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The Virial Coefficients of Five Binary Mixtures of Fluorinated Methanes and Ethanes

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We have made new measurements of the gas-phase *PVT* surface of five binary mixtures of hydrofluorocarbons (HFCs) in a Burnett/isochoric apparatus. The components chosen all have moderate to large reduced dipole moments. We present *PVT* data, derived mixture virial coefficients, cross second virial coefficients, and binary interaction parameters for these systems, and we compare the results with a recently published model for calculating second and third viral coefficients of polar gases and their mixtures. That model accounts for the polar nature of the molecules with a term containing the reduced dipole moment, μ_R, and it contains mixing rules for the substance-specific parameters needed to calculate the second and third cross virial coefficients. The model and data are in satisfactory agreement, and the model can be used to greatly extend the useful range of the limited set of data.

KEY WORDS: hydrofluorocarbons; *PVT*; polar gases; refrigerants; virial coefficients.

1. INTRODUCTION

In a previous paper, one of the authors (L.A.W.) [1] presented a model for calculating second and third virial coefficients of polar gases and their mixtures. That model was used successfully to correlate measurements in the literature for the virial coefficients of the hydrofluorocarbons (HFCs) and hydrochlorofluorocarbons (HCFs). Virial coefficients of their mixtures were calculated by using conventional mixing rules. However, no generally accepted mixing rules exist for expressions which account for the polar

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nature of molecules. Furthermore, the data required to test candidate rules for the second virial coefficients of polar/polar binary mixtures are scarce. Data for the corresponding third virial coefficients are virtually nonexistent.

Therefore, we have used the National Institute of Standards and Technology (NIST) Burnett/isochoric *PVT* apparatus to make selected gas-phase measurements of the densities of five equimolar binary mixtures of partially fluorinated methanes and ethanes: difluoromethane (R32) + 1,1,1,2-tetrafluoroethane (R134a), R32 + pentafluoroethane (R125), R125 + R134a, R134a + 1,1-difluoroethane (R152a), and R125 + 1,1,1-trifluoroethane (R143a). The molecules chosen all have moderate to large dipole moments, therefore, they provide a good test of mixing rules for that property. Several of the mixture are also of potential interest to the refrigeration industry.

In the next sections, we describe the measurements and present the density data and derived virial coefficients. We also compare the experimental results with the virial coefficients calculated from the model. Finally, we compare our results with the very limited data which have recently become available in the literature.

2. EXPERIMENTS

The apparatus has been described many times before [2, 3], and we dispense with any further description here. Samples having approximately equimolar composition were prepared by measuring the components with a gas burette and distilling them into a transfer cylinder that was cooled with liquid nitrogen. The exact composition was determined gravimetrically with an estimated standard uncertainty of $\pm 5 \times 10^{-4}$ in mole fraction. Approximately 10-g quantities were prepared and stored in the cylinders, and in each case, the entire sample was loaded into the apparatus. This procedure avoided fractionation of the components during the loading process. For each binary mixture, one set of Burnett expansions was measured at 373 K. The pressure was measured with a deadweight pressure balance, which had a sensitivity of about ±20 Pa. After each expansion, the apparatus was cooled along the isochore, and pressure measurements were made at 338 and 353 K; for one mixture, measurements were also made at 333 and 363 K, and for another, measurements were also made at 303 and 323 K. A quartz Bourdon pressure gauge with an estimated standard uncertainty of 150 Pa was employed for these latter measurements.

In this way, 11 to 27 *PVT* data were measured for each binary system. The results and the exact compositions of the mixtures appear in Table I. The isochoric slopes were used to adjust the data for small temperature

Table I. Experimental Results for the Compositions and Densities of the Mixtures

Pressure (MPa)	Density ($\text{mol} \cdot \text{L}^{-1}$)
R32(1) + R134a(2) $x1 = 0.5084$	
$T = 373.15$ K	
0.36659	0.12115
0.64060	0.21589
1.10224	0.38473
1.84466	0.6856
2.93780	1.2217
4.29680	2.1772
$T = 353.15$ K	
0.34576	0.12125
0.60234	0.21607
1.0303	0.38505
1.7042	0.6861
2.6569	1.2228
3.7092	2.1790
$T = 338.15$ K	
0.3301	0.12133
0.5733	0.21621
0.9748	0.38530
1.5973	0.6866
2.4361	1.2236
R32(1) + R125(2) $x1 = 0.5456$	
$T = 373.15$ K	
0.32245	0.10591
0.56631	0.18873
0.98307	0.33633
1.67174	0.5993
2.74171	1.0680
4.22672	1.9033

