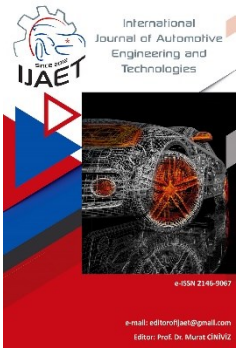
e-ISSN: 2146 - 9067

International Journal of Automotive Engineering and Technologies

journal homepage:
https://dergipark.org.tr/en/pub/ijaet



Original Research Article

Prediction and optimization of biodiesel production by using ANN and RSM

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ARTICLE INFO

Orcid Numbers
1. 0000-0002-4072-6784

Doi: 10.18245/ijaet.1057170

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Received: Jan 17, 2022
Accepted: Apr 22, 2022

Published: 01 July 2022

Published by Editorial Board Members of IJAET

ABSTRACT

This experimental work examined the prediction and optimization of biodiesel production from pomegranate seed oil using Artificial Neural Networks (ANN) and Response Surface Methodology (RSM) with central composite design and The transesterification method chosen for biodiesel production. The Central Composite Design (CCD) optimization conditions were methanol/oil molar ratio (3:1 to 11:1), catalyst rate (0.5 wt% to 1.50 wt%), temperature (50 °C to 70 °C) and time (45 min to 105 min). The process factors were optimized by using CCD based on the RSM method and developed an ANN model to predict biodiesel yield. The optimum yield was found 95.68% with optimum process parameters as 8.01:1 methanol/oil molar ratio, 1.08 wt% catalyst rate, 70 °C temperature and 45 min time. The coefficient of determination (R^2) acquired from the response surface methodology model is 0.9887 and is better when compared to the coefficient of determination (R^2) of 0.9691 acquired from the Artificial neural network model. According to the results, using RSM and ANN models is beneficial for optimizing and predicting the biodiesel production process.

Keywords: ANN; optimization; prediction; pomegranate biodiesel; RSM

1. Introduction

Renewable fuels are an important issue for the reason of increasing population, energy demand, and global warming [1]. For that reason, researchers have given more attention to finding new alternative fuels for diesel engines [2]. Biodiesel is the most useful renewable source for diesel engines [3]. It has important advantages such as biodegradability, non-toxic, and renewable [4,5]. It can be also used in diesel engines without modification [6]. Many diverse methods are used in biodiesel production. Alcoholysis or transesterification method is the most popular method. In transesterification reaction triglycerides and short chain alcohol although catalyst is used to produce monoester [7]. Optimization of the production process is an important and considerable issue and it provides to increase bio-diesel efficiency and decrease cost production. This study is focused on optimizing and modelling of pomegranate seed oil biodiesel production by using two different

mathematical tools named by response surface methodology (RSM) and artificial neural network (ANN).

RSM model is useful tool for optimization of many fields. RSM is used for developing, improving, and optimizing complex processes. Response surface methodology is used to find optimum parameters for transesterification process [8,9]. The advantage of RSM is reduced the experimental runs to provide sufficient data statistically acceptable outcomes [10].

ANN is one of the best methods for the prediction and modeling of biodiesel yield. This method had been used in many parts of engineering because it can be analyzing incomplete data successfully [11]. In recent years, the ANN model has been used for the prediction of biodiesel yield [12].

There are many studies in which prediction and optimize the process parameters in biodiesel production by using ANN and RSM software [13-17]. Gautam et. al. [18] used response surface methodology for optimization of jatropha oil ethyl ester. They obtained maximum production of jatropha biodiesel at the process parameters viz molar ratio 8.5, reaction time 89.67 min, reaction temperature 70.1°C, and catalyst.0.62wt%. Naidoo et. al. [19] studied the optimization of biodiesel production from waste cooking oils. The biodiesel production parameters for optimization were methanol to oil ratio, catalyst amount, temperature, and time. They found that the optimum parameters for the biodiesel production process were a 0.3:1 methanol to oil ratio, catalyst amount of 0.75% wt, a reaction temperature of 68.4 °C and a reaction time of 1.9 hours. Kumar et. al. [20] investigated the transesterification process of the jatropha algae oil blend and they used the ANN technique for the prediction of biodiesel yield. Ayola et. al. [21] used ANN and RSM optimization tools to compare the relationship between biodiesel yield from waste soybean oil and its process parameters. They found that the coefficient of determination values (R^2) are 0.93 and 0.98 for RSM and ANN respectively. Soji-Adekunle et. al. [22] studied the ANN and RSM modeling tools to predict waste cooking biodiesel yield. They calculated the coefficient of determination values (R^2) and average absolute deviation (AAD). They found that the coefficient of determination values (R^2) are 0.9950 and 0.9843 and the average absolute deviation (AAD) is 0.4930 and 0.9376 for ANN and RSM, respectively. Bharadwaj et. al., [23] produced biodiesel from rubber seed oil by using heterogeneous catalyst fluorite. They used a central composite design based on response surface methodology to identify the important parameters which affect the methyl ester yield and optimizing the process parameters. They used ANN for creating a network model. They comprised RSM and ANN responses to find the best model for predicting bio-diesel production.

