

Reference Correlation for the Viscosity of Ethane-1,2-diol (Ethylene Glycol) from the Triple Point to 465 K and up to 100 MPa

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We present a new wide-ranging correlation for the viscosity of ethane-1,2-diol (ethylene glycol) based on critically evaluated experimental data. The correlation is designed to be used with an existing equation of state, and it is valid from the triple point to 465 K, at pressures up to 100 MPa. The estimated uncertainty is 4.9 % (at the 95 % confidence level), except in the dilute-gas region which is estimated to be 15 %, as there are no measurements in this region for comparison. The correlation behaves in a physically reasonable manner when extrapolated to 750 K and 250 MPa, however care should be taken when using the correlations outside of the validated range.

Key words: ethane-1,2-diol; ethylene glycol; transport properties; viscosity.

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1 Introduction

Ethane-1,2-diol (IUPAC name) also known as 1,2-ethanediol, ethylene glycol, monoethylene glycol, or 1,2-dihydroxyethane, CAS 107-21-1, $C_2H_6O_2$, is a very common heat-transfer fluid. Nevertheless, reference correlations for its viscosity and its thermal conductivity do not exist, and the current formulations employed in REFPROP [1] are based on extended corresponding states [2]. The viscosity formulation is based on limited experimental data that cover a temperature range (288 to 373) K and is valid only to 10 MPa. Hence there is a need for a reference correlation for the viscosity of ethane-1,2-diol covering the widest possible range.

In a series of recent papers, reference correlations for the viscosity of selected common fluids [3-10] have been developed that cover a wide range of temperature and pressure conditions, including the gas, liquid, and supercritical phases. In this paper, the methodology adopted in the aforementioned papers is extended to developing a new reference correlation for the viscosity of ethane-1,2-diol.

The analysis that will be described is based on 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 [11]. 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 higher uncertainty, provided they are consistent with other lower uncertainty data or with theory. In all cases, the uncertainty claimed for the final recommended data must reflect the estimated uncertainty in the primary information.

2 The Correlation

The viscosity η can be expressed [3, 5-10] 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 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 [12-14] 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. This term for viscosity is significant only in the region very near the critical point, as shown in Vesovic et al. [15] and Hendl et al. [16]. Since we do not have data close to the critical point, $\Delta\eta_c(\rho, T)$ will be set to zero in Equation 1 and not discussed further in this work. 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.

Table 1 summarizes, to the best of our knowledge, the experimental measurements of the viscosity of ethane-1,2-diol reported in the literature. In the same table, the technique employed, the purity, the uncertainty as reported by the original authors, the number of measurements, as well as the range of temperatures and pressure investigated, are also shown.

Table 1 Viscosity theoretical predictions and measurements of ethane-1,2-diol.

Investigators / Reference	Technique employed ^a	Purity ^b (%)	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Primary Data						
Li et al. [17]	RBal	99.24	1.0	11	303-353	0.1
Moosavi et al. [18]	RBal	99.0	0.5	3	293-303	0.1
Hemmat et al. [19]	RBal	99.0	0.5	3	293-303	0.1
Losetty et al. [20]	RCyl	99.0	1.0	8	293-363	0.1
Fan et al. [21]	Cap	99.99	1.0	4	303-333	0.1
Zhao et al. [22]	Cap	99.0	1.0	6	298-323	0.1
Sun et al. [23]	Cap	99.9	0.2	5	298-318	0.1
Carvalho et al. [24]	RCyl	99.5	2.0	18	288-373	0.1
Li et al. [25]	Cap	na	0.1	4	313-343	0.1
Kondalah et al. [26]	Cap	99.5	0.2	1	308	0.1
Quijada-Maldonado et al. [27]	Cap	99.8	0.3	7	298-328	0.1
Sagdeev et al. [28]	FCyl	99.7	0.7-1.0	53	293-422 ^c	0.1-245

Sagdeev et al. [29]	FCyl	99.7	0.7-1.0	9	293-465	0.1
Li et al. [30]	Cap	na	1.0	3	298-308	0.1
Ge et al. [31]	RCyl	99.7	1.0	9	298-338	0.1
Sun and Teja[32]	Cap	99.8	2.0	8	296-428	0.1
Lech et al. [33]	RCyl	99.0	1.0	15	293-359	0.1
Marchetti et al. [34]	Cap	99.9	0.1	19	263-353	0.1
Lee and Teja [35]	Cap	99.8	1.5	8	293-423	0.1
Tanaka et al. [36]	FCyl	99.8	2.0	26	298-323	0.1-118
Bohne et al. [37]	Cap	na	2.0	9	263-373	0.1
Weber [38]	RBal	98.7	1.5	16	293-373	0.1-49
Sadykov et al. [39]	Cap	na	1.0	17	303-463	0.1
Litovitz et al. [40]	Cap	na	1.5	6	267-316	0.1

Secondary Data

Hoga et al. [41]	RCyl	99.9	14.0	4	293-308	0.1
Ciocirlan et al. [42]	FBal	99.5	5.0	3	293-303	0.1
Chasib [43]	Cap	99.3	na	1	298	0.1
Kinart et al. [44]	Cap	99.0	0.005	3	293-303	0.1
Doghaei et al. [45]	Cap	99.7	0.01	3	298-308	0.1
Zorębski and Lubowiecka-Kostka [46]	Cap	99.5	na	4	298-313	0.1
Mehta et al. [47]	Cap	99.5	0.01	1	308	0.1
Nain [48]	Cap	99.4	na	6	293-318	0.1
Zhang et al. [49]	Cap	99.4	1.0	4	308-323	0.1
Gurung and Roy [50]	Cap	99.5	0.003	3	298-318	0.1
Song et al. [51]	Cap	na	na	4	288-303	0.1
Wu et al. [52]	RCyl	na	2.0	5	333-413	0.1
Sinha and Roy [53]	Cap	99.0	0.003	3	298-318	0.1
Yang and Yu [54]	Cap	99.4	0.003	7	308-353	0.1
Naidu et al. [55]	Cap	98.5	0.5	1	308	0.1
Nayak et al. [56]	Cap	99.0	0.003	3	298-308	0.1
Sastry and Patel [57]	Cap	99.5	0.003	2	298-308	0.1
Yang et al. [58]	Cap	99.8	0.003	7	293-353	0.1
Cocchi et al. [59]	Cap	99.5	na	19	263-353	0.1
Saleh et al. [60]	Cap	99.0	na	5	303-323	0.1
Aminabhavi and Banerjee [61]	Cap	99.0	0.003	3	298-308	0.1
Tsierkezos and Molinou [62]	Cap	99.5	0.006	4	283-313	0.1
Pal and Singh [63]	Cap	na	na	2	303-308	0.1
Bilkis et al. [64]	Cap	na	na	5	303-323	0.1
Corradini et al. [65]	Cap	99.5	0.005	19	263-353	0.1
Reddy et al. [66]	Cap	na	na	1	308	0.1
Kumagai et al. [67]	RBal	99.0	3.4	4	273-333	0.1
Lux and Stockhausen [68]	na	na	na	1	293	0.1
Oyevaar et al. [69]	Cap	99.0	0.0001	1	298	0.1
Barbetova [70]	Cap	na	0.02	4	283-318	0.1
Idriss-Ali and Freeman [71]	Cap	99.0	na	4	298-358	0.1
Thomas et al. [72]	Cap	Pur	3.0	16	281-441	0.1
Jerome et al. [73]	Cap	na	0.3	1	298	0.1
Marks [74]	Cap	na	na	1	298	0.1

