

Reference Correlation of the Viscosity of para-Xylene from the Triple Point to 673 K and up to 110 MPa

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Reference Correlation of the Viscosity of *para*-Xylene from the Triple Point to 673 K and up to 110 MPa

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A new correlation for the viscosity of *para*-xylene (*p*-xylene) 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 110 MPa. 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 to 5% for the highest temperatures and pressures of interest. Tables of the viscosity generated by the relevant equations, at selected temperatures and pressures and along the saturation line, are provided. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4908048>]

Key words: correlation; *p*-xylene; transport properties; viscosity.

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1. Introduction

There is a growing industrial need to establish reference values of thermophysical properties of pure fluids that are both accurate and thermodynamically consistent.¹ Not only are such values useful in their own right, but they also serve as the starting point for the prediction of thermophysical properties of mixtures. For thermodynamic properties, the reference values are obtained by recourse to a substance-specific equation of state (EOS) that provides a general framework to correlate the measured properties and ensures thermodynamic consistency. For transport properties, no such general framework is available and one develops separate correlations for different transport properties.

Recently, research and development of state-of-the-art viscosity correlations have gained renewed impetus. Under the auspices of 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,² and since then a plethora of viscosity correlations have been produced, using the same philosophy, covering among others: simple fluids,^{3–5} alkanes,^{6–13} and water.¹⁴ Recently, the work has been extended to cyclic and aromatic hydrocarbons.^{15,16} 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 *p*-xylene that is valid over a wide range of temperature and pressure, covering the vapor, liquid, and supercritical fluid states.

para-Xylene (C₁₀H₈) is an aromatic hydrocarbon that consists of a benzene ring and two–CH₃ groups in positions 1 and 4. At ambient conditions it is a colorless liquid that is primarily used in industry as a raw material and solvent.

Xylene isomers occur naturally in crude oil and are also found in gasoline and to some extent kerosene. The thermodynamic properties are well catered for by an up-to-date EOS,¹⁷ while the thermal conductivity correlation has also become recently available.¹⁸ At present, no correlation of viscosity, valid over a wide range of temperature and pressure, is available and if one wants to predict the viscosity of *p*-xylene, one has to rely on generic correlations^{19,20} developed for a wide variety of fluids that have invariably traded the range of applicability for accuracy.

2. Experimental Viscosity Data

The Appendix summarizes, to the best of our knowledge, the experimental measurements of the viscosity of *p*-xylene reported in the literature,^{21–103} 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 *p*-xylene were reported in 83 papers resulting in 706 data points. Unsurprisingly, the vast majority of researchers (65 papers, 263 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. The Appendix also contains two reference works^{104,105} that report recommended tabulated values of the viscosity of *p*-xylene. There are a number of other papers^{106–119} in the literature that purport to report experimental data, but on closer inspection, the data either have been published previously, or refer to an unspecified xylene mixture, rather than *p*-xylene.

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 criteria¹²⁰ 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, 11 datasets were considered primary data. Table 1 summarizes the primary data^{27,30,31,35,38,39,41,50,54,57,58} detailing the temperature and pressure ranges, the authors' claimed uncertainty and purity of the sample, and the technique employed to perform the measurements. The choice of primary data is discussed in more detail in Sec. 3 that also provides a comparison of the data by different workers.

Figure 1 shows the temperature and pressure range of the measurements outlined in the Appendix with primary and

TABLE 1. Primary data used in developing the viscosity correlation of *p*-xylene

Authors	Year of publication	Technique employed ^a	Purity (%)	Claimed uncertainty (%)	Number of data	Temperature range (K)	Pressure range (MPa)
Mamedov and Panchenkov ²⁷	1955	C	99	...	7	293–353	0.1
Mamedov <i>et al.</i> ³⁰	1968	C	99.78	1.2	129	323–548	0.1–40
Nissema and Koskenniska ³¹	1972	C	...	0.4	4	293–323	0.1
Mamedov <i>et al.</i> ³⁵	1975	C	99.78	1.2	124	323–548	0.1–40
Kashiwagi and Makita ³⁸	1982	TC	99	2	60 ^b	298–348	0.1–110
Abdullaev and Akhundov ³⁹	1983	C	...	1.5	27 ^b	473–673	0.1–3.9
Dymond and Robertson ⁴¹	1985	C	99	0.5	6	298–393	P_{sat}
Serrano <i>et al.</i> ⁵⁰	1990	C	99.98	0.4	4	288–303	0.1
Vogel and Hendl ⁵⁴	1992	OD	99.9	0.3	66	338–635	0.0–0.2
Et-Tahir <i>et al.</i> ⁵⁷	1995	FB	99	2	12	313–353	20–100
Exarchos <i>et al.</i> ⁵⁸	1995	C	99.5	0.4	5	293–313	0.1

^aC, capillary; FB, falling body; OD, oscillating disk; TC, torsional crystal.

^bOne outlier point removed from the analysis.

secondary data distinguished. The primary data cover a wide range of temperatures and pressures of interest. The secondary data are extensive in the liquid phase at ambient pressure, but only two sets report measurements at high pressures.^{24,91} One of these datasets²⁴ was 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.*¹⁷ that covers the thermodynamic space from the triple point to 700 K and up to 200 MPa. Uncertainties in density are estimated to be $\pm 0.2\%$ in the liquid region and $\pm 1.0\%$ elsewhere.

