Vapor−Liquid Equilibria in the Nitrogen + Carbon Dioxide + Propane System from 240 to 330 K at Pressures to 15 MPa

Belgin Yucelen and Arthur J. Kidnay*

Department of Chemical Engineering and Petroleum-Refining, Colorado School of Mines,
Golden, Colorado 80401

Vapor−liquid equilibria were measured for the nitrogen, carbon dioxide, and propane ternary system and its constituent binaries over the temperature range 240 K to 330 K. The binary systems were measured at 240, 270, and 330 K, and the ternary system was studied at 240 K for 2.0 and 13.0 MPa, 270 K for 2.0, 6.0, and 13.0 MPa, and 330 K and 5 MPa. The Peng−Robinson equation of state was used to model the system.

Introduction

The design and development of separation processes in the natural gas industry requires accurate knowledge of the vapor−liquid equilibria of multicomponent systems consisting of the alkane hydrocarbons with carbon dioxide and nitrogen. Binary data are required for the development of thermodynamic models that are used for the prediction of phase equilibria in multicomponent mixtures, while ternary data are widely used to test the predictive models (Prausnitz, 1985). This is the tenth in a series of experimental studies designed to provide accurate vapor−liquid equilibria data for the gas processing industry.

Review of Previous Work

The binary isotherms for systems composed of nitrogen, carbon dioxide, and propane have been studied extensively, but there are no measurements on the ternary system. The literature is summarized in Table 1.

Table 1. Experimental Data for Systems of Nitrogen, Carbon Dioxide, and Propane

temp/K	ref
	Nitrogen + Carbon Dioxide
288−303	Krichevskii et al., 1962
218−273	Zenner and Dana, 1963
273	Muirbrock and Prausnitz, 1965
273	Yorizane et al., 1970
253−288	Arai et al., 1971
250−320	Altunin et al., 1974
270	Somait and Kidnay, 1978
220, 240	Al-Sahhaf, 1983
223−273	Dorau et al., 1983
250, 270	Brown et al., 1989a,b
293	Xu et al., 1992
	Carbon Dioxide + Propane
290−366	Poettman and Katz, 1945
278−344	Reamer et al., 1951
233−273	Akers et al., 1954
305−361	Roof and Baron, 1967
232−273	Nagahama et al., 1974
244, 266	Hamam and Lu, 1976
213−273	Acosta et al., 1984
311−361	Niesen and Rainwater, 1990
230, 270	Webster, 1996
	Nitrogen + Propane
90	Vellinger and Pons, 1943
92−128	Chueng and Wang, 1964
103−353	Schindler et al., 1966
312−365	Roof and Baron, 1967
172−255	Cannon et al., 1968
114−122	Poon and Lu, 1974
230−290	Grausø et al., 1977
183−303	Hudziak et al., 1984
117−127	Llave et al., 1985

Experimental Section

The gases used in this study were purchased commercially and used without further purification. The nitrogen, carbon dioxide, and propane had minimum purities of 99.995, 99.99, and 99.95 mol %, respectively.

Two separate experimental units were used, one for temperatures below ambient and one for temperatures above.

Low-Temperature Apparatus. The equipment and procedure described by Wei et al. (1995) were used, with slight modifications to the low-temperature bath and the analytical procedures. Temperatures were measured to ±0.01 K with a standard platinum resistance thermometer (IPTS-68), and pressures were measured by four Heise bourdon tube gauges (0−100 psia, 0−500 psia, 0−100 atm, and 0−2500 psia), accurate to ±0.1 % of the full scale reading.

Vapor and liquid samples were analyzed by a Hewlett-Packard 5890 series II gas chromatograph equipped with a thermal conductivity detector. A Hewlett Packard 3392 integrator was used for peak area evaluation. The composition of each sample was calculated using the calibration curves determined for each of the components by injecting pure gas samples at a series of pressures. All three components exhibited linear relationships between the partial pressure and the peak area. The calibrations were checked by injecting mixtures prepared gravimetrically in this laboratory. The estimated error in the measured mole fractions is ±0.002.

