

# Vapor Pressure of Propylene Oxide

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The saturated vapor pressure of propylene oxide in the temperature range 20° to 70° C. has been determined. Between 40° and 70° C., the results are correlated by the equation

$$\log P = 7.658 - \frac{1472}{T}$$

**S**ATURATED VAPOR PRESSURES of propylene oxide at temperatures up to 48.5° C. have been reported in the literature (2, 3, 4). The correlation up to a temperature of about 40° C. given by Kireev and Popov (3) is:

$$\log P = -\frac{1722.73}{T} + 8.48693 \quad (1)$$

where  $T$  = absolute temperature, °K.

The increasing interest in this material has prompted the confirmation and extension of the data.

## APPARATUS AND EXPERIMENTAL METHOD

The apparatus, which is essentially a modified Reid vapor pressure apparatus (1) consisted of a 150-ml. chromium-plated brass cylindrical pressure vessel, immersed in a constant temperature bath. A mercury manometer was connected to the vessel by a flexible stainless steel tube. A "tee" junction in this line, having a valve with a connection to a vacuum pump via a cold trap (solid carbon dioxide), enabled the apparatus to be exhausted prior to the determination of vapor pressures.

The vessel and its contents of propylene oxide were held at constant temperature until equilibrium had been established when the level of mercury in the manometer was noted. Corrections were made for barometric pressure and the effects of liquid condensed in the manometer. Accuracy of pressure measurement was better than 0.25% while the temperature was recorded to  $\pm 0.1^\circ$  C. The purity of the propylene oxide (I.C.I.Ltd., H.O.C. Division) is greater than 99.9%.

## RESULTS

The results, given in Table I, when plotted graphically, gave a straight-line relationship up to approximately 40° C. and a slightly different straight-line plot between 40° C. and 70° C.

Table I. Saturated Vapor Pressure of Propylene Oxide at Different Temperatures

Temp., ° C.	Vapor Pressure, Mm. Hg	Temp., ° C.	Vapor Pressure, Mm. Hg
19.0	416	50.2	1265
27.0	574	54.0	1452
36.3	806	59.2	1694
40.8	950	64.7	1994
46.8	1157	71.8	2403

The data obtained in the present work agree well with the data of Kireev and Popov in the range up to about 35° to 40° C.

The results presently reported in the range 40° to 70° C. are correlated by the following equation.

$$\log P = 7.658 - \frac{1472}{T} \quad (2)$$

## ACKNOWLEDGMENT

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# Phase Behavior in the Hydrogen-Cyclohexane System

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**T**HERE HAS BEEN a gradual accumulation of information concerning the phase behavior of binary systems involving hydrogen and a paraffin hydrocarbon. However, data upon the hydrogen-cyclohexane system are limited. An experimental study involving measurements of the specific volume of three mixtures of hydrogen and cyclohexane at four temperatures between 100° and 280° F. was carried out at pressures as high

as 10,000 p.s.i.a. The composition of the coexisting gas phase in heterogeneous mixtures of hydrogen and cyclohexane was determined throughout the above-mentioned temperature and pressure interval. This binary system followed the trends of other hydrogen-paraffin hydrocarbon systems, and the critical pressure was above 10,000 p.s.i.a. for all of the temperatures investigated.

Measurements of the composition of the coexisting phases in the hydrogen-cyclohexane system have been carried out at temperatures between 100° and 280° F. and at pressures up to 10,000 p.s.i.a. Limited measurements of the specific volume of the liquid phase were made throughout the above-mentioned temperature interval for three mixtures of hydrogen and cyclohexane. The experimental results are presented in tabular form, and the properties of the coexisting phases have been recorded.

Studies of the phase behavior of the hydrogen-propane (4) and of the hydrogen-*n*-hexane system (6) are available. Benham, Katz, and Williams (2) reviewed the general characteristics of hydrogen-light hydrocarbon systems and presented a tentative method of predicting the phase behavior of such mixtures. Recently, data concerning the liquid-vapor equilibrium of the hydrogen-cyclohexane system became available at temperatures of 150° and 250° F. (11).

