

# Reference Correlation of the Viscosity of Propane

E. Vogel, C. Küchenmeister, and E. Bich

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

A. Laesecke<sup>a)</sup>

Physical and Chemical Properties Division, National Institute of Standards and Technology, Boulder, Colorado 80303

Received June 15, 1998; final manuscript received June 29, 1998

A new representation of the viscosity of propane includes a zero-density correlation and an initial-density dependence correlation based on the kinetic theory of dilute gases and on the Rainwater–Friend theory. The higher density contributions of the residual viscosity in the representation are formed by a combination of double polynomials in density and reciprocal temperature, and a free-volume term with a temperature-dependent close-packed density. The full surface correlation is based on a set of primary experimental data selected as a result of a critical assessment of the available information from 37 original viscosity studies. The review refers to 96 citations altogether. The validity of the representation extends from the triple point to 600 K and 100 MPa in accordance with the modified Benedict–Webb–Rubin equation of state. The uncertainty of the representation varies from  $\pm 0.4\%$  for the viscosity of the dilute gas phase between room temperature and 600 K, to about  $\pm 2.5\%$  for the range 100–475 K up to about 30 MPa, and to about  $\pm 4\%$  for points outside this range. Tables of the viscosity according to the representative equations at selected temperatures and pressures and along the saturation line provide easy reference as well as the validation of computer codes. © 1998 American Institute of Physics and American Chemical Society. [S0047-2689(98)00405-X]

Key words: critical data assessment, free-volume model, liquid viscosity, propane, vapor viscosity, viscosity correlation.

## Contents

1. Introduction.....	948
2. Equation of State.....	948
3. Experimental Viscosity Data.....	949
3.1. Low-Density Viscosity Data.....	949
3.2. Viscosity Data at Higher Densities.....	951
4. Methodology.....	952
4.1. Viscosity in the Critical Region.....	952
4.2. Viscosity in the Zero-Density Limit.....	953
4.3. Initial Density Dependence of Viscosity.....	954
4.4. Higher-Density Terms of the Residual Viscosity.....	955
5. Results and Discussion.....	956
5.1. Zero-Density Limit and Initial Density Dependence.....	956
5.2. High-Density Region.....	957
5.3. Comparison with the Younglove–Ely Representation.....	959
6. Tabulations and the Overall Representation.....	960
7. Conclusion.....	967
8. Acknowledgments.....	969

## List of Tables

1. List of available data from measurements of the viscosity of propane at low densities $\rho < 0.10$ mol L <sup>-1</sup> .....	950
2. List of available data from viscosity measurements on propane at densities $\rho > 0.10$ mol L <sup>-1</sup> .....	951
3. Coefficients for the representation of the viscosity of propane.....	954
4. Viscosity and density of propane.....	961
5. Viscosity and density of propane along the saturation line.....	968

## List of Figures

1. Distribution of the available experimental viscosity data of propane at densities $\rho > 0.10$ mol L <sup>-1</sup> ; primary data.....	952
2. Distribution of the available experimental viscosity data of propane at densities $\rho > 0.10$ mol L <sup>-1</sup> ; secondary data.....	952
3. Deviations ( $\Delta$ ) of the viscosity data by Starling <i>et al.</i> (1959) and by Starling (1960) from a preliminary correlation.....	953
4. Reduced second viscosity virial coefficient $B_{\eta}^*$	

<sup>a)</sup>Electronic mail: Arno.Laesecke@Boulder.NIST.Gov

©1998 by the U.S. Secretary of Commerce on behalf of the United States. All rights reserved. This copyright is assigned to the American Institute of Physics and the American Chemical Society.

