

## Effect of Supercritical Carbon Dioxide Condition on Oil Yield and Solubility of *Pithecellobium Jiringan* (Jack) Prain Seeds

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### Article history

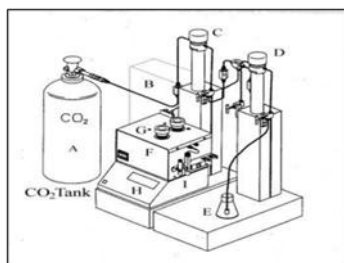
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### Graphical abstract



### Abstract

*Pithecellobium Jiringan* (Jack) Prain (*P.Jiringan*) is one of the medicinal plant sources that consist of antioxidant compounds. The aim of this study was to investigate the effects of supercritical carbon dioxide (SC-CO<sub>2</sub>) extraction parameters on oil yields and to measure the solubility of *P.Jiringan* oil in SC-CO<sub>2</sub> and correlated the data with Chrastil and del Valle & Aguilera models. The experimental were conducted at temperatures ranging from 40 °C to 70 °C and pressure from 27.58 MPa to 44.82 MPa. The extraction duration was 60 minutes. The result shows that the oil yields increase as the pressure and temperature increase. The solubility of oil increased as the density of CO<sub>2</sub> increased at constant temperature. An analysis of the results show that the solubility experimental data correlated with the Chrastil's model was successfully matched. In addition, SC-CO<sub>2</sub> give the highest oil yields compared to accelerated solvent extraction (ASE) and soxhlet extraction process.

**Keywords:** Chrastil's model; *Pithecellobium Jiringan* (Jack) Prain; solubility; supercritical fluid extraction

### Abstrak

*Pithecellobium Jiringan* (Jack) Prain (*P.Jiringan*) merupakan salah satu daripada sumber tumbuhan-tumbuhan yang mengandungi komponen antioksidan. Tujuan kajian ini dijalankan adalah untuk mengkaji kesan-kesan parameter kaedah penyarian bendalir lampau genting dengan menggunakan karbon dioksida sebagai pelarut (SC-CO<sub>2</sub>) ke atas hasil minyak dan untuk mengukur kebolehlarutan minyak *P.Jiringan* didalam (SC-CO<sub>2</sub>) dan mengkorelasi data kebolehlarutan minyak *P.Jiringan* dengan model Chrastil dan del Valle & Aguilera. Kajian ini telah dijalankan pada suhu antara 40 °C sehingga 70 °C dan pada tekanan antara 27.58 MPa sehingga 44.82 MPa. Tempoh pengekstrakan adalah 60 minit. Penemuan hasil kajian ini menunjukkan bahawa hasil minyak meningkat dengan peningkatan tekanan dan peningkatan suhu. Pada keadaan suhu malar, kebolehlarutan minyak meningkat dengan peningkatan kepadatan CO<sub>2</sub>. Analisis keputusan menunjukkan bahawa data keterlarutan minyak *P.Jiringan* telah berjaya dipadankan dengan model Chrastil. Di samping itu, SC-CO<sub>2</sub> memberikan hasil minyak tertinggi berbanding dengan ASE dan proses pengekstrakan Soxhlet.

**Kata kunci:** Kebolehlarutan; Model Chrastil; penyarian bendalir lampau genting; *Pithecellobium Jiringan* (Jack) Prain

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### 1.0 INTRODUCTION

Supercritical carbon dioxide extraction (SC-CO<sub>2</sub>) is the most commonly use for supercritical fluid extraction (SFE) application because of its moderate critical properties that include: 1) critical temperature at 31.1 °C and 2) critical pressure at 7.38 MPa. SFE is known to be dependent on the density of the fluid that in turn can be manipulated through control of the system pressure and temperature. The dissolving power of a SFE increases with isothermal increase in density or an isopycnic (i.e. constant

density) increase in temperature. In practical terms this means a SFE can be used to extract a solute from a feed matrix as in conventional liquid extraction. However, unlike conventional extraction, once the conditions are returned to ambient the quantity of residual solvent in the extracted material is negligible.

