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Optimization of Essential Oil Extraction of Beluntas *(Pluchea Indica L.)* Leaves by Using *Solvent-Free Microwave Extraction*

Nur Karima[1], Nova Chintya Kurniawati[1], Boy Arief Fachri[1], Istiqomah Rahmawati[1], Bekti Palupi[1], Mahfud Mahfud[2], Ditta Kharisma Yolanda Putri [1], Atiqa Rahmawati[3], Badril Azhar[4], Maktum Muharja[1,*]

[1]Department of Chemical Engineering, Faculty of Engineering, University of Jember, Jalan Kalimantan 37, 68121 Jember, Indonesia
[2]Department of Chemical Engineering, Sepuluh Nopember Institute of Technology, Sukolilo Surabaya East Java 60111 Indonesia
[3]Department of Skin Processing Technology, Politeknik ATK Yogyakarta, Bantul 55188, Yogyakarta
[4]Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei City 10607, Taiwan

*e-mail: maktum@unej.ac.id

ABSTRAK

Beluntas *(Pluchea Indica L.)* yang biasa digunakan sebagai astringent dan antipiretik memiliki potensi yang tinggi sebagai bahan baku produksi minyak atsiri. Tujuan dari penelitian ini adalah untuk mengoptimalkan *solvent-free microwave extraction* (SFME) dari daun beluntas menggunakan *response surface metodology* (RSM). Desain Box-Behnken dengan variasi waktu ekstraksi (60-120 menit), rasio bahan/labu distilat (0,06-0,1 g/ml), dan daya pemanas (150-450 Watt) digunakan untuk mengoptimalkan produksi minyak atsiri. Faktor rasio bahan/penyuling memiliki pengaruh signifikan paling tinggi terhadap rendemen minyak atsiri (P<0,05). Rendemen minyak atsiri meningkat seiring dengan meningkatnya daya pemanasan minyak dan waktu ekstraksi, dan sebaliknya. Di sisi lain, peningkatan rasio bahan/labu distilat memberikan dampak negatif terhadap rendemen minyak atsiri. Hasil minyak atsiri maksimum menggunakan metode SFME sebesar 0,2728 b/b% diperoleh untuk kondisi optimal waktu ekstraksi 90 menit, daya pemanasan 450 W, dan rasio bahan/labu distilat 0,06.

Kata kunci: Desain Box-Behnken, minyak atsiri, *Pluchea Indica L.*, response surface metodology, solvent-free microwave extraction

ABSTRACT

Beluntas *(Pluchea Indica L.)* which commonly used as astringent and antipyretic has a high potential for the feedstock of essential oil production. The objective of this work is to optimize solvent-free microwave extraction (SFME) of Beluntas leaves by using response surface methodology (RSM). Box-Behnken Design with the variations of extraction time (60-120 min), feed/distiller ratio (0.06-0.1 g/ml), and heating power (150-450 W) was utilized to optimize essential oil yield. The feed/distiller ratio factor had the highest significant effect on the essential oil yield (P<0.05). Essential oil yield increased as the increase of oil heating power and time extraction, and vice versa. On the other hand, the increase in the feed/distiller ratio gave a negative impact on the essential oil yield. The maximum essential oil yield using SFME method of 0.2728 b/b% was obtained for the optimized condition of extraction time of 90 min, microwave heating power of 450 W, and feed/distiller ratio of 0.06.

***Keywords*:** Box-Behnken design, essential oil, *Pluchea Indica L.*, response surface methodology, solvent-free microwave extraction

1. INTRODUCTION

Beluntas *(Pluchea indica L.)* is one of Indonesia's native plants that is widespread in several regions in Indonesia. Beluntas belongs to the family Asteraceae which has been used as food and traditional medicine. Beluntas generally grows wild in dry areas on hard and rocky soil or planted as hedges [1],

Corresponding author: Maktum Muharja
Department of Chemical Engineering, Universitas Jember
Jalan Kalimantan 37, 68121 Jember, Indonesia
E-mail: maktum@unej.ac.id

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[2]. Beluntas leaves are essential oil-producing plants that have not been maximally utilized in Indonesia. Beluntas leaves contain alkaloids, flavonoids, tannins, sodium, potassium, aluminum, calcium, magnesium, phosphorus, and essential oils. Beluntas leaves have an aromatic aroma and taste bitter which is efficacious to increase appetite (stomatic), fever-lowering (antipyretic), sweating (diaphoretic), refresher, tuberculosis glands, pain in rheumatism and vaginal discharge [3,4].

