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Article

The Extraction of Essential Oil from Piper Cubeba by Using Solvent Free Microwave Extraction Method

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Abstract. This research aims to study the production of essential oil from Piper Cubeba using the solvent-free microwave extraction (SFME) method. Before extraction, the raw materials are prepared in the form of grains and powders to vary the size of the raw materials. Time variation was carried out at microwave power 180 Watt Analyses were then performed on the resulting essential oil using both physical and chemical tests of density and solubility with 96% alcohol. Analyses of chemical compounds within the essential oil were then performed using the Gas Chromatograph Mass Spectrometry (GC-MS). It was found that the production of essential oil from Piper Cubeba using the SFME method was affected by microwave power, grain size, and treatment duration. Results from physical and chemical analyses revealed that the resulting cubeb oil has 0.86 g/mL density, which means that essential oil produced using the SFME method is lighter compared than those produced using MAHD methods. Results from GC-MS analyses showed eight components detected from the production of essential oil from Piper cubeba using the SFMEmethod; Copaene (39.28%), Cubebene (23.83%), Isoledene (11.66%), Naphthalene (6.65%), Phellandrene (5.81%), Asarone (5.71%), Cadidene (4.90%), and Caryophyllene (2.16%) which contents belong to the sesquiterpene group..

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1. Introduction

Essential oil is one of Indonesia non-oil and gas export commodities that have been cultivated since before World War II. According to data from DAI (Indonesian Essential Oil Council), Indonesia is still listed as one of the world's largest suppliers of essential oil raw materials, and has even supplied up to 90% of patchouli essential oil (around 1600 tons/year). Of the 150 types of essential oils traded on the international market, 40 of them can be produced in Indonesia, but only a few are used commercially and which meet export quality standards, only 12 are available, such as cinnamon oil, vetiver oil, sandalwood oil, oil. cubeb, patchouli oil, cananga oil, nutmeg oil, clove oil, eucalyptus oil. Until now, Indonesia's total essential oil production capacity can reach 5.000 to 6.000 tons per year with the number of business actors reaching 3.000 businesses. Data from industry records that even though Indonesia is the world's largest producer of essential oil [1].

Indonesia is a tropical country that has high biodiversity, rich in flora and fauna [2]. Indonesia has great potential to have a large scale essential oil industry. There are all sorts of essential oils available in here. One with promising economic potential is (Cubeb Oil). Piper cubeba is also used to treat bronchitis and cough, other than for diuretic and antiseptic agent. Piper cubeba shows the activity as an antioxidant, anti-bacterial, and functional, and antinuclear [3]. Cubeb oil heals throat infection and eases breathing difficulty [4]. It can also be used to create the fragrance of spices and other scents [5]. The chemical compounds responsible for fragrance in Piper cubeba include cubebene, dipentene, cadinene, and chamfer [6].

Traditionally, essential oil from *Piper cubeba* can be extracted using steam distillation. However, this method comes with some drawbacks, including easily evaporated compounds, low extraction efficiency, lengthy processing, less ester content due to heat, and toxic residues [7]. Therefore, this process will reduce production costs, be efficient, and also be environmentally friendly. Among the most anticipated new methods that are capable of providing expected results include supercritical extraction [8], vacuum microwave-assisted extraction (VMAE), Microwave-assisted hydrodistillation (MAHD) [9], and solvent-free microwave extraction [10]. Supercritical extraction is separating components at critical pressure and temperature levels of fluids [11]; hence, this process is expensive and risky. VMAE is an extraction process using a microwave in a vacuum. In this method, the extraction boiling point of the solvent must be lower than the boiling point at air temperature. Therefore, extraction using VMAE results in comparatively more essential oil than using distillation [12]. MAHD is a new method of extracting essential oil. It can speedily produce quality results, and the process is itself is environmentally friendly [13].

