



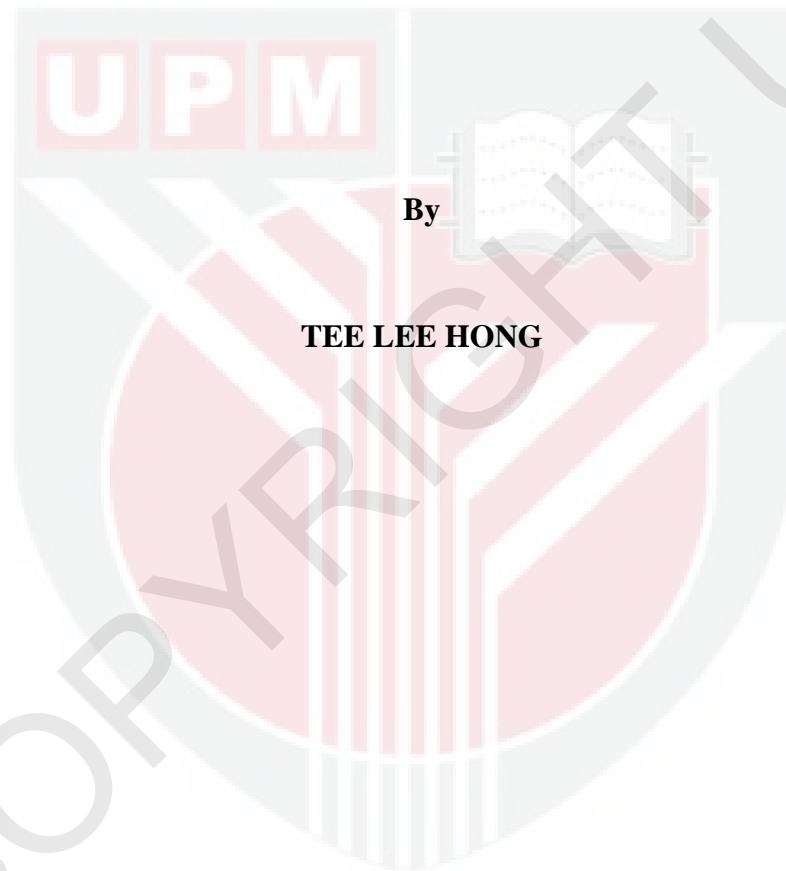
**UNIVERSITI PUTRA MALAYSIA**

***RHEOLOGICAL PROPERTIES AND OPTIMAL SPRAY DRYING  
PROCESS PARAMETERS OF PIPER BETLE L. (BETEL) LEAF  
EXTRACTS COATED WITH DIFFERENT EXCIPIENTS***

**TEE LEE HONG**

**FK 2011 167**

**RHEOLOGICAL PROPERTIES AND OPTIMAL SPRAY DRYING  
PROCESS PARAMETERS OF *PIPER BETLE L.* (BETEL) LEAF  
EXTRACTS COATED WITH DIFFERENT EXCIPIENTS**



By

**TEE LEE HONG**

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,  
in Fulfillment of the Requirements for the Degree of Master of Science**

**November 2011**

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

**RHEOLOGICAL PROPERTIES AND OPTIMAL SPRAY DRYING  
PROCESS PARAMETERS OF *PIPER BETLE L.* (BETEL) LEAF  
EXTRACTS COATED WITH DIFFERENT EXCIPIENTS**

By

**TEE LEE HONG**

**November 2011**

**Chair: Professor Luqman Chuah Abdullah, PhD**

**Faculty: Engineering**

*Piper betle L.*, more commonly known as betel or local name of *Sirih*, belongs to the family *Piperaceae*. Previous research has shown that the leaves of *P. betle* possess tremendous beneficial effects including antimicrobial, antioxidant, anti-diabetic, wound healing and gastro-protective properties. The presence of these beneficial properties indicates that leaf extract of betel has great potential for development into a wide range of health food supplements. However, there is a lack of research on the processing aspects to produce its bioactive component.

