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Research Article

Solubility Modeling of Some Chlorophenols in Supercritical Carbon Dioxide

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Abstract: The basic information required to implement any supercritical processes is solubility. The solubility data is correlated with various models. For binary systems (solute-solvent), solubility data is limited in the literature, which makes it's modelling essential. Equation of state (EoS) method is one of the best techniques to model solubility data. For EoS modelling to be implemented, critical properties of the solute and the solvent are essential but, the exact realistic properties are sparse for solutes. The present work deals with the modelling of the solubility of solid solutes in supercritical carbon dioxide solvent with and without critical properties of the solute. Redlich-Kwong (RK) EoS along with Kwak-Mansoori (KM) mixing rules is used for the solubility data modelling. In this work, three substituted phenols and supercritical carbon dioxide systems are considered for modelling. Finally, the correlating abilities of RK EoS along with KM mixing rules with and without critical properties of the solute are indicated in terms of the average absolute relative deviation percentage (AARD%) and Akaike information criterion (AIC).

***Keywords*:** supercritical CO_2, solubility modeling, Kwak-Mansoori mixing rules, RK EoS, chlorophenols

Symbols

a [J·m^3/mol]	Pure component property in PREOS
b [m^3/mol]	RK EoS volume correction parameter
C_1, C_2	Molar volume expression constants
k_{ij} [-]	Binary interaction parameter in EOS
l_{ij} [-]	Binary interaction parameter in PREOS
N_i [-]	Number of data points
P [MPa]	Pressure
Q [-]	Number of data points
R [J/mol·K]	Universal gas constant
T [K]	Temperature
v [m^3/mol]	Molar volume in RK EoS

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v_2 [m^3/mol]	Molar volume of the solute
y_2 [-]	Mole fraction paraben compound

Greek letters

ρ_1 [$kg \cdot m^{-3}$]	SC-CO_2 Density
α [-]	RK EoS parameter
ω [-]	Acentric factor
$\hat{\phi}$ [-]	Fugacity coefficient of component in mixture

Superscript and Subscript

1	Solvent
2	Solute
cal	Calculated mole fraction
exp	Experimental mole fraction
S	Pure solid
SC-CO_2	Supercritical Carbon Dioxide

Abbreviations

AARD	Average Absolute relative deviation
AIC	Akaike's information criterion
AIC_c	Corrected Akaike's information criterion
EoS	Equation of state
KM	Kwak-Mansoori
RK	Redlich-Kwong

1. Introduction

Industries using supercritical fluid technologies are increasing which include petrochemical, biotechnology, pharmaceuticals, natural products and dyeing industries.[1] In all industrial applications the supercritical fluid acts as green solvent that targets the desired solutes. The vividly used supercritical fluids are carbon dioxide and water.[1,2] However, carbon dioxide can be taken to the supercritical state with lesser energy requirements than water. This makes it a well-sought supercritical solvent in the process industry. Due to this, its applications have been quite evident in the dyeing, food processing, and pharmaceutical industries in the recent past.[3-7] For proper implementation, one of the basic properties required is the solubility information of the solute in the supercritical fluid (solvent). Supercritical fluids replace conventional organic solvents, which are widely used in process industries and the end products of supercritical processes are nontoxic due to the absence of the organic solvent.[8-11] Supercritical carbon dioxide (SC-CO_2) differs from other solvents in that it is non-toxic, non-flammable, and has a tuneable density. It can be used instead of conventional organic solvents. It has a moderate critical pressure of 7.39 MPa and a mild critical temperature of 304.12 K.[3] Complete understanding of the affecting variables on the compound's solubility is required for the successful application of SC-CO_2 based technology in industry. Since getting solubility data at the necessary conditions (pressure and temperature) is a difficult process, modelling is essential for solubility data interpolation.[3]

