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PVT Property Measurements for R143a, R125, and R32 in the Gaseous Phase

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PVT property measurements for the hydrofluorocarbons R143a, R125, and R32 are presented in the gaseous phase including the region near saturation. The measurements were performed by means of magnetic suspension densimeter. One hundred and two *PVT* properties for R143a, 144 *PVT* properties for R125, and 101 *PVT* properties for R32 were obtained in the range of temperature from (283 to 323) K and pressures up to 3.2 MPa. The experimental uncertainties are estimated to be 10 mK for temperature, (0.74 to 0.84) kPa for pressure and (0.03 % + 0.005 $kg\cdot m^{-3}$) for density. The sample purities of R143a, R125, and R32 were better than 99.99 %. The measurements are compared with the available data including the values calculated from existing equations of state.

Introduction

For the development of energy conversion systems, accurate thermodynamic properties of the working fluids are required. These properties are calculated from thermodynamic equations of state. To develop the equations of state that represent accurately the whole thermodynamic surfaces, reliable measurements of the thermodynamic properties must be available.

For hydrofluorocarbons, many thermodynamic equations of state have been already reported. They can precisely reproduce the existing experimental data over the entire fluid phase. However, it has been pointed out by Narukawa et al.[1] from our group in 2000 that a large discrepancy, sometimes more than 5 %, exists among the calculated values of the specific heat capacity in the gaseous phase near saturation from existing equations of state developed for hydrofluorocarbons. Our group has also pointed out that the second and third virial coefficients may affect the discrepancy. The third virial coefficients derived from existing equations of state show different behaviors at low temperatures.[2] One of the reasons for these discrepancies is a lack of experimental data near saturation. Only a few measurements of the *PVT* properties have been reported in the region near saturation.

We report precise measurements for the gaseous *PVT* properties of R143a, R125, and R32 in the region near saturation with a magnetic suspension densimeter. On the basis of our measurements, including those in the region near saturation, we show that most of the existing equations of state for R143a, R125, and R32 do not rationally represent our measurements near saturation. There are obviously systematic deviations in the calculation results near saturation from existing equations of state.

Experimental Apparatus

An experimental apparatus consisting of two densimeters with a magnetic suspension balance was used for the measurements. The diagram of the apparatus is shown in Figure 1. The principle of the densimeter was originally given by Klimeck et al.[3] The apparatus was assembled by our group for measuring *PVT* properties of new refrigerants including those near saturation in both the liquid and gaseous phases. This apparatus consists of a temperature control/measurement system, a pressure control/measurement system, and a density measurement system.

A quartz digital pressure gauge (E) covered with a container as shown in Figure 1 was used. The pressure gauge was calibrated by using a dead-weight pressure gauge (W; model 5201, DH Instruments). The temperature was measured by a standard platinum resistance thermometer (L). The temperature values were processed in accordance with ITS-90. The thermometer was installed in the middle between two cells of the A and B densimeters in the thermostatic bath.

The expanded uncertainty of temperature measurements with a coverage factor of 2, having a level of confidence of 95 %, is estimated to be not greater than 10 mK. The expanded uncertainty of the pressure measurements is estimated to be not greater than 0.84 kPa. The expanded uncertainty of the density measurement is estimated to be not greater than (0.03 % + 0.005 $kg\cdot m^{-3}$) in density. The sample purities of R143a, R125, and R32 were better than 99.99 % according to calibration by the manufacturer.

Experimental Results

To confirm the reliability of the apparatus, density measurements for Ar and CO_2 were carried out. We have 107 *PVT* properties of Ar and 59 *PVT* properties of CO_2 in the range of temperature of (283 to 313) K and pressure of (0.4 to 3) MPa. All of our measurements for Ar agree with the reliable equation of state developed of Tegeler et al.[4] within (0.03 % + 0.005 $kg\cdot m^{-3}$), the uncertainty of the density measurements without consideration of the uncertainties for temperature and pressure measurements as shown in Figure 2. The same result was obtained for CO_2.

