Speed-of-Sound Measurements in Gaseous Binary Refrigerant Mixtures of Difluoromethane (R-32) + 1,1,1,2-Tetrafluoroethane (R-134a)

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One hundred ninety-three speed-of-sound values in gaseous difluoromethane (R-32, CH_2F_2) + 1,1,1,2-tetrafluoroethane (R-134a, CF_3CH_2F) have been measured using a spherical resonator. The measurements have been carried out at temperatures from 303 K to 343 K, pressures up to 240 kPa, and mole fractions of R-32 from 0.16 to 0.90. The experimental uncertainties in the temperature, pressure, and speed of sound for the binary mixture are estimated to be not greater than ±8 mK, ±0.1 kPa, and ±0.0072%, respectively. The samples purified and analyzed by the manufacturers were used and were better than 99.99 mass % for R-32 and 99.98 and 99.99 mass % for two different R-134a samples. We have accurately determined the compositions of the binary refrigerant mixture, R-32 + R-134a, and the second acoustic virial coefficients from the speed-of-sound measurements.

Introduction

The measurement of speed of sound in gases using a spherical resonator is recognized as one of the most accurate methods for determining the thermodynamic properties of very dilute gases such as the ideal-gas heat capacities and the second virial coefficients. Accurate knowledge of the second virial coefficient is essential not only to develop a reliable equation of state but also to evaluate the intermolecular potential parameters. The speed-of-sound measurements with a spherical resonator were reported by Trusler (1984), Moldover et al. (1988), Goodwin and Moldover (1990), Beckermann and Kohler (1995), Trusler et al. (1996), and Estrada-Alexanders and Trusler (1996). Especially, Trusler et al. (1996) reported about the binary mixtures of the (methane + propane) system. In addition, it should be noted that in the case of binary mixtures, we can accurately determine the composition of mixtures on the basis of the ideal-gas heat-capacity values for each component derived from the speed-of-sound measurements. The speed-of-sound measurements for pure components of the present binary refrigerant mixture, i.e., in gaseous 1,1,1,2-tetrafluoroethane (R-134a, CF_3CH_2F) (Hozumi et al., 1993, 1996a) and difluoromethane (R-32, CH_2F_2) (Hozumi et al., 1994b, 1996a), have previously been reported with an experimental uncertainty of ±0.01%. The present system is expected to be a promising refrigerant mixture to replace chlorodifluoromethane (R-22, $CHClF_2$) due to its zero ozone-depleting potential. This paper will provide reliable experimental data on the speed of sound in these mixtures.

Experimental Procedure for Binary Mixtures

The explanation of the experimental procedure used in the present study was given in our earlier publication (Hozumi et al., 1993) for pure refrigerants. As illustrated in Figure 1, in the case of the binary mixture, R-32 + R-134a, the sample was introduced in the vapor phase into the vessel Q from the supply bottle X in which R-32 and R-134a were premixed using a mass balance. The inside and outside of the spherical resonator S were filled with the sample gas. After the thermodynamic equilibrium conditions of the temperature T (±0.1 mK) and pressure P (±0.01 kPa) were confirmed, the frequency, amplitude, and phase difference were measured at the condition of radially symmetric-mode resonance of the sample gas in the spherical resonator. The speed of sound, W, was determined from the values of resonance frequency $f_{l,n}$ and half-width $g_{l,n}$ which were calculated from the values of the frequency, amplitude, and phase difference. The relation among W, $f_{l,n}$, and $g_{l,n}$ is given by a complex resonance expression reported by Moldover et al. (1988) and Ewing et al. (1986),where a and $Z_{l,n}$ in the first term on the right

