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Compressed and Saturated Liquid Densities for 18 Halogenated Organic Compounds†

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The pressure−density−temperature $P(\rho,T)$ behavior of 18 liquids that are potential working fluids in thermal machinery has been measured using a vibrating tube densimeter. For each liquid, the data were taken on isotherms spaced at intervals of 5 K to 10 K spanning the temperature range 245 K to 370 K. The pressures ranged from just above the vapor pressure (or the critical pressure) to 6500 kPa. The results of measurements at more than 12 000 thermodynamic points are summarized by correlating functions. Comparison with data from other laboratories indicates that the relative expanded uncertainty in the measured densities is less than 0.05%, except in the critical region. The repeatability of the measured densities is on the order of 0.005%. For each liquid, the $P(\rho,T)$ data were extrapolated to the vapor pressure to obtain the density of the liquid at the vapor pressure. The fluids studied (and their designations by the refrigeration industry) were trichlorofluoromethane (R11), chlorodifluoromethane (R22), 1,1-dichloro-2,2,2-trifluoroethane (R123), 1,2-dichloro-1,2,2-trifluoroethane (R123a), 1-chloro-1,2,2,2-tetrafluoroethane (R124), 1,1,2,2-tetrafluoroethane (R134), 1,1,1,2-tetrafluoroethane (R134a), 1,1-dichloro-1-fluoroethane (R141b), 1,1,1-trifluoroethane (R143), 1,1,2-trifluoroethane (R143a), pentafluorodimethyl ether (E125), 1,1-difluoroethane (R152a), octafluoropropane (R218), 1,1,1,2,3,3,3-heptafluoropropane (R227ea), 2-(difluoromethoxy)-1,1,1-trifluoroethane (E245), 1,1,1,2,2-pentafluoropropane (R245cb), 1,1,1,3,3-pentafluoropropane (R245fa), and propane (R290).

Introduction

In response to the Montreal Protocol and the 1990 Amendments to the Clean Air Act, the Physical and Chemical Properties Division of The National Institute of Standards and Technology (NIST) has been pursuing a program to acquire, correlate, and distribute thermophysical property data for fluids being considered for use in air-conditioning and refrigeration equipment. The present work is a result of this NIST program and it reports measurements of liquid densities as a function of temperature and pressure for 18 potential working fluids.

This manuscript describes the scope of the measurements, the sources and characterization of the fluid samples, the apparatus and procedures used to acquire the data, and comparisons with selected data from other sources to assess the accuracy of the present results.

The present measurements were made at more than 12 000 state points; thus, it was not practical for this manuscript to include a tabulation of all the data. Instead, the results are available in two forms. First, most of the results for each fluid are summarized within their precision by correlating functions $P(\rho,T)$. For convenience, the correlating function chosen is the widely used 32-parameter mBWR (modified Benedict−Webb−Rubin) function (Jacobsen and Stewart, 1973) or a 14-term subset of the mBWR function. However, the present mBWR functions were deduced from $P(\rho,T)$ data spanning a very limited range of conditions; thus, these functions should not be used outside the range of the present data. To emphasize this caveat, we will refer to the present correlating functions as “pseudo-mBWR functions”. Secondly, the original compressed liquid data are also available as Supporting Information and are deposited in the TRC source database.

For all of the 18 liquids, we extrapolated the $P(\rho,T)$ data to the vapor pressure to obtain $\rho_s(T)$, the density of the liquid at the vapor pressure. Because the extrapolations were short, they introduced negligible uncertainties into the functions $\rho_s(T)$. The representations of $\rho_s(T)$ required values of the critical density ρ_c and the critical temperature T_c; these were taken from the literature. We do not recommend using the functions $\rho_s(T)$ within 10 K of the critical point because neither the present representations of our $P(\rho,T)$ data nor the vapor pressure data from the literature are accurate in the immediate vicinity of the critical point.

Scope of the Measurements

The pressure−density−temperature $P(\rho,T)$ data were acquired over a period of 5 years using a vibrating tube densimeter. For each liquid, the data were taken on isotherms spaced at intervals of 5 K to 10 K spanning the temperature range 245 K to 370 K. The pressures on each isotherm ranged from just above the vapor pressure (or the critical pressure) to 6500 kPa.

For brevity, we will refer to the 18 halogenated hydrocarbons studied by the “R numbers” assigned to them by the refrigeration industry. We will refer to the two ethers studied by “E numbers”, as suggested by ASHRAE (1992).

The fluids studied (and, in parentheses, their R or E numbers) were trichlorofluoromethane (R11), chlorodifluoromethane (R22), 1,1-dichloro-2,2,2-trifluoroethane (R123), 1,2-dichloro-1,2,2-trifluoroethane (R123a), 1-chloro-1,2,2,2-tetrafluoroethane (R124), 1,1,2,2-tetrafluoroethane (R134), 1,1,1,2-tetrafluoroethane (R134a), 1,1-dichloro-1-fluoroethane (R141b), 1,1,1-trifluoroethane (R143), 1,1,2-trifluoroethane (R143a), pentafluorodimethyl ether (E125), 1,1-difluoroethane (R152a), octafluoropropane (R218), 1,1,1,-2,3,3,3-heptafluoropropane (R227ea), 2-(difluoromethoxy)-

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† Brand names and commercial sources of materials and instruments, when noted, are given for scientific completeness. Such information does not constitute a recommendation by the National Institute of Standards and Technology nor does it suggest that these products or instruments are the best for the described application.

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Table 1

substance	supplier/purity	source for the critical parameters	source for the vapor pressure
R-11	DuPont/99.9%	T_c Perelshtein (1980) ρ_c Perelshtein (1980)	Altunin et al. (1985)
R-22	Allied Signal/99.9%	T_c JAR (1975) ρ_c JAR (1975)	JAR (1975)
R-123	Halocarbon/99.9%	T_c McLinden et al. (1989) ρ_c McLinden et al. (1989)	McLinden et a. (1989)
R-123a	Allied Signal/99.9%	T_c Chae et al. (1990) ρ_c Chae et al. (1990)	Kubota et al. (1989)
R-124	PCR/99.9%	T_c Schmidt et al. (1995) ρ_c Schmidt et al. (1995)	Weber and Silva (1994)
E-125	3M Chemicals/99.9%	T_c Schmidt et al. (1995) ρ_c Schmidt et al. (1995)	Weber and Defibaugh (1996a)
R-134	PCR/99.9%.	T_c Chae et al. (1990) ρ_c Chae et al. (1990)	Maezawa et al. (1991)
R-134a	DuPont/99.88%	T_c McLinden et al. (1989) ρ_c McLinden et al. (1989)	McLinden et al. (1989)
R-141b	ELF Atochem/99.9%	T_c Chae et al. (1990) ρ_c Chae et al. (1990)	Defibaugh et al. (1993)
R-143	ELF Atochem/99.9%	T_c Schmidt (1996) ρ_c Schmidt (1996)	Van Poolen and Holcomb (1994)
R-143a	ELF Atochem/99.9%	T_c Schmidt et al. (1995) ρ_c Schmidt et al. (1995)	Weber and Defibaugh (1996b)
R-152a	PCR/99.9%	T_c Chae et a. (1990) ρ_c Chae et al. (1990)	Defibaugh and Morrison (1996)
R-218	PCR/99.9%	T_c Schmidt (1996) ρ_c Schmidt (1996)	Brown (1963)
R-227ea	PCR/99.9%	T_c Schmidt (1996) ρ_c Schmidt (1996)	Weber (1996)
E-245	Halocarbon Corp./99.9%	T_c Schmidt et al. (1995) ρ_c Schmidt et al. (1995)	Weber (1996)
R-245cb	PCR/99.9%	T_c Schmidt et al. (1995) ρ_c Schmidt et al. (1995)	Weber and Defibaugh (1996c)
R-245fa	Allied Signal/99.9%	T_c Schmidt (1996) ρ_c Schmidt (1996)	Weber (1996)
R-290	Matheson/99.999%	T_c Younglove and Ely (1987) ρ_c Younglove and Ely (1987)	Younglove and Ely (1987)

