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

BY
D. E. DRAYER AND T. M. FLYNN

U. S. DEPARTMENT OF COMMERCE
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NATIONAL BUREAU OF STANDARDS

Technical Note

May 1961

A Compilation of the Physical Equilibria

and

Related Properties of the Hydrogen-Carbon Monoxide System

by

Dennis E. Drayer

and

Thomas M. Flynn

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Abstract

Literature data have been used to calculate K-factors for the hydrogen-carbon monoxide system over the range of 68.2 to 122.2°K and 10 to 225 atmospheres. K-factors are presented graphically for eight isotherms over this range.

Published data on the solid-vapor region are presented separately as composition versus pressure at constant temperature.

A bibliography of approximately 450 references is also presented on related properties for this system and for the pure components.

1. Introduction

1.1 Purpose

Hydrogen gas for liquefaction purposes frequently contains large amounts of other gases. Typical impurities or contaminants found in hydrogen-source streams include nitrogen, carbon monoxide, methane, ethane, and higher hydrocarbons. Purification of these various sources of hydrogen requires a considerable knowledge of the physical equilibria of the systems involved.

As an initial step in the study of the physical equilibria of hydrogen systems, a review of the literature was undertaken to determine what has been done and is known in this area. In this paper, the carbon monoxide-hydrogen system was so studied. (Previously, the nitrogen-hydrogen system was examined). The purpose of this paper is to determine what is known about the physical equilibria relationships and to present an extensive compilation of known related references for data for this system. It is hoped that this paper will thus provide a firm basis for the conduct of research programs in this area.

Future publications in this series will be concerned with the physical equilibria of hydrogen and other important components. Binary systems will be studied initially. When such a program is completed, it is hoped that the results will be of significant value in the design of cryogenic equipment.

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 such data are to be found. Part (3), the Bibliography, lists the references alphabetically by author.

1.3 Scope

The scope of this work is as follows: an exhaustive literature search, as summarized in NBS Technical Note No. 56, revealed much of the pertinent data; such data were abstracted and presented in the form of K-factor charts and a concentration-pressure chart 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 and prior to July of 1960.

1.4 Acknowledgements

The authors especially appreciate the aid of the staff of the Data Center of the National Bureau of Standards Cryogenic Engineering Laboratory who provided the majority of the original papers for review.

2. Survey of Literature

The literature search revealed three important references for carbon monoxide-hydrogen vapor-liquid equilibria data. These were Akers and Eubanks (4), Ruhemann and Zinn (368), and Verschoyle (423). K-factor charts were prepared from the data of these references. In all cases the data had to be re-interpreted to arrive at K-factors.

Of noteworthy interest is the article by Verschoyle (423) who also presents solid-vapor and solid-liquid-vapor data for this same system. Also, the extensive work of Dokoupil et al. (88) presents solid-gas equilibria data for this system.

No related physical data are actually presented in this report; only references for such material are listed. Other areas so covered include adsorption phenomena, purification processes, solubility relationships, density and compressibility data, equations of state, thermodynamic and transport properties, P-V-T data, critical constants, virial coefficients, Beattie-Bridgeman constants, analytical techniques, and various processing references. Such material for the pure components as well as for mixtures of carbon monoxide and hydrogen is included in many cases. A general phenomena category is also presented to aid in the theoretical study of adsorption, phase equilibria, purification, solubility relationships, and other important

areas.

3. Discussion of Available Data

For this system one could expect the vapor-liquid data, if complete, to range roughly from the triple point temperature of carbon monoxide to the critical temperature of carbon monoxide (68.1 to 132.9°K). The three articles mentioned previously provided data for eight isotherms between 68.2°K and 122.2°K. The isotherms so presented are at temperatures of 68.2°, 73.2°, 78°, 83°-83.3°, 88.2°, 90°, 100°, and 122.2°K. (The vapor-liquid equilibria data for 83°K and for 83.3°K are plotted as one isotherm and so labeled 83°-83.3°K). Thus, the data available do present a rather complete picture of the vapor-liquid equilibria for this system.

The solid-vapor region has been explored at 58.2° and 63.2°K and from 20 to 175 atmospheres by Verschoyle (423) and from 32° to 70°K and 1.3 to 50 atmospheres by Dokoupil et al. (88).

The P-T regions covered by the published data are presented in Figure 1. This figure indicates that this system has been rather well explored in both the vapor-liquid region and the solid-vapor region. (The P-T data of hydrogen and carbon monoxide needed for the construction of Figure 1 were obtained from Johnson (189), Verschoyle (423) and Woolley, Scott and Brickwedde (446)).

The original data were treated to arrive at the corresponding K-factors. 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. After plotting the K-factors derived from the various investigations, a smooth curve was drawn for the given isotherm. Finally, the smoothed, individual K-factors were transferred to a plot of K versus total pressure with temperature as a parameter.

