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The Viscosity of Gaseous *n*-Butane and Its Initial Density Dependence[1]

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New relative high-precision measurements of the viscosity η of gaseous *n*-butane were carried out in an oscillating-disk viscometer. Seven series of measurements were performed between 298 and 627 K in the density range from 0.01 to 0.05 $\mathrm{mol \cdot L^{-1}}$. Isotherms recalculated from the original experimental data were analyzed with a first-order expansion, in terms of density, for the viscosity. Reduced values of the second viscosity virial coefficient deduced from the zero-density and initial-density viscosity coefficients for *n*-butane are in good agreement with the representation of the Rainwater–Friend theory. The new experimental data and some data sets from the literature were used to develop a representation for the viscosity of *n*-butane in the limit of zero density on the basis of the extended principle of corresponding states. It has been shown that an individual correlation is needed to represent the experimental data between 293 and 627 K with an uncertainty of ±0.4%.

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

1. INTRODUCTION

In a program initiated under the auspices of IUPAC, the viscosity surfaces of ethane [1] and propane [2] were studied in the series of the *n*-alkanes. This program is now extended to *n*-butane, which as a natural gas component and an environmentally friendly alternative refrigerant, is an important industrial fluid for which thermophysical properties are required over the widest range of thermodynamic states possible.

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Earlier viscosity measurements on *n*-butane in the low-density range suffered from three problems. First, the measurements were not extended to high temperatures. Second, there existed only a limited number of reliable values to be included in the primary data sets for the correlation. Third, isotherms with a sufficient number of precise data to evaluate with a first-order expansion in terms of density have not been measured. Such data are needed, because the correlation of a transport-property surface should be started at low densities.

The new viscosity measurements at relatively low densities are suitable to determine both leading terms in the density expansion for the viscosity. The viscosity in the limit of zero density is to be correlated on the basis of the extended theorem of corresponding states. But the specific interactions between *n*-butane molecules require an individual correlation. Individual parameters are also needed for the description of the initial density dependence of the viscosity. The representation proposed in this paper for the low-density range differs from the earlier correlation given by Younglove and Ely [3] by considering the initial density dependence of viscosity in a novel and correct way [4].

2. METHODOLOGY OF DATA CORRELATION

The viscosity η of a moderately dense gas may be represented as a function of the temperature T and molar density ρ by a density expansion limited to the first power:

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

Here η_0 and η_1 are the zero-density and initial-density viscosity coefficients, and B_η is the second viscosity virial coefficient.

$\eta_0(T)$ is not directly accessible experimentally, but it follows from extrapolation of the results of low-density measurements. According to the well-established kinetic theory of dilute gases, $\eta_0(T)$ is given, in practical engineering form [5], by

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

where M is the molar mass in $\mathrm{g \cdot mol^{-1}}$, T is in K, and η is in units of $\mu\mathrm{Pa \cdot s}$.

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

The notation $\mathfrak{S}_\eta^*$ represents the reduced effective collision cross section $\mathfrak{S}^*(2000)$ and the correction factor for higher-order approximations. T^* is the reduced temperature, and ε/k_B (in K) is an energy scaling parameter or the well depth of an assumed intermolecular potential. σ is a length scaling parameter or the collision diameter in nm.

The temperature dependence of $\mathfrak{S}_\eta$ may be represented in reduced functional form:

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

To develop a viscosity correlation the available experimental data are divided into primary and secondary data sets based on a critical assessment of the methods of measurements with regard to the completeness of the measurement theory and to the achieved precision. Then experimental $\mathfrak{S}_\eta$ values ($\mathfrak{S}_\eta = \pi\sigma^2\mathfrak{S}_\eta^*$) are derived from selected η data and fitted, using appropriate statistical weights, to Eq. (4). Since the kinetic theory for polyatomic gases like *n*-butane is still in its formative stages, a modified analysis has to be performed in two steps based on an empirical extension of the extended corresponding-states principle. We adopted a universal correlation on the basis of this principle derived by Bich et al. [6], who determined simultaneously the coefficients a_i of Eq. (4) as well as σ and ε/k_B in Eqs. (2) and (3) for the noble gases in the temperature range $1.2 < T^* < 10$. In the first step, σ and ε/k_B are determined from $\eta_0(T)$ using the known coefficients a_i. In the second step, the determined scaling factors are assumed to be fixed and new coefficients a_i are deduced by fitting the quasi-experimental $\mathfrak{S}_\eta$ values to Eq. (4). The resulting representation is specific for the considered substance and is called an individual correlation.

