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Modelling of synthesis of waste cooking oil methyl esters by artificial neural network and response surface methodology

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ABSTRACT

This present study was carried out to investigate the application of artificial neural network (ANN) and response surface methodology (RSM) as modelling tools for predicting the waste cooking oil methyl esters (WCOME) yield obtained from alkali-catalysed methanolysis of waste cooking oil (WCO). The impact of process parameters involved was studied by a central composite rotatable design. A comparison of the two developed models for the methanolysis process was carried out based on pertinent statistical parameters. The calculated values of coefficient of determination (R^2) of 0.9950 and the average absolute deviation (AAD) of 0.4930 for the ANN model compared with R^2 of 0.9843 and AAD of 0.9376 for the RSM model demonstrated that the ANN model was more accurate than the RSM model. The actual maximum WCOME yield of 94 wt% was obtained at a reaction temperature of 55°C, a catalyst amount of 1 w/v, a reaction time of 70 min and a methanol-to-oil ratio of 6:1.

Abbreviations/Nomenclature CV: coefficient of variance; FFA: free-fatty acid; *R*: correlation coefficient; R^2: coefficient of determination

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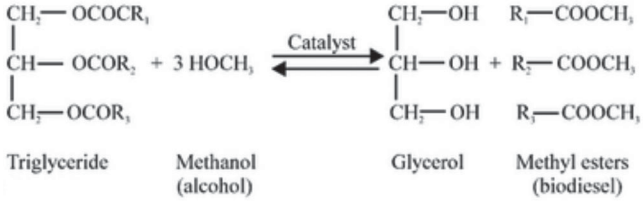
Waste cooking oil; transesterification; biodiesel; modelling; artificial neural network; response surface methodology

1. Introduction

One of the major sources of fuel for generating energy in the world is the fossil fuel, which is the fuel obtained from the remains of dead animals and organic matter. Environmental problems associated with the enormous use of energy derived from fossil fuel have triggered the quest for alternative fuels, especially renewable energy source. Biodiesel, which is a mixture of fatty acid alkyl esters, is a form of renewable energy that is basically derived from different biological sources such as animal fat or vegetable oil (Ramírez-Verduzco, Rodríguez-Rodríguez, and del Rayo Jaramillo-Jacob 2012; Wang et al. 2012). Biodiesel possesses numerous advantages: it is biodegradable, non-toxic, ecofriendly and has low emission profile (McCormick, Ross, and Graboski 1997; Jeong, Yang, and Park 2009; Wang et al. 2012). This has made it a good substitute for petrol–diesel fuel.

Biodiesel cannot fully substitute the conventional diesel fuel because of the high cost of feedstock used in its production (Knothe 2002). It has been reported that 70–80% of the total cost of biodiesel production arises from the cost of its feedstock oil (Wang et al. 2012). To troubleshoot this major challenge, a low and cheap feedstock such as non-food oil, waste cooking oil (WCO), animal fat and cheap alcohol such as methanol could be used for the effective production of biodiesel. WCO is obtained from fresh cooking oil that has been subjected to one form of cooking or the other and such that it is no longer suitable to be used for cooking purposes. It is of low economic value and hence considered waste.

The most cost-effective method of producing biodiesel is via the transesterification process or alcoholysis reaction. The transesterification process involves a reversible equilibrium reaction between a mole of triglyceride and three moles of alcohol to give three moles of fatty acid alkyl esters and a mole of glycerol (Equation (1)). It has been reported that in order to obtain a high yield of biodiesel, the stoichiometry ratio in the reaction could be altered by increasing the number of moles of the alcohol used (Demirbas 2006; Hameed, Lai, and Chin 2009). The chain of reaction in the transesterification reaction involves a step-by-step conversion of triglyceride molecule into diglycerides, monoglycerides and glycerol.



(1)

Biodiesel production using WCO as feedstock through the transesterification process has been reported by several authors (Patil et al. 2010; Baskar and Soumiya 2016; Bala, Misra, and Chidambaram 2017). While some authors have reported modelling of the transesterification process involved using the response

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surface methodology (RSM) (Omar and Amin 2011; Atapour, Kariminia, and Moslehabadi 2014), only a few reports exist on the use of artificial neural network (ANN) (Talebian-Kiakalaieh et al. 2013) or adaptive neuro-fuzzy inference system (ANFIS) (Mostafaei, Javadikia, and Naderloo 2016) for this purpose. RSM is a statistical tool used to develop the empirical model. It is widely used to design an experiment and to perform local optimisation (Chen, Chen, and Lin 2005). However, ANN is the soft computing technique which originated from mathematical neurobiology. ANN has been used as a predictive tool in numerous disciplines due to its capability to use its learning algorithm to create a relationship between input and output for complex nonlinear systems (Alavala 2007; Zobel and Cook 2011). ANN has found a wide application in biochemical processes such as modelling of curdlan production from *Paenibacillus polymyxa* (Rafigh et al. 2014), extraction of artemisinin from *Artemisia annua* (Pilkington, Preston, and Gomes 2014), prediction of process parameters for reduction of acid value of sunflower oil (Rajendra, Jena, and Raheman 2009) and palm kernel oil (Betiku et al. 2016). It has also been applied to the modelling of transesterification processes used for biodiesel production with great success (Betiku and Ajala 2014; Avramović et al. 2015; Sarve, Sonawane, and Varma 2015). Both ANN and RSM have been compared in terms of their prediction capabilities (Betiku and Ajala 2014; Avramović et al. 2015; Sarve, Sonawane, and Varma 2015; Betiku et al. 2016). However, it has been reported that ANN outperformed the RSM in prediction effectiveness (Betiku and Ajala 2014; Avramović et al. 2015; Sarve, Sonawane, and Varma 2015; Betiku et al. 2016).

