Enhancement of Quality of Nutmeg Oil Using Rotary Vaccumm Evaporator

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Abstract. One business opportunity in terms of increasing the economic value of nutmeg is the content of essential oils in nutmeg. The nutmeg oil producing area in Aceh province is centralized in South Aceh Region. Nutmeg oil has a distinctive aroma and high oil yield. The quality of nutmeg oil is determined by the content of myristicin which gives a distinctive aroma to nutmeg oil [1]. In terms of improving the quality of nutmeg oil, purification process are carried out with a rotary vaccumm evaporator in order to isolate myristicin which is an active compound of nutmeg oil. The isolation process to purify nutmeg oil was carried out at an operating pressure of 300 mbar and temperature of 450 °C was increase myristicin content up to 53.41%. Test results of characteristics such as specific gravity, refractive index have meet the requirement of the Indonesian National Standard (SNI) 06-2388-2006.

1. Introduction

One business opportunity in terms of increasing the economic value of nutmeg is the content of essential oils in nutmeg. Nutmeg essential oils produce from nutmeg seeds and mace are widely used as aromatherapy, traditional medicine and in the perfume industry [2]. Furthermore, essential oils from several types of plants have antibacterial and antifungal agent so that they can be used as preservatives in food and as natural antibiotics [3]. Nutmeg has high antioxidants due to the presence of phenolic compounds, especially in nutmeg seeds [1]. Nutmeg seeds are also known to have bactericidal activity due to the content of myristicin compounds, terpene hydrocarbons, and phenylpropane derivatives [4].

Aceh became one of the most nutmeg producing regions in Indonesia. In Aceh Province the nutmeg oil producing region is concentrate in South Aceh Region, wherein the most widely cultivated is myristica fragans nutmeg. This type of nutmeg has a higher economic quality and higher prices on the international market. Data from the Office of Food Crops and Horticulture in South Aceh District shown that in 2001, South Aceh District was able to produce 4,937 tons of nutmeg with foreign exchange of at least 6.5 billion IDR [5].

Nutmeg consists of 77.8% flesh of fruit, 4% mace, 5.1% shell and 13.1% seed [6]. Nutmeg seed and mace are the most important part of nutmeg and can be made into a variety of products including essential oils and oleoresins. According to [7], nutmeg oil from Aceh has distinctly clear and flavourful, and has a high oil yield range from 5-15%, sensitive to heat and light, soluble in alcohol.
but insoluble in water. This superiority of Aceh nutmeg oil, characteristic, and more derivatives product made nutmeg has great economic value which is able to generate considerable foreign exchange [5].

However, the quality of nutmeg oil produced is still not good because of the conventional refining process. The low quality of nutmeg oil as well as the low content of myristicin, resulting decrease the price value of the nutmeg oil. Many factors can cause the quality of nutmeg oil is poor. These factors can start from the unfavorable businesses handling such as the method of harvesting, harvesting time, post-harvest treatment, including handling of raw materials to the refining process.

Nutmeg oil is produce from the distillation of nutmeg seed or mace, or both. One thing to be considered is the quality of nutmeg oil produced must meet the applicable quality requirements. The quality of nutmeg oil produced is strongly influenced by the methods and conditions of nutmeg oil refining carried out by the producer. And also, handling nutmeg oil during distribution process such as oil purification, mixing, packaging and storage, which can be seen from the different specifications of nutmeg oil listed in the Certificate of Analysis [8]. Nutmeg essential oil obtained from distillation of nutmeg seed or mace must meet the requirements of the Indonesian National Standard (SNI) 06-2388-2006.

One indicator that shows the quality of nutmeg seed or mace oil is determined by the content of myristicin content in which is produces a distinctive aroma. Myristicin is a volatile compound, derivative of the phenylpropanoid compound, a colorless liquid, insoluble in water but soluble in organic solvents, the smell is typical like spices and the aroma is sharp. Its molecular weight is 192 gr/mol. Another name for myristicin is 3-methoxy, 4,5-methylenedioxy-allylbenzene or 5-methoxy safrol. Myristicin is a natural organic compound found in essential oils especially in nutmeg oil. Naturally, myristicin contains insecticides and pesticide agent allows a neurotoxic effect on neuroblastoma tissue and contains psychoactive ingredients with doses higher than those commonly used for cooking. Myristicin is a weak inhibitor of monoamine oxidase. Besides that it can function as anti cholinergic. Myristicin contained in nutmeg oil is toxic, easily absorbed in other constituents in nutmeg oil and has a very intense odor. Myristicin can be used as an anesthetic and a mixture of certain drugs in the pharmaceutical field. This substance is a hallucinogenic and toxic agent that can cause poisoning at excessive doses. However, myristicin is very useful in preventing tumor formation, preventing liver damage to carbon tetrachloride in mice [9].

