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Reference Correlation of the Viscosity of *n*-Heptane from the Triple Point to 600 K and up to 248 MPa

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This paper contains a new wide-ranging correlation for the viscosity of *n*-heptane based on critically evaluated experimental data. The correlation is valid from the triple point (182.55 K) to 600 K, and at pressures up to 248 MPa. The estimated uncertainty at a 95% confidence level is 3.5% over the whole range (with the exception of the near-critical region). Along the saturated liquid curve, the estimated uncertainty is 1% below 292 K, 0.6% in the region from 292 to 346 K, rising to 2% between 346 and 363 K, and 0.3% for the low-density gas at temperatures from 317 to 600 K and pressures to 0.3 MPa. © *2014 by the U.S. Secretary of Commerce on behalf of the United States. All rights reserved.* [http://dx.doi.org/10.1063/1.4875930]

Key words: critical phenomena; *n*-heptane; transport properties; viscosity.

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1. Introduction

In a series of recent papers, new reference correlations were reported for the thermal conductivity of a series of fluids[1–8] and for the viscosity of *n*-hexane,[9] covering a wide range of temperature and pressure. In this paper, the methodology adopted for the viscosity of *n*-hexane[9] is extended to developing a new reference correlation for the viscosity of *n*-heptane.

The goal of this work is to critically assess the available literature data, and to provide a wide-ranging correlation for the viscosity of *n*-heptane that is valid over gas, liquid, and supercritical states and that incorporates densities provided by a short-form Helmholtz equation of state of Span and Wagner.[10]

2. Methodology

The viscosity η can be expressed[9,11] as the sum of four independent contributions, as

$$\eta(\rho,T) = \eta_0(T) + \eta_1(T)\rho + \Delta\eta(\rho,T) + \Delta\eta_c(\rho,T), \tag{1}$$

where ρ is the molar density, T is the absolute temperature, and the first term, $\eta_0(T) = \eta(0,T)$, is the contribution to the viscosity in the dilute-gas limit, where only two-body molecular interactions occur. The linear-in-density term, $\eta_1(T)\rho$, known as the initial density dependence term, can be separately established with the development of the Rainwater-Friend theory[11–13] for the transport properties of moderately dense gases. The critical enhancement term, $\Delta\eta_c(\rho,T)$, arises from the long-range density fluctuations that occur in a fluid near its critical point, which contribute to divergence of the viscosity at the critical point. Finally, the term $\Delta\eta(\rho,T)$, the residual term, represents the contribution of all other effects to the viscosity of the fluid at elevated densities, including many-body collisions, molecular-velocity correlations, and collisional transfer.

The identification of these four separate contributions to the viscosity and to transport properties in general is useful because it is possible, to some extent, to treat $\eta_0(T)$, $\eta_1(T)$, and $\Delta\eta_c(\rho,T)$ theoretically. In addition, it is possible to derive information about both $\eta_0(T)$ and $\eta_1(T)$ from experiment. In contrast, there is little theoretical guidance concerning the residual contribution, $\Delta\eta(\rho,T)$, and therefore its evaluation is based entirely on an empirical equation obtained by fitting experimental data.

The analysis described above should be applied to the best available experimental data for the viscosity. Thus, a prerequisite to the analysis is a critical assessment of the experimental data. For this purpose, two categories of experimental data are defined: primary data, employed in the development of the correlation, and secondary data, used simply for comparison purposes. According to the recommendation adopted by the Subcommittee on Transport Properties (now known as The International Association for Transport Properties) of the International Union of Pure and Applied Chemistry, the primary data are identified by a well-established set of criteria.[14] These criteria have been successfully employed to establish standard reference values for the viscosity and thermal conductivity of fluids over wide ranges of conditions, with uncertainties in the range of 1%. However, in many cases, such a narrow definition unacceptably limits the range of the data representation. Consequently, within the primary data set, it is also necessary to include results that extend over a wider range of conditions, albeit with a poorer accuracy, provided they are consistent with other more accurate data or with theory. In all cases, the accuracy claimed for the final recommended data must reflect the estimated uncertainty in the primary information.

3. The Correlation

Recently Sagdeev *et al.*[15] measured the viscosity of heptane and provided a literature search for measurements in the liquid phase. We identified some additional data sources, included the vapor phase, and in Table 1 summarize, to the best of our knowledge, the experimental measurements[15–137] of the viscosity of *n*-heptane including sample purity and the uncertainties ascribed by the original authors. 121 sets are included in the table. From these sets, 11 were considered as primary data. Table 1 shows that most of the secondary data sets consist of a few measurements over a limited temperature range near ambient pressure.

Absolute vibrating-wire instruments were employed by the group of Wakeham[21] in London and the group of Assael[22] in Thessaloniki. These data sets, backed by a full theoretical analysis, extend over a wide range of conditions with an uncertainty of 0.5%, and thus are primary data. The measurements of Vogel and Holdt[23] were performed in the very-low-density vapor range in an oscillating-disk viscometer designed exactly for that region. These measurements were obtained with an uncertainty of 0.3% and were also part of the primary dataset. Knapstad *et al.*[24] performed very accurate viscosity measurements at atmospheric pressure in an absolute oscillating-cup viscometer with an uncertainty of 0.3%–0.5%, and these measurements were also included in the primary data set. Capillary viscometers, but of slightly higher uncertainty, were employed by Abdulagatov and Azizov,[16] Abdulagatov and Rasulov,[19,20] and Zéberg-Mikkelsen *et al.*[17] These three sets, performed with an uncertainty of 1.6%, 1.0%–2.0%, and 1.2%

TABLE 1. Viscosity measurements of *n*-heptane.

