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Original Research Article

Process Optimization of the Re-Refining of Spent Lubricating Oil Using Response Surface Methodology

***[1]Tang'an, D.B., [1,2]Osha, O.A. and [1]El-Nafaty, U.A.**

[1]Department of Chemical Engineering, Abubakar Tafawa Balewa University Bauchi, P.M.B. 0248, Bauchi, Nigeria.
[2]Department of Chemical Engineering, University of Calabar, P.M.B. 1115, Calabar, Nigeria.
*direttangan@gmail.com

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ABSTRACT

Lubricating oils used in automotive engines become contaminated with impurities and residues. Improper management of these oils is a leading cause of pollution worldwide. Re-refining with solvent extraction is a competitive process for recycling them. To optimize this process, used engine oils were collected and homogenized representing typical feedstock for re-refining plants. The oil was characterized and heated to 260°C to remove emulsified water, gas oils, and fuel diluents. It was then mixed with ethanol at 10:1 and subjected to different temperatures, mixing times and speed conditions using Box-Behnken design-of-experiment. The mixture was centrifuged, and the extract distilled to separate and weigh the base oil. An optimal yield of 39.21 wt.% was achieved at 70°C, 20 minutes, and 200 rpm. The quadratic model fitting using ANOVA showed a coefficient of determination (R^2) of 92.86% and standard deviation (S) of 1.54, indicating that the model sufficiently explained the variation in base oil yield. The p-value regression (0.021) was statistically significant, demonstrating the model's validity. There was no evidence of lack-of-fit as p-value (0.570) was statistically insignificant. The recovered base oil met acceptable SAE standards, with specific gravity (0.8924), kinematic viscosity (7.63 cSt at 100°C), flash point (194°C), and pour point (-11°C).

1. INTRODUCTION

The rising environmental pollution due to global growth in using lubricating oil for automotive engines and the more stringent environmental regulations has driven investigations into the recycling of used lubricating oils (Osman *et al.*, 2017; Botas *et al.*, 2017; Maceiras *et al.*, 2017). Lubricating oils which comprise base oil and additives are viscous fluids used to minimize friction and wear between interactive solid surfaces in order to facilitate the relative motion between them (Aremu *et al.*, 2015). These oils separate the metallic parts of an engine thereby minimizing the friction existing between them, reducing material wear, improving machine efficiency, enhancing fuel and energy savings and further keeping the engine clean (Ogbeide, 2010; Merai, 2015).

During application in lubrication, impurities such as dirt, water, chemicals, degraded additives and very fine heavy metals due to engine erosion get mixed with the oil thus reducing its lubricating performance and rendering it unsuitable for use (Sarkar *et al.*, 2023). At this point, the oil which contains about 25% impurities and 75% active base oil is referred to as spent (Sener 2012, Tang'an *et al.*, 2017). Moreover, the chemical breakdown of the additives present in the oil during use results in the buildup of halogenated hydrocarbons (Aremu *et al.*, 2015). Spent lubricating oil is therefore a petroleum-based oil which through usage, storage or handling becomes unfit for the purpose for which it was primarily designed (Bamiro and Osibanjo, 2013). When inappropriately disposed of into the environment because of improper handling or management, it poses a major threat to human health and the ecosystem i.e., soil, water and air (Sarkar, *et al.*, 2023, Sarkar *et al.*, 2024). Spent lubricating oil is considered a significant liquid hazardous waste due to the presence of Polychlorinated Biphenyls (PCBs), heavy metals and Polyaromatic Hydrocarbons (PAHs) (Jafari and Hassanpour, 2015; Pinheiro *et al.*, 2017). Inhaling or ingesting these contaminants would endanger the lives of animals (Sarkar *et al.*, 2024).

Each year, forty (40) million tons of lubricant oils are consumed around the world, generating twenty (20) million tons of used lubricating oils (Sener, 2012). This suggests that about half of the oil generated is unaccounted for probably due to improper management and disposal. Disposing 1 volume of spent oil pollutes at least 250 000 volumes of water (Bridjanian and Sattarin, 2006). To solve this menace, the regeneration of used oil is considered the most cost-effective and environmentally friendly approach (Sarkar *et al.*, 2024). Different recycling techniques employed such as the acid-clay method generate a significant amount of hazardous waste sludge. Bridjanian and Sattarin (2006) reported that for every 1 000 tons of used oil processed using this method, about 200 tons of environmentally harmful by-product is generated. Recent recovery method such as catalytic hydrogenation which is an effective substitute for the acid-clay method is capital intensive and only justifiable for large-capacity projects (Sener, 2012). This necessitates the need for an environmentally friendly recovery method for low to medium recycling capacity process that avoids the acid-clay finishing step.

As a replacement for the acid clay treatment process, re-refining using solvent extraction has attracted interest because of its selectivity in extracting and recovering the active base oil component of spent lubricating oil (Hussein *et al.*, 2014; Hussein *et al.*, 2015). This process conveniently removes additives, water, wear metals and other impurities from the spent lubricating oil and returns its active base oil component which could now be blended with new additives and restored to its initial design specification for use without causing further pollution (Antony, 2012). Additionally, the spent oil re-refining process is not as rigorous as crude oil refining because it only uses one-third of the energy requirement to produce virgin base oil from crude oil (Merai, 2015; Chemical Engineering Partners, 2016). However, care must be taken to select solvents that allow high base oil solubility and minimal solubility of additives (Osman *et al.*, 2017). Alcohols have been found to have high removal performance in liquid-liquid extraction compared to other solvents like ketones (Hussein *et al.*, 2014), thus, ethanol is used for this study.

Presently, there exist few reports in the literature on re-refining spent lubricating oil using ethanol. However, no such comprehensive work is available in the literature on the optimization of the re-refining technique using ethanol solvent. Therefore, this study utilized the Response Surface Methodology (RSM) to determine operating conditions that improve base oil extraction from spent lubricating oil using ethanol with the aim of optimizing the recycling efficiency of the re-refining process and giving a product with desired properties.

Other optimization techniques applied in product development exist, however, RSM has gained popularity in recent years as it could be applied in chemical, biochemical, metallurgical, petrochemical processes, food processing etc. (Brown, 2013). Overall, RSM could be used in finding operating conditions (factor settings) that produce optimum response, meet process specifications, establish new operating conditions that improve product quality and to model a relationship between operating conditions and responses (Minitab Inc., 2007).

Re-refining spent lubricating oil shall serve as a means to convert waste to wealth in addition to meeting stringent environmental laws, thereby opening opportunities for job creation if explored on a larger scale hence preserving our crude oil reserves and conserving the environment.

