

establishing which of the infinite dilution coefficients should be the largest. An exact matching of this relationship was not expected since no data were obtained in the dilute C₄-unsaturated region. The vinyl acetylene-*cis*-butene-2 and vinyl acetylene-1,3-butadiene binaries adhered to the above relationship, whereas the vinyl acetylene-*trans*-butene-2 did not. Attempts to incorporate Equation 13 as a constraint in the *trans*-butene-2 system proved unsuccessful, the resulting *F* ratio being 15 times larger than with the original Wilson parameters (although the average % relative error in calculating the vapor composition only increased from 1 to 2.6 %).

Regular solution theory predicts that the temperature variation of infinite dilution activity coefficients can be obtained from

$$\gamma_1^\infty = \gamma_2^\infty \left(\frac{v_1 T_2}{v_2 T_1} \right) \quad (14)$$

For all three binaries, the Wilson equations were in agreement with regular solution theory in predicting the temperature dependence of the infinite dilution activity coefficients.

Prediction of binary azeotropes. The Wilson equations were used to generate relative volatilities for each binary. These are presented in Figure 3 for a temperature of 110.5°F. This plot shows that at 34 mol %, vinyl acetylene forms a low-boiling azeotrope with *trans*-butene-2. Similarly at 46 mol %, vinyl acetylene forms a low-boiling azeotrope with *cis*-butene-2. Note that these predicted azeotropes are outside the range of the experimental data. A degree of uncertainty exists as to the actual composition of the azeo-

tropes, since, experimentally, little information was generated for the activity coefficients of the C₄-unsaturates in vinyl acetylene.

Acknowledgment

This work was performed at Phillips Petroleum Co., and the author is indebted to them for permission to publish these results.

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Vapor-Liquid Equilibrium Relationships of Binary Systems *n*-Butane-*n*-Pentane and *n*-Butane-*n*-Hexane

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The *P*-*ρ*-*T*-*x* relationships of the *n*-butane-*n*-pentane and *n*-butane-*n*-hexane systems are determined. The experimental results cover a range from about 100 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 experimental study reports the *P*-*ρ*-*T*-*x* relationships along the liquid-vapor phase boundaries of the binary systems of *n*-pentane and *n*-hexane with *n*-butane as the common component. It is the fourth of a series of studies (4-6) whose primary object has been to explore the effect of the relative size of the components of a binary system on its phase diagram in the liquid-vapor region.

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Experimental

The *P*-*ρ*-*T*-*x* relationships of the two *n*-butane-*n*-alkane systems were obtained by the determination of the *P*-*T* and *V*-*T* border curves of a series of mixtures of known composition. The relationships between any set of the variables were then obtained by appropriate cross plots of these curves.

The apparatus and experimental procedures employed were the same as those employed in the studies previously reported (4-6). Very briefly, the operation consisted in confining a measured amount of an air-free sample over mercury in a glass capillary tube, hereafter referred to as the experimental tube, which was fastened in one leg of a mercury-in-steel, U-tube. The other leg was connected through a manifold to a compressed gas cylinder which served as a source of pressure and to a sensitive spring gauge for the measurement of the applied pressure. The tube was surrounded by a constant-temperature jacket whose temperature was measured by means of a copper-constantan thermocouple in conjunction with a potentiometer. By a precalibration of the experi-