This research studied two main aspects including the rheological properties of betel leaves extract and optimization of spray drying process coated with three excipients which were xanthan gum, maltodextrin and lactose. The main purpose for studying the rheological properties of betel leaves is to prepare data that would be useful in designing processing equipment for betel leaves extract products. Spray drying was

chosen for easy handling and the preservation of bioactive compounds. The process parameters of spray drying were inlet hot air temperature, feed flow rate or pump flow rate and aspirator rate. The properties of dried powder produced were investigated in terms of particle size distribution, moisture content and bioactive component; hydroxychavicol (HC) content, hygroscopicity and powder yield. The experimental run and optimization work were designed using Box-Behnken method of Response Surface Methodology (RSM).

Proximate analysis on *Piper betle L.* freeze-dried extract was successfully conducted. The compositions consist of 16.45% moisture, 28.18% ash, 9.36% protein, 0.11% crude fiber, 0.16% crude fat and 45.74% carbohydrate. Rheology behavior of 10 °Brix and 12 °Brix concentration of betel leaves extract at temperature 5, 10, 20, 30, 40 and 50 °C was successfully investigated. Bingham Plastic Model and Casson model illustrated that betel leaves extract showed a plastic behavior. Power Law model was suitable to represent the rheology model of betel leaves extract.

The optimum operation conditions for the highest hydroxychavicol content with the lowest moisture content and the smallest particle size for xanthan gum-coated powder were obtained at inlet drying temperature of 160 °C, the pump flow rate of 3 rpm, and airflow rate of 86%. The optimum responses of the process were: moisture content of 10.14%, particle size distribution of 7.37 µm, powder yield of 6.72 g, powder hygroscopicity of 26.76 g/100g dry powder and HC content of 383.19 ppm. For maltodextrin-coated spray drying process, the optimum processing parameters were as follow: the inlet drying temperature of 159.53°C, the pump flow rate of 6.13

rpm, and airflow rate of 98.33%. The optimum responses of the process were: moisture content of 6.99%, particle size distribution of 5.48  $\mu\text{m}$ , powder yield of 10.53 g, powder hygroscopicity of 28.88 g/100g dry powder and HC content of 229.33 ppm. While for lactose-coated spray drying process, the optimum processing parameters were as follow: the inlet drying temperature of 160 °C, the pump flow rate of 5.33 rpm, and airflow rate of 100%. The optimum responses of the process were: moisture content of 6.96%, particle size distribution of 6.58  $\mu\text{m}$ , powder yield of 16.54 g, powder hygroscopicity of 16.58 g/100g dry powder and HC content of 80.23 ppm. Xanthan gum was found out to be the most suitable carrier for the spray drying process of betel leaves extract as it contained the highest amount of HC content and showed better morphological properties which are more spherical in shape, more consistent in size and less shrinkage present on the surface of the particle.

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

**RHEOLOGICAL PROPERTIES AND OPTIMAL SPRAY DRYING  
PROCESS PARAMETERS OF *PIPER BETLE L.* (BETEL) LEAF  
EXTRACTS COATED WITH DIFFERENT EXCIPIENTS**

Oleh

**TEE LEE HONG**

**November 2011**

**Pengerusi: Profesor Luqman Chuah Abdullah, PhD**

**Fakulti: Kejuruteraan**

*Piper betle L.*, lebih dikenali sebagai nama betel atau nama tempatan Sirih; tergolong dalam keluarga *Piperaceae*. Kajian sebelum ini menunjukkan bahawa daun sirih mempunyai kesan berfaedah termasuk antioksidan, antimikrob, anti-diabetes, dan penyembuhan luka. Kehadiran sifat-sifat yang bermanfaat ini menunjukkan bahawa ekstrak daun sirih mempunyai potensi besar dalam pembangunan pelbagai produk.

Tetapi, penyelidikan dalam aspek-aspek pemrosesan untuk menghasilkan komponen bioaktifnya masih tidak mencukupi.