1st author	Year publ.	Technique employed[a]	Purity (%)	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Primary data							
Sagdeev[15]	2013	FB	99.997	1.5–2.0	52	298–470	0.09–245
Abdulagatov[16]	2006	Cap	99.95	1.6	15	298–473	0.1–10
Zéberg-Mikkelsen[17]	2006	FB/Cap	99.5	1.0–2.0	24	293–353	0.1–100
Pensado[18]	2005	RB	99.5	2.0	84	303–353	0.1–60
Abdulagatov[19,20]	1992	Cap	99.98	1.2[b]	148	298–548	0.1–50
Assael[21]	1992	VBW-Abs	99.5	0.5	46	303–348	0.1–248
Assael[22]	1991	VBW-Abs	99.5	0.5	28	303–323	0.1–69
Vogel[23]	1991	OD	99.96	0.3	80	317–632	0.01–0.21
Knapstad[24]	1989	OCup-Abs	99.5	0.3–0.5	9	292–346	0.1
Kessel'man[25]	1976	Cap	na	1.0	7	211–302	0.1
Giller[26]	1949	Cap	Pure grade	0.5	15	183–293	0.1
Secondary data							
Guerrero[27]	2011	Cap	99.0	0.1	3	283–313	0.1
Rathnam[28]	2010	Cap	99.0	0.8	3	303–313	0.1
Yang[29]	2010	Cap	99.0	na	4	298–313	0.1
Zivkovic[30]	2010	VBT	99.5	1.5	5	303–323	0.1
El-Sayed[31]	2009	Cap	99.0	0.1	2	303–313	0.1
Fang[32]	2008	Cap	99.0	0.8	5	288–308	0.1
Totchasov[33]	2008	Cap	na	0.1	4	293–313	0.1
Landaverde-Cortes[34]	2007	Cap	99.6	1.0	9	273–313	0.1
Reddy[35]	2007	Cap	99.5	0.3	1	303.15	0.1
Wang[36]	2007	Cap	99.98	0.1	5	293–313	0.1
Baragi[37]	2006	Cap	99.0	0.26	3	298–308	0.1
Iloukhani[38]	2006	Cap	99.0	0.5	1	298	0.1
Reddy[39]	2006	Cap	na	0.3	1	303	0.1
Al-Jimaz[40]	2005	Cap	99.0	na	3	293–303	0.1
Ma[41]	2005	Cap	98.5	0.8	7	293–343	0.1
Yang[42]	2004	Cap	99.5	0.8	6	298–343	0.1
Yang[43]	2004	Cap	99.5	0.8	7	293–343	0.1
Ma[44]	2004	Cap	na	1–2	18	298–403	0.1,5.1
Oswal[45]	2003	Cap	99.8	0.5	1	303	0.1
Queimada[46]	2003	RB	99.0	2.0	4	293–323	0.1
Peng[47]	2002	Cap	99.6	0.8	1	298	0.1
Matos[48]	2001	Ubb	99.8	0.2	4	283–313	0.1
Nayak[49]	2001	Cap	99.7	2.0	3	298–308	0.1
Sastry[50]	1999	Cap	99.8	0.3	3	298–318	0.1
Dandekar[51]	1998	Cap	na	na	1	293	0.1
Jimenez[52]	1998	Cap	99.0	na	4	293–308	0.1
Wu[53]	1998	Cap	99.0	0.1	4	293–313	0.1
Aminabhavi[54]	1997	Cap	99.8	0.3	3	298–308	0.1
Baylaucq[55]	1997	FB	99.0	2.0	18	303–343	0.1–100
Baylaucq[56]	1997	FB	99.0	2.0	6	323	0.15–100
Nath[57]	1997	Cap	na	1.0	1	298	0.1
Orge[58]	1997	RB	99.0	1.0	1	298	0.1
Will[59]	1997	DLS	99.5	2.0	8	293–353	0.1
Abdulagatov[60]	1996	Cap	99.98	1.2[b]	49	298–540	0.006–2.7
Aminabhavi[61]	1996	Cap	99.5	0.3	3	298–308	0.1
Sastry[62,63]	1996	Cap	na	0.8	2	298–308	0.1
Sastry[64]	1996	Cap	na	0.8	2	303–313	0.1
Aminabhavi[65]	1995	Cap	99.6	0.3	4	298–313	0.1
Aucejo[66]	1995	Cap	99.0	0.6	1	298	0.1
Aminabhavi[67]	1994	Cap	99.5	0.3	5	298–318	0.1
Dymond[68]	1994	FB	na	0.4	1	298	0.1
Pandey[69]	1994	Cap	na	0.1	1	298	0.1
Papaioannou[70]	1994	FB	99.8	1.0–2.5	18	298	0.1–51
Blanco[71]	1993	Cap	99.5	0.8	4	288–318	0.1
Cooper[72]	1991	Cap	na	na	1	293	0.1
Kanti[73]	1991	FB	99.0	1.0	15	313–353	0.1–40
Papaioannou[74]	1991	Cap	99.0	0.4	1	298	0.1
Zhang[75]	1991	FB	na	2.0	7	298	0.1–29.5
Chevalier[76]	1990	Cap	na	0.1	1	298	0.1
Fermeglia[77]	1990	Cap	99.0	na	1	298	0.1
Iwahashi[78]	1990	Cap	99.9	na	2	298–323	0.1

TABLE 1. Viscosity measurements of *n*-heptane.—Continued

1st author	Year publ.	Technique employed[a]	Purity (%)	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Brunson[79]	1989	DLS	99.0	1.0	14	292–348	0.1
Kouris[80]	1989	Cap	99.0	0.4	1	298	0.1
Schrodt[81]	1989	Cap	na	na	1	297	0.1
Chandrashekhar[82]	1988	Cap	na	1.0	1	303	0.1
Rao[83]	1988	Cap	na	0.5	1	298	0.1
Wakefield[84]	1988	Cap	99.0	0.5	2	303–308	0.1
Celda[85]	1987	Cap	99.0	0.3	1	293	0.1
Senanayake[86]	1987	Cap	99.9	na	1	298	0.1
Awwad[87]	1986	Cap	99.5	na	1	298	0.1
Awwad[88]	1985	Cap	99.5	na	1	298	0.1
Martin[89]	1983	Cap	na	na	5	303–343	0.1
Kashiwagi[90]	1982	TCr	99.0	2.0	48	298–348	0.1–110
Sreenivasulu[91]	1982	Cap	na	0.5	1	303	0.1
Rabe[92]	1981	Cap	na	na	47	185–300	0.1–10
Dusart[93]	1978	na	na	na	1	298	0.1
Gouel[94]	1978	RB	na	5.0	52	293–392	0.1–40
Gunchuk[95]	1977	Cap	na	na	13	208–363	0.1
Öz[96]	1976	Cap	99.6	0.3	5	288–308	0.1
Badalov[97]	1975	Cap	na	na	73	290–540	0.1–49
Mussche[98]	1975	FB	na	1.0	2	293,298	0.1
Diaz Pena[99]	1974	Cap	99.5	2.0	10	333–423	0.02–0.37
Moore[100]	1974	Cap	99.0	na	4	293–313	0.1
Ratkovics[101]	1974	na	na	na	1	293	0.1
Shikhaliev[102]	1974	Cap	99.35	na	14	223–353	0.1
Nauruzov[103]	1973	RB	na	na	4	268–298	0.1
Zhigunov[104]	1972	Cap	na	3.0	3	293–313	0.1
Konobeev[105]	1970	Cap	na	2.0	3	293–333	0.1
Kuss[106]	1969	Cap	na	0.2	6	313	0.1–147
Darmois[107]	1967	na	na	na	1	293	0.1
Carmichael[108]	1966	RCyl	99.98	1.0	2	310,344	0.0027
Bidlack[109]	1964	Cap	na	na	1	298	0.1
Bagdasarian[110]	1964	na	na	na	7	223–353	0.1
Agaev[111]	1963	Cap	na	na	1	540	0.1
Golubev[112]	1963	Cap	na	1.0	2	540	0.1–2.7
McAllister[113]	1960	na	na	0.5	1	311	0.1
Albright[114]	1956	RB	99.0	na	2	397–423	0.1
Ledneva[115]	1956	Cap	na	na	11	293–493	0.004–1.4
Belinskaya[116]	1955	na	na	na	11	233–333	0.1
Golik[117]	1955	Cap	na	na	7	303–423	0.1–0.4
Hammond[118]	1955	Cap	na	1.0	1	298	0.1
Koshkin[119]	1955	na	na	na	12	223–333	0.1
Lambert[120]	1955	Cap	na	1.0	4	338–364	0.03–0.08
Toporov[121]	1955	Cap	na	na	3	293–333	0.1
Johnson[122]	1953	Cap	99.8	na	6	199–255	0.1
Koshkin[123]	1953	na	na	na	6	223–273	0.1
Doolittle[124]	1951	Cap	99.74	na	4	263–371	0.1
Fawcett[125]	1946	na	na	na	1	293	0.1
Geist[126]	1946	Cap	na	0.5	3	273–313	0.1
Khalilov[127]	1939	Cap	na	na	48	293–539	0.005–2.7
Kyropoulos[128]	1939	RCon	na	na	3	293–362	0.1
Timmermans[129]	1935	Cap	na	na	2	288,303	0.1
Melaven[130]	1932	Cap	na	na	4	373–525	0.1
Shepard[131]	1931	Cap	na	0.5	1	298	0.1
Tausz[132]	1930	na	na	na	1	293	0.1
Smyth[133]	1928	Cap	na	na	1	293	0.1
Lewis[134]	1925	Cap	na	0.5	1	298	0.1
Miller[135]	1924	Cap	na	na	2	282–293	0.1
Batschinski[136]	1913	Cap	na	na	10	273–363	0.1
Thorpe[137]	1894	Cap	na	na	13	279–365	0.1

