Turnitin Originality Report

CEJ by Suryadi Ismadji

From Hippo (Hippo-03)

Processed on 21-Aug-2018 14:33 WIB
ID: 991773345
Word Count: 6746

Similarity Index	Similarity by Source	
20%	Internet Sources:	13%
	Publications:	20%
	Student Papers:	5%

sources:

1 1% match (publications)
Wojciech Plazinski, Wladyslaw Rudzinski, Anita Plazinska. "Theoretical models of sorption kinetics including a surface reaction mechanism: A review", Advances in Colloid and Interface Science, 2009

2 1% match (publications)
Md. Ahmaruzzaman. "Adsorption of phenolic compounds on low-cost adsorbents: A review", Advances in Colloid and Interface Science, 2008

3 1% match (publications)
Kurniawan, A.. "Removal of basic dyes in binary system by adsorption using rarasaponin-bentonite: Revisited of extended Langmuir model", Chemical Engineering Journal, 20120501

4 1% match (publications)
Prahas, D.. "Activated carbon from jackfruit peel waste by H"3PO"4 chemical activation: Pore structure and surface chemistry characterization", Chemical Engineering Journal, 20080701

5 1% match (publications)
A. Dąbrowski, P. Podkościelny, Z. Hubicki, M. Barczak. "Adsorption of phenolic compounds by activated carbon—a critical review", Chemosphere, 2005

6 1% match (Internet from 05-Sep-2013)
http://rsce2012.its.ac.id/search.php

7 1% match (publications)
Lan Zhang, Jiying Wei, Xuan Zhao, Fuzhi Li, Feng Jiang, Meng Zhang, Xuzhou Cheng. "Competitive adsorption of strontium and cobalt onto tin antimonate", Chemical Engineering Journal, 2016

8 1% match (publications)
İlhami Gülçin, Zübeyr Huyut, Mahfuz Elmastaş, Hassan Y. Aboul-Enein. "Radical scavenging and antioxidant activity of tannic acid", Arabian Journal of Chemistry, 2010

1% match (Internet from 06-Nov-2014)

9 http://widyamandala.academia.edu/SuryadiIsmadji/Papers

10 1% match (publications)
SpringerBriefs in Molecular Science, 2015.

11 1% match (Internet from 13-Oct-2016)
http://www.omicsonline.com/open-access/subcritical-water-extraction-of-xanthone-from-mangosteen-garcinia-mangostana-linn-pericarp-2090-4568-1000117.php?aid=44509

12 1% match (publications)
Alfin Kurniawan, Hogiartha Sutiono, Nani Indraswati, Suryadi Ismadji. "Removal of basic dyes in binary system by adsorption using rarasaponin–bentonite: Revisited of extended Langmuir model", Chemical Engineering Journal, 2012

13 1% match (Internet from 20-Jul-2018)
http://shura.shu.ac.uk/21636/8/Chen-AmmoniaRemovalFromWater%28AM%29.pdf

14 1% match (publications)
Yujia Liu, Danyang Ying, Luz Sanguansri, Yanxue Cai, Xueyi Le. "Adsorption of catechin onto cellulose and its mechanism study: Kinetic models, characterization and molecular simulation", Food Research International, 2018

15 1% match (Internet from 01-Dec-2015)
http://www.jeaconf.org/UploadedFiles/Document/897510bf-1f90-4689-968c-40d7b71c0011.pdf

16 1% match (publications)
Chandra, T.C.. "Adsorption of basic dye onto activated carbon prepared from durian shell: Studies of adsorption equilibrium and kinetics", Chemical Engineering Journal, 20070301

17 1% match (publications)
Keith K. H. Choy, John F. Porter, Gordon McKay. "Langmuir Isotherm Models Applied to the Multicomponent Sorption of Acid Dyes from Effluent onto Activated Carbon", Journal of Chemical & Engineering Data, 2000

18 < 1% match (student papers from 20-Mar-2018)
Submitted to Georgia State University on 2018-03-20

19 < 1% match (Internet from 21-Jul-2018)
https://www.cheric.org/research/tech/periodicals/searchresult.php?articlesearch=Soetaredjo+FE&searchtype=author

20 < 1% match (Internet from 10-Aug-2018)
https://www.yumpu.com/en/scalaris

21 < 1% match (publications)
Bin Tang, Yanwen Lin, Ping Yu, Yunbai Luo. "Study of aniline/ε-caprolactam mixture adsorption from aqueous solution onto granular activated carbon: Kinetics and equilibrium", Chemical Engineering Journal, 2012

22 < 1% match (publications)
Hokkanen, Sanna, Eveliina Repo, Song Lou, and Mika Sillanpää. "Removal of arsenic(V) by magnetic nanoparticle activated microfibrillated cellulose", Chemical Engineering Journal, 2015.

23 < 1% match (publications)
Tabaraki, Reza, and Ashraf Nateghi. "Multimetal biosorption modeling of Zn2+, Cu2+ and Ni2+ by Sargassum ilicifolium", Ecological Engineering, 2014.

24 < 1% match (publications)
Borneman, Z.. "Selective removal of polyphenols and brown colour in apple juices using PES/PVP membranes in a single ultrafiltration process", Separation and Purification Technology, 20010301

25 < 1% match (student papers from 11-Dec-2013)
Submitted to The University of Manchester on 2013-12-11

26 < 1% match (Internet from 28-Jan-2017)
https://www.omicsonline.org/2157-7048/images/2157-7048-4-169-t003.html

27 < 1% match (publications)
Yesi, Fiona Patricia Sisnandy, Yi-Hsu Ju, Felycia Edi Soetaredjo, Suryadi Ismadji. "Adsorption of Acid Blue 129 from Aqueous Solutions onto Raw and Surfactant-Modified Bentonite: Application of Temperature-Dependent Forms of Adsorption Isotherms", Adsorption Science & Technology, 2010

28 < 1% match (publications)
Putra, E.K.. "Performance of activated carbon and bentonite for adsorption of amoxicillin from wastewater: Mechanisms, isotherms and kinetics", Water Research, 200905

29 < 1% match (publications)
Andi Kurniawan Rahardjo, Maria Josephine Jeannette Susanto, Alfin Kurniawan, Nani Indraswati, Suryadi Ismadji. "Modified Ponorogo bentonite for the removal of ampicillin from wastewater", Journal of Hazardous Materials, 2011

30 < 1% match (publications)
SHIRLENE M. ANTHONYSAMY. "BROWNING OF SAGO (METROXYLON SAGU) PITH SLURRY AS INFLUENCED BY HOLDING TIME, pH AND TEMPERATURE", Journal of Food Biochemistry, 5/2004

