Vapor-Liquid Equilibrium Relationships of Binary Systems Propane–*n*-Octane and *n*-Butane–*n*-Octane

Webster B. Kay,[1] Joseph Genco,[2] and David A. Fichtner[3]
Department of Chemical Engineering, The Ohio State University, Columbus, Ohio 43210

The *P-ρ-T-x* relationships of the propane–*n*-octane and the *n*-butane–*n*-octane systems are determined. The experimental results cover a range from about 150 lb/in.^{2} abs and room temperature to the highest pressure and temperature at which liquid and vapor can coexist. The data are presented in tabular form. *P-T-x*, *ρ-T-x*, and isobaric *T-x* diagrams are given.

This paper presents the *P-ρ-T-x* relationships of the binary systems of propane–*n*-octane and *n*-butane–*n*-octane. It is the third of a series (*5*, *6*) in a study of the effect of the relative size of the molecules on the phase diagram of mixtures of *n*-alkanes.

Experimental

The apparatus and experimental procedure for the determination of the *P-ρ-T-x* phase diagrams of binary systems at elevated temperatures and pressures were the same as described in previously published papers (*5*, *6*).

A small air-free sample was contained over mercury in the sealed end of a precision bore glass capillary tube, hereafter termed the experimental tube, which was surrounded by a constant-temperature bath. The tube was fastened in a compressor block which was essentially a mercury-in-steel U-tube. This, in turn, was connected through a manifold to a high-pressure gas cylinder. By admitting nitrogen gas to the system, pressure was applied via the mercury to the sample in the capillary.

For the measurement of the pressure, a sensitive precision spring gauge with a 16-in. dial marked in 2-lb divisions, was attached to the manifold. The gauge had a sensitivity of 0.2 lb/in.^{2} and could be read accurately to 0.5 lb/in.^{2}. The constant-temperature bath surrounding the experimental tube consisted of a double-walled, silvered, and evacuated glass jacket with viewing slits and with a side-arm boiling flask attached below the double wall. By refluxing the vapors of a series of organic compounds of different boiling points contained in the flask and by controlling the pressure over the boiling liquid, a range of temperature, constant to within 0.02°C, could be obtained.

The temperature was measured by means of an iron-constantan thermocouple in conjunction with a sensitive potentiometer capable of reading the millivolt equivalent

[1] To whom correspondence should be addressed.
[2] Present address, Battelle Memorial Institute, Columbus, Ohio 43201.
[3] Present address, Goodyear Tire & Rubber Co., Akron, Ohio 44305.