High-Temperature Apparatus. The apparatus used by Brown et al. (1989a,b) was rebuilt with modifications in the vapor and liquid sampling lines and the vapor circulation pump. A schematic diagram is represented in Figure 1. The equilibrium cell, vapor pump, and both the liquid and vapor sample valves were inside a Hotpack oven, which

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* To whom correspondence should be addressed.

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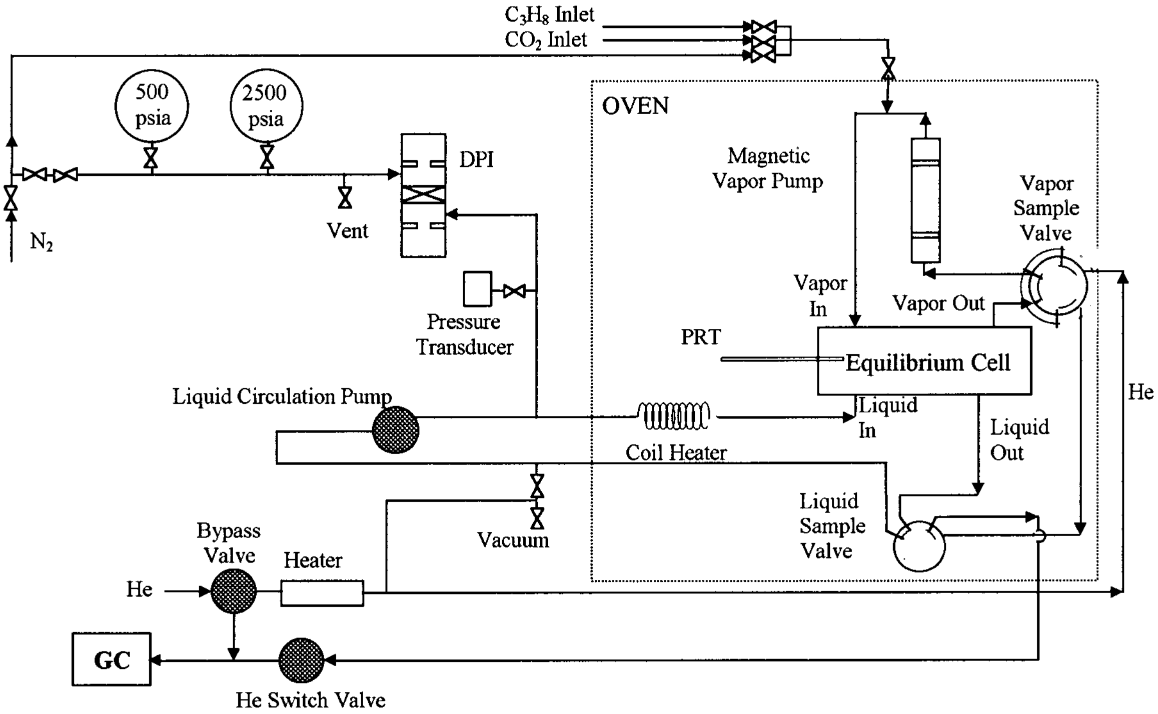

Figure 1. Schematic of high-temperature apparatus.

Table 2. Comparison of the Measured and Literature Vapor Pressures for Carbon Dioxide, Propane, and *n*-Butane

temp/K	vapor pressure/MPa measured	vapor pressure/MPa lit.	dev/MPa
	Carbon Dioxide[a]		
230.00	0.8946	0.8935	+0.0011
240.00	1.2841	1.2830	+0.0012
246.00	1.5710	1.5698	+0.0012
250.00	1.7837	1.7856	−0.0019
254.00	2.0243	2.0223	+0.0020
256.00	2.1477	2.1489	−0.0012
260.00	2.4173	2.4194	−0.0021
266.00	2.8723	2.8705	+0.0019
270.00	3.2018	3.2034	−0.0016
	Propane[b]		
240.00	0.1508	0.1479	+0.0029
270.00	0.4336	0.4304	+0.0032
330.00	1.9795	1.9822	−0.0027
343.15	2.5848	2.5868	−0.0020
344.26	2.6414	2.6417	−0.0003
	n-Butane[c]		
297.44	0.2386	0.2390	−0.0004
327.57	0.5585	0.5578	+0.0007
336.41	0.6933	0.6931	+0.0002
351.11	0.9715	0.9702	+0.0013

