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EXTRACTION OF ESSENTIAL OILS FROM ZINGIBERACEACE FAMILI USING MICROWVE TECHNIQUES

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By

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February 2017

Chairman : Assoc. Prof.Jumiah Hassan, PhD
Faculty : Science

The essential oil from ZingiberaceaceFamili has many potential, especially in medicine, cosmetic and food industries. Therefore, therequirement to get the highest yield as well as good quality of essential oil gives the motivation of this research. Nowadays, the conventional extraction, especially in the commercial factory uses a lot of energy, high cost and a lot of extraction times. To overcome these disadvantages, microwave extraction technique (MET) which known as the best alternative extraction methods in terms of the producing of essential oil at shorter time was used at various extractions process parameters. Besides that, the undertaken in this thesis includes the performance of heating process in terms of the absorption power and the electric field strength. The main goal of this research is to study the various factors using microwave-assisted extraction that influences the production of essential oil in terms of the total yield and the total percentages of the oxygenated compound from Zingiberaceace Family's essential oil (Java turmeric, Mango ginger, Black turmeric and Turmeric). Laboratory studies were carried out at different factors such as the different power of heating source using microwave-assisted extraction, the amount of water added into the fresh and frozen samples, the soaking time of the sample, and the dry sample with different drying methods. Basically, the sample with solvent (distilled water) was put in the container with a hole on top of the cover and the container will placed inside the Microwave Extraction Laboratory System (MELs) which is the laboratory microwave oven. The distillation unit was connected with the container and the extraction process parameter was controlled by using the terminal controller-personal computer where the EasyWave 3.5 software program was installed. The yield of essential oil was collected and stored until used.For Java turmeric, the unique parameter that gives higher yields (6.37 %) is at the combination of dried sample mixture using an open air drying method, 200 ml of water and 600 W of microwave power heating. For Mango ginger, the unique parameter that gives higher yields (1.22 %) is at the combination of dried sample mixture using the electric oven drying method, 200 ml of water and 600 W of microwave power heating. For Black turmeric, it produces higher yields (1.26 %) with the combination of these unique parameter; dried sample mixture using a microwave oven drying method, 200 ml of water and 600 W of microwave power heating. For Turmeric, the highestyields (2.25

%) were obtained with the combination of these unique parameter; dried sample mixture using an open air drying method, 200 ml of water and 600 W of microwave power heating. This indicates that every sample has its own parameter to produce the highest yield of essential oil and among those samples, the dried sample was found to be a unique parameter that produce high yield. This happens due to the cell wall of the sample's structures that already destroy and allow the extraction of essential oil easily. The second goal is to analyzechemical compounds, especially the percentage of oxygenated compound. This analysis was using gas chromatography –mass spectrometry (GC-MS) and found that at every experimental order, it reveals the same compound but different abundance. This happens due to the heating conditions as some compounds were very sensitive. Following the optimum parameter that give higher yield of essential oils, the total percentages of oxygenated compound that used to identify the quality of essential oilis at64.16%, 73.24%, 64.54% and 77.20% for Java turmeric, Mango ginger, Black turmeric and Turmeric. That means, among all the sample, the essential oil from Turmeric was estimated to be the best oil compared to others. Every experimental order successfully extracted the compounds known to have many advantages such as antimicrobial, analgesic, and antifungal like cedr-8-ene, ar-curcumene, camphor, caryophylleneoxide, α-curcumene, eucalyptol, *ar*-turmerone, curlone and turmerone. The third goal was comparing theperformance of the extraction microwave method (solvent free microwave extraction, SFME and microwave-assisted extraction, MAE) and conventional method (Hydro-distillation, HD)in terms of the yield and the percentages of oxygenated compound.The extraction was done in one combination of extraction process parameter. Microwave-assisted extraction method was determined to give the best result in terms of the total yieldas water assist in the extraction process. However, the hydro-distillation was found to produce high percentage of oxygenated compound. These indicate that different techniques of extraction play an important role in the production of essential oil. From the research, microwave-assisted extractionwas revealed as a best extraction method in terms of the producing the total yield of essential oil and as many factors influencing the production of essential oil, the result of the combination of extraction process parameter can be used for further research. The extraction process parameter does not affect the compound in the essential oil, but affect the total abundance. This happens due to the compound that very fragile and vanishes at certain condition.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Doktor Falsafah

