours and its sediments and wastes were deposited on the bottom, which were then gently separated

from oil. Oil was stirred for 2 hours at 100°C to evaporate its moisture. Then, for the determination of free fatty acids and water content of oil, titration method (Canakci and Gerpen, 2001) and distillation method were used. Free fatty acids and water content of oil were calculated to be 0.8 and 0.5%, respectively which are suitable for transesterification reaction (Freedman *et al.*, 1984; Ma *et al.*, 1998). The free fatty acid percentage was calculated with Equation 1:

$$FFA\% = \frac{V \times N \times 28.2}{W} \quad (1)$$

Where, *V*, is the volume in ml of titration solution (KOH + H₂O); *N*, is the normality of the titration solution; *W*, is the weight of the oil sample in grams and 28.2, is the molecular weight of oleic acid divided by ten.

Preparation of Methoxide Solution

Since one of the goals of using ultrasonic energy in biodiesel production is to improve the quality and costs reduction, in all experiments the molar ratio of alcohol to oil and catalyst concentration were 5 to 1 and 0.7 wt %, respectively. For this purpose, the total weights of oil for all tests were estimated and the total required methanol and potassium hydroxide were estimated according to the molar weight of oil (859.97 g), molar weight of methanol (32.04 g), molar weight of potassium hydroxide (56.11 g) and ratio of alcohol to oil molar and

catalyst concentration. The total methoxide solution was prepared using magnetic stirring.

Biodiesel Production by Using Ultrasonic Energy

In order to produce biodiesel by ultrasonic energy, the Hielcsher UPS 400 processor was used. This device has a maximum power of 400 watts, working frequency of 24kHz, adjustable vibration pulses (Cycle 0–100%) and amplitude of vibration (20–100%). In all experiments, the continuous vibration of the probe (Cycle= 1) and 40% of maximum amplitude were used. Oil and methoxide solution were prepared according to the experimental design and were transferred into the reactor. In order to avoid probe penetration depth and sonication time effects on the reaction, they were selected as 15 mm and 3 minutes, respectively. Also to set reaction temperature at 35°C, the oil was heated slightly and a water circulator with adjustable temperature was used. Figure 1 shows the experimental set up.

Reaction Efficiency Measuring Method

At the end of each experiment, 2 ml samples were taken from the reactor and the transesterification reaction was stopped using diluted sulfuric acid solution. Then the samples were transferred into a freezer at -5°C and maintained until being analyzed by

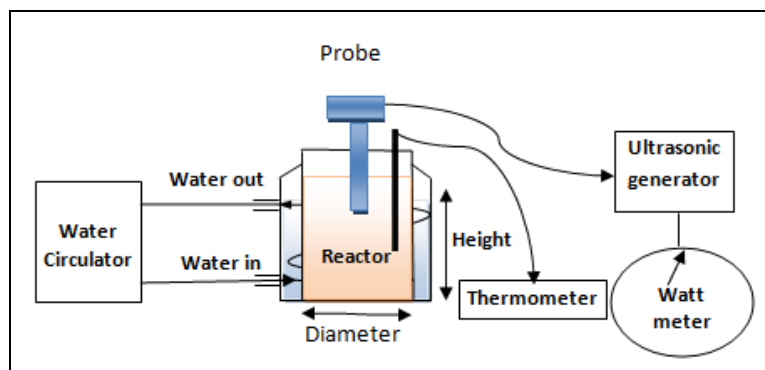


Figure 1. The set-up for experiments.



GC. In order to separate the produced biodiesel, firstly the test tubes were centrifuged at 4,000 rpm for 2 minutes and then the upper phase was separated and weighed carefully. Finally, about 50 mg of the biodiesel phase was transferred into micro-tubes and 1 ml of internal standard solution (C17) with 7mg/ml concentration was added. Approximately 0.5 micro-liter of the prepared solution was injected into the GC system. The Gas Chromatograph of Clarus 580, Manufactured by Perkin Elmer, was equipped with a FID detector and biodiesel capillary column (Varian CP9080) with a length of 30 m, internal diameter of 0.32 mm and the stationary phase thickness of 0.25 μm . Helium was used as the mobile phase. Column temperature program was adjusted according to the EN 14103 standard as follows: The column temperature was firstly set at 60°C and kept for 2 minutes, finally with a rate of 10 °C min⁻¹ it reached 210°C and immediately at a rate of 5 °C min⁻¹ reached 230°C where it remained constant for 10 minutes. The total required time for analysis was 31 minutes. All used parameters in GC analysis have been given in Table 1.

