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Measurement of the Compressibility and Sound Velocity of Helium up to 1 GPa

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By using a gas expansion technique, the density of helium has been determined at 298.15 K as a function of pressure from 100 MPa to 1 GPa. The precision of the measurements is 0.02%, while the estimated absolute accuracy is about 0.08%. The sound velocity has been measured by a phase-comparison pulse-echo technique between 98 and 298 K with intervals of 25 K and at pressures up to 1 GPa, with an accuracy generally better than 0.04%. By combining pVT with velocity-of-sound data at 298 K, the adiabatic compressibility and the ratio of the specific heats are calculated. The experimental sound velocities are compared with the values, predicted from an equation of state as proposed by Hansen.

KEY WORDS: adiabatic compressibility; equation of state; density; high pressure; isothermal compressibility; helium; pVT; sound velocity; ultrasonics.

1. INTRODUCTION

Helium, although belonging to the group of rare gases, is the second most abundant element in the universe. In all states of aggregation—gas, liquid, and solid—it has unique properties of importance from both the scientific and the technical point of view. Many standard works are devoted to this element [1–3], but there still is a need for accurate equation-of-state data over a wide pressure and temperature range. The first, rather crude study of the fluid compressibility at elevated pressures (above 100 MPa) was undertaken in 1923 by Bridgman [4, 5]. With pressures up to 1.5 GPa in the temperature range of 303–368 K, Bridgman certainly was a pioneer in this field. Later studies were made by Tsiklis et al. [6] up to 0.7 GPa in the temperature range of 293–423 K. Vidal et al. [7] (up to 1 GPa at 298 K)

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and Mills et al. [8] (up to 2 GPa at 75–300 K) combined compressibility with velocity-of-sound measurements. The velocity of sound was also measured by Vereshchagin et al. [9] up to 350 MPa at 298 K, Pitaevskaya and Bilevich [10] up to 400 MPa (298–473 K), and Nishitake and Hanayama [11] up to 1 GPa at 298 K. At pressures below 100 MPa, several studies were undertaken. A review of this work was given by Gammon [12], who measured the velocity of sound at pressures up to 15 MPa (100–423 K) with a precision of 0.001%, in order to obtain the parameters for his equation of state, based on perturbation theory.

The most accurate of the direct measurements of the compressibility of helium below 100 MPa (268–353 K) were reported by Briggs et al. [13] and Briggs [14]. The uncertainty in the compressibility factor (0.05 %) in this pressure region is at least 10 times smaller than the most accurate measurements at pressures above 100 MPa.

As a continuation of the study at the Van der Waals Laboratory of the acoustic and thermodynamic properties of simple gases (argon [15, 16], neon [17], nitrogen [18], methane [19]) at pressures up to 1 GPa, the results on helium are presented here. Our aim was to measure the density up to 1 GPa, with an accuracy comparable to that of the measurements in the pressure range up to 100 MPa, and to obtain reliable thermodynamic properties of helium by combining compressibility with sound-velocity data.

2. EXPERIMENTAL

An extensive description of the method and the equipment for the determination of the compressibility isotherms of gases upto 1 GPa has been given previously [15]. Briefly, the method involves the expansion of the gas from an initially pressurized vessel of volume V_A into an evacuated vessel V_B of nearly the same volume. In contrast with the well-known Burnett method of repeated expansions, we do not reevacuate vessel V_B and make new expansions, but return to the initial situation, with vessel V_A charged with a different density. In a total expansion run, the pressure after expansion is equal in both vessels, whereas in a stepwise expansion run the gas is partly expanded. The pressure in V_B is generally lower than in V_A. After each expansion the pressure is recorded in both vessels. The density on the high-pressure side (vessel V_A) is calculated from the density on the low-pressure side (vessel V_B), which in its turn is obtained from the literature. In the case of helium, we used the results of Briggs [14], who determined with a Burnett apparatus low-pressures pVT data in the temperature range from 268 to 353 K and at pressures up to 80 MPa, with an accuracy of the compressibility factor of better than 0.05%. These data

were also used to determine the volume ratio V_B/V_A of the pressure vessels, whereas the pressure distortion coefficients β_A and $\beta_B[\beta \equiv (1/V)(\Delta V/\Delta p)_T]$ of the pressure vessels were determined previously by calibration with argon up to 280 MPa.

