

MASS TRANSFER AND SOLUBILITY OF CORIANDER (*Coriandrum sativum* L.) SEED

Muhammad Hakim Bin Salahudin, Miss Sitinoor Adeib Binti Idris

Faculty of Chemical Engineering, Universiti Teknologi Mara

Abstract—The experimental data was obtained from literature to be analyzed with the application of two correlation models which are Chrastil's model (CH) and Del Valle and Aguilera's model (DVA) to estimate the solubility data. Coriander was extracted at temperature of 313, 328 and 343 K and operating pressure of 100, 150 and 200 bar in supercritical carbon dioxide (SC-CO₂). Both solubility and density of solvent are closely related with each other. The results proved that as the density of SC-CO₂ increased, the solubility of coriander increased but decreasing with temperature. It can be conclude that the relationship between the temperature and the solubility of coriander were not linear to each other. The modified model, DVA model it is expected have smaller value of AARD (%) that is 0.01 less than CH model. Smaller value of AARD indicates more satisfactory correlation and more accuracy of the correlation models to predict the solubility of coriander in SC-CO₂.

Keywords— Coriander, , Chrastil's Model, Del Valle and Aguilera's Model, Solubility, AARD

I. INTRODUCTION

Coriander (*Coriandrum sativum* L.) also known as cilantro or Chinese parsley. Coriander is a culinary and medicinal plant originating from the Mediterranean region, and is nowadays grown widely all over the world. The leaves are variously referred to as coriander leaves in Britain; cilantro in the United States, and "yanshui" in China (Chen et al., 2011).

Coriander is a soft plant growing to 50 cm (20 in) tall. The leaves are variable in shape, broadly lobed at the base of the plant, and slender and feathery higher on the flowering stems. The oil and various extracts from Coriander have also shown the activities of antibacterial, anti-cancerous and anti-mutagenic (Chithra & Leelamma, 2000), antioxidant, and anti-diabetic (Gallagher, Flatt, Duffy, & Abdel Wahab, 2003). Thus, this plant displays a sufficient of biological activity and is generally used in folk medicine and the human diet, it is important to extend the study to preparative scale.

Currently, oil of aromatic herbs is traditionally obtained by hydro distillation, steam-distillation or solvent extraction. Wang et al. (2009) reported that as an alternative, supercritical fluid extraction (SFE) is a particularly suitable method. Carbon dioxide is an ideal solvent because it is non-toxic, non-explosive, readily available and easy to remove from extracted products. SFE has the ability to use low temperatures, leading to less deterioration of the thermally labile components in the extract with comparable or better recovery, and minimal alteration of the active ingredients; thus the curative properties can be preserved.

Nowadays, the application using supercritical fluid extraction towards plants is extensively research because of the fact that other methods have huge disadvantages for example hydro-distillation

methods. This method undergoes chemical alterations and the heat-sensitive compounds can easily be destroyed. Therefore, the quality of the oil extracts is extremely impaired (Daood et al., 200).

Thus, supercritical fluid extraction is more suitable for process that involving extracting oils for it quality. The parameters used for the extraction in this study are temperature and pressure as these two variables play an important role to the overall extraction process performance.

For solubility study, there are many model proposed by many researcher in the past. The example of modeling for solubility behavior using supercritical fluid extraction includes Chrastil model, dell Valle and Aguilera model, Gordillo model, Sovova model and Yu model.

There was not much study on solubility data of coriander seed which is important due to the characteristics which indicates the equilibrium solubility of solutes in supercritical fluids corresponds to the limit of the total amount of solutes that can be extracted at saturation equilibrium. This research is conducted due to the lack of solubility data in previous literature about the supercritical carbon dioxide extraction. This data is very important for future references and also for scale-up production of others plant seeds.

II. METHODOLOGY

A. Data to be Analyzed

The data (Table 1) is obtained from previous related studies by Zeković. Z [11]. Samples of 50.0 g coriander were placed in an extractor vessel. Extraction process was carried out and extraction yield was measured after 15, 30, 45, 60, 90, 120, 150, 180 and 240 min of extraction. Pressure (100, 150 and 200 bar), temperature (40, 55 and 70°C) and CO₂ flow rate (0.2, 0.3 and 0.4 kg/h) were independent variables in the process, while all other SFE parameters were held constant [11].

