

A Reference Quality Equation of State for Nitrogen¹

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A new formulation describing the thermodynamic properties of nitrogen has been developed. New data sets which have been used to improve the representation of the p - ρ - T surface of gaseous, liquid and supercritical nitrogen, including the saturated states are now available. New measurements on the speed of sound from spherical resonators have been used to improve the accuracy of caloric properties in gaseous and supercritical nitrogen. State-of-the-art algorithms for the optimization of the mathematical structure of the equation and special functional forms for an improved description of the critical region were used to represent even the most accurate data within their experimental uncertainty. The uncertainty in density of the new reference equation of state ranges from $\pm 0.01\%$ between 270 and 350 K at pressures less than 12 MPa, within $\pm 0.02\%$ over all other temperatures less than 550 K and pressures less than 12 MPa, and up to a maximum of $\pm 0.6\%$ at the highest pressures. The equation is valid from the triple point to temperatures of 1000 K and pressures up to 2200 MPa. The new formulation yields a reasonable extrapolation up to the limits of chemical stability of nitrogen as indicated by comparison to experimental shock tube data. Constraints regarding the structure of the equation ensure reasonable extrapolated properties up to temperatures and pressures of 5000 K and 25 GPa. For typical calibration applications, the new reference equation is supplemented by a simple but also highly accurate formulation, valid only for supercritical nitrogen between 270 and 350 K at pressures up to 30 MPa.

KEY WORDS: caloric properties; density; equation of state; nitrogen; thermodynamic properties.

¹ Paper presented at the Thirteenth Symposium on Thermophysical Properties, June 22–27, 1997, Boulder, Colorado, U.S.A.

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1. INTRODUCTION

Nitrogen has been one of the most important reference fluids both for tests of physical models and for calibration purposes. The high demands on the availability and accuracy of thermophysical data resulting from this special role have led to the development of different reference equations of state which were based on available state-of-the-art data sets and correlation techniques. During the last 10 years, major improvements have taken place with respect to both of these aspects.

The equation of state presented by Jacobsen et al. [1] has been an accepted standard for the thermodynamic properties of nitrogen since its publication. The equation of state developed in that work was based on the Helmholtz energy of the fluid. A comprehensive evaluation of the experimental data available up to 1986 was reported by Jacobsen et al. [1]. Their work superseded the 32-term modified BWR equation of state published by Jacobsen and Stewart [2] in 1973 and the IUPAC international tables of properties for nitrogen published by Angus et al. [3] in 1979. Correlations for nitrogen prior to 1973 are reported by Jacobsen and Stewart [2]. The equations of Jacobsen et al. and of Stewart and Jacobsen were reported on the International Practical Temperature Scale of 1968 (IPTS-68). The new equation presented here is given on the International Temperature Scale of 1990 (ITS-90).

The new nitrogen equation of state presented here is based on many new data sets which have become available in recent years. These data include the p - ρ - T measurements of Nowak et al. [4, 5], Klimeck et al. [6], Fenghour et al. [7], Pieperbeck et al. [8], Jaeschke and Hinze [9], Duschek et al. [10], and Achtermann et al. [11], the isochoric heat capacity data of Magee [12], the speed of sound data of Costa Gomez and Trusler [13], Boyes [14], Ewing and Trusler [15], and Kortbeek et al. [16], and the enthalpy data of Grini and Owren [17]. New values of the critical parameters as well as the vapor pressures and coexisting densities were reported by Nowak et al. [5]. These new data along with several data sets used previously by Jacobsen et al. [1] form the basis of the new equation of state for nitrogen. In order to represent the available accurate data within their uncertainty, while maintaining the least number of coefficients, the new equation was developed using state-of-the-art optimization and multiproperty fitting algorithms.

