# CERTIFICATION OF APPROVAL

## Modelling of Supercritical Carbon Dioxide Extraction of Oil from Oilseeds

By

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# CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own except as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources or persons.

**MUHAMMAD FARIS BIN ROSLAN**

# ABSTRACT

This study is aimed to learn the underlying behaviour of the Supercritical Carbon Dioxide ($CO_2$) Extraction on the oilseeds or any solid without chemical reaction. CFD model will be developed to better understand the mass transfer phenomenon and complex flow field around a single particle in porous medium.

Wide range of species has been explored such as wheat germ, oats, tomato, grape seed and soybean. Despite the relatively large number of species processed, only some models of the SFE of oil seed have been published.

CFD numerically solves the Navier-Stokes equations and the energy and species balances. Some important variables in extraction process such as solvents flow rate and solid particle length will be modeled. Volume of Fluid (VOF) method will be used to predict the mentioned fields in a 2D geometry.

This project focus is to model the the concentration profile of oil under Supercritical Carbon Dioxide Extraction condition via CFD. In the end, it will predict extract yields as a function of flow rate and particle diameter. Thus, determine the most optimum operating conditions and improve the efficiency of the extraction process. Experimental result referred is to validate the proposed model.

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All praises to Allah the Almighty who help me and gave me strength, inspiration and patience to finish this research. It is only with His help, blessings and guidance and only by His will that I finally managed to complete this project.

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## LIST OF SYMBOLS

| | |
|---|---|
| $a$ | specific surface of the solid (1/cm) |
| $C$ | solute concentration in the solvent in terms of mass of solute per unit mass of solvent (g/g) |
| $C_0$ | solute concentration in the solvent at the beginning of extraction (g/g) |
| $D_L$ | axial dispersion ($m^2$/s) |
| $d_p$ | particle diameter (cm) |
| $k_f$ | mass transfer coefficient from the free oil phase to the solvent (m/s) |
| $k_i$ | mass transfer coefficient from the tied oil phase to the solvent (m/s) |
| $K_p$ | seed oil equilibrium constant between the tied oil phase and the solvent (g/g) |
| $K$ | seed oil equilibrium constant between the c free oil phase and the solvent (g/$cm^3$) |
| $m_0$ | mass of seed charged (g) |
| $m_e$ | mass of extracted oil (g) |
| $m_s$ | mass of solvent used (g) |
| $P$ | solute concentration in the solid in terms of mass of solute per unit mass of nonsoluble solid (g/g) |
| $P_0$ | solute concentration in the solid at the beginning of extraction (g/g) |
| $t$ | extraction time (min) |
| $u$ | superficial velocity of the solvent (cm/min) |
| $Y$ | extraction yield ( - ) |
| $Y$ | asymptotic extractable oil yield ( - ) |
| $z$ | axial coordinate in the extractor (cm) |

Greek Letter

| | |
|---|---|
| $\varepsilon$ | voidage of the extraction bed ( - ) |
| $\phi_f$ | fraction of the particle volume filled by the free oil phase ( - ) |
| $\phi_t$ | fraction of the particle volume filled by the tied oil phase ( - ) |
| $\rho_e$ | density of the untreated particles (g/$cm^3$) |
| $\rho_f$ | solvent density (g/$cm^3$) |
| $\rho_o$ | vegetable oil density (g/$cm^3$) |
| $\rho_s$ | density of the non-soluble solid (g/$cm^3$) |

# CHAPTER 1

# INTRODUCTION

## 1.1 PROJECT BACKGROUND

Supercritical fluid extraction (SFE) processes have been lately successfully applied as a preferred alternative to conventional separation methods in the chemical, food, pharmaceutical, biochemical and environmental industries (Verónica Arancibia et al., 2004). The supercritical $CO_2$ extraction of essential or volatile oils from herbs and spices operates at low operating temperature, is free from contamination of products by the solvents.

The supercritical fluid $CO_2$ is commonly used to extract flavor from compounds, essential oil or unnecessary fats from food. This process is more environmentally friendly compared to other existing conventional processes. Plus, It is widely used in food industries since it is non-toxic (G. Brunner, 2005). Eventhough it is an expensive process, for the complex products that require high purity such as medicines, the costs can be considered as competitive compared to conventional ones such as distillation and solvent extraction which are less efficient.

Computational Fluid Dynamics (CFD) has been found to be a very useful tool in the understanding and optimization of such complex processes. CFD provides a means of visualizing your system and gives an enhanced understanding of the fluid process.

In this project, VOF method will be used to predict the concentration profile and liquid velocity field around a single sphere particle. This study will assist us to understand the behaviour of the Supercritical Carbon Dioxide ($CO_2$) Extraction on the oilseeds or any solid without chemical reaction.

## 1.2 PROBLEM STATEMENT

In the past few decades, The Supercritical Carbon Dioxide ($CO_2$) Extraction has been studied as a potential alternative to existing conventional extraction techniques. It is found that it is a very promising for improving the traditional techniques which are mostly based on solvent extraction.

Thus, SFE of oil seed has been thoroughly explored from the processing perspective. A wide range of species has been studied such as soybean, tomato, oats, corn germ, grape seed and so on. Most of the researches which had been done were to study the influence of temperature and pressure on process rate and yield, mass transfer and heat transfer.

However, despite the relatively large number of species processed, only a few models of the SFE of oil seed have been published. Furthermore, most of the researches are experimental based. Only a few researches which are conducted to study the behavior of SFE of seed oils by using Computational Fluid Dynamics (CFD).

Modeling Supercritical Carbon Dioxide Extraction on the oilseeds is very challenging since the pathway towards a more efficient extraction is not obvious yet due to the complexity and poorly understood kinetics involved in the extraction process (Zabaleta, G.A., 2007). Building prototypes is both time consuming and expensive. Thus, one of the best options left to study SFE of seed oils behavior is via CFD.

Generally, it operates on quite simple laws such as laws of thermodynamics, momentum, density different, and Euler equations. By using CFD, we can study the extraction behavior and produce many design approach with that, we can achieve best solution in a very economical and interactive ways.

## 1.3 OBJECTIVES

The main objective of this project is to simulate the concentration profile of oil under Supercritical Carbon Dioxide Extraction condition.

In order to attain the main objective, the following specific objectives will be done:

a) To investigate the effect of the process parameters, which are flow rate and solid particle length relative to yield.

## 1.4 SCOPE OF STUDY

This project is a study and research-based which emphasizes the modelling of the concentration profile of oil under Supercritical Carbon Dioxide Extraction condition. The concentration profile will be determined to understand the process.

The models produced will be validated against available experimental works which predicts extract yields as a function of flow rate, and particle diameter. Few parameter combinations will be studied to understand the extraction efficiency.

## 1.5 RELEVANCY OF THE PROJECT

Research on this area of study rarely involves CFD simulation to understand the process behavior. By conducting this project, the model produced can be referred to improve the efficiency of extraction process upon oilseeds or any solid under Supercritical $CO_2$ Extraction conditions.

