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Acoustic Determination of Ideal-Gas Heat Capacity and Second Virial Coefficients of Some Refrigerants Between 250 and 420 K[1]

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An apparatus for speed-of-sound measurements with a spherical resonator was adapted for temperatures up to 420 K. This included new microphones with a special wiring, a pressure indicator which can be thermostatted to 420 K, and some installations to avoid temperature gradients. Calibration of the radius of the resonator with argon was extended to higher temperatures. Speed-of-sound measurements up to 420 K and 0.5 MPa were done on 1,1-difluoroethane (R152a), 1,1,1-trifluoroethane (R143a), 1,1,1-chlorodifluoroethane (R142b), 1,1,1,2-tetrafluoroethane (R134a), and 2,2,2-trifluoroethanol. The ideal-gas heat capacities coincide with the statistical mechanical values, except for R134a, where our values as well as recent literature data are below the values calculated from spectroscopy. The reduced second virial coefficients can be interpreted in terms of the dipole moment and the angle between dipole moment and molecular axis. For the associated substance trifluoroethanol values of the third virial coefficient are given, which are appreciably negative at low temperatures.

KEY WORDS: speeds of sound; ideal-gas heat capacity; second virial coefficient; refrigerants.

1. INTRODUCTION

In a previous paper [1], the construction of an apparatus for speed-of-sound measurements with a spherical resonator was described and results on nitrogen, methane, ethane, chlorodifluoromethane (R22), and propane were presented. For less volatile substances, the extension of the technique to higher temperatures is essential. In the present paper, this step is performed up to 420 K.

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The evaluation of the measurements of the speed of sound c is based on the pressure series of c^2:

$$c^2 = A_0 + A_1 p + A_2 p^2 + A_3 p^3 \tag{1}$$

where

$$A_0 = k_0 RT/M \tag{2}$$

and

$$A_1 = (k_0/M)[-2B + 2B'(k_0 - 1) + B''(k_0 - 1)^2/k_0] \tag{3}$$

with

$$B' = T\,dB/dT \qquad \text{and} \qquad B'' = T^2\,d^2B/dT^2$$

Equation (2) gives the connection to the ideal-gas heat capacity

$$c_p^0/R = k_0/(k_0 - 1) \tag{4}$$

and Eq. (3) allows the calculation of the second (thermal) virial coefficient B. This is done usually by assuming a certain function $B(T)$ and determining the parameters of this function such that $A_1(T)$ corresponds as nearly as possible to the experimental results. Therefore, the task is to get accurate values of $A_0(T)$ and $A_1(T)$ in the limit of low pressures. We have measured c^2 between 0.025 and 0.5 MPa and have used two different methods of evaluation. The first is a fit of c^2 with pressure according to Eq. (1) up to the square term (isothermal fit). In the second method, we fit all points in the T, p plane (simultaneous fit), assume temperature functions for $A_0(T)$ and $A_1(T)$, and calculate A_2 and A_3 for each temperature via the appropriate functions of virial coefficients, as will be explained below. The disadvantage of the isothermal fit is a partial compensation of errors in A_1 and A_2. The disadvantage of the simultaneous fit is the arbitrariness of the functions $A_0(T)$ and $A_1(T)$. But this fit smooths small inaccuracies in the temperatures and ensures consistency between A_1 and A_2, and A_3. The connection to A_2 to the virial coefficients is

$$A_2 = (a_{2B} + a_{2C})/(MRT) \tag{5}$$

$$a_{2B} = -(k_0 + 1)\,B^2 + (k_0 - 1)(2k_0 - 1)^2\,B'^2 + (k_0 - 1)^3\,B''^2$$
$$+ (k_0 - 1)^2\,[2BB' + BB'' + (4k_0 - 2)\,B'B''] \tag{5a}$$