Table I. *(Continued)*

$T = 353.15$ K	
0.3043	0.10600
0.5335	0.18889
0.9221	0.33661
1.5553	0.5998
2.5112	1.0689
3.7536	1.9049
$T = 338.15$ K	
0.5087	0.18901
0.8755	0.33682
1.4653	0.6002
2.3325	1.0696
3.3827	1.9061
R125(1) + R134a(2) $x1 = 0.4950$	
$T = 373.15$ K	
0.21116	0.069193
0.37150	0.12330
0.64701	0.21973
1.10629	0.39158
1.83039	0.6978
2.85573	1.2435
4.03194	2.2159
$T = 353.15$ K	
0.6077	0.21992
1.0317	0.39191
1.6840	0.69840
2.5541	1.2445
$T = 338.15$ K	
0.5778	0.22006
0.9746	0.39216
1.5704	0.6988
$T = 323.15$ K	
0.5475	0.2202
0.9164	0.3924

Table I. *(Continued)*

$T = 303.15$ K	
0.1697	0.0694
0.2959	0.1237
R134a(1) + R152a(2) $x1 = 0.4970$	
$T = 373.15$ K	
0.28385	0.09399
0.49560	0.16749
0.85041	0.29848
1.41525	0.5319
2.22810	0.9479
3.16723	1.6892
$T = 353.15$ K	
0.26765	0.09405
0.46579	0.16760
0.79405	0.29867
1.30511	0.5322
2.00303	0.9485
R125(1) + R143a(2) $x1 = 0.5090$	
$T = 373.15$ K	
0.24521	0.08044
0.43103	0.14335
0.74954	0.25545
1.27816	0.45522
2.10669	0.8112
3.27418	1.4456
$T = 363.15$ K	
0.2384	0.08047
0.4188	0.14340
0.7268	0.25555
1.2348	0.45541
2.0217	0.8115
3.1003	1.4462

Table I. *(Continued)*

$T = 353.15$ K	
0.2314	0.08047
0.4064	0.14341
0.7037	0.25556
1.1910	0.45541
1.9353	0.8115
2.9230	1.4462
$T = 343.15$ K	
0.2248	0.08055
0.3938	0.14354
0.6805	0.25579
1.1466	0.45583
1.8472	0.8123
2.7413	1.4475
$T = 333.15$ K	
0.2179	0.08050
0.3812	0.14346
0.6572	0.25566

differences from each isotherm. Then the data on each isotherm were analyzed with a truncated virial equation,

$$P = RT\rho[1 + B_m\rho + C_m\rho^2] \tag{1}$$

by means of a Burnett analysis routine. Here, B_m and C_m are the mixture virial coefficients. Maximum densities used in the analysis were in the range 1.0–1.3 mol · L^{-1}. At the lowest temperatures, when there were insufficient data for a Burnett analysis, we fitted a polynomial to the data. In that case the densities of the isochores were taken from the higher temperature isotherm fits. Because we have made measurements on only one composition for each system, the cross third virial coefficients, C_{112} and C_{122}, cannot be determined. The cross second virial, B_{12}, can be calculated (albeit, without desirable redundant data), and B_m, C_m, and B_{12} are tabulated for each system in Table II. The nominal estimated standard uncertainties are ± 0.5 cm^3 · mol^{-1} for B_m, ± 1 cm^3 · mol^{-1} for B_{12}, and ± 1000 (cm^3 · mol^{-1})^2 for C_m. These estimates are the results of numerical experiments that estimate the effects of systematic and random errors in the experimental pressures.

Table II. Mixture Second and Third Virial Coefficients and the Cross Second Virial Coefficients for the Five Systems