[a] Angus et al. (1976). [b] Goodwin and Haynes (1982a). [c] Goodwin and Haynes (1982b).

provided the temperature controlled environment. The temperature was measured with a platinum resistance thermometer (PRT) calibrated from 230 K to 373 K against the standard thermometer used for the low-temperature measurements. The PRT was placed inside the equilibrium cell in direct contact with the liquid and vapor. The PRT had a precision of ±0.01 K; however, shifts in the oven temperature decreased the accuracy of the temperature measurements to ±0.1 K. Pressures were measured from the liquid line using the Heise gauges of the low-temperature apparatus; the gauges were separated from the liquid by a Ruska differential null indicator.

Table 3. Nitrogen + Carbon Dioxide Binary System

pressure/MPa	x_{N_2}	y_{N_2}	pressure/MPa	x_{N_2}	y_{N_2}
		240 K			
1.285	0.0000	0.0000	3.341	0.0325	
1.696	0.0094		8.07		0.7002
1.707	0.0082		10.24	0.1705	
2.000		0.3186	10.93	0.1830	0.6977
2.647	0.0265		13.00	0.2467	0.6692
3.066	0.0307				
		270 K			
3.209	0.0000	0.0000	4.108	0.0164	
3.567	0.0064	0.0759	5.076	0.0358	0.2629
3.688	0.0096		6.986		0.3728
3.719	0.0088	0.1036	10.90	0.1850	
3.871	0.0125		11.97	0.2456	0.3822

A magnetic piston pump circulated the vapor from the top of the cell through a 2.8 μL sample valve and back to the bottom of the cell. The liquid was transferred by a diaphragm pump through a 0.2 μL sample valve and a stainless steel heat exchanger before returning to the cell. The samples were analyzed with the equipment and procedures used for the low-temperature measurements.

Results and Discussion

The accuracy of the temperature and pressure measurements for both experimental units was verified by comparing the measured vapor pressures of the pure components with accepted values from the literature (Table 2).

The results of all the binary and ternary measurements are given in Tables 3−6. The data were modeled with the Peng−Robinson (1976) equation of state, using the standard linear mixing rule for the van der Waals volume b and quadratic mixing rule for the attraction parameter a, with one binary interaction coefficient, k_{ij}. The interaction coefficients (Table 7) were obtained by selecting the k_{ij} which minimized the differences between calculated and experimental vapor and liquid mole fractions for the binary isotherms.

Nitrogen + Carbon Dioxide Binary System. Figure 2 compares the data of this study at 270 K with previous

Table 4. Carbon Dioxide + Propane Binary System

pressure/MPa	x_{CO_2}	y_{CO_2}	pressure/MPa	x_{CO_2}	y_{CO_2}	pressure/MPa	x_{CO_2}	y_{CO_2}	pressure/MPa	x_{CO_2}	y_{CO_2}
240 K											
0.1508	0.0000	0.0000	0.4640	0.1282	0.6935	0.8370	0.3354	0.8488	1.057	0.5700	0.8933
0.2020	0.0220		0.5896		0.7570	0.8811	0.3736	0.8583	1.200	0.8206	0.9302
0.3160	0.0604	0.5424	0.7277		0.8122	0.9740		0.8808	1.284	1.0000	1.0000
0.4480	0.1203	0.6787	0.7440	0.2727	0.8184	1.017	0.5165	0.8902			
270 K											
0.4340	0.0000	0.0000	1.014	0.1129		2.022	0.4161	0.8095	2.287	0.5326	0.8374
0.6990	0.0524		1.331		0.6843	2.055		0.8102	2.460		0.8574
0.9130	0.0940		1.700	0.3107	0.7658	2.249	0.5106	0.8377	3.202	1.0000	1.0000
330 K											
1.980	0.0000	0.0000	2.886	0.0876	0.2640	4.439	0.2710		5.438	0.3860	
2.189	0.0197	0.0873	3.722	0.1890	0.4000	5.000	0.3340	0.5150	5.852	0.4430	0.5690
2.479	0.0458	0.1810									

