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Thermodynamic Properties of 1-Chloro-1,2,2,2-tetrafluoroethane (R124)

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The critical temperature and pressure, vapor pressure, and *PVT* relations for gaseous and liquid 1-chloro-1,2,2,2-tetrafluoroethane (R124) were determined experimentally. The vapor pressure was measured in the temperature range from 278.15 K to the critical temperature. The *PVT* measurements were carried out using two types of volumeters in the temperature range from 278.15 to 423.15 K, at pressure up to 100 MPa. The numerical *PVT* data of gaseous state are fitted as a function of density to a modified Benedict–Webb–Rubin equation. The pressure–volume relations of the liquid at each temperature are correlated satisfactorily as a function of pressure by the Tait equation. The critical density and saturated vapor and liquid densities are also determined and some of the thermodynamic properties are derived from the experimental results.

KEY WORDS: Benedict–Webb–Rubin (BWR) equation; 1-chloro-1,2,2,2-tetrafluoroethane; compressibility factor; critical pressure; critical temperature; equation of state; R124; specific volume; vapor pressure.

1. INTRODUCTION

The chlorofluorocarbons, such as R11, R12, and R113, are employed widely as excellent foaming and spraying agents, working fluids, and cleaning solvents. In recent years, it has become clear that these compounds containing no hydrogen atoms in their molecules have long atmospheric resistance times and are decomposed by photodecomposition in the stratosphere. This process has serious environmental consequences through the release of chlorine atoms, which act as the chain catalyst of ozone removal reactions. Furthermore, another obvious problem is the enhanced atmospheric adsorption in the infrared spectral region, which

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arises from the increase of chlorofluorocarbons in the normal atmosphere. This affects the temperature of the global surface, the so-called "greenhouse effect." Therefore, it is urgently needed to find new compounds as substitutes, in order to protect the global environment, and to determine the various physical and chemical properties of the new compounds. Although the present test fluid, 1-chloro-1,2,2,2-tetrafluoroethane (R124), is not so well known, it would probably be one of the candidates among the substitutive chlorofluorocarbon compounds as a promising working fluid for refrigeration and heat pump cycles or a foaming agent for various plastic materials. In this paper, the thermodynamic properties, such as the critical temperature and pressure, vapor pressure, and densities of gaseous and liquid phases, have been measured over wide ranges of temperature and pressure. Based on the experimental data obtained, the equations of state are formulated and thermodynamic quantities are calculated.

2. EXPERIMENTAL

2.1. Sample

The sample used in the experiments was synthesized and purified specially by Daikin Industries, Ltd. Its purity is better than 99.97%, and the impurities would not affect the experimental results.

2.2. Vapor Pressure and Critical Constants

The saturated vapor pressure and the critical pressure were measured using a strain gauge-type pressure transducer which was connected to a borosilicate glass cylinder about 6.5 cm^3 in volume. The pressure gauge was calibrated against a deadweight gauge. The accuracy of the pressure measurements was within ±0.01 MPa. The sample cylinder was placed in a liquid bath thermostatically controlled to within ±10 mK. The temperature was determined with a standard mercury thermometer calibrated by the National Research Laboratory of Metrology. The uncertainty of the temperature measurements was less than 0.03 K and this would introduce an uncertainty of ±2 kPa to the vapor pressure. The total uncertainty of the vapor pressure measurements was less than 12 kPa. The temperature range of the vapor pressure measurements was from 278.15 K to the critical temperature.