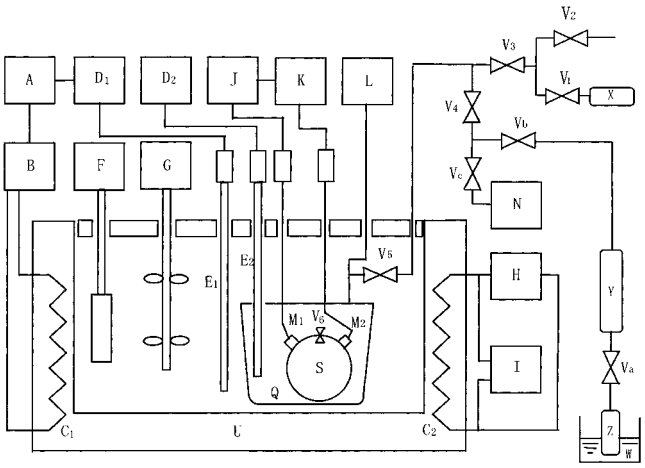

Figure 1. Experimental apparatus: (A) PID controller; (B) thyristor regulator; (C_1, C_2) heaters; (D_1, D_2) thermometer bridges; (E_1, E_2) platinum resistance thermometers; (F) refrigeration unit; (G) stirrer; (H) transformer; (I) voltmeter; (J) frequency synthesizer; (K) lock-in amplifier; (L) pressure gauge; (M_1, M_2) transducers; (N) vacuum pump; (Q) pressure vessel; (S) spherical resonator; (U) thermostated bath; (V_{1-6}, V_{a-c}) valves; (W) liquefied nitrogen; (X) supply bottle; (Y) expansion bottle; (Z) recovery bottle.

$$f_{l,n} + \mathrm{i}g_{l,n} = \frac{WZ_{l,n}}{2\pi a} + \sum_j (\Delta f_{l,n} + \mathrm{i}\Delta g_{l,n})_j \qquad (1)$$

$$l = 0, 1, 2, ...; n = 0, 1, 2, ...$$

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hand side are the radius of the spherical resonator, about 50 mm, and the nth root of the lth order Bessel function, respectively. Each mode is expressed by (l, n), where the value of l is zero in the case of a radially symmetric mode. The second term on the right hand side is a series of perturbation terms expressed by the frequency shift $\Delta f_{l,n}$ and the uncertainty of the half-width $\Delta g_{l,n}$ to represent various nonideal conditions. In the present study, four radially symmetric modes (0, 2) through (0, 5) are used for the data processing.

When a series of measurements was completed at a certain pressure and temperature, we reduced the pressure step by step under the isothermal condition. In the case of the present binary mixture, careful consideration was paid to prevent a composition change in the process. The pressure is reduced by expanding the sample gas into the expansion bottle Y and into the recovery bottle Z. The inner volumes of the bottles Z and Y are about 50 cm^3 and 500 cm^3, respectively. Before reducing the pressure, first we must evacuate the line between valve V_b, and the bottle Z. Second, the pressure of the sample confined within the vessels Q and S is slowly reduced so as to transfer the sample under the gaseous phase into the bottle Y. Finally, the gas-phase sample in the expansion bottle Y is then introduced into the recovery bottle Z which is immersed in liquefied nitrogen, when the sample pressure within the expansion bottle Y is maintained at very low pressure. We repeated this procedure to achieve a desired pressure level in the spherical resonator S.

Composition Determination for Binary Mixtures

We can accurately determine the ideal-gas heat capacity and the second virial coefficient from the speed-of-sound measurements for the pure substances. For binary mixtures, however, we can determine the composition of the sample instead of the ideal-gas heat capacity. We have used the ideal combination rule of the ideal-gas heat capacity for the determination of the composition.

Speed of sound W is expressed by the following truncated virial expansion expression.

$$W(T,P,y)^2 = \frac{\kappa^0(T,y)}{M}(RT + \beta_a(T,y)P + \gamma_a(T,y)P^2) \quad (2)$$

where superscript zero denotes the ideal-gas value, R is the universal gas constant, M is the molar mass, β_a and γ_a are the second and third acoustic-virial coefficients, respectively, κ is the specific-heat ratio c_P/c_v, y is the gaseous mole fraction, and R = 8.314 471 J·mol^{-1}·K^{-1}, which was determined from the similar acoustic method by Moldover et al. (1988). This value differs from a CODATA (Cohen and Taylor, 1986) value, 8.314 510 J·mol^{-1}·K^{-1}, by about 0.0005%.

