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PVTx Measurements of the Carbon Dioxide + 2,3,3,3-Tetrafluoroprop-1-ene Binary System

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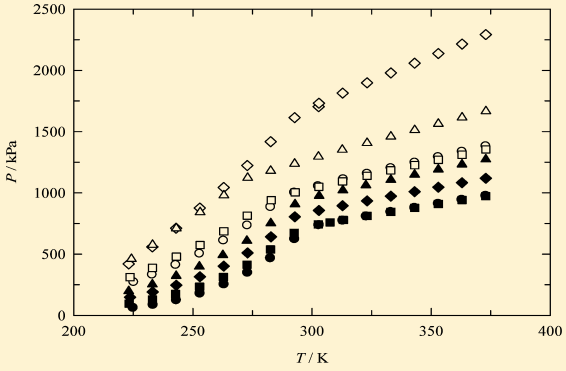
ABSTRACT: In this work, the *PVTx* properties of carbon dioxide + 2,3,3,3-tetrafluoroprop-1-ene were measured by an isochoric apparatus. The experiment covered both two-phase and superheated vapor regions and have been performed a total of eight isochores within a temperature range that spanned from (223 to 373) K and within a pressure range from (62 to 2292) kPa. Our experimental results were correlated with the Carnahan−Starling−DeSantis equation of state (CSD EoS) and predicted with REFPROP 9.0. Coefficients for the CSD EoS were derived from the experimental data. Results in the two-phase region showed a good agreement with the CSD equation and a strong disagreement with the REFPROP 9.0. Data in the superheated vapor region were also predicted by extrapolating coefficients for the CSD EoS at higher temperatures. The agreement with CSD EoS for the superheated vapor region experimental data was also good. The vapor−liquid equilibrium (VLE) behavior of the studied binary system from the experimental data was also derived.



INTRODUCTION

For a lower impact on the environment, the European Union decided to ban refrigerants with a global warming potential (GWP) over 150 in automobile air conditioning. This has resulted in a search to find alternatives to a commonly used fluid, R134a (GWP = 1430 for a 100 year time horizon).

R1234yf is a hydrofluoroolefin, a new refrigerant, recently proposed by the chemical industry. It is a non-ozone-depleting substance, having an atmospheric lifetime of 11 days. Its GWP is approximately 12 for a 20 year time horizon and 4 for a 100 year time horizon. Its thermophysical properties have been studied by our laboratory in previous papers.[1,2] One of the main drawbacks of R1234yf is its mildly flammability, having a small gap between lower and upper flammability limits.

Among natural refrigerants, we focused our interest on carbon dioxide,[3,4] particularly because it is nonflammable and it has a low toxicity. Although carbon dioxide has a GWP = 1 and an ozone depletion potential (ODP) that is zero, it has some restrictions due to its thermophysical properties. Additionally, technical issues regarding the use of carbon dioxide remain unresolved by the industry, and the very high system pressures require a total redesign of several system components.

To overcome the drawbacks of both carbon dioxide and R1234yf considered as pure fluids and searching for new perspective refrigerant mixtures for low temperature industrial applications, we focused our attention on a binary system containing both carbon dioxide and R1234yf.

In this paper, the *PVTx* properties of the carbon dioxide + R1234yf were measured by an isochoric apparatus over a wide temperature and pressure range.

EXPERIMENTAL SECTION

Chemicals. Data for each chemical sample are reported in Table 1. Carbon dioxide was supplied by Sol SpA. The sample of R1234yf was produced by the French group Arkema and donated by Centro Ricerche FIAT, Italy. It was then degassed to remove air and other noncondensable gases by immersing it in liquid nitrogen and evacuating. The sample was then brought

Table 1. Information for Each Chemical Sample

chemical name	source	initial mole fraction purity	purification method	final mole fraction purity	analysis method
carbon dioxide	Sol SpA	0.9999			
R1234yf[a]	Archema	0.9987	freezing, evacuating, and thawing	0.9995	GC[b]

[a]2,3,3,3-Tetrafluoroprop-1-ene. [b]Gas chromatography.