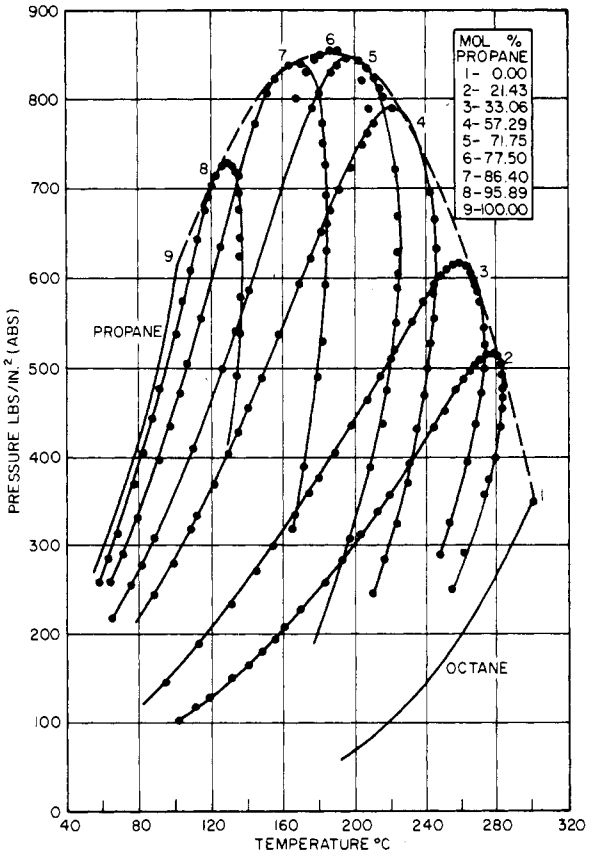

Figure 1. Pressure-temperature diagram of propane–*n*-octane system

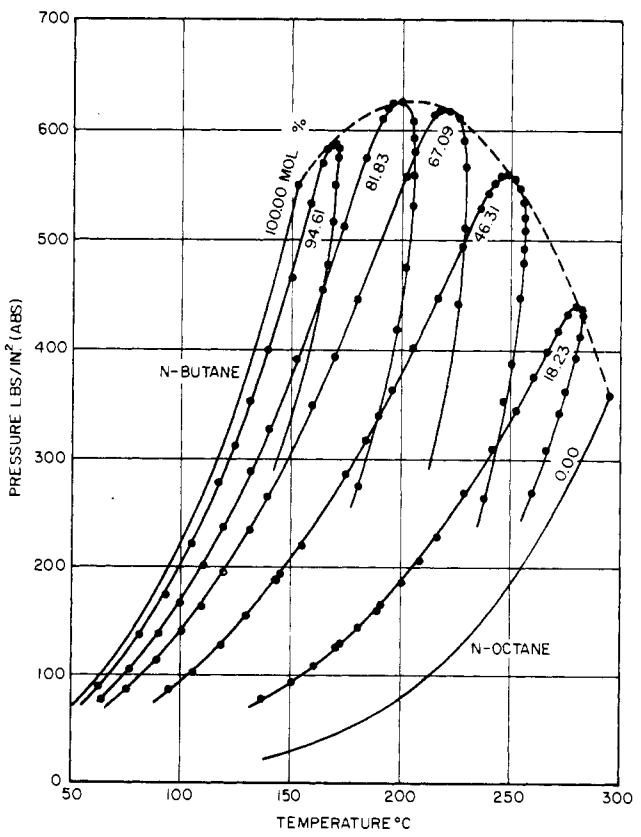

Figure 2. Pressure-temperature diagram of *n*-butane–*n*-octane system

of 0.005°C. Both the pressure gauge and the thermocouple were calibrated; the former by comparison with a calibrated dead-weight gauge and the latter by comparison with a platinum resistance thermometer which had been calibrated by the National Bureau of Standards. A previous calibration of the experimental tube made it possible to determine the volume of the sample by measuring the length of the tube which it occupied to within 0.02 mm by means of a cathetometer. The total volume of the tube was expressed analytically as a function of the distance from the sealed end.

The coefficients of the equation were determined by a least-squares procedure with experimental values of the mass of mercury required to fill the tube to various levels.

