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Optimization of the Extraction Process of Gmelina Seed Oil using Response Surface Methodology

Samuel E. Ogbeide[1], Emmanuel U. Aniekwe[1]*, Chinaza E. Nwanno[2]

Department of Chemical Engineering, University of Benin, P.M.B. 1154, Benin City, Nigeria
Department of Industrial Physics, University of Benin, P.M.B. 1154, Benin City, Nigeria

Abstract The optimization of the extraction process conditions of gmelina seed oil was studied via the response surface methodology using Box Behnken Design. The effect of various process variables such as agitation time (10-60mins), volume of solvent (50-150ml), particle size (150-1000µm), and their interaction on oil yield was investigated. A total of 17 experimental runs were carried out as determined by design of experiment. A Predictive model describing the oil yield in terms of process variables was derived and was found to be a Quadratic model. Optimum yield of 52.09% was obtained at extraction time of 60mins, seed particle size of 150µm and 150ml volume of solvent with a constant extraction temperature of 60°C, for the process. It was found that oil yield increased with increase in agitation time and volume of solvent but decreased with increase in seed particle size. The oil content was significant and would be highly suitable economically for industrial applications.

Keywords Gmelina Seed Oil; Process Optimization; Solvent Extraction; Box-Behnken design

Introduction

The rapidly growing global demand for energy and industrial raw materials from crude oil and the consequent depletion of crude oil reserves in addition to adverse environmental concerns and unstable nature of the international market make imperative the need to explore alternative sources of fuel and industrial raw materials. There has been an increase in the world production of oil seeds over the last 30years [1]. This would appear to be related to the increasing demand for oil seed products and by-products. Most seeds are cultivated primarily for their oil and meal. The oil from most seeds can be used for edible purposes (example is groundnut oil, fluted pumpkin seed oil, soya bean seed oil) and about 80% of the world production of vegetable oil is for human consumption while the remaining 20% is shared between animals and chemical process industries [2].

As a result of the extensive demand of oil for consumption and industrial uses, analysis of many oils has been carried out. Most vegetable seed oils find wide applications in the production of soaps, paints, varnishes, lacquers, lubricants, hydraulic fluids, printing inks, dyes, pesticides, and insecticides [1,3-4]. Also, with increase in petroleum prices and uncertainties surrounding petroleum availability, vegetable oil can be employed as an alternative fuel and for biodiesel production which aims to overcome energy crisis problem [5]. Due to the increasing applications of vegetable oils, a number of seed oils have been characterized but the vast majority have not been adequately evaluated and this is particularly valid for gmelina, which falls into this group of under-utilized species of plant. The ability of a particular oil seed to fit into the growing industry depends on its utilization potential, rate of production and availability of processing technology [6].

Gmelina arborea is a fast growing tree, which grows on different localities and prefers moist fertile valleys, they attain moderate to large height up to 40 m and 140 cm in diameter [7]. It is occurring naturally throughout greater part of India at altitudes up to 1500 m. It also occurs naturally in Myanmar, Thailand, Laos, Cambodia, Vietnam, and in southern provinces of China, and has been planted extensively and is widely available in Sierra Leone, Nigeria and Malaysia [8]. A lot of researches are being carried to find alternative ways of producing oil for process industries and for food industry. It has been found that almost all the seeds contain oil, hence these gives ground for other researchers to consider studies on the possible uses of other oil producing substances found in people's everyday lives.

There are various ways of extracting oil from oilseeds but solid-liquid extraction (Leaching) has been reported to be most efficient technique [9]. Now, gmelina seeds are already proven to produce non-edible oil [10], this fact itself is already useful information for researchers who seek to find alternative sources of oil. The ability of the oil to fit depends on its constituents, its composition, and the rate of production and availability of the processing technology. Response surface methodology has been successfully applied industrially to different processes for achieving its optimization using experimental designs [11-14] and this research is geared towards achieving the experimental set of data for industrial application needed in optimizing the yield in gmelina seed oil extraction process.