T (K)	B_m ($\mathrm{cm^3 \cdot mol^{-1}}$)	C_m ($(\mathrm{cm^3 \cdot mol^{-1}})^2$)	B_{12} ($\mathrm{cm^3 \cdot mol^{-1}}$)	k_{12}
		R32(1) + R134a(2) $x1 = 0.5084$		
373.15	−206.3	18,102	−195.4	0.040
353.15	−239.4	21,860	−227.6	0.038
338.15	−268.6	24,641	−255.5	0.036
		R32(1) + R125(2) $x1 = 0.5456$		
373.15	−177.3	14,705	−171.2	0.038
353.15	−205.1	16,991	−199.0	0.035
338.15	−230.7	19,522	−225.2	0.029
		R125(1) + R134a(2) $x1 = 0.4950$		
373.15	−237.4	22,938	−227.9	0.032
353.15	−273.9	25,742	−263.7	0.030
338.15	−306.9	28,159	−296.7	0.027
323.15	−349.3	33,200	−337	0.022
		R134a(1) + R152a(2) $x1 = 0.4970$		
373.15	−282.9	28,843	−285.0	0.000
353.15	−326.4	31,795	−330.1	0.000
		R125(1) + R143a $x1 = 0.5090$		
373.15	−219.1	22,344	−214.4	0.020
363.15	−234.6	23,547	−230.6	0.016
353.15	−251.2	24,457	−246.2	0.020
343.15	−271.8	27,251	−267.6	0.016
333.15	−298		−280	0.027

3. THE MODEL

The model for the second virial coefficient is a development of the earlier work of Pitzer and Curl [4] and of Tsonopoulos [5]. The reduced second virial coefficient is given by

$$BP_c/RT_c = f^{(0)}(T_r) + \omega f^{(1)}(T_r) + a/T_r^6 \tag{2}$$

where P_c and T_c are the critical pressure and temperature, ω is the Pitzer acentric factor, and $T_r = T/T_c$. The f's are "universal" polynomials; that is, they apply to all gases. They are fully described in Ref. 1. The first two terms are sufficient to describe the second virial coefficients of nonpolar gases; the only required substance-specific parameters are the critical parameters and ω.

The third term, introduced by Tsonopoulos [5], accounts for the additional contribution to B due to permanent dipole moments. The parameter a is taken to be proportional to the reduced dipole moment squared, defined here as

$$\mu_R = 0.9869 \times 10^5 \, \mu^2 P_c / T_c^2 \tag{3}$$

where μ is the dipole moment in debye, T_c is in kelvin, and P_c is in bar. We showed in Ref. 1 that for many polar molecules a can be represented quite satisfactorily by the expression

$$a = -9 \times 10^{-7} \mu_R^2 \tag{4}$$

Values for the necessary parameters, μ, P_c, T_c, v_c, and ω, for all of the components were given in Ref. 1.

The form of the third virial coefficient was taken from the work of Kohler and co-workers [6]. In summary, C is given by the expression

$$C = C_h + (B - B_h)^2 \mathscr{T}_c \mathscr{F}(T_r) \tag{5}$$

where B is the second virial coefficient, $\mathscr{T}_c$ is an amplitude which determines the value of C at T_c, and $\mathscr{F}(T_r)$ is a simple function of temperature which has the value unity at the critical temperature. We showed in Ref. 1 that for nonassociating fluids, $\mathscr{T}_c$ and $\mathscr{F}(T_r)$ sould be weak functions of μ_R,

$$\mathscr{T}_c = c_1 + c_2 \mu_R^3 \tag{6a}$$

and

$$\mathscr{F}(T_r) = \mathscr{C} + (1 - \mathscr{C})/T_r^4 \tag{6b}$$

with

$$\mathscr{C} = c_3 + c_4 \mu_R^3 \tag{6c}$$

Values of $c_1 - c_4$ were given in Ref. 1.

We simplified the implementation of Eq. (5) by letting B_h be the hard-sphere volume, b, and $C_h = 0.625\,b^2$. An adequate approximation for b, found to work well for our purposes, is $b = 0.36v_c$, where v_c is the critical molar volume.

When the above relationships are applied to binary mixtures, mixing rules are required for the substance-specific parameters T_c, P_c, ω, and μ_R. For the first three, we employed widely used formulae [5], and only the first is important to note here,

$$T_{c12} = (T_{c1} T_{c2})^{1/2}(1 - k_{12}) \tag{7}$$

where k_{12} is the binary interaction parameter, which must be found from a measurement. For the reduced dipole moment of mixtures, we follow O'Connell and Prausnitz [7],

$$\mu_{R_{12}} = 0.9869 \times 10^5 \mu_1 \mu_2 P_{c12}/T_{c12}^2 \tag{8}$$

where μ_1 and μ_2 are the component dipole moments, and P_{c12} is calculated as shown in [5]; units are the same as in Eq. (3).