Kishimoto and Nomoto [75]	Cap	na	na	6	283-308	0.1
Timmermans and Hennaut-Roland [76]	na	na	na	5	288-298	0.1

^a Cap, capillary; FCyl, falling cylinder; RBal, rolling ball; RCyl, rotating cylinder; na, not available.

^b na, not available.

^c 5 points at pressures above atmospheric at the highest temperature of 465 K were not included, as discussed in the text.

There are three sets of viscosity measurements performed under high pressures.

- a) The 2012 measurements of Sagdeev et al. [28] were performed in a falling-cylinder viscometer up to 245 MPa and in the temperature range (293 to 465) K, with an uncertainty of 1 %. Measurements from this group have successfully been employed in previous correlations [4, 9]. However, we do note that in this case, it was impossible to fit the 5 measurements above atmospheric pressure at the highest temperature of 465 K, which is possibly attributed to high uncertainties in density, as it will be discussed at the end of this section. Hence these 5 measurements were excluded from the correlation.
- b) The 1988 measurements of Tanaka et al. [36] were also performed in a falling-cylinder viscometer up to 118 MPa pressure in the temperature range (298 to 323) K, with an uncertainty of 2 %. Measurements from this group have also been successfully employed in a previous correlation [4].
- c) Finally, in 1975, Weber et al. [38] employed a rolling-ball viscometer to perform measurements up to a pressure of 49 MPa in the temperature range (293 to 373) K, with an uncertainty of 1.5 %. This set was the only other set of measurements performed under higher pressure. Hence, these three sets were included in the primary data set.

As already mentioned in the introduction, ethane-1,2-diol is a very common heat transfer liquid. Indeed, it has lately been extensively employed as the base fluid in investigations of the increase in viscosity and thermal conductivity [77, 78] when very small amounts of nanoparticles or nanotubes are added. In such studies the viscosity of ethane-1,2-diol is also measured, but the emphasis is on the relative increase resulting from the addition of the nanoparticles. In recent literature there are many papers that include a single measurement of the viscosity of ethane-1,2-diol at room temperature with inadequate assessments of uncertainty. As water is the only liquid whose viscosity is known to an uncertainty as low as 0.17 % (at the 95 % confidence level) [11], all measurements in which the authors quote uncertainties of less than 0.01 % (e.g. 0.003 %!), characteristic of investigators that do not understand how to assess their measurement uncertainty, have been placed in the secondary data set. In addition, measurements quoting uncertainties higher than 2 %, and those without any assessment of uncertainty, were placed in the secondary data set. Finally, the capillary measurements of Zhang et al. [49], Wu et al. [52], Jerome et al. [73], and Naidu et al.

[55] were placed in the secondary data set, as they were 10 to 20 % lower than all other measurements. The remaining sets were kept in the primary data set.

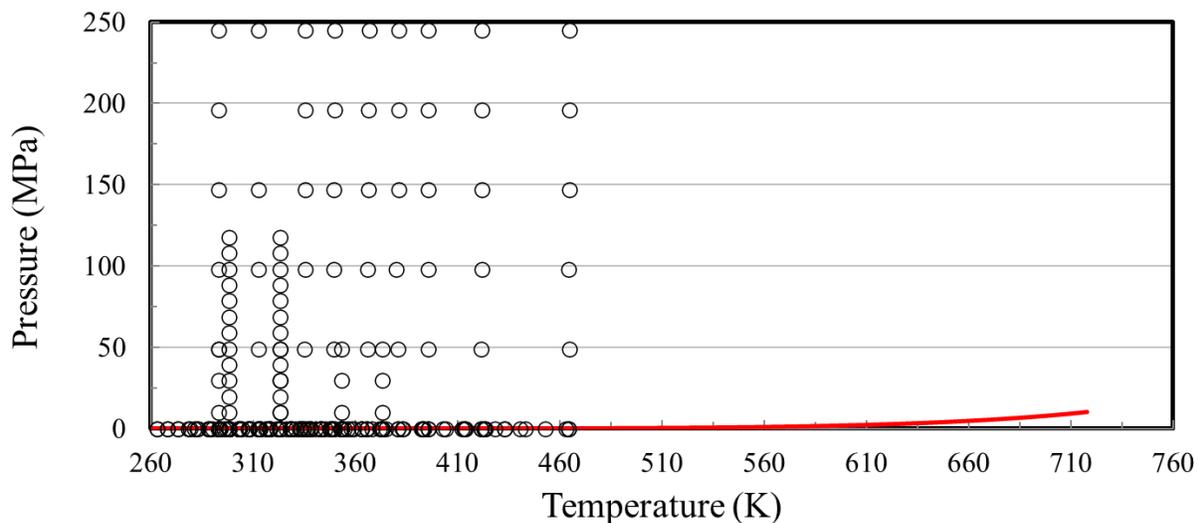


FIG. 1 Temperature-pressure ranges of the primary experimental viscosity data for ethane-1,2-diol.

