Modelling of the Carbon Dioxide + Cyclohexane Binary System with Cubic Equations of State

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This study compares the ability of different thermodynamic models to calculate the phase behaviour of the carbon dioxide + cyclohexane binary system. All available literature data (47 data sets, 348 experimental points) for the aforementioned system were modelled with the ***G****eneral* ***E****quation* ***o****f* ***S****tate (GEOS) and Soave-Redlich-Kwong (SRK) equations of state coupled with classical van der Waals (two-parameter conventional mixing rule, 2PCMR) mixing rules or with Huron-Vidal (HV) mixing rules and a reduced UNIQUAC model. All models perform similarly when correlating the data and the absolute average deviations in pressure (AADP, %) are small, though sometimes the type of phase behaviour is wrong. A semi-predictive approach is also shown, which leads to the correct type of phase behaviour and to higher errors in the predicted pressures.*

Keywords: carbon dioxide, cyclohexane, GEOS, SRK, EoS, high-pressures

The phase behaviour of the carbon dioxide + naphthenic hydrocarbons is of interest in several fields such as processing of petroleum products, enhanced oil recovery, carbon capture and storage, production of coal liquids, etc. [1]. In particular, the carbon dioxide + cyclohexane system has been also studied in the context of the removal of organic contaminants or for improving the amount of smoke, soot, and invisible particles emitted with the exhaust of engines running on diesel fuel [2]. Besides its practical importance, the carbon dioxide + cyclohexane system was chosen as vapour-liquid equilibrium data are available in wide range of temperature, from 273.15 to 533.15 K, as well as liquid-vapour critical points [3-16].

Recently [16,17], we started a study on carbon dioxide + naphthenic hydrocarbons and we analysed the phase behaviour of the carbon dioxide + cyclopentane and carbon dioxide + cyclohexane systems with a three-parameters equation of state, RKPR [18], and with Peng-Robinson (PR) [19], coupled classical van der Waals quadratic mixing rules, using a special objective function (O.F.) [20]. The results were good, but they rely on the quality of the experimental data, as they are used in the objective function [16]. Therefore, in this work we compare the modelling results for the carbon dioxide + cyclohexane system by different cubic equations of state, namely the **G**eneral **E**quation **o**f **S**tate (GEOS) [21-24] and Soave-Redlich-Kwong (SRK) [25], coupled with classical van der Waals and excess Gibbs free energy mixing rules [26-28] using a different approach.

Although there is no experimental evidence regarding the type of phase behaviour, except liquid-vapour critical points, the carbon dioxide + cyclohexane system can be attributed to type I or type II [29, 30]. Both type I and II phase behaviour are very similar, being characterized by a continuous liquid vapour critical curve stretching between the critical points of the pure components. The difference is the appearance of a second liquid-liquid critical curve, intersecting in an upper critical endpoint (UCEP) with a three phases liquid-liquid-vapour equilibrium curve extending to lower temperatures, in the case of the type II phase behaviour [29,30]. Thus, following the same modelling procedure as in our previous papers [17,31], unique sets of interaction parameters, representing well the critical pressure maximum (CPM) and avoiding a false upper critical end point (UCEP) at high temperatures, were used to model the phase behaviour of the carbon dioxide + cyclohexane system. The calculations results were compared to all available literature VLE data. The agreement between the models result and the experimental data is reasonably good.

Modelling

The modelling of phase behaviour of this system was made with the GEOS [21-24] and SRK [25] equations of state (EoS) coupled with classical van der Waals mixing rules (2PCMR) and with Huron-Vidal (HV)–residual UNIQUAC mixing rules [26-28].