With the necessary pure component parameters and a value for k_{12}, one can calculate the cross second virial coefficient B_{12} from Eqs. (2)–(4), (7), and (8) with the assumption that it has the same corresponding-states behavior as the B's of the pure components, and also with the assumption that the mixing rules are physically realistic. Then the mixture second virial coefficient, B_m, can be calculated for any given composition. Conversely, from an experimental value for B_m we can find the value of k_{12}.

For the mixture third virial coefficient, C_m, no other experimental parameters are needed, but we must define a mixing rule for μ_R for three molecule interactions. We define

$$\mu_{R_{112}} = (\mu_{R_1} \mu_{R_{12}}^2)^{1/3} \tag{9a}$$

$$\mu_{R_{122}} = (\mu_{R_2} \mu_{R_{12}}^2)^{1/3} \tag{9b}$$

$$B_{h12} = (B_{h1} + B_{h2})/2 \tag{9c}$$

$$C_{h112} = 0.625(B_{h1}^2 + 2B_{h12}^2)/3 \tag{9d}$$

$$C_{h122} = 0.625(B_{h2}^2 + 2B_{h12}^2)/3 \tag{9e}$$

and in Eq. (5) we replace

$$(B - B_h) \quad \text{with} \quad [(B_{ii} - B_{hii})(B_{ij} - B_{hij})^2]^{1/3} \tag{9f}$$

Equations (9c)–(9f) are reasonable rules for hard spheres. They were tested in Ref. 1, and the results justify their use. Equations (8), (9a), and (9b) have the property that for a polar/nonpolar mixture, $\mu_{R_{12}} = \mu_{R_{112}} = \mu_{R_{122}} = 0$ in keeping with the idea that induced dipoles are relatively unimportant for these calculations. The model predicts the same values for B_m and C_m that result for a mixture of otherwise similar nonpolar components. This result was also tested against available data in Ref. 1, and the agreement in the case of polar/nonpolar systems was satisfactory. However, no published data could be found for the third virial coefficients of polar/polar systems. Therefore, the data given here make possible the first test of Eqs. (9a) and (9b).

4. COMPARISONS

4.1. Second Virial Coefficient

Reference 1 gave comparisons between the model and the experimental data for 15 candidate refrigerants whose molecules varied in polarity from nonpolar to highly polar. In addition, comparisons have been made with data for several monatomic and diatomic gases, with light hydrocarbons, with carbon dioxide, and with several nonpolar and nonpolar/polar

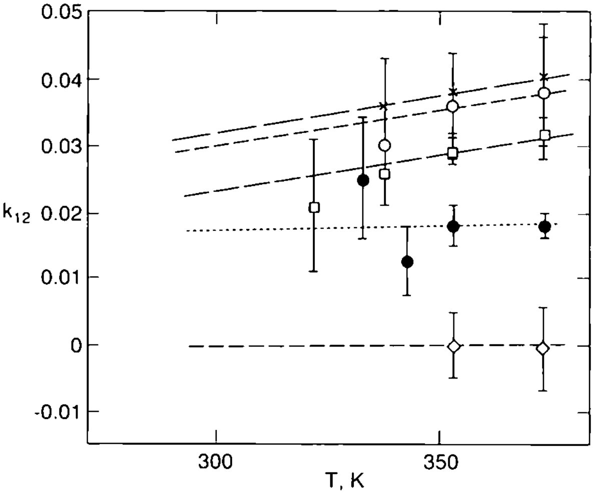

Fig. 1. Binary interaction parameters for the five systems: (×) R32 + R134a; (○) R32 + R125; (□) R125 + R134a; (●) R125 + R143a; (◇) R134a + R152a.

binary mixtures. In general, the agreement for B was of the order of 1–2% or better for pure fluids. In the case of mixtures, the experimental values of B_m were used to obtain k_{12}. A single value of k_{12} sufficed to reproduce the experimental B_m or B_{12} data over a wide temperature range, where data were available for comparison.