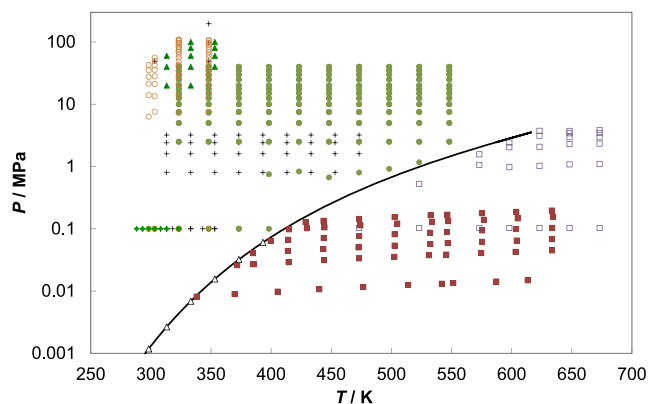


FIG. 1. Distribution of the available experimental viscosity data of *p*-xylene. Primary data: (○) Mamedov *et al.*;^{27,30,35} (●) Kashiwagi and Makita;³⁸ (□) Abdullaev and Akhundov;³⁹ (△) Dymond and Robertson;⁴¹ (■) Vogel and Hendl;⁵⁴ (▲) Et-Tahir;⁵⁷ (◆) data at 0.1 MPa;^{31,50,58} Secondary data: (+).

3. Methodology and Analysis

It is customary¹²¹ 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. 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.¹²² To a lesser extent, we also understand the behavior of fluids at low density.¹²¹ 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,^{2,121} 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.¹²¹ Here, we refrain from naming $\Delta\eta$ as the “higher density terms of the residual viscosity,”^{7,8} and opt for a simpler “residual viscosity,” which adequately describes it, as we did in our previous work on cyclohexane.¹⁵

3.1. The zero-density limit

Two teams of investigators have measured the viscosity of *p*-xylene in the vapor phase.^{39,54} For the development of

the correlation at zero density, we have chosen the data of Vogel and Hendl⁵⁴ as the only primary data set. It covers an extensive temperature range 338–635 K; it was measured in a quartz oscillating-disk viscometer which 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. As the viscosity in the zero-density limit is not a quantity directly accessible from experiment, the temperature range of the primary zero-density viscosity data is actually limited to 383–633 K, as only in this temperature range, the number of data points along each isotherm was sufficient to perform an extrapolation to zero density.⁵⁴

The viscosity in the zero-density limit $\eta_0(T)$ was represented by means of a standard relationship in kinetic theory,^{121,122} that in practical engineering form is given by

$$\eta_0(T) = \frac{1}{4} \sqrt{\frac{k_B}{\pi N_A}} \frac{\sqrt{MT}}{S_\eta} = 0.22005 \frac{\sqrt{T}}{S_\eta}, \quad (2)$$

where T is the temperature in K, M is the molar mass, S_η is the effective collision cross section in nm², and $\eta_0(T)$ is in $\mu\text{Pa s}$. The numerical constant in Eq. (2) was obtained by the use of the recommended values of fundamental constants¹²³ and a relative molecular mass of 106.165.¹²⁴ 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,

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

where A_0 , B_0 , and C_0 are the adjustable parameters. The primary data set of Vogel and Hendl,⁵⁴ consisting of ten 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 0.17%, 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 functional form for the effective cross section, S_η , and then using two scaling parameters, one for length, σ , and the other for energy, ε , to perform the fitting.¹²¹ 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) potential¹²⁵ and the other that employs a universal correlation for S_η that is based on experimental data.^{126,127} 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

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

A_0 (-)	B_0 (K)	C_0 (K ²)
-1.4933	473.2	-57 033

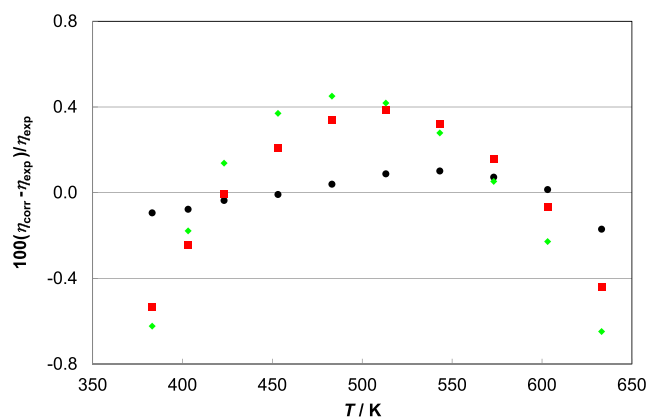


FIG. 2. Percentage deviations $[100(\eta_{\text{corr}} - \eta_{\text{exp}})/\eta_{\text{exp}}]$ from the experimental data of Vogel and Hendl⁵⁴ in the limit of zero density, of the values calculated by: (●) Eqs. (2) and (3); (■) Lennard-Jones correlation;¹²⁵ and (◆) universal correlation.^{126,127}

fitting the primary data for *p*-xylene using the functional forms of S_η corresponding to Lennard-Jones and the universal correlation.^{125–127} By a judicious fitting of the scaling parameters (σ and ε), both Lennard-Jones and universal correlation can be made to represent the data within systematic deviations of 0.65%, which is approximately twice as large as the claimed uncertainty of the measured data. This confirms our previous finding for cyclohexane 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.^{121,128,129} The decrease in viscosity, which for *p*-xylene occurs at temperatures below 606 K and reaches a maximum value at 453 K of approximately 1.7%, necessitates a separate analysis. We have based the analysis of initial density dependence on the data set of Vogel and Hendl,⁵⁴ consisting of 66 data points measured in the temperature range 338–635 K and pressures up to 0.2 MPa. The claimed uncertainty of these data ranges from 0.15% at lower temperatures to 0.3% at the highest temperature. We have supplemented this data set with the data of Abdullaev and Akhundov,³⁹ measured in a capillary viscometer, at atmospheric pressure in the temperature range 473–673 K. Although this data set, consisting of seven data points, has a much larger claimed uncertainty of 1.5%, it allows us to extend the temperature range of the primary data set to 673 K.

