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4 **SUPERCritical CO2 EXTRACTION OF PHENOLIC COMPOUNDS IN ROSELLE (HIBISCUS SABDARIFFA L.)**

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4Supercritical CO₂ Extraction of Phenolic Compounds in Roselle (Hibiscus sabdariffa L.)

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1of Chemical Engineering, Widya Mandala Surabaya Catholic University, Kalijudan, Surabaya, Indonesia **2**Department of Chemical Engineering, National Taiwan University of Science and Technology,

Taipei, Taiwan The extraction of phenolic compounds from roselle using supercritical CO₂ was studied. The supercritical fluid extraction experiments were conducted at a range of

4pressures from 10 to 24 MPa and temperatures of 323.15, 333.15, and 343.15 K.

The extraction time was 2.5 h. Three density-based equations, Chrastil, del Valle and Aguilera, and Méndez-Santiago and Teja, were used to correlate the experimental data. All three equations correlate with the actual the data quite well. The maximum yield obtained from the supercritical extraction experiment was 8.63 mg GAE=g dried rosella at 343.15 K and 24 MPa. Keywords Density-based model; Extraction; Phenolic compounds Introduction Roselle (Hibiscus sabdariffa L.) is a species of native hibiscus belonging to the family of Malvaceae. This crop is widely grown in Southeast Asia, West Africa, and Central America. Currently, it is cultivated extensively for food, beverage, and pharmaceutical applications (Abou-Arab et al.,

2011; Hussein et al., 2010; Mahadevan et al., 2009; Mohamed et al., 2007; Odigie et al., 2003; Tseng et al., 2000). Roselle contains many chemical compounds that have antioxidant activity and potential health benefits (Mahadevan et al., 2009). The roselle flower contains anthocyanins, flavonoids, and polyphenols (Abou-Arab et al., 2011; Tzu-Lin et al., 2007), and the petals are a good source of ascorbic acid and anthocyanins (Abou-Arab et al., 2011; Prenesti et al., 2007). The calyx is a rich source of organic acids, phytosterols, and polyphenols (Abou-Arab et al., 2011). Because of its high antioxidant content, roselle has been used for medical purposes for a long time. Furthermore, the beneficial effect of antioxidants in roselle for managing some human diseases has been reported in the literature (Cisse et al., Address correspondence to Suryadi Ismadji, Department of Chemical Engineering, Widya Mandala Surabaya Catholic University, Kalijudan 37, Surabaya 60114, Indonesia. E-mail: suryadiismadji@yahoo.com 1187 2011; Mohd-Esa et al., 2010). Because of its potential beneficial health effects in humans, studies on the recovery of antioxidants and other compounds from different parts of the roselle crop have been conducted by many research groups (Abou-Arab et al., 2011; Mohamed et al., 2007). The most widely used extraction method features various polar solvents (water, methanol, ethanol, etc), and only a few studies have been conducted using supercritical fluid extraction methods (Mourtzinou et al., 2008; Nyam et al., 2010). The main problem of antioxidants obtained by solvent extraction is their stability during extraction at high temperature and also the presence of various endogenous enzymes (Cisse et al., 2011). Extraction of antioxidants using supercritical fluid as the solvent has received much attention over the past three decades since it offers several advantages over conventional liquid extraction: energy savings, higher selectivity and purity, more rapid extraction and phase separation, and solvent-free product. Supercritical extraction of chemical compounds or antioxidants from roselle is still limited, and we have found only two studies using this technology to extract oil from roselle seeds (Mourtzinou et al., 2008; Nyam et al., 2010). In this study we utilized supercritical fluid technology to extract flavonoid (phenolic) compounds from dried calyces of roselle (*Hibiscus sabdariffa* L.). The supercritical fluid extraction of phenolic compounds from dried calyces was conducted at 323.15, 333.15, and 343.15 K in a pressure range of 10 to 24 MPa, and acetone was used as a co-solvent. The selectivity and solvent effectiveness of supercritical fluid was shown to be affected by small changes in both pressure and temperature. Materials and Methods Dried calyces of roselle (*Hibiscus sabdariffa* L.) were obtained from Malang, East Java, Indonesia. The chemical composition of dried roselle is: water content 7.8%, protein 34.1%, total dietary fiber 18.8%, carbohydrate 14.3%, ash 2.6%, and fat 22.4%. Prior to extraction, dried calyces were crushed into powder using a micro hammer mill (Janke & Kunkel) to obtain a particle size of 40–60 mesh. The powdered dried calyces were then stored in a freezer until extraction. Food-grade carbon dioxide with a minimum purity of 99.95% was obtained from PT Aneka Gas (Surabaya, Indonesia). This gas was obtained

