

Compressibility and Sound Velocity Measurements on N₂ up to 1 GPa

P. J. Kortbeek,¹ N. J. Trappeniers,¹ and S. N. Biswas¹

Received July 31, 1987

A gas expansion technique has been used to determine the pVT properties of N₂ up to 1 GPa at 298.15 K, with an accuracy of 0.08% in density, 1 mK in temperature, and 0.05% + 0.2 MPa in pressure. The sound velocity has been measured by a phase-comparison pulse-echo technique between 123 and 298 K at intervals of 25 K and at pressures up to 1 GPa, with an accuracy of better than 0.02% in sound velocity, 10 mK in temperature, and 0.05% + 0.2 MPa in pressure. An equation of state is presented that correlates the density data over the wide pressure range of 36–1000 MPa with maximum deviations between the calculated and the experimental densities of less than 0.05%.

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

1. INTRODUCTION

In this paper, the experimental results on the thermodynamic properties of nitrogen (N₂) up to 1 GPa are presented. Nitrogen, a diatomic molecular fluid, is of interest both for technical applications and for theoretical calculations. Many compressibility and sound velocity experiments have been carried out, pushing pressures higher and higher [1–14]. A survey of this work is given in Table I. The present study was undertaken to extend the range of very accurate density data from 200 up to 1000 MPa and because no sound velocity measurements at elevated pressures have been carried out previously at temperatures below 247 K. The availability of density data at higher pressures has led to the formulation of empirical equations of state [11, 15, 17]. More recently, the theoretical microscopic models for strictly spherical molecules such as the noble gases have been

¹ Van der Waals Laboratory, University of Amsterdam, Valckenierstraat 67, 1018 XE Amsterdam, The Netherlands.

Table I. Review of Experimental Data on Nitrogen at Elevated Pressures:
Density is Denoted by ρ and Sound Velocity by w

Reference	Temperature range, K (interval)	Pressure range (MPa)	Measured quantity
Kanishchev et al. [13]	298, 323, 373, 423	200–700	w
Kimura et al. [31]	295	300–2680	ρ, w
Lacam [4]	298	0–100	w
Malbrunot and Vodar [10]	473–1273 (100)	80–500	ρ
Michels et al. [1–3]	148–448 (25)	0–300	ρ
Mills et al. [11]	247–320 (25)	300–2200	ρ, w
Morris and Wylie [14]	253–283 (10), 298, 308	200–500	ρ
Nishitake and Hanayama [12]	298	300–1500	w
Robertson and Babb [9]	308, 373–673 (100)	200–1000	ρ
Tsiklis and Polyakov [7]	294, 453, 473–673 (100)	150–1000	ρ
Vereshchagin et al. [6]	298	0–350	w
Volarovich and Balashov [5]	293	0–500	w
Voronov et al. [8]	298–448 (25)	20–400	w
This work	298	200–1000	ρ
	123–298 (25)	85–1000	w

adapted for diatomic molecules [16]. From the field of molecular dynamics (MD) there is an increasing demand for equation-of-state data. The pressure of a MD system is a quantity which is extremely sensitive to the form of the potential. In particular, the low-pressure range of the system probes the attractive part of the potential, while the high-pressure range probes the repulsive part [24]. Furthermore, very accurate data in the moderate-pressure region (up to 1 GPa) are necessary, in order to establish a successful link between experimental data at moderate pressures and data at very high pressures as obtained from diamond anvil cell and shock-wave experiments. Also, in the study of mixtures of simple gases, a field with increasing popularity, a good knowledge of the behavior of the pure components is indispensable.

This work is a continuation of the study of acoustic and thermodynamic properties of simple gases from the Van der Waals laboratory at pressures up to 1 GPa.

2. EXPERIMENTAL

2.1. Compressibility

The compressibility data and the sound velocity data were determined in two separate experimental setups. The pVT data were measured using a

gas expansion technique, in which a gas is expanded from an initially pressurized vessel V_A into an evacuated pressure vessel V_B . In the case of a total expansion, the pressures after one expansion in both vessels are equal. In a stepwise expansion run, the gas is expanded step by step and the pressure in the expansion vessel is accumulated. At each stage, the pressure is recorded in both vessels and the densities at the high-pressure side (vessel V_A) are derived from the densities at the low-pressure side (vessel V_B), which are taken from the literature. An extensive description of the method will be given in a separate paper [19], in which data on argon will be reported. The equipment was also used for measurements on methane [20].