2.3. *PVT* Measurements of the Superheated Vapor

A schematic diagram of the constant-volume apparatus for measuring the *PVT* relations of vapor and supercritical fluid is shown in Fig. 1. The

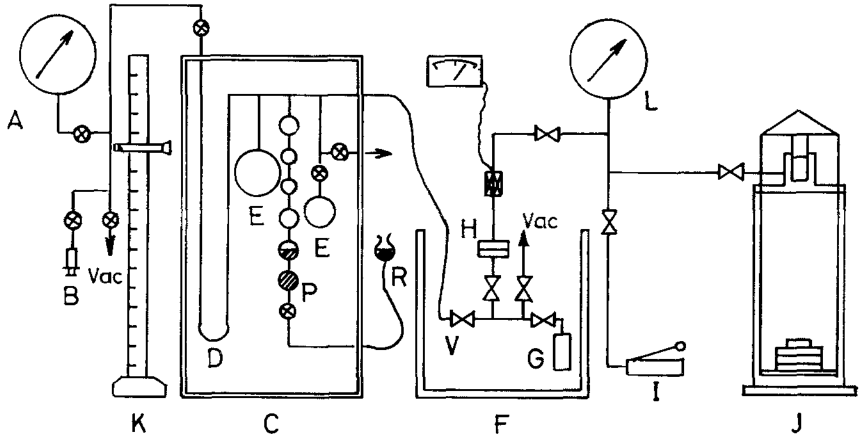

Fig. 1. Schematic diagram of the constant-volume apparatus: A, low-pressure gauge; B, air injector; C and F, constant-temperature bath; D, manometer; E, glass bulb; G, constant-volume cell; H, diaphragm-differential-pressure indicator; I, oil pump; J, deadweight gauge; K, cathetometer; L, Bourdon tube gauge; P, glass burette; R, Toepler pump; V, valve.

apparatus consisted essentially of three main sections. The first one was a pressure measurement section, which was composed of a hand-operated pump, a deadweight gauge, and a diaphragm-differential-pressure indicator. The uncertainty of the pressure measurements by this deadweight gauge was ±3.5 kPa and the sensitivity of the diaphragm-differential-pressure indicator was ±0.5 kPa. The second one was a high-pressure constant-volume section $21.81 \pm 0.02\ \mathrm{cm}^3$ in volume at 298.15 K. The temperature of this section was maintained constant within ±10 mK by a constant-temperature liquid bath (F). The third one was a low-pressure gas expansion section. The volume of this section could be varied from 200 to $900\ \mathrm{cm}^3$ by changing a glass bulb (E). The temperature of this section was maintained constant within ±50 mK using an air bath (C).

Basically, the experimental procedure involves charging a known mass of the test fluid to a cell of known volume and measuring the pressure and temperature of the test fluid.

A known mass of the sample was introduced into the high-pressure constant-volume section and the temperature of the constant-temperature bath (F) was adjusted to the experiment temperature. The temperature was measured with a standard mercury thermometer and the pressure was measured using a deadweight gauge (J). The sample gas and pressure-transmitting oil in the deadweight gauge were separated from each other by the diaphragm-differential-pressure indicator (H). By changing the experimental temperatures from 298.15 to 423.15 K, *PVT* relations at a constant mass of the sample were obtained. Then a small amount of the sample gas in the high-pressure constant-volume section was allowed to expand into the low-pressure expansion section and the pressure of this expanded gas was measured using a mercury U-tube (D). Then the low-pressure expansion section was evacuated to less than 0.2 Pa. The procedures mentioned above were repeated several times until the pressure of the sample gas remaining in the constant-volume section became about 40 kPa. The uncertainty of the isothermal compressibility factors obtained was estimated to be less than 0.35 %. The *PVT* measurements were made at temperatures from 298.15 to 423.15 K and pressures up to 5.5 MPa.

2.4. *PVT* Measurements of the Liquid

The measurements of *PVT* relations of the liquid were performed using a "high-pressure burette" apparatus, which is shown schematically in Fig. 2. The sample liquid of known weight was introduced into a vessel (A). The pressure was transmitted through mercury filled in a high-pressure burette (F) and in the lower part of A from an oil pump (I). The volume change of the test liquid was detected from the displacement of a small iron