8 **gas liquid CO₂ (supplied with a deep tube liquid supply cylinder).**

Analytic-grade acetone was obtained from Sigma Aldrich Singapore, and Folin-Ciocalteu phenol reagent and sodium bicarbonate were obtained from Merck (Darmstadt, Germany). Supercritical Fluid Extraction Procedure Supercritical fluid extractions were carried out in a semi-pilot plant supercritical fluid extractor. The extractor system consisted of a high pressure pump (Eldex AA-100-S-2-CE, USA) and pressure transducer (Druck PTX 611, USA) with digital process indicator (Druck DPI 280, USA). The deviation of pressure measurement was 0.01 MPa. A Memmert oven equipped with temperature controller was used as a heating chamber. The deviation of temperature measurement in the heating chamber was 1 K. The supercritical fluid extraction system was also equipped with a vacuum pump (Gast DOA-P504 BN, USA), volume calibrator (Zeal DM3B, UK), and high-pressure extraction column. The volume of the extraction column was 150 cm³. All fittings and tubing used in the system were made of stainless steel (Swa-gelok, USA).

8 **The maximum working pressure and temperature of the supercritical extraction system were 40 MPa and 373.15 K, respectively. The**

procedure of supercritical fluid extraction of roselle calyx powder was as follows. Ten grams of roselle calyx powder were introduced into a high-pressure extraction column. At both ends of the extraction column, 0.5

mm filters were placed to prevent any carryover of particles. Before the supercritical fluid extraction proceeded, the

1 system was evacuated using a vacuum pump (Gast DOA-P504 BN,

USA) until an appropriate vacuum was attained. Subsequently, the system was heated to different temperatures (323.15, 333.15, and 343.15 K). When the desired temperature was reached, the liquid CO₂ was pumped into the system using a high-pressure pump (Eldex AA-100-S-2-CE, USA). The ranges of pressures used in this study were 10 to 24 MPa. The supercritical fluid extraction was carried out for 2.5 h in batch mode method. Acetone was employed as co-solvent (5% V=V). After the extraction was completed, the product gas (CO₂ þ phenolic compounds þ co-solvent) was bubbled into a collecting glass vessel filled with 100 mL of acetone. Analysis

3 Total phenolic content was determined with Folin-Ciocalteu reagent using the method of Kahkonen et al. (1999). One mL of each sample extract was mixed with 4 mL of a 10-fold dilution of Folin-Ciocalteu reagent and 5 mL of 7.5% (w=v) bicar- bonate solution and allowed to stand at room temperature for 30 min. The absor- bance was measured at 760 nm, using a Shimadzu UV- vis spectrophotometer.

The total phenolic content was expressed as gallic acid equivalents (GAE) in milligrams per gram of dry material. Results and Discussion

5 Supercritical fluids have densities much greater than those of typical gases and slightly less than those of organic liquids. On the other hand, the viscosities of super- critical fluids are nearer to those of typical gases and less than those of liquids. These features ensure high fluid phase capacity concomitant with favorable transport properties, making supercritical fluids

attractive as solvents for extraction. The den- sity of a supercritical fluid plays an important role in the extraction process. By increasing density, the solvating power of the supercritical fluid also increases. The densities of supercritical CO₂ at various temperatures and pressures were calculated using the Peng-Robinson equation of state as modified by Stryjek-Vera (Stryjek and Vera, 1986): $P = \frac{RT}{v - b} - \frac{a}{v(v + b) + c}$ where $a = \frac{0.457235 R^2 T_c^2}{P_c} \alpha$, $b = \frac{0.77796 RT_c}{P_c} \beta$, $c = \frac{0.1 P_c}{P_c} \beta$, $\alpha = \left[1 + \frac{0.5 P_c}{P_c} \left(1 - \frac{0.5 P_c}{P_c} \right) \right]^2$, $\beta = \frac{0.378893}{P_c} \left[1 - \frac{0.17131848 x^2}{P_c} + \frac{0.0196554 x^3}{P_c} \right]$. Here P is pressure (bar),

16 R is gas constant (8.314 J=mol K), and T is temperature (K). The molar volume of the

system given by v (cm³=mol), while parameter a_0

12 is a measure of the attractive force between the molecules and parameter b_0 is related to the size of the molecules. The

critical temperature (K) and pressure (bar) are given as T_c and P_c , respectively, T_R is reduced temperature, x is an acentric factor, and j

12 is an adjustable parameter characteristic for each pure component.