The relation between κ^0 and the ideal-gas heat capacity, c_P^0 is given by

$$\kappa^0 = \frac{1}{1 - 1/(c_P^0(T,y)/R)} \quad (3)$$

M and c_P^0 are given by the following ideal combination rule.

$$M = y_1M_1 + (1 - y_1)M_2 \quad (4)$$

$$\frac{c_P^0}{R} = \frac{1}{R}[y_1c_{P1}^0 + (1 - y_1)c_{P2}^0] \quad (5)$$

where subscripts 1 and 2 represent R-32 and R-134a, y_1 denotes the gaseous mole fraction of substance R-32. In the present study, the mole fraction, y_1, has been determined from eqs 2 through 5 using measured values of W and previously determined values for c_{P1}^0 and c_{P2}^0.

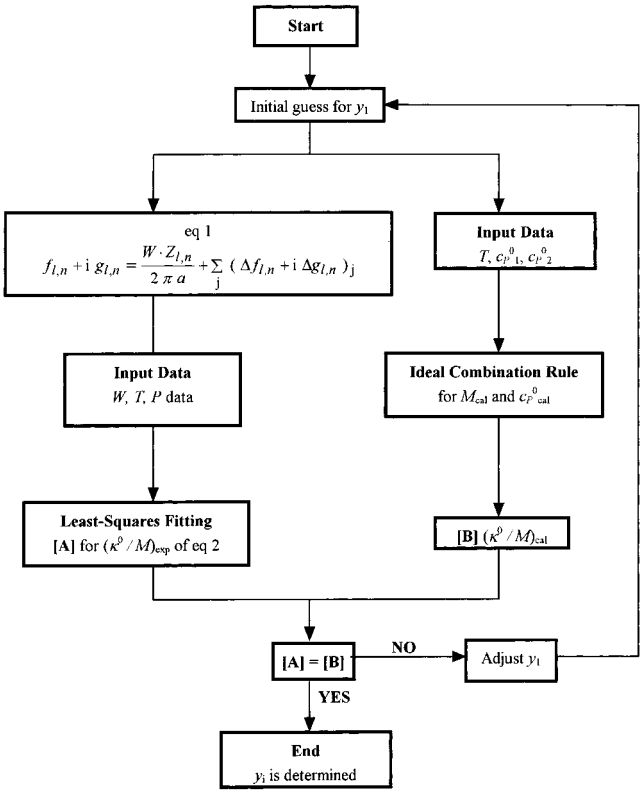

Figure 2. Determination of the compositions for binary mixtures.

The method described above is a direct procedure without the sampling process to determine the mole fraction of gaseous binary mixtures for high accuracy. Namely, we have employed the following procedure shown in Figure 2: the first term in eq 2, $(\kappa^0/M)_{exp}$, was determined by least-squares fitting from the measured speed-of-sound values. c_{P1}^0 and c_{P2}^0 were determined for each pure component. The frequency shift $\Delta f_{l,n}$ in eq 1 depend on the composition, so W values in eq 1 and the first term in eq 2, $(\kappa^0RT/M)_{exp}$, change with different composition values y_1. Therefore, we have to determine both composition y_1 and the correction terms simultaneously by the trial and error procedure in Figure 2.

Experimental Uncertainties and Sample Purities

We follow ISO (International Organization for Standardization) guidelines (ISO, 1993) for the experimental uncertainty. The expanded uncertainty, U, of the measured values can be represented by the following equation.

$$U = k\sqrt{\sum(u_i)^2} \quad (6)$$

where k and u are the coverage factor and the standard uncertainty, respectively. The subscript, i, is the component of the uncertainty. When k is from 2 to 3, then the level of confidence corresponds to 95% to 99%.

