



UNIVERSITI PUTRA MALAYSIA

***EXTRACTION OF EUGENOL FROM BETEL LEAVES (PIPER BETLE L.)
USING HYDRODISTILLATION AND SUPERCRITICAL CARBON
DIOXIDE TECHNIQUE***

NOOR HADZUIN BINTI NIK HADZIR

FK 2012 126

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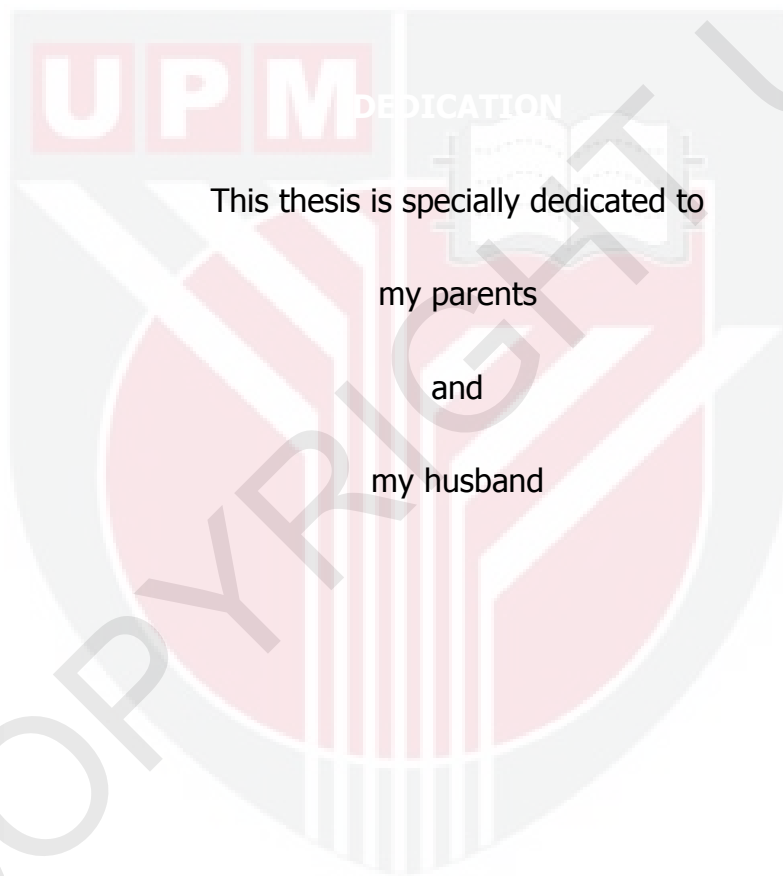


By

NOOR HADZUIN BINTI NIK HADZIR

**Thesis Submitted to the School of Graduate Studied, Universiti
Putra Malaysia, in Fulfilment of the Requirement for the Degree of
Master of Science**

December 2012



This thesis is specially dedicated to
my parents
and
my husband

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Master of Science

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USING HYDRODISTILLATION AND SUPERCRITICAL CARBON
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December 2012

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Faculty: Engineering

Betel (*Piper betle* L.) is one of the precious herb plants originated from Malaysia. The leaves of betel have been used traditionally for various medicinal purposes. Scientific research on the leaf of this plant claims that it possesses many beneficial bioactivities. Extract from betel leaves has a great potential to be used in developing commercial products. However, there is lack of research on the processing aspects to produce its bioactive extract.

Many extraction methods are widely used in extracting bioactive compounds. However, each method is different since it has advantages and disadvantages. This study focuses on two processes involved in producing

bioactive extract of betel leaves namely hydrodistillation and supercritical fluid extraction. Different experiments were designed and carried out to look into the effects of various operating parameters.

The first part of this study investigated the effect of type of leaves and extraction time on yield and composition of eugenol by using conventional hydrodistillation method. Results of the study showed that fresh betel leaves were more preferable over dried betel leaves. Six hours was compatible for extraction of fresh betel leaves.

The second part of this study is to examine effect of pressure and temperature on yield and concentration of eugenol by using supercritical fluid extraction. Response surface methodology (RSM) was applied to obtain the optimum process parameter. Optimized pressure and temperature which were suggested by RSM are 190 bar and temperature 50 °C. The predicted properties of extract are 0.5% yield and 21.57 mg/ml concentration of eugenol.

