

## **Thermophysical Properties of 1,1,1,2-Tetrafluoroethane (R-134a)<sup>1</sup>**

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The present hypothesis of depletion of the stratospheric ozone layer by some chlorofluorocarbons has prompted a lot of research and development of new stratospherically safe fluids in various uses such as refrigerants, blowing agents in foams, aerosol propellants, solvents, and many other uses. In the areas of certain refrigeration needs 1,1,1,2-tetrafluoroethane (R-134a) has been considered as a possible alternate to the use of dichloro-difluoromethane (R-12), the most commonly used refrigerant. R-12 is estimated to have a higher potential for ozone depletion. This will require a large number of thermophysical property data to help in designing equipment and also in manufacturing R-134a. This paper is intended to fill that need. The paper details the measurement and correlation of some of the important thermophysical properties such as vapor pressure, liquid density, and pressure-volume-temperature. The measured  $P$ - $V$ - $T$  data have been used to generate a Martin-Hou-type equation of state for this fluid over a wide range of temperature and pressure. Correlating equations are also developed for vapor pressure, liquid density, and ideal-gas specific heat. Ideal-gas specific heat has been estimated from measured spectroscopic data. The correlating equations can be used to generate the thermodynamic tables and charts. The critical temperature of R-134a has also been measured. Critical density and pressure have been estimated from measured data. The data and the correlations presented here are expected to be very useful to the refrigeration industry in the development of R-134a as a working fluid for refrigeration applications.

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**KEY WORDS:** critical constants; ideal-gas heat capacity; liquid density;  $PVT$ ; refrigerant; 1,1,1,2-tetrafluoroethane; vapor pressure.

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## 1. INTRODUCTION

Chlorofluorocarbons (CFCs) are commonly used as refrigerants, as propellants in aerosols, as blowing agents in foams, and as solvents in degreasing and defluxing materials such as printed circuit boards. These fluids are generally inert on the earth's surface, but when they diffuse into the stratosphere, they break down into free radical fragments by intense ultraviolet rays of the sun. The chlorine free radicals released by this process are speculated to deplete the earth's stratospheric ozone layer by a chain reaction with ozone molecules. This theory was first proposed by Molina and Rowland [1]. Recent scientific studies indicate a small reduction in ozone over the last 15 years. As a result of these findings an international agreement has been reached called the Montreal Protocol [2]. The protocol calls for the regulation of consumption of CFCs and halons that deplete the ozone layer.

Among the regulated CFCs the present use of R-12 in the United States is close to 300 million lb per year. It also has a high ozone depletion potential. R-134a (1,1,1,2-tetrafluoroethane) has so far shown to be a promising candidate to replace R-12 in a large number of applications. R-134a does not contain chlorine and therefore has zero ozone depletion potential. It is also found to be reasonably stable and its thermodynamic properties are found to be similar to those of R-12.

In order to assist designers and operators of refrigeration and air-conditioning equipment in evaluating R-134a, basic thermophysical properties and thermodynamic tables must be available. Very few data have been published to date. This paper is intended to fill the void of key property information on R-134a and also to provide future users with correlating equations. The data reported here are critical constants, vapor pressure, liquid density, ideal-gas heat capacity, and  $P$ - $V$ - $T$ . Along with the measured data, correlations with various equations are also shown.

## 2. EXPERIMENTAL METHODOLOGY

The procedures used in the determination of the thermophysical properties of R-134a are described below. The details of apparatuses and methodologies used are described in greater detail in Refs. 3 and 4. In these experiments the samples used had a purity of 99.95 wt% with a moisture level of less than 10 ppm. The amount of noncondensables in the sample was reduced to a pressure of less than 50  $\mu$ m Hg. Temperature measurements were made by calibrated platinum resistance thermometers accurate to  $\pm 0.01^\circ\text{C}$ . Resistances were constant to  $\pm 0.001 \Omega$ , which corresponds to a precision of  $\pm 0.01^\circ\text{C}$ . Pressure measurements were done

with Bourdon tube gauges and pressure transmitters. Depending on the system pressure, gauges encompassing various ranges were used. For most measured pressures this error was less than 0.05% of the full-scale value. Gauges have been periodically calibrated with a primary standard deadweight tester.