The GEOS [21] equation of state is:

$$P = \frac{RT}{V-b} - \frac{a(T)}{(V-d)^2 + c} \tag{1}$$

with the classical van der Waals mixing rules:

$$a = \sum_i \sum_j x_i x_j a_{ij} \qquad b = \sum_i \sum_j x_i x_j b_{ij} \tag{2}$$

$$c = \sum_i \sum_j x_i x_j c_{ij} \qquad d = \sum_i x_i d_i \tag{3}$$

$$a_{ij} = (a_i a_j)^{1/2}(1-k_{ij}) \qquad b_{ij} = \frac{b_i + b_j}{2}(1-l_{ij})$$

$$c_{ij} = \pm(c_i c_j)^{1/2} \tag{4}$$

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with + for c_i, $c_j > 0$ and – for c_i, $c_j < 0$. Generally, negative values are common for the c parameter of pure components.

The four parameters *a, b, c, d* for a pure component are expressed by:

$$a = \frac{R^2T_c^2}{P_c}\beta(T_r)\Omega_a \qquad b = \frac{RT_c}{P_c}\Omega_b \qquad (5)$$

$$c = \frac{R^2T_c^2}{P_c^2}\Omega_c \qquad d = \frac{RT_c}{P_c}\Omega_d \qquad (6)$$

Setting four critical conditions, with α_c as the Riedel's criterion:

$$P_r = 1 \qquad \left(\frac{\partial P_r}{\partial V_r}\right)_{T_r} = 0$$

$$\left(\frac{\partial^2 P_r}{\partial V_r^2}\right)_{T_r} = 0 \qquad \alpha_c = \left(\frac{\partial P_r}{\partial T_r}\right)_{V_r} \qquad (7)$$

at $T_r = 1$ and , the expressions of the parameters Ω_a, Ω_b, Ω_c, Ω_d are obtained:

$$\Omega_a = (1-B)^3 \qquad \Omega_b = Z_c - B$$

$$\Omega_c = (1-B)^2(B-0.25) \qquad (8)$$

$$\Omega_d = Z_c - \frac{(1-B)}{2} \qquad B = \frac{1+m}{\alpha_c + m} \qquad (9)$$

where , T_r, V_r are the reduced variables and Z_c is the critical compressibility factor.

The temperature function used is:

$$a(T) = a\cdot\beta(T_r);\ \beta(T_r) = T_r^{-m} \qquad (10)$$

The GEOS parameters *m* and α_c were estimated by constraining the EoS to reproduce the experimental vapour pressure and liquid volume on the saturation curve between the triple point and the critical point [21].

The SRK [25] EoS is:

$$P = \frac{RT}{V-b} - \frac{a(T)}{V\cdot(V+b)} \qquad (11)$$

As pointed out previously [32], the relations (8) and (9) are general forms for all the cubic equations of state with two, three, and four parameters.

The parameters of the SRK EoS can be obtained from the eqs. (8) and (9) by setting [21-24] the following restrictions: $\Omega_c = -(\Omega_b / 2)^2$ and $\Omega_d = -\Omega_b / 2$. It follows

$$\Omega_c = (1-B)^2(B-0.25) = -\frac{(Z_c - B)^2}{4} \qquad (12)$$

$$\Omega_d = Z_c - 0.5(1-B) = -\frac{(Z_c - B)}{2} \qquad (13)$$

It results

$$Z_c(SRK) = 1/3 \qquad (14)$$

and the relation for *B* (SRK)

$$B = 0.25 - \frac{1}{36}\left(\frac{1-3B}{1-B}\right)^2 \qquad (15)$$

Solving iteratively this equation gives B(SRK)=0.2467, and correspondingly Ω_a(SRK)=$(1\text{-}B)^3$ =0.42748 and Ω_b(SRK)=Z_c - B = 0.08664.

The Huron and Vidal mixing rules [26] for the SRK EoS are given by:

$$b = \sum_i x_i b_i \qquad (16)$$

$$\frac{a}{bRT} = \sum_i x_i\left(\frac{a_i}{b_iRT} - \frac{\ln\gamma_i(P\to\infty)}{\ln 2}\right) \qquad (17)$$

where $\gamma_i(P\to\infty)$ is the activity coefficient of the component in the mixture at infinite pressure.

