High-Pressure Vapour-Liquid Equilibria of Carbon Dioxide + 1-Pentanol System

Experimental Measurements and Modelling

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High-pressure vapour-liquid equilibria (VLE) were measured for the binary mixture carbon dioxide + pentanol at 313.15 and 353.15 K. The pressure range under investigation was between 0.59 and 11.22 MPa. The experimental method used in this work was a static-analytical method with liquid and vapour phase sampling. The new experimental results are discussed and compared with available literature data. Measured and literature VLE data for carbon dioxide + 1-pentanol system were correlated with the General Equations of State (GEOS), the Peng-Robinson (PR), and the Soave-Redlich-Kwong (SRK) equations of state (EoS) using classical van der Waals (two-parameters conventional mixing rule - 2PCMR), Huron Vidal at infinite dilution (HVID), MHV1, and MHV2 mixing rules.

Keywords: high-pressure, VLE, carbon dioxide, 1-pentanol, EoS

This work is a part of an extensive study of carbon dioxide + alcohols binary mixtures of interest in the design and implementation of many processes [1-3]. Carbon dioxide is the most used fluid in supercritical extraction. Application of supercritical carbon dioxide in extraction, reaction and separation is recognized as a green technology in order to replace the use of organic solvents. Very often a small amount of a polar modifier such as an alcohol is added to the supercritical carbon dioxide to enhance the extraction efficiency of polar compounds. Therefore, data for carbon dioxide + alcohol systems are of great importance in SFE (supercritical fluid extraction) and SFC (supercritical fluid chromatography), in the oil and natural gas industry, and in the cosmetic, pharmaceutical, surfactant, and food industries [4-6]. A literature search has identified few papers on the carbon dioxide + 1-pentanol binary system: Jennings et al. [7] reported results at 314.6, 325.9, and 337.4 K, Staby and Mollerup [8] at 283.2, 313.2, 343.2, and 373.2 K, and Silva-Oliver et al. [9] at 333.08, 343.69, 374.93, 414.23, and 426.86 K. Raeissi et al. [10] found that this system shows type IV fluid phase behavior, according to the classification of van Konynenburg and Scott [11]. In this work VLE data are reported at 313.15 and 353.15 K and pressures ranging from 0.59 to 11.22 MPa.

Measured and literature VLE data for carbon dioxide + 1-pentanol system were correlated with different equation of state (GEOS [12], PR [13], SRK [14]) using classical van der Waals (2PCMR) and G^E - EoS (HVID [15,16], MHV1 [17], MHV2 [18]) mixing rules.

Experimental part

Materials

Carbon dioxide (mass fraction purity >0.997) was provided by Linde Gaz Romania, and 1-pentanol (mass fraction purity >0.998) was a Fluka product. The chemicals were used as supplied.

Apparatus and Procedure

A detailed description of the experimental apparatus was presented in previous papers [19,20]. The apparatus used in this work is based on the static analytical method with liquid- and vapour-phase sampling. The procedure is the same as described elsewhere [19-21]. The entire internal loop of the apparatus including the equilibrium cell was rinsed several times with carbon dioxide. Then, the equilibrium cell was evacuated with a vacuum pump. The cell was charged with alcohol; then, it was slightly pressurized with carbon dioxide to the experimental pressure and was heated to the experimental temperature. To facilitate the approach to an equilibrium state, the mixture in the cell was stirred for a few hours. Then the stirrer was switched off, and about 1 h was allowed to pass until the coexisting phases were completely separated. Samples of the liquid and vapor phases were collected by depressurization and expansion into glass traps by using manually operated valves. The valves were operated in such a way as to keep the pressure in the visual cell almost constant. The total amounts of the organic substance in the glass trap were about 0.05 and 0.2 g for the vapour and liquid phases, respectively. The amount of carbon dioxide in each phase was obtained by expansion in a glass bottle of calibrated volume. In a typical experiment, the measured volumes of carbon dioxide were about 100 cm^3 from the vapour phase and 50 cm^3 from the liquid phase. The liquid samples of both phases were weighed with a precision balance (A&D Instruments Ltd, type HM-200, Tokyo, Japan) with an accuracy of 0.0001 g.

