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The Optimization Ratio and Size of Raw Materials in Patchouli Oil Extraction Process Using Hydrodistillation Method with Addition of $MgCl_2$

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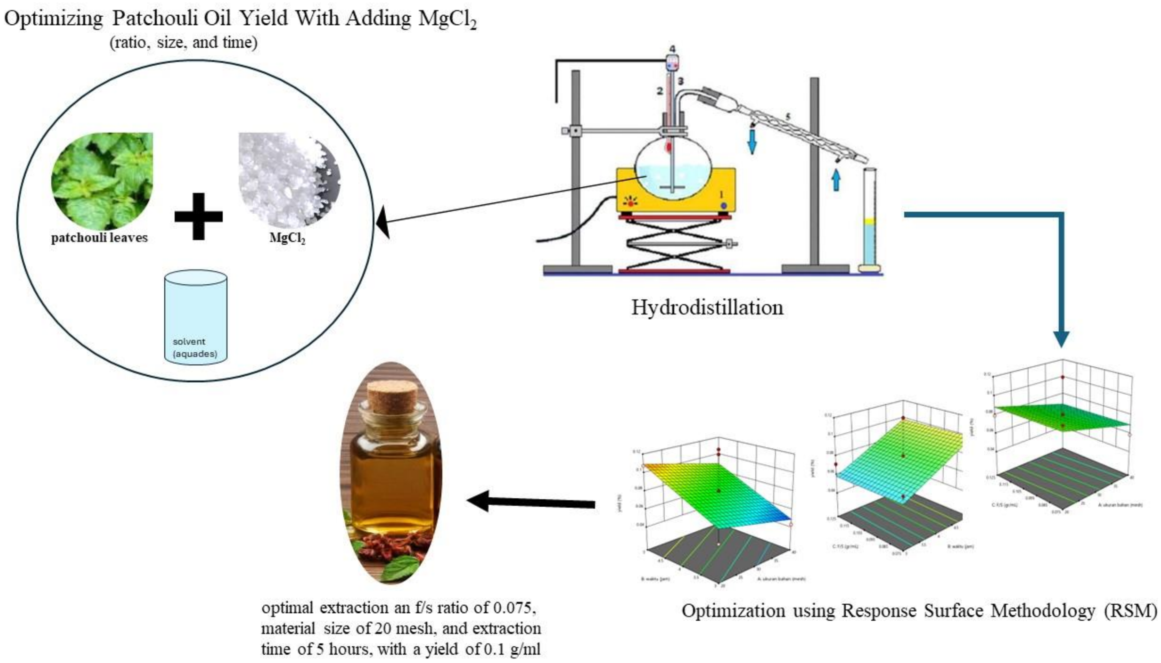
GRAPHICAL ABSTRACT



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ABSTRACT

This research focuses on optimizing patchouli oil yield using the hydrodistillation method and adding $MgCl_2$. The RSM method is used to analyze model variance, including the sum of squares model, less of the fit model, and ANOVA analysis. The research results show that adding $MgCl_2$ can increase the yield of patchouli oil by reducing water content and undesirable non-volatile elements, thereby producing more stable patchouli oil. ANOVA analysis of patchouli oil yield was conducted to determine the optimal results from factors such as f/s, material size, and time. The extraction time factor had a p-value of less than 0.05, indicating that the model was suitable for explaining the results. The research results showed that the yield of patchouli oil was optimal with an f/s ratio of 0.075, material size of 20 mesh, and extraction time of 5 hours, with a yield of 0.1 g/ml being the best choice. The research results showed that using specific methods, namely distilled water and $MgCl_2$ in the extraction process, effectively increased the yield of patchouli oil.

