Optimization Process of Parameters for the Transesterification of *Jatropha curcas* Seed Oil using Response Surface Methodology (RSM)

Abbas Abubakar[1*], Auwal A. Mahmoud[1], Dahiru A. Ajiya[1], Umar F. Hassan[1], Khuzaifa Y. Muhammad[1], Sha'aban Sallau[2] and Salim Yushau[1], Abubakar H.I[1]

[1.] *Department of Chemistry Abubakar Tafawa Balewa University Bauchi, Nigeria.*
[2.] *Department of Pure and Applied Chemistry, Bayero University Kano, Kano State, Nigeria.*
**Corresponding author*

Abstract:- **Biodiesel comprises of monoalkyl esters of long chain fatty acids. It is produced using edible oil, non-edible oil and animal fats by acid or base catalyzed transesterification with ethanol or methanol. In this research work, the oil was extracted using n-hexane. Response surface Methodology (RSM) with central composite design (CCD) was applied to optimize the biodiesel production from seed oil. An optimum biodiesel yield of 98.32 % was obtained by transesterifying *Jatropha* seed oil with 0.30 g of catalyst and ethanol to oil molar ratio of 12:1 at 65 ^{o}C for 2 hrs. The experimental yield is in good agreement with the predicted yield, with relatively small percentage error (0.58 %). This shows that the proposed statistical model is suitable for prediction of optimized biodiesel yield and for optimization of transesterification process.**

Keywords: **Jatropha curcas, Biodiesel, Response Surface Methods, Optimization, Seed Oil.**

I. INTRODUCTION

A great emphasis has been placed on global warming, environmental pollution and the ever- depleting fossil fuel resources in this climate- sensitive period. These have become major global issues and various methods have been suggested to curtail the undesired effects of fossil fuel emission (Moyo *et al.,* 2020). Biodiesel is a clean and renewable fuel which is considered to be the best substitution for diesel fuel (Singh and Singh, 2010; Latchubugata *et al.*, 2018; Mahmoud *et al.*, 2020). Currently, Biodiesel is a less-toxic, biodegradable fuel from renewable sources, which is an important alternative for fossil fuel for use in a sustainable approach involving economic and environmentally friendly aspects (Fant *et al.*, 2011; Yunus *et al.*, 2013; Abubakar *et al.*, 2020). It is used as an alternative for diesel fuel in the automotive industry, commonly known as No. 2 diesel. The advantage of this biofuel over the conventional diesel fuel are high cetane number, low smoke and particulates, low carbon dioxide and hydrocarbon emissions (Encinar, 2007; Kumar and Kant, 2013). It reduces emissions of carbon (II) oxide, unburned hydrocarbons and smoke. On the other hand, vegetable oil has high density, high viscosity, lower calorific value and poor non-volatility, which leads to atomization problem, pumping problem and poor combustion inside the combustion chamber of a diesel engine. In case of long-term use of vegetable oils in diesel engines, problems such as gumming, injector fouling, piston ring sticking and contamination of lubricating oils are bound to occur. All these problems mentioned are due to the high viscosity of vegetable oils. It is therefore necessary to find a means of reducing the viscosity of vegetable oil to a more approximate value of diesel. The approaches such as preheating the oils, blending them with diesel, thermal cracking and transesterification are the solutions to the problems (Pramanik, 2003; Knothe and Steidley, 2007; Kalpana *et al.*, 2019).

Biodiesel comprises of monoalkyl esters of long chain fatty acids. It is produced using edible oil, non-edible oil and animal fats by acid or by base catalyzed transesterification with ethanol or methanol. Significant efforts have been made for obtaining biodiesel by transesterification of oil obtained from Jatropha curcas, soybean, sunflower, cotton seed, rapeseed and palm oils (Abubakar *et al.*, 2020)

II. MATERIALS AND METHOD

2.1 Materials / Equipment

The equipment used for this experiment include NYC12 muffle furnace, Soxhlet extractor set-up, rotary shaker (Bio Techno Lab Mumbai India) and water bath HHW420 (B-scientific England), heating mantle and Reflux condenser.

2.2 Chemicals and Reagents

All the reagents/chemicals that were used in this research were of analytical grade.

2.3 Sample Collection and Preparation

Fresh seeds of Jatropha curcas was collected from Railway Quarters, Bauchi, Bauchi State and was identified at the Department of Biological Sciences, Abubakar Tafawa Balewa University, Bauchi. Samples were washed with distilled water 3 times, dried under shade and stored for further use.

Fig. 1 Jatropha seed.

Fig. 2 Extracted Oil

2.4 Soxhlet Extraction

Jatropha curcas seeds were cracked and the shells carefully removed. The kernels obtained was used for oil extraction. The method described by Mahmoud *et al.,* 2019 with a little modification was used. The seed kernels were ground using mechanical method and defatted in Soxhlet apparatus. The extraction was carried out using N-hexane. The process was continued for 6 hrs. Solvent was removed by vacuum evaporation and exposure to heat in drying oven at 50 °C. The amount of oil recovered was calculated as percentage of total oil present in Jatropha curcas seed kernels. Each extraction was run in triplicate and the final value which is the average of all. Extracted seed oil was stored in freezer at -20 °C for subsequent physicochemical analyses.

Fig. 3: Soxhlet extraction Set-up

Fig. 4: Biodiesel produced

2.5 Experimental Design

Response surface methodology (RSM) with central composite design (CCD) was applied to optimize the biodiesel production from seed oil. In this research, three independent parameters were evaluated (reaction time, catalysts loading and methanol to oil molar ratio) whilst the dependent variable was fatty acid methyl esters (FAME) yield or biodiesel yield. The range and levels of the independent variables for transesterification process was determined (Lee *et al.,* 2020).

2.6 Model Fitting and Statistical Analysism

Design Expert software version 7.0. (STAT-EASE Inc., Minneapolis, USA) was employed for regression analysis of the experimental data to fit the equations. The quality of the developed model was determined from the value of correlation (R^2), while evaluation of the statistical significance of the equations developed was determined using an analysis of variance (ANOVA).