(-) saturation curve.

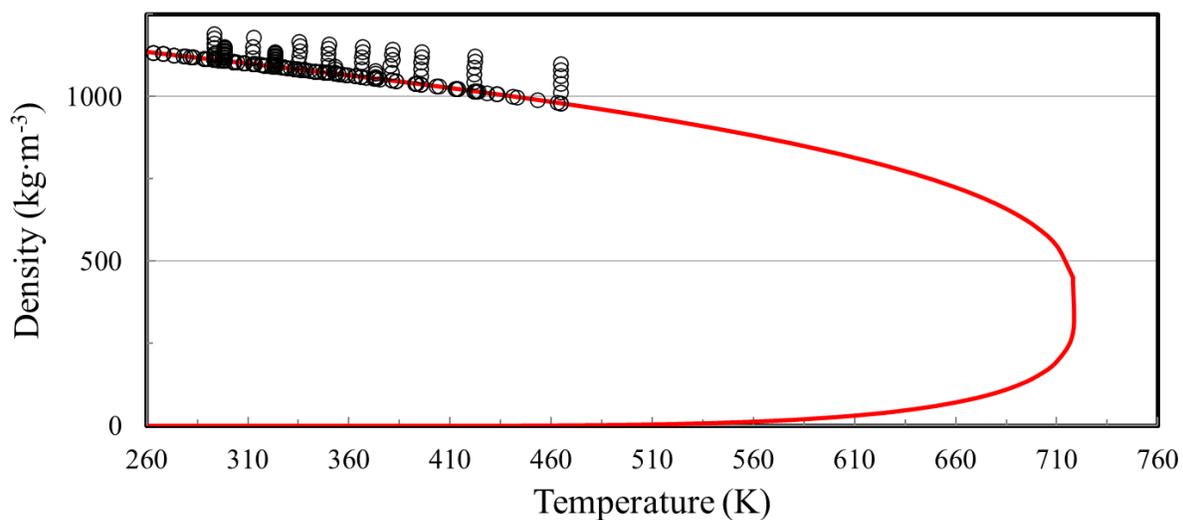


FIG. 2 Temperature-density ranges of the primary experimental viscosity data for ethane-1,2-diol. (-)

saturation curve.

Figures 1 and 2 show the ranges of the primary measurements outlined in Table 1, and the phase boundary. The lack of data in the vapor phase, near the critical temperature, and in the supercritical region is apparent. The development of the correlation requires densities; Zhou and Lemmon [79] developed an accurate, wide-ranging equation of state that is valid from the triple point up to 750 K and 100 MPa. It is available in REFPROP [1] but is not yet published. The equation of state has an uncertainty in density [79] of 0.15% in the liquid phase from 260 K to 360 K with pressures to 100 MPa, 0.3% in the liquid phase at pressures above 100 MPa and for all liquid states between 360 K to 400 K, and increase to 1% at higher temperatures, although the values above 400 K are not fully known due to a lack of high-temperature experimental data. Uncertainties in density in the vapor phase are also not known. We adopt the values for the critical point from their equation of state; the critical temperature, T_c , and the critical density, ρ_c , are 719.0 K and 364.9589 kg m⁻³, respectively. We also adopt the value used by Zhou and Lemmon [79] for the triple-point temperature, 260.6 K.

2.1 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 and is only a function of temperature. Assuming that the Lennard-Jones potential is applicable, one can use Chapman-Enskog theory [80] to express the dilute-gas viscosity as

$$\eta_0(T) = 0.02669 \frac{\sqrt{M_w T}}{\sigma^2 \Omega^{(2,2)}} \quad (2)$$

where M_w (g mol⁻¹), is the molar mass, and σ is the Lennard Jones collision diameter in nm, T is the temperature in Kelvin, and the resulting viscosity is in $\mu\text{Pa s}$. $\Omega^{(2,2)}$ is the Lennard-Jones collision integral that can be calculated by the empirical correlation developed by Neufeld [81] as a function of dimensionless temperature $T^* = k_B T / \varepsilon$ (where k_B is Boltzmann's constant and ε is the Lennard Jones energy parameter),

$$\begin{aligned} \Omega^{(2,2)}(T^*) = & 1.16145(T^*)^{-0.14874} + 0.52487e^{-0.7732T^*} + 2.16178e^{-2.43787T^*} \\ & - 6.435 \times 10^{-4} (T^*)^{0.14874} \sin \left[18.0323(T^*)^{-0.7683} - 7.27371 \right], \end{aligned} \quad (3)$$

Since there are no vapor-phase viscosity measurements for ethane-1,2-diol available, we employed the empirical equations proposed by Chung et al. [82, 83] to predict the scaling parameters σ (m) and ε/k_B (K), from

$$\sigma = 8.09 \times 10^{-8} (M_w / \rho_c)^{1/3} \quad \varepsilon / k_B = T_c / 1.2593 . \quad (4)$$

The resulting values are shown in Table 2. Eqs. 2-4 present a consistent scheme for the calculation of the dilute-gas limit viscosity of ethane-1,2-diol. Figure 3 shows the dilute-gas viscosity as a function of the temperature.

For ease of use in calculations η_0 was fit to a polynomial form:

$$\eta_0(T) = \sum_0^3 a_i (T / T_c)^i \quad (5)$$

where the units for η_0 are $\mu\text{Pa s}$, T is in K and the coefficients a_i are in Table 2. The above equation reproduces the scheme of Eqs. 2-4 to within 0.2 %, and thus it will be employed hereafter. Eq.(5) is easy to implement since it is a simple polynomial, but due to this fact it does not extrapolate well to high temperatures. Any investigative studies that involve behavior above 1500 K should employ the full scheme of Eq. (2)-(4) that has the correct extrapolation behavior. Since there are no experimental measurements in the vapor phase, the uncertainty in this region is estimated to be about 15 %. The application of the technique to other compounds, where vapor-phase measurements exist, e.g. *n*-hexane [10], and *n*-heptane [9] have demonstrated an uncertainty of 10%; however due to the polar nature of ethylene glycol the uncertainty of the method may be slightly higher.

