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Ideal-Gas Heat Capacity Values and Equations for Hydrofluorocarbon (HFC) Refrigerants Based on Speed-of-Sound Measurements

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Final values of ideal-gas heat capacity c_p^0 derived from speed-of-sound measurements using an acoustic spherical resonator and equations of c_p^0 as a simple function of temperature are provided from an overall assessment of speed-of-sound measurements for five hydrofluorocarbon (HFC) refrigerants, difluoromethane (R32), pentafluoroethane (R125), 1,1,1,2-tetrafluoroethane (R134a), 1,1,1-trifluoroethane (R143a), and 1,1-difluoroethane (R152a). Some of the experimental results had systematic errors in comparison with theoretical calculations based on spectroscopic data, which seem to result from the impurity of the sample fluids. The agreement of the experimentally determined and theoretically calculated c_p^0 values was confirmed for HFC refrigerants. The uncertainty of c_p^0 values calculated from the proposed equations is estimated to be 0.1 or 0.2% corresponding to an ISO uncertainty with a coverage factor of $k=1$. An erratum for Table I in a previous report by Yokozeki et al. in 1999 is provided as an appendix.

KEY WORDS: heat capacity; ideal gas; R32; R125; R134a; R143a; R152a; speed of sound.

1. INTRODUCTION

In 1990, Goodwin and Moldover [1] reported c_p^0 values for R134a based on speed-of-sound measurements using an acoustic spherical resonator. The principle of the measurements was established in a study on the universal (molar) gas constant by Moldover et al. [2] that was used to establish the international standard value [3]. Our apparatus was developed by

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Koga [4] on the basis of their studies in 1990 for determining c_p^0 values and the thermodynamic properties of new refrigerants proposed as replacements for chlorofluorocarbons. The principle, experimental apparatus, and procedure of our measurements have been explained in detail in theses [4, 5]. The results measured by our group for hydrofluorocarbons were reported in papers for R152a in 1994 [6], R134a in 1994 [6] and 1996 [6, 7]; R32 in 1996 [7]; R125 in 1996 [8] and 1998 [9]; and R143a in 1997 [10] and 2001 [11].

In 1998, our group, Yokozeki et al. [12], reported the theoretical calculation results for c_p^0 of six HFC refrigerants including trifluoromethane (R23) based on spectroscopic data and provided equations [13] having seven or eight reduced temperature terms. The reason for recalculation of the theoretical c_p^0 values was due to a large discrepancy between the values derived from the speed-of-sound measurements using the acoustic spherical resonator and the previously calculated c_p^0 values based on spectroscopic data, especially for R134a [14]. On the other hand, the agreement between c_p^0 values determined from the speed-of-sound measurements using two different spherical resonators by Goodwin and Moldover [1] and by ourselves [6, 7] was very good.

For R125 [8] and R143a [10], the experimentally derived c_p^0 values did not agree with the calculations by Yokozeki et al. Yokozeki carefully re-examined the calculation procedure but did not find any problems. Simultaneously, we re-examined our experimental speed-of-sound measurements and re-measured R125 [9] and R143a [11] using completely different sample fluids provided by a different manufacturer. The speed-of-sound measurements for R143a have already been reported by Ogawa et al. [11] in 2001. Those for R125 will be submitted soon by Kojima et al. [15].

This current paper will demonstrate the agreement between experimentally and theoretically determined c_p^0 values and will provide a simple four-term correlation of c_p^0 for five HFC refrigerants of R32, R125, R134a, R143a, and R152a. An erratum for Table I in a previous paper for numerical parameters of seven- or eight-term c_p^0 correlations by Yokozeki et al. in 1999 is provided as an appendix at the request of the authors.