This research extracted essential oil from Piper cubeba using the microwave extraction (SFME) method. It is the same method as MAHD without the addition of water or solvent. This means that the process is cost-effective, efficient, and environmentally friendly. Those features are also supported by results from experiments using MSFE, and one of them is by Chen [14]. Extraction of essential oil from aromatic herbal ingredients (basil, crispate mint, thyme) and seasonings (ajowan, cumin, star anise) with yields of basil 0.03% v/w, crispate mint 0.03% v/w, thyme 0.16% v/w extracted for 30 minutes, ajowan 1.41% v/w, cumin 0.63% v/w, star anise 1.38% v/w extracted using SFME for 30 and 60 minutes, respectively. Meanwhile, results obtained from steam distillation are the same for both 4-hour and 8-hour distillations [15].

Solvent-free microwave extraction used to extract Schisandra Chinensis essential oil for 50 minutes came with a 0.92% v/w result. Yields gained from water distillation are almost the same with SFME. The only difference is the time it takes .

Steamed essential oil has great potential for various industries, it requires optimal operating conditions to produce oil with a higher yield compared to conventional methods or other new methods. Several operating conditions to obtain high yields for cubed seed extraction using SFME were microwave power, raw material size and time. In addition, the essential oil of cubeb seed oil obtained will be observed for its chemical compound composition and physico-chemical properties. So, from the two references above, it can be concluded that the SFME method is better than the water distillation method and the extraction of cubeb essential oil using the SFME method can be done to fulfill this research.

2. Experimental Section

2.1. Material and Instrument

The instrument used for this research are: microwave, hose, glass extractor, graduated glass, beaker glass, balance, blender, electric oven, thermometer, stative clamp, soxhlet extractor, water bath, boiling flask, sieve, porcelain cup, glass funnel, 50 mm pipette, sample bottle. Meanwhile, the materials used for this research are Piper cubeba, n-hexane, filter paper, water, yarn, and 96% ethanol.

2.2. Procedure

1). Material Preparation

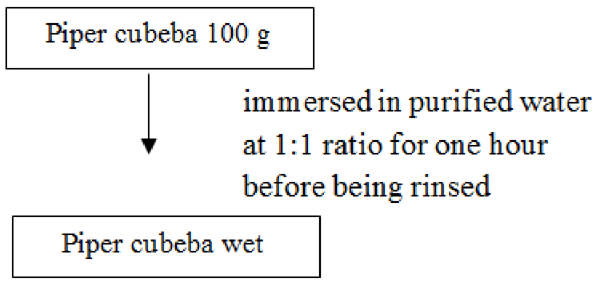

Figure 1. Material preparation I

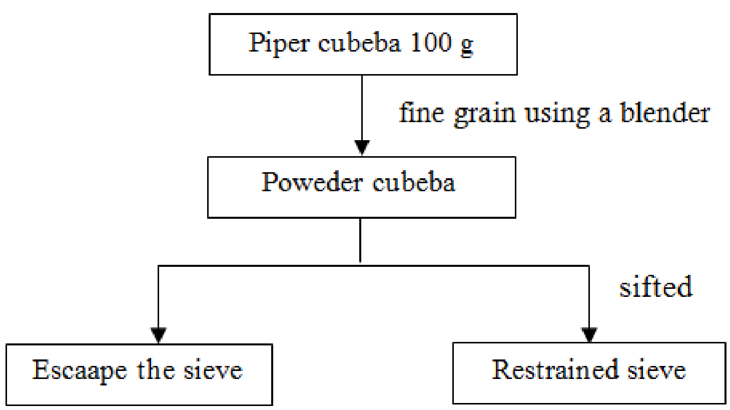

Figure 2. Material preparation II

2). Analysis of Essential Oil Content in Piper cubeba (Standard Procedure)

