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## Full Length Article

## Optimization of the operating parameters for the extractive synthesis of biolubricant from sesame seed oil via response surface methodology

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## ABSTRACT

This study examined the optimization of the transesterification reaction of sesame methyl esters (SMEs) with trimethylolpropane (TMP) to synthesise sesame biolubricant (SBL). Response surface methodology (RSM) using central composite design (CCD) was employed for the experimental design. Effects of temperature, mole ratio and reaction time on the yield of SBL were evaluated. The predicted yield after process optimization was found to agree satisfactorily with the experimental value. The optimum conditions were obtained at temperature of 150 °C, mole ratio of 6:1 and time of 197.26 min for 80.02% biolubricant yield. Mole ratio and reaction time were found to be the most significant variables. SBL characteristics complied with ISO VG 22 standard and holds high potential as base stock for biolubricant formulation.

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## 1. Introduction

Lubricants used globally are commonly made from petroleum, coals or natural gases [1,2]. Due to their high utilization, there is the need to search for alternative sources for producing lubricants [3]. As a result of the long-term pollution effects associated with mineral oil-based lubricants on the environment, there is need for cheap and renewable feedstock for the production of biodegradable lubricant.

There are many alternatives to petroleum based lubricant, such as synthetic or animal fat lubricants. However, lubricants derived from vegetable oil have received greater attention due to their favourable and acceptable physical properties. Among the advantages of biobased lubricants is its high lubricity as well as much lower coefficient of friction when compared to petrobased lubricants. Furthermore, biobased lubricants have high flash points, which makes them effective in high temperature environment to impede evaporation or dissipation [4–6].

They also have relatively stable viscosity indexes, so that they are useful over a large range of temperatures. In addition, biobased lubricants are generally derived from vegetable oils, and its processing involves clean and pollution free method as well as being

renewable. Finally, the non-toxic and biodegradable nature of bio-based lubricant ensures easy disposal to the environment, unlike petroleum based lubricant. These properties make biobased lubricants an attractive alternative to petroleum based lubricants [7].

Despite a number of useful properties of biobased lubricant, there are few draw backs associated with them, such as high composition of organic matters that causes easy oxidation of the lubricant. This rapid oxidation leads to frequent changing of the lubricant, there by modifying its properties adversely. Secondly, the initial cost per volume of a biobased lubricant is generally two or three times more than that of a petrobased lubricant. Thirdly, biobased lubricants can only be applied over a moderate range of temperatures as they have high pour points in cold and low thermal stability in heat. Finally, specialized crops (rather than food by products) are the raw materials for higher quality lubricants, thus diminishing sources that would otherwise be applied for food production [8,9].

In general, biolubricant is produced by transesterification reaction of fatty acid methyl esters (FAME) and polyhydric alcohol. Transesterification reaction may be carried out with an acid catalyst, a base catalyst or an enzyme. There has been a lot of work reported on the transesterification reaction of vegetable oil methyl esters with trimethylolpropane (TMP) as the polyol [10–12]. However, reported model compound studies in this area of research are very few.

In this study, we present a novel synthetic approach for transesterification reaction to produce a high yield of biolubricant. It is

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typically investigated by optimizing the following reaction variables: mole ratio, reaction temperature, and reaction time involved in the process. Both the effects of the variables and their reciprocal interactions have been evaluated using response surface methodology (RSM).

Response surface methodology is a collection of mathematical and statistical techniques used for the modelling and analysis of problems in which a response of interest is influenced by several variables. The objective is to optimize the response (output variable) which is influenced by several independent variables (input variables or factors) [13]. Different levels or values of the operating conditions comprise the factors in each experiment [14]. Some may be categorical (e.g., the supplier of raw material) and others may be quantitative (feed rates, temperatures, time, etc.). In practice, categorical variables must be handled separately by comparing our best operating conditions with respect to the quantitative variables across different combinations of the categorical ones. The fundamental methods for quantitative variables involve fitting first-order (linear) or second-order (quadratic) functions of the predictors to one or more response variables, and then examining the characteristics of the fitted surface to decide what action is appropriate [14].

## 2. Materials and methods

### 2.1. Materials

*Sesame* samples were collected from Idah, Kogi State, Nigeria. Chaffs were separated from the oilseeds by winnowing. All the chemicals and reagents used for this work were of analytical grade.

### 2.2. Extraction of oil from sesame seed

Dried sesame seeds were crushed and tied in a white piece of cloth. This was later soaked in hexane in a tightly sealed bucket for 3 days before collecting the extract through filtration. The cloth containing the crushed sesame seeds was further rinsed with fresh hexane to extract more oil. Hexane contained in the extracted sesame oil was removed by distillation and the crude oil extracts was collected in a beaker [15].

### 2.3. Synthesis of sesame methyl ester

Sesame oil extracted from sesame seed was transesterified to form sesame methyl ester (SME). In this method, a mixture of 300 g of sesame oil, 100 g methanol and 1% wt/wt orthophosphoric acid catalyst was poured into continuously stirred reactor equipped with a water-cooled reflux condenser and heated up to 65 °C for 90 min. The mixture was dosed with 0.2 M of sodium trioxocarbonate IV, which on neutralizing the acid catalyst, stopped the reaction. The neutralized mixture was later transferred to a separating funnel and subsequently allowed to stand overnight to ensure complete separation of methyl esters and glycerol phases. Glycerol phase (bottom phase) was emptied into a clean container and then allowed to stand. The obtained sesame methyl esters were heated at 65 °C to remove methanol. Entrained catalyst in the SME was removed by successive rinses with hot and distilled water. Finally, water present in the SME was eliminated by oven-heating at 70 °C [15].

