Measurements of Gas-Phase *PVTx* Properties for the Ternary Mixtures Difluoromethane (1) + Pentafluoroethane (2) + 1,1,1-Trifluoroethane (3)

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As one of the promising alternative refrigerants to replace R-502 [R-22 + R-115 ($x_{\mathrm{R-22}} = 0.63$, where x denotes mole fraction)], the gas-phase thermodynamic properties of the ternary mixture R-32 (difluoromethane) (1) + R-125 (pentafluoroethane) (2) + R-143a (1,1,1-trifluoroethane) (3) have been studied. A total of 220 gas-phase *PVTx* properties for the present ternary system, R-32 + R-125 + R-143a, was obtained at four different compositions, ($x_1 = 0.174$, $x_2 = 0.340$), ($x_1 = 0.313$, $x_2 = 0.203$), ($x_1 = 0.343$, $x_2 = 0.445$), and ($x_1 = 0.484$, $x_2 = 0.216$), by using the Burnett apparatus. The present measurements range widely in temperature (305 to 380) K, pressure up to 4.6 MPa, and density up to 200 kg·m^{-3}. The uncertainties of the present study are estimated to be within 7 mK in temperature, (0.8 to 2.5) kPa in pressure, 0.15% in density, and 0.1 mol % in composition. The second and third virial coefficients for the mixtures, B_{m} and C_{m}, were also obtained from the present isothermal *PVTx* data. The consistency of the present measurements was confirmed by comparing them with the virial equation of state for the mixture of interest developed by the present authors and REFPROP. An excellent agreement between the present measurements and those models was confirmed to be within ±0.4% for pressure and ±0.1% for the mixture second virial coefficients.

Introduction

It is strongly recommended to replace remaining HCFCs (hydrochlorofluorocarbons) with zero ozone depletion potential (ODP) refrigerants such as HFCs (hydrofluorocarbons). R-502 [R-22 + R-115 ($x_{\mathrm{R-22}} = 0.630$, where x denotes the mole fraction being used throughout the present paper)] is an azeotropic refrigerant mixture that is being widely used in various commercial refrigeration systems especially for low-temperature application, and its replacement is in progress. Among the several alternative candidates to replace R-502, HFC mixtures such as R-507A [R-125 + R-143a ($x_{\mathrm{R-125}} = 0.412$)], R-404A [R-125 + R-134a + R-143a ($x_{\mathrm{R-125}} = 0.358$, $x_{\mathrm{R-134a}} = 0.604$)], or the ternary R-32 (difluoromethane) (1) + R-125 (pentafluoroethane) (2) + R-143a (1,1,1-trifluoroethane) (3) system have been suggested. However, for the ternary R-32 + R-125 + R-143a system, there are few published reports on its VLE property measurements by Koester et al.[1] and Fujimine et al.[2] and no thermodynamic property study in the single-phase region has ever been reported.

In the present study, therefore, we aim to measure a first set of *PVTx* properties in the gas phase of this important ternary blend. We have performed a series of Burnett measurements at four different compositions of R-32 + R-125 + R-143a [($x_1 = 0.174$, $x_2 = 0.340$), ($x_1 = 0.313$, $x_2 = 0.203$), ($x_1 = 0.343$, $x_2 = 0.445$), and ($x_1 = 0.484$, $x_2 = 0.216$)]. A total of 220 *PVTx* properties was obtained in a wide range of temperatures (305 to 380) K, pressures up to 4.6 MPa, and densities up to 200 kg·m^{-3}.

To examine the validity and reliability of the present measurements, they were compared with available thermodynamic models including REFPROP[3] and a virial equation of state which was developed by the present authors[6] on the basis of *PVT* and *PVTx* property data obtained for the single-component, binary, and ternary systems.

