# Measurements of the p-v-T Behavior of Refrigerant R134a in the Liquid Phase<sup>1</sup>

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A short description of an improved *p-v-T* apparatus with a constant-volume piezometer is given. *p-v-T* data of the environmentally safe refrigerant 1, 1, 1, 2-tetrafluoroethane (R134a), were measured for temperatures between 205 and 309 K at pressures up to 56 MPa. The uncertainty of temperature and pressure measurements was estimated to be  $\pm 0.05^{\circ}$ C and  $\pm 0.1\%$ , respectively; uncertainty in the specific volume was estimated to be  $\pm 0.15\%$ . Purity of the sample used was 99.9 + wt%. The data are represented analytically in order to demonstrate experimental accuracy and to facilitate calculation of thermodynamic properties.

**KEY WORDS:** equation of state; p-v-T measurements; R134a; refrigerants; 1, 1, 1, 2-tetrafluoroethane.

# **1. INTRODUCTION**

The hydrofluorocarbon R134a ( $CH_2FCF_3$ ) has been proposed as a substitute for R12 ( $CCl_2F_2$ ), which is used in refrigeration and heat-pump technology. In contrast to R12, which has an ozone depletion potential (ODP) of 1, the refrigerant R 134a does not deplete the stratospheric ozone layer. Information on its thermodynamic properties is available along the saturation curve [1–8] and in the gaseous phase [1, 2, 9, 10], but in the liquid phase very few data have been published to date.

In the present study, measurements of the *p*-v-T surface are given for R134a in its liquid phase. The measurements were made on an improved constant-volume apparatus developed for measurements in the range between -70 and  $40^{\circ}$ C at pressures up to 60 MPa.

<sup>&</sup>lt;sup>1</sup> Paper dedicated to Professor Joseph Kestin.

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A brief description of the apparatus, measuring techniques, and discussion of results are presented. This paper is intended to fill the void of property information on R134a in the liquid region and, also, to provide future users with an interpolating equation.

# 2.. EXPERIMENTS

### 2.1. Apparatus

An apparatus based on the constant-volume method has been used for combined measurements along quasi-isochores and isotherms. Measurements along quasi-isochores enable one to obtain several points with one filling at various temperatures; measurements along isotherms require a new filling for each measured point.

The experimental device consists of a piezometer, pressure and temperature measuring system, temperature control system, filling system, and combined air/liquid cryostat. A schematic diagram of the apparatus is given in Fig. 1.

The piezometer is a thick-walled cylindrical vessel with a spherical bottom, made from stainless steel 17 246 (equivalent to H 11 ISO 4955-83), with a capacity of about 200 cm<sup>3</sup>. The closure is a flat flange cover with six screws and flat copper gasket.

The piezometer volume was determined by calibration with mercury and water. It is connected with a shutoff valve and a differential pressure null indicator-DPI (Ruska, Type 2413-703), located outside the air cryostat, and a control box/null indicator (Ruska, Type 2416-711). A reference U-tube is used for setting "zero" on the DPI. The total volume of the piezometer, shutoff valve, tubing to the DPI, and its volume is 205.650 cm<sup>3</sup>. The "noxious" volume, i.e., volume of the connecting tube and of the DPI, kept at 25°C, is 1.473 cm<sup>3</sup>. The diaphragm DPI is connected to the pressure measuring system. This consists of a two-range deadweight oil-operated gauge (Budenberg, Type 380 D) accurate within 0.012% for the lower and 0.017% for the upper range. The second part of the system consists of a set of Bourdon tube gauges (WIKA, Type 342.11) with an accuracy of 0.1% FSR, and ranges of 600, 250, 100, 25 and 10 bars, calibrated before and after measurements to the deadweight gauge. Pressure in the positive displacement pump was measured by a pressure transducer with digital pressure indication (WIKA Tronic).

