THE EXTRACTION OF ESSENTIAL OIL FROM *Quercus infectoria*
(MANJAKANI) GALLS USING SUPERCritical CARBON DIOXIDE
PRESSURE SWING TECHNIQUE

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(MANJAKANI) GALLS USING SUPERCRITICAL CARBON DIOXIDE
PRESSURE SWING TECHNIQUE

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A thesis submitted in fulfilment of the
requirements for the award of the degree of
Master of Engineering (Bioprocess)

Faculty of Chemical Engineering
Universiti Teknologi Malaysia

JUNE 2013
ACKNOWLEDGEMENT

In the name of Allah, the Most Gracious and the Most Merciful

Alhamdulillah, all praises to Allah for the strengths and His blessing in completing this thesis. This thesis would not have been possible without the guidance and the help of several individuals who in one way or another contributed and extended their valuable assistance in the preparation and completion of this study. First and foremost, I would like to express my sincere gratitude to my supervisor Dr. Liza Md. Salleh, with her enthusiasm, her inspiration and her great efforts to explain things clearly and simply. Throughout my thesis writing, she provided encouragement, good teaching, good company and lots of good ideas, without her, this thesis would not have been completed or written.

I am obliged to many of my colleagues who supported me, their friendship and assistance has meant more to me than I could ever express. Their guidance has served me well and I owe them my heartfelt appreciation. Special thanks to Madam Siti Zalita for her friendly assistance in practical analysis of this thesis. Thank you very much.

Last but not least, I owe sincere and earnest thankfulness to my beloved family. To my father, Mr. Rosland Abel Richard Lee, my mother, Mrs. Jumatiah Sagulu, my dear husband, Mohd Azmi Pang Thien Soong, my dearest sisters, Sabrina, Stasha Eleanor, Synthia Attilah and Sylvia Viviena for helping me get through the difficult times and for all the emotional support, I owe them everything and wish I could show them how much I love and appreciate them. To them I dedicate this thesis.
ABSTRACT

The study of essential oil of *Quercus infectoria* (Manjakani) was carried out using supercritical carbon dioxide (SC-CO$_2$) extraction. Two techniques were tested: Soxhlet extraction and SC-CO$_2$ extraction with pressure swing technique. The experiments of SC-CO$_2$ extraction investigated the effect of pressure and temperature on extraction yields and solubility of galls oil. The extraction were performed at pressures range of 35 to 48 MPa, temperatures between 40 and 60°C with extraction time of 30 to 60 minutes. The experiments were designed using Response Surface Methodology (RSM) to determine the optimum conditions. In this study, SC-CO$_2$ extraction combined with the pressure swing technique produced higher oil extracted compared to the continuous SC-CO$_2$ extraction with 2.58 % and 1.12 % yield, respectively. The analysis of RSM indicated that the extraction temperature has a major linear effect on the galls oil extraction with 17.92 % yield. The optimum extraction process parameters were for pressure of 38 MPa, temperature of 75°C and extraction time of 54 minutes with 1.12 % yield. The experimental solubility data was successfully correlated using Chrastil model with the coefficient of determination ($R^2$) value of 0.95. SC-CO$_2$ extraction combined with the pressure swing technique proved of potential to produce greater yield by using much lower SC-CO$_2$ compared to the continuous SC-CO$_2$ extraction. The identification of bioactive compounds from essential oil was analyzed using High Performance Liquid Chromatography (HPLC) analysis, and it was observed that *Quercus infectoria* galls oil contained gallic acid as the major component at the retention time of 2.79 min.
Kajian pengekstrakan minyak galls dari *Quercus infectoria* (Manjakani) dijalankan menggunakan kaedah pengekstrakan karbon dioksida bendalir lampau genting (SC-CO₂). Pengekstrakan cecair ini diuji menggunakan pengekstrakan Soxhlet dan SC-CO₂ dengan teknik ayunan tekanan. Eksperimen melibatkan penyiasatan tentang kesan tekanan dan suhu pada hasil pengekstrakan dan kebolehlarutan minyak galls menggunakan pengekstrakan SC-CO₂. Keadaan proses pengekstrakan yang digunakan pada tekanan 35 hingga 48 MPa, suhu diantara 40 dan 60 °C manakala masa pengekstrakan adalah diantara 30 dan 60 minit. Eksperimen telah direka menggunakan Kaedah Tindakbalas Permukaan (RSM) bagi menentukan keadaan proses yang optimum. Pengekstrakan menggunakan SC-CO₂ yang digabungkan dengan teknik ayunan tekanan menghasilkan produk ekstrak minyak yang lebih tinggi (2.58 % hasil ekstrak) berbanding pengekstrakan berterusan SC-CO₂ (1.12 % hasil ekstrak). Analisis menunjukkan bahawa suhu pengekstrakan mempunyai kesan utama pada hasil ekstrak minyak galls dengan peratus hasil ekstrak 17.92 %. Keadaan proses yang optimum bagi proses perahan minyak galls adalah pada tekanan 48 MPa, suhu 75 °C dengan masa pengekstrakan 54 minit bagi menghasilkan ekstrak minyak galls 1.12 %. Data kelarutan dari eksperimen telah berjaya dikaitkan menggunakan model Chrastil dengan pekali penentuan, $R^2$ bernilai 0.95. Pengekstrakan SC-CO₂ yang digabungkan dengan teknik ayunan tekanan berpotensi tinggi mengeluarkan hasil ekstrak minyak galls yang lebih tinggi dengan penggunaan SC-CO₂ yang lebih rendah berbanding pengekstrakan berterusan SC-CO₂. Pengenalpastian sebatian bioaktif dari minyak galls dianalisis menggunakan analisis Kromatografi Cecair Prestasi Tinggi (HPLC) dengan minyak *Quercus infectoria* galls mengandungi asid gallic sebagai komponen utama telah dikesan, manakala masa tahanan dikesan pada 2.79 minit.
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LIST OF ABBREVIATIONS

