SUPERCRITICAL CARBON DIOXIDE EXTRACTION OF CASTOR OIL SEED

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To Allah (SWT), my beloved mother and father

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ABSTRACT

This study investigates the extraction of castor oil using supercritical carbon dioxide (SC-CO_2). The response surface methodology (RSM) was employed to show explicitly the influence of the process parameters such as temperature, pressure and CO_2 flow rate on the oil yield using the Box-Behnken design. The linear terms of pressure, CO_2 flow rate and temperature, and the quadratic terms of temperature, pressure and CO_2 flow rate had a significant effect on the oil yield. The maximum oil yield obtained from the mathematical model was predicted to be 9.29% under the conditions of temperature at 63.7 °C and pressure of 29.9 MPa with CO_2 flow rate of 4.15 mL/min. Comparing the oil yield with the conventional soxhlet extraction, yields of 59.8%, 52.3% and 49.9% were obtained using ethanol, n-hexane and petroleum ether as solvents for optimized average particle sizes and extraction times of 1.30 mm and 2.88 h, 1.26 mm and 2.65 h and 1.25 mm and 2.55 h, respectively. In addition, the solubility of castor oil was measured at temperatures ranging from 313 to 335 K, and pressures 20 to 36 MPa, respectively. The measured solubilities ranged from 1.00×10^{-3} to 4.88×10^{-3} g of oil/g of CO_2. The measurements confirmed that temperature and pressure have direct effects on solubility-enhancement factors. Five semi-empirical models were tested for correlating the experimental data from SC-CO_2 extraction: the Chrastil, Del Valle Aguilera (VA), Bartle, Kumar and Johnston (KJ) and Mendez-Santiago and Teja (MST) models. The solubilities from these models had the following average absolute relative deviations (AARD%) from experimental data: 0.05% (Chrastil), 0.30% (VA), 0.38% (Bartle), 5.98 % (KJ) and 28.5% (MST). Thus, the Chrastil, VA and Bartle models correlated to the castor oil solubility data with the lowest AARD%. The physico-chemical properties of the seed oil extracted using the SC-CO_2 and soxhlet methods were determined. Palmitic, stearic, oleic, linoleic, linolenic and ricinoleic acids were identified by gas chromatography-mass spectrometry (GC-MS) analysis after the formation of fatty acid methyl ester (FAME). For oxidation stability, castor oils derived from these extraction methods were heated in an oven at 70 °C for 0–12 weeks. The quality and oxidative stability of oil recovered by SC-CO_2 were generally found to be better than that recovered by the soxhlet method. In addition, the parameters considered in this study, namely peroxide value, refractive index, pH, conductivity, acid value and free fatty acid offer an appropriate tool for evaluating the quality of castor oil during storage as well as its oxidative stability; the study also sheds light on the different responses of castor oil during storage.

ABSTRAK

Kajian ini meneliti pengekstrakan minyak kastor menggunakan karbon dioksida lampau genting (SC-CO_2). Kaedah gerak balas permukaan menggunakan reka bentuk *Box-Behnken* digunakan untuk menunjukkan dengan jelas pengaruh parameter proses seperti suhu, tekanan dan kadar alir CO_2 ke atas hasil minyak. Pekali linear dan kuadratik bagi suhu, tekanan dan kadar alir CO_2 mempunyai kesan ketara ke atas hasil minyak. Hasil minyak maksimum yang diperoleh daripada model matematik dijangkakan sebanyak 9.29% pada keadaan suhu 63.7 °C dan tekanan 29.9 MPa dengan kadar alir CO_2 4.15 mL/min. Membandingkan hasil minyak dengan kaedah pengekstrakan *soxhlet* menggunakan etanol, n-heksana dan petroleum eter sebagai pelarut, hasil sebanyak 59.8%, 52.3% dan 49.9% diperoleh untuk saiz zarah dan masa pengekstrakan masing-masing 1.30 mm dan 2.88 jam, 1.26 mm dan 2.65 jam, dan 1.25 mm dan 2.55 jam. Selain itu, kebolehlarutan minyak diukur pada suhu di antara 313 hingga 335 K, dan tekanan 20 hingga 36 MPa. Kebolehlarutan adalah di antara 1.00×10^{-3} hingga 4.88×10^{-3} g minyak/g CO_2. Pengukuran mengesahkan bahawa suhu dan tekanan mempunyai kesan langsung ke atas faktor peningkatan kebolehlarutan. Lima model separa empirik telah diuji untuk menghubungkaitkan data ujikaji daripada pengekstrakan SC-CO_2: model *Chrastil, Del Valle Aguilera (VA), Bartle, Kumar dan Johnston (KJ),* dan *Mendez-Santiago dan Teja (MST)*. Kebolehlarutan diperoleh daripada model mempunyai sisihan relatif mutlak purata (AARD%) berikut: 0.05% *(Chrastil)*, 0.30% *(VA)*, 0.38% *(Bartle)*, 5.98% *(KJ)* dan 28.5% *(MST)*. Oleh itu, model *Chrastil*, *VA* dan *Bartle* dapat dikaitkan dengan data kebolehlarutan minyak kastor dengan AARD% terendah. Sifat fiziko-kimia minyak diekstrak menggunakan SC-CO_2 dan kaedah *soxhlet* ditentukan. Asid palmitik, stearik, oleik, linoleik, linolenik dan risinoleik dikenalpasti melalui analisis kromatografi gas-spektrometri jisim (GC-MS) selepas pembentukan asid lemak metil ester (FAME). Untuk kestabilan pengoksidaan, minyak kastor diperoleh daripada kaedah-kaedah pengekstrakan telah dipanaskan di dalam ketuhar pada 70 °C selama 0-12 minggu. Kualiti dan kestabilan pengoksidaan minyak yang diperoleh dengan SC-CO_2 secara umumnya didapati lebih baik berbanding kaedah *soxhlet*. Di samping itu, parameter yang dipertimbangkan dalam kajian ini iaitu nilai peroksida, indeks biasan, pH, kekonduksian, nilai asid dan asid lemak bebas bersesuaian untuk menilai kualiti minyak semasa penyimpanan serta kestabilan pengoksidaan; kajian ini memberikan gambaran perubahan sifat minyak kastor semasa penyimpanan.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

a	-	Solvato complex
AARD	-	Absolute relative deviation percentage
ANOVA	-	Analysis of variance
AOM	-	Active oxygen
AP	-	Adequate precision
ASTM	-	American Society for Testing and Materials
atm	-	Atmosphere
AV	-	Acid value
b	-	Indicates solute extraction
BHT	-	Butylated hydroxyl toluene
BHA	-	Butylated hydroxyl anisole
BS	-	British standard
c	-	Constant
c	-	Speed of light in a vacuum
^oC	-	Degree Celsius
C_1	-	Weight of castor seed before drying
C_2	-	Weight of castor seed after drying
CCD	-	Central composite design
CE	-	Capillary electrophoresis
CO_2	-	Carbon dioxide
CP	-	Critical point
cst	-	Centi stroke
CV	-	Coefficient of variance
DIN	-	Deutsche standard
DoE	-	Design of experiment
DSC	-	Differencial scanning colorimetry
Ea	-	Activation number
EFSA	-	European food safety authority
EoS	-	Equation of state
EtOH	-	Ethanol

FAMEs	-	Fatty acid methyl esters
FESEM	-	Field emission scanning electron microscopy
FFA	-	Free fatty acid
FTIR	-	Fourier transform infrared spectroscopy
g	-	grams
GC	-	Gas chromatography
GC-MS	-	Gas chromatography–mass spectrometry
GC	-	Gas chromatography
h	-	Hour
H_2O	-	Water
ΔH	-	Heat of reaction
ΔH_{solv}	-	Heat of salvation
ΔH_{vap}	-	Heat of vaporization
HPLC	-	High- pressure liquid chromatography
I_2	-	Iodine value
IP	-	Industrial practice
K	-	Kelvin
k	-	Association number
KJ	-	Kumar and Johnson
KOH	-	Potassium hydroxide
m	-	Mass
Meq	-	Milliequivalents
mg	-	Milligram
mm	-	Milli meter
MPa	-	Mega Pascal
MST	-	Mendez- Santiago and Teja
n	-	Number of data points
N	-	Normality
NaOH	-	Sodium hydroxide
OSI	-	Oil stability instrument
P	-	Pressure
p	-	Probability
PG	-	Propyl gallate
P_C	-	Critical pressure
pH	-	Hydrogen ion concentration
P_0	-	Independent real value

P_{ref}	-	Standard reference pressure
Pr	-	Reduced pressure
P_{sub}	-	Sublimation pressure
PV	-	Peroxide value
R	-	Gas constant
RI	-	Refractive index
RSM	-	Response surface methodology
S	-	Solubility
S_{calc}	-	Solubility calculated using model
S_{exp}	-	Solubility obtained from experimental data
S_I	-	Volume of solution
S_0	-	Volume of blank
SC-CO_2	-	Supercritical carbon dioxide
SCF	-	Supercritical fluid
SFE	-	Supercritical fluid extraction
T	-	Temperature
T_i	-	Actual independent variable
ΔT_i	-	Step change
TAN	-	Total acid number
TBHQ	-	Mono-tert-buyl-hydroquinone
T_C	-	Critical temperature
TGA	-	Thermal gravimetric analysis
TP	-	Triple point
Tr	-	Reduced temperature
v	-	Speed of light in a substance
v^1	-	Kinematic viscosity
VA	-	Del Valle Aguilera
VI	-	Viscosity index
X_i	-	Independent variable
y_1	-	First y–value above
y_2	-	Second y–value below
ZDTC	-	Zinc dithiocarbomates
ZDTP	-	Zinc dithiophosphates
ρ	-	Density
ρ_{ref}	-	Reference density
%	-	Percentage

µL	-	Microlitre
µ	-	Dynamic viscosity
µm	-	Microns

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Castor oil plant (*Ricinus communis L*), a drought-resistant shrub belonging to the family *Euphorbiaceae* is extracted from castor seed. The extraction involves both mechanical and chemical separation processes (Anderson, 2005; Belgacem and Gandini, 2008; Kemper, 2005). Castor oil is viscous, pale yellow and non-drying oil with a bland taste and in most times generally used as a purgative (Mutlu and Meier, 2010). The mechanical separation process is often associated with low yields as against a chemical separation process which has (>99 weight %) of oil yield (Guoliang et al., 2011; Zhao and Zhang, 2013). Chemical extraction methods in most cases employ solvents such as *n*–hexane. However, its volatility is high and toxic in nature at low concentration. The vapors need to be monitored as uncontrolled amount could lead to an explosion during industrial oil extraction. Moreover, conventional extraction methods are time consuming, laborious, no selectivity and low extraction yields, no fractionation capabilities, solvent residue remained in the product, manipulations of limited variables, damaged the heat-sensitive components of the materials and the requirement of post-extraction process for solvent removal (Danh et al., 2009; Hossein et al., 2014; Luque de Castro and Priego, 2010; Miao et al., 2013; Rezzoug et al., 2005).

On the other hand, supercritical fluid extraction (SFE) allows the extraction of oil from seeds that are as good as, or even better than, those of conventional extraction. Thermal degradation and decomposition of labile compounds are avoided, due to the operation at reduced temperatures, whereas the absence of light and oxygen prevents the occurrence oxidation reaction (Turek and Stintzing, 2013). More also, the supercritical fluid processed materials do not require any sterilization, since gram positive and gram negative bacteria can be inactivated at mild temperatures. The high pressure gradient during pressure release can yield extracts free of microorganisms and their spores, with a longer shelf life than standard solvent extraction (Foster et al., 2003; Perrut, 2004).

Supercritical fluid extraction (SFE) is a technique that can overcome these drawbacks of the conventional extraction process (Danlami et al., 2014). The negligible environmental impact of this process represents a prospect for changing the relative concentration of the various lipid moieties (Perretti et al., 2003). SFE is fast becoming a powerful means of extraction of solid samples especially seeds oil. It can be considered a technological revolution in the extraction industry (Sahena et al., 2009). Supercritical carbon dioxide (SC-CO_2) is formed when liquids and gases are heated at a temperature and pressure higher than their critical points. Generally, substances that are heated above their critical temperature, regardless of the pressure, liquid phase cannot be recognized, as such when temperature and pressure are higher than their critical temperature and pressure, such are called supercritical fluid. Another description refers to substance in the state above its critical value. That critical value represents the temperature and pressure where it is not possible to clearly distinguish the boundary phases. That is the vapour-liquid critical point is in line and there are no separate phases of gas and liquid. Supercritical fluids effuse both solids like vapours and dissolve it like a liquid. In fact, the fluid property is neither that of a gas or liquid, but something in between. As a result, supercritical fluids provide the option for a better extraction feasibility, better products quality and efficiency. A supercritical fluid shows specific properties which are different from properties of gas and liquid states (Clifford, 1998).