### 2.4. Synthesis of sesame biolubricant

This was as described by Surapoj et al. [16] with modifications. A batch transesterification at predetermined conditions: TMP:SME (3:1, 4:1, 5:1, 6:1 and 7:1), temperature (80, 100, 120, 140 and

160 °C) and time (1, 2, 3, 4 and 5 h) at 110 °C was carried out in a 50 mL three-necked round bottom flask equipped with a water-cooled reflux condenser, a thermometer, kipp's apparatus and a magnetic stirrer. The mixture contained in the flask was stirred at 1000 rpm, 110 °C for 15 min under CO<sub>2</sub> flow. 110 °C was maintained to evolve moisture from the TMP. A required amount of catalyst was then introduced in the reaction mixture, which was allowed to react for 5 h. At the end of the reaction, the product mixture was brought to room temperature and filtered to separate the solid catalyst from the liquid mixture (SBL). The filtered sesame bio-based stock was analysed using the gas chromatography (GC) to determine the product composition. Pour point, viscosities, flash point, total base number (TBN) and viscosity index were also determined by appropriate analysis [15].

### 2.5. Analysis of transesterification product

Identification of functional groups present in SBL was determined using Fourier Transform Infrared (FTIR) Resonance. Samples were collected after 1, 2, 3, 4 and 5 h and analysed for SME, monoester (ME), diesters (DE), triesters (TE) and TMP by gas chromatography. The yield of each product was determined from the GC chromatogram calibrated against the known samples according to the procedure described by Yunus et al. [17].

### 2.6. Lubricating characteristics

The following lubricating characteristics were determined based on the corresponding referred ASTM procedures: Pour Point [18]; Kinematics Viscosities [19]; Viscosity Index [20] and Flash Point [21].

### 2.7. Design of experiment

The Central Composite Design (CCD) was used to study the effects of the variables of the transesterification reaction and subsequently in the optimization of the process. This method is suitable for fitting a quadratic surface and it helps to optimize the effective parameters with a minimum number of experiments, as well as to analyse the interaction between the parameters. In order to describe the effects of temperature, time and mole ratio on the yield of biolubricant, batch experiments were conducted based on the CCD. The coded values of the process parameters were determined by the following equation:

$$x_i = \frac{X_i - X_0}{\Delta X} \quad (1)$$

where  $x_i$  – coded value of the  $i$ th variable,  $X_i$  – uncoded value of the  $i$ th test variable and  $X_0$  – uncoded value of the  $i$ th test variable at centre point. A second-order polynomial Eq. (2) was used to express the biolubricant yield ( $Y$ ) as a function of the independent variables,

$$Y = b_0 + \sum_{i=1}^k b_i X_i + \sum_{i=1}^k b_{ij} X_i^2 + \sum_{i=1}^k \sum_{j=1}^k b_{ij} X_i X_j + e \quad (2)$$

where  $Y$  is response factor (% yield), and  $i$  and  $j$  denote linear and quadratic coefficients, respectively;  $b_0$  is the intercept,  $b_i$  is the first order model coefficient,  $k$  is the number of factors, and  $e$  is a random number.

The factors levels of independent variables for SBL synthesis is given in Table 1. The regression analysis was performed to estimate the response function as a second order polynomial. A statistical software package, Design Expert 8.7.0.1 was used for regression analysis of the data obtained and to estimate the coefficient of the regression equation. Analysis of variance (ANOVA) was further carried out to determine the adequacy of the model. The

**Table 1**  
Factors levels of independent variables for SBL synthesis.

Independent factors	$-\alpha$	Low level (–)	Medium level (0)	High level (+)	$+\alpha$
Temp, °C	123.82	130	140	150	156.818
Time, min	146.364	160	180	200	213.636
Mole ratio	3.31821	4	5	6	6.61879

significance of each term in the model equation was used to estimate the goodness of fit in each case. Response surfaces were generated to determine the individual and interactive effects of the test variable on the yield of bio-lubricant. The optimal values of the test variables were first obtained in coded units and then converted to the uncoded units. The high (+) and low (–) values for TMP ester synthesis variables were temperature (130 and 150 °C), time (160 and 200 min) and mole ratio (3.0 and 5.0). The alpha ( $\alpha$ ) value used was 1.414. Table 2 shows the complete design matrix of the experiments carried out, together with the results obtained.

### 3. Results and discussion

#### 3.1. Optimisation of the process parameters using response surface methodology

##### 3.1.1. Model fitting and analysis of variance (ANOVA)

The optimization process of the transesterification reaction was done using the Central Composite Design (CCD). Three important factors: temperature, time, and mole ratio were used as the independent variables. Their combined effects were examined while the percentage yield of the bio-lubricant was the response (dependent) variable used to develop the empirical model. This was done to determine the best conditions for optimum yield of biolubricants. CCD generally consists of six centre runs, 2n factorial runs, and 2(n) axial runs, where n is the number of factors. It was used to analyse the correlation between bio-lubricant preparation variables and the percentage yield. In this work, a set of 20 experiments were performed including 6 centre points or null points. The experiments were performed at random to avoid systematic error.

Table 2 shows the combined effects of temperature (A), time (B) and mole ratio (C). The highest percentage of sesame triester (STE) yield of 80.02% was obtained at 150 °C, 200 min, and mole ratio of 6:1 for SME:TMP.

The summary of P-values statistics is shown in Table 3. A quadratic model was suggested from the CCD module. The cubic model is always aliased because the CCD does not contain enough runs to support a full cubic model. A significance level of 95% was used hence all terms whose P-value are less than 0.05 were considered significant. The model summary test and the lack of fit test for the synthesis of biolubricant are also presented in Tables 4 and 5.

The ANOVA results are presented in Table 6 and the model variables from the table show that A, B, C, B<sup>2</sup> and C<sup>2</sup> were all significant, while the regression F-value of 11.29 implies that the model was significant, which was validated by the P-values of less than 0.0001 [22]. The tests for adequacy of the regression models, significance of individual model coefficients and the lack of fit test were performed using the same statistical package. The P values were used as a tool to check the significance of each of the coefficients, which in turn were necessary to understand the pattern of the mutual interactions between the test variables [23]. Therefore, the higher the F-test value and the smaller the P-values, the higher the significance of the corresponding coefficient [24].