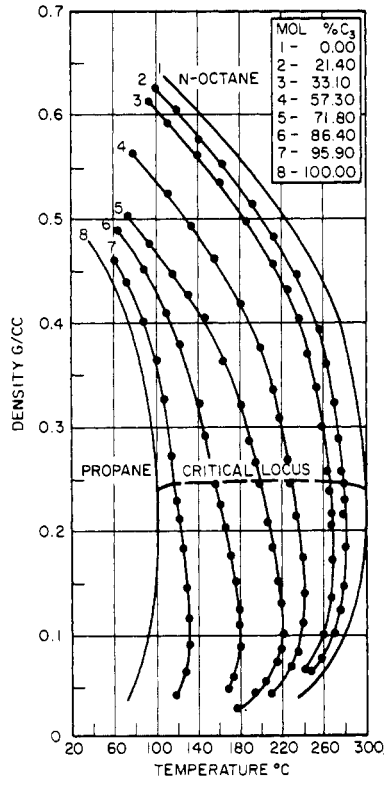

Figure 3. Density-temperature diagram of propane–*n*-octane system

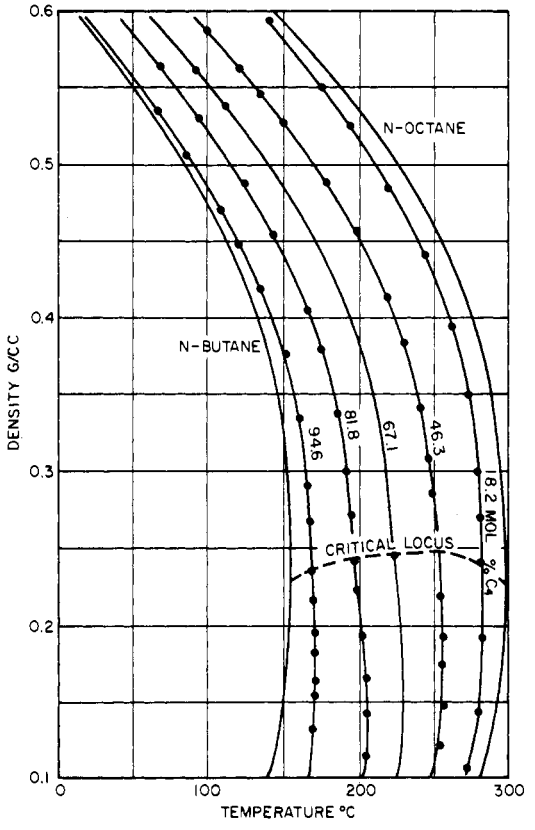

Figure 4. Density-temperature diagram of *n*-butane–*n*-octane system

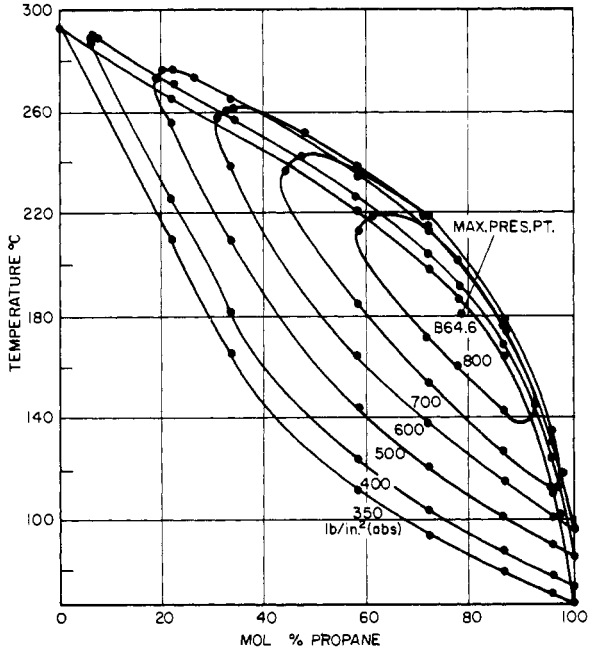

Figure 5. Isobaric temperature-composition diagram of propane–*n*-octane system

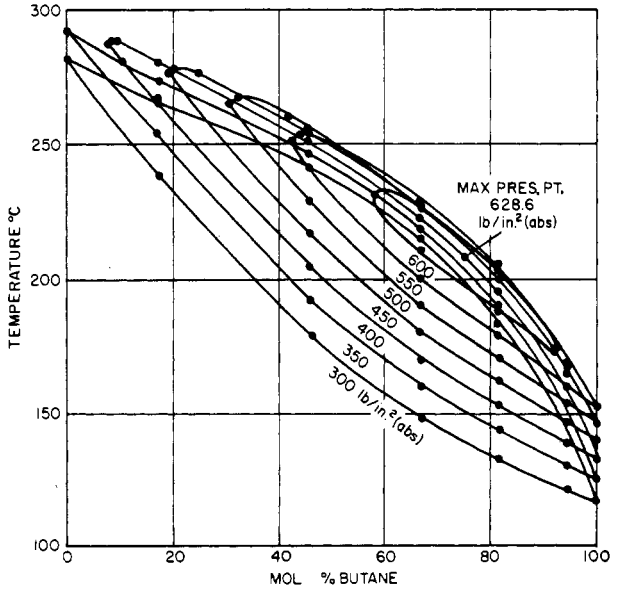

Figure 6. Isobaric temperature-composition diagram of *n*-butane–*n*-octane system

Equilibrium between the liquid and vapor phases was attained by moving a small steel ball enclosed in the tube, by means of a magnet around the outside of the jacket. Pressure and volume were measured at a series of constant temperatures covering the range desired.

Materials and Preparation of Mixtures

The propane, *n*-butane, and *n*-octane were supplied with a purity of better than 99.90 mol % by the Phillips Petroleum Co. They were used without further purification except for the removal of air. Deaeration was accomplished by a series of operations which involved freezing the sample with liquid nitrogen and pumping off the residual gas over the solid, followed by melting the solid sample and distilling it at low pressure.