Reprints available from ACS; see Reprints List at back of issue.

as a function of the reduced temperature $T^*$ .....	955
5. Deviations ( $\Delta$ ) of the primary viscosity data and of correlations from the present individual zero-density correlation.....	956
6. Deviations ( $\Delta$ ) of older secondary viscosity data from the present individual zero-density correlation.....	957
7. Deviations ( $\Delta$ ) of more recent secondary viscosity data from the present individual zero-density correlation.....	957
8. Deviations ( $\Delta$ ) of the primary viscosity data by Starling <i>et al.</i> (1959) from the correlation.....	957
9. Deviations ( $\Delta$ ) of the primary viscosity data by Starling (1960) from the correlation.....	958
10. Deviations ( $\Delta$ ) of the secondary viscosity data by Starling <i>et al.</i> (1959) and by Starling (1960) from the correlation.....	958
11. Deviations ( $\Delta$ ) of the primary viscosity data of Carmichael <i>et al.</i> (1964) from the present correlation.....	958
12. Deviations ( $\Delta$ ) of the primary viscosity data of Giddings <i>et al.</i> (1966) from the present correlation.....	958
13. Deviations ( $\Delta$ ) of the primary viscosity data of Huang <i>et al.</i> (1966) and of Strumpf <i>et al.</i> (1974) from the present correlation.....	959
14. Deviations ( $\Delta$ ) of the primary viscosity data of Diller (1982) from the present correlation.....	959
15. Deviations ( $\Delta$ ) of secondary viscosity data from the present correlation (Part I).....	959
16. Deviations ( $\Delta$ ) of secondary viscosity data from the present correlation (Part II).....	959
17. Deviations ( $\Delta$ ) of secondary viscosity data from the present correlation (Part III).....	960
18. Comparison with the correlation by Younglove and Ely (1987).....	960
19. Deviations ( $\Delta$ ) of the viscosity data of Diller (1982) from the Younglove-Ely correlation.....	960
20. The extent of the viscosity representation and its estimated uncertainty.....	968
21. Viscosity surface correlation of propane.....	968

## 1. Introduction

In the last 20 years most assessments of the viscosity of propane in the dense fluid region were confined to limited temperature and pressure ranges. Thus, the correlation by Tanaka and Makita<sup>1</sup> was restricted to the gas phase between 300 and 500 K and 0.1 and 55 MPa. Using graphical smoothing techniques, Stephan and Lucas<sup>2</sup> generated in 1979 a table of the viscosity of propane in the temperature range 175–750 K between 0.1 and 35 MPa. In the same year Holland *et al.*<sup>3</sup> developed a correlation within the temperature range 140–500 K and the pressure range 0.1–50 MPa using the data available to them at that time. The latest representation by Younglove and Ely<sup>4</sup> from 1987 has been extended from the triple point to 600 K and 100 MPa based on the modified

Benedict–Webb–Rubin (MBWR) equation of state. This correlation suffers from having no information about the selection and analysis of the data and little indication of the development of the representative equations and of the tables. But it includes recent experimental data by Diller<sup>5</sup> in the saturated and compressed liquid regions down to 90 K.

The development of state-of-the-art representations of thermophysical properties has gained renewed attention. Therefore, a research program has been initiated under the auspices of the International Union of Pure and Applied Chemistry (IUPAC) in order to develop accurate, consistent and theoretically sound correlations for the transport properties of fluids of industrial importance. Propane has been included in this program for two reasons. An improved viscosity surface correlation is needed as a standard reference formulation to represent the most accurate experimental data within their estimated uncertainty. There is also considerable interest in such a correlation, because propane is used as a reference fluid for the prediction of thermodynamic and transport properties of mixtures on the basis of the extended theorem of corresponding states, such as in the computer program SUPERTRAPP developed by the National Institute of Standards and Technology (NIST).<sup>6–9</sup> The correlation of the reference fluid should safely extrapolate beyond the range, on which it is based on data and sometimes even into its two-phase region. In particular, the second objective influences the functional terms to be selected for an improved correlation.

