

Reference Correlation of the Viscosity of Cyclohexane from the Triple Point to 700 K and up to 110 MPa

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Reference Correlation of the Viscosity of Cyclohexane from the Triple Point to 700 K and up to 110 MPa

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A new correlation for the viscosity of cyclohexane is presented. The correlation is based upon a body of experimental data that has been critically assessed for internal consistency and for agreement with theory. It is applicable in the temperature range from the triple point to 700 K at pressures up to 110 MPa. In the dilute gas region, at pressures below 0.3 MPa, the correlation is valid up to 873 K. The overall uncertainty of the proposed correlation, estimated as the combined expanded uncertainty with a coverage factor of 2, varies from 0.5% for the viscosity of the dilute gas and of liquid at ambient pressure to 5% for the viscosity at high pressures and temperatures. Tables of the viscosity generated by the relevant equations, at selected temperatures and pressures and along the saturation line, are provided. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4891103]

Key words: correlation; cyclohexane; transport properties; viscosity.

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1. Introduction

Cyclohexane is a colorless liquid that has widespread industrial use primarily as a raw material and nonpolar solvent, as well as being used in laboratories as a standard. It is, due to its simplicity, a prototypical cyclic molecule that provides a useful proxy molecule for modelling cycloalkanes and their interactions in mixtures. This is of special interest to the oil and gas sector, where cycloalkanes (or naphthenes) make up a high proportion of crude oil.¹

There is thus a need to establish reference values of thermophysical properties of cyclohexane that are both accurate and thermodynamically consistent. For some time, the thermodynamic properties have been described by the existing equation of state (EOS),² and their representation is being improved by the development of a new EOS (Ref. 3) that is both more accurate and is valid over a wider range of thermodynamic conditions. The situation concerning transport properties and, in particular, viscosity is not so good. At present, no correlation of viscosity is available and if one wants to predict the viscosity of cyclohexane, one has to rely on generic correlations^{4,5} developed for a wide variety of fluids that have invariably traded the range of applicability for accuracy.

Recently, research and development of state-of-the-art viscosity correlations have gained renewed impetus. Under the auspices of the International Union of Pure and Applied Chemistry (IUPAC), a research program has been initiated to develop representations of the viscosity and thermal conductivity of industrially important fluids. The basic philosophy of the program is to make use of the best available experimental data, selected on the basis of a critical analysis of the measurement methods. This information is complemented with guidance available from theory to produce accurate, consistent, and theoretically sound representations of the transport properties over the widest range of thermodynamic states possible. The first fluid studied in this program was carbon dioxide,⁶ and since then a plethora of viscosity correlations have been produced, using the same philosophy, covering among others: simple fluids,^{7–9} alkanes,^{10–17} and water.¹⁸ The present study is a continuation of this effort. The aim of this work is to critically assess the data available in the literature, and provide a correlation for the viscosity of cyclohexane that is valid over a wide range of temperature and pressure, covering the vapor, liquid, and supercritical fluid states.

2. Experimental Viscosity Data

Appendix A summarizes, to the best of our knowledge, the experimental measurements of the viscosity of cyclohexane reported in the literature, ^{19–205} detailing the temperature and pressure ranges, number of data points measured and the technique employed to perform the measurements. Overall, measurements of the viscosity of cyclohexane were reported in 186 papers resulting in 1427 data points. Unsurprisingly, the vast majority of researchers (165 papers, 504 data points) have measured only the value of the liquid viscosity at atmospheric pressure mostly around room temperature, although some of the measurements extend to higher temperatures. Appendix A

VISCOSITY OF CYCLOHEXANE

TABLE 1. Primary	data used in	developing the	e viscosity	correlation of	cyclohexane C
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Authors	Year publ.	Technique employed ^a	Purity (%)	Claimed uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Isdale <i>et al.</i> ⁸¹	1979	FB	99	2	14	298-373	$P_{\rm sat}$ -100
Dymond and Young ⁸⁴	1981	С	99	1	8	283-393	$P_{\rm sat}$
Kashiwagi and Makita ⁸⁸	1982	TC	99	2	51	298-348	0.1-110
Vogel et al. ¹¹³	1988	OB	99.9	0.3	97	298-632	0.0-0.3
Berstad ¹¹⁴	1989	OB	99.8	2	32	295-441	1.0-40
Grachev et al. ¹¹⁵	1989	С	_	-	285	473-673	0.1-60
Knapstad <i>et al.</i> ¹¹⁶	1989	OB	99.5	0.3	13	289-334	0.1
Papanastasiou and Ziogas ¹²⁹	1991	С	99.5	0.2	5	288-308	0.1
Tanaka et al. ¹³¹	1991	TC	99.8	2	26	298-348	0.1-100
Matsuo and Makita ¹³⁸	1993	С	99.5	2	18	298-313	0.1-48
Padua et al. ¹⁵²	1996	VW	99.85	3	25	298-348	0.1-38
Ihmels ¹⁵⁶	1998	С	99.99	0.5	11	293-343	0.1
Guzman et al. ¹⁵⁹	1999	С	99.9	-	3	283-313	0.1
Gascon <i>et al.</i> ¹⁶¹	2000	С	99.9	-	2	298-313	0.1
Roy et al. ¹⁹³	2009	С	99.7	-	1	298	0.1

^aC - capillary; FB - falling body; OB - oscillating body; TC - torsional crystal; VW - vibrating wire.

also contains a short list of reference works^{206–208} that report recommended tabulated values of the viscosity of cyclohexane. Not all the papers²⁰⁷ provide information on what experimental data were used to generate the tables.^{206–208}

Following the recommendation adopted by the IUPAC Subcommittee of Transport Properties [now known as The International Association for Transport Properties (IATP)], a critical assessment of the experimental data was performed to classify the data as primary and secondary. For this purpose, we used a set of well-established criteria²⁰⁹ that among other things classify primary data as data obtained with an experimental apparatus for which a complete working equation is available and for which a high precision in measuring the viscosity has been achieved. Furthermore, the criteria stipulate that guarantee of the purity of the sample, including the description of purification methods, should be available. 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 poorer accuracy, provided they are consistent with other more accurate data or with theory. Based on these criteria, 15 datasets were considered primary data. Table 1 summarizes the primary data^{81,84,88,113–116,129,131,138,152,156,159,161,193} detailing the temperature and pressure ranges, the authors' claimed uncertainty and purity of the sample, and the technique employed to perform the measurements. Some of the authors have only quoted the precision of their data, rather than uncertainty, in which case we left the relevant entry in Table 1 blank. The choice of primary data is discussed in more detail in Sec. 3, which also provides a comparison of the data by different workers.

Figure 1 shows the temperature and pressure range of the measurements outlined in Appendix A with primary and secondary data distinguished. The primary data cover a wide range of temperatures and pressures of interest. In the ranges where no primary data were available, the secondary data were

used to validate whether the proposed correlation extrapolates correctly.

Experimental measurements of viscosity are usually reported at a given temperature and pressure. In some cases, experimentally determined densities were also provided. For the development of a viscosity correlation that makes use of the available theory to provide guidance, temperature and density are the natural variables. Hence one requires an EOS to convert (*T*, *P*) pairs into corresponding (*T*, ρ) pairs. The use of EOS-generated density, rather than the one reported as part of the viscosity measurements, provides an additional level of consistency and further reduces the uncertainty of the developed viscosity correlation. For the purposes of this work, we have used a recent EOS developed by Zhou *et al.*³ that covers the thermodynamic space from the triple point to 700 K, and up



FIG. 1. Distribution of the available experimental viscosity data of cyclohexane. Primary Data: (\bullet) Isdale *et al.*;⁸¹ (\diamond) Dymond and Young;⁸⁴ (\bullet) Kashiwagi and Makita;⁸⁸ (\bullet) Vogel *et al.*;¹¹³ (\Box) Berstad;¹¹⁴ (Δ) Grachev *et al.*;¹¹⁵ (\blacksquare) Tanaka *et al.*;¹³¹ (\blacktriangle) Matsuo and Makita;¹³⁸ (x) Padua *et al.*;¹⁵² (\circ) data at 0.1 MPa. ^{116,129,156,159,161,193} Secondary data: (+).

to 250 MPa. Uncertainties in density are estimated to be $\pm 0.1\%$ up to 500 K, and $\pm 0.2\%$ above 500 K.