$$a_{2C} = (2k_0 + 1)\,C + (k_0^2 - 1)\,C' + (k_0 - 1)^2\,C''/2 \tag{5b}$$

with $C' = T\,dC/dT$ and $C'' = T^2\,d^2C/dT^2$. The a_{2C} term is usually much smaller than the a_{2B} term. Therefore, an approximate correlation for the third virial coefficient C suffices [2]. Compared to A_2 from the isothermal fit, A_2 from Eq. (5) was less negative at low temperatures and less positive at high temperatures. These systematic deviations prompted us to calculate the next coefficient in Eq. (1), A_3, which is given as

$$A_3 = (a_{3B} + a_{3BC} + a_{3D})/(MR^2T^2) \tag{6}$$

$$\begin{aligned} a_{3B} = {} & 2(k_0+1)\,B^3 - 2(2k_0-1)(k_0-1)\,BB'^2 + (k_0-1)^2\,[2(2k_0-1)^2\,B'^3 \\ & + (12k_0^2 - 16k_0 + 5)\,B'^2B'' + (6k_0-4)(k_0-1)\,B'B''^2 + (k_0-1)^2\,B''^3 \\ & - 2B^2B' - B^2B'' - 2BB'B''] \end{aligned} \tag{6a}$$

$$\begin{aligned} a_{3BC} = {} & -4(k_0+1)\,BC - 2(k_0-1)[BC' - k_0(2k_0-1)\,B'C'] \\ & + (k_0-1)^2\,[2B'C + (2k_0-1)\,B'C'' + B''C \\ & + 2k_0B''C' + (k_0-1)\,B''C''] \end{aligned} \tag{6b}$$

$$a_{3D} = 2(k_0+1)\,D + 2(k_0+2)(k_0-1)\,D'/3 + (k_0-1)^2\,D''/3 \tag{6c}$$

with $D' = T\,dD/dT$ and $D'' = T^2\,d^2D/dT^2$. An approximation to D could be obtained from a graph of Ref. 3:

$$D = (B - B_h)^3\,\mu^{*4}(0.1003 - 0.0893\,T/T_c) \tag{7}$$

where B_h is the second virial coefficient of the hard body and μ^* is the reduced dipole moment [4]. This relation is restricted to nonassociated fluids. It turned out that A_3 is nonnegligible at low temperatures and could explain therefore the discrepancies between A_2 from the isothermal fit and A_2 from Eq. (5). The differences at high temperatures remain unexplained.

2. EXPERIMENTS

The basic apparatus has been described before [1]. Here the changes for the extension of measurements to higher temperatures are listed. Most important was the employment of precision microphones (Brüel & Kjær type 4135). The original protecting grid was screwed off and replaced by a protecting mantle which allowed the insertion of the microphones in such a way that the membrane of the microphones was aligned with the inner spherical wall of the resonator. The electric connection to the microphone was made with a copper wire, insulated by Teflon, and surrounded by a steel mantle. For measuring pressure, a differential pressure indicator (MKS type 315BD-10000 Torr) was used, which could be thermostatted to 420 K. In order to avoid temperature gradients coming from the various connections going through the cover of the apparatus, a Pt_{100} sensor was installed at the cover and the temperature difference between cover and resonator minimized by special heating and cooling devices.

Calibration of the resonator radius was done with argon at 400 K and pressures between 0.025 and 0.5 MPa. The values of A_1 and A_2 were $A_1 = 1113\ \mathrm{m^2 \cdot s^{-2} \cdot MPa^{-1}}$ and $A_2 = 40\ \mathrm{m^2 \cdot s^{-2} \cdot MPa^{-2}}$.

Together with the previous calibration between 260 and 350 K [1] this gives for the radius a (in mm)

$$a = 39.9774\{1 + \alpha_0(T - 300) + \alpha_1(T - 300)^2\} \pm 0.0008 \tag{8}$$

with

$$\alpha_0 = 12.0 \times 10^{-6}\ \mathrm{K}^{-1} \quad \text{and} \quad \alpha_1 = 2.1 \times 10^{-9}\ \mathrm{K}^{-2}$$

The evaluation of the resonance curves and the calculation of the speed of sound have been described before [1]. The contribution of the bulk viscosity to the half-width could be neglected in all cases. With the new microphones the resonance curves were less disturbed so that in most cases four modes could be measured. Pressures varied from 0.025 MPa to pressures near the vapor pressure or to 0.5 MPa.

