# Assessment of Production Rate and Quality Analysis of Essential Oil of Clove Oil Obtained from Hydro-Distillation of Clove Leaves

*Novi* Caroko[1*], *Indah* Hartati[2]

[1]Mechanical Engineering Department Magister Program, 55183 Universitas Muhammadiyah Yogyakarta, Indonesia
[2]Chemical Engineering Department, 50232 Universitas Wahid Hasyim, Semarang, Indonesia

**Abstract.** Clove is one of the leading and potential commodities of the plantation in Madigondo. Currently, the community earns the economic advantage of clove plantation only from the buds. However, other parts of the clove plant have varied potential utilization. For instance, the abundant clove leaves are a valuable and potential source of essential oil. The community considered clove leaves as plantation waste due to a lack of knowledge on further clove leaves processing. The isolation of essential oil from clove leaves can be conducted by applying hydrodistillation. In this research, the hydrodistillation of clove leaves was conducted to characterize the essential oil of clove leaves as well as to investigate the production potency of clove leaves essential oil. The research was also conducted to determine the production rate and capacity of the hydrodistillation of clove leaves. The research shows that the optimum duration of the clove leaves hydrodistillation is 90 minutes. The yield of clove leaves hydrodistillation is 5.4%. The obtained essential oil of clove leaves has to have a density of 1.019 gram/mL and a typical odor of clove oil with a bright and yellow color. Those criteria meet the SNI 06-2387-2006 for oil of clove leaves.

## 1 Introduction

Kulon Progo is one of the regencies within the Yogyakarta Special Region (DIY). It is located on the west side of Yogyakarta. Kulon Progo topography varies from 0-1000 meters above sea level. The plantation sector is one of the economic sub-sectors that is making a high contribution to economic growth in the Regency of Kulon Progo. Cacao, coconut, coffee, cane, and clove are some of the prime plantation crop commodities planted in several locations in Kulon Progo regency. Among the plantation crop commodities commonly planted in Kulon Progo, coffee and clove are the two dominant crops found in Madigondo Village.

Madigondo is located in the urban village of Sidoharjo, sub-district of Samigaluh, Kulon Progo Regency. Madigondo lies at an altitude of 500-1000 above sea level. It covers an area of ±727.87 ha and is situated in the Menoreh Mountain in the north of Kulon Progo. Seen

* Corresponding author: novicaroko@umy.ac.id

from the territorial boundaries, Madigondo Village is surrounded by four districts which are Wonogiri of Central Java on the north side, as well as Wonotawang, Munggang Lor, and Nyemani Villages on the east, south, and west sides, respectively [1].

Based on its location and altitude, Madigondo is suitable for plantation areas. Coffee is the dominant plantation crop found in Madigondo. However, the local people also plant coconut, clove, cacao, durian, and pepper [2]. Being a native plant, clove (Syzygium aromaticum) has been cultivated for centuries in Indonesia including in Yogyakarta. Central Bureau of Statistics of Indonesia listed that there is 56.544 ha of people's plantation area of clove in Indonesia (2020) with a total production of clove buds up to 145.984 ton per year in 2020. Among the overall plantation area, 3.120 ha of it was in Yogyakarta with total production of clove buds up to 724 tons/ year in 2020.

Being one of the most valuable spices that have been used for centuries, people usually plant, harvest, and trade clove buds as the main commodity of clove crops. The local people of Madigondo also prefer to harvest and trade only the clove buds. However, due to the lack of knowledge of the postharvest processing technology, they are not aware of the other potential parts of clove crops i.e., the clove leaves. Nonetheless, each clove tree could produce on average 0.45-0.88 kg of dry leaves in a week [3]. At the moment, the dry leaves of clove are being abandoned and unprocessed. Whereas, the dry leaves of clove are a valuable and potential source of essential oil. The essential oil of clove leaves is up to 1-4% with eugenol and β-caryophyllene as the two major chemical components of the oil. Typically, the eugenol content is reported as high as 70-80% [4], [5]. Eugenol and β-caryophyllene are reported to have a myriad of pharmacological activities such as anti-oxidant, anti-fungal, anti-inflammatory, analgesic, and anesthetic which therefore promote the wide application of clove oil. Clove oil is mostly used in perfume industries, flavor concentrates, and in the pharmaceutical industries [6]. The wide spectrum of clove oil applications has also promoted the economic value of the oil. It is reported that the price of clove oil is $31.5/kg.

