$\Delta\beta$	average error in solute distribution ratio
γ	activity coefficient

Subscripts

i, j	components i and j
j	phase j, eq 1 and 2
k	tie line k
n	1, 2, ..., NP (parameters, eq 1)
s	solute
∞	infinite dilution

Superscripts

$\hat{}$	calculated values
I, II	phases I and II, eq 1

Registry No. Acetic acid, 64-19-7; methyl isopropyl ketone, 563-80-4.

Literature Cited

(1) Renon, H.; Prausnitz, J. M. *AIChE J.* **1968**, *14*, 135–144.
(2) Abrams, D. S.; Prausnitz, J. M. *AIChE J.* **1975**, *21*, 116–128.
(3) Fredenslund, Aa.; Jones, R. L.; Prausnitz, J. M. *AIChE J.* **1975**, *21*, 1086–1099.
(4) Magnussen, T.; Rasmussen, P.; Fredenslund, Aa. *Ind. Eng. Chem. Process Des. Dev.* **1981**, *20*, 331–339.
(5) Othmer, D. F.; White, R. E.; Trueger, E. *Ind. Eng. Chem.* **1941**, *33*, 1240–1248.
(6) Correa, J. M. Determinación, correlación y predicción del equilibrio líquido-líquido. Sistemas Agua + Acido acético + Cetonas. Doctoral Thesis, Universidad de Santiago de Compostela, Spain, 1985.
(7) Gross, P. M.; Rintelen, J. C.; Saylor, J. M. *J. Phys. Chem.* **1939**, *43*, 197–205.
(8) Ginnings, P. M.; Plonk, D.; Carter, E. *J. Am. Chem. Soc.* **1940**, *62*, 1923–1924.
(9) Sørensen, J. M. Correlation of liquid-liquid equilibrium data. Ph.D. Thesis, Technical University of Denmark, Lyngby, Denmark, 1980.
(10) Correa, J. M.; Arce, A.; Blanco, A.; Correa, A. *Fluid Phase Equilib.* **1987**, *32*, 151–162.

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High-Pressure Vapor–Liquid Equilibria of Mixtures of Nitrogen, Carbon Dioxide, and Cyclohexane

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Data are reported for the equilibrium phase compositions and densities of the carbon dioxide + cyclohexane and the nitrogen + cyclohexane systems at 366.5 and 410.9 K and for the nitrogen + carbon dioxide + cyclohexane system at the same temperatures at a variety of isobars. These data were modeled with the Peng–Robinson equation of state with generalized parameters. Good fits were obtained for the compositions in all three systems, but the predicted liquid-phase densities were in error by about 5%, as expected.

Introduction

The motivation for this research is to obtain experimental data on systems consisting of hydrocarbons and carbon dioxide, nitrogen, and other non-hydrocarbons, and to utilize these data in evaluating equations of state and their mixing rules. By determining both the compositions and the densities of the coexisting phases, equations of states and their mixing rules can be more stringently tested than with composition data alone.

The equipment used in these measurements is the double-recirculation cell with a maximum temperature of 300 °F and maximum pressure of 5000 psia described earlier (*1*). The key components of the apparatus are a 100-cm^3 through-windowed visual cell, recirculation pumps, and vibrating-tube density meters. The operating procedure for this equipment has also been described earlier (*1*) and will not be repeated here.

Experimental Results

Table I contains our unsmoothed, experimental data for the CO_2 + cyclohexane system at 366.5 and 410.9 K. Data for this system, at different temperatures, have been reported previously by Krichevskii and Sorina (*2*). Figures 1 and 2 are plots of composition and density for both data sets. Since the temperatures of the data sets are different, the data cannot be directly compared.

Table I. Vapor–Liquid Equilibrium Data for the Carbon Dioxide + Cyclohexane Binary System

T, K	P, bar	x_{CO_2}	y_{CO_2}	ρ_L, g/cm^3	ρ_V, g/cm^3
366.5	1.71	0.0000	0.0000	0.7050	0.0077
366.4	17.52	0.0959	0.8984	0.7059	0.0288
366.4	34.69	0.1681	0.9374	0.7071	0.0593
366.5	51.91	0.2657	0.9495	0.7074	0.0940
366.5	68.86	0.3484	0.9515	0.7060	0.1349
366.6	86.37	0.4598	0.9500	0.7019	0.1831
366.5	103.60	0.5698	0.9402	0.6894	0.2476
366.5	125.11	0.7526	0.8977	0.6222	0.3986
366.4	128.00	0.7901	0.8732	0.5807	0.4515
410.8	4.43	0.0000	0.0000	0.6566	0.0114
410.8	17.18	0.0671	0.7166	0.6584	0.0294
410.9	34.44	0.1470	0.8295	0.6544	0.0570
410.9	51.51	0.2050	0.8593	0.6553	0.0866
410.8	69.28	0.2887	0.8850	0.6488	0.1198
410.9	86.51	0.3578	0.8861	0.6434	0.1614
410.9	104.02	0.4329	0.8777	0.6312	0.2046
410.9	120.56	0.5165	0.8636	0.6151	0.2579
410.9	138.14	0.6107	0.8320	0.5733	0.3434
410.8	144.41	0.6850	0.7846	0.5232	0.4115
410.9	145.10	0.7448	0.7671	0.4893	0.4321

The data for the N_2 + cyclohexane system are tabulated in Table II. Two isotherms, 366.5 and 410.9 K, were measured for this system. The previously measured bubble-point data of Khodeeva (*3*) are plotted and compared with our experimental data in Figure 3, though the temperatures are not quite the same. Our density data are shown in Figure 4; there are no published density data for this system.