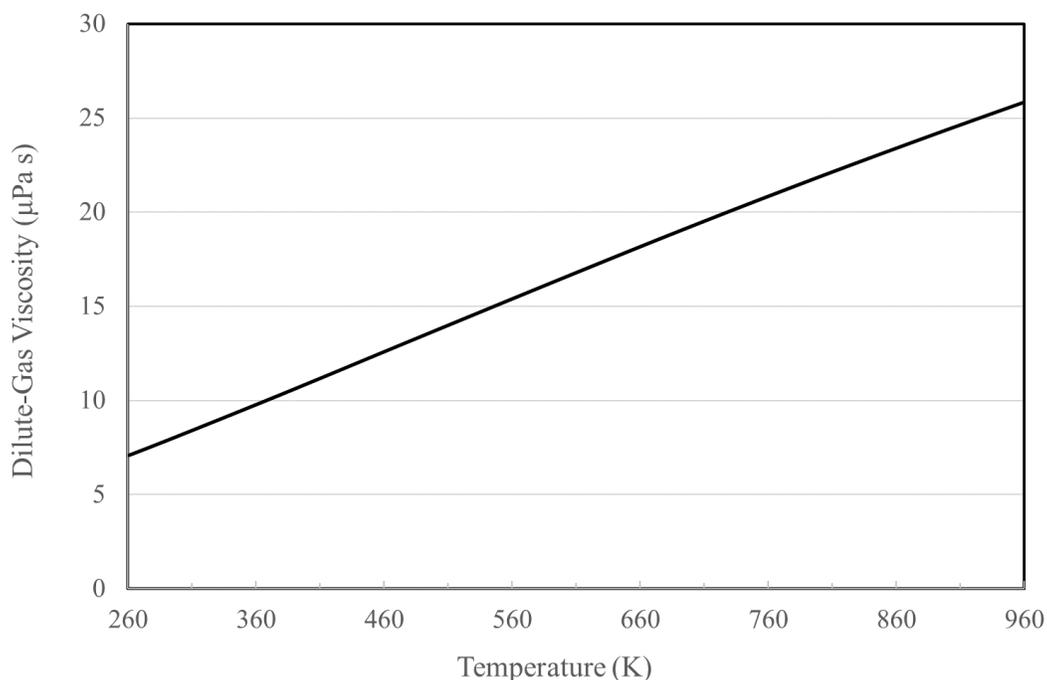


FIG. 3 Dilute-gas viscosity as a function of the temperature, calculated by the scheme of Eqs. 2-4.

The temperature dependence of the linear-in-density coefficient of the viscosity, $\eta_1(T)$, in Eq. 1 is large at subcritical temperatures and must be considered 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 [84, 85]. Vogel et al. [85] 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 $B_\eta(T)$ in $\text{m}^3 \text{kg}^{-1}$, as

$$B_\eta(T) = \frac{\eta_1(T)}{\eta_0(T)}. \quad (6)$$

Note that in the above equation, if the dilute-gas limit viscosity, $\eta_0(T)$, is expressed in $\mu\text{Pa s}$, then the initial-density viscosity coefficient, $\eta_1(T)$, will be expressed in $\mu\text{Pa s m}^3 \text{kg}^{-1}$. The second viscosity virial coefficient can be obtained according to the theory of Rainwater and Friend [12, 14] as a function of a reduced second viscosity virial coefficient, $B_\eta^*(T^*)$, as

$$B_\eta^*(T^*) = \frac{B_\eta(T)M_W}{N_A \sigma^3}, \quad (7)$$

where [85]

$$B_\eta^*(T^*) = \sum_{i=0}^6 b_i (T^*)^{-0.25i} + b_7 (T^*)^{-2.5} + b_8 (T^*)^{-5.5}. \quad (8)$$

In Eq. 7, N_A is the Avogadro constant. The coefficients b_i from Ref. [85] are given in Table 2. Figure 4 shows the calculated values of the initial-density viscosity coefficient, employing the scheme described by Eqs. 5-8.

Table 2 Coefficients and parameters for Eqs 2, 5, and 8.

ε/k_B (K)	570.9
σ (nm)	0.4482
M_w (g mol ⁻¹)	62.06784
T_c (K)	719.0

ρ_c (kg m⁻³) 364.9589

Coefficients a_i ($\mu\text{Pa s}$) for Eq. 5

0	4.14421×10^{-1}
1	1.69125×10^1
2	4.88979
3	-2.46114

Coefficients b_i (-) for Eq. 8 [85]

0	-1.9572881×10^1
1	2.1973999×10^2
2	-1.0153226×10^3
3	2.4710125×10^3
4	-3.3751717×10^3
5	2.4916597×10^3
6	-7.8726086×10^2
7	1.4085455×10^1
8	$-3.4664158 \times 10^{-1}$

