MICROWAVE ASSISTED EXTRACTION OF PATCHOULI ESSENTIAL OIL USING ETHANOL AS SOLVENT

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“Saya/Kami* akui bahawa saya telah membaca karya ini dan pada pandangan saya/kami* karya ini adalah memadai dari segi skop dan kualiti untuk tujuan Penganugerahan Ijazah Sarjana Muda Kejuruteraan Kimia.”

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DEDICATION

Thanks to ALLAH SWT for His Blessing…

Specially dedicated to my beloved family, lecturers and friends…

ACKNOWLEDGEMENT

First and foremost, thanks to Allah SWT for His guidance and blessing. This thesis cannot be done smoothly without support from everybody around me. In particular, I wish to express my sincere appreciation to my main supervisor, Mr Shawn Cheng Chin Kui for encouragement, guidance, critics and friendship.

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ABSTRACT

Patchouli or *pogostemon cablin* essential oil is extracted from the leaves of the patchouli plant. Patchouli is the important ingredient in about a third of modern, high-end perfume. It also has been claimed as an aramotheraphy material. In this project, the essential oil of patchouli will be obtained by microwave assisted extraction using ethanol as solvent. Because microwave extraction uses a combination of higher temperature, closed vessel, magnetic stirrer, and precise computer control, it can gives better extraction performance compared to the conventional method. It is a simple technique that provides a novel way of extracting soluble products into a fluid, from a wide range of materials, helped by microwave energy. From GC analysis, the main component of patchouli essential oil is patchouli alcohol. It existence is shown as a high peak in all of the analysis. Other components that presence in the essential oil is Caryophyllene, β-Patchoulene α-guaiene and α-Bulnesene. This mean, using microwave assisted extraction method for extract patchouli essential oil is feasible. It's also cut the extraction time and solvent usage compared to other conventional methods.

ABSTRAK

Pati minyak Patchouli atau *pogostemon cablin* diekstrak dari daun pokok patchouli. Patchouli adalah bahan yang penting dalam pembuatan minyak wangi. Ia juga dikatakan sebagai satu bahan aromaterapi. Dalam projek ini, pati minyak patchouli ini didapati dengan menggunakan kaedah pengekstrakkan dengan bantuan gelombang mikro dan etanol sebagai pelarut. Kerana kaedah ini menggunakan kombinasi suhu tinggi, balang tertutup, pengacau magnetik dan kawalan komputer yang tepat, ia dapat memberikan kualiti pengekstrakkan yang lebih baik dari kaedah lain. Ia adalah satu teknik mudah yang memberi hasil pengekstrakkan yang terbaik dalam mengekstrak bahan boleh larut kepada cecair, dari pelbagai bahan, dengan bantuan gelombang mikro. Dari analisis GC yang dijalankan, komponen utama dalam pati minyak patchouli ialah patchouli alcohol. Kehadirannya ditunjukkan sebagai puncak tertinggi di dalam kesemua analisis yg dibuat. Komponen lain yang hadir dalam pati minyak ini antaranya ialah Caryophyllene, β-Patchoulene α-guaiene and α-Bulnesene. Ini bermakna, kaedah pengekstrakkan dengan bantuan gelombang mikro adalah sesuai. Ia juga dapat menjimatkan masa dan juga kandungan pelarut yang digunakan jika dibandingkan dengan kaedah-kaedah lain.

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CHAPTER 1

INTRODUCTION

1.1 Background

Patchouli or *pogostemon cablin* essential oil is extracted from the leaves of the patchouli plant. Patchouli has a very heavy and camphoraceous odor. It is widely grown in West and East Indies and used primarily as perfume for centuries. Table 1.1 shows the scientific classification and it scientific name.

Table 1.1: Scientific Classification of Patchouli

Kingdom	Plantae
Division	Magnoliophyta
Class	Magnoliopsida
Order	Lamiales
Family	Lamiaceae
Genus	*Pogostemon*
Species	*P. cablin*

Nowadays, patchouli is the important ingredient in about a third of modern, high-end perfume, including more than half of perfume for men (Bauer *et al* 1997). It is also used as scent in household products such as paper towels, laundry detergent, and air fresheners. Despite its usage as a perfume in many of our daily products, patchouli has been claimed as an aramotheraphy material where some people claim that the scent of patchouli is very relaxing.