31 < 1% match (student papers from 15-Apr-2014)
Submitted to Stockholms universitet on 2014-04-15

32 < 1% match (Internet from 15-Dec-2015)
http://arjournals.org/index.php/ijpm/article/download/113/107

33 < 1% match (publications)
S.B. REVANAPPA. "PHENOLIC ACID PROFILES AND ANTIOXIDANT ACTIVITIES OF DIFFERENT WHEAT (TRITICUM AESTIVUM L.) VARIETIES : PHENOLIC ACID PROFILES IN WHEAT VARIETIES", Journal of Food Biochemistry, 05/2010

34 < 1% match (publications)
Muhammad, , T.G. Chuah, Yunus Robiah, A.R. Suraya, and T.S.Y. Choong. "Single and binary adsorptions isotherms of Cd(II) and Zn(II) on palm kernel shell based activated carbon", Desalination and Water Treatment, 2011.

35 < 1% match (Internet from 24-May-2018)
https://idus.us.es/xmlui/bitstream/handle/11441/27154/Tesis%20SMG_revFJGO.pdf?isAllowed=y&sequence=4

36 < 1% match (Internet from 24-Sep-2014)
http://www.cyut.edu.tw/~ijase/2010/7(2)/2_016003.pdf

37 < 1% match (Internet from 21-Nov-2017)
http://upcommons.upc.edu/bitstream/handle/2117/96251/TJLV1de1.pdf?isAllowed=y&sequence=1

38 < 1% match (Internet from 03-Jan-2016)
http://file.scirp.org/Html/3-2200837_56183.htm

39 < 1% match (publications)
Felycia Edi-Soetaredjo, Suryadi Ismadji, Yi-Hsu Ju. "Measurement and modeling of epicatechin solubility in supercritical carbon dioxide fluid", Fluid Phase Equilibria, 2013

40 < 1% match (publications)
Weiwei Cheng, Guoqin Liu, Xuede Wang, Lipeng Han. "Adsorption Removal of Glycidyl Esters from Palm Oil and Oil Model Solution by Using Acid-Washed Oil Palm Wood-Based Activated Carbon: Kinetic and Mechanism Study", Journal of Agricultural and Food Chemistry, 2017

41 < 1% match (publications)
Edi-Soetaredjo, Felycia, Suryadi Ismadji, and Yi-Hsu Ju. "Measurement and modeling of epicatechin solubility in supercritical carbon dioxide fluid", Fluid Phase Equilibria, 2013.

< 1% match (publications)

42 Jacobsen, S.. "Single-step reactive extrusion of PLLA in a corotating twin-screw extruder promoted by 2-ethylhexanoic acid tin(II) salt and triphenylphosphine", Polymer, 200004

43 < 1% match (Internet from 24-Sep-2017)
https://dr.ntu.edu.sg/bitstream/handle/10220/19432/Hybrid%20Multi-Walled%20Carbon%20Nanotubes-Alginate-Polysulfone%20Beads%20for%20Adsorption%20of%20Bisphenol-A%20from%20Aqueous%20Solution.pdf?isAllowed=y&sequence=1

44 < 1% match (publications)
Kosasih, A.N.. "Sequestering of Cu(II) from aqueous solution using cassava peel (Manihot esculenta)", Journal of Hazardous Materials, 20100815

paper text:

35**Chemical Engineering Journal** 231 **(2013)** 406–413 **Contents lists available at ScienceDirect Chemical Engineering Journal journal homepage: www.elsevier.com/locate/cej**

17**Recovery of catechin and epicatechin from sago waste effluent: Study of kinetic and binary adsorption isotherm studies Felycia Edi Soetaredjo** a, **Suryadi Ismadji** b, **Shella Permatasari Santoso** a, **Ong Lu Ki** b, **Alfin Kurniawan**

b,

6**Yi-Hsu** Ju **a,⇑** a **Department of Chemical Engineering, National Taiwan University of Science and Technology, 43, Sec 4., Keelung Rd., Taipei, Taiwan b Department of Chemical Engineering, Widya Mandala Surabaya Catholic University, Kalijudan 37, Surabaya 60114, Indonesia**

highlights

19**Recovery of catechin and epicatechin from sago waste effluent.** Supercritical CO2 desorption **of** catechin **and**

epicatechin from activated carbon. Adsorption of catechin and epicatechin in binary system.

31**article info Article history: Received** 27 May **2013 Received in revised form** 16 **July 2013 Accepted 17 July 2013 Available online 26 July 2013 Keywords:**

Catechin Epicatechin Binary adsorption Supercritical CO2 extraction abstract The recovery of valuable phenolic compounds (catechin and epicatechin) from sago waste effluent by activated carbon adsorption was studied in this work. Single and binary component of catechin and epi- catechin was studied in static mode using synthetic effluent.

12**Langmuir model was** applied **to evaluate** the **adsorption equilibrium data in** single **system.** In binary system, **the** modified **extended Langmuir model**

by inclusion of surface coverage was used to represent the adsorption data. All experimental data were well represented either by Langmuir model or its modified form. For kinetic study, it was found that pseudo-second order model represents the experimental data better than pseudo-first order model. Adsorption study using real sago waste effluent indicated that activated carbon adsorption process is promising for recovery of valuable phenolic compounds. Supercritical CO2 extraction of phenolic com- pounds from activated carbon resulted in low recovery. The addition of ethanol as co-solvent significantly enhanced the desorption of catechin and epicatechin from activated carbon. Ó 2013 Elsevier B.V. All rights reserved. 1. Introduction Phenolic compounds are universally distributed in plants such as leave, trunk, root, seed, and fruit. They are usually are bounded within plant tissues. Phenolic compounds as antioxidants are be- lieved

33**to contribute to health benefits through several possible mechanisms** such as **by quenching free radicals, chelating** transi- tion **metals, reducing peroxides and stimulating antioxidative** en- zyme **defense.**

32**Natural antioxidants are known to exhibit a wide range of biological effects including** anti-bacterial, **antiviral, anti- inflammatory, anti-allergic, antithrombotic and vasodilatory**

activ- ities [1].

