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OPTIMIZATION OF THE PRODUCTION OF LUBRICATING OIL FROM RE-REFINED USED LUBRICATING OIL USING RESPONSE SURFACE METHODOLOGY

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ABSTRACT

Response surface methodology was successfully employed in the optimization of the production of lubricating oil from re-refined used lubricating oils. The re-refined lubricating oil was obtained from caustic treatment and vacuum distillation of used automotive gasoline engine lubricating oils. A 2^3 factorial design augmented with seven experimental points to form an orthogonal central composite design (CCD) to accommodate all second order effects was employed in this study. Lubricating oil additives namely oxidation/corrosion inhibitor, detergent and dispersant were used as independent variables. The extent of deterioration of the lubricating oil was investigated using neutralization number, sludge deposition and corrosion of carbon steel as responses. Optimal additive levels of 0.99% (v/v) oxidation/corrosion inhibitor, 1.33% (v/v) detergent and 3.0% (v/v) dispersant; were used with a blend of re-refined used lubricating oil and virgin bright lube stock in the ratio 3:1 to obtain an SAE 40 grade lubricating oil suitable for automotive gasoline engines.

Keywords: lubricating oil, response surface methodology, central composite design, optimization, neutralization number.

INTRODUCTION

Large amounts of used lubricating oils are generated in Ghana from automobile engines, plant and machinery, transformers, etc. Unfortunately however, apart from only a small percentage that is used as form oil to lubricate the inside of form work in the construction industry, the large quantity of used oil generated is dumped around the cities in drains which ultimately contaminate and pollute water bodies including rivers, lagoons streams, etc. Examples of water bodies polluted by the dumping of used lubricating oils, greases, etc; from automobile service garages abound and these include the Odaw River and Korle Lagoon in Accra, Fosu Lagoon in Cape Coast, etc.

Waste lubricating oils contain a lot of metal contaminants like calcium, magnesium, copper, lead, iron, etc; [1-4] and the current disposal practice of just dumping used oils in drains and rivers and lagoons poses serious environmental and health hazards. Thus, it is imperative to find more environmentally sustainable ways of disposing of the used lubricating oils or recycling them. It is worthy of note that lubricating oils are only partially consumed during their service while their quality is degraded by oxidation and decomposition of the mineral oil and/or additives and by contamination by such components as gasoline, dirt, metallic particles and carbon as soot [4, 5]. A high proportion of the used oil consists of high quality hydrocarbons in the original oil and recovery and reuse of these hydrocarbons provide an opportunity to reduce significantly the need for the production of virgin lube blending stock. In Ghana, all the lubricating oil is imported and huge sums of foreign exchange are required to import these lubricating oils for automobile engines as well as for plant and machinery. Economic and environmental considerations demand that it is worthwhile to explore the possibilities of reclaiming used lubricating oils to produce lube blending stock to supplement blending stock imports. The objective of this paper among others is to investigate and develop the optimal levels of additives to be added to lube blending stock obtained from re-refined used lubricating oils using experimental design techniques namely, response surface methodology.

Response surface methodology (RSM) has been one of the most popular optimization techniques in recent years and its applications in various fields including chemical, biochemical, metallurgical, petrochemical processes, food processing, to mention only a few; abound.

RSM is a collection of statistical and mathematical techniques that are useful for modeling and analysis of problems in which the response of interest is influenced by several parameters and the objective is to optimize the response [6]. The application of RSM can often be used to achieve the objectives of quality improvement, including reduction of variability and improved process and product performance [7].

Concept of response surface methodology

This section will give a brief introduction to the concept of RSM as developed over the years by earlier workers including Box and Wilson [7], Cox and Cochran [8], Box, Hunter and Hunter [9], Box and Draper [10, 11].

RSM defines the effect of the independent variables alone or in combination on the processes or more specifically on the performance measure or the quality characteristics called the response of the processes. In general, a statistical modeling technique is employed to develop an appropriate approximating model between the response and the independent variables such that [6]:

$$\eta = f(x_1, x_2, \ldots\ldots, x_n) + \varepsilon \quad (1)$$

where η is the response, f is the unknown function of the response, $x_1, x_2, \ldots\ldots, x_n$ are the independent variables

(also called the natural variables, especially when expressed in the natural units of measurement such as degree Celsius (^oC), Pascal (Pa), pound per square inch (psi), etc); and ε is the statistical error which represents other sources of variability not accounted for in f. The sources of error usually may include measurement error on the response, background noise, the effect of other variables, etc. In general, ε is assumed to have a normal distribution with mean zero and variance σ^2 [6].