1,1,1-trifluoroethane (E245), 1,1,1,2,2-pentafluoropropane (R245cb), 1,1,1,3,3-pentafluoropropane (R245fa), and propane (R290).

Some of our data for R22, R134a, R141b, and R152a have been published previously. Because these fluids have been studied in several other laboratories, our data for these fluids are used here to assess the accuracy of our results.

Sources and Characterization of Fluid Samples

Before use, every fluid sample was analyzed with a gas chromatograph. In most cases, the analysis was consistent with the manufacturer's stated purity. In every case, the sample was used without further purification. The pseudo-mBWR correlating equations require the critical temperature T_c of each fluid. The values of T_c were taken from the literature as noted in Table 1. The representations of $\rho_s(T)$ required values of the critical density ρ_c and values of the vapor pressure as a function of temperature. These too were taken from the literature (see Table 1).

Apparatus and Procedures

The vibrating tube densimeter that was used to measure compressed liquid densities was model DPR412Y manufactured by Anton-PAAR. Below, evidence is provided that the present data from this specific densimeter are accurate to ±0.5 kg/m^3. The densimeter has been thoroughly discussed in an earlier publication, (Defibaugh and Morrison, 1992); a brief description is given here. The vibrating tube together with the manifold containing the fluid sample had an internal volume of 3.5 cm^3. The vibrating tube was made of stainless steel with a wall thickness of 0.3 mm and an inside diameter of 2.4 mm. The densimeter was operated with the electronics package provided by the manufacturer. This package measured the vibration period to seven significant figures.

The temperature of the densimeter was regulated by a thermostated circulating liquid. For temperatures above 273 K a water/ethylene glycol solution was pumped from a thermostated bath through a heat exchanger surrounding the vibrating tube. At temperatures below 273 K, methanol was used in the circulating bath. The heat exchanger surrounding the vibrating tube was enclosed in an insulated, air-filled box. A platinum resistance thermometer monitored the temperature of the liquid exiting the heat exchanger. The temperatures (ITS-90) have a standard uncertainty of ±0.01 K; however, temperature gradients across the densimeter may have exceeded this value at the highest and lowest temperatures. Thermocouples were used to measure temperature gradients in the air bath. We found that the water entering the densimeter was 0.022 K warmer than the water exiting the densimeter at 370 K, the highest temperature at which the apparatus was operated.

The pressure measurements have a standard uncertainty of ±0.5 kPa. A manifold located outside the air bath consisted of mercury reservoirs and a mercury manometer that separated the fluid sample from the pressurized argon. A glass capillary in the manometer allowed us to locate the mercury level in the manometer. The pressures of the argon and the sample were maintained above the vapor pressure of the test fluid at temperatures above ambient. When the vibrating tube was below room temperature, the lower pressure limit was determined by the vapor pressure of the test fluid at the temperature of the manometer. The pressure of the argon was monitored with a quartz pressure transducer, calibrated to a standard uncertainty of ±0.5 kPa.

Table 2

chemical formula	refrig no.	T_{range}/K	P_{range}/KPa	ρ_{range}/kg·m^{-3}	no. of points	T_c/K	ρ_c/kg·m^{-3}	MW	mBW no. of params	RMS/%
CCl_3F	R11	274−370	200−6500	1282−1523	444	471.15	556.1	137.37	14	0.001
$CHClF_2$	R22	263−373	1000−6200	376−1334	162	369.3	513	86.47	32	0.122
$CHCl_2-CF_3$	R123	274−371	200−6500	1292−1535	450	456.94	550	152.93	14	0.001
$CHClF-CClF_2$	R123a	277−367	110−5800	1279−1528	77	461.7	625	152.93	14	0.021
$CHClF-CF_3$	R124	256−370	600−6500	1019−1504	1068	395.62	559.8	136.48	32	0.004
$CF_3-O-CHF_2$	E125	275−370	1500−6200	311−1348	1437	354.49	576.1	136.02	32	0.204
CHF_2-CHF_2	R134	273−367	500−5000	986−1388	59	392.1	539	102.03	14	0.006
CF_3-CH_2F	R134a	243−371	800−6500	756−1407	924	374.3	515.3	102.03	32	0.014
CCl_2F-CH_3	R141b	278−369	105−6100	1084−1280	70	477.3	464.1	116.94	14	0.015
CHF_2-CH_2F	R143	265−373	500−6000	996−1307	171	429.8	469	84.04	14	0.003
CF_3-CH_3	R143a	243−371	1500−6500	341−1035	816	346.75	432.7	84.04	32	0.037
CHF_2-CH_3	R152a	243−372	700−6500	641−1035	1011	386.41	368	66.05	14	0.006
$CHF_2-O-CH_2-CF_3$	E245	273−370	300−6500	1169−1461	855	444.03	515.6	150.05	14	0.005
$CF_3-CF_2-CF_3$	R218	243−373	1200−6500	277−1600	987	345.1	627.1	188.02	32	0.137
$CF_3-CHF-CF_3$	R227ea	243−370	600−6500	983−1611	1014	375.95	580	170.03	14	0.009
$CF_3-CF_2-CH_3$	R245cb	243−372	550−6500	805−1360	1008	380.38	498.8	134.05	14	0.011
$CF_3-CH_2-CHF_2$	R245fa	253−371	500−6500	1103−1467	1041	427.0	517	134.05	14	0.005
$CH_3-CH_2-CH_3$	R290	245−373	1200−6500	274−572	945	369.85	220.5	44.09	32	0.030

Prior to loading the densimeter with the sample fluid, the apparatus was rinsed with ethanol and then acetone to remove any residue from previous experiments. Once the apparatus was filled, compressed liquid densities for the test fluid were measured and recorded by a computer. The temperature and pressure of the densimeter were monitored by the computer and changed after each measurement was completed.