It is not the purpose of this report to present a test of the data for thermodynamic consistency. However, some general comments regarding the agreement between investigators is in order. Most discrepancies appear to lie in the pressure range from 10 to 30 atmospheres. In this range, there is some inconsistency in the hydrogen data as evidenced by cross-over of the isotherms of different investigators. The carbon monoxide data were not subject to these

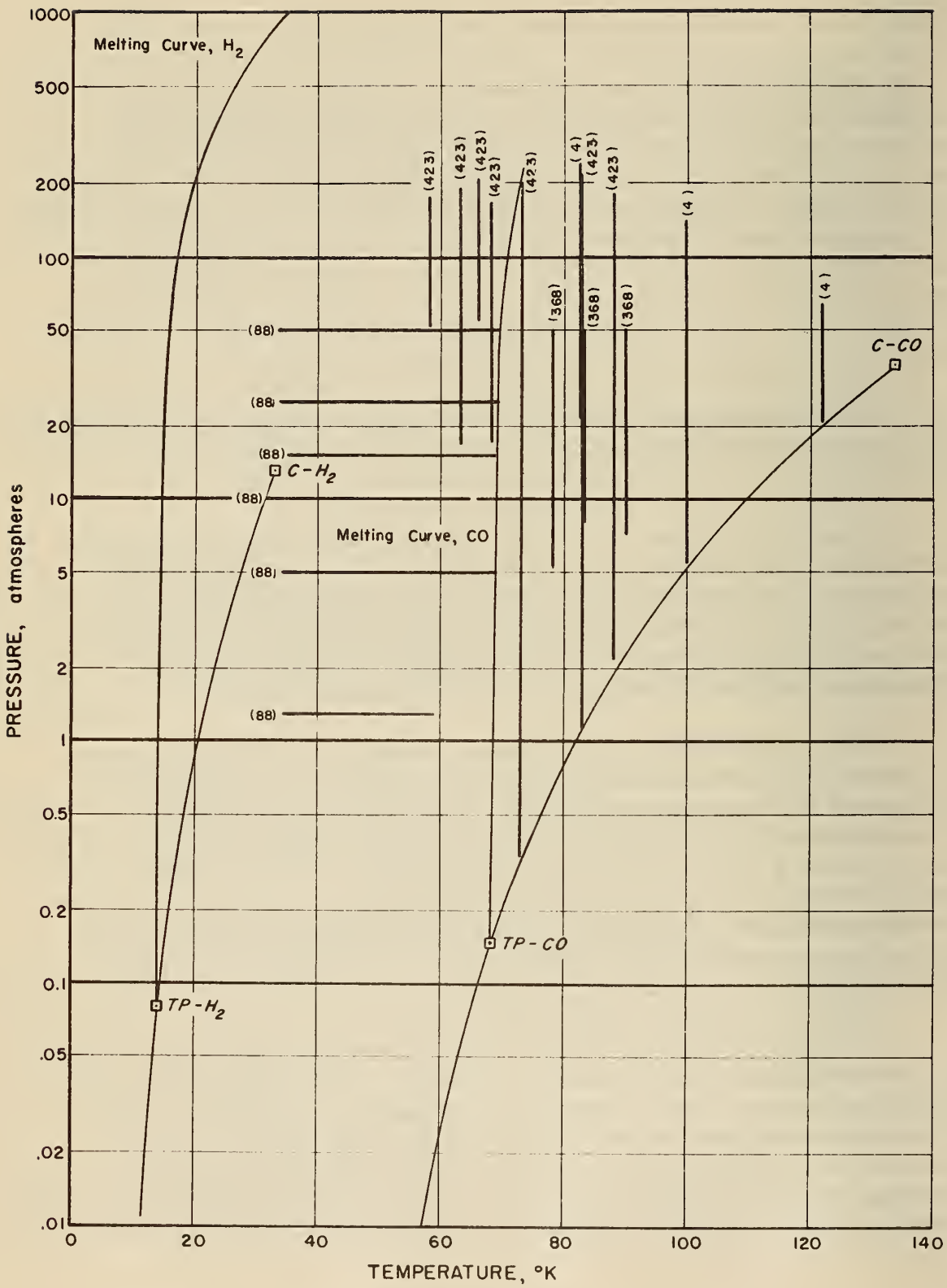

Figure 1. Regions Covered by Published Data. Parenthetical Numbers Refer to Sources in Bibliography.

variations. This scatter of data for hydrogen may stem partly from the analytical techniques used.

The 78° and 90°K isotherms, as contributed by Ruhemann and Zinn, extend only to 50 atmospheres. By using the adjacent isotherms as guide lines, one could, if required, probably make a reasonable extrapolation of these two isotherms to higher pressures.

It must be emphasized that this report is based on the original data of the investigators. These data, in most cases, have not been tested for thermodynamic consistency and should be used only with thorough awareness of this fact.

4. K-Factor Charts

Presented in Tables I, III and IV in the Appendix are the data used in computing K-factors.

Figures 2 and 3 are plots of the K-factors of hydrogen and carbon monoxide, respectively. Dotted portions on these figures indicate extrapolated areas. Figure 4 shows, finally, the curves for both hydrogen and carbon monoxide as taken from Figures 2 and 3. In Figure 4, hydrogen K-factors are situated above the line K = 1 and carbon monoxide K-factors are below this line. The intersection of an isotherm with the line K = 1 is called the plait point for that isotherm. The plait points for the 68.2° and 73.2°K isotherms were estimated by Verschoyle to be at pressures of 380 and 325 atmospheres, respectively. Critical constants for this system have been estimated by Verschoyle (423) and are presented in Table II.