The required low-pressure data were taken from Michels et al. [1–3] and were determined up to 200 MPa with a piezometer, with an accuracy of 0.02 %. Using these data, the volume ratio V_B/V_A of the pressure vessels was determined following a low-pressure gas expansion method [19]. The value obtained agrees well with the value determined with argon, the difference being as small as 0.01 %. The average pressure distortion coefficients β_A and β_B [$\beta \equiv 1/V(\Delta V/\Delta p)_T$] of the pressure vessels were determined by calibration with argon up to 280 MPa, instead of nitrogen, the gas to be investigated, since the uncertainty of the compressibility data of nitrogen at pressures up to 200 MPa is twice that of argon.

2.2. Sound Velocity

The velocity of sound in nitrogen was measured by using the well-known phase-comparison pulse-echo technique, operating with one transducer and two reflectors at unequal distances. The pressure vessel containing the ultrasonic cell is part of a metal cryostat, in which temperature gradients can be reduced to 10 mK by using a cold finger system. The frequencies applied lie between 9.5 and 10.5 MHz. A detailed description of the apparatus and data on argon can be found elsewhere [21].

In both experiments nitrogen with a purity of 99.9995 %, purchased from Matheson Co., U.S.A., was used.

3. RESULTS

In total nine runs of stepwise expansions with 69 data points and an equal number of total expansion runs were taken. The consistency of the combined stepwise expansion runs is good, the maximum deviations being of the order of 0.03 %. The total expansion data scatter by about 0.05 %, but the values are systematically lower than those obtained by the stepwise

expansion runs by about 0.06%. This discrepancy is discussed in the next section.

The data are smoothed with a polynomial of the fourth degree in pressure, using all the data points. The standard deviation in the density, representing the random error, is less than 0.04% and the systematic error, caused by the uncertainties in the volume ratio and the distortion coefficients, is estimated to be 0.04%. This gives a total uncertainty of 0.08%. No corrections have been made for systematic errors in the data of Michels et al. [1-3], as no estimates are given by these authors. Their data are probably the most accurate ones in the lower-pressure region and our density values are based mainly on the densities at pressures below 75 MPa. The polynomial is used for the interpolation of the density to round values of the pressure and for the calculation of the isothermal compressibility χ_T [$\equiv 1/\rho (\delta\rho/\delta p)_T$].

The sound velocity measurements were carried out at pressures down to 85 MPa, where the mismatch of acoustic impedance between the quartz transducer and the gas puts a limit to the detection of the echoes. The

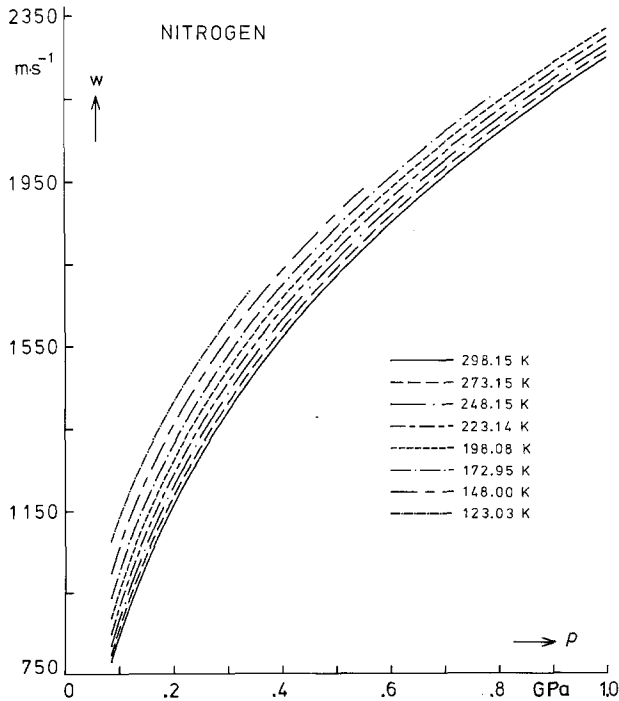


Fig. 1. Sound velocity in nitrogen as a function of pressure at several temperatures.