[a] Abs, Absolute; Cap, Capillary; DLS, Dynamic Light Scattering; FB, Falling Body; na, not available; OCup, Oscillating Cup; OD, Oscillating Disk; RB, Rolling Ball; RCyl, Rotating Cylinders; RCon, Rotating Cones; TCr, Torsional Crystal; VBW, Vibrating Wire; VBT, Vibrating Tube.

[b] Rising to 5% at the critical region.

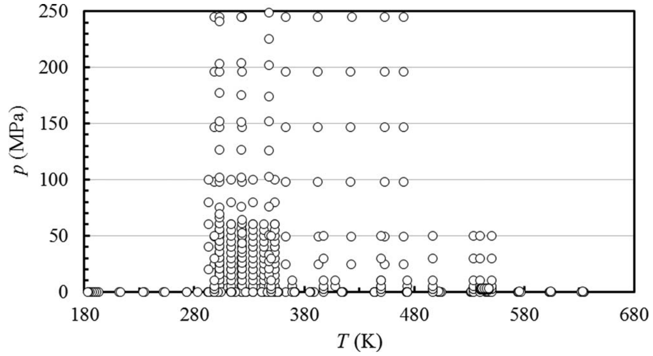

FIG. 1. Temperature-pressure ranges of the primary experimental viscosity data for *n*-heptane.

respectively, were included in the primary data set to extend the temperature and pressure range of the other aforementioned sets. Unfortunately, there are many errors in the data tables in both the Russian-language[19] and English-language[20] versions of the 1992 work of Abdulagatov and Rasulov; we therefore include the correct values (used in this work) in the Appendix. A falling-body viscometer was employed for the high-pressure (to 100 MPa) viscosity measurements of Zéberg-Mikkelsen *et al.*[17] A falling-body viscometer was also employed by Sagdeev *et al.*[15] with a 2% uncertainty for measurements up to 245 MPa pressure. These measurements agree well with the other high-pressure sets. The measurements of Pensado *et al.*[18] were performed in a rolling-ball viscometer with an uncertainty of 2% and extend up to 60 MPa. Although rolling-ball viscometers are not usually taken as primary instruments, the laboratory of Professor J. Fernández has produced accurate measurements of very low uncertainty; thus these measurements were also included in the primary data set. Finally, the capillary measurements of Kessel'man *et al.*[25] and of Giller and Drickamer,[26] that extend down to 211 and 183 K, respectively, were also included in the primary dataset to extend the temperature range to low temperatures.

Figures 1 and 2 show the ranges of the primary measurements outlined in Table 1, and the phase may be seen as well. Temperatures for all data were converted to the ITS-90 (Ref. 138) temperature scale. The development of the correlation requires densities; Span and Wagner[10] in 2003 reviewed the thermodynamic properties of *n*-heptane and developed an accurate, wide-ranging equation of state from the triple point up to 600 K and 100 MPa, with an uncertainty of 0.2% in density. Recently, Sagdeev *et al.*[15] measured the density of *n*-heptane at 298 to 470 K at pressures up to 250 MPa, with an uncertainty of better than 0.3% at high pressures. Sagdeev *et al.*[15] reported that their measurements agreed with the Span and Wagner[10] densities up to 250 MPa, within 0.07%–0.25%. Hence even though the original upper pressure limit was given as 100 MPa, we use the Span and Wagner[10] equation up to 600 K and 248 MPa, which is also the highest pressure of the data in Table 1. Finally, we adopt the values for the critical point and triple point from their equation of state; the critical temperature, T_c, and the critical density, ρ_c, are 540.13 K and 232.00 kg m^{-3}, respectively.[10] The triple-point temperature is 182.55 K.[10]

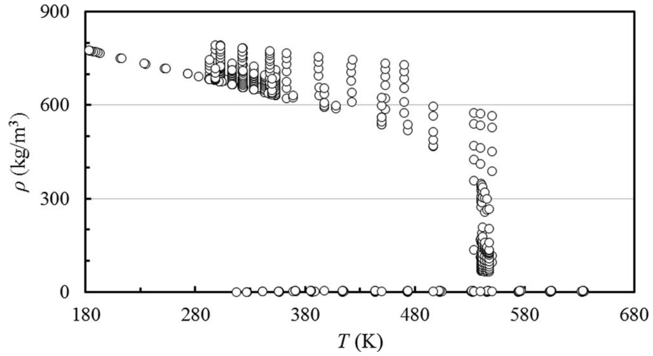

FIG. 2. Temperature-density ranges of the primary experimental viscosity data for *n*-heptane.