In the literature, various methods have been used in recent years to model solubility data.[12-15] These methods are explicitly categorised into five types namely density models (or) mathematical models, equation of state models, and solid-liquid equilibrium models, intelligent models, and artificial neural networks models.[4,16-18] Wherein three models are simple to use namely, mathematical, equations of state (EoS), and density models.[5-7] The EoS methods requires information about the compounds' physical characteristics, like vapour pressure, molar volume, critical values, and acentric factor.[8-12] The mathematical and density models require pressure, temperature, and solvent density information. If all the necessary data were available the EoS model would be the best. Unfortunately, the necessary physiochemical properties are unavailable for many compounds.[5-7] On the other side, the other methods that don't require this information have drawn greater interest and proven to be more useful.[3,10]

Density and mathematical models are quite effective at modelling solubility due to their ease of use.[3,10] However, equilibrium solubility is a phase equilibrium study, and understanding phase behaviour with EoS approach is meaningful. Hence, it is necessary to address solubility through the EoS models.

In literature, there are a numerous number of EoS models and all those models requires critical properties of the solute, molar volume of the solute and sublimation pressure of the solute along with fugacity coefficient, system pressure and system temperature.[5,6,13,17] The required critical properties are estimated using group contribution methods.[6] For many complex solutes' critical properties, molar volume and sublimation pressure are unavailable and at the same time existing group contribution methods are also not applicable due to the lack of functional group contributions in their techniques.[7] Due to this, there is a need to address EoS methodology without using solute's critical properties. Therefore in the present study, a new EoS methodology is proposed for which critical properties of the solute, molar volume of the solute are not required. Thus, this study is focused on the development of new solubility model using equation of state in its limiting form.

In the present work, the Redlich-Kwong (RK) EoS along with KM mixing rules[18] is used to model the equilibrium solubility of three chlorophenols (4-chlorophenol, 2,4-dichlorophenol and 2,4,6-trichlorophenol)[19] without considering the solute's critical properties (T_C & P_C), vapour pressure, acentric factor, and molar volume. Chlorophenol are used in synthesis of dyes, drugs, insecticides and germicides thus their solubility modelling in SC-CO_2 is important.[19] Micronization/nanonization of chlrophenols using supercritical carbon dioxide (SC-CO_2) requires exact solubility data, but it is a quite difficult task to get the exact required solubility data at every pressure and temperature. Therefore, there is a need to focus on the model development that gives necessary solubility data. Thus, the proposed work fulfils the requirement. To know the efficacy of the proposed modelling, the correlations are compared to the same modelling done with the solutes' critical properties taken into account. More details about the RK EoS, KM mixing rules with and without the solute's critical properties are discussed in the following section.

2. Modelling

EoS model for the solubility of solids in SC-CO_2.

The solubility of drug i(solute)in SC-CO_2 (solvent) is expressed as[20,21]:

$$y_2 = \frac{p_2^S \hat{\phi}_2^S}{P\hat{\phi}_2^{SC-CO_2}} \exp\left[\frac{\left(P - p_2^S\right)V_2}{RT}\right] \tag{1}$$

where p_2^S is the sublimation pressure of the solute and other parameters have usual meanings. The fugacity coefficient of the solid solute at saturation ($\hat{\phi}_2^S$) is usually taken to be unity. The fugacity coefficient($\hat{\phi}_2^{SC-CO_2}$) of the solute in SC-CO_2 is from eq. (2) for the combination of RK EoS and KM mixing rules

$$\ln\left(\hat{\phi}_2^{SC-CO_2}\right) = \frac{1}{RT}\int_v^\infty \left[\left(\frac{\partial P}{\partial n_1}\right)_{T,V,n_2} - \frac{RT}{v}\right] dv - \ln Z \tag{2}$$

2.1 *RK EoS with KM mixing rules*[18]

RK EoS in terms of compressibility factor (Z) is given by

$$Z = \frac{v}{v-b} - \frac{a}{RT^{1.5}\left(v+b\right)} \tag{3}$$

where $a = 0.42748R^2T_c^{2.5}/P_c$; $b = 0.8664RT_c/P_c$.