From the experimental values of B_m and the mixture compositions given in Table I and Eqs. (2)–(4), (7), and (8), we obtained values for k_{12}

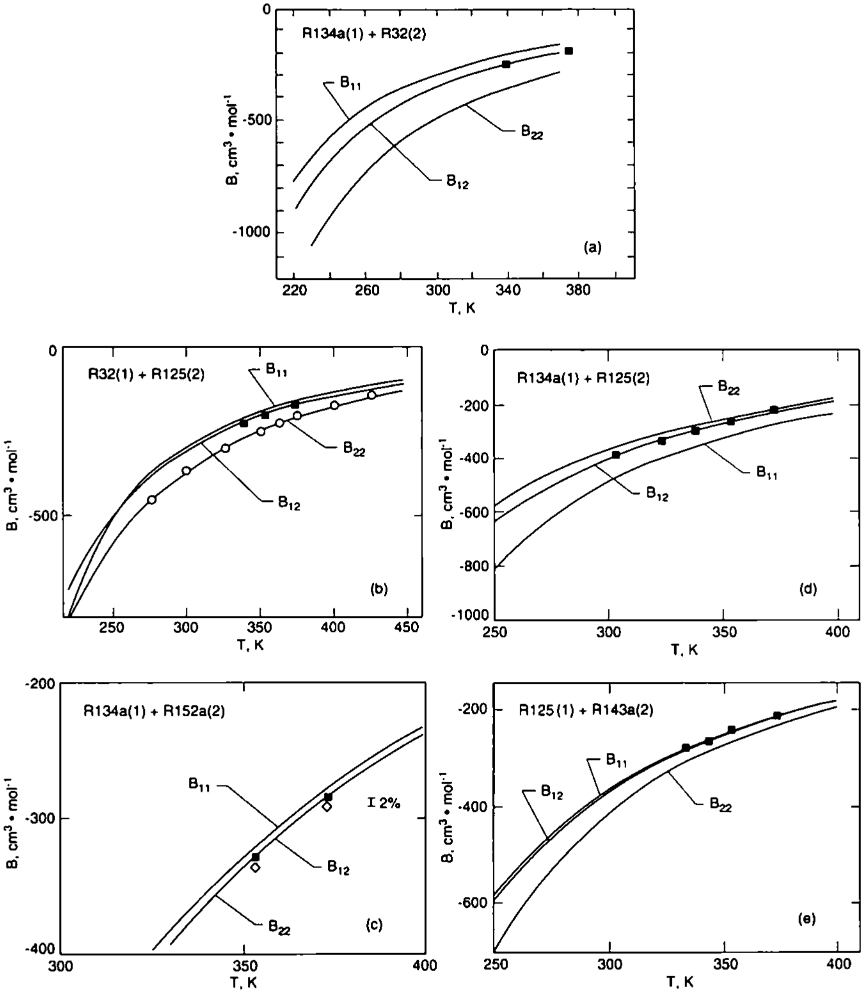

Fig. 2. Experimental cross second virial coefficients and calculated values for the cross virials and the pure fluid virials for the five systems: (■) this work; (○) Ref. 8; (◇) B_{12} from Ref. 10.

and for B_{12} for each system at each experimental temperature. The results for k_{12} are shown in Fig. 1. The error bars shown reflect the estimated uncertainties in B_{11}, B_{22}, and B_m. The figure suggests that k_{12} may be a function of temperature for the three systems having the largest values of the binary interaction parameter, R32 + R134a, R32 + R125, and R125 + R134a. However, due to the size of the uncertainty bars, constant values are not ruled out. Measurements at lower temperatures would help to settle this issue. The other two systems, R125 + R143a and R134a + R152a, have smaller values for k_{12}, and there is no apparent variation with temperature. Numerical values for the binary parameters are given in Table II.

Figures 2a–e show the calculated curves for B_{11}, B_{22}, and B_{12} for each system. In this figure averaged values for k_{12} (not functions of T) were used. In each figure the derived B_{12} data are shown but not the pure fluid virial coefficient data. The reader is referred to Ref. 1 for those comparisons. The one exception is the comparison with data for R125, from Ref. 8, which was not shown in Ref. 1. For the system R134a + R152a we also show the cross virials derived from the data of Baehr and Tillner-Roth [9, 10] at our experimental temperatures. These comparisons are discussed below. The location of B_{12} reltive to the B's of the pure components is not a simple function of the dipole moments. In some cases the B_{12} curve is closer to the B of the more polar component; in other cases it is not. This behavior is summarized by the values of k_{12} for each system.