In terms of improving the quality of nutmeg oil from the results of the traditional distillation it is necessary to purify with vacuum distillers, which are able to isolate myristicin which is an active compound in nutmeg oil. Purification of nutmeg oil using vacuum distillers aims to shorten the time of refining process so that the essential oils obtained have high content of myristicin. Research conducted by [10] which extracted essential oil from nutmeg seeds by steam distillation obtained 7.5% yield and after isolation of myristicin using 70% methanol, yield increased to 12.8%. Likewise, observations made by Marzuki, et al (2014) of 3 nutmeg types from Maluku, the three extracted samples had high myristicin content up to 13.76%. Myristicin, safrol and elimicin are the main aromatic components in determining the quality and distinctive aroma of nutmeg oil, so that the myristine component is used as a determinant indicator of nutmeg oil quality [7].

The method for producing nutmeg oil is generally by steam distillation method, water distillation, steam-water distillation and extraction using solvents. The refining method commonly used is steam distillation due to operation time and oil production capacity. Steam distillation for extracting nutmeg seed oil is conditioned under atmospheric pressure because nutmeg seeds have a fat oil component. High pressure distillation can lead to the possibility of subsequent fat oil, thereby reducing the quality of the volatile oil [11]. Steam distillation is a method for the isolation and purification of compounds. This method is used for liquids that are not mixed or only slightly mixed [12].

The purpose of this research was to improve the quality of traditional refined nutmeg oil by isolating myristicin using a rotary evaporator to increase the quantity and quality of nutmeg oil according to the Indonesian National Standard (SNI) 06-2388-2006.
2. Methodology

The material used in this research are: nutmeg oil from traditional refinery in South Aceh District, cooking oil used as heating media from local market, NaCl (Merck), and distilled water. The equipment on this research are one set Rotary Vacuum Evaporator (Heidolph) complete with chiller for cooling water used in refining process, Centrifuge (Effendorf) used to separate nutmeg oil and other impurities. The myristicin content in nutmeg oil analyzed using Gas Chromatography Mass Spectrometer (GCMS Shimadzu QP2010), Refractometers as refractive index test equipment, and other glass appliances for testing.

Distillation Sequences

There are two distillation sequences of nutmeg oil were carried out in this research. The first distillation sequences (called concentration stage) was carried out at a temperature of 145 °C using a rotary vacuum evaporator for 1 hour at a pressure of 300 mbar with an initial oil sample volume of 200 ml. The volume of residue obtained is measured and further processed at the second distillation sequences (called purification stage).

In the purification stage, as much as 20 ml residue obtained from the concentration stage mixed with 100 ml of water and 10 grams of NaCl, so that it form a two-phase mixture that does not interfere. Purification was carried out using a rotary vacuum evaporator at a pressure of 300 mbar with the same temperature (145 °C) and a distillation time of 40 minutes. Flow chart of the vacuum distillation process as shown in Figure 3.1.

Characterization of Nutmeg Oil

The study begins with the quality test for nutmeg oil which is used as the main raw material, includes color parameters, density, refractive index, solubility in alcohol, and myristicin content and other component in nutmeg oil analyzed using Gas Chromatography Mass Spectrometer (GCMS Shimadzu QP2010). Although, the residu from distillation will be analyzed with the same parameter.