2. MATERIALS AND METHODS

2.1. Materials and Reagents

Spent lubricating oils were obtained from service stations in the Bauchi metropolis, Nigeria. Analytical grade ethanol (99%), potassium hydroxide (86.0% assay), and ammonium chloride (99.5% assay) were obtained respectively from Loba Chemie Pvt Ltd, Titan Biotech Ltd and Burgoynes & Co, India and used throughout the experiment without further purification.

2.2. Experimental Design

In this research, RSM using the Box-Behnken technique was adopted to study three parameters affecting the yield of lubricating base oil from its spent using ethanol solvent. In addition to its advantage of graphical visualization in 3D, this method allows the optimization of extraction yield and also enables the understanding of how extraction yield changes in a given direction with adjustment in design variables (Hussein *et al.*, 2015). The experimental design involves twelve (12) runs with three center point runs making a total of fifteen (15) experimental runs. The factors i.e. temperature, extraction time and mixing speed were coded x_1, x_2 and x_3 with levels -1, 0 and +1 representing, low level, center point and high level respectively. Table 1 shows the summary of these factors, their levels and codes.

Table 1: Coding of independent variables

	Temperature (x_1)		Extraction time (x_2)		Mixing speed (x_3)	
Levels	Value (°C)	Coded	Value (min.)	Coded	Value (rpm)	Coded
Low	30	-1	10	-1	200	-1
Medium	50	0	20	0	250	0
High	70	1	30	1	300	1

The temperature range (30-70°C) was selected to allow investigation from a low temperature of 30°C (average ambient temperature in Bauchi metropolis) to a higher temperature of 70°C less than the boiling point of the ethanol (78°C) to prevent boiling and subsequent loss of solvent during extraction. A mixing time range of 10-30 minutes was adopted from the method used by Hussein *et al.* (2015). However, the mixing speed range of 200-300 rpm was chosen based on the capacity of the magnetic stirrer to allow the study of the effect of mixing on base oil extraction yield.

2.3. Analysis of Results

Statistical analysis of the Box-Behnken model using analysis of variance (ANOVA) describes the interaction between the process variables (i.e. mixing temperature, extraction time and mixing speed) and response (i.e. percentage base oil yield). From the statistical analysis of the model, the response as a function of independent variables (x_i) is generated and expressed as shown in Equation (1).

$$Y = f(x_1, x_{2,\dots,}x_3) \tag{1}$$

The sole aim is to optimize the response (Y) as a function of the independent factors x_i. A second-order polynomial response equation for yield comprising the linear, quadratic and interaction terms is generated as shown in Equation (2).

$$Y = b_o + \sum b_i x_i + \sum {b_i}^2 {x_i}^2 + \sum b_{ij} x_i x_j \tag{2}$$

Where Y is the response (extraction yield), b_o is the intercept, b_i is the coefficient for linear direct effect and b_i^2 is the coefficient for quadratic effect responsible for model curvature. b_{ij} is the coefficient for interactive effect (Agarry and Ogunleye, 2012; Hussein *et al.*,2015).

2.4. Experimental Procedure

In this work, spent lubricating oil samples were collected from points within the Bauchi metropolis and homogenized by mixing to represent a typical feedstock for a re-refining plant. After physical screening for solid objects, the spent oil sample was characterized. Oil pretreatment was carried out by heating to 260°C to remove emulsified water, fuel diluents and raw gas oil. Analytical grade ethanol was mixed with potassium hydroxide (KOH) in the ratio 2 g KOH/Kg of solvent to enhance the base oil extraction rate (Hussein *et al.*, 2015). Base oil extraction was carried out by mixing 5 grams of pretreated oil with the prepared ethanol solvent at a solvent-oil mass ratio of 10:1 (established from a preliminary investigation at a temperature of 50ºC, mixing speed of 250 rpm for 20 minutes) and maintained throughout the experiment. To establish the operating conditions with the optimum base oil recovery, the solvent-oil mixture was subjected to various experimental conditions of temperature (30, 50 and 70ºC), extraction time (10, 20 and 30 minutes) and mixing speed (200, 250 and 300 rpm). After the extraction step, the mixture was centrifuged for 30 minutes at 1500 rpm and 30ºC using Eppendorf Centrifuge where a clear separation of the sludge from the extract-solvent mixture was visually observed (Figure 1).

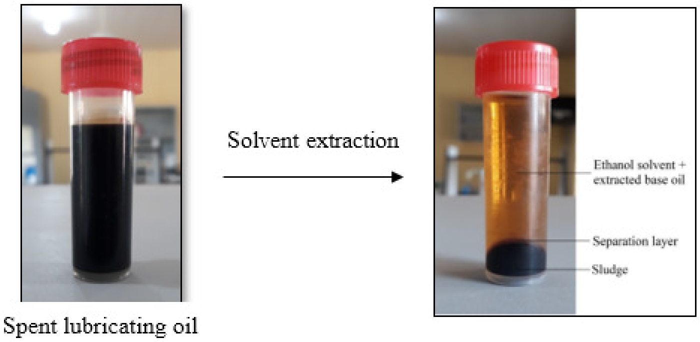

Figure 1: Spent lubricating oil re-refining using ethanol solvent

The extract-solvent (base oil and ethanol) was separated by distillation using a rotary evaporator at 95ºC to sufficiently allow the ethanol solvent to boil off leaving the base oil. The distillate (ethanol) was recovered and reused while the extracted base oil was collected and weighed to determine the % base oil yield using Equation (3) and was further analyzed for its suitability as a lubricant.

$$Base\ oil\ extraction\ yield\ (wt.\%) = \left(\frac{W_{extracted\ base\ oil}}{W_{spent\ oil\ used}}\right) * 100 \tag{3}$$

Measurements for density, specific gravity, API gravity, kinematic viscosity and viscosity index were carried out in quadruplets and their average values and standard deviations were reported.

2.5. Characterization of Lubricating Oil

2.5.1. Determination of density, specific gravity and API gravity

A clean dry 2 mL syringe was weighed (w_o) and filled with distilled water to 1 mL (V_o) and reweighed to give (w_1). The water was then substituted with the lubricating oil after drying and weighed to give (w_2). The density (ρ), specific gravity (SG) and API gravity were determined respectively from Equations (4), (5) and (6). This was repeated four times, and the average values were reported. Note that both the distilled water and lubricant were cooled to 15ºC (60/60ºF) in an ice water bath before measurement to conform to ASTM D 1298 standard temperature.

$$Density\ \left(\frac{g}{mL}\right) = \frac{w_2 - w_o}{v_o} \tag{4}$$

$$Specific\ gravity\ = \frac{w_2 - w_o}{w_1 - w_o} \tag{5}$$

$$API\ gravity\ = \left(\frac{141.5}{Specific\ gravity\ @\ 60/60°\mathrm{F}}\right) - 131.5 \tag{6}$$

2.5.2. Determination of kinematic viscosity and viscosity index

Kinematic viscosity (at 40 and 100ºC) and viscosity index (VI) of the lube oil were determined using the ASTM D446 and D 445 standard methods of testing. The Cannon-Fenske routine viscometer for transparent liquids was used.

