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Prediction of the solid solubility in supercritical carbon dioxide using a new mixing rule with Lenard-Jones parameters

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ABSTRACT

This study is performed to provide a new mixing rule based on the Lenard-Jones parameters for calculation of the solid solubility in supercritical carbon dioxide. The solubility of solid solutes in supercritical fluid is an important thermo-physical property which is required to be determined. Since generally there are few data about solubility, we need to provide methods to estimate the solubility of solid solutes in supercritical solvents by using limited information. In the current study, the cubic Peng–Robinson (PR) and Soave–Redlich–Kwong (SRK) equations of state are used to estimate the solid solubilities of 18 solutes in supercritical carbon dioxide. This estimation is performed by using four mixing rules which are called the van der waals one fluid rule with one (VDW1) and two (VDW2) adjustable parameters, the covolume dependent (CVD) rule and the new mixing rule. The prediction of the new mixing rule is more accurate than the other mixing rules in the same equations of state for calculating solid solubility in supercritical carbon dioxide and the results of various calculations demonstrate that the proposed mixing rule is in good agreement with the 665 experimental data points that are used in this work. © 2013 Trade Science Inc. - INDIA

KEYWORDS

Mixing rule;
Equation of state;
Solid solubility;
Supercritical carbon dioxide;
Lenard-Jones parameters.

INTRODUCTION

Supercritical fluids (SCF) are widely used in chemical processes such as extractions, purifications, separations, crystal growth, reactions, fractionations, remove impurity of chemical products, regenerating activated carbon, industrial food applications and many other processes^[1]. Carbon dioxide is a promising solvent since it is inexpensive, nontoxic, inflammable, and environmentally acceptable while it possesses a low critical temperature and a moderate critical pressure ($T_c=304.25$

K and $P_c=73.8$ bar)^[1,2].

The design of the extraction processes with SCF's depends upon the ability to predict the solubility of solid in supercritical fluids^[2]. In order to design optimized supercritical processes, solubility data of the considered compounds are needed. It is difficult to determine solid solubility in supercritical fluid accurately. Due to the difficulties of experimental measurements and also time-consuming and costly nature of such techniques^[3], it is desirable to develop predictive methods for estimating the phase behavior of these kinds of systems.

To predict the solubility of a solute in supercritical fluids, Equation of State (EOS) models are widely incorporated. Cubic EOS's are the simplest equations capable of predicting and representing fluid phase equilibrium of pure and fluid mixtures^[2]. Therefore thermodynamic models including EOS's with different mixing rules are widely used to predict solid solubilities at equilibrium conditions. A number of empirical mixing rules have been proposed for the representation of solid-supercritical fluid equilibrium, which have been discussed in reviews by Johnston et al.^[4] and Brennecke and Eckert^[5]. Several researchers have suggested that the selection of the mixing rules is more important than the EOS itself^[6,7]. Therefore, the focus of this study is to propose a new mixing rule for the determination of the solid solubilities of aromatic hydrocarbons, aliphatic carboxylic acids, aromatic acids, and heavy aliphatic and aromatic alcohols in supercritical carbon dioxide. The cubic Peng–Robinson (PR)^[8] and Soave–Redlich–Kwong (SRK)^[9] equations of state have been used to calculate the solid solubilities of 18 solutes in supercritical CO₂, by using four mixing rules, namely, the van der Waals one fluid rule with one (VDW1) and two (VDW2) adjustable parameters, the covolume dependent (CVD) rule and the new mixing rule.

In the new mixing rule, the surface of solid is considered as a plate that is surrounded by molecules of supercritical carbon dioxide; then the interaction energy of sorbent molecule with a single infinite layer plane of solid molecule is calculated using Kirkwood–Müller formula and distance between the layers is related on diameters of molecule and then a volume correction term is proposed on the basis of second virial coefficient and Lenard-Jones 6-12 potential.

The model is applied to calculate solubilities of various solid compounds in supercritical CO₂. The optimal values of the model adjustable parameters have been obtained for 665 experimental data points. Finally, the most accurate combination of the mentioned equations of state with the mixing rules, which leads to the least absolute average deviation of the results from experimental values (AARD %) is reported.

CALCULATION OF SOLUBILITY OF SOLIDS IN SUPERCRITICAL FLUID

First equilibrium condition is equality of the solute

fugacity in both solid and supercritical fluid phases:

$$\hat{f}_i^{\text{solid}} = \hat{f}_i^{\text{supercriticalfluid}} \quad (1)$$

Solubility of supercritical fluid in solid phase is negligible; therefore, solid phase is supposed as a pure solid. By assuming that the solid phase is incompressible, and that the molar volume of the solid is constant, the following correlation is obtained:

$$\hat{f}_i^{\text{solid}} = P_i^{\text{sat}} \hat{\phi}_i^{\text{sat.s}} \exp \left[\frac{v_i^s (P - P_i^{\text{sat}})}{RT} \right] \quad (2)$$

where P is pressure, P_i^{sat} is sublimation pressure, $\hat{\phi}_i^{\text{sat.s}}$ is the fugacity coefficient of solute i at saturation, R is the universal constant, T is temperature and v_i^s is solid molar volume.

Fugacity of solute “ i ” in the supercritical fluid phase is obtained from the following equation:

$$\hat{f}_i^{\text{supercritical}} = y_i \hat{\phi}_i P \quad (3)$$

where $\hat{\phi}_i$ is the fugacity coefficient of solute i in the supercritical fluid solvent.

Following relationship is used to calculate mole fraction of solid:

$$y_2 = \frac{P^{\text{sat}} \hat{\phi}_2^{\text{sat}}}{P \hat{\phi}_2} \exp \left[\frac{v_2^s (P - P_2^{\text{sat}})}{RT} \right] \quad (4)$$

where subscript 2 indicates the heavy solute component.

The sublimation pressure is obtained from the Antoine equation:

$$\log P^{\text{sat}} = A' - \frac{B'}{T(K) - C'} \quad (5)$$

where A, B, C are constant.

The fugacity coefficient of solute in the supercritical fluid solvent is determined by using a cubic equation of state (EOS). The general form of a cubic EOS is given by:

$$P = \frac{RT}{v-b} - \frac{a}{v^2 + Uv + W} \quad (6)$$

Depending on numerical values of U and W , various equations of state can be obtained.

For the case of $U=b$ and $W=0$, the Soave-Redlich-Kwong (SRK) EOS with the following two parameters is derived:

$$a = 0.42747R^2 T_c^2 \alpha(T_r) / P_c \quad (7)$$

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$$b = 0.08664RT_C/P_C \quad (8)$$

where

$$\alpha(T_r) = \left(1 + m(1 - T_r^{0.5})\right)^2 \quad (9)$$

$$m = 0.480 + 1.574\omega - 0.176\omega^2 \quad (10)$$

ω is acentric factor.

