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Reference Correlation of the Viscosity of Benzene from the Triple Point to 675 K and up to 300 MPa

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This paper contains new, representative reference equations for the viscosity of benzene. The equations are based in part upon a body of experimental data that has been critically assessed for internal consistency and for agreement with theory whenever possible. The correlation is valid from the triple point (278.647 K) to 675 K, and at pressures up to 300 MPa, with the exception of temperatures lower than 350 K where the pressure is restricted to 200 MPa. For the liquid phase, at temperatures from 288 to 373 K at pressures up to 80 MPa, we estimate the uncertainty (at a 95% confidence level) to be 1.8%, increasing to 3.4% at 200 MPa, and 5% at pressures up to the correlation maximum. For the liquid at temperatures from 373 to 523 K, the uncertainty is 2.7% at pressures from saturation to 50 MPa, rising to 3.6% at 300 MPa. For temperatures above 523 K, we estimate the uncertainty in the liquid phase to be 5%. The uncertainty for the low-density fluid phase at temperatures from 305 to 640 K and pressures to 0.3 MPa is estimated to be 0.2%. *© 2014 by the U.S. Secretary of Commerce on behalf of the United States. All rights reserved.* [http://dx.doi.org/10.1063/1.4892935]

Key words: benzene; critical phenomena; transport properties; viscosity.

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4. Percentage deviations of the experimental low-density viscosity, $\eta_0 + \eta_1\rho$, of benzene, from those calculated with Eqs. (2)–(7) with the parameters in Table 2, as a function of density. 7
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1. Introduction

In a series of recent papers, new reference correlations for the thermal conductivity of a series of fluids[1–9] and the viscosity of water,[10] *n*-hexane,[11] and *n*-heptane[12] covering a wide range of conditions of temperature and pressure, were reported. In this paper, the methodology adopted for the viscosity of *n*-hexane[11] and *n*-heptane[12] is extended to developing a new reference correlation for the viscosity of benzene.

The goal of this work is to critically assess the available literature data, and provide a wide-ranging correlation for the viscosity of benzene that is valid over gas, liquid, and supercritical states, incorporating densities provided by the recent equation of state of Thol *et al.*,[13] while maintaining consistency with our recent reference correlation for the thermal conductivity of benzene.[4]

2. Methodology

The viscosity η can be expressed[11,12,14] as the sum of four independent contributions, as

$$\eta(\rho,T) = \eta_0(T) + \eta_1(T)\rho + \Delta\eta(\rho,T) + \Delta\eta_c(\rho,T), \quad (1)$$

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[14–16] 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 residual 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.

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. Finally, it should be stated that, relative to other types of viscosity models commonly used in engineering calculations, this viscosity model has a strong phenomenological basis and is best suited for providing a high precision correlation of pure-component viscosity over a very broad temperature and pressure space.

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.[17] 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 wide 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

Table 1 summarizes, to the best of our knowledge, the experimental measurements[18–167] of the viscosity of benzene including sample purity and the uncertainties ascribed by the authors. 150 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.

The measurements of Vogel *et al.*[22] were performed in the very-low-density vapor range in an absolute oscillating-disk viscometer designed exactly for that region. These measurements were obtained with an uncertainty of 0.15% rising to 0.3% at the highest temperatures, and were part of the primary dataset. Knapstad *et al.*[21] performed very accurate viscosity measurements at atmospheric pressure in an absolute oscillating-cup viscometer with an uncertainty of 0.33%,

TABLE 1. Viscosity measurements of benzene

1st author	Year Publ.	Technique employed[a]	Purity (%)	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
			Primary data				
dos Santos[18]	1997	TCr	99.99	0.5	30	298–348	0.1–178
Matsuo[19]	1993	Cap	99.5	2	20	298–313	0.1–49
Assael[20]	1991	VBW	99.80	0.5	28	303–323	0.1–70
Knapstad[21]	1989	OCup	99.5	0.33	6	288–332	0.1
Vogel[22]	1986	OD	99.92	0.15	75	305–637	0–0.3
Bauer[23]	1984	Cap	99.7	0.1	2	293, 298	0.1
Abdullaev[24b]	1983	Cap	99.98	1.2	303	353–673	0.3–40
Kashiwagi[25]	1982	TCr	99	2	28	298–348	0.1–110
Dymond[26]	1981	FB	99.5	2	27	298–373	0.1–402
Parkhurst[27]	1975	RB	na	2.5	35	303–433	0.1–400
Mamedov[28]	1971	FB	na	1	130	293–548	0.1–41
			Secondary data				
Bhalodia[29]	2013	Cap	na	0.1	3	303–313	0.1
Bhatia[30]	2013	Cap	99.7	0.53	3	298–308	0.1
de Doz[31]	2011	Cap	99.8	0.35	1	298	0.1
Mariano[32]	2011	Cap	99.7	0.8	3	283–303	0.1
Pradhan[33]	2011	Cap	na	0.03	1	298	0.1
Rahman[34]	2011	Cap	99	1	3	303–323	0.1
Shukla[35]	2011	Cap	99	1.7	4	298–313	0.1
Dubey[36]	2010	Cap	na	0.5	1	298	0.1
Rathnam[37]	2010	Cap	99.5	0.7	3	303–313	0.1
Yadava[38]	2010	Cap	99.7	na	1	308	0.1
Ali[39]	2009	Cap	99	0.3	1	298	0.1
El-Sayed[40]	2009	Cap	99.95	0.34	2	308–313	0.1
Sastry[41]	2009	Cap	99.98	0.53	2	298–308	0.1
Awwad[42]	2008	Cap	99.8	0.76	4	298–323	0.1
Song[43]	2008	Cap	99.5	0.64	7	303–333	0.1
Wankhede[44]	2008	Cap	na	na	3	288–308	0.1
Hernandez[45]	2007	RB	99.9	2	40	313–393	0.69–60
Kharat[46]	2007	Cap	99	0.55	4	298–313	0.1
Nain[47]	2007	Cap	99.8	na	3	298–318	0.1
Reddy[48]	2007	Cap	99.8	0.18	1	303	0.1
Ren[49]	2007	Cap	na	0.64	6	298–343	0.1
Song[50]	2007	Cap	99.8	0.6	5	298–333	0.1
Yang[51]	2007	Cap	99.5	0.64	4	303–333	0.1
Ali[52]	2006	Cap	99	0.9	1	303	0.1
Al-Kandary[53]	2006	Cap	99.5	0.47	4	288–303	0.1
Baragi[54]	2006	Cap	99.80	na	3	298–308	0.1
Han[55]	2006	Cap	na	na	1	298	0.1
Varshney[56]	2006	Cap	99.3	0.02	1	303	0.1
Agarwal[57]	2004	Cap	99	0.49	1	298	0.1
Jain[58]	2004	Cap	99	0.56	1	313	0.1
Ma[59]	2004	VBW	99.5	1.60	22	303–445	0.1–6.1
Oswal[60]	2004	Cap	99.5	0.38	2	303–313	0.1
George[61]	2003	Cap	99.5	0.5	2	298–308	0.1
Lark[62]	2003	Cap	na	0.35	2	298–303	0.107
Nikam[63]	2003	Cap	99	0.55	4	298–313	0.1
Nayak[64]	2002	Cap	99.9	0.88	3	298–308	0.1
Chowdhury[65]	2001	Cap	99	0.05	3	303–323	0.1
Pandey[66]	2001	Cap	na	0.02	1	298	0.1
Swain[67]	2001	Cap	98	na	5	298–318	0.1
Tsierkezos[68]	2000	Cap	99.5	0.15	1	293	0.1
Viswanathan[69]	2000	na	99.5	na	3	288–298	0.1
Aminabhavi[70]	1999	Cap	99.8	0.19	3	298–308	0.1
Aralaguppi[71]	1999	Cap	99.8	0.18	3	298–308	0.1
Guzman[72]	1999	Cap	99.9	na	3	283–313	0.1
Sastry[73]	1999	Cap	99.8	0.2	2	308–318	0.1
Casas[74]	1998	Cap	99.5	0.08	1	298	0.1
Emmerling[75]	1998	Cap	99	na	4	293–323	0.1
Exarchos[76]	1995	Cap	99.5	0.3	4	293–308	0.1
Krishnan[77]	1995	Cap	99.95	0.1	1	303	0.105
Petrino[78]	1995	Cap	na	0.1	1	298	0.1
Rived[79]	1995	Cap	99.7	0.15	1	303	0.1
Singh[80]	1995	Cap	na	0.5	1	298	0.1

