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SOLVENT EXTRACTION OF OIL FROM SOURSOP OILSEEDS & ITS QUALITY CHARACTERIZATION

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ABSTRACT

This study focused on optimization of oil extraction from Soursop oilseeds using Box-Behnken design an allied of Response Surface Methodology (RSM), it also examine the physicochemical properties and fatty acid profile of the oil. Based on the design, 17 experimental runs were conducted to investigate the effects of variables and their reciprocal interactions on the oil yield. A quadratic polynomial and the ANOVA test showed the model to be remarkably significant ($p< 0.05$). A statistical model predicted the highest oil yield to be 34.6074% (w/w) at the optimal condition of X_1 = 38.10 min, X_2 = 30.00g and X_3 = 100 ml. The experiment was validated as 33.593% (w/w) of oil. The fatty acid profile of the oil revealed the oil to be highly unsaturated (73.42%). The physicochemical properties of extracted oil revealed that the oil is edible and could serve as feedstock for many industrial applications. This may provide useful information regarding the development of economic and efficient processes using solvent extraction method.

Keywords: Optimization, Soursop, Box-behnken, Fatty acid, Physicochemical properties.

1. INTRODUCTION

The massive disposal of biomass wastes (skin, pulp, seeds etc.) that always occurred in most fruit processing units and underutilization of products has become noticeable in recent years (Federici *et al.*, 2009). Meanwhile, recent studies revealed that bio-recovery of valuable byproducts like ethanol, enzymes, pharmaceuticals and essential oils from agro-biomass wastes like mango, beniseed, sorrel, pineapple and papaya have been carried out (Arumugam and Manikandan, 2011;

Betiku and Adepoju, 2012; Kumar *et al.*, 2012; Adepoju and Abiodun, 2013). Soursop has emerged as a potential candidate for production of essential oil.

Soursop is a fruit of *Annona muricata* a broadleaf, flowering, evergreen tree native to Mexico, Cuba, Central America, the Caribbean, and northern South America, primarily Colombia, Brazil, Peru, Ecuador, Venezuela as well as in some parts of Africa, Southeast Asia and the Pacific. It is in the same genus as the chirimoya and the same family as the papaya. The flesh of the fruit consists of an edible, white pulp, some fiber, and a core of indigestible, black seeds (Arumugam and Manikandan, 2011). Soursop seeds constitute 20% - 25% mass of the fruit and have potential to produced 33.87% oil with nutritional and functional properties highly similar to those of edible oils (Okoro, 2013). Different extraction methods (aqueous enzymatic extraction, extrusion expelling process and solvent extraction) have been examined for the recovery of oils from oilseeds (Lee *et al.*, 2011; Adepoju *et al.*, 2013; Okoro, 2013). Previous researchers applied solvent extraction technique for the extraction of oils from different plant oilseeds due to it cost effectiveness, simplicity, remarkable reduction in solvent volume and short time needed for the extraction (Tan *et al.*, 2009; Betiku and Adepoju, 2012; Bimakr *et al.*, 2012; Adepoju *et al.*, 2013). Moreover, the optimization of extraction conditions (extraction time, sample weight, solvent volume and particle size) in solvent extraction methods have been studied in order to obtain extra yield and better quality index of the product under the optimum extraction conditions (Mani *et al.*, 2007; Li *et al.*, 2009; Adepoju *et al.*, 2013).

RSM a good optimizer is a collection of statistical techniques for designing experiments, building models, evaluating the effects of factors and searching for the optimum conditions (Kalil *et al.*, 2000). Based on RSM, the observed responses to design of experiments (DOEs) are fitted to a quadratic function. The models assume that a second-order polynomial relation can reasonably approximate many of the extraction process dynamics. The main advantage of RSM is the capability to minized the number of experimental runs needed to give adequate evidence for statistically acceptable results. Physicochemical properties of oils such as colour, refractive index, acid value, saponification value, iodine value, higher heating value, etc. as well as the fatty acid profile are important quality characteristics used in determining their potential uses. In the current study, solvent extraction of oil from Soursop seeds was carried out, to optimize the extraction conditions for the process, RSM was employed to determine the effects of three-level-three factors and their reciprocal interactions on the oil extracted. In addition, the quality of oil extracted was evaluated by carrying out physicochemical properties and fatty acid analysis with a view to determining its potential applications.