8**The antioxidant activity of phenolic compounds is mainly attributed to their redox properties, which allow them to act as reducing agents, hydrogen donors and quenchers of singlet oxygen. In addition, they may also possess metal** chelating **properties.**

Phe- nolic compounds possess aromatic structure along with hydroxyl ⇑ Corresponding author. Tel.: +886 (2) 2737 6611; fax: +886

18**(2) 2737 6644. E-mail address: yhju@mail.ntust.edu.tw (Y.-H. Ju).** 1385-8947/$ - **see front matter** Ó 2013 **Elsevier** B.V. **All rights reserved. http://dx.doi.org/10.1016/j.** cej **.2013.**

07.048 substituent

11**which enable them to protect human tissues from damages caused by oxygen or free radicals, and consequently** re- duce **the risk of** various **diseases, and offer beneficial** effect **against** cancer, **cardiovascular disease, diabetes, and Alzheimer's disease**

[2–7]. In starch extraction from sago pith, the sago pith slurry during sedimentation is browning due to the

30**oxidation of phenolic** com- pounds **by the polyphenol** oxidase (PPO) **enzyme** [8]. **Phenolic compounds are located mainly in vacuoles** while the **PPO**

is found in plastids. Browning is initiated with the disruption of cell due to

30**cutting or bruising** and the **compartmentalization is lost. The**

pri- mary phenolic compounds related to browning are DL-epicatechin and D-catechin [8,9]. Pei-Lang et al. [9] reported that the content of phenolic compounds in sago pith depends on its growth stages and height; and the amount of phenolic compound extracted is less than 1%. Recovery of phenolic compounds from the waste effluents is beneficial, but it is a challenging process. Adsorption is efficient for removing

2**organic matter from waste effluents. Activated carbons are the most widely used adsorbents due to their** high **adsorption** capacities **for organic pollutants** which are

2**related to their high surface area, pore volume and porosity. The adsorption**

capacities also strongly depend on the activation method and the nature of source materials.

2**Activated carbon is an effective** adsor- bent **for organic compounds removal, especially for phenolic**

com- pounds [10–15].

40**However, the** utilization **of activated carbon** as adsorbent **in the adsorption of**

epicatechin and catechin has never been reported. The focus of this work was to evaluate the adsorption perfor- mance of activated carbon for the recovery of epicatechin and cat- echin in single and binary component systems. The surface area was obtained based on Brunauer, Emmet, and Teller (BET) mea- surements, while the pore volume and pore diameter of activated carbon were calculated from the adsorption experimental data using density functional theory (DFT). The effect of various vari- ables, such as initial adsorbate concentrations, adsorption time and mass of adsorbent were studied using batch equilibrium tech- nique.

27**Pseudo first and second order** kinetic models **were used to correlate the adsorption** dynamic or **kinetic data. The**

10**adsorption equilibrium data** was evaluated **using Langmuir model**

for single component, while

9**modified extended-Langmuir model with** incor- poration **of solute's selectivity factor**

was used to study the corre- lation [16]. The feasibility and adsorption performance of activated carbon in recovering epicatechin and catechin was also tested in real effluent. 2. Experimental section

39**2.1.** Chemicals **HPLC grade (?)-epicatechin (purity**

98 wt.%) and (+)-catechin hydrate (purity P

39**98 wt.%)** were **obtained from** Sigma **Aldrich** (Sin- gapore) **and was used without any further purification.**

Pellet acti- vated carbon (0.5–1.5) produced by Norit (The Netherland) was used as the adsorbent. Double distilled water was used in adsorp- tion experiments. 2.2. Characterizations of activated carbon Since

4**structural heterogeneity of activated carbon plays an important role** during **adsorption**

therefore it is crucial to charac- terize the pore structure of activated carbon.

3**Pore structure of the** activated carbon **was** characterized **by nitrogen** sorption tech- nique **in a Quadrasorb SI at**

boiling point of nitrogen gas (?196 °C). Prior to the nitrogen sorption measurement, the activated carbon was degassed in vacuum condition at 200 °C for 48 h. The measure- ment of the nitrogen sorption isotherms (adsorption and desorp- tion) were conducted

44**over a relative pressure (p/po)** of **0.** 001– **0.** 990. **The BET surface area** of the carbon **was determined by**

stan- dard BET equation at a

28**relative pressure of 0.06–0.3.** Total of **pore** volume **of the** activated carbon **was**

evaluated at the highest rela- tive pressure (0.990).

4**At this relative pressure all pores**

within the activated carbon were already completely filled with nitrogen gas.

4**Pore size distribution of the** carbon **was** determined **by** standard **DFT method** with **medium regularization.**

2.3. Static adsorption experiments The adsorbate solution was prepared by mixing a known amount of adsorbate ((?)-epicatechin or (+)-catechin hydrate) with double distilled water to produce the desired initial concen- trations (100–150 mg/L). The adsorption was carried out isother- mally in static mode at 303.15 K. A fixed amount of activated carbon was added to

16**a series of 250 mL** glass-stoppered **flasks each filled with 100 mL**

diluted solutions. The glass stoppered flasks were then put

38**in a thermostatic** shaker **bath at** 120 **rpm for** 1 **h (the**

equilibrium condition was achieved in about 40 min). The adsorption isotherm for binary systems of epicatechin and catechin was also obtained isothermally at 303.15 K, using the same proce- dure as for the single component adsorption. To study the adsorption kinetics, a series of 250 mL glass-stop- pered flasks contained 100 mL of phenolic compound solution with certain initial concentration (100–250 mg/L) were prepared. Sub- sequently,

34**a known amount of activated carbon** (0.2 g) **was added into** each glass-stopper **flask. The**

glass-stopper flasks were placed in thermostatic water-bath shaker for 150 min at 303.15 K. During kinetic experiments, at intervals of 10 min, one of glass-stopper flask was taken and the concentration of phenolic compound was analyzed. The adsorption of phenolic compounds from waste effluent was carried out using waste water from sago starch extraction. Briefly, sago pith was pulverized into coarse powder and soaked in water (1 h), and then filtered using Buchner funnel to obtain the waste effluent. One gram of activated carbon was added to 250 mL glass-stopper

16**flasks each filled with 100 mL**

waste effluent. The glass stopper flask was placed

38**in a thermostatic** shaker **bath at** 120 **rpm for** 1 **h. The**

initial and equilibrium concentrations of the single and bin- ary solutions and waste effluent were analyzed

41**using a HPLC (Jasco HPLC PU-2089 plus) with a UV–vis detector (UV-2077 plus)**

set at 280 nm. Separations were achieved in an Enduro C18, 5 lm, 250 4.6 mm. The

21**flow rate** was **0.** 8 **ml/min. The mobile phase used was**

isocratic elution comprised of 3% acetic acid in double distilled water (60%) and acetonitrile (40%) for a total running time of 10 min. The contents of epicatechin and catechin were deter- mined using calibration curves. The