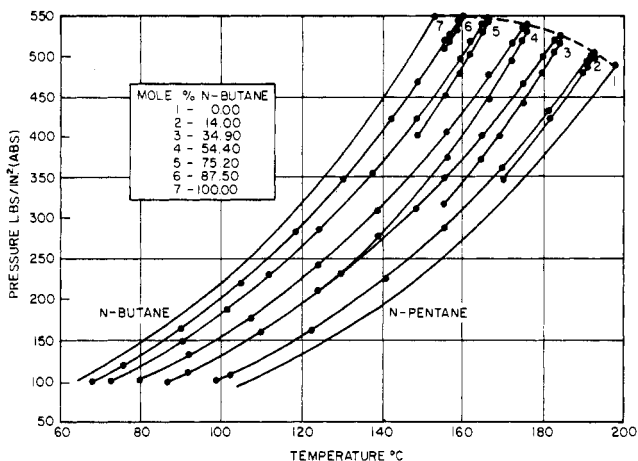


Figure 1. Pressure-temperature diagram of *n*-butane-*n*-pentane system

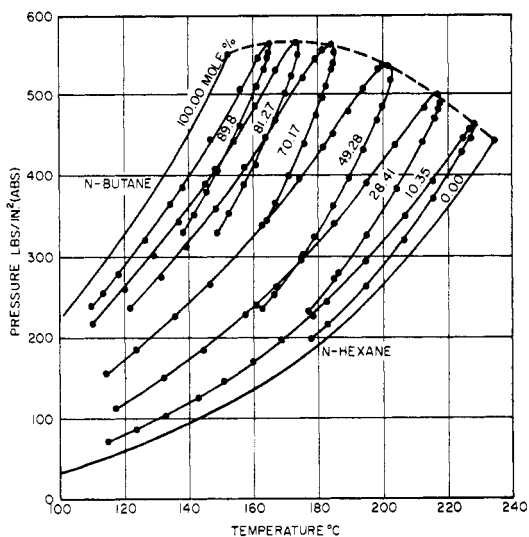


Figure 2. Pressure-temperature diagram of *n*-butane-*n*-hexane system

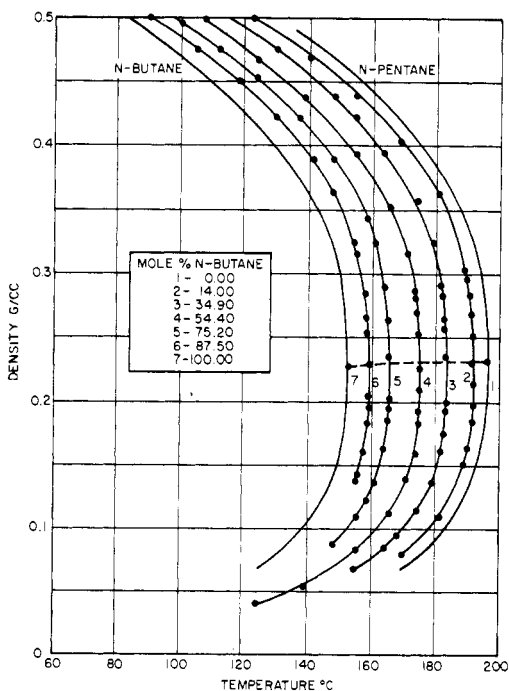


Figure 3. Density-temperature diagram of *n*-butane-*n*-pentane system

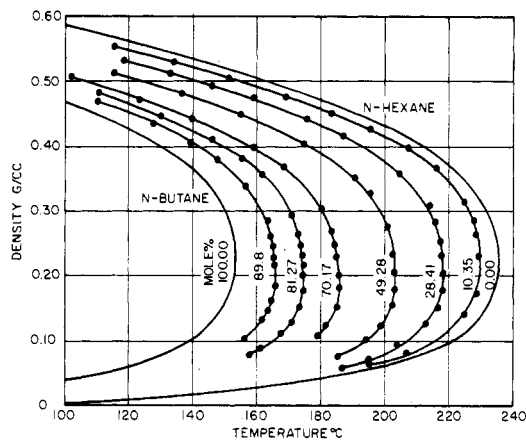


Figure 4. Density-temperature diagram of *n*-butane-*n*-hexane system

mental tube, its total volume was known in terms of the distance from the sealed end, so that the volume of the sample at any temperature and pressure was obtained by measuring the length of the tube occupied by the sample with a cathetometer. Equilibrium between the liquid and vapor phases was attained by moving a small steel ball enclosed in the tube by a magnet around the outside of the constant-temperature jacket. Pressure and volume were measured at a series of constant temperatures covering the range desired.

The pressure gauge and the thermocouple were calibrated; the former by comparison with a precision dead-weight gauge and the latter by comparison with a Pt-resistance thermometer which had been calibrated at the National Bureau of Standards. The emf of the thermocouple was measured with a potentiometer capable of measuring the millivolt equivalent of 0.005°C. The total volume of the experimental 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. The length of the tube occupied by the sample was measured with a cathetometer to within 0.02 mm. For details of the apparatus and experimental procedures, see refs. 7 and 8.