2. MATERIALS AND METHODS

2.1. Materials

Soursop seeds were collected from Omu-Aran market, Kwara State, Nigeria. The seeds were washed to remove particles and dried at room temperature for. The black hard epicarp seeds were cracked manually and the fleshy part was removed. The fleshy part was sun dried for 3 days until the constant weight was obtained. The cleaned seeds were milled to powder by grinding with grinding machine. All chemicals and reagents used for this extraction work were of analytical grades and needs no further purification.

2.2. Methods

2.2.1. Experimental Design

In order to optimize oil extraction from Soursop oilseeds, Box-Behnken experimental design, an allied of RSM was employed. Three-level-three-factors design which generated 17experimental runs was applied. Selected extraction variables were samples were sample weight (g): X_1, extraction time (min): X_2, and solvent volume (ml): X_3. Table 1 showed the coded independent variables levels, while Table 2 showed the 17 experimental runs generated with the oil yields, the predicted oil yields and the residual values. The randomization of experiments was used to minimize the effect of unexplained variability in the observed response.

2.2.2. Description of Oil Extraction

A 250-ml Soxhlet extractor apparatus and n-hexane as solvent were used for this work. The apparatus was charged with a known weight of the oilseeds powder in a muslin cloth placed in a thimble of the apparatus. A reactor containing known volume of n-hexane was stationed to the end of the apparatus and a condenser

Table-1. Factors and their levels for Box-Behnken design.

Factor	Symbol	Coded factor levels		
		-1	0	+1
Extraction time (min)	X_1	30	40	50
Sample weight (g)	X_2	30	40	50
Solvent volume (ml)	X_3	100	125	150

Table-2. Experimental design matrix by Box-Behnken for three-level-three-factors response surface study

Std. run	X_1	X_2	X_3	Experimental yield% (w/w)	Predicted yield % (w/w)	Residual values
1	-1	-1	0	24.00	23.85	0.15
2	1	-1	0	27.20	27.19	0.013
3	-1	1	0	30.00	30.01	-0.012
4	1	1	0	31.16	31.31	-0.15
5	-1	0	-1	26.00	25.75	0.25
6	1	0	-1	36.30	35.92	0.38
7	-1	0	1	35.80	36.18	-0.38
8	1	0	1	30.40	30.64	-0.24
9	0	-1	-1	34.50	34.89	-0.39
10	0	1	-1	40.20	40.43	-0.23
11	0	-1	1	38.10	37.87	0.23
12	0	1	1	43.00	42.61	0.39
13	0	0	0	34.50	34.22	0.28
14	0	0	0	34.00	34.22	-0.22
15	0	0	0	34.50	34.22	0.28
16	0	0	0	34.10	34.22	-0.12
17	0	0	0	34.00	34.22	-0.22

was tightly fixed at the bottom end of the extractor. The whole set up was heated up in a heating mantle at a constant temperature of 70 ^oC and the seedoils was collected in the flask. The excess solvent in the seedoils was recycled by heating at the same temperature. The quantity of the seedoils yield was determined gravimetrically as the ratio of the weight of the extracted seedoils to the weight of the Soursop seed powder sample used (Eq.1). The obtained oil was kept properly for further processing.

$$Oil\ yield\ \%\ (w/w) = \frac{Weight of seedoils\ produced}{Weight\ of\ soursop\ oilseeds\ powder\ used} X\ 100 \qquad (1)$$

2.2.3. Physicochemical Analysis of the Soursop Seedoils

The physicochemical properties of the crude seedoils namely, refractive index, moisture content, density, viscosity, acid value, saponification value, peroxide value, specific gravity and % FFA (oleic) were determined using (AOAC, 1990) methods.