This present study developed and compared RSM and ANN models that were used to predict waste cooking oil methyl esters (WCOME) yield obtained from the alcoholysis reaction of WCO with methanol in the presence of NaOH as catalyst. The parameters studied using design of experiments (DoE) by RSM were reaction temperature (°C), methanol-to-oil ratio, reaction time (min) and catalyst amount (w/v). The interaction between these parameters was investigated by using the surface plots of RSM. The developed models by RSM and ANN were appraised using statistical parameters such as correlation coefficient (R), coefficient of determination (R^2), AAD, mean average error (MAE), standard error of prediction (SEP), root-mean-square error (RMSE). The ASTM D6751 and EN 14241 standard specifications were used to determine the quality of the biodiesel produced.

2. Experimental

2.1. Materials

The WCO used for this study was obtained from a fast food industry in Ilesa, Osun State, Nigeria. All the chemicals and reagents (sulphuric acid, potassium iodide, calcium chloride, diethyl ether, ethanolic sodium hydroxide, ethanol (95%), methanol and sodium sulphate) used in this study were of analytical grades.

2.2. Methods

2.2.1. DoE and RSM modelling for WCOME production

The central composite rotatable design (CCRD) of RSM was used to model the production of WCOME. A five-level–four-factor design was employed, which produced 30 experimental conditions. The CCRD design includes 16 factorial points, 8 axial points, and 6 central points to provide information regarding the interior of the experimental region, making it possible to evaluate the curvature effect (Jeong, Yang, and Park 2009). The process parameters and their ranges that were considered for the transesterification process of WCO were reaction temperature (40–60°C), catalyst amount (0.9–1.3 w/v), reaction time (40–80 min) and methanol-to-oil ratio (3:1–7:1). Tables 1 and 2 show the coded levels of the independent factors and CCRD for transesterification step, respectively. To fit the coefficient of the quadratic polynomial regression model of the response (dependent variable), multiple regressions were employed. Both Analysis of variance and significance test were applied to determine the quality of the quadratic polynomial response model. Equation (2) describes the fitted model.

Table 1. Factors and their levels for CCRD.

		Coded factors level				
Variable	Unit	−2 (-α)	−1	0	+1	+2 (+α)
Temperature (X_1)	°C	40	45	50	55	60
Catalyst amount (X_2)	w/v	0.9	1.0	1.1	1.2	1.3
Time (X_3)	min	40	50	60	70	80
Methanol-to-oil ratio (X_4)	–	3:1	4:1	5:1	6:1	7:1

$$Y = \alpha_0 + \alpha_1 X_1 + \alpha_2 X_2 + \alpha_3 X_3 + \alpha_4 X_4 + \alpha_{12} X_1 X_2 + \alpha_{13} X_1 X_3 + \alpha_{14} X_1 X_4 + \alpha_{23} X_2 X_3 + \alpha_{24} X_2 X_4 + \alpha_{34} X_3 X_4 + \alpha_{11} X_1^2 + \alpha_{22} X_2^2 + \alpha_{33} X_3^2 + \alpha_{44} X_4^2, \quad (2)$$

where Y is the dependent variable (WCOME yield), α_0 is the intercept value, α_1, α_2, α_3 and α_4 are the first-order coefficients, α_{12}, α_{13}, α_{23} and α_{34} are the interaction coefficients, α_{11}, α_{22}, α_{33} and α_{44} represent the quadratic coefficients and X_1, X_2 and X_3 denote the independent variables.

2.2.2. ANN modelling of the alkali-catalysed methanolysis

The ANN tool can serve as an alternative to the polynomial regression-based modelling tool because it has the ability to model complex and nonlinear relationships (Maran et al. 2013; Rafigh et al. 2014). The accuracy of this modelling tool is achieved by including all the experimental data (Nagata and Chu 2003). The process variables investigated for the ANN model developed were reaction temperature, catalyst amount, reaction time and methanol-to-oil ratio. The ANN model was developed by using MATLAB (The Mathworks, Inc., Natick, MA, USA, ver. 9.0). The ANN training was made more efficient by scaling the inputs and targets data set in the range of −1 to 1. In order to obtain the desired model, the data set containing the output and the inputs was split into three subsets, whereby 70% was employed for training, 15% was used for validation, while the remaining 15% was selected for testing the reliability of the model (Betiku et al. 2016). A two-layer feed forward network also known as multi-layered perceptron with the Levenberg–Marquardt back-propagation algorithm (trainlm) was selected. The choice of the number of hidden neurons was evaluated 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)

Table 2. Actual and predicted acid values by RSM and ANN for the transesterification process.

Run	X_1 (°C)	X_2 (w/v)	X_3 (min)	X_4	Actual WCOME yield(wt. %)	RSM predicted WCOME yield(wt. %)	ANN Predicted WCOME yield(wt. %)
1	40(-2)	1.1(0)	60(0)	5(0)	84.70	84.56	83.74
2	50(0)	1.1(0)	60(0)	5(0)	92.00	91.61	91.75
3	50(0)	1.1(0)	60(0)	5(0)	92.20	91.61	91.75
4	50(0)	1.1(0)	60(0)	5(0)	91.53	91.61	91.75
5	45(−1)	1.2(1)	70(1)	6(1)	88.00	89.10	88.00
6	45(−1)	1.2(1)	50(−1)	6(1)	88.30	89.25	88.29
7	50(0)	1.1(0)	60(0)	7(2)	93.00	92.20	93.16
8	50(0)	1.1(0)	60(0)	5(0)	92.73	91.61	91.75
9	60(2)	1.1(0)	60(0	5(0)	86.76	86.95	86.75
10	50(0)	1.1(0)	80(2)	5(0)	93.00	92.18	92.98
11	45(−1)	1.2(1)	70(1)	4(−1)	88.80	87.41	88.79
12	55(1)	1.2(1)	70(1)	4(−1)	85.62	86.78	85.62
13	50(0)	1.3(2)	60(0)	5(0)	81.00	81.51	81.00
14	45(−1)	1(−1)	50(-1)	6(1)	83.00	81.83	82.99
15	55(1)	1(−1)	70(1)	4(−1)	84.20	83.22	84.19
16	50(0)	1.1(0)	60(0)	5(0)	90.10	91.61	91.75
17	50(0)	1.1(0)	40(-2)	5(0)	79.92	80.78	79.91
18	50(0)	0.9(−2)	60(0)	5(0)	71.00	70.54	70.99
19	45(−1)	1(−1)	70(1)	6(1)	87.30	87.36	88.00
20	45(-1)	1(-1)	50(-1)	4(-1)	66.10	66.63	68.87
21	55(1)	1.2(1)	50(-1)	6(1)	83.15	81.95	82.90
22	50(0)	1.1(0)	60(0)	5(0)	91.10	91.61	91.75
23	50(0)	1.1(0)	60(0)	3(−2)	76.50	77.34	77.39
24	55(1)	1.2(1)	50(-1)	4(−1)	81.00	80.91	79.93
25	55(1)	1(-1)	70(1)	6(1)	94.00	95.01	93.99
26	45(−1)	1.2(1)	50(−1)	4(−1)	87.20	86.17	86.37
27	55(1)	1.2(1)	70(1)	6(1)	87.00	86.44	86.99
28	55(1)	1(−1)	50(-1)	6(1)	83.50	84.86	83.49
29	55(1)	1(−1)	50(-1)	4(−1)	72.80	71.68	72.79
30	45(−1)	1(−1)	70(1)	4(−1)	72.35	73.54	72.35

