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Response Surface Optimization of Essential Oils Production from Clove Leaf Waste by Microwave-Assisted Hydro Distillation

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Response Surface Optimization of Essential Oils Production from Clove Leaf Waste by Microwave-Assisted Hydro Distillation

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Abstract. Essential oils can be obtained from leaves, flowers, stems, and roots. One of the ingredients that has the potential to become essential oils is clove plants. The content of essential oils in clove leaves is between 1-4%. The process of refining essential oils can use the microwave assisted hydrodistillation method. The use of microwaves is to speed up processing time. Optimization using the Response Surface Methodology (RSM) method is based on Central Composite Design (CCD). The research variables of micro wave power are 400-800 watt and clove leaf mass 70-90 gram. The optimization results obtained at 599.5 watts of microwave power and clove leaf mass of 65.86 grams with a yield of 1.1835%. The second order polynomial equation is obtained $y = 0.827502 + 0.493345X_1 - 0.179890X_2 - 0.3363901X_1^2 + 0.228974X_2^2 + 0.227850X_1X_2$.

1. Introduction

Clove (Syzygium aromaticum) is widely cultivated in Indonesia, India, Malaysia, Sri Lanka, West India, Madagascar and Tanzania [1]. Clove plants aged more than 20 years old have dry leaves that can be collected an average of 0.96 kg / tree each week, while the plants aged less than 20 years can be collected as much as 0.46 kg / tree [2]. In 2006-2016 the total area of clove plantations in Indonesia was increasing every year, in 2016 the total area was 542,281 hectares. This was followed by an increase in the amount of production in 2006-2016, the amount of clove production in 2016 was 139,552 tons [3]. The abundant clove plants have the potential to produce essential oils. Clove is more often used as a mixture of clove cigarettes, but the leaves, stems, and flowers can be used as a source of clove oil or essential oil in the pharmaceutical, cosmetic, perfume, food and beverage industries [4], and it also can be used for therapy in biomedical applications [5]. Clove oil has biological activities, such as antibacterial, antifungal, insecticide, and antioxidant properties, and it is used traditionally as a reinforcing agent and antimicrobial agent in food [6]. According to Nurdjannah [4], clove leaves have an essential oil content of 1-4%.

The process of extracting essential oils using the distillation method is the oldest method, but it is still used by craftsmen in developing countries including Indonesia [7]. Distillation methods, either steam distillation or hydrodistillation, have been shown its ability to extract and characterize clove oil from buds or clove leaves [8]. The method often used is hydrodistillation in a conventional and microwave-assisted way. The conventional method requires a long time to obtain oil, so the process becomes less economical. Microwave-assisted hydrodistillation is extracting dissolved substances in plant material with the help of microwave energy [9]. Microwave-assisted hydrodistillation (MAHD) uses a microwave as an eco-friendly source of energy, and the extraction process is fast, so it is more economical and efficient [10]. Microwave power is one of the factors that influence the efficiency of extraction using MAHD because microwave energy will significantly affect molecular interactions between one compound and another [11].

The determination of optimum result can be done with Response Surface Methodology (RSM) which is a mathematical and statistical technique to analyze problems and to get a mathematical formula. RSM is an effective way for the optimization process [12]. Several variables affect the response of the effect of interaction between variables [13]. The advantage of using RSM is the reduced number of experiments that will be carried out to evaluate various parameters and their interactions [14].

This study aims to obtain the optimum operating condition of the microwave-assisted distillation process on clove leaves and obtain the second-order polynomial equation with Response Surface Methodology (RSM).

2. Materials and Methods

2.1. Materials.

The main material used is water and clove leaf waste which has been falling and it is obtained from the Gebugan Village area, Bergas District, Semarang Regency.

2.2. Method

2.2.1. The Distillation of Essential Oils by Microwave Assisted Hydrodistillation (MAHD) Method. The clove leaves that had become powder were then weighed by a certain mass. Next, it was put into the reactor and 300 ml of water was added. The distillation equipment was arranged, the adjustment of the microwave according to certain power was also set. The next step was holding the distillate that came out from the condenser in the separating funnel and being observed for 30 minutes. Oil and water were separated using a separating funnel, the water was collected in an Erlenmeyer, while the oil was collected in a sample bottle. Calculating the yield of essential oils was also done by weighing the mass of the oil and weighing the mass of the sample material with an analytical balance, the calculation of the yield used the following formula:

$$\textbf{Yield}\ (\%) = \frac{\textbf{mass of the extracted essential oil}}{\textbf{initial dried mass of plant}} \times \mathbf{100}\% \tag{1}$$

2.2.2. Experimental Design Using Response Surface Methodology. The optimization using Response Surface Methodology was based on Central Composite Design (CCD) was used to design experiments and to determine the effect of the independent variables used, which were microwave power (400, 600 and 800 watts) and clove leaf mass (70, 80 and 90 grams) towards the dependent variable, that was the essential oil yield. In this study, the independent variables were microwave power (x1) and clove leaf mass (x2), as well as the dependent variable was the yield of essential oil in which were described in the form of a second order polynomial equation [15], i.e.

