Reference Correlation of the Viscosity of *n*-Hexadecane from the Triple Point to 673 K and up to 425 MPa

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Abstract

A new correlation for the viscosity of *n*-hexadecane 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 673 K at pressures up to 425 MPa. The overall uncertainty of the proposed correlation, estimated as the combined expanded uncertainty with a coverage factor of 2, varies from 1% for the viscosity at atmospheric pressure to 10% for the viscosity of the vapor phase at low temperatures. Tables of the viscosity, generated by the relevant equations, at selected temperatures and pressures, and along the saturation line, are provided.

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1. Introduction

There is a growing industrial need to establish accurate and reliable reference values of viscosity of pure fluids.[1,2] Not only are such values useful in their own right, but they are also a precursor for developing models for the predicting viscosity of mixtures. This is especially important for naturally occurring mixtures that contain a myriad of species, where the presence of long-chain species increases viscosity significantly. For a number of industrially important fluids (simple fluids,[3-6] water,[7] normal alkanes,[8-17] cyclic and aromatic hydrocarbons[18-24] and refrigerants[25-27]) there exist accurate and reliable correlations of viscosity that cover a wide range of temperatures and pressures, with well-defined estimates of uncertainty. The aim of the present study is to critically assess the viscosity data available in the literature and provide a correlation for the viscosity of *n*-hexadecane that is valid over a wide range of temperature and pressure, covering the vapor, liquid, and supercritical fluid states. Thus, expanding the set of viscosity correlations that are available for *n*-alkanes that currently encompass methane to *n*-butane[8-11] and *n*-hexane to *n*-dodecane.[12-17] The work is continuation of the program to develop representations of the viscosity and thermal conductivity of pure fluids that is carried out under the auspices of the International Association for Transport Properties (IATP) a Subcommittee of the International Union of Pure and Applied Chemistry (IUPAC).

n-Hexadecane (*n*-$C_{16}H_{34}$) is a normal, paraffinic alkane that is naturally found in oil and is an important constituent of aviation and diesel fuel. It is sometimes referred by its old name, cetane, and is assigned a cetane number of 100, thus serving as a reference fuel for measuring the quality of diesel. *n*-Hexadecane has been shown to exhibit anti-inflammatory behaviour leading to its increasing medical and microbiological applications. The thermodynamic properties of *n*-hexadecane are well catered for, by an up-to-date Equation of State (EoS),[28] while at present, no correlation of viscosity is available.

2. Experimental Viscosity Data

Appendix A summarizes, to the best of our knowledge, the experimental measurements of the viscosity of *n*-hexadecane reported in the literature,[29-107] 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 *n*-hexadecane were reported in 77 papers[29-105] and 2 unpublished sources[106,107] resulting in 738 data points. Appendix A also contains a list of reference works[108-111] that report recommended tabulated values of the viscosity of *n*-hexadecane. Following the recommendation adopted by the IATP, a critical assessment of the experimental data was performed to classify the data as primary and secondary, using well-established criteria[112] that have been widely disseminated.[3-27] Based on these criteria, 10 datasets were considered primary viscosity data. Table 1 summarizes the primary data[30,51,52,61,63,69,77,93,97,106] detailing the temperature and pressure ranges, the authors′ uncertainty attributed to the measurements, claimed purity of the sample, and the technique employed to perform the measurements. The choice of primary data is discussed in more detail in section 3 that also provides a comparison of the data by different workers.

TABLE 1. Primary data used in developing the viscosity correlation of *n*-hexadecane

Authors	Year publ.	Technique employed[a]	Purity (%)	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Nederbragt and Boelhouwer[30]	1947	C	--	1.5	5	298-518	0.1
Dymond *et al.*[51]	1980	FB	99.6	2	28	298-373	0.1-425
Dymond and Young[52]	1980	C	99.6	1.4	10	298-393	P_{sat}
Wakefield and Marsh[61]	1987	C	99-99.9	0.5	3	318-338	0.1
Wakefield[63]	1988	C	99	0.5	2	303-308	0.1
Tanaka *et al.*[69]	1991	TC	98	2	16	298-348	0.1-151
Wu *et al.*[77]	1998	C	99	0.1	4	293-313	0.1
Ciotta[93]	2010	VW	99.8	2	53[b]	298-474	1-103
Baled *et al.*[97]	2014	RB	99	2	17[c]	304-326	3-227
Sanchez-Vicente[106]	2018	VW	99.6	3	36[d]	323-673	1-4

[a] C, capillary; FB, falling-body; TC, torsional crystal; VW, vibrating wire; RB, rolling body.
[b] One data point at 323.21 K and 102.81 MPa was excluded from the primary data set.
[c] Data above 429 K were excluded from the primary data set.
[d] Data at 723 K were excluded from the primary data set.

Figure 1 shows the temperature and pressure range of the measurements outlined in Appendix A with primary and secondary data distinguished. The primary data of Dymond and Young[52] along the saturation line is not shown, as the temperature range covered corresponds to pressures lower than 0.01 MPa. The primary data cover a wide range of temperatures and pressures of interest. The data is extensive in the liquid phase, but in the vapor phase only two data sets are available.

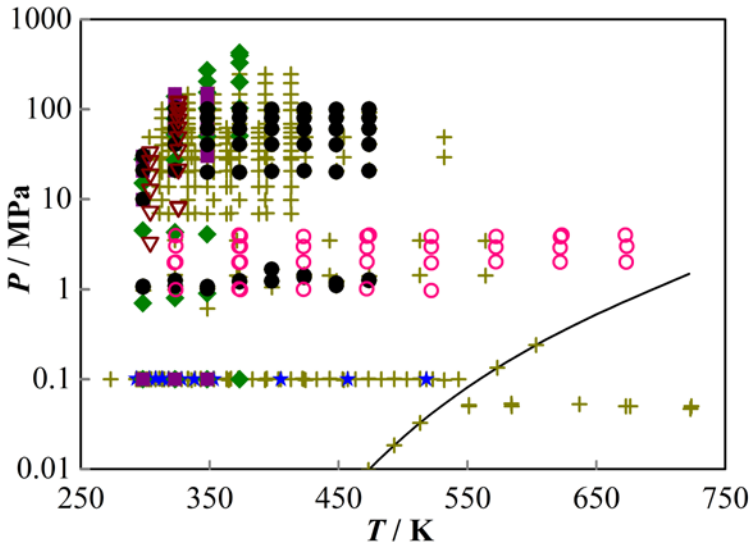

FIG. 1. Distribution of the available experimental viscosity data of *n*-hexadecane. Primary data: (◆) Dymond *et al.*;[51] (■) Tanaka *et al.*;[69] (●) Ciotta;[93] (▽) Baled *et al.*;[97] (○) Sanchez-Vicente *et al.*;[106] (★)data at 0.1 MPa;[30,61,63,77] (+) Secondary data; The solid line shows the vapor-liquid saturation boundary.

