OPTIMISATION OF OPERATING PARAMETERS FOR THE REMOVAL OF ETHANOL FROM ZINGIBER OFFICINALE ROSCOE (GINGER) OLEORESIN USING SHORT-PATH DISTILLATION

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Dedicated to my beloved husband, Ahmad Ridzuan bin Mohamed Said,
my darling daughter, Alya Syahmina, parents and family

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ABSTRACT

Due to the importance of exploiting the natural sources to enhance healthier lifestyle, cultivated herbs such as *Zingiber Officinale* Roscoe or ginger has become our main interest in this research in order to accomplish all the objectives mentioned. Using a statistical software called MINITAB, optimisation on short-path distillation experiments was done and the results showed that feed temperature, along with evaporating temperature and distillate rate were the three operational parameters that influenced the purification process of ginger oleoresin. Feed temperature should be set at 65°C and the evaporating temperature also at the same temperature to maximise the oleoresin concentration. The result of the study also point to the advantages and efficiency of feed pre-heating to temperature close or equals to that of evaporating temperature under steady-state conditions for the given operating conditions, prior to its entering the evaporation surface. Although feed rate effect is not statistically significant, it does have linear effect on the oleoresin concentration. The vast difference between theoretical separation efficiency and calculated separation efficiency was probably due to the thickness of liquid film, initial concentration and viscosity of the liquid feed. The theoretical value of recovery yield at feed and evaporating temperature of 65°C is 0.99, and experimental value of recovery yield is 0.95. Based on this integrated knowledge on various type of ginger samples, pre-processing methods and more importantly the experience of applying technologies in optimising final purification of ginger oleoresin, would serve to enhance the transfer of advanced technology to local producers.

ABSTRAK

Kepentingan ‘mengeksplotasikan’ sumber semulajadi ke arah kehidupan yang lebih sihat, penanaman herba seperti *Zingiber Officinale* Roscoe atau halia telah menjadi fokus utama dalam penyelidikan ini. Dengan menggunakan perisian statistik MINITAB, pengoptimuman penyulingan laluan pendek telah dijalankan dan keputusan eksperimen telah menunujukkan bahawa suhu suapan, bersama dengan suhu penyejatan dan kadar pendistilan adalah tiga parameter yang penting dalam proses penulenan oleoresin halia. Suhu suapan harus ditetapkan pada 65°C dan begitu juga dengan suhu penyejatan untuk memaksimakan kepekatan oleoresin di akhir proses penulenan. Keputusan eksperimen juga menunjukkan kelebihan dan kepetingan pra-pemanasan terhadap suhu suiapan sama atau hampir sama dengan suhu penyejatan di bawah keadaan stabil, sebelum cecair masuk ke dalam permukaan penyejatan. Walaupun kadar suapan tidak signifikan secara statistik, ia sebenarnya mempunyai kesan secara linear terhadap kepekatan oleoresin. Perbezaan yang ketara antara kecekapan pemisahan secara teori dengan kecekapan pemisahan yang diperolehi dari eksperimen mungkin disebabkan oleh ketebalan cecair filem, kepekatan asal and kelikatan cecair suapan. Nilai teori hasilan pada suhu suapan 65°C adalah 0.99, dan nilai hasilan secara eksperimen ialah 0.95. Berdasarkan pelbagai pengetahuan yang dapat diteroka melalui penyelidikan berkenaan berjenis-jenis sampel halia, kaedah pra-pemprosesan dan yang pentingnya pengoptimuman penyulingan turus pendek ini, akan dapat meningkatkan pengetahuan serta pemindahan teknologi secara lebih efisien kepada penanam serta pengeluar halia tempatan.