3. Methodology and Analysis

It is customary²¹⁰ in developing correlations of transport properties to take advantage of theoretical guidance on the functional form of the correlation as a function of temperature and density. Hence we express the viscosity η as the sum of four contributions,

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

where ρ is the molar density, T is the temperature, and the different contributions to viscosity, η_0 , η_1 , $\Delta\eta$, and $\Delta\eta_c$, are the zero-density viscosity, the first-density coefficient, the residual viscosity, and the critical enhancement, respectively. The advantage of decomposing the viscosity in this fashion is that it is possible to examine each contribution in turn. The current theories provide us with a good understanding of the dilute-gas regime, where only binary molecular interactions occur, and of the behavior near the critical point, where the long-range density fluctuations contribute to a divergence of the viscosity at the critical point.²¹¹ To a lesser extent we also understand the behavior of fluids at low density.²¹⁰ Hence, it is possible to make use of these theoretical developments, in conjunction with the available experimental data, to provide a more robust analysis of the zero-density viscosity, the first-density coefficient, and the critical enhancement than would have been possible by simply fitting to empirical functional forms.

In earlier works,^{6,210} the residual viscosity also contained the first density term $\eta_1(\rho)$. The separation became a sensible step once the theory of initial-density dependence advanced sufficiently to allow for an independent analysis.²¹⁰ Here, we refrain from naming $\Delta \eta$ as the "higher density terms of the residual viscosity,"^{11,12} and opt for a simpler "residual viscosity" which adequately describes it.

3.1. The zero-density limit

The viscosity in the zero-density limit is not a quantity directly accessible from experiment. It is obtained by extrapolating the measured data at low density, along a given isotherm. Several investigators have measured the viscosity of cyclohexane at low pressure, see Appendix A, but only the data of Refs. 26, 37, and 113 qualify for the development of the correlation at zero density. We have chosen the data of Vogel et al.¹¹³ as the only primary data set, as it covers an extensive temperature range (298-632) K, it was measured in a viscometer that satisfies all the requirements of a primary instrument, and based on previous measurements for other fluids it is demonstrably reliable within the low quoted uncertainty. However, note that the temperature range of the primary zero-density viscosity data is actually limited to (323 to 623) K, as only in this temperature range was the number of data points along each isotherm sufficient to perform an extrapolation to zero density.¹¹³

TABLE 2. Coefficients for the representation of the zero-density viscosity, Eqs. (2) and (3).

$A_0(-)$	B_0 (K)	$C_0 (\mathrm{K}^2)$
-1.5093	364.87	-39537

The viscosity in the zero-density limit $\eta_0(T)$ was represented by means of a standard relationship in kinetic theory,^{210,211} that in practical engineering form is given by

$$\eta_0(T) = \frac{1}{4} \sqrt{\frac{k_{\rm B}}{\pi N_{\rm A}}} \frac{\sqrt{MT}}{S_{\eta}} = 0.19592 \frac{\sqrt{T}}{S_{\eta}}, \qquad (2)$$

where *T* is the temperature in Kelvin, *M* is the molar mass, S_η is the effective collision cross-section in nm² and $\eta_0(T)$ is in µPa s. The numerical constant in Eq. (2) was obtained by the use of the recommended values of fundamental constants²¹² and a relative molecular mass of 84.15948.²¹³ In developing the viscosity correlation, experimental values of S_η have been derived from each of the primary data points and fitted to the empirical form,

$$\ln(S_{\eta}/\mathrm{nm}^{2}) = A_{0} + \frac{B_{0}}{T} + \frac{C_{0}}{T^{2}}, \qquad (3)$$

where A_0 , B_0 , and C_0 are the adjustable parameters. The primary data set of Vogel *et al.*,¹¹³ consisting of 11 data points, was fitted by means of Eqs. (2) and (3). The fitting parameters are given in Table 2 and the deviations of experimental data from the zero-density viscosity correlation, Eqs. (2) and (3), are given in Fig. 2. All the data are fitted within 0.15%, well within the claimed experimental uncertainty of extrapolated zero-density data of 0.2%–0.35%.

Traditionally, the development of a viscosity correlation in the zero-density limit relied on choosing a particular



FIG. 2. Percentage deviations $[100(\eta_{corr} - \eta_{exp})/\eta_{exp}]$ from the experimental data of Vogel *et al.*¹¹³ in the limit of zero density, of the values calculated by: (•) Eqs. (2) and (3); (•) Lennard-Jones correlation;²¹⁴ (•) universal correlation.^{215,216}

functional form for the effective cross-section, S_n , and then using two scaling parameters, one for length, σ , and the other for energy, ε , to perform the fitting.²¹⁰ Historically, two functional forms were used for this purpose: one that arises from assuming that molecular interactions can be represented by an effective spherical Lennard-Jones (12-6) potential²¹⁴ and the other that employs a universal correlation for S_{η} that is based on experimental data.^{215,216} The advantage of both functional forms is that extrapolation of the viscosity correlation to temperatures where measured data are not available is more reliable. Figure 2 illustrates the results of fitting the primary data for cyclohexane using the functional forms of S_n corresponding to Lennard-Jones and the universal correlation.^{214–216} By a judicious fitting of the scaling parameters (σ and ε), both the Lennard-Jones and universal correlation can be made to represent the data within systematic deviations of 1%, which is approximately 3 times as large as the claimed uncertainty of the measured data. This indicates that for large, cyclic molecules, the use of an effective spherical Lennard-Jones potential or the universal correlation is only valid at the 1% level of uncertainty.

3.2. The initial density dependence

At subcritical temperature, in the vapor phase, the viscosity below a certain temperature initially decreases with increasing density before increasing at higher densities.^{210,217,218} The existence of a viscosity minimum in density, which for cyclohexane occurs at temperatures below 520 K and is at 298 K of the order of 3% of the viscosity value, necessitates a separate analysis. It is customary to express the initial-density coefficient, η_1 , in terms of the second viscosity virial coefficient, B_{η} , by means of²¹⁰

$$\eta_1(T) = B_\eta(T)\eta_0(T).$$
 (4)

A number of workers^{217–220} have proposed functional forms for the second viscosity virial coefficient. These were initially based on modeling the moderately dense gas as a mixture of monomers and dimers which interact according to the LJ (12-6) potential, but are nowadays based on universal correlations developed by a judicious choice of experimental data and theory.^{12,220}

Although eight data sets exist in the vapor phase, only the data of Vogel *et al.*¹¹³ are sufficiently detailed to allow for the development of an initial-density correlation. We have, thus, used it as our only primary data set and have initially made use of the latest universal correlation¹² to represent it. The developed correlation with an optimal choice of scaling parameters σ and ε was found to predict the values quoted by Vogel *et al.*¹¹³ outside their uncertainties. An empirical approach was therefore adopted and the initial density dependence was represented by a simple functional form

$$\eta_1(T)\rho = \left(A_1 + \frac{B_1}{T} + \frac{C_1}{T^2}\right)\rho,$$
(5)

TABLE 3. Coefficients for the representation of the initial density dependence, Eq. (5)

A_1 (µPa s mol ⁻¹ l)	B_1 (μ Pa s K mol ⁻¹ l)	$\frac{C_1}{(\mu \text{Pa s } \text{K}^2 \text{mol}^{-1} \text{l})}$
5.09643	-3387.21	337 477

where A_1 , B_1 , and C_1 are the adjustable coefficients and ρ is the molar density in units of mol 1⁻¹. The primary data set of Vogel *et al.*,¹¹³ consisting of 97 data points, was fitted by means of Eq. (5). For this purpose the zero-density viscosity correlation was extrapolated to 298 K in order to include the 11 data points in this temperature range. The fitted parameters are given in Table 3 and deviations of experimental data from the initial density correlation, Eq. (5), are given in Fig. 3. All the data are fitted within 0.15%, which is within the claimed experimental uncertainty of 0.15%–0.3%. As the 11 data points in the temperature range (298–323) K were reproduced within their experimental uncertainty, one can surmise that the extrapolation of the zero-density correlation to 298 K is valid.