2.7 Preparation of $CaTiO_3$ Catalyst using CaO from Calcined Bone

$CaTiO_3$ catalyst was prepared: calcined bone (CaO) and TiO_2 in a molar ratio of 1:1 (20.0 g $CaCO_3$ and 20.0 g TiO_2) were milled in a mortar for 10- 15 min. The mixture was calcined at 550 ^oC for 3 h and at 1050 ^oC for 3.5 h to obtain $CaTiO_3$ as white solid which was stored in a dried container and kept in a desiccator.

2.8 Production of Biodiesel Fuel

During biodiesel production process, two phases were used: Trans-esterification phase as well as separation and washing phase

2.8.1 Trans-Esterification Phase

Reactions in the trans-esterification phase involves glyceride (oil) and alcohol in the presence of a catalyst. Biodiesel was produced from crude *Jatropha curcas* seed oil (triglycerides) by transesterification with ethanol in the presence of $CaTiO_3$ catalyst. This process was optimized by applying response surface methodology (RSM). A second-order mathematical model was used to predict the yield as a function of ethanol/oil molar ratio, catalyst concentration and reaction time.

III. RESULTS AND DISCUSSION

3.1 Results

Table 1: The Levels and Range of Independent Variables of Transesterification Process

Variables				**Levels**			
	Coding	**Unit**	**-α**	**-1**	**0**	**1**	**+ α**
Catalyst	A	G	0.04	0.10	0.3	0.50	0.64
Molar ratio	B	M	6.95	9	12	15	17.02
Time	C	Hr	0.32	1	2	3	3.68

Table 2: Design Matrix of Experiments and their Respective Experimental and Predicted Yields

Run	**A: Catalyst(g)**	**B:molar ratio (M)**	**C: Time(hr)**	**Experimental yield (%)**	**Predicted Yield (%)**
1	0.10	9.00	1.00	97.00	96.65
2	0.50	9.00	1.00	96.00	96.09
3	0.10	15.00	1.00	96.10	95.69
4	0.50	15.00	1.00	97.00	97.20
5	0.10	9.00	3.00	96.00	95.62
6	0.50	9.00	3.00	95.75	95.98
7	0.10	15.00	3.00	93.50	93.24
8	0.50	15.00	3.00	95.50	95.68
9	0.04	12.00	2.00	94.00	95.07
10	0.64	12.00	2.00	95.20	94.79
11	0.30	6.95	2.00	97.00	97.16
12	0.30	17.05	2.00	96.00	96.06
13	0.30	12.00	0.32	98.00	98.20
14	0.30	12.00	3.68	96.00	96.05
15	0.30	12.00	2.00	98.50	98.36
16	0.30	12.00	2.00	98.70	98.36
17	0.30	12.00	2.00	98.60	98.36
18	0.30	12.00	2.00	98.80	98.36
19	0.30	12.00	2.00	98.00	98.36
20	0.30	12.00	2.00	98.00	98.36

Table 3: Sequential Model Sum of Squares

Source	**Some of square**	**Degree of freedom**	**Mean square**	**F-value**	**p-value Prob> F**	
Mean vs. Total	1.870 E + 005	1	1.870E +005			
Linear vs. Mean	7.51	3	2.50	1.07	0.3906	
2FI vs. Linear	3.60	3	1.20	0.46	0.7154	
Quadratic vs. 2FI	31.67	3	10.42	39.20	<0.000 1	Suggested
Cubic vs. Quadratic	2.05	5	0.41	3.33	0.1069	Aliased
Residual	0.61	5	0.12			
Total	1.870 E+5	20	9349.7 6			

"Sequential Model Sum of Squares ": Select the highest order polynomial where the additional terms are significant and the model is not aliased.

Table 4: Interaction Variene Effects

Source	**Sum of square**	**Degree of freedom**	**Mean square**	**F-value**	**P-value Prob> F**	
Model	42.38	9	4.71	17.71	<0.0001	Significant
A-catalyst	2.65	1	2.65	9.97	0.0102	
B-molar ratio	1.37	1	1.37	5.17	0.0463	
C- Time	5.56	1	5.56	20.91	0.0010	
AB	2.15	1	2.15	8.10	0.0174	
AC	0.43	1	0.43	1.61	0.2334	
BC	1.02	1	1.02	3.82	0.0792	
A^2	25.30	1	25.30	95.17	<0.0001	
B^2	5.46	1	5.46	20.53	0.0011	
C^2	2.76	1	2.76	10.40	0.0091	
Residual	2.66	10	0.27			
Lack of fit	2.05	5		3.33	0.1061	Not significant
Pure Error	0.61	5	0.12			
Cor Total	45.04	19				

Table 5: Optimization Criteria for Transesterification Process

Factors	**Goal**	**Lower limit**	**Upper limit**
Molar ratio	Minimize	9	15
Time	Minimize	1	3
Catalyst	Minimize	0.1	0.5
Biodiesel	Maximize	93.50	98.80

Table 6: Validation Model at the Optimum Conditions.

Catalyst (g)	Molar ratio (M)	Time (hr)	Experimental yield (%)	Predicted yield	Percentage error (%)
0.30	12	2	98.80	98.36	0.58