AARD - Average absolute relative deviation
ACE - Angiotensin converting enzyme
BBD - Box-Behnken Design
CNSL - Cashew nut shell liquid
CO₂ - Carbon dioxide
HPLC - High performance liquid chromatography
MOX - Malaysian Oxygen
\( P_c \) - Critical pressure
PT - Pressure-Temperature
RSM - Response Surface Methodology
SCF - Supercritical fluid
SC-CO₂ - Supercritical carbon dioxide
SEM - Scanning electronic microscope
SFE - Supercritical fluid extraction
\( T_c \) - Critical Temperature
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CHAPTER 1

INTRODUCTION

Essential oils are aromatic liquids derived from various parts of different plants (Dorie, 2001). Depending on the plants, essential oils are located in the bark, leaves, flowers, seeds and roots. There are different methods of extracting essential oils, based on the plant parts from which the individual oils are derived. The extraction process, which separates oils from their sources, has been employed for more than 40 centuries. The extraction methods include cold press, steam distillation, microwave extraction and solvent extraction. Nevertheless, a fairly new modern technology supercritical fluid extraction (SFE) with carbon dioxide (CO₂) may increase the production efficiency and contributes to preservation of the environment by reducing the use of solvents and the generation of hazardous substances (Masayoshi, 2010).

Over the last decade, the extraction of essential oil by supercritical fluids technology has re-emerged, mainly due to the dramatic increased in the research and development activities focusing on innovative and new trends approaches. SFE technology is growing at rapid pace because it can overcomes many disadvantages associated with conventional technologies and meet the consumer demand for natural products. In addition, large amounts of organic solvents used, particularly in the pharmaceutical, food and chemical industries can cause toxicity and generate
hazardous wastes to the environment. Therefore, in the search for environmentally friendly solvents, supercritical fluid extraction has been given high attention in variety of applications (Jose et al., 2007).

*Quercus infectoria* or locally known as Manjakani in Malaysia is a small tree native of Greece and Asia Minor, with four to six feet in height. The stems are crooked, shrubby looking with smooth and bright-green leaves borne on short petioles of 1 to 1.5 inches long. The leaves is bluntly mucronate, rounded, smooth, unequal at the base and shiny on the upper side, as shown in Figure 1.1a. Meanwhile, *Quercus infectoria* (Figure 1.1b) galls are corrugated and can be used as a thickener in stews or mixed with cereals for making bread.

![Figure 1.1](image1.png)  
(a) Oak gall tree leaves  
(b) *Quercus infectoria* galls

In India, it is known as Majuphal and is widely used as Indian traditional medicine for treatment of toothache and gingivitis. Compared with other plant seeds, Manjakani is relatively less known. This plant is also efficacious for women health. Manjakani is also known as "herbal magic" which are rich with tannins, calcium, protein, vitamins A and C (Rina et al., 2011). Additionally, the galls contain elements of astringent which can help to inhibit bacterial growth in urinary tract of women. Galls are mainly imported from Syria and Turkey, with some high grades are brought in smaller quantity from other countries namely China and Japan. Galls
are purely effective astringent and scarcely stimulant. They can be used as an injection in bad leucorrhrea where they arrest putrefactive tendencies, and can be combined with suitable stimulants (Hwang et al., 2010).

