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Optimization of Clove Flower Oil Extraction *(Syzgium aromaticum L.)* with Factorial Design Method

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Abstract - *Solid-liquid extraction or commonly called leaching is the removal of the solute fraction (solute) from a solid to a liquid solvent. This extraction has become a widespread oil extraction process due to the high percentage of oil produced. Soxhletation method is an extraction method that contacts a solid and a liquid by dissolving it in the liquid phase at reflux. Clove oil is an essential oil that can be produced from distillation method on parts of the clove plant, especially the leaves and flowers of cloves. All parts of the clove plant contain oil, but the flowers contain the most oil. Because the leaves and twigs of cloves also produce oil, they also become a side income for clove farmers who harvest clove flowers for cigarettes. One of the most important ingredients in clove oil is eugenol, eugenol has many benefits from antiseptic to stimulant. Isopropyl alcohol is a unwell-known solvent specially to essential oil solvent, isopropyl alcohol is a solvent that can be recovered back into a solvent form usually using distillation method. The selection of isopropyl alcohol as a solvent is based on its high boiling point compared to other solvents and has elements of carbon, hydrogen, and oxygen that can dissolve polar molecules and their alkyl groups.*

Keywords- *clove oil, leaching, soxhletation, isopropyl alcohol*

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1. Introduction

Essential oil is a type of vegetable oil that is multi-purpose. The raw material for this oil is obtained from various plant parts such as leaves, flowers, fruits, seeds, seed coats, stems, roots or rhizomes. One of the main characteristics of essential oils is that they are volatile and have a distinctive aroma. World export-import statistics show that consum-ption of essential oils and their derivatives increases by around 8 - 10% from year to year [1]. Clove oil comes from the clove plant (Eugenia aromaticum) both from the flowers, stems, leaves, and stems of cloves. Cloves belong to the Myrtaceae tribe which are widely planted in several countries including Indonesia [2].

Clove oil contains the active compound eugenol which is the raw material for perfumes and various types of medicines. Cloves have a distinctive odor derived from the essential oil found in flowers (10–20%), stalks (5–10%) and leaves (1–4%) [3]. Clove flower oil resulting from steam distillation that has met the quality requirements of SNI 06-2387-2006 means that the clove flower oil has met the quality standards for the essential oil trade in Indonesia [4].

A solvent is a liquid or gas that dissolves a solid, liquid or gas, resulting in a solution. Solvents are also commonly used are organic chemicals (containing carbon) which are also called organic solvents [5]. The solvent greatly affects the extraction process. The choice of solvent is generally influenced by factors such as selectivity, boiling point, solubility, inert nature, and economicals [6].

Isopropyl alcohol is a solvent that is widely used in industry. It is estimated that 50% of IPA was applied as a solvent in 1992 [7]. Generally, clove oil producers in Indonesia still use water solvents with an inefficient time span from 6 until 8 hours [8]. Considering the relatively higher price of IPA compared to other alcohol solvents, the common method is to recover IPA as solvent again by distillation. Due to its high boiling point, IPA is safer than other polar solvents. Isopropyl alcohol is an alcohol compound with a secondary type, because of the ability of its COH group to bind 2 carbons [9].

The extraction process consists of three basic steps, namely the addition of a mass of solvent, the separation of solutes to form the extract, and the separation of the extract phase [10]. In the extraction process using solvents, the number and types of compounds that enter the solvent liquid are largely determined by the type of solvent used [11]. Soxhletation is a method of extracting plants using a Soxhlet device. The tools used are boiling flask, extractor, and condenser. Samples in soxletation need to be dried before extraction. The purpose of drying is to remove the water content contained in the sample while mashing is to facilitate the dissolved any compounds in the solvent. Soxhletation is used in certain organic solvents. Extraction is carried out by sequentially using organic solvents with increasing polarity [12].

2. Methodology

2.1. Materials

The basic ingredient of Essential Clove Flower Oil in this Research is Isopropyl Alkohol, Clove flo-wer, and water. Tools that used in this research is soxhletation kit (including Soxhlet, round bottom flask, condenser, and isomantle), vacuum distillation, grinder, and screener.

2.2. Procedure

Material Preparation

Grind the dried clove flowers with a grinder until it becomes a fine powder.

Soxhletation

Assemble a soxhlet kit, by inserting a variable mass of clove flower powder which has been wrapped with filter paper and then tied with wool thread. Put the clove flower powder into the soxhlet and the isopropyl solvent according to the variable into a three-neck flask. Then, Carry out the clove flower extraction process for 10 cycles and 20 cycles with a heating temperature of ±83°C.

