Subscripts

1 = component 1, n-pentane
2 = component 2, acetone

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RECEIVED for review June 30, 1961. Accepted February 9, 1962. Work submitted by Teh Cheng Lo in 1959 to Stevens Institute of Technology in partial fulfillment of the requirements for the degree of master of science in chemical engineering.

Phase Behavior in a Six-Component Hydrocarbon System

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IN DESIGNING EQUIPMENT for the separation of hydrocarbons of similar molecular weight and volatility, information on phase behavior of the system is important. Because hydrocarbons deviate significantly from ideal solutions (*14*), it is difficult to predict the contribution of each of several components to the phase behavior of the system as a whole. Experimental investigation of the phase behavior of the hydrocarbon system is usually necessary if a detailed knowledge of the composition of the coexisting phases is required.

The basis for this study was an industrial interest in the phase behavior of a six-component system made up of hydrocarbons containing four carbon atoms per molecule. Little information appears to be available involving more than binary systems of such hydrocarbons, although the phase behavior of the 1-butene-n-butane system has been studied (*22*), and Gerster and coworkers (*9*) investigated the phase behavior of the n-butane-1-butene-furfural and isobutane-1-butene-furfural systems.

Ten different mixtures of isobutane, isobutene, n-butane, 1-butene, *trans*-2-butene, and *cis*-2-butene were investigated. Experimental work was carried out at temperatures from 126° F. to 220° F., and samples of each of the coexisting phases were obtained at more than one temperature for most of the mixtures.

Raoult's law was used as a frame of reference in depicting the phase behavior of the six-component hydrocarbon system. Only a knowledge of the vapor pressure of each of the components is required in predicting the behavior described by this relation. The deviations from Raoult's law were treated as systematic and simple functions of temperature, pressure, and composition. Results are pre-

Table I. Purity of Components

	Composition							
Component	Isobutane	Isobutene	n-Butane	1-Butene	trans-2-Butene	cis-2-Butene	Butadiene	Purity[a] Mole Fraction
			(Mole Fraction of Component)					
Isobutane	0.998	...	0.002	...	...	...	...	0.9994
Isobutene	...	0.995	0.001	0.004	...	...	...	0.9952
n-Butane	0.001	...	0.999	...	...	...	...	0.9991
1-Butene	0.005	0.003	0.003	0.987	...	...	0.002	0.999+
trans-2-Butene	...	...	0.004	...	0.992	0.004	...	0.9937
cis-2-Butene	...	...	...	...	...	0.999	0.001	0.9933

[a] Reported by Phillips Petroleum Co.

sented both in terms of the quotient of the equilibrium ratio determined experimentally and the equilibrium ratio predicted from Raoult's law.

MATERIALS

The hydrocarbons employed were research grade (Phillips Petroleum Co.). They were received at the laboratory in steel containers and were used without further purification. Care was exercised to avoid contamination of the samples by air. Table I gives the reported purity of each of the components and includes the results of special analyses upon subsamples. The purity of each of the components was such that in the six-component mixture the total extraneous material was less than 0.003 mole fraction. The composition of each mixture, determined gravimetrically during the addition process, was corrected for the traces of impurities in each of the several components involved. Part of the impurities in the individual hydrocarbon samples consisted of other components of the system, and these were taken into account in calculating the composition of a given mixture.

Experimental vapor pressure data for each of the six components are recorded in Table II at intervals of 5° F. These data are given in order that appropriate values of equilibrium ratio may be calculated from the tabular experimental results as a function of temperature and pressure. These data are presented more precisely than is justified by the accuracy of the primary experimental data. The references for the tabular values are included in this table.

EXPERIMENTAL METHODS

The hydrocarbons were confined in a stainless-steel vessel which was partially filled with mercury. This vessel was immersed in an agitated oil bath whose temperature was controlled within 0.02° F. in relation to the international platinum scale. The pressure was measured with a balance (*23*) which had been calibrated against the vapor pressure of carbon dioxide at the ice point (*4*). Relative to this standard, the pressures were known within either 0.1 pound per square inch or 0.1%, whichever was the larger measure of uncertainty. Equilibrium was hastened by mechanical agitation of the contents within the pressure vessel. Details of construction and methods of operation of the apparatus have been described (*23*).