CONTENT

LIST OF TABLES

LIST OF FIGURES

NOMENCLATURE

α	separation efficiency
[A]	reactant concentration
C, C_w, C_{d1}, C_{d2}	concentration of species in liquid
C_∞	equilibrium concentration
D	Diffusion coefficient
K, K_{d1}, k_{d2}	rate constant
M, M_A, M_B	Molecular weight
η	mass transfer efficiency
$p^o, p^o{}_A, p^o{}_B$	saturation vapour pressure, component
R	Universal gas constant
r'	rate of reaction
r	radius of gland/oil globule
T	Temperature
t	time
w_o, w_{oA}, w_{oB}	rate of molecular evaporation
x_A, x_B	mole fraction of component A at evaporating surface
y_A, y_B	mole fraction of component A at condensing surface
ρ	density
σ	cross-section
λ	mean-free path
Ω	volume interaction
N	number of target particles in Ω
A	Atomic weight
N_A	Avogadro number

E_i	Evaporation rate
C_{is}	Mole fraction of the liquid in the film surface
F	surface ratio
n	number of components in the mixture
k	anisotropy of vapour
G	residual output
X	molar fraction in the residual flow
Xo	molar fraction in the feed
u	separation ratio
S	film thickness
η	viscosity
m_o	mass flow rate

GLOSSARY

alcohol	any of a class of organic compounds with the general formula ROH, where R represents an alkyl group made up of carbon and hydrogen in various proportions and OH represents one or more **hydroxyl groups**.
aliphatic	any of a large class of organic compounds whose carbon atoms are joined together in straight or branched open chains rather than in rings.
amber	a heavy, full bodied, powdery, warm fragrance note.
anti-inflammatory	medicines that relieve pain, swelling, stiffness, and inflammation.
aspirin	a kind of drug used to relieve pain and fever,by blocking the production of hormone-like substances known as prostoglandins
ayurvedic	the ancient Hindu art of medicine and of prolonging life
bleaching	uniform coating of lime after ginger is being dried and limed for several days.

blanching	blanching is a heat treatment process applied to fruit and vegetables prior to freezing, drying or canning like hot steaming, hot water boiling.
chromatography	resolution of a chemical mixture into its component compounds by passing it through a system that retards each compound to a varying degree.
comminution	process of grinding.
concrete	The mixture of volatile oil, waxes and colour that is obtained after an atomatic raw material such as flower petals are extracted with a highly volatile solvent.
condenser	a apparatus of which a change of a substance from the gaseous (vapor) to the liquid state can be observed
cortex	in botany, term generally applied to the outer soft tissues of the leaves, stems, and roots of plants.
decorticated	peeling of outer layer of a plant.
desirability function	A desirability function translates each response scale to a zero-to-one desirability scale in MINITAB software. The most desirable values of the response have desirability one. The least desirable values have desirability zero.
diffusion	the spontaneous migration of substances from regions where their concentration is high to regions where their concentration is low

distillation	process of heating a liquid until its more volatile constituents pass into the vapor phase, and then cooling the vapor to recover such constituents in liquid form by condensation.
distillate rate	volume divide by seconds of the more volatile liquid condensed
eddy diffusion	turbulent diffusion, characterized by motion of the fluid particle, which is irregular with respect to direction and time.
extraction	The method by which essential oils are separated from the plant using solvents which can then be removed by evaporation.
equilibrium	reversible chemical reactions in which the reactions involved are occurring in opposite directions at equal rates, so that no net change is observed.
evaporating temperature	the arithmetic mean of evaporator inlet and dew point
evaporation	change of a liquid into vapor at any temperature below its **boiling point**
halogen	any of the chemically active elements found in group VIIa of the **periodic table.**
hydrocarbon	any organic compound composed solely of the elements hydrogen and carbon.
homogeneous	process in which a mixture is made uniform throughout. Generally this procedure involves reducing the size of the particles of one component of the

	mixture and dispersing them evenly throughout the other component.
hydro-distillation	the type of distillation in which the botanic material is completely immersed in water and the still is brought to the boil. This method protects the oils so extracted to a certain degree since the surrounding water acts as a barrier to prevent it from overheating.
laminar	When fluid flows smoothly without vortices or other turbulence, typically when a fluid is flowing this way it flows in straight lines at a constant velocity.
mean-free path	The mean free path of a particle in a medium is a measure of its probability of undergoing interactions of a given kind.
molar fraction	The ratio of the amount of substance (number of moles) of substance A to the total amount of substance in a mixture.
molecular distillation	by a short exposure of the distilled liquid to elevated temperatures, high vacuum with below than 0.001 mbar, in the distillation space and a small distance between the evaporator and the condenser
morphology	scientific study of the form and structure of animals and plants.
nonpolar solvents	of which carbon tetrachloride is an example, have molecules whose electric charges are equally distributed.

