data are in the opposite direction for this system from the deviations seen for CO_2 + 2,2-dimethylpropane (*3*).

List of Symbols

a,b	coefficients in cubic equation of state
a_{ij},b_{ij}	coefficients in quadratic mixing rules for equation of state
j_{ij}	interaction parameter describing deviations from the linear combining rule for covolumes in the SRK equation
k_{ij}	interaction parameter describing deviations from the geometric mean combining rule for the unlike pair attraction parameter in cubic equations
K_i	$\equiv y_i/x_i$
p	pressure
R	molar gas constant
rms(x,y)	root mean square deviation between calculated and experimental mole fractions on one isotherm
T	absolute temperature
V	molar volume
x_i	liquid mole fraction of component i
y_i	vapor mole fraction of component i

Registry No. CO_2, 124-38-9; cyclopentane, 287-92-3.

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High-Pressure Vapor–Liquid Equilibrium of Some Binary Mixtures of Cyclopentane, Argon, Nitrogen, *n*-Butane, and Neopentane

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Accurate high-pressure vapor–liquid equilibrium and phase density data for binary systems of cyclopentane with carbon dioxide, nitrogen, and argon and of argon also with neopentane and *n*-butane are reported, each at two isotherms. These data are correlated with the Peng–Robinson equation of state using one- and two-parameter versions of the van der Waals one-fluid mixing rules with both generalized and fluid-specific equation of state parameters.

Introduction

This study is the continuation of a research program on the measurement of vapor–liquid equilibrium data for mixtures consisting of hydrocarbons and inorganic gases. A set of mixtures has been chosen for this purpose based on their relative interaction energies (as reflected in the values of the critical temperatures T_c) and on molecular size (based on values of critical molar volumes V_c). The mixtures consist of the inorganic gases Ar, N_2, and CO_2 with the hydrocarbons *n*-butane, cyclopentane, and neopentane. The experimental data we obtain consist of pressure P, temperature T, liquid x_i, and vapor y_i, equilibrium mole fractions, and phase densities ρ, along two isotherms. Further we use these data to test the correlative ability of a simple cubic equation of state with various mixing rules. In this work we use the P–T–x–y data to determine the optimal value of the binary equation of state interaction parameters, and then we predict the phase densities, which are compared with our measured values. This is an additional test of the accuracy of the equation of state and its mixing rules. Also, we compare the results of using both the generalized equation of state parameters and temperature-dependent parameters determined specifically for each fluid.

Equipment and Experiment

The apparatus used for the experiments was described by Shibata (*1*) and Shibata and Sandler (*2*). It consists of a visual equilibrium cell, recirculation pumps (vapor and liquid), and vibrating-tube density meters. The circulating vapor is bubbled into the bottom of the cell, and the liquid is sprayed from the top, which ensures good vapor–liquid contact in the equilibrium cell; both phases are also circulated through the density meters. The entire system is enclosed in a constant-temperature air oven. System temperatures are monitored by five thermocouples located in the cell with an accuracy of ±0.05 K. The pressure is measured with a dead weight tester coupled to a null differential pressure indicator with an accuracy of ±0.14 bar at the high pressures measured here. Small separate samples of the equilibrated gas and liquid mixtures are drawn into evacuated and preheated gas bombs that contain motor-driven mixers. The samples are analyzed with a gas chromatograph. The chromatograph, other ancillary equipment, and the calibration procedures used have all been described previously (*2*). The sampling procedures used usually lead to area fractions in the three sample injections with a standard deviation of 0.003 in mole fraction.