2. IDEAL-GAS HEAT CAPACITY FROM SPEED-OF-SOUND MEASUREMENTS

Speed-of-sound measurements were obtained by means of an acoustic spherical resonator. An explanation of the procedure, which was also applied for the present measurements, was reported by Hozumi et al. (1993) [6] and in his thesis (1997) [5]. The inner diameter was about 0.1 m. The original idea of the apparatus and the data procedure were those of

Moldover et al. [2] whose work established the international standard value of the molar gas constant of $8.314472 \pm 0.000015\ \mathrm{kJ \cdot kmol^{-1} \cdot K^{-1}}$ by their speed-of-sound measurements in 1988. They reported a detailed explanation of the principle, apparatus, experimental procedure, corrections, and data processing. Our apparatus was built almost the same as the one developed by Moldover et al., but the flatness of the inner surface of the spherical resonator was very rough (within 50 μm) compared with 0.5 μm for that of Moldover et al. because of different objectives of the work.

We measured the speed of sound in HFC refrigerants while carefully avoiding any impurity introduced in the process of filling and expanding the sample gas in the apparatus. For measurements at equilibrium, the temperature was controlled within ± 0.5 mK while the pressure was controlled within ± 0.01 kPa for several hours. These were the common conditions established for a series of data at a certain temperature and pressure with the highest purity sample fluids.

Speed-of-sound measurements were performed at four different resonance radial modes; the maximum difference among the different modes was about 100 ppm in the speed-of-sound value as shown in Fig. 13 of the literature for R152a [6]. The reproducibility of the c_p^0 measurements using the same sample fluid was within 0.026% as shown in Fig. 4 of the literature for R143a [11]. When we correlate the measured speed-of-sound results squared with a quadratic pressure function at a certain temperature, the maximum deviation was 26 ppm as shown in Fig. 3 of the literature for the case of R143a [11]. We estimated the experimental uncertainties for all the measurements for the five refrigerants as follows: 4 mK in temperature measurements, 0.1 kPa in pressure measurements, and 36 ppm in the speed-of-sound measurements according to the ISO guide (1993) with a coverage factor of $k = 1$ (coverage factor of 68%).

The c_p^0 values were determined from the constant parameter of a quadratic pressure-series equation, Eq. (1), fitted to speed-of-sound measurements at different pressures between 10 and 500 kPa along each isotherm.

$$W^2 = \frac{\gamma^0 RT}{M}\left\{1 + \beta_a\left(\frac{p}{RT}\right) + \gamma_a\left(\frac{p}{RT}\right)^2\right\} \tag{1}$$

$$\frac{c_p^0}{R} = \frac{\gamma^0}{\gamma^0 - 1} \tag{2}$$

where the superscript zero denotes the ideal-gas condition; R is the molar gas constant; M is the molar mass; β_a and γ_a are the second and third acoustic-virial coefficients; and γ is the specific-heat ratio.

All of the c_p^0 values derived for each isotherm using Eq. (1) are listed in Table I for each substance along with the mean value of the standard deviations uc_p^0 derived from the uncertainty of γ^0 in Eq. (1) on the basis of measured speed-of-sound data along each isotherm.

In the following sections, c_p^0 equations and the comparison of experimental and theoretical data will be explained for each refrigerant. The equations were correlated on the basis of the theoretically calculated values by Yokozeki et al. between 200 and 500 K with a simple power series temperature function.

$$c_p^0 = c_0 + c_1 T_r + c_2 T_r^2 + c_3 T_r^3 \tag{3}$$

where T_r is the reduced temperature, T/T_c. The critical temperature, T_c, was taken from JARef [16]. The numerical values of the parameters, c_0 to c_3, and T_c are listed in Table II. On the basis of the equations, the agreement of c_p^0 values calculated from spectroscopic data and from speed-of-sound data will be discussed for R32, R125, R134a, R143a, and R152a in the following sections.