The major advantages of using supercritical fluids for extraction purposes are (Baysal et al., 2002).

1. The extraction method is extremely rapid.
2. The solvent for extraction is easily removed.
3. The pressure, temperature and other variable conditions can be used to control the process
4. The solvent such as carbon dioxide, ethanol and methanol are reasonably cheap.
5. Supercritical fluid extraction does not produce any contamination.

Various studies were conducted in developing new extraction processes of increasing both quality and quantity of active natural products (Birmakr et al., 2012; Sajfrtova et al., 2010). Supercritical fluid extraction (SFE) has received great attention as a potential substitute for conventional technology for separating numerous valuable compounds from plant sources (Gomes et al., 2007; Liu et al., 2010). This is because the technique is carried out at low temperatures, at minimal extraction times and a little amount of solvent in comparison to other conventional solvent extraction methods that have been used for extracting valuable compounds for a very long time, which also involves using large amount of solvents with higher temperatures (Liza et al., 2010).

SC-CO_2 extraction is still receiving attention globally because carbon dioxide (CO_2) is an inert, low-cost, harmless and ecologically-friendly which allows extraction at relatively low temperatures and pressures. More also, CO_2 evaporates easily when subjected to atmospheric conditions, as a result, the extract is freed from both chemicals and thermal degradation (Gomez et al., 1996). In fact, SC-CO_2 extract are by and large recognized as generally safe to be used in food industries (Gerard and May, 2002). Furthermore, CO_2 is also referred to be a nonpolar solvent, therefore, adding a little amount of co-solvent can improve significantly the extraction of polar compounds. Among these solvents ethanol (EtOH) is mostly used due to its miscibility with CO_2, non-toxic and allowed usage in the food and pharmaceutical products (Gomez et al., 1996). Previously, SC-CO_2 has been used by many researchers for extracting valuable compounds from different natural sources such as hazelnut (Bernado-Gil et al., 2002), grape seed (Cao and Ito, 2003), watermelon (Vaughn et al., 2008), winter melon (Bimakr et al., 2013), orange pomace (Benelli et al., 2010), peach (Prunus persica) (Herrero et al., 2010), Bidens pilosa Linné (Kviecinski et al., 2011) and Mitragyna speciosa leaves (Orio, 2012). Sanchez et al. (2009), presented an overview on some new advances and applications of SFE. In addition, SFE has been used for separating antioxidant compounds from sage, brazilian plants and some rosemary leaves (Monica et al., 2011; Ollanketo et al., 2002; Veggi et al., 2011). It is remarkable that very few reports on the extraction of castor oil using SC-CO_2 have been published. Turner et al. (2004) reported on the extraction of castor oil using CO_2 over the range of 40 – 80 ^{o}C and 20 - 40 MPa. They found that the fatty acid methyl esters (FAMEs) in castor seeds were similar to those obtained using conventional methodology based on solvent extraction.

The extraction rate of seed materials with high oil is often limited by the solubility of oils in supercritical carbon dioxide (SC-CO_2) (Dauksas et al., 2002; Molero et al., 2002; Marongiu, 2004; Illes et al., 2000; Leeke et al., 2002; Menaker et al., 2004). Therefore, oil solubility data are key to the development of separation processes, including feasibility evaluations, process design, scale up and establishing optimum operating conditions (Sovova et al., 2001; Abaroudi et al., 2002; Carr et al., 2011; Iwai and Yamamoto, 2013). However, experimental determination of the solubility of oil in SC-CO_2 is a difficult and an expensive task. Therefore, models to correlate and predict solubilities of oil in SC-CO_2 are very desirable; such models usually take the form of theoretical equations of state or semi-empirical equations because experimental determinations of the solubilities of oil in SC-CO_2 at various temperatures and pressures are time consuming. Cubic and non-cubic equations of state have been used to model the solubilities of oil in SC-CO_2 with and without co-solvents (Tabernero et al., 2011; Ashour et al., 2000). However, these equations require robust computational methods and the knowledge of several properties such as sublimation pressures, molar volumes, critical temperatures and pressures. These properties are scarce in the literature, thus group contribution methods are usually applied, although their use introduces additional uncertainties in the use of these equations (Coimbra et al., 2006).

1.2 Problem Statement

The utilization of vegetable oil in engineering is declining due to oxidative, hydrolytic and thermal stability. The greatest challenge however is the oxidative stability which has become a major concern that hinders the useful service life of the plant or vegetable oil. This is due to the presence of polar compounds which could irreversibly produce insoluble deposits throughout the whole volume, thus increasing the oil acidity and viscosity, which are not good for industrial purposes. These results from the type of extraction method used for the oil production. Valuable compounds are identified and isolated by means of extraction from plant sources (Stevigny et al., 2007). Oil extraction of plant or seed material is done using solvent extraction (Akpan et al., 2006; Salimon et al., 2010; Shridhar et al., 2010). Solvent extraction removes almost all organic phases inclusive of polar and non-polar groups and it becomes very difficult to separate compounds of interest, thereby leading to subsequent processing to improve the oxidative stability of oil is unavoidable.

Until now, attention has been mainly focussed on improving the oxidation stability of plant or seed oils obtained from solvent extraction (Akpan et al., 2006; Salimon et al., 2010; Shridhar et al., 2010) and mechanical expression with antioxidants. Examples of such antioxidant are butylated hydroxyl toluene (BHT), butylated hydroxyl anisole (BHA), mono-tert-buyl-hydroquinone (TBHQ), propyl gallate (PG), some naturally occurring tocopherols, zinc dithiophosphates (ZDTP) and zinc dithiocarbomates (ZDTC) (Fox and Stachowiak, 2007). Tocopherols are widely used as natural antioxidants, although their protective ability is not always sufficient (Milovanovi et al., 2002). However, the safety of these synthetic antioxidants has been a cause of concern because of their toxicity. They also decompose and become less effective at high temperature (Fox and Stachowiak, 2007; Akoh and Min, 2008; Gunstone, 2011; Shahidi, 2005). There is therefore need for further researches into finding a more environmentally friendly approach in improving oxidation stability. This research will therefore employ supercritical fluid extraction techniques to limit the presence of polar compounds in plant based oil from the castor oil. The castor oil has better properties than other vegetable oil due to its high content of ricinoleic acid (Scarpa and Guerci, 1982; Sorin, 2012). The main idea behind using supercritical carbon dioxide as a solvent is for selective extraction of non-polar compounds, thereby leaving behind the polar compounds that can accelerate the oxidation of the oil.

To date, there is no technical report has been published on the use of supercritical carbon dioxide extraction as a green approach to remove polar groups at the early stage of oil extraction in plant based oil processing to achieve oxidation stability improvement of the resultant oil without the addition of antioxidants. Similarly, there has been no research on the development of a mathematical model and optimization of the supercritical carbon dioxide extraction of castor oil using statistical experimental design method. Such a determination is crucial to understanding the effects of process variables such as temperature, pressure and CO_2 flow rate on extraction and oxidation stability.

Oil solubility data are also important at several stages in the development of SFE processes, including feasibility evaluations, process design, equipment sizing and establishing optimum operating conditions. However, experimental determination of the solubility of oils in SC-CO_2 is a difficult and an expensive task. Therefore, models to correlate and predict solubilities of oils in SC-CO_2 are very much desirable. Gupta and Shim (2007) reported the solubility data of various compounds in SC-CO_2 from over 1200 data. The solute including lipids, solids, polymer, food, drugs, pesticides, dyes, metal complexes have been reported (Gupta and Shim, 2007). However, there are no studies reported on the solubility of the castor oil in SC-CO_2.

1.3 Objectives of the Study

The research is aimed at studying the extraction of castor oil seed using supercritical carbon dioxide.

In achieving these objectives, there are specific objectives that have to be fulfilled, which are:

1. To extract castor oil using supercritical carbon dioxide.
2. To establish the solubility profile of castor oil by supercritical carbon dioxide.
3. To characterize the physio-chemical properties of castor oil products.

1.4 Scope of the Thesis

This research covers extraction of castor seed oil using supercritical carbon dioxide and also, comparing the oil yield to conventional solvent extraction. Modelling and optimization of the effect of independent variables such as average particle size, extraction time and temperature, pressure, flow rate of CO_2 on oil yield by employing conventional and supercritical fluid carbon dioxide were also carried out respectively, using response surface methodology (RSM) to design the experiment in order to reduce the number of experimental runs. In addition, the solubility of castor seed oil in CO_2 was measured and the density model was applied to determine the correlation of the solubility. Finally, the characterization of physiochemical properties of the castor oil such as the acid value, saponification value, iodine value, specific gravity, viscosity, refractive index, pH value and their fatty acid by employing supercritical carbon dioxide techniques. Moreover, oxidative stability changes of the castor oil were also studied by the schaal oven method at a temperature of 70 ^{o}C for 12 weeks.

1.5 Significant Contribution

The results obtained from this research will reveal the best method for extracting castor oil to achieve enriched oil at optimum yield and this will also lead to the adoption of green approaches of supercritical CO_2 in improving the oxidative stability of castor oil seed. This will provide better understanding of primary oxidative compounds, which will allow the development of more effective and durable vegetable oil. In addition, this study will go long the way in understanding the fundamental aspects affecting the operating variables in achieving the desired product, so as to tailor the process to produce the desired product in a controlled manner.

The limitations of the conventional methods of oil extraction have favoured oxidation which has led to the growing interest of scientists in the development of a more efficient method. The SC-CO_2 is expected to serve as an alternative method in improving the oxidative stability of the oil. In terms of intellectual merit, a mathematical model showing the effect of average particle size, extraction time and temperature, pressure, flow rate of CO_2 was developed for both conventional and supercritical carbon dioxide extraction.

The empirical data on the solubility behavior of castor oil extraction from castor seed using supercritical carbon dioxide will provide a significant impetus for further SFE studies, especially in SC-CO_2. Moreover, the use of a density model of the solubility approach will be envisioned as a simpler method for further prediction of solute solubility in supercritical carbon dioxide as an alternative and economical solvent in the extraction and improving oxidation stability.

1.6 Organization of the Thesis

This thesis consists of five chapters and each chapter gives specific information about the research area as follows:

Chapter One: covers the background of the study, research aims and objectives. Moreover, the chapter highlights the scope of the research and the significance of the study.

Chapter Two: presents the literature reviews relevant to the study. The use of solvent extraction, supercritical carbon dioxide, response surface methodology (RSM) for optimization of the oil yield, solubility and oxidation stability studies were also explored.

Chapter Three: covers the raw material, description of the experimental methods and the general procedures in conducting this research work. These include oil extraction and characterization. It also explains the use of design expert in optimizing the oil yield.

Chapter Four: presents and discusses the comparisons between the physiochemical properties of conventional and supercritical fluid carbon dioxide extraction method. The effects of average particle size, extraction time, and temperature, pressure, flow rate of CO_2 on the optimum oil yield were also discussed. The chapter also discussed the solubility of castor oil in CO_2 and the application of the density model on solubility. Finally, an oxidation stability study of conventional and supercritical fluid carbon dioxide extraction were also discussed.

Chapter Five: deals with conclusions and recommendations. It presents the conclusions derived from the study and proposes several recommendations for future study for better understanding of oil solubility and oxidation stability.

REFERENCES

Abaroudi, K., Trabelsi, F., Recasens, F. (2002). Screening of cosolvents for a supercritical fluid: a fully predictive approach. *American Institute of Chemical Engineers Journal*. 48 (3), 551–560.

Abayeh, O.J., Aina, E.A. and Okuonghae, C.O. (1998). Oil content and oil quality characteristics of some Nigerian oil seeds. *Journal of Pure Applied Science*. 1, 17- 23.

Abbasi, H., Rezaei, K. and Rashidi, L. (2008). Extraction of essential oils from the seeds *of pomegranate* using organic solvents and supercritical CO_2. *Journal of American Oil Chemist's Society*. 85, 83–89.

Adachi, Y. and Lu, B.C.Y. (1983). Supercritical fl uid extraction with carbon dioxide and ethylene. *Fluid Phase Equilibria*. 14, 147 – 156.