PENGEKSTRAKAN MINYAK PATI DARI KELUARGA ZINGIBERACEACE MENGGUNAKAN TEKNIK MICROGELOMBANG

Oleh

NOR AZILA BINTI ABD AZIZ

Februari 2017

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Minyak pati dari Keluarga Zingiberaceace mempunyai banyak potensi, terutamanya dalam industi perubatan, cosmetic dan makanan. Oleh itu, keperluan untuk mendapatkan hasil minyak yang banyak dan berkualiti telah memberi motivasi untuk membuat kajian ini. Pada masa ini, penggunaan pengekstrakan konvensional masih lagi digunakan terutamanya kilang komersial. Ia menggunakan banyak tenaga, kos dan masa pengekstrakan.Untuk mengatasi kelemahan pengekstrakan konvensional, teknik pengekstrakan gelombangmikro (MET) yang dikenali sebagai kaedah pengekstrakan alternative terbaik dari segi penghasilan minyak pada masa yang singkat telah digunakan dalam kajian ini menggunakan pengekstrakan gelombang mikrodengan pelarut (MAE) dengan pelbagai faktor proses pengekstrakan. Selain itu, penyelidikan yang dijalankan didalam tesis ini termasuklah prestasi proses pemanasan dari segi kuasa penyerapan dan kekuatan medan elektrik. Matlamat utama kajian ini adalah untuk mengkaji faktor yang mempengaruhi menghasilkan minyak pati dari segi jumlah hasil dan komposisi minyak pati terutamanya jumlah peratusan sebatian oksigen dari Keluarga Zingiberaceace (Temu lawak, Temu mangga, Temu hitam dan Kunyit). Kajian makmal telah dijalankan pada factor yang berbeza seperti kuasa penyinaran yang berbeza, jumlah air yang ditambah ke dalam sampel segar dan beku, masa rendaman sampel, dan sampel kering dengan kaedah pengeringan yang berbeza. Pada asasnya, sampel dengan pelarut (air suling) dimasukkan kedalam bekas dimana bahagian atas penutupnya ada lubang dan bekas akan diletakkan di dalam Sistem Microwave Extraction Laboratory (MELs) yang merupakan ketuhar gelombang mikro makmal. Unit penyulingan disambung pada bekas yang berisi sampel dan parameter proses pengekstrakan dikawal dengan menggunakan computer pengawal peribadi terminal di mana program perisian EasyWave 3.5 telah dipasang. Hasil minyak pati dikumpulkan dan disimpan sehinggadigunakan. Temu lawak memberikan hasil yang lebih tinggi (6.37%) dengan kombinasi campuran sampel dikeringkan menggunakan kaedah pengeringan udara terbuka, 200 ml air dan 600 W pemanasan kuasa gelombang mikro. Hasil yang lebih tinggi diperoleh untuk Temu mangga (1.22%) adalah dengan kombinasi campuran sampel dikeringkan menggunakan kaedah ketuhar elektrik pengeringan, 200 ml air dan 600 W pemanasan kuasa gelombang mikro. Temu hitam pula mengeluarkan hasil yang lebih tinggi (1.26%) dengan kombinasi campuran sampel

dikeringkan menggunakan ketuhar gelombang mikro kaedah pengeringan oven, 200 ml air dan 600 W pemanasan kuasa gelombang mikro. Kadar penghasilan minyak yang tertinggi bagi Kunyit (2.25%) diperoleh dengan kombinasi campuran sampel dikeringkan menggunakan kaedah pengeringan udara terbuka, 200 ml air dan 600 W pemanasan kuasa gelombang mikro. Ini menunjukkan bahawa setiap sampel mempunyai parameter sendiri untuk menghasilkan minyak pati yang banyak dan sampel yang dikeringkan didapati menghasilkan hasil yang tinggi. Ini berlaku kerana dinding sel struktur sampe ltelah memusnahkan dan ini membolehkan pengekstrakan minyak pati dengan mudah. Matlamat kedua adalah untuk menganalisis komposisi minyak terutamanya peratusan sebatian oksigen. Analisis ini menggunakan kromatografi gas spektrometri -mass (GC-MS) dan diperhatikan bahawasanya setiap experiment mempunyai kompaun sama namun berbeza jumlahnya. Ini berlaku disebabkan oleh keadaan pemanasan dimana beberapa kompaun adalah sangat sensitif. Mengikuti optimum parameter yang memberikan hasil minyak pati yang tinggi, jumlah peratusan sebatian oksigen yang digunakan untuk mengenal pasti kualiti minyak pati adalah pada 64.16%, 73.24%, 64.54% dan 77.20% untuk Temu lawak, Temu mangga, Temu hitam dan Kunyit. Setiap eksperimen telah Berjaya mengekstak sebatian yang diketahui mempunyai banyak kelebihan seperti antimikrob, analgesic dan anti-kulat seperti cedr-8-ene, ar-curcumene, camphor, caryophyllene oksida, α-curcumene, eucalyptol, ar-turmerone, curlonedan turmerone. Matlamat ketiga adalah membandingkan prestasi kaedah pengekstrakan gelombang mikro (pengekstrakan gelombang mikro tanpa pelarut, SFME dan pengekstrakan gelombang mikro dengan pelarut, MAE) dan kaedah konvensional (penyulingan air, HD) dari segi hasil dan peratusan sebatian oksigen. Pengekstrakan dilakukan dengan satu kombinasi proses pengekstrakan parameter. Pengekstrakan gelombang mikro dengan pelarut telah menghasilkan minyak yang banyak.Walau bagaimanapun, penyulingan air menghasilkan peratusan sebatian oksigen yang tertinggi. Ini menunjukkan bahawa teknik pengekstrakan memainkan peranan yang penting dalam pengeluaran minyak pati. Dari penyelidikan ,pengekstrakan gelombang mikro dengan pelarut adalah kaedah pengekstrakan terbaik dari segi menghasilkan jumlah hasil minyak dan disebabkan banyak factor mempengaruhi pengeluaran minyak pati, keputusan penyelikan yang diperolehi boleh digunakan pada masa hadapan. Parameter proses pengekstrakan tidak menjejaskan sebatian namun iamenjejaskan jumlah peratusan. Ini berlaku disebabkan oleh sebatian yang sangat rapuh dan hilang pada keadaan tertentu.

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I certify that a Thesis Examination Committee has met on 7 February 2017 to conduct the final examination of Nor Azila binti Abd Aziz on her thesis entitled "Extraction of Essential Oils from Zingiberaceace Famili using Microwave Techniques" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Doctor of Philosophy.