The following are the mixing rules that are statistically consistent with van der Walls

$$a = \frac{\left(\sum_i^n\sum_j^n x_i x_j a_{ij}^{2/3} b_{ij}^{1/3}\right)^{3/2}}{\left(\sum_i^n\sum_j^n x_i x_j b_{ij}^{1/2}\right)^{1/2}} \tag{4}$$

$$b = \sum_i^n\sum_j^n x_i x_j b_{ij} \tag{5}$$

$$a_{ij} = \left(1-k_{ij}\right)\sqrt{a_{ii}a_{jj}} \tag{6}$$

$$b_{ij} = \left(1-l_{ij}\right)\frac{\left(b_{ii}^{1/3} + b_{jj}^{1/3}\right)^3}{8} \tag{7}$$

eq. (4) to eq. (7), when combined with RK EoS, represents EoS for mixtures.

The Fugacity coefficients for RK EoS is

$$\ln\left(\hat{\phi}_2^{SC-CO_2}\right) = \ln\left(\frac{v}{v-b}\right) + \left(\frac{2\sum x_j b_{ij} - b}{v-b}\right) - \ln\left(Z\right) +$$

$$\left(\frac{a\left(2\sum x_j b_{ij} - b\right)}{b^2RT^{3/2}}\right)\left[\ln\left(\frac{v+b}{v}\right) - \frac{b}{v+b}\right]\left(3\alpha^{1/2}\left(\sum x_j a_{ij}^{2/3} b_{ij}^{1/3}\right)\Big/ b^{1/2} - \alpha^{2/3}\left(\sum x_j b_{ij}\Big/ b^{3/2}\right)\right)\Big/ bRT^{3/2} \tag{8}$$

From the above it is clear that the adjustable constants are k_{ij} and l_{ij} when RK EoS is used in the eq. (1).

2.2 *RK EoS (limiting case)*

This approach can be used when the solute data is not available**.** The major limitations in using eq. (1) is the availability of necessary data such as critical properties of the solute, molar volume of the solute and vapour pressure**.** Limiting case solution is obtained by keeping $x_1 \rightarrow 1$ and $x_2 \rightarrow 0$.

The corresponding mixing rules and activity coefficient reduce to the following:

$$a = a_{11} \tag{9}$$

$$b = b_{11} \tag{10}$$

$$a_{12} = \sqrt{a_{11} a_{22}} \tag{11}$$

$$b_{12} = \left(\frac{b_{11}^{1/3} + b_{22}^{1/3}}{2} \right)^3 \tag{12}$$

$$Z = Z_{11} \tag{13}$$

$$\ln\left(\hat{\phi}_2^{SC-CO_2}\right) = \ln\left(\frac{v}{v-b_{11}}\right) + \left(\frac{2\sum x_j b_{ij} - b_{11}}{v-b_{11}}\right) - \ln\left(Z_{11}\right) +$$

$$\left(\frac{a\left(2\sum x_j b_{ij} - b_{11}\right)}{b_{11}{}^2 RT^{3/2}}\right)\left[\ln\left(\frac{v+b_{11}}{v}\right) - \frac{b_{11}}{v+b_{11}}\right]\left(3\alpha^{1/2}\left(\sum x_j a_{ij}^{2/3} b_{ij}^{1/3}\right) \Big/ b_{11}{}^{1/2} - \alpha^{2/3}\left(\sum x_j b_{ij} \Big/ b_{11}{}^{3/2}\right)\right) \Big/ b_{11} RT^{3/2} \tag{14}$$

where $\alpha = a_{11}{}^{2/3} b_{11}$.