Figure 3 also illustrates the deviation of all the other data sets available in the vapor phase, at pressures of 0.3 MPa and below, from the correlation given by Eqs. (2)–(3) and (5). We observe that four data points by Craven and Lambart³⁷ are underestimated by 0.5%–1.0% by the proposed correlation. The deviation is just outside the claimed uncertainty of the experimental data and is of the same order as observed for other gases. All the other experimental data points lie in a cluster which is on average underestimated by 2.5% by the correlation, with no obvious systematic trend either in density or temperature. Only the data of Golubev⁶² cover a



FIG. 3. Percentage deviations $[100(\eta_{corr} - \eta_{exp})/\eta_{exp}]$ of the available experimental data in the vapor phase at pressures of 0.3 MPa and below from the calculated values. (•) Nasini;²⁶ (□) Titani;³¹ (•) Craven and Lambart;³⁷ (•) Golubev;⁶² (▲) Zhdanov and Lyusternik;⁶⁸ (*) Guseinov *et al.*;⁶⁹ (○) Vogel *et al.*¹¹³

This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: htp Physa Chem, Befa Data, Yol, 43, No. 3, 2014ed to IP: 129.31.244.5 On: Sun. 09 Nov 2014 11:20:08 temperature range (273-873) K that is outside the range covered by the primary data used in developing the correlation. The data⁶² consist of 12 smoothed viscosity values in the vapor phase quoted at atmospheric pressure. The tabulated values⁶² are based on the experimental measurements performed by Golubev and co-workers. We have used these smoothed viscosity values, as no measured values are available in the open literature. The value quoted at 273 K is most likely an extrapolated value, as it is unlikely that any measurements were made 6 K below the triple-point temperature. It would have required performing the measurements at a pressure below 5 mbar. As can be observed in Fig. 3, the data of Golubev⁶² do not display any systematic density trends, although they do have a bias. As these data are all at atmospheric pressure, the trend in density is the same as in temperature. Thus, it is safe to assume that the developed correlation has the correct temperature dependency when extrapolated to lower and higher temperatures.

Based on the agreement with the primary data set, we ascribe a combined expanded uncertainty with coverage factor of 2 to the viscosity correlation in the vapor phase, below 0.3 MPa, of 0.5% in the temperature range (323–673) K. Outside this temperature range, we estimate the uncertainty to increase to 1% at the triple point and 2% at 873 K.

3.3. The critical enhancement

In the vicinity of the critical point, the viscosity of a pure fluid exhibits an enhancement that diverges at the critical point.²²¹ Unlike thermal conductivity and heat capacity, where the critical enhancement is evident in a rather large region, the viscosity critical enhancement is of local character. The studies on CO₂ and C₂H₆ indicate that the ratio $\Delta \eta_c / \eta$ exceeds 0.01 only in the region which is approximately within 2% in temperature and 35% in density of the critical point.^{6,11} For cyclohexane this region would cover 542 < T/K < 565 and 2.10 $< \rho/(\text{mol l}^{-1}) < 4.35$. Only 2 of the viscosity data points measured by Grachev¹¹⁵ at 554.15 K fall within the critical region. However, our initial analysis of the data by Grachev appeared to indicate that the magnitude of the critical enhancement is much larger than expected. Figure 4 illustrates the behavior of Grachev's data¹¹⁵ along three isotherms (554.15, 554.65, and 558.15) K in the vicinity of the critical isotherm, 553.6 K. Although the viscosity data along the 554.15 K isotherm does indicate the presence of critical enhancement, its magnitude is inconsistent with previous studies. For instance, for ethane at 0.3 K above the critical temperature, the maximum critical enhancement is of the order of 1 µPa s, which corresponds to an increase of about 3% in viscosity.¹¹ For cyclohexane, however, the experimental data¹¹⁵ by Grachev at 2.567 mol 1^{-1} indicates a critical enhancement of 60%, which corresponds to an increase in viscosity of 13.5 µPa s. More importantly, the data along the 558.15 K isotherm show an unphysical decrease in viscosity around the critical region. This casts further doubt on the accuracy of the measurements made in this apparatus¹¹⁵ near the critical temperature. It is thus not possible to classify these data as primary nor to



FIG. 4. The ratio of experimental to the correlated viscosity, (η_{exp}/η_{corr}) , as a function of molar density, along the near-critical isotherms: (**u**) 554.15 K;¹¹⁵ (**b**) 554.65 K;¹¹⁵ (**c**) value of the critical density;³ (- -) boundary of the region where $\Delta \eta_c / \eta$ should be larger than 0.01.^{6,11}

determine the critical viscosity enhancement with any degree of certainty.

Hence, the lack of reliable data prevents the development of the critical enhancement contribution for cyclohexane. In line with the development of other correlations,^{7,9,10,12–17} the critical enhancement contribution is taken as zero.

3.4. The residual viscosity

There is no theoretical guidance for the residual-viscosity contribution and hence the existence of accurate experimental data covering a wide range of temperature and pressure is paramount for developing reliable correlations. Out of the available viscosity data sets, only 19 cover a wide range of temperature and pressure. These measurements were performed using a variety of instruments, not all of which could be considered as primary. Initially, the data of Matsuo and Makita¹³⁸ and Padua et al.,¹⁵² obtained in a capillary and in a vibrating-wire viscometer, respectively, were chosen. The primary data set, thus, covered the temperature range (298-348) K and pressures from 0.1 up to 48 MPa. Further analysis of the data of Isdale *et al.*,⁸¹ Kashiwagi and Makita,⁸⁸ Berstad,¹¹⁴ and Tanaka *et al.*¹³¹ indicated that these data agree with the initial selection of primary data within the claimed uncertainty over the whole range of temperatures and pressures. Hence, these data were also included in the primary data set. The main advantage of including these data is that the primary data covered pressures up to 110 MPa and temperatures up to 441 K. However, the upper temperature limit of 441 K still placed all the data firmly in the liquid part of the phase diagram. In order to extend the temperature range, we made use of data by Grachev et al.¹¹⁵ measured in a capillary viscometer that covered the temperature range (280-673) K and pressures up to 60 MPa. Although capillary viscometers are considered to be primary instruments, as the full working



FIG. 5. Percentage deviations $[100(\eta_{exp} - \eta_{fit})/\eta_{exp}]$ of the primary experimental viscosity data at 323 K and 348 K from the quadratic fit in density to Tanaka *et al.*¹³¹ data at each temperature. 323 K data: () Isdale *et al.*;⁸¹ (**n**) Kashiwagi and Makita;⁸⁸ (**•**) Grachev *et al.*;¹¹⁵ (×) Tanaka *et al.*;¹³¹ (**•**) Padua *et al.*;¹⁵² 348 K data: (**Δ**) Isdale *et al.*;⁸¹ (**n**) Kashiwagi and Makita;⁸⁸ (**•**) Grachev *et al.*;¹¹⁵ (×) Tanaka *et al.*;⁸⁸ (**•**) Grachev *et al.*;¹⁵²

equation is available, it was not clear from Ref. 115 if all the necessary corrections have been applied. Figure 5 illustrates the comparison of Grachev *et al.*¹¹⁵ data at 323 and 348 K with the other primary data measured at those temperatures.^{81,88,131,152} In order to perform the comparison between the different data sets, we used at each temperature a quadratic polynomial fit in density to Tanaka *et al.*¹³¹ data as the base line. It is clear from Fig. 5 that the Grachev *et al.*¹¹⁵ data exhibit a systematic deviation from the primary data set at all temperatures indicates differences of the order of 5%.