Temperature was measured by means of two  $25-\Omega$  platinum resistance thermometers (Tinsley, Type 5187 SA, and Sensing Devices Ltd./ Rosemount). The first thermometer was located in the liquid cryostat bath; the second one, in the body of the piezometer. Two further thermometers

served for the temperature measurements of the DPI and volumeter which were thermostated to approx. 25°C. The thermometers were interconnected through four leads each, with a selector switch (Tinsley, Type 5840 CS/6) and an automatic AC thermometer bridge (Tinsley, Type 5840D-Senator). Analog output of the bridge was used for the registration of the time



Fig. 1. Experimental apparatus. (A) Evaporator of a cascade refrigerator, (B) fan, (C) evaporator for the liquid bath, (D) piezometer, (E) air cryostat with insulation, (F) filling bomb, (G) liquid cryostat with cover, (H) heaters, (I) insert of the liquid cryostat, (J) control box of the freezer, (K) pump with stirrer and distributing tube ring, (L) platinum resistance thermometers, (M) thermometer selector switch, (N) thermocouple selector switch, (O) thermometer bridge F 16, (P) digital voltmeter, (Q) thermometer bridge Senator, (R) external standard resistor, (S) pen recorder, (T) thermostat, (U) pressure transducer with digital indication, (V) supply vessel, (VP) vacuum pump with gauges, (W) wattmeter, (X) oil reservoir, (Y) reference U-tube, (Z) distribution piece, (BTG) Bourdon gauges, (CB) control box/null indicator, (DPI) differential pressure indicator, (DWG) deadweight gauge, (PDP) positive displacement pump/volumeter, (PID) PID controller, and (VD) voltage divider.

history of the piezometer and the volumeter temperature by means of a pen recorder. This enabled us to follow the stabilization of temperature, long-term stability of the control system, and temperature fluctuations around the set value. The accuracy of temperature measured with the Tinsley thermometer was  $\pm 5$  mK, that with S.D. Ltd.  $\pm 10$  mK, and that with the others  $\pm 0.02$  K. The maximum uncertainty in the piezometer temperature was estimated to be  $\pm 0.05$ °C at the lowest temperatures.

In addition to the mentioned thermometers, another Pt thermometer, located on the lower inner part of the liquid cryostat cylindrical insert, served as a sensor for the temperature control. Distribution of temperatures in the area of the liquid and air cryostat was monitored by thermocouples (Hereaus, Ni–NiCr,  $\phi$  1.5 mm) connected through a selector switch to the digital voltmeter.

A modified freezer (Frigera, Type NZ 280/75) with working temperatures down to  $-75^{\circ}$ C and an output of 150 W at an ambient temperature of 25°C was used as a cryostat. It consisted of a cascade two-stage cycle using refrigerants R12 and R13. Inside the freezing room an evaporator with a fan was located. In the middle of the freezing room a liquid bath with approx. 50 L of ethanol was situated. A pump with a stirrer and distribution tube ring located in the upper part of the bath agitated and circulated the bath. The insert of the liquid cryostat was furnished with an evaporating spiral tube for rapid cooling down. Four heaters in series-parallel connection with an output of 2 kW were located in the bath. The requested temperature was set by a built-in resistance decade of a thermometer bridge (ASL Ltd., Model F 16) furnished with an external standard resistor in a thermostated bath. The control voltage for the PID controller (UNIPAN, Type 650H) was taken from the analog output of the bridge F 16 through a voltage divider. The output of the heater, monitored by a wattmeter, was between 20 and 30 W at the steady state. The deviations of the set temperature were registered on a pen recorder. A built-in control box in the freezer-regulated air temperature in the cryostat. The air temperature was kept approx. 2°C lower than the liquid bath temperature. The temperature was stable within several millikelvins.

A positive displacement pump/volumeter (Ruska, Model 2270-803) with a total discharge volume of  $250 \text{ cm}^3$  was used for the filling of the piezometer. The discharged volume was read off on a scale and a dial with resolution of 1 and 0.01 cm<sup>3</sup>, respectively; the vernier enabled determination of 0.001 cm<sup>3</sup>. The volumeter was connected with a five-valve distributing piece, which was linked with the piezometer, a filling bomb, and a high-vacuum pump consisting of a rotary and diffusion pump with vacuum gauges and valve system.

