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Reply to Comments by Stanislaw K. Malanowski and Roman Stryjek on *J. Chem. Eng. Data* 1999, *44*, 1226−1230

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We appreciate the interest and comments from S. K. Malanowski and R. Stryjek on our publication. Following are our answers to the comments.

1. You are correct. We had made a mistake. The unit of the pressures at 313.2 K for the HFC-152a + isobutane system in Table 3 was bar in the original publication. Correct values can be obtained in MPa by multiplying the reported data by the factor 0.1.

2. We remeasured several times the vapor pressures of HFC-134a and isobutane at 323.2 K. At this time, we obtained a vapor pressure of HFC-134a P = 1.313 MPa and for isobutane P = 0.6836 MPa, respectively, within a repeatability of 0.003 MPa. We appear to have made a measurement error.

3. The gas chromatograph was recalibrated with pure components of known purity and with mixtures of known composition that were prepared gravimetrically. The analysis uncertainty of the liquid and vapor phase composition is estimated to be within ±0.005 mole fraction, including the uncertainty of the calibration curve. Recalculation of

Table 2. Vapor−Liquid Equilibrium Measurements for the HFC-134a (1) + Isobutane (2) System

x_1	y_1	P/MPa
	T/K = 303.2	
0.0000	0.0000	0.4070
0.0330	0.1710	0.4830
0.0731	0.2855	0.5600
0.1440	0.4039	0.6540
0.2230	0.4790	0.7320
0.3575	0.5635	0.8070
0.4521	0.6074	0.8440
0.5225	0.6327	0.8620
0.5585	0.6453	0.8720
0.6531	0.6845	0.8760
0.7422	0.7246	0.8810
0.8452	0.7893	0.8630
0.8938	0.8310	0.8460
0.9265	0.8769	0.8280
0.9675	0.9387	0.7980
1.0000	1.0000	0.7630
	T/K = 323.2	
0.0000	0.0000	0.6836
0.0301	0.1314	0.7700
0.0688	0.2408	0.8820
0.1294	0.3407	1.0070
0.2088	0.4374	1.1480
0.3258	0.5280	1.2710
0.4267	0.5782	1.3420
0.4976	0.6159	1.3840
0.5418	0.6356	1.4040
0.6372	0.6770	1.4210
0.7337	0.7340	1.4330
0.8486	0.8121	1.4130
0.8969	0.8559	1.3930
0.9375	0.9031	1.3600
0.9727	0.9550	1.3280
1.0000	1.0000	1.3130

Table 3. Vapor−Liquid Equilibrium Measurements for the HFC-152a (1) + Isobutane (2) System

x_1	y_1	P/MPa
	T/K = 303.2	
0.0000	0.0000	0.4070
0.0492	0.1633	0.4820
0.1969	0.4005	0.6290
0.4048	0.5517	0.7210
0.5716	0.6368	0.7570
0.6751	0.6936	0.7670
0.8008	0.7747	0.7590
0.8405	0.8089	0.7580
0.9165	0.8828	0.7300
1.0000	1.0000	0.6860
	T/K = 313.2	
0.0000	0.0000	0.5300
0.2659	0.4625	0.8598
0.5152	0.6151	0.9690
0.7212	0.7281	0.9950
0.7624	0.7549	0.9912
0.8066	0.7858	0.9904
0.9004	0.8685	0.9632
0.9632	0.9432	0.9334
1.0000	1.0000	0.9058
	T/K = 323.2	
0.0000	0.0000	0.6870
0.1264	0.2985	0.9270
0.2810	0.4703	1.1200
0.6014	0.6631	1.2570
0.7242	0.7361	1.2730
0.7564	0.7550	1.2720
0.7599	0.7576	1.2700
0.8511	0.8285	1.2580
0.9285	0.9057	1.2200
0.9897	0.9857	1.1790
1.0000	1.0000	1.1770
	T/K = 333.2	
0.0000	0.0000	0.8724
0.1291	0.2852	1.1896
0.2875	0.4664	1.4020
0.5689	0.6434	1.5682
0.6581	0.6967	1.5972
0.7425	0.7517	1.6058
0.7977	0.7922	1.6048
0.8734	0.8556	1.5900
0.9365	0.9201	1.5500
1.0000	1.0000	1.5000