Note: X_1 – temperature, X_2 – catalyst amount, X_3 – time and X_4 – methanol-to-oil ratio.

is identified. Hence, the topology of the network was based on the number of hidden neurons chosen. In this study, a tangent sigmoid transfer function (tansig) at the hidden layer and a linear transfer function (purelin) at the output layer was used for developing the ANN model. The equations used for developing the ANN model are illustrated in Equations (3)–(5).

$$X = W^{h}V + b^{h}, \tag{3}$$

$$g(t) = \text{tansig}(t) = \frac{1 - e^{-t}}{1 + e^{-t}}, \tag{4}$$

$$Y = W^{out}f(x) + b^{out}, \tag{5}$$

where X is the hidden layer output, V is the vector of network input, W^{h} is the hidden layer weight, b^{h} is the hidden layer bias, $g(t)$ is the activation function, Y is the network output, W^{out} is the output layer weight and b^{out} is the output layer bias.

2.2.3. Statistical analysis of the developed models

Statistical parameters such as R, R^2, RMSE, MAE, SEP and AAD were employed assessing the predictive capability of the developed models. Equations (6)–(11) were employed for the statistical indices (Table 3). The results obtained were used to compare the effectiveness and superiority of the two modelling tools.

Table 3. Statistical parameters and their relations.

Index name	Equation	Number
Correlation coefficient	$R = \frac{\sum_{i=1}^{n}(y_{p,i} - y_{p,ave}).(y_{a,i} - y_{a,ave})}{\sqrt{\left[\sum_{i=1}^{n}(y_{p,i} - y_{p,ave})^2\right]\left[\sum_{i=1}^{n}(y_{a,i} - y_{a,ave})^2\right]}}$	6
Coefficient of determination	$R^2 = \frac{\sum_{i=1}^{n}(y_{a,i} - y_{p,i})^2}{\sum_{i=1}^{n}(y_{p,i} - y_{a,ave})^2}$	7
Root mean square error	$\text{RMSE} = \sqrt{\frac{\sum_{i=1}^{n}(y_{a,i} - y_{p,i})^2}{n}}$	8
MAE	$\text{MAE} = \frac{\sum_{i=1}^{n}\lvert(y_{a,i} - y_{p,i})\rvert}{n}$	9
SEP	$\text{SEP} = \frac{\text{RMSE}}{y_{a,ave}} \times 100$	10
AAD	$\text{AAD} = \frac{100}{n}\sum_{i=1}^{n}\left(\frac{\lvert y_{a,i} - y_{p,i}\rvert}{(y_{a,i})}\right)$	11

Note: Where n is the number of data sets, $y_{a,i}$ and $y_{p,i}$ are actual and predicted output values of the ith set, respectively, $y_{a,ave}$ is the average actual output values and $y_{p,ave}$ is the average predicted values.

2.2.4. Alkali-catalysed methanolysis of WCO

The alkali-catalysed methanolysis was carried out for WCOME production due to the low acid value of the WCO (Betiku and Adepoju 2013). NaOH pellets of known weight were carefully dissolved in a known volume of methanol. The mixture was then transferred into the WCO in the reactor and the reaction was monitored based on the design variables in Table 2. After the reaction was completed, the resulting product was transferred to a separating funnel for WCOME and glycerol separation under gravity. Glycerol was tapped off and the WCOME left was

washed with distilled water to remove residual catalyst, glycerol, methanol and soap. The washed WCOME was further dried over heated $CaCl_2$ powder. The WCOME yield was determined gravimetrically as described in Equation (12).

$$\text{WCOME (wt. \%)} = \frac{\text{weight of WCOME produced}}{\text{weight of WCO used}} \times 100. \quad (12)$$

2.2.5. Quality characterization of WCO and WCOME

To evaluate the physical and chemical properties of the WCO and WCOME, the AOAC methods were used. The properties determined include acid value, FFA, kinematic viscosity, specific gravity, peroxide value and iodine value. Cetane number, aniline point, diesel index and API were determined by the methods earlier reported by Haldar, Ghosh, and Nag (2009), while the higher heating value was determined using the method reported by Demirbas (1998).

3. Results and discussion

3.1. Quality characterisation of WCO and WCOME

The physicochemical properties of WCO and WCOME produced are shown in Table 4. The properties of the WCO showed that it is a good candidate for the synthesis of biodiesel. Although the iodine value and cetane number of the WCO were within the specified limits for biodiesel by ASTM D 6751 and EN 14214, its kinematic viscosity was too high to be used directly in an internal combustion engine. The WCO had an acid value of 1.06 mg KOH/g oil, which corresponds to %FFA of 0.53, which suggests one-step transesterification to convert it into biodiesel. The fuel properties of WCOME produced are also summarised in Table 4. The properties of the WCOME agreed satisfactorily with biodiesel standard specifications (ASTM D 6751 and EN 14214).