The HV mixing rule is based on a reduced UNIQUAC model suitable for infinite pressure conditions [27,28]. The model is reduced to its residual part only:

$$\ln\gamma_i(P\to\infty) = q_i\left[1 - \ln\left(\sum_j \theta_j\tau_{ji}\right) - \sum_j \frac{\theta_j\tau_{ij}}{\sum_k \theta_k\tau_{kj}}\right] \qquad (18)$$

$$\theta_i = \frac{q_i x_i}{\sum_j q_j x_j} \qquad (19)$$

$$\tau_{ij} = \exp\left(-\frac{u_{ij}}{T}\right) \qquad (20)$$

The interaction parameters are considered temperature dependent [27,28]:

$$u_{ij} = u_{ij}^0 + u_{ij}^1\frac{1}{T} \qquad (21)$$

The calculations were made using the software packages PHEQ, developed in our laboratory [33], and GPEC [34,35]. The calculation of the critical curves implemented in PHEQ is based on the method proposed by Heidemann and Khalil [36], with numerical derivatives given by Stockfleth and Dohrn [37].

Results and discussions

Firstly all available experimental data (47 data sets, 348 experimental points) were correlated with GEOS/2PCMR, SRK/2PCMR, and SRK/HV-residual UNIQUAC models. The correlations by all models lead to small errors in bubble-point pressures, most of them being smaller than 1.0% (the average is less than 1.2 % for all sets), except for the data reported in ref. [4] which did not agree with other measurements too [16]. Although experimental data are available in a wide range of temperatures, many papers present only pressures-liquid phase compositions [16]. A high degree of scatter can be also observed. Figure 1 illustrates an example of correlations by four models (GEOS/2PCMR, PR/2PCMR, SRK/2PCMR, and SRK/HV-residual UNIQUAC) at two temperatures, 323.15 and 353.15 K. It can be seen that both bubble- and dew-points are very well correlated by all models, except for the critical region where GEOS/2PCMR leads to smaller critical pressures.

However, if the values of the optimised binary interaction parameters, k_{12} and l_{12} or u_{12} and u_{21} obtained at each temperature, are used to calculate the critical curve(s) of the carbon dioxide + cyclohexane binary system, the results are not always the expected ones. For instance, if the optimised binary interaction parameters at 323.15 and 353.15 K [16] are used to calculate the critical curve(s), the GEOS/2PCMR leads to type I of phase behaviour, while SRK/2PCMR leads to type II phase behaviour, as shown in figure 2.

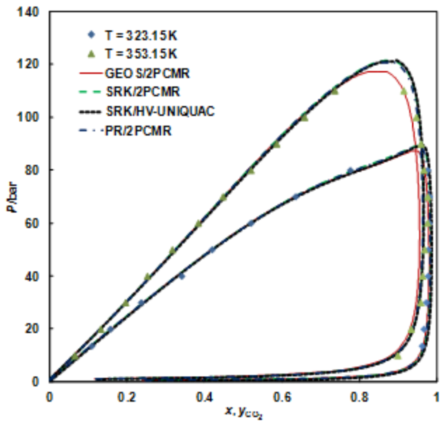

Fig. 1. Comparison of literature VLE data [16] and calculations by thermodynamic models for carbon dioxide + cyclohexane at 323.15 and 353.15 K temperatures: symbols, experimental data [16]; lines, correlations by GEOS, PR. and SRK EoSs.