It is customary to express the initial-density coefficient, η_1 , in terms of the second viscosity virial coefficient, B_η , by means of¹²¹

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

We have already shown¹⁵ that this functional form could not reproduce the cyclohexane data of Vogel *et al.*¹³⁰ within

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

A_1 ($\mu\text{Pa s mol}^{-1}$ l)	B_1 ($\mu\text{Pa s K mol}^{-1}$ l)	C_1 ($\mu\text{Pa s K}^2 \text{mol}^{-1}$ l)
13.2814	-10 862.4	1 664 060

their claimed uncertainty. In this work, we reached the same conclusion concerning the current viscosity data for *p*-xylene. Therefore, as in our previous work,¹⁵ we opted to represent the initial density dependence by a simple empirical functional form

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

where A_1 , B_1 and C_1 are the adjustable coefficients and ρ is the molar density in units of mol l^{-1} . In order to calculate $\eta_1\rho$ term from the measured viscosity, the zero-density viscosity correlation was extrapolated to 338 K at the lower end and to 673 K at high-temperature end. Hence, five experimental data points that lie in the extrapolated temperature range were also included in the analysis. The primary data set, consisting of 73 data points, was fitted by means of Eq. (5). 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 of Vogel and Hendl are fitted within 0.2%, which is within the claimed experimental uncertainty of 0.15–0.3%. The data of Abdullaev and Akhundov³⁹ were also correlated well within its claimed uncertainty. In the temperature region 473–635 K, where the data from two laboratories overlap the agreement between the two sets of data is within 0.7%. However, as illustrated in Fig. 3, the data of Abdullaev and Akhundov do display a different density trend than data of Vogel and Hendl. As the five data points in the temperature range 338–383 K and 633–673 K were reproduced within their experimental uncertainty, one can surmise that the extrapolation of the zero-density correlation is valid.

Based on the agreement with the primary data set, we ascribe a combined expanded uncertainty with coverage factor

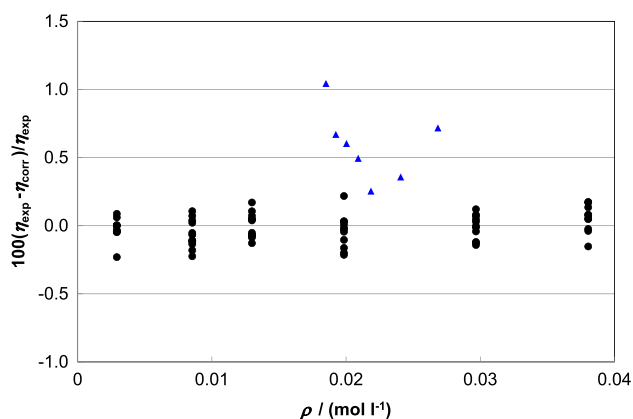


FIG. 3. Percentage deviations $[100(\eta_{\text{exp}} - \eta_{\text{corr}})/\eta_{\text{exp}}]$ of the available experimental data in the vapor phase at pressures of 0.2 MPa and below from the calculated values. (\blacktriangle) Abdullaev and Akhundov³⁹ and (\circ) Vogel and Hendl.⁵⁴

of 2 to the viscosity correlation in the vapor phase, below 0.2 MPa, of 0.5% in the temperature range 338–635 K. In the high temperature limit, we estimate the uncertainty to increase to 1% at 673 K. We do not recommend the use of Eqs. (2) and (5) to predict the viscosity of the vapor *p*-xylene at temperatures below 338 K. The lack of experimental data and the empirical nature of the equations makes the extrapolation rather uncertain. However, the use of Eqs. (2) and (5), as part of Eq. (1), to predict the liquid viscosity from the triple point to 338 K is recommended since the contribution of low-density terms to the overall liquid viscosity is small.

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.¹³¹ Based on the previous studies,^{2,7} we would expect the ratio $\Delta\eta_c/\eta$ for *p*-xylene to exceed 0.01 only in the region that would cover $604 < T/\text{K} < 628$ and $1.8 < \rho/(\text{mol l}^{-1}) < 3.6$. Only one viscosity data point, measured at 623 K and 1.945 mol l^{-1} by Abdullaev and Akhundov,³⁹ exists in this region. In principle, it would be possible to estimate the viscosity critical enhancement based on the model of Bhattacharjee *et al.*¹³² This would require a knowledge of $(\partial p/\partial \rho)_T$ and $(\partial p/\partial T)_{\rho=\rho_c}$ and values of two wave number cutoff parameters, q_C and q_D . If there are no viscosity data in the critical region, the value of cutoff parameter q_D can be taken from the analysis of thermal conductivity data. Although such a value of q_D for *p*-xylene is available,¹⁸ the recent analysis of benzene viscosity data concluded that the usefulness of using such a model to estimate the viscosity critical enhancement is negated by the complexity and high uncertainty of the resulting correlation. Thus, in line with the development of other correlations,^{3,5,6,8–13,15,16} the viscosity critical enhancement contribution of *p*-xylene is taken as zero. The total lack of industrial applications of *p*-xylene near its critical temperature further supports this choice.

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 seven cover a wide range of temperature and pressure. Based on the analysis of the measurement techniques and the authors measurements on other fluids, we have chosen four datasets as primary in the liquid region. Mamedov and coworkers^{30,35} performed the experiments using a capillary viscometer with a claimed uncertainty of 1.2%. Kashiwagi and Makita³⁸ used a torsional crystal viscometer, while Et-Tahir *et al.*⁵⁷ made the measurements by means of the falling body viscometer. Both sets of authors claimed uncertainty of 2%, which is well-supported by their measurements on other fluids.^{15,38,133} The primary data in the liquid state thus covered the temperature range 298–548 K and pressures from 0.1 MPa up to 110 MPa. We note that