3.1. The dilute-gas limit and the initial-density dependence terms

The dilute-gas limit viscosity, $\eta_0(T)$ in μPa s, can be analyzed independently of all other contributions in Eq. (1). According to kinetic theory, the viscosity of a pure polyatomic gas may be related to an effective collision cross section, which contains all the dynamic and statistical information about the binary collision. For practical purposes, this relation is formally identical to that of monatomic gases and can be written as[139]

$$\eta_0(T) = \frac{0.021357\sqrt{MT}}{\sigma^2 S_\eta^*(T^*)}, \tag{2}$$

where $S_\eta^* = S(2000)/(\pi\sigma^2 f_\eta)$ is a reduced effective cross section, M is the molar mass in g mol^{-1}, σ is the length scaling parameter in nm, and f_η is a dimensionless higher-order correction factor according to Chapman and Cowling.[140,141] In the above expression for S_η^*, $S(2000)$ is a generalized cross section that includes all of the information about the dynamics of the binary collisions that govern transport properties, which in turn are governed by the intermolecular potential-energy surface.[139] The effective cross section is usually expressed in the functional form

$$\ln S_\eta^*(T^*) = \sum_{i=0}^{3} \alpha_i (\ln T^*)^i, \tag{3}$$

$$T^* = k_B T/\varepsilon, \tag{4}$$

where T^* is the reduced temperature, ε/k_B is an energy scaling parameter in K, and k_B is Boltzmann's constant.

The temperature dependence of the linear-in-density coefficient of the viscosity $\eta_1(T)$ in Eq. (1) is very large at subcritical temperatures and must be taken into account to obtain an accurate representation of the behavior of the viscosity in the vapor phase. It changes sign from positive to negative as the temperature decreases. Therefore, the viscosity along an isotherm should first decrease in the vapor phase and subsequently increase with increasing density.[139]

TABLE 2. Coefficients and parameters for Eqs. (2) and (3)–(7).

Molar mass
$M = 100.202$ g/mol

Scaling parameters
$\varepsilon/k_B = 426.118$ K $\sigma = 0.61362$ nm

Coefficients α_i for Eq. (3)
$\alpha_0 = 0.339\,74$ $\alpha_1 = -0.493\,96$
$\alpha_2 = 0.0$ $\alpha_3 = 0.080\,50$

Coefficients b_i for Eq. (7)[139]
$b_0 = -19.572\,881$ $b_1 = 219.739.99$ $b_2 = -1015.322\,6$
$b_3 = 2471.012\,5$ $b_4 = -3375.171\,7$ $b_5 = 2491.659\,7$
$b_6 = -787.260\,86$ $b_7 = 14.085\,455$ $b_8 = -0.346\,641\,58$

Vogel *et al.*[142] have shown that fluids exhibit the same general behavior of the initial density dependence of viscosity, which can also be expressed by means of the second viscosity virial coefficient $B_\eta(T)$ as

$$B_\eta(T) = \frac{\eta_1(T)}{\eta_0(T)}. \tag{5}$$

The second viscosity virial coefficient can be obtained according to the theory of Rainwater and Friend[12,13] as a function of a reduced second viscosity virial coefficient, $B_\eta^*(T^*)$, as

$$B_\eta^*(T^*) = \frac{B_\eta(T)}{N_A\,\sigma^3}, \tag{6}$$

where[139]

$$B_\eta^*(T^*) = \sum_{i=0}^{6} b_i (T^*)^{-0.25i} + b_7 (T^*)^{-2.5} + b_8 (T^*)^{-5.5}. \tag{7}$$

In Eq. (6), N_A is Avogadro's constant. The coefficients b_i for Eq. (7) are taken from Ref. 139 and are given in Table 2.

Equations (2)–(7) present a consistent scheme for the correlation of the dilute-gas limit viscosity, $\eta_0(T)$, and the initial density dependence term, $\eta_1(T)$. In the particular case of n-heptane, very accurate dilute-gas limit and initial-density-dependence viscosity values have been presented by Vogel and Holdt.[23] These values have been used with the regression package ODRPACK (Ref. 143) in order to fit the coefficients α_i in Eq. (3) and the scaling parameters σ and ε/k_B. This procedure was employed previously in correlating successfully the dilute-gas limit and the initial density dependence viscosity values of n-hexane.[9]

The values obtained are shown in Table 2. Figures 3 and 4 show the percentage deviations of the viscosity at low density $\eta_0 + \eta_1\rho$, (calculated with Eqs. (2)–(7) and the parameters in Table 2) with the experimental viscosity values of Vogel and Holdt,[23] as a function of temperature and density. Although Eq. (7) was originally developed for propane, as in its recent application to n-hexane,[9] the agreement is excellent. Figure 5 shows a comparison of the initial density dependence, η_1, viscosity data of Vogel and Holdt[23] with the values calculated

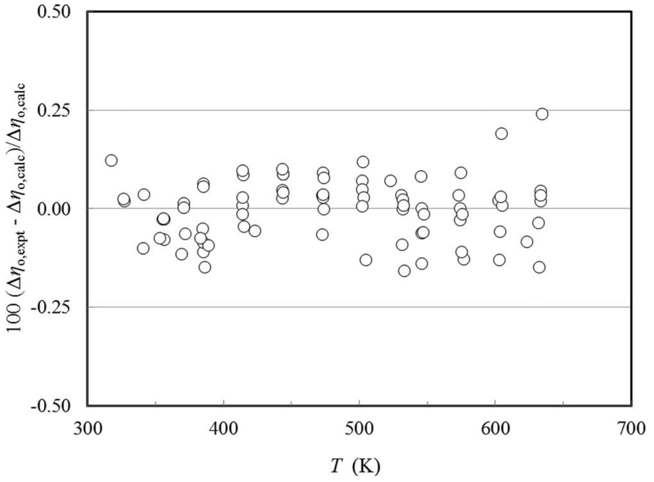

FIG. 3. Percentage deviations of the experimental low-density viscosity, $\eta_0 + \eta_1\rho$, of n-heptane, from those calculated with Eqs. (2)–(7) with the parameters in Table 2, as a function of temperature. (o) Vogel and Holdt.[23]

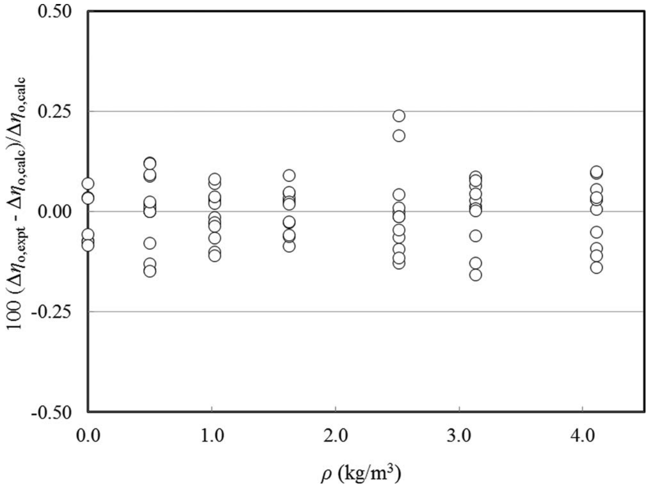

FIG. 4. Percentage deviations of the experimental low-density viscosity, $\eta_0 + \eta_1\rho$, of n-heptane, from those calculated with Eqs. (2)–(7) with the parameters in Table 2, as a function of density. (o) Vogel and Holdt.[23]