Penyelidikan ini merangkumi dua aspek utama termasuk kajian tentang sifat-sifat reologi ekstrak daun sirih dan pengoptimuman proses pengeringan semburan ekstrak daun sirih yang disalut dengan tiga agen lapisan iaitu xanthan gum, maltodextrin dan laktosa. Tujuan utama untuk mengkaji sifat-sifat reologi daun sirih adalah untuk menyediakan data yang berguna dalam mereka bentuk peralatan pemrosesan untuk

produk ekstrak daun sirih. Pengeringan semburan telah dipilih untuk mengendalimudahkan dan pemeliharaan sebatian bioaktif. Parameter proses pengeringan semburan kajian ini termasuk suhu udara panas, aliran kadar atau kadar aliran pam dan kadar penyedut. Serbuk yang dihasilkan telah dianalisis untuk kandungan sebatian aktif, Hydroxychavicol (HC), taburan saiz zarah, kandungan lembapan, keupayaan untuk menyerap air dan hasil serbuk. Larian eksperimen dan pengoptimuman telah direka bentuk menggunakan kaedah Box Behnken seperti yang disarankan oleh Response Surface Methodology (RSM).

Komposisi ekstrak sirih terdiri daripada kelembapan 16.45%, 28.18% abu, protein 9.36%, 0.11% serat mentah, 0.16% lemak dan karbohidrat 45.74%. Rheologi untuk 10 ° Brix dan 12 ° Brix ekstrak daun sirih pada 5 suhu, 10, 20, 30, 40 dan 50 °C telah berjaya disiasat. Bingham Model Plastik menggambarkan bahawa ekstrak daun sirih menunjukkan kelakuan plastik. Power law tidak sesuai untuk mewakili model rheologi daun sirih ekstrak. Casson model mendedahkan bahawa daun sirih ekstrak mempamerkan sifat plastik.

Keputusan yang diperolehi daripada RSM menunjukkan bahawa process parameter optima; suhu pengering 160 °C, aliran pam kadar 3 rpm, dan kadar aliran udara sebanyak 86%. Respons optima proses ialah: kandungan kelembapan 10.14%, saiz taburan zarah 7.37 µm, hasil serbuk 6.72 g, keupayaan menyerap air, 26.76 g/100g serbuk kering dan kandungan HC, 383.19 ppm. Untuk maltodextrin, parameter pemprosesan yang optima adalah seperti berikut: suhu pengering 159.53 °C, kadar aliran pam 6.13 rpm, dan kadar aliran udara dari 98,33%. Respons optima proses ialah: kandungan kelembapan 6.99%, saiz taburan zarah, 5.48 µm, hasil serbuk 10.53

g, keupayaan menyerap air, 28.88 g/100g serbuk kering dan kandungan HC, 229.33 ppm. Manakala bagi process bersalut dengan laktosa, parameter pemprosesan yang optima adalah seperti berikut: suhu pengeringan 160 °C, kadar aliran pam 5.33 rpm, dan kadar aliran udara sebanyak 100%. Respons optima proses ialah: kandungan kelembapan 6.96%, saiz taburan zarah 6.58  $\mu\text{m}$ , hasil serbuk 16.54 g, keupayaan menyerap air, 16.58 g/100g serbuk kering dan kandungan HC, 80.23 ppm. Xanthan gum didapati merupakan agent lapisan yang paling sesuai untuk proses semburan pengeringan daun sirih ekstrak. Dari segi morfologi, ia lebih berbentuk sfera, lebih konsisten dari segi saiz zarah dan tiada pengecutan hadir di permukaannya.



## ACKNOWLEDGEMENT

First of all, I need to thank my University, Universiti Putra Malaysia (UPM) for giving me a chance to carry out my Master research project to fulfill the requirement for completing my Master Science of Process Engineering. Through this research study, I've gained lots of precious experiences about the real research methodology, which I cannot get it through lectures and textbooks.

I would like to take this opportunity to express my heartfelt gratitude to my project advisor and the chairman of my supervisory committee, Professor Dr. Luqman Chuah Abdullah and my co-supervisor, Associate Prof. Dr. Yus Aniza Yusof and Dr. Pin Kar Yong who gave me a lot of expert guidance, opinions and their willingness to spend their precious time to help me in accomplishing this master research project. Without their guidance, this project may not have reached completion. I would also like to thank the staff from Medicinal Plants Division of Forest Research Institute Malaysia (FRIM) especially Tn. Haji Abdull Rashih Ahmad and staff from Food Science and Technology Faculty, UPM, for their technical assistance and kind knowledge sharing.