2.5.3. Determination of flash point

The flash point is the lowest temperature of the lubricating oil at which the application of a test flame will cause the oil vapor to ignite (Obasi *et al.*, 2014). At room temperature, the lube oil sample was fed into a beaker and placed on a magnetic stirrer and heater. The bulb of an alcohol-in-glass thermometer capable of measuring up to 300°C was immersed into the sample with the support of a stand and clamp. The procedure outlined by ASTM D92-78 was then followed in carrying out the test. The heater control was set to heat the sample rapidly first and then at a slow constant rate. At specified temperature intervals, a small test flame across the heated sample was passed to see whether it would flash. The temperature reading on the thermometer at which the lubricant flashes is recorded as the flash point.

2.5.4. Determination of Pour Point

The pour point which signifies the lowest temperature at which the lube oil specimen flows under the prescribed condition of the test is determined by an initial heating of the sample in a test tube after which it is cooled at a specified rate using an ice bath mixed with Ammonium chloride-to further reduce the temperature of the bath to enable pour point measurements at very low temperatures of up to -12°C using a mercury in glass thermometer. The lubricating oil sample is then examined at intervals of 3°C for flow characteristics as outlined by ASTM D 97–IP standard method of testing. The minimum temperature at which the specimen flows was taken as the pour point.

3. RESULTS AND DISCUSSION

3.1. Optimization of Process Parameters Using Response Surface Methodology

A total of fifteen experimental runs were performed in accordance with the Box-Behnken DOE to optimize and study the effect of process parameters on the extraction yield of lubricating oil bases. The result obtained is shown in Table 2. The lowest and highest base oil recovery of 23.95 wt.% and 39.21 wt.% were observed at a temperature, time and mixing speed of 50°C, 20 minutes, 300 rpm and 70°C, 20 minutes, 200 rpm respectively.

Table 2: Box-Behnken's design matrix and results of experimental and predicted values for percentage base oil extraction yield at 10:1 solvent to spent oil mass ratio

Run order	Block	Process condition						Extracted base oil yield (wt.%)	
		Temperature (x_1)		Time (x_2)		Mixing speed (x_3)			
		Code	Value (°C)	Code	Value (min.)	Code	Value (rpm)	Experimental	Predicted
1	1	-1	30.0	0	20.0	1	300.0	32.01	32.65
2	1	0	50.0	1	30.0	-1	200.0	35.07	34.60
3	1	0	50.0	0	20.0	0	250.0	31.75	30.74
4	1	0	50.0	0	20.0	0	250.0	28.90	30.74
5	1	0	50.0	-1	10.0	1	300.0	23.95	24.42
6	1	1	70.0	0	20.0	1	300.0	31.52	31.53
7	1	1	70.0	0	20.0	-1	200.0	39.21	38.57
8	1	1	70.0	-1	10.0	0	250.0	29.76	29.28
9	1	1	70.0	1	30.0	0	250.0	34.54	35.66
10	1	-1	30.0	-1	10.0	0	250.0	30.63	29.51
11	1	0	50.0	0	20.0	0	250.0	31.58	30.74
12	1	0	50.0	1	30.0	1	300.0	33.40	32.28
13	1	-1	30.0	0	20.0	-1	200.0	35.56	35.55
14	1	-1	30.0	1	30.0	0	250.0	33.06	33.54
15	1	0	50.0	-1	10.0	-1	200.0	30.92	32.04

3.2. Effect of Process Variables on Base Oil Extraction Yield

3.2.1. Effect of temperature on extraction yield

As temperature increases from 30ºC to about 50ºC at a mixing speed of 200 rpm, a decrease in base oil extraction from spent lubricating oil was observed (Figure 2). However, with further temperature increment, extraction yield begins to rise leading to a maximum yield of 39.21 wt.% recorded at a high temperature of 70ºC. Visual observation revealed a darker extract obtained at this temperature as compared with other lower temperatures. High extraction yields of 36-38 wt.% as represented by the dark green color on the contour plot in Figure 3 could only be obtained within a temperature range of 58-70ºC and time of 12-30 minutes whereas extraction yields >38 wt.% as shown by the darkest green color were only detected at a temperature range of 67-70ºC and extraction time range of 17.5-30 minutes.

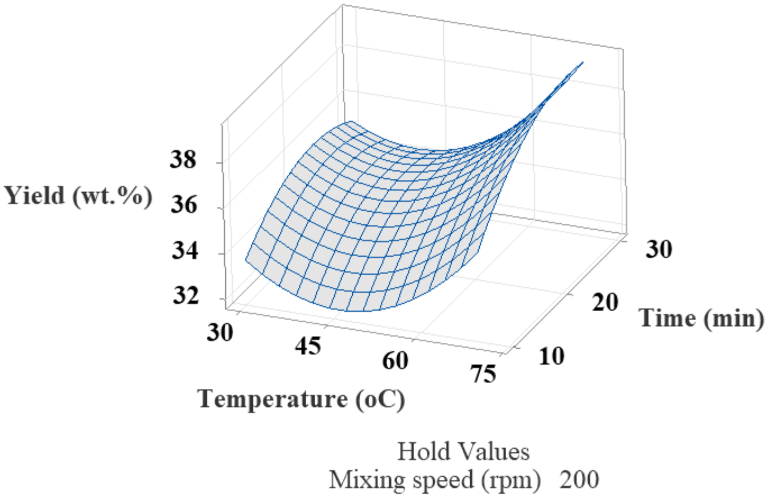

Figure 2: Surface plot of base oil extraction yield (wt.%) against extraction time (min) and temperature (ºC) at 200 rpm and 10:1 solvent to spent oil mass ratio