Measured amounts of the degassed propane and *n*-butane were then transferred to glass ampuls, sealed, and stored for use in the preparation of mixtures with *n*-octane. The procedure for preparing and loading the experimental tube with a mixture of known composition has

Table I. Summary of Temperature, Pressure, and Density Relationships at Phase Boundaries for Propane–*n*-Octane System: Data by Genco (*3*)

	Liquid		Vapor	
Press, lb/in.² abs	Temp, °C	Density, g/cc	Temp, °C	Density, g/cc
	Composition: 21.43 mol % propane			
100	98.0	0.628	...	...
150	127.5	0.592	...	...
200	153.0	0.561	...	...
250	174.0	0.533	247.4	0.0642
300	194.0	0.508	257.0	0.0799
350	210.0	0.481	265.7	0.0971
400	227.0	0.453	272.5	0.1200
450	242.5	0.418	277.3	0.1590
500	258.0	0.374	276.7	0.2199
510	262.0	0.357	276.0	0.2399
520	266.5	0.333	...	...
520	274.0	0.270	...	...
525.8	270.0	0.317	...	...
	Composition: 33.06 mol % propane			
150	92.5	0.611	...	...
200	113.0	0.590	...	...
250	132.5	0.566	...	...
300	149.5	0.545	243.5	0.0700
350	166.0	0.529	251.0	0.0820
400	181.5	0.502	257.5	0.0999
450	196.0	0.480	263.0	0.1180
500	210.0	0.455	266.5	0.1499
550	224.0	0.427	266.5	0.1699
600	238.9	0.385	262.5	0.247
610	242.7	0.370	...	...
610	260.5	0.270	...	...
620	247.0	0.355	...	...
620.0	258.0	0.293	...	...
626.7	253.0	0.328	...	...
	Composition: 57.29 mol % propane			
200	71.8	0.568	...	...
250	87.0	0.550	206.0	0.0400
300	100.5	0.533	214.5	0.0490
350	112.0	0.518	222.0	0.0580
400	122.5	0.504	227.5	0.0690
450	134.0	0.489	232.0	0.0780
500	144.2	0.474	235.5	0.0910
550	154.3	0.460	238.3	0.1110
600	164.5	0.450	239.7	0.1200
650	174.5	0.425	240.0	0.1300
700	185.0	0.405	238.2	0.1749
750	196.5	0.376	232.5	0.2069
770	202.0	0.360	229.0	0.2300
780	205.0	0.350	...	...
780	225.5	0.258	....	...
790	208.5	0.337	...	...
790	222.0	0.276	...	...
784	215.3	0.311	...	...

	Liquid		Vapor	
Press, lb/in.² abs	Temp, °C	Density, g/cc	Temp, °C	Density, g/cc
	Composition: 71.75 mol % propane			
200	56.0	0.519	177.0	0.0280
250	70.5	0.501	185.6	0.0349
300	83.5	0.487	193.0	0.0420
350	94.3	0.474	199.0	0.0500
400	104.0	0.462	205.0	0.0561
450	113.0	0.450	211.0	0.0600
500	121.2	0.440	215.6	0.0780
550	129.5	0.427	218.5	0.0879
600	137.5	0.415	219.6	0.1020
650	145.6	0.403	219.6	0.1080
700	154.0	0.387	218.9	0.1200
750	162.2	0.371	216.5	0.1450
800	171.1	0.348	212.0	0.1631
820	176.0	0.332	208.3	0.1940
840	182.3	0.312	202.8	0.2199
850	185.5	0.293	...	...
850	198.0	0.247	...	...
854.8	193.2	0.268	...	...
	Composition: 86.40 mol % propane			
300	69.0	0.480	...	...
350	78.5	0.468	164.5	0.0429
400	86.7	0.454	168.8	0.0481
450	94.4	0.439	172.5	0.0570
500	101.4	0.425	175.5	0.0660
550	108.6	0.411	177.8	0.0750
600	114.8	0.395	179.3	0.0870
650	121.5	0.377	179.5	0.1049
700	128.0	0.358	179.5	0.1049
750	134.9	0.335	178.4	0.1200
800	143.0	0.305	176.1	0.1450
820	147.7	0.280	174.5	0.1560
840	...	...	170.0	0.1760
840	...	...	115.0	0.2439
848	...	...	162.8	0.2090
	Composition: 95.89 mol % propane			
300	61.6	0.454	...	...
350	70.0	0.439	115.0	0.0370
400	77.1	0.423	122.6	0.0481
450	83.5	0.409	127.5	0.0629
500	89.8	0.392	130.0	0.0770
550	95.9	0.375	131.8	0.1049
600	101.8	0.353	132.1	0.1100
650	107.4	0.319	131.9	0.1089
700	114.0	0.220	130.4	0.1749
710	115.8	0.250	130.0	0.1800
720	...	...	118.1	0.2300
720	...	...	129.2	0.1499
730	...	...	120.3	0.2149
730	...	...	128.0	0.1600
738.4	.....	.....	124.9	0.1829

