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*Technical Note*

*No. 109*

*Boulder Laboratories*

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A COMPILATION OF THE PHYSICAL  
EQUILIBRIA AND RELATED PROPERTIES  
OF THE  
HYDROGEN-HELIUM SYSTEM

BY

D. E. DRAYER AND T. M. FLYNN



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U. S. DEPARTMENT OF COMMERCE  
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# NATIONAL BUREAU OF STANDARDS

## *Technical Note*

June 1961

A Compilation of the  
Physical Equilibria and Related Properties  
of the  
Hydrogen-Helium  
System

by

Dennis E. Drayer

and

Thomas M. Flynn

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## Abstract

Published data have been used to calculate K-factors for the helium-hydrogen system over the range of  $17.4^{\circ}$  to  $21.8^{\circ}$  K and 2 to 32 atmospheres pressure. K-factors are presented graphically for three isotherms over this range. A bibliography of approximately 290 references is also presented on related properties for this system and for the pure components.



## 1. Introduction

### 1.1 Purpose

Hydrogen is frequently found as a companion gas in helium sources. If this helium is to be liquefied, a most rigorous purification is required to prevent blocking of the liquefier by solid impurities. The exacting design of such a helium purification system requires a knowledge of the physical equilibria behavior of the hydrogen-helium system.

As an initial step in the study of the physical equilibria of the hydrogen-helium system, a literature survey was made. This paper presents the results of that survey, which includes the current knowledge of the vapor-liquid equilibria, and selected properties of the pure components.

### 1.2 Organization

The information is presented in three principal parts: (1) physical equilibria with major emphasis on vapor-liquid equilibria; (2) properties related to physical equilibria; and (3) a bibliography of references. Some discussion is presented with Part (1). The information of Part (2) is presented in tabular form showing the reference where important data are to be found. Part (3), the Bibliography, lists the references alphabetically by author.

### 1.3 Scope

A literature search, as summarized in NBS Technical Note No. 56, revealed most of the pertinent data. Such data were abstracted, and presented in the form of K-factor charts, and as a bibliography of references for related areas of interest. The areas searched are presented in the above reference and will not be enumerated here. Generally speaking, the literature was searched extensively and includes articles published up to 1960. This report does, however, include additional references obtained from a search of Volumes 1 through 5 of the series "Advances in Cryogenic Engineering".

## 2. Survey of Literature

Only one reference was found relating to the vapor-liquid equilibria for the helium-hydrogen system. This reference, Smith (247), presented pressure-concentration data for three temperatures, 17.4°, 20.4° and 21.8°K.

No related physical data are actually presented in this paper; only the references for such material are listed. Other areas so covered include adsorption phenomena, purification processes, solubility relationships, density and compressibility data, thermodynamic and transport properties, P-V-T data, critical constants, virial coefficients, analytical techniques and various processing references. Such material for the pure components as well as for mixtures of helium and of hydrogen is included in many cases.

The P-T region explored by Smith (247) is shown in Figure 1. (The data needed to prepare the P-T diagrams for hydrogen and helium were taken from Johnson (134), Scott et al. (243) and Keesom (151)).

## 3. Discussion of Available Data

Admittedly, the data presented herein cover only a narrow temperature range: namely, from 17.4° to 21.8°K. However, for this system one could expect the data, at best, to vary over a narrow range. Complete data would extend roughly from the triple-point temperature of hydrogen (13.9°K) to the critical temperature (33.0°K). The difficulties of investigating this system completely are obvious.

The treatment given the original data of Smith (247) consisted of making appropriate calculations to arrive at K-factors for each component. (In some cases it was necessary to supplement the tabulated data of Smith with P-x and P-y values from his various plots). K is defined as  $y/x$ ; where y is the mole fraction of a component in the vapor phase and x is the mole fraction of that component in the liquid phase. K-factors were calculated for each component at a given temperature and pressure. Initially a plot of K versus P (total pressure) was prepared for each component. The data did show some scatter, especially for helium K-factors. The best smooth curve was drawn through the plotted points and this curve was then transferred to another plot (Figure 4).