The objective of this work is to optimize pomegranate seed oil biodiesel production parameters and investigate the impacts of production parameters on biodiesel conversion rate by using RSM based on a central composite design. And also artificial neural network is used for predicting of the experimental results. Finally, a comparison of RSM and ANN methods for the prediction of Pomegranate seed oil methyl ester yield has been made.

2. Material and Method

2.1. Materials

Pomegranate seed oil was used as feedstock in the transesterification reaction. Pomegranate seed oil was purchased from local markets. Sodium hydroxide pellets (NaOH) and methanol (CH_3OH) were used as catalysts and alcohol, respectively.

2.2. Experimental set up

The setup used for biodiesel production consisted of a 250 ml spherical glass reactor equipped with a condenser, contact thermometer, and magnetic stirrer. For each run, 50 ml of pomegranate seed oil was placed in the glass reactor and heated up to the desired temperature. Next, necessary amounts of methyl alcohol and sodium hydroxide were used to obtain the sodium methoxide mixture. Sodium methoxide solution was prepared using different alcohol molar ratios and catalyst amounts. Then this solution was added to the preheated pomegranate seed oil. The transesterification reaction was carried out at different temperatures and times. After completion of the transesterification reaction, the crude biodiesel was transferred into a separation funnel for 8

hours. Later, crude glycerine was removed from methyl ester, and biodiesel was washed and heated up to 105 ℃ to remove residuals and water contents.

2.3. Analysis of fuel properties method

The fuel properties of biodiesel at optimum conditions were investigated according to the standards. Kinematic viscosity, density, flash point, and higher heating value were measured as fuel properties.

Table 1. Independent parameters and their levels

	Levels				
Parameters	-2	-1	0	1	2
Methanol to oil molar ratio	3	5	7	9	11
Catalyst amount (wt%)	0.5	0.75	1.0	1.25	1.5
Reaction temperature (℃)	50	55	60	65	70
Reaction time (min)	45	60	75	90	105

2.4. Response surface methodology (RSM)

Response surface methodology was used in this work for statistical analysis of experimental values using Minitab Statistical Software. A central composite design (CCD) of RSM was used to optimize biodiesel production parameters. A total of 30 experiments were performed. The design involves 16 factorial points, 8 axial points, and 6 center points. The process parameters chosen- methanol to oil molar ratio (A), catalyst rate (B), reaction temperature (C), and reaction time (D) were selected based on literature. The four independent variables and their levels were given in Table 1. RSM model uses quadratic polynomial model to estimate response surface which is shown in Eq. (1) [24].:

$$Y = \beta_0 + \sum_{i=1}^{n} \beta_i x_i + \sum_{i=1}^{n} \beta_{ii} x_i^2 + \sum_{i>j}^{n} \sum_{j}^{n} \beta_{ij} x_i x_j + e \quad (1)$$

Where Y is predicted output response, n is the number of factors, β_0 constant, β_i, β_{ii} and βij are linear, quadric and interaction coefficients, respectively, xi and xj are independent factors and e is error.

2.5. Artificial neural network

ANNs are well-known for being a powerful and nonlinear tools. The nonlinear characteristic of ANN makes them important since most the real-life issues are nonlinear [25]. The essence of the system comes from the imitation of the human nervous system. ANNs can learn from previously supplied data by building input-output mapping for the prediction of various parameters [26]. A multilayer neural network system mainly consists of three-layer; namely; input, hidden, and output. In some cases, there may be more than one hidden layer [27].

