Reference correlation of the viscosity of ethylbenzene from the triple point to 673 K and up to 110 MPa

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Abstract

A new correlation for the viscosity of ethylbenzene 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 1% for the viscosity at atmospheric pressure 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.

Comparison of viscosity of xylene isomers indicated that at very high temperatures the viscosity correlation of *para*-xylene has higher uncertainty than previously postulated. Thus, in this work we also provide a revised viscosity correlation for *p*-xylene.

CONTENTS

1. Introduction
2. Experimental Viscosity Data
3. Methodology and Analysis
 - 3.1. The zero-density and initial-density terms
 - 3.2. The critical enhancement and the residual viscosity terms
4. Overall Viscosity Correlation
5. Computer-Program Verification
6. Comparison of viscosity of xylene isomers
7. Conclusion
- Acknowledgments
8. Appendix A: Viscosity measurements of ethylbenzene
9. Appendix B: Revised reference correlation of the viscosity of *p*-xylene from triple point to 673 K and up to 110 MPa
10. References

List of Tables

1. Primary data used in developing the viscosity correlation of ethylbenzene
2. Coefficients for the representation of the residual viscosity, Eqs. (6) and (7)
3. Evaluation of the ethylbenzene viscosity correlation against the primary experimental data
4. Recommended viscosity values
5. Recommended viscosity values along the saturation line
6. Sample points for computer verification of the correlating equations
7. Viscosity measurements of ethylbenzene
8. Coefficients for the representation of the residual viscosity of *p*-xylene, Eq. (9)
9. Evaluation of the revised *p*-xylene viscosity correlation against the primary experimental data
10. Recommended viscosity values for *p*-xylene
11. Recommended viscosity values for *p*-xylene along the saturation line
12. Sample points for computer verification of the correlating equations

List of Figures

1. Distribution of the available experimental viscosity data of ethylbenzene.
2. Percentage deviations $[100(\rho_{\text{exp}} - \rho_{\text{corr}}) / \rho_{\text{exp}}]$ of the experimental density data from the calculated values using EoS developed by Zhou *et al.*¹
3. Percentage deviations $[100(\eta_{\text{exp}} - \eta_{\text{corr}}) / \eta_{\text{exp}}]$ of the available experimental data of Abdullaev and Akhundov⁴¹ in the vapor phase at 0.1 MPa.
4. Comparison of the experimental liquid viscosity data at high pressure.
5. Comparison of the experimental liquid viscosity data measured at 0.1 MPa.
6. 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) - (7).
7. 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) - (7).
8. Viscosity of ethylbenzene as a function of density along a couple of isotherms.
9. The extent of the viscosity representation and its estimated uncertainty.
10. 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) - (7).
11. Percentage deviations $[100(\eta_{\text{exp}} - \eta_{\text{corr}}) / \eta_{\text{exp}}]$ of the selected secondary experimental viscosity data at high pressures from the calculated values using Eqs. (1) - (7).
12. Viscosity of four xylene isomers as a function of density at 300 K.
13. Viscosity of four xylene isomers as a function of density at 450 K.
14. Viscosity of four xylene isomers as a function of temperature at 0.1 MPa.
15. Viscosity of *p*-xylene as a function of density at 473 K.
16. Viscosity of *p*-xylene as a function of density at 673 K.

1. Introduction

Ethylbenzene (C_8H_{10}) is an aromatic hydrocarbon that consists of a benzene ring and $-C_2H_5$ group. At ambient conditions it is a colorless, flammable liquid that has been widely used in the production of polystyrene. The thermodynamic properties of ethylbenzene are well catered for, by an up-to-date EoS,¹ while the thermal conductivity correlation has also become recently available.² At present, no correlation of viscosity is available and if one wants to predict the viscosity of ethylbenzene, one has to rely on generic correlations^{3,4} that have invariably traded the range of applicability for accuracy. 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 ethylbenzene that is valid over a wide range of temperature and pressure, covering the vapor, liquid, and supercritical fluid states. Thus, it is a continuation of our recent work on viscosity of xylene isomers⁵⁻⁷ (*para*-, *meta*-, *ortho*-xylene) and is carried out under the auspices of the International Union of Pure and Applied Chemistry (IUPAC), as part of the program to develop representations of the viscosity and thermal conductivity of industrially important fluids that has so far produced viscosity correlations for: simple fluids,⁸⁻¹¹ water¹², normal alkanes¹³⁻²⁰ and cyclic and aromatic hydrocarbons.^{5-7,21-23}

2. Experimental Viscosity Data

Appendix A summarizes, to the best of our knowledge, the experimental measurements of the viscosity of ethylbenzene reported in the literature,²⁴⁻⁸⁷ 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 ethylbenzene were reported in 64 papers resulting in 684 data points. However, like for the other xylene isomers studied, most of the papers (58 papers, 233 data points) report only the value of liquid viscosity at atmospheric pressure, around room temperature, usually as part of a measurement program of viscosity of mixtures containing ethylbenzene. Appendix A also contains two reference works^{88,89} that report recommended tabulated values of the viscosity of ethylbenzene. 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, using well-established criteria⁹⁰ that have been widely disseminated.⁵⁻²³ Based on these criteria, 9 datasets were considered primary viscosity data. Table 1 summarizes the primary data^{25,26,31,36,40,41,54,69,87} detailing the temperature and pressure ranges, the uncertainty attributed to the measurements, authors 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 also contains the estimates of the uncertainty ascribed by the authors of the present work to each data set, following the analysis presented in section 3.