Table 1: Secondary data from literature (Zeković. Z et al., 2016)

Run	P [bar]	T [°C]	CO ₂ flow rate [kg/h]	ρ [kg/m ³]	Y [%]
1	150	70	0.2	515.35	2.05 ^a
2	200	40	0.3	839.90	5.95 ^a
3	100	55	0.4	337.20	1.20 ^a
4	150	70	0.4	515.35	3.50 ^a
5	200	70	0.3	658.95	5.36 ^a
6	150	40	0.2	780.30	4.31 ^a
7	200	55	0.2	754.10	4.90 ^a
8	150	55	0.3	651.85	3.77 ^a
9	150	40	0.4	780.30	5.64 ^a
10	150	55	0.3	651.85	4.02 ^a
11	100	55	0.2	337.20	0.95 ^a
12	200	55	0.4	754.10	7.00 ^a
13	100	40	0.3	628.70	2.69 ^a
14	150	55	0.3	651.85	4.00 ^a
15	100	70	0.3	255.80	0.59 ^a
16	200	55	0.3	754.10	7.16
17	100	55	0.3	337.20	1.47
18	150	40	0.3	780.30	5.53
19	150	55	0.2	651.85	3.89
20	150	55	0.4	651.85	5.54
21	150	70	0.3	515.35	4.63

B. Solubility Correlation

The solubility depicts the maximum possible capacity of the SC-CO₂ at certain temperature, pressure and also concentration of the co-solvent in SC-CO₂. A reliable correlation can be utilized to estimate the solubility of the oil in SC-CO₂ since the experimental measurement of solubility is tedious and also time consuming at different conditions.

$$\ln y = k \ln \rho + a/T + b \quad (1)$$

Equation above (1), represents the solubility Chrastil's model (CH). The y represents the solubility of solute in g/L expression, T is the temperature of the system (K) and ρ is the solvent density (g/L). The k constant represents the number of CO₂ molecules present in the complex solute-solvent while a and b parameters indicate the vaporizing enthalpy and molecular weight dependant. a , b and k in the equation represent the adjustable constants of the model that are specific for each solute-supercritical solvent pair and not dependent on temperature and pressure.

$$\ln y = k \ln \rho + a/T + b/T^2 + c \quad (2)$$

Equation (2) is Del Valle and Aguilera mode (DVA) that proposed adequate for temperature in a range of 293 K to 353 K and pressure varying from 150 bar to 880 bar. It is also the modification of the Chrastil's model.

C. Average Absolute Relative Deviations (AARD)

All the experimental values from previous related studies, the operating condition and the solvent correspond density will be tabulated for each models and solubility will be calculated for each temperature variation and density changes during the extraction process. The comparison of the experimental and calculated solubility will be done in a plotted graph of correlated solubility ($\ln y$) versus density of CO₂ ($\ln \rho$).

$$AARD = \frac{100}{n} \sum \frac{y_{\text{experimental}} - y_{\text{calculated}}}{y_{\text{experimental}}} \quad (3)$$

Where n is number of data, y experimental is solubility data obtained from experimental respectively at i th condition and y calculated is solubility data obtained from model respectively at i th condition.

II. RESULTS AND DISCUSSION

A. Correlation by using Solubility Models

Fig.1 and fig.2 shows the relationship between coriander solubility and the SC-CO₂ density according to CH model and DVA model, respectively. Both solubility and density of solvent are closely related with each other. The plots proved that as the density of SC-CO₂ increased, the solubility of coriander increased. However, as shown in both figure 1 and 2, the plots were decreasing with temperature. It can be said that the relationship between the temperature and the solubility of coriander were not linear to each other.

Both figures really resemble with each other due to the fact that DVA model is a model that modified from CH model. Thus, the comparison is hard to tell. It is recommended for further studies to use more than 4 models for easier comparisons.

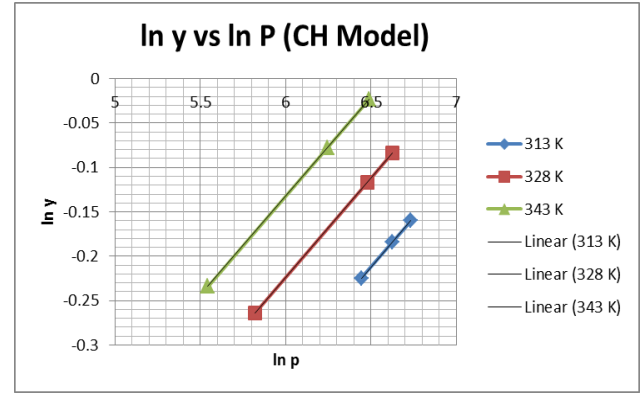


Fig.1: Correlation of Coriander solubility in SC-CO₂ with Chrastil's model (CH)

