Compressed Liquid Density Measurements for 1,1,1,2,3,3,3-Heptafluoropropane (R227ea)

L. Fedele,* F. Pernechele, S. Bobbo, and M. Scattolini

Consiglio Nazionale delle Ricerche, Istituto per le Tecnologie della Costruzione, Corso Stati Uniti 4, I-35127 Padua, Italy

1,1,1,2,3,3,3-Heptafluoropropane (R227ea) is considered an alternative fluid in the refrigeration and air conditioning industry to replace traditional CFC refrigerants. Limited information is available in the literature about the $P\rho T$ behavior of this fluid, so the purpose of the experimental analysis described in the present paper is to provide a set of compressed liquid density data. A vibrating tube densimeter (Anton Paar DMA 512) was used to measure the density of R227ea along six isotherms between (283.15 and 333.15) K and up to 35 MPa, with an estimated uncertainty of about 0.05 %. Saturated liquid densities were extrapolated from the measured data using a polynomial. The results of the compressed liquid density measurements and the extrapolated values of saturated liquid density were correlated with fitting equations and compared to existing experimental data sets.

Introduction

During the past several years, the need to find solutions to environmental questions connected with ozone depletion and the greenhouse effect has led researchers to investigate new possibilities in various fields of industry and HVAC&R (heating, ventilation, air conditioning, and refrigeration) systems. The recent introduction of HFCs as refrigerants, the group to which 1,1,1,2,3,3,3-heptafluoropropane (R227ea) belongs, is particularly due to their zero ozone depletion potential (ODP).

The main characteristics of R227ea allow its use as an alternative to refrigerants R114, R12B1, and R12, probably in blends together with R365mfc in applications with a high condensing temperature that are still under testing. Other applications of this fluid are as a propellant in the production of aerosol sprays and as a blowing agent in the production of rigid polyurethane foams.[1]

The use of a fluid in technical applications requires good knowledge of its thermophysical properties, which must be correlated by accurate equations of state. An extensive database of experimental measurements of properties such as vapor pressure, specific heat, density, viscosity, surface tension, and critical parameters is necessary. Unfortunately, the literature reports only a few works about the measurements of thermophysical properties of R227ea. In particular, vapor pressure measurements were conducted by Shi et al.,[2] by Hu et al.,[3] by Salvi-Narkhede et al.,[4] and by Wang at al.[5] Liu et al.[6] studied liquid viscosity along the saturation line, and Zhang et al.[7] studied the speed of sound and the ideal gas heat capacity at constant pressure. Recently, Fröba et al.[1] completed a work about the thermal diffusivity, sound of speed, viscosity, and surface tension. A limited number of works are available concerning $P\rho T$ behavior of this fluid, in particular, on compressed liquid densities,[4,8−13] on vapor phase densities,[4,10,12−14] and on supercritical phase densities.[10,15] These density data do not completely agree, though it should be very difficult to understand which data set is more reliable than the others. The aim of the present study was to supply accurate compressed and saturated liquid density data along six isotherms between (283.15 and 333.15) K and up to 35 MPa. Measurements were obtained by means of an apparatus based on a vibrating tube densimeter with an estimated density uncertainty of about 0.05 %. The compressed liquid density data were regressed with a Tait equation of state. This correlation was subsequently used for comparison with all the data sets found in the literature.

* Corresponding author. Tel.: +39 049 8295831. Fax: +39 049 8295728. E-mail: laura.fedele@itc.cnr.it.

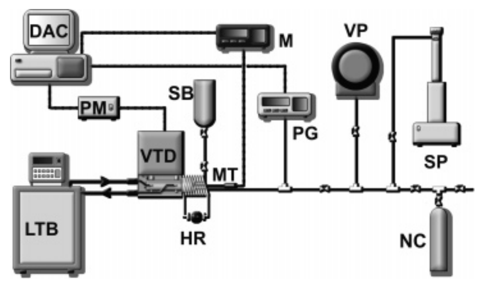

Figure 1. Density measurements apparatus scheme. VTD, vibrating tube densimeter; PM, frequency meter; DAC, data acquisition and control; MT, temperature measurement sensor; M, multimeter; LTB, liquid thermostatic bath; HR, heating resistance; SB, sample bottle; PG, pressure gauge; VP, vacuum pump; SP, syringe pump; NC nitrogen cylinder.