Patchouli oil is also used in medical as a cure to many illnesses. It is used to stimulate appetite, for diarrhea, and its balancing effect on the endocrine system. It also have a great effect to skin by reduces inflammations such as acne, eczema, athlete's foot, fungal infection and dandruff. Figure 1.1 shows a fully grown patchouli plant that is ready for harvesting.

Figure 1.1: Patchouli plant

The essential oil of patchouli is usually obtained by steam distillation (Xiao *et al,* 2004), a process which provides a relatively high yield of the oil. However, in this project, the essential oil of patchouli was obtained using microwave assisted extraction employing ethanol as solvent. Using this method, a better extraction performance was achieve compared to the conventional method. This is because microwave extraction uses a combination of higher temperature, closed vessel, magnetic stirrer, and precise computer control.

1.2 Objective

To examine the feasibility of microwave assisted extraction using ethanol solvent method as an alternative way for Patchouli essential oil extraction process.

1.3 Scopes of Study

In order to achieve the objective as stated above, the following scopes have been identified and to be studied:

1. To investigate the influence of power and extraction time to the yield of patchouli essential oil.
2. To study the major component of patchouli essential oil using GC analysis

1.4 Problem Statement

Steam distillation is the common method employed to extract patchouli essential oil. It uses steam or water to depress the boiling point of material, in this case patchouli. But this method requires lots of time to separate the essential oil from the waste. Water boil at temperature of 100ºC and it takes time to boil it to that temperature. Because of this, steam distillation use a lot of power to operate.

By using microwave assisted method, essential oil from patchouli become easy to extract. Microwave extractions can be completed within minutes—a fraction of the time, compared to conventional methods. Lower solvent usage reduces the costs of solvent purchase and disposal.

In this project, the experimental result of patchouli essential oil is compared to the standard result in term of their chromatogram result from analysis with gas chromatography-mass spectrometry (GC-MS). The standard was taken from the literature review from various kind of source to make sure the standard is the best.

CHAPTER 2

LITERATURE REVIEW

2.1 Patchouli

2.1.1 Introduction

Patchouli originally comes from East and West Indies. The word patchouli is derived from the Tamil *patchai* (green) and *ellai* (leaf). It come from *pogostemon* species of the family *Lamiaceae*. This fragrant herb, with soft, opposite, egg-shaped leaves and square stems, grows from two to three feet in height, giving out the peculiar, characteristic odor of patchouli when rubbed. The crop is cut two or three times a year, the leaves being dried and packed in bales and exported for distillation of the oil (Deguerry *et al.,* 2006).

Patchouli has a strong, earthy, musty and spicy smell, and is used for its base note in many perfumes with an oriental or exotic smell. The smell or aroma is reported to improve with age, and this attribute has been compared to that of a good wine improving as it matures. Patchouli has a deep reddish-brown color and is very similar to myrrh and benzoin oil in color and consistency, and is thick oil (Robbins, 1983).

In aromatherapy, patchouli is considered a great balancer, relaxing yet stimulating, particularly relevant for conditions of weak immunity where

overwork and anxiety have left the individual in a susceptible state. It is said to bring the three principal forces at work within the body - the Creative at the navel, the Heart center, and transcendental wisdom at the crown - into harmony. Due to its remarkable staying power, Patchouli is a great fixative ingredient (a fixative is a substance that binds other compounds, slowing down their evaporation and thereby making the fragrance last longer).

Patchouli essential oil is available by 10 mL (1/3 oz), 1oz, 4oz, 8oz and 16 oz. The pricing can be as low as RM 36.50 to as high as RM 460.00 according to the volume. (buyaromatherapy.com). This pricing is based on steam distillation process and using patchouli that are planted in Indonesia.

2.1.2 Component of Patchouli Essential Oil

Based on literature reading, there are five main components in patchouli oil including β-patchoulene, caryophyllene, α-guaiene, α-bulnesene (δ-guaiene) and patchouli alcohol were identified by using GC–MS. (Hu *et al*, 2005), (Tsai et al, 2006).

But the major constituent of patchouli oil is patchoulol or patchouli alcohol. It also known as patchouli camphor. It is responsible for the typical smell of patchouli.

Figure 2.1 show the chemical structure of patchoulol, the main component in patchouli oil. Figure 2.2, 2.3, 2.4 and 2.5 shows the chemical structure of Caryophyllene, β-Patchoulene, α-guaiene and α-Bulnesene respectively. Table 2.1 show the properties for the major components.