In order to calculate the methyl esters weight in biodiesel phase and the reaction efficiency, the following formulas were used (Yin *et al.*, 2012 and Thanh *et al.*, 2010):

$$W_{FAME} = \frac{\sum A - A_{IS}}{A_{IS}} \times \frac{C_{IS} \times V_{IS} \times M}{m} \quad (2)$$

Table 1. Parameters used in biodiesel GC analysis.

Gas Chromatograph: Perkin Elmer Clarus 580 GC			Column: Select Biodiesel CP9080 30 m x 0.32 mm x 0.25 μm film
Inlet temperature: 250°C Mobile phase flow: 1 ml min ⁻¹ Split flow: 50 ml min ⁻¹			Mobile phase: Helium Gas (purity 99.999) Detector: FID 250°C Air flow: 450 ml min ⁻¹ Hydrogen flow: 45 ml min ⁻¹
Column temperature program			Volume of material injected: 0.5 μl
Hold (min)	Temp. (°C)	Rate (°C min ⁻¹)	
2	60	-	
-	210	10	
10	230	5	

$$Y = \frac{W_{FAME} \times M_{Oil}}{3M_{FAME} \times W_{Oil}} \times 100 \quad (3)$$

Where, W_{FAME} = Total produced methyl esters weight (mg); A_{IS} = Peak area of internal standard; V_{IS} = Volume of internal standard solution (ml); M = Total weight of biodiesel phase in the reaction mixture (mg); M_{Oil} = Oil average molar weight (g mol⁻¹); $\sum A$ = Total peaks area; C_{IS} = Internal standard solution concentration (mg ml⁻¹); m = Weight of biodiesel for GC analysis (mg); Y = The reaction yield percent; W_{Oil} = Used oil weight (mg), M_{FAME} = Produced methyl esters average molar weight (g mol⁻¹);

Response Surface Methodology and Statistical Analysis

In this study, a central composite experimental design (CCED) with two independent variables (height and ratio of reactor diameter to the horn diameter) at 5 levels and a dependent variable (yield response) were used. To investigate the relationship between the process variables and obtained responses and optimization of reactor dimensions, the response surface method was applied. Encoded levels and actual levels of the independent variables are given in Table 2.

The central composite experimental design (CCED), for two independent variables,

predicts 13 experiments with 5 replicates at the center of the experimental design to obtain experimental error, and 8 factorial experiments consisting of four experiments in the corners and four experiments on the experimental design sides (Figure 2).

In order to model the reactor dimensions and reaction efficiency, based on the proposing Design-Expert software, a quadratic equation was used. The general form of a quadratic polynomial equation is as follows:

$$Y = \beta_0 + \sum_{i=1}^k \beta_i X_i + \sum_{i=1}^k \beta_{ii} X_i^2 + \sum_{i=1}^k \sum_{j=i+1}^k \beta_{ij} X_i X_j + \varepsilon \tag{4}$$

Where; Y , β_0 , β_i , β_{ii} , and β_{ij} are response (dependent variable), constant, linear, square and interaction parameters coefficients respectively. X_i and X_j are independent variables and ε is unanticipated error.

In order to analyze the data and draw the corresponding diagrams, the Design-Expert software was used.

RESULTS AND DISCUSSION

Prior to the main tests, the fatty acids of waste cooking oil were extracted using BF_3 method and were then identified by gas

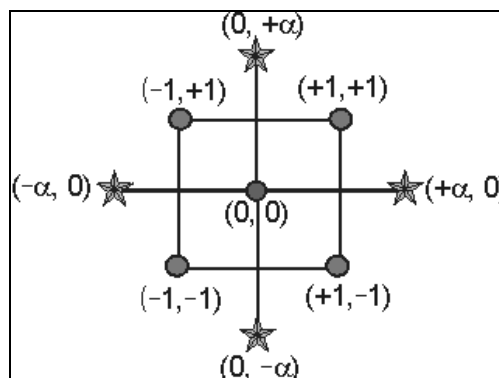


Figure 2. The position of each experiment in the experimental design.

chromatography (David *et al.*, 2003). The oil chromatogram is shown in Figure 3, and the percent of fatty acids in used cooking oil is shown in Table 3. As it is seen, oleic acid, palmitic acid, and lineolic acid are the most dominating with the oil contents of 32.7, 25.6 and 20.82%, respectively.