4.2. Third Virial Coefficient

In Ref. 1 the model calculations were compared with the available data for C and for the very limited data for cross third virial coefficients. The deviations could be approximated by, $\Delta C \leqslant 0.05C + 500$ $(\mathrm{cm}^3 \cdot \mathrm{mol}^{-1})^2$. The present experimental results for the mixture third virial coefficients are compared with the results from Eqs. (5)–(9) for each system in Figs. 3a–e. Overall the agreement is quite satisfactory, and there are no deviations greater than our estimated uncertainties for the pure fluids, given above. Figure 3b includes results from the Burnett-isochoric measurements of Ref. 8 for pure R125, which were not previously available in Ref. 1. Figure 3c shows some of our own unpublished data for pure R134a [17] and also some values of C_m derived from the data of Tillner-Roth [10] for the nearly equimolar mixture. These comparisons are discussed further below.

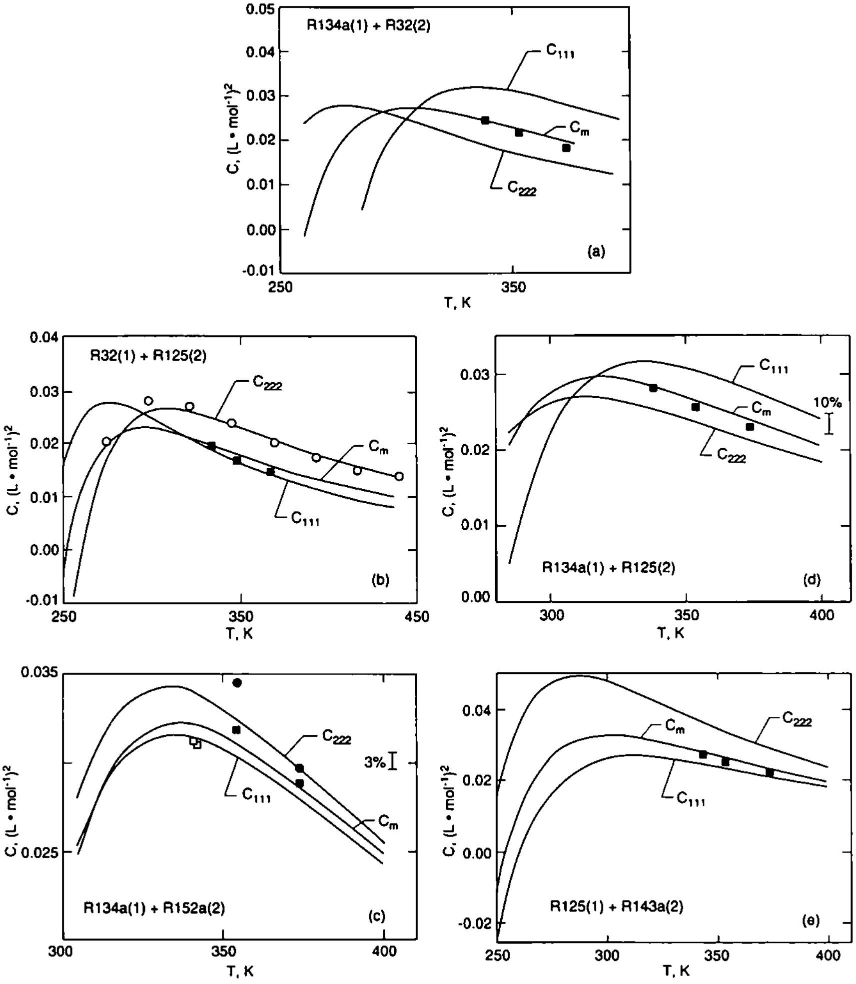

Fig. 3. Experimental mixture third virial coefficients and calculated values: (■) C_m, this work; (○) Ref. 8; (●) C_m, Ref. 10; (□) Ref. 17.

4.3. Comparison with Other Results

PVTx data have recently been reported for two of these systems. Tillner-Roth [10] made measurements on the system R134a + R152 between 293 and 433 K for three compositions. Sato et al. [11] reported virial coefficients for the system R134a + R32 from 320 to 440 K at four compositions as well as virial coefficients for pure R32.

