

Viscosity Correlation for Isobutane over Wide Ranges of the Fluid Region¹

E. Vogel,^{2,3} C. Küchenmeister,² and E. Bich²

A new representation of the viscosity of isobutane has been developed. The representative equations include zero-density and initial-density dependence correlations. The higher density contributions to the residual viscosity are formed by a combination of double polynomials in density and reciprocal temperature and of a free-volume term with a temperature-dependent close-packed density. The new full surface correlation is based on primary experimental data sets selected as a result of a critical assessment of the available information. The validity of the representation extends from the triple point to 600 K and 35 MPa in accordance with the modified Benedict–Webb–Rubin equation of state by Younglove and Ely (1987). The uncertainty of the representation varies from $\pm 0.4\%$ in the dilute gas phase between room temperature and 600 K to $\pm 3\%$ in the thermodynamic ranges in which the equation of state is valid as well as where primary experimental data are available.

KEY WORDS: critical data assessment; free-volume model; isobutane; liquid viscosity; vapor viscosity; viscosity correlation.

1. INTRODUCTION

In a research program initiated under the auspices of IUPAC, consistent and theoretically sound correlations for the transport properties are to be developed for fluids of industrial importance. Since the theory of the transport properties of fluids is not completely worked out, a state-of-the-art representation corresponds to a correlation which is based on a critical

¹ Paper presented at the Fifth Asian Thermophysical Properties Conference, August 30–September 2, 1998, Seoul, Korea.

² Fachbereich Chemie, Universität Rostock, D-18051 Rostock, Germany.

³ To whom correspondence should be addressed.

analysis of the available experimental data and is developed under the guidance of the kinetic theory. The viscosity surfaces of ethane [1], propane [2, 3], and *n*-butane [4] were studied in the series of *n*-alkanes. The program should also be extended to the series of isoalkanes in which isobutane as a liquefied gas and environmentally friendly refrigerant represents a commercially important fluid. Its thermophysical properties should reliably be known over temperature and pressure ranges as wide as possible.

In 1979 Stephan and Lucas [5] generated a table of the viscosity of isobutane in the temperature range from 310 to 850 K between 0.1 and 50 MPa using recommended values by Gonzalez and Lee [6] between 311 and 511 K up to pressures of 55 MPa. The experimental data by Gonzalez and Lee [7] were actually restricted up to 444 K and down to 0.7 MPa. Their values at atmospheric pressure [6] represent an average of rather unreliable experimental data between 308 and 378 K and of values extrapolated with semiempirical correlations. That means the table by Stephan and Lucas corresponds to an extrapolation from 378 to 850 K at atmospheric pressure and from 444 to 850 K in the range of higher pressures.

In 1987 Younglove and Ely [8] reported a representation of the viscosity of isobutane from the triple point to 600 K and 35 MPa based on the modified Benedict–Webb–Rubin (MBWR) equation of state. This correlation suffers from lack of information about the selection and analysis of the data base and from little indication of the development of the viscosity formulation. It includes recent experimental data by Diller and Van Poolen [9] in the saturated and compressed liquid regions down to 115 K. But it is unclear whether the data by Agaev and Yusibova [10] between 273 and 548 K up to pressures of 69 MPa were considered.

In the present paper a new correlation is proposed specifically designed to take into account the steep increase of viscosity at low temperatures and high densities as well as a continuous increase in the two-phase region. The smooth behavior in the two-phase region is of importance for the prediction of the viscosity of fluid mixtures using the corresponding-states principle. The suitability of such a correlation has already been proven for propane [3] and *n*-butane [4]. The contributions to the viscosity in the low-density region reported separately [11] have been based on new results of high-precision measurements with an oscillating-disk viscometer up to 627 K and on an improved way to predict data in the vapor phase at moderately low densities. The analysis of the contributions in the high-density region has been based on a critical assessment of the available experimental data. It shows that the representative equations describe the selected primary data sets within their ascribed uncertainties.

2. METHODOLOGY

The representation of the viscosity is based on the residual viscosity concept. The viscosity $\eta(\rho, T)$ of a pure fluid at density ρ and temperature T may be considered as composed of four contributions [3, 12]:

$$\eta(\rho, T) = \eta^{(0)}(T) + \eta^{(1)}(T) \rho + \Delta\eta_h(\rho, T) + \Delta\eta_c(\rho, T) \quad (1)$$

where $\eta^{(0)}(T)$ is the viscosity in the limit of zero density, the sum of $\eta^{(1)}(T) \rho$ and $\Delta\eta_h(\rho, T)$ represents the residual viscosity, and $\Delta\eta_c(\rho, T)$ corresponds to the critical enhancement of viscosity in the immediate vicinity of the vapor-liquid critical point. The procedure enables an independent analysis of each contribution using the most recent advances of the kinetic theory as a guidance in the development of the viscosity surface correlation.