TABLE 1. Primary data used in developing the viscosity correlation of ethylbenzene

Authors	Year publ.	Technique employed ^a	Purity (%)	Uncertainty (%)	No. of data	Temperature range (K)	Pressure range (MPa)
Thorpe and Rodger ²⁵	1894	C	--	1.0	24	273-405	0.1
Thorpe and Rodger ²⁶	1897	C	--	1.0	22	273-405	0.1
Geist and Cannon ³¹	1946	C	--	0.5	3	273-313	0.1
Barlow <i>et al.</i> ³⁶	1966	C	--	1.0	10 ^b	178-303	0.1
Kashiwagi and Makita ⁴⁰	1982	TC	99	2.0	48	298-348	0.1-110
Abdullaev and Akhundov ⁴¹	1983	C	--	1.5	29	473-673	0.1-4.6
Krahn and Luft ⁵⁴	1994	RB	99	2.0	7 ^c	298-453	0.1-80
Yang <i>et al.</i> ⁶⁹	2005	C	99.3	1.0	7	298-353	0.1
Meng <i>et al.</i> ⁸⁷	2016	VW	99	2.0	117	253-373	0.1-35

^a C, capillary; TC, torsional crystal; VW, vibrating wire; RB, rolling body.

^b Data below triple point were excluded from the primary data sets.

^c Data above 110 MPa were excluded from the primary data sets.

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. The data is extensive in the liquid phase, but in the vapor phase only one dataset of measurements is available.

FIG. 1. Distribution of the available experimental viscosity data of ethylbenzene. Primary data: (■) Kashiwagi and Makita;⁴⁰ (◇) Abdullaev and Akhundov;⁴¹ (▼) Krahn and Luft;⁵⁴ (▲) Meng *et al.*;⁸⁷ (◆) data at 0.1 MPa;^{25,26,31,36,69} Secondary data: (+).

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 Zhou *et al.*¹ that covers the thermodynamic space from the triple point to 700 K, and up to 60 MPa. The uncertainties in density generated by this EoS range from 0.1% in the compressed-liquid region to 1.0% in the critical and vapor regions.¹ As some of the primary viscosity data has been measured at

pressures higher than 60 MPa, we made use of the Zhou *et al.*¹ EoS to estimate the densities for pressures as high as 110 MPa, like we did for *o*-xylene.⁷ The accuracy of the extrapolation was checked by comparison to the unpublished data⁹¹ measured in a vibrating tube densimeter with a typical uncertainty of 0.1%. Figure 2 illustrates that although there is a systematic trend the agreement between extrapolated and measured density data is within 0.15%, which is within the mutual uncertainty of the two sets of data.

FIG. 2. Percentage deviations $[100(\rho_{\text{exp}} - \rho_{\text{corr}}) / \rho_{\text{exp}}]$ of the unpublished experimental density data⁹¹ from the calculated values using EoS developed by Zhou *et al.*¹ (∇) 284 K; ($\square$) 323 K; ($\diamond$) 363 K.

3. Methodology and Analysis

It is customary⁹² 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.⁵⁻²³

3.1. The zero-density and initial-density terms

The situation for ethylbenzene is similar to that encountered with *m*-xylene⁶ and *o*-xylene⁷ that the paucity of data in the vapor phase does not allow for obtaining the zero-density and initial-density terms by fitting to the experimental data. Hence, we take the same approach as we took in developing the correlations for *m*- and *o*-xylene and we make use of the zero-density and initial density viscosity of *p*-xylene, developed earlier,⁵ to estimate $\eta_0(T)$ and $\eta_1(T)$ terms for ethylbenzene. We note that the low density correlation, $\eta_0(T) + \eta_1(T)\rho$, for *p*-xylene was based on the accurate and extensive data of Vogel and Hendl⁹³ that covered a temperature range (338 to 635) K and were measured in a quartz oscillating-disk viscometer with a claimed experimental uncertainty of 0.15 - 0.3%. The adjustment for ethylbenzene involved increasing the zero-density viscosity by 0.5% such that the low-density ethylbenzene data of Abdullaev and Akhundov⁴¹ is recaptured within its quoted uncertainty.

Figure 3, illustrates the deviations of the primary data of Abdullaev and Akhundov⁴¹ for two xylene isomers from their respective correlations. It is clear that the developed ethylbenzene correlation for $\eta_0(T) + \eta_1(T)\rho$ reproduces the available experimental data with the same uncertainty as was the case for *p*-xylene.

FIG. 3. Percentage deviations $[100(\eta_{\text{exp}} - \eta_{\text{corr}})/\eta_{\text{exp}}]$ of the available experimental data of Abdullaev and Akhundov⁴¹ in the vapor phase at 0.1 MPa. (■) *p*-xylene (▲) ethylbenzene.

For completeness we present the equations for the two terms and the relevant coefficients. The viscosity in the zero-density limit was represented using a practical engineering form as,⁵

$$\eta_0(T) = 1.005\eta_{0,p\text{-xylene}} = 0.22115 \frac{\sqrt{T}}{S_\eta} \quad (2)$$

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

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

where A_0 , B_0 and C_0 are the adjustable parameters and take the value of $A_0 = -1.4933$, $B_0 = 473.2$ K and $C_0 = -57033$ K².

The initial-density dependence is given by a simple empirical function,

$$\eta_1(T)\rho = \left(A_1 + \frac{B_1}{T} + \frac{C_1}{T^2}\right)\rho \quad (4)$$

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 $A_1 = 13.2814$ $\mu\text{Pa s mol}^{-1}$ l, $B_1 = -10862.4$ $\mu\text{Pa s K mol}^{-1}$ l and $C_1 = 1664060$ $\mu\text{Pa s K}^2 \text{ mol}^{-1}$ l.

