

# Thermophysical Properties of Fluid D<sub>2</sub>O

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The present publication contains data on the thermophysical properties of deuterium oxide (heavy water). It is a companion to the paper on the thermophysical properties of fluid H<sub>2</sub>O published earlier in this journal by the same authors. The properties are represented by equations which can be readily programmed on a computer and incorporated in data banks. All data have been carefully and critically analyzed. The compendium represents the best available data for fluid D<sub>2</sub>O.

Key words: critical parameters; equation of state; fundamental equation; heavy steam; heavy water; thermal conductivity; thermodynamic properties; viscosity.

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## 1. Introduction

The present publication contains data on the thermophysical properties of deuterium oxide alias heavy water

(D<sub>2</sub>O). It is a companion to the paper on the thermophysical properties of fluid H<sub>2</sub>O published earlier in this journal by the same authors.<sup>1</sup> The properties are represented by equations which can be readily programmed on a computer and incorporated in data banks. Most of the numerical values and formulations have been studied quite extensively by the International Association for the Properties of Steam (IAPS) and several of the formulations have been endorsed as inter-

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national standards.<sup>2</sup> It must, however, be emphasized that the present publication, like the preceding publication on the thermophysical properties of light water,<sup>1</sup> is not issued under the authority of IAPS. The aim is to supply the best contemporary values for the thermophysical properties of fluid D<sub>2</sub>O as judged by the present authors regardless of international agreements.

Heavy water is of interest in a number of applications in engineering and physics. In engineering, heavy water is used for its excellent nuclear and thermal properties. These make it suitable as a moderator in nuclear reactors because D<sub>2</sub>O possesses a small cross section for the capture of neutrons. Since the thermal properties of heavy water are very much like those of light water, it is very suitable for use both as a moderator in homogeneous reactors using natural uranium and as a working fluid in nuclear reactors for power applications (CANDU—Canadian deuterium reactor for the production of electricity). For example, heavy water freezes at 3.8 °C under a pressure of 1 atm and its normal boiling point is 101.42 °C; the maximum density of the saturated liquid occurs at 11.2 °C. At 20 °C, its density is 1.1050 kg/dm<sup>3</sup> compared to 0.9982 kg/dm<sup>3</sup> for H<sub>2</sub>O.

In view of the applications of heavy water, many research workers have measured the thermodynamic and transport properties of heavy water and prepared computer codes for their representation.<sup>3</sup> The material thus generated is of sufficient reliability to justify the present publication.

From the point of view of a physicist, the two versions of water, H<sub>2</sub>O and D<sub>2</sub>O, whose properties are probably better known than for any other isotopic pair of fluids, constitute a valuable system for the study of the relationships between isotopes.

The discovery of the heavy isotope D of hydrogen, whose atomic mass is double that of the latter, was made in 1931 by H. C. Urey, G. M. Murphy, and F. G. Brickwedde.<sup>4,5</sup> The leader of the team, Harold Urey, was awarded the Nobel Prize for this discovery in 1934. It is interesting to note that, not unlike the case of the discovery of argon by Lord Rayleigh<sup>6</sup> (for which he was also honored with the Nobel Prize), the possibility of the existence of an isotopic version of water was foreshadowed by the very accurate measurements of the density of pure water performed by A. B. Lamb and R. E. Lee in 1913.<sup>7</sup> Their measurements of various samples of water,<sup>5</sup> carefully prepared and purified, varied in density by as much as (or as little as!)  $8 \times 10^{-7}$  g/cm<sup>3</sup>, in the face of a precision of measurement of  $2 \times 10^{-7}$  g/cm<sup>3</sup>. This difference was later quoted as evidence of the existence of an isotope.

Heavy hydrogen gives rise to two isotopes of water, namely HDO with a molar mass of 19 g/mol and D<sub>2</sub>O with a molar mass of 20 g/mol, against 18 g/mol for H<sub>2</sub>O. The first sample of practically pure D<sub>2</sub>O was produced in 1933 by G. N. Lewis and R. T. MacDonald.<sup>8</sup> This marked the beginning of the research into its thermophysical properties.

It is remarkable that the isotopic composition of naturally occurring water on earth varies very little from place to place. In order to secure very accurate reproducibility, H. Craig created a standard known as Standard Mean Ocean Water (SMOW).<sup>9-11</sup> Since oxygen possesses three stable iso-

topes, <sup>16</sup>O, <sup>17</sup>O, <sup>18</sup>O, the composition specification for SMOW is quite complex, as shown hereunder:

Component	Mole fraction
<sup>1</sup> H	0.999 842
<sup>2</sup> H(D)	0.000 158
<sup>3</sup> H(T)	0
<sup>16</sup> O	0.997 640
<sup>17</sup> O	0.000 371
<sup>18</sup> O	0.001 989

In the present paper, it is assumed that heavy water consists of 100% D<sub>2</sub>O with the same relative abundance of oxygen isotopes as in SMOW.

A survey of various thermophysical and chemical properties of heavy water was published in 1951 in the U.S. in a book by Kirshenbaum.<sup>12</sup> Subsequently, a comprehensive collection of thermophysical property information, including tables of thermodynamic properties of heavy water, was published by V. A. Kirillin and co-workers in the Soviet Union.<sup>13</sup> At the time, the authors could claim that they had taken into account "all experimental and theoretical data published in their country and abroad," even though the work was done without the availability of computers.