2. Materials and Methods

2.1. Materials

The gmelina fruit were collected locally from St. Mary's secondary school, Abagana, Anambra State in Nigeria. It was soaked in water for eight days so as to easily separate the fruit pulp from the seed (de-pulp). The seeds were sun-dried and crushed mechanically using corona blender; the crushed samples were then separated into different particle sizes using laboratory test sieves (150μm, 575μm and 1mm). The samples were then dried using the electric oven to a 5% moisture content, stored in air tight containers and were labeled adequately.

The organic solvent used for the oil extraction was n-hexane. It was purchased from chemical store, behind Fidelity Bank, Ugbowo campus. The reagent was commercial grade and was used without further purification.

2.2. Experimental Design for Extraction Process Optimization

Design expert software from Stat-Ease Inc. Minneapolis, Minnesota, USA was used to obtain the permutation of the different process variables in order to know how the variables will be varied in the performing the experiment. Response surface methodology was utilized in the course of using the software and the three variables Box Behnken was used to obtain the permutation of the different process variables by inputting the range of the variables to be considered as studied below:

Table 2.1: Experimental range of the independent variables for gmelina seed oil extraction

Independent Variable	**Range**	
	-1 level	**1 level**
Seed Particle (μm) (A)	150	1000
Volume of Solvent (ml) (B)	50	150
Time (min) (C)	10	60

By inputting the ranges in the Box Behnken, the software arranges the different runs of experiment to be performed in order to obtain the optimum parameter required for the extraction of oil from gmelina seed. Solid-liquid extraction method was used as against other methods of oil extraction, such as distillation, expression etc, because it gives higher efficiency, up to 95% [15] and can easily be set up in the laboratory. The solvent used in this experiment was n-hexane, because it is relatively cheap, non-toxic and has a tolerable odor.

Particle size, volume of solvent and agitation time was the variables considered in the experiment. The agitation time was varied between 10-60 minutes. The volume of solvent to solid ratio was investigated from 2:1- 5:1 and particle size was varied from 150μm -1000μm. The temperature during the whole experiment was kept constant at 60^oC. The percentage oil yield was calculated using the expression below:

$$Y = \frac{W_0}{W} * 100 \quad (1.0)$$

Where, Y is the oil yield (%),

W_0 is the weight of pure oil extracted (g) and

W is the weight of the sample of gmelina seed used in the experiment.

3. Results and Discussion

3.1. Result of the Experimental Design for Optimizing the Extraction Process Variables as Provided using Box Behnken

Table 3.1: Three variable Box behnken with experimental and predicted responses of the Yield (%)

Runs	Independent Variables			Yield (%)	
	Seed particle(μm)	Volume of solvent (ml)	Time (mins)	Experimental	Predicted
1	575.00	150.00	60.00	38.54	38.89
2	575.00	150.00	10.00	30.75	29.75
3	150.00	100.00	10.00	40.82	39.92
4	1000.00	100.00	10.00	17.93	18.78
5	150.00	50.00	35.00	40.08	39.92
6	150.00	100.00	60.00	52.09	51.24
7	575.00	50.00	60.00	27.15	28.15
8	575.00	100.00	35.00	31.67	31.65
9	575.00	100.00	35.00	31.62	31.65
10	575.00	100.00	35.00	31.65	31.65
11	150.00	150.00	35.00	50.82	51.31
12	1000.00	100.00	60.00	23.02	22.51
13	1000.00	150.00	35.00	22.54	22.70
14	575.00	100.00	35.00	31.65	31.65
15	575.00	50.00	10.00	24.01	23.66
16	575.00	100.00	35.00	31.65	31.65
17	1000.00	50.00	35.00	17.75	17.26

As studied from literature, the yield of oil (leaching operation) is dependent on the particle size, solvent, agitation of fluid and temperature. However, only the first three of these factors were studied in this work. With the aid of a statistically designed experiment, the combined effect of these factors were studied and experiment was conducted at different level of combinations of these parameters as shown in Tables 3.1. The experimental result associated with the interactions between each independent variable as well as the predicted response was also shown in Table 3.1. Minimum response occurs for run 17 and the maximum for run 6. A second order polynomial equation was derived to represent the gmelina seed oil yield as a function of the three parameters varied:

$$\mathbf{Y = 30.97673 - 0.032416A + 0.17989B + 0.17556C - 7.00000E\text{-}005AB - 1.45412E\text{-}004AC + 9.30000E\text{-}004BC + 1.24678E\text{-}005A^2 - 4.40200E\text{-}004B^2 - 6.92800E\text{-}004C^2} \quad (2.0)$$