The equation of state is valid for temperatures from the triple point to 1000 K at pressures up to 2200 MPa. Experimental shock tube measurements [18–20] verify that the equation extrapolates reasonably to the limits of chemical stability of nitrogen up to 5000 K and 25 GPa, at least with respect to basic properties like pressure, fugacity, and enthalpy. Further details are given in Ref. 21.

2. THE NEW EQUATION

The selected parameters for the critical point and triple point for nitrogen were reported by Nowak et al. [5]. These values are

$$\begin{aligned} T_c &= 126.192 \text{ K} \\ p_c &= 3.3958 \text{ MPa} \\ \rho_c &= 11.1839 \text{ mol} \cdot \text{dm}^{-3} \end{aligned} \quad (1)$$

The triple-point temperature is 63.151 K and the triple-point pressure is 12.523 kPa [5]. The molar mass is 28.01348 g · mol⁻¹ [22].

The equation for the ideal gas heat capacity used in this work was developed using the contributions from translation, rotation, and vibration of the nitrogen molecule. This equation is valid from 40 to 5000 K, at which point the effects of dissociation are no longer negligible. At temperatures below 40 K, the deviation between heat capacities calculated from the equation used here and those calculated from statistical thermodynamics increases reaching a maximum of 2.7% at 2.4 K. The ideal gas heat capacity passes through a maximum value of 3.597R at this temperature. The equation presented here does not account for this maximum, nor does it account for the decrease in heat capacity to 2.5R near 0 K, where the rotational modes are not excited. The equation is expressed as

$$\begin{aligned} \frac{c_p^0}{R} &= 3.5 + 3.066\,469 \times 10^{-6}T + 4.701\,240 \times 10^{-9}T^2 - 3.987\,984 \times 10^{-13}T^3 \\ &+ 1.012\,941 \frac{u^2 \exp(u)}{[\exp(u) - 1]^2} \end{aligned} \quad (2)$$

where u is 3364.011 K/T and the gas constant R is 8.31451 J · mol⁻¹ · K⁻¹ [23]. Since the vibrational relaxation time for nitrogen is very long compared to the period of sound waves, the vibrational modes of the molecule are not excited by acoustical measurements, and values of ideal gas heat capacity calculated from such measurements do not account for vibrational effects. These effects become noticeable in measurements at temperatures above 300 K. Thus, for calculations of the acoustical speed of sound w_{acs} in nitrogen gas, the thermodynamic speed of sound, $w_{\text{eos}} = \sqrt{(\partial p / \partial \rho)_s}$, calculated from the equation of state presented here can be adjusted according to

$$w_{\text{acs}} = w_{\text{eos}} \sqrt{\frac{c_v}{c_p} \left(\frac{c_p - c_{\text{vib}}}{c_v - c_{\text{vib}}} \right)} \quad (3)$$

where c_{vib} is the exponential term in Eq. (2). Further information is given by Costa Gomez and Trusler [13] and Span et al. [24].

The functional form of the new equation of state for nitrogen is explicit in the dimensionless Helmholtz energy α using independent variables of reduced density and temperature. The form of this equation is

$$\frac{a(\rho, T)}{RT} = \alpha(\delta, \tau) = \alpha^0(\delta, \tau) + \alpha^r(\delta, \tau) \quad (4)$$

where a is the Helmholtz energy, $\delta = \rho/\rho_c$, $\tau = T_c/T$, the superscript 0 refers to the ideal gas Helmholtz energy, and the superscript r refers to the residual Helmholtz energy. The ideal gas Helmholtz energy is given by

$$\alpha^0 = \frac{h_0^0 \tau}{RT_c} - \frac{s_0^0}{R} - 1 + \ln \frac{\delta \tau_0}{\delta_0 \tau} - \frac{\tau}{R} \int_{\tau_0}^{\tau} \frac{c_p^0}{\tau^2} d\tau + \frac{1}{R} \int_{\tau_0}^{\tau} \frac{c_p^0}{\tau} d\tau \quad (5)$$