The velocity of sound in helium was measured with the well-known phase-comparison pulse-echo technique, operating with one X-cut quartz transducer and two reflectors at unequal distances, having a ratio of 3:2. Two succesive longitudinal sound-wave pulses are introduced in the pressurized fluid by applying to the transducer two electrical pulses, produced by a synthesizer, a pulse former, and a timer. At certain frequencies, the so-called null frequencies, the phase difference between the echo of the first pulse reflected by the distant reflector and the echo from the second pulse reflected by the near reflector equals $(2n+1)\pi$, where the integer n is the order of interference. The two echoes cancel each other, provided that the amplitudes are equal. From these null frequencies, one can calculate very accurately the difference in transit time of the sound pulses necessary to cover the difference in path lengths. The systematic error in the sound velocity in this method is estimated to be less than 0.02% and random errors are caused mainly by the uncertainty in the pressure measurement. The null frequencies range from 9.5 to 10.5 MHz. Details of the equipment and the method are given elsewhere [16].

The gas under investigation was purchased from Matheson Co, U.S.A., and was stated to be 99.9999% pure.

3. RESULTS

Since we initially intended to use the results of Michels and Wouters [20] as reference data, 12 total expansions were performed between 20 and 68 MPa. In the high-pressure range, from 68 to 1000 MPa, two stepwise expansions, each with eight intermediate data points and 35 total expansion runs, were carried out. Due to the lack of an analytical equation, the cubic spline method, taking into account 17 knots, was used to calculate the density at round values of the pressure. The standard deviation in the density as a function of pressure is 0.02% and the deviations are randomly distributed. The systematic error, due to the uncertainty in the volume ratio and the pressure distortion coefficients, is estimated to be 0.03%. If we assume that the systematic error in the Briggs compressibility factor data is 0.04% and that the error in the pressure determination is 0.01%, then the systematic error in the reference density data is 0.03%. By combining the random and the systematic errors, the total accuracy of the data presented here is found to be 0.08%. In contrast with the results for

nitrogen [18], for helium there were no systematic deviations between the data obtained from stepwise and those obtained from total expansions.

The sound-velocity measurements were carried out from 1 GPa down to 100 MPa, in steps of 50 MPa at higher pressures and in steps of 25 MPa below 250 MPa. The isotherms are shown in Fig. 1 over the full pressure range and in Fig. 2 up to 400 MPa, whereas isobars are shown in Fig. 3. The data of the sound velocity at round values of the pressure along the isotherms were obtained by fitting the data to polynomials: $p = \sum_{i=0}^{4} a_i w^i$. The standard deviations of the sound velocity isotherms, due to random errors, range from 0.005 to 0.02%. Only the 173 K isotherm has a larger standard deviation, 0.05%. As the systematic errors are estimated to be about 0.02%, the overall accuracy will generally be better than 0.04%. For the 173 K isotherm this will be 0.08%. Normally the isotherms end at about 100 MPa, due to the mismatch of the acoustical impedance between the quartz transducer and the sample. In the case of the isotherms at 298 and 273 K, the lowest pressure at which the sound velocity could be determined was about 130 MPa and the values at 100 MPa were obtained by fitting our data together with those of Gammon [12] to polynomials of the same type as described above. Although the gap in pressure is rather large,