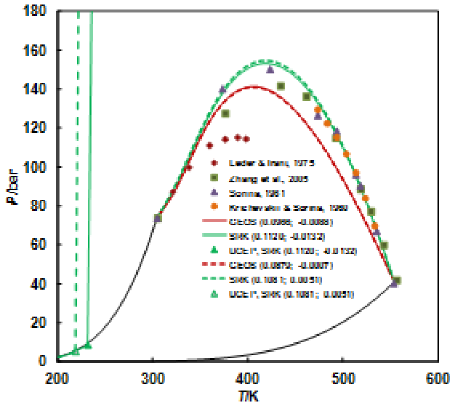

Fig. 2. *P-T* fluid phase diagram of carbon dioxide (1) + cyclohexane (2) system: symbols, literature data [15,38-40]; ■, vapour pressure curves of pure components; ——,- - , critical line calculated by GEOS/2PCMR; - - - -, ■■, critical lines calculated by SRK/2PCMR

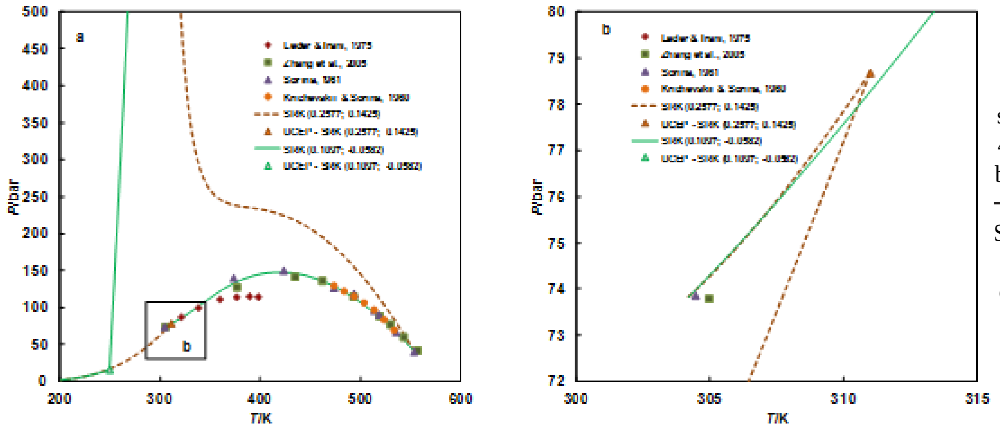

Fig. 3. a. *P -T* fluid phase diagram of carbon dioxide (1) + cyclohexane (2) system: symbols, literature data [15,38-40]; - -, critical lines calculated by SRK/2PCMR (0.2577; 0.1425); ——, critical lines calculated by SRK/2PCMR (0.1097; -0.0582); b. detail of the vapour-liquid critical curve, UCEP, and three phases liquid-liquid-vapour equilibrium line calculated by SRK/2PCMR (0.2577; 0.1425)

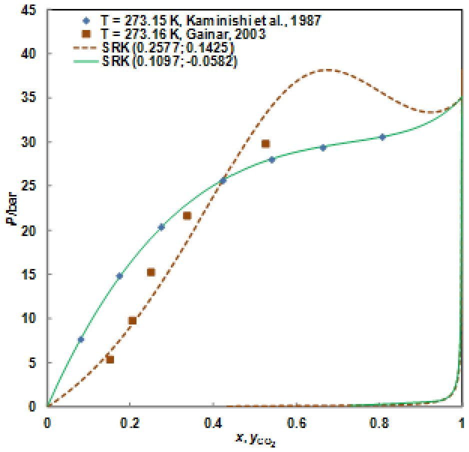

Fig. 4. Comparison of literature VLE data [3,4] and correlations by SRK/2PCMR EoS for carbon dioxide + cyclohexane at 273.15 K.