Three main stages namely (i) the determination of independent variables and their levels, (ii) the selection of the experimental design, prediction and verification of the model equation, and (iii) obtaining the response surface plot and contour plot of the response as a function of the independent variables and the determination of the optimal points [9, 12] are essential when employing RSM in optimization studies for the development of optimal systems. These three stages of analysis/study were employed in this study to optimize the production of lubricating oil from re-refined used automotive engine lubricating oils.

Lubricant characteristic parameters

Lubricating oil may be classically defined as a substance capable of reducing friction heat, wear, etc; when introduced as a film between solid surfaces [13, 14]. Its function among others may be summarized as follows:

a) lubricate by forming a fluid film between highly loaded moving parts;
b) receive and carry away contaminants from both internal and external sources;
c) protect against wear of highly loaded parts under conditions of boundary lubrication;
d) protect against the accumulation of deposits, sludge and vanish in lubricating systems; and
e) protect against rust, corrosion of precision parts of varying metallurgy.

MATERIALS AND METHODS

Experimental work

A 2^3 factorial design augmented with seven experimental points to form an orthogonal central composite design (CCD) to take care of all second order effects was employed in this study. The experimental points are shown in Table-1. Minitab® Release 14 software with experimental study variable number K = 3, for independent variables including oxidation/corrosion inhibitor (X_1), detergent (X_2) and dispersant (X_3) was used for the design and analysis of the results. The response surfaces were subsequently approximated with a second-order polynomial equation of the form:

$$Y=\beta_o+\beta_1X_1+\beta_2X_2+\beta_3X_3+\beta_{12}X_1X_2+\beta_{13}X_1X_3+\beta_{23}X_2X_3+\beta_{11}X_1^2+\beta_{22}X_2^2+\beta_{33}X_3^2 \quad (2)$$

where Y is the predicted response, β_0 the constant (intercept), β_i the linear coefficient, β_{ii} the quadratic coefficient and β_{ij} the cross product coefficient. X_i and X_j are the coded independent variables ranging from -1 to +1 so that they affected the responses more evenly [6]. The following equation was used to effect the coding.

$$X = [2x-(x_{max}+x_{min})]/[x_{max}-x_{min}] \quad (3)$$

where x is the independent variable in natural units such as degree Celsius (^oC), Pascal (Pa), percent volume-by-volume (%, v/v), etc; and X is the coded variable while x_{max} and x_{min} are the maximum and minimum values of the independent variables, respectively.

Sampling and testing of used oil

The samples of used lubricating oil used in the study were drained from the crankcase of cars that had used SAE 20W-50 grade automobile lubricating oil and travelled on the average 6000 kilometers.

The used crankcase lubricating oil collected was initially tested to establish the level of deterioration of the oil while in used in the automobile engine. The experimental procedure followed among others included: the characterization of the used oil, re-refining of the used oil, characterization of the re-refined oil, treatment of the blended lube stock with additives, and oxidation stability test of additive treated blended lube stock. The tests and methods of characterization were American Society for Testing and Materials (ASTM) and /or Institute of Petroleum (IP) Standard Test Methods.

Table-1. Three-factor central composite design and responses.