parenchyma	These are cells in a tissue or tissues in an organ that are concerned with function.
pharmacology	scientific study of drugs and their use in medicine. body, and of the mathematical relationships required to develop models to interpret such data.
polar solvent	of which water is an example, have molecules whose electric charges are unequally distributed, leaving one end of each molecule more positive than the other.
purification process	the process of removing impurities (as from oil or metals or solvent etc.)
refractive index	The behaviour of light entering a crystal that is fundamentally controlled by the crystal structure.
residence time	The average amount of time that an element or substance remains in the column at steady state conditions
recovery yield	The quantity of product obtained from a process or reaction
residual rate	The rate of the lees volatile component exiting the evaporator column
rhizomes	root-like stems with nodes which grow under or along the ground
separation efficiency	a measure of the short-path evaporator's performance during purification process

short-path distillation	a short exposure of the distilled liquid to elevated temperatures, high vacuum performed at pressures between 0.5 and 0.01 mbar in the distillation space and a small distance between the evaporator and the condenser,
steroids	any of a number of organic compounds naturally produced in the body, including certain hormones and vitamins.
steam distillation	This process involves the use of steam to percolate and vapourise out the essential oils from the plant material, with the subsequent condensation of steam and essential oil prior to their separation.
solvent	constituent of a solution that acts as a dissolving agent.
super critical fluid extraction	supercritically performed extraction method under relatively severe pressure and temperature conditions to remove all oleoresin components quickly and efficiently
turbulent	The irregular, chaotic flow of a fluid that results in random velocity fluctuations and in mixing.

LIST OF APPENDIXES

CHAPTER 1

INTRODUCTION

1.1 Research background

Since the time man started gathering and adding varieties in food, spices seem to have been used to make the food more appetizing: they add aroma and taste to the cereal or meat dishes. In the modern world today, spices and those aromatic herbs continue to be extensively used as a ‘versatile’ spice throughout India, the Middle East and the Far East in food preparation and also in alternative medicine. Ancient belief in the medicinal properties of ginger existed in Indian and oriental cultures where ginger was used alone or as a component in herbal remedies. In recent times there has been scientific research undertaken to test out the validity of the medicinal claims about ginger. There have been some exciting results from the experiments conducted by researchers worldwide with respect to the medicinal properties of ginger as new alternative for ordinary chemicals like steroids and aspirin (Katiyar, *et.al*,1996).

The rhizome of ginger (*Zingiber officinale Roscoe*) is widely cultivated in the tropic and semi-tropic regions of the world. The high moisture content of the ginger and low shelf life of raw ginger makes the transportation and marketing of ginger an expensive proposition (Bartley and Jacobs, 2000). Drying of ginger is not practiced in many ginger cultivar countries because of the adverse climatic conditions. Making of value added products which find market within the country as well as abroad is the only answer for the poor farmers of these countries (Kumar and Arumughan, 2000). Ginger oil is meant for the export market only and the demand within the country is marginal. A new entrepreneur will find it difficult to penetrate the international market but with the support of the governmental agencies can achieve some break through (Varghese, 1998). If proper promotional efforts are put in, the world demand can be totally met by these countries.

In order to obtain high quality ginger extract as an alternative chemicals, ginger as raw material needs to undergo thorough processing methods from sample preparations right to the quality assessment of its end product. Ginger extract preparations vary widely in their concentration and purity, depending on the source of the root, extraction methodology and formulation (Lawrence and Tobacco, 1984). The first step in oleoresin preparation is the extraction of dried, ground ginger with solvent, usually acetone or ethanol (Connell,1970). In 1999, Lewis, et.al, patented the method of making processed fresh herbs. They claimed that, edible salts such as sodium chloride assist in preventing the rapid discolouration and flavour change of herbs. Wu, *et.al* (2001) patented a method of preparing an extract from *Zingiber officinale*, which is potent in anti-inflammation and anti-platelet aggregation. They prepared a crude liquid from rhizomes of ginger by extraction with several steps of using organic solvents like alcohol and acetone.

