

In both of these systems, the values of γ_2 ranged from 0.85 to 1; while the values of γ_1 were unity or slightly higher. These data are thermodynamically inconsistent, but results of this kind have been reported earlier (1, 2, 9) in studies of ideal systems.

All the compounds were redistilled in a rectification column of over 70 theoretical plates; fractional values of γ_2 again were obtained. Pure liquids were then run in the Othmer still and the refractive indices of vapor and liquid determined. These were identical for each of the two liquids, thus suggesting the absence of impurities (at least of the kind that could be detected by refractive index measurements). These observations confirm the view of Van Winkle and coworkers (2, 9) that for systems approaching ideality such thermodynamic inconsistencies are likely.

NOMENCLATURE

P = vapor pressure of pure component, mm. of Hg.
 x = mole fraction in the liquid phase
 y = mole fraction in the vapor phase
 γ = activity coefficient

Subscripts

1 = refers to low boiling component
2 = refers to high boiling component

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Low Temperature Phase Equilibria of the Gas-Liquid System Helium-Neon-Nitrogen

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Vapor and liquid composition data were obtained for the gas-liquid systems helium-nitrogen, neon-nitrogen, and helium-neon-nitrogen at 82.70° K. and 113.13° K. from 6.4 to 50 atm. From the observed linear relation between the compositions of helium and neon in the vapor phase for the binary and ternary mixtures at constant temperature and pressure, semiempirical equations relating the compositions of the phases were derived from which smoothed and interpolated liquid composition data were calculated.

THE SOLUBILITY of helium gas in liquid nitrogen (1-3, 6) and of helium-neon gas mixtures in liquid nitrogen (7) have been reported. The literature data for the ternary gas-liquid system (7) are of limited value and cannot be used for process calculations. No data have been reported for the solubility of neon gas in liquid nitrogen. Measurements were undertaken, therefore, on the binary solutions of neon and helium gases in liquid nitrogen, and on the ternary solutions of helium-neon gas mixtures in liquid nitrogen at pressures up to 50 atm. The liquid phase compositions were correlated and extended throughout the experimental pressure range by semiempirical equations derived from standard thermodynamic relations and the Henry's Law constant for neon in helium-nitrogen mixtures.

EXPERIMENTAL

Materials. The purity of the gases neon, nitrogen, and helium were determined by mass spectrometer. The analyses

given in Table I show that no further purification of the gases was necessary for the phase equilibrium studies. Liquid and vapor samples taken from the equilibrium cell were also analyzed by mass spectrometer using calibration mixtures prepared with an accuracy of $\pm 0.4\%$.

Apparatus and Procedure. The technique used for obtaining equilibrium between a liquid solution and its gas phase is known as the "once-through-flow" method. In this procedure, a feed gas of composition lying between the estimated equilibrium compositions of the two phases is brought near to the temperature of the system and passed through the liquid phase. The vapor and liquid phases are withdrawn continuously at a rate such that the mass balance of each component in the system is maintained. The advantages of this method are that the feed gas composition needs to be known only approximately, and the technique used for withdrawing samples ensures that the liquid sample truly represents the equilibrium composition. The latter is a particularly important consideration at low

Table I. Analysis of Gases Used for Vapor-Liquid Equilibrium Studies

Gas	Source and Grade	Impurity Concentrations, P.P.M. V/V						
		N ₂	O ₂	H ₂	CO ₂	C.H.	Ar	Ne
Neon	Airco, spectroscopic	<50	<50	<50	<0.5	<0.5	ND	...
Nitrogen	Airco, prepurified	...	ND ^a	ND	12	ND	11	ND
Helium	Bureau of Mines	23	5	<4	2	1	<4	<10

^a ND—none detected, indicating less than 5 p.p.m.

temperatures owing to uncertainties involved in removing representative liquid samples from liquefied gas systems.