Table 5. Nitrogen + Propane Binary System

pressure/MPa	x_{N_2}	y_{N_2}	pressure/MPa	x_{N_2}	y_{N_2}	pressure/MPa	x_{N_2}	y_{N_2}	pressure/MPa	x_{N_2}	y_{N_2}
240 K											
0.1508	0.0000	0.0000	2.768	0.0537	0.9291	9.315	0.1793	0.9462	14.00	0.2721	0.9313
1.191	0.0200	0.8652	4.823	0.0942	0.9434	12.21		0.9386	15.08	0.2845	0.9252
2.069	0.0378	0.9125	7.094	0.1385	0.9486	12.84	0.2447	0.9375			
270 K											
0.4336	0.0000	0.0000	4.277	0.0785	0.8485	6.577	0.1240		13.70	0.2837	0.8655
2.006	0.0305	0.7482	4.433	0.0820	0.8504	7.988	0.1590	0.8920	14.98	0.3168	0.8628
2.279	0.0376	0.7785	5.023	0.0920		10.16		0.8817			
3.547	0.0651	0.8350	5.249	0.0976	0.8673	11.49	0.2369	0.8806			
330 K											
1.981	0.0000	0.0000	3.975	0.0471	0.3721	5.711	0.0985	0.4705	9.808	0.2180	0.5530
2.979	0.0248	0.2325	4.845	0.0774	0.4219	6.468	0.1192	0.4981	12.50	0.3180	0.5460
3.518	0.0390	0.3085	4.995	0.0798	0.4302	7.919	0.1600	0.5339			