# CHAPTER 2

# LITERATURE REVIEW

## 2.1 DEVELOPMENT OF SUPERCRITICAL $CO_2$ EXTRACTION

There is an increasing public awareness of the health, environment and safety hazards associated with the use of chemical solvents in the industry. Hannay and Horgarth (1879) found that gases could be good solvents under supercritical conditions. Slight changes in pressure continuously altered the density of these fluids from gas-like to liquid-like, thus allowing their solubility power to be adjusted over wide ranges.

A few years later, in 1896, a review of supercritical fluid solubility phenomena has been published by Villard. He described the strange of fluid when it changed its state from low pressure and temperature to high temperature and pressure including its density variation. At that time, he could not describe the supercritical state as specific as it is nowadays. However, it is clear enough to explain the concept of supercritical phenomena.

Then, E.H. Buchner reviewed several existing literature and made a significant improvement in supercritical fluid research. He carried out his studies over a wide temperature range, and he used observations of cloud points, freezing points, and the number of phases present for his solubility determinations. From here, he managed to explain very clearly the supercritical state. He proved the phenomena that gas changed to supercritical state and he gave some supercritical conditions (temperature and pressure) of some substances, such as $CO_2$.

Then, Supercritical Fluid technique has been introduced and applied into extraction process. It is considered as a powerful technique which offers a few advantages over conventional methods in term of low operating temperature, no contamination of product by usual solvents and lower physical plant area space requirements (J.L. Humphrey and G.E. Keller, 1997).

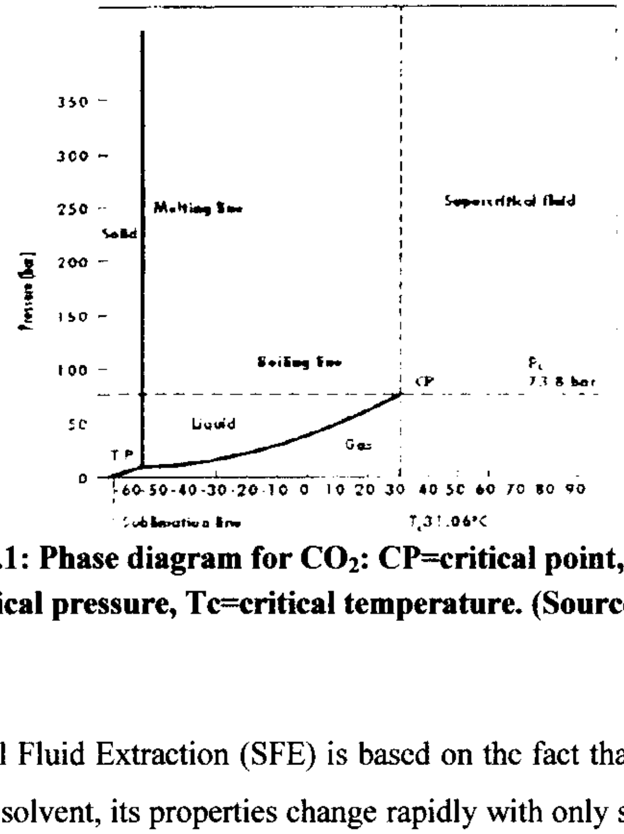

**Figure 2.1: Phase diagram for $CO_2$: CP=critical point, TP=triple point, Pc=critical pressure, Tc=critical temperature. (Source: Brogle, 1982)**

Supercritical Fluid Extraction (SFE) is based on the fact that, near the critical point of the solvent, its properties change rapidly with only slight variations of pressure.

## 2.2 APPLICATION OF SUPERCRITICAL EXTRACTION

### 2.2.1 Food Technology

Supercritical Fluid Extraction (SFE) of oil seeds has been studied by several authors from the processing point of view. One of the earliest and most important fields that SFE was applied is in food technology and essential oil extraction.

For example, the application of SFE in the decaffeination of coffee and tea is the first process that SFE has been used in food technology. Previously, organic solvents such as ethyl acetate or methylene chloride are used to extract and reduce the level of caffeine in coffee bean from approximately 1% w/w to 0.06%.

Potentially harmful effects of residual levels of these solvents have created an urge to find alternative safer solvents such as Supercritical $CO_2$. Eventhough the solubility of caffeine in SCF $CO_2$, is relatively low (< 0.2% w/w), Ebelling and Franck (1984) state that this predicament is offset by the high added value of the process.

Moreover, supercritical $CO_2$ is a very selective solvent for decaffeination which does not remove many of the desirable flavor-precursor components compared to organic solvents which make it a very interesting alternative.

### 2.2.2 Medicine

Other than that, one of the most important applications of SFE is in medicine production. SFE was used to extract vitamin from natural sources. It was also used for the process of drug extraction from plants or animals or in the encapsulation process (Y. Wang et al, 2002). Many papers were published in this field such as:

1. Extraction and isolation of avermectins and milbemycins from liver samples (Martin Danahera et al., 2001),
2. "Formulation of an effective mosquito – repellent topical product from Lemongrass Oil" (A.O.Oyedele et al., 2002),
3. Extraction of cholesterol from cattle brain (N. Vedaraman et al., 2004).

### 2.2.3 Cosmetics

SFE process can be applied in cosmetics industry as well. Especially in extracting fragrances and flavors to be used in cosmetics products. Most of the early work on the use of liquid CO2 for flavor extraction was carried out in the

Soviet Union in the 1960s and reported in Russia. There are many researches that have been carried out in this field such as extraction of Evening Primrose (J.W. King et al., 1997), or extraction of flavor and essential oil from orange peel (A. Berna et al., 2000).

### 2.2.4 Environmental Engineering

Pollution becomes more alarming nowadays. Therefore, many organizations have taken initiatives to curb this problem and avoid further environmental disaster. Many researches which applies SFE in environmental engineering have been listed as examples, such as:

1. Cleaning degreasing process (A. Marsal et al., 2000),
2. Purification of used frying oil (Jungro Yoon et al., 2000),
3. Removal of organophosphate pesticides from wastewater (Jya-Jyun Yu et al., 2002).

Supercritical Fluid can also be used as cleaning solvent for some special cases.