The volumetric behavior of cyclohexane is known (8), as is the vapor pressure (12). Likewise, the volumetric behavior of hydrogen has been established with a relatively high degree of accuracy (5, 12). As a result of the absence of data upon the volumetric or phase behavior of mixtures of hydrogen and cyclohexane, an investigation was started nearly a decade ago to establish the composition of the coexisting phases of mixtures of hydrogen and cyclohexane at pressures up to 10,000 p.s.i.a.

Table I. Experimental Volumetric Measurements

Pressure, P.S.I.A.	Volume, Cu. Ft./Lb.	Pressure, P.S.I.A.	Volume, Cu. Ft./Lb.	Pressure, P.S.I.A.	Volume, Cu. Ft./Lb.	Pressure, P.S.I.A.	Volume, Cu. Ft./Lb.
100° F.		160° F.		220° F.		280° F.	
Mole Fraction Hydrogen 0.0502							
(1855) <sup>a</sup>		(1610)		(1350)		(1080)	
100.8	0.062144	1339.7	0.022490	144.21	0.063821	189.8	0.062746
1052.3	0.023090	1461.0	0.022260	832.6	0.025540	851.6	0.026303
1320.0	0.022119	1501.8	0.022150	995.2	0.024467	1012.6	0.025064
1635.4	0.021538	1531.7	0.022000	1161.1	0.023657	1063.2	0.024760
1860.98	0.021177	1633.9	0.021922	1214.6	0.023492	1099.0	0.024483
2065.2	0.021037	2014.4	0.021802	1309.7	0.023151	1202.7	0.024349
2328.1	0.020983	3114.2	0.021508	1418.2	0.023031	1311.5	0.024270
2664.9	0.020924	4021.8	0.021419	1515.6	0.022980	1535.8	0.024154
3001.6	0.020813	4855.2	0.021338	1630.6	0.022915	1815.4	0.024015
4858.2	0.020390			2049.6	0.022783	2528.7	0.023697
		933.9	0.02416	3018.1	0.022505	3484.5	0.023335
		1077.1	0.023502	4074.9	0.022237	4635.5	0.023021
		1235.9	0.022943	4887.6	0.022099		
		1370.9	0.022504				
		1731.3	0.021927				
		1850.0	0.021892				
		1957.6	0.021842				
		2069.2	0.021792				
		2318.8	0.021764				
		2454.1	0.021719				
		2554.0	0.021657				
		3018.3	0.021485				
		3601.8	0.021375				
		4082.1	0.021385				
		4594.1	0.021332				
		4875.6	0.021325				
Mole Fraction Hydrogen 0.1338							
(5290) <sup>a</sup>		(4482)		(3670)		(3010)	
118.0	0.120469	279.2	0.067259	349.8	0.063327	1021.0	0.035382
265.7	0.063455	1637.8	0.027115	1203.3	0.031308	1436.3	0.030509
1470.0	0.027009	2639.7	0.024033	1464.3	0.029043	1923.9	0.027575
1904.4	0.025104	3535.3	0.022846	2027.0	0.026311	2332.1	0.026069
2010.3	0.024766	3738.6	0.022659	2662.2	0.024602	2504.8	0.025569
3007.8	0.022867	3912.9	0.022498	3038.7	0.023901	2618.1	0.025261
3397.7	0.022339	4012.7	0.022399	3243.3	0.023575	2717.7	0.025061
3831.6	0.022010	4112.2	0.022327	3406.0	0.023384	2822.2	0.024847
4102.8	0.021778	4209.1	0.022242	3512.1	0.023297	3033.2	0.024548
4446.1	0.021598	4329.0	0.022187	3623.3	0.023183	3228.5	0.024455
4659.3	0.021555	4516.0	0.022086	3763.1	0.023115	3408.3	0.024377
4816.9	0.021423	4985.7	0.022019	3886.6	0.023088	3508.1	0.024317
4998.4	0.021376	5990.1	0.021867	4161.4	0.023012	4035.1	0.024138
5194.6	0.021248	6921.7	0.021734	4988.9	0.022830	5133.8	0.023771
5345.2	0.021130	7479.1	0.021665	6094.5	0.022636	5970.3	0.023543
5532.3	0.021056			7099.1	0.022470	7010.3	0.023299
6027.3	0.021090			7415.9	0.022374		
7096.8	0.020947						
7702.0	0.020966						

(Continued on page 27)

in the temperature interval between 100° and 280° F. In addition, a limited study of the volumetric behavior of mixtures of hydrogen and cyclohexane in the liquid phase has been made. This latter work was carried out within the composition interval between pure cyclohexane and 0.022 mole fraction hydrogen. The latter data were employed to establish the bubble-point pressures of these mixtures throughout the above-described range of temperature and pressure.

