SOLUBILITY OF *SWIETENIA MACROPHYLLA* SEEDS OIL IN SUPERCRITICAL CARBON DIOXIDE EXTRACTION

MOHAMMAD LOKMAN BIN HILMI

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School of Chemical and Energy Engineering
Faculty of Engineering
Universiti Teknologi Malaysia

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ABSTRACT

Swietenia macrophylla (*S.macrophylla*) is one of the members of the family Meliaceae. *S.macrophylla* is commonly known as *Swietenia mahagoni* in Indonesia and locally known as Tunjuk Langit in Malaysia. It is very popular for various treatments and it contains many useful health properties such as antibacterial, antioxidant, anti-inflammatory and antidiabetic. The main purpose of this research was to investigate the effect of temperature and pressure on the oil yield and solubility of *S.macrophylla* seeds oil using supercritical carbon dioxide (SC-CO_2). Understanding the behavior of the SC-CO_2 separation process is fundamental in designing this process. Another objective was to determine the best suited solubility model for *S.macrophylla* seeds oil using five semi-empirical solvent density-based models. The solubility of *S. macrophylla* seeds oil in SC-CO_2 was determined by using the dynamic method at 20-30 MPa and 40-60 ^oC at the fixed CO_2 flow rate of 2 mL/min. The results showed that an increase in temperature and pressure would increase the oil yield. The highest extract and solubility of *S.macrophylla* oil was obtained at the highest extraction condition of 60 ^oC and 30 MPa with 803 mg oil/g sample and 4.77 mg/g CO_2 respectively. In this study, five semi-empirical models were tested to fit the experimental data. Results showed that Sparks model was the best-fitted model for *S. macrophylla* seeds oil solubility data with an average absolute relative deviation of 7.76% and a high coefficient of determination, R^2, of 0.992. It can be concluded that the result of this study can be used as reference in widening the scope of research in the area of pressure and temperature with a better improvement of predictive ability.

ABSTRAK

Swietenia macrophylla (*S.macrophylla*) adalah salah satu daripada ahli keluarga Meliaceae. *S.macrophylla* biasanya dikenali sebagai *Swietenia mahagoni* di Indonesia atau lebih dikenali sebagai Tunjuk Langit di Malaysia. Ia adalah sangat popular untuk pelbagai rawatan dan ia mempunyai pelbagai sifat kesihatan seperti antibakteria, antioksidan, anti-radang dan antidiabetik. Tujuan utama kajian ini adalah untuk mengkaji kesan suhu dan tekanan terhadap hasil minyak dan keterlarutan minyak biji *S.macrophylla* menggunakan karbon dioksida superkritikal (SC-CO_2). Pemahaman tingkah laku proses pemisahan SC-CO_2 merupakan asas dalam merekabentuk proses. Objektif yang seterusnya adalah untuk menentukan model keterlarutan yang terbaik untuk minyak biji *S.macrophylla* menggunakan lima model berdasarkan ketumpatan pelarut semi-empirikal. Keterlarutan minyak biji *S.macrophylla* dalam SC-CO_2 telah ditentukan dengan menggunakan kaedah dinamik pada 20-30 MPa dan 40-60 ^oC pada kadar aliran tetap 2 mL / min CO_2. Hasil kajian menunjukkan bahawa peningkatan suhu dan tekanan meningkatkan pengeluaran hasil minyak. Ekstrak tertinggi dan keterlarutan *S.macrophylla* telah diperolehi pada keadaan pengeluaran yang tertinggi 60 ^oC dan 30 MPa masing-masing dengan 803 minyak mg/g sampel dan 4.77 mg/g CO_2. Dalam kajian ini, lima model semi-empirikal telah diuji untuk dipadankan dengan data eksperimen. Hasil kajian menunjukkan model Sparks adalah yang terbaik untuk keterlarutan minyak biji *S.macrophylla* dengan data purata sisihan mutlak relatif dengan 7.76% dan pekali yang tinggi penentuan, R^2, 0.992. Ini dapat disimpulkan bahawa hasil kajian ini boleh digunakan sebagai rujukan untuk meluaskan skop penyelidikan dalam bahagian tekanan dan suhu dengan peningkatan yang lebih baik keupayaan ramalan.

