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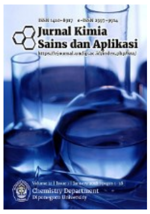

Study on the Upgrading Citronella (*Cymbopogon nardus* L.) Oil Isolation Using Microwave Assisted Hydro-distillation Method

Diana [1,*], Venitalitya Alethea Sari Augustia [1], Arif Hidayat [1], Kiki Adi Kurnia [2]

[1] Department of Chemical Engineering, Faculty of Industrial Engineering, Universitas Islam Indonesia, Yogyakarta, Indonesia
[2] Department of Chemical Engineering, Faculty of Industrial Technology, Institut Teknologi Bandung, Bandung, Indonesia

* Corresponding author: diana@uii.ac.id

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Abstract

Isolation of essential oil using microwave-assisted hydro-distillation (MAHD) method could enhance the yield compared to steam-distillation or conventional hydro-distillation methods. This study explored the isolation of essential oil from Citronella (*Cymbopogon nardus* L.) using the MAHD method with several parameters, such as solid-to-liquid ratio, power of the microwave, and extraction time. The essential oils produced were evaluated based on yield, density, refractive index, and composition. The result showed that the density of the essential oil product was in the range of 0.866–0.902 g/mL, while the refractive index was in the range of 1.470–1.475, which met the ISO standard. Chemical compounds were identified using Gas Chromatography-Mass Spectrometry (GC-MS) analysis. It was found that the total amount of citronellal, citronellol, and geraniol in the product obtained by MAHD is higher than the one from conventional methods. This study aims to identify the most efficient operating conditions to maximize yield while analyzing energy consumption to determine the most cost-effective conditions.

1. Introduction

Indonesia has abundant and diverse biological natural resources, such as many plants that produce essential oils, which have not been optimally explored. Indonesia produces 40-50 types of essential oil-producing plants from 80 types of essential oils traded globally. These oils are the active ingredient of cosmetics, perfumes, medicines, and foodstuffs and are widely used in industry as a flavor and aroma agent. One of the essential oils that have the potential to be developed is citronella oil, which consists of 30-40 chemical compounds, including alcohols, hydrocarbons, esters, aldehydes, ketones, oxides, and terpenes [1, 2]. Citronella oil can be used as an analgesic, hemolytic, anti-enzymatic, sedative and stimulant for stomach aches, and raw material for insect bite prevention lotions [3, 4].

Generally, essential oils are obtained by extraction using solvents or distillation. Two common distillation methods are hydro-distillation and steam distillation. This conventional method has been widely used because it is simple and easy to operate. However, it still has problems related to its high energy, long processing time, and low yield [5, 6].

Currently, many methods are developed to increase the yield of essential oil production. One widely used method is hydro-distillation, which uses microwave energy. The basic microwave heating mechanism involves stirring polar molecules or ions oscillating under electric and magnetic fields. As a result, the particles produce random motion that generates heat. The advantage of choosing a microwave as a heating medium is that the microwave can work quickly and efficiently. This is due to the presence of electromagnetic waves that can penetrate the material and evenly excite the material's molecules [7].

Several studies using microwave-assisted hydro-distillation (MAHD) have been reported, such as essential oils extraction of rosemary [8], magnolia [9], mas cotek (*Ficus deltoidea*) [10, 11], mace [12], lemongrass [13, 14], cinnamon [15], patchouli [16, 17], fennel [18, 19] and vetiver [20]. Most studies reported an increase in the yield of essential oils and a reduction in processing time while using the MAHD method compared to conventional distillation. Several studies stated that the MAHD method also produced better-quality essential oils. However, it should be highlighted that the MAHD method has not been reported to isolate citronella oil from *Cymbopogon nardus* (L.) and its energy evaluation.

Therefore, this study aims to apply the microwave-assisted hydro-distillation method to isolate essential oils from citronella leaves and compare it with the conventional methods (hydro-distillation and steam distillation). Furthermore, factors that affect the process are also studied to find operating conditions that can produce optimal oil yield and quality.