A global fundamental equation, and new tables of thermodynamic properties of heavy water derived from it, were recently published by P. G. Hill, R. D. C. MacMillan, and V. Lee of the University of British Columbia in Canada.<sup>14,15</sup> This fundamental equation has been adopted by IAPS as the *Provisional IAPS Formulation 1983 for the Thermodynamic Properties of Heavy Water Substance*.<sup>2</sup> The global fundamental equation quoted in this paper is a dimensionless version of the fundamental equation of Hill *et al.*

Present-day understanding of the thermodynamic properties of any substance, including D<sub>2</sub>O, indicates that the thermodynamic surface becomes nonanalytic at the critical point. Thus it is convenient to exclude a small near-critical region from the thermodynamic surface specified by the analytic global fundamental equation mentioned above. Inside this critical region, recourse is made to the scaling laws predicted by theory for the critical behavior of substances.<sup>16</sup>

The question may be raised whether the properties of heavy water could not have been simply predicted from those of light water with the aid of the law of corresponding states. The answer is that, although this law seems to hold well for several of the properties of these two substances, there are notable exceptions. Properties that do show correspondence are the critical compressibility factor  $Z_c = P_c V_c / RT_c$  and the compressibility factors in the vapor and liquid phases from 20 to 300 °C. The differences of a few tenths of a percent noted by Hill and Lee<sup>17</sup> are within the uncertainty of the critical parameters used as reduction factors. The anomalous (scaled) parts of the critical properties of the two substances are identical within the accuracy of the data when expressed in reduced units.<sup>16</sup> On the other hand, significant departures from corresponding states have been found for the vapor-pressure curves. Although the critical temperature of heavy water is 3.2 K lower than that of light water, its triple point temperature is 3.8 K higher. The reduced slopes of the vapor pressure curves differ by 0.5% at

the critical point. At reduced temperatures from 0.85 down to 0.6, the reduced vapor pressure of D<sub>2</sub>O is a few percents lower than that of H<sub>2</sub>O. At lower temperatures, the differences become larger; near the triple point the reduced vapor pressure of D<sub>2</sub>O is approximately 30% below that of H<sub>2</sub>O.

Thus, although the assumption of corresponding states for D<sub>2</sub>O and H<sub>2</sub>O is quite sensible, the departures definitely exceed the accuracy of the data for some of the properties. Hence, an accurate thermodynamic surface cannot be obtained on the basis of this assumption.

Equations for the transport properties of fluid D<sub>2</sub>O have been developed by N. Matsunaga and A. Nagashima of Keio University in Yokohama.<sup>18</sup> These equations have been adopted by IAPS as the international representation of the transport properties of heavy water substance<sup>2</sup> and we have incorporated these equations in this paper.

All equations have been programed on computers and

calculated results were checked against critically evaluated experimental data. A short numerical table is included in this paper solely for the purpose of verifying computer codes.

The authors hope that this selection of formulas will be of use to all those who need computer codes for the thermophysical properties of fluid D<sub>2</sub>O.

## 2. Molecular Data

$$\text{Molar mass}^{11}: \quad M = 0.020\,027\,478 \text{ kg/mol}, \quad (2.1)$$

$$\text{universal gas constant}^{19}: \quad R = 8.314\,41 \text{ J/mol K}, \quad (2.2)$$

$$\text{specific gas constant}: \quad r = 415.150 \text{ J/kg K}. \quad (2.3)$$

The molar mass refers to 100% D<sub>2</sub>O with the same relative abundance of oxygen isotopes as in standard mean ocean water.<sup>11</sup>

## 3. Critical Constants

$$\text{Critical temperature:} \quad T_{cr} = (643.89 + \delta)K = (370.74 + \delta)^\circ\text{C} \text{ with } -0.2 \leq \delta \leq +0.2, \quad (3.1)$$

$$\text{critical pressure:} \quad P_{cr} = (21.671 + 0.27\delta \pm 0.010)\text{MPa}, \quad (3.2)$$

$$\text{critical density:} \quad \rho_{cr} = (356 \pm 5)\text{kg/m}^3, \quad (3.3)$$

$$\text{critical specific volume:} \quad V_{cr} = (0.002\,81 \pm 0.000\,04)\text{m}^3/\text{kg}, \quad (3.4)$$

as adopted by the International Association for the Properties of Steam.<sup>2</sup>

The above values for the critical constants are the results of measurements which may be repeated and improved. On the other hand, critical parameters are often used as reference quantities in correlations. Once adopted, such refer-

ence constants need not be revised when our knowledge of the critical constants improves. In order to distinguish the reference constants from the critical constants (to which they are close), we denote the reference constants by asterisks:  $P^*$ ,  $T^*$ ,  $\rho^*$  and treat them as empirical correlation constants throughout the remainder of the publication.

## 4. Triple Point<sup>14,20-21</sup>

$$\text{Triple-point temperature:} \quad T_{tr} = (276.97 \pm 0.02)K = (3.82 \pm 0.02)^\circ\text{C}, \quad (4.1)$$

$$\text{triple-point pressure:} \quad P_{tr} = (661 \pm 3) \text{ Pa}, \quad (4.2)$$

$$\text{density of liquid at triple point:} \quad \rho_{tr,L} = (1105.5 \pm 0.2)\text{kg/m}^3, \quad (4.3)$$

$$\text{density of vapor at triple point:} \quad \rho_{tr,G} = (0.005\,75 \pm 0.000\,03)\text{kg/m}^3. \quad (4.4)$$

## 5. Dimensionless Variables and Reference Constants

All equations in this publication are presented in nondimensional form. The dimensionless version of each quantity

is denoted by a symbol with a bar. The reference quantities consist of two classes: five primary reference quantities (denoted by an asterisk) and three secondary reference quantities (denoted by a double asterisk) which are simple combinations of the primary reference quantities.