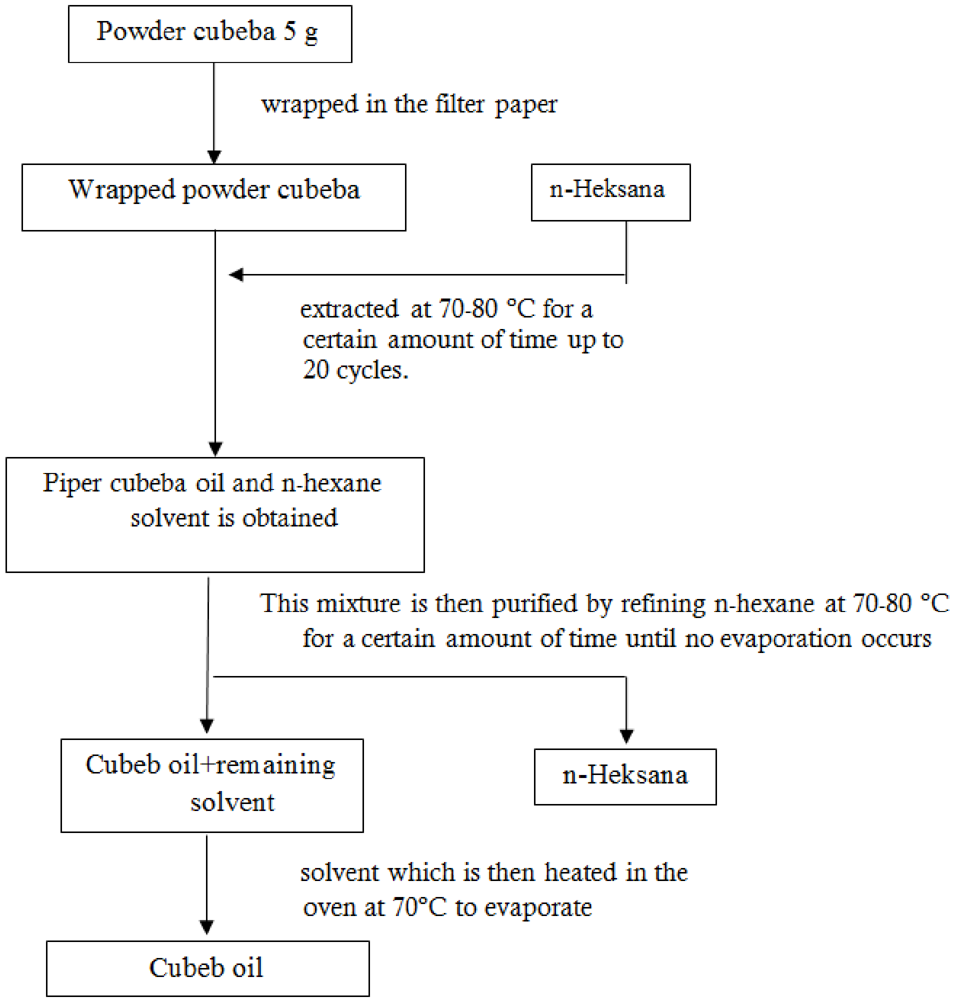

Figure 3. Analysis of Essential Oil Content in Piper cubeba (Standard Procedure)

3). Obtaining Essential Oil from Piper cubeba using Solvent-Free Microwave Extraction Method