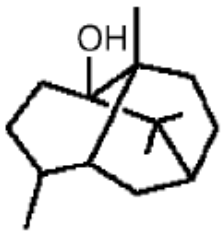

Figure 2.1: patchoulol

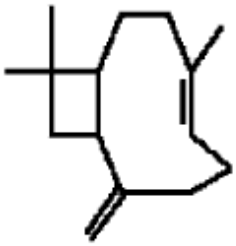

Figure 2.2: Caryophyllene

(Hu *et al.,* 2005)

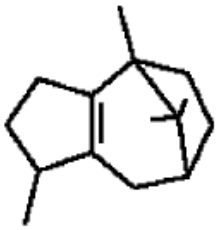

Figure 2.3: β-Patchoulene

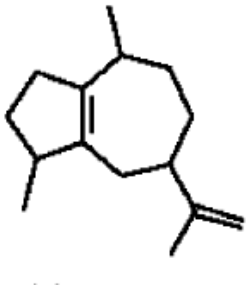

Figure 2.4: α-guaiene

(Hu *et al.,* 2005)

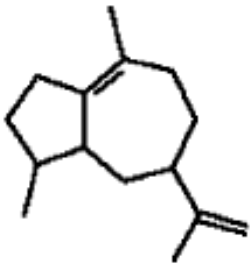

Figure 2.5: α-Bulnesene

(Hu *et al.,* 2005)

Table 2.1: Properties of Patchouli Essential Oil Main Component

Component	Formula	Molecular Weight	Boiling Point (^{0}C)	Density (g/cm^3)	Vapor Pressure (mmHg)
Patchoulol	$C_{15}H_{26}O$	222.3663	287.4	1.001	0.000278
Caryophyllene	$C_{15}H_{24}$	204.3511	268.4	0.89	0.0128
B-Patchoulene	$C_{15}H_{24}$	204.3511	260.7	0.95	0.0196
α -Guaiene	$C_{15}H_{24}$	204.3511	281.1	0.89	0.0062
α –Bulnesene	$C_{15}H_{24}$	204.3511	274.5	0.89	0.00904

2.2 Microwave assisted Extraction using Ethanol solvent

2.2.1 Microwave assisted Extraction

Microwave assisted extraction is one of the possible method to extract patchouli essential oil. This method uses a microwave applicator as the energy source during solvent extraction leading to: faster processing time, improved yield and quality, direct extraction capability, lower energy consumption, reduced solvent levels, and lower capital investment, when compared to conventional extraction methods. Microwave assisted extraction is a process that is based on direct application of electromagnetic radiation to materials such as organic solvent or plant tissue. It has the ability to absorb electromagnetic energy in this case microwave energy and transform it into heat (Leetellier and Budzinski, 1999).

The technique can be applied to both liquid phase extraction (when a liquid is used as solvent) and gas phase extraction (when a gas acts as extractant), (Luque *et al*, 1999). If practical, samples may be air-dried and ground to fine

powder prior to extraction. The technique can be applied to both liquid phase extraction and gas phase extraction (Lopez-Avila *et al*, 1994).

The advantages of using microwave energy, a non contact heat source, for the extraction of essential oils from plant materials, include: more effective heating, faster energy transfer, reduced thermal gradients, selective heating, and reduced equipment size, faster response to process heating control, faster start-up, increased production, and elimination of process steps (Metaxes *et al.*, 1993).

Compared with conventional methods like ultrasonic extraction and Soxhlet extraction, the advantages of using microwave extraction are reported to be higher recovery of analyte, shorter extraction time, and the use of smaller quantities of solvent, (Pastor *et al.*, 1997; Tomaniova *et al.*, 1998; Blanco *et al.*, 2000). Table 2.2 shows the comparison between microwave assisted extraction methods with other possible method.