Experimental Section

Materials. R227ea (or HFC-227ea, CAS number 431-89-0) was donated by Solvay with a declared purity > 99.5 %, whereas R32 was supplied by Air Liquide, with a declared purity of 99.5 %. For these fluids, no impurities were detected by gas chromatography using both the flame ionization (FID) and the thermal conductivity (TCD) detectors. The samples underwent several cycles of freezing with liquid nitrogen, evacuation, thawing, and ultrasound and then were used with no further purification.

Experimental Apparatus. The compressed liquid density measurements were performed with an apparatus whose main component is the vibrating tube densimeter (Anton Paar DMA 512). A complete description of the system and the theory used for the calibration of the instrument are given elsewhere;[16−17] here only the most important points and some differences from the original apparatus are outlined. A scheme of this apparatus is shown in Figure 1. The method of measuring density with a vibrating U-tube densimeter is based on the correlation existing

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Table 1. Total Uncertainty in the Density Measurements

uncertainty of pressure measurements in kPa	
accuracy of the pressure gauge	20
stability	3
total	23
uncertainty of temperature measurements in K	
accuracy of thermometer	0.020
stability of the thermostatic bath	0.003
total	0.023
uncertainty of period of oscillation measurements in μs	
variation	0.020
counter accuracy	0.001
total	0.021
uncertainty of the calibration eq 1 in kg·m^{-3}	
uncertainty of the equation of state	0.14
influence of period of oscillation fluctuations under vacuum	0.10
deviation from eq 1	0.10
total	0.34
uncertainty in density calculation in kg·m^{-3}	
uncertainty in pressure measurements	0.20
uncertainty in temperature measurements	0.10
uncertainty in period of oscillation measurements	0.10
uncertainty of the calibration equation	0.34
total	0.74

between the oscillation period (π) of a hollow resonating stainless steel tube and the density of the fluid contained in it, varying temperature T, and pressure P. This correlation is determined during the calibration, evaluating the periods of oscillation of the U-tube under a vacuum, and filled with a fluid of known density in the temperature and pressure range considered in the measurements. The densimeter was connected to an electronic evaluation unit for the measurements of the oscillation period (Anton Paar mPDS 2000) and filled with the sample through a circuit of stainless steel tubes connecting the cell with the refrigerant bottle. The pressure was measured by means of a differential pressure gauge (Druck DPI 145) with a scale up to 35 MPa and an uncertainty of 20 kPa. The temperature of the vibrating tube was controlled with a stability of 0.003 K by a PID control system. Measurements of temperature were obtained by means of a PT 100 Ω resistance thermometer, with an estimated uncertainty within 0.02 K. Considering the full experimental procedure, as indicated in Table 1, the density measurement uncertainty is estimated to be around 0.74 kg·m^{-3}. The fluid was pressurized directly by a syringe pump (Isco Pump, model 260D) connected to the circuit. Dedicated software, developed in the LabView environment, allowed the continuous acquisition, visualization, and elaboration of the main experimental parameters (period, temperature, and pressure).

Experimental Procedure. After purging the circuit by flowing nitrogen and putting it under a vacuum overnight, the sample was charged and the fluid was pressurized by activating the syringe pumps up to about 35 MPa. Then, thermal stabilization was expected, fixing a temperature by means of a thermostatic bath, and after reaching the desired condition, a controlled pressure bleeding, of about (5 to 10) kPa·s^{-1}, was performed by lowering the syringe pump piston, trying to approach a near-static process. Temperature, pressure, and oscillation period were acquired continuously until a discontinuity was observed in the diagram of pressure, revealing the vapor phase formation.