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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LIST OF ABBREVIATIONS AND SYMBOLS

Glossary of terms

ANA	Automatic Network Analyzer
AIRTC	Automatic infrared temperature controller
Cal	Calculation
CET	Conventional extraction technique
EO	Essential oils
GC	Gas Chromatography
GC-MS	Gas Chromatography couple with Mass Spectrometry
HD	Hydro distillation
HM	Heating mantle
HP	Hewlett Packard
IEEE	Institute Electric Electronic Engineering
MAE	Microwave Assisted Extraction
MET	Microwave Extraction Technique
MELs	Microwave Extraction Labstation System
NDT	Non-destructive measurement technique
NMR	Nuclear Magnetic Resonance
OECP	Open-ended coaxial line probe
P	Power
PTEE	Polytrtraflouroethylene
SFME	Solvent free Microwave Extraction
Th	Theory
WD	Wet distillation

Symbols

MC_{fs}	Moisture content of fresh samples
EM_{Avgfs}	Average evaporated moisture of fresh sample
EM_{fs}	Evaporated moisture of fresh sample
MC_{Js}	Moisture content of Java turmeric
MC_{Ms}	Moisture content of Mango ginger
MC_{Bs}	Moisture content of Black turmeric
MC_{Ts}	Moisture content of Turmeric
ρ_{ds}	Density of dry samples
ρ_{fs}	Density of wet samples
V_{mfs}	Volume fraction of fresh samples mixture
V_a	Volume fraction of air

V_w	Volume fraction of water
ε^*_{mfs}	Dielectric properties of fresh samples mixture
ε'_{mfs}	Dielectric constant of fresh samples mixture
ε''_{mfs}	Dielectric loss factor of fresh samples mixture
$\varepsilon^*_{mfs(SFME)}$	Dielectric properties of fresh samples mixture during solvent free microwave extraction
$\varepsilon'_{mfs(SFME)}$	Dielectric constant of fresh samples mixture during solvent free microwave extraction
$\varepsilon''_{mfs(SFME)}$	Dielectric loss factor of fresh samples mixture during solvent free microwave extraction
$\varepsilon^*_{mfs(MAE)}$	Dielectric properties of fresh samples mixture during microwave-assisted extraction
$\varepsilon'_{mfs(MAE)}$	Dielectric constant of fresh samples mixture during microwave-assisted extraction
$\varepsilon''_{mfs(MAE)}$	Dielectric loss factor of fresh samples mixture during microwave-assisted extraction
ε^*_w	Dielectric properties of water
ε'_w	Dielectric constant of water
ε''_w	Dielectric loss factor of water
$\tan\delta_w$	Loss tangent of water
ε^*_{fs}	Dielectric properties of fresh samples
ε'_{fs}	Dielectric constant of fresh samples
ε''_{fs}	Dielectric loss factor of fresh samples
ε^*_{ds}	Dielectric properties of dry samples
ε'_{ds}	Dielectric constant of dry samples
ε''_{ds}	Dielectric loss factor of dry samples
ε'_{sJ}	Dielectric constant of Java turmeric
ε''_{sJ}	Dielectric loss factor of Java turmeric
$\tan\delta_{sJ}$	Loss tangent of Java turmeric
ε^*_{fsJ}	Dielectric properties of fresh Java turmeric
ε'_{fsJ}	Dielectric constant of fresh Java turmeric
ε''_{fsJ}	Dielectric loss factor of fresh Java turmeric
ε^*_{dsJ}	Dielectric properties of dry Java turmeric
ε'_{dsJ}	Dielectric constant of dry Java turmeric
ε''_{dsJ}	Dielectric loss factor of dry Java turmeric
ε'_{sM}	Dielectric constant of Mango ginger
ε''_{sM}	Dielectric loss factor of Mango ginger

$\tan\delta_{sM}$	Loss tangent of Mango ginger
ε^{*}_{fsM}	Dielectric properties of fresh Mango ginger
ε'_{fsM}	Dielectric constant of fresh Mango ginger
ε''_{fsM}	Dielectric loss factor of fresh Mango ginger
ε^{*}_{dsM}	Dielectric properties of dry Mango ginger
ε'_{dsM}	Dielectric constant of dry Mango ginger
ε''_{dsM}	Dielectric loss factor of dry Mango ginger
ε'_{sB}	Dielectric constant of Black turmeric
ε''_{sB}	Dielectric loss factor of Black Turmeric
$\tan\delta_{sB}$	Loss tangent of Black turmeric
ε^{*}_{fsB}	Dielectric properties of fresh Black turmeric
ε'_{fsB}	Dielectric constant of fresh Black turmeric
ε''_{fsB}	Dielectric loss factor of fresh Black turmeric
ε^{*}_{dsB}	Dielectric properties of dry Black turmeric
ε'_{dsB}	Dielectric constant of dry Black turmeric
ε''_{dsB}	Dielectric loss factor of dry Black turmeric
ε'_{sT}	Dielectric constant of Turmeric
ε''_{sT}	Dielectric loss factor of Turmeric
$\tan\delta_{sT}$	Loss tangent of Turmeric
ε^{*}_{fsT}	Dielectric properties of fresh Turmeric
ε'_{fsT}	Dielectric constant of fresh Turmeric
ε''_{fsT}	Dielectric loss factor of fresh Turmeric
ε^{*}_{dsT}	Dielectric properties of dry Turmeric
ε'_{dsT}	Dielectric constant of dry Turmeric
ε''_{dsT}	Dielectric loss factor of dry Turmeric
j	Constant value (-1)
Vol_{w}	Volume of water
$T_{w(f)}$	Specific temperature
$PO_{w(MELs)}$	Power output of water using MELs
$PO_{w(HM)}$	Power output of water using HM
Δt	Time consumption (min)
$(PA/Vol)_{w}$	Absorption power of water
POC	Power output consumption