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to room temperature and was again subjected to the freezing, evacuating, and thawing process. This procedure was repeated several times. The purity of carbon dioxide and R1234yf was checked by gas chromatography using a thermal conductivity detector and was found to be better than 99.99 % and 99.95 % on a molar basis, respectively, basing all estimations on an area response.

Apparatus. The experimental setup is illustrated in Figure 1. The main modification to the original apparatus concerned the twin

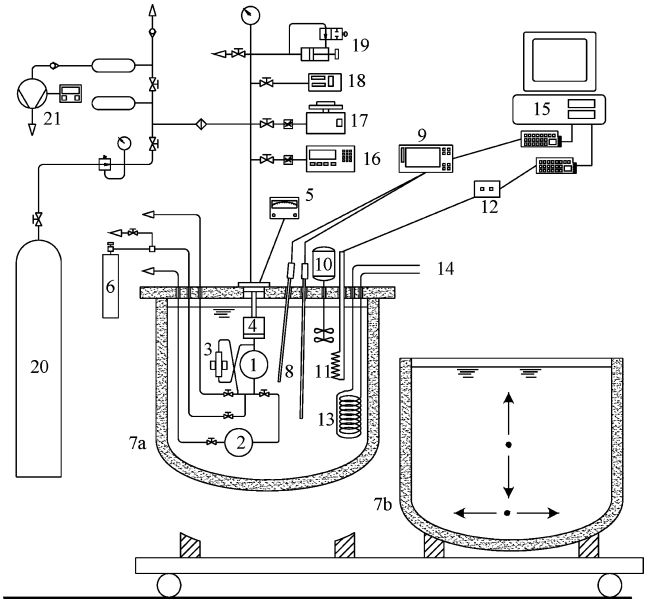

Figure 1. Schematic view of the experimental setup.

thermostatic baths (7) filled with different silicone oils (Baysilone M10 and Baysilone M100, Bayer). After charging with the sample mixture, the setup can be operated over two temperature ranges, from (210 to 290) K and from (290 to 360) K, depending on which bath is used. The thermostatic baths are easy to move due to the new configuration of the system. The spherical cells and pressure transducer are immersed in one of the two thermostatic baths (7). An auxiliary thermostat (14) is used to reach the below-ambient temperatures. The pressure and temperature data acquisition systems remained the same as in the previous apparatus[5] and will be only briefly described here.

An AISI 304 stainless steel spherical cell (1) containing the refrigerant sample is connected to a differential diaphragm pressure transducer (4) coupled to an electronic null indicator (5). In the new setup the transducer and sphere were placed vertically, and a magnetic pump (3) for mixing the sample was connected to the sphere. A second spherical cell (2) was also connected and used for volume calibration. Because of the complex volume of the isochoric cell (1), its total volume (including the piping, the pressure transducer cavity, and the magnetic pump volumes) was calibrated according to the classic Burnett calibration procedure, adopting helium as the reference fluid. After estimating the apparatus constant $N = (V_1 + V_2)/V_1$, the auxiliary cell volume, V_2, was measured by filling it with distilled water. Finally, the volume of the measurement cell, V_1, was found to be 273.5 ± 0.3 cm^3 at room temperature, based on several expansions from cell (1) to cell (2).

A PID device was used to control the temperature, which was measured using a calibrated resistance thermometer; the total uncertainty of the temperature measurements was ± 0.02 K. The uncertainty in the pressure measurements stems from the uncertainty of the transducer and null indicator system and the pressure gauges. The uncertainty of the digital pressure indicator (Ruska, model 7000) is ± 0.003 % of its full scale (6000 kPa). Temperature fluctuations due to bath instability can also affect the total uncertainty in the pressure measurement, which was nonetheless found to be less than ± 1 kPa.

