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Viscosity of H_2O in the Critical Region

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Abstract It has been well established that thermodynamic and transport properties of fluids exhibit singular behavior near the critical point. In this article, the theoretical predictions for the enhancement of the viscosity near the critical point are reviewed. It is then shown how these predictions can be used to obtain a representative equation for the viscosity of H_2O in the critical region.

Keywords Critical dynamics · Critical region · H_2O · Steam · Viscosity · Water

1 Introduction

One of the goals of the International Association for the Properties of Water and Steam (IAPWS) is to recommend state-of-the-art procedures for calculating thermodynamic and transport properties of H_2O over a large range of temperatures and pressures. The recommended procedure for calculating the thermodynamic properties of H_2O is provided by the IAPWS Formulation 1995 for the Thermodynamic Properties of Ordinary Water Substance for General and Scientific Use (IAPWS-95) [1–3]. The

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IAPWS formulations for the transport properties of H_2O that are currently available do not extend to the higher temperatures and pressures covered by the IAWPS-95 formulation for the thermodynamic properties. Moreover, they are based on experimental information that was available prior to about 1980 [4,5]. Hence, IAPWS has embarked on a research effort, in collaboration with the International Association for Transport Properties (IATP), to develop new formulations for the transport properties of H_2O. For this purpose, the available experimental information for the transport properties has been updated with the temperatures converted to the current temperature scale (ITS-90) and densities computed from the experimental pressures with the aid of IAPWS-95 [6]. A new correlating equation for the viscosity of H_2O has been developed and has been adopted recently as the IAPWS Formulation 2008 for the Viscosity of Ordinary Water Substance [7]. Research to develop an updated correlating equation for the thermal conductivity of H_2O is still being pursued.

We find it convenient to make the temperature T, the density ρ, and the pressure p dimensionless in terms of their values T_c, ρ_c, p_c at the critical point:

$$\overline{T} = T/T_{\mathrm{c}}, \quad \overline{\rho} = \rho/\rho_{\mathrm{c}}, \quad \overline{p} = p/p_{\mathrm{c}}. \tag{1}$$

The recommended interpolating equation for the viscosity μ has the form:

$$\overline{\mu} \equiv \frac{\mu}{1\mu\mathrm{Pa}\cdot\mathrm{s}} = \overline{\mu}_0(\overline{T}) \times \overline{\mu}_1(\overline{T},\overline{\rho}) \times \overline{\mu}_2(\overline{T},\overline{\rho}). \tag{2}$$

In this equation, $\overline{\mu}_0(\overline{T})$ represents the viscosity in the dilute-gas limit as a function of the reduced temperature $\overline{T}$. The factor $\overline{\mu}_1(\overline{T},\overline{\rho})$ represents the so-called excess or residual viscosity as a function of the reduced density $\overline{\rho}$ and the reduced temperature $\overline{T}$, i.e., the increase of the viscosity over its dilute-gas value at the same temperature T. The viscosity of fluids diverges, albeit weakly, at the critical point [8]. This phenomenon is accounted for by the viscosity-enhancement factor $\overline{\mu}_2(\overline{T},\overline{\rho})$ that is very close to unity everywhere except in a small range of densities and temperatures near the critical point. The recommended interpolating equations for $\overline{\mu}_0(\overline{T})$ and $\overline{\mu}_1(\overline{T},\overline{\rho})$ of H_2O [9] are given in the Appendix to this article. Here we focus our attention on the critical viscosity-enhancement factor $\overline{\mu}_2(\overline{T},\overline{\rho})$. Reliable theoretical predictions for the viscosity in the critical region are available for this work.

2 Asymptotic Critical Power Laws

In the vicinity of a critical point, fluids exhibit large fluctuations in the order parameter associated with the critical phase transition. For fluids near the vapor–liquid critical point, the order parameter can be asymptotically identified with the density, and the corresponding ordering field with the chemical potential. As a consequence, the behavior of thermodynamic and transport properties becomes singular at the critical point. Asymptotically close to the critical point, this singular behavior can be described by power laws with universal critical exponents [10].