The molar volume of the solute is expressed as a function of density[22] as

$$v_2 = \exp\left(C_1 \ln(\rho_1) + C_2\right) \tag{15}$$

From the above, it is clear when the solute vapour pressure data is available, the adjustable constants are a_{22}, b_{22}, C_1 and C_2 when RK EoS (limiting case) is used in the eq. (1)

3. Results and discussion

In the work, two forms of RK EoS and KM mixing rules are examined for the solubility modelling. The validation of both forms was done with three literature available substituted chlorophenols' solubility data. Table 1 indicates the summary of the compounds used in the work. The main reasons to consider these compounds are, EoS correlations for substituted phenols are not reported in the literature, and the vapour pressure data of solutes are readily available in the literature.[23] The summary of vapour pressure data is indicated in Table 2. The AARD% is used to obtain the proposed model parameters, and it is given as[24] eq. (16)

$$AARD\% = \frac{100}{N} \sum_{i=1}^{N} \frac{\left| y_{2i}^{\exp} - y_{21}^{calc} \right|}{y_{2i}^{\exp}} \tag{16}$$

Table 1. Summary of Chlorophenol solubility data in SC-CO_2[19]

System Solute-solvent	Solute formula	Temperature range, K	Pressure range, bar	Mole Fraction range	Solubility in g/l
4-Chlorophenol-SC-CO_2	C_6H_5ClO	308-313	88-156	0.0038-0.0238	1.485-56.291
2,4-Dichlorophenol-SC-CO_2	$C_6H_4Cl_2O$	308-313	88-156	0.0097-0.0702	15.778-221.036
2,4,6-Trichlorophenol-SC-CO_2	$C_6H_3Cl_3O$	308-318	88-156	0.0013-0.02	1.843-69.063

Table 2. Chlorophenols sublimation pressures[23]

Compound	Sublimation pressures of Chlorophenols (Where P^*/Pa and T/K)
4-Chlorophenol	$\ln(P^*) = (297.9/R) - (84019.4/(RT)) - (23.1/R)\ln(T/298.15)$
2,4-Dichlorophenol	$\ln(P^*) = (309.5/R) - (85326.1/(RT)) - (24.7/R)\ln(T/298.15)$
2,4,6-Trichlorophenol	$\ln(P^*) = (306.3/R) - (90278.2/(RT)) - (26.9/R)\ln(T/298.15)$

The required critical properties and acentric factor data are estimated using group contribution methods (Ambrose and Lee-Kesler methods)[25-27] and are presented in Table 3. The model results for the RK EoS along with KM mixing rules and the RK EoS limiting case, are shown in terms of the average absolute relative deviation percentage (AARD %) in Table 4. Figures 1, 2 and 3 indicate the correlating ability of both the models. It can be inferred that both models are able to correlate the data with similar AARD%.The two forms have a variable number of parameters in their model. The relative efficacy is examined in terms of the Akaike information criterion (AIC) and the corrected AIC (AIC_c).[28-32] They are represented as

$$AIC_c = AIC + \frac{2Q(Q+1)}{N-Q-1} \tag{17}$$

where

$$AIC = N\ln\left(\sigma^2\right) + 2Q \tag{18}$$

In eqs. (17) and (18), σ, N, and Q represent the variance of deviations, number of data points, and model parameters, respectively.

The data used for each compound is less than 40. AIC_c is recommended to know the best model. Table 5 indicates the AIC_c values for the models considered in the work. From the tabulated values, both models are performing on par with each other. Thus, the RK EoS limiting case, which only uses solvent critical properties, is also recommended for data correlation.

Table 3. Summary of critical properties of Chlorophenols

Solute	${}^{a}T_c$, K	${}^{a}P_c$, bar	${}^{b}\omega$	${}^{c}V_2$, m^3/mol
4-Chlorophenol	735	51.8	0.4677	0.89047×10^{-4}
2,4-Dichlorophenol	712.8	45.5	0.476	1.02289×10^{-4}
2,4,6-Trichlorophenol	766.1	30.03	0.489	1.1554×10^{-4}

[a]Ambrose method[25]; [b]Lee-Kesler method[25]; [c]Immirzi and Perini method[26,27]