Vogel and Küchenmeister<sup>10</sup> developed recently a viscosity surface correlation which included new results of precise measurements as well as theoretical advances for the low-density vapor.<sup>11,12</sup> However, the correlation exhibited large loops in the two-phase region which precluded its use as a reference fluid formulation for corresponding-states predictions.<sup>13</sup> These loops were caused by exponential density terms which had been selected for the viscosity correlation in analogy to the terms in the MBWR equation of state by Younglove and Ely.<sup>4</sup>

In this paper a new correlation has been designed to take into consideration the steep increase at low temperatures and higher densities as well as a smooth continuous increase in the two-phase region. It is based on a revised critical assessment of the published experimental data. The representative equations describe the most reliable data sets within their estimated uncertainties and reproduce the phenomenological behavior of propane in the same temperature and pressure ranges as the correlation by Younglove and Ely.<sup>4</sup> But the correlation of this paper has been improved in the vapor and gas phases at moderately low densities and in the compressed liquid region at low temperatures. The analysis presented in the following sections conforms extensively to that adopted in earlier work<sup>14–16</sup> on the representation of the viscosity of fluids.

## 2. Equation of State

An equation of state is an essential part of a pure fluid transport property correlation, since experimental data are

measured in terms of pressure and temperature while the correlation uses the independent variables density and temperature as suggested by kinetic theory. The thermodynamic formulation used for propane in this paper is the MBWR equation of state by Younglove and Ely.<sup>4</sup> All the constants in that reference have to be used to calculate densities or pressures with the MBWR equation of state, and the temperatures in this equation are expressed in terms of the earlier IPTS-68 scale. The most reliable experimental viscosity data were measured mainly on the same temperature scale, so that density and viscosity could be evaluated according to the IPTS-68 scale in the course of the development of the viscosity representation. However, the results of this paper are, of course, reported due to the ITS-90 temperature scale. Therefore, it could be necessary to convert temperatures between both temperature scales.

### 3. Experimental Viscosity Data

The dynamic viscosity of propane in the gaseous and liquid states has been studied in about 40 papers. In principle, analyses of the data for the dilute gas<sup>11</sup> as well as for the high density region<sup>10</sup> have already been performed to select primary data sets. As a consequence of the examination of the viscosity surface of *n*-butane to be published shortly, however, we felt compelled to realize several essential changes in the selection of the primary data. Therefore, a recapitulation pointing out the changes is desirable. We will be guided by details of the low-density and high-density regions. Data at densities up to 0.10 mol L<sup>-1</sup> are now considered to be low-density data, whereas 0.15 mol L<sup>-1</sup> was used as limiting value in the previous papers.

To qualify for the primary class the data should be measured with a high-precision instrument, for which a full working equation is available and all necessary corrections can be applied. According to these requirements only data obtained from methods using either an oscillating disk or a capillary flow apparatus may be classified as primary. The theory and mechanics of operation of both kinds of instruments have been studied in detail and are well established.<sup>17,18</sup> Nevertheless, in practice all of the capillary results in the low-density region and some in the regions of higher densities had to be excluded from the primary data sets, mostly because the analyses of the experiments were performed with simplified working equations. On the other hand, data obtained using rotating and falling cylinders as well as oscillating quartz-crystal viscometers had to be included in order to cover as much as possible of the phase space. This inclusion of data at higher densities, which have not met the stringent ideal measurement conditions and have led to a relaxation of the constraints for primary data, has been taken into account by a decrease of the weights assigned according to the ascribed relative uncertainty  $\delta\eta_r$ . Generally,  $\delta\eta_r$  of the data at elevated densities is about five times larger than that of the data in the low-density region.

The relative uncertainty was assigned based on the measurement method, the quoted experimental errors, the check

on precision of the data and the discrepancies to other independent data in overlapping thermodynamic ranges. Then, the statistical weights  $w_k$  follow from

$$w_k = \left( \frac{100}{\eta_{\text{exp},k} \cdot \delta\eta_{r,k}} \right)^2, \quad (1)$$

where  $\eta_{\text{exp},k}$  is the experimental viscosity value  $k$  and  $\delta\eta_{r,k}$  is its ascribed relative uncertainty.