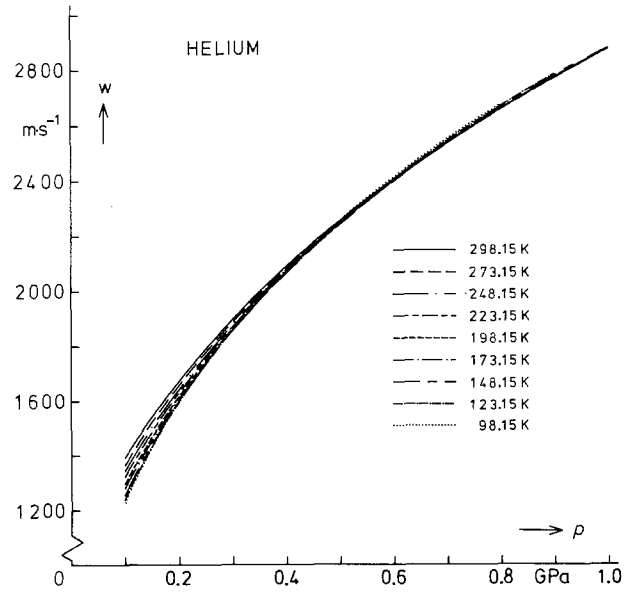

Fig. 1. Sound velocity in helium as a function of pressure up to 1 GPa, at several temperatures.

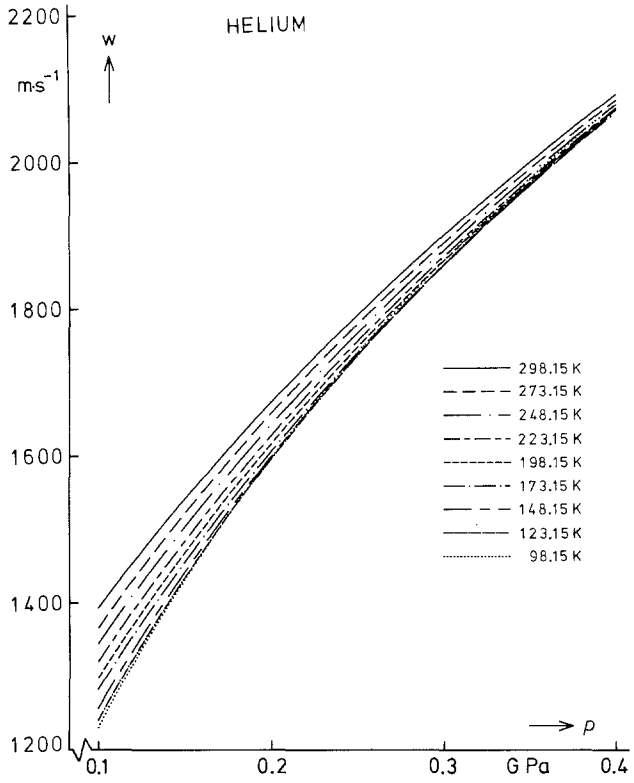

Fig. 2. Sound velocity in helium as a function of pressure up to 0.4 GPa, at several temperatures.

the fit is excellent. The isotherms of 98.15, 123.15, and 148.15 K terminate at 790, 900, and 960 MPa, respectively, due to experimental difficulties with the pressure vessel.

From the density (ρ) and the sound velocity (w), one obtains the adiabatic compressibility χ_S ($\equiv 1/\rho w^2$). In order to calculate the derivative of the density with respect to pressure, to obtain the isothermal compressibility χ_T $[\equiv(1/\rho)(\partial\rho/\partial p)_T]$, we fitted the data to an empirical equation, which also was applied successfully to our compressibility data for argon and for nitrogen:

$$\rho = A + Bp^{-1} + Cp^{-2} + Dp^{-m} \tag{1}$$

The values of the parameters, as found from a least-squares analysis, are $A = -1.898471 \times 10^2$, $B = 1.1739284 \times 10^3$, $C = -1.3849161 \times 10^4$, $D =$