In order to confirm the selection of alcohol to oil molar ratio, the amount of catalyst, and ultrasonic energy application time, some tests were arranged and carried out in a 255 ml glass vessel. As shown in Figure 4, at 5:1 molar ratio, reaction is more efficient than that at 6:1 molar ratio after 3 minutes elapsed time. This observation can be

Table 2. Actual and encoded independent variables levels in the CCED.

Variables	Label	Levels				
		-1.414 (- α)	-1	0	1	1.414 (α)
Height (mm)	A	27.9	42	76	110	124.1
Diameter (mm)	B	56.8	63	78	93	99.2

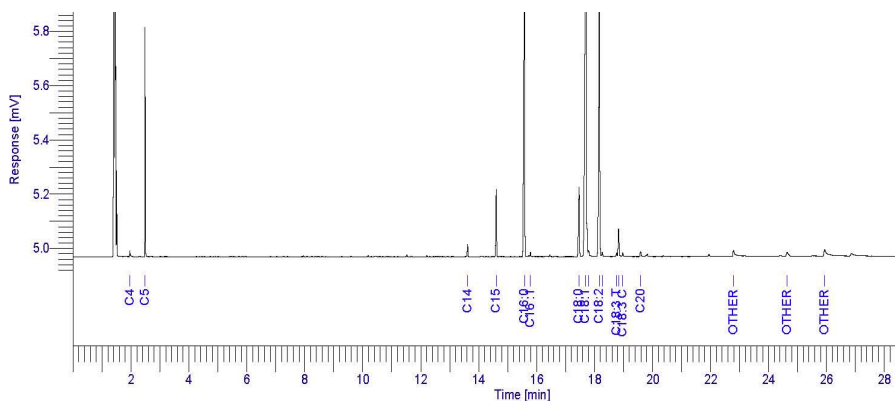


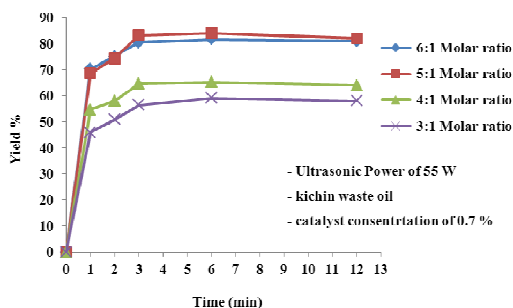
Figure 3. Chromatogram of the kitchen waste oil.

**Table 3.** Waste oil components obtained from GC.

Component	Ret. Time	Area	%	g mol ⁻¹
C4	1.961	28.4	0.22	88
C5	2.487	986.44	7.62	102
C12	13.619	67.74	0.52	200
C15	14.605	390.51	3.01	242
C16:0	15.585	3342.12	25.8	256
C16:1	15.785	22.76	0.18	254
C18:0	17.465	617.44	4.77	284
C18:1	17.695	4266.85	32.94	282
C18:1	17.737	43.04	0.33	282
C18:2	18.167	2717.43	20.98	280
C18:2	18.266	31.11	0.24	280
C18:3	18.761	27.11	0.21	278
C18:3	18.83	198.26	1.53	278
C18:3	18.975	29.95	0.23	278
C20	19.587	39.55	0.31	312
others	22.792	47.97	0.37	340
others	24.674	30.06	0.23	338
others	25.942	66.1	0.51	338

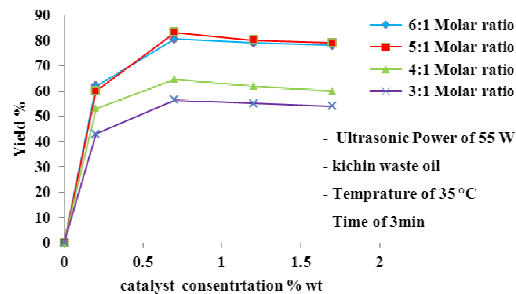
attributed to greater solubility of alcohol in the produced glycerin and reversible transesterification reaction. Similar results have been reported by Hingu and colleagues who selected 6:1 against 7:1 molar ratio (Hingu *et al.*, 2010).