The established primary data sets in both density regions have been employed in the development of the representative equations for the viscosity of propane. The secondary data have been used for comparison only.

#### 3.1. Low-Density Viscosity Data

The primary and secondary data sets are summarized for the dilute gas in Table 1 together with the year of publication, the method of measurement, the temperature range covered, the number of experimental points with a density  $\rho < 0.10$  mol L<sup>-1</sup>, and the ascribed relative uncertainty of the data. In some cases higher uncertainties have been given to data at higher temperatures.

The most important change in the primary data between our work and the paper by Vogel<sup>11</sup> is that all data above room temperature by Kestin and collaborators<sup>20-22</sup> have now been excluded, because of a temperature measurement error which affected all data measured by Kestin and co-workers with their high-temperature oscillating-disk viscometer.<sup>37</sup> The effect of this error on the measured viscosities has already been demonstrated earlier by Bich *et al.*<sup>38,39</sup> Assuming the concept by Kestin, Ro, and Wakeham<sup>40-43</sup> concerning the extended theorem of corresponding states for the rare gases (KRW), Bich *et al.* developed an improved version of this principle. It was based on quasi-experimental values for the viscosity and diffusion coefficients calculated with the best Hartree-Fock dispersion (HFD) potentials of the noble gases known at that time and resulting from a multiproperty fit. The deviations of the experimental viscosity data of Kestin *et al.*<sup>44</sup> from the HFD-based corresponding states values have been presented in Figs. 3-7 of the paper by Bich *et al.*<sup>39</sup> Kestin *et al.*<sup>44</sup> claimed for their measurements an uncertainty of  $\pm 0.1\%$  at room temperature and  $\pm 0.2\%$  up to  $\pm 0.3\%$  at higher temperatures (up to almost 1000 K). The figures by Bich *et al.* demonstrate that the experimental data by Kestin *et al.* agree with the HFD values at room temperature within this uncertainty. But in the temperature range 350-650 K large systematic differences are evident; they show maxima at about 400 K between 0.5% for neon and 1.2% for xenon and decrease at higher temperatures. Furthermore, the same figures illustrate that the experimental viscosity data by Vogel<sup>45</sup> agree with the HFD corresponding states values within their experimental uncertainties of  $\pm 0.15\%$  at room temperature and of  $\pm 0.2\%$  -  $\pm 0.3\%$  at higher temperatures up to 650 K. On the other hand, a comparison of the experimental data by Kestin *et al.*<sup>44</sup> with the KRW corresponding states values has been illustrated in Figs. D2, D8, D16, D24, and D32 by Kestin *et al.*<sup>43</sup> Remarkably, the data at room temperature character

TABLE 1. List of available data from measurements of the viscosity of propane at low densities  $\rho < 0.10 \text{ mol L}^{-1}$ 

Author(s)	Year	Technique <sup>a</sup>	Temperature (K)	Number of points	Ascr. rel. uncertainty (%)
Primary data sets:					
Kestin <i>et al.</i> <sup>19</sup>	1971	OD	296–303	2	0.2
Kestin <i>et al.</i> <sup>20</sup>	1977	OD	299	1	0.2
Abe <i>et al.</i> <sup>21</sup>	1978	OD	298	1	0.2
Abe <i>et al.</i> <sup>22</sup>	1979	OD	298	1	0.2
Vogel <sup>11</sup>	1995	OD	297–625	14	0.2–0.4
Secondary data sets:					
Klemenc and Remi <sup>23</sup>	1923	C	273	1	2.0
Titani <sup>24</sup>	1930	C	293–393	6	2.0
Trautz and Kurz <sup>25</sup>	1931	C	301–549	6	1.0–2.2
Trautz and Sorg <sup>26</sup>	1931	C	291–524	6	1.0–2.2
Adzumi <sup>27</sup>	1937	C	303–373	8	3.0
Sage and Lacey <sup>28</sup>	1938	RB	311–378	5	5.0
Wobser and Mueller <sup>29</sup>	1941	RB	293–371	5	1.0
Senftleben and Gladisch <sup>30</sup>	1949	HT	313	1	5.0
Golubev and Petrov <sup>31</sup>	1953	C	298–523	8	1.0–2.2
Lambert <i>et al.</i> <sup>32</sup>	1955	OP,C	308–364	7	3.0
Carmichael <i>et al.</i> <sup>33</sup>	1964	RC	278–478	5	1.5
Diaz Pena and Cheda <sup>34</sup>	1975	C	304–408	11	3.0
Diaz Pena and Cheda <sup>35</sup>	1975	C	373	1	3.0
Kestin <i>et al.</i> <sup>20</sup>	1977	OD	335–478	4	0.7
Abe <i>et al.</i> <sup>21</sup>	1978	OD	333–468	4	0.7
Abe <i>et al.</i> <sup>22</sup>	1979	OD	333–468	5	0.7
Nagaoka <i>et al.</i> <sup>36</sup>	1986	RB	298–348	3	2.0

