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Virial Coefficients from Burnett Measurements for the Carbon Dioxide + Fluoromethane System

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This work reports the experimental results for the carbon dioxide (CO_2) + fluoromethane (R41) system obtained using the Burnett method. The apparatus was calibrated using helium. For mixtures, the composition was found by gas chromatographic analysis of the sample collected during expansions. The gas chromatograph was calibrated precisely using samples prepared by the gravimetric method. From the *PVT* data collected, the second and third virial coefficients were derived for R41 covering the temperature range from 303 to 343 K. The second and third cross virial coefficients were derived from the *PVTx* measurements for the CO_2 + R41 system at T = (303, 313, 323, 333, and 343) K. The second virial coefficients for pure R41 show a good consistency with the literature. Due to lack of experimental data in the literature for the CO_2 + R41 system, our density measurements were compared with the REFPROP 6.01 prediction, the differences being within 0.6%, which would confirm the good predictive power of the model for this system and property.

Introduction

Carbon dioxide has received much attention as a refrigerant for industrial applications in both low and transcritical temperature ranges. It is neither flammable nor toxic, and it is inexpensive and widely available. One of the main disadvantages of this natural fluid is its relatively high melting temperature for the low temperatures and high pressures of transcritical applications, in which case the plant components need to be designed for much higher pressures (the maximum operating pressures are typically 150 bar). Fluoromethane is a hydrofluorocarbon used in the refrigeration industry as a constituent in several binary or ternary mixtures substituting difluorochloromethane (R22). The main drawback of this refrigerant fluid lies in its flammability and far from negligible GWP, despite a zero ODP. However, a combination of fluoromethane and CO_2 might make it possible to achieve the basic requirements for industrially applicable blends.

In a first attempt to study the properties of the CO_2 + fluoromethane (R41) system, the *PVTx* property was measured by the Burnett method and virial coefficients were derived from the experimental data collected. To our knowledge, no experimental *PVTx* data are reported in the literature for this specific binary system.

Experimental Section

Reagents. CO_2 and R41 were supplied by Sol SpA and Lancaster Inc., respectively; their purities were checked by gas chromatographic analysis, using a thermal conductivity detector. They was found to be 99.99% for the CO_2 and 99.9% for the R41 on an area-response basis.

Apparatus. A diagram of the apparatus is shown in Figure 1. It is the same as the one described elsewhere[1−3] and used with only minimal modifications already described.[4] It consists of two pressure vessels, the measurement chamber, V_A, and the expansion chamber, V_B, both spherical in shape, with a volume of approximately 70 and 35 cm^3, respectively, plus some auxiliary systems for filling and mixing the compounds in the Burnett vessels and for controlling and measuring the pressure and temperature. The vessels are made of Invar, given its excellent corrosion resistance and low thermal expansion coefficient. The four-