The basic principle of SFE is that the solubility of a given compound (solute) in a solvent varies with both temperature and pressure. At ambient conditions (25 °C and 1 bar) the solubility of a solute in a gas is usually related directly to the vapor pressure of the solute and is generally negligible. Moreover, one of the main

advantages of supercritical fluids is the ability to modify their selectivity by varying the pressure and temperature (modify fluid density). Therefore, supercritical fluids are often used to extract selectively or separate specific compounds from a mixture. On the other hand, accelerated solvent extraction (ASE) and soxhlet extraction were used to make a comparison with SC-CO<sub>2</sub> on the global oil yield. The comparison was study to determine the best process to extract oil from *P.Jiringan* seeds.

Malaysia has about 12,000 species of plants of which about 1,300 are said to be medicinal and only a small number of these plants have been fully investigated for their potential [1]. One of them is *Pithecellobium Jiringan* (Jack) Prain (*P.Jiringan*). *P.Jiringan* jack is a local delicacy in Indonesia, Malaysia, southern Thailand, and Myanmar. The beans are commercially available in markets during most of the year and are consumed raw, roasted, or fried. The djenkol bean has a broad, round shape and reddish color. The beans grow in large, dark purple pods (3-9 beans/pods) on the djenkol tree, which grows to 25 meters (82 ft) in height. These beans resemble a large, flattened horse chestnut, and the crushed bean emits a faint sulfurous odor. *P.Jiringan* is not normally considered to be a medicinal plant as it contains undesired compounds such as volatile oil and toxic acid namely djenkolic acid [2]. Even though the *P.Jiringan* seeds contain undesired compounds, it is also rich in essential compounds including protein, carbohydrates, fiber, calcium, phosphorus, vitamins and sterols which make it possible to be developed into medicinal drugs especially for anti-diabetics and anti-hypertension [3].

Solubility measurement can be obtained both experimental and modeling. It is important to obtain the solubility of solutes in the SC-CO<sub>2</sub> extraction in order to determine the optimal operating conditions. Most models can be classified as being either semi-empirical or equation-of-state based. However, semi-empirical models are often utilized because their relative ease of application equated to equations of state. The most common semi-empirical models are established upon providing a correlation between solubility and density; hence, they are referred to as density-based models. The most common semi-empirical methods are those developed by Chrastil [4]. This study, focus more on correlating the solubility of some compounds in SC-CO<sub>2</sub> using the Chrastil's model and del Valle and Aguilera model [5]. The aim of this study was to investigate the effects of extraction conditions namely, pressure and temperature especially on the solubility of *P.Jiringan* oil in SC-CO<sub>2</sub>, besides extraction yield. Furthermore, the correlation of experimental solubility data correlated with Chrastil and del Valle and Aguilera models were in this study.

## 2.0 MATERIALS AND METHODS

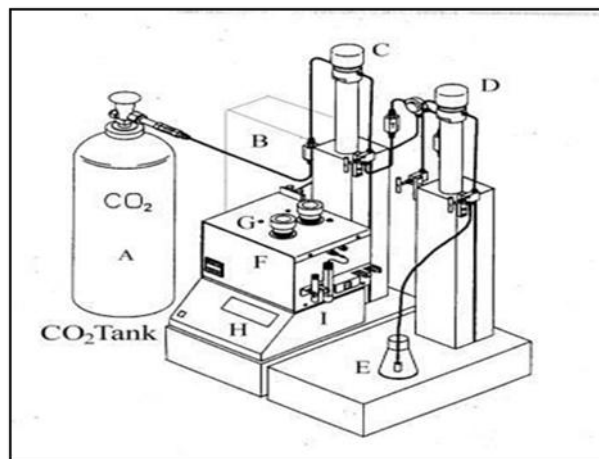
### 2.1 Sample Preparation

Matured and fresh *P.Jiringan* seeds were used and obtained from a local market at Taman Universiti, Skudai, Malaysia. Seeds was separated from fruits and thoroughly washed with tap water followed by distilled water. Next, the seeds were cut into small pieces (2-3 cm diameter and 1mm thickness) and were sun-dry conventionally all day long. *P.Jiringan* seeds were finely ground achieved powder form. The particle size was determined by sieving and was fixed at 215  $\mu\text{m}$ . 215  $\mu\text{m}$  was the best particle size to obtain the maximum yield [2].