Results and Discussion

The equilibrium compositions for the carbon dioxide + 1-pentanol binary system were measured at 313.15 and 353.15 K, and the results are summarized in table 1. The values are averages of two or three measurements. For the VLE measurements, the uncertainty of the mole fraction is typically ± 0.001 and always < 0.003. Equilibrium measured data obtained at 313.15 K are used to confirm the accuracy of the measurement. This shows that the data are in good agreement with the existing data at the same temperature, as shown in figure 1.

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Table 1

MOLE FRACTION OF COMPONENT 1 IN THE LIQUID PHASE, X_1, AND MOLE FRACTION OF COMPONENT 1 IN THE VAPOR PHASE, Y_1 AT PRESSURE (P) AND TEMPERATURE (T) FOR THE BINARY SYSTEM CARBON DIOXIDE (1) + 1-PENTANOL (2)

P/MPa	X_1	Y_1	P/MPa	X_1	Y_1
T/K = 313.15 ± 0.1					
0.59	0.0400	0.9937	5.16	0.3544	0.9985
1.75	0.1227	0.9970	6.35	0.4507	0.9984
3.07	0.2060	0.9982	7.02	0.5175	0.9979
3.98	0.2661	0.9985			
T/K = 353.15 ± 0.1					
0.83	0.0351	0.9889	7.01	0.2989	0.9921
2.01	0.0834	0.9924	8.01	0.3466	0.9910
3.03	0.1292	0.9930	9.00	0.3968	0.9900
3.99	0.1744	0.9932	10.00	0.4477	0.9885
4.99	0.2152	0.9929	11.22	0.4950	0.9866
6.00	0.2513	0.9924			

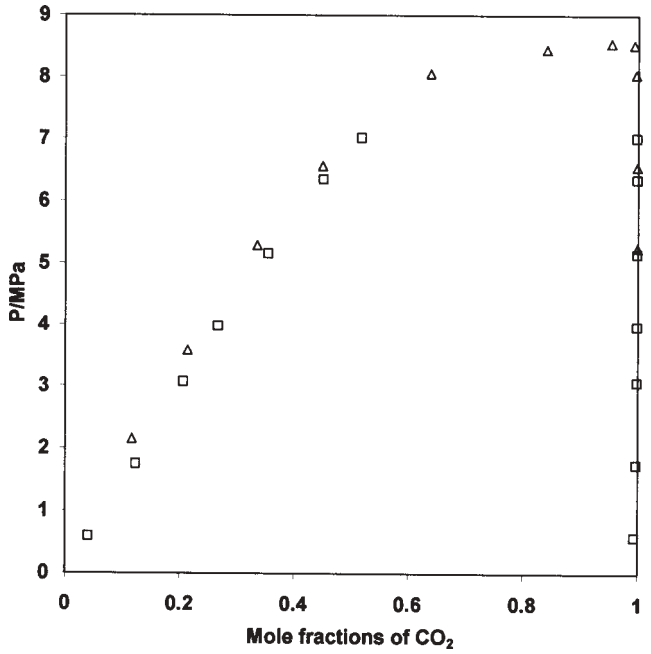


Fig. 1. Pressure-composition data for carbon dioxide + 1-pentanol at 313.15 K: (□) experimental, this work; (Δ) Staby and Mollerup [8]