In the present study, the coverage factor, k, equals 2. The standard uncertainties, u_i, and the combined uncertainties, $u_c = (\sum(u_i)^2)^{1/2}$, are shown in Table 1. The experimental uncertainties in temperature, pressure, and speed-of-sound measurements are estimated to be not greater than ±8 mK, ±0.1 kPa, and ±0.0072%, respectively. The uncertainty of the composition of R-32 determined from speed-of-sound measurements is estimated to

Table 1. Standard Uncertainties, u_i, for Temperature, Pressure, Speed of Sound, and Composition

Standard Uncertainties for Temperature	
u_1, the standard platinum resistance thermometer E1	1.0 mK
u_2, the platinum resistance thermometer E2 calibrated with the platinum resistance thermometer E1	1.0 mK
u_3, the temperature stability combined standard uncertainty, u_c	4.0 mK
Standard Uncertainties for Pressure	
u_1, dead weight pressure gauge (DH Instruments Inc., Model 5200)	0.03 kPa
u_2, the pressure gauge L calibrated with the dead weight pressure gauge	0.04 kPa
u_3, the pressure stability	0.01 kPa
combined standard uncertainty, u_c	0.05 kPa
Standard Uncertainties for Speed of Sound	
u_1, $f_{l,n}$ and $\sum(\Delta f_{l,n})_j$	0.0012%
u_2, the inner radius of the spherical resonator a	0.0034%
combined standard uncertainty, u_c	0.0036%
Standard Uncertainties for Composition	
u_1, c_P^0	0.10%
u_2, c_{P32}^0	0.04%
u_3, c_{P134a}^0	0.08%
combined standard uncertainty, u_c	0.03 mol %

be not greater than ±0.05 mol %. When the coverage factor, k, equals 3, the uncertainty in speed-of-sound measurements for R-32 + R-134a is ±0.01%, which is similar to values of the earlier measurements (Hozumi et al., 1993, 1994b), ±0.01%.

The samples purified and analyzed by the manufacturers were used and were better than 99.99 mass % for R-32 and 99.98 and 99.99 mass % for two different R-134a samples. We have not purified the sample by ourselves, and we could not find any effect of impurities by filling pure samples from the gas and the liquid phases of the sample bomb. So we can simply rely on the available purity data supplied by the chemical manufacturers.

Results and Discussion

One hundred and ninety-three speed-of-sound values in the gaseous R-32 (1) + R-134a (2) system have been measured for y_1 = 0.16, 0.34, 0.58, 0.76, and 0.90 along five isotherms of (303, 313, 323, 333, and 343) K and pressures from (30 to 240) kPa. The thermophysical properties of the R-32 + R-134a system used were the equation of state developed by Piao et al. (1995), the viscosity values calculated from the equations of Brokaw, Chapman-Enskog, and Reichenberg appearing in Reid et al. (1977), and the thermal conductivity values estimated from the modified Eucken equation appearing in Reid et al. (1977). The correlations used for c_{P1}^0 and c_{P2}^0 were developed by the present authors (Hozumi et al., 1996a). These thermophysical properties were used for the perturbation terms in eq 1. The values of the perturbation terms have an effect of about 0.01% on the speed of sound. We consider that the uncertainty of the perturbation terms has a very small effect on the present measurements.

The present speed-of-sound values are listed in Table 2 and shown in Figure 3. The present results are the average values of (0, 2) to (0, 5) radially symmetric modes. The solid line in Figure 3 is the calculated values from eq 2. Equation 2 represents the experimental speed-of-sound values in Table 2 within standard deviation 0.003%.

Figure 4 shows the present speed-of-sound values in Table 2 for mixtures with y_1 = 0.16−0.90 along the 333 K isotherm and at a pressure of 100 kPa. The solid line is