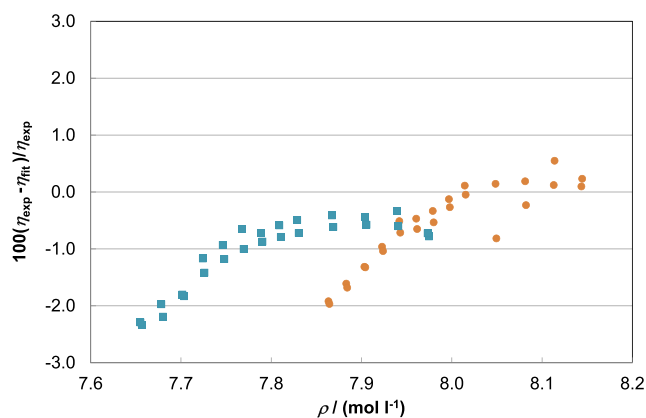


FIG. 4. Percentage deviations $[100(\eta_{\text{exp}} - \eta_{\text{fit}})/\eta_{\text{exp}}]$ of Mamedov *et al.*^{30,35} experimental viscosity data at 323 K and 348 K from the quadratic fit in density to data of Kashiwagi and Makita³⁸ at each temperature. (●) 323 K and (■) 348 K.

the later work of Mamedov and coworkers¹¹⁰ contains no new experimental data, but only the analysis of the data obtained in 1968.

Figure 4 illustrates the comparison of the data of Mamedov *et al.*^{30,35} at 323–348 K with the data of Kashiwagi and Makita³⁸ that were measured along the same isotherms. In order to perform the comparison, we used at each temperature a quadratic polynomial fit in density to the data of Kashiwagi and Makita³⁸ as the baseline. It is clear from Fig. 5 that the two sets of data exhibit a systematic deviation as we approach the saturation line. However, the deviations are within the combined claimed uncertainty of both sets of data.

The four primary datasets covered the liquid part of the phase diagram. We have also included the data of Abdullaev and Akhundov³⁹ that report the measurements of the viscosity of *p*-xylene in the vapor phase in the temperature range 473–673 K and pressures up to 3.9 MPa. The outlier at 623.15 K and 3.68 MPa, that initial analysis uncovered, was removed from the primary data set. Good agreement of these data at atmospheric pressure with the measurements of Vogel and Hendl, illustrated in Fig. 3, indicates that the claimed uncertainty of 1.5% is justified.

The primary data set also contains four sets of viscosity measurements^{27,31,50,58} of liquid *p*-xylene at atmospheric pressure covering the temperature range 288–353 K. The choice was based on careful analysis of the available data that involved: (i) use of viscometer capable of producing primary data set; (ii) low quoted uncertainty that is supported by other measurements by the same authors; in this instance, measurements of viscosity of cyclohexane¹⁵ and benzene¹⁶ were used; (iii) large temperature range. Finally, the primary data were supplemented with one set of measurements⁴¹ along the liquid saturation line covering the temperature range 298–393 K. In summary, 371 data points covering the temperature range 288–673 K and pressures up to 110 MPa measured in ten 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

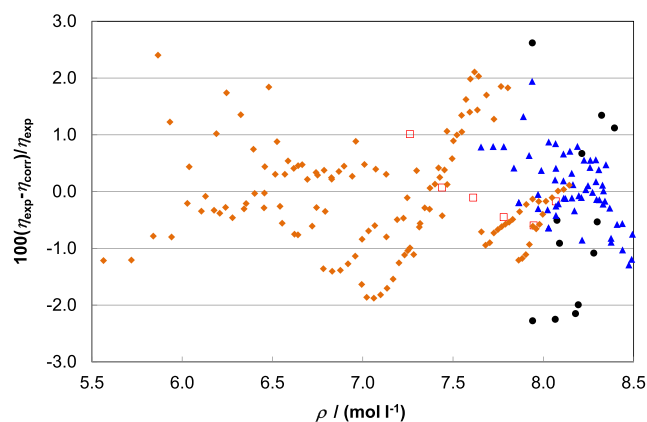


FIG. 5. Percentage deviations $[100(\eta_{\text{exp}} - \eta_{\text{corr}})/\eta_{\text{exp}}]$ of the primary experimental viscosity data in the liquid region from the values calculated by Eqs. (1)–(3), (5), and (6): (◆) Mamedov *et al.*;^{30,35} (▲) Kashiwagi and Makita;³⁸ (□) Dymond and Robertson;⁴¹ and (●) Et-Tahir *et al.*⁵⁷

*et al.*¹⁷ 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. As discussed previously,^{8,15} this makes the choice of the functional form to fit the data rather difficult. As a result, a number of existing viscosity correlations exhibit nonmonotonic 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 states¹²¹ or in Vesovic-Wakeham (VW) models.^{134–136}

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, 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

$$\Delta\eta(\rho, T) = \rho_r^{2/3} \sum_{i=1,5,2,3,4,5,11} D_i \rho_r^i + \frac{\rho_r^{2/3}}{\sqrt{T_r}} \sum_{i=1,5,11,15} E_i \rho_r^i, \quad (6)$$

where D_i and E_i 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. We have also adopted a slightly different functional form to that used in the analysis of cyclohexane viscosity data.¹⁵ In essence, we have taken advantage of the hard sphere result¹³⁷ $\Delta\eta(\rho, T)$

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

i	D_i	E_i
1.5	122.919	15.337
2	-282.329	...
3	279.348	...
4	-146.776	...
5	28.361	...
11	-0.004 585	-0.000 438 2
15	...	0.000 023 07

^a $T_c = 616.168$ K; $\rho_c = 2.693 92$ mol l⁻¹ obtained from Ref. 17

= $\rho_r^{2/3}/T_r^{1/2} f(\rho_r, T_r)$ as already used in correlating the viscosity of benzene¹⁶ and the fact that excess viscosity of supercritical fluids is independent of temperature.^{19,138} One can thus, very broadly, claim that the viscosity behavior at supercritical and high-pressure vapor conditions is governed by the coefficients D_i , while the coefficients E_i govern the behavior in the liquid region.