E_{in}	Electric strength inside
$E_{in(w)}$	Electric strength inside of water
$E_{o(w)}$	Electric strength outside of water (air)
$(PA/Vol)_{mfs}$	Absorption power of fresh samples mixture
ε^*_{mfs}	Dielectric properties of fresh samples mixture
ε^*_{mfsMAE}	Dielectric properties of fresh samples mixture during microwave-assisted extraction
$\varepsilon^*_{mfsSFME}$	Dielectric properties of fresh samples mixture during solvent free microwave extraction
ε''_{mfs}	Dielectric loss factor of fresh samples mixture
$E_{in(mfsMAE)}$	Electric strength inside of fresh samples mixture during microwave-assisted extraction
$E_{in(mfsSFME)}$	Electric strength inside of fresh samples mixture during solvent free microwave extraction

CHAPTER 1

INTRODUCTION

This chapter coversthe research background with the information about essential oils and microwave extraction technique (MET), some benefits and potentials of this research, problem statement, limitations, objectives and hypothesis of the research.

1.1 Research Background

Curcuma xanthorrhiza Roxb or known as 'Temu lawak' (Java turmeric) in Malaysia is a member of the ginger family (Zingiberaceae). It is a native Indonesian plant and grown in Thailand, Philippines, Sri Lanka and Malaysia. Curcuma xanthorrhiza's rhizome (root) is similar to ginger with bitter taste, aromatic and pungent odor (Devaraj *et al.*, 2013). The color and shape of rhizome are similar to turmeric.

Java turmeric has been chosen as one from nine 'unusual' plants in Indonesia. It has many applications and can be used as food, medical purposes and as a tonic (Hwang and Rukayadi, 2006). Curcuminoid is the component that gives the yellow colour and usually it is used in cosmetic. Atsiri oil is used for cosmetic, medicine, and aroma. Both of these components have antibacterial effect (Dzen *et al.*). Starch is the biggest component in the Java turmeric. It has theability for food digestion and is mixed in baby food (Br *et al.*, 2006).

Curcuma xanthorrhiza has been reported to be useful to treat hepatitis, liver disease, cancer, diabetes, rheumatism, hypertension and heart disorder. It also has anti-bacterial, anti-spasmodic, anti-inflammatory, anti-oxidant and antifungal effects (Devaraj *et al.*, 2013).

Curcuma amada is apart from Zingiberaceae family and commonly known as mango ginger. It originated from Indo-Malayan and has been distributed widely from Asia to Africa and Australia. It is called mango ginger and is quite famous for this type of species because the smell and flavor are the same as mango and morphologically similar to ginger.

In thefood industry, mango ginger has been used in the manufacture of pickles, culinary preparations for salads as a flavor, candy and sauce (Kullu *et al.*, 2013). In medical application, it can be used to treat psychological problems. It isalso used in traditional medicine and Ayurvedic medicine. Mango ginger has many bioactive molecules where it demonstrates antibacterial, antifungal, anti-inflammatory, anti-hypercholesterolemic, insecticidal, antipyretic and antioxidant properties (Singh *et al*., 2010). Mangiferin is one of the important bioactive molecules. Mangiferin very useful in the treatment of skin diseases, asthma, bronchitis and inflammation because of xanthone-C-glycoside inside the mangiferin has antidiabetic, cardioprotective, immunomodulatory, antioxidant, antitumor, hepatoprotective and vasorelaxant properties (Karchuli and Pradhan, 2011).

Curcuma aeruginosa, the member of Zingiberaceae family is popular known as 'Temu Hitam' in Malaysia, wild arrowroot or East Indian arrowroot in India and Waan-Ma-Haa-Mek in Thailand. It is available in Malaysia, Burma, Indonesia, and in South India (Ranjini and Vijayan, 2005).

In medicine, the rhizome of this plant is used to treat asthma, cough, scurvy, and mental derangements. It also helps women in confinement to accelerate the lochia and decrease pain and inflammation of uterus by adding in beverage. Other than internal consumption, this rhizome can be used for external application. For example, it is used as poultice for inching (Reanmongkol *et al.*, 2006).

The 1,8-cineol, curzerenone, zedoarol, furanodienone, curcumenol and germacrone are some of the compounds identified from the essential oil of the rhizomes and leaves of Curcuma aeruginosa. The 1,8-cineol compound displayed antinociceptive and anti-inflammatory effects in experimental animals(Santos *et al.*, 2000).

Turmeric (the common name for Curcuma longa) is derived from the rhizomes of the plant. It is a perennial herb and member of the Zingiberaceae (ginger) family. It is cultivated extensively in Asian countries, especially in India and China. It is known by many names such as Curcum (Arab), Indian saffron, Haridra (Sanskrit, Ayurvedic), Jianghuang (yellow ginger in Chinese) and Kyoo or Ukon (Japanese).

In Asian cuisines, turmeric is used for color and flavor and became an important ingredient in curry powder. In Ayurvedic and Chinese medicine, it is used as an anti-inflammaory and to treat jaundice, menstrual difficulties, hematuria, hemorrhage, and colic (Labban, 2014). For internal, it is used as a stomachic, tonic and blood purifier. For external, it is used in the prevention and treatment of skin diseases (Jayaprakasha *et al.*, 2005).