Table 2.2: Comparison of Microwave Assisted Extraction with Other Available Methods

	SOXHLET	**SONICATION**	**MICROWAVE**	**S/CRITICAL FLUID**
Sample* Weight (gram)	5.00-10.00	5.00-30.00	0.50-1.00	1.00-10.00
Solvent	**	**	Hexane/Ethanol	CO_2
Solvent Volume (mL)	>300.00	300.00	10.00-20.00	5.00-25.00
Vessel Volume (mL)	500.00-1000.00	500.00	<100.00	5.00-25.00
Temperature (^{0}C)	Boiling Point	Room Temp.	40, 70, 100	50, 200
Time	16 hours	30 minutes	30-45 seconds	30-60 minutes
Pressure (atm.)	Ambient Atm.	Ambient Atm.	1.0-5.0	150.0-650.0
Relative Energy Consumption	1.00	0.05	0.05	0.25

* Dependent on the concentration and type of sample

* * Dichloromethane, acetone, hexane, cyclohexane, toluene, etc.

2.2.2 Component of microwave assisted extraction.

There are several important components in microwave assisted extraction. These parts are vital to make sure that the product will meet the customer standard.

The first part is pump and mixer. This is where raw material is added with solvent. Next part is the microwave itself. This is the core and the most important part of this system. Its provided appropriate temperature to heat the material to its boiling point. Figure 2.6 shows the simplify figure of microwave assisted extraction process and its basic component.

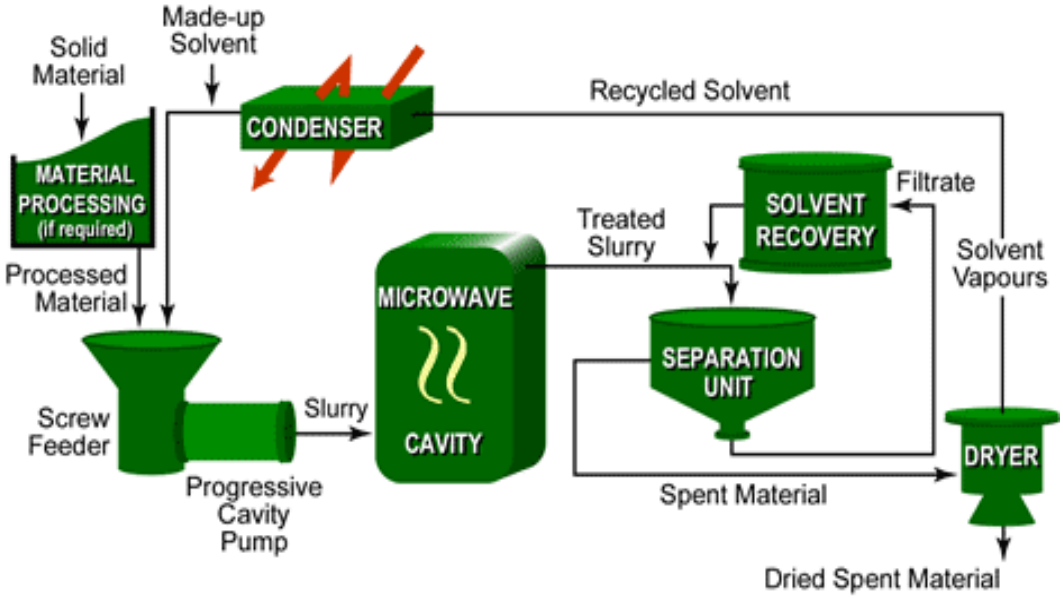

Figure 2.6: Flowchart of typical microwave assisted extraction process

Then there is a distillation tower to separate between the solvent and the product. Big companies have a big and high distillation tower to get high amount of product. These two components need a constant maintenance to ensure a good quality of products.

Condenser is also needed in this system to recycle any solvent from the separation process. This component will make sure any waste will not be wasted and still can be use in the process.

2.2.3 Ethanol solvent

Ethanol, also known as ethyl alcohol, drinking alcohol or grain alcohol, is a flammable, colorless, slightly toxic chemical compound, and is best known as the alcohol found in alcoholic beverages. In dilute aqueous solution, it has a somewhat sweet flavor, but in more concentrated solutions it has a burning taste. It is a group of chemical compounds, whose molecules contain a hydroxyl group, –OH, bonded to a carbon atom. Ethanol is classified as a primary alcohol, meaning that the carbon to which its hydroxyl group is attached has at least two hydrogen atoms attached to it as well. The chemistry of ethanol is largely that of its hydroxyl group.

Ethanol is used as a solvent in dissolving medicines, food flavorings and colorings that do not dissolve easily in water. Ethanol has boiling point of 78.4ºC which is lower compared to water boiling point (100ºC). With this physical property theres no doubt that ethanol as solvent is better than water solvent. Figure 2.7 shows a comparison between ethanol and other solvents available. It been different in term of their physical properties.

