Faculty of Engineering

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OPTIMIZATION STUDY ON MATHEMATICAL MODEL OF CYMBOPOGON *WINTERIANUS* ESSENTIAL OIL EXTRACTION BY STEAM DISTILLATION

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OPTIMIZATION STUDY ON MATHEMATICAL MODEL OF CYMBOPOGON WINTERIANUS ESSENTIAL OIL EXTRACTION BY STEAM DISTILLATION

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A dissertation submitted in partial fulfilment
of the requirement for the degree of
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Dedicated to my beloved late father and mother, supervisor, lecture and friends who always bestow me sustainable motivations, inspirations and encouragements

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ABSTRACT

N.A.Farhana

Citronella essential oil derived from Cymbopogon *winterianus* is one of the most widely traded essential oil in the aroma sector. As these oil content more than 50 terpenoidal blends and constituents, the oils are high in demand in aroma industry either as perfumery agent or as source of lead molecules to derived more useful value-added product that are required in high-grade cosmetics and drugs. The content of citronella oil in a freshly harvested raw material is only 0.5% to 1.0% and decreases as the plant dries out while waiting to be processed. Other than the limited life of the plantation and low yield, citronella oil also had been witnessing demand and price fluctuations as the result of proliferation of inexpensive synthetic isolates in the market. Therefore, it is important to study and develop optimum mathematical model for the extraction of Cymbopogon *winterianus* essential oil as mathematical model plays important factors in predicting the extraction of citronella oil. Yield is a major indicator of financial and productivity. Inefficient mathematical model may result in failure and inexact prediction of citronella yield thus leading to waste of capital cost expenditure. Thus in order to optimize the prediction of essential oil, five mathematical models developed by Cassel and Vargas (2006), Hervas et al (2006), Garkal et al (2012), Ana et al (2007) and Milojevi et al (2008) were studied and optimized. The adequacy of the fit of the models to the experimental data are analyzed by using three statistical criteria that are correlation coefficient (r) ,the root mean square error (RMSE) and the mean relative deviation modulus (E). The result has shown that the mathematical model developed by Ana et al (2007) based on mass transfer fundamentals is the optimum mathematical model for the extraction of Cymbopogon *winterianus* essential oil by steam distillation.

Keywords: Essential Oils, Cymbopogon *winterianus* essential oil, Optimization of mathematical model of extraction of citronella essential oils

ABSTRAK

N.A.Farhana

Minyak pati serai wangi yang diperolehi dari Cymbopogon *winterianus* adalah salah satu daripada minyak terpenting dalam sektor aroma. Oleh disebabkan kandungan minyak ini mempunyai lebih daripada 50 campuran terpenoidal dan kompaun kimia, minyak ini mendapat permintaan yang tinggi di dalam industri aroma sama ada sebagai sumber minyak wangi atau sebagai sumber molekul membawa kepada perolehan produk nilai tambah yang lebih berguna untuk meghasilkan kosmetik dan ubat-ubatan bergred tinggi. Kandungan minyak serai wangi dalam bahan mentah yang segar adalah di antara 0.5% sehingga 1.0% dan akan berkurangan sementara menunggu untuk diproses. Selain daripada jangka hayat ladang yang terhad dan hasil yang rendah, minyak serai wangi juga telah mengalami kadar turun naik permintaan dan harga akibat daripada penghasilan "*synthetic isolates*" yang murah di pasaran. Oleh itu, adalah penting untuk mengkaji dan membangunkan model matematik yang optimum untuk pengekstrakan minyak pati Cymbopogon winterianus oleh kerana model matematik memainkan faktor penting dalam meramalkan pengeluaran minyak serai wangi. Hasil pengeluaran minyak pati adalah petunjuk utama kewangan dan produktiviti. Model matematik yang tidak cekap boleh menyebabkan kegagalan dan ramalan hasil serai wangi yang tidak tepat membawa kepada pembaziran perbelanjaan kos modal. Oleh itu untuk mengoptimumkan ramalan minyak pati, lima model matematik yang dibangunkan oleh Cassel dan Vargas (2006), Hervas dkk (2006), Garkal dkk (2012), Ana dkk (2007) dan Milojevi dkk (2008) telah dikaji dan dioptimumkan. Kecukupan patut model kepada data ujikaji dianalisis dengan menggunakan tiga kriteria statistik iaitu "*correlation coefficient (r)*", "*root mean square error (RMSE)*" dan "*mean relative deviation modulus (E)*". Keputusan kajian menunjukkan bahawa model matematik yang dibangunkan oleh Ana dkk (2007) berdasarkan asas-asas pemindahan jisim adalah model matematik yang optimum untuk pengekstrakan minyak pati Cymbopogon winterianus melalui penyulingan wap.