The temperature function, $B_\eta(T)$, which characterizes the viscosity of moderately dense gases, can be represented by a microscopically based theoretical model derived by Rainwater and Friend [7, 8] using the Lennard-Jones 12-6 potential. In this theory two potential parameter ratios, δ and θ, had to be determined by fitting the theoretical expressions to experimental B_η data for monatomic and polyatomic gases as well as to experimental B_λ values for monatomic gases (λ—thermal conductivity). Bich and Vogel [9] presented tables of B_η^* [Eq. (5)] as a function of T^* [see Eq. (3)] for two sets of δ and θ. For practical applications in computer codes, B_η^* can be represented using the correlation

$$B_\eta^* = \frac{B_\eta}{N_A\sigma^3} = \sum_{i=0}^{n} c_i(T^*)^{-i/2} \tag{5}$$

The coefficients c_i are given in Table 5.3. of Ref. 9 for the optimized parameter ratios, $\delta = 1.04$ and $\theta = 1.25$ by Bich and Vogel [4] (B_η in units of $\mathrm{L \cdot mol^{-1}}$).

3. EXPERIMENTAL RESULTS

The experiments were carried out in a quartz oscillating-disk viscometer with small gaps. The basic principles of design and construction [10], as well as the details of performance of relative measurements [11, 12], have been described previously. In accordance with the experiments on propane [13], the uncertainty of the viscosity data has been estimated to be ±0.15% at room temperature and ±0.2 to 0.3% at the highest temperatures, whereas the reproducibility has been found to be about ±0.1% covering the whole temperature range. Seven series of measurements were performed on *n*-butane for densities between 0.009 and 0.047 $\mathrm{mol \cdot L^{-1}}$ in the temperature range from 298 to 627 K. The actual densities were determined during the filling process by $p\rho T$ measurements taking into account the real-gas behavior [3]. The certified purity of *n*-butane supplied by UCAR, USA, amounts to 99.95%. Table I summarizes the experimental data.

As the values in Table I are not exactly isothermal, the isochoric data were adjusted to isothermal values by means of a first-order Taylor series, in terms of temperature:

$$\eta(T_{\mathrm{int}}) = \eta(T_{\mathrm{exp}}) + \left(\frac{\partial \eta}{\partial T}\right)_\rho \Delta T + R_n \tag{6}$$

where

$$\Delta T = T_{\mathrm{int}} - T_{\mathrm{exp}} \tag{7}$$

The interpolation temperature T_{int} corresponds to the mean temperature for the points of the seven series. The temperature derivative of η needed in Eq. (6) was evaluated with the coefficients of Eq. (8), which were obtained by a fit to the original isochoric data.

$$\eta(T) = S \exp\left(A \ln T_{\mathrm{R}} + \frac{B}{T_{\mathrm{R}}} + \frac{C}{T_{\mathrm{R}}^2} + D\right) \quad \text{and} \quad T_{\mathrm{R}} = \frac{T}{298.15} \tag{8}$$

Then η_0 and η_1 were deduced by fitting Eq. (1) to the quasi-experimental viscosity values of the isotherms. In Table II the least-squares values of η_0 and η_1 are given, together with their individual standard deviations s_{η_0} and s_{η_1} and with the standard deviation s_η for each isotherm.

Table I. Viscosity of *n*-Butane

Temperature T (K)	Viscosity η (μPa · s)
Series 1 $\rho = 8.79 \times 10^{-3}$ mol · L^{-1}	
298.75	7.432
325.44	8.093
353.67	8.769
382.95	9.471
410.83	10.136
438.85	10.805
470.13	11.541
496.47	12.140
526.82	12.845
546.66	13.313
568.13	13.810
597.66	14.490
626.84	15.136
298.15	7.419
Series 2 $\rho = 8.98 \times 10^{-3}$ mol · L^{-1}	
298.24	7.426
325.68	8.103
353.92	8.782
382.81	9.474
410.00	10.131
440.11	10.842
467.36	11.481
496.84	12.158
527.31	12.841
547.92	13.327
568.78	13.802
597.47	14.454
627.13	15.116
330.04	8.198
Series 3 $\rho = 17.72 \times 10^{-3}$ mol · L^{-1}	
298.87	7.420
325.02	8.087
353.55	8.779
381.52	9.457

