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STATISTICAL APPROACH TO THE OPTIMIZATION OF  
BIODIESEL PRODUCTION FROM *JATROPHA CURCAS* OIL

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### ABSTRACT

Biodiesel is a renewable and alternative source of energy that can minimise the environmental pollution associated with burning of fossil fuels. In this work, optimization of biodiesel production from *Jatropha curcas* oil was investigated. Biodiesel was produced via base catalyzed transesterification reaction of the oil with methanol. This process was optimized by the application of two-level- four factor ( $2^4$ ) factorial design of response surface methodology (RSM) requiring 16 experiments. A linear model of  $Y = 84.86 + 4.981X_1 - 3.50X_2 - 2.50X_3 + 3.10X_4 + 5.13X_1X_2 - 0.71X_1X_4 + 3.02X_2X_3 + 0.13X_2X_4 - 2.31X_1X_2X_4$  was obtained to predict the yield of biodiesel (Y) as a function of reaction time ( $X_1$ ), KOH catalyst concentration ( $X_2$ ), methanol to oil ratio ( $X_3$ ) and temperature ( $X_4$ ). A modified statistical model comprising of all significant factors obtained by multiple regression predicted that the highest yield of biodiesel was 98.3% with the following optimization rules:  $X_1 = 2.50$ h,  $X_2 = 1.1\%$  (w/v),  $X_3 = 3:1$  and  $X_4 = 70^\circ\text{C}$ . Also, there was significant interaction between  $X_1X_2$ ,  $X_2X_3$  and  $X_2X_4$ . Gas chromatographic analysis of the JCO biodiesel identified myristate (1.50%) and palmitoleate (98.5%) as the major fatty acid methyl esters. The model has been found to describe the experimental range studied adequately.

**Keywords:** Optimization; Biodiesel; *Jatropha curcas* oil; Transesterification; Response surface methodology.

### 1. INTRODUCTION

Energy consumption has increased tremendously over the years as a result of increase in world population, urbanization and industrialization. In Nigeria, there is need to search for alternate fuel like biodiesel that will replace the conventional fossil fuel as this will reduce their contribution to global warming through emission, cut down dependence on the petroleum sector and diversify our economy (Akintude et al., 2015). According to Betiku et al. (2004), wastes and combustible renewable materials with their highest potential to supply the global energy requirements accounted for more than three quarters of global energy supply in 2007 and 2008.

Biodiesel can be used to replace the conventional fossil fuel because it is biodegradable, sustainable, non-toxic, and environmentally friendly with less pollutant in emissions and can be used as an additive in fossil fuel (Achten et al., 2008; Rodriguez et al., 2008). Biodiesel when compared to fossil fuel reduces the emission of green house gas by 41% during combustion because the carbon dioxide that is released during the process can be broken down and re-used during the process of photosynthesis (Reefat., 2010; Vincente., 1998).

Biodiesel also known as mono alkyl ester of long chain fatty acids can be produced from waste oils, plant oils (edible and non edible e.g moringa seed oil, palm oil, coconut oil, soy bean oil, Neem oil, rapeseed oil, tung oil, yellow oleander oil, sorrel oil,

and jobba oil e.t.c), micro algae oils and animal fats (Ighose et al., 2016; Offor et al., 2015). The most preferred raw material for the production of biodiesel are the non edible oil and waste vegetable oil because of the tremendous demand for edible oil as food there by making it far too expensive to be used as fuel presently (Adebayo et al., 2011; Baroi et al., 2009; Encinar et al., 2010; Offor et al., 2015; Tiwari et al., 2007).

*Jatropha curcas* oil (JCO), a non edible oil that is considered as a feed stock for production of biodiesel because of its availability and cost is processed from *Jatropha curcas* seeds. It belongs to the family of *Euphorbiaceae* and is a second generation cropping solution for biofuel production with its seeds yielding between 30 to 60% of oil (Ma&Hanna., 1999; Rashid et al., 2010). The presence of some antinutritional factors such as toxic phorbol esters and stearic acid render *jatropha* oil unfit for human consumption (Rashid et al., 2010). *Jatropha curcas* oil is a non drying oil that is odorless and colorless when fresh but can change to yellow when left for a long time (Pramanik., 2003). *Jatropha curcas* oil is used commercially as natural pesticide, cosmetics and in candle production (Martinez et al., 2006).

Biodiesel is commonly produced using transesterification reaction because it is simple when compared to other methods and can give high yields of biodiesel (Attanatho et al., 2004). There are

factors that affect the transesterification process and the yield of biodiesel which include reaction time, catalyst concentration, methanol/ethanol to oil molar ratio, temperature etc (Ma & Hanna., 1999). Most commercial processes use base catalyst such as NaOH, KOH or related alkoxides for biodiesel production because base catalyzed reaction is faster than acid catalyzed ones (Offor et al., 2015). The major problem associated with base catalyzed reaction is the formation of soap. However, subsequent washing of biodiesel is required to remove the soap in order to maintain its quality (Baroi et al., 2009).

