Table XI. Compromise UNIQUAC Parameters

Size and Shape Parameters

	r	q	q'
SBA	3.9235	3.6640	4.0643
DSBE	6.0909	5.1680	5.7409
water	0.9200	1.4000	1.6741

Interaction Coefficients, a_{ij}, cal/mol

	SBA	DSBE	water
SBA	0.000	−193.141	424.025
DSBE	415.855	0.000	315.312
water	103.810	3922.50	0.000

mental measurements for the tower (*1*).

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Glossary

a_{ij}	adjustable interaction coefficients
c	regression parameter
K	vapor–liquid equilibrium constant
P	pressure, bar
q	structural area parameter
q'	modified structural area parameter
r	structural size (volume) parameter
T	temperature, K
x'	mole fraction in first liquid phase
x''	mole fraction in second liquid phase
y	vapor mole fraction

Subscripts

i, j	chemical species indices

Registry No. SBA, 78-92-2; MEK, 78-93-3; DSBE, 6863-58-7.

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Measurements of *PVTx* Properties for the R 13B1 + R 114 System

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The *PVTx* properties for the R 13B1 + R 114 system have been measured by the constant-volume method coupled with several isothermal expansion procedures. The results for four different compositions of 25, 50, 70, and 80 wt % R 13B1 cover the range of temperatures 303–443 K and the range of pressures 0.5–10.3 MPa, which corresponds to the density variation from 150 to 1200 kg/m^3. The experimental uncertainties of the temperature, pressure, density, and mass fraction measurements were estimated to be no greater than 8 mK, 2.0 kPa, 0.15%, and 0.12%, respectively. The dew and bubble points were determined.

Introduction

The *PVTx* properties of refrigerant mixtures have been recently of particular interest not only to the refrigeration industry but also to the power industry since refrigerant mixtures have become regarded as new, prospective working substances. We have been measuring the *PVTx* properties of refrigerant mixtures systematically and the results on the R 12 + R 22 system (*1*) and the R 22 + R 114 system (*2*) were published. In succession we have measured the *PVTx* properties of the R 13B1 + R 114 system. This paper reports the *PVTx* measurements for four different compositions of 25, 50, 70, and 80 wt % R 13B1 in the range of temperatures 303–443 K and the range of pressures 0.5–10.3 MPa, which covers the density range 150–1200 kg/m^3. The dew and bubble points are also reported. The purity of each component supplied was either 99.965 or 99.990 wt % R 13B1 (bromotrifluoromethane, $CBrF_3$) and 99.94 wt % R 114 (dichlorotetrafluoroethane) being an isomeric blend of 95% $CClF_2CClF_2$ + 5% CCl_2FCF_3.

Experimental Section

The *PVTx* measurements of this work have been made by the constant-volume method coupled with several isothermal expansion procedures for a mixture of a given composition. The apparatus and experimental procedure used here have

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Table I. Experimental *PVTx* Data for the R 13B1 + R 114 System

