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ORIGINAL RESEARCH PAPER

OPTIMIZATION OF SESAME SEEDS OIL EXTRACTION OPERATING CONDITIONS USING THE RESPONSE SURFACE DESIGN METHODOLOGY

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Abstract: This paper applies Response Surface Design (RSD) to model the experimental data obtained from the extraction of sesame seeds oil using n-hexane, chloroform and acetone as solvents under different operating conditions. The results obtained revealed that n-hexane outperformed the extraction obtained using chloroform and acetone.
The developed model predicted that n-hexane with a rotational speed of 547 rpm and a contact time between the solvent and seeds of 19.46 hours with solvent: seeds ratio of 4.93, yields the optimum oil extracted of 37.03 %, outperforming chloroform and acetone models that gave prediction for 4.75 and 4.21 respectively.
While the maximum predictions yield for chloroform is 6.73 %, under the operating conditions of 602 rpm, and 24 hours contact time, with a ratio of solvent: seeds of 1.74. On the other hand the acetone maximum prediction is only 4.37 %, with operational conditions of 467 rpm, and 6.00 hours contact time, with a ratio of solvent: seeds of 1.
It is has been found that the maximum oil extraction yield obtained from the chloroform (6.73 %) and Acetone (4.37 %) is much lower than that predicted by n-hexane 37.03 %.

Keywords: *ANOVA statistical test, operating conditions, process optimization, sesame seeds oil extraction, response surface design*

INTRODUCTION

The use of solvents in oil extraction has been a subject that gained the attention of researchers' worldwide for a very long time. The need for a clean, high-efficiency process that provides higher extraction yield was the motivation for most of the work carried in this area.

Sesame seeds have been used as a healthy food and for disease prevention in many countries for centuries. The literature reports indicate many health benefits associated with the consumption of sesame seeds; for example, they significantly increase plasma γ-tocopherol and enhance vitamin E activity, which are believed to prevent human aging-related diseases such as cancer and heart disease. Studies have also shown that including sesame in the diet can improve antioxidant capacity [1].

Sesame seeds have higher oil content (around 42-50 %) than most of the known oil seeds, see Table 1.

Table 1 .*Oil content of selected seeds/fruits*

	Product name	Oil content in [%]	Water content in [%]
1	Mustard seeds	15.0 - 35.0	5.0 - 9.0
2	Sesame seed	42.0 - 50.0	5.0 - 13.0
3	Shea nuts	46.0 - 48.0	5.0 - 8.0
4	Soybeans	13.0 - 24.0	8.0 - 10.0
5	Sunflower seeds	19.0 - 56.0	7.0
6	Olives, black	15.0 - 31.0	52.0
7	Olives, kernels	12.0 - 15.0	8.0

Sesame oil is generally regarded as high-priced and high-quality oil. It is one of the most stable edible oil despite its high degree of unsaturation [2]. Sesame oil is rich in monounsaturated and polyunsaturated fatty acids [1]. The most abundant fatty acids in sesame oil are oleic, linoleic, palmitic, and stearic acids, which together comprised about 96 % of the total fatty acids.

Extraction of sesame oil has developed significantly over the years; mechanical methods are an early means of separation by physical pressure to 'squeeze the oil out'. The most energy efficient, practical embodiment of that method is the modern screw press. This is a conveyor screw with a slotted cage surrounding it and a screw with diminishing space for the solid material as the material proceeds from pitch to pitch of the screw. Eventually, as the free space is progressively restricted, the oil is squeezed out of the solids and through the slots. More than half of the oil is easily removed in this way, but at least 7 - 18 % residual oil is left in the solids, the process uses considerable horsepower, there is considerable wear and maintenance, and it takes many machines for high capacity.

In a nutshell such techniques are no longer prevailing currently due to the higher cake oil contents. Solvent extraction, providing higher yields 98 - 99 %, is nowadays the dominant technique applied in most extraction processes. Solvent extraction became the commonly used commercial technique to recover oil from oilseeds, presently n-hexane is the preferred solvent throughout the world due to its extraction efficiency and ease of availability [3]. In comparison, solvent extraction with hexane will leave about 0.5 % of residual oil. In addition, it uses less horsepower, and requires less maintenance; hence it is more efficient and reliable.