where $\delta_0 = \rho_0/\rho_c$, $\tau_0 = T_c/T_0$, and ρ_0 is the ideal gas density at $T_0 = 298.15$ K and $p_0 = 0.101325$ MPa. The values of $h_0^0 = 8670$ J·mol⁻¹ and $s_0^0 = 191.5$ J·mol⁻¹·K⁻¹ were chosen to coincide with the values reported by Cox et al. [25]. Combining Eqs. (2) and (5), the resulting expression is

$$\begin{aligned} \alpha^0 = & \ln \delta + 2.5 \ln \tau - 12.769\,53 - 0.007\,841\,630\tau - 1.934\,819 \times 10^{-4}\tau^{-1} \\ & - 1.247\,742 \times 10^{-5}\tau^{-2} + 6.678\,326 \times 10^{-8}\tau^{-3} \\ & + 1.012\,941 \ln[1 - \exp(-26.657\,88\tau)] \end{aligned} \quad (6)$$

The functional forms of preliminary equations for the residual part of the Helmholtz energy were optimized with a modified version of the algorithm developed by Setzmann and Wagner [26]. In this algorithm, nonlinear data are linearized [27]. To improve the representation of the highly accurate speed of sound data and of the shock tube data available for nitrogen, the final functional form was developed with the nonlinear regression analysis developed by Tegeler et al. [28]. Using a combination of linear and nonlinear techniques, this algorithm determines the functional form which yields the best representation of the selected experimental data.

The bank of terms used as basis for both the linear and the nonlinear optimization procedure contained a total of 838 terms, including simple polynomial terms, combinations of polynomials with exponential expressions, and modified Gaussian terms which were introduced by Setzmann and Wagner [27] to improve the representation of data in the critical region. Neither the importance of the critical region of nitrogen nor the

data situation in this region made it necessary to use the complex non-analytical terms developed by Span and Wagner [29] for an improved description of caloric data in the immediate vicinity of the critical point.

The residual Helmholtz energy contribution is given by

$$\alpha^r(\delta, \tau) = \sum_{k=1}^6 N_k \delta^{i_k} \tau^{j_k} + \sum_{k=7}^{32} N_k \delta^{i_k} \tau^{j_k} \exp(-\delta^{l_k}) + \sum_{k=33}^{36} N_k \delta^{i_k} \tau^{j_k} \exp(-\varphi_k(\delta-1)^2 - \beta_k(\tau-\gamma_k)^2) \quad (7)$$

where the coefficients and parameters of this equation are given in Tables I and II. The coefficients N_k were determined using the selected data summarized in Table III. In total, 3393 of the 8244 data points available for p - ρ - T data were used and 1096 out of the 2393 data points available for speed of sound were used. In addition to those data shown in Table III, 44 isobaric heat capacity data, 14 heat of vaporization data, 203 Joule-Thomson data, and 5 shock tube data points were used in the fit. Calculated vapor pressure and saturated liquid and vapor density values were used in the linear optimization algorithm from ancillary equations

Table I. Parameters and Coefficients of the Equation of State for Nitrogen, Eq. (7)