The densimeter was calibrated by measuring the oscillation period of the U-tube under a vacuum and filled with a fluid of known density, in this case water. The measured oscillation period π was correlated to the known density of water, along

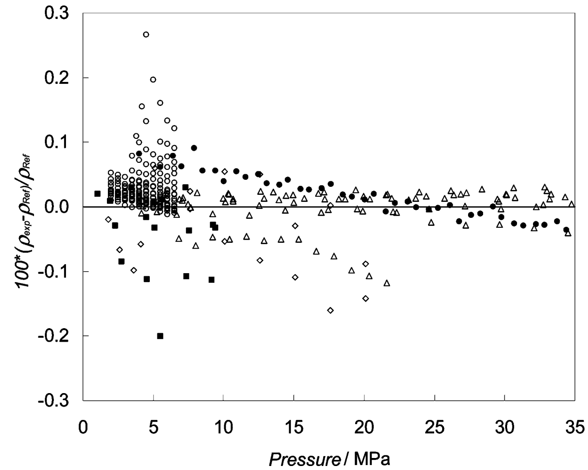

Figure 2. Deviations of the selected compressed liquid density data for R32 from the Refprop 8.0[19] database: ●, experimental; ○, Defibaugh et al.;[21] Δ, Magee;[22] ■, Bouchot and Richon;[23] and ◇, Malbrunot et al.[24]

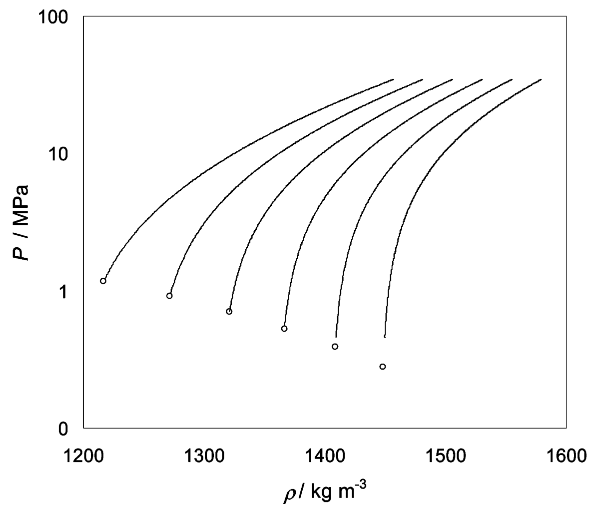

Figure 3. Distribution of −, the experimental compressed liquid density data; and ○, the estimated saturated liquid density data for R227ea.

all the six isotherms considered in the present study. These experimental data were used to regress the a, b, c, and B parameters in the following calibration equation, applied at constant temperature

$$\pi^2 = (aP^2 + bP + c)\rho + B \tag{1}$$

where π is the oscillation period in μs; P is the calibration pressure in kPa; and ρ is the density in kg·m^{-3}. The equation proposed by Wagner and Pruss[18] was used to determine the water density at the calibration pressure and temperature.

To perform a system validation before starting the investigation on R227ea, density measurements for difluoromethane (R32) were performed at two temperatures, (323.15 and 333.15) K. The results were compared to those calculated with the Refprop 8.0 database,[19] using an equation of state[20] for this fluid, which is claimed to have a declared uncertainty of 0.05 % over the experimental temperature and pressure ranges. The deviations between a selected set of experimental data and the values calculated with Refprop are shown in Figure 2. Deviations are within 0.1 % for all the considered data, but for both isotherms, a deviation trend with pressure is evident. For this reason, other data sets already published in the literature[21−24] were considered for the comparison. Not all these data were measured at the same experimental temperature and pressure ranges, and the deviations between them and Refprop seem to be more scattered,

Table 2. Selection of Experimental Compressed Liquid Density Values for R227ea Evenly Sampled from All of the Data Sets