Results and Discussion

The experimental data obtained in this work are reported in Table I. These data have been correlated with the Peng–Robinson equation of state (*3*)

$$P = \frac{RT}{V-b} - \frac{a}{V(V+b)+b(V-b)} \qquad (1)$$

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Table I. Vapor–Liquid Equilibrium Data Measured in This Work: Pressure P, Liquid-Phase Mole Fraction x_1, Vapor-Phase Mole Fraction y_1, Liquid Density ρ_L, and Vapor Density ρ_V

P/bar	x_1	y_1	ρ_L/(g cm^{-3})	ρ_V/(g cm^{-3})	P/bar	x_1	y_1	ρ_L/(g cm^{-3})	ρ_V/(g cm^{-3})
Argon (1) + Neopentane (2) at 367.3 K									
9.97	0.000	0.000	0.4942	0.0312	102.97	0.281	0.778	0.4867	0.1945
22.18	0.051	0.442	0.4946	0.0489	120.16	0.332	0.786	0.4778	0.2293
35.54	0.070	0.606	0.4952	0.0703	131.19	0.367	0.753	0.4686	0.2575
62.83	0.146	0.726	0.4941	0.1159	147.04	0.449	0.702	0.4404	0.3111
87.67	0.229	0.764	0.4912	0.1635	151.38	0.516	0.686	0.4162	0.3358
Argon (1) + Neopentane (2) at 411.1 K									
21.93	0.000	0.000	0.4036	0.0807	47.46	0.092	0.294	0.3889	0.1385
29.20	0.034	0.126	0.4032	0.0990	55.73	0.123	0.336	0.3782	0.1583
39.68	0.060	0.241	0.3966	0.1207	74.61	0.332	0.368	0.3015	0.2520
Argon (1) + *n*-Butane (2) at 366.2 K									
13.42	0.000	0.000	0.4821	0.0373	97.63	0.229	0.697	0.4788	0.1853
22.10	0.026	0.309	0.4828	0.0518	109.76	0.273	0.687	0.4701	0.2079
29.67	0.050	0.419	0.4833	0.0641	125.19	0.320	0.668	0.4635	0.2483
44.16	0.095	0.543	0.4838	0.0879	134.01	0.360	0.657	0.4544	0.2714
53.94	0.111	0.588	0.4839	0.1028	142.84	0.411	0.638	0.4403	0.3026
70.96	0.157	0.656	0.4830	0.1329	146.58	0.564	0.586	0.3486	0.3485
84.05	0.191	0.675	0.4816	0.1584					
Argon (1) + Cyclopentane (2) at 366.5 K									
3.58	0.000	0.000	0.6694	0.0120	179.91	0.257	0.917	0.6904	0.2678
26.38	0.039	0.847	0.6735	0.0434	207.06	0.304	0.912	0.6926	0.3081
42.09	0.058	0.882	0.6755	0.0658	234.90	0.359	0.904	0.6944	0.3504
69.65	0.092	0.910	0.6788	0.1050	262.46	0.400	0.900	0.6955	0.3927
97.08	0.137	0.923	0.6820	0.1449	285.76	0.454	0.890	0.6947	0.4306
122.99	0.178	0.922	0.6849	0.1908	309.67	0.506	0.880	0.6942	0.4682
152.34	0.227	0.920	0.6878	0.2293					
Argon (1) + Cyclopentane (2) at 410.4 K									
9.21	0.000	0.000	0.6110	0.0248	180.40	0.266	0.818	0.6122	0.2672
22.38	0.024	0.510	0.6175	0.0418	207.06	0.317	0.812	0.6080	0.3094
43.13	0.053	0.700	0.6134	0.0702	233.73	0.401	0.791	0.5989	0.3557
68.83	0.096	0.762	0.6143	0.1054	247.23	0.437	0.779	0.5926	0.3822