#### METHODS AND APPARATUS

The equipment employed in determining the volumetric behavior of this system has been described (9). In principle, it consists of a stainless steel cylinder in which the hydrocarbon and hydrogen were confined over mercury. Mechanical agitation within the chamber was provided and this chamber was immersed in an agitated oil bath. The samples were withdrawn from the gas phase under isobaric, isothermal conditions, and the quantity of cyclohexane present was determined by partial condensation techniques (7). A description of the apparatus used for the gas phase withdrawals is available (10).

Determination of the compositions of the liquid phase by direct analysis was not attempted because of the relatively small amount of hydrogen present in the liquid phase at the lower pressures. For this reason, the discontinuous change in slope of the specific volume-pressure diagram was employed as a means of determining the bubble-point pressure for a system

of known composition. The equipment employed for these measurements has been described (9). In principle, this equipment was similar to that used for confining the samples for the composition of the coexisting phases. Gravimetric techniques were employed for the introduction of the cyclohexane, and the quantity so introduced was also established from limited volumetric measurements upon the pure cyclohexane. Hydrogen was introduced utilizing volumetric methods to establish the quantity so introduced. The quantity of cyclohexane employed probably was known within 0.15%, while the quantity of hydrogen was known within 0.25% of the total quantity added. Mechanical agitation was provided to permit the attainment of physical equilibrium.

The pressures for both the composition of the coexisting gas phase and the volumetric measurements were determined by means of a balance involving a piston-cylinder combination (9). Calibration of this instrument over a period of two decades against the vapor pressure of carbon dioxide at the ice point (8) has indicated a probable uncertainty in the measured values of the pressure of 0.15% or 0.1 p.s.i., whichever is the larger measure of uncertainty. Temperature was determined by means of a strain-free platinum resistance thermometer. Its indications were compared against those of a similar instrument calibrated by the National Bureau of Standards. The temperature for both the gas-phase composition and volumetric measurements was known relative to the international platinum scale within 0.02° F. throughout the temperature interval between 100° and 280° F.

Table I. Experimental Volumetric Measurements (Continued)

Pressure, P.S.I.A.	Volume, Cu. Ft./Lb.	Pressure, P.S.I.A.	Volume, Cu. Ft./Lb.	Pressure, P.S.I.A.	Volume, Cu. Ft./Lb.	Pressure, P.S.I.A.	Volume, Cu. Ft./Lb.
100° F.		160° F.		220° F.		280° F.	
Mole Fraction Hydrogen 0.2203							
(9520) <sup>a</sup>		(7769)		(6605)		(5610)	
473.4	0.063006	446.2	0.072015	600.2	0.063497	543.6	0.062498
1431.7	0.033810	1316.3	0.037015	1688.1	0.034654	698.0	0.049446
5328.2	0.023215	2336.4	0.029254	2733.1	0.028751	1429.1	0.039512
6270.3	0.022606	4420.9	0.024573	3759.0	0.026193	2356.8	0.031399
6752.9	0.022362	5616.1	0.023525	4741.0	0.024777	3415.6	0.027646
7185.6	0.022224	6461.0	0.023010	5544.0	0.023996	4379.3	0.025823
7609.6	0.022071	7082.4	0.022703	6015.1	0.023713	5242.9	0.024825
8056.1	0.021931	7489.2	0.022520	6432.5	0.023501	5353.1	0.024770
8318.4	0.021816	7726.9	0.022453	6547.9	0.023460	5427.0	0.024698
8546.7	0.021802	7843.4	0.022412	6629.9	0.023416	5496.9	0.024651
8707.5	0.021780	7900.1	0.022406	6850.1	0.023375	5597.1	0.024582
8809.3	0.021738	8015.2	0.022368	6946.4	0.023360	5908.3	0.024537
8932.6	0.021712	8155.9	0.022354	7088.7	0.023320	7074.5	0.024212
9402.3	0.021606	8315.5	0.022333	7276.3	0.023273	4082.5	0.026257
9545.5	0.021568	8586.2	0.022284	8162.5	0.023137	4607.8	0.025505
9770.9	0.021540	9034.0	0.022229	9097.4	0.022978	4884.7	0.025127
9938.3	0.021536	9133.4	0.022215	9559.1	0.022939	5029.3	0.024952
475.2	0.063198	9617.0	0.022144	9807.0	0.022901	5107.3	0.024905
655.6	0.050915	9711.0	0.022149			6121.3	0.024467
4214.3	0.024124					7234.5	0.024191
6227.5	0.022615					8001.3	0.023983
7181.3	0.022235					9010.8	0.023792
8149.2	0.021890					9282.4	0.023755
8676.3	0.021775						
8968.4	0.021690						
9024.1	0.021668						
9153.8	0.021616						
9259.4	0.021547						
9301.8	0.021552						
9421.5	0.021484						
9440.4	0.021473						
9575.2	0.021461						
9887.8	0.021422						
9958.4	0.021415						