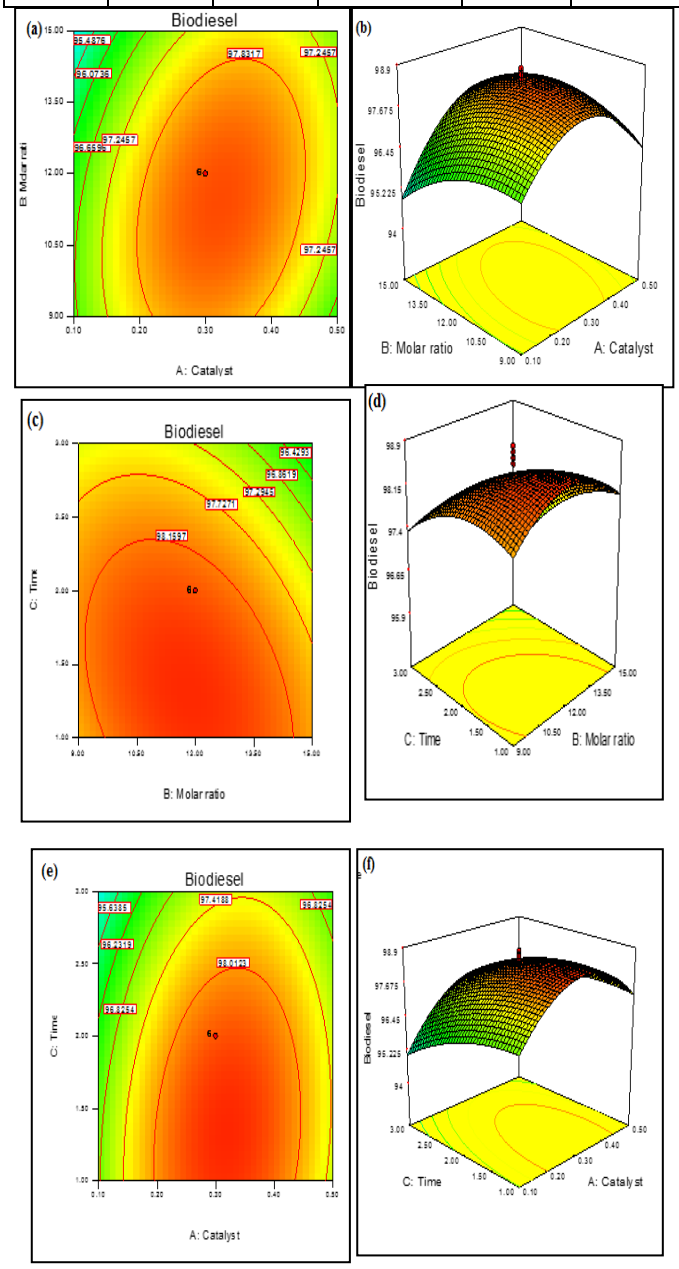

Figure 5: Counter plot and 3D of experiment.

Table 6: Physicochemical properties of Jatropha curcas L. seed oil

Oil Quality Parameters	***Jatropha curcas* L. seed oil**	**ASTMD Standard**
Moisture content (%)	2.60	-
Oil yield (%)	65.00	-
Colour	Brownish-black	-
Viscosity (mm^2/s)	22.80 ± 0.03	35.00
Density (g/mL)	0.88 ± 0.02	0.86 – 0.90
Cloud point	4.56 ± 0.01	– 3.00 – 12.00
Pour point	1.83 ± **0. 01**	– 15.00 – 10.00
Flash point	155.00 ± 0.02	130.00 – 170.00
Acid value mgKOH/g	13.76 ± 0.02	0.00 – 0.80
Saponification value mgKOH/g	212.67 ± 0.01	189.00 – 198.00
Iodine value $gI_2/100g$	86.88 ± 0.04	≥ 130.00
Peroxide value mEq/kg	6.55 ±0.01	2.00-10.00
Specific gravity	0.89 ± 0.02	0.92
Free fatty acid (mgKOH/g)	4.76 ±0.12	25.00

Values are mean ± standard deviation (n=3)

Table 7: Functional groups present in *Jatropha curcas* L biodiesel

Wave number (cm^{-1})	**Types of vibration**	**Nature of functional group**
3004.280	CH=C-H	Stretching of methyl ester
2922.520	C-H (stretching)	Alkanes
2853.210	C-H (stretching)	Alkanes
1724.140	C=O (stretching)	Esters
1435.320	C-H (bending)	Alkane
1462.140	C-H (bending)	Alkanes
1195.500	C-O (stretching/ bending)	Esters
1169.520	C –O (stretching)	esters
1016.060	C-O-C antisymmetric	Alcohols, ethers esters
850.880	RHC=CHR	Methylene group
722.350	R_2CH_2	Alkenes

3.2 Discussion

In this research work, 20 sets of experiments were generated which include 23 factorial experiments, 6 axial points and 6 replicates of centre points.

Each response obtained from the transesterification process was used to develop a mathematical model that correlates the biodiesel yield with the independent reaction variables via second-order polynomial equation as given below (Wong *et al.*, 2015).

$$\boldsymbol{Y = b_o \sum_{i=1}^{n} b_i x_i + \sum_{i=1}^{n} b_o x_i^2 + \sum_{i=1}^{n=1} \sum_{j=i+1}^{n} b_o x_i x_j} \quad 1$$

Where Y is the predicted biodiesel yield, bo is the constant coefficient, bi is the linear coefficients, bij is the interaction coefficients, bii is the quadratic coefficients and xi, while xj are the coded values of the experimental variables.

3.3 Model Fitting and Statistical Analysis

Design Expert software version 7.0. (STAT-EASE Inc., Minneapolis, USA) was employed for regression analysis of the experimental data to fit the equations. The quality of the developed model can be determined from the value of correlation (R^2), while evaluation of the statistical significance of the equations developed can be determined using analysis of variance (ANOVA).

3.4 Development of Experimental Regression Model

The complete design matrix of the experiments attached with the experimental yield and predicted yield is shown in the Table 2. From the Table, it can be seen that the biodiesel yield found is in the range of 93.50 to 98.80 %.

The RSM software produced a series of models (linear, two factor interaction (2FI), quadratic and cubic polynomial) that fitted to the response as well as recommend the best fitted model as shown in Table 3. According to the sequential model sum of square, the best model to fit the response is quadratic model owing to its highest order polynomial with significance of additional terms and the model was not aliased. The final equation in terms of actual factor for the biodiesel production was calculated below using the equation 2.

$$\text{Biodiesel} = +86.13722 +12.73807A +1.50759B +2.18577C +0.86458AB +1.15625AC -.11875BC -38.47693A^2 - 0.068133B^2 -0.43642C^2 \quad \text{----------2}$$

Where the coded terms A, B and C represent the concentration of catalyst, ethanol to oil molar ratio and reaction time respectively. Positive sign in front of the terms indicates synergic effect, while negative sign indicates antagonistic effect (Shuit *et al.,* 2010). The terms A, B, C, AC and AB therefore, play an important role in increasing the biodiesel yield, whilst the other terms A^2, BC, B^2 and C^2 play important role in decreasing the biodiesel concentration.

