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2**Supercritical CO2 Extraction of Phytochemical Compounds from Mimosa pudica Linn Astrid** Rahmawatia, **Debora** Panga, **Yi-Hsu** Jub, **Felycia Edi** Soetaredjoa, **Ong Lu**

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2**Supercritical CO2 Extraction of Phytochemical Compounds from Mimosa pudica Linn,**

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2**Supercritical CO2 Extraction of Phytochemical Compounds from Mimosa pudica Linn ASTRID** RAHMAWATI1, **DEBORA** PANG1, **YI-HSU** JU2, **FELYCIA EDI** SOETAREDJO1, **ONG LU** KI1, and **SURYADI**

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The extraction of phytochemicals from Mimosa pudica Linn (MPL) using supercritical CO2(SC-CO2) has been studied, and the results are presented in this article. The significant variables affecting this extraction were investigated by design of experiment analysis. The effect of temperature and pressure on phytochemical yields was explored by measuring the total phenolic content (TPC), total flavonoid content

(TFC), and total alkaloid compound (TAC). The maximum yield of TPC, TFC, and TAC extracted from MPL was 74.83 mg gallic acid equivalent (GAE)=g dry MPL, 30.93 mg quercetin equivalent=g dry MPL, and 14.23 mg hyos- cyamine (HE)=g dry MPL, respectively. The work of Chrastil was used

20**to correlate the experimental data. It was** found **that**

tem- perature and pressure greatly affected the yield of phytochemicals, whereas the effect of extraction time on the extraction yield diminished after 2 h. Qualitative analysis of the phytochemicals extracted from MPL was performed by high-performance liquid chromatography (HPLC). The SC-CO2 extraction was more selective than Soxhlet extraction based on the HPLC spectra. Keywords: Alkaloid; Extraction; Flavonoid; Mimosa pudica Linn; Phenolic; Supercritical CO2 Introduction Different kinds of plant varieties can be found in Indonesia, and they can be used for various kinds of applications such as the raw materials for specialty chemicals and biofuel (Zhou et al., 2011). One of them is Mimosa pudica Linn (MPL). This plant is commonly known as a sensitive or sleepy plant since its leaves fold inward when touched. It is available throughout Indonesia as a wild bush. The plant originally migrated from Brazil, and its weed distributed throughout the tropics (Elango et al., 2012). The stem of MPL is erect as a young plant but trails along the ground with age.

12**All parts of the** MPL **plant contain**

phytochemicals which have medicinal value. The benefits of phytochemicals in MPL have been investigated by various authors (Bendgude et al., 2012; Cheng et al., 2007; Saraswat and Pokharkar, 2012; Sowmya and Ananthi, 2011; Tanaka and Takeshi, 2004). Saraswat and Pokharkar (2012) extracted the phytochem- icals from MPL leaves using methanol as the solvent, and the extract was found to be an antidiabetic agent. Extracts of MPL leaf extract also have anthelmintic (Bendgude et al., 2012) and anti-inflammatory properties (Cheng et al., 2007). The aqueous extract of this plant (using decoction Address

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9**yahoo.com Color versions of one or more of the figures in the article can be found online at www.tandfonline.com/gcec.** method) **has an**

anti-nociceptive benefit (Tanaka and Takeshi, 2004), and the root extract contains high levels of antioxidants (Sowmya and Ananthi, 2011). Previous extraction studies of phytochemicals from MPL have been conducted using the Soxhlet extraction method which used polar organic solvents such as methanol (Saraswat and Pokharkar, 2012; Sowmya and Ananthi, 2011), ethanol (Sowmya and Ananthi, 2011; Tanaka and Takeshi, 2004), and even water (Cheng et al., 2007; Tanaka and Takeshi, 2004). Even though these extractions are quite simple, they require a complex separation process to obtain high purity of active compounds. Supercritical fluid extraction (SFE) is an attractive alternative for obtaining valuable compounds for use in pharmaceuticals (McHugh and Krukonis, 1986; Huang et al., 2004), nutraceutical (Riha and Brunner, 2000; Senorans et al., 2001), and flavor (Doker et al., 2010; Haas et al., 1989; Kim et al., 2008; Monteiro et al., 1997; Rodrigues et al., 2003). Supercritical fluids have unique properties that support effective extraction. The density of such fluid is much greater than gases and slightly less than organic solvents; the viscosity is close to the typical gases and less than organic solvents. The excellent mass

transfer property of supercritical fluids is easily controlled by temperature and pressure or a modifier. For most SFE, carbon dioxide is the most widely used gas as the solvent since it has a relatively low critical temperature and pressure, is nontoxic, inexpensive, and environmental friendly. Until now, there is no information available