Trial		Independent variables (Level codes)			Responses		
		X_1	X_2	X_3	**Neutralization number** (mg KOH/g)	**Sludge deposition** (10^{-4} g/cm^2) ‡	**Corrosion of carbon steel** (10^{-4} g/cm^2) †
2^3 Factorial	1	-1	-1	-1	1.296	52	-4
	a	+1	-1	-1	1.313	32	-6
	b	-1	+1	-1	1.462	38	-6
	ab	+1	+1	-1	1.149	19	8
	c	-1	-1	+	2.690	25	-6
	ac	+1	-1	+1	2.837	43	1
	bc	-1	+1	+1	2.147	71	1
	abc	+1	+1	+1	1.478	101	4
Augmenting points	9	0	0	0	1.942	20	-12
	10	+1	0	0	1.410	29	5
	11	-1	0	0	1.283	-37	5
	12	0	+1	0	1.519	22	12
	13	0	-1	0	1.166	10	-7
	14	0	0	+1	1.524	-16	25
	15	0	0	-1	1.423	13	5
SAE 40 (Mobil Oil)					1.5	17	2

‡Negative sludge deposition indicates there was loss in weight of the metal. †Negative corrosion on carbon steel indicates an increase in weight of the catalyst.

Re-refining of used lubricating oil

The used oil drained from the crankcase of cars was re-refined using the caustic treatment/distillation process. First, the used oil was heated to about 300^{o}F (149^{o}C) with stirring and treated with 1 % (w/w) sodium hydroxide pellets to dehydrate, remove all fuel dilution and to neutralize all acids. The caustic treatment also aided in the breaking of oil-water emulsion and allowed the easy precipitation of solids contained in the used oil. This was followed by vacuum distillation and the recommended ASTMD 1160-77 standard method for distillation of petroleum products at reduced pressure was used. Distillation was carried out at 65 mmHg (86.7 mbar) to produce lube stock and the yield was estimated to be 70% of input used oil.

Selection of additives

Chemical, biochemical, metallurgical processes and in general all processes are affected by several parameters. In a study such as this, it is impossible to identify the effects of all parameters and it is therefore imperative to select a limited set of parameters that have major effects on the processes or mechanism under study. A screening experiment was conducted to elucidate the effect of each factor on those of the others and on the response(s). Wagner and Gorman as far back as 1962 suggested factorial designs and simplex lattice designs for specification of treatment combinations as especially useful in experiments with fuels, lubricants and engines [10, 11]. Factorial designs were therefore employed to assist in the determination of the independent variables (additives) in this investigation.

Automobile engine lubricating oil mainly contains a base stock largely comprising of hydrocarbons whose lubricating properties are enhanced by motor oil additives. The main functions of motor oil additives are to: (i) reduce wear, corrosion and oxidation, (ii) control deposition of lacquer and sludge, and (iii) modify physical properties including viscosity index, pour point, etc; and chemical properties like neutralization number of base oils; and these informed the selection of the set of independent variables.

The following additives - oxidation/corrosion inhibitor (x_1), detergent (x_2) and dispersant (x_3) were selected as independent variables and used within the recommended applications ranges of 0.38 - 1.14 (%, v/v), 1.0 - 2.0 (%, v/v) and 1.0 - 5.0 (%, v/v), respectively. The additives were supplied by Lubrizol Corporation, Ohio, USA.

Response selection

The responses considered in the study were (i) neutralization number, (ii) sludge deposition and (iii) corrosion of carbon steel catalyst. All three responses are indicative of the extent of oxidation or deterioration of oil [13] and oxidation stability test was carried on the samples to determine the selected responses.

Oxidation stability test

The recommended IP 48/67 standard test for testing oxidation stability of lubricating oils was adopted. The STANHOPE-SETA Series 1660 apparatus specially designed for testing oxidation stability of lubricating oils was used. Specially prepared catalysts - carbon steel, copper and aluminum plates tied into a triangular configuration were used to simulate the materials for the fabrication of automobile engine block and bearings.

Neutralization number determination

Oxidation is the primary cause of oil deterioration and forms acidic products which increase the acidity of used oils. Hence the neutralization number gives an indication of the extent of oxidation and for that matter oil deterioration [13]. Neutralization number is defined as the weight in milligrams of potassium hydroxide required to neutralize one gram of oil. The recommended IP 136/65 - ASTMD 974-64 standard test method was used for the determination of neutralization number.

Sludge deposition

Sludge deposition was measured to assess the extent of deterioration of the oil. The catalysts were carefully removed from the oxidation tube, cleaned with naphtha and dried in an oven at 90^{o}C for six hours to a constant weight. A Mettler Toledo balance with a precision of ±0.0001g was used in weighing.