The procedure adopted during this analysis used the R software for statistical computing¹³⁹ to fit Eq. (6) to the primary data. The uncertainties quoted in Table 1 were used to determine relative weights. The optimal coefficients D_i and E_i are shown in Table 4 together with the critical temperature and critical density of *p*-xylene.

Figures 5 and 6 illustrate the percentage deviation of the primary viscosity data used in developing the correlation from the developed viscosity correlation, Eqs. (1)–(3), (5), and (6). Figure 5 illustrates the agreement with the experimental data in the liquid region for pressures higher than atmospheric. All the experimental data^{30,35,38,41,57} are reproduced by the proposed correlation within 2.6%, with no systematic trend in any of the data sets. The data of Dymond and Young⁴¹ measured along the saturation line are reproduced within 0.6%, which is just outside the claimed uncertainty of the data. The only exception being a datum at the highest temperature measured (393 K) which was reproduced within 1%. The data of Et-Tahir *et al.*⁵⁷ exhibit deviations from the correlation that are just outside the claimed uncertainty, with maximum deviation of 2.6%. It is worth noting that we only used the high-pressure data of Et-Tahir *et al.* as primary, see Table 1. Et-Tahir *et al.*⁵⁷ also report three measurements at atmospheric pressure in the temperature range 313–353 K that were performed in a different viscometer, capillary rather than falling body. These data points exhibit average absolute deviation (AAD) of 2.6% and maximum deviation of 3.0%. Based on the deviations illustrated in Fig. 5, the uncertainty of the data of Mamedov and coworkers^{27,30,35} is nearer to 2%, rather than the claimed 1.2%.

Figure 6 illustrates the agreement of the developed viscosity correlation with the primary experimental data at atmospheric pressure that cover the temperature range 288–373 K, in the liquid phase. All of the data are reproduced within 1.5%, with the exception of a single datum of Mamedov and Panchenkov²⁷ at 353 K. It is reassuring that the proposed viscosity correlation predicts the data of Exarchos *et al.*,⁵⁸ Serrano *et al.*,⁵⁰ and Nissema and Koskenniska³¹ just outside their claimed uncertainty of 0.4%.

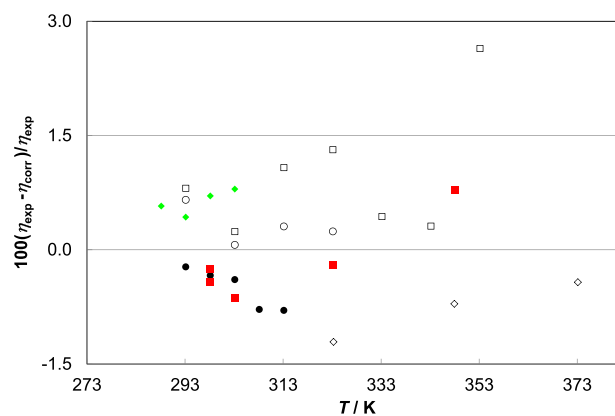


FIG. 6. Percentage deviations $[100(\eta_{\text{exp}} - \eta_{\text{corr}})/\eta_{\text{exp}}]$ of the primary experimental viscosity data measured at 0.1 MPa from the calculated values using Eqs. (1)–(3), (5), and (6). (□) Mamedov and Panchenkov;²⁷ (◇) Mamedov *et al.*;³⁰ (○) Nissema and Koskenniska;³¹ (■) Kashiwagi and Makita;³⁸ (◆) Serrano *et al.*;⁵⁰ and (●) Exarchos *et al.*⁵⁸

Table 5 summarizes the agreement between the primary experimental data and the proposed viscosity correlation for *p*-xylene in the liquid, dense vapor, and supercritical regions. The correlation recaptures the entire set of primary data with an AAD of 0.6%, zero bias and maximum deviation of 2.6%. We have estimated the overall uncertainty of the correlation, 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 point (286.4 K)¹⁷ to 373 K, we estimate the uncertainty to be 1.0%; (ii) along the liquid saturation line in the same temperature range 286.4–373 K, we estimate the uncertainty to be also 1.0%; (iii) in the rest of the liquid region, we estimate the uncertainty to be 2.0%; (iv) everywhere else, in the high-pressure vapor and supercritical

TABLE 5. Evaluation of the *p*-xylene viscosity correlation against the primary experimental data

Authors	Year of publication	AAD ^a (%)	Bias ^b (%)	MD ^c (%)
Mamedov and Panchenkov ²⁷	1955	1.0	1.0	2.6
Mamedov <i>et al.</i> ³⁰	1968	0.8	-0.1	2.4
Nissema and Koskenniska ³¹	1972	0.3	0.3	0.7
Mamedov <i>et al.</i> ³⁵	1975	0.7	-0.2	-2.2
Kashiwagi and Makita ³⁸	1982	0.5	0.0	1.9
Abdullaev and Akhundov ³⁹	1983	1.0	0.5	2.6
Dymond and Robertson ⁴¹	1985	0.4	0.0	1.0
Serrano <i>et al.</i> ⁵⁰	1990	0.6	0.6	0.8
Vogel and Hendl ⁵⁴	1992	0.1	0.0	-0.2
Et-Tahir <i>et al.</i> ⁵⁷	1995	1.5	-0.5	2.6
Exarchos <i>et al.</i> ⁵⁸	1995	0.5	-0.5	0.8
Entire primary data set		0.6	0.0	2.6

^aAAD, Average Absolute Deviation = $100/N \sum |(\eta_{\text{exp}} - \eta_{\text{corr}})/\eta_{\text{exp}}|$