The equilibrium assembly and auxiliary equipment are shown schematically in Figure 1. The inner chamber of the cell, *A*, consists of a heavy walled cylindrical copper vessel with an internal volume of about 140 ml. For control of temperature, there is mounted around the cell two cylindrical shells, one of which, *B*, is electrically heated and the other, *C*, is cooled by means of liquid nitrogen pumps. The cell is insulated from the boiling nitrogen vapors by the insulated double walled vessel, *D*. The entire assembly is suspended in a large metal Dewar by means of stainless steel tube, *E*. Stirring of the cell contents is effected with a reciprocating magnetic stirrer, *F*, operated at 4 cycles per second whose shaft passes through the stainless steel suspension tube, *E*, to a magnetic core. The suspension tube also serves as a take-off line for the vapor sample stream. The liquid is withdrawn continuously at high velocity through copper coil, *G*, which is immersed in the liquid phase. The liquid is then passed through a short length of $\frac{1}{16}$ inch tubing and then into a large heated vaporization chamber. The feed gas is contacted with the outer body of cell *A* in order to attain the selected temperature, and then passed continuously into the liquid phase.

The temperature of the cell is regulated by counterbalancing the cooling produced by the liquid nitrogen pumps with a high-low heater control circuit. The sensing element for this control circuit is a 30 B & S gage bare wire copper-constantan thermocouple, *H*, one junction of which is immersed in the liquid phase contained in the cell and the other junction in a carefully prepared ice bath. The EMF produced by the couple is balanced through a K-3 potentiometer, and any deviation from the selected null point is amplified by a D.C. breaker amplifier which operates the relays for the high-low heater control circuit. Temperature control for the range -195° to -100° C. is $\pm 0.04^{\circ}$ C.

The liquid level in the cell is observed either by means of an illuminated peep sight (not shown) which is operable to 1000 p.s.i. at -195° C. or by a thermistor sensor, *I*, calibrated for the gas mixture. The level can be measured to one-half mm. or better with either technique. Control of liquid level is achieved by regulation of the flow rates of the feed and sample streams at constant temperature and pressure.

The thermocouple, *H*, suspended in cell *A* was calibrated using a platinum resistance thermometer previously calibrated by the National Bureau of Standards. Additionally, the couple, *H*, is calibrated at the selected temperature by measuring the vapor pressure of pure liquid nitrogen and using the temperature-vapor pressure relations reported by Hilsenrath and coworkers (4). The agreement between the two calibration techniques is within 0.2° C., which is taken as the accuracy of the temperature measurements.

Pressures measured with two gages to cover the range from atmospheric to 1000 p.s.i. Below 100 p.s.i.a., the accuracy of the pressure readings is ± 0.1 p.s.i.; from 100 to 1000 p.s.i., the accuracy is ± 1.0 p.s.i.

Consistent compositions for vapor and liquid samples over a wide range of sample flow rates established the condition of equilibrium and the reliability of the sampling procedure. Vapor sampling rates ranging from 0 to 250 ml. of gas per minute and liquid sampling rates (as gas) from 125 to 500 ml. per minute gave compositions which were independent of flow rate. As an example of the consistency of the data, the reliability of the mean for 30 samples of vapor containing a constant concentration of neon plus helium of 28.5 mole % (113.13° K., 30 atm.) is better than ± 0.15 mole %. For liquid samples of equal concentration taken within the range of flow rates stated above, the compositions are constant within the limits of precision of the mass spectrometer and range in average deviation from $\pm 2\%$ of composition at 1.0 mole % of helium to $\pm 10\%$ of composition at 0.03 mole % helium.

RESULTS

The experimental and correlated data for the binary and ternary solutions are contained in Figures 2 and 3. Each circle represents the average of three or more liquid and vapor sample sets taken at a single temperature and pressure for one gas feed mixture. The solid lines for the vapor phase compositions, drawn between the binaries of neon and helium in nitrogen at constant pressure, represent the best linear fit of the experimental data. The dashed lines for the liquid phase compositions were calculated at constant pressure by the semiempirical correlation procedure described below. Ternary compositions at each indicated pressure fall on the isobaric lines. The tie lines connect corresponding liquid and vapor compositions which are in equilibrium. The nitrogen concentrations in mole per cent for each phase are found by subtracting the sum of neon plus helium concentrations from 100.