TABLE 1. Viscosity measurements of benzene—Continued

1st author	Year Publ.	Technique employed[a]	Purity (%)	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Lafuente[81]	1994	Cap	99.9	na	2	298, 313	0.106
Rout[82]	1994	Cap	na	0.53	5	298–318	0.1
Shastri[83]	1993	Cap	99.8	0.2	1	293	0.1
Klofutar[84]	1992	Cap	na	0.58	6	293–333	0.102
Tasioula-Margari[85]	1992	Cap	99.7	na	5	298–308	0.1
Papaioannou[86]	1991	Cap	99.7	0.25	1	298	0.1
Zhang[87]	1991	FB	na	1.5	21	298–348	0.1–30
Chevalier[88]	1990	Cap	99.95	na	1	298	0.1
Joshi[89]	1990	Cap	na	0.13	5	298–313	0.1
Schumpe[90]	1990	Cap	na	na	1	293	0.1
Sekar[91]	1990	Cap	na	0.5	1	303	0.1
Sivaramprasad[92]	1990	Cap	na	0.5	1	298	0.1
Kouris[93]	1989	Cap	99.7	0.25	1	298	0.1
Ruiz[94]	1989	Cap	99.7	0.2	3	303–323	0.1
Schrodt[95]	1989	Cap	na	na	1	297	0.1
Shah[96]	1989	Cap	na	0.55	3	298–319	0.1
Chandrasekhar[97]	1988	Cap	na	na	1	303	0.1
Rao[98]	1988	Cap	na	na	1	298	0.1
Manjeshwar[99]	1987	Cap	na	1.5	2	298–318	0.1
Ramanjaneyulu[100]	1987	Cap	na	na	1	303	0.1
Fischer[101]	1986	Cap	99.7	na	7	286–343	0.1
Al-Mandfai[102]	1985	Cap	99.5	na	1	298	0.1
Albright[103]	1983	Cap	99.94	na	1	298	0.1
Martin[104]	1983	Cap	na	3	5	303–343	0.1
Nath[105]	1983	Cap	na	0.18	1	303	0.1
Asfour[106]	1981	Cap	99	0.13	1	298	0.1
Teja[107]	1981	Cap	na	0.1	3	298–323	0.1
Yadava[108]	1981	Cap	na	0.19	1	307	0.1
Bich[109]	1979	OD	na	na	47	307–628	0.01–0.1
Gouel[110]	1978	Cap	na	5	51	292–392	0.1–40
Medani[111]	1977	RB	na	na	12	353–463	0.1–1.2
Vyunnik[112]	1977	Cap	na	na	5	278–328	0.1
Dhillon[113]	1976	Cap	na	1	2	298–308	0.1
Bulanov[114]	1974	Cap	na	2	12	293–402	0.1–0.37
Mekhtiev[115]	1974	Cap	99.98	0.5	7	283–343	0.1
Alms[116]	1973	Cap	na	na	1	298	0.1
Collings[117]	1971	TCr	na	1	19	303–323	0.1–171
Konobeev[118]	1970	Cap	na	na	5	288–333	0.1
Heric[119]	1967	Cap	na	0.1	1	298	0.1
Ridgway[120]	1967	Cap	na	na	1	298	0.1
Fort[121]	1966	Cap	na	0.2	1	298	0.1
Katti[122]	1966	Cap	na	na	1	298	0.1
Bagdasarian[123]	1964	na	na	na	9	293–453	0.1–1
Howard[124]	1959	Cap	na	0.1	6	293–343	0.1
Teller[125]	1959	Cap	na	na	2	283–299	0.1
Ling[126]	1958	Cap	na	na	7	303–423	0.01–0.58
Rabinovich[127]	1958	Ultr	na	na	12	288–343	0.1
Airapetova[128]	1956	na	na	na	3	298–338	0.1
Heiks[129]	1956	na	na	na	12	363–561	0.2–5
Toropov[130]	1956	na	na	na	3	293–333	0.1
Belinskaya[131]	1955	Cap	na	0.5	11	279–333	0.1
Koshkin[132]	1955	na	na	na	5	283–323	0.1
Kuss[133]	1955	Cap	na	na	7	293–343	0.1
Mamedov[134]	1955	Cap	na	0.04	7	283–343	0.1
Dixon[135]	1954	Cap	na	na	3	293–333	0.1
Grunberg[136]	1954	Cap	na	0.2	1	298	0.1
Keeling[137]	1954	Cap	na	na	3	303–348	0.1
Mukherjee[138]	1953	Cap	na	na	6	298–323	0.1
Subnis[139]	1952	Cap	na	na	6	293–323	0.1
Craven[140]	1951	Pend	na	1	4	308–351	0.1
Jobling[141]	1951	FB	na	na	10	303–363	0.1
Golik[142]	1950	Cap	na	na	7	293–303	0.1–0.4
Teitelbaum[143]	1950	Cap	98	na	1	298	0.1
Geist[144]	1946	Cap	na	0.1	2	293–313	0.1
Linke[145]	1941	FB	na	na	13	286–353	0.1