Table I. *(Continued)*

Temperature T (K)	Viscosity η (μPa · s)
409.78	10.140
438.45	10.822
466.85	11.481
495.83	12.156
526.76	12.860
546.87	13.320
568.04	13.801
595.83	14.438
625.29	15.100
325.20	8.092
Series 4 $\rho = 27.28 \times 10^{-3}$ mol · L^{-1}	
298.45	7.418
325.17	8.076
353.06	8.758
383.28	9.496
410.45	10.136
438.82	10.821
467.00	11.477
495.93	12.146
525.26	12.802
546.27	13.295
567.18	13.771
595.75	14.436
626.10	15.118
297.68	7.397
Series 5 $\rho = 34.19 \times 10^{-3}$ mol · L^{-1}	
299.36	7.433
325.43	8.080
353.36	8.757
384.10	9.515
409.63	10.126
438.90	10.819
467.43	11.488
496.15	12.156
527.60	12.872

Table I. *(Continued)*

Temperature T (K)	Viscosity η (μPa · s)
546.69	13.325
568.33	13.815
598.25	14.498
626.87	15.123
298.27	7.407
Series 6 $\rho = 40.26 \times 10^{-3}$ mol · L^{-1}	
299.12	7.433
324.89	8.070
352.94	8.766
381.88	9.463
409.87	10.141
438.55	10.824
467.14	11.494
496.29	12.165
525.60	12.844
545.96	13.324
568.43	13.846
596.55	14.482
625.15	15.142
297.77	7.397
Series 7 $\rho = 46.47 \times 10^{-3}$ mol · L^{-1}	
297.85	7.392
324.49	8.054
354.14	8.792
381.44	9.449
409.67	10.135
437.94	10.812
466.46	11.488
496.03	12.175
525.50	12.854
546.71	13.346
567.43	13.832
595.92	14.491
625.74	15.155
297.72	7.390

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

Temperature T (K)	Zero-density viscosity $\eta_0 \pm s_{\eta_0}$ (μPa · s)	Initial-density viscosity $\eta_1 \pm s_{\eta_1}$ (μPa · s · L · mol^{-1})	Standard deviation, $10^3 s_\eta$ (μPa · s)
298.52	7.437 ± 0.005	−0.588 ± 0.158	5.78
325.16	8.094 ± 0.003	−0.497 ± 0.109	4.01
353.52	8.768 ± 0.006	0.132 ± 0.204	7.50
382.57	9.467 ± 0.005	0.301 ± 0.170	6.25
410.03	10.124 ± 0.008	0.435 ± 0.259	9.49
438.80	10.807 ± 0.007	0.527 ± 0.221	8.11
467.48	11.476 ± 0.005	0.636 ± 0.182	6.68
496.22	12.135 ± 0.007	0.820 ± 0.249	9.14
526.41	12.819 ± 0.011	1.018 ± 0.355	13.01
546.73	13.295 ± 0.008	0.998 ± 0.275	10.10
568.05	13.780 ± 0.012	1.199 ± 0.415	15.23
596.78	14.438 ± 0.012	1.225 ± 0.417	15.31
626.16	15.091 ± 0.016	1.388 ± 0.529	19.40
297.92	7.417 ± 0.003	−0.459 ± 0.078	2.27

4. ANALYSIS OF $B_\eta(T)$

Our experimental results and the values of Kestin and Yata [14] are the only data at moderate densities that are suitable for an accurate evaluation in the limit of zero density. Both data sets are characterized by a sufficient number of low-density points of high accuracy along isotherms to resolve both the slope η_1 and the zero-density limit η_0. In order to compare B_η [Eq. (1)] with predicted reduced values B_η^* [Eq. (5)] from the Rainwater–Friend theory, Lennard–Jones 12–6 potential parameters for *n*-butane are needed. They have been determined from $\eta_0(T)$ in this paper, using Eqs. (2) and (3), to be

$$\varepsilon/k_B = 328.68 \text{ K} \qquad \text{and} \qquad \sigma = 0.53272 \text{ nm}$$

Figure 1 compares the curves of the theoretical results for the ratios, δ and θ, obtained originally by Rainwater and Friend [7, 8] and more recently by Bich and Vogel [4], with B_η^* values for *n*-butane of this paper and of Kestin and Yata [14] as well as for ethane [15] and propane [13]. The comparision shows that the B_η^* values for the three *n*-alkanes, all reduced with Lennard–Jones 12–6 parameters from $\eta_0(T)$, do not completely agree