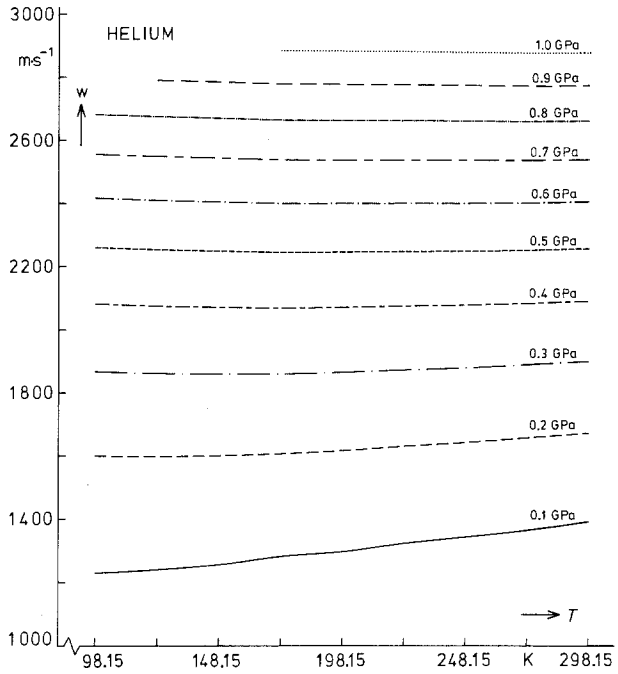

Fig. 3. Sound velocity in helium as a function of temperature at round values of the pressure.

1.0864384×10^2, and $m = -0.1406$, where p is expressed in MPa and ρ is in $\text{kmol} \cdot \text{m}^{-3}$. The standard deviation is 0.03%, but th deviation is 0.03%, but the deviations below 500 MPa are not randomly distributed. The ratio of χ_T over χ_S provides γ, the ratio of specific heats ($\gamma = \chi_T/\chi_S = C_p/C_V$). In Table I the data at 298.15 K and at round values of the pressure with intervals of 50 MPa are shown. The ratio of specific heats γ is a monotonously decreasing function of pressure. The sound velocities for all isotherms at round values of the pressure are given in Table II.

4. COMPARISON WITH PREVIOUS WORK

4.1. Compressibility

For the results from 20 to 68 MPa, we initially used the data of Michels and Wouters [20] as reference data for densities below 20 MPa. It turned out that our results were not consistent with these data. For example, the density that we obtain from a total expansion with an initial pressure of 20 MPa (and a pressure after expansion of about 8.5 MPa) is

Table I. Thermodynamic Properties of Helium at 298.15 K

p (MPa)	ρ (kmol · m^{-3})	w (m · s^{-1})	$\chi_T 10^5$ (MPa^{-1})	$\chi_S 10^5$ (MPa^{-1})	γ
100	28.104	1393.35	719.58	457.90	1.571
150	37.134	1542.77	435.79	282.67	1.542
200	44.497	1674.74	303.24	200.19	1.515
250	50.755	1793.45	228.12	153.04	1.491
300	56.187	1901.57	180.72	122.97	1.470
350	60.991	2001.02	148.47	102.30	1.451
400	65.293	2093.24	125.28	87.33	1.435
450	69.184	2179.31	107.91	76.03	1.419
500	72.745	2260.10	94.47	67.24	1.405
550	76.048	2336.29	83.79	60.19	1.392
600	79.129	2408.44	75.13	54.43	1.380
650	81.989	2477.01	67.97	49.66	1.369
700	84.699	2542.37	61.97	45.64	1.358
750	87.268	2604.85	56.88	42.19	1.348
800	89.667	2664.73	52.51	39.24	1.338
850	91.988	2722.24	48.72	36.65	1.329
900	94.199	2777.58	45.40	34.38	1.321
950	96.264	2830.94	42.48	32.38	1.312
1000	98.264	2882.46	39.89	30.60	1.304

Table II. Velocity of Sound (in m · s^{-1}) in Helium at Various Pressures (in MPa)