After confirming the reliability, we performed measurements for R143a, R125, and R32. The *PVT* property measurements

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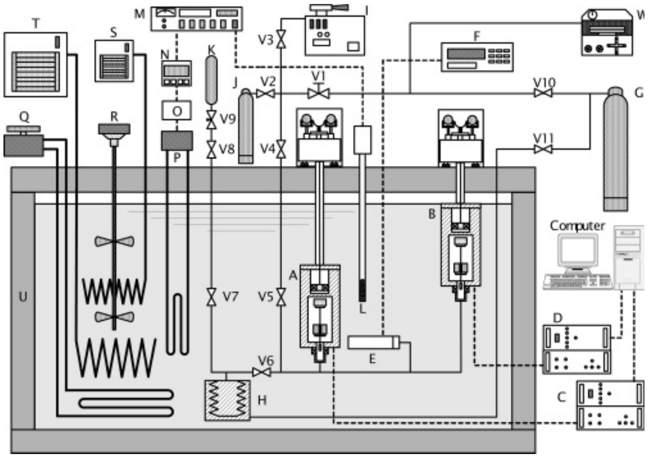

Figure 1. Experimental apparatus: A, densimeter with magnetic suspension balance A; B, densimeter with magnetic suspension balance B; C, control box for (A); D, control box for (B); E, quartz pressure transducer; F, pressure computer; G, nitrogen gas cylinder; H, variable volume vessel with metallic bellows; I, vacuum pump; J, argon gas cylinder; K, sample cylinder; L, standard platinum resistance thermometer; M, thermometer bridge; N, PID controller; O, thyristor regulator; P, subheater; Q, main heater; R, stirrer; S, subcooler; T, main cooler; U, thermostatic bath; V1−V11, valves; W, dead weight pressure-gauge.

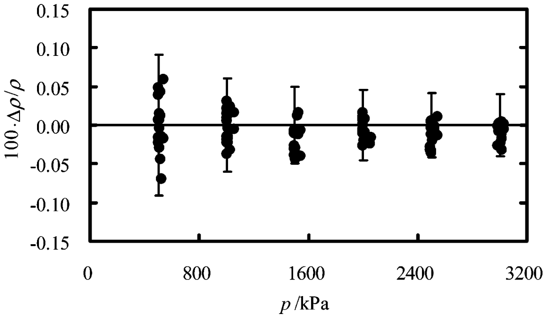

Figure 2. Fractional deviation $\Delta\rho = \{\rho(\text{calcd}) - \rho(\text{exptl})\}$ of the calculated value ρ(calcd) from the equation of state developed by Tegeler et al.[4] from the experimental density ρ(exptl) of Ar at T = (283, 293, 303, and 313) K: baseline, this work with error bars of the expanded uncertainty of (0.03 % + 0.005 kg·m^{-3}); ●, calculated value from equations of state at the state parameters of this work.