Experimental Procedure. Mixtures were prepared using the gravimetric method. First of all, the pure samples were charged in different bottles, degassed to remove noncondensable gases and air, and then weighed with an analytical balance (uncertainty ± 0.3 mg). After evacuating the cell, the bottles were then emptied into the cell immersed in the bath. The mixtures were charged in the vapor phase. The bottles were weighed again, and the mass of the charge was calculated from the difference between the two masses. The dispersion of the mass inside the duct was estimated to be between (0.01 and 0.06) g, depending on the charging temperature, pressure, and molar mass of the fluid, and finally subtracted from the total mass of the sample. The uncertainty in the mass was estimated to be lower than ± 0.9 mg.

The uncertainty for the mass yields an uncertainty for the molar fraction that can be estimated to be constantly lower than 0.001 in mole fraction. Taking into account the uncertainty in the charged mass and in the isochoric cell volume, we estimated the uncertainty in molar volume to be always lower than ± $5 \cdot 10^{-3}$ $dm^3 \cdot mol^{-1}$. In addition, it is worth it to mention that all changes of molar volumes along temperature depend only on changes in the isochoric cell volumes, also considering the correction for thermal expansion and pressure distortion, as reported elsewhere.[6]

After reaching the experimental temperature, the mixing pump was activated for about 15 min, and, next, the mixture was allowed to stabilize for about 20 min before the data recording. After having charged each mixture composition, the temperature was increased step by step.

RESULTS AND DISCUSSION

The temperature and pressure ranges are shown in Table 2, along with the mixture's composition and the number of moles charged. The *P*,*T* isochoric sequence is also shown in Figure 2.

Table 2. Measurements at Bulk Compositions z_1 and Masses m_1 and m_2 for the CO_2 (1) + R1234yf (2) System over the Temperature Range ΔT and Pressure Range ΔP

		ΔT	ΔP	n	m_1	m_2
series	z_1	K	kPa	mol	g	g
1	0.051	225−373	62−976	0.0954	0.215	10.322
2	0.160	223−373	96−973	0.0940	0.660	9.005
3	0.266	224−373	148−1119	0.1075	1.256	8.999
4	0.365	223−373	198−1273	0.1225	1.968	8.872
5	0.480	225−373	271−1381	0.1323	2.796	7.838
6	0.574	224−373	312−1354	0.1282	3.236	6.232
7	0.662	223−373	420−2292	0.2250	6.559	8.664
8	0.754	224−373	460−1667	0.1580	5.245	4.433

On the basis of the analysis of the slope of each *T*,*P* sequence, the experimental points were each attributed either to the superheated or to the two-phase region. Table 3 shows the experimental data within the vapor−liquid equilibrium (VLE) boundary, while Table 4 contains the *PVTx* data.

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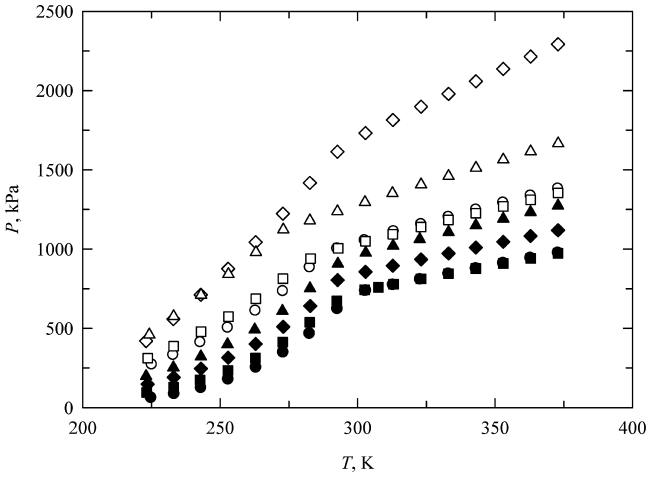

Figure 2. Experimental $P-T$ data: ●, z_1 = 0.051; ■, z_1 = 0.160; ◆, z_1 = 0.266; ▲, z_1 = 0.365; ○, z_1 = 0.480; □, z_1 = 0.574; ◇, z_1 = 0.662; △, z_1 = 0.754.