## 2.1. Critical Constants

The critical temperature of Refrigerant 134a was determined by the observation of appearance and disappearance of the meniscus of the fluid contained in a heavy-walled, sealed glass tube immersed in an oil bath regulated to  $\pm 0.02^\circ\text{C}$ . The sample was both heated and cooled and passed through the critical temperature to determine the actual appearance and disappearance of the meniscus. The tube was carefully loaded such that just below the critical temperature, approximately equal volumes of liquid and vapor coexisted. The critical pressure was determined by using a six-constant vapor pressure equation to fit saturated vapor pressure data up to  $5^\circ\text{C}$  from the critical point and constraining the slope of the curve to match the critical  $P$ - $V$ - $T$  isochore. The pressure calculated at the critical temperature was determined to be the critical pressure. The critical density was calculated from a rectilinear diameter equation correlating measured liquid and vapor densities. Critical constants with estimated errors are reproduced in Table I.

## 2.2. Vapor Pressure Measurement

The saturated vapor pressure properties of R-134a were determined by condensing the desired weight of sample into a high-pressure bomb fitted with a null-point diaphragm transmitter and pressure balancing circuit, which is a modified version of transmitters described in the literature [5, 6]. By means of a bypass system, effects of temperature and pressure on the diaphragm null point could be quantitatively accounted for.

## 2.3. Saturated Liquid Density Measurement

The saturated liquid densities were determined by standard density floats guaranteed by the manufacturer to be accurate to  $\pm 0.2 \text{ kg} \cdot \text{m}^{-3}$ . The

**Table I.** Critical Constants of R-134a

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$$\begin{aligned}T_C &= 374.25 \pm 0.15 \text{ K} \\P_C &= 4067 \pm 2.7 \text{ kPa} \\ \rho_C &= 512.2 \pm 5 \text{ kg} \cdot \text{m}^{-3}\end{aligned}$$

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float technique, derived from ASTM D1505, was previously tested in our laboratory on  $\text{CCl}_2\text{F}_2$  and  $\text{CCl}_4$  and found to be accurate to  $\pm 0.05\%$  over the range covered. Refrigerant 134a was condensed into a glass tube containing density floats, then given one final degassing to less than  $50\ \mu\text{m Hg}$ , and the end of the tube was sealed. The tube was then thawed and suspended vertically in a liquid bath thermostatted to  $\pm 0.01^\circ\text{C}$ . The temperature required to hold the float freely suspended in the liquid for up to 1 h was recorded. The density of the float was corrected for the thermal expansion of glass.

#### 2.4. Pressure–Volume–Temperature ( $P$ – $V$ – $T$ ) Measurements

$P$ – $V$ – $T$  measurements were made using the constant-volume, constant-mass technique, where a high-pressure stainless-steel cylinder was used as the confinement cell. The cell volume was experimentally determined from the weight of water contained at constant temperature plus a measured volume for connecting fittings. The uncertainty of the measured volume was  $\pm 0.08\%$ . During  $P$ – $V$ – $T$  runs, the cubical expansion coefficient for stainless steel was used to correct the volume of the cell at the measurement temperature.

Isochoric  $P$ – $V$ – $T$  measurements were made by distilling a known quantity of R-134a into the cell, randomly varying the cell temperature by increasing or decreasing increments, and measuring the associated equilibrium pressure. After the isochore determination, the material was distilled back into the tared cylinder and reweighed. The initial and final sample weights agreed within 0.0002 kg. It was estimated that the uncertainties in the  $P$ – $V$ – $T$  measurements were less than 0.3% in specific volume, 0.1% in pressure, and  $0.03^\circ\text{C}$  in temperature.