“Adeq Precision” measures the signal to noise ratio. A ratio greater than 4 is desirable. The ratio of 12.312 indicates an adequate signal [25]. This model can be used to navigate the design space. The coefficient of regression R<sup>2</sup> was used to validate the fitness of the model equation. For SBL, the R<sup>2</sup> has a high value of 0.9104, showing that 91.04% of the variability in the response can be explained by the model. This implies that the prediction of experimental data was quite satisfactory. The obtained quadratic model equation after eliminating non-significant terms for the transesterification is Eq. (3):

$$\text{Yield (\%)} = +77.23 + 1.10A + 1.47B + 1.39C + 0.65B^2 + 0.63C^2 \quad (3)$$

In a regression equation, when an independent variable has a positive sign, it means that an increase in the variable will cause an increase in the response while a negative sign will result in a decrease in the response [25]. Hence, an increase in temperature,

**Table 2**  
Central composite design arrangement and response for SBL synthesis.

Run	A	B	C	A = Temp	B = Time	C = Mole ratio	% Yield	Predicted value
1	–1	–1	–1	130	160	4	72.28	71.79
2	1	–1	–1	150	160	4	74.15	73.9
3	–1	1	–1	130	200	4	75.19	75.41
4	1	1	–1	150	200	4	76.84	76.99
5	–1	–1	1	130	160	6	75.59	74.64
6	1	–1	1	150	160	6	78.49	77.46
7	–1	1	1	130	200	6	77.99	77.43
8	1	1	1	150	200	6	80.02	79.7
9	–1.41	0	0	123.18	180	5	74.62	75.29
10	1.412	0	0	156.82	180	5	78.51	78.98
11	0	–1.41	0	140	146.36	5	71.69	72.92
12	0	1.412	0	140	213.64	5	77.95	77.86
13	0	0	–1.41	140	180	3.32	73.27	73.1
14	0	0	1.412	140	180	6.68	76.47	77.78
15	0	0	0	140	180	5	77.49	77.23
16	0	0	0	140	180	5	76.86	77.23
17	0	0	0	140	180	5	78.02	77.23
18	0	0	0	140	180	5	76.59	77.23
19	0	0	0	140	180	5	76.92	77.23
20	0	0	0	140	180	5	77.7	77.23

**Table 3**  
Summary of P-values for SBL synthesis.

Source	Sequential p-value	Lack of Fit p-value	Adjusted R-squared	Predicted R-squared	Remark
Linear	<0.0001	0.0379	0.7439	0.6592	Not suggested
2FI	0.9156	0.0225	0.6966	0.5787	Not suggested
Quadratic	0.0324	0.0665	0.8297	0.4234	Suggested
Cubic	0.4460	0.0227	0.8346	−6.8419	Aliased

**Table 4**  
Summary of model statistics for SBL synthesis.

Source	Std. Dev.	R-Squared	Adjusted R-squared	Predicted R-squared	PRESS	Remark
Linear	1.12	0.7843	0.7439	0.6592	31.45	Not suggested
2FI	1.21	0.7924	0.6966	0.5787	38.89	Not suggested
Quadratic	0.91	0.9104	0.8297	0.4234	53.21	Suggested
Cubic	0.90	0.9478	0.8346	−6.8419	723.71	Aliased

**Table 5**  
Lack of fit test for SBL synthesis.

Source	Sum of squares	df	Mean square	F Value	p-value Prob > F	Remarks
Linear	18.36	11	1.67	5.39	0.0379	Not suggested
2FI	17.61	8	2.20	7.11	0.0225	Not suggested
Quadratic	6.72	5	1.34	4.34	0.0665	Suggested
Cubic	3.27	1	3.27	10.57	0.0227	Aliased
Pure Error	1.55	5	0.31			

**Table 6**  
ANOVA for SBL synthesis.

Source	Sum of squares	df	Mean square	F value	p-value Prob > F
Model	84.02	9	9.34	11.29	<0.0001
A-Temperature	16.46	1	16.46	19.90	0.0012
B-Time	29.46	1	29.46	35.62	0.0001
C-Mole ratio	26.47	1	26.47	32.00	0.0002
AB	0.15	1	0.15	0.18	0.6807
AC	0.25	1	0.25	0.30	0.5956
BC	0.35	1	0.35	0.42	0.5308
A <sup>2</sup>	0.016	1	0.016	0.019	0.8923
B <sup>2</sup>	6.09	1	6.09	7.37	0.0218
C <sup>2</sup>	5.77	1	5.77	6.97	0.0247
Residual	8.27	10	0.83		
Lack of fit	6.72	5	1.34	4.34	0.0665
Pure error	1.55	5	0.31		
Cor total	92.29	19			

Std. Dev. = 0.91; mean = 76.33; C.V. = 1.19%; PRESS = 53.53.  
R-Squared = 0.9104; Adj R-Sq = 0.8297; Pred R-Sq = 0.4234; Adeq precision = 12.312.

time, and mole ratio will cause an increase in the percentage yield. Time and mole ratio will have more significant effect in the increment of the response since their coefficients were higher.

Normal plot of residuals (Fig. 1) and the predicted vs actual plots (Fig. 2) were used to check the distribution of residuals. The close distributions of the points along the straight lines indicate a good relationship between the experimental values and the predicted values of the response. These plots also confirm that the chosen model was adequate for prediction of the response variables in the experimental values.

### 3.1.2. Effect of process parameter on transesterification reaction

The two factors interactions response surface plots were drawn for the three reaction parameters of temperature, mole ratio and time. The TE yield was found to vary between 71.69 and 80.02 wt%. Fig. 3 shows the plots of the effect of temperature and mole ratio on TE yield at a fixed time of 3 h. Fig. 4 shows the plots of the effect of temperature and time at fixed mole ratio of 5:1. Fig. 5 shows the plots of the effect of mole ratio and time at a fixed temperature of 140 °C.

The response surface contours which are graphical results of interactive effects are shown in Figs. 6–8. The optimum value of Y was found to be 80.02%, which was higher than the highest value amongst the calculated values based on experimental design. The surface plot of conversion (Y) of TMP based biolubricant as a function of FAME/TMP molar ratio and temperature demonstrated maximum conversion of Y = 80.02% as obtained from the contour plot at maximum value of FAME/TMP ratio and temperature.