The samples were introduced by weighing bomb techniques (*21*), and the composition of the mixture as a whole was known within 0.001 mole fraction. After introduction of the components the bubble-point pressure at a single temperature was determined, and the system was then maintained in the heterogeneous region. Samples of the gas and liquid phases were withdrawn under isobaric, isothermal conditions. The volume of the system was decreased during withdrawal to maintain isobaric conditions. After the sample was taken, the glass sample bulbs were immersed in liquid nitrogen and sealed. Duplicate samples of the gas and of the liquid phases were taken at each state.

ANALYSES

Compositions of samples from the liquid and gas phases of the mixtures were determined by combined chromatographic and mass spectrographic analysis. It is estimated from duplicate measurements that the mole fraction of each component was established within 0.002 mole fraction.

EXPERIMENTAL RESULTS

Table III records experimental information pertaining to each mixture investigated. Included are the composition of liquid and gas phases as determined from samples withdrawn under isobaric, isothermal conditions; the equilibrium ratios based both upon the experimental measurements and upon Raoult's law; and β_k, the quotient of the two latter quantities. The following equations define the equilibrium ratios recorded in Table III:

$$K_k = \frac{y_k}{x_k} = \frac{\beta_k P_k'}{P} \qquad (1)$$

Table II. Vapor Pressure of Components

	Component					
Temp., ° F.	Isobutane	Isobutene	*n*-Butane	1-Butene	*trans*-2-Butene	*cis*-2-Butene
100	71.9[a]	63.2	51.5	62.5	49.6	45.9
105	77.6	68.1	55.9	67.3	53.4	49.8
110	83.2	73.6	60.1	72.6	57.7	53.8
115	89.4	78.7	65.2	78.1	62.5	58.1
120	95.8	85.2	69.8	83.9	67.5	62.6
125	102.7	91.6	75.3	89.8	72.5	67.6
130	109.8	98.6	80.6	96.5	78.3	73.0
135	117.5	105.4	86.7	103.2	83.7	78.0
140	125.3	112.8	92.7	110.4	89.6	83.5
145	134.1	120.7	99.2	118.0	95.8[b]	89.3[b]
150	142.3	127.9	106.0	125.8	104.2	97.2
155	151.9	138.0	113.1	135.4	111.9	101.5
160	161.1	146.3	120.8	142.7	118.8	111.5
165	171.6	156.2	128.5	152.2	124.6	117.9
170	181.6	165.7	137.1	161.3	132.7	124.8
175	193.3	174.9	145.7	171.3	140.9	133.0
180	204.0	186.6	154.8	181.6	149.8	141.4
185	216.2	197.9	164.6	192.6	159.1	150.6
190	228.3	208.9	173.3	203.8	172.1	162.9
195	241.4	221.6	185.1	215.5	178.6	171.1
200	254.6	234.0	195.5	227.9	189.3	181.3
205	269.1	246.9	205.7	241.4	201.0	192.4
210	283.1	260.2	218.4	254.1	213.1	204.8
215	298.1	274.1	229.2	268.2	225.6	217.1
220	313.7	288.4	243.2	282.6	239.9	229.6
225	329.6	304.0	254.7	297.9	253.3	241.8

Source of data: isobutane (*15, 19, 20*); isobutene (*2, 8, 12, 19*); *n*-butane (*1, 3, 11, 16, 19, 24*); 1-butene (*7, 12, 13, 17, 19*); trans-2-butene (*8, 10, 12, 19*); *cis*-2-butene(*8, 12, 19, 25*).

[a] Pressure, p.s.i.

[b] Vapor pressures at temperatures higher than 145° F. extrapolated.

$$K_{Rk} = \frac{y_{Rk}}{x_{Rk}} = \frac{P_k''}{P} = \frac{\phi_k(T)}{P} \qquad (2)$$

$$\beta_k = \frac{K_k}{K_{Rk}} = \frac{y_k x_{Rk}}{x_k y_{Rk}} = \frac{y_k P}{x_k P_k''} = \phi_k(T,\ x_k, \frac{x_p}{x_p + x_o}) \qquad (3)$$

In Equation 3, β_k was assumed to be a function of the temperature, the relative quantities of paraffin and olefin in the liquid phase other than component k, and its own mole fraction. In effect, this treats the six-component system as a restricted ternary system. Hereafter the phrases "mole fraction paraffin" and "mole fraction olefin" will imply the meaning stated above.

