EXTRACTION OF GAHARU ESSENTIAL OIL USING ULTRASONIC ASSISTED DISTILLATION

ZUBAIR BIN MAT ISA

UNIVERSITI MALAYSIA PAHANG

UNIVERSITI MALAYSIA PAHANG

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EXTRACTION OF GAHARU ESSENTIAL OIL USING ULTRASONIC ASSISTED HYDRODISTILLATION

ZUBAIR BIN MAT ISA

A Thesis submitted in fulfillment of the requirement for the award of the degree of Bachelor of Chemical Engineering

Faculty of Chemical Engineering and Natural Resources
Universiti Malaysia Pahang

MAY 2008

“ I declare that this thesis is the result of my own research except as cited references. The thesis has not been accepted for any degree and is concurrently submitted in candidature of any degree.”

Signature : ………………………………

Name of Candidate : ZUBAIR MAT ISA

Date : ……………………………...

DEDICATION

Dedicated to my beloved father, mother and brothers……

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First of all, thanks to Allah s.w.t for all His guidance and blessing through all the hardship encountered whilst completing this thesis. In preparing this thesis, I was in contact with many people, researchers, academicians and practitioners. They have contributed towards my understanding and thoughts. In particular, I wish to express my sincere appreciation to my main supervisor, Miss Mazni Ismail and Mrs. Zuraida Ali for encouragement, guidance, critics and friendship.

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ABSTRACT

Gaharu is one of the most highly valuable, non-timber products harvested from tropical forest that produces from the *Aquilaria* species trees naturally or mechanically. Its essential oil is used in varies industries as perfumes and also been used in religious occasion for centuries. The gaharu that was used in this research is grade C came from the peninsular of Malaysia and known as ‘karas’ among the locals. Recently, the gaharu essential oil traditionally extracted using hydro distillation. However, the method is not effective in producing the higher yield of oil and it is consumed a lot of time. The ultrasonic assisted hydro-distillation is introduced to enhance the process where the gaharu sample is put into an ultrasonic bath before distillate. The parameter that has been studied in this research are extraction time which are 1 hour, 3 hours, 6 hours and 9 hours, and the solid to solvent ratio which are from 1:8, 1:12, 1:16 and 1:20. Solvent been used in this research are water. From the result, it showed that the gaharu oil yield is increase along with the increase of extraction time and solid to solvent ratio and it is proved that this method are feasible to extract the gaharu oil. From the results, the highest oil yield percentage for solid to solvent ratio is for 1:20 which is 0.139 %. For extraction time, the highest percentage oil yield is for 9 hours which is 0.169 %.

ABSTRAK

Gaharu merupakan sumber yang paling berharga dan banyak terdapat di hutan tropika di mana ia dihasilkan daripada spesies Aquilaria secara semulajadi atau mekanik. Penggunaan gaharu tidak terhad di mana ia digunakan sebagai minyak wangi, aromaterapi dan ia juga digunakan dalam upacara keagamaan sejak berkurun lamanya. Gred gaharu yang digunakan dalam kajian ini adalah dari gred C yang diperolehi dari semenanjung Malaysia di mana ia di kenali sebagai 'karas' di kalangan penduduk tempatan. Dewasa ini, pati minyak gaharu diekstrak secara tradisional menggunakan penyulingan hidro. Walau bagaimanapun, kaedah ini kurang efektif dalam menghasilkan pati minyak dan memakan masa yang lama. Penyulingan dengan bantuan ultrasonik telah diperkenalkan bagi memperbaiki proses ini di mana sampel gaharu diletakkan di dalam bekas ultrasonik sebelum disulingkan. Parameter yang dikaji ialah masa pengestrakan iaitu dari 1 jam, 3 jam, 6 jam dan 9 jam dan juga nisbah gaharu kepada pelarut iaitu air di mana nisbahnya adalah 1:8, 1:12, 1:16 dan 1:20. Daripada keputusan ujikaji yang telah dijalankan, menunjukkan bahawa peratusan minyak gaharu yang diperolehi meningkat berkadar langsung dengan peningkatan masa pengestrakan dan juga nisbah gaharu kepada pelarut. Ini menunjukkan cara pengestrakan ini adalah sesuai untuk dipraktikkan dalam proses pengestrakan gaharu. Berdasarkan keputusan eksperimen, bagi nisbah gaharu kepada pelarut, peratus minyak paling tinggi ialah 0.139 % untuk 1:20 manakala bagi masa, peratus paling tinggi ialah 0.169 % untuk 9 jam pengestrakan.

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CHAPTER 1

INTRODUCTION

1.1 BACKGROUND

Agarwood is a mystical and precious resinous wood formed in heartwood of *Aquilaria* tree, an archaic tropical evergreen tree which came from countries of Southeast Asia. It is called Candan or Gaharu in the Malay language where it refers to the resin produced by the trees from the *Aquilaria* species. Apart from *Aquilaria* genus, there are also six genera that can probably produce Agarwood: *Gyrinops, Gonystylus, Aetoxylon, Phaleria, Wikstroemia,* and *Enkleia*. The taxonomy of Gaharu is shown on Table 1.1 below.