Materials and Preparation of Mixtures

n-Butane, *n*-pentane, and *n*-hexane were supplied by Phillips Petroleum Co. with a purity of 99.9 mol % or better. They were used without further purification except that they were deaerated by a cyclic operation which involved freezing, pumping off the released gas, melting, and distilling the sample at low pressure. Measured amounts of the pure *n*-butane were transferred to glass ampuls attached to the loading line. The ampuls were sealed off and stored for use in the preparation of mixtures with degassed *n*-pentane and *n*-hexane. The procedure for preparing and loading the experimental tube with a mixture of known composition has been described elsewhere (7, 8). The purity of the individual components was tested by determining the pressure difference between the bubble and dew points at constant temperature. This difference was always less than 2.0 lb/in.².

Equilibrium Data

The pressure, temperature, and volume at the bubble and dew points for a series of five mixtures of *n*-butane and *n*-pentane and six mixtures of *n*-butane and *n*-hexane were measured over a temperature range from room temperature to the highest temperature at which the liquid and vapor can coexist. For each mixture, 25-35 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 from data taken from the literature (3, 9, 10). 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 composi-

Table I. Summary of Temperature, Pressure, and Density Relationships at Phase Boundaries for *n*-Butane-*n*-Pentane System: Data by Hoffman (2)

Press, lb/in. ² abs	Liquid		Vapor	
	Temp, °C	Density, g/cc	Temp, °C	Density, g/cc
Composition: 13.99 mol % <i>n</i> -butane				
150	118.0	0.506
200	133.2	0.482
250	146.2	0.460
300	157.3	0.433
350	166.9	0.408	169.3	0.080
400	175.5	0.376	177.7	0.098
450	183.2	0.351	184.8	0.125
500	190.7	0.278	191.3	0.195
Composition: 34.93 mol % <i>n</i> -butane				
150	106.4	0.513
200	121.3	0.490
250	134.2	0.467
300	145.3	0.444
350	155.0	0.420	160.2	0.078
400	163.6	0.394	167.8	0.094
450	171.6	0.366	175.8	0.116
500	178.9	0.323	181.0	0.157
510	180.3	0.308	182.1	0.173
520	181.8	0.285	183.0	0.190
Composition: 54.44 mol % <i>n</i> -butane				
150	98.3	0.512
200	113.0	0.488
250	125.5	0.467	132.4	0.048
300	136.5	0.444	142.5	0.060
350	146.0	0.420	151.3	0.074
400	154.2	0.397	158.8	0.091
450	161.8	0.469	165.7	0.113
500	169.1	0.330	171.6	0.144
520	171.9	0.307	173.7	0.165
530	173.3	0.280	174.7	0.190
Composition: 75.18 mol % <i>n</i> -butane				
150	90.2	0.512
200	104.4	0.489
250	115.3	0.469
300	126.6	0.447
350	135.8	0.425
400	144.1	0.401	147.8	0.087
450	151.5	0.376	154.7	0.108
500	158.5	0.343	160.8	0.137
520	161.3	0.320	163.1	0.157
540	164.0	0.288	165.1	0.202
Composition: 87.45 mol % <i>n</i> -butane				
150	85.3	0.508
200	99.4	0.484
250	111.2	0.463
300	121.3	0.443
350	130.2	0.422
400	138.3	0.398
450	145.7	0.371
500	152.2	0.336
520	154.7	0.321	156.0	0.148
540	157.4	0.288	158.2	0.190

Table II. Summary of Temperature, Pressure, and Density Relationships at Phase Boundaries for *n*-Butane-*n*-Hexane System: Data by Davies (1)