Although this would eliminate Ref. 115 from being considered as primary data, it was deemed useful to include a selection of these data in order to extend the temperature range of the correlation. Bearing in mind that this would also involve including measurements of uncertain provenance near the critical region, as discussed in Sec. 3.3, we have eliminated 18 data points that fall in the temperature range 548 < T/K < 558 and density range $2 < \rho/(\text{mol } 1^{-1}) < 5$. Thus, we have included the remaining 285 data points measured by Grachev *et al.*¹¹⁵ at temperatures from 473 to 673 K in the primary data set, but have increased the uncertainty to 5%. This choice did not jeopardize the low uncertainty of the developed correlation up to 441 K and it allowed for extension to higher temperature, albeit with the higher uncertainty.

Finally, the primary data set was supplemented with six sets of viscosity measurements^{116,129,156,159,161,193} of liquid cyclohexane at atmospheric pressure covering the temperature range (283–348) K and one set of measurements⁸⁴ along the liquid saturation line covering the temperature range (283–393) K. In summary, 495 data points covering the temperature range (283–673) K and pressures up to 110 MPa measured in 14 different viscometers were used as the primary data for the development of the residual viscosity contribution.

All the viscosity data were converted from the $\eta(T,P)$ to $\eta(T,\rho)$ representation by means of the recent EOS of Zhou et al.3 The residual viscosity was generated by subtracting from each data point the zero-density value, Eqs. (2) and (3), and the initial density contribution, Eq. (5). The resulting data set exhibits classical features of the $\eta(T,\rho)$ representation: (i) viscosity increases steeply at temperatures and densities near the solidification line and (ii) there are no data along subcritical isotherms at densities that lie within the two-phase region. This makes the choice of the functional form to fit the data rather difficult. As a result, a number of existing viscosity correlations exhibit non-monotonic behavior in the two-phase region. This is not surprising as there are no viscosity data at these densities to guide the correlation. Although this is not an issue if one is only interested in the viscosity of a pure substance, it limits the use of such viscosity correlations as a reference equation or to represent a particular species when calculating mixture viscosity. Hence, it precludes their use in corresponding states²¹⁰ or in VW models.^{222–224}

In this work, we have constrained the fitting of the experimental viscosity data in such a way that the resulting correlation within the two-phase region is a continuous, monotonically increasing function of density at all temperatures.

The residual viscosity is represented as a function in reduced temperature, $T_r = T/T_c$, and reduced density, $\rho_r = \rho/\rho_c$, as

$$\Delta \eta(\rho, T) = \sum_{i=2.2,2.5,2.8} \left(D_{i,0} + \frac{D_{i,1}}{T_{\rm r}} \right) \rho_{\rm r}^{i} + \sum_{i=10}^{14} \left(D_{i,0} + \frac{D_{i,1}}{T_{\rm r}} \right) \rho_{\rm r}^{i}, \tag{6}$$

where $D_{i,0}$ and $D_{i,1}$ are adjustable coefficients. In this instance, we have used fractional powers to allow us more flexibility in fitting the experimental data with the constraint imposed on the behavior in the two-phase region.

The procedure adopted during this analysis used the R software for statistical computing²²⁵ to fit Eq. (6) to the primary data. The uncertainties quoted in Table 1 were used to determine relative weights. As described previously, data from Grachev *et al.*¹¹⁵ were assigned an uncertainty of 5%. The primary data at atmospheric pressure where the authors have not quoted the uncertainty ^{159,161,193} were, for fitting purposes, assigned an uncertainty of 0.5%, in line with the other primary atmospheric data. The optimal coefficients $D_{i,0}$ and $D_{i,1}$ are shown in Table 4. The critical temperature T_c (553.6 K) and critical density ρ_c (3.224 mol l⁻¹) were obtained from Ref. 3.

Figures 6–8 illustrate the percentage deviations of the primary viscosity data used in developing the correlation from the developed viscosity correlation, Eqs. (1)–(3), (5), and (6). Figure 6 illustrates the agreement with the experimental data in the liquid region for temperatures from 283 to 441 K and pressures from saturation to 110 MPa. All the experimental data^{81,88,114,131,138,152} are reproduced by the proposed correlation within 2.5%, with no systematic trend in any of the data sets. The data of Dymond and Young⁸⁴ measured along the

TABLE 4. Coefficients for the representation of the residual viscosity, Eq. (6)

i	$D_{i,0}$	$D_{i,1}$
2.2	0	335.23400
2.5	7.8494803	-687.39760
2.8	0	362.08680
10	-10.4793856	2.5521774
11	17.2734993	-5.9372242
12	-10.6186149	4.3982781
13	2.8894928	-1.3468174
14	-0.2938491	0.1487134

saturation line are reproduced within 1%, which is within the claimed uncertainty of the data. The data of Isdale *et al.*⁸¹ exhibit an average absolute deviation (AAD) of 0.9%, bias of 0.7 and maximum deviation of 2.4%. Although the deviations of these data from the correlation are within the claimed uncertainty, there is a bias at high densities that was not fully reconciled within the proposed correlation. The deviations between different authors tend to increase with increasing density, leading to a larger uncertainty of the proposed correlation at higher pressures and at lower temperatures.

Figure 7 illustrates the agreement with the experimental data of Grachev *et al.*¹¹⁵ that were used to supplement the primary data set in the temperature range (473–673) K. The data are reproduced with an AAD of 1.7% and a maximum deviation of -8.4%. There was one datum (573.15 K, 60 MPa), not shown in Fig. 7, where a deviation of -18% was observed. Based on further analysis of the correlation in the vicinity of this point and the deviations of neighboring data points, it was concluded that this particular experimental point is an outlier. There are a couple of isotherms (523 K and 548 K) that exhibit systematic deviations from the correlation as one approaches



FIG. 6. Percentage deviations $[100(\eta_{exp} - \eta_{corr})/\eta_{exp}]$ of the primary experimental viscosity data in the liquid region from the values calculated by Eqs. (1)–(3), (5), and (6): (\bullet) Isdale *et al.*;⁸¹ (\Box) Dymond and Young;⁸⁴ (\bullet) Kashiwagi and Makita;⁸⁸ (\blacktriangle) Berstad;¹¹⁴ (\times) Tanaka *et al.*;¹³¹ (+) Matsuo and Makita;¹³⁸ (\bullet) Padua *et al.*¹⁵²



FIG. 7. Percentage deviations $[100(\eta_{exp} - \eta_{corr})/\eta_{exp}]$ of the primary experimental viscosity data of Grachev *et al.*¹¹⁵ from the values calculated by Eqs. (1)–(3), (5), and (6): (•) 473.15 K; (**n**) 523.15 K; (•) 548.15 K; (**A**) 598.15 K; (**D**) 623.15 K; (+) 648.15 K; (×) 673.15 K.

the liquid saturation density. Furthermore, at low densities the deviations systematically increase with decreasing pressure, reaching a maximum value of 4% at the lowest pressure measured (1 MPa). All this confirms that the Grachev data are of much higher uncertainty than the rest of the primary data and that assigning it a value of 5%, as we did earlier, is a reasonable conservative choice.