	T (K)								
p (MPa)	298.15	273.15	248.12	223.17	198.15	173.15	148.15	123.10	98.10
100	1393.35	1368.80	1344.92	1320.50	1297.84	1282.32	1256.36	1240.14	1229.98
150	1542.77	1523.13	1504.81	1485.66	1470.01	1457.02	1443.67	1435.79	1433.14
200	1674.74	1659.57	1644.28	1629.59	1618.51	1608.28	1601.40	1598.11	1599.69
250	1793.45	1781.09	1768.60	1757.39	1749.50	1741.66	1738.48	1737.98	1742.00
300	1901.57	1891.16	1881.15	1872.56	1867.01	1861.11	1860.24	1861.54	1866.97
350	2001.02	1992.10	1984.21	1977.57	1973.80	1969.42	1970.12	1972.62	1978.84
400	2093.24	2085.52	2079.42	2074.23	2071.85	2068.67	2070.48	2073.81	2080.44
450	2179.31	2172.62	2168.03	2163.93	2162.64	2160.39	2163.04	2166.93	2173.74
500	2260.10	2254.29	2250.97	2247.70	2247.28	2245.75	2249.05	2253.33	2260.18
550	2336.29	2331.26	2329.00	2326.38	2326.64	2325.69	2329.49	2334.03	2340.84
600	2408.44	2404.10	2402.73	2400.63	2401.43	2400.92	2405.13	2409.83	2416.55
650	2477.01	2473.28	2472.64	2470.99	2472.20	2472.05	2476.58	2481.36	2487.98
700	2542.37	2539.18	2539.16	2537.90	2539.42	2539.56	2544.34	2549.15	2555.66
750	2604.85	2602.13	2602.62	2601.72	2603.48	2603.84	2608.81	2613.61	2620.01
800	2664.73	2662.41	2663.33	2662.78	2664.69	2665.24	2670.35	2675.10	2681.41
850	2722.24	2720.26	2721.53	2721.33	2723.33	2724.04	2729.24	2733.91	
900	2777.58	2775.89	2777.44	2777.61	2779.63	2780.49	2785.73	2790.30	
950	2830.94	2829.47	2831.25	2831.81	2833.81	2834.79	2840.04		
1000	2882.46	2881.16	2883.13	2884.11	2886.03	2887.13			

systematically lower than the density reported by Michels and Wouters at 20 MPa. The discrepancy is larger than can be explained by the errors in our procedure. It proves, in our opinion, that Michels and Wouters' results are not consistent within the stated accuracy. However, our data were found to be consistent with those of Briggs. A comparison of the results of this author with those of Michels and Wouters shows that the relative deviations of Michels and Wouters' data increase almost linearly from 0 at $p=0$ to 0.18% at 20 MPa. Michels and Wouters used a glass tube piezometer and probably there was a loss of helium due to diffusion through the glass wall. Therefore, in this work the results of Briggs have been taken for the low-pressure densities.

In Fig. 4, the relative deviations of the data of Mills et al. [8] and of Vidal et al. [7] from our data are shown. Both Mills et al. and Vidal et al. report an accuracy in the molar volume, e.g. the density of 0.3%. The data of Mills used for the comparison are calculated from their equation of state, which describes the experimental molar volumes with an accuracy of 0.5%. The deviation curve is similar as in the case of nitrogen [18]. According to the deviation plot of the authors, the experimental data at 295 K and 200 MPa are about 0.5% lower than the equation of state predicts, so the difference between their experimental values and ours is smaller. The agreement at higher pressures is fair. The agreement with the

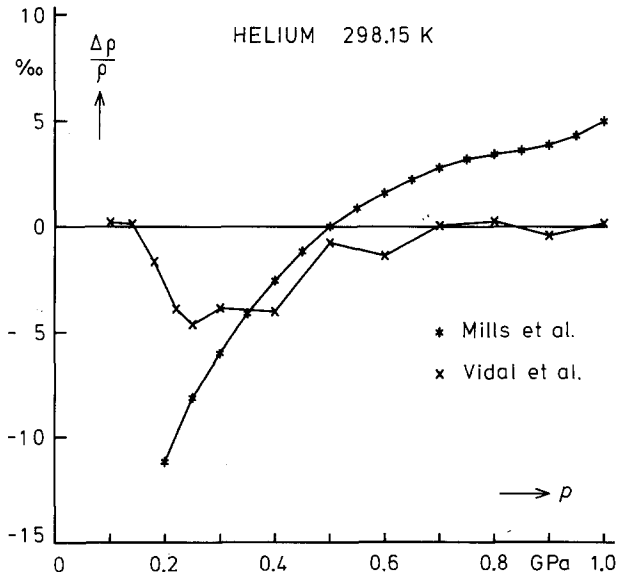

Fig. 4. Comparison between previous and the present density data at 298.15 K.