Press, lb/in. ² abs	Liquid		Vapor	
	Temp, °C	Density, g/cc	Temp, °C	Density, g/cc
Composition: 10.35 mol % <i>n</i> -butane				
90	125.5	0.539
125	143.1	0.518
150	153.1	0.502
175	161.9	0.489
200	169.9	0.476	177.7	...
225	177.3	0.463	184.9	...
250	184.2	0.450	191.1	...
275	190.5	0.437	196.9	0.063
300	196.3	0.423	202.3	0.071
325	201.9	0.409	207.4	0.081
350	207.3	0.394	212.1	0.091
375	212.4	0.378	216.6	0.104
400	217.1	0.360	220.7	0.120
425	221.7	0.338	224.4	0.139
440	224.3	0.321	226.4	0.154
450	225.9	0.307	227.7	0.169
460	227.6	0.280	228.8	0.198
Composition: 28.41 mol % <i>n</i> -butane				
115	118.1	0.534
125	122.9	0.528
150	133.1	0.514
175	141.9	0.501
200	150.0	0.489
225	157.3	0.477
250	164.1	0.465	181.0	...
275	170.3	0.453	185.9	0.059
300	176.2	0.440	190.6	0.065
325	181.8	0.427	195.0	0.073
350	187.1	0.414	199.2	0.081
375	192.1	0.401	203.3	0.091
400	196.9	0.387	207.1	0.103
425	201.7	0.369	210.6	0.115
440	204.6	0.358	212.6	0.125
450	206.5	0.350	213.9	0.131
475	211.1	0.324	216.7	0.115
500	215.9	0.276	217.7	0.217
Composition: 49.28 mol % <i>n</i> -butane				
150	112.5	0.516
175	121.8	0.504
200	129.6	0.492
225	136.6	0.481
250	143.0	0.471	165.8	...
275	149.1	0.460	170.8	...
300	154.8	0.449	175.3	...
325	160.2	0.439	179.4	...
350	165.4	0.427	183.3	0.074
375	170.4	0.415	186.9	0.082
400	175.2	0.403	190.3	0.091
425	179.8	0.390	193.6	0.101
440	182.5	0.382	195.4	0.107
450	184.3	0.376	196.6	0.113

(Continued on page 336)

Table II. Continued

Press, lb/in. ² abs	Liquid		Vapor	
	Temp, °C	Density, g/cc	Temp, °C	Density, g/cc
Composition: 49.28 mol % <i>n</i> -butane				
475	188.7	0.359	199.3	0.127
500	193.0	0.339	201.5	0.145
525	197.5	0.307	203.1	0.176
535	199.7	0.282	202.7	0.163
Composition: 70.17 mol % <i>n</i> -butane				
165	102.1	0.503
175	105.5	0.498
200	113.0	0.487
225	119.7	0.477
250	125.7	0.468
275	131.4	0.458
300	136.8	0.448
325	141.9	0.438	160.9	...
350	146.9	0.427	164.5	...
375	151.7	0.415	167.9	...
400	156.3	0.403	171.1	...
425	160.7	0.390	174.2	...
440	163.1	0.383	176.0	...
450	164.7	0.378	177.1	0.101
475	168.5	0.365	179.7	0.114
500	172.4	0.349	182.1	0.126
525	176.4	0.328	184.2	0.144
550	180.5	0.297	185.6	0.179
560	182.6	0.272	185.2	0.207
Composition: 81.27 mol % <i>n</i> -butane				
215	110.0	0.484
225	112.5	0.479
250	118.4	0.468
275	123.9	0.458
300	129.1	0.448
325	134.0	0.438	148.3	...
350	138.6	0.427	152.1	...
375	143.1	0.416	155.6	...
400	147.3	0.405	158.9	0.083
425	151.4	0.394	162.0	0.091
440	153.8	0.385	163.8	0.097
450	155.3	0.381	164.9	0.100
475	159.1	0.366	167.7	0.111
500	162.7	0.350	170.2	0.125
525	166.3	0.329	172.4	0.142
550	170.1	0.299	174.0	0.164
560	171.6	0.276	174.3	0.187
Composition: 89.8 mol % <i>n</i> -butane				
240	110.4	0.468
250	112.7	0.464
275	118.1	0.454
300	123.1	0.444
325	127.7	0.434	137.3	...
350	132.2	0.423	141.5	...
375	136.5	0.412	145.2	...
400	140.5	0.401	148.6	...
425	144.4	0.390	151.7	...
440	146.7	0.380	153.5	...
450	148.1	0.377	154.6	0.101
475	151.7	0.362	157.4	0.111
500	155.2	0.345	160.1	0.125
525	158.6	0.325	162.6	0.143
550	162.1	0.294	164.6	0.173
560	163.6	0.270	165.1	0.192

Table III. Isobaric Temperature-Composition Relations of *n*-Butane-*n*-Pentane System

