

## The Viscosity of Gaseous Propane and Its Initial Density Dependence

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Results of five series of high-precision viscosity measurements on gaseous propane, each differing in density, are reported. The measurements were performed in a quartz oscillating-disk viscometer with small gaps from room temperature up to about 625 K and for densities between 0.01 and 0.05 mol · L<sup>-1</sup>. The experimental data were evaluated with a first-order expansion, in terms of density, for the viscosity. Reduced values of the second viscosity virial coefficients deduced from the zero-density and initial-density viscosity coefficients for propane and for further *n*-alkanes are in close agreement with the theoretical representation of the Rainwater-Friend theory for the potential parameter ratios by Bich and Vogel. A new representation of the viscosity of propane in the limit of zero density is provided using the new experimental data and some data sets from literature. The universal correlation based on the extended principle of corresponding states extends over the temperature range 293 to 625 K with an uncertainty of  $\pm 0.5\%$  and deviates from earlier representations by about 1% at the upper temperature limit.

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**KEY WORDS:** *n*-alkanes; propane; second viscosity virial coefficient; transport properties; viscosity.

### 1. INTRODUCTION

The development of state-of-the-art representations of thermophysical properties has gained renewed attention. Thus, a research program has been started under the auspices of IUPAC in order to develop procedures for representing the transport properties of fluids of industrial importance on the basis of a critical analysis of the available experimental data including recent results of kinetic theory. The aim of the program in which carbon dioxide [1] and ethane [2, 3] have been studied is accurate, consistent, and theoretically sound representations of the transport properties over the widest range of thermodynamic states possible.

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The correlation of a transport-property surface should be started at low densities including possibly improved measurements and recent results of the kinetic theory as in the case of the viscosity surface of ethane [4]. Now we extend the investigation to propane and present new viscosity measurements at relatively low densities. The experimental results are used to determine both leading terms in the density series of the viscosity.

Earlier viscosity correlations in the low-density range, for example [5, 6], suffer from two problems. First, although there exist a great number of experimental data at low densities in the existing literature, most of them have been influenced by the results of Trautz et al. [7, 8], which obviously are subject to a systematic error with increasing temperature. Thus, the data of Trautz et al. were used to calibrate the instruments and to test their performance. Holland et al. [6] have tried to account for the error in those data, but it is difficult to estimate an error without independent values for the same substance. The new experimental data cover a larger temperature range than measured up to now and should be more reliable, too. Second, a novel way of evaluating the data in the vapor phase at low densities has been developed [9]. Incorporating this method of correlating and predicting the initial density dependence of viscosity and using the experimental results presented here, the viscosity correlation in the low-density range can now be greatly improved.

## 2. METHODOLOGY OF DATA CORRELATION

The viscosity  $\eta$  of a moderately dense gas may be represented at temperature  $T$  and density  $\rho$  by a density expansion limited to the first power:

$$\eta(T, \rho) = \eta_0(T) + \eta_1(T) \rho + \dots = \eta_0(T) [1 + B_\eta(T) \rho + \dots] \quad (1)$$

Here  $\eta_0$  is the viscosity in the limit of zero density,  $\eta_1$  the initial-density viscosity coefficient, and  $B_\eta$  the second viscosity virial coefficient.

The viscosity in the zero-density limit  $\eta_0(T)$  is not directly experimentally accessible but can be obtained by extrapolation of the results of low-density measurements. Since the well-established kinetic theory of dilute gases [10] relates to the zero-density viscosity coefficient, this limit is the natural starting point for any viscosity correlation. The viscosity of a pure gas is characterized in practical engineering form by

$$\eta_0(T) = \frac{0.021357(MT)^{1/2} f_\eta}{\sigma^2 \mathfrak{S}^*(2000)(T^*)} = \frac{0.021357(MT)^{1/2}}{\sigma^2 \mathfrak{S}_\eta^*(T^*)} \quad (2)$$

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

where  $\mathfrak{S}^*(2000)$  is the reduced effective collision cross section, which includes all the dynamical and statistical information about the binary collisions.  $f_\eta$  is the correction factor that accounts for high-order terms of the kinetic theory, and  $T^*$  is the reduced temperature.  $\mathfrak{S}^*(2000)$  is related to the earlier frequently used reduced collision integral  $\Omega^{(2,2)*}$  by  $\Omega^{(2,2)*} = (5/4) \mathfrak{S}^*(2000)$ . The notation  $\mathfrak{S}_\eta^*$  has been introduced in order to incorporate  $\mathfrak{S}^*(2000)$  and  $f_\eta$  for convenient use with respect to data correlation. In Eqs. (2) and (3),  $T$  is the temperature in K,  $M$  the relative molecular mass,  $\sigma$  the collision diameter of an assumed intermolecular potential or a length scaling parameter in nm,  $\varepsilon/k$  the potential well depth or an energy scaling parameter in K, and  $\eta_0$  is in units of  $\mu\text{Pa} \cdot \text{s}$ . The numerical constant results by the use of the recommended values of fundamental constants [11].