values were fitted to polynomials of the fifth degree in pressure for isotherms above 223 K and of fourth degree for temperatures between 148 and 198 K, whereas the 123 K isotherm was fitted to a polynomial of the third degree. The isotherms are shown in Fig. 1, whereas in Fig. 2 the isobars are displayed. From Fig. 2 it is clear that the experimental sound velocities decrease with increasing temperature, indicating that compressed nitrogen shows a liquid-like behavior within our pressure range.

From the density (ρ) and the sound velocity (w), one can easily calculate the adiabatic compressibility $\chi_s (\equiv 1/\rho w^2)$, whereas the ratio of χ_T over χ_s gives γ , the ratio of specific heats ($\gamma = \chi_T/\chi_s = C_p/C_v$). In Table II the data at 298.15 K and at round values of pressures at intervals of 50 MPa are shown. The densities and isothermal compressibilities below 200 MPa are taken from Michels et al. The ratio of specific heats γ is a monotonously decreasing function of pressure. The present data indicate that at 340 MPa the value of γ for fluid nitrogen has decreased to 7/5, which is the value of an ideal diatomic gas, and at 740 MPa to 9/7, the value of γ of an ideal triatomic gas. The sound velocities for all isotherms at round values of the pressure are tabulated in Table III.

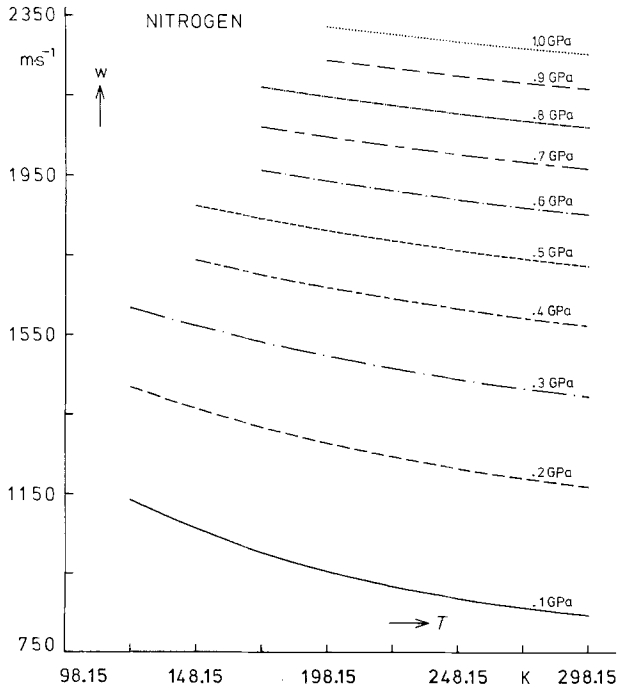


Fig. 2. Sound velocity in nitrogen as a function of temperature at several pressures.

Table II. Thermodynamic Properties of Nitrogen at 298.15 K

p (MPa)	ρ (kmol · m ⁻³)	w (ms ⁻¹)	$\chi_T \cdot 10^4$ (MPa ⁻¹)	$\chi_S \cdot 10^4$ (MPa ⁻¹)	γ
85	19.1490	780.82	51.0018	30.5761	1.668
100	20.4547	844.27	40.0701	24.4835	1.637
150	23.6455	1023.69	22.4815	14.4061	1.561
200	25.8569	1168.02	15.1843	10.1194	1.501
250	27.5906	1289.42	11.2756	7.7819	1.449
300	29.0118	1394.85	8.9814	6.3242	1.420
350	30.2209	1488.54	7.4440	5.3310	1.396
400	31.2766	1573.20	6.3468	4.6116	1.376
450	32.2159	1650.68	5.5265	4.0667	1.359
500	33.0638	1722.29	4.8912	3.6397	1.344
550	33.8381	1788.99	4.3853	3.2962	1.330
600	34.5514	1851.53	3.9733	3.0137	1.318
650	35.2137	1910.47	3.6314	2.7774	1.307
700	35.8324	1966.27	3.3433	2.5767	1.297
750	36.4134	2019.30	3.0973	2.4042	1.288
800	36.9616	2069.85	2.8848	2.2543	1.280
850	37.4809	2118.18	2.6995	2.1227	1.272
900	37.9744	2164.51	2.5366	2.0064	1.264
950	38.4450	2209.01	2.3921	1.9028	1.257
1000	38.8948	2251.84	2.2632	1.8099	1.250