The response surface of extent of conversion showed (Figs. 6–8) a clear peak, suggesting that the optimum condition for maximum yield (Y) was well inside the design boundary. It could be observed from the 3D plot (Figs. 6–8) that the conversion increased when molar ratio and temperature were increased. It was observed that for temperature, yield (Y) increased with increasing mole ratio before ceasing; whereas for lower values of temperature, the pattern followed a parabolic path. At fixed value of molar ratio, the increased temperature increased the value of the yield (Y) until a point where there was no significant increase in yield (Fig. 8) [26].

Fig. 6 shows response for the interaction of mole ratio and temperature, while Fig. 7 shows response for interaction of mole ratio

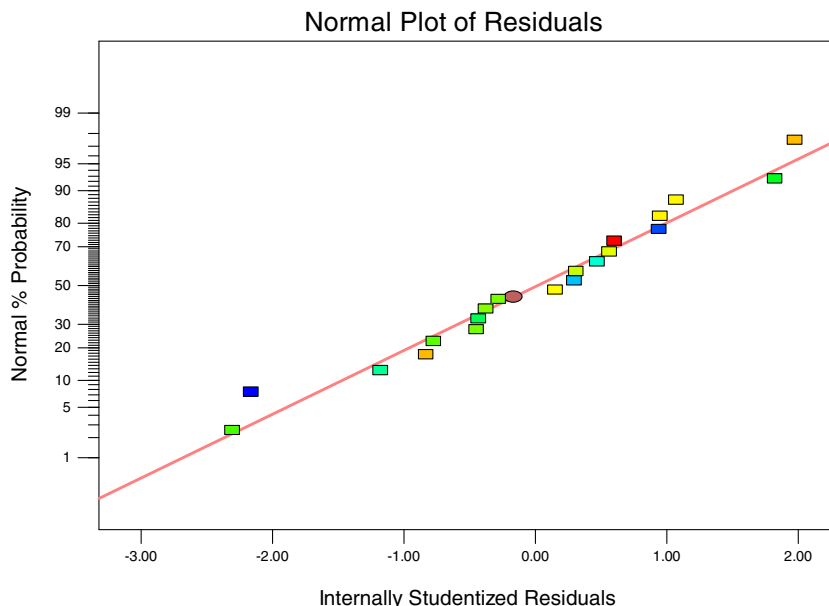


Fig. 1. Normal plot of residuals SBL synthesis.

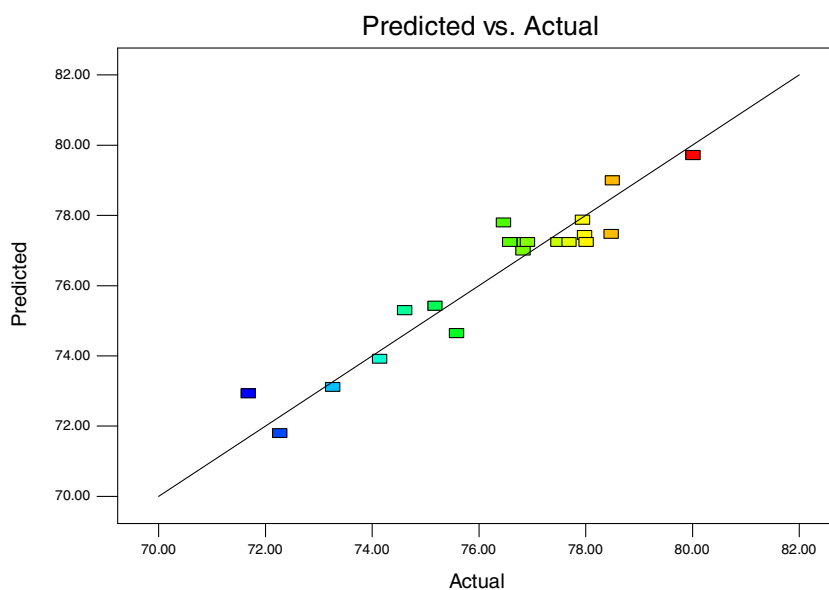


Fig. 2. Predicted vs actual plot for SBL synthesis.

and time. The 3D responses (Figs. 6 and 7) indicate that the biolubricant synthesis increased when FAME concentration increases. Therefore, the maximum biolubricant conversion was obtained for high mole ratio. This was due to the stoichiometry of transesterification, which requires a 3:1 ratio for FAME and TMP, since the reaction involves the conversion of one ester and an alcohol towards another ester and another alcohol. Thus, an excess FAME could be used to drive the reaction near completion. Consequently, the FAME concentration results in a greater biolubricant conversion within a shorter time as indicated in Fig. 6.

Higher yield of biolubricant is strongly favoured when high molar ratio is employed for certain time of reaction (B) and temperature (A). The mole ratio (FAME/TMP) is a fundamental variable in the transesterification process for biolubricant synthesis [27].

3D response surfaces (Figs. 6 and 8) revealed that increment of reaction temperature leads to increase in biolubricant yield with

reduced time. On the contrary, increase in reaction time does not improve the catalytic activity at low temperature level. Meanwhile, high temperature improved the dispersion of catalyst particle in liquid medium with better mass transfer between the reactants. The contour plot shows that high biolubricant yield was favoured by higher level of reaction temperature with low or intermediate level of reaction time.

Furthermore, the 3D response surface revealed that increment of reaction temperature leads to increase in biolubricant yield with reduced reaction time. On the other hand, increase in reaction time does not have significant effect on the yield after about 3 h of reaction. Meanwhile, high temperature improved dispersion of catalyst particle in liquid medium with better mass transfer between reactants. Also contour plots show high biolubricant yield at high level of temperature with low intermediate of reaction time.