In order to convert Temperature-Pressure (T, P) pairs, at which most measured viscosities are quoted, into Temperature-Density (T, ρ) pairs, we have used a recent EoS developed by Romeo and Lemmon[28] that covers the thermodynamic space from the triple point to 800 K, and pressures up to 50 MPa. The available density data

are represented on average with deviations within 0.3%, with a maximum deviations not exceeding 2%.[28] The uncertainties in the saturated liquid density at temperatures up to 490 K, generated by this EoS, are lower than 0.1%.[28] As some of the primary viscosity data has been measured at pressures higher than 50 MPa, we made use of the Romeo and Lemmon[28] EoS to estimate the densities for pressures as high as 425 MPa, like we did for *o*-xylene.[23] The accuracy of the extrapolation was checked by comparison to Dymond *et al.*,[51] Dymond and Young[52] and Wu *et al.*[113] density data. Figure 2 illustrates that the agreement between extrapolated and measured density data is within 0.5%, which is within their mutual uncertainty.

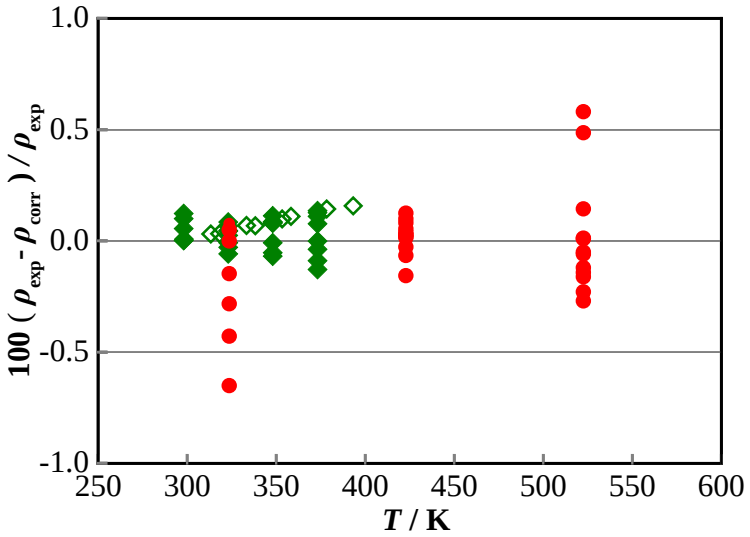

FIG. 2. Percentage deviations [100(ρ_{exp}–ρ_{corr})/ ρ_{exp}] of the experimental density data from the calculated values. (◆) Dymond *et al.*;[51] (◇) Dymond and Young;[52] (●) Wu *et al.*[113]

3. Methodology and Analysis

It is customary[114] in developing correlations of transport properties to take advantage of theoretical guidance to 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 and by making use of current theoretical developments, in conjunction with the available experimental data, 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.[3-27]

3.1. The zero-density and initial-density terms

There are no reliable measurements of viscosity of *n*-hexadecane in the vapor phase. The low vapor pressure at room temperature, of the order of a few tenths of a Pascal, the experimental difficulties of obtaining accurate values at high temperature and the lack of industrial applications are some of the main reasons for the paucity of data. Instead of relying on scarce secondary viscosity data, we take advantage of the recent developments in the modelling to estimate the values of the zero-density and initial-density terms.

Riesco and Vesovic[115] have recently developed a semi-theoretical way of estimating the zero-density viscosity of long-chain *n*-alkanes. The model is based on representing the intermolecular interactions by means of an effective Lennard-Jones 12-6 (LJ) potential. The LJ length scaling parameter, σ, is obtained by taking advantage of the relationship between the average end-to-end length of the alkane molecule and the number of carbon atoms. Constraining the length scaling parameter in this fashion allows for a unique determination of the energy scaling parameter, ε, for each *n*-alkane, by making use of the available zero-density viscosity measurements. The resulting energy scaling parameters, when plotted as a function of carbon number, exhibit a monotonic increase that reaches a plateau for long-chain alkanes. Hence the model can be used to allow viscosity prediction for *n*-alkanes for which no measurements are available. The resulting model predicts the zero-density viscosity within 2.4%, based on a comparison with reliable and accurate experimental viscosity data.[115] We have used Riesco and Vesovic[115] model to predict the zero-density viscosity of *n*-hexadecane in the temperature range (290 to 750 K). For ease of use we have fitted the calculated values to a standard and practical engineering representation,[22-24]

$$\eta_0(T) = 0.32137\frac{\sqrt{T}}{S_\eta} \tag{2}$$

where $\eta_0(T)$ is given in units of μPa·s, T is the temperature in Kelvin and S_η is the effective collision cross-section in nm^2 given by

$$\ln(S_\eta/\mathrm{nm}^2) = A_0 + \frac{B_0}{T} + \frac{C_0}{T^2} \tag{3}$$

where A_0, B_0 and C_0 are the adjustable parameters and take the value of A_0 = -0.6131, B_0 = 414.4 K and C_0 = -39759 K^2.

At subcritical temperature, in the vapor phase, the viscosity below a certain temperature initially decreases with increasing density before increasing at higher densities.[114,116,117] It is customary to express the initial-density coefficient, η_1, in terms of the second viscosity virial coefficient, B_η, by means of [114]

$$\eta_1(T) = B_\eta(T)\eta_0(T) \tag{4}$$

A number of workers have proposed functional forms for the second viscosity virial coefficient that are based on either LJ (12-6) potential[116-118] or on universal correlations.[11,114] In this work we opted for universal

correlations and used the scaling parameters σ and ε obtained from aforementioned Riesco and Vesovic model.[115] Although the expressions for the second virial coefficients were developed using data for much smaller molecules, the use of scaling parameters of Riesco and Vesovic model that successfully employs effective spherical potentials to mimic the interaction between long chains, give us some justification in estimating the initial density term by means of the second viscosity virial coefficient.

As the expressions for the second viscosity virial coefficient can be cumbersome, we have fitted the calculated values of the initial-density term a simple functional form, that we have successfully employed previously, [22-24]

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

where ρ is the molar density in units of mol l^{-1} and A_1, B_1 and C_1 are the adjustable parameters, with the values of A_1 = 7.4345 μPa s mol^{-1} L, B_1 = -739.4 μPa s K mol^{-1} L and C_1 = -2,255,587 μPa s K^2 mol^{-1} L.

The only available data in open literature, for the viscosity of *n*-hexadecane in the vapor phase, have been measured by Zhdanov and Lyusternik.[44,46] The first set of measurements[44] were carried out in the capillary viscometer operating in the absolute mode at pressure of 0.5 bar in the temperature range (551 to 723) K. The purity of the sample was indicated to be in the range of 98-99%.[44] The second set of measurements were performed in a similar, if not the same capillary viscometer in the same temperature range at the same pressure, but the purity of the sample was in the range 99.4-99.8%. The later measurements[46] are on average 1.0-3.5% higher than the former,[44] the agreement improving with temperature. Lyusternik and Zhdanov also produced tables of recommended data.[46] Although the recommended values are within 2% of the measured data at lower temperatures, at temperatures higher than 636 K, the deviations of 10-13% are observed.

Figure 3, illustrates the deviations of the data of Zhdanov and Lyusternik[44,46] from the vapor-phase viscosity correlation proposed in this work. It is clear that the developed *n*-hexadecane correlation for $\eta_0(T) + \eta_1(T)\rho$ reproduces the recommended tabulated data[46] within 2.5%.