In this work, MATLAB (R2020a) software was used to develop the ANN model. For network training, TRAINLM (Lavenberg–Marquardt) algorithm was used. The architecture of the ANN model has occurred to an input layer, an output layer, and a hidden layer with 9 neurons. The selection of the number of hidden neurons was calculated by a trial-and-error procedure. This was done by testing different numbers of neurons until the minimum value of the mean-square error (MSE) is determined. Figure 1. shows the architecture of ANN for pomegranate seed oil biodiesel.

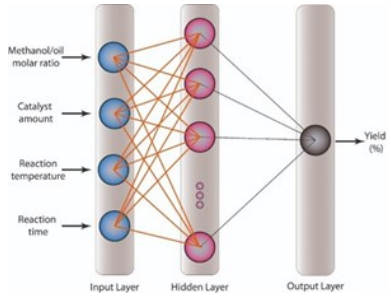

Figure 1. ANN Architecture

3. Results and Discussions

3.1 Optimization by response surface methodology

The impacts of methanol/oil molar ratio, catalyst rate, reaction temperature and reaction time were determined using RSM based central composite design method. For modeling and optimization of biodiesel production, four independent variables with five levels were employed to central composite design. Table 2 gives the process parameters and their levels with the responses. Eq. (2) was adopted represent the quadratic model (coded-basis).

$$Y = -87.9 + 8.09A + 26.08B + 3.751C + 0.323D - 0.4154A^2 - 38.38B^2 - 0.02246C^2 + 0.000449D^2 + 3.238AB - 0.0619AC - 0.01312AD + 0.295BC + 0.2317BD - 0.0892CD \quad (2)$$

Where Y is biodiesel yield, a (methanol/oil molar ratio), B (catalyst amount), C (reaction

temperature), D (reaction time) are input parameters.

Analysis of variance (ANOVA) used to find the importance of the model is given in Table 3. P-value and F-value are demonstrated the model significance and its consistency, respectively [22]. According to Table 3, F-value is 82.02 and the p-value is <0.0001 means the designed model is significant. The value of $p<0.05$ demonstrates model terms are significant, so that A, B, C, A^2, B^2 and C^2, AB, AC, AD, BC, BD, CD are significant model terms. The coefficient terms with p-values more than 0.05 are insignificant [28] and abrogates from the regression model. This model can be used to navigate the design space. The model reduces to Eq. (3), after eliminating the insignificant coefficient terms:

$$Y = -87.9 + 8.09A + 26.08B + 3.751C - 0.4154A^2 - 38.38B^2 - 0.02246C^2 + 3.238AB - 0.0619AC - 0.01312AD + 0.295BC + 0.2317BD - 0.0892CD \quad (3)$$

The quality of model fit is assessed by the coefficient of determination (R^2). The analysis of the regression equation by ANOVA showed that the R^2 value was 0.9887. The adjusted R^2 and predicted R^2 values were 0.9767 and 0.9461 respectively. There is a reasonable agreement between the predicted R^2 and adjusted R^2 values because the difference is less than 0.2. Therefore, this model could be used in the theoretical prediction of the pomegranate seed oil biodiesel production process.

3.2. Analysis of response parameters

The effects of process parameters on biodiesel production were analyzed by three- dimensional surface plots. Figure 2-7 demonstrates the three-dimensional surface plots representing two independent variables while keeping other variables constant. According to the Table 3, methanol/oil molar ratio catalyst amount and reaction temperature are the most significant variables for biodiesel conversion rate whereas reaction temperature had little importance and catalyst amount shows highest 197.21 sums of squares than molar ratio (168.50) and temperature (98.53) and therefore catalyst amount proves to be dominating factor.

Table 2. CCD Response Surface Design and Artificial Neural Network values for pomegranate seed biodiesel yield