**Table 2.1: Summary of Supercritical $CO_2$ extraction process**

| References | Materials | Description | Conditions |
|---|---|---|---|
| Sovová et al., 1994 | Grape seed | Oil extraction, effect of solvent flow rate and flow direction of extraction | T = 40°C<br>P = 280 atm |
| Stastova et al., 1996 | Sea buckthorn berries | Oil extraction from seed and pulp, effect of extraction conditions on the solubility and mass transfer rate | T = 25 – 60°C<br>P = 96 - 270 atm |
| B. Mira et al., 1998 | Orange peel | Investigation effect of $CO_2$ feed rate, pressure, temperature and average length on composition and linalool. | T=293-232K<br>P=80-280 atm<br>G=0.5-3.5kg/h<br>S=0.1-10mm |

| | | | |
|---|---|---|---|
| Bravi et al., 2002 | Sunflower | High quality edible oil from sunflower seed optimized in continuous process model with 3, 4 and 5 extractors. | 40°C; 280 atm<br>110 – 220 atm<br>0.15 – 0.24 $m^3$/h |
| Giuseppe Vasapollo et al., 2004 | Tomato | Extraction of lycopene from tomato using supercritical carbon dioxide in the presence of vegetable oil as co-solvent. | T=45-700C<br>P=335-450 atm |
| Patel et al., 2006 | Cashew nut shells | Effect of temperature and pressure on yield. Significant effect of extraction time on chemical composition. | 303 – 333K<br>200 – 300 atm<br>30 – 180 min |

## 2.3 GENERAL PROCESS DESIGN

Essential oils are mainly formed by hydrocarbon and oxygenated terpenes and by hydrocarbon and oxygenated sesquiterpenes. Its high solubility in hexane make hexane extraction process from ground seeds a quite convenient method to produce essential oils. The process is very efficient, but its major problem is represented by hexane elimination after extraction. Three distillation units in series, operated under vacuum and other ancillary apparatuses (deodorizer, degumming, etc.), have to be used. To make it worse, the thermal degradation of the oil and the incomplete hexane elimination (from 500 to 1000 ppm residue) may possibly happen.

Therefore, several authors have proposed the replacement of the traditional process by Supercritical $CO_2$ extraction of oil from seeds (B. Marongiu et al, 2004, G. Vasapollo et al, 2004, E.U. Franck et al, 1984). Indeed, triglycerides forming seed oils are readily soluble in Supercritical $CO_2$ at 40 ◦C and at pressures larger than about 280 bar. The main parameters to be taken into account for this process are particle size, pressure and residence time. Small particles (1mm mean diameter or less) and high pressures (300–500 bar) can strongly reduce the extraction time.

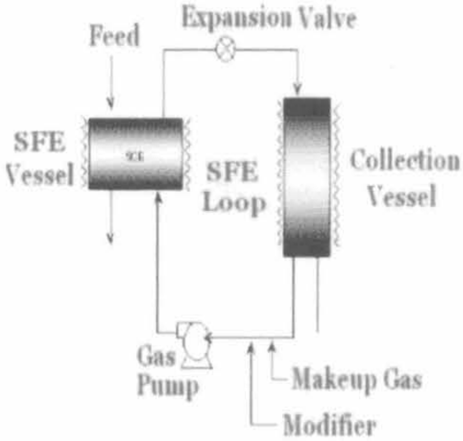

**Figure 2.2: General Process Setup**

After extraction, the SC-$CO_2$ tryglicerides solution is sent to a separator working at subcritical conditions. This operation reduces to near zero the solvent power of $CO_2$ and allows the recovery of oil. The complete elimination of gaseous $CO_2$ from oil is also obtained in the separator. The SFE of several seed oils has been successfully performed up to the pilot scale.

An alternative process has also been proposed, in which the extraction is performed at a fixed pressure and only temperature variations are used to reduce the oil solubility and obtain its recovery. The advantage of this scheme coupled to heat exchangers networking is in the reduction of energy consumption in the overall extraction process (E. Reverchon and L. Sesti Oss`eo, 1994).

## 2.4 MODEL DEVELOPMENT

### 2.4.1 Computational Fluid Dynamics

Computational Fluid Dynamics (CFD) is a method that is becoming more and more popular in the modeling of flow systems in many fields. It is the science of predicting fluid flow, heat transfer, mass transfer, chemical reactions, and related phenomena by solving the mathematical equations which govern these processes using algorithms and numerical methods.

By using CFD, we can build a computational model that represents a system or device that we wanted to study then apply the fluid flow physics and chemistry to this virtual prototype. The software will produce a prediction of the fluid dynamics and related physical phenomena (FLUENT Website, 2010).

The result of CFD analyses is relevant engineering data used in (André Bakker, 2002-2008):

a) Conceptual studies of new designs.

b) Detailed product development.

c) Troubleshooting.

d) Redesign.

The fundamental basis of almost all CFD problems is the Navier-Stokes equations, which define any single-phase fluid flow. These equations can be simplified by removing terms describing viscosity to yield the Euler equations. The most fundamental consideration in CFD is how one treats a continuous fluid in a discretized fashion on a computer.

### 2.4.2 Applications of CFD

Areas of research where CFD has taken an important role include the aerospace and automotive industries where CFD has become a relatively cheap alternative to wind tunnel testing.. The other applications of CFD are (André Bakker, 2002-2008):

a) Film coating, thermoforming in material processing applications.
b) Ventilation, heating, and cooling flows in buildings.
c) Flow and heat transfer in propulsion and power generation systems.

### 2.4.3 ANSYS FLUENT Software

ANSYS FLUENT (Figure 2) is a "Flow Modeling Software" owned by and distributed by ANSYS, Inc. It is used to model fluid flow within a defined geometry using the principles of computational fluid dynamics. It contains the broad physical modeling capabilities needed to model flow, turbulence, heat transfer, and reactions for wide range of industrial applications including turbulent flows, heat transfer, reacting flows, chemical mixing, combustion, and multiphase flows.

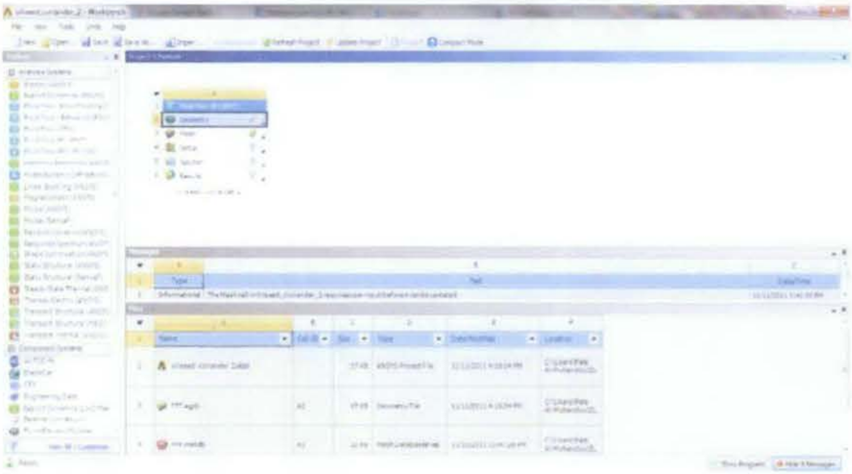

**Figure 2.3: ANSYS Workbench General User Interface**

Its advanced solver technology provides fast, accurate CFD results, flexible moving and deforming meshes, and superior parallel scalability. The interactive solver setup, solution and post-processing capabilities of ANSYS FLUENT make it easy to pause a calculation, examine results with integrated post-processing, change any setting, and then continue the calculation within a single application.