Table 4. Correlation results of Chlrophenols-SC-CO_2 systems

System	Correlation constants	R^2	AARD %
	RK EoS correlations		
4-Chlorophenol-SC-CO_2	$k_{12} = -0.1685$; $l_{12} = -0.1802$	0.982	4.21
2,4-Dichlorophenol-SC-CO_2	$k_{12} = -0.0278$; $l_{12} = -0.0559$	0.965	5.68
2,4,6-Trichlorophenol-SC-CO_2	$k_{12} = 0.0378$; $l_{12} = -0.0155$	0.979	7.32
	RK EoS correlations for limiting case		
4-Chlorophenol-SC-CO_2	$a_{22} = 157.44$; $b_{22} = 2.2379 \times 10^{-4}$; $C_1 = -6.9848$; $C_2 = 3.1574$	0.981	4.42
2,4-Dichlorophenol-SC-CO_2	$a_{22} = 159.46$; $b_{22} = 2.1776 \times 10^{-4}$; $C_1 = -7.2252$; $C_2 = 3.2623$	0.951	6.91
2,4,6-Trichlorophenol-SC-CO_2	$a_{22} = 189.68$; $b_{22} = 2.4901 \times 10^{-4}$; $C_1 = -2.3398$; $C_2 = 4.5515$	0.982	6.56

Table 5. Statistical indicator of the Chlorophenol-SC-CO_2 systems

System	Parameters	Data points	SSE	AIC	AIC_c
		RK EoS correlations			
4-Chlorophenol-SC-CO_2	2	16	9.07×10^{-6}	-226	-226
2,4-Dichlorophenol-SC-CO_2	2	16	1.99×10^{-4}	-177	-176
2,4,6-Trichlorophenol-SC-CO_2	2	24	1.61×10^{-5}	-337	-337
		RK EoS correlations for limiting case			
4-Chlorophenol-SC-CO_2	4	16	7.79×10^{-6}	-225	-223
2,4-Dichlorophenol-SC-CO_2	4	16	2.76×10^{-4}	-167	-166
2,4,6-Trichlorophenol-SC-CO_2	4	24	1.41×10^{-5}	-336	-335