Vacuum Distillation

Set the vacuum distillation kit, then put the extracted clove oil into a three neck flask. Inserting a vacuum/pump pressure source into the line before the erlenmeyer. Heating to 83 C until there e no drips

Variable

By setting to this variable, variable straightly to factorial design method that using (+) and (-) symbols t-hat can be easier ways to get this methods. By choosing 2 variable fron 3 data. To get 8 trials and 5 Optimization to get complete step factorial design methods.

Table 1. Factorial Design Method

No	Solvent Volume	Extract Cycle	Clove Mass
1	(-)200	(-)10	(-)50
2	(+)400	(+)20	(+)100

Table 2. Variable To Factorial Design

Run	Interaction			
	Cycle+Mass	Cycle+Volume	Mass+Volume	VCM
1	+	+	+	-
2	-	-	+	+
3	-	+	-	+
4	+	-	-	-
5	+	-	-	+
6	-	+	-	-
7	-	-	+	-
8	+	+	+	+

3. Results

3.1. Yield

From fig .1, it can be seen that the yield of variables 1,2,3,4,5,6,7, and 8 is 13.86%; 15.61%; 15.74%; 17.93%; 18.00%; 19.13%; 19.45%; and 20.55%. The highest yield was obtained from the 8th variable with a mass of 100 grams of clove flower, the number of cycles 20 and the volume of isopropyl 400 mL which was 20.55%. Optimum variable can be detected using factorial design method by combinating all variable using interaction calculation by normal p-probability graphics.

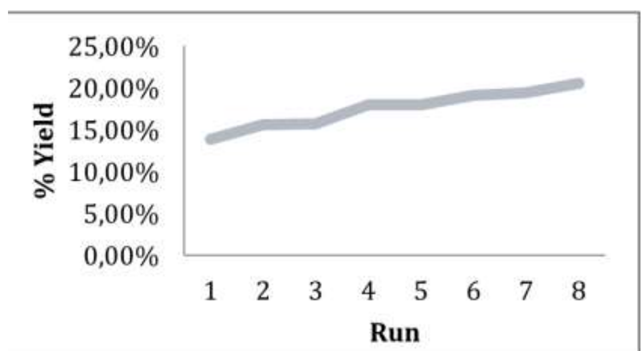

Figure 1. Yield Result Graph

3.2. Density

Table 3. shows that the density of clove flower oil variable to 3,4,5,6,7, and 8 meets SNI-06-2387-2006, which is in the range of 1.025-1.049 gr/mL. The density of clove flower oil variable 1 and 2 did not meet SNI-06-2387-2006 due to the large number of light fraction components contained in the oil. Density data obtained from calculations using the pycnometer method, where the difference in density data in Table 3 is due to the variation of the variables in each experiment carried out. On variables that are not classified as SNI, influenced by several data including mass and volume data. The in-accuracy of the data that it is not included in the SNI is a

natural thing in research which is the reference for the success value of variable optimization.

Table 3. Density Result

Run	Density (gr/mL)	SNI (gr/mL)
1	1,009	
2	1,015	
3	1,032	
4	1,028	1,025-1,049
5	1,035	
6	1,039	
7	1,042	
8	1,044	

3.3. Refractive Index

Table 4 shows that the refractive index of clove fl-ower oil variable to 1, 2, 3,4,5,6,7, and 8 meets SNI-06-2387-2006, which is in the range of 1.528-1,535. The refractive index is affected by the length of the carbon chain and the number of double bonds. The refractive index is obtained from a refractometer which uses the light refraction method uses the principle of refraction of light where light is passed through a solution.

Table 4. Refractive Index Result

Run	Refractive Index	SNI
1	1,528	
2	1,528	
3	1,529	
4	1,528	1,528-1,535
5	1,530	
6	1,532	
7	1,533	
8	1,535	

3.4. Optimization

In the optimization of isopropyl alcohol solvent volume as a variable changes with running as much as 5x, while the variable number of cycles and mass as a fixed variable. Fixed variable number of cycles is 20 cycles and mass variable is 100 grams. The data is obtained from the calculation of the main effects and iterations.

From fig 3., seen that the yield of optimization 1,2,3,4, and 5 is 19.75%; 19.84%; 20.47%; 21.04%; and 21.30%. The effect of isopropyl alcohol solvent volume between 200 mL to 300 mL has increased the yield of clove flower oil. When the volume of isopropyl alcohol solvent is increased, the yield of clove flower oil formed also increases. This happens because the ratio results in an increase in the amount of oil produced, the solubility of clove oil components is not affected by the amount of solvent. This is causes by the oil component in the raw material is limited in number and the solvent used has a limited ability to dissolve the existing material, so that with the large number of solvents in the material it cannot dissolve anymore.