3.2. Process modelling of WCOME production by RSM

The results of the methanolysis of the WCO to WCOME are presented in Table 5. The WCOME yield varied from 66.10 to 94 wt.%. The minimum WCOME yield was obtained at a reaction temperature of 45°C, a catalyst amount of 1 w/v, a reaction time of 50 min and a methanol-to-oil ratio of 4:1. The maximum WCOME yield was obtained at a reaction temperature of 55°C, a catalyst amount of 1 w/v, a reaction time of 70 min and a methanol-to-oil ratio of 6:1. Equation (13) shows the results obtained from the RSM modelling when the multiple regression was applied to the experimental data. The equation describes the actual relationship between the independent variables (process parameters) and response.

$$\begin{aligned} Y(\text{wt.\%}) = {} & -1197.41 + 10.77X_1 + 2.39X_2 + 61.31X_3 - 5.16X_4 \\ & + 0.023X_1X_2 - 0.10X_1X_3 - 1.42X_1X_4 - 30.33X_2X_3 - 0.03X_2X_4 \\ & - 0.06X_3X_4 - 0.059X_1^2 - 389.67X_2^2 - 0.013X_3^2 - 1.71X_4^2, \end{aligned} \quad (13)$$

Table 4. Physicochemical properties of WCO and WCOME.

Parameter	WCO	WCOME	ASTM D6751	EN 14214
Physical properties				
Physical state at 28°C	Liquid/ dark-brown	Liquid/ light brown		
Moisture content (%)	0.086	0.012	0.03 max	0.05 max
Specific gravity	0.962	0.852	0.87-0.90	0.85
Kinematic viscosity (cSt) at 40°C	32.20	5.87	1.9-6.0	3.5-5.0
Chemical properties				
%FFA (as oleic acid)	0.53	0.16	–	
Acid value (mg KOH/g oil)	1.06	0.32	0.5 max	0.5 max
Iodine value (g I_2/100 g oil)	67.6	74.67	-	120 max
Saponification value (mg KOH/g oil)	187.30	58.10	–	–
Higher heating value (MJ/kg)	40.74	45.93	–	–
Other fuel properties				
Cetane number	60.23	58.64	47 min	51 min
Flash point (°C)	–	176	100	–
Cloud point (°C)	–	12	–	–
Pour point (°C)	–	9	–	–

Table 5. Significance tests and ANOVA results.

Source	Coefficient estimate	Sum of square	df	Mean square	*F*-value	*p*-value
Model		1505.87	14	107.56	67.49	< 0.0001
Linear						
Intercept	−1197.41					
Temperature (X_1)	10.77	8.57	1	8.57	5.38	0.0349
Catalyst amount (X_2)	2.39	180.51	1	180.51	113.26	< 0.0001
Time (X_3)	61.31	194.83	1	194.83	122.24	< 0.0001
Methanol-to-oil ratio (X_4)	−5.16	331.38	1	331.38	207.92	< 0.0001
Interaction						
X_1X_2	0.023	106.50	1	106.50	66.82	< 0.0001
X_1X_3	−0.10	21.44	1	21.44	13.45	0.0023
X_1X_4	−1.42	4.12	1	4.12	2.59	0.1287
X_2X_3	−30.33	32.15	1	32.15	20.17	0.0004
X_2X_4	−0.03	147.14	1	147.14	92.32	< 0.0001
X_3X_4	−0.06	1.90	1	1.90	1.19	0.2916
Quadratic						
X_1^2	−0.059	58.80	1	58.80	36.89	< 0.0001
X_2^2	−389.67	416.48	1	416.48	261.31	< 0.0001
X_3^2	−0.013	45.06	1	45.06	28.27	< 0.0001
X_4^2	−1.71	80.13	1	80.13	50.27	< 0.0001
Residual		23.91	15	1.59		
Lack of fit		19.61	10	1.96	2.28	0.1880
$R^2 = 0.9844$ adjusted $R^2 = 0.9698$; predicted $R^2 = 0.9221$; C.V. = 1.49%						

Note: df – degree of freedom, C.V. – coefficient of variance.

where Y denotes the WCOME yield, X_1 is the temperature, X_2 is the catalyst amount, X_3 is the time and X_4 is the methanol-to-oil ratio. Equation (13) shows the terms that have either positive or negative impact on the WCOME yield. From the equation, all the linear terms and the interaction between the temperature and reaction time have a positive influence on the WCOME yield, while all other terms have a negative influence on the WCOME yield. The accuracy and the effectiveness of the model were subjected to ANOVA and the results are displayed in Table 5. The values of 67.49 and 0.0001 obtained for F-value and p-value, respectively, indicate that the model was significant (Stamenković et al. 2013) at the 95% confidence level ($p < .05$) (Table 4). All the terms are significant except the interaction between the temperature and methanol-to-oil ratio (X_1X_4) and the interaction between time and the methanol-to-oil ratio(X_3X_4). The model accuracy was determined by calculating the R^2 and adjusted R^2. The value of 0.9844 obtained from the model implies that more than 98% of the whole data set was consistent with the observed values. The adjusted R^2, which was used to assessed the alignment of R^2 for the number of terms and sample size involved in the model, was 0.9698, indicating good fitness of the model. The predicted R^2 of 0.9221 is in reasonable agreement with the adjusted R^2 of 0.9698. The lower value of CV (1.49%) shows that variation between the observed and predicted values was low (Betiku and Ajala 2014). The measure of signal-to-noise ratio is determined by