Table 1.1 The Taxonomy of Gaharu Tree

KINGDOM	Plantae
PHYLUM	Tracheophyta
CLASS	Magnoliopsida
ORDER	Myrtales
FAMILY	Thymelaeaceae
GENUS	*Aquilaria*
SPECIES	*malaccensis*
SPECIES AUTHORITY	Lamk.

(Source: International Union for Conservation of Nature and Natural Resources.)

The popularity of Gaharu was spread worldwide and well-known in some countries with other names like in Chinese they called it “Chen Xiang”, “Jin-Ko” in Japanese and “Oud” in Arabic. Gaharu also known to be used since the Pharaoh in Egypt and it is used in religious ceremony and as perfumes. In Malaysia, Gaharu have been a national treasure the type have been found is *Aquilaria Malaccensis* and can be found rarely in the nation rainforest because of it’s decreasing in population. The essential oil produced from the tree contains high value of oil and it is mentioned that the values are more worth than gold (Mohd Haikal Mohd Isa, 2006).

The Aquilaria tree generally grows up to 40 meters high and 80 centimeters in diameter. The formation of Gaharu was naturally produces by becoming randomly infected with some parasite fungi or molds and begins to produce Gaharu in heartwood due to immune response to this attack. But this biologic process develops very slowly over several tens of years. Not all Aquilaria trees produce agarwood, only approximately 10% of wild mature Aquilaria tree (from the age of 20 years onwards and with above 40 cm diameter at breast height) can naturally produce resin. The ecological interaction between the host tree and the fungi or molds in order to produce Agarwood is poorly understood. Other factors such as the age of the tree, differences in the tree caused by seasonal variation, environmental variation and genetic variation of Aquilaria trees may also play an important role in agarwood formation.

Recently scientific research have shown that Gaharu formation can occur in cultivated trees as young as four years of age by using inducement or wounding technique. After wounding, by drilling in tree-trunk, a bio-agent consisting of special isolated fungi and biochemicals are put into the drillhole. After 20-24 months a dark brown resinous wood part was observed around injury sites. Accurate estimation of the quality and yield of such “Gaharu” is still a pending question.

Figure 1.1 below show sample of Gaharu tree that commonly found in the rainforest in Malaysia.

Figure 1.1 ***Aquilaria malaccensis*** **Plantation in Malaysia**

(Source: Natural Perfumery And Health.)

In Malaysia, the Malaysian Institute of Nuclear Technology (Mint) has applied nuclear irradiation technology to mass-produce plantlets via tissue culture. Seeds were screened for fast-growth and single-bole characteristics at the cellular level claimed that a secret formula was found after one year of experimentation (Hilary Chiew, 2006).

Majority, most commonly method of genuine essential oils are extracted by distillation. The production of gaharu essential oil has been a cottage industry in East of Peninsular Malaysia and also traditionally distilled by the indigenous people mainly by using water distillation or hydro distillation. It is the oldest form of essential oil extraction. The process operate with the resin is chipped, dried, ground into powder and soaked for a week before being boiled in vats with the resultant steam being captured and condensed to separate between oil and water (Hilary Chiew, 2005).

1.2 PROBLEM STATEMENT

Nowadays, the most popular method to extract gaharu essential oil is the traditional hydro distillation method. This method involves submerging the desired by using raw material (gaharu chips) in water in the still and brought to boil, and the steam produced is collected and condensed to get the essential oil.

This extraction method acquires long extraction times almost 96-hour of distillation that consume a lot of fuel for heating purposes. The extraction process did not produce the maximum yield of oil because the efficiency of the method itself is relatively low and because of this, the process need a lot of material from the plant to be extracted to get the required yield of oil. Thus, this will increase the operating cost and the long time consuming is needed.

To improve this lack ness some enhancement are need to be made such as adding some active enhancement like an ultrasonic to the distillation process to reduce the extraction time and to produce the higher yield of the oil.

1.3 OBJECTIVES

- To examine the feasibility of ultrasonic assisted extraction as an improved method for gaharu essential oil extraction process.

1.4 SCOPE OF RESEARCH

In order to achieve the objective, these following scopes have been identified and to be applied:

- The effect of pretreatment time on the gaharu essential oil yield.
- The effect of solid to solvent ratio on the gaharu oil yield

CHAPTER 2

LITERATURE REVIEW

2.1 Gaharu

2.1.1 Gaharu Population

Gaharu is known throughout many Asian countries. This large evergreen tree was found in primary or secondary forest up to 1000 m. In Malaysia, the species can be found up to 750 m on plane, hillsides and ridges in primary and secondary forest (Normahwati, 2005).

Populations are widespread but patchy in distribution in Indonesia and Malaysia. In Malaysia, estimates lie at 2.5 individuals per hectare (Soehartono, 2000). As the most important source of gaharu population are heavily exploited throughout the species range, only 10% of the trees in any population are likely to be infected with the fungus that causes the wood to decay, producing gaharu. Traditionally, local people have harvested only infected trees but demand in the last ten years has led to excessive harvesting of both diseased and healthy trees (Soehartono, 2000).