P/MPa	ρ/kg·m^{-3}	P/MPa	ρ/kg·m^{-3}	P/MPa	ρ/kg·m^{-3}	P/MPa	ρ/kg·m^{-3}	P/MPa	ρ/kg·m^{-3}	P/MPa	ρ/kg·m^{-3}
T = 283.15 K				T = 293.15 K				T = 303.15 K			
34.504	1578.9	17.370	1527.0	34.546	1554.6	17.485	1498.2	34.526	1530.1	17.451	1468.7
33.830	1577.3	16.678	1524.6	33.879	1552.6	16.801	1495.6	33.838	1528.0	16.775	1465.7
33.158	1575.5	15.994	1521.8	33.182	1550.8	16.118	1492.8	33.172	1526.0	16.079	1462.6
32.476	1573.6	15.328	1519.4	32.494	1548.7	15.425	1489.9	32.481	1523.9	15.389	1459.5
31.782	1571.7	14.643	1516.8	31.815	1546.8	14.759	1487.1	31.806	1521.7	14.687	1456.0
31.105	1569.9	13.943	1514.1	31.136	1544.7	14.065	1484.2	31.117	1519.6	14.003	1452.9
30.425	1567.9	13.258	1511.5	30.456	1542.8	13.371	1481.1	30.437	1517.3	13.317	1449.6
29.745	1566.1	12.576	1508.7	29.782	1540.6	12.670	1478.0	29.746	1515.0	12.631	1446.1
29.044	1564.1	11.901	1506.1	29.104	1538.5	11.967	1474.9	29.074	1512.8	11.952	1442.7
28.371	1562.0	11.232	1503.2	28.411	1536.5	11.292	1471.8	28.394	1510.6	11.258	1439.1
27.673	1560.0	10.559	1500.4	27.728	1534.3	10.597	1468.5	27.705	1508.1	10.559	1435.3
27.001	1558.1	9.875	1497.5	27.045	1532.2	9.915	1465.1	27.035	1505.9	9.878	1431.5
26.327	1556.3	9.194	1494.6	26.349	1529.9	9.213	1461.8	26.361	1503.5	9.192	1427.6
25.630	1554.1	8.520	1491.6	25.678	1527.7	8.542	1458.3	25.686	1501.0	8.526	1423.9
24.950	1552.0	7.851	1488.5	24.991	1525.5	7.862	1454.8	24.987	1498.7	7.850	1419.8
24.269	1549.9	7.185	1485.5	24.326	1523.2	7.196	1451.2	24.303	1496.1	7.171	1415.7
23.603	1547.8	6.517	1482.3	23.648	1520.9	6.508	1447.5	23.621	1493.5	6.493	1411.3
22.904	1545.6	5.822	1478.8	22.959	1518.6	5.818	1443.6	22.934	1490.9	5.795	1406.8
22.231	1543.5	5.147	1475.5	22.291	1516.2	5.137	1439.6	22.251	1488.4	5.116	1402.4
21.541	1541.2	4.454	1472.0	21.593	1513.7	4.452	1435.7	21.556	1485.5	4.444	1397.6
20.840	1538.9	3.764	1468.5	20.907	1511.2	3.750	1431.3	20.880	1482.9	3.765	1392.7
20.142	1536.6	3.097	1464.7	20.215	1508.7	3.082	1427.2	20.179	1480.1	3.088	1387.7
19.435	1534.2	2.431	1461.1	19.526	1506.1	2.402	1422.6	19.490	1477.3	2.418	1382.4
18.754	1531.8	1.760	1457.3	18.859	1503.7	1.714	1418.1	18.803	1474.4	1.732	1376.9
18.060	1529.4	1.091	1453.4	18.164	1500.9	1.048	1413.5	18.125	1471.6	1.055	1371.1
T = 313.15 K				T = 323.15 K				T = 333.15 K			
34.489	1505.4	17.294	1437.8	34.387	1480.4	17.330	1407.5	34.570	1456.7	17.654	1378.4
33.807	1503.2	16.615	1434.6	33.689	1477.9	16.649	1403.9	33.872	1453.9	16.964	1374.2
33.127	1500.8	15.947	1431.2	33.019	1475.7	15.979	1400.1	33.196	1451.6	16.281	1370.1
32.447	1498.5	15.246	1427.5	32.334	1473.2	15.307	1396.3	32.524	1448.8	15.593	1365.8
31.739	1496.2	14.559	1423.9	31.664	1470.7	14.624	1392.3	31.857	1446.3	14.922	1361.4
31.073	1494.0	13.891	1420.4	30.970	1468.1	13.940	1388.1	31.190	1443.7	14.224	1356.7
30.376	1491.5	13.220	1416.6	30.280	1465.6	13.263	1384.2	30.521	1440.9	13.551	1352.2
29.682	1489.1	12.532	1412.9	29.581	1462.9	12.589	1379.7	29.848	1438.1	12.846	1347.1
28.993	1486.5	11.860	1408.9	28.904	1460.2	11.908	1375.3	29.171	1435.3	12.167	1342.2
28.311	1484.0	11.193	1405.0	28.217	1457.6	11.232	1370.9	28.484	1432.4	11.481	1337.0
27.616	1481.6	10.519	1401.0	27.549	1454.9	10.553	1366.0	27.818	1429.5	10.813	1331.6
26.928	1478.9	9.823	1396.6	26.866	1452.2	9.876	1361.3	27.151	1426.6	10.119	1326.1
26.230	1476.2	9.130	1392.1	26.196	1449.4	9.191	1356.1	26.469	1423.6	9.422	1320.1
25.516	1473.5	8.461	1387.6	25.518	1446.5	8.516	1350.8	25.788	1420.5	8.732	1313.9
24.848	1470.8	7.768	1382.8	24.828	1443.6	7.837	1345.4	25.111	1417.2	8.057	1307.7
24.163	1468.1	7.079	1377.9	24.156	1440.7	7.168	1339.9	24.436	1414.2	7.368	1301.1
23.463	1465.3	6.406	1372.9	23.484	1437.7	6.489	1334.1	23.763	1411.0	6.668	1293.9
22.796	1462.5	5.739	1367.9	22.801	1434.6	5.812	1327.8	23.068	1407.5	5.970	1286.4
22.100	1459.6	5.048	1362.3	22.114	1431.5	5.129	1321.4	22.370	1404.1	5.294	1278.8
21.405	1456.7	4.382	1356.8	21.424	1428.1	4.445	1314.6	21.704	1400.7	4.623	1270.6
20.721	1453.6	3.702	1350.9	20.735	1424.9	3.770	1307.5	21.033	1397.1	3.950	1261.9
20.037	1450.7	3.012	1344.6	20.047	1421.4	3.088	1299.8	20.352	1393.5	3.261	1252.3
19.333	1447.5	2.329	1337.9	19.359	1418.1	2.412	1291.7	19.677	1389.8	2.576	1241.8
18.662	1444.3	1.653	1331.4	18.688	1414.7	1.733	1283.1	19.009	1386.3	1.898	1230.4
17.971	1441.1	0.985	1324.0	17.998	1411.2	1.055	1273.7	18.321	1382.2	1.386	1221.1

