Extraction of citronella oil using an ohmic heating method

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Abstract. The basic principle of ohmic heating is the dissipation of electrical energy into heat which causes warming effect and membrane cell permeability. The aim of this study was to explore the tools that use the ohmic heating method to extract essential oils from lemongrass. The extraction process of citronellal and geraniol was using several temperature variations which were at 90 °C using aquadest (citronellal 31.77% and geraniol 18.49%) and at 80 °C using ethanol-n-hexane (citronella 31.75% and geraniol 18.65%). The ohmic heating method based extraction process resulted in a variety of electrodes that were producing the alpha-cadinol compounds which were not found in the control. The average electrical power in the treatment using copper plates were 1287.7 watts and 1116.04 watts. Moreover, the result showed that the produced citronella oil has fulfilled the criteria for citronella oil quality standard according to SNI 06-39531995 which has a density value in the range of 0.850 - 0.892 g/cm³.

1. Introduction
Essential oil is a natural extract from certain types of plants, these natural extracts could be found in the leaves, flowers, wood, seeds and even the pistils of a certain plant. The essential oil also has many other titles such as etheric oil and flying oil which is one of the prospective export commodities. Moreover, the perfume, cosmetics, pharmaceuticals and food industries use a lot of essential oils. The essential oil commodities are very potential in Indonesia, approximately 40 plant species from about 150,200 plant species that are estimated could produce essential oils are in Indonesia [1].

The essential oil commodities potential received considerable attention from the Indonesian government. Citronella oil is a potential oil and one of Indonesia's export commodities. Citronella oil has a high demand, besides it has a stable price and tends to increase. The market needs always increase by about 3 - 5% every year, this shows a high market demand. Indonesia was a major exporter of citronella oil before the Second World War. Unfortunately, the production of citronella oil in Indonesia tends to decrease. Besides that, the quality of Indonesia’s citronella oil is unable to compete with China and Taiwan [2]. Therefore, the main producer of citronella oil is China nowadays.

There are several methods in the process of extracting essential oils from plants, such as distillation, solvent extraction and extraction with supercritical fluids [3]. This study was exploring the extraction tools using the ohmic heating method to extract essential oils from lemongrass. The basic principle of ohmic heating is the dissipation of electrical energy into heat. The advantages of ohmic heating are causing the warming effect and also causing the cell wall permeability. Increase of cell wall permeability can play a role in accelerating the reaction process, increasing the rate of compounds diffusion across the cell wall [4].
2. Materials and Methods

2.1 Materials and Tools
The materials used in this study were mahapengiri variety lemongrass, aquadest, ethanol-n-hexane (4:1). The tools used in this study were ohmic heating extraction tool (Figure 1), copper plate, stainless steel plate, digital scales (Type PW-254), DMA 4500 Density Meter M. Agilent Technologies 7890 Gas Chromatograph with Auto Sampler and 5975 Mass Selective Detector and Chemstation Data System.

![Figure 1. Extraction tool ohmic heating](image)

2.2 Sample Preparation
Mahapengiri variety lemongrass was cut to a uniform size of ± 2 cm.

2.3 Extraction using ohmic heating
This study used two variations of the extraction process. The first extraction process was carried out directly. The sample was soaked with a solvent in an extraction container and then inserted into the extraction container electrode. At this stage, the samples that have been cut with uniform size were weighed as much as 500 grams. Variations carried out at this stage were differences in temperature and solvent. Temperature variations used at this stage were 60 °C, 70 °C, 80 °C, and 90 °C, while the variations of solvents used were aquadest and ethanol-n-hexane. The extraction process at this stage was carried out for 120 minutes. The second extraction process used two variations of electrodes, the electrodes used were copper plate and stainless steel. Samples were put into a cylindrical closed container made of stainless steels and inside it were placed two plates as electrodes. The solvent used in the second stage was distilled water. The extraction process was carried out for 4 hours.

2.4 Analysis of chemical components
The analysis was conducted to determine the chemical components of citronella oil by using Gas Chromatography-Mass Spectrometry (GC-MS). The content of each compound contained in citronella oil has a different retention time and peak area in the chromatogram according to the type of compound analyzed.

2.5 Density Test
The sample density test was conducted using a 4500 M DMA density meter. The densimeter must be calibrated internally by measuring the specific gravity of water until the results are 0.99820 ± 0.00002 g/cm$^2$ before being used to determine the specific gravity of a substance or sample. After the internal calibration is conducted, then the sample is injected in a 4500 DMA density meter.