Run	Methanol to oil	Catalyst amount (wt%)	Reaction temperature (°C)	Reaction time (min)	Experimental yield (%)	RSM yield (%)	ANN yield (%)
1	5	0.75	55	60	83.7	83.9858	83.6927
2	5	1.25	65	90	88.5	88.9400	88.4731
3	5	0.75	65	90	86.2	85.9692	86.1956
4	7	1.00	60	75	91.2	91.9900	91.4930
5	9	1.25	55	60	89.9	89.9858	89.8971
6	5	1.25	55	90	85.3	85.0025	86.1203
7	9	1.25	65	60	94.1	93.7733	92.8071
8	9	0.75	55	90	84.3	84.1358	84.3317
9	7	1.00	60	75	91.5	91.9900	91.4930
10	9	0.75	55	60	86.0	85.6400	85.9954
11	9	0.75	65	60	88.0	87.5525	87.2638
12	5	0.75	65	60	87.9	88.1733	87.8967
13	9	1.25	55	90	92.7	92.1067	91.2233
14	9	1.25	65	90	93.3	93.7692	93.2939
15	5	0.75	55	90	83.6	83.9067	83.5917
16	9	0.75	65	90	83.7	83.9233	83.6974
17	7	1.00	60	75	91.8	91.9900	91.4930
18	7	1.00	60	75	91.5	91.9900	91.4930
19	5	1.25	65	60	87.9	87.5192	86.1099
20	5	1.25	55	60	81.8	81.4567	82.5339
21	3	1.00	60	75	82.0	81.7908	81.9968
22	7	1.00	70	75	91.3	92.9075	90.6027
23	7	1.50	60	75	85.1	85.4908	85.0863
24	7	1.00	60	75	91.5	92.0700	91.4930
25	7	0.50	60	75	79.0	78.1742	81.5343
26	7	1.00	50	75	87.5	87.0575	87.4919
27	7	1.00	60	75	93.1	92.0700	91.4930
28	7	1.00	60	45	92.1	92.4748	92.0956
29	7	1.00	60	105	92.0	92.3908	91.9878
30	11	1.00	60	75	88.0	88.2742	87.9978

Table 3. ANOVA for response surface quadratic model for pomegranate seed oil biodiesel

Source	DF	Sum of squares	Mean square	F-value	P-value	
Model	15	465.102	31.007	82.02	0.000	significant
A-methanol to oil	1	63.700	63.700	168.50	0.000	
B-catalyst amount	1	74.554	74.554	197.21	0.000	
C-reaction temperature	1	37.250	37.250	98.53	0.000	
D-reaction time	1	0.150	0.150	0.40	0.538	
AB	1	41.926	41.926	110.90	0.000	
AC	1	6.126	6.126	16.20	0.001	
AD	1	2.481	2.481	6.56	0.023	
BC	1	2.176	2.176	5.75	0.031	
BD	1	12.076	12.076	31.94	0.000	
CD	1	7.156	7.156	18.93	0.001	
A^2	1	75.715	75.715	200.28	0.000	
B^2	1	157.851	157.851	417.54	0.000	
C^2	1	8.646	8.646	22.87	0.000	
D^2	1	0.280	0.280	0.74	0.404	
Error	14	5.293	0.378			
Lack of fit	10	3.833	0.383	1.05	0.526	
Pure Error	5	1.460	0.365			

Two and three-dimensional plots indicate the interactions between the variables and biodiesel conversion. It was observed that the optimum conditions acquired from the model equation are methanol to oil molar ratio 8.01:1, catalyst amount 1.08wt%, reaction temperature 70°C, and reaction time 45 min, and the maximum yield was found 95.68% at these optimum conditions

3.3. Fuel properties of pomegranate seed oil biodiesel

The fuel properties of diesel fuel and the pomegranate seed oil biodiesel are produced at optimum conditions are given in Table 4.

Table 4. Fuel Properties of diesel and pomegranate seed biodiesel

Property	Diesel	Biodiesel	EN 14214
Kinematic viscosity at 40°C (cSt)	3.02	4.95	3.5-5.0
Density at 15°C (kg/m^3)	840.8	886	860-900
Flash Point (°C)	58	134	Min 120
Higher heating value (kj/kg)	45.732	40.015	-