### 2.2 Supercritical Carbon Dioxide System

Supercritical fluid extraction was performed using SFX TM 220 extraction system (ISCO, Lincoln, NE, US) consisting of high

pressure syringe pump (Model: 100DX), extractor chamber, 2.5 mL stainless steel extraction cell and extractor controller (Model: SFX 200) was used in this study. Others part involved were a chiller (Model: Yih Der BL-730). CO<sub>2</sub> with 99.99% purity (MOX Gases Sdn. Bhd. Selangor) was used as a solvent throughout the process. The equipment set up of the extraction process SFX TM 220 is shown in Figure 1.



**Figure 1** The equipment set up of the extraction process SFX TM 220. A. CO<sub>2</sub> tank; B. Chiller; C. Solvent pump; D. Modifier pump; E. Modifier reservoir; F. Extraction chamber; G. Sample cartridge; H. Controller panel; I. Analyte receiver

2.0 g of ground dried *P.Jiringan* seeds that have reached room temperature were loaded into the extraction cell and tightly sealed. Next, the extraction cell was placed in the extraction chamber and let the system to reach the desired condition. The extraction started after reached the desired condition. Volume of CO<sub>2</sub> used and extracted oil was recorded every 10 minutes of extraction process. Extracts are finally separated from the CO<sub>2</sub> phase and collected in collector at ambient temperature and atmospheric pressure. The CO<sub>2</sub> gas was depressurized in order to remove from the separator. The variable parameters involved in this process are temperature and pressure. The interest responses were oil yield and solubility. On the other hand, the constant parameters were mass of sample, extraction time, particle size and flow rate of CO<sub>2</sub>. The ranges of parameters were presented in Table 1 meanwhile Table 2 presents the constant parameter used.

**Table 1** Range of process parameter applied in extraction process

Process Parameter	Unit	Range	Increment ( $\Delta$ )	Responses
Temperature, T	$^{\circ}\text{C}$	$40.00 \leq T \leq 70.00$	$\Delta T = 10.00$	1. Oil yield 2. Solubility
Pressure, P	MPa	$27.58 \leq P \leq 44.82$	$\Delta P = 3.45$	

**Table 2** Constant parameter applied in extraction process

Constant Parameter	Unit	Value
Particle size	$\mu\text{m}$	215
Mass of Sample	g	2.0
Flow rate	mL/min	2.2
Extraction time	min	60

### 2.3 Soxhlet Extraction

Soxhlet extractions were performed using 15 g weighed of powdered *P.Jiringan* seeds and were placed in a thimble holder. Soxhlet extraction was carried out using 300 mL of solvents (Hexane and Water). The heating power was set slightly above the boiling point of the solvent used. The arrangement is such that vapors of the solvent are generated from the round-bottomed flask, pass through the thimble and get condensed in the condenser. The condensed solvent comes in contact with the *P.Jiringan* seeds in the thimble, where extraction occurs. When the liquid reaches the overflow level in the thimble, the liquid moves through the siphon back into the round-bottomed flask, carrying extracted solutes into the bulk liquid. The solvent was removed by rotary evaporator at temperature slightly above the boiling point of the solvent.

### 2.4 Accelerated Solvent Extraction (ASE)

10 g of grounded *P. Jiringan* seeds were extracted using ASE 150 system (Dionex, Sunnyvale, CA, USA) with hexane as a solvent. Extractions were performed at three different parameters which are temperatures (70, 90 and 110 °C), extraction times (5, 10 and 15 minutes) and cycles (1, 2 and 3). Sample mixed homogeneously with 5 g weight of diatomaceous earth was loaded into 34 mL stainless-steel extraction cells and filled with solvent up to a pressure of 1500 psi. The extraction cell was placed into the carousel. Next, initial heat-up time was applied and a static extraction with all system valves closed was performed. In order to overcome any extract carry-over, the cell was rinsed (with 60% cell volume using extraction solvent) and by using N<sub>2</sub> gas, solvent was purged from the cell. Solvent used were removed by rotary vacuum evaporator (RVE) at 70°C.