Incorporation of $U=2b$, $W=b^2$ in Eq. (6) leads to the Peng-Robinson (PR) EOS with the following parameters:

$$a = 0.45724R^2T_C^2\alpha(T_r)/P_C \quad (11)$$

$$b = 0.07778RT_C/P_C \quad (12)$$

where

$$m = 0.37464 + 1.54226\omega - 0.26992\omega^2 \quad (13)$$

PR and SRK equations of state with four mixing rules (VdW1, VdW2, CVD, and new mixing rules) have been applied to estimate solid solubility in supercritical carbon dioxide. The mixing rules are summarized in TABLE 1.

TABLE 1 : Summary of the mixing rules used in this work.

Mixing rules	Functional form	Refs.
VdW1	$a = \sum \sum x_i x_j \sqrt{a_i a_j} (1 - k_{ij}), b = \sum x_i b_i$	[14]
VdW2	$a = \sum \sum x_i x_j \sqrt{a_i a_j} (1 - k_{ij})$ $b = \sum \sum x_i x_j \left(\frac{b_i + b_j}{2}\right) (1 - l_{ij})$	[14]
CVD	$a = \sum \sum x_i x_j a_{ij} \left(\frac{b}{b_i}\right)^{n_{ij}}, b = \sum x_i b_i$ $a = \sqrt{(a_i a_j)} \quad b = \sqrt{(b_i b_j)}$	[15]

In this study, a new mixing rule has been developed based on the Lenard-Jones parameters that obtained from Kirkwood-Müller formula. At first solid surface is assumed as a plate contacted with carbon dioxide molecules then interaction energy between CO₂ can be calculated by Lenard-Jones 6-12 potential^[10,11]:

$$\varepsilon_{12}(z) = 4\varepsilon_{12}^* \left[\left(\frac{\sigma}{z}\right)^{12} - \left(\frac{\sigma}{z}\right)^6 \right] \quad (14)$$

The interaction of one molecule CO₂ and an infinite layer plane of solid molecules is obtained^[10-12]:

$$\varepsilon(z) = \frac{N_s A_s}{2\sigma^4} \left[\left(\frac{\sigma}{z}\right)^{10} - \left(\frac{\sigma}{z}\right)^4 \right] \quad (15)$$

where n is the number of solid molecules/surface unit, is dispersion constant which is calculated by Kirkwood-Müller formula as follows:

$$A_s = \frac{6m'c^2\alpha_s\alpha_A}{\chi_s + \frac{\alpha_A}{\chi_s}} \quad (16)$$

where m is the mass of one electron, c is the speed of light, χ_A and χ_s are polarizabilities of CO₂ and solid respectively, and α_A and α_s are magnetic susceptibilities of CO₂ and solid respectively.

σ is the intermolecular distance at zero-interaction energy $\sigma = \left(\frac{2}{5}\right)^{1/6} d_0 = 0.858d$ and d_0 is the average of CO₂ and solid molecule diameters. z is the intermolecular distance between CO₂ and solid molecule. In this study, we suppose that the solid surface z is covered by CO₂; As a result z is summation of radius of solid and CO₂; then we have:

$$\frac{\sigma}{z} = \frac{0.858 \times d_0}{\frac{1}{2}(d_1 + d_2)} = \frac{0.858 \times \frac{1}{2}(d_1 + d_2)}{\frac{1}{2}(d_1 + d_2)} = 0.858 \quad (17)$$

where d_1 and d_2 are CO₂ and solid molecule diameters. and:

$$\varepsilon(z) = \frac{N_s A_s}{2\sigma^4} [0.858^{10} - 0.858^4] = -0.162864 \frac{N_s A_s}{\sigma^4} \quad (18)$$

polarizability can be predicted by the following correlation^[13]:

$$\alpha = \left(\frac{3}{4\pi N_A}\right) \times \left(\frac{M}{\rho}\right) \times \left(\frac{n^2 - 1}{n^2 + 2}\right) \quad (19)$$

where N_A is Avogadro's number, M is molecular weight, ρ is absolute density and n is refractive index.

Calculation of second virial coefficient

We have following equations for second virial coefficient:

$$B = \lim_{\rho \rightarrow 0} \frac{\partial Z}{\partial \rho} \quad (20)$$

$$\frac{\partial Z}{\partial \rho} = \frac{b}{(1 - b\rho)^2} - \frac{a}{RT} \left[\frac{1 - c_1 c_2 b^2 \rho^2}{(c_1 c_2 b^2 \rho^2 + (c_1 + c_2) b \rho + 1)^2} \right] \quad (21)$$

$$B = b - \frac{a}{RT} \quad (22)$$

$$B_{ij} = 2\pi N_A \int_0^\infty \left[1 - e^{-\Gamma_{ij}(r)/kT} \right] r^2 dr \quad (23)$$

$$\left(b - \frac{a}{RT} \right)_{ij} = 2\pi N_A \int_0^\infty \left[1 - e^{-\Gamma_{ij}(r)/kT} \right] r^2 dr \quad (24)$$

The integral equation that is mentioned above is replaced by two function in which one of them is in terms of a_{ij} and another one is in terms of b_{ij} .

$$2\pi N_A \int_0^\infty \left[1 - e^{-\Gamma_{ij}(r)/kT} \right] r^2 dr \approx H(\varepsilon / \omega P_c T^* / \sigma^3) - F(\varepsilon / \omega P_c T^* / \sigma) \quad (25)$$

where H and F are equivalent to b_{ij} and a_{ij} , respectively.

$$H = p \left(\frac{g_0 \varepsilon_{12}}{\omega P_c} \right)^q \left(\frac{T^*}{\sigma} \right)^r - L' \quad (26)$$

$$F = p \left(\frac{g_0 \varepsilon_{12}}{\omega P_c} \right)^q \left(\frac{T^*}{\sigma} \right)^r - K' \text{ and } g_0 = 10^{22} \quad (27)$$

$$T^* = \frac{T}{T_c} \quad (28)$$

where P_c and T_c are critical pressure and temperature, T is temperature, ω is acentric factor, ε_{12} is interaction energy between solid and CO_2 , and p, q, r, K' and L' are constant. After optimization with experimental and theoretical values, constant parameters are obtained and p, q and r are shown in TABLE 2. These values are in general condition when there is no experimental data. It is better to optimize the parameters with experimental data for each solid using the values in TABLE 2 as an initial guess.

TABLE 2 : Constant of H and F function for PR and SRK EOS's.