Since the correlations of the zero-density and the initial-density viscosity coefficients have separately been treated [11], a short summary is all that is necessary here. The kinetic theory of dilute gases [13] may be used to relate the viscosity of a pure polyatomic gas to an effective collision cross section which contains all the dynamic and statistical information about the binary collisions. Then, the relationship is formally identical to that of monatomic gases and is written in practical engineering form [14] as

$$\eta^{(0)}(T) = \frac{0.021357(MT)^{1/2}}{\sigma^2 \mathfrak{S}_\eta^*(T^*)} \quad (2)$$

$$\mathfrak{S}_\eta^*(T^*) = \exp \left[\sum_{i=0}^4 a_i (\ln T^*)^i \right] \quad (3)$$

$$T^* = k_B T / \varepsilon \quad (4)$$

\mathfrak{S}_η^* represents a reduced effective cross section for which the dependence on the reduced temperature T^* is given by Eq. (3). When the molar mass M is in $\text{g} \cdot \text{mol}^{-1}$, $\eta^{(0)}$ is in units of $\mu\text{Pa} \cdot \text{s}$. The length scaling factor σ is in nm, and the energy scaling factor ε/k_B is in K. It has been found that the primary experimental viscosity data can only be represented within their ascribed experimental uncertainties by means of an individual correlation because polyatomic molecules like isobutane do not conform to the corresponding states principle for the monatomic species. Adopting the same scaling factors deduced in the analysis of the initial-density viscosity coefficient (see below), the coefficients a_i of the functional \mathfrak{S}_η^* were determined by fitting Eqs. (2)–(4) to the experimental $\eta^{(0)}$ values.

The Rainwater–Friend theory [15–18] can be used to establish separately the linear-in-density term $\eta^{(1)}\rho$ and the temperature function of the second viscosity virial coefficient $B_\eta(T)$:

$$B_\eta(T) = \frac{\eta^{(1)}(T)}{\eta^{(0)}(T)} \quad (5)$$

Recently, Vogel et al. [3] recommended an improved empirical equation for the reduced second viscosity virial coefficient B_η^* as a function of the reduced temperature T^* in the range $0.5 \leq T^* \leq 100$ which can be reliably extrapolated down to $T^* \approx 0.3$:

$$B_\eta^*(T^*) = \frac{B_\eta(T)}{0.6022137\sigma^3} = \sum_{i=0}^6 b_i T^{*-0.25i} + b_7 T^{*-2.5} + b_8 T^{*-5.5} \quad (6)$$

Here, B_η is in units of $\text{L} \cdot \text{mol}^{-1}$. Küchenmeister and Vogel [11] have shown that the initial density dependence of isobutane can be reasonably well represented by means of an individual correlation. They have determined optimized scaling factors σ and ε/k_B by fitting Eqs. (4)–(6) to their experimentally determined $B_\eta(T)$ values.

The coefficients a_i [Eq. (3)] and b_i [Eq. (6)] of both individual correlations as well as the scaling factors σ and ε/k_B are summarized in Table I.

In principle, both the thermal conductivity and viscosity of pure fluids diverge at the vapor–liquid critical point due to long-range fluctuations, but the enhancement in viscosity is small and the ratio $\Delta\eta_c(\rho, T)/\eta(\rho, T) > 0.01$ only within 1% of the critical temperature of the fluid [1, 19]. Furthermore, measurements near the critical point are often influenced by experimental difficulties inherent in the technique used. The analysis of the experimental data for isobutane has shown that there are no data to be used to develop a surface correlation that includes the critical enhancement of the viscosity.