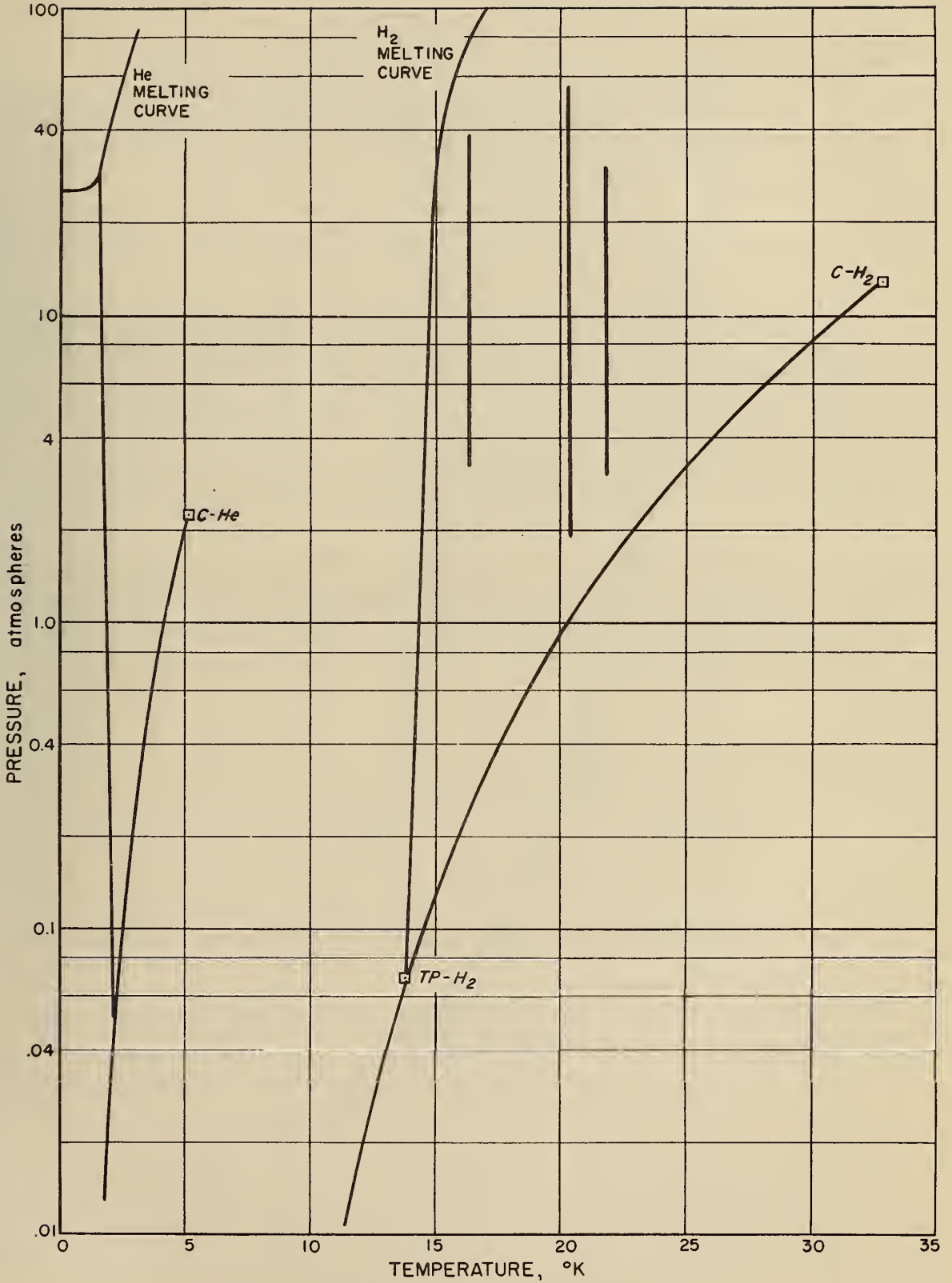


Figure 1. Regions Covered by Published Data

It should be noted that the 20.4° and 21.8°K isotherms for helium cross at approximately 10 atmospheres. Since the Boyle point of helium is near 20°K, one would expect a reversal of the gas solubility in that region. This is in fact shown by the data.