the data given in the original publication shows some deviations; the new values are given in the following Tables 2−4. For the correlation work in the original publication, we obtained model parameters at 303.2 K or 301.8 K and then predicted VLE at 323.2 K (for HFC-134a + isobutane) and 313.2, 323.2, and 333.2 K (for HFC-152a + isobutane) or 321.8 K (for HFC-32 + isobutane) with these parameters, respectively. Because the temperature ranges of binary systems studied here were relatively short, k_{ij}, A_{ij}, and A_{ji} were fixed with the values obtained at 303.2 K (or 301.8 K for HFC-32 + isobutane). So, the deviations seem
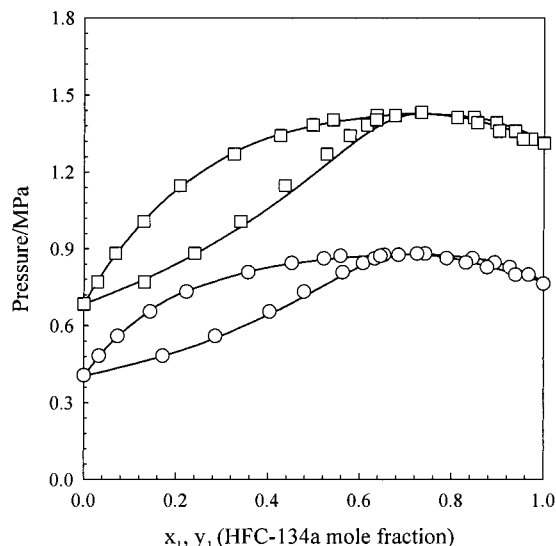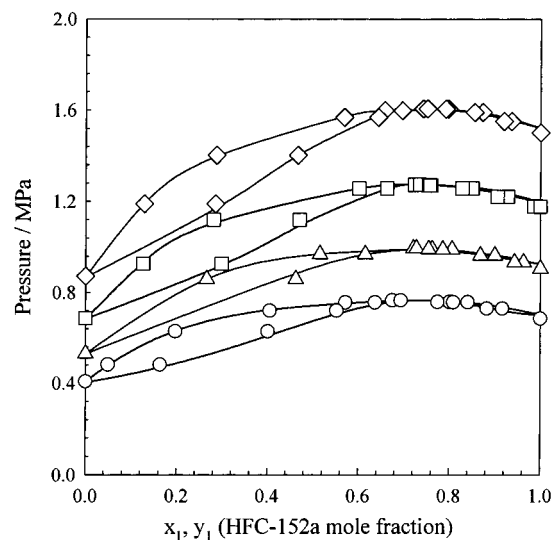
Table 4. Vapor−Liquid Equilibrium Measurements for the HFC-32 (1) + Isobutane (2) System

x_1	y_1	P/MPa
	T/K = 301.8	
0.0000	0.0000	0.3920
0.0341	0.2992	0.5790
0.0504	0.3843	0.6690
0.0958	0.5277	0.8660
0.1275	0.5803	0.9940
0.2048	0.6675	1.2420
0.2490	0.6952	1.3500
0.3786	0.7496	1.5750
0.4845	0.7837	1.6970
0.5996	0.8073	1.7900
0.6978	0.8352	1.8590
0.7804	0.8555	1.9030
0.8281	0.8702	1.9200
0.8709	0.8877	1.9320
0.9072	0.9087	1.9370
0.9154	0.9102	1.9380
0.9360	0.9301	1.9330
0.9627	0.9534	1.9180
0.9883	0.9848	1.9010
1.0000	1.0000	1.8950
	T/K = 321.8	
0.0000	0.0000	0.6520
0.0240	0.2227	0.8730
0.0374	0.3076	0.9920
0.0846	0.4310	1.2260
0.1182	0.4980	1.4030
0.1907	0.5986	1.7590
0.2290	0.6317	1.9060
0.3786	0.7119	2.3440
0.4462	0.7420	2.4980
0.6110	0.7919	2.7790
0.6729	0.8121	2.8690
0.7722	0.8470	2.9740
0.8257	0.8671	3.0220
0.8640	0.8877	3.0560
0.9034	0.9124	3.0660
0.9124	0.9183	3.0750
0.9360	0.9367	3.0770
0.9627	0.9591	3.0730
0.9876	0.9869	3.0330
1.0000	1.0000	3.0220

Table 6. Values of Binary Parameters and AAD of P and y

system	T/K	NRTL parameters	AAD(P)[a]	AAD(y)[b]
HFC-134a + isobutane	303.15	k_{ij} = 0.173 A_{ji} = 2.634 A_{ij} = 3.016	0.481	0.008
	323.15 0.014	k_{ij} = 0.485 A_{ji} = 5.272 A_{ij} = −2.429	0.464	0.014
HFC-152a + isobutane	303.15	k_{ij} = 0.434 A_{ji} = 5.328 A_{ij} = −2.304	0.676	0.010
	313.15	k_{ij} = 0.632 A_{ji} =18.638 A_{ij} = −4.087	0.469	0.007
	323.15	k_{ij} = 0.430 A_{ji} = 6.213 A_{ij} = −2.873	0.737	0.003
	333.15	k_{ij} = 0.347 A_{ji} = 6.185 A_{ij} = −2.236	0.378	0.004
HFC-32 + isobutane	301.8	k_{ij} = 0.318 A_{ji} = 2.189 A_{ij} = 4.611	0.258	0.006
	321.8	k_{ij} = 0.533 A_{ji} = 6.382 A_{ij} = −1.630	0.998	0.011

[a] AAD%(P) = (1/N)Σ|(P_{exp} − P_{cal})/P_{exp}| × 100. [b] AAD%(y) = (1/N)Σ|y_{exp} − y_{cal}|. The unit of A_{ij} and A_{ji} is kJ·mol^{-1}.