	H (PR)	H (SRK)	F(PR & SRK)
p	0.0082	0.0168	21.5288
q	-0.5697	-0.5822	-0.4951
r	-0.5813	-0.4557	-0.1255

In order to simplify the mixing rule, an optimization program was coded to find an alternative mixing rule in terms of equation of state parameters, as a result, the following mixing rules was obtained with the same result compared to above b_{ij} and a_{ij} in the calculations:

$$a_m = \sum \sum x_i x_j \sqrt{a_i a_j} Q \left(\frac{b_i + b_j}{2} \right)^{m_{ij}} (1 - k_{ij})$$

$$\text{if } \begin{cases} i \neq j \rightarrow m_{ij} = 0 \\ i \neq CO_2 \rightarrow m_{ij} = 0 \\ i = j \rightarrow k_{ij} = 0 \end{cases} \quad (29)$$

$$b_m = \sum \sum x_i x_j \left(\frac{b_i + b_j}{2} \right) (1 - l_{ij})$$

$$\text{if } i = j \rightarrow l_{ij} = 0 \quad (30)$$

where k_{ij}, l_{ij} and m_{ij} are interaction coefficients between components of i and j and Q is a fitting parameter that is highly close to one.

In this work, 18 different hydrocarbon solids are studied. Experimental data of solid solubility in supercritical carbon dioxide at different pressure and temperature are applied to calculate and to compare between various mixing rules with PR and SRK EOS's.

The adjustable parameters of each mixing rule are optimized by simplex method and the Absolute Average Relative Deviation is defined as follows:

$$AARD = \sum_i^N \left(\left| \frac{y_{solid,i}^{exp.} - y_{solid,i}^{calc.}}{y_{solid,i}^{exp.}} \right| \frac{1}{n} \right) \cdot 100 \quad (31)$$

where N is number of solubility data point, $y_{solid,i}^{exp.}$ is i th experimental data point of solid solubility and $y_{solid,i}^{calc.}$ is i th calculated data point of solid solubility

RESULTS AND DISCUSSION

The critical constants, acentric factors and solid molar volumes of 18 compounds are listed in TABLE 3. TABLE 4 shows the constants of Antoine equation. Different combinations of the PR and the SRK EOS's with four mixing rules are mentioned in this study to calculate the solubilities of solid compounds in supercritical carbon dioxide. TABLE 5 shows the comparison of our calculated results with two EOS's and four mixing rules based on Average Absolute Relative Deviation Percent (AARD (%)) and the optimized interaction coefficients between components of i and j (k_{ij}, l_{ij}, m_{ij}) are presented too. TABLE 5 shows that AARD (%) for the new mixing rule in the same equations of state, is minimum in compare with the other mixing rules that are discussed here.

Figure 1 and Figure 2 show the comparison of our calculated results with the experimental data for 1-Hexadecanol, 1-Octadecanol, Stearic acid and Palm-

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itic acid in supercritical carbon dioxide by using PR EOS at 338K and SRK EOS at 328K, respectively. Figure 3 shows the comparison of our calculated results with

the experimental data for 2,5-Xylenol, Benzoin, 3-4-Xylenol, Fluorene, Mandelic Acid, Naphthalene, Pyrene and Propyl-4-hydroxy benzoate in supercritical carbon

TABLE 3 : Physical properties of the studied components

Component	T_c (K)	P_c (bar)	ω	Refs.	v_2^s (l/mol)	Refs.
Carbon dioxide (solvent)	304.2	73.7	0.225	[16]	–	–
Benzoin	853.52	26.6	0.599	[17,18,19]	0.162	[20]
Anthracene	869.15	30.8	0.353	[15]	0.1426	[15,17,21-23]
Pyrene	936	25.7	0.509	[24]	0.1585	[17]
Mandelic Acid	903.79	34.73	0.645	[17,18,19]	0.117	[25]
Propyl-4-hydroxy benzoate	815.92	31.3	0.722	[17,18,19]	0.1316	[26]
3-4-Xylenol	729.8	49	0.576	[16,27]	0.1243	[27]
2,5-Xylenol	706.9	48	0.569	[16,27]	0.1257	[27,28]
Naphthalene	748.4	40.51	0.302	[29,30]	0.111	[31]
Phenanthrene	882.65	31.715	0.437	[32]	0.182	[31]
Fluorene	826.4	29.5	0.406	[15]	0.1393	[17,33]
2,6-Dimethyl naphthalene	777	31.8	0.42	[34]	0.1392	[17]
2,7-Dimethyl naphthalene	777	32.2	0.42	[31,35]	0.136	[35,36]
O-hydroxy benzoic acid	739	51.8	0.832	[37]	0.0957	[38]
P-hydroxy benzoic acid	739	51.8	0.832	[37]	0.0924	[38]
1-Hexadecanol	761	14.9	0.748	[4]	0.2965	[39]
1-Octadecanol	777	13.4	0.863	[4]	0.333	[39]
Palmitic acid	776	14.9	1.083	[4]	0.2857	[40]
Stearic acid	779	13.4	1.084	[4]	0.3024	[39,41]

TABLE 4 : Constant of Antoine equation

Component	\hat{A}	\hat{B}	\hat{C}	Pressure unit	Refs.
Naphthalene	8.583	3733.9	0	bar	[42]
Phenanthrene	9.631	4873.4	0	bar	[43]
Anthracene	9.775	5313.7	0	bar	[29]
Fluorene	9.429	4419.5	0	bar	[25]
Pyrene	8.3496	4904	0	bar	[29]
2,6-Dimethyl naphthalene	9.429	4419.5	0	bar	[29]
2,7-Dimethyl naphthalene	12.431	4388.11	0	mmHg	[35]
1-Hexadecanol	22.773	8736	0	kPa	[44]
1-Octadecanol	24.99	9787	0	kPa	[44]
Palmitic acid	19.342	8069	0	kPa	[44]
Stearic acid	6.171	2157.5	153.78	kPa	[44]
2,5-Xylenol	15.495	4438.6	0	Pa	[45]
3-4-Xylenol	15.298	4478.2	0	Pa	[45]
Mandelic acid	49.83 ^a	17256.62 ^a	0 ^a	Pa	[46]
Benzoin	36.38 ^a	13160.97 ^a	0 ^a	Pa	[46]
Propyl-4-hydroxy benzoate	41.15 ^a	14209.69 ^a	0 ^a	Pa	[46]
O-hydroxy benzoic acid	2.25E-6@45°C ^b 6.65E-6@55°C ^b			bar	[37]
P-hydroxy benzoic acid	1.37E-8@45°C ^b 3.76E-8@55°C ^b			bar	[37]

^a $\ln p^{\text{sat}} = A - \frac{B}{T(\text{K}) - C}$; ^bSublimation pressure

TABLE 5 : Optimized interaction coefficients between components of i and j and average absolute relative deviation percent for the solubility of pure component in supercritical CO₂ with four different mixing rules using the PR and SRK EOS's.