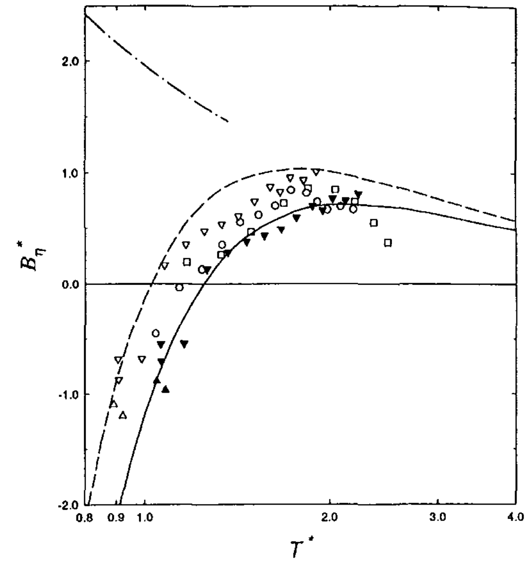

Fig. 1. Reduced second viscosity virial coefficient B^*_η as a function of the reduced temperature T^* for the Lennard-Jones 12-6 potential. (□) Ethane [15]; (○) propane [13]; (▽) *n*-butane, present paper; (△) *n*-butane [14]; (▼) *n*-butane, optimized scaling factors, present paper; (▲) *n*-butane, optimized scaling factors [14]; (-·-·-) *n*-butane, optimized scaling factors, Younglove-Ely correlation [3], (---) Rainwater-Friend theory [7, 8] ($\delta = 1.02$, $\theta = 1.15$); (—) Rainwater-Friend theory [4] ($\delta = 1.04$, $\theta = 1.25$).

with any of the theoretical curves. The disagreement is due to the uncertainty of the experimental data and to the fact that the Lennard-Jones 12-6 potential model is inadequate to represent the intermolecular interactions of polyatomic gases. Since the experimental information is to be used for a viscosity surface correlation with the goal of extrapolating to temperatures outside the measurement ranges, it is advisable to derive scaling factors ε/k_B and σ in a fit of one of the theoretical curves to the experimental $B_\eta(T)$ data. Thus, we chose $B^*_\eta(T^*)$ according to Eq. (5) with the coefficients c_i of Bich and Vogel [9] and obtained, using the data from this paper and those of Kestin and Yata for *n*-butane:

$$\varepsilon/k_B = 280.51 \text{ K} \qquad \text{and} \qquad \sigma = 0.57335 \text{ nm}$$

The temperature dependence of the experimental B^*_η values using this procedure agrees, in general, with that of the Rainwater-Friend theory. But

Fig. 1 shows that the B_η^* values from the Younglove–Ely correlation [3] are also in disagreement with that theory.

5. ZERO-DENSITY CORRELATION

Our data cover a larger temperature range than those of all other sources combined. A list of data from the literature on viscosity measurements of *n*-butane at low densities has been compiled (Table III). The list includes the method of measurement, the temperature range, the number of data points at densities up to 0.10 $\text{mol} \cdot \text{L}^{-1}$, the ascribed accuracy of the data, and the classification as primary or secondary data sets.

In order to compare our data with those from the literature on a common basis, the last-mentioned values were corrected to the limit of zero

Table III. Available Data from Viscosity Measurements on *n*-Butane at Low Density

First author [Ref. No.]	Technique[a]	Temperature (K)	Number of points	Ascribed accuracy (%)
Primary data sets				
Kestin (1968) [14]	OD	293–303	7[b]	0.2
Kestin (1971) [16]	OD	297–303	2	0.2
Abe (1979) [20]	OD	298–468	7	0.2–0.6
Present paper	OD	298–627	14	0.2–0.6
Secondary data sets				
Kuenen (1913) [21]	C	289–373	2	3.0
Titani (1930) [22]	C	293–393	6	2.0
Sage (1939) [23]	RB	311–378	23[c]	5.0
Wobser (1941) [24]	RB	293–371	5	1.0
Senftleben (1949) [25]	HT	313	1	5.0
Senftleben (1953) [26]	HT	303	1	5.0
Lambert (1955) [27]	OP, C	308–363	11	3.0
Carmichael (1963) [28]	RC	278–478	9[d]	1.5
Dolan (1963) [29]	C	300–444	10	3.0
Diaz Pena (1975) [30]	C	313–403	10	3.0
Diaz Pena (1975) [31]	C	373	1	3.0
Kestin (1977) [17]	OD	299–478	5	0.2–0.6
Abe (1978) [18]	OD	298–468	5	0.2–0.6
Abe (1979) [19]	OD	298–468	6	0.2–0.6

[a] C, capillary; HT, heat transfer between concentric cylinders; OD, oscillating disk; OP, oscillating pendulum; RB, rolling ball; RC, rotating cylinder.

[b] Two values by extrapolation of isotherms.

[c] Five values by extrapolation of isotherms.