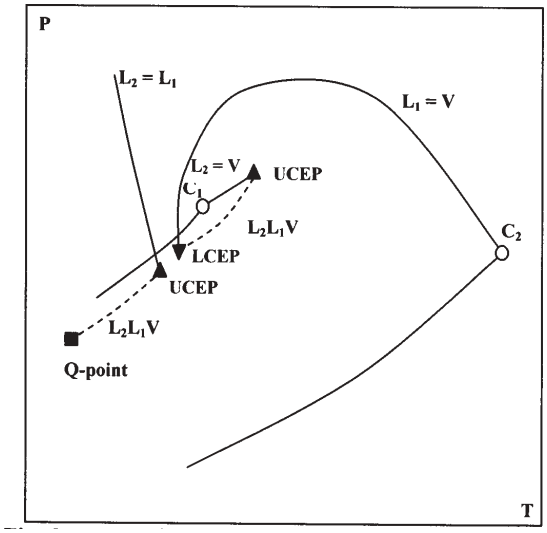


Fig. 2. P, T projection of type IV fluid phase behaviour

As Raeissi et al. [10] have shown, the carbon dioxide + 1-pentanol system is type IV fluid phase behaviour, according to the classification of van Konynenburg and Scott [11]. Type-IV fluid phase behaviour is characterized by having two separate three-phase loci (fig. 2). The low temperature branch shows an upper critical endpoint (UCEP), the high-temperature branch shows a lower critical endpoint (LCEP) and an UCEP. The critical endpoint (CEP) of the low temperature three-phase locus and the lower CEP of the three-phase locus situated at higher temperature both have the nature $L_2 = L_1V$, whereas the higher CEP of the high temperature three-phase equilibrium has the nature $L_2L_1 = V$. The UCEP of the lower temperature branch of the three-phase equilibrium LLV is located at 273.45 K [10] and the higher temperature branch of the three-phase equilibrium LLV is bounded by the two critical endpoints at 316.02 K, respectivelly 317.06 K [10]. The isotherms shown in this work (fig. 3 a, b) were measured at temperatures between the two three-phase equilibrium liquid-liquid-vapour (LLV) lines (313.15 K), respectivelly above the second three-phase equilibrium line (353.15 K).

Measured VLE data for carbon dioxide + 1-pentanol system were correlated with the GEOS, PR and SRK equations of state coupled with 2PCMR, HVID, MHV1, and MHV2 mixing rules.The correlations by EoS were compared with the new experimental data and the calculations predicted a false liquid-liquid splitting at 313.15 K. By restricting the interaction parameters in the optimization routine to avoid the false liquid-liquid splitting, in order to agree with the experimental observed behavior, the correlations are satisfactory only for SRK/HVID model (fig. 4), all other models lead to higher average absolute deviations in bubble point pressure (AADP, %), as can be seen in table 2. The AADP is calculated by the equation:

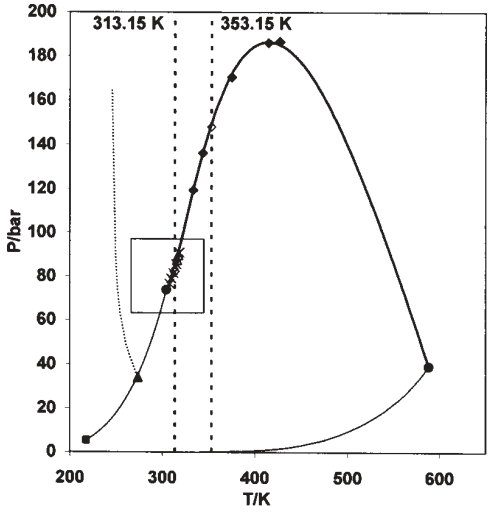
$$AADP(\%) = \frac{1}{N_{exp}} \sum_{i=1}^{N_{exp}} \left| \frac{P_i^{exp} - P_i^{calc}}{P_i^{exp}} \right| \cdot 100 \qquad (1)$$

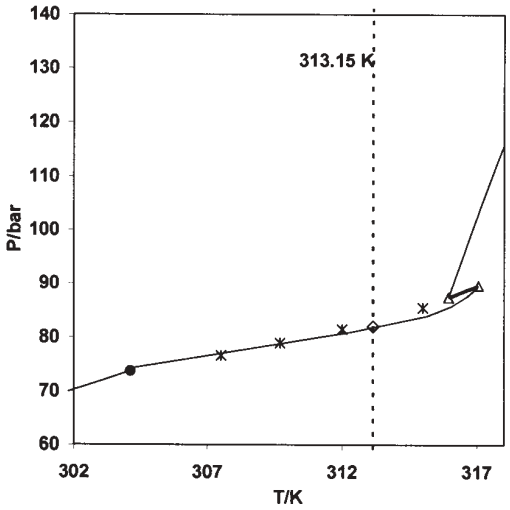
The correlations at 353.15 K are in good agreement with the experimental data (table 2) for all models. The optimized parameter values and the modified parameters are presented in table 3.