^bBias = $100/N \sum (\eta_{\text{exp}} - \eta_{\text{corr}})/\eta_{\text{exp}}$

^cMD, Maximum deviation

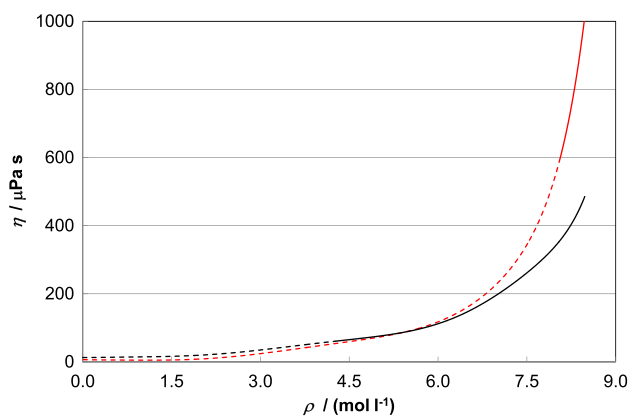


FIG. 7. Viscosity of *p*-xylene as a function of density along a couple of isotherms. (red solid line) 300 K, liquid phase; (red dashed line) 300 K, two-phase region; (black solid line) 600 K, liquid phase; and (black dashed line) 600 K, two-phase region.

region, we estimate the uncertainty to be 2.5%; (v) in the region (>548 K and >40 MPa) where no experimental data are available, we conservatively estimate the uncertainty to be 5%.

4. Overall Viscosity Correlation

The viscosity correlation of *p*-xylene 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 286.4–673 K and pressure (up to 110 MPa) range. In the vapor phase, the lower temperature limit corresponds to 338 K. Figure 7 illustrates the behavior of the viscosity correlation as a function of density along the 300 K and 600 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 monotonic increase with

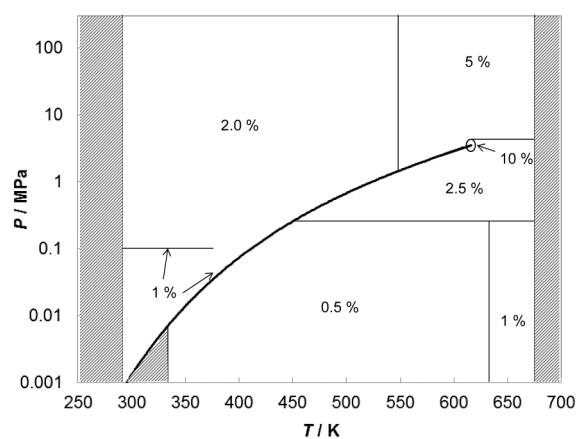


FIG. 8. The extent of the viscosity representation and its estimated uncertainty. No representation is available in the hatched region.

density except at low densities, of up to 1.1 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 cyclic hydrocarbons¹²¹ or as part of the VW model^{134–136} to predict the viscosity of mixtures containing *p*-xylene.

It is possible to extrapolate the proposed viscosity correlation up to 200 MPa by making use of the densities calculated from the EOS of Zhou *et al.*¹⁷ 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 a single measurement of Bridgman²⁴ performed at 196 MPa and 348 K that exhibits a deviation of -1.3% . The deviation compares favorably with the other data of Bridgman²⁴ measured in the temperature range 303 K–348 K and pressures up to 98 MPa. The AAD and MD of these five points are 1.0% and -2.2% , respectively.

TABLE 6. Recommended viscosity values in $\mu\text{Pa s}$

P MPa	T/K										
	290	300	310	320	330	340	350	370	470	570	670
0	7.36	7.55	7.96	10.05	12.15	14.21
0.1	670.7	593.3	528.6	474.2	428.2	389.0	355.3	300.4	145.6	12.14	14.22
0.5	672.9	595.2	530.3	475.8	429.6	390.3	356.5	301.5	146.6	69.16	14.34
1	675.6	597.6	532.5	477.7	431.4	391.9	358.0	302.8	147.8	72.24	14.59
2	681.1	602.5	536.8	481.6	434.9	395.1	361.0	305.5	150.3	76.36	15.33
4	692.2	612.3	545.6	489.5	442.0	401.6	367.0	310.7	155.1	82.31	17.49
6	703.4	622.2	554.4	497.3	449.1	408.1	373.0	316.0	159.6	87.18	32.89
8	714.7	632.2	563.2	505.3	456.3	414.6	378.9	321.2	164.1	91.55	53.54
10	726.1	642.2	572.1	513.2	463.5	421.2	384.9	326.4	168.3	95.61	62.47
30	845.0	747.1	665.0	596.0	537.6	488.1	445.9	378.3	206.1	129.7	92.57
50	974.5	861.0	765.6	685.1	617.1	559.3	510.0	431.5	238.6	158.3	114.8
70	1115.4	984.9	874.9	781.9	703.1	636.0	578.7	487.4	268.5	183.6	136.1
90	1268.6	1119.8	994.0	887.3	796.5	719.1	653.0	547.3	297.2	206.6	156.4
110	1434.6	1266.3	1123.4	1001.8	898.1	809.5	733.5	612.0	325.6	227.6	175.3

TABLE 7. Recommended viscosity values along the saturation line

T/K	P_v /MPa	Vapor		Liquid	
		ρ /(mol l ⁻¹)	η /(μ Pa s)	ρ /(mol l ⁻¹)	η /(μ Pa s)
293.15	0.0009	0.0004	...	8.1100	644.2
303.15	0.0016	0.0006	...	8.0283	571.2
313.15	0.0027	0.0010	...	7.9462	510.0
323.15	0.0043	0.0016	...	7.8637	458.5
333.15	0.0069	0.0025	...	7.7805	414.9
343.15	0.0105	0.0037	7.40	7.6966	377.5
353.15	0.0156	0.0054	7.59	7.6118	345.4
403.15	0.0807	0.0251	8.56	7.1702	233.8
453.15	0.2746	0.0803	9.53	6.6834	164.5
503.15	0.7131	0.2073	10.65	6.1182	116.3
553.15	1.5487	0.4883	12.30	5.3990	84.23

Figure 8 summarizes the estimated combined expanded uncertainty with coverage factor of 2 of the proposed viscosity correlation as a function of temperature and pressure.