without any regular trend with pressure, except for the compressed liquid density data from Defibaugh et al.[21] taken at temperatures lower than 343 K. However, the maximum deviations are higher than those for our measurements, being 0.26 % for Defibaugh, −0.12 % for Magee,[22] −0.20 % for Bouchot and Richon,[23] and −0.16 % for Malbrunot et al.,[24] as indicated in Figure 2.

Results and Discussion

Compressed Liquid Density. The total number of experimental R227ea measured density data is 6615. A selection of these data is summarized in Table 2, and Figure 3 shows the distribution of total measurements on a (ρ,P) plane. All the measured data are available in the Supporting Information on the Web as explained at the end of the paper.

All the experimental data in the temperature range between (283.15 and 333.15) K were correlated with a generalized Tait equation in the form[25]

$$1/\rho = 1/\rho_{sat}\left(1 - c\ \ln\frac{\beta + P}{\beta + P_{sat}}\right) \quad (2)$$

where ρ is the molar density (mol·m^{-3}); ρ_{sat} is the estimated saturated molar density; P is the pressure (kPa); and P_{sat} is the vapor pressure at a given temperature.

Moreover

$$\beta = P_c[-1 + a(1 - T_r)^{1/3} + b(1 - T_r)^{2/3} + d(1 - T_r) + e(1 - T_r)^{4/3}] \quad (3)$$

$$e = \exp(f + g\omega + h\omega^2) \quad (4)$$

$$c = j + k\omega \quad (5)$$

where the reduced temperature $T_r = T/T_c$ and ω is the acentric factor calculated through the basic equation

$$\omega = -\log_{10} P_{rsat} - 1 = 0.357 \quad (6)$$

where P_{rsat} is the reduced vapor pressure at the reduced temperature $T_r = 0.7$. The following values of critical properties were used: T_c = 374.9 K[1], P_c = 2923 kPa; and ρ_c = 590 kg·m^{-3}. The ρ_c value was regressed by fitting a dedicated Helmholtz equation of state (EoS) for R227ea,[26] and P_c was simply calculated from this EoS at T_c and ρ_c.