All available literature data (91 experimental points) were also correlated with the GEOS/2PCMR, PR/2PCMR, SRK/2PCMR, SRK/HVID, SRK/MHV1, and SRK/MHV2 and the calculations model lead, with few exception, to reasonable AADP (fig. 5) and average absolute deviations in vapor phase compositions (AADY, %). The AADY is calculated by the equation:

$$AADY(\%) = \frac{1}{N_{exp}} \sum_{i=1}^{N_{exp}} \left| Y_i^{exp} - Y_i^{calc} \right| \cdot 100 \qquad (2)$$

a





b

Fig. 3. P, T projection of carbon dioxide + 1-pentanol system: (●) critical points of pure components; (▲) UCEP of low temperature branch of LLV line [10]; (Δ) LCEP and UCEP of high-temperature branch of LLV line [10]; (■) quadruple point [23]; (-) vapor pressure curves of pure components; (.....) liquid-liquid critical line; (→) critical line $L_1 = V$(calculated with GEOS/2PCMR), and $L_2 = V$; (⇀) LLV line []; (- -) temperatures of this study, 313.15 and 353.15 K; (◇) critical points estimated from SRK/HVID calculations; (◆), experimental critical points [9]; (*), experimental critical points]241

T/K / EoS/Mixing rules	313.15		353.15	
	AADP (%)	AADY (%)	AADP (%)	AADY (%)
GEOS/2PCMR	2.0	0.1	1.4	0.1
GEOS/2PCMR*	7.9	0.1		
PR/2PCMR	1.8	0.1	1.3	0.1
PR/2PCMR*	12.8	0.1		
SRK/2PCMR	1.8	0.1	1.3	0.1
SRK/2PCMR*	8.6	0.1		
SRK/HVID	1.3	0.1	1.2	0.1
SRK/HVID*	3.5	0.1		
SRK/MHV1	1.3	0.1	1.2	0.1
SRK/MHV1*	10.7	0.1		
SRK/MHV2	1.2	0.1	1.2	0.1
SRK/MHV2*	10.6	0.1		

*Avoiding LL splitting

Table 2
AVERAGE ABSOLUTE DEVIATIONS IN BUBBLE POINT PRESSURE (AADP, %) AND AVERAGE ABSOLUTE DEVIATIONS IN VAPOR PHASE COMPOSITIONS (AADY, %) FOR THE CARBON DIOXIDE + 1-PENTANOL SYSTEM AT 313.15 AND 353.15 K

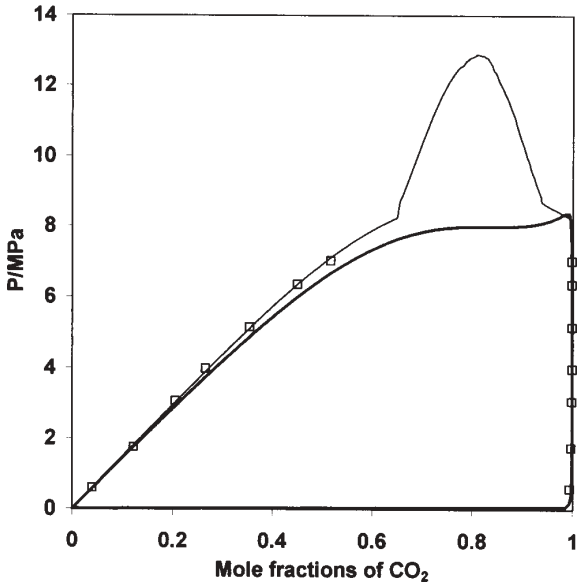


Fig. 4. Pressure-composition data for carbon dioxide (1) + 1-pentanol (2), comparing the experimental results at 313.15 K with the SRK/HVID model: (□), experimental data; (—), calculated with the SRK/HVID model, predicting false liquid-liquid splitting; (—),calculated with the SRK/HVID model with modified interaction parameters in order to avoid the false liquid-liquid splitting