Previously, FLUENT is distributed along with GAMBIT, which is a tool to generate or import geometry so that it can be used as a basis for simulations run in FLUENT. The latest version of the software, FLUENT 12.0, integrates ANSYS Workbench in it. It provides users with an innovative project schematic view ties together the entire simulation process, guiding the user through even complex multiphysics analyses with drag-and-drop simplicity which simplify the work of the user.

# CHAPTER 3

# METHODOLOGY

## 3.1 RESEARCH METHODOLOGY

The methodology of this project as follows (FLUENT Software Training, 2001):

- Problem identification and Pre-Processing
    I. Modeling goals: To simulate the concentration profile and liquid velocity field around a single sphere particle.
    II. Domain of the model: 3D.
    III. Grid design: created using ANSYS Workbench
- Solver Execution
    IV. Setting up the numerical method: Volume of Fluids (VOF) multiphase model will be used.
    V. Computing and monitoring the solution.
- Post processing
    VI. Examining the results.
    VII. Plotting the results.
    VIII. Compare results with experimental results or existing literature values.

## 3.2 PROJECT ACTIVITIES AND PROGRESS

### 3.2.1 Model Analysis

The modeling of the extraction process is based on the following hypotheses (C. Marrone, M. Poletto, E. Reverchon and A. Stassi, 1998):

1. We suppose that the behavior of all compounds extracted is similar and can be described by a single pseudo-component with respect to the mass transfer phenomena.
2. Concentration gradients in the fluid phase develop at larger scales than the particle size.
3. The solvent flow rate, with superficial velocity u, is uniformly distributed in all the sections of the extractor.
4. The volume fraction of the fluid, $\varepsilon$, is not affected by the reduction of the solid mass during extraction. Further hypotheses regard the natural matrix.
5. The solute in the solid is present in two separate phases. One phase includes the solute contained inside the internal structure of the particles (the 'tied solute' phase). It fills a fraction $\phi_t$ of the overall volume occupied by the seed particles. This value does not change during the extraction process and, therefore, $\phi_t$ is considered constant. The average tied solute concentration is called P. The other phase is made of the solute freely available on the particle surface. The concentration here is always the same and, according to our hypotheses, it is equal to the pure solute density $p_o$.
6. The fraction of the seed volume filled by the free solute before extraction is $\phi_f=1-\phi_t$.
7. The fraction of the seed occupied by the free solute during the extraction is $\psi\ \phi_f$ where is $\psi \leq 1$.
8. A linear equilibrium relationship applies between phases.

According to the above hypotheses, the mass balance on the solute (seed oils) is:

$$\varepsilon \cdot \rho_f \cdot D_L \cdot \frac{\partial^2 C}{\partial z^2} + \varepsilon \cdot \rho_f \cdot \frac{\partial C}{\partial t} + \rho_f \cdot u \cdot \frac{\partial C}{\partial z}$$

$$+ (1-\varepsilon)\phi_t \cdot \rho_s \cdot \frac{\partial P}{\partial t} + (1-\varepsilon)\phi_f \cdot \rho_0 \cdot \frac{\partial \psi}{\partial t} = 0 \qquad (1)$$

The general mass balance on the phase of the free solute alone is:

$$\rho_s \frac{\partial \psi}{\partial t} = -\frac{k_1 a(\rho_s - K_\psi C)}{(1-\varepsilon)\phi_1} \quad \text{until } \psi > 0,$$

$$\text{otherwise} \frac{\partial \psi}{\partial t} = 0 \qquad (2)$$

The mass balance on tied solute is:

$$\frac{\partial P}{\partial t} = -\frac{k_1 \cdot a(P - K_p C)}{(1-\varepsilon)\phi_1} \qquad (3)$$

The set of differential Equations (1)–(3) was numerically integrated using a finite difference method. A Wendroff numerical cell was used. This method is characterized by an implicit computational cell that assures calculation stability whichever is the interval (of time and of space) used.

### 3.2.2 Preliminary Modeling Identification and Pre-processing

#### 3.2.2.1 Modelling Goals

The modeling goal is to study the behavior of supercritical $CO_2$ extraction process of seed oils. In this project, we will specifically simulate the concentration profile of oil under Supercritical Carbon Dioxide Extraction condition. The inlet flow is continuous. The solvent flow rate, with superficial velocity u, is uniformly distributed in all the sections of the extractor.

### 3.2.2.2 Model Assumptions

The model is based on the design of extraction basket provided by Marrone *et al* and Catchpole *et al*. The object is originally Cylindrical in shape and will be created on 2D Plane.

| Dimensions | Coriander Seed | Almond Seed |
|---|---|---|
| **Height (cm)** | 28 | 16 |
| **Diameter (cm)** | 11.6 | 4 |

**Table 3.1: Extraction Basket Dimensions**

Porosity of each model is calculated based on mean particle diameter using Carmen-Kozeny Equation:

$$\frac{dp}{dx} = -\frac{180\mu u(1-\varepsilon)^2}{d_s^2\varepsilon^3}$$

SEM Analysis shows that Coriander and Almond seeds particle are spherical. Thus, it increases the accuracy of the bed porosity calculation.

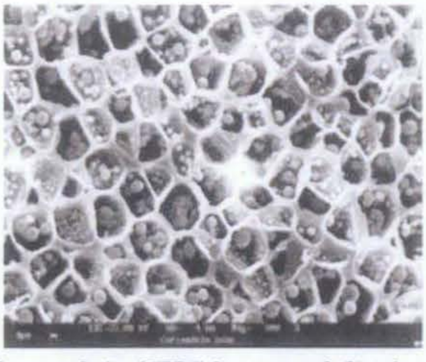

**Figure 3.1: SEM image of Coriander Seed particle (Catchpole et. al., 1996)**

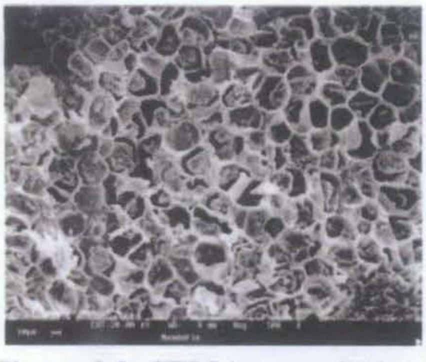

**Figure 3.2: SEM image of Almond Seed particle (Marrone et. al., 1998)**

#### 3.2.2.3 Grid Designs

The topology of the simulation model is established in the initial CAD geometry design phase. In this initial phase the major solid and fluid region interfaces are established. An appropriate choice of grid type depends on the geometric complexity, flow filed and the cell and element types supported by solver. (Gambit Software Tutorial Guide, 2001)

The quality of the grid is extremely important and can strongly influence the solution sometimes also determining whether or not a valid or a converged solution can be achieved at all. The most important qualification about a computational meshing is that it must define enough points to capture everything of interest that is happening in the computational domain without becoming so extensive and that unreasonable computation times are required.