14**effect of pH on the adsorption of** epicatechin and **catechin was** investigated **in**

single component systems. It was found that as the initial solution pH was reduced to pH 3, the solution concen- tration decreased up to 50% of the initial concentration. When the initial solution pH was increased to pH 10, the solution concentra- tion also decreased up to 60–70% of the initial concentration. This is due to oxidation of the phenolic compounds. Therefore,

40**the adsorption of** epicatechin **and** catechin **in single and binary** compo- nent **were**

carried out at pH 7, which is similar to the waste effluent pH. 2.4. Calculations The amounts of epicatechin and catechin adsorbed onto acti- vated carbon in single component system were calculated using the following equation: qe ¼ ðC0 ? CeÞ m V ð1Þ where C0

25**and Ce are the initial and equilibrium concentration of** phenolic compounds **in** the **liquid phase (mg/L), respectively, V is the volume of** phenolic compounds **solution (L),** qe **is the** amount **of**

metal ions adsorbed per unit of biosorbent mass (mg/g), and m is the mass of activated carbon

23**(g). The equilibrium concentration of** phenolic compound **i**

species (Ce,i) in binary mixture was calcu- lated

23**by the same manner using following mass balance equation:** qe **;i** ¼ ðC0 **;i** ? Ce;iÞ **m**

V ð2Þ where qe

23**,i is the equilibrium amount of** solute i in **the adsorbed phase** (mg/g), C0,i and Ce

3**,i are the initial and equilibrium** concentra- tion **of** solute **i in the** liquid phase (mg **/L), respectively.** All data are **the** average results **of**

triplicates experiments. 408 F.E. Soetaredjo et al. / Chemical Engineering Journal 231 (2013) 406–413 3. Results and discussion 70 3.1. Activated carbon characterization 60 Analysis of nitrogen sorption isotherms and DFT-pore size dis- tribution of the activated carbon used in this study indicates that the NORIT carbon possess both micropore and mesopore structures with micropore structure more dominant than mesopore as indi- cated by the rapid filling of pores at low relative pressure and fol- lowed by the gradual increase of adsorption at moderate relative pressure (S1). The existence of the mesopores

28**is indicated by the presence of hysteresis** loop **between adsorption and desorption**

isotherms, an indication of capillary condensation in the mesop- ores. The

13**BET surface area, micropore volume, and total pore** vol- ume **of the**

activated carbon (NORIT nrs carbon Ea 0.5–1.5) were 890 m2/g, 0.431 cm3/g, and 0.506 cm3/g, respectively. The pore distribution of activated carbon represents a model of carbon internal structure which represents non-interacting of reg- ularly shape model pores and

4**complex void spaces within the** car- bon. **The pore size distribution** of the carbon **is**

one of its most important characteristics of the carbon in studying adsorption equilibrium and kinetic. The

37**pore size distribution of the activated carbon** confirms that the **carbon**

possesses both micropore and mesopore structure (S2). 50 40 30 qe (mg.g-1) 20 10 0 0 20 25 20 qe (mg.g-1) 15 10 (a) 40 60 Ce (mg.L-1) 3.2. Adsorption isotherms 5 (b) The isotherm shape can be used as quasi-qualitative informa- tion

5**on the nature of the solute-surface interaction.** In **the** case **of** 0 **adsorption of** phenolic compounds **from** water, it is characterized 40 50 60 70 80 90 100 by initial region (high uptake at low concentration); and Langmuir Ce (mg.L -1) model is best fitted for this case [17,18]. Based on the classification proposed by Giles et al. [17,18], the curve shape in Fig. 1 is class L2 Fig. 1. Adsorption isotherm of catechin and epicatechin onto activated carbon NORIT nrs carbon Ea 0.5–1.5 and Langmuir isotherm model (solid lines). according to the Giles classification

5**suggests that the aromatic ring** adsorbed is **parallel to the surface and no strong competition exists between the adsorbate and the solvent to occupy the adsorption**

time consuming and much more difficult to obtain. Therefore it sites [12].

22**Langmuir isotherm is characterized by a plateau curve**

will be convenient if the multi-component isotherm can be con- which indicates that equilibrium has been achieved. The Langmuir structed based on single adsorption isotherms. Various models isotherm was originally developed for describing gas adsorption have been developed for this purpose and one of the most widely phenomena but it has been extensively used for the correlation used is the extended Langmuir and its modified forms [16,22,23]. of

22**adsorption equilibrium data of various solute-sorbent** interac- **The**

extended Langmuir model for a two-component system can tions in liquid phase [19–21]. The Langmuir isotherm model is gi- be expressed as: ven as follows: qe ¼ qm 1 þKLKLCeCe ð3Þ qe;1 1 þ KqLm;1;C1Ke;1L;1þCKe;1L;2Ce;2 ¼ ð4Þ where qm

22**is the maximum** adsorption **capacity of the solid** (mg **/g)** corresponding **to monolayer surface coverage**

and KL is Langmuir qe;2 ¼ 1 þ KqLm;1;C2Ke;1L;2þCKe;2L;2Ce;2 ð5Þ constants of adsorption affinity (L/mg). where qm,i and Ce,i are the amount adsorbed and equilibrium con- Fig. 1 shows that the adsorption equilibrium data of phenolic centration of component i, respectively. The parameters qm,i and compounds are well fitted by the Langmuir isotherm model and KL,i are the adsorption capacity of component i in the adsorbent the correlation coefficients for epicatechin and catechin are 0.97 and adsorption affinity of component i, respectively. All these and 0.94, respectively. The maximum adsorption capacity (qm) of parameters are obtained from single adsorption isotherms. catechin (73.56 mg/g) obtained by fitting calculation using Sigma- In a multi-component system, competition for vacant adsorp- Plot 12.0 is higher than that of epicatechin (29.04 mg/g). The tion sites in the surface of adsorbent occurs not only between adsorption affinity parameter (KL) of catechin and epicatechin ob- the same molecules but also between different adsorbate species. tained are 0.1257 L/mg and 0.041 L/mg, respectively. Higher This competition may act as the controlling factor for the adsorp- adsorption affinity parameter of catechin than epicatechin indi- tion process [16,23] and such phenomenon is not captured by ori- cates that catechin is easier adsorbed onto the surface interior of ginal extended Langmuir model. Recently, Kurniawan et al. [23] the carbon. and Soetaredjo et al. [16] modified the extended Langmuir model Industrial Adsorption process usually involves multi-compo- by including fractional surface loading or coverage [23] and selec- nent, therefore, information on adsorption equilibria in multi-com- tivity factor [16]. Since both epicatechin and catechin are organic ponent system is required for engineering design of the adsorption compounds, the fractional surface loading approach by Kurniawan process. Compare to the single component adsorption, reliable et al. [23] is more