There is even a belief that the diseased wood developed in fell trees. The increasing rarity of the species has led to traders searching for populations in more remote areas and in some cases outside the species range (Normahwati, 2005).

2.1.2 Formation of Gaharu Resin

Gaharu formation investigation was first initiated in 1926 by Bose but it is uncertain whether the fragrant wood result from fungal infection which brings about pathological conditions, certain chemical changes in the tree, or environmental factors. Research conducted so far focused mainly on pathological and non-pathological conditions (L.T. Ng, Y.S. Chang and Azizol Abdul Kadir, 1996).

Commonly, gaharu formation is caused by the tree response to mechanical or natural injury associated with the wood. In brief, the tree has two response mechanisms to injury. The first line of defense is for the phloem cells to produce callus growth over the injury. If the formation of callus prevented then the tree will produce resin as a chemical defense to the injury (Normahwati, 2005). The fungus that probably infected in gaharu formation is *Epicoccum granulatum* or *Cytosphaera mangiferae* (L.T. Ng, Y.S. Chang and Azizol Abdul Kadir, 1996).

The degree to which the resin saturates the heartwood phloem fibers determines the market value of this product. In lesser quality specimen, the resin create a mottled or speckled appearance in the naturally pale wood, but higher quality specimen are nearly solid in color, glossy and black (Donovan *et al.*, 2004). Figure 2.1 below showed the method on producing gaharu resin.

Figure 2.1 Method of Producing Gaharu Resin

(Source: Yip and Lai, Hong Kong Herbarium, 2005)

2.2 Grading and Prizing of Gaharu

Grading gaharu is a complicated process and very laborious process that needs time consuming and it is an important process in order to optimize its financial returns. Grading process are based on the size, color, odor, shape, weight, density, and even flammability, irregular shapes with angled features fetch more than regular shaped pieces because of the greater ease of lighting them to burn (Normahwati, 2005). The application of grade codes are shown in Table 2.1 below varies between buyers (Ahmad Fadzli, 2006).

Table 2.1 Gaharu Price per Kilogram in May 2001

Gaharu Grade	Price per Kilogram (USD)
A	341
B	237
C	172
D	111

(Source: Zich *et al.,* 2001)

The resin of gaharu usually tested by igniting the wood and smelling the odor while watching for bubble of the burning resin. In a bigger scale of the gaharu resin, water test are usually being used by separating the pieces that float because of the lower resin content from those that sink that have a high resin content and better quality. After drying, the pieces are graded based on colour and sizes (Zich *et al.,* 2001).

2.3 Gaharu in Malaysia

Gaharu has been recognized by the local Malaysian since a long time and its valuable oil has been collected and extracted traditionally as a 'backyard industry' by the local people. Nowadays, Malaysian extracting the essential oil of gaharu by using distillation unit made from stainless steel vat as a container that contains ground-up gaharu that will undergo a 96 hour distillation process to get its essence. High quality gaharu can fetch RM10,000 per kg depending to the grade of the resin. A 12 g bottle of oil is sold at between RM50 and RM200 (Hilary Chiew, 2005).

Malaysia has been known as a country that produce gaharu. According to TRAFFIC, in 2000, it is estimated that nearly 700 tonnes of gaharu were produced in the international market mostly came from the jungle of Malaysia and Indonesia. The price is estimated at least RM 3.5 billion. Gaharu has been Malaysian natural treasure because of its rarity and its high value (M. Haikal, 2006). In Malaysia, there is a report that gaharu can be found in heart of Kelantan, Perak, Pahang, Terengganu jungle even though it is a rare species (Hilary Chiew, 2005). Figure 2.2 shown the distillation process of gaharu in Malaysia.

Figure 2.2 Gaharu oil extraction process

(Source: All Malaysia Info, The Star Online, 2005)

As it is a rare species, hard to found, and because of its high value, the federal Forestry Department has urged the state governments to regulate the collection, trade and processing of gaharu through a licensing system where the Gaharu collectors or buyers have to pay a royalty fee amounting to 10% of the raw material market price and an extraction permit is issued and this will facilitate the traders in obtaining export and CITES (Convention on International Trade in Endangered Species) permit (Hillary Chiew, 2005).

2.4 Essential Oil

2.4.1 Physical Properties of Essential Oil

Essential oil also referred to as "essence" are botanical extracts of various plant materials such as leaves, flowers, roots, buds, twigs, rhizomes, heartwood, bark, resin, seeds and fruits. Essential oils are found in special secretory glands or cells within plant life. The aromatic substances are formed and stored in certain organs of a plant as a by-product or because of its metabolism. The combinations of the plant's very own unique blueprints, the energy of the sun, soil, air and water gives each of the oil its individual perfumes and beneficial healing properties.