k	N_k	i_k	j_k	l_k	k	N_k	i_k	j_k	l_k
1	0.924 803 575 275	1	0.25	0	19	$-0.435\ 762\ 336\ 045 \times 10^{-1}$	1	4.0	2
2	$-0.492\ 448\ 489\ 428$	1	0.875	0	20	$-0.723\ 174\ 889\ 316 \times 10^{-1}$	2	6.0	2
3	0.661 883 336 938	2	0.5	0	21	$0.389\ 644\ 315\ 272 \times 10^{-1}$	3	6.0	2
4	$-0.192\ 902\ 649\ 201 \times 10^1$	2	0.875	0	22	$-0.212\ 201\ 363\ 910 \times 10^{-1}$	4	3.0	2
5	$-0.622\ 469\ 309\ 629 \times 10^{-1}$	3	0.375	0	23	$0.408\ 822\ 981\ 509 \times 10^{-2}$	5	3.0	2
6	0.349 943 957 581	3	0.75	0	24	$-0.551\ 990\ 017\ 984 \times 10^{-4}$	8	6.0	2
7	0.564 857 472 498	1	0.5	1	25	$-0.462\ 016\ 716\ 479 \times 10^{-1}$	4	16.0	3
8	$-0.161\ 720\ 005\ 987 \times 10^1$	1	0.75	1	26	$-0.300\ 311\ 716\ 011 \times 10^{-2}$	5	11.0	3
9	$-0.481\ 395\ 031\ 883$	1	2.0	1	27	$0.368\ 825\ 891\ 208 \times 10^{-1}$	5	15.0	3
10	0.421 150 636 384	3	1.25	1	28	$-0.255\ 856\ 846\ 220 \times 10^{-2}$	8	12.0	3
11	$-0.161\ 962\ 230\ 825 \times 10^{-1}$	3	3.5	1	29	$0.896\ 915\ 264\ 558 \times 10^{-2}$	3	12.0	4
12	0.172 100 994 165	4	1.0	1	30	$-0.441\ 513\ 370\ 350 \times 10^{-2}$	5	7.0	4
13	$0.735\ 448\ 924\ 933 \times 10^{-2}$	6	0.5	1	31	$0.133\ 722\ 924\ 858 \times 10^{-2}$	6	4.0	4
14	$0.168\ 077\ 305\ 479 \times 10^{-1}$	6	3.0	1	32	$0.264\ 832\ 491\ 957 \times 10^{-3}$	9	16.0	4
15	$-0.107\ 626\ 664\ 179 \times 10^{-2}$	7	0.0	1	33	$0.196\ 688\ 194\ 015 \times 10^2$	1	0.0	—
16	$-0.137\ 318\ 088\ 513 \times 10^{-1}$	7	2.75	1	34	$-0.209\ 115\ 600\ 730 \times 10^2$	1	1.0	—
17	$0.635\ 466\ 899\ 859 \times 10^{-3}$	8	0.75	1	35	$0.167\ 788\ 306\ 989 \times 10^{-1}$	3	2.0	—
18	$0.304\ 432\ 279\ 419 \times 10^{-2}$	8	2.5	1	36	$0.262\ 767\ 566\ 274 \times 10^4$	2	3.0	—

Table II. Parameters of the Gaussian Terms in the Equation of State for Nitrogen, Eq. (7)

k	φ_k	β_k	γ_k
33	20	325	1.16
34	20	325	1.16
35	15	300	1.13
36	25	275	1.25

given by Nowak et al. [5]. In the final nonlinear optimization of the functional form [28], the 53 vapor pressures, 52 saturated liquid densities, and 50 saturated vapor densities published by Nowak et al. were used directly. The melting line equation of Jacobsen et al. [1] was adopted in this work. All thermodynamic properties such as pressure, heat capacities, and speed of sound can be calculated from Eq. (4) through differentiation of the Helmholtz energy with respect to temperature and/or density. The expressions for these properties can be found in Jacobsen et al. [1] or in Setzmann and Wagner [27]. The following properties calculated using Eqs. (6) and (7) for the specified temperature and density are given for validation of computer programs developed by others: (1) $T = 270$ K, $\rho = 11$ mol·dm⁻³, $p = 27.0621$ MPa, $h = 6517.95$ J·mol⁻¹, and $w = 459.222$ m·s⁻¹; (2) $T = 126.2$ K, $\rho = 11.2$ mol·dm⁻³, $p = 3.39712$ MPa, $h = 816.780$ J·mol⁻¹, and $w = 135.571$ m·s⁻¹. The second point is in the critical region and includes the effect of the Gaussian terms [terms 33–36 in Eq. (7)].