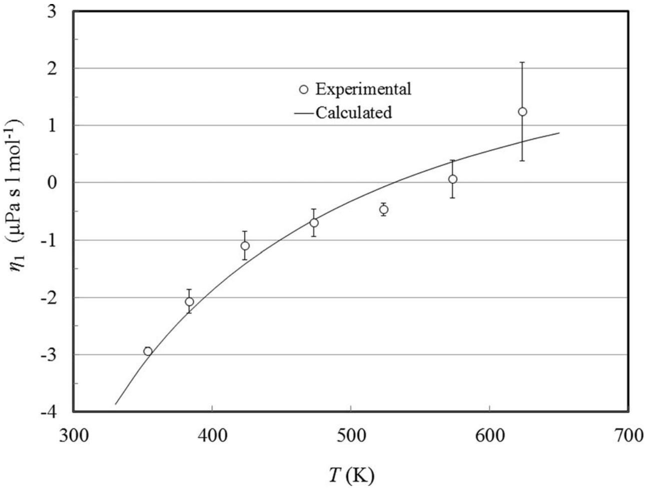

FIG. 5. Initial density dependence viscosity, η_1, as a function of the temperature. (o) Vogel and Holdt,[23] (__) values calculated with Eqs. (2)–(7) with the parameters in Table 2.

with Eqs. (2)–(7). Again, the agreement is good. Based on comparisons with the data of Vogel and Holdt,[23] we estimate the uncertainty of the correlation for the low-density gas viscosity at temperatures from 317 to 632 K to be 0.3%, at a 95% confidence level. Although 632 K is slightly beyond the recommended range of the equation of state (stated as 600 K), we feel it can safely be extrapolated to 632 K. Therefore, Eqs. (2)–(7) can be employed for the calculation of the dilute-gas limit viscosity, $\eta_0(T)$, and the initial density dependence term, $\eta_1(T)$.

3.2. The critical enhancement term

The viscosity and thermal conductivity of pure fluids diverge at the critical point due to long-range fluctuations. The critical enhancements can be described by a theoretical crossover model originally developed by Bhattacharjee *et al.*[144] and Olchowy and Sengers[145] and modified by Luettmer-Strathmann *et al.*[146] Unlike the critical enhancement in thermal conductivity, the enhancement in viscosity is confined to a small region, becoming relevant only at temperatures and densities very close to the critical point. For some fluids,[147,148] the ratio $\Delta\eta_c(\rho,T)/\eta(\rho,T)$ exceeds 0.01 only within 1% of the critical temperature of the fluid. There are very few reliable data for the viscosity of *n*-heptane in the critical region. The study by Abdulagatov and Rasulov[19,20] has data along isotherms at 540.16, 541, and 542 K where the viscosity critical enhancement would be significant near the critical density. Unfortunately, there are no viscosity data available at densities from 190 to 273 kg m^{-3} along the two isotherms closest to the critical point at 540.13 K and 232 kg m^{-3}. However, as indicated in Fig. 6, there appears to be a critical enhancement visible in some of the near-critical data.

The critical enhancement models require the derivative $(\partial\rho/\partial p)_T$. Unfortunately, testing of the equation of state[10] in the critical region found that it gives negative, nonphysical, values of the derivative $(\partial\rho/\partial p)_T$ at temperatures within 1.2 K of the critical temperature and at densities near the critical density. Since this is the region where the critical enhancement of viscosity is significant and nonphysical behavior of this derivative is likely to persist beyond this region, our recommendation is to simply set the critical enhancement $\Delta\eta_c = 0$. Hence, the critical enhancement is considered negligible and it is not further taken into consideration in this work.

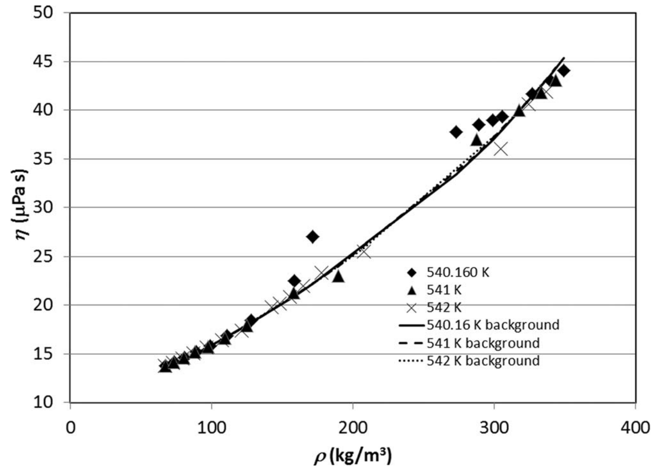

FIG. 6. Selected isotherms showing viscosity in the near-critical region.

3.3. The residual term

As stated in Sec. 2, the residual viscosity term $\Delta\eta(\rho,T)$ represents the contribution of all other effects to the viscosity of the fluid at elevated densities including many-body collisions, molecular-velocity correlations, and collisional transfer. Because there is little theoretical guidance concerning this term, its evaluation here is based entirely on experimentally obtained data.

The procedure adopted during this analysis used symbolic regression software[149] to fit all the primary data to the residual viscosity. Symbolic regression is a type of genetic programming that allows the exploration of arbitrary functional forms to regress data. The functional form is obtained by use of a set of operators, parameters, and variables as building blocks. Most recently, this method has been used to obtain correlations for the viscosity of *n*-hexane[9] and hydrogen.[150] In the present work, we restricted the operators to the set (+,−,*,/) and the operands (constant, T_r, ρ_r), with $T_r = T/T_c$ and $\rho_r = \rho/\rho_c$. Various choices of a scaling factor for density were tested, but the best results were obtained using the critical density. In addition, we found the best results when we adopted a form suggested from the hard-sphere model employed by Assael *et al.*,[151] $\Delta\eta(\rho_r,T_r) = (\rho_r^{2/3}T_r^{1/2})F(\rho_r,T_r)$, where the symbolic regression method was used to determine the functional form for $F(\rho_r,T_r)$. For this task, the dilute-gas limit and the initial density dependence terms were calculated for each experimental point (employing Eqs. (2)–(7)) and subtracted from the experimental viscosity to obtain the residual term. The density values employed were obtained by the equation of state of Span and Wagner.[10] The final equation obtained was

$$\Delta\eta(\rho,T) = \left(\rho_r^{2/3}T_r^{1/2}\right)\left\{c_1\rho_r + c_2\rho_r^2 + c_3\rho_r^3 + \frac{c_4\ \rho_r}{c_5 + c_6T_r + c_7\rho_r + \rho_r^2 + c_8\rho_rT_r}\right\}. \tag{8}$$

Coefficients c_i are given in Table 3.