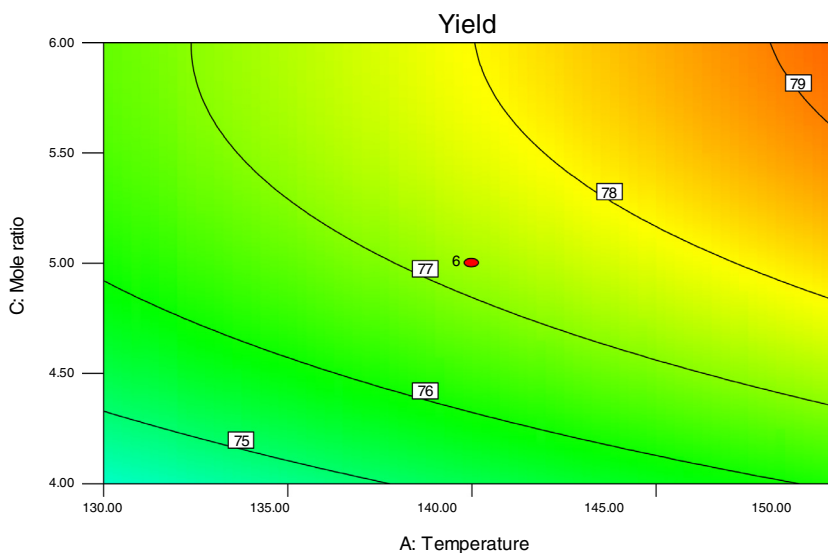


Fig. 3. 2D surface plot for effect of temperature and mole ratio on SBL yield.

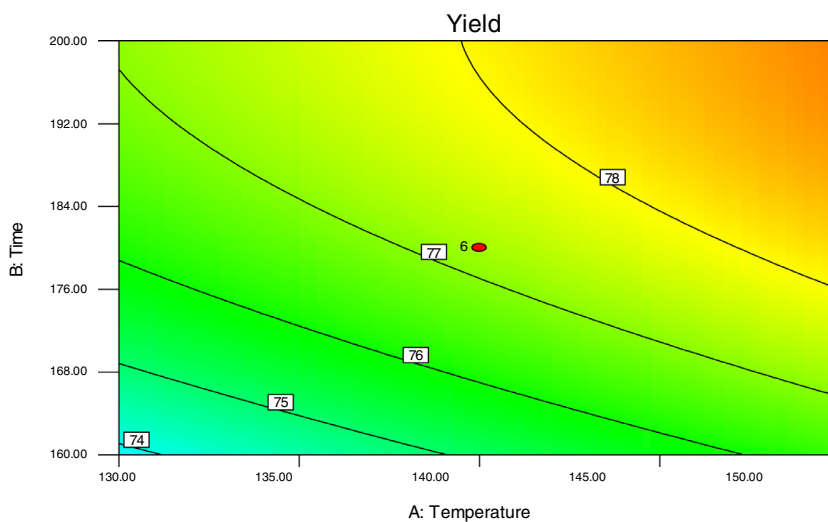


Fig. 4. 2D surface plot for effect of temperature and time on SBL yield.

### 3.1.3. Process variable optimization and validation

Table 7 shows the optimization and validation results for SBL synthesis. Predicted responses were generated using point prediction node (under optimization node in the CCD module). Temperature, molar ratio of reactants and time were all fixed in the range of 80–160 °C, 4–6 and 1–5 h respectively. TE yield was maximized within the experimental range of 71.69–80.02 wt%. Using these criteria, the software suggested (see Table 7) the following optimum conditions: temperature of 150 °C, mole ratio of 6, time of 197.26 min and yield of 80.02 wt% at a desirability value of 1.00. Fig. 9 shows surface and contour plots for desirability, wherein desirability is plotted against temperature and mole ratio respectively at a fixed time of 200 min while Fig. 10 shows 3D contour plot for desirability against temperature and mole ratio for SBL synthesis. Sripada [27] applied this method to the transesterification reaction between methyl oleate and TMP and determined the optimum condition to be temperature of 139 °C, mole ratio 4.9 and catalyst loading of 1.42 wt% and obtained a TE yield of 91.2 wt%. Yunus et al. [9] determined the optimum synthesis conditions for maximum conversion of palm oil methyl esters to TMP

triester to be temperature of 130 °C, vacuum pressure of 20 mbar, catalyst (sodium methoxide) at 0.8% w/w; mole ratio of palm oil methyl esters to TMP at 3.9:1. Ghazi et al. [12], utilized a temperature of 150 °C, with a pressure of 10 mbar, mole ratio of JME-to-TMP of 4:1.

A confirmatory experiment was conducted (at the optimum conditions) to verify the accuracy and applicability of the developed optimum conditions at TE yield of 80.02 wt%. The validation experiment indicated TE yield of 79.07 wt% as shown in Table 7. Hence, the significant closeness between 80.02 and 79.07 wt% confirmed that optimum reaction conditions developed were found to be valid for the experimental data. Fig. 2 shows a comparison plot of predicted versus actual values which indicated that relationship between modelled and experimental values was excellent. Fig. 12 shows plots for standard error of design as a function of temperature and time.

Studies were performed to check for the reproducibility of data obtained from transesterification reaction for the synthesis of SBL. The centre point experiment was performed 6 times as indicated by the CCD model. The final TE yield varied between 76.59 and

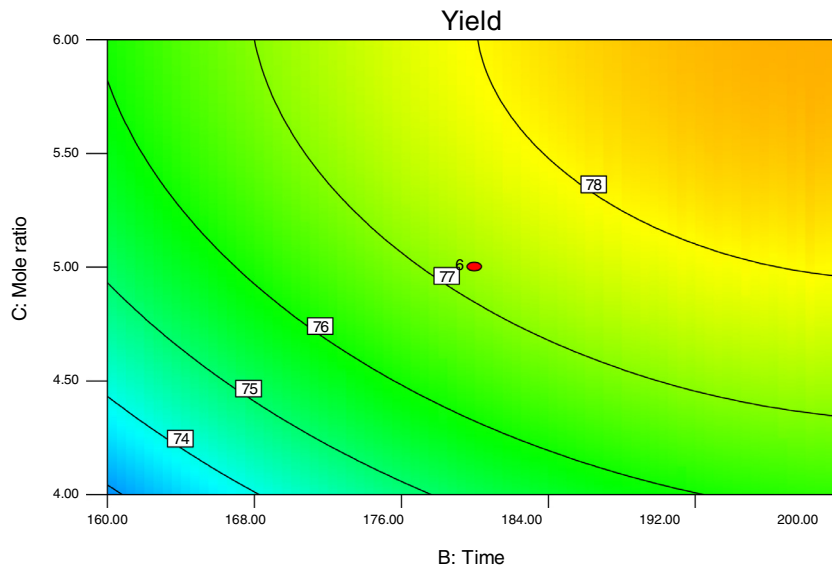


Fig. 5. 2D surface plot for effect of mole ratio and time on SBL yield.