Calibration

The vibrating tube densimeter is not an absolute instrument. We measured the period of vibration of the tube and deduced the density of the test fluids using the calibration function:

$$\rho = \frac{A((\tau/\tau_0)^2 - 1)}{(1 + BT)(1 + CP) + (DTP) + (ET^2P) + (FT^2)} \tag{1}$$

Here τ_0 is the period of the empty vibrating tube and τ is the period when the sample is in the tube. Coefficient A is related to the mean density of the vibrating tube including the empty inner cavity. The coefficients $B-F$ account for the temperature and pressure dependence of the period. The pressure dependence results from the elastic response of the vibrating tube to changing pressure. The temperature dependence results from the thermal expansion of the tube and from the temperature dependence of its elastic constants. The parameters in the calibration function were determined from measuring the temperature dependence of the tube's oscillation frequency when it was evacuated and from the temperature and pressure dependencies of the oscillation frequency when the tube was filled with either water (278 K to 368 K) or propane (245 K to 278 K). The calibration was performed over the pressure range 1.0 MPa to 6.5 MPa. For the calibration, the densities of water were taken from Haar (1984) and the densities of propane were taken from Ely and Huber (1990). The fractional root mean square (rms) deviation of the water data from the calibration was 0.000 09. The fractional rms deviation of the propane data from the calibration was 0.000 13. The most recent calibration (October 1994) resulted in the values

$$A = 4.126\ 575\ 44\ \mathrm{g/cm^3}$$

$$D = -5.424446 \times 10^{-7}\ \mathrm{K^{-1}\ Pa^{-1}}$$

$$B = 7.13672 \times 10^{-5}\ \mathrm{K^{-1}}$$

$$E = 2.312152 \times 10^{-9}\ \mathrm{K^{-2}\ Pa^{-1}}$$

$$C = 2.122165 \times 10^{-4}\ \mathrm{Pa^{-1}}$$

$$F = -1.666759 \times 10^{-7}\ \mathrm{K^{-2}}$$

with τ_0 represented by the expression

$$\tau_0/\mu\mathrm{s} = 6678123 + 1300.782(T/\mathrm{K}) + 0.7726851(T/\mathrm{K})^2$$

Correlation of $P(\rho,T)$ and $\rho_s(T)$

The temperature, pressure, and density ranges for all fluids in this work are listed in Table 2. As mentioned above, we chose the widely-used, modified Benedict−Webb−Rubin equation of state to correlate the compressed liquid density data. The mBWR equation is

$$P = \sum_{n=1}^{9} a_n(T)\rho^n + \mathrm{e}^{-(\rho/\rho_c)^2} \sum_{n=10}^{15} a_n(T)\rho^{2n-17} \tag{2}$$

where P is in kPa, T is in Kelvin, and ρ is in kg·m^{-3}. The temperature dependence of the coefficients is given by

$$a_1 = RT$$

$$a_2 = b_1T + b_2T^{0.5} + b_3 + \frac{b_4}{T} + \frac{b_5}{T^2}$$

$$a_3 = b_6T + b_7 + \frac{b_8}{T} + \frac{b_9}{T^2}$$

$$a_4 = b_{10}T + b_{11} + \frac{b_{12}}{T}$$

$$a_5 = b_{13}$$

$$a_6 = \frac{b_{14}}{T} + \frac{b_{15}}{T^2}$$

$$a_7 = \frac{b_{16}}{T}$$

$$a_8 = \frac{b_{17}}{T} + \frac{b_{18}}{T^2}$$

$$a_9 = \frac{b_{19}}{T^2} \tag{3}$$

$$a_{10} = \frac{b_{20}}{T^2} + \frac{b_{21}}{T^3}$$

$$a_{11} = \frac{b_{22}}{T^2} + \frac{b_{23}}{T^4}$$

$$a_{12} = \frac{b_{24}}{T^2} + \frac{b_{25}}{T^3}$$

$$a_{13} = \frac{b_{26}}{T^2} + \frac{b_{27}}{T^4}$$

$$a_{14} = \frac{b_{28}}{T^2} + \frac{b_{29}}{T^3}$$

$$a_{15} = \frac{b_{30}}{T^2} + \frac{b_{31}}{T^3} + \frac{b_{32}}{T^4}$$

Table 3 lists the coefficients for the mBWR correlation for each fluid.

The density of the liquid at the saturated vapor pressure $\rho_s(T)$ is particularly important in the design of thermal machinery. We represented $\rho_s(T)$ with the function

$$\rho_s/\text{kg}\cdot\text{m}^{-3} = \rho_c(1 + Ar^{1/3} + Br^{2/3} + Cr + Dr^{4/3}) \tag{4}$$

where $r = (T_c - T)/T_c$. The coefficients for this representation for the 18 fluids are listed in Table 4. For each fluid, these coefficients were obtained in the following manner. First, the values of T_c, ρ_c, and the vapor pressure curve $P_s(T)$ were obtained from the literature. Then the pseudo-mBWR functions $P(\rho,T)$ were solved for the values of ρ_s along the vapor pressure curve at the temperatures for which we had taken compressed liquid density data. Finally, the values of ρ_s and r were fitted by eq 4 to obtain values of A, B, C, and D. Table 5 lists all the values of the saturated liquid densities that were used to obtain the coefficients found in Table 4.

In the regions spanned by the present data, the extrapolation of the pseudo-mBWR functions to the vapor pressure curve was very short. Thus, the extrapolation was not sensitive to the specific vapor pressure curve taken from the literature and the resulting values of $\rho_s(T)$ have essentially the same uncertainty (approximately ±0.05%) as the underlying $P(\rho,T)$ data. However, within the last 10 deg of the critical temperature of each fluid, where we have not taken extensive data, eq 4 has not been tested and should not be used.

Accuracy and Precision

Figure 1 displays the temperature and density ranges spanned by the present data for R22, R134a, R141b, R152a, and R290. These fluids have been well-studied in other laboratories; thus, we may compare our results for these fluids with independent measurements to assess the accuracy of our results.

The wide density range spanned by the data in Figure 1 (270 kg/m^3 to 1400 kg/m^3) contrasts sharply with the very narrow ranges of the data used to calibrate the densimeter. Calibrations were done with the densimeter evacuated, filled with water at temperatures in the range 278 K to 368 K (corresponding to 961 kg/m^3 < ρ < 1003 kg/m^3), and filled with propane at temperatures in the range 245 K to 278 K (corresponding to 525 kg/m^3 < ρ < 572 kg/m^3). Thus, it is particularly important to establish the accuracy of the results away from the conditions of the calibration. In comparing data sets, the pseudo-mBWR equations of state are convenient intermediaries. These functions are used as the base line for deviation plots. However, the pseudo-mBWR equations cannot be extrapolated reliably; thus, in those cases where the data sets from the literature extend beyond the range of the present data, the data from the literature outside the range of overlap are not displayed on the deviation plots.