18**on the extraction of** phytochemicals **from** MPL **using supercritical carbon dioxide**

(SC-CO2). In this work, the effect of 1012 temperature, pressure, CO2 flow rates, and extraction times on phytochemical yields has been studied. The results are presented as total phenolic compound (TPC) and total alka- loid content (TAC), and the optimum extraction condition was obtained using 24 full factorial experimental designs. The SC-CO2 extraction of MPL was modeled using density- based correlation (Chrastil, 1982).The correlation of pressure, temperature, and density on TPC and TAC is useful for the design of SC-CO2 extraction process. The identification of phytochemical components in the extract was conducted by high-performance liquid chromatography (HPLC). Materials and Methods Raw Material and Sample Preparation The whole plants of MPL were collected from a local area in Surabaya, Indonesia. They were collected in November 2012 and dried in an oven (Memmert type UNB 500, Büchenbach, Germany) at 323.15 K for 48 h until the moisture content was about 8.5%. The raw materials were then crushed using a domestic grinding mill and then sieved with a vibrating screen (Retsch AS 200, Haan, Germany) to 80=100 mesh. The dried MPL powder was stored in a desiccator contain- ing silica gel to maintain the moisture content at the desired level (8.5%). Chemical Reagents Food grade carbon dioxide (>99.9% purity) was supplied as liquid CO2 by Aneka Gas Pty Ltd, Indonesia. Analytical grade ethanol (>99.5%) and Folin–Ciocalteu reagent were supplied by Merck (Darmstadt, Germany). Gallic acid (98% purity), sodium carbonate, and DPPH (1,1-diphenyl-2- picrylhydrazyl-hydrate) were obtained from Sigma-Aldrich (Singapore, Singapore). Bromocresol green (American Chemical Society reagent with a purity of 95%), mimosine (CAS 500-44-7, 3-hydroxy-4-pyridine-1-yl L-alanine, 98% purity), catechin (CAS 18829-70-4, 98% purity), quercetin (CAS 117-39-5, 2-(3,4-dihydroxyphenyl)-3,5,7-trihydroxy- 4H-1-benzopyran-4-one, 95% purity), and potassium acetate (99% purity) were obtained from Sigma-Aldrich. Supercritical Fluid Extraction Procedure Extraction of phytochemicals from MPL was performed in a SC-CO2 extractor system. The extraction system consists of high-pressure pump (Eldex AA-100-S-2-CE, Napa, CA, USA), a pressure transducer (Druck PTX 611, Boston, MA, USA), and an oven (Memmert type UNB 500, Büchenbach, Germany). All valves and tubes were made from 316 SS Grade (Swagelok, Solon, OH, USA). A known mass of MPL powder (10 g) was placed in SC-CO2 stain- less steel extractor. The heating chamber was heated to the desired temperature (313.15 K, 323.15 K, or 333.15 K). Liquid CO2 was then pumped then into the system until the desired pressure was reached (10–28 MPa) using high-pressure pump with constant CO2 flow rate (6–8 mL=min). When the extraction conditions were reached, the dynamic extraction was started. Finally, the CO2-containing phytochemicals were A. Rahmawati

17**et al. Fig. 1. Schematic diagram**

of the experimental setup. A, deep tube CO2 cylinder; B, high-pressure pump; C, pressure transducer; D, oven; E, extraction column; F, sample collector. introduced into a collector containing a known amount of ethanol (200 mL) and the CO2 was released into the air. A sche- matic of the experimental setup is given in Figure 1. Design of Experiment (DOE) In this study, 24 full factorial of design of experiment (DOE) analysis was used to investigate the effect of significant vari- ables on the response of the TPC. The controlled variables in this study were temperature (T), pressure (P), CO2 flow rate (C), and dynamic extraction time (X). Each of the controlled variables

5**was kept at three levels—high level** (þ1), the **center point (0), and lower level (1)—as listed in Table I.** Based on **the**

experimental design, 33 experiments were carried out in duplicate.