10**suitable to represent the** binary **adsorption** data **experimental**

multi-component adsorption isotherm data are more of this study. Since

37**competition between** catechin **and** epicatechin **molecules for** vacant **adsorption sites in the** activated **carbon** occurred during **the**

adsorption process, catechin and epicatechin were adsorbed in certain amount, and the adsorption capacity of the activated for both adsorbates (binary) can be written as qm;bin ¼ qm;1h1 þ qm;2h2 ð6Þ Here h1 and h2 are the fractional loading of catechin and epicatechin, respectively, and h1 + h2 = 1. The adsorption affinity parameter in Langmuir equation represents

27**how strong** an **adsorbate molecule is attracted** onto **the surface** of **the**

adsorbent. In a binary system, the competition between different molecules weakens the adsorp- tion potential

36**of the adsorbate** toward **the surface of adsorbent,** therefore, **the**

affinity parameter of binary compounds can be writ- ten as: KL;1ðbinÞ ¼ KL;1 exp ? hh21 ð7Þ KL;2ðbinÞ ¼ KL;2 exp ? hh12 ð8Þ where KL,1(bin) and KL,2(bin) are the affinity parameter of catechin and epicatechin

12**in binary system,** respectively. **The** final expressions **of extended Langmuir model**

with inclusion of fractional loading con- cept are qe;1 ¼ 1 þ KLq;1mðb;bininÞCKeL;;11ðþbinKÞCLe;2;1ðbinÞCe;2 ð9Þ qe;2 ¼ 1 þ KLq;1mðb;bininÞCKeL;;12ðþbinKÞCLe;2;2ðbinÞCe;2 ð10Þ The applicability of the modified extended Langmuir model was tested against the liquid phase binary adsorption data of catechin and epicatechin. The parameter è1 and è2 were obtained by the simultaneously fitting of Eqs. (9) and (10) using nonlinear least square method by minimizing the following objective function 2 #0:5 SSE ¼ "P ðqexp ? qcalÞ n ð11Þ where qexp and qcal are the actual amount and the calculated values of catechin and epicatechin adsorbed by activated carbon, respec- tively; n is the total number of experimental data used in the fitting procedure. Fig. 2 shows the 3D plots of binary adsorption of catechin and epicatechin at various initial concentrations and the model calcu- lated by Eqs. (9) and (10). The fitted parameters h1 and h2 with cal- culate values of qm,(bin), KL,1(bin), and KL,2(bin) are summarized in Table 1. In this work an improvement was made in the fitting pro- cedure of modified extended Langmuir model by Kurniawan et al. [23]. Both Eqs. (9) and (10) were simultaneously fitted by nonlin- ear least square method with h1 and h2 as the fitted parameters, with boundary condition of the fitting procedure as h1 + h2 = 1. It is obvious that this fitting procedure

34**improved the** applicability **of the modified** extended **Langmuir model** in correlating **the** binary experimental data **of**

catechin and epicatechin as indicated in Fig. 2. The maximum amount of adsorbates adsorbed in binary system (qm(bin)) was lower than in single system (qm) due to the effect of molecular competition between both adsorbate molecules. This competition was accommodated in the modified extended Lang- muir by taking into account of fractional loading coverage. The fractional coverage of catechin (h1) and the maximum amount adsorbates adsorbed (qm(bin)) decreased with increasing initial epi- catechin concentration as shown in Table 1. This result is consis- (a) g/ g m,50 d r b e osd 40 s a dnu 30 op oc 20 m icl n o hepf 10 80 6070 g/L otnuo 0 a m 90 80 30 4050atechin,m Ce epicatech7i0n, mg/L60 20 Ce c 50 10 adsorption experimental data of catechin adsorption experimental data of epicatechin modified extended Langmuir model (b) g/g m,35 d b e o r sd 30 a d s un 25 p o m o c 20 cil n o he 15 p 80 f o / L t g nu 10 o 40 m 140 a 130 120 Ce epicatechin11,m0g/L 20 100 C e c6at0echin,m 90 adsorption experimental data of catechin adsorption experimental data of epicatechin modified extended Langmuir model Fig. 2. 3D plot of binary adsorption data and modified extended Langmuir model. (a)

24**Catechin 100 mg/L + epicatechin 100 mg/L,** (b) catechin **100 mg/L** + epicatechin 150 **mg/L.**

tent

3**with adsorption characteristics in single** adsorption **system that the** adsorption capacity **of** epicatechin **is** less **than**

that of cat- echin, therefore increasing the concentration of epicatechin in bin- ary system caused the interaction of adsorbate molecules to become even higher and reduce the value of qm(bin) . 3.3. Adsorption kinetics The effective design of an adsorption system requires informa- tion about the

1**rate at which the** organic compounds **removal takes place in a given** sorbent **/solution system.**

Several adsorption kinetic

10**models have been developed to describe the** adsorption **kinetics** in liquid phase. **The**

development of adsorption kinetic models mostly is based on fundamental approach to interfacial kinetics and its modifications [24]. Several models which have been developed 410 F.E. Soetaredjo et al. / Chemical Engineering Journal 231 (2013) 406–413 Table 1 Fitted and calculated parameters for catechin and epicatechin adsorption at several initial concentrations. Phenolic compounds mixture Fitted parameter Calculate parameter R2 h1 h2 qm,(bin), mg/g KL,1(bin), L/mg KL,2(bin), L/mg

24**Catechin 100 mg/ L + Epicatechin 100 mg/L** Catechin **100 mg/ L** + Epicatechin 150 **mg/L**

0.632 0.368 0.524 0.476 57.1 0.0699 52.2 0.0499 0.0075 0.0139 0.998 0.997 based on this approach are Langmuir kinetics, first and second or- der, reversible first and second order, and irreversible reactions [24]. Another approach which is used to develop the kinetic mod- els is based on assuming that surface reaction is the rate control- ling step in sorption. Many simple and compact models have been proposed based on this approach and the famous and

43**widely used models** are **pseudo-first** (Lagergren) **and pseudo-second order** [24]. **Pseudo-first order model** which **is** also known **as**

Lagergren equation is the earliest empirical model describing the rate of adsorption in a liquid phase system [25]. The differential form of this model is dqðtÞ dt ¼ k1ðqe ? qðtÞÞ ð12Þ The