Based on the agreement with the primary data and uncertainty associated with *p*-xylene correlation we ascribe uncertainty of 1% to the viscosity correlation in the vapor phase, below 0.2 MPa, in the temperature range (338 to 673) K. We do not recommend the use of Eqs (2) and (4) to predict the viscosity of the ethylbenzene vapor at temperatures below 338 K. The lack of experimental data and the empirical nature of the equations make the extrapolation rather uncertain. However, the use of Eqs (2) and (4), as part of Eq. (1), to predict the liquid viscosity from the triple point (178.2 K)¹ to 338 K is recommended since the contribution of low density terms to the overall liquid viscosity is small.

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 enhancement⁹⁴ that is significant only in a relatively narrow window in temperature and density round the critical point.^{8,14} Based on the previous studies,^{5-7,15-23} the viscosity critical enhancement of ethylbenzene is taken as zero. The total lack of industrial applications of ethylbenzene near its critical temperature and the existence of only a single experimental viscosity datum⁴¹ 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. Five sets of authors^{38,40,43,54,87} have measured the viscosity of ethylbenzene at a wide range of temperatures and at pressures higher than atmospheric, as illustrated in Fig. 1. Kashiwagi and Makita⁴⁰ performed the measurements in the temperature range (298 to 348 K) at pressures up to 110 MPa in a torsional crystal viscometer, while Meng *et al.*⁸⁷ used the vibrating wire viscometer in an extended temperature range (253 to 373 K), up to 35 MPa. Both sets of authors claimed uncertainty of 2% which is well-supported by their measurements on other fluids.^{5-7,21-23} Figure 4 illustrates that their data are mutually consistent, although Meng *et al.*⁸⁷ measurements are on average 1.5% higher than those of Kashiwagi and Makita.⁴⁰

FIG. 4. Comparison of the experimental liquid viscosity data at high pressure. ($\square$) Kashiwagi and Makita at 298 K;⁴⁰ ($\blacksquare$) Kashiwagi and Makita at 348 K;⁴⁰ (∇) Krahn and Luft at 298 K;⁵⁴ ($\blacktriangledown$) Krahn and Luft at 353 K;⁵⁴ ($\triangle$) Meng *et al.* at 303 K;⁸⁷ ($\blacktriangle$) Meng *et al.* at 353 K.⁸⁷

Krahn and Luft⁵⁴ performed the measurements in a rolling ball viscometer in the temperature range 298 to 453 K and at pressures up to 195 MPa. Although the rolling ball viscometer is not considered a primary instrument, the data of Krahn and Luft⁵⁴ are consistent with the data of Kashiwagi and Makita⁴⁰ and Meng *et al.*⁸⁷, as illustrated in Figure 4. Furthermore, Krahn and Luft⁵⁴ measurements of the viscosity of 2,2,4-trimethylpentane, in the same viscometer, agree with the values measured by Dymond *et al.*⁹⁵ within 2% over an extended pressure range (0.1-200 MPa). We have thus included in the primary dataset the 7 data points measured up to pressures of 110 MPa, which is the limit of the density extrapolation of Zhou *et al.*¹ EoS, as discussed in section 2. The inclusion of Krahn and Luft⁵⁴ measurements extends the temperature range of the primary ethylbenzene data to (298 to 453 K). It is worth noting that the measurements of Krahn and Luft⁵⁴ of more viscous fluids deviate from recommended values, as illustrated by the work on squalane.⁹⁶

The primary data set also contains five sets of viscosity measurements^{25,26,31,36,69} of liquid ethylbenzene at atmospheric pressure covering the temperature range (178-405) K. The choice of data followed our previous work on xylene isomers⁵⁻⁷ and was based on careful analysis of the available data deploying a number of criteria, as described previously.⁵⁻⁷ Three of the viscometers used to measure the primary ethylbenzene data solely at atmospheric pressure^{25,26,31,69} were also used to measure the viscosity of other xylene isomers that were included in the primary data set.⁵⁻⁷ We have also designated ethylbenzene data measured by Barlow *et al.*³⁶ in a capillary viscometer as primary, after analyzing the described workings of the viscometer and taking into account that their viscosity of toluene obtained in the same viscometer is reproduced with AAD of 1.4% by the current toluene viscosity correlation.²³ The data has the quoted uncertainty of 1.0% and covers the temperature range (178 to 303) K. Its inclusion in the primary data set allows the present correlation to be valid down to the triple point temperature. The primary data in the liquid state, thus covered the temperature range (178 to 453) K and pressures from 0.1 MPa up to 110 MPa. It is worth noting that

the upper temperature limit is much lower than for other xylene isomers. The only other data set that covers an extended range of pressure (0.1 to 60 MPa) and temperature (298 to 548 K) in the liquid state, and that could have been used to extend the temperature range of the primary data, has been measured by Akhundov³⁸ and Abdullaev and Akhundov⁴³ using the capillary viscometers. The comparison with the primary data at atmospheric pressure, as shown in Figure 5, indicates that the former is (6 to 10%) above, while the latter is up to 5% below, the data consensus.

FIG. 5. Comparison of the primary experimental liquid viscosity data measured at 0.1 MPa. (●) Primary data;^{26,31,36,40,54,69,87} (◆) Akhundov;³⁸ (▴) Abdullaev and Akhundov.⁴³

In the development of the recommended viscosity correlation for toluene²³ the authors used the viscosity data in the liquid state measured by Akhundov *et al.*,^{97,98} presumably in the same, if not similar viscometers, as primary. At temperatures above 480 K the data were correlated within 5%. Even if we assign 5% uncertainty to ethylbenzene data of Akhundov³⁸ and Abdullaev and Akhundov⁴³ it proved impossible to reconcile these data, either together or separately, with the other primary data sets. Hence the data of Akhundov³⁸ and Abdullaev and Akhundov⁴³ were classified as secondary for the purposes of developing the current correlation. Instead we have used the behavior of the excess viscosity, where the isotherm stratification should decrease as we enter the supercritical region,^{92,94} and the viscosity behavior of other three xylene isomers⁵⁻⁷ to guide the viscosity correlation in the temperature region (453 to 673 K) where no experimental data in the liquid state is available.