In the procedure of developing the viscosity correlation, the available experimental data are divided into primary and secondary data sets starting with a critical assessment of the methods of measurements with regard to the completeness of the measuring theory and to the achieved precision. Then experimental  $\mathfrak{S}_\eta$  data ( $\mathfrak{S}_\eta = \pi\sigma^2\mathfrak{S}_\eta^*$ ) are deduced from each value selected for the primary data sets. With appropriate statistical weights the complete set of primary experimental data for  $\mathfrak{S}_\eta^*(T^*)$  are fitted to the functional form

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

It has been found that extensive and accurate correlations for the viscosity of many simple gases are possible by means of so-called universal correlations on the basis of the extended principle of corresponding states [10, 12]. Bich et al. [13] developed a universal correlation, using quasi-experimental values calculated from the best HFD potentials of the noble gases, and determined simultaneously the coefficients  $a_i$  of Eq. (4) and the scaling factors  $\sigma$  and  $\varepsilon/k$  in Eqs. (2) and (3), valid for two intermediate reduced temperature ranges. The individuality of the noble gases is only to take into account outside these ranges. With the coefficients  $a_i$  of a universal correlation the scaling factors  $\sigma$  and  $\varepsilon/k$  for any other substance can be deduced from its temperature function  $\eta_0(T)$ . Such a universal correlation has the advantage of a possible extrapolation to a practical upper temperature limit without direct measurements. The universal correlation derived for the reduced temperature range  $1.2 < T^* < 10$  by Bich et al. [13] with the coefficients  $a_i$  given in Table I is used in the present paper.

The viscosity in the vapor phase is characterized by the temperature function of the second viscosity virial coefficient  $B_\eta(T)$  and the linear-density correction  $\eta_1(T)$ , respectively. Rainwater and Friend [14, 15]

**Table I.** Coefficients for Representation of the Reduced Effective Collision Cross Section  $\Theta_{\eta}^*$  [Eq. (4)] and of the Reduced Second Viscosity Virial Coefficient  $B_{\eta}^*$  [Eq. (9)] with  $\delta = 1.04$  and  $\theta = 1.25$  by Bich and Vogel [9]

Coefficient for $\Theta_{\eta}^*$	
$i$	$a_i$
0	$0.2218816 \times 10^0$
1	$-0.5079322 \times 10^0$
2	$0.1285776 \times 10^0$
3	$-0.8328165 \times 10^{-2}$
4	$-0.2713173 \times 10^{-2}$
Coefficient for $B_{\eta}^*$	
$i$	$c_i$
0	$-0.17999496 \times 10^1$
1	$0.46692621 \times 10^2$
2	$-0.53460794 \times 10^3$
3	$0.33604074 \times 10^4$
4	$-0.13019164 \times 10^5$
5	$0.33414230 \times 10^5$
6	$-0.58711743 \times 10^5$
7	$0.71426686 \times 10^5$
8	$-0.59834012 \times 10^5$
9	$0.33652741 \times 10^5$
10	$-0.12027350 \times 10^5$
11	$0.24348205 \times 10^4$
12	$-0.20807957 \times 10^3$

developed a microscopically based theoretical model, which is limited to pure gases, to viscosity  $\eta$  and thermal conductivity  $\lambda$ , to monatomics in the case of  $\lambda$ , and to the Lennard-Jones 12-6 potential for representing the intermolecular interactions. They started to model the moderately dense gas as a mixture of monomers and dimers and identified three contributions to  $B_{\eta}$ , namely, one from two-monomer collisions  $B_{\eta}^{(2)}$ , another from collisions among three monomers  $B_{\eta}^{(3)}$ , and a contribution resulting from collisions between a monomer and a dimer  $B_{\eta}^{(MD)}$ .