Where: Y = Gmelina seed oil yield, A, B and C are the values of seed particle **(**μm), Volume of solvent (ml) and Agitation time (mins) respectively. The coefficient of A, B, C and D are the main linear effects of the independent process variables, AB, AC and BC represent the linear interaction effects between seed particle/volume of solvent, seed particle/time and volume of solvent/time, respectively. A^2, B^2 and C^2 are the quadratic effects of the respective process variables. The regression equation was also used to calculate the predicted response presented in Table 3.1. A comparison of the predicted values and the experimental values shows insignificant deviations which imply that these data are in correlation, this is displayed in figure 3.1. It can be observed that all data points aggregate close to the straight y=x line. This point to the fact that the quadratic regression model obtained was able to predict the gmelina seed oil extraction process to a high level of accuracy or confidence. Hence, this equation can be used for both predictive and design purposes.

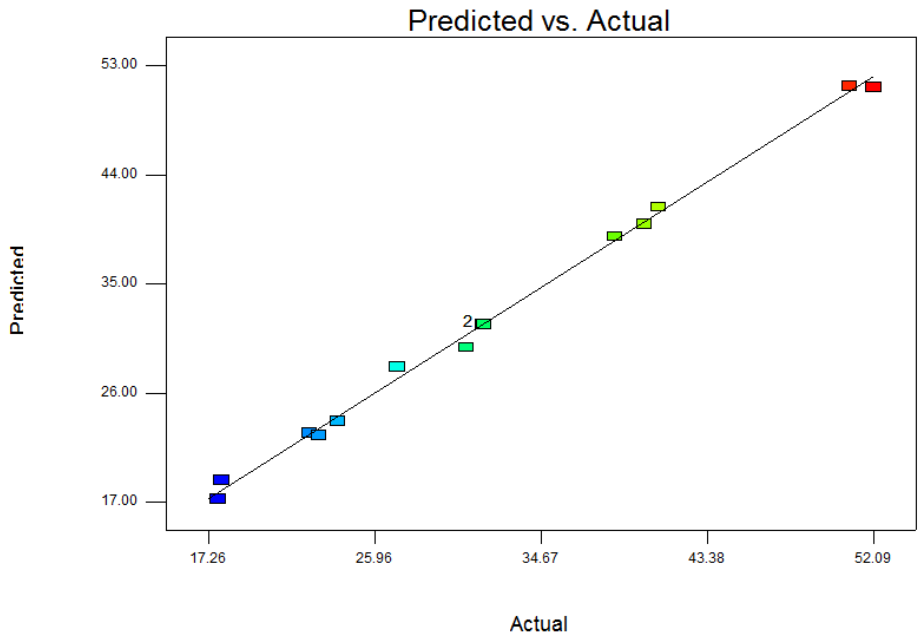

Figure 3.1: Comparison plot showing experimental (actual) and predictive values for gmelina seed oil extraction process

The Normal plot of residuals was used to check whether the points will follow a straight line. The data points should be approximately linear; a non-linear pattern indicates abnormality in the error term which may be corrected by transformation. From Figure 3.2, it is seen that the points were closely distributed to the straight line of the plot, it confirms the good relationship between the experimental and predicted values of the response, so to say, the plot equally confirm that the selected model was adequate in predicting the response variables in the experimental values.