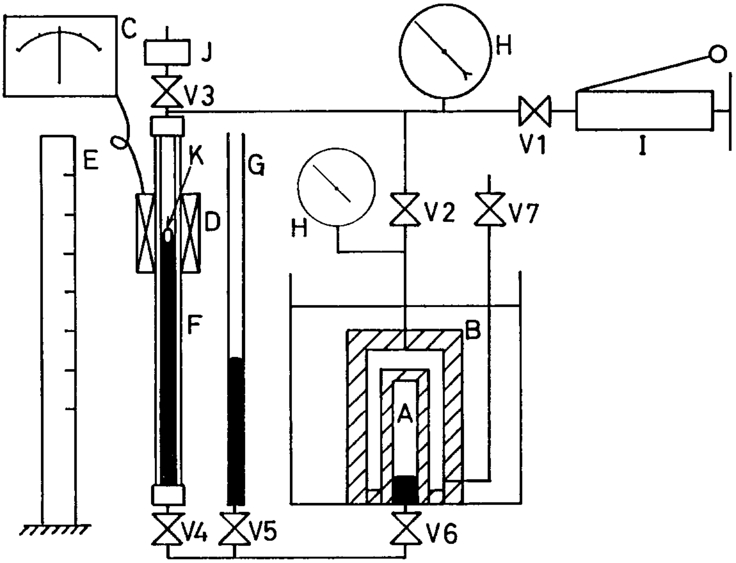

Fig. 2. Schematic diagram of the "high-pressure burette" apparatus: A and B, high-pressure vessel; C, ammeter; D, differential transformer; E, cathetometer; F, high-pressure burette; G, mercury reservoir; H, Bourdon tube gauge; I, oil pump; J, oil reservoir; K, steel float; V, valves.

float (K) on the surface of the mercury column by means of a differential transformer (D). The sample vessel consisted of coaxial double cylinders. The inner cylinder (A) was a thin-walled sample vessel about 84 cm^3 in volume. To the outside of vessel A, a pressure somewhat lower than that of the sample was applied separately from the oil pump, so as to reduce the volume change of vessel A at elevated pressures. The sample vessel was placed in a liquid bath, thermostatically controlled to within ±30 mK. The accuracy of the temperature measurements was within ±50 mK. The pressure was measured by Heise Bourdon tube gauges (H) which were calibrated against a deadweight gauge. The uncertainty in the pressure measurements was estimated to be less than 0.35 MPa.

Using the displacement of the iron float, the specific volume of the sample liquid was calculated [1]. The uncertainty of the specific volume

Table I. Vapor Pressure of R124

Temp. (K)	Obs. *P* (MPa)	Calc. *P* (MPa)
278.15	0.21	0.21
283.15	0.24	0.24
288.15	0.28	0.28
293.15	0.33	0.32
298.15	0.38	0.38
303.15	0.44	0.44
308.15	0.50	0.50
313.15	0.57	0.58
318.15	0.66	0.66
323.15	0.76	0.76
328.15	0.86	0.86
333.15	0.98	0.98
338.15	1.11	1.11
343.15	1.24	1.24
348.15	1.40	1.39
353.15	1.56	1.56
358.15	1.74	1.74
363.15	1.92	1.93
368.15	2.13	2.14
373.15	2.37	2.37
378.15	2.62	2.61
383.15	2.89	2.88
388.15	3.16	3.17
393.15	3.48	3.49
395.65	3.66	3.66

data obtained was estimated to be less than 0.09%. The *PVT* measurements were made at temperatures from 283.15 to 398.15 K and pressures up to 100 MPa.

3. RESULTS AND DISCUSSION

3.1. Critical Constants

The critical temperature and pressure were determined by observation of the meniscus in a glass cylinder which was slowly heated and cooled in a stirred liquid bath. The critical density was determined with the aid of the law of the rectilinear diameter.

$$\begin{aligned} T_c &= 395.65 \pm 0.05 \text{ K} \\ P_c &= 3.66 \pm 0.01 \text{ MPa} \\ \rho_c &= 560 \pm 2 \text{ kg} \cdot \text{m}^{-3} \end{aligned} \tag{1}$$

3.2. Vapor Pressure

The vapor pressure data obtained experimentally are given in Table I. These data were fitted to the following equation [2]:

$$\ln P = \ln P_c + (T_c/T)(aX + bX^{1.5} + cX^3 + dX^6) \tag{2}$$

where P is in MPa, T is in K, $X = (1 - T/T_c)$, $a = -7.76544$, $b = 2.36923$, $c = -9.33775$, and $d = 217.292$. The calculated pressures of this equation are also given in Table I. This equation is found to reproduce experimental data within standard and maximum deviations of 0.39 and 0.87%, respectively.