$$B_{\eta} = B_{\eta}^{(2)} + B_{\eta}^{(3)} + B_{\eta}^{(MD)} \quad (5)$$

Because of the uncertainty and complexity of the monomer-dimer collisional process, two potential parameter ratios,  $\delta$  and  $\theta$ , have been

optimized by making use of the theoretically calculated results of the two-monomer and three-monomer contributions to the viscosity and to the translational part of the thermal conductivity for the Lennard-Jones 12-6 potential and by fitting the corresponding expressions of the monomer-dimer contributions to experimental data for both second transport virial coefficients  $B_\eta$  and  $B_\lambda$  ( $B_\lambda$  only for monatomic gases). In this procedure it is assumed that the effective intermolecular potential for the monomer-dimer interaction is related to that of the monomer-monomer one by appropriate scaling.

$$\delta = \frac{\sigma_{\text{MD}}}{\sigma_{\text{M}}} \quad (6)$$

$$\theta = \frac{\varepsilon_{\text{MD}}}{\varepsilon_{\text{M}}} \quad (7)$$

Originally, Friend and Rainwater [14] determined the ratios to be  $\delta = 1.02$  and  $\theta = 1.15$ . The most recent analysis of this kind [9] resulted in the values  $\delta = 1.04$  and  $\theta = 1.25$ . For a comparison of the theoretically predicted values with experimental data, it is convenient to use reduced second viscosity virial coefficients  $B_\eta^*$  and reduced temperatures  $T^*$  [see Eq. (3)].

$$B_\eta^* = \frac{B_\eta}{N_{\text{Av}} \sigma^3} \quad (8)$$

Here  $N_{\text{Av}}$  is Avogadro's constant. The theorem of corresponding states is the basis for the choice of a unique intermolecular pair potential and of common values of  $\delta$  and  $\theta$  for the behavior of a wide variety of substances. A particular advantage of the approach is that the coefficient  $B_\eta(T)$  can be evaluated for a gas for which no experimental viscosity data as a function of density are available. In principle, only knowledge of the Lennard-Jones 12-6 potential parameters  $\sigma$  and  $\varepsilon$  of the substance is needed to calculate  $B_\eta(T)$ .  $\sigma$  and  $\varepsilon$  should be deduced from the temperature dependence of the zero-density viscosity coefficient using preferably values for the temperature range for which  $B_\eta$  is also needed.

For practical applications  $B_\eta^*$  can be represented as a function of the reduced temperature  $T^*$  by the following relation.

$$B_\eta^* = \frac{B_\eta}{0.6022137 \sigma^3} = \sum_{i=0}^n c_i (\sqrt{T^*})^{-i} \quad (9)$$

Here  $B_\eta$  is in units of  $\text{L} \cdot \text{mol}^{-1}$  and  $\sigma$  is in nm. The coefficients  $c_i$  obtained by fitting to the values, which were tabulated by Vogel and Hendl [16] for the optimized parameter ratios  $\delta$  and  $\theta$  by Bich and Vogel, are also given in Table I.

### 3. RESULTS

The experiments were carried out in a quartz oscillating-disk viscometer with small gaps. The basic principles of design and construction [17] and details of calibration and of performance and the evaluation of the measured data have been described previously [18, 19]. The recalibration at room temperature according to the quasi-absolute theory by Newell [20] was performed on nitrogen and argon with reference viscosity values by Kestin et al. [21, 22] characterized by an uncertainty of  $\pm 0.15\%$ . Measurements on these gases between room temperature and 690 K were used as a test of the performance of the viscometer. The uncertainty of the experimental viscosity has been estimated to be  $\pm 0.15\%$  at room temperature and  $\pm 0.3\%$  at the highest temperatures, whereas the reproducibility has been found to be about  $\pm 0.1\%$  covering the whole temperature range.

Five series of measurements, each differing in density  $\rho$ , were performed on propane between room temperature and about 625 K. The viscometer was directly filled from a lecture bottle supplied by Linde AG, Germany, with a certified purity of 99.95%. The actual densities were determined during the filling process by  $p\nu T$  measurements taking into account the real-gas behavior by using either the temperature function of the second pressure virial coefficient  $B$  given by Goodwin and Haynes [23] or, more generally, a 32-term modified Benedict–Webb–Rubin (MBWR) equation of state [24]. The results are recorded in Table II together with the mass and molar densities  $\rho'$  and  $\rho$ , respectively. The temperatures refer to ITS-90.