The theoretical guidance to the functional form of the higher terms of the residual viscosity $\Delta\eta_h(\rho, T)$ is rather weak. Thus, a correlation with double polynomials in density and reciprocal temperature was successfully utilized for ethane [1, 14]. In this procedure a structural optimization was carried out to determine the most suitable combination from a large number of the double polynomials using the SEEQ algorithm based on the stepwise linear least-squares technique. But in the case of propane, such a correlation proved unsuitable to describe appropriately the steep viscosity increase at high densities and low temperatures. An attempt by Vogel and Küchenmeister [2] to include additional exponential terms for this

Table I. Coefficients for the Representation of the Viscosity of Isobutane

Molar mass

$$M = 58.1222 \text{ g} \cdot \text{mol}^{-1}$$

Critical constants

$$T_c = 407.817 \text{ K}, \rho_c = 3.860 \text{ mol} \cdot \text{L}^{-1}$$

Scaling factors

$$\varepsilon/k_B = 307.55 \text{ K}, \sigma = 0.46445 \text{ nm}$$

Coefficients a_i for Ξ_η^* according to Eq. (3)

$$a_0 = 0.53583008 \quad a_1 = -0.45629630$$

$$a_2 = 0.049911282 \quad a_3 = 0$$

$$a_4 = 0$$

Coefficients b_i for B_η^* according to Eq. (6)

$$b_0 = -19.572881 \quad b_1 = 219.73999$$

$$b_2 = -1015.3226 \quad b_3 = 2471.01251$$

$$b_4 = -3375.1717 \quad b_5 = 2491.6597$$

$$b_6 = -787.26086 \quad b_7 = 14.085455$$

$$b_8 = -0.34664158$$

Coefficients e_{ij} , f_1 , and g_l for $\Delta\eta_h$ according to Eqs. (7) and (8)

$$e_{20} = 103.511763411 \quad e_{21} = -312.670896234$$

$$e_{22} = 145.253750239 \quad e_{30} = -210.649894193$$

$$e_{31} = 386.269696509 \quad e_{32} = -214.963015527$$

$$e_{40} = 112.580360920 \quad e_{41} = -223.242033154$$

$$e_{42} = 119.114788598 \quad e_{50} = -18.1909745900$$

$$e_{51} = 36.0438957232 \quad e_{52} = -21.3960184050$$

$$f_1 = 1940.37606990$$

$$g_1 = 2.33859774637 \quad g_2 = 1.00596672174$$

increase also turned out to be inadequate since the correlation was characterized by large loops in the two-phase region at low temperatures. In this case problems arise when using propane as a reference fluid to predict the viscosity of other fluids or fluid mixtures within the scope of the extended corresponding states principle. Finally, Vogel et al. [3] proposed to test a combination of double polynomials in reduced density and reduced reciprocal temperature and of a modified Batschinski-Hildebrand term with a temperature-dependent reduced close-packed density:

$$\Delta\eta_h(\delta, \tau) = \sum_{i=2}^n \sum_{j=0}^m e_{ij} \frac{\delta^i}{\tau^j} + f_1 \left[\frac{\delta}{\delta_0(\tau) - \delta} - \frac{\delta}{\delta_0(\tau)} \right] \quad (7)$$

$$\delta_0(\tau) = g_1 \left[1 + \sum_{l=2}^{n_l} g_l \tau^{(l-1)/2} \right] \quad (8)$$

$$\delta = \rho/\rho_c, \quad \tau = T/T_c \quad (9)$$

Here, ρ_c is the critical density and T_c is the critical temperature. The higher density contribution to the residual viscosity was deduced from each experimental datum by means of Eq. (1) subtracting the zero-density viscosity $\eta^{(0)}$ and the linear-in-density term $\eta^{(1)}\rho$.

The NIST package ODRPACK by Boggs et al. [20] based on a non-linear least-squares regression was used to determine the parameters and their significance in Eqs. (7) and (8). For propane [3] and also for *n*-butane [4], it turned out that the higher terms in the temperature function $\delta_0(\tau)$ as well as the number of double polynomials had to be restricted because the uncertainties of the experimental data are too large.

The viscosity surface of isobutane was expected to be represented appropriately by a correlation nearly equivalent to that of propane since the viscosity rise in the liquid phase at low temperatures and high densities is similarly strong.