data of Vidal et al. is excellent, except in the region of 200 to 400 MPa. A similar deviation is found for the sound velocity, as discussed in the next section. We were not able to compare our results directly with those of Tsiklis et al. [6], because their isotherms are at 293, 323, 373, and 423 K. Compared to the data of Mills et al. at 293 K, the densities at 200 MPa of Tsiklis et al. are 0.2% lower and the difference increases to 2% at 700 MPa. The estimated deviation between Tsiklis and co-workers' densities and our data lies accordingly between 0.7% at lower pressures and 1.5% at higher pressures. We compared the predicted values for the density from the equation of state given by Gammon [12] with our experimental values. The deviations increase from less than 0.1% at 100 MPa to 4% at 1 GPa. Taking in to account the fact that the equation-of-state parameters were adjusted to fit velocity-of-sound data at pressures up to 15 MPa, the agreement is surprisingly good.

4.2. Sound Velocity

A comparison of the present results for sound velocities with those of other authors is shown in Fig. 5. The agreement with the data of Vidal et al. is again excellent, but also here we see the largest deviations between 200 and 400 MPa. Since in our case, separate experimental setups were

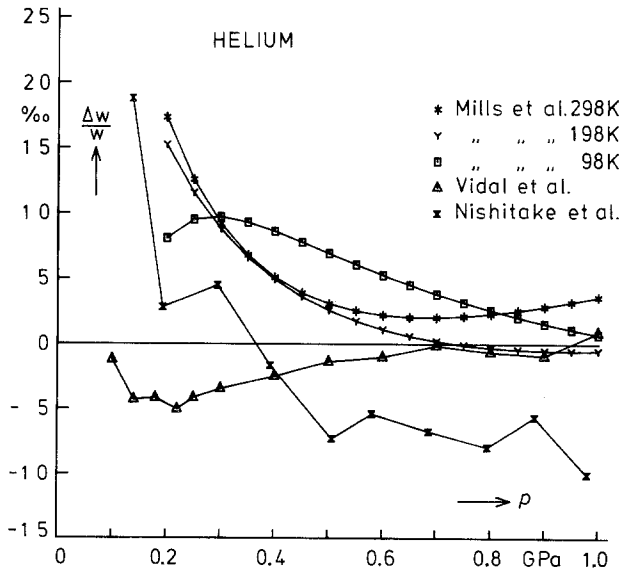

Fig. 5. Comparison between previous and the present sound-velocity data.

used to determine the compressibility and the sound velocity, we do not expect this deviation to be due to our equipments. From the equation of state given by Mills et al., we calculated the corresponding sound-velocity isotherms. The deviations are shown for only three isotherms but the deviation curves of the other isotherms show a similar shape. The agreement with Nishitake and Hanayama [11] is, except for the lowest pressure, within the 1% margin claimed by the authors, although the deviation curve shows a strong systematic tendency. A comparison with the data of Pitaevskaya and Bilevich [10] is not shown but their values are 2% lower at 100 MPa and the difference decreases steadily to 0.3% at 400 MPa. No comparison is made with Vereshchagin et al. [9] because numerical data are not given by these authors.

4.3. Ratio of Specific Heats

In Fig. 6 the values of γ, the ratio of specific heats C_p/C_V as a function of pressure, are shown. No smoothing procedure is applied to the present data. The data of Vidal et al. are in close agreement, as would be expected from the comparisons of density and sound velocity. The values of Mills et al. are systematically higher. The data of Nishitake and Hanayama [11] show large deviations.