2.1. R32 (Difluoromethane)

In 1996, our group Hozumi et al. [7] reported the speed of sound in gaseous R32 whose purity was 99.99 mass% by using the same apparatus and procedure as those of our other measurements. The c_p^0 data, except for that at 273 K, agree with Eq. (3) with a maximum deviation of 0.076% and a standard deviation of 0.031% as shown in Fig. 1. By reanalyzing all of

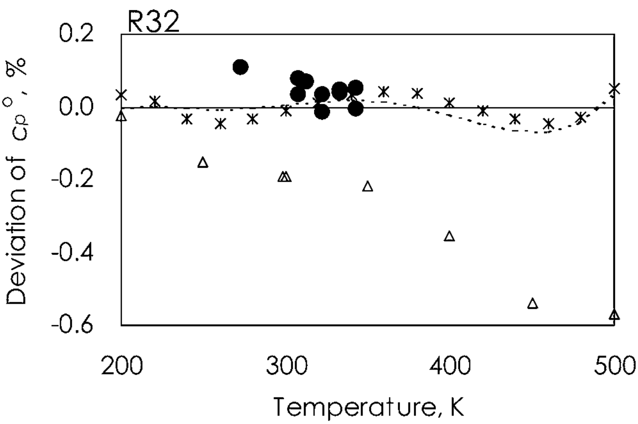

Fig. 1. Deviation of c_p^0 values from Eq. (3) for R32: (●) Hozumi et al. [7]; (∗, ···) Yokozeki et al. [12, 13]; (△) Chase, Jr. et al. [17].

Table I. c_p^0 Values Experimentally Determined on the Basis of Speed-of-sound Data for R32, R125, R134a, R143a, and R152a Measured by Our Group

T (K)	c_p^0/R	uc_p^0 (%) ($k=2$)
	R32	
273.155	4.949	0.014
308.157	5.261	0.012
313.151	5.308	0.012
323.149	5.399	0.012
333.149	5.499	0.015
343.147	5.595	0.018
308.183	5.259	0.014
323.174	5.402	0.013
333.141	5.499	0.014
343.136	5.598	0.012
	R125	
273.153	10.725	0.095
303.144	11.485	0.038
313.153	11.713	0.034
323.137	11.927	0.047
333.148	12.129	0.040
343.141	12.329	0.056
	R134a	
273.154	9.673	0.037
298.217	10.210	0.067
323.15	10.721	0.040
343.138	11.149	0.055
	R143a	
303.143	9.487	0.033
313.144	9.704	0.028
323.133	9.903	0.025
333.141	10.098	0.025
343.142	10.295	0.029
	R152a	
273.165	7.699	0.024
298.151	8.139	0.034
313.156	8.407	0.045
318.158	8.512	0.030
328.153	8.700	0.031
338.156	8.881	0.031
348.158	9.057	0.043

Table II. Numerical Parameters and the Critical Temperature of Eq. (3)

	R32	R125	R134a	R143a	R152a
c_0	4.4971	3.0614	3.1610	1.8567	3.4943
c_1	−2.8987	10.7918	8.7589	10.1478	3.6118
c_2	5.8251	−1.2173	1.0384	−1.4867	4.6738
c_3	−1.74767	−0.36795	−1.18189	−0.16996	−2.04672
T_c (K)	351.255	339.165	374.083	345.860	386.410

the measurements for the alternative refrigerants, we found an unexpected error in our measurements at 273 K that was not included in the data at higher temperatures. The actual error source was not clear, but the smaller signals of sound resonance, the unexpected heat loss, or temperature gradient in the spherical resonator were suspect. At present we are constructing a new thermostat bath for minimizing the heat loss and the temperature gradient. Equation (3) reproduces Yokozeki's values [12] within $\pm 0.052\%$ and his equation [13] within $\pm 0.041\%$ between 200 and 500 K. Chase et al. [17] reported the theoretical calculations in 1985 whose values have systematic errors of 0% at 200 K and -0.6% at 500 K. Because of the good agreement of experimentally and theoretically determined c_p^0 values, the uncertainty of calculated c_p^0 values from Eq. (3) can be estimated to be better than 0.1% for $k=1$ over a temperature range at least within our experimental range from 310 to 343 K. The result is consistent with theoretical calculations of Yokozeki et al. from 200 to 500 K (or the temperature at which dissociation starts).