#### 3.2.2.3 Solver Executions - CFD Simulation Parameters

The simulations were based upon standard "k-ε" turbulence and volume of fluid (VOF) models. To model the movement of the fluids (supercritical $CO_2$) inside the extractor a special User Defined Function (UDF) was developed. The UDF allowed us to evaluate the fluids flow in the separator and enable to study the separation behavior. (FLUENT Software Tutorial Guide, 2001)

Currently, no simulation has been done to show the result and description of this project. The new mesh for oilseeds is still under construction and design phase. Every mesh produces must be tested and evaluated properly before it can be simulated in FLUENT software.

### 3.2.3 Project Progress

*Generating a Simple 2-Dimensional Model of an extraction basket*

Step 1: Create a Fluid Flow Analysis System in ANSYS Workbench

1. In this project, the geometry is designed using ANSYS Workbench. Thus, ANSYS Workbench is started to open the application window. Toolbox can be seen on left and Project Schematic on the right.

**Figure 3.3: Selecting the Fluid Flow (FLUENT) Analysis System in ANSYS Workbench**

2. Fluid Flow (FLUENT) option in the toolbox is chosen as the fluid flow analysis system. Then, it is dragged and dropped into the project schematic.

Step 2: Create the Geometry in ANSYS DesignModeler.

1. ANSYS DesignModeler is started by double-clicking the Geometry cell in the fluid flow analysis system. Unit is set to millimeter.

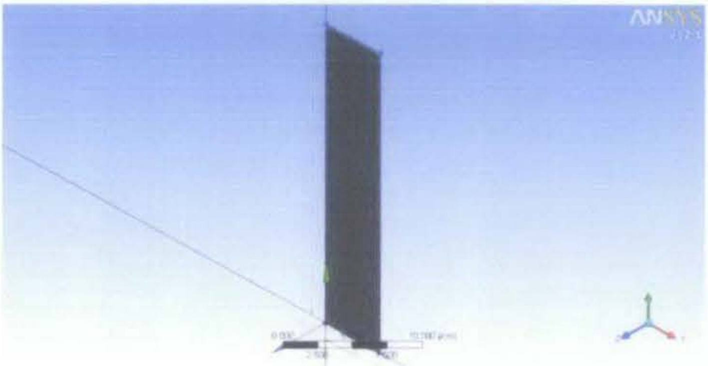

**Figure 3.4: Rectangle generated at origin**

2. Create a rectangle. Put the necessary parameters and click Generate.

Step 3: Meshing the Geometry in ANSYS Meshing

1. ANSYS Meshing is opened by double-clicking the Mesh cell.
2. To perform surface meshing, this geometry is edited in Meshing.
3. Basic meshing parameters is set and improved from time to time to ensure the mesh file created is relevant to the project and acceptable.

**Figure 3.5: Surface Mesh in Meshing**

Step 4: Setting Up the CFD Simulation in ANSYS FLUENT

1. Mesh checking.
2. Model setup.
3. Materials setup for CFD simulation.

| **Materials** | Supercritical $CO_2$ (Fluid) | Triglycerides (Fluid) | Wood (Solid) |
|---|---|---|---|
| **Density** | 880 kg/m$^3$ (Coriander)<br>934 kg/m$^3$ (Almond) | 918.8 kg/m$^3$ | Default |
| **Viscosity** | 8.8e-5 kg/m.s (Coriander)<br>1.23e-6 kg/m.s (Almond) | 0.04914kg/m.s | Default |

**Table 3.2: Material Properties**

4. Boundary conditions setup for CFD analysis.

| **Zone** | **Type** |
|---|---|
| Inlet | Velocity-inlet |
| Outlet | Pressure-Outlet |
| Interior-surface_body | Interior (porous zone) |
| Wall | Wall |
| Axis | Axis |

**Table 3.3: Boundary Conditions**

| **Simulations** | **1** | **2** | **3** | **4** |
|---|---|---|---|---|
| **Seeds** | Coriander | Coriander | Almond | Almond |
| **Temperature ($^{o}C$)** | 40 | 40 | 40 | 40 |
| **Pressure (MPa)** | 25 | 25 | 35 | 35 |
| **Particle Diameter (mm)** | 0.56 | 0.92 | 0.70 | 1.90 |
| **Flow Rate (kg/h)** | 5.16 | 5.16 | 1.43 | 1.43 |

| Simulations | 5 | 6 | 7 | 8 |
|---|---|---|---|---|
| Seeds | Coriander | Coriander | Almond | Almond |
| Temperature (°C) | 40 | 40 | 40 | 40 |
| Pressure (MPa) | 25 | 25 | 35 | 35 |
| Particle Diameter (mm) | 0.56 | 0.56 | 0.30 | 0.30 |
| Flow Rate (kg/h) | 5.16 | 12.78 | 0.72 | 1.43 |

**Table 3.4: Parameters Applied on the Model**

5. Solution parameters setup.
6. Solution calculation

Step 5: Displaying Results in ANSYS FLUENT and ANSYS CFD-Post

Step 6: Duplicating the ANSYS FLUENT-based Fluid Flow Analysis System

1. Duplicate the title cell in Project Schematic.
2. Alter the geometry and parameters to run another simulation.
3. Similar steps are used simulate the flow of supercritical CO2 in extraction vessel, with a extraction basket containing Coriander Seed and another containing Almond Seed.

Step 7: Validation of Simulation Results Against Experimental Data

All simulation results will be compared with experimental data from the work of Catchpole (J. Catchpole, B. Grey and M. Smallfield, 1996) for Coriander Seed and Marrone (C. Marrone, M. Poletto, E. Reverchon and A. Stassià, 1998)

### 3.3 Activities/Gantt Chart and Milestone

| No | Detail/Week | 1 | 2 | 3 | 4 | 5 | 6 | | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | Selection of Project Topic: Modelling of Supercritical Carbon Dioxide Extraction of Oils | | | | | | | Mid-Semester Break | | | | | | | | |
| 2 | Preliminary Research Work: Research on literatures related to the topic | | | | | | | | | | | | | | | |
| 3 | Submission of Extended Proposal to Supervisor | | | | | | ■ | | | | | | | | | |
| 4 | Proposal Defence | | | | | | | | | | | | | | | |
| 5 | Project work continues: Further investigation on the project and do modification if necessary | | | | | | | | | | | | | | | |
| 6 | Submission of Interim Draft Report to Supervisor | | | | | | | | | | | | | | ■ | |
| 7 | Submission of Final Report to Coordinator | | | | | | | | | | | | | | | ■ |