#### p-v-T Behavior of Refrigerant R134a

For the evaluation of the measurements all necessary corrections such as change of piezometer volume with pressure and temperature, "noxious" volumes, differences in hydrostatic heads of pressure gauges and of the piezometer, and so on, were taken into account. The noxious volumes were determined by calculation or with calibration, where it was possible.

The purity of the studied sample, supplied by the courtesy of Hoechst A.G. Frankfurt am Main, FRG, was 99.9 wt%. Composition reported by the manufacturer corresponds to the standard DIN 8960: noncondensable gases, <1.5 vol%; moisture, <10 H<sub>2</sub>O mg·kg<sup>-1</sup>; change of boiling point temperature, <0.3 K; and admixtures, <50 mg·kg<sup>-1</sup>. The gas sample was slowly distilled from the supplier bomb into a filling bomb solidified by means of liquid nitrogen and degassed by vacuum sublimation.

#### 2.2. Procedure

After a tightness test the whole apparatus was evacuated and the volumeter was filled to the reference state 25°C and 1 MPa. This state was carefully maintained at the filling or withdrawal of a sample mass in/from the piezometer. Then the piezometer was filled from the volumeter. At measurements along quasi-isochores, the initial point was always remeasured to confirm that the device was tight. Before starting new measurements along isotherms, the liquid in the volumeter was compressed to 50 MPa and relieved to 1 MPa to dissolve the released gases, if any. The



Fig. 2. Detailed chart of measurements for R134a Normal font, points measured along isochores; *italic font*, points measured along isotherms; **bold font**, points common for isochores and isotherms.

transition from the quasi-isochore to the isotherm was carried out at suitable state parameters to cover the studied region evenly. Measuring along an isotherm was carried out by pressing or withdrawing of a defined quantity of the sample using the volumeter. The entire course of the measurement is shown in Fig. 2. In the crucial points, i.e., the starting points of the quasi-isochores and isotherms, as well as in transition points from quasi-isochores to isotherms, the state values were measured several times.

For calculating density it is necessary to determine the density at the reference state. This was done by weighing the filling bomb of a well-known volume filled with R134a at the reference state. The density obtained for 24.994°C and 1 MPa was 1.2091 g  $\cdot$  cm<sup>-3</sup>, which is in fair agreement with the density 1.2087 g  $\cdot$  cm<sup>-3</sup> gained by interpolation in data reported by Morrison and Ward [11]. The percentage deviation was 0.035%.

# 3. RESULTS

Measurements have been performed along five quasi-isochores and four isotherms in the liquid region from 204.3 to 309.6 K at pressures between approx. 1 and 56 MPa. Results obtained by the two-way classification method [12, 13], evaluating measured quantities and taking into account all necessary corrections, are presented in Table I. The columns show the number of the measured point, temperature in K (ITS-90), pressure in MPa, density in  $g \cdot cm^{-3}$ , and deviation of the measured density and the calculated one from the derived equation of state Eq. (1). Through multiple measured points, the maximum percentage uncertainty was evaluated to be of about 0.15%.

A short-range equation of state in the form

$$\rho = \rho_{\text{ref}} \sum_{j=1}^{11} b_j \pi^{m_j} \tau^{n_j/2}$$
(1)

where  $\pi = p/p_1$ ,  $\tau = T/T_1$ ,  $p_1 = 100$  MPa,  $T_1 = 1000$  K, and  $\rho_{ref} = 1.2091$  g cm<sup>-3</sup>, was derived by maximum-likelihood fit, independently on the above-mentioned evaluation. The coefficients and exponents of Eq. (1) are in Table II. In Table III, densities calculated from Eq. (1) are listed at roundoff pressures and temperatures.