2.2.4. Fatty Acid Compositions Analysis of Soursop Seedoils

Fatty acid composition of the crude oil was determined using gas chromatography (HP6890 powered with HP ChemStation Rev. A 09.01 [1206] Software). Oil sample (50 mg) was esterified for five minutes at 95 ^oC with 3.4 ml of the 0.5 M KOH in dry methanol. The mixture was neutralized using 0.7 M HCl and 3 ml of 14% boron triflouride in methanol was then added, the mixture was heated for 5 min at 90 ^oC to achieve complete methylation process. The fatty acids were extracted in triplicate from the mixture with redistilled n-hexane. The content was concentrated to 1 µl for gas chromatography analysis which was injected into the injection port of GC.

2.3. Numerical Data Analysis

The results obtained from 17 experimental runs were statistically analyzed using RSM, so as to fit the quadratic polynomial equation generated by the Design-Expert software version 8.0.3.1 (Stat-Ease Inc., Minneapolis, USA). To correlate the response variable (Y) to the independent variables (X_1, X_2 and X_3), multiple regressions was used to fit the coefficient of the polynomial model of the response. The quality of the fit of the model was evaluated using test of significance and analysis of variance (ANOVA). The fitted polynomial equation is as follows:

$$Y = b_0 + \sum_{i=1}^{k} b_i X_i + \sum_{i=1}^{k} b_{ii} X_i^2 + \sum_{i<j}^{k} b_{ij} X_i X_j + e \qquad (2)$$

Y is response factor (Oil yield)

b_o = intercept value. (i= 1, 2, ..…… k)

b_i = first order model coefficient

b_{ij} = interaction effect,

b_{ii} = qaudratic coefficients of X_i

e = random error.

3. THE DISCUSSION OF RESULTS

3.1 Optimization of Soursop Oil Extraction

Depicted in Table 2 are the coded variables, the experimental and predicted values obtained from the experimental. Table 3 shows the results of test of significance for every regression analysis. The results showed that the p-values of the model terms were remarkably significant, i.e. $p<0.05$.

The three linear terms (X_1, X_2, X_3), two cross-products (X_1X_2 and X_1X_3) and the two quadratic terms (X_1^2, X_3^2) were all found to be significant model terms at 95% confidence level. In order to minimize error, all the coefficients were considered in the design, and based on the large Fisher *F-test* (i.e. F-value) and low corresponding probability values (p-values), all the linear terms, the cross products (X_1X_3) and the quadratic terms ($X_1^2$and X_3^2) have very strong effects on the experimental oil yield. The results of the second-order response surface model fitting in the form of ANOVA are shown in Table 4. The model F-value of 385.50 with $p<0.0001$ showed that the regression was significance (Yuan *et al.*, 2008; Adepoju *et al.*, 2013).

The goodness of fit was checked by the coefficient of determination (R^2). In this study, the R^2 value of 0.9971 indicated the sample variation of 99.71% for oil extraction can be attributed to the independent variables and only 0.29% of the total variations were not explained by the model. The value of the adjusted determination coefficient (Adj. R^2 of 0.9933) and the predicted determination coefficient (Pred. R^2 of 0.9930) was also very high, supporting a high significance of the model (Khuri and Cornell, 1987).

The Adj. R^2 and Pred. R^2 should not be more than approximately 0.20 of each to be in "reasonable agreement". The value (0.0003) obtained showed that the model is in perfect agreement with each other. Meanwhile, the lack-of-fit term of 0.0956 was not significant relative to the pure error. In this case, a non-significant lack of fit is good. Table 5 shows the coefficient of estimate for the variable and their interactions, X_1X_2, X_1X_3, X_2X_3 and X_1^2 have negative effects on the oil yield at 95% CI. Hence, the model could be used in theoretical prediction of the oil extraction. The final equation in terms of coded variables for the Box-Behnken response surface second-order model is expressed in Eq. (3).

$$Y = 34.22 + 1.16X_1 + 2.57X_2 + 1.29X_3 - 0.51\ X_1X_2 - 3.92X_1X_3 + 0.20X_2X_3 - 6.48X_1^2 + 0.35X_2^2 + 4.38X_3^2 \quad (3)$$

The graph of predicted against the actual yield of the experimental runs are shown in Fig. 1. Observation from the graph suggested that the actual yield and the predicted values were in line with each other.