Several extraction methods to extract essential oils have been carried out. One of them is solvent extraction where the Soxhlet extraction method is generally used. This method is proven to be able to extract the contents of a material to the maximum [5], [6]. However, this soxhlet extraction method has several disadvantages including the need for organic solvents to extract essential oil content thereby increasing the cost of oil production. In addition, it takes a long time to extract the entire essential oil content, making it less efficient [7]. Therefore, the latest extraction method was developed using a distillation system by utilizing microwave heating known as Microwave-Assisted Extraction [8]. This method was further developed into four methods namely Microwave Hydrodistillation, Microwave Steam Distillation, Microwave Steam Diffusion and Solvent-Free Microwave Extraction [9, 10].

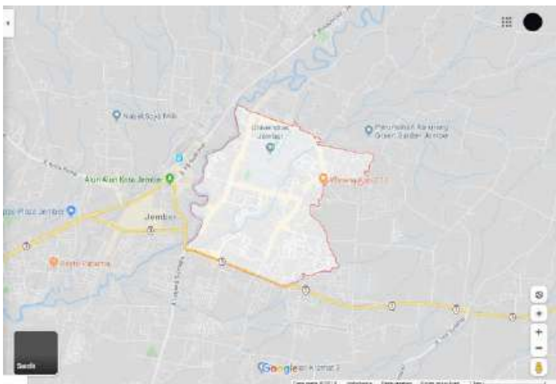
Microwave oven extraction is strongly influenced by solvents. Solvents that are able to absorb microwaves are solvents which have high dielectric coefficient and dissipation factors [11] . Several studies on the extraction of bioactive compounds by microwave ovens have been carried out using water solvents that are able to absorb microwaves. The use of water solvents replaces organic solvents which have negative effects on the environment and health because they are toxic and non-renewable. Water can be used as a solvent for extraction because water has a dielectric coefficient of 78.3 so that it can absorb microwaves [12, 13]. The advantage of water as a solvent for extraction is the availability of abundant, non-toxic and will minimize the use of organic solvents. Therefore this study uses the Solvent-Free Microwave Extraction (SFME) method. Although having great potential, from the literature studies, there is no previous report about the utilization of SFME for essential oil extraction from *Pluchea Indica L*.

This study aims to determine the optimum conditions for extracting Beluntas using SFME. The optimum extraction conditions include power, time, and the ratio of raw material: distillate volume. This study also aims to determine the power, time and ratio of the material to the volume of the distiller to the yield obtained from extraction using the SFME method.

2. RESEARCH METHODS

2.1. STUDY AREA

The material used in this study was dried Beluntas (Pluchea indica Less) leaves obtained from Sumbersari sub-district, Jember district, Indonesia and Maesan sub-district, Bondowoso district, Indonesia as shown in Figure 1a and 1b. To bind oil from extracted water using n-hexane. N-hexane used is technical n-hexane.



(a)

Figure 1. a) Location of Beluntas leave is in Sumbersari, Jember, Jawa Timur, $6^0$27’29” s/d $7^0$14’35” East Longitude and $7^0$59’6” s/d $8^0$33’56” South Latitude.

(b)

Figure 1. b) Location of Beluntas leaves is in Pujer Baru, Maesan, Bondowoso, Jawa Timur 113°48'10" - 113°48'26" East Longitude and between 7°50'10" - 7°56'41" South Latitude.

2.2. PROCEDURES

2.2.1 PRETREATMENT

Beluntas leaves were dried and weighed 60, 80, 100 g. The dried leaves were soaked with water for about 30 minutes before being put into a distillation flask.

2.2.2 EXTRACTION

Beluntas leaves that have been soaked are then put into a distillation flask to be extracted. In carrying out this solvent-free microwave extraction method, the microwave is modified by perforating the top part. A round bottom flask with a capacity of 1000 mL is placed in the microwave and connected to the Clevenger condenser through the hole. The procedure for the solvent-free microwave extraction method is carried out at atmospheric pressure with a ratio between the mass of the raw material to the volume of the distiller that is 0.06; 0.08; and 0.1 g/mL. Beluntas oil extraction is carried out at 150 W, 300 W, and 450 W power with time-variable 1; 1,5; and 2 h.

2.2.3 PURIFICATION

Since the extraction results obtained are quite small, the separation process between Beluntas oil and water is added, N-Hexane is added which functions to bind Beluntas oil. After that, N-Hexane is separated from Beluntas oil using a rotary evaporator.