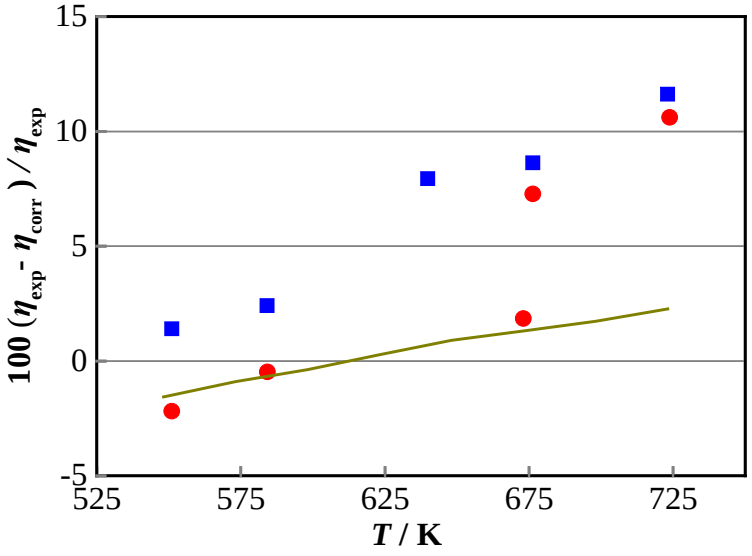

FIG. 3. Percentage deviations [100(η_{exp}–η_{corr})/η_{exp}] of the available experimental and recommended tabulated data in the vapor phase. (●) Zhdanov and Lyusternik;[44] (■) Lyusternik, and Zhdanov;[46] (—) Lyusternik, and Zhdanov (tabulated data).[46]

Based on the agreement with the recommended tabulated data of Zhdanov and Lyusternik[44,46] and uncertainty associated with Riesco and Vesovic model[115] and initial-density model, we ascribe uncertainty of 5% to the viscosity correlation in the vapor phase, above 500 K. At lower temperatures, because of uncertainty in estimating the initial-density term, we conservatively estimate the uncertainty to be 10%.

3.2. The critical enhancement and the residual viscosity terms

In the vicinity of the critical point the viscosity of the pure fluid exhibits an enhancement[2] that is significant only in a relatively narrow window in temperature and density round the critical point.[3,9] Based on the previous studies,[13-27] the viscosity critical enhancement of *n*-hexadecane is taken as zero. The total lack of industrial applications of *n*-hexadecane near its critical temperature and the lack of the experimental viscosity data in the neighborhood of the critical point further supports this choice.

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. Twelve sets of authors[37,45,51,53,57,59,69,92,93,97,100,106] have measured the viscosity of *n*-hexadecane at a wide range of temperatures and at pressures higher than atmospheric, as illustrated in Fig. 1 and summarized in Appendix A. Based on the analysis of the viscometers employed and agreement with measurements on other fluids we have designated as primary five sets of data, as illustrated in Table 1. The measurements cover an extended temperature range (293 to 673 K) at pressures up to 425 MPa. The 3 data points measured by Sanchez-Vicente[106] at 723 K were excluded from the primary data set, as according to the authors[106] hexadecane underwent partial thermal decomposition at this temperature. As shown in Fig. 1, the temperature and pressure range of primary data encompasses that of the secondary data. A variety of techniques have been used to measure designated primary data, including falling body, torsional crystal, vibrating wire and capillary viscometer with the uncertainty claimed by authors of 0.1-3%, see Table 1. Comparison of data along the isotherms, at which more than a single set of measurements has been made, indicates that all the data below 373 K are mutually consistent, to within 2.5%. Figure 4 illustrates the comparison at 323 K.

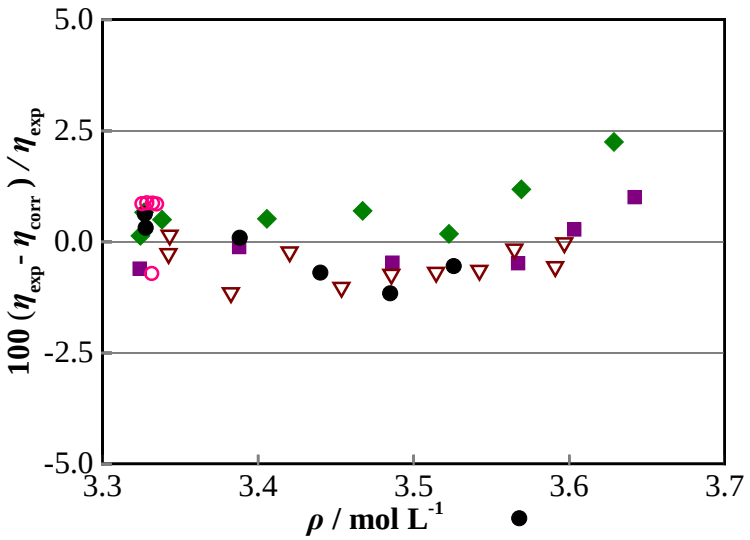

FIG. 4. Comparison of the experimental liquid viscosity data at 323 K. (◆) Dymond *et al.*;[51] (■) Tanaka *et al.*;[69] (●) Ciotta;[93] (▽) Baled *et al.* (326 K);[97] (○) Sanchez-Vicente *et al.*[106]

At higher temperatures we observe systematic deviations, between the data of Baled *et al.*[97] , Ciotta[93] and Sanchez-Vicente *et al.*[106], as illustrated in Figs. 5 and 6. We observe that at both temperatures Baled *et al.*[97] data is 2 to 7% lower than the data measured by Ciotta[93] and Sanchez-Vicente *et al.*[106] in the density region where the data overlap. It was not possible to reconcile the two sets of data nor was it possible to use only Baled *et al.*[97] data at high temperatures and obtain the viscosity correlation that would recapture lower temperature data within its claimed uncertainty. Thus, we have eliminated Baled *et al.*[97] data measured at the two highest isotherms (423 and 534) K from the primary data set, as indicated in Table 1.

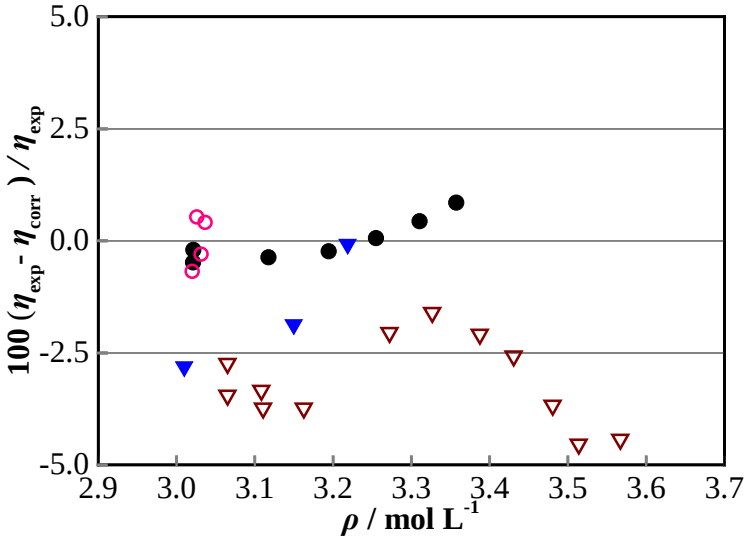

FIG. 5. Comparison of the experimental liquid viscosity data at 423 K. (▼) Rastorguev and Keramidi (secondary data at 424K);[45] (●) Ciotta;[93] (▽) Baled *et al.* (429 K);[97] (○) Sanchez-Vicente *et al.*[106]