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

ized by the lowest uncertainties show differences from the KRW values up to  $\pm 0.5\%$ . The somewhat better agreement at higher temperatures between the experimental data by Kestin *et al.*<sup>44</sup> and the KRW values<sup>43</sup> arises because only these data were fitted to when the corresponding states principle was applied. However, a simultaneous deterioration of the description of the room temperature data results obviously. From all these findings, we concluded that the experimental results by Kestin *et al.*<sup>44</sup> deviate from the extremely reliable HFD corresponding states values<sup>39</sup> in a systematic manner associated with the thermal conductivity of the gas concerned.

Consequently, the deviations should be caused by the construction of the thermostat and of the temperature measurement device applied in the high-temperature oscillating-disk viscometer used in Kestin's experiments. In that thermostat a relatively strong temperature gradient was kept to achieve a stable stratification of the gas inside the viscometer. According to Figs. 5 and 9 in the paper by Di Pippo *et al.*<sup>37</sup> the temperature measurement was performed by means of three platinum/platinum-rhodium (90Pt, 10Rh) thermocouples spaced radially in the measuring gap of 2.83 mm and led out along the temperature gradient through the bottom of the viscometer, where the mirror of the viscometer was kept at about ambient temperature. The vertical temperature difference in the measurement gap amounted to 0.06–0.15 K, corresponding to a temperature gradient of 20–50 K/m. A large

problem may have resulted from the arrangement of the thermocouple leads along the temperature gradient, which was still increasing below the oscillating-disk system. In the discussion of a possible temperature measurement error, we assumed that thermocouples with conventional asymmetric hot junctions were employed. The heat loss at the measurement position due to heat conduction of the wires in the direction of the negative temperature gradient cannot completely be compensated by heat conduction of the gas and by radiation. For thermocouple leads of 0.4 mm diameter commonly used by Kestin and co-workers, a speculative estimation of the temperature measurement error has been performed.<sup>46</sup> In that procedure radiation has not been taken into account and the heat-transfer coefficient could only be assessed approximately. Nevertheless, the temperatures in the measuring gap should have been measured too low by 1–5 K depending on the measuring temperature itself and above all on the thermal conductivity of the gas. If the deviations were completely due to a temperature measurement error, the temperatures would be too low by another 2 K. This makes it clear that our estimation cannot exactly explain the differences between the experimental data by Kestin and co-workers and the HFD corresponding states values as should be expected. But the estimation gives an idea of the reason for the deviations of the experimental data by Kestin *et al.*<sup>44</sup> from the HFD corresponding state values and from the experimental data determined by Vogel.<sup>11,45</sup> The most convincing fact for

TABLE 2. List of available data from viscosity measurements on propane at densities  $\rho > 0.10 \text{ mol L}^{-1}$ 