Figure 4 thus contains sufficient information to enable one to calculate the vapor and liquid compositions under given temperature and pressure conditions. After the K-factors are obtained, one simply substitutes into the following formulae to obtain phase compositions:

$$K_1 = y_1/x_1 \tag{1}$$

$$K_2 = y_2/x_2 \tag{2}$$

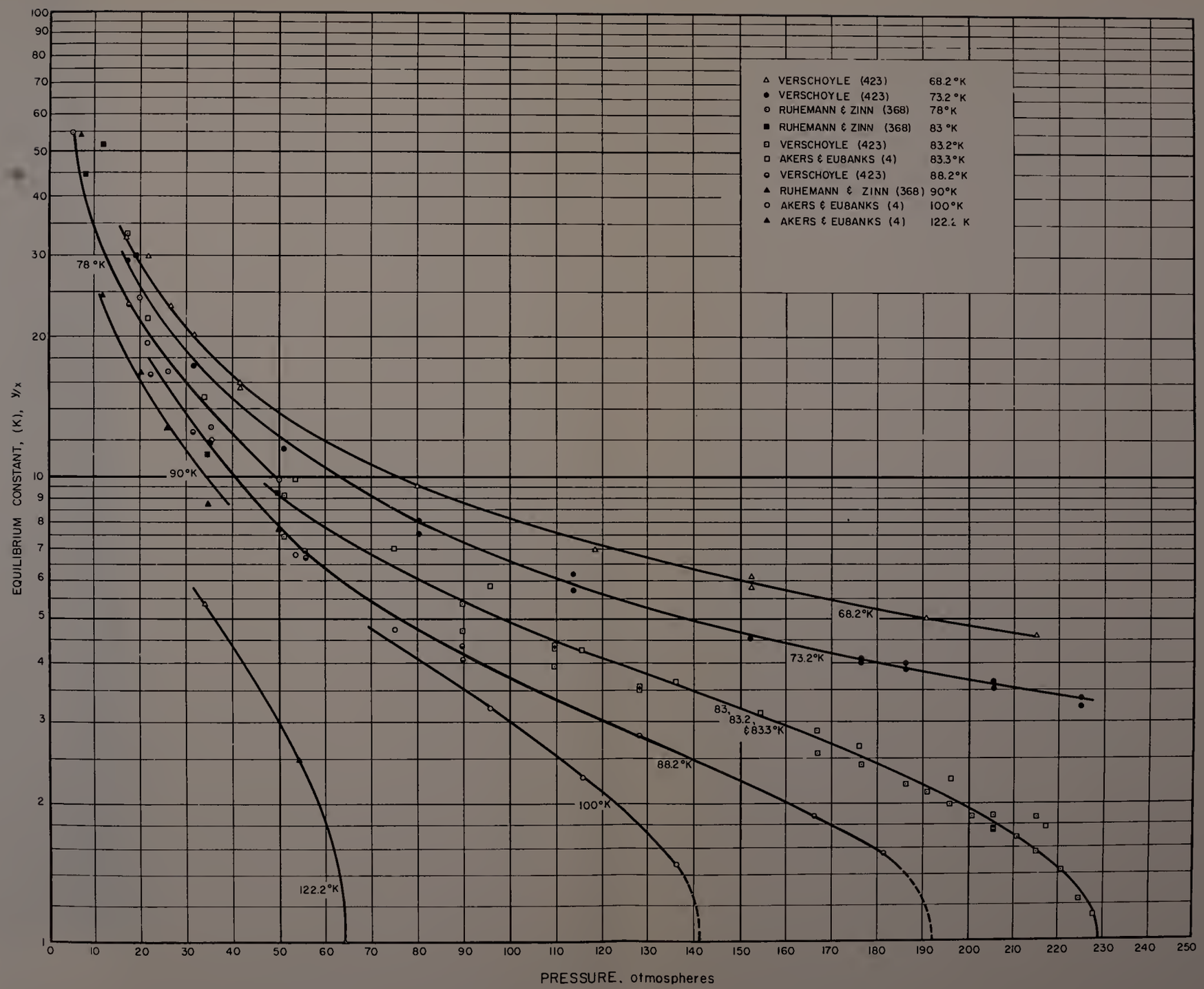

Figure 2. Carbon Monoxide-Hydrogen Vapor-Liquid Equilibria Data. Hydrogen K-Factors. Parenthetical Numbers Refer to Sources in Bibliography.