Essential oil is not oily compared with vegetables oil expressed in nuts and seed for they are not fat-based oil. Some essential oils are viscous; others are fairly solid and most are watery. They have lipid-soluble molecular structure which allows them to pass easily through the skin. They penetrate into the fat layers of the skin quickly which is why massage is such an effective treatment.

Essential oil provides a concentrated dose of nature's vast pharmacological active ingredients in a single drop of oil. Hundred percent of essential oil are distinguished by remarkable diversity of substances that only nature could produce. Essential oil also volatile, they easily evaporate into air. In addition, essential oil is sensitive to heat and light. There are also certain essential oil that can be toxic and public awareness and proper use is required when using it for the first time or experiment it with other oil.

2.4.2 Physical Properties of Gaharu

2.4.2.1 Colour and Scent

Traders are look for blackened, resinous and aromatic gaharu that believe has a higher grade and resin content (F. Zich, J. Compton, 2001). The colour mention for gaharu is green, dark green, yellow, golden, red, black, brown and white. The scent of gaharu also affecting the grade of gaharu which a softer scent is consider as a higher grade and higer price than those with more intense scent.

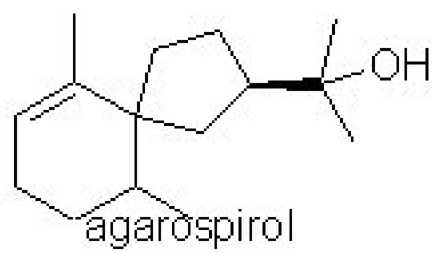
2.4.2.2 Size and Form

For the pieces of the same level of grade of gaharu, the larger pieces carrying a higher value than the smaller pieces according to their respective weight.

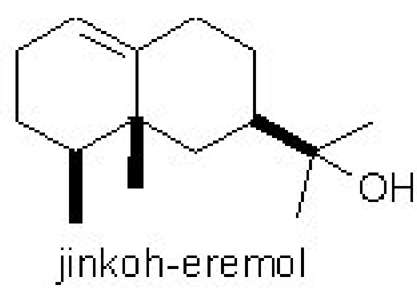
2.4.3 Chemical Component of Gaharu

Gaharu contains a sesquiterpene alcohol which produces its characteristics aroma. The component in gaharu is depending on the types of species of the wood respectively. It was reported that 2-[2-(4-methoxyphenyl)ethyl]chromone and 2-(2-phenylethyl)chromone through pyrolysis at 150 °C produces 4-methoxybenzaldehyde and benzaldehyde respectively and this molecules are odourless at room temperature but produce a long lasting fragrance upon burning (L.T. Ng, Y.S. Chang and Azizol Abdul Kadir, 1996).

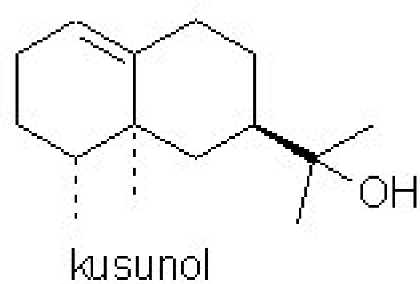
There are three major components that may contribute to the characteristic of aroma in gaharu. The major components are agarospirol, jinkohol-eremol and kusenol (Nakanishi *et al.,* 1984, Ishihara *et al.,* 1993).Chemical profile for each gaharu grade such as A, B, and C were different. In peninsular of Malaysia, the gaharu were mostly of grade C quality. Figure 2.3 shown some chemical component structure in gaharu essential oil.



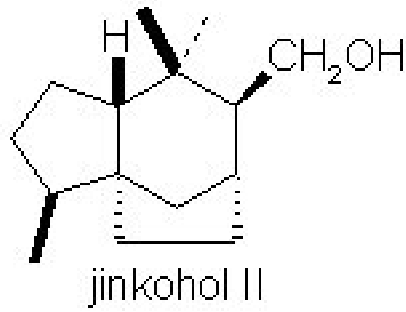
IUPAC: 2-(6,10-dimethyl-2-spirol[4.5]dec-9-enyl)propan-2-ol



IUPAC: 2-(8,8a-dimethyl-2,3,4,6,7,8-hexahydro-1H-elicoids-e-2-yl)propan-2-ol



IUPAC: 2-(2,4-dihydroxyphenyl)-3-7-dihydroxy-8-(5-elicoid-5-methyl-2-prop-1-en-2-yl-hexyl)-5-methoxy-chroman-4-one



MF: $C_{15}H_{26}O$

Figure 2.3 Chemical component structures in gaharu essential oil.

(Source: www.equitech.biz)

2.5 Extraction

2.5.1 Definition of Extraction

Extraction is a selective transfer of a compound or compounds from liquid (usually water) to another immiscible liquid (usually organic) or from a solid to a liquid. It is a separation process to separate the desired solute or removed undesirable solute component from the solid where the solid is contact with the liquid phase with an intimate contact and the solute can diffuse from the solid to the liquid phase.