Extent of corrosion

The extent of corrosion is also indicative of the extent of deterioration of the lubricating oil. The triangular arrangement of the catalysts was broken and each catalyst gently wiped with a "rubber policeman" to remove the sludge deposit and any corroded metal from the catalyst surface. The catalysts were washed in naphtha, dried in an oven at 90^{o}C for six hours, cooled in a desiccator and weighed using a Mettler Toledo balance with a precision of ±0.0001g.

RESULTS AND DISCUSSIONS

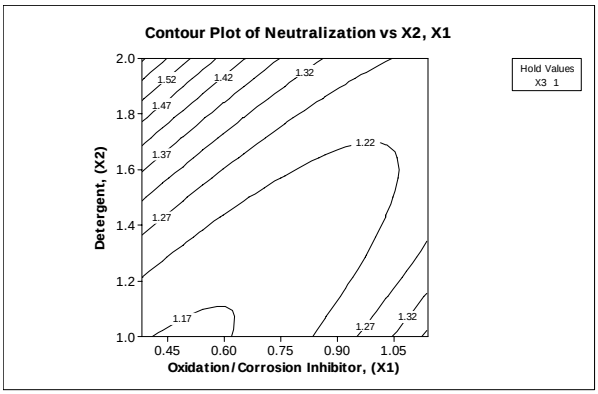
The effect of the independent variables: oxidation/corrosion inhibitor, detergent and dispersant on the performance of lubricating oil produced from re-refined used lubricating oils was studied using Minitab® Release 14 (Minitab Inc., USA) software.

Effect of treatment factors on neutralization number

The response surface regression model obtained for neutralization number of the lubricating oil was:

$$Y = 1.5887 - 0.0691X_1 - 0.2596X_2 + 0.4033X_3 - 0.14325X_1X_2 + 0.02825X_1X_3 - 0.2380X_2X_3 - 0.1539X_1^2 + 0.3665X_2^2 - 0.0269X_3^2$$ with $R^2 = 84.1\%$.

There was a significant ($p \leq 0.05$) quadratic effect of oxidation/corrosion inhibitor and detergent on the neutralization number of the lubricating oil. The analysis also showed a linear effect of the detergent and dispersant on neutralization number. The relatively small value of the coefficient of X_1 compared to those of X_2 and X_3 suggests an attenuation along the X_1 axis and that there is a stationary ridge along this axis. The practical significance of this is that for very large increases along the X_1 axis a very small change in neutralization number is achieved. This is markedly shown in Figure-1(a) at $X_3 = -1$. The elongation of the response surface along the X_1 axis also indicates that there is dependence between the variables and that there is some synergism in the variables (factors). The model could explain about 84.1% of the variations in neutralization, thus, about 15.9% of the observed variations in neutralization number could be due to other factors that were not included in the model. Figure-1 (a)-(c) show the contour plots of neutralization number versus oxidation/corrosion inhibitor, (X_1) and detergent, (X_2) at fixed values of the dispersant, (X_3) = -1, 0 and +1 for Figure-1(a), Figure-1(b) and Figure-1(c) respectively.



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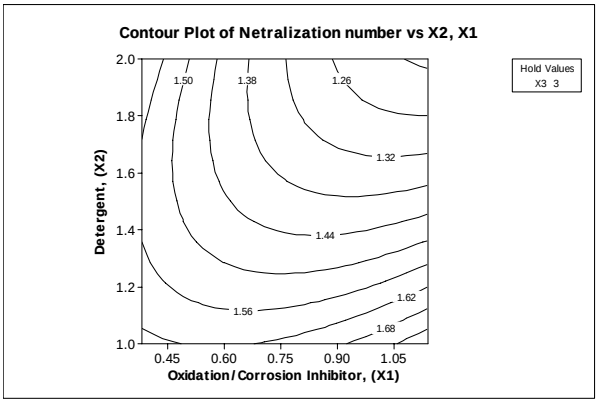