### 5.1. Primary Reference Constants

$$\text{Reference temperature:} \quad T^* = 643.89 \text{ K}, \quad (5.1)$$

$$\text{reference density:} \quad \rho^* = 358 \text{ kg/m}^3, \quad (5.2)$$

$$\text{reference pressure:} \quad P^* = 21.671 \times 10^6 \text{ Pa}, \quad (5.3)$$

$$\text{reference viscosity:} \quad \eta^* = 55.2651 \times 10^{-6} \text{ Pa s}, \quad (5.4)$$

reference thermal conductivity:  $\lambda^* = 0.742\,128 \times 10^{-3} \text{ W/K m}$ . (5.5)

It is emphasized that the three reference constants  $T^*, \rho^*, P^*$  are close to but not identical with the critical parameters  $T_{cr}, \rho_{cr}, P_{cr}$ .

## 5.2 Secondary Reference Constants

Reference constant for Helmholtz function, energy, enthalpy, Gibbs function:

$$A^{**} \equiv \frac{P^*}{\rho^*} = 60\,533.52 \text{ J/kg}, \quad (5.6)$$

reference constant for entropy, specific heats:

$$S^{**} \equiv \frac{P^*}{\rho^* T^*} = 94.012\,21 \text{ J/kg K}, \quad (5.7)$$

reference constant for sound velocity:

$$w^{**} \equiv \left( \frac{P^*}{\rho^*} \right)^{1/2} = 246.036 \text{ m/s}. \quad (5.8)$$

## 5.3. Thermophysical Properties in Dimensionless Form

Temperature:  $\bar{T} = T/T^*$ , (5.9)

pressure:  $\bar{P} = P/P^*$ , (5.10)

density:  $\bar{\rho} = \rho/\rho^*$ , (5.11)

specific volume:  $\bar{V} = V/\rho^*$ , (5.12)

specific Helmholtz function:  $\bar{A} = A/A^{**}$ , (5.13)

specific energy:  $\bar{U} = U/A^{**}$ , (5.14)

specific enthalpy:  $\bar{H} = H/A^{**}$ , (5.15)

specific Gibbs function:  $\bar{G} = G/A^{**}$ , (5.16)

chemical potential:  $\bar{\mu} = \mu/A^{**}$ , (5.17)

specific entropy:  $\bar{S} = S/S^{**}$ , (5.18)

specific heat at constant volume:  $\bar{C}_v = C_v/S^{**}$ , (5.19)

specific heat at constant pressure:  $\bar{C}_p = C_p/S^{**}$ , (5.20)

isothermal compressibility:  $\bar{K}_T = K_T P^*$ , (5.21)

symmetrized compressibility:  $\bar{\chi}_T = \bar{\chi}_T P^*/\rho^{*2}$ , (5.22)

speed of sound:  $\bar{w} = w/w^{**}$ , (5.23)

viscosity:  $\bar{\eta} = \eta/\eta^*$ , (5.24)

thermal conductivity:  $\bar{\lambda} = \lambda/\lambda^*$ . (5.25)

## 6. Equilibrium Thermodynamic Properties in the Analytic Range

All equilibrium thermodynamic properties in the analytic region, that is, to the exclusion of a small area near the critical point, are implied by the canonical equation

$$A = A(T, \rho), \quad (6.1)$$

in which the Helmholtz free energy  $A$  is represented as a function of temperature and density.

The equation is based on the formulation constructed by Hill, MacMillan, and Lee.<sup>14,15</sup> It has been provisionally adopted by the International Association for the Properties of Steam as the *Provisional IAPS Formulation 1983 for the Thermodynamic Properties of Heavy Water Substance*.<sup>2</sup> For the present publication the original equation<sup>15</sup> has been converted to a nondimensional form for convenience of programming. In order to change the system of units in the output, it is sufficient to perform the conversion in the primary

and secondary reference constants given earlier in Secs. 5.1 and 5.2.

## 6.1. Fundamental Equation in Canonical Form

$$\bar{A}(\bar{T}, \bar{\rho}) = \bar{A}_0(\bar{T}, \bar{\rho}) + \bar{A}_1(\bar{T}, \bar{\rho}) \quad (6.2)$$

with

$$\bar{A}_0(\bar{T}, \bar{\rho}) = (A_{00} + A_{01}\bar{T})\ln \bar{T} + \sum_{j=2}^7 A_{0j}\bar{T}^{j-2} + A_{08}\bar{T}\ln \bar{\rho}, \quad (6.3)$$

$$\bar{A}_1(\bar{T}, \bar{\rho}) = \bar{T}\bar{\rho} \left( \frac{1}{\bar{T}} - \frac{1}{\bar{T}_1} \right) \sum_{i=1}^7 \left( \frac{1}{\bar{T}} - \frac{1}{\bar{T}_i} \right)^{i-2} \times \left[ \sum_{j=1}^8 A_{ij}(\bar{\rho} - \bar{\rho}_i)^{j-1} + e^{-1.5394\bar{\rho}} \sum_{j=9}^{10} A_{ij}\bar{\rho}^{j-9} \right]. \quad (6.4)$$

The fundamental Eq. (6.2) covers the range

$$P < 100 \text{ MPa}, T_{cr} < T < 800 \text{ K}. \quad (6.5)$$

We recommend the analytic fundamental equation everywhere in this range except for a region around the critical point bounded by

$$0.991 < \bar{T} < 1.06, \quad 0.7 < \bar{\rho} < 1.3. \quad (6.6)$$

Inside the range bounded by Eq. (6.6), a more accurate representation of the derivatives of the thermodynamic surface is obtained from the nonanalytic equation presented in Sec. 8.