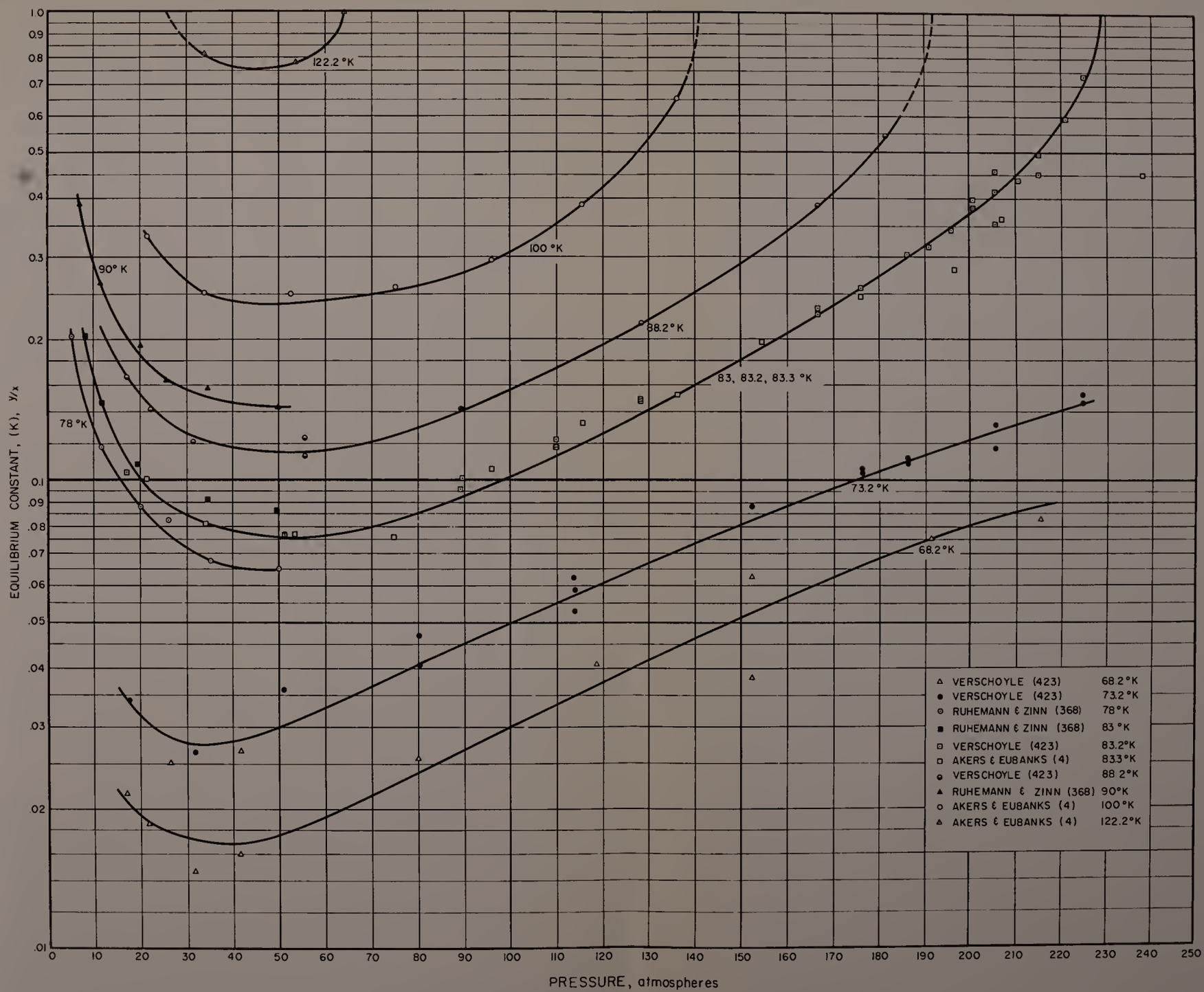

Figure 3. Carbon Monoxide-Hydrogen Vapor-Liquid Equilibria Data. Carbon Monoxide K-Factors. Parenthetical Numbers Refer to Sources in Bibliography.