Figure 8 illustrates the agreement of the developed viscosity correlation with the primary experimental data at atmospheric pressure that cover the temperature range (283–373) K, in the



FIG. 8. Percentage deviations $[100(\eta_{exp} - \eta_{corr})/\eta_{exp}]$ of the primary experimental viscosity data measured at 0.1 MPa from the calculated values using Eqs. (1)–(3), (5), and (6). (•) Isdale *et al.*;⁸¹ (•) Kashiwagi and Makita;⁸⁸ (•) Grachev *et al.*;¹¹⁵ () Knapstad *et al.*;¹¹⁶ (*) Papanastasiou and Ziogas;¹²⁹ (+) Tanaka *et al.*;¹³¹ (×) Matsuo and Makita;¹³⁸ (-) Padua *et al.*;¹⁵² (\circ) Ihmels;¹⁵⁶ (\Box) Guzman *et al.*;¹⁵⁹ (\diamond) Gascon *et al.*;¹⁶¹ (Δ) Roy *et al.*;¹⁹³

TABLE 5. Evaluation of the cyclohexane viscosity correlation against the primary experimental data

	Year	AAD ^a	Bias ^b	MD ^c
Authors	publ.	(%)	(%)	(%)
Isdale <i>et al.</i> ⁸¹	1979	0.9	0.7	2.4
Dymond and Young ⁸⁴	1981	0.5	0.4	0.9
Kashiwagi and Makita ⁸⁸	1982	0.4	0.0	2.1
Vogel et al. ¹¹³	1988	0.1	0.0	-0.4
Berstad ¹¹⁴	1989	0.7	-0.7	-2.0
Grachev et al. ¹¹⁵	1989	1.7	0.2	-8.4
Knapstad et al. ¹¹⁶	1989	0.3	0.3	0.5
Papanastasiou and Ziogas ¹²⁹	1991	0.4	0.3	0.6
Tanaka <i>et al.</i> ¹³¹	1991	0.4	0.0	-1.0
Matsuo and Makita ¹³⁸	1993	0.7	-0.7	-1.7
Padua <i>et al.</i> ¹⁵²	1996	0.8	-0.7	-1.6
Ihmels ¹⁵⁶	1998	0.2	-0.1	-0.5
Guzman et al. ¹⁵⁹	1999	0.3	0.2	0.4
Gascon et al. ¹⁶¹	2000	0.5	-0.5	-0.6
Roy et al. ¹⁹³	2009	0.1	0.1	0.1
Entire primary data set		1.0	0.0	-8.4

^aAAD – Average absolute deviation; AAD = $100/N \sum |\eta_{exp} - \eta_{corr}/\eta_{exp}|$. ^bBias = $100/N \sum (\eta_{exp} - \eta_{corr})/\eta_{exp}$.

 $^{\rm c}MD$ – Maximum deviation.

liquid phase. All of the data are reproduced within 0.6%, with the exception of a single datum of Grachev et al.¹¹⁵ at 280 K. Although this is outside the claimed uncertainty of a number of authors, especially those who measured around room temperature, the spread of data between different laboratories does indicate that the proposed correlation represents the primary experimental data as well as could have been expected. Knapstad et al.¹¹⁶ and Papanastasiou and Ziogas¹²⁹ claim low uncertainty for their data, of the order of 0.2%-0.3% (see Table 2), and the mutual agreement observed between them (see Fig. 8) confirms it. It is reassuring that the proposed viscosity correlation predicts these two sets just outside their claimed uncertainty. The only systematic deviation is observed when comparing with the data of Ihmels.¹⁵⁶ However, the deviations are within the claimed uncertainty of the data, see Table 1. At low temperature the correlation underpredicts Ihmels's¹⁵⁶ data by 0.3%, while at high temperature it overpredicts by 0.45%. So although the data of Ihmels¹⁵⁶ agree with Knapstad et al.¹¹⁶ and Papanastasiou and Ziogas¹²⁹ at low temperatures, at 334 K the difference between their data¹⁵⁶ and that of Knapstad et al.¹¹⁶ is approximately 0.8%, which is just outside the combined uncertainty of both sets of data.

Table 5 summarizes the agreement between the primary experimental data and the proposed viscosity correlation for cyclohexane in the liquid and supercritical regions. The correlation recaptures the entire set of primary data with an AAD of 1.0%, zero bias and maximum deviation of -8.4%. Although the uncertainty of the correlation in the liquid and supercritical region can be estimated based on the overall agreement with the primary data set, we have taken a different approach and examined the agreement for different ranges of temperature and pressure. The choice of the temperature and pressure ranges was based on the availability of primary data and their quality, measured in terms of ascribed uncertainty. Thus we have estimated the overall uncertainty of the correla-

tion defined as the combined expanded uncertainty with a coverage factor of 2 as follows: (i) at atmospheric pressure in the temperature range from the melting point³ (279.52 K) to the boiling point³ (353.865 K), we estimate the uncertainty to be 0.5%; (ii) for pressures lower than 0.1 MPa, we estimate the uncertainty in the liquid region to be 1%; (iii) for pressures higher than 0.1 MPa and up to 110 MPa in the temperature range from the triple point to 441 K, we estimate the uncertainty to be 2%; (iv) everywhere else, in the liquid and supercritical region, we estimate the uncertainty to be 5%.

4. Overall Viscosity Correlation

The viscosity correlation of cyclohexane as a function of temperature and density is represented by Eqs. (1)–(3), (5), and (6) with the coefficients given in Tables 2–4. The correlation is valid in an extended temperature range. The lower temperature limit corresponds to the triple point of cyclohexane³ (279.45 K), while the upper temperature limit depends on the pressure. At densities corresponding to pressures below 0.3 MPa, the viscosity correlation is valid up to 873 K, while at higher pressures the viscosity correlation is valid to 700 K.

Figure 9 illustrates the behavior of the viscosity correlation as a function of density along the 300 K and 700 K isotherms. We observe a 100-fold increase in viscosity over the range of densities covered, with a steep increase in viscosity at the highest densities. Nevertheless, the proposed correlation is well-behaved within the two-phase region, where no data are available to constrain the correlation; for all isotherms, viscosity exhibits a monotonic increase with density. The behavior at densities corresponding to the two-phase region makes the present correlation suitable as the basis of developing a reference corresponding-states correlation for cyclic hydrocarbons²¹⁰ or as part of the VW model^{222–224} to predict the viscosity of mixtures containing cyclohexane.



FIG. 9. Viscosity of cyclohexane as a function of density along a couple of isotherms. (----) 300 K, liquid phase; (---) 300 K, 2-phase region; (----) 700 K.



Fig. 10. The extent of the viscosity representation and its estimated uncertainty. No representation is available in the hatched region.

It is possible to extrapolate the proposed viscosity correlation up to 200 MPa by making use of the densities calculated from the Zhou *et al.*³ EOS. The correlation extrapolates in a well-behaved fashion and the viscosity increases monotonically as a function of density along all isotherms. The validity of the extrapolation is supported to a certain extent by comparison with the secondary data. Jonas et al.⁸³ measured 25 points in the temperature range from 313 to 383 K, up to 210 MPa. Six measurements of viscosity were performed at pressures above 110 MPa. The AAD and MD of these six points are 4.0% and -7.6%, respectively, which compares favorably with the AAD (3.6%) and MD (-9.8%) of the 19 points at pressures that fall within the pressure range of the current correlation. Furthermore, the single data point of Isdale *et al.*⁸¹ at 373 K and 200 MPa, that was not part of the primary data set, is reproduced with a deviation of 1.8%.