3.3. Saturated Liquid Density

The saturated liquid densities were extrapolated by introducing the experimental vapor pressure data to the Tait equation, which is described later. The saturated liquid densities are fitted to the following equation:

$$\rho/\rho_c = 1 + AX^{1/3} + BX^{2/3} + CX + DX^{4/3} \tag{3}$$

where $X = (1 - T/T_c)$, $A = 4.3725$, $B = -15.239$, $C = 32.289$, and $D = -21.211$.

Table II. Compressibility Factor and Density of Superheated Vapor

P (MPa)	Z	ρ (kg · m^{-3})
	298.15 K	
0.376	0.8961	23.10[a]
0.360	0.8939	22.17
0.333	0.9011	20.31
0.292	0.9198	17.47
	323.15 K	
0.761	0.8241	46.93[a]
0.705	0.8477	42.22
0.659	0.8612	38.88
0.565	0.8851	32.43
0.473	0.9070	26.48
0.405	0.9197	22.39
0.300	0.9431	16.15
0.249	0.9516	13.30
	348.15 K	
1.401	0.7292	90.57[a]
1.391	0.7270	90.22
1.315	0.7564	81.10
1.303	0.7607	80.73
1.208	0.7815	72.85
1.191	0.7878	71.25
1.099	0.8061	64.25
1.077	0.8131	62.44
0.936	0.8393	52.60
0.797	0.8669	43.32
0.758	0.8754	40.84
0.656	0.8926	34.62
0.611	0.9034	31.91
0.539	0.9157	27.77
0.420	0.9337	21.21
0.304	0.9557	14.97
0.261	0.9583	12.82

[a] Saturated state calculated by Eq. (4).

Table II. *(Continued)*

P (MPa)	Z	ρ (kg · m^{-3})
	373.15 K	
2.367	0.6076	171.35[a]
2.199	0.6550	147.69
2.076	0.6857	133.16
1.931	0.7178	118.31
1.831	0.7354	109.49
1.773	0.7493	104.08
1.682	0.7647	96.75
1.570	0.7861	87.88
1.496	0.7891	82.44
1.365	0.8206	73.17
1.285	0.8335	67.80
1.136	0.8554	58.42
1.051	0.8684	53.26
0.881	0.8922	43.44
0.614	0.9246	29.21
0.325	0.9621	14.86
	398.15 K	
5.590	0.2521	914.20
4.848	0.2302	868.31
4.357	0.2191	819.74
4.099	0.2183	774.04
3.953	0.2246	725.56
3.880	0.2360	677.76
3.844	0.2519	629.06
3.822	0.2713	580.82
3.809	0.2944	533.42
3.789	0.3220	485.06
3.759	0.3550	436.56
3.712	0.3947	387.72
3.639	0.4424	339.08
3.509	0.4978	290.58
3.290	0.5614	241.63
3.113	0.5996	214.01
2.986	0.6251	196.96
2.959	0.6330	192.74
2.840	0.6535	179.16
2.676	0.6817	161.85
2.488	0.7121	144.06
2.485	0.7134	143.59
2.275	0.7427	126.30
2.040	0.7749	108.53
1.850	0.8023	95.03
1.775	0.8085	90.49
0.981	0.8982	45.01