Table II. Summary of Temperature, Pressure, and Density Relationships at Phase Boundaries for *n*-Butane–*n*-Octane System: Data by Fichtner (*2*)

Press, lb/in.²abs	Liquid Temp, °C	Liquid Density, g/cc	Vapor Temp, °C	Vapor Density, g/cc
	Composition: 18.23 mol % *n*-Butane			
100	153.7	0.573	...	...
150	182.7	0.536	...	...
200	205.1	0.504	...	...
250	223.3	0.475	...	...
300	238.9	0.446	264.7	...
350	253.5	0.412	273.0	0.110
400	266.5	0.372	280.5	0.150
420	271.8	0.348	282.3	0.179
430	274.5	0.332	282.3	0.219
440	277.7	0.307	281.0	0.256
	Composition: 46.31 mol % *n*-butane			
100	102.7	0.579	...	...
150	127.2	0.551	...	...
200	146.8	0.527	...	...
250	163.6	0.506	...	...
300	178.3	0.485	242.0	...
350	192.1	0.463	246.6	...
400	204.8	0.440	250.8	...
450	216.8	0.412	254.1	0.124
500	228.7	0.380	256.3	0.157
520	233.6	0.364	256.4	0.175
540	238.8	0.345	255.2	0.199
550	241.5	0.332	253.5	0.228
555	243.2	0.332	251.9	0.248
560	245.5	0.309	250.0	0.270
	Composition: 67.09 mol % *n*-butane			
100	81.3	0.569	...	...
150	103.2	0.545	...	...
200	120.4	0.525	...	...
250	135.0	0.506	...	...
300	148.0	0.487	214.1	...
350	159.7	0.467	218.7	...
400	170.5	0.447	222.6	...
450	180.6	0.426	226.2	...
500	190.5	0.402	228.2	0.176
550	200.2	0.347	229.4	...
600	210.9	0.331	227.5	0.186
610	213.2	0.319	225.8	0.207
615	214.5	0.311	223.9	0.231
	Composition: 81.83 mol % *n*-butane			
100	73.8	0.551	...	...
150	93.3	0.526	...	...
200	108.8	0.504	...	...
250	121.6	0.483	...	...
300	133.5	0.463	183.8	0.050
350	143.8	0.444	190.4	0.060
400	153.5	0.425	195.9	0.072
450	162.4	0.405	200.2	0.085
500	170.9	0.384	203.2	0.099
550	179.3	0.357	205.4	0.128
600	188.5	0.313	205.4	0.157
610	190.5	0.301	204.8	0.167
620	193.0	0.282	203.0	0.189
625	195.0	0.255	201.0	0.207
	Composition: 94.61 mol % *n*-butane			
100	66.3	0.527	...	...
150	84.5	0.501	...	...
200	98.7	0.479	...	...
250	110.6	0.4595	...	...
300	121.1	0.440	...	...
350	130.3	0.421	151.6	...
400	138.8	0.402	158.5	...
450	146.5	0.381	163.9	...
500	153.7	0.356	167.4	0.231
550	160.6	0.319	169.4	0.195
570	163.4	0.296	170.2	0.175
580	165.1	0.275	170.4	0.159
585	166.3	0.254	170.0	0.143
587.3	166.85	0.237	166.85	0.237
588.5	167.5			
589.8	168.7			

been described elsewhere (*7*, *8*). The purity of each of the components was checked by measuring the difference in the pressure between the isothermal dew and bubble points. This difference was always less than 2.0 lb/in.².

Equilibrium Data

The pressure, temperature, and volume at the bubble and dew points of five mixtures each of propane–*n*-octane and *n*-butane–*n*-octane were measured over a temperature range from room temperature to the highest temperature at which liquid and vapor could coexist. For each mixture, 25–35 data points were determined. In the compilation of the data, the measured specific volumes were expressed as densities. The experimental points were plotted, and the *P-T* border curves and the density-temperature curves are shown in Figures 1–4.