### 2.5 Oil Yield Measurement

Extracted oil can be defined as the weight difference between the sample before and after extraction at given temperature and pressure at 60 minutes of time extraction. The expression of extracted oil as follows:

$$W_{oil} = W_{t,i} - W_{t,f} \quad (1)$$

Where:

$$\begin{aligned} W_{oil} &= \text{weight of extracted oil, mg} \\ W_{t,i} &= \text{weight of vial + weight of sample, g} \\ W_{t,f} &= \text{weight of vial, g} \end{aligned}$$

### 2.6 Solubility of *P.Jiringan* oil in SC-CO<sub>2</sub>

In the experimental work, the solubility data of *P.Jiringan* oil in supercritical CO<sub>2</sub> for temperatures ranges from 40 °C to 70 °C and pressures ranges from 27.58 MPa to 44.82 MPa were measured by plotting experimental data of mass of oil extracted against the mass of CO<sub>2</sub> used (overall extraction curves). The curves were developed at 2.2 mL/min of CO<sub>2</sub> flow rate. Therefore, the solubility (y\*) was the slope of the constant extraction rate period of the overall extraction curves. Chrastil and Del Vale & Aguilera models were selected to evaluate the use of empirical model to correlate solubility data of *P.Jiringan* oil in supercritical CO<sub>2</sub>. These models are described to be appropriate for the range of temperature and pressure from which the experimental values were obtained.

A multilinear regression was performed by using Solver in Excel 2007 program to determine the model constants. The accuracy of Chrastil models and del Valle and Aguilera model were quantified by analysis of average absolute relative deviation

percentage (AARD). Equation 2 represents the method in calculating AARD%.

$$\text{AARD \%} = \frac{1}{n} \sum_{i=1}^n \left| \frac{y_{i,\text{exp}} - y_{i,\text{calc}}}{y_{i,\text{exp}}} \right| \times 100 \quad (2)$$

Where n = number of data  
 Y<sub>i,exp</sub> = solubility data obtained from experimental respectively at i<sup>th</sup> condition  
 Y<sub>i,calc</sub> = solubility data obtained from Chrastil model respectively at i<sup>th</sup> condition

#### 2.6.1 Chrastil Model

The Chrastil's model studies the formation of a solvation complex between the solvent and the solute molecules in the equilibrium, establishing a linear relation between solubility, solvent density and process temperature. The equation that represents the solubility model derived by Chrastil showed as follows:

$$Y^* = \rho^k \exp\left(\frac{a}{T} + b\right) \quad (3)$$

Where y\* = the solute solubility  
 T = temperature (K)  
 ρ = solvent density (g/L)

Meanwhile, for coefficient of a and b represents:

$$\begin{aligned} a &= \frac{\Delta H}{RT} \\ b &= \ln(M_A + kM_B) + q - k \ln M_B \end{aligned}$$

Where; ΔH = ΔH<sub>solv</sub> + ΔH<sub>vap</sub> and q = q<sub>s</sub> + q<sub>v</sub>  
 M<sub>A</sub> = molecular weights of the solute  
 M<sub>B</sub> = molecular weights of the gas

Also, a, b and k represent the adjustable constants of the model. The constant k indicates the number of CO<sub>2</sub> molecules present in the complex solute-solvent. The a and b parameters are vaporizing enthalpy and molecular weight dependants, respectively.

#### 2.6.2 Del Valle & Aguilera's Model

The del Valle and Aguilera model is a modification of the Chrastil model. The equation of del Valle and Aguilera model can be expressed as follows:

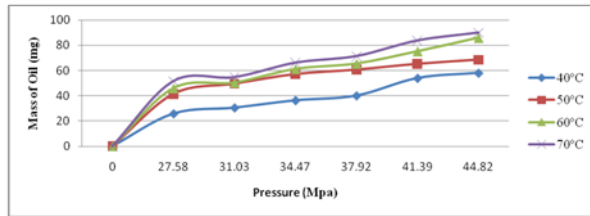
$$\ln Y^* = k \ln \rho + \frac{a}{T} + \frac{b}{T^2} + c \quad (4)$$