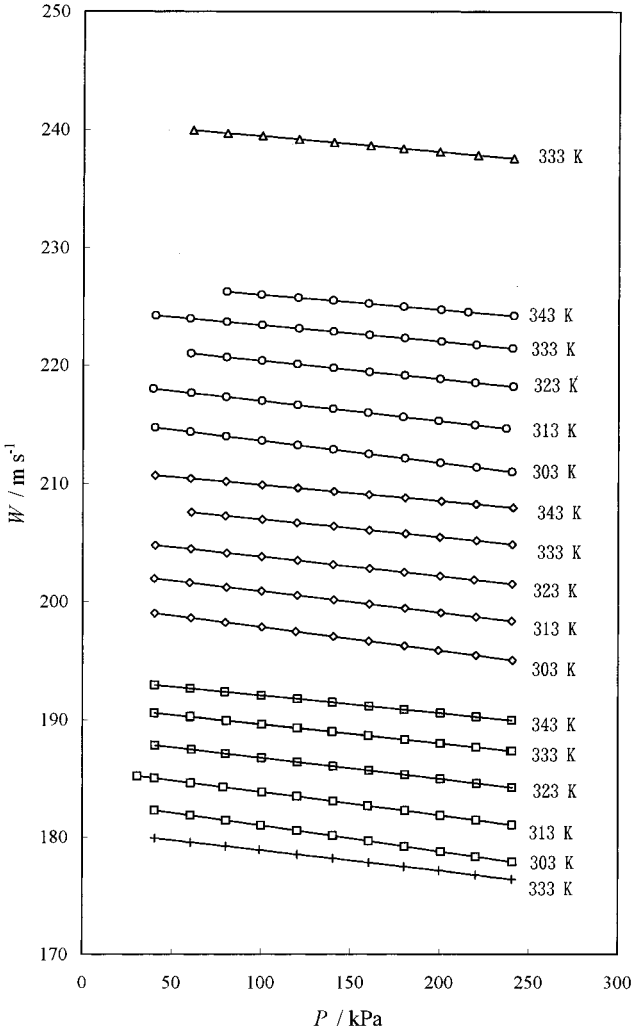

Figure 3. Speed-of-sound data for R-32 (1) + R-134a (2) mixtures: (+) y_1 = 0.16; (□) y_1 = 0.34; (◇) y_1 = 0.58; (○) y_1 = 0.76; (△) y_1 = 0.90; (—) eq 2.

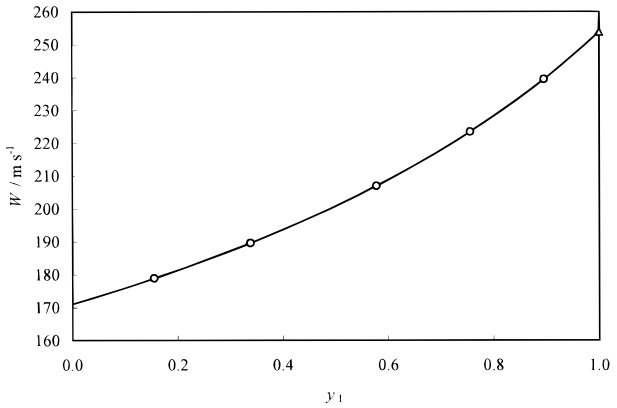

Figure 4. Composition dependence of speed-of-sound data for R-32 (1) + R-134a (2) mixtures along the 333 K isotherm at 100 kPa: (○) present work; (△) y_1 = 1.0 (Hozumi et al., 1996a); (—) present authors (Hozumi et al., 1996b).

the calculated value from the equation of state. This equation of state (Hozumi et al., 1996b) represents the experimental speed-of-sound values in Table 2 within standard deviation 0.0053%. It is found that the present measurements are thermodynamically consistent, which represents the continuous speed-of-sound values for the entire composition range including pure R-32 and R-134a.