The production of clove oil from leaves can be conducted by distillation method. Distillation is defined as the separation of a component from its mixture that has a different boiling point. The volatile component will evaporate at a lower temperature than a less volatile compound [7]. The volatile fraction of clove leaves evaporates and condenses [8]. Three types of distillation are commonly applied in the separation of essential oil from various biomasses i.e., hydrodistillation, steam distillation, and hydro-steam distillation [9]. Other methods can be applied in essential oil separation such as pressing and extraction (leaching). However, distillation is reported to give several advantages over the other two methods [10]. Loppies et al. [11] mentioned that essential oil obtained from 2-4 hours of distillation of clove leaves meet the SNI criteria i.e., the oil color is yellow with the typical odor of clove oil and has eugenol content in the range of 34.01-86.33%. The other benefit of distillation is that the distillation apparatus is simple, so it can be applied easily by the clove farmer.

Considering the abundant availability of clove leaves, the potential production of clove oil from clove leaves, the economical potency of clove oil, and the advantages of the distillation method for clove oil separation; therefore, this research aims to characterize the clove oil obtained from hydrodistillation of clove leaves and determine its production rate.

## 2 Methodology

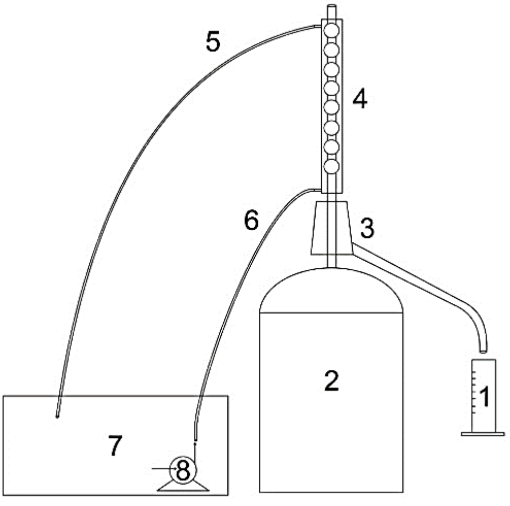
## 2.1 Materials

The clove leaves (Fig. 1) were given by a local farmer from a clove plantation in Madigondo Village, the urban village of Sidoharjo, sub-district of Samigaluh, Kulon Progo Regency, DIY.

**Fig. 1.** Clove leaves.

## 2.2 Distillation

This research was conducted by using a series of distillations (Fig. 2) comprised of steam ketel, condensor, refrigerator, thermocouple, and data logger.




1. Measuring cylinder
2. Distillation chamber
3. Destilate chamber
4. Condenser
5. Outlet of cooling water
6. Inlet of cooling water
7. Cooling water chamber
8. Pump

**Fig. 2.** Distillation scheme.

## 2.3 Research Procedures

The separation of clove oil from clove leaves was conducted by applying the hydrodistillation method. The distillation was performed at a constant temperature for 3.5 hours. The volume of the clove oil obtained from distillation was recorded every 10 minutes. The data obtained was then collected and used for the rate and production capacity analysis. The obtained clove oil was then also subjected to GC-MS (Gas Chromatography–Mass Spectrometry) to determine the chemical component of the oil.

### 2.4 Equations and Mathematics

The rate of the oil separation was calculated by using a series of equations as reported by Caroko et al. [12]:

$$\frac{dM}{dt} = -k \quad (1)$$

$$\int dM = \int -k\, dt \quad (2)$$

$$Mt - Mo = -kt \quad (3)$$

$$CapMt = Mo - kt \quad (4)$$

Equation 2 is used to calculate the oil content in the initial and final phases. In the initial phase, the mass of the biomasses is constant. It will then decrease and in a certain time, the reduction will reach equilibrium. In this phase, the oil content in the sample can be calculated by using equations that are derived from the ratio of the oil content (equation 5-9).

$$\frac{dMt}{dt} = -k\,(Mt - Me) \quad (5)$$

$$MR = \frac{(Mt-Me)}{(Mo-Me)} = e^{-kt} \quad (6)$$

$$\ln\frac{(Mt-Me)}{(Mo-Me)} = -kt \quad (7)$$

$$Mt - Me = (Mo - Me)\cdot e^{-kt} \quad (8)$$

$$Mt = (Mo - Me)\cdot e^{-kt} + Me \quad (9)$$

## 3 Results and Discussion

### 3.1 The Characterization of Clove Oil

The obtained clove oil (Fig. 3) was subjected into organoleptic and density analysis. The color of the clove oil is bright yellow and has the typical odor of clove oil. The color of the clove oil meets the criteria of clove oil as stated and required in SNI 06-2387-2006. It was stated that the color of the clove oil should be in the range of yellow-dark brown. Several literatures reported the separation of clove oil from various parts of the clove plant. Pratiwi et al. [13] mentioned that the color of the clove oil obtained from solvent extraction by using ethanol and hexane is brown and yellow, respectively. It was mentioned that the dark color could be attributed to the undesirable impurities that are also extracted by the solvent. Therefore, it can be concluded that the clove oil obtained from this research is free from impurities.