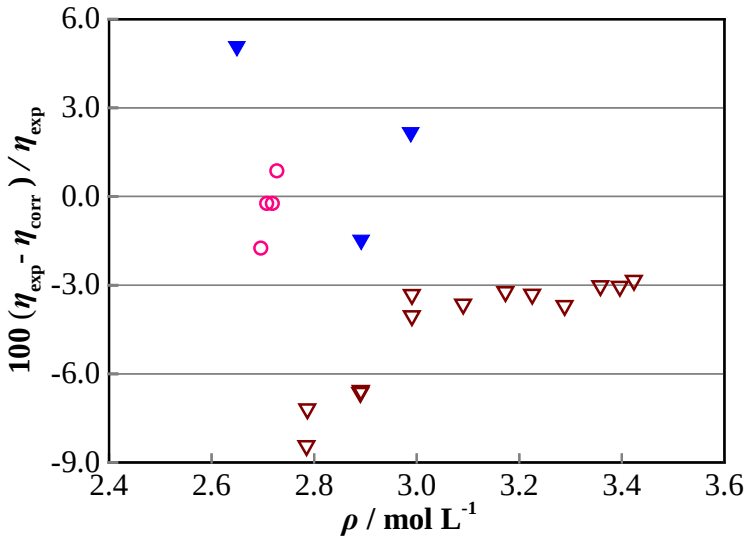

FIG. 6. Comparison of the experimental liquid viscosity data at 534 K. (▼) Rastorguev and Keramidi (secondary data at 532K);[45] (▽) Baled *et al.*;[97] (○) Sanchez-Vicente *et al.* (522 K)[106]

The primary data set also contains one set of data along the liquid saturation line[52] and four sets of viscosity measurements[30,61,63,77] in the liquid phase at atmospheric pressure covering the temperature range (293-518) K. The measurement along the saturation line was performed by Dymond and Young[52] in a capillary viscometer of proven capabilities from 298 to 393 K. Two of the capillary viscometers used to measure the primary *n*-hexadecane data solely at atmospheric pressure[61,63,77] were also used to measure accurately the viscosity of other fluids. Based on the comparison obtained for other fluids and the purity of the samples employed we estimate that the uncertainty of these data are nearer to 1-1.5% rather than the 0.1-0.5% , as quoted by the authors. In order to extend the temperature range of primary data at atmospheric pressure, to temperatures above 373 K, we have also included, in the primary data set, the measurements made by Nederbragt and Boelhouwer[30] performed in a capillary viscometer. The choice was guided by our analyses of the described workings of the employed viscometer and by good agreement of their data with other primary sources in the region of temperature overlap. Numerous measurements of the viscosity of *n*-hexadecane at atmospheric pressure have been performed at a single temperature, in the vicinity of the room temperature. Although quite a few authors claim the uncertainty in the region of 0.1-0.2%, the comparative analysis of the viscosity data, purity of the sample employed and measurements on other fluids do not sustain this claim. For instance, five measurements of viscosity by different labs[73,74,77-79] at 298 K differ by 2.2% which is an order of magnitude larger than the claimed uncertainty. As it was not possible to identify a high-precision measurement, no viscosity measurements consisting of a single datum was taken as primary.

In summary 174 data points covering the temperature range (293 to 673) K and pressures up to 425 MPa measured in nine different viscometers were used as the primary data for the development of the residual viscosity contribution. 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). In line with previous work, [18, 21-24] 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, except at low

densities where the decreasing initial-density dependence extends partially into the two-phase region. 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,

$$\Delta\eta(\rho_r, T_r) = (\rho_r^{2/3}\, T_r^{1/2}) f(\rho_r, T_r) \quad (6)$$

where function $f(\rho_r, T_r)$ is given by,

$$f(\rho_r, T_r) = (D_0 + E_0/T_r^{k_0})\rho_r^{n_0} + D_1\rho_r^{n_1} + E_2\rho_r^{n_2}/T_r^{k_2} + (D_3\rho_r + E_3 T_r)\rho_r^{n_3} + D_4\rho_r^{n_4} \quad (7)$$

Following the development of other correlations,[18,21-24] 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 1stOpt (First Optimization) software for statistical computing[119] to fit primary data to Eqs. (6) and (7). The uncertainties quoted in Table 1 were used to determine relative weights for all the primary data. The optimal coefficients D_i, E_i, k_i and n_i are shown in Table 2, while the critical temperature T_c (722.1 K) and critical density ρ_c (1.000 mol L^{-1}) were obtained from Ref. 28.

TABLE 2. Coefficients for the representation of the residual viscosity, Eqs. (6) and (7)

i	D_i	n_i	E_i	k_i
0	0.000692129	9	0.00645721	1.3
1	-0.000305913	11	--	--
2	--	24	1.27×10^{-12}	5.5
3	21.851	0.6	-30.2533	--
4	21.0853	0	--	--

Figures 7 and 8 illustrate the percentage deviation of the primary viscosity data from the developed viscosity correlation, Eqs. (1)-(7).

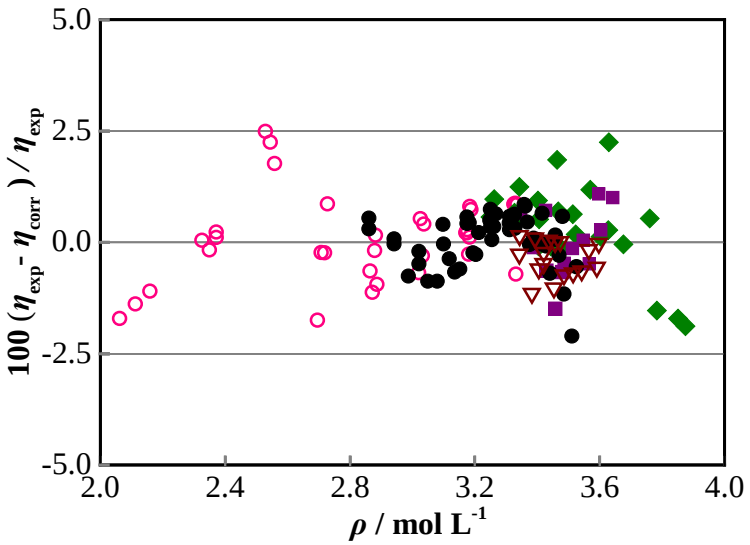

FIG. 7. Percentage deviations $[100(\eta_{exp}-\eta_{corr})/\eta_{exp}]$ of the primary experimental viscosity data in the liquid region for pressures higher than atmospheric from the values calculated by Eqs. (1) - (7). Dymond *et al.*;[51] (■) Tanaka *et al.*;[69] (●) Ciotta;[93] (▽) Baled *et al.*;[97] (○) Sanchez-Vicente *et al.*[106]