Tables III and IV contain our experimental, unsmoothed data for the N_2 + CO_2 + cyclohexane system at 366.5 and 410.9 K with data taken at four pressures, 68.9, 137.9, 206.8, and 275.7 bar, at both temperatures. No data for this system have been reported previously. Figures 5–8 are composition and density plots of these data sets.

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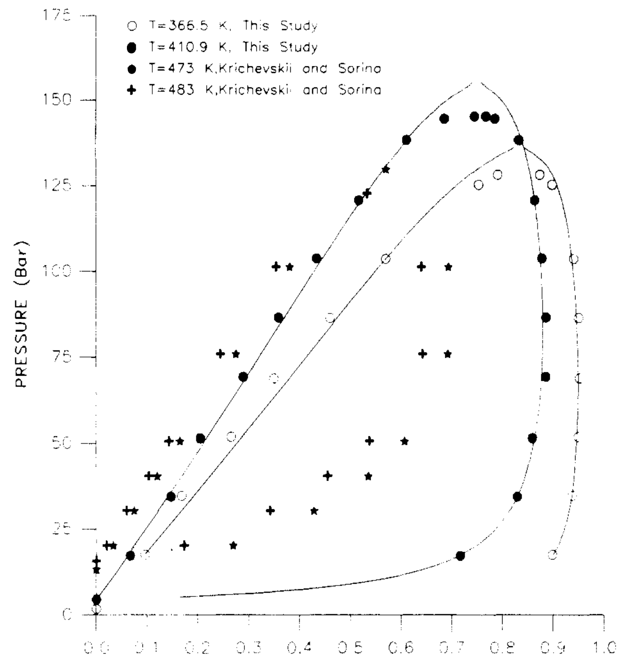

Figure 1. T–P–x–y data for carbon dioxide + cyclohexane. A comparison of experimental data from this study with that of Krichevskii and Sorina (*2*) at somewhat different temperatures. The data have been fit with the Peng–Robinson equation of state (solid lines).

Table II. Vapor–Liquid Equilibrium Data for the Nitrogen + Cyclohexane Binary System

T, K	P, bar	x_{N_2}	y_{N_2}	ρ_L, g/cm^3	ρ_V, g/cm^3
366.5	1.71	0.0000	0.0000	0.7050	0.0077
366.5	17.99	0.0094	0.9064	0.7034	0.0194
366.5	34.85	0.0337	0.9422	0.7032	0.0358
366.4	69.83	0.0678	0.9673	0.7031	0.0685
366.4	103.88	0.0985	0.9713	0.7024	0.1004
366.4	138.76	0.1286	0.9721	0.7017	0.1324
366.4	172.53	0.1571	0.9719	0.7017	0.1626
366.5	207.00	0.1802	0.9676	0.7015	0.1929
366.3	241.94	0.2088	0.9694	0.7011	0.2204
366.6	275.93	0.2438	0.9664	0.7015	0.2485
410.8	4.43	0.0000	0.0000	0.6566	0.0114
410.8	17.53	0.0169	0.7133	0.6606	0.0226
410.9	35.27	0.0343	0.8381	0.6612	0.0388
410.9	69.14	0.0721	0.9124	0.6606	0.0689
410.8	103.74	0.1116	0.9154	0.6594	0.0997
410.8	138.21	0.1478	0.9230	0.6574	0.1297
410.9	172.74	0.1821	0.9224	0.6542	0.1593
410.8	206.86	0.2161	0.9202	0.6522	0.1877
410.8	241.46	0.2552	0.9198	0.6494	0.2163
410.9	275.79	0.2906	0.9121	0.6473	0.2449

The data for the CO_2 + cyclohexane and N_2 + cyclohexane binary systems have been correlated with the Peng–Robinson equation of state (*4*) and the van der Waals one-fluid mixing rules

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

$$a = \sum_{i=1}^{n_c}\sum_{j=1}^{n_c} x_i x_j a_{ij} \tag{2}$$

$$b = \sum_{i=1}^{n_c} x_i b_i \tag{3}$$

and

$$a_{ij} = (a_i a_j)^{0.5}(1 - k_{ij}) \tag{4}$$

The fits of the CO_2 + cyclohexane compositional data with the Peng–Robinson EOS and van der Waals one fluid mixing rules are shown as the solid lines in Figures 1 and 2. One adjustable interaction parameter k_{ij} fitted to the T–P–x–y data is adequate for modeling the compositional data for this system, though the critical pressure is overpredicted. An isothermal flash routine was used in the fitting of the binary interaction parameter, since we feel the error in our vapor and liquid compositions to be equal, at about 0.005 mole fraction. The objective function used in the fitting was

$$\mathrm{OBJ} = \sum_{i=1}^{\mathrm{NPTS}}\sum_{n=1}^{n_c} (|x_i^{\mathrm{CALC}} - x_i^{\mathrm{EXP}}| + |y_i^{\mathrm{CALC}} - y_i^{\mathrm{EXP}}|) \tag{5}$$