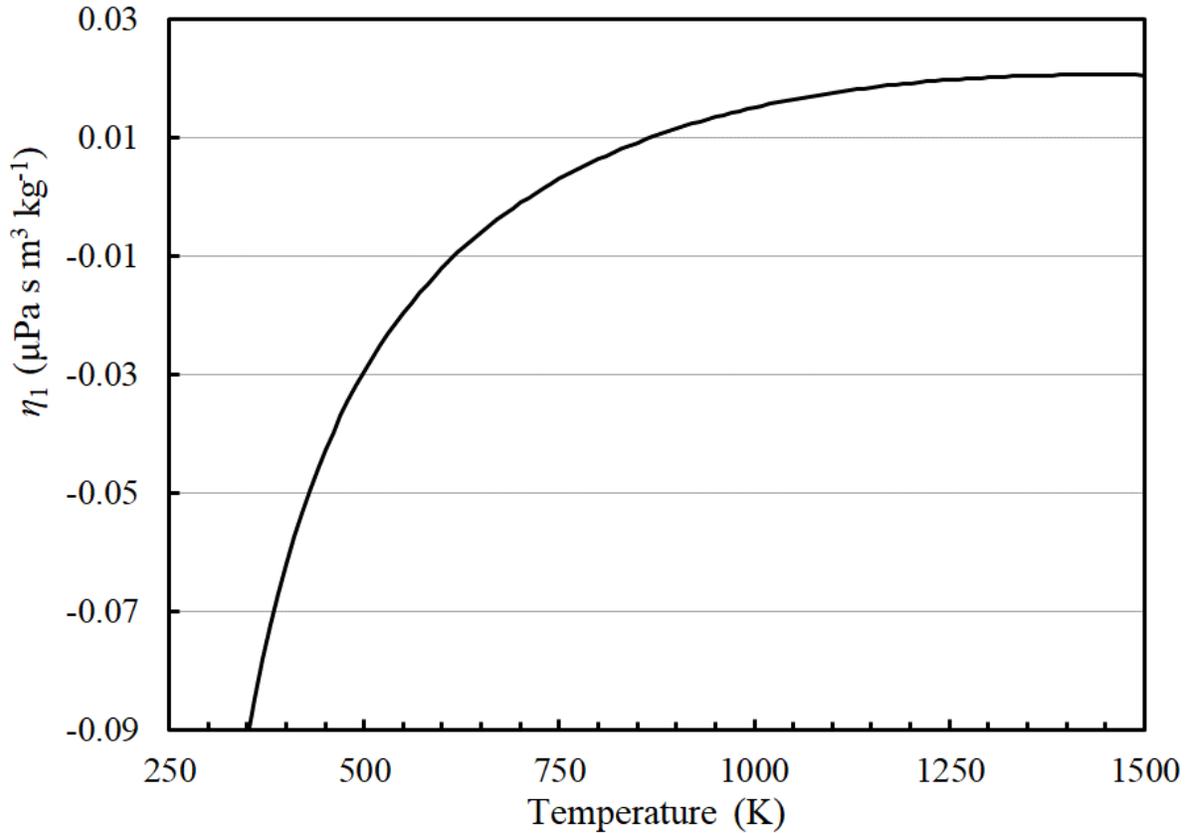


FIG. 4 Initial-density viscosity coefficient η_1 calculated from the scheme of Eqs. 5-8.

2.2 The residual term

As stated in Section 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. As there is no rigorous theory to calculate this term, it was evaluated almost entirely on experimentally obtained data, as discussed in the next paragraph.

The procedure adopted during this analysis used symbolic regression software [86] 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 R161 [3], *n*-undecane [6], R1234yf and R1234ze(E) [5], ammonia [87], and xenon [88]. 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 $\rho_r = \rho/\rho_c$. As mentioned earlier, the critical temperature $T_c = 719.0$ K and critical

density $\rho_c = 364.9589 \text{ kg m}^{-3}$ are from the equation of state of Zhou and Lemmon [79]. In addition, we adopted a form suggested by the hard-sphere model employed by Assael *et al.* [89], $\Delta\eta(\rho_r, T_r) = (\rho_r^{2/3} T_r^{1/2}) F(\rho_r, T_r)$, 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 term were calculated for each experimental point, employing Eqs. 5-8, and subtracted from the experimental viscosity to obtain the residual term, $\Delta\eta(\rho_r, T_r)$. The density values employed were obtained by the equation of state of Zhou and Lemmon [79]. The final equation obtained was

$$\Delta\eta(\rho, T) = \eta_{\text{ref}} (\rho_r^{2/3} T_r^{1/2}) \exp \left\{ c_0 + c_1 \rho_r + \frac{c_2 \rho_r}{T_r^2} + c_3 \frac{\rho_r^4}{T_r^3} + c_4 T_r + c_5 \rho_r^2 \right\} \quad (9)$$

Coefficients c_i are given in Table 3, and $\Delta\eta$ is in $\mu\text{Pa s}$.

Table 3 Coefficients c_i for Eq. 9.

c_0	2.5357387
c_1	1.8519087
c_2	6.9643296×10^{-2}
c_3	1.9804739×10^{-3}
c_4	-1.5781297
c_5	$-1.8187941 \times 10^{-1}$
$\eta_{\text{ref}} (\mu\text{Pa s})$	1.0

2.3 Comparison with data

Tables 4 and 5 summarize comparisons of the primary and secondary data with the correlation. We have defined the percent deviation as $\text{PCTDEV} = 100(\eta_{\text{exp}} - \eta_{\text{fit}})/\eta_{\text{fit}}$, where η_{exp} is the experimental value of the viscosity and η_{fit} is the value calculated from the correlation. Thus, the average absolute percent deviation (AAD) is found with the expression $\text{AAD} = (\sum |\text{PCTDEV}|)/n$, where the summation is over all n points, the bias percent is found with the expression $\text{BIAS} = (\sum \text{PCTDEV})/n$. The average absolute percentage deviation of the fit for all primary data is 1.83 %, with a bias of -0.03 %. The percentage standard deviation of the correlation from the triple point up to 465 K and 245 MPa is 4.9 % (at the 95% confidence level). Points at pressures above 100 MPa exceed the recommended limits of the equation of state but are included

as we feel the extrapolation of the equation of state from 100 MPa to 245 MPa behaves in a physically reasonable manner.

Table 4 Evaluation of ethane-1,2-diol viscosity correlation for the primary data.

Investigators / Reference	AAD (%)	BIAS (%)
Li et al. [17]	0.31	-0.31
Moosavi et al. [18]	1.90	1.70
Hemmat et al. [19]	3.28	3.28
Losetty et al. [20]	2.26	2.26
Fan et al. [21]	0.88	0.88
Zhao et al. [22]	1.99	1.34
Sun et al. [23]	2.48	0.33
Carvalho et al. [24]	1.50	1.50
Li et al. [25]	1.50	-1.50
Kondalah et al. [26]	3.56	-3.56
Quijada-Maldonado et al. [27]	1.29	-1.29
Sagdeev et al. [28]	1.16	0.41
Sagdeev et al. [29]	1.91	1.33
Li et al. [30]	1.01	-1.01
Ge et al. [31]	1.26	1.02
Sun and Teja[32]	2.47	-1.07
Lech et al. [33]	3.47	-0.28
Marchetti et al. [34]	1.70	1.68
Lee and Teja [35]	1.58	-1.58
Tanaka et al. [36]	3.13	-2.92
Bohne et al. [37]	1.89	-1.89
Weber [38]	1.28	-0.93
Sadykov et al. [39]	2.17	0.27
Litovitz et al. [40]	2.14	-0.17
Entire set	1.83	-0.03