Adhvaryu, A., Erhan, S. Z., Liu, Z. S. and Perez, J. M. (2000). Oxidation kinetics Studies of Oils Derived from Unmodified and Genetically Modified Vegetables Using Pressurized Differential Scanning Calorimetry and Nuclear Magnetic Resonance Spectrocopy. *Thermochimica. Acta.* 364, 87-97.

Akande, T. O., Oduns A. A., Olabode O. S. and Ojediran T. K. (2012). Physical and Nutrient Characterisation of Raw and Processed Csator (Ricinus communis L.) Seeds in Nigeria. *World journal of Agricultural Science*. 8(1): 89-95.

Akpan, U. G., Jimoh, A. and Mohammed, A. D. (2006). Extraction, characterization and modification of castor seed oil. *Leonardo Journal Science*. 8:43–52.

Alberto, G. D. and Philips, D. T. (1995). *Principle of experimental design and analysis*. London: Chapman and Hall.

Anderson, A. (2005). *A primer on oils processing technology. In: Bailey's Industrial Oil and Fat Products*. Vol.5. Edible Oil and Fat Products: Processing Technologies. Ed. F. Shahidi, John Wiley & Sons, Hoboken, NJ (USA), pp. 1–56.

Araus, K., Uquiche, E. and del Valle, J. M. (2009). Matrix effects in supercritical CO_2 extraction of essential oils from plant material. *Journal of Food Engineering*. 92: 438–447.

Asaudauskas, S. J., Biresaw, G. and Meclure, T. G. (2010). Effects of chlorinated paraffin and ZDDP concentrations on boundary lubrication properties of mineral and soybeans oils. *Tribology*. 37, 111-121.

Ashaye, O.A. and Olusoji, O.C. (2006). Evaluation of some quality attributes of soybean oils in Ibadan. *African Journal of Agricultural Research*. 1, 17-19.

Ashour, I., Almehaideb, R., Fateen, S.E. and Aly, G. (2000). Representation of solid-supe-rcritical fluid phase equilibria using cubic equations of state. *Fluid Phase Equilibria*. 167, 41–6.

ASTM. (2002). *International Standard test method for oxidation onset temperature of hydrocarbons by differential scanning calorimetry*. Annual Book of Standards, section 12.10, ASTM International, West Conshohocken.

Atkins, P. W. (2002). Kurzlehrbuch Physikalische Chemie. 3rd ed. Weinheim, Germany: Wiley-VCH.

Azizi, C.Y. M. (2007) Extraction, identification and separation of vitamin E and djenkolic acid from pithecellobium jiringan (jack) prain seed using supercritical carbon dioxide. Penang, Malaysia: *Universiti Sains Malaysia*.

Azmir, J., Zaidul I.S.M., Rahman, M.M., Sharif, K.M., Sahena, F., Jahurul, M.H.A. and Mohamed, A. (2014). Optimization of oil yield of Phaleria macrocarpa seed using responsesurface methodology and its fatty acids constituents. *Industrial Crops and Products*. 52, 405– 412.

Bagheri, H., Abdul Manap, M. Y. and Zeinab Solati, Z. (2014). Response surface method-ology applied to supercritical carbon dioxide extraction of Piper nigrum L. essential oil. *LWT - Food Science and Technology*. 57, 149-155.

Bamberger, T., Erickson, J. C., Cooney, C. L. and Kumar, S. K. (1988). Measurement and model prediction of solubilities of pure fatty-acids, pure triglycerides, and mixtures of triglycerides in supercritical carbon dioxide*Journal Chemical Engineering Data*. 33, 327-333.

Bartle, K.D., Clifford, A. A., Jafar, S. A. and Shilstone, G. F. (1999). Solubilities of solids and liquids of low volatility in supercritical carbon dioxide. *Journal of Physical and Chemical Reference Data*. 20, 201–219.

Barwick, V. J. (1997). Strategies for solvent selection – a literature review. *Trends Analytical Chemistry*. 16, 293-309.

Bas, D. and Boyaci, I. H. (2007). Modeling and optimization I: Usability of response surf-ace methodology. *Journal of Food Engineering*. 78,836–845.

Baysal, T., Eesus, S. and Starmans, D. A. J. (2002). Supercritcal extraction of B-carotene and lycopene from tomato paste waste. *Journal of Agriculture and Food Chemistry*. 48: 5507-5511.

Belgacem, M. N. and Gandini, A. (2008). Materials from vegetable oils: Major sources, properties and applications. In: *Monomers, Polymers and Composites from Renewable Resources.* Eds. M N. Belgacem, A. Gandini, Elsevier, Amsterdam (The Netherlands), pp. 39–66.

Bello, E. I. and Makanju, A. (2011). Production, Characterization and Evaluation of Castor oil Biodiesel as Alternative Fuel for Diesel Engines. *Journal of Emerging Trends in Engineering and Applied Sciences (JETEAS).* 2(3): 525-530.

Benelli, P., Riehl, C. A. S., Smânia, A. Jr., Smânia, E. F. A. and Ferreira, S. R. S. (2010). Bioactive extracts of orange (Citrus sinensis L. Osbeck) pomace obtained by SFE and low pressure techniques: Mathematical modeling and extract composition. *Journal of Supercritical Fluids*. 55: 132-141.

Berger, T. A, Fogleman, K., Staats, T., Bente, P., Crocket, I., Farrell, W. and Osonubi, M. (2000). The development of a semi-preparatory scale supercritical fluid chromatograph for high-throughput purification of ‘combi-chem’ libraries. *Journal of Biochemistry Biophysic Methods*. 43: 87–111.

Berna, A., Cha´fer, A. and Monto´n, J. B. (2000). Solubilities of Essential Oil Components of Orange in Supercritical Carbon Dioxide. *Journal of Chemical Engineering Data*. 45, 724-727.

Bernardo-Gil, M. G. (2013). *Engineering aspects of food biotechnology*. In: Jose, A.T. Antonio, A.V, editors. Contemporary Food Engineering. Boca Raton, FL: CRC Press, Taylor & Francis Group. pp 215–235.

Bernado-Gil, M. G. and Lopes, L.M.C. (2004). Supercritical fluid extraction of Curcurbita ficifolia seed oil. *European Food Res. Technology.* 219, 593.

Bernado-Gil, G. et al. (2001). Extraction of lipids from cherry seed oil using supercritical carbon dioxide. *European Food Res Technology*. 212, 170.

Bernado-Gil, M. G., Grenha, J., Santos, J. and Gardos, P. (2002). Supercritical fluid extraction and characterisation of oil from hazelnut. *European Journal of Lipid Science Technology*. 104: 402-409.

Bettelheim, F. A., Brown, W. H., Campbell, M. K. and Farrell, S. O. (2010). *Introduction to general organic and biochemistry.* 9th edition. ed. Belmont, USA: Brooks/Cole, Cengage Learning.

Bezerra, M. A., Santelli, R. E., Oliveira, E. P., Villar, L. S. and Escaleira, L. A. (2008). R-esponse surface methodology (RSM) as a tool for optimization in analytical chemistry. *Talanta*. 76, 965-977.

Bharath, R., Inomata, H., Arai, K., Shoji, K. and Noguchi, Y. (1989). Vapor-liquid Equilibria for binary mixtures of carbon dioxide and fatty acid and ethyl esters. *Fluid Phase Equilibria.* 50, 315.

Bimakr, M., Rahman, R. A., Ganjloo, A., Taip, F. S., Saleh, L.M. and Sarker, Z. I. (2012). Optimization of supercritical carbon dioxide extraction of bioactive flavonoid compounds from spearmint (Mentha spicata L.) leaves by using response surface methodology. *Food Bioprocess Technology*. 5: 912-920.

Bimakr, M., Rahman, R. A., Saleena, T. F., Adzahan, N. M., Sarker, Md. Z. I. and Ganjlo-o A. (2013). Supercritical Carbon Dioxide Extraction of Seed Oil from Winter

Melon (*Beninca-sa hispida*) and Its Antioxidant Activity and Fatty Acid Composition. *M-olecules*. 18, 997-1014.

Bisunada, M. (1993). Extraction of iol from oil palm fruits using supercritical carbon dio-xide. MSc thesis, University of Sains Malaysia, Penang Malaysia.

Blakeway, J.M., Frey, M.L., Lacroix, S. and Salerno, M.S. (1987). Chemical reaction in perfume ageing. *International Journal of Cosmetic Science*. 9, 203–14.

Bocevska, M. and Sovová, H. (2007). Supercritical CO_2 extraction of essential oil from y-arrow. *Journal of supercritical Fluid*. 40: 360–367.

Bozan, B. and Temelli, F. (2002). Supercritical CO_2 extraction of flaxseed. *Journal of American Oil Chemist's Society*. 79, 231.

Bravi, M., Spinoglio, F., Verdone, N., Adami, M., Aliboni, A., D'Andrea, A., de Santis, A. and Ferri, D. (2007). Improving the structure of α-tocopherolenriched oil from grape seeds by supercritical CO_2. Optimisation of the extraction conditions. *Journal of Food Enginering.* 78: 488–493.

Brennecke, J.F. and Eckert, C.A. (1989). Phase equilibria for supercritical fluid process d-esign. *American Institute of Chemical Engineering Journal*. 35, 1409 – 1427.

Brown, W.H., Foote, C.S. and Iverson, B.L. (2005). *Organic Chemistry.* (4^{th} ed.) States: Thomson Learning, Inc.

Brunner, G. (2005). Supercritical fluids: technology and application to food processing. *J-ournal of Food Engineering.* 67: 21–33.

Cao, X. and Ito, Y. (2003). Supercritical fluid extraction of grape seed oil and subsiquent s-eparation of free fatty acids by high-speed counter-current chromatography. *Journal of Chromatography*. 1021: 117-124.

Carr, A. G., Mammucari, R., Foster, N. R. (2011). A review of subcritical water as a solvent and its utilisation for the processing of hydrophobic organic compounds. *Chemical Engineering Journal*. 172(1), 1–17.

Catchpole, O.J., Grey, J.B. and Noemark, K.A. (2000). Fractionation of fish oils using supercritical CO_2 and CO_2 plus ethanol mixtures. *Journal of Supercritical Fluids*. 19, 25–37.

Catchpole, O. J., Perry, N. B., da Silva, B. M. T., Grey, J..B. and Smallfield, B. M. (2002). Supercritical extraction of herbs I: saw palmetto, St John's wort, kava root, and Echinacea. *Journal of Supercritical Fluids*. 22: 129–138.

Catchpole, O. J. and Proells, K. (2001). Solubility of Squalene, Oleic Acid, Soya Oil, and Deep Sea Shark Liver Oil in Subcritical R134a from 303 to 353 K. *Ind. Eng. Chem. Res*. 40, 965-972.

Chaiyasit, W., Elias, R., McClements, D. J. AND Decker, E. A. (2007). Role of physical structures in bulk oils on lipid oxidation. *Crit. Rev. Food Sci. Nutr*. 47, 299-317.

Chapard, C., Mesnard, P. and Fourtillan, J.B. (1972). Analyse comparee des essences de pin, de niaouli et d'eucalyptus. Essai d'identification des composes formes au cours de l'oxydation de ces essences. Bull Soc Pharm Bordeaux. 111,183–99.

Cheah, L. and Chin, E. (2009). *Study of extraction processes and their impact on bio-activity of botanicals. Singapore*: National University of Singapore.

Chen, Q., Ruan, H., Zhang, H., Ni, H. and He, G. (2007). Enhanced production of elastase by Bacillus Lichenformis ZJUEL31410: Optimization of cultivation conditions using response surface methodology. *Journal of Zhejiang University, Science B.* 8(11), 845-852.

Chen, X.W., Li, X.M., Chen, H. and Li, S.G. (2002). Researches on the optimal working parameters in the extraction of wheat germ oil with supercritical carbon dioxide. *China Food Science*. 23, 151–153.

Chiavaro, E., Rodriguez-Estrada, M.T., Vittadini, E. and Pellegrini, N. (2010). Microwave heating of different vegetable oils: relation between chemical and thermal parameters. *LWT-Food Scence Technology*. 43, 1104–12.

Chow, C. K. E. (2007). *Fatty Acids in Foods and their Health Implications.* 3rd. ed. University of Kentucky, Lexington, USA,: CRC Press.

Chrastil, J. (1982). Solubility of solids and liquids in supercritical gases. *Journal of Physical Chemistry*. 86, 3016–3021.

Clifford, T. (1998). *Fundamentals of supercritical Fluids*. New York, USA. Oxford University Press.