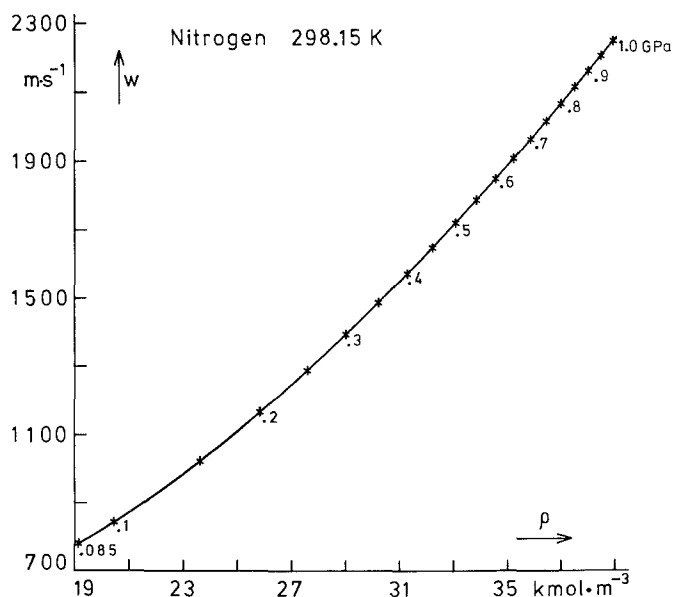
In Fig. 3, the sound velocity against the density is plotted at 298.15 K. In an attempt to correlate the data, it turns out that a polynomial of the type $w = \sum_{i=0}^3 a_i \rho^i$ gives better results than a power law of type $w = b\rho^c + d$. From a least-squares procedure we obtained the following values for the polynomial: $a_0 = 6.387434 \times 10^2$, $a_1 = -3.8142556 \times 10^1$, $a_2 = 2.699940$, and $a_3 = -1.6787661 \times 10^{-2}$, the standard deviation being 0.02% in density and 0.03% in sound velocity. In the case of the power law the following values are obtained: $b = 1.6225$, $c = 1.9414$, $d = 275.25$, and the standard deviation in sound velocity = 0.15%. Because the two quantities, density and sound velocity, are not measured simultaneously, some smoothing effects by the interpolating polynomials in pressure may occur.

As mentioned above, a systematic deviation was found between the densities obtained by stepwise expansions and those obtained by total expansions. This discrepancy was not observed in our previous measurements, either on argon [19] or on methane [20]. Two possible explanations can be given.

The first is that the discrepancy is caused by adsorption/absorption of nitrogen on/into the walls of the pressure vessels or the connecting

Table III. Velocity of Sound (in m·s⁻¹) in N₂ at Various Pressures (in MPa)

<i>p</i> (MPa)	T (K)							
	298.15	273.15	248.15	223.14	198.08	172.95	148.00	123.03
85	780.82	796.78	819.47	848.78	888.16	937.96	998.85	1075.41
100	844.27	862.93	886.74	916.84	955.06	1002.93	1063.44	1134.35
150	1023.69	1045.16	1069.63	1099.88	1135.84	1179.02	1231.78	1292.89
200	1168.02	1189.42	1213.79	1243.04	1277.04	1316.94	1363.45	1418.32
250	1289.42	1310.27	1334.47	1362.53	1394.66	1432.01	1474.67	1524.39
300	1394.85	1415.14	1439.07	1465.98	1496.42	1531.68	1571.92	1617.41
350	1488.54	1508.34	1531.87	1557.72	1586.68	1620.17	1658.65	1700.90
400	1573.20	1592.58	1615.59	1640.48	1668.15	1700.12	1737.05	
450	1650.68	1669.68	1692.09	1716.11	1742.67	1773.31	1808.60	
500	1722.29	1740.95	1762.69	1785.92	1811.52	1840.99	1874.42	
550	1788.99	1807.32	1828.38	1850.89	1875.66	1904.07	1935.33	
600	1851.53	1869.54	1889.91	1911.76	1935.80	1963.26		
650	1910.47	1928.17	1947.86	1969.10	1992.49	2019.10		
700	1966.27	1983.65	2002.70	2023.39	2046.19	2072.02		
750	2019.30	2036.34	2054.82	2074.99	2097.25	2122.37		
800	2069.85	2086.56	2104.53	2124.22	2145.97	2170.44		
850	2118.18	2134.56	2152.09	2171.33	2192.59			
900	2164.51	2180.53	2197.72	2216.54	2237.33			
950	2209.01	2224.68	2241.60	2260.03	2280.35			
1000	2251.84	2267.14	2283.91	2301.95	2321.82			