Primary data sets have to be selected from the experimental data in the literature to develop an accurate and consistent representation for the viscosity. To qualify for the primary class, the data should have been measured with a high-precision instrument for which a full working equation is available and all necessary corrections can be applied. Then, a relative uncertainty $\delta\eta_{r,k}$ is ascribed to the selected experimental viscosity value $\eta_{\text{exp},k}$ according to the measurement method used, the quoted experimental error, the check on precision of the data, and the discrepancies with other independent data in overlapping thermodynamic ranges. The appropriate statistical weight w_k of the value k needed in the development of the viscosity correlation follows from

$$w_k = \left(\frac{100}{\eta_{\text{exp},k} \delta\eta_{r,k}} \right)^2 \quad (10)$$

3. EXPERIMENTAL VISCOSITY DATA AT HIGH DENSITY

The transport properties are formulated as functions of temperature and density from the kinetic theory, while experimental data are reported at specific temperatures and pressures. Consequently, a satisfactory thermodynamic formulation is an essential part of a pure fluid transport property correlation. For isobutane the modified Benedict–Webb–Rubin equation of state by Younglove and Ely [8] including a correction to the ITS-90 temperature scale has been applied to adjust the densities to which the viscosity data from earlier measurements have been referred. The viscosity itself has not been reevaluated. Primary and secondary data sets from the literature at densities $\rho > 0.10 \text{ mol} \cdot \text{L}^{-1}$ are summarized in Table II.

Table II. Data from Viscosity Measurements on Isobutane at High Densities
 $\rho > 0.10 \text{ mol} \cdot \text{L}^{-1}$

Author(s)	Year	Technique ^a	<i>T</i> (K)	<i>p</i> (MPa)	Phase ^b	No. of points	$\delta\eta_r^c$ (%)
Primary data sets:							
Gonzalez and Lee [7]	1966	C	311–444	0.7–55.2	L, S	50 ^d	2.5
Gonzalez and Lee [6]	1966	C	311–444	0.7–55.2	L, S	^e	
Agaev and Yusibova [10]	1969	C	273–548	0.49–68.6	V, L, S, sL	425 ^f	2.0 ^g
Diller and Van Poolen [9]	1985	OQ	115–300	0.41–34.5	L, sL	141 ^h	2.5
Secondary data sets:							
Sage et al. [21]	1939	RB	311–378	0.28–13.8	V, L, sL, sV	496	8.0
Lipkin et al. [22]	1942	C	200–278		sL	8	6.0
Babb and Scott [23]	1964	RB	303	200–800	L	4	8.0
Agaev and Yusibova [10]	1969	C	273–409 ⁱ	3.4–4.9	L, S, sV	71	8.0

^a C, capillary; OQ, oscillating quartz crystal; RB, rolling ball.

^b V, vapor; L, liquid; S, supercritical; sL, saturated liquid; sV, saturated vapor.

^c Ascribed relative uncertainty.

^d Individual experimental points at the same or nearly the same pressure reduced to averaged values. One value at 378 K, one value at 411 K → secondary data.

^e Recommended values on the basis of the experimental data reported by Gonzalez and Lee [7].

^f One value at 408 K, one value at 410 K, one value at 413 K → secondary data.

^g Experimental points at pressures >35 MPa (>range of EOS) → relative uncertainty: 4%.

^h Individual experimental points for the saturated liquid reduced to averaged values.

ⁱ L and S: six isotherms near the critical temperature (404–409 K); sV: 273–408 K.

The list includes the year of publication, the measurement technique, the temperature and pressure ranges covered together with the fluid phase investigated, the number of experimental points, and the ascribed relative uncertainty of the data. These viscosity data are also illustrated in the temperature–pressure diagrams of Fig. 1.

In principle, only data obtained with a capillary-flow apparatus could be classified as primary. Unfortunately, a fair number of experimental data by Agaev and Yusibova [10] had to be excluded from the primary data sets. These authors performed measurements in the liquid phase near the saturation line and in the neighborhood of the critical point, but some of the reported temperatures and pressures correspond to the gaseous state according to the MBWR equation of state. Thus, all the data in this region were rejected including values extrapolated to the saturated vapor. The uncertainty of the data by Gonzalez and Lee [6, 7] was ascribed to be