First, the coefficients of the following equation were fitted to the experimental points of each isochoric series of measurements.

$$\eta(T) = S \exp\left(A \ln T_R + \frac{B}{T_R} + \frac{C}{T_R^2} + D\right) \quad (10)$$

$$S = 10 \mu\text{Pa} \cdot \text{s}, \quad T_R = \frac{T}{298.15}$$

The resulting values of the coefficients of Eq. (10), of the standard deviation  $s_\eta$ , and of the root-mean-square deviation rms are listed for the series in Table III. Because the original experimental points in Table II are not exactly isothermic, the isochoric data were recalculated into isothermic by means of a first-order Taylor series, in terms of temperature:

$$\eta(T_{\text{int}}) = \eta(T_{\text{exp}}) + \left(\frac{\partial \eta}{\partial T}\right)_\rho \Delta T + R_n \quad (11)$$

$$\Delta T = T_{\text{int}} - T_{\text{exp}} \quad (12)$$

Table II. Viscosity of Propane

Temperature $T$ (K)	Viscosity $\eta$ ( $\mu\text{Pa}\cdot\text{s}$ )
Series 1	
$\rho' = 0.455 \text{ kg}\cdot\text{m}^{-3}$	
$\rho = 10.31 \times 10^{-3} \text{ mol}\cdot\text{L}^{-1}$	
297.30	8.117
324.60	8.846
352.99	9.587
381.30	10.330
409.34	11.042
438.07	11.768
466.69	12.476
495.60	13.169
524.95	13.854
546.34	14.364
567.34	14.840
596.43	15.488
625.80	16.125
297.48	8.122
Series 2	
$\rho' = 0.964 \text{ kg}\cdot\text{m}^{-3}$	
$\rho = 21.85 \times 10^{-3} \text{ mol}\cdot\text{L}^{-1}$	
297.42	8.124
324.68	8.860
353.00	9.602
381.03	10.336
409.48	11.065
437.80	11.782
466.24	12.484
495.27	13.185
525.02	13.881
545.47	14.372
567.06	14.863
596.59	15.521
625.64	16.156
297.36	8.124

Table II. (Continued)

Temperature $T$ (K)	Viscosity $\eta$ ( $\mu\text{Pa} \cdot \text{s}$ )
Series 3	
$\rho' = 1.464 \text{ kg} \cdot \text{m}^{-3}$	
$\rho = 33.19 \times 10^{-3} \text{ mol} \cdot \text{L}^{-1}$	
297.51	8.122
324.85	8.857
353.11	9.599
380.94	10.328
409.42	11.061
438.95	11.792
466.63	12.490
495.62	13.190
526.17	13.899
546.11	14.385
566.89	14.855
595.93	15.505
625.37	16.145
297.36	8.120
Series 4	
$\rho' = 1.955 \text{ kg} \cdot \text{m}^{-3}$	
$\rho = 44.32 \times 10^{-3} \text{ mol} \cdot \text{L}^{-1}$	
298.96	8.150
324.65	8.852
352.93	9.593
380.86	10.330
409.72	11.068
437.74	11.778
466.25	12.487
495.37	13.189
524.73	13.879
545.58	14.376
567.04	14.861
596.40	15.521
625.65	16.159
297.24	8.111



Table II. (Continued)

Temperature $T$ (K)	Viscosity $\eta$ ( $\mu\text{Pa}\cdot\text{s}$ )
Series 5 $\rho' = 2.238 \text{ kg}\cdot\text{m}^{-3}$ $\rho = 50.75 \times 10^{-3} \text{ mol}\cdot\text{L}^{-1}$	
297.47	8.116
324.61	8.848
352.51	9.582
380.60	10.326
408.98	11.057
437.70	11.788
466.06	12.492
494.94	13.194
523.47	13.859
545.29	14.378
566.45	14.857
595.44	15.503
625.00	16.145
297.48	8.112

Table III. Parameters of Eq. (10) for the Series of Measurements on Propane

Series	$A$	$B$	$C$	$D$	Root mean square deviation	
					SD $10^3 s_\eta$ ( $\mu\text{Pa}\cdot\text{s}$ )	$10^2$ rms (%)
1	0.450660	-1.078766	0.278452	0.594581	4.45	3.00
2	0.476486	-1.002215	0.249464	0.547659	4.52	3.11
3	0.469036	-1.031275	0.261557	0.564098	6.84	4.48
4	0.476905	-0.993872	0.242082	0.545207	4.51	3.66
5	0.400821	-1.198300	0.307594	0.684060	4.01	3.00
+ <sup>a</sup>	0.478941	-1.006374	0.257241	0.544167	5.02	3.20
+ + <sup>b</sup>	0.477222	-0.987967	0.240322	0.541409	4.85	3.34

<sup>a</sup> At zero density ( $297 \leq T \leq 625$  K).<sup>b</sup> At 0.1013 MPa ( $297 \leq T \leq 625$  K).