	Т	р	ρ	$\Delta  ho /  ho^a$
No.	(K)	(MPa)	$(g \cdot cm^{-3})$	(%)
26	236.045	54.753	1.5055	0.03
27	229,120	44.353	1.5062	0.04
28	223.337	35.623	1.5068	0.03
29	216 734	25 503	1 5075	0.04
30	210.499	15875	1 5082	0.04
31	204 289	6173	1.5082	0.04
22	236.040	54 803	1.5055	0.04
32	200.040	6 5 2 2	1.5095	0.03
33	204.451	0.323	1.5009	0.02
40	204.414	0.073	1.5092	0.05
41	235.992	54.904	1.5058	0.05
15	267.602	54.304	1.4389	0.02
16	260.669	45.854	1.4395	0.03
17	252.501	35.744	1.4403	0.02
18	244.493	25.704	1.4411	0.02
19	236.404	15.530	1.4419	0.02
20	227.701	4,343	1.4429	0.03
21	225 267	1 202	1.4432	0.02
22	267 690	54 503	1 4389	0.00
22	236,030	15.055	1.4420	0.00
18	236.039	14 455	1.4406	0.02
40	250.028	52 254	1.4375	0.01
47	207.571	55.554	1.4375	0.03
4	298.303	54.904	1.3764	0.02
5	287.437	43.909	1.3774	0.05
6	277.149	33.304	1.3783	0.02
7	267.595	23.395	1.3792	0.01
8	259.520	14.865	1.3800	0.03
9	251.784	6.683	1.3807	0.03
10	246.640	1.180	1.3813	0.01
11	298.293	54.954	1.3764	0.01
12	267.600	23,465	1.3792	-0.00
57	267.573	24.616	1.3820	0.01
58	298.295	56.254	1.3792	0.01
66	298.301	25.854	1.3083	0.05
67	287.085	16.331	1.3093	0.09
68	275.005	6.014	1.3104	0.11
69	269.647	1.446	1.3109	0.07
70	298.296	25.784	1.3083	0.07
71	309.698	35.394	- 1.3074	0.08
72	298.332	25.894	1.3083	0.05
73	309.729	35.404	1.3073	0.08
74	200 722	16 521	1 3457	0.12
/4	309.733	10.531	1.2457	0.12
15	292.972	4.699	1.24/1	0.13
/6	288.240	1.343	1.24/5	0.08
11	309.730	16.596	1.2457	0.10

Table I.Experimental p- $\rho$ -T Data of R134a

 $^{a} \Delta \rho / \rho = 100(\rho_{\text{exp.}} - \rho_{\text{Eq.}(1)}) / \rho_{\text{Eq.}(1)}.$ 

	Т	D	Ø	$\Delta \rho / \rho^a$	
No.	(K)	(MPa)	$(g \cdot cm^{-3})$	(%)	
33	204.431	6.523	1.5089	0.02	
34	204.461	1.552	1.5005	0.00	
35	204.430	15.680	1.5227	0.01	
36	204.441	25.105	1.5358	-0.00	
37	204.450	35.704	1.5495	-0.02	
38	204.411	45.864	1.5616	-0.02	
39	204.411	54.154	1.5712	0.03	
40	204.414	6.673	1.5092	0.03	
23	236.039	15.055	1.4420	0.02	
24	236.040	55.703	1.5068	0.03	
25	236.040	15.005	1.4422	0.05	
26	236.045	54.753	1.5055	0.03	
41	235.992	54.904	1.5058	0.03	
42	236.028	1.048	1.4118	-0.02	
43	236.025	6.803	1.4250	0.01	
44	236.025	14.705	1.4411	0.01	
45	236.026	25.304	1.4607	0.02	
46	236.027	35.454	1.4774	0.02	
47	236.028	45.854	1.4930	0.02	
48	236.028	14.455	1.4406	0.01	
12	267.600	23.465	1.3792	-0.00	
13	267.599	56.204	1.4421	0.01	
14	267.600	23.095	1.3783	-0.01	
15	267.602	54.304	1.4389	0.02	
49	267.571	53.354	1.4375	0.03	
50	267.572	1.399	1.3173	0.05	
51	267.572	5.243	1.3302	0.06	
52	267.574	15.005	1.3586	0.03	
53	267.574	24.345	1.3813	0.00	
54	267.574	34.754	1.4037	0.02	
55	267.573	45.304	1.4234	0.03	
56	267.573	54.704	1.4394	0.00	
1	298.303	1.139	1.2090	-0.03	
2	298.304	54.504	1.3755	0.02	
3	298.302	1.110	1.2095	0.03	
4	298.303	54.904	1.3764	0.02	
58	298.295	56.254	1.3792	0.01	
59	298.295	1.731	1.2126	0.01	
60	298.298	6.323	1.2366	0.13	
61	298.297	14.146	1.2696	0.11	
62	298.299	24.456	1.3041	0.05	
63	298.295	34.954	1.3327	0.05	
64	298.297	46.404	1.3591	0.08	
65	298.297	56 104	1 3791	0.03	