Fig. 5 shows the percentage deviations of all primary viscosity data of ethane-1,2-diol from the values calculated by Eqs. 1 - 6, as a function of temperature, while Figs. 6 and 7 show the same deviations but as a function of the pressure and the density. The deviations of the experimental data from the present correlation are within about 5 %. This fluid would benefit from additional measurements, as some of the existing data sets do not agree with each other to within their experimental uncertainties. For example, both

Marchetti et al. [34] and Bohne et al. [37] provide data at atmospheric pressure, but at temperatures near 263 K they differ by about 7%, exceeding the estimated uncertainty of the original authors. Additional measurements at low temperatures could help resolve these differences. Similarly, the measurements of Sadykov et al. [39] and those of Sun and Teja[32] are at atmospheric pressure and differ by 6 % at ~428 K, again exceeding the uncertainty estimates of the original authors, and additional measurements at these conditions would be beneficial. We also observed that the 323 K isotherm of Tanaka et al. [36] shows a systematic deviation from the other data, and additional measurements at pressures above atmospheric could help clarify the behavior at higher pressure. Finally, there are no vapor-phase measurements at all. The availability of vapor-phase measurements would assist in better understanding of the gas-phase behavior of glycols.

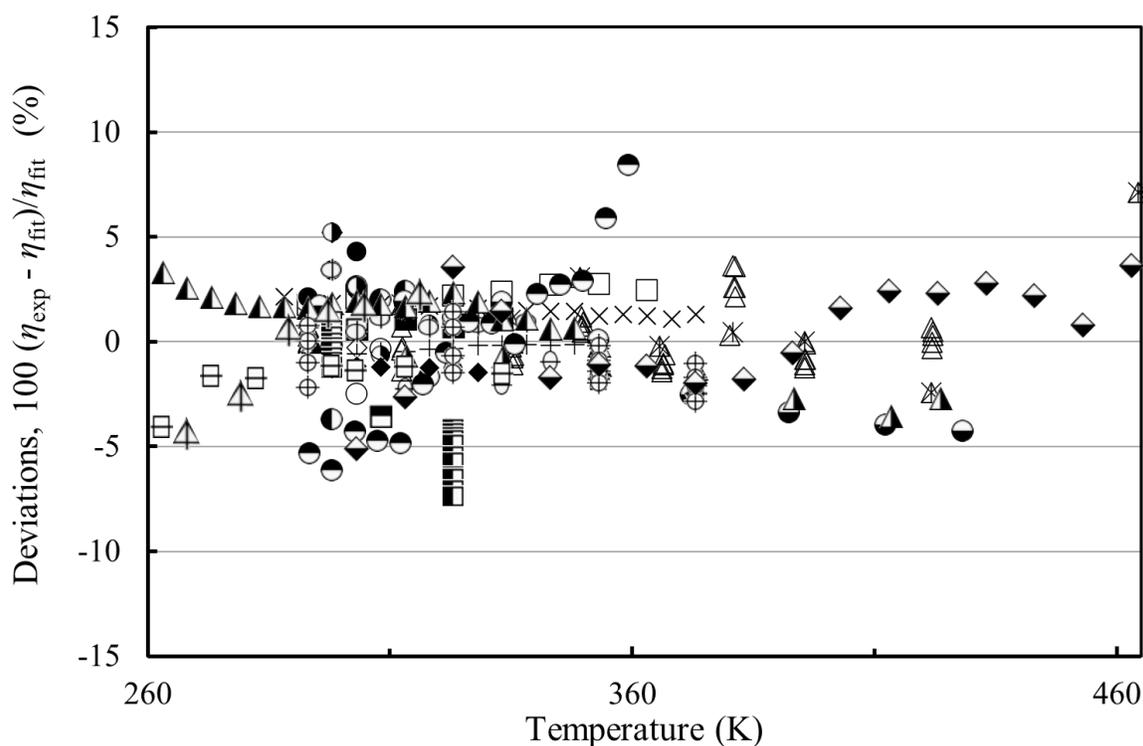


FIG. 5 Percentage deviations of primary experimental data of ethane-1,2-diol from the values calculated by the present model as a function of temperature. Li et al. [17] (+), Moosavi et al. [18] (\diamond), Hemmat et al. [19] (\bullet), Losetty et al. [20] (\square), Fan et al. [21] (\blacksquare), Zhao et al. [22] (\odot), Sun et al. [23] (\ominus), Carvalho et al. [24] (\times), Li et al. [25] (\oplus), Kondalah et al. [26] (\boxplus), Quijada-Maldonado et al. [27] (\blacklozenge), Sagdeev et al. [28] (\triangle), Sagdeev et al. [29] (\star), Li et al. [30] (\circ), Ge et al. [31] (Φ), Sun and Teja[32] (\bullet), Lech et al. [33] (\ominus), Marchetti et al. [34] (\blacktriangle), Lee and Teja [35] (\blacktriangle), Tanaka et al. [36] (\blacksquare), Bohne et al. [37] (\boxplus), Weber [38] (\oplus), Sadykov et al. [39] (\blacklozenge), Litovitz et al. [40] (\blacktriangle).