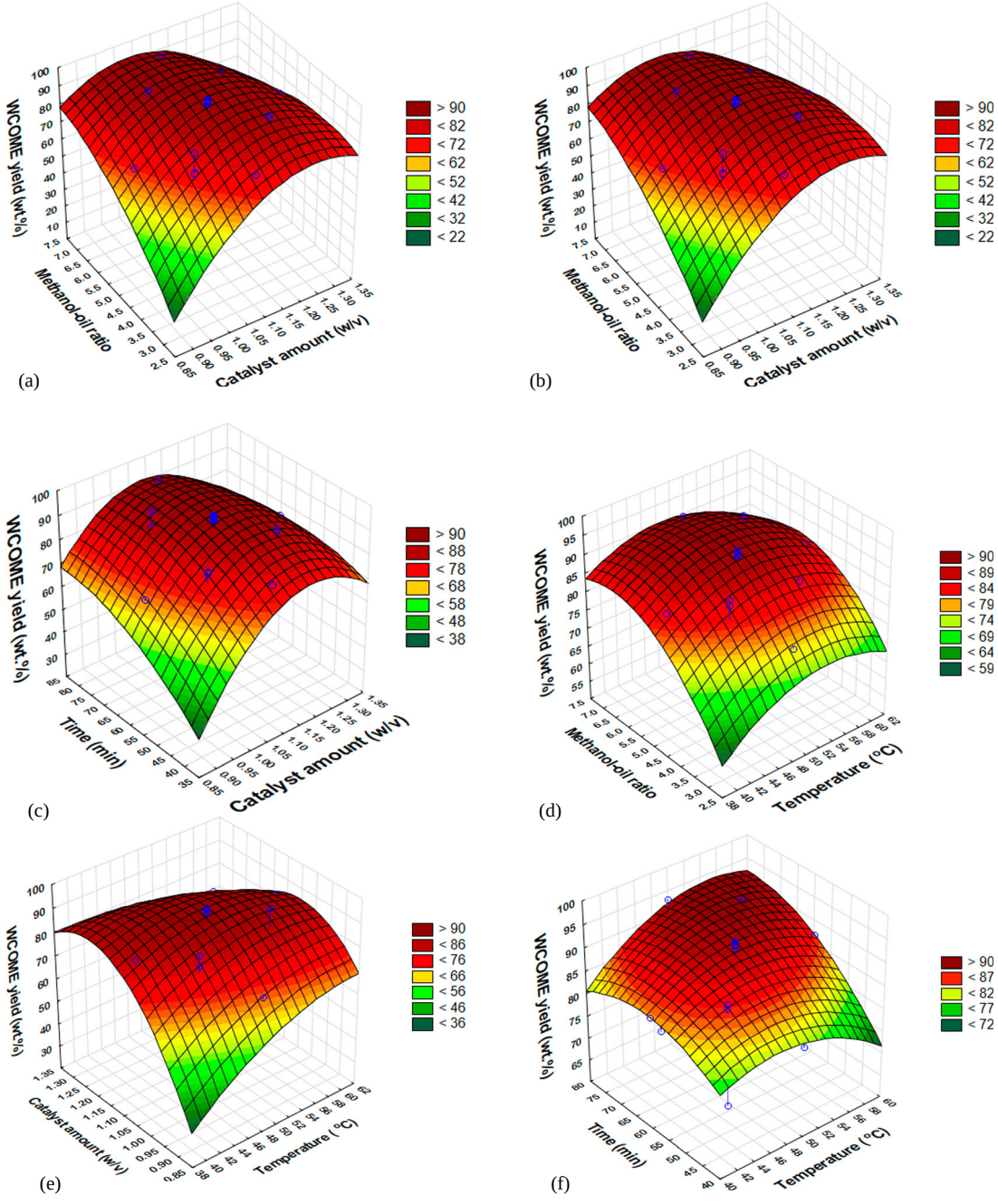

Figure 1. Response surface plot.

the adequate precision and a value greater than 4 is required (Noordin et al. 2004). The value obtained in this study was 31.7, which indicates an adequate signal. The 'lack of fit *F*-value' of 2.28 implies an insignificant lack of fit relative to the pure error. The insignificant lack of fit suggests that the model is significant. The prediction of WCOME yields by the ANN model is shown in Table 2. The WCOME yield ranged between 66.63 and 95.01 wt.%. The maximum WCOME yield of 95.01 wt.% predicted by the RSM model was obtained at a reaction temperature of 55°C, a catalyst amount of 1 w/v, a reaction time of 70 min and a methanol-to-oil ratio of 6:1.

3.2.1. Process parameters interactions

Statistica 12 software (StatSoft Inc., Tulsa, OK, USA) was further used to generate response surface plots in order to investigate the interactions between the process variables investigated in this study. Figure 1(a) shows the surface plot for the WCOME yield as a function of catalyst amount and methanol-to-oil ratio, when the temperature and reaction time are kept constant. The figure shows that as the methanol-to-oil ratio increases from 3:1 to 7:1, there is an increase in the WCOME yield. This is because an increase in the methanol-to-oil ratio favours the WCOME yield. It has been reported that a sufficient methanol-to-oil ratio increases the biodiesel yield (Demirbas 2008; Hameed, Lai, and Chin 2009; Olutoye, Lee, and Hameed 2011; Betiku, Akintunde, and Ojumu 2016). It can be seen from Figure 1(a) that beyond 7:1, there was no significant increase in the WCOME yield. Although an excess methanol-to-oil ratio quickens the reaction, beyond certain required ratio, it may result in esters recovery complication (Goff et al. 2004; Rashid et al. 2008). Hence, an optimum methanol-to-oil ratio should be selected on the overall economics and equilibrium WCOME yield (Sarve, Sonawane, and Varma 2015). Figure 1(a) also reveals that the catalyst had a significant impact on the WCOME yield. The yield increases from 71 to 94 wt.% as the catalyst amount was increased from 0.9 to 1.1 w/v, but beyond certain amount of catalyst, the yield decreases. This may be attributed to the saturation of catalyst particle inside the reaction medium and hence lower the yield of WCOME (Sarve, Sonawane, and Varma 2015).