The need to optimize the biodiesel produced is very important as it will save time, reduce production cost, increase the yield of oil and reduce the number of experiments needed to provide sufficient results (Bas & Boyaci., 2007). The most widely used multivariate statistic technique used is response surface methodology (RSM) which helps to solve the problem of optimizing one factor at a time (Bezzer et al., 2008; Fristak et al., 2002; Stamenkovic et al., 2011). Response surface methodology is as statistical tool or group of mathematical and independent statistical technique used to study the effect of variables and their interactions in a process (Akintude et al., 2015; Fristak et al., 2002). The RSM has been used in the production and optimization of biodiesel produced from different seed oils such as castor oil, cotton seed oil, soya bean oil, rapeseed oil waste cooking oil and lard among others (Olutoye et al., 2011; Shaw et al., 2003). From the literature, most authors optimized biodiesel yield using two or three process variables in the optimization of biodiesel produced from *J. curcas* but there is relatively scarce indigenous research on the optimization of *Jatropha* biodiesel using two-level-four-factors factorial design. This work focused on the modeling and optimization of biodiesel yield from locally sourced *J. curcas* oil using RSM. The effect of the reaction time, methanol to oil ratio, catalyst concentration and reaction temperature and their reciprocal interaction on the biodiesel yield were investigated.

## 2.0 MATERIALS AND METHODS

### 2.1 Preparation of Materials

Freshly matured seeds of *Jatropha curcas* were obtained from Afikpo, Ebonyi State Nigeria. The seeds were cleaned, de-shelled and dried in a hot airoven (SM9053, Uniscope, England) at 50°C for 72h. All chemicals and reagents used were of analytical (AR) grades and obtained from BDH England.

### 2.2 Extraction of Oil from *Jatropha curcas* Seeds

The oil was extracted as described by Betiku, Omole & Aluko (2012). The seeds of *J. curcas* were milled with a hard mill (Victoria Columbia) of which 30g

was carefully transferred into a thimble prior to soxhlet extraction. The round bottom flask of the apparatus filled with 100cm<sup>3</sup> of n-hexane (bp 68°C) was fixed at the bottom of the extractor. The whole set up was heated on a heating mantle at a temperature of 70°C. Water was made to circulate from the frigostat at 9°C. The extractor was allowed to run for an hour in each set before the heat source was removed. The round bottom flask with the extract was allowed to cool before weighing. The oil was separated from the solvent by using distillation assembly and the extracted oil was measured. The oil composition based on the fatty acid content was determined by gas chromatography (GC).

### 2.3 Experimental Design and RSM Modeling

The experimental design used in this work was 2<sup>4</sup> factorial design of RSM. The independent variables studied were reaction time (1.5-2.5h), catalyst concentration (1-3%), methanol to oil molar ratio (1:3-9:1) and temperature (50-70°C). The four input variables were studied at low(-1) and high (+1) levels. The effect of these variables on the biodiesel produced was studied using RSM. The order of experimentation was completely randomized to avoid systematic errors. The coefficient of determination (R<sup>2</sup>) (Equation 1), adjusted R<sup>2</sup> (Equation 2), probability value at 95% confidence interval, predicted R<sup>2</sup>, coefficient of variation and analysis of variance were employed as statistical indicators. The modeling and the optimization of the *Jatropha* based diesel were done using 2<sup>4</sup> factorial design-RSM of the Design Expert software version 8.0.7.1 (Stat-ease Inc., MN, USA).

$$R^2 = 1 - \frac{\sum_{i=1}^n (a_{e,i} - a_{p,i})^2}{\sum_{i=1}^n (a_{p,i} - a_{e,ave})^2} \quad (1)$$

$$\text{Adjusted } R^2 = 1 - \left[ (1 - R^2) \times \frac{n - 1}{n - j - 1} \right] \quad (2)$$

### 2.4 Experimental Protocols

A preliminary titration was carried out with JCO sample, methanol and phenolphthalein indicator to determine the amount of KOH catalyst of given concentration was needed for complete transesterification reaction. Due to the high acid value of 8.52 mgKOH/g oil corresponding to a FFA level of 4.26% which was high, the FFA was reduced to less than 1% via esterification reaction prior to transesterification proper.

#### 2.4.1 Acid catalyzed esterification step

Esterification of the *J. curcas* oil was performed according to the method described by Jaliliannosrati (2013) with some modifications. Exactly 20g of JCO

was measured into a 250ml glass reactor and heated to appropriate temperature on a magnetic stirrer hot plate. Exactly 7.5%(v/v) of H<sub>2</sub>SO<sub>4</sub> was added to the oil in the reactor and allowed to stir for 5mins after which 10.5% (v/w) of methanol to oil ratio was added to the reactor. The contents were stirred and heated on a magnetic stirrer hot plate for 35mins. The esterified oil was transferred into a separating funnel and the mixture separated into two layers (methanol-water layer and *J.curcas* oil). The pretreated oil, which was at the bottom layer was decanted into a flask and the excess methanol in esterified JCO was rid off by heating prior to determination of acid value. The high free fatty FFA's were reduced to 0.34% and the resulting mixture subjected to base catalyzed transesterification reaction.