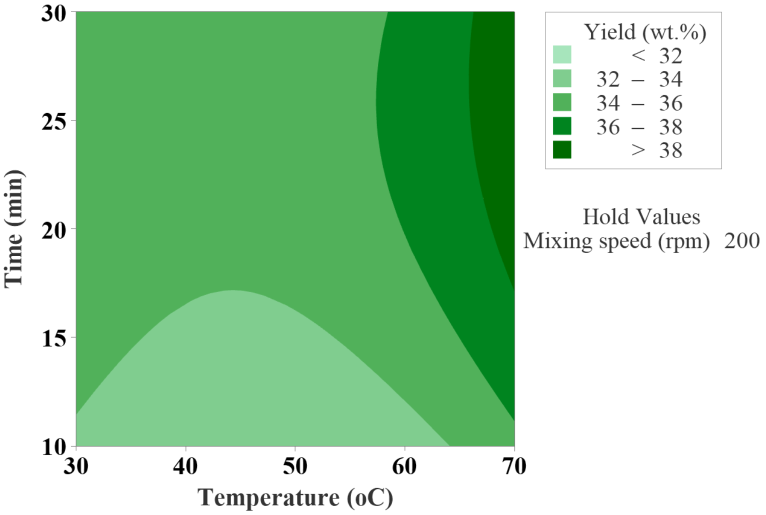

Figure 3: Contour plot of extraction yield (wt.%) against extraction time (min) and temperature (ºC) at 200 rpm and 10:1 solvent to spent oil mass ratio

3.2.2. Effect of mixing time on extraction yield

Figure 4 shows a surface plot of the effect of extraction time and mixing speed on base oil extraction yield. It was observed that the extraction yield was directly proportional to the extraction time as represented by the upward curve (slope) on the plot. This implies that high extraction time allows sufficient interaction between the solvent and base oil leading to more base oil recovery. As shown on the contour plot (Figure 5), two dark green regions were observed at a temperature of 30°C to represent higher extraction yields from 34-35 wt.% at a mixing speed of 200-228 rpm and 280-300 rpm respectively and an extraction time of 11.5-30 minutes and 25-30 minutes respectively. The darkest green area represents yields greater than 35 wt.% within a mixing speed and extraction time range of 200-210 rpm and 15.5-30 minutes respectively.

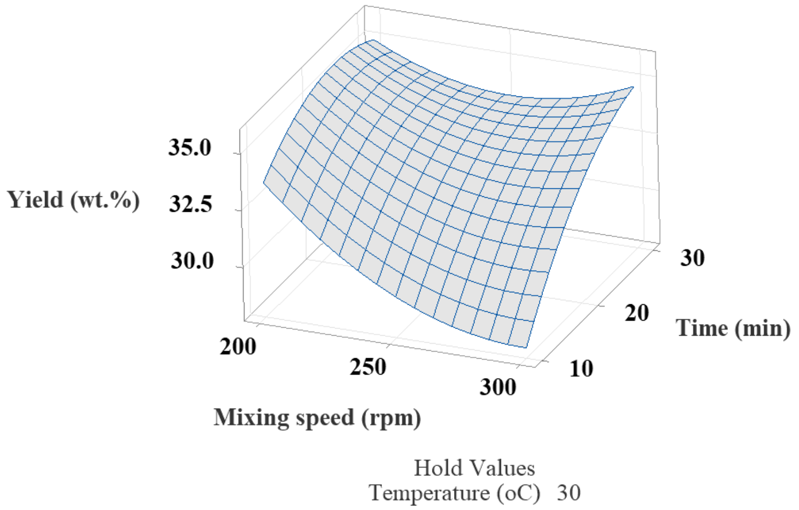

Figure 4: Surface plot of extraction yield (wt.%) against extraction time (min.) and mixing speed (rpm) at 30°C and 10:1 solvent to spent oil mass ratio

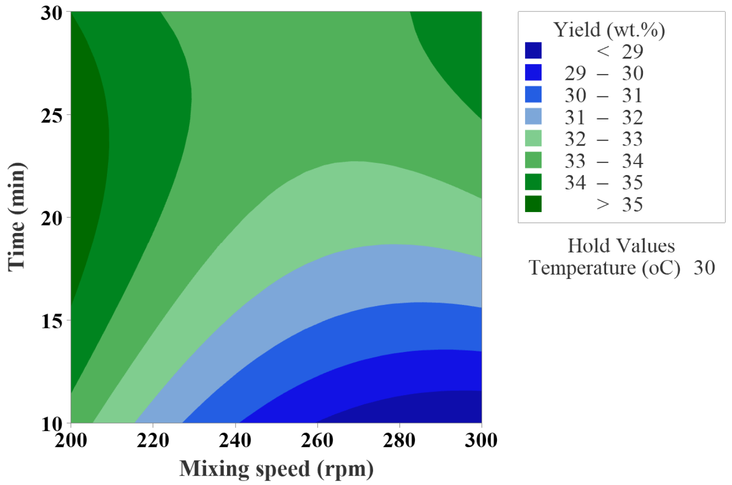

Figure 5: Contour plot of extraction yield (wt.%) against extraction time (min) and mixing speed (rpm) at 30°C and 10:1 solvent to spent oil mass ratio

3.2.3. Effect of mixing speed on extraction yield

Overall, an inverse proportionality relationship was observed between the base oil extraction yield and mixing speed at an extraction temperature of 30°C (Figure 4). The highest yields >35 wt.% were only gotten within a mixing speed range of 200-210 rpm (Figure 6). However, relatively high yields of 34-35 wt.% were recorded not only within lower mixing speeds of 200-228 rpm but also at higher mixing speeds of about 280-

300 rpm. This could be attributed to the longer extraction time (i.e. 25-30 minutes) within which these yields were obtained. This trend agrees with the finding of Hussein *et al.* (2015) who studied base oil extraction using a 1:1 wt.% solvent blend of 1 butanol and methylethylketone within a mixing speed range of 50-250 rpm. The decrease in base oil extraction with increasing mixing speed indicates that slow mixing favors base oil extraction as it allows room for sufficient interaction between the ethanol and base oil.

3.2.4. Interactions of process variables and their combined effect on base oil yield

To further understand the individual and the combined effect of the process factors on the extraction yield, the main effect plot was used. This examines the level means for each factor, compares the level means for the different factors and also compares the relative strength of the effect across factors (Minitab Inc., 2007). The factor means on the main effect plot (Figure 6) showed a gradual decrease in extraction yield i.e. from ≈ 32.8 wt.% to 31 wt.% as temperature increased from 30 to 50ºC respectively. However, an increase in yield of ≈ 31 wt.% to 34 wt.% represented by the steeper slope is observed with further increment in temperature between 50 and 70ºC. As extraction time increases from 10 to 20 minutes, extraction yield increases rapidly from ≈ 28.5 wt.% to 32.8 wt.%. An increase in yield from 32.8 wt.% to 34 wt.% was registered with further increment in extraction time from 20 to 30 minutes respectively. However, as shown by the less steep slope, the extraction yield was not drastic within this time range as compared with the yield at early extraction times. This behavior possibly indicates a gradual approach to the point of equilibrium within the system where no significant increment in base oil extraction yield would be noticed. Similar behavior was observed with mixing time. However, instead of increasing, the base oil yield decreased rapidly from ≈ 35.2 wt.% to 31.3 wt.% between 200-250 rpm respectively and then gradually from ≈ 31.3 wt.% to 30.2 wt.% between 250-300 rpm respectively.