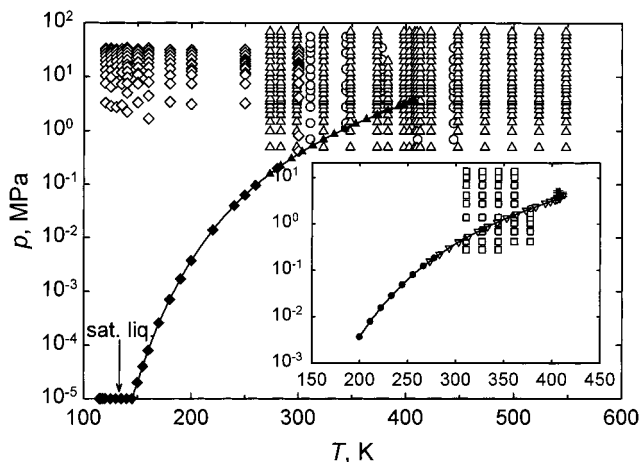


Fig. 1. Distribution of the available experimental viscosity data of isobutane at densities $\rho > 0.10 \text{ mol} \cdot \text{L}^{-1}$. Primary data: (○) Gonzalez and Lee (1966) [7]; Agaev and Yusibova (1969) [10]; (Δ) compressed vapor and liquid, (\blacktriangle) saturated liquid; Diller and Van Poolen (1985) [9]; (\diamond) compressed liquid, (\blacklozenge) saturated liquid data. Secondary data: Sage et al. (1939) [21]: (\square) compressed vapor and liquid, (\blacksquare) saturated vapor and liquid; (●) Lipkin et al. (1942) [22], saturated liquid; (\odot) Gonzalez and Lee (1966) [7], primary rejected; Agaev and Yusibova (1969) [10]: ($\#$) primary rejected, (+) near-critical region, (∇) saturated vapor data. (—) Liquid-vapor boundary.

$\pm 2.5\%$ due to a rather large scatter compared with the results by Starling et al. [24] obtained for propane with a capillary viscometer of the same kind. The capillary data by Lipkin et al. [22] were excluded since these measurements were analyzed with a simplified working equation. On the other hand, to cover as much as possible of the phase space, data by Diller and Van Poolen [9] obtained with an oscillating quartz-crystal viscometer were included although this technique does not meet the stringent conditions of primary instruments. These data are consistent with the other primary data near room temperature.

4. RESULTS AND DISCUSSION

The best correlation of the selected primary data sets was obtained with a representation of the higher density terms of the residual viscosity according to Eqs. (7)–(9) in which the double polynomials are restricted to $n=5$ and $m=2$ and only the first two coefficients in the temperature function $\delta_0(\tau)$ are considered. The significant coefficients are listed in Table I.

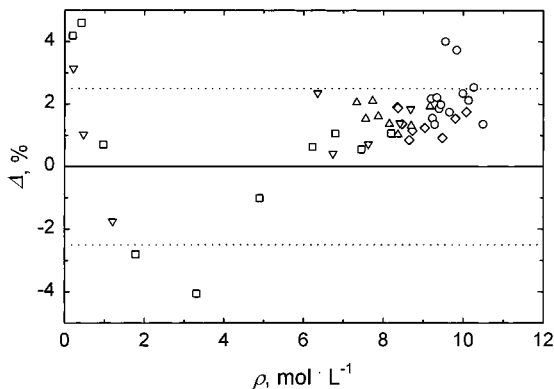


Fig. 2. Deviations Δ of the primary viscosity data by Gonzalez and Lee (1966) [7] from the present correlation. (○) 311 K, (◇) 344 K, (△) 378 K, (▽) 411 K, (□) 444 K. $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$.

In the course of the analysis, it was decided to transfer to secondary data such experimental points of the data sets which were supposed in the beginning to be primary and have subsequently been characterized by deviations of $\pm 5\%$. Thus, two outliers of Gonzalez and Lee [7] and three of Agaev and Yusibova [10] had to be rejected. The deviations from the final correlation of the total viscosity coefficient $\eta(\rho, T)$ for the remaining

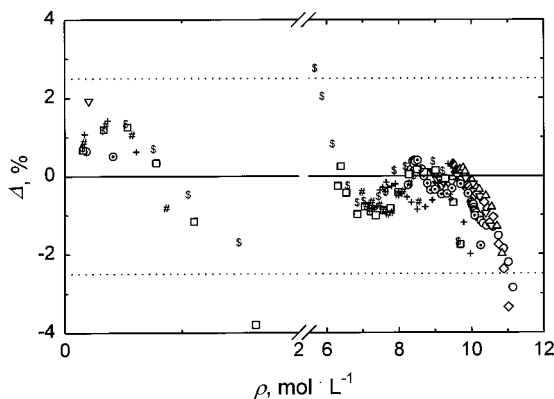


Fig. 3. Deviations Δ of the primary viscosity data by Agaev and Yusibova (1969) [10] from the present correlation, part I. (○) 273 K, (◇) 283 K, (△) 298 K, (▽) 323 K, (⊙) 348 K, (+) 373 K, (#) 383 K, (□) 398 K, (S) 403 K. $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$.