A susceptibility χ may be defined as the derivative of the order parameter with respect to the ordering field. In terms of dimensionless variables, the susceptibility is related to the isothermal compressibility such that $\overline{\chi} = \overline{\rho}\,(\partial\overline{\rho}/\partial\overline{p})_{\overline{T}}$ [11]. The spatial extent of the fluctuations is characterized by a correlation length ξ [12]. Along the critical isochore $\overline{\rho} = 1$ at $\overline{T} \geq 1$, the correlation length ξ and the susceptibility $\overline{\chi}$ diverge as a function of $t = \left(\overline{T} - 1\right)/\overline{T}$ according to power laws of the form,

$$\xi \approx \xi_0 t^{-\nu} \quad \text{and} \quad \bar{\chi} \approx T_0 t^{-\gamma}, \tag{3}$$

where ξ_0 and Γ_0 are system-dependent amplitudes, and ν and γ are universal critical exponents. In this article, we use the symbol $\approx$ to designate equality asymptotically close to the critical point. Fluids belong to the critical universality class of Ising-like systems for which [10,13]

$$v \simeq 0.630 \quad \text{and} \quad \gamma \simeq 1.239. \tag{4}$$

From Eq. 3, it follows that

$$\xi \approx \xi_0(\overline{\chi}/\Gamma_0)^{\nu/\gamma}. \tag{5}$$

Although Eq. 5 is strictly valid only for $\rho = \rho_c$, it is also used as an approximation for $\rho \neq \rho_c$ [14]. The viscosity is predicted to diverge as [15]

$$\overline{\mu} \approx \overline{\mu}_b\,(Q_0\xi)^{x_\mu}, \tag{6}$$

where Q_0 is an effective wave number that determines the amplitude of the power-law divergence of the viscosity, while x_μ is a universal dynamic critical exponent, for which we have adopted the most recent theoretical value [16],

$$x_\mu \simeq 0.068. \tag{7}$$

In Eq. 6, $\overline{\mu}_b$ is a so-called background viscosity, i.e., the viscosity in the absence of any critical fluctuations. From Eq. 2 it follows that

$$\overline{\mu}_b\left(\overline{T}, \overline{\rho}\right) = \overline{\mu}_0(\overline{T}) \times \overline{\mu}_1(\overline{T}, \overline{\rho}). \tag{8}$$

The viscosity exhibits a so-called multiplicative critical anomaly, i.e., the critical enhancement is proportional to the background viscosity $\overline{\mu}_b$ [17].

3 Crossover Equation for Critical Viscosity Enhancement

Equation 6 represents the divergent behavior of the viscosity asymptotically close to the critical point. To apply the theory to experimental data, one needs a theoretically based “crossover” equation that incorporates not only the asymptotic power law of Eq. 6 but also reduces to the normal background viscosity $\overline{\mu}_b$ away from the critical point. This problem has been solved by Bhattacharjee and coworkers [15,18], who derived the following crossover equation to include the critical behavior of the viscosity:

$$\overline{\mu} = \overline{\mu}_{\mathrm{b}} \times \overline{\mu}_2 \quad \text{with} \quad \overline{\mu}_2 = \exp\left(x_\mu Y\right). \tag{9}$$

The function Y is defined by

$$\begin{aligned} Y = {} & \frac{1}{12}\sin(3\psi_{\mathrm{D}}) - \frac{1}{4q_{\mathrm{C}}\xi}\sin(2\psi_{\mathrm{D}}) + \frac{1}{(q_{\mathrm{C}}\xi)^2}\left[1 - \frac{5}{4}(q_{\mathrm{C}}\xi)^2\right]\sin(\psi_{\mathrm{D}}) \\ & - \frac{1}{(q_{\mathrm{C}}\xi)^3}\left\{\left[1 - \frac{3}{2}(q_{\mathrm{C}}\xi)^2\right]\psi_{\mathrm{D}} - \left|(q_{\mathrm{C}}\xi)^2 - 1\right|^{3/2} L(w)\right\} \end{aligned} \tag{10}$$

with

$$\psi_{\mathrm{D}} = \arccos\left[\left(1 + q_{\mathrm{D}}^2\xi^2\right)^{-1/2}\right], \tag{11}$$

and with the function $L(w)$ given by

$$L(w) = \begin{Bmatrix} \ln\frac{1+w}{1-w}, & \text{for } q_{\mathrm{C}}\xi > 1 \\ 2\arctan|w|, & \text{for } q_{\mathrm{C}}\xi \le 1 \end{Bmatrix}. \tag{12}$$

The variable w is defined as

$$w = \left|\frac{q_{\mathrm{C}}\xi - 1}{q_{\mathrm{C}}\xi + 1}\right|^{1/2} \tan\left(\frac{\psi_{\mathrm{D}}}{2}\right). \tag{13}$$

