

The Viscosity of Gaseous Isobutane and Its Initial Density Dependence¹

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Results of new relative high-precision measurements on gaseous isobutane are reported. Six series, each differing in density, were performed in a quartz oscillating-disk viscometer from 297 to 627 K and for densities between 0.010 to 0.048 mol · L⁻¹. Isothermal values recalculated from the original experimental data were evaluated with a first-order expansion, in terms of density, for the viscosity. Reduced values of the second viscosity virial coefficient derived from the zero-density and initial-density viscosity coefficients agree with the representation of the Rainwater–Friend theory when using energy and length scaling factors specific for the interactions of the isobutane molecules. With the same scaling factors an individual correlation in the limit of zero density was developed including only a few values from the literature. The uncertainty of the zero-density viscosity correlation is estimated to be ±0.4%.

KEY WORDS: isobutane; second viscosity virial coefficient; transport property; viscosity.

1. INTRODUCTION

In a program initiated under the auspices of IUPAC, the viscosity surfaces of ethane [1] and propane [2, 3] in the series of *n*-alkanes were studied. The program needs to be extended to *n*-butane and isobutane as important industrial fluids. Isobutane has gained renewed attention as an environmentally friendly alternative refrigerant and as a natural gas component.

The development of a viscosity surface has to be started in the low-density region in which the correlation can be based on established

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theoretical guidance and on accurate experimental data. Recently, results of high-precision viscosity measurements on *n*-butane required for that purpose were reported and evaluated including some data from the literature to deduce a zero-density and an initial-density viscosity correlation [4, 5]. Very recently, relative measurements were performed with our oscillating-disk viscometer for isobutane between 297 and 597 K at low densities. The results were reported at the 5th Asian Thermophysical Properties Conference [6]. Unfortunately, there was an error in the temperature control regime of the thermostat of the viscometer at higher temperatures. Therefore, we repeated the complete measuring program and now present high-precision results for isobutane in the low-density region at temperatures from 297 to 627 K. On the other hand, only a few of the earlier experimental results fulfil the requirements on primary data. The shortcomings of most older data were low accuracy and, above all, a limited number of experimental points on isotherms which are needed for the determination of both leading terms in the density expansion of the viscosity.

The interactions between the isobutane molecules are specific, so that individual correlations have to be derived in the limit of zero density as well as for the initial density dependence of the viscosity. The proposed representation differs from an earlier correlation by Younglove and Ely [7] in that the initial density dependence has been taken into account in an appropriate way [8, 9] and in that the experimental database has been expanded.

2. METHODOLOGY

The viscosity $\eta(\rho, T)$ of a moderately dense pure gas at molar density ρ and temperature T may be represented by a density expansion restricted to the first power:

$$\eta(\rho, T) = \eta^{(0)}(T) + \eta^{(1)}(T) \rho + \dots = \eta^{(0)}(T) [1 + B_\eta(T) \rho + \dots] \quad (1)$$

Here, $\eta^{(0)}$ and $\eta^{(1)}$ are the zero-density and initial-density viscosity coefficients, whereas B_η is the second viscosity virial coefficient.

The best way to determine $\eta^{(0)}(T)$ is to extrapolate isotherms with a sufficient number of experimental points at moderately low densities to the limit of zero density, because measurements in that limit are not feasible. However, the results at low pressures, such as 0.1 MPa, cannot be equated with zero-density viscosity coefficients within the experimental uncertainties at all temperatures. Knowledge of the initial density dependence of viscosity can be used to correct such data in order to compare them with actual zero-density viscosity values.