Over the years, researchers investigated different solvents for sesame seeds oil extraction, Durma et al.[4] investigated the impact of roasting seeds on the oil yield and has concluded that antioxidant capacity of the roasted seeds and oxidative stability of the extracted oil could be greater than that of the unroasted counterpart. Elkhaleefa et al. [5] have studied the operating condition of sesame seeds oil extraction in more comprehensive details as they examined the effect of sesame seeds particle sizes, the ratio of solvent to seeds mass, contact time, stirring effect, roasting impact and extraction temperatures.
The application of experimental design using response surface methodology (RSM) has been widely used for optimization of experimental parameters. RSM is a collection of mathematical and statistical techniques widely applied in the food industry to evaluate the relationship between the predicted values of independent and dependent variables. RSM is not only time and reagent saving, but it also provides information about the interactions of parameters [6, 7]. Experimental design for process optimization via response surface methodology has been studied by many researchers [8 – 12].
The objective of this study was to analyze the effects of: seeds/solvent ratio (1 to 5), rotation speed (0, 150, 300 and 700 rpm) and contact time (6, 12, 24 hours) on the oil extraction yields. An effective technique to study and evaluate the effect of some of the above variables on the extraction yield and extract composition, which allows identification of the main effects with limited experimental works, is response surface analysis of results of carefully planned experiments, or so-called response surface design (RSD) of experimental data. Data obtained by Elkhaleefa et al. [5] are used as operational conditions parameters.

MATERIALS AND METHODS

The experimental design is based on a face centered (FCD), three-variable (operation conditions in this case), three-level 3^3 fractional factorial design with star points and triplicates at the center [13]. Experiments were carried out in a randomized order to minimize the effect of unexpected variability in the observed response due to extraneous factors. A second-order (quadratic) polynomial equation was used to express each response variable Y as a function of the independent variables X in the general form:

$$Y = \sum_{i=1}^{3} \sum_{j=1}^{3} A_{ij}^{y} X_i X_j \tag{1}$$

where: X_0 (dummy variable) equals 1; A_{01}, A_{02}, and A_{03} are linear coefficients; A_{12}, A_{13}, and A_{23} are cross-product coefficients; A_{11}, A_{22} and A_{33} are quadratic coefficients, and the super index *y* defines the dependent variables (*e. g.* concentration of A, B, and C).
The best fitting of the model was evaluated by determining the coefficient of determination R^2 and by the analysis of the variance (ANOVA). The coefficients of the response surface equation were estimated by using Design-Expert Software. The statistical significance was based on the total error criteria with a confidence level of 95 %.

The mathematical model

Experimental data from literature [5] were modelled using the Design-Expert Software. Response Surface Design (RSD) was used to evaluate and find the optimum model, the three variables; seeds/solvent ratio**,** contact time (hour) and stirring speed (rpm) were coded to ensure dimensionless of the variables by applying the general equation:

$$The\ dimensionless\ variables = \frac{variables\ value - mean\ variable}{range} between\ variables\ (int\,ervals)$$

Thus obtain: Seeds/solvent ratio X_1 = (A-3)
Contact time X_2 = (B-12)/6
Stirring speed X_3 = (C-150)/150

as. The design model can be written in a function of each independent variable (X_1, X_2 and X_3) as follows:

$$Y = \beta_o + \sum_{i=1}^{3} \beta_i X_i + \sum_{i=1}^{3} \beta_{ii} X_i^2 + \sum \sum_{i<j=1}^{3} \beta_{ii} X_i X_j \quad (2)$$

where: β_0, β_i, β_{ii}, and β_{ij} are the regression coefficients for intercept, and the notations X_1 = A, X_2 = B and X_3 = C are the dimensionless independent variables as presented in Table 2.