Fragrance extraction are process which involve extracting aromatic compounds from the raw materials using various methods such as distillation, solvent extraction, expression, or enfleurage. These techniques will distort the odour of the aromatic compounds obtain due to the use of heat, harsh solvents, or through exposure to oxygen which will denature the aromatic compounds either change their odour character or renders them odourless.

2.5.2 Types of Extraction Method

Nowadays, there are many types of extracting essential oil that has its very own advantages and disadvantages depend on the material that needs to be extracted.

2.5.2.1 Hydro Distillation

Hydro distillation is the most popular process in extracting the essential oil in gaharu. In hydro distillation, the aromatics materials are fully submerged in water, producing a “soup”, the steam of which contains the aromatic plant molecules. These methods are most often used in primitive countries because of its versatile and an ancient method in separating substances.

The resultant steam that contains the aromatic plant molecules is being captured and condensed. The oil will normally float on top of the ‘hydrosol’ (the distilled water component) and may be separated off. When the condensed material cooled down, the oil and hydrosol is separated and the decanted oil to be used as essential oil. These method has its risk is that the still can run dry or be overheated, burning the aromatics and resulting in an essential oil with a burnt smell. This method is also a time consuming process and its need a large amount of plant material to operate it.

2.5.2.2 Steam Distillation

Steam distillation is done in a still. Fresh or sometimes dried, botanical material is placed in the plant chamber of the still, and pressurized steam is generated in a separate chamber and circulated through the plant material. The heat of the steam forces the tiny intercellular pockets that hold the essential oils to open and release them. The temperature of the steam must be high enough to open the pouches, yet not so high that it destroys the plants or burns the essential oils. As they are released, the tiny droplets of essential oil evaporate and, together with the steam molecules, travel through a tube into the still's condensation chamber.

As the steam cools, it condenses into water and the essential oil forms a film on the surface of the water. The film is then decanted or skimmed off the top to separate the essential oil from the water. The remaining water, a byproduct of distillation, is called floral water, distillate, or hydrosol. It retains many of the therapeutic properties of the plant, making it valuable in skin care for facial mists and toners. In certain situations, floral water may be preferable to pure essential oil, such as when treating a sensitive individual or a child, or when a more diluted treatment is required.

2.5.2.3 Spinning Band Distillation

Spinning band distillation has two main types which is Teflon and metal. The most common is Teflon where it has a maximum of 50 theoretical plates at atmospheric pressure. This method suitable for distilling solvents that can be distilled up to 225 °C but Teflon does not suitable above this temperature because it becomes soft and can come apart under the spinning force.

If the temperature of the boiler going above 225 °C a metal spinning band can be used. The most common metal used is Monel, a stainless steel with a high content of molybdenum to maximize corrosion resistance.

2.5.2.4 Ultrasonic Distillation

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

Ultrasonic enhancement in distillation will improve the mixing and chemical reactions in the material plant that will undergo the process. It will generates alternating 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. This process will be use for the disintegration of cells or the mixing of reactants.

This active enhancement 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.

The enhancement by ultrasonic cavitation is mainly contributed by acoustically excited bubble break-ups on the membrane surface. Those bubble break-ups result in the hydraulic pressure impulses by the impinging actions similar to the water hammer effect. The hydraulic pressure impulses further lead to a corresponding impulsive elevation of vapor partial pressure in the dissolved vapor phase. As a result, an increase in vapor pressure difference over the membrane is obtained provided that the vapor pressure on the other side of the membrane remains unchanged. Consequently, the vapor permeated through the membrane is increased or enhanced (C. Zhu, 2000).

Figure 2.4 below showed the ultrasonic irradiation for modeling system on the porous membrane for the membrane distillation.

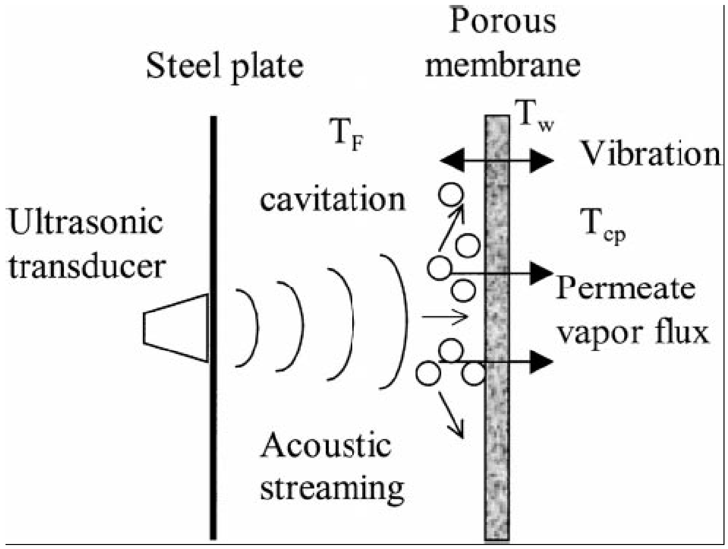


Figure 2.4 Ultrasonic irradiation medolling systems

(Source: Chao Zhu, 2000)

2.5.2.5 Carbon Dioxide Extraction

Supercritical carbon dioxide extraction uses carbon dioxide under extremely high pressure to extract essential oils. Plants are placed in a stainless steel tank and, as carbon dioxide is injected into the tank, pressure inside the tank builds. Under high pressure, the carbon dioxide turns into a liquid and acts as a solvent to extract the essential oils from the plants. When the pressure is decreased, the carbon dioxide returns to a gaseous state, leaving no residues behind.

