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EXPERIMENTAL AND MODELING STUDIES OF VIETNAM LEMONGRASS ESSENTIAL OIL EXTRACTION PROCESS USING RESPONSE SURFACE METHODOLOGY

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ABSTRACT

Natural precious products such as aroma compounds, essential oils, and bio-activated materials are usually extracted from about 30,000 botanical species. These extracts are often high competitive market due to their small content (less than 1 %) in plants and high purification cost. Thus, development of a modeling for the optimization of the crude oil extraction is highly paid attention. In this work, a modeling of Vietnam lemongrass oil extraction using steam distillation is developed and the optimization of the process parameters is performed using response surface methodology (RSM). The operating parameters considered for the modeling and optimization are specific area of raw materials, moisture content of feedstock, and steam rate. Experimental data show that the oil yield from steam distillation of Vietnam lemongrass is significantly affected by the three mentioned factors. Box-Behnken design (BBD) and analysis of variance (ANOVA) are used to examine the effects of operating parameters on the extraction efficiency. On the basis of the measurements and RSM, a quadric regression model as a function of steam rate, specific area and moisture content of materials is estimated. The optimized operating conditions of the lemongrass hydrodistillation are also obtained by applying the proposed modeling.

Keywords: lemongrass essential oil, steam distillation, modeling, optimization, response surface methodology (RSM).

1. INTRODUCTION

Response surface methodology (RSM) has been largely used for optimizing the operating conditions of a process. RSM consists of a group of mathematical and statistical techniques that can be used to evaluate the relationships between the response and the independent variables in which effects of individual or combination of variables can be examined. The main advantage of RSM is the reduction of experimental runs to evaluate multiple parameters while capturing well their interactions. Therefore, RSM is a useful tool that can be applied to the development, improvement and optimization of operating conditions for a process [1].

Lemongrass is one of the largely cultivated medicinal plants for its essential oil in parts of tropical and subtropical areas of Asia. However the low content of essential oil in plant material (less than 1 %) needed an extraction technique with optimal performance to achieve maximum yield, thus process parameters are often to be optimized [2, 3]. In the present study, RSM was carried out to evaluate the optimized operating conditions for the maximum yield of essential oil from lemongrass (*Cymbopogon Citratus)* using steam distillation. The parameters considered for the analysis were steam rate, specified area, and moisture content of initial materials. The composition of essential oil of lemongrass was also analyzed.

2. MATERIALS AND METHODS

2.1. Materials

Fresh leaves and bulb of lemongrass (10 kg) used in the study was collected from Quang Nam province. The plant material was dried at room temperature (25 ^{o}C). It was kept in a sealed plastic bag at ambient temperature after the drying step. The samples were then cut, grind at room temperature prior to extraction.

2.2. Experimental design and statistical analysis

According to the Box-Behnken design, 17 experimental runs were prepared accounting for combinations of independent variables including steam rate (3 kW, 4.5 kW, and 6 kW), material size (5 cm, 8.5 cm, and 12 cm), and moisture of material (15%, 37.5%, and 60%). The extraction time of each experiment is 180 min under atmospheric pressure.

The relationship between the response and the input is given by Eq. (1) [1, 4]:

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

where η is the response, f is the unknown function of response, x_1, x_2, …, x_n denote the independent variables, also called natural variables, n is the number of the independent variables and ε is the statistical error that represents other sources of variability not accounted for by f.

Because it is not possible to identify the effects of all parameters, therefore only selected parameters that have major effects can be analyzed. The major parameters selected in this work are size of material (x_1), moisture of material (x_2), and steam rate (x_3). Each of the coded variables is forced to the range from -1 to 1. Commonly used equation for coding is expressed as Equation (2)

$$X = \frac{x - (x_{max} + x_{min})/2}{(x_{max} - x_{min})/2} \qquad (2)$$

and the second order model (quadratic function) can be written as Equation (3)

$$y = \beta_0 + \sum_{j=1}^{k} \beta_j X_j + \sum_{j=1}^{k} \beta_{jj} X_j^2 + \sum_{i<j} \beta_{ij} X_i X_j \qquad (3)$$

where $\beta_0, \beta_j, \beta_{jj}$ and β_{ij} are regression coefficients for intercept, linear, quadratic and interaction coefficients respectively and X_i and X_j are coded independent variables.