Table I. (Continued)

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j	$m_j$	$n_j$	$b_j$
1	2	8	$-3.305 416 276 \times 10^{2}$
2	0	3	$-1.529\ 376\ 192 \times 10^{12}$
3	1	6	$1.510\ 546\ 636 \times 10^2$
4	3	8	$1.414\ 152\ 223 \times 10^2$
5	0	2	$1.054~978~718 \times 10^{1}$
6	3	6	$-2.929\ 172\ 880 \times 10^{1}$
7	2	7	$1.444\ 138\ 672 \times 10^2$
8	2	-8	$-4.591\ 144\ 886 \times 10^{-5}$
9	1	5	$-9.989\ 153\ 616 \times 10^{1}$
10	1	2	$7.937\ 265\ 555  imes 10^{0}$
11	0	-2	$0.101\ 363\ 421 \times 10^{0}$

Table II. Coefficients and exponents in Eq. (1)

Table III. Densities of R134a Calculated from Eq. (1)

			Density (k	$g \cdot m^{-3}$ ) at	a pressure	of (MPa)		
t(°C)	1	5	10	20	30	40	50	55
-65	1489.6	1496.3	1504.4	1519.6	1533.7	1546.6	1558.3	1563.7
-60	1476.0	1483.0	1491.5	1507.4	1522.0	1535.3	1547.5	1553.1
- 55	1462.3	1469.6	1478.5	1495.1	1510.2	1524.1	1536.8	1542.7
-50	1448.4	1456.2	1465.5	1482.8	1498.5	1512.9	1526.0	1532.2
45	1434.4	1442.6	1452.4	1470.5	1486.9	1501.7	1515.4	1521.8
-40	1420.2	1428.9	1439.2	1458.1	1475.2	1490.6	1504.7	1511.4
-35	1405.7	1415.0	1425.8	1445.7	1463.4	1479.4	1494.0	1501.0
-30	1391.1	1400.8	1412.4	1433.2	1451.7	1468.2	1483.4	1490.6
- 25	1376.1	1386.5	1398.7	1420.7	1439.9	1457.0	1472.7	1480.2
-20	1360.9	1372.0	1384.9	1408.0	1428.1	1445.8	1462.0	1469.8
15	1345.4	1357.2	1370.9	1395.3	1416.2	1434.6	1451.4	1459.4
-10	1329.5	134.2	1356.8	1382.4	1404.3	1423.4	1440.7	1449.1
-5	1313.4	1326.9	1342.4	1369.5	1392.3	1412.1	1430.1	1438.7
0	1296.9	1311.3	1327.8	1356.4	1380.3	1400.8	1419.4	1428.4
5	1280.1	1295.5	1313.1	1343.3	1368.3	1389.6	1408.7	1418.0
10	1262.9	1279.4	1298.1	1330.0	1356.2	1378.3	1398.1	1407.6
15	1245,3	1263.0	1282.9	1316.7	1344.0	1366.9	1387.4	1397.3
20	1227.4	1246.3	1267.5	1303.2	1331.9	1355.6	1376.7	1387.0
25	1209.1	1229.3	1251.9	1289.7	1319.6	1344.3	1366.1	1376.7
30	1190.5	1212.0	1236.0	1276.0	1307.4	1333.0	1355.4	1366.3
35	1171.4	1194.4	1220.0	1262.3	1295.1	1321.6	1344.8	1356.0