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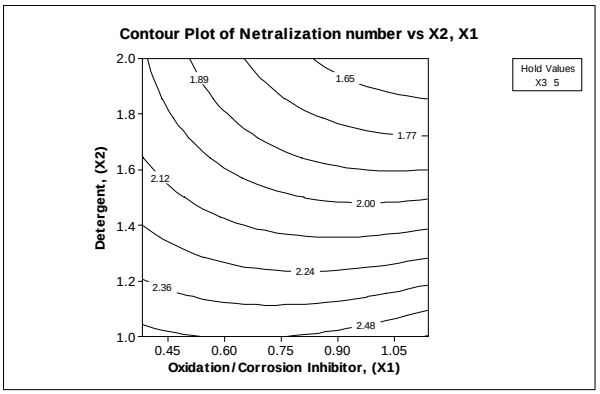


(c)

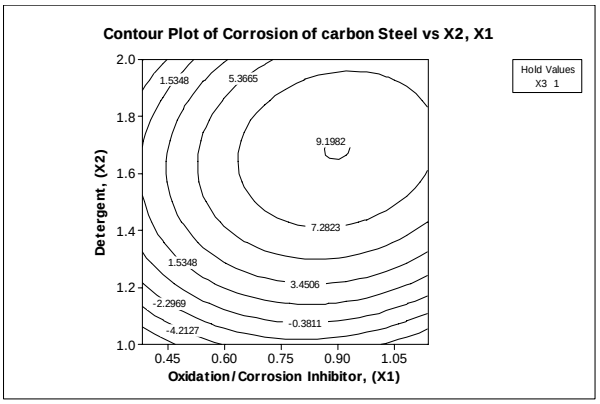
Figure-1. Response surface contours for neutralization number mgKOH/g at (a) x_3 = 1% v/v dispersant, (b) x_3 = 3% v/v dispersant and (c) x_3 = 5% v/v dispersant.

Effect of treatment factors on corrosion of carbon steel

The response surface regression model obtained for corrosion of carbon steel was:

$Y = 5.36 + 2.20X_1 + 3.835X_2 + 2.80X_3 + 1.50X_1X_2 - 0.25X_1X_3 - 0.25X_2X_3 - 4.70X_1^2 - 5.875X_2^2 + 5.30X_3^2$ with $R^2 = 40.9\%$.

There was a significant ($p \leq 0.05$) quadratic effect of all three factors - oxidation/corrosion inhibitor, dispersant and detergent on the corrosion of carbon steel in the lubricating oil. The analysis also showed a linear effect of the three factors on corrosion of carbon steel with the effect of the detergent being most significant. The model could explain about 40.9% of the variations in corrosion of carbon steel, thus, about 59.1% of the observed variations in corrosion of carbon steel could be due to factors other than those that were not included in the model. Figure-2 (a)-(c) show the contour plots of corrosion of carbon steel versus oxidation/corrosion inhibitor, (X_1) and detergent, (X_2) at fixed values of the dispersant, (X_3) = -1, 0 and +1 for Figure-2(a), Figure-2(b) and Figure-2(c), respectively.



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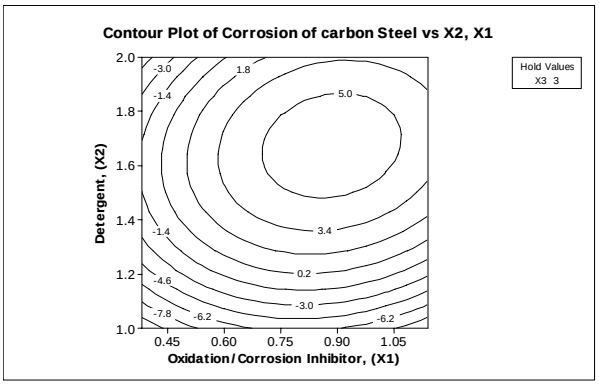

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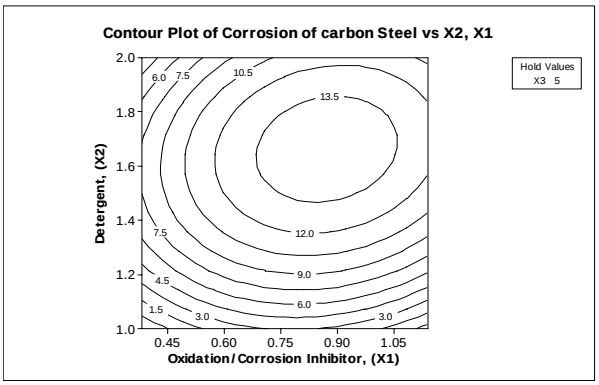