Figure 5 shows the deviation of the present measurements at four different radially systematic modes of (0, 2)

Table 2. Speed-of-Sound Values in Gaseous R-32 (1) + R-134a (2) Mixtures

y_1	T/K[a]	P/kPa	W/m·s^{-1}	y_1	T/K[a]	P/kPa	W/m·s^{-1}
			Series A[b]				
0.155 45	333.146	240.55	176.389	0.155 45	333.146	120.51	178.516
		220.23	176.753			99.40	178.884
		199.86	177.117			80.49	179.213
		180.27	177.464			60.58	179.557
		160.53	177.813			40.44	179.905
		140.46	178.167				
			Series B[c]				
0.339 36	303.150	240.92	177.836	0.339 36	303.150	120.45	180.536
		220.55	178.301			100.18	180.980
		200.78	178.748			80.52	181.408
		180.71	179.200			60.58	181.839
		160.64	179.647			40.60	182.269
		140.53	180.094				
			Series C[b]				
0.337 58	313.161	240.69	180.998	0.337 58	313.161	120.47	183.427
		220.62	181.410			100.79	183.816
		200.73	181.815			79.16	184.242
		180.90	182.216			60.72	184.603
		160.23	182.633			40.42	184.999
		140.63	183.025			30.89	185.185
			Series D[b]				
0.337 68	323.161	240.74	184.163	0.337 68	323.161	120.64	186.354
		220.84	184.531			100.75	186.711
		200.72	184.900			80.49	187.073
		180.88	185.262			61.15	187.418
		160.74	185.629			40.69	187.781
		140.78	185.990				
			Series E[b]				
0.338 08	333.147	240.17	187.262	0.338 08	333.147	120.85	189.235
		220.79	187.586			100.57	189.567
		200.84	187.918			80.56	189.892
		180.87	188.249			60.60	190.215
		160.38	188.587			40.39	190.542
		140.08	188.921				
			Series F[b]				
0.338 30	333.147	240.65	187.269	0.338 30	333.147	140.38	188.930
		220.69	187.602			120.55	189.255
		200.43	187.939			100.60	189.580
		180.88	188.264			80.88	189.901
		160.35	188.602			60.60	190.230
			Series G[b]				
0.332 94	343.141	240.47	189.895	0.332 94	343.141	120.60	191.703
		220.70	190.196			100.52	192.003
		200.59	190.500			79.91	192.309
		180.36	190.806			60.64	192.595
		160.68	191.102			40.47	192.897
		140.31	191.408				
			Series H[c]				
0.579 03	303.151	240.65	194.948	0.579 03	303.151	119.40	197.419
		220.27	195.368			100.53	197.796
		199.42	195.796			79.88	198.208
		180.37	196.185			60.59	198.590
		160.27	196.594			40.37	198.989
		140.52	196.994				
			Series I[c]				
0.577 93	313.151	240.31	198.285	0.577 93	313.151	120.22	200.499
		220.29	198.659			100.11	200.864
		200.69	199.023			80.53	201.218
		180.63	199.394			59.82	201.591
		160.46	199.765			40.08	201.944
		140.68	200.127				
			Series J[c]				
0.576 20	323.174	240.47	201.474	0.576 20	323.174	120.47	203.476
		219.08	201.834			100.24	203.809
		200.30	202.149			80.66	204.129
		180.09	202.487			60.52	204.459
		160.68	202.811			40.59	204.782
		140.38	203.147				

Table 2 (Continued)