Figure 1(b) represents the surface plot showing the interaction between the methanol-to-oil ratio and reaction time with the WCOME yield. From the plot, the WCOME yield increases as both methanol-to-oil ratio and time increase. As the reaction time increases from 40 to 75 min, there is a sharp increase in the WCOME yield. This is because sufficient time is needed for the reaction to attain equilibrium since transesterification reaction is a reversible process (Ma and Hanna 1999). Beyond 75 min, the WCOME yield decreases. It has been reported that a longer reaction time enhanced the hydrolysis of esters and hence may result in the loss of esters and also cause soap production due to fatty acids formation (Eevera, Rajendran, and Saradha 2009). Figure 1(b) reveals that the WCOME yield attained $>$ 90 wt.% at a reaction time of 75 min and a catalyst amount of 1 w/v.

Figure 1(c) depicts the interaction between the catalyst amount and the reaction time. The surface plot reveals that the reaction time and the catalyst amount have a strong influence on the WCOME yield, but the reaction time had more influence than the catalyst amount. This is because in order to overcome the slow reaction rate, sufficient time is needed to foster the dispersion of methanol in the reaction medium (Leung, Wu, and Leung 2010). The maximum WCOME yield of 94 wt.% was attained at a reaction time of 70 min and 1 w/v catalyst amount. The plot also shows that increasing the catalyst amount beyond certain level leads to a decrease in the WCOME yield. The decrease in WCOME can be attributed to the saturation of catalyst particle.

Figure 1(d) shows the mutual interaction between the temperature and methanol-to-oil ratio on the WCOME yield. The increase in the WCOME yield is observed when the temperature increases from 40°C to 45°C and methanol-to-oil ratio increases from 3:1 to 7:1. The plot reveals that the WCOME yield of $>$ 90 wt.% was achieved at a temperature $>$ 50°C and at a methanol-to-oil ratio $>$ 5:1. The decrease in WCOME is observed when the temperature is high and the methanol-to-oil ratio is low. The influence of catalyst amount and reaction temperature on the WCOME yield is illustrated in Figure 1(e). The plot reveals that both catalyst amount and temperature had a significant impact on the WCOME yield, but that the catalyst amount had more influence than the reaction temperature. The effect of catalyst amount was more pronounced than the reaction temperature at a lower catalyst amount. The maximum yield (94 wt.%) was attained at a catalyst amount of 1 w/v and a reaction temperature of 55°C. The shape of the surface plot in Figure 1(f) reveals the significance of the interaction between the reaction time and reaction temperature. The effect of reaction time was more obvious at low reaction temperatures. A higher yield of WCOME is observed at higher reaction times because sufficient time is needed for the reaction media to attain equilibrium, and hence favours the production of WCOME.

3.3. Process modelling of WCOME production by ANN

The prediction of the WCOME yield was obtained by ANN with Levenberg–Marquardt backpropagation algorithm consisting of an input layer with four input process variables (temperature, catalyst amount, reaction time and methanol-to-oil ratio) and one output layer with single output variable (WCOME yield). The best combination of the transfer function for the input to hidden layer and hidden to output layer was determined by studying nine different pairs of these transfer functions (Table 6). Due to the lower and higher values obtained for MSE (0.5406) and R^2 (0.9901), respectively (Table 6), the hyperbolic tangent transfer function (tansig) was chosen for the input to hidden

Table 6. Comparison of different transfer functions of Levenberg–Marquardt backpropagation with 10 neurons in hidden layers.

Transfer function					
Input-hidden	Hidden-output	Number of iteration	MSE	R^2	Linear equation
Logsig	Logsig	179	41.45	0.3396	$y = 0.5099x + 43.47$
Tansig	Tansig	124	18.26	0.6869	$y = 0.7895x + 16.65$
Purelin	Purelin	158	28.16	0.4543	$y = 0.4779x + 44.859$
Logsig	Tansig	102	4.37	0.9342	$y = 0.8966x + 9.7535$
Logsig	Purelin	65	7.59	0.8579	$y = 0.8196x + 15.831$
Tansig	Purelin	38	0.5406	0.9901	$y = 0.9657x + 2.9811$
Tansig	Logsig	159	15.91	0.7758	$y = 0.5812x + 36.977$
Purelin	Tansig	187	19.99	0.6345	$y = 0.6778x + 28.46$
Purelin	Logsig	186	33.92	0.4114	$y = 0.3174x + 59.652$

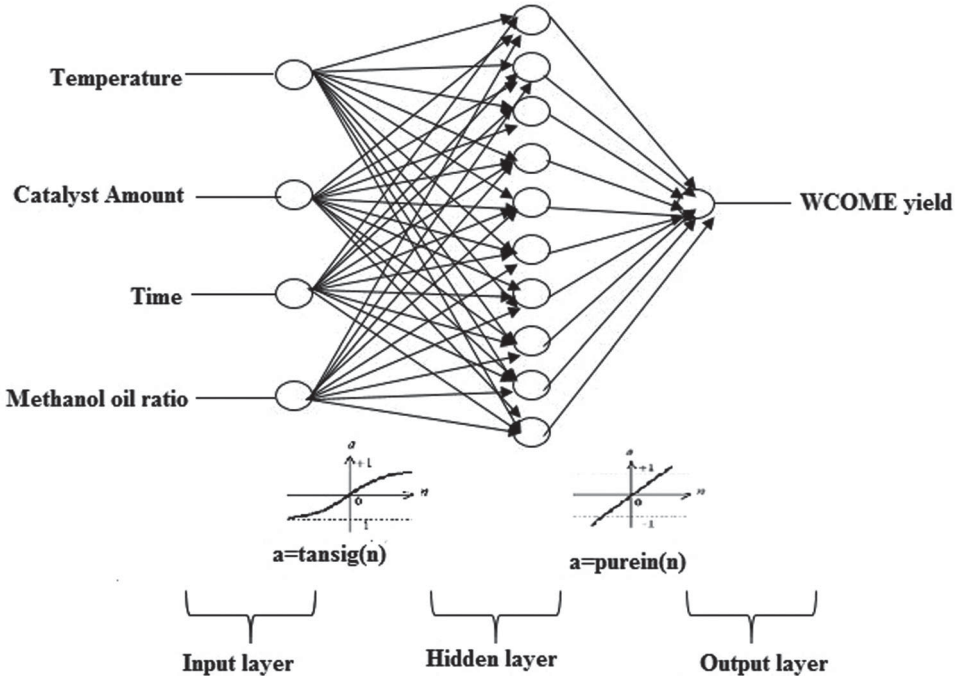

Figure 2. Optimal ANN architecture used for the prediction of the WCOME yield.