5**The results were analyzed** by **analysis of variance (ANOVA) technique** using MINITAB 16 **.0 software**

to a 95% confidence level. Total Phenolic Content Assay The determination of total phenolic content of the MPL extract was based on Habila et al. (2010) and Cavalcanti et al. (2012) using a Folin–Ciocalteu assay. Briefly, 5 mL of Folin–Ciocalteu reagent (1:10; v=v) was added to 1 mL of the extract to oxidize it. After an interval of 1–5 min, 4

23**mL of sodium carbonate (7.5%, w=v) was added** to **the mixture** solution to stop **the** oxidation, and subsequently **the** solution **was**

allowed to stand in a dark place at room Table I. Variables and levels used in the design of experiment with TPC response Low level Variables (1) Center High level point (0) (þ1) Temperature (C) (T) Pressure (MPa) (P) CO2 flow rate (mL=min) (C) Dynamic extraction time (h) (X) 40 50 12 20 6 7 2 6 60 28 8 10 Supercritical CO2 Extraction of Phytochemical Compounds 1013 temperature for 60 min. The absorbance was measured at a wavelength of 739 nm with a UV=Vis spectrophotometer (Shimadzu mini UV 1240, Kyoto, Japan). Using gallic acid as standard; the TPC was expressed as mg GAE=L of extract. Data are reported for three replications.

16**Total Flavonoid Content** Assay **Total flavonoid content** (TFC) **of** each **extract was determined**

in triplicate by aluminum chloride colorimetric method as modified by Woisky and Salatino (1998). Briefly, 0.5 mL of extract was mixed with

7**0.1 mL of 10% aluminum chloride, 0.1 M potassium acetate, and 2.8 mL of distilled water.** The mixtures **were** allowed to stand **at room temperature for 30 min.** The **absorbance of**

standard and extract solutions was measured at 415 nm using a spectrophotometer (Shimadzu UV mini 1240). The

16**flavonoid content was expressed as** milligram **quercetin equivalents (QE)=**

gram sample. Total Alkaloid Content Assay TAC was determined by titrimetric method (Evan and Partridge, 1952) using bromocresol green as an indicator. About 1 mL of each extract was dissolved in 2.0 mL distilled water and 2 mL buffer, then the mixture solution was trans- ferred into a

24**conical flask, and** three **drops of** bromocresol green indicator **were added to the** solution. **The mixture was titrated with 0.**

005 N H2SO4 until the appearance of a light green color (end point). Each milliliter of acid used is equivalent to 0.00145 g of alkaloid calculated as hyoscyamine. Data are reported for three replications. The calculation of percent- age total alkaloid is given below (Nuhu and Ghani, 2002): malkaloid TAC ¼ msample HPLC Analysis The identification of phytochemicals was conducted on a HPLC column using a modification of the procedure described by Tangendjadja and Wills (1980) and Weisz et al. (2009). The analysis of phytochemicals, namely gallic acid, catechin, mimosine, caffeic acid, epicatechin, p-Coumaric acid, ferulic, and quercetin, was conducted by HPLC (Jasco LC-Net II=ADC, Tokyo, Japan), in conjunction with a dual pump (Jasco PU-2089 plus) and a UV detector (Jasco PU-2077 plus). The column used for separation was an Enduro C-18 250 mm 4.6 mm, with 5-mm particle diameters. The mobile phases were solvent A (water=acetic acid with a volume ratio 97=3) and solvent B (acetonitrile=acetic acid with a volume ratio 97=3) at a constant flow rate of 1 mL= min. The calibration curves were selected at a good detection wavelength of 280 nm. A single injection of the solvent was performed to specify the retention time of the solvent. Result and Discussion Effect of Variables on TPC The extraction of phytochemicals using SC-CO2 extraction is influenced by temperature, pressure, CO2 flow rate, and time. Temperature and pressure significantly influenced the density, viscosity, and diffusivity of SC-CO2, hence affecting the amount of phytochemicals in the extract. The flow rate of CO2 also affected the yield of phytochemicals in some cases. The time for SC-CO2 extraction greatly influenced the yield of phytochemicals extracted prior to equilibrium. The determination of the significant variables affecting the extraction of phytochemicals using SC-CO2 was carried out by performing two-way ANOVA. The statistical DOE was a simultaneous study of several process variables to determine the influence of controlled variables on the amount of phytochemicals extracted (presented as TPC) into a measurable response (Table II). The results were analyzed by ANOVA technique using Minitab 16.0. The estimated constants, coefficients of linear regression, and interaction effects are presented in Table III. In ANOVA,