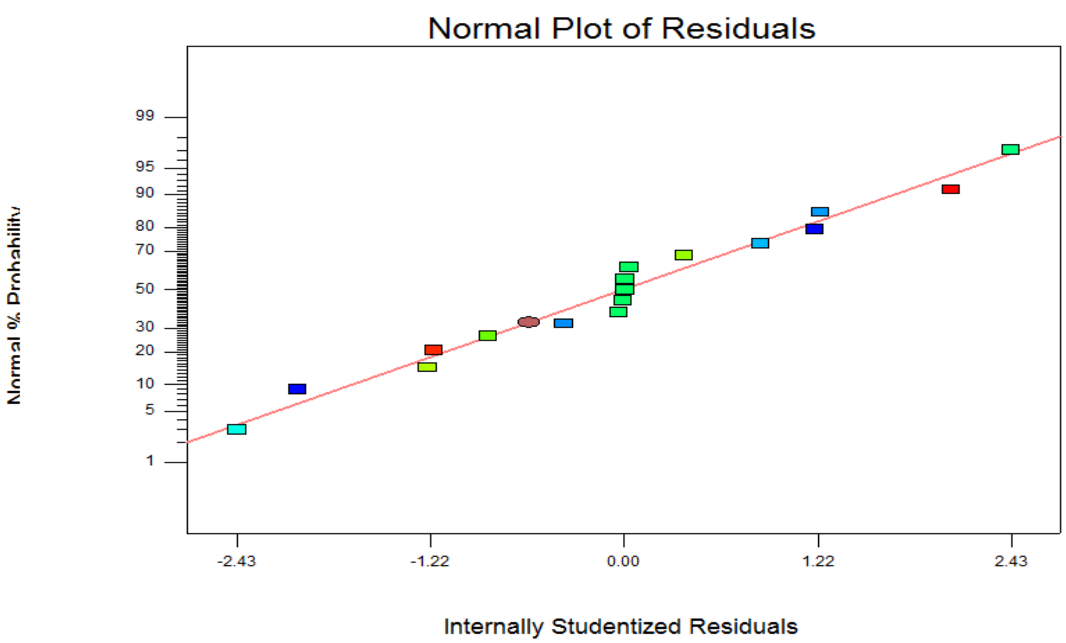

Figure 3.2: Normal plot of residuals

3.1.1. Analysis of variance for Gmelina seed extraction process

The statistical significance of the model was checked using F test analysis of variance (ANOVA) using the Design Expert 7.0 as shown in Table 3.2. The analysis shows that the regression model is significant with a computed F value of 261.39 and Probability > F value less than 0.0001. This suggests that there is only a 0.01% chance that a 'Model F-Value' this large could occur due to noise. The very low probability value (<0.0001) of the model is an indication that the model is significant. Each term in the model was also checked for its significance. Value of Prob< 0.05 is an indication that the model term has significant effect on the response while for P-value>0.1 is an indication of the insignificance of the model. From the ANOVA result, it can be seen that all the model terms were significant: seed particle (A), volume of solvent (B) and agitation time(C). Statistically, the influence of specific independent parameter on an output response is indicated by its F-value. From the ANOVA Table, the very high F-value of seed particle (A) is an indication that it had the greatest effect on Gmelina seed oil yield. Time and Volume of solvent did not have much influence on the yield as indicated by their low F-values. Uzoh and Onukwuli [16] also report seed particle as the most significant factor.

Table 3.2: Analysis of variance (ANOVA) table for the quadratic model for gmelina seed extraction process

Source	Sum of Squares	Df	Mean Square	F value	p-value (prob > F)
Model	1599.60	9	177.73	261.39	< 0.0001
A – Seed particle	1315.08	1	1315.08	1934.04	< 0.0001
B – Volume of solvent	141.62	1	141.62	208.28	< 0.0001
C – Time	93.09	1	93.09	136.91	< 0.0001
AB	8.85	1	8.85	13.02	0.0086
AC	9.55	1	9.55	14.04	0.0072
BC	5.41	1	5.41	7.95	0.0258
A^2	21.35	1	21.35	31.40	0.0008
B^2	5.10	1	5.10	7.50	0.0290
C^2	0.79	1	0.79	1.16	0.3170
Residual	4.76	7	0.68		
Lack of Fit	4.76	3	1.59	4806.49	< 0.0001
Pure error	1.320E-003	4	3.300E-004		
Cor Total	1604.36	16			