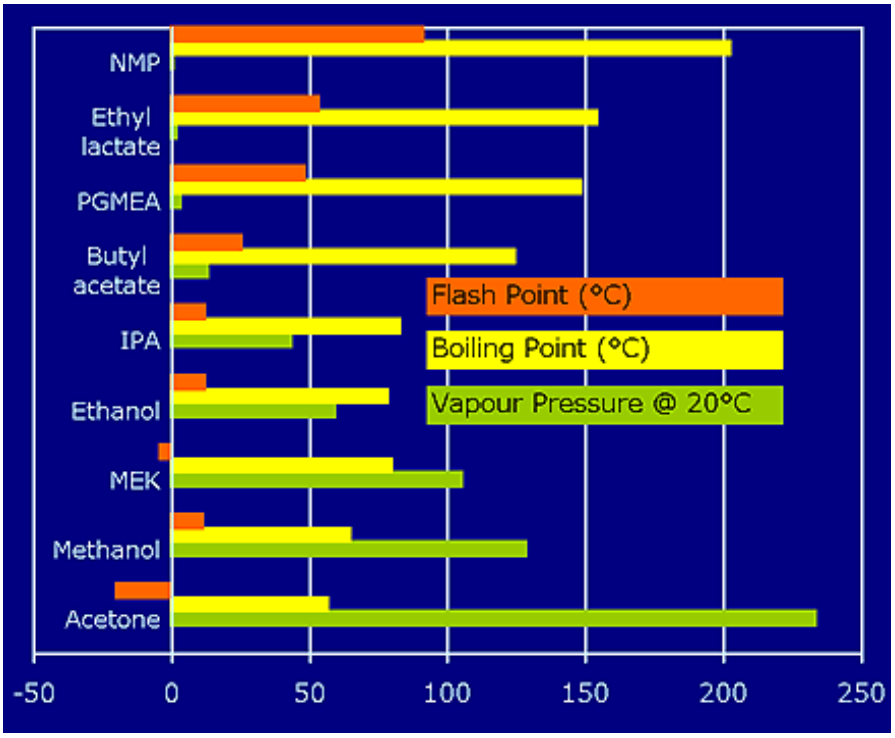

Figure 2.7: comparison of solvent physical properties

2.3 Other method of extracting essential oil

2.3.1 Steam Distillation

This is the common method of extracting essential oil. It is believe to be the only method to extract oils. This process should be easy to do, as long as the extraction process is closely monitored. This is to make sure the temperature will not exceed the standardize temperature of this process.

The process start when the sample is placed onto a vessel. Steam is added into the vessel and passed through the sample. Heat from the steam helps to open the pocket of the sample. This pocket contain its aromatic molecules or oils. When this pocket opened, it release the aromatic molecules and it rise along with the steam. The vapors carrying the aromatic molecules will then be cool using

cold water. It will then transform into liquid form as a result of condensation. This is product is known as the highly condensed patcouli essential oil. Figure 2.8 shows the overview of thestem distillation.

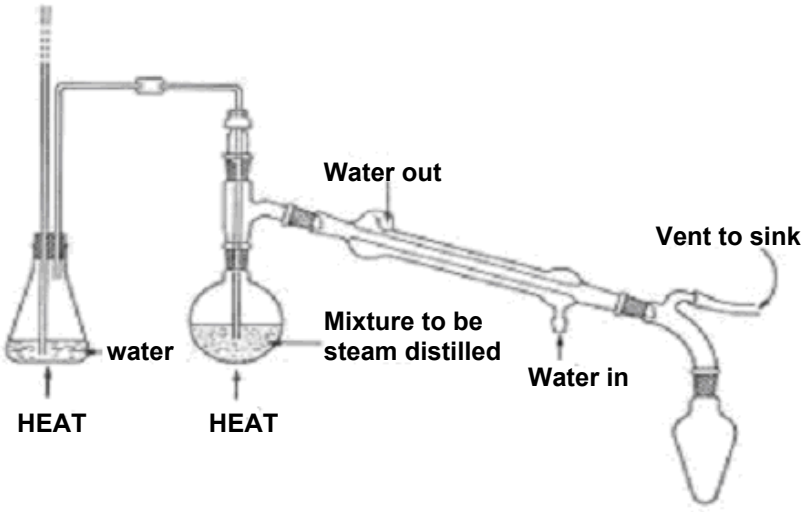

Figure 2.8: Main components in steam distillation

2.3.2 Water Distillation

This method is almost similar with steam distillation except it uses water instead of steam. Sample is placed in specialized equipment call still and then submerging it in water. The water is then brought to boil and the rest of the process is not much different from steam distillation.