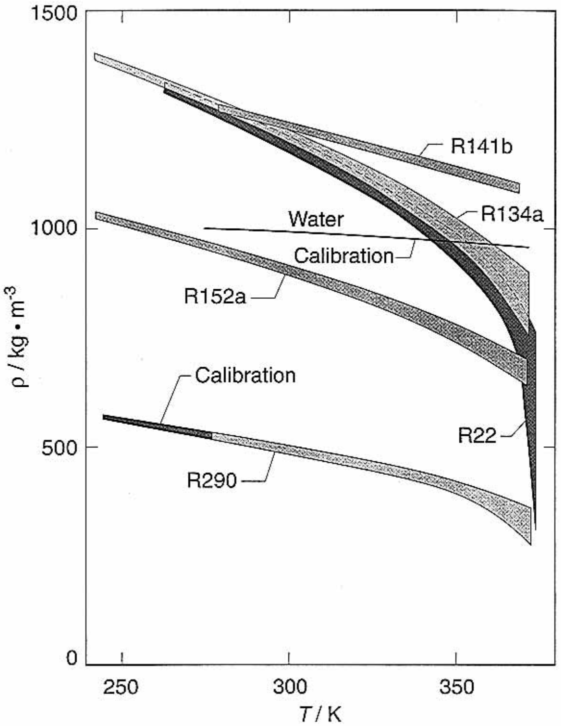

Figure 1. Density−temperature region where compressed liquid densities were measured for six fluids. The figure shows calibrating fluids water above 278 K and propane (R290) below 278 K and the present data for R22, R134a, R152a, R141b, and R290 above 278 K.

The fluid that we studied first and most extensively was R134a. Our most recent densimeter data for R134a span the temperature range from 243 K to 371 K and the pressure range from 800 kPa to 6500 kPa. The densities ranged between 756 kg/m^3 and 1400 kg/m^3. These most recent $P(\rho,T)$ data were correlated with a 32-parameter pseudo-mBWR function. Figure 2 shows the deviation of the 984 data points from the correlation. The fractional root mean square deviation is 0.00017%, or 0.017% of the density.

Figure 2 also displays two previously published data sets for R134a that were collected by Morrison and Ward (1991) using the same vibrating tube densimeter as that used here. The first and the last data sets were taken 3.5 years apart. The present data extend to temperatures as low as 243 K in contrast with the earlier data that were all at 280 K or above. Between the present data and the earlier data, the air bath surrounding the densimeter was greatly improved to reduce temperature gradients and the densimeter was recalibrated using both water and propane. Except in the critical region, deviations of the older R134a data from the more recent data do not exceed 0.1%.

In Figure 3, the R134a data of Tillner-Roth and Baehr

Table 3. mBWR Coefficients

	R11	R123	R123a	R134	R141b
b_{14}	2.59802691207158e−12	1.95338427191828e−12	1.296002851279e−10	2.19715166282643e−12	−3.07263250372138e−11
b_{20}	4.91813268056429e+04	2.86007265915542e+04	5.71932690760094e+05	1.06214020023476e+03	−1.28445914216689e+06
b_{21}	8.89780353160381e+05	1.06863729760879e+06	−9.48336496074027e+06	−2.37020816165854e+05	3.25248530413215e+07
b_{22}	−1.40564548888342e−01	−8.863328904328322e−02	−1.61073751190051e+00	−2.67394811411127e−03	5.05588208276460e+00
b_{23}	−4.52440887656748e+02	−7.781332285821495e+02	−1.87835876855343e+03	1.31107340246379e+02	1.54077371003263e+03
b_{24}	1.58364739537017e−07	1.01294590534417e−07	1.8964409889646e−06	4.38237556253348e−09	−8.39715292976777e−06
b_{25}	7.10199172847341e−07	1.83250394892046e−06	1.79511288208096e−05	−7.04913798333610e−07	−7.24477045836768e−05
b_{26}	−9.15960002889870e−14	−6.41368506265472e−14	−1.18347515739854e−12	2.29067462186431e−15	7.32669340074242e−12
b_{27}	3.24571308974469e−10	5.02527402222898e−10	9.54982649457656e−10	7.98576464462187e−11	−1.85908653340474e−09
b_{28}	2.76521663502760e−20	2.22121734685937e−20	3.91463228503453e−19	9.63363917248841e−22	−3.3410.8387788429e−18
b_{29}	−1.08799694693242e−18	−2.01833400858493e−18	−1.11192274960794e−17	−2.21877996686096e−19	7.56327512517859e−17
b_{30}	−3.18016466044885e−27	−2.85950454894567e−27	−5.58246832581451e−26	7.83510350370720e−29	6.41431343070481e−25
b_{31}	2.04581224270512e−25	4.85288787422495e−25	4.11788401143126e−24	−9.56147418391697e−27	−3.80263781086511e−23
b_{32}	−1.94567037424688e−23	−3.87003034138381e−23	−1.72140262159606e−22	−3.96902733058980e−24	1.09090973979833e−21

	R22	R124	E125	R134a	R143a
b_1	5.99293595624396e−03	2.02195483987853e−02	3.41257775276893e−03	−9.60635512242551e−03	−2.89472915858034e−02
b_2	−1.41895710756076e−01	−7.96289456830664e−01	2.58843483856190e−02	−3.54882780456864e−02	1.15699896438244e+00
b_3	5.01300150685640e−01	5.61855732197923e+00	−4.91781708461723+00	9.05682796999541e+00	−1.19448339350874e+01
b_4	−1.29703166497865e+03	−1.05581570520265e+03	1.64064907501529e+03	−3.26494254245859e+03	−4.42576532322608e+02
b_5	4.83434744225677e+05	−8.75912863827025e+05	−2.00120974374653e+05	−1.65531532300335e+05	1.81953918798157e+05
b_6	9.97706147050999e−07	−1.44302887514260e−05	−3.74825212713009e−06	8.0849644821822e−06	1.54386978141875e−05
b_7	−3.00246461686050e−03	1.964293197824254e−02	3.26815248896427e−03	2.94781852380527e−04	−6.36003826552930e−03
b_8	2.17030857215789e+00	1.51597042613618e+00	−1.256363936804362e−01	−1.415159718416787e−01	1.692422749205681e+00
b_9	−3.96510138021838e+02	−4.494542583239743e+02	2.9745267110792e+02	5.83012937137583e+02	3.07335070181262e+02
b_{10}	1.1646655457644e−09	4.16162281334443e−09	1.3319587293987e−09	−5.42722078945313e−09	−8.4942914640736e−09
b_{11}	1.39030758318177e−06	−5.98148567039040e−06	6.87392484995264e−08	−1.21684940974768e−06	−2.52751772104977e−06
b_{12}	−6.67017210984917e−04	−2.773321862393749e−03	−1.27974691997548e−03	4.15449502348678e−04	−2.61100356191879e−03
b_{13}	−1.62844060714784e−09	−1.89924455108166e−09	−3.07672999214504e−10	1.4894398913476e−09	4.96924752044153e−09
b_{14}	6.36404238896989e−10	1.80031090171851e−09	1.09311339652977e−10	−6.24848231178252e−10	4.47586295951566e−09
b_{15}	−3.03901236996692e−07	3.338189333188340e−07	−7.11410800520614e−08	−1.26688159330256e−08	−4.56775773902390e−07
b_{16}	6.12889167126144e−13	−2.21014305634304e−13	2.81122851294963e−13	5.05619454900261e−14	−4.32187572392042e−12
b_{17}	−4.88708792859445e−16	−1.942936620263946e−16	−1.12229968421366e−16	1.40249481568506e−16	1.17556887183533e−15
b_{18}	2.01614106881277e−13	−1.11167221677931e−13	1.97411311135380e−14	−4.738060461601393e−14	3.39468366109504e−13
b_{19}	−5.74019827994368e−17	2.80440494963943e−17	−3.80962684796229e−18	1.72060768220206e−17	−1.23006362740269e−16
b_{20}	1.04329137654355e+03	−4.90770054482784e+03	−4.56295024872136e+02	1.50059735526743e+03	−1.42548173049122e+02
b_{21}	−4.1962655148324e+05	30.3933166730659e+06	2.87708492947044e+04	8.90968045462685r+04	−1.23382990177305e+05
b_{22}	1.70851899168271e−04	1.23685406673772e−02	−6.56785606122923e−04	−1.48945446529556e−03	−4.32763711184833e−04
b_{23}	−9.83298346494205e−01	−1.55810462725056e+03	1.68599718196297e+01	−1.54045927341391e+02	6.34198963576591e+01
b_{24}	2.50377395536088e−09	−3.136377666954226e−08	−3.33162021579154e−10	−2.4863513411151e−09	1.02189392866185e−08
b_{25}	−9.6803053991829e−07	1.33012482325473e−05	−1.41562463520261e−07	3.2188619204373e−06	−4.77467169450896e−06
b_{26}	2.06628914251366e−15	1.20872889988449e−14	−2.52031398052359e−16	−2.87789855419208e−15	−8.5125917944386e−16
b_{27}	−5.33799514877737e−11	−7.19136676704500e−10	2.20467966013757e−11	−5.26774409834169e−10	7.52140468714866e−10
b_{28}	−1.05125721322578e−21	−4.46952405317974e−21	1.54899390774597e−22	−2.55003969393437e−22	1.96910603137520e−20
b_{29}	1.57449097674197e−19	8.62154122704459e−19	−1.57490934231532e−19	1.50943720684636e−18	−9.51014038072415e−18
b_{30}	3.81790239122965e−27	−4.28193725553803e−28	2.72900464100949e−30	−2.89247266966223e−27	1.12715421500994e−27
b_{31}	−1.78654722159252e−24	1.10301411972382e−24	3.32511426937776e−26	9.7483417327004e−25	3.17080756215014e−24
b_{32}	2.97136739417353e−22	3.18306608785001e−23	−5.37283538480285e−24	−1.85369971146266e−22	−5.31994394748418e−22