CODEX (1982). Alimentarius Commission. *Recommended International Standards for edible Arachis oil*,1st ed, FAO/WHO: Rome.

Coimbra, P., Duarte, C.M.M. and de Sousa, H.C. (2006). Cubic equation-of-state correlation of the solubility of some anti-inflammatory drugs in supercritical carbon dioxide. *Fluid Phase Equilibria*. 239, 188–199.

Cornell, J.A. (1990). *How to Apply Response Surface Methodology,* Revised ed. Tammy Griffin, United State of America: Book Crafters.

Cosmetic Ingredient Review Expert Panel (CIREP). (2007). Final report on the safety assessment of Ricinus communis (Castor) seed oil. *International Journal of Toxicology*. 26:31–77.

Danh, L.U., Mammucari, R., Truong, P. and Foster, N. (2009). Response surface method applied to supercritical carbon dioxide extraction of *vetiveria zizanoides* essential oil. *Chemical Engineering Journal*. 155, 617-626.

Danh, L. T, Truong, P. and Mammucari, R., Foster, N. (2010). Extraction of vetiver essential oil by ethanol-modified supercritical carbon dioxide. Chem Eng J. 165: 26–34.

Danielski, L., Brunner, G., Schwänke, C., Zetzl, C., Hense, H. and Donoso, J. P. M. (2008) .Deterpenation of mandarin (*Citrus reticulata*) peel oils by means of countercurrent multistage extraction and adsorption/ desorption with supercritical CO_2. *Journal of Supercritical Fluid*. 44, 315–324.

Danlami, J. M., Agus, A., Muhammad, A. A. Z and Hanizam, S. A. (2014). Comparative study of various oil extraction techniques from plants. *Reviews in Chemical Engineering*. 30, 605–626.

Danlami J.M., Agus, A., Muhammad, A. A. Z. and Azizi, M.C.Y. (2015). A parametric investigation of castoroil (*Ricinus comminis* L) extraction using supercritical carbondioxide *via* response surface optimization, *Journal of the Taiwan Institute of Chemical Engineers* .http://dx.doi.org/10.1016/j.jtice.2015.02.033

Daood, H. G., Illes, V., Gnayfeed, M.H., Meszaros, B., Horvath, D. and Biacs, P.A. (2002) .Extraction of pungent spice paprika by supercritical carbon dioxide and subcritical propane. *Journal of supercritical fluids*. 23, 143-152.

Dauksas, E., Venskutonis, P. R. and Sivik, B. (2002). Supercritical fluid extraction of borage (*Borago officinalis* L.) seeds with pure CO_2 and its mixture with caprylic acid methyl ester. *Journal of Supercritical Fluids*. 22, 211–219.

de Azevedo, A.B.A., Kieckbush, T. G., Tashima, A. K., Mohamed, R. S., Mazzafera, P. and Melo, S. A. B. V. D. (2008). Extraction of green coffee oil using supercritical carbon dioxide. *Journal of Supercritical Fluids*. 44, 186–192.

Dean, J. R. (1993). Application of supercritical fluids in industrial analysis. *Blackie Academic and professional publishers.* Glassgow.

del Valle, J.M. and Aguilera, J.M. (1988). An improved equation for predecting the solubility of vegetable oils in supercritical CO_2, *Industrial and Engineering Chemistry Research*. 27, 1551–1553.

Del Valle, J. M., Jimenez, M. and De la Fuente, J. C. (2003). Extraction kinetics of prepelltized jalapeno peppers with supercritical CO_2. *Journal of Supercritical Fluids.*14, 247-256.

Doker, O. (2004). Modelling of extraction of b-carotene from apricot bagasse using supercritical CO_2 in packed bed extractor. *Journal of Supercritical Fluids.* 28, 11-19.

Döker, O., Salgin, U., Yildiz, N., Aydo~mu, M., Çalimli, A. (2010). Extraction of sesame seed oil using supercritical CO2 and mathematical modeling. *Journal of Food Engineering.* 97, 360-366.

Donelian, A., Carlson, L., Lopes, T. and Machado, R. (2009). Comparison of extraction of patchouli (*Pogostemon cablin*) essential oil with supercritical CO_2 and by steam distillation. *Journal of Supercritical Fluids*. 48, 15–20.

Effendi, C., Shanty, M., Ju, Y. H, Kurniawan, A., Wang, M. J., Indraswati, N. and Ismadji, S. (2013). Measurement and mathematical modeling of solubility of buttery-odor substance (acetoin) in supercritical CO_2 at severalpressures and temperatures. *Fluid Phase Equilibria*. 356, 102– 108.

Essien, E.A, Umoren, S.A, Essien, E.E. and Udoh, A.P. (2012). Preparation and evaluation of *Cucumeropsis mannii* Naud seed oil metallic soaps as driers in gloss paint. *Journal of Material and Environmental Science*. 3, 477-484.

European food safety authority. (2008). Ricin (from Ricinus communis) as undesirab-le subtances in animal feed Scientific Opinion of the Panel on Contaminants in the Food Chain. *European food safety authority journal.* 726, 1-38.

European Union (2003). EC Regulation 1989/2003 of 6 November 2003. Official *Journal of European Communities*, L295.

Evans, W. C. (2002). *General methods associated with the phytochemical investigation of herbal products*. In: E. Daphne, editor. Trease and Evans pharmacognosy, 15th ed. New Delhi: Saunders (Elsevier). 137–148.

Fang, T., Goto, T., Sakaki, M. D. and Yang, D. (2008). *Extraction and purification of natural tocopherols by supercritical CO_2*, in: J.L. Martinez (Ed.), Supercritical Fluid Extraction of Nutraceuticals and Bioactive Compounds, CRC Press, Taylor & Francis Group, Boca Raton, FL. 103–140.

Favati, F., King, J. W. and Mazzanti, M. (1991). Supercritical carbon dioxide extraction of evening primrose oil. *Journal of the American Oil Chemists' Society*. 68(6), 422-427.

Food and agriculture organization of the united nations (FAOSTAT). FAO. (2008). Online http://faostat.fao.org.

Foster, N., Mammucari, R., Dehghani, F., Barrett, A., Bezanehtak, K., Coen, E., Combes, G., Meure, L., Ng, a., Regtop, H. L. and Tandya, A. (2003). Processing pharmaceutical compounds using dense gas technology. *Industrial and Engineering Chemistry Research.* 42: 6476-6493.

Fox, N. J. and Stachowiak, G. W. (2007). Vegetable oil based lubricant- A review of oxidation. . *Tribology International.* 40: 1035-1046.

Gaspar, F., Tiejun Lu, T., Marriott, R., Mellor, S., Watkinson, C., Al-Duri, B., Santos, R. and Seville, J. (2003). Solubility of Echium, Borage, and Lunaria Seed Oils in Compressed CO_2. *Journal of Chemical. Engineering Data*. 48, 107-109.

Geier, K. (2006). Qualitassicrung von atherischen olem (PhD thesis) Munich: Veriag Dr. Hut.

Gemene, K. L. and Bakker E. (2008). Direct Sensing of Total Acidity by Chronopotentiometric Flash Titrations at Polymer Membrane Ion-Selective Electrodes. *Analytical Chemistry*. 80, 3743-3750.

Gerard, D. and May, P. (2002). Herb and spice carbon dioxide extracts-versatile, safe ingr-dients for premium food and health food. *Food Technology*. 2, 1-5.

Ghasemi, E., Yamini, Y., Bahramifar, N. and Sefidkon, F. (2007). Comparative analysis of the oil and supercritical CO_2 extract of *Artemisia sieberi*. *Journal of Food Engineering*. 79, 306–311.

Giovanni, M. (1983). Response surface methodology and product optimization. *Food Te-chnology*. 37, 41–45.

Glišić, S. B., Mišić, D. R., Stamenić, M. D., Zizovic, I. T., Ašanin, R. M. and Skala, D. U. (2007). Supercritical carbon dioxide extraction of carrot fruit essential oil: chemical composition and antimicrobial activity. *Food Chemistry*. 105, 346–352.

Goodrum, J.W. and Kilgo, M.B. (1987). Peanut oil extraction with SC-CO_2: solubilility and kinetic functions. *Transactions of the American Society of Agricultural Engineers*. 30, 1865-1868.

Gomes, P. B., Mata, V. G. and Rodrigues, A. E. (2007). Production of rose geranium oil u-sing supercritical fluid extraction. *Journal of Supercritical Fluids.* 41, 50-60.

Gomez, A. M. and de la Ossa, E. M. (2002). Quality of borage seed oil extracted by liquid and supercritical carbon dioxide extraction. *Chemical Engineering Journal*. 88, 103.

Gomez, A. M., Lopez, C.P. and de la Ossa, E. M. (1996). Recovery of grape seed oil by liquid and supercritical carbon dioxide extraction: A comparison with conventional solvent extraction. *Chemical Engineering Journal*. 61, 227-231.

Gomez, A. M., and Martinez, de la O. (2000). Quality of wheat germ oil extracted by liquid and supercritical carbon dioxide. *Journal of American oil Chemist's Society*. 77(9), 969-976.

Gracia, I., García, M. T, Rodríguez, J. F., Fernández, M. P. and de Lucas, A. (2009). Modelling of the phase behaviour for vegetable oils at supercritical conditions. *Journal of Supercritical Fluids*. 48, 189–194.

Grosso, C., Ferraro, V., Figueiredo, A., Barroso, J., Coelho, J. and Palavra, A. (2008). Supercritical carbon dioxide extraction of volatile oil from Italian coriander seeds. *Food Chemistry*. 111, 197–203.

Guan, W., Li, S., Yan, R., Tang, S. and Quan, C. (2007). Comparison of essential oils of clove buds extracted with supercritical carbon dioxide and other three traditional extraction methods. *Food Chemistry*. 101: 1558–1564.

Guan, X. and Yao, H. (2008). Optimization of *viscozyme L* assisted extraction of oat bran protein using respond surface methodology. *Food Chemistry*. 106, 345 – 35.

Guclu-Ustundag, O. and Temelli, F. (2000). Correlating the solubility behaviour of fatty acids, mono-, di-, and triglycerides, and fatty acid esters in supercritical carbon dioxide. *Industrial and Engineering Chemistry Research*. 39, 4756– 4766.

Guclu-Ustundag, O. and Temelli, F. (2006). Solubility behavior of ternary systems of lipids in supercritical carbon dioxide. *Journal of Supercritical Fluids*. 38, 275–288.

Guclu-Ustundag, O. and Temelli, F. (2007). Column fractionation of canola oil deodorizer distillate using supercritical carbon dioxide. *Journal of American Oil Chemist's Society*. 84, 953–961.

Guo, H., Zhang, Z., Qian, J. and Liu, Y. (2013). Optimization of the liquid–liquid extraction of phosphatidyl-choline from rapeseed oil gums by response surface methodology. *Industrial Cropsand Products*. 42, 500– 506.

Guoliang, L., Junyou, S., Yourui, S., Zhiwei, S., Xia Lian, X., Zheng Jie, Z., You Jinmao, Y, and Liu Yongjun, L. (2011). Supercritical CO_2 cell breaking extraction of *Lycium barbarum* seed oil and determination of its chemical composition by HPLC/APCI/MS and antioxidant activity. *LWT - Food Science and Technology*. 44, 172-1178.

Gupta, R. B. and Shim, J. J. (2007). *Solubility in supercritical carbon dioxide.* Boca Raton, CRC Press, Taylor & Francis Group. 1-18.

Gupta, A., Madhu, N. and Vijay, K. (2012). Modern extraction methods for preparation of bioactive plant extracts. *International Journal of Applied and Natural Sciences*. 1, 8–26.

Hagvall, L. (2009). Formation of skin sensitizers from fragrance terpenes via oxidative activation routes [dissertation]. Gothenburg: University of Gothenburg. 62 p. Available from: Department of Chemistry, *University of Gothenburg*, Gothenburg.

Handel, A. P. and Guerrieri, S. A. (1990). Evaluation of heated frying oils containing added fatty acids. *Journal of Food Science*. 55, 1417–1420.

Hamburger, M., Baumann, D. and Adler, S. (2004). Supercritical carbon dioxide extractio-n of selected medicinal plants – effects of high pressure and added etha- ethanol on yield of extracted substances. *Phytochemical Analysis*. 15, 46–54.

Han, X., Cheng, L., Zhang, R. and Bi, J. (2009). Extraction of safflower seed oil by supercritical CO_2. *Journal of Food Engineering.* 92, 370-376.