95.78	0.129	0.818	0.6151	0.1430	261.21	0.474	0.763	0.5760	0.4207
123.20	0.183	0.825	0.6152	0.1818	275.70	0.548	0.716	0.5402	0.4628
152.27	0.222	0.828	0.6144	0.2245					
Carbon Dioxide (1) + Cyclopentane (2) at 366.8 K									
3.66	0.000	0.000	0.6689	0.0120	70.41	0.364	0.872	0.6675	0.1550
10.25	0.044	0.587	0.6706	0.0220	80.13	0.434	0.875	0.6615	0.1830
17.56	0.070	0.733	0.6713	0.0387	90.46	0.510	0.872	0.6515	0.2189
27.27	0.118	0.811	0.6722	0.0560	100.39	0.606	0.852	0.6326	0.2681
37.82	0.175	0.847	0.6725	0.0736	107.11	0.657	0.849	0.6100	0.3126
49.12	0.218	0.864	0.6718	0.0980	112.17	0.735	0.825	0.5675	0.3670
59.11	0.271	0.871	0.6709	0.1211					
Carbon Dioxide (1) + Cyclopentane (2) at 412.1 K									
9.21	0.000	0.000	0.6110	0.0249	70.34	0.240	0.740	0.5938	0.1453
19.76	0.033	0.430	0.6105	0.0474	80.27	0.280	0.756	0.5875	0.1689
25.48	0.054	0.531	0.6098	0.0542	86.95	0.308	0.760	0.5794	0.1880
30.72	0.076	0.602	0.6084	0.0636	95.91	0.351	0.764	0.5695	0.2178
42.30	0.115	0.674	0.6055	0.0837	104.80	0.419	0.758	0.5535	0.2512
49.74	0.140	0.695	0.6034	0.0989	113.83	0.492	0.709	0.5103	0.3319
56.22	0.181	0.719	0.6002	0.1106	119.96	0.584	0.690	0.4356	0.3705
61.04	0.196	0.728	0.5990	0.1238					
Nitrogen (1) + Cyclopentane (2) at 366.4 K									
3.58	0.000	0.000	0.6694	0.0120	112.24	0.106	0.903	0.6644	0.1212
13.63	0.021	0.713	0.6693	0.0232	124.09	0.140	0.901	0.6644	0.1324
21.69	0.026	0.791	0.6689	0.0311	136.65	0.157	0.902	0.6629	0.1597
28.79	0.028	0.810	0.6688	0.0385	178.35	0.193	0.901	0.6613	0.1867
42.71	0.040	0.855	0.6681	0.0525	202.93	0.220	0.897	0.6596	0.2106
55.32	0.051	0.872	0.6674	0.0651	235.11	0.261	0.895	0.6569	0.2371
70.20	0.067	0.886	0.6663	0.0722	261.77	0.287	0.890	0.6555	0.2611
83.91	0.092	0.896	0.6657	0.0925	289.75	0.321	0.883	0.6530	0.2860
99.28	0.097	0.896	0.6649	0.1081	312.83	0.338	0.882	0.6510	0.3061
Nitrogen (1) + Cyclopentane (2) at 410.2 K									
9.21	0.000	0.000	0.6113	0.0249	207.81	0.235	0.812	0.5810	0.2199
29.55	0.025	0.601	0.6092	0.0447	234.89	0.275	0.807	0.5741	0.2483
42.64	0.041	0.689	0.6075	0.0569	261.77	0.302	0.792	0.5649	0.2776
70.00	0.069	0.764	0.6041	0.0832	274.73	0.343	0.778	0.5603	0.2934
97.08	0.097	0.797	0.6004	0.1096	290.92	0.355	0.776	0.5520	0.3080
122.43	0.130	0.814	0.5968	0.1360	296.64	0.366	0.771	0.5496	0.3197
153.30	0.159	0.821	0.5919	0.1651	304.15	0.374	0.770	0.5420	0.3296
180.45	0.193	0.817	0.5871	0.1922					