Component	T(K)	P(bar)	N ^a	EOS	AARD(%)				k_{ij}	l_{ij}	m_{ij}
					VdW1	VdW2	CVD	This Work			
Benzoin [46]	308.15	121.6–236.1	6	PR	2.8292	2.6338	2.673	2.4928	0.0906	0.0090	-0.0012
				SRK	3.7486	2.6898	4.6249	2.5954	0.1008	0.0385	-0.0023
	318.15	111.3–244.3	7	PR	3.9081	4.3922	3.5001	3.0957	0.0911	0.0402	-0.0001
				SRK	5.2831	3.3924	5.0384	3.4223	0.0987	0.0385	-0.0008
328.15	114.8–244.3	6	PR	10.2256	4.366	10.2674	1.1958	0.0901	0.0215	0.0043	
			SRK	10.8436	5.949	8.6901	2.4733	0.0973	0.0210	0.0043	
Anthracene [47]	318.15	84.4–564.4	4	PR	28.4037	24.7712	27.8199	6.3541	2.10E-16	0.1850	-0.0250
				SRK	28.8286	24.6342	28.2802	4.1721	0.0300	0.2450	-0.0300
	323.15	89–836.3	6	PR	24.3418	25.5412	24.5633	7.2509	0.0500	0.2850	-0.0350
				SRK	31.3374	27.1708	33.4898	6.5453	0.0800	0.3450	-0.0400
	328.15	94.7–89.09	5	PR	27.5037	22.7695	29.1631	4.0347	0.0450	0.2850	-0.0400
				SRK	35.6341	24.0610	37.7778	5.5060	0.0650	0.3300	-0.0450
	358.15	129–930.3	6	PR	18.1184	17.1583	18.2654	14.4283	0.0100	0.2350	-0.0300
				SRK	25.7956	18.6818	24.1200	14.3251	0.0350	0.3000	-0.0400
	363.15	132.6–975.7	6	PR	16.9620	16.4691	16.7251	13.5154	0.0050	0.2300	-0.0300
				SRK	26.4216	17.8756	24.829	13.9934	0.0300	0.3000	-0.0450
	368.15	137–1020.5	5	PR	21.3167	20.1808	21.4619	14.8580	0.0150	0.2700	-0.0550
				SRK	28.2421	22.4729	27.8522	14.8614	0.0350	0.3300	-0.0700
Pyrene [19,48]	308.15	83.6–483.4	7	PR	30.6695	13.7620	30.9967	7.3232	0.2600	0.3600	-0.0100
				SRK	37.5088	16.0310	35.8357	7.9719	0.2850	0.4050	-0.0150
	308.2	80.4–203.5	45	PR	11.6926	11.4037	12.1831	5.4511	0.2050	0.2350	-0.0050
				SRK	13.2584	12.5155	13.6199	6.4711	0.2200	0.2500	-0.0050
318.2	95–254	20	PR	9.4866	8.7079	9.4178	3.6209	0.2300	0.3000	-0.0100	
			SRK	11.2353	10.1302	11.2394	4.8605	0.2600	0.3550	-0.0150	
Pyrene [19,48]	323.15	104.3–483.4	7	PR	36.0100	7.4995	34.1467	8.0844	0.2450	0.3400	-0.0100
				SRK	38.2325	10.7713	41.7837	6.6957	0.2600	0.3650	-0.0100
	323.2	100.1–228.5	35	PR	5.1136	4.7663	4.4540	3.2411	0.1850	0.2000	-0.0050
				SRK	6.0415	6.9306	5.4578	3.2414	0.2200	0.2650	-0.0100
	328.2	105.245	20	PR	6.7645	6.1578	5.8100	3.4319	0.2300	0.3050	-0.0150
				SRK	7.1908	7.5917	6.9991	2.6587	0.2350	0.3050	-0.0150
	343.15	104.3–483.4	8	PR	55.5147	24.7147	53.3595	21.5114	0.2300	0.3250	-0.0150
				SRK	52.5814	22.1344	53.1025	17.3982	0.2550	0.3750	-0.0200
Mandelic Acid [46]	308.15	101–228.5	7	PR	37.0743	6.2913	37.4339	7.8740	0.2000	0.4700	0.0050
				SRK	37.2863	6.652	39.6686	7.4203	0.2200	0.5150	0.0050
	318.15	102.3–225.7	7	PR	31.6307	5.1715	33.2679	2.6531	0.2750	0.6500	-0.0050
				SRK	36.1244	5.1098	36.4133	2.7383	0.2850	0.6800	-0.0050
328.15	104.4–230.6	7	PR	31.5318	10.3430	30.3295	9.1046	0.2800	0.6800	-0.0050	
			SRK	29.0384	10.8282	30.4613	9.8902	0.2850	0.7000	-0.0050	
Propyl-4-hydroxy benzoate [46]	308.15	94.1–220.9	7	PR	13.5994	2.1176	14.3637	1.5054	0.2350	0.4200	-0.0050
				SRK	15.6949	2.6009	17.2696	1.1920	0.2400	0.4200	-0.0050