Hanmoungjai, P., Pyle, L. and Niranjan, K. (2000). Extraction of rice bran oil using acqu-eous media. *Journal of Chemical Technology and Biotechnology*. 75, 348–352.

Harlalka, R. (2008). Solid phase micro-extraction and headspace trapping extraction. In: S. S Handa, S.P.S Khanuja, G. Longo, D.D. Rakesh, editors. Extraction technologies for medicinal and aromatic plants. *International centre for science padriciano* 99. Triesta, Italy, UNIDO. 145–167.

Harvey, A. H. (1990). Supercritical solubility of solids from near-critical dilute-mixture t-heory. *Journal of Physical Chemistry*. 94, 8403 – 8406.

Hassan, M.N., Ab. Rahman, N.N and Ab. Kadir, M.O. (1999). Major chemical constituen-ts of supercritical carbon dioxide extract of pandanus odorus leave. *Journal of Natural product science*. 5(2), 75-79.

Hellerstrom, S., Thyresson, N., Blohm, S.G. and Widmark, G. (1955). On the nature of the eczematogenic component of oxidized carene. *Journal of Investigative Dermatologt*. 24, 217–24.

Herrero, M., Mendiola, J. A., Cifuentes, A. and Ibáñez, E. (2010). Supercritical fluid extra-ction: Recent advances and applications. *Journal of Chromatography A.* 1217: 2495-2511.

Higuera-Ciapara, I., Toledo-Guillen, A. R., Noriega-Orozco, L., Martinez-Robinson, K.G. and Esqueda-Valle, M.C. (2005). Production of a low-cholesterol shrimp using supercritical extraction. *Journal of Food Process Engineering*. 28, 526–538.

Hossein, B., Mohd Yazid, B. A. M. and Zeinab, S. (2014). Response surface methodology applied to supercritical carbon dioxide extraction of *Piper nigrum L*. essential oil. *LWT-Food Science and Technology*. 57, 149-155.

Huie, C. W. (2002). A review of modern sample-preparation techniques for the extraction and analysis of medicinal plants. *Analytical and Bioanalytical Chemistry*. 373, 23–30.

Ibanez, E., Benavides, A.M.H. Senorans, F.J. and Reglero, G. (20020. Concentration of sterols and tocopherols from olive oil with supercritical carbon dioxide. *Journal of Americal Oil Chemist's. Society*. 79, 1255–1260.

Ibánez, E., Herrero, M., Mendiola, J. and Castro-Puyana, M. (2012). Extraction and characterization of bioactive compounds with health benefits from marine resources: macro and micro algae, cyanobacteria and invertebrates. In: Hayes M, editor. Marine bioactive compounds. New York: *Springer*. 55–98.

Ill´es, V., Daood, H.G., Perneczki, S. and Szokonya, L.M. (2000). Then, Extraction of coriander seed oil by CO_2 and propane at super- and subcritical conditions. *Journal of Supercritical Fluids*. 17, 1- 77–186.

Inomata, H., Kondo, T., Hirohama, S., Arai, K., Suzuki, Y. and Konno, M. (1989). Vapor-liquid Equlibria for binary mixtures of carbon dioxide and fatty acid and methyl esters. *Fluid Phase Equilibria*. 46, 41.

Iwai, Y., Yamamoto, Y. (2013) Concentration dependent surface area parameter model for calculation of activity coefficients. *Fluid Phase Equilib*. 337 (0), 165–173.

Ixtaina, V., Veja, A, Nolasco, S.M., Tomas, M.C, Gimeno, M., Barzana, E. and Tecante, A. (2010). Supercritical carbon dioxide extraction of oil from Mexican chia seed (Salvia hispanica L.): characterization and process optimization. *Journal of Supercritical Fluids*. 55, 192–9.

Izuagie, A., Akpambang, V.O.E. and Amoo, A. (2008). Comparative compositional analysis on two varieties of melon (*Colocynthis citrullus* and *Cucumeropsis edulis*) and a variety of almond (*Prunus amydalus*). *Research Journal of Agricultural and Biological Sciences*. 4, 639-642.

Jadhav, D., Rekha, B.N., Gogate, P.R. and Rathod, V.K. (2009). Extraction of vanillin from vanilla pods: a comparison study of conventional Soxhlet and ultrasound assisted extraction. *Journal of Food Engineering*. 93, 421–426.

Jaswir I., Kitts, d. D., Che Man, Y. B. and Hassan, T. H. (2005). Physio-Chemical stability of flaxseed oil with natural antioxidants mixture during heating. *Journal of Oleo Science*. 54, 71-79.

Jebe, T.A., Matlock, M.G. and Sleeter, R.T. (1993). Collaborative study of the oil stability index analysis. *Journal of American Oil Chemist's Society*. 70, 1055–61.

Jose, A. C., Alberto, C. H. and Denise, M. G. F. (2013). A potential biodegrable lubricant from castor biodiesel esters. *Lubrication Science Wiley online Library*. 25 (1), 53-61.

Joseph, P. V., Saxena, D. and Sharma, D. K. (2007). Study of some non-edible vegetable oils of Indian origin for lubricant application. *Journal of Synthetic Lubrication.* 24: 181-197.

Kamal-Eldin, A. and Yanishlieva, N.V. (2002). n-3 fatty acids for human nutrition: stability considerations. *European Journal of Lipid Science Technology*. 104, 825-836.

Kapchie, V. N., Yao, L., Hauck, C.C., Wang, T. and Murphy, P. A. (2013). Oxidative stability of soybean oil in oleosomes as affected by pH and iron. *Food Chemistry*. 141, 2286-93.

Kemper, T. G. (2005). *Oil extraction*. In: Bailey's Industrial Oil and Fat Products. Edible Oil and Fat Products: Processing Technologies. Ed. F. Shahidi, John Wiley & Sons, Hoboken, NJ (USA), 185: 57–98.

Khattab,R., Rempel, C., Suh, M., Thiyam, U. (2012). Quality of Canola Oil Obtained by Conventional and Supercritical Fluid Extraction. *American Journal of Analytical Chemistry*. 3, 966-976.

Kiriamiti, H., Rascol, E., Marty, A. and Condoret, J. (2001). Extraction rates of oil from high oleic sunflower seeds with supercritical carbon dioxide. *Chemical Engineering Process*. 41, 711–718.

King, M.B. and Bott, T.R. (1993). *Extraction of Natural Products Using Near- Critical s-olvents.* Chapman and Hall, Glaskow, UK, pp: 1-25.

King, J.W. and Dunford, N.T. (2002). Phytosterol-enriched triglyceride fractions from ve-getable oil deodorizer distillates utilizing supercritical fluid fractionation technology. *Seperation. Science Technology*. 37, 451–462.

Kitzberger, C. S. G., Lomonaco, R. H., Michielin, E. M. Z., Danielski, L., Correia, J. and Ferreira, S. R. S. (2009). Supercritical fluid extraction of shiitake oil: Curve modeling and extract composition. *Journal of Food Engineering*. 90, 35–43.

Klein, T. and Schulz, S. (1989). Measurement and model production of vapour-liquid Eq-uilibria of mixtures of rapeseed oil and supercritical carbon dioxide. *Industrial and Engineering Chemistry and Research*. 28, 1073.

Kong, Y., Fu, Y. J., Zu, Y. G., Liu, W., Wang, W., Hua, X. and Yang, M. (2009). Ethanol modified supercritical fluid extraction and antioxidant activity of cajaninstilbene acid and pinostrobin from pigeonpea *Cajanus cajan* (L.) Millsp. leaves. *Food Chemistry*. 117: 152–159.

Kroon, J. J. and Raynie, D. E. (2010). *Supercritical fluid extraction*. In: Pawliszyn J, edit-or. Handbook of sample preparation. New Jersey: John Wiley & Sons Inc., pp. 191.

Kumar, S. and Johnston, K. P. (1988). Modelling the solubility of solids in supercritical f-luids with density as independent variable. *Journal of Supercritical Fluids*. 1, 15–22.

Kumoro, A. C., Hassan, M. and Singh, H. (2009). Effects of solvent properties on soxhlet extraction of *diterpenoid lactones* from *andrograhis paniculata* leaves. *Science Asia*. 35, 306-309.

Kviecinski, M. R., Benelli, P., Felipe, K. B., Correia, J. F. G., Pich, C.T., Ferreira, S. R. S. and Pedrosa, R. C. (2011). SFE from Bidens pilosa Linné to obtain extracts rich in cytotoxic polyacetylenes with antitumor activity. *Journal of Supercritical Fluids*. 56, 243-248.

Laguerre, M., Lecomte, J. and Villeneuve, T. (2007). Evaluation of ability of antioxidants to counteract lipid oxidation: Existing methods, new trends and challenges. Prog. *Lipid Research.* 46, 244-282.

Lang, Q. and Wai, C. M. (2001). Supercritical fluid extraction in herbal and natural produ-ct studies – a practical review. *Talanta*. 53, 771–782.

Langa, E., Porta, G. D., Palavra, A. M. F., Urieta, J. S. and Mainar, A. M. (2009). Supercr-itical fluid extraction of Spanish sage essential oil: optimization of the process parameters and modelling. *Journal of Supercritical Fluids*. 49, 174–181.

Lee, J. H., Asaduzzaman, A. K. M., Yun, J. H., Yun, J. H., and Chun, B. S. (2012). Char-acterization of the Yellow Croaker *Larimichthys polyactis* muscle Oil Extracted with Supercritical Carbon Dioxide and an Organic Solvent. *Fisheries and Aquatic Sciences*. 15(4), 275-281.

Leeke, G., Gaspar, F. and Santos, R. (2002). Influence of water on the extraction of essen-tial oils from a model herb using supercritical carbon dioxide. *Industrial and Engineering Chemistry Research*. 41, 2033–2039.

Leo, L., Rescio, L., Ciurlia., L. and Zacheo, G. (2005). Supercritical carbon dioxide extr-action of oil and α-tocopherol from almond seeds. *Journal of the Science of Food and Agriculture.* 85, 2167-2174.

Li, H., Pordesimo, L. and Weiss, J. (2004). High intensity ultrasound-assisted extraction of oil from soybeans. *Food Research International*. 37,731–738.

Li, M., Bellmer, D. D. and Brusewitz, G. H. (1999). Pecan kernel brekage and oil extracte-d by supercritical CO_2 as affected by moisture content. *Journal of Food Sciences*. 64, 1084.

Liu, G. and Nagahama, K. (1996). Solubility of organic solid mixture in supercritical fluid . *Journal of Supercritical Fluids*. 9, 152 – 160.

Liu, G., Xu, X., Hao, Q. and Gao, Y. (2009). Supercritical CO_2 extraction optimization of pomegranate (*Punica granatum* L.) seed oil using response surface methodology. *LWT-Food Science Technology*. 42, 1491-1495.

Liu, W., Yu, Y., Yang, R., Wan, C., Xu, B. and Cao, S. (2010). Optimization of total flavonoid compound extraction from Gynura medica leaf using response surface methodology and chemical composition analysis. *International Journal of Molecular Science.* 11, 4750-4763.

Liu, Z., Lijuan Mei, L., Wang, Q., Shao, Y. and Tao, Y. (2014). Optimization of subcritical fluid extraction of seed oil from Nitraria tangutorum using response surface methodology. *LWT - Food Science and Technology*. 56, 168-174.

Liza, M. S., Abdul Rahman, R. A., Mandana, B. , Jinap, S. , Rahmat, A. , Zaidul, I. and Hamid, A. (2010). Supercritical carbon dioxide extraction of bioactive flavonoid from Strobilanthes crispus (Pecah Kaca). Food Bioprod. Process. 88, 319-326.

Louli, V., Folas, G., Voutsas, E. and Magaulas, K. (2004). Extraction of parsley seed oil by supercritical CO_2. *Journal of Supercritical Fluids.* 30, 163-174.

Lu, X. and Rasco, B. (2010). Rapid determination of antioxidant content and activity in foods using infrared spectroscopy and chemometrics: a review. *Critical Reviews in Food Science Nutrition*. 52, 853–885.

Lucas, J. M. (1994). How to achieve a robust process using response surface methodology. .*Journal of Quality Technology*. 24(6), 248-260.

Lucas de, A., Martinez de la. O. E., Rincon, J., Blanco, M. A. and Gracia, I. (2002). Supercritical fluid extraction of of tocopherol concentrates from olive tree leaves. *Journal of Supercritical Fluids*. 22, 221-228.