3. RESULTS AND DISCUSSION

The details of the experimental results (resonance frequencies, half-widths, speed of sound) and the details of the input for the corrections are given elsewhere [5]. In the following tables we present the acoustic virial coefficients A_i from isothermal as well as simultaneous fit, the relative standard deviation δ in the measured speed of sound

$$\delta = 100\left[\frac{1}{n}\sum_{i=1}^{n}\left(\frac{c_{i,\exp} - c_{i,\mathrm{fit}}}{c_{i,\exp}}\right)^2\right]^{1/2} \tag{9}$$

and the number of points n of each fit (Tables I, III, V, VII, and IX). Furthermore, we give c_p^0/R from both fits and the values of the second (thermal) virial coefficient. The functions $A_0(T)$ and $A_1(T)$ for the simultaneous fit were

$$A_0(T) = a_{01}T + a_{02}T^2 + a_{03}T^3 + a_{04}T^4 \tag{10}$$

$$A_1(T) = a_{10} + a_{11}/T + a_{12}/T^2 + a_{13}/T^3 \tag{11}$$

In Eq. (11), a nonzero value of a_{12} was used only when δ showed a significant improvement.

Table I. Acoustic Virial Coefficients A_i ($\mathrm{m^2 \cdot s^{-2} \cdot MPa^{-i}}$) for R152a

T (K)	Fit	A_0	A_1	A_2	A_3	n	δ (%)
255	isoth	37149.35	−19229.01	−8512.69		24	0.0023
	simult	37147.46	−19550.04	−5455.18	−5248.64		
260	isoth	37785.31	−18236.25	−6761.03		28	0.0022
	simult	37799.35	−18478.50	−4771.12	−4205.57		
270	isoth	39099.23	−16657.35	−3820.70		32	0.0038
	simult	39101.89	−16618.36	−3646.89	−2709.17		
280	isoth	40380.67	−14780.07	−4044.65		25	0.0037
	simult	40403.08	−15062.75	−2781.03	−1749.58		
290	isoth	41691.42	−13603.40	−2768.21		30	0.0051
	simult	41703.22	−13743.09	−2111.99	−1130.14		
300	isoth	42991.07	−12491.86	−2059.40		31	0.0047
	simult	43002.48	−12607.24	−1594.10	−728.58		
320	isoth	45622.56	−10795.11	−1005.86		32	0.0040
	simult	45598.59	−10736.48	−883.53	−298.58		
340	isoth	48193.82	−9217.39	−630.65		32	0.0035
	simult	48190.74	−9228.84	−464.37	−118.58		
360	isoth	50788.88	−8062.12	7.29		25	0.0040
	simult	50776.59	−7951.16	−228.71	−44.86		
380	isoth	53338.32	−6947.34	297.83		24	0.0023
	simult	53352.07	−6822.23	−111.62	−15.20		
400	isoth	55926.50	−5959.42	475.14		24	0.0048
	simult	55911.45	−5792.07	−72.87	−2.50		
420	isoth	58472.19	−5137.77	484.45		22	0.0091
	simult	58442.36	−4829.38	−86.67	4.87	329	0.0111

Table II. Ideal-Gas Heat Capacities and Second Virial Coefficients for R152a

T (K)	$(c_p^0/R)_{\mathrm{isoth}}$	$(c_p^0/R)_{\mathrm{simult}}$	B ($\mathrm{cm^3 \cdot mol^{-1}}$)
255.0	7.3564	7.3595	−819.8
260.0	7.4727	7.4555	−776.7
270.0	7.6491	7.6465	−699.3
280.0	7.8650	7.8360	−632.1
290.0	8.0390	8.0238	−573.4
300.0	8.2248	8.2099	−521.9
320.0	8.5421	8.5771	−436.4
340.0	8.9338	8.9394	−368.7
360.0	9.2816	9.3014	−314.3
380.0	9.6911	9.6707	−270.0
400.0	10.0317	10.0585	−233.5
420.0	10.4351	10.4804	−203.1