The well-established kinetic theory of dilute monatomic gases by Chapman and Enskog [10] can be used in the analysis since $\eta^{(0)}(T)$ is almost independent of the existence of the internal degrees of freedom and nearly unaffected by inelastic collisions. The incomplete knowledge of the intermolecular potential energy surface of polyatomic molecules like isobutane is taken into account via an effective collision cross section which contains all the dynamic and statistical information about the binary collision. Then, $\eta^{(0)}(T)$ is given, in practical engineering form [11], by

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

$$T^* = k_B T / \varepsilon \quad (3)$$

where \mathfrak{S}_η^* represents a reduced effective cross section and T^* the reduced temperature. M is the molar mass in $\text{g} \cdot \text{mol}^{-1}$, T is the temperature in K, $\eta^{(0)}$ is in units of $\mu\text{Pa} \cdot \text{s}$, whereas σ is a length scaling parameter in nm and ε/k_B is an energy scaling parameter in K. The temperature dependence of the reduced effective cross section is usually expressed in the functional form

$$\ln \mathfrak{S}_\eta^*(T^*) = \sum_{i=0}^4 a_i (\ln T^*)^i \quad (4)$$

To develop an accurate and consistent representation for the viscosity, primary data sets have to be selected from the available experimental data in the literature. Primary class data are qualified by measurements with a high-precision instrument for which a full working equation is available and all necessary corrections can be applied. The relative uncertainty of the data $\delta\eta_r$ was assigned according to the analysis performed by the present authors and was based on the measurement method, the quoted experimental error, the check on precision of the data, and the discrepancies with other independent data in overlapping temperature ranges. Then, the statistical weights w_k follow from

$$w_k = \left(\frac{100}{\eta_{\text{exp},k} \delta\eta_{r,k}} \right)^2 \quad (5)$$

where $\eta_{\text{exp},k}$ is the experimental viscosity value k .

The \mathfrak{S}_η ($\mathfrak{S}_\eta = \pi\sigma^2 \mathfrak{S}_\eta^*$) values have been calculated from the selected primary experimental data with the help of Eq. (2) to develop the representation of the zero-density viscosity coefficient. The fit could be carried out in two versions. The adoption of a universal correlation requires that the fluid under discussion obeys the extended principle of corresponding states

for the noble gases. In that case, values of the scaling factors ε/k_B and σ are derived using the known coefficients a_i of the functional \mathfrak{S}_η^* for the rare gases by Bich et al. [12]. But polyatomics like isobutane require an individual correlation specific for the considered molecules, because they do not conform to the corresponding-states principle for the monatomics. Then, the scaling factors determined for the universal correlation could be kept fixed and new values of the coefficients a_i could be derived by fitting Eq. (4) to the quasiexperimental \mathfrak{S}_η values. Since the values of the scaling factors ε/k_B and σ are in principle arbitrary and the number of parameters and coefficients in a full surface correlation should be minimized, we have adopted the same scaling factors deduced in the analysis of the second viscosity virial coefficient (see below). This procedure corresponds to that applied to propane [3] and *n*-butane [5].

With the development of the Rainwater–Friend theory for the transport properties of the moderately dense gas [9, 13, 14], it is possible to establish separately the linear-in-density term $\eta^{(1)}(T)\rho$ and the temperature function $B_\eta(T)$. Rainwater and Friend have modeled the moderately dense gas as a mixture of monomers and dimers which interact according to the Lennard-Jones (12–6) potential. Two potential parameter ratios, δ and θ , relating the monomer–dimer interaction to that of the monomer–monomer interaction have been determined by fitting the theoretical expressions to selected second transport virial coefficients. Thus, Bich and Vogel [8, 9] presented tables of the reduced second viscosity virial coefficient B_η^* as a function of T^* for an optimized set of δ and θ . Recently, Vogel et al. [3] recommended an improved empirical equation in the reduced temperature range $0.5 \leq T^* \leq 100$ which can safely be 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 $L \cdot \text{mol}^{-1}$. The coefficients b_i were reported in Ref. 3.