The interpolation temperature  $T_{\text{int}}$  corresponds to the mean temperature of the experimental values of the five series of measurements. The temperature derivative of viscosity needed in Eq. (11) was evaluated with the coefficients of Eq. (10) given in Table III. It was proven that the remainder  $R_n$  is negligible compared with the experimental uncertainty. Then zero-density and initial-density viscosity coefficients  $\eta_0$  and  $\eta_1$  were deduced by fitting Eq. (1) to the quasi-experimental viscosity values of the isotherms between 297 and 625 K. In Table IV the least-squares values of  $\eta_0$  and  $\eta_1$  are given, together with their individual standard deviations  $s_{\eta_0}$  and  $s_{\eta_1}$  and with the standard deviation  $s_\eta$  for the isotherm. In Fig. 1 the initial density dependence of viscosity is shown for some isotherms. Whereas open circles are the quasi-experimental values, filled circles represent values at densities corresponding to atmospheric pressure and including the temperature function of second pressure virial coefficient  $B$ . Viscosity values resulting from Eq. (1) are plotted as straight lines. The error bars in this figure indicate an uncertainty of 0.1%. The viscosity coefficients in the limit of zero density  $\eta_0$  and at 0.1013 MPa were again correlated using Eq. (10). The coefficients obtained are also recorded in Table III.

**Table IV.** Zero-Density and Initial-Density Viscosity Coefficients of Propane for the Isotherms According to Eq. (1)

Temperature $T$ (K)	Zero-density viscosity $\eta_0 \pm s_{\eta_0}$ ( $\mu\text{Pa} \cdot \text{s}$ )	Initial-density viscosity $\eta_1 \pm s_{\eta_1}$ ( $\mu\text{Pa} \cdot \text{s} \cdot \text{L} \cdot \text{mol}^{-1}$ )	SD $10^3 s_\eta$ ( $\mu\text{Pa} \cdot \text{s}$ )
297.73	$8.134 \pm 0.005$	$-0.262 \pm 0.144$	4.71
324.68	$8.853 \pm 0.006$	$-0.024 \pm 0.159$	5.22
352.91	$9.590 \pm 0.006$	$0.089 \pm 0.177$	5.80
380.95	$10.322 \pm 0.005$	$0.257 \pm 0.135$	4.43
409.39	$11.045 \pm 0.007$	$0.437 \pm 0.199$	6.52
438.05	$11.765 \pm 0.012$	$0.520 \pm 0.327$	10.73
466.37	$12.466 \pm 0.006$	$0.625 \pm 0.178$	5.84
495.36	$13.160 \pm 0.008$	$0.790 \pm 0.238$	7.80
524.87	$13.849 \pm 0.009$	$0.806 \pm 0.252$	8.28
545.76	$14.351 \pm 0.009$	$0.755 \pm 0.255$	8.36
566.96	$14.833 \pm 0.010$	$0.706 \pm 0.281$	9.22
596.16	$15.483 \pm 0.009$	$0.770 \pm 0.267$	8.77
625.49	$16.121 \pm 0.012$	$0.770 \pm 0.332$	10.90
297.38	$8.126 \pm 0.005$	$-0.272 \pm 0.131$	4.31