(c)

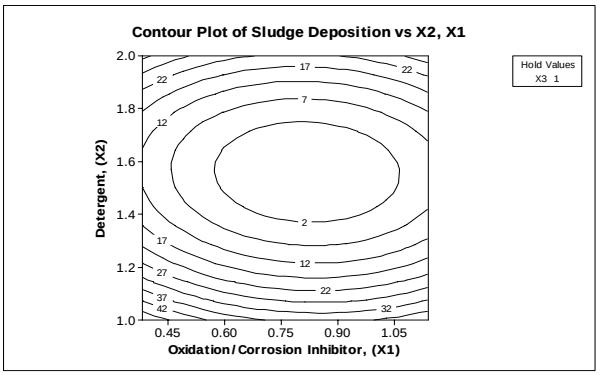
Figure-2. Response surface contours for corrosion of carbon steel (10^{-4} g/cm^2) at (a) x_3 = 1% v/v dispersant, (b) x_3 = 3% v/v dispersant and (c) x_3 = 5% v/v dispersant.

Effect of treatment factors on sludge deposition

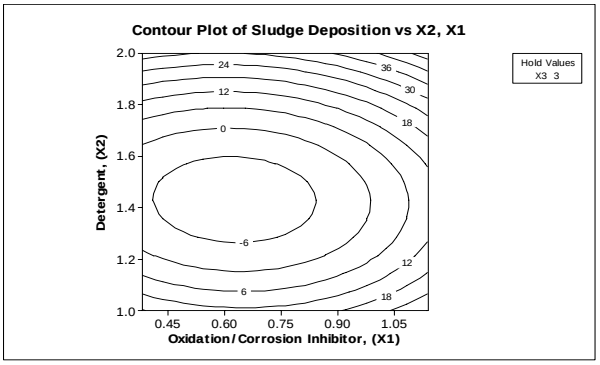
The response surface regression model obtained for sludge deposition was:

$Y = -36.188 - 13.540X_1 + 116.934X_2 - 8.270X_3 - 21.353X_1^2 - 40.645X_2^2 + 24.231X_3^2$ with $R^2 = 91.1\%$.

There was a significant ($p \leq 0.05$) quadratic effect of all three factors - oxidation/corrosion inhibitor, dispersant and detergent on the sludge deposition in the lubricating oil. However, the quadratic effect of the detergent is the most significant. The analysis also showed strong linear effect of the detergent (X_2) on sludge deposition in the lubricating oil. The model could explain about 91.1% of the variations in sludge deposition, thus, about 8.9% of the observed variations in sludge deposition may be due to factors that were not included in the model. Figure-3 (a)-(c) show the contour plots of corrosion of sludge deposition in the lubricating oil versus oxidation/corrosion inhibitor, (X_1) and detergent, (X_2) at fixed values of the dispersant, (X_3) = -1, 0 and +1 for Figure-3(a), Figure-3(b) and Figure-3(c), respectively.



(a)



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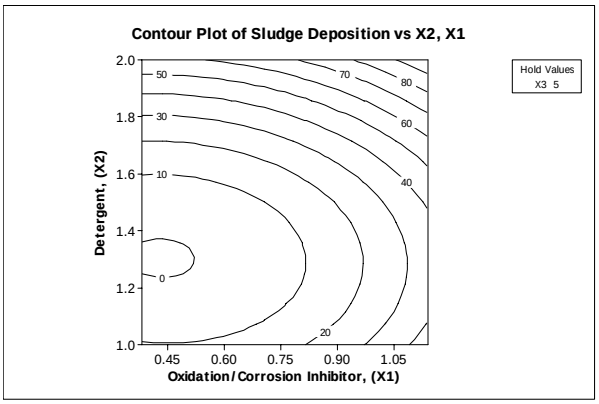

(c)

Figure-3. Response surface contours for sludge deposition in lubricating oil (10^{-4} g/cm^2) at (a) x_3 = 1% v/v dispersant, (b) x_3 = 3% v/v dispersant and (c) x_3 = 5% v/v dispersant.