Table-3. Test of significance for all regression coefficient terms

Source	Sum of squares	df	Mean Square	F-value	p-value
X_1	10.72	1	10.72	65.99	<0.0001
X_2	52.84	1	52.84	325.34	<0.0001
X_3	13.26	1	13.26	81.65	<0.0001
X_1X_2	1.04	1	1.04	6.41	0.00392
X_1X_3	61.62	1	61.62	379.42	<0.0001

Continue

X_2X_3	0.16	1	0.16	0.99	0.3540
X_1^2	176.67	1	176.67	1087.74	<0.0001
X_2^2	0.51	1	0.51	3.13	0.1202
X_3^2	80.87	1	80.87	497.92	<0.0001

Table-4. Analysis of variance (ANOVA) of regression equation

Source	**Sum of Squares**	**df**	**Mean Square**	**F-value**	**p-value**
Model	385.50	9	42.83	263.73	<0.0001
Residual	1.14	7	0.16		
Lack of fit	0.87	3	0.29	4.32	0.0956
Pure error	0.27	4	0.067		
Cor total	386.64	16			
R^2 = 99.71%, R^2 (adj.) = 99.33%, Pred. R^2 = 96.30%					

Table-5. Regression coefficients and significance of response surface quadratic

Fact.	**Coefficient estimate**	**df**	**Standard error**	**95% CI Low**	**95% CI high**	**VIF**
Intercept	34.22	1	0.18	33.79	34.65	-
X_1	1.16	1	0.14	0.82	1.49	1.00
X_2	2.57	1	0.14	2.23	2.91	1.00
X_3	1.29	1	0.14	0.95	1.62	1.00
X_1X_2	-0.51	1	0.20	-0.99	-0.034	1.00
X_1X_3	-3.92	1	0.20	-4.40	-3.45	1.00
X_2X_3	-0.20	1	0.20	-0.68	0.28	1.00
X_1^2	-6.48	1	0.20	-6.94	-6.01	1.01
X_2^2	0.35	1	0.20	-0.12	0.81	1.01
X_3^2	4.38	1	0.20	3.92	4.85	1.01

The graphical representations of the regression equation for the optimization of Soursop oil extracted are displayed as contour and 3-D dimensional surfaces plot in Fig. 2(a-c). Fig. 2b and 2c showed a perfect mutual interaction between solvent volume with extraction time and solvent volume with sample weight and their reciprocal interaction on oil yield. The optimal values of the independent variables selected for the Soursop oil extraction were obtained by solving the regression equation (Eq. (3)) using the dx8 software. The optimal condition for this process was established at X_1= 38.10 g, X_2 = 30.00 min cm and X_3= 100 ml. The predicted oil yield under this optimal condition was Y = 34.6074 % (w/w) at 95% PI High. Using these optimal condition values for three independent replicates, a mean of 33.593 % (w/w) oil yield was achieved, which was within the range predicted by the model. The results of this work demonstrated that RSM with appropriate experimental design can be effectively applied to the optimization of the process variables in oil extraction from oilseeds.