### 3. RESULTS

#### 3.1. Vapor Pressure

Vapor pressure data encompassed a range of temperature given by  $-62.19$  to  $95.99^\circ\text{C}$ , which is about  $5^\circ\text{C}$  below the critical point ( $|\Delta T^*| \simeq 0.01$ ,  $|\Delta T^*| = 1 - T_r$ , where  $T_r$  is the reduced temperature,  $T/T_c$ ). A total of 32 data points is measured in this range. The vapor pressure data have been correlated using two equations. The first form of the equation follows from Hou and Martin [7] and is similar in form to the vapor pressure equation used for a number of CFCs [8, 9]. This form is used widely in the refrigeration industries and is given by

$$\ln P_{\text{vp}} = A + \frac{B}{T} + CT + DT^2 + \frac{E(F-T)}{T} \ln(F-T) \quad (1)$$

**Table II.** Constants for Vapor Pressure Equation (1)

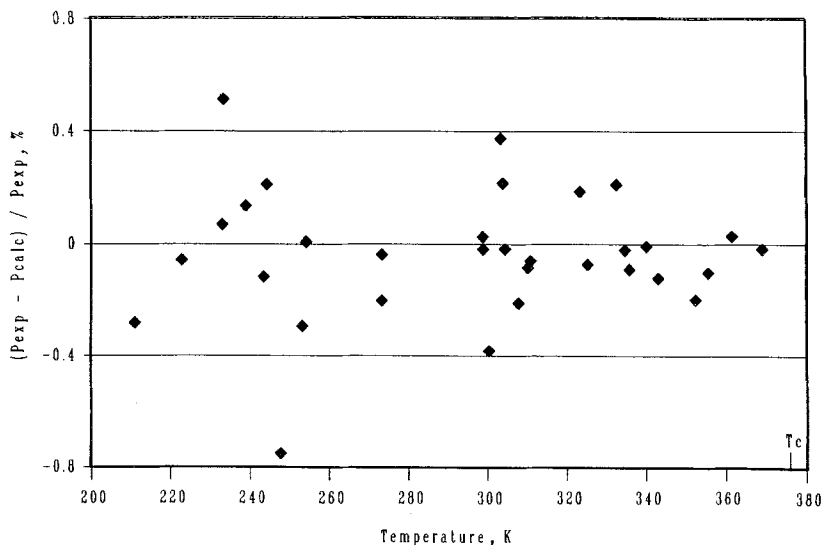
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$$\begin{aligned}
 A &= 24.8033988 \\
 B &= -0.3980408 \times 10^4 \\
 C &= -0.2405332 \times 10^{-1} \\
 D &= 0.2245211 \times 10^{-4} \\
 E &= 0.1995548 \\
 F &= 0.3748473 \times 10^3
 \end{aligned}$$


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Here  $P_{vp}$  is the vapor pressure of R-134a in kPa,  $T$  is in K, and  $A$  through  $F$  are fitting constants shown in Table II. The last term in this equation is an empirical term used to fit the vapor pressure data and this term has been found to be reasonably good in representing the data near the critical point, especially for a large number of refrigerants. A standard deviation of  $\pm 1.37$  kPa in experimental and calculated pressure was obtained using this form of the equation. No systematic deviation of the fit was found. The observed and calculated data are shown in Table III. The deviation plot of the experimental and calculated values using Eq. (1) is shown in Figure 1. The constant  $F$  is constrained to be greater than the critical temperature, therefore no singularity is expected at the critical point.

The second form of the equation used is more scientifically correct. The form of the equation used in this case had the usual analytic part and



**Fig. 1.** Deviation plot of experimental and calculated vapor pressures of R-134a. Vapor pressure is calculated using Eq. (1).

a term proportional to  $|\Delta T^*|^{(2-\alpha)}$  which is expected to give the correct singularity of  $C_v$  near the critical point. This equation is given by

$$P_{vp} = \exp\left(A' + \frac{B'}{T} + C'T + D'T^2\right) + E' |\Delta T^*|^{2-\alpha} \cdot e^{-F' \cdot |\Delta T^*|^2} \quad (2)$$

here  $P_{vp}$  is in kPa,  $T$  is in K,  $A'$  through  $F'$  are fitting constants, and  $\alpha$  is the critical exponent. The values of these constants are presented in