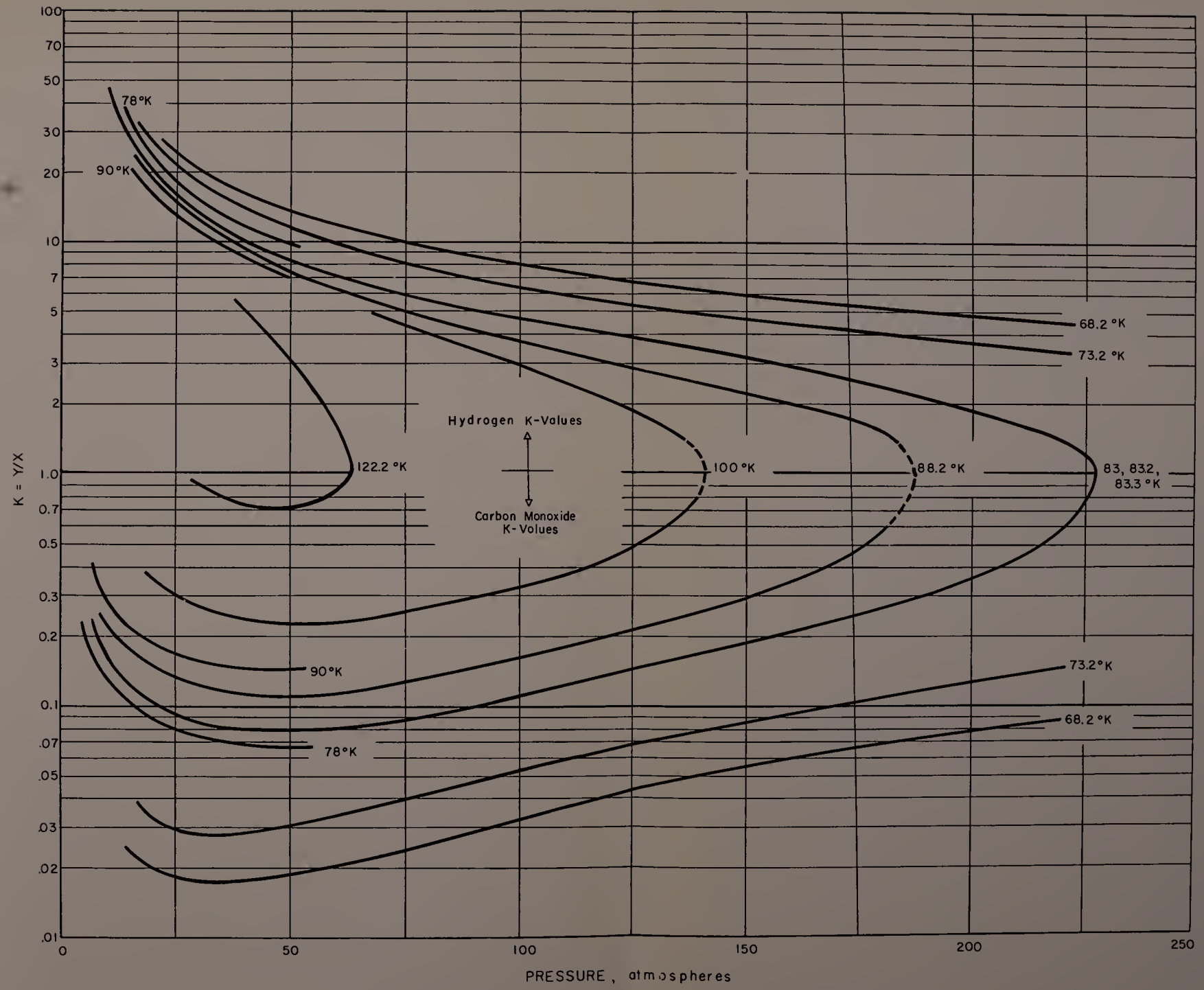

Figure 4. Vapor-Liquid Equilibria. Carbon Monoxide-Hydrogen System.

$$y_1 + y_2 = 1.0 \quad (3)$$

$$x_1 + x_2 = 1.0 \quad (4)$$

The subscripts refer to hydrogen and carbon monoxide. For example, at a system pressure of 100 atm., the phase compositions at 88.2°K could be found as follows:

$$K_{CO} = 0.159 = y_{CO}/x_{CO} \quad ; \quad y_{CO} = 0.159\, x_{CO}$$

$$K_{H_2} = 3.72 = y_{H_2}/x_{H_2} \quad ; \quad y_{H_2} = 3.73\, x_{H_2}$$

Solving equations (3) and (4), one obtains

$$y_{CO} = 0.122 \quad ; \quad x_{CO} = 0.764$$

$$y_{H_2} = 0.878 \quad ; \quad x_{H_2} = 0.236$$

Similarily, dew points and bubble points of given hydrogen-carbon monoxide mixtures can be calculated.

5. Solid-Vapor Equilibria

The data of Verschoyle (423) are given in Table V. These data have been plotted in Figure 5 to show vapor phase composition versus total pressure for the isotherms of 58.2° and 63.2°K.

The extensive solid-gas equilibria data of Dokoupil et al. (88) are given in Table VI. These data have been replotted (not shown) to arrive at P-y curves at 5°K temperature intervals from 35° to 65°K. The derived data are shown in Table VII and also on Figure 5. Of noteworthy interest is the minimum shown by each isotherm. The locus of these minimum points in the y-P curves thus allow the selection of the optimum total pressure at a given temperature to yield a minimum CO concentration in the gas phase.

6. Three-Phase Equilibria

Verschoyle has also presented data showing the locus of the three-phase curve. Table VIII contains this information.