ρ, kg/m^3	T, K	P, MPa	ρ, kg/m^3	T, K	P, MPa	ρ, kg/m^3	T, K	P, MPa	ρ, kg/m^3	T, K	P, MPa
(A) 25 wt % R 13B1 + 75 wt % R 114											
1193	371.148	2.7158	780.6	443.156	7.3982	453.2	406.118	3.7457	259.4	398.158	2.9381
1193	378.148	4.3448	734.8	406.138	4.0610	453.1	409.119	3.8742	259.3	403.158	3.0443
1193	383.150	5.5182	734.5	413.141	4.5942	453.0	413.120	4.0457	259.2	413.156	3.2503
1192	388.147	6.6991	734.1	423.153	5.3831	452.8	423.119	4.4703	258.9	433.159	3.6479
1192	393.147	7.8821	733.7	433.155	6.1845	452.5	433.120	4.8836	258.8	443.157	3.8414
1192	398.149	9.0766	733.3	443.142	6.9936	452.3	443.118	5.2917	216.4	443.151	3.4242
1191	403.149	10.2732	613.1	443.148	6.1885	378.1	443.137	4.8367	170.3	393.146	2.5215
996.1	403.145	4.9602	576.9	409.141	4.0611	355.9	403.152	3.4362	160.0	393.154	2.4601
936.0	398.559	3.8243	576.8	413.144	4.2876	355.8	413.145	3.7665	159.9	403.150	2.6043
935.7	403.144	4.4195	576.5	423.143	4.8514	355.6	423.148	4.0795	159.8	413.151	2.7450
935.2	413.147	5.6510	576.2	433.122	5.4134	355.4	433.157	4.3792	159.8	423.160	2.8828
934.7	423.152	6.9198	575.9	443.121	5.9794	355.2	443.135	4.6799	159.7	433.155	3.0178
934.2	433.156	8.2102	481.7	433.141	5.0140	275.8	423.160	3.5665	159.6	443.151	3.1511
933.7	443.149	9.5144									
(B) 50 wt % R 13B1 + 50 wt % R 114											
1092	371.145	3.6272	664.8	403.138	5.3215	513.9	393.143	4.3234	321.3	423.146	4.4444
1092	373.152	3.9407	664.4	413.137	5.9920	513.6	403.152	4.8131	320.9	443.148	4.9449
1091	383.132	5.5480	664.1	423.139	6.6645	513.4	413.146	5.3042	265.9	443.152	4.3839
1090	393.141	7.2117	663.8	433.146	7.3384	513.1	423.155	5.7858	220.3	443.139	3.8368
1089	407.615	9.6722	663.4	443.143	8.0109	512.9	433.157	6.2384	221.1	377.148	2.7593
904.0	381.159	4.0824	620.4	389.149	4.2826	512.6	443.155	6.7384	220.9	393.159	3.0328
903.9	383.146	4.2665	620.2	393.150	4.5283	456.6	389.198	4.0316	220.8	403.150	3.1986
903.4	393.141	5.2946	619.9	403.132	5.1452	456.5	391.152	4.1227	220.7	413.601	3.3614
903.0	403.155	6.3883	619.6	413.148	5.7641	456.5	393.155	4.2079	220.6	423.160	3.5220
902.5	413.156	7.5062	619.3	423.156	6.3819	456.3	403.147	4.6224	220.4	433.140	3.6801
902.0	423.156	8.6398	619.0	433.163	6.9990	456.0	413.153	5.0281	182.6	433.172	3.2144
901.5	433.136	9.7800	618.6	443.159	7.6148	455.8	423.152	5.4282	151.9	359.158	2.0068
748.6	393.157	4.7924	551.2	391.132	4.3226	455.6	433.149	5.8224	151.9	363.148	2.0521
748.2	403.150	5.5692	551.1	393.131	4.4323	455.3	443.175	6.2136	151.7	383.161	2.2702
747.8	413.153	6.3730	550.8	403.143	4.9644	321.9	385.165	3.4443	151.6	393.148	2.3757
747.4	423.148	7.1858	550.5	413.130	5.4881	321.8	393.147	3.6630	151.5	413.150	2.5824
747.0	433.163	8.0044	550.3	423.132	6.0105	322.1	373.154	2.9729	151.4	423.148	2.6838
746.6	443.152	8.8276	550.0	433.069	6.5250	322.0	377.146	3.1392	151.3	433.150	2.7843
665.3	389.142	4.3945	550.0	443.139	7.0441	321.6	403.146	3.9288	151.3	443.148	2.8838
665.1	393.132	4.6542	514.0	391.124	4.2268						
(C) 70 wt % R 13B1 + 30 wt % R 114											
1195	353.137	3.7786	732.5	393.149	6.0808	451.8	376.136	4.2508	354.6	393.150	3.8854
1195	358.133	4.7288	732.2	403.149	6.8523	451.8	383.139	4.5504	354.4	403.155	4.1026
1195	363.141	5.7012	731.8	413.151	7.6310	451.5	393.141	4.9680	354.2	413.150	4.3164
1194	368.132	6.6668	731.4	423.151	8.4105	451.2	403.143	5.3759	354.0	423.162	4.5281
1194	373.141	7.6705	731.0	433.152	9.1907	451.0	413.144	5.7779	353.7	443.154	4.9454
933.9	368.142	4.4330	611.2	433.149	8.0203	450.8	423.146	6.1761	153.7	343.141	1.9855
933.7	373.149	4.9631	575.9	373.150	4.3469	450.5	433.148	6.5707	153.6	353.149	2.0954
933.2	383.150	6.0739	575.3	393.156	5.4774	450.3	443.138	6.9612	153.5	363.147	2.2030
932.7	393.153	7.2066	574.7	413.155	6.5908	376.5	443.140	6.1559	153.4	383.151	2.4122
932.2	403.161	8.3639	574.1	433.156	7.6909	355.1	363.153	3.2073	153.3	403.153	2.6161
931.7	413.160	9.5353	480.0	433.152	6.8380	354.9	373.139	3.4385	153.1	423.155	2.8156
779.0	413.149	7.9840	451.9	373.139	4.1237	354.7	383.151	3.6649	152.9	443.160	3.0120
732.9	383.148	5.3165									
(D) 80 wt % R 13B1 + 20 wt % R 114											
1188	347.152	3.6829	741.2	433.159	9.9134	353.6	403.163	4.7298	218.4	373.158	2.9719
1187	349.146	4.0338	619.7	433.155	8.6005	353.5	413.161	4.9952	218.3	383.166	3.1237
1187	353.147	4.7499	584.4	357.154	3.9173	353.3	423.159	5.2571	218.2	393.160	3.2731
1187	358.144	5.6618	584.2	363.153	4.2682	353.1	433.158	5.5159	218.1	403.160	3.4212
1186	363.145	6.5877	583.9	373.153	4.8607	352.9	443.162	5.7745	218.0	413.162	3.5675
1186	368.144	7.5237	583.7	383.153	5.4342	295.0	443.152	5.0551	217.9	423.165	3.7125
1186	373.150	8.4697	583.4	393.148	6.0003	278.6	351.152	3.0638	217.8	433.160	3.8560
1186	378.154	9.4212	583.1	403.143	6.5611	278.5	355.147	3.1572	217.7	443.165	3.9984
983.4	357.165	4.2000	582.5	423.143	7.6736	278.3	373.148	3.5632	182.0	443.160	3.4584
983.3	359.148	4.4231	581.9	443.138	8.7761	278.0	393.147	3.9956	171.9	333.147	1.7462
982.6	373.146	6.0948	486.5	443.138	7.6998	277.9	403.156	4.2055	171.8	343.152	1.8378
982.1	383.150	7.3380	458.9	359.151	3.8894	277.7	413.150	4.4161	171.7	353.155	1.9275
981.6	393.152	8.6052	458.8	363.152	4.0753	277.6	423.151	4.6244	171.6	363.150	2.0157
981.1	403.160	9.8863	458.6	373.154	4.5160	277.4	433.150	4.8274	171.5	373.151	2.1019
812.9	403.150	8.1651	458.4	383.156	4.9421	277.3	443.153	5.0307	171.5	383.151	2.1885
743.9	363.151	4.4556	458.1	393.155	5.3581	229.8	443.156	4.3514	171.4	393.146	2.2733
743.5	373.152	5.2114	457.9	403.149	5.7682	218.8	339.161	2.3595	171.3	403.147	2.3575
743.2	383.154	5.9865	457.5	423.156	6.5751	218.7	343.156	2.4861	171.2	413.141	2.4409
742.8	393.156	6.7676	457.0	443.155	7.3689	218.7	347.157	2.5618	171.1	423.147	2.5238
742.4	403.156	7.5523	354.1	373.158	3.9088	218.6	353.156	2.6590	171.0	433.149	2.6060
742.0	413.162	8.3388	354.0	383.163	4.1873	218.5	363.161	2.8172	170.9	443.156	2.6879
741.6	423.154	9.1261	353.8	393.159	4.4609						