3.5. ANOVA

Analysis of variance was further carried out to determine the significance and the fitness of the linear model. The ANOVA for the response surface linear model was presented in Table 4. The Model F-value of 17.71 implies that the model is significant. There is only a 0.01% chance that a "Model F-Value" this large could occur due to noise. Values of "Prob > F" less than 0.0500 indicate that the model terms are significant at 95 % confidence level. In this case A, B, C, AB, A^2, B^2 and C^2 are significant model terms. Values greater than 0.1000 indicate the model terms are not significant. If there are many insignificant model terms, model reduction may improve the model. The "Lack of Fit F-value" of 3.33 implies that the Lack of Fit is not significant relative to the pure error. Non-significant lack of fit is considered good.

3.6 Study of parameters

Figures 1a and 1b display the contour plot and the three dimensional (3D) plot for the interaction effect between A (Catalyst) and B (molar ratio). The reaction temperature and time were kept constant at 65 ^oC and 2 hr respectively, throughout the experiments. The contour plot Figure 1a shows that higher biodiesel yield (> 97 %) was obtained between low and intermediate molar ratio (9 to 12 M) and between the intermediate and low catalyst mass (0.225 to 0.39 g). The 3D respond surface showed that the biodiesel yield increased with increasing catalyst mass and decreasing molar ratio. Increase in conversion of the oil to biodiesel was perceived with increased mass of catalyst and decreased with increased molar ratio. At 0.30 g catalyst and 9.00 molar ratio, a yield of 97.83 % biodiesel was achieved. Increasing the molar ratio from 9 to 15 molar ratio reduced the biodiesel conversion to about 95 %. This implies that ethanol has an inverse relation with the biodiesel conversion. Excess ethanol led to difficulty in separation because it increases the glycerol solubility in product phase (Hamza *et al.,* 2015). With an increasing catalyst mass beyond 0.30 g, a decreased conversion was found due to meagre diffusion among oil, ethanol and solid catalyst phases which is based on excess mass of catalyst. A similar trend was also reported with CaO derived from chicken and ostrich egg shells for biodiesel production from WCO ratio (Tan *et al.,* 2017).

The contour plot and the three dimensional (3D) plot for the interaction effect between B (molar ratio) and C (time) is shown in Figures 1c and 1d. The reaction temperature and catalyst mass were kept constant at 65 ^oC and 0.30 g respectively throughout the experiment. The contour plot 1c shows that higher biodiesel yield (> 98 %) was obtained at a minimum molar ratio (9.5 molar ratio) and reaction time (1 hr). The 3D respond surface show that the biodiesel yield increased with decreasing reaction time and molar ratio. Increase in ethanol decreased the biodiesel conversion, which clearly shows that ethanol has greater impact on biodiesel conversion. Excess ethanol has negative effects on biodiesel conversion and this agrees with the findings of Lee *et al.,* (2011).

The contour plot and the three dimensional (3D) plot for the interaction effect between A (catalyst) and C (time) is depicted in Figures 1e and 1f. The reaction temperature and molar ratio were kept constant at 65 ^oC and 12 molar respectively throughout the experiment. The counter plot 1e shows that higher biodiesel yield (> 98 %) was obtained between intermediate and high mass of catalyst (0.19 to 0.45) and at low values of time (1 to 0.5 hr). The 3D respond surface shows that the biodiesel yield increased with decrease in the reaction time and catalyst.

At 1 hr reaction time and 0.10 g catalyst mass, the biodiesel conversion was 96 %, when the time was increased to 3 hrs, a

minor decline was observed (95 %). Increase in catalyst mass beyond 0.45 g and maintaining reaction time at 2 hr, yielded biodiesel conversion of 96 %. However, increasing the time resulted in decreased biodiesel conversion of 95 %. At 1 hr and 0.50 g catalyst, a biodiesel conversion of 96 % was achieved. It was also observed that between 0.20 and 0.40 g catalyst and 1 hr reaction time 98 % biodiesel conversion was obtained. This indicates that excess reaction time resulted in decreased biodiesel yield.

In the present experiment, biodiesel yield was set to maximum value, while the other reaction parameters were set at a minimum values (Table 5). The experimental conditions with the highest predicted biodiesel yield were selected for further validation. The result of model validation is shown in Table 6. An optimum biodiesel yield of 98.32 % was obtained by transesterifying *Jatropha* seed oil with 0.30 g of catalyst and ethanol to oil molar ratio of 12 at 65 ^{o}C for 2 h. The experimental yield is in good agreement with the predicted yield with relatively small percentage error (0.58 %). This shows that the proposed statistical model is suitable for prediction of optimized biodiesel yield and for optimization of transesterification process.

The results of the physicochemical characteristics of Jatropha curcas L. seed oil were given in Table 6. The oil is Brownish-black in colour and has a viscosity of (22.80 mm^2/s) at room temperature it was considered as less viscous when compared to Jatropha curcas seed oil (32.00 mm^2/s), Jatropha. podagrica (36.47 mm^2/s)(Premjet *et al.*, 2021), Malaysian Jatropha, Indonesian Jatropha and Thailanian Jatropha (47.50 mm^2/s 53.94 mm^2/s and 39.20 mm^2/s) respectively (Emil et al., 2010), Bebra seed oil (40.59 mm^2/s), cashew nut oil (56.00 mm^2/s) (Andualem and Gassesse, 2014), but it was in turn more viscous than Terminalia mantaly seed oil (4.80 mm^2/s) Jatropha curcas seed oil (20.55 mm^2/s) (Abubakar *et al.*, 2020) and Tropical almond (14.10 mm^2/s) (Orhevba *et al*., 2016) .

The density of oil in this study was (0.877 g/mL) which is lower than the density of Jatropha podagrica (0.90 g /mL) (Premjet *et al.*, 2021), Tropical almond (0.90 g/cm3) (Orhevba *et al.*, 2016) Terminalia mantaly seed oil (0.92 g/mL) (Abubakar *et al.*, 2020), Jatropha curcas (0.90 g/mL) (Emil *et al.*, 2010)soybean and sunflower (0.91 g/mL and 0.92 g/mL) respectively (Karmakar *et al.*, 2017) and Bebra seed oil (0.94 g/mL) (Akpan and Muhammed, 2007). The specific gravity of oil in this study (0.89) which is lower than that of Sorrel and Okra seed oils (1.00 and 0.90 respectively (Umar *et al*., 2020), Bebra seed oil (0.926), Cashew nut (0.964) (Aremu *et al*., 2006) and Castor seed oil (0.958) (Andualem and Gassesse, 2014) and the same as that of Tropical almond (0.89) (Orhevba *et al.*, 2016).