The values for the vapor phase compositions, shown as the smaller solid circles in Figures 2 and 3, have an average deviation of ± 0.2 mole % at 8 mole % total vapor composition and ± 0.5 mole % at 95 mole % vapor composition, although the data at 50 atm. and 113.13° K. show a somewhat larger error. The experimental liquid phase concentrations which are shown as the larger circles in the figures have an average deviation ranging from 2% to 10% of liquid composition, the deviation value being dependent upon the concentration of neon or helium in the liquid phase. The source of the errors in determining the liquid phase compositions is primarily analytical and not due to nonequilibrium conditions or sampling procedure. As will be shown below, the correlation procedure developed here makes use of the greater reliability of the vapor composition data to assist in smoothing and correlating the less precise liquid composition data.

Equilibrium compositions were calculated at 5 atm. intervals of pressure for selected incremental values of the concentration of neon in the liquid phase. The calculations were made using the same relations derived for the correlation procedure. Tables II and III contain the calculated tie line data for the binary and ternary mixtures. Equilib-

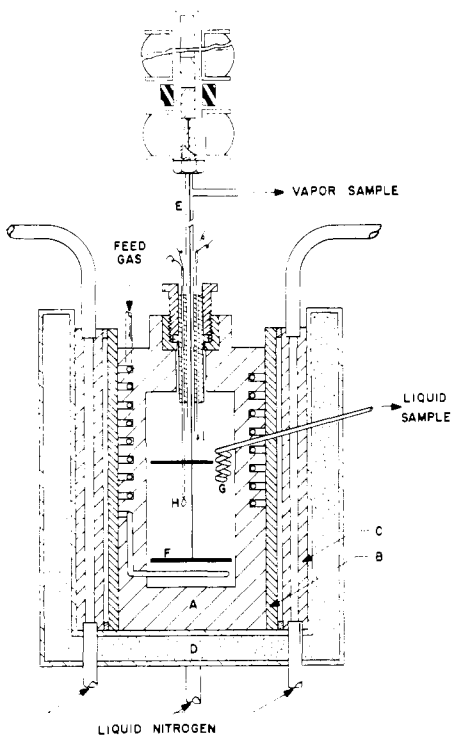


Figure 1. Phase equilibrium cell

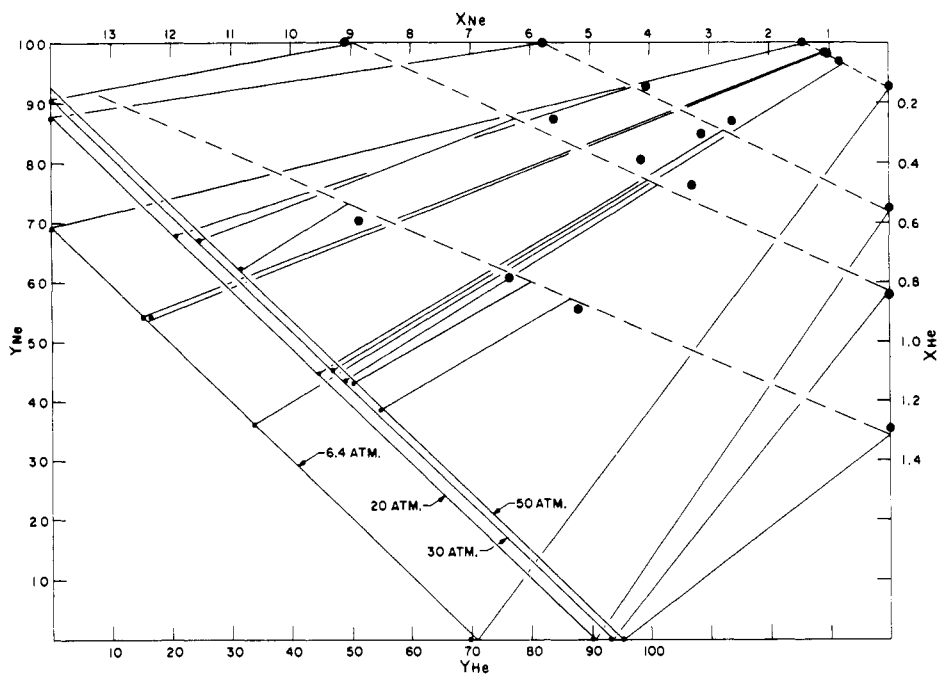