2.2. R125 (Pentafluoroethane)

In 1996, Hozumi et al. [8] reported the speed of sound in gaseous R125 whose purity was 99.998 mass% while using the same apparatus and procedure as those of other measurements. The c_p^0 data have a systematic error of about -0.8% from Eq. (3) as shown in Fig. 2. Ichikawa et al. [8] re-measured the speed of sound by using the same apparatus but with a different sample fluid whose purity was 99.953 mass% in 1998 obtained from a different manufacturer. The impurity in the sample fluid used by Hozumi et al. is unknown, but it is clear that the sample fluid contained some impurities. Based on the same reasons as discussed in the previous section, the measurements at 273 K by Ichikawa et al. also have an unexpected error. Equation (3) reproduces Yokozeki's values [12] within $\pm 0.030\%$ and his equation [13] within $\pm 0.051\%$ between 200 and 500 K.

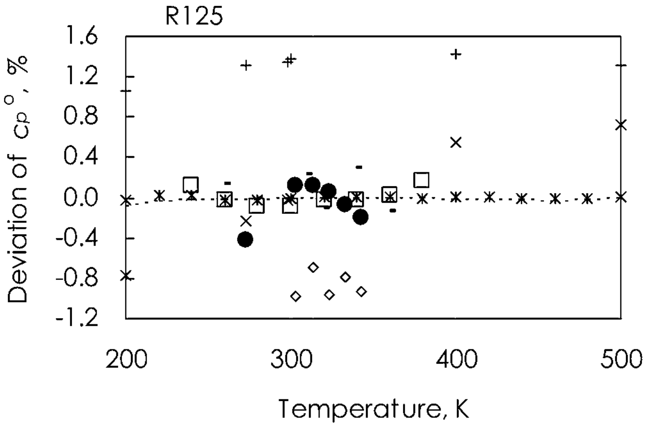

Fig. 2. Deviation of c_p^0 values from Eq. (3) for R125: (●) Ichikawa et al. [9]; (◇) Hozumi et al. [8]; (□) Gillis [18]; (–) Grigiante et al. [19]; (∗, ···) Yokozeki et al. [12, 13]; (×) Chao and Rodgers (TRC) [20]; (+) Chen et al. [14].

Gillis [18] and Grigiante et al. [19] also reported c_p^0 values derived from similar speed-of-sound measurements. The data of Gillis agree extremely well with our data and theoretical values calculated by Yokozeki et al. within ±0.17% as shown in Fig. 2. The data of Grigiante et al. agree with Eq. (3) within ±0.29%. The uncertainty of calculated c_p^0 values from Eq. (3) can be estimated to better than 0.1% for $k = 1$ for the temperature range at least within our experimental range. This result is consistent with calculations of Yokozeki et al. from 200 to 500 K. On the other hand, Chen et al. [14] and Chao and Rodgers (TRC) [20] reported theoretical calculations in 1975 and 1989 based on spectroscopic data whose values have systematic errors of +1.4% or −0.8% at 200 K and +0.75% at 500 K. The reason for the systematic error was discussed by Yokozeki et al. [12].

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

In 1993 [6] and 1996 [7], Hozumi et al. [6] reported speed-of-sound measurements for R134a, whose purity was 99.98 mass% and 99.95 mass%, respectively, by means of the same apparatus and procedure. The measurement at 273 K does not shows the systematic error that appeared in the data for other refrigerants as shown in Fig. 3. We cannot explain the reason. Equation (3) reproduces Yokozeki's values [12] within ±0.061% and his equation [13] within ±0.058% between 200 and 500 K. Goodwin