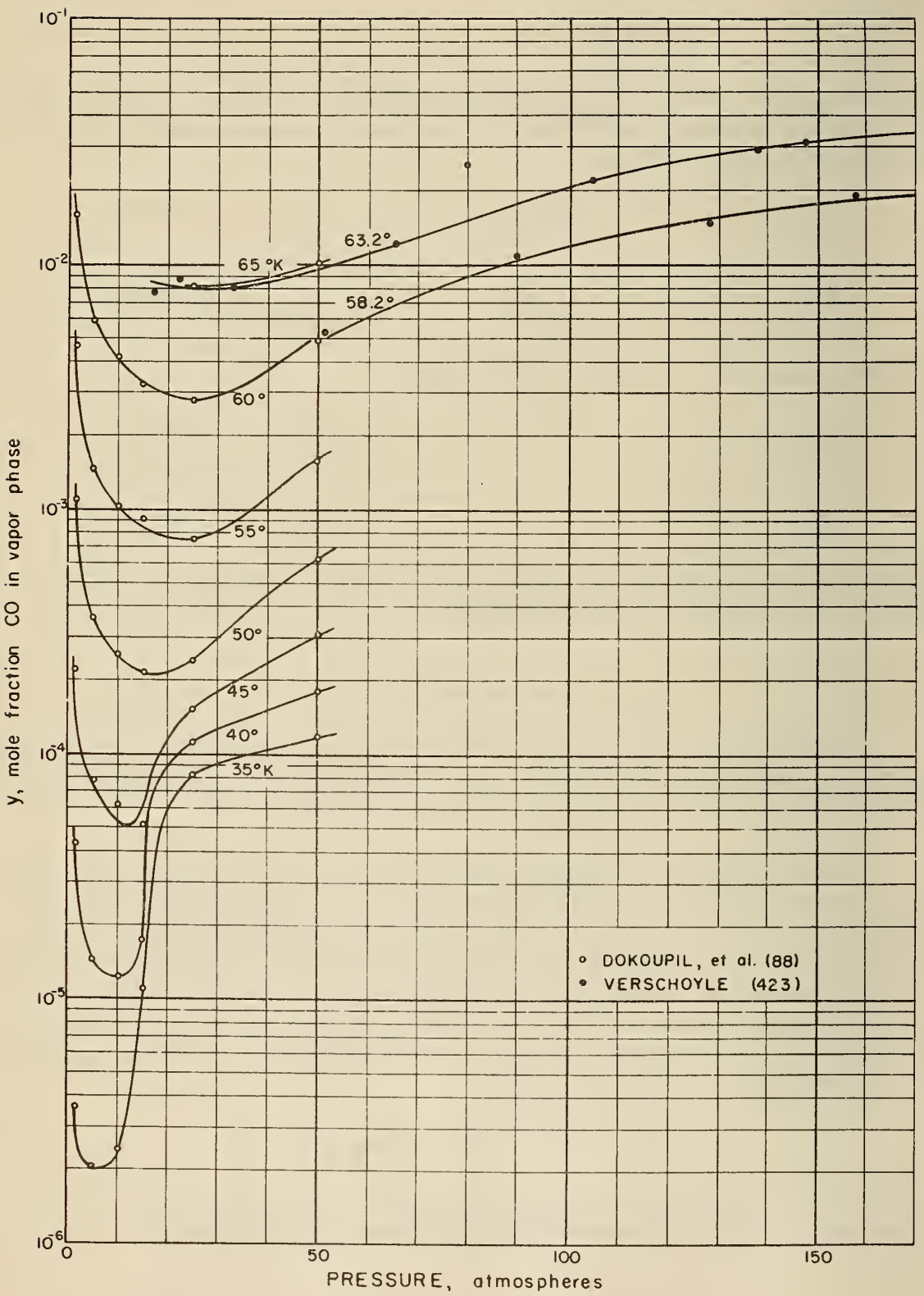

Figure 5. Solid-Vapor Equilibria. Concentration of Carbon Monoxide in the Vapor Phase.

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Phenomena Index

Phenomena

MAJOR COMPONENT

HYDROGEN

Category	Other Components	References
Adsorption		8, 11, 84, 87, 107, 119, 176, 183, 187, 208, 209, 211, 212, 249, 250, 266, 296, 318, 332, 333, 351, 395, 414, 419
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	Nitrogen	188, 419
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Phenomena

MAJOR COMPONENT

GENERAL

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Properties

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Processes

MAJOR COMPONENT

HYDROGEN

Category	Other Components	References
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	Carbon Monoxide-Nitrogen	109
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	Nitrogen-Carbon Monoxide-Methane	185
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Processes

MAJOR COMPONENT

CARBON MONOXIDE

Category	Other Components	References
Analytical		34, 35, 131, 203, 307, 322, 426, 427, 430
	Carbon Dioxide-Oxygen-Hydrogen	110
	Ethylene	325
	Hydrogen	317, 380
	Hydrogen-Methane	272
	Hydrogen-Nitrogen	109
	Hydrogen-Nitrogen Methane	185

Processes

MAJOR COMPONENT

GENERAL

Category	Other Components	References
Analytical		38, 69, 142, 196, 286, 384, 403, 404, 425

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Appendix

TABLE I

CO-H_2 PRESSURE-CONCENTRATION DATA

Reference: Verschoyle (423)

Temperature		Pressure	Liquid		Vapor		K	
°C	°K	Atm	Mole % H_2	Mole % CO	Mole % H_2	Mole % CO	H_2	N_2
-183	88.2	181.3	45.4	54.6	70.4	29.6	1.55	0.542
"	"	166.7	41.0	59.0	77.1	22.9	1.88	0.388
"	"	128.2	30.3	69.7	84.8	15.2	2.80	0.218
"	"	89.6	21.7	78.3	88.7	11.3	4.09	0.144
"	"	89.3	20.3	79.7	88.8	11.2	4.37	0.141
"	"	55.9	13.4	86.6	90.2	9.8	6.73	0.113
"	"	55.8	12.9	87.1	89.3	10.7	6.92	0.123
"	"	31.4	7.1	92.9	88.8	11.2	12.5	0.121
"	"	22.1	5.2	94.8	86.6	13.4	16.6	0.141
"	"	17.2	3.6	96.4	84.0	16.0	23.3	0.166
-190	83.2	224.8	54.1	45.9	66.3	33.7	1.23	0.734
"	"	220.9	48.6	51.4	69.4	30.6	1.43	0.595
"	"	215.1	47.0	53.0	73.7	26.3	1.57	0.496
"	"	210.4	44.8	55.2	75.9	24.1	1.69	0.437
"	"	205.4	43.5	56.5	76.6	23.4	1.76	0.414
"	"	200.6	41.1	58.9	76.7	23.3	1.87	0.396
"	"	200.6	41.5	58.5	77.7	22.3	1.87	0.381
"	"	195.8	40.1	59.9	79.5	20.5	1.98	0.342
"	"	190.9	38.1	61.9	80.5	19.5	2.11	0.315
"	"	186.1	36.8	63.2	80.8	19.2	2.20	0.304
"	"	176.4	34.4	65.6	83.1	16.9	2.42	0.258
"	"	166.8	32.9	67.1	84.3	15.7	2.56	0.234
"	"	128.1	24.9	75.1	88.9	11.1	3.57	0.148
"	"	128.1	25.4	74.6	88.8	11.2	3.50	0.150
"	"	109.8	22.8	77.2	90.4	9.6	3.96	0.124
"	"	109.8	21.0	79.0	90.7	9.3	4.32	0.118
"	"	109.6	20.6	79.4	90.6	9.4	4.40	0.118
"	"	89.5	19.5	80.5	91.8	8.2	4.71	0.102
"	"	89.3	17.0	83.0	92.0	8.0	5.41	0.0964
"	"	51.2	12.5	87.5	93.2	6.8	7.46	0.0777
"	"	51.2	10.2	89.8	93.1	6.9	9.13	0.0768
"	"	17.2	2.7	97.3	89.9	10.1	33.3	0.104
-200	73.2	224.9	27.5	72.5	89.0	11.0	3.24	0.152
"	"	224.8	26.5	73.5	89.3	10.7	3.37	0.146
"	"	205.6	25.0	75.0	91.2	8.8	3.65	0.117
"	"	205.4	25.7	74.3	90.2	9.8	3.51	0.132
"	"	186.2	22.9	77.1	91.6	8.4	4.00	0.109
"	"	186.2	23.6	76.4	91.4	8.6	3.87	0.113
"	"	176.4	23.0	77.0	92.0	8.0	4.00	0.104
"	"	176.4	21.8	78.2	91.7	8.3	4.21	0.106
"	"	152.4	20.6	79.4	93.0	7.0	4.51	0.0882
"	"	113.9	16.6	83.4	95.1	4.9	5.73	0.0588
"	"	113.8	15.4	84.6	95.5	4.5	6.20	0.0532
"	"	113.7	16.5	83.5	94.8	5.2	5.75	0.0623
"	"	80.2	12.7	87.3	95.9	4.1	7.55	0.0470
"	"	80.1	12.0	88.0	96.4	3.6	8.03	0.0409
"	"	51.0	8.4	91.6	96.7	3.3	11.5	0.0360
"	"	31.8	5.6	94.4	97.5	2.5	17.4	0.0265
"	"	17.3	3.3	96.7	96.7	3.3	29.3	0.0341
-205	68.2	215.2	20.2	79.8	93.4	6.6	4.62	0.0827
"	"	200.0	18.8	81.2	93.9	6.1	4.99	0.0751
"	"	152.3	16.3	83.7	94.6	5.4	5.80	0.0645
"	"	118.4	13.8	86.2	96.5	3.5	6.99	0.0406
"	"	79.9	10.2	89.8	97.7	2.3	9.58	0.0256
"	"	41.4	6.3	93.7	97.5	2.5	15.5	0.0267
"	"	41.4	6.2	93.8	98.5	1.5	15.9	0.0160
"	"	31.7	4.9	95.1	98.6	1.4	20.1	0.0147
"	"	26.6	4.2	95.8	97.6	2.4	23.2	0.0251
"	"	21.7	3.3	96.7	98.2	1.8	29.8	0.0186
"	"	17.0	3.0	97.0	97.9	2.1	32.6	0.0216