#### 2.4.2 Base catalyzed transesterification of the esterified JCO

The base catalyzed transesterification of the esterified *J. curcas* oil was performed according to the method described by Offor *et al.* (2015) with some modifications. The reaction was carried out in a reactor which is 250cm<sup>3</sup> conical flask equipped with reflux condenser (to minimize ethanol loss). A predetermined volume of KOH catalyst was dissolved in 6cm<sup>3</sup> of methanol to obtain potassium methoxide which was gently added to 2cm<sup>3</sup> of JCO in the reactor. The amount of KOH dissolved in methanol was based on the amount needed to neutralize the un-reacted acids. The mixture was heated to a certain temperature and the reaction monitored according to the factorial design variables to yield biodiesel and glycerol. At the end of the reaction, the products were cooled at room temperature and transferred to a separating funnel. The product was allowed to settle overnight before the glycerol was tapped off and the *jatropha* biodiesel was washed three times with hot distilled water to remove residual catalyst, glycerol, methanol and soap. The biodiesel yield was calculated as described in equation (3)

$$\text{Yield of biodiesel} = \frac{\text{Volume of the } Jatropha \text{ biodiesel produced}}{\text{Volume of the esterified } Jatropha \text{ curcas oil used}} \times 100 \quad (3)$$

#### 2.4.3 Physicochemical analysis of crude JCO and quality of JCO- based biodiesel

The refractive index, specific gravity, saponification vaue, peroxide value, relative density, viscosity, acid value, FFA and pour point were determined using the method of Association of official Analytical Chemists (1990) Iodine value was dertermined using Wij's method (Firestone, 1994).

### 3.1 Physicochemical analysis of crude JCO and quality of JCO- basedbiodiesel

The results of the physicochemical properties of the *J.curcas* oil and the the biodiesel produced are presented in Table I.

**Table I: Physicochemical properties of JCO and Jatropha biodiesel.**

| Parameter                                | JCO         |
|--|-------------|
| Jatropha biodiesel                       | ASTM D 6751 |
| Refractive index at 40°C                 | 1.49        |
| 1.47                                     | NS          |
| Specific gravity at 40/25°C              | 0.91        |
| 0.90                                     | 0.86-0.90   |
| Saponification value (mg/KOH)            | 196.60      |
| ND                                       | NS          |
| Iodine value (I <sub>2</sub> /100g)      | 102.70      |
| ND                                       | NS          |
| Kinematic viscosity at 40°C              | 44.38       |
| 4.23                                     | 1.9-6.0     |
| Peroxide value                           |             |
| (O <sub>2</sub> equivalent/1000g sample) | 9.42        |
| ND                                       | ND          |
| Pour point (°C)                          | -3.0        |
| -5.0                                     | NS          |
| Density (g/cm <sup>3</sup> )             | 0.91        |
| ND                                       | ND          |
| Acid value                               | 8.52        |
| ND                                       | ND          |
| %FFA                                     | 4.26        |
| ND                                       | ND          |
| Flash point(°C)                          | ND          |
| 138.00                                   | ND          |

ND-not determined, NS-not specified

The refractive index is an intrinsic characteristic and an indication of purity of the oil (Arrelano *et al.*, 2016).The refractive indexwas 1.49 andis similar to the value (1.4692) reported by Montes *et al* (2011) and 1.46 reported by Gandhi & Senthilkumarin (2015).

The specific gravity of the oil(0.9064Kg/L) is in agreement with value of 0.917Kg/L reported by Gandhi &Senthilkumarin, (2015) and 0.913Kg/L reported by Joshi *et al.*(2011) The density of 0.914gcm<sup>-3</sup> is lower than the reported value of 0.920gcm<sup>-3</sup>by Odesanya *et al.* (2015) and value of 0.914 gcm<sup>-3</sup> obtained by Karaj *et al.*, 2008).

Iodine value is a measure of the average amount of unasaturated fats and oil (Adebayo *et al.*, 2011; Arellano *et al.*, 2016). The oil indicates a high content of unsaturated fatty acid due to its high iodine value (102.69I<sub>2</sub>/100g). The iodine value obtained has shown that the oil is a non-drying oil and may find useas a raw material in the manufacture of oil paints. The iodine value obtained is similar to the value (110.5I<sub>2</sub>/100g) reported by Gandhi&Senthilkumarin (2015) 104.9I<sub>2</sub>/100g reported by Abdulhamid *et al.* (2013). but lower

than the value 135.85I<sub>2</sub>/100g reported by Jumat&Abdullahi (2008).