Figure 10 summarizes the estimated combined expanded uncertainty with coverage factor of 2 of the proposed viscosity correlation as a function of temperature and pressure.

TABLE 7. Recommended viscosity values along the saturation line

		Vap	or	Liqu	ıid
<i>T</i> (K)	P _v (MPa)	$\frac{\rho}{(\text{mol } l^{-1})}$	η (μPas)	$\frac{\rho}{(\text{mol } l^{-1})}$	η (μPa s)
293.15	0.0103	0.0043	6.91	9.2515	971.3
303.15	0.0162	0.0065	7.11	9.1393	818.9
313.15	0.0246	0.0096	7.30	9.0258	700.4
323.15	0.0363	0.0137	7.49	8.9111	606.4
333.15	0.0519	0.0192	7.69	8.7949	530.2
343.15	0.0726	0.0262	7.89	8.6770	467.4
353.15	0.0992	0.0351	8.09	8.5574	414.8
403.15	0.3625	0.1191	9.16	7.9250	245.5
453.15	0.9664	0.3143	10.62	7.1971	158.86
503.15	2.1061	0.7500	13.12	6.2406	103.24
542.15	3.5328	1.6610	17.11	4.9161	61.19

Table 6 contains the recommended values of viscosity of cyclohexane at a selected number of temperatures and pressures which broadly cover the range of the proposed viscosity correlation. Table 7 contains the recommended values of viscosity of cyclohexane along the saturation line.

Appendix B contains Figs. 13–15 that summarize the deviations of the secondary data from the current correlation. Although some data exhibit deviations that are similar to those observed for the primary data, there are a number of experimental data sets that exhibit large and systematic deviations that cannot be reconciled with the other available data.

4.1. Comparison with the available recommended data

Although no other viscosity correlation of cyclohexane is available in open literature, there are a number of tables of recommended values^{206–208} together with the Refprop software implementation.²²⁶ In this section, a brief comparison is carried out. Figure 11 illustrates the deviation of the viscosity values calculated using the Refprop correlation²²⁶ from the

TABLE 6. Recommended viscosity values in µPa s

Р	<i>T</i> (K)										
(MPa)	290	300	310	320	330	340	350	400	500	600	700
0	6.86	7.06	7.26	7.46	7.66	7.87	8.07	9.11	11.19	13.19	15.09
0.1	1029.7	863.7	735.5	634.4	553.0	486.2	430.5	252.0	94.84	13.20	15.11
0.5	1035.3	868.3	739.4	637.7	555.9	488.8	432.9	253.7	97.76	13.30	15.22
1	1042.4	874.2	744.3	641.9	559.6	492.1	435.8	255.7	100.93	13.52	15.42
2	1056.6	885.9	754.2	650.4	567.0	498.7	441.8	259.9	106.34	14.27	16.00
4	1085.5	909.7	774.1	667.5	581.9	511.9	453.7	268.1	115.16	16.77	17.63
6	1114.9	933.9	794.4	684.8	597.0	525.3	465.7	276.3	122.58	24.27	19.70
8	1144.7	958.4	815.0	702.4	612.3	538.8	477.8	284.4	129.22	46.82	22.48
10	1175.1	983.4	835.8	720.2	627.7	552.4	490.0	292.5	135.33	60.27	26.85
30	1509.1	1256.6	1062.8	912.0	792.6	696.5	617.9	374.4	185.32	113.67	74.45
50	1901.4	1576.0	1326.3	1132.5	979.9	858.1	759.3	460.6	230.5	148.20	104.96
70	2356.6	1945.9	1630.1	1384.9	1192.6	1039.8	916.7	552.7	277.2	178.53	131.18
90	2878.6	2369.6	1977.0	1671.9	1432.7	1243.3	1091.5	651.1	327.0	208.3	154.63
110	3470.8	2850.1	2369.7	1995.5	1702.1	1469.9	1284.6	755.6	380.0	239.5	176.75

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FIG. 11. Percentage deviations $[100(\eta_{corr} - \eta_{Refprop})/\eta_{corr}]$ of the viscosity values calculated using the Refprop correlation²²⁶ from the current correlation along a selected isotherms: $(\cdot \cdot \cdot \cdot)$ 280 K; (--) 300 K; $(-\cdot \cdot \cdot)$ 400 K; $(-- \cdot)$ 500 K; (--) 600 K; (--) 700 K.

current correlation along a selected number of isotherms. The agreement at low temperatures in the liquid phase is good, while at higher temperatures (T > 400 K) larger and more systematic differences are observed.

Figure 12 illustrates the deviation of the available tabulated values of the viscosity of cyclohexane from those obtained using the current correlation. The tabulated data given in Refs. 207 and 208 are in good agreement at low temperatures, but at higher temperatures the tabulated values exhibit increasingly higher deviations. The tabulated data of Stephan and Lucas²⁰⁶ show systematic deviations that increase with tem-

FIG. 12. Percentage deviations $[100(\eta_{exp} - \eta_{corr})/\eta_{exp}]$ of the selected tabulated viscosity data^{206–208} from the calculated values using Eqs. (1)–(3), (5), and (6). (\circ) Stephan;²⁰⁶ (**■**) Liessmann;²⁰⁷ (\blacklozenge) TRC 2003.²⁰⁸

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

Т	ρ	η
(K)	$(\text{mol } l^{-1})$	(µPa s)
300	0	7.058
300	0.0430	6.977
300	9.1756	863.66
300	9.9508	2850.18
500	0	11.189
500	6.0213	94.842
500	8.5915	380.04
700	0	15.093
700	7.4765	176.749

perature for any pressure. The trend can be traced to the choice of primary data by Stephan and Lucas who relied on the data of Guseinov *et al.*⁶⁹ at higher temperatures and pressures.

5. 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.

6. Conclusion

A new wide-ranging correlation for the viscosity of cyclohexane has been developed based on critically evaluated experimental data. The correlation is valid from the triple point to 873 K at pressures below 0.3 MPa, and from the triple point to 700 K at pressures up to 110 MPa. The correlation is expressed in terms of temperature and density, where the densities were obtained from the equation of state of Zhou *et al.*³ The overall uncertainty, using a coverage factor of 2, of the proposed correlation is less than 5%, however this uncertainty varies depending on thermodynamic state and is summarized in more detail in Fig. 10.