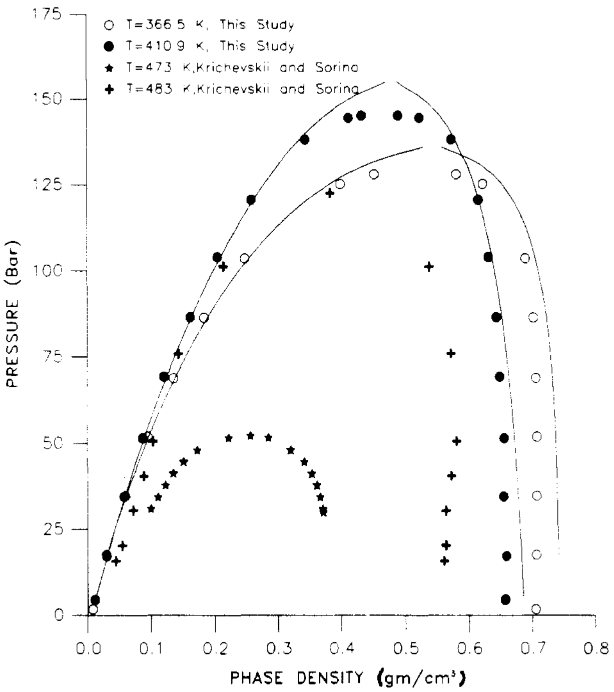

Figure 2. Phase density data for carbon dioxide + cyclohexane. A comparison of experimental data from this study with that of Krichevskii and Sorina (*2*) at somewhat different temperatures. The solid lines result from the Peng–Robinson equation of state with the binary parameter determined from fitting compositional data.

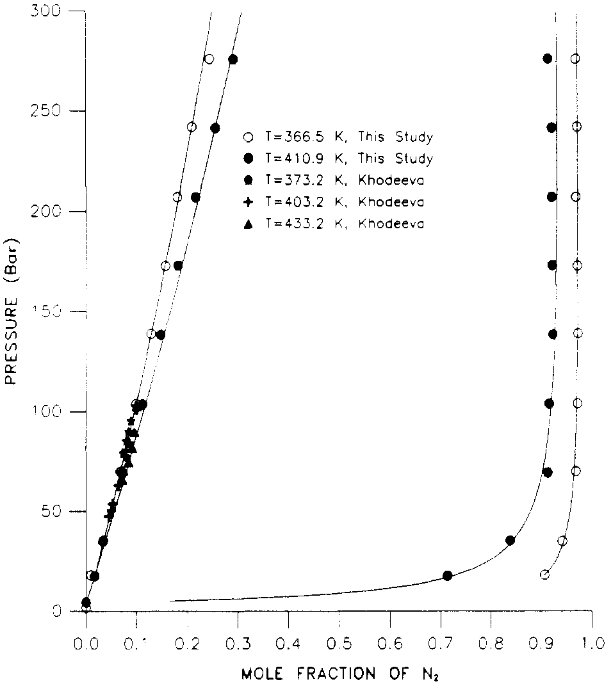

Figure 3. T–P–x–y data for nitrogen + cyclohexane. A comparison of experimental data from this study with that of Khodeeva (*3*) at somewhat different temperatures. The data have been fit with the Peng–Robinson equation of state and a single binary parameter.

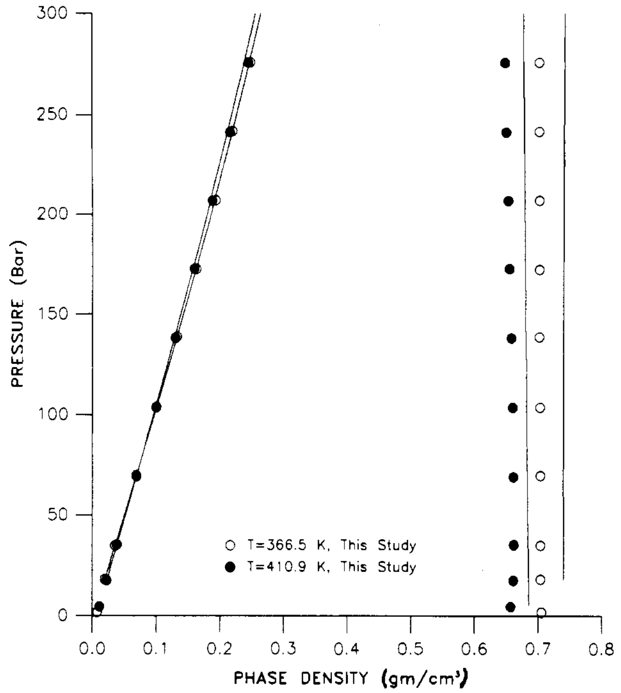

Figure 4. Phase density data for the nitrogen + cyclohexane system. The solid lines are the predictions of the Peng-Robinson equation of state with the single binary parameter fit to compositional data.