The response surfaces are the theoretical three dimensional plots showing the relationship between the response and the independent variables. These plots give useful information about the model fitted.

Table 1 shows the 17 combinations of three independent variables. The significance of each coefficient was determined by using F-test and p-value. The corresponding variables would be more significant if the F-value becomes greater and the p-value becomes smaller.

Table1. Experimental arrangement, response, and predicted values for lemongrass oil yield.

Run	Coded variables			Independent variables			Essential oil yield (Y) (%)		
	X_1	X_2	X_3	x_1	x_2	x_3	Experimental (Y_0)	Predicted (Y_i)	Y_0 - Y_i
1	-1	0	-1	5	37.5	3	1.095	1.224	-0.129
2	1	0	-1	12	37.5	3	0.452	0.469	-0.017
3	0	-1	-1	8.5	15	3	0.901	0.928	-0.027
4	0	1	-1	8.5	60	3	0.771	0.677	0.094
5	1	1	0	12	60	4.5	0.773	0.885	-0.112
6	0	0	0	8.5	37.5	4.5	1.347	1.334	0.013
7	-1	1	0	5	60	4.5	0.859	0.826	0.033
8	0	0	0	8.5	37.5	4.5	1.347	1.334	0.013
9	0	0	0	8.5	37.5	4.5	1.326	1.334	-0.008
10	0	0	0	8.5	37.5	4.5	1.337	1.334	0.003
11	0	0	0	8.5	37.5	4.5	1.347	1.334	0.013
12	-1	-1	0	5	15	4.5	1.533	1.421	0.112
13	1	-1	0	12	15	4.5	0.858	0.891	-0.033
14	1	0	1	12	37.5	6	1.033	0.865	0.168
15	0	1	1	8.5	60	6	0.358	0.360	-0.002
16	0	-1	1	8.5	15	6	0.673	0.796	-0.123
17	-1	0	1	5	37.5	6	0.571	0.515	0.056

2.3. GC-MS conditions

Composition of the extracted essential oil from lemongrass was analyzed by gas chromatography–mass spectrometry (GC-MS). The analysis of the essential oils was performed in the capillary column (30 m, 0.32 mm i.d., 0.25 μm film thickness). Column temperature was initially 40 ^oC for 2 minutes, and then gradually increased to 225 ^oC at the rate of 4 ^oC/min. The extracts were diluted 3:100 (v/v) with acetone 99.99 %. Temperature of the injector and detector were set at 290 ^oC and 175 ^oC, respectively. Split ratio was set at 1:100 and the carrier gas was helium operated at a flow rate of 2.2 ml/min [2].

3. RESULTS AND DISCUSSION

The equation is an empirical relationship between lemongrass essential oil of yield and the test variable in coded unit is given by Equation (4) as follows.

$$Y = -0{,}778 - 0{,}186x_1 + 0{,}0167x_2 + 1{,}321x_3 - 9{,}104.10^{-3}x_1^2 - 4{,}415.10^{-4}x_2^2 - 0{,}196x_3^2 + 1{,}869.10^{-3}x_1x_2 + 0{,}0526x_1x_3 - 1{,}37.10^{-3}x_2x_3 \quad (4)$$

The predicted yield of lemongrass essential oil obtained the regression model in comparison with experimental data is shown in Figure 1.

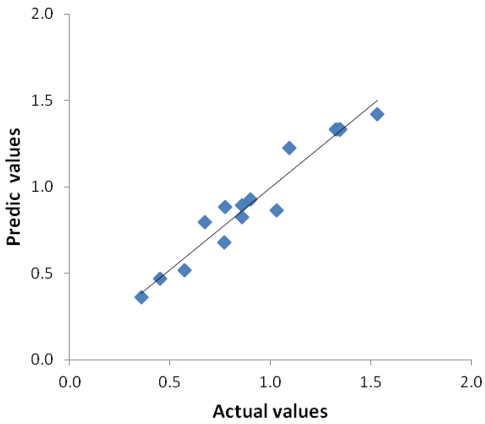

Figure 1. Comparison between predicted and actual lemongrass essential oil yield.