2 The Peng- Robinson Stryjek-Vera (PRSV) equation of state is a twofold modification of the PR equation of state that extends the application of the original PR method for moderate nonideal systems. It has been shown to match vapor pressure curves of pure components and mixtures more accurately than the PR method, especially at low vapor pressures

(Stryjek and Vera, 1986). The

1 densities of supercritical CO₂ at various pressures and temperatures

are given in Figure 1. Figure 2 depicts the amount of phenolic compounds extracted by supercritical CO₂ at various pressures and temperatures. The total phenolic compounds extracted are given by mg of GAE (galic acid equivalent)=g of dried roselle. In general, the amount of phenolic compounds increased with the increase of temperature and pressure. The maximum yield obtained from the supercritical extraction experiment was 8.63 mg GAE=g dried rosella at 343.15 K and 24 MPa. This result is slightly higher than that obtained by extraction using water as solvent at 95 C (Wongsa et al., 2012). In their experiment the total amount of phenolic compounds recovered from dry roselle was 8.31 mg GAE=g of dried roselle. With the increase of pressure, the density of the supercritical fluid also increased, and, as mentioned before, the solvating power of supercritical carbon dioxide increases with increase in density. Figure 1. Densities of supercritical CO₂ at various pressures and temperatures. Figure 2. Total phenolic compounds extracted at various pressures and temperatures. Increasing pressure increases solvent density allowing for greater permeation through the matrix. This leads to an increase in the amount of phenolic compounds extracted from the solid matrices. At the critical condition, an increase of temperature will increase the diffusivity of carbon dioxide. Meanwhile, viscosity decreases with an increase in temperature. This phenomenon enhances the penetration of carbon dioxide molecules into the interior of the solid matrix of roselle. Since more carbon dioxide molecules are in contact with the phenolic compounds within the interior of roselle, more phenolic compound molecules were dissolved into the supercritical carbon dioxide. The total amount of phenolic compound extracted in supercritical carbon dioxide is correlated with several density-based models. The advantage of using these density-based correlations is that it does not require any physical or chemical data from the compounds extracted. The earliest density-based model was developed by Chrastil (1982), which has the following form: $c = \frac{1}{4} k \exp \left(\frac{b}{T} \right) \rho$

8 where c is the solubility of the solute in gas,

q

6 is the density of gas, k is an association number, a is a function of the enthalpy of solvation and enthalpy of vaporization, and b is a function of the association number and molecular weights of the solute and supercritical

fluids. Based on the density model

1 by Chrastil, del Valle and Aguilera (1988) proposed another density-based

model. Their model considered

1 the change in enthalpy of vaporization with temperature:

The physical meaning of the parameters k , b , and a is similar to that of the parameters in the Chrastil model. The parameter d is a parameter associated with the change in enthalpy of vaporization. A more complex model (which includes the sublimation pressure of the solid) was proposed by Méndez-Santiago and Teja (1999). The Méndez-Teja equation is based on dilute solution theory. However, if the sublimation pressure of the solute is not known, a modified version

7 of the Méndez-Santiago and Teja model in the

following form can be used: $T \ln y_2 P = A_0 + B_0 q + C_0 T$. Here A_0 , B_0 , and C_0 are the parameter constants, while y_2 is the fraction of solid in supercritical fluid CO_2 . The parameters of the models were estimated by a nonlinear least squares analysis of the solubility data. The calculation or fitting procedure was done for the solubility data for all temperatures simultaneously by minimizing the sum of squared errors as objective function: $\text{SSE} = \sum_{i=1}^N (P_{\text{cal}} - P_{\text{exp}})^2$. Here c_{exp} is the actual

13 solubility of phenolic compound in supercritical CO_2 , c_{cal} is the

calculated solubility,

16 and N is the number of experimental data. Figure 3 shows the solubility data and a plot of

the Chrastil model, with the parameters of the Chrastil equation obtained by nonlinear regression. The results are summarized in Table I. In general, the Chrastil model can represent the total Figure 3.

13 Solubility of total phenolic compounds in supercritical CO_2 and

plots of Chrastil model. Table I. Parameters of Chrastil equation Temperature (K) a b k SSE 323.15 5745 10.75 3.13 0.097 333.15 343.15 amount of phenolic compounds extracted in supercritical carbon dioxide quite well (Figure 3). The Chrastil model is a semi-empirical equation that was developed based on the assumption that the interaction of the solute and gas molecules causes

9 the formation of a solvato complex that is in equilibrium with the gas

(Chrastil, 1982). As stated by Chrastil, parameter k is an association number. The physical meaning of parameter

7 k is the number of molecules of gas associated with one molecule of solute to form a solvato complex that is in equilibrium with the

system (Chrastil, 1982): $S = kG / (SG + k)$. And the equilibrium constant for Equation (10) can be written as $K = \frac{1}{2} S \frac{1}{2} G \frac{1}{2} k \frac{1}{2} SG \frac{1}{2} k$. Figure 4.