$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_{11} {x_1}^2 + \beta_{22} {x_2}^2 + \beta_{12} x_1 x_2 + \varepsilon \tag{2}$$

where x_1 represented the microwave power, x_2 represented the mass of clove leaves and y represened the yield of essential oil, β_0, β_1, β_{11}, β_{22}, β_{12} were the regression coefficients and ε was the residual component (error).

2.2.3. Statistical Analysis. The resulting data from the distillation of clove leaf essential oil was analyzed using Statistica 10 software. The software could display response graphs in 3-dimensional graphics. The evaluation of the significance of the influence of the variables used during the experiment was statistically defined at (p) <0.05. The validity of developing model equations was verified by experimental research.

3. Results and Discussion

3.1. Developing Model for the Essential Oil Yield Prediction.

The optimization method used is Response Surface Methodology (RSM) which is based on Central Composite Design (CCD). The encoding values for the independent variables are presented in table 1 and the resulting experimental design is presented in table 2.

Table 1. The Code Level and The Experiment Value

Free Variables	Level Code				
	-α	-1	0	1	+α
Microwave Power	317	400	600	800	882
Clove Leaf Mass	65.86	70	80	90	94.14

Table 2. Experimental Design and Experiment Results

Run	Power	Mass	Yield (%)	Prediction (%)
1	400	70	0.6857	0.7172
2	400	90	0.54	0.3094
3	800	70	0.88	0.9827
4	800	90	1.19	1.0306
5	317	80	0	0.1141
6	882	80	0.8	0.8135
7	600	65.86	1.305	1.1835
8	600	94.14	0.68	0.9292
9 (C)	600	80	0.8375	0.8275
10 (C)	600	80	0.7875	0.8275
11 (C)	600	80	0.8125	0.8275
12 (C)	600	80	0.825	0.8275
13 (C)	600	80	0.875	0.8275

The results of the second order polynomial equation analysis to predict the yield of essential oils are described in the following equation:

$$y = 0.82750 + 0.49334X_1 - 0.17989X_2 - 0.36390X_1^2 + 0.22897X_2^2 + 0.22787X_1X_2 \quad (3)$$

In table 3, the coefficient from the parameter is used to obtain the second order polynomial equation. The parameter that has a value of $p < 0.05$, then the significant parameter influences the oil yield. Based on table 3, that the two variables which are microwave power (X_1) and clove leaf mass (X_2) have a significant effect on oil yield because a value of $p < 0.05$ is obtained. The R^2 value is obtained $R^2 = 0.84028$, which means that 84.028% of the total variations is corresponding to the model. The comparison graph between observations and predictions is presented in figure 1.

Table 3. The prediction of the estimated effects for polynomial equations.

Factor	Effect	Std Err.	T	p
Mean/Interc.	0.827502	0.014470	57.1883	0.000001
X_1	0.493345	0.022893	21.5503	0.000027
X_1^2	-0.363901	0.024579	-14.8053	0.000121
X_2	-0.179890	0.022880	-7.8621	0.001414
X_2^2	0.228974	0.024535	9.3313	0.000734
X_1X_2	0.227850	0.032355	7.0421	0.002143
Pure Error				
R^2	0,84028			

3.2. The Influence of Variables on Yield of Essential Oils.

The results of the influence of independent variables which are microwave power and mass of clove leaves on the yield of essential oils that have been analyzed using the response surface methodology (RSM) are presented in figure 2. The power of microwave is one of the factors that influence the efficiency of extraction using MAHD because microwave energy will significantly affect molecular interactions between one compound and another [11]. The results show reaching the optimum point. Oil yield tends to increase along with the increase of microwave power, but it tends to decrease with the increase of the leaf mass, so the variable that greatly influences the yield is microwave power. The higher the microwave power the more significantly affects the results. At high power, it will produce high temperatures which will speed up the process and the yield will rise [16].

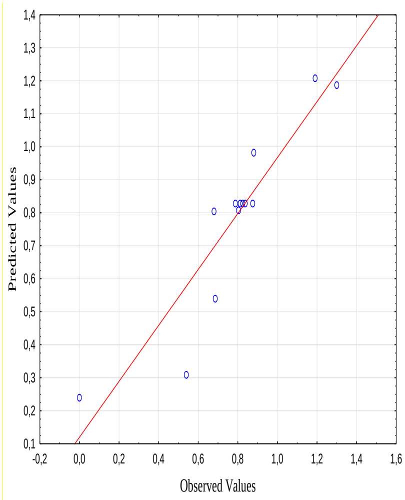

Figure 1. The relationship between the predicted value and the observed value in the oil yield.