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LIST OF ABBREVIATIONS

AAD	-	Absolute average deviation
AARD	-	Absolute average relative deviation
ASE	-	Accelerated solvent extraction
CER	-	Constant extraction rate
CLEAR	-	Centre of Lipids Engineering & Applied Research
CO_2	-	Carbon dioxide
COD	-	Coefficient of determination
FER	-	Falling extraction rate
MAE	-	Microwave assisted extraction
RSM	-	Response surface methodology
SC-CO_2	-	Supercritical carbon dioxide
SFE	-	Supercritical fluid extraction
TGC	-	Total glucosinolate content
TPC	-	Total phenolic content
S.D	-	Standard deviation

LIST OF SYMBOLS

c_0,c_1,c_2,c_3,c_4,c_5	-	Constant in Gordillo model
a,b ,c	-	Constant in solubility equation
^oC	-	Degree celsius
ρ	-	Density
ρ_{scs}	-	Density supercritical solvent
ΔH_{solv}	-	Heat of solvation
ΔH_{vap}	-	Heat of vaporization
W_{oil}	-	Mass of oil extract
W_i	-	Mass of sample (before)
W_f	-	Mass of sample (final)
MPa	-	Mega Pascal
P	-	Pressure
Y*	-	Solubility
T	-	Temperature
y_2	-	Solubility (Mole fraction)
C_2	-	Solubility (g L^{-1})
T	-	Absolute temperature (K)
T_1	-	Temperature (^OC)
ρ_1	-	Molar density of fluid (mol L^{-1})
ρ_2	-	Density (g mL^{-1})
α	-	Dimensionless function of reduce temperature and acentric factor, unity at critical temperature
α_{cr}	-	Attraction parameter at critical temperature
b	-	Van der Waals covolume
c	-	Equation of state parameter
k	-	Association number of solute
ω	-	Acentric factor
z	-	Compressibility factor

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CHAPTER 1

INTRODUCTION

1.1 Background of Research

Swietenia macrophylla is well-known in Asian countries, South America, Mexico and Hawaii. According to U.S National Plant Germplasm System (2017), *S.macrophylla* is usually known as mahogany, Honduran mahogany, aguano, caoba and hondurasmahogny. *S.macrophylla* is local to the couple of tropical districts of the Americas, including Southern Mexico, Central America and Bolivia (Tan *et al.*, 2009). The characteristic of *S.macrophylla* fruit seem to hang upwards to the sky from the tree and thus commonly called as "sky fruit" or in Malaysia, locally known as 'Tunjuk Langit'. It is a tropical timber tree that can develop to a stature of 40-60 m, with straight trunk and barrel shaped with 100 to 200 cm tallness

Assoc.Prof.Dr. Liza Md Salleh began her study about *S.macrophylla* on 2014 with research on the effect of supercritical fluid extraction parameters on the *S.macrophylla* seed oil extraction and its cytotoxic properties. Then, in 2015, she furthered her research by doing solubility data study using 2 semi-empirical model namely as Chrastil (1982) model and del Valle-Aguilera (1988) model. Nowadays, there is continuation on the research regarding solubility data with addition several and latest model which are Adachi-Lu (1983), Gordillo (1999) and Sparks model (2008). With the data gathered along the years of research, she managed to create few products from *S.macrophylla* seeds oil known as Softgel and Niomahogel. *S.macrophylla* is one of the plants that are commonly used in the community far and wide particularly in a few nations which have high dissemination of this species. Over the years, traditional medicines extracted from plant have been practiced by local neighborhood networks since long time prior to treat diverse sort of ailment, for example, diabetes without adverse effect. It is very popular for various treatment and have example of properties that are very useful in pharmaceutical such as antibacterial, antioxidant, anti-inflammatory and antidiabetic activity (Salsabila *et al.*, 2018).

Extraction process is the first step in analysis of plants as it is important in order to extract bioactive compound for further research. In general, extraction technique can be divided into two which is; conventional/traditional method and modern extraction method. Most of the conventional method has been known by others but for modern extraction method which is still new, not yet fully known. Well known conventional method consist of Soxhlet, sonification and maceration whereas modern method such as supercritical fluid extraction (SFE), pressurized-liquid extraction and microwave assisted extraction. In this study, SFE method was used and its function to separate the extractant from the matrix utilizing supercritical liquids as the separating solvent. Typically, extraction is from a solid matrix, yet it can likewise from fluids. As per Djas and Henczka (2018), elimination of organic solvents from modern process and their substitution with supercritical liquids, especially, supercritical carbon dioxide (SC-CO_2) have been seriously researched. This substitution is essential strategy so as to make to make the modern procedures all the more environmentally friendly and reducing the amounts of hazardous waste.