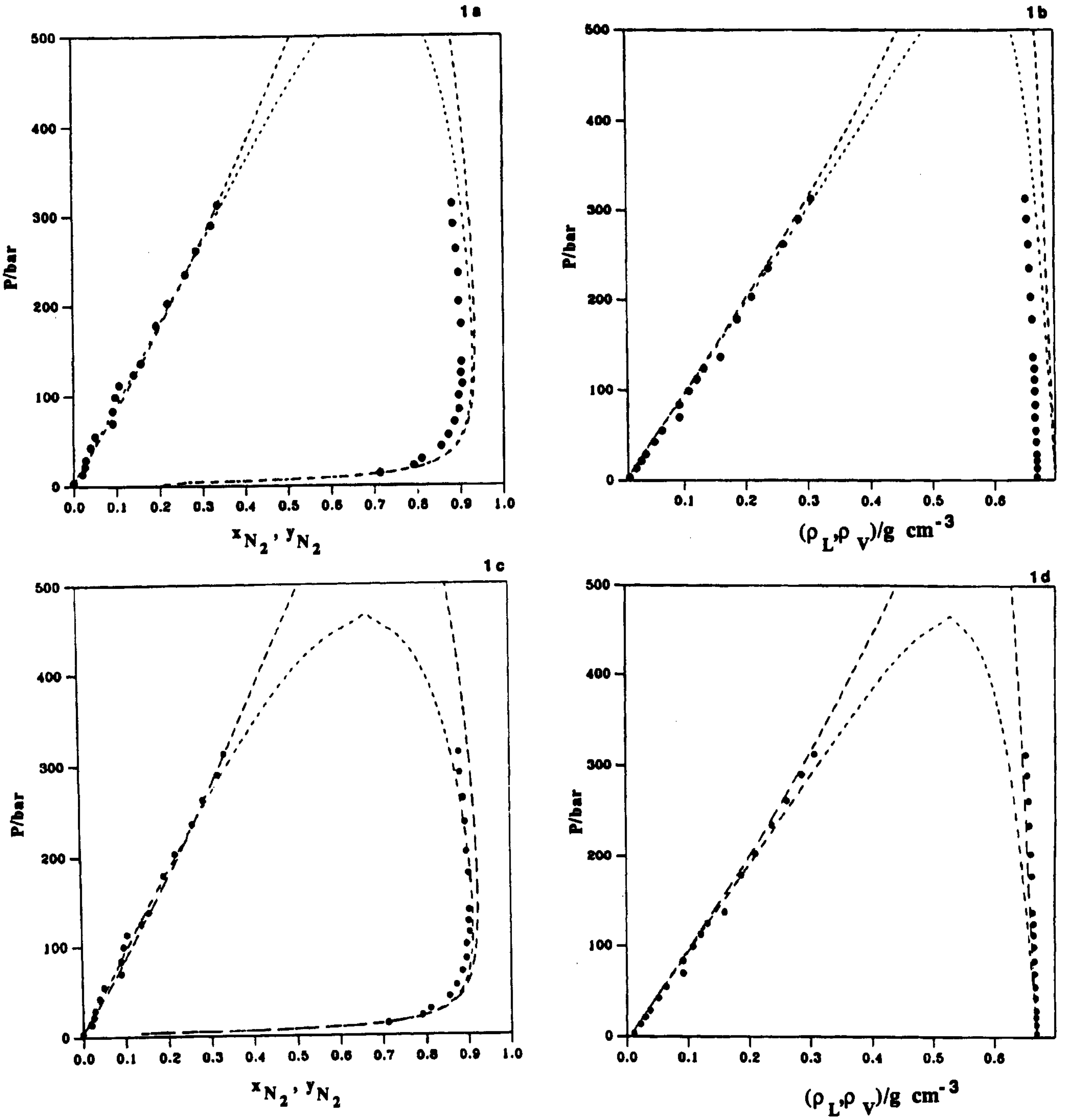

Figure 1. Experimental data and correlations for the nitrogen-cyclopentane system at 366.4 K. Parts a and c show P-x-y data, and parts b and d show pressure P and density ρ data. Points are experimental data; lines — — and - - are calculated from the Peng-Robinson equation of state with one and two adjustable parameters, respectively. The correlations in parts a and b are obtained by using the generalized equation of state parameters and in parts c and d by using the fluid-specific parameters.

where a and b are the parameters of the equation of state, V is the molar volume, and R is the molar gas constant. The generalized parameters for this equation of state are (3)

$$b(T) = b(T_c) = 0.07780RT_c/P_c \tag{2a}$$

$$a(T) = \alpha(T)\, a(T_c) = 0.45724R^2T_c^2\alpha(T)/P_c^2 \tag{2b}$$

$$\alpha(T) = [1 + \kappa(1 - T_r^{1/2})]^2 \tag{2c}$$

$$\kappa = 0.37464 + 1.54226\omega - 0.26992\omega^2 \tag{2d}$$

where P_c is the critical pressure, $T_r = T/T_c$, the reduced temperature, and ω the acentric factor.