Figure 2. Isobars and tie lines for the He-N₂, Ne-N₂, and Ne-He-N₂ systems at 82.70° K. Compositions in mole per cent

rium composition data have also been calculated for selected incremental concentrations of helium in the liquid phase, and are available upon request from the author.

CORRELATION AND EXTENSION OF DATA

The relation between two phases in equilibrium is expressed by

$$x_i \gamma_i^l f_i^{o,l} = y_i \gamma_i^v f_i^{*v} \quad (1)$$

where x and y are the mole fraction compositions of the

liquid and vapor phases, respectively, γ is the activity coefficient and f is the fugacity of the pure component. Assuming as a first approximation that the gas mixture is an ideal mixture of real gases, then $\gamma_i^v = 1$. The partial fugacities of helium and neon, therefore, were computed for the gas phase using the empirically smoothed vapor compositions corresponding to the experimental points in Figures 2 and 3 and the fugacities of the pure gases. The latter fugacities were evaluated by a graphical integration procedure using the P - V - T data of Keesom (5) and Yendall (8).

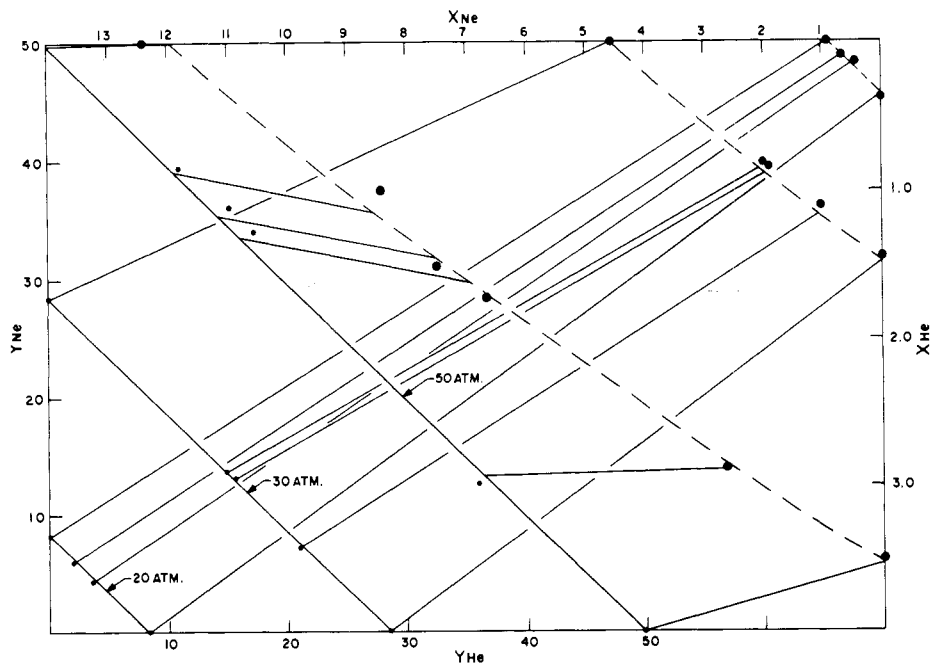


Figure 3. Isobars and tie lines for the He-N₂, Ne-N₂, and Ne-He-N₂ systems at 113.13° K. Compositions in mole per cent

Table II. Vapor-Liquid Composition Data for the Binary and Ternary Solutions of Neon (1) and Helium (2) Gases in Liquid Nitrogen, 82.70° K.