According to Figure 4, ultrasonic energy initiation time in all experiments was selected to be 3 minutes. Reaction efficiency was increased for 3 minutes, but then it gradually reached a constant value. To meet such reaction efficiency using a mechanical mixer under the same conditions, a time more than 1 hour is required (Kim *et al.*,

**Figure 4.** Changes of reaction efficiency versus ultrasonic energy application time.

2004).

Also, some experiments were performed for selecting the proper concentration of the catalyst. The results of these experiments in Figure 5 show that the reaction efficiency initially increased with increasing catalyst concentration, but then the main reaction did not increase and overall reaction efficiency dropped because of saponification reaction between extra catalyst and released fatty acids from triglycerides branches. This result is consistent with earlier studies where the best obtained yields in the investigation of alkaline catalyst concentration effect on

**Figure 5.** Changes of reaction efficiency versus catalyst concentration.

the transesterification reaction by using ultrasonic energy, has been reported to be low concentration of the catalyst (0.5 wt% oil) (Stavarache *et al.*, 2005). For similar reasons, Hingu *et al.* chose 1 wt% catalyst concentration against higher concentrations (Hingu *et al.*, 2010).

After obtaining proper experimental conditions (concentration of catalyst, time and alcohol to oil molar ratio), the main tests were conducted in accordance with Table 4 and regression model between independent variables and dependent variable was as follows:

$$\text{Yield} = 82.84 + 0.20 \times A - 0.98 \times B - 2.7 \times A \times B - 0.24 \times A^2 + 0.34 \times B^2 \quad (5)$$

The positive and negative sign in each part show the increasing and decreasing effects of the parameters on each response, respectively. Therefore, an increase in height (A) has a positive effect whereas an increase in diameter has a negative effect on the reaction efficiency. Also, simultaneous increases or decreases in height and diameter lead to negative effects on the reaction efficiency (note the negative coefficient mark). As shown in Table 4, the most efficient reaction occurred at 110 mm of height and 63 mm of diameter.

Applying surface response methodology and comparing the observed values with their predicted ones in Figure 6, the

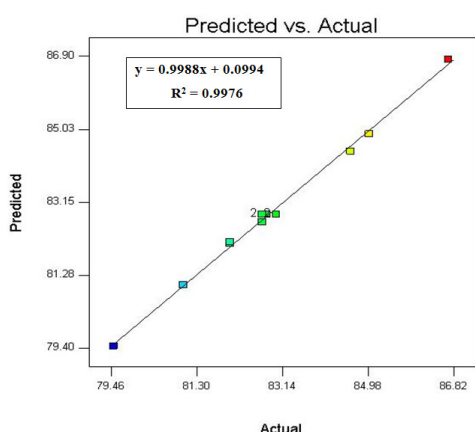


Figure 6. The predicted reaction efficiency versus the observed reaction efficiency.

convergence of these numbers could be observed.

The linear relationship among these values shows a satisfactory correlation between the obtained results and the empirical method on one hand, and correlation between the predicted values and the statistical method on the other. The results of regression analysis are shown in Table 5. The regression coefficient of variance indicates reaction efficiency ($P < 0.0001$).

The diagram of dependent variable response level (reaction efficiency) for all the variation of independent variables is shown in Figures 7 and 8 in 3D and contour lines.

The F term 329.03 for the model shows

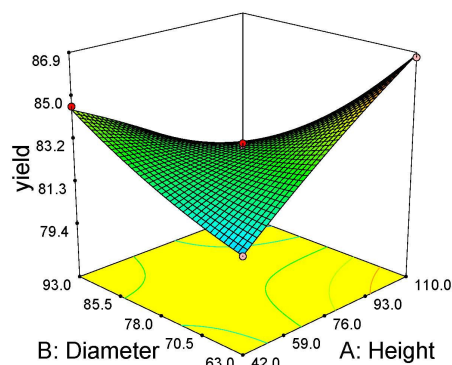


Figure 7. Three-dimensional response surface diagram of the reaction efficiency of biodiesel production as a function of independent variables.