36**integration of Eq.** (12) **with initial** conditions: **q(t) = 0 at t = 0**

gives qðtÞ ¼ qeð1 ? expð?k1tÞÞ ð13Þ where

21**q(t) is the amount of** adsorbate **adsorbed at time t** (mg **/g), and** k1 **is the rate constant (1/min).**

Pseudo-second order was first proposed by Blanchard et al. [26]. This model assumes that

1**direct adsorption/desorption (seen as chemical reaction)** is **the** rate controlling step in the overall sorp- tion kinetics [24], and has the differential form as dqdðttÞ ¼ k2ðqe ? qðtÞÞ2 ð14Þ Here

16**k2 is rate constant of pseudo-second order (g/mg min).** Inte- gration of **Eq.** (14) gives **the**

following result qðtÞ ¼ qe 1 þk2kq2etqet ð15Þ To obtain the parameters of pseudo-first and pseudo-second or- der equations, non-linear least square method was employed by fitting the kinetic experimental data with Eqs. (13) and (15)

12**and the results are** summarized **in Table** 2. **Figs. 3 and 4**

present the plots of catechin and epicatechin uptake as the function of time, respectively. The

13**symbols represent the** kinetic experimental **data while the solid lines represent**

the pseudo-first and pseudo-second order kinetic models. As shown in Table 2, the values of R2 ob- tained from the fitting of kinetic experimental data using both models were similar, however the pseudo-second order kinetic model represents kinetic experimental data better than the pseu- do-first order since the predicted amount of adsorbate adsorbed in equilibrium condition was closer to that of the measured one. This is confirmed by the fact that from the values of fitting param- eter qe of pseudo-second order model agree with the values of qe from kinetic experimental data. As mentioned by Plazinski et al. [24] that one of the potential advantages of using pseudo-second order kinetic model to correlate the adsorption kinetic experimen- tal data

10**is its small sensitivity to the influence of the random experimental error,**

especially for the fitting parameter qe. Another advantage of using the pseudo-second order is that the value of fit- ted parameter qe can be obtained from the independent plot of ki- netic adsorption experimental data [24,27]. In many adsorption kinetic studies [24,27–29],

7**failure of the pseudo-first order** in correlating adsorption **kinetic experimental data is due to** the **theoretical**

interpretations of this equation. In pseudo-first order it is based on the common

1**assumption that the overall sorption process is controlled by the rate of** adsorp- tion **/desorption processes** which is **seen** as **a chemical reaction on the**

surface of

adsorbent [24]. However in many cases, the rate of diffusion of solute into interior of the adsorbent also controls the adsorption mechanism and this phenomenon is not captured by the pseudo-first order model. Table 2, Figs. 3 and 4 clearly indicate that the parameter k2

1**plays a role** as **time scaling factor.**

At high k2, shorter

1**time is required to reach equilibrium.**

As can be seen in Table 2, k2 decreased with increasing initial concentration. A simple correlation between of k 2 and initial concentration of the adsorbate (Co) is given as follows: k2 ¼ k2;o þ a expð?bCoÞ ð16Þ where k2,o, a and b are empirical constants. Plots of Co versus k2 are given in Fig. 5. The fitted empirical constants for catechin are k2,o = 0.0013 g/mg min, a = 0.0194 g/mg min, and b = 0.0092 L/mg, while for epicatechin are k2,o = 0.0004 g/mg min, a = 0.0147 g/ mg min, and b = 0.0207 L/mg. Table 2 Fitted parameters of pseudo-first and pseudo-second order of catechin and epicatechin adsorption onto activated carbon NORIT nrs carbon Ea 0.5–1.5. Phenolic compound Co, mg/L

26**qe, mg/g,** experiment **Pseudo-first order Pseudo-second order k1, 1/min qe, mg/g R2 k2, g/mg min qe, mg/g R2**

Catechin 101.3 122.5 139.8 158.7 203.6 251.9 Epicatechin 100.5 118.4 141.2 161.5 201.7 252.3 43.3 43.9 44.5 44.7 45.3 46.1 0.0631 0.0471 0.0441 0.0337 0.0275 0.0185 21.2 21.9 23.2 25.4 28.2 28.9 0.1037 0.1096 0.0974 0.0795 0.0708 0.0656 39.1 38.6 38.3 38.2 35.6 33.6 0.996 0.997 0.998 0.998 0.996 0.994 19.6 22.2 21.1 22.5 25.4 26.5 0.993 0.996 0.996 0.995 0.994 0.995 0.0022 0.0014 0.0013 0.0008 0.0007 0.0004 43.4 44.3 44.3 46.1 44.2 45.1 0.0086 0.0083 0.0069 0.0053 0.0040 0.0034 20.9 23.6 22.8 24.5 27.9 29.3 0.997 0.998 0.999 0.998 0.997 0.996 0.994 0.998 0.996 0.996 0.997 0.995 50 (a) 40 qt (mg.g-1) 30 20 10 0 0 20 40 60 80 100 120 140 160 T (min) 50 (b) 40 qt (mg.g-1) 30 20 10 0 0 20 40 60 80 100 120 140 160 T (min) Fig. 3. Kinetic adsorption experimental data of catechin onto activated carbon NORIT nrs carbon Ea 0.5–1.5 at various initial concentration

7**and plots** of **(a) pseudo- first order, and (b) pseudo-second order.**

3.4. Recover of phenolic compounds in sago waste effluent To verify the potential application of recovering phenolic com- pounds from sago waste effluent by adsorption, adsorption exper- iments using real sago waste effluent were also conducted. The adsorption of catechin and epicatechin from real sago waste efflu- ent was conducted isothermally at 303.15 K under static condition. Prior to the adsorption experiment, the sago waste effluent was fil- tered to remove solid particles. The initial concentration of cate- chin and epicatechin in sago waste effluent was 321.8 mg/L and 158.2 mg/L, respectively. The recovery of catechin and epicatechin in sago waste effluent

13**is given in Table** 3. **From Table** 3 **it can be seen that the**

recovery of both phenolic compounds increases with increasing activated carbon mass due to the increase of adsorption active sites. At an adsorbent dose of 2%, the

19**recovery of catechin and epicatechin from sago waste effluent**

was 85% and 53%, respec- tively. It can be seen that the amount of phenolic compounds ad- sorbed by the activated carbon from real waste sago effluent (Table 3) is less than that of the synthetic one (Fig. 2). In waste sago effluent, in addition to catechin and epicatechin other compounds such as protein and soluble starch are also present which compete for adsorption active sites

29**resulting in the** observed decrease **of adsorption capacity**

toward catechin and epicatechin.