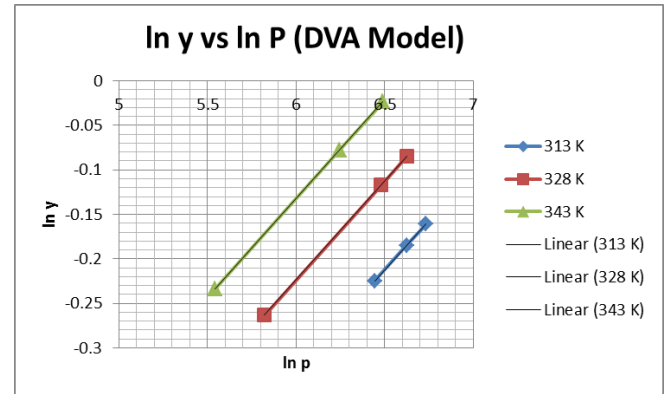


Fig.2: Correlation of Coriander solubility in SCCO₂ with Del Valle and Aguilera's model (DVA)

There is 0.01 different between AARD CH model and DVA model (Table 2). Thus, higher the numbers of parameters present in each model, the higher the accuracy of the correlation models, hence, this is one of the important factors of the precision of the correlation [5]. Usually, the more parameters, the calculated results will have high precision and the better the results calculated.

Table 2: Correlated Results of Empirical Models for the Solubility of Coriander in SC-CO₂

Model	a	b	c	d	k	AARD (%)
Chrastil	- 684.67	0.52367	-	-	0.22332	4.4980
Del Valle	- 683.33	0.5000	0.52893	-	0.22180	4.4859

III. CONCLUSION

The modified model, DVA model is the empirical models which has a higher number of parameters present than the CH model. Thus, it is to be expected to have the minimum AARD (%) value that indicates more satisfactory correlation results that shows more accuracy of the correlation models to predict the solubility of coriander in SCCO₂. This matched with one of the objective of the study to determine the most suitable correlation to predict the solubility of coriander in SCCO₂.

ACKNOWLEDGMENT

I would like to express the deepest appreciation to all the lecturers of Faculty of Chemical Engineering of UiTM Shah Alam for the guidance and encouragement.

References

- [1] Bušić, A., Komes, D., Vladić, J., & Pavlić, B. (2015). Coriander seeds processing: Sequential extraction of non-polar and polar fractions using supercritical carbon dioxide extraction and ultrasound-assisted extraction. *Food and Bioproducts Processing*, 218-227.
- [2] Chen, Q., Yao, S., Huang, X., & Wang, J. (2009). Supercritical fluid extraction of *Coriandrum sativum* and subsequent separation of isocoumarins by high-speed counter-current chromatography. *Food Chemistry*, 504-509.
- [3] Grosso, C., Ferraro, V., Coelho, J., & Palavra, A. (2008). Supercritical carbon dioxide extraction of volatile oil from Italian coriander seeds. *Food Chemistry*, 197-203.
- [4] Illeš, V., Daood, H., Perneczki, S., Szokonya, L., & Then, M. (2000). Extraction of coriander seed oil by CO₂ and propane at super- and subcritical conditions. *Journal of Supercritical Fluids* 17, 177-186.
- [5] Tang, Z., Jin, J., Zhang, Z., & Liu, H. (2012). New Experimental Data and Modeling of the Solubility of Compounds in Supercritical Carbon Dioxide. *Industrial & Engineering Chemistry Research*, 51(15), 5515–5526. <https://doi.org/10.1021/ie2016224>
- [6] Laribi, B., Kouki, K., M'Hamdi, M., & Bettaieb, T. (2015). Coriander (*Coriandrum sativum* L.) and its bioactive constituents. *Fitoterapia*, 9-26.
- [7] Mandal, S., & Mandal, M. (2015). Coriander (*Coriandrum sativum* L.) essential oil: Chemistry and biological activity. *Asian Pacific Journal of Tropical Biomedicine*, 421-428.
- [8] Msaada, K., Hosni, K., Taarit, M., Chahed, T., & Marzouk, B. (2007). Changes on essential oil composition of coriander (*Coriandrum sativum* L.) fruits during three stages of maturity. *Food Chemistry*, 1131-1134.
- [9] Tomita, K., Sasaki, M., Goto, M., & Fukuzato, R. (2013). Extraction and solubility evaluation of functional seed oil in supercritical carbon. *The Journal of Supercritical Fluids*, 109-113.
- [10] Vaughn Katherine, L. S., Clausen Edgar, C., & King Jerry, W. (2008). Extraction conditions affecting supercritical fluid extraction (SFE) of lycopene from watermelon. *Bioresource Technology* 99, 7835–7841.
- [11] Wangensteen, H., Samuelsen, A., & Malterud, K. (2004). Antioxidant activity in extracts from coriander. *Food Chemistry* 88, 293-297.
- [12] Z. Zeković, B. Pavlić, A. Cvetanović, S. Đurović, Supercritical fluid extraction of coriander seeds: process optimization, chemical profile and antioxidant activity of lipid extracts, *Ind. Crops Prod.* 94 (2016) 353–362.