TABLE 1. Viscosity measurements of benzene—Continued

1st author	Year Publ.	Technique employed[a]	Purity (%)	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Udovenko[146]	1940	na	na	na	1	298	0.1
Khalilov[147]	1939	na	na	na	18	293–483	0.1–1.7
Salceanu[148]	1939	Cap	na	na	1	293	0.1
Lemonde[149]	1938	Cap	na	na	2	284, 288	0.1
Adzumi[150]	1937	Cap	na	na	1	293	0.1
Kireev[151]	1936	Cap	na	na	3	293–333	0.1
Titani[152]	1933	Cap	na	na	7	404–586	0.1
Piatti[153]	1931	na	na	na	7	273–333	0.1
Madge[154]	1930	na	na	na	7	283–343	0.1
Tausz[155]	1930	Cap	na	na	2	293	0.1
Lowry[156]	1929	na	na	na	2	293–373	0.1
Nasini[157]	1929	Cap	na	na	15	288–519	0.008–3
Bridgman[158]	1926	FB	na	na	6	303–348	0.1–98
Kolossowsky[159]	1925	Cap	na	na	8	285–313	0.1
Lewis[160]	1925	Cap	na	na	2	298	0.1
Bingham[161]	1920	na	na	na	5	288–348	0.1
Faust[162]	1912	Cap	na	na	4	273–323	0.1
Rappenecker[163]	1910	Cap	na	na	2	373, 485	0.2, 1.8
Findlay[164]	1909	Cap	na	na	1	352	0.1
Dunstan[165]	1907	Cap	na	na	1	298	0.1
Heydweiller[166]	1896	Cap	na	na	38	288–463	0.1–1.2
Thorpe[167]	1894	Cap	na	na	12	280–348	0.1

[a]Abs, Absolute; Cap, Capillary; FB, Falling Body; na, not available; OCup, Oscillating Cup; OD, Oscillating Disk; Pend, Pendulum; RB, Rolling Ball; TCr, Torsional Crystal; Ultr, Ultrasonic; VBW, Vibrating Wire.
[b]Low-density points were excluded from the primary data set.

and these measurements were also included in the primary data set. A capillary viscometer was employed by Bauer and Meerlender[23] for two very-low-uncertainty absolute measurements of the viscosity of benzene at atmospheric pressure. The uncertainty was 0.1% and these measurements were also part of the primary set. An absolute vibrating-wire instrument was employed by Assael *et al.*[20] These measurements, backed by a full theoretical analysis, extend up to 70 MPa pressure with an uncertainty of 0.5%, and thus are primary data. Also of similar low uncertainty were the torsional-crystal viscosity measurements of dos Santos and Nieto de Castro,[18] performed up to 178 MPa; these were also part of the primary data set. The remaining sets included in the primary data were of slightly inferior uncertainty. Capillary viscometers were employed by Matsuo and Makita[19] and Abdullaev *et al.*[24] with quoted uncertainties of 2% and 1.2%, respectively. In the low-density fluid region where the zero-density and first viscosity virial terms contribute more than 99% of the total viscosity, we excluded points from Abdullaev *et al.*[24] from the primary data set (about 20 points in total) and exclusively used the measurements of Vogel *et al.*[22] The measurements of Kashiwagi and Makita,[25] Dymond *et al.*,[26] and Parkhurst and Jonas[27] were also included as they extend to high pressures, and they seem to agree adequately with each other. They employed a torsional-crystal viscometer, a falling-body and a rolling-body viscometer, with corresponding uncertainties of 2%, 2%, and 2.5%. Finally, the measurements of Mamedov *et al.*,[28] performed in a falling-body viscometer with a 1% uncertainty, were also included as they extend to higher temperatures.

Figures 1 and 2 show the ranges of the primary measurements outlined in Table 1, and the phase boundary may be seen as well in Fig. 2. Examining Fig. 1 it can be seen that above 300 MPa pressure, only very few measurements exist. Hence it was preferred to restrict the pressure range of the present correlation to 300 MPa. Furthermore, below 350 K, pressure measurements exist only up to 200 MPa. As this is a range where the viscosity increases sharply with pressure, extrapolation of the correlation in this region should be avoided. Temperatures for all data were converted to the ITS-90 (Ref. 168) temperature scale. The development of the correlation requires densities; recently, Thol *et al.*[13] reviewed the thermodynamic properties of benzene and developed an accurate, wide-ranging equation of state up to 725 K and 500 MPa. The approximate uncertainties of the density (at a coverage factor of 2) calculated with this

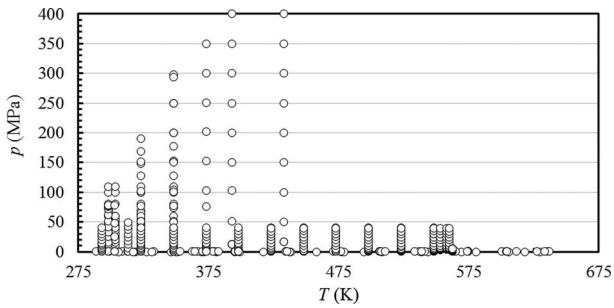


FIG. 1. Temperature-pressure ranges of the primary experimental viscosity data for benzene.