Table 3. Coefficients of the Generalized Tait Equation for R227ea (Equations 2 to 5)

parameter	numerical value
a	−25.714565
b	167.91085
c	0.08.1102697
d	−366.68485
e	311.05707
f	4.7610868
g	2.2808043
h	1.2801268
j	0.55480357
k	−1.3251606

Table 4. Percent Average Absolute Deviation (AAD %) and Maximum Absolute Deviation ($\Delta\rho_{max}$ %) of Experimental Values for R227ea from Equation 2[a]

	present work	Klomfar et al.[8]	Defibaugh et al.[9]
AAD ($\Delta\rho$ %)	0.01	0.09	0.02
BIAS ($\Delta\rho$ %)	0.00	0.09	−0.01
$\Delta\rho$ %$_{max}$	0.05	0.22	0.04
	Scalabrin et al.[10]	**Ihmels et al.[11]**	**Refprop 8.[19]**
AAD ($\Delta\rho$ %)	0.04	0.26	0.04
BIAS ($\Delta\rho$ %)	0.02	0.26	0.04
$\Delta\rho$ %$_{max}$	0.20	0.56	0.11

[a] $\text{AAD}\,(\Delta\rho\,\%) = 100\cdot\dfrac{\sum_{i=1}^{N_p}\left|\dfrac{\rho_{exptl}-\rho_{calcd}}{\rho_{calcd}}\right|}{N_p}$ where N_p = number of points.

Table 5. Coefficients of Equation 7

A	*B*	*C*	*D*
1.7607	1.2730	−1.1385	0.8747

The *a* to *k* coefficients, regressed from the experimental data, are given in Table 3. Equation 2 represents the data presented here with an absolute average deviation (AAD) of 0.01 % and a maximum deviation of about 0.05 %.

A search in the open literature of publications about compressed liquid density data of R227ea was conducted with the purpose of making a comparison with the data obtained in this work. Equation 2 was used as a reference for comparison with all the experimental data found in the temperature and pressure range of the present regression, and the deviations between experimental and calculated values are plotted in Figure 4 as a function of temperature and summarized in Table 4.

As evident from the figure and table, eq 2 represents data from Defibaugh et al. and Scalabrin et al. with absolute average deviations (AADs) lower than 0.05 %, data from Klomfar et al. with an AAD of 0.09 %, and data from Ihmels et al. with an AAD of 0.26 %. Moreover, the regressed Tait equation was compared with Refprop 8.0, obtaining a good agreement, with an AAD of 0.04 % and a maximum deviation of 0.11 %.

Saturated Liquid Density. The compressed liquid density data were correlated as a function of pressure with a third degree polynomial, for pressures lower than 2.0 MPa. The saturated density values for R227ea were calculated extrapolating this polynomial at the saturated pressures that were determined by means of the Refprop database. This procedure seems to be very reliable because the experimental measurements were taken until the pressure was only (0.1 to 0.3) MPa higher than saturation. The density values at saturation are reported in Table 5 and included in Figure 3 (empty circles).

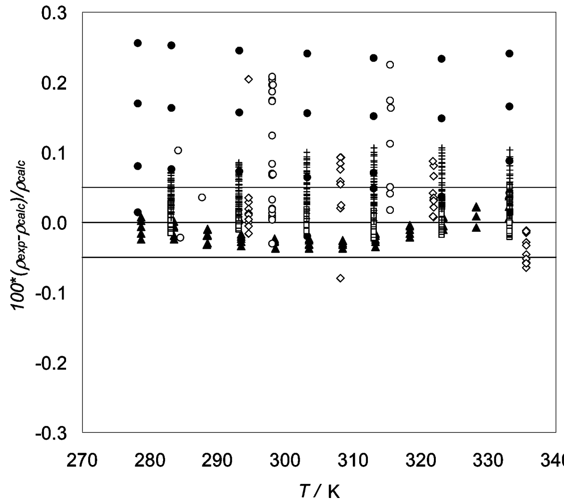

Figure 4. Deviations of all of the available experimental compressed liquid density data for R227ea from eq 2: □, present work; ○, Klomfar et al.;[8] ▲, Defibaugh et al.;[9] ◇, Scalabrin et al.;[10] ●, Ihmels et al.;[11] +, Refprop 8.0.[19]