Experimental Section

For the gas-phase *PVTx* property measurements for the ternary R-32 + R-125 + R-143a system, we have employed the Burnett method. The Burnett method enables the densities of the gaseous fluid to be determined with only pressure measurements along a fixed isotherm. Under a constant temperature condition, the sample gas in the cell is expanded to another vessel evacuated in advance. To repeat this isothermal expansions close to the ideal gas state, the compressibility factor at the initial state, Z_0, is calculated by extrapolating $P_n/P_0\mathrm{N}_n$ for n, the repeated number of the expansion, to infinity, where N, the cell constant, denotes the inner volume ratio before and after the expansion. In addition to this expansion procedure at constant temperature, the Burnett−isochoric coupled method originally proposed by Pope et al.[4] and Hall and Eubank[5] was also employed, and this method is known to be effective to obtain numerous gas-phase *PVT* properties in a wide range of temperatures and pressures, not only for pure substances but also for mixtures.

Figure 1 shows a schematic diagram of the present Burnett experimental apparatus. The two cells are the

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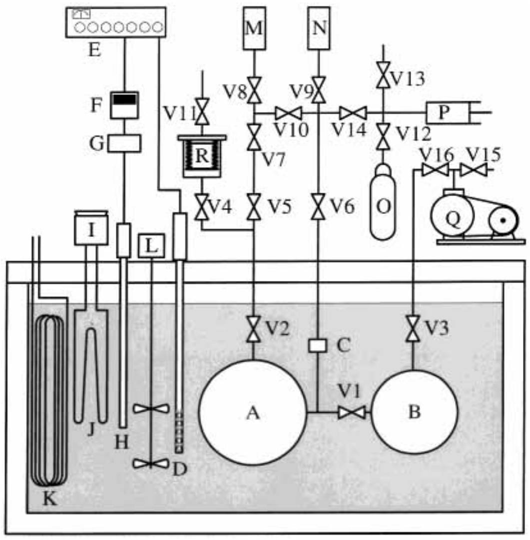

Figure 1. Schematic diagram of the present Burnett apparatus: (A) sample cell; (B) expansion vessel; (C) differential pressure transducer; (D) standard platinum thermometer; (E) precision thermometer bridge; (F) PID controller; (G) thyristor regulator; (H) subheater; (I) slide trance; (J) main heater; (K) water cooler; (L) stirrer, (M, N) quartz pressure transducer; (O) nitrogen gas bomb; (P) hand piston; (Q) vacuum pump; (R) variable volume vessel with metallic bellows; (V1) constant volume valve; (V2−V13) valves.

kernel of the Burnett apparatus, and the sample cell (A) and the expansion vessel (B) are thick-walled spherical vessels made of stainless steel. To avoid any minor pressure deviation after the expansion, these two cells are connected by a constant-volume valve (V1) which is carefully operated. The inner volume ratio of the two cells at zero pressure is called the cell constant, N_∞, and it was determined precisely using gaseous helium with its known density values. In the present study, we have used the value $N_\infty = 1.503\,00 \pm 0.000\,10$[13] for the majority of the present measurements, except those for the composition ($x_1 = 0.343$, $x_2 = 0.445$), for which we have applied the recalibrated value $N_\infty = 1.501\,05 \pm 0.000\,10$, which was a result of a minor change of the inner volume of the sample cell (A) caused by a modification of the pressure measuring system to be discussed later. It should be noted that a possible inner volume change due to thermal expansion and pressure deformation was corrected.

The Burnett apparatus described above is set in a temperature-controlled liquid bath, and the mixture sample inside the cells is assumed to be in thermal equilibrium with the bath fluid (silicone oil). The bath fluid temperature in the vicinity of the sample cell is detected by a standard platinum resistance thermometer (D) and a thermometer bridge (E). The platinum resistance thermometer was calibrated against IPTS-68, and then the resistance values of the thermometer bridge were converted into those of ITS-90. A PID controller (F) associated with the thermometer bridge maintains the fluid temperature uniform and constant with fluctuation less than ±3 mK by controlling the current supplied to the subheater (H).