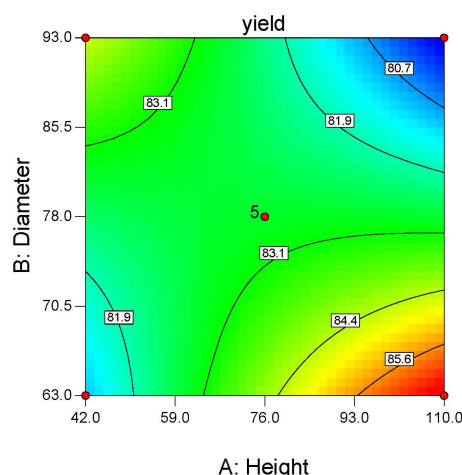


Figure 8. Contour lines per output response for changes in reactor dimensions.

**Table 4.** Tests design based on the CCD including independent variables (two) and dependent variable (one).

Test number	Height	Diameter	Y% response (Reaction yield)	Yield/Energy 1 KJ ⁻¹
	A	B		
1	0	0	83	8.30
2	0	0	82.7	8.44
3	0	-1.414	82	8.20
4	0	1.414	82.7	8.35
5	1	-1	86.7	8.68
6	0	0	82.7	8.27
7	1	1	79.5	8.11
8	-1.414	0	85	8.50
9	-1	1	84.6	8.74
10	0	0	82.8	8.41
11	1.414	0	82	8.31
12	-1	-1	81	8.09
13	0	0	83	8.39

Table 5. The results of regression variance analysis and the related terms.

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob> F
Model	38.516	5	7.703	329.035	< 0.0001
A-Height	0.316	1	0.316	13.497	0.0079
B-Diameter	7.688	1	7.688	328.401	< 0.0001
AB	29.160	1	29.160	1245.538	< 0.0001
A ²	0.397	1	0.397	16.937	0.0045
B ²	0.787	1	0.787	33.596	0.0007
Residual	0.164	7	0.023		
Lack of Fit	0.072	3	0.024	1.042	0.4649
Pure Error	0.092	4	0.023		
Cor Total	38.680	12			

that this model is significant and only 0.01% of variations in F is related to the errors. Also, less than 0.05 p -values of the model indicate that every part of the model is significant.

According to the obtained results, a decrease in reactor diameter leads to an increase in efficiency of biodiesel production reaction such that the most efficient diameter was selected to be 63 mm. Similar results were reported indicating that increasing the ratio of horn diameter to reactor diameter (up to a definite level) or decreasing reactor diameter at constant horn diameter, increases the cavitation activity, and this efficient ratio would be different based on the chemical activity (Nanzai *et al.*,

2009; Kumar *et al.*, 2006). Moreover, increasing the diameter and decreasing the height, increases the reaction efficiency which was definitely not the most efficient value.

The investigation by Kleima and coworkers to develop a 20kHz reactor based on the numerical simulation of ultrasonic energy local distribution and its quality comparison, showed that the maximum cavitation was at 25 mm of depth and 90 mm of reactor diameter as well as 13 mm of height for vibrating probe with 13 mm of diameter and in water condition (Kleima *et al.*, 2007). The difference between the obtained results might be due to the different conditions (water and oil), different

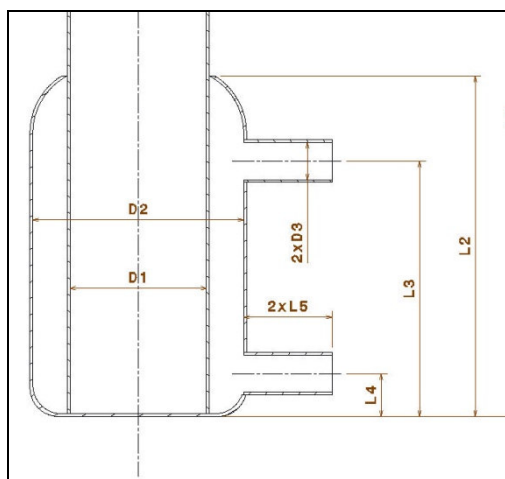


Figure 9. The optimized ultrasonic reactor with $D1=63$ mm and $L2=110$ mm.

penetration depths (15 and 25 mm) and different probe diameters (13 and 14 mm). The optimized ultrasonic reactor is shown in Figure 9 schematically.