The percentage oil yield of the seed was (65.70 %) which is almost similar to Terminalia catappa seed oil (63.65 %) (Monnet *et al*., 2012), this value is in turn higher than the percentage oil yields of Z-spinachristi (Nabag) Seed Crude Oil (28.96%), (Saeed and Abbas, 2016). Terminalia Catappa (38 %, melon seed 38.30 % Moringa seed 40.60 %, Cashew seed 49.34 % sesame seed 47.80 %, Bitter kola seed 11.92 %) as reported by (Abdulhamid *et al.*, 2014; Saeed and Shola, 2015; Ebenezer, 2015). Cotton seed (15.0-24.0%), soybean (17.0-21.0%) (Ebenezer *et al.*, 2016), J. curcas L. (46. 31 %) Afzelia africana seed oil (33.32 %) (Ejikeme *et al.*, 2010) J. gossyypiifolia and Jatropha curcas L. (23 % and 31 %) respectively (Jefferson *et al*., 2009), Terminalia belerica R. seed oil (12.28 %) (Hossain *et al*., 2007), mustard seed oil (44.67 %), Canola (43.87 %) Corn (4.23 %) Cotton (17.83 %) (Arif *et al.*, 2012) and is lower than that of M. myristica seed oil (79.93 %) (Ajayi *et al*., 2013) and it is within the A.O.A.C 1990. This high percentage oil yield indicates its potential use in detergent/ soap making industries and edible purposes.

The Saponification value of the oil was found to be (212.67 mgKOH/g) which is higher than that of Bebra seed oil (174.95 mgKOH/g), Terminalia mantaly seed oil (196 mgKOH/g) and some of common oil like castor seed oil (185.83 mgKOH/g), palm oil (196-205 mgKOH/g), ground nut oil (188-196 mgKOH/g), corn oil (187-196 mgKOH/g) (Abubakar *et al.*, 2020), Z-spinachristi (Nabag) Seed Crude Oil (181.39 mgKOH/g) (Saeed and Shola, 2015), Moringa Seed (182.89 mgKOH/g), cashew seed (169.42 mgKOH/g), sesame seed (192.70 mgKOH/g,) water melon seed (192.09 mgKOH/g), (Saeed and Shola, 2015), and also Terminalia Catappa (140.275 mgKOH/g), beeswax (93.0 mgKOH/g) which is commonly used in soap making and lower than Jatropha curcas (216.09 mgKOH/g) (Emil *et al*., 2010), Coconut oil (253 mgKOH/g) and palm kernel oil (247 mgKOH/g) as reported by (Andualem and Gassesse, 2014), bitter kola seed (229.45 mgKOH/g) and melon seed (247.0 mgKOH/g) (Saeed and Shola, 2015), this is within the A.O.A.C. Standard 1990. Oils with lower saponification values contains high amount of long chain fatty acids.

The acid value in this study was (13.76 mgKOH/g) which is higher than that of T. mantaly seed oil (0.56 mgKOH/g), Bebra seed oil (0.052 mgKOH/g), melon seed 0.43 (mgKOH/g), water melon seed (0.51 mgKOH/g) moringa seed (0.51 mgKOH/g), bitter kola 0.06 (Abubakar *et al.*, 2020), Terminalia belerica seed oil (3.69 mgKOH/g, Linseed oil 6.0 mgKOH/g, Sunflower oil 0.5-5.0 mgKOH/g) (Hossain *et al.*, 2007).This value exceeds the limit of AOAC Standard of < 4.0. According to (Aremu *et al*., 2015). High acid value in oil (e.g. luffa gourd) showed that the oil may not be suitable for use in cooking (edibility), but however, be useful for production of paints, biodiesel, liquid soap and shampoos (Aremu *et al.*, 2006).

The peroxide value in this study was (6.55 mEq/Kg) which is higher than that of T. mantaly (4 mEq/Kg) Terminalia catappa (2.60 mEq/Kg) as reported by (Abubakar *et al.*, 2020), Malaysian, Indian and Thailanian Jatropha (1.93 mEq/Kg, 3.70 mEq/Kg and 1.08 mEq/Kg) respectively (Emil *et al.*, 2010), Moringa seed 5.82 mEq/Kg, Sesame seed 8.33 mEq/Kg, Bitter kola 10.22 mEq/Kg, Melon seed 7.19 mEq/Kg

and water melon seed 13.41 mEq/Kg and the value falls within the AOAC 1990 standard (Saeed and Shola, 2015). Which in turn is lower than that of Nigerian and Indian jatropha (56.00 mEq/Kg and 39.20 mEq/Kg) respectively (Belewu et al., 2010). Peroxide value (PV) is the most common indicator of lipid oxidation. The unrefined vegetable oils are characterized by greater PV, compared to refined oils. High values of PV are indicative of high levels of oxidative rancidity of the oils and also suggest absence or low levels of antioxidant (Aremu *et al.*, 2015).

The iodine value in this study was (86.88 gI_2/100g), which is higher to that of bitter kola 53.99, Terminalia Catappa (54.567 gI_2 /100g) 44.4 gI_2/100g cashew nut oil (Aremu *et al*., 2006), J. curcas L. (40.91 gI_2/100g)(Abubakar *et al.*, 2020). On the other hand, the Iodine value of this study was lower than the range of J. curcas (134.11 gI_2/100g) (Emil *et al.*, 2010), soybean (128-143 gI_2/100g) (Karmakar *et al*., 2017). The iodine value could be used to quantify the amount of double bond present in the oil which reflects the susceptibility of the oil to oxidation. Oils with iodine value less than 100 gI_2/100g of oil are non-drying oils, correspondingly, (Aremu *et al.*, 2006) reported that the lower the iodine value the lesser the number of unsaturated bonds, thus, the lower the susceptibility of such oil to oxidative rancidity.