An indirect pressure measuring system which consists of a differential pressure indicator, DPI (C), within the thermostated bath was employed. The sample pressure is measured by transmitting it to the nitrogen gas pressure through the DPI made of a thin stainless steel diaphragm which separates the mixture sample from the nitrogen gas. We used a DPI of our own design with ±0.3 kPa uncertainty for most of the present measurements, except those for the mixtures with the composition ($x_1 = 0.343$, $x_2 = 0.445$), for which we used a commercially available diaphragm-type differential pressure transducer (DPT) (Validyne, DP-15−38) after a series of careful calibrations. The uncertainty of the differential pressure measurements of the DPT is within ±0.4 kPa. Two quartz pressure transducers (M, N) were used to measure the absolute pressure of nitrogen gas with ±0.1 kPa accuracy.

It is needless to mention that the mole fraction of the sample mixture must remain unchanged throughout the measurements. For this purpose, we have employed the following process. At first, three different component samples were mixed in a sample cylinder on a mass basis. The mixed sample was then transferred to the variable volume vessel with metallic bellows (R), which is cooled by liquefied nitrogen so that all the mixture was charged in to the variable volume vessel. After recovering the vessel to the room-temperature condition, the charged sample outside the bellows was maintained at the compressed liquid state by pressurizing it above the bubble-point pressure by means of the nitrogen gas introduced inside the bellows. Then, the specified quantity of sample was charged into the sample cell little by little by intermittent operation of the valve (V2).

Experimental Results

A total of 220 *PVTx* measurements of the ternary R-32 + R-125 + R-143a system was obtained at four different compositions: 53 measurements along 10 isochores for the pressures (0.14 to 4.5) MPa and temperatures (305 to 380) K for the composition ($x_1 = 0.174$, $x_2 = 0.340$), 52 data points along 9 isochores for the pressures (0.15 to 3.7) MPa at the same temperatures for the ($x_1 = 0.313$, $x_2 = 0.203$) mixture, 58 measurements along 10 isochores for the pressures (0.13 to 4.9) MPa and temperatures (305 to 380) K for ($x_1 = 0.343$, $x_2 = 0.445$), and 57 data points along 10 isochores for the pressures (0.13 to 4.4) MPa at the same temperatures for the ($x_1 = 0.484$, $x_2 = 0.216$) mixture, respectively. These *PVTx* property values are tabulated in Table 1 and plotted in a pressure−temperature diagram of Figure 2, where the vapor pressure curve for each pure component calculated from REFPROP (ver. 6.01)[3] is also included.

An estimation of the experimental uncertainties was made on the basis of the ISO recommendation[14] on the expanded uncertainty with the coverage factor $k = 2$. The estimated temperature uncertainty is not greater than 7 mK. The pressure uncertainty is 0.8 kPa, except for the series for the composition ($x_1 = 0.313$, $x_2 = 0.203$). For this composition, the uncertainty is 2.5 kPa because of the instability of the diaphragm. The density determination uncertainty is 0.15%, whereas the uncertainty of the composition is less than 0.1 mol %. The pure-component samples were of research grade purities, 99.99 mol % for R-32, 99.953 mass % for R-125, and either 99.999 mol % or 99.95 mol % for R-143a, respectively. These purities are those provided by the chemical manufacturers, and samples were used as received.

The experimental values of the mixture second virial coefficient, B_m, and the third virial coefficient, C_m, were determined by fitting eq 1 to the respective isothermal data.

$$(Z - 1)/\rho = B_m + C_m\rho \tag{1}$$

These virial coefficient values are tabulated in Table 2.