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	328.15	114.8–244.3	6	PR	10.2256	4.366	10.2674	1.1958	0.0901	0.0215	0.0043	
				SRK	10.8436	5.949	8.6901	2.4733	0.0973	0.0210	0.0043	
Anthracene [47]	318.15	84.4–564.4	4	PR	28.4037	24.7712	27.8199	6.3541	2.10E-16	0.1850	-0.0250	
				SRK	28.8286	24.6342	28.2802	4.1721	0.0300	0.2450	-0.0300	
	323.15	89–836.3	6	PR	24.3418	25.5412	24.5633	7.2509	0.0500	0.2850	-0.0350	
				SRK	31.3374	27.1708	33.4898	6.5453	0.0800	0.3450	-0.0400	
	328.15	94.7–89.09	5	PR	27.5037	22.7695	29.1631	4.0347	0.0450	0.2850	-0.0400	
				SRK	35.6341	24.0610	37.7778	5.5060	0.0650	0.3300	-0.0450	
	358.15	129–930.3	6	PR	18.1184	17.1583	18.2654	14.4283	0.0100	0.2350	-0.0300	
				SRK	25.7956	18.6818	24.1200	14.3251	0.0350	0.3000	-0.0400	
	363.15	132.6–975.7	6	PR	16.9620	16.4691	16.7251	13.5154	0.0050	0.2300	-0.0300	
				SRK	26.4216	17.8756	24.829	13.9934	0.0300	0.3000	-0.0450	
	368.15	137–1020.5	5	PR	21.3167	20.1808	21.4619	14.8580	0.0150	0.2700	-0.0550	
				SRK	28.2421	22.4729	27.8522	14.8614	0.0350	0.3300	-0.0700	
Pyrene [19,48]	308.15	83.6–483.4	7	PR	30.6695	13.7620	30.9967	7.3232	0.2600	0.3600	-0.0100	
				SRK	37.5088	16.0310	35.8357	7.9719	0.2850	0.4050	-0.0150	
	308.2	80.4–203.5	45	PR	11.6926	11.4037	12.1831	5.4511	0.2050	0.2350	-0.0050	
				SRK	13.2584	12.5155	13.6199	6.4711	0.2200	0.2500	-0.0050	
	318.2	95–254	20	PR	9.4866	8.7079	9.4178	3.6209	0.2300	0.3000	-0.0100	
				SRK	11.2353	10.1302	11.2394	4.8605	0.2600	0.3550	-0.0150	
Pyrene [19,48]	323.15	104.3–483.4	7	PR	36.0100	7.4995	34.1467	8.0844	0.2450	0.3400	-0.0100	
				SRK	38.2325	10.7713	41.7837	6.6957	0.2600	0.3650	-0.0100	
	323.2	100.1–228.5	35	PR	5.1136	4.7663	4.4540	3.2411	0.1850	0.2000	-0.0050	
				SRK	6.0415	6.9306	5.4578	3.2414	0.2200	0.2650	-0.0100	
	328.2	105.245	20	PR	6.7645	6.1578	5.8100	3.4319	0.2300	0.3050	-0.0150	
				SRK	7.1908	7.5917	6.9991	2.6587	0.2350	0.3050	-0.0150	
	343.15	104.3–483.4	8	PR	55.5147	24.7147	53.3595	21.5114	0.2300	0.3250	-0.0150	
				SRK	52.5814	22.1344	53.1025	17.3982	0.2550	0.3750	-0.0200	
	Mandelic Acid [46]	308.15	101–228.5	7	PR	37.0743	6.2913	37.4339	7.8740	0.2000	0.4700	0.0050
					SRK	37.2863	6.652	39.6686	7.4203	0.2200	0.5150	0.0050
		318.15	102.3–225.7	7	PR	31.6307	5.1715	33.2679	2.6531	0.2750	0.6500	-0.0050
					SRK	36.1244	5.1098	36.4133	2.7383	0.2850	0.6800	-0.0050
328.15		104.4–230.6	7	PR	31.5318	10.3430	30.3295	9.1046	0.2800	0.6800	-0.0050	
				SRK	29.0384	10.8282	30.4613	9.8902	0.2850	0.7000	-0.0050	
Propyl-4-hydroxy benzoate [46]	308.15	94.1–220.9	7	PR	13.5994	2.1176	14.3637	1.5054	0.2350	0.4200	-0.0050	
				SRK	15.6949	2.6009	17.2696	1.1920	0.2400	0.4200	-0.0050	
	318.15	96.8–214.7	7	PR	14.2196	8.1911	14.0412	2.7169	0.2850	0.5400	-0.0100	
				SRK	15.4313	8.8246	15.1762	3.0757	0.2850	0.5350	-0.0100	
	328.15	105.1–220.2	7	PR	18.0685	17.7706	18.0141	2.0883	0.4800	0.9700	-0.0350	
				SRK	18.6907	18.3623	19.0544	2.1674	0.4800	0.9800	-0.0400	
3-4-Xylenol [27]	308.15	82–262	7	PR	23.0053	8.6598	22.7541	1.5214	-0.0600	-0.1250	-0.0050	
				SRK	22.0859	9.2925	21.601	2.2491	-0.0050	-8.21E-16	-0.0100	
2,5-Xylenol [28]	308.15	87–267	7	PR	26.6876	6.0457	26.5655	1.8053	-0.0500	-0.0850	-0.0100	
				SRK	25.4857	7.1183	25.7471	1.5935	-0.0350	-0.0650	-0.0100	

Component	T(K)	P(bar)	N ^a	EOS	AARD(%)				k_{ij}	l_{ij}	m_{ij}
					VdW1	VdW2	CVD	This Work			
Naphthalene [49]	308.15	86.8–255.3	9	PR	11.2797	4.9591	11.1225	1.4325	0.0800	0.2000	-0.0100
				SRK	10.7920	5.6464	9.5160	1.6425	0.0850	0.2050	-0.0100
Naphthalene [49]	333.55	108.4–291.4	19	PR	5.9033	4.3151	5.63	3.8172	-0.1850	-0.2900	0.0400
				SRK	6.5240	4.1782	6.6265	4.0712	-0.0500	0.0550	0.0200
	338.05	151.8–232.2	7	PR	3.7213	1.9292	3.7518	1.0550	0.2150	0.6350	-0.0200
				SRK	5.0398	2.1961	4.2844	0.9533	0.1900	0.6000	-0.0200
Phenanthrene [48,50]	308.2	78.3–203.5	47	PR	19.3415	12.3779	18.7357	4.8184	0.1050	0.0900	-0.0050
				SRK	18.1881	13.1176	17.8897	5.6411	0.1200	0.1050	-0.0050
	318.2	95–254	20	PR	17.4822	7.7594	17.1394	4.2555	0.1100	0.1200	-0.0100
				SRK	15.9964	8.9711	15.1638	3.8964	0.1250	0.1400	-0.0100
	323.2	89.4–228.5	38	PR	17.7502	5.7279	18.0258	5.1786	0.0700	0.0250	-0.0050
				SRK	19.2049	6.7354	17.9363	5.4091	0.0800	0.0300	-0.0050
	328.2	90–245	23	PR	19.9165	4.8743	19.3509	3.1093	0.0850	0.0650	-0.0100
				SRK	19.9606	7.7785	19.3720	3.6395	0.1300	0.1600	-0.0150
Fluorene [19,48]	308.15	83.7–414.5	6	PR	30.0807	5.2473	28.0864	5.2473	0.2250	0.2800	4.34E-17
				SRK	33.891	6.0112	34.2151	6.3855	0.2500	0.3200	4.34E-17
	308.2	78.3–203.5	47	PR	14.2369	9.7064	13.1427	5.4411	0.2650	0.3650	-0.0050
				SRK	14.9606	10.5292	14.1889	5.0675	0.2650	0.3550	-0.0050
	323.15	83.7–414.5	8	PR	42.7946	15.2145	43.1622	15.2145	0.2350	0.2950	4.34E-17
				SRK	43.7543	11.7959	44.4848	11.7907	0.2550	0.3300	4.34E-17
	323.2	83.7–414.5	38	PR	16.3774	8.2093	16.9244	7.9118	0.1950	0.1950	0.0050
				SRK	15.9664	6.5014	16.5766	6.7457	0.2050	0.2050	0.0050
	328.2	85–245	24	PR	26.9329	11.2076	24.5408	10.0447	0.2650	0.3550	-0.0050
				SRK	21.3068	8.4739	22.7499	7.8974	0.2650	0.3500	-0.0050
2,6-Dimethyl naphthalene [35]	308.2	79–146	4	PR	33.3766	23.4231	31.8858	5.6955	-0.0300	-0.1350	-0.0050
				SRK	31.8981	24.1483	32.606	5.7398	0.1300	0.2300	-0.0150
	328.2	100–127	4	PR	6.6612	2.7170	6.2782	2.2257	0.0300	0.0350	-0.0050
				SRK	8.6343	2.4292	9.0369	2.8443	-0.0850	-0.2900	0.0050
2,7-Dimethyl naphthalene [35]	308.2	88–242	5	PR	14.8662	8.8744	14.7062	1.0942	0.1150	0.2000	-0.0150
				SRK	13.6194	9.8150	13.8143	1.0943	0.1400	0.2450	-0.0200
	328.2	100–249	5	PR	16.8066	3.3291	15.5319	1.9836	0.0400	0.0500	-0.0050
				SRK	16.1314	5.4781	15.8082	0.3379	0.0750	0.1250	-0.0100
O-hydroxy benzoic acid [51,52]	318.15	81.1–202.6	12	PR	22.9849	5.9596	22.9985	6.7869	-0.0500	-0.0900	-0.0050
				SRK	23.5465	6.2956	22.7284	6.7809	-0.0400	-0.0950	-0.0050
	328.15	101.3–202.6	11	PR	17.6547	3.4977	17.9277	3.3700	-0.0700	-0.1050	-0.0050
				SRK	17.5422	4.538	17.9908	4.1159	-0.0550	-0.0850	-0.0050
	328.15	101.3–202.6	6	PR	21.6606	1.7615	22.2208	1.6937	-0.0850	-0.1650	-0.0050
				SRK	21.7743	2.5635	22.3821	1.9378	-0.0700	-0.1500	-0.0050
P-hydroxy benzoic acid [52]	318.15	101.3–202.6	6	PR	19.8779	4.8131	19.5393	1.5374	-0.0450	-0.0650	-0.0050
				SRK	19.0202	5.2828	18.7005	1.9857	-0.0300	-0.0500	-0.0050
	328.15	101.3–202.6	6	PR	15.8269	4.6075	14.3667	3.9840	-0.0250	0.0450	-0.0050
				SRK	15.9321	3.4474	14.6654	2.7469	-0.0250	0.0200	-0.0050
1-Hexadecanol [53]	318	152.1–415.1	7	PR	32.3703	2.1009	32.3461	1.5887	0.1300	0.2750	-0.0050
				SRK	37.2722	3.0008	40.4952	1.6716	0.1700	0.3350	-0.0150
	328	141.8–415.9	5	PR	57.0294	9.8866	52.303	8.3203	0.1950	0.3500	-0.0050
				SRK	58.1061	9.9483	55.8279	7.9504	0.2300	0.4050	-0.0150
	338	147.1–373	6	PR	48.781	4.4550	53.802	2.4078	0.2700	0.4350	-0.0100
				SRK	54.967	4.7066	54.1103	2.8447	0.3100	0.5000	-0.0250