Table 3. Experimental Molar Volumes V as a Function of Pressure P and Temperature T at Charged Composition z_1 in the Two-Phase Region for the CO_2 (1) + R1234yf (2) System

T	P	V	T	P	V
K	kPa	$dm^3 \cdot mol^{-1}$	K	kPa	$dm^3 \cdot mol^{-1}$
z_1 = 0.051			272.66	609.0	2.258
224.91	61.7	2.894	282.69	751.6	2.259
233.25	86.7	2.895	z_1 = 0.480		
243.07	124.7	2.896	225.14	271.4	2.063
252.97	179.0	2.897	232.92	331.8	2.063
263.06	253.3	2.899	242.84	412.5	2.064
272.93	348.1	2.900	252.82	504.1	2.065
282.56	466.4	2.901	262.86	610.6	2.066
292.70	623.0	2.902	272.84	734.4	2.067
z_1 = 0.160			282.62	884.2	2.068
223.28	95.7	2.938	z_1 = 0.574		
233.00	129.7	2.939	223.63	312.0	2.153
242.68	174.3	2.940	233.01	387.5	2.154
252.82	234.8	2.942	243.08	479.4	2.155
262.80	313.0	2.943	253.03	574.3	2.156
272.75	412.9	2.944	262.96	686.0	2.157
282.57	537.9	2.945	272.82	813.7	2.158
z_1 = 0.266			z_1 = 0.662		
223.62	147.7	2.569	223.07	420.2	1.227
233.09	190.8	2.570	232.97	559.0	1.227
243.03	246.0	2.571	242.95	711.4	1.228
252.95	315.1	2.572	252.90	876.2	1.228
262.95	401.7	2.573	262.94	1044.0	1.229
272.93	509.9	2.574	272.83	1223.1	1.229
282.75	641.4	2.575	282.64	1417.7	1.230
292.75	804.6	2.577	z_1 = 0.754		
z_1 = 0.365			224.20	460.3	1.747
223.08	197.6	2.253	233.06	576.0	1.747
232.97	252.9	2.254	243.04	710.6	1.748
242.99	322.1	2.255	253.04	842.3	1.749
252.69	399.3	2.256	263.01	980.1	1.750
262.58	492.3	2.257			

VLE Derivation. The method used to derive VLE data from the isochoric measurements using the Carnahan–Starling–DeSantis equation of state (CSD EoS) was described elsewhere.[7] The CSD EoS was adopted in the following form:

$$\frac{PV}{RT} = \frac{1 + Y + Y^2 - Y^3}{(1 - Y)^3} - \frac{a}{RT(V + b)} \quad (1)$$

where

$$Y = \frac{b}{4V} \quad (2)$$

The method involves deriving the VLE parameters for each data point in the two-phase region using the "flash method" with the CSD EoS. For this method to be applied to isochoric data, we also

Table 4. Experimental Molar Volumes V as a Function of Pressure P and Temperature T at Composition z_1 in the Superheated Vapor Region for the CO_2 (1) + R1234yf (2) System