The vapor pressure and saturated density curves of the pure components were constructed by use of data taken from the literature (*1*, *4*, *9*). Large-scale plots of these diagrams were constructed from which values of the temperature and density at the bubble and dew points were read at regular intervals of the pressure. These values are listed in Tables I and II. *T-x* data at different pressures were obtained from cross plots of Figures 1 and 2, and are listed in Tables III and IV, where the temperatures at the bubble and dew points at regular intervals of the composition are given. Isoberic temperature-composition curves are shown in Figures 5 and 6. From the *T-x* diagrams, the vapor-liquid equilibrium ratios, $K = y/x$, for each of the components were derived and are given in Tables V and VI.

The critical point was determined visually by the disappearance-of-the-meniscus method, whereas the coördinates of pressure, temperature, and density at the maximum pressure and maximum temperature points were obtained graphically from large-scale plots of the *P-T* and *ρ-T* border curves in the critical region of the mixture. The critical constants of the pure compounds and mixtures and the coordinates of the maximum pressure (cricondenbar) and maximum temperature (criconten-therm) points of each system are listed in Tables VII and VIII. The coordinates of the maximum pressure point on the *P-T* critical locus curve are as follows: Propane–*n*-octane: $P = 864 \pm 1.0$ lb/in.² abs; $T = 182.6 \pm 0.5$°C; mol % propane, 77.5 ± 0.3. *n*-Butane–*n*-octane: $P = 628.6 \pm 1.0$ lb/in.² abs; $T = 208.6 \pm 0.2$°C; mol % *n*-butane, 75.4 ± 0.3.

Table III. Isobaric Temperature-Composition Relations of Propane–n-Octane System

Compn, mol % propane	Temp, °C					
	Press, lb/in.² abs					
	350		400		500	
	Liquid	Vapor	Liquid	Vapor	Liquid	Vapor
0	293.8	293.8	...	...	...	...
5.0	...	...	291.0	291.0	...	...
10	254.9	280.8	271.3	286.1	...	...
18.0	...	...	...	...	273.0	273.0
20	216.0	267.8	232.6	273.7	262.6	277.8
30	177.2	256.3	193.8	261.4	222.2	269.6
40	146.0	244.4	160.8	249.1	186.4	258.2
50	125.0	232.2	138.2	236.8	159.9	245.9
60	108.8	218.8	120.0	224.0	140.1	232.6
70	95.8	202.2	106.1	208.2	123.5	218.0
80	84.8	181.9	94.5	186.8	109.1	195.4
90	75.0	150.4	83.6	156	96.7	161.8
100	66.4	66.4	73.0	73.0	85.6	85.6
	600		700		800	
	Liquid	Vapor	Liquid	Vapor	Liquid	Vapor
29.8	255.0	255.0	...	...	...	...
30	252.4	257.6	...	...	...	...
40	210.4	260.3	...	...	...	...
43.1	...	...	233.2	233.2	...	...
50	182.2	249.0	205.6	243.6	...	...
57.8	...	...	...	...	212.0	212.0
59	...	...	...	...	203.6	218.0
60	159.4	236.1	179.8	235.4	200.0	219.5
70	140.6	222.3	157.4	221.5	174.9	216.2
80	124.0	200.0	138.2	200.8	155.0	196.4
85	...	...	...	...	145.8	181.3
90	108.9	166.6	120.2	166.2	137.2	162.6
91	...	...	...	...	136.4	157.6
92.6	...	...	...	...	143.0	143.0
95	...	...	112.0	137.6	...	...
97.7	...	...	116.0	116.0	...	...
100	95.0	95.0	...	...	...	...