The bioactive compound from the turmeric was found to be antimicrobial, anti-inflammatory, anticancer and antiviral (Pa *et al.*, 2012). ar- Turmerone, zingiberene, tumerone and curlone are major compounds found in volatile oil. It can be identifiedusing GC-MS (Jayaprakasha *et al.*, 2005).

To obtain good quality essential oils with high yield, the extraction process is very laborious. For many decades, conventional method of extraction such as hydro-distillation method was used. Generally, the conventional technique required a long experimental process and a large quantity of solvent. Because of this, the operation cost increased and environmental problem occurred (Tatke and Rajan, 2014). In addition, the temperature of extraction cannot be controlled and can cause overheating of the sample. Some volatile compounds will be lost to the environment in this condition. However, it depends on the type of sample used (Nurdin, 2007). The research was done to overcome the disadvantages of the conventional extraction technique and come up with alternative extraction techniques such as the microwave extraction technique, supercritical fluids and ultrasound.

Microwave extraction technique is the one of the alternative technique developed to overcome the disadvantages of the conventional extraction technique. This technique has the ability to minimize the time of extraction process due to the microwave energy that can penetrate the materials. The volumetrically heat source is produced as the molecules start to collide with each other (Zhou and Liu, 2006). This can be done automatically with the reduction in organic consumption (Zygmunt and Namieśnik, 2003).

1.2 Research Benefit and Potential

Microwave energy has been known for heating and drying in microwave chemistry, and stellar application. The microwave energy involved in the extraction of essential oils is in the microwave extraction technique (MET). Because of the advantages of MET, many researches were done using this technique and the essential oil extracted was studied based on the chemistry and biological aspects such as antioxidant, antimicrobial, antivenom, anti-tumor and anti-inflammatory properties. However, the studies using MET to extract the essential oil from Zingiberaceace family (Java turmeric, Mango ginger, Black turmeric and Turmeric) has been known from time immemorial in medicine, and food industry was so limited. Therefore, this project has high potential in raising the level of the country's economy because apart from the low cost, energy and time saving to produce the best quality essential oil with a bigger amount than the conventional method. It is also a green technology where chemical usage is unnecessary and the handling process is safe. This project results in appropriate parameters needed to produce good quality essential oil in large amounts and more time saving.

1.3 Microwave Extraction Technique and Conventional Extraction Technique

The demand for a new extraction technique with shortened extraction times, reduced organic solvent consumption, pollution prevention and low operating cost was increased. Driven by these goals, the alternative extraction techniques such as ultrasonic-assisted extraction (UAE), supercritical fluid extraction (SFE) and microwave extraction technique (MET) were developed (Abert *et al.*, 2008).

MET was found to the best method in terms of the production of essential oil in shorter period of time compared with other alternative extraction techniques (Tatke and Rajan, 2014). This technique involves the use of microwave energy which is electromagnetic wave with frequencies ranging between 300 MHz to 1000 GHz and this leads to the fact that MET is a fast extraction technique compared with others. The MET depends on many factors like types of solvent used rather it is non-polar or polar solvent, duration of extraction process, the irradiation power, the temperature and the matrix (Devgun *et al.*, 2012).

The conventional processes or the conventional extraction technique (CET) have been used for decades in the extraction of essential oils. Until today, it is still used especially in industrial scale. The main disadvantages of this technique are the long extraction time and the large amount of solvent used. Furthermore, this technique is not suitable to extract a thermo-sensitive compound as the possibility to decomposition is high due to the long extraction time at the boiling point of solvent used (Bimakr *et al.*, 2010). Usually, this technique could extract 0.005 to 10% of the essential oils from plants. The distillation duration, the temperature, the operating pressure, the type and quality of raw plant materials are the factors that can influence the production of essential oils (Li *et al.*, 2014).

1.4 Essential Oils

Essential oil is a complex mixture. It can be defined as the volatile material extracted from trees, flowers, stems, herbs and roots through distillation. This mixture consists of oxygenated terpenes, terpenes, oxygenated sesquiterpenes and sesquiterpes. Some of the compounds obtain in the essential oil cannot be classified as it can belong to any of the family of compounds mentioned earlier. There are other compounds that can be extracted especially from vegetable and usually in a small amount. There are fatty acid methyl esters (FAMES), coloring matters (p-carotene), sterols, coumarins and flavones (Reverchon, 1997).

Essential oil is a valuable natural product. It can be used as raw materials as spices, in cosmetics, perfumes, aromatherapy and nutrition. For many decades, it was claimed to have useful effect in aromatherapy together with the additional aromatic compounds (George *et al.*, 2015). It has also been used as food preservatives, alternative medicine, pharmaceuticals and natural therapies for thousands of years. Until today, it has various functions including conferring pest and disease resistant. In the cosmetic industry, it is used for the production of shampoo, lotion, cologne, cream and other make-up tools (Lis-Balchin and Deans, 1997)

The quality of essential oil is described by the presence of the aromatic compounds in the essential oils such as oxygenated and terpenes compound. If the oxygenated compound is of high value or a major compound, the essential oil can be acknowledged as a good quality essential oil. This is because the oxygenated compound is highly odoriferous. Alcohols, aldehydes, ketones, acids and ester are the usually oxygenated compound found in the essential oils (Ranasinghe *et al.*, 2003)

1.5 Statement of the Problem

The essential oil from Zingiberaceace family has much goodness, especially in medicine, cosmetic and food industries. However, this goodness was not used extensively as the knowledge about it so limited. So, this research was conducted as an attempt to introduce this kind of sample as well as the right way to extract and get the high yield and good quality of essential oil. In this research, microwave technique was used to extract the essential oil as it has many advantages compared with other alternative and conventional technique. Besides providing low cost and saving time and energy, microwave technique also the best method among the alternative technique to extract the high yield of essential oil at a shorter time. There are many factors that affect the extraction, such as extraction of solvent, time of extraction process, microwave power level, temperature and contact surface area. Therefore, this research, undertaken various extraction process parameters to obtain high yield and good quality of essential oil. Various extraction methods which are SFME and MAE in microwave technique and HD in conventional technique were done to compare the performance in terms of total yield and quality of essential oil.