Table 6. Nitrogen + Carbon Dioxide + Propane Ternary System

pressure/MPa	x_{N_2}	x_{CO_2}	$x_{C_3H_8}$	y_{N_2}	y_{CO_2}	$y_{C_3H_8}$	pressure/MPa	x_{N_2}	x_{CO_2}	$x_{C_3H_8}$	y_{N_2}	y_{CO_2}	$y_{C_3H_8}$
240 K and 2.0 MPa													
2.000	0.0294	0.0048	0.9658	0.8953	0.0094	0.0951	2.000	0.0238	0.5375	0.4387	0.4325	0.4968	0.0707
2.000	0.0284	0.3539	0.6177	0.5205	0.4037	0.0758	2.000	0.0211	0.6317	0.3472	0.4030	0.5374	0.0596
2.000				0.3896	0.5545	0.0560	2.000	0.0175	0.7413	0.2412	0.3507	0.6092	0.0401
2.000				0.4746	0.4543	0.0712	2.000	0.0266	0.9238	0.0496	0.8216	0.0859	0.0925
2.000	0.0265	0.4229	0.5506	0.4849	0.4424	0.0727	2.000	0.0130	0.9870	0.0000	0.3186	0.6814	0.0000
240 K and 13.0 MPa													
13.00	0.2500	0.0000	0.7500	0.9370	0.0000	0.0630	13.00	0.3114	0.2817	0.4069	0.7649	0.1598	0.0753
12.99				0.8490	0.0729	0.0781	13.00	0.2612	0.6986	0.0403	0.9006	0.0263	0.0731
13.00				0.8013	0.1240	0.0747	13.00	0.2531	0.7450	0.0019	0.6679	0.3316	0.0005
13.00				0.7878	0.1366	0.0756	13.00	0.2467	0.7533	0.0000	0.6692	0.3308	0.0000
12.99	0.3014	0.2485	0.4501	0.7810	0.1444	0.7460							
270 K and 2.0 MPa													
2.006	0.0305	0.0000	0.9695	0.7320	0.0000	0.2680	2.000	0.0069	0.3019	0.6862	0.1179	0.6772	0.2049
2.000	0.0286	0.0182	0.9532	0.6812	0.0676	0.2512	2.000	0.0038	0.3621	0.6341	0.0613	0.7381	0.2006
2.000	0.0204	0.1234	0.8562	0.4046	0.3373	0.2581	2.000	0.0008	0.3956	0.6037	0.0062	0.8055	0.1883
2.000	0.0141	0.2087	0.7790	0.2607	0.4994	0.2399	2.000	0.0000	0.8170	0.1830	0.0000	0.4150	0.5850
2.000	0.0096	0.2774	0.7130	0.1669	0.6133	0.2198							
270 K and 6.0 MPa													
6.00	0.1115	0.0000	0.8885	0.8720	0.0000	0.1280	6.00	0.0882	0.4861	0.4260	0.4845	0.4205	0.0951
6.00	0.1133	0.0079	0.8787	0.8533	0.0106	0.1363	6.01	0.0933	0.5475	0.3592	0.4606	0.4493	0.0900
6.01	0.0974	0.2085	0.6921	0.6595	0.2197	0.1208	6.01	0.0904	0.5770	0.3326	0.4463	0.4667	0.0871
5.99	0.1012	0.3038	0.5950	0.5894	0.2935	0.1171	6.00	0.0682	0.8435	0.0882	0.3543	0.6107	0.0351
6.00	0.1031	0.3069	0.5901	0.5966	0.2945	0.1088	6.00	0.0530	0.9470	0.0000	0.3250	0.6750	0.0000
6.00	0.0937	0.4002	0.5061	0.5270	0.3675	0.1055							
270 K and 13.0 MPa													
13.00	0.2646	0.0000	0.7354	0.8612	0.0000	0.1388	13.00	0.2990	0.2990	0.4020	0.6657	0.1828	0.1515
13.00	0.2663	0.0217	0.7120	0.8386	0.0192	0.1423	13.00				0.6312	0.2166	0.1522
13.00				0.7839	0.0727	0.1434	12.99				0.6060	0.2400	0.1541
13.00	0.2854	0.2046	0.5100										
330 K and 5.0 MPa													
5.00	0.0000	0.3340	0.6660	0.0000	0.5150	0.4850	5.00	0.0357	0.1479	0.8164	0.1650	0.2967	0.5383
5.00	0.0090	0.2989	0.6921				5.00	0.0450	0.1297	0.8253	0.2273	0.2428	0.5299
5.00	0.0149	0.2681	0.7170				5.00				0.2634	0.1977	0.5389
5.00	0.0204	0.2258	0.7538	0.0300	0.4460	0.5240	5.00	0.0712	0.0367	0.8921	0.3713	0.0715	0.5572
4.99	0.0229	0.2216	0.7555	0.0400	0.4236	0.5364	5.01	0.0792	0.0000	0.9208	0.4339	0.0000	0.5661
5.00	0.0320	0.1884	0.7796	0.1031	0.3603	0.5366							

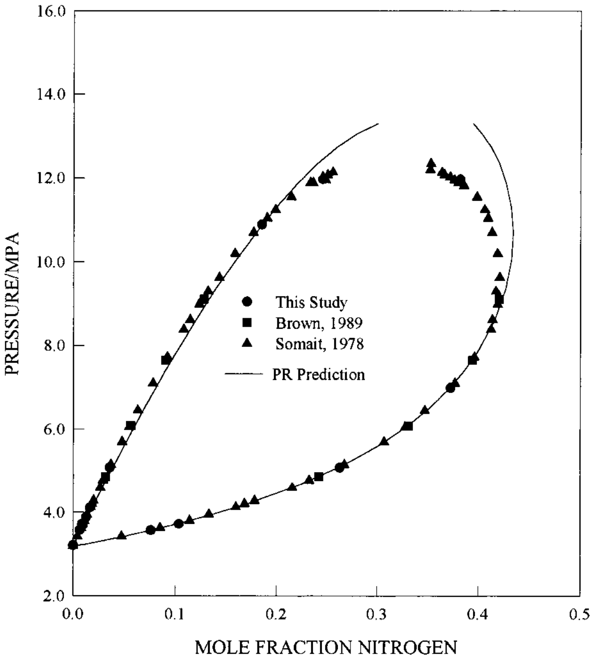

Figure 2. Nitrogen + carbon dioxide at 270 K.