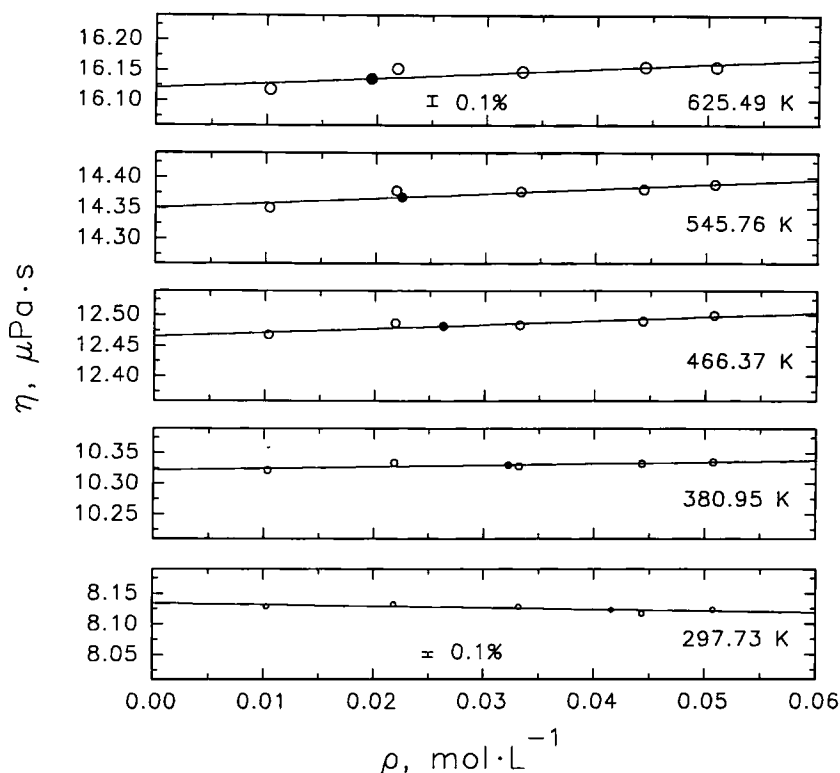


Fig. 1. Viscosity of propane as a function of molar density for several isotherms.  $\eta = \eta_0 + \eta_1 \rho$ ; (●) at 0.1013 MPa, calculated.

#### 4. ANALYSIS AND LOW-DENSITY DATA CORRELATION

The Lennard-Jones 12-6 potential parameters for propane were deduced from the temperature function  $\eta_0(T)$  of the present paper using Eqs. (2) and (3).

$$\varepsilon/k = 285.60 \text{ K}, \quad \sigma = 0.49154 \text{ nm}$$

Reduced values of  $B_\eta$  and of  $T$  were calculated via Eqs. (8) and (3) by means of these potential parameters. In Fig. 2 the reduced second viscosity virial coefficients for propane as well as for ethane [25], *n*-pentane [26], *n*-hexane [27], and *n*-heptane [26], also experimentally determined in our laboratory, are compared with the Rainwater-Friend theory. The theoretical curve for the ratios  $\delta$  and  $\theta$  obtained by Bich and Vogel [9] gives an excellent representation of the experimental  $B_\eta$  data of the *n*-alkanes.