Table II. Dew and Bubble Points for the R 13B1 + R 114 System[a]

compn of R 13B1					
wt %	mol %	ρ, kg/m^3	T, K	P, MPa	
25.00	27.67	160	383	2.31	(D)
		259	393	2.82	(D)
		356	400	3.33	(D)
		453	404	3.66	(D)
		577	406	3.86	(D)
		735	405	3.93	(B)
		936	396	3.52	(B)
		1194	370	2.38	(B)
50.00	53.44	221	374	2.70	(D)
		322	382	3.37	(D)
		457	388	3.99	(D)
		514	389	4.09	(D)
		551	389	4.17	(D)
		620	388	4.22	(D)
		665	387	4.22	(D)
		749	387	4.22	(B)
		904	380	4.01	(B)
		1091	370	3.49	(B)
70.00	72.81	154	341	1.98	(D)
		355	361	3.18	(D)
		452	369	3.94	(D)
		576	372	4.17	(D)
		733	370	4.28	(D)
		934	365	4.24	(B)
		1195	351	3.32	(B)
80.00	82.12	172	326	1.68	(D)
		219	338	2.40	(D)
		279	349	3.02	(D)
		354	355	3.36	(D)
		459	357	3.78	(D)
		584	357	3.91	(D)
		744	357	4.03	(D)
		984	355	4.00	(B)
		1188	346	3.49	(B)

[a] B, bubble point; D, dew point.

been reported upon in detail in our previous publications (*2*, *3*). The mass fraction of the sample mixture in the sample cell was determined by weighing the mass of each component on a chemical balance before mixing. The density of the sample was determined to be the ratio of the mass of the sample to the inner volume of the sample cell. The temperature of the sample was measured by a 25-Ω platinum resistance thermometer which was mounted in the vicinity of the cell in a thermostated fluid bath. The pressure of the sample was measured by dead-weight pressure gauges with the aid of a differential pressure detector. The experimental uncertainties of the temperature, pressure, density, and mass fraction measurements were estimated to be no greater than 8 mK, 2.0 kPa, 0.15%, and 0.12%, respectively.