**Fig. 3.** The clove oil obtained from hydrodistillation of clove leaves.

Meanwhile, the density analysis shows that the clove oil density is 1.019 g/mL. This density is slightly less than the density required in SNI 06-2387-2006 which stated that the density should be in the range of 1.025 – 1.049 g/mL. However, the density of the clove oil obtained from this research was higher than the one obtained from solvent extraction of clove buds in which the range is 0.72-1.008 g/mL. The other literature mentions that hydrodistillation of clove was able to produce clove oil with a density of 1.0420-1.0217 g/mL while the other distillation for 8 hours produced clove oil with a density of 1.0663 g/mL [13]. The low density of the clove oil obtained from this research could be due to the shorter duration of distillation applied.

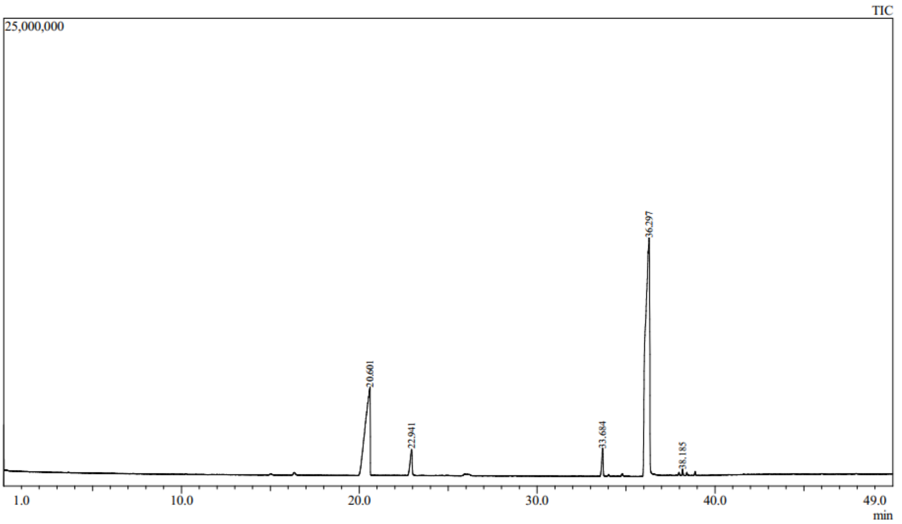



**Fig. 4.** GC-MS spectra of clove oil.

**Table 1.** The chemical component of the clove oil.

| Peak | Compounds | % Area | Molecular weight (g/mol) |
|---|---|---|---|
| 1 | Cis-Caryophyllene | 28.63 | 204 |
| 2 | Alpha-Humulene | 3.46 | 204 |
| 3 | Caryophyllene Oxide | 2.55 | 220 |

| Peak | Compounds | % Area | Molecular weight (g/mol) |
|---|---|---|---|
| 4 | Phenol, 2-Methoxy-4-(2-Propenyl-(CAS)-Eugenol | 64.95 | 164 |
| 5 | Chavicol | 0.40 | 134 |

Furthermore, the chemical composition of the clove oil obtained from this research was analyzed using GC-MS. The chemical profile of the clove oil can be used to determine the quality of the oil. Based on GC-MS analysis (Fig. 4), there are 5 compounds detected in the clove oil obtained from hydrodistillation of clove leaves from Madigondo Village of Kulon Progo Regency. The two major components are eugenol and cis-caryophyllene.

Eugenol or phenol, 2-methoxy-4-(2-propenyl) is a phenylpropanoid compound with a molecular weight of 164.2 g/mol. It is pale yellow and has a spicy taste and special odor. Eugenol is reported to have various pharmacological activities such as insecticide activity, analgesic, antimicrobial, anti-inflammatory, wound healing, antiviral, antioxidant, and anticancer potential. It was also reported that eugenol possesses anticancer activity against different types of cancers such as colon, gastric, breast, prostate, melanoma, leukemia, or skin cancer [14].