3. COMPARISONS TO EXPERIMENTAL DATA

The uncertainty of the equation of state was determined by comparisons of property values calculated with the equation of state to experimental data. These comparisons are based on the percent deviation in any property X defined as

$$\% \Delta X = 100 \left(\frac{X_{\text{data}} - X_{\text{calc}}}{X_{\text{data}}} \right) \quad (8)$$

Using this definition, the average absolute deviation is defined as

$$\text{AAD} = \frac{1}{n} \sum_{i=1}^n |\% \Delta X_i| \quad (9)$$

Table III. Comparisons of the Equation of State for Nitrogen with Selected Experimental Data

Author	No. of selected points	Pressure range (MPa)	Temp. range (K)	AAD
<i>p</i> - ρ - <i>T</i> data				
Achtermann et al. [11]	35	1.07–28.7	323	0.019
Duschek et al. [10]	127	0.50–8.01	273–323	0.004
Fenghour et al. [7]	50	3.54–37.0	290–680	0.077
Jaeschke and Hinze (Burnett method) [9]	129	0.22–30.2	273–353	0.010
Jaeschke and Hinze (Interferometer) [9]	499	0.24–28.7	269–353	0.011
Klimeck et al. [6]	264	1.11–30.1	240–520	0.002
Michels et al. [30]	56	1.93–8.58	273–423	0.016
Michels et al. [31]	147	19.5–300.0	273–423	0.017
Nowak et al. [4] ^a	813	0.10–12.0	66–340	0.002
Nowak et al. [4] ^b	107	2.30–5.60	120–140	0.003
Nowak et al. [5] ^b	172	3.20–3.39	125–126	0.002
Pieperbeck et al. [8]	124	0.10–12.1	273–323	0.003
Saurel [32]	85	1.01–91.2	423–1074	0.091
Straty and Diller [33]	174	0.83–34.8	80–300	0.016
Other data ^c	611	0.40–2200.0	248–673	0.242
Speed of sound data				
Boyes [14] ^d	112	0.05–6.64	250–325	0.001
Ewing and Trusler [15] ^d	98	0.00–0.58	80–373	0.001
Kortbeek et al. [16]	134	85.0–1000.0	123–298	0.402
Costa Gomez and Trusler [13] ^d	72	0.00–30.1	250–350	0.003
Younglove and McCarty [34]	237	0.03–1.51	80–350	0.065
Other data ^c	443	0.10–2200.0	64–1001	0.311
Calorimetric data				
Magee [12] (c_v)	173	6.1–31.0 ^e	66–307	0.625
Weber [35] (c_v)	61	10.7–27.5 ^e	91–242	0.404
Magee [12] (c_{sat})	102		56–101	0.318
Grini and Owren [17] (Δh)	19	0.31–15.0	160–240	0.128

^a Data in the critical region are reported separately.

^b Deviations in pressure for data in the critical region.

^c Due to space limitations, 8 *p*- ρ -*T* and 12 speed of sound data sets have been grouped together as “other data” References for these data sets are given by Span et al. [24].

^d Data corrected according to Eq. (3).

^e Density range ($\text{mol} \cdot \text{dm}^{-3}$).