Luengthanaphol, S., Mongkholkhajornsilp, D., Douglas, S., Douglas, P., Pengsopa, L. and Pongamphai, S. (2004). Extraction of antioxidant from sweet Thai tamarind seed coat-preliminary experiments. *Journal of food Engineering*. 63, 247-252.

Luque de Castro, M. D., Valcarcel, M. and Tena, M. T. (1994). *Analytical supercritical fluid extraction*. Butterworth Publisher's. Boton.

Luque de Castro, M. and Priego, C. (2010). Soxhlet extraction: Past and Panacea. *Journal of Chromatography A.* 1217, 2383-2389.

Madaeni, S. S., Arast, N., Rahimpour, F. and Arast, Y. (2011). Fabrication optimization of acrylonitrile butadiene styrene (ABS)/polyvinylpyrrolidone (PVP) nanofilteration membrane using response surface methodology. *Desalination*. 280, 305-312.

Majewski, W., Mengal, P., Perrut, M., Ecalard, J. P. (1991). "Supercritical fluid fractionation of butter oil," *Proceedings: Supercritical fluid processing of biomaterials: Basics of process design and applications symposium, Toronto*. 123.

Malakahmada, A. and Chuan, S. Y. (2013). Application of response surface methodology to optimize coagulation-flocculation treatment of anaerobically digested palm oil mill effluent using alum. *Desalination and Water Treatment*. 51(34-36), 6729-6735.

Mamidipally, P. K. and Liu, S. X. (2004). First approach on rice bran oil extraction using limonene. *European Journal of Lipid Science Technology*. 106, 122–125.

Maria, S. G. P., Maria, L. R. D. C., Gema, F., Guillermo, S. M. and Gracia, P. B., (2007). Applicacation of Chrastil's Model to the Extraction in SC-CO_2 OF β-carotene and Lutein in *Mentha spicata* L. *The Journal of Supercritical Fluids*. 43: 32–36.

Marilena, M., Cristina, M. Q., Alessandra dos, S. T., Humbervania, R. G., da S.Jaques, D. S., Pedro da, C. N., Luri, M. P., Mariana, A. S. and Luciana, L. N. (2011). Determination of the oxidation stability of biodiesel and oils by spectrofluorimetry and multivariate calibration. *Talanta*. 85, 430-434.

Mariod, A., Ismail, M., Abd Rahman, N. F., and Matthaus, B. (2013).Stability of rice bra-n oil extracted by SFE and soxhlet methods during accelerated shelf life storage. *Grasas Aceites* 65 1 e013. doi: http://dx.doi.org/10.3989/gya.109413.

Marongiu, B., Porcedda, S., Piras, A., Rosa, A., Deiana, M. and Dess`I, M. A. (2004). An-ioxidant activity of supercritical extract of *Melissa officinalis* subsp. *officinalis* and *Melissa officinalis* subsp. *inodora*, *Phytotheraphy Research*. 18, 789–792.

Martínez, M. L., Mattea, M. A. and Maestri, D. M. (2008). Pressing and supercritical carb-on dioxide extraction of walnut oil. *Journal of Food Engineering*. 88, 399–404.

Mazalli, M.R. and Bragagnolo, N. (2007). Validation of two methods for fatty acid analysis in eggs. *Lipids*. 42, 483–490.

McClements, D. J. and E. A. Decker. (2008). *Lipids*. In: Damodaran, S., K. L. Parkin and O. R. Fennema, editors, Fennema's Food Chemistry, 4th ed. CRC Press, Boca Raton, FL. pp. 155-216.

McHugh, M. A. and Krukonis, V. J. (1994). *Supercritical fluid extraction: principles and practice*, 2nd ed. Boston, MA: Butterworth-Heine..

McHugh, M. A., Watkins, J. J., Doyle, B. T. and Krukonis, V. J. (1988). High-pressure naphthalene-xenon phase behavior. *Industrial and Engineering Chemistry Research.* 27, 1025 – 1033.

Megan, M., Robert, H. and Stephanie, F. (2002). *Effect of operating conditions on static/ dynamic extraction of peanut oil using supercritical carbon dioxide.* Glassboro, NJ: Rowan University. pp. 1–8.

Menaker, A., Kravets, M., Koel, M. and Orav, A. (2004). Identification and characterizat-ion of super-critical fluid extracts from herbs. *Comptes Rendus Chimie*. 7, 629–633.

Mendez-Santiago, J. and Teja, A. S. (1999). The solubility of solids in supercritical fluids. *Fluid Phase Equilibria*. 158, 501–510.

Mendoza, G., Igartua, A., Fernandez, D., Urquiola, V. S. and Arguizoniz, R. (2011). Vegetable oils as hydraulic fluids for agricultural application. *Grasas Y Aceites*. 62, 29-38.

Metherel, A. H., Taha, A. Y., Izadi, H. and Stark, K. D. (2009). The application of ultrasound energy to increase lipid extraction throughput of solid matrix samples (flaxseed)*. Prostaglandins Leukotrienes and Essential fatty acids.* 81, 417-423.

Mezzomo, N., Mileo, B. R., Friedrich, M. T., Martínez, J. and Ferreira, S. R. S. (2010). Supercritical fluid extraction of peach (Prunus persica) almond oil: Process yield and extract composition. *Bioresource Technology*. 101, 5622–5632.

Miao, J., Che, K., Xi, R., He, L., Chen, X., Guan, X., Zhuang, X., Wen, X. and Cao, Y. (2-013). Characterization and *benzopyrene* content analysis of *camellia* seed oil extracted by a novel subcritical fluid extraction. *Journal of American Oil Chemist's Society*. 90, 1503–1508.

Min, D. B. and Boff, J. M. (2002). *Lipid oxidation of edible oil. In: Food Lipids:*. Akoh, C.C., and Min, D.B., Eds., Marcel Dekkered., ed. N. Chemistry, and Biotechnology. New York.

Mitra, S. and Wilson, N. K. (1991). An empirical-method to predict solubility in supercritical fluids. *Journal of Chromatography Science*. 29, 305 – 309.

Miyashita, K. and Takagi, T. (1986). Study on the oxidative rate and prooxidant activity of free fatty acids. *Journal of American Oil Chemists' Society*. 63, 1380–1384.

Mohamed, R. S. and Mansoori, G. A. (2002). *The use of supercritical fluid extraction technology in food processing*. Food Technology Magazine. London, UK: The World Markets Research Centre.

Mohammedi, T., Razmi, A. and Sadrzadeh, M. (2004). Effect of operating parameters on pb2+ separation from wastewater using elecrodialysis. *Desalination*. 167, 379-385.

Molero, A., G´omez, E. and Martinez de la Ossa. (2002). Quality of borage seed oil exatrcted by liquid and supercritical carbon dioxide. Chemical Engineering Journal. 88, 103–109.

Mónica, R., García-Risco, E. J. H., Vicente, G., Fornari, T., Señoráns, F. J. and Reglero, G. (2011). Kinetic study of pilot-scale supercritical CO_2 extraction of rosemary (Rosmarinus officinalis) leaves. *Journal of Supercritical Fluids.* 55, 971-976.

Montgomery, D. C. (1997). *Design and analysis of experiments*. (4^{th} ed.) New York: John Wiley and Sons.

Montgomery, D. C. (1999). Experimental design for product and process design and development. *The Statistician*. 48(2), 159-177.

Muralidhar, R. V., Chirumamila, R. R., Marchant, R., & Nigam, P. (2001). A response surface approach for the comparison of lipase production by *Candida cylindracea* using two different carbon sources. *Biochemical Engineering Journal*. 9, 17-23.

Murga, R., Sanz, M. T., Beltran, S. and Cabezas, J .L. (2005). Solubility of three hydroxycinnamic acids in supercritical carbon dioxide. *Journal of supercritical fluids*. 35, 106-113.

Murga, R., Sanz, M.T., Beltran, S. and Cabezas, J. L. (2003). Solubility of some phenolic compounds contained in grape seeds in supercritical carbon dioxide. *Journal of supercritical fluids*. 23, 113-121.

Mutlu, H. and Meier, A.R.M. (2010). Castor oil as a renewable resource for the chemical industry. European Journal of Lipid Science Technology. 112, 10–30.

Myers, R. H. and Montgomery, D. C. (1995). *Response surface methodology: Process and product optimization using designed experiments* (1st ed). New York: Wiley, Inc.

Nahar, L. and Sarkar, S. D. (2005). *Supercritical fluid extraction*. In: Sarka SD, Zahid L, Gray AI, editors. Method of biotechnology, 20. National products isolation, 2nd ed. Totowa, NJ: Humana Press. pp. 44–77.

Nguyen, H. N., Gaspillo, P. D., Maridable, J. B., Malaluan, R. M. Hinode, H., Salim, C. a-nd Ha Huynh, H. K. P. (2011). Extraction of oil from Moringa oleifera kernels using supercritical carbon dioxide with ethanol for pretreatment: Optimization of the extraction process. *Chemical Engineering and Processing*. 50, 1207– 1213

Nsa, E. E. and Ukachukwu, S. N. (2007). Determination of the true metabolizable energy of raw and thermal treated castor bean (Ricinus communis) using finisher broilers. *Annual Conference of the Nigerian Society forAnimal Production, Calarbar.* 18-22 March,. Calaber. 287-289.

Nurhan, T. D., Jerry, W. K. and Gary, R. (2003). *Supercritical Fluid Extraction in Food Engineering.*: Marcel Dekker, Inc.

Oderinde, R.A., Ajayi, I.A. and Adewuyi, A. (2009). Characterization of seed and seeds oil of *Hura Crepitans* and the kinetics of degradation of the oil during heating. Electronic *Journal of Environmental, Agricultural and Food Chemistry*. 8, 201-208.

Official Methods of Analysis (AOAC). (1999). (16^{th} ed.), AOAC International, Gaithersburg.

Ogunniyi, D. S. (2006). Castor oil: a vital industrial raw material. *Bioresource Technology*. 97, 1086-1091.

Ollanketo, M., Peltoketo, A.,Hartonen, K., Hiltunen, R. and Riekkola, H. (2002). Extraction of sage (Salvia officinalis L.) by pressurized hot water and conventional methods: Antioxidant activity of the extracts. *European Food Research Technology*. 215, 158-163.

Orav, A., Koel, M., Kailas, T. and Müürisepp, M. (2010). Comparative analysis of the co-mposition of essential oils and supercritical carbon dioxide extracts from the berries and needles of Estonian juniper (*Juniperus communis* L.). *Procedia Chemistry*. 2, 161–167.

Orio, L., Alexandru, L., Cravotto, G., Mantegna, S. and Barge, A. (2012). SFE-CO_2 and cl-assical methods for the extraction of Mitragyna speciosa leaves. *Ultrasonics Sonochemistry.* 19, 591-595.

Ozkal, S. G., Yener, M. E., Salgin, U. and Mehmetoglu, U. (2005a). Response surfaces of hazelnut oil yield in supercritical carbon dioxide. *European Food Research and Technology*. 220, 74 -78.

Ozkal, S.G. (2004). Supercritical carbon dioxide extraction of apricot kernel oil. PhD thes-is. *Middle East Technical University*.

Ozkal, S. G., Salgin, U. and Yener, M. E. (2005b). Supercritical carbon dioxide extraction of hazelnut oil. Journal of Food Engineering. 69, 217- 223.

Özkal, S. G., Yener, M. E. and Baymdirh, L. (2005). Mass transfer modeling of apricot kernel oil extraction with supercritical carbon dioxide. *Journal of Supercritical Fluids.* 35, 119-127.

Palazoğlu, T. K. and Balaban, M. E. (1998). Supercritical CO_2 extraction of roasted pistac-chio nuts. *Transactions of the American Society of Agricultural Engineers*. 41, 679-684.

Papamichail, I., Louli, V. and Magoulas, K. (2000). Supercritical fluid extraction of celery seed oil. *Journal of Supercritical Fluids*. 18, 213–226.

Passey, C. A. and Patil, N. D. (1994). *Process for preparing low calorie nuts*. U. S Patent 5290578.

Perakis, C., Louli, V., Voutsas, E. and Magoulas, K. (2010). Supercritical CO_2 Extraction of Dittany Oil: Experiments and Modelling. *Journal Supercritical Fluids*. 55, 573-578.

Perretti, G., Miniati, E., Montanari, L. and Fantozzi, P. (2003). Improving the value of rice by products by SFE. *Journal of Supercritical Fluids*. 26, 63-71.