Table 6 contains the recommended values of viscosity of *p*-xylene 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 *p*-xylene along the saturation line.

Figure 9 summarizes the deviations of the selected secondary data, measured at atmospheric pressure in the last fifteen years, from the current correlation. Although many measurements are within the acceptable 1-2%, there are a number of data sets that exhibit much larger deviations. Only two sets of experimental data, classified as secondary, extend to higher pressure. The deviation of data of Bridgman²⁴ has already been discussed, while the data of Fang *et al.*⁹¹ display the AAD of 2% with the maximum deviation of 10.2% from the proposed correlation.

Although no other viscosity correlation of *p*-xylene is available in the open literature, there are a couple of tables of recommended values^{104,105} for liquid viscosity at atmospheric

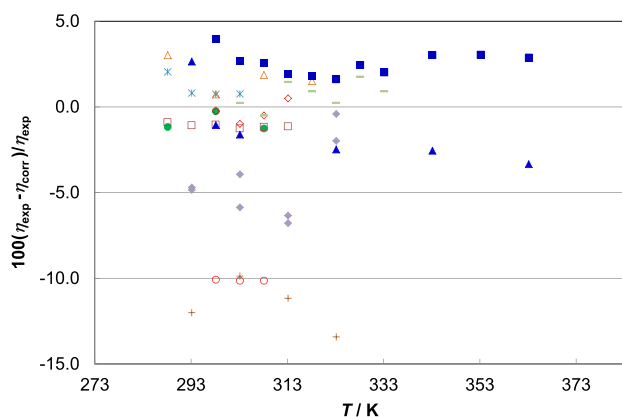


FIG. 9. Percentage deviations $[100(\eta_{\text{exp}} - \eta_{\text{corr}})/\eta_{\text{exp}}]$ of the selected secondary experimental viscosity data measured at 0.1 MPa from the calculated values using Eqs. (1)–(3), (5), and (6). (○) Aminabhavi and Banerjee;⁶⁴ (◆) Prasad *et al.*;^{63,67} (■) Yang *et al.*;⁷² (▲) Feng *et al.*;⁸⁰ (◇) Kapoor and Rattan;⁸¹ (*) Al-Kandary *et al.*;⁸⁴ (Δ) Nain;⁸⁸ (●) Wankhede *et al.*;⁹⁰ (–) Song *et al.*;⁹² (□) Ikeuchi *et al.*;⁹⁸ and (+) Dikio *et al.*¹⁰³

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

T (K)	ρ (mol l ⁻¹)	η (μ Pa s)
300	0	6.604
300	0.0490	6.405
300	8.0548	593.272
300	8.6309	1266.337
400	0	8.573
400	7.1995	239.202
400	8.0735	484.512
600	0	12.777
600	7.0985	209.151

pressure together with the Yaws recommended equations²⁰ for both vapor and liquid viscosity, also at atmospheric pressure. The agreement between the three correlations for the liquid viscosity and the present correlation is very good and the deviations do not exceed $\pm 1\%$. However, Yaws' proposed equation²⁰ for the vapor viscosity underpredicts the current correlation by 8%–18% in the temperature range 403–283 K. This is not surprising as the proposed equation was fitted to viscosity values calculated by one of the generic methods.^{19,20}

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 *p*-xylene has been developed based on critically evaluated experimental data. The correlation is valid from the triple point to 673 K and pressures up to 110 MPa in the liquid part of the phase diagram. In the vapor part of the phase diagram, the correlation is valid in the temperature range 338–673 K and pressures up to 110 MPa. The correlation is expressed in terms of temperature and density, and the densities were obtained from the equation of state of Zhou *et al.*¹⁷ The overall uncertainty, using a coverage factor of 2, of the proposed correlation is less than 5.0%; however, this uncertainty varies depending on thermodynamic state and is summarized in more detail in Fig. 8.

Acknowledgments

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7. Appendix: Viscosity Measurements of *p*-Xylene