The flash point of the oil was found to be (155 °C) which is higher than that of the Jatropha curcas L. (152 ^oC) (Emil *et al*., 2010), and in turn lower than that of Karanja, Polanga, Rubber, and Mahua (205 °C, 221°C, 198 °C and 232 °C) respectively (Puhan *et al*., 2005; Karmee and Chadha, 2006; Akma *et al.*, 2012), H. crepitans and P. angollensis (272 °C and 260 °C) respectively, (Micheal and Cynthia, 2019), Neem oil (265 ^oC) (Idris *et al.*, 2018).

The pour point in this study was (1.83 ^oC), which is lower than that of Neem oil (25 ^oC) (Idris and Usman, 2018), Citrus sinensis Seed oil (2.5 ^oC) (Adekunle *et al.*, 2019), Coconut oil, Rubber seed oil, Palm oil and Neem oil (12.7 ^oC, 18 ^oC, 23.6 ^oC and 7 ^oC) respectively, and higher than that of Soybean, Sunflower, Rapeseed oil, Cotton seed oil, Sesame seed oil, Castor seed oil and Olive oil (-12 ^oC, -15 ^oC, -15 ^oC, -4.5 ^oC, -11 ^oC, -21 ^oC and -14 ^oC) (Karmakar *et al*., 2017). The result of Cloud point was found to be (4.56 ^oC) which is similar to that of pea nut and ground nut oil (4.5 ^oC) and in turn lower than that of palm oil (25.20 ^oC), Rice brand oil (16 ^oC), Jatropha oil (11 ^oC), Neem oil (13 ^oC) which is higher than that of Soybean oil, Sunflower oil, Rape seed oil, Cotton seed oil, Castor and Olive seed oil (-4 ^oC, -9.5 ^oC, -2 ^oC, -0.5 ^oC, -18 ^oC, and -11 ^oC) (Karmakar *et al.*, 2017).

FT-IR is an important analysis technique which detects the presence and the various characteristic of functional groups present in the oils. The results of FT-IR was shown in Table 7. The peak at 3004.280 cm^{-1} indicates the presence of Stretching of methyl ester CH=C-H. The C-H (stretching) at 2853.210 cm^{-1} to 2922.520 cm^{-1} shows the presence of alkanes. C=O stretching vibration at 1724.140 cm^{-1} indicates the presence of esters. C-H bending vibrations at 1462.140 cm^{-1} shows the presence of alkanes while at 1435.320 cm^{-1} was due to ethyl ester group. C-O bending and stretching at 1195.500 cm^{-1} to 1169.520 cm^{-1} indicate the presence of Alcohols and esters and confirms the biodiesel conversion. RCH=CHR bending at 993.430 cm^{-1} indicates the presence of Alkenes (cis disubstituted alkenes), the presence of $R_2C=CH_2$ Peak at 892.218 cm^{-1} indicates the presence of alkenes. This results agrees with the results C-H stretch (~2900 cm^{-1}), C=O stretch (~1700 cm^{-1}), broad O-H stretch (~3400 cm^{-1}) and C-O stretch (~1100 cm^{-1}) 886 cm^{-1}, 1436 cm^{-1} and 1644 cm^{-1} reported by Snezana *et al.* (2020). 2923.40 cm^{-1} Alkyls (CH_3, CH_2, CH), 1722.68 cm^{-1} ketone, 1598.10 cm^{-1} Alkenes and 1374.83 cm^{-1}Methyl (Ogwuche and Edema, 2020), and also similar to what has been reported by Elkady *et al.* (2015) 721 cm^{-1} -CH_2 rocking, 1373.70 cm^{-1} Bending vibrations of CH_2 groups, 1745 cm^{-1} C=O ester stretch, 1163 cm^{-1} C-O stretching, 1456 cm^{-1} Bending vibrations of the CH_2, 2925 -CH_2 stretching, Latchubugata *et al.* (2018) 1115.85 cm^{-1} for -O-CH_2-C, 1117.75 cm^{-1} for -O-CH_2-C, 1743.87 cm^{-1} for C=O ester 1742.07 cm^{-1} C=O ester, 1160.11 cm^{-1} C-O stretching, 1169.37 cm^{-1} C-O ester, 3005.44 cm^{-1} fore –CH, 2922.24 cm^{-1} for-CH_2, 2922.41 cm^{-1} for -CH_2, 2853.43 cm^{-1} for -CH_2.

IV. CONCLUSION

In summary, RSM was successfully applied to assess the effects of multiple variables, including alcohol/oil molar ratio, catalyst mass and reaction time for the production of biodiesel from Jatropha curcas L. seed oil. The experimental results suggested the optimal condition as follows: 0.30 g of catalysts and ethanol to oil molar ratio of 12:1 at 65 ^oC for 2 hrs. This optimized condition was validated with the actual biodiesel yield at 98.32 %. Increasing the molar ratio from 9 to 15 molar ratio while keeping other variables in their respective optimal conditions produced biodiesel with a yield of 95 %. Since increasing the biodiesel yield by 3.5 % with the cost of significantly increasing the molar ratio of methanol versus oil does not appear to be cost-effective, it was suggested that using methanol/oil molar ratio at 12.0 for the optimal production of biodiesel from Jatropha curcas L. seed oil.

ACKNOWLEDGEMENTS

My sincere appreciation goes to my supervisors, Dr. A.A. Mahmoud and Dr. D.A. Ajiya for their time, tireless corrections and constructive criticisms aimed at making this work a success. The authors also acknowledge the entire staff of the Department of chemistry Abubakar Tafawa Balewa University Bauchi, Nigeria for assisting me either directly or indirectly during the course of this research work.

REFERENCES

[1] Abubakar, A., Mahmoud, A.A., Yakubu, H., and Yushau, S. (2020). Extraction and Physicochemical Characterization of Seed oil from Terminalia mantaly Seed. International Journal of Research and Innovation in Applied Science 5, (1), 60-64.

[2] Abubakar, H., Jauro, A., Abubakar, I.M., Abubakar, A., Yushau, S., and Sallau, S. (2020). Production and biodegradability of biodiesel from Lagenaria siceraria seed oil. International Journal of research and innovation in applied science 5, (3), 132-134.