Utilizing Wohl's equation for a symmetrical two-suffix system (*26*) and taking into account the effect of temperature upon the coefficients of the Wohl equation (*6*), the following expression was developed:

$$\log \beta_k = x_p^2(\alpha_p + \tau_p/T) + x_o^2(\alpha_o + \tau_o/T) + x_p x_o(\alpha_{po} + \tau_{po}/T) \qquad (4)$$

Equation 4 relates the coefficient β_k for each component to the mole fraction paraffin, the mole fraction olefin, and the temperature expressed in degrees Rankine. For each component there are a total of six coefficients which were evaluated from the experimental data by the method of least squares (*18*). Automatic computing methods (*5*) were employed to attain the solution of the appropriate matrix equations.

The standard error of estimate for the experimental values of β_k from Equation 4 is shown in Table IV. Values of the standard error of estimate are in some cases significant when compared to the value of β_k, which is approximately unity.

Values of β_k at 130° F., calculated from Equation 4 and using the coefficients of Table IV, are shown in Figures 1, 2, and 3 for the components investigated. The symmetrical two-suffix equation developed by Wohl de-

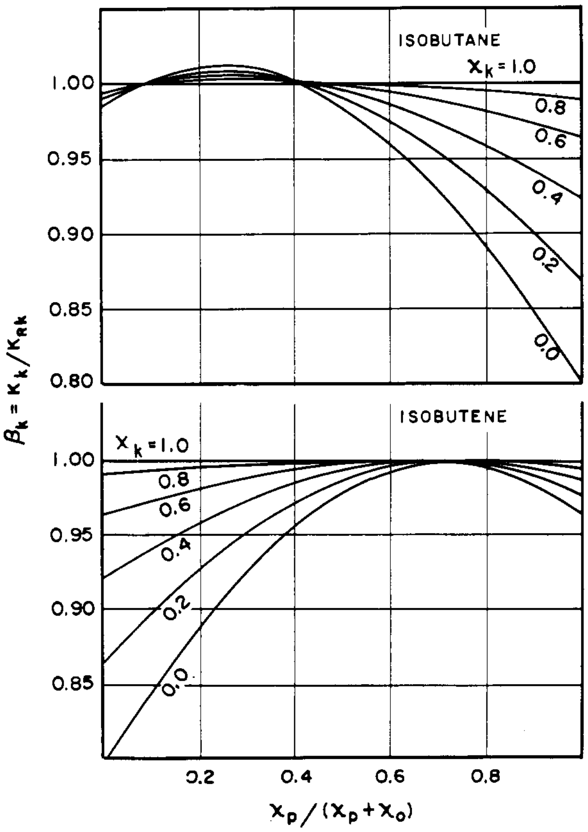

Figure 1. Phase behavior of isobutane and isobutene at 130° F. in the six-component system

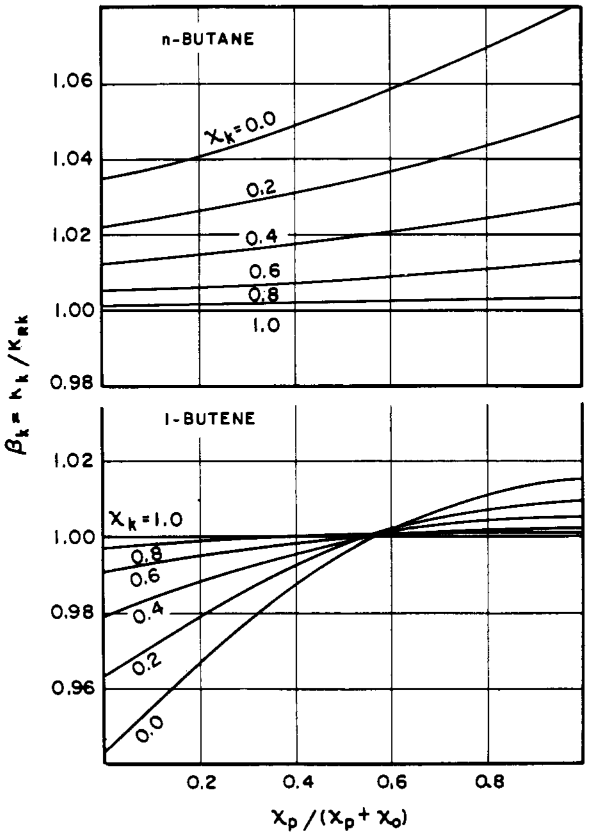

Figure 2. Phase behavior of *n*-butane and 1-butene at 130° F. in the six-component system

