Microwave assisted hydro-distillation (mhd) of citronella oil from lemongrass plants (*Cymbopogon nardus*): effect of distiller size on oil yield

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**Abstract.** The essential oil from *Cymbopogon nardus* is widely used in industry, especially medicine and perfume. The microwave assisted method has been successfully applied to a laboratory extraction scale, namely Microwave Hydrodistillation. Therefore, it is necessary to increase the distiller capacity to increase the amount of essential oil obtained. The purpose of this study was to increase the scale of the extraction space through hydrodistillation made from microwaves to produce citronella oil with high yield. It was extracted compared to three distillers 1, 2 and 3 L. The feed to solvent ratio marked as F/S was 0.05 - 0.15 g / and microwave power was 400 – 800 W and extraction time 20 – 60 min. The yield obtained slightly decreased with increasing distiller volume for power parameters and F/S ratio. The highest extraction results using the MHD method occurred at 0.5 cm of material size, 800 W of power and 0.05 g / ml of F/S ratio was 1.81%. GC-MS analysis of citronella oil obtained from the dry leaf extract of *Cymbopogon nardus* showed that the main bioactive composition of citronella oil was citronell, geraniol, and citronellol at 73.76%. Therefore, it makes sense to extract citronella oil from the dried leaves of *Cymbopogon nardus* by using a larger distiller size.

**1. Introduction**

Essential oil is also known as etheric oil or volatile oil produced from some variety plants by some extraction methods. The oil is volatile at room temperature without decomposition of components, has a bitter taste, smells good and distinctive smell in accordance with the type of plants, is generally soluble in organic solvents and insoluble in water [1].

Essential oils are commodities desirable in various industries such as in the perfume, cosmetics, pharmaceutical and food, beverage and beverage industries and can also be used as natural insecticides [2]. As one of the megabiodiversiri centers, Indonesia produces 40 types of 80 types of essential oils traded on the world market. Of these, 13 have entered the world essential market, namely patchouli, lemongrass, cloves, ginger, nutmeg, pepper, cinnamon, sandalwood, jasmine, vetiver, cananga, cajuput, and kemukus. Most of the essential oils produced by farmers are exported with a market share of patchouli 64%, kenanga 67%, vetiver 26%, fragrant lemongrass 12%, nutmeg 72%, clove 63%, ginger 0.4%, and pepper 0.9% from world exports [3]. In the world of trade, this commodity is seen as having a strategic role in producing primary and secondary products and still exists despite price fluctuations, including citronella oil. However, the essential oil industry in Indonesia is still
undeveloped so that extraction of essential oils has not been optimal both in terms of quantity and quality [4,5].

Citronella is a plant that has many benefits, including as a producer of essential oils. This plant is easily cultivated and does not require special treatment, so that people who are interested in developing citronella cultivation have great opportunities. It is currently widely cultivated in tropical and subtropical regions in Asia (Indonesia, Malaysia, India, Ceylon), Africa (Madagascar) and almost all countries in Latin America. At present, 300-350 tons of oil have been produced in India over the past 6-8 years, especially in areas that receive good rainfall and are distributed throughout the year such as in the states of Assam, Karnataka, Maharashtra, Tamil Nadu, and West Bengal, due to these area it is suitable for planting Citronella [6–8]. Therefore, the development of fragrant citronella oil has great potential in Indonesia, moreover the fragrant raw material of citronella is abundant. The production of fragrant citronella oil in Indonesia is produced from various regions dominated by the provinces of Nanggroe Aceh Darussalam, East Java, West Java and Lampung with an area of 3,492 hectares [3].