Compositions, Mole % ^a				Compositions, Mole % ^a			
x ₁	x ₂	y ₁	y ₂	x ₁	x ₂	y ₁	y ₂
5 Atm.				20 Atm.			
0.00	0.108	0.00	63.8	2.50	0.322	40.7	48.8
0.100	0.097	6.26	57.1	3.00	0.274	48.4	40.9
0.200	0.087	12.5	50.7	3.50	0.225	55.9	33.2
0.300	0.076	18.7	44.4	4.00	0.178	63.1	25.8
0.400	0.065	24.9	38.0	5.00	0.081	77.3	11.3
0.500	0.055	31.0	31.8	5.80	0.00	88.3	0.00
0.600	0.044	37.2	25.4	30 Atm.			
0.700	0.033	43.3	19.2	0.00	0.825	0.00	92.9
0.800	0.022	49.3	13.0	1.00	0.731	11.9	80.6
0.900	0.012	55.4	6.76	2.00	0.631	23.4	67.8
1.010	0.00	62.0	0.00	3.00	0.547	34.4	57.6
10 Atm.				4.00	0.456	45.0	46.7
0.00	0.268	0.00	81.8	5.00	0.366	55.0	36.5
0.050	0.260	1.62	80.1	6.00	0.274	64.6	26.6
0.200	0.247	6.45	75.2	7.00	0.180	74.0	17.0
0.500	0.218	15.9	65.5	8.00	0.092	82.5	8.30
0.800	0.187	25.5	55.6	8.96	0.00	90.6	0.00
1.10	0.156	34.8	46.1	40 Atm.			
1.40	0.115	44.1	36.6	0.00	1.07	0.00	93.5
1.70	0.094	53.2	27.3	1.00	0.990	9.46	84.9
2.00	0.063	62.2	18.0	2.00	0.900	18.6	75.5
2.30	0.031	71.1	8.91	3.00	0.809	27.4	66.5
2.60	0.00	79.8	0.00	4.00	0.718	35.8	57.9
15 Atm.				5.00	0.630	43.9	49.6
0.00	0.418	0.00	87.7	6.00	0.543	51.7	41.6
0.200	0.399	4.43	83.3	7.00	0.453	59.1	34.0
0.500	0.370	11.0	76.6	8.00	0.365	66.2	26.7
1.00	0.321	21.8	65.5	10.0	0.195	79.3	13.3
1.50	0.272	32.4	54.7	12.2	0.00	92.3	0.00
2.00	0.222	42.8	44.1	50 Atm.			
2.50	0.172	52.9	33.7	0.00	1.31	0.00	94.8
3.00	0.122	62.8	23.5	1.00	1.22	8.01	86.8
3.50	0.071	72.5	13.6	2.00	1.13	15.7	78.9
4.00	0.021	82.0	3.83	3.00	1.05	23.2	71.3
4.20	0.00	85.7	0.00	4.00	0.957	30.4	63.9
20 Atm.				5.00	0.870	37.2	56.9
0.00	0.560	0.00	90.5	7.00	0.700	50.1	43.7
0.50	0.514	8.48	81.8	9.00	0.527	62.0	31.5
1.00	0.467	16.8	73.3	11.0	0.360	72.6	20.6
1.50	0.418	25.0	64.9	13.0	0.193	82.5	10.5
2.00	0.369	32.9	56.8	15.4	0.00	92.7	0.00

^a Nitrogen concentrations obtained by subtracting the sum of the concentrations of components 1 and 2 from 100.