Table III. Vapor Pressure of R-134a

Temperature (°C)	Pressure (kPa)				
	Observed	Calculated [Eq. (1)]	Difference (%)	Calculated [Eq. (2)]	Difference (%)
-62.19	14.13	14.20	-0.495	14.19	-0.425
-50.35	29.30	29.30	0.000	29.39	-0.307
-40.23	51.09	51.09	0.000	51.14	-0.098
-39.93	52.12	51.85	0.518	51.95	0.326
-34.32	68.95	68.88	0.101	68.92	0.044
-29.78	85.56	85.63	-0.082	85.69	-0.156
-28.86	89.63	89.42	0.234	89.46	0.190
-25.42	103.97	104.73	-0.731	104.72	-0.722
-20.00	132.65	133.07	-0.317	132.90	-0.186
-19.05	138.58	138.58	0.000	138.40	0.130
0.00	292.34	292.96	-0.212	292.14	0.068
0.00	292.82	292.96	-0.048	292.14	0.232
25.59	677.41	677.20	0.031	676.83	-0.086
25.68	678.92	679.06	-0.021	678.64	0.041
27.27	708.92	711.61	-0.368	711.34	-0.341
30.08	774.97	772.07	0.374	771.99	0.384
30.69	787.38	785.65	0.220	785.66	0.219
31.23	797.72	797.86	-0.018	797.90	-0.023
34.58	874.94	876.80	-0.213	877.09	-0.246
36.91	934.24	934.99	-0.080	935.52	-0.137
37.57	951.47	952.02	-0.059	952.58	-0.117
50.08	1323.10	1320.62	0.187	1321.54	0.118
51.92	1381.70	1382.67	-0.070	1383.56	-0.134
59.35	1659.56	1656.05	0.212	1656.13	0.207
61.46	1740.23	1740.58	-0.020	1740.34	-0.007
62.66	1788.49	1790.08	-0.090	1789.66	-0.065
66.91	1973.96	1974.10	-0.007	1972.87	0.055
70.04	2115.99	2118.54	-0.121	2116.75	-0.036
79.39	2593.80	2598.90	-0.198	2596.15	-0.091
82.60	2779.27	2782.09	-0.101	2779.49	-0.008
88.48	3145.38	3144.41	0.031	3143.00	0.076
95.99	3663.86	3664.41	-0.016	3664.56	-0.019

**Table IV.** Constants for Vapor Pressure Equation (2)

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$A' = 26.3531$
$B' = -0.378072 \times 10^4$
$C' = -0.3536896 \times 10^{-1}$
$D' = 0.3780304 \times 10^{-4}$
$E' = 0.248828 \times 10^4$
$F' = 66.0$
$\alpha = 0.11$

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Table IV. The vapor pressure data fit this equation as well as Eq. (1). The standard deviation between observed and calculated pressures are found to be  $\pm 1.36$  kPa. In Eq. (2) the last term contains the nonanalytic term with an exponential damping factor [10]. This term can be viewed as an empirical crossover function for the nonanalytic term. The comparison between the experimental and the calculated values using Eq. (2) is also shown in Table III.

### 3.2. Liquid Density

Nine liquid density data points were determined between  $-34.2$  and  $98.5^\circ\text{C}$ . The data were fitted to a standard liquid density correlation [7] given by Eq. (3) below:

$$\rho_L = \rho_C + \sum_{N=1}^4 D_N (1 - T_r)^{N/3} \quad (3)$$

here  $\rho_L$  is the density of the saturated liquid,  $\rho_C$  is the critical density shown in Table I,  $T_r$  is the reduced temperature, and  $D_N$ 's are fitting constants given in Table V; densities are expressed in  $\text{kg} \cdot \text{m}^{-3}$ . In this case, density data were taken within  $3^\circ\text{C}$  ( $|\Delta T^*| \simeq 0.007$ ) of the critical point. A comparison of calculated and observed liquid densities is shown in Table VI. The standard deviation of the fit of the measured data with Eq. (3) was  $\pm 0.5 \text{ kg} \cdot \text{m}^{-3}$ . Figure 2 shows the deviation of the data as a function of temperature. Again, the deviations do not exhibit any systematic trends.