2.3. DATA ANALYSIS

Beluntas oil yield is obtained from the calculation of the mass of essential oil divided by the mass of raw material multiplied by one hundred percent [14], as following Equation (1). ANOVA was utilized to analyze the effect of linear, quadratic, and interaction variables on the SFME of Beluntas. RSM and data analysis were carried out with a commercial statistical package, Minitab 16 statistical software (Minitab Inc., ITS Surabaya, Indonesia). Table 1 shows the run order, variable conditions, and experimental data of Box-Behnken Design (BBD) which was consisted of 17 experiments including 5 repetitions on the center point and was run randomly [15, 16].

$$Yield = \frac{The\ obtained\ oil\ (g)}{The\ initial\ beluntas\ (g)} x\ 100\% \quad (1)$$

Table 1. Box-Behnken design and the obtained oil yield

Run	Time (h)	F/D Ratio (g/mL)	Power (W)	Yield (%)
1	1.5	0.08	300	0.2170
2	2	0.08	450	0.1350
3	1.5	0.08	300	0.1517
4	1.5	0.1	150	0.1552
5	1.5	0.06	450	0.2728
6	2	0.08	150	0.1778
7	1	0.08	150	0.1736
8	1.5	0.08	300	0.1779
9	1	0.08	450	0.1333
10	2	0.06	300	0.2182
11	2	0.1	300	0.0245
12	1	0.1	300	0.1479
13	1	0.06	300	0.1364
14	1.5	0.08	300	0.0604
15	1.5	0.1	450	0.1798
16	1.5	0.06	150	0.2656
17	1.5	0.08	300	0.0797

3. RESULTS AND DISCUSSION

Based on Table 1, there are 5 repetitions on the center point that have different result. Differences can be caused by inaccurate weighing of materials and differences in material conditions. The results were analyzed using Analysis of Variance (ANOVA). Table 1 presents the experimental parameters produced by BBD design. To determine the factors effects in the model are statistically significant, the p-values less than 0.05 indicate model terms are significant, whereas the lack of fit was not significant at the 0.05 [17]. The insignificant Lack of fit value is requirement for a good model because it shows the suitability response data with the model [18]. The quadratic model described by the following as in equation:

$$Yield = 0.132 + 0.818\ Time - 6.7\ Ratio - 0.00154\ Power - 0.132\ Time*Time + 72.3\ Ratio*Ratio + 0.000002\ Power*Power - 5.13\ Time*Ratio - 0.000008\ Time*Power + 0.00145\ Ratio*Power \quad (2)$$

Table 2. Analysis of variance (ANOVA) obtained oil yield.

Source	DF	Adj SS	Adj MS	F-Value	P-Value	
Model	9	0.049149	0.005461	1.97	0.192	Not significant
Linear	3	0.019074	0.006358	2.29	0.165	
Time	1	0.000159	0.000159	0.06	0.818	
Ratio	1	0.018586	0.018586	6.70	0.036	
Power	1	0.000329	0.000329	0.12	0.741	
Square	3	0.019471	0.006490	2.34	0.160	
Time*Time	1	0.005014	0.005014	1.81	0.221	
Ratio*Ratio	1	0.003521	0.003521	1.27	0.297	
Power*Power	1	0.011426	0.011426	4.12	0.082	
2-Way Interaction	3	0.010604	0.003535	1.27	0.355	
Time*Ratio	1	0.010527	0.010527	3.79	0.093	
Time*Power	1	0.000002	0.000002	0.00	0.982	
Ratio*Power	1	0.000076	0.000076	0.03	0.873	
Error	7	0.019428	0.002775			
Lack-of-Fit	3	0.001988	0.000663	0.15	0.923	Not significant
Pure Error	4	0.017439	0.004360			
Total	16	0.068577				

The results in Table 2. displayed the Analysis of Variance results for the quadratic model of essential oils yield. Three independent factors in the ANOVA table include: time, ratio, power, square and 2-way interaction. The result show that the model are not significant, aproved by P-Value more than 0.05. Same as the model P-Value from lack of fit more than 0.05.