## 3.0 RESULTS AND DISCUSSION

### 3.1 Effect of Pressure and Temperature on the Overall Extraction Oil Yield

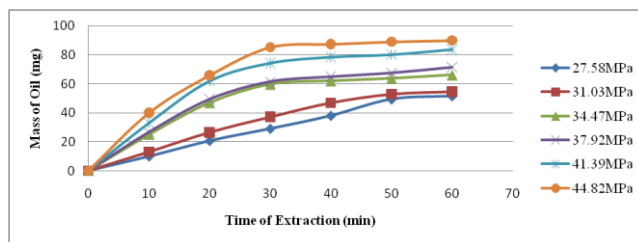
Figure 2 presents the effect of pressure and temperature on extraction yield of *P.Jiringan* in SC-CO<sub>2</sub> at different pressure for the ranges from 27.58 MPa to 44.82 MPa and different temperature at the ranges from 40 °C to 70 °C. It can be seen that the oil yield increased with the increase of the pressure. This

phenomenon can be explained by the basic principle of supercritical fluid extraction process. According to De Castro [6] at constant temperature, the density of CO<sub>2</sub> increased with the increase in pressure. Hence, the interaction between intermolecular solvent and solute which is known as solvating power increased. In this condition, the extraction rate would increase with the increase in the solubility of oil in the supercritical carbon dioxide extraction due to the increase in solvating power. In addition, vapor pressure would increase at high temperature and it did facilitate the transportation of oil onto the seeds surfaces.



**Figure 2** The effect of pressure on extraction yield (mg) at constant temperature (°C)

The highest extracted oil yield obtained (89.89 mg) at the highest pressure and of 44.82 MPa and highest temperature of 70 °C. Meanwhile, the lowest oil yield obtained (25.87 mg) at the lowest pressure of 27.58 MPa and lowest temperature of 40 °C. Figure 3 shows the effect of different pressure at highest temperature of the process (70 °C). It shows that the curve becomes uniform with the extracted oil yield of 73.09 mg, in percentage of 3.65% after 30 minutes of extraction time. From this point, the oil extracted reached the asymptotic yield at the end of the extraction process after 60 minutes of extraction time. Asymptotic yield represents the maximum amount of oil obtained at the maximum condition of extraction [7]. The asymptotic yield obtained was 4.49% (89.89 mg of oil per 2.0 g sample used). Furthermore, by reaching at an asymptotic value, it can be assumed that the extraction was complete where the oil from the seeds was considered completely removed.



**Figure 3** Effect of pressure on extracted *P.Jiringan* oil at constant 70°C as a function of extraction time

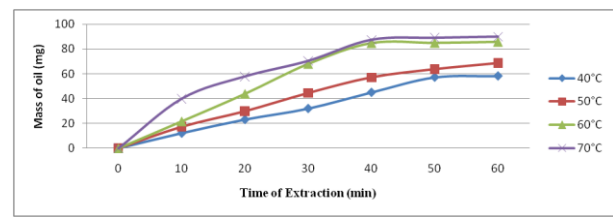
It was observed that the asymptotic yield obtained 40 minutes after the extraction time at pressures ranging from 34.47 MPa to 44.82 MPa. Meanwhile at low pressures ranging from 27.58 MPa to 31.03 MPa, the asymptotic value achieved 50 minutes after the extraction time. It can be assumed that a decrease in pressure would prolong the extraction time. This was due to the increase in the solvating power as the pressure increased. As the solvating power increased, the interaction between the fluid and the matrix became stronger [8].

From the Figure 2, it is obvious that the oil yield increased with the increase of the temperature. These phenomena occurred

due to the increase of the oil volatility in this condition was more dominant compared to the drop in the density of CO<sub>2</sub>. In addition, as the temperature increased, the solvent approached the gas-like state, facilitating in the increase of extraction rate which produced a faster extraction to reach asymptotic yield particularly at high pressure condition [2].