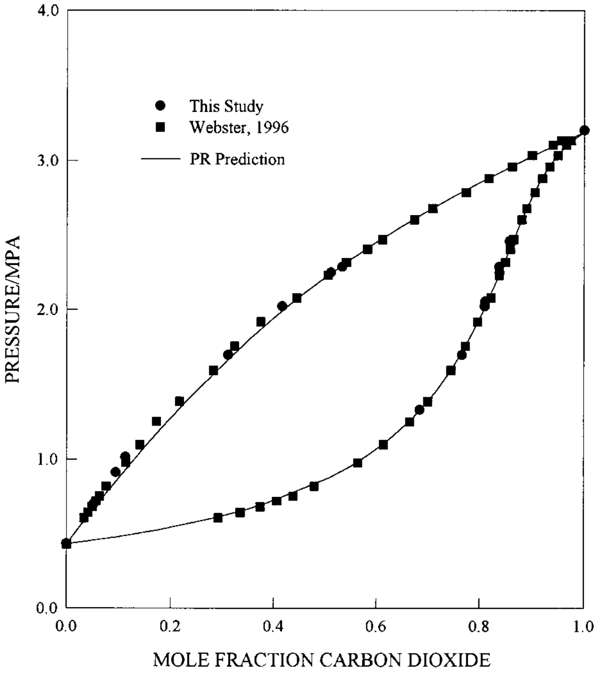

Figure 3. Carbon dioxide + propane at 270 K.

Table 7. Binary Interaction Parameters (k_{ij}) for the Peng–Robinson Equation of State

temp/K	k_{ij}	AAD
	Nitrogen + Carbon Dioxide	
240.00	−0.029	0.0043
270.00	−0.024	0.0045
	Carbon Dioxide + Propane	
240.00	0.126	0.0044
270.00	0.120	0.0050
330.00	0.124	0.0061
	Nitrogen + Propane	
240.00	0.069	0.0043
270.00	0.054	0.0079
330.00	0.089	0.0024

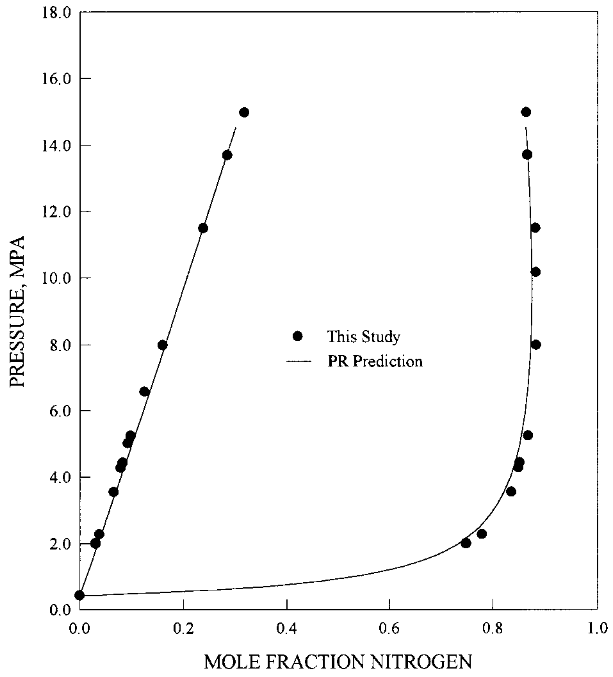

Figure 4. Nitrogen + propane at 270 K.