The extraction of essential oils contained in fragrant lemongrass on an industrial scale is currently carried out by steam and water distillation. The selection of this distillation system is because the material used is in the form of leaves and stems so that the essential oil produced is more, the distillation is shorter and the distilled material does not burn [4]. However, in this system there are disadvantages, namely the yield obtained cannot be optimum and the time taken is longer. Different methods can be used for extracting essential oils from plant materials, for example hydrodistillation (HD), steam distillation, Soxhlet extraction, and distillation extraction simultaneously [9,10] But these molecules are thermally sensitive and susceptible to chemical changes. This extraction method can cause the loss of some volatile compounds, low efficiency extraction, long extraction times, degradation of unsaturated compounds or esters through thermal or hydrolytic effects and toxic solvent residues in extracts. To overcome this problem several new environmentally friendly technologies for extracting essential oils have been developed by researchers [11]. These techniques usually use less solvents and energy, such as supercritical fluids, ultrasound and microwave extraction. The microwave assistance process has been successfully applied in extracting essential oils on a laboratory scale and in various configuration schemes. Microwave-assisted hydrodistillation (MHD) and solvent-free microwave extraction (SFME) are apparently industries important because of their simple configuration, economy and high product yield. [11].

The problem that remains unclear for the new technique is how to increase the capacity of refiners. Several studies have been carried out to increase the capacity of refiners for Microwave-assisted hydrodistillation systems, but the results have not been satisfactory [12]. Therefore, this research is specifically for Microwave-assisted hydrodistillation systems studying the effect of the size of the distillers together with the microwave power and the ratio of feed to solvents for the extraction of fragrant citronella essential oils (Cymbopogon nardus)

2. Material and methods

2.1. Materials
The materials used referred to lemongrass leaves (Cymbopogon nardus) obtained from Pacet Mojokerto, East Java, Indonesia and distilled water as a solvent in the distillation process. The chemicals such as Ethanol, n Hexane and Na$_2$SO$_4$ were obtained from PT. Brataco Chemicals (Surabaya branch).

2.2. Experimental apparatus for Microwave Hydro-Distillation
This study applied microwave hydro-distillation for citronella oil extraction. EMM2308X, Electrolux microwave oven was used. It has a maximal power of 1000 W and a wavelength frequency of up to 2450 MHz. Its dimensions referred to 48.5 cm x 37.0 cm x 29.5 cm. It was modified by drilling a hole
at its top. A 1000 mL round bottom flask connected to Clevenger condenser through the hole was placed inside. The hole was closed with a PTFE to prevent the entry of any contaminants.

2.3. Experimental Procedure

The extraction work carried out consisted of the following steps. The plant material was packed in the distiller and then water was added in a sufficient amount. The samples extracted strung together with Clevenger apparatus. The pre-heating was measured from the moment the microwave was turned on till that of the evaporation beginning. The steam which came out from the extraction chamber was condensed by a condenser apparatus and transported to a separator, where the oil separated automatically from the water and collected a vial bottle. The water in the extraction yield was removed using anhydrous sodium sulfate and cooled to 4°C. The maximum temperature in the distillery was boiling water at atmospheric pressure. The yield was followed every 10 min after preheating. The process continued until reaching the maximum extraction time of 150 min. The extraction was repeated three times for each variable. The extraction yield of Citronella Oil was calculated according to the equation given:

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Yield\ (\%\ ,\ w/w) = \frac{mass\ of\ extracted\ citronella\ oil}{mass\ of\ dry\ citronella\ leaves} \times 100\%\quad (1)
\]

2.4. Operating conditions and variables

- Operating Pressure: Atmospheric
- Materials size: 0.5 cm
- Solvent volume: 0.5 Distiller volume
- Feed to solvent volume ratio (F/S): 0.05, 0.1, 0.15 g/ml.
- Microwave power: 400, 600, 800 Watt.
- Distiller size: 1000, 2000, 3000 mL

2.5. GC-MS Identification

Citronella oils composition was determined by gas chromatography coupled to mass spectrometry (GC–MS) analysis Instrument Agilent 6890N Network GC System, Agilent J & W Capillary GC Column (30 m x 0.320 mm x 0.25μm). The sample was shaken and taken 0.11 μL and injected into the instrument. The conditions maintained were: carrier gas Alpha Gaz Helium Ultra pure; split 1:100; Injection temperature 280°C; oven temperature programmed from 50 to 230°C at 5°C min⁻¹ and from 230 to 280°C at 10°C min⁻¹. Identification of Citronella Oil component.