Discussion

A comparison of the present measurements with the available thermodynamic model was made. On the basis

Table 1. Experimental *PVTx* Data for the R-32 (1) + R-125 (2) + R-143a (3) System

$x_1 = 0.174$, $x_2 = 0.340$			$x_1 = 0.313$, $x_2 = 0.203$			$x_1 = 0.343$, $x_2 = 0.445$			$x_1 = 0.484$, $x_2 = 0.216$		
T/K	P/kPa	ρ/kg·m^{-3}	T/K	P/kPa	ρ/kg·m^{-3}	T/K	P/kPa	ρ/kg·m^{-3}	T/K	P/kPa	ρ/kg·m^{-3}
380.000	4537.8	2.182	380.000	3751.9	1.602	380.000	4895.3	2.353	380.000	4442.9	1.994
380.000	3451.4	1.452	380.000	2755.7	1.066	380.000	3726.8	1.568	380.000	3312.2	1.327
380.000	2521.1	0.9661	380.000	1959.8	0.7093	380.000	2724.6	1.044	380.000	2381.0	0.8826
380.000	1787.2	0.6428	380.000	1363.3	0.4720	380.000	1933.6	0.6958	380.000	1669.2	0.5873
380.000	1240.9	0.4277	380.000	934.5	0.3140	380.000	1343.4	0.4635	380.000	1150.2	0.3907
380.000	849.5	0.2846	380.000	634.2	0.2089	380.000	921.1	0.3088	380.000	783.5	0.2600
380.000	576.0	0.1893	380.000	427.6	0.1390	380.000	625.5	0.2057	380.000	529.5	0.1730
380.000	388.1	0.1260	380.000	287.1	0.09249	380.000	422.1	0.1371	380.000	356.0	0.1151
380.000	260.4	0.0838	380.000	192.1	0.06154	380.000	283.5	0.09131	380.000	238.5	0.07657
380.000	174.3	0.05576	380.000	128.4	0.04094	380.000	190.0	0.06083	380.000	159.5	0.05094
380.000	116.4	0.03710	365.000	3468.4	1.604	380.000	127.0	0.04052	365.000	4073.2	1.995
365.000	4110.0	2.184	365.000	2581.1	1.067	365.000	4430.3	2.352	365.000	3089.8	1.328
365.000	3196.0	1.453	365.000	1852.1	0.7099	365.000	3448.9	1.567	365.000	2245.4	0.8833
365.000	2367.8	0.9669	365.000	1296.1	0.4723	365.000	2556.4	1.044	365.000	1584.9	0.5877
365.000	1692.8	0.6433	365.000	891.8	0.3143	365.000	1829.8	0.6955	365.000	1096.9	0.3910
365.000	1181.4	0.4280	365.000	606.8	0.2091	365.000	1278.6	0.4634	365.000	749.2	0.2602
365.000	811.3	0.2848	365.000	409.8	0.1391	365.000	879.3	0.3087	365.000	507.5	0.1731
365.000	551.3	0.1895	365.000	275.4	0.09256	365.000	598.4	0.2057	365.000	341.5	0.1152
365.000	371.9	0.1261	365.000	184.4	0.06158	365.000	404.3	0.1370	365.000	229.1	0.07663
365.000	249.8	0.08388	350.000	3174.9	1.607	365.000	271.8	0.09128	365.000	153.2	0.05098
365.000	167.2	0.05581	350.000	2401.9	1.069	365.000	182.2	0.06081	350.000	3694.1	1.997
350.000	3664.2	2.181	350.000	1742.6	0.7113	350.000	3948.4	2.354	350.000	2861.8	1.329
350.000	2930.6	1.4563	350.000	1228.1	0.4733	350.000	3162.7	1.568	350.000	2106.5	0.8839
350.000	2208.5	0.969	350.000	848.9	0.3149	350.000	2384.2	1.045	350.000	1498.9	0.5881
350.000	1595.9	0.6447	350.000	579.4	0.2095	350.000	1724.2	0.696	350.000	1042.7	0.3913