3. Results and discussion
3.1 Effect of temperature on the extraction process
In this study, the extraction process was using temperature variations of 60 °C, 70 °C, 80 °C, and 90 °C. The GC-MS test results can be seen in Figures 2 and 3, these results indicate a tendency that the higher the temperature used, the more content of geraniol and citronella is produced. In the extraction using aquadest solvent, the highest geraniol and citronella were obtained at a temperature of 90 °C.
which are 18.49% and 31.77%, respectively. On the other hand, in the ethanol-n-hexane solvent, the highest geraniol and citronellal obtained at the extraction temperature of 80 °C, which are 18.65% and 31.75%.

Geraniol and citronellal produced from the extraction of citronella oil are the compounds produced from the secondary metabolites. Secondary metabolites are the chemical compounds that generally have bioactivity and have a function as a plant protector from disease pest disorders both for the plant itself and the environment. Moreover, geraniol and citronellal compounds have antibacterial properties. These antibacterial properties interfere with the process of forming a membrane or bacterial cell wall which results in an incomplete bacterial cell membrane or wall [5]. Essential oils of lemongrass leaves from Tawangmangu are able to produce inhibitory zones against S. aureus and E. coli bacteria [6].

![Figure 2](image1.png)

**Figure 2.** Effect of temperature on the percentage of geraniol and citronella in aquadest solvents

![Figure 3](image2.png)

**Figure 3.** Effect of temperature on the percentage of geraniol and citronella in n-hexane ethanol solvents

The principle of ohmic heating is heating the sample by passing the flow of electric current through an electrode. In this condition, there is a heat transfer process from the electrode. The more heat transfer results in an increment in temperature. This temperature increment could result in an increment in the kinetic energy of a molecule. An increment temperature could also cause damage to proteins or protein denaturation. Protein is one of the main compounds that make up cell membranes, where the essential oils are stored. The presence of proteins in cell membranes reaches half to 2/3 of the total dry weight of the membrane [7].
3.2 Effect of solvent on the extraction process

The effect of the various solvents (aquadest and ethanol-n-hexane) on the extraction process is shown in Figures 2 and 3. Hexane is an alkane hydrocarbon which has the chemical formula of $\text{C}_6\text{H}_{14}$. Hexane is the result of refining crude oil in which composition and fraction are influenced by oil sources. Generally, it ranges from 50% of the weight of the isomer chain. Moreover, all hexane isomers are often used as organic solvents which are inert because of their non-polar properties. Ethanol is a volatile solvent for organic compounds, which is semipolar because it could dissolve both polar and non-polar compounds so that they could dissolve with water. This polarity is caused by the presence of $-\text{OH}$ and non-polar polar groups namely ethyl ($-\text{C}_2\text{H}_5$). Semipolar solvent could induce the polarity level of non-polar solvents. Moreover, it could be used as an intermediate solvent for mixing the polar and non-polar solvents [8]. The hexane solvent serves to dissolve citronella and geraniol in carbon chains contained in the structures of citronella and geraniol, while the ethanol serves to dissolve the aldehyde group.

3.3 Effect of electrode differences in the extraction process

Copper and stainless steel plates were used as electrode variations in this study. The results of the study using electrode variations can be seen in Tables 1, 2 and 3. Based on the results of the GC-MS test, it was found that there were compounds detected in the ohmic heating method but were not detected in the control sample. The compound was alpha-cadinol, this compound functions as an anti-fungal and hepatoprotective. Besides that, it is also known that the yield value in the control sample was 1% while the extracted sample using copper plate had a yield value of 0.66% and the sample extracted using stainless steal had a yield value of 1%.

| Sample Type          | Retention time (second) | Compound                  | Percentages (%) |
|----------------------|------------------------|----------------------------|-----------------|
| Aquades & Stainless  | 8.597                  | dl-Limonene                | 1.71            |
|                      | 21.121                 | Citronella                | 51.61           |
|                      | 31.206                 | 2,6-Octadiene, 2,6-dimethy | 1.95            |
|                      | 36.116                 | Geranyl acetate           | 1.20            |
|                      | 36.406                 | Beta-citronellol          | 17.53           |
|                      | 40.507                 | Geraniol                  | 22.28           |
|                      | 48.978                 | Elemol                    | 1.47            |
|                      | **51.202**             | **Alpha-Cadinol**         | **1.30**        |