Table 6. Extrapolated Values of Saturated Liquid Density for the Six Isotherms Analyzed by Means of Equation 7

T / K	*P* / MPa	ρ_{sat} / kg·m^{-3}
283.15	0.2796	1448.2
293.15	0.3891	1408.7
303.15	0.5284	1366.4
313.15	0.7025	1321.0
323.15	0.9164	1271.6
333.15	1.1760	1216.5

Table 7. Comparison between Saturated Liquid Density for R227ea and Values Calculated by Equations Proposed by Defibaugh et al.[9] and by Scalabrin et al.[10] and from Refprop 8.0[19]

	this work	Defibaugh et al.		Scalabrin et al.		Refprop 8.0	
T	ρ_{sat}	ρ_{sat}	$\Delta\rho$	ρ_{sat}	$\Delta\rho$	ρ_{sat}	$\Delta\rho$
K	kg·m^{-3}	kg·m^{-3}	%[a]	kg·m^{-3}	%[a]	kg·m^{-3}	%
283.15	1448.2	1448.2	0.00	1447.5	0.05	1448.2	0.00
293.15	1408.7	1408.3	0.03	1408.0	0.05	1408.4	0.02
303.15	1366.4	1365.9	0.03	1366.0	0.03	1366.2	0.01
313.15	1321.0	1320.6	0.03	1320.9	0.01	1320.9	0.01
323.15	1271.6	1271.4	0.02	1271.7	−0.01	1271.4	0.01
333.15	1216.5	1216.8	−0.03	1217.1	−0.05	1216.5	0.00

[a] $\Delta\rho\,\% = 100\cdot(\rho_{exptl} - \rho_{calcd})/\rho_{calcd}$.

Then, the following general correlation was used to represent the densities at saturation

$$\rho_{sat} = \rho_c(1 + A\tau^{1/3} + B\tau^{2/3} + C\tau + D\tau^{4/3}) \qquad (7)$$

where

$$\tau = 1 - \frac{T}{T_c} = 1 - T_r$$

The parameters of eq 7 are calculated regressing the six data extrapolated from each isotherm, and they are reported in Table 6. Equation 7 is able to estimate the saturated liquid densities of the isotherms of this study with an AAD of 0.004 %. A comparison of these saturated liquid density data was made, in the temperature range considered in the present study, with an analogue equation proposed by Defibaugh et al.[9] and with another proposed by Scalabrin et al.[10] and again with the values provided by Refprop. The results of this comparison are proposed in Table 7.

Conclusions

Compressed liquid density values for 1,1,1,2,3,3,3-heptafluoropropane (R227ea) were measured along six isotherms in the range of (283.15 to 333.15) K with a resonating U-tube densimeter.

To validate the measurements and procedure, an experimental investigation on a limited set of conditions was realized for the fluid R32, comparing the measured data with the literature. The obtained results were satisfactory.

All the experimental data related to R227ea were used to regress the parameters of a Tait equation of state. This equation is able to represent the data of this study with an AAD of 0.01 % and a maximum deviation of 0.05 %; on the basis of this equation, a comparison between the present work and the literature was made, obtaining a good agreement.

An extrapolation of each isotherm allowed the estimation of saturated liquid densities at the vapor pressures determined with Refprop. Subsequently, the values obtained by means of the extrapolation were correlated with a four-parameter equation, able to represent the extrapolated values with an AAD of 0.004 %. Finally, the results obtained by eq 7 were satisfactorily compared to some data available in the literature for the saturated liquid density.

Supporting Information Available:

All of the experimental data. This material is available free of charge via the Internet at http://pubs.acs.org.

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