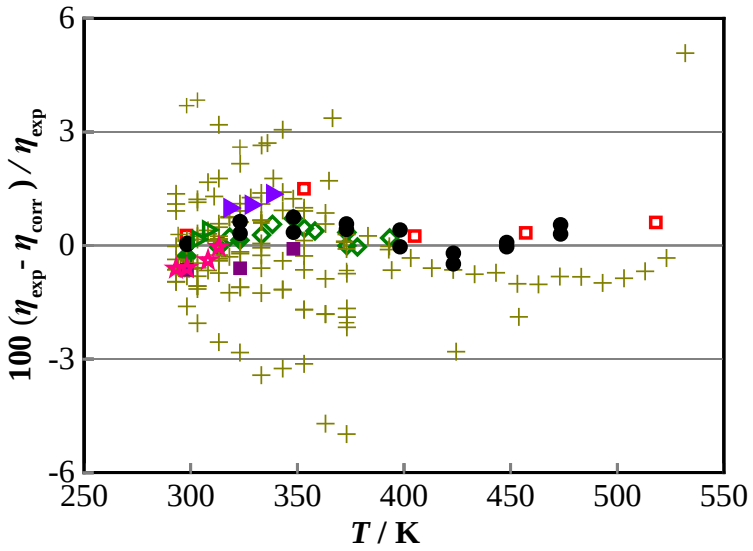

FIG. 8. Percentage deviations $[100(\eta_{exp}-\eta_{corr})/\eta_{exp}]$ of the experimental viscosity data measured at 0.1 MPa and along the saturation line from the calculated values using Eqs. (1) - (7). (□) Nederbragt and Boelhouwer;[30] (◆) Dymond *et al.*;[51] (◇) Dymond and Young;[52] (►) Wakefield and Marsh;[61] (▹) Wakefield;[63] (■) Tanaka *et al.*;[69] (☆) Wu *et al.*;[77](●) Ciotta;[93] (+) Secondary data.[29, 32,36,37,41,45 ,50,53,57,60,81,82,94,95,96,98,99,107]

Figure 7 illustrates the agreement with the experimental data in the liquid region for pressures higher than atmospheric. We observe that most of the data are reproduced within 2.5% which is within the overall claimed uncertainty of the data. Figure 8 illustrates the agreement with the experimental data in the liquid region for atmospheric pressures. We observe that most of the data are reproduced within 1.0%, with the exception of a single datum measured by Nederbragt and Boelhouwer[30] at 353 K and Wakefield and Marsh[61] data at the highest temperature, 338 K. The former is most likely the outlier, while Wakefield and Marsh data[61,63] seems to exhibit a stronger temperature dependence than the proposed correlation. We have also included in Fig. 8 a selection of

data classified as secondary where viscosity was reported at more than a single temperature. Although as expected the deviations are on average larger than those of primary data, there are no systematic temperature trends, indicating that the current correlation correctly depicts the temperature dependence.

As a further test of the validity of our choice of primary data set at high temperature and pressures higher than, atmospheric, we have tested the developed correlation against a selected set of secondary data that consists of measurements performed by Rastoguev and Keramidi[45] and Mathews [59], that cover the temperature range (323 to 564) K at pressures up to 49 MPa. Both sets of authors performed the measurements in a capillary viscometer which, providing a full working equation is employed, is capable of producing primary data. As illustrated in Fig. 9 at temperatures below 400 K the data are predicted to within 2.5%, with no systematic trends. At higher temperatures we observe that the data of Rastoguev and Keramidi[45] and Mathews[59] concur with the proposed correlation. This is further illustrated in Figs. 5 and 6 where Rastoguev and Keramidi[45] data along 424 K and 532 K isotherm is plotted as a function of density. The other sets of secondary data,[53,92] that cover the temperature range up to 523 K, could not be used in this manner as they are not consistent with the primary data set at lower temperatures. Thus, the comparison with a selected secondary data provides further, albeit circumstantial evidence, for the choice of primary viscosity data at high temperature.

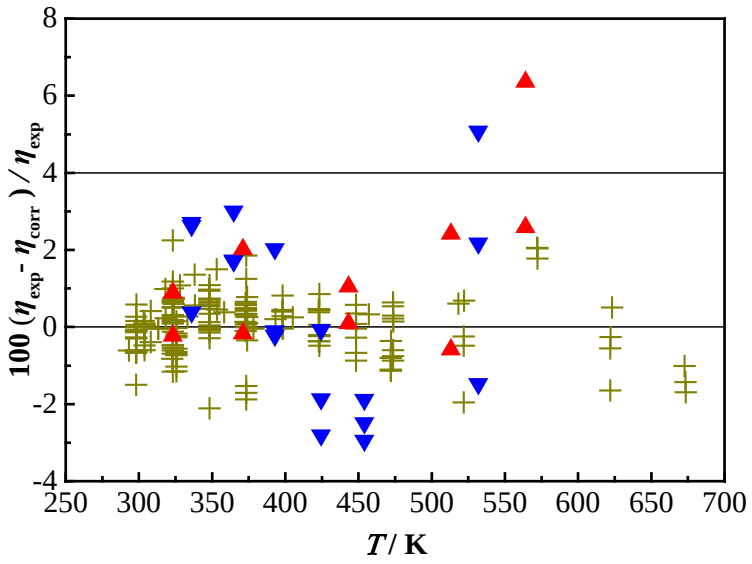

FIG. 9. Percentage deviations [$100(\eta_{exp}-\eta_{corr})/\eta_{exp}$] of the primary experimental viscosity data and the selected secondary data[45,59] from the values calculated by Eqs. (1) - (7). (+) primary data;[30,51,52,61,63,69,77,93,97,106] (▼) Rastorguev and Keramidi;[45] (▲) Mathews *et al.*[59]

Table 3 summarizes the agreement between the primary experimental data and the proposed viscosity correlation for *n*-hexadecane in the liquid, dense vapor and supercritical regions. The correlation recaptures the entire set of primary data with an average absolute deviation (AAD) of 0.56 %, bias of 0.08% and maximum deviation of 2.49%.

TABLE 3. Evaluation of the *n*-hexadecane viscosity correlation against the primary experimental data

Authors	Year publ.	AAD[a] (%)	Bias[b] (%)	MD[c] (%)
Nederbragt and Boelhouwer[30]	1947	0.59	0.59	1.50
Dymond *et al.*[51]	1980	0.72	0.31	2.25
Dymond and Young[52]	1980	0.25	0.17	0.56
Wakefield and Marsh[61]	1987	1.14	1.14	1.36
Wakefield[63]	1988	0.29	0.29	0.41
Tanaka *et al.*[69]	1991	0.57	-0.10	-1.50
Wu *et al.*[77]	1998	0.41	-0.41	-0.61
Ciotta[93]	2010	0.44	0.04	-2.11
Baled *et al.*[97]	2014	0.40	-0.37	-1.15
Sanchez-Vicente *et al.*[106]	2016	0.74	0.10	2.49
Entire primary data set		0.56	0.08	2.49

[a]AAD, Average Absolute Deviation = $^{100}/_{N}\sum|(\eta_{\mathrm{exp}}-\eta_{\mathrm{corr}})/\eta_{\mathrm{exp}}|$.
[b]Bias = $^{100}/_{N}\sum(\eta_{\mathrm{exp}}-\eta_{\mathrm{corr}})/\eta_{\mathrm{exp}}$.
[c]MD, Maximum deviation.