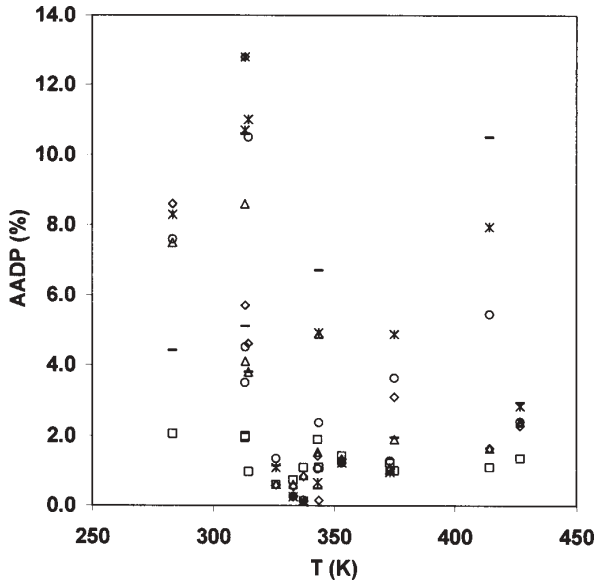


Fig. 5. Average absolute deviations in bubble point pressure (AADP, %) at temperatures from 283.2 to 426.86 K: (□), GEOS/2PCMR; (◇), PR/2PCMR; (Δ), SRK/2PCMR; (o), SRK/HVID; (*), SRK/MHV1; (-), SRK/MHV2.

Table 3

INTERACTION PARAMETERS FOR DIFFERENT EoS/MIXING RULES MODELS AT 313.15 AND 353.15 K FOR THE CARBON DIOXIDE + 1-PENTANOL BINARY SYSTEM

EoS/Mixing rules	T/K	NEXP	NCONV	k_{12}	l_{12}	u_{12}/K	u_{21}/K
GEOS/2PCMR	313.15	7	7	0.0815	-0.0099	-	-
GEOS/2PCMR*	313.15	7	7	0.0630	-0.0146	-	-
PR/2PCMR	313.15	7	7	0.1051	-0.0135	-	-
PR/2PCMR*	313.15	7	7	0.0820	-0.1450	-	-
SRK/2PCMR	313.15	7	7	0.1057	-0.0127	-	-
SRK/2PCMR*	313.15	7	7	0.0640	-0.0385	-	-
SRK/HVID	313.15	7	7	-	-	108.1	114.9
SRK/HVID*	313.15	7	7	-	-	61.1	144.9
SRK/MHV1	313.15	7	7	-	-	47.0	95.6
SRK/MHV1*	313.15	7	7	-	-	47.0	66.6
SRK/MHV2	313.15	7	7	-	-	-62.3	395.9
SRK/MHV2*	313.15	7	7	-	-	-63.3	355.9
GEOS/2PCMR	353.15	11	11	0.0734	-0.0212	-	-
PR/2PCMR	353.15	11	11	0.0902	-0.0335	-	-
SRK/2PCMR	353.15	11	11	0.0948	-0.0316	-	-
SRK/HVID	353.15	11	11	-	-	-10.9	233.5
SRK/MHV1	353.15	11	11	-	-	-83.9	243.6
SRK/MHV2	353.15	11	11	-	-	-134.3	450.1

*Avoiding LL splitting

Table 4

AVERAGE ABSOLUTE DEVIATIONS IN BUBBLE POINT PRESSURE (AADP, %), AVERAGE ABSOLUTE DEVIATIONS IN VAPOR PHASE COMPOSITIONS (AADY, %) AND INTERACTION PARAMETERS FOR GEOS/2PCMR RULES FROM LITERATURE DATA

T/K	NEXP	NCONV	k_{12}	l_{12}	AADP (%)	AADY (%)	Ref.
283.20	7	7	0.0831	-0.0126	2.1	-	8
283.20*	7	7	0.0659	-0.0126	10.2	-	8
313.20	7	6	0.0670	-0.0271	2.0	-	8
313.20*	7	7	0.0645	-0.0271	4.5	-	8
314.60	5	5	0.0694	-0.0245	1.0	0.1	7
314.60*	5	5	0.0594	-0.0245	7.5	0.0	7
325.90	8	8	0.0648	-0.0306	0.6	0.4	7
333.08	8	7	0.0610	-0.0300	0.7	0.6	9
337.40	9	9	0.0633	-0.0278	1.1	0.4	7
343.20	6	6	0.0640	-0.0305	1.9	-	8
343.69	5	4	0.0630	-0.0519	1.1	0.5	9
373.20	11	11	0.0587	-0.0322	1.2	1.2	8
374.93	6	5	0.0535	-0.0400	1.0	0.7	9
414.23	6	5	0.0508	-0.0345	1.1	1.2	9
426.86	13	12	0.0421	-0.0151	1.4	0.8	9