The saponification value is a measure of the acid present and indication of the purity of the oil. The saponification value of 194.64mg/KOH is comparable to the value of 195.7mg/KOH reported by Joshi *et al.*(2011) but lower than the value 203.36mg/KOH reported by Abdulhamidet *al* (2013).The kinematic viscosity 44.38cSt is higher than the value 40.28cSt reported Gandhi& Senthilkumarin (2015)

The flash point of the biodiesel was 138°Cand is in agreement with ASTM and EN specification for biodiesel (min 130°C). The flash point value is lower than the value of170°C reported by Adebayo *et al.* (2011) and higher than the value (105°C) reported by Ghandi & Senthilkumarin (2015), Flash point helps to monitor the safe handling of the storage of fuel. The higher the flash point, the safer the fuel and vice versa (Adebayo *et al.*,201) The kinematic viscosity of 4.2cst demonstrates that the transesterification process reduced the kinematic viscosity of the oil by 10-fold (Ighose *et al.*, 2016) and is in agreement with the manufacturer standard given and falls within ASTM limits of biodiesel. The viscosity is within the values 4.2cts reported by Ghandi & Senthilkumarin (2015) and 4.8 cts reported by Adebayo *et al.* (2011). The viscosity of biodiesel is higher compared to that of fossil diesel (3.6cts) which implies that the biodiesel will have lubricating effect on the engines which will be an added advantage to the users since it will reduce wear and tear in the engine (Adebayo *et al.*, 2011).

The refractive index value of 1.466 is similar to the value 1.43 reporetd by Ghandi & Senthilkumarin (2015).The specific gravity of biodiesel obtained in this study (0.9077) is similar to the value reported byGhandi & Senthilkumarin (2015). 0.88 reported by Belewu *et al.* (2010)

### 3.2 Fatty acid composition analysis of crude JCO

The oil content of the *Jatropha curcas* seed was 48.5%, which is higher than the value reported by Adebayo *et al* (2011).

**Table II: Fatty acid composition and percentage yield**

| Fatty acid       | composition |
|------------------|-------------|
| Percentage yield |             |
| Oleic            | 50.25       |
| Linoleic         | 17.03       |
| Myristoleic      | 10.25       |
| Caprylic         | 8.83        |
| Capric           | 6.35        |
| Lauric           | 3.93        |

|               |      |
|---------------|------|
| Myristic      | 3.36 |
| Others        | 1.2  |
| % Saturated   | 33   |
| % Unsaturated | 67   |

Table II shows the major long chain fatty acids present in the *J curcas* seed oil; they are:oleic, linoleic, myristoleic, caprylic, capric, lauric and myristic acids.*Jatropha curcas* seed oil contains high percentage of unsaturated fatty acid. The presence of high unsaturated oleic and linoleic acid makes the *Jatropha curcas* oil prone to oxidation during storage (Arellano *et al.*, 2016).The free fatty acid(FFA) of the oil was found to be 4.26%. The value of the free fatty acid obtained in this study is similar to the report ofJain & Sharma (2011)

### 3.3. Regression model and statistical analysis

The coded variables (-1,+1) and their two-level-four factor design,the result for the experimental biodiesel yield, predicted biodiesel yield and residual values are presented in Table III.

**Table III: Experimental Design, Predicted and Residual for the 2<sup>4</sup>Factorial Design**

| Run | X <sub>1</sub> | X <sub>2</sub> | X <sub>3</sub> | X <sub>4</sub> | Experimental Predicted Residual Value(%) | Biodiesel |
|-----|----------------|----------------|----------------|----------------|--|-----------|
|     | (h)            | %(w/v)         |                | °C             |  |           |
|     | Yield (%)      | Yield (%)      |                |                |  |           |
| 1   | -1 (1.5)       | -1(1)          | -1(3:1)        | -1(50)         | 93.00                                    |           |
|     | 92.65          | 0.35           |                |                |  |           |
| 2   | -1(1.5)        | -1(1)          | -1(3:1)        | +1(70)         | 87.26                                    |           |
|     | 89.15          | -1.89          |                |                |  |           |
| 3   | -1(1.5)        | -1(1)          | +1(9:1)        | -1(50)         | 65.00                                    |           |
|     | 65.48          | 0.52           |                |                |  |           |
| 4   | -1(1.5)        | -1(1)          | +(9:1)         | +(70)          | 92.25                                    |           |
|     | 90.73          | 1.77           |                |                |  |           |