The optimized sonochemical reactor generating acoustic cavitation was compared with the magnetic stirring operation (magnetic rod with a diameter of 6 mm, length of 26 mm and rotational speed of 400 rpm) and the obtained results are shown in Figure 10. It can be seen from the figure that acoustic cavitation results in 89% conversion whereas the magnetic stirring method results in a much lower extent of conversion (50%) over similar time of operation as 12 minutes. This can be attributed to the fact that as the reaction is

mass transfer controlled, the micro level turbulence generated due to cavitation bubbles results in higher availability of the interfacial area and hence higher conversion. The mass transfer of triglycerides from oil phase towards the methanol–oil interface could be a rate limiting step and hence poor mass transfer between the two phases results in a slow reaction rate (Noureddini *et al.*, 1997).

CONCLUSIONS

In this research work, the effect of ultrasonic reactor height and diameter on the efficiency of biodiesel production reaction was studied using surface response method and central composite design (CCD). In order to confirm the selection of alcohol to the oil molar ratio, catalyst concentration and sonication time, some tests were carried out. Reaction was more efficient at 5:1 molar ratio than the one with 6:1 and 4:1 molar ratio after 3 minutes elapsed time. The results showed that the reaction efficiency increased with catalyst concentration initially, but after 0.7 % wt, overall reaction efficiency dropped. In addition, reaction efficiency increased for 3 minutes, but then gradually leveled off. Therefore, molar ratio of alcohol to oil, catalyst concentration, and sonication time in all experiments were 5:1,

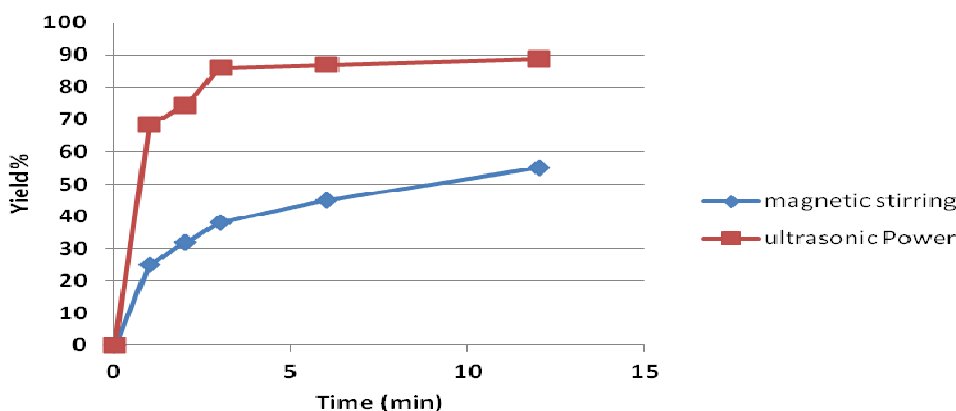


Figure 10. Comparison of reaction yield using ultrasonic power and magnetic stirring (methanol to oil ratio of 5:1, catalyst concentration of 0.7% by wt., reactor volume of 343 ml, ultrasonic power of 55 W, stirring rod with diameter of 6 mm, length of 26 mm which operating at 400 rpm).



0.7% and 3 minutes, respectively. The diameter size and height of ultrasonic reactor had a significant effect on the reaction efficiency. Thus, the produced methyl esters during the process were increased by the decrease in diameter provided that the reactor height was increased simultaneously. In other words, decreasing the reactor height and diameter simultaneously led to significant decrease in reaction efficiency. The most efficient combination of reactor height and diameter to maximize the reaction efficiency were 63 and 110 mm, respectively, which resulted in 87% efficiency. Having the influencing parameters (molar ratio, catalyst concentration, time, etc) constant, we conducted 5 tests on efficient points (reactor height) for the validation of the results and obtained an average reaction efficiency of 87.2%. Also acoustic cavitation resulted in 89% conversion whereas the magnetic stirring method brought about much lower extent of conversion (50%) over similar time of operation as 12 min.

ACKNOWLEDGEMENTS

This study has been carried out at Agriculture Faculty of Tarbiat Modares University using equipment and facilities of the Bioenergy Research Center. The authors would hereby appreciate the manager and the officers-in-charge for providing financial supports.