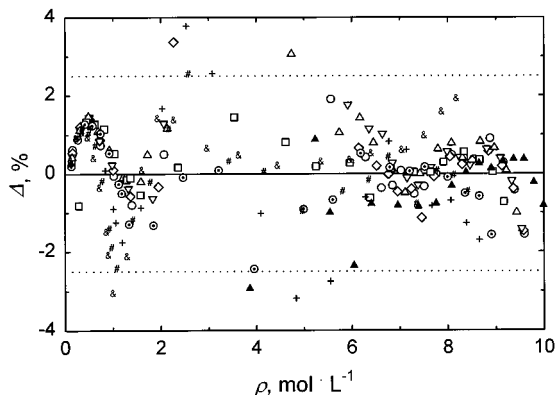


Fig. 4. Deviations Δ of the primary viscosity data by Agaev and Yusibova (1969) [10] from the present correlation, part II. (○) 408 K, (◇) 410 K, (▽) 413 K, (△) 423 K, (□) 448 K, (⊙) 473 K, (+) 498 K, (#) 523 K, (&) 548 K, (▲) saturated liquid data. $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$.

611 primary experimental data in the high-density region are illustrated in Figs. 2–5. The figures demonstrate that about 95% of the primary data are represented within $\pm 2.5\%$ and 99.5% within $\pm 4\%$. It emerges from Fig. 3 that there is a trend to lower values in the measurements by Agaev and Yusibova [10] at high densities and lower temperatures. This tendency

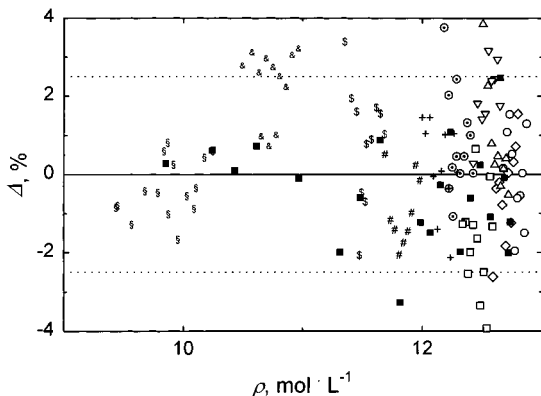


Fig. 5. Deviations Δ of the primary viscosity data by Diller and Van Poolen (1985) [9] from the present correlation. (○) 120 K, (◇) 125 K, (△) 130 K, (▽) 135 K, (□) 140 K, (⊙) 150 K, (+) 160 K, (#) 180 K, (§) 200 K, (&) 250 K, (§) 300 K, (■) saturated liquid data. $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$.

could be due to the application of the MBWR equation of state to evaluate the densities of the experiments for pressures which exceed the range of validity of the equation. An attempt was made to take this into account with an increase in the ascribed uncertainty of these data.

As remarked in the introduction, the correlation by Younglove and Ely [8] was largely undocumented with respect to the development of its formulation and to the data base selected. A comparison of the present correlation with the Younglove–Ely representation is shown for several isotherms in Fig. 6. The deviations between the two correlations exceed 2% at temperatures below 200 K in the vapor phase due to the fact that both correlations could not be based on any experimental data and the values at these temperatures were obtained by an uncertain extrapolation. On the contrary, differences larger than 2% emerge in the vapor phase at ambient temperature and at moderately low densities since Younglove and Ely did not consider the initial density dependence according to the Rainwater–Friend theory and based on the new experimental data by Küchenmeister and Vogel [11]. Thus, we believe that in this region the present correlation is an improvement over the previous one. The agreement in the liquid phase at 300 K and at lower temperatures is not surprising because both correlations are based on the same data sets by Diller and Van Poolen [9]. Remarkably, the deviations for the isotherms from 400 to 600 K show a

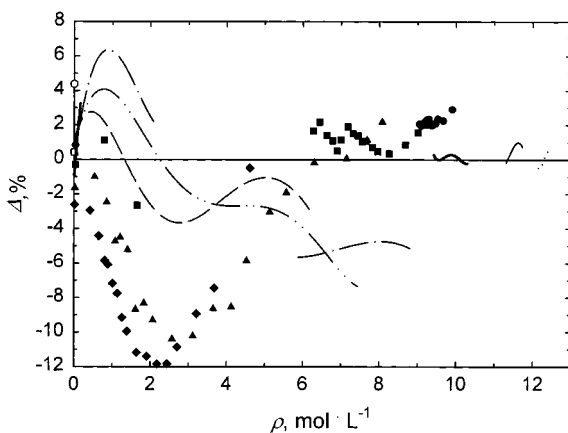


Fig. 6. Comparison with the recommended values by Stephan and Lucas (1979) [5] as well as the correlation by Younglove and Ely (1987) [8]. Stephan and Lucas: (●) 320 K, (■) 400 K, (▲) 500 K, (◆) 600 K. Younglove and Ely: (○) 150 K, (□) 200 K, (—) 300 K, (···) 400 K, (— · —) 500 K, (---) 600 K. $\Delta = 100(\eta_{\text{litcor}} - \eta_{\text{newcor}})/\eta_{\text{litcor}}$.