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Component	T(K)	P(bar)	N ^a	EOS	AARD(%)				k_{ij}	l_{ij}	m_{ij}
					VdW1	VdW2	CVD	This Work			
1-Octadecanol [54]	318	152–437.9	4	PR	9.5284	4.1363	9.5890	3.0092	0.0450	0.1250	-0.0100
				SRK	24.0953	4.2891	23.5500	3.3346	0.0900	0.1750	-0.0100
	328	139.9–447.7	7	PR	56.8142	8.4207	53.5752	4.9818	0.1750	0.3250	-0.0150
				SRK	58.2835	9.2823	59.0386	5.2869	0.2100	0.3700	-0.0200
	338	145.8–452.8	6	PR	66.9715	3.4795	67.0302	3.4795	0.2150	0.3400	4.34E-17
				SRK	71.4502	6.1522	66.6701	3.2837	0.2600	0.4100	-0.0150
Palmitic acid [53]	318	142.1–360.6	5	PR	30.4772	3.7517	33.0766	2.2303	0.1600	0.3050	-0.0100
				SRK	42.1376	5.1102	41.0469	1.8748	0.2200	0.3950	-0.0300
	328	144.1–573.5	7	PR	73.5056	5.9974	74.4539	6.6069	0.1500	0.2800	0.0050
				SRK	84.9557	6.6040	79.5333	6.6701	0.1950	0.3400	4.34E-17
	338	142.5–574.8	7	PR	95.1375	14.0898	93.5112	8.3035	0.2450	0.4400	-0.0200
				SRK	99.4231	15.3601	94.1093	7.8808	0.2800	0.4850	-0.0250
Stearic acid [54]	318	145.4–361.5	6	PR	11.0718	7.9148	10.8864	2.7284	0.0500	0.1500	-0.0400
				SRK	10.2141	9.0883	8.8103	3.2388	0.1150	0.2450	-0.0700
	328	154.8–467.5	6	PR	57.1116	18.3888	60.0238	4.8699	0.0250	0.0900	0.0350
				SRK	71.1858	16.6831	71.7080	7.2859	0.0900	0.1850	0.0250
	338	161.5–463.8	5	PR	56.9730	11.7971	53.8029	4.3336	0.1950	0.4150	-0.0400
				SRK	63.6315	13.3366	59.1502	3.6326	0.2250	0.4500	-0.0500

a N is the number of data points

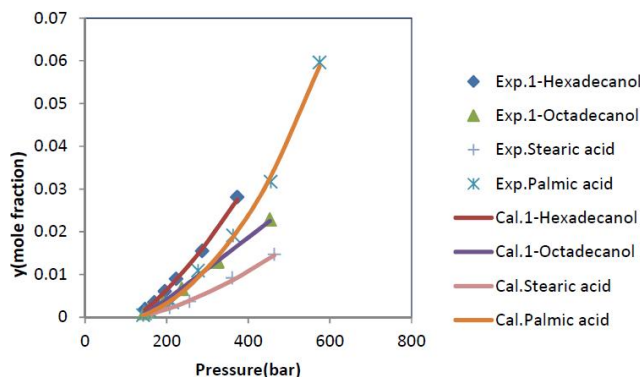


Figure 1 : Solubility of solid components in supercritical CO₂ using the PR EOS at T = 338K in the new mixing rule (this work)

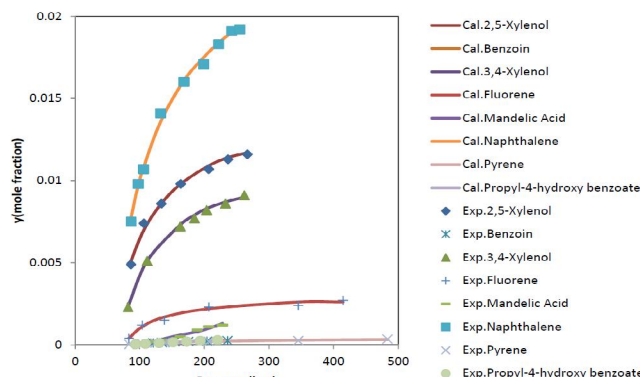


Figure 3 : Solubility of solid components in supercritical CO₂ using the PR EOS at T = 308.15K in the new mixing rule (this work).