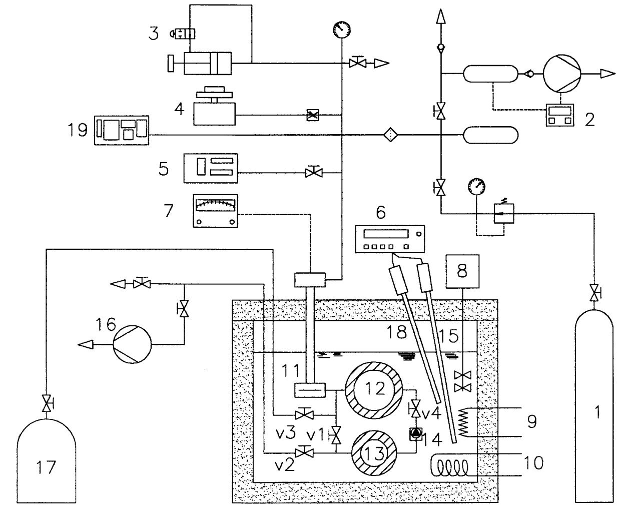

Figure 1. Schematic view of the experimental apparatus: 1, nitrogen reservoir; 2, vacuum pump (Vacuubrand, mod. RZ2); 3, precision pressure controller (Ruska, mod. 3981); 4, gas lubricated dead weight gauge (Ruska, mod. 2465); 5, vibration cylinder pressure gauge (Ruska, mod. 6220); 6, digital temperature indicator (Corradi, RP 7000); 7, electronic null indicator (Ruska, mod. 2416); 8, stirrer; 9, heater; 10, cooling coil connected with an auxiliary bath; 11, differential pressure transducer (Ruska, mod. 2413); 12, measurement chamber (V_A); 13, expansion chamber (V_B); 14, nagnetic recirculating pump; 15, Pt resistance thermometer (Tersid, Pt 100); 16, vacuum pump for V_B (Vacuubrand, mod. RZ2); 17, charging fluid reservoir; 18, Pt resistance thermometer (Hart Scientific, Pt 25); 19, digital pressure indicator (Ruska, mod. 7000); V_1, V_2, V_3, and V_4, constant volume valves.

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Table 1. Experimental Pressures Measured during Burnett Expansions of R41

P/kPa								
series 1	series 2	series 3	series 4	series 5	series 6	series 7	series 8	series 9
303.15 K	303.15 K	313.15 K	323.15 K	323.15 K	333.15 K	333.15 K	343.15 K	343.15 K
3354.8	3286.6	3622.7	3876.0	3706.5	3836.8	3764.6	3753.1	4203.3
2551.6	2493.0	2729.4	2890.7	2746.2	2810.2	2751.0	2709.0	3067.9
1859.0	1812.8	1977.2	2081.0	1968.1	1999.0	1953.6	1907.6	2177.6
1316.1	1281.8	1394.9	1462.2	1378.5	1393.4	1360.2	1320.6	1515.7
914.2	889.6	966.6	1010.6	950.8	958.0	934.3	903.7	1041.0
626.9	609.6	661.9	690.9	649.1	652.6	636.0	613.6	708.5
426.2	414.3	449.6	468.8	440.0	441.8	430.3	414.4	479.2
288.2	280.1	303.9	316.6	297.0	298.0	289.9	278.9	322.7
194.1	188.7	204.6	213.1	199.9	200.4	194.9	187.3	216.8
130.5	126.9	137.6	143.3	134.3	134.6	130.7	125.6	145.4

valve arrangement enables the vessels V_A and V_B to be filled or emptied separately and, in addition to the expansion experiment, allows for the compounds in the Burnett vessels to be mixed using a magnetic recirculating pump. The packing surfaces of valves V_1 and V_4 are exposed to the expansion volume V_B, and the packing surfaces of valves V_2 and V_3 are exposed in the opposite direction to the volumes V_A and V_B. Thus, the principal volume V_A and its fixtures are all-metal to prevent contact between the sample gas in vessel V_A and the Teflon packing throughout the lengthy Burnett experiment, except for the time it takes to reach thermal equilibrium after expansion.

The measurement vessel is connected to a diaphragm-type differential pressure transducer (Ruska Model 2413), coupled to an electronic null indicator (Ruska Model 2416). The pressure on either side of the diaphragm is balanced with nitrogen by means of a precision pressure controller (Ruska Model 3981), and the pressure is read by a digital pressure gauge (Ruska Model 7000).

The vessels with the magnetic pump and the pressure transducer are immersed in a thermostatic bath filled with about 45 L of an ethylene glycol and water mixture. The temperature of the bath is kept constant by means of a PID device, piloted by a computer to which the temperature measurement system is also connected. The temperature control and acquisition system relies on two platinum resistance thermometers calibrated according to ITS 90 at the Istituto Metrologico G. Colonnetti (IMGC), of Turin. A Hart Scientific Pt 25 Ω resistance thermometer (model Hart 5680) and a Tersid Pt 100 Ω resistance thermometer are used, respectively, for temperature measurement and control. Both thermometers are connected to a digital temperature indicator (Corradi, RP 7000).

The Burnett constant, N, defined as the ratio of the sum of the volumes of cells A and B and of the volume of cell A at zero pressure, was determined using gaseous helium measurements. After taking measurements at several isotherms, the constant was found to be $N = 1.4969 \pm 0.0001$. This value of the Burnett constant reproduces the pressures of helium with absolute average deviations AAD = 0.418 kPa or AAD = 0.142 as a percentage.