<sup>a</sup> Values in parentheses represent bubble-point pressures expressed in p.s.i.a.

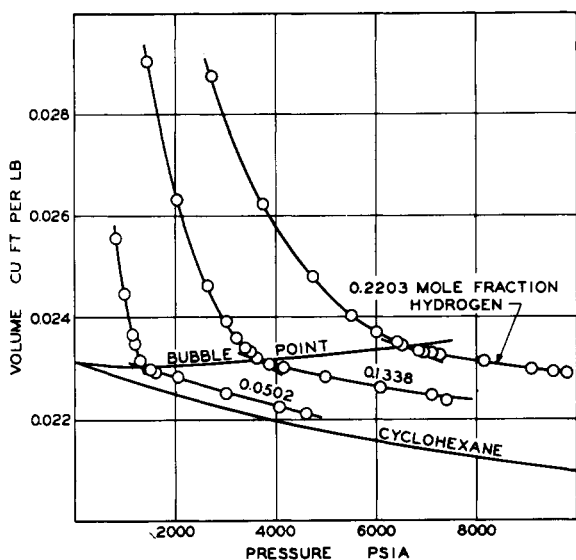


Figure 1. Experimental volumetric measurements at 220° F.

The investigation was not extended to higher temperatures because of thermal rearrangement of the cyclohexane at temperatures significantly in excess of 280° F. This occurred at a sufficiently rapid rate as to render either volumetric or phase composition measurements of questionable accuracy. It was not feasible to extend the measurements to lower temperatures because of the proximity of the freezing point of cyclohexane (1).

The analytical techniques yielded a reproducibility of the order of 0.0002 mole fraction and the uncertainty in measurement appears to be of the order of 0.001 mole fraction. Throughout the range of temperatures and pressures covered in this investigation, the volumetric measurements in the liquid phase are believed to involve uncertainties of the order of 0.02% and the bubble-point pressures were established with a precision of 0.5 p.s.i.

#### MATERIALS

The cyclohexane was obtained as research grade from the Phillips Petroleum Co., which reported it to contain less than 0.0006 mole fraction of impurities. The hydrocarbon was dried over metallic sodium and solidified at liquid nitrogen temperatures and maintained at a relatively high vacuum for an extended period to aid in the removal of noncondensable gases. The specific weight at 77° F. and atmospheric pressure was 48.307 pounds per cubic foot for an air-free sample as compared with a value of 48.311 pounds per cubic foot reported (1) for an air-saturated sample at the same temperature. The index of refraction relative to the D-lines of sodium at 77° F. was 1.42355 as compared with a value of 1.42354 reported for an air-saturated sample (1). The material used in this investigation contained less than 0.002 mole fraction of impurities and these impurities are probably hydrocarbons containing six carbon atoms per molecule.

The hydrogen, obtained from a commercial manufacturer who had prepared it by electrolytic techniques, was reported to contain 0.00002 mole fraction of oxygen and 0.001 mole fraction of materials other than hydrogen and water. The gas was passed through a coil immersed in liquid nitrogen, through a chamber containing platinum wire heated to approximately 800° F., and again through a second coil immersed in liquid nitrogen. This treatment was followed by contact with anhydrous calcium sulfate and activated charcoal at pressures in excess of 500 p.s.i. A mass spectrographic analysis of the purified hydrogen indicated it to contain less than 0.0014 mole fraction of materials other than hydrogen.