Kata Kunci: Minyak Pati, Minyak Pati Cymbopogon *winterianus*, Pengoptimuman Model Matematik Pengekstrakan Minyak Pati Cymbopogon *winterianus*

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ABBREVIATIONS

E	Mean Relative Deviation Modulus
EDA	Electro-dermal Activity
EEG	Electroencephalogram
GC-MS	Gas Chromatography _ Mass Spectrometry
HIV	Human Immunodeficiency Virus
r	Correlation Coefficient
RMSE	Root Mean Square Error
SFE	Supercritical Fluid Extraction
USD	United States Dollar

NOMENCLATURE

˚C	Degree celcius
g	Gram
g/g	Gram per Gram
in	inch
J/kg.k	Joule per kilogram.kelvin
Kg	Kilogram
Kg/h	Kilogram per hour
kg/t	Kilogram per tonne
kJ/kg	Kilojoule per kilogramme
kPa	kiloPascal
kW/°C	Kilowatt/degree celcius
m	Meter
m^3/s	Meter cube per second
mg/in^2	Milligram per inch square
Mg/L	Milligram per litre
MHz	Mega Hertz
min	Minute
ml	Mililiter
MPa	Mega Pascal
MT	Mega Tonnes
s	Second
t	Tonne
t/yr	Tonne per year
W/m^2K	Watt per meter square kelvin

CHAPTER 1

INTRODUCTION

1.1 General

The purpose of this research is to optimize the mathematical model of Cymbopogon *winterianus* essential oil extraction by steam distillation developed by Cassel and Vargas (2006) based on their research "*Experiments and Modelling of Cymbopogon winterianus Essential Oil Extraction by Steam Distillation*". By optimization this mathematical model, the mathematical model is expected to be optimized by 5% in predicting yield of the essential oil extracted.

1.1.1 Cymbopogon *winterianus*

Cymbopogon winterianus is commonly known as Citronella. Citronella Grass and Java Citronella grass is a lemongrass species that is believed to have originated from Cymbopogon nardus that often referred to Ceylonese, a Sri Lankan commercial citronella. Cymbopogon winterianus was named after the Winter whom presented the plant as a separate species in 19^{th} century. It is later than introduced to Indonesia and commercially known as Javanese citronella. The plant later was further introduced to India in 1959 (Shasany *et al*, 2000).

Essential oils are subtle, natural, aromatic and volatile compounds which are extracted from the flower, seeds, leaves, stems, bark and roots of herbs (Peng *et al*, 2012). As agreed by Tajidin *et al* (2012), essential oils are natural products which can be extracted from plants. They were formed through mixture of varied and complex volatile chemical compounds with high proportion of terpene associated with aldehyde, ketone and alcohols. The different mixtures of the chemical then were deposited in different structure of the plant (Tajidin *et al*, 2012).

Essential oil can be extracted traditionally by hydrodistillation, steam distillation, or solvent extraction (Yusof, 2010). Kabuba (2013) also stated that essential oils derived from aromatic plant are typically extracted by steam distillation as this method is simple and relatively inexpensive process. The essential oil in the plant is removed by steam of water vapour and can be separate easily in the next phase (Kabuba, 2013). The aromatic industry often employed this extraction method as it is cheaper than advanced methods such as supercritical fluid extraction (SFE). The proportion of different essential oil extracted by steam distillation in the industry is 93% and the remaining 7% employed other extraction methods such as SFE and microwaves extraction (Kabuba J, 2013). This method is also preferred by Amenaghawon *et al* (2014) because steam distillation is flexible, versatile, do not lead to decomposition of essential oils and provides the possibility of operating with small volumes (Amenaghawon *et al*, 2014).

Essential oils played an important role in the personal and social hygiene of mankind in terms of their application in cosmetics, toiletries, medicinal formulation, surface coatings and aroma therapy (Koul *et al*, 2003). Essential oils have been largely applied in various areas due to their natural properties which exhibit antibacterial, antifungal, insecticidal, antiviral and anti-herbivore characteristics. To date, approximately 3000 essential oils are known at the and 200 of the essential oils are commercially important especially in pharmaceutical, agronomic, sanitary, food, cosmetics and perfume industries (Yusof, 2010).

Cymbopogon spp is an herb that grows in most tropical countries such as Malaysia, India, Afrika and Sri Lanka (Yusof, 2010). Lemongrass can tolerate a wide range of soils and climatic conditions. However, optimum growth can be obtained when the plant is harvested on well-drained sandy loam soil with high fertility and exposed to ample of sunlight (Sugumaran *et al*, 2005). The genus Cymbopogon is the grass species that responsible in yielding citronella essential oil. There are about 140 Cymbopogon species discovered and about 20 species of Cymbopogon breed in India namely Cymbopogon *wintwerianus* Jowitt and Cymbopogon *nardus* Rendle commercially known as Java citronella and Ceylon citronella (Shasany *et al*, 2000).