1. INTRODUCTION

Essential oils are oils that come from plants. Essential oils are also known as ethereal or volatile oils, usually produced by plants. Components that quickly evaporate make essential oils, commonly known as flying oils. Indonesia has various plants that have great potential to be processed into essential oils [1]. Essential oils have multiple benefits, including fragrances (in cosmetics and body care products), aromatherapy oils, rubbing oils (for colds, body warmers, and carminatives), room fresheners, and insect repellents. However, only a portion of this amount is used as a commercial source of essential oils, some of which come from plantation commodities, namely patchouli and citronella [2]. Patchouli (*Progestemon cablin*) belongs to the labiate family of plants. Patchouli plants were cultivated in Aceh and then expanded to several provinces in Indonesia. Patchouli essential oil is obtained by steam distillation or hydrodistillation of dried leaves of Pogostemon cablin Benth. Patchouli oil has a unique smell and is necessary to replace the constituents of fragrances and cosmetics. Indonesia is one of the producers of patchouli oil. Around 90% of global production of 1200-1300 metric tons per year is realized in Indonesia [3]. Considering the commercial demands for cosmetics, aromatherapy and its therapeutic potential, patchouli oil is still necessary. One of the processes for making essential oils is to use the distillation technique. Distillation separates components contained in plants or other materials based on the vapor point of two or more components [4].

In general, there are three types of distillation techniques: water (hydrodistillation), wet steam (steam and water distillation), and dry steam. The method often used is distillation using water (hydrodistillation) because it is the simplest method. Hydrodistillation is a method commonly used in the chemical industry, where the working principle is to separate high boiling points or thermally unstable components. The distillation technique using water (hydrodistillation) has several problems, such as long reaction times, high energy consumption, and low yields. Based on this problem, a salting out method was carried out: adding certain inorganic salts to the system to improve the essential oil distillation process. It has been shown in various literature that the addition of lithium salts to the hydrodistillation technique is effective in increasing the yield of essential oils [5]. So, in this research, patchouli oil will be extracted using $MgCl_2$ salt. $MgCl_2$ salt is added to cause a salting out effect, namely adding certain inorganic salts to the system to improve the essential oil distillation process.

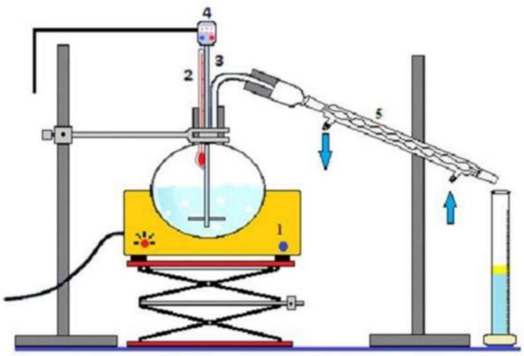
Hydrodistillation is the best method for producing essential oil from patchouli because it is more stable in producing patchouli alcohol [6]. Patchouli extraction has several problems: long reaction time, high energy consumption, and low yield. The extraction process must also maintain the essential components in patchouli essential oil. Based on GCMS analysis, ten dominant component compounds make up patchouli essential oil, namely Patchouli Alcohol (39.46%), Delta-Guaiene (21.51%), Alpha Guaiene (20.13%), Alpha-patchouli (9.01%) [7]. In this study, Response Surface Methodology (RSM) will be used as one of the methods that can provide optimal conditions for the patchouli oil extraction process using hydrodistillation with a salting out system.

2. EXPERIMENTAL METHODS

Response Surface Methodology (RSM) will be used to analyze the extraction results using the hydrodistillation method with the addition of $MgCl_2$ salt. RSM can identify the perfect conditions for optimal results or find an acceptable operating range. The samples used in this research were patchouli leaves that had been air-dried and uniform in size according to analytical needs.

2.1. Extraction Of Patchouli Oil

Patchouli oil is extracted using the hydrodistillation method by adding $MgCl_2$ salt to the hydrodistillation apparatus (Figure 1). The extraction stages using the hydrodistillation method can be seen in Figure 2.