| Sample Type | Retention time (second) | Compound                  | Percentages (%) |
|-------------|------------------------|----------------------------|-----------------|
| Control     | 8.601                  | dl-Limonene                | 3.67            |
|             | 21.130                 | Citronella                | 57.39           |
|             | 25.592                 | Isopulegol                | 2.09            |
|             | 31.206                 | 2,6-Octadiene, 2,6-dimethy | 1.70            |
|             | 36.130                 | Nerol (CAS)               | 1.10            |
|             | 36.116                 | Geranyl acetate           | 1.20            |
|             | 36.406                 | beta-Citronellol          | 16.22           |
|             | 40.492                 | Geraniol                  | 15.38           |
### Table 3. Result of GC-MS test of citronella oil extraction using aquades solvent and copper plate

| Sample Type | Retention time (second) | Compound                  | Percentages (%) |
|-------------|-------------------------|----------------------------|-----------------|
|             | 8.606                   | dl-Limonene                | 1.15            |
|             | 21.135                  | Citronella                 | 53.86           |
|             | 31.216                  | 2,6-Octadiene, 2,6-dimethy | 1.89            |
| Aquades & Copper | 36.130            | Geranyl acetate            | 1.13            |
|              | 36.416                  | beta-Citronellol           | 16.27           |
|              | 40.506                  | Geraniol                   | 20.41           |
|              | 45.235                  | Cubedol                    | 1.62            |
|              | 48.978                  | Hedycaryol                 | 1.31            |
|              | **51.202**              | alpha-Cadinol              | **1.47**        |

The extraction process using ohmic heating causes an increment in diffusion through the cell membrane. The heat produced could also damage the cell wall [9]. The increment in mass transportation due to electroplasmolysis is caused by two mechanisms, the first mechanism is the cell membrane electroporation and the second is the thermal denaturation of the membrane. In conventional heating conditions, the second mechanism is more dominant while in the ohmic heating both mechanisms are possible [10]. Plant cell walls consist mostly of cellulose fibers, polysaccharides, and proteins which form an additional barrier to water diffusion. When there is no electric field, the cell wall is usually damaged due to the thermal treatment, then results in changes in physical properties in the temperature range of 60-75 °C [11]. Physical changes that cause the rupture of a cell could increase the point of permeability in the porosity of the cell wall [12]. The permeability of cell membranes and electrical conductivity becomes a linear function of temperature. This process shows that cell walls are susceptible to electrical interference.

### 3.4 Electrical power

Based on the results of voltage and current measurements, it could determine the amount of electrical power used. The time used in the extraction process using copper electrodes and stainless steel was 4 hours. The relationship between power and time can be seen in Figure 4.

![Figure 4. Graph of power and time relationship](image)

The average usage of electric power in the treatment using copper plates was 1287.7 watts and for treatment using stainless steel plates was 1116.04 watts. It is known that the electrical power required in the treatment that was using a copper plate was larger than using a stainless steel plate, due to the higher thermal conductivity of copper if compared to stainless steel. Copper has a thermal conductivity of 401 J/s.m.°C while the stainless steel has a thermal conductivity of 15.1 J/s.m.°C [13].
3.5 Density
Density is one of the important criteria for determining the quality or purity of essential oils. Density is often associated with a heavy fraction of the components contained in it. The greater the weight fraction contained in the oil, the greater the density value [14]. Based on the result of density measurements in Table 4, it is known that the density of essential oils with treatment using electrode variation is larger compared to the control sample. It was identified that the essential oils using a variety of electrodes extracted more chemical components of lemongrass. In addition, results of citronella oil extraction in this study have met the criteria for citronella oil quality standard according to SNI 06-39531995 which has a density value in the range of 0.850 - 0.892 g/cm$^3$.

| No. | Samples                  | Density (g/cm$^3$) |
|-----|--------------------------|--------------------|
| 1   | Aquades + Stainless Steel| 0.87040            |
| 2   | Control                  | 0.86976            |
| 3   | Aquades + Copper         | 0.87061            |

4. Conclusion
The ohmic heating extraction tool could be used to extract the citronella oil. The compound of citronella and geraniol has the highest value at the temperature of 90 °C using aquadest (citronellal 31.77% and geraniol 18.49%) and has the highest value at the temperature of 80 °C using ethanol-n-hexane (citronellal 31.75% and geraniol 18.65%). The extraction process that was using electrodes variation produced the alpha-cadinol compounds which are not found in the control sample. The average electric power used in the treatment that was using copper plate was 1287.7 watts and for treatment using a stainless steel plate was 1116.04 watts. Besides that, citronella oil in this study has fulfilled the criteria for citronella oil quality standard according to SNI 06-39531995 which has a density value in the range of 0.850 - 0.892 g/cm$^3$.

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