In 1997, Kreuter and Steiner invented and patented a process for the preparation of a stable, homogeneous extract of plants. They used a 400 kg dried herbs sample mixed with 1600 kg ethanol and extraction was done at a temperature between 60-70°C. Solvent removal of the patented process was done under evaporation, reduced pressure and elevated temperature. Andrews and Basu (2000), patented a process of preparing a pharmaceutically-active extract of valerian root. The process comprises the steps of adding the roots to an alcoholic extraction solvent to form a mixture, wherein the alcoholic extraction solvent comprises of a mixture of 50% to 100% (v/v) in water and heating the mixture to 80°C for two hours.

In 1990, Spiro, *et.al*. (2000), extracted the ginger oleoresins with supercritical fluids (SCF) which proceeds through three stages; washing, fast extraction and slow extraction. They assumed that the extraction is completed at the end of the fast extraction stage, which required six hours. However, oleoresins extracted with supercritical fluids have a higher price because of higher quality and less variations in the final products despite having extreme operating pressure (up to 127 bar) compared to those extracted with organic solvents. In 1997, Balladin used pilot plant extraction to extract oleoresin from ginger. The solvent used followed its designated route to the vertically oriented water cooled 20 litres leaching vessel. The process continued for 10 hours until the majority of the oleoresin was extracted, determined by the transparent appearance of the extracting solvent. In this research, short path distillation is the main focus in final purification of ginger oleoresin extraction. Short-path distillation or molecular distillation is generally acknowledged to be the safest method to separate and purify thermally unstable compounds and substances having low volatility (Lutisan and Cvengros, 1995) The number of applications for short path distillation is relatively high Baker and Olejniczak (1997). For instance, short path distillation has been used in solvent removal and reducing the pigmentation process of paprika oleoresin for years (Albers, 2001). By using short path distillation, the solvent traces can be removed up to 10 ppm. In 1996, a company named after a Norwegian pharmacist Peter Moller, developed a new process using short path distillation for extracting the oil from cod liver, which can increase the omega 3 acid up to 60% (Kukla, 1997). In flavour enhancement process, Mehnert (1997) patented methods and composition to fractionate the milk fats and fatty acids into mixture of chain lengths and bond saturations under the conditions of 190°C, at 0.003 mm Hg by using short path distillation.

Modelling the process of molecular distillation has been considered by several authors, focused mostly on organic and pure compounds, either binary or ternary mixture. However, studies on multi-component or complex organic mixtures such as the ginger oleoresin have never been documented. Bose and Palmer (1984) have given reasons for a separation efficiency decrease by distillation in jet tenzimeter using binary mixtures. This decrease is caused by temperature gradients in the distilled liquid mixture as a consequence of intense evaporation. Cvengros, et.al., (2001), developed a model for modelling various design and operating conditions in a molecular evaporator, such as the fractionation in the evaporator with a divided condenser, the influence of feed temperature on the evaporator performance, the effect of an adiabatic separator in the distillation gap on the separation efficiency and the effect of inert gas pressure on the molecular distillation process, also using binary mixtures.

Three operational parameters which were studied in this research, namely feed temperature, evaporating temperature and feed rates, were optimised by a statistical software called MINITAB®. Comparative analysis between the optimised operational settings and theoretical model were then described by empirical and theoretical separation efficiency and recovery yield in chapter 3.

1.2 Objectives and scopes

The objective of this research was to identify the most influential operational parameters or combined parameters that affect the yield of ginger oleoresin collected in short-path distillation by using a statistical software, MINITAB®, as a mean of final stage of ethanol removal.

Three important scopes have been identified in achieving the above mentioned objective:

1. Using pilot scale extractor in ethanol extraction and partial ethanol removal of ginger oleoresin. For the final stage of ethanol removal, short-path distillation process was used.