3.4. Effects of process variables on biodiesel yield

Figure 2 represents the interaction of molar ratio and catalyst. The reaction temperature and reaction time were kept constant at 60 °C and 75 min, respectively. At a higher molar ratio, the biodiesel yield increased with an increase in catalyst amount. Figure 3 shows the 3D plot between reaction temperature and molar ratio for the fixed catalyst amount of 1.0 wt% and reaction time of 60 min. According to the figure, biodiesel yield is low at low methanol to oil molar ratio and the conversion of biodiesel increases with an increase in reaction temperature. However, it was also observed that there is a decrease in biodiesel yield at higher reaction temperatures. The effects of reaction time and the molar ratio are given in Figure 4. It can be seen from the figure, that biodiesel yield decreases with the increment in reaction temperature. Figure 5 depicts the effect of reaction temperature and catalyst amount and their interference on the process at a 7:1 methanol to oil molar ratio and 75 min reaction time. According to the figure, the biodiesel yield starts to in-creasing as the catalyst amount increased from 0.5wt% to 1.0wt%. Beyond this level, it is observed that there is a drop-in biodiesel yield. The effects of reaction time and catalyst amount on biodiesel yield are given Figure 6. Increases in reaction time did not significantly affect the conversion to biodiesel yield at any catalyst amount. Figure 7 shows the effects of reaction time and reaction temperature on biodiesel yield. Biodiesel yield increases with the increase in the reaction temperature.

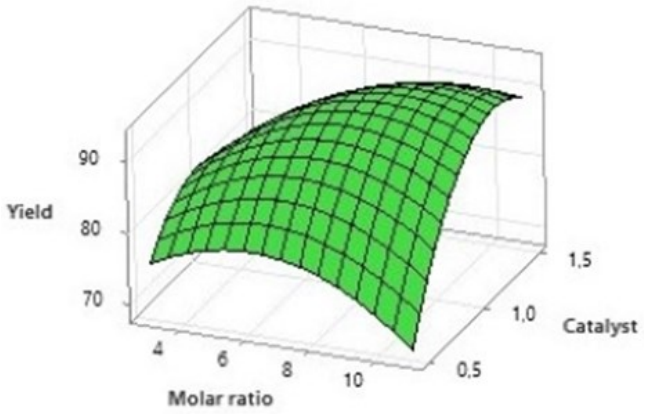

Figure 2. 3D plot between catalyst amount and molar ratio on biodiesel yield

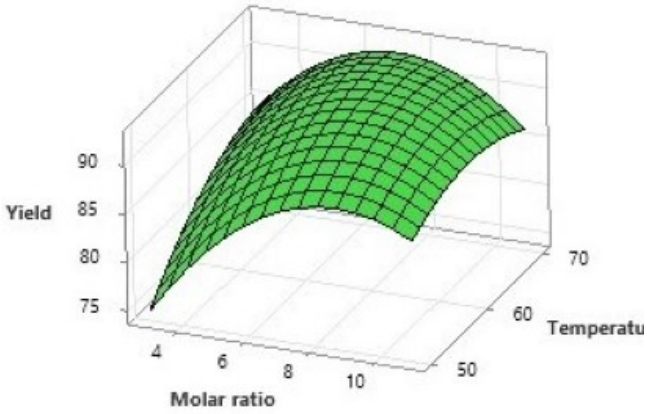

Figure 3. 3D plot between reaction temperature and molar ratio on biodiesel yield

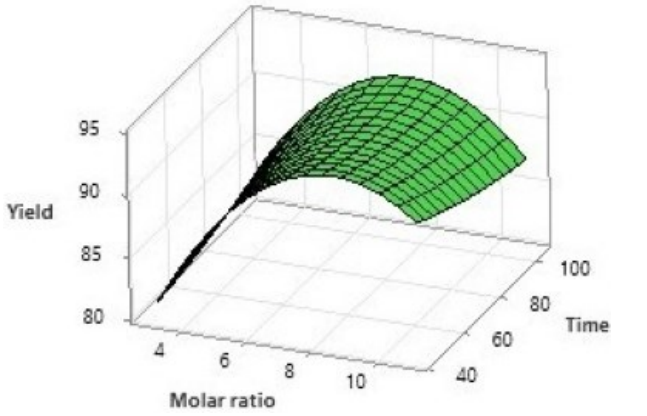

Figure 4. 3D plot between reaction time and molar ratio on biodiesel yield

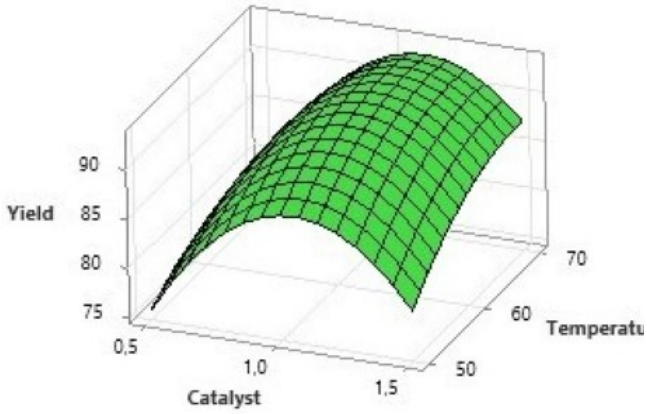

Figure 5. 3D plot between reaction temperature and catalyst amount on biodiesel yield