29**Reusability of adsorbent is crucial in industrial** application. **The**

recovery of catechin and epicatechin from activated carbon was performed by supercritical CO2 at 313.15 K and 26 MPa under sta- tic and dynamic conditions. Details about the supercritical system can be found elsewhere [30]. At static condition, the recovery of catechin from activated carbon was only 1.2% while for epicatechin it was 14.5%. This low recovery is consistent with the solubility 30 (a) 25 20 qt

15**(mg.g** -1) **15 10 5 0 0 20 40 60 80 100** 120 140 160 **T (min)**

30 (b) 25 20 qt

15**(mg.g** -1) **15 10 5 0 0 20 40 60 80 100** 120 140 160 **T (min) Fig. 4.**

Kinetic adsorption experimental data of epicatechin onto activated carbon NORIT nrs carbon Ea 0.5–1.5 at various initial concentration

7**and plots** of **(a) pseudo- first order, and (b) pseudo-second order. data** of both phenolic compounds in supercritical CO2 under static condition [30,31]. In order to increase the recovery of catechin and epicatechin in supercritical CO2, dynamic extraction experiments were also conducted with flow rates of CO2 from 0.1 to 1 kg/h. By increasing CO2 flow rate from 0 kg/h (static mode) to 0.1 kg/h, the recovery increased from 1.2% to 2.3% and from 14.5% to 27% for catechin and epicatechin, respectively. Maximum recovery (catechin 9.7% and epicatechin 87.3%) was achieved

42**at a** CO2 **flow rate of 0.** 7 **kg/h.**

Further increase of CO2 flow rate to 1 kg/h had neg- ligible effect on the recovery both compounds. 0.010 0.008 Catechin Epicatechin k2, g/mg.min 0.006 0.004 0.002 0.000 80 100 120 140 160 180 200 220 240 260 Initial concentration, mg/L Fig. 5. Correlation between Co and k2. 412 F.E. Soetaredjo et al. / Chemical Engineering Journal 231 (2013) 406–413 Table 3 Adsorption of catechin and epicatechin from sago waste effluent. Co,1 catechin, mg/ Co,2 epicatechin, mg/ m, mass of activated carbon, L L g Ce,1, mg/ Ce,2, mg/ L L qe,1, mg/ qe,2, mg/ % Recovery g g catechin % Recovery epicatechin 321.8 158.2 0.112 0.483 0.689 0.921 1.104 1.328 1.573 1.921 274.4 143.3 141.2 113.8 110.3 103.8 99.8 100.2 95.5 96.4 85.4 93.1 71.2 87.4 48.3 74.2 42.3 13.3 37.4 9.2 30.7 7.9 24.1 6.3 20.5 5.6 17.8 4.9 15.9 4.5 14.2 4.3 14.7 9.4 56.1 28.1 65.7 34.4 68.9 36.7 70.3 39.1 73.5 41.1 77.7 44.7 84.8 52.3 Note: volume of sago waste effluent 100 mL. Low recovery of phenolic compound, especially catechin is caused by the difference in the polarity between the solute (pheno- lic compound) and the solvent (supercritical CO2). Catechin is highly polar while supercritical CO2 is non-polar. In order to in- crease the polarity of supercritical CO2, ethanol as co-solvent (5– 25%) was used in this study. By adding 5% ethanol

42**at a** CO2 **flow rate of 0.** 7 **kg/h,**

the recovery of catechin increased almost two fold (from 9.7% to 18.2%), and while for epicatechin the recovery reached 95.7%. Increasing the percentage of co-solvent increased the recovery of both compounds. For epicathecin the maximum recovery was 99.2% at 20% of co-solvent. Further increase in co-sol- vent concentration had negligible effect on the recovery of epicat- echin. The un-recovered epicatechin may be strongly bound on carbon surface or epicatechin molecule may be adsorbed inside small or ultra micropore so it could not be desorbed by supercrit- ical CO2. At 30% co-solvent, the recovery of catechin was 93.1%. Since further increase of co-solvent concentration changed super- critical condition into subcritical, therefore maceration (303.15 K, 24 h) was used to obtain maximum removal of catechin (97.8%) and epicatechin (99.4%). 4. Conclusion The

19**recovery of catechin and epicatechin from sago waste**

efflu- ent by adsorption onto activated carbon followed

9**by supercritical CO2** desorption **was demonstrated in this work.** To study **the** ki- netic and equilibria of adsorption, synthetic aqueous catechin and epicatechin solution was used instead of sago waste effluent. The adsorption equilibria and kinetic were obtained isothermally at 303.15 K in static mode condition. The adsorption equilibria of single and binary system were modeled using Langmuir and mod- ified extended Langmuir, respectively. It was found that both of

29**models could represent the** experimental **data well. In** adsorption kinetic experiments, **the**

well-known

9**pseudo-first and pseudo-** sec- ond **models were** employed **to correlate the experimental data. The pseudo-second**

model gave better representation of experimental data than the pseudo-first model. The effect of initial concentration on the value of fitting parameters k1 and k2 was also studied. The results indicate that k2

1**plays a role** as **time scaling factor,**

at high k2, shorter

1**time is required to reach equilibrium.**

Adsorption using real sago waste effluent followed by supercrit- ical CO2 extraction indicates that this method has potential appli- cation in recovery catechin and epicatechin in industrial scale. Ethanol as co-solvent was required to obtain high recovery of cat- echin from the activated carbon. References [1] M.A. Soobrattee, V.S. Neergheen, A. Luximon-Ramma, O.I. Aruoma, T. Bahorun, Phenolics as potential antioxidant therapeutic agents: mechanism and actions, Mutat. Res. 579 (2005) 200–213. [2] L. Yu, K. Zhou, Antioxidant properties of bran extracts from ‘‘platte’’ wheat grown at different locations, Food Chem. 90 (2004) 311–316. [3] P. Vitaglione, A. Napolitano, V. Fogliano, Cereal dietary fibre: a natural functional ingredient to deliver phenolic compounds into the gut, Trends Food Sci. Technol. 19 (2008) 451–463. [4] A.R. Rechner, M.A. Smith, G. Kuhnle, G.R. Gibson, E.S. Debnam, S.K.S. Srai, K.P. Moore, C.A. Rice-Evans, Colonic metabolism of dietary polyphenols: influence of structure on microbial fermentation products, Free Radical Biol. Med. 36 (2004) 212–225. [5] A.R. Rechner, G. Kuhnle, P. Bremner, G.P. Hubbard, K.P. Moore, C.A. Rice-Evans, The metabolic fate of dietary polyphenols in humans, Free Radical Biol. Med. 33 (2002) 220–235. [6] O. Pourali, F.S. Asghari, H. Yoshida, Production of phenolic compounds from rice bran biomass under subcritical water conditions, Chem. Eng. J. 160 (2010) 259–266. [7] H. Epstein, Cosmeceuticals and polyphenols, Clin. Dermatol. 27 (2009) 475– 478. [8] S.M. Anthonysamy, N.B. Saari, K. Muhammad, F.A. Bakar, Browning of sago (Metroxylon sagu) pith slurry an influenced by holding time, pH and temperature, J. Food Biochem. 28 (2004) 91–99. [9] A.T. Pei-Lang, A.M.D. Mohamed, A.A. Karim, Sago starch and composition of associated components in palms of different growth stages, Carbohyd. Polym. 63 (2006) 283–286. [10] M. Ahmaruzzaman, Adsorption of phenolic compounds on low-cost adsorbents: a review, Adv.