15**a large regression coefficient and a small p-value indicate a significant effect on the respective response variables**

(Table III). Our results show that the individual effects of pressure and temperature and the Table II. TPC results from the experimental runs of 24 full factorial design Standard order T P C X TPC response 5 2 9 27 32 15 10 31 23 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 18.14 27.20 18.11 27.13 74.94 27.77 27.48 27.69 27.55 4 26 14 16 25 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 74.71 27.32 27.59 74.85 18.10 3 30 1 1 1 1 1 1 1 1 27.18 27.59 7 13 18 17 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 27.52 18.13 27.48 18.05 1 12 11 1 1 1 1 1 1 1 1 1 1 1 1 17.76 74.72 27.18 8 24 33 29 1 1 0 1 1 1 0 1 1 1 0 1 1 1 0 1 74.94 74.72 35.80 18.11 20 19 22 21 28 6 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 74.75 27.18 27.48 17.94 74.73 27.68 1014 Table III. The result of DOE analysis of SC-CO2 extraction of MPL Standard error (SE) Term Effect Coefficient coefficient

10**T P** Constant **T** P **C X T P T C** T **X P** C P **X C X T P** C **T** P **X T** C **X** P **C X T P** C **X**

— 28.4148 28.3364 0.2220 0.0746 18.9769 0.0474 0.0407 0.0768 0.0145 0.0151 0.1155 0.0143 0.0104 0.0576 0.0263 S ¼ 0.287205 R2 ¼ 99.59% % R2 (adjusted) ¼ 98.83% 36.8943 14.2074 14.1682 0.1110 0.0373 9.4884 0.0237 0.0203 0.0384 0.0073 0.0076 0.0577 0.0072 0.0052 0.0288 0.0132 0.05000 0.05077 0.05077 0.05077 0.05077 0.05077 0.05077 0.05077 0.05077 0.05077 0.05077 0.05077 0.05077 0.05077

0.05077 0.05077 737.94 0.000 279.83 0.000 279.06 0.000 2.19 0.043 0.74 0.472 186.89 0.000 0.47 0.646 0.40 0.694 0.76 0.460 0.14 0.888 0.15 0.884 1.14 0.271 0.14 0.889 0.10 0.920 0.57 0.578 0.26 0.798 interaction effect of pressure with temperature are very significant in the case of TPC in MPL extract ($p < 0.01$). There was a slightly significant individual effect of extraction time ($0.04 < p < 0.05$), which indicated that 2 h is sufficient to extract the phytochemicals from MPL solid matrices (Figure 2).

22**There was no significant effect of** CO2 flow **rate on**

TPC as individual variable and in the interaction with other variables ($p > 0.05$) (Figure 2). The Taylor series of expansion can be used to express the regression model with the interaction terms:

11**Y ¼ b0 þ b1x1 þ b2x2 þ b3x3 þ b4x4 þ b12x1x2 þ b13x1x3 þ b14x1x4 þ b23x2x3 þ b24x2x4 þ b34x3x4 þ** b123x1x2x3 **þ**

b124x1x2x4 þ b134x1x3x4 þ b234x2x3x4 þ b1234x1x2x3x4 ð1Þ where Y is TPC; bi is model coefficients; and x1; x2; x3; and x4 are dimensionless coded factors for the controlled variables (i.e., temperature, pressure, CO2 flow rate, and dynamic extraction time, respectively). Neglecting the coefficients of nonsignificant terms at 95% confidence level, the regression becomes: Y ¼ 36:8943 þ 14:2074x1 þ 14:1682x2 þ 0:1110x3 þ 0:0373x4 þ 9:4884x1x2 ð2Þ The yield of phenolic compounds (TPC) as a function of the controlled variables within the experimental range was generated by using the empirical model in Equation (2). The statistical model indicated that the

17**TPC of the** extract **increased with** temperature, **pressure,**

and time. A. Rahmawati et al.