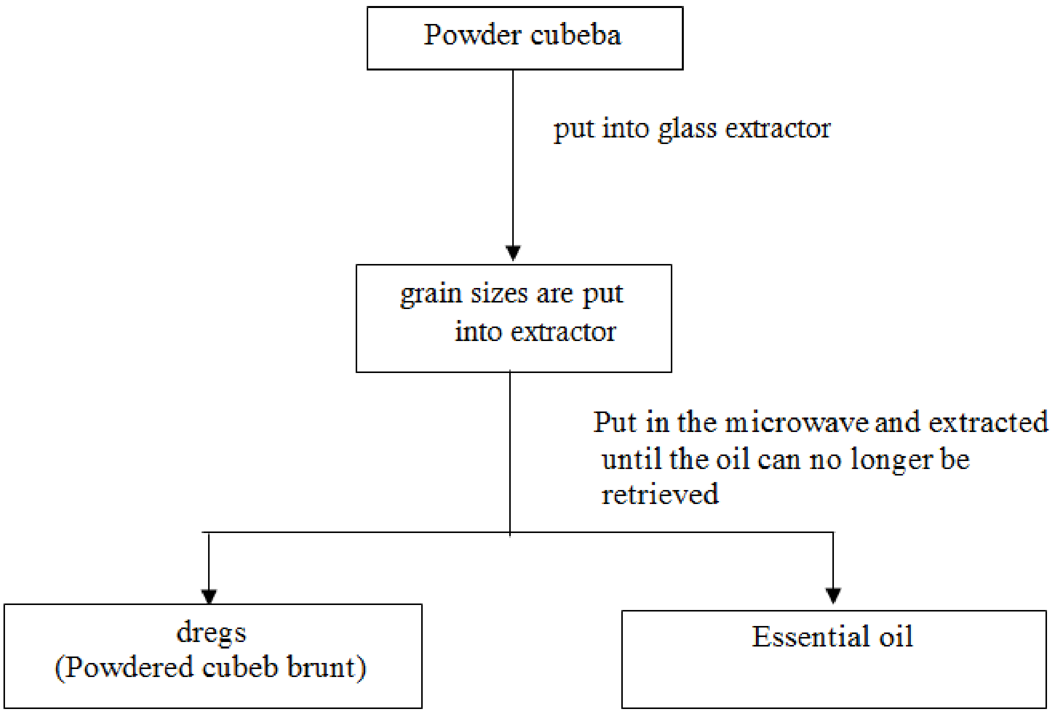

Figure 4. Obtaining Essential Oil from Piper cubeba using Solvent-Free Microwave Extraction Method

4). GC-MS Testing for Essential Oil from Piper cubeba

GC-MS testing in, principle is about identifying compounds based on their molecular weight. This method gives good results in the analysis of essential oil quality [16]. These identified compounds are then compared to the data available in GC-MC memory. GC-MS analysis for essential oil from Piper cubeba made use of GC Perkin Elmer device with GC Clarus 680 & MS SQ 8T, with the following conditions: PE Auto System GC instrument type, sampling speed of 1.5625, carrier P Flow-He control, 30 meter column length, split flow of 20 mL/minute, initial set point at 1 mL/minute, diameter of 250 µm, programmed oven temperature at 50-350^{o}C, total running time of 16.5 minutes, and library The NIST Mass Spectral Search Process for The NIST/EPA/NIH Mass Spectral Library Version 2.