13 Solubility of total phenolic compounds in supercritical CO_2 and

plots

1 of del Valle and Aguilera model.

Table II. Parameters of

1 del Valle and Aguilera equation Temperature (K) a b d k

SSE 323.15 1.496 861.3 256541 3.13 0.089 333.15 343.15 where the

9 equilibrium constant K can be expressed as $K = \frac{1}{4} \frac{DH_{solv}}{RT}$

$\ln K = \frac{1}{4} \frac{DH_{solv}}{RT} - \frac{a}{RT}$ Here DH_{solv} and a are the heat of solvation and a constant parameter, respectively.

9 In most cases solvato complexes are not stoichiometric

due to the formation of more or less stable solvate complexes (Chrastil, 1982); therefore, the value of association number k will not be an integer. Thus, the association constant k expresses an average equilibrium association number, which is a characteristic constant for a given gas and solute (Chrastil, 1982). The value of the association number for CO_2 -phenolic compound system is 3.13. As mentioned before, a is a function of the enthalpy of solvation and the enthalpy of vaporization (Chrastil, 1982). Parameter a can be defined as: $a = \frac{1}{4} DH_{solv} - R \ln K$, where DH_{solv} is the total reaction heat, which is equal to $DH_{solv} + DH_{vap}$. The value of parameter a , obtained from the fitting of experimental data, is 5745. Therefore, the total reaction heat for CO_2 -phenolic compound system is 24.014 kJ/mol K. Figure 5.

13 Solubility of total phenolic compounds in supercritical CO_2 and

plots of Méndez-Santiago and Teja model. Table III. Parameters of Méndez-Santiago and Teja equation Temperature (K) A0 B0 C0 SSE 323.15 3271 2058 0.6891 0.098 333.15 343.15 Parameter b in the Chrastil model is a function of the association number and molecular weights of the solute and supercritical fluids (Chrastil, 1982): $b = \frac{1}{4} \ln \left(\frac{M_s}{M_g} \right) + \frac{1}{4} \ln \left(\frac{M_g}{M_s} \right) + \frac{1}{4} \ln \left(\frac{M_g}{M_s} \right) + \frac{1}{4} \ln \left(\frac{M_g}{M_s} \right) + \frac{1}{4} \ln \left(\frac{M_g}{M_s} \right)$ where M_s and M_g are the molecular weights of the solute and gas, respectively. The value of parameter b is 10.75. If the phenolic compounds in the system are assumed to be gallic acid with a molecular weight of 170.12, the constant q is 3.94. The fitted parameters of k , a , and b are then consistent with their physical meaning and have reasonable values and are comparable to other systems (Chrastil, 1982). Figure 4 shows the total amount of phenolic compounds extracted in supercritical carbon dioxide and plots of the del Valle and Aguilera model. The fitted parameters of this model are summarized in Table II. It seems that

4 del Valle and Aguilera model can represent the experimental data

better than the Chrastil model. This is not surprising since the del Valle and Aguilera model contains one more parameter than the Chrastil equation. Since this equation was derived from the Chrastil model, it gives a trend similar to the Chrastil equation in predicting the experimental data. The

4 plots of the Méndez-Santiago and Teja model to

the total amount of phenolic compounds obtained from supercritical fluid extraction with CO₂ are given in Figure 5. The fitted parameter values obtained from a simultaneous nonlinear fitting procedure are summarized in Table III. The parameters A₀, B₀, and C₀ are constants and therefore should be specific for a given gas and solute at supercritical condition. The model of