#### 4. DISCUSSION

Measurements for R134a in the liquid region are very scarce. Morrison and Ward [11] have measured compressed liquid by means of a vibrating tube densimeter between 279 and 367 K at pressures from 0.7 to 6 MPa. Forty-four points are overlapping with our measurements. Maezawa et al. [4] reported 10 measured data points determined by means of a magnetic densimeter between 280 and 340 K at 0.5 to 2 MPa in the liquid region, only four points overlap with our measurements. Kesselman et al. [14] measured 65 points in the liquid region between 212 and 368 K at pressures from 0.5 to 20 MPa, only 4 points are over 10 MPa; 29 data points overlap with our range. They also published an equation of state and tabulated densities from 210 to 345 K and 0.1 to 20 MPa. Two other papers [15, 16] also mention measurements, but do not present experimental data. Ruvinskij et al. [15] reported measurements from 26 to 1383 kg  $m^{-3}$  along isochores up to 7 MPa and an equation of state based on these 229 measured points. The same case is with the paper by Dobrochotov et al. [16], referring to measurements carried out in the Moscow Power Institute from 200 to 315 K and 1.1 to 55 MPa with densities 1140 to 1510 kg · m<sup>-3</sup> as well as to measurements carried out at the Odessa Institute of Low-Temperature Technology and Energetics in the liquid region. Data were used for construction of an equation of state but are not available.

The latest paper obtained shortly before publication of the present measurements by Hou et al. [17] presents density measurements obtained with a continuously weighed pycnometer along 14 isotherms between 180 and 379 K at pressures up to 70 MPa. Magee [19] measured the *p*-*v*-*T* behavior simulaneously with the heat capacity at constant volume in the range from 172 to 343 K at pressures up to 35 MPa.

Comparisons of our data calculated from Eq. (1) with the experimental data of Morrison and Ward [11], Maezawa et al. [4], Kesselman et al. [14], Hou et al. [17], and Magee [19] are presented in Table IV.

There is fairly close agreement between our results and those of by Morrison and Ward [11]. Deviation greater than 0.13% is in 4 points only, the maximum deviation is 0.225%. Points with these large deviations are at the beginning of two isotherms. Densities by Maezawa et al. [4] in all four points are smaller than those calculated from Eq. (1) by 0.138 to 0.205%. Densities by Keselman et al. [14] except for four points are greater than those calculated from Eq. (1). The deviations of 13 points are between -0.028 and 0.071%, 9 points deviate within 0.161 to 0.36%; 4 points along the isotherm 274.14 K yield deviations of about 0.42% and at 3 points of the quasi-isochore 1301 kg  $\cdot m^{-3}$  deviations increase from 0.46

	Deviation (%)				
Author(s)	$N^a$	Standard <sup>b</sup>	Systematic <sup>c</sup>	Mean <sup>d</sup>	
Hou et al. [17]	128	0.142	-0.137	0.137	
Morrison & Ward [11]	44	0.068	-0.005	0.050	
Kesselman et al. [14]	29	0.339	0.0226	0.230	
	22*	0.160	0.114	0.119	
Maezava et al. [4]	4	0.179	-0.177	0.177	
Magee [19]	104	0.119	-0.095	0.108	

 Table IV.
 Comparison of Reported Experimental Data by Various Authors with Eq. (1)

<sup>a</sup> Number of compared points.

<sup>b</sup> Standard deviation:  $((1/N) \sum [100(\rho_{exp.} - \rho_{Eq.(1)})/\rho_{Eq.(1)}]^2)^{1/2}$ .

<sup>c</sup> Systematic deviation:  $(1/N) \sum 100(\rho_{exp.} - \rho_{Eq.(1)})/\rho_{Eq.(1)}$ 

<sup>d</sup> Mean deviation:  $(1/N) \sum |100(\rho_{exp.} - \rho_{Eq.(1)})/\rho_{Eq.(1)}|$ .

to 1.078%. After the omission of these seven aforementioned points, supposing them to be unsound, the resulting deviations run down as shown in Table IV, in the row denoted by asterisk. Densities by Hou et al. [17] are systematically lower. Densities reported by Magee [19] with exception of 12 points are also lower then densities calculated from Eq. (1). Deviations are within the band -0.193 to 0.076%.

Comparison of our experimental data with the equations of state proposed by Huber and Ely [18] yields for all 77 data points a standard deviation 0.20%, the mean deviation 0.19%, and a systematic deviation also 0.19%.

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