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7. Appendix A: Viscosity Measurements of Cyclohexane

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Authors	Year publ.	Technique employed ^a	No. of data	Temperature range (K)	Pressure range (MPa)
Drapier ¹⁹	1911	С	5	290-308	0.1
Thole ²⁰	1914	С	1	298	0.1
Chavanne and Risseghem ²¹	1922		3	288-303	0.1
Vorlaender and Walter ²²	1925		1	293	0.1
Bridgman ²³	1926		6	303–348	0.1–147
Timmermans and Martin ²⁴	1926		3	288–333	0.1
Lowry and Nasini ²⁵	1929		2	293–377	0.1
Nasini ²⁶	1929	С	16	319–478	0.01-0.1
Godchot and Cauquil ²⁷	1931	С	1	287	0.1
Locket ²⁰	1932	С	1	298	0.1
Vorlaender and Fischer ²⁹	1932		1	290	0.1
MacFarlane and Wright ³⁰	1933	С	1	298	0.1
Titani ³¹	1933	С	7	395–580	0.1
Waller ³²	1934		2	280–355	0.1
Evans ³³	1938		6	281-348	0.1
Linke ³⁴	1941		5	287-313	0.1
Friend and Hargreaves ³⁶	1944		5	284-349	0.1
Geist and Cannon ³⁵	1946	0.5	2	293-313	0.1
Craven and Lambert ³⁷	1951	OB	4	308-351	0.03
Hase ³⁰	1953		1	298	0.1
Mukherjee ³³	1953	~	7	303-333	0.1
Dixon and Schliessler ⁴⁰	1954	С	3	293–333	0.1
Grunberg ⁴¹	1954	С	1	298	0.1
Kuss ⁴²	1955	FB/C	22	293–353	0.1–130
Donald and Ridgway ⁴⁵	1956	C	l	298	0.1
Donald and Ridgway ⁴⁴	1958	С	1	298	0.1
Lutskii and Obukhova ⁴⁵	1961		1	298	0.1
Toropov and Ermokhina ⁴⁰	1961		2	313–333	0.1
Homes <i>et al.</i> ⁴⁷	1962		1	298	0.1
Bamelis <i>et al.</i> ⁴⁰	1965		4	298-328	0.1
Belinskii and Khodzhaev ⁴⁹	1965		7	303	0.1–59
Fort and Moore ⁵⁰	1966	С	1	298	0.1
Katti <i>et al.</i> ⁵¹	1966	С	1	298	0.1
Kozlov <i>et al.</i> ⁵²	1966		15	283-413	$P_{\rm sat}$ -101
Meyer and Metzger ⁵⁵	1967	C.	1	298	0.1
Ridgway and Butler ³⁴	1967	С	1	298	0.1
Meyer <i>et al.</i> ⁵⁶	1968		l	298	0.1
Neduzhii and Khmara ⁵⁰	1968		8	280-350	0.1
Raman <i>et al.</i> ⁵⁷	1968		1	303	0.1
Brzostowski ⁵⁰	1969		l z	298	0.1
Mato and Coca ⁵⁵	1969		5	298-318	0.1
Mato and Hernandez	1969		5	288-323	0.1
Prados ⁶¹	1969		5	298-318	0.1
Golubev ⁵²	1970		22	2/3-8/3	0.1
Konobeev and Lyapin ⁴⁴	1970	TC	4	293-353	0.1
Collings and McLaughlin [®]	1971	IC	1/	303-323	0.1-/8
Prokorev <i>et al.</i>	1971		11	293-493	0.1
Campbell and Anand	1972		14	298-323	0.1
Nissema	1972		3	293-313	0.1
Zhdanov and Lyusternik	1972	C	15	348-662	0.05-0.1
Guseinov et al.	1973	U	110	290-523	0.1-49
Ishihara <i>et al.</i>	1974	C	1	313	0.1-59
Jain et al.	1974	Ľ	1	303	0.1
Kuskova and Matizen	1974		5	293-333	0.1
Meknuev et al.	1974	TC	0	293-343	0.1
Strumpi <i>et al.</i>	1974	IC	8	288-323	0.1
Dhillon and Bhatia	1975		I	298	0.1
Mekhtiev <i>et al.</i> ⁷⁷	1975		6	293-343	0.1
Zakurenov <i>et al.</i>	1975	C	15	293-508	0.1
Dhillon and Chugh	1976	U	2	298-308	0.1
INISSEMA and Saeynaejaekangas	19/6		4	293-323	0.1
Lega-Dalkowska <i>et al.</i>	1977	FD	3	280-323	U.1
Isuale <i>et al.</i>	1979	FB	15	298-373	P_{sat} -100
Keddy and Naidu ²²	1979	DD	1	303	0.1
Jonas <i>et al.</i> ⁵⁵	1980	KB	25	313-383	0.1–210
Dymond and Young	1981	C	8	283-393	$P_{\rm sat}$
r adava and r adava	1981	C	1	307	0.1
Ammadnavi et al.	1982	C	1	293	0.1

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TABLE . —Continued

Authors	Year publ.	Technique employed ^a	No. of data	Temperature range (K)	Pressure range (MPa)
Bauer and Meerlender ⁸⁷	1982	С	1	298	0.1
Kashiwagi and Makita ⁸⁸	1982	TC	51	298-348	0.1-110
Albright et al. ⁸⁹	1983	С	1	298	0.1
Cholpan <i>et al.</i> ⁹⁰	1983		4	293-323	0.1
Dhillon and Kaur ⁹¹	1983		1	308	0.1
Martin <i>et al.</i> ⁹²	1983		5	303-343	0.1
Nath and Narain ⁹³	1983	С	1	303	0.1
Yamada <i>et al.</i> ⁹⁴	1983		1	353	0.1
Bauer and Meerlender ⁹⁵	1984	С	3	293–298	0.1
Martin <i>et al.</i> ⁹⁶	1984		5	293-333	0.1
Nath and Dixit ⁹⁷	1984	С	1	303	0.1
Oswal and Rathnam ⁹⁸	1984	С	1	303	0.1
Wei and Rowley ⁹⁹	1984	С	1	298	0.1
Durov and Artykov ¹⁰⁰	1985		4	283-313	0.1
Lavrent'ev and Yakovlev ¹⁰¹	1985		21	293–547	0.1
Oswald and Rao ¹⁰²	1985		1	303	0.1
Awwad and Salman ¹⁰³	1986	С	1	298	0.1
Fischer and Weiss ¹⁰⁴	1986		8	291-341	0.1
Nath and Tripathi ¹⁰⁵	1986		1	303	0.1
Manjeshwar and Aminabhavi ¹⁰⁶	1987	С	2	298	0.1
Rathnam ¹⁰⁷	1987		1	313	0.1
Subha and Brahmaji Rao ¹⁰⁸	1987		1	308	0.1
Yadava and Yadava ¹⁰⁹	1987		1	307	0.1
Chandrasekhar <i>et al.</i> ¹¹⁰	1988		1	303	0.1
Manjeshwar and Aminabhavi ¹¹¹	1988	С	1	298–318	0.1
Rattan <i>et al.</i> ¹¹²	1988		1	303	0.1
Vogel <i>et al.</i> ¹¹⁵	1988	OB	97	298-632	0.0-0.3
Berstad ¹¹⁴	1989	OB	32	295-441	1.0-40
Grachev <i>et al.</i> ¹¹⁵	1989		306	280-673	0.1-60
Knapstad <i>et al.</i>	1989	OB	13	289-334	0.1
Ramanjaneyulu <i>et al.</i> ¹¹⁷	1989		1	303	0.1
Ruiz <i>et al.</i> ¹¹⁰	1989	-	3	303–323	0.1
Chevalier <i>et al.</i> ¹¹⁹	1990	С	1	298	0.1
Joshi et al. 120	1990	С	4	298–313	0.1
Joshi <i>et al.</i> 177	1990	С	4	298–313	0.1
Mazurkiewicz and Tomasik ¹²²	1990		1	298	0.1
Sekar <i>et al.</i> 124	1990	С	l	303	0.1
Schumpe and Lunring ¹²	1990	U	1	293	0.1
Subha et al.	1990	C	1	308	0.1
Krishnaiah and Viswanath	1991	C	2	298-308	0.1
Distance Pater	1991	C	1	303	0.1
Paparoannou <i>et al.</i> Paparoantosiou and Ziagaa ¹²⁹	1991	C	1	298	0.1
Papallastasiou and Ziogas Remelvanth et at^{130}	1991	C	5	200-300	0.1
Tanakanun et al.	1991	TC	1	298	0.1
Ali and Pahman ¹³²	1991	IC	20	298-348	0.1-100
Noth and Chaudhary ¹³³	1992	C	1	290	0.1
Nath and Saini ¹³⁴	1992	C	1	303	0.1
Oswal and Patel ¹³⁵	1002	C	1	303	0.1
Oswal and Phalak ¹³⁶	1992	C	1	303	0.1
Blasco <i>et al</i> ¹³⁷	1003	C	1	298	0.1
Matsuo and Makita ¹³⁸	1993	C	18	298_313	0.1_48
Pandev et al ¹³⁹	1993	e	1	298	0.1
Raikar <i>et al</i> 140	1993		3	298-308	0.1
De Lorenzi <i>et al</i> ¹⁴¹	1994	С	1	298	0.1
Rajendran ¹⁴²	1994	e	1	303	0.1
Aminabhavi and Gopalakrishna ¹⁴³	1995	С	4	298–313	0.1
Artigas et al 144	1995	-	1	303	0.1
Haase and Tillmann ¹⁴⁵	1995		1	298	0.1
Neumann ¹⁴⁶	1995		15	283-353	0.1
Petrino <i>et al.</i> ¹⁴⁷	1995	С	1	298	0.1
Yavada ¹⁴⁸	1995	-	1	293	0.1
Aminabhavi et al. ¹⁴⁹	1996	С	3	298-308	0.1
Easteal ¹⁵⁰	1996	Č	1	298	0.1
Latifi ¹⁵¹	1996		1	298	0.1
Padua et al. ¹⁵²	1996	VW	25	298-348	0.1–38