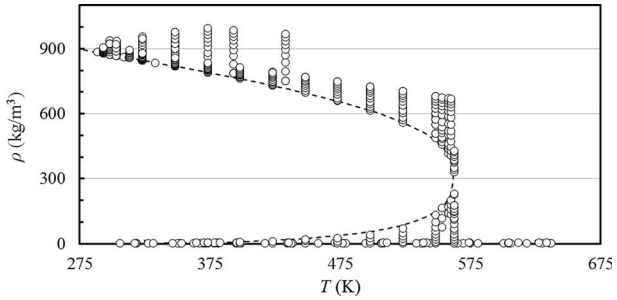


FIG. 2. Temperature-density ranges of the primary experimental viscosity data for benzene. Saturation line (- - -).

equation are 0.1% below 350 K and 0.2% above 350 K for liquid densities, 1% for saturated vapor densities, 0.1% for densities up to 350 K and 100 MPa, and 0.1%–0.5% in density above 350 K. Uncertainties in the critical region are higher for the above properties. We also adopt the values for the critical point and triple point from their equation of state;[13] the critical temperature, T_c, and the critical density, ρ_c, were taken to be equal to 562.02 K and 304.792 kg m^{-3}, respectively. The triple-point temperature is 278.674 K.[13]

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

The dilute-gas limit viscosity, $\eta_0(T)$, can be analyzed independently of all other contributions in Eq. (1). According to the 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[169]

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

where η_0 is in μPa s, $S_\eta^* = S(2000)/(\pi\sigma^2 f_\eta)$ 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.[170,171] In the definition of 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, and in turn are governed by the intermolecular potential energy surface.[169] 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.[169] Vogel *et al.*[22] 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[15,16] 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[169]

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

In Eq. (6), N_A is Avogadro's constant. The coefficients b_i from Ref. 169 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 benzene, very accurate dilute-gas limit and initial-density-dependence viscosity values have been presented by Vogel *et al.*[22] These values have been used with the regression package ODRPACK (Ref. 172), in order to fit the coefficients α_i in Eq. (3). The scaling parameters σ and ε/k_B were the same as those employed in the calculation of the dilute-gas limit contribution in the recent thermal conductivity reference correlation for benzene.[4] This procedure was also employed previously in correlating successfully the dilute-gas limit and

TABLE 2. Coefficients and parameters for Eqs. (2)–(4) and (7)

Molar mass		
78.11184 g/mol		
Scaling parameters		
$\varepsilon/k_B = 412.0$ K	$\sigma = 0.540$ nm	
Coefficients α_i for Eq. (3)		
$\alpha_0 = 0.234\,018$	$\alpha_1 = -0.476\,136$	
$\alpha_2 = 0.0$	$\alpha_3 = -0.015\,269$	
Coefficients b_i for Eq. (7) (Ref. 169)		
$b_0 = -19.572\,881$	$b_1 = 219.739\,99$	$b_2 = -1015.322\,6$
$b_3 = 2471.012\,5$	$b_4 = -3375.171\,7$	$b_5 = 2491.659\,7$
$b_6 = -787.260\,86$	$b_7 = 14.085\,455$	$b_8 = -0.346\,641\,58$

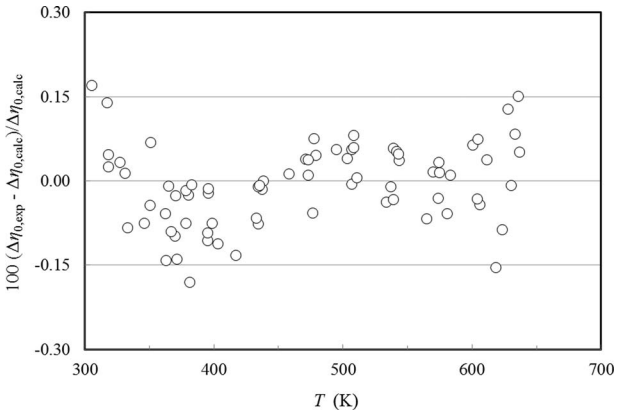


FIG. 3. Percentage deviations of the experimental low-density viscosity, $\eta_0 + \eta_1\rho$, of benzene, from those calculated with Eqs. (2)–(7) with the parameters in Table 2, as a function of temperature. (o) Vogel *et al.*[22]

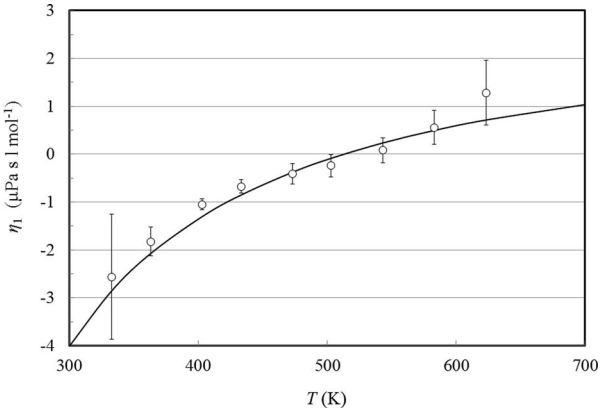


FIG. 5. Initial density dependence viscosity, η_1, as a function of the temperature. (o) Vogel *et al.*[22], (—) values calculated with Eqs. (2)–(7) with the parameters in Table 2.

the initial density dependence viscosity values of *n*-hexane[11] and *n*-heptane.[12]

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 *et al.*,[22] as a function of temperature and density. Although Eq. (7) was originally developed for propane, as in its recent application to *n*-hexane[11] and *n*-heptane,[12] the agreement is excellent. Figure 5 shows a comparison of the initial density dependence, η_1, viscosity data of Vogel *et al.*[22] with the values calculated with Eqs. (2)–(7). Again, the agreement is good. Based on comparisons with the data of Vogel *et al.*,[22] we estimate the uncertainty of the correlation for the low-density gas viscosity at temperatures from 305 to 640 K to be 0.2%, at a 95% confidence level. 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