## 6.2. Parameter Values for Fundamental Equation

The values of the parameters  $A_{ij}$ ,  $\bar{T}_i$ , and  $\bar{\rho}_i$  in the equations for  $\bar{A}_0$  and  $\bar{A}_1$  are given in Tables 1–3. In all tables presented in this paper, we have adopted the “E-notation.” That is, the integer which follows E indicates the power of ten by which the listed coefficient value should be multiplied. Thus E + 2 represents the factor  $10^2 = 100$ , etc.

The choice of the coefficients  $A_{02}$  and  $A_{03}$  in Eq. (6.3) is related to the convention adopted for the zero points of energy and entropy. Unlike the case of  $\text{H}_2\text{O}$ , the choice of the reference point for internal energy and entropy presents us with a problem. In the case of  $\text{H}_2\text{O}$ , the values of specific

TABLE 1. Coefficients  $A_{0j}$

$j$	$A_{0j}$
0	0.5399322597E - 2
1	- 0.1288399716E + 2
2	0.3087284587E + 2
3	- 0.3827563059E + 2
4	0.4424799189E + 0
5	- 0.1256336874E + 1
6	0.2843343470E + 0
7	- 0.2401555088E - 1
8	0.4415884023E + 1

TABLE 2. Coefficients  $A_{ij}$

$i$	$j$	$A_{ij}$
1	1	0.115623643567E + 3
1	2	- 0.161413392951E + 3
1	3	0.108543003981E + 3
1	4	- 0.471342021238E + 2
1	5	0.149218685173E + 2
1	6	- 0.360628259650E + 1
1	7	0.686743026455E + 0
1	8	- 0.951913721401E - 1
1	9	- 0.157513472656E + 4
1	10	- 0.433677787466E + 3
2	1	0.607446060304E + 2
2	2	- 0.927952190464E + 2
2	3	0.632086750422E + 2
2	4	- 0.264943219184E + 2
2	5	0.905675051855E + 1
2	6	- 0.578949005123E + 0
2	7	0.665590447621E + 0
2	8	- 0.525687146109E - 1
2	9	- 0.341048601697E + 4
2	10	- 0.146971631028E + 4
3	1	0.444139703648E + 2
3	2	- 0.580410482641E + 2
3	3	0.354090438940E + 2
3	4	- 0.144432210128E + 2
3	9	- 0.102135518748E + 4
3	10	- 0.136324396122E + 4
4	1	0.157859762687E + 2
4	2	- 0.194973173813E + 2
4	3	0.114841391216E + 2
4	4	- 0.196956103010E + 1
4	9	- 0.277379051954E + 3
4	10	- 0.481991835255E + 3
5	1	- 0.619344658242E + 2
5	2	0.791406411518E + 2
5	3	- 0.484238027539E + 2
5	4	0.191546335463E + 2
5	9	0.128039793871E + 4
5	10	0.186367898973E + 4
6	1	- 0.749615505949E + 2
6	2	0.947388734799E + 2
6	3	- 0.575266970986E + 2
6	4	0.173229892427E + 2
6	9	0.137572687525E + 4
6	10	0.231749018693E + 4
7	1	- 0.260841561347E + 2
7	2	0.328640711440E + 2
7	3	- 0.186464444026E + 2
7	4	0.484262639275E + 1
7	9	0.430179479063E + 3
7	10	0.822507844138E + 3

Note: Coefficients  $A_{ij}$  omitted from table are zero identically.

TABLE 3. Parameters  $\bar{T}_i$  and  $\bar{\rho}_i$

$i$	$\bar{T}_i$	$\bar{\rho}_i$
1	0.1000038832E + 1	0.1955307263E + 1
2	0.6138578282E + 0	0.3072625698E + 1
3	0.6138578282E + 0	0.3072625698E + 1
4	0.6138578282E + 0	0.3072625698E + 1
5	0.6138578282E + 0	0.3072625698E + 1
6	0.6138578282E + 0	0.3072625698E + 1
7	0.6138578282E + 0	0.3072625698E + 1

energy and entropy of the liquid at the triple point are set equal to zero.<sup>1</sup> The temperature of the triple point of H<sub>2</sub>O need not be measured, because it constitutes the (single) fixed point on the internationally accepted Thermodynamic Scale and is, naturally, incorporated in the International Practical Temperature Scale. In the case of D<sub>2</sub>O, the triple-point temperature must be obtained from accurate measurements which are subject to improvement. Following Hill *et al.*,<sup>15</sup> we have adopted as the reference point the liquid state at  $T_t = 3.80$  °C and at  $P_t = 660.066$  Pa which is the vapor-liquid saturation pressure at 3.80 °C implied by the surface. Hence, at this pseudo-triple point we have

$$T_t = 276.95 \text{ K}, \quad P_t = 660.066 \text{ Pa}, \quad (6.7)$$

$$U_{t,L} = 0, \quad S_{t,L} = 0, \quad (6.8)$$

$$H_{t,L} = 0.597 \text{ J/kg}. \quad (6.9)$$

Comparing Eq. (6.7) with Eqs. (4.1) and (4.2), we note that this pseudo-triple point may differ slightly from the "true" triple point and, in fact, it may correspond to a metastable liquid state. We do not think that this specific choice of the absolute values of  $U$  and  $S$  is important for most users for whom this paper is intended.