REFERENCES

1. Canakci, M. and Van Gerpen, J. 2001. Biodiesel Production from Oils and Fats with High Free Fatty Acids. *Trans. ASAE*, **44(6)**: 1429–1436.
2. Carraretto, C., Macor, A., Mirandola, A., Stoppato, A. and Tonon, S. 2004. Biodiesel as Alternative Fuel: Experimental Analysis and Energetic Evaluations, *Energy Sour.*, **29**: 2195–2211.
3. Klíma, J. Frias-Ferrer, A. González-García, J. Ludvík, J. Sa'ez, V. and Iniesta, J. 2007. Optimisation of 20kHz Sonoreactor Geometry on the Basis of Numerical Simulation of Local Ultrasonic Intensity and Qualitative Comparison with Experimental Results, *Ultrason. Sonochem.* **14**: 19–28.
4. David, F. Sandra, P. And Wylie, P. L. 2003. Improving the Analysis of Fatty Acid Methyl Esters Using Retention Time Locked Methods and Retention Time Databases: Application Food, Agilent Application Note. I. Agilent Technologies. Available on: <http://www.chem.agilent.com/Library/applications/5988-5871EN.pdf>
5. Demirbas, A. 2006. Biodiesel Production via Non-catalytic SCF Method and Biodiesel Fuel Characteristics. *Energy Conv. Mgmt.* **47**: 2271–82.
6. Freedman, B. Pryde, E. H. and Mounts, T. L. 1984. Variables Affecting the Yields of Fatty Esters from Transesterified Vegetable Oils. *J. Am. Oil Chem. Soc.* **61**: 1638–1643.
7. Gogate, P. R. Sutkar, V. S. and Pandit, A. B. 2011. Sonochemical Reactors: Important Design and Scale up Considerations with a Special Emphasis on Heterogeneous Systems, *Chem. Eng. J.*, **166**: 1066–1082.
8. Ji, J. Wang, J., Li, Y., Yu, Y. and Xu, Z. 2006. Preparation of Biodiesel with the Help of Ultrasonic and Hydrodynamic Cavitation. *Ultrasonics*, **44**: e411–e414.
9. Kim, H. J., Kang, B. S., Kim, M. J., Park, Y.M., Kim, D. K., Lee, J. S. and Lee, K. Y. 2004. Transesterification of Vegetable Oil to Biodiesel Using Heterogeneous Base Catalyst. *Catal. Today*, **93–95**: 315–20.
10. Kumar, A. Kumaresan, T. Pandit, A. B. and Joshi, J. B. 2006. Characterization of Flow Phenomena Induced by Ultrasonic Horn. *Chem. Engin. Sci.*, **61**: 7410-7420.
11. Hanh, H. D., Dong, N. T., Starvarache, C., Okitsu, K., Maeda, Y. and Nishimura, R. 2008. Methanolysis of Triolein by Low Frequency Ultrasonic Irradiation, *Energy Converse. Manage.*, **49**: 276–280.
12. Hingu, M. H. Gorate, P. R. and Rathod, V. K. 2010. Synthesis of Biodiesel from Waste Cooking Oil Using Sonochemical Reactors, *Ultrason. Sonochem.*, **17**: 827–832.
13. Ma, F., Clements, L. D. and Hanna, M. A. 1998. The Effects of Catalyst, Free Fatty Acids, and Water on Transesterification of Beef Tallow. *Trans. ASAE*, **41**: 1261–1264.
14. Mootabadi, H., Salamatinia, B., Bhatia, S. and Abdullah, A. Z. 2010. Ultrasonic Assisted Biodiesel Production Process from