Viscosity and thermal conductivity of pure fluids diverge at the critical point due to long-range fluctuations. The critical enhancements can be described by theoretical crossover models originally developed by Bhattacharjee *et al.*[173] and Olchowy and Sengers[174] and modified by Luettmer-Strathmann *et al.*[175] 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,[176,177] the ratio $\Delta\eta_c(\rho,T)/\eta(\rho,T)$ exceeds 1% only within ±1% of the critical temperature of the fluid. There are only very few reliable data for benzene in the critical region. The study by Abdullaev *et al.*[24] has data along a subcritical isotherm near 560.20 K and supercritical isotherms near 562.65, 563.20, 564.20, and 566.20 K where the viscosity critical enhancement is significant near the critical density. These viscosity isotherms are shown in Fig. 6, after the reported temperatures were shifted by +0.09 K. This temperature shift is required to center the viscosity maximum in the data near the

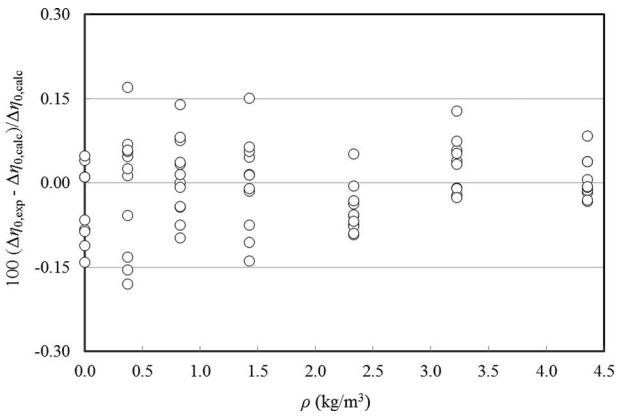


FIG. 4. Percentage deviations of the experimental low-density viscosity, $\eta_0 + \eta_1\rho$, of benzene, from those calculated with Eqs. (2)–(7) with the parameters in Table 2, as a function of density. (o) Vogel *et al.*[22]

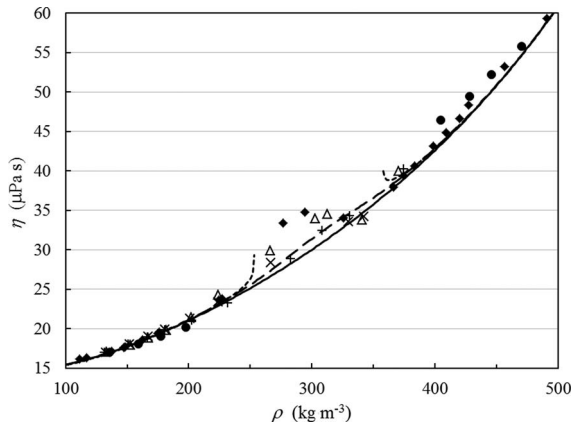


FIG. 6. Selected isotherms showing viscosity in the near-critical region. 562.65 K (♦), 562.65 K theoretical calculation (— —), 563.20 K (Δ), 564.20 K (×), 566.20 K (+), 560.20 K (●), 560.20 K theoretical calculation (- - -), background (—).

critical density of 304.792 kg m^{-3} as expected based on the theory. Even though the claimed temperature stability was ±0.01 K, this temperature shift could be due to a temperature gradient in the viscometer or uncertainty in the temperature that was measured with a thermocouple. It is the temperature difference between the experimental temperature and the actual sample critical point that is important for the critical enhancement. The apparent critical temperature of the sample studied might also have been different from the critical point of pure benzene due to small concentrations (~0.02%) of impurities.

The critical enhancement model for viscosity of Bhattacharjee *et al.*[173] requires knowledge of the critical temperature, pressure, and density, and several critical parameters that are available from our earlier correlation for the thermal conductivity of benzene,[4] along with the derivative $(\partial\rho/\partial p)_T$ from the equation of state of Thol *et al.*[13] The viscosity model[173] additionally requires the wave number q_C that is calculated from the critical properties and knowledge of $(\partial p/\partial T)$ at the critical density from Thol *et al.*[13] and the background values for the viscosity (this work) and thermal conductivity[4] at the critical point. We calculate $q_C = 1.66 \times 10^{-8}$ m^{-1} and assume that the only remaining adjustable parameter, the wave number cutoff $q_D = 1.63 \times 10^{-8}$ m^{-1}, is the same as that found for the thermal conductivity critical enhancement of benzene.[4] In general, the values for the wave number cutoff are different for the viscosity and thermal conductivity models, but this is a reasonable estimate to calculate theoretical values for the viscosity critical enhancement. Figure 6 shows curves calculated for the critical enhancement of viscosity along the nearest subcritical isotherm at 560.20 K and the nearest supercritical isotherm at 562.65 K relative to the background viscosity and the data of Abdullaev *et al.*[24]

It is apparent in Fig. 6 that the experimental data have much more critical enhancement relative to the background viscosity than that expected from the theoretically based model. Our theoretical estimates indicate a 4.6% critical enhancement for viscosity at the critical density along the 562.65 K isotherm of Abdullaev *et al.*[24] This is the maximum critical enhancement that is expected for any of the available experimental viscosity data. Figure 6 also shows that the scatter in the experimental data in the critical region exceeds the magnitude of the calculated critical enhancement, so 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.