The function Y contains two system-dependent constants, namely, the wave numbers q_{C} and q_{D}. Asymptotically close to the critical point, i.e., in the limit of large ξ, Eq. 9 reproduces Eq. 6 with an amplitude Q_0 that is related to q_{C} and q_{D} such that [18]

$$Q_0^{-1} = (q_{\mathrm{C}}^{-1} + q_{\mathrm{D}}^{-1})e^{4/3}/2. \tag{14}$$

The wave number q_{C} is related to a background contribution to the decay rate of the critical fluctuations and is given by

$$q_{\mathrm{C}} = \frac{k_{\mathrm{B}}T_{\mathrm{c}}^2}{16\mu_{\mathrm{b}}^{\mathrm{c}}\lambda_{\mathrm{b}}^{\mathrm{c}}p_{\mathrm{c}}^2}\frac{\Gamma_0}{\xi_0^2}\left(\frac{\partial p}{\partial T}\right)_{\rho=\rho_{\mathrm{c}}}^2, \tag{15}$$

where k_{B} is Boltzmann's constant, and $\mu_{\mathrm{b}}^{\mathrm{c}}$ and $\lambda_{\mathrm{b}}^{\mathrm{c}}$ are the values of the background viscosity and background thermal conductivity, respectively, at the critical point, while $(\partial p/\partial T)_{\rho=\rho_{\mathrm{c}}}$ is the slope of the critical isochore at the critical temperature. The wave number q_{D} represents a "Debye" cutoff of the mode-coupling integrals for critical dynamics and is the only adjustable parameter in the theory.

For small ξ, the function Y approaches zero, so that $\overline{\mu}$ approaches $\overline{\mu}_{\mathrm{b}}$ in this limit. Around $\xi = 0$, the function Y has a Taylor expansion of the form,

$$Y = \frac{1}{5} q_{\mathrm{C}}\xi \ (q_{\mathrm{D}}\xi)^5 \left(1 - q_{\mathrm{C}}\xi + (q_{\mathrm{C}}\xi)^2 - \frac{765}{504} (q_{\mathrm{D}}\xi)^2\right). \tag{16}$$

The approximations in the derivation of Eq. 9 for the critical enhancement of the viscosity have been discussed by Luettmer-Strathmann et al. [19]. One of the approximations is that the isobaric specific heat capacity c_p in the mode-coupling integral for the viscosity has been replaced by the difference $c_p - c_V$, where c_V is the isochoric specific heat capacity. These approximations are well justified in the small region around the critical point where a critical viscosity enhancement is observed.

Equation 5 represents the behavior of the correlation length ξ near the critical point. In the theory of critical phenomena, ξ is to be interpreted as that part of the actual correlation length associated with the long-range critical fluctuations. Hence, the correlation length ξ in Eq. 10 should vanish far away from the critical point. To accomplish this goal, we have adopted a procedure proposed by Olchowy and Sengers [20] by generalizing Eq. 5 to

$$\xi = \xi_0 \left(\frac{\Delta\overline{\chi}}{\Gamma_0}\right)^{\nu/\gamma} \tag{17}$$

in terms of $\Delta\overline{\chi}$ (≥ 0), which is defined by

$$\Delta\overline{\chi} = \left[\overline{\chi}\left(\overline{T}, \overline{\rho}\right) - \overline{\chi}\left(\overline{T}_{\mathrm{R}}, \overline{\rho}\right) \frac{\overline{T}_{\mathrm{R}}}{\overline{T}}\right] . \tag{18}$$

In Eq. 18, $\overline{T}_{\mathrm{R}}$ is a reference temperature sufficiently high above the critical temperature where the critical fluctuations can be assumed to be small. In practice one may select $\overline{T}_{\mathrm{R}} = 1.5$ [19–21]. Furthermore, $\Delta\overline{\chi}$ is to be taken to be zero when the right-hand side of Eq. 18 becomes negative. This procedure assures that the correlation length ξ in Eqs. 10 and 16 will vanish far away from the critical point, while Eq. 17 still reduces to Eq. 5 asymptotically close to the critical point.

An accurate experimental determination of the viscosity of fluids near their critical point is hampered by the presence of gravitationally induced density profiles [22]. To avoid this complication, the viscosity of xenon near the critical density and critical temperature has been measured by Berg et al. [23] at low-gravity conditions in the Space Shuttle. In the hydrodynamic limit of zero frequency, the experimental viscosity data are well represented by Eq. 9 with an experimental value for the critical exponent x_μ that is in agreement with the theoretical value 0.068 within its uncertainty. We conclude that the crossover Eq. 9 for the critical behavior of the viscosity has a sound theoretical basis and has been validated experimentally.