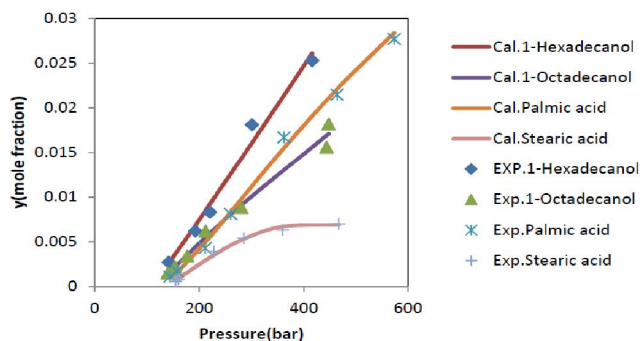


Figure 2 : Solubility of solid components in supercritical CO₂ using the SRK EOS at T = 328K in the new mixing rule (this work)

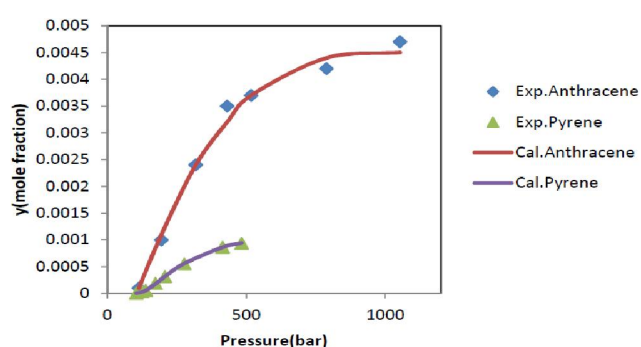


Figure 4 : Solubility of solid components in supercritical CO₂ using the PR EOS at T = 343.15K in the new mixing rule (this work)

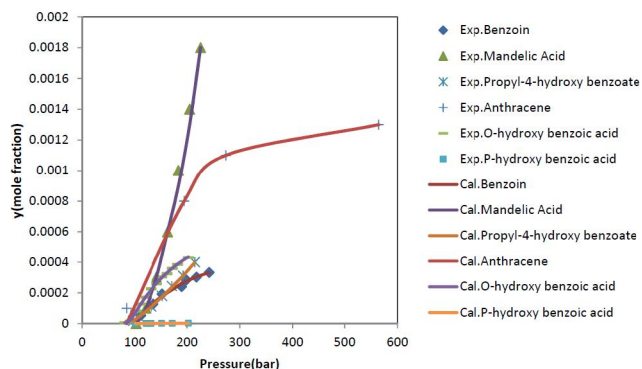


Figure 5 : Solubility of solid components in supercritical CO₂ using the PR EOS at T = 318.15K in the new mixing rule (this work)

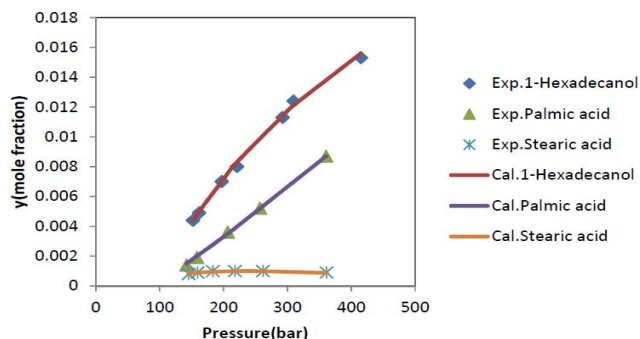


Figure 9 : Solubility of solid components in supercritical CO₂ using the SRK EOS at T = 318K in the new mixing rule (this work)

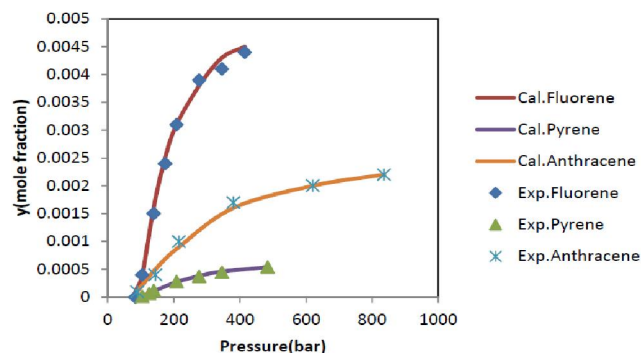


Figure 6 : Solubility of solid components in supercritical CO₂ using the PR EOS at T = 323.15K in the new mixing rule (this work).

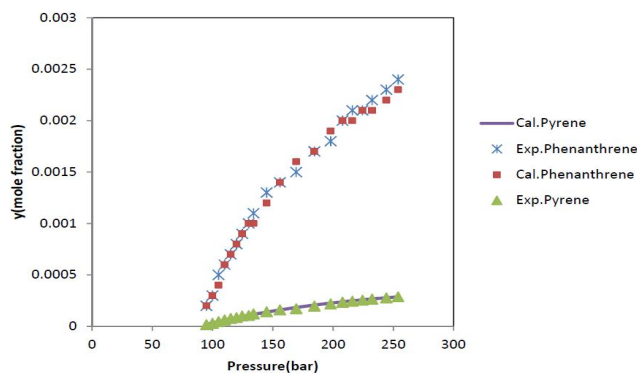


Figure 10 : Solubility of solid components in supercritical CO₂ using the SRK EOS at T = 318.2K in the new mixing rule (this work)

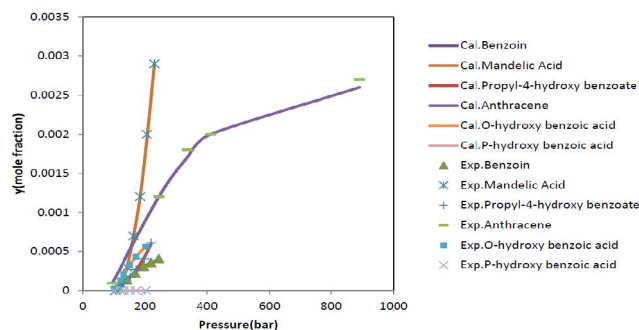


Figure 7 : Solubility of solid components in supercritical CO₂ using the PR EOS at T = 328.15K in the new mixing rule (this work)