Table II. *(Continued)*

P (MPa)	Z	ρ (kg · m^{-3})
	423.15 K	
5.694	0.3766	586.49
5.470	0.3962	535.54
5.278	0.4197	487.78
5.069	0.4490	437.97
4.838	0.4845	387.36
4.575	0.5250	338.07
4.261	0.5722	288.85
3.861	0.6262	239.17
3.616	0.6590	212.83
3.372	0.6871	190.39
3.251	0.7017	179.69
2.831	0.7468	147.05
2.741	0.7561	140.62
2.333	0.7944	113.93
1.949	0.8310	90.97
1.834	0.8400	84.70
1.579	0.8660	70.71
1.341	0.8842	58.85

3.4. *PVT* Relations of the Superheated Vapor

The *PVT* relations of the superheated vapor obtained are given in Table II. The data were correlated by the modified BWR equation of state [3]:

$$\begin{aligned} P = {} & RT\rho' + (A_1 T + A_2 + A_3/T^2)\,\rho'^2 + (A_4 T + A_5)\,\rho'^3 \\ & + (A_6 + A_7/T^5)\,\rho'^4 + (A_8 + A_9/T^3)\,\rho'^5 + (A_{10} + A_{11}/T)\,\rho'^6 \\ & + (A_{12} + A_{13}/T^2 + A_{14}/T^4)(1 + \beta\rho'^2)\,\rho'^3 \exp(-\beta\rho'^2) \end{aligned} \tag{4}$$

Table III. Numerical Constants of the Modified BWR Equation

A_1	0.1511379136×10^4	A_2	$-0.1598255008 \times 10^7$
A_3	$-0.1180134912 \times 10^{11}$	A_4	0.3623657600×10^8
A_5	$0.3982231232 \times 10^{11}$	A_6	$-0.1516338624 \times 10^{12}$
A_7	$0.6304719872 \times 10^{24}$	A_8	$0.4768674304 \times 10^{14}$
A_9	$-0.8002150400 \times 10^{21}$	A_{10}	$-0.8665377792 \times 10^{16}$
A_{11}	$0.6872051200 \times 10^{18}$	A_{12}	$-0.6670848512 \times 10^{11}$
A_{13}	$0.2926245728 \times 10^{16}$	A_{14}	$-0.1452375392 \times 10^{21}$
β	4570.0		

Table IV. Specific Volume of Liquid R124

P (MPa)	v ($\mathrm{dm^3 \cdot kg^{-1}}$)
283.15 K	
0.2	0.7130[a]
0.6	0.7124
1.4	0.7105
2.3	0.7087
3.6	0.7063
5.9	0.7020
6.0	0.7018
8.9	0.6967
12.2	0.6916
17.0	0.6847
24.6	0.6751
32.0	0.6670
32.3	0.6667
38.6	0.6607
47.1	0.6532
54.3	0.6478
60.6	0.6433
67.2	0.6388
75.1	0.6339
82.3	0.6296
86.5	0.6272
91.4	0.6247
100.5	0.6202
108.0	0.6164
298.15 K	
0.4	0.7365[a]
0.9	0.7351
5.4	0.7237
8.6	0.7164
8.7	0.7165
14.6	0.7052
20.6	0.6956
26.4	0.6874
34.4	0.6778
40.4	0.6713
47.4	0.6646
56.5	0.6565
93.7	0.6316
107.8	0.6241

[a] Saturated state calculated by Eq. (5).

Table IV. *(Continued)*

P (MPa)	v ($\mathrm{dm^3 \cdot kg^{-1}}$)
323.15 K	
0.8	0.7828[a]
1.6	0.7791
5.6	0.7617
8.4	0.7516
15.3	0.7325
21.8	0.7182
27.3	0.7076
35.2	0.6952
40.8	0.6874
54.5	0.6714
63.7	0.6625
68.9	0.6577
82.9	0.6465
95.9	0.6373
106.1	0.6309
348.15 K	
1.4	0.8640[a]
2.3	0.8552
3.7	0.8423
4.8	0.8336
7.0	0.8193
14.7	0.7839
21.2	0.7629
26.6	0.7494
32.4	0.7376
36.6	0.7295
36.7	0.7294
45.3	0.7159
47.7	0.7123
53.4	0.7047
59.7	0.6971
69.1	0.6873
75.0	0.6817
80.6	0.6772
87.7	0.6708
91.7	0.6677
101.5	0.6607
109.5	0.6550
109.7	0.6553