1.6 Purpose of the Study

The primary purpose of this study is to investigate the optimized parameters of microwave extraction technique in order to obtain high yield and better quality of essential oils from Zingiberaceace family. The performance of microwave extraction technique in terms of their rapidity, the total yield and the quality of essential oil was compared with conventional extraction method. The physical properties of water and samples such as the moisture content, wet and dry density and dielectric properties were measured. It is important to measure the physical properties as it is the parameter needed to estimate the absorption power of the fresh sample mixture during microwave and conventional extraction technique at frequency 2.45 GHz. The objectives of this research are:

- to extract essential oils from Zingiberaceace family (Java turmeric, Mango ginger, Black turmeric and Turmeric) using microwave extraction technique by controlling the parameter of extraction process to obtain high yield essential oils. The parameters include the power of extraction, volume of solvent used, the nature of the samples (fresh and frozen samples), time of soaking and the drying method of samples.
- to analyze chemical compounds in the essential oils from Zingiberaceace family at different parameters of the extraction process and identified the valuable essential oils
- to compare the extraction performance between microwave (Solvent-free Microwave Extraction and Microwave-assisted Extraction) and conventional (Hydro-distillation) extraction technique in terms of rapidity, quality and quantity.

1.7 Hypothesis

The hypotheses for the research are:

1. Each sample would have its own extraction process parameter (power of extraction, volume of solvent used, the nature of the samples (fresh and frozen samples), time of soaking and the drying method of samples) to obtain high yield and good quality of essential oil.
2. Microwave-assisted extraction (MAE) would produce more essential oil in a shorter time.

1.8 Scope and Limitations of the Study

This study was limited by the following characteristics:

1. The sample consisted of the four types of rhizome in the Zingiberaceace family, which is Java turmeric, Mango ginger, Black turmeric and Turmeric. The samples were choosing because it has more medical values. It was assumed originated from the same place as it bought from same market.
2. The temperature of extraction was specific to 100°C, as this is the boiling point of water where it act as solvent to the sample.
3. The extraction time is about 1 and 4 hours, as this is the duration normally no more essential oil can be extracted for microwave and conventional, respectively.
4. For the dielectric properties of water, the frequency measurement was set at the range between 0.2 to 20 GHz, because it is the microwave frequency.
5. The total percentage of oxygenated compound was used to define the quality of essential oil as it is one of the methods.
6. The comparison between the extraction method (SFME, MAE and HD) was done in one combination of extraction process parameter as it is enough to determine the best method among them.

1.9 Thesis outline

The literature review and theory related to the study were discussed in Chapter 2. This includes the relationship between the characteristics of the sample which is the moisture content, density and dielectric properties with the microwave mechanism during the extraction using Solvent-free Microwave Extraction (SFME) and Microwave-assisted Extraction (MAE). The tools and equipment used during the experiment as well as the experimental methodology were discussed in Chapter 3. This chapter also discussed the errors that can influence the data together with their precautionary steps to minimize it during the experiment. In Chapter 4, the results of all the data including the characteristics of the sample, the yield of essential oil with the compound composition from the various samples used (Java turmeric, Mango ginger, Black turmeric and Turmeric), the rapidity of the extraction tools are presented and discussed in detailed. Finally, Chapter 5 concluded all the results obtained and the conclusions were drawn relating it to the original purpose of the study.

REFERENCES

Abert, M., Fernandez, X., Visinoni, F., Chemat, F. 2008. Microwave hydrodiffusion and gravity , a new technique for extraction of essential oils, 1190, 14–17.

Alitonou, G. A., Sessou, P., Tchobo, F. P., Noudogbessi, J. P., Avlessi, F., Ou, B. Y., Sohounhloue, D. C. K. 2012. Chemical composition and biological activities of essential oils of Chenopodium ambrosioides L. collected in two areas of Benin. International Journal of Biosciences, 2(8), 58–66.

Bimakr, M., Abdul, R., Saleena, F., Ganjloo, A., Salleh, L., Selamat, J., Zaidul, I. S. M. 2010. Comparison of different extraction methods for the extraction of major bioactive flavonoid compounds from spearmint (Mentha spicata L .) leaves, Food and Bioproducts Processing. *9*(March), 67–72.

Br, B., Ginting, I., Penelitian, B., Obat, T. (2006). Pengaruh kehalusan bahan dan lama ekstraksi terhadap mutu ekstrak Temulawak (Curcuma xanthorriza Roxb). Bul. Littro, XVII(2), 53–58.

Cardoso-ugarte, G. A., Juárez-becerra, G. P., Sosa-, M. E. 2013. Microwave-assisted extraction of essential oils from herbs, *47*(1), 63–72.

Chuvieco, E., Cocero, D., Riaño, D., Martin, P., Martínez-Vega, J., De La Riva, J., Pérez, F. 2004. Combining NDVI and surface temperature for the estimation of live fuel moisture content in forest fire danger rating. Remote Sensing of Environment, 92(3), 322–331.