	R143	R152a	E245	R227ea	R245cb
b_{14}	4.464339607929153e−12	1.01703404124189e−11	2.42036517848205e−12	6.80081432029707e−13	1.88682781378583e−12
b_{20}	5.3103479032458e+03	−3.75801783918968e+03	1.27701783619188e+04	−8.54381500033543e+02	−8.61391331846689e+02
b_{21}	9.55319185482259e+05	1.04878861317634e+06	2.56428635867820e+06	2.61116769258407e+05	2.53960984745572e+05
b_{22}	−2.73830455708215e−02	1.58168190683583e−02	−5.1478269231755e−02	1.18525176865025e−03	1.74782145012688e−03
b_{23}	−4.55274679992309e+02	−7.92670883899185e+02	−7.81588440405865e+02	−7.5586729273449e+01	−0.8990575346272e+01
b_{24}	4.63826136597111e−08	−3.70022490451931e−08	6.39690298622463e−08	−1.24243874551359e−09	−2.44630088809043e−09
b_{25}	−1.09807541799605e−06	−3.85208083194138e−06	−2.74312070381779e−07	−2.26243563922185e−08	−1.97785043910395e−07
b_{26}	−3.49339932637218e−14	5.92899462487730e−14	−3.7027523714071e−14	7.21055476145966e−16	2.15318338633885e−15
b_{27}	1.35845527334554e−09	7.1969913587838e−09	6.66581091402539e−10	7.54406830189679e−11	2.66438230664774e−10
b_{28}	1.57640535552913e−20	9.03250427144772e−21	1.03176707338059e−20	−7.754799669594698e−23	−2.03377901029119e−23
b_{29}	−5.10566800746815e−18	−5.50489130545338e−17	−1.37368301052988e−18	−2.16057301881082e−19	−1.11772508314657e−18
b_{30}	1.12822598023419e−27	2.75281100082799e−26	−6.45667908918141e−29	8.55834023655390e−29	4.84423355588955e−28
b_{31}	6.02980633018529e−25	2.30664217775455e−23	1.83419257058395e−26	2.272297608630248e−06	2.17307189158206e−25
b_{32}	−1.17791237307706e−22	−2.55717208647218e−21	−3.65170077719492e−23	−3.86699377610803e−24	−2.7084155724471e−23

	R218	R290	R245fa
b_1	−1.34384369175194e−03	1.75140105010012e−01	
b_2	5.35802867203334e−01	−5.73070479098014+00	
b_3	−1.78610039095098e+01	2.36325049217481e+01	
b_4	4.07996591279173e+03	1.77667483292775e+04	
b_5	−4.34602086515198e+05	5.17986148276483e+06	
b_6	−6.17628780522705e−06	−1.36579910783538e−04	
b_7	5.54299407867743e−03	2.10448422993521e−02	
b_8	−9.83406758050058e−01	4.91872596394195e+00	
b_9	1.96447133246926e+02	−2.5620157058025e+03	
b_{10}	4.49170484603634e−10	1.56140367643666e−07	
b_{11}	9.10745267642029e−07	5.12182114714349e−05	
b_{12}	−1.08421929439530e−03	−7.57936930219950e−02	
b_{13}	−3.82503611430890e−10	6.37534762302583e−08	

Table 3. (Continued)

	R218	R290	R245fa
b_{14}	2.1157451905297e−10	−9.73142013992819e−08	2.20260572797671e−12
b_{15}	3.91301257078449e−08	−2.20995856434408e−05	
b_{16}	−4.92272389355912e−14	−3.224491391915073e−11	
b_{17}	9.29382844441493e−18	2.78990730266247e−13	
b_{18}	−1.82701567347095e−14	9.50202989498035e−11	
b_{19}	4.60348581772166e−18	−9.50559871133783e−14	
b_{20}	−2.21893231718545e+02	−4.956432343113264e+04	3.46536031458487e+01
b_{21}	2.12583619782393e+04	−9.92975036648170e+06	9.4039113782514e+05
b_{22}	−2.63079473873963e−04	−1.07282971399773e−01	−4.93086939015568e−03
b_{23}	1.75270565891928e+01	4.61234034319472e+04	−4.35435322354341e+02
b_{24}	−1.31750958449550e−10	−1.10110244730699e−06	7.65658467153751e−09
b_{25}	−4.90790129570993e−09	−3.23791908406416e−03	3.12424885971007e−07
b_{26}	−1.21877313463312e−16	8.80376629460018e−12	−6.54333485725902e−15
b_{27}	1.5378222198406e−11	2.14421553997804e−06	5.39075803197599e−10
b_{28}	8.70846219878236e−23	−6.4761209807067e−17	4.17432473081135e−21
b_{29}	−4.26145255775775e−20	−3.251733112026375e−14	−1.9829213649773e−18
b_{30}	−7.03485105182703e−29	1.06101484487086e−22	−1.93105887373005e−28
b_{31}	4.14520685681596e−26	−2.24400343664914e−20	4.18430358092105e−25
b_{32}	−4.72431129341521e−24	−8.18307840376474e−18	−4.75422511841959e−23