Table IV. *(Continued)*

P (MPa)	v (dm$^3 \cdot$ kg^{-1})
373.15 K	
2.4	0.9918[a]
2.9	0.9760
3.3	0.9640
3.7	0.9554
4.1	0.9465
4.5	0.9385
5.0	0.9285
6.2	0.9135
6.8	0.9045
8.4	0.8868
10.3	0.8699
12.7	0.8526
15.3	0.8381
17.5	0.8271
19.9	0.8163
23.2	0.8036
27.2	0.7905
31.1	0.7794
35.8	0.7684
41.2	0.7568
46.1	0.7475
50.5	0.7398
57.4	0.7292
64.1	0.7204
64.6	0.7198
70.4	0.7126
77.5	0.7049
82.5	0.6996
88.1	0.6942
93.5	0.6894
99.0	0.6849
105.9	0.6795

Table IV. *(Continued)*

P (MPa)	v ($\mathrm{dm^3 \cdot kg^{-1}}$)
398.15 K	
4.4	1.2112
4.6	1.1773
5.0	1.1284
5.2	1.1145
5.8	1.0671
6.5	1.0369
7.9	0.9978
9.4	0.9622
12.9	0.9131
15.4	0.8898
18.5	0.8681
22.9	0.8417
28.6	0.8162
31.5	0.8053
35.8	0.7918
40.3	0.7728
46.0	0.7664
53.4	0.7510
59.1	0.7409
73.1	0.7205
85.6	0.7056
93.8	0.6971
99.8	0.6908

where P is in MPa, T is in K, and ρ' is in $\mathrm{mol \cdot cm^{-3}}$. The numerical constants of Eq. (4) were determined by a least-squares method and are listed in Table III. The deviations of the experimental superheated vapor densities from those values calculated by Eq. (4) are shown in Fig. 3. The pressure dependence of the compressibility factor is shown in Fig. 5.

3.5. *PVT* Relations of the Liquid

The specific volume data obtained are listed in Table IV. These data were correlated as a function of pressure by the Tait equation [4]:

$$k = \frac{v_0 - v}{v_0} = C \ln \frac{B + P}{B + P_0} \tag{5}$$

where k is the isothermal compression, v_0 is the specific volume at a

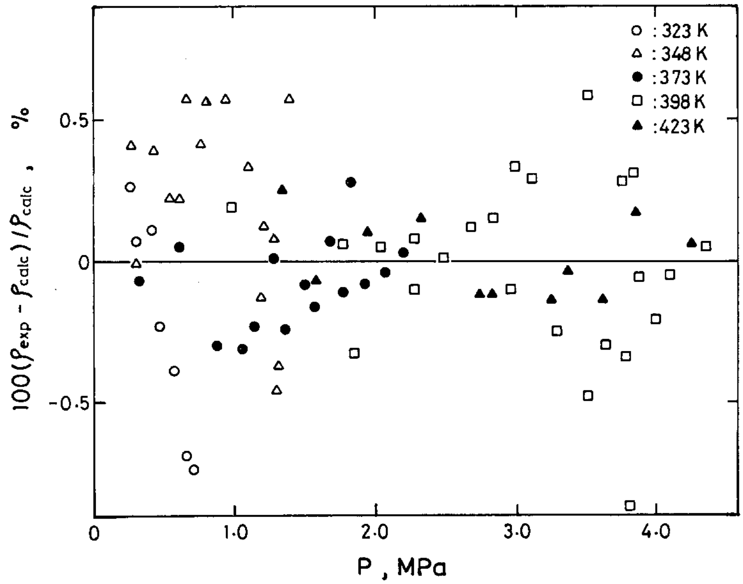

Fig. 3. Deviations of the superheated vapor densities of R124 from Eq. (4).

reference pressure P_0, and B and C are constants. The empirical constants B and C were determined by a least-squares method as given in Table V together with the standard and maximum deviations of the experimental data from those calculated by the above equation. The pressure dependence of the isothermal compression is shown in Fig. 4. The variation of compressibility factors of R124 in the gaseous and liquid phases is shown in Fig. 5.