T	P	V	T	P	V
K	kPa	$dm^3 \cdot mol^{-1}$	K	kPa	$dm^3 \cdot mol^{-1}$
z_1 = 0.051			302.61	1055.6	2.070
302.81	736.5	2.904	313.12	1111.4	2.071
303.05	738.0	2.904	323.11	1157.2	2.071
312.94	773.7	2.905	333.08	1202.8	2.072
322.75	808.1	2.906	343.10	1248.0	2.073
333.09	843.3	2.907	352.97	1292.5	2.074
343.06	877.3	2.909	362.96	1337.1	2.075
353.02	910.7	2.910	372.92	1381.3	2.076
362.92	943.3	2.911	z_1 = 0.574		
372.92	975.7	2.912	282.88[a]	940.5[a]	2.159[a]
z_1 = 0.160			292.91	1004.6	2.160
292.47[a]	673.8[a]	2.947[a]	302.86	1049.3	2.161
302.61	742.4	2.948	312.79	1094.3	2.162
307.54	758.8	2.949	323.10	1139.4	2.163
313.12	778.4	2.949	333.06	1183.4	2.164
323.09	811.8	2.951	343.04	1227.0	2.165
333.08	844.9	2.952	352.99	1269.8	2.166
343.05	877.5	2.953	362.94	1311.6	2.167
352.99	909.5	2.954	372.92	1354.3	2.167
362.96	941.6	2.956	z_1 = 0.662		
372.89	973.3	2.957	292.69[a]	1614.0[a]	1.231[a]
z_1 = 0.266			302.89	1732.2	1.231
302.88	856.1	2.578	312.84	1814.8	1.232
312.82	895.4	2.579	323.10	1898.7	1.232
323.10	934.8	2.580	333.08	1979.1	1.233
333.08	972.6	2.581	343.05	2058.6	1.233
343.04	1010.2	2.582	352.99	2136.6	1.234
353.00	1046.2	2.583	362.97	2214.8	1.234
362.92	1082.5	2.584	372.93	2291.9	1.235
372.89	1119.0	2.586	z_1 = 0.754		
z_1 = 0.365			272.98	1122.7	1.750
292.85[a]	906.8[a]	2.260[a]	282.63	1179.9	1.751
302.96	976.1	2.261	292.67	1237.7	1.752
312.92	1020.4	2.262	302.67	1296.0	1.753
322.60	1062.1	2.263	312.70	1352.4	1.753
333.07	1106.6	2.264	323.10	1407.6	1.754
343.03	1149.4	2.265	333.08	1460.2	1.755
352.99	1191.4	2.266	343.07	1512.6	1.756
362.92	1232.6	2.267	353.02	1564.1	1.756
z_1 = 0.480			362.96	1615.2	1.757
292.60	1003.3	2.069			

[a]Not considered in the regression.

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Table 5. Composition at Bubble Point x (1), Composition at Dew Point y (1), Bubble-Point Pressure (P_B), and Dew-Point Pressure (P_D) Derived with the CSD EoS[6] in the Two-Phase Region for the CO_2 (1) + R1234yf (2) System