Experimental Procedure, Gas Chromatographic Calibration, and Experimental Uncertainties

Experimental Procedure. To measure the system components, the classical Burnett experimental procedure was followed, repeating expansions until low pressures were achieved. When measuring mixtures, the two vessels were separately filled with different compounds. Then the sample was mixed to homogenize the composition with the aid of the magnetic pump while the expansion valve was kept open. A sample for composition measurement, collected during the first expansion, was used for gas chromatographic analysis.

Gas Chromatographic Calibration. The mixtures for the calibration were prepared in stainless steel bottles of about 250 cm^3 capacity; the mass of charged components, depending on the composition, was varied from (8 to 15) g and was weighed using an analytical balance to ±0.1 mg. On that basis, the mole fraction of the sample was found; then the prepared samples were connected on-line to the gas chromatograph and, after preheating the sample to about 323 K (for sample homogenization), a series of gas chromatographic analyses was performed. The reproducibility of the gas chromatograph for each sample was within 0.1%. The results of the analysis for the series of samples covering a wide range of compositions were then regressed to find the relationship between area ratio and composition in terms of molar fraction. A third-degree polynomial expression, obtained by forcing it to the points corresponding to pure compounds, provides the unknown composition when its peak area ratio is measured. The Carlo Erba Mega series 5380 gas chromatograph was used with a thermal conductivity detector.

Experimental Uncertainties. The uncertainty in the temperature measurements is due to the thermometer and any instability of the bath. The stability of the bath was found to be less than ±0.015 K, and the uncertainty of the thermometer was found to be less than ±0.010 K in our temperature range. The total uncertainty in the temperature measurements was thus lower than ±0.03 K.

Any uncertainty in the pressure measurements is due to the transducer and null indicator system, and to the pressure gauge. The digital pressure indicator (Ruska, model 7000) has an uncertainty of ±0.003% of the full scale. The total uncertainty in the pressure measurement, also influenced by temperature fluctuations due to bath instability, was found to be lower than ±1 kPa.

The uncertainty of the mixture's composition was found to be constantly lower than 0.5% in mole fraction.

Results for CO_2 and R41

For CO_2, 234 experimental points along 7 isotherms (17 sets in all) were collected in the temperature range from 283 K to 363 K and in the pressure range from 11 kPa to 5700 kPa, and they are reported elsewhere.[4] These results were adopted in the present work because the same sample was used. For R41, 90 experimental points along 5 isotherms (9 sets in all) were collected in the temperature range from 303 K to 343 K and for pressures from 125 kPa up to 4200 kPa, and they are shown in Table 1.

The experimental PVT measurements were used to derive the second, B, and third, C, virial coefficients of the virial equation:

$$P = \frac{RT}{V}\left(1 + \frac{B}{V} + \frac{C}{V^2}\right) \tag{1}$$

Table 2. Second and Third Virial Coefficients for R41: $\rho(1)$ Denotes Regressed Initial Density

series	T/K	B/cm^3·mol^{-1}	C/cm^6·mol^{-2}	$\rho(1)$/mol·dm^{-3}	abs(dP)/kPa	$100(P - P_{calc})P^{-1}$/%
1	303.15	−196.1	16100	1.967 18	0.3	0.08
2	303.15	−197.5	16330	1.911 07	0.3	0.09
3	313.15	−182.2	14750	2.005 35	0.4	0.09
4	323.15	−169.0	13480	2.021 89	0.4	0.09
5	323.15	−169.5	13740	1.894 41	0.3	0.08
6	333.15	−158.0	12830	1.840 02	0.3	0.09
7	333.15	−157.1	12510	1.791 02	0.2	0.04
8	343.15	−146.0	11400	1.669 23	0.1	0.04
9	343.15	−144.7	10930	1.936 69	0.1	0.00

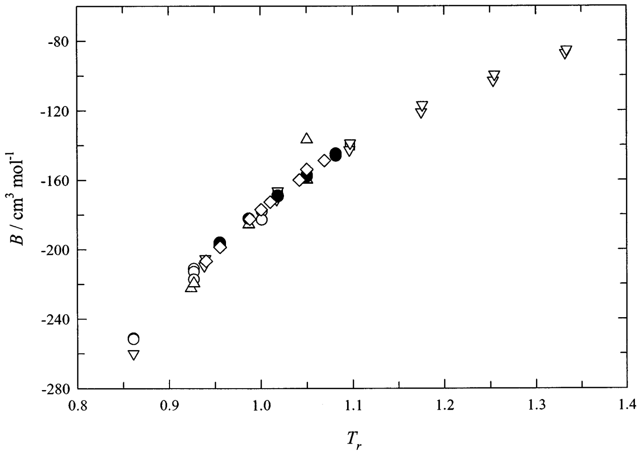

Figure 2. Experimental second virial coefficients for R41 against reduced temperature, T/T_c, where T_c is the critical temperature: ●, present work; ○, Cawood et al.;[5] □, David et al.;[5] △, Hamann et al.;[5] ▽, Michels et al.;[5] ◇, Demiriz et al.[6]