Description:
1. Induction heater (heater)
2. Thermometer
3. Stirring rod
4. Distillation adapter
5. Condenser

Figure 1. Schematic apparatus of hydrodistillation

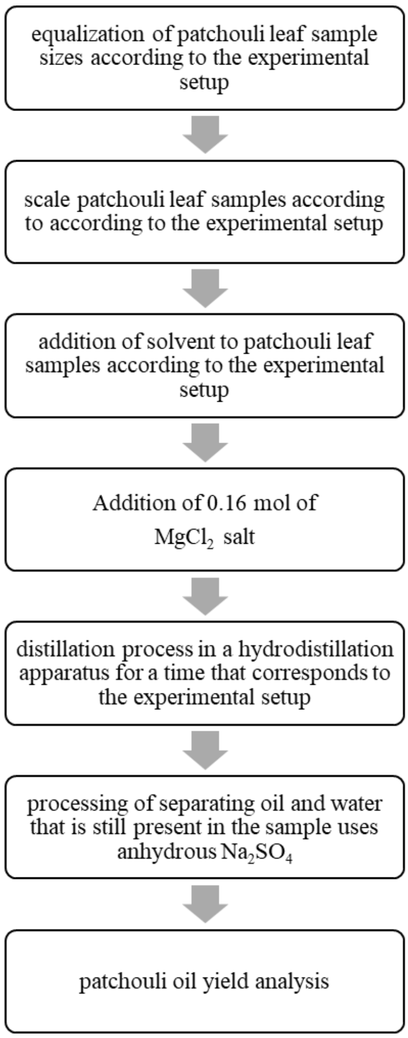


Figure 2. Hydrodistillation process flow diagram

2.2. Experimental Setup For Factorial Analysis

In this study, the response observed was a combination of the ratio of raw materials to solvent (feed to solvent), size of raw materials, and extraction time to obtain the optimal composition for extracting patchouli oil. Data processing and analysis will use statistical software Design-Expert 13.0.5.0 (Stat-Ease, Inc.) to get a model for patchouli oil purification. Experimental design with three factors where variations in feed to solvent (f/s) used was 0.04 g/ml‑0.012 g/ml; variations in raw material sizes are 20 mesh‑40 mesh; time variation 3-5 hours. At the extraction stage, after entering each factor value in the software by selecting Box Behnken Design (BBD), 15 experimental designs were obtained, as in Table 1. In the extraction process, 0.16 mol $MgCl_2$ salt is added, and anhydrous Na_2SO_4 is added, which is used to bind (absorb) the water content that is still contained in the essential oil, then accommodate the oil in a sample bottle and close it. The essential oil yield was calculated using Equation 1 [8].

$$Yield\ (\%) = \frac{Massa\ minya\ (gram)}{massa\ sampel\ (gram)\ x\ (1 - kadar\ air (\%))}\ x\ 100\% \quad (1)$$

2.3. Validation Experiment

Validation experiments were carried out to validate the optimal conditions suggested by the Design-Expert software. Criteria for selecting optimal conditions include f/s, raw material size, and time, with all responses being in maximum conditions. An optimal experiment is a factor condition that can produce the highest yield response value.

3. RESULTS AND DISCUSSIONS

The essential oil in patchouli leaves is surrounded by oil glands, vessels, oil sacs, or glandular hairs. If patchouli leaves are left intact, the essential oil can only be extracted if water vapor makes its way through the tissue of the plant material and forces it to the surface. Therefore, the treatment of materials is also included in the experimental variables. In addition, a larger plant surface area can cause the evaporation rate of essential oils from plant material to be faster [9]. In this study, samples of dried patchouli leaves were chopped to a size of 20-40 mesh. According to the existing experimental design, the sample was added with distilled water as a solvent with an f/s ratio. Apart from the distilled water solvent, in the extraction process, 0.16 mol of $MgCl_2$ salt was added, and then the extraction time was adjusted based on the experimental design that had been made. The patchouli oil obtained was then analyzed for its yield value. Based on Table 1, the yield value obtained is 0.043%‑0.120%.