17**Effect of Temperature and Pressure on**

TPC, TFC, and TAC Based on a full factorial design analysis, the significant variables influencing the SC-CO2 extraction of MPL were pressure and temperature. Since the extraction time slightly affected the TPC of extract, further investigation was carried out at constant dynamic extraction time (2 h). The flow rate of CO2 was not a significant variable and was kept at 6 mL=min during the experiments. The TPC of MPL extract significantly increased with pressure (12–28 MPa) at 313.15 K, 323.15 K, and 333.15 K. The lowest TPC was 17.76 mg GAE=g MPL at the lowest operating condition 12 MPa and 313.15 K and the highest TPC was 74.94 mg GAE=g MPL at the highest operating condition 28 MPa and 333.15 K. At a constant temperature, the density of SC-CO2 increased, approaching the density of organic solvent with increasing pressure; as a result, the yield of extraction increases since its ability to dissolve phenolic compounds from the solid matrices increased. The effect of SC-CO2 pressure on TPC of MPL extract was more pronounced at higher temperature (Figure 3). At 313.15 K, the TPC increases from 17.76 mg GAE=g MPL at 12 MPa to 27.18 mg GAE=g MPL at 28 MPa, while at 333.15 K, the TPC increases from 27.21 mg GAE=g MPL at 12 MPa to 74.94 mg GAE=g MPL at 28 MPa. At constant pressure, the amounts of phenolic compounds dissolved in SC-CO2 increased with temperature (Figure 3) because the solute vapor pressures, as well as the diffusivities of both solvent and solute, increased.

20**The effect of pressure and temperature on the**

TFC of MPL extract was similar to TPC since flavonoid is one of the phenolic compounds (Figure 4). The TFC of MPL extract was expressed as milligram QE=g MPL, and the value was lower than TPC, which indicated that other types of phenolic compounds were also extracted. The SC-CO2 also extracted alkaloid compounds even though the result was lower than TPC and TFC. The TAC were presented as milligram hyoscyamine=g MPL. As expected, the lowest TAC was obtained at 313.15 K and 12 MPa (5.24 mg hyoscyamine=g MPL) and the highest was Fig. 2. Normal plot of the standardized effect. Supercritical CO2 Extraction of Phytochemical Compounds 1015 Fig. 3. The effect of pressure and temperature on TPC of MPL extract. obtained at 333.15 K and 28 MPa (14.23 mg hyoscyamine=g MPL). The total content of alkaloids in MPL was obtained from Soxhlet extraction (29.83 mg hyoscyamine=g MPL), which is lower than the total content of phenolics (171.49 mg GAE=g MPL). The HPLC analysis (Figure 5) on the MPL extract from SC-CO2 extraction showed that it contains gallic acid (phenolic compound) and mimosine (alkaloid), and the result from Soxhlet extraction using water ethanol as the solvent contained gallic acid and catechin (phenolic compounds) and mimosine (alkaloid). The

12**solubility of** solutes **in SC-CO2 is**

affected by temperature and pressure and is generally a function of the density of SC-CO2 as the solvent.

12**The effect of temperature on solubility is complex due to the** opposing factors **of**

solute vapor pressure and solvent density. The solute vapor press- ure increased with temperature and causes the solubility to Fig. 4. The effect of pressure and temperature on TFC on MPL extract. Fig. 5. HPLC chromatogram analysis of (a) Soxhlet and (b) SC-CO2 extract from MPL at 280 nm. increase, while the solvent density decreased resulting in decreased solubility. In this study, both factors influenced the yield of phenolic and alkaloid compounds. The yield was also influenced by the polarity of CO2. Although CO2 is nonpolar, it has a large quadrupole that is able to interact with other polar liquids and solids, especially at higher press- ure; the proximity between CO2 and solute molecules is shortened. This leads to the creation of substantial dipole (induced or not)—quadrupole interactions that explain the CO2—philic character of many solutes (de Melo et al., 2012). Density-Based Modeling Density-based modeling is a semiempirical correlation of the logarithm of the solubility of the solute and the density. This approach is quite simple since it does not require knowledge of solute properties and permits easy analysis of correlations with existing solubility data. The Chrastil model was based on the formation of solvation complex model between the solvent and the solute molecules at 1016 an equilibrium state (Chrastil, 1982): A þ kB $ ABk ð3Þ This model assumes that one molecules of solute (A) is associated with k molecules of solvent (B) to form a solva- tion complex (ABk) at equilibrium. The Chrastil model is a model which correlates the