The increase of temperature from 40 °C to 70 °C at the pressure of 27.58 MPa shows that the increment of mass of oil obtained was 50.72%. On the other hand, at the constant pressure of 44.82 MPa, the maximum mass of oil obtained was 89.89 mg at 70 °C. The increment of oil mass from 40 °C to 70 °C at this condition was 35.34%. It shows that the increment at high pressure was lower than increment at low pressure. This is because the condition of high pressure and temperature facilitated the extraction rate to obtain a high performance in extraction.

Figure 4 shows the effect of different temperature at highest pressure (44.82 MPa). From the observation, it shows that the asymptotic yield achieved after 40 minutes of extraction time. This phenomenon was due to the increase of pressure resulting in the increase of the solvating power. Meanwhile, the increase in temperature resulted in the increase of vapor pressure which contributed to the increase of mass transfer rate of oil into the solvent. In fact, the highest mass of oil was obtained at high pressure and temperature.



**Figure 4** Effect of temperature on extracted *P.Jiringan* oil at constant 44.82 MPa as a function of extraction time

### 3.3 Solubility of *P.Jiringan* Oil in SC-CO<sub>2</sub>

The solubility of *P.Jiringan* oil in SC-CO<sub>2</sub> was evaluated at temperatures ranging from 40 °C to 70 °C and pressures ranging from 27.58 MPa to 44.82 MPa. The solubility data was obtained using a dynamic method and overall extraction curves were plotted as mass of carbon dioxide used versus mass of oil extracted. The solubility data for each condition were obtained from the slope of the linear portion of the overall extraction curve.

Figure 5 shows the effects of pressure and temperature on the solubility of *P.Jiringan* oil in SC-CO<sub>2</sub>. The result shows that at constant temperature, the solubility of oil increased as the density of CO<sub>2</sub> increased. Meanwhile, at constant pressure, the solubility of oil increased as the temperature increased even though the density of CO<sub>2</sub> decreased. From the observation, it showed that as the pressure increased from 27.58 MPa to 31.03 MPa, the solubility increased drastically at temperature 60 °C and 70 °C. However, the solubility was slightly increased at the temperatures of 40 °C and 50 °C. This was due to the competing effect which the increase of vapor pressure was more dominant than the decrease of CO<sub>2</sub> density.

A similar result studied on the effects of pressure and temperature on the solubility of *P.Jiringan* oil in SC-CO<sub>2</sub> had been reported [2]. The parameters used in his study were pressures at the range of 20.68 MPa to 55.15 MPa and temperatures at 40 °C, 50 °C, 60 °C, 70 °C and 80 °C. The resulted show that the solubility increased as the temperature increased at a constant pressure. Moreover, the solubility effect as a function of pressure

was less significant compared to the solubility effect as a function of temperature. The solubility in his study increased from 1.10 to 2.60 mg oil per g CO<sub>2</sub> at the constant pressure of 55.15 MPa as the temperature increased from 40 °C to 80 °C. It shows that the effect of temperature was more dominant than the pressure.

This trend was similarly reported by Ana Najwa [9] findings which showed the increment of solubility where the increase in pressure was slower than as the increase in temperature. Besides that, the solubility of palm oil in R134a at 60 °C and 80 °C suddenly increased from 45 to 60 bar. This is due to the balancing between the increase in the solute vapor pressure and the decrease in solvent density which is known as competing effect. This phenomenon occurred at the lowest and the highest temperature. In her study, she assumed that the solute vapor pressure played a major role in increasing the oil solubility and she concluded that temperature had a dominant effect on palm oil solubility.

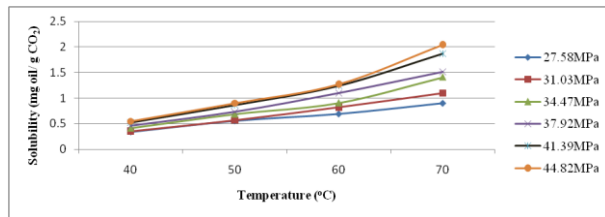


Figure 5 *P.Jiringan* oil solubility as a function of temperature at different pressure

The solubility data of *P.jiringan* oil in SC-CO<sub>2</sub> in this study were correlated with models proposed by Chrastil (Equation 3) and by Del Valle and Aguilera (Equation 4). Figure 6 shows the correlation of solubility experimental data of *P.Jiringan* oil solubility in SC-CO<sub>2</sub> by chrastil model and by del Valle and Aguilera model.