***Table 2.** Experimental design variables used for optimizing sesame seeds oil yield.*

Independent variables X_n	**Symbol**	**Coded levels**		
		-1	**0**	**1**
Seeds to solvent ratio in [%] *(r)*	A	1	3	5
Contact time in [h] *(t)*	B	6	12	24
Stirring speed in [rpm] *(S)*	C	0	350	750

RESULTS AND DISCUSSION

In this paper the Response Surface Design (RSD) is used as an effective technique to study, model and compare between the effects of various variables. These variables are the operating conditions for oil extraction using three different solvents; n-hexane, chloroform and acetone, the variables are: seeds/solvent ratio (1 to 5), rotational speed (0, 150, 300 and 700 rpm) and contact time (6, 12, 24 hours) and their effect on the overall oil extraction yield.

n-Hexane solvent extraction model

The operating conditions of sesame seeds oil extraction by hexane solvent have been studied by Elkhaleefa et al. [5] in a more comprehensive details as they examined the effect of sesame seeds particle sizes, the ratio of solvent to seeds mass, contact time, stirring effect, roasting impact and extraction temperatures. The experimental data consist of 60 runs. This paper models the experimental data using the above regression for the three variables.

Pre-model results for n-hexane

When applying the developed model using the previously defined data, the following Pre-model has been obtained:

$$Oil\ yield\% = 33.66 + 4.26 \times A + 0.43 \times B + 0.25 \times C - 0.26 \times A \times B - 0.098 \times A \times C - 0.013 \times B \times C - 0.93 \times A^2 - 0.060 \times B^2 - 0.32 \times C^2 \quad (3)$$

The model has been verified using ANOVA statistical test where Table 3 was obtained.

***Table 3**. ANOVA Test for response surface quadratic model analysis of variance*

	F-Value	P-Value
Model	1263.63	< 0.0001
A - seeds/solvent	9975.61	< 0.0001
B - contact time	136.80	< 0.0001
C - stirring speed	39.83	< 0.0001
AB	27.63	< 0.0001
AC	3.13	0.0830
BC	0.075	0.7857
A^2	178.63	< 0.0001
B^2	0.72	0.3997
C^2	21.48	< 0.0001

From Table 3 it has been found that the calculated F-value of 1263.63 implies that the model is significant (as the largest value is desirabled). However, the P-value should be less than 0.0500 to indicate that model terms are significant. In this case A, B, C, AB, A^2, C^2 are considered significant model terms. Values greater than 0.1000 indicate that the model terms are not significant, the model reduction will improve this model.

***Table 4.** R^2 Coefficient values*

Std. Dev.	0.23	R^2	0.9956
Mean	32.87	Adj R^2	0.9948
C.V. %	0.68	Pred R^2	0.9938
PRESS	3.59	Adeq Precision	109.090

The "Pred R^2" of 0.9938 is in reasonable agreement with the "Adj R^2" of 0.9948 as presented in Table 4. "Adeq Precision" measures the signal to noise ratio. A ratio greater than 4 is desirable. The model ratio of 109.090 indicates an adequate signal and this model can be used to navigate the design space. The determination coefficient (R^2) and the adjusted determination coefficient (Adj. R^2) were 0.9956 and 0.9948, respectively, which implied that the model could explain 99.56 % variability of the response variable.

The experimental and predicted values were compared in order to determine the validity of the model, the values of "Prob > F" less than 0.0500 indicate model terms are significant. In this case A, B, C, AB, A^2, C^2 are significant model terms. This model can be visualised in 3D graphical shape as in Figure 1A and 1B.