To complete the primary data set we have also included the data of Abdullaev and Akhundov⁴¹ measured in the vapor phase. The measurements, carried out in a different capillary viscometer, for that used for liquids, cover the temperature range (473 to 673) K and pressures up to 4.6 MPa. Good agreement of the viscosity data measured by the same authors in the same viscometer for *p*-xylene⁵ with that of Vogel and Hendl⁹³ indicates that the claimed uncertainty of 1.5% is justified.

In summary 267 data points covering the temperature range (178 to 673) K and pressures up to 110 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. (4). In line with previous work,^{5-7,21} 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,

$$\Delta\eta(\rho_r, T_r) = (\rho_r^{2/3} T_r^{1/2})f(\rho_r, T_r) + \exp(\rho_r^2) g(\rho_r, T_r) \quad (5)$$

The first term on the right-hand side of Eq. (5) takes advantage of the hard sphere result,^{99,100} as already used in correlating the viscosity of other fluids.^{5-7,22,23} The second term had to be introduced to account for a steep increase in viscosity observed at very low temperatures and high densities. The functions $f(\rho_r, T_r)$ and $g(\rho_r, T_r)$ are given by,

$$f(\rho_r, T_r) = (D_0 + E_0/T_r^{k_0})\rho_r^{n_0} + D_1\rho_r^{n_1} + E_2\rho_r^{n_2}/T_r^{k_2} + (D_3\rho_r + E_3T_r)\rho_r^{n_3} + D_4\rho_r^{n_4} \quad (6)$$

$$g(\rho_r, T_r) = \sum_{i=5}^{i=8} E_i\rho_r^{n_i}/T_r^{k_i} \quad (7)$$

Following the development of other correlations,^{5-7,21} 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 computing¹⁰¹ to fit primary data to Eqs. (5)-(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 (617.12 K) and critical density ρ_c (2.741016 mol l⁻¹) were obtained from Ref. 1.

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

i	D_i	n_i	E_i	k_i
0	-0.0376893	6.3	0.168877	1.1
1	17.9684	0.3	--	--
2	--	23.7	3.57702×10^{-11}	3.4
3	29.996	0.3	-8.00082	--
4	-25.7468	0.85	--	--
5	--	4.6	-3.29316×10^{-13}	20.8
6	--	11.1	-2.92665×10^{-13}	10.6
7	--	5.6	2.97768×10^{-13}	19.7
8	--	12.4	1.76186×10^{-18}	21.9

Figures 6-7 illustrate the percentage deviation of the primary viscosity data from the developed viscosity correlation, Eqs. (1)-(7).

FIG. 6. 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) - (7). (■) Kashiwagi and Makita;⁴⁰ (▼) Krahn and Luft;⁵⁴ (▲) Meng *et al.*⁸⁷

FIG. 7. 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) - (7). (◇) Thorpe and Rodger;²⁵ (○) Thorpe and Rodger;²⁶ (●) Geist and Cannon;³¹ (◄) Barlow *et al.*;³⁶ (■) Kashiwagi and Makita;⁴⁰ (▼) Krahn and Luft;⁵⁴ (▽) Yang *et al.*;⁶⁹ (▲) Meng *et al.*⁸⁷

Figure 6 illustrates the agreement with the experimental data in the liquid region for pressures higher than atmospheric. The experimental data of Krahn and Luft⁵⁴, Kashiwagi and Makita⁴⁰ and Meng *et al.*⁸⁷ are reproduced by the proposed correlation within their claimed experimental uncertainty of 2.0%. Figure 7 illustrates the agreement of the developed viscosity correlation with the primary

experimental data at atmospheric pressure that cover the temperature range (178 to 405) K, in the liquid phase. Most of the data are reproduced within 1.5%.

Table 3 summarizes the agreement between the primary experimental data and the proposed viscosity correlation for ethylbenzene in the liquid, dense vapor and supercritical regions. The correlation recaptures the entire set of primary data with an average absolute deviation (AAD) of 0.9%, bias of 0.3% and maximum deviation of -2.5%.

TABLE 3. Evaluation of the ethylbenzene viscosity correlation against the primary experimental data

Authors	Year publ.	AAD ^a (%)	Bias ^b (%)	MD ^c (%)
Thorpe and Rodger ²⁵	1894	0.7	-0.7	-1.0
Thorpe and Rodger ²⁶	1897	1.2	-1.2	-1.7
Geist and Cannon ³¹	1946	1.2	1.2	1.7
Barlow <i>et al.</i> ³⁶	1966	0.4	0.3	1.0
Kashiwagi and Makita ⁴⁰	1982	0.2	0.0	0.6
Abdullaev and Akhundov ⁴¹	1983	0.7	-0.1	-2.5
Krahn and Luft ⁵⁴	1994	0.7	-0.6	-1.9
Yang <i>et al.</i> ⁶⁹	2005	0.5	0.4	0.9
Meng <i>et al.</i> ⁸⁷	2016	1.2	0.9	-1.9
Entire primary data set		0.9	0.3	-2.5

^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.