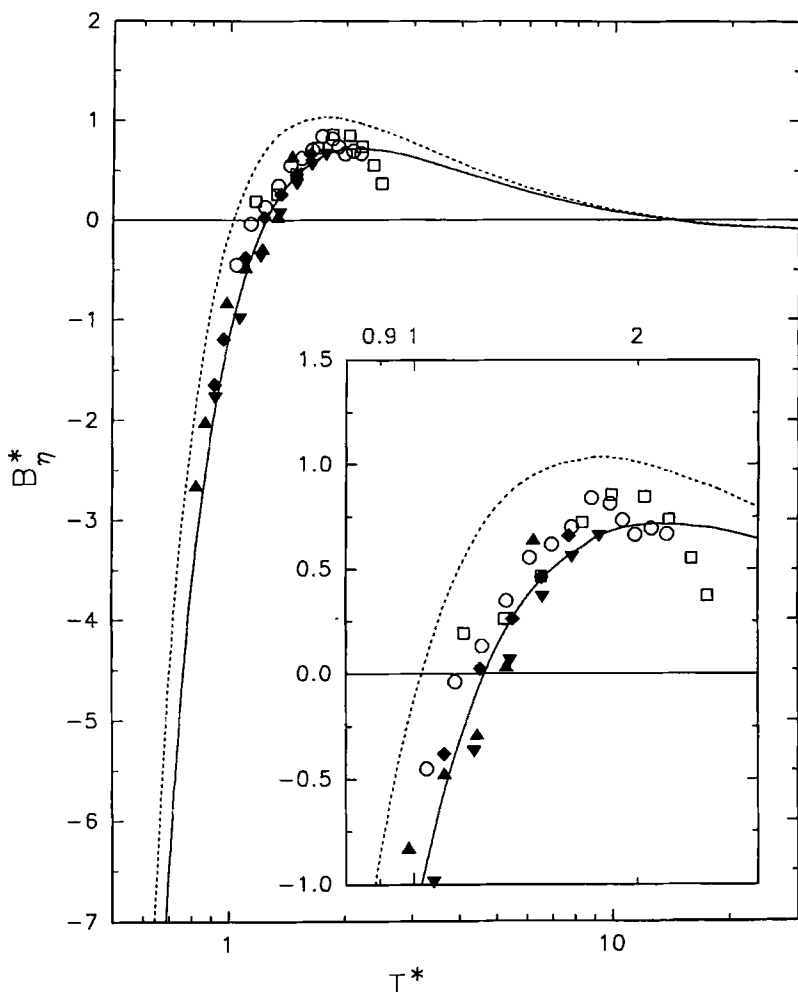


Fig. 2. Reduced second viscosity virial coefficient  $B_{\eta}^*$  as a function of the reduced temperature  $T^*$  for the Lennard-Jones 12-6 potential. (○) Propane, present work; (□) ethane [25]; (▼) *n*-pentane [26]; (◆) *n*-hexane [27]; (▲) *n*-heptane [26]; (.....) Rainwater-Friend theory [14, 15] ( $\delta = 1.02$ ,  $\theta = 1.15$ ); (—) Rainwater-Friend theory, Bich and Vogel [9] ( $\delta = 1.04$ ,  $\theta = 1.25$ ).

The new experimental results are the only data at moderate densities in the open literature that are suitable for an exact evaluation in the limit of zero density because a sufficient number of low-density points of high accuracy along isotherms are available to resolve both slope  $\eta_1$  and zero-density limit  $\eta_0$ . Furthermore, these data cover a larger temperature range

Table V. List of Available Data from Measurements of the Viscosity of Propane at Low Density

Reference	Technique <sup>a</sup>	Temperature (K)	Number of points	Ascribed accuracy (%)
Klemenc and Remi [28]	C	273	1	2.0
Titani [29]	C	293–393	6	2.0
Trautz and Kurz [7]	C	301–549	6	1.0–2.2
Trautz and Sorg [8]	C	291–524	6	1.0–2.2
Adzumi [30]	C	303–373	8	3.0
Sage and Lacey [31]	RB	311–378	10	5.0
Wobser and Mueller [32]	RB	293–371	5	0.5
Senftleben and Gladisch [33]	HT	313	1	5.0
Golubev and Petrov [34]	C	298–523	8	1.0–2.2
Lambert et al. [35]	OP, C	308–364	7	3.0
Carmichael et al. [36]	RC	278–478	6	2.0
Kestin et al. [21]	OD	296–303	2	0.2
Diaz Pena and Cheda [37]	C	304–408	11	3.0
Diaz Pena and Cheda [38]	C	373	1	3.0
Kestin et al. [39]	OD	299–478	5	0.2–0.6
Abe et al. [40]	OD	298–468	5	0.2–0.6
Abe et al. [41]	OD	298–468	6	0.2–0.6
Nagaoka et al. [42]	RB	298–348	3	2.0
Vogel (present paper)	OD	297–625	14	0.2–0.4

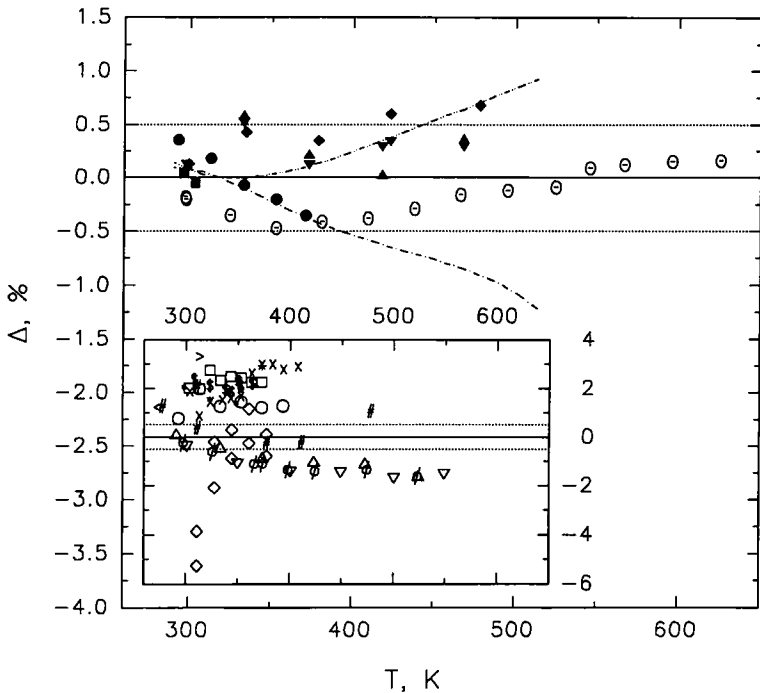
<sup>a</sup> C, capillary; RB, rolling ball; HT, heat transfer between concentric cylinders; OP, oscillating pendulum; RC, rotating cylinder; OD, oscillating disk.

than those of all other sources [7, 8, 21, 28–42] listed in Table V together with the method of measurement, the temperature range covered, and the ascribed accuracy of the data.