|       |         |       |         |        |       |
|-------|---------|-------|---------|--------|-------|
| 5     | -1(1.5) | +(3)  | -1(3:1) | -1(50) | 81.26 |
| 81.61 |         | -0.35 |         |        |       |
| 6     | -1(1.5) | +(3)  | -1(3:1) | +(70)  | 80.00 |
| 78.11 |         | 1.89  |         |        |       |
| 7     | -1(1.5) | +(3)  | +(9:1)  | -1(50) | 65.00 |
| 65.52 |         | -0.52 |         |        |       |
| 8     | -1(1.5) | +(3)  | +(9:1)  | +(70)  | 90.00 |
| 91.77 |         | -1.77 |         |        |       |
| 9     | +1(2.5) | -1(1) | -1(3:1) | -1(50) | 97.25 |
| 95.39 |         | 1.86  |         |        |       |
| 10    | +1(2.5) | -1(1) | -1(3:1) | +1(70) | 98.00 |
| 98.32 |         | 0.32  |         |        |       |
| 11    | +1(2.5) | -1(1) | +1(9:1) | -1(50) | 75.00 |
| 76.98 |         | -1.98 |         |        |       |
| 12    | +1(2.5) | -1(1) | +1(9:1) | +1(70) | 90.85 |
| 91.16 |         | -0.31 |         |        |       |
| 13    | +1(2.5) | +1(3) | -1(3:1) | -1(50) | 82.50 |
| 84.36 |         | -1.86 |         |        |       |
| 14    | +1(2.5) | +1(3) | -1(3:1) | +1(70) | 87.60 |
| 87.28 |         | 0.32  |         |        |       |
| 15    | +1(2.5) | +1(3) | +1(9:1) | +1(70) | 80.00 |
| 98.02 |         | 1.98  |         |        |       |
| 16    | +1(2.5) | +1(3) | +(9:1)  | +1(70) | 92.25 |
| 92.19 |         | 0.31  |         |        |       |

$X_1$  – Reaction time;  $X_2$  – Catalyst concentration;  $X_3$  – Methanol to oil molar ratio;  $X_4$  - Temperature

The actual values are the obtained response data from experimental runs while the predicted values were generated by using the approximation functions based on the linear mode. The model was demonstrated to cover the experimental range of studies adequately (Jaliliannosrati et al., 2013). The results showed that the experimental biodiesel yield ranged from 65% to 95%. The results of the regression analysis are presented in Table IV.

**Table 4: Analysis of Variance (ANOVA) for Model Regression of 2<sup>4</sup> Design**

| Source      | Sum of Squares | df                   | Mean Square        |
|-------------|----------------|----------------------|--------------------|
| F-Value     | P-Value        | Coefficient Estimate |                    |
| Model       | 1507.26        | 9                    | 167.47             |
| 34.28       | 0.0002         | 84.86                |                    |
| $X_1$       | 397.01         | 1                    | 397.01             |
| 81.27       | <0.0001        | 4.98                 |                    |
| $X_2$       | 196.14         | 1                    | 196.14             |
| 40.15       | 0.0007         | -3.50                |                    |
| $X_3$       | 100.00         | 1                    | 100.00             |
| 20.47       | 0.0040         | -2.50                |                    |
| $X_4$       | 154.26         | 1                    | 154.26             |
| 31.58       | 0.0014         | 3.10                 |                    |
| $X_1X_2$    | 420.25         | 1                    | 420.25             |
| 86.03       | <0.0001        | 5.13                 |                    |
| $X_1X_4$    | 7.98           | 1                    | 7.98               |
| 1.63        | 0.2484         | -0.71                |                    |
| $X_2X_3$    | 145.81         | 1                    | 145.81             |
| 29.83       | 0.0016         | 3.02                 |                    |
| $X_2X_4$    | 0.26           | 1                    | 0.26               |
| 0.052       | 0.8269         | 0.13                 |                    |
| $X_1X_2X_4$ | 85.56          | 1                    | 85.56              |
| 17.52       | 0.0058         | -2.31                |                    |
| Residual    | 29.31          | 6                    |                    |
| 4.88        |                |                      |                    |
| Lack of Fit | 13.49          | 5                    | 8.65               |
| 2.05        | 0.3239         |                      |                    |
| Pure Error  | 54.26          | 4                    | 2.86               |
| Cor total   | 1536.57        | 15                   | R-Squared          |
| 0.9809      |                |                      |                    |
| Std dev     | 2.21           |                      | Adjusted R-Squared |
| Squared     | 0.9523         |                      |                    |
| Mean        | 84.86          |                      | Pred R-Squared     |
| 0.8644      |                |                      |                    |
| CV          | 2.60           |                      | Adeq Precision     |
| 19.366      |                |                      |                    |

$X_1$  – Reaction time;  $X_2$  – Catalyst concentration;  $X_3$  – Methanol to oil molar ratio;  $X_4$  - Temperature

The four independent variables ( $X_1, X_2, X_3$  and  $X_4$ ) and two interactive terms:  $X_1X_2$ ,  $X_2X_3$  were significant at the 95% confidence level. large  $F$ -values and low  $p$ -values expressed the significant ( $p < 0.05$ ) attribute of the four independent