In this analysis, the experimental data was accommodated with both models to obtain the parameters involved. The parameters of Chrastil’s model (*a*, *b* and *k*) and del Valle and Aguilera model (*a*, *b*, *c* and *k*) were determined by minimizing the objective function (Equation 2) which was known as average absolute percent deviation (AAPD%) using Solver in Excel 2007 program.

In Chrastil’s model, the parameters obtained represent the important term. For parameter *a*, it represents the heat of salvation and vaporization of the solute, meanwhile parameter *b* represents the molecular weight and melting point of solute involved and *k* represents the average number of molecules that form the solvate-complex. From the analysis of the results, the AAPD% of correlation between the solubility experimental data with the Chrastil’s model obtained was 0.206%.

Moreover, in del Valle and Aguilera model, the parameters used were the same as Chrastil’s model and *c* was the additional constant for the model. The analysis was done by minimizing the AARD% and directly determined the constant parameters which were *a*, *b*, *c* and *k* in the equation. The results from the analysis show that the AAPD% of correlation between the solubility experimental data with the del Valle and Aguilera model obtained was 0.5818%.

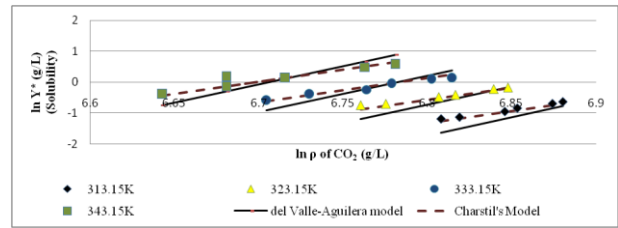


Figure 6 Correlation of *P.Jiringan* oil solubility in SC-CO<sub>2</sub> experimental data with Chrastil and del Valle-Aguilera model

Constant parameter of *a* showed the influence of the temperature change inside the extraction vessel. According to Maria Salud [10] as an absolute value of parameter *a* is high, it relates with high temperature influence. Moreover, they said that lower *k* value indicates that most of the oil was extracted with SC-CO<sub>2</sub> from the sample itself. Meanwhile, constant parameter *b* indicates that the solute had been extracted. In addition, according to Wahyu B. Sentianto [11], parameter *k* and *b* can be assumed as parameters that are independent of temperature. Following this, the solubility parameters *a*, *b*, *c* and *k* of Chrastil and del Valle-Aguilera model calculated are shown in Table 3.

Previous researchers had reported the solubility correlation by Chrastil’s model and by del Valle and Aguilera model in literature. The Chrastil model and del Valle-Aguilera model were used to correlate solubility data of virgin coconut oil (VCO) in CO<sub>2</sub> at temperature ranges of 40 °C to 80 °C and pressure ranges of 20.7 MPa to 34.5 MPa. The result shows that AAPD% of del Valle and Aguilera model obtained was 0.39% meanwhile, AAPD% of Chrastil model obtained was 0.93%. Even though both models gave good correlation of the data, he conclude that del Valle and Aguilera model as the best model correlated with the experimental data since the AAPD% of del Valle and Aguilera model is lowest compared to AAPD% of chrastil model [12]. The results show that most of the data obtained from Chrastil model approaching nearly to the solubility of experimental data compared to data obtained from del Valle and Aguilera model. Therefore, it shows that the Chrastil model was successful matched to the solubility experimental data compared to del Valle and Aguilera model.

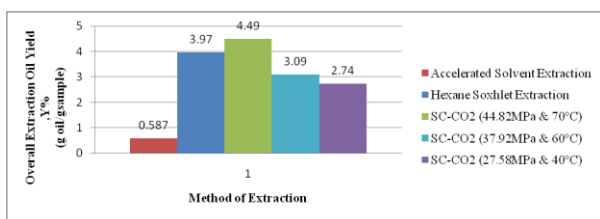
In addition, the value of AAPD% of Chrastil’s and del Valle and Aguilera models was used as a comparison. The result of AAPD% calculated for Chrastil model and del Valle and Aguilera model are shown in Table 3. The AAPD% from del Valle-Aguilera model was poor compared to the value from Chrastil model. The best model that fitted the experimental data was obtained by Chrastil model (Equation 2) with the smallest AAPD% value.