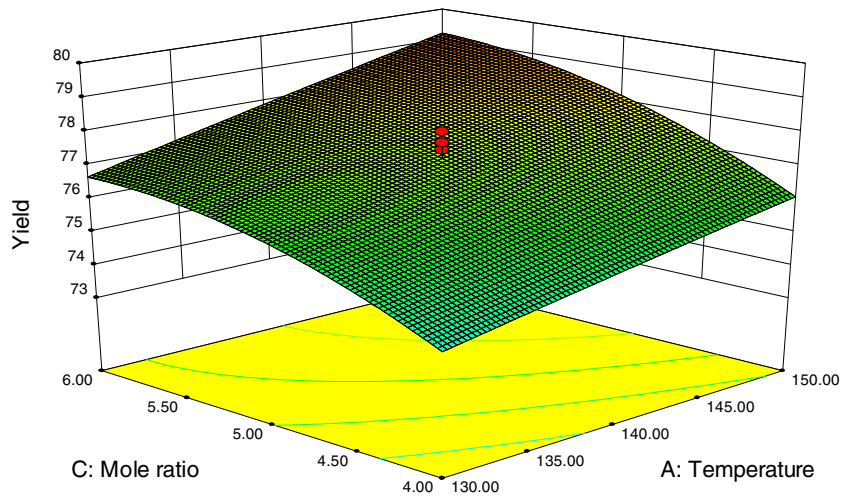


Fig. 6. Surface and contour plot between mole ratio and temperature against SBL yield.

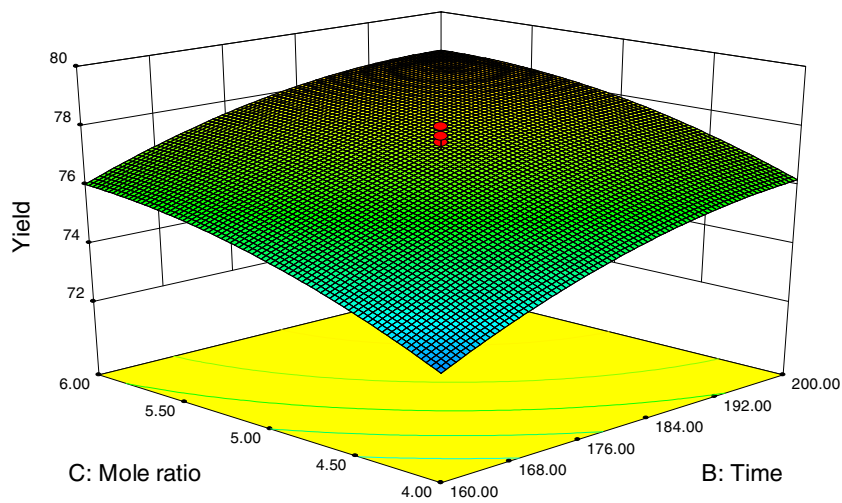


Fig. 7. Surface and contour plot between mole ratio and time against SBL yield.

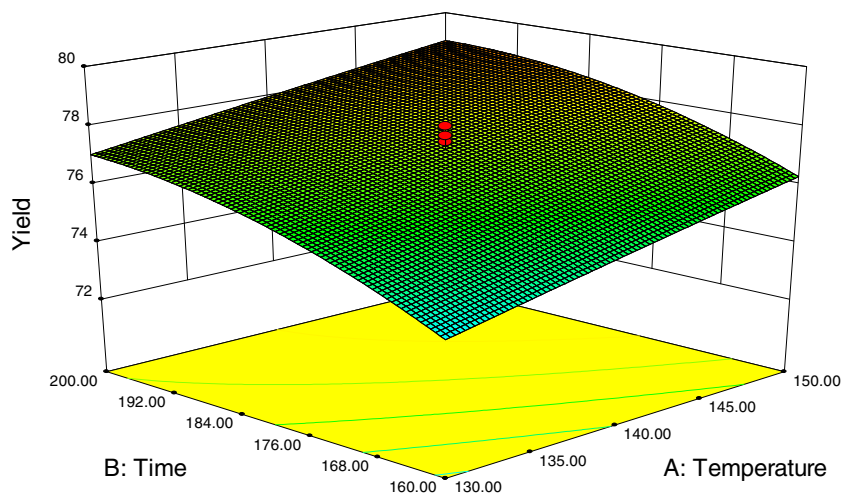


Fig. 8. Surface and contour plot between time and temperature against SBL yield.

**Table 7**  
Optimization/validation results for SBL synthesis.

Parameter	Optimum operating condition
Temperature (°C)	150
Time (min)	197.26
Mole ratio	6:1
Yield (optimum) wt%	80.02
Yield (validated) wt%	79.70 ± 0.03

78.02 wt%. Fig. 11 show scatter plots of the six experiments at the centre point. It was observed that the replicates showed excellent similarities in product development. It was indicated that these yields differed by less than 2.0 wt%, while the standard deviation between these values was found to be less than 0.75%. Thus, it was confirmed that the transesterification experiments were repeatable.

All the remaining 14 experiments were each performed twice to check if they were repeatable. The percentage error for each was found to be less than 3%. Thus, it was confirmed that the transesterification experiments were repeatable.

### 3.2. Fourier transform infrared (FTIR) analysis of the SBL

The spectrum from the FTIR (figure not shown) analysis displays several absorption peaks between 4000 and 700  $\text{cm}^{-1}$ . FTIR peaks of SBL indicated presence of absorption band at 1737.388  $\text{cm}^{-1}$  which belongs to the C=O carboxylic acid. Another evidence of trimethylolpropane ester produced was the appearance of C–O–C stretching vibration peak of trimethylolpropane ester at 1170.148–1050  $\text{cm}^{-1}$ . The peaks at 1176, 1173, and 1117  $\text{cm}^{-1}$  of SBL are referred to as C–O stretching ester. FTIR spectrum also showed absorption bands at 714.6795  $\text{cm}^{-1}$  of C–H group. A similar observation has been reported for the FTIR elsewhere [15,28,29].

