OPTIMIZING THE PROCESSING OF GINGER ESSENTIAL OIL YIELD (Zingiber collinsii) FROM PUMAT NATIONAL PARK OF VIETNAM USING THE RESPONSE SURFACE METHODOLOGY

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ABSTRACT

In Vietnam, ginger (Zingiber) belonging to ginger family (Zingiberaceae) is a valuable spice plant. Extraction is the first step of great importance for the recovery and purification of essential oil from plant materials. In this paper, we studied on the factors that affect the process of total essential oil extraction from ginger roots originated from Pu Mat National Park, Nghe An province. The total essential oil yield extraction from ginger roots was optimized using Response Surface Methodology (RSM) with experimental order of Box-Behnken design. The final optimization value for yield of ginger essential oil is at 570 ml water to 100 g sample at 290 minutes of steam distillation using 1.09 mm material thickness size. In this condition, the maximum total essential oil yield was predicted to be 1.23 %.

Keywords: optimization, Zingiber collinsii, Box-Behnken Design, fresh ginger.

1. INTRODUCTION

The genus Zingiber (Zingiberaceae), which has about 150 species distributed in almost all tropical and subtropical countries (Southeast Asia, China, India, Nigeria...). In Vietnam, the genus is diverse with about 10 endemic species [1, 2].

Ginger roots and its essential oil have been used in food, spice, beverages, medicine and many religious activities... with many types fresh, dried, essential oil, oleoresin... [1-3].

Essential oil from ginger can be isolated by hydro-distillation, steam distillation; steam distillation and hydro-distillation are traditional methods for removal of essential oils for food and medicinal [4-6]. Ida Hasmita et al. have studied particle size and duration on the amount of essential oil obtained [7]. Fhtaheya Buang et al. reported that the yield of ginger oil was significantly affected by water volume, hydro-distillation time and particle size [8]. Besides
that, the total essential oil yield depends on other factors such as: drying method of material, parts of plant for distillation, genus of ginger, environment, and time of stored, etc. [4 - 6].

The objective of this work was to study the extraction process of ginger essential oil using steam distillation and the desirability methodology was employed to optimize total essential oil distilled from fresh roots of *Zingiber collinsii*.

2. MATERIALS AND METHODS

2.1. Plant materials

The fresh rhizomes of ginger plant (*Zingiber collinsii*) were collected in Pu Mat National Park, Nghe An province. Voucher specimens were identified and deposited at the Botany Museum Vinh University, Vietnam. Ginger roots were washed and unpeeled.

2.2. Distillation of essential oil

The fresh gingers (100 g) were sliced with different thickness material size of 1, 1.5, 2, 2.5, 3 and 3.5 mm. Then the material was treated by steam distillation using Clevenger apparatus at different water/material ratio of 100, 200, 300, 400, 500, 600, 700 and 800 mL for 100 g material, different distillation times of 60, 120, 180, 240, 300 and 360 minutes. The extracted essential oil was dehydrated with anhydrous sodium sulfate to isolate ginger oil.

The total essential oil yield is performed as:

\[
Y = \frac{v \times d}{100 - w} \times 100 \%
\]

Y (%): total ginger essential oil yield; v (ml): volume of ginger essential oil from distillation; d: ginger essential oil density; w (g): moisture of material, determined by distillation with toluene method.

2.3. Experimental design

Statistical experimental design method: Using the statistical Box-Behnken design with three factors: material size (mm), water/material ratio (v/w) and distillation time (min) (Table 1).

| Factors             | Symbols | Units | Range and level |
|---------------------|---------|-------|-----------------|
| Material size       | A       | mm    | 1, 1.5, 2       |
| Water/material ratio| B       | v/w   | 4/1, 5/1, 6/1   |
| Distillation time   | C       | min   | 180, 240, 300   |

This design has 17 actual experiments with 3 factors (k = 3), 3 level with 5 center points to form a central composite design with response: total essential oil (%) (Table 2).
Table 2. Experimental design and total oil yield.