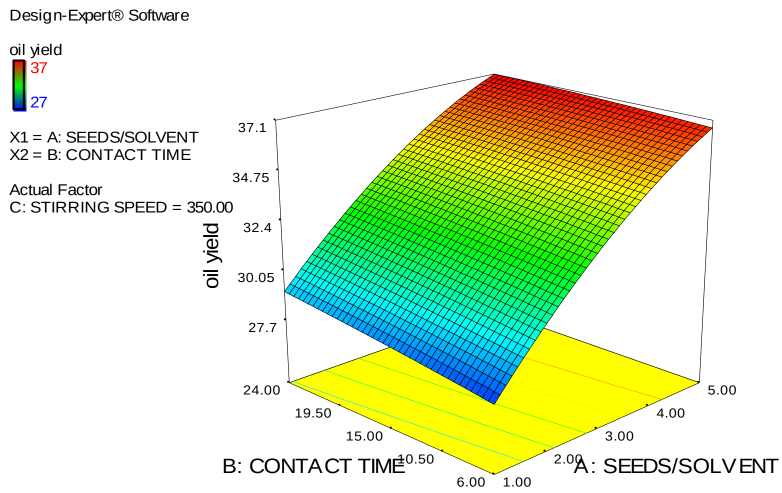

Figure 1A. *3D Prediction of oil extraction yield using n-hexane at different seeds/solvent ratio vs. contact time at constant stirring speed of 350 rpm*

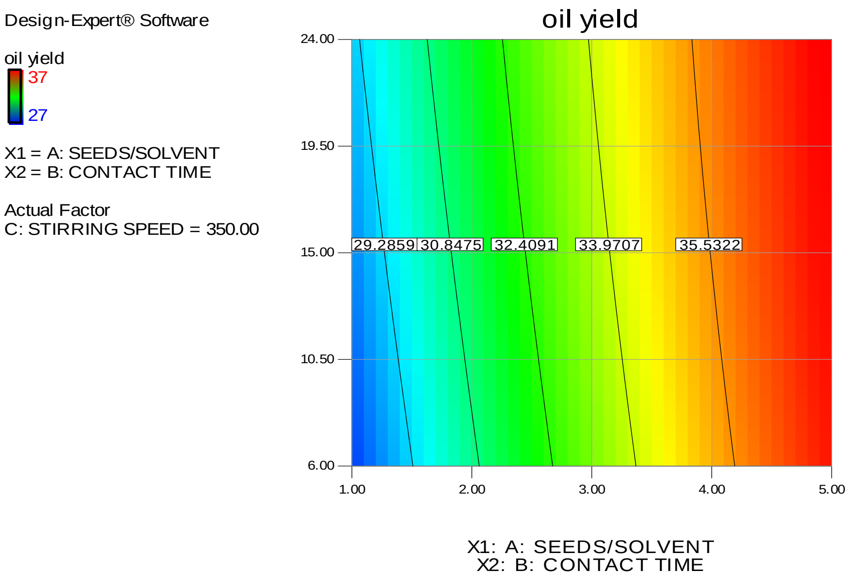

Figure 1B. *Contour prediction of oil extraction yield using n-hexane at different seeds/solvent ratio vs. contact time at constant stirring speed of 350 rpm*

The experimental data and predicted values were compared in order to determine the validity of the model and have showed good agreement as presented in Figure 2.

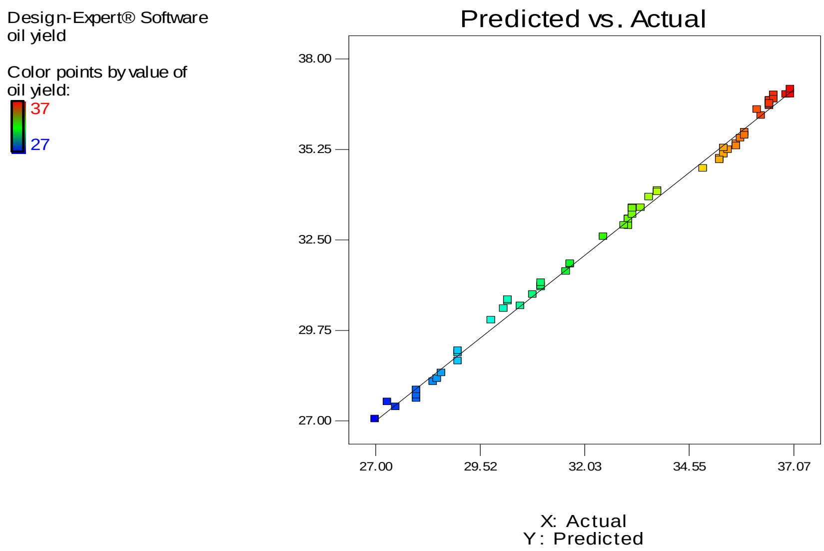

Figure 2. *Oil extraction yield prediction vs. oil extraction yield experimental data using n-hexane*