Table V. Coefficients of the Tait Equation

T(K)	P_0 (MPa)	v_0 (dm$^3 \cdot$kg^{-1})	B (MPa)	$C \times 10^2$	SD (−)[a]	Max. dev. (%)
283.15	0.24	0.7130	28.78	8.719	9.4×10^{-5}	3.4×10^{-2}
298.15	0.38	0.7365	22.29	8.726	9.9×10^{-5}	2.7×10^{-2}
323.15	0.76	0.7827	13.46	9.100	2.3×10^{-4}	5.7×10^{-2}
348.15	1.40	0.8640	5.338	8.513	2.0×10^{-4}	7.3×10^{-2}
373.15	2.37	0.9918	−0.03815	8.257	3.4×10^{-4}	9.3×10^{-2}
398.15	4.37	1.211	−3.782	8.663	8.3×10^{-3}	2.2

[a] SD = $[(v_{exp} - v_{calc})^2/n]^{1/2}$.

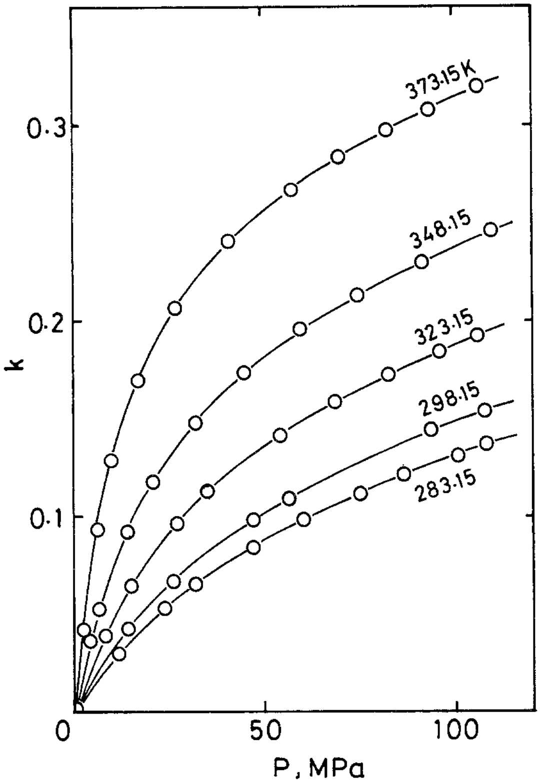

Fig. 4. Isothermal compression of liquid R124 against pressure.

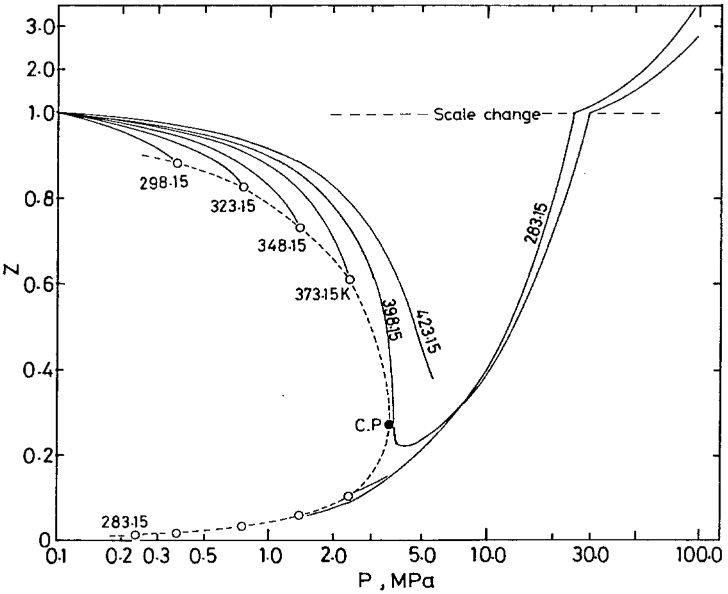

Fig. 5. Compressibility factor of R124 against pressure.