**Fig. 3.** Sound velocity in nitrogen as a function of density at 298.15 K.

capillaries or in the valve sealings. If it were possible to determine the pressure distortion coefficients of the pressure vessels with sufficient accuracy for nitrogen as a pressure medium, the effects of sorption would be incorporated to some extent in these coefficients.

The second is that nitrogen, possibly by dissolving in the manganin wire, may influence the reading of a manganin pressure gauge, as has been reported previously [7, 18]. With a recently developed 1-GPa differential pressure null indicator as a phase separator, it is now possible to calibrate a manganin gauge with nitrogen as pressure medium, against a 1.5-GPa controlled-clearance pressure balance. In this way, the effect of nitrogen on a manganin gauge can be investigated. Preliminary results do not seem to support this possibility, however.

4. COMPARISON WITH PREVIOUS WORK

4.1. Compressibility

The density data are compared with those reported in the literature. In Fig. 4 the relative deviations are shown. The data of Morris and Wylie [14] are systematically higher by about 0.06%, which is within our accuracy limit. Our results support their conclusion that the density data of Mills et al. [11] increase too rapidly with increasing pressure. The difference at 1 GPa amounts to 0.7%, which exceeds a few times the accuracy claimed by these authors. Considering that the data between 200 and 600 MPa presented by Lunbeck et al. [3] were obtained by extrapolation of the experimental data below 200 MPa, a deviation of 1.5% at the highest pressure is still not too bad. A comparison with other authors is

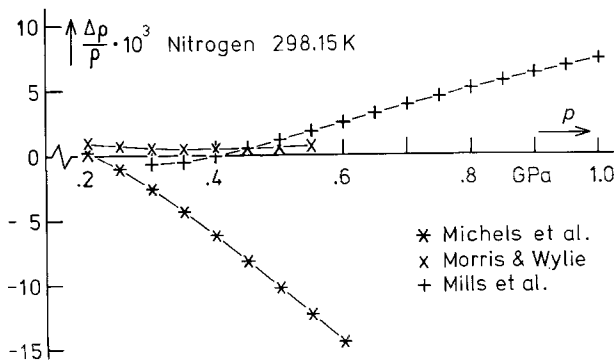


Fig. 4. Comparison between previous and present density data at 298.15 K.

not realistic, because the experimental temperatures are different and temperature corrections on the density are not possible with sufficient accuracy.

4.2. Sound Velocity

The comparison of the present data for sound velocities and the previous data is shown in Figs. 5 and 6. Our values do not agree with those of Lacam [4] within 1% or with the data of Voronov et al. [8] within 0.3%. Except at the lowest pressure, the agreement with the data of Nishitake and Hanayama [12] is within the stated 1% error margin, although the data scatter rather a lot. For all temperatures that are compared, the data of Mills et al. [11] show a remarkably constant deviation, which is in contrast with the comparison of the densities. The fact that these authors found systematic deviations between the experimental sound velocities and those calculated from the equation of state may be due to the systematic error in the molar volume determinations. A comparison with the data of Kanishchev et al. [13] is not given, because we suspect some error in their tabulated coefficients. Even after adjustment, deviations of about 1% from our data at 300 MPa are found and the values do not coincide with those of Mills, as stated by the authors. Below 300 MPa the present data are compared with those calculated from pVT data of Michels et al. [2, 3] and the agreement is fair below 200 MPa, but the deviations increases very rapidly above this pressure. It should be noted