[d] Two measuring points, four values by extrapolation of isotherms.

density using the information on the initial density dependence of viscosity given in the foregoing section. Although for the temperature range under discussion this effect is small compared with the experimental uncertainties of most authors, the correction was performed because it is a systematic one. In this procedure the densities were calculated with the MBWR equation of state [3] for temperatures corresponding to the IPTS-68 from the information given in the papers followed by a correction of the temperatures to ITS-90.

In principle, only data of Kestin and co-workers [14, 16–20] might be classified as primary ones. But there are two problems with these data. First, several data sets exist in the same temperature range, so that their influence would be overrated. Second, the deviations between different data sets amount to about 1%, although Kestin and coworkers claimed an accuracy of ±0.1 to 0.3% for their data. Thus, we were forced to increase the ascribed accuracy to ±0.2 to 0.6% and to include only the data at room temperature [14, 16] and the most recent data of Abe et al. [20] between 298 and 468 K into the primary data sets.

According to the methodology described in Section 2, the scaling factors of the universal correlation have been determined in the first step to be

$$\varepsilon/k_B = 330.51 \text{ K} \qquad \text{and} \qquad \sigma = 0.53649 \text{ nm}$$

It clearly emerges that the universal correlation is not suitable to represent the primary data sets within their uncertainties in the temperature range covered by the experiments (see Fig. 2). For this reason an individual correlation was carried out with ε/k and σ to be fixed. The following coefficients a_i of Eq. (4) were derived:

$$a_0 = 0.2229039, \qquad a_1 = -0.4809052, \qquad a_2 = 0.1339312,$$

$$a_3 = -0.2548478, \qquad a_4 = 0.1941395$$

The deviations of the primary and secondary data sets from the individual correlaion are shown in Fig. 2, including a comparison with the universal correlation and with the recommended data of Younglove and Ely [3] and by Tarzimanov et al. [32]. The values of Tarzimanov et al. turn out to be generally too high. The agreement with the Younglove–Ely correlation is quite reasonable, although it is based on experimental data only up to 480 K. However, there is a tendency to higher values for Younglove and

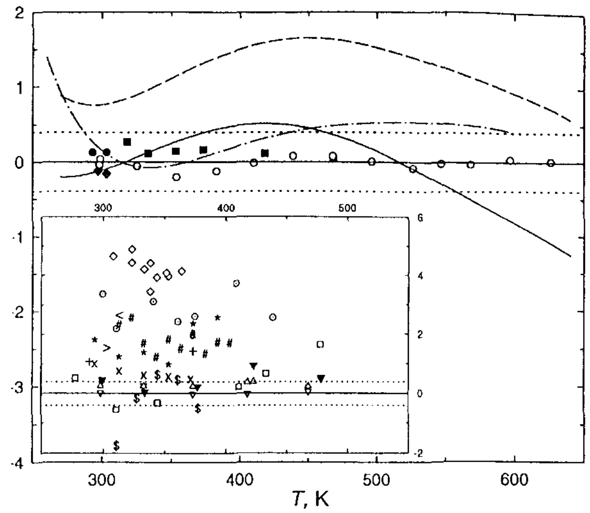

Fig. 2. Deviations [$\Delta = 100 \cdot (\eta_{exp} - \eta_{corr})/\eta_{exp}$] of viscosity data from the present correlation. Primary data: (●) Kestin and Yata [14]; (◆) Kestin et al. [16]; (■) Abe et al. [20]; (○) present paper. Correlations: (- -) universal correlation; (-----) Younglove and Ely [3]; (——) Tarzimanov et al. [32]. Secondary data (in set): (+) Kuenen and Visser [21]; (★) Titani [22]; ($) Sage et al. [23]; (×) Wobser and Mueller [24]; (<) Senftleben and Gladisch [25]; (>) Senftleben [26]; (◇) Lambert et al. [27]; (□) Carmichael and Sage [28]; (⊙) Dolan et al. [29]; (#) Diaz Pena and Cheda [30]; (&) Diaz Pena and Cheda [31]; (▼) Kestin et al. [17]; (▽) Abe et al. [18]; (△) Abe et al. [19].

Ely at low temperatures. The uncertainty of the new correlation for the zero-density viscosity between 293 and 625 K is estimated to be ±0.4%.

6. CONCLUSION

The representation of the viscosity surface should separately take into account both leading terms of the density series. In the case of polyatomic gases the representation of the viscosity in the limit of zero density is preferably to be based on an individual correlation instead of a universal one according to the extended principle of corresponding states. The initial density dependence of viscosity is to be modeled from the Rainwater–Friend theory including optimized scaling factors. Only a small body of experimental values for *n*-butane from the literature could be characterized as primary data.

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