TABLE I. Results of yield analysis *patchouli oil extraction* using the RSM method

No.	Factor 1 A: material size (mesh)	Factor 2 B: time (hours)	Factor 3 C: f/s (g/ml)	Response 1 yield (%)
1	20	4	0.075	0.096
2	20	4	0.125	0.080
3	20	3	0.1	0.054
4	20	5	0.1	0.108
5	30	3	0.075	0.068
6	30	3	0.125	0.071
7	30	5	0.075	0.083
8	30	5	0.125	0.100
9	30	4	0.1	0.080
10	30	4	0.1	0.120
11	30	4	0.1	0.073

12	40	4	0.075	0.059
13	40	4	0.125	0.047
14	40	3	0.1	0.043
15	40	5	0.1	0.106

Table 2 shows the difference in obtaining the yield of patchouli oil using hydrodistillation with the addition of $MgCl_2$ salt and without the addition of $MgCl_2$ salt. Adding $MgCl_2$ salt can improve the quality of essential oils by reducing water content and other undesirable compounds, resulting in purer and higher-quality essential oils. $MgCl_2$ salt has good oxidation resistance, which can help maintain the quality of essential oils stable in the extraction process using the hydro distillation method. The presence of $MgCl_2$ salt helps reduce the viscosity of crucial oils, which is vital in the processing and applying essential oils so that the processing and application process is easier and more efficient [10]. ow-viscosity essential oils have more mobile molecules, so they can quickly reach the surface and evaporate into the air. On the other hand, oil with a high viscosity will evaporate more slowly because of obstacles in its movement. In addition, viscosity can affect the stability and quality of essential oils during storage. Thicker essential oils may be more stable and less susceptible to evaporation [6].

TABLE II. The yield of patchouli oil using hydrodistillation

No.	Factor 1 A: material size (mesh)	Factor 2 B: time (hours)	Factor 3 C: f/s (g/ml)	With $MgCl_2$ yield (%)	Without $MgCl_2$ yield (%)
1	20	4	0,075	0,096	0,061
2	20	4	0,125	0,079	0,072
3	20	5	0,1	0,108	0,072
4	30	3	0,075	0,068	0,026
5	30	5	0,125	0,100	0,049
6	30	4	0,1	0,120	0,060
7	40	4	0,075	0,059	0,031
8	40	5	0,1	0,106	0,087

Salt separates essential oil components through several mechanisms, namely controlling oxidation, water content, and non-essential compounds. In controlling oxidation, $MgCl_2$ salt has good oxidation resistance, which can help reduce the oxidation of essential oils during the extraction and storage processes. These are necessary steps to ensure the quality of the essential oil remains stable. In controlling water content, $MgCl_2$ salt can reduce the water content in essential oils, which improves quality and reduces the risk of damage during storage. $MgCl_2$ salt can control the levels of undesirable non-essential compounds in essential oils so that essential oils are purer and of higher quality [11].

3.1 Model Variance Analysis (ANOVA)

A statistical model analysis of the yield response of patchouli oil was carried out to determine and evaluate the type of model that best suited the data in the research. Optimization in this research aims to determine the optimum yield point from factors (f/s, material size, and time) to the response (yield). In the RSM method, there are several criteria used in selecting a statistical model, such as the sum of squares of the model sequence (Sequential Model Sum of Squares), testing model mismatch (Lack of Fit), and analysis of variation (ANOVA).

Based on the results of the values from the analysis of variation (ANOVA) in Table 3, it can be seen that the model and factor B show a significant value of <0.05. This proves that factor B has a real influence on the yield response. The Lack of Fit value for the response p-value of 0.0022 indicates that $p < 0.05$ with no significant status is by the trend or plot used in the Quadratic model. The main criterion for assessing model suitability is the Lack of Fit test. This is because a model is considered suitable if the test results show a model mismatch. Therefore, model mismatch data that shows insignificant results is considered adequate to explain the problem. Conversely, if the model mismatch shows significant results, then the resulting data is considered inadequate for explaining the problem [12].