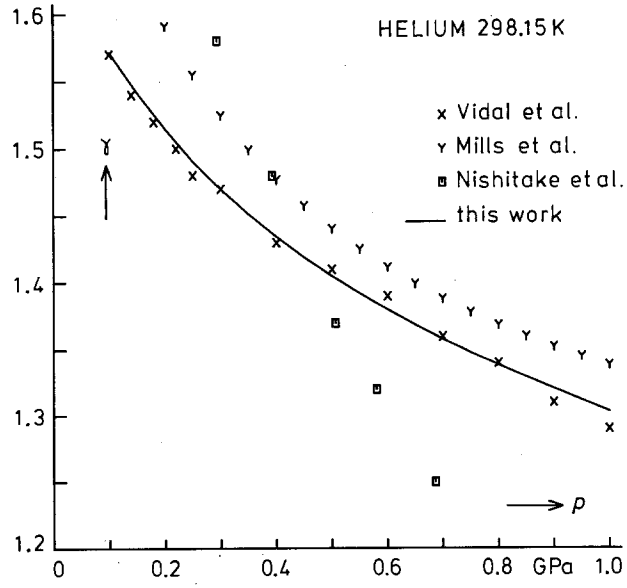

Fig. 6. Comparison between previous and the present data for γ, the ratio of specific heats.

5. ANALYSIS OF THE RESULTS

From the fact that the sound velocity increases with increasing temperature, the conclusion can be drawn that fluid helium has a gas-like character, even at pressures up to 1 GPa for higher temperatures and up to 200 MPa at 98 K. The isobaric slope of the sound velocity $(\partial w/\partial T)_p$ is positive for lower pressures, but the isobars show a minimum at higher pressures, as can be seen in Fig. 3. The velocity of sound is nearly temperature independent at higher pressures. A comparison with other simple gases is shown in Table III, in which the average relative change per degree kelvin in sound velocity, expressed by $\varepsilon = (1/w)(\Delta w/\Delta T)_p$, is given. Here $\Delta w = (w_{max} - w_{min})/w_{min}$, and w_{max} and w_{min} are the maximum and minimum values of the sound velocity along the isobar. In the case of helium these values do not necessarily lie at the end points of the temperature interval. Since the temperature intervals ΔT of the isobars may differ, the relative change is divided by ΔT. Due to the presence of the melting line, no sufficient data are available for argon along the 1000-MPa isobar up to 298.15 K. At elevated pressures the temperature dependence for helium is about 10 times smaller than for the other light noble gases.

Our data do not support the conclusion of Mills et al. that along an isotherm the product of the molar volume V and the sound velocity w is almost constant; e.g., for 298.15 K and 100 MPa the product $wV = 49.6$ $m^4 \cdot kmol^{-1} s^{-1}$, whereas at 1 GPa the value has decreased to 29.4 $m^4 \cdot kmol^{-1} \cdot s^{-1}$. But the deviations from Rao's rule, expressed by $Vw^{1/3} =$ constant, as well as the free volume model, $wV^{1/3} =$ constant, are even larger. Kimura et al. [21] proposed a modification of the sound velocity equation for an ideal gas, $w = (\gamma_0 pV/M)^{1/2}$, in the form $w = (\gamma_0 pV/M + b(V_0/V)^2)^{1/2}$, where $\gamma_0 = 1.67$, M is the molecular weight, and V_0 is the molar volume at atmospheric pressure. While deviations between the sound velocity calculated by the ideal-gas law and the experimental values range from 18 to 35%, the largest deviation of the modified equation is about 1.2% and is found at the lowest pressure (100 MPa). The deviations are positive and decrease to about 0.3% with increasing pressure up to 1 GPa

Table III. The Relative Change of the Sound Velocity per Degree K, $\varepsilon \times 10^4$, Along Several Isobars

p (MPa)	He	Ne	Ar	N_2	CH_4
100	6.64	4.35	18.3	19.6	24.3
500	0.321	4.22	5.32	5.89	6.25
1000	0.165	2.47	—	3.11	3.66

but do not show a random distribution. In the low-pressure limit, the modified equation converts to the ideal-gas situation. We tried a new modification, in which V_0/V is replaced by $(V_0 - V)/V$, resulting in a correction term on the ideal gas law: $b[(V_0 - V)/V]^2$. Still, the corrections at lower pressures are overestimated. Further improvement was obtained by replacing the integer value of the power (2) by a noninteger value, m. The best result was obtained with the value $m = 1.74$. The maximum deviations are reduced to within 1% over the full pressure range and a standard deviation of 0.23%, but the deviations are still not randomly distributed. Because in our case the compressibility and the sound velocity were not measured at exactly the same pressure, smoothing by interpolation cannot be avoided. Although we believe that our data are accurate, this situation is not suitable for the examination of an improved isothermal relation between sound velocity and density.