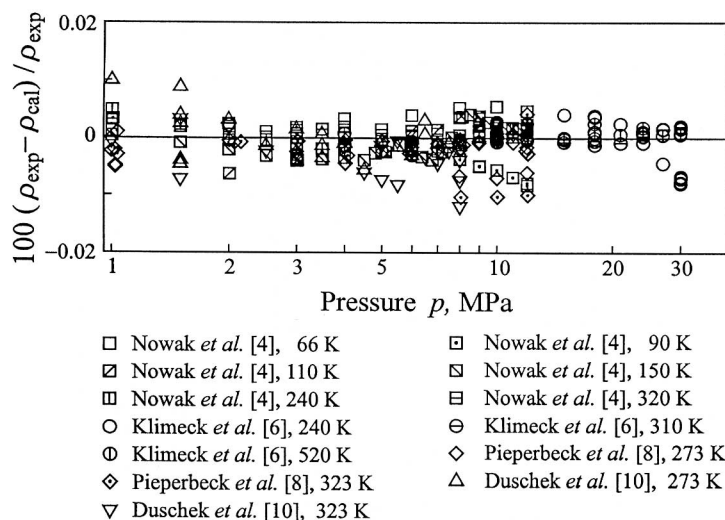


Fig. 1. Comparisons of experimental p - ρ - T data measured with single- and two-sinker densimeters to calculations from the new equation of state for nitrogen.

where n is the number of data points. Table III shows comparisons of selected experimental data sets that were used in fitting the equation of state for p - ρ - T , isochoric and saturation heat capacities, speed of sound, and enthalpy data. Figure 1 shows density deviations calculated using the equation of state from p - ρ - T data measured with single- and two-sinker densimeters.

4. UNCERTAINTIES

The new reference equation of state describes the p - ρ - T surface with an uncertainty of less than $\pm 0.02\%$ in density from the triple point up to temperatures of 523 K and pressures up to 12 MPa and from temperatures of 240 to 523 K at pressures less than 30 MPa. The uncertainty is less than $\pm 0.02\%$ in pressure in the critical region. Further information is given in Fig. 2. In the gaseous and supercritical region, highly accurate speed-of-sound data are represented within ± 0.005 to $\pm 0.02\%$ as shown in Fig. 3. The estimated uncertainty for the speed of sound in the liquid phase and at pressures above 30 MPa is $\pm 0.6\%$ and the estimated uncertainty for heat capacities is ± 0.5 to $\pm 1\%$ over the range of validity of the equation, except in the critical region.

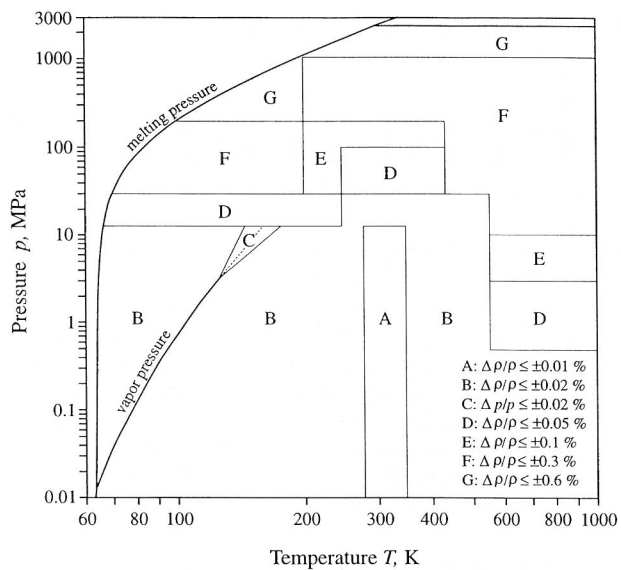


Fig. 2. Uncertainties in density for the new equation of state for nitrogen.

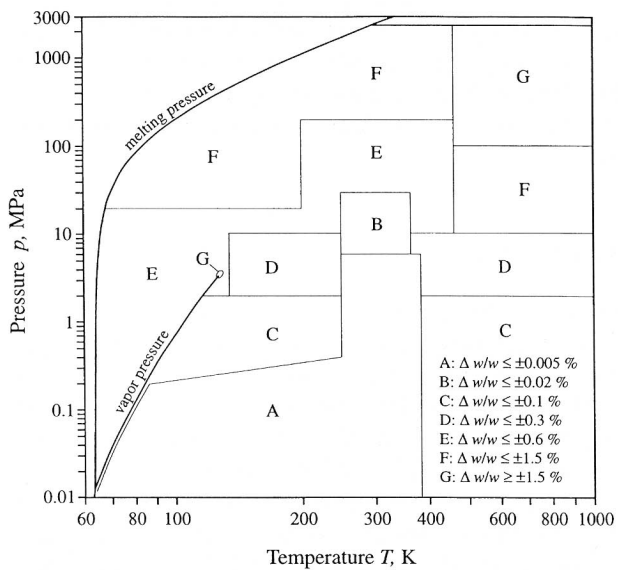


Fig. 3. Uncertainties in speed of sound for the new equation of state for nitrogen.