2. Experimental

2.1. Materials

Citronella leaves were obtained from plantations in Sleman, Yogyakarta, Indonesia. First, the leaves were cleaned and then dried in an open shade for three days to reduce the moisture content without damaging the material. The preliminary research found that more extended time drying would produce dark-colored oil. After drying, the leaves were cut into 2 cm lengths, put in a tightly closed container, and stored in the refrigerator. The 2 cm length of leaves was chosen based on the result published by Tran *et al.* [13], where the authors stated that a particular size could produce an optimal volume of essential oil.

2.2. Methods

Citronella oil extraction was carried out in a microwave oven (Electrolux EMM2308X) connected to a Clevenger apparatus that could continuously return condensate water to the flask, as shown in Figure 1a. For comparison, citronella oil was extracted using hydro-distillation, employing a mantle heater (Figure 1b) operated for 240 minutes. The distillation process was terminated at the 240-minute mark, as no further oil was obtained beyond that point.

At the end of the process, the oil layer was separated and cooled in the freezer. Once the water entrained in the oil was frozen, the water-free oil was separated for analysis. The microwave oven can be operated in various conditions with a maximum power of 800 watts at a 220 v –50 Hz voltage. The microwave-assisted hydro-distillation operation was carried out with variations in the ratio of solvent: solid ratio of 2.5:1, 5:1, 10:1, and 15:1. This variation was selected with the condition of the raw material being partially submerged (2.5:1), half-immersed (5:1), fully immersed (10:1), and floating (15:1).

In the fully submerged condition, the lowest microwave oven power setting capable of generating steam was 400 watts (50%). Thus, experiments were conducted using power settings at 50, 60, 75, 95, and 100% of the maximum power. Simultaneously, the operation was run for 15, 30, 60, 90, 120, and 240 minutes.

A flask containing 50 grams of citronella leaves and distilled water in a specific ratio was placed inside a microwave oven. The steam produced during the procedure was directed into the Clevenger apparatus, positioned outside the oven. Processing time was calculated starting from the formation of the first drops of condensate. At the end of the process, the oil layer was separated and cooled in the freezer. The water entrained in the oil solidified, allowing for the separation of the water-free oil for subsequent analysis.

2.3. Sample Analysis

The volume of oil obtained from each variation was measured, and the yield was calculated using Equation (1).

$$Y = \frac{V}{m} \times 100 \quad (1)$$

Where, Y is yield (%, v/w), V is the oil volume (mL), and m is the weight of raw material (g).

In addition, a density test was also carried out using an Anton Paar DMA 4100 M densitometer and a refractive index test with a Kruss DR600 refractometer. A Gas Chromatography-Mass Spectrometry (GC–MS) Shimadzu GP-2010 equipped with an Rtx-5ms column with a diameter of 0.25 mm and helium as a carrier gas was used to analyze the components in the essential oil. The GC-MS analysis was obtained with the following settings: 0.75 mL/min of flow rate, 130 of split ratio, 200°C of injection temperature, 0.2 L of injection volume, and heating was conducted in the temperature range 60–200°C at a rate of 10°C/minute.

3. Results and Discussion

In this study, the extraction of citronella oil using microwave-assisted hydro-distillation (MAHD) at an 800-watt power setting (100%) was compared with two other methods: hydro-distillation (HD) and steam distillation (SD). The distillations were conducted using the same raw materials and conditions, with a processing time of 4 hours and a solvent-to-solid ratio of 10. This ratio ensured that all the leaves were fully immersed in distilled water during hydro-distillation. Subsequently, microwave-assisted distillation experiments were performed with various parameters, including different solvent-to-solid ratios, microwave power settings, and processing times. The results from all experiments are presented in Table 1.