### 3.3. Lubricating properties of sesame biolubricant

A lubricant's viscosity is generally considered one of its most important properties. This is because if the viscosity of a lubricant is even modestly different from what is needed for a particular component and application, the lubricant will not be able to lubri-

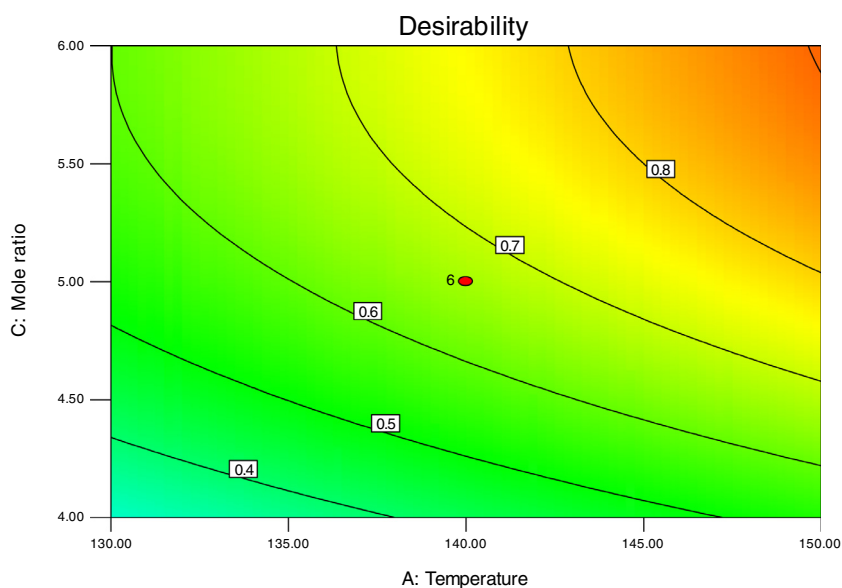


Fig. 9. 2D surface plot for desirability against temperature and mole ratio for SBL.



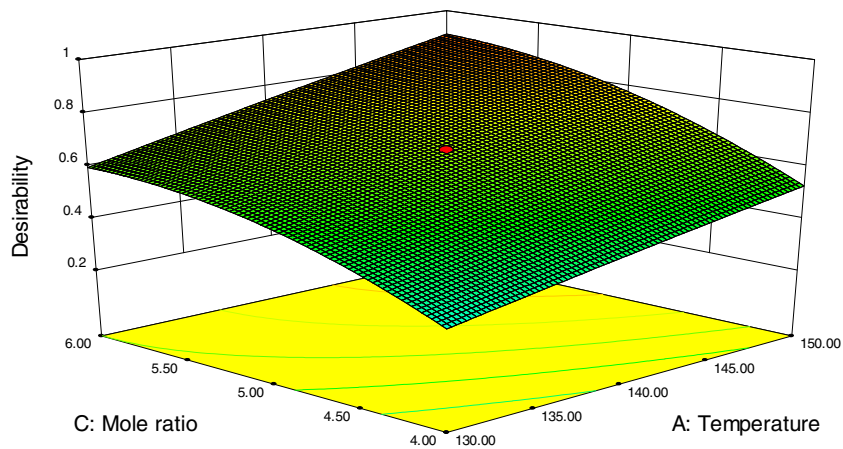


Fig. 10. 3D contour plot for desirability against temperature and mole ratio for SBL synthesis.

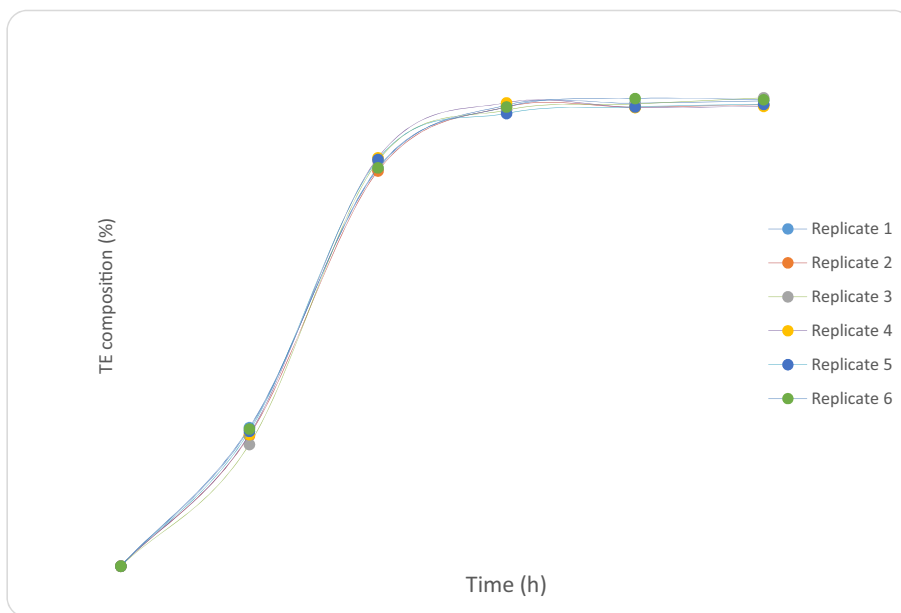


Fig. 11. Scatter plot of STE composition at different reaction times from six replicate reactions.