In the regression, each run was treated separately and $(dP)^2$ was used as an objective function, applying the Burnett constant from the helium calibration. The pressure distortion of the Burnett cells was taken into account, as explained elsewhere.[1−3]

Defining the average absolute deviation in pressure as

$$\mathrm{AAD} = \sum_{i=1}^{N} \mathrm{abs(dP)}/N \tag{2}$$

the AAD = 0.273 kPa for R41 was found to be within the estimated experimental uncertainty.

The second and third virial coefficients for R41 are shown in Table 2, together with the pressure deviations from the fit, that always proved to be lower than 0.1%.

In Figure 2, the second virial coefficients for R41 are plotted and compared with values in the literature.[5,6] A good consistency with most of the data in the literature was evident for the second virial coefficient (refs 5 and 6).

For CO_2, all the comparisons with the literature are reported elsewhere.[4]

Results for the Mixtures

For the CO_2 + R41 system, 209 experimental points along 20 sets and 5 isotherms were collected within a temperature range from 303 K to 343 K and a pressure range from 120 kPa to 5600 kPa. The experimental findings are given in Table 3, together with the compressibility factor values, z. The virial coefficients for the mixtures were found by applying the same procedure as that for pure compounds. The values of the second and third virial coefficients (Table 4), along with the virial coefficients for the pure compounds (smoothed as a function of reduced temperature), were used to derive cross virial coefficients. The results are shown in Table 5. The second cross virial coefficients were calculated from the expression

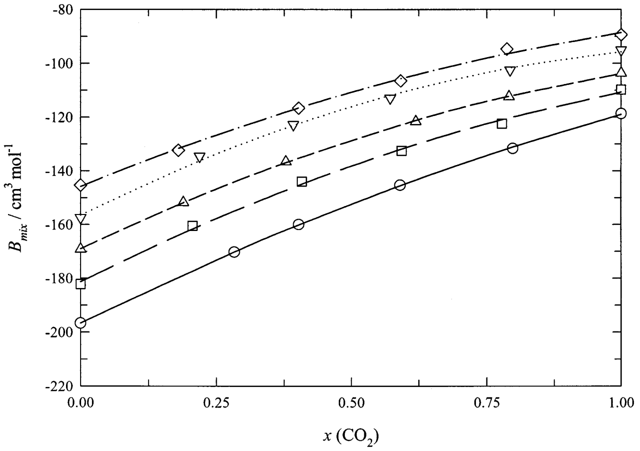

Figure 3. Second virial coefficients for the CO_2 (1) + R41 (2) system against the mole fraction at the following temperatures, T: ○, 303.15; □, 313.15; △, 323.15; ▽, 333.15; ◇, 343.15 K. The lines represent the correlated values.

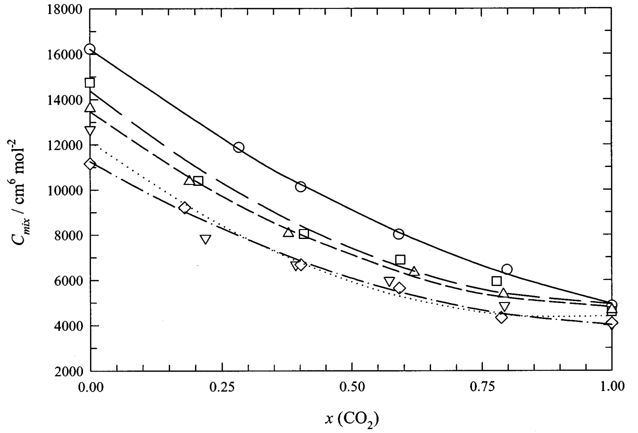

Figure 4. Third virial coefficients for the CO_2 (1) + R41 (2) system against the mole fraction at the following temperatures, T: ○, 303.15; □, 313.15; △, 323.15; ▽, 333.15; ◇, 343.15 K. The lines represent the correlated values.