Author(s)	Year	Technique <sup>a</sup>	Temperature (K)	Pressure (MPa)	Phase <sup>b</sup>	Number of points	Ascr. rel. uncertainty (%)
Primary data sets:							
Eakin and Ellington <sup>47</sup>	1959	C	298	0.7–62.1	V,L	<sup>c</sup>	...
Starling <i>et al.</i> <sup>48</sup>	1959	C	298–411	0.7–62.1	V,L,S	152 <sup>d,e</sup>	1.5–2.0
Starling <i>et al.</i> <sup>49</sup>	1960	C	298–411	0.7–55.1	V,L,S	<sup>f</sup>	...
Starling <sup>50</sup>	1960	C	363–374	3.4–5.1	V,L,S	79 <sup>g</sup>	1.5–2.0
Starling <i>et al.</i> <sup>51</sup>	1962	C	363–374	3.4–5.1	V,L,S	<sup>h</sup>	...
Carmichael <i>et al.</i> <sup>33</sup>	1964	RC	278–478	0.34–34.4	V,L,S	17 <sup>e</sup>	1.5
Giddings <i>et al.</i> <sup>52</sup>	1966	C	278–378	0.7–55.2	V,L,S	74	1.5
Huang <i>et al.</i> <sup>53</sup>	1966	FC	173–273	6.9–34.5	L	30	2.0
Strumpf <i>et al.</i> <sup>54</sup>	1974	OQ	311	3.2–7.2	L	5	2.0
Diller <sup>5,55</sup>	1982	OQ	90–300	1.7–31.5	L,sL	84	2.0
Secondary data sets:							
Sage and Lacey <sup>28</sup>	1938	RB	311–378	0.34–13.8	V,L,S,sL	86	8.0
van Wijk <i>et al.</i> <sup>56</sup>	1940	FN	304–422	16.1–145	L	20	4.0
Gerf and Galkov <sup>57</sup>	1940	C	83–170	...	sL	14	6.0
Galkov and Gerf <sup>58</sup>	1941	C	112–176	...	sL	5	4.0
Lipkin <i>et al.</i> <sup>59</sup>	1942	C	200–289	...	sL	9	6.0
Smith and Brown <sup>60</sup>	1943	RB	295–463	0.7–34.5	V,L,S	111	8.0
Bicher and Katz <sup>61</sup>	1943	RB	298–498	2.8–34.5	L,S	40	8.0
Comings <i>et al.</i> <sup>62</sup>	1944	C	303–378	0.44–4.2	V,S	29 <sup>e</sup>	3.0
Golubev and Petrov <sup>31</sup>	1953	C	298–523	2.0–81.1	L,S	123	3.0
Baron <i>et al.</i> <sup>63</sup>	1959	C	325–408	0.7–55.2	V,L,S	40	3.0
Swift <i>et al.</i> <sup>64</sup>	1959	FC	88–363	0.34–3.9	L	29	5.0
Swift <i>et al.</i> <sup>65</sup>	1960	FC	243–370	0.34–4.3	L	14	2.5
Babb and Scott <sup>66</sup>	1964	RB	303	200–1000	L	5	8.0
Nasarenko and Golubev <sup>67</sup>	1976	C	284–423	9.8–448	L,S	43 <sup>e</sup>	6.0

<sup>a</sup>(C) capillary, (FC) falling cylinder, (FN) falling needle, (OQ) oscillating quartz crystal, (RB) rolling ball, (RC) rotating cylinder.

<sup>b</sup>(V) vapor, (L) liquid, (S) supercritical, (sL) saturated liquid.

<sup>c</sup>24 experimental points included by Starling *et al.* (Ref. 48).

<sup>d</sup>One value at 370 K, one value at 377 K and three values at 411 K → secondary data.

<sup>e</sup>Individual experimental points at the same or nearly the same pressure reduced to averaged values.

<sup>f</sup>Recommended values on the basis of the experimental data reported by Starling *et al.* (Ref. 48).

<sup>g</sup>11 values at 370.0 K → secondary data.