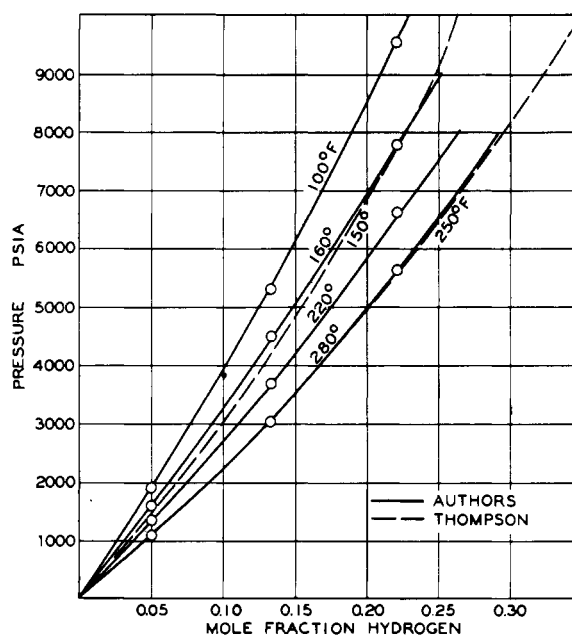


Figure 2. Pressure-composition diagram for bubble point

#### EXPERIMENTAL RESULTS

Measurements of the volumetric behavior of three mixtures of hydrogen and cyclohexane were made at pressures up to 10,000 p.s.i.a. A sample of the experimental results at 220° F. is presented in Figure 1. The information shown in this figure is typical of that obtained at the other three temperatures investigated. The corresponding values of the specific volume of cyclohexane have been included. Results of these volumetric measurements are recorded in Table I for each of the three mixtures investigated experimentally. The bubble-point pressures obtained from the isothermal discontinuities are also included in this table.

Figure 2 shows the bubble-point pressure of the hydrogen-cyclohexane system as a function of mole fraction hydrogen for temperatures between 100° and 280° F. The corresponding values from the work of Thompson and Edmister (11) have been included. The curvature shown in Figure 2 for the 150° F. isotherm results from following the experimental points of Thompson. The experimentally determined compositions of the dew-point gas are set forth in Table II for each temperature of measurement.

Table II. Composition of Gas Phase in Heterogeneous System

Pressure, P.S.I.A.	Mole Fraction Hydrogen	Pressure, P.S.I.A.	Mole Fraction Hydrogen
100° F.		220° F.	
30.0	0.9113	249.5	0.8742
81.3	0.9585	507.4	0.9341
249.3	0.9852	1004.0	0.9626
898.4	0.9952	2012.5	0.9762
2740.7	0.9958	2725.8	0.9799
3292.8	0.9966	4318.5	0.9823
5470.2	0.9976	6086.5	0.9836
7857.9	0.9957	8157.8	0.9834
160° F.		280° F.	
205.8	0.9433	779.2	0.8993
450.3	0.9717	1604.4	0.9420
986.4	0.9850	3063.8	0.9606
1989.8	0.9900	6154.2	0.9680
1993.0	0.9901	7158.9	0.9681
4030.7	0.9917	9531.7	0.9684
6146.9	0.9920		
9280.0	0.9920		