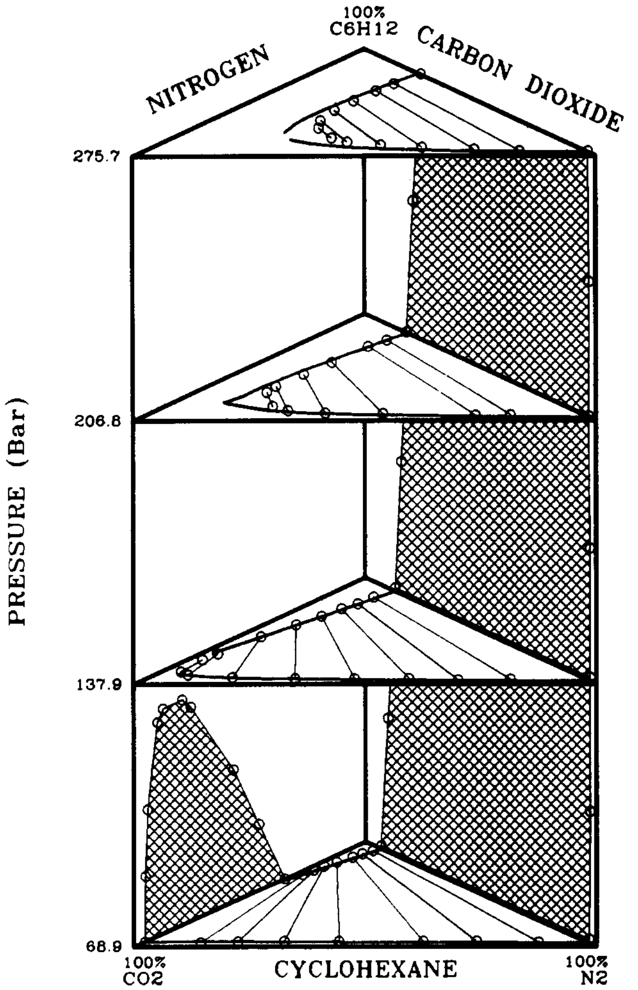

Figure 5. Phase behavior for the ternary system of nitrogen, carbon dioxide, and cyclohexane at T = 366.5 K and four pressures.

Optimum fits of the isotherms were determined by allowing the k_{ij} parameter to be temperature dependent (Table V). Predictions of the CO_2 + cyclohexane phase density data using the k_{ij} determined from fitting the T-P-x-y data are less satisfactory, as is expected. The differences between the experimental and calculated densities are most noticeable in the liquid and near the mixture critical point of each isotherm. [Note that the phase densities were not used in fitting the binary interaction parameter, nor would including them have improved the fit significantly.]

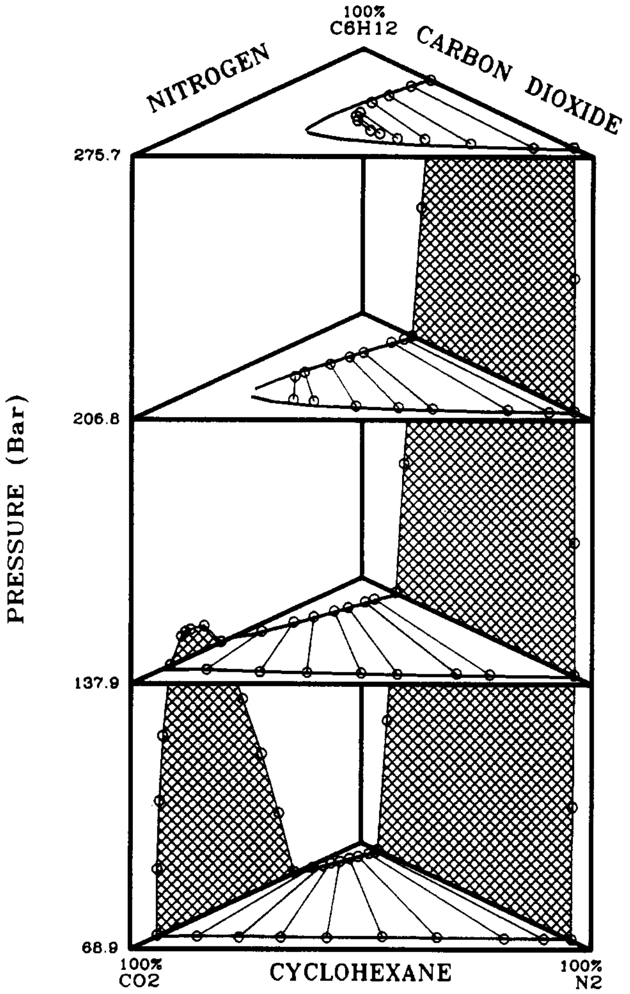

Figure 6. Phase behavior for the ternary system of nitrogen, carbon dioxide, and cyclohexane at T = 410.9 K and four pressures. The solid lines are the Peng-Robinson predictions based on binary data.

The fits of the N_2 + cyclohexane data with the Peng-Robinson EOS and van der Waals one-fluid mixing rules are shown in Figures 3 and 4. The fit of the compositional data is generally quite good up to the highest pressure (~276 bar) which is well below the critical point for this mixture at the temperatures studied. Again the liquid density predictions are in error by as much as 5%. In a subsequent paper we will discuss alternative modeling techniques to improve the density predictions.