3. Results and Discussion

This research begins with analyze the characteristics of raw materials of nutmeg oil which obtained from SME refinery nutmeg oil. This test is carried out as a comparison of the characteristics of nutmeg oil after the later stages of the distillation process, with focused on myristicin content obtained from the results of the study. Table 1 shows the results of the characteristics of initial nutmeg oil as the raw material in this study.

| No | Parameter                              | Unit        | Result       |
|----|----------------------------------------|-------------|--------------|
| 1  | Colour                                 | -           | Colorless – pale yellow |
| 2  | Odor                                   | -           | Typical of nutmeg oil |
| 3  | Density 20°C/20°C                      | -           | 0,91         |
| 4  | Refractive Index (nD20)                | -           | 1,5          |
| 5  | Solubility in ethanol 90% at 20°C      | -           | 1,3          |
| 6  | Evaporation residue                    | %           | 1,5          |
| 7  | Myristicin                             | %           | 14,26 %      |
The nutmeg oil used as raw material in this research obtained from SME refinery with conventional process in South Aceh District. This study focuses on increasing of myristicin content as a determinant factor for the quality of refined nutmeg oil. The distillation sequences was also carried out in two sequences using a rotary vacuum evaporator. Before refining, this raw nutmeg oil was analyzed to determine the content of myristicin components using GCMS.

**Table 2.** The chemical composition of nutmeg oil from SME refinery

| Compound          | %    |
|-------------------|------|
| Alpha terpinen    | 13,71|
| Beta pinene       | 18,74|
| Gama terpinene    | 4,21 |
| D- limonene       | 9,12 |
| Trans sabinene    | 6,03 |
| Compound                  | %    |
|--------------------------|------|
| Alpha terpinolene        | 3.01 |
| Linalool                 | 1.46 |
| Terpinol                 | 1.31 |
| Trans terpinol           | 10.08|
| Safrole                  | 5.02 |
| Alpha terpinene          | 1.74 |
| Trans phenol             | 1.89 |
| Beta phenol              | 2.14 |
| Ester (geranyl acetate)  | 2.36 |
| Caryophyllene            | 0.77 |
| Eugenol                  | 3.15 |
| Farnesene                | 0.44 |
| Myristisin               | 14.84|

The results of GCMS testing of raw materials found 18 main components of nutmeg oil. From the test results, the 5 largest components were obtained in the components are alpha terpinene 13.71%, beta pinene 18.74%, delta limonene 9.12%, trans terpineol 10.08%, and myristisin obtained at 14.84%. The results of this test are the basic reference for reviewing levels of myristicin on nutmeg oil results of the study.

**Concentration phase (first sequence of distillation)**

The first sequences distillation were conducted at a temperature of 145°C with a pressure of 300 mbar for 1 hour. This condition process refers to [13] which reported the best yield of 6.72%, myristicin content 70.10% and a concentration of 786.99 g/L were obtained at a temperature of 145°C with a pressure of 15 mmHg (under ambient pressure). This phase distillation produce two kind nutmeg oil, namely the top product as distillate and bottom product as oil residue. The two products obtained are then analyzed using GCMS to obtain the chemical composition for each nutmeg oil. Table 3 shows the results of the GCMS test from the top and bottom products resulted from first sequence distillation with a temperature of 145°C, a pressure of 300 mbar for 1 hour. The test results in Table 3 show a significant effect on the content of myristicin obtained. Myristicin levels obtained in the bottom product or residue are much higher than the percent components of myristicin in the top product or distillate.

From this stage, myristicin content in the bottom product (residue) is slightly increase compare to initial myristicin content at nutmeg oil as raw material. This is due to during distillation, there are still many other components wich more volatil compound evaporated to distillate product. Table 3 shows that the terpene component is the most evaporated component, this is because the components of terpenes are compounds that have the lowest boiling point of other compounds of nutmeg oil so that these compounds become the first evaporated compound [14].
Table 3. The compound of nutmeg oil from first sequences distillation indentified by GCMS (5 biggest compound).

| Compound   | Distillate (%) | Bottom product (residue) (%) |
|------------|----------------|-----------------------------|
| alpha terpinene | 17.96          | 13.17                       |
| terpineol   | 11.36          | 12.09                       |
| Safrole     | 8.38           | 7.36                        |
| limonene    | 12.28          | 9.07                        |
| Myristisin  | 6.74           | 19.28                       |

This increase in myristicin content also affects by specified vacuum pressure which used in this study. Vacuum pressure in a distillation process aims to shorten the operating time, where the operation time of nutmeg oil refining conventionally takes a long time. Using this vacuum pressure, the operating time can be shortened so that it can easily isolate the myristicin component in nutmeg oil. The determinant quality factor of nutmeg oil is determined by the content of myristicin because it gives a distinctive aroma to nutmeg oil [1]. Myristicin is a derivative of phenylpropanoid compounds. Myristicin is a clear liquid, insoluble in water but in organic solvents. It smells like spices and aroma is sharp and volatile. Its molecular weight is 192 gr/mol. Another name for myristicin is 5 allyl-1 methoxy-2,3 methylene dioxybenzene or 5 methoxy safo1 [2].