5). Density Test for Essential Oil from Piper cubeba

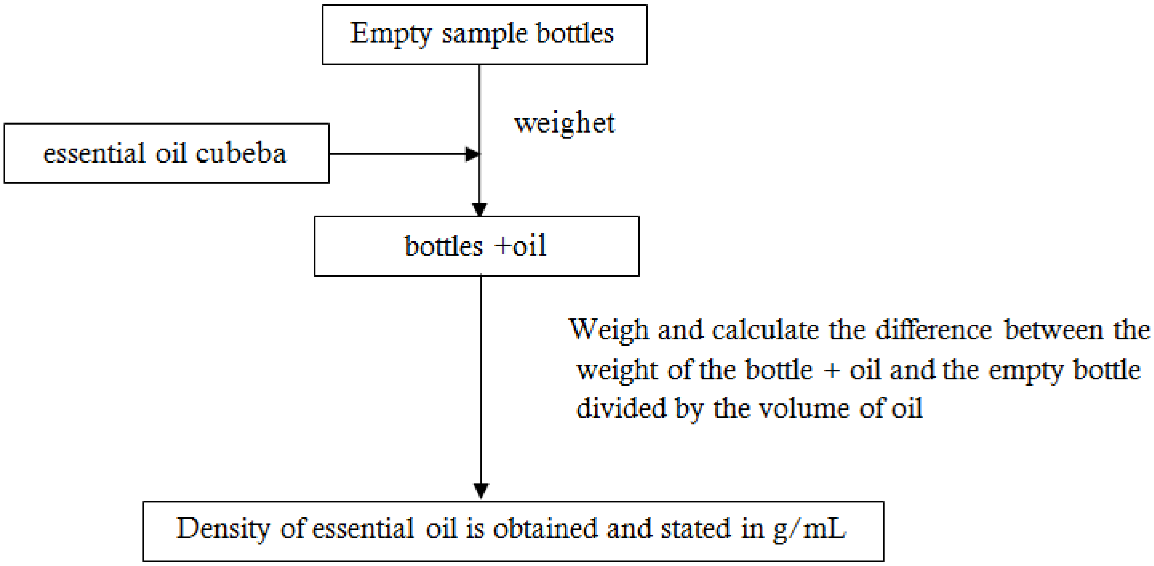

Figure 5. Density Test for Essential Oil from Piper cubeba

6). Solubility Test for Essential Oil from Piper cubeba

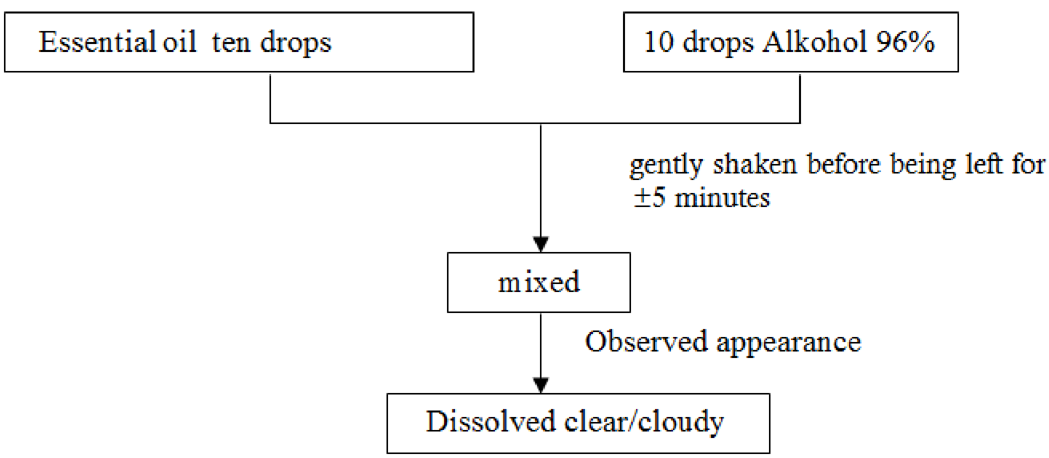

Figure 6. Solubility Test for Essential Oil from Piper cubeba

3. Results and Discussion

3.1 Analysis of Essential Oil Content in Piper cubeba (Standard Procedure)

Analysis of essential oil content in cubeb seeds was carried out to determine the quality of cubeb seeds and calculate the efficiency of SFME extraction. It is performed using the n-hexane extraction method with the help of a Soxhlet extractor. After extraction, the resulting essential oil must be purified by heating using an oven to evaporate n-hexane. Heating with an oven is performed until a constant oil weight is reached. the theoretical value is then calculated as a comparison between oil mass and raw material times 100%. The essential oil content obtained in this research is at 18.2% (m/m). One research found that essential oil content in Piper cubeba is 10-18% dry weight [17], while another research discovered that value to be 12.5-20%. Therefore, it can be concluded that the result obtained here is almost the same as those of Heyne and Guenther.