The fluid-specific a and b parameters for cyclopentane and neopentane were found by fitting the equation of state with the following expressions

$$a(T) = \psi_a R^2T_c^2/P_c \tag{3a}$$

and

$$b(T) = \psi_b RT_c/P_c \tag{3b}$$

where

$$\psi_a = \sum_{i=0}^{2} a_i T_r^{\,i} \tag{4a}$$

and

$$\psi_b = \sum_{i=0}^{2} b_i T_r^{\,i} \tag{4b}$$

Table II. Parameters a_i and b_i in Equations 4a and 4b for Cyclopentane and Neopentane[a]

	a_0	a_1	a_2
cyclopentane	0.894 746 352	−0.547 657 011	0.133 775 278
neopentane	0.970 495 711	−0.617 222 465	0.077 897 712
	b_0	b_1	b_2
cyclopentane	0.067 131 293	0.040 835 730	−0.030 995 670
neopentane	0.061 361 188	0.067 882 317	−0.057 542 925

[a] Note that the parameters are unitless.

to vapor pressure and liquid density data in the temperature range of 330–415 K. The coefficients a_i and b_i were obtained by using the procedure of Wu and Sandler (*4*). The vapor pressure and liquid density data used for neopentane were reported by Dawson et al. (*5*), and the vapor pressure data for cyclopentane were from Willingham et al. (*6*). The saturated liquid density ρ_L data at three temperatures obtained from our experiments were correlated by using the expression of Spencer and Danner (*7*)

$$\rho_L = P_c R T_c Z_{RA}[1 + (1 - T_r)^{2/7}] \qquad (5)$$

where Z_{RA} is a constant determined from experimental data. Densities from this correlation were then used to obtain the equation of state parameters for cyclopentane. The a_i and b_i values for cyclopentane and neopentane calculated in this way are given in Table II. The fluid-specific parameters for the other fluids of interest here are reported by Xu and Sandler (*8*).

The equation of state parameters for mixture a_m and b_m are related to the pure-component parameters with mixing and combining rules. The mixing and combining rules used here are as follows:

$$a_m = \sum_1^2 \sum_1^2 x_i x_j a_{ij} \qquad (6a)$$

and

$$b_m = \sum_1^2 \sum_1^2 x_i x_j b_{ij} \qquad (6b)$$

where

$$a_{ij} = (a_i a_j)^{1/2}(1 - k_{ij}) \qquad (7a)$$

and

$$b_{ij} = (b_i + b_j)(1 - d_{ij})/2 \qquad (7b)$$

Since the experimental errors in our vapor and liquid compositions are approximately equal and, though small, are larger than in temperature and pressure, we used an isothermal flash routine in the data fitting to obtain the binary interaction parameters k_{ij} and d_{ij}. In this calculation temperature and pres-

Table III. Average Deviations Δ between Calculated and Experimental Values of Liquid and Vapor Mole Fractions, x and y, and densities, ρ_L and ρ_V, at Saturation