Table 1. Experimental *PVT* Properties of R143a

T/K	p/MPa	ρ/kg·m^{-3}	T/K	p/MPa	ρ/kg·m^{-3}
283.15	0.2036	7.588	303.15	0.8016	31.031
283.15	0.2036	7.589	303.15	1.0010	40.662
283.15	0.3999	15.605	303.15	1.0010	40.666
283.15	0.3999	15.605	303.15	1.0999	45.879
283.15	0.5058	20.282	303.15	1.0999	45.880
283.15	0.5058	20.278	303.15	1.1973	51.364
283.15	0.6050	24.932	303.15	1.1973	51.363
283.15	0.6050	24.935	303.15	1.2492	54.458
283.15	0.6516	27.227	303.15	1.2995	57.578
283.15	0.6517	27.238	303.15	1.2995	57.581
283.15	0.7008	29.728	303.15	1.3406	60.231
283.15	0.7008	29.729	303.15	1.3406	60.229
283.15	0.7508	32.361	303.15	1.3698	62.181
283.15	0.7508	32.363	303.15	1.3698	62.183
283.15	0.7815	34.030	303.15	1.4015	64.363
283.15	0.7816	34.039	303.15	1.4015	64.364
283.15	0.8017	35.150	303.15	1.4237	65.925
283.15	0.8018	35.164	303.15	1.4239	65.952
283.15	0.8202	36.196	313.15	0.2015	6.696
283.15	0.8203	36.205	313.15	0.2018	6.718
293.15	0.2017	7.219	313.15	0.2051	6.831
293.15	0.2018	7.222	313.15	0.2051	6.828
293.15	0.4003	14.916	313.15	0.2062	6.869
293.15	0.4003	14.915	313.15	0.4016	13.776
293.15	0.5993	23.356	313.15	0.4017	13.775
293.15	0.5993	23.360	313.15	0.6003	21.275
293.15	0.7013	28.031	313.15	0.6003	21.282
293.15	0.7013	28.028	313.15	0.8003	29.410
293.15	0.7982	32.729	313.15	0.8003	29.413
293.15	0.7982	32.732	313.15	0.9991	38.197
293.15	0.8987	37.925	313.15	0.9991	38.204
293.15	0.8987	37.921	313.15	1.2003	47.988
293.15	0.9510	40.774	313.15	1.2003	48.002
293.15	0.9510	40.774	313.15	1.4002	58.893
293.15	0.9996	43.532	313.15	1.4002	58.909
293.15	0.9996	43.532	313.15	1.5008	64.963
293.15	1.0309	45.365	313.15	1.5008	64.977
293.15	1.0309	45.355	313.15	1.5409	67.443
293.15	1.0487	46.429	313.15	1.5409	67.442
293.15	1.0488	46.429	313.15	1.5980	71.290
293.15	1.0710	47.788	313.15	1.5981	71.312
293.15	1.0711	47.784	313.15	1.6483	74.652
293.15	1.0890	48.896	313.15	1.6483	74.647
293.15	1.0890	48.901	313.15	1.7013	78.498
303.15	0.2000	6.895	313.15	1.7013	78.510
303.15	0.2000	6.903	313.15	1.7408	81.508
303.15	0.4009	14.305	313.15	1.7408	81.517
303.15	0.4009	14.313	313.15	1.7711	83.908
303.15	0.6003	22.265	313.15	1.7711	83.916
303.15	0.6004	22.265	313.15	1.7932	85.730
303.15	0.8016	31.034	313.15	1.7944	85.811

on R143a (102 points), R125 (144 points), and R32 (101 points) were carried out at 4 isotherms for R143a, at 5 isotherms for R125, and at 2 isotherms for R32 in the range of (283 to 323) K at pressures up to 3.2 MPa. The measured data for R143a, R125, and R32 are listed in Tables 1 to 3, respectively. The experimental data distributions for R143a, R125, and R32 are shown on a pressure−temperature plane in Figures 3 to 5, together with those by de Vries.[5] As shown in Figures 3 to 5, our measurements contain many experimental data points in the region near saturation.

Discussion

R143a. Figure 6 shows deviations of the existing *PVT* properties[5−8] including the present measurements from the equation of state developed by Lemmon and Jacobsen,[9] which is an internationally accepted equation for R143a recommended by the International Energy Agency (IEA). Most of our measurements are represented to within ± 0.2 %. The present measurements are compared with four different equations of state in Figure 7, where the baseline is each equation of state. The equations of state are those developed by Lemmon and Jacobsen,[9] Li et al.,[10] Span and Wagner,[11] and Outcalt and McLinden.[12] As shown in Figure 7, the deviations of the present density measurements show systematic trends close to saturation on all isotherms. The deviations of the present data from the equation of state developed by Lemmon and Jacobsen are the smallest, which is also shown in Figure 6, and the other equations of state show a systematic trend along each isotherm from our measurements, especially near saturation.

R125. Figure 8 shows deviations of the existing *PVT* properties[5,13−16] including the present measurements from the equation of state developed by Lemmon and Jacobsen,[17] which is an internationally accepted equation for R125 recommended by the IEA. All of our measurements are represented to within ± 0.2 %. The present measurements are compared with four different equations of state in Figure 9. The baseline is each equation of state, developed by Lemmon and Jacobsen,[17] Sunaga et al.,[18] Piao and Noguchi,[19] and Outcalt and McLinden.[20] As shown in Figure 9, the deviations of our density measurements show systematic trends close to saturation on all isotherms. There are obviously systematic trends along all isotherms near