Table III. Some Properties of the Coexisting Phases

Pressure, P.S.I.A.	Mole Fraction Hydrogen		Equilibrium Ratio		Volume Cu. Ft./Lb. Mole Bubble Point
	Dew point	Bubble point	Hydrogen	Cyclohexane	
100° F.					
3.2 <sup>a</sup>	0.0000	0.0000	...	1.00000	1.775
500	0.9921	0.0135	73.58	0.00800	1.748 <sup>b</sup>
1000	0.9949	0.0269	37.00	0.00526	1.724
1500	0.9964	0.0400	24.89	0.00373	1.700
2000	0.9969	0.0530	18.80	0.00330	1.678
2500	0.9970	0.0658	15.16	0.00320	1.656
3000	0.9971	0.0784	12.72	0.00313	1.635
3500	0.9972	0.0906	11.00	0.00302	1.615
4000	0.9973	0.1028	9.703	0.00300	1.598
4500	0.9973	0.1145	8.711	0.00309	1.578
5000	0.9972	0.1260	7.914	0.00324	1.560
6000	0.9970	0.1480	6.736	0.00348	1.525
7000	0.9967	0.1692	5.891	0.00401	1.492
8000	0.9960	0.1893	5.261	0.00496	1.462
160° F.					
10.9 <sup>a</sup>	0.0000	0.0000	...	1.0000	1.851
500	0.9734	0.0157	61.97	0.0270	1.821 <sup>b</sup>
1000	0.9851	0.0316	32.00	0.0153	1.791
1500	0.9888	0.0473	20.91	0.0119	1.762
2000	0.9903	0.0627	15.80	0.0104	1.735
2500	0.9912	0.0774	12.80	0.00956	1.709
3000	0.9915	0.0927	10.70	0.00933	1.683
3500	0.9919	0.1068	9.286	0.00911	1.660
4000	0.9920	0.1215	8.163	0.00912	1.635
4500	0.9920	0.1357	7.311	0.00924	1.613
5000	0.9920	0.1494	6.639	0.00944	1.591
6000	0.9920	0.1760	5.637	0.00972	1.550
7000	0.9919	0.2020	4.910	0.00102	1.511
8000	0.9918	0.2273	4.374	0.0106	1.47
9000	0.9917	0.2520	3.936	0.0111	...
220° F.					
28.4 <sup>a</sup>	0.0000	0.0000	...	1.0000	1.945
1500	0.9350	0.0189	49.42	0.0662	1.906 <sup>b</sup>
1000	0.9622	0.0384	25.18	0.0393	1.868
500	0.9716	0.0574	16.93	0.0301	1.832
2000	0.9763	0.0760	12.84	0.0257	1.797
2500	0.9789	0.0941	10.40	0.0233	1.764
3000	0.9804	0.1119	8.762	0.0221	1.733
3500	0.9815	0.1281	7.660	0.0212	1.704
4000	0.9822	0.1446	6.792	0.0208	1.676
4500	0.9827	0.1599	6.144	0.0206	1.649
5000	0.9830	0.1758	5.593	0.0207	1.623
6000	0.9832	0.2058	4.777	0.0211	1.574
7000	0.9834	0.2362	4.163	0.0217	1.53
8000	0.9835	0.2644	3.719	0.0225	...
9000	0.9832	...	...	...	...
280° F.					
62.0 <sup>a</sup>	0.0000	0.0000	...	1.000	2.055
500	0.8582	0.0224	38.26	0.145	2.009 <sup>b</sup>
1000	0.9196	0.0466	19.61	0.0843	1.962
1500	0.9402	0.0697	13.48	0.0643	1.917
2000	0.9503	0.0914	10.41	0.0547	1.873
2500	0.9564	0.1118	8.556	0.0491	1.834
3000	0.9603	0.1340	7.248	0.0458	1.793
3500	0.9630	0.1505	6.400	0.0435	1.759
4000	0.9651	0.1673	5.721	0.0419	1.725
4500	0.9664	0.1858	5.202	0.0412	1.691
5000	0.9673	0.2020	4.789	0.0410	1.659
6000	0.9681	0.2343	4.148	0.0417	1.598
7000	0.9681	0.2652	3.633	0.0434	1.55
8000	0.9683	0.2919	3.244	0.0442	...
9000	0.9684	...	...	...	...

<sup>a</sup> Vapor pressure of cyclohexane.<sup>b</sup> Bubble-point volumes are estimated and subject to larger uncertainties.