**Table 3.5: FYP1 Gantt chart and Key Milestone**

| No | Detail/Week | 1 | 2 | 3 | 4 | 5 | 6 |  | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | Geometry design/Pre-processiong |  |  |  |  |  |  | Mid-Semester Break |  |  |  |  |  |  |  |  |
| 2 | Solver execution |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| 3 | Submission of Progress Report |  |  |  |  |  |  |  |  | ■ |  |  |  |  |  |  |
| 4 | Post-Processing Activities |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| 5 | Pre-SEDEX |  |  |  |  |  |  |  |  |  |  |  | ■ |  |  |  |
| 6 | Submission of Draft Report |  |  |  |  |  |  |  |  |  |  |  |  | ■ |  |  |
| 7 | Submission of Final Draft Report (Softbound) |  |  |  |  |  |  |  |  |  |  |  |  |  | ■ |  |
| 8 | Submission of Technical Paper |  |  |  |  |  |  |  |  |  |  |  |  |  | ■ |  |
| 9 | Viva Presentation |  |  |  |  |  |  |  |  |  |  |  |  |  |  | ■ |
| 10 | Submission of Final Report (Hardbound) |  |  |  |  |  |  |  |  |  |  |  |  |  |  | ■ |

**Table 3.6: FYP2 Gantt chart and Key Milestone**

# CHAPTER 4

# RESULT AND DISCUSSION

## 4.1 EXPERIMENTAL VALIDATION

For validation purpose, experimental data set from (J. Catchpole, B. Grey and M. Smallfield, 1996) and (C. Marrone, M. Poletto, E. Reverchon and A. Stassi, 1998) are used.

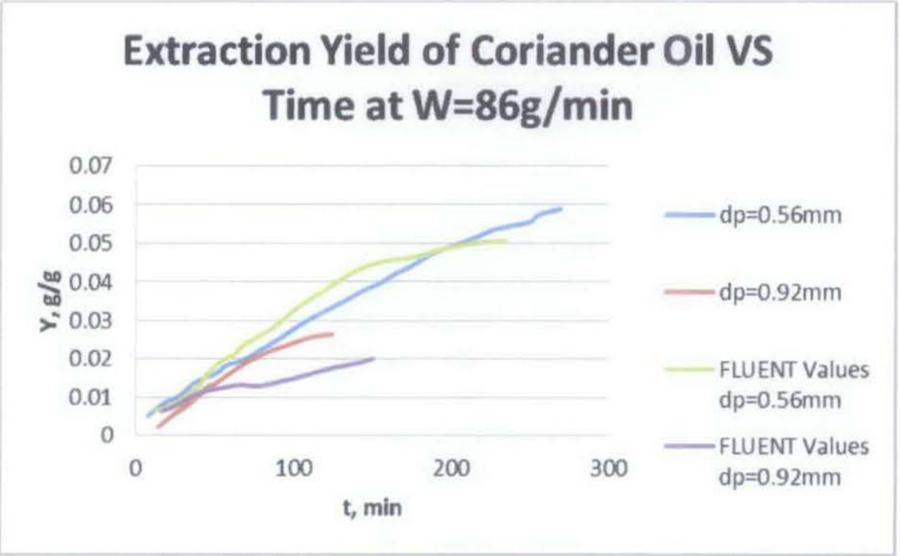

**Figure 4.1: Extraction Yield of Coriander Oil as a function of particle diameter with w=86 g/min**

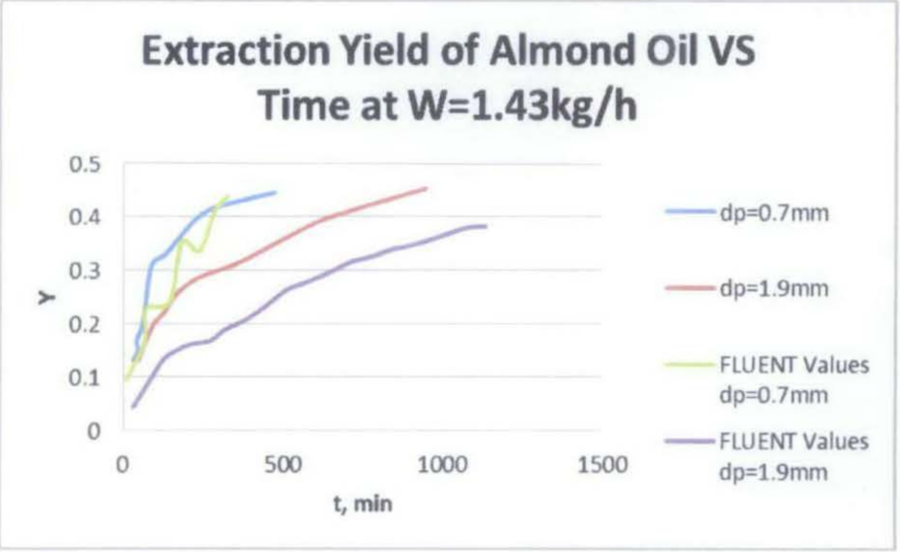

**Figure 4.2: Extraction Yield of Almond Oil as a function of particle diameter with w=1.43 kg/h**

Figure 4.1 and 4.2 shows the extraction yield of Coriander Oil and Almond Oil, Y, divided by the weight of the initial charges, a function of particle diameter at fixed extraction time. From the graph above, both oilseeds shows similar yield extraction pattern. The yield of extract for all oilseeds as a function of extraction time followed the form of an inverted exponential decay curve. These curves are typical of essential oil extraction, and similar to those obtained from the works of (B. Mira, M. Blasco, A. Berna, S. Subirats, 1999), (E. Reverchon and L. Sesti Oss'eo, 1994) and (H. Sovova', J. Kukeera and J. Jez, 1994).

Both figures prove that larger particle cause reduction of yield. It happens due to lower surface areas which reduce the contact between solvent and oil inside the pores. Thus, the most inner part of the solid is not efficiently reached by the solvent. On the other hand, the figure also shows that an increase of particle size increases the extraction time as it is expected from a corresponding decrease of the mass transfer rate. Indeed, in comparison the simulation results are in good agreement with experimental results.