Perrut, M. (2004). *Supercritical fluid extraction/fractionation: Industrial development and economics issues*. In state of the Art Book on supercritical fluids; Ainia: Spain,: pp. 171-178.

Porto,C. Da, Voinovich, Decortia, D., Natolino, A. (2012). Response surface optimization of hemp seed (Cannabis sativa L.) oil yield and oxidation stability by supercritical carbon dioxide extraction. *Journal of Supercritical Fluids*. 68, 45– 51.

Prieto, G., Okumoto, M., Takashima, K., Katsura, S., Mizuno, A., Prieto, O. and Gay, C.R (2003a). Non-thermal plasma reactors for the production of light hydrocarbon olefins from heavy oil. *Braziillian Journal of Chemical Engineering.* 20(1).

Prieto, G., Prieto, O., Gay, C. R. and Yamamoto, T. (2003b). Destruction of residual fumigant using a plasma reactor. *IEEE Transaction on Industry Applications*. 39(1), 72-78.

Puri, M., Sharma, D. and Barrow, C. J. (2012). Enzyme-assisted extraction of bioactives from plants. *Trends Biotechnology*. 30, 37–44.

Raisi, S. (2009). Developing New Processes and Optimizing Performance Using Response Surface Methodology. *World Academy of Science, Engineering and Technology*. 25, 1039-1042.

Rahman, N.A., Hassan, M.N, Anuar, B.O., Ibrahim, M.H. and Omar, A.K. (2001). The effect of dehulling on supercritical extraction of palm kernel oil. *Journal of Chemical Engineering of Japan*. 34(3), 407-410.

Rama Rao, S.V. (2004). *Vegetable protein supplements in poultry diets*. htt:// www. poul-vet.com/index.php.

Rebolleda, S., Rubio, N., Beltrán, S., Teresa Sanz, M., González-Sanjosé, M. L. (2012). Supercritical fluid extraction of corn germ oil: Study of the influence of process parameters on the extraction yield and oil quality. *Journal of Supercritical Fluids.* 72, 270– 277.

Regueira, T., Carvalho, P. J., Oliveira, M. B., Lugo, L., Coutinho, J. A. P. and Fernandez, J. (2013). Experimental measurements and modeling of CO_2 solubility in sunflower, castor and rapeseed oils. *Journal of Supercritical Fluids*. 82, 191– 199.

Reverchon, E. (1997). Supercritical fluid extraction and fractionation of essential oils and related products. *Journal of Supercritical Fluids*. 10, 1-37.

Reverchon, E. and Marco, I. (2006). Supercritical fluid extraction and fractionation of natural matter. *Journal of Supercritical Fluids*. 38, 146–166.

Reverchon, E., Poletto, M., Osseo, L.S. and Somma, M. (2000). Hexane elimination from soybean oil by continuous packed tower processing with supercritical CO_2. *Journal of American Oil Chemist's Society*. 77, 9–14.

Rezzoug, S. A., Boutekedjiret, C. and Allaf, K. (2005). Optimization of operating conditions of *rosemary* essential oil extraction by a fast controlled pressure drop process using response surface methodology. *Journal of Food Engineering*. 71, 9-17.

Rial-Otero, R., Gaspara, E. M., Moura, I. and Capeloa, J. L. (2007). Gas chromatography mass spectrometry determination of acaricides from honey after a new fast ultrasonic-based solid phase micro-extraction sample treatment. *Talanta*. 71, 1906–1914.

Rohman, A. and Che Man, Y. B. (2010). Fourier transform infrared (FTIR) spectroscopy for analysis of extra *virgin olive* oil adulterated with palm oil. *Food Research International*. 43, 886–892.

Rostami, M., Farzaneh, V, Boujmehrania, A., Mohammadi, M. and Bakhshabadi, H. (201-4). Optimizing the extraction process of sesame seed's oil using responsesurface method on the industrial scale. *Industrial Crops and Products*. 58, 160–165.

Rozzi, N. I., Singn, R. K., Vierling, R. A. and Watkins, B. A. (2002). Supercritical fluid extraction of lycopene from tomato processing by-product. *Journal of Agricultural and Food Chemistry*. 50, 2638-2643.

Ruhan, A. and Motonobu, G. (2009). Mitsuru, S. *Supercritical fluid extraction in food an-alysis*. In: Otles,S., editors. Handbook of food analysis instruments. Boca Raton, FL: CRC Press, Taylor and Francis. pp. 25–54.

Ruttarattanamongkol, K., and Petrasch, A. (2015). Oxidative susceptibility and thermal pr-operties of Moringa Oleifera seed oil obtained by Pilot-Scale subcritical and supercritical carbon dioxide extraction. *Journal of food process Engineering*. DOI: 10.1111/jfpe.12213.

Sabinus, O. O. E. (2012). Physico-chemical properties of oil from some selected under util-ized oil seeds available for biodiesel preparation. *African Journal of Biotechnology*. 11(42), 10003-10007.

Safaralie, A., Fatemi, S. and Sefidkon, F. (2008). Essential oil composition of *Valeriana officinalis* L. roots cultivated in Iran Comparative analysis between supercritical CO_2 extraction and hydrodistillation. *Journal of Chromatograp-hy A*. 1180, 159–164.

Safaralie, A., Fatemi, S. and Salimi, A. (2010). Experimental design on supercritical extra-ction of essential oil from valerian roots and study of optimal conditions. *Food Bioproduct Process*. 88, 312–318.

Sahena, F., Zaidul, I. S. M, Jinap, S., Karim, A. A, Abbas, K. A, Norulaini, N. A. N. and Omar, A. K. M. (2009). Application of supercritical CO_2 in lipid extraction, A review. *Journal of Food Engineering*. 95:240–253.

Sajfrtová, M., Ličková, I., Wimmerová, M., Sovová, H. and Zdeněk, W. (2010). Supcritica-cal carbon dioxide extraction from sea buckthorn (Hippophae rhamnoides L.) seeds. International. *Journal Molecular Science*. 11, 1842-1850.

Salgin, U., Doker, O. and Calimli, A. (2006). Extraction of sunflower oil with supercritical CO_2: Experiments and modeling. *Journal of Supercritical Fluids.* 38: 326-331.

Salgın, U. and Korkmaz, H. (2011). A green separation process for recovery of healthy oil from pumpkin seed. *Journal of Supercritical Fluids*. 58, 239– 248.

Salgın, U. and Salgın, S. (2013). Effect of main process parameters on extraction of pine kernel lipid using supercritical green solvents: Solubility models and lipid profiles. *Journal of Supercritical Fluids*. 73, 18– 27.

Salimon, J., Dina, A. M. N., Nazrizawati, A. T., Firdausi, M. Y. M. and Noraishah, A. (2-010). Fatty acid composition and physicochemical properties of Malaysian castor beans Ricinus communis L. seed oil. *Sains Malaysiana*. 39, 761–4.

Sánchez-Vicente, Y., Cabañas, A., Renuncio, J. A. R. and Pando, C. (2009). Supercritical fluid extraction of peach (Prunus persica) seed oil using carbon dioxide and ethanol. *Journal of Supercritical Fluids*. 49, 167-173.

Sandra, B. G., Dusan, R. M., Marko, D. S., Irena, T. Z., Ruzica, M. A. and Dejan, U. S. (2007). Supercritical carbon dioxide extraction of carrot fruit essential oil: Chemical composition and antimicrobial activity. *Food Chemistry*. 105, 346-352.

Sawsan, S. R., Ibrahim, A. H., Majid, A. S. A., Abdul Majid, A.M.S. and Mohd Omar, A. K. (2013). Comparison of yields and quality of nutmeg butter obtained by extraction of nutmeg rind by soxhlet and supercritical carbon dioxide (SC-CO_2). *Journal of Food Engineering*. 119, 595-601.

Scarpa, A. and Guerci, A. (1982). Various uses of castor oil Plant (Ricinus Communist L)- a review. *Journal of Ethnopharmacology*. 5, 117-137.

Scarpellini, A., Cerretani, L., Bendini, A. and Toschi, T. G. (2005). Effetto dell'acidità libera sulla stabilità ossidativa di un olio neutralizzato. *Industrie Alimentari*. 44, 22–25.

Schmitt, W. J. and Reid, R. C. (1986). Solubility of monofunctional organic solids in chemically diverse supercritical fluids. *Journal of Chemical Engineering Data*. 31, 204 – 212.

Scholz, V. and Silva, J.N.D. (2008). Prospect and risks of the use of castor oil as a fuel. *Biomas and Bioenergy*. 32, 95-100.

Selva, P., Susana, B. B. and Esteban, A. B. (2008). *Fundamentals of supercritical fluid technology*. In: Mertinez JL, editor. Supercritical fluid extraction of nutraceuticals and bioactive compounds. Boca Raton, FL: CRC Press, Taylor & Francis Group; pp. 1–49.

Shahipurullah, A. (2013). *Investigation of extracting oil from waste palm fruit fibre*. Bachelor Degree, Universiti Teknikal Malaysia.

Shehu, M.S. Abdul Manan, Z. and Wan Alwi, S. R. (2012). Optimization of thermo-alkaline disintegration of sewage sludge for enhanced biogas yield. *Bioresource Technology*. 114, 69-74.

Shen, J. and Shao, X. (2005). A comparison of accelerated solvent extraction, Soxhlet extraction, and ultrasonic-assisted extraction for analysis of terpenoids and sterols in tobacco. *Analytical and Bioanalytical Chemistry*. 383, 1003–1008.

Shi, J., Yi, C., Xue, S. J., Jiang, Y., Ma, Y. and Li, D. (2009). Effects of modifiers on the profile of lycopene extracted from tomato skins by supercritical CO_2. *Journal of Food Engineering*. 93, 431–436.

Shi, J., Yi, C., Ye, X., Xue, S., Jiang, Y., Ma, Y. and Liu, D. (2010). Effects of supercritical CO_2 fluid parameters on chemical composition and yield of arotenoids extracted from pumpkin. *Food Science and Technology*. 43, 39–44.

Shridhar, B. S., Beena, K.V., Anita, M. V. and Paramjeet, K. B. (2010). Optimization and charaterization of castor seed oil. *Leonardo Journal of Sciences*. 17:59–70.

Shirsath, S., Sonawane, S. and Gogate, P. (2012). Intensification of extraction of natural products using ultrasonic irradiations – a review of current status. *Chemical Engineering Processing: Process Intensification*. 53, 10–13.

Silva, D. C. M. N., Bresciani, L. F. V., Dalagnol, R. L., Danielski, L., Yunes, R. A. and Ferreira, S. R. S. (2009). Supercritical fluid extraction of carqueja (*Baccharis trimera*) oil: process parameters and composition profiles. *Food and Bioproducts Processing*. 87, 317–326.

Silverstein, R. M., Bassler, G. C. and Morrill, T. C. (1981). *Spectrometric Identification of Organic Compounds*, 4th ed. John Wiley and Sons, New York.

Skerget, M., Knez, Z. and Habulin, M. (1995). Solubility of β -carotene and oleic acid in dense CO_2 and data correlation by a density based model. *Fluid Phase Equilibria*. 109, 131 – 138.

Skerget, M., Knez, Z. and Knez-Hrncic, M. (2011). Solubility of solids in sub and supercritical fluids: a review. *Journal of Chemical Engineering Data*. 56, 694 – 719.

Skold, M., Borje, A., Harambasic, E. and Karlberg, A. T. (2004). Contact allergens formed on air exposure of linalool. Identification and quantification of primary and secondary oxidation products and the effect on skin sensitization. Chemical Research Toxicology. 17, 1697–705.

Soares, B. M. C., Gamarra, F. M. C., Paviani, L. C., Goncalves, L. A. G., Cabral, F.A. (2007). Solubility of triacylglycerols in supercritical carbon dioxide. *Journal of Supercritical Fluids*. 43, 25–31.

Sodeifian, G., Azizi, J. and Ghoreishi, S. M. (2014). Response surface optimization of Smyrnium cordifolium Boiss (SCB) oil extraction via supercritical carbon dioxide. *Journal of Supercritical Fluids*. 95, 1–7.

Sorin, T. (2012). Castor oil induces laxation concentration via ricinoleic acid activatingprostag landin Ep3 receptors,. *Institute of Pharmacology, J. W. Euethe university Frankfurth, Germany.* 1-6.