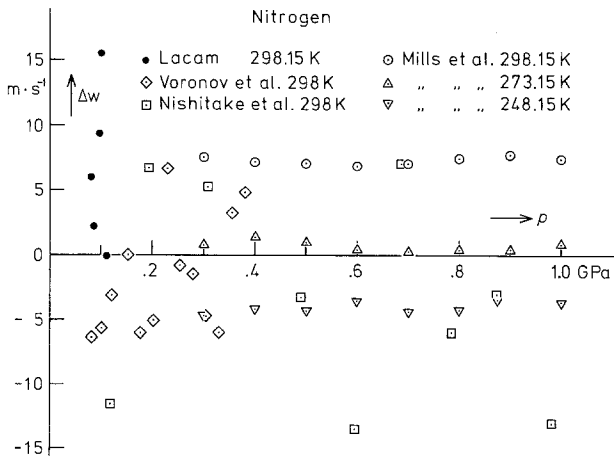


Fig. 5. Comparison between previous and present sound velocity data up to 1 GPa.

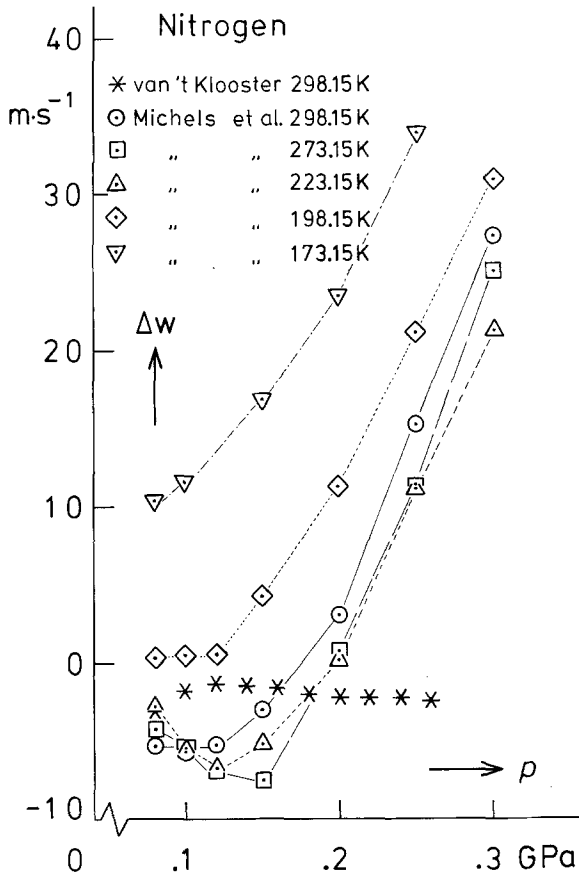


Fig. 6. Comparison between previous and present sound velocity data below 300 MPa.

that the sound velocities calculated by Michels et al. at temperatures below 273.15 K are based on pVT data obtained by Benedict [32]. The agreement with the data of van 't Klooster [22] is excellent. These measurements were performed in an apparatus, operating at 2 MHz, that was designed especially for the investigation of the sound velocity in liquids [23]. The ultrasonic frequencies at which the reported sound velocity data were measured range from 900 kHz [4] to 30 MHz [11] and no indications for anomalous dispersion are found within the experimental range.

5. ANALYSIS OF THE COMPRESSIBILITY DATA

A number of empirical equations of state have been proposed in the past to describe the volumetric behavior of gases as a function of pressure and temperature. Some of these equations are well known in the literature, namely, the Redlich–Kwong equation, the Beattie–Bridgeman equation, and the Benedict–Webb–Rubin equation. Others are less well known, such as the Becker equation, although this was previously used by Bridgman to represent the equation-of-state data of nitrogen at high pressures [25]. However, it was previously shown by Robertson et al. [26] and Malbrunot [18] that most of these empirical equations of state are not capable of representing the compressibility data of a gas at pressures up to 1000 MPa with any reasonable degree of precision. Recently, a very simple equation of state in the form

$$\sqrt{\rho} = a + b \ln p \quad (1)$$

in which a and b are adjustable parameters, has been suggested by Antanovich [17] for pressures above 200 MPa. However, it is found that the relative standard deviation of a least-squares fit of the present data to this equation is 0.7%, which is nearly an order of magnitude larger than the accuracy of the present measurements.