350.000	1121.2	0.4289	350.000	392.0	0.1394	350.000	1212.5	0.4636	350.000	714.4	0.2604
350.000	773.3	0.2854	350.000	263.8	0.09275	350.000	837.1	0.3089	350.000	484.9	0.1732
350.000	526.9	0.1899	350.000	176.8	0.06171	350.000	571.1	0.2058	350.000	326.9	0.1153
350.000	356.2	0.1263	335.000	2863.1	1.606	350.000	386.5	0.1371	350.000	219.4	0.07668
350.000	239.5	0.08406	335.000	2216.0	1.069	350.000	260.2	0.09133	350.000	146.9	0.05102
350.000	160.4	0.05593	335.000	1631.3	0.7111	350.000	174.6	0.06085	335.000	3289.9	1.998
335.000	2655.9	1.454	335.000	1159.6	0.4731	335.000	3440.6	2.354	335.000	2625.7	1.330
335.000	2042.1	0.9673	335.000	805.5	0.3148	335.000	2854.9	1.568	335.000	1964.0	0.8846
335.000	1494.9	0.6436	335.000	551.3	0.2095	335.000	2206.8	1.045	335.000	1411.1	0.5886
335.000	1058.4	0.4282	335.000	373.7	0.1394	335.000	1616.0	0.6960	335.000	987.4	0.3916
335.000	733.3	0.2849	335.000	251.8	0.09272	335.000	1145.5	0.4637	335.000	679.6	0.2605
335.000	501.0	0.1896	335.000	168.9	0.06169	335.000	794.7	0.3089	335.000	462.3	0.1733
335.000	339.2	0.1261	320.000	2029.1	1.069	335.000	543.8	0.2058	335.000	312.2	0.1153
335.000	228.3	0.08392	320.000	1516.5	0.7112	335.000	368.7	0.1371	335.000	209.7	0.07674
335.000	153.1	0.05583	320.000	1089.1	0.4732	335.000	248.5	0.09134	335.000	140.4	0.05106
320.000	1870.0	0.9687	320.000	761.5	0.3148	335.000	166.9	0.06085	320.000	2372.8	1.331
320.000	1391.9	0.6442	320.000	523.3	0.2095	320.000	2543.6	1.570	320.000	1817.1	0.8852
320.000	995.2	0.4286	320.000	355.6	0.1394	320.000	2022.4	1.046	320.000	1321.0	0.5890
320.000	693.7	0.2852	320.000	240.0	0.09272	320.000	1505.0	0.6967	320.000	931.1	0.3919
320.000	475.7	0.1897	320.000	161.1	0.06169	320.000	1076.9	0.4641	320.000	643.8	0.2607
320.000	322.9	0.1262	305.000	1399.3	0.7119	320.000	751.5	0.3092	320.000	439.3	0.1735
320.000	217.6	0.08399	305.000	1017.5	0.4737	320.000	516.0	0.2060	320.000	297.0	0.1154
320.000	146.1	0.05588	305.000	716.9	0.3151	320.000	350.8	0.1372	320.000	199.9	0.07679
305.000	927.4	0.4279	305.000	495.0	0.2097	320.000	236.8	0.09142	320.000	134.0	0.05109
305.000	650.9	0.2847	305.000	337.4	0.1395	320.000	159.2	0.06091	305.000	1662.5	0.8858
305.000	448.4	0.1894	305.000	228.1	0.09282	310.000	1893.7	1.048	305.000	1228.2	0.5894
305.000	206.4	0.08385	305.000	153.4	0.06176	305.000	1389.7	0.6965	305.000	873.8	0.3921
305.000	138.7	0.05579				305.000	1006.3	0.4643	305.000	607.6	0.2609
						305.000	707.6	0.3093	305.000	416.1	0.1736
						305.000	488.2	0.2061	305.000	282.2	0.1155
						305.000	332.6	0.1373	305.000	190.0	0.07684
						305.000	224.9	0.09147	305.000	127.6	0.05113
						305.000	151.3	0.06093			