Compn, mol % <i>n</i> -C ₄	Temp, °C					
	Press, lb/in. ² abs					
	350		400		450	
	Liquid	Vapor	Liquid	Vapor	Liquid	Vapor
0	174.7	174.7	183.1	183.1	190.8	190.8
10	169.3	170.9	177.6	179.2	185.3	186.5
13.99	166.9	169.3	175.5	177.6	183.2	184.8
20	163.9	166.9	172.2	174.8	179.7	182.0
30	158.5	162.4	166.7	170.2	174.3	177.3
34.93	155.0	160.2	163.9	167.9	171.6	174.8
40	153.2	158.0	161.3	165.6	169.0	172.5
50	148.1	153.4	156.3	161.0	164.0	167.7
54.44	146.0	151.2	154.2	158.8	161.8	165.6
60	143.0	148.3	151.5	156.0	159.0	162.7
70	138.1	142.8	146.6	150.7	154.0	157.4
75.18	135.8	139.8 ^a	144.1	147.8	151.5	154.6
80	133.4	137.0	141.8	145.0	149.2	151.8
87.45	130.2	132.5 ^a	138.3	140.5 ^a	145.7	147.5 ^a
90	129.1	131.0	137.2	139.0	144.5	146.0
100	125.0	125.0	132.7	132.7	139.8	139.8
500						
	Liquid	Vapor				
13.99	190.6	191.3				
20	187.4	188.4				
30	182.1	183.4				
34.93	178.8	181.1				
40	176.8	178.6				
50	171.5	173.8				
54.44	169.1	171.6				
60	166.3	168.8				
70	161.1	163.5				
75.18	158.6	160.8				
80	156.0	158.0				
87.45	152.3	153.8				
90	151.0	152.2				
100	146.1	146.1				

^a Extrapolated.Table IV. Isobaric Temperature-Composition Relations of *n*-Butane-*n*-Hexane System

Compn, mol % <i>n</i> -C ₄	Temp, °C					
	Press, lb/in. ² abs					
	325		350		375	
	Liquid	Vapor	Liquid	Vapor	Liquid	Vapor
0	214.0	214.0	218.9	218.9	223.4	223.4
10	202.5	207.7	207.6	212.5	212.6	216.9
20	191.1	200.9	196.3	205.4	201.9	209.7
30	179.2	193.8	185.5	193.2	191.1	202.2
40	169.6	186.4	179.8	190.5	180.3	194.4
50	159.5	178.9	164.7	182.7	169.6	186.4
60	150.2	170.5	155.2	174.1	160.0	177.8
70	142.0	161.0	146.9	164.6	151.7	168.2
80	134.9	150.0	139.6	153.7	144.2	157.3
90	127.8	137.1	132.3	141.2	136.5	145.1
100	120.7	120.7	125.0	125.0	128.9	128.9

Table IV. Continued

Compn, mol % <i>n</i> -C ₄	Temp, °C					
	Press, lb/in. ² abs					
	400		425		450	
	Liquid	Vapor	Liquid	Vapor	Liquid	Vapor
0	227.9	227.9	232.2	232.2
10	217.5	221.0	222.1	224.7	226.3	227.9
20	206.4	213.6	211.1	217.1	215.5	220.4
30	195.2	205.9	199.9	209.3	204.7	212.5
40	184.7	197.9	189.2	201.3	193.8	204.5
50	174.5	189.7	179.1	193.2	186.4	195.9
60	165.1	180.9	169.7	184.2	173.7	187.1
70	156.5	171.2	160.8	174.4	164.8	177.6
80	148.5	160.4	152.4	163.6	156.4	166.6
90	140.6	148.4	144.2	151.6	148.0	154.3
100	132.7	132.7	136.2	136.2	139.7	139.7
	500		550			
	Liquid	Vapor	Liquid	Vapor		
30	214.2	216.6		
40	203.2	209.3		
50	192.5	200.9		
60	182.2	192.1	192.0	193.8		
70	172.6	182.4	180.7	185.8		
80	163.8	171.7	171.5	175.5		
90	155.0	160.0	161.9	164.5		
100	146.1	146.1	152.2	152.2		

tion are given. Isobaric temperature-composition 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.