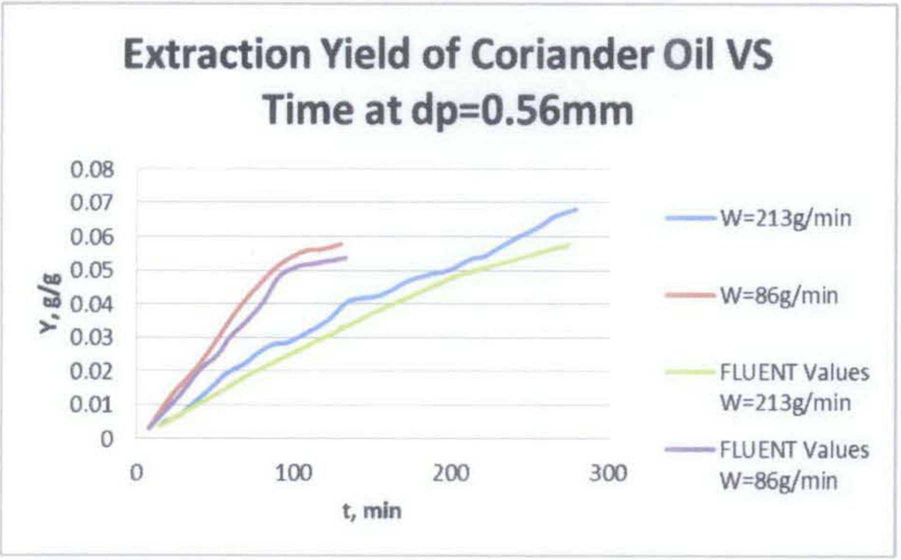

**Figure 4.3: Extraction Yield of Coriander Oil as a function of flow rate with dp=0.56 mm**

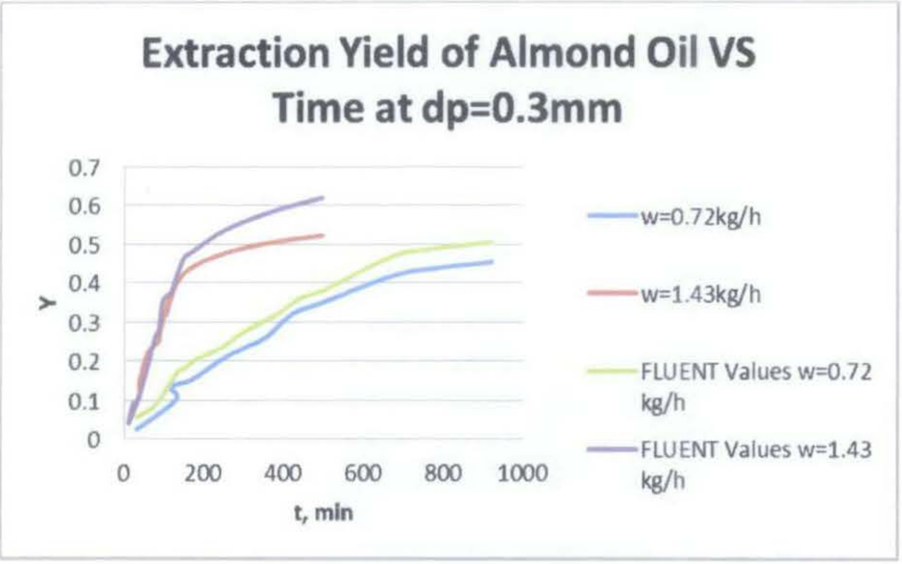

**Figure 4.4: Extraction Yield of Almond Oil as a function of flow rate with dp=0.3 mm**

Figure 4.3 and 4.4 also show similar pattern to previous two figures. Increasing the flow rate caused a reduction in the mass transfer rate. Thus, it implies that higher flow rate will increase the time required to approach completion of extraction. The simulation results also in good agreement with experimental results.

# CHAPTER 5

# CONCLUSION AND RECOMMENDATION

## 5.1 CONCLUSION

In this study, computational fluid dynamics was performed to predict the extract yields as a function of flow rate and particle diameter which are useful for the efficiency of Supercritical Fluid Extraction Process. Results show that for prediction of extract yields as a function of flow rate and particle diameter in the liquid-liquid systems via VOF method could be applied as an investigative tool.

It has been shown that complete extraction is almost impossible since the curve follows the inverted exponential decay pattern. It was found that the influence of particle size and solvent flow rate are significant on the efficiency of the supercritical fluid extraction process. Larger particle size as well as higher solvent flow rate could result in lower extraction rate. This finding seems to have a significant impact in the design of extractor. Thus, these investigations have merit as a scientific tool and will have practical applications in Supercritical Fluid Extraction Process.

## 5.2 RECOMMENDATIONS

For a better research work, this project should incorporate more important process parameters such as temperature, pressure, extraction time, purity of solvents and extractor size. The modeling should also consider other type of fat as part of the seed oil instead of Triglycerides alone. The final finding will be an accurate and consistent result. This project should vary the oilseeds species which have variety of cells shape instead of spherical alone. Thus, more conclusions can be made to improve this interesting process.

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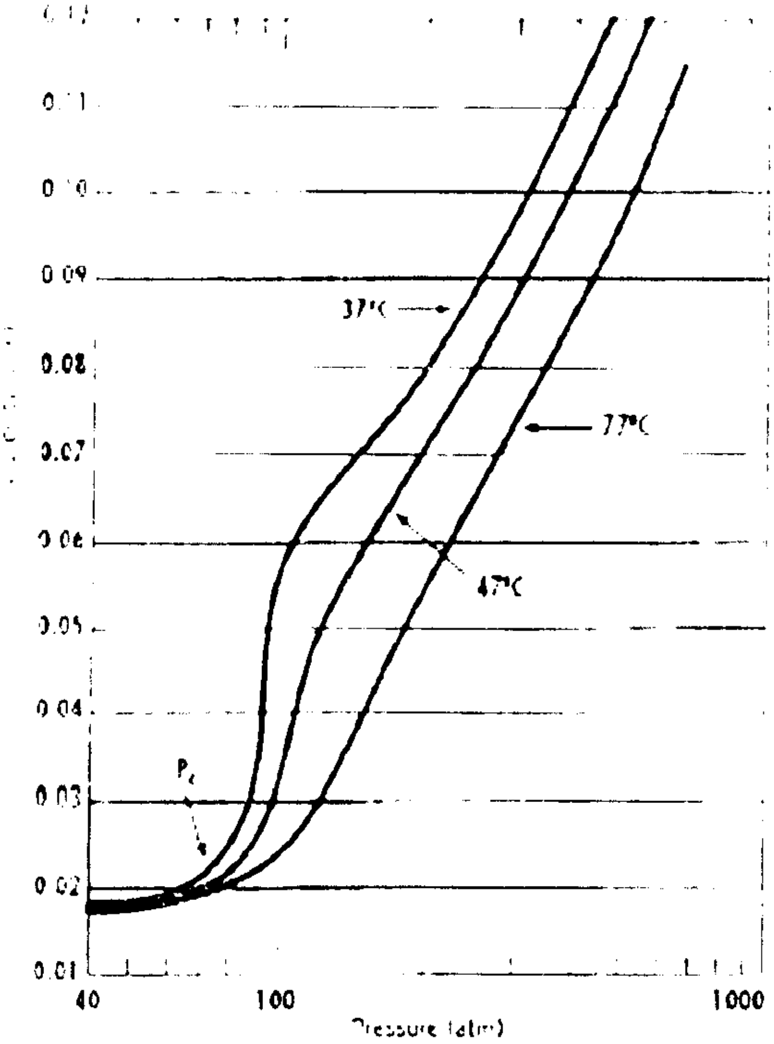
## Appendix A: Density-Temperature-Pressure Relationship for $CO_2$