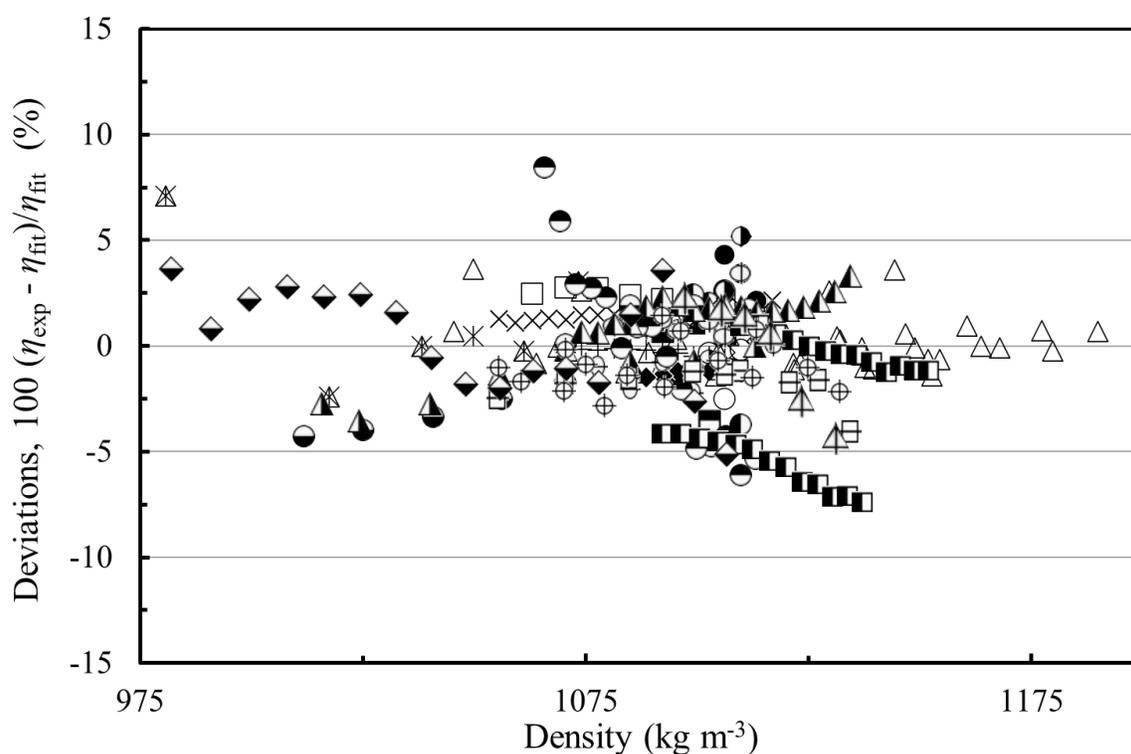


FIG. 7 Percentage deviations of primary experimental data of ethane-1,2-diol from the values calculated by the present model as a function of density. Li et al. [17] (+), Moosavi et al. [18] (\diamond), Hemmat et al. [19] (\bullet), Losetty et al. [20] (\square), Fan et al. [21] (\blacksquare), Zhao et al. [22] (\bullet), Sun et al. [23] (\bullet), Carvalho et al. [24] (\times), Li et al. [25] (\oplus), Kondalah et al. [26] (\blacksquare), Quijada-Maldonado et al. [27] (\blacklozenge), Sagdeev et al. [28] (\triangle), Sagdeev et al. [29] (\times), Li et al. [30] (\circ), Ge et al. [31] (\oplus), Sun and Teja [32] (\bullet), Lech et al. [33] (\bullet), Marchetti et al. [34] (\blacktriangle), Lee and Teja [35] (\blacktriangle), Tanaka et al. [36] (\blacksquare), Bohne et al. [37] (\boxplus), Weber [38] (\oplus), Sadykov et al. [39] (\blacklozenge), Litovitz et al. [40] (\blacktriangle).

Table 5 Evaluation of ethane-1,2-diol viscosity correlation for the secondary data.

Investigators/ Reference	AAD (%)	BIAS (%)
Hoga et al. [41]	1.66	-0.72
Ciocirlan et al. [42]	3.52	3.52
Chasib [43]	19.52	-19.52
Kinart et al. [44]	1.62	0.62
Doghaei et al. [45]	7.34	-7.34
Zorębski and Lubowiecka-Kostka [46]	0.63	0.63
Mehta et al. [47]	0.17	-0.17
Nain [48]	1.41	0.32
Zhang et al. [49]	19.47	-19.47
Gurung and Roy [50]	4.79	-4.79
Song et al. [51]	4.52	-4.52
Wu et al. [52]	18.84	-17.86
Sinha and Roy [53]	4.04	-4.04
Yang et al. [54]	1.47	-1.47
Naidu et al. [55]	6.88	-6.88
Nayak et al. [56]	29.57	-29.57
Sastry and Patel [57]	9.89	-9.89
Yang et al. [58]	1.27	-0.52
Cocchi et al. [59]	1.66	1.63
Saleh et al. [60]	2.10	-2.10
Aminabhavi and Banerjee [61]	10.82	-10.82
Tsierkezos and Molinou [62]	2.58	-2.04
Pal and Singh [63]	2.05	-2.05
Bilkis et al. [64]	2.11	-2.11
Corradini et al. [65]	1.70	1.69
Reddy et al. [66]	7.91	-7.91
Kumagai et al. [67]	7.50	-7.50
Lux and Stockhausen [68]	4.59	-4.59
Oyevaar et al. [69]	0.39	0.39
Barbetova [70]	3.70	3.27
Idriss-Ali and Freeman [71]	3.10	3.10
Thomas et al. [72]	6.00	-6.00
Jerome et al. [73]	12.18	-12.18
Marks [74]	116.18	116.18

Kishimoto and Nomoto [75]	10.39	-10.39
Timmermans and Hennaut-Roland [76]	1.66	0.70