*Avoiding LL splitting

Table 5

AVERAGE ABSOLUTE DEVIATIONS IN BUBBLE POINT PRESSURE (AADP, %), AVERAGE ABSOLUTE DEVIATIONS IN VAPOR PHASE COMPOSITIONS (AADY, %) AND INTERACTION PARAMETERS FOR PR/2PCMR RULES FROM LITERATURE DATA

T/K	NEXP	NCONV	k_{12}	l_{12}	AADP (%)	AADY (%)	Ref.
283.20	7	7	0.1024	-0.0245	1.8	-	8
283.20*	7	7	0.0854	-0.0245	8.6	-	8
313.20	7	6	0.0867	-0.045	1.1	-	8
313.20*	7	7	0.0778	-0.045	5.7	-	8
314.60	5	5	0.0842	-0.0424	0.9	0.1	7
314.60*	5		0.0768	-0.0424	4.6	0.1	7
325.90	8	8	0.0776	-0.0507	0.6	0.4	7
333.08	8	7	0.0734	-0.0481	0.6	0.6	9
337.40	9	9	0.0767	-0.0449	0.8	0.3	7
343.20	6	6	0.0747	-0.0504	1.4	-	8
343.69	5	4	0.0689	-0.0702	0.2	0.4	9
373.20	11	11	0.0649	-0.0543	1.0	0.6	8
374.93	6	5	0.0561	-0.0399	3.1	1.7	9
414.23	6	5	0.0344	-0.0869	1.7	0.5	9
426.86	13	12	0.0286	-0.0366	2.3	1.0	9

*Avoiding LL splitting

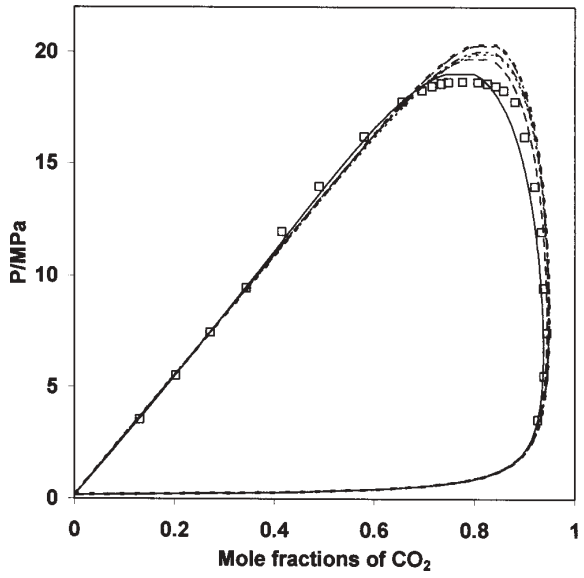


Fig. 6. Comparison of literature VLE data for carbon dioxide (1) + 1-pentanol (2) system at 426.86 K [9] with model results:(□) experimental data [9]; (—), calculated with GEOS / 2PCMR; (— —), calculated with PR / 2PCMR; (...), calculated with SRK /2PCMR; (— .—), calculated with SRK / HVID; (— .. —), calculated with SRK / MHV1; (- -), calculated with SRK/MHV2