Table III. Acoustic Virial Coefficients A_i ($m^2 \cdot s^{-2} \cdot MPa^{-i}$) for R143a

T (K)	Fit	A_0	A_1	A_2	A_3	n	δ (%)
250	isoth	28105.90	−12716.04	−4170.52		26	0.0108
	simult	28103.18	−12762.69	−2954.17	−1989.86		
270	isoth	30134.91	−10401.17	−1797.31		26	0.0078
	simult	30126.90	−10471.46	−1654.89	−788.59		
290	isoth	32155.84	−8875.13	−961.43		28	0.0053
	simult	32155.87	−8787.35	−916.82	−307.97		
310	isoth	34190.68	−7556.40	−525.47		28	0.0077
	simult	34191.40	−7490.18	−500.10	−116.48		
330	isoth	36235.56	−6365.22	−450.81		31	0.0024
	simult	36233.05	−6447.17	−269.81	−42.28		
350	isoth	38284.52	−5602.14	−35.03		26	0.0103
	simult	38278.68	−5576.20	−146.79	−14.50		
370	isoth	40322.99	−4909.73	136.60		27	0.0033
	simult	40324.40	−4825.18	−82.67	−3.51		
390	isoth	42359.55	−4182.54	121.49		26	0.0072
	simult	42364.62	−4160.38	−46.86	2.80		
410	isoth	44408.04	−3745.08	334.07		28	0.0035
	simult	44392.03	−3559.55	−19.42	9.04	246	0.0222

3.1. 1,1-Difluoroethane (R152a)

Purity was 99.7% as checked by gas chromatography. No difference of the speed of sound could be detected when the sample was taken from the liquid or gas phase of the storage tank. The ideal-gas heat capacities (Table II) agree completely with the spectroscopic values [6]. The second virial coefficients, given here by

$$B(T) = 70.00 - 49{,}163/T - 1.15579 \times 10^{10}/T^3 \qquad (12)$$

Table IV. Ideal-Gas Heat Capacities and Second Virial Coefficients for R143a

T (K)	$(c_p^0/R)_{\text{isoth}}$	$(c_p^0/R)_{\text{simult}}$	B ($cm^3 \cdot mol^{-1}$)
250.0	8.3348	8.3407	−650.4
270.0	8.8051	8.8235	−532.3
290.0	9.2811	9.2811	−442.4
310.0	9.7111	9.7093	−372.4
330.0	10.1020	10.1084	−316.8
350.0	10.4682	10.4834	−271.9
370.0	10.8483	10.8446	−235.2
390.0	11.2215	11.2078	−204.6
410.0	11.5507	11.5948	−179.0

Table V. Acoustic Virial Coefficients A_i ($m^2 \cdot s^{-2} \cdot MPa^{-1}$) for R142b

T (K)	Fit	A_0	A_1	A_2	A_3	n	δ (%)
270	isoth	25001.73	−13229.71	−7275.36		22	0.0136
	simult	25003.10	−13487.77	−4281.71	−2942.46		
290	isoth	26707.96	−11235.85	−3342.97		28	0.0094
	simult	26714.06	−11355.27	−2728.01	−1420.95		
310	isoth	28419.56	−9641.58	−2068.71		28	0.0040
	simult	28423.04	−9692.35	−1747.32	−696.16		
330	isoth	30137.40	−8433.69	−1109.34		28	0.0061
	simult	30131.69	−8370.66	−1113.35	−339.39		
350	isoth	31844.24	−7328.73	−629.08		27	0.0046
	simult	31841.22	−7302.49	−699.32	−161.23		
370	isoth	33554.04	−6489.57	−329.07		26	0.0058
	simult	33552.43	−6426.34	−431.76	−73.29		
390	isoth	35271.71	−5769.78	−98.78		25	0.0064
	simult	35265.70	−5698.17	−266.85	−32.29		
410	isoth	36984.95	−5170.62	478.81		27	0.0055
	simult	36980.95	−5085.80	−177.78	−16.19	211	0.0093