variables in the biodiesel. However, Time  $X_1$  ( $F=81.27$ ,  $p<0.0001$ ) was the most significant model term. To minimize error, all the coefficients were considered in the design. Analysis of variance (ANOVA) for the response model was used to justify the adequacy of the models (Jaliliannosrati et al., 2013). The coefficient of the linear response surface model fitting are presented in Table IV. The model  $F$ -value of 34.28 with  $p=0.0002$  showed that the regression model was significant. The high value of  $R^2$  (0.9809) and non significant lack- of- fit (0.3239) showed that there were adequate correlation between the independent variables; and the fitted model can sufficiently describe the effect of the independent variables on the dependent variables. The  $R^2$  describes the ratio of the explained variation to the total variation, a measure of the degree of fit (Fasusn et al., 2018). Thus,  $R^2$  indicated that 98.09% of the variability in the data accounted for the model. The adjusted  $R^2$  is a corrected value for  $R^2$  after the elimination of non significant model terms. The absence of multiple non-significant terms in a model would increase the value of adjusted  $R^2$  appreciably (Stamenkovic et al., 2002). The adjusted  $R^2$  (0.9523) supports the high significance and adequacy of the model. Hence, the model can be used in theoretical production of the biodiesel yield. The mathematical model for the yield of biodiesel are presented in equation (4)

$$Y(\%)=84.86+4.98X_1-3.50X_2-2.50X_3+5.13X_1X_2-0.71X_1X_4+3.02X_2X_3+0.136X_2X_4-2.31X_1X_2X_4$$

(4)

The positive sign in before the terms signifies synergistic effect while negative sign reveals antagonistic effect that would indicate the influence of independent variables on the process. In statistics, a studentized residual is the proportion resulting from the division of a residual by an estimate of its standard deviation (Jaliliannosrati et al., 2013). The studentized residual predicted by the best fit normal distribution was plotted against the experimentally obtained studentized residual in Figure 1(a) and straight line formed shows that the studentized residuals follow a normal distribution (Korbahti & Rauf., 2008). In cases where the residuals do not follow a normal distribution, an S-shape curve is often formed and this type of curve often results from the use of an incorrect model or if an additional transformation of the response is necessary (Noshadi et al., 2012). Figure 1(b) is the plot of the studentized residuals versus predicted biodiesel yield. The plot is expected to be a random scatter, indicating that the variation in the original observations is not related to the value of the response and the suggested model is an appropriate description of the Process (Noshadi et al., 2012). Figure 1(c) is the plot of the actual versus the

biodiesel yield. Actual values are data for each specific run from Table III and predicted values were produced by the model equation (3). In experimental design,  $R^2$  is a calculation of amount of variation around the mean described by the model. However, a large value of  $R^2$  can be misleading if the model contains extraneous terms (Montgomery, 1996). Generally, the adjusted  $R^2$  value does not increase as factors are added to the model. Large differences between  $R^2$  and Adj.  $R^2$  indicate that non significant terms are involved (Myers & Montgomery., 1996). The data in Figure 1 (c) leads to values of  $R^2$  and Adj.  $R^2$  of 0.9809 and 0.9523, respectively. In addition, the Adj.  $R^2$  was very high to prove the high significance of the model. In Figure 1(d), the outlier plot for all runs of the biodiesel production is shown. The outlier  $t$  plot simply indicates the magnitude of the residuals for each run to determine if any of the runs had particular large residuals ((Noshadi et al., 2012). Typically, a threshold of three standard deviations is employed as a definition of an outlier (Noshadi et al., 2012). Most of the standard residuals should lie in the interval of  $\pm 3.00$ . Any observation outside this interval indicates a potential error in the model or an operational error in the experimental data (Noshadi et al., 2012). In figure 1(d), there was no data beyond the interval 3, which reveals that the fitted model is consistent with all the data and there was no recording error.