1.1 Background of the Problem

In the past few years, a published article highlighted the potential of antioxidant activity from *Quercus infectoria* galls with an addition of ethno botanical that can act as a stimulant and also can be used in treating skin disorders (Umachig et al., 2008). In addition to medicinal properties for all parts of plants, the galls can live for more than 10 years before undergo the seedling process. In Indonesia, few researches have been carried out in the extraction of *Quercus infectoria* galls by using solvent extraction. The most important components extracted from these techniques were gallotannin, gallic acid and ellagic acid (Ikram and Nowshad, 1977). Nevertheless, the study on extraction of essential oils from *Quercus infectoria* galls using SFE has never been reported before. Therefore, this research will be conducted to emphasize the investigation on the extraction of essential oils from *Quercus infectoria* by using supercritical carbon dioxide (SC-CO$_2$) method coupled with pressure swing technique.

Supercritical fluid extraction has been documented as an effective method for extraction of essential oils from plant materials (Modey et al., 1996). In Malaysia, the use of supercritical fluid extraction is still unpopular even though several local researchers have studied the feasibility of this technique especially in the herbs industry such as *Strobilanthes crispus* (Pecah Kaca) (Azizi et al., 2007, Norulaini et al., 2009 and Liza 2010). Therefore, the result from this research is important to be revealed and documented in order to evaluate the therapeutic properties of essential oils and their potentials in pharmaceutical or food industries.
1.2 Problem Statement

Generally, it is feasible to obtain quality results by using the SC-CO₂ extraction process, as well as selective extraction results. As the galls are usually coated by a hard shell, there is a limitation on the efficiency of the extraction system. The pressure swing technique approach is considered as an improvement of SC-CO₂ process, where the quantity and quality of extraction can be improved, particularly for hard grains, which is influenced the botanical structure of the material used. The pressure swing method of SC-CO₂ is considered as an economical process as it only consumes a small amount of CO₂ gas for the extraction process.

1.3 Research Objectives

The objectives of this research are to study the effects of process parameters on the yield of *Quercus infectoria* (Manjakani) galls oil and also the ability of SC-CO₂ with pressure swing technique for extraction of galls oil.

1.4 Scopes of Work

The extraction of *Quercus infectoria* galls was performed at selected range of conditions by using both conventional SC-CO₂ and SC-CO₂ pressure swing techniques. Therefore, the scopes of the work are as follows:

a) Conducting experiments to determine the effects of process parameters which include pressure, temperature and extraction time on the extraction yield at
selected range of conditions and analysis of crude yield composition was carried out using High Performance Liquid Chromatography (HPLC) analysis.

b) Optimization of variable condition (pressure, temperature and extraction time) by using Response Surface Model, with Box-behnken design (BBD) was used in the design of the experiment.

c) Investigation on the correlation of the solubility behaviour by using Chrastil model.

d) Comparison of extraction yield between conventional SC-CO$_2$ and SC-CO$_2$ with pressure swing techniques.

1.5 Research Contributions

Supercritical fluid extraction techniques are widely investigated in many industrial areas, and most of the published materials are concerned with the extraction of analytes from solid or semi-solid materials matrices. Several extraction techniques available over the past decades such as steam distillation extraction, ultrasound-assisted extraction, Soxhlet extraction and water steam distillation have been used to extract various natural compounds and essential oils from plant materials (seed, galls or leaves). Unfortunately, the oldest methods of extractions are very time consuming and require large quantities of solvents. Recently, the industry comes out with new observations, where dissolution of solutes in supercritical fluid (SCF) media has introduced the possibility of a new solvent medium. Consequently, SFE using CO$_2$ as a solvent is known as an efficient extraction technique for solid materials and it is extensively studied for separation of active compounds from herbs and other plants. The findings of this study, together with the pressure swing technique, will be viewed as a new approach for comparison with the current SC-
CO₂ continuous extraction process. In addition, there is lack of information on pressure swing extraction reported on SFE study. Thus, the information obtained from this study may provide a new approach to disclose the essential oils obtained from *Quercus infectoria* galls extraction and also as a reference to the SFE industry involving extraction of essential oils from seed plants.
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