At a constant temperature and pressure, the liquid phase activity coefficients in Equation 1, as well as the product $\gamma_i^l f_i^{\circ l}$, are functions only of the concentrations of the components in a ternary solution. There is also indicated from Figures 2 and 3 that since there is a nearly linear relationship between the concentrations of components, from the binary of neon to that of helium, then the activity coefficient can be adequately expressed as a function of the concentration of a single component. For neon (gas 1) values for the term $y_1 f_1^{*v} / x_1$, were plotted as a function of the concentration of neon ranging from the binary of neon in nitrogen to the lowest experimental concentration of neon in the liquid phase. At each temperature and pressure, a linear equation was fitted to the data by least squares. The intercept at $x_1 = 0$ (binary of helium in nitrogen) is given by

$$K_1 = (\gamma_1^l f_1^{\circ l})_{x_1=0} \quad (2)$$

where K_1 is the Henry's Law constant at infinite dilution for neon in a solvent consisting of low concentrations of helium gas in liquid nitrogen, and $f_1^{\circ l}$ is the hypothetical fugacity of pure liquid neon under its vapor pressure P_1°

and at the temperature of the solution. From the slope of the curves, m , Equation 1 for neon then becomes

$$x_1 K_1 - m x_1^2 = y_1 f_1^{*v} \quad (3)$$

The constant K_1 in Equation 2 is a function of the total pressure of helium insofar as the activity coefficient changes with pressure. The effect of variations in the amount of dissolved helium on K_1 is assumed negligible since the concentration of helium is never larger than 3.5%. Then from the relation,

$$\ln K_1 = \ln f_1^{\circ l} \gamma_1^{\circ l} + \bar{V}_1 (P - P_1^\circ) / RT \quad (4)$$

smoothed values of K_1 are obtained from a plot of $\ln K_1$ vs. P . In Equation 4, $\gamma_1^{\circ l}$ is the activity coefficient at P_1° , and \bar{V}_1 is the partial molar volume of neon at infinite dilution, which is assumed to be independent of pressure. The values of K_1 obtained from the graph are given in Table IV as a function of pressure together with the values for the slope m . The data in Table IV were used with Equation 3 to calculate liquid compositions from the smoothed vapor compositions given in Figures 2 and 3.

Table III. Vapor-Liquid Composition Data for the Binary and Ternary Solutions of Neon (1) and Helium (2) Gases in Liquid Nitrogen, 113.13° K.

Compositions, Mole % ^a			
x_1	x_2	y_1	y_2
20 Atm.			
0.00	0.340	0.00	8.47
0.100	0.298	0.95	7.37
0.200	0.262	1.89	6.43
0.300	0.224	2.83	5.49
0.400	0.189	3.76	4.56
0.500	0.152	4.69	3.63
0.600	0.115	5.60	2.72
0.700	0.077	6.51	1.81
0.800	0.039	7.42	0.90
0.900	0.00	8.32	0.00
25 Atm.			
0.00	0.930	0.00	20.2
0.400	0.800	3.20	17.0
0.800	0.670	6.33	13.8
1.00	0.602	7.87	12.3
1.20	0.536	9.38	10.8
1.40	0.468	10.9	9.27
1.60	0.398	12.4	7.79
1.80	0.329	13.8	6.33
2.20	0.186	16.7	3.47
2.70	0.00	20.2	0.00
30 Atm.			
0.00	1.47	0.00	28.4
0.50	1.32	3.56	24.9
1.00	1.17	7.01	21.5
1.50	1.02	10.4	18.1
2.00	0.860	13.6	14.9
2.50	0.700	16.8	11.7
3.00	0.533	19.9	8.70
3.50	0.365	22.8	5.70
4.00	0.187	25.7	2.80
4.52	0.00	28.5	0.00
40 Atm.			
0.00	2.51	0.00	40.4
0.50	2.39	3.05	37.7
1.00	2.25	6.03	34.7
2.00	1.96	11.8	28.9
3.00	1.68	17.2	23.5
4.00	1.39	22.3	18.4
5.00	1.09	27.1	13.6
6.00	0.768	31.7	9.00
7.00	0.440	35.9	4.80
8.22	0.00	40.7	0.00
50 Atm.			
0.00	3.54	0.00	49.9
1.00	3.26	5.49	44.1
2.00	3.00	10.8	38.8
3.00	2.73	15.7	33.9
4.00	2.45	20.6	29.0
5.00	2.17	25.1	24.5
6.00	1.88	29.4	20.2
8.00	1.29	37.3	12.3
10.0	0.65	44.2	5.40
11.9	0.00	49.6	0.00