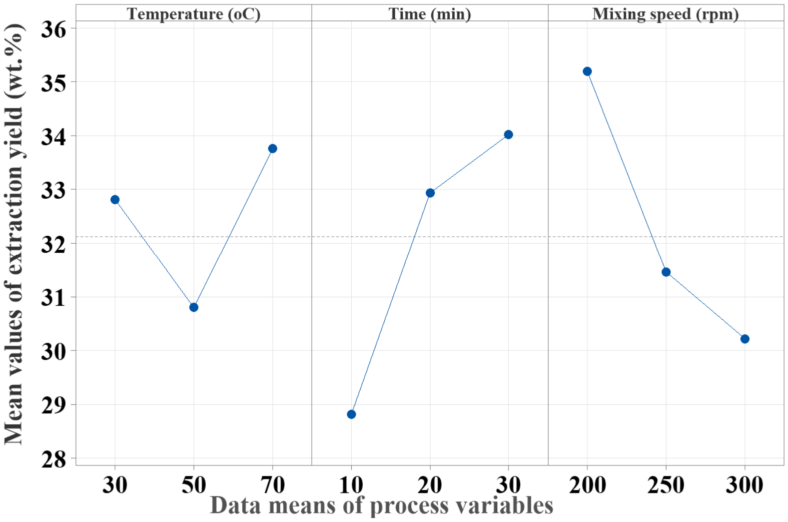

Figure 6: Main effects plot for extraction yield (wt.%) at 10:1 solvent/oil mass ratio

3.2.5. Statistical Analysis of Results

3.2.5.1. Model fitting and testing of the model using regression analysis

In regression analysis, the standard deviation (S), coefficient of determination (R^2) and adjusted R^2 measure how well the model fits the data (Minitab Inc., 2007). The coefficient of determination (R^2) measures the significance of the regression while the adjusted coefficient of determination (Adj - R^2) estimates the model's accuracy fitness as well as corrects the coefficient of determination value for the sample size and number of terms in the model (Dasari *et al.*, 2017; Minitab Inc., 2007). From Table 3, the coefficient of determination (R^2 = 92.86%) indicates a high degree of correlation between the model and the experimental data for the extraction of base oil from spent lubricating oil. This is because the more R^2 is to 1 the better the model which fits the experimental data. It suggests that more than 92.86% of experimental data were compatible whereas only 7.14% of the total variations were not explained by the model. Adjusting the coefficient of determination for the number of terms (i.e. linear, quadratic and interactive terms) in the model gives a modified R^2 (Adj - R^2 = 80.01%) which is a decrease in R^2 by 12.85%. This high value of Adj - R^2 = 80.01% and low standard deviation S = 1.54 confirms the significance of the model.

Table 3: Regression analysis for coded response surface quadratic model

Term	Coefficient	SE coefficient	*T*-value	*P*-value	Remark
Constant	30.7433	0.8897	34.555	0.000	Significant
x_1 - Temperature	0.4713	0.5448	0.865	0.427	Insignificant
x_2 – Extraction time	2.6012	0.5448	4.775	0.005	Significant
x_3 – Mixing speed	-2.4850	0.5448	-4.561	0.006	Significant
$x_1 * x_1$	2.4971	0.8020	3.114	0.026	Significant
$x_2 * x_2$	-1.2429	0.8020	-1.550	0.182	Insignificant
$x_3 * x_3$	1.3346	0.8020	1.664	0.157	Insignificant
$x_1 * x_2$	0.5875	0.7705	0.763	0.480	Insignificant
$x_1 * x_3$	-1.0350	0.7705	-1.343	0.237	Insignificant
$x_2 * x_3$	1.3250	0.7705	1.720	0.146	Insignificant
Standard deviation = 1.54098, R^2_{model} = 92.86%, $R^2_{Adjusted}$ = 80.01%					

Key: SE, Standard error

The coefficient value for P (p-value) tells whether the association between the response and predictors (factors) is statistically significant. This is determined by comparing the coefficient of p-value to the commonly used α-level value (α-level = 0.05) at a 5% significance level. If the p-value is less than the α-value selected (i.e., $p < \alpha$-level), then the association is said to be statistically significant (Minitab Inc, 2007). However, if $p > \alpha$-level then the association is statistically insignificant and could be neglected in the model. It could be seen that the coefficient/constant of the model (intercept), x_2 (extraction time), x_3 (mixing speed) and x_1^2 $(\text{temperature})^2$ have a statistically significant effect on the base oil extraction yield for the coded response surface quadratic model as their p-values of 0.000, 0.005, 0.006 and 0.026 respectively were less than 0.05. However, the remaining linear term, x_1 (p = 0.427), quadratic terms, x_2^2 (p = 0.182), x_3^2 (p = 0.157), and interactive terms, $x_1 * x_2$ (p = 0.480), $x_1 * x_3$ (p = 0.237), and $x_2 * x_3$ (p = 0.146) were insignificant and therefore could be neglected in the model. Overall, the relative strength of the effect of individual factors on extraction yield is in the order, of extraction time > mixing speed > temperature as revealed by their respective absolute coefficients, 2.6012, 2.4850 and 0.4713 from the coded model.

3.2.5.2. ANOVA for the response surface quadratic model fitting

The amount of variation in response data explained by the predictors (factors) and those variations unexplained is best revealed by the ANOVA table using the two most important p-values i.e., p-value (regression) and the p-value (lack of fit). The p-value regression determines whether or not the regression coefficients are significantly different from zero while the lack of fit p-value determines whether the selected model adequately describes the experimental data or a more complicated model should be selected. (Minitab Inc., 2007; Agarry and Ogunleye, 2012). ANOVA results (Table 4) explain that the fitted model is considered sufficient to explain the variation in base oil extraction yield data as the p-value regression (p = 0.021) obtained from the *F* test was significant ($p < 0.05$). Additionally, no evidence of lack of fit was observed as the Lack of fit p-value (p = 0.570) was much greater than 0.05 (i.e., p » 0.05) and thus considered insignificant.