TABLE III. ANOVA Analysis for Patchauli Oil Yield

Source	Sum of Squares	df	Mean Square	F-value	p-value	
Model	0.0040	3	0.0013	4.31	0.0307	significant
A-size material	0.0008	1	0.0008	2.70	0.1286	
B-time	0.0032	1	0.0032	10.20	0.0085	
C- f/s	5.823E-06	1	5.823E-06	0.0186	0.8938	
Residual	0.0034	11	0.0003			
Lack of Fit	0.0022	9	0.0002	0.3889	0.8692	not significant
Pure Error	0.0012	2	0.0006			
Total Cast	0.0075	14				

The f/s ratio, raw material size, and extraction time were used to create the surface response diagram generated by Design-Expert software. The yield value response in patchouli oil extraction was used as an index to see the effect of adding $MgCl_2$ salt in the extraction process of patchouli oil from patchouli leaves. The maximum yield value is taken as the optimization goal in testing. Figure 3 shows the patchouli oil yield response value at an f/s ratio of 0.1 g/ml. When the extraction time is a minimum of 3 hours, as the size of the material increases, the patchouli oil yield value decreases. However, when using the same material size at a maximum condition of 20 mesh, the patchouli oil yield value increases significantly as the extraction time increases.

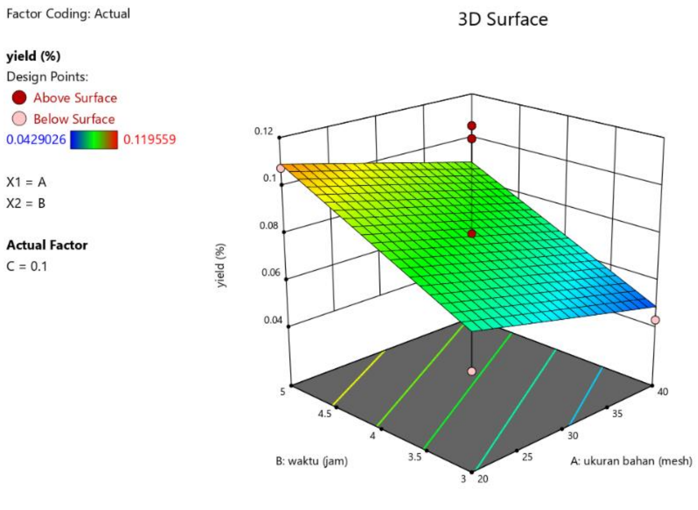
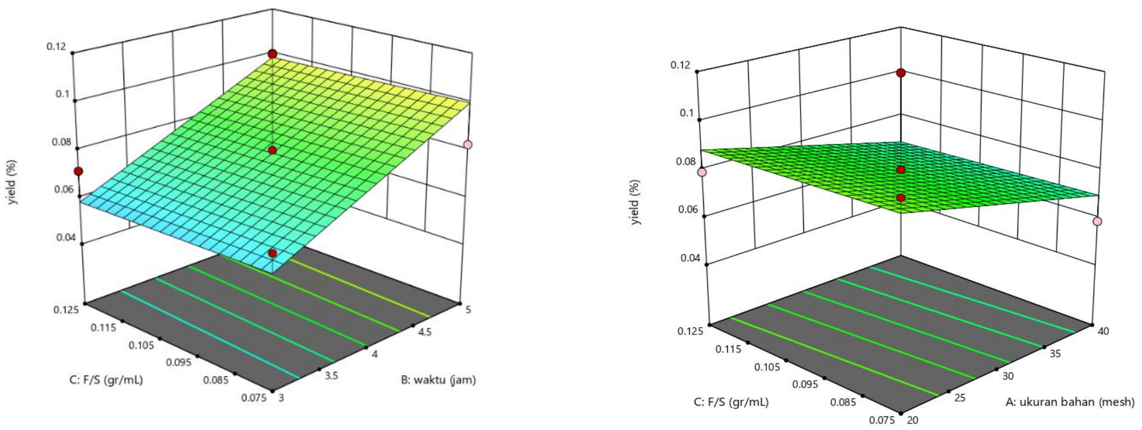