In the liquid region, small changes in density along an isotherm cause large changes in pressure. For this reason, due to an accumulation of small errors, a particular computer code may fail to return the zeros [Eq. (6.8)] at the liquid density which corresponds to the values of  $P_t$  and  $T_t$  imposed by Eq. (6.7). This problem can be solved by adjusting the constants  $A_{02}$  and  $A_{03}$  in Eq. (6.3) so that condition (6.8) is satisfied with the desired accuracy.

### 6.3. Thermodynamic Relations

All thermodynamic properties of interest can be derived from the fundamental Eq. (6.2) by the use of the following thermodynamic relations

$$\bar{V} = \bar{\rho}^{-1}, \quad (6.10)$$

$$\bar{P} = \bar{\rho}^2 \left( \frac{\partial \bar{A}}{\partial \bar{\rho}} \right)_{\bar{T}}, \quad (6.11)$$

$$\left( \frac{\partial \bar{P}}{\partial \bar{\rho}} \right)_{\bar{T}} = \frac{2\bar{P}}{\bar{\rho}} + \bar{\rho}^2 \left( \frac{\partial^2 \bar{A}}{\partial \bar{\rho}^2} \right)_{\bar{T}}, \quad (6.12)$$

$$\bar{K}_T^{-1} = \bar{\rho} \left( \frac{\partial \bar{P}}{\partial \bar{\rho}} \right)_{\bar{T}}, \quad (6.13)$$

$$\bar{\chi}_T = \bar{\rho}^2 \bar{K}_T, \quad (6.14)$$

$$\left( \frac{\partial \bar{P}}{\partial \bar{T}} \right)_{\bar{\rho}} = \bar{\rho}^2 \frac{\partial^2 \bar{A}}{\partial \bar{\rho} \partial \bar{T}} \quad (6.15)$$

$$\bar{S} = - \left( \frac{\partial \bar{A}}{\partial \bar{T}} \right)_{\bar{p}}, \quad (6.16)$$

$$\bar{U} = \bar{A} + \bar{T}\bar{S}, \quad (6.17)$$

$$\bar{H} = \bar{U} + \frac{\bar{P}}{\bar{\rho}}, \quad (6.18)$$

$$\bar{G} = \bar{\mu} = \bar{A} + \frac{\bar{P}}{\bar{\rho}}, \quad (6.19)$$

$$\bar{C}_v = -\bar{T} \left( \frac{\partial^2 \bar{A}}{\partial \bar{T}^2} \right)_{\bar{p}}, \quad (6.20)$$

$$\bar{C}_p = \bar{C}_v + \frac{\bar{T}}{\bar{\rho}^2} \left( \frac{\partial \bar{P}}{\partial \bar{T}} \right)_{\bar{p}}^2, \quad (6.21)$$

$$\bar{w} = \left[ \frac{\bar{C}_p}{\bar{C}_v} \left( \frac{\partial \bar{P}}{\partial \bar{p}} \right)_{\bar{T}} \right]^{1/2}. \quad (6.22)$$

## 7. Ideal-Gas Properties

The ideal-gas properties of D<sub>2</sub>O are obtained from

$$\begin{aligned} \bar{A}_{id} = \bar{A}_0(\bar{T}, \bar{\rho}) = & (A_{00} + A_{01}\bar{T}) \ln \bar{T} \\ & + \sum_{j=2}^7 A_{0j} \bar{T}^{j-2} + A_{08} \bar{T} \ln \bar{\rho} \end{aligned} \quad (7.1)$$

as a function of temperature and density, or from

$$\begin{aligned} \bar{A}_{id} = & (A_{00} + A_{01}\bar{T} - A_{08}\bar{T}) \ln \bar{T} + \sum_{j=2}^7 A_{0j} \bar{T}^{j-2} \\ & - A_{08} \bar{T} \ln A_{08} + A_{08} \bar{T} \ln \bar{P} \end{aligned} \quad (7.2)$$

as a function of temperature and pressure.

We remark that Hill and co-workers adopted the value  $r = 415.147$  J/kg K for the specific gas constant,<sup>22</sup> rather than the recommended value  $r = 415.150$  J/kg K given in Eq. (2.2). The dimensionless analytic equation given here is identical with the dimensional equation of Hill *et al.*<sup>14</sup> provided that one uses in the latter equation  $r = 415.147$  J/kg K. The differences, obtained when the more accurate value  $r = 415.150$  J/kg K is used, are unimportant.

## 8. Equilibrium Thermodynamic Properties in the Critical Region

In the critical region, Eq. (6.6), it is more convenient to represent the thermodynamic properties by means of a canonical equation among intensive variables. The form chosen here is<sup>16</sup>

$$\frac{P}{T} = \frac{P}{T} \left( \frac{1}{T}, \frac{\mu}{T} \right). \quad (8.1)$$

### 8.1. Revised Dimensionless Variables for Critical Region

For a representation of the thermodynamic properties in the critical region, it is convenient to transform the reference temperature, density, and pressure to the value of these parameters at the critical point of the thermodynamic surface. Its coordinates now assume the role of revised reference constants and are defined by

$$T_c = 643.89 \text{ K} \quad \text{or} \quad \bar{T}_c = 1.000 \ 000, \quad (8.2)$$

$$\rho_c = 356.238 \text{ kg/m}^3 \quad \text{or} \quad \bar{\rho}_c = 0.995 \ 078, \quad (8.3)$$

$$P_c = 21.6713 \times 10^6 \text{ Pa} \quad \text{or} \quad \bar{P}_c = 1.000 \ 014. \quad (8.4)$$

The canonical equation is so structured that its critical point is located at  $T_c, \rho_c, P_c$ .