In principle, Lennard-Jones (12–6) potential parameters derived from the zero-density viscosity coefficient $\eta^{(0)}(T)$ could be used for a representation of $B_\eta(T)$ in reduced form. In accordance with the fact that isobutane conforms neither to the corresponding-states principle nor to the Lennard-Jones (12–6) potential, specific scaling factors ε/k_B and σ should be optimized by fitting the theoretical results by Bich and Vogel [9] or the representative correlation of Eq. (6) to experimental B_η values of the present paper. In that way a set of individual scaling factors for isobutane will be applied for the temperature functions of both the zero-density and initial-density viscosity coefficients.

3. EXPERIMENTAL RESULTS

The measurements were performed in a quartz oscillating-disk viscometer with small gaps. The basic principles of design and construction of the instrument [15] as well as details of performance of the relative measurements and of the evaluation of the measured data were reported previously [16, 17]. In analogy to the experiments on propane [18] and *n*-butane [4], the uncertainty of the viscosity data has been estimated to be $\pm 0.15\%$ at room temperature increasing up to $\pm 0.3\%$ at the highest temperatures. The reproducibility has been determined to be about $\pm 0.1\%$ in the whole temperature range.

Six isochoric series of measurements were carried out on isobutane in a temperature range from 297 to 627 K for densities between 0.010 and 0.048 mol · L⁻¹. The densities were determined during the filling process by $p\rho T$ measurements considering the real-gas behavior according to the MBWR equation of state [7] including a correction of the temperatures to ITS-90. The certified purity of isobutane supplied by UCAR (Union Carbide, Spezialgase, Biebesheim, Germany) amounted to 99.95%. The experimental data are recorded in Table I.

The isochoric data of Table I were adjusted to isothermal values by means of a first-order Taylor series in terms of temperature,

$$\eta(T_{\text{int}}) = \eta(T_{\text{exp}}) + \left(\frac{\partial \eta}{\partial T} \right)_\rho \Delta T + R_n \quad (7)$$

$$\Delta T = T_{\text{int}} - T_{\text{exp}} \quad (8)$$

The interpolation temperature T_{int} followed from averaging the corresponding temperatures T_{exp} of the six series. The temperature derivative $(\partial \eta / \partial T)_\rho$ needed in Eq. (7) was calculated with the coefficients of the following equation, which were deduced by means of a fit to the original isochoric data and are listed in Table II:

$$\eta(T) = S \exp \left(A \ln T_{\text{R}} + \frac{B}{T_{\text{R}}} + \frac{C}{T_{\text{R}}^2} + D \right) \quad (9)$$

$$T_{\text{R}} = \frac{T}{298.15}, \quad S = 10.0 \mu\text{Pa} \cdot \text{s} \quad (10)$$

The remainder R_n in Eq. (7) was proven to be negligible in comparison with the experimental uncertainty.

The zero-density and initial-density viscosity coefficients $\eta^{(0)}$ and $\eta^{(1)}$ were determined by fitting Eq. (1) to the quasiexperimental values of the