Finally, I would like to express my warmest appreciation my family and my beloved boyfriend, Dr. Liew Yuan Hwen for their never ending support. My family's love and support were crucial for me to pass through all difficult stages in the completion of this final year project. A million thanks also goes to all my friends and coursemates who have made the journey of completing this training a successful and memorable one.

I certify that an Examination Committee has met on 14<sup>th</sup> November 2011 to conduct the final examination of Tee Lee Hong on her Master of Science thesis titled “Rheological Properties and Optimization of Spray Drying Process of *Piper betle L.* (Sirih) Leaves Extract” in accordance with the Universities and University Colleges Act 1971 and the Constitution of Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

Members of the Thesis Examination Committee were as follows:

**Farah Saleena Taip, PhD**

Senior Lecturer  
Faculty of Engineering  
Universiti Putra Malaysia  
(Chairman)

**Mohd Nordin Ismail, PhD**

Associate Professor  
Faculty of Engineering  
Universiti Putra Malaysia  
(Internal Examiner)

**Zurina Zainal Abidin, PhD**

Senior Lecturer  
Faculty of Engineering  
Universiti Putra Malaysia  
(Internal Examiner)

**Ramlan Abd. Aziz, PhD**

Professor  
Faculty of Engineering  
Universiti Teknologi Malaysia  
(External Examiner)

---

**SEOW HENG FONG, PhD**

Professor and Deputy Dean  
School of Graduate Studies  
Universiti Putra Malaysia

Date: 21 May 2012

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

**Luqman Chuah Abdullah, PhD**

Professor  
Faculty of Engineering  
Universiti Putra Malaysia  
(Chairman)

**Yus Aniza Yusof, PhD**

Associate Professor  
Head, Department of Process and Food Engineering  
Faculty Engineering  
Universiti Putra Malaysia  
(Member)

**Pin Kar Yong, PhD**

Medicinal Plants Division  
Herbal Technology Centre  
Forest Research Institute Malaysia  
(Member)

---

**BUJANG BIN KIM HUAT, PhD**

Professor and Dean  
School of Graduate Studies  
Universiti Putra Malaysia

Date:

## DECLARATION

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



---

**TEE LEE HONG**

Date: 14 November 2011

## TABLES OF CONTENTS

	<b>Page</b>
<b>ABSTRACT</b>	ii
<b>ABSTRAK</b>	v
<b>ACKNOWLEDGEMENTS</b>	viii
<b>APPROVAL</b>	ix
<b>DECLARATION</b>	xi
<b>LIST OF TABLES</b>	xvi
<b>LIST OF FIGURES</b>	xviii
<b>CHAPTER</b>	
<b>1 INTRODUCTION</b>	<b>1</b>
1.1 Herbal industry in Malaysia	1
1.2 Herbal processing	2
1.3 Problem statement	3
1.4 Objectives	5
1.5 Research Scope	5
<b>2 LITERATURE REVIEW</b>	<b>7</b>
2.1 <i>Piper betle</i> Linn (betel)	7
2.1.1 Hydroxychavicol and Eugenol of betel leaves extract	10
2.2 Extraction of <i>Piper betle</i> L. leaves	11
2.3 Proximate analysis	12
2.3.1 Determination of ash content	13
2.3.2 Determination of moisture content	13
2.3.3 Determination of crude protein by Kjeldahl method	13
2.3.4 Determination of crude fiber by Weede method	14
2.3.5 Determination of crude fat by Soxhlet method	15
2.3.6 Determination of carbohydrates	15
2.4 Rheological properties analysis	15
2.4.1 Newtonian behavior	17
2.4.2 Non-Newtonian behavior	17
2.4.3 Bingham Plastic Model	18
2.4.4 Power Law Model	19
2.4.5 Casson Model	19
2.4.6 Time Independent Fluid Behavior in Steady Shear Flow	20
2.5 Spray Drying	21
2.6 Spray Drying Principles	24
2.6.1 Atomization	25
2.6.2 Mixing and Drying	26
2.6.3 Powder Separation	29
2.7 Key Parameters Affecting Spray Drying Process	29
2.8 Advantages of Spray Drying	30