Table 3 Coefficient parameter and AAPD% for Chrastil and del Valle-Aguilera model

Model	Model Coefficient				Average absolute percent deviation (AAPD%)
	<i>a</i>	<i>b</i>	<i>c</i>	<i>k</i>	
Chrastil	-7549.77	-29.9029	-	7.74925	0.206
Del Valle-Aguilera	-10001.2	-20000	-49.7187	11.78452	0.5818

### 3.4 Comparison of the SC-CO<sub>2</sub> Extraction with ASE and Hexane Soxhlet Extraction

An overall extracted oil yield obtained from the various condition of SC-CO<sub>2</sub> was compared with those obtained from different methods of extraction, hexane soxhlet extraction and ASE. The various condition of SC-CO<sub>2</sub> was selected at the higher conditions (44.82 MPa and 70 °C), low conditions (22.58 MPa and 40 °C) and moderate conditions (37.92 MPa and 60 °C) in order to compare the performance of SC-CO<sub>2</sub> method than others. The highest overall extraction oil yield is 4.49 % with the SC-CO<sub>2</sub> at highest condition, followed by hexane soxhlet extraction yield of 3.97%. Finally the lowest oil yield of 0.587 % obtained from ASE, as presented in Figure 7. The SC-CO<sub>2</sub> conditions at 44.82 MPa and 70 °C was generated higher oil yield than other extraction mediums. This is because high oil solubilization capability than hexane soxhlet and ASE as well as capable to extract the groups of oleo resin and waxes (typically waxy materials in the outer part of the seed). According to Taylor [13] in the Stahl's extraction rules with pure CO<sub>2</sub>, the more strongly polar (e.g amino acid) only could be extracted in the range above 40.00 MPa. Therefore, at the higher SC-CO<sub>2</sub> extraction conditions, CO<sub>2</sub> solvent was able to extract both, the polar and non-polar compounds.



**Figure 7** Comparison of an overall extraction oil yield obtained from different conditions of SC-CO<sub>2</sub> with Hexane Soxhlet and ASE

### 4.0 CONCLUSION

The extraction of *P.Jiringan* seeds using supercritical carbon dioxide extraction was performed at temperatures ranging from 40°C to 70°C and pressures ranging from 27.58MPa to 44.82MPa. Results showed that the mass of *P.Jiringan* oil increased as the pressure increased at constant temperature. Similarly, the mass of extracted oil also increased as the temperature increased at constant pressure. The maximum mass of oil obtained was 89.89mg at the highest supercritical extraction condition of pressure at 44.82MPa and the temperature at 70°C, respectively.

The solubility of *P. Jiringan* oil in supercritical carbon dioxide was measured from the slope of the linear portion of the overall extraction curve. At constant temperature, the solubility of *P.Jiringan* oil increased as the density of CO<sub>2</sub> increased. In the meantime, at constant pressure, the solubility of oil increased as the temperature increased even though the density of CO<sub>2</sub> decreased. The maximum solubility of *P.Jiringan* oil was 2.0462 mg oil per g of CO<sub>2</sub> obtained at the temperature of 70°C and the pressure of 44.82MPa. On the other hand, the minimum solubility of *P.Jiringan* oil was 0.3394 mg oil per g of CO<sub>2</sub> obtained at the lowest supercritical condition which was at the temperature of 40°C and the pressure of 27.58MPa.

The experimental solubility data was correlated with two different empirical equations proposed correspondingly by Chrastil and del Valle and Aguilera. The lowest deviation between the experimental data and correlated values was obtained from the results taken at 60°C and 37.92MPa using the Chrastil model. From the observation, the solubility data were successfully fitted to the Chrastil model compared to del Valle and Aguilera model based on the AARD% value. The AARD% of Chrastil model which was 0.206% was lower than AARD% of del Valle and Aguilera model which was 0.5818%.

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