Model reduction and modification for n-hexane

Table 5 below summarizes the statistical parameters for the three Response Surface Design (RSD), it shows that the parameters are estimated adequately because of (p-value < 0.05). High coefficients of determination (R^2 > 0.90) for all Response Surface Desing models, and high signal-to-noise (> 4) for all responses, indicate that the models can be used to navigate the design space. This statistical indicators are complemented in Figure 2 by scatter plots demonstrating the correlation between predicted and experimental responses.

Table 5. *ANOVA for response surface reduced quadratic model analysis of variance table [partial sum of squares - type iii]*

	F-Value	P-Value
Total Model Variables	1862.24	< 0.0001
A - seeds/solvent	10442.82	< 0.0001
B - contact time	144.81	< 0.0001
C - stirring speed	40.28	< 0.0001
AB	27.15	< 0.0001
A^2	175.57	< 0.0001
C^2	21.11	< 0.0001

Model verification

The experimental and predicted values were compared in order to determine the validity of the model in the (Figure 2). The researchers [14] found that solvent concentration plays a critical role in the extraction of soluble solids; Also, Kwon et al. [15] reported that solvent concentration was the most important factor contributing to the extraction components using RSM. The plot of predicted versus experimental values for the concentration shows more correlations, due to the quadratic equation.

The F-value model of 1862.24 implies that the model is significant. There is only a 0.01% chance that this large "F-Value" could occur due to noise. Moreover the P-values of less than 0.0500 indicate that the model terms are significant. In this case A, B, C, AB, A^2 and C^2 are the significant model terms. Values greater than 0.1000 indicate that the model terms are not significant, concluding that this is the best model terms that can be achieved using ANOVA test since all terms are significant.
Table 6, presents the "Pred R-Squared" of 0.9941 which is in reasonable agreement with the "Adj R2" of 0.9947. On the other hand, "Adeq Precision" measures the signal to noise ratio where ratio value greater than 4 is desirable. This model ratio of 128.012 indicated an adequate signal, which it higher than the previous pre-model.

Table 6. *Modified model R^2 coefficient values*

Std. Dev.	0.23	R^2	0.9953
Mean	32.87	Adj R^2	0.9947
C.V. %	0.69	Pred R^2	0.9941
PRESS	3.43	Adeq Precision	128.012

From what has been found, the final equation model for the n-hexan in terms of coded factorsis presented in the following format:

$$Oil\,yield\% = 33.66 + 4.26 \times A + 0.43 \times B + 0.25 \times C - 0.26 \times A \times B - 0.098 \times A \times C - 0.013 \times B \times C - 0.93 \times A^2 - 0.060 \times B^2 - 0.32 \times C^2 \quad (4)$$

The final equation in terms of actual factors is:

$$Oil\,yield\% = 23.18267 + 3.74661 \times r + 0.090446 \times t + 2.55737E - 003 \times S - 0.014435 \times r \times t - 0.23214 \times r^2 - 2.61689E - 6 * S^2 \quad (5)$$

Optimization of the final model resulted in (Figures 3A and 3B), where it can be revealed that the optimum value for the maximum oil yield is 37.0458, the seeds to solvent ratio of 4.93 and with contact time of 19.46 h and strring speed of 547 rpm as summarised in Table 7.