Table 4 summarizes comparisons of the primary data with the correlation. We have defined the percent deviation as PCTDEV $= 100*(\eta_{\rm exp}-\eta_{\rm fit})/\eta_{\rm fit}$, where $\eta_{\rm exp}$ is the experimental value of the viscosity and $\eta_{\rm fit}$ is the value calculated from the correlation. Thus, the average absolute percent deviation (AAD) is found with the expression AAD $= (\sum|\text{PCTDEV}|)/n$, where the summation is over all n points, and the bias percent is found with the expression BIAS

TABLE 3. Coefficients for Eq. (8).

$c_1 = 22.150\,00$	$c_2 = -15.008\,70$	$c_3 = 3.717\,91$
$c_4 = 77.728\,18$	$c_5 = 9.734\,49$	$c_6 = 9.519\,00$
$c_7 = -6.340\,76$	$c_8 = -2.519\,09$	

TABLE 4. Evaluation of the *n*-heptane viscosity correlation for the primary data.

1st Author	Year publ.	AAD (%)	BIAS (%)
Sagdeev[15]	2013	1.95	1.18
Abdulagatov[16]	2006	1.07	−0.54
Zéberg-Mikkelsen[17]	2006	1.02	−0.03
Pensado[18]	2005	0.92	−0.83
Abdulagatov[19,20 a]	1992	1.75	−0.82
Assael[21]	1992	0.54	−0.11
Assael[22]	1991	0.52	−0.46
Vogel[23]	1991	0.14	−0.12
Knapstad[24]	1989	0.30	−0.03
Kessel'man[25]	1976	0.63	0.55
Giller[26]	1949	0.29	−0.19
Entire data set		**1.1**	**−0.3**

[a] Excludes 11 data points in the near-critical region.

$= (\sum \mathrm{PCTDEV})/n$. The average absolute percent deviation of the fit is 1.1%, and its bias is −0.3%. We estimate the uncertainty at a 95% confidence level to be 3.5%, except for the very-near-critical region where the deviations are larger. As mentioned in Sec. 3.1, we estimate the expanded uncertainty of the correlation at a 95% confidence level for the low-density gas at temperatures from 317 to 632 K and pressures to 0.3 MPa to be 0.3%.

Figure 7 shows the percentage deviations of all primary viscosity data from the values calculated by Eqs. (1)–(8) as a function of temperature, while Figs. 8 and 9 show the same deviations but as a function of the pressure and of the density.

In Fig. 7, a previous reference correlation by Dymond and Øye[68] along the saturated liquid line is also shown. It has an estimated uncertainty of 0.4% and is based on two data sets, those of Knapstad *et al.*[24] and Assael *et al.*[21] The Dymond and Øye correlation is restricted to a limited temperature range, from 292 to 346 K. The present correlation was based on additional data sets as given in Table 1, and may be used over a much wider temperature range. Along the liquid-phase saturation curve, we estimate the uncertainty is 1% below 292 K, 0.6% in the region from 292 to 346 K, rising to 2% between 346 and 363 K. In Table 5, values are given along the saturated liquid line, calculated from the present proposed correlation between 250 and 360 K.

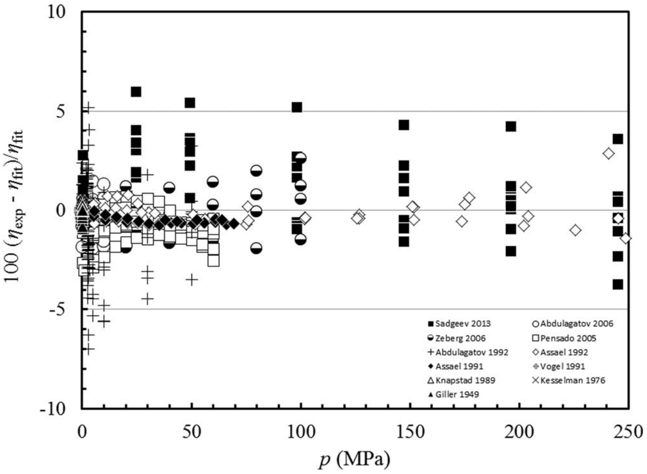

FIG. 8. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by the present model as a function of pressure.

Table 6 shows the average absolute percent deviation and the bias for the secondary data. Finally, Fig. 10 shows a plot of the viscosity of *n*-heptane as a function of temperature for different pressures. The plot demonstrates the smooth extrapolation behavior at temperatures that exceed the 600 K limit of the correlation.

4. Computer-Program Verification

Table 7 is provided to assist the user in computer-program verification. The viscosity calculations are based on the tabulated temperatures and densities.

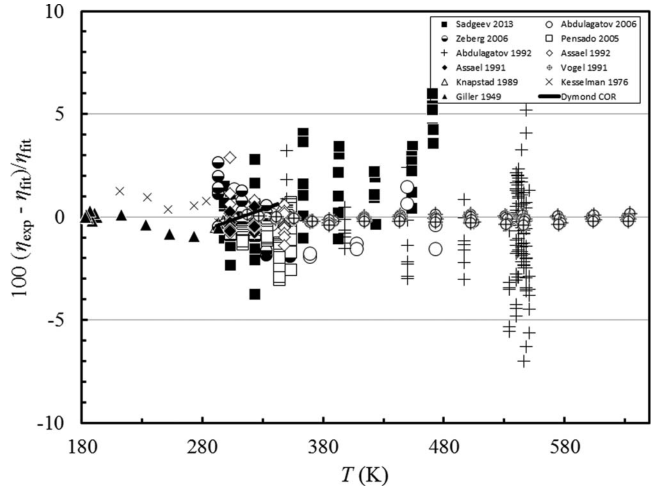

FIG. 7. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by the present model as a function of temperature.

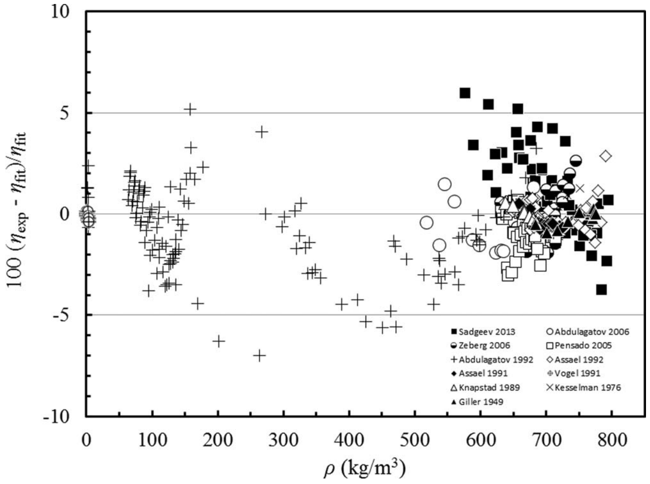

FIG. 9. Percentage deviations of primary experimental data of *n*-heptane from the values calculated by the present model as a function of density.