Fig-1. Actual yield vs Predicted values

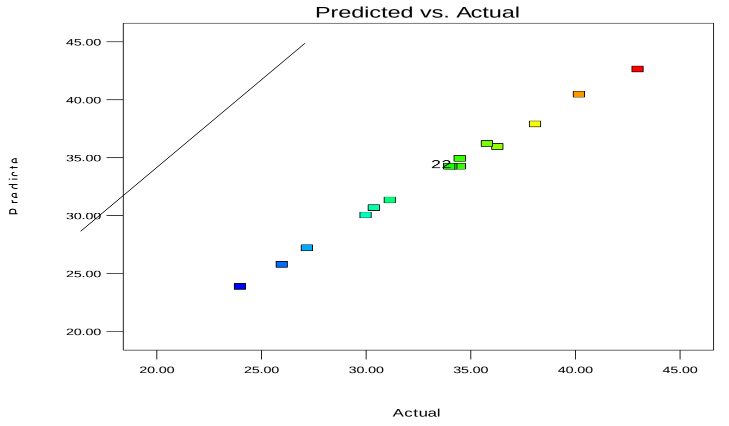
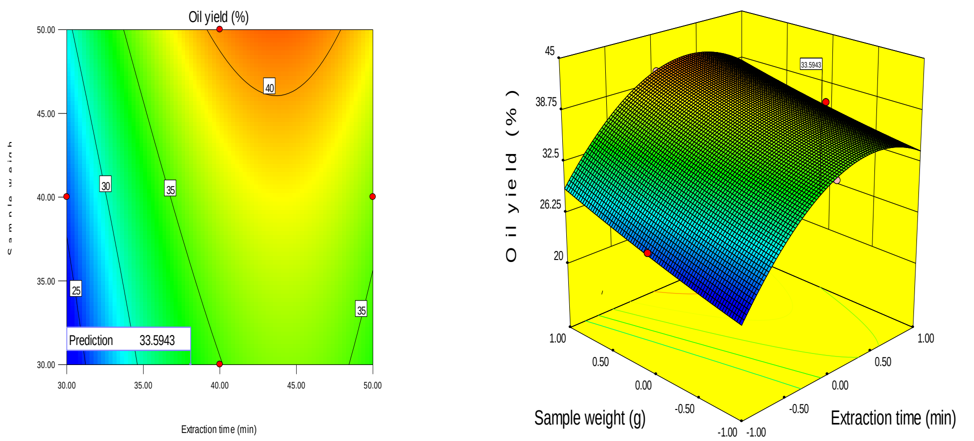
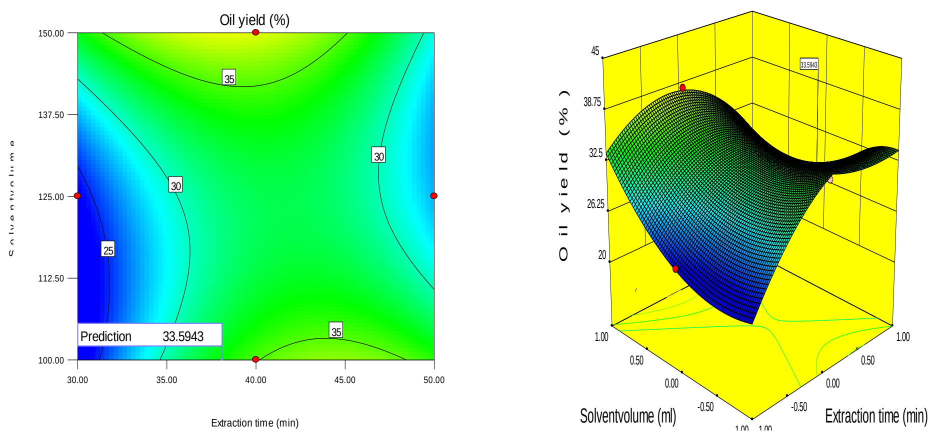


Fig-2(a-c). Contour and 3-D dimensional surfaces plots

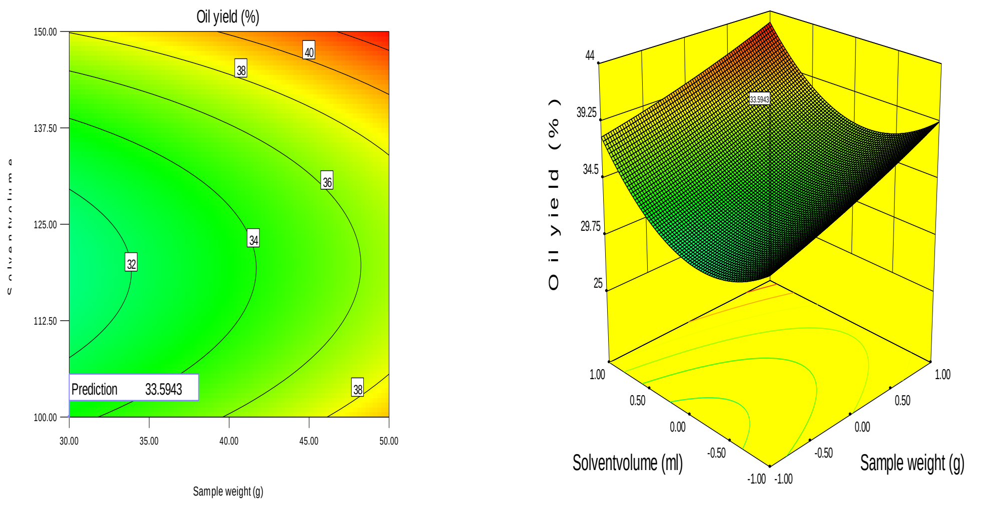
(a)