Figure 1. $P−x−y$ diagram for the HFC-134a (1) + isobutane (2) system: ○, experimental at 303.2 K; □, experimental at 323.2 K; −, calculated at 303.2 K and 323.2 K.

Figure 2. $P−x−y$ diagram for the HFC-152a (1) + isobutane (2) system: ○, experimental at 303.2 K; △, experimental at 313.2 K; □, experimental at 323.2 K; ◇, experimental at 333.2 K; −, calculated at (303.2, 313.2, 323.2 and 333.2) K.

Table 7. Azeotropic Data of HFC-134a + Isobutane, HFC-152a + Isobutane, and HFC-32 + Isobutane Systems

system	T/K	x_1	P/MPa
HFC-134a + isobutane	303.2	0.729	0.883
	323.2	0.740	1.436
HFC-152a + isobutane	303.2	0.722	0.769
	313.2	0.733	0.997
	323.2	0.746	1.274
	333.2	0.767	1.608
HFC-32 + isobutane	301.8	0.909	1.939
	321.8	0.943	3.079

to be very large. At this time, we refit the model parameters to VLE at each temperature for three systems. All the binary parameters and the average absolute deviations (AADs) between measured and calculated values of P and y are listed in Table 6. Figures 1−3 show the comparison of measured and calculated values. The newly obtained azeotropic data are shown in Table 7.
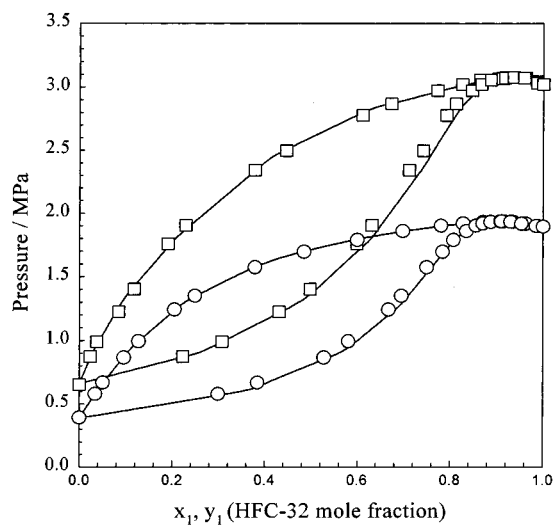
Figure 3. $P-x-y$ diagram for the HFC-32 (1) + isobutane (2) system: ○, experimental at 301.8 K; □, experimental at 321.8 K; —, calculated at (301.8 and 321.8) K.

4. Figure 4 shows comparison between our revised result and published values[1] for the HFC134a + isobutane system.

Literature Cited

Bobbo, S.; Stryjek, R.; Elvassore, N.; Bertucco, A. A Recirculation Apparatus for Vapor-Liquid Equilibrium Measurements of Refriger-
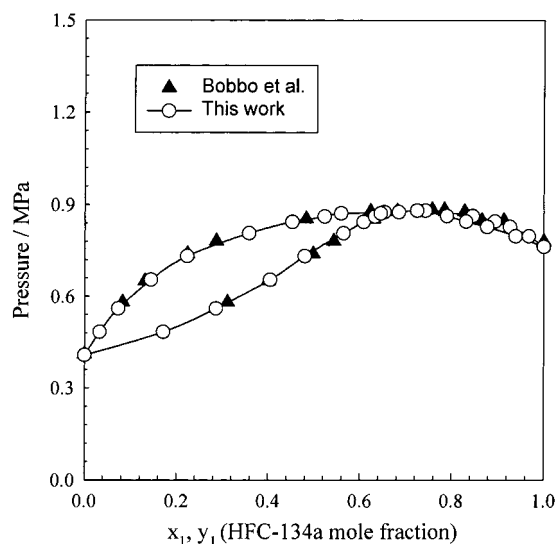

Figure 4. $P-x-y$ diagram of the HFC-134a (1) + isobutane (2) system: ○, present work at $T = 303.2$ K; ▲, Bobbo et al.[1] at $T = 303.68$ K.

ants. Binary Mixtures of R600a, R134a and 236fa. *Fluid Phase Equilib.* **1998**, *150−15*1, 343−352.