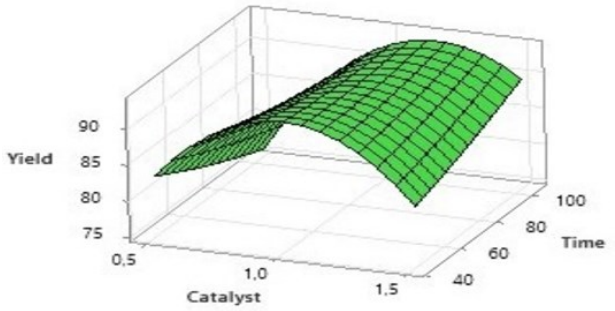

Figure 6. 3D plot between reaction time and catalyst amount on biodiesel yield

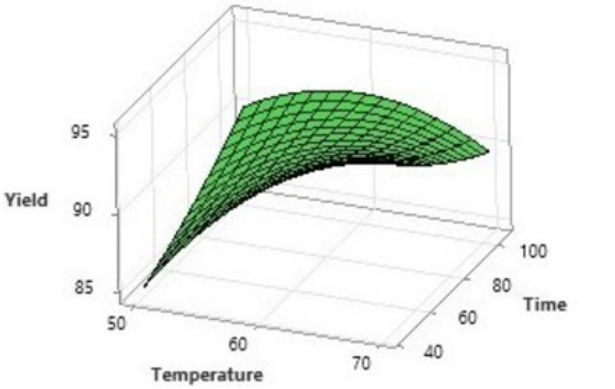

Figure 7. 3D plot between reaction time and reaction temperature on biodiesel yield

3.5. Prediction by artificial neural network

In biodiesel production, Artificial Neural Network is one of the most proper predictive models. The prediction of biodiesel yield from pomegranate oil was acquired by using ANN with Levenberg–Marquardt backpropagation algorithm. The input parameters were catalyst ratio, molar ratio, and reaction temperature and the yield of biodiesel was used as a target. Statistical variables such as R, R^2, RMSE, SEP, and MAE have used in the evaluation of the model. The accuracy of ANN and RSM methods is determined using Eq. (4–8) [17, 29]. In this work, R, R^2, RMSE, MAE, and SEP were selected as performance parameters.

$$R = \frac{\sum_{i=1}^{n}(Y-Y_{ave})(X-X_{ave})}{\sqrt{[\sum_{i=1}^{n}(Y-Y_{ave})^2][\sum_{i=1}^{n}(X-X_{ave})^2}} \quad (4)$$

$$R^2 = \frac{\sum_{i=1}^{n}(X-Y)^2}{\sum_{i=1}^{n}(Y-X_{ave})^2} \quad (5)$$

$$RMSE = \frac{\sum_{i=1}^{n}(X-Y)^2}{n} \quad (6)$$

$$MAE = \sum_{i=1}^{n}\frac{(X-Y)}{n} \quad (7)$$

$$SEP = \frac{RMSE}{Y_{ave}} \quad (8)$$

where X→experimental outputs, Y → predicted outputs, and n → total number of training data. Table 5 shows the operating conditions

performed for the training network of ANN. The coefficient of determination (R^2) indicates the degree of relationship between the used parameters. When the coefficient of determination (R^2) value is between 0.7 to 1.0 considers satisfactory results [17]. For obtaining the best method the iteration of weight parameters was done several times. Figure 8 demonstrated the plots of ANN regression of experimental yield and predicted yield for training, validation, test, and overall. As you can see the Figure 8, The R-value for training data, validation data, test data, and overall data is 0.99361, 0.98686, 0.98975, and 0.98443, respectively, showing that there is a good relationship between the experimental and predicted biodiesel yield. The mean square (MSE) of $6.16141x10^{-1}$ and the coefficient of determination R^2 value of 0.9691 was obtained from the ANN. Network training of ANN is given in Figure 9. The best validation performance of 1.625 was obtained at epoch 4 as given in Figure 10.