systematic trend with increasing density. The Younglove–Ely representation underestimates the present correlation by approximately 5% at high densities and leads to an overestimate of about 4% at moderate densities of about $1 \text{ mol} \cdot \text{L}^{-1}$. It is to be assumed that Younglove and Ely did not include the experimental values by Agaev and Yusibova [10] in their data base and simply extrapolated their correlation to high temperatures. In our opinion, the experimental data by Agaev and Yusibova are characterized by a reasonably small uncertainty so that an extrapolation with an empirical functional form cannot be preferred.

Figure 6 also includes a comparison with the tabulated values by Stephan and Lucas [5]. The negative deviations at very low densities which increase with increasing temperature are explained by the fact that these authors had no reliable data for their use. At 500 and 600 K, Stephan and Lucas underestimate the new correlation up to about 10% at densities lower than the critical density since they did not take into account the values by Agaev and Yusibova [10] as primary data, but trusted more their graphical extrapolation procedure. The agreement at 320 and 400 K in the liquid phase is within $\pm 3\%$ as expected.

The final representation of the viscosity of isobutane is given by Eqs. (1)–(9) including the coefficients of Table I and using the MBWR equation of state [8] with a correction to the ITS-90 temperature scale. Figure 7 illustrates its range of applicability as well as the uncertainty in various

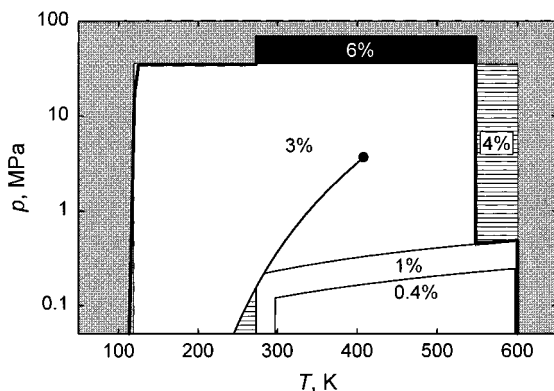


Fig. 7. Range of the viscosity representation and estimated uncertainty by region. \square , Validity range of MBWR equation of state; \blacksquare , range based on experimental viscosity data; ▨ , validity range of MBWR equation of state, but no experimental viscosity data available; \blacksquare , range based on experimental viscosity data, but outside the validity of the MBWR equation of state; \bullet , critical point.

thermodynamic regions. In the temperature range 297–627 K, the uncertainty is $\pm 0.4\%$ for densities below $0.05 \text{ mol} \cdot \text{L}^{-1}$, increasing to $\pm 1\%$ for densities to $0.10 \text{ mol} \cdot \text{L}^{-1}$ between 273 and 600 K. In the thermodynamic regions in which both the MBWR equation of state is valid and primary experimental data are available, the uncertainty is estimated to be $\pm 3\%$ corresponding to a twofold standard deviation. The uncertainty is estimated to increase to $\pm 4\%$ when extrapolating to the other thermodynamic ranges of the MBWR equation of state. The uncertainty increases further to $\pm 6\%$, in the case of extrapolations to higher pressures where only experimental viscosity data are available.

5. CONCLUSION

A new representation of the viscosity of isobutane encompassing large regions of thermodynamic states has been presented. The formulation is based on a critical assessment of the available experimental data, guided by theoretical considerations and results. The correlation represents 95% of the primary experimental data for the validity range of the MBWR equation of state within their ascribed relative uncertainties of $\pm 2.5\%$. Nevertheless, it would be advantageous to have more measurements with other primary instruments in the vapor phase, along the saturation line in the vapor phase above room temperature, and in the vicinity of the critical point in order to advance further our understanding of the behavior of the viscosity in these regions. Because mutual overlaps of the primary data sets are rather limited, the uncertainty ascribed to the present correlation is increased to $\pm 3\%$ to be on the safe side.