6. SECONDARY PRESSURE STANDARD

As stated above, the temperature dependence of the sound velocity in helium at elevated pressures is very small. The averaged increase of the sound velocity with pressure is about 0.15–0.20 $\mathrm{m \cdot s^{-1}}$ per 0.1 MPa. This is well within the experimental possibilities. These facts would make the sound velocity in helium very suitable to serve as a secondary pressure standard, the more so as helium is widely used as a pressure medium.

7. EQUATION OF STATE

In the past, several equations have been proposed for helium. Some of these equations have an empirical nature, whereas others are based on theoretical assumptions, concerning the interaction of the molecules. An example of the first type is Eq. (1). An interesting equation of the latter type is derived by Hansen [22] and applied to the group of noble gases by Vidal et al. [7]. In the calculation of the free energy for a hot Lennard-Jones fluid ($T^* = kT/\varepsilon > 2.5$; for helium $T > 26$ K), the contribution of the repulsive part of the potential and the first-order $1/T$ term of the attractive part (considered as a weak perturbation) were calculated numerically.

The results were fitted by a polynomial expressing the reduced pressure $p^* = p\sigma^3/\varepsilon$ in terms of the reduced density $\rho^* = N\rho\sigma^3$, N being Avogadro's number:

$$\begin{aligned} p^* = {} & \rho^* T^* + \rho^{*2}(B_1 T^{*3/4} - C_1 T^{*1/4}) + \rho^{*3}(B_2 T^{*1/2} - 2C_2) \\ & + \rho^{*4}(B_3 T^* 1/4 - 3C_3 T^{*-1/4}) + \rho^{*5}(B_4 - 4C_4 T^{*-1/2}) \\ & - \rho^{*6}(5C_5 T^{*-3/4}) + \rho^{*11} B_{10} T^{*-3/2} \end{aligned} \tag{2}$$

where $B_1 = 3.629$, $B_2 = 7.2641$, $B_3 = 10.4924$, $B_4 = 11.459$, $B_{10} = 2.17619$, $C_1 = 5.3692$, $C_2 = 6.5797$, $C_3 = 6.1745$, $C_4 = -4.2685$, and $C_5 = 1.6841$. The parameters of the Lennard-Jones potential, given by Hirschfelder et al. [23], are $\sigma = 2.556 \times 10^{-10} m$ and $\varepsilon/k = 10.22$ K. The calculated values of the density are systematically about 1.7% higher than the experimental values. In a subsequent article a more detailed comparison of the performance of the equation of state of Hansen with respect to a series of gases (He, Ne, Ar, N_2, and CH_4) at 298 K will be given and, also, other equations of state will be tested. It is interesting to compare the experimental velocity-of-sound data with the values predicted by the equation of state. We calculated the sound velocity using the relation

$$w^2 = 1/\{M[(\partial\rho/\partial p)_T - T(\partial\rho/\partial T)_p^2/\rho^2 C_p(p, T)]\} \tag{3}$$

Here we did not calculate the $C_p(p, T)$ values by using values for the thermal expansion coefficient α_p reported in the literature, as was done by Vidal et al., but instead, we used the data obtained from the equation of state of Mills et al., which are valid above 200 MPa. The change of C_p is only a few percent over the pressure range from 0.2 to 1 GPa. At 298 K, the predicted values for the sound velocity exceed the experimental values by about 0.8% at 200 MPa, increasing to 8% at 1 GPa. At lower temperatures the deviations increase steadily and at 98 K values are reached of 5% at 200 MPa and 15% at 800 MPa. The conclusion can be drawn that, at lower pressures, the equation of state predicts reasonable values for the sound velocity but that, at higher pressures, the increase in sound velocity is overestimated. The velocity of sound proves to be a very sensitive quantity for testing an equation of state.

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