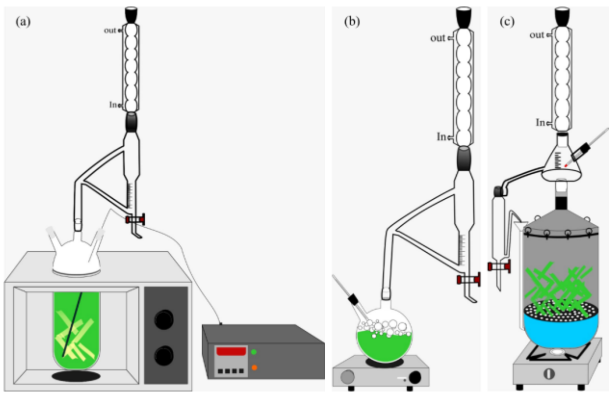

Figure 1. Apparatus for (a) microwave-assisted hydro-distillation, (b) hydro-distillation, and (c) steam-distillation

Table 1. Yield of citronella oil on various parameters

No	Method	Power (watt)	Ratio	Time (hour)	Yield (%, v/w)
1	SD	0	10	4	3.2
2	HD	0	10	4	3
3	MAHD	800	10	4	3.7
4	MAHD	800	15	4	3.6
5	MAHD	800	5	4	3.4
6	MAHD	800	2,5	4	3.2
7	MAHD	720	10	4	3.6
8	MAHD	600	10	4	3.5
9	MAHD	480	10	4	3.2
10	MAHD	400	10	4	3
11	MAHD	800	10	0.25	2.4
12	MAHD	800	10	0.5	2.9
13	MAHD	800	10	1	3.2
14	MAHD	800	10	1.5	3.6
15	MAHD	800	10	2	3.7

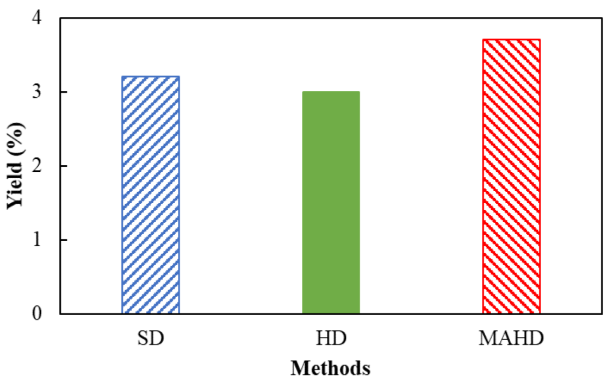

Figure 2. The yield of citronella oil using different methods

Figure 2 illustrates a comparison of essential oil yields obtained from three different methods. Notably, the MAHD method demonstrated the highest yield at 3.7%. This outcome represents a 15% increase compared to the SD method and a substantial 23% increase compared to the HD method.

Microwave heating heats the distilled water used as a solvent and the in-situ water content within the material matrix. Radiation heating occurs from the inside out, aligning with the direction of mass transfer during the extraction process. The combination of heat and mass transfer directions, which occur from the inside out, facilitates the diffusion of essential oils in the material matrix. In addition to radiation, convection, and conduction heat transfer mechanisms occur around the water and materials. Combining the three heat transfer mechanisms supports the process of extracting essential oils.

3.1. Effect of Solvent: Solid Ratio

Figure 3 illustrates the effect of varying ratios of distilled water to raw materials on the yield of citronella oil, using raw materials of 2 cm in size, 800 W of microwave power, and a distillation time of four hours. The mass of the raw materials for all experiments was consistently 50 g. As shown in Figure 3, the oil yield increases with the water-to-raw-material ratio, rising from 3.2% at a ratio of 2.5 to 3.7% at a ratio of 10. However, beyond this point, the yield subsequently decreases to 3.6% at a ratio of 15.

During the process with MAHD, microwave heat is released within the material, causing the essential oil pockets to rupture and drawing the oil along with the steam. If there is inadequate water to dissolve the colloidal mixture and the essential oil trapped within the salt, the essential oil cannot be effectively separated. Utilizing a larger quantity of water results in increased oil diffusion. Water readily permeates the material, effectively carrying away soluble components and enhancing the essential oil extraction yield. However, excessive solvent absorbs microwave energy for heating the solvent, consequently diminishing the amount of oil extracted. As depicted in Figure 3, a ratio of 10 provided optimal results and was therefore selected for subsequent experiments.