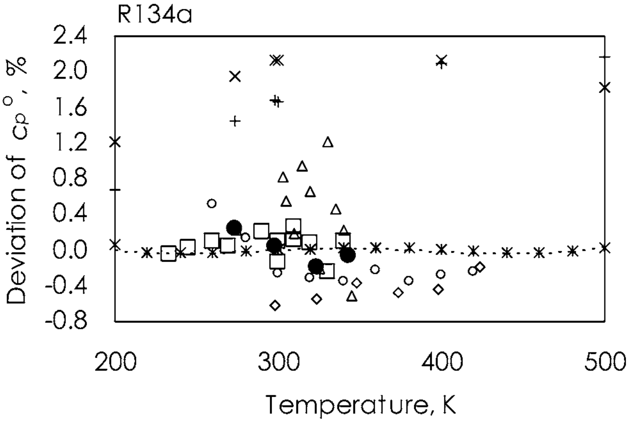

Fig. 3. Deviation of c_p^0 values from Eq. (3) for R134a: (●) Hozumi et al. [6, 7]; (∘) Beckermann and Kohler [21]; (□) Goodwin and Moldover [1]; (△) Zhu et al. [23]; (◇) Türk et al. [22]; (∗, ···) Yokozeki et al. [12, 13]; (×) Chao and Rodgers (TRC) [20]; (+) Chen et al. [14].

and Moldover [1] reported c_p^0 values derived from speed-of-sound measurements using a spherical resonator in 1990. Hozumi's data agree extremely well with the data of Goodwin and Moldover. Similar data reported by Beckermann and Kohler [21] are smaller, with a deviation of −0.35%. Türk et al. [22] and Zhu et al. [23] measured the speed of sound in gaseous R134a, and the deviations of the c_p^0 values are −0.5% systematically smaller or scattered within ±1%, respectively. The uncertainty of calculated c_p^0 values from Eq. (3) can be estimated to be better than 0.1% for $k = 1$ in the temperature range at least within our experimental range. This result is consistent with the theoretical calculations of Yokozeki et al. from 200 to 500 K.

Because the uncertainties of measurements are greater than those in the recent measurements for other refrigerants, the uncertainty of Eq. (3) seems rather large compared to that for other refrigerants. We estimate the final uncertainty to be 0.1% because of the reliabilities of the data of Goodwin and Moldover and of the theoretical calculations carefully performed by Yokozeki et al. [12] in 1998. On the other hand, Chen et al. [14] and Chao and Rodgers (TRC) [20] calculated c_p^0 values based on spectroscopic data in 1975 and 1989. The data have systematic errors of +0.68% at 200 K and +2.1% at 400 K or about +2%, respectively. The reason for the systematic error was discussed in detail by Yokozeki et al. [12].

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

In 1997, Ichikawa et al. [10] reported the speed of sound for gaseous R143a whose purity was 99.97 mass% (originally reported as 99.99 area% G.C.). The data have a systematic error of about +0.4% from Eq. (3). Yokozeki et al. tried to find the reason explaining the difference between the data of Ichikawa et al. and the theoretical c_p^0 values. Yokozeki et al. believed their theoretical values at the time in 1998. Ogawa et al. [11] re-measured the speed of sound in 2001 using the same apparatus but with a different sample fluid provided by a different manufacturer for which the purity was 99.95 G.C. area%. These data agree with Yokozeki's theoretical values within ±0.1% as shown in Fig. 4. It should be noted that the c_p^0 values were measured after they were calculated by Yokozeki et al. in 1998 [12]. The impurity in the sample fluid used by Ichikawa et al. is still unknown. For an understanding of the difference between the two sets of measurements, Ogawa et al. measured the speed of sound in the earlier sample fluid at 343 K. The data reproduced previous c_p^0 values within 0.026%. Equation (3) reproduces Yokozeki's spectroscopic-data values [12] within ±0.016% and his equation [13] within ±0.049% between 200 and 500 K. Gillis [18] also reported c_p^0 values derived from similar speed-of-sound measurements. His data agree, in general, with Yokozeki's theoretical values within ±0.2% at temperatures below 360 K except at 250 K, where the data deviate by −0.35% as shown in Fig. 4. Beckermann