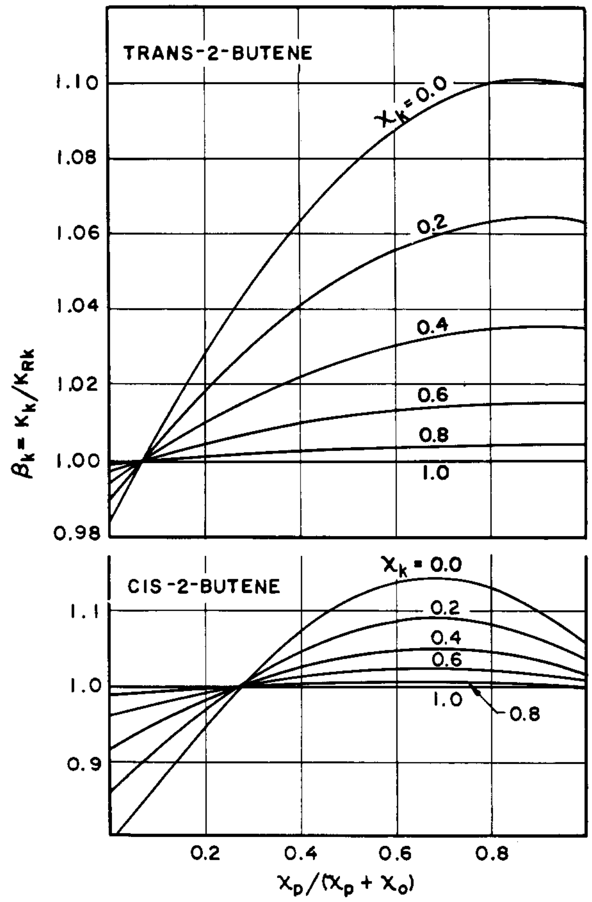

Figure 3. Phase behavior of *trans*-2-butene and *cis*-2-butene at 130° F. in the six-component system

Table III. Composition of Coexisting

	Compn. Mole Fraction		Equil. Ratio			Compn. Mole Fraction		Equil. Ratio	
	Gas	Liquid	Exptl.	Raoult's Law	β_k	Gas	Liquid	Exptl.	Raoult's Law
Mixture No.			A					B	
Temp., ° F.			131					139	
Withdrawal pres., p.s.i.			99.01					99.83	
Bub. pt. pres., p.s.i.			99.52					101.39	
Paraffin, mole fraction			0.517					0.440	
Isobutane	0.4378	0.4022	1.088	1.118	0.973	0.1029	0.0812	1.267	1.214
Isobutene	0.1575	0.1567	1.005	0.999	1.006	0.1905	0.1755	1.086	1.097
n-Butane	0.0989	0.1149	0.861	0.824	1.045	0.3386	0.3585	0.944	0.903
1-Butene	0.1299	0.1222	1.063	0.983	1.081	0.1841	0.1742	1.057	1.076
trans-2-Butene	0.0964	0.1130	0.853	0.798	1.069	0.1036	0.1155	0.897	0.882
cis-2-Butene	0.0795	0.0911	0.873	0.741	1.178	0.0804	0.0951	0.845	0.818
Mixture No.			E					F	
Temp., ° F.			130					160	
Withdrawal pres., p.s.i.			92.48					134.12	
Bub. pt. press., p.s.i.			93.46					137.77	
Paraffin, mole fraction			0.795					0.794	
Isobutane	0.4445	0.3910	1.137	1.175	0.968	0.4324	0.3823	1.131	1.169
Isobutene	0.0557	0.0515	1.082	1.055	1.026	0.0566	0.0533	1.062	1.062
n-Butane	0.3570	0.4044	0.883	0.862	1.024	0.3690	0.4112	0.897	0.877
1-Butene	0.0538	0.0505	1.065	1.032	1.032	0.0535	0.0513	1.043	1.036
trans-2-Butene	0.0454	0.0514	0.883	0.838	1.054	0.0455	0.0505	0.901	0.862
cis-2-Butene	0.0437	0.0512	0.854	0.781	1.093	0.0431	0.0517	0.834	0.809
Mixture No.			J					K	
Temp., ° F.			160					130	
Withdrawal pres., p.s.i.			130.06					92.34	
Bub. pt. pres., p.s.i.			130.27					93.37	
Paraffin, mole fraction			0.094					0.593	
Isobutane	0.0554	0.0453	1.201	1.237	0.971	0.4499	0.3917	1.149	1.176
Isobutene	0.0499	0.0491	1.016	1.123	0.905	0.1073	0.1016	1.056	1.056
n-Butane	0.0479	0.0484	0.990	0.927	1.068	0.1776	0.2012	0.883	0.863
1-Butene	0.4136	0.3938	1.050	1.095	0.959	0.0514	0.0505	1.018	1.034
trans-2-Butene	0.3915	0.4171	0.939	0.912	1.030	0.0443	0.0496	0.893	0.839
cis-2-Butene	0.0415	0.0463	0.846	0.856	0.981	0.1695	0.2053	0.826	0.782
Mixture No.			N					O	
Temp., ° F.			220					130	
Withdrawal press., p.s.i.			248.6					93.74	
Bub. pt. pres., p.s.i.			250.1					94.08	
Paraffin, mole fraction			0.197					0.404	
Isobutane	0.1105	0.0945	1.169	1.254	0.932	0.2239	0.1973	1.135	1.167
Isobutene	0.1086	0.0994	1.093	1.153	0.948	0.4166	0.4077	1.022	1.048
n-Butane	0.1010	0.1024	0.986	0.973	1.013	0.1792	0.2064	0.868	0.857
1-Butene	0.1045	0.0972	1.075	1.130	0.951	0.0986	0.0996	0.990	1.026
trans-2-Butene	0.1952	0.1998	0.977	0.959	1.019	0.0428	0.0432	0.991	0.832
cis-2-Butene	0.3802	0.4067	0.935	0.918	1.019	0.0388	0.0456	0.851	0.776
Mixture No.			R						
Temp., ° F.			220						
Withdrawal pres., p.s.i.			243.6						
Bub. pt. press., p.s.i.			244.2						
Paraffin, mole fraction			0.903						
Isobutane	0.0010	0.0012	0.833	1.284	0.649				
Isobutene	0.1102	0.0970	1.136	1.181	0.962				
n-Butane	0.8888	0.9018	0.986	0.996	0.990				
1-Butene	...	...	...	...	...				
trans-2-Butene	...	...	...	...	...				
cis-2-Butene	...	...	...	...	...				

