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Optimization of Steam Distillation of Essential Oil of *Eucalyptus tereticornis* by Response Surface Methodology

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ABSTRACT: This paper deals with optimization of yield of *Eucalyptus tereticornis* oil in steam distillation using response surface methodology (RSM). The factors considered were mass of solute/solvent ratio (A), extraction time (B) and steam rate (C). These parameters were varied at two levels. Conditions of optimum oil yield predicted were 105 min of extraction time and 0.032 kg/h steam rate. These factors gave an optimum oil yield of 2.05 %. Significant model terms were time, steam rate, and the interactions between them. Analysis of variance (ANOVA) indicates that the model was significant as evidenced from R^2 of 0.9844 and the model F-value of 49.13. The oil yield predicted by the model was closed to the experimentally determined values (1.85 % and 1.87 % respectively); hence the model can be used for prediction of oil yield in essential oil extraction from *E. tereticornis* leaf via steam distillation method.

Keywords: RSM, *Eucalyptus tereticornis*, steam distillation, Box-Bohnken

INTRODUCTION

Essential oils are volatile compounds from aromatic plants that are widely used in foods, cosmetics, and pharmaceuticals. Generally, essential oils are complex mixtures of hydrocarbon monoterpenes, oxygenated monoterpenes, hydrocarbon sesquiterpenes, oxygenated sesquiterpenes, and related compounds that are derived from the secondary metabolism of plants (Reverchon, 1997). The oils are formed in green (chlorophyll bearing) parts of the plant and, with plant maturity are transported to other tissues particularly the flowering shots (Wijesekera *et al*., 1983). Studies have been conducted on optimization of essential oil yield using one factor at a time (OFAT) design by Okonkwo *et al*. (2006) and using 2^n factorial design (Silou *et al*., 2009; Muazu *et. al.,* 2012;) and Response surface methodology (Liu *et al*., 2009; Khajeh, 2011). However, conditions were not optimized for extraction of essential oil from *Eucalyptus tereticornis* specie. Moreover, the low concentration of essential oils in plants material requires extraction technique of high performance to achieve higher yields. These techniques and distillation parameters need optimization for each crop as plant matrix, oil content and constituents affect the production kinetics (Johns *et al*., 1992; Babu and Singh 2009).

Response Surface Methodology (RSM) is a collection of mathematical and statistical techniques useful for modelling and analyzing of problems in which a response of interest is influenced by several quantitative variables with the objective of optimizing the response (Montgomery, 2005). In this study, an attempt was made at optimizing steam distillation of essential oil of *E. tereticonis* specie using response surface methodology.

MATERIALS AND METHODS

Fresh *Eucalyptus tereticonis* leaves were obtained from Zaria, Nigeria (Latitude 11^{o} 05^{I} N, Longitude 7^{o} 43^{I} E) and dried at room temperature for seven days. Steam distillation was conducted using a 5 liters capacity stripping chamber connected to a 2000 liters capacity round bottom flask with a heating mantle. Steam temperature was maintained at 97^{o}C and the pressure was atmospheric. Cooling water was available at 10 cm^3/s at a temperature of 20^{o}C. The yield was obtained from steam distillation with the combination of the actual variables for each experimental run. Two experimental runs were conducted for each case and average value of the yield was reported.

Design of experiment

A 2^3 Box-Behnken Design (BBD) was employed resulting in a total of 17 experiments using the Design-Expert 6.0.6 version to optimize the chosen key factors namely mass of solute/solvent ratio A, distillation time B, and steam rate C. These variables each at two levels, low and high: A (0.0667 – 0.1333), B (1200 s – 7200 s) and C (8.30E-06 – 5.80E-05) are presented in Table 1. These levels were chosen based on capacity of the experimental set up for variables A and C, while B was selected based on experimental run time. The experimental design is shown in Table 2.

The regression analysis was performed to estimate the response function as a second order polynomial

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i^2 + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_{i=1}^{k-1} \sum_{j=2}^{k} \beta_{ij} X_i X_j \tag{1}$$

where Y is the predicted response, X_i is the uncoded value of the ith test variable, β_0, β_j, and β_{ij} are coefficients estimated from the regression according to Rajasimman *et al*. (2009).

Table 1: Design summary

Factors	Name	Units	Low Actual	High Actual	Low Coded	High Coded
A	Solute/solvent ratio (mass)	kg/kg	0.0667	0.1333	-1	1
B	Time	S	1200	7200	-1	1
C	Steam rate	kg/s	8.300E-06	5.80E-05	-1	1

A statistical program package Design Expert 6.0.6 was used for regression analysis of the data and for estimating the coefficient of the regression equation. The equations were validated by the analysis of variance (ANOVA) test. Model and regression coefficients were considered significant when P-values were lower than 0.05 (Liu *et al*., 2009; Wijingaard and Brunton, 2010).