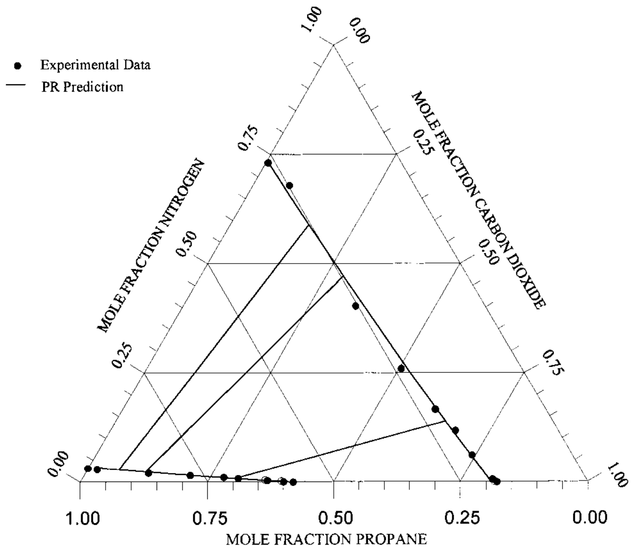

Figure 5. Nitrogen + carbon dioxide + propane at 270 K and 2.0 MPa.

work, and shows the results of calculations using the Peng–Robinson (PR) equation with k_{ij} from Table 7. The PR equation is not capable of accurately modeling the critical region for this system, a result that is commonly observed.

Carbon Dioxide + Propane Binary System. The results at 270 K are shown in Figure 3. The data at 240 K and 270 K are below the critical temperature of carbon dioxide (304.21 K), while the 330 K isotherm is considerably above. The PR equation is quite satisfactory in modeling at all temperatures, although the critical region at 330 K is not well-defined experimentally. Limited measurements were also made at 344.26 K to compare with the earlier work of Reamer et al. (1951). The agreement was excellent.

Nitrogen + Propane Binary System. The data at 270 K are shown in Figure 4, together with the PR predictions. The measurements were made to pressures of 15 MPa, which was the maximum limit for the pressure gauges used. For all isotherms the fits from the PR equation of

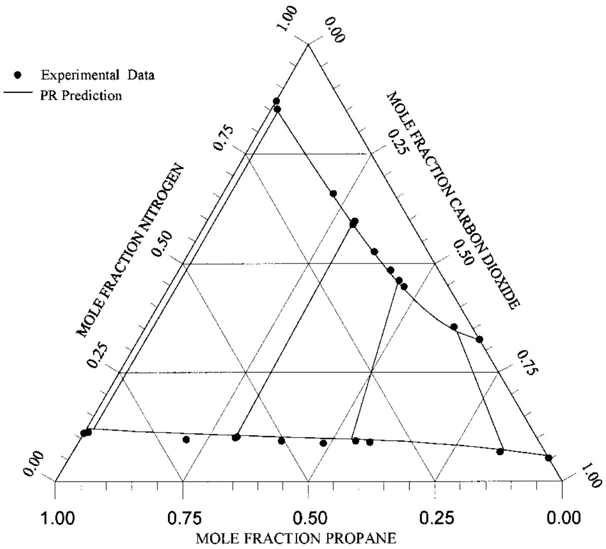

Figure 6. Nitrogen + carbon dioxide + propane at 270 K and 6.0 MPa.