It is not the purpose of this article to make a test of the data for thermodynamic consistency. Smith does treat his data from several thermodynamic viewpoints.

#### 4. K-Factor Charts

Figures 2 and 3 are plots of the K-factors for helium and for hydrogen, respectively. Figure 4 shows the curves for both helium and hydrogen as taken from Figures 2 and 3. In Figure 4, helium K-factors are above the line  $K = 1$  and those for hydrogen are below. If the data extended to higher pressures, the isotherm could be expected to close at the high-pressure end. Such closure would occur at the line  $K = 1$ . This line thus represents the locus of the plait points.

Figure 4 permits the calculation of the vapor and liquid phase equilibrium composition at a given system temperature and pressure. After obtaining the individual K-factors, one simply substitutes into the following series of equations:

$$K_1 = y_1/x_1 \quad (1)$$

$$K_2 = y_2/x_2 \quad (2)$$

$$x_1 + x_2 = 1.0 \quad (3)$$

$$y_1 + y_2 = 1.0 \quad (4)$$

The subscripts refer to the components, helium and hydrogen. As an example, at  $T = 20.4^\circ$  and  $P = 20$  atm.

$$K_{\text{He}} = 37$$

$$K_{\text{H}_2} = 0.094$$

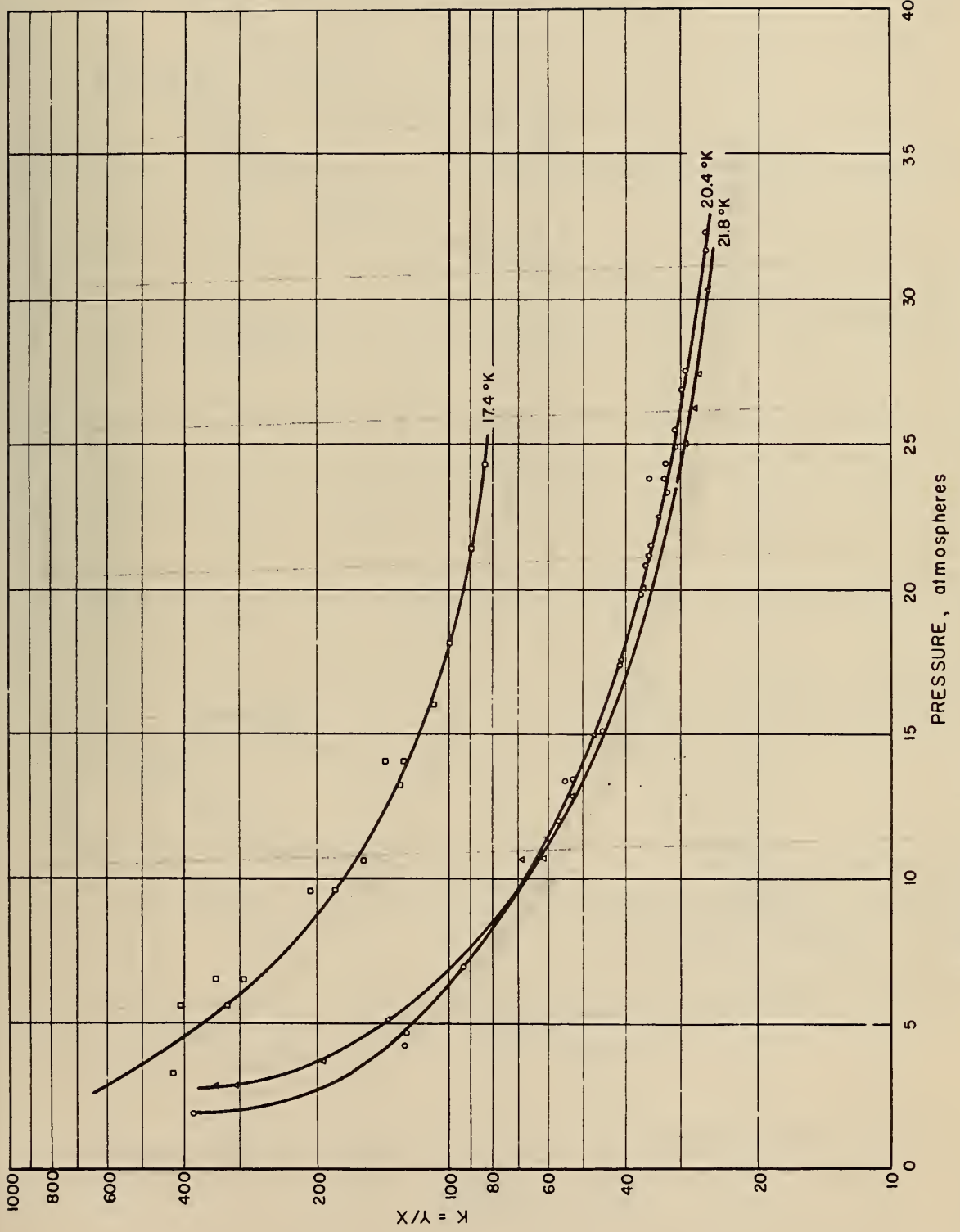


Figure 2. Helium-Hydrogen Vapor-Liquid Equilibria  
Data: Helium K-Factors

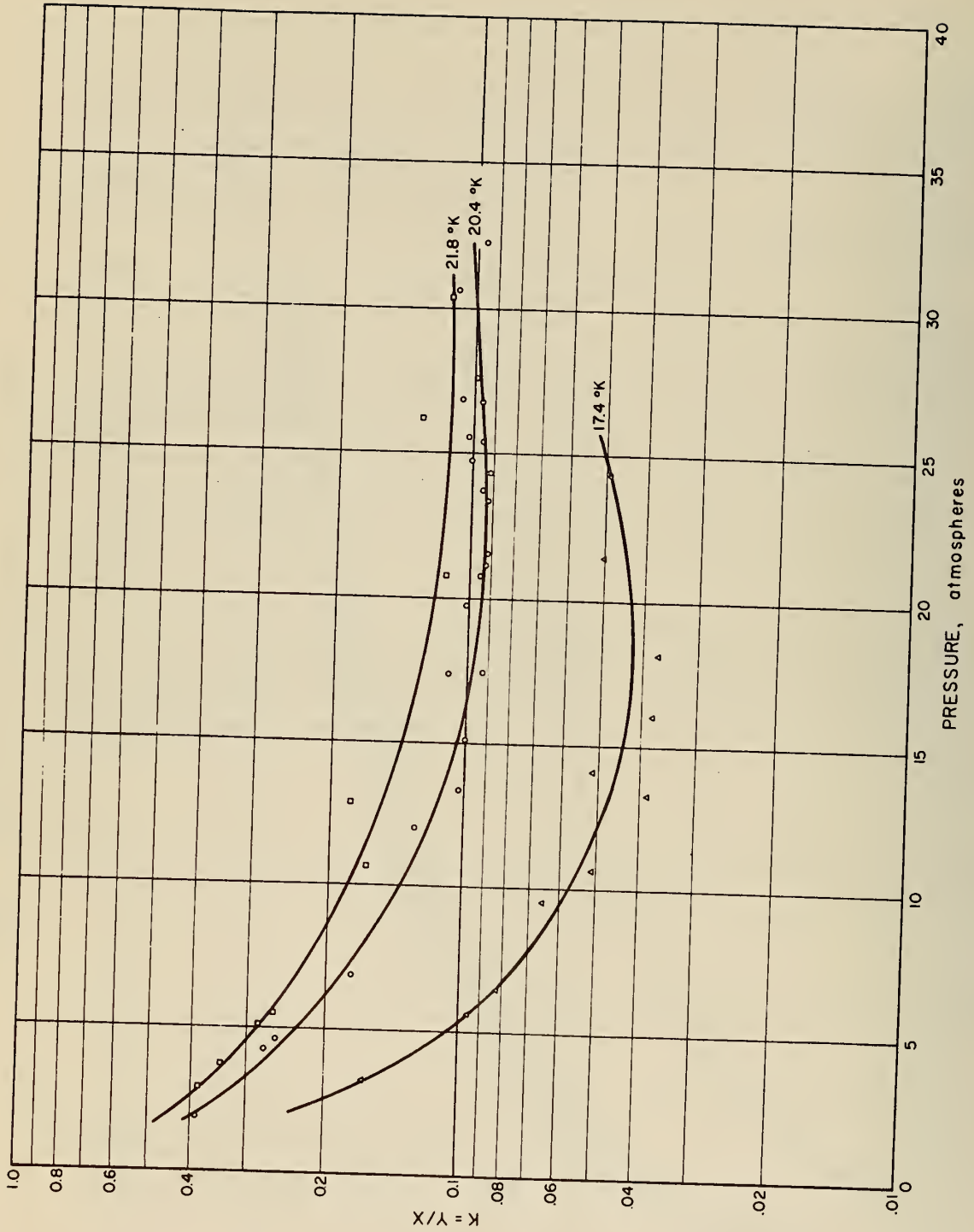


Figure 3. Helium-Hydrogen Vapor-Liquid Equilibria  
Data: Hydrogen K-Factors



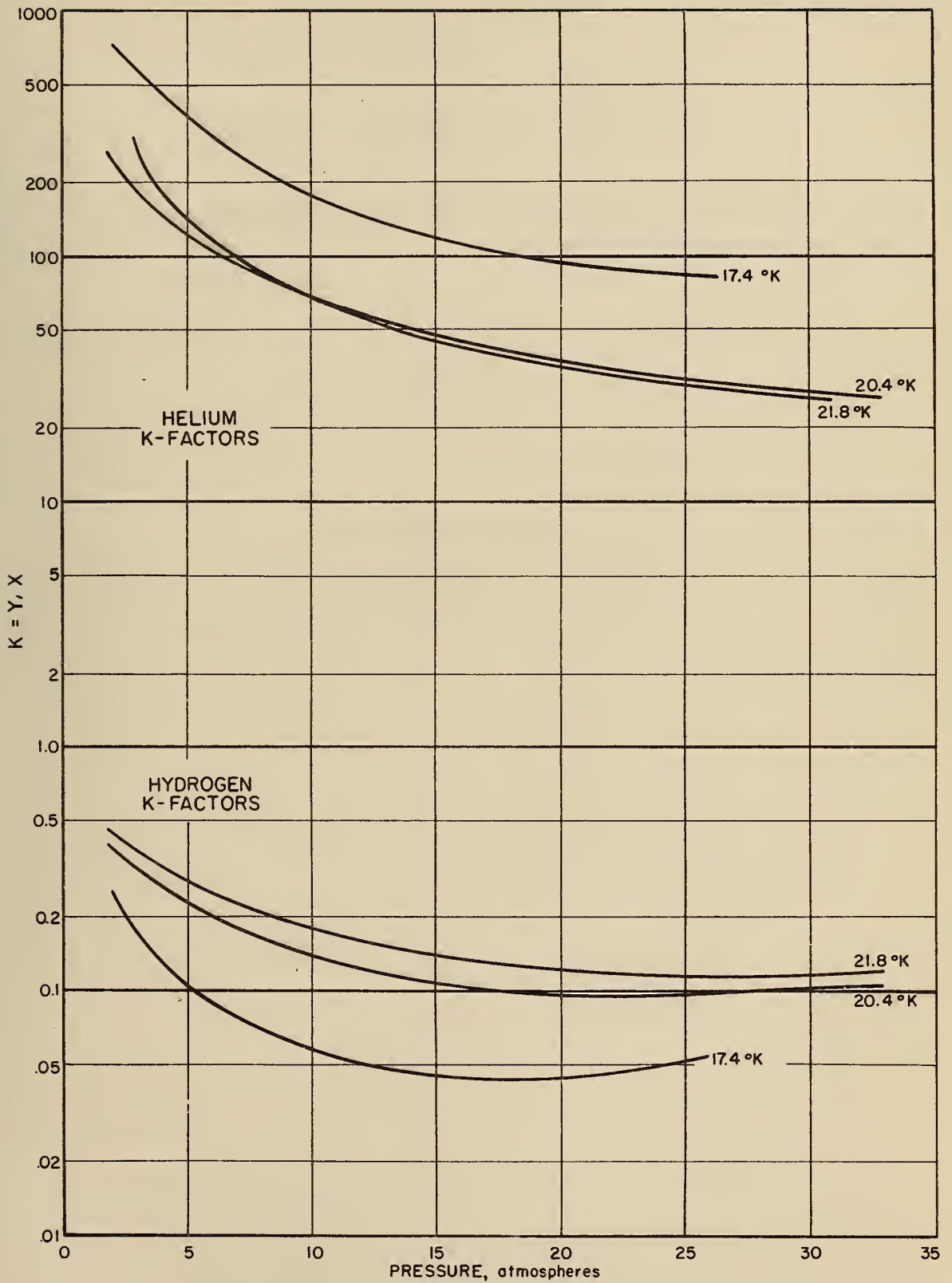


Figure 4. Vapor-Liquid Equilibria Helium-Hydrogen System

Solving equations (1) - (4) gives

$$x_{\text{He}} = 0.0245; \quad x_{\text{H}_2} = 0.9755$$

$$y_{\text{He}} = 0.9083; \quad y_{\text{H}_2} = 0.0917$$

Other relationships involving K-factors, e.g. relative volatilities, could also be easily calculated.

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Phenomena Index



Phenomena

## MAJOR COMPONENT

## HYDROGEN

Category	Other Components	References
Adsorption		9, 12, 59, 62, 75, 86, 126, 130, 133, 158, 159, 165, 166, 179, 180, 181, 188, 204, 218, 220, 221, 226, 249, 255, 257
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	Nitrogen	135, 257
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	Deuterium	170
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Phenomena

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## Properties

## MAJOR COMPONENT

## HELIUM

Category	Other Components	References
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## Properties (Continued)

## MAJOR COMPONENT

## HELIUM

Category	Other Components	References
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## Processes

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HYDROGEN

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## Processes

## MAJOR COMPONENT

## HELIUM

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Handling		30, 182, 214
Heat Transfer		81, 229
	Hydrogen	267
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Appendix





TABLE I

 HELIUM-HYDROGEN  
 PRESSURE-CONCENTRATION DATA

Reference: Smith (247)