The reduced variables employed in the canonical equation for the critical region are defined by

$$\tilde{T} = - \frac{\bar{T}_c}{\bar{T}}, \quad (8.5)$$

$$\tilde{\mu} = \frac{\bar{\mu}}{\bar{T}} \frac{\bar{\rho}_c \bar{T}_c}{\bar{P}_c}, \quad (8.6)$$

$$\tilde{P} = \frac{\bar{P}}{\bar{T}} \frac{\bar{T}_c}{\bar{P}_c}, \quad (8.7)$$

$$\tilde{\rho} = \frac{\bar{\rho}}{\bar{\rho}_c}, \quad (8.8)$$

$$\tilde{U} = \bar{\rho} \bar{U} \cdot \frac{1}{\bar{P}_c}, \quad (8.9)$$

$$\tilde{S} = \bar{\rho} \bar{S} \cdot \frac{\bar{T}_c}{\bar{P}_c}, \quad (8.10)$$

$$\tilde{A} = \frac{\bar{\rho} \bar{A}}{\bar{T}} \cdot \frac{\bar{T}_c}{\bar{P}_c}, \quad (8.11)$$

$$\tilde{H} = \frac{\bar{\rho} \bar{H}}{\bar{T}} \cdot \frac{\bar{T}_c}{\bar{P}_c}, \quad (8.12)$$

$$\tilde{\chi}_T = \bar{T} \chi_T \cdot \frac{\bar{P}_c}{\bar{T}_c \bar{\rho}_c^2}, \quad (8.13)$$

$$\tilde{C}_v = \bar{\rho} \bar{C}_v \cdot \frac{\bar{T}_c}{\bar{P}_c}, \quad (8.14)$$

$$\tilde{C}_p = \bar{\rho} \bar{C}_p \cdot \frac{\bar{T}_c}{\bar{P}_c}, \quad (8.15)$$

$$\tilde{w} = \frac{\bar{w}}{\bar{T}^{1/2}} \left( \frac{\bar{\rho}_c \bar{T}_c}{\bar{P}_c} \right)^{1/2}. \quad (8.16)$$

### 8.2. Fundamental Equation in the Critical Region

The canonical equation in the critical region is written in two parts, a regular part and a nonanalytic part which accounts for the critical anomalies. The former is expressed in terms of the natural variables  $\tilde{T}$  and  $\tilde{\mu}$ , defined in Eqs. (8.5) and (8.6). The latter part is parametrized with the aid of the following transformation.

$$\Delta \tilde{\mu} \equiv \tilde{\mu} - \tilde{\mu}_0(\tilde{T}) = r^{\beta\delta} a \theta (1 - \theta^2), \quad (8.17a)$$

$$\Delta \tilde{T} \equiv \tilde{T} + 1 = r(1 - b^2 \theta^2) - cr^{\beta\delta} a \theta (1 - \theta^2), \quad (8.17b)$$

where  $a$ ,  $b$ , and  $c$  are constants. The variables  $r$  and  $\theta$  replace the original variables  $\tilde{T}$  and  $\tilde{\mu}$  in the nonanalytic part in such a way that there are no singularities in the single-phase region except at the critical point itself.

The fundamental equation reads

$$\tilde{P} = \tilde{P}_{reg} + \tilde{P}_{sc}, \quad (8.18)$$

with

$$\tilde{P}_{reg} = \tilde{P}_0(\tilde{T}) + \Delta \tilde{\mu} + \tilde{P}_{11} \Delta \tilde{\mu} \Delta \tilde{T}, \quad (8.18a)$$

$$\tilde{P}_{sc} = ak_0 r^{\beta(\delta+1)} p_0(\theta) + ak_1 r^{[\beta(\delta+1)+\Lambda]} p_1(\theta). \quad (8.18b)$$

The scaled part of the potential, Eq. (8.18b), is identical with that of light water. For this property, a law of corresponding states appears to hold very well. The functions  $\tilde{\mu}_0(\tilde{T})$  and

TABLE 4. Coefficients for fundamental equation in the critical region

$a =$	23.667
$k_0 =$	1.4403
$k_1 =$	0.2942
$c =$	0.017 76
$b^2 =$	1.3757
$\tilde{P}_1 =$	6.9107
$\tilde{P}_2 =$	-25.2370
$\tilde{P}_3 =$	8.6180
$\tilde{P}_{11} =$	0.547 64
$\tilde{\mu}_c =$	-11.948
$\tilde{\mu}_1 =$	-24.219
$\tilde{\mu}_2 =$	-21.552
$\tilde{\mu}_3 =$	17.308

TABLE 5. Coefficients for polynomials  $p_i(\theta)$

$p_{00} =$	0.586 535
$p_{20} =$	-1.026 243
$p_{40} =$	0.612 903
$p_{01} =$	0.103 25
$p_{21} =$	0.160 32
$p_{41} =$	-0.169 86

$\tilde{P}_0(\tilde{T})$  are defined by

$$\tilde{\mu}_0(\tilde{T}) = \tilde{\mu}_c + \sum_{j=1}^3 \tilde{\mu}_j (\Delta \tilde{T})^j, \quad (8.19)$$

$$\tilde{P}_0(\tilde{T}) = 1 + \sum_{j=1}^3 \tilde{P}_j (\Delta \tilde{T})^j. \quad (8.20)$$

The functions  $p_0(\theta)$  and  $p_1(\theta)$  are polynomials of the form

$$p_i(\theta) = p_{0i} + p_{2i} \theta^2 + p_{4i} \theta^4 \quad (i = 0, 1). \quad (8.21)$$