In the absence of a suitable empirical equation of state at pressures up to 1000 MPa and in view of the importance of such an equation for smoothing and interpolating the experimental data as well as for calculating thermodynamic properties, it is considered worthwhile to examine whether the equations of state normally used for describing liquid densities are applicable for representing the present data for nitrogen at high densities. Accordingly, the present data are fitted to the well-known Tait equation, expressed in the form

$$\rho = \rho_0 + C \ln(1 + p/B) \quad (2)$$

where B and C are parameters which depend only on the temperature. It turns out, however, that the deviations between calculated and experimental densities are not random but smooth functions of the pressure and that the relative standard deviation is about 0.2%, which is several times the estimated error of the present measurement. Thus the Tait equation, having the advantage of simplicity, may be used for the purpose of interpolation if use is made of an appropriate deviation curve.

The best analytical representation of the experimental data for nitrogen at 298.15 K and over the wide pressure range of 36–1000 MPa

(which include the present data and those reported by Michels et al. [1]) is achieved by the following five-parameter equation:

$$\rho = A + B(1/p) + C(1/p)^2 + D(1/p)^m \quad (3)$$

in which ρ is the density in $\text{kmol} \cdot \text{m}^{-3}$, p is the pressure in MPa, and A , B , C , D , and m are adjustable parameters. The values of these parameters, as found from a least-squares analysis, are as follows: $A = -1.8323751 \times 10^1$, $B = -2.3498275 \times 10^2$, $C = 2.2397355 \times 10^3$, $D = 2.0728567 \times 10^1$, and $m = -1.4754851 \times 10^{-1}$. The maximum relative deviations between the calculated and the experimental densities are found to be less than 0.05%, which is within the accuracy of the present measurements.

In the past, several theoretical equations of state for dense fluids were developed, which are based on a noninteracting rigid-sphere model and which describe only the effect of repulsive forces. These include the equations of Thiele and Wertheim [27] obtained from a closed-form solution of the Percus–Yevick equations, the equation of Ree and Hoover [28] developed by using the Padé approximation, and the Carnahan–Starling equation derived from considerations of the nearly geometric form of the virial series for rigid spheres [29]. As can be expected, none of these equations is found to describe the experimental data at high densities within the precision of the measurements; and the relative deviations even at higher densities are not less than about 6% in each case. Therefore, an attempt is made to fit the experimental data for nitrogen at 298.15 K covering the pressure range of 36–1000 MPa with the following semiempirical equation:

$$Z = pV/RT = (1 + y + y^2 - y^3)/(1 - y)^3 + aV^{-1} + bV^{-m} \quad (4)$$

where a , b , and m are adjustable parameters and V is the volume in $\text{m}^3 \cdot \text{kmol}^{-1}$. This equation was successfully used recently for describing the equation-of-state data of argon up to 1000 MPa [19]. Here, the first term on the right side is the contribution of repulsive forces as given by Carnahan and Starling and the remaining two terms (of which aV^{-1} is a Van der Waals type of term) describe the contribution due to attractive forces. The quantity y is the packing fraction defined by

$$y = (1/6) \pi d^3 (N/V) \quad (5)$$

N being Avogadro's number and d the diameter of the rigid sphere. It turns out from a least-squares analysis that the best fit is obtained with $a = -3.6011430 \times 10^{-2}$, $b = 1.6292193 \times 10^{-6}$, $m = 4.15$, and $d = 3.072 \times 10^{-10} \text{m}$. The rigid-sphere diameter obtained from the present fit is found to be in

reasonable agreement with those evaluated from the transport coefficients [30]. The relative standard deviations in Z are mostly less than 0.15% at higher densities ($p > 200$ MPa), which is about the experimental error of the present measurements and 0.25% at lower densities.