Eugenol is also the main component of clove oil that is utilized for the synthesis of isoeugenol. Furthermore, isoeugenol is used in the synthesis and production of a myriad of products such as fragrances for perfumery, skincare, deodorant, soap, shampoo, and detergent. Isoeugenol is also applied as an antiseptic and analgesic in drugs and medicine; raw material for vaniline synthesis, as well as a stabilizer and antioxidant in plastic and rubber [15]. Meanwhile, β-caryophyllene, a natural bicyclic sesquiterpene, is reported to have varied pharmacological activities including neuroprotective, anti-inflammatory, antioxidant, sedative, anxiolytic, anti-depressive, muscle relaxant, anti-atherogenic; anti-tumor and anti-convulsant [16].

The clove oil obtained from hydrodistillation of clove leaves from Madigondo Village of Kulon Progo Regency has eugenol and cis-caryophyllene content of 64.95% and 28.95%, respectively (Table 1). The eugenol content as required in SNI 06-2387-2006 is 78% minimum. Accordingly, the eugenol content of the clove oil obtained from this research is below the SNI requirement. This finding is similar to the one reported by Widayat et al. [17] who reported that the clove oil produced by UMKM in Batang regency also has a lower eugenol content (70%). Therefore, the clove oil produced needs to be further purified.

### 3.2 The Rate and Production Capacity of Clove Oil from Clove Leaves

The hydrodistillation of clove leaves for 3.5 hours produces 54 mL of clove oil. The distillation was conducted by applying 1 kg of clove leaves. Therefore, the yield of the distillation is 5.4% v/w. The yield obtained from this research is relatively high since typically, the essential oil of clove leaves is up to 1-4%. The data on the mass reduction of the clove leaves along the distillation process is shown in Fig. 5.

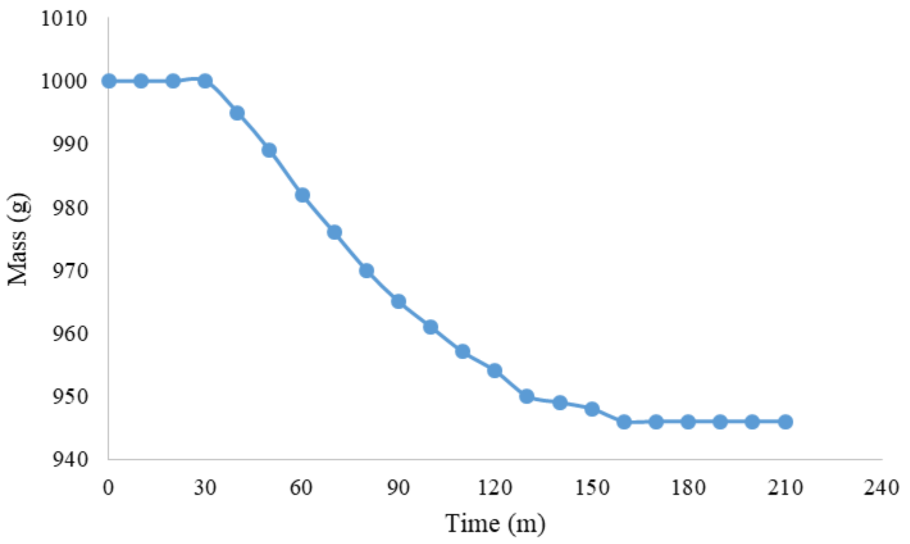



**Fig. 5.** The mass reduction of the clove leaves.

Fig. 5 shows the mass reduction of the clove leaves during the 3.5 hours of the distillation process. The data show that after 160 minutes of the distillation process, there is no further mass reduction of the clove leaves. In the initial phase, the mass reduction is fast and constant. It is shown by the value of the k of 0.6. The k value is obtained from the gradient of the curve from a distillation duration of 30 minutes up to 90 minutes (Fig. 6).