The critical exponents are

$$\beta = 0.325, \quad \delta = 4.82, \quad \Delta = 0.50. \quad (8.22)$$

The values of the other constants in Eqs. (8.17)–(8.21) are listed in Table 4. The coefficients  $p_{0i}$  of the polynomials  $p_i(\theta)$  defined in Eq. (8.21) are related to the critical exponents  $\beta, \delta, \Delta$  and the constant  $b^2$  as discussed elsewhere<sup>16</sup>; the numerical values of the coefficients  $p_{0i}$  are given in Table 5. Note that the critical exponents  $\beta, \delta, \Delta$ , the constants  $a, k_0, k_1, c, b^2$  and the polynomials  $p_0(\theta), p_1(\theta)$  are identical with those of light water.<sup>1,23</sup>

### 8.3. Thermodynamic Relations

The various thermodynamic properties can be deduced from the fundamental Eq. (7.18) with the aid of the following

TABLE 6. Coefficients  $H_i$

$H_0 =$	1.000 00
$H_1 =$	0.940 695
$H_2 =$	0.578 377
$H_3 =$	-0.202 044

thermodynamic relations.<sup>23</sup>

$$\tilde{\rho} = \left( \frac{\partial \tilde{P}}{\partial \tilde{\mu}} \right)_{\tilde{T}}, \quad (8.23)$$

$$\tilde{U} = \left( \frac{\partial \tilde{P}}{\partial \tilde{T}} \right)_{\tilde{\mu}}, \quad (8.24)$$

$$\tilde{A} = \tilde{\rho} \tilde{\mu} - \tilde{P}, \quad (8.25)$$

$$\tilde{H} = \tilde{P} - \tilde{T} \tilde{U}, \quad (8.26)$$

$$\tilde{S} = \tilde{H} - \tilde{\rho} \tilde{\mu} = -\tilde{A} - \tilde{T} \tilde{U}, \quad (8.27)$$

$$\tilde{\chi}_T = \left( \frac{\partial^2 \tilde{P}}{\partial \tilde{\mu}^2} \right)_{\tilde{T}}, \quad (8.28)$$

$$\left( \frac{\partial \tilde{P}}{\partial \tilde{T}} \right)_{\tilde{\rho}} = \tilde{U} - \frac{\tilde{\rho}}{\tilde{\chi}_T} \frac{\partial^2 \tilde{P}}{\partial \tilde{T} \partial \tilde{\mu}}, \quad (8.29)$$

$$\tilde{C}_v = \tilde{T}^2 \left[ \left( \frac{\partial^2 \tilde{P}}{\partial \tilde{T}^2} \right)_{\tilde{\mu}} - \frac{1}{\tilde{\chi}_T} \left( \frac{\partial^2 \tilde{P}}{\partial \tilde{T} \partial \tilde{\mu}} \right)^2 \right], \quad (8.30)$$

$$\tilde{C}_p = \tilde{C}_v + \frac{\tilde{\chi}_T}{\tilde{\rho}^2} \left[ \tilde{P} - \tilde{T} \left( \frac{\partial \tilde{P}}{\partial \tilde{T}} \right)_{\tilde{\rho}} \right]^2, \quad (8.31)$$

$$\tilde{w} = \left[ \frac{\tilde{C}_p}{\tilde{C}_v} \left( \frac{\partial \tilde{P}}{\partial \tilde{\rho}} \right)_{\tilde{T}} \right]^{1/2}. \quad (8.32)$$

Note: Explicit expressions for the derivatives of the canonical equation can be found in Ref. 23.

## 9. Viscosity

The viscosity is represented by the equation<sup>18</sup>

$$\bar{\eta} = \bar{\eta}_0(\bar{T}) \times \bar{\eta}_1(\bar{T}, \bar{\rho}). \quad (9.1)$$

The first term of the product gives the viscosity in the dilute-gas limit and has the form

$$\bar{\eta}_0(\bar{T}) = \frac{\sqrt{\bar{T}}}{\sum_{i=0}^3 \frac{H_i}{\bar{T}^i}}, \quad (9.2)$$

with the coefficients  $H_i$  given in Table 6. The second multi-

TABLE 7. Coefficients  $H_{ij}$

$i =$	0	1	2	3	4	5
$j = 0$	0.486 419 2	-0.244 837 2	-0.870 203 5	0.871 605 6	-1.051 126	0.345 839 5
1	0.350 900 7	1.315 436	1.297 752	1.353 448	0.0	0.0
2	-0.284 757 2	-1.037 026	-1.287 846	0.0	0.0	-0.021 482 29
3	0.070 137 59	0.466 012 7	0.229 207 5	-0.485 746 2	0.0	0.0
4	0.016 412 20	-0.028 849 11	0.0	0.160 717 1	0.0	-0.009 603 846
5	-0.011 638 15	-0.008 239 587	0.0	0.0	0.0	0.004 559 914
6	0.0	0.0	0.0	-0.003 886 659	0.0	0.0

plicative factor is

$$\bar{\eta}_1(\bar{T}, \bar{\rho}) = \exp \left[ \bar{\rho} \sum_{i=0}^5 \sum_{j=0}^6 H_{ij} \left( \frac{1}{\bar{T}} - 1 \right)^i (\bar{\rho} - 1)^j \right], \quad (9.3)$$

with the coefficients  $H_{ij}$  given in Table 7. Equation (9.1) for the viscosity covers the range

$$P \leq 100 \text{ MPa}, T_{tr} \leq T \leq 775 \text{ K}. \quad (9.4)$$

The viscosity is expected to show a small enhancement in the immediate vicinity of the critical point as it does for  $\text{H}_2\text{O}$ .<sup>24</sup> The effect is small and is neglected in Eq. (9.1).