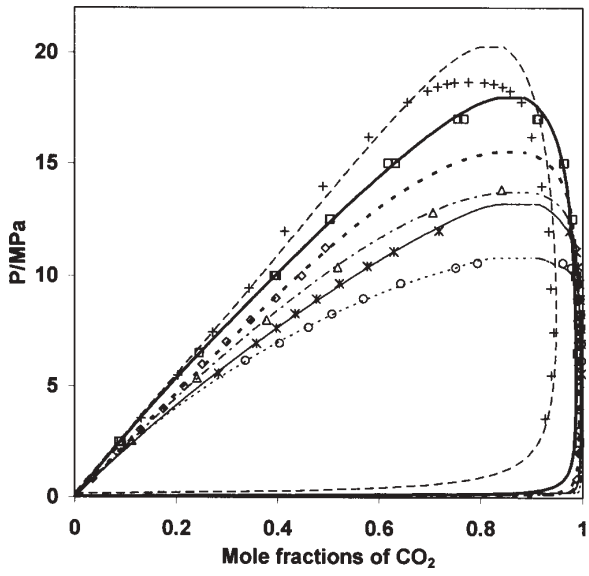


Fig. 7. Comparison of measured and literature VLE data for carbon dioxide (1) + 1-pentanol (2) system at different temperatures: (o), 325.9 K [7]; (·····), calculated with GEOS/2PCMR; (*), 337.4 K [7]; (—),calculated with PR/2PCMR; (Δ), 343.2 K [8]; (-·-·),calculated with SRK/MHV2; (◇), 353.15 K , this work; (---),calculated with SRK/ HVID; (□), 373.2 K [8]; (—),calculated with SRK/2PCMR; (+), 426.86 K [9]; (— —),calculated with SRK/MHV2

T/K	NEXP	NCONV	k_{12}	l_{12}	AADP (%)	AADY (%)	Ref.
283.20	7	7	0.1000	-0.0257	1.5	-	8
283.20*	7		0.0845	-0.0257	7.5	-	8
313.20	7	6	0.0835	-0.0476	1.3	-	8
313.20*	7		0.0784	-0.0476	4.1	-	8
314.60	5	5	0.0849	-0.0421	0.9	0.1	7
314.60*	5	8	0.0784	-0.0421	3.8	0.0	7
325.90	8	8	0.0792	-0.0499	0.6	0.2	7
333.08	8	7	0.075	-0.0478	0.6	0.9	9
337.40	9	9	0.0795	-0.0431	0.9	0.2	7
343.20	6	6	0.0777	-0.0487	1.5	-	8
343.69	5	4	0.0694	-0.1242	4.9	2.8	9
373.20	11	11	0.0698	-0.0526	1.1	0.5	8
374.93	6	5	0.0589	-0.0664	1.9	1.3	9
414.23	6	5	0.0386	-0.0959	1.7	0.5	9
426.86	13	12	0.0285	-0.0402	2.4	1.7	9

*Avoiding LL splitting

Table 6
AVERAGE ABSOLUTE DEVIATIONS IN BUBBLE POINT PRESSURE (AADP, %), AVERAGE ABSOLUTE DEVIATIONS IN VAPOR PHASE COMPOSITIONS (AADY, %) AND INTERACTION PARAMETERS FOR SRK/2PCMR RULES FROM LITERATURE DATA

T/K	NEXP	NCONV	u_{12}/K	u_{21}/K	AADP (%)	AADY (%)	Ref.
283.20	7	7	106.0	157.3	1.7	-	8
283.20*	7	7	71.5	157.3	7.6	-	8
313.20	7	5	26.3	237.4	2.5	-	8
313.20*	7	7	15.2	237.4	4.5	-	8
314.60	5	5	128.0	82.3	4.8	0.2	7
314.60*	5	5	95.2	84.3	10.5	0	7
325.90	8	6	18.2	215.1	1.4	0.1	7
333.08	8	7	19.2	190.5	0.3	0.8	9
337.40	9	9	4.5	213.9	0.2	0.2	7
343.20	6	6	-16.1	255.4	1.1	-	8
343.69	5	3	53.5	100.9	2.4	2.6	9
373.20	11	11	-81.0	317.2	1.3	0.6	8
374.93	6	4	69.3	49.6	3.6	2.6	9
414.23	6	5	15.7	72.6	5.4	3.9	9
426.86	13	12	-185.3	349.9	2.4	2.4	9

*Avoiding LL splitting

Table 7
AVERAGE ABSOLUTE DEVIATIONS IN BUBBLE POINT PRESSURE (AADP, %), AVERAGE ABSOLUTE DEVIATIONS IN VAPOR PHASE COMPOSITIONS (AADY, %) AND INTERACTION PARAMETERS FOR SRK/HVID RULES FROM LITERATURE DATA