Table 5. Operation conditions of ANN

Definations	Descriptions
Software used	MATLAB R2020a
Algorithm	TRAINLM (Lavenberg-Marquardt)
Transfer function	Tansig
Tool used	Nftool
Number of input layers	3
Training data	70%
Validation data	15%
Testing data	15%
Mean square error (MSE)	$1x10^{-5}$ (default)

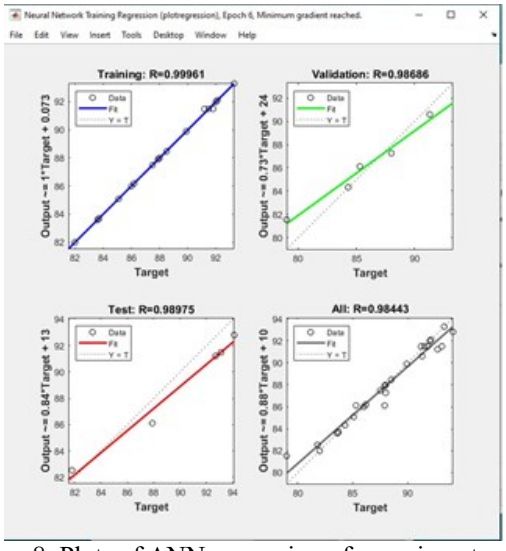

Figure 8. Plots of ANN regression of experiment versus predicted biodiesel yield for training, validation, test and overall

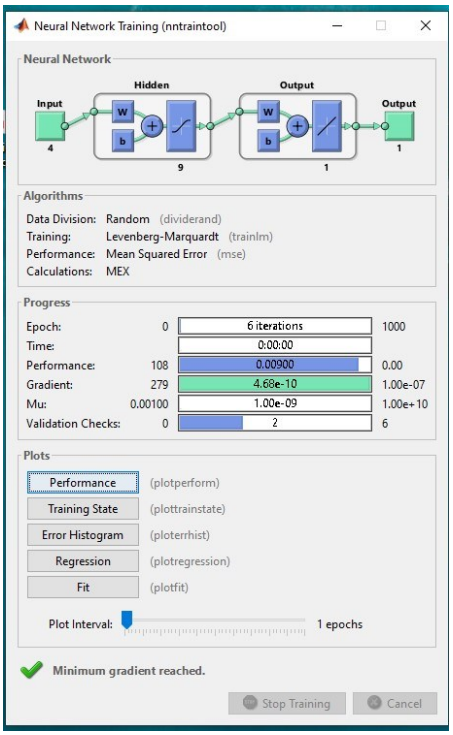

Figure 9. Summary of ANN model

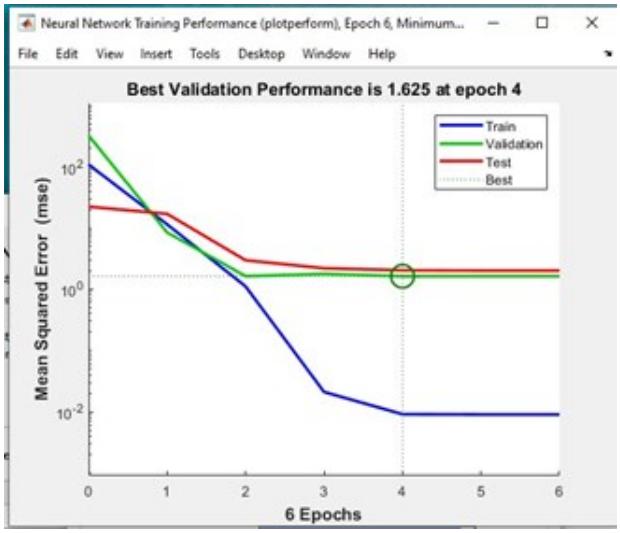

Figure 10. Plot of ANN performance

3.6. Validation of biodiesel production by using RSM method

The optimal level of independent variables was evaluated by solving the regression Equation (1) using the response surface method. Figure 11 is given the obtained optimum values from the RSM model. These are 8.01:1 methanol to oil molar ratio, 1.08 wt% catalyst amount, 70°C reaction temperature, and 45 min reaction time. The predicted optimum biodiesel yield at these optimal conditions was 95.68 %. An experiment was done at optimal process parameters and the obtained yield was %94.90. According to the results, there is a good agreement between the predicted and experimental values.