7 Méndez-Santiago and Teja also can represent **the solubility data** quite **well**

(Figure 5). Conclusion The extraction of flavonoids or phenolic compounds from roselle calyces under supercritical condition was conducted at various pressures and temperatures. In order to increase the polarity of CO₂, acetone was employed as a co-solvent (Foster et al., 1993). The amount of phenolic compounds extracted increased with an increase in pressure and temperature. The solubility data of phenolic compounds in supercritical CO₂ were correlated using density-based models. All the models used in this study correlated quite well with the experimental data. Acknowledgments We thank Dr. David Barkley from N. Simonson & Company (<http://www.virtlab.com>) for assistance in language editing. References Abou-Arab, A. A., Abu-Salem, F. M., and Abou-Arab, E. A. (2011). Physico-chemical properties of natural pigments (anthocyanin) extracted from roselle calyces (*Hibiscus sabdariffa* L.), *J. Am. Sci.*, 7, 445–456. Chrastil, J. (1982). Solubility of solids and liquids in supercritical gases, *J. Phys. Chem.*, 86, 3016–3021. Cisse, M., Vaillant, F., Soro, D., Reynes, M., and Dornier, M. (2011). Crossflow microfiltration for the cold stabilization of roselle (*Hibiscus sabdariffa* L.) extract, *J. Food Eng.*, 106, 20–27. del Valle, J. M., and Aguilera, J. M. (1988). An improved equation for predicting the solubility of vegetable oils in supercritical CO₂, *Ind. Eng. Chem. Res.*, 27, 1551–1553. Foster, N. R., Singh, H., Yun, S. L. J., Tomasko, D. L., and Macnaughton, S. J. (1993). Polar and non-polar cosolvent effect on the solubility of cholesterol in supercritical fluids, *Ind. Eng. Chem. Res.*, 32, 2849–2853. Hussein, R. M., Shahein, Y. E., El Hakim, A. E., and Awad, H. M. (2010). Biochemical and molecular characterization of three colored types of roselle (*Hibiscus sabdariffa* L.), *J. Am. Sci.*, 6, 726–733. Kahkonen, M. P., Hopia, A. I., Vuorela, H. J., Rauha, J. P., Pihlaja, K., Kujala, T. S., and Heinonen, M. (1999). Antioxidant activity of plant extracts containing phenolic compounds, *J. Agric. Food Chem.*, 47, 3954–3962. Mahadevan, N., Shivali, and Kamboj, P. (2009). *Hibiscus sabdariffa* Linn.—An overview, *Nat. Prod. Radiance*, 8, 77–83. Méndez-Santiago, J., and Teja, A. S. (1999). The solubility of solids in supercritical fluids, *Fluid Phase Equilib.*, 158, 501–510. Mohamed, R., Fernandez, J., Pineda, M., and Aguilar, M. (2007). Roselle (*Hibiscus sabdariffa* L.) seed oil is a rich source of α -tocopherol, *J. Food Sci.*, 72, 207–211. Mohd-Esa, N., Hern, F. S., Ismail, A., and Yee, C. L. (2010). Antioxidant activity in different part of roselle (*Hibiscus sabdariffa* L.) extracts and potential exploitation of the seeds, *Food Chem.*, 122, 1055–1060. Mourtzinis, I., Makris, D. P., Yannakopoulou, K., Kalogeropoulos, N., Michali, I., and Karathanos, V. T. (2008). Thermal stability of anthocyanin extract of *Hibiscus sabdariffa* L. in the presence of β -cyclodextrin, *J. Agric. Food Chem.*, 56, 10303–10310. Nyam, K. L., Tan, C. P., Lai, O. M., Long, K., and Man, Y. B. C. (2010). Optimization of supercritical fluid extraction of phytosterol from roselle seeds with a central composite design model, *Food Bioprod. Process.*, 88, 239–246. Odigie, I. P., Ettarh, R. R., and Adigun, S. (2003). Chronic administration of aqueous extract of *Hibiscus sabdariffa* attenuates hypertension and reverses cardiac hypertrophy in 2 K-1C hypertensive rats, *J. Ethnopharmacol.*, 86, 181–185. Prenesti, E., Berto, S., Daniele, P. G., and Toso, S. (2007). Antioxidant power quantification of decoction and cold infusions of *Hibiscus sabdariffa* flowers, *Food Chem.*, 100, 433–438. Stryjek, R., and Vera, J. H. (1986). PRSV: An improved Peng-Robinson equation of state for pure compounds and mixtures, *Can. J. Chem. Eng.*, 64, 323–333. Tseng, T., Kao, T., Chu, C., Chou, F., Lin, W., and Wang, C. (2000). Induction of apoptosis by *Hibiscus* protocatechuic acid in human leukemia cells via reduction of retinoblastoma (RB) phosphorylation and Bcl-2 expression, *Biochem. Pharmacol.*, 60, 307–315. Tzu-Lin, L., Lin, H. H., Chen, C. C., Lin, M. C., Chou, M. C., and Wang, C. J. (2007). *Hibiscus sabdariffa* extract reduces serum cholesterol in men and women, *Nutr. Res.*, 27, 140–145. Wongsap, P., Chaiwarit, J., and Zamaludien, A. (2012). In vitro screening of phenolic compounds, potential inhibition against α -amylase and α -glucosidase of culinary herbs in Thailand, *Food Chem.*, 131, 964–971. 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