are slightly more negative below 350 K and slightly more positive above 350 K than the values of Tillner-Roth and Baehr [7] and Schramm et al. [8, 9], the largest discrepancies being around 5%.

3.2. 1,1,1-Trifluoroethane (R143a)

Gas chromatography showed a purity of 99.3% (the impurity having a peak very close to the main peak). The sample was gas-free. The ideal-gas heat capacities (Table IV) agree with the spectroscopic values [6]. The second virial coefficients can be only compared between 290 and 310 K

Table VI. Ideal-Gas Heat Capacities and Second Virial Coefficients for R142b

T (K)	$(c_p^0/R)_{\text{isoth}}$	$(c_p^0/R)_{\text{simult}}$	B ($cm^3 \cdot mol^{-1}$)
270.0	9.3881	9.3838	−793.4
290.0	9.8385	9.8186	−664.8
310.0	10.2541	10.2425	−564.5
330.0	10.6317	10.6512	−484.7
350.0	11.0310	11.0415	−420.2
370.0	11.4057	11.4113	−367.3
390.0	11.7383	11.7598	−323.3
410.0	12.0733	12.0877	−286.3

Table VII. Acoustic Virial Coefficients A_i ($\mathrm{m^2 \cdot s^{-2} \cdot MPa^{-i}}$) for R134a

T (K)	Fit	A_0	A_1	A_2	A_3	n	δ (%)
260	isoth	23708.97	−11621.30	−4750.92		23	0.0047
	simult	23715.09	−11874.80	−3135.53	−2235.77		
280	isoth	25404.94	−9561.73	−2430.24		31	0.0073
	simult	25413.71	−9678.47	−1799.40	−933.22		
300	isoth	27099.31	−8051.46	−1332.99		29	0.0048
	simult	27102.99	−8097.67	−1019.24	−384.06		
320	isoth	28781.48	−6827.53	−736.61		28	0.0047
	simult	28785.09	−6901.03	−559.50	−150.57		
340	isoth	30462.19	−5923.43	−334.98		27	0.0054
	simult	30462.23	−5949.65	−291.38	−53.14		
360	isoth	32137.17	−5181.36	−58.31		26	0.0022
	simult	32136.76	−5158.37	−141.31	−14.79		
380	isoth	33819.83	−4571.19	30.93		27	0.0037
	simult	33811.10	−4474.05	−65.64	−1.58		
400	isoth	35496.86	−4010.59	307.14		27	0.0051
	simult	35487.80	−3863.03	−37.56	1.66		
420	isoth	37175.89	−3442.32	356.61		26	0.0066
	simult	37169.51	−3303.73	−40.05	1.67	244	0.0087

[10], where the discrepancy is not more than 1.5%. Our correlation equation is

$$B(T) = 85.71 - 63{,}863/T - 7.50947 \times 10^9/T^3 \tag{13}$$

3.3. 1,1,1-Chlorodifluoroethane (R142b)

Gas chromatography showed a purity of 99.5%; the sample was carefully degassed. The ideal-gas heat capacity (Table VI) agrees with the

Table VIII. Ideal-Gas Heat Capacities and Second Virial Coefficients for R134a

T (K)	$(c_p^0/R)_{\mathrm{isoth}}$	$(c_p^0/R)_{\mathrm{simult}}$	B ($\mathrm{cm^3 \cdot mol^{-1}}$)
260.0	9.4017	9.3829	−723.4
280.0	9.8166	9.7885	−593.0
300.0	10.2164	10.2054	−493.1
320.0	10.6404	10.6296	−415.1
340.0	11.0537	11.0557	−353.0
360.0	11.4733	11.4772	−302.9
380.0	11.8503	11.8861	−261.8
400.0	12.2354	12.2733	−227.8
420.0	12.6003	12.6283	−199.2