| Run | Material size (mm) | Water/material ratio (v/w) | Distillation time (min) | Total oil yield (%) |
|-----|-------------------|--------------------------|-------------------------|-------------------|
| 1   | 1.00              | 4.00                     | 240.00                  | 0.80              |
| 2   | 2.00              | 4.00                     | 240.00                  | 0.75              |
| 3   | 1.00              | 6.00                     | 240.00                  | 1.20              |
| 4   | 2.00              | 6.00                     | 240.00                  | 0.84              |
| 5   | 1.50              | 4.00                     | 180.00                  | 0.51              |
| 6   | 1.50              | 6.00                     | 180.00                  | 0.91              |
| 7   | 1.50              | 4.00                     | 300.00                  | 1.05              |
| 8   | 1.50              | 6.00                     | 300.00                  | 1.06              |
| 9   | 1.00              | 5.00                     | 180.00                  | 0.75              |
| 10  | 2.00              | 5.00                     | 180.00                  | 0.64              |
| 11  | 1.00              | 5.00                     | 300.00                  | 1.22              |
| 12  | 2.00              | 5.00                     | 300.00                  | 0.97              |
| 13  | 1.50              | 5.00                     | 240.00                  | 0.98              |
| 14  | 1.50              | 5.00                     | 240.00                  | 0.96              |
| 15  | 1.50              | 5.00                     | 240.00                  | 0.97              |
| 16  | 1.50              | 5.00                     | 240.00                  | 0.95              |
| 17  | 1.50              | 5.00                     | 240.00                  | 0.99              |

2.4. Statistical analysis

The statistical Design-Expert 7.1 software (Stat-Ease, Inc., Minneapolis, USA) was used for regression analysis of experimental data to plot response surface and to optimize according to the desirability methodology [9, 10]. ANOVA (Analysis of variance) was used to estimate the statistical parameters of regression model. The RSM method makes use of an objective function, $d(x)$, called the desirability function. In this paper, the "Goal" field for response must be "maximum":

$$d_i = \begin{cases} 0 & \text{if } Y_i < Y_{\text{min}} \text{ (min value accepted)} \\ \left[ \frac{Y_i - Y_{\text{min}}}{Y_d - Y_{\text{min}}} \right]^8 & \text{if } Y_{\text{min}} \leq Y_i \leq Y_d \text{ (target value)} \\ 1 & \text{if } Y_i > Y_d \text{ (target value)} \end{cases}$$

3. RESULTS AND DISCUSSION

3.1. Study on the factors influencing on total essential oil
Optimization of ginger essential oil yield (Zingiber collinsii) from North Central region of Vietnam

The total essential oil amount depended on three factors: material size (mm) (Figure 1), water/material ratio (v/w) (Figure 2) and distillation time (min) (Figure 3). Based on the experimental data, we decided to choose value ranges for particular factors as follows: material size 1 - 2 mm, water/material ratio 4/1 - 6/1, time of distillation 180 - 300 minutes.

3.2. Model building

The results of experimental design are shown in Table 2. The response of total essential oil yield was fitted with the second-order polynomial equation of three factors. The significance of the model and the coefficients are controlled by regression analysis (Table 3). The F-value of the model is 116.29 showing that the model is completely significant of 99.99% (p < 0.0001) confidence level. The p-value of less than 0.05 indicates that the coefficient is significant. F-value for “lack of fit” of the model was 3.5 (p = 0.1288). It showed that the model was fit with the experiment. In addition, the coefficient of determination (R²), which was 0.9934, showed that the model was well matched with the designed experiment. The prediction R² of 0.9202 is also in reasonable agreement with adjusted R² of 0.9848.

The total essential oil yield is performed in the second-degree model as follows:

\[ Y = 0.97 - 0.096A + 0.11B + 0.19C - 0.077AB - 0.035AC - 0.098BC - 0.03A^2 - 0.042B^2 - 0.045C^2 \]
Table 3. Regression analysis of total essential oil.

| Source            | Mean Square | F value | p-value (prob > F) |
|-------------------|-------------|---------|-------------------|
| Model             | 0.060       | 116.29  | <0.0001           |
| A- material size  | 0.074       | 143.11  | <0.0001           |
| B- water/material ratio | 0.10     | 195.52  | <0.0001           |
| C- distillation time | 0.28     | 535.89  | <0.0001           |
| AB                | 0.024       | 46.39   | 0.0003            |
| AC                | 4.900E-003  | 9.46    | 0.0179            |
| BC                | 0.038       | 73.43   | <0.0001           |
| A²                | 3.789E-003  | 7.32    | 0.0304            |
| B²                | 7.605E-003  | 14.69   | 0.0064            |
| C²                | 8.526E-003  | 16.46   | 0.0048            |
| Lack of Fit      | 8.750E-004  | 3.50    | 0.1288            |