<sup>h</sup>Only graphical illustration of the experimental points by Starling (Ref. 50).

the temperature measurement error is the systematic order in the deviations for the different noble gases connected with their thermal conductivity coefficients.

Consequently, Vogel's assessment<sup>11</sup> of the propane data by Kestin and collaborators<sup>20–22</sup> as primary is no longer justified because these data should be affected in the same manner by that measurement error. It is estimated that a systematic error between 0.5% and 1% resulted from it.

In addition, the data by Wobser and Mueller<sup>29</sup> were excluded, since the rolling-ball viscometer is not a primary instrument.

### 3.2. Viscosity Data at Higher Densities

The data sets for the higher density region are listed in Table 2, which includes the year of publication, the measurement technique, the temperature and pressure ranges and the fluid phases, the number of experimental points, and the relative uncertainty of the data. The primary and secondary viscosity data at higher densities are also illustrated in the temperature–pressure diagrams of Figs. 1 and 2.

All data by Comings *et al.*<sup>62</sup> and by Swift *et al.*<sup>65</sup> are now classified as secondary data sets, because sufficient primary data are available in the same thermodynamic ranges. The data by Starling *et al.*<sup>48</sup> are considered as primary except for five points. In addition, 11 data points by Starling<sup>50</sup> close to the critical point are excluded from the primary data as explained in Section 4.1.

According to Fig. 1 the liquid region below 270 K and up to 1 MPa is not adequately covered with primary data. The secondary data in that region by Swift *et al.*<sup>64</sup> are characterized by very large uncertainties at low temperatures according to a subsequent paper by Swift *et al.*<sup>65</sup> (see Fig. 2). The supercritical region above 410 K is sparsely covered with primary data, although further experimental data were reported for this region as shown in Fig. 2. The data by Golubev and Petrov<sup>31</sup> determined with a capillary flow viscometer could not be classified as primary, as they show distinct deviations from primary data in overlapping ranges. The shortage of primary data has to be overcome by more accurate and extensive measurements, particularly in the supercritical region.

In all, 425 of 1009 experimental data points represent the

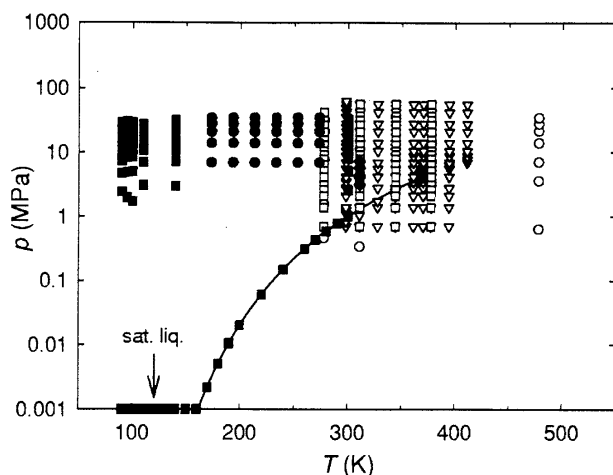


FIG. 1. Distribution of the available experimental viscosity data of propane at densities  $\rho > 0.10 \text{ mol L}^{-1}$ ; primary data. ( $\nabla$ ) Starling *et al.* (1959) (Ref. 48), ( $\Delta$ ) Starling (1960) (Ref. 50), ( $\circ$ ) Carmichael *et al.* (1964) (Ref. 33), ( $\square$ ) Giddings *et al.* (1966) (Ref. 57), ( $\bullet$ ) Huang *et al.* (1966) (Ref. 53), ( $\blacklozenge$ ) Strumpf *et al.* (1974) (Ref. 54), ( $\blacksquare$ ) Diller (1982) (Ref. 5), (—) liquid-vapor boundary.

primary data base for the development of the representation of the viscosity surface at higher densities.