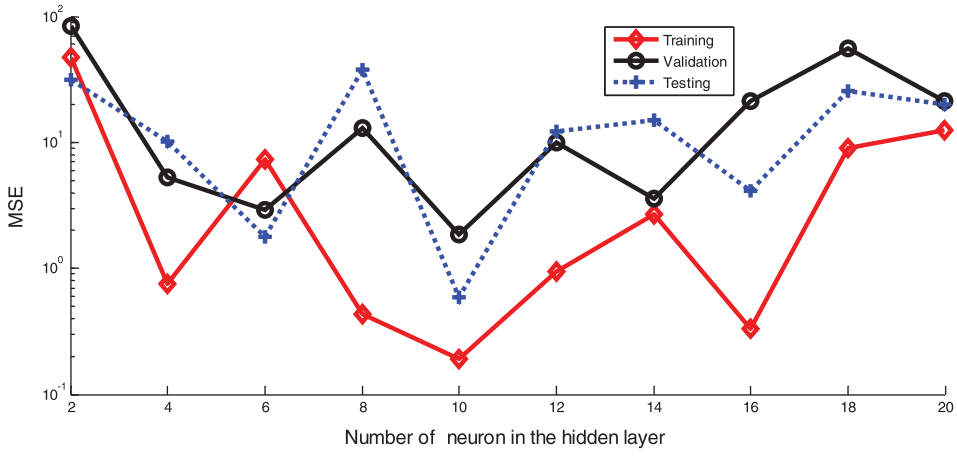

Figure 3. Optimal number of hidden neurons.

layer mapping, while purelin was chosen for the hidden to output layer mapping. Hence, tansig-purelin transfer function was considered for the developed ANN model. The optimal number of hidden neurons was carefully selected by trial-and-error procedure. For this purpose, 2–20 hidden neurons were investigated for constructing the network architecture. Each one of this hidden neuron was then trained several times and further appraised to produce the least output error based on MSE and the highest value of *R*. The best topology chosen for the estimation of WCOME yield was 3-10-1. Figure 2 shows the ANN optimum architecture for the obtained topology for the estimation of the WCOME yield. Figure 3 illustrates the variations in the MSE value for training, validation and testing of the developed ANN model. It is obvious from the plot that the minimum MSE obtained for training, validation and testing of the data sets are at hidden neuron 10. Table 7 shows the MSE and *R* of the hidden neuron 10 for training, validation and testing of the data sets. Correlation coefficient plots for the training, testing, validation and whole data sets are presented in Figure 4. The prediction of WCOME yields by the ANN model is shown in Table 2. The WCOME yield ranged between 68.87 and 93.99 wt.%. The maximum yield predicted by the ANN model was under the same condition as predicted by the RSM model, i.e. at a reaction temperature of 55°C, a catalyst amount of 1 w/v, a reaction time of 70 min and a methanol-to-oil ratio of 6:1.

Table 7. Predicted model *R* and MSE analysis for hidden neuron 10.

	Sample	*R*	MSE
Training	20	0.9956	0.1974
Validation	5	0.9866	1.8676
Testing	5	0.9769	0.5879