4. Overall Viscosity Correlation

The viscosity correlation of ethylbenzene 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 (178 to 673 K) and pressure (up to 110 MPa) range. In the vapor phase the lower temperature limit corresponds to 338 K, while in the liquid phase, in the temperature range (178 to 250 K), the upper pressure limit is 0.1 MPa. 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 (178.2 K)¹ nor when extrapolated in the liquid phase to pressures as high as 110 MPa in the temperature range (178 to 250 K).

Figure 8 illustrates the behavior of the viscosity correlation as a function of density along the 300 and 600 K isotherms. We observe a 190 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 1.3 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 hydrocarbons⁹² or as part of the VW model¹⁰²⁻¹⁰⁴ to predict the viscosity of mixtures containing ethylbenzene.

FIG. 8. Viscosity of ethylbenzene 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) at atmospheric pressure we estimate the uncertainty to be 1.0% and 1.5%, in the vapor and liquid phase, respectively; (ii) in the liquid region for pressures above atmospheric and temperature in the range (253 to 453 K), we estimate the uncertainty to be 2.0%; (iii) in the high pressure vapor and supercritical region, we estimate the uncertainty to be 2.5%; (iv) at other temperatures and pressures where no primary experimental data are available we conservatively estimate the uncertainty to be 5% based on analysis of secondary data and our previous work on *p*-, *m*- and *o*-xylene.⁵⁻⁷ Figure 9 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 ethylbenzene 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 ethylbenzene along the saturation line.

FIG. 9. The extent of the viscosity representation and its estimated uncertainty. No representation is available in the hatched region.

TABLE 4. Recommended viscosity values in $\mu\text{Pa s}$

P	T / K											
MPa	200	250	280	300	320	360	400	450	500	550	600	650
0	--	--	--	--	--	7.79	8.62	9.67	10.73	11.79	12.84	13.87
0.1	5215.7	1299.2	797.1	616.8	495.5	343.1	250.3	9.70	10.78	11.85	12.91	13.95
0.5	--	1303.6	799.7	618.7	497.0	344.4	251.4	175.4	10.89	12.02	13.12	14.18
1	--	1309.1	802.8	621.1	499.0	345.9	252.7	176.7	125.1	12.19	13.34	14.44
2	--	1320.1	809.1	625.9	502.9	348.9	255.4	179.3	127.9	89.51	13.88	14.99
4	--	1342.4	821.8	635.5	510.7	354.9	260.8	184.5	133.4	96.30	63.60	18.03
6	--	1364.9	834.6	645.2	518.5	360.9	266.0	189.4	138.5	102.2	73.26	41.67
8	--	1387.6	847.5	655.0	526.4	366.8	271.2	194.3	143.3	107.6	80.30	56.06
10	--	1410.6	860.5	664.8	534.2	372.8	276.3	199.0	148.0	112.5	86.18	64.61
20	--	1529.6	927.3	714.6	573.9	402.1	301.1	221.4	169.4	133.8	108.6	90.08
50	--	1928.8	1146.6	875.3	698.7	489.3	371.4	281.1	223.2	183.7	155.9	135.9
110	--	2948.3	1690.3	1191.1	989.1	675.8	509.9	389.8	315.5	265.3	229.7	203.5

T/K	P_V/MPa	Vapor		Liquid	
		$\rho/(\text{mol l}^{-1})$	$\eta/(\mu\text{Pa s})$	$\rho/(\text{mol l}^{-1})$	$\eta/(\mu\text{Pa s})$
273.15	0.0003	0.0001	--	8.3287	879.4
293.15	0.0010	0.0004	--	8.1649	669.6
313.15	0.0029	0.0011	--	7.9999	531.8
333.15	0.0074	0.0027	--	7.8329	435.1
353.15	0.0168	0.0058	7.65	7.6630	363.4
373.15	0.0343	0.0113	8.06	7.4891	307.9
393.15	0.0643	0.0203	8.48	7.3104	263.5
413.15	0.1122	0.0343	8.91	7.1256	227.1
433.15	0.1845	0.0549	9.34	6.9332	196.7
453.15	0.2884	0.0842	9.79	6.7317	171.0
473.15	0.4319	0.1247	10.26	6.5188	149.0
493.15	0.6238	0.1797	10.75	6.2918	130.1
513.15	0.8736	0.2539	11.27	6.0464	113.6
533.15	1.1917	0.3542	11.84	5.7764	98.96
553.15	1.5899	0.4922	12.52	5.4711	85.66

- 15 -

Figure 11 exhibits the only three data sets of secondary experimental data that extend to higher pressure. The data of Krahn and Luft⁵⁴ illustrated in Fig. 11 correspond to measurements at pressures higher than 110 MPa, outside the range of validity of Zhou *et al.* EoS. At pressures of 120 MPa the deviations are still within 2-3%, while at 200 MPa they increase to as high as 9%. Thus giving some credence that the correlation extrapolates in a well-behaved fashion to higher pressures. The two data sets measured by Akhundov and co-worker^{38,43} are not only inconsistent with each other, but also with the developed correlation both in the temperature range where the primary data are available (288 to 448 K) and at higher temperatures, as illustrated in Fig. 11. The systematic deviations extend over the whole pressure range.