		P_B	P_D			P_B	P_D
x (1)	y (1)	kPa	kPa	x (1)	y (1)	kPa	kPa
z_1 = 0.051				0.108	0.500	1331.7	483.5
0.024	0.324	82.4	43.6	0.096	0.430	1677.5	674.6
0.021	0.264	116.4	66.3	z_1 = 0.480			
0.018	0.205	169.6	104.2	0.301	0.880	404.1	78.5
0.016	0.159	240.9	157.8	0.269	0.846	534.5	115.9
0.014	0.122	335.5	232.7	0.233	0.794	740.4	183.1
0.012	0.095	453.7	330.0	0.201	0.732	997.6	278.8
0.011	0.075	597.2	452.4	0.175	0.664	1311.0	411.4
0.010	0.059	782.9	615.9	0.154	0.592	1680.8	588.4
z_1 = 0.160				0.137	0.523	2101.4	815.3
0.077	0.625	156.7	45.0	z_1 = 0.574			
0.066	0.540	225.2	73.7	0.377	0.910	442.0	87.2
0.057	0.457	313.4	115.1	0.328	0.876	622.5	139.9
0.049	0.377	430.7	176.5	0.281	0.828	868.7	222.2
0.042	0.308	574.7	259.4	0.241	0.770	1171.5	337.6
0.037	0.249	750.2	369.5	0.209	0.705	1538.0	496.0
0.033	0.202	958.2	510.6	0.183	0.637	1968.3	706.4
z_1 = 0.266				z_1 = 0.662			
0.142	0.760	235.2	52.2	0.550	0.949	486.0	104.7
0.123	0.693	331.3	84.2	0.507	0.930	701.6	172.6
0.106	0.615	459.7	133.1	0.459	0.903	980.4	272.7
0.091	0.536	619.8	202.1	0.410	0.869	1327.5	414.2
0.079	0.458	816.8	297.2	0.365	0.825	1754.0	610.9
0.070	0.387	1053.0	424.2	0.326	0.775	2253.5	871.2
0.062	0.324	1326.7	586.6	0.294	0.720	2829.8	1211.1
0.056	0.269	1650.6	798.0	z_1 = 0.754			
z_1 = 0.365				0.590	0.954	563.7	147.2
0.217	0.838	298.2	58.2	0.529	0.934	784.7	228.8
0.188	0.783	426.2	96.2	0.458	0.903	1101.3	360.2
0.162	0.718	591.5	152.6	0.393	0.862	1500.2	546.7
0.141	0.649	790.0	229.7	0.339	0.811	1987.7	802.6
0.123	0.575	1034.6	337.0	0.590	0.954	563.7	147.2

Table 6. Coefficients of the CSD EoS[7] for R1234yf

	a_0	a_1	a_2	b_0	b_1	b_2
carbon dioxide	1324.8179	$-3.8522\cdot10^{-3}$	$-8.8413\cdot10^{-7}$	0.0680	$-1.0770\cdot10^{-4}$	$-2.3529\cdot10^{-8}$
R1234yf	3939.9449	$-2.6782\cdot10^{-3}$	$-1.5475\cdot10^{-6}$	0.1717	$-2.1467\cdot10^{-4}$	$-5.0706\cdot10^{-8}$

need the volumetric properties of both phases, which were calculated from the CSD EoS. T, z_i, and n (number of moles charged) were kept constant during the correlation having the isochoric cell volume from the gravimetric calibration, the objective function:

$$Q = \sum_i \left(\frac{dP}{P}\right)^2 \tag{3}$$

was minimized by tuning the binary interaction parameter value, K_{12}. The average of the value was found to be $K_{12} = -0.010$, with a statistical uncertainty of ± 0.005. The correlation gives also the parameters of VLE (pressure and composition of both phases) which were considered, obviously, as dependent variables. The values of composition at bubble point, composition at dew point, bubble-point pressures, and dew-point pressures derived with the CSD EoS[6] are reported in Table 5.

For pure compounds, the temperature dependence of parameters a and b in the CSD EoS[6] is described by the following expressions:

$$a = a_0\cdot\exp(a_1T + a_2T^2) \tag{4}$$

$$b = b_0 + b_1T + b_2T^2 \tag{5}$$

The a_i and b_i coefficients for R1234yf and carbon dioxide are reported in Table 6. Figure 3 shows the scatter diagram of the relative pressure deviations. The pressure deviations were found to be within ± 1 % for all series, except few points at lower temperatures measured for series 3. A very different trend of results was obtained comparing the experimental data together with the REFPROP 9.0[8] prediction, which calculations are based on an equation of state explicit in reduced Helmholtz energy.[9]

Here, deviations up to 40 % and a generally strong disagreement was evident with experimental data, increasing with the lowering of temperatures, as shown in Figure 4. Since both pure fluids' behavior was well-predicted by the REFPROP 9.0, the revealed discrepancy was probably due to the predictive method