Considering in turn the effect of each factor on the total essential oil yield distilled (when other factors are remained at the central level) (Figure 4), it showed that the distillation time (C) has played a significant role on the total essential oil yield, while the material size (A) less impacted than distillation time (C) on total essential oil yield. Meanwhile, water/material ratio (B) took impact as the least role on the total essential oil yield. This can be shown obviously in the response surface of Y function (Figures 5a, 5b, 5c).

![Perturbation diagram](image)

Figure 4. Influence of factors on total essential oil yield.
3.3. Optimization of the total essential oil yield distillation

The most importantly is to obtain the highest yield of essential oil. To this point, the optimization using algorithm of desirability methodology invented by Derringer and Suich was applied. The results after using the Design-Expert 7.1 Software were the followings: material size 1.09 mm, water/material ratio 5.7/1 (v/w) and time of distillation 290 minutes. In this condition, the predicted maximum total essential oil distillation was 1.23%. The total essential oil has obtained 100% desirability of objective proposed (Figure 6). This result of our research has matched with the reports of other authors about optimization of essential oil yields for other genus ginger (zingiber) in the ginger family (zingiberaceae) [8, 7, 11, 12].

3.4. Validation of the model

The trial experiments were conducted (three times) under optimized process conditions with material size 1.09 mm, water/material ratio 5.7/1 (v/w) and time of distillation 290 minutes. The mean value of the total essential oil has reached 1.21 ± 0.03 % (Table 4), which result very
close to the predicted total essential oil obtained from regression equation (1.23%). Thus, the model could be used to predict the total essential oil yield obtained from the hydodistillation.

Table 4. The distillation results under optimal condition.

| Number | Material size (mm) | Water/material ratio (v/w) | Distillation time (min) | Total essential oil (%) |
|--------|--------------------|---------------------------|------------------------|------------------------|
| 1      | 1.09               | 5.7/1                     | 290                    | 1.21                   |
| 2      | 1.09               | 5.7/1                     | 290                    | 1.20                   |
| 3      | 1.09               | 5.7/1                     | 290                    | 1.23                   |

Average 1.21 ± 0.03

4. CONCLUSIONS

Based on the statistical experimental design using response surface and desirability methodology, the optimal conditions for hydro distillation of ginger rhizome were determined: material size was of 1.09 mm; water/material ratio was 570 ml to 100 g; distillation time was of 290 minutes and the maximum ginger essential oil was predicted to be 1.23%.

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TÓM TÁT

TOI UU HÓA QUÁ TRÌNH CHUNG CÁT TINH DẦU GỪNG (Zingiber collinsii) TỪ RÚNG QUỐC GIA PUMAT VIỆT NAM BẰNG PHƯƠNG PHÁP BÉ MẤT ĐẠP ỦNG

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Ở Việt Nam, các loại gừng (zingiber) thuộc họ gừng (zingiberaceae) là một loại gia vị có giá trị và được ưa dùng nhiều trong đời sống. Chất xuất là bước đầu tiên rất quan trọng để thu hài tinh dầu từ các nguyên liệu thực vật. Trong bài báo này, chúng tôi trình bày nghiên cứu về các yếu tố ảnh hưởng đến quá trình chưng cất tinh dầu rõ lợi gừng (Zingiber collinsii) thu há ở Vườn Quốc gia Pù Mát, tỉnh Nghệ An. Tổng lượng tinh dầu thu được từ rõ gừng đã được tối ưu hóa bằng cách sử dụng phương pháp bè mát đáp ứng với trình tự thực nghiệm theo thiết kế Box-Behnken. Giảm tỷ lệ trụ ưu của tinh dầu gừng được dự đoán là 1,23 % tại điều kiện thực nghiệm ở 570 ml nước cho 100 g mấu, thời gian chưng cất 290 phút với chiều dày nguyên liệu là 1,09 mm.

Từ khóa: tòi ưu hóa, Zingiber collinsii, thiết kế thí nghiệm theo Box Behnken, gừng tươi.