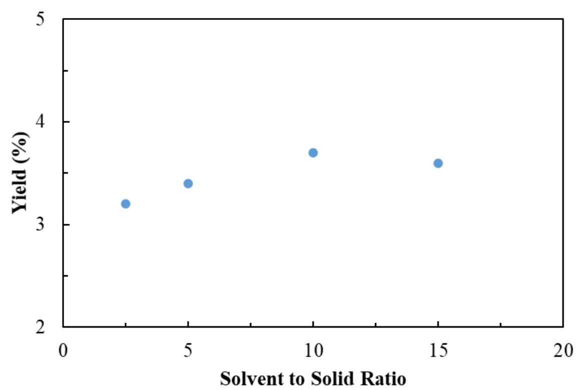

Figure 3. Effect of solvent-to-solid ratio on essential oil yield

Table 2. Energy requirement rank

Method	Power (watt)	Ratio	Time (hour)	Energy (kWh/kg oil)	Rank
MAHD	800	10	0.5	204	1
MAHD	800	10	0.25	225	2
MAHD	800	10	1	281	3
MAHD	800	10	1.5	375	4
MAHD	800	10	2	486	5
MAHD	400	10	4	580	6
SD	400	10	4	605	7
HD	400	10	4	658	8
MAHD	480	10	4	674	9
MAHD	600	10	4	770	10
MAHD	720	10	4	899	11
MAHD	800	10	4	972	12
MAHD	800	15	4	999	13
MAHD	800	5	4	1058	14
MAHD	800	2.5	4	1124	15

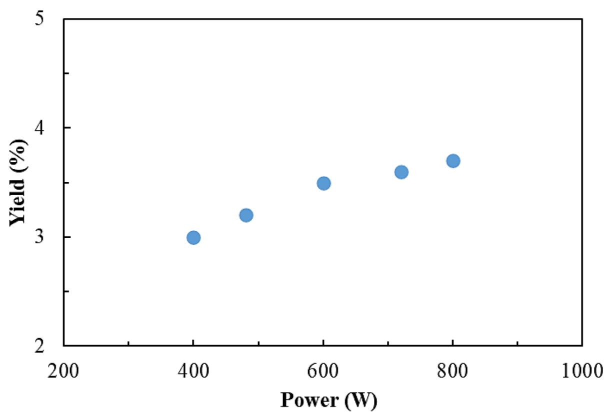

Figure 4. Effect of microwave power on essential oil yield

3.2. Effect of Microwave Power

Microwave energy plays a significant role in the citronella oil isolation process. Experiments were conducted using 2 cm-sized lemongrass leaves, a 4-hour duration, and a water-to-raw-material ratio of 10. It was observed that power levels below 400 watts failed to yield essential oil. Consequently, a range of power variations from 400 to 800 watts was employed in this study.

As depicted in Figure 4, when a power level of 400 watts is employed, the oil yield matches that of the hydro-distillation method, which is 3%. Furthermore, Figure 4 illustrates that an increase in the power level correlates with a higher yield of citronella oil. The escalation in power from 400 watts to 800 watts is capable of elevating the yield from 3% to 3.7%. Consequently, higher microwave power levels lead to increased oil yield. This is because of the higher energy supply, which enhances the movement of the constituents and accelerates the motion of the components within the mixture. This, in turn, promotes more effective diffusion and facilitates greater water penetration into the material layer, ultimately enhancing oil extraction efficiency. The most favorable outcomes were achieved at 800 watts of power; therefore, this power setting was adopted for further research.

3.3. Effect of Processing Time

Figure 5 shows the effect of time on citronella oil extraction. The experiment used an 800-watt power level and a solvent-to-solid ratio of 10. It is evident that as the duration of the process increases, the yield of essential oil also rises. The yield reaches 3.6% at 1.5 hours and 3.7% at 2 hours. However, no further addition of essential oil is observed beyond the 4-hour timeframe.

The incremental rise in yield between 1.5 hours and 2 hours of extraction time was not notably significant. Visually, it was observed that the oil obtained after a longer duration turned darker. This color change is attributed to the denaturation of certain substances caused by prolonged exposure to high temperatures, which can impact the quality of the final product. Additionally, extended extraction times result in higher energy consumption and increased production costs. Therefore, the optimal extraction time for citronella oil using the MAHD method was 1.5 hours.