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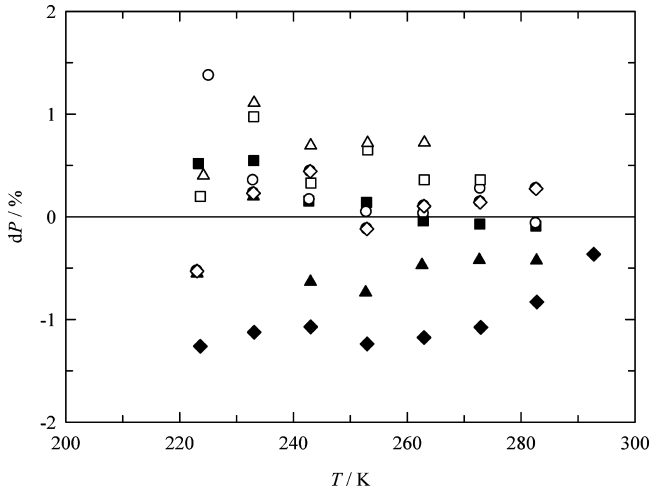

Figure 3. Pressure deviations between experimental values and those calculated with the CSD EoS[7] for the CO_2 (1) + R1234yf (2) system in the two-phase region. Symbols are denoted as in Figure 2.

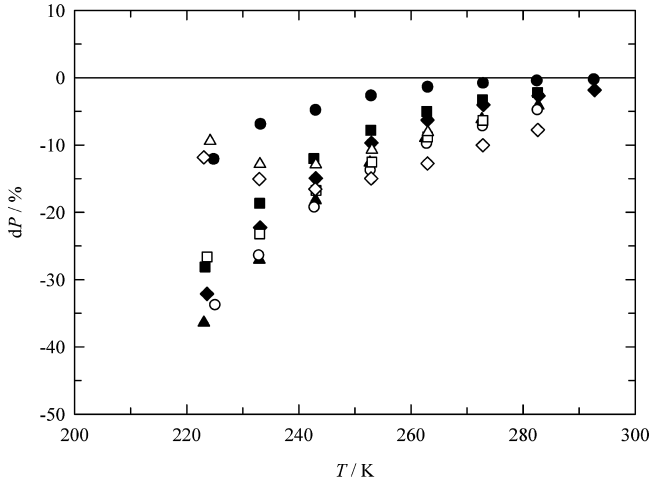

Figure 4. Pressure deviations between experimental values and those calculated with the REFPROP 9.0[8] for the CO_2 (1) + R1234yf (2) system in the two-phase region. Symbols are denoted as in Figure 2.

applied in the estimation of binary interaction parameters, since no experimental mixture data were available for this system.

The VLE behavior of the system was also calculated at three fixed temperatures, (233.15, 253.15, and 273.15) K, with the CSD EoS[7] adopting the found binary interaction parameter $K_{12} = -0.010$. The system showed small deviations from Raoult's law for all of the temperatures, as shown in Figure 5.

PVTx. Data on the superheated vapor region for the binary systems considered were also compared with the CSD EoS[6] and REFPROP 9.0[8] prediction. In this case, the averaged value of $K_{12} = -0.010$ obtained with the flash method in the two-phase region data was used for the CSD EOS calculations. Not considering the points in the proximity of the change of phase that showed much higher deviations (these points were denoted in Table 4 with an asterisk), an AAD (dP%) = 0.24 % was obtained. Again, deviations within ± 1 % for all series were obtained with the CSD EoS, except few points at lower temperatures. Slightly higher deviations were achieved comparing the experimental findings with the REFPROP 9.0 prediction, obtaining an AAD (dP%) = 0.64 %. Results are summarized in Figures 6 and 7.