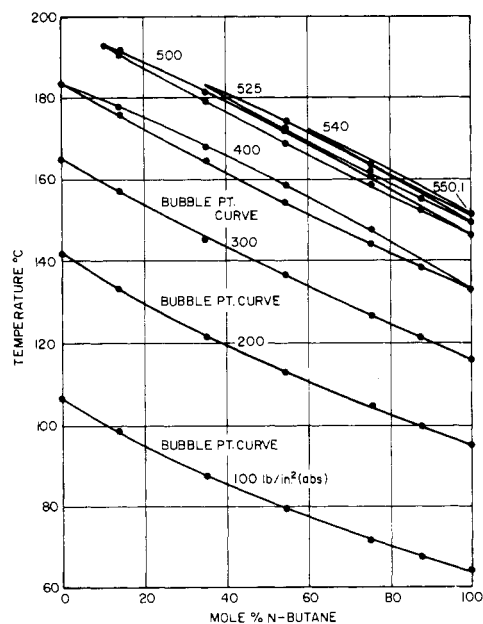
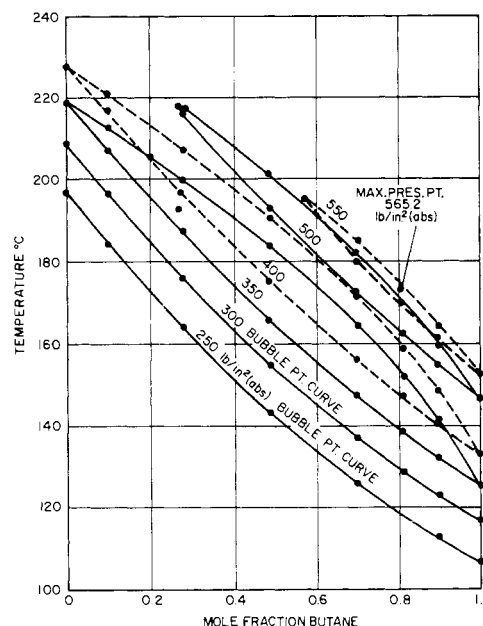
The critical point was determined visually by the disappearance-of-the-meniscus method, whereas the coordinates of the pressure, temperature, and density at the maximum pressure and maximum temperature points were obtained graphically from large-scale plots of the *P*-*T* and ρ -*T* border curves in the critical region of the mixture. The critical constants of the pure components and mixtures and the coordinates of the maximum pressure (circondenbar) and maximum temperature (circondentherm) points of each system are listed in Tables VII and VIII. The coordinates at the maximum pressure point on the *P*-*T* locus curve for the *n*-butane-*n*-hexane system are as follows: $P = 565.2 \pm 1.0$ lb/in.² abs; $T = 173.9 \pm 1.0$ °C; $x = 0.807$ C₄. The *P*-*T* critical locus curve of the *n*-butane-*n*-pentane system has no maximum pressure point.

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

Acknowledgment is made to the Phillips Petroleum Co. for samples of the pure hydrocarbons.

Figure 5. Isobaric temperature-composition diagram of *n*-butane-*n*-pentane systemFigure 6. Isobaric temperature-composition diagram of *n*-butane-*n*-hexane systemTable V. Vapor-Liquid Equilibrium Ratios of *n*-Butane-*n*-Pentane System

Press, lb/in. ² abs	Temp, °C					
	140	150	160	170	180	190
	$K_i = y_i/x_i$ for <i>n</i> -butane					
350	1.131	1.229	1.300	1.414
400	1.055	1.130	1.223	1.271	1.428	...
450	1.001	1.063	1.124	1.183	1.256	1.357
500	...	1.017	1.057	1.089	1.094	1.113
	$K_j = y_j/x_j$ for <i>n</i> -pentane					
350	0.743	0.803	0.893	0.961
400	0.718	0.778	0.835	0.914	0.975	...
450	0.750	0.774	0.829	0.887	0.938	0.995
500	...	0.800	0.852	0.900	0.952	0.980