y_1	T/K[a]	P/kPa	W/m·s^{-1}	y_1	T/K[a]	P/kPa	W/m·s^{-1}
			Series K[b]				
0.577 99	333.147	240.41	204.843	0.577 99	333.147	140.42	206.358
		220.50	205.147			120.06	206.663
		200.10	205.455			100.62	206.954
		180.72	205.750			79.91	207.263
		160.64	206.053			60.68	207.549
			Series L[c]				
0.577 48	343.140	240.93	207.945	0.577 48	343.140	120.39	209.603
		220.27	208.231			100.34	209.876
		200.78	208.500			80.22	210.149
		180.61	208.778			60.27	210.419
		160.30	209.057			40.37	210.686
		140.68	209.326				
			Series M[c]				
0.756 07	303.150	240.41	210.958	0.756 07	303.150	120.43	213.250
		220.70	211.339			100.36	213.627
		200.50	211.728			80.41	214.000
		180.50	212.111			60.41	214.373
		160.42	212.493			40.52	214.742
		140.50	212.871				
			Series N[c]				
0.756 38	313.151	237.24	214.642	0.756 38	313.151	140.39	216.313
		219.78	214.946			120.46	216.652
		199.46	215.298			100.34	216.994
		179.63	215.640			80.38	217.330
		159.93	215.979			60.80	217.660
						39.39	218.019
			Series O[c]				
0.757 55	323.160	241.27	218.169	0.757 55	323.160	140.47	219.747
		219.77	218.508			120.28	220.060
		200.26	218.815			100.41	220.366
		180.32	219.127			80.65	220.669
		160.56	219.435			60.56	220.978
			Series P[b]				
0.756 09	333.149	240.81	221.428	0.756 09	333.149	121.05	223.118
		220.49	221.716			100.49	223.405
		200.87	221.991			80.51	223.679
		180.38	222.284			60.03	223.968
		160.59	222.562			40.77	224.233
		140.53	222.845				
			Series Q[c]				
0.751 11	343.139	241.34	224.185	0.751 11	343.139	140.31	225.491
		215.79	224.516			120.65	225.743
		200.64	224.713			99.97	226.007
		179.70	224.983			80.52	226.256
		160.18	225.235				
			Series R[b]				
0.895 92	333.156	240.96	237.532	0.895 92	333.156	140.22	238.888
		220.83	237.804			120.74	239.149
		199.64	238.090			100.30	239.421
		179.11	238.367			80.71	239.681
		160.68	238.614			61.37	239.937

[a] ITS-90. [b] The purities of R-32 and R-134a are better than 99.99 mass % and 99.98 mass %, respectively. [c] The purity of R-32 and R-134a is better than 99.99 mass %.

to (0, 5) by the deviations from eq 2 at 333.147 K and $y_1 =$ 0.577 99. The values of (0, 2) to (0, 5) radially symmetric modes agree very well with the calculated values from eq 2 within ±0.005%. Similar agreements were also found for the measurements along other isotherms and other isopleths.

We have measured two independent series of speed of sound at a temperature of 333.147 K, but with similar compositions, $y_1 =$ 0.338 08 and 0.338 30. The difference between the two series of measurements is shown in Figure 6. In order to compare the results, the measurements (Δ) at 0.338 30 have been converted to the values (×) at 0.338 08. The two series behave similarly even if the pressure is very low. The reliability of the present measurements was thus confirmed from the fact that the speed-of-sound values with slightly higher composition, $y_1 =$ 0.338 30, are always greater than those for lower composition, $y_1 =$ 0.338 08 and the converted values agree very well with those at $y_1 =$ 0.338 08 within ±0.0010%.

Eighteen second acoustic virial coefficients, β_a, were determined by fitting eq 2 to the squared speed-of-sound measurements at $y_1 =$ 0.16 through 0.90, as listed in Table 3. The standard deviation of β_a is 0.50%, which is rather great because the absolute value of the second term of eq 2 is very small. Figure 7 shows the present β_a values and the values for R-32 and R-134a given in previous publica-

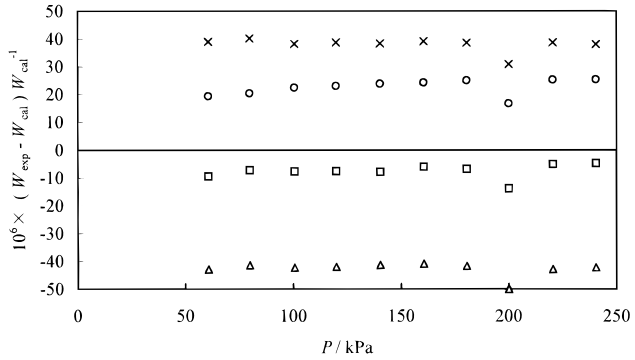

Figure 5. Deviation of the present measurements from eq 2 at 333.147 K, $y_1 = 0.577\ 99$: (○) (0, 2); (△) (0, 3); (□) (0, 4); (×) (0, 5); (—) eq 2.