| Density (g/mL) | 313 | 323 | 333 | 343 | 353 | 363 | 373 | 383 | 393 |
|---|---|---|---|---|---|---|---|---|---|
| 1.000 | 52.6 | 61.8 | n.a. | n.a. | n.a. | n.a. | n.a. | n.a. | n.a. |
| 0.95 | 36.3 | 46.3 | 54.4 | 64.4 | 68.0 | n.a | n.a. | n.a. | n.a. |
| 0.90 | 28.1 | 35.0 | 42.0 | 48.9 | 51.8 | n.a | n.a. | n.a. | n.a. |
| 0.85 | 21.1 | 26.9 | 32.9 | 40.1 | 44.7 | n.a | n.a. | n.a. | n.a. |
| 0.80 | 16.4 | 21.3 | 26.4 | 31.4 | 36.5 | 41.6 | 46.7 | n.a. | n.a. |
| 0.75 | 13.4 | 17.5 | 21.6 | 26.1 | 30.5 | 34.8 | 39.2 | 43.6 | 51.0 |
| 0.70 | 11.5 | 15.0 | 18.7 | 22.3 | 26.0 | 29.7 | 33.4 | 37.2 | 42.5 |
| 0.65 | 10.4 | 13.3 | 16.5 | 19.6 | 22.7 | 25.9 | 29.0 | 32.2 | 35.4 |
| 0.60 | 9.7 | 12.2 | 14.9 | 17.6 | 20.3 | 22.9 | 25.6 | 28.4 | 31.1 |
| 0.55 | 9.3 | 11.5 | 13.6 | 16.1 | 18.3 | 20.6 | 23.0 | 25.2 | 27.6 |
| 0.50 | 9.1 | 10.9 | 12.9 | 14.8 | 16.8 | 18.8 | 20.7 | 22.7 | 24.6 |
| 0.45 | 8.9 | 10.4 | 12.2 | 13.9 | 15.6 | 17.2 | 18.6 | 20.5 | 22.1 |
| 0.40 | 8.7 | 10.0 | 11.5 | 12.9 | 14.3 | 15.7 | 17.1 | 18.5 | 19.7 |
| 0.35 | 8.4 | 9.6 | 10.8 | 12.0 | 13.2 | 14.4 | 15.5 | 16.7 | 17.8 |
| 0.30 | 8.1 | 9.0 | 10.1 | 11.1 | 12.1 | 13.0 | 14.0 | 14.9 | 15.8 |
| 0.25 | 7.7 | 8.4 | 9.3 | 10.0 | 10.8 | 11.6 | 12.3 | 13.0 | 13.7 |
| 0.20 | 7.0 | 7.5 | 8.2 | 8.8 | 9.4 | 9.9 | 10.5 | 11.0 | 11.6 |

Source: Hewlett Packard Co. (Wilmington, DE USA)
n.a = data not available
pressure is given in MPa
temperature is given in K

Source: Taylor, 1996

**Appendix B: Viscosity behaviour of CO2 at various temperatures and pressures.**



Source: Taylor, 1996

# Appendix E: Experimental Conditions in the Literature for the SFE of Oil from Different Seeds

| | P (bar) T (°C) | $D_i$ (cm) | H (cm) | $m_s$ (g) | $d_p$ (mm) | W (g/min) |
|---|---|---|---|---|---|---|
| *Sunflower* | | | | | | |
| Test 1 | 280 (40) | 8.2 | 28 | 500 | 3 | 750 |
| Test 2 | 280 (40) | 8.2 | 28 | 500 | 3 | 4166 |
| Test 3 | 280 (40) | 8.2 | 28 | 500 | 3 | 1666 |
| Test 4 | 280 (40) | 8.2 | 28 | 500 | 3 | 83.3 |
| *Coriander* | | | | | | |
| Test 1 | 250 (40) | 10 | 27 | 1050 | 0.56 | 86 |
| Test 2 | 250 (40) | 10 | 27 | 1050 | 0.42 | 86 |
| Test 3 | 250 (40) | 10 | 27 | 1050 | 0.56 | 213 |
| Test 4 | 250 (40) | 10 | 27 | 1050 | 0.42 | 184 |
| *Grape* | | | | | | |
| Test 1 | 280 (40) | 0.8 | 12.2 | 4.5 | 0.36 | 0.9 |
| Test 2 | 280 (40) | 0.8 | 15.5 | 0.57 | 0.36 | 1.7 |
| Test 3 | 280 (40) | 0.8 | 14.5 | 5.32 | 0.81 | 1.7 |
| Test 4 | 280 (40) | 0.8 | 14.6 | 5.38 | 0.78 | 1.7 |
| Test 5 | 280 (40) | 0.8 | 15.2 | 5.58 | 0.45 | 1.7 |
| *Tomato* | | | | | | |
| Test 1 | 280 (40) | 0.75 | 5 | 1.26 | 0.25 | 1.5 |
| Test 2 | 280 (40) | 0.75 | 5 | 1.26 | 0.25 | 3.18 |
| Test 3 | 280 (40) | 0.75 | 5 | 1.26 | 0.25 | 5.88 |
| Test4 | 280 (40) | 0.75 | 5 | 1.26 | 1.02 | 3.42 |
| Test5 | 280 (40) | 0.75 | 5 | 1.26 | 0.65 | 3.42 |
| Test 6 | 280 (40) | 0.75 | 5 | 1.26 | 0.46 | 3.42 |
| Test 7 | 280 (40) | 0.75 | 5 | 1.26 | 0.25 | 3.42 |
| *Peanut* | | | | | | |
| Test 1 | 550 (25) | 5 | 38 | 500 | 1.1 | 72 |
| Test 2 | 550 (25) | 5 | 38 | 500 | 1.4 | 72 |
| Test 3 | 550 (25) | 5 | 38 | 500 | 2 | 72 |
| Test 4 | 550 (25) | 5 | 38 | 500 | 2.8 | 72 |
| Test 5 | 550 (25) | 5 | 38 | 500 | 4 | 72 |
| *Almond* | | | | | | |
| Test 1 | 350 (50) | 4 | 16 | 160 | 0.30 | 12 |
| Test 2 | 350 (50) | 4 | 11.5 | 72 | 0.70 | 23.3 |
| Test 3 | 350 (50) | 4 | 11.5 | 84 | 0.30 | 23.3 |
| *Fennel*[1] | | | | | | |
| Test 1 | 200 (40) | 5.5 | 16.5 | 281 | 0.372 | 8.3 |
| Test 2 | 200 (40) | 5.5 | 16.5 | 304 | 0.372 | 16.6 |
| Test 3 | 200 (40) | 5.5 | 16.5 | 275 | 0.372 | 24.9 |

Source: E. Reverchon and C. Marrone, 2001