scribes the experimental results in a reasonable fashion, as is apparent from the standard errors of estimate (Table IV).

Figure 4 records the effect of temperature on β_k for a single set of conditions where the mole fractions of the paraffin, the olefin, and the component in question are equal to 0.4, 0.4, and 0.2, respectively. With the exception of 1-butene, the total change in the values of β_k with temperature is much larger than the standard error of estimate. Therefore, the trends with respect to temperature are believed significant.

ACKNOWLEDGMENT

The assistance of the Beacon Laboratory of Texaco, Inc. in carrying out the chromatographic analysis is gratefully

Phases and Equilibrium Ratios

	Compn. Mole Fraction		Equil. Ratio			Compn. Mole Fraction		Equil. Ratio		
β_k	Gas	Liquid	Exptl.	Raoult's Law	β_k	Gas	Liquid	Exptl.	Raoult's Law	β_k
			C					D		
			136 105.77 105.91 0.427					126 91.96 92.76 0.444		
1.044	0.3690	0.3289	1.122	1.126	0.996	0.3209	0.2810	1.142	1.122	1.018
0.990	0.2450	0.2491	0.984	1.008	0.976	0.2599	0.2603	0.998	0.997	1.001
1.045	0.0827	0.0980	0.844	0.829	1.018	0.1412	0.1625	0.869	0.823	1.056
0.982	0.1948	0.1916	1.017	0.989	1.028	0.2356	0.2426	0.971	0.984	0.987
1.017	0.0701	0.0841	0.834	0.808	1.032	0.0322	0.0403	0.799	0.796	1.004
1.033	0.0384	0.0485	0.792	0.749	1.057	0.0102	0.0133	0.767	0.738	1.039
			G					H		
			220 270.5 272.4 0.794					130 85.89 87.66 0.095		
0.967	0.4092	0.3780	1.082	1.151	0.940	0.0589	0.0468	1.258	1.253	1.004
1.000	0.0566	0.0537	1.054	1.059	0.995	0.0529	0.0571	0.926	1.125	0.823
1.023	0.3883	0.4156	0.934	0.893	1.046	0.0453	0.0483	0.936	0.920	1.017
1.007	0.0530	0.0506	1.047	1.038	1.009	0.4325	0.3834	1.128	1.101	1.025
1.045	0.0472	0.0511	0.924	0.881	1.049	0.3735	0.4172	0.895	0.893	1.002
1.031	0.0463	0.0510	0.908	0.843	1.077	0.0368	0.0470	0.783	0.833	0.887
			L					M		
			220 270.6 273.4 0.587					130 82.43 82.87 0.201		
0.977	0.4160	0.3784	1.099	1.147	0.958	0.1302	0.0990	1.315	1.325	0.992
1.000	0.1052	0.1009	1.043	1.055	0.989	0.1174	0.1014	1.158	1.190	0.973
1.023	0.1906	0.2082	0.916	0.890	1.029	0.1010	0.1019	0.991	0.973	1.018
0.985	0.0514	0.0494	1.040	1.034	1.006	0.1096	0.0978	1.121	1.164	0.963
1.064	0.0488	0.0516	0.946	0.878	1.077	0.1889	0.1995	0.947	0.945	1.002
1.056	0.1879	0.2115	0.888	0.840	1.057	0.3528	0.4003	0.881	0.881	1.000
			P					Q		
			220 276.5 277.8 0.402					130 80.50 80.67 0.898		
0.973	0.2055	0.1926	1.067	1.129	0.945	0.0009	0.0012	0.750	1.361	0.551
0.975	0.4132	0.4037	1.024	1.038	0.987	0.1190	0.1014	1.174	1.222	0.961
1.013	0.1927	0.2090	0.922	0.876	1.053	0.8801	0.8973	0.981	0.999	0.982
0.965	0.0989	0.0966	1.024	1.018	1.006	...	...	...	...	...
1.191	0.0458	0.0481	0.952	0.864	1.102	...	...	...	...	...
1.097	0.0438	0.0500	0.876	0.827	1.059	...	...	...	...	...