Table 3.3: Statistical information for the Gmelina seed extraction process

Standard deviation	Mean	Coefficient of Variance (%)	PRESS	R Squared	Adjusted R-Squared (correlation coefficient)	Predicted R-Squared	Adequate Precision
0.82	31.98	2.58	76.14	0.9970	0.9932	0.9525	53.851

From Table 3.3, the coefficient of variance (CV) shows the degree of precision by which parameters where compared. A lower value means higher reliability of the experiment hence the obtained CV value of 2.58% indicates a great reliability in the comparison. The reliability of the model was checked using the coefficient of determination (R^2). The R^2 value of 0.9970 means that the model suitably describes the actual response and adjusted R^2 value of 0.9932 further validates the model's adequacy. The 'Predicted R-Squared' of 0.9525 is in reasonable agreement with the "Adjusted R-Squared" of 0.9932. "Adequate Precision" measures the signal to noise ratio. A ratio greater than 4 is desirable. The design ratio of 53.851 indicates an adequate signal. This model can be used to navigate the design space.

3.1.2. 3-D Response Surface Plots for the Optimization Process

From statistical analysis one can tell the extent to which an independent parameter influences a response but cannot describe the influence (whether positive, negative or otherwise), hence the need for response surface plot. The 3-D response surface plots are graphical representations of the interactive effects of any two variables. The nature of the response surface curves shows the interaction between the variables. An elliptical shape of the curve indicates good interaction of the two variables and circular shape indicates no interaction between the variables. The data were generated using the RSM package by keeping two of the independent variables at a constant (central) level and varying the other two within their experimental ranges. The 3-D response surface plots are shown in Figure 3.3 to Figure 3.5 for the chosen model equation and it shows the relationship between the independent and the dependent variables.

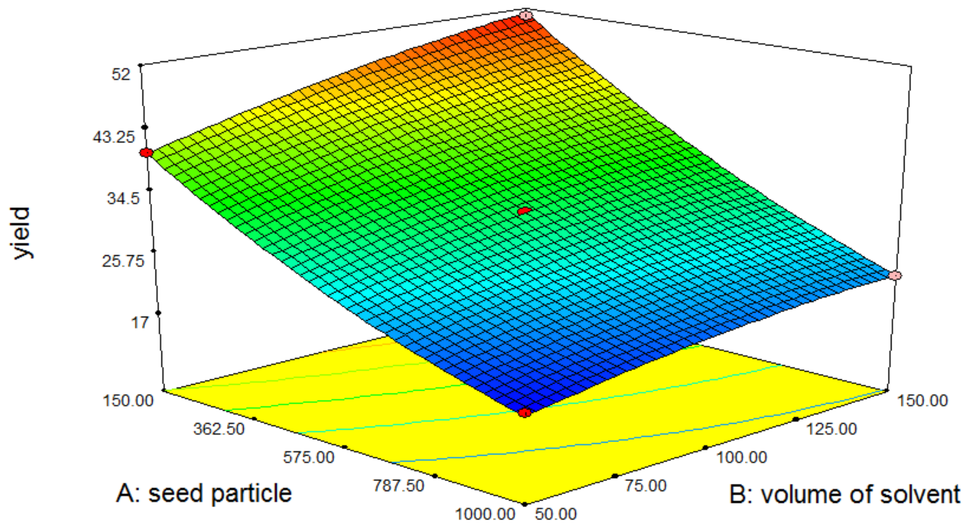

Figure 3.3: The effect of particle size (μm) and volume of solvent (ml) on the oil yield (%) at constant time of 35mins

Figure 3.3 shows a plot of the oil yield as a function of seed particle size and volume of solvent. It can be observed that the oil yield has a progressive increase with increase in solvent volume and decrease in particle size. The negative effect of seed particle size on oil yield could be attributed to the fact that smaller particles have larger amount of surface area coupled with increased number of ruptured cells resulting in a high oil concentration at the particle surface and low or little diffusion into the particles surface [17]. Sayyar et al, [2], while investigating the extraction of oil from Jatropha seed, suggested also that large particles have smaller amount of surface areas and are more resistant to intrusion of solvent and oil diffusion. Therefore, small amount of oil will be carried from inside the large particles to the surrounding solution.