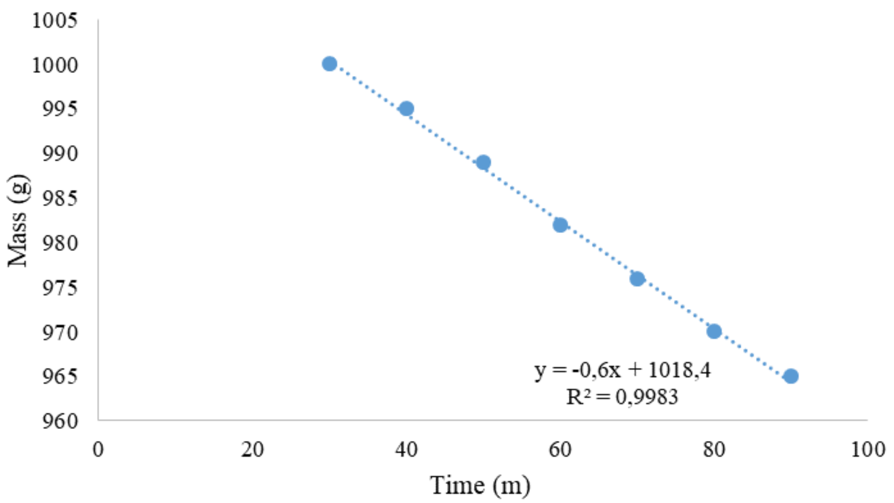



**Fig. 6**. The graph for the k value.

The analysis of the production rate also shows that after the distillation process takes place for 90 minutes, the clove leaves mass reduction rate is not as fast as seen before 90 minutes. This phenomenon appears because the oil of the clove leaves is already separated from the biomass matrix at that condition. Furthermore, on the duration of distillation longer than 160 minutes, there was no longer oil obtained. Moreover, the mass of the clove leaves was no longer reduced. Fig. 7 shows the m value of the final distillation process. The m value obtained is 0.08. This value shows that since the duration is 140 minutes, the production of clove oil is no longer significant. However, the clove oil is still produced until 160 minutes of the distillation process.

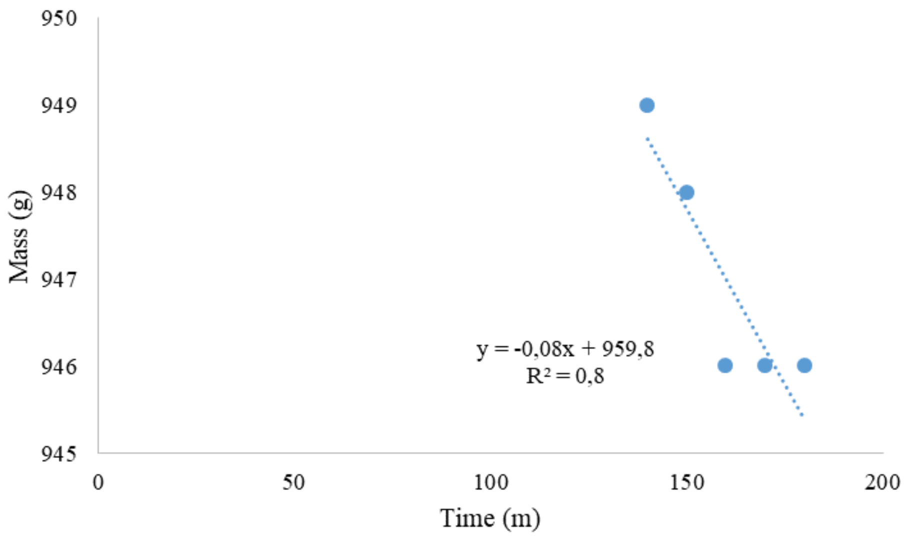



**Fig. 7**. The graph for the m value.

## 4 Conclusions

The hydrodistillation of clove leaves obtained from Madigondo village of Kulon Progo Regency is successfully performed. The hydrodistillation was conducted for 3.5 hours. The following are the conclusions of this research:

1. Based on the k value, it can be concluded that the optimum duration of hydrodistillation for 1kg of clove leaves is 90 minutes.
2. The maximum duration for hydrodistillation of 1 kg of clove leaves is 160 minutes
3. The yield of the process is 5.4%
4. The obtained clove oil has a bright yellow color and a typical odor of clove oil which fulfils the criteria of clove oil as mentioned in SNI 06-2387-2006. However, the density and the eugenol content of the oil (64.95%) is slightly lower than the one of SNI 06-2387-2006 (70%).
5. The clove oil obtained is comprised of two major compounds i.e., eugenol and cis-caryophyllene in which the eugenol content is up to 64.95% and the cis-caryophyllene content is up to 28.63%. The two components are very valuable because both are reported to possess a myriad of pharmacological activities.
6. The clove oil obtained from this research needs to be further purified.

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