Table 2. Experimental *PVT* Properties of R125

T/K	p/MPa	ρ/kg·m^{-3}	T/K	p/MPa	ρ/kg·m^{-3}
283.15	0.2072	10.978	293.15	1.2010	77.644
283.15	0.2072	10.980	303.15	0.2251	11.079
283.15	0.4427	24.667	303.15	0.2251	11.067
283.15	0.4427	24.675	303.15	0.4119	20.874
283.15	0.6174	35.920	303.15	0.4119	20.883
283.15	0.6174	35.904	303.15	0.6023	31.549
283.15	0.7107	42.388	303.15	0.6023	31.562
283.15	0.7108	42.398	303.15	0.8001	43.526
283.15	0.7583	45.853	303.15	0.8001	43.535
283.15	0.7583	45.855	303.15	0.9992	56.733
283.15	0.8117	49.877	303.15	0.9993	56.740
283.15	0.8117	49.881	303.15	1.2024	71.762
283.15	0.8280	51.137	303.15	1.2024	71.771
283.15	0.8280	51.141	303.15	1.2992	79.657
283.15	0.8570	53.417	303.15	1.2994	79.674
283.15	0.8570	53.429	303.15	1.4066	89.099
283.15	0.8722	54.640	303.15	1.4067	89.124
283.15	0.8723	54.652	303.15	1.4502	93.212
283.15	0.8800	55.270	303.15	1.4502	93.199
283.15	0.8800	55.283	303.15	1.4812	96.221
283.15	0.8891	56.021	303.15	1.4813	96.231
283.15	0.8892	56.023	303.15	1.5052	98.609
283.15	0.9017	57.048	303.15	1.5052	98.623
283.15	0.9017	57.055	303.15	1.5205	100.175
293.15	0.2059	10.502	303.15	1.5206	100.195
293.15	0.2060	10.492	303.15	1.5373	101.933
293.15	0.4131	21.853	303.15	1.5374	101.917
293.15	0.4132	21.867	303.15	1.5651	104.879
293.15	0.6050	33.264	303.15	1.5658	104.956
293.15	0.6050	33.278	313.15	0.2034	9.623
293.15	0.7997	45.976	313.15	0.2034	9.618
293.15	0.7998	45.993	313.15	0.4045	19.688
293.15	0.9015	53.187	313.15	0.4046	19.685
293.15	0.9015	53.196	313.15	0.6027	30.212
293.15	1.0004	60.669	313.15	0.6027	30.217
293.15	1.0005	60.666	313.15	0.8015	41.496
293.15	1.0515	64.739	313.15	0.8015	41.504
293.15	1.0515	64.718	313.15	1.0020	53.760
293.15	1.1012	68.839	313.15	1.0020	53.773
293.15	1.1013	68.860	313.15	1.1993	66.915
293.15	1.1332	71.588	313.15	1.1993	66.926
293.15	1.1332	71.578	313.15	1.3982	81.578
293.15	1.1496	73.019	313.15	1.3983	81.585
293.15	1.1497	73.016	313.15	1.6044	98.700
293.15	1.1705	74.869	313.15	1.6045	98.710
293.15	1.1706	74.882	313.15	1.7059	108.152
293.15	1.1818	75.887	313.15	1.7059	108.153
293.15	1.1820	75.908	313.15	1.8141	119.180
293.15	1.1921	76.823	313.15	1.8141	119.179
293.15	1.1922	76.841	313.15	1.9007	128.932
293.15	1.2009	77.624	313.15	1.9007	128.938
313.15	1.9519	135.193	323.15	1.5997	90.927
313.15	1.9519	135.196	323.15	1.7989	106.993
313.15	1.9767	138.387	323.15	1.7990	106.996
313.15	1.9767	138.379	323.15	2.0003	125.421
313.15	1.9903	140.173	323.15	2.0003	125.435
313.15	1.9903	140.201	323.15	2.1004	135.708
313.15	2.0018	141.759	323.15	2.1004	135.695
313.15	2.0020	141.763	323.15	2.2014	147.071
323.15	0.1962	8.951	323.15	2.2015	147.082
323.15	0.1962	8.953	323.15	2.3013	159.592
323.15	0.4958	23.505	323.15	2.3013	159.610
323.15	0.4958	23.498	323.15	2.3500	166.311
323.15	0.7981	39.455	323.15	2.3501	166.298
323.15	0.7981	39.458	323.15	2.4002	173.712
323.15	0.9995	50.950	323.15	2.4002	173.740
323.15	0.9996	50.954	323.15	2.4300	178.427
323.15	1.1990	63.156	323.15	2.4301	178.468
323.15	1.1990	63.162	323.15	2.4518	182.037
323.15	1.3990	76.417	323.15	2.4518	182.009
323.15	1.3990	76.411	323.15	2.4693	185.047
323.15	1.5997	90.933	323.15	2.4693	185.021

saturation in the existing equations of state including the internationally accepted equation.