Table 2: Box-Behnken Design matrix

Run	A	B	C	Yield %
1	0	0	0	1.84
2	0	0	0	1.97
3	-1	0	1	1.6
4	-1	0	-1	1.93
5	0	-1	1	0.8
6	1	-1	0	0.5
7	1	0	1	1.78
8	-1	-1	0	0.86
9	0	0	0	1.87
10	0	1	1	1.66
11	0	-1	-1	0.74
12	0	0	0	1.81
13	1	0	-1	1.84
14	1	1	0	1.98
15	0	0	0	1.75
16	0	1	-1	2.27
17	-1	1	0	2.03

RESULTS AND DISCUSSION

Table 2 presents the Box-Behnken Design matrix and the oil yield obtained for each experimental run.

Analysis of variance for the response surface model is presented in Table 3. The analysis indicate that the model F-value of 49.13 implies that the model is significant. There is only a 0.01% chance that a “Model F-Value” this large could occur due to noise, the model also has satisfactory level of adequacy (R^2). Model terms B, C, B^2 and BC are significant terms ($P < 0.05$). The lack of fit F-Value of 2.16 implies the lack of fit is not significant relative to the pure error. There is a 23.54 % chance that a “Lack of fit-Value this large could occur due to noise.

The “Pred R^2 of 0.8365 is in reasonable agreement with the “Adj R^2 of 0.9644. Adeq Precision measures the signal to noise ratio; the ratio of 22.10 obtained indicates an adequate signal (a ratio greater than 4 is desirable). The model can therefore be used to navigate the design space.

The response surface curves are plotted to understand the interaction of the variables and the optimum level of each variable for maximum response (Rajasimman *et. al*., 2009). The response surface curves for extraction of essential oil from *Eucalyptus tereticornis* using steam distillation are presented in Figures 1 to 3. Figure 1 shows interaction of mass and time on the yield, while Figures 2 and 3 indicate interaction of mass and steam rate on the yield as well as that of steam rate and time on the yield, respectively. The interaction effects between the variables are significant, as evidenced from the elliptical nature of the contours.

Table 3: Analysis of variance (ANOVA) for response surface quadratic model to identify significant factors affecting the oil yield

Source	Coefficient Estimate	Sum of Square	DF	Mean square	F Value	Prob > F
Model		4.38	9	0.49	49.13	< 0.0001*
Intercept	1.85		1			
A	-0.040	0.013	1	0.013	1.29	0.2932
B	0.63	3.18	1	3.18	320.36	< 0.0001*
C	-0.12	0.11	1	0.11	11.14	0.0125*
A^2	-0.043	0.0077	1	0.0077	0.78	0.4075
B^2	-0.46	0.90	1	0.90	90.97	< 0.0001*
C^2	-0.018	0.00133	1	0.00133	0.13	0.7253
AB	0.078	0.024	1	0.024	2.42	0.1634
AC	0.067	0.018	1	0.018	1.84	0.2172
BC	-0.17	0.11	1	0.11	11.32	0.0120*
Residual		0.069	7	0.00991		
Lack of Fit		0.043	3	0.014	2.16	0.2354
Pure Error		0.026	4	0.00662		
Cor Total		4.45	16			

Standard deviation: 0.1; R^2: 0.984; mean: 1.60; adj R^2: 0.964; pred R^2: 0.8365; adeq precision: 22.100
*significant variable
Final equation in terms of coded and real variables are given by equations (2) and (3) respectively. Significant model terms ($P < 0.05$) were the variables used in the model to describe the response (yield).

$$\text{Yield} = 1.85 + 0.63B - 0.12C - 0.46B^2 - 0.17BC \qquad (2)$$

$$= 0.20218 + (6.38804 \times 10^{-4})B - 1543.08088C - (5.14167 \times 10^{-8})B^2 - 2.24681BC \qquad (3)$$

Where: A = solute/solvent mass ratio, B = time a nd C= steam rate.

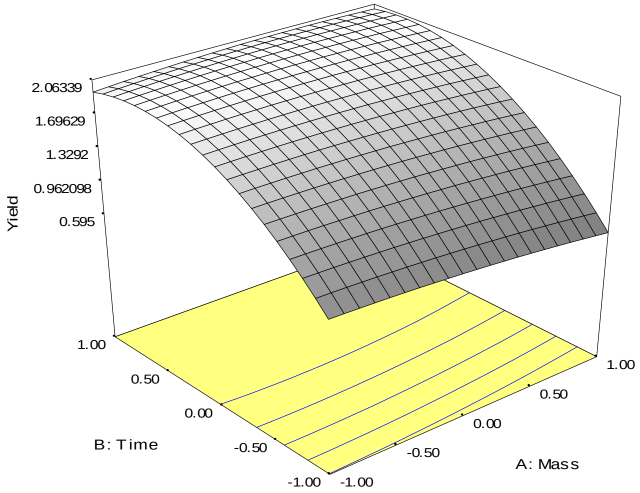

Figure 1: Response surface showing effect of mass and time on the yield at constant steam rate