(b)



(c)



3.2. Physical and Chemical characteristics of the Crude Seed Oil

3.2.1. Physical characteristics of the Crude Seed Oil

In order to evaluate the quality of Soursop oil, the content and compositions of the oil were subjected to physicochemical analysis and the results obtained are shown in Table 6. At room temperature, the oil was pale yellow in colour with refractive index and moisture content of 1.526 and 1.20%, respectively. Observations on the colour, moisture content and refractive index of the oil agreed with previously published report by Okoro (2013). The specific gravity of the oil obtained was 0.8032. The viscosity, which is a measure of the resistance of oil to shear obtained in this work was 32.54 mPa.s.

3.2.2. Chemical characteristics of the Crude Seed Oil

Showed also in Table 6 is the results obtained for the chemical properties of crude Soursop oil. Low FFA content (0.91%) of the seed oil indicated good resistance to hydrolysis. The small acid value (1.82 mg KOH/g oil) of the seed oil showed that it is not only edible but could also have a long shelf life. A high saponification value of 235.46 (mg of KOH/g of oil) obtained, suggested high concentration of triglycerides.

The iodine value of the seed oil was high (115.30 g of I_2/100g oil), which signified the oil contained a substantial level of unsaturation. This observation is supported by the high level of unsaturated fatty acids present in the seed oil. Peroxide value measures the content of hydroperoxides in the oil and its low value indicates high resistance to oxidation. The value obtained for the oil in this work was 1.26 meq O_2/kg oil, which is within the limit stipulated for vegetable oils. The combination of high iodine value and low peroxide value suggests that the oil could also be stored for a long period without deterioration. These also demonstrate that the oil possesses the desirable qualities of edible oils. The Soursop oil could therefore be used for food purposes and as a feedstock in the industries.

Table-6. Physicochemical properties and other characteristics of seed oil

Parameters	Mean values
Physical properties	
Physical state at 28 °C	Pale yellow in colour
Refractive index at 25 °C	1.5260
Moisture content (%. wet.b)	0.012
Specific gravity	0.8032
Viscosity (mpa.s) at 40 °C	32.54
Chemical properties	
%FFA (as oleic acid)	0.91
Acid value (mg KOH/g oil)	1.82
Saponification value (mg KOH/g oil)	235.46
Iodine value (g I_2/100g oil)	115.30
Peroxide value (meq O_2/kg oil)	1.26

3.2.3 Fatty Acid Profile of the Crude Seed Oil

The results of Gas chromatography analysis of fatty acids present in the Soursop oil indicated that the oil does not only highly unsaturated (73.42%) but also have some degree of saturation (26.68%). Hence, the oil can be classified in the oleic-linoleic group.

4. CONCLUSIONS

The study showed that the Soursop seed is a good source rich in oil. Also, this work demonstrated Response Surface Methodology is a good tool for optimization in oil extraction processes. From design software employed in this work, the maximum seed oil yield of 34. 6074% (w/w) was established at the optimal condition of extraction time, 38.10 min, sample weight, 30.0 g and solvent volume, 100 ml. The optimized condition was validated with the actual seed oil yield of 33.593% (w/w). The fatty acid profile of the seed oil showed it is highly unsaturated (73.42%). Physicochemical analysis of the seed oil advocates it could serve as useful oil for food purposes and as a feedstock in many food and chemical industries.

5. ACKNOWLEDGEMENTS

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