## 10. Thermal Conductivity

The thermal conductivity is represented by the equation<sup>18</sup>

$$\bar{\lambda} = \bar{\lambda}_0(\bar{T}) + \bar{\lambda}_1(\bar{\rho}) + \bar{\lambda}_2(\bar{T}, \bar{\rho}) + \bar{\lambda}_3(\bar{T}, \bar{\rho}). \quad (10.1)$$

The first term  $\bar{\lambda}_0(\bar{T})$  gives the thermal conductivity in the dilute-gas limit and has the form

$$\bar{\lambda}_0(\bar{T}) = \sum_{i=0}^5 L_{0i} \bar{T}^i. \quad (10.2)$$

The other terms in Eq. (10.1) are defined by the equations

$$\bar{\lambda}_1(\bar{\rho}) = L_{10} [1 - \exp(-2.506\bar{\rho})] + \sum_{i=1}^4 L_{1i} \bar{\rho}^i, \quad (10.3)$$

$$\begin{aligned} \bar{\lambda}_2(\bar{T}, \bar{\rho}) &= L_{20} f g \\ &\times \left[ 1 + g^2 \left\{ \frac{L_{21} f^4}{1 + \exp[60(\tau - 1) + 20]} \right. \right. \\ &\left. \left. + \frac{L_{22} g}{1 + \exp[100(\tau - 1) + 15]} \right\} \right], \quad (10.4) \end{aligned}$$

with

$$\tau = \frac{\bar{T}}{|\bar{T} - 1.1| + 1.1}, \quad (10.4a)$$

$$\bar{\lambda}_3(\bar{T}, \bar{\rho}) = L_{30} f^{6/5} \{1 - \exp[-(0.4\bar{\rho})^{10}]\}. \quad (10.5)$$

The auxiliary functions  $f \equiv f(\bar{T})$  and  $g \equiv g(\bar{\rho})$  are

$$f(\bar{T}) = \exp[\bar{T}(f_0 + f_1 \bar{T})], \quad (10.6)$$

$$g(\bar{\rho}) = \exp[g_0(\bar{\rho} - 1)^2] + g_1 \exp[g_2(\bar{\rho} - \bar{\rho}_0)^2]. \quad (10.7)$$

The values of the coefficients in these equations are given in Table 8. Equation (10.1) for the thermal conductivity covers the range

$$P \leq 100 \text{ MPa}, T_{tr} \leq T \leq 825 \text{ K}. \quad (10.8)$$

TABLE 8. Coefficients for  $\bar{\lambda}(\bar{T}, \bar{\rho})$

$L_{00} =$	1.000 00	$L_{20} =$	$0.354\ 296 \times 10^5$
$L_{01} =$	37.3223	$L_{21} =$	$0.5 \times 10^{10}$
$L_{02} =$	22.5485	$L_{22} =$	3.5
$L_{03} =$	13.0465	$f_0 =$	0.144 847
$L_{04} =$	0.0	$f_1 =$	-5.644 93
$L_{05} =$	-2.607 35	$g_0 =$	-2.800 00
		$g_1 =$	-0.080 738 543
		$g_2 =$	-17.9430
$L_{10} =$	-167.310	$\bar{\rho}_0 =$	0.125 698
$L_{11} =$	483.656	$L_{30} =$	-741.112
$L_{12} =$	-191.039		
$L_{13} =$	73.0358		
$L_{14} =$	-7.574 67		

The thermal conductivity of any fluid, including  $\text{D}_2\text{O}$ , exhibits a critical enhancement in a large range of densities and temperatures around the critical point. This critical enhancement is approximated by the term  $\bar{\lambda}_2(\bar{T}, \bar{\rho})$  in Eq. (10.1). However, while the actual thermal conductivity should become infinite at the critical point as it does for  $\text{H}_2\text{O}$ ,<sup>25</sup> the thermal conductivity calculated from Eq. (10.1) remains finite at the critical point.

## 11. Computer-Program Verification

To assist the user in computer-program verification, we present a short table of values calculated for a number of thermophysical properties. The numerical values in Table 9 are given to six digits which is considered adequate for all practical purposes.

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TABLE 9. Thermophysical property values calculated for selected values of  $\bar{T}$  and  $\bar{\rho}$

$\bar{T}$	$\bar{\rho}$	$\bar{A}$	$\bar{P}$	$\bar{C}_v$	$\bar{\eta}$	$\bar{\lambda}$
0.50	0.0002	-0.264519E + 1	0.440215E - 3	0.142768E + 2	0.197298E + 0	0.270065E + 2
0.50	3.1800	-0.217597E + 0	0.435497E + 1	0.414463E + 2	0.124679E + 2	0.891182E + 3
0.75	0.0295	-0.727350E + 1	0.870308E - 1	0.201586E + 2	0.295148E + 0	0.552168E + 2
0.75	2.8300	-0.429366E + 1	0.447530E + 1	0.334367E + 2	0.304176E + 1	0.869672E + 3
1.00	0.3000	-0.151650E + 2	0.801404E + 0	0.308587E + 2	0.442482E + 0	0.143422E + 3
1.00	1.5500	-0.126455E + 2	0.109763E + 1	0.330103E + 2	0.110384E + 1	0.502847E + 3
1.20	0.4000	-0.254738E + 2	0.149910E + 1	0.236594E + 2	0.609454E + 0	0.160059E + 3
1.20	1.6100	-0.212805E + 2	0.456438E + 1	0.254800E + 2	0.127119E + 1	0.471748E + 3



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