The water in this process provides protection for the plant because it acts as a barrier. This method use less pressure and lower temperature compared to steam distillation. So water distillation is suitable for extract plants that cannot tolerate high heat.

2.3.3 Solvent Extraction

Plant sample sometime cannot stand high temperature when using steam distillation. High heat can damage this plant and once damage, their essential oil will also damage and can no longer can be extracted.

Sample is first washed by solvent such as ether, methanol, ethanol, hexane and other solvent. This process will dissolve the needed material including the aromatic molecules. The solvent mixture is then filtered and distilled using low pressure. Additional processing using alcohol helps extract the essential oil needed.

This is one of a very fast and effective essential oil extraction method. However, the residual solvents remains in the essential oil. Consequently, the essential oil becomes impure.

2.3.4 Supercritical CO_2 Extraction.

Super critical Carbon Dioxide (CO_2) is a new developed technic of extracting oil from plant. The final product is very concentrated and proven to be of high quality essential oil.

Carbon dioxide is an inert solvent which is non-reactive and doesn't form other type of chemical compounds. When the extraction process completes, the carbon dioxide will be returned back to gas phase and released to air. Therefore it doesn't leave any residual in the essential oil and pure essential oil can be obtain.

But this new technology is very expensive due to its sophiscated operation and control. Consequently, it is not yet widely used.

2.3.5 Cold Press

This is an alternative way to extracting essential oil from plant. This ancient method uses mechanical pressure to force the oils out from the leaves. The essential oil extracted with this method contain water, but eventually this water will evaporate, leaving just the essential oil. The essential oil that extracted by using this method spoil more quickly than other methods.

2.3.6 Ultrasonic Distillation

Ultrasonic distillation is the newest method for being used in extracting essential oil. It can be applied in distillation process to reduce the time consuming in the reaction and to increase the percentage of yield of essential oil.

Ultrasonic enhancement in distillation will improve the mixing and chemical reactions in the plant sample that will go through the process. It will generate low pressure and high pressure waves in liquid, leading the formation and violent collapse of small vacuum bubbles that was called cavitations that causes high speed impinging liquid jets and strong hydrodynamic shear-forces.

It is use as an alternative to high-speed mixers and agitator bead-mills. . Ultrasonic gives a benefited in chemical reactions from free radicals created by cavitations that will leads to a substantial reduction in reaction time.

CHAPTER 3

METHODOLOGY

3.1 Introduction

Microwave assisted extraction is a simple technique that provides a novel way of extracting soluble products into a fluid, from a wide range of materials, helped by microwave energy. It offers a rapid delivery of energy to a total volume of solvent and solid plant matrix with subsequent heating of the solvent and solid matrix, efficiently and homogeneously. Because water within the plant matrix absorbs microwave energy, cell disruption is promoted by internal superheating, which facilitates desorption of chemicals from the matrix, improving the recovery of nutraceuticals (Kaufmann, Christen, & Veuthey, 2001).

MAE has been considered as a potential alternative to traditional solid-liquid extraction for the extraction of metabolites from plants. It has been used to extract patchouli because it can reduced extraction time, reduced solvent usage and improved extraction yield.

3.2 Sample Preparation

Plant sample used in this experiment were patchouli leaves. It easily can be bought through local supplier and it price is affordable because patchouli plant is easy to found. This project used the patchouli leaves that have been planted in the university area.

3.2.1 Drying

Patchouli leaves left dried to prevent other moisture to distract the outcome of the experiment. It done by two ways; either exposes it to sunlight or by the usage of dryer until it completely dry. In this research, patchouli leaves were dried by placing it in desiccators for two days.

3.2.2 Grinding

The dried patchouli leaves were grinded using blender and ground to a fine powder prior to extraction. To maximize the contact area between the particles and the solvent, it grinded until the particle size is less than 1mm. Finely ground sample were more easily extracted than large particles.