of the present *PVTx* property measurements for ternary compositions of R-32 + R-125 + R-143a, together with those for each binary mixture, R-32 + R-125,[7] R-125 + R-143a,[8] and R-32 + R-143,[9] and single components, R-32,[10] R-125,[11] and R-143a,[13] a mixture virial equation of state, eq 2, was developed by the present authors.[6] On the other hand, we have the thermodynamic property calculation software, REFPROP (ver. 6.01),[3] which consists of numbers of equations of state for several refrigerants and is widely used for satisfactory prediction for unstudied mixtures. Figure 3 shows a relative pressure deviation of the present measurements from eq 2[6] and REFPROP.[3] Both models reproduce the present measurements within ±0.4% in pressure. Since eq 2 was developed by taking the present *PVTx* properties as a part of the input data, its agreement is better than that of REFPROP. Equation 2 represents the present measurements within ±0.3%, except for a single datum, whereas calculated pressure values by REFPROP deviate by about ±0.4%, which is very good, as it is not based on the ternary data for the system.

It is very meaningful to compare the experimental and calculated values of the second virial coefficient. As shown

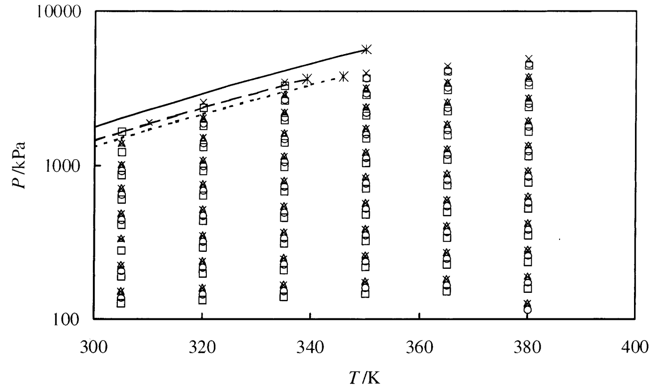

Figure 2. Experimental data distribution of the R-32 + R-125 + R-143a system: ○, ($x_1 = 0.174$, $x_2 = 0.340$); △, ($x_1 = 0.313$, $x_2 = 0.203$); ×, ($x_1 = 0.343$, $x_2 = 0.445$); □, ($x_1 = 0.484$, $x_2 = 0.216$); —, vapor pressure curve for R-32; — — —, vapor pressure curve for R-125; - - -, vapor pressure curve for R-143a; ∗, critical points.

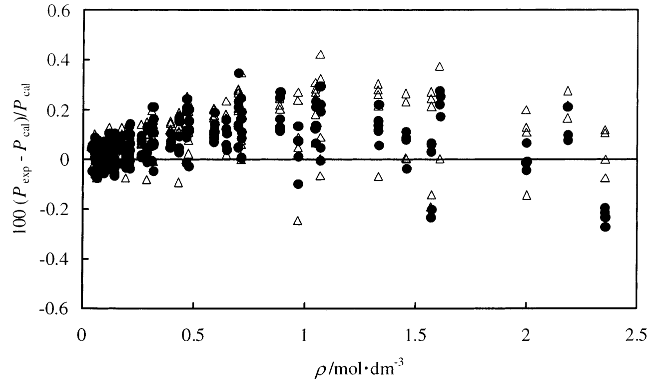

Figure 3. Relative pressure deviation of the present *PVTx* properties from the different models: ●, virial equation of state;[6] △, REFPROP.[3]

Table 2. Experimental Second and Third Virial Coefficients for the R-32 (1) + R-125 (2) + R-143a (3) System