Table I. Viscosity of Isobutane

T (K)	η ($\mu\text{Pa}\cdot\text{s}$)	T (K)	η ($\mu\text{Pa}\cdot\text{s}$)	T (K)	η ($\mu\text{Pa}\cdot\text{s}$)
Series 1 $\rho = 0.00997 \text{ mol}\cdot\text{L}^{-1}$		Series 2 $\rho = 0.01023 \text{ mol}\cdot\text{L}^{-1}$		Series 3 $\rho = 0.02138 \text{ mol}\cdot\text{L}^{-1}$	
297.29	7.492	297.53	7.500	297.60	7.486
324.50	8.144	324.33	8.142	324.63	8.136
352.67	8.809	352.37	8.805	352.63	8.803
381.02	9.477	380.98	9.475	381.95	9.495
409.34	10.135	409.10	10.132	409.51	10.129
438.20	10.802	437.80	10.793	437.73	10.788
467.29	11.456	466.75	11.446	467.08	11.466
495.87	12.106	495.57	12.100	495.89	12.102
525.03	12.742	524.86	12.734	526.32	12.772
546.35	13.209	546.29	13.207	546.36	13.212
568.24	13.677	567.46	13.664	568.57	13.698
597.26	14.290	597.12	14.283	597.62	14.317
626.99	14.910	626.48	14.911	626.84	14.927
297.26	7.493	298.39	7.520	297.52	7.494
Series 4 $\rho = 0.02955 \text{ mol}\cdot\text{L}^{-1}$		Series 5 $\rho = 0.03796 \text{ mol}\cdot\text{L}^{-1}$		Series 6 $\rho = 0.04789 \text{ mol}\cdot\text{L}^{-1}$	
297.69	7.491	297.54	7.480	297.42	7.471
325.05	8.150	324.92	8.142	324.69	8.133
352.89	8.812	352.50	8.810	352.56	8.797
381.21	9.480	380.80	9.469	380.92	9.473
409.91	10.153	410.35	10.165	409.62	10.148
438.50	10.816	438.20	10.813	437.90	10.804
467.31	11.472	466.95	11.456	466.61	11.458
496.21	12.123	495.49	12.104	495.67	12.115
525.52	12.771	524.60	12.753	524.93	12.754
546.43	13.233	545.89	13.218	546.78	13.244
568.36	13.709	567.68	13.689	567.74	13.688
597.31	14.317	597.06	14.311	597.02	14.297
627.17	14.938	626.62	14.915	626.01	14.906
297.77	7.496	297.72	7.484	297.15	7.464

isotherms. They are summarized together with their individual standard deviations s_{η_0} and s_{η_1} and with the standard deviation s_{η} for each isotherm in Table III. In Fig. 1 the initial density dependence of viscosity is demonstrated for some isotherms. Viscosity values resulting from Eq. (1) are plotted as straight lines or, in the case of atmospheric pressure, as filled circles. $\eta^{(0)}$ and viscosity values at 0.1013 MPa were again correlated with Eq. (9), and the coefficients deduced are given in Table II.

Table II. Parameters of Eq. (9) for the Series of Measurements on Isobutane

Series	A	B	C	D	$10^3 s_\eta^a$ ($\mu\text{Pa}\cdot\text{s}$)	10^2 RMS (%)
1	0.585734	-0.813409	0.228387	0.299241	2.93	2.30
2	0.637142	-0.670093	0.180186	0.204235	4.44	2.98
3	0.619423	-0.732450	0.202323	0.243020	5.08	4.24
4	0.554898	-0.906822	0.258122	0.361666	3.26	2.48
5	0.582842	-0.806590	0.216173	0.302243	6.12	4.67
6	0.541116	-0.916117	0.249594	0.377440	4.49	3.12
^b	0.636348	-0.684479	0.190243	0.209293	2.84	2.20
^c	0.619992	-0.696793	0.177573	0.230658	2.55	2.00

^a Standard deviation.^b At zero density ($297 \leq T \leq 627$ K).^c At 0.1013 MPa ($297 \leq T \leq 627$ K).

4. LOW-DENSITY VISCOSITY CORRELATION

The present experimental data are the only values suitable to resolve both $\eta^{(0)}$ and $\eta^{(1)}$. All other data from the open literature are characterized by a low accuracy and/or an insufficient number of isothermal values at low densities.