parameter values					
k_{ij}	d_{ij}	Δx	$\Delta\rho_L$/(g cm^{-3})	Δy	$\Delta\rho_V$/(g cm^{-3})
		Argon + Neopentane at 367.3 K With Generalized Parameters			
0.091		0.023	0.0492	0.028	0.0377
0.05	−0.09	0.010	0.0175	0.022	0.0231
		With Fluid-Specific Parameters			
0.11		0.025	0.0273	0.030	0.0394
0.07	−0.09	0.010	0.0142	0.024	0.0251
		Argon + Neopentane at 411.1 K With Generalized Parameters			
−0.083		0.038	0.0736	0.032	0.0201
−0.03	−0.28	0.021	0.0640	0.013	0.0409
		With Fluid-Specific Parameters			
−0.131		0.042	0.0222	0.025	0.0306
−0.06	−0.22	0.022	0.0202	0.010	0.0243
		Argon + *n*-Butane at 366.2 K With Generalized Parameters			
0.094		0.025	0.0283	0.028	0.0457
0.07	−0.09	0.019	0.0406	0.014	0.0364
		With Fluid-Specific Parameters			
0.105		0.024	0.0309	0.025	0.0420
0.08	−0.07	0.017	0.0441	0.014	0.0321
		Argon + Cyclopentane at 366.5 K With Generalized Parameters			
0.069		0.017	0.0471	0.011	0.0207
0.05	−0.02	0.013	0.0402	0.007	0.0199
		With Fluid-Specific Parameters			
0.0967		0.017	0.0057	0.005	0.0216
0.07	−0.03	0.012	0.0081	0.004	0.0193
		Argon + Cyclopentane at 410.4 K With Generalized Parameters			
0.052		0.029	0.0503	0.028	0.0249
0.00	−0.092	0.010	0.0191	0.011	0.0148
		With Fluid-Specific Parameters			
0.065		0.028	0.0204	0.025	0.0294
0.02	−0.08	0.011	0.0121	0.013	0.0168
		Carbon Dioxide + Cyclopentane at 366.8 K With Generalized Parameters			
0.119		0.026	0.0328	0.034	0.0476
0.10	−0.06	0.007	0.0372	0.022	0.0632
		With Fluid-Specific Parameters			
0.14		0.021	0.0201	0.020	0.0546
0.13	−0.03	0.009	0.0426	0.017	0.0691
		Carbon Dioxide + Cyclopentane at 412.1 K With Generalized Parameters			
0.175		0.014	0.0205	0.024	0.0845
0.06	−0.04	0.008	0.0213	0.015	0.0542
		With Fluid-Specific Parameters			
0.184		0.020	0.0201	0.029	0.0884
0.14	−0.08	0.007	0.0387	0.021	0.0463
		Nitrogen + Cyclopentane at 366.4 K With Generalized Parameters			
0.01		0.008	0.0443	0.032	0.0609
−0.06	−0.04	0.006	0.0369	0.026	0.0627
		With Fluid-Specific Parameters			
0.02		0.008	0.0060	0.021	0.0551
−0.1	−0.07	0.007	0.0166	0.012	0.0658
		Nitrogen + Cyclopentane at 410.2 K With Generalized Parameters			
0.047		0.014	0.0614	0.047	0.0177
−0.13	−0.13	0.004	0.0295	0.009	0.0227
		With Fluid-Specific Parameters			
0.03		0.015	0.0248	0.036	0.0255
−0.08	−0.09	0.006	0.0042	0.008	0.0104