The data for the system R134a + R152a from Ref. 10 for the composition $x_1 = 0.498$ were analyzed with our Burnett analysis program, and the derived virial coefficients are compared with our own coefficients (for $x_1 = 0.497$) in Figs. 2c and 3c. The difference in B_m is $3\ \mathrm{cm^3 \cdot mol^{-1}}$, and the corresponding difference in B_{12} is $6\ \mathrm{cm^3 \cdot mol^{-1}}$, or 2%, as seen in Fig. 2c. In Fig. 3c the difference in C_m at 373 K is about 1000 $(\mathrm{cm^3 \cdot mol^{-1}})^2$, or about 3%. It should be noted that these differences represent the level of agreement not just between the model and data, but also between two sets of data. The differences are no larger than the combined estimated uncertainties in the two data sets. These two sets of virial coefficients would predict the same densities within 0.1–0.2% at pressures

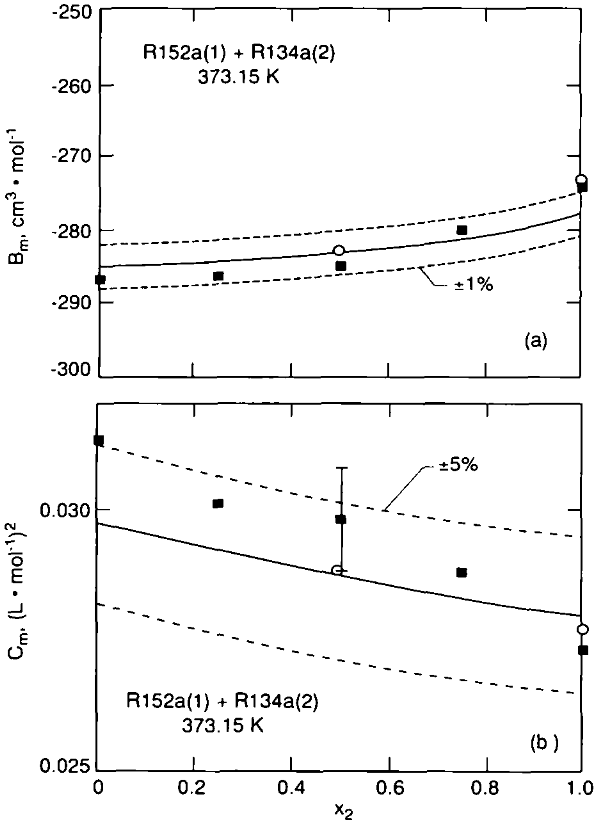

Fig. 4. Mixture virial coefficients for R152a + R134a as a function of composition at 373.15 K; (■) Refs. 9 and 10; (○) this work; (——) model calculation.

up to about 30 bar. Figures 4a and b show how B_m and C_m vary with composition at 373 K for this mixture. The agreement between the model and the data is satisfactory.

Our calculated results for the system R32 + R134a are compared with the experimental data of Sato et al. [11] in Figs. 5a and 5b. Figure 5a shows that the agreement for B is quite good for pure R32, but there is some disagreement for the mixtures. The experimental B_m's from Ref. 11

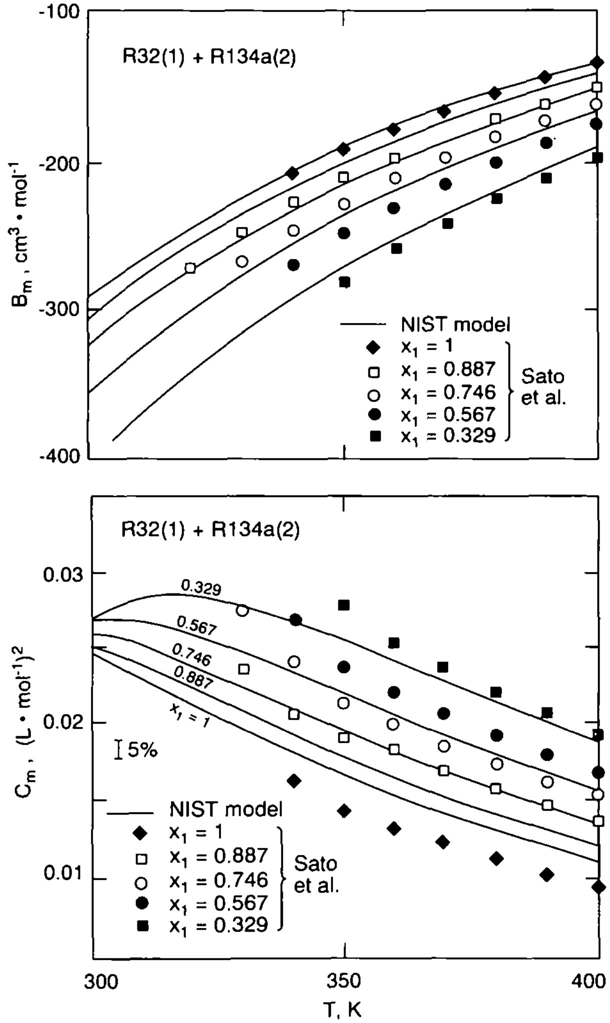

Fig. 5. Mixture virial coefficients for the system R32 + R134a: symbols, Ref. 11; ——, model.