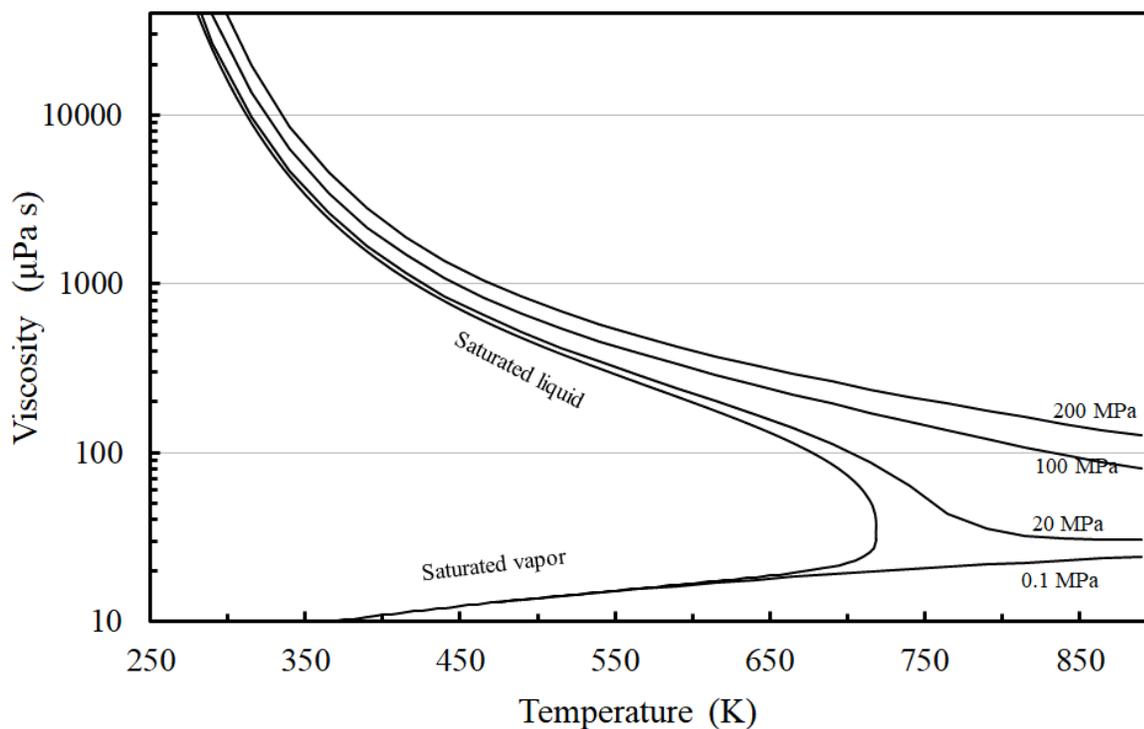


FIG. 8 Viscosity of ethane-1,2-diol as a function of the temperature for different pressures.

3 Recommended Values

In Table 6, viscosity values are given along the saturated liquid and vapor lines, calculated from the present proposed correlations between 261 and 465 K, while in Table 7 viscosity values are given for temperatures between 261 and 465 K at selected pressures. Saturation pressure and saturation density values for selected temperatures, as well as the density values for the selected temperature and pressure, are obtained from the equation of state of Zhou and Lemmon [79]. The calculations are performed at the given temperatures and

densities. For computer verification of values, the following points may be used for the given T , ρ conditions: $T = 350$ K, $\rho = 0.0$ kg m⁻³, $\eta = 9.522$ μ Pa s; $T = 350$ K, $\rho = 0.01$ kg m⁻³, $\eta = 9.525$ μ Pa s; $T = 350$ K, $\rho = 1100.0$ kg m⁻³, $\eta = 4246.78$ μ Pa s.

Table 6 Viscosity values of ethane-1,2-diol along the saturation line, calculated by the present scheme.

T (K)	p (MPa)	ρ_{liq} (kg m ⁻³)	ρ_{vap} (kg m ⁻³)	η_{liq} (μ Pa s)	η_{vap} (μ Pa s)
265	4.0017×10^{-7}	1133.065	1.1273×10^{-5}	95888.	7.19
290	5.6273×10^{-6}	1115.539	1.4486×10^{-4}	24038.	7.87
310	3.2874×10^{-5}	1101.469	7.9164×10^{-4}	10631.	8.42
330	1.5010×10^{-4}	1087.232	3.3956×10^{-3}	5604.6	8.97
350	5.6009×10^{-4}	1072.734	1.1948×10^{-2}	3352.7	9.53
370	1.7685×10^{-3}	1057.889	3.5694×10^{-2}	2200.9	10.08
390	4.8564×10^{-3}	1042.616	9.3043×10^{-2}	1548.8	10.64
410	1.1857×10^{-2}	1026.837	2.1631×10^{-1}	1148.8	11.21
430	2.6202×10^{-2}	1010.468	4.5663×10^{-1}	887.06	11.77
450	5.3184×10^{-2}	993.418	8.8837×10^{-1}	706.43	12.34

Table 7 Viscosity values of ethane-1,2-diol at selected temperatures and pressures, calculated by the present scheme.

p (MPa)	T (K)	ρ (kg m ⁻³)	η (μ Pa s)	p (MPa)	T (K)	ρ (kg m ⁻³)	η (μ Pa s)
0.1	265	1133.103	95949.	75	305	1133.741	18055.
	300	1108.560	15586.		330	1118.048	7607.9
	330	1087.278	5607.1		360	1098.992	3542.5
	360	1065.410	2690.6		390	1079.514	2004.6
	390	1042.670	1549.4		420	1059.464	1290.4
	420	1018.783	1005.4		450	1038.713	906.66
	450	993.452	706.56				
25	275	1135.429	60053.	100	315	1136.145	13671.
	300	1118.590	17636.		330	1127.043	8336.8
	330	1098.240	6242.6		360	1108.695	3842.3

	360	1077.452	2966.4		390	1090.016	2160.3
	390	1055.983	1699.1		420	1070.873	1385.3
	420	1033.620	1100.5		450	1051.159	971.68
	450	1010.149	774.31				
50	290	1134.499	31072.				
	300	1128.013	19842.				
	330	1108.476	6910.1				
	360	1088.614	3250.6				
	390	1068.218	1851.0				
	420	1047.111	1195.5				
	450	1025.131	840.96				

4 Conclusions

A new wide-ranging correlation for the viscosity of ethane-1,2-diol was developed based on critically evaluated experimental data and theoretical results. The dilute-gas limit viscosity in the present work was calculated from the Chapman-Enskog theory [80], by employing the empirical equations of Neufeld et al. [81] and Chung et al. [82, 83]. The initial-density dependence viscosity is based on the scheme proposed by Vogel et al. [85], and the residual term is based on a critically evaluated set of measurements. Due to the lack of experimental data in the vapor phase, the uncertainty in the dilute-gas region is estimated to approximately 15 %, while in all other cases it is 4.9 % (at the 95% confidence level). Since we could not locate any gas-phase measurements we recommend experimental measurements be made to assist in the understanding of the vapor-phase behavior of glycols. The correlation is valid for temperatures from the triple-point temperature to 465 K, a limit imposed by the range of the primary experimental data, while its pressure range extends to 100 MPa, a value up to which the equation of state is valid. Although there are multiple data sets at atmospheric pressure, many of these disagree by more than their uncertainties, especially for temperatures lower than 280 K and above 400 K; additional measurements in these regions, and at pressures above atmospheric, would be helpful for the development of an improved correlation. The correlation behaves in a physically realistic manner at temperatures and pressures up to 750 K and 250 MPa, respectively, and we feel the correlation may be extrapolated to this limit, although the uncertainty will be larger, and caution is advised.

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