$$B_{\mathrm{m}} = \sum_{i=1}^{n}\sum_{j=1}^{n} B_{ij}x_i x_j \tag{3}$$

for each experimental datum point. Next, the B_{12} values found for each temperature were averaged; the averaged B_{12} values, given in Table 5, were used to calculate the deviations of B_{m} from the experimental values shown in Table 4. The third cross virial coefficients were calculated from the defining equation

$$C_{\mathrm{m}} = \sum_{i=1}^{n}\sum_{j=1}^{n}\sum_{k=1}^{n} C_{ijk}x_i x_j x_k \tag{4}$$

Table 3. Experimental Pressures Measured during Burnett Expansions and Regressed Compressibility Factors of the CO_2 (1) + R41 (2) System

T/K = 303.15

series 1		series 2		series 3		series 4	
$x_1 = 0.283$		$x_1 = 0.402$		$x_1 = 0.590$		$x_1 = 0.798$	
P/kPa	z	P/kPa	z	P/kPa	z	P/kPa	z
3455.9	0.7182	3745.5	0.7091	4156.5	0.7033	4628.3	0.6983
2578.9	0.8022	2808.4	0.7959	3127.5	0.7921	3491.8	0.7886
1854.5	0.8635	2025.1	0.8591	2259.3	0.8566	2527.0	0.8543
1300.8	0.9066	1423.2	0.9038	1589.6	0.9021	1779.6	0.9006
897.8	0.9368	983.5	0.9349	1099.1	0.9337	1231.1	0.9326
613.1	0.9576	672.2	0.9564	751.4	0.9555	842.0	0.9547
415.8	0.9721	456.1	0.9715	510.0	0.9708	571.5	0.9700
280.7	0.9823	308.0	0.9821	344.5	0.9815	386.0	0.9807
188.9	0.9896	207.3	0.9896	231.9	0.9892	259.9	0.9886
126.9	0.9950	139.3	0.9955	155.9	0.9951	174.7	0.9946

T/K = 313.15

series 5		series 6		series 7		series 8	
$x_1 = 0.206$		$x_1 = 0.408$		$x_1 = 0.593$		$x_1 = 0.778$	
P/kPa	z	P/kPa	z	P/kPa	z	P/kPa	z
4622.8	0.6302	5645.6	0.5668	5581.0	0.6277	5382.3	0.6845
3600.8	0.7348	4556.4	0.6847	4357.3	0.7335	4083.3	0.7773
2665.8	0.8143	3455.2	0.7772	3229.5	0.8138	2967.7	0.8457
1907.6	0.8722	2512.5	0.8460	2311.9	0.8721	2096.6	0.8943
1334.2	0.9132	1775.9	0.8950	1617.0	0.9130	1453.8	0.9282
919.1	0.9417	1231.8	0.9293	1113.9	0.9415	996.0	0.9519
626.8	0.9613	843.8	0.9529	759.4	0.9608	676.8	0.9683
424.5	0.9746	573.2	0.9689	514.3	0.9740	457.5	0.9798
286.2	0.9837	387.2	0.9799	346.7	0.9829	308.2	0.9881
192.3	0.9894	260.7	0.9874	233.0	0.9890	207.3	0.9946
128.9	0.9928	175.0	0.9921	156.2	0.9925	139.2	1.0000

T/K = 323.15

series 9		series 10		series 11		series 12	
$x_1 = 0.189$		$x_1 = 0.378$		$x_1 = 0.619$		$x_1 = 0.791$	
P/kPa	z	P/kPa	z	P/kPa	z	P/kPa	z
4257.2	0.7144	5138.4	0.6793	5587.6	0.6915	5596.0	0.7197
3176.6	0.7980	3899.6	0.7717	4217.2	0.7813	4170.0	0.8027
2286.6	0.8598	2838.8	0.8409	3058.6	0.8482	2997.8	0.8639
1606.1	0.9040	2009.1	0.8909	2158.3	0.8959	2102.7	0.9070
1109.6	0.9349	1394.9	0.9259	1495.6	0.9293	1451.1	0.9369
758.3	0.9564	956.3	0.9502	1023.9	0.9523	990.6	0.9574
514.3	0.9710	650.1	0.9670	695.4	0.9682	671.5	0.9715
347.1	0.9809	439.4	0.9783	469.9	0.9793	453.0	0.9812
233.4	0.9875	295.9	0.9860	316.4	0.9871	304.7	0.9877
156.6	0.9918	198.7	0.9914	212.6	0.9926	204.4	0.9921
		133.2	0.9948	142.6	0.9967	137.0	0.9950