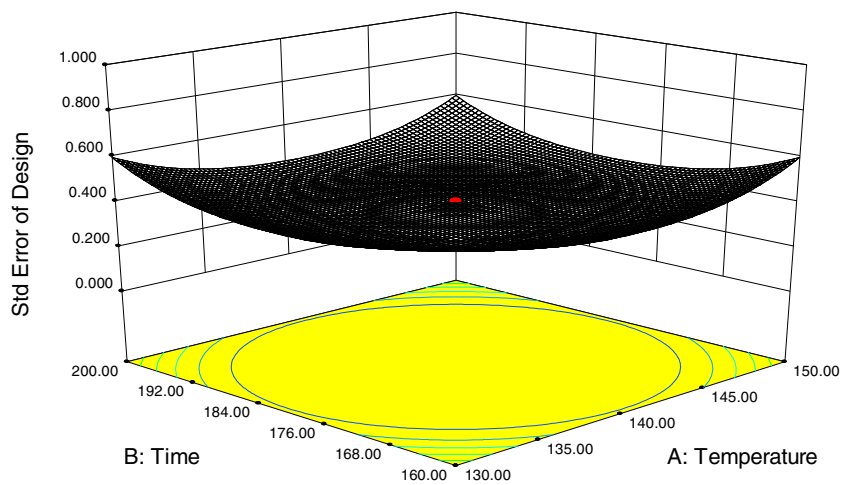


Fig. 12. 3D contour plot for standard error of design a function of temperature and time (SBL).

**Table 8**  
Properties of SBL and comparison with other plant based biolubricants and ISO viscosity grades.

Property	SBL	CSO	Sesame biolube [33]	Rapeseed biolube [10]	Palm Oil biolube [9]	ISO VG32 [34]	ISO VG46 [34]	ISO VG68 [34]	ISO VG100 [34]
<i>Viscosity (Cst)</i>									
40 °C	35.55	27.33	35.43	35.34	52.4	>28.8	>41.1	>61.4	>90
100 °C	7.66	6.3	7.93	7.99	10.2	>4.1	>4.1	>4.1	>4.1
<i>Viscosity index (VI)</i>									
Flash point (°C)	193	193	206	209.2	186	>90	>90	>198	>216
Flash point (°C)	196	130	–	–	–	–	–	–	–
Pour point (°C)	–21	–15	–12	–15.5	–5	<–10	<–10	<–10	<–10

cate the component effectively. Thus, this may lead to significant damage and possibly equipment failure.

In particular, kinematic viscosities were determined at 40 and 100 °C. SBL was found to have kinematic viscosities of 35.55 and 7.66 cSt at 40 and 100 °C, respectively. Various kinematic viscosities have been reported by different authors for different biobased lubricants at 40 °C. Such reports include 39.7 cSt and 54.1 cSt for TMP esters of palm oil and palm kernel oil [9], 43.9 cSt for TMP esters of jatropha curcas oil [12] and 11.2–36.1 cSt for TMP esters of 10-undecenoic acid [30]. In comparison, TMP-based biolubricant from SME exhibited very favourable viscosities. Furthermore, when compared with ISO VG 32 standard specification for light gear lubricant [31], the properties of the SBL based oil met the stipulated specification.

Ideal oil for most purposes is one that maintains a constant viscosity throughout temperature changes. The viscosity index (V.I.) of oil is a number that indicates the effect of temperature changes on the viscosity of the oil. A low V.I. signifies a relatively large change of viscosity with changes of temperature. In other words, the oil becomes extremely thin at high temperatures and extremely thick at low temperatures. On the other hand, a high V.I. signifies relatively little change in viscosity over a wide temperature range. The higher the VI, the more preferable the lubricant is. Using the kinematic viscosity values at 40 and 100 °C, the viscosity index of SBL was found to be 193. Sripada [27] reported VI of 193 and 204 for the synthesis of biolubricant from methyl oleate and canola biodiesel, respectively. Meanwhile, Gryglewicz et al. [10] recorded a VI range of 209–235 for the synthesis of neopentyl glycol and trimethylolpropane esters of olive oil, rapeseed oil and lard fatty acids. Yunus et al. [9] found that TMP esters of palm and palm kernel oils exhibited very high VI of 167–187. Accordingly, Ghazi et al. [12] and Rao et al. [30] obtained VI of 180 for jatropha curcas-derived TMP esters and 162–172 for polyol esters of 10-undecenoic acid respectively. Furthermore, Åkerman et al. [32] reported that TMP esters of C<sub>5</sub>–C<sub>18</sub> fatty acids had VI of 80–208. When compared with the results obtained by other researchers, SBL had shown a relatively favourable VI.

For a lubricant to perform in extremely cold environments, low-temperature fluidity is a highly essential property. The pour point, or the temperature at which a lubricant ceases to flow, is important in appraising flow properties at low temperature. As such, it can become the determining factor in selecting one lubricant from among a group with otherwise identical properties. Pour point is the lowest temperature at which movement is observed in the lubricant. The values of pour point, viscosities (at 40 °C and 100 °C) and the viscosity index of SBL were presented in Table 8 and compared with other plant based lubricant and ISO viscosity grade requirement. Oil extracted from sesame seed and sesame crude oil (CSO), were tested for pour point and viscosity and further compared with SBL. The obtained pour points for SBL and CSO were –21 °C and –15 °C, respectively. This indicated that the tranesterification reaction enhanced the pour point. In addition,

the pour point for the SBL was found to be comparable to other plant based biolubricants.

Flash point of SBL showed great improvement over the CSO and thus justified the chemical modification of the oil. However, palm oil biolubricant showed much more stability than SBL due to the higher saturated molecules present in the palm oil. There was not much difference between the viscosities of CSO and SBL. However, their viscosity index were found to be slightly lower than other plant based biolubricants. Lubricants are usually identified according to their grades.

#### 4. Conclusion

RSM based on CCD was successfully applied for optimizing the reaction parameters for the synthesis of trimethylolpropane based sesame biolubricant. The effects of reaction temperature, mole ratio and reaction time on the transesterification reaction yield were also studied and optimized. A polynomial quadratic model was used to predict maximum yield and the optimization of reaction parameters. The model predicted the optimal conditions for the selected transesterification variables as reaction temperature of 150 °C, mole ratio of 6:1 and reaction time of 197.26 min with an actual sesame biolubricant yield of 80.02 wt%. The ANOVA implied that mole ratio and time were the most significant factors affecting the yield of biolubricant. The resulting sesame biolubricant properties also satisfied the ISO VG 22 biolubricant standards. The optimized sesame biolubricant production using calcium hydroxide as a catalyst could be applied in a sesame biolubricant pilot plant.

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