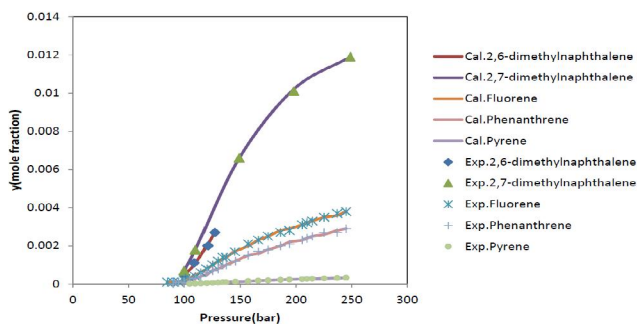


Figure 11 : Solubility of solid components in supercritical C₉O₂ using the SRK EOS at T = 328.2K in the new mixing rule (this work)

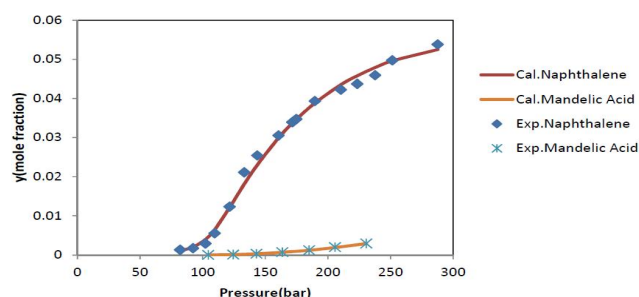


Figure 8 : Solubility of solid components in supercritical CO₂ using the PR EOS at T = 328.15K in the new mixing rule (this work)

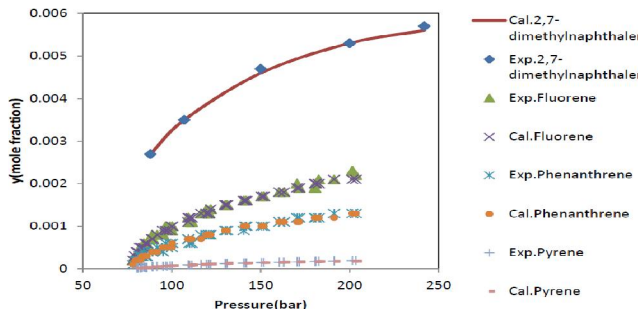


Figure 12 : Solubility of solid components in supercritical CO₂ using the SRK EOS at T = 308.2K in the new mixing rule (this work)

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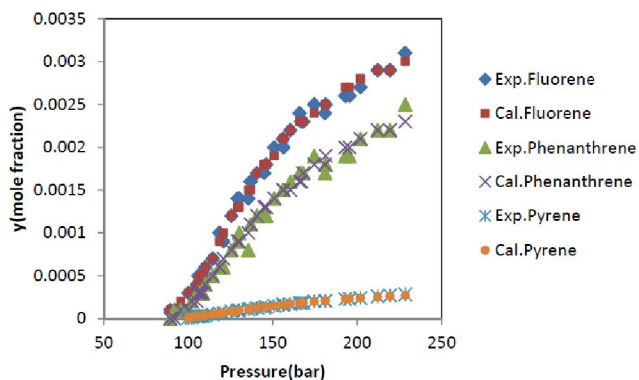


Figure 13 : Solubility of solid components in supercritical CO₂ using the SRK EOS at T = 323.2K in the new mixing rule (this work)

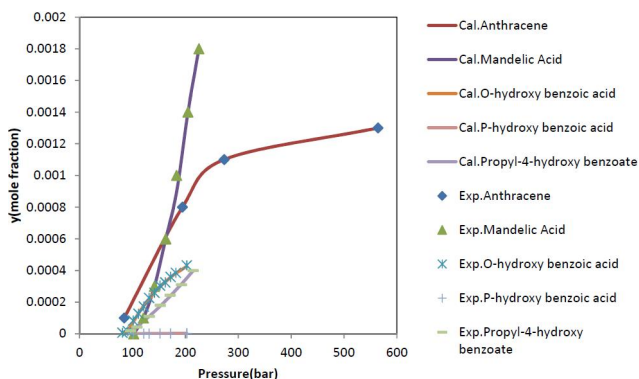


Figure 14 : Solubility of solid components in supercritical CO₂ using the SRK EOS at T = 318.15K in the new mixing rule (this work)

dioxide by using PR EOS at 308.15K. In this way, Figures 4-14 demonstrate our calculated results with the experimental data for different solids in supercritical carbon dioxide at various temperature and pressure conditions. In all systems, our predicted results are in close agreement to the experimental data.

NOMENCLATURE

a	energy parameter of a cubic equation of state
AARD	absolute average relative deviation
b	co-volume of a cubic equation of state
B	second virial coefficient
c	speed of light in vacuum [m/s]
$\dot{A}, \dot{B}, \dot{C}$	Antoine equation parameters
A_s	dispersion constant [J.cm ⁶ /molecule ²]
d	molecular diameter [\AA]
EOS	equation of state
f	fugacity

i, j	components i, j
k_{ij}, l_{ij}, m_{ij}	interaction coefficients between components of i and j
m	parameter in Eq. (10)
m	rest mass of electron [kg]
M	molecular weight
n	refractive index
N	number of experimental points
N_A	avogadro's number [molecule/mol]
N_S	number of solid molecules/surface unit
P	pressure [bar]
P_C	critical pressure [bar]
PR	Peng-Robinson equation of state
Q	fitting parameter [m ³]
R	universal gas constant
p, q, r, \hat{K}, \hat{L}	parameters in Eqs. (26,27)
SRK	Soave-Redlich-Kwong equation of state
T	temperature [K]
T_c	critical temperature [K]
T_r	reduced temperature
U, W	parameters in Eq.(6)
y_i	mole fraction of component i in the supercritical phase
Z	intermolecular distance between CO ₂ and solid molecule
Z	compressibility factor

Greek symbols

v	molar volume [liter mol ⁻¹]
ϕ	fugacity coefficient
ω	acentric factor
σ	intermolecular distance at zero-interaction energy [\AA]
α	parameter in Eq. (9)
α_A, α_s	polarizabilities of CO ₂ and solid [cm ³ /molecule]
χ	magnetic susceptibility [cm ³ /molecule]
ϵ	potential energy interaction [10 ³² J/molecule]
ρ	absolute density

Superscripts

exp	experimental
calc	calculation
s	solid phase
sat	saturation

CONCLUSION

In the current work, the cubic Peng–Robinson (PR) and Soave–Redlich–Kwong (SRK) equations of state have been used to estimate the solid solubilities of 18 solutes in supercritical carbon dioxide by using four mixing rules called the van der Waals one fluid rule with one (VDW1) and two (VDW2) adjustable parameters, the covolume dependent (CVD) rule and the new mixing rule. Furthermore, the optimized adjustable parameters of the new mixing rule are reported for 665 experimental data points. The prediction of the new mixing rule model for solid solubility calculations in supercritical carbon dioxide is more accurate than the other mixing rules in the same equations of state.

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