Authors	Year of publication	Technique employed ^a	No. of data	Temperature range (K)	Pressure range (MPa)
Thorpe and Rodger ²¹	1894	C	12	281–408	0.1
Batschinski ²²	1913	–	14	273–403	0.1
Kremann <i>et al.</i> ²³	1914	C	2	285–337	0.1
Bridgman ²⁴	1926	–	6	303–348	0.098–196.1
Timmermans and Martin ²⁵	1926	C	2	288–303	0.1
Geist and Cannon ²⁶	1946	–	2	293–313	0.1
Mamedov and Panchenkov ²⁷	1955	C	7	293–353	0.1
Panchenkov ²⁸	1957	C	7	293–353	0.1
Panchenkov <i>et al.</i> ²⁹	1958	C	7	281–408	0.1
Mamedov <i>et al.</i> ³⁰	1968	C	129	323–548	0.1–40
Nissema and Koskenniska ³¹	1972	C	4	293–323	0.1
Schmack <i>et al.</i> ³²	1973	C	3	293–323	0.1
Alms <i>et al.</i> ³³	1973	C	1	297	0.1
Friedel and Raetzsch ³⁴	1973	C	1	293	0.1
Mamedov <i>et al.</i> ³⁵	1975	C	124	323–548	0.1–40
Dhillon and Chugh ³⁶	1976	C	2	298–308	0.1
Yadava and Yadava ³⁷	1981	–	1	307	0.1
Kashiwagi and Makita ³⁸	1982	TC	61	298–348	0.1–110
Abdullaev and Akhundov ³⁹	1983	C	28	473–673	0.1–3.9
Nath and Narain ⁴⁰	1983	C	1	303	0.1
Dymond and Robertson ⁴¹	1985	C	6	298–393	P_{sat}
Al-Madfai <i>et al.</i> ⁴²	1985	C	1	298	0.1
Aminabhavi <i>et al.</i> ⁴³	1987	–	1	298	0.1
Manjeshwar and Aminabhavi ⁴⁴	1987	C	2	298–318	0.1
Ramanjaneyulu <i>et al.</i> ⁴⁵	1987	C	1	303	0.1
Chandrasekhar <i>et al.</i> ⁴⁶	1988	C	1	303	0.1
Fermeglia and Lapasin ⁴⁷	1988	C	1	298	0.1
Rathnam ⁴⁸	1988	C	2	303–313	0.1
Ruiz <i>et al.</i> ⁴⁹	1989	–	3	303–323	0.1
Serrano <i>et al.</i> ⁵⁰	1990	C	4	288–303	0.1
Chevalier <i>et al.</i> ⁵¹	1990	C	1	298	0.1
Fermeglia <i>et al.</i> ⁵²	1990	C	1	298	0.1
Sivaramprasad <i>et al.</i> ⁵³	1990	C	1	298	0.1
Vogel and Hendl ⁵⁴	1992	OD	66	338–635	0.008–0.2
Wanchoo and Narayan ⁵⁵	1992	C	4	293–318	0.1
Tasioula-Margari and Demetropoulos ⁵⁶	1992	C	5	298–318	0.1
Et-Tahir <i>et al.</i> ⁵⁷	1995	FB	12	313–353	20–100
Et-Tahir <i>et al.</i> ⁵⁷	1995	C	3	313–353	0.1
Exarchos <i>et al.</i> ⁵⁸	1995	C	5	293–313	0.1
Petrino <i>et al.</i> ⁵⁹	1995	C	1	298	0.1
Ramachandran <i>et al.</i> ⁶⁰	1995	C	1	303	0.1
Singh <i>et al.</i> ⁶¹	1995	C	1	298	0.1
Oswal <i>et al.</i> ⁶²	1996	C	1	303	0.1
Prasad <i>et al.</i> ⁶³	1999	FB	4	303–323	0.1
Aminabhavi and Banerjee ⁶⁴	1999	C	3	298–308	0.1
Aralaguppi <i>et al.</i> ⁶⁵	1999	C	3	298–308	0.1
Goud <i>et al.</i> ⁶⁶	1999	C	1	303	0.1
Prasad <i>et al.</i> ⁶⁷	2000	FB	4	298–323	0.1
Tsierkezos <i>et al.</i> ⁶⁸	2000	C	1	293	0.1
Tu <i>et al.</i> ⁶⁹	2001	C	1	298	0.1
Nayak <i>et al.</i> ⁷⁰	2002	C	3	293–313	0.1
Peng and Tu ⁷¹	2002	C	1	298	0.1
Yang <i>et al.</i> ⁷²	2002	C	11	298–363	0.1
Moreiras <i>et al.</i> ⁷³	2003	RB	1	298	0.1
George and Sastry ⁷⁴	2003	C	2	298–308	0.1
Lark <i>et al.</i> ⁷⁵	2003	C	2	298–303	0.1
Oswal <i>et al.</i> ⁷⁶	2004	C	1	303	0.1
Singh <i>et al.</i> ⁷⁷	2004	C	1	298	0.1
Yang <i>et al.</i> ⁷⁸	2004	C	3	298–323	0.1
Yang <i>et al.</i> ⁷⁹	2004	C	7	298–353	0.1
Feng <i>et al.</i> ⁸⁰	2005	C	6	298–363	0.1
Kapoor and Rattan ⁸¹	2005	C	3	303–313	0.1
Oswal <i>et al.</i> ⁸²	2005	C	1	298	0.1
Rathnam <i>et al.</i> ⁸³	2005	C	2	303–313	0.1

Authors	Year of publication	Technique employed ^a	No. of data	Temperature range (K)	Pressure range (MPa)
Ali <i>et al.</i> ⁸⁴	2006	C	5	298–318	0.1
Al-Kandary <i>et al.</i> ⁸⁵	2006	C	4	288–303	0.1
Baragi and Aralaguppi ⁸⁶	2006	C	3	298–308	0.1
Di <i>et al.</i> ⁸⁷	2006	C	9	298–353	0.1
Nain ⁸⁸	2007	C	4	288–318	0.1
Yang <i>et al.</i> ⁸⁹	2007	C	7	298–353	0.1
Wankhede <i>et al.</i> ⁹⁰	2008	C	3	288–308	0.1
Fang <i>et al.</i> ⁹¹	2008	FB	39	313–473	0.1–3.2
Song <i>et al.</i> ⁹²	2008	C	7	303–333	0.1
Dominguez-Perez <i>et al.</i> ⁹³	2009	C	1	298	0.1
Sastry <i>et al.</i> ⁹⁴	2009	C	2	298–308	0.1
Yang <i>et al.</i> ⁹⁵	2009	C	3	298–318	0.1
Habibullah <i>et al.</i> ⁹⁶	2010	C	3	303–323	0.1
Habibullah <i>et al.</i> ⁹⁷	2010	C	2	308–318	0.1
Ikeuchi <i>et al.</i> ⁹⁸	2010	C	6	288–313	0.1
Yadava and Yadav ⁹⁹	2010	C	1	308	0.1
Bhatia <i>et al.</i> ¹⁰⁰	2011	C	2	298–308	0.1
Yadava <i>et al.</i> ¹⁰¹	2011	C	1	308	0.1
Zarei and Salami ¹⁰²	2012	C	1	298	0.1
Dikio <i>et al.</i> ¹⁰³	2013	C	4	293–323	0.1
Tables of collected data					
Golubev ¹⁰⁴	1970	–	13	283–403	0.1
NIST/TRC database 2008 ¹⁰⁵	2003	–	26	288–413	0.1

^aC, capillary; FB, falling body; OD, oscillating disc; RB, rolling body; TC, torsional crystal.

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