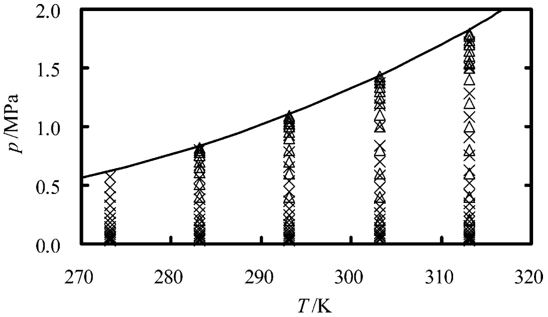

Figure 3. Experimental data distribution of R143a: △, this work; ×, ref 5; the line is the saturation curve calculated from ref 9.

Table 3. Experimental *PVT* Properties of R32

T/K	p/MPa	ρ/kg·m^{-3}	T/K	p/MPa	ρ/kg·m^{-3}
313.15	0.2018	4.120	323.15	0.9983	21.393
313.15	0.2019	4.121	323.15	1.0952	23.732
313.15	0.4908	10.342	323.15	1.2003	26.337
313.15	0.4908	10.336	323.15	1.2004	26.341
313.15	0.7910	17.277	323.15	1.3974	31.424
313.15	0.7910	17.275	323.15	1.3974	31.416
313.15	0.9966	22.355	323.15	1.5906	36.664
313.15	0.9966	22.349	323.15	1.5906	36.670
313.15	1.2033	27.770	323.15	1.7972	42.644
313.15	1.2033	27.759	323.15	1.7972	42.658
313.15	1.3965	33.159	323.15	2.0049	49.091
313.15	1.3965	33.148	323.15	2.0049	49.105
313.15	1.6029	39.331	323.15	2.2006	55.637
313.15	1.6029	39.322	323.15	2.2006	55.653
313.15	1.8018	45.760	323.15	2.4023	62.990
313.15	1.8019	45.748	323.15	2.4023	62.978
313.15	1.9981	52.687	323.15	2.5961	70.738
313.15	1.9981	52.676	323.15	2.5961	70.733
313.15	2.1036	56.702	323.15	2.6990	75.190
313.15	2.1036	56.692	323.15	2.6991	75.207
313.15	2.2015	60.637	323.15	2.7990	79.805
313.15	2.2015	60.629	323.15	2.7990	79.816
313.15	2.2510	62.720	323.15	2.8029	80.005
313.15	2.2510	62.711	323.15	2.8029	80.017
313.15	2.2981	64.762	323.15	2.9006	84.820
313.15	2.2981	64.748	323.15	2.9006	84.839
313.15	2.3557	67.348	323.15	2.9012	84.882
313.15	2.3558	67.343	323.15	2.9012	84.872
313.15	2.4027	69.541	323.15	2.9470	87.258
313.15	2.4030	69.545	323.15	2.9470	87.244
313.15	2.4350	71.084	323.15	2.9603	87.971
313.15	2.4351	71.095	323.15	2.9603	87.990
313.15	2.4505	71.838	323.15	3.0022	90.240
313.15	2.4506	71.855	323.15	3.0023	90.261
313.15	2.4720	72.903	323.15	3.0102	90.716
313.15	2.4720	72.918	323.15	3.0102	90.730
323.15	0.2079	4.101	323.15	3.0411	92.478
323.15	0.2079	4.103	323.15	3.0412	92.495
323.15	0.4992	10.125	323.15	3.0746	94.445
323.15	0.4992	10.143	323.15	3.0746	94.453
323.15	0.4992	10.143	323.15	3.0995	95.942
323.15	0.4992	10.132	323.15	3.0995	95.952
323.15	0.8011	16.793	323.15	3.1255	97.558
323.15	0.8012	16.794	323.15	3.1256	97.549
323.15	0.9983	21.387	323.15	3.1342	98.084