4 Application to H_2O

The currently accepted values for the critical parameters of H_2O are [1,11]

$$T_c = 647.096\,\text{K},\ \rho_c = 322\,\text{kg}\cdot\text{m}^{-3},\ p_c = 22.064\,\text{MPa}. \tag{19}$$

The amplitudes in Eq. 3 for the correlation length ξ and the dimensionless susceptibity $\overline{\chi}$ of H_2O are [11,24]

$$\xi_0 \simeq 0.13\,\text{nm},\ \Gamma_0 \simeq 0.06. \tag{20}$$

With $(\partial p/\partial T)_{\rho=\rho_c} \simeq 0.267\,\text{MPa}\cdot\text{K}^{-1}$ [11] and $\mu_b^c = 39.3\,\mu\text{Pa}\cdot\text{s}$ as calculated from Eq. 8 at $T = T_c$ and $\rho = \rho_c$, along with $\lambda_b^c = 0.197\,\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ as estimated from the current interpolating equation for the thermal conductivity of H_2O [1,25], we find from Eq. 15

$$q_C^{-1} \simeq 1.9\ \text{nm}. \tag{21}$$

The equation for the critical enhancement of the viscosity of H_2O is now completely specified except for one system-dependent constant, namely, the wave number q_D.

5 Comparison with Experimental Data

A detailed experimental study of the viscosity of H_2O in the critical region was made by Rivkin et al. [26]. In this experiment, the kinematic viscosity μ/ρ was determined by measuring the flow rate through a capillary with an internal diameter of 0.3 mm and a length of 50 cm as a function of the pressure drop over the capillary. In principle, the method can lead to some complications due to the large compressibility near the critical point. However, the investigators made measurements with various pressure differences ranging from 13 kPa to 1.1 kPa and verified that the measured kinematic viscosity became independent of the applied pressure difference below 1.6 kPa [27]. The measurements seem to have been executed with great care, and we have adopted the values determined by Rivkin et al. for the kinematic viscosity with an uncertainty estimate of 2 %, compared to 1 % suggested by the authors. Another attempt to measure the viscosity of H_2O was made by Oltermann [28] with an oscillating-disk viscometer. Near the critical density, his data are consistent with those of Rivkin et al., but the agreement is not good at lower and higher densities. We have adopted the data of Rivkin et al. as the primary data source for characterizing the behavior of the viscosity in the critical region. The same decision was earlier made by Watson et al. [4] in developing the previous correlating equation for the viscosity of H_2O in the critical region.

Rivkin et al. obtained the kinematic viscosity as a function of temperature in terms of the International Practical Temperature of 1948 (IPTS-48) and of pressure. We have converted the experimental temperatures to ITS-90, calculated the densities from the

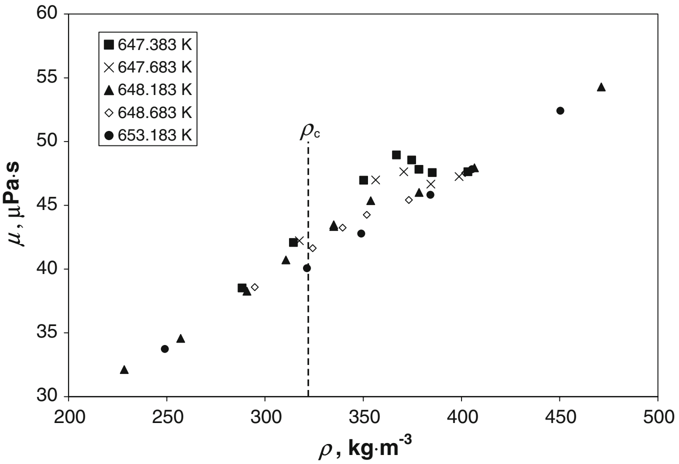

Fig. 1 Viscosity data obtained by Rivkin et al. [26] as a function of density with the density calculated from the IAPWS-95 equation of state and the temperatures converted to ITS-90