Figure 3. 3D response surface of the yield of patchouli oil using hydrodistillation

The size of the sample material and the f/s ratio in this study did not significantly affect the extraction results of patchouli oil with the addition of salt. The effectiveness of the material measure depends on the extraction method used. In maceration or reflux methods, smaller material sizes can increase extraction efficiency, whereas other methods may not have the same impact. As in extraction using microwaves, smaller material sizes can increase extraction efficiency, whereas in different methods, such as distillation, larger material sizes may be more appropriate to avoid blockages [13].

In the patchouli oil extraction process using hydrodistillation with the addition of $MgCl_2$ salt, the size of the ingredients and the f/s ratio did not significantly affect the yield value of patchouli oil (Figure 4b). However, the extraction time factor is different; the longer the extraction time, the more the yield value of patchouli oil increases significantly (Figure 4a). $MgCl_2$ salt functions to increase extraction efficiency by reducing viscosity, binding water, and assisting in the separation of essential oil components. Optimizing the effect of adding $MgCl_2$ salt will increase with the length of the extraction cycle in the hydrodistillation method. Each extraction cycle involves the process of separating essential oils from raw materials. In industrial practice, one cycle may seem more efficient regarding energy use, but multiple cycles can increase the total yield [14].



(a) (b)

Figure 4. 3D response surface of patchouli oil yield based on factors a) f/s and time b) f/s and material size

3.2. Validation Experiment

The optimization criteria for selecting optimal functional conditions using a quadratic BBD-based model is to obtain optimal patchouli oil yield values with process factors limited to experimental values. Numerical optimization, graphical optimization, and point prediction express optimal process conditions and their results. The best conditions suggested by the Design-Expert software to produce maximum patchouli oil yield for each response are f/s 0.075 g/ml, material size of 20 mesh, and extraction time of 5 hours. The validity of the model was carried out in three replicate experiments for each variable under optimum conditions. The actual average value under optimal conditions is compared with the predicted value to verify the suitability of the model [15]. Errors were calculated between predicted data and actual data on each response, where all error values were within the acceptable rule-of-thumb range, as shown in Table 4.

TABLE IV. Validation results of patchouli oil yield values using RSM

Parameter	Lowest prediction	Prediction	Highest prediction	Verification Results	difference	Accuracy (%)
Yield	0.043	0.110	0.120	0.109	0.001	99.98%

Based on Table 4, the validation results show that the yield response value is 0.109, while from the Design Expert program calculations, the yield response is 0.110. The results of this comparison show that the difference in verification predictions is smaller than 5%, which means that the verification value follows the program prediction value. The results of the comparison between the actual research value and the prediction interval (PI) value, which shows that the actual research value is still in the range of 95%, shows that it proves that the optimum formula with the highest desirability value has test results that match the predictions recommended by the program. The difference in value is not very significant [16]. The yield response verification results also show an accuracy level of 99.98. Based on this response, it produces an excellent value between the predicted value and the verification results. This shows the validity of the resulting response model is correct.

4. CONCLUSIONS

The results showed that using the hydrodistillation method with the addition of $MgCl_2$ in the extraction process effectively increased the yield of patchouli oil. ANOVA analysis of patchouli oil yield was conducted to determine the optimal results of factors such as f/s, material size, and time. The f/s ratio and material size did not show any differences in optimal yield. The results of the ANOVA analysis showed that the model and factor B (extraction time) had significant differences in results, with a p value of less than 0.05, indicating that the model was suitable to explain the results. The optimal conditions offered by RSM with an f/s ratio of 0.075 g/ml, material size of 20 mesh, and extraction time of 5 hours had optimal yield results. The results showed that the patchouli oil yield was optimal, with 0.1 g/ml yield being the best choice.

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