The phase behavior of the ternary N_2 + CO_2 + cyclohexane system was predicted by using the binary parameters determined from our binary data, though setting k_{ij} for the N_2-CO_2 system equal to zero as reported elsewhere (*1*). The accuracy of the composition predictions for ternary mixtures depends on the fit of the binary systems (i.e., the N_2 + cyclohexane, CO_2 + cyclohexane, and N_2 + CO_2 systems). Since the fit of the N_2 + cyclohexane and CO_2 + cyclohexane binary systems with the Peng-Robinson equation of state, the van der Waals one-fluid mixing rules, and a single binary interaction parameter was quite good, the quality of the ternary predictions is satisfactory (Figures 5 and 6). However, similar to the fits of the binary data,

Table III. Vapor–Liquid Equilbrium Data for the Nitrogen + Carbon Dioxide + Cyclohexane Ternary System at 366.5 K (200 °F)

T, K	P, bar	x_{N_2}	x_{CO_2}	y_{N_2}	y_{CO_2}	ρ_L, g/cm^3	ρ_V, cm^3
366.4	69.83	0.0678	0.0000	0.9673	0.0000	0.7031	0.0685
366.5	69.00	0.0603	0.0284	0.8568	0.1048	0.7041	0.0729
366.5	69.00	0.0533	0.0675	0.7187	0.2386	0.7045	0.0792
366.5	69.00	0.0459	0.1021	0.6007	0.3514	0.7052	0.0856
366.4	69.14	0.0374	0.1621	0.4219	0.5351	0.7057	0.0971
366.4	69.28	0.0303	0.2100	0.3054	0.6562	0.7060	0.1055
366.5	68.86	0.0251	0.2486	0.2057	0.7559	0.7060	0.1134
366.3	68.86	0.0179	0.2894	0.1253	0.8349	0.7057	0.1201
366.5	68.86	0.0000	0.3484	0.0000	0.9515	0.7060	0.1349
366.4	138.76	0.1286	0.0000	0.9721	0.0000	0.7017	0.1324
366.4	138.20	0.1138	0.0776	0.7974	0.1678	0.7031	0.1499
366.5	137.79	0.1041	0.1360	0.6794	0.2788	0.7030	0.1644
366.5	138.07	0.0961	0.1981	0.5710	0.3830	0.7024	0.1804
366.4	137.93	0.0862	0.2750	0.4518	0.4975	0.7010	0.2019
366.4	137.93	0.0705	0.3697	0.3224	0.6265	0.6970	0.2333
366.5	138.07	0.0539	0.5041	0.1842	0.7572	0.6847	0.2877
366.5	137.79	0.0365	0.6722	0.0705	0.8357	0.6394	0.3856
366.5	137.93	0.0339	0.7358	0.0434	0.8387	0.5952	0.4449
366.5	207.00	0.1802	0.0000	0.9676	0.0000	0.7015	0.1929
366.5	206.86	0.1737	0.0784	0.7911	0.1594	0.7014	0.2150
366.6	206.86	0.1623	0.1481	0.7155	0.2383	0.7001	0.2384
366.5	206.86	0.1591	0.3010	0.5072	0.4289	0.6927	0.2956
366.6	206.58	0.1481	0.4124	0.3752	0.5463	0.6786	0.3542
366.5	206.58	0.1443	0.5287	0.2831	0.6155	0.6478	0.4256
366.3	207.00	0.1564	0.5802	0.2341	0.6320	0.6106	0.4821
366.6	275.93	0.2438	0.0000	0.9664	0.0000	0.7015	0.2485
366.5	275.65	0.2274	0.0987	0.8140	0.1413	0.6991	0.2823
366.4	275.79	0.2252	0.1736	0.7085	0.2341	0.6955	0.3151
366.5	275.65	0.2229	0.2677	0.5858	0.3378	0.6877	0.3577
366.5	275.79	0.2244	0.3525	0.4861	0.4153	0.6733	0.4082
366.4	275.65	0.2404	0.4287	0.3977	0.4705	0.6447	0.4698
366.5	275.65	0.2711	0.4687	0.3453	0.4862	0.6081	0.5202

Table IV. Vapor–Liquid Equilibrium Data for the Nitrogen + Carbon Dioxide + Cyclohexane Ternary System at 410.9 K (280 °F)