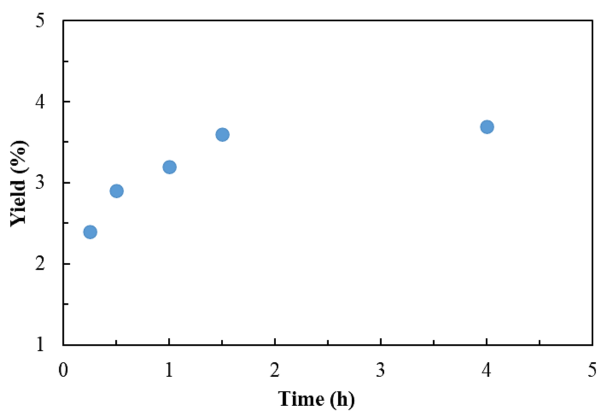

Figure 5. Yield of citronella oil during processing time

Table 3. Compounds in the essential oil of citronella

Peak#	Retention time (min)	Component
1	3.906	Tricyclene
2	4.027	Alpha pinene
3	4.222	Camphene
4	4.655	Myrcene
5	5.215	Limonene
6	5.27	Ocimene
7	5.761	Propyl amyl ketone
8	6.056	Alpha terpinolene
9	6.177	Linalool
10	6.949	Citronellal
11	7.387	Limonene oxide
12	8.082	Isopulegol
13	8.145	Z citral
14	8.281	Citronellol
15	8.444	Citronellol
16	8.519	Geraniol
17	8.713	Geranial
18	8.835	E citral
19	8.968	Bornyl acetate
20	9.776	Citronellyl acetate
21	10.208	Geranyl acetate
22	10.914	Caryophyllene
23	11.726	Germacrene
24	12.135	Elemol

3.4. Energy Requirements

The energy requirements for all experiments have been assessed, and the findings are detailed in Table 2. When evaluating the three methods under identical operational conditions—specifically, using a power level of 400 watts and a processing time of 4 hours—the MAHD method ranks 6th, with an energy requirement of 580 kWh/kg of oil. In comparison, the SD method ranks 7th, requiring 605 kWh/kg of oil, while the HD method ranks 8th, necessitating 658 kWh/kg of oil. Furthermore, the top five positions are held by the full-powered microwave-assisted hydro-distillation method (800 watts) when the processing time is limited to 2 hours. These methods exhibit significantly lower energy consumption than full-powered steam distillation and hydro-distillation methods (400 watts).

3.5. Product Characterization

All samples obtained were then analyzed for density, refractive index, and composition. The results of the density and refractive index tests are shown in Figures 6 and 7. It can be seen that the majority of the essential oils produced exhibit densities within the specified standard range (ISO 3848:2016.279) of 0.880–0.902 g/mL. Furthermore, all products conform to the citronella oil refractive index standard (ISO 3848:2016.280), falling within the range of 1.446–1.477.