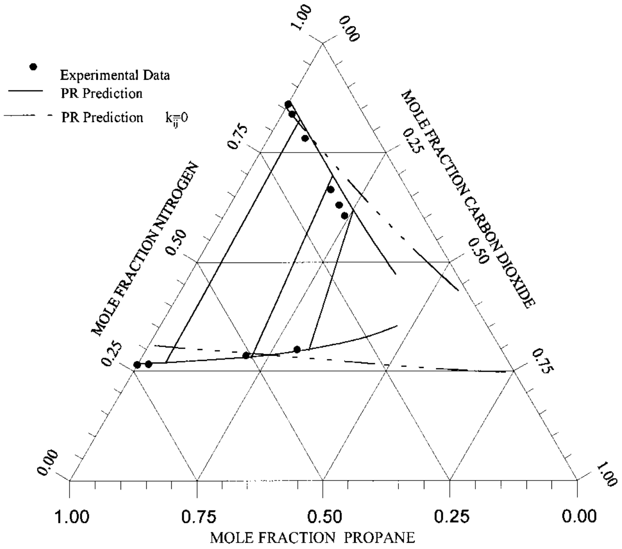

Figure 7. Nitrogen + carbon dioxide + propane at 270 K and 13.0 MPa.

state were good for both the vapor and the liquid phases, but again the critical region at 330 k is not well-defined experimentally.

Nitrogen + Carbon Dioxide + Propane Ternary System. **T**he ternary data and the PR predictions at 270 K for 2.0, 6.0, and 13.0 MPa, at 240 K for 2.0 and 13.0 MPa, and at 330 K for 5.0 MPa are shown in Figures 5−9. The binary end points were checked by comparing them with the previously generated binary curves at each pressure and were found to be within ±0.002 mole fraction of the previous points.

At 270 K and 13.0 MPa the ternary system has a critical point since the system pressure was above the critical pressure of the nitrogen + carbon dioxide binary system. For all other conditions two binary end points exist. The accuracy of the predictions for the ternary system depends on the goodness of the fit of the binary systems which form the boundaries of the ternary system. At low pressures the PR equation models the data very well; this is reflected in the quality of the ternary predictions. At 13.0 MPa, the PR fits to the binary data were particularly poor for the liquid phase at 240 K and for the vapor phase at 270 K, and as a result the ternary predictions were also poor in these regions. Peng−Robinson predictions for 13.0 MPa and 270 K using k_{ij} values of 0.0 are shown in Figure 7, demonstrating the importance of the interaction coefficients.

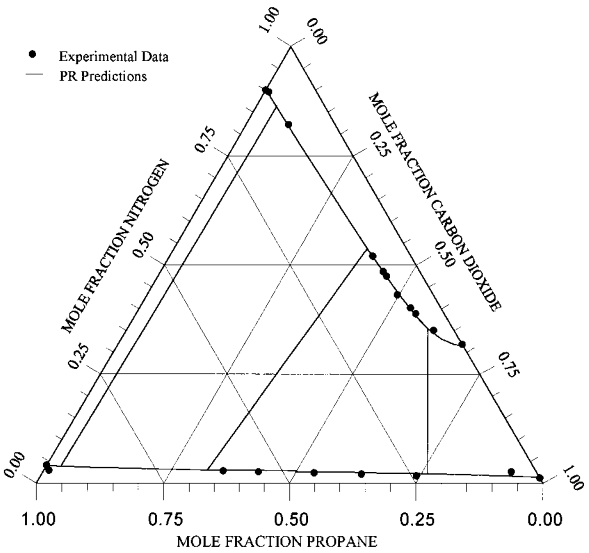

Figure 8. Nitrogen + carbon dioxide + propane at 240 K and 2.0 MPa.

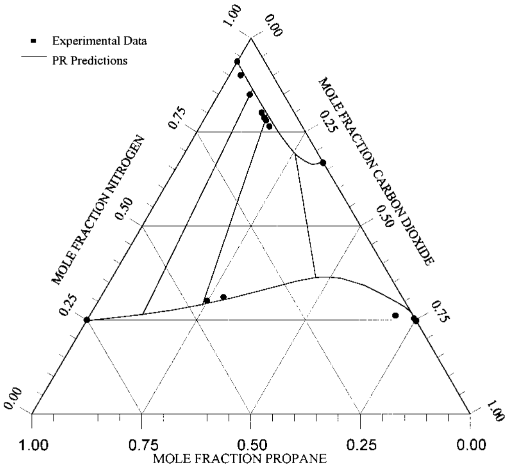

Figure 9. Nitrogen + carbon dioxide + propane at 240 K and 13.0 MPa.

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