- Palm Oil Using Alkaline Earth Metal Oxides as the Heterogeneous Catalyst. *Fuel*, **89**: 1818–1825.
15. Murugesan, A., Umarani, C., Chinnusamy, T. R., Krishnan, M., Subramanian, R. and Neduzchezain, N. 2009. Production and Analysis of Bio-diesel from Non-edible Oils: A Review. *Renew. Sust. Energ. Rev.*, **13**: 825–834.
 16. Nanzai, B., Okitsu, K., Takenaka, N., Bandow, H., Tajima, N. and Maeda, Y. 2009. Effect of Reaction Vessel Diameter on Sonochemical Efficiency and Cavitation Dynamics, *Ultrason. Sonochem.*, **16**:163–168.
 17. Nouredini, H. and Zhu, D. 1997. Kinetics of Transesterification of Soybean Oil. *J. Am. Oil Chem. Soc.*, **74(11)**: 1457–1463.
 18. Rashid, U., Anwar, F., Ansari, T. M., Arif, M. and Ahmad, M. 2009. Optimization of Alkaline Transesterification of Rice Bran Oil for Biodiesel Production Using Response Surface Methodology, *J. Chem. Technol. Biotechnol.*, **84**: 1364–1370.
 19. Singh, A. K., Fernando, S. D. and Hernandez, R. 2007. Base Catalyzed Fast-transesterification of Soybean Oil Using Ultrasonication. *Energy Fuels*, **21**: 1161–1164.
 20. Stavarache, C., Vinatoru, M., Nishimura, R. and Maeda, Y. 2005. Fatty Acids Methyl Esters from Vegetable Oil by Means of Ultrasonic Energy. *Ultrason. Sonochem.*, **12**:367–72.
 21. Sutkar, V. S. and Gogate, P. R. 2009. Design Aspects of Sonochemical Reactors: Techniques for Understanding Cavitation Activity Distribution and Effect of Operating Parameters. *Chem. Engin. J.*, **155**: 26–36.
 22. Thanh, L. T., Okitsu, K., Sadanaga, Y., Takenaka, N., Maeda, Y. And Bandow, H. 2010. Ultrasound-assisted Production of Biodiesel Fuel from Vegetable Oils in a Small Scale Circulation Process. *Bioresource Technol.*, **101**: 639–645.
 23. Worapun, I., Pianthong, K. and Thaiyasuit, P. 2011. Optimization of Biodiesel Production from Crude Palm Oil Using Ultrasonic Irradiation Assistance and Response Surface Methodology, *J. Chem. Technol. Biot.*, **87(2)**: 189–197.
 24. Yin, X., Ma, H., You, Q., Wang, Z. and Chang, J. 2012. Comparison of Four Different Enhancing Methods for Preparing Biodiesel through Transesterification of Sunflower Oil. *Appl. Energy*, **91**: 320–325.

بهبود سازی رآکتور فراصوتی به منظور تولید بیودیزل با کمک روش سطح پاسخ

ب. مصطفایی، ب. قبادیان، م. برزگر و ا. بناکار

چکیده

سوخت بیودیزل محصول واکنش ترانس استریفیکاسیون بین الکل و روغن بوده و به عنوان سوختی پاک و جایگزین گازوئیل نفتی مطرح است. امروزه یکی از فناوری‌های نوین تولید بیودیزل استفاده از انرژی فراصوت است که باعث تولید سریع تر، با کیفیت تر و ارزان تر آن می‌شود. عوامل مختلفی در طراحی رآکتور فراصوتی موثر هستند که از آن جمله می‌توان نسبت قطر میله ارتعاشی به قطر رآکتور، ارتفاع رآکتور، عمق نفوذ میله درون مایع و ویژگی‌های محفظه (جنس و شکل رآکتور) را نام برد. در این مقاله دو عامل نسبت قطر میله ارتعاشی به قطر رآکتور و ارتفاع رآکتور به منظور افزایش راندمان تولید مورد بررسی قرار گرفت. در تمامی آزمایش‌ها از میله ارتعاشی با قطر ۱۴ میلی متر، نسبت مولی



الکل به روغن ۵ به ۱، غلظت کاتالیزور ۰/۷ درصد وزنی روغن، عمق نفوذ میله درون مایع ۱۵ میلی متر و شکل استوانه‌ای رآکتور استفاده شد. طرح آزمایشی شامل استفاده از طرح آزمایشی مرکب مرکزی (CCD) و روش آماری سطح پاسخ (RSM) بود. با توجه به مدل تجربی به دست آمده، ارتباط معنی داری میان متغیرهای مستقل و وابسته با ضریب تبیین ۰/۹۹ وجود داشت. با توجه به مطلوبیت افزایش راندمان، نقاط بهینه تابع قطر رآکتور ۶۳ میلی متر، ارتفاع رآکتور ۱۱۰ میلی متر با راندمان واکنش ۸۷ درصد تعیین گردید. در حالی که برای رسیدن به چنین بازدهی در شرایطی مشابه و با استفاده از روش های مرسوم به صرف زمان و انرژی چندین برابر نیاز است. به منظور راست آزمایی مدل، پاسخ های تابع در نقاط مذکور با ۵ تکرار آزمایش شده و میانگین راندمان واکنش ۸۷/۲ درصد به دست آمد. همچنین مدل به دست آمده حاکی از تأثیر معکوس همزمان قطر و ارتفاع رآکتور بر راندمان واکنش می باشد.