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

Press, lb/in. ² abs	Temp, °C										
	130	140	150	160	170	180	190	200	210	220	230
	$K_i = y_i/x_i$ for <i>n</i> -butane										
325	1.091	1.207	1.326	1.433	1.530	1.605	1.683	1.756	1.846
350	1.044	1.145	1.258	1.359	1.442	1.516	1.581	1.647	1.721
375	1.009	1.095	1.191	1.294	1.374	1.428	1.465	1.507	1.581	1.672	...
400	...	1.057	1.137	1.221	1.302	1.367	1.423	1.455	1.474	1.496	...
425	...	1.028	1.100	1.170	1.243	1.313	1.364	1.391	1.383	1.354	1.289
450	...	1.002	1.063	1.129	1.203	1.263	1.301	1.323	1.322	1.293	1.106
500	1.018	1.068	1.116	1.160	1.189	1.191	1.157
550	1.020	1.043	1.072	1.057
	$K_j = y_j/x_j$ for <i>n</i> -hexane										
325	0.395	0.447	0.505	0.575	0.652	0.738	0.819	0.896	0.969
350	0.402	0.442	0.496	0.565	0.643	0.721	0.799	0.870	0.938
375	0.433	0.441	0.499	0.560	0.631	0.711	0.790	0.859	0.918	0.978	...
400	...	0.446	0.513	0.574	0.636	0.705	0.774	0.843	0.905	0.959	...
425	...	0.433	0.517	0.584	0.640	0.698	0.765	0.833	0.898	0.952	0.993
450	...	0.51	0.551	0.597	0.638	0.698	0.767	0.832	0.892	0.945	0.993
500	0.593	0.637	0.685	0.733	0.792	0.856	0.920
550	0.770	0.810	0.826	0.909

Table VII. Critical Constants of *n*-Butane-*n*-Pentane System

Mol %, <i>n</i> -C ₄	Critical point			Point of max press			Point of max temp		
	<i>T_c</i> , °C	<i>P_c</i> , lb/in. ² abs	Density, g/cc	<i>T_{Pmax}</i> , °C	<i>P_{Pmax}</i> , lb/in. ² abs	Density, g/cc	<i>T_{Tmax}</i> , °C	<i>P_{Tmax}</i> , lb/in. ² abs	Density, g/cc
0.00	196.57 ^a	488.8 ^a	0.2315 ^a						
13.99	191.44	503.6	0.2309	191.4	503.7	0.252	191.5	503.5	0.214
34.93	183.06	525.0	0.2362	182.8	525.3	0.257	183.2	523.8	0.210
54.44	174.74	536.7	0.2250	174.5	537.0	0.253	174.9	535.0	0.209
75.18	165.08	545.7	0.2341	165.0	546.0	0.252	165.2	544.1	0.202
87.45	158.8	548.6	0.2279	158.6	549.0	...	158.9	548.0	0.204
100.00	152.2 ^b	550.1 ^b	0.2278 ^b						

^a Ref. 3. ^b Ref. 9.

Table VIII. Critical Constants of *n*-Butane-*n*-Hexane System

Mol % <i>n</i> -C ₄	Critical point			Point of max press			Point of max temp		
	<i>T_c</i> , °C	<i>P_c</i> , lb/in. ² abs	Density, g/cc	<i>T_{Pmax}</i> , °C	<i>P_{Pmax}</i> , lb/in. ² abs	Density, g/cc	<i>T_{Tmax}</i> , °C	<i>P_{Tmax}</i> , lb/in. ² abs	Density, g/cc
0.00	234.7 ^a	440.1 ^a	0.232 ^a						
10.35	228.61	462.3	0.2307	228.4	462.9	0.255	228.8	460.8	0.205
28.41	217.34	501.5	0.2307	216.9	502.1	0.251	218.0	495.4	0.192
49.28	202.01	537.6	0.2317	201.3	539.1	0.251	203.1	529.4	0.185
70.17	184.39	562.0	0.2297	183.9	562.5	0.241	185.6	553.0	0.184
81.27	173.37	565.1	0.2319	173.2	565.2	0.239	174.3	559.6	0.185
89.8	164.50	562.4	0.2306	164.5	562.4	0.240	165.1	559.6	0.191
100.00	152.2 ^b	550.1 ^b	0.2281 ^b						

^a Ref. 10. ^b Ref. 9.

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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 X 148 mm, 24X 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 \$4.50 for photocopy or \$2.50 for microfiche, referring to code number JCED-75-333.