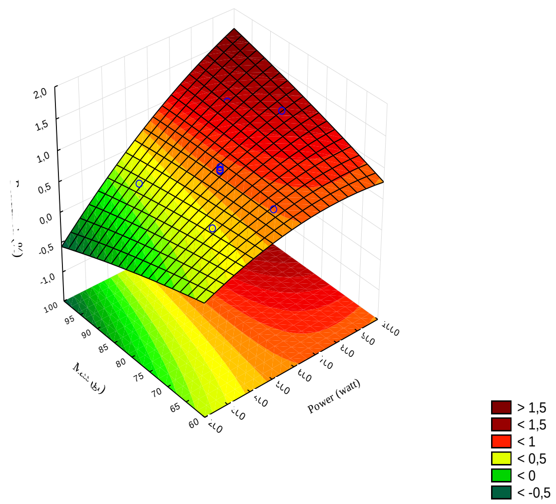

Figure 2. The response surface of essential oils is affected by microwave power and clove leaf mass.

The surface results that have been analyzed using the response surface method predict the optimal value of the yield of essential oils of 1.1834% with the optimal value of the independent variable that is microwave power of 599.5 watts and leaf mass of 65.86 grams.

4. Conclusion

The optimization of the distillation process of clove leaf essential oil by microwave assisted hydro distillation method using Response Surface Methodology (RSM) is based on Central Composite Design (CCD). The results show that microwave power and clove leaf mass significantly affect the yield of essential oils. The optimum operating condition for distillation of essential oil in 30 minutes reaches 599.5 watts of microwave power and 65.86 grams of leaf mass and the second order polynomial equation obtained is $y = 0{,}827502 + 0{,}493345X_1 - 0{,}179890X_2 - 0{,}363901X_1^2 + 0{,}228974X_2^2 + 0{,}227850X_1X_2$. The second order polynomial equation is obtained to predict percentage conversion. Trial validation is conducted to check the availability and accuracy of the model. The results show that the predicted value has been verified according to the experimental value.

References

[1] Kamatou G P, Vermaak I and Viljoen A M 2012 *Mol.* **17** 6953–6981
[2] Supriatna A, Rambitan U N, Sumangat D and Nurdjannah N, 2004 *Bul. TRO* **15** 1-18
[3] Direktorat Jenderal Perkebunan 2016 *Statistik Perkebunan Indonesia Komoditas Cengkeh 2015-2017* (Jakarta : Sekretariat Direktorat Jenderal Perkebunan, Kementerian Pertanian) 1-40
[4] Nurdjannah N 2004 *Diversifikasi Penggunaan Cengkeh* (Bogor: Balai Besar Penelitian and Pengembangan Pasca Panen Pertanian. Perspektif) **3** 61-70
[5] Gonzalez-Rivera J, Duce C, Falconieri D, Ferrari C, Ghezzi L, Piras A and Tine M R 2015 *Inv. Food Sci. Em. Tech.* 1-34
[6] Wenqiang G, Shufen L, Ruixiang Y, Shaokun T and Can Q 2007 *Food Chem* **101** 1558–1564
[7] Pratama I B 2012 *Metode Pengambilan Minyak Atsiri dalam Rimpang Kencur (Kaempferia Galanga L.) menggunakan Ekstraksi Gelombang Mikro* (Tugas Akhir Fakultas Teknik Universitas Diponegoro)
[8] Bhuiyan N, Begum J, Nandi N C and Akter F 2010 *Afr J Pharm Pharmacol* **4** 451–454
[9] Andayani T, Yufuf H and Rini Y 2014 *JBKT **2** 123-130*
[10] *Fong O H* 2012 *Extraction Of Essential Oil From Orange Peels* (Thesis Faculty of Chemical & Natural Resources Engineering University Malaysia Pahang)
[11] Mathialagan R, Nour A H, Sulaiman Z A, Nour A H and S T R 2014 *Int. J. Chem. Eng. App.* **5** 104–108
[12] Bas D and Boyaci I H 2007 *J. Food Eng.* **78** 836–845
[13] Montgomery D C 1997 *Design and Analysis of Experiments*. (New York: 4th edition John Wilwy & Sons)
[14] Liu Y, Wei S and Liao M 2013 *Ind. Crops Prod.* **49** 837–843
[15] Nugroho A D, Sianto M E and Asrini L J 2017 *JIWT* **16** 97-104
[16] Zhu C P, Zhai X C, Li L Q, Wu X X, and Li B 2015 *Food Chem.* **177** 139–146