T/K = 333.15

series 13		series 14		series 15		series 16	
$x_1 = 0.219$		$x_1 = 0.392$		$x_1 = 0.572$		$x_1 = 0.793$	
P/kPa	z	P/kPa	z	P/kPa	z	P/kPa	z
4136.0	0.7679	5046.6	0.7372	5488.2	0.7409	5565.8	0.7639
3016.4	0.8383	3728.6	0.8154	4045.3	0.8174	4063.7	0.8348
2137.2	0.8891	2666.7	0.8729	2888.7	0.8738	2882.0	0.8863
1484.9	0.9247	1863.4	0.9130	2018.3	0.9138	2004.2	0.9226
1017.9	0.9489	1282.7	0.9408	1389.2	0.9416	1375.1	0.9475
692.1	0.9657	874.8	0.9605	946.8	0.9606	935.3	0.9647
467.7	0.9770	592.5	0.9737	641.1	0.9736	632.5	0.9765
314.8	0.9843	399.3	0.9824	432.2	0.9824	426.0	0.9846
211.2	0.9886	268.3	0.9879	290.4	0.9884	286.2	0.9901
141.4	0.9909	179.8	0.9911	194.8	0.9922	191.9	0.9938
		120.3	0.9924	130.5	0.9947	128.5	0.9965

T/K = 343.15

series 17		series 18		series 19		series 20	
$x_1 = 0.180$		$x_1 = 0.402$		$x_1 = 0.591$		$x_1 = 0.787$	
P/kPa	z	P/kPa	z	P/kPa	z	P/kPa	z
4064.7	0.7916	5337.5	0.7508	5565.1	0.7652	5631.7	0.7905
2930.2	0.8542	3914.8	0.8243	4057.2	0.8350	4064.8	0.8541
2061.5	0.8996	2787.5	0.8786	2876.3	0.8861	2861.0	0.8998
1426.1	0.9315	1943.4	0.9169	1999.6	0.9222	1979.2	0.9318
975.3	0.9536	1336.1	0.9436	1372.4	0.9474	1353.5	0.9539
662.0	0.9690	910.0	0.9620	933.5	0.9646	918.6	0.9691
447.0	0.9794	615.9	0.9747	631.2	0.9764	620.2	0.9794
300.8	0.9865	415.1	0.9834	425.2	0.9845	417.3	0.9864
202.0	0.9914	279.1	0.9895	285.7	0.9901	280.1	0.9911
135.4	0.9953	187.3	0.9939	191.6	0.9938	187.7	0.9941
		125.5	0.9971	128.3	0.9967	125.6	0.9960

Table 4. Experimental and Calculated Second and Third Virial Coefficients for the CO_2 (1) + R41 (2) System

T/K	x_1	B_m(exp)[a]	B_m(calc)[a]	dB_m[a]	C_m(exp)[b]	C_m(calc)[b]	dC_m[b]
303.15	0.283	−170.3	−170.4	0.1	11880	11740	140
	0.402	−160.0	−160.4	0.3	10130	10210	−80
	0.590	−145.3	−145.7	0.3	8020	8150	−130
	0.798	−131.7	−131.2	−0.6	6460	6310	150
313.15	0.206	−160.6	−162.2	1.6	10400	10420	−20
	0.408	−144.0	−145.1	1.1	8040	8050	−10
	0.593	−132.6	−131.7	−0.8	6890	6850	40
	0.778	−122.4	−120.5	−1.9	5940	5990	−50
323.15	0.189	−151.8	−152.0	0.2	10380	10370	10
	0.378	−136.6	−137.0	0.4	8070	8100	−30
	0.619	−121.6	−121.2	−0.4	6330	6260	70
	0.791	−112.4	−112.1	−0.2	5370	5420	−50
333.15	0.219	−134.6	−136.7	2.1	7860	8190	−330
	0.392	−122.8	−123.1	0.3	6670	6480	190
	0.572	−112.9	−111.6	−1.3	5980	5720	260
	0.793	−102.5	−101.2	−1.4	4860	5180	−320
343.15	0.180	−132.4	−131.3	−1.1	9210	9100	110
	0.402	−116.7	−116.3	−0.4	6690	6900	−210
	0.591	−106.5	−105.7	−0.8	5640	5430	210
	0.787	−94.7	−96.8	2.1	4330	4430	−100