[3] Andualem, B and Gassesse, A. (2014)."Proximate composition mineral content and antinutritional factors of Bebra (Milletiaferruginea) seed flour as well as physicochemical characterization of its seed oil. ," A springa open journal vol. 3, pp. 1-10.

[4] Akpan, U. G. and Mohammed, J. A. (2007). "Extraction, characterization and modification of castor seed oil.," Leonardo Journal of Sciences vol. 8, pp. 43–52.

[5] Aremu, M., Olaofe, O., and Akintayo, E. (2006). "Chemical Composition and Physicochemical Characteristics of two Varieties of Bambara Groundnut (Vigna subterrenea) flours. ," Journal of Applied Sciences vol. 6, pp. 1900 - 1903.

[6] Abdulhamid, A., Sani, I., and Fekal, I. M. (2014). "Physicochemical Analysis of Soxhlet Extracted Oils from Selected Northern Nigerian Seeds.," International Journal of Biological, Biomolecular Agricultural, Food and Biotechnological Engineering vol. 8, pp. 1122-1124.

[7] Arif, N., Masood, T., and S. Syed, S. (2012). "Evaluation of oil seeds for their potential nutrients. ," ARPN Journal of Agricultural and Biological Science vol. 7,, pp. 730-734.

[8] Aremu, M., Ibrahim, H., and T. Bamidele, T. (2015). "Physicochemical Characteristics of the Oils Extracted from some Nigerian Plants Foods. ," A Review Chemical and Engineering Research vol. 32, pp. 36 - 52.

[9] Aremu, M., lonisakin, O., Bako, D., and P. Madu, P. (2006). "Compositional studies and physicochemical characteristics of cashew nut (Anarcadium occidentale) flour. ," Pakistan Journal of Nutrition vol. 5, pp. 328–333.

[10] Akma, I., Yaakob, Z., Anuar, N., Primandari, S., and Osman, M. (2012). "Physiochemical properties of Jatropha curcas seed oil from different origins and candidate plus plants (CPPs) " Journal of the American oil Chemists' Society vol. 89, pp. 293-300.

[11] Adekunle, O., Eluwale, E., Saheed, A., Samson, O., and Jide, O. O. (2019). " Biodiesel Potentials and Lubricating Properties of Citrus sinensis Seed Oil. ," International Journal of Bioorganic Chemistry vol. 4, pp. 84-92.

[12] Belewu, M. A., Adekola, F. A., Adebayo, G. B., G. B. Ameen, G. B., Muhammed, N. O. Olaniyan, et al. (2010). "Physico-chemical characteristics of oil and biodiesel from Nigerian and Indian Jatropha curcas seeds. International " Journal of Biological and Chemical Sciences vol. 4, pp. 524-529.

[13] Ebenezer, K. (2015). "Extraction and Physicochemical Characterization of oil extract from the seed of umbrella tree (Terminalia mantalis)," International Journal of Scientific and Engineering Research, vol. 6, pp. 144-147.

[14] Ejikeme, L., Obasi, N., and A. Egbuonu, A. (2010)."Physico-chemical and toxicological studies on Afzelia africana seed and oil. African Journal of Biotechnology " 9, vol. 13, pp. 1959-1963.

[15] Elkady, M.F., Ahmed, Z., & Ola, B. (2015). Production of Biodiesel from Waste Vegetable Oil via KM Micromixer. Journal of chemistry, 9, 1-9.

[16] Emil, A., Zahira, Y., Kumar, M., Jahim, J. and Salimon, J. (2010)"Comparative Evaluation of Physicochemical Properties of Jatropha Seed Oil from Malaysia, Indonesia and Thailand. ," Journal of American oil Chemists' Society vol. 87, pp. 689–695, 2010.

[17] Fan, X., Wang, X., and Chen, F. (2011). Biodiesel Production from Crude Cottonseed Oil: An Optimization Process Using Response Surface Methodology. The Open Fuels & Energy Science Journal 4, 1-8.

[18] Hamze, H., Akia, M., & Yazdani, F. (2015). Optimization of biodiesel production from waste cooking oil using response surface methodology. Process Safety and Environmental Protection, 4, 1-10.

[19] Hoekman, S.K., Broch, A., Robbins, C., Ceniceros, E., & Natarajan, M. (2012). Review of biodiesel composition, properties, and specifications. Renewable and Sustainable Energy Review 16, 143–169.

[20] Hossain, M. Alam, M. and M. Islam, M.(2007). "Physico-chemical and nutritional studies of Terminalia belerica Roxb. Seed oil and seed kernel.," Journal of bio-sciences vol. 15, pp. 117-126.

[21] Idris, M., Usman, M., and Igbafe, I. (2018). "Experimental Studies on Neem Seed (Azadirachta Indica) as a Possible Engineering Lubricating Fluid. ," International Journal of Agriculture and Earth Science, vol. 4, pp. 25-34.

[22] Jefferson, S., De, O., Polyanna, M., Lincoln, B., De, S., and M. Vinıcius, M. (2009)."Characteristics and composition of J. gossypiifolia and J. curcas L. oils and application for biodiesel conversion," Biomass and bioenergy vol. 33, pp. 449-453.

[23] Kalpana, J.G., Bajaj, S.B., & Sumedh, S.A.I., (2019). Analysis of Performance and Emission of Biodiesel with Perovskite Nanomaterial in Diesel Engine using Taguchi Approach. International Research Journal of Science & Engineering 7, (1), 29-33.

[24] Karmakar, G., Ghosh, P., and Sharma, B. (2017). "Chemically modifying vegetable oils to prepare green lubricants," Lubricants vol. 5, p. 44.

[25] Karmee, S., and Chadha, A. (2006). "Preparation of biodiesel from crude oil of Pongamia pinnata," Bioresource Technology, vol. 96, p. 416.

[26] Knothe, G., & Steidley, R. K. (2007). Kinematic viscosity of biodiesel fuel component and related compounds. Influence of compound structure and comparison to petro-diesel fuel components. Fuel, 84, 1059-1065.