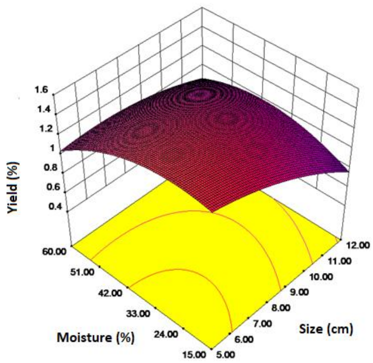

Figure 2. Response surface plots showing the effect of material size (x_1) and material moisture (x_2) on extraction yield of lemongrass.

The three dimension surface plots were drawn to illustrate the main and interactive effects of the independent variables on the objective function. These graphs were obtained by fixing one variable at coded zero level while varying the other two variables.

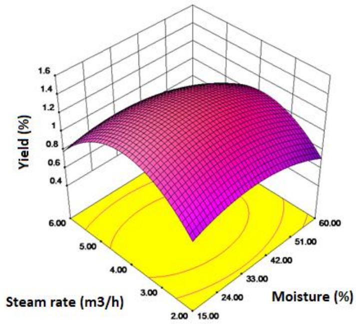

Figure 3. Response surface plots showing the effect of steam rate (x_3) and material moisture (x_2) on extraction yield of lemongrass

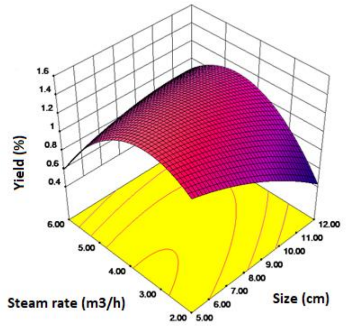

Figure 4. Response surface plots showing the effect of material size (x_1) and steam rate (x_3) on extraction yield of lemongrass

Figure 2 shows the effects of both material size (x_1) and moisture (x_2) on extraction yield. Effects of steam rate (x_3) and moisture (x_2) on oil yield is presented in Figure 3 and Figure 4 presents influence of steam rate (x_3) and material size (x_1). The results revealed that the decrease of material size extracted higher amount of lemongrass oil yield. This phenomenon can be explained that increasing specific area of material will increase contacting ability between steam-rate and material. Also, the results indicate that extraction yield increases when steam rate increase between 2 to 4 m3/h. However if we continue increasing steam rate to 6 m3/h the extraction yield will be decrease. This result can be explained that essential oil dissolved into steam rate.

Figure 5. Optimization of the process by “Ramp function”.

The suitability of the model equation for predicting the optimum response value using Ramp function was tested using the recommended optimum conditions (Figure 5). When optimum values of independent variables (material size 5 cm, moisture content 46.54 %, steam rate 3.15 m^3/h) were applied, predicted extraction yield was 1.5296 % whereas the yield of 1.4029 % was obtained from the experiment. Thus, predicted values from fitted equations and observed values were in good agreement.

According to the GC/MS analysis, Limonene, Citronellal, Citronellol and Geraniol are major components obtained from Quang Nam lemongrass. The composition of sample was compared to VietNam standard of lemongrass oil (TCVN 11425:2016) and other work [5, 6] in which Limonene, Citronellal, and Citronellol is within the standard range while Geraniol is much higher. Detailed measurements are shown in Table 2.

Table 2. GC/MS of lemongrass essential oil obtained by steam distillation.

No	**Component**	**Composition (%)**	
		Quang Nam lemongrass oil	**TCVN 11425:2016**
1	Limonene	3.29510	2.0 - 5.0
2	Citronellal	31.0432	31.0 - 40.0
3	Citronellol	10.0025	8.5-14.0
4	Geraniol	27.8635	20.0-25.0

4. CONCLUSIONS

RSM was utilized to describe and predict the extraction process of bioactive compounds from lemongrass oil. The extraction of essential oils from Quang Nam lemongrass was extracted by steam distillation with 17 combinations of material size, moisture content, and steamrate. The experimental values of lemongrass yield varied from 0.358 % to 1.533 %. The variable with the largest effect was material size. The optimal operating condition obtained for the maximum oil yield from the extraction of Quang Nam lemongrass is material size 5 cm, moisture content 46.54 %, steam rate 3.15 m^3/h.

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