In figure 2 are plotted the calculated critical curves by the optimised binary interaction parameters at each temperature (323.15 and 353.15 K) for the two models, GEOS and SRK. It can be noticed that both the critical experimental data from different groups did not agree, as well as the calculations by the two models. The critical curves obtained with these parameters are still satisfactory, tough SRK leads to type II phase behaviour, as the calculated upper critical endpoint (UCEP) is located at low temperatures. In other cases, the values of optimised binary interaction parameters lead to a wrong type of phase behaviour, e.g. type III. This is the case when the optimised binary interaction parameters obtained by correlating the data reported by [4], as shown in figure 3a,b. It must be remarked that the false liquid-liquid splitting can be observed in the pressure-compositions diagram too, as a maximum and minimum of the calculated bubble-points curve. In figure 4 are plotted two experimental data sets at the same temperature measured by two different groups together with their correlations by SRK/2PCMR. It can be immediately spotted that the two experimental sets did not agree each other and the correlation of Gainar's [4] data leads to the aforementioned behaviour. In this particular case the average absolute deviation in bubble point pressure (AADP, %) is high (> 10%), but there are situations when the errors are small and the maximum – minimum in bubble-point pressures are not simply observed.

The average absolute deviations in bubble point pressure (AADP, %) and vapour phase compositions (AADY, %) for the carbon dioxide + cyclohexane system were calculated for the thermodynamic models used by the following equations:

$$AADP(\%) = \frac{1}{N_{exp}} \sum_{i=1}^{N_{exp}} \left| \frac{P_i^{exp} - P_i^{calc}}{P_i^{exp}} \right| \cdot 100 \qquad (22)$$

$$AADY(\%) = \frac{1}{N_{\exp}} \sum_{i=1}^{N_{\exp}} \left| Y_i^{\exp} - Y_i^{calc} \right| \cdot 100 \qquad (23)$$

The AADP, % as function of temperature from the correlations by GEOS/2PCMR, PR/2PCMR, SRK/2PCMR, and SRK/HV-UNIQUAC are plotted in figure 5 for all available data sets. It can be seen that with few exceptions (here are also included the data reported by ref. [4]), most of the errors are less than 2% for all models.

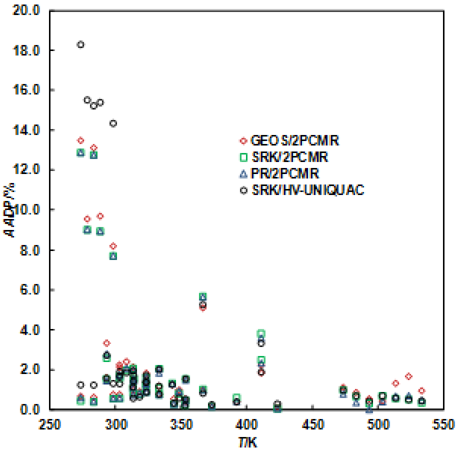

Fig. 5. Absolute average deviations in bubble point pressures by GEOS, PR, SRK/2PCMR, and SRK/HV-residual UNIQUAC correlations (regression of experimental data at each temperature from each data source separately) for the carbon dioxide + cyclohexane system

The reasons mentioned above demonstrate why just correlating the data, without taking into account the entire phase behaviour, is not satisfactory. Therefore, in this study we present a second approach to model in a semi predictive way the phase behaviour of the carbon dioxide + cyclohexane system. Similarly with our previous work on carbon dioxide + cyclopentane binary system [17], we used unique sets of binary interaction parameters for each model (GEOS/2PCMR, SRK/2PCMR, SRK/HV-UNIQUAC) representing well the critical pressure maximum (CPM) and avoiding an upper critical end point (UCEP), when possible, or at least its occurrence at high temperatures.

Thus, figure 6 presents the predicted critical curves calculated with the binary interaction parameters from table 1. For comparison reasons we also plotted the critical curves predicted by PR/2PCMR with the set of parameters from [16]. It can be seen that GEOS with the set of parameters k_{12} = 0.1205, l_{12} = 0.0050 predicts a type I phase behaviour diagram and its predicted critical pressure maximum is the closest to the experimental value, if the data reported by Sorina [40] and Krichevskii and Sorina [15] respectively are the reference ones. All other models (PR/2PCMR, SRK/2RPCMR, and SRK/HV-UNIQUAC) overestimate the CPM, but they represent better the maximum critical temperature. It can be also noticed that both SRK and PR predict type II phase behaviour, but the UCEP is located at low temperatures. The critical data [41], acentric factor [41], and cubic GEOS parameters of the pure components used in the calculations are given in table 2.