T	B_m	C_m	T	B_m	C_m
K	dm^3·mol^{-1}	dm^6·mol^{-2}	K	dm^3·mol^{-1}	dm^6·mol^{-2}
$x_1 = 0.174$, $x_2 = 0.340$			$x_1 = 0.343$, $x_2 = 0.445$		
380.000	−0.2013	0.020 85	380.000	−0.1867	0.018 22
365.000	−0.2235	0.023 01	365.000	−0.2073	0.020 11
350.000	−0.2492	0.025 72	350.000	−0.2314	0.022 60
335.000	−0.2791	0.029 65	335.000	−0.2596	0.025 37
320.000	−0.3145	0.033 80	320.000	−0.2929	0.029 35
305.000	−0.3563	0.038 86	305.000	−0.3329	0.034 11
$x_1 = 0.313$, $x_2 = 0.203$			$x_1 = 0.484$, $x_2 = 0.216$		
380.000	−0.1947	0.020 45	380.000	−0.1832	0.017 98
365.000	−0.2160	0.022 59	365.000	−0.2030	0.019 91
350.000	−0.2407	0.025 18	350.000	−0.2262	0.022 21
335.000	−0.2695	0.028 26	335.000	−0.2535	0.024 95
320.000	−0.3035	0.032 69	320.000	−0.2858	0.029 08
305.000	−0.3440	0.037 70	305.000	−0.3246	0.033 73

in eq 2, the second virial coefficient of the present mixture, B_m, consists of each cross virial coefficient, B_{ij}, where i and j denote the components.

$$B_m = \sum_{i=1}^{3}\sum_{j=1}^{3} x_i x_j B_{ij} \tag{2}$$

Note that the cross virial coefficients, B_{ij}, of eq 2 were obtained without ternary data so that the mixture second virial coefficients, B_m, are exclusively derived only with data of single components and binary mixtures. Figure 4 shows the experimental second virial coefficients obtained in this study together with the calculated curve by eq 2. The experimental values and calculated curves of the second virial coefficients agree well within ±0.1%. This fact implies the thermodynamic consistency of the present measurements for ternary mixtures and that for each single-component or binary mixture, as well as confirms the sound temperature dependence of the second virial coefficients as well-known.

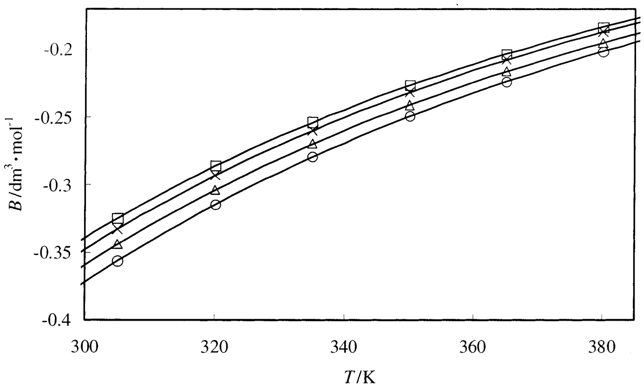

Figure 4. Temperature dependence of the experimentally obtained second virial coefficients for the ternary R-32 + R-125 + R-143a system: ○, ($x_1 = 0.174$, $x_2 = 0.340$); △, ($x_1 = 0.313$, $x_2 = 0.203$); ×, ($x_1 = 0.343$, $x_2 = 0.445$); □, ($x_1 = 0.484$, $x_2 = 0.216$); —, calculated curve by the virial equation of state.[6]

Conclusion

Two hundred and twenty *PVTx* property measurements in the gaseous phase of the ternary system R-32 + R-125 + R-143a at four different compositions were performed in the present study by employing the Burnett−isochoric coupled method. Experimental values of the second and third virial coefficients for the mixtures were also presented. Comparison of the virial equation of state[6] and REFPROP[3] model with the present *PVTx* property data has shown sufficient agreement with the calculated values by these two models. Confirming the present experimental values of the second virial coefficient and calculated values that are obtained with data for pure components and ternary mixtures, it was found that the present measurements do not contradict those for single-component and binary systems.

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