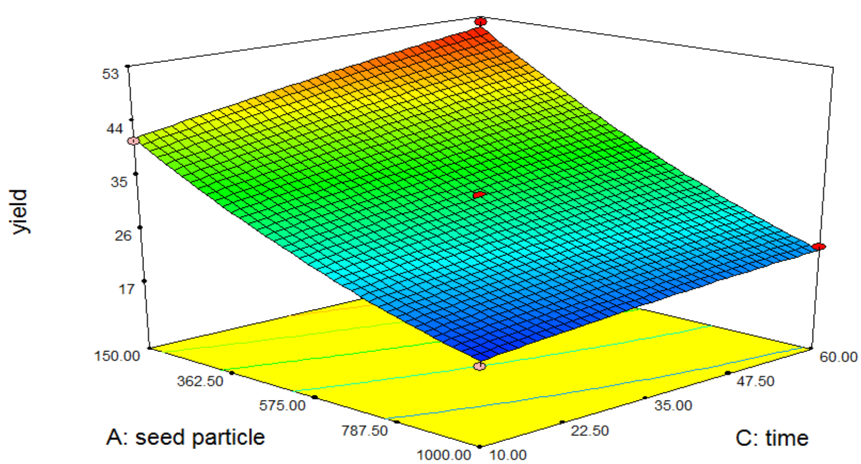

Figure 3.4: The effect of particle size (μm) and agitation time (mins) on the oil yield (%) at constant volume of 100ml

Figure 3.4 shows a plot of the oil yield as a function of seed particle size and agitation time. It can be observed that the oil yield has a progressive increase with increase in agitation time and decrease in particle size. Agitation time of the solvent is important because this increases the eddy diffusion and therefore the transfer of material from the surface of the particles to the bulk of the solution [18]. Further, agitation of suspensions of fine particles prevents sedimentation and more effective use is made of the interfacial surface [19].

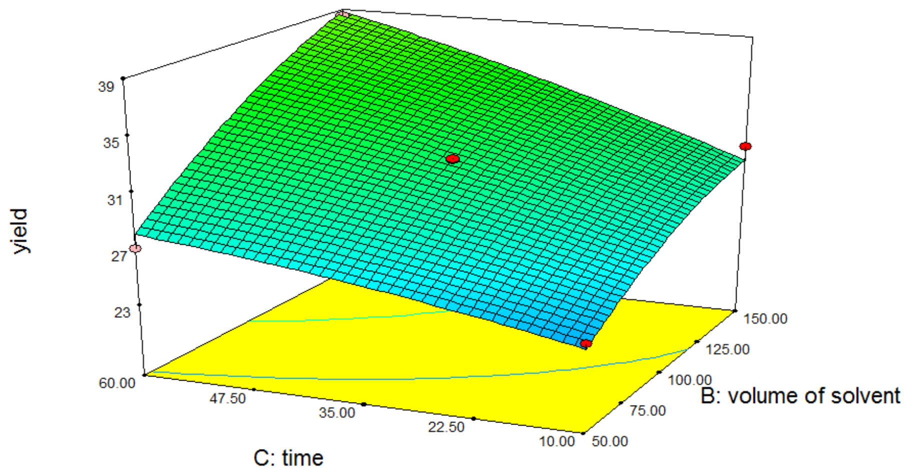

Figure 3.5: The effect of agitation time (mins) and volume of solvent (ml) on the oil yield (%) at constant particle size of 575µm

Figure 3.5 shows a plot of the oil yield as a function of agitation time and volume of solvent. It can be observed that the oil yield has a progressive increase with increase in solvent volume and increase in agitation time. The positive effect of volume of solvent on oil yield was as a result of increase in the concentration driving force as volume of solvent increases, it was also as a result of increased washing of the oil extracted, away from the particle surface by the solvent as a result of increased volume. This is in accordance with the report obtained by Meziane and Kadi that studied kinetics and thermodynamics of oil extraction from olive cake.