4. Overall Viscosity Correlation

The viscosity correlation of *n*-hexadecane as a function of temperature and density is represented by Eqs. (1)-(7) with the coefficients given in Table 2. The correlation is valid in an extended temperature range (293 to 673 K) and pressure up to 425 MPa. The upper pressure limit of the correlation is primarily determined by the availability of experimental data. In the high temperature range (500 to 673 K) the upper pressure limit is 4 MPa, increasing to 100 MPa, at the intermediate temperature range (400 to 500 K) and reaching its maximum value of 425 MPa in the temperature range (293 to 400 K). The lack of experimental data, the empirical nature of the correlating equations and the steep increase in the liquid viscosity at very low temperatures makes extrapolation rather uncertain. However, the proposed correlation does not exhibit any unphysical behavior when extrapolated in the vapor phase to temperatures as low as the triple point (291.3 K)[28] nor when extrapolated in the liquid phase to pressures as high as 425 MPa in the temperature range (400 to 673 K).

Figure 10 illustrates the behavior of the viscosity correlation as a function of density along the 300 and 600 K isotherms. We observe a 1800 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 monotonic increase with density except at low densities, of up to 0.7 mol L^{-1}, where the decreasing initial-density dependence extends into the two-phase region. 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 *n*-alkanes[114-115] or as part of the VW model[120-122] to predict the viscosity of mixtures containing *n*-hexadecane.

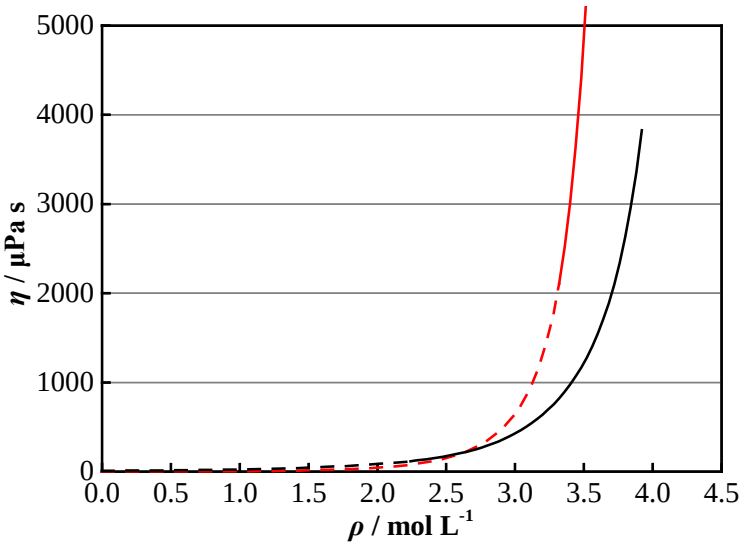

FIG. 10. Viscosity of *n*-hexadecane as a function of density along two isotherms. (red solid line) 300 K, liquid phase; (red dashed line) 300 K, two-phase region; (black solid line) 600 K, liquid phase; and (black dashed line) 600 K, two-phase region.

Based on deviations of the primary data, we have estimated the overall uncertainty of the correlation defined as the combined expanded uncertainty with a coverage factor of 2 as follows: (i) in the liquid phase we estimate the uncertainty to be 2.5%, except at atmospheric pressure or below where we estimate it to be 1% (ii) at other temperatures and pressures in the liquid phase where no primary experimental data are available we conservatively estimate the uncertainty to be 5% (iii) in the vapor phase we estimate the uncertainty to be 5%, above 500 K and 10% below it. Figure 11 summarizes the estimated combined expanded uncertainty with coverage factor of 2 of the proposed viscosity correlation as a function of temperature and pressure. Table 4 contains the recommended values of viscosity of *n*-hexadecane at a selected number of temperatures and pressures which broadly cover the range of the proposed viscosity correlation. Table 5 contains the recommended values of viscosity of *n*-hexadecane along the saturation line.

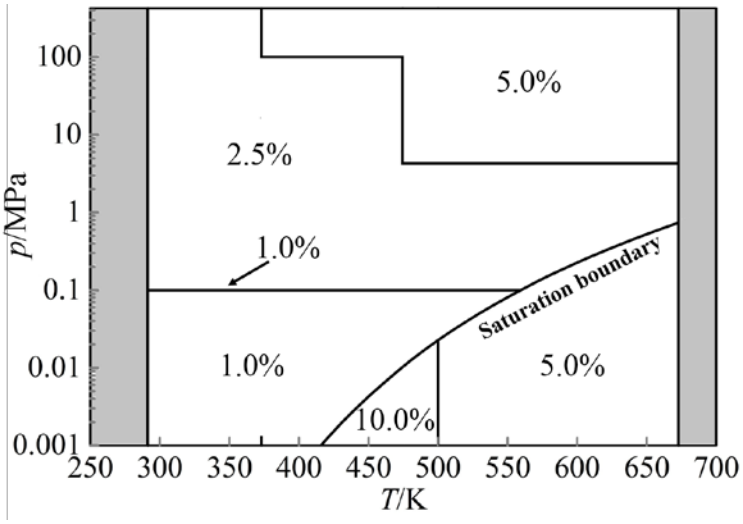

FIG. 11. The extent of the viscosity representation and its estimated uncertainty. No representation is available in the hatched region.

TABLE 4. Recommended viscosity values in μPa s

P	*T*(K)										
MPa	300	320	340	360	380	400	450	500	550	600	650
0	4.02	4.29	4.56	4.84	5.12	5.40	6.10	6.79	7.47	8.13	8.78
0.1	2952.3	1949.7	1389.8	1049.4	825.2	667.5	423.9	286.9	201.3	9.4	10.1
0.5	2967.3	1959.9	1397.1	1054.6	829.4	671.2	426.7	289.3	203.5	145.1	12.2
1	2987.8	1973.3	1405.9	1061.6	834.9	675.8	430.1	292.2	206.3	147.9	103.4
2	3029.4	1999.6	1424.4	1074.9	845.9	684.9	437.0	298.0	211.6	153.3	110.0
4	3111.8	2052.0	1460.6	1102.4	867.4	703.1	450.5	309.3	221.8	163.4	121.1
6	3195.4	2106.1	1497.5	1129.9	889.4	721.3	464.0	320.4	231.6	172.7	130.7
8	3281.7	2160.2	1535.0	1157.6	911.3	739.7	477.3	331.3	241.1	181.5	139.3
10	3367.6	2215.1	1572.5	1185.4	933.2	757.7	490.5	342.0	250.3	189.8	147.3
20	3820.8	2501.2	1767.6	1328.1	1044.4	849.1	554.9	393.2	293.5	227.7	181.9
50	5399.8	3485.5	2425.3	1796.9	1400.0	1133.1	744.7	537.5	410.2	325.8	266.8
100	8874.7	5626.4	3822.0	2761.4	2103.6	1673.1	1075.4	774.3	594.2	475.5	392.1
200	19827.8	12345.6	8131.3	5649.5	4127.5	3153.8	1878.5	1297.9	976.9	774.2	635.0
400	64703.4	40106.7	25883.6	17370.6	12118.3	8775.9	4545.1	2798.8	1945.7	1463.8	1158.9