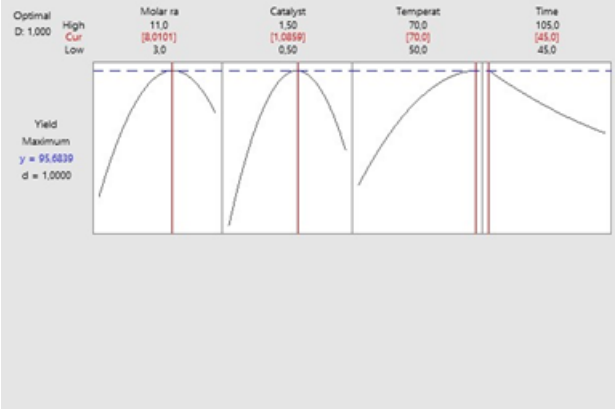

Figure 11. Optimized results of biodiesel production

3.7. Comparison of the prediction capability of RSM and ANN methods

The prediction capabilities of RSM and ANN were determined by comparing the values of R, R^2, RMSE, SEP, and MAE. Table 6 is given the results of the predictive indices for the RSM and ANN methods. The values of R^2 for RSM and ANN were 0.9887 and 0.9691, respectively. According to these results, the RSM method showed higher accuracy and precision than the ANN method and the RSM method is a better mathematical tool to determine the biodiesel yield from pomegranate seed oil compared to the ANN method. The values of RSME, MAE, and SEP acquired from both models demonstrated that the RSM had lower error values than the ANN. Figure 12 shows the experimental biodiesel yield vs predicted biodiesel yield obtained from RSM and ANN methods. Moreover, the figure gives that the RSM predicted values closely aligned to the experimental results than the ANN method. These results displayed that the RSM method has better performance than the ANN method and although these two methods performed satisfactorily.

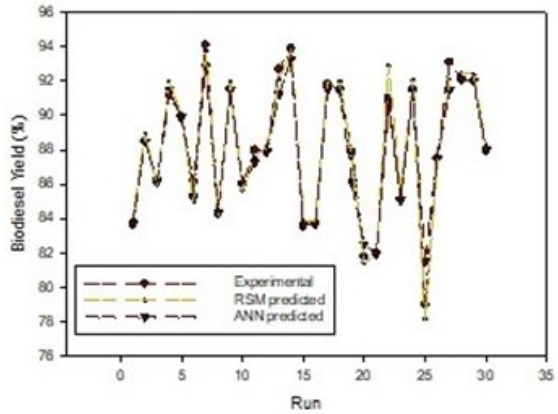

Figure 12. Experimental biodiesel yield vs RSM and ANN predicted

Table 6. Results of statistical benchmark of ANN and RSM methods

Parameters	RSM	ANN
R	0.9943	0.9844
R^2	0.9887	0.9691
RMSE	0.5582	0.7858
SEP	0.00633	0.00893
MAE	0.01474	0.4250

4. Conclusion

In this work, the Central Composite Design of RSM and ANN methods was successfully applied for predicting and optimization of process parameters for biodiesel production from pomegranate seed oil. The obtained results are given below:

- The optimized results of the biodiesel production process were 8.01:1 methanol/oil molar ratio, 1.08 wt% catalyst rate, 70 °C temperature, and 45 min time for conversion of 95.68% pomegranate seed oil to biodiesel was observed.
- Error analysis between predicted and experimental values for the RSM model was determined and the error percentage was found too low.
- The predictive capability of these two methods was determined according to the R, R^2, RMSE, SEP, and MAE. The acquired results were used to compare the effectiveness and superiority of the two method tools. According to the results, the developed RSM has a good predicting capability than the ANN. The R^2 values from RSM and ANN methods were obtained at 0.9887 and 0.9691, respectively and the two methods performed reasonably well.

CRediT authorship contribution statement

Ceyla ÖZGÜR: Writing - original draft, International Journal of Automotive Engineering and Technologies, Investigation, Visualization, Supervision, Conceptualization, Methodology, Software, Formal analysis. Investigation, Supervision, Writing - review & editing. Funding acquisition.

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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