From the regression analysis of experimental results, a regression model (Equation (7)) for determining responses was developed using uncoded factors at a 95% confidence limit.

$$\begin{aligned} Yield\ (wt.\%) = {} & 84.0374 - 0.4007 * (Temp.) - 0.0521 * (Time) - 0.3179 \\ & * (Speed) + 0.0062 * (Temp.)^2 - 0.0124 * (Time)^2 + 0.0005 \\ & * (Speed)^2 + 0.0029 * (Temp. * Time) - 0.0010 * (Temp. * Speed) \\ & + 0.0026 * (Time * Speed) \end{aligned} \tag{7}$$

Note that all terms irrespective of their significance were included in the model. To validate the model, results obtained by Equation (7) were compared with experimental results. The results were found to be in close agreement thus the model is valid and could be applied within the experimental boundary.

Table 4: ANOVA for the response surface quadratic model

Source	DF	Seq SS	Adj SS	Adj MS	*F*-value	*P*-value
Regression	9	154.457	154.457	17.1619	7.23	0.021
Linear	3	105.310	11.222	3.7408	1.58	0.306
Temp. (°C)	1	1.777	4.540	4.5403	1.91	0.225
Time (min)	1	54.132	0.022	0.0221	0.01	0.927
Speed (rpm)	1	49.402	8.485	8.4847	3.57	0.117
Square	3	36.458	36.458	12.1528	5.12	0.055
Temp. (°C) * Temp. (°C)	1	23.157	23.023	23.0231	9.70	0.026
Time (min) * Time (min)	1	6.725	5.704	5.7040	2.40	0.182
Speed (rpm) * Speed (rpm)	1	6.576	6.576	6.5764	2.77	0.157
Interaction	3	12.688	12.688	4.2293	1.78	0.267
Temp. (°C) * Time (min)	1	1.381	1.381	1.3806	0.58	0.480
Temp. (°C) * Speed (rpm)	1	4.285	4.285	4.2849	1.80	0.237
Time (min) * Speed (rpm)	1	7.022	7.023	7.0225	2.96	0.146
Residual Error	5	11.873	11.873	2.3746	-	-
Lack-of-Fit	3	6.762	6.762	2.2539	0.88	0.570
Pure Error	2	5.111	5.111	2.5556	-	-
Total	14	166.330	-	-	-	-

Key: DF = Degree of freedom, Adj = Adjusted, SS = Sum of square, MS = Mean square

3.2.5.3. Normal probability and residual plots

Residuals indicate differences between the observed results from experimentation and the predicted results from the model. The normal probability graph plots the residuals versus their expected values when at a normal distribution. From Figure 7, it can be seen that there is no evidence of the existence of non-normality (not a straight-line pattern), skewness (curve in line tails), outliers (a point far away from the line) and unidentified variable (changing slope) as the residuals appear to follow a straight line hence confirming the validity of the model.

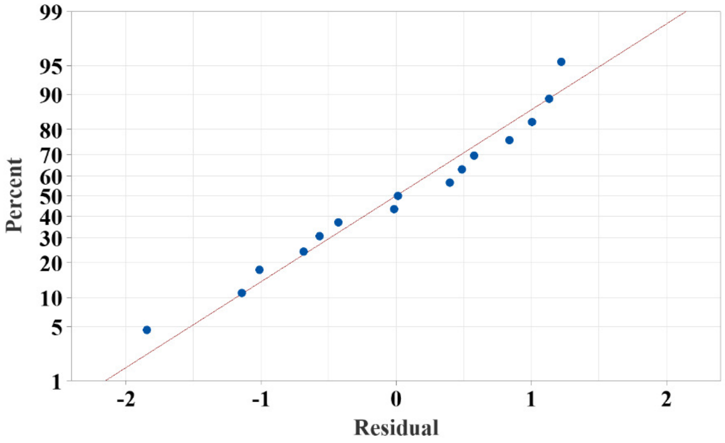

Figure 7: Normal probability plot for response (% base oil extraction yield)

Similarly, the residual plots (Figure 8) were scattered randomly about zero indicating no evidence of the existence of non-constant variance (uneven spreading of residuals across fitted values), missing higher-order terms (curvilinear-curve pattern) and outliers. This suggests that there is no need to consider including the higher-order terms of predictors to get a better fit.

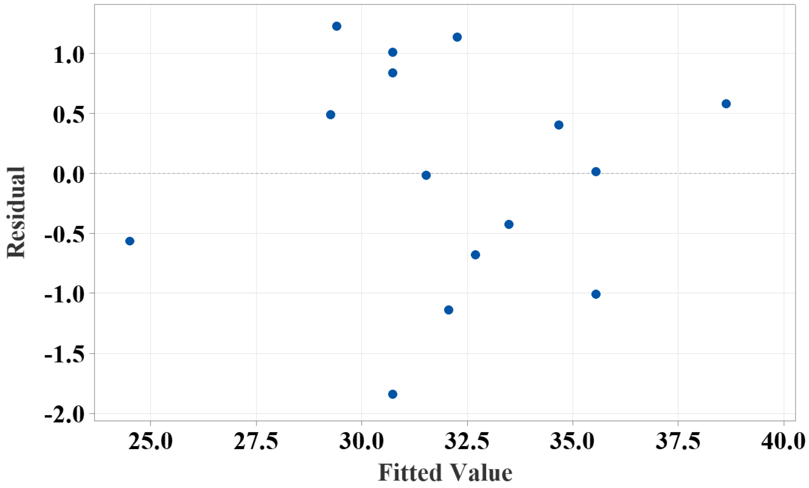

Figure 8: Residual plots for extraction yield (wt.%)

3.2.6. Typical response optimization of base oil yield at a set target

To determine the settings that optimizes the extraction yield of lubricating oil bases from spent lubricating oil using the operating conditions of temperature, extraction time and mixing speed as factors and ethanol as solvent, a typical response optimization was carried out using Minitab® 16.2.4 optimizer. Note that the set target and boundaries chosen for extraction yield must fall within the yield range obtained from experimentation outside which the optimization results would be considered invalid. A goal of 39 wt.% yield was set as a target within a lower and upper boundary of 30 and 40 wt.% yield respectively as shown in Table 5.