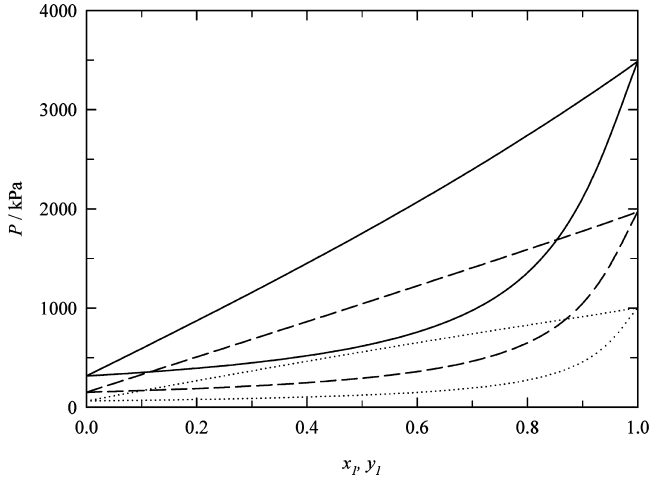

Figure 5. VLE representation from the correlation with the CSD EoS[6] for the CO_2 (1) + R1234yf (2) system at three temperatures: dotted lines, T = 233.15 K; dashed lines, T = 253.15 K; solid lines, T = 273.15 K.

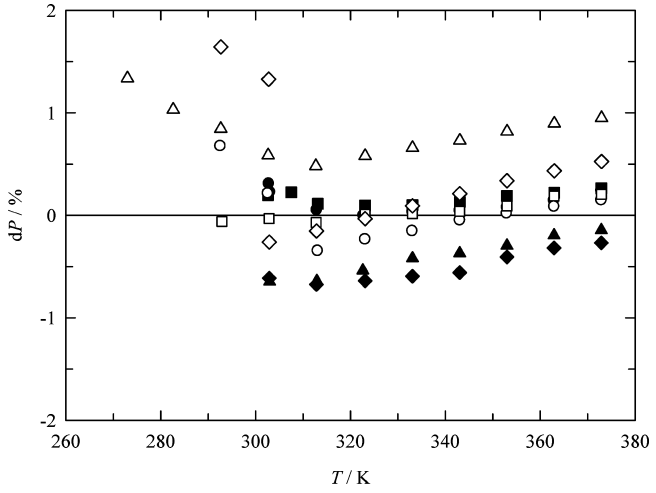

Figure 6. Pressure deviations between experimental values and those calculated with the CSD EoS[7] for the CO_2 (1) + R1234yf (2) system in the superheated vapor region. Symbols are denoted as in Figure 2.

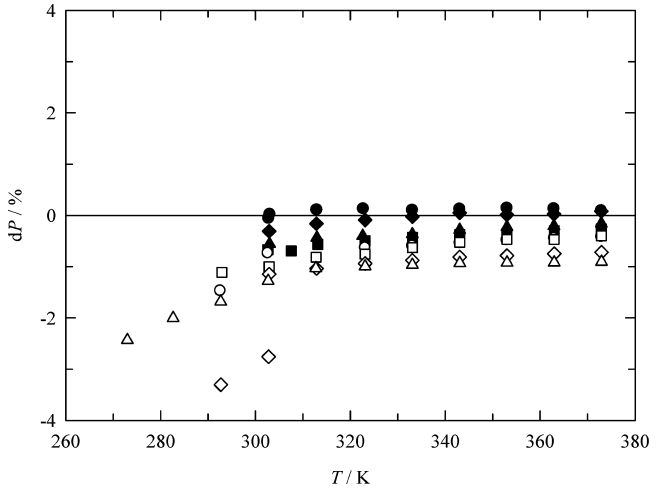

Figure 7. Pressure deviations between experimental values and those calculated with the REFPROP 9.0[8] for the CO_2 (1) + R1234yf (2) system in the superheated vapor region. Symbols are denoted as in Figure 2.

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CONCLUSIONS

An isochoric apparatus has been used to obtain *PVTx* measurements on CO_2 + R1234yf. The binary interaction parameters were derived from experimental data in the two-phase region, applying the flash method and the CSD EoS. The *PVTx* data at superheating region were compared by the CSD EoS and by REFPROP 9.0 prediction.

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