Many carbon dioxide extractions have fresher, cleaner, and crisper aromas than steam-distilled essential oils, and they smell more similar to the living plants. Scientific studies show that carbon dioxide extraction produces essential oils that are very potent and have great therapeutic benefits. This extraction method uses lower temperatures than steam distillation, making it gentler on the plants. It produces higher yields and makes some materials, especially gums and resins, easier to handle. Many essential oils that cannot be extracted by steam distillation are obtainable with carbon dioxide extraction.

CHAPTER 3

METHODOLOGY

3.1 Introduction

In this research the ultrasonic assisted extraction is used to extract gaharu oil. This method of distillation will enhance the recent process of extracting gaharu and will reduce the time consuming of extraction and will get a higher yield of gaharu essential oil. The technique of ultrasonic will assisted the process by produces the ultrasonic cavitation that causing the disintegration of gaharu cells that leads the releasing of the valuable odour of gaharu. This method will also reduce the extraction time. There are two parameters that are going to be studied in this research which is the extraction time and the solid to solvent ratio. Before running the process, there are pretreatment processes which are drying, grinding and soaking.

3.2 Sample Preparation

The extraction of gaharu has a selectivity of the grade of gaharu to be extracted. Gaharu wood has a three main grade that is grade A, B and C (Chang Y.S. *et al.,* 2002). Grade A and grade B are the highest in the gaharu level because it quality and demand for it (Hilary Chiew, 2005). In this process, grade C gaharu is used because it is cheaper and reduce the cost of process.

3.2.1 Drying

Drying process need to be done with the chips of gaharu are putted into dryer so that the chips are completely dry from any moisture and also to get rid any substance that can distract the impurities of oil and to avoid any blockade in grinding.

3.2.2 Grinding

The grinding process is done with the dry sample of gaharu are ground into sawdust to get the particle size of 1 mm as a purpose to maximize the contact area between the gaharu particle and the solvent.

3.3 Extraction Process

A mixture of 500 grams of readily soaking gaharu and four liters of dionized water is put into an ultrasonic bath. The ratio of solid to solvent is 1:8. The sample is sonicated in ultrasonic cleaner for treatment time that has been set which is one, three, six, and nine hours. Then the mixture going through a cooling step and the raw extract was transferred to a flask to running the distillation process. Then the vaporized mixture is condensate and the effluent is collected in a receiving vessel and collected in a sample bottle. A small amount of hexane is pipette into the receiving vessel functioning as a medium to separate the oil from water. The distillation process is run for three days. Then the experiment is repeated with the manipulate variable of solid to solvent ratio of 1:8, 1:12, 1:16, and 1:20. The time for distillation process is constant that is three days. Figure 3.1 show the ultrasonic cleaner.

Figure 3.1 Ultrasonic Cleaner

3.3.1 Data Collecting

The yield of essential oil is calculated using equation below:

$$\text{Oil Yield (\%)} = \frac{\text{Weight of Oil Extracted (g)}}{\text{Dry Weight Sample (g)}} \times 100$$

Table 3.1 below show the experimental matrix for ultrasonic assisted distillation.

Table 3.1 The Experimental Matrix for Ultrasonic assisted distillation

VARIABLE	EXPERIMENT	MASS OF SAMPLE (kg)	SOLID TO SOLVENT RATIO (w/v)	TREATMENT TIME, t (hour)
SOLID TO SOLVENT RATIO (w/v)	R1	0.50	1:08	3
	R2	0.30	1:12	3
	R3	0.25	1:16	3
	R4	0.20	1:20	3
TREATMENT TIME, t (hour)	T1	0.50	1:08	1
	T2	0.50	1:08	3
	T3	0.50	1:08	6
	T4	0.50	1:08	9

Figure 3.2 below show the summary of methodology of the process;

1. Grind the gaharu wood by using grinder into sawdust with size of 1 mm.
2. Weight 500 g sawdust of gaharu wood as the sample.
3. Put the mixture of 500 g sawdust of gaharu wood and 4000 mL of dionized water into a flask of distillation unit.
4. Put the sample in ultrasonic bath depend on its time duration which is 1, 3, 6, and 9 hours.
5. Set up the apparatus of distillation and the sample from ultrasonic bath is put into distillation unit.
6. Run experiment for 3 days before collect the essential oil.
7. Repeat the experiment with solid to solvent ratio of 1:08, 1:12, 1:16, and 1:20 with the treatment time of 3 hours respectively.