2.9	Spray drying carrier or wall material	31
2.9.1	Maltodextrin	33
2.9.2	Lactose	35
2.9.3	Xanthan gum	35
2.10	Response Surface Methodology	36
2.10.1	Box-Behnken Design (BBD)	39
<b>3</b>	<b>METHODOLOGY</b>	42
3.1	Introduction	42
3.2	Materials	42
3.3	Preparation of betel leaves extract	44
3.4	Proximate analysis on the <i>Piper betle L.</i> extract	44
3.4.1	Determination of ash content	44
3.4.2	Determination of moisture content	45
3.4.3	Determination of crude protein content by Kjeldahl method	46
3.4.4	Determination of crude fiber by Weede method	46
3.4.5	Determination of crude fat by Soxhlet method	47
3.4.6	Determination of carbohydrates	48
3.5	Rheological Behavior Analysis	48
3.5.1	Material	48
3.5.2	Rheological Analysis Experiments	49
3.6	Spray drying process	50
3.6.1	Preparation of betel leaves extract	50
3.6.2	Preparation of additives	50
3.6.3	Preparation of feed solution	52
3.6.4	Spray drying of betel leaves extract	52
3.7	Analytical methods	53
3.7.1	Content of hydroxychavicol	53
3.7.2	Moisture content	54
3.7.3	Particle size distribution	54
3.7.4	Hygroscopicity test	55
3.7.5	Yield	55
3.8	Experimental design	55
3.8.1	Statistical Analysis	57
3.9	Validation of the optimization model	57
3.10	Optimized particle's morphology	57
<b>4</b>	<b>RESULTS AND DISCUSSIONS</b>	58
4.1	Proximate Analysis	58
4.2	Rheological study	59
4.3	Comparative performance of selected rheological model	59
4.3.1	Bingham Plastic Model	59
4.3.2	Power Law Model	61
4.3.3	Casson Model	63

4.4	Spray drying process	67
4.4.1	Introduction of spray drying process	67
4.5	Preliminary studies on selecting the concentration of different carriers	68
4.6	RSM analysis of <i>Piper betle</i> extract spray drying process coated with xanthan gum	69
4.6.1	Hydroxychavicol content	71
4.6.2	Moisture content	74
4.6.3	Powder particle size distribution	77
4.6.4	Powder hygroscopicity	79
4.6.5	Powder yield	81
4.6.6	Optimization of process parameters on spray drying process coated with xanthan gum	83
4.6.7	Validation of the predictive models	84
4.6.8	Optimized powder's morphology	84
4.7	RSM analysis of <i>Piper betle</i> extract spray drying process coated with maltodextrin	87
4.7.1	Hydroxychavicol content	88
4.7.2	Moisture content	91
4.7.3	Powder particle size distribution	93
4.7.4	Powder hygroscopicity	95
4.7.5	Powder yield	97
4.7.6	Optimization of process parameters on spray drying process coated with maltodextrin	99
4.7.7	Validation of the predictive models	100
4.7.8	Optimized powder's morphology	101
4.8	RSM analysis of <i>Piper betle</i> extract spray drying process coated with lactose	103
4.8.1	Hydroxychavicol content	105
4.8.2	Moisture content	107
4.8.3	Powder particle size distribution	108
4.8.4	Powder hygroscopicity	111
4.8.5	Powder yield	112
4.8.6	Optimization of process parameters on spray drying process coated with lactose	115
4.8.7	Validation of the predictive models	116
4.8.8	Optimized powder's morphology	117
4.9	Comparisons of three carriers; xanthan gum, maltodextrin and lactose	119
<b>5</b>	<b>CONCLUSIONS</b>	<b>121</b>
5.1	Conclusions on Proximate analysis and Rheological analysis	121
5.2	Conclusion on optimization of Spray drying process parameters	122
5.2.1	Spray drying of betel leaves extract coated with xanthan gum	122
5.2.2	Spray drying of betel leaves extract	

	coated with maltodextrin	123
5.2.3	Spray drying of betel leaves extract coated with lactose	125
5.3	Comparisons of three carriers	126
5.4	Future Recommendations	126
5.4.1	Feed formulation	127
5.4.2	Powder specifications to be studied	127
5.4.3	Study on other drying methods	127
5.4.4	Study on other potential medicinal plants	128
	<b>REFERENCES</b>	129
	<b>APPENDICES</b>	138
	<b>BIODATA OF STUDENT</b>	146