Generally, solubility defined as amount of substances (solute) that dissolve in a substances (solvent) to form a saturated solution under specified conditions of temperature and pressure. The solubility of the solutes in a supercritical solvents depends on several factors namely temperature, pressure, modifier, solvent flow rate, extraction time and solvent density. For solubility behavior, the elevation of temperature leads to reduce in solvation power at constant pressure but increasing the volatility of solute, hence increase the solubility of solute. Furthermore, according to Ahmad Ramdan (2015), to increase the solubility of the solute, modifiers or co-solvents can be used to decrease the crossover pressure for the system. There are fundamentally three kinds of methods utilized which are static, distribution and continuous flow (dynamic) techniques to gauge the solubility of solutes in supercritical liquids. Solubility is generally estimated either dynamic or static method, in which, solvent is straightforwardly contact with either liquid or ground material (N. Nguyen *et al.*, 2011).

Modeling can be defined in mathematical term as method of simulating real-life with mathematical equations to forecast their future behavior. Mathematical model are used to correlate with solubility data in order to determine the equilibrium solubility of solute in a supercritical fluid in different kinds of operating conditions. This is because determination of solubility data for solid solubility in every different point of temperature and pressure experimentally is difficult, time consuming and not possible due to some limitation such as maximum operating conditions on equipment (Yaw Choong *et al.*, 2013). Empirical fit is the most straight-forward and easiest way of correlating data, where relationship between solubility data and thermodynamic properties such as pressure and density is fitted directly either linear or other form.

The first model proposed for solubility data modeling was by Chrastil (1982). The study expressed a model equation as the correlation between temperature, density of supercritical fluid and concentration of solute in supercritical fluids. Then, the model was modified by others such as Adachi and Lu (1983) which the authors modified Chrastil (1982) equation to better model the solubility of triglyceride. The equation was proposed with assumption density has big influences in the extraction process. Other modified Chrastil (1982) equation was then proposed by del Valle and Aguilera (1988) which considered the change in the enthalpy of vaporization with temperature and assumed that the association number k is independent of density and c is the additional constant for the model. Gordillo (1999) proposed new semi-empirical model in order to improve correlation of the experimental data and introduced 6 adjustable parameter in the equation. In addition to that, Sparks (2008) proposed another modification which has been considered as latest modification on Chrastil (1982) model to binary system which combined two correction from Adachi-Lu (1983) and del Valle-Aguilera (1988).

As of late, the incredible development of pharmaceutical market and sustenance industry resulting high demand in solubility data information in extraction for better analysis and results. Their application in pharmaceutical and food industry are numerous, and also used in cosmetic industry. Solubility data gives valuable indication about the biological activity and behavior of the drug along the process, and therefore most important in extraction studies. Furthermore, having solubility data with high percent correlation with mathematical modeling will be able to boost up the throughput of extraction process and able to do valid prediction with wide range of parameters such as pressure and temperature.

1.2 Problem Statement

Extraction term commonly used in separation process, consisting in the separation of solute or substances from a matrix. Extraction methods such as Soxhlet and sonification are commonly used and have significant disadvantages. The major disadvantages of this conventional methods are extremely time-consuming, large volume of expensive and inconsistent (Tankeviciute *et al.*, 2001). Due to this disadvantages of conventional method, the supercritical fluid extraction (SFE) method is becoming more, and more important. Recently, the application SFE is extensively researched. SFE provides many interesting features overcoming conventional method. Due to their characteristic of low viscosity and relatively high diffusivity, SFE enhanced the transport properties than liquid, can diffuse through small pore medium easily and therefore give faster extraction rates (da Silva *et al.*, 2016). Pressure and temperature are the main parameter for SFE extraction as it plays importance role to the overall extraction process (Hartati *et al.*, 2014).

Solubility data are essential for better understanding of behaviour in separation process which is fundamental in the designing process and development of new drug. Other than that, oil solubility data are critical for future reference, for example, for scale up creation and at a few phases in the advancement of SC-CO2 extractions in term of process plan, hardware measuring, achievability assessments and building up ideal working conditions. Existing research on oil from *S.macrophylla* seeds in SC-CO_2 are still limited in term of solubility data for wide range of temperatures and pressures due to limitations of equipment itself such as maximum pressure, expensive and complex.