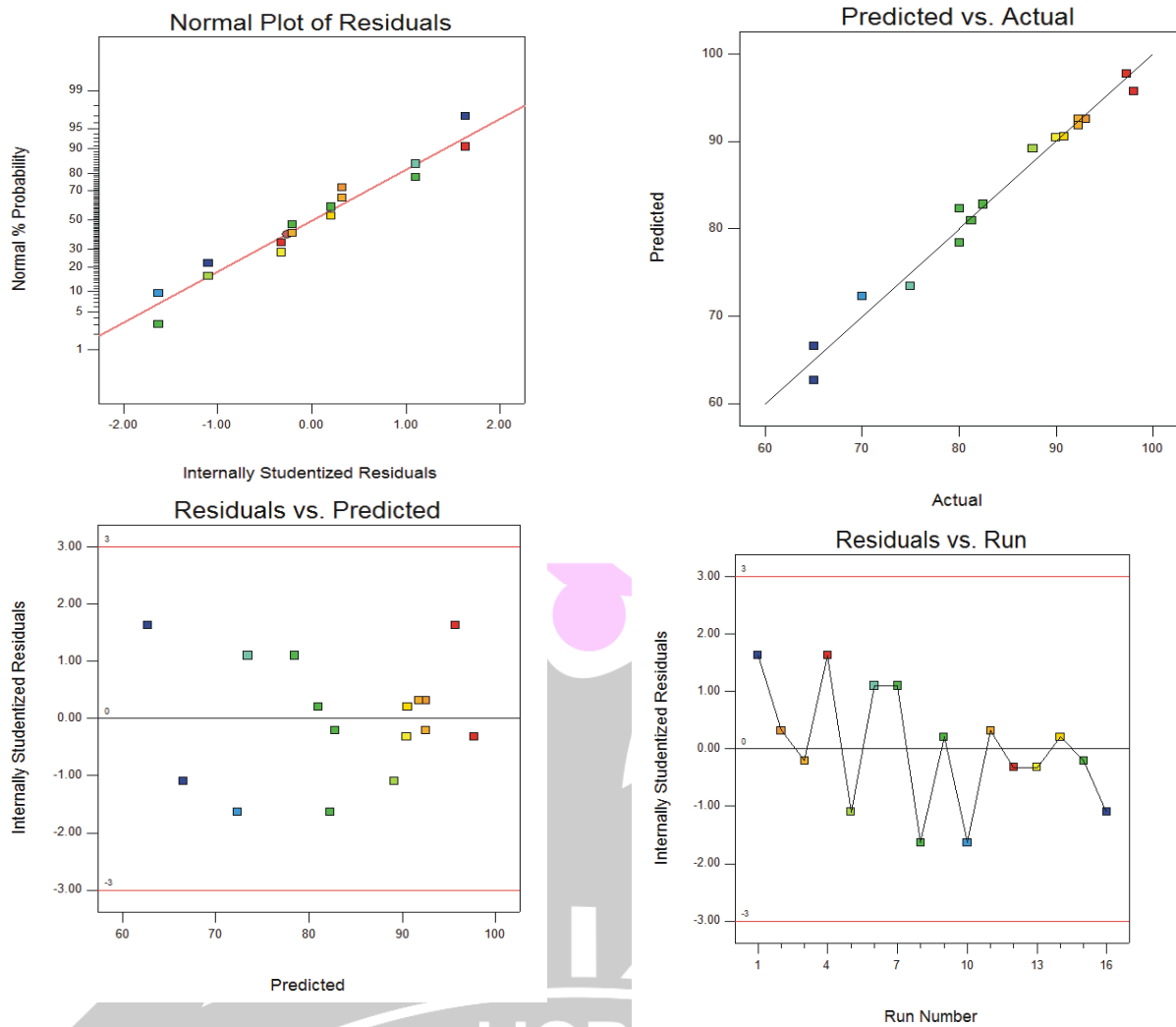


Figure 1: (a) The studentized residuals and (b) Studentized residuals and predicted response plots normal % probability plot

Figure 1(c) Predicted values against experimental values 1 (d) Outlier  $t$  plots

### 3.4 Interaction of process parameters and optimization

The interaction between the parameters were demonstrated clearly by plotting two variables with the biodiesel yield on a three dimensional surface and a contour (two dimensional) plot. Since the model has more than two variables, four plots were created, each with two targeted variables while the remainder of the variables were held constant (Jaliliannosrati et al., 2013). Figure 2 (a) exhibits the contour plots of the effect of time (h), methanol to oil ratio and their reciprocal interaction on the biodiesel yield at constant methanol to oil ratio and catalyst. Increase in reaction time and increase in temperature increase the yield of biodiesel. Fig 2(b) contour plots representing the effect of methanol to oil ratio and catalyst and their effect on biodiesel yield at constant time and temperature. Increase in methanol to oil ratio and decrease in catalyst favors the biodiesel yield. In figure 2(c), the effect of catalyst, temperature and their reciprocal interaction

on the yield of biodiesel yield at constant reaction time and methanol to oil ratio is shown. From the figure, decrease in catalyst and increase in temperature increases the yield of biodiesel. Figure 2(d) exhibits the effect of time, catalyst and their reciprocal interaction on the biodiesel produced at constant temperature increase the yield while time seem to have more influence on the biodiesel yield.