TABLE 5. Viscosity values of the liquid along the saturation line, calculated by the present scheme.

T (K)	ρ (kg m^{-3})	η (μPa s)
250.00	719.62	721.7
260.00	711.35	622.9
270.00	703.06	543.9
280.00	694.74	479.7
290.00	686.37	426.6
300.00	677.94	382.0
310.00	669.42	344.1
320.00	660.82	311.6
330.00	652.09	283.3
340.00	643.24	258.6
350.00	634.23	236.9
360.00	625.05	217.5

TABLE 6. Evaluation of the *n*-heptane viscosity correlation for the secondary data.

1st author	Year publ.	AAD (%)	BIAS (%)
Guerrero[27]	2011	0.28	0.04
Rathnam[28]	2010	1.43	−0.49
Yang[29]	2010	3.26	3.26
Zivkovic[30]	2010	2.63	2.63
El-Sayed[31]	2009	0.61	−0.61
Fang[32]	2008	1.33	1.33
Totchasov[33]	2008	0.38	0.38
Landaverde-Cortes[34]	2007	0.63	0.63
Reddy[35]	2007	4.65	4.65
Wang[36]	2007	0.80	0.80
Baragi[37]	2006	1.45	1.45
Iloukhani[38]	2006	1.25	1.25
Reddy[39]	2006	4.65	4.65
Al-Jimaz[40]	2005	0.15	0.15
Ma[41]	2005	1.54	−1.54
Yang[42]	2004	1.54	1.54
Yang[43]	2004	1.54	−1.54
Ma[44]	2004	2.04	2.04
Oswal[45]	2003	1.49	1.49
Queimada[46]	2003	2.55	2.13
Peng[47]	2002	1.25	1.25
Matos[48]	2001	0.53	−0.33
Nayak[49]	2001	3.33	3.33
Sastry[50]	1999	0.95	0.95
Dandekar[51]	1998	1.17	1.17
Jimenez[52]	1998	1.76	−1.76
Wu[53]	1998	1.37	−1.37
Aminabhavi[54]	1997	0.53	−0.53
Baylaucq[55]	1997	3.05	1.92
Baylaucq[56]	1997	0.63	−0.63
Nath[57]	1997	0.03	−0.03
Orge[58]	1997	2.53	2.53
Will[59]	1997	0.51	−0.51
Abdulagatov[60]	1996	3.31	1.01
Aminabhavi[61]	1996	0.53	−0.53
Sastry[62,63]	1996	0.14	0.14
Sastry[64]	1996	1.11	1.11
Aminabhavi[65]	1995	1.42	1.42
Aucejo[66]	1995	0.39	−0.39
Aminabhavi[67]	1994	0.34	−0.34
Dymond[68]	1994	0.29	−0.29
Pandey[69]	1994	1.05	−1.05
Papaioannou[70]	1994	0.82	−0.75
Blanco[71]	1993	0.71	0.71
Cooper[72]	1991	0.36	−0.36
Kanti[73]	1991	1.88	1.81
Papaioannou[74]	1991	0.80	−0.80
Zhang[75]	1991	0.33	0.17
Chevalier[76]	1990	4.76	4.76
Fermeglia[77]	1990	2.02	2.02
Iwahashi[78]	1990	2.38	2.38
Brunson[79]	1989	5.57	−4.67
Fermeglia[77]	1990	2.02	2.02
Kouris[80]	1989	0.80	−0.80
Schrodt[81]	1989	0.39	−0.39
Chandrashekhar[82]	1988	1.39	−1.39
Rao[83]	1988	1.50	1.50
Wakefield[84]	1988	1.23	−1.23
Celda[85]	1987	0.77	−0.77
Senanayake[86]	1987	0.28	−0.28
Awwad[87]	1986	3.09	3.09
Awwad[88]	1985	1.05	1.05
Martin[89]	1983	0.76	−0.76
Kashiwagi[90]	1982	0.97	−0.97
Sreenivasulu[91]	1982	0.49	−0.49
Rabe[92]	1981	2.37	2.01
Dusart[93]	1978	0.87	0.87
Gouel[94]	1978	5.86	−5.47
Gunchuk[95]	1977	1.24	1.24
Öz[96]	1976	1.28	1.28
Badalov[97]	1975	1.73	−0.97
Mussche[98]	1975	1.64	1.64
Diaz Pena[99]	1974	6.83	6.83
Moore[100]	1974	0.51	−0.51
Ratkovics[101]	1974	0.20	0.20
Shikhaliev[102]	1974	7.28	7.03
Nauruzov[103]	1973	0.70	0.46
Zhigunov[104]	1972	5.05	5.05
Konobeev[105]	1970	1.62	1.62
Kuss[106]	1969	2.94	−2.94
Darmois[107]	1967	2.96	−2.96
Carmichael[108]	1966	2.45	2.45
Bidlack[109]	1964	0.22	−0.22
Bagdasarian[110]	1964	1.55	1.53
Agaev[111]	1963	7.37	7.37
Golubev[112]	1963	9.10	−7.73
McAllister[113]	1960	0.04	−0.04
Albright[114]	1956	10.97	10.97
Ledneva[115]	1956	1.32	−0.93
Belinskaya[116]	1955	1.79	1.69
Golik[117]	1955	0.83	−0.47
Hammond[118]	1955	0.55	−0.55
Koshkin[119]	1955	1.17	0.83
Lambert[120]	1955	7.71	7.71
Toporov[120]	1955	0.57	−0.46
Johnson[122]	1953	1.81	1.81
Koshkin[123]	1953	2.03	1.62
Doolittle[124]	1951	2.59	2.59
Fawcett[125]	1946	0.29	−0.29
Geist[126]	1946	0.29	−0.29
Khalilov[127]	1939	3.33	0.14
Kyropoulos[128]	1939	2.55	2.02
Timmermans[129]	1935	1.61	−1.61
Melaven[130]	1932	2.29	0.30
Shepard[131]	1931	1.09	−1.09
Tausz[132]	1930	31.10	31.10
Smyth[133]	1928	0.77	−0.77
Lewis[134]	1925	1.20	−1.20
Miller[135]	1924	16.56	16.56
Batschinski[136]	1913	0.43	0.04
Thorpe[137]	1894	0.43	0.10

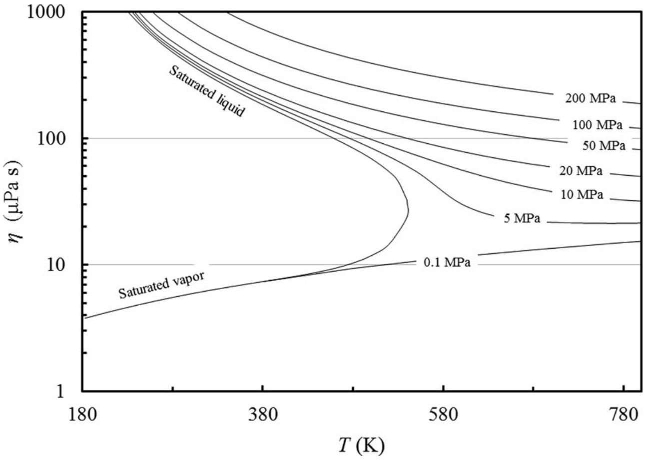

FIG. 10. Viscosity of *n*-heptane as a function of temperature for different pressures.