R32. Figure 10 shows deviations of the existing *PVT* properties[5,21−24] including the present measurements from the equation of state developed by Tillner-Roth and Yokozeki,[25] which is an internationally accepted equation for R32 recommended by the IEA. Most of our measurements are represented within ± 0.1 %. The present measurements are precisely compared with three different equations of state in Figure 11. The baseline is each equation of state, developed by Tillner-Roth and Yokozeki,[25] Span and Wagner,[11] and Outcalt and McLinden.[20] As shown in Figure 11, most of the present data are represented without large systematic trend along each

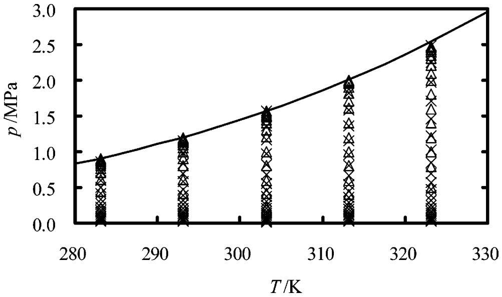

Figure 4. Experimental data distribution of R125: △, this work; ×, ref 5; the line is the saturation curve calculated from ref 17.

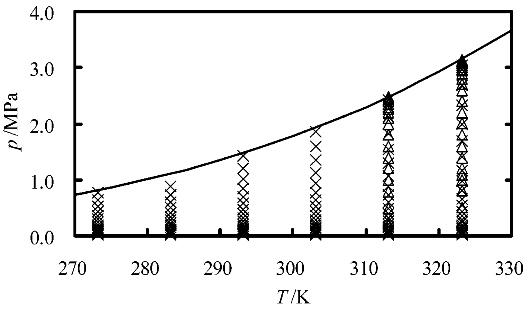

Figure 5. Experimental data distribution of R32: △, this work; ×, ref 5; the line is the saturation curve calculated from ref 25.

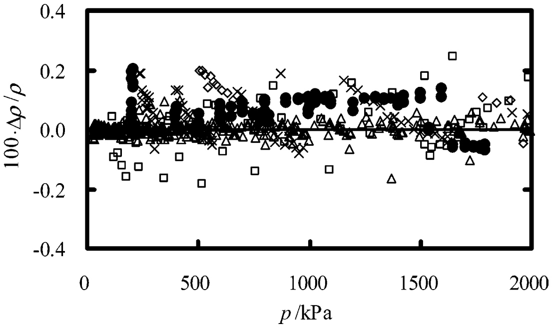

Figure 6. Fractional deviation $\Delta\rho = \{\rho(\text{exptl}) - \rho(\text{calcd})\}$ of the experimental density ρ(exptl) of R143a from values ρ(calcd) calculated from the equation of state developed by Lemmon and Jacobsen:[9] ●, this work; △, ref 5; □, ref 6; ×, ref 7; ◇, ref 8.

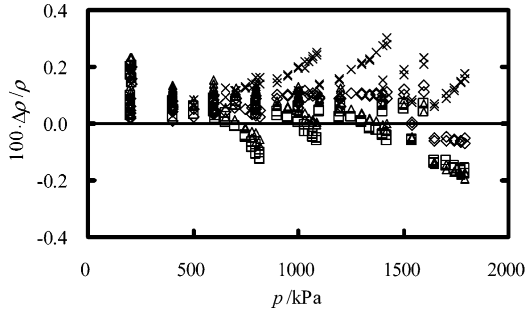

Figure 7. Fractional deviation $\Delta\rho = \{\rho(\text{exptl}) - \rho(\text{calcd})\}$ of the experimental density ρ(exptl) of R143a from values ρ(calcd) calculated from the following four different equations of state: ◇, Lemmon and Jacobsen;[9] □, Li et al.;[10] △, Span and Wagner;[11] ×, Outcalt and McLinden.[12]

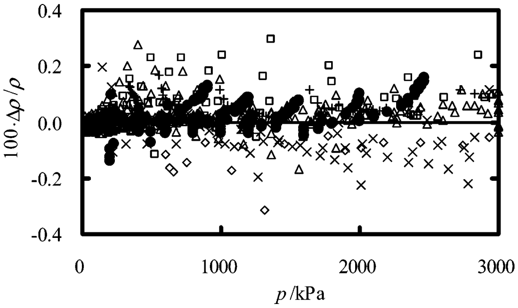

Figure 8. Fractional deviation $\Delta\rho = \{\rho(\text{exptl}) - \rho(\text{calcd})\}$ of the experimental density ρ(exptl) of R125 from values ρ(calcd) calculated from the equation of state developed by Lemmon and Jacobsen:[17] ●, this work; △, ref 5; ×, ref 13; +, ref 14; ◇, ref 15; □, ref 16.