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[178] to fit all the primary data to the residual viscosity. It should be noted that, according to the discussion in Sec. 3.2, the measurements of Abdullaev *et al.*[24] performed very near the critical temperature were not included in the correlation. To obtain the residual viscosity, symbolic regression was employed. 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,[11] *n*-heptane,[12] and hydrogen.[179] 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.*,[180] $\Delta\eta(\rho_r,T_r) = (\rho_r^{2/3}T_r^{1/2})F(\rho_r,T_r)$, 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 Thol *et al.*[13] The final equation obtained was

$$\Delta\eta(\rho,T) = \left(\rho_r^{2/3}T_r^{1/2}\right) \times \left\{c_0\rho_r^2 + \frac{c_1\rho_r}{c_2 + c_3T_r + c_4\rho_r} + \frac{c_5\rho_r + c_6\rho_r^2}{c_7 + c_8\rho_r^2}\right\}. \quad (8)$$

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*$(\eta_{exp}-\eta_{fit})/\eta_{fit}$, where η_{exp} is the experimental value of the viscosity and η_{fit} is the value calculated from the

TABLE 3. Coefficients of Eq. (8)

$c_0 = -9.989\ 45$	$c_1 = 86.062\ 60$	$c_2 = 2.748\ 72$
$c_3 = 1.111\ 30$	$c_4 = -1.0$	$c_5 = -134.133\ 0$
$c_6 = -352.473$	$c_7 = 6.609\ 89$	$c_8 = 88.417\ 4$

TABLE 4. Evaluation of the benzene viscosity correlation for the primary data

1st Author	Year Publ.	AAD (%)	BIAS (%)
dos Santos[18]	1997	1.66	0.82
Matsuo[19]	1993	0.57	−0.42
Assael[20]	1991	0.25	−0.16
Knapstad[21]	1989	0.33	−0.27
Vogel[22]	1986	0.06	−0.01
Bauer[23]	1984	0.36	0.36
Abdullaev[24]	1983	1.56	−0.14
Kashiwagi[25]	1982	0.58	0.05
Dymond[26]	1981	1.26	0.84
Parkhurst[27]	1975	1.75	−0.10
Mamedov[28]	1971	1.31	0.42
Entire data set		**1.23**	**−0.14**

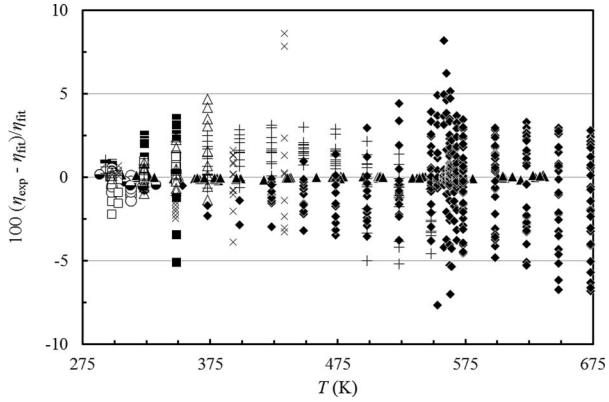


FIG. 7. Percentage deviations of primary experimental data of benzene from the values calculated by the present model as a function of temperature. dos Santos and Nieto de Castro[18] (■), Matsuo and Makita[19] (○), Assael *et al.*[20] (●), Knapstad *et al.*[21] (◒), Vogel *et al.*[22] (▲), Bauer and Meerlender[23] (◊), Abdulaev *et al.*[24] (♦), Kashiwagi and Makita[25] (□), Dymond *et al.*[26] (Δ), Parkhurst and Jonas[27] (×), Mamedov *et al.*[28] (+).

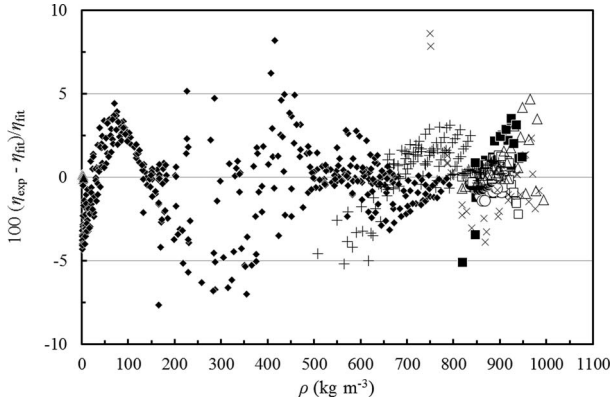


FIG. 9. Percentage deviations of primary experimental data of benzene from the values calculated by the present model as a function of density. dos Santos and Nieto de Castro[18] (■), Matsuo and Makita[19] (○), Assael *et al.*[20] (●), Knapstad *et al.*[21] (◒), Vogel *et al.*[22] (▲), Bauer and Meerlender[23] (◊), Abdulaev *et al.*[24] (♦), Kashiwagi and Makita[25] (□), Dymond *et al.*[26] (Δ), Parkhurst and Jonas[27] (×), Mamedov *et al.*[28] (+).

correlation. Thus, the average absolute percent deviation (AAD) is found with the expression AAD = (∑|PCTDEV|)/*n*, where the summation is over all *n* points, the bias percent is found with the expression BIAS = (∑PCTDEV)/*n*. The average absolute percent deviation of the fit is 1.34, and its bias is −0.02. For the liquid phase, at temperatures from 288 to 373 K at pressures up to 80 MPa, we estimate the uncertainty (at a 95% confidence level) to be 1.8%, increasing to 3.4% at 200 MPa, and 5% at pressures up to the correlation maximum. For the liquid at temperatures from 373 to 523 K, the uncertainty is 2.7% at pressures from saturation to 50 MPa, rising to 3.6% at 300 MPa. For temperatures above 523 K, we estimate the uncertainty in the liquid to be 5%. As mentioned previously, we estimate the expanded uncertainty of the correlation at a 95% confidence level for the low-density gas at temperatures from 305 to 640 K and pressures to 0.3 MPa to be 0.2%.

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 the density.

In Table 5, values are given for the viscosity along the saturation line, calculated from the present proposed correlation between 250 and 360 K. Values of the viscosity as a function of temperature and pressure are also given in Table 6.

Table 7 shows the AAD and the bias for the secondary data. It can be seen that, except for a few isolated cases, most of the secondary data agree very well with the proposed correlation. Finally, Fig. 10 shows a plot of the viscosity of benzene as a function of the temperature for different pressures. Figure 10 should be employed only within the limits of the correlation.