TABLE 5. Recommended viscosity values along the saturation line

		Vapor		Liquid	
T/K	*P*/MPa	ρ/(mol L^{-1})	η/(μPa s)	ρ/(mol L^{-1})	η/(μPa s)
293.15	1.157×10^{-7}	4.747×10^{-8}	3.92	3.4168	3468.4
323.15	2.396×10^{-6}	8.918×10^{-7}	4.33	3.3237	1837.4
353.15	2.643×10^{-5}	9.003×10^{-6}	4.75	3.2312	1147.7
383.15	1.828×10^{-4}	5.740×10^{-5}	5.18	3.1384	795.6
413.15	8.868×10^{-4}	2.587×10^{-4}	5.64	3.0449	586.5
443.15	3.275×10^{-3}	8.944×10^{-4}	6.15	2.9497	448.4
473.15	9.779×10^{-3}	2.521×10^{-3}	6.72	2.8521	351.2
503.15	2.470×10^{-2}	6.072×10^{-3}	7.37	2.7509	279.9
533.15	5.462×10^{-2}	1.297×10^{-2}	8.14	2.6445	226.0
563.15	1.087×10^{-1}	2.528×10^{-2}	9.03	2.5309	184.0
593.15	1.987×10^{-1}	4.611×10^{-2}	10.05	2.4067	150.3

623.15	3.398×10^{-1}	8.053×10^{-2}	11.19	2.2667	122.2
653.15	5.507×10^{-1}	1.381×10^{-1}	12.44	2.1011	97.4

Figure 12 summarizes the deviations of the selected secondary data,[35,37,48,53,57,70,92,100,101-103,105] consisting of at least four data points, from the current correlation. Although the number of measurements are within the acceptable 5%, there are a number of data sets that exhibit much larger deviations.

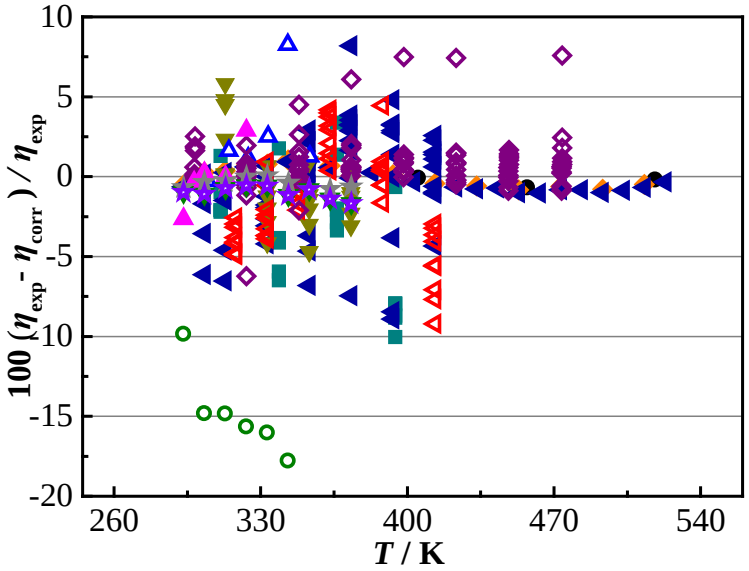

FIG. 12. Percentage deviations [$100(\eta_{exp}-\eta_{corr})/\eta_{exp}$] of the selected secondary experimental viscosity data from the calculated values using Eqs. (1) - (7). (●) Nederbragt and Boelhouwer;[30] (◆) Golubev and Petrov;[35] (■) Rolling and Vogt;[37](○) Ratkovics *et al.*[48] (◀) Annesini et al.;[53] (▼) Ducoulombier *et al.*;[57] (△) Nakazawa *et al.*;[70] (◁) Rajagopal et al.;[92] (◇) Mohammed *et al.*;[100] (▲) Esteban *et al.*;[101] (▽) Luning Prak; [102] (★) Luning Prak *et al.*;[103] (☆) Luning Prak *et al.*[105]

Although no other viscosity correlation of *n*-hexadecane is available in the open literature there are several tables of recommended values[108-111] and Yaws recommended equation[123], all for liquid viscosity at atmospheric pressure. As illustrated in Fig. 13, the agreement between the tabulated values of Vargaftik[108], Scholz and Kley[109], Pugachevich *et al.* [110] and the present correlation is very good and the deviations do not exceed ±5%, except for a couple of points. The deviations of Liessmann *et al.*[111] exhibit a strong temperature trend at temperatures higher than 450 K, while the equation of Yaws[123] agrees reasonably well at temperatures below 550 K, but exhibits increasing deviations at higher temperatures, leading to a difference of -32% at 720 K.

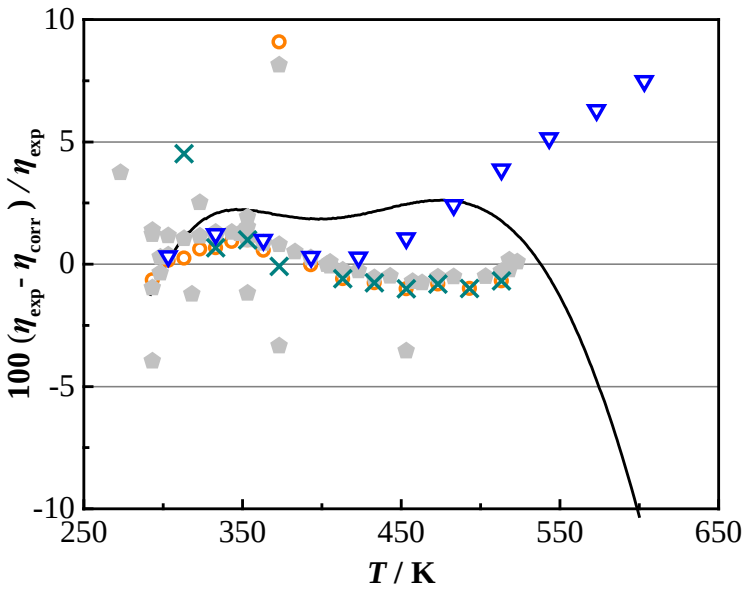

FIG. 13. Percentage deviations [100(η_{exp}-η_{corr})/η_{exp}] of the recommended viscosity data from the calculated values using Eqs. (1) - (7). (○) Vargaftik;[108] (⬟) Scholz and Kley;[109] (×) Pugachevich et al.;[110] (▽) Liessmann *et al.*;[111] (-)Yaws.[123]

5. Computer-Program Verification

Table 6 is provided to assist the user in computer-program verification. The viscosity calculations are based on the tabulated temperatures and densities.