Table 5: Optimization parameters to get a target of 39 wt.% extraction yield using a lower and upper boundary of 30 and 40 wt.% respectively

Parameters	Goal	Lower	Target	Upper	Weight	Importance
Extraction yield (wt.%)	Target	30	39	40	1	1

The factors comprise a temperature range of 30-70°C, mixing time range of 10-30 minutes and mixing speed range of 200-300 rpm as used in the experimental design. Table 6 revealed 9 local solutions which signify the "best" combination of factor settings found within the variable range that could achieve the set target. The best local solution also referred to as the global solution is the best combination of factor settings that achieves the desired response (extraction yield).

Table 6: Summary of the local and global solution to get a target of 39 wt.% extraction yield using a lower and upper boundary of 30 and 40 wt.% respectively

Solution	Process conditions			Predicted response		
	Temp. (°C)	Time (min)	Speed (rpm)	Yield (wt.%)	Desirability	Composite desirability
Local 1	69.6406	25.5556	200.910	38.9977	0.999746	0.999746
Local 2	70.0000	22.8393	200.115	38.9819	0.997989	0.997989
Local 3	30.0000	22.6367	200.000	35.6489	0.627660	0.627660
Local 4	70.0000	30.0000	300.000	34.7971	0.533009	0.533009
Local 5	30.0000	30.0000	300.000	34.7496	0.527731	0.527731
Local 6	30.0000	22.8703	200.120	35.6401	0.626673	0.626673
Local 7	69.4671	30.0000	200.000	39.0000	1.000000	1.000000
Local 8	30.0000	30.0000	300.000	34.7496	0.527731	0.527731
Local 9 (Global)	70.0000	22.8807	200.000	39.0000	1.000000	1.000000

From Table 6, it is observed that local solutions 3, 4, 5, 6 and 8 have lower yields of 35.6489, 34.7971, 34.7496, 35.6401 and 34.7496 wt.% respectively which are below the set target of 39 wt.%, hence the solutions are discarded as their desirability of 0.627660, 0.533009, 0.527731, 0.626673 and 0.527731 respectively are considered low compared to others. Local solutions 1 (38.9977 wt.%), 2 (38.9819 wt.%), 7 (39.0000 wt.%) and 9 (39.0000 wt.%) have high desirability of 0.999746, 0.997989, 1.000000 and 1.000000 respectively on a scale of 0 to 1, hence satisfy the response target. However, it could be noticed that solution 1 was achieved at a longer extraction time of 25.5556 minutes and a higher mixing speed of 200.910 rpm while solution 7 at an even longer time of 30 minutes. Although these two solutions achieved the set target, they are considered unsuitable as working under these operating conditions would impact negatively on process economics from the point of higher energy consumption due to longer extraction time and increased mixing speed. Solutions 2 and 9 have similar settings for temperature and time, however, 9 has the advantage of lower energy consumption due to a decrease in mixing speed and is therefore considered the global solution with temperature, mixing time and mixing speed of 70.00ºC, 22.88 min. and 200.00 rpm respectively and a desirability of 1.00.

The optimization plot for the global solution (Figure 9) showed that increasing the extraction temperature initially moves the extraction yield farther away from its target of 39 wt.% and then subsequently closer to the target denoted by the initial downward and subsequent upward curved line. Increasing extraction time also moved the yield closer to its target. However, increasing mixing speed moved the extraction yield farther away from its set target denoted by the downward steep slope. This implies that the optimization of the set target is achieved at a lower mixing speed (200 rpm), moderate extraction time (22.88 min.) and high extraction temperature (70.0ºC) as shown by the red vertical line and numbers which represent the current optimized factor settings.

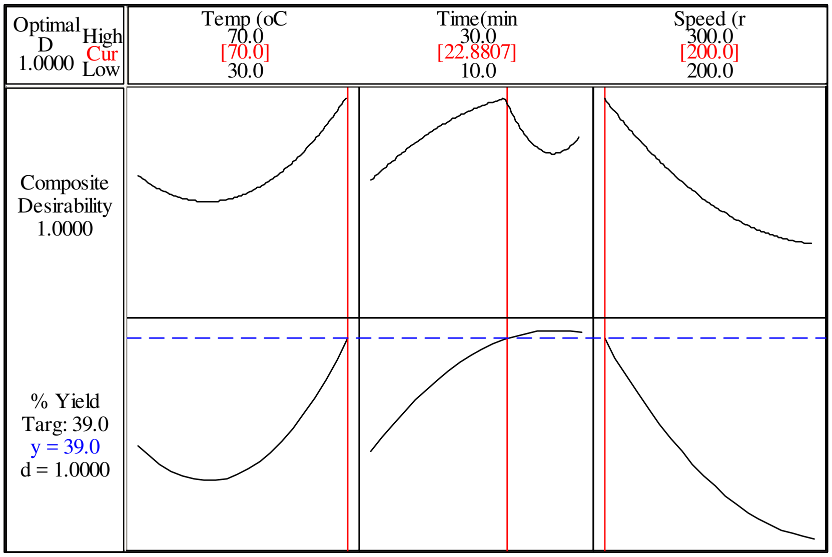

Figure 9: The optimal parameters to get a target of 39 wt.% extraction yield using a lower and upper boundary of 30 and 40 wt.% respectively

3.2.7. Properties of re-refined lubricating oil as compared with SAE standards

After re-refining and analyzing the base oil, its suitability for use as a lubricant was determined by comparing its properties with the widely used standard specified by the Society of Automotive Engineers (SAE) as shown in Table 7. The difference in specification for the various SAE classifications of lubricating oil is determined by the most important property i.e. the viscosity of the lubricant measured at 100ºC (Rizvi, 2009). Lubricating oil's viscosity increases with increasing SAE number. This implies that engines with SAE 30 specification require lube oils with higher viscosity compared to those of SAE 20 specification.

Table 7: Comparison of the properties of re-refined spent lube oil bases with spent lube oil and SAE

Parameter	Spent lube oil	Re-refined lube oil base	Standard grade lube oil[a]			Lube oil base stocks[b]		
			SAE 20	SAE 30	SAE 40	100N	250N	500N
Density (g/mL) @ 15 °C	0.9731 ± 0.0059	0.9590 ± 0.0037	0.8700	0.8801	0.8826	0.8600	0.8720	0.8880
Specific gravity @ 15 °C	0.9004 ± 0.0055	0.8924 ± 0.0034	–	–	–	–	–	–
API gravity	25.6627 ± 0.9601	27.0632 ± 0.6084	–	–	–	–	–	–
Kinematic viscosity @ 40 °C, cSt	146.12 ± 0.27	37.36 ± 0.38	37	88	110	21.5	56.1	107
Kinematic viscosity @ 100 °C, cSt	19.34 ± 0.12	7.63 ± 0.11	5.6 - <9.3	9.3 - <12.5	12.5 - <16.3	3.9 - 4.3	6.5-7.7	11.0
Viscosity Index	129 ± 1	150 ± 1	95	94	96	95	95	95
Flash point, °C	222	194	224	264	268	193	210	221
Pour point, °C	-8	-11	-10	-1	-12	-9	-9	-9
Appearance	Black and homogenous	Dark brown and homogenous	Clear and homogenous			Clear and bright		

[a]Bridjanian and Sattarin, 2006; ExxonMobil, 2017.[b]Rizvi, 2009; Ogbeide, 2010; Behran oil co., 2018.