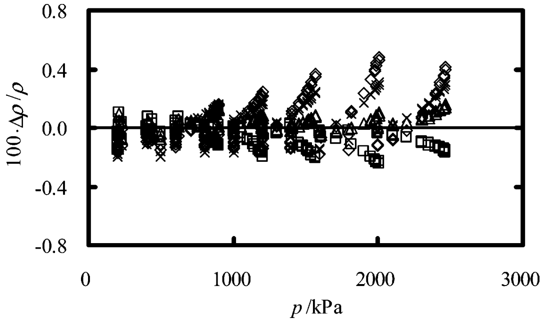

Figure 9. Fractional deviation $\Delta\rho = \{\rho(\text{exptl}) - \rho(\text{calcd})\}$ of the experimental density ρ(exptl) of R125 from values ρ(calcd) calculated from the following four different equations of state: △, Lemmon and Jacobsen;[17] □, Sunaga et al.;[18] ◇, Piao and Noguchi;[19] ×, Outcalt and McLinden.[20]

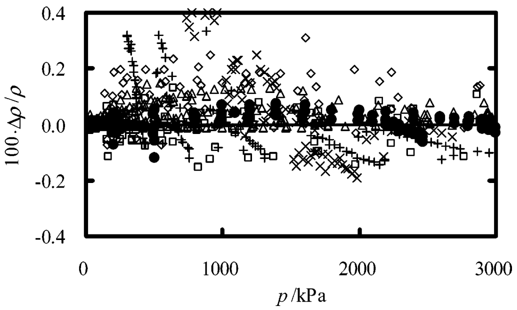

Figure 10. Fractional deviation $\Delta\rho = \{\rho(\text{exptl}) - \rho(\text{calcd})\}$ of the experimental density ρ(exptl) of R32 from values ρ(calcd) calculated from the equation of state developed by Tillner-Roth and Yokozeki:[25] ●, this work; △, ref 5; ♦, ref 21; +, ref 22; ×, ref 23; □, ref 24.

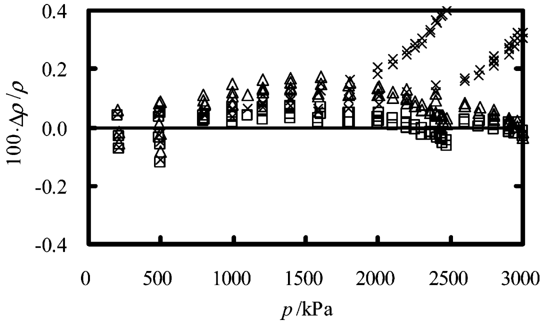

Figure 11. Fractional deviation $\Delta\rho = \{\rho(\text{exptl}) - \rho(\text{calcd})\}$ of the experimental density ρ(exptl) of R32 from values ρ(calcd) calculated from the following three different equations of state: □, Tillner-Roth and Yokozeki;[25] △, Span and Wagner;[11] ×, Outcalt and McLinden.[20]

isotherm near saturation in comparison with the other two substances by existing equations of state except for large systematic trends in the equation of state developed by Outcalt and McLinden for (313 and 323) K isotherms.

Conclusions

PVT properties of R143a, R125, and R32 have been measured in the gaseous phase including near saturation with a magnetic suspension densimeter. One hundred and two *PVT* properties for R143a, 144 *PVT* properties for R125, and 101 *PVT* properties for R32 with an uncertainty of (0.03 % + 0.005 kg·m^{-3}) were obtained. Most of the existing equations of state for R143a, R125, and R32 do not represent the present measurements within experimental uncertainties near saturation. Systematic deviations were found in the region near saturation. The reason could be that they were developed without the experimental data measured near saturation. Measurements of *PVT* properties in the vicinity of saturation in the gaseous phase are important to develop the equation of state.

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