IAPWS-95 formulation, and converted the experimental values for the kinematic viscosity μ/ρ into values for the dynamic viscosity μ. The viscosities thus obtained are plotted as a function of density in Fig. 1. The figure shows that μ exhibits an enhancement at a density substantially larger than the critical density ρ_c. Watson et al. [4] adopted the hypothesis that this apparent shift in the location of the maximum of the viscosity enhancement may be due to an offset of the temperature or, more precisely, to an inaccurate value of the temperature difference $T - T_c$ attributed to the experimental data. Such an offset can result from a temperature difference between the sample in the cell and the location of the thermometer, mechanical stresses in thermocouple connections, or similar problems, while the critical temperature can depend on the purity of the sample. Adopting the best available value for the critical temperature at that time and a scaled equation of state, Watson et al. showed that the data did become consistent with the expected diverging critical behavior of the viscosity, if the temperatures reported by Rivkin et al. were shifted by +0.040 K [4]. We have performed a statistical analysis to determine the optimum temperature shift using the IAPWS-95 formulation for the equation of state, which assumes a critical temperature $T_c = 647.096\,\mathrm{K}$ (ITS-90). This analysis yielded a temperature shift of +0.051 K with a standard deviation of 0.002 K. Figure 2 shows the critical enhancement of the viscosity deduced from the measurements of Rivkin et al. when the temperatures on ITS-90 plus the offset of 0.051 K are used to compute the densities with IAPWS-95. The curves in this figure represent the values calculated from the theoretical Eq. 9 with

$$q_D^{-1} \simeq 1.1\ \mathrm{nm}. \tag{22}$$

We conclude that, with the temperature correction, the data are consistent with the theoretical predictions for the behavior of the viscosity in the critical region. The reported uncertainty in the experimental temperature is 0.02 K and the reported uncertainty in

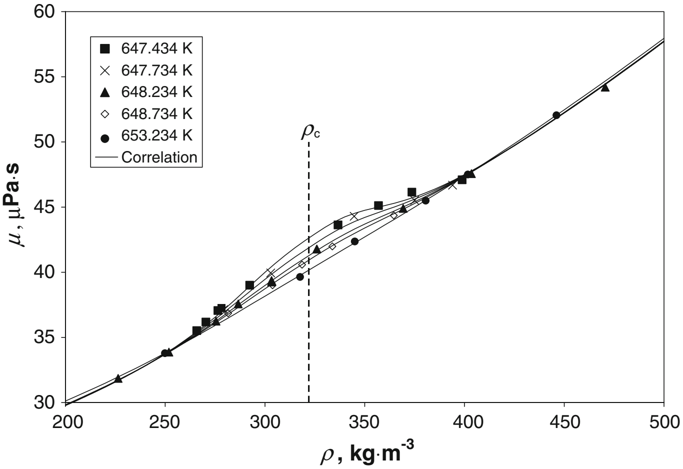

Fig. 2 Viscosity data obtained by Rivkin et al. [26] as a function of density with the density calculated from the IAPWS-95 equation and the temperatures converted to ITS-90 plus 0.051 K. Curves represent the proposed correlation equation for the viscosity in the critical region

the pressure is 0.01 MPa, which is equivalent to the effect of a temperature uncertainty of 0.04 K on density (based on the 0.267 MPa $\cdot$K^{-1} slope of the vapor-pressure curve at the critical point). The uncertainty in the critical temperature in the IAPWS-95 equation of state is 0.100 K [29]. The combined uncertainty from these three sources is 0.110 K. Hence, the temperature offset of 0.051 K is well within the uncertainty of $T - T_c$ to be attributed to the experimental data of Rivkin et al. In principle, the density where the critical enhancement has a maximum can be moved to the critical density by applying either a temperature shift or a pressure shift. We have verified that replacing the temperature shift by a pressure shift does not yield satisfactory agreement between theory and experiment. We also found that an optimized combination of temperature and pressure shifts does not significantly improve agreement between theory and experiment relative to that found for the temperature shift alone.

One comment should be made concerning the technical application of Eq. 9 for the behavior of the viscosity in the critical region. While the function Y defined by Eq. 10 does become zero in the limit as ξ goes to zero, some individual terms diverge in this limit. Hence, Eq. 10 is no longer suitable for numerical calculations at small values of ξ; specifically, for $\xi \leq 0.3817016416$ nm it is recommended that the function Y be evaluated by using its truncated Taylor expansion given by Eq. 16.