**Table V.** Constants for Liquid Density Equation

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$D_1 = 819.6183$
$D_2 = 1023.582$
$D_3 = -1156.757$
$D_4 = 789.7191$

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Table VI. Liquid Density of R-134a

Temperature (°C)	$10^{-3} \cdot \text{Density}$ ( $\text{kg} \cdot \text{m}^{-3}$ )		Difference (%)
	Observed	Calculated	
-34.21	1.40077	1.40077	0.000
-1.60	1.30031	1.30048	-0.013
27.06	1.19995	1.19953	0.035
50.77	1.09971	1.09969	0.002
69.51	0.99955	1.00035	-0.080
83.65	0.89948	0.89920	0.031
83.66	0.89948	0.89911	0.041
93.05	0.79946	0.79928	0.022
98.48	0.69949	0.69968	-0.027

### 3.3. Pressure-Volume-Temperature Behavior

A Martin-Hou-type equation of state [8] has been used to fit 52  $P$ - $V$ - $T$  data points generated in our laboratory. These  $P$ - $V$ - $T$  data were measured along five isochores at specific volumes of  $1.92 \times 10^{-3}$ ,

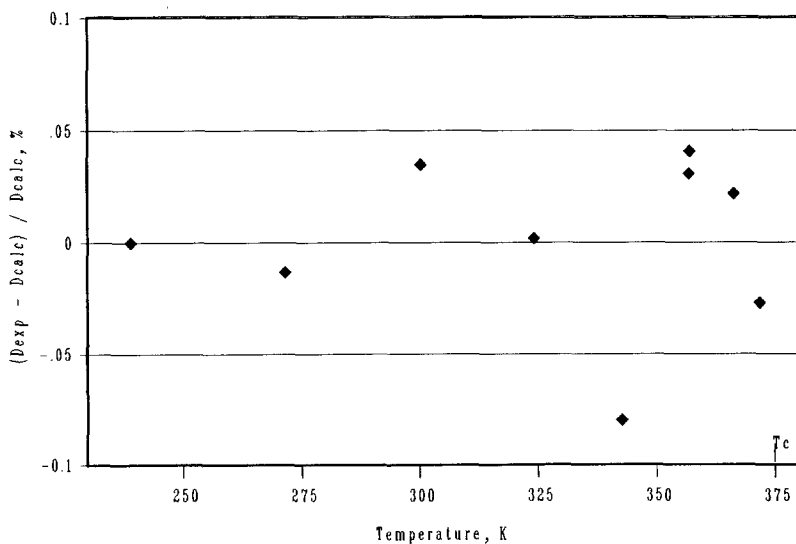


Fig. 2. Deviation plot of experimental and calculated liquid densities of R-134a. Liquid density values are calculated using Eq. (3).



$2.77 \times 10^{-3}$ ,  $3.83 \times 10^{-3}$ ,  $6.38 \times 10^{-3}$ , and  $19.5 \times 10^{-3} \text{ m}^3 \cdot \text{kg}^{-1}$  over a temperature range of 44 to 175°C. The equation of state is given below:

$$P = \frac{RT}{(v-b)} + \sum_{i=2}^5 \frac{A_i + B_i T + C_i \cdot e^{-kT_i}}{(v-b)^i} \quad (4)$$

In Eq. (4),  $P$  is the pressure in kPa,  $v$  is the vapor volume in  $\text{m}^3 \cdot \text{kg}^{-1}$ ,  $R$  is the gas constant,  $k$  is a constant, and  $A_i$ ,  $B_i$ , and  $C_i$  are fitting parameters shown in Table VII. A comparison of a selected set of measured  $P$ - $V$ - $T$  data versus values calculated from Eq. (4) is shown in Table VIII. The complete set of  $P$ - $V$ - $T$  data is given by Wilson and Basu [11]. The data show that within the thermodynamic range investigated, the experimental data are fairly well represented by the form of the Martin-Hou equation used, giving a standard deviation of  $\pm 4.14$  kPa. Figure 3 shows the deviation plot of the data; no systematic trends in the correlation were observed over the thermodynamic range explored.