T/K	NEXP	NCONV	u_{12}/K	u_{21}/K	AADP (%)	AADY (%)	Ref.
283.20	7	7	44.4	135.1	1.7	-	8
283.20*	7	7	15.4	135.1	8.3	-	8
313.20	7	6	29.9	109.9	8.5	-	8
313.20*	7	7	12.3	109.3	12.8	-	8
314.60	5	5	72.4	59.6	4.5	0.2	7
314.60*	5	5	45.3	59.5	11.0	0.0	7
325.90	8	8	-49.6	213.7	1.1	0.6	7
333.08	8	7	-58.0	205.8	0.3	0.7	9
337.40	9	9	-71.3	228.7	0.1	0.2	7
343.20	6	6	-94.3	275.1	0.7	-	8
343.69	5	4	-154.5	468.3	4.9	1.8	9
373.20	11	11	-162.5	357.2	1.0	0.6	8
374.93	6	5	54.0	-13.2	4.9	3.5	9
414.23	6	5	151.6	-152.2	7.9	3.6	9
426.86	13	12	-285.1	462.6	2.8	2.6	9

*Avoiding LL splitting

Table 8
AVERAGE ABSOLUTE DEVIATIONS IN BUBBLE POINT PRESSURE (AADP, %), AVERAGE ABSOLUTE DEVIATIONS IN VAPOR PHASE COMPOSITIONS (AADY, %) AND INTERACTION PARAMETERS FOR SRK/MHV1 RULES FROM LITERATURE DATA

T/K	NEXP	NCONV	u_{12}/K	u_{21}/K	AADP (%)	AADY (%)	Ref.
283.20	7	7	-50.0	543.4	4.4	-	8
313.20	7	6	-77.7	478.1	2.0	-	8
313.20*	7	7	-87.7	478.1	5.1	-	8
314.60	5	5	-81.3	452.3	0.2	0.1	8
314.60*	5	5	-89.3	452.3	3.8	0.0	7
325.90	8	8	-98.1	449.7	1.2	0.9	7
333.08	8	7	-109.7	430.2	0.3	0.7	7
337.40	9	9	-118.2	447.3	0.1	0.2	9
343.20	6	6	-129.5	476.8	0.5	-	7
343.69	5	4	0.1687	99.3	6.7	3.8	8
373.20	11	11	-185.1	513.1	0.9	0.6	9
374.93	6	5	-208.4	608.9	2.0	0.7	8
414.23	6	5	105.7	-115.6	10.5	6.0	9
426.86	13	12	-289.4	550.0	2.9	2.6	9

*Avoiding LL splitting

Table 9
AVERAGE ABSOLUTE DEVIATIONS IN BUBBLE POINT PRESSURE (AADP, %), AVERAGE ABSOLUTE DEVIATIONS IN VAPOR PHASE COMPOSITIONS (AADY, %) AND INTERACTION PARAMETERS FOR SRK/MHV2 RULES FROM LITERATURE DATA

The results obtained with these models are similar, as can be seen în figura 6.

False liquid-liquid splitting occurs also for literature data at temperatures near the CEPs. The interaction parameters were also restricted and the correlations are less accurate, but in agreement with the experimental behavior. The correlations were done for different temperatures, as presented in tables 4-9, between 283.2 K and 426.86 K. The AADP (%) for the 109 available experimental points are 2.8 for GEOS/2PCMR, 3.2 for PR/2PCMR, 2.9 for SRK/2PCMR, 3.2 for SRK/HVID, 4.8 for SRK/MHV1, and 3.6 for SRK/MHV2. The interaction parameters are also done in tables 4-9. Figure 7 presents the comparison of some literature data and our data at 353.15 K with the models calculated results.

Conclusions

New VLE experimental data were measured at 313.15 and 353.15 K for the carbon dioxide + 1-pentanol system, with high-pressure static apparatus. The obtained experimental data and all available literature data were correlated by different EoS coupled with classical van der Waals and G^E-EoS mixing rules.

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