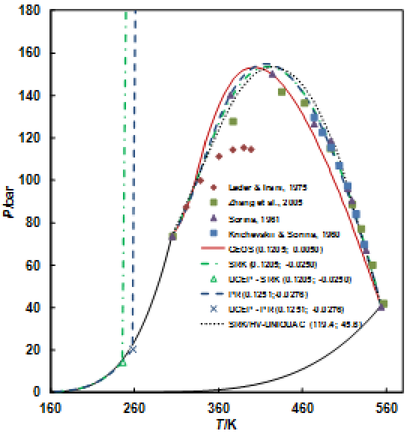

Fig. 6. *P* - *T* fluid phase diagram of carbon dioxide (1) + cyclohexane (2) system: symbols, literature data [15,38-40]; lines, predictions by GEOS, SRK, and PR EoSs

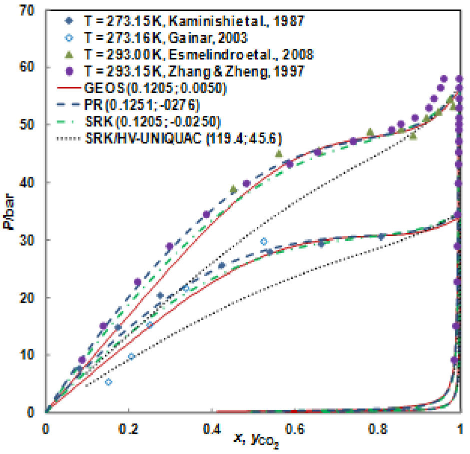

Fig. 7. Comparison of literature VLE data [3-6] and predictions by GEOS, PR, SRK/2PCMR, and SRK/HV-residual UNIQUAC models for carbon dioxide + cyclohexane

The parameters reported in table 1 were then used to calculate all available data. Several examples are shown in the next figures.

In figure 7 are plotted two sub-critical isotherms (273 and 293 K) together with the prediction results by all models, while in figure 8 are plotted several experimental data sets measured at 313 K and the predictions by the same models. It can be observed that the predictions are reasonable good for all models, except for SRK/HV-residual UNIQUAC, but we must recall that the set of parameters used to calculate the critical curve was constant, instead of a temperature function [42].

Table 1

BINARY INTERACTION PARAMETERS USED TO PREDICT THE PHASE BEHAVIOUR OF THE CARBON DIOXIDE + CYCLOHEXANE SYSTEM

Model (EoS/Mixing rule)	k_{12}	l_{12}	u_{12}/K	u_{21}/K
GEOS/2PCMR	0.1205	0.0050		
SRK/2PCMR	0.1205	-0.0250		
SRK/HV-UNIQUAC			119.4	45.6
PR/2PCMR*	0.1251	-0.0276		

*Parameters from ref. [16].

Table 2

CAS REGISTRY NUMBER, CRITICAL DATA, ACENTRIC FACTOR, AND GEOS PARAMETERS FOR CARBON DIOXIDE + CYCLOHEXANE

Component	CAS	P_c/bar	T_c/K	ω	α_c	m
CO_2	124-38-9	73.83	304.21	0.22362	7.0517	0.3146
C_6H_{12}	110-82-7	40.80	553.80	0.20805	7.0100	0.3228

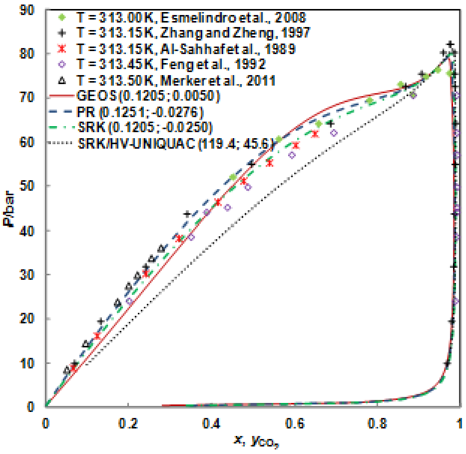

Fig. 8. Comparison of literature VLE data [5,6,8-10] and predictions by GEOS, PR, SRK/2PCMR, and SRK/HV-residual UNIQUAC models for carbon dioxide + cyclohexane.