The perturbation plot for the model is shown in fig 3.6. Perturbation provides the outline views of the response. For the response surface designs, perturbation plot shows how the response changes as any of the parameters moves from the reference point, with all other factors held constant at the reference value.

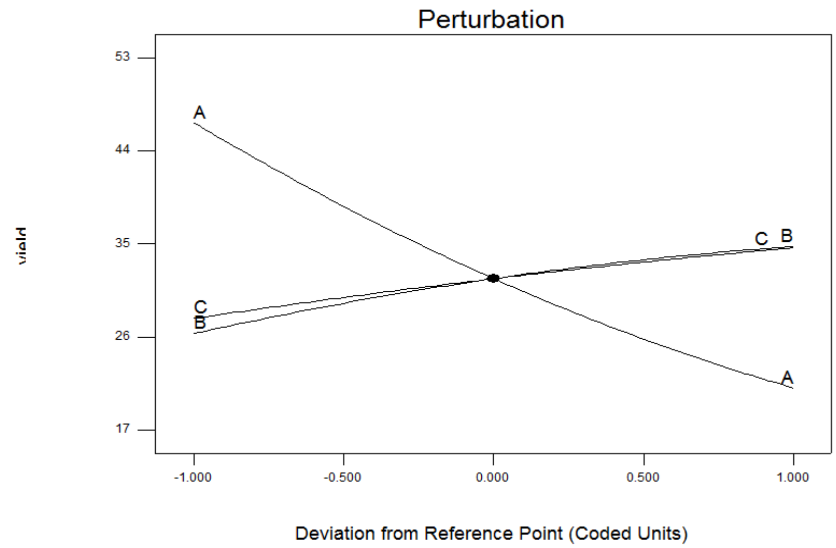

Figure 3.6: Perturbation plot for oil yield (%) as a function of Particle size (A), Volume of solvent (B) and Agitation time(C)

In Fig 3.5, it is shown that factor A (seed particle size) produces higher effect on the response as compared to factors B (Volume of solvent) and C (agitation time) because factor A shows higher slope than factors B and C. It has been shown that this higher effect is a negative one from the 3-D response surface plots in Figures 3.3 to 3.5. The perturbation plot also shows that factors B and C affects the oil yield almost equally; B is slightly higher than C.

The maximum response predicted from the model was an oil yield of 57%. The final optimized parameters influencing the yield of oil obtained were seed particle size of 150µm, solvent volume of 150ml and agitation time of 60mins. The maximum predicted oil yield of 57% obtained was almost close to the experimental value of 52.09%. This proves the validity of the model which indicates that there is excellent correlation between experimental and predicted values.

The sensory analysis of the gmelina seed oil indicated physical state of the oil to be liquid and amber yellow at room temperature. The oil content of gmelina arborea seed was found to be 52.09%wt. The oil content is significant and compares favourably with seed oil of other plants such as Hevea brasiliensis (51%wt), Hematostaphis berter (54.5%wt), Jatropha curcas (30-50%wt), Sapindus mukorossi (51%wt), Mellia azadirachta (33-45%wt), and 55-65% wt for Simarouba glauca. On the basis of the oil content, gmelina arborea seed would be highly suitable economically for industrial applications, as any oil bearing seed that can produce up to 30% oil are regarded as suitable [6].

4. Conclusion

From the experimental study conducted, the following conclusions were drawn;

I. Extraction of oil from gmelina seed is influenced by seed particle size, volume of solvent and agitation time.

II. The use of statistical tools like Box-Behnken design of experiment helps in the optimization of process parameters.

III. The percentage oil yield obtained from the extraction of oil from gmelina seed is related to the seed particle size, volume of solvent and agitation time by a quadratic regression model equation.

IV. The combination of the optimum process conditions were:
   - seed particle size of 150µm
   - solvent volume of 150ml and
   - agitation time of 60mins

   To give a 52.09% oil yield.

V. On the basis of the oil content, gmelina arborea seed would be highly suitable economically for industrial applications.

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