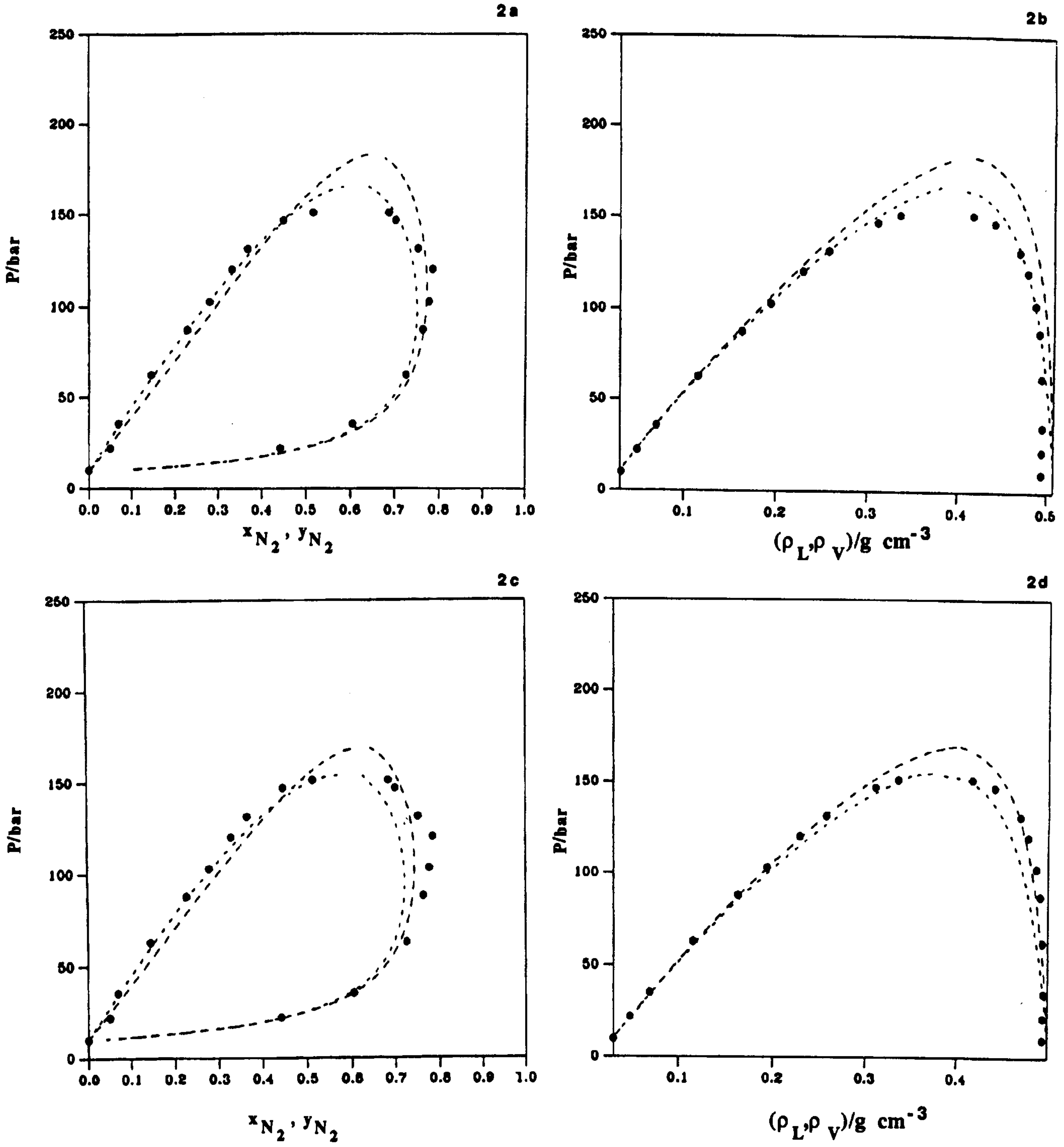

Figure 2. Experimental data and correlations for the argon–neopentane system at 367.3 K. Parts a and c show P–x–y data, and parts b and d show pressure P and density ρ data. Points are experimental data; lines -- and -- are calculated from the Peng–Robinson equation of state with one and two adjustable parameters, respectively. The correlations in parts a and b are obtained by using the generalized equation of state parameters and in parts c and d by using the fluid-specific parameters.

sure were taken as the independent variables, and the following objective function OBJ was used in the optimization procedure

$$\mathrm{OBJ} = \sum_{1}^{N}\sum_{1}^{2}(|x_j^{\mathrm{calcd}} - x_j^{\mathrm{expt}}| + |y_j^{\mathrm{calcd}} - y_j^{\mathrm{expt}}|) \qquad (8)$$

where N is the number of data points along an isotherm. Optimum fits for the parameters at each isotherm were determined by minimizing OBJ for the N data points. A simple minimization procedure was used when only k_{ij} was to be found; a grid search technique was used in the minimization procedure when both k_{ij} and d_{ij} were being fit. The results of the fitting appear in Table III. Note that only composition data, not our measured density data, were used in the optimization. In terms of the average derivations $\Delta\rho$, Δx, and Δy are given in Table III.

Discussion

As can be seen from Table III, the fit of the experimental data when the fluid-specific temperature-dependent pure component parameters are used is better than that obtained when the generalized parameters are used. This is not surprising since the temperature-dependent fluid-specific parameters were developed to give more accurate pure-component vapor pressures and densities than the generalized parameters. Also, in general, the use of two binary interaction parameters leads to better correlations than the use of only one parameter, though sometimes the improvement is small.