^a Nitrogen concentrations obtained by subtracting the sum of the concentrations of components 1 and 2 from 100.

The procedure used for determining the function for the change in activity coefficient with concentration of helium was the same as for neon with certain modifications. Equation 1 as applied to helium can be written in a form similar to that for neon,

$$-x_2 k_2 + F(x_2) = y_2 f_2^{*v} \quad (5)$$

where $F(x_2)$ is an empirical function of the concentration of helium. Figures 2 and 3 indicate a linear relationship between the concentrations of neon and helium in the vapor phase. Solving Equation 5 with Equation 3, gives a linear

Table IV. Constants for Equation 3 (Neon) and Equation 7 (Helium) Used for Computations

T , ° K.	P , Atm.	K_1 , Atm.	k_2 , Atm.	m , Atm.	b , Atm.	c , Atm.
82.70	6.4	313	328	6.37	3204	895
	20.0	331	363			
	30.0	345	390			
	50.0	375	452			
113.13	20.0	189	195	5.90	670	107
	30.0	214	224			
	50.0	277	299			

expression relating the vapor phase compositions at constant temperature,

$$y_1 = \left(-\frac{K_1 f_2^{*c}}{k_2 f_1^{*c}} \right) y_2 + I \quad (6)$$

where the values for k_2 and the intercept I depend upon the pressure selected. At each temperature, the slopes of the y_1 vs. y_2 plots were found to be independent of pressure (Figures 2 and 3) and equal to -0.976 at 82.70° K. and -1.000 at 113.13° K. From the Henry's Law constant for neon given in Table IV and the fugacities of the pure gases, values of k_2 in Equation 6 were calculated. Values for the term $F(x_2)$ in Equation 5 were calculated from the data at constant temperature for all experimental pressures, and plotted as a function of x_2 . The least squares fit of the data gave for Equation 5 the relation for helium,

$$x_2(b - k_2) + cx_2^2 = y_2 f_2^{*c} \quad (7)$$

The correlated liquid phase data in Figures 2 and 3 (dashed lines) were calculated by substituting into Equations 3 and 7 the data of Table IV and the smoothed vapor compositions which correspond as closely as possible to the experimental vapor composition points contained in the figures. Tie lines were then drawn between the calculated liquid compositions and these vapor compositions.

The smoothed values for the concentrations of neon and helium in the liquid phase of the binary solutions (see Tables II and III) were nearly linear functions of pressure. These calculated liquid composition data were plotted against total pressure, and liquid compositions at 5 or 10 atm. increments were read from the graphs. The corresponding vapor compositions for the binary solutions in Tables II and III were then calculated from Equations 3 and 7.

Beginning from the calculated binary composition data, values of decreasing mole fraction of neon in the ternary liquid mixtures were selected, and the corresponding values for the vapor compositions were read from large sized xy graphs prepared from Equation 3 for neon. Tie line vapor compositions for the helium in the ternary mixtures were calculated from Equation 6 or read from large plots of Figures 2 and 3, and corresponding liquid compositions for helium on the tie lines were taken from xy graphs prepared from Equation 7. The smoothed and interpolated binary and ternary composition data are collected in Tables II and III.