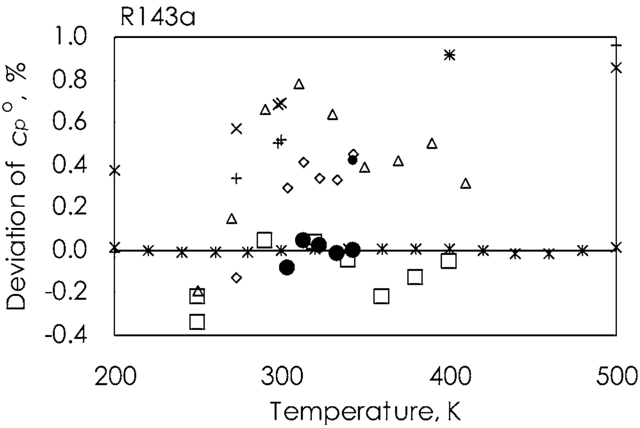

Fig. 4. Deviation of c_p^0 values from Eq. (3) for R143a: (●) Ogawa et al. [11]; (◇) Ichikawa et al. [10]; (•) Ogawa et al. (re-measured point) [11]; (△) Beckermann and Kohler [21]; (□) Gillis [18]; (∗, ···) Yokozeki et al. [12, 13]; (+) Chen et al. [14]; (×) Chao and Rodgers (TRC) [20].

and Kohler [21] also reported similar speed-of-sound measurements. Their c_p^0 values are about 0.5% larger than these from Eq. (3). The uncertainty of calculated c_p^0 values from Eq. (3) can be estimated to be better than 0.1% for $k = 1$ for the temperature range at least within our experimental range. This result is consistent with the theoretical calculations by Yokozeki et al. from 200 to 500 K. On the other hand, Chen et al. [14] and Chao and Rodgers (TRC) [20] reported the results of theoretical calculations in 1975 and 1989 based on spectroscopic data for which the values have systematic errors of +0.4% at 273 K to +1% at 500 K or +0.37% at 200 K and +0.86% at 500 K, respectively. The reason for the systematic error was discussed in detail in the previous paper of Yokozeki et al. [12].

2.5. R152a (1,1-Difluoroethane)

In 1993, Hozumi et al. [6] reported the speed of sound for gaseous R152a whose purity was 99.8 mass% using the same apparatus and procedure. Equation (3) reproduces Yokozeki's spectroscopic-data values [12] within ±0.044% and his equation [13] within ±0.040% between 200 and 500 K. The derived c_p^0 values from the speed-of-sound data by Hozumi et al., except for that at 273 K, agree with Eq. (3) within +0.1% to +0.25% as shown in Fig. 5. Gillis [18] also reported speed-of-sound data in 1997. His derived c_p^0 data agree with Eq. (3) within ±0.21%. Türk et al. [22]

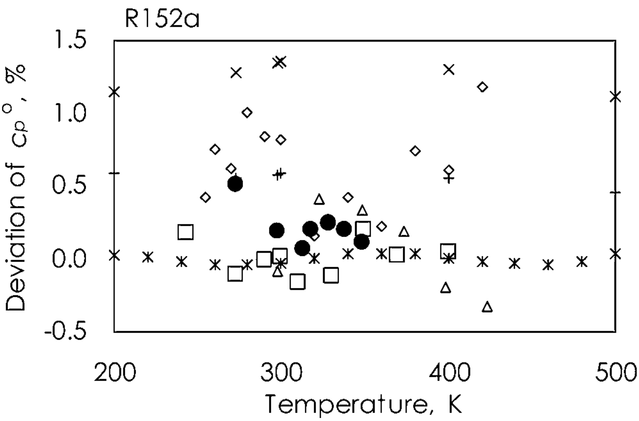

Fig. 5. Deviation of c_p^0 values from Eq. (3) for R152a: (●) Hozumi et al. [6]; (◇) Beckermann and Kohler [21]; (□) Gillis [18]; (*, ···) Yokozeki et al. [12, 13]; (△) Türk et al. [22]; (+) Chen et al. [14]; (×) Chao and Rodgers (TRC) [20].