T, K	P, bar	x_{N_2}	x_{CO_2}	y_{N_2}	y_{CO_2}	ρ_L, g/cm^3	ρ_V, g/cm^3
410.9	69.14	0.0721	0.0000	0.9124	0.0000	0.6606	0.0689
410.8	69.00	0.0770	0.0144	0.8499	0.0569	0.6591	0.0703
410.9	68.86	0.0670	0.0327	0.7601	0.1385	0.6591	0.0743
410.8	68.86	0.0537	0.0704	0.6087	0.2776	0.6587	0.0804
410.8	69.00	0.0511	0.1017	0.4897	0.3958	0.6581	0.0870
410.8	68.86	0.0423	0.1333	0.3724	0.5194	0.6568	0.0932
410.8	69.00	0.0333	0.1635	0.2722	0.6194	0.6569	0.0999
410.8	69.00	0.0257	0.1930	0.1816	0.7120	0.6565	0.1059
410.9	69.07	0.0150	0.2239	0.0876	0.8009	0.6556	0.1132
410.8	69.28	0.0000	0.2887	0.0000	0.8850	0.6488	0.1198
410.8	138.21	0.1478	0.0000	0.9230	0.0000	0.6574	0.1297
410.8	138.07	0.1308	0.0758	0.7348	0.1796	0.6542	0.1471
410.8	138.07	0.1242	0.1094	0.6577	0.2469	0.6523	0.1555
410.9	137.93	0.1114	0.1704	0.5305	0.3748	0.6504	0.1712
410.8	137.93	0.0964	0.2161	0.4465	0.4478	0.6468	0.1845
410.9	137.93	0.0809	0.2889	0.3270	0.5636	0.6415	0.2068
410.9	138.00	0.0630	0.3574	0.2230	0.6654	0.6333	0.2315
410.8	138.07	0.0382	0.4719	0.0978	0.7723	0.6152	0.2769
410.9	138.14	0.0000	0.6107	0.0000	0.8320	0.5733	0.3434
410.8	206.86	0.2161	0.0000	0.9202	0.0000	0.6522	0.1877
410.9	206.65	0.2095	0.0285	0.8645	0.0551	0.6504	0.1962
410.9	207.00	0.2040	0.0783	0.7706	0.1405	0.6451	0.2177
410.9	206.72	0.1867	0.1812	0.5983	0.2932	0.6376	0.2458
410.9	206.65	0.1768	0.2319	0.5177	0.3597	0.6319	0.2658
410.9	206.58	0.1675	0.3061	0.4180	0.4449	0.6194	0.2984
410.9	207.14	0.1597	0.4099	0.3142	0.5248	0.5913	0.3542
410.8	207.00	0.1571	0.4480	0.2602	0.5586	0.5688	0.3870
410.9	275.79	0.2906	0.0000	0.9121	0.0000	0.6473	0.2449
410.9	275.93	0.2796	0.0738	0.8294	0.0938	0.6394	0.2722
410.9	275.93	0.2756	0.1648	0.6742	0.2109	0.6251	0.3116
410.9	276.20	0.2756	0.2398	0.5594	0.2929	0.6069	0.3534
410.9	275.51	0.2848	0.2989	0.4850	0.3378	0.5808	0.3960
410.8	275.93	0.2990	0.3306	0.4311	0.3605	0.5512	0.4292
410.9	276.07	0.3253	0.3508	0.3989	0.3686	0.5230	0.4630

the critical point at each pressure is overpredicted, as is common with calculations involving cubic equations of state. Figures 7 and 8 contain the density predictions. Once again, the densities in the liquid and near-critical regions are poorly predicted. It should be emphasized that in these ternary mixture calculations the values of the interaction parameters used were

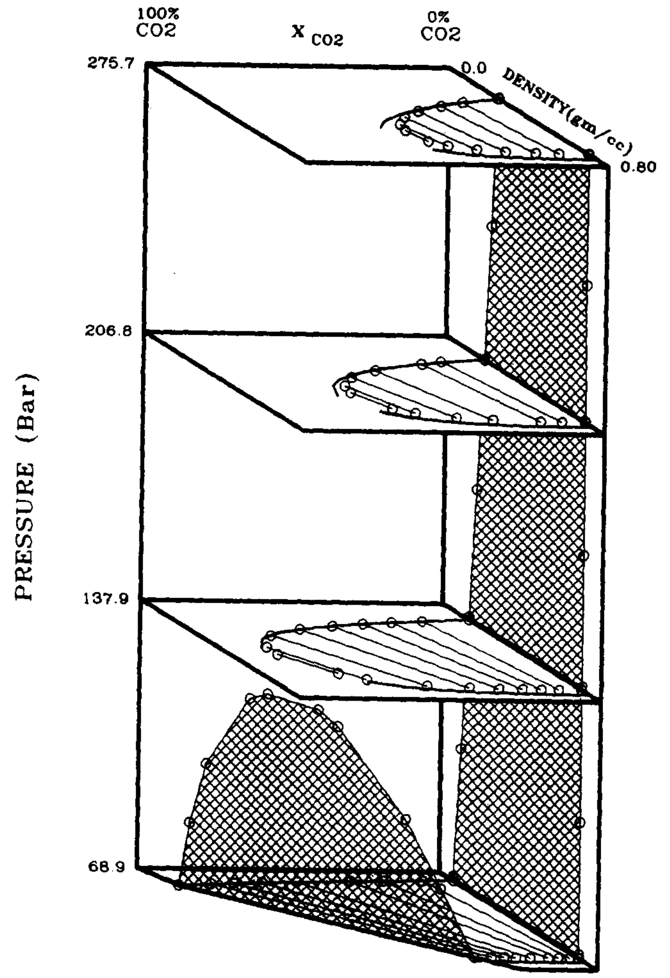

Figure 7. Three-dimensional view of phase density data for the ternary system of nitrogen, carbon dioxide, and cyclohexane at T = 366.5 K and three pressures. The crosshatched areas represent the binary two-phase regions.