TABLE 6. Sample points for computer verification of the correlating equations

T (K)	ρ (mol L^{-1})	η (μPa s)
300	0.0000	4.015
300	3.3958	2952.341
300	3.5204	5399.820
300	3.9643	64703.310
400	0.0000	5.398
400	3.0865	667.472
400	3.2773	1133.059
400	3.8145	8775.892
600	0.0000	8.135
600	2.8585	325.830
600	3.5780	1463.804
700	0.0000	9.418
700	2.6752	223.559
700	3.4806	949.425

6. Conclusion

A new wide-ranging correlation for the viscosity of *n*-hexadecane has been developed based on critically-evaluated experimental data. The correlation is valid to pressures up to 425 MPa and temperatures from the triple point to 673 K. The correlation is expressed in terms of temperature and density, and the densities were obtained

from the equation of state of Romeo and Lemmon.[28] The overall uncertainty, using a coverage factor of 2, of the proposed correlation is less than 5%, in the temperature and pressure range where experimental data are available, however this uncertainty varies depending on the thermodynamic state and is summarized in more detail in Fig. 11.

Acknowledgments

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7. Appendix A: Viscosity Measurements of *n*-Hexadecane

Table 7. Viscosity measurements of *n*-hexadecane

Authors	Year publ.	Technique employed[a]	No. of data	Temperature range (K)	Pressure range (MPa)
Evans[29]	1938	-	6	293-363	0.1
Nederbragt and Boelhouwer[30]	1947	C	5	298-518	0.1
Nederbragt and Boelhouwer[30]	1947	FB	5	298-518	0.1
Tuot and Guyard[31]	1947	-	1	293	0.1
Koelbel *et al.*[32]	1949	-	4	293-353	0.1
Mumford and Phillips[33]	1950	C	2	293-298	0.1
Bak and Ansersen[34]	1958	C	2	293-303	0.1
Golubev and Petrov[35]	1959	-	16	293-513	p_{sat}
Sanin and Melenteva[36]	1959	C	6	294-373	0.1
Rolling and Vogt[37]	1960	-	23	311-394	0.1-35
Gollis *et al.*[38]	1962	-	3	311-422	0.1
Bidlack and Anderson[39]	1964	C	1	298	0.1
Heric and Brewer[40]	1967	C	1	298	0.1
Anonymous[41]	1968	-	4	293-372	0.1
Jagannathan *et al.*[42]	1968	-	1	293	0.1
Coursey and Heric[43]	1969	C	1	298	0.1
Zhdanov and Lyusternik[44]	1971	-	5	551-724	0.05
Rastorguev and Keramidi[45]	1972	C	18	336-532	0.1-49
Lusternik and Zdanov[46]	1973	-	5	551-723	0.05
Ratkovics *et al.*[47]	1974	-	1	293	0.1
Ratkovics *et al.*[48]	1974	-	6	293-343	0.1
Delmas *et al.*[49]	1975	C	1	298	0.1
Pugachevich and Khvorov[50]	1977	-	8	303-373	0.1
Dymond *et al.*[51]	1980	FB	28	298-373	0.1-425
Dymond and Young[52]	1980	C	10	298-393	P_{sat}
Annesini *et al.*[53]	1982	-	71	293-523	0.1-245
Awwad and Allos[54]	1985	C	1	298	0.1
Awwad[55]	1986	C	1	298	0.1
Awwad and Salman[56]	1986	C	1	298	0.1
Ducoulombier *et al.*[57]	1986	FB	24	313-373	0.1-100
Celda *et al.*[58]	1987	C	1	293	0.1
Matthews *et al.*[59]	1987	C	10	323-564	1.4-4
Soliman[60]	1987	-	10	293-343	0.1
Wakefield and Marsh[61]	1987	C	3	318-338	0.1
Awwad *et al.*[62]	1988	C	1	298	0.1
Wakefield[63]	1988	C	2	303-308	0.1
Asfour *et al.*[64]	1990	C	2	293-298	0.1
Chevalier *et al.*[65]	1990	C	1	298	0.1
Aralaguppi *et al.*[66]	1991	C	3	298-308	0.1
Cooper and Asfour[67]	1991	C	1	293	0.1

Masy and Cournil[68]	1991	-	1	293	0.1
Tanaka *et al.*[69]	1991	TC	16	298-348	0.1-151
Nakazawa *et al.*[70]	1992	RC	6	305-353	0.1
Raikar *et al.*[71]	1993	-	3	298-308	0.1
Aminabhavi and Bindu[72]	1994	C	3	298-318	0.1
De Lorenzi *et al.*[73]	1994	C	1	298	0.1
Aucejo *et al.*[74]	1995	C	1	298	0.1
Aucejo *et al.*[75]	1995	C	1	298	0.1
Dandekar *et al.*[76]	1998	RB	1	293	0.1
Wu *et al.*[77]	1998	C	4	293-313	0.1
Fermeglia and Torriano[78]	1999	C	1	298	0.1
Lal *et al.*[79]	2000	C	1	298	0.1
Nhaesi and Asfour[80]	2000	C	2	293-298	0.1
Mehra and Israni[81]	2002	C	4	298-313	0.1
Queimada *et al.*[82]	2003	RB	6	293-343	0.1
Knothe and Steidley[83]	2005	C	1	313	0.1
Nhaesi and Asfour[84]	2005	C	2	308-313	0.1
Mehra[85]	2005	C	3	298-318	0.1
Tripathi[86]	2005	C	1	298	0.1
Aguila-Hernandez *et al.*[87]	2008	C	3	293-303	0.1
Dubey and Sharma[88]	2008	C	3	298-308	0.1
Dubey and Sharma[89]	2008	C	3	298-308	0.1
Dubey and Sharma[90]	2009	C	3	298-308	0.1
Dubey *et al.*[91]	2009	C	3	298-308	0.1
Rajagopal *et al.*[92]	2009	TC	54	318-413	6.9-62
Ciotta[93]	2010	VW	54	298-474	1.0-103
Luning Prak *et al.*[94]	2013	RC	9	293-373	0.1
Luning Prak *et al.*[95]	2013	RC	5	293-373	0.1
Luning Prak *et al.*[96]	2014	RC	8	293-373	0.1
Baled *et al.*[97]	2014	RB	42	304-534	3.3-227
Wang *et al.*[98]	2016	C	6	298-323	0.1
Wang *et al.*[99]	2016	C	6	298-323	0.1
Mohammed *et al.*[100]	2016	VW	99	298-474	0.6-103
Esteban *et al.*[101]	2016	CP	5	293-323	0.1
Luning Prak[102]	2016	RC	9	293-373	0.1
Luning Prak *et al.*[103]	2017	RC	9	293-373	0.1
Dragoescu *et al.*[104]	2017	RB	1	298	0.1
Luning Prak *et al.*[105]	2017	RC	9	293-373	0.1
Sanchez-Vicente *et al.*[106]	2018	VW	39	323-723	0.97-4
Quiñones-Cisneros *et al.*[107]	-	RC	18	293-373	0.1
Tables of collected data					
Vargaftik[108]	1972	-	16	293-513	0.1
Scholz and Kley[109]	1981	-	45	273-523	0.1
Pugachevich *et al.*[110]	1988	-	10	313-513	0.1

Liessmann *et al.*[111]	1995	-	11	303-603	0.1

[a] C, capillary; FB, falling-body; TC, torsional crystal; VW, vibrating wire; RB, rolling body; RC, rotating cylinder; CP, cone and plate rheometer.

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