NOMENCLATURE

- 1 = neon
- 2 = helium
- x_i = mole % of component i in the liquid phase
- y_i = mole % of component i in the vapor phase
- γ_i^{sl} = activity coefficient in liquid phase at the temperature and pressure of the solution
- γ_i^{sl} = activity coefficient in the liquid phase at pressure P_l and the temperature of the solution
- γ_i^{sv} = activity coefficient in vapor phase
- f^* = fugacity of pure component at the temperature and pressure of the solution

f° = fugacity of pure component at its vapor pressure and the temperature of the solution
 f_i = partial fugacity of component in mixture
 K_1 = Henry's constant for neon
 V_i = partial molar volume of component i at infinite dilution
 P = total pressure of the solution
 P° = vapor pressure of pure component
 m, b, c = empirical constants for Equations 3 and 7
 $F(x_2)$ = a function of the concentration of helium
 k_2 = a pressure dependent empirical constant for helium

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Phase Equilibria in Hydrocarbon Systems

Phase Behavior in the n -Butane- n -Decane System

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Measurements of the composition of the gas phase in heterogeneous mixture of n -butane and n -decane were made throughout the two-phase region at temperatures between 160° and 460° F. These new measurements coupled with earlier volumetric investigations serve to establish the phase behavior of this binary system pressures up to 700 pounds per square inch in the temperature interval between 160° and 460° F. The results are presented in graphical and tabular form.

STUDIES OF THE volumetric and phase behavior of hydrocarbon systems remain of importance to the petroleum industry since satisfactory methods of predicting the behavior of multicomponent hydrocarbon systems of interest to the industry continue to be based upon experimental investigation. The volumetric behavior of n -butane has been investigated in detail (2, 3, 5, 6). Similarly, the volumetric behavior of decane in the liquid phase has been studied in some detail (8, 14). The vapor pressures of n -butane (6) and of decane (8) have also been established. Measurements of the volumetric behavior of mixtures of n -butane and decane in the liquid phase have been carried out at pressures up to 10,000 p.s.i.a. in the temperature interval between 100° and 460° F. (11). The properties of the bubble-point liquid have also been studied (11). However, there has not been any investigation of the composition of the gas phase in heterogeneous mixtures of n -butane and n -decane at elevated pressures and temperatures. In order to complete the investigation of the phase behavior of this system, a study of the composition of the gas phase in heterogeneous mixtures of n -butane and n -decane has been carried out at pressures throughout the two-phase region at temperatures between 160° and 460° F.

APPARATUS AND PROCEDURE

The apparatus and techniques employed in this investigation are similar to those used in earlier investigations (13). The heterogeneous mixtures of n -butane and n -decane were confined over mercury in a stainless steel container and samples of the gas phase were withdrawn under isobaric, isothermal conditions. The composition of the material

withdrawn was determined by a partial condensation technique (7) carried out at temperatures of solid carbon dioxide in equilibrium with acetone. After separation the relative quantities of n -decane and n -butane were determined by conventional gravimetric techniques (7).

The temperature of the heterogeneous equilibrium was related to the international platinum scale by use of a strain-free, platinum resistance thermometer. The behavior of this instrument was compared with the indications of a similar device calibrated by the National Bureau of Standards. Experience over a period of several decades indicates the temperature of the stainless steel container was known within 0.1° F. relative to the international platinum scale throughout the temperature interval between 160° and 460° F.

Pressure was determined by means of a balance whose construction has been described in some detail (12). This instrument has been calibrated at least annually against the vapor pressure of samples of carbon dioxide at the ice point. The variation in calibration of this balance over a 20-year period has been less than 0.05% (10). It appears that the pressures were established within 0.02% or 0.2 p.s.i. throughout the entire range of this investigation. It was not necessary to determine the volumetric behavior of the samples investigated during this series of measurements.

MATERIALS

The n -butane utilized in this study was obtained from the Phillips Petroleum Co. as research grade and reportedly contained less than 0.001 mole fraction of impurities. The change in vapor pressure corresponding to a change in qual-