Myristicin obtained from the first sequences distillation of residue product was 19.28%, while myristicin content in distillate product obtained 6.74%. The concentrated fraction obtained still contains mild components of nutmeg oil so that further distillation is needed. The next step after concentration is the purification process, where the sample used is the residue at the concentration stage.

Purification stage

In the purification stage, the residues obtained from the concentration stage are mixed with NaCl solution to form a two-phase mixture that does not interfere. The purpose of adding NaCl solution is to increase the boiling point of the water because salt will only dissolve in water, while the oil does not dissolve. The increase in boiling point of this water is to give a longer time to evaporate light compounds in nutmeg oil so as to obtain an increase in myristicin levels in the fraction. Purification was carried out using a rotary vacuum evaporator at a pressure of 300 mbar with the same temperature and a distillation time of 40 minutes. The product obtained from this stage is the top product (distillate) and bottom product (residue). The use of time is 40 minutes shorter, because the oil sample used from the residue is only 20 ml with the addition of 100 ml NaCl solution which is operated at vacuum pressure with the same temperature causing the oil to evaporate faster. The oil of the nutmeg residue in the purification stage was obtained as much as 1 ml.

The GCMS test results of the 5 largest components of nutmeg oil from distillate and residue obtained from this purification stage can be seen in Table 4. The test results showed a good increase in the levels of myristicin from nutmeg oil distilled from this purification stage. Myristicin level in distillate was 7.54% while at the residue from this purification process was obtained at 53.41%.

The GCMS test results show that the components of alpha terpinene hydrocarbons and aromatic components safo1 are evaporated in refined distillates. Similarly, other compounds
that make nutmeg oil, such as terpene hydrocarbons, have decreased in composition at the residue because they have been evaporated and participated in the distillate product. The operating conditions used in this purification stage are able to isolate myristicin which is the heaviest component of nutmeg oil, this is seen in the residual results, more isolated myristicin than distillate. These results have been able to prove that the isolation of myristicin using vacuum pressure was able to isolate myristicin levels by 53.41%. [14] isolating myristicin from people's nutmeg oil through three stages of distillation at a temperature of 1450°C and a vacuum pressure of 0.2 bar for 1 hour obtained myristicin level of 83.45%.

Table 4. The compound of nutmeg oil from second sequences distillation indentified by GCMS (5 biggest compound)

| Compound          | Distillate (%) | Bottom product (residue) (%) |
|-------------------|---------------|-----------------------------|
| alpha terpinene   | 34.85         | 1.62                        |
| sabine            | 0.56          | 3.29                        |
| Eugenol           | 0.86          | 13.59                       |
| safrole           | 18.69         | 2.29                        |
| Myristisin        | 7.36          | 53.41                       |

The use of vacuum pressure at the distillation of essential oils is quite beneficial in terms of production, which is able to shorten the distillation time so that the active component of nutmeg essential oil is more easily isolated. The results table of nutmeg oil characteristics test results of this study are shown in Table 5. The characteristics of nutmeg oil show the values that are in accordance with SNI 06-2388-2006.

Table 5. Characteristic test results of nutmeg oil after purification process

| No | Parameter                  | Unit         | Result      |
|----|----------------------------|--------------|-------------|
| 1  | Colour                     | Brownis yellow |             |
| 2  | Odor                       | Typical of nutmeg oil |             |
| 3  | Density 20°C/20°C          | -            | 0.88        |
| 4  | Refractive Index (nD20)    | -            | 1.47        |
| 5  | Solubility in ethanol 90% at 20°C | -            | 1:4         |
| 6  | Evaporatiron residue       | %            | 0.1         |
| 7  | Myristicin                 | %            | 53.41 %     |

4. Conclusion

From the results of the above study it was concluded that, the concentration process until the nutmeg oil purification was carried out at a pressure of 300 mbar with a temperature of 450°C capable of isolating myristicin to 53.41% myristicin. Test results of characteristics such as specific gravity, refractive index and other tests have followed the Indonesian National Standard (SNI) 06-2388-2006.

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