[a] In $cm^3 \cdot mol^{-1}$. [b] In $m^6 \cdot mol^{-2}$.

Table 5. Averaged Second and Third Virial Coefficents for the CO_2 (1) + R41 (2) System

T/K	B_{11}[a]	B_{12}[a]	B_{22}[a]	C_{111}[b]	C_{112}[b]	C_{122}[b]	C_{222}[b]
303.15	−118.8	−147.3	−196.8	4860	7029	10174	16219
313.15	−109.8	−130.4	−182.2	4600	7434	5771	14748
323.15	−103.6	−120.7	−169.2	4670	5792	6910	13612
333.15	−95.1	−105.5	−157.5	4020	6980	3298	12674
343.15	−89.5	−103.7	−145.4	4090	3942	7190	11165

[a] $cm^3 \cdot mol^{-1}$. [b] $cm^6 \cdot mol^{-2}$.

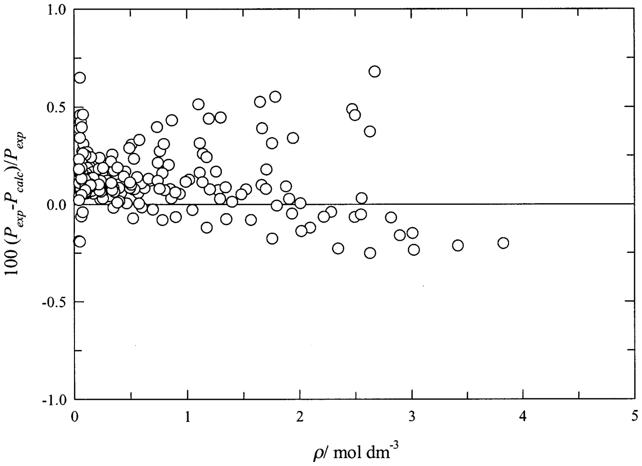

Figure 5. Deviations in pressure produced by the REFPROP 6.01[7] against density.

The averaged values of the third cross virial coefficients are included in Table 5. Using these values, the third virial coefficients for the mixtures were recalculated for each point. The results, together with the deviations, are also given in Table 4. The overall AAD for B_m was evaluated at 0.9 $cm^3 \cdot mol^{-1}$, while the AAD for C_m was 126 $cm^6 \cdot mol^{-2}$.

For the system being plotted against the mole fraction in Figures 3 and 4, respectively, the second and third virial coefficients calculated from eqs 3 and 4, with the cross virial coefficients given by eqs 5−7,

$$B_{12} = (B_{11} + B_{22})/2 \tag{5}$$

$$C_{112} = (2C_{111} + C_{222})/3 \tag{6}$$

$$C_{122} = (C_{111} + 2C_{222})/3 \tag{7}$$

show very small positive and negative deviations.

The experimental findings were also compared with the prediction obtained with the REFPROP 6.01[7] software. The calculations were performed at the highest experimental pressures of each run, adopting regressed values for $\rho(1)$, B_m, and C_m. The differences in pressure are given in Figure 5, showing a consistency within 0.6%.

Conclusions

This work reports the experimental findings for the CO_2 + R41 system obtained with the Burnett method. The performance of the apparatus was checked by means of measurements for R41 and CO_2, and a good consistency was observed between the virial coefficients obtained and those reported in the literature. The CO_2 + R41 system was studied over five isotherms, deriving second and third cross virial coefficients. Over the entire temperature range, the second virial coefficients showed positive deviations, while the third virial coefficients revealed negative deviations. The prediction obtained using REFPROP 6.01 proved consistent with our results.

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