The kinetic model was used to describe the mass transfer phenomena. The highest value of mass transfer coefficient was found to be 0.208 min^{-1} at pressure 160 bar and temperature 50 °C. Del Valle-Aguilera model showed the best fit with experimental data with lowest average absolute relative

deviation (AARD). This equation is recommended for betel oil solubility in supercritical fluid extraction.

Comparison of supercritical carbon dioxide technique and hydrodistillation was studied. Supercritical carbon dioxide technique offers many important advantages over hydrodistillation. Therefore, this technique can be considered as a distinguished technology for the extraction of betel leaves, not only due to proclaimed advantages over the conventional techniques but also due to the high target compound concentration and yield with short extraction time (1 hour).

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan untuk ijazah Master Sains

**PENGEKSTRAKAN EUGENOL DARI DAUN SIRIH (*PIPER BETLE L.*)
MENGUNAKAN PENYULINGAN HIDRO DAN TEKNIK BENDALIR
LAMPAU GENTING KARBON DIOKSIDA**

Oleh

NOOR HADZUIN BINTI NIK HADZIR

Disember 2012

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Fakulti: Kejuruteraan

Sirih (*Piper betle L.*) ialah merupakan salah satu tumbuhan herba bernilai yang berasal dari Malaysia. Daun sirih telah digunakan secara tradisional untuk pelbagai tujuan perubatan. Kajian saintifik ke atas daun pada tumbuhan ini menunjukkan bahawa ianya mempunyai banyak bioaktiviti yang bermanfaat. Ekstrak daripada daun sirih ini mempunyai potensi yang besar untuk digunakan dalam pembangunan produk komersial. Tetapi, terdapat kurangnya penyelidikan dalam aspek pemprosesan untuk menghasilkan ekstrak yang bioaktif ini.

Banyak kaedah pengekstrakan digunakan secara meluas untuk mengekstrak sebatian bioaktif. Walaubagaimanapun, setiap kaedah adalah berbeza-beza kerana mempunyai kebaikan dan keburukan. Penyelidikan ini berfokus kepada dua proses yang terlibat dalam penghasilan ekstrak bioaktif daripada daun sirih iaitu kaedah penyulingan hidro dan ekstraksi bendalir lampau-genting. Eksperimen yang berbeza telah direka dan dijalankan untuk mengkaji kesan pelbagai parameter proses.

Bahagian pertama kajian ini mengkaji kesan jenis daun dan masa ekstraksi terhadap hasil dan komposisi eugenol dengan menggunakan kaedah penyulingan hidro yang konvensional. Hasil kajian menunjukkan bahawa daun sirih yang segar lebih dipilih daripada daun sirih yang kering. Enam jam adalah bersesuaian untuk mengekstrak daun segar.

Bahagian kedua penyelidikan ini adalah mengkaji kesan tekanan dan suhu terhadap hasil dan kepekatan eugenol dengan menggunakan kaedah ekstraksi bendalir lampau-genting. Kaedah Tindak Balas Permukaan (RSM) telah digunakan untuk mendapatkan parameter proses yang optimum. Tekanan dan suhu optimum yang telah dicadangkan oleh RSM adalah 190 bar dan suhu 50 °C. Sifat yang dijangka pada ekstrak adalah 0.5% hasil dan 21.57 mg/ml kepekatan eugenol.

Model kinetik telah digunakan untuk menghuraikan fenomena pemindahan jisim. Nilai tertinggi untuk pekali pemindahan jisim ialah 0.208 min^{-1} pada tekanan 160 bar dan suhu $50 \text{ }^\circ\text{C}$. Model Del Valle-Aguilera menunjukkan padanan yang terbaik dengan data eksperimen di mana nilai purata sisihan relatif mutlak AARD ialah paling rendah. Persamaan ini disyorkan untuk kelarutan minyak sirih di dalam ekstraksi bendalir lampau-genting.