Obtaining Essential Oil from Piper cubeba using Solvent-Free Microwave Extraction (SFME)
Effects of Microwave Power on Extraction of Essential Oil from Piper cubeba

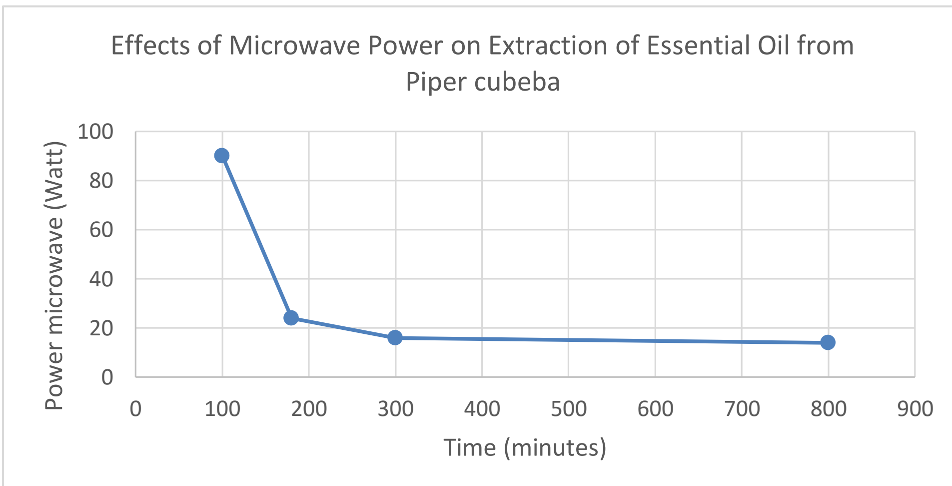

Figure 7. Effects of Microwave Power on Extraction of Essential Oil from Piper cubeba

Varied power used in this research is at a range of 100-800 Watt, or 800, 300, 180, and 100 Watt, to be precise. The experiment was performed at 800 watt in order to obtain maximum yield. It was found that after 14 minutes of treatment, the 100 g raw material, either grain or powder, started to burn. The reason for this is completely evaporated water molecule from the grain, which means the essential oil is properly extracted. Some literature suggest that essential oil evaporates at higher temperature (>150°C). This is the reason why the raw material starts to burn prior to evaporation of essential oil.

This research also used 300 watt power variation. It was found that after 16 minutes of treatment, the 100 g raw material in the microwave, either grain or powder, started to burn. At a variation of 100 watt microwave power, found that even after 1.5 hour, no essential oil was obtained. Another variation at 180 watt showed that after 24 minutes of treatment, the 100 g raw material in the microwave, either grain or powder, started to burn. Hence, the optimum microwave power for 100 g of raw material is 180 watt. At 300 watt, the raw material already burned even before essential oil was evaporated. In contrast, at 100 watt and treatment duration of 1.5 hour, water content in the raw material did not evaporate. This means that a longer duration is required to obtain the essential oil.

Effects of Grain Size on Extraction of Essential Oil from Piper cubeba

Varied grain sizes of 100 g weight were used for 800 watt microwave power. This varied grain sizes is meant to obtain maximum extraction yield. 100 g of Piper Cubeba grain extracted using SFME at 800 watt results in burned raw material after 8 minutes. Meanwhile, 100 g of Piper cubeba powder extracted using SFME at 800 watt results in burned raw material after 4 minutes. This shows that Piper cubeba powder allows for easy evaporation of essential oil, along with water, compared to its grain counterpart.

Effects of Duration on Extraction of Essential Oil from Piper cubeba

The effect of duration is studied at 180 Watt for 100 g of raw material, as can be seen in Table. More essential oil is obtained every 4 minute. In the first 4 minutes, obtained essential oil is at 0.8 mL. Meanwhile, the subsequent 4-minute intervals yield 0.8, 0.9, 0.9, 1.1, and 1.2 mL respectively.