FIG. 11. Percentage deviations $[100(\eta_{\text{exp}} - \eta_{\text{corr}})/\eta_{\text{exp}}]$ of the selected secondary experimental viscosity data at high pressures from the calculated values using Eqs. (1) - (7). (◆) Akhundov (302 to 448 K);³⁸ (◇) Akhundov (473 to 548 K);³⁸ (►) Abdullaev and Akhundov (288 to 448 K);⁴³ (▷) Abdullaev and Akhundov (473 to 548 K);⁴³ (▼) Krahn and Luft.⁵⁴

Although no other viscosity correlation of ethylbenzene is available in the open literature there are a couple of tables of recommended values^{88,89} and Yaws recommended equation⁴ all for liquid viscosity at atmospheric pressure. The agreement between the tabulated values of Golubev,⁸⁸ NIST/TRC database⁸⁹ and Yaws⁴ and the present correlation is very good and the deviations do not exceed $\pm 1.3\%$, except for one data point of Golubev.⁸⁸

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 (μ Pa s)
250	8.9814	2948.109
300	8.1093	616.814
300	8.4082	875.361
300	8.6762	1262.986
350	0.000	7.591
400	0.000	8.616
400	7.2481	250.283
400	8.1196	509.868
500	0.000	10.734
500	0.020	10.776
600	0.000	12.841
600	6.4831	155.940
600	7.1427	229.686

6. Comparison of viscosity of xylene isomers

The development of the viscosity correlation for ethylbenzene, presented in this paper, complements our work on viscosity correlations of the other three xylene isomers (*p*-, *m*-, *o*-xylene)⁵⁻⁷. Figures 12 and 13 illustrate the behavior of the viscosity of four xylene isomers along two different isotherms. In general, in the liquid phase, the differences, at the same density and temperature, do not exceed 8-13%. At atmospheric pressure *o*-xylene displays by far the highest viscosity, as illustrated in Fig. 14, being approximately 30% more viscous than the other three isomers at 293 K. The higher viscosity is a direct result of higher density of liquid *o*-xylene at atmospheric conditions. We have limited the comparison to the liquid phase only, as the development of the viscosity of correlation in the vapor phase of ethylbenzene, *m*-xylene and *o*-xylene was based on experimental measurements of *p*-xylene and differences between the viscosities of four isomers are of the order of 1%.

FIG. 12. Viscosity of four xylene isomers as a function of density at 300 K: (—) *p*-xylene;⁵ (- - -) *m*-xylene;⁶ (••••) *o*-xylene;⁷ (• - - •) ethylbenzene.

FIG. 13. Viscosity of four xylene isomers as a function of density at 450 K: (—) *p*-xylene;⁵ (- - -) *m*-xylene;⁶ (••••) *o*-xylene;⁷ (• - - •) ethylbenzene.

FIG. 14. Viscosity of four xylene isomers as a function of temperature at 0.1 MPa: (—) *p*-xylene;⁵ (- - -) *m*-xylene;⁶ (••••) *o*-xylene;⁷ (• - - •) ethylbenzene.

In performing the comparison between the viscosities of different xylene isomers we have observed that the viscosity of *p*-xylene exhibits less smooth behavior at densities corresponding to the two phase region, compared to the other xylene isomers. As there are no data to guide the correlation at these densities, the behavior is determined by the form of the fitting equation, which for *p*-xylene was different to that used subsequently for the other three xylene isomers. We have thus re-fitted the primary data for *p*-xylene using the same residual viscosity representation as that for *m*- and *o*-xylene. Figure 15 illustrates the behavior of the viscosity of *p*-xylene at 473 K using the published⁵ and the revised formulation.

FIG. 15. Viscosity of *p*-xylene as a function of density at 473 K: (- - -) published correlation;⁵ (—) new correlation.

At densities lower than vapor saturation density ($\rho \leq 0.1243 \text{ mol l}^{-1}$) and higher than liquid saturation density ($\rho \geq 6.5205 \text{ mol l}^{-1}$) the difference between the two formulations is minimal. In fact at temperatures below the highest temperature at which the high pressure experimental data are available ($T = 548 \text{ K}$) the two formulations reproduce viscosity for the most part within deviations of less than 1%, except at the highest pressures where maximum deviation of 1.8% is observed. However, at higher temperatures the less smooth behavior of the published correlation⁵ in the two phase region manifests itself at supercritical density, as illustrated in Fig 16 for 673 K isotherm. It is clear that the two correlations produce very different values of viscosity, highlighting that extrapolation to regions where there are no experimental data is fraught with difficulties. The lack of a physical basis for all the functions used in the literature⁵⁻²³ to represent the residual viscosity contribution makes the issue of extrapolation rather acute and indicates that the estimated uncertainty, quoted for the ranges of temperature and pressure where there are no experimental data, might be too optimistic.

FIG. 16. Viscosity of *p*-xylene as a function of density at 673 K: (---) published correlation;⁵ (—) new correlation.

As the new correlation for *p*-xylene exhibits smoother behavior and the values of viscosity at high temperature are compatible with the viscosity values for the other three xylene isomers, we recommend its use instead of the published one.⁵ For this purpose we have collated the relevant information in Appendix B.

7. Conclusion

A new wide-ranging correlation for the viscosity of ethylbenzene has been developed based on critically-evaluated experimental data. The correlation is valid to pressures up to 110 MPa and temperatures up to 673 K. In the liquid part of the phase diagram the lower temperature limit is 178 K, while in the vapor part of the phase diagram it is 338 K. The correlation is expressed in terms of temperature and density, and the densities were obtained from the equation of state of Zhou *et al.*¹ The overall uncertainty, using a coverage factor of 2, of the proposed correlation is less than 5.0%, however this uncertainty varies depending on the thermodynamic state and is summarized in more detail in Fig. 9.

Comparison of viscosity of xylene isomers indicated that at very high temperatures the viscosity correlation of *para*-xylene has higher uncertainty than previously postulated. Thus, in this work we also provide a revised viscosity correlation for *p*-xylene.