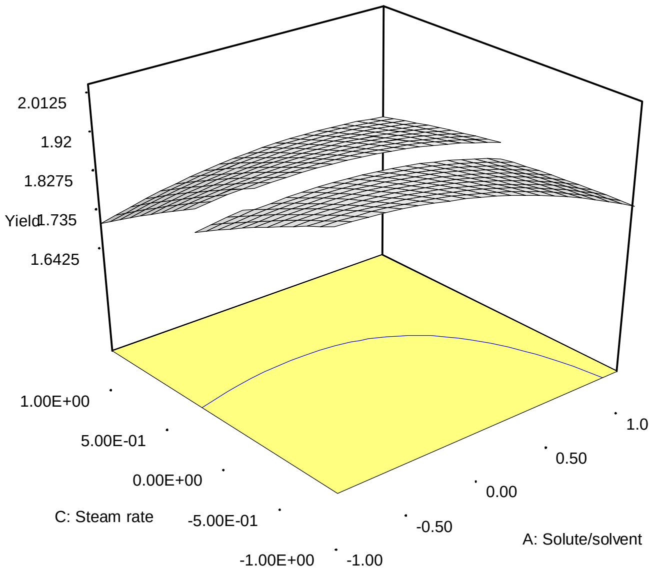

Figure 2: Response surface showing effect of mass and steam rate on the yield at constant time

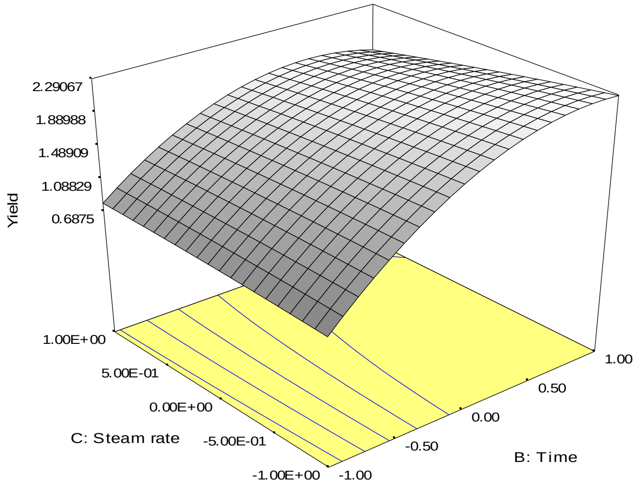

Figure 3: Response surface showing effect of steam rate and time on the yield at constant solute/solvent ratio

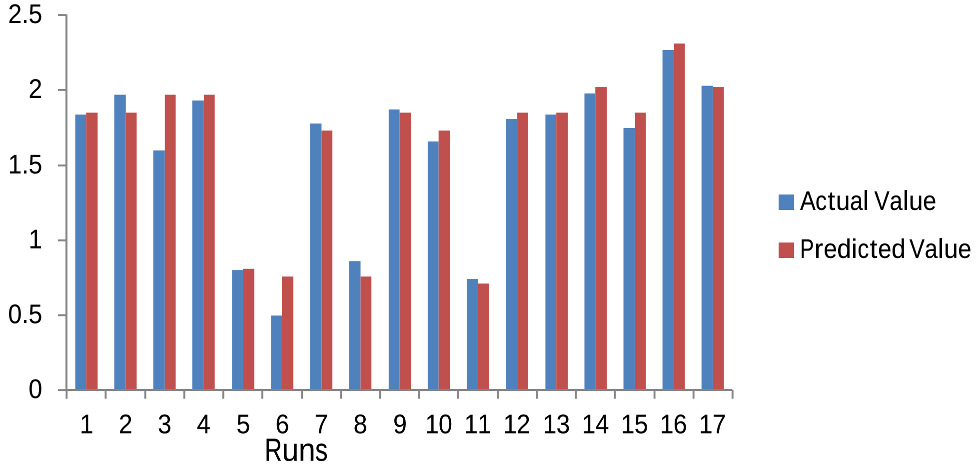

Figure 4: Actual oil yield and prediction from the model

Experimental data generated in Table 2 in terms of coded values were substituted into equation 2 and the predicted oil yield was obtained. Actual oil yield and the predicted oil yield are presented in Figure 4; Comparison of these values indicates that the 2 sets of values are in close agreement. This suggest good reliability of the model as also evidenced from the statistical parameters of the model such as standard deviation of 0.1, R^2 of 0.9844 and F-value of 49.13. This shows fitness of the data to the model.

Numerical optimization method was used to predict optimum condition for the response using the Design-Expert package 6.0.6. Extraction time of 105 min and 0.032 kg/h steam rate were obtained among the solutions for the optimum conditions for the yield. This condition gives a yield of 2.05 %.

CONCLUSION

Optimization model was developed using response surface methodology technique for prediction of essential oil yield in steam distillation of *Eucalyptus tereticornis* leaf. The model fits experimental data. Extraction time, steam rate and their interactions were the major process parameters found to significantly influence the oil yield. Conditions of optimum oil yield predicted were 105 min of extraction time and 0.032 kg/h steam rate, which gives 2.05 % oil yield.

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