Devaraj, S., Ismail, S., Ramanathan, S., Yam, M. F.2013. In vivo toxicological investigations of standardized ethanolic extract of Curcuma xanthorrhiza Roxb . rhizome, *3*(1), 67–73.

Devgun, M., Nanda, A., & Ansari, S. H. 2012. Comparison of conventional and non conventional methods of extraction of heartwood of Pterocarpus Marsupium Roxb., 69(3), 475–485.

Dorthe, A. M. 1999. Optimization by factorial design of focused microwave assisted extraction of polycyclic aromatic hydrocarbons from marine sediment, 228–237.

Dzen, S. M., Wibowati, S., Purwarini, A. W. Efek antimikroba ekstrak rimpang Temulawak (Curcuma xanthorrhiza) terhadap Salmonella typhi secara in vitro. Fk Ub, 1–12.

Flamini, G., Tebano, M., Luigi, P., Ceccarini, L., Simone, A., Longo, I. 2007. Comparison between the conventional method of extraction of essential oil of Laurus nobilis L . and a novel method which uses microwaves applied in situ , without resorting to an oven, 1143, 36–40.

Gao, M., Song, B., Liu, C. 2006. Dynamic microwave-assisted extraction of flavonoids from Saussurea medusa Maxim cultured cells, 32, 79–83.

George, M., Britto, S. J., Arulappan, M. T., Marandi, R. R., Kindo, I., Herbarium, R., Joseph, S. 2015. Research article phytochemical, antioxidant and antibacterial studies on the essential oil of the rhizome of Curcuma amada Roxb.

Hu, Z., Cai, M., Liang, H. 2008. Desirability function approach for the optimization of microwave-assisted extraction of saikosaponins from Radix Bupleuri, 61, 266–275.

Hwang, J., Rukayadi, Y. 2006. Challenges and opportunities in applying temulawak (Curcuma xanthorrhiza Roxb.) for industrial oral care products. Bogor Agricultural University, 25–32.

Jayaprakasha, G. K., Jagan Mohan Rao, L., Sakariah, K. K. 2005. Chemistry and biological activities of C. longa. Trends in Food Science and Technology, 16(12), 533–548.

Karchuli, M. S., Pradhan, D. 2011, *952*, 947–952

Khajeh, M., Reza, A., Moghaddam, A. 2010. Application of Doehlert Design in the optimization of microwave-assisted extraction for determination of zinc and copper in cereal samples using FAAS, 133–137.

Khalid, K. 1988. The application of microstrip sensor for determination of moisture content in Hevea Rubber Lates. J. Microwave Power and EM Energy 45: 23-31.

Khalid, K., Hassan, J., Daud, W.M. 1994. Dielectric properties of Hevea Latex at various moisture content. Journal Natural Rubber 9(3): 172-189.
Kraszewski, A. 1974. Determination of the strength of water suspension using microwave bridge technique. J. Microwave Power. 9:295

Kullu, J., Dutta, A., Constales, D., Chaudhuri, S., Dutta, D. 2013. Experimental and modeling studies on microwave-assisted extraction of mangiferin from Curcuma amada. *3 Biotech*, *4*, 107–120. doi:10.1007/s13205-013-0125-5

Labban, L. 2014. Medicinal and pharmacological properties of Turmeric (Curcuma longa): A review, 5(1), 17–23.

Li, Y., Fabiano-Tixier, A-S,; Chemat, F. 2014. Essential oils as reagents in green chemistry. 9–21.

Lis-Balchin, M., Deans, S. G. 1997. Bioactivity of selected plant essential oils against listeria monocytogenes. Journal of Applied Microbiology, 82(6), 759–762.

Lucchesi, M. E., Smadja, J., Bradshaw, S., Louw, W., Chemat, F. 2007. Solvent free microwave extraction of Elletaria cardamomum L .: A multivariate study of a new technique for the extraction of essential oil, 79, 1079–1086.

Lucchesi, M. E., Chemat, F., Smadja, J. 2004. Solvent-free microwave extraction of essential oil from aromatic herbs: comparison with conventional hydro-distillation. Journal of Chromatography A, 1043(2), 323–327.

Ma, W., Lu, Y., Dai, X., Liu, R.,Hu, R. and Pan, Y. 2009. Determination of anti-tumor constitute mollugin from traditional Chinese medicine Rubia cordifolia: Comparative study of classical and microwave extraction techniques. Separation Science and Technology. 44(4), 995-1006.

Pa, M. H., S, J. S., Sm, M. A., Jacob, A. 2012. Phytochemical characterization and Antimicrobial activity of oil and solvent extracts of Curcuma longa. Research Journal of Pharmaceutical Biological and Chemical Sciences. 3(3), 49–55.

Padmapriya, K., Dutta, A., Chaudhuri, S., Dutta, D. 2012. Microwave assisted extraction of mangiferin from Curcuma amada. 3 Biotech, 2(1), 27–30.

Quan, P. T., Hang, T. Van, Ha, N. H., De, N. X., Tuyen, T. N. 2006. Microwave-assisted extraction of polyphenols from fresh tea shoot, (1), 69–75.

Ranasinghe, L. S., Jayawardena, B., Abeywickrama, K. 2003. Use of waste generated from cinnamon bark oil (Cinnamomum zeylanicum Blume) extraction as a post harvest treatment for Embul banana, *1*(April), 340–344.

Ranjini, C. E., & Vijayan, K. K. 2005. Structural characterization of a glucan from the tubers of Curcuma aeruginosa, *44*(March), 643–647.