In order to compare the older data with the new ones on a common basis, all data at densities up to  $0.15 \text{ mol} \cdot \text{L}^{-1}$ , whose numbers are also listed in Table V, were corrected to the limit of zero density. For this purpose the densities were calculated with the MBWR equation of state [24] for temperatures corresponding to the IPTS-68 from the information given in the papers. With the coefficients  $c_i$  of Eq. (9) the second viscosity virial coefficients  $B_{\eta}$  were calculated by means of the Lennard–Jones 12–6 potential parameters and used in Eq. (1) to calculate the zero-density viscosity coefficients at temperatures corrected to ITS-90. Although for the temperature range under discussion the effect of the initial density dependence of viscosity is small compared with the experimental uncertainties of most

authors, the correction was performed because it is a systematic one. Experimental low-density viscosity data have been treated in this manner for the first time.

In the process of data analysis most data sets have been classified as secondary data, because these measurements have been performed in instruments for which either a complete working equation was unavailable or not all necessary corrections were applied. Furthermore, the precision of the data was tested on the basis of a comparison for monatomic gases mostly measured by the same authors. The consistency test with independent values derived from the HFD potentials for the monatomics



**Fig. 3.** Deviations of viscosity data from the present correlation. Primary data: (●) Wobser and Mueller [32]; (■) Kestin et al. [21]; (◆) Kestin et al. [39]; (▲) Abe et al. [40]; (▼) Abe et al. [41]; (⊖) Vogel (present paper). Correlations: (---) Tanaka and Makita [5]; (-·-·-) Holland et al. [6]. Secondary data (inset): (<) Klemenc and Remi [28]; (○) Titani [29]; (▽) Trautz and Kurz [7]; (△) Trautz and Sorg [8]; (□) Adzumi [30]; (◇) Sage and Lacey [31]; (>) Senftleben and Gladisch [33]; (φ) Golubev and Petrov [34]; (\$) Lambert et al. [35]; (#) Carmichael et al. [36]; (×) Diaz Pena and Cheda [37]; (+) Diaz Pena and Cheda [38]; (\*) Nagaoka et al. [42].  $\Delta = 100(\eta_{exp} - \eta_{corr})/\eta_{exp}$ .

confirmed that, apart from the data by Kestin and collaborators [21, 39–41] and the 14 zero-density viscosity values of the present paper, only the data of Wobser and Mueller [32], could be included in the primary data sets. In the fitting procedure based on the least-squares method an individual statistical weight according to the ascribed accuracy has been assigned to each experimental  $\xi_{\eta}$  value, and the scaling parameters have been determined to be

$$\varepsilon/k = 295.85 \text{ K}, \quad \sigma = 0.49119 \text{ nm}$$

The deviations of the primary data sets from the correlation are shown in Fig. 3. It emerges that the mutual inconsistencies between the data of Kestin and co-workers [21, 39–41] and the present experimental results are a little larger than the ascribed experimental uncertainties. The reason is a temperature measurement error in the viscometer of Kestin and collaborators that could be verified by a consistency test on the basis of the extended theorem of corresponding states between their data and values following from the HFD potentials for monatomic gases [13]. Although the influence of Kestin's data on the correlation is relatively strong, we can estimate the uncertainty of the representation between 293 and 625 K to be  $\pm 0.5\%$ . Figure 3 also shows a comparison with two correlations. The correlation of Tanaka and Makita [5] is obviously badly influenced by the error in the data of Trautz et al. [7, 8], whereas that of Holland et al. [6] is practically based only on the data of Kestin et al. [39]. The deviations of the secondary data sets presented in the inset to Fig. 3 are larger, amounting mostly to positive values of 1–3%. It emerges that the data of Golubew and Petrov [34] do not seem to have been determined independently. They correspond completely to the data of Trautz et al. [7, 8].

## 5. CONCLUSIONS

The zero-density limit is the natural starting point for the development of a representation of any viscosity surface. Viscosity measurements should be performed in a range of moderately low densities in order to obtain both leading terms in the density series. The representation of the initial density dependence of viscosity by the Rainwater–Friend theory can be used to deduce viscosity values in the limit of zero density from earlier experiments, particularly in the vapor phase, provided that there is enough information about the measurements. The representation of the viscosity in the limit of zero density should preferably be based on a universal correlation according to the extended principle of corresponding states.

The correlation presented for propane fulfills these requirements. It is based on a small body of experimental data that have been critically assessed for internal consistency and for agreement with theory in the zero-density limit.

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