[27] Kumar, V., & Kant, P. (2013). Study of Physical and Chemical Properties of Biodiesel from Sorghum Oil. Research Journal of Chemical Sciences 3, (9), 64-68.

[28] Latchubugata, C.S., Kondapaneni, RV, Patluri KK, Virendra, U & Vedantam S (2018) Kinetics and Optimization Studies using Response Surface Methodology in Biodiesel Production using Heterogeneous Catalyst. Chemical Engineering Research and Design 18, 1-30.

[29] Lee, H.V., Yunus, R., Juan, J.C., & Taufiq-Yap, Y.H. (2011). Process optimization design for jatropha-based biodiesel production using response surface methodology. Fuel Processing Technology, 92, 2420–2428.

[30] Mabrouk, S. (2005). "Making Useable, Quality Opaque or Transparent Soap," Journal of Chemical Education vol. 82, pp. 1534 – 1537, 2005.

[31] Mahmoud, A. A., Abubakar, A., Yushau, S., and Yakubu, H. (2019). Extraction and characterization of the seed oil from Terminalia mantaly seed. International Journal of research and innovation in applied science 5, (1), 60-64.

[32] Mahmoud, A. A., Kwada, S. I., Jauro, A., and Abubakar, A. (2020). Production and biodegradability of biodiesel from Lagenaria siceraria seed oil. International Journal of research and innovation in applied science 5, and (3), 94-98.

[33] Michael, A., and Cynthia, F. (2019). "Physicochemical properties of Hura crepitans and Pycnanthus angollensis Seed Oils and Their Possible Uses " Scholars Bulletin vol. 5, pp. 184-187.

[34] Monnet, Y., Gbogouri, A., Kouadio, B., and Patrice, K. (2012). "The chemical characterization of seeds and seed oils from mature Terminalia catappa fruits harvested in Côte d'Ivoire," International Journal of Biosciences vol. 2, pp. 110-124.

[35] Moyo, L.B, Iyuke, S.E., Muvhiiwa, R.F., Simate, G.S., & Hlabangana, N. (2021). Application of response surface methodology for optimization of biodiesel production parameters from waste cooking oil using a membrane reactor. South African Journal of Chemical Engineering 35, 1-7.

[36] Murugesan, A., Umaran, C., Chinnusamy, T.R., Krishnan. M., Subramanian, R., & Neduzchezhain, N. (2009). Renewable and Sustainanable. Energy Review 13, 825–834.

[37] Ogwuche, C. E., & Edema, M. O. (2020). GC-MS and FTIR Characterization of essential oil from the fresh leaves of Pandanus candalabrum obtained from Bayelsa state, Nigeria. Nigerian Journal of Chemical Research, 25(1), 1-10.

[38] Orhevba, B., Adebayo, S., and A. Salihu, A. (2016). "Synthesis of biodiesel from tropical almond (Terminalia catappa) seed oil. Current " Research in Agricultural Sciences vol. 3, pp. 57-63.
[39] Puhan, S., Vedaraman, N., Ram, B., Sankarnarayanan, G., and K. Jeychandran, K. (2005). "Mahua oil (Madhua indica seed oil) methyl ester as biodiesel-preparation and emission characteristics. ," Biomass and Bioenergy vol. 28, pp. 87-93.
[40] Premjet, D., Abraham, K., Hah, Y., Seung, W. and Premjet, S. (2021). "Physicochemical Characterization of Jatropha podagrica Seed Oil for Potential Biodiesel Production and other Industrial Applications in Thailand," Sains Malaysiana vol. 50, pp. 85-92, 2021.
[41] Ramadhas, A.S., Jayaraj, S., & Muraleedharan, C. (2005). Biodiesel production from FFA rubber seed oil. Fuel 84, 335-340.
[42] Saeed, K., and Abbas, A. (2016)."Determination of the Physico-Chemical Properties of Z-spinachristi (Nabag) Seed Crude oil," Journal of Agricultural Research, vol. 2, pp. 22-35.
[43] Saeed. M., and E. Shola, E. (2015). "Extraction and physico chemical properties of some edible seed oils sampled in keno metropolis, Kano State," Bayero Journal of pure and applied sciences vol. 8, pp. 239 – 244.
[44] Shuit, S.H., Lee, K.T., Kamaruddin, A.H. & Yusup, S. (2010). Reactive extraction of Jatropha curcas L. seed for production of biodiesel: Process optimization study. Environmental Science & Technology, 44, 4361-4367.
[45] Singh, S.P., & Singh, D. (2010). Biodiesel production through the use of different sources and characterization of oils and their esters as the substitute of diesel: A review. Renewable and Sustainable Energy Reviews14, (1), 200-216.
[46] Snezana A.K., Petar, R., Vladimir, G., Tatiana M. L., & David W. M. (2020). Essential Oil Quality and Purity Evaluation via FT-IR Spectroscopy and Pattern Recognition Techniques. Applied sciences, 10 (7294), 1-12.
[47] Tan, Y,H., Abdullah, M.O., Nolasco-Hipolito, C., and Zauzi, N.S.A., (2017). Application of RSM and Taguchi mothods for optimizing the transesterification of waste cooking oil catalyzed by solid ostrich and chicken-eggshell derived CaO. Renewable energy, 114, 437-447.
[48] Umar, D. M., Jauro, A., Yushau, S., Abubakar, A., H. Abubakar, H., Abubakar, et al.(2020). "Characterization and Determination of Nutritional and Anti-Nutritional Values of the Seed Oils of Sorrel (Hibbiscus Sabdariffa) and Okro (Abelmoschus Esculentus) " International Journal of Research and Innovation in Applied Science vol. 5, pp. 1-7.
[49] Wong, Y.C., Tan, Y.P., Taufiq-yap, Y.H., & Ramli, I. (2015). An Optimization Study for Transesterification of Palm Oil using Response Surface Methodology (RSM).Sains Malaysiana, 44(2), 281–290.
[50] Yunus, M.M., Zuru, A.A., Faruq, U.Z., & Aliero, A.A, (2013) Assessment of Physicochemical Properties of Biodiesel from African Grapes (Lannea microcarpa Engl. & K.Krause) Nigerian Journal of Basic and Applied Science 21, (2), 127-130.