Figure 3.2 Summary of Flow Method of the Experiment

Figure 3.3 below show the set up of distillation unit;

Figure 3.3 The distillation unit

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Introduction

In this research, the ultrasonic distillation method is developed to enhance the previous method of extracting the gaharu essential oil. This method is also to get the alternative method that is more effective and produce a higher yield of essential oil. In the process, the consuming time for the process is expected to be shorter than other extraction method; the yield that produced is higher because it is improved the recent distillation with the ultrasonic. The oil yield will increase along with the increasing time until it reaches a time where its oil yield increment will be constant. It is because the entire sample has been completely extracted. The solid to solvent ratio affecting the oil yield with increasing oil yield due to increase of the solid to solvent ratio. The higher solvent ratio that been used, the higher product obtained (Bulyamin, 2005).

In this experiment, hexane is used in the receiving vessel during the distillation process as a medium to avoiding the oil that was extracted from mixing the water. Hexane is use because of the reactivity of hexane. After the sample was collected, hexane is removed by putting the sample in the fume hood because it has a lower boiling point than the oil. In the fume hood, hexane was left for an amount of time for its fully vaporized. To make sure that the hexane is fully vaporized, the sample is weighted every hour until the weight of the sample reaching a constant value where it assumes that the hexane is fully vaporized. Figure 4.1 below show the oil sample of gaharu;

Figure 4.1 Sample of gaharu oil

4.2 Results and Discussion

From the experiment, the result are obtained for the following experiment and showed in the Table 4.1 below;

Table 4.1 The result from the experiment

Variable	Experiment	Time in ultrasonic (hours)	Ratio sample water (w/v)	Time, t (days)	Result % yield
Solid to solvent ratio (w/v)	R1	3	1 : 8	3	0.111
	R2	3	1 : 12	3	0.122
	R3	3	1 : 16	3	0.131
	R4	3	1 : 20	3	0.139
Extraction time (hours)	T1	1	1 : 8	3	0.960
	T2	3	1 : 8	3	0.111
	T3	6	1 : 8	3	0.144
	T4	9	1 : 8	3	0.169

From the data in Table 4.1, graph for the extraction of gaharu for solid to solvent ratio are plot in the figure 4.2 below;

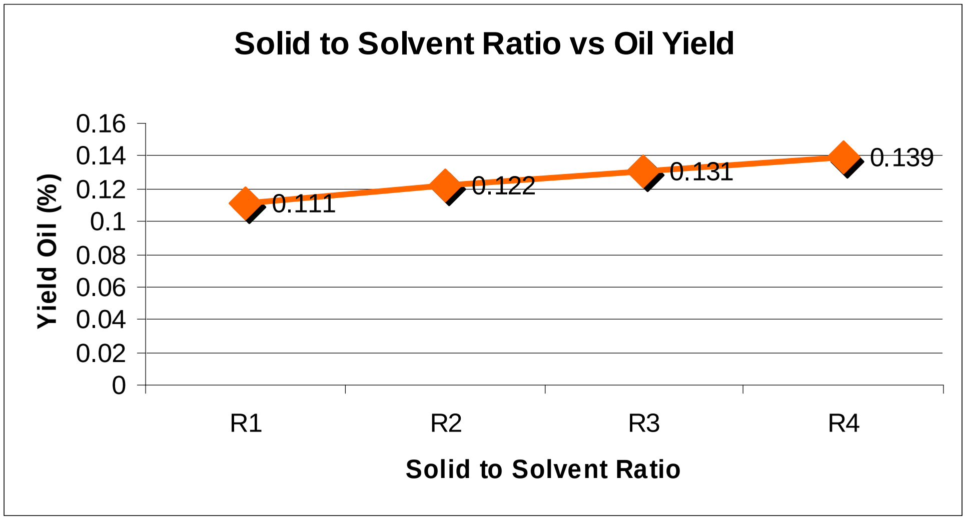


Figure 4.2 Graph for percentage oil yield versus solid to solvent ratio

From the graph, the legend R1, R2, R3 and R4 are stands for solid to solvent ratio of 1:8, 1:12, 1:16 and 1:20 where the ratio is between 1 parts of gaharu sample in kilograms to 8 parts of solvent which is water in milliliters. The graph shows that the solid to solvent ratio of 1 to 20 gives the highest percentage of oil yield that is 0.139 percent. The experiment are started at the ratio of 1 to 8 because it is the ideal ratio and the ratio cannot lower than 1 to 8 where the sample will be saturated and the concentration are to high to undergo the distillation process. The graph shows that the differential between the value of oil percentage for each ratio are too small and the result cannot be consider in making the comparison between the variables.

From the graph, the percentage yield of oil increasing along with the solid to solvent ratio. This is because of the solvent ratio that will assist the breaking of the gaharu cell wall in the ultrasonic process. When the amount of solvent is increase, the kinetic energy during the ultrasonic process is easily transmitted to the gaharu sample producing the higher vibration of the cell of the gaharu sample (Bulyamin, 2005).