ACKNOWLEDGMENTS

C.K. is grateful to the Ministry of Culture of Mecklenburg-Vorpommern, Germany, for the award of the Landesgraduiertenförderung. We acknowledge the support by the Fonds der Chemischen Industrie im Verband der Chemischen Industrie, Germany. The project was carried out under the auspices of the Subcommittee on Transport Properties of Commission I.2 of the International Union of Pure and Applied Chemistry.

REFERENCES

1. S. Hendl, J. Millat, E. Vogel, V. Vesovic, W. A. Wakeham, J. Luettmmer-Strathmann, J. V. Sengers, and M. J. Assael, *Int. J. Thermophys.* **15**:1 (1994).
2. E. Vogel and C. Küchenmeister, *High Temp. High Press.* **29**:397 (1997).
3. E. Vogel, C. Küchenmeister, E. Bich, and A. Laesecke, *J. Phys. Chem. Ref. Data* **27**:947 (1998).

4. E. Vogel, C. Küchenmeister, and E. Bich, *High Temp. High Press.* **31**:173 (1999).
5. K. Stephan and K. Lucas, *Viscosity of Dense Fluids* (Plenum, New York, 1979), p. 62.
6. M. H. Gonzalez and A. L. Lee, *J. Chem. Eng. Data* **11**:357 (1966).
7. M. H. Gonzalez and A. L. Lee, *Viscosity of isobutane*, Document No. 8919 deposited (American Documentation Institute, Library of Congress, Washington, D.C., 1966).
8. B. A. Younglove and J. F. Ely, *J. Phys. Chem. Ref. Data* **16**:577 (1987).
9. D. E. Diller and L. J. Van Poolen, *Int. J. Thermophys.* **6**:43 (1985).
10. N. A. Agaev and A. D. Yusibova, *Gazov. Prom.* **14**:41 (1969).
11. C. Küchenmeister and E. Vogel, *Int. J. Thermophys.* **21**:329 (2000).
12. J. H. Dymond, E. Bich, E. Vogel, W. A. Wakeham, V. Vesovic, and M. J. Assael, in *Transport Properties of Fluids*, J. Millat, J. H. Dymond, and C. A. Nieto de Castro, eds. (Cambridge University Press, Cambridge, 1996), p. 66.
13. G. C. Maitland, M. Rigby, E. B. Smith, and W. A. Wakeham, *Intermolecular Forces: Their Origin and Determination* (Clarendon Press, Oxford, 1987).
14. V. Vesovic and J. Millat, in *Transport Properties of Fluids*, J. Millat, J. H. Dymond, and C. A. Nieto de Castro, eds. (Cambridge University Press, Cambridge, 1996), p. 348.
15. D. G. Friend and J. C. Rainwater, *Chem. Phys. Lett.* **107**:590 (1984).
16. J. C. Rainwater and D. G. Friend, *Phys. Rev. A* **36**:4062 (1987).
17. E. Bich and E. Vogel, *Int. J. Thermophys.* **12**:27 (1991).
18. E. Bich and E. Vogel, in *Transport Properties of Fluids*, J. Millat, J. H. Dymond, and C. A. Nieto de Castro, eds. (Cambridge University Press, Cambridge, 1996), p. 72.
19. V. Vesovic, W. A. Wakeham, G. A. Olchowy, J. V. Sengers, J. T. R. Watson, and J. Millat, *J. Phys. Chem. Ref. Data* **19**:763 (1990).
20. P. T. Boggs, R. H. Byrd, J. E. Rogers, and R. B. Schnabel, *User's Reference Guide for ODRPACK Version 2.01 Software for Weighted Orthogonal Distance Regression*, NISTIR 4834 (National Institute of Standards and Technology, Gaithersburg, Maryland, 1992).
21. B. H. Sage, W. D. Yale, and W. N. Lacey, *Ind. Eng. Chem.* **31**:223 (1939).
22. M. R. Lipkin, J. A. Davison, and S. S. Kurtz, Jr., *Ind. Eng. Chem.* **34**:976 (1942).
23. S. E. Babb, Jr., and G. J. Scott, *J. Chem. Phys.* **40**:3666 (1964).
24. K. E. Starling, B. E. Eakin, and R. T. Ellington, *Liquid, gas, and dense-fluid viscosity of propane*, Document No. 6346 deposited (American Documentation Institute, Library of Congress, Washington, D.C., 1959).