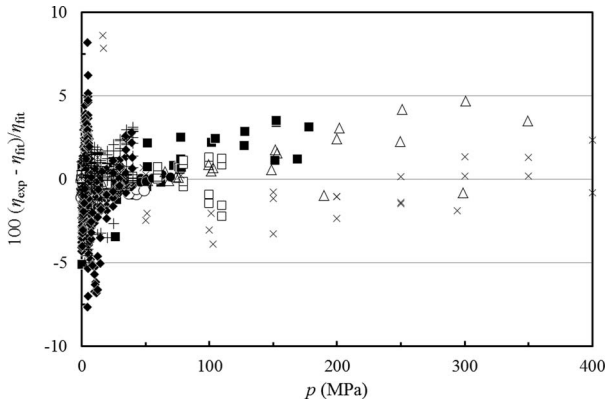


FIG. 8. Percentage deviations of primary experimental data of benzene from the values calculated by the present model as a function of pressure. dos Santos and Nieto de Castro[18] (■), Matsuo and Makita[19] (○), Assael *et al.*[20] (●), Knapstad *et al.*[21] (◒), Vogel *et al.*[22] (▲), Bauer and Meerlender[23] (◊), Abdulaev *et al.*[24] (♦), Kashiwagi and Makita[25] (□), Dymond *et al.*[26] (Δ), Parkhurst and Jonas[27] (×), Mamedov *et al.*[28] (+).

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)
280.00	892.702	795.3
300.00	871.470	586.4
320.00	849.999	455.2
340.00	828.145	365.1
360.00	805.753	299.4
380.00	782.656	249.3
400.00	758.650	209.9
420.00	733.477	178.1
440.00	706.783	151.8
460.00	678.042	129.6
480.00	646.421	110.5
500.00	610.458	93.57
520.00	567.192	77.93
540.00	508.839	62.26

TABLE 6. Viscosity values in μPa s, as a function of temperature and pressure

	T (K)										
p (MPa)	300	320	340	360	380	400	450	500	550	600	675
0	7.62	8.12	8.62	9.11	9.61	10.1	11.3	12.6	13.8	15.0	16.9
0.1	588.2	456.4	365.8	9.04	9.55	10.1	11.3	12.6	13.8	15.0	16.9
0.5	590.3	458.0	367.2	300.9	250.4	210.6	11.2	12.6	13.8	15.1	17.0
1	592.9	460.0	368.9	302.4	251.8	211.8	140.5	12.5	13.9	15.1	17.0
2	598.3	464.1	372.3	305.4	254.4	214.3	142.9	12.5	13.9	15.3	17.2
5	614.2	476.4	382.4	314.2	262.3	221.6	149.8	101.7	60.4	17.5	18.3
10	641.2	497.1	399.3	328.7	275.2	233.5	160.4	113.1	79.1	51.5	27.5
20	696.8	539.0	433.1	357.3	300.4	256.2	179.8	131.6	99.0	76.1	54.0
50	879.9	671.8	537.3	443.3	374.1	321.1	231.0	175.4	138.5	112.9	87.6
100	1268.3	931.6	729.9	595.5	499.8	428.1	309.5	237.7	190.1	156.8	123.1
200	2763.6	1734.3	1253.9	976.0	794.9	667.7	470.2	357.1	284.3	233.6	181.8
300				1552.1	1200.0	974.1	653.8	484.8	380.3	309.3	237.7

TABLE 7. Evaluation of the benzene viscosity correlation for the secondary data

1st author	Year Publ.	AAD (%)	BIAS (%)
Bhalodia[29]	2013	1.53	1.53
Bhatia[30]	2013	0.76	0.14
de Doz[31]	2011	0.15	0.15
Mariano[32]	2011	1.40	1.40
Pradhan[33]	2011	0.52	−0.52
Rahman[34]	2011	0.55	0.55
Shukla[35]	2011	0.43	−0.43
Dubey[36]	2010	0.31	0.31
Rathnam[37]	2010	1.02	1.02
Yadava[38]	2010	0.51	−0.51
Ali[39]	2009	0.15	0.15
El-Sayed[40]	2009	0.09	0.01
Sastry[41]	2009	0.14	0.01
Awwad[42]	2008	0.28	−0.22
Song[43]	2008	0.53	0.31
Wankhede[44]	2008	2.99	−1.83
Hernandez[45]	2007	1.85	0.72
Kharat[46]	2007	0.77	0.77
Nain[47]	2007	0.86	0.55
Reddy[48]	2007	0.66	0.66
Ren[49]	2007	0.90	−0.90
Song[50]	2007	0.44	−0.44
Yang[51]	2007	0.62	−0.62
Ali[52]	2006	0.39	0.39
Al-Kandary[53]	2006	0.93	0.93
Baragi[54]	2006	0.37	−0.37
Han[55]	2006	0.31	0.31
Varshney[56]	2006	0.30	0.30
Agarwal[57]	2004	0.98	0.98
Jain[58]	2004	4.14	−4.14
Ma[59]	2004	1.66	0.26
Oswal[60]	2004	0.27	−0.23
George[61]	2003	0.07	−0.07
Lark[62]	2003	0.63	−0.56
Nikam[63]	2003	0.77	0.77
Nayak[64]	2002	1.23	1.23
Chowdhury[65]	2001	0.73	0.73
Pandey[66]	2001	1.00	−1.00
Swain[67]	2001	14.78	14.78
Tsierkezos[68]	2000	1.57	1.57
Viswanathan[69]	2000	6.28	6.28
Aminabhavi[70]	1999	7.53	−7.53
Aralaguppi[71]	1999	0.46	0.46
Guzman[72]	1999	0.83	0.62
Sastry[73]	1999	1.46	1.46
Casas[74]	1998	1.68	−1.68
Emmerling[75]	1998	0.47	0.47
Exarchos[76]	1995	0.50	0.03

TABLE 7. Evaluation of the benzene viscosity correlation for the secondary data—Continued