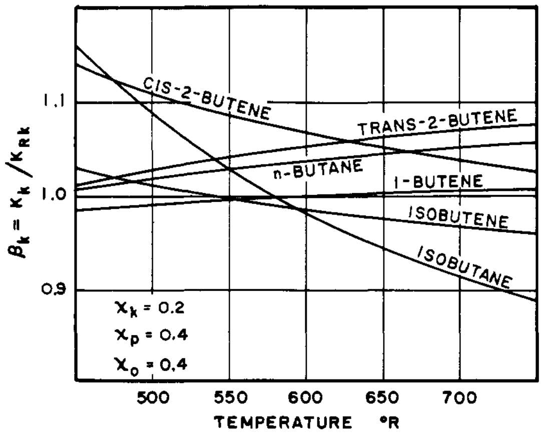

Figure 4. Effect of temperature upon deviations from Raoult's Law

recognized. H.H. Reamer was responsible for the general supervision of the laboratory activities.

NOMENCLATURE

K_k = equilibrium ratio of component k
N = total number of experimental points
P = pressure, lb./sq. in.
P_k' = vapor pressure of component k, lb./sq. in.
T = absolute temperature, ° R.
x_k = mole fraction of component k in liquid phase
x_p = mole fraction paraffins in liquid phase excluding component k
x_o = mole fraction olefins in liquid phase excluding component k
y_k = mole fraction of component k in gas phase
α = coefficient of Equation 4, dimensionless
β_k = quotient defined by Equation 3
$\sum_{n=1}^{N}$ = sumation operator

Table IV. Coefficients for Two-Suffix Equation[a]

Component	α_p	α_o	α_{po}	τ_p	τ_o	τ_{po}	Standard Error of Estimate[b]
Isobutane	−0.1810	−0.1920	−1.044	50.16	109.8	659.5	0.0195
Isobutene	0.2138	0.5172	−1.125	135.6	363.4	708.7	0.0193
n-Butane	0.5869	0.0909	−0.3472	326.3	44.82	229.4	0.0195
1-Butene	−0.1587	−0.3677	0.6539	97.33	201.8	379.2	0.0277
trans-2-Butene	−0.3440	−0.1809	0.9741	227.2	102.4	518.8	0.0276
cis-2-Butene	0.2195	1.300	−1.872	116.0	828.5	1261.0	0.0345

[a] Equation 4

[b] $\sigma = [\sum_{n=1}^{N} (\beta_{ke} - \beta_{kc})^2/N]^{1/2}$

σ = standard error of estimate
τ = coefficient of Equation 4, ° R.
$\phi_k()$ = function of () for component k

Subscripts

c = calculated
e = experimental
n = individual points used in analysis
o = olefin
p = paraffin
R = Raoult's law

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RECEIVED for review June 22, 1959. Revised December 26, 1961. Accepted March 1, 1962. Program was sponsored by Texaco, Inc., which provided financial support.