Table 1. Obtained essential oil

Time (minutes)	Volum (mL)
4	0.8
8	0.8
12	0.9
16	0.9
20	1.1
24	1.2

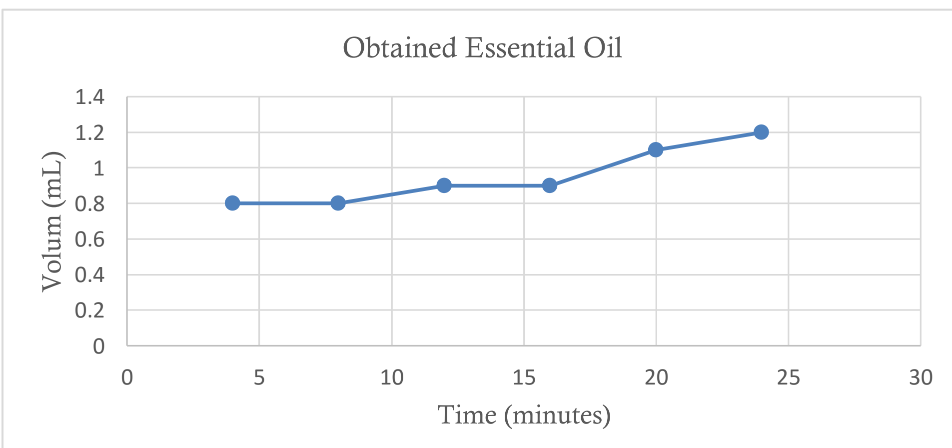

Figure 8. Oil volume versus time at 180 Watt power

Analysis of Density Physical and Chemical Properties of Essential Oil from Piper cubeba

Physical and chemical analyses of essential oil from Piper cubeba extracted using SFME method involves density and solubility tests. Density is obtained from discrepancy between the weight of empty bottles and those with oil in them, and then divided by droplet volume. A result of 0.86 g/mL is obtained, Keteren an earlier study suggests values between 0.911-0.919 g/mL. It shows that the essential oil extracted using SFME here is slightly lighter. This finding is supported by that of [18], suggesting that extraction with SFME method yields more oxygenated components.

In the meantime, testing for solubility was performed by mixing Piper cubeba with 96% alcohol at 1:1 ratio in a bottle with proper shaking as to allow for homogeneous content. Solubility testing is aimed at finding out sample purity based on its solubility in alcohol. Examination results showed that the sample appearance belong to category (b), with the sample being muddy and some oil is not dissolved. One study revealed that essential oil from Piper cubeba can be dissolved in alcohol at 1:1 ratio [19] In this research, essential oil is not fully dissolved in alcohol because some water was presence in the mix, that the alcohol did not dissolve the essential oil, but the water instead.

GC-MS Test

Results of GC-MS for essential oil from Piper cubeba using SFME method can be seen in Figure 9 and Table 2.