Table 7. *n-Hexane optimised extraction prarmeters*

Solvent	**Contact time *t* [h]**	**Stirring speed *S* [rpm]**	**Seeds: Solvant ratio *r***	**Oil yield [%]**
n-Hexane	19.46	547	4.93	37.04

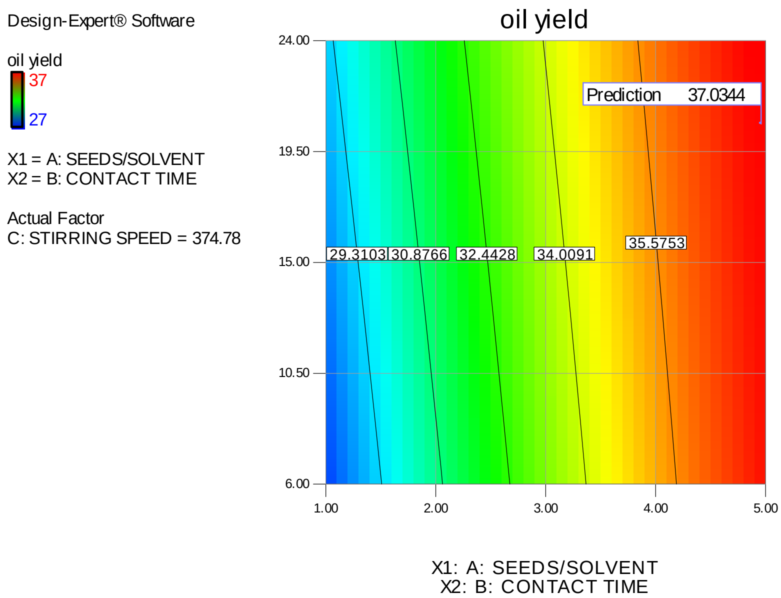

Figure 3A. *Contour optimum predicted extracted oil value with respect to seeds: n-hexane solvant ratio vs. contact time*

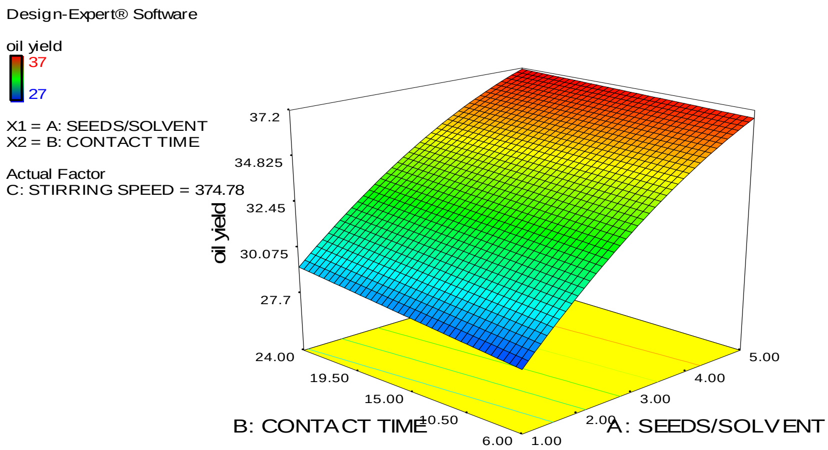

Figure 3B. *3D optimum predicted extracted oil value with respect to seeds: n-hexane solvant ratio vs. contact time*

Chloroform extraction model

Similar to the work done on the n-hexane the results for the final chloroform extraction model is presented as:

$$Oil\ yield\% = 5.85537 + 0.057500 \times (r-3) + 0.027976 \times \frac{t-12}{6} + 1.58605E - 003 \times \frac{S-150}{150} - 6.01190E - 003 \times (r-3) \times \frac{t-12}{6} - 1.63559E - 006 \times \left[\frac{S-150}{150}\right]2 \quad (6)$$

The above equation predict the oil yield in term of (as function of) all independent variables the response surface as can be seen from the 3D Figure 3B below is semi-flat.

Optimization of chloroform model

The maximum value for the oil yield is 6.72259 at the ratio of 1.59, contact time of 23.97 and stirring speed of 310.12 as summarised in Table 8 and (Figure 4A and 4B).