Table 4. Saturated Liquid Density Coefficients for Equation 4

refrigerant	T_c/K	ρ_c/kg/m^3	A	B	C	D	RMS/%
R11	471.15	556.1	1.815711	0.530415	−0.234150	0.631059	0.003
R22	369.3	513	1.887739	0.5985853	−0.071134	0.4032765	0.025
R123	456.94	550	1.974631	0.216241	0.164865	0.446553	0.004
R123a	461.7	625	1.379072	0.003318	1.576401	−0.753463	0.029
R124	395.62	559.8	11.056560	−31.120400	40.175240	−18.186200	0.022
E125	354.49	576.1	2.995996	−5.297620	10.120490	−5.451210	0.058
R134	392.1	539	1.551954	1.743686	−1.564189	0.974518	0.001
R134a	374.3	515.3	1.525758	2.342191	−3.048260	2.127285	0.006
R141b	477.3	464.1	1.967908	−0.229620	1.089664	−0.187400	0.007
R143	429.8	469	2.980305	−3.545429	4.988171	−1.586307	0.007
R143a	346.75	432.7	1.673047	1.106419	−0.204215	0.185364	0.067
R152a	386.41	368	2.228153	−1.022100	2.706948	−1.104830	0.034
E245	444.03	515.6	1.815682	1.449995	−1.839450	1.531503	0.004
R218	345.1	627.1	2.949734	−5.287324	10.215643	−5.655360	0.035
R227ea	375.95	580	1.822471	1.333985	−1.454100	1.211983	0.005
R245cb	380.38	498.8	1.300037	3.492634	−5.089040	3.257889	0.058
R245fa	427.0	517	1.614125	2.154118	−2.864340	1.985377	0.008
R290	369.85	220.5	2.223294	−1.682330	3.454879	−1.442340	0.018

Table 5. Saturated Liquid Densities (kg/m^3)

T/K	R11	R22	R123	R123a	R124	E125	R134	R134a	R141b	R143	R143a	R152a	E245	R218	R227ea	R245cb	R245fa	R290
245						1493.8		1384.2			1114.9	1020.2			1585.3	1339.7		564.4
250						1475.6		1369.0			1100.1	1009.9		1556.2	1568.4	1325.6		558.3
255						1457.2		1353.5			1085.0	999.3		1535.8	1551.2	1311.3	1449.6	552.1
260					1625.7	1438.2		1337.8			1069.5	988.6		1514.6	1533.7	1296.9	1437.3	545.7
265		1308.6			1617.9	1418.7		1321.8		1296.6	1053.5	977.7		1492.8	1515.9	1282.2	1424.9	539.2
270		1292.1			1609.9	1398.6		1305.5		1285.2	1037.0	966.6		1470.1	1497.8	1267.3	1412.3	532.6
275	1529.4	1275.2	1521.4		1601.7	1377.8	1371.1	1289.0		1273.7	1019.9	955.3	1443.6	1446.7	1479.3	1252.1	1399.6	525.8
280	1517.8	1257.9	1509.2	1509.7	1593.4	1356.3	1355.8	1272.0	1267.6	1262.0	1002.2	943.7	1431.2	1422.3	1460.4	1236.7	1386.8	518.8
285	1506.1	1240.1	1496.9	1498.1	1584.9	1334.2	1340.2	1254.7	1258.4	1250.3	983.8	931.9	1418.6	1397.0	1441.0	1220.9	1373.8	511.7
290	1494.3	1221.8	1484.4	1486.4	1576.3	1311.2	1324.2	1236.9	1249.1	1238.4	964.6	919.8	1406.0	1370.6	1421.1	1204.8	1360.6	504.4
295	1482.4	1202.9	1471.9	1474.5	1567.5	1287.3	1307.8	1218.6	1239.7	1226.3	944.6	907.4	1393.1	1343.0	1400.6	1188.2	1347.2	496.8
300	1470.4	1183.5	1459.1	1462.4	1558.6	1262.5	1291.0	1199.8	1230.2	1214.1	923.4	894.7	1380.1	1314.1	1379.6	1171.2	1333.6	489.1
305	1458.2	1163.3	1446.2	1450.1	1549.7	1236.7	1273.7	1180.3	1220.6	1201.7	901.0	881.6	1367.0	1283.7	1357.8	1153.7	1319.8	481.0
310	1445.9	1142.3	1433.2	1437.7	1540.6	1209.6	1255.9	1160.2	1210.8	1189.2	877.2	868.2	1353.6	1251.5	1335.3	1135.6	1305.7	472.7
315	1433.5	1120.4	1419.9	1425.1	1531.5	1181.2	1237.5	1139.2	1201.0	1176.4	851.5	854.3	1340.1	1217.1	1311.8	1116.9	1291.4	464.1
320	1420.8	1097.4	1406.5	1412.2	1522.3	1151.1	1218.5	1117.3	1191.0	1163.5	823.5	840.0	1326.3	1180.0	1287.4	1097.4	1276.8	455.0
325	1408.1	1073.2	1392.8	1399.2	1513.1	1118.9	1198.7	1094.3	1180.8	1150.4	792.4	825.1	1312.3	1139.2	1261.7	1077.0	1261.9	445.6
330	1395.1	1047.4	1379.0	1385.9	1503.9	1084.2	1178.2	1070.1	1170.6	1137.0	757.0	809.6	1298.0	1093.1	1234.7	1055.5	1246.7	435.6
335	1382.0	1019.9	1364.9	1372.4	1494.7	1046.0	1156.7	1044.2	1160.1	1123.4	715.0	793.5	1283.5	1038.1	1206.0	1032.8	1231.1	424.9
340	1368.7	990.0	1350.6	1358.6	1485.5	1002.6	1134.2	1016.4	1149.5	1109.5	660.8	776.5	1268.7	963.7	1175.3	1008.6	1215.0	413.5
345	1355.1	957.1	1336.0	1344.6	1476.3	950.2	1110.4	986.2	1138.7	1095.3		758.7	1253.5		1142.0	982.6	1198.5	401.0
350	1341.4	920.1	1321.1	1330.2	1467.1	877.2	1085.2	952.9	1127.8	1080.8		739.7	1238.0		1105.5	954.3	1181.5	387.1
355	1327.3	877.1	1305.9	1315.6	1457.9		1058.2	915.2	1116.6	1065.9		719.3	1222.1		1064.6	923.0	1163.9	371.1
360	1313.1	823.8	1290.4	1300.7	1448.4		1029.1	871.1	1105.3	1050.7		697.1	1205.8		1017.4	887.7	1145.6	351.6
365	1298.5	748.4	1274.4	1285.4	1438.5		997.2	816.4	1093.7	1034.9		672.5	1189.0		960.2	846.5	1126.6	324.5
370	1283.6		1258.1	1269.7	1427.6		961.7	738.9	1081.8	1018.7		644.6	1171.7		883.6	796.0	1106.7	

(1993), Uematsu et al. (1987), Fukushima et al. 1990, and Piao et al. 1990 are compared with the present data. The data of Tillner-Roth and Baehr agree with the present data to within 0.05% or less between 260 K and 330 K. At 243 K, the lowest temperature, the Tillner-Roth data differ from the present data by 0.1%. The densities of Uematsu et al. are systematically 0.2% smaller than the present densities and those of Tillner-Roth and Baehr. Data of Fukushima et al. 1990 have a positive deviation of 0.1% and the data of Piao et al. (1990) have a negative deviation of 0.1%.