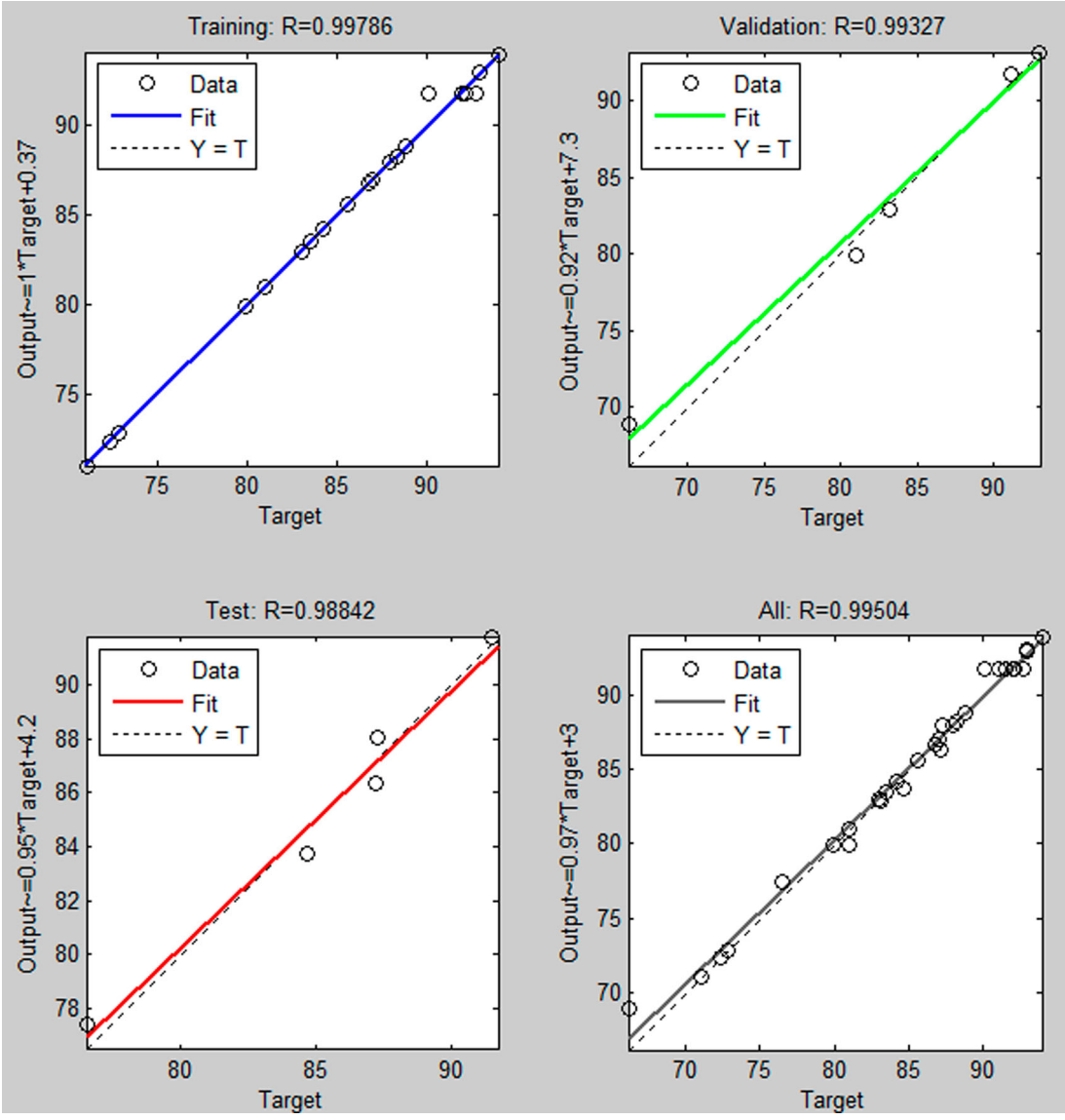

Figure 4. Regression plots of predicted values against actual for the ANN model.

Table 8. Evaluation of predictive of ANN and RSM models.

Parameter	RSM	ANN
R	0.992140	0.995035
R^2	0.984342	0.990095
RMSE	0.893551	0.735273
MAE	0.790667	0.398667
SEP	1.052119	0.865754
AAD	0.937611	0.493019

3.4. Comparison of the predictive capability of RSM and ANN models

The prediction capabilities of RSM and ANN models were evaluated and compared based on statistical parameters: R, R^2, RMSE, MAE, SEP and AAD. The results obtained are shown in Table 8. These revealed that the ANN model outperformed the RSM model, although both models performed satisfactorily. The values of R (0.9950), R^2 (0.9901) obtained for the ANN model demonstrated higher precision and accuracy compared with R (0.992140) and R^2 (0.984342) obtained for the RSM model. The values of RSME, MAE, and SEP obtained for both models showed that the ANN model had lower error values than the RSM model. Furthermore, Figure 5 shows the plots of the actual and predicted values of both ANN and RSM models against the experimental runs. The figure shows that ANN predicted values closely aligned to the actual values than the RSM predicted values. The superiority of ANN over RSM observed in this study is in agreement with previous reports (Avramović et al. 2015; Betiku and Ajala 2014; Betiku et al. 2016; Sarve, Sonawane, and Varma 2015). Betiku et al. (2016) evaluated the performance of RSM, ANN and ANFIS in the prediction of acid value of palm kernel oil and proved that ANN was the best of the three modelling tools. Betiku and Ajala (2014) demonstrated the superior power of ANN over RSM in predicting the biodiesel yield from methanolysis of yellow oleander oil with plantain peels as the heterogeneous base catalyst. Avramović et al. (2015) showed that ANN is more consistent in capturing the nonlinear relationship between the process parameters and biodiesel yield obtained from ethanolysis of sunflower oil with calcium oxide as the catalyst, while Sarve et al. (2015) showed in their studies of ultrasound-assisted

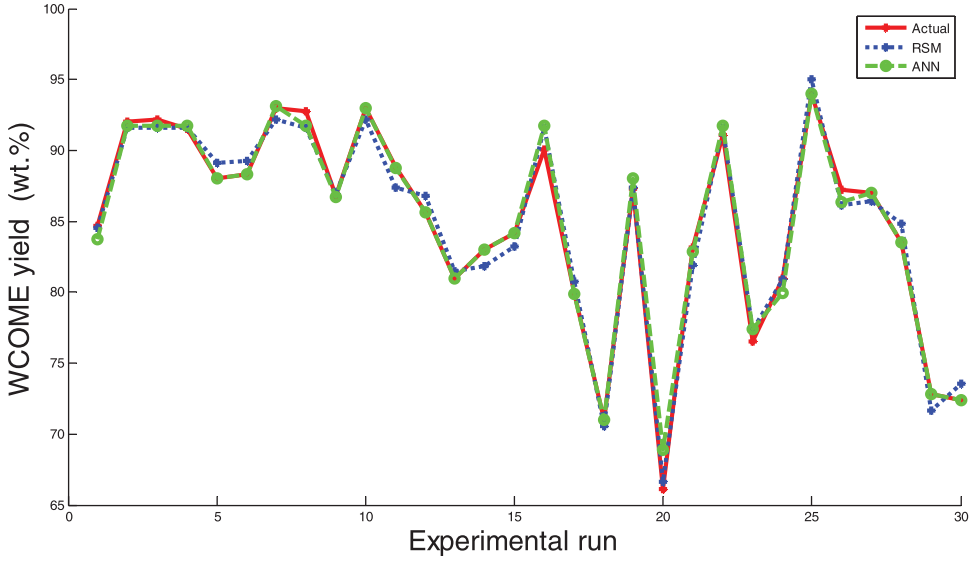

Figure 5. Experimental runs as a function of predicted and actual values.

biodiesel production from sesame oil with barium hydroxide as a catalyst that ANN was a better predicting tool than RSM.

4. Conclusions

In this study, RSM and ANN models were developed for predicting the conversion of WCO into WCOME using a homogeneous base catalyst. DoE of RSM was employed to generate 30 experimental conditions via the use of a four-factor–five-level CCRD. The predictive capability of RSM and ANN was evaluated and compared using statistical parameters viz: R, R^2, RMSE, MAE, SEP and AAD. The results of these statistical parameters confirmed that the ANN model was more accurate and precise than the RSM model, although both models performed reasonable well. The properties of the WCOME produced satisfied the biodiesel standard specifications (ASTMD 6751 and EN 14214).

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No potential conflict of interest was reported by the author(s).

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