At the volume of 300 mL isopropyl alcohol solvent is the best condition that produces a maximum clove oil yield of 21.30%. If the volume of isopropyl alcohol solvent is continuously increased to 400 mL, the yield of clove flower oil that is formed actually decreases. This is because the addition of the amount of solvent after the optimal point is reached, is no longer able to significantly increase the clove flower extract.

Table 5. Density Result

Optimization	Refractive Index	SNI
1	1,031	
2	1,034	
3	1,047	1,528-1,535
4	1,045	
5	1,042	

Table 5 shows that the density of clove flower oil optimization to 1,2,3,4, and 5 complies with SNI-06-2387-2006 which is in the range of 1.025-1.049 gr/mL. The effect of increasing density gives a tendency to increase the solubility of clove oil. The effect of raw materials can affect the specific gravity of clove oil, the higher the specific gravity indicates the oil has good quality.

Table 6. Refractive Index Result

Component	Rate %
Eugenol	59,16
Phenol	18,3
β-caryophillene	8,06
Cycloheptane	1,42
Allopurinol	2,23
Dimethylamino	3,27
Methyl Slicylate	1,15
⍰-caryophillene	1,06
⍰-cubebene	1,35
Thienylcyclohexene	2,02
Caryophyllene Oxide	1,98

Table 6. shows that the optimization of clove flower oil's refractive index to 1,2,3,4, and 5 meets SNI-06-2387-2006, which is in the range of 1.528-1,535. The refractive index is influenced by the length of the carbon chain and the number of double bonds. The higher the refractive index, the longer the carbon chain, and the more double bonds. Clove oil with a large refractive index value is better than clove oil with a small refractive index value. It is possible that the eugenol content contained in the oil is getting bigger. The refractive index of this optimization variable uses the same method as the non-optimized variable.

Table 7. GC-MS Result

Component	Rate %
Eugenol	59,16
Phenol	18,3
β-caryophillene	8,06
Cycloheptane	1,42
Allopurinol	2,23
Dimethylamino	3,27
Methyl Slicylate	1,15
⍰-caryophillene	1,06
⍰-cubebene	1,35
Thienylcyclohexene	2,02
Caryophyllene Oxide	1,98

In this study, the extract of clove flower oil contained 59.16% eugenol which was not in accordance with SNI-06-2387-2006 which was 78%. This is because there is eugenol that is lost during the material treatment process and extraction time.

Table 8. Comparison Result

Parameter	Research Product	SNI-06-2387-2006
Bau	Khas cengkeh	Khas cengkeh
Color	Hitam	Kuning
Density	1,047	1,025 – 1,049
R. Index	1,535	1,528-1,535
Eugenol	59,16%	Min 78%, v/v
β-caryop	8,06%	Maks 17%, v/v

Based on table 8 the odor parameters, density, refr-active index, and beta-caryophillene content have met SNI-06-2387-2006. The color parameter of the black clove flower oil extraction research is not in accordance with SNI-06-2387-2006 which states that the color of clove flower oil is yellow to dark brown, this is due to the presence of metal ions. Which then reacts with compounds in the oil, especially eugenol. The metals contained in clove leaf oil include Fe, Mg, Mn, Zn, and Pb. The metals come from leaves and distillers, metal accumulation in leaves occurs due to metal absorption from the soil through roots and metal absorption from the air through leaf stomata.

4. Conclusion

The clove flower oil extraction process consists of 3 stages, namely; pretreatment of raw materials, extraction using soxhlet, and purification of clove flower oil using vacuum distillation. The clove flower powder extracted was obtained from dried clove flowers which were ground using a grinder.

In the clove flower oil extraction process using the factorial design method with 2 levels of 3 variables, namely the number of cycles, the mass of the material, and the volume of the isopropyl alcohol solvent. Normal probability graph is the volume of isopropyl alcohol solvent of 400 mL with the resulting yield of 20.55%. Then the optimization of the process is carried out with the variable changing is the volume of isopropyl alcohol solvent with a size of 200 mL; 250 mL; 300 mL; 350 mL; and 400 mL, the fixed variables are the number of cycles of 20 and the mass of the material of 100 grams.

From the results of the optimization of the process carried out, the most optimum result to produce the maximum yield is the volume of 300 mL isopropyl alcohol solvent of 21.30%. The results of the optimization process have met SNI-06-2387-2006 on odor parameters, density, refractive index, and -caryophillene content, while the color parameters and eugenol content have not met SNI-06-2387-2006. The level of eugenol from the optimization of the clove flower oil extraction process was 59.16%.

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