In a preliminary analysis of B_η derived from $\eta^{(0)}$ and $\eta^{(1)}$ according to Eq. (1), reduced values B_η^* were calculated with Lennard-Jones (12-6)

Table III. Zero-Density and Initial-Density Viscosity Coefficients of Isobutane for the Isotherms According to Eq. (1)

T (K)	$\eta_0 \pm s_{\eta_0}$ ($\mu\text{Pa}\cdot\text{s}$)	$\eta_1 \pm s_{\eta_1}$ ($\mu\text{Pa}\cdot\text{s}\cdot\text{L}\cdot\text{mol}^{-1}$)	$10^3 s_\eta$ ($\mu\text{Pa}\cdot\text{s}$)
297.51	7.503 ± 0.003	-0.638 ± 0.099	3.39
324.69	8.152 ± 0.003	-0.416 ± 0.098	3.35
352.60	8.810 ± 0.005	-0.150 ± 0.157	5.34
381.15	9.479 ± 0.001	-0.032 ± 0.041	1.39
409.64	10.138 ± 0.005	0.208 ± 0.177	6.02
438.06	10.795 ± 0.003	0.311 ± 0.107	3.66
467.00	11.450 ± 0.005	0.357 ± 0.164	5.59
495.78	12.099 ± 0.004	0.355 ± 0.130	4.42
525.21	12.739 ± 0.006	0.578 ± 0.188	6.41
546.35	13.201 ± 0.004	0.737 ± 0.147	5.02
568.01	13.671 ± 0.007	0.627 ± 0.222	7.56
597.23	14.289 ± 0.010	0.533 ± 0.324	11.04
626.68	14.911 ± 0.007	0.260 ± 0.243	8.27
297.64	7.510 ± 0.002	-0.699 ± 0.065	2.22

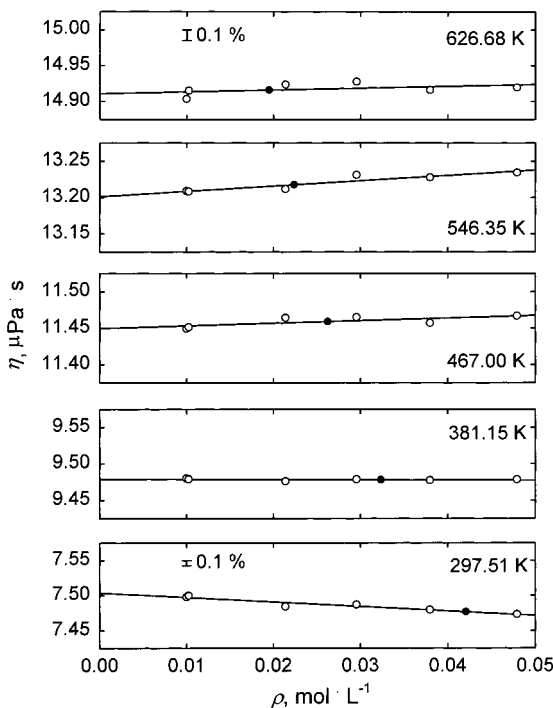


Fig. 1. Viscosity of isobutane as a function of molar density for several isotherms. $\eta = \eta_0 + \eta_1 \rho$: (○) quasiexperimental values; (●) at 0.1013 MPa, calculated.

potential parameters which were determined from $\eta^{(0)}(T)$ of this paper to be

$$\varepsilon/k_B = 284.29 \text{ K}, \quad \sigma = 0.54965 \text{ nm}$$

In a second step, individual scaling factors were deduced by fitting the theoretical results of B_η^* , for the ratios δ and θ given by Bich and Vogel [9], to the experimental B_η values:

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

In Fig. 2 the curves of the theoretical results for δ and θ , obtained originally by Rainwater and Friend [13, 14] and more recently by Bich and Vogel [8, 9], are compared with B_η^* values for isobutane based either on the Lennard-Jones (12-6) potential parameters or on the optimized scaling factors. B_η^* values of propane [3] and *n*-butane [4, 5] also determined