1st author	Year Publ.	AAD (%)	BIAS (%)
Krishnan[77]	1995	0.05	−0.05
Petrino[78]	1995	0.73	−0.73
Rived[79]	1995	1.41	−1.41
Singh[80]	1995	0.15	0.15
Lafuente[81]	1994	1.60	−1.60
Rout[82]	1994	7.74	7.74
Shastri[83]	1993	0.13	−0.13
Klofutar[84]	1992	1.07	−1.07
Tasioula-Margari[85]	1992	0.49	0.49
Papaioannou[86]	1991	0.45	0.45
Zhang[87]	1991	1.83	1.73
Chevalier[88]	1990	0.20	0.20
Joshi[89]	1990	1.96	−1.04
Schumpe[90]	1990	0.88	0.88
Sekar[91]	1990	2.08	2.08
Sivaramprasad[92]	1990	6.96	6.96
Kouris[93]	1989	0.43	0.43
Ruiz[94]	1989	0.89	−0.89
Schrodt[95]	1989	0.09	0.09
Shah[96]	1989	1.73	1.73
Chandrasekhar[97]	1988	1.32	1.32
Rao[98]	1988	1.79	1.79
Manjeshwar[99]	1987	1.28	1.08
Ramanjaneyulu[100]	1987	0.06	−0.06
Fischer[101]	1986	10.74	10.74
Al-Mandfai[102]	1985	0.20	−0.20
Albright[103]	1983	0.18	0.18
Martin[104]	1983	0.72	−0.72
Nath[105]	1983	0.47	0.47
Asfour[106]	1981	0.08	0.08
Teja[107]	1981	0.63	−0.63
Yadava[108]	1981	0.41	−0.41
Bich[109]	1979	0.54	0.10
Gouel[110]	1978	6.08	6.01
Medani[111]	1977	2.21	−2.21
Vyunnik[112]	1977	0.58	−0.36
Dhillon[113]	1976	0.16	0.16
Bulanov[114]	1974	1.17	−1.02
Mekhtiev[115]	1974	0.82	−0.54
Alms[116]	1973	0.80	0.80
Collings[117]	1971	1.60	0.68
Konobeev[118]	1970	0.67	−0.02
Heric[119]	1967	0.34	0.34
Ridgway[120]	1967	0.61	0.61
Fort[121]	1966	0.03	−0.03
Katti[122]	1966	0.43	−0.43

TABLE 7. Evaluation of the benzene viscosity correlation for the secondary data—Continued

1st author	Year Publ.	AAD (%)	BIAS (%)
Bagdasarian[123]	1964	2.10	−2.05
Howard[124]	1959	0.51	−0.22
Teller[125]	1959	2.73	2.73
Ling[126]	1958	1.07	−1.07
Rabinovich[127]	1958	0.52	0.48
Airapetova[128]	1956	1.04	−0.64
Heiks[129]	1956	9.31	8.19
Toropov[130]	1956	0.88	−0.88
Belinskaya[131]	1955	1.83	1.83
Koshkin[132]	1955	0.74	0.74
Kuss[133]	1955	0.39	0.11
Mamedov[134]	1955	0.80	−0.64
Dixon[135]	1954	0.45	0.09
Grunberg[136]	1954	0.43	−0.43
Keeling[137]	1954	0.60	−0.60
Mukherjee[138]	1953	1.10	−0.89
Subnis[139]	1952	4.31	2.16
Craven[140]	1951	1.48	0.82
Jobling[141]	1951	1.68	−0.60
Golik[142]	1950	10.96	−10.96
Teitelbaum[143]	1950	7.27	7.27
Geist[144]	1946	0.43	−0.11
Linke[145]	1941	9.33	9.33
Udovenko[146]	1940	0.05	0.05
Khalilov[147]	1939	2.53	−1.54
Salceanu[148]	1939	0.32	0.32
Lemonde[149]	1938	0.44	0.44
Adzumi[150]	1937	2.33	2.33
Kireev[151]	1936	1.68	1.18
Titani[152]	1933	1.53	1.53
Piatti[153]	1931	63.39	40.69
Madge[154]	1930	0.48	−0.26
Tausz[155]	1930	0.43	0.28
Lowry[156]	1929	0.92	−0.15
Nasini[157]	1929	1.76	0.44
Bridgman[158]	1926	2.31	1.61
Kolossowsky[159]	1925	1.11	0.06
Lewis[160]	1925	1.16	−1.16
Bingham[161]	1920	0.81	−0.60
Faust[162]	1912	3.44	−3.22
Rappenecker[163]	1910	1.67	1.39
Findlay[164]	1909	1.58	−1.58
Dunstan[165]	1907	0.27	0.27
Heydweiller[166]	1896	3.30	−2.28
Thorpe[167]	1894	0.44	−0.24

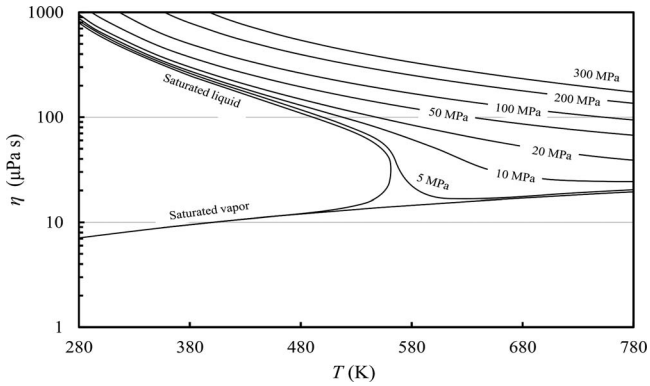


FIG. 10. Viscosity of benzene as a function of temperature for different pressures.

TABLE 8. Sample points for computer verification of the correlating equations.

T (K)	ρ (kg m^{-3})	η (μPa s)
300.00	0.00	7.625
400.00	0.00	10.102
550.00	0.00	13.790
300.00	875.00	608.52
400.00	760.00	211.74
550.00	500.00	60.511

4. Computer-Program Verification

Table 8 is provided to assist the user in computer-program verification. The viscosity calculations are based on the tabulated temperatures and densities.

5. Conclusion

A new wide-ranging correlation for the viscosity of benzene was developed based on critically evaluated experimental data. It consists of Eqs. (1)–(8), the coefficients found in Tables 2 and 3, and the equation of state of Thol *et al.*[13] to calculate density. The correlation is valid from the triple point to 675 K, and at pressures up to 300 MPa. The correlation is expressed in terms of temperature and density, with the density calculated from the equation of state of Thol *et al.*[13] The uncertainty estimate for the liquid phase at a 95% confidence level varies from 1.8% to 5%, and the uncertainty of the low-density gas at temperatures from 305 to 640 K and pressures to 0.3 MPa is estimated to be 0.2%.

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6. References

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