The saturation values can be calculated from the equation of state through the use of the Maxwell criterion. The estimated uncertainties of vapor pressure, saturated liquid density, and saturated vapor density are $\pm 0.02\%$ for each property. The new formulation yields a reasonable extrapolation up to the limits of chemical stability of nitrogen as indicated by comparison to experimental shock tube data.

5. THE CALIBRATION EQUATION

In order to describe the thermodynamic behavior of nitrogen in the so-called “natural-gas region” for instrument calibration purposes, a limited equation of state has been developed. This equation is valid from 270 to 350 K at pressures up to 30 MPa and its general functional form is identical to Eq. (4). For the ideal gas heat capacity used in Eq. (5), the simplified equation

$$\frac{c_p^0}{R} = 3.500\,571 + 1.115\,488 \times 10^{-20} T^7 \quad (10)$$

was used, which is valid from 270 to 350 K. The ideal gas Helmholtz energy equation can then be expressed as

$$\begin{aligned} \alpha^0 = & \ln(\delta) + 2.500\,571 \ln(\tau) - 12.769\,41 - 0.008\,137\,875\tau \\ & - 1.015\,078\,5 \times 10^{-7} \tau^{-7} \end{aligned} \quad (11)$$

The residual Helmholtz energy contribution to the equation of state is given by

$$\alpha^r(\delta, \tau) = \sum_{k=1}^{10} M_k \delta^{i_k} \tau^{j_k} \quad (12)$$

The coefficients and parameters of this equation are given in Table IV. The associated equation for pressure is given by

$$p = \rho RT \left[1 + \sum_{k=1}^{10} i_k M_k \delta^{i_k} \tau^{j_k} \right] \quad (13)$$

The maximum deviation between these equations and Eqs. (6) and (7) is 0.001% in density in the specified range, and for the speed of sound, the maximum deviation is 0.002% at pressures less than 7 MPa and 0.006% for higher pressures. Thus, the wide-range equation and the simplified

Table IV. Parameters and Coefficients of the Short Equation of State for Nitrogen, Eq. (12)

k	M_k	i_k	j_k	k	M_k	i_k	j_k
1	-0.409 226 050 427	1	-1.0	6	$0.112\ 593\ 677\ 045 \times 10^{-1}$	3	0.0
2	0.583 733 818 214	1	-0.875	7	$-0.604\ 379\ 290\ 033 \times 10^{-1}$	3	2.875
3	$-0.132\ 040\ 812\ 535 \times 10^1$	1	1.625	8	$0.567\ 224\ 683\ 248 \times 10^{-2}$	4	-0.125
4	$0.854\ 602\ 646\ 673 \times 10^{-1}$	2	0.125	9	$-0.496\ 167\ 879\ 044 \times 10^{-2}$	6	-1.0
5	0.207 794 266 769	2	3.5	10	$0.572\ 786\ 635\ 566 \times 10^{-2}$	6	-0.875

equation can be used alternatively without causing significant inconsistencies. The following properties calculated using Eqs. (11) and (12) for the specified temperature and density are given for validation of computer programs developed by others: $T = 270$ K, $\rho = 11$ mol · dm⁻³, $p = 27.0621$ MPa, $h = 6518.00$ J · mol⁻¹, and $w = 459.220$ m · s⁻¹.

ACKNOWLEDGMENTS

R. Span and W. Wagner thank the Deutsche Forschungsgemeinschaft for their financial support of this project.

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