Reanmongkol, W., Subhadhirasakul, S., Khaisombat, N., Fuengnawakit, P., Jantasila, S., Khamjun, A. 2006. Investigation the antinociceptive, antipyretic and anti-inflammatory activities of Curcuma aeruginosa Roxb. extracts in experimental animals. Songklanakarin Journal of Science and Technology, 28(5), 999–1008.

Reverchon, E. 1997. Supercritical fluid extraction and fractionation related products. Journal of Supercritical Fluids, 10, 1–37

Routray, W., Orsat, V. 2012. Microwave-assisted extraction of flavonoids : A Review, 409–424.

Santos, F. a, Rao, V. S. N. 2000. Antiinflammatory and antinociceptive effects of 1, 8-cineole a terpenoid oxide present in many plant essential oils. Phytotherapy Research, 14(4), 240–244.

Singh, S., Kumar, J. K., Saikia, D., Shanker, K., Thakur, J. P., Negi, A. S., Banerjee, S. 2010. A bioactive labdane diterpenoid from Curcuma amada and its semisynthetic analogues as antitubercular agents. European Journal of Medicinal Chemistry. 45(9), 4379–4382.

Suresh, N., Callanghan, J.C. Creelman, A.E. 1976. Microwave measurement of the degree of binding of water absorbed in soils. J. Microwave Power. 129, 2-4.

Tatke, P., & Rajan, M. 2014. Comparison of conventional and novel extraction techniques for the Extraction of Scopoletin from Convolvulus Pluricaulis. 48(1), 27–31.

Technology, B. 2012. Lymphocyte proliferation by Temu Lawak (Curcuma xanthorrhiza ROXB) Essential Oil. *29*, 205–209.

Veggi, P. C., Martinez, J., Meireles, M. A. A. 2013. Fundamentals of Microwave Extraction.

Venkatesh, M. S., & Raghavan, G. S. V. 2004. An overview of microwave processing and dielectric properties of agri-food materials. Biosystems Engineering, 88(1), 1–18.

Wang, S., Tang, J., Johnson, J. A., Mitcham, E., Hansen, J. D., Hallman, G.,Wang, Y. 2003. Dielectric properties of fruits and insect pests as related to radio frequency and microwave treatments, 85, 201–212.

Wang, Y., You, J., Yu, Y., Qu, C., Zhang, H., Ding, L.,Li, X. 2008. Analysis of ginsenosides in Panax ginseng in high pressure microwave-assisted extraction, 110, 161–167.

Xiao, W., Han, L., Shi, B. 2008. Microwave-assisted extraction of flavonoids from Radix Astragali, 62, 614–618.

White, J.R. 1970. Measuring the strength of the microwave field in a cavity. Journal of Microwave Power 5: 145-147.

Zhou, H.-Y., Liu, C.-Z. 2006. Microwave-assisted extraction of solanesol from tobacco leaves. Journal of Chromatography. A, 1129(1), 135–9.

Zygmunt, B., Namieśnik, J. 2003. Preparation of samples of plant material for chromatographic analysis. Journal of Chromatographic Science, 41(3), 109–116.

Books

Allan W. S. 1993. Undersatanding Microwaves. Published by John Wiley & Sons,Inc., Hoboken, New Jersey. 5-7.

Brennan, J.G. 1990. Dehydration of foodstuffs, In: Water and Food Quality, Hardman, T.M. (ed), Elsevier Applied Science, New York. 33-70

Debye, P. 1929. Polar Molecules. New York:Chemical Catalog Co.

Kraszewski, A. 2005. Recent Development in Electromagnetic Aquametry. Electromagnetic Aquametry:Electromagnetic Wave Interaction with Water and Moist Substances, Published by Springer. 1-14.

Matthew, N.O.S. 2007. Prinsiples of Electromagnetics Fourth Edition, Published by Oxford University Press Inc. 133-161.

Thesis

Dai. J. 2006. Microwave-assisted Extraction and Synthesis Studies and The Scale-up Study with The Aid of FDTD Simulation. PhD Thesis, Department of Bioresource Engineering, McGill University Montreal, QC, Canada.

Nurdin, R. 2007. Microwave Extraction of Essential Oils from 'Penaga Lilin'(Mesua Ferrea L.) Leaves. Master Thesis, Department of Physic, Universiti Putra Malaysia

Routray, W. 2014. Effect of Different Extraction Methods, Environmental and Post-harvest Factors on Yield of Phenolic Compounds From Blueberry Leaves. PhD Thesis, Department of Bioresource Engineering, McGill University Montreal, QC, Canada.

Conferences

Khalid, K. 2004. Microwave Aquametry: A Growing Technology, Inaugural Lecture. Kuala Lumpur, Universiti Putra Malaysia:Bahagian Komunikasi korporat UPM.

Webpage

Food and Agriculture Organization of the United Nations (1995). Basic Principles of Steam Distillation. Retrieved August 18, 2005, from http://www.fao.org/docrep/V5350e/V5350e13.htm (accessed on 26th Febuary 2016)

http://chestofbooks.com/health/materia-medica-drugs/Textbook-Materia-Medica/Ginger-Zingiber-Rhizoma-Zingiberis.html#ixzz4bAx54oJO (accessed on 13th March 2017)

http://chestofbooks.com/health/materia-medica-drugs/Textbook-Materia-Medica/Turmeric-Rhizoma-Curcumas.html#ixzz4bAxHFpH0 (accessed on 13th March 2017)