Perbezaan antara teknik ekstraksi bendalir lampau genting dan penyulingan hidro telah dikaji. Teknik ekstraksi bendalir lampau genting memberi banyak kelebihan yang penting berbanding penyulingan hidro. Oleh itu, teknik ini boleh dianggap sebagai teknologi yang berbeza untuk pengekstrakan daun sirih, bukan hanya diisytiharkan mengenai kebaikannya berbanding teknik konvensional tetapi juga kerana kepekatan sasaran sebatian dan hasil yang tinggi bersama masa pengekstrakan yang pendek (1 jam).

ACKNOWLEDGEMENT

First and foremost, I wish to acknowledge my supervisor, Professor Dr. Luqman Chuah Abdullah for the encouragement and endless guidance I have received from him, are much appreciated. Besides my advisor, I would like to thank the rest of my supervisory committee: Associate Professor Dr. Yus Aniza Yusof, Professor Dr. Md. Zaidul Islam Sarker and Dr. Nor Azah Mohamad Ali, for their helps, encouragement and insightful comments.

My appreciation is also extended to staff from Forest Research Institute Malaysia (FRIM) especially Dr. Pin Kar Yong and Saidatul Husni Saidin for their technical assistant and kind knowledge-sharing. Also special thanks to all the staff members of Department of Process & Food Engineering, Department of Chemical & Environmental Engineering and Faculty of Food Science and Technology, UPM for their helps and corporation.

Last but not least, I would like to express my deepest gratitude to my family especially to my husband Ariff Izuddin Zakaria for their constant encouragement, scarifies and understanding.

I certify that a Thesis Examination Committee has met on **27 December 2012** to conduct the final examination of Noor Hadzuin Bt Nik Hadzir on her thesis entitled "**Extraction of Eugenol from Betel Leaves (*Piper betle* L.) using Hydrodistillation and Supercritical Carbon Dioxide Extraction Technique**" in accordance with the Universities and University Colleges Act 1971 and the Constituent of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

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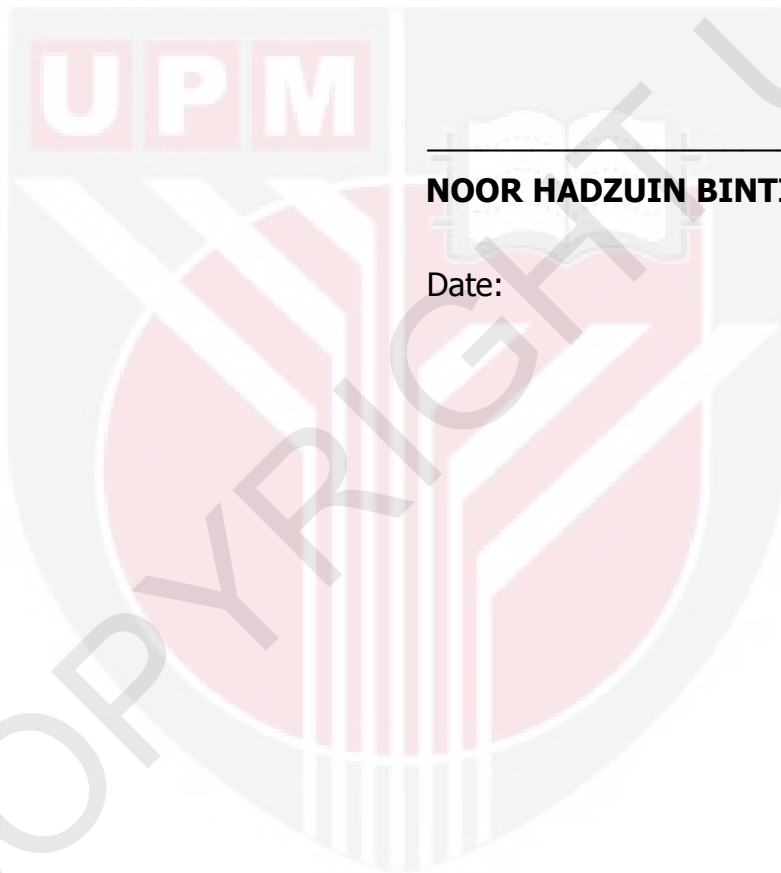
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DECLARATION

I declare that the thesis is my original work except for quotations and citation which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any degree at Universiti Putra Malaysia or at any other institutions.



NOOR HADZUIN BINTI NIK HADZIR

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