TABLE 7. Sample points for computer verification of the correlating equations.

T (K)	ρ (kg m^{-3})	η (μPa s)
250.00	0.00	4.9717
400.00	0.00	7.8361
550.00	0.00	10.7394
250.00	720.00	725.69
400.00	600.00	175.94
550.00	500.00	95.105

5. Conclusion

A new wide-ranging correlation for the viscosity of *n*-heptane was developed based on critically evaluated experimental data. The correlation is valid from the triple point to 600 K, and at pressures up to 248 MPa. The correlation is expressed in terms of temperature and density, with the density calculated from the equation of state of Span and Wagner.[10] We estimate the uncertainty at a 95% confidence level to be 3.5% over the whole range (with the exception of the near-critical region). Along the saturated liquid curve, we estimate the uncertainty is 1% below 292 K, 0.6% in the region from 292 to 346 K, rising to 2% between 346 and 363 K, and 0.3% for the low-density gas at temperatures from 317 to 600 K and pressures to 0.3 MPa.

Acknowledgments

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6. Appendix: Corrected Values for the Viscosity of *n*-Heptane from Abdulagatov and Rasulov[19,20]

p (MPa)	T (K)	η (μPa s)	p (MPa)	T (K)	η (μPa s)	p (MPa)	T (K)	η (μPa s)
0.100	298.300	387.50	0.100	550.400	10.90	2.700	540.160	22.52
2.500	298.300	398.70	2.500	550.400	15.20	2.700	542.000	19.78
2.715	298.300	398.90	2.715	550.400	17.04	2.700	544.000	18.31
5.000	298.300	408.70	5.000	550.400	52.00	2.700	546.000	17.72
10.000	298.300	429.90	10.000	550.400	69.10	2.700	548.000	17.13
30.000	298.300	516.50	30.000	550.400	107.40	2.720	540.160	22.00
50.000	298.300	607.10	50.000	550.400	136.50	2.720	541.000	21.33
0.100	350.100	238.10	2.000	540.160	13.81	2.720	542.000	20.21
2.500	350.100	245.60	2.000	541.000	13.79	2.720	544.000	18.73
2.715	350.100	246.00	2.000	542.000	13.76	2.720	546.000	18.03
5.000	350.100	253.80	2.000	544.000	13.75	2.720	548.000	17.37
10.000	350.100	270.10	2.000	546.000	13.65	2.740	540.160	37.75
30.000	350.100	333.50	2.000	548.000	13.58	2.740	542.000	20.84
50.000	350.100	399.30	2.100	540.160	14.18	2.740	544.000	19.15
0.100	397.800	7.70	2.100	541.000	14.15	2.740	546.000	18.32
2.500	397.800	165.30	2.100	542.000	14.11	2.740	548.000	17.63
2.715	397.800	165.40	2.100	544.000	14.04	2.760	540.160	34.54
5.000	397.800	171.20	2.100	546.000	13.96	2.760	541.000	23.01
10.000	397.800	185.60	2.100	548.000	13.91	2.760	542.000	21.95
30.000	397.800	235.10	2.200	540.160	14.65	2.760	544.000	19.85
50.000	397.800	282.20	2.200	541.000	14.60	2.760	546.000	18.86
0.100	449.600	9.00	2.200	542.000	14.54	2.760	548.000	18.07
2.500	449.600	110.70	2.200	544.000	14.43	2.780	540.160	39.01
2.715	449.600	111.20	2.200	546.000	14.31	2.780	542.000	23.35
5.000	449.600	116.70	2.200	548.000	14.24	2.780	544.000	20.84
10.000	449.600	129.20	2.300	540.160	15.25	2.780	546.000	19.41
30.000	449.600	173.00	2.300	541.000	15.18	2.780	548.000	18.44
50.000	449.600	212.30	2.300	542.000	15.10	2.800	540.160	39.32
0.100	496.700	9.80	2.300	544.000	14.92	2.800	541.000	37.06
2.500	496.700	75.60	2.300	546.000	14.75	2.800	542.000	25.51
2.715	496.700	76.30	2.300	548.000	14.66	2.800	544.000	21.82
5.000	496.700	84.00	2.400	540.160	15.84	2.800	546.000	19.72

Continued

p (MPa)	T (K)	η (μPa s)	p (MPa)	T (K)	η (μPa s)	p (MPa)	T (K)	η (μPa s)
10.000	496.700	96.90	2.400	541.000	15.74	2.800	548.000	18.75
30.000	496.700	138.30	2.400	542.000	15.63	2.900	540.160	41.65
50.000	496.700	172.50	2.400	544.000	15.45	2.900	541.000	40.02
0.100	534.000	10.40	2.400	546.000	15.27	2.900	542.000	36.05
2.500	534.000	18.00	2.400	548.000	15.08	2.900	544.000	34.67
2.715	534.000	45.10	2.500	540.160	16.84	2.900	548.000	22.30
5.000	534.000	60.20	2.500	541.000	16.62	3.000	540.160	43.05
10.000	534.000	75.90	2.500	542.000	16.41	3.000	541.000	41.80
30.000	534.000	115.60	2.500	544.000	16.08	3.000	542.000	40.61
50.000	534.000	148.70	2.500	546.000	15.82	3.000	544.000	37.50
0.100	540.100	10.70	2.500	548.000	15.63	3.000	546.000	30.24
2.500	540.100	16.50	2.600	540.160	18.45	3.000	548.000	23.94
2.715	540.100	20.95	2.600	541.000	17.93	3.100	540.160	44.13
5.000	540.100	57.60	2.600	542.000	17.42	3.100	541.000	43.12
10.000	540.100	73.90	2.600	544.000	17.05	3.100	542.000	41.93
30.000	540.100	113.30	2.600	546.000	16.63	3.100	544.000	39.81
50.000	540.100	144.90	2.600	548.000	16.26	3.100	546.000	37.03
						3.100	548.000	34.43

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