Figure 3 shows a plot of the multiplicative critical viscosity enhancement μ/μ_b as a function of the correlation length ξ. The critical enhancement of the viscosity is significant only in a very small region in temperatures and densities around the critical point. Although exactly at the critical point the viscosity is infinite, the enhancement term, $\overline{\mu_2} = \exp(x_\mu Y)$ in Eq. 9, contributes an amount greater than the uncertainty of 2 % of our correlating equation only at values of ξ larger than $\xi = 3.51$ nm. This condition is satisfied only at temperatures and densities within the following boundaries:

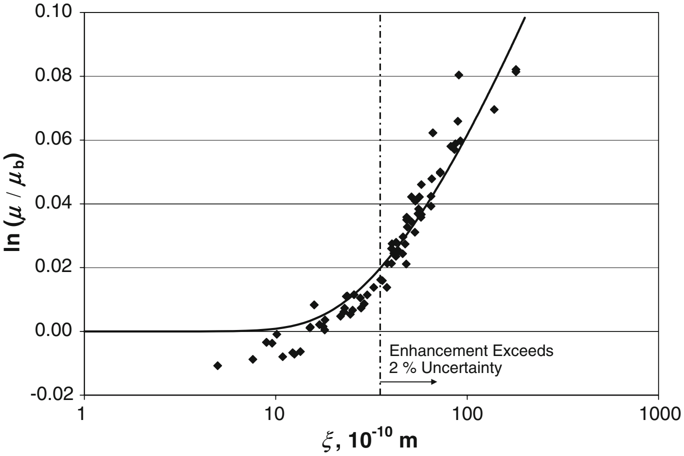

Fig. 3 Multiplicative critical viscosity enhancement $\mu_2 = \mu/\mu_b$ plotted as a function of the correlation length ξ. Symbols represent the experimental data of Rivkin et al. [26] with densities calculated from the IAPWS-95 equation and the temperatures converted to ITS-90 plus 0.051 K. Curve represents the theoretical expression for this viscosity enhancement

$$\begin{aligned} &645.91\,\mathrm{K} < T < 650.77\,\mathrm{K}, \\ &245.8\,\mathrm{kg}\cdot\mathrm{m}^{-3} < \rho < 405.3\,\mathrm{kg}\cdot\mathrm{m}^{-3} \end{aligned} \tag{23}$$

Thus, the critical enhancement is significant only within the boundaries specified by Eq. 23. Outside this region, the critical viscosity enhancement is always less than the uncertainty of the formulation. For further details, the reader is referred to a forthcoming publication [9].

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Appendix: Equations for the Background Viscosity μ_b [7,9]

The background viscosity is given by

$$\overline{\mu}_b\left(\overline{T},\overline{\rho}\right) = \overline{\mu}_0(\overline{T}) \times \overline{\mu}_1(\overline{T},\overline{\rho}). \tag{A1}$$

The viscosity of the dilute gas is given by

$$\overline{\mu}_0\left(\overline{T}\right) = \frac{100\sqrt{\overline{T}}}{\sum_{i=0}^{3}\frac{H_i}{\overline{T}^i}}. \tag{A2}$$

Table 1 provides the coefficients for the dilute-gas viscosity.

Table 1 Coefficients H_i for $\overline{\mu_0}(\overline{T})$

i	H_i
0	1.67752
1	2.20462
2	0.6366564
3	−0.241605

Table 2 Coefficients H_{ij} for $\overline{\mu_1}(\overline{T}, \overline{\rho})$[a]

i	j	H_{ij}
0	0	5.20094×10^{-1}
1	0	8.50895×10^{-2}
2	0	-1.08374
3	0	-2.89555×10^{-1}
0	1	2.22531×10^{-1}
1	1	9.99115×10^{-1}
2	1	1.88797
3	1	1.26613
5	1	1.20573×10^{-1}
0	2	-2.81378×10^{-1}
1	2	-9.06851×10^{-1}
2	2	-7.72479×10^{-1}
3	2	-4.89837×10^{-1}
4	2	-2.57040×10^{-1}
0	3	1.61913×10^{-1}
1	3	2.57399×10^{-1}
0	4	-3.25372×10^{-2}
3	4	6.98452×10^{-2}
4	5	8.72102×10^{-3}
3	6	-4.35673×10^{-3}
5	6	-5.93264×10^{-4}

[a] *Note*: Coefficients H_{ij} omitted from the table are identically equal to zero

The excess contribution to the viscosity is given by

$$\overline{\mu}_1\left(\overline{T}, \overline{\rho}\right) = \exp\left[\overline{\rho} \sum_{i=0}^{5} \left(\frac{1}{\overline{\overline{T}}} - 1\right)^i \sum_{j=0}^{6} H_{ij}\left(\overline{\rho} - 1\right)^j\right]. \tag{A3}$$

Table 2 provides the coefficients for the excess viscosity.

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