From the data in Table 4.1, graph for the extraction of gaharu for time variable are plotted in the Figure 4.3 below;

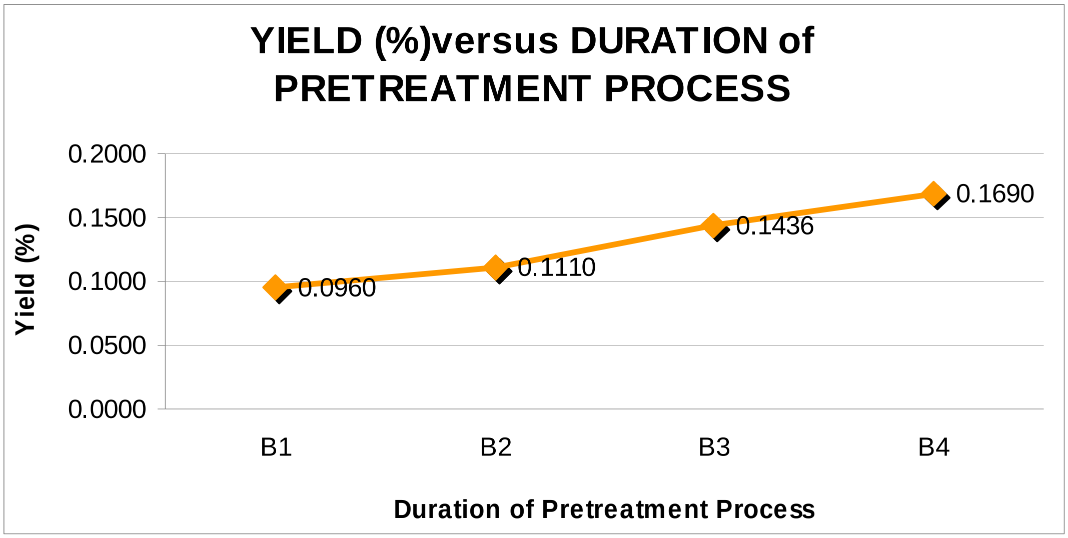


Figure 4.3 Graph for percentage oil yield versus extraction time

From the graph, the legend B1 is stands for 1 hour extraction time, B2 is for 3 hour extraction time, B3 is for 6 hour extraction time, and B4 is for 9 hour extraction time. The graphs showed that B1 is the lowest value and B4 is the peak and the biggest value of percentage oil yield. The graphs will continue rising for the value that larger than B4 until it will came to a constant value as the entire sample are completely extracted. The longer the extraction time, the higher the oil yields percentage.

From Figure 4.3, the oil yield percentage are increasing along with the pretreatment time because of the sample are exposed in the ultrasonic bath for a period of time. The ultrasonic process assisted the extraction of oil by the acoustic stream and cavitation process where the process will breaking up the gaharu cell wall and make it easier for the extraction of the oil. The longer exposure of the sample in the ultrasonic bath, the larger amount of cell wall were broken resulting the higher percentage of oil produce during the hydro-distillation process.

When doing this research, to get the value of the oil yield percentage, the experiment need to be repeated where the value are not acceptable according to the journal because there might being some error when running the experiment. The error might occur as parallax error when determining the measurement of the amount of the solvent. Error might also occur in the process of extracting and collecting the oil such as where the shortage in electricity and water supply that interfere the process.

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

In this experiment, for the conclusion, the ultrasonic assisted distillation is consider as an improved method for the extraction of gaharu essential oil where it can produce a higher yield of oil compare to the traditional and conventional method that still applied nowadays. The extraction of gaharu by using ultrasonic assisted distillation is a feasible way to produce the oil yield percentage.

Furthermore, the used of ultrasonic cleaner as a pretreatment will reduce the time consuming for the process in extracting the oil because of its sonication and cavitation mechanism that will disintegrate the cells of the gaharu sample and making the process of extracting the oil easier.

In this experiment, in order to achieve the objective, the parameter which is solid to solvent ratio and treatment time are needed to be manipulated according to the experimental matrix in the methodology of the experiment.

According to the result, the optimum condition for the experiment are at 9 hours extraction time and the ratio of 1 to 20 of solid to solvent.

5.2 Recommendations

Gaharu oil extraction by using ultrasonic assisted distillation is an important research that must be continued in the future due to the value and the demands of gaharu that increasing nowadays. The method in this research can improve the recent method which is hydro-distillation where it can produce the higher yield of oil thus it also can help the local entrepreneur who involve in gaharu oil extraction. Based on the studies, there are several recommendation in order to improve this method of extraction which are;

1) It is quite hard to collect the oil that produce because of the yield of the oil is too small and some of the oil might stick at the wall of the apparatus. The solution is to run the extraction process in pilot scale rather in laboratory scale.

2) The product of the gaharu oil should be analyzing using GC/MS to ensure the main component of gaharu is well extracted.

3) Some enhancement is needed to be added into the ultrasonic assisted distillation to ensure that the process will produce a high yield of oil and increase its efficiency. The enhancement is such as enzymatic assisted extraction where enzyme will be used in the sample preparation.

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