reported c_p^0 values that agree with Eq. (3) within $\pm 0.42\%$. The values measured by Beckermann and Kohler [21] show systematic deviations of 0.25% and 1.2% from Eq. (3). The uncertainty of calculated c_p^0 values from Eq. (3) can be estimated to be better than 0.2% for $k = 1$ for the temperature range at least within our experimental range. This result is consistent with the theoretical calculations of Yokozeki et al. from 200 to 500 K. On the other hand, Chen et al. [14] and Chao and Rodgers (TRC) [20] reported the results of theoretical calculations in 1975 and 1989 based on spectroscopic data whose values have systematic errors of about +0.6% or +1.3, respectively. The reason for the systematic error was discussed in detail in the previous paper of Yokozeki et al. [12].

3. SUMMARY

c_p^0 values for five HFC refrigerants, R32, R125, R134a, R143a, and R152a, are discussed from an experimental viewpoint. The final experimentally determined c_p^0 values are summarized in Table I. In addition, an erratum for the work of Yokozeki et al. [13] is presented in the appendix. A theoretical approach was reported in a previous paper by Yokozeki et al. [12] After the theoretical results reported by Yokozeki et al. in 1998, we continued to measure the speed of sound in gaseous R125 and R143a using different sample fluids provided by a different manufacturer to solve the discrepancy with theoretical results. The experimentally and theoretically determined c_p^0 values finally agreed to within $\pm 0.1\%$ for R32, R125, R134a, and R143a.

The uncertainty of c_p^0 values is always difficult to be estimated because c_p^0 values are usually determined from a theoretical model, and these cannot be directly measured but must be derived from spectroscopic data or speed-of-sound measurements. As a conclusion, using either the careful theoretical calculations based on spectroscopic data or the experimental determination from speed-of-sound measurements, the c_p^0 values can be reliably determined. The best way to determine the c_p^0 values might be to be derived from spectroscopic data for results over a wide temperature range, and the calculated results should be confirmed with data derived from speed-of-sound measurements for estimating the uncertainty. c_p^0 equations with a simple temperature function are provided for five refrigerants. The uncertainty of c_p^0 values calculated from the equations is estimated to be 0.1% for R32, R125, R134a, and R143a, and 0.2% for R152a corresponding to an ISO uncertainty for $k = 1$ on the basis of statistical considerations for the data treatment in the process of fitting c_p^0 values obtained independently at different temperatures .

APPENDIX

Table AI. Corrections for Table I in Ref. 13. Coefficients in Eq. (1) for Ideal-Gas Heat Capacities

	R32	R125	R134a	R143a	R152a
a_0	6.168087	1.367753×10	1.245089×10	1.151193×10	1.014258×10
a_1	1.008226	1.833764	1.801270	1.706299	1.704048
a_2	-1.577759×10^{-2}	-2.225800×10^{-1}	-2.023163×10^{-1}	-1.684880×10^{-1}	-1.242065×10^{-1}
a_3	-3.224254×10^{-2}	-3.333652×10^{-3}	-7.333155×10^{-3}	-2.426148×10^{-3}	-2.195255×10^{-2}
a_4	6.782532×10^{-3}	4.226685×10^{-3}	7.263184×10^{-3}	-4.261583×10^{-4}	8.522034×10^{-3}
a_5	7.322812×10^{-4}	5.010408×10^{-4}	5.424829×10^{-5}	1.632690×10^{-3}	4.943119×10^{-4}
a_6	-2.994537×10^{-4}	-1.964569×10^{-4}	-2.632141×10^{-4}	3.220782×10^{-4}	-3.929138×10^{-4}
a_7	—	—	—	-1.759529×10^{-4}	—
T_0	4.0	4.1	4.1	4.1	4.1
T_1	100.0	100.0	100.0	100.0	100.0

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