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8. Appendix A: Viscosity measurements of ethylbenzene

Table 7. Viscosity measurements of ethylbenzene

Authors	Year publ.	Technique employed ^a	No. of data	Temperature range (K)	Pressure range (MPa)
Gartenmeister ²⁴	1890	--	1	293	0.1
Thorpe and Rodger ²⁵	1894	C	24	274-405	0.1
Thorpe and Rodger ²⁶	1897	C	22	273-405	0.1
Batschinski ²⁷	1913	C	14	273-403	0.1
Dunstan <i>et al.</i> ²⁸	1913	C	1	298	0.1
Timmermans and Martin ²⁹	1926	--	3	288-304	0.1
Schmidt <i>et al.</i> ³⁰	1939	--	7	283-343	0.1
Geist and Cannon ³¹	1946	C	3	273-313	0.1
Koelbel <i>et al.</i> ³²	1949	--	5	303-393	0.1
Mamedov and Panchenkov ³³	1955	C	11	253-253	0.1
Petro and Smyth ³⁴	1957	C	3	293-333	0.1
Panchenkov and Erchenkov ³⁵	1962	--	6	283-353	0.1
Barlow <i>et al.</i> ³⁶	1966	C	15	160-303	0.1
Blank ³⁷	1968	--	1	298	0.1
Akhundov ³⁸	1973	C	131	302-548	0.1-40
Shikhaliev ³⁹	1977	--	1	293	0.1
Kashiwagi and Makita ⁴⁰	1982	TC	48	298-348	0.1-110
Abdullaev and Akhundov ⁴¹	1983	C	29	473-673	0.1-4.6
Oswal and Rathnam ⁴²	1984	C	1	303	0.1
Abdullaev and Akhundov ⁴³	1985	C	110	288-548	0.1-60
Al-Madfai <i>et al.</i> ⁴⁴	1985	C	1	298	0.1
Plaskunova <i>et al.</i> ⁴⁵	1986	--	11	223-353	0.1
Dewan <i>et al.</i> ⁴⁶	1988	C	1	303	0.1
Fermeglia and Lapasin ⁴⁷	1988	C	1	298	0.1
Subha and Rao ⁴⁸	1988	C	1	308	0.1
Rattan <i>et al.</i> ⁴⁹	1989	C	2	298-308	0.1
Singh <i>et al.</i> ⁵⁰	1989	C	5	298-333	0.1
Asfour <i>et al.</i> ⁵¹	1990	C	2	293-298	0.1
Schumpe and Luhning ⁵²	1990	C	1	293	0.1
Vavanellos <i>et al.</i> ⁵³	1991	C	2	308-313	0.1
Krahn and Luft ⁵⁴	1994	RB	16	298-453	0.1-195
Chen <i>et al.</i> ⁵⁵	1995	C	2	313-333	0.1
Ramadevi <i>et al.</i> ⁵⁶	1996	C	2	303-313	0.1
Nhaesi and Asfour ⁵⁷	2000	C	2	293-298	0.1
George and Sastry ⁵⁸	2003	C	2	298-308	0.1
Katyal <i>et al.</i> ⁵⁹	2003	C	3	293-313	0.1
Lark <i>et al.</i> ⁶⁰	2003	C	2	298-303	0.1
Oswal <i>et al.</i> ⁶¹	2003	C	1	303	0.1
Oswal <i>et al.</i> ⁶²	2004	C	1	303	0.1
Rattan <i>et al.</i> ⁶³	2004	C	2	298-308	0.1

Yang <i>et al.</i> ⁶⁴	2004	C	3	298-323	0.1
Yang <i>et al.</i> ⁶⁵	2004	C	5	298-333	0.1
Knothe and Steidley ⁶⁶	2005	C	1	313	0.1
Nhaesi and Asfour ⁶⁷	2005	C	2	308-313	0.1
Rathnam <i>et al.</i> ⁶⁸	2005	C	2	303-313	0.1
Yang <i>et al.</i> ⁶⁹	2005	C	7	298-353	0.1
Al Gherwi <i>et al.</i> ⁷⁰	2006	C	2	303-313	0.1
Al-Kandary <i>et al.</i> ⁷¹	2006	C	2	293-303	0.1
Baskaran and Kubendran ⁷²	2008	--	3	303-323	0.1
Song <i>et al.</i> ⁷³	2008	C	7	303-333	0.1
Wankhede <i>et al.</i> ⁷⁴	2008	C	3	288-308	0.1
Dominguez-Perez <i>et al.</i> ⁷⁵	2009	C	1	298	0.1
El-Sayed and Asfour ⁷⁶	2009	C	2	308-313	0.1
Rathnam <i>et al.</i> ⁷⁷	2009	C	2	303-313	0.1
Ikeuchi <i>et al.</i> ⁷⁸	2010	C	6	283-308	0.1
Rathnam <i>et al.</i> ⁷⁹	2010	C	2	303-313	0.1
Bhatia <i>et al.</i> ⁸⁰	2011	C	2	298-308	0.1
Hasan <i>et al.</i> ⁸¹	2011	C	2	298-308	0.1
Rathnam <i>et al.</i> ⁸²	2011	C	2	303-313	0.1
Saravanakumar <i>et al.</i> ⁸³	2012	--	1	303	0.1
Barega <i>et al.</i> ⁸⁴	2013	C	4	298-313	0.1
El-Sayed and Asfour ⁸⁵	2013	C	2	293-298	0.1
Prak <i>et al.</i> ⁸⁶	2014	RC	8	293-373	0.1
Meng <i>et al.</i> ⁸⁷	2016	VW	117	253-373	0.1-35

Tables of collected data

Golubev ⁸⁸	1970	--	14	273-413	0.1
NIST/TRC database 85 ⁸⁹	2003	--	29	248-413	0.1

^a C, capillary; TC, torsional crystal; VW, vibrating wire; RB, rolling body; RC, rotating cylinder.