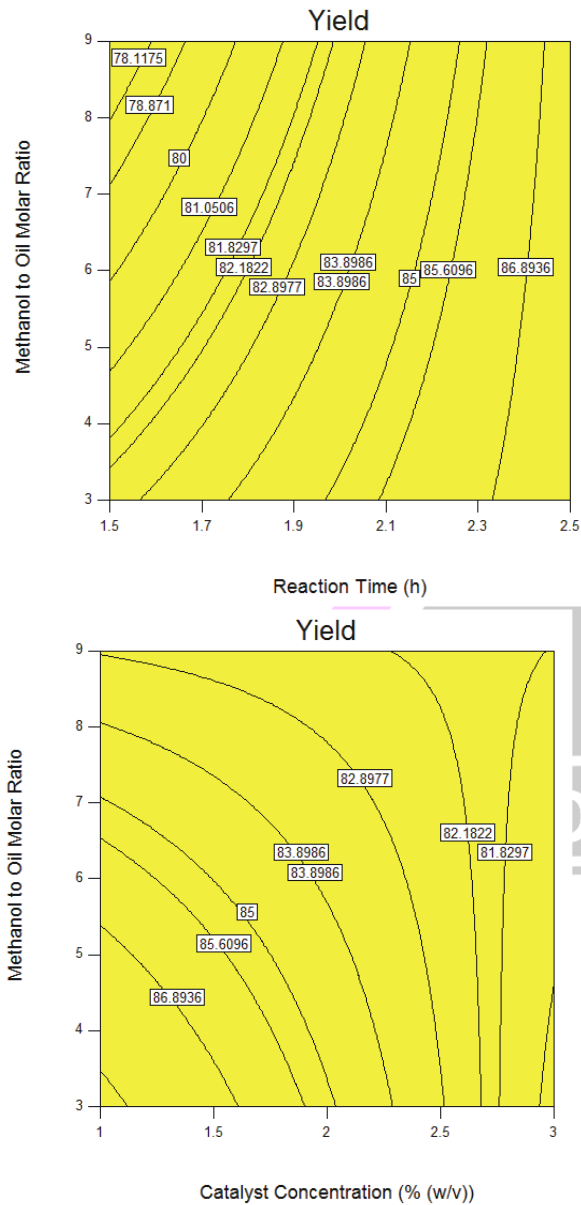


Figure 2(a) and Figure 2(b)

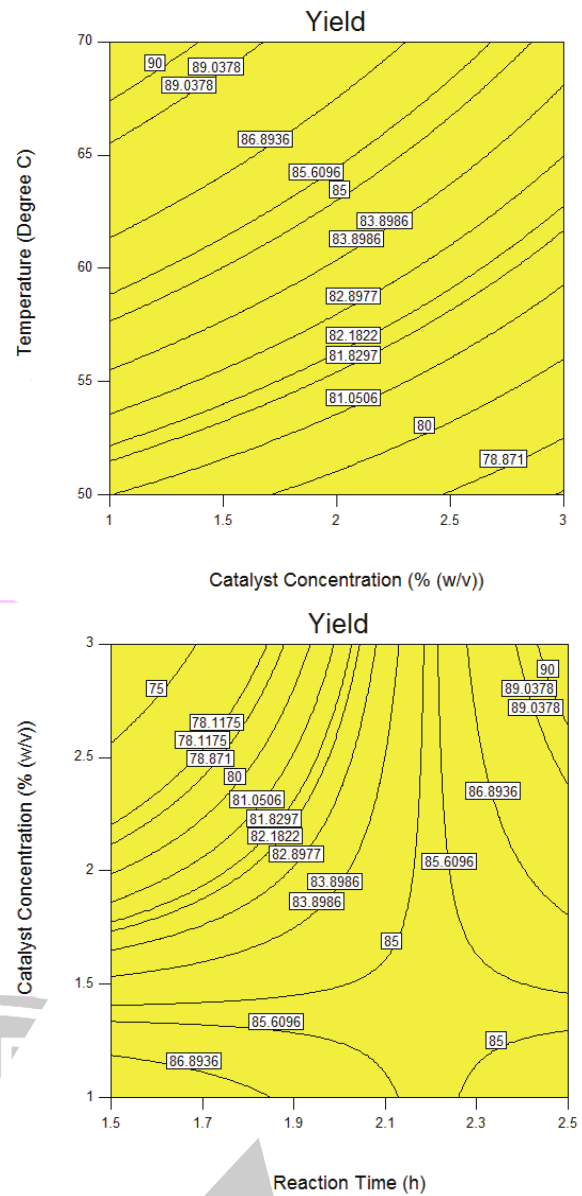


Figure 2(c) and Figure 2(d)

Figure 2: Response surface contourplots for optimization of biodiesel production from *J. curcas* oil

### 3.5 Optimization process

In the optimization process, the limits (lower and upper) of each process parameters (reaction time, catalyst concentration, methanol to oil ratio and temperature) and its response (biodiesel yield) provided by the surface and contour plots are used in the optimization procedure. The optimal conditions of the two step method for biodiesel production from *J. curcas* oil with high free fatty acid gave a yield of 98.3% at reaction time (2.50h), catalyst concentration (1%, w/v), methanol to oil ratio (3:1) and temperature (70°C). From the optimal conditions stated above, the JCO conversion to



biodiesel which gave a yield of 98.3% confirmed the efficiency of the RSM model used in this study.

#### **4. CONCLUSION**

Statistical approach to the optimization of biodiesel produced from *Jatropha curcas* oil seeds by two step reactive extraction was carried out to determine the interaction of various factors and their effect on the yield of biodiesel. Transesterification of the pretreated JCO using KOH as catalyst was successfully optimized by RSM. The RSM predicted that the optimal condition for the transesterification was time (2.5h), catalyst concentration (1%, w/v), methanol to oil ratio (3:1) and temperature (70°C) with actual biodiesel yield of 98.3%. This study clearly showed that response surface methodology was a suitable method of optimizing the operating conditions in order to maximize the biodiesel production. Graphical response was used to locate the optimum point. A full factorial  $2^4$  design (16 assays) was successfully employed for experimental biodiesel and results analysis. Satisfactory prediction equation was derived for the biodiesel yield using RSM.

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#### **CONFLICT OF INTEREST**

The authors have declared no conflicts of interest for this article.