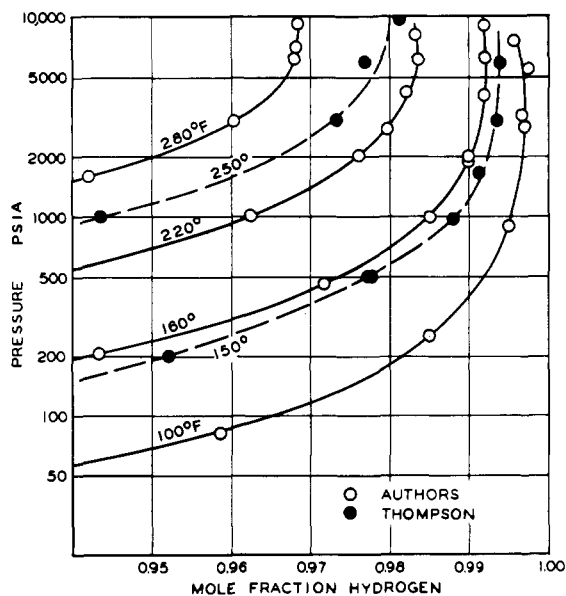


Figure 3. Composition of coexisting gas phase

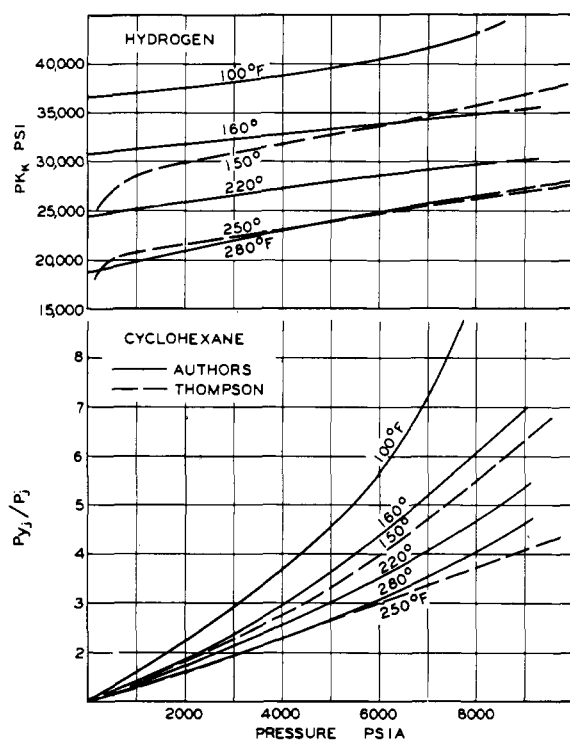


Figure 4. Equilibrium ratios for hydrogen and cyclohexane

Figure 3 depicts the experimentally determined composition of the coexisting gas phase in samples of heterogeneous mixtures of hydrogen and cyclohexane. A number of duplicate measurements were made, and these yielded a degree of agreement which would not permit the points to be differentiated from those shown on the diagram. The standard error of estimate of the experimental points from the smooth curves was 0.0007 mole fraction, assuming all of the uncertainty in the composition and none in the measured values of pressure and temperature. Following conventional smoothing techniques, the compositions

of the coexisting phases in the hydrogen-cyclohexane system have been evaluated and are presented for even values of pressure and temperature in Table III. Included also are the equilibrium ratios for hydrogen and cyclohexane. As a result of the number of significant figures reported for dew-point gas compositions, some discrepancies in the tabulated values of the equilibrium ratio for cyclohexane, computed from the compositions, may exist. In the upper part of Figure 4 is shown the product of the pressure and the equilibrium ratio,  $PK_k$ , for hydrogen in the hydrogen-cyclohexane system. These data were compared with similar information based upon the measurements of Thompson and Edmister (11) and a comparison has been indicated in the upper part of Figure 4. The quantity  $Py_j/P''_j$  has been used to portray the phase behavior of cyclohexane and is shown in the lower part of Figure 4. In this expression  $P$  is the pressure,  $y_j$  is the mole fraction of cyclohexane in the gas phase, and  $P''_j$  is the vapor pressure of cyclohexane. In the case of relatively nonvolatile materials such a function yields a smaller variation with respect to state than the use of the more conventional equilibrium ratio. In the lower part of Figure 4 the measurements of Thompson and Edmister (11) are shown for comparison.

The current measurements and those of Thompson and Edmister (11) appear to agree within an uncertainty in mole fraction of approximately 0.0066 in the gas phase and 0.0083 in the liquid phase for temperatures of 150° and 250° F. The current investigation extended over a sufficiently limited range of temperatures so as not to permit the evaluation of the behavior at higher temperatures approaching the critical state of cyclohexane.

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