3. Results and discussion

3.1 Effect of microwave power on citronella oil yield

Power is the amount of energy per unit time (joules / second). Power has an influence on citronella oil yield in the extraction process. In the extraction process using microwave, microwave power acts as a controller of the amount of energy that will be emitted by a magnetron and then captured by water to be converted into heat energy. This power helps the process of the release of essential oils from plant material. The MHD method is closely related to processing power, where the greater the power produced by the microwave, the increase in temperature of the system will more quickly reach the boiling point of water. High power and low time can cause thermal degradation of the product so that it helps the process of removing essential oils from the material to be extracted. The profile of power to yield used by the MHD method in the extraction process can be seen in Figure 1.

In this study it was seen that the most optimal microwave power to produce citronella oil yield varied. Based on Figure 1, the tendency of the increase in yield is seen as the power increases so that it can be said that the greater the energy received by the material to be converted into heat, causing the yield of essential oils to be obtained more and more. Optimum power occurs at a power of 800 W on the MHD method for a yield of 1.312%.
In the MHD method, fragrant lemongrass is submerged by the solvent so that there is an increase in the heating speed in the citronella by direct absorption of microwave energy which causes the pressure gradient to be higher in the material when the microwave power is increased. This happens because microwave energy significantly influences molecular interactions between compounds and others.

High power affects the temperature of the material to be increased and the yield can decrease if more power is used than the optimum point. The rate of evaporation becomes faster so that the oil glands in citronella overheating and cell walls undergo lysis. This causes the material to experience dry product combustion or can be said to be scorched and can reduce yield gains and damage the composition of the essential oils taken. Material characteristics also have an important role in this regard, especially in the pseudo stem section. Because the pseudo stem part is more easily degraded at high temperatures.

Microwave power needs to be controlled to avoid loss of product quality. Furthermore, the oil that has been obtained will be diffused with water solvent and evaporates with the solvent vapor which is then condensed. This study also showed the occurrence of microwave heating in the rapid mass transfer in the sample due to the formation of heat which results in a large vapor pressure difference between the product center and the product surface [10].

![Figure 1. Effect of power on yield (F / S = 0.1 Material Size 0.5 cm)](image1)

![Figure 2. Effect of F/S ratio on yield (D = 2L, size 0.5 cm)](image2)

3.2. Effect of raw materials to distiller ratio on citronella oil yield

In this study, the ratio of raw materials used per volume of solvents for the MHD method is 0.05; 0.1; 0.15; g / ml so that the mass of material used in a 1000 mL flask with a solvent volume of 500 mL is 25, 50 and 75 grams. The material mass ratio affects the density of the material present in the distillation flask. The effect of the ratio of raw materials to yield can be seen in Figure 2. The figure 2 shows that the highest yield was obtained at the lowest F / S ratio (F / S = 0.05), while at the higher F / S ratio the relative yield values were constant. Hence the highest yield value is in the ratio of the weight of the material: the solvent is 0.05 which is equal to 1.81%. Based on the graph, it can be seen that the yield tends to decrease as the mass of raw material increases. This is caused by the increasing mass of material causing the material in the flask to become more dense so that the heat distribution that occurs in the flask is not optimal, as a result part of the material surface is not optimally exposed to heat. The less the mass of the material, the more hollow the mass density of the material, heat can enter through the gaps between the material, heating surface area to increase. This causes the yield to increase as the mass of the extracted material decreases. In the MHD method the amount of solvent added must be in accordance with the optimal ability of the solvent to diffuse to bind the oil that comes out of the lysis oil glands due to microwave heating.
3.3. Effect of distiller size on citronella oil yield

In this study, the size of the material used was the length of fragrant lemongrass 0.5, 1, and 2 cm at each power and ratio. The effect of material size on fragrant lemongrass yield can be seen in Figure 4. Microwave power is also applied to the distillers' different volumes of 1L, 2 L and 3 L. In figure 3 shows that with the increase in the volume of the distillers there is a decrease in yield even though the amount of oil obtained in quantity increases. This shows that with the increase in the size of the distiller, the microwave power divided per unit of solvent or per unit distiller volume will decrease, but this decrease is not as large as the increase in the size of the distiller. This laboratory experiment provides sufficient justification to start increasing the scale of the microwave energy industry as a tool for large capacity extraction. But what needs to be considered is the heat distribution that arises when the wave source is fixed but the volume of the distiller increases.