Results

The *PVTx* measurements for four different compositions of 25, 50, 70, and 80 wt % R 13B1 were carried out along the 34 isochores from 150 to 1200 kg/m^3 in the range of temperatures 303–443 K and of pressures 0.5–10.3 MPa. The results in the single-phase region are tabulated in Table I. Those in the two-phase region of vapor–liquid coexistence are available in the supplementary material. (See Supplementary Material Available paragraph at the end of the text.) Because the results of the measurements for both pure components of R 114 and R 13B1 were reported in previous publications (*2*, *4*), we did not repeat the measurements for these pure components in the present work. Thirty-four isochores measured were carefully examined with respect to the curvature on a pressure–temperature diagram. The breaking point in the isochore was regarded as the dew or bubble point. In the case of the isochores whose breaking points were hardly found, each measurement along the respective isochores was carefully distinguished by the state condition, whether in the two-phase region or in the single-phase region. The temperature of dew or bubble point for these isochores was then determined to be the intermediate value of two neighboring measurements; one is in the two-phase region and the other in the single-phase region. The dew and bubble points for each composition are tabulated in Table II. Molar masses of 148.910 g/mol for R 13B1 and 170.922 g/mol for R 114 were used for converting mass fraction into mole fraction. The mole fraction of each composition is also shown in Table II. The uncertainty in temperature of the dew and bubble points determined by the breaking point on the isochore is estimated to be 1 K, although the uncertainty of the rest is larger by the factor of 3 or more.

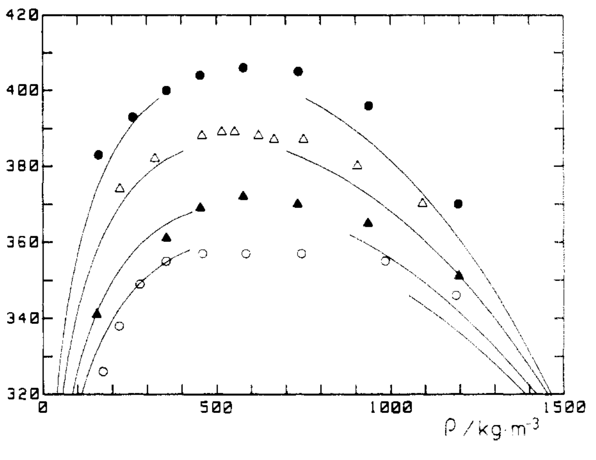

Figure 1. Coexistence curves of the R 13B1 + R 114 system: ●, our results for 25 wt % R 13B1; △, our results for 50 wt % R 13B1; ▲, our results for 70 wt % R 13B1; ○, our results for 80 wt % R 13B1; —, calculated by Peng-Robinson eq (*6*) with $k_{ij} = 0$.

Our careful observation found the difference of time duration for the sample mixture in the cell to reach a state of thermodynamic equilibrium at each measurement due to the sample state whether the sample was in the two-phase region of vapor–liquid coexistence or in the single-phase region. The sample state was monitored by measuring pressure variation of the sample after the bath temperature approached the prescribed temperature. When the sample was in the single-phase region, it took 0.5–1 h until the pressure became stable and constant, whereas it required more than 3 h for the measurements in the two-phase region. The time duration to reach thermodynamic equilibrium increased as the sample state was close to the dew or bubble point. The monitored pressure values for low densities in the two-phase region increased as the thermodynamic equilibrium state was approached but decreased for high densities.