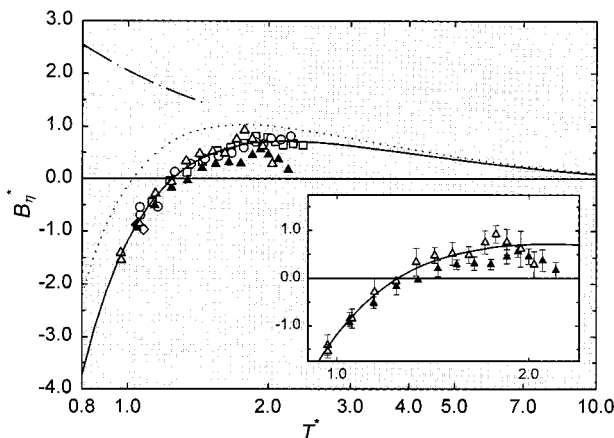


Fig. 2. Reduced second viscosity virial coefficient B_{η}^* as a function of the reduced temperature T^* . Optimized parameters: (\square) propane, Vogel (1995) [18]; (\diamond) *n*-butane, Kestin and Yata (1968) [19]; (\circ) *n*-butane, Küchenmeister and Vogel (1998) [4, 5]; (\triangle) isobutane, present paper. Lennard-Jones (12-6) parameters from $\eta^{(0)}(T)$: (\blacktriangle) isobutane, present paper. (\cdots) Rainwater-Friend theory [13, 14] ($\delta=1.02$, $\theta=1.15$); ($-$) Rainwater-Friend theory, Bich and Vogel [8, 9] ($\delta=1.04$, $\theta=1.25$); ($-\cdot-$) Younglove and Ely [7].

with optimized scaling factors are included. The B_{η}^* values for isobutane taking into account the standard deviation of η_1 as error bars are presented in the inset of the figure. It demonstrates, on the one hand, the suitability of the Rainwater-Friend theory and, on the other hand, the inability of the Younglove-Ely correlation [7] for representing the initial density dependence of viscosity.

A list of viscosity measurements on isobutane at low densities from the literature is compiled in Table IV. The list includes the method of measurement, the temperature range, the number of points at densities $\rho < 0.10$ mol \cdot L $^{-1}$, the ascribed relative uncertainty of the data, and the classification as primary or secondary data sets.

In order to analyze all data in the limit of zero density, the data from the literature were corrected for the initial density dependence according to the results of the present paper, although the experimental uncertainties of most references are larger than this systematic effect.

The reliability of the results of Kestin and collaborators measured above room temperature with their high-temperature oscillating-disk viscometer [27] is problematic because all these data have been affected in a systematic way by a temperature measurement error. In the case of

Table IV. List of Available Data from Viscosity Measurements of Isobutane at Low Densities $\rho < 0.10 \text{ mol} \cdot \text{L}^{-1}$

Author(s)	Year	Technique ^a	<i>T</i> (K)	No. of points	$\delta\eta_r$ ^b (%)
Primary data sets:					
Abe et al. [20]	1979	OD	298–468	7	0.2, 0.4 ^c
Dunlop [21]	1994	C	298	1	0.3
present paper	1998	OD	297–627	14	0.2–0.4
Secondary data sets:					
Ishida [22]	1923	FO	296	1	3.0
Titani [23]	1930	C	293–373	6	2.0
Sage et al. [24]	1939	RB	311–378	12 ^d	5.0
Lambert et al. [25]	1955	OP, C	308–363	8	3.0
Agaev and Yusibova [26]	1969	C	273–548	18	1.5

^a C capillary; FO, falling oil–drop; OD, oscillating disk; OP, oscillating pendulum; RB, rolling ball.

^b Ascribed relative uncertainty.

^c 0.2% only at 298 K.

^d Five values by extrapolation of isotherms.

propane [3] and *n*-butane [5] we felt compelled to exclude all data of Kestin and collaborators above room temperature from the primary data sets since most of them are characterized by systematic positive deviations of 0.4–1.0% from our values.

For isobutane the values by Abe et al. [20] are the only data from Kestin's group. They have been accepted as primary with a slightly increased uncertainty above 298 K since these data are in reasonably good agreement with those of the present paper. It is to be stressed that this agreement could be fortuitous. In addition, one data point by Dunlop [21] could be included in the primary data.