Table VI. Thermodynamic Properties of R124[a]

P (MPa)	ρ ($kg \cdot m^{-3}$)	H ($kJ \cdot kg^{-1}$)	S ($kJ \cdot kg^{-1} \cdot K^{-1}$)	C_p ($kJ \cdot kg^{-1} \cdot K^{-1}$)	C_v ($kJ \cdot kg^{-1} \cdot K^{-1}$)	u ($m \cdot s^{-1}$)
303.15 K						
0.10	5.532	346.91	1.569	0.7324	0.6659	139.4
0.20	11.342	345.51	1.524	0.7530	0.6764	136.5
0.30	17.518	343.75	1.495	0.7885	0.6948	133.2
0.40	24.190	341.51	1.471	0.8455	0.7232	129.4
313.15 K						
0.10	5.344	354.31	1.593	0.7470	0.6813	141.8
0.20	10.923	353.09	1.548	0.7624	0.6890	139.2
0.30	16.791	351.62	1.520	0.7876	0.7023	136.3
0.50	29.734	347.71	1.479	0.8824	0.7500	129.6
323.15 K						
0.10	5.170	361.85	1.617	0.7615 0.7697[b]	0.6965	144.1
0.30	16.148	359.51	1.545	0.7915	0.7116	139.2
0.50	28.274	356.38	1.507	0.8551 0.8329[b]	0.7449	133.6
0.70	42.185	352.07	1.475	0.9801	0.8043	126.8
348.15 K						
0.10	4.783	381.33	1.675	0.7968 0.8034[b]	0.7327	149.7
0.50	25.505	377.45	1.570	0.8388 0.8402[b]	0.7532	141.6
1.00	56.755	370.42	1.512	0.9920 0.9242[b]	0.8311	129.3
1.30	80.865	364.08	1.481	1.2237	0.9290	119.7
373.15 K						
0.10	4.451	401.67	1.731	0.8304 0.8335[b]	0.7670	155.0
0.50	23.409	398.55	1.628	0.8518 0.8631[b]	0.7749	148.3
1.00	50.388	393.89	1.577	0.9072 0.9098[b]	0.8011	139.1
2.00	125.243	379.85	1.506	1.2814	0.9592	114.7
398.15 K						
0.10	4.164	422.84	1.786	0.8627 0.8669[b]	0.7997	160.2
1.00	45.918	416.37	1.635	0.8978 0.9175[b]	0.8068	147.1
2.00	105.648	407.19	1.577	0.9896	0.8274	130.3
3.00	198.055	393.50	1.525	1.2915	0.8726	107.3
423.15 K						
0.10	3.912	444.80	1.840	0.8940 0.8977[b]	0.8312	165.1
1.00	42.351	438.97	1.690	0.9127 0.9358[b]	0.8268	154.4
2.00	93.801	431.11	1.635	0.9402	0.8028	142.8
4.00	254.835	406.23	1.545	1.2120	0.5729	132.8

[a] Complete sets of data covering temperatures from 303.15 to 423.15 K and pressures up to 4.4 MPa are available on request.

[b] Experimental value of Bender et al. [5].

3.6. Thermodynamic Properties

Equations (2)–(4), representing the physical properties of R124, are used to derive the thermodynamic properties of the saturated and superheated vapor. The ideal-gas heat capacity data are obtained from the literature [5]. The saturated liquid at 303.15 K was chosen as a reference state, where the enthalpy and entropy values are defined as $200\ \mathrm{kJ \cdot kg^{-1}}$ and $1\ \mathrm{kJ \cdot kg^{-1} \cdot K^{-1}}$, respectively. The thermodynamic properties such as enthalpy (H), entropy (S), isobaric specific heat capacity (C_p), isochoric specific heat capacity (C_v), and speed of sound (u) for the superheated state are tabulated in Table VI.

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