3.2.7.1. Density, relative density and API gravity

Density is simply the mass of a lubricant sample per unit volume at a specific temperature while relative density or specific gravity shows the ratio of the mass of a given volume of liquid sample to the mass of an equal volume of pure water at a specified temperature. The gravity specified by the American Institute of Petroleum (API gravity) is a special function of specific gravity at 15°C (60°C/60°F). For a lubricant, the knowledge of these properties is vital as it indicates the energy that would be required to pump the lubricant (Rizvi, 2009). 0.9590 mg/L, 0.8924 and 27.0632 were determined as the density, specific gravity and API gravity of the re-refined lube base oil respectively. Although slightly higher than the SAE standard, they were found to be more comparable to the standard compared to those obtained for the spent lube oil i.e. 0.9731 mg/L, 0.9004 and 25.6627 respectively for density, specific gravity and API gravity. This shows that a considerably lower amount of energy would be required to pump the re-refined oil in engines compared to that required for its spent.

3.2.7.2. Viscosity and viscosity index

The viscosity of an oil lubricant represents its level of resistance to flow. It is the most important controlling property to consider when manufacturing or choosing a lubricant for a particular application (Speight, 2002; American Society of Heating Refrigerating and Air-Conditioning Engineers (ASHRAE), 2010). The kinematic viscosity of the re-refined lube oil base at 100°C was 7.63 cSt with an improvement of about 11.71 cSt from its spent whose viscosity is 19.34 cSt. This drop-in resistance to flow resulted in a kinematic viscosity that meets the SAE-20 specification for lube oils and the specification for 250N base oil stock who's minimum and maximum kinematic viscosity range are 5.6-9.3 cSt and 6.5-7.7 cSt respectively (Rizvi, 2009; ExxonMobil, 2017; Behran oil. Co., 2018). This implies that the re-refined lube oil's viscosity is enough to adequately lubricate the moving parts of the specified automotive engine across its various operating temperatures. A similar trend was noticed at 40°C where the kinematic viscosities of the re-refined and spent lube oils were 37.36 and 146.12 cSt respectively. Viscosity index (VI) measures the lube oil's sensitivity to temperature (Sote, 2003). It is an arbitrary number used in characterizing the variation of the kinematic viscosity of a petroleum product with temperature and was determined as 150. This high viscosity index indicates that the effect of temperature on the lubricant's kinematic viscosity would be small (Institute of Petroleum, London, 1993). It also suggests the absence of aromatic and volatile compounds which have a low viscosity index (Hamawand *et al.*, 2013; Rizvi, 2009). Although this was observed to be greater than the specifications of monograde oils (i.e. SAE-20, 30 and 40) with typical VI ranging between 85 and 105, it however falls within the VI range of multigrade oils whose VI is within 130-190 (Speight, 2002). This therefore suggests that multigrade automotive oils (e.g. SAE 20W/50) constitute a larger portion of the spent lubricating oil sample collected within the Bauchi metropolis. This offers an advantage as it assures proper lubrication of engines by the re-refined lube oil across a broad range of temperatures since the change in its kinematic viscosity would be minimal.

3.2.7.3. Pour point

The pour point indicates the lowest temperature at which the lube oil will flow when cooled and examined under specified conditions. It shows the minimum temperature at which the lubricant could be stored and still be capable of flowing under low forces. The pour point of the re-refined lube base oil was improved from -8^{o}C in spent lube oil to -11^{o}C in re-refined lube oil. This indicates the ability of the lubricating oil to flow while stored or in use even during cold (winter) periods.

3.2.7.4. Flash point

The flash point represents the highest temperature at which the lubricating oil can be stored and handled without causing serious fire hazards. A low flash point minimizes the loss of a lubricant through evaporation at high temperatures and lowers the hazards during handling and use (Rizvi, 2009). The flash point of the re-refined lube oil was 194^{o}C.

3.2.7.5. Visual appearance

The appearance of the re-refined lubricating oil was observed to be dark brown. This suggests a possible extraction of other dark (black) pigments resulting from the mixing of these oils with the by-products of combustion fuels. This agrees with the findings of Osman *et al.* (2017) who regenerated spent lubricating oil using composite solvents (Toluene, 1-butanol and methanol, Toluene, 1-butanol and ethanol, and Toluene, 1-butanol and isopropanol). However, the color could be improved using activated alumina at a proportion 1:5 alumina (Osman *et al.*, 2017).

4. CONCLUSION

In this work, spent lubricating oil was collected from different points and homogenized to represent a typical feedstock for a re-refining plant. From the main parameters influencing the rate of base oil extraction in the re-refining of spent lubricating oil using liquid-liquid extraction, temperature, mixing time and speed were investigated, while solvent type (ethanol), solvent-oil mass ratio (10:1), spent oil mass (5 g) and KOH-concentration (2 g KOH/Kg) were kept constant. Optimization of the base oil extraction yield by varying temperature, mixing time and mixing speed at solvent-oil mass ratio (10:1) achieved an optimal base oil recovery of 39.21 wt.%. ANOVA for the response surface quadratic model fitting revealed the *p-value* regression ($p = 0.021$) to be statistically significant and, thus, is sufficient to explain the variation in base oil extraction yield. Additionally, there was no evidence of lack of fit as the lack of fit *p-value* ($p = 0.570$) was found to be statistically insignificant. Viscosity, being the most important property in lubricant manufacture and selection, met the SAE standard as it was found to be 37.36 and 7.63 cSt at 40^{o}C and 100^{o}C respectively, thus considered suitable for use as lubricant. Overall, the re-refining process proved feasible as ethanol solvent recovered up to 39.21 wt.% of the active base oil content of the spent oil. The condition under which this process must be carried out to achieve high yield is 10:1 solvent to oil mass ratio, 70^{o}C extraction temperature, 20 minutes mixing time and 200 rpm mixing speed.

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6. CONFLICT OF INTEREST

There is no conflict of interest associated with this work.

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