Table V. Binary Interaction Parameters for the Peng–Robinson Equation of State

Carbon Dioxide + Cyclohexane Binary System	
T = 366.5 K	$k_{CO_2+C_6H_{12}} = 0.110$
T = 410.9 K	$k_{CO_2+C_6H_{12}} = 0.103$
Nitrogen + Cyclohexane Binary System	
T = 366.5 K	$k_{N_2+C_6H_{12}} = 0.109$
T = 410.9 K	$k_{N_2+C_6H_{12}} = 0.076$
Nitrogen + Carbon Dioxide + Cyclohexane Ternary System	
T = 366.5 K	$k_{CO_2+C_6H_{12}} = 0.110$
	$k_{N_2+C_6H_{12}} = 0.109$
	$k_{CO_2+N_2} = 0.000$
T = 410.9 K	$k_{CO_2+C_6H_{12}} = 0.103$
	$k_{N_2+C_6H_{12}} = 0.076$
	$k_{CO_2+N_2} = 0.000$

obtained from fitting the binary systems. The predictions of the ternary data involved no adjustment of the binary interaction parameters.

Conclusions

Data are reported for the compositions and densities of the coexisting vapor and liquid phases in equilibrium for binary and ternary mixtures of nitrogen, carbon dioxide, and cyclohexane at temperatures up to 411 K and pressures to 276 bar. The compositional data for the binary systems containing cyclohexane were fit well with the Peng–Robinson equation of state with van der Waals one-fluid mixing rules, though (as is expected) the liquid density predictions were less satisfactory. Since the compositional data for constituent binary systems were reasonably well fit with the Peng–Robinson equation of state, the composition data for the ternary mixture were satisfactorily predicted by using the equation of state parameters obtained from the binary data.

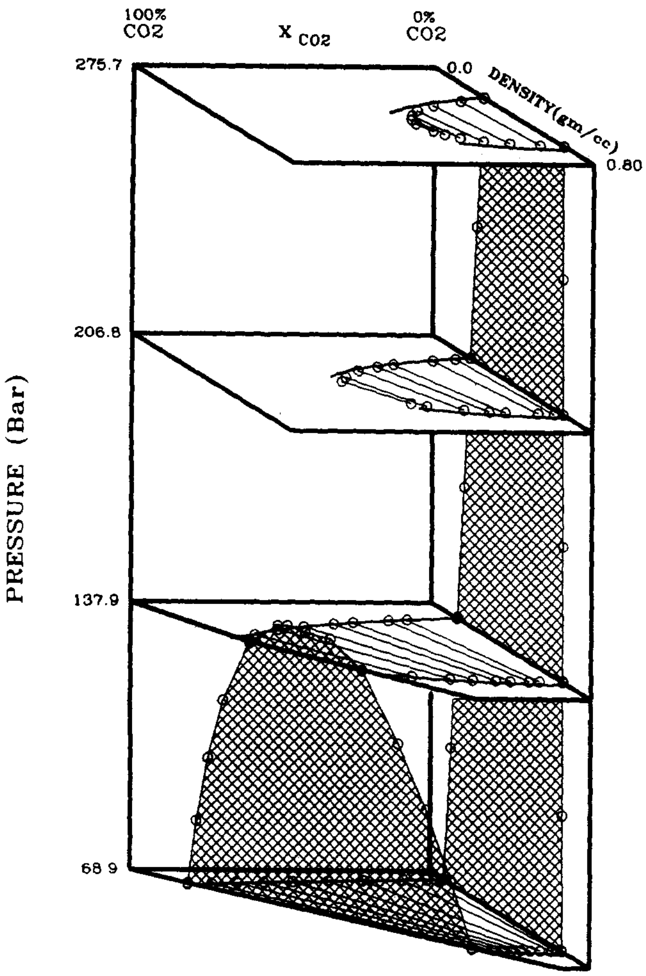

Figure 8. Three-dimensional view of phase density data for the ternary system of nitrogen, carbon dioxide, and cyclohexane at T = 410.9 K and three pressures. The crosshatched areas represent the binary two-phase regions.

Glossary

a	Peng–Robinson EOS parameter for mixture
a_{ii}	Peng–Robinson EOS parameter for component i
a_{ij}	cross parameter defined by eq 4
b	Peng–Robinson EOS parameter for mixture
b_i	Peng–Robinson EOS parameter for component i
k_{ij}	binary interaction parameter
n_c	number of components in mixture
NPTS	number of experimental data points
OBJ	objective function defined by eq 5
P	pressure
R	universal gas constant
T	temperature
V	volume
x_j	liquid mole fraction of component j
y_j	vapor mole fraction of component j

Subscripts/superscripts

i, j	component
CALC	calculated value
EXP	experimental value

Registry No. Carbon dioxide, 124-38-9; cyclohexane, 110-82-7; nitrogen, 7727-37-9.

Literature Cited

(1) Shibata, S. K.; Sandler, S. I.; Kim, H. High-Pressure Vapor–Liquid Equilibria Involving Mixtures of Nitrogen, Carbon Dioxide, and *n*-Butane. *J. Chem. Eng. Data* **1989**, *34*, 291. (An erratum will follow including H. Kim as an author of this paper.).
(2) Krichevskii, I. R.; Sorina, G. A. Liquid-Gas Phase Equilibria in the Cyclohexane–Carbon Dioxide and Cyclohexane–Nitrous Oxide Systems. *Russ. J. Phys. Chem.* **1960**, *34*, 679.
(3) Khodeeva, S. M. *Tr. Naucho-Issled. Pr. I. Azotn. Prom. Org. Sin.* **1971**, *12*, 18.
(4) Peng, D.-Y.; Robinson, D. B. New Two-Constant Equation of State. *Ind. Eng. Chem. Fundam.* **1976**, *15*, 59.