Table IV. Isobaric Temperature-Composition Relations of n-Butane–n-Octane System

Compn, mol % butane	Temp, °C					
	Press, lb/in.² abs					
	300		350		400	
	Liquid	Vapor	Liquid	Vapor	Liquid	Vapor
0	282.4	282.4	293.6	293.6		
8.6	...	...	...	...	287.6	287.6
10	258.0	272.5	271.6	282.2	283.5	288.5
20	234.4	263.2	249.6	271.2	262.2	278.7
30	211.3	255.0	227.0	261.8	239.0	268.2
40	190.2	247.0	205.3	252.4	217.6	257.9
50	172.0	238.0	185.4	242.8	197.8	246.1
60	156.8	225.3	169.2	229.6	181.0	233.0
70	144.8	209.0	156.4	213.2	166.8	218.0
80	134.8	188.0	145.8	194.0	155.2	199.8
90	125.8	161.0	135.2	167.8	144.2	174.4
100	116.4	116.4	125.0	125.0	133.0	133.0
	450		500		550	
	Liquid	Vapor	Liquid	Vapor	Liquid	Vapor
20	277.0	277.0	...	...	...	...
22.0	271.4	278.4	...	...	...	...
30	252.5	272.6	...	...	...	...
31.2	...	...	266.0	266.0	...	...
32.0	...	...	263.2	267.6	...	...
40	230.1	261.5	242.8	262.8	...	...
43.1	...	...	...	...	251.2	251.2
45.0	...	...	...	...	244.4	253.8
50	209.2	249.6	220.8	251.7	233.0	251.2
60	191.2	236.6	202.0	238.4	212.8	239.9
70	176.6	221.5	186.5	223.6	195.6	225.0
80	164.4	203.6	173.2	206.6	181.6	208.5
90	152.2	180.0	160.0	182.4	167.8	184.4
100	140.0	140.0	146.4	146.4	152.0	152.0
	600					
	Liquid	Vapor				
58.7	231.5	231.5				
60	226.2	233.0				
70	205.8	223.8				
80	191.1	208.5				
90	176.0	186.2				
92	173.2	179.2				
92.8	174.0	174.0				

The accuracy of the tabulated data is estimated to be as follows: temperature, ±0.5°C; pressure, ±2.0 lb/in.²; density, ±0.001 g/cc for the liquid; and ±0.0001 g/cc for the vapor. In the critical region, the uncertainties in the values reported may be somewhat greater because of the difficulty in assessing the accuracy of the measurements in this region.

The unsmoothed experimental data have been deposited with the ACS Microfilm Depository Service.

Acknowledgment

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Literature Cited

(1) Deschner, W., Brown, G. G., *Ind. Eng. Chem.*, **32**, 836 (1940).
(2) Fichtner, David, MS thesis, Ohio State University, Columbus, Ohio, 1962.
(3) Genco, Joseph, MS thesis, Ohio State University, Columbus, Ohio, 1962.
(4) Kay, W. B., *Ind. Eng. Chem.*, **32**, 358 (1940).
(5) Kay, W. B., *J. Chem. Eng. Data*, **15**, 46 (1970).
(6) Kay, W. B., *ibid.*, **16**, 137 (1971).
(7) Kay, W. B., Donham, R., *Chem. Eng. Sci.*, **4**, 1 (1955).
(8) Kay, W. B., Rambosek, G. M., *Ind. Eng. Chem.*, **45**, 221 (1953).
(9) McMicking, J. H., Kay, W. B., *Proc. Amer. Petrol. Inst., Sect. III*, **45** (3), 75 (1965).

Table V. Vapor-Liquid Equilibrium Ratios of Propane–n-Octane System

Press, lb/in.² abs	Temp, °C					
	90	100	110	120	150	200
	$K_i = y_i/x_i$ for propane					
350	1.30	1.45	1.62	1.79	2.34	2.94
400	1.18	1.30	1.45	1.60	2.05	2.61
500	1.04	1.13	1.24	1.34	1.69	2.18
600	...	1.04	1.11	1.18	1.44	1.85
700	...	...	...	1.08	1.26	1.54
800	000	...	...	...	1.11	1.31
	$K_j = y_j/x_j$ for n-octane					
350	0.0843	0.0928	0.103	0.114	0.161	0.38
400	0.0688	0.0729	0.0818	0.0952	0.155	0.354
500	0.0714	0.0859	0.0922	0.108	0.167	0.339
600	...	0.105	0.120	0.132	0.190	0.353
700	...	...	...	0.253	0.263	0.411
800	...	...	...	...	0.457	0.538