9. Appendix B: Revised Reference correlation of the viscosity of *p*-xylene from triple point to 673 K and up to 110 MPa

Appendix B contains the new residual viscosity function, that replaces the function given by Eq. (6) in the original ref 5. Table 8 summarizes the optimal coefficients D_i , n_i , E_i and k_i that were obtained by fitting to the same primary data used in the original development.⁵ Table 9 summarizes the agreement between the primary experimental data and the revised viscosity correlation for *p*-xylene. For completeness we have included Tables 10-11 that contain the recommended values of viscosity at selected temperatures and pressures and along the saturation lines. Table 12 is provided to assist users in computer- program verification of the revised correlation.

$$\Delta\eta(\rho_r, T_r) = (\rho_r^{2/3} T_r^{1/2})f(\rho_r, T_r) \quad (8)$$

$$f(\rho_r, T_r) = (D_0 + E_0/T_r^{k_0})\rho_r^{n_0} + D_1\rho_r^{n_1} + E_2\rho_r^{n_2}/T_r^{k_2} + (D_3\rho_r + E_3T_r)\rho_r^{n_3} + D_4\rho_r^{n_4} \quad (9)$$

TABLE 8. Coefficients for the representation of the residual viscosity of *p*-xylene, Eq. (9)

i	D_i	n_i	E_i	k_i
0	-0.0258436	8.3	0.0323284	0.7
1	15.4854	0.6	--	--
2	--	23.5	4.30133×10 ⁻¹¹	3
3	13.8162	1.2	-19.9037	--
4	-0.000112615	11	--	--

TABLE 9. Evaluation of the revised *p*-xylene viscosity correlation against the primary experimental data

Authors	Year publ.	AAD (%)	Bias (%)	MD (%)
Mamedov and Panchenkov ³³	1955	0.7	0.7	2.1
Mamedov <i>et al.</i> ¹⁰⁵	1968	0.8	-0.5	2.1
Nissema and Koskenniska ¹⁰⁶	1972	0.2	0.2	0.6
Mamedov <i>et al.</i> ¹⁰⁷	1975	0.7	-0.6	-2.2
Kashiwagi and Makita ⁴⁰	1982	0.5	0.3	1.7
Abdullaev and Akhundov ⁴¹	1983	0.8	0	2.2
Dymond and Robertson ¹⁰⁸	1985	0.6	-0.3	1
Serrano <i>et al.</i> ¹⁰⁹	1990	0.6	0.6	0.7
Vogel and Hendl ¹¹⁰	1992	0.3	-0.3	-0.7
Et-Tahir <i>et al.</i> ¹¹¹	1995	1.6	-0.3	-2.8
Exarchos <i>et al.</i> ¹¹²	1995	0.6	-0.6	-1
Entire primary data set		0.7	0.3	-2.8

TABLE 10. Recommended viscosity values for *p*-xylene in $\mu\text{Pa s}$

<i>P</i> MPa	<i>T</i> / 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	671.0	593.5	529.4	475.7	430.1	391.0	357.2	301.5	10.02	12.17	14.25
0.5	673.2	595.4	531.1	477.2	431.5	392.3	358.4	302.7	147.5	12.30	14.47
1	676.0	597.9	533.3	479.2	433.3	394.0	360.0	304.1	148.7	12.53	14.78
2	681.5	602.7	537.6	483.1	436.9	397.3	363.1	306.8	151.1	68.72	15.47
4	692.6	612.5	546.4	491.0	444.1	404.0	369.3	312.4	155.8	77.34	17.25
6	703.8	622.4	555.1	498.9	451.3	410.6	375.5	317.9	160.3	83.95	24.46
8	715.1	632.3	563.9	506.8	458.5	417.3	381.6	323.4	164.6	89.55	36.91
10	726.4	642.2	572.8	514.8	465.8	423.9	387.8	328.8	168.7	94.51	45.47
30	845.2	745.8	664.2	596.4	539.5	491.1	449.7	382.5	205.7	130.2	86.92
50	974.6	857.7	762.1	683.0	616.8	560.9	513.1	436.3	238.3	156.5	111.2
70	1116.1	979.4	867.8	775.8	699.0	634.4	579.5	491.6	269.0	179.2	130.6
90	1271.4	1112.2	982.6	875.9	787.3	712.8	649.7	549.2	298.8	200.0	147.3
110	1441.7	1257.4	1107.6	984.5	882.4	796.8	724.5	609.9	328.3	219.6	162.5

TABLE 11. Recommended viscosity values for *p*-xylene along the saturation line

<i>T</i> /K	<i>P_v</i> /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	644.4
303.15	0.0016	0.0006	--	8.0283	571.6
313.15	0.0027	0.0010	--	7.9462	511.0
323.15	0.0043	0.0016	--	7.8637	460.1
333.15	0.0069	0.0025	--	7.7805	416.8
343.15	0.0105	0.0037	7.40	7.6966	379.6
353.15	0.0156	0.0054	7.60	7.6118	347.2
403.15	0.0807	0.0251	8.58	7.1702	233.5
453.15	0.2746	0.0803	9.62	6.6834	164.9
503.15	0.7131	0.2073	10.82	6.1182	117.6
553.15	1.5487	0.4883	12.44	5.3990	80.31

TABLE 12. Sample points for computer verification of the correlating equations

<i>T</i> (K)	ρ (mol l ⁻¹)	η (μ Pa s)
300	0	6.604
300	0.049	6.450
300	8.0548	593.513
300	8.6309	1257.494
400	0	8.573
400	7.1995	239.081
400	8.0735	488.777
600	0	12.777
600	7.0985	199.160

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