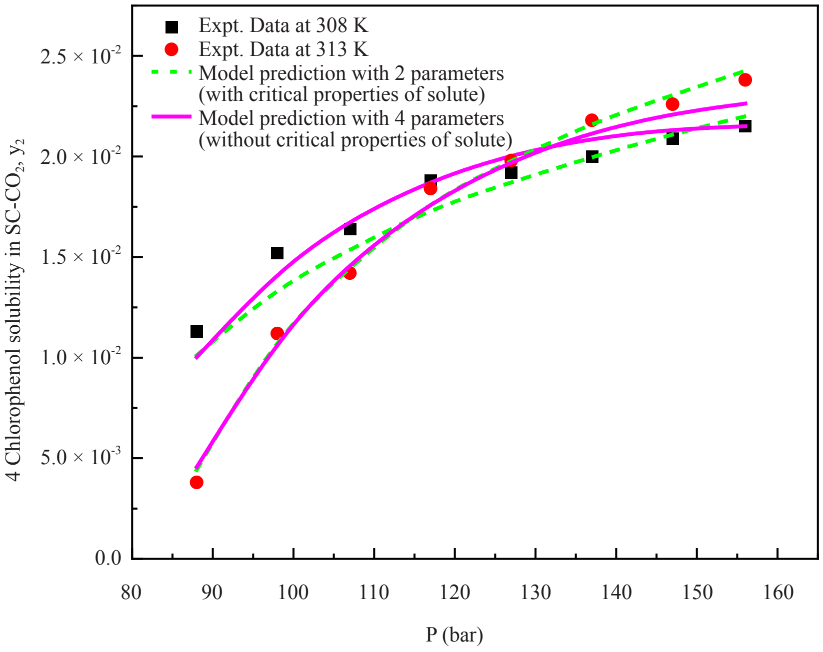

Figure 1. 4-Chlorophenol solubility in SC-CO$_2$, vs. P

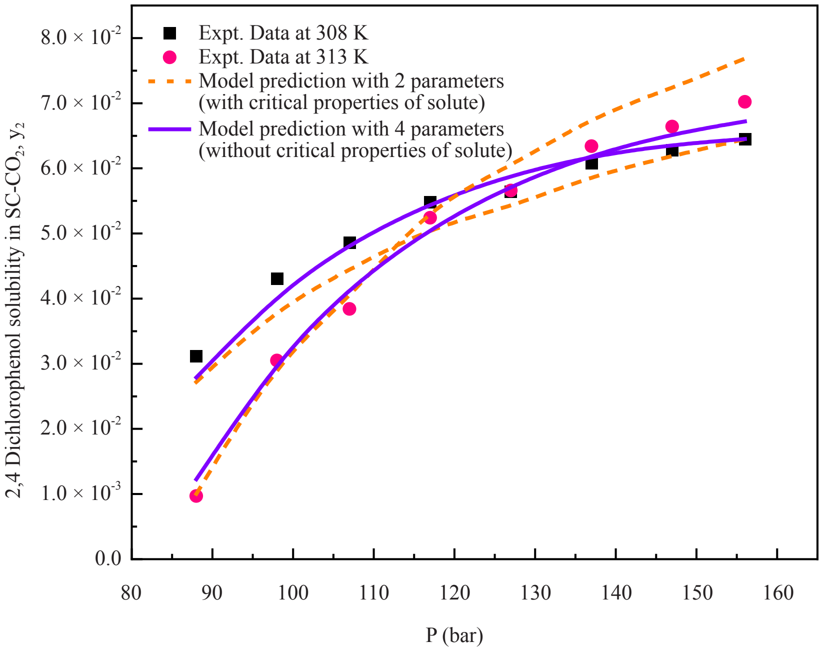

Figure 2. 2,4-Dichlorophenol solubility in SC-CO$_2$, vs. P

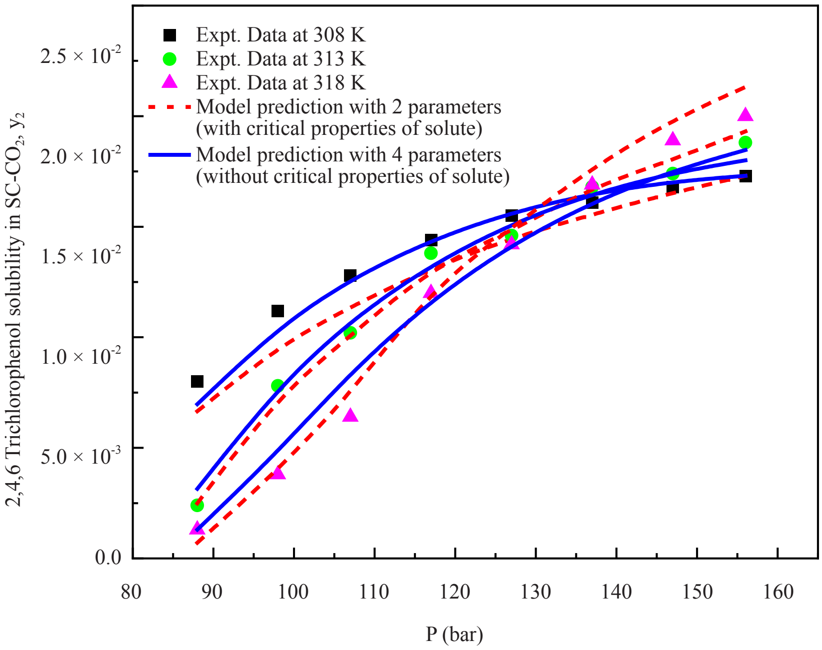

Figure 3. 2,4,6-Trichlorophenol solubility in SC-CO$_2$, vs. P

4. Conclusion

The solubilities of some chlorophenols in SC-CO$_2$ are examined with the RK EoS along with the KM mixing rules and their limiting case as well. The correlations were developed with and without the data of chlorophenols' critical properties. It is observed that both forms can correlate the data satisfactorily. The Akaike information criterion (AIC) also reveals that both forms can correlate the data on par. Thus, the limiting case method is recommended for data correlation when the solute's critical properties are unavailable. The same methodology can be extended to any solute-supercritical fluid solvent system with appropriate EoS and mixing rules.