![Figure 3. Effect of distiller size on yield (F/S = 0.05 g/ml, material size = 0.5 cm)](image)

3.4 Gas Chromatography / Mass Spectrometer (GC-MS) Analysis Results

To determine the components contained in citronella essential oils, Gas Chromatography - Mass Spectrometer (GC-MS) analysis is used. GC-MS analysis not only shows the components contained in citronella oil, but also the levels of these components. Based on the analysis that has been done, the results shown in table 1. discussion of the results obtained, focused on the main components of citronella oil, namely Geraniol, Citronellal and Citronelol.

In the Microwave Hydro-Distillation method, it was found that the content of the main components of citronella oil was greater than generally obtained from conventional distillation. From table 1 shows that the largest components of citronella oil obtained from the Microwave Hydro-Distillation method are Geraniol, Geraniol Formate, Citronellal and Citronelol. From the% composition obtained, it can be seen that the Geraniol and Citronellal content from the MHD method is above the standard maximum limit of 69.73% and 7.82%. The component that meets the standard of the MHD method is Citronelol which is 3.3%.

Ranitha (2014) reports that the chemical profile of Lemongrass essential oil from Malaysia and other scientists' reports, shows that there is a significant quantitative difference between the chemical profiles of the main components of essential oils extracted by conventional hydrodestillation methods and microwave hydrodestillation methods [10]. In Hydrodestillation and microwave hydrodestillation methods, the number of components obtained is more than the conventional method. From the results of GC-MS analysis of the oil in this study, it shows that the number of citronella oil components is 25 as shows in table 1. This difference shows that the MHD method is more effective in extracting essential oil content both in terms of yield and oil quality.
Table 1. Gas Chromatography - Mass Spectrometer (GC-MS) analysis

| No. | Components                          | Compositions |
|-----|-------------------------------------|--------------|
| 1   | O-Methan- 8-ol                      | 0.32%        |
| 2   | Linalool                            | 1.09%        |
| 3   | Rose oxide trans                    | 0.19%        |
| 4   | alpha.- terpinolene                 | 0.33%        |
| 5   | Iso pulegol                         | 1.42%        |
| 6   | (R)-(+-)Citronellal                 | 7.82%        |
|     | 4-Methyl- 1,4-Heptadiene            | 0.42%        |
|     | nonan-8-one                         | 0.18%        |
| 7   | Geraniol formate                    | 12.42%       |
| 8   | (R)-(+)-beta.-Citronellol           | 3.3%         |
| 9   | Geraniol                            | 69.73%       |
| 10  | cis-2,6-Dimethyl-2,6-octadiene      | 0.44%        |
| 11  | Trans-Caryophyllene oxide           | 0.35%        |
| 12  | Camphene                            | 0%           |
| 13  | Methyl Isougenol                    | 0%           |
| 14  | Borneol                             | 0%           |
| 15  | Limonene                            | 0%           |
|     | TOTAL                                | 100%         |

4. Conclusions
The results of extraction of fragrant lemongrass essential oils by the Microwave Hydro-Distillation (MHD) method show that increasing the power will increase the yield of oil. The optimal power in this study is 800 W. Likewise, the smaller the ratio of the mass of the material to the solvent (F / S) the greater the results obtained. And the maximum F / S for the Microwave Hydro-Distillation method is 0.05 g / ml. Optimal extraction conditions for 1 L distillers are 800 watts of power, F / S ratio of 0.05 g / ml and yield of 1.81%. Whereas in the case of an increase in the size of the distillers, it is shown that an increase in the size of the distillers results in a decrease in oil yield, this occurs because the microwave power is divided per unit of solvent or per unit of the volume of the distillers will decrease, but this decrease is not as much as an increase in the size of the distillers. In the Microwave Hydro-Distillation method, it was found that the content of the main components of citronella oil was greater than generally obtained from conversional distillation.

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