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High-Pressure Vapor–Liquid Equilibria of the Binary Mixtures Nitrogen + *n*-Butane and Argon + *n*-Butane

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High-pressure vapor–liquid equilibrium composition and density data of the binary systems nitrogen + *n*-butane (at 339.4 and 380.2 K) and argon + *n*-butane (at 340.0 and 380.1 K) are reported. The data sets were fit with the Peng–Robinson equation of state using both generalized and fluid-specific temperature-dependent parameters. The binary interaction parameters so obtained are reported.

Introduction

This study continues our research on the vapor–liquid equilibrium of mixtures of molecules asymmetric in size and critical properties (*1*, *2*). The data obtained in this study, in addition to being of direct interest, are of use for testing and improving equations of state and their mixing rules, especially for the simultaneous prediction of the equilibrium compositions, densities, and other thermodynamic properties of mixtures.

Experiment

A dynamic high-pressure vapor–liquid equilibrium apparatus (maximum pressure 5000 psi and maximum temperature 300 °F), which allows the simultaneous measurement of the compositions and densities of the coexisting phases, was used to obtain the data reported below. Shibata et al. (*1*) give a detailed description of the apparatus and its operation.

Grade 5 nitrogen, argon with a purity of 99.998%, and instrument grade *n*-butane, supplied by Matheson were used for the measurements. As described earlier (*1*), temperatures are measured with an error of less than 0.05 K and pressures with an error of 0.1 psi in the range below 800 psi and 2 psi above 800 psi. The error in the measurement of the phase compositions with a calibrated gas chromatograph is smaller than 0.005 mole fraction. Densities were measured with calibrated Mettler/Paar vibrating tube densimeters with an error of less than 0.0001 g/cm^3.

Table I. Experimental Data for the Nitrogen + *n*-Butane System at 339.4 K

T, K	P, bar	x_N	y_N	ρ_L, g/cm^3	ρ_V, g/cm^3
339.37	12.41	0.023	0.342	0.5242	0.0263
339.18	18.82	0.024	0.524	0.5230	0.0333
339.26	26.17	0.038	0.632	0.5226	0.0413
339.43	34.35	0.051	0.700	0.5212	0.0514
339.36	41.66	0.068	0.738	0.5203	0.0596
339.41	56.16	0.092	0.781	0.5190	0.0766
339.41	68.26	0.117	0.800	0.5175	0.0916
399.33	89.18	0.165	0.822	0.5144	0.1196
339.39	103.42	0.180	0.826	0.5120	0.1356
339.57	120.52	0.211	0.824	0.5095	0.1578
339.35	135.96	0.249	0.824	0.5042	0.1788
339.37	152.17	0.282	0.815	0.4869	0.2001
339.50	168.85	0.315	0.801	0.4810	0.2249
339.48	182.64	0.352	0.792	0.4746	0.2479
339.37	193.88	0.381	0.778	0.4685	0.2649
339.35	199.95	0.400	0.761	0.4640	0.2770
339.35	206.29	0.424	0.751	0.4563	0.2888
339.15	215.18	0.459	0.726	0.4467	0.3098
339.35	221.05	0.505	0.688	0.4201	0.3360

Table II. Experimental Data for the Nitrogen + *n*-Butane System at 380.2 K

T, K	P, bar	x_N	y_N	ρ_L, g/cm^3	ρ_V, g/cm^3
380.15	17.58	0.000	0.000	0.4518	0.0483
380.02	24.27	0.020	0.190	0.4501	0.0566
380.20	31.92	0.037	0.319	0.4478	0.0649
380.26	39.44	0.051	0.400	0.4458	0.0748
380.07	51.64	0.077	0.483	0.4409	0.0904
380.09	65.50	0.109	0.540	0.4348	0.1105
380.21	82.74	0.153	0.573	0.4254	0.1349
380.20	103.42	0.211	0.586	0.4097	0.1700
380.20	124.04	0.287	0.563	0.3833	0.2153
380.30	130.66	0.317	0.544	0.3634	0.2369
380.25	134.45	0.349	0.514	0.3489	0.2560
380.35	135.76	0.373	0.500	0.3370	0.2707

Results and Discussion

Tables I and II give the unsmoothed data for the equilibrium compositions and densities for the nitrogen + *n*-butane system at 339.4 and 380.0 K, respectively; these same data are plotted in Figures 1 and 2. The data for the argon + *n*-butane system at the same temperatures are shown in Tables III and IV and in Figures 3 and 4. The data were fit with the Peng–Robinson equation of state (PR-EOS) (*3*) with both generalized parameters and the fluid-specific temperature-dependent parameters of Xu and Sandler (*4*). The PR-EOS can be written as

$$P = \frac{RT}{v-b} - \frac{a}{v(v+b)+b(v-b)} \tag{1}$$

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