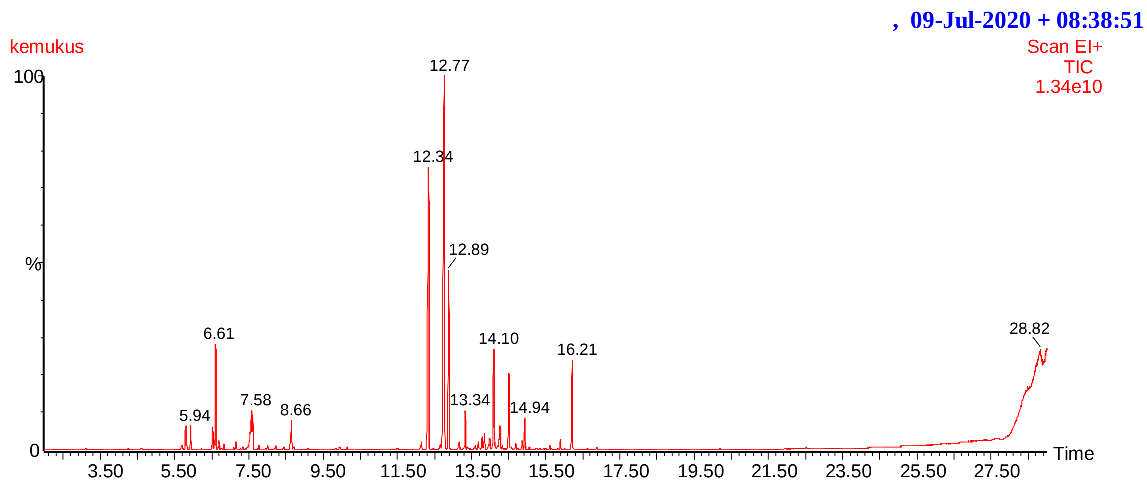

Figure 9. Chromatogram of Essential Oil from Piper cubeba

Table 2. GC-MS Results for Essential Oil From Pipper cubeba

IT	FT	RT	Area	Height	Concentration (%)
6.565	6.655	6.605	86515520	3733161984	5.81
12.258	12.403	12.338	354839808	10115657728	23.83
12.693	12.838	12.773	584889920	13391035392	39.28
12.838	12.948	12.888	173669808	6436752384	11.66
13.293	13.368	13.338	32186544	1365948544	2.16
14.028	14.163	14.098	99025296	3592855296	6.65
14.453	14.543	14.508	72948976	2696205824	4.9

16.109	16.284	16.214	84978424	3196828160	5.71

Results from GC-MS test show chemical compounds in the essential oil extracted from Piper cubeba. Detected chemical compounds are shown in Table 3.

Table 3. Chemical compounds of essential oil from Piper cubeba from GC-MS

Time	Area	Component
6.605	86515520	*β-Phellandrene*
12.338	354839808	*α-Cubebene*
12.773	584889920	*Copaene*
12.888	173669808	*Isoledene*
13.338	32186544	*Caryophyllene*
14.098	99025296	*Naphthalene*
14.508	72948976	*β-Cadinene*
16.214	84978424	*Asarone*

Table 4 shows quantitative components of chemical compounds in essential oil extracted from Piper cubeba using microwave-assisted hydrodistillation (MAHD) method.

Table 4. Quantitative comparison of chemical compounds from essential oil obtained from the experiment and reference

Chemical compound	Results	References
Cadinene	4.90%	-
Cubebene	23.83%	26.54%
Naphthalene	6.65%	25.85%
Azulene	-	10.39%
Copaene	39.28%	9.23%
Caryophyllene	2.16%	4.34%
Carene	-	1.45%
Asarone	5.71%	-
Isoledene	11.66%	-
Phellandrene	5.81%	-

GS-MS testing on the samples of this research revealed 8 components of essential oil from Piper cubeba; Copaene (39.28), Cubebene (23.83%), Isoledene (11,66%), Naphthalene (6,65%), Phellandrene (5.81%), Asarone (5,71%), Cadidene (4,90%), and Caryophyllene (2,16%), with most contents belong to the sesquiterpene group. found dominant contents of Cubebene (26.54%), Naphthalene (25.85%), Azulene (10.39%), Copaene (9.23%), Caryophyllene (4.34%), and Carene (1.34%). It can be seen in the Table that essential oil from Piper cubeba in this research has similar contents, with most contents belonging to the sesquiterpene group, despite differences in the composition of components. This means that quantitatively, the components of essential oil available in different Piper cubeba lead to different yields for each particular extraction. Different Piper cubeba treated with different methods results in different yield. This is only reasonable as compounds available in different Piper cubeba is affected by environmental conditions such as climate, weather, soil, rainfall, and elevation. Nonetheless, the chemical components found in most Piper cubeba are capable of healing throat infection and breathing difficulties.

4. Conclusion

Microwave power, material size, and extraction time of essential oil on the steamed seeds using the SFME method against yield, namely using an effective power of 180 Watts with a mass of 100 g of material, to achieve a stable extraction it takes 16 minutes to increase the volume obtained. While the test results of the Physico-chemical properties of the essential oil of cubeb seed oil, obtained data, among others, the yield of essential oil of 18.2%, density 0.86 g / mL. And the GC-MS test results obtained 8 largest components of essential oil of cubed seeds with the solvent-free microwave extraction method, namely Copaene (39.28), Cubebene (23.83%), Isoledene (11.66%), Naphthalene (6.65%).), Phellandrene (5.81%), Asarone (5.71%), Cadidene (4.90%), and Caryophyllene (2.16%) whose largest content belongs to the sesquiterpene group.

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