#### 4. Methodology

The correlation of the viscosity is based on the residual viscosity concept. For theoretical and practical reasons, the viscosity  $\eta(\rho, T)$  of a pure fluid at density  $\rho$  and temperature  $T$  may be considered to be composed of three or four contributions:<sup>14-16,68</sup>

$$\eta(\rho, T) = \eta^{(0)}(T) + \Delta\eta(\rho, T) + \Delta\eta_c(\rho, T), \quad (2)$$

$$\eta(\rho, T) = \eta^{(0)}(T) + \eta^{(1)}(T)\rho + \Delta\eta_h(\rho, T) + \Delta\eta_c(\rho, T). \quad (3)$$

Here,  $\eta^{(0)}(T)$  is the viscosity in the zero-density limit,  $\Delta\eta(\rho, T)$  is a residual viscosity that accounts for the increase in viscosity at elevated density over  $\eta^{(0)}$  at the same temperature, and  $\Delta\eta_c(\rho, T)$  is a critical enhancement which represents the viscosity increase in the immediate vicinity of the vapor-liquid critical point. The first two terms of Eq. (2) may be summarized, and the resulting quantity is called background viscosity. With the development of the Rainwater-Friend theory for the transport properties of moderately dense gases,<sup>69-71</sup> it became possible to establish the linear-in-density term  $\eta^{(1)}(T)\rho$  separately. The higher-density terms of the residual viscosity are represented by  $\Delta\eta_h(\rho, T)$ . As a consequence, this approach enables an independent treatment of each contribution. The most recent advances of the kinetic theory may be used as a guide in the development of the viscosity correlation and for the assessment of the internal consistency of the experimental data from the literature.

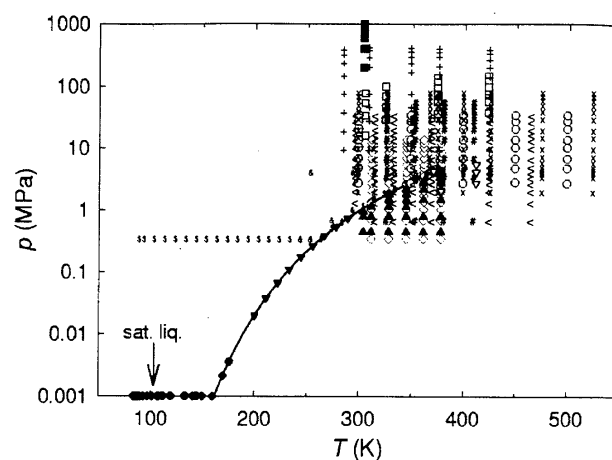


FIG. 2. Distribution of the available experimental viscosity data of propane at densities  $\rho > 0.10 \text{ mol L}^{-1}$ ; secondary data. ( $\diamond$ ) Sage and Lacey (1938) (Ref. 28), ( $\square$ ) van Wijk *et al.* (1940) (Ref. 56), ( $\blacklozenge$ ) Gerf and Galkov (1940) (Ref. 57), ( $\bullet$ ) Galkov and Gerf (1941) (Ref. 58), ( $\blacktriangledown$ ) Lipkin *et al.* (1942) (Ref. 59), ( $\blacktriangleleft$ ) Smith and Brown (1943) (Ref. 60), ( $\circ$ ) Bicher and Katz (1943) (Ref. 61), ( $\blacktriangle$ ) Comings *et al.* (1944) (Ref. 62), ( $\times$ ) Golubev and Petrov (1953) (Ref. 31), ( $\#$ ) Baron *et al.* (1959) (Ref. 63), ( $\$$ ) Swift *et al.* (1959) (Ref. 64), ( $\&$ ) Swift *et al.* (1960) (Ref. 65), ( $\nabla$ ) Starling *et al.* (1959) (Ref. 48), ( $\Delta$ ) Starling (1960) (Ref. 50), ( $\blacksquare$ ) Babb and Scott (1964) (Ref. 66), ( $+$ ) Nasarenko and Golubev (1976) (Ref. 67), (—) liquid-vapor boundary.

#### 4.1. Viscosity