$P$ - $V$ - $T$  data are also fitted to other equations such as the Soave-Redlich-Kwong (SRK)-type equation and Carnahan-Stirling-deSantis (CSD)-type equation [12] proposed for other fluids by Morrison and McLinden [13]. Of all these equations studied, the Martin-Hou type of equation has been found to fit the  $P$ - $V$ - $T$  data best over the range of temperatures and pressures studied.

### 3.4. Ideal-Gas Heat Capacity

Ideal-gas heat capacities can be calculated from thermodynamic derivations based on statistical mechanical treatments of molecular struc-

Table VII. Constants for Equation of State

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$R = 81.4881629 \times 10^{-3} \text{ kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$
$b = 0.3455467 \times 10^{-3}$
$K = 5.475$
$A_2 = -0.1195051$
$B_2 = 0.1137590 \times 10^{-3}$
$C_2 = -3.531592$
$A_3 = 0.1447797 \times 10^{-3}$
$B_3 = -0.8942552 \times 10^{-7}$
$C_3 = 0.6469248 \times 10^{-2}$
$A_4 = -1.049005 \times 10^{-7}$
$B_4 = 0.0$
$C_4 = 0.0$
$A_5 = -6.953904 \times 10^{-12}$
$B_5 = 1.269806 \times 10^{-13}$
$C_5 = -2.051369 \times 10^{-9}$

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ture and vibrational-rotational assignments. The vibrational fundamentals used for R-134a were previously reported [14]. Using the rigid-rotor, harmonic oscillator approximation, the ideal-gas heat capacities at constant pressure were obtained from the 18 reported fundamental assignments from Harnish and Hirschman [14], which were based on analysis of infrared and Raman spectral data. Ideal-gas heat capacity at constant pressure  $C_p^{(0)}$  was calculated in the range of 125 to 600 K using the following mathematical relationship:

$$C_p^{(0)} = C_{p1} + C_{p2} T + C_{p3} T^2 + C_{p4} T^3 + \frac{C_{p5}}{T} \quad (5)$$

Table VIII.  $P$ - $V$ - $T$  Data of R-134a

$10^3 \cdot \text{volume}$ ( $\text{m}^3 \cdot \text{kg}^{-1}$ )	Temperature (K)	Pressure (kPa)		Difference (%)
		Observed	Calculated (MH Eq.)	
1.9209	373.36	3989.29	3988.60	0.017
1.9212	376.55	4266.46	4264.39	0.048
1.9219	383.44	4852.51	4860.79	-0.171
1.9234	397.90	6114.94	6113.56	0.023
2.7658	373.21	3969.30	3971.37	-0.052
2.7667	379.28	4330.58	4325.76	0.111
2.7676	385.11	4665.67	4661.53	0.089
2.7695	397.94	5379.96	5384.79	-0.090
2.7712	409.55	6019.10	6021.86	-0.046
2.7730	421.62	6672.72	6670.65	0.031
3.8256	373.32	3787.97	3783.83	0.109
3.8282	385.76	4276.11	4267.84	0.193
3.8303	396.39	4674.63	4667.73	0.147
3.8333	410.88	5197.94	5195.18	0.053
3.8365	426.39	5738.49	5741.93	-0.060
3.8409	447.73	6472.09	6468.64	0.053
6.3771	355.03	2682.74	2685.50	-0.103
6.3790	360.42	2802.02	2801.33	0.025
6.3832	372.77	3055.75	3058.50	-0.090
6.3947	406.17	3705.92	3707.99	-0.056
6.4011	424.74	4043.07	4047.90	-0.119
6.4090	447.77	4451.93	4454.69	-0.062
19.4090	317.19	1059.03	1057.65	0.130
19.4340	341.21	1190.72	1191.41	-0.058
19.4620	367.13	1325.86	1326.55	-0.052
19.4890	393.52	1456.17	1458.24	-0.142
19.5180	420.86	1589.93	1589.93	0.000
19.5450	446.83	1709.89	1711.96	-0.121