Temperature °K	Pressure atm	Liquid		Vapor		K	
		Mole % He	Mole % H <sub>2</sub>	Mole % He	Mole % H <sub>2</sub>	He	H <sub>2</sub>
17.4	3.27	0.19	99.81	83.40	16.60	439	0.166
17.4	3.27	0.20	99.80	83.50	16.50	418	0.165
17.4	5.65	0.22	99.78	90.10	9.90	410	0.0992
17.4	5.65	0.28	99.72	90.80	9.20	324	0.0923
17.4	6.53	0.27	99.73	91.76	8.24	340	0.0826
17.4	9.59	0.45	99.55	93.47	6.53	208	0.0656
17.4	9.59	0.51	99.49	93.41	6.59	183	0.0662
17.4	14.08	0.67	99.33	94.31	5.69	141	0.0573
17.4	14.08	0.74	99.26	94.44	5.56	128	0.0560
17.4	21.50	1.05	98.95	95.04	4.96	90.5	0.0501
17.4	21.50	1.08	98.92	95.02	4.98	88.0	0.0503
17.4	24.35	1.11	98.89	95.16	4.84	85.7	0.0489
17.4	24.35	1.15	98.85	95.21	4.79	82.8	0.0485
20.4	4.29	0.59	99.41	72.90	27.10	124	0.273
20.4	6.94	0.87	99.13	82.50	17.50	94.8	0.177
20.4	12.04	1.53	98.47	87.20	12.80	56.9	0.130
20.4	17.41	2.16	97.84	89.00	11.00	41.2	0.112
20.4	19.80	2.47	97.53	90.16	9.84	36.5	0.101
20.4	23.81	2.77	97.23	90.70	9.30	32.7	0.0956
20.4	24.83	2.95	97.05	90.20	9.80	30.6	0.101
20.4	24.83	2.97	97.03	90.20	9.80	30.4	0.101
20.4	25.51	2.89	97.11	90.70	9.30	31.4	0.0958
20.4	27.62	3.06	96.94	90.40	9.60	29.5	0.0990
21.8	2.93	0.18	99.82	61.3 *	38.7 *	341	0.388
21.8	3.74	0.34*	99.66*	65.8	34.2	194	0.343
21.8	5.17	0.52*	99.48*	71.5	28.5	138	0.286
21.8	5.58	0.29	99.71	74.0 *	26.0 *	255	0.261
21.8	10.68	1.21	98.79	83.6	16.4	69.1	0.166
21.8	10.68	1.36	98.64	83.7	16.3	61.5	0.165
21.8	12.86	1.55*	98.45*	82.2	17.8	53.0	0.181
21.8	20.82	2.44	97.56	88.9 *	11.1 *	36.4	0.114
21.8	26.26	3.06	96.94	87.4	12.6	28.6	0.130
21.8	26.26	3.13	96.87	87.2	12.8	27.9	0.132
21.8	30.41	3.38*	96.62*	88.9	11.1	26.3	0.115

NOTE: \* Values taken from Smith's Figures





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**Electricity.** Resistance and Reactance. Electrochemistry. Electrical Instruments. Magnetic Measurements. Dielectrics.

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**Analytical and Inorganic Chemistry.** Pure Substances. Spectrochemistry. Solution Chemistry. Standard Reference Materials. Applied Analytical Research.

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**Organic and Fibrous Materials.** Rubber. Textiles. Paper. Leather. Testing and Specifications. Polymer Structure. Plastics. Dental Research.

**Metallurgy.** Thermal Metallurgy. Chemical Metallurgy. Mechanical Metallurgy. Corrosion. Metal Physics. Electrolysis and Metal Deposition.

**Mineral Products.** Engineering Ceramics. Glass. Refractories. Enameled Metals. Crystal Growth. Physical Properties. Constitution and Microstructure.

**Building Research.** Structural Engineering. Fire Research. Mechanical Systems. Organic Building Materials. Codes and Safety Standards. Heat Transfer. Inorganic Building Materials.

**Applied Mathematics.** Numerical Analysis. Computation. Statistical Engineering. Mathematical Physics. Operations Research.

**Data Processing Systems.** Components and Techniques. Digital Circuitry. Digital Systems. Analog Systems. Applications Engineering.

**Atomic Physics.** Spectroscopy. Infrared Spectroscopy. Solid State Physics. Electron Physics. Atomic Physics.

**Instrumentation.** Engineering Electronics. Electron Devices. Electronic Instrumentation. Mechanical Instruments. Basic Instrumentation.

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**Radio Standards.** High Frequency Electrical Standards. Radio Broadcast Service. Radio and Microwave Materials. Atomic Frequency and Time Interval Standards. Electronic Calibration Center. Millimeter-Wave Research. Microwave Circuit Standards.

**Radio Systems.** High Frequency and Very High Frequency Research. Modulation Research. Antenna Research. Navigation Systems.

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