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Data sharing policy

The data that supports the findings of this study are available from authors of this article.

Compliance with ethical standards

This article does not contain any studies with human or animal subjects.

Conflict of interest

The authors declare no competing financial interest.

References

[1] Sodeifian, G.; Usefi, M. M. B. Solubility, extraction, and nanoparticles production in supercritical carbon dioxide. *Chem. Bio. Eng. Rev.* **2022,** *10*, 1-35.

[2] Martin, A.; Cocero, M. Fundamentals of modelling of supercritical precipitation process. In *Current Trends of Supercritical Fluid Technology in Pharmaceutical, Nutraceutical and Food Processing Industries*; Duarte Ana, R. C.; Duarte Catarina, M. M., Eds.; Bentham Science Publishers, Sharjah, United Arab Emirates, 2010; pp 41-51.

[3] Reddy, T. A.; Garlapati, C. Dimensionless empirical model to correlate pharmaceutical compound solubility in supercritical carbon dioxide. *Chem. Eng. Technol*. **2019,** *42*, 2621-2630.

[4] Sodeifian, G.; Garlapati, C.; Hazaveie, S. M.; Sodeifian, F. Solubility of 2,4,7-triamino-6-phenylpteridine (triamterene, diuretic drug) in supercritical carbon dioxide: experimental data and modeling. *J. Chem. Eng. Data.* **2020,** *65*, 4406-4416.

[5] Sodeifian, G.; Garlapati, C.; Roshanghias, A. Experimental solubility and modelling of crizotinib (anti-cancer medication) in supercritical carbon dioxide. *Sci. Rep.* **2022,** *12*, 17494.

[6] Alwi, R. S.; Garlapati, C. New correlations for the solubility of anticancer drugs in supercritical carbon dioxide. *Chem. Pap.* **2022,** *76*, 1385-1399.

[7] Sodeifian, G.; Garlapati, C.; Razmimanesh, F.; Nateghi, H. Solubility measurement and thermodynamic modelling of pantoprazole sodium sesqihydrate in supercritical carbon dioxide. *Sci. Rep.* **2022,** *12*, 7758.

[8] Sodeifian, G.; Garlapati, C.; Razmimanesh, F.; Nateghi, H. Experimental solubility and thermodynamic modelling of empagliflozin in supercritical carbon dioxide. *Sci. Rep*. **2022,** *12*, 9008.

[9] Alwi, R. S.; Garlapati, C. A new semi empirical model for the solubility of dyestuff in supercritical carbon dioxide. *Che. Pap.* **2021,** *75*, 2585-2595.

[10] Alwi, R. S.; Garlapati, C. A new model for the estimation of thermodynamic parameters for the solubility of azobenzene and anthraquinone derivatives in supercritical carbon dioxide. *Che. Pap.* **2021,** *75*, 4589-4610.

[11] Mahesh, G.; Garlapati, C. Modelling of solubility of some parabens in supercritical carbon dioxide and new correlations. *Arab. J. Sci. Eng.* **2021,** *47*, 5533-5545.

[12] Garlapati, C.; Madras, G. Temperature independent mixing rules to correlate the solubilities of antibiotics and anti-inflammatory drugs in SC-CO_2. *Thermochemica Acta.* **2009,** *496*, 54-58.

[13] Valderrama, J. O. The state of the cubic equations of state. *Ind. Eng. Chem. Res.* **2003,** *42*, 1603-1618.

[14] Alwi, R. S.; Rojas, A.; Esfandiari, N.; Sajadian, S. A.; Ardestani, N. S.; Jouyban, A. Experimental study and thermodynamic modeling of clonazepam solubility in supercritical carbon dioxide. *Fluid Phase Equilb.* **2023,** *574*, 113880.

[15] Ardestani, N. S.; Sajadian, S. A.; Esfandiari, N.; Rojas, A.; Garlapati, C. Experimental and modeling of solubility of *sitagliptin phosphate*, in supercritical carbon dioxide: proposing a new association model. *Sci. Rep.* **2023,** *13*, 17506.