Sousa, E. M. B. D., Chiavone-Filho, O., Moreno, M. T.,D. Silva, D. N., Marques, M. O.M. and Meireles, M. A. A. (2002). Experimental results for the extraction of essential oil from *Lippia sidoides* cham using pressurised carbon dioxide. *Brazilian Journal of Chemical Engineering*. 19, 229-241.

Sovova, H., Stateva, R. P. and Galushko, A. A. (2001). Solubility of b-carotene in supercritical CO_2 and the effect of entrainers. *Journal of Supercritical Fluids*. 21 (3), 195-203.

Sovova, H., Stateva, R. P. and Galushko, A. A. (2001). Essential oils from seeds: solubility of limonene in supercritical CO_2 and how it is affected by fatty oil. *Journal of Supercritical Fluids*. 20, 113–129.

Sovova, H., Zarevu´cka, M., Vacek, M. and Stra´nsky, K. (2001). Solubility of two vegetable oils in supercritical CO_2. *Journal of Supercritical Fluids*. 20, 15–28.

Sparks, D.L., Hernandez, R. and Estévez, L.A. (2008). Evaluation of density-based models for the solubility of solids in supercritical carbon dioxide and formulation of a new model. *Chemical Engineering Science*. 63, 4292–4301.

Stahl, K., Quirin, W. and Gerald, G. (1988). *Dense gases for extraction and refining*. Springer-Verlag. New York.

Stevigny, C., Rolle, L., Valentini, N. and Zeppa, G. (2007). Optimization of extraction of phenolic content from hazelnut shell using response surface methodology. *Journal of the Science and Food Agriculture*. 87, 2817-2822.

Stroescu, M., Stoica-Guzun, A., Ghergu, S., Chira, N. and Jipa, I. (2013). Optimization of fatty acids extraction from Portulaca oleracea seed using response surface methodology. *Industrial Crops and Products*. 43, 405– 411.

Tabernero, A., Martín del Valle, E.M. and Galán, M.A. (2011). On the use of semi empirical models of (solid + supercritical fluid) systems to determine solid sublimation properties. *Journal of Chemical Thermodynamics*. 43, 711–718.

Tan, C. P. and Che Man, Y. B. (2002). Recent Developments in Differential Scanning Calorimetry for Assessing Oxidative Deterioration of Vegetable oils. *Trends in Food Science and technology*. 13(9-10): 312-318.

Tan, C. P., Che Man, Y. B., Selamat, J. and Yusoff, M. S. A. (2002). Comparative Studies of oxidation Stability of Edible Oils by Differential Scanning Calorimetry and Oxidative Stability Index Methods. *Food Chemistry.* 76, 385-389.

Tariq, M., Ali, S., Ahmad, F., Ahmad, M., Zafar, M., Khalid, N. and Khan, M.A. (2011). Identification, FT-IR, NMR (1H and13C) and GC/MS studies of fatty acid methyl *estersin* biodiesel from *rocket seed* oil. *Fuel Processing Technology.* 92, 336–341.

Temelli, F. (2009). Perspectives on supercritical fluid processing of fats and oils. *Journal of Supercritical Fluids*. 47: 583–590.

Terada, A., Kitajima, N., Machmudah, S., Tanaka, M., Sasaki, M. and Goto, M. (2010). C-old- pressed yuzu oil fractionation using countercurrent supercritical CO_2 e xtraction column. *Seperation and Purification Technology*. 71, 107–113.

Tonthubthimthong, P., Chuaprasert, S., Douglas, P. and Luewisutthichat, W. (2001). Supercritical CO_2 extraction of nimbin from neem seeds an experimental study. *Journal of Food Engineering*. 47, 289–293.

Tonthubthimthong, P., Douglas. P., Douglas, S., Luewisutthichat, W., Teppaitoon, W. and Pengsopa, L. (2004). Extraction of nimbin fron neem seeds using supercritical CO_2 and a supercritical CO_2-menthanol mixture. *Journal of supercritical Fluids*. 30, 287-301.

Trinh, T.K. and Kang, L.S. (2011). Response surface methodological approach to optimiz-e the coagulation–flocculation process in drinking water treatment. *Chemical engineering research and design.* 89, 1126-1135.

Turek, C., Kirschmann, N. and Stintzing, F. C. (2012). Quality monitoring of selected ess-ential oils upon storage at different temperature regimes. z.*Arznei Gewurzpfia*. 17, 73-79.

Turek, C. , Stintzing F. C. (2013). Stability of Essential Oils: A Review. *Comprehensive Reviews in Food Science and Food Safety*. 12, 40-53.

Turner, C., Whitehand, L. C., Nguyen, T. and Mckeon, T., (2004). Optimization of a supe-rcritical fluid extraction/reaction methodology for the analysis of castor oil using experimental design. *Journal of Agricultural and Food Chemistry*. 26(*52*), 26−32.

Vargas, C. E., Mendes, M. F., Azevedo, D. A., Pessoa, F. L. P. and Uller, A. C. (2010). E-xtraction of the essential oil of abajeru (*Chrysobalanus icaco*) using supercritical CO_2. *Journal of Supercritical Fluids*. 54, 171–177.

Vaughn Katherine, L. S., Clausen Edgar, C., King Jerry, W., Howard Luke, R. and Julie, C. D. (2008). Extraction conditions affecting supercritical fluid extraction (SFE) of lycopene from watermelon. *Bioresource Technology*. 99, 7835-7841.

Vealsco, J. and Dobarganes, C. (2002). Oxidative stability of virgin olive oil. *European Journal of lipid Science Technology*. 104, 661-676.

Vlachos, N., Skopelitis, Y., Psaroudaki, M., Konstantinidou, V., Chatzilazarou, A. and Tegou, E. (2006). Applications of Fourier transform-infrared spectroscopy to edible oils. *Analytica Chimica Acta*. 573-574, 459–465.

Veggi, P. C., Cavalcanti, R. N., Angela, M. and Meireles, A. (2011). Modifier effects on s-upercritical fluid extraction (SFE) of some Brazilian plants: antioxidant activity and economical evaluation. *Procedia Food Science*. 1, 1717-1724.

Wabner, D., Geier, K. and Hauck, D. (2006). For a deeper understanding of tea tree oil: fresh is best—why we should only use fresh oil at any concentration. *Internation Journal of Aromatherapy*. 76, 109–15.

Wade, L. G. Jr. (1987). *Organic chemistry*. Prentice-Hall. NJ: Englewood Cliffs.

Wagner, H., Luther, R. and Mang, T. (2001). Lubricant base fluids based on renewable raw materials: Their catalytic manufacture and modification. *Applied Catalysis A: General*. 221, 429-442.

Wahyu, B. S., Nik Norulaini, N.A.R., Mohd Azizi, C.Y. and Mohd Omar, A. K. (2007). Medium-chain Triglycerides Rich Oil Extraction using Supercritical Carbon Dioxide. *International Conference on Chemical Sciences (ICCS-2007)*. 24-26. Yogyakarta, Indonesia, ANL/17-2.

Wahyu, B., S., Priyo, A., Didi, D., A., 2012. Solubility examination of palm kernel oil in supercritical CO_2 and its correlation with solvent density based model. *Proceeding of International Conference on Chemical and Material Engineering*. Semarang Indonesia, ISBN : 978-602-097-281-7.

Wang, H. C., Chen, C. R. and Chang, C. J. (2001). Carbon dioxide extraction of ginseng r-oot hair oil and ginsenosides. *Food Chemistry*. 72, 505–509.

Wang, L. and Weller, C.L. 2006. Recent advances in extraction of nutraceuticals from pla-nts.Trends Food Science and Technology. 17, 300–312.

Wang, X. and Xue, W.T. (2003). Research on wheat embryo oil extracted by supercritical CO_2. *Journal of China Agricutural University*. 8, 58–60.

Waynick, J. A. (2005). *Characterization of biodiesel oxidation and oxidation products*. Technical Literature Review, SwRI Project No. 08-10721. Department of Energy. National Renewable Energy Laboratory,: U.S.

Weiss, E. A. (2005). *Castor*. In Oilseed Crops, 2nd Edition, Blackwell Scientific Ltd, Oxf-ord. pp. 13-52.

Xing, H., Yang, Y., Su, B., Huang, M. and Ren, Q. (2003). Solubility of artemisinin in su-percritical carbon dioxide. *Journal of Chemical Engineering Data*. 48, 330–332.

Yamini, Y., Khajeh, M., Ghasemi, E., Mirza, M. and Javidnia, K. (2008). Comparison of essential oil compositions of *Salvia mirzayanii* obtained by supercritical carbon dioxide extraction and hydrodistillation methods. *Food Chemistry*. 108, 341–346.

Yin, J. Z., Wang, A. Q., Wei, W., Liu, Y. and Shi, W. H. (2005). Analysis of the operation conditions for supercritical fluid extraction of seed oil. *Seperation and Purification Technology*. 43, 163–167.

Yong, O.Y. and Salimon, J. (2006). Characteristics of *Elateriospermum tapos* seed oil as a new source of oilseed. Industrial Crops and Products. 24, 146–151.

Yu, Z. R., Rizvi, S. S. H. and Zollweg, J. A. J. (1992). Phase equilibrium of oleic acid, m-ethyl oleate, and anhydrous milk fat in supercritical CO_2. *Journal of Supercritical Fluids*. 5, 114.

Yun, S.L.J., Liong, K.K., Gurdial, G.S. and Foster, N.R. (1991). Solubility of cholesterol in supercritical carbon dioxide. *Industrial and Engineering Chemistry Research*. 30, 2476 – 2482.

Zaidul, I. S. M. (2003). Supercritical carbon dioxide as a process fractionation technology of palm kernel oil for cocoa butter quality fat. Ph. D. thesis, University Sains Malaysia, Penang Malaysia.

Zaidul, I., Nik Norulaini, N., Mohd Omar, A. and Smith, R. (2007a). Supercritical carbon dioxide (SC-CO_2) extraction of palm kernel oil from palm kernel. *Journal of Food Engineering*. 79, 1007–1014.

Zaidul, I. S. M., Norulaini, N. A. N., Mohd Omar, A. K., Sato, Y. and Smith, R. L. Jr (20-07b). Separation of palm kernel oil from palm kernel with supercritical carbon dioxide using pressure swing technique. *Journal of Food Engineering*. 81, 419–428.

Zarnowski, R. and Suzuki, Y. (2004). Expedient Soxhlet extraction of resorcinolic lipids from wheat grains. *Journal of Food Composition and Analysis*. 17, 649–664.

Zermane, A., Larkeche, O., Meniai, A. H, Crampon, C. and Badens, E. (2014). Optimizat-ion of essential oil supercritical extraction from Algerian Myrtus communis L. leaves using response surface methodology. *Journal. of Supercritical Fluids*. 85, 89– 94.

Zhang, S., Zu, Y. G., Fu, Y. J., Luo, M., Liu, W., Li, J. and Efferth, T. (2010). Supercritic-al carbon dioxide extraction of seed oil from yellow horn (*Xanthoceras sorbifolia* Bunge.) and its anti-oxidant activity. *Bioresource Technology*. 101, 2537–2544.

Zhang, Q., Tang, L., Zhang, J., Mao, Z. and Jiang, L. (2011). Optimization of thermal-dilute sulphuric acid pretreatment for enhancement of methane production from cassava residues. *Bioresource Technology*. 102, 3958-3965.

Zhao, S. and Zhang, D. (2013). A parametric study of supercritical carbon dioxide extraction of oil from *Moringa oleifera* seeds using a response surface methodology. *Seperation and Purification Technology*. 113, 9–17.

Zhao, S. and Zhang, D. (2014). Supercritical CO_2 extraction of Eucalyptus leaves oil and comparison with Soxhlet extraction and hydro-distillation methods. *Separation and Purification Technology*. 133, 443–451.

Zheng, H., Tshukudu, T. and Yang, J. (2012). Optimization of the Coagulation- flocculation Process for WastewaterTreatment Using Polymeric Ferric Sulfate (PFS) - Poly-diallyldimethyl Ammonium Chloride (PDADMAC) Composite Coagulant. *Hydrology current research*. 3(139), doi.org/10.4172/2157-7587.1000139.

Zhinmin, X. and Samuel, J. G. (2000). Comparison of supercritical fluid and solvent extraction methods in extracting *Oryzanol* from rice bran. *Journal of American Oil Chemist's*. 77, 547-551.

Zizovic, I., Stamenic, M. and Orlovic, A. (2007). Supercritical carbon dioxide extraction of essential oils from plants with secretory ducts: Mathematical modelling on the micro-scale. *Journal of Supercritical Fluids.* 39, 338-346.