ACKNOWLEDGMENTS

The authors are particularly indebted to Mr. J. J. van de Ridder, who carried out the compressibility measurements, to Professor C. A. ten Seldam for his help with the computer calculations, and to Dr. J. A. Schouten for many valuable discussions. This paper is the 330th publication of the Van der Waals Fund.

REFERENCES

1. A. Michels, H. Wouters, and J. de Boer, *Physica* **3**:535 (1936).
2. A. Michels, R. J. Lunbeck, and G. J. Wolkers, *Physica* **17**:801 (1951).
3. R. J. Lunbeck, A. Michels, and G. J. Wolkers, *Appl. Sci. Res.* **A3**:197 (1952).
4. A. Lacam, *J. Phys. Rad.* **14**:351 (1953).
5. M. P. Volarovich and D. B. Balashov, *Primenenie Ultra Akoustiki K Issled Veschestva* **13**:63 (1961) (Russian).
6. L. F. Vereshchagin, N. A. Yuzefovich, and A. V. Chelovskii, *Sov. Phys. Doklady* **7**:541 (1962).
7. D. S. Tsiklis and E. V. Polyakov, *Sov. Phys. Doklady* **13**:901 (1968).
8. F. F. Voronov, L. L. Pitaevskaya, and A. V. Bilevich, *Russ. J. Phys. Chem.* **43**:321 (1969).
9. S. L. Robertson and S. E. Babb, Jr., *J. Chem. Phys.* **50**:4560 (1969).
10. P. Malbrunot and B. Vodar, *Physica* **66**:351 (1973).
11. R. L. Mills, D. H. Liebenberg, and J. C. Bronson, *J. Chem. Phys.* **63**:1198 (1975).
12. T. Nishitake and Y. Hanayama, *J. Phys. Soc. Jap.* **39**:1065 (1975).
13. B. E. Kanishchev, L. L. Piteavskaya, and S. L. Gutman, *Sov. Phys. Doklady* **26**:418 (1981).
14. E. L. Morris and R. G. Wylie, *J. Chem. Phys.* **79**:2982 (1983).
15. R. T. Jacobsen and R. B. Stewart, *J. Phys. Chem. Ref. Data* **2**:757 (1973).
16. G. I. Kerley and J. Abdallah, Jr., *J. Chem. Phys.* **73**:5337 (1980).
17. A. A. Antanovich, *J. Eng. Phys. (U.S.A.)* **40**:449 (1981).
18. P. Malbrunot, *Experimental Thermodynamics, Vol. III*, B. Le Neindre and B. Vodar, eds. (Butterworths, London, 1975), Chap. 8.
19. S. N. Biswas, N. J. Trappeniers, P. J. Kortbeek, and C. A. ten Seldam, *Rev. Sci. Instr.* (in press).
20. P. J. Kortbeek, S. N. Biswas, and N. J. Trappeniers, *Physica* **139/140B**:109 (1986).
21. P. J. Kortbeek, M. J. P. Muringer, N. J. Trappeniers, and S. N. Biswas, *Rev. Sci. Instr.* **56**:1269 (1985).
22. P. van 't Klooster, Private communication.
23. M. J. P. Muringer, N. J. Trappeniers, and S. N. Biswas, *Phys. Chem. Liq.* **14**:273 (1985).
24. R. Vogelsang and C. Hoheisel, *Phys. Chem. Liq.* **16**:189 (1987).
25. P. W. Bridgman, *Proc. Am. Acad.* **59**:173 (1923).
26. S. L. Robertson, S. E. Babb, Jr., and G. J. Scott, *J. Chem. Phys.* **50**:2160 (1969).

27. E. Thiele, *J. Chem. Phys.* **39**:474 (1963).
28. F. H. Ree and W. G. Hoover, *J. Chem. Phys.* **40**:939 (1964).
29. N. F. Carnahan and K. E. Starling, *J. Chem. Phys.* **51**:635 (1969).
30. J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids* (John Wiley & Sons, New York, 1964), p. 545.
31. M. Kimura, Y. Hanayama, and T. Nishitake, *Jap. J. Appl. Phys.* **26**:1366 (1987).
32. M. Benedict, *J. Am. Chem. Soc.* **59**:2224 (1937).