Citronella oil is a clear liquid that possesses strong lemon-like odour and pale yellow colour. The total amount of essential oils present in lemongrass varies between 0.28% and 1.4%. Maximum recovered recorded is at 3% (Amenaghawon *et al*, 2014).

Citronella oil can be fractionated into three major constituent chemicals which are citronellal, citronelol and geraniol. The three major constituent chemicals are used commercially in the industry for many purposes such as soaps, sprays, disinfectants and polishes (Shasany *et al*, 2000). The major constituent of the essential oil is citral which is more than 75% by weight of the essential oil (Yusof, 2010). Citral is the natural combination of two isomeric aldehydes namely isomers geranial (α-citral) and neral (β-citral). Other unusual active components are limonene, geraniol, citronella and β-myrcene (Tajidin *et al*, 2012). The essential oil quality is judged by its citral content and the oil solubility in alcohol. The citronella essential oil can be in reddish-yellow to reddish brown colour with strong lemon odour properties. It is used in perfume, cosmetics and soap. Citronella essential oil serves as important raw material to pharmaceutical preparation such as pain balm, disinfectants and mosquito-repellent creams (Yusof N, 2010).

1.1.2 Mathematical Modelling

Simulation of a mathematical model is used to predict how the behaviour of the designed process. After simulation model is developed, the input parameter such as pressure, temperature and flow rates can be inserted by assumption to predict others relating parameters such as amount of end product. Simulation model can predict as well the process in a reversible way such as amount of raw material required and energy used to achieve the specific amount of product. The performance of the design can be changed in the next stage to improve the simulation model performance known as optimization (Babu, 2004).

In many industrial applications such as control theory, economic modelling, climate modelling and fuzzy system, mathematical model is major unit operation that are frequently used to design effective control system, handle fault diagnostics and train operating personnel (Amenaghawon *et al*, 2014). Mathematical modelling of steam distillation is considered an inevitable step to project industrial plants that have good operational conditions. The advantage of using mathematical models is the process can be simulated without running the experimental procedures in order to know the extraction process behaviour. Therefore the mathematical model allows alternative strategies to be tested in order to evaluate the selection of the process variable conditions (Cassel *et al*, 2008).

Another advantage of having a well-accepted mathematical model is its usefulness in the development of up-scaling procedures from laboratory to pilot and then to industrial scale (Kabuba J., 2013). By modelling the kinetics of distillation or any other process, it can contribute not only toward the fundamental understanding of the process but can also contribute towards better control and higher efficiency (Pornpunyaat *et al*, 2011).

Mathematical modelling is flexible as it can be varied from simple mathematical model to complex mathematical model. Many researchers construct their own mathematical model by using various physical laws with the aid of mathematical software such as Matlab, ANOVA and Designer Software 6.0.6. Benyoussef *et al* (2002) modelled the steam distillation of essential oil from coriander by using two diffusion models that take into account both diffusion and transfer of species (Benyoussef *et al*, 2002) whereas Cassel and Vargas modelled the steam distillation of lemon grass using a model based on Fick's law steady state for one-dimensional rectangular geometry (Cassel and Vargas, 2006). On the other hand, Ha *et al* modelled the extraction of essential oil from lemon grass stems by using two-factor interaction model and a linear mode in terms of coded variables for the extraction of essential oil from lemon grass stem (Ha *et al*, 2010)

1.1.3 Optimization of Mathematical Modelling

Throughout the years, chemical industry has evolved and undergone significant changes due to the increased cost of energy and increasingly stringent environmental regulations. Thus various plants design procedure and plant operating conditions have been modified in order to reduce the cost and meet the constraints. Most of the industries are currently focusing in improving the efficiency and the profitability of existing plants rather than expansion (Babu, 2004).

The ultimate goal in applying optimization is to find the optimum values in the variables of a process that can produce the best value of the performance criterion. Nevertheless there is no single method or algorithm of optimization that can be applied efficiently to all the problems. The method chosen for any case will depends on (i) nature of the constrain, (ii) the character of the objective function and whether it is known explicitly, and (iii) the number of independent and dependent variables (Babu, 2004).

Despite the advance of technology that successfully applies the extraction of essential oil, there is still a need to consider the method or procedure that can yield essential oil at its upmost optimum output in order to achieve targeted production rate, energy consumption, process minimization and others (Hazwan *et al*, 2012).