are more negative than the calculated values by about 6–12 $cm^3 \cdot mol^{-1}$ (2.5–5%). In Fig. 5b the third virial coefficients from Ref. 11 are more positive than our calculated values by 1800–2200 $(cm^3 \cdot mol^{-1})^2$, or about 10%, except for those of pure R32, which are more negative. Densities calculated with their mixture virials and with the model would agree within 0.7% for pressures up to about 25 bar. The results of Sato et al. agree well with our own experimental results [2] for pure R32.

The binary interaction parameter, k_{12}, has been measured several times for the system R134a + R152a. Tillner-Roth [10] made vapor liquid equilibrium (VLE) measurements in the temperature range 313–373 K, and he found values for k_{12} ranging from −0.0068 to −0.0085 for the

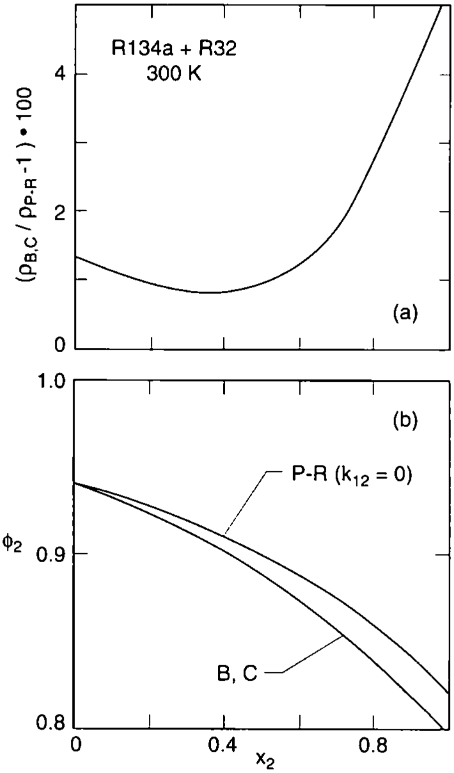

Fig. 6. Comparison of results from the virial coefficient model with a Peng–Robinson (P–R) equation with $k_{12} = 0$ for the saturated vapor (dew curve) of the system R134a + R32 at 300 K; (a) relative density difference; (b) fugacity coefficient.

Redlich–Kwong equation of state. Kleiber [13] also made VLE measurements at temperatures from 255 K to 298 K and found average values from −0.0056 to −0.0065 using three cubic equations of state. Sand et al. [14] made P-x measurements at 273 K and found values from −0.005 to +0.016 with the Carnahan–Starling–DeSantis equation. Morrison [15] found a value of −0.007 from P-x measurements. Our value for this quantity, 0.000 ± 0.003, from virial coefficients, is in reasonably good agreement with these determinations, and together these results support the conclusion that there is no temperature dependence for this system.

The main uses for the results of this work are calculations of mixture densities and fugacity coefficients. In Figs. 6a and b we compare some selected results of this work with values calculated with a Peng–Robinson equation for the system R134a + R32. Comparisons are made at 300 K for the saturated vapor along the dew curve. In Fig. 6a density differences vary from about 1% for R134a-rich compositions up to about 6% for pure R32. In Fig. 6b the calculated fugacity coefficients of R32 agree well at the R134a-rich end, and they disagree by about 3% for pure R32. In these calculations for the Peng–Robinson calculations we used a value $k_{12} = 0$ in agreement with the results of Ref. 16.

5. SUMMARY

We have applied a previously developed model for the virial coefficients to new data on five binary refrigerant mixtures. Data on most of these systems have not been published previously. The agreement was found to be satisfactory, and the model can be used to greatly extend the range of usefulness of the rather limited data set. The low-temperature extrapolation of the model for B has been found to be accurate in the few cases where reliable experimental data are available for comparison. The extrapolation of the third virial coefficient to low temperatures is less certain, but the model provides a rational way to calculate C in a region where even the best data have large uncertainties. In any event, at reduced temperatures less than about 0.7, C has limited effect on the calculations for density and fugacity. With the binary interaction parameters determined here the model should be useful for calculations on several binary and multicomponent mixtures of potential use to the refrigeration industry.

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