Table IX. Acoustic Virial Coefficients A_i ($m^2 \cdot s^{-2} \cdot MPa^{-i}$) for Trifluoroethanol

T (K)	Fit	A_0	A_1	A_2	n	δ (%)
360	isoth	32574.08	−14538.19	−14007.20	24	0.0089
	simult	32555.81	−14410.48	−14077.08		
370	isoth	33399.71	−13056.05	−9795.87	32	0.0067
	simult	33406.16	−12878.08	−10301.16		
380	isoth	34229.78	−11134.29	−8556.54	31	0.0053
	simult	34240.16	−11622.27	−7383.57		
390	isoth	35065.94	−10700.81	−4717.61	23	0.0077
	simult	35066.74	−10602.47	−5153.41		
400	isoth	35908.69	−10045.16	−2990.04	28	0.0084
	simult	35896.55	−9784.43	−3476.62		
410	isoth	36755.86	−9160.39	−2251.42	28	0.0067
	simult	36741.14	−9139.24	−2248.26		
420	isoth	37605.52	−8560.08	−1482.33	28	0.0074
	simult	37613.14	−8642.32	−1385.91	194	0.0250

spectroscopic calculation [11]; the second virial coefficients are a bit more positive than the values of Schramm et al. [8, 9], especially for low and high temperatures, where the discrepancy is 4–6%. Our correlation equation is

$$B(T) = 104.02 - 97.055T - 1.05880 \times 10^{10}/T^3 \qquad (14)$$

3.4. 1,1,1,2-Tetrafluoroethane (R134a)

Purity was 99.7% according to gas chromatography; the sample was carefully degassed. The ideal-gas heat capacities (Table VIII) are about 2% below the spectroscopic values [6], but agree substantially with the acoustic determinations of Goodwin and Moldover [12] and Zhu et al.

Table X. Ideal-Gas Heat Capacities and Second Virial Coefficients for Trifluoroethanol

T (K)	$(c_p^0/R)_{\text{isoth}}$	$(c_p^0/R)_{\text{simult}}$	B ($cm^3 \cdot mol^{-1}$)	C ($cm^6 \cdot mol^{-2}$)
360.0	12.2744	12.2601	−878.7	−1802000
370.0	12.6117	12.6041	−798.7	−1407500
380.0	12.9302	12.9277	−727.2	−1073700
390.0	13.2209	13.2259	−663.3	−795400
400.0	13.4795	13.4937	−605.9	−567960
410.0	13.7149	13.7265	−554.3	−386461
420.0	13.9355	13.9201	−507.8	−246070

[13] and the calorimetric determination of Ernst et al. [14], though the latter two are ca. 0.5% higher. The second virial coefficients agree with various other determinations [7–10, 12] within 2.5%. Our correlation equation is

$$B(T) = 81.45 - 61{,}088/T - 1.00156 \times 10^{10}/T^3 \tag{15}$$

3.5. 2,2,2-Trifluoroethanol

Purity was 99.96%; the sample was carefully dried and degassed. Here the a_{2C} term [Eq. (5b)] is very important. It was determined via the isothermal fit of A_1 and A_2, and calculation of $B(T)$ and a_{2B}. Then a polynomial for $C(T)$ was fitted to reproduce a_{2C} as well as possible. With the smoothed values of a_{2C}, A_1, and B were calculated iteratively for the simultaneous fit. The results are shown in Tables IX and X. No comparison to literature is possible.

It is interesting to plot the second virial coefficients of R152a, R143a, R142b, and R134a on a reduced diagram B/v_c vs. T/T_c and to realize the importance not only of the reduced dipole moment, but also of the angle between dipole moment and molecular axis [4].

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