1.2 Problem Statement

Lemongrass oil, citronella oil, palmarosa oil, gingergrass oil and karnkusa oil derived from C.*flexuosus*, C.*winterianus*, C.*martinii* and C.*jwarancusa* are the five most widely traded essential oil in the aroma sector. As these oils content more than 50 terpenoidal blends and constituents, the oils are high in demand in aroma industry either as perfumery agent or as source of lead molecules to derived more useful value-added product that are required in high-grade cosmetics and drugs. Among the five commercially cultivated species of Cymbopogon mention previously, C.*flexuosus*, C.*winterianus*, C.*martinii* are the most widely used as primary source of geraniol, citronellol, citral, cironellal, linalool, 1,8-cineole, limonene, beta-caryophyllene, geranyl acetate and geranyl formate in the perfumery industry (Akhila, 2010).

Citronella plantation can only last on average of six years depending on the climate and soil conditions. The yield of the oil is low during the first year and increase gradually until it reaches the maximum at three to four years. The yield started to decline after the fourth year. Thus for economy purposes, farmers and citronella oil producers only maintained the plantation for six years as the soil becomes unproductive after six years. Up to 50 ton per hectare of raw material of citronella oil can be possibly produced annually but 25 to 30 ton per hectare is commonly achieved annually which only produces approximately 80kg of oil. The content of citronella oil in a freshly harvested raw material is only 0.5% to 1.0% and decreases as the plant dries out while waiting to be processed (Akhila, 2010).

Other than the limited life of the plantation and low yield, citronella oil also had been witnessing demand and price fluctuations as the result of proliferation of inexpensive synthetic isolates in the market. Eucalyptus *citriodora* has become a major competitor of citronella oil due to its high content of citronella oil. Synthetic turpentine oil and E.*citriodora* oil had affected the price and demand of citronella oil derived from Cymbopogon *winterianus*. However, natural citronella oil and its derivatives still remains the preferred choice in the perfumery industries as it stable properties that are vital in blending perfumes and compounding industrially important essences (Akhila, 2010).

Thus it is important for citronella oil manufacturers to have a mathematical model in order for the process to be simulated without the need to run experimental procedures to understand the extraction process behaviour thus allowing alternative strategies to be tested to evaluate the selection of the process variable conditions (Cassel *et al*, 2008). In order to predict, mathematical modelling plays important factors in predicting the extraction of citronella oil. Inefficient mathematical model may cause failure and inexact prediction of citronella yield thus leading to waste of capital cost expenditure. Therefore, it is important to study and develop optimum mathematical model for extraction of citronella oil from Cymbopogon *winterianus*. Thus, with the aim of optimize the prediction of citronella oil extraction, five mathematical models will be study in this research and further optimized. Statistical tools are used in analysing the adequacy of the fit of the models to the experimental data. The main objective of this study is to improve the mathematical modelling of Cymbopogon *winterianus* essential oil extraction by steam distillation proposed by Cassel and Vargas (2006) by minimum 5% error reduction.

1.3 Objectives

The main objective of this study is to improve the mathematical modelling of Cymbopogon *winterianus* essential oil extraction by steam distillation proposed by Cassel and Vargas (2006) by minimum 5% error reduction. By using diffusional model and Fick's Law in steady-state for one-dimensional rectangle geometry, the optimum D parameter proposed in the research is used to simulate the prediction of Cymbopogon *winterianus* essential oil yield.

1.3.1 Project Objective

The objective of this study is to simulate the mathematical model of Cymbopogon Winterianus essential oil extraction by steam distillation developed by Cassel and Vargas (2006) through their previous research "*Experiments and Modelling of the Cymbopogon winterianus Essential Oil Extraction by Steam Distillation*". The proposed optimum D parameter value in the previous research is used to simulate and verify the prediction value of the mathematical model. The study has been divided into several stages and the objective as follows:

1. To simulate the mathematical model prediction of the essential oil yield extracted using steam distillation.
2. To optimize the value of parameter D in predicting the essential oil yield extracted using steam distillation.
3. Improve minimum of 5% of the prediction essential oil yield extracted using steam distillation by proposing a new parameter D value and new mathematical model.

1.3.2 Project Scope

There are various methods of essential oil extraction. In this research, the study of the optimization of Cymbopogon *winterianus* essential oil synthesis will cover the following specific subject matter:

a) Only extraction of Cymbpogon *winterianus* essential oil by steam distillation will be cover in this study.
b) Laboratory scale experiment data in previous research done by Cassel and Vargas (2006) in their research "*Experiments and Modelling of the Cymbopogon winterianus Essential Oil Extraction by Steam Distillation*" were used as a benchmark and reference.
c) Mathematical model developed by Cassel and Vargas (2006) and the proposed optimum parameter D value is simulated by using Microsoft Excel.
d) Optimization of the mathematical model is done by using Microsoft Excel Solver.
e) New mathematical model proposed is evaluated by using three statistical criteria that are correlation coefficient (r), the root mean square error (RMSE) and the mean relative deviation modulus (E) to analyze the adequacy of the fit of the models to the experimental data.