The scaling factors of a universal correlation with the coefficients a_i of the functional \mathfrak{S}_η^* for the monatomics deduced for the reduced temperature range $1.2 \leq T^* \leq 10$ by Bich et al. [12] were determined to be

$$\varepsilon/k_B = 292.41 \text{ K}, \quad \sigma = 0.55059 \text{ nm}$$

Finally, the optimized scaling factors ε/k_B and σ obtained in the analysis of the initial density dependence were used to establish new coefficients a_i of an individual functional \mathfrak{S}_η^* . The significant coefficients a_i of Eq. (4) in the temperature range 297–627 K were deduced to be

$$a_0 = 0.53583008, \quad a_1 = -0.45629630, \quad a_2 = 0.049911282, \quad a_3 = 0, \quad a_4 = 0$$

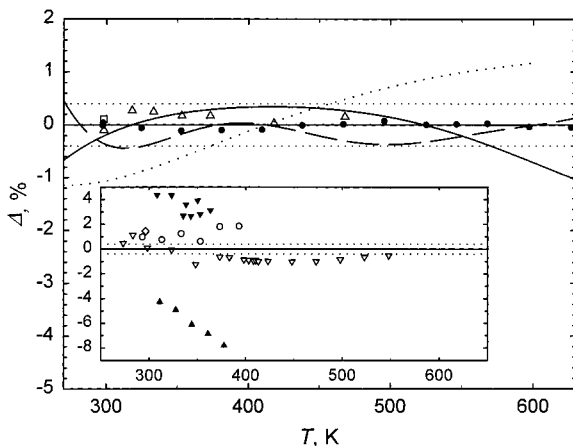


Fig. 3. Deviations $\Delta = 100.0(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$ of viscosity data from the present individual zero-density viscosity correlation. Primary data: (Δ) Abe et al. (1979) [20]; (\square) Dunlop (1994) [21]; (\bullet) present paper; (\cdots) Younglove and Ely (1987) [7]; ($--$) Tarzimanov et al. (1987) [28]; ($-$) universal correlation. Secondary data: (\diamond) Ishida (1923) [22], (\circ) Titani (1930) [23], (\blacktriangle) Sage et al. (1939) [24], (\blacktriangledown) Lambert et al. (1955) [25], (∇) Agaev and Yusibova (1969) [26].

The deviations of the primary data sets from the individual correlation are presented in the main part of Fig. 3, whereas the deviations of the secondary data sets are illustrated in its inset. The figure reveals clearly that the universal correlation is not suitable to represent the primary data within their uncertainties. The Younglove–Ely correlation [7] disagrees completely with the final representation. In particular, the discrepancy of 1% at 298 K is unexpected because Younglove and Ely had the data of Abe et al. [20] at their disposal. The values recommended by Tarzimanov et al. [28] agree within $\pm 0.4\%$ with the new correlation. This is certainly due to the inclusion of the data of Agaev and Yusibova [26], which are in rather good agreement with the present experimental data. Another set of values recommended by Golubev [29] as well as by Gonzalez and Lee [30] is not included in the comparison because they were extensively extrapolated beyond the temperature range of the experimental data on which they were based.

5. CONCLUSION

The representation of the viscosity in the low-density region includes a zero-density correlation based on the kinetic theory of dilute gases and

an initial-density dependence correlation according to the Rainwater-Friend theory. Both of them represent individual correlations due to the specific interactions between the polyatomic molecules of isobutane. To minimize the number of coefficients and parameters of a full surface correlation, the same scaling factors ε/k_B and σ are employed in both correlations. Whereas only the present experimental data are suitable to be used for the determination of the initial-density dependence correlation, a few data from the literature were included to derive the zero-density correlation. Its uncertainty is estimated to be $\pm 0.4\%$.

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