Now turning to specific systems we see that the data for the argon–cyclopentane mixture are correlated poorly compared to the data correlation of mixtures of cyclopentane with CO_2 or N_2. The nitrogen + cyclopentane experimental data at 366.4 K and the correlations are shown in Figure 1, where the difference between using the generalized and fluid-specific equation of state parameters is clearly seen, especially in the liquid densities. The molecular sizes (or V_c values) of CO_2 and N_2 are close and larger than that of the Ar molecule. Thus the mixture of cyclopentane and Ar has a large size difference, as well as a difference in molecular shape, and on the basis of the value of the d_{ij} parameter, the simple linear mixing rule (i.e., $d_{ij} = 0$) for the b parameter is not adequate for this system. In contrast, the size ratios in the cyclopentane–carbon dioxide and cyclopentane–nitrogen mixtures are smaller, and the linear mixing rule for the b parameter ($d_{ij} = 0$) is satisfactory.

To further examine the b_{ij} parameter mixing rule, two isotherms for the argon + neopentane mixture and one isotherm for the argon + n-butane mixture were also measured. The results, both experimental and correlated, for the 367.3 K isotherm of the argon + neopentane system are shown in Figure 2. Both Ar and neopentane can be considered to be approximately spherical, but there is a large difference in their molecular sizes. On comparing the correlated data for the argon + n-butane and argon + neopentane binary mixtures, we see both data sets are well correlated with similar k_{ij} parameter values when using the van der Waals one-parameter mixing rule, as might be expected since the critical temperatures (and thus interaction energies) of n-butane and neopentane are very close. Somewhat surprising is that the error between the calculated and experimental values is not reduced substantially with the introduction of the second adjustable interaction parameter d_{ij}, even though the largest size difference in the mixtures we have studied occurs here. This suggests that it is predominantly the differences in molecular shape that necessitate the use of a binary adjustable parameter in the b_{ij} parameter mixing rule, rather than merely a difference in sizes of the molecules.

Overall, the differences between the calculated and measured values of the liquid densities when the generalized parameters in the Peng–Robinson equation of state are used are 7–9%, compared to 2 to 4% in the vapor densities. However, the liquid density estimates are greatly improved by using the fluid-specific parameters that reduce the errors between the calculated and experimental values to 2–4%.

Conclusions

We have measured the high-pressure vapor–liquid equilibria for binary mixtures of N_2, CO_2, and Ar separately with cyclopentane and of Ar separately with neopentane and n-butane. We report the compositions and densities of the coexisting phases.

We have correlated our data with the Peng–Robinson equation of state, using both the original, generalized parameters and temperature-dependent parameters specific to each fluid. The fluid-specific parameters generally lead to a better correlation of the experimental data. Also, much more accurate densities of the liquid phase are predicted with these parameters.

Finally we considered the use of the van der Waals one-fluid mixing rules for the equation of state parameters with one binary parameter (in the a parameter combining rule) and two binary parameters (one in each of the a and b parameter combining rules). We find that, for the mixtures studied, only when both the size and shape of the molecules are very different are two binary parameters needed. For mixtures containing similar size molecules, or even for spherical molecules of widely differing size, the data are well correlated with only a single binary parameter.

Glossary

a	equation of state parameters, bar m^6 mol^{-2}
b	equation of state parameters, m^3 mol^{-1}
d	interaction parameter, eq 7b
k	interaction parameter, eq 7a
P	pressure, bar
R	molar gas constant, 8.31451×10^{-5} bar m^3 K^{-1} mol^{-1}
T	temperature, K
V	molar volume, m^3 mol^{-1}
x	liquid-phase mole fraction

Greek Letters

y	vapor-phase mole fraction
Δ	average deviation

Subscripts

ρ	density at saturation, g cm^{-3}
c	critical property
i,j	component
L	liquid
m	mixture property

Superscripts

V	vapor
calcd	calculated
expt	experimental

Registry No. CO_2, 124-38-9; N_2, 7727-37-9; Ar, 7440-37-1; cyclopentane, 287-92-3; neopentane, 463-82-1; n-butane, 106-97-8.

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