TABLE II

CO-H_2 CRITICAL CONSTANTS

Reference: Verschoyle (423)

		Plait-Point		Critical Point of Contact	
Temperature		Pressure	Mole % H_2	Pressure	Mole % H_2
°C	°K	Atm		Atm	
-185	88.2	187	58	54	90
-190	83.2	228	60	48	93
-200	73.2	(325)	(64)	34	97.5
-205	68.2	(380)	(66)	30	99.5

TABLE III

CO-H_2 PRESSURE-CONCENTRATION DATA

Reference: Ruhemann and Zinn (368)

Temperature °K	Pressure Atm	Liquid		Vapor		K	
		Mole % H_2	Mole % CO	Mole % H_2	Mole % CO	H_2	N_2
90	50	11.2	88.8	87.2	12.8	7.79	0.144
"	35	9.5	90.5	85.9	14.1	9.04	0.156
"	25.8	6.5	93.5	84.5	15.5	13.0	0.166
"	20	4.6	95.4	81.9	18.1	17.8	0.190
"	11.7	2.9	97.1	74.5	25.5	25.7	0.263
"	6.9	1.2	98.8	61.6	38.4	51.3	0.389
83	49.7	10.3	89.7	92.1	7.9	8.94	0.0881
"	34.5	8.1	91.9	91.5	8.5	11.3	0.0925
"	19.5	3.1	96.9	89.5	10.5	28.9	0.108
"	12.0	1.6	98.4	85.4	14.6	53.4	0.148
78	50	9.3	90.7	94.1	5.9	10.1	0.0650
"	35	6.9	93.1	93.8	6.2	13.6	0.0666
"	25.8	5.0	95.0	92.1	7.9	18.4	0.0832
"	20	3.8	96.2	91.5	8.5	24.1	0.0884
"	11.7	1.9	98.1	88.1	11.9	46.4	0.121

TABLE IV

CO-H_2 PRESSURE-CONCENTRATION DATA

Reference: Akers and Eubanks (4)

Temperature		Pressure	Liquid		Vapor		K	
°F	°K	Atm	Mole % H_2	Mole % CO	Mole % H_2	Mole % CO	H_2	CO
-240	122.2	64.1	30.0	70.0	30.0	70.0	1.0	1.0
"	"	54.1	13.4	86.6	33.4	66.6	2.49	0.769
"	"	34.0	4.1	95.9	22.1	77.9	5.36	0.812
-280	100	136.1	41.2	58.8	61.1	38.9	1.48	0.662
"	"	115.5	32.2	67.8	73.5	26.5	2.28	0.391
"	"	95.7	24.1	75.9	77.5	22.5	3.22	0.296
"	"	75.0	16.5	83.5	78.2	21.8	4.74	0.261
"	"	53.8	11.4	88.6	77.8	22.2	6.82	0.251
"	"	34.0	6.5	93.5	76.5	23.5	11.8	0.251
"	"	21.4	3.5	96.5	67.4	32.6	19.3	0.338
-310	83.3	238.1	66.3	33.7	74.7	25.3	1.13	0.751
"	"	217.1	45.4	54.6	80.3	19.7	1.77	0.361
"	"	175.9	31.3	68.7	83.1	16.9	2.65	0.246
"	"	154.3	27.3	72.7	85.5	14.5	3.13	0.199
"	"	136.1	24.2	75.8	88.4	11.6	3.65	0.153
"	"	115.5	21.0	79.0	89.5	10.5	4.26	0.133
"	"	95.7	15.6	84.4	91.0	9.0	5.83	0.107
"	"	75.0	13.3	86.7	93.4	6.6	7.02	0.0761
"	"	53.7	9.4	90.6	93.0	7.0	9.91	0.0772
"	"	34.0	6.2	93.8	92.4	7.6	14.9	0.0810
"	"	21.4	4.1	95.9	90.3	9.7	21.9	0.101