Optimal experimental region

As noted earlier, a lubricant is a multifunctional product and to assess its quality it is necessary to measure as many significant responses as possible. Three responses have been considered in this study to investigate the nature of synergism inherent in a compounded product like a lubricant.

Table-2. Responses of treatment combinations in the optimal region.

Treatment combination	Independent variables			Responses					
				Neutralization number (mg KOH/g)		Sludge deposition (10^{-4} g/cm^2)		Corrosion of carbon steel (10^{-4} g/cm^2)	
	X_1	X_2	X_3	Predicted value	Experimental value	Predicted value	Experimental value	Predicted value	Experimental value
1	0.7	-0.5	0	1.552	1.527	5.5	10	0.33	6
2	-0.75	0.5	0	1.497	1.461	14.2	16	0.27	6
3	0.6	0.5	0	1.227	1.309	14.9	21	5.55	3
4	0.7	0.0	0	1.370	1.284	5.1	14	4.95	5
5	-0.6	0.35	0	1.448	1.532	8.3	24	3.69	3
6	0.6	-0.35	0	1.492	1.475	2.9	6	2.97	2
7	0.85	0.15	0	1.320	1.342	9.6	20	3.69	2
8	1.05	0.25	0	1.290	1.363	15.5	11	0.91	4

The quality of the oil can be associated with the total synergistic effect of the mineral oil (i.e., the base stock), and the individual additives used in the formulation of the final blend. Following the procedure of Lind, *et al*. [15] superimposition of the response surfaces was done to obtain the optimal experimental region. Further experimentation in the optimal experimental region led to a more precise estimate of the optimum additive package. Table-2 gives the treatment combinations and their responses in the optimal region shown in Figure-4.

Constraints in the responses used in obtaining the optimal region were chosen with reference to values obtained for new SAE 40 (Mobil) oil in the oxidation stability test.

Thus the 'standard' values were 1.5 mg KOH/g oil, $17x10^{-4}$ g/cm^2 and $2x10^{-4}$ g/cm^2 for neutralization number, sludge deposition and corrosion of carbon steel respectively. While the differences in individual responses for treatment combinations 4 and 6 in Table-2 are marginal the additive requirement for treatment combination 4 is greater than that of treatment 6. Thus, the optimal additive package for the formulation of lubricant obtained in this investigation using a base stock with viscosity index (VI = 110) on volume-by-volume basis was as follows: base stock (94.79%), oxidation/corrosion inhibitor (0.988%), detergent additive (1.325%) and dispersant additive (3.0%).

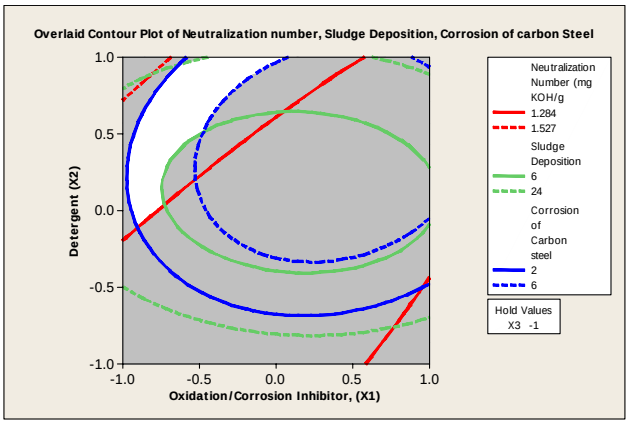

Figure-4. Response surface contours showing optimal experimental region.

CONCLUSIONS

Response surface methodology was an efficient and effective tool in the development of the optimal additive package for a base stock blended at 75% re-refined used oil stock and 25% bright stock. An orthogonal central composite design formed by augmenting a 2^3 factorial design approximated the response surfaces with a second degree polynomial equation with very little lack of fit at the 5% level of significance.

The dispersant additive was observed to have the same general effect on all the responses i.e., in moving along its axis towards the individual centers of the corresponding response surfaces. The elongation of the response surface along X_1 axis indicated dependence between the additives thus confirming the existence of some synergism between the additives used in the investigation. However a general correlation between the responses could not be arrived at.

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