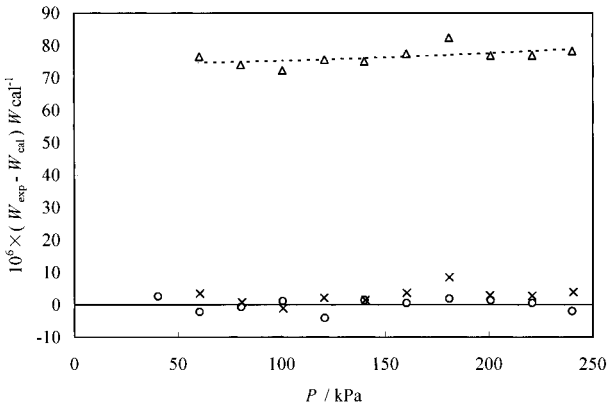

Figure 6. Deviation of the present measurements from eq 2 at 333.147 K, $y_{32} = 0.338\ 08$: (○) $y_1 = 0.338\ 08$; (△) $y_1 = 0.338\ 30$; (×) converted values from $y_1 = 0.338\ 30$ to 0.338 08 by using the ideal mixing rule; (—) eq 2 for $y_1 = 0.338\ 08$; (- - -) eq 2 for $y_1 = 0.338\ 30$.

Table 3. Second Acoustic Virial Coefficients for R-32 (1) + R-134a (2) Mixtures

y_1	T/K	β_a/cm^3·mol^{-1}
0.155 45	333.146	−522.7
0.339 36	303.150	−581.7
0.337 58	313.161	−538.7
0.337 68	323.161	−500.1
0.338 08	333.147	−463.3
0.338 30	333.147	−463.0
0.332 94	343.141	−434.2
0.579 03	303.151	−491.6
0.577 93	313.151	−454.8
0.576 20	323.174	−420.9
0.577 99	333.147	−391.1
0.577 48	343.140	−361.2
0.756 07	303.150	−428.6
0.756 38	313.151	−394.5
0.757 55	323.160	−365.8
0.756 09	333.149	−338.4
0.751 11	343.139	−317.0
0.895 92	333.156	−302.0

tions (Hozumi et al., 1993, 1996a). The solid line is the calculated values from the equation of state (Hozumi et al., 1996b). It was found that the present values for mixtures fall between the values of R-32 and R-134a and agree well with the values of the equation of state (Hozumi et al., 1996b) within ± 0.50%.

Conclusions

The speed of sound in gaseous binary R-32 (1) + R-134a (2) mixtures has been measured by means of a spherical resonator. A total of 193 speed-of-sound values have been measured for the mixtures at y_1 from 0.16 to 0.90 at temperatures from 303 K to 343 K and pressures up to 240 kPa. We could accurately determine the compositions of the binary mixture on the basis of the ideal-gas heat capacity. Eighteen second acoustic virial coefficients have been determined at compositions y_1 from 0.16 to 0.90.

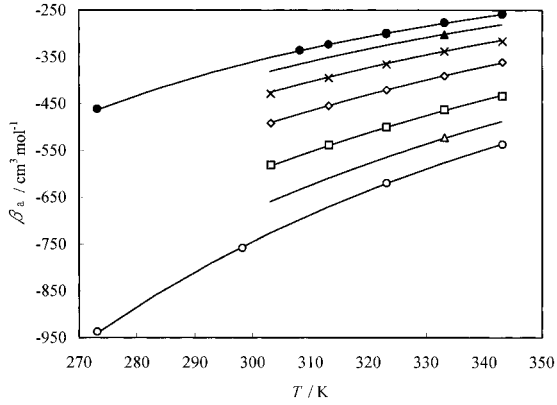

Figure 7. Comparison of second acoustic virial coefficients for R-32 (1) + R-134a (2): (○) $y_1 = 0.0$ (Hozumi et al., 1993, 1996a); (△), $y_1 = 0.16$; (□) $y_1 = 0.34$; (◇) $y_1 = 0.58$; (×) $y_1 = 0.76$; (▲) $y_1 = 0.90$; (●) 1.0 (Hozumi et al., 1996a); (—) present authors (Hozumi et al., 1996b).

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