Discussion

The envelopes of dew and bubble points of the present system are the widest among those of the other two systems we measured, i.e., the R 12 + R 22 system (*1*) and the R 22 + R 114 system (*2*). Three different cubic equations of state by Soave (*5*), by Peng–Robinson (*6*), and by Patel–Teja (*7*) were fitted to the present results. These three equations describe roughly the envelopes of dew and bubble points measured, and no significant difference among them was found. Introduction of the binary interaction parameter, k_{ij}, into these equations for better overall description of the envelopes of four compositions measured described the dew and bubble points of one composition better, but those of other compositions worse. Therefore an optimum value of k_{ij} for each equation examined was not determined. The behavior of coexistence curves calculated by these three equations was also examined by projecting onto a temperature–density diagram. None of these equations represents well the density dependence of the present system. Figure 1 shows the comparison of the coexistence curves for four compositions on the temperature–density diagram between measured values and calculated values by the Peng–Robinson equation with $k_{ij} = 0$ as a typical example. The effect of introducting k_{ij} was contradictory for each composition as mentioned above, and it was not efficient to improve description of liquid density. We might conclude that the cubic equations of state would not be good enough to reproduce the density values measured particularly for the liquid state.

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Registry No. R 13B1, 75-63-8; R 114, 76-14-2.

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Supplementary Material Available: Complete form of Table I including the experimental data in the two-phase region (22 pages). Ordering information is given on any current masthead page.

Measurements of the Vapor–Liquid Coexistence Curve for the R 13B1 + R 114 System in the Critical Region

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Measurements of the vapor–liquid coexistence curve in the critical region for the refrigerant mixture of bromotrifluoromethane ($CBrF_3$, R 13B1) and 1,2-dichloro-1,1,2,2-tetrafluoroethane ($CClF_2CClF_2$, R 114) were made by visual observation of the disappearance of the meniscus at the vapor–liquid interface within an optical cell. Eighteen saturated-vapor densities and 21 saturated-liquid densities for four different compositions of 25, 50, 70, and 80 wt % R 13B1 between 345 and 406 K were obtained in the range of densities 356–1166 kg/m^3. The experimental error of temperature, density, and mass fraction was estimated within ±15 mK, ±0.5%, and ±0.05%, respectively. On the basis of these measurements, the critical curve of the R 13B1 + R 114 system is determined and compared with several predictive methods. In addition, new correlation so as to represent the composition dependence of the critical parameters for the R 13B1 + R 114 system is proposed.

Introduction

Nonazeotropic refrigerant mixtures, so-called NARBs (nonazeotropic refrigerant blends), of two or three kinds of halogenated hydrocarbons have recently been of interest as prospective working fluids in the reversed Rankine cycle systems. For example, Cooper et al. (*1*) showed that the R 13B1 (bromotrifluoromethane, $CBrF_3$) + R 152a (1,1-difluoroethane, CH_3CHF_2) system was an attractive choice to replace R 22 (chlorodifluoromethane, $CHClF_2$) in an air-to-air heat pump with an accumulator and capillary tube expander, and that the R 13B1 + R 152a system supplied approximately 40% more heating capacity than R 22 in a 7-kW heat pump. Moreover, the low-temperature medical freezer using the ternary refrigerant mixture of the R 502 (azeotropic mixture of R 22 + R 115 ($CClF_2CF_3$, chloropentafluoroethane)) + R 12 (dichlorodifluoromethane, CCl_2F_2) system as well as the ternary refrigerant mixture of the R 21 (dichlorofluoromethane, $CHCl_2F$) + R 13B1 + R 14 (tetrafluoromethane, CF_4) system as a working fluid is commercialized (*2*).

We have been carrying out investigations on the thermophysical properties of several refrigerant mixtures. Some of the results were published: measurements of the vapor–liquid coexistence curve in the critical region and the critical curve for the R 12 + R 22 system (*3*) and the R 22 + R 114 system (*4*), the *PVTx* measurements of the R 12 + R 22 system (*5*–*7*), the R 22 + R 114 (1,2-dichloro-1,1,2,2-tetrafluoroethane, $CClF_2CClF_2$) system (*8*), and the R 13B1 + R 114 system (*9*). In the present paper, measurements of the vapor–liquid coexistence curve and the critical curve for the R 13B1 + R 114 system in the critical region are reported, and the assessment of the predictive methods of the critical parameters is also discussed on the basis of the experimental results of the critical parameters for refrigerant mixtures. The correlations of the composition dependence of the critical parameters are proposed.

Experimental Section

The vapor–liquid coexistence curve in the critical region and the critical point for a certain composition of binary refrigerant mixtures were measured by observing the meniscus disappearance. The experimental apparatus used and procedures were described in detail in previous publications (*10*, *11*). The apparatus was composed of an optical cell, an expansion vessel, and a supplying vessel. In order to measure the saturation temperature successively for a series of densities along the coexistence curve, we introduced an expansion technique (*10*) throughout the measurements. The optical cell was connected with the expansion vessel to repeat the expansion procedures of the sample fluid from the optical cell to the ex-

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