Table 8. *Chloroform optimised extraction prarmeters*

Solvent	Contact time t [h]	Stirring speed S [rpm]	Seeds: Solvant ratio r	Oil yeild [%]
Chloroform	23.97	310.12	1.59	6.722

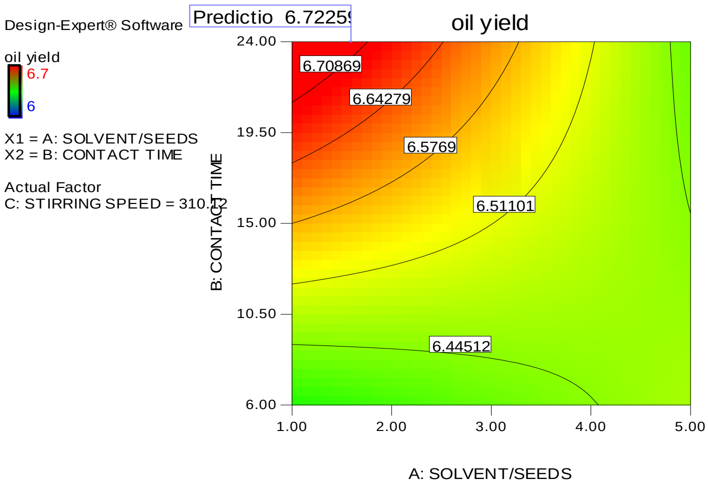

Figure 4A. *Contour optimum oil extracted yield with respect to constant contact time and chloroform solvent to seeds ratio*

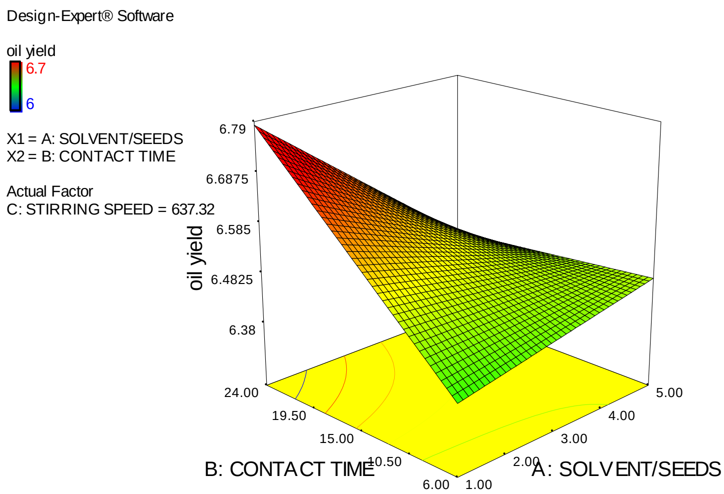

Figure 4B. *3D optimum oil extracted yield with respect to constant contact time and chloroform solvent to seeds ratio at different stirring speeds*

Acetone extraction model

Similarly when applying the same technique on acetone extraction experimental data, the following final equation in terms of coded factors is obtained.

$$Oil\ yield = +4.17 - 0.018 \times A - 0.023 \times B + 0.065 \times C + 0.051 \times A^2 + 0.048 \times B^2 - 0.097 \times C^2 \quad (7)$$

Final equation in terms of actual factors is:

$$Oil\ yield\% = 4.31700 - 0.085536 \times r - 0.020208 \times t + 7.41100E - 004 \times S + 0.012798 \times r^2 + 5.90278E - 004 \times t^2 - 7.91606E - 007S^2 \quad (8)$$

The 3D model can be visualised at this point as in (Figure 5).