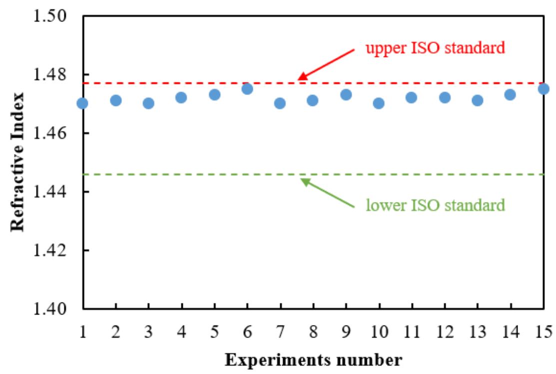

Figure 6. The density of citronella oil from experiment products

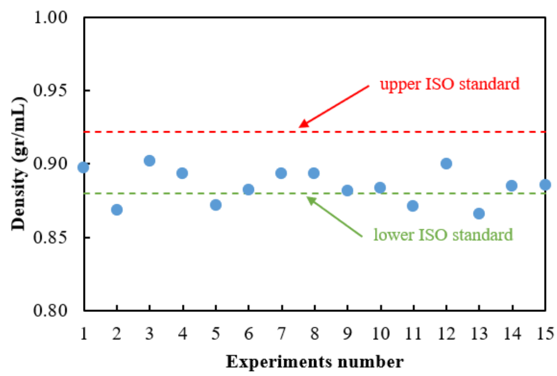

Figure 7. Refractive index of citronella oil from experiment products

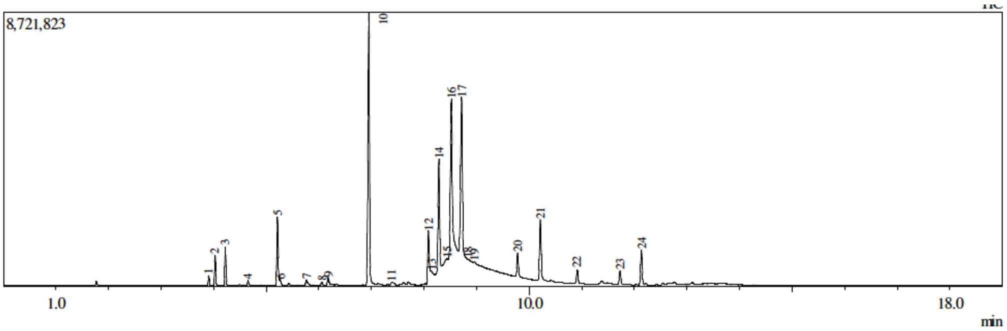

Figure 8. Citronella oil product chromatogram (MAHD 1.5 h)

Table 4. Comparison of SD, HD, and MAHD methods

Parameter	Method		
	SD	HD	MAHD
Time (h)	4	4	1.5
Yield (%, v/w)	3.2	3	3.6
Density (g/mL)	0.8972	0.8685	0.8847
Refractive index	1.470	1.471	1.475
Citronellal (%)	20.47	7.43	19.83
Citronellol (%)	7.42	9.16	14.96
Geraniol (%)	24.79	35.99	36.38

The composition of the ingredients contained in citronella oil was identified by the Gas Chromatography-Mass Spectrometry (GC MS) test. One of the chromatographic results is presented in Figure 8 and detailed in Table 3. Key compounds identified in citronella oil include citronellal at a retention time of 6.947, citronellol at 8.281, and geraniol at 8.519. Additionally, limonene, isopulegol, geranial, citronellyl acetate, and geranyl acetate were also detected.

Table 4 provides a comparison of the oil characteristics obtained through three different methods: steam distillation (SD), hydro-distillation (HD), and microwave-assisted hydro-distillation (MAHD). Among these methods, the HD method yields the least amount of citronella oil; its density falls outside the ISO standard and contains the lowest citronellal content. In contrast, the MAHD method demonstrates the ability to generate a higher oil yield in a shorter duration. It offers elevated levels of valuable compounds, including citronellol, citronellal, and geraniol. As indicated in Table 1, the MAHD method requires only 1 hour to attain a citronella oil yield of 3.2%, while the SD method necessitates 4 hours to achieve the same yield. This implies that the MAHD method reduces processing time and conserves energy usage.

4. Conclusion

The utilization of the microwave-assisted hydro-distillation (MAHD) method for citronella oil extraction was conducted and compared with traditional methods, specifically steam distillation (SD) and hydro-distillation (HD). These experiments involved 2 cm-sized pieces of dried citronella leaves. The MAHD method yielded 3.6% citronella oil within a mere 1.5 hours, while the SD and HD methods yielded 3.3% and 3%, respectively, necessitating a lengthier 4-hour extraction process. In terms of energy consumption, the full-powered MAHD method, used for 1.5 hours, consumed 375 kWh/kg oil, which is significantly lower than the 4-hour full-powered SD (605 kWh/kg oil) and HD (658 kWh/kg oil) methods. Furthermore, based on GCMS analysis, the MAHD method yielded a total citronellol, citronellol, and geraniol content of 71.17%, surpassing the SD (52.68%) and HD (52.58%) methods. In conclusion, the MAHD method saves time and energy and enhances the quality of the extracted citronella oil.

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