As T_c is approached [for R134a, T_c = 374.3 K (McLinden, 1989)] the deviations of our data from the pseudo-mBWR function increase to 0.15%. This is a symptom of the

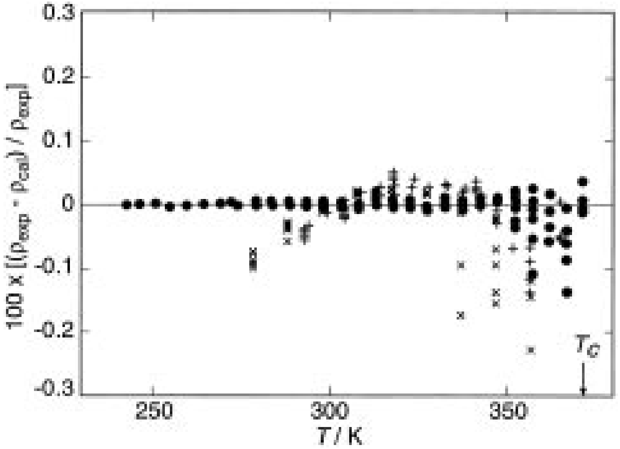

Figure 2. Density deviations from the pseudo-mBWR equation for R134a: (●) this work; (×, +) Morrison and Ward, 1991.

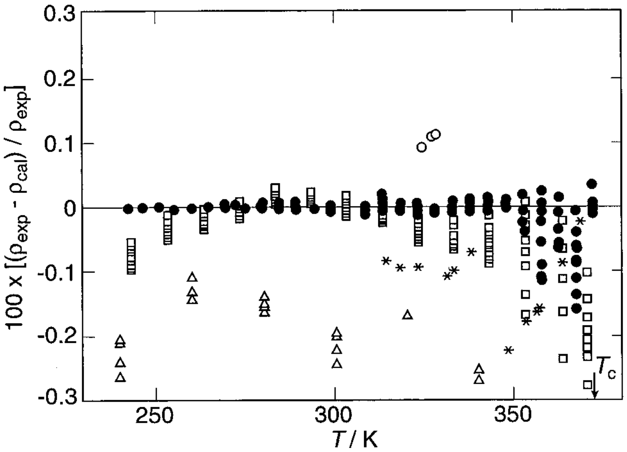

Figure 3. Density deviations from the pseudo-mBWR equation for R134a: (□) data of Tillner-Roth and Baehr 1993; (△) data of Uematsu et al., 1987; (○) data of Fukushima et al., 1990; (*) data of Piao et al., 1990; (●) this work.

inadequacies of the pseudo-mBWR function in the critical region. However, the deviations of the Tillner-Roth and Baehr (1993) data become as large as 0.25% in the same region. A plausible explanation for this additional increase is that the critical temperature of Tillner-Roth and Baehr sample for R134a differed from that of our sample. Differing concentrations of a volatile impurity in either the two sample could explain the density deviations, especially around the critical region. In Figure 4, we explore this possibility by adding 0.5 K to all of the temperatures in the Tillner-Roth and Baehr data. With this temperature shift, the deviations of Tillner-Roth and Baehr's data from the pseudo-mBWR function are comparable to the deviations of our own data throughout the critical region while the deviations at lower temperatures are changed by less than 0.05%. With this shift in temperature, essentially all of Tillner-Roth and Baehr's data fall within 0.05% or our own.

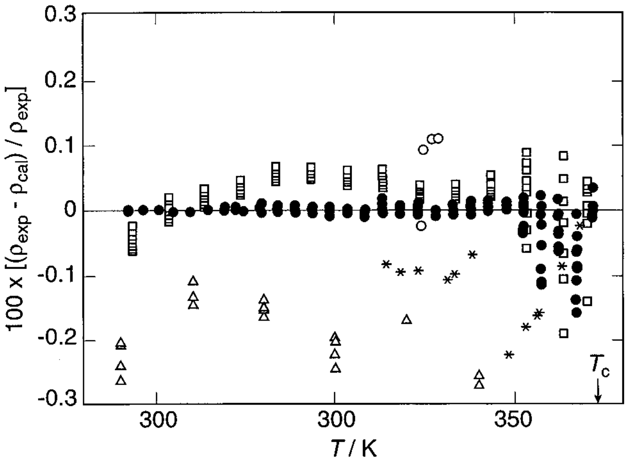

Figure 4. Density deviations of R134a data from the pseudo-mBWR equation: (□) after adding 0.5 K to the data of Tillner-Roth and Baehr, 1993; (△) data of Uematsu et al., 1987; (○) data of Fukushima et al., 1990; (*) data of Piao et al., 1990; (○) this work.

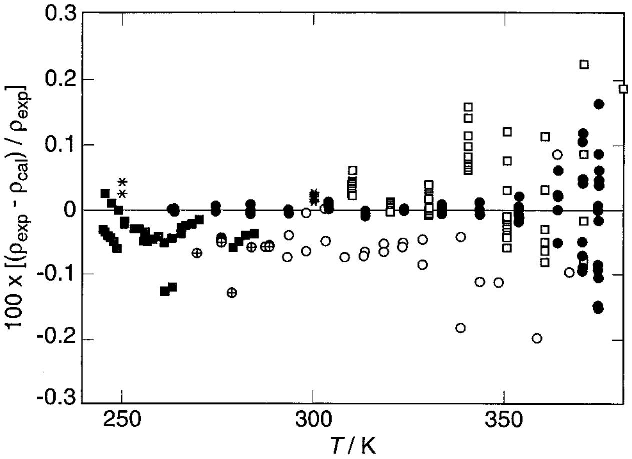

Figure 5. Density deviations of R22 data from the pseudo-mBWR equation: (●) Defibaugh and Morrison, 1992, (○) data of Zander, 1968; (□) data of Uematsu and Fukuizumi, 1991; (⊕) data of Blanke et al., 1988; (■) data of Blanke and Weiss, 1990; (*) data of Haendel et al., 1992.