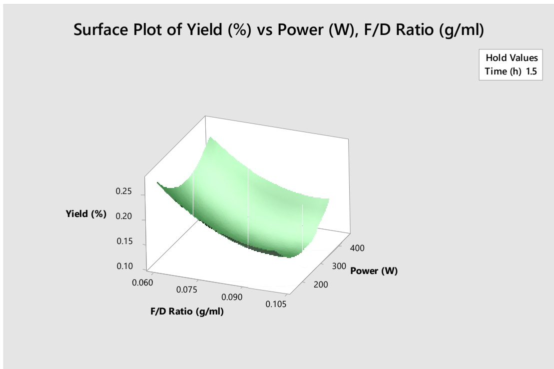


Figure 2. Surface plot of the oil yield of fixed extraction time of 1.5 h

Based on Figure 2, the yield of Beluntas oil tends to increase in yield along with the increase in power. The increase of oil yield by power may be attributed to the enhancement of penetration into the sample matrix and solubilize the analyte [19]. The thermal energy via irradiation also drove the breakdown of the structure of membranes cell in the Beluntas leaves and facilitate the analyte diffusion from the matrix surface [20]. The highest yield is obtained at a power of 450 W. Generally, it can be said that the greater the energy received by the material to be converted into heat, causing the yield of Beluntas oil to be obtained more and more. Basically, microwaves can accelerate the extraction process for the desorption of targeted compounds from low power and high power matrices Z [21].

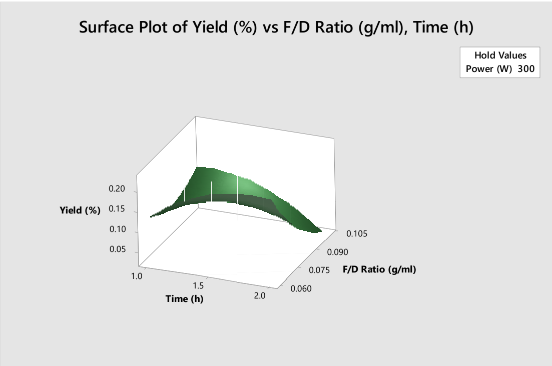


Figure 3. Surface plot of the oil yield of fixed power of 300 W

Based on Figure 3, the higher the ratio of raw material: volume distiller, the lower the yield obtained. The optimum yield for Beluntas oil extraction with the solvent-free microwave extraction method is found at an F/D ratio of 0.06 g/mL. This happens because the smallest ratio can be extracted well with a density level that is not too high. The material density factor is the ratio between the mass of the material and the volume capacity of the distiller flask used. The ratio used is related to how dense (the amount of) raw material entered in the distiller flask, so that the process of extracting and evaporation of oil can run perfectly. The material density that is too high and uneven can cause the formation of "rat holes" steam pathways which can reduce the yield and quality of essential oils. This was in agreement from previous studies by Kusuma et al. [22] on patchouli oil extraction by the solvent-free microwave extraction method, where the highest yield is also obtained at the lowest ratio.

The effect of the extraction time on the Beluntas oil yield was presented in Figure 4. As shown in Figure 4, at low heating power (< 300 W), oil yield increased significantly and reached maximum value during 1.5 h extraction time. Furthermore, extending the duration of extraction gave a negative effect on the obtained oil yield. The same trend of the yield was also shown on the high heating power. The lowest oil yield was gained on the power of 300 W at all reaction times. The oil yield decreased significantly which might be attributed to the possible degradation as the longer extraction time [23]. Volatile components in the oil yield might also be evaporated, leading to a decrease in the extraction yield of oil as reported in the literature [24]. Jesus et al. [25] reported that the longer time of extraction leads to the degradation of the essential oil.

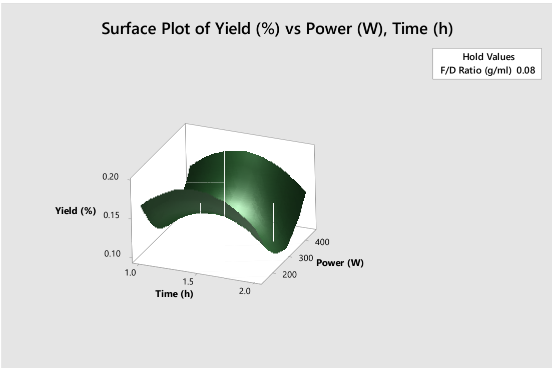


Figure 4. Surface plot of the oil yield of fixed F/D Ratio of 0.08 g/ml

On the other hand, this SFME method has lower production cost than another method. In this study, only required material and electricity costs for production cost with total cost 6.26 US$/kg. Compared with production cost for steam distillation, water distillation, solvent extraction and supercritical fluid extraction varies from 15.85-76.50 US$/kg, 7.05-86.4 US$/kg, 8.35-8.53 US$/kg and 6.71-42.69 US$/kg, respectively [26].

4. CONCLUSION

This report shows that the extraction of beluntas essential oil using SFME method was affected by power, time, and ratio distiller volume. The optimum conditions in Beluntas oil extraction are at the highest power that is 450 W, the ratio between the mass of raw material with the lowest distiller volume is 0.06 g/ mL, and the longest extraction time is 1.5 h.

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