Colloid Interfaces 143 (2008) 48–67. [11] K. Baransi, Y. Dubowski, I. Sabbah, Synergetic effect between photocatalytic degradation and adsorption processes on the removal of phenolic compounds from olive mill wastewater, Water Res. 46 (2012). [12] A. Dabrowski, P. Podkoscielny, Z. Hubicki, M. Barczak, Adsorption of phenolic compounds by activated carbon – a critical review, Chemosphere 58 (2005) 1049–1070. [13] J. Qi, Y. Guo, H. Xu, Adsorption of phenolic compounds on micro- and mesoporous rice husk-based active carbons, Mater. Chem. Phys. 87 (2004) 96– 101. [14] C. Michailof, G.G. Stavropoulos, C. Panayiotou, Enhanced adsorption of phenolic compounds, commonly encountered in olive mill wastewaters, on olive husk derived activated carbons, Bioresour. Technol. 99 (2008) 6400– 6408. [15] M.L. Soto, A. Moure, H. Dominguez, J.C. Parajo, Recovery, concentration and purification of phenolic compounds by adsorption: a review, J. Food Eng. 105 (2011) 1–27. [16] F.E. Soetaredjo, A. Kurniawan, L.K. Ong, S. Ismadji, Incorporation of selectivity factor in modeling binary component adsorption isotherms for heavy metals-biomass system, Chem. Eng J. 219 (2013) 137–148. [17] C.H. Giles, D. Smith, A. Huitson, A general treatment and classification of the solute adsorption, I., Theor. J. Colloid Interface Sci. 47 (1974) 755–765. [18] C.H. Giles, A.P. D'Silva, I.A. Easton, A general treatment and classification of the solute adsorption isotherm, II, J. Colloid Interface Sci. 47 (1974) 766– 778. [19] Y. Ding, D. Jing, H. Gong, L. Zhou, X. Yang, Biosorption of aquatic cadmium (II) by unmodified rice straw, Bioresour. Technol. 114 (2012) 20–25. [20] H. Muhamad, H. Doan, A. Lohi, Batch and continuous fixed-bed column biosorption of Cd2+ and Cu2+, Chem. Eng. J. 158 (2010) 369–377. [21] C.G. Rocha, D.A.M. Zaia, R.V.d.S. Alfaya, A.A.d.S. Alfaya, Use of rice straw as biosorbent for removal of Cu(II), Zn(II), Cd(II) and Hg(II) ions in industrial effluents, J. Hazard. Mater. 166 (2009) 383–388. [22] K.K.H. Choy, J.F. Porter, G. McKay, Langmuir isotherm models applied to the multicomponent sorption of acid dyes from effluent onto activated carbon, J. Chem. Eng. Data 45 (2000) 575–584. [23] A. Kurniawan, H. Sutiono, N. Indraswati, S. Ismadji, Removal basic dyes in binary system by adsorption using rarasaponin–bentonite: revisited extended Langmuir model, Chem. Eng. J. 189–190 (2012) 264–274. [24] W. Plazinski, W. Rudzinski, A. Plazinska, Theoretical models of sorption kinetics including a surface reaction mechanism: a review, Adv. Colloid Interfaces 152 (2009) 2–13. [25] S. Lagergren, Zur theorie der sogenannten adsorption geloster stoffe Kungliga Svenska Vetenskapsakademiens, Handlingar 24 (1898) 1–39. [26] G. Blanchard, M. Maunaye, G. Martin, Removal of heavy metals from waters by means of natural zeolites, Water Res. 18 (1984) 1501–1507. [27] A.N. Kosasih, J. Febrianto, J. Sunarso, Y.-H. Ju, N. Indraswati, S. Ismadji, Sequestering of Cu(II) from aqueous solution using cassava peel (Manihot esculenta), J. Hazard. Mater. 180 (2010) 366–374. [28] A. Kurniawan, H. Sutiono, Y.-H. Ju, F.E. Soetaredjo, A. Ayucitra, A. Yudha, S. Ismadji, Utilisation of rarasaponin natural surfactant for organo-bentonite preparation: application for methylene blue removal from aqueous effluent, Micropor. Mesopor. Mater. 142 (2011) 184–193. [29] A. Kurniawan, S. Ismadji, Potential utilization of Jatropha curcas L press cake residue as new precursor for activated carbon preparation: application in methylene blue removal from aqueous solution, J. Taiwan Inst. Chem. Eng. 42 (2011) 826–836. [30] F.E. Soetaredjo, S. Ismadji, Y.-H. Ju, Measurement and modeling of epicatechin solubility in supercritical carbon dioxide fluid, Fluid Phase Equilib. 340 (2013) 7–10. [31] F.E. Soetaredjo, S. Ismadji, M.J. Liauw, Y.-H. Ju, Catechin sublimation pressure and solubility in supercritical carbon dioxide, Fluid Phase Equilib. (2013). submitted for publication.

20**Author's personal copy Author's personal copy**

F.E. Soetaredjo et al. /

14**Chemical Engineering Journal 231 (2013) 406–413**

407

20**Author's personal copy Author's personal copy**

F.E. Soetaredjo et al. /

14**Chemical Engineering Journal 231 (2013) 406–413**

409

20**Author's personal copy Author's personal copy**

F.E. Soetaredjo et al. /

14**Chemical Engineering Journal 231 (2013) 406–413**

411

20**Author's personal copy**

Author's personal copy F.E. Soetaredjo et al. / Chemical Engineering Journal 231 (2013) 406–413 413