Nowadays, mathematical model are used to overcome this problem which valid for a wide range of pressure and concentrations prediction (Dwi *et al.*, 2016). Previous study by Ahmad Ramdan (2015) in the same research applied two semi-empirical density- based models which are Chrastil (1982) model and del Valle-Aguilera (1988) model. As for this study, improvement had been made with addition of several and updated models which are Gordillo (1999) model, Adachi-Lu (1983) model and Sparks (2008) model which can improve the correlation with experimental data, hence more accurate prediction for solubility data. Therefore, this research is conducted due to the lack of data from previous literature regarding mathematical model of solubility data from *S.macrophylla* seeds oil using SC-CO_2 extraction.

1.3 Research Objectives

This study is the continuation research on parameters effect and solubility data of *S.macrophylla* seeds oil using semi-empirical modeling. The objectives of this research are:

1. To investigate the effect of temperature and pressure on the oil yield and solubility of *S.macrophylla* seeds oil using SC-CO_2.
2. To determine best suited solubility model for *S.macrophylla* seeds oil using five semi-empirical solvent density-based model.

1.4 Scope of Research

In order to the achieve objectives as stated in section 1.3, the scopes of study were as below:

1. Determination of extraction yield and solubility data of *S.macrophylla* seeds oil using supercritical carbon dioxide extraction at CO_2 flow rate 2 ml/min for 120 min with temperature (40-60^{o}C) and pressure (20-30 MPa). Pressure was

limited until 30 MPa due to constraints of equipment used whereas for extraction temperature was at 60^oC for maximum as above that denatured bioactive compound in seeds oil.

2. Correlation of solubility data using five semi-empirical solvent density- based model which are Chrastil (1982), Gordillo (1999), Adachi-Lu (1983) and del Valle-Aguilera (1988) and Sparks (2008) and validated using coefficient of determination (COD) and absolute average relative deviation (AARD).

1.5 Significance of Research

The significance of this study can be seen in several ways. First, the research will give benefits to local farmer as this research revealed the importance of this *S.macrophylla* plants. In addition to that, the uses of *S.macrophylla* and the usage of seeds can be promoted for its medicinal benefits such as antioxidant, antidiabetic and antimicrobial. Besides that, the abundance availability of the plant in Malaysia made it even more suitable for economic growth in the near future.

Other than that, an environmentally friendly technique is applied using SC-CO_2 which free from any dangerous chemical at low temperature and high pressure to ensure the bioactive compound safe and not denatured due to exposure of heat. Moreover, this research focused on obtaining the best conditions for oil extracted from *S.macrophylla* seeds and this conditions eventually can be used for future reference in up-scaling process. As high quality product is obtained using SC-CO_2, it can be used for product marketing although the initial set up cost is high, but the finished product quality can be sold at higher price. A recycle CO_2 pump also can be installed in the future to reduce the product's cost drastically.

Next is the modeling data can be used as a guideline for further up-scaling study. The establishment of database for solubility behavior provides a significant reference for further studies of supercritical carbon dioxide extraction of *S.macrophylla* seeds oil at different conditions.

1.6 Thesis Outline

This thesis is organize in 5 chapters. In Chapter 1, the background of research was discussed, the research objective, the research scope and the significance of the study. Two research objectives were stated in this chapter with their scopes of research.

In Chapter 2, the overview of general description of *S. macrophylla* plant, type of extraction involved, mathematical model for solubility modeling of *S. macrophylla* seeds oil in SC-CO_2.

Chapter 3 defines the method that were used for solubility data modeling of oil *Swietenia macrophylla* seeds oil using SC-CO_2. The method proposed focused on extraction of oil from the seeds, establishing solubility data of extract and correlation with five semi-empirical solvent density-based models.

In Chapter 4, the overall extraction yield and solubility data of *S. macrophylla* seeds oil using SC-CO_2 based on two factors namely pressure (MPa) and temperature (^oC). Furthermore, mathematical modeling was done by several semi-empirical models for solubility data correlation with experimental data which were Chrastil (1982), Adachi-Lu (1983), del Valle-Aguilera (1988), Gordillo (1999) and Sparks (2008) model.

Conclusion and recommendation were discussed in Chapter 5. It concludes on the findings in this research and future prospect of the research. Recommendations are also provided for future research idea and improvement.

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