Figure 5 displays the previously published (Defibaugh and Morrison, 1992) R22 data from this laboratory. They span the wide density range 376 kg/m^3 < ρ < 1334 kg/m^3 and were acquired specifically to test the accuracy of the densimeter at densities significantly different from 1000 kg/m^3. Measurements were made along isotherms at pressures from 1000 kPa to 6200 kPa and temperatures between 263 K and 373 K. The measurements are correlated with a 32-parameter pseudo-mBWR which fit the data with a fractional rms deviation of 0.12%. Below the critical temperature [T_c = 369.3 K (JAR, 1975)], our data agree with Zander's data (1968) well within the uncertainty claimed by Zander (±0.25%). On the average, Zander's densities are lower than ours. Our measurements agree particularly well with the recent measurements of Uematsu and Fukuizumi (1991). Data of Blanke et al. (1988), Blanke and Weiss (1990), and Haendel et al. (1992), all agree to within 0.06% of this work between 240 K and 330 K. Near the critical point (369.3 K) the deviations from the mBWR surface are as large as 2% and fall outside of Figure 5. However, both Zander's and Uematsu and Fukuizumi's data show deviations that are similar in magnitude but opposite in sign. In this region, where small uncertainties in the temperature, pressure, and purity of samples become important, we consider the agreement to be acceptable.

Figure 6 displays our data for R141b together with those of Maezawa et al. (1991), Arnaud et al. (1991), and Matsuo et al. (1994). The base line is a 14-term pseudo-mBWR which is appropriate because all of the data are well away from the critical point. The abbreviated mBWR fit the data with a fractional rms deviation of 0.007%. The data of Maezawa et al. deviate up to 0.1% at the lowest temperature range of this work. Data of Arnaud et al. and Matsuo et al. range between ±0.1% of this work.

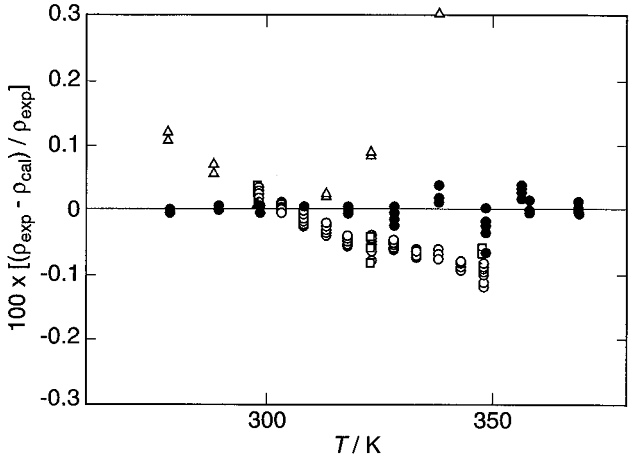

Figure 6. Density deviations of R141b data from the pseudo-mBWR equation: (●) this work; (△) data of Maezawa et al., 1991; (□) data of Arnaud et al., 1991; (○) Matsuo et al., 1994.

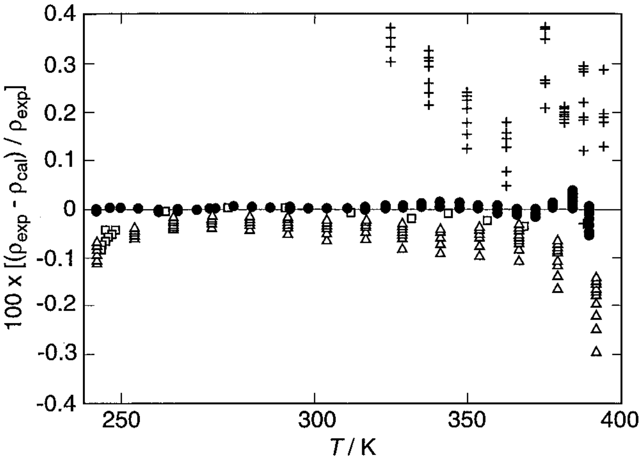

Figure 7. Density deviations of R152a data from the pseudo-mBWR equation: (●) this work; (△) data of Blanke and Weiss, 1992; (□) data of Tillner-Roth and Baehr, 1993a,b; (+) data of Iso and Uematsu, 1989.

Figure 7 displays the present data for R152a together with those of Blanke and Weiss (1992), Tillner-Roth and Baehr (1993), and those of Iso and Uematsu (1989). The deviation of the densities from the mBWR is shown in Figure 7. The compressed liquid data for R152a were correlated with a 14-term mBWR that reproduces the density with a fractional rms deviation of only 0.008% of the density. Remarkably, the data of Blanke and Weiss (1992) and those of Tillner-Roth and Baehr (1993a,b) have a fractional rms deviation of 0.02% and 0.075%, respectively. It is extraordinary for density data from different laboratories to agree so well. In the worst cases, at temperatures below 260 K and near the critical point (T_c = 386.41 K) the deviations approach 0.1% and 0.25%, respectively. The data of Iso and Uematsu (1989) show positive deviations up to 0.35% above 320 K.

Figure 8 displays the present data for propane (R290) together with the data from Ely and Kobayashi (1978), Reamer et al. (1949), and Haynes (1983). A correlating function from the literature (Ely and Huber, 1990), representing the low-temperature (240 K to 278 K) portion of the data, was used in the present calibration. As expected, the present data below 278 K are in good agreement with the data from the literature. Perhaps half of the 0.1% deviations in this low-temperature region are accounted for by the imperfections of the function taken from the literature. In the range 278 K to 360 K, the data from the literature have a fractional rms deviation of only 0.08% from the function that represents our data. In this range, the density of propane is less than 500 kg/m^3, while the density of water used for calibration is near 1000 kg/m^3.

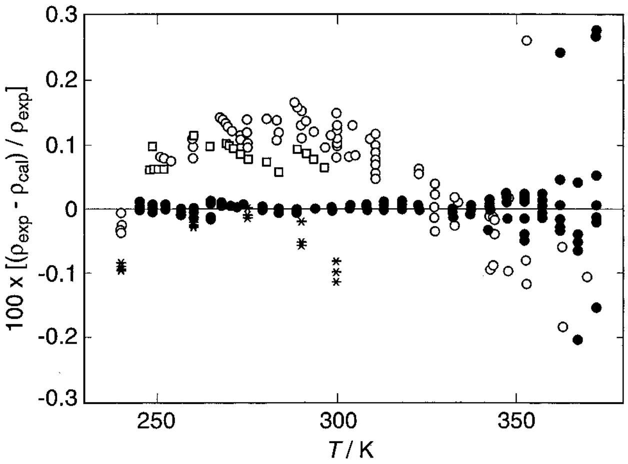

Figure 8. Density deviations of R290 data from the pseudo-mBWR equation: (●) this work; (□) data of Ely and Kobayashi 1978; (○) data of Reamer et al., 1949; (*) data of Haynes, 1983.

In summary, Figures 3–8 demonstrate that for the five fluids R22, R134a, R141b, R152a, and R290, nearly all the present data differ by less than 0.05% of the density from independent measurements in other laboratories. In a few instances, notably at the lowest temperatures, the differences between the present data and those from the literature increase to approximately 0.1%. Near critical points, the comparisons with the data from the literature are inconclusive because the correlating functions do not represent either the present data or the literature data adequately and because samples from different laboratories appear to have different critical temperatures resulting from different concentrations of impurities.

Supporting Information Available:

Tables of data for all 18 fluids are available on the Internet. See any current masthead page for ordering information.

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