18**solubility of a solute in a** super- critical **solvent** to **the** density **of**

solvent and temperature process (Chrastil, 1982): s ¼ qk exp þ b a T ð4Þ where s indicates the concentration of solute in SC-CO2 (g=L), q indicates the density of SC-CO2 (g=L), k indicates the number of CO2 molecules performed in the complex solute of solvent, a is

6**a function of** the **enthalpy of solvation** (DHsolvation) (J=mol) **and enthalpy of vaporization**

(DHvaporation) (J=mol), and b is

6**a function of** the **average association number that depends on the molecular** weight **of** the **solute and solvent. The** mathematical form **of** a as **a**

function of enthalpy of vaporization and solvation is a¼ DHsolvation þ DHvaporation DH R ¼ R ð5Þ

22**where R is** an ideal **gas constant and** DH (J=mol) **is the**

enthalpy of solution or reaction of the substance in supercri- tical fluid. The calculation of constants for the semiempirical density-based correlation (Chrastil) was conducted using multivariable nonlinear regression analysis. The quality of all data correlations was quantified by the sum of squared error, defined as follows: "P sexp scal 2 #0:5 SSE ¼ N ð7Þ Fig. 6. The total phenolic compounds extracted at supercritical conditions and plots of Chrastil model. A. Rahmawati et al. Fig. 7. The total flavonoid compounds extracted at supercritical conditions and plots of Chrastil model. where sexp is the actual data of TPC of the MPL extract, scal is the calculated data of TPC of MPL extract, and N is the number of experimental data. The multivariable non- linear regression is applied with the iteration curve-fitting procedure using SigmaPlot 12.0. An initial estimation for each parameter was provided, and then iteration calculation was conducted until the convergence criteria for each final parameter were satisfied. Figures 6–8 show the two-dimensional graphs of experimental data of phenolic, flavonoid, and alkaloid extracted in SC-CO2 and the plot using Chrastil model. These figures clearly show that the model by Chrastil represents the data well. The fitting parameters of Chrastil model obtained from nonlinear least-square method are summarized in Table IV. The value of k indicates the number of CO2 molecules (B) associated with the solute (A) to form a solvation complex (ABk). The enthalpy of solution for phenolic, flavonoid, and alkaloid in Chrastil model was 50,263.15 J=mol, 53,390.21 J=mol, and 53,286.28 J=mol, respectively. Fig. 8. The total alkaloid compounds extracted at supercritical conditions and plots of Chrastil model. Supercritical CO2 Extraction of Phytochemical Compounds 1017 Table IV. Parameters of Chrastil model of phytochemical com- pounds from MPL Chrastil model Parameter Phenolic Flavonoid Alkaloid k0 1.5512 b0 7.3493 a0 6045.24 SSE 0.0918 1.5660 7.7469 6461.34 0.0222 1.5853 6.5755 6407.84 0.0094 Conclusion According to the DOE results, the significant variables for the SC-CO2 extraction of phytochemicals from MPL were temperature and pressure. The maximum amounts of TPC, TFC, and TAC were 74.83 mg GAE=g dry MPL, 30.93 mg QE=g dry MPL, and 14.23 mg HE=g dry MPL, respectively. The experimental data of phenolic, flavonoid, and alkaloid extracted from MPL were correlated by density-based mod- els. The Chrastil model can represent the experimental data very well. From the identification of phytochemicals by HPLC, the SC-CO2 extract is more selective than the Soxhlet extraction method, in which gallic acid and mimosine were detected in the SC-CO2 extract of MPL. Acknowledgments The authors thank

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