TABLE V

SOLID-VAPOR EQUILIBRIA

Reference: Verschoyle (423)

Temperature		Pressure	Vapor Phase	
°C	°K	Atm	Mole % H_2	Mole % CO
-210	63.2	195.75	96.31	3.69
"	"	176.41	96.53	3.47
"	"	147.39	96.88	3.12
"	"	137.76	97.03	2.97
"	"	104.01	97.80	2.20
"	"	79.80	97.42	2.58
"	"	65.74	98.76	1.24
"	"	32.03	99.18	0.82
"	"	21.85	99.12	0.88
"	"	16.81	99.22	0.78
-215	58.2	176.37	98.18	1.82
"	"	157.03	98.08	1.92
"	"	128.05	98.50	1.50
"	"	89.52	98.90	1.10
"	"	51.37	99.47	0.53

TABLE VI

SOLID-VAPOR EQUILIBRIA

Reference: Dokoupil, Van Soest and Swenker (88)

Pressure Atm	Temperature °K	Vapor Phase Mole % CO
50	69.9	1.86
"	65.7	1.03
"	60.8	0.569
"	57.9	0.330
"	55.4	0.171
"	49.7	0.0623
"	45.6	0.0329
"	35.6	0.0124
25	67.7	1.36
"	66.5	1.038
"	62.8	0.515
"	60.0	0.284
"	56.0	0.0983
"	50.4	0.0234
"	41.7	0.0126
"	35.5	0.00850
15	60.7	0.400
"	59.4	0.266
"	55.6	0.104
"	54.6	0.0834
"	51.4	0.0328
"	46.1	0.00624
"	44.9	0.00480
"	42.5	0.00374
"	42.5	0.00358
"	42.1	0.00320
"	38.1	0.00139
"	35.0	0.00112
"	34.1	0.00102
"	34.1	0.00103
10	63.0	0.763
"	56.2	0.152
"	54.2	0.0784
"	51.4	0.0427
"	51.4	0.0430
"	45.8	0.00605
"	42.1	0.00314
"	37.0	0.000522
"	36.0	0.000356
"	33.9	0.000172
"	31.9	0.0000873
5	62.6	1.22
"	56.4	0.234
"	54.7	0.120
"	51.4	0.0549
"	46.2	0.00765
"	42.2	0.00320
"	37.6	0.000552
"	35.8	0.000362
"	33.4	0.0000925
1.3	58.8	1.25
"	55.3	0.527
"	50.5	0.103
"	47.2	0.0445
"	44.2	0.0167
"	39.8	0.00361
"	38.2	0.00145
"	36.4	0.000620
"	34.4	0.000288
"	32.3	0.000158

TABLE VII

SOLID-VAPOR EQUILIBRIA

Reference: Dokoupil, Van Soest and Swenker (88)
Table VI, This Report

Temperature, °K	Pressure, Atm.	y, Mole Fraction CO in Vapor Phase
35	1.3	0.00000355
"	5	0.00000205
"	10	0.00000240
"	15	0.0000110
"	25	0.000083
"	50	0.000119
40	1.3	0.000044
"	5	0.0000145
"	10	0.0000125
"	15	0.0000175
"	25	0.000112
"	50	0.000180
45	1.3	0.000223
"	5	0.0000780
"	10	0.0000620
"	15	0.0000510
"	25	0.000153
"	50	0.000310
50	1.3	0.00110
"	5	0.000361
"	10	0.000253
"	15	0.000212
"	25	0.000241
"	50	0.000630
55	1.3	0.00470
"	5	0.00148
"	10	0.00103
"	15	0.000910
"	25	0.000750
"	50	0.00160
60	1.3	0.0160
"	5	0.0059
"	10	0.00420
"	15	0.00325
"	25	0.00280
"	50	0.00480
65	25	0.0081
"	50	0.0100

TABLE VIII

THREE-PHASE DATA

Reference: Verschoyle (423)

Pressure	Temperature	
Atm	°C	°K
55.2	-206.12	67.04
55.2	-206.15	67.01
104.1	-206.54	66.62
104.1	-206.59	66.57
104.1	-206.73	66.43
147.4	-206.48	66.68
147.4	-206.67	66.49
147.4	-206.72	66.44
147.4	-206.87	66.29
205.5	-206.34	66.82
205.5	-206.38	66.78
205.5	-206.39	66.77

U. S. DEPARTMENT OF COMMERCE
Luther H. Hodges, *Secretary*

NATIONAL BUREAU OF STANDARDS
A. V. Astin, *Director*

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