2. Varying the feed temperature, feed rate and evaporating temperature on the short-path evaporator performance, pairing the all the parameters involved to be optimised by MINITAB® with dried, mature, blanched and bleached ginger as the raw material.

3. To relate and analyse the findings of each combined operational parameters to the theory and empirical equations of separation efficiency and recovery yield.

1.3 Statement of problem

The affluent lifestyle has led to the increasing demand for health food. Scientific evidence support claims that prevention is better than cure (Morad, *et.al*, 2000). Numerous scientific findings are in support of healthier lifestyle through consuming natural ingredients to prevent and cure modern diseases. In order to accommodate the fast pace in modern life whilst maintaining the nutritious values in food, the health food market ranging from personal care products and health food is becoming the present norm of life (Schulick,1993). The Malaysian herbal market was estimated to be worth RM2.5 billion annually, with the local herbal industry capturing only 5% to 10% of the market. Statistics in 1994 shows in terms of sales of herbal medicines by region, Europe topped the chart with sales of US$6 billion, Asia US$2.3 billion, Japan US$2.1 billion and North America US1.5 billion. It is projected that the value of the global market for herbal products would reach US$200 billion by 2008 (The Sun, 2001).

Due to the importance of exploiting the natural sources to enhance healthier lifestyle, cultivated herbs such as the *Zingiberaceae* family has become our main interest in this research in order to accomplish all the objectives mentioned. Most Zingiberaceae family species are fibrous rooted perennial which are cultivated in many tropical and subtropical area. There are about 18 genera with more than 160 species of *Zingiberaceae* in Peninsular Malaysia (Schulick, 1993). Among these species, *Zingiber Officinale* (also known as ginger) is widely used in food industry for flavouring purposes as well as foods and beverages and as fragrances in pharmaceutical and industrial products. Apart from the identification of constituents in the common species of ginger by some of our local chemists, there is no interest as yet to commercialise the findings (Morad, *et.al*, 2001).

In this research, feed temperature, evaporating temperature and feed rates were the three operational parameters experimented to optimise the operating conditions for the ethanol removal of ginger oleoresin. According to Cvengros *et.al*., in 1999, practical experience over many years with the operation of molecular evaporators have shown that the optimum temperature of the entering liquid feed should not differ too much from the working temperature inside the evaporator. This results in evaporation at a non-reduced rate from the very first moment the distilled liquid gets onto the evaporation surface, without using the evaporation surface to post-heat liquid to the operation temperature. In 1996, Batistella and Maciel studied the increase in the liquid film temperature caused by heating of the evaporator governed by the evaporating temperature, the feed rate and the initial pre-heating of feed. If the supply enters the machine at a lower temperature than the distillation temperature, a part of the surface is used to heat the solution to the later one, causing a temperature gradient. This temperature change also makes it very difficult to maintain a constant liquid flow unless an optimum feed rate is used. Nguyen and Goffic, in 1997 suggested that these factors: feed temperature, feed rate and evaporating temperature can only be estimated through experimental work.

By completing the optimisation of operational parameters using short- path distillation, it is anticipated that once optimised operational parameters of short path distillation have been established, ginger oleoresin mixtures can be extracted efficiently. In this way, Malaysia can develop our own standardised ginger products based on different combination of ginger.

1.4 Research Contribution

Based on the present knowledge on various type of ginger sample, the pre-processing methods and more importantly the experience of applying technologies in optimising final purification of ginger oleoresin, would serve to enhance the creativity development and transfer of advanced technology to local producers. It is anticipated that once these methods have been established, thermally instable constituents such as 6-gingerol can be extracted. Apart from that, it is believed that with the help from this recent technology, the local producers can actually formulate a better ingredients of natural based healing ailments since the raw materials are actually originated and widely cultivated in this part of the region. It would also be a great challenge for all of the cultivars of natural herbs in the tropical countries to be one of the great competitors generating high quality natural based product such as Zinaxin®(ginger extract which promotes joint health).

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