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$\ensuremath{\mathsf{T}\mathsf{ABLE}}$. —Continued

Authors	Year publ.	Technique employed ^a	No. of data	Temperature range (K)	Pressure range (MPa)
Pintos et al. ¹⁵³	1997		3	288-308	0.1
Abramovic and Klofutar ¹⁵⁴	1998		6	293-323	0.1
Aminabhavi and Banerjee ¹⁵⁵	1998	С	3	298-308	0.1
Ihmels ¹⁵⁶	1998	С	11	293-343	0.1
Nath and Mishra ¹⁵⁷	1998	С	1	303	0.1
Gascon et al. ¹⁵⁸	1999	С	2	298-313	0.1
Guzman et al. ¹⁵⁹	1999	С	3	283-313	0.1
Dumitrescu and Vilcu ¹⁶⁰	2000	С	5	288-308	0.1
Gascon <i>et al.</i> ¹⁶¹	2000	С	2	298-313	0.1
Gomez-Diaz et al. ¹⁶²	2001	С	6	298-323	0.1
Palaniappan ¹⁶³	2001		3	303-313	0.1
George and Sastry ¹⁶⁴	2003	С	2	298-308	0.1
Rodriguez et al. ¹⁶⁵	2003	RB	4	293-313	0.1
Ali et al. ¹⁶⁶	2004	C	1	308	0.1
Haro <i>et al.</i> ¹⁶⁷	2004	Č	2	298-313	0.1
Ma et al 168	2004	VW	16	323-413	0.1-6.4
Oswal <i>et al</i> 169	2004	C	2	303-313	0.1
Yang et al. ¹⁷⁰	2004	Č	5	298-333	0.1
Iloukhani and Rezaei-Sameti ¹⁷¹	2005	C	1	298	0.1
Knothe and Steidlev ¹⁷²	2005	Č	1	313	0.1
Oswal <i>et al</i> 173	2005	C	1	303	0.1
Oswal et al. ¹⁷⁴	2005	C	1	298	0.1
Singh ¹⁷⁵	2005	C	1	298	0.1
Singh at al ¹⁷⁶	2005	C	3	208 308	0.1
Tripathi ¹⁷⁷	2005	C	1	298-308	0.1
Al Chorwi <i>et al</i> 178	2005	C	1	200 212	0.1
Al Olici wi et al. Conzoloz et al 1^{79}	2000	C	2	202 202	0.1
Surash Baddy at al ¹⁸⁰	2000	C	3	295-505	0.1
Sinche and Pigwag ¹⁸¹	2000	C	1	208	0.1
Conzeloz et al ¹⁸²	2007	C	1	298	0.1
Mohra and Danahali ¹⁸³	2007	C	3	295-505	0.1
Vong et al. ¹⁸⁴	2007	G	1	298	0.1
A www.d and Aby Daches ¹⁸⁵	2007	C	3	298-333	0.1
Awwad and Abu-Daabes	2008	C	4	298-323	0.1
Failg $et at$.	2008	C	5	200-508	0.1
Charles and Oswal	2008	C	1	303	0.1
Shukia et al.	2008	ť	1	298	0.1
Check of 190	2008	C	1	308	0.1
Gnael <i>et al.</i>	2009		1	303	0.1
Hernandez-Galvan <i>et al.</i>	2009	RB	40	313-393	0.7-60
Rajagopal <i>et al.</i>	2009	OB	82	298-413	0.1-62
Roy et al.	2009		I z	298	0.1
Silva et al.	2009	RB	5	283-323	0.1
All et al. 196	2010	DD	6	293-318	0.1
Horstmann <i>et al.</i> 197	2010	кв	7	283-343	0.1
Kathnam <i>et al.</i> ¹⁹⁸	2010	C	3	303-313	0.1
Yadava and Yadava ¹⁹⁰	2010	С	l	308	0.1
Zheng <i>et al.</i> ²⁰⁰	2010		3	293-303	0.1
Pradhan and Roy ²⁰⁰	2011	-	l	298	0.1
El-Hadad <i>et al.</i> ²⁰¹	2012	C	2	293–298	0.1
Hamzehlouia and Asfour ²⁰²	2012	С	2	308–313	0.1
Vanathi <i>et al.</i> ²⁰⁴	2013	~	3	303-313	0.1
Sastry <i>et al.</i> ²⁰⁴	2013	С	2	298-303	0.1
$L1 \ et \ al.^{203}$	2013	RB	6	293–318	0.1
Tables of collected data					
Stephan and Lucas ²⁰⁶	1979		183	290-520	0 1–50
Liessmann <i>et al.</i> ²⁰⁷	1995		11	273-473	0.1
NIST/TRC database ²⁰⁸	2003		51	278–553	0.1

^aC - capillary; FB - falling body; OB - oscillating body; RB - rolling body; TC - torsional crystal; VW - vibrating wire.

FIG. 13. Percentage deviations $[100(\eta_{exp} - \eta_{corr})/\eta_{exp}]$ of selected secondary experimental viscosity data in the liquid region from the calculated values using Eqs. (1)–(3), (5), and (6). (•) Bridgman;²³ (•) Collings and McLaughlin;⁶⁴ (*) Ishihara *et al.*;⁷⁰ (•) Jonas *et al.*;⁸³ (+) Ma *et al.*¹⁶⁸

ρ / (mol·l⁻¹)

8.5

9.0

9.5

10.0

7.5

8.0

FIG. 14. Percentage deviations $[100(\eta_{exp} - \eta_{corr})/\eta_{exp}]$ of selected secondary experimental viscosity data in the liquid region from the calculated values using Eqs. (1)–(3), (5), and (6). (**n**) Belinski and Khodzhaev;⁴⁹ (×) Guseinov *et al.*;⁶⁹ (\blacklozenge) Hernandez *et al.*;¹¹¹ (\blacktriangle) Rajagapol *et al.*¹¹²

FIG. 15. Percentage deviations $[100(\eta_{exp} - \eta_{corr})/\eta_{exp}]$ of the secondary experimental viscosity data of Grachev *et al.*¹¹⁵ in the liquid region from the calculated values using Eqs. (1)–(3), (5), and (6) with selected isotherms highlighted. (**1**) 373 K; (**A**) 398 K; (x) 448 K; (**•**) other isotherms.

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