[16] Esfandiari, N.; Ardestani, N. S.; Alwi, R. S.; Rojas, A.; Garlapati, C.; Sajadian, S. A. Solubility measurement of verapamil for the preparation of developed nanomedicines using supercritical fluid. *Sci. Rep.* **2023,** *13*, 17089.

[17] Ardestani, N. S.; Sajadian, S. A.; Rojas, A.; Alwi, R. S.; Estévez, L. A. Solubility of famotidine in supercritical carbon dioxide: experimental measurement and thermodynamic modeling. *J. Supercrit. Fluids* **2023,** *201*, 106031.

[18] Kwak, T. K.; Mansoori, G. A. Van der waals mixing rules for cubic equations of state: Applications for supercritical fluid extraction modelling. *Chem. Eng. Sci.* **1986,** *41*, 1303-1309.

[19] Garlapati, C.; Madras, G. Solubility of some chlorophenols in supercritical CO_2 in the presence and absence of cosolvents. *J. Chem. Eng. Data*. **2010,** *55*, 273-277.

[20] Orbey, H.; Sandler, S. I. *Modeling Vapor-Liquid Equilibria: Cubic Equations of State and Their Mixing Rules*; Cambridge University Press, New York, 1998.

[21] Prausnitz, J. M.; Lichtenthaler, R. N.; Gomes de Azevedo, E. *Molecular Thermodynamics of Fluid-Phase Equilibria*; Prentice-Hall, Inc: New Jersey, 1999.

[22] Iwai, Y.; Koga, Y.; Kukuda, T.; Arai, Y. Correlation of solubilities of high-boiling components in supercritical carbon dioxide using a solution model. *J. Chem. Eng. Jpn.* **1992,** *25*, 757-760.

[23] Verevkin, S. P.; Emel'yaneko, V. N. Thermochemistry of chlorobenzenes and chlorophenols: ambient temperature vapor pressures and enthalpies of phase transitions. *J. Chem. Eng. Data*. **2007,** *52*, 499-510.

[24] Valderrama, J. O.; Alvarez, V. H. Correct way of reporting results when modelling supercritical phase equilibria using equations of state. *Can. J. Chem. Eng*. **2005,** *83*, 578-581.

[25] Reid, R. C.; Prausnitz, J. M.; Poling, B. E. *The Properties of Gases and Liquids*; McGraw-Hill, 1987.

[26] Lyman, W.; Reehl, W.; Rosenblatt, D. *Handbook of Chemical Property Estimation Methods*; Mc Graw Hill, 1982.

[27] Immirzi, A.; Perini, B. Prediction of density in organic crystals. *Acta Crystallogr., Sect. A: Cryst. Phys., Diffr., Theor. Gen. Crystallogr.* **1977,** *33*, 216-218.

[28] Akaike, H. Information theory and an extension of the maximum likelihood principle. In *Proceedings of the 2nd International Symposium on Information Theory*; Petrov, B. N.; Csaki, F., Eds.; AkademiaiKiado, Budapest, 1973; pp 267-281.

[29] Akaike, H. A new look at the statistical model identifcation. *IEEE Trans. Autom. Control.* **1974,** *19*, 716-723.

[30] Kletting, P.; Glatting, G. Model selection for time-activity curves: the corrected akaike information criterion and the F-test. *J. Med. Phys.* **2009,** *19*, 200-206.

[31] Burnham, K. P.; Anderson, D. R. Multimodel inference: Understanding AIC and BIC in model selection. *Sociol. Methods Res*. **2004,** *33*, 261-304.

[32] Deepitha, J.; Pitchaiah, K. C.; Brahmmananda Rao, C. V. S.; Madras, G.; Sivaraman, N. Solubilities of dialkylhydrogen phosphonates in supercritical carbon dioxide and their correlation using semi-empirical equations. *Sep. Sci. Technol.* **2019,** *54*, 1650-1660.