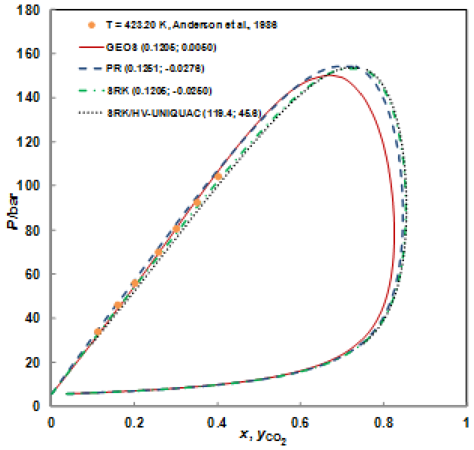

Fig. 9. Comparison of literature VLE data [12] and predictions by GEOS, PR, SRK/2PCMR, and SRK/HV-residual UNIQUAC models for carbon dioxide + cyclohexane.

In figure 9 are compared the predictions by the thermodynamic models and experimental data measured at 423.15 K. The best results are obtained with GEOS followed by PR and SRK. The absolute average deviations in pressure for all available experimental data and by all models are presented in figure10. Although there are some errors very high, as for instance for the data reported by ref. [4], in most cases the errors are reasonable taking into account the semi-predictive modelling procedure.

Conclusions

GEOS and SRK EoS coupled with classical van der Waals and G^E mixing rules were used to analyse the phase behaviour of the carbon dioxide + cyclohexane binary system. One set of interaction parameters for each thermodynamic model was used to predict the critical and sub-critical phase behaviour in the binary mixture carbon dioxide + cyclohexane in a wide range of temperatures. The predicted results were compared with the all available literature data for carbon dioxide + cyclohexane binary system. The phase behaviour was satisfactory reproduced, taking into account the semi predictive procedure used.

Acknowledgements: The work of Anca-Simona DEACONU was supported/funded by the Sectorial Operational Programme Human Resources Development 2007-2013 of the Ministry of European Funds through the Financial Agreement POSDRU/159/1.5/S/134398 (KNOWLEDGE) while the work of Aurel-Ionuþ CHIRILÃ was supported/funded by the Sectorial Operational Programme Human Resources Development 2007-2013 of the Ministry of European Funds through the Financial Agreement POSDRU/159/1.5/S/138963 (PERFORM). These authors would also like to thank the support given by the Applied Thermodynamics Research Laboratory.

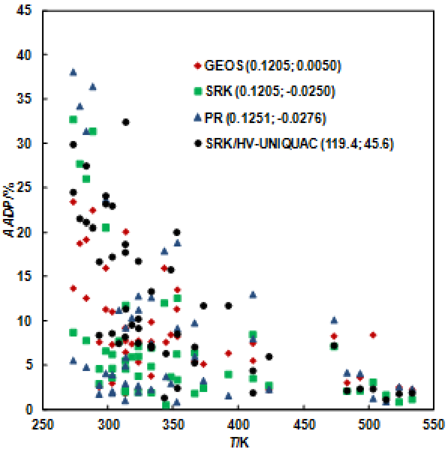

Fig. 10. Absolute average deviations in bubble point pressures by GEOS, PR, SRK/2PCMR, and SRK/HV-residual UNIQUAC predictions (with parameters from table 1 at each temperature from each data source separately) for the carbon dioxide + cyclohexane system

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Manuscript received: 19.10.2015