Table VI. Vapor-Liquid Equilibrium Ratios of *n*-Butane–*n*-Octane System

Press, lb/in.² abs	Temp, °C						Press, lb/in.² abs	Temp, °C					
	150	175	200	225	250	275		150	175	200	225	250	275
	$K_i = y_i/x_i$ for *n*-butane							$K_j = y_j/x_j$ for *n*-octane					
300	1.425	1.77	2.09	2.51	2.78	2.55	300	0.201	0.284	0.410	0.600	0.726	0.954
350	1.250	1.56	1.81	2.03	2.16	1.95	350	0.215	0.280	0.398	0.536	0.714	0.913
400	1.136	1.40	1.64	1.79	1.86	1.66	400	0.238	0.281	0.395	0.543	0.711	0.892
450	1.070	1.29	1.48	1.60	1.595	1.35	450	0.265	0.289	0.404	0.557	0.730	0.910
465.5	...	...	...	...	...	1.000	465.5	...	...	...	...	...	1.000
500	1.020	1.19	1.36	1.445	1.385		500	0.300	0.310	0.433	0.591	0.773	...
531.0	1.000	...	...	...	...	...	531.0	1.000	...	...	...	...	...
550	...	1.09	1.25	1.30	1.19		550	...	0.404	0.479	0.648	0.855	
560	...	...	...	...	1.000		560	...	...	...	...	1.000	...
600	...	1.02	1.15	1.145			600	...	0.678	0.578	0.781	...	...
602.6	...	1.000					602.6	...	1.000	...	...	...	...
613.8	...	...	...				613.8	...	...	...	1.000	...	...
627.5	...	...	1.000	1.000	...	...	627.5	...	...	1.000	...	...	...

Table VII. Critical Constants of Propane–*n*-Octane System

Mol %, C_3	Critical point			Point of max press			Point of max temp		
	T_c, °C	P_c, lb/in.² abs	Density, g/cc	$T_{P_{max}}$, °C	$P_{P_{max}}$, lb/in.² abs	Density, g/cc	$T_{T_{max}}$, °C	$P_{T_{max}}$, lb/in.² abs	Density, g/cc
0	295.6[a]	360.7[a]	0.232[a]	...	...	...	...	...	...
21.4	274.8	514.0	0.257	270	525.8	0.321	278.2	479	0.186
33.1	262.1	601.7	0.247	253	626.7	0.325	266.9	537	0.169
57.3	226.1	778.1	0.248	215	798.4	0.310	240.1	639	0.139
71.8	198.0	849.9	0.240	192	854.8	0.264	219.5	678	0.0996
77.5	182.6	864.5	0.247	183	864.6	0.253	...	...	...
86.4	154.7	839.2	0.245	164	848.0	0.211	179.8	669	0.106
95.9	116.8	714.9	0.245	125	738.4	0.182	132.2	646.2	0.104
100	96.87[b]	617.9[b]	0.266[b]	...	...	...	...	...	...

[a] Ref. 9. [b] Ref. 5.

Table VIII. Critical Constants of *n*-Butane–*n*-Octane System

Compn, mol % *n*-butane	Critical point			Point of max press			Point of max temp		
	T_c, °C	P_c, lb/in.² abs	Density, g/cc	$T_{P_{max}}$, °C	$P_{P_{max}}$, lb/in.² abs	Density, g/cc	$T_{T_{max}}$, °C	$P_{T_{max}}$, lb/in.² abs	Density, g/cc
0	295.6[a]	360.7[a]	0.232[a]	...	...	...	...	...	...
18.23	280.89	437.9	0.2398	278.83	442.1	0.2811	282.49	426.9	0.1914
46.31	251.80	555.1	0.2480	248.09	562.1	0.2854	256.51	511.3	0.1626
67.07	221.89	618.3	0.2463	219.51	619.6	0.2670	229.82	565.9	...
81.83	195.42	625.6	0.2423	198.72	627.0	0.2238	205.74	582.1	0.1413
94.61	166.85	587.3	0.2369	168.86	589.9	0.2045	170.53	575.1	0.1584
100	152.2[b]	550.5[b]	0.228[b]	...	...	...	...	...	...

[a] Ref. 9. [b] Ref. 4.

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Supplementary Material Available. An Appendix containing two tables of unsmoothed experimental data will appear following these pages in the microfilm edition of this volume of the journal. Photocopies of the supplementary material from this paper only or microfiche (105 × 148 mm, 24× reduction, negatives) containing all of the supplementary material for the papers in this issue may be obtained from the Journals Department, American Chemical Society, 1155 16th St., N.W., Washington, D.C. 20036. Remit check or money order for $4.00 for photocopy or $2.00 for microfiche, referring to code number JCED-74-275.