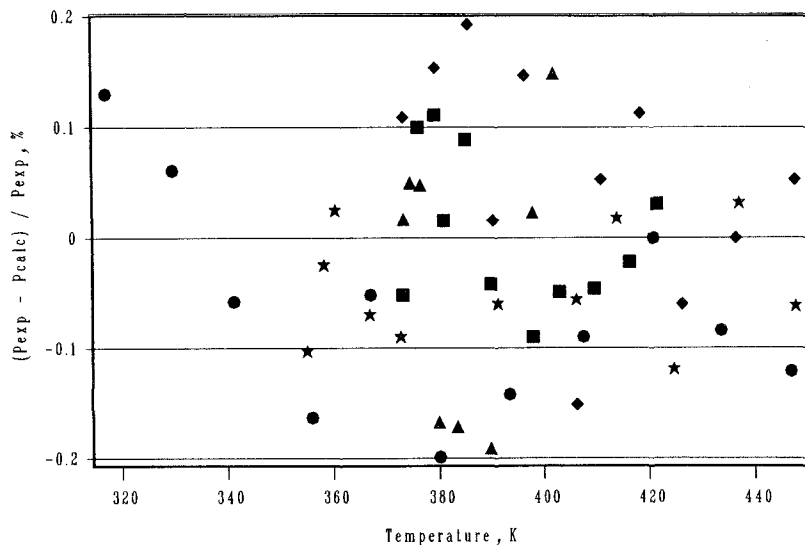


Fig. 3. Deviation plot of R-134a *PVT* data. (▲) Critical isochore; (■) isochore at specific volume  $2.77 \times 10^{-3} \text{ m}^3 \cdot \text{kg}^{-1}$ ; (◆) isochore at specific volume  $3.83 \times 10^{-3} \text{ m}^3 \cdot \text{kg}^{-1}$ ; (★) isochore at specific volume  $6.28 \times 10^{-3} \text{ m}^3 \cdot \text{kg}^{-1}$ ; (●) isochore at specific volume  $19.5 \times 10^{-3} \text{ m}^3 \cdot \text{kg}^{-1}$ .

Uncertainty of fit is estimated to be about  $\pm 0.4\%$ . The constants  $C_{p_i}$ 's are shown in Table IX.  $C_p$  is expressed in  $\text{kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ . The calculated values agree very well (to within 1%) with the work of Chen et al. [15]. The accuracy of vibrational spectra assignments has been a subject of discussion in the past, where significant disagreements in interpreting the data for R-134a have occurred between various laboratories [14, 16–18]. These differences are related to both the symmetry classification of the fundamentals and to proper identification as to which absorption bands are actual fundamentals. This was due to the fact that Raman polarization data for this molecule were not highly definitive.

Table IX. Constants for the Ideal-Gas Specific Heat

$$\begin{aligned}
 C_{p_1} &= -0.5257455 \times 10^{-2} \\
 C_{p_2} &= 0.3296570 \times 10^{-2} \\
 C_{p_3} &= -2.017321 \times 10^{-6} \\
 C_{p_4} &= 0.0 \\
 C_{p_5} &= 15.82170
 \end{aligned}$$

#### 4. DISCUSSION

In this paper we have presented the first set of data on the critical and thermodynamic properties of R-134a. Critical constants are shown in Table I. No other data on critical constants have been found in the open literature for comparison. Measured vapor pressure data are correlated using two equations, both of which fit the data with a reasonable degree of accuracy. Both equations should be considered equivalent to represent the data but one of them has the proper nonanalytic form and should be more accurate near the critical point. Liquid density is fitted to the standard form of equation which has proper singularity near the critical point.  $P$ - $V$ - $T$  data presented here encompass the critical isochore to a density one-tenth of the critical density and a fairly broad range in temperature and pressure. A Martin-Hou-type equation has been found to fit the data better than the CSD or RKS type of equation.

A comparison of the thermophysical data of R-12 and R-134a is shown in Table V of the paper by Sukornick [19] in this issue. It shows that the thermodynamic data compare favorably and R-134a should be a viable alternate to R-12 in refrigeration and other applications. There is a paucity of data on R-134a in the open literature. Additional transport property data, compatibility with materials and water, thermal stability characteristics, and lubricant solution behavior have to be determined before R-134a can be used as a substitute for R-12 in various applications.

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