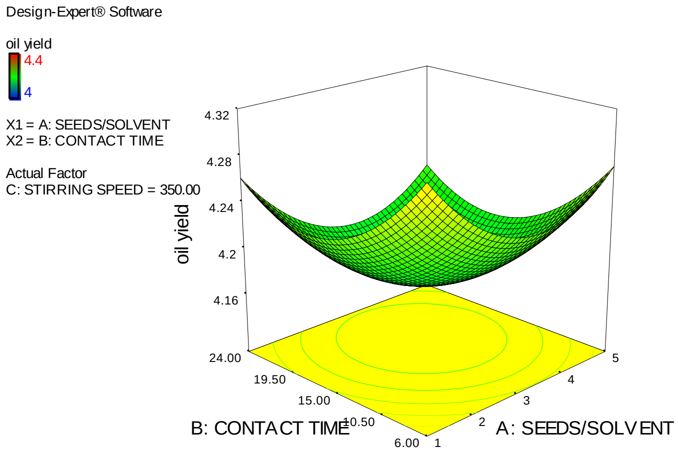

Figure 5. *3D optimum oil extracted yield with respect to constant contact time and acetone solvent to seeds ratio at different stirring speeds*

The best model achived a maximum value for the oil yield of 4.31772 the ratio 1.00 the contact time is 6 h the strring speed is 468.64, Table 9 and Figure 6.

Table 9. *Acetone optimised extraction prarmeters*

Solvent	Contact time *t* [h]	Stirring speed *S* [rpm]	Seeds: solvant ratio *r*	Oil yeild [%]
Acetone	6	468.64	1.00	4.31772

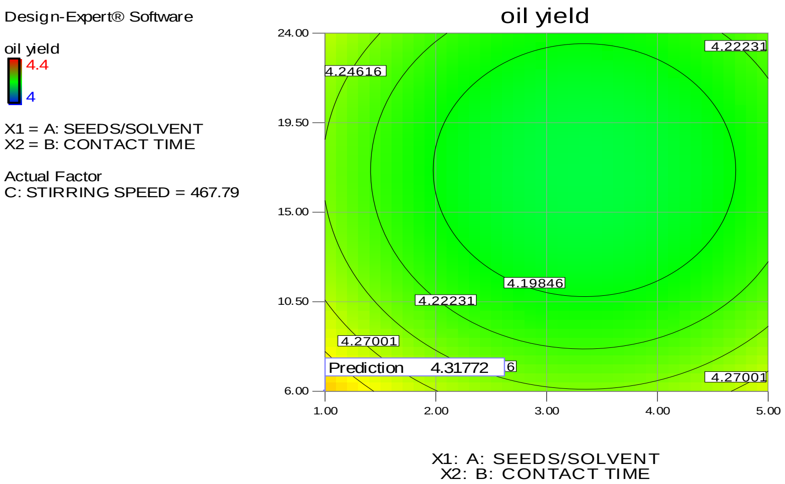

Figure 6. *Contour optimum oil extracted yield with respect to constant contact time and acetone solvent to seeds ratio at different stirring speeds*

CONCLUSIONS

This study showed that RSM is a powerful modelling technique capable of describing and predicting solvent extraction operational process parameters. Contours and surface plots in RSM estimated the effects of three independent variables- contact time, stirring speed and seeds to solvent ratio- efficiently.
Table 10 summarizes the optimised predictions for the model using the solvents: n-hexane, chloroform and acetone.

Table 10. *Prediction of the extraction for the solvents*

Solvent	**Contact time [h]**	**Stirring speed [rpm]**	**Seeds: Solvent ratio *r***	**Oil yield [%]**
n-Hexane	19.46	547	4.93	37.0063
Chloroform	23.94	602	1.74	6.73632
Acetone	6.00	467	1.00	4.31772

In a nutshell, hexane has shown significant extraction yields in comparison with other solvents. The developed model was able of predicting extraction yields that are in good agreement with experimental data.

SYMBOLS

A_{ij}	linear coefficient
X_{ij}	independent variables
Y	oil yield [%]
β	independent coefficient
A	seeds/solvent ratio [%]
X_1	seeds/solvent, dimensionless
t	contact time, [h]
X_2	contact time, dimensionless
S	stirring speed, [rpm]
X_3	stirring speed, dimensionless
R^2	determination coefficient

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