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

Muh Asror Muwaffaq*, Edy Supriyo

Industrial Chemical Engineering, Vocational School, Diponegoro University, Jalan Prof. Sudarto, Tembalang, Semarang, Central Java 50275

*email: asrormuwaffaq24@gmail.com

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 an 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

1. Introduction

Essential oil is a type of vegetable oil that is multipurpose. 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 consumption 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 condensor. 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-ower, 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

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 | 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 pycnometr 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 inaccuracy 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 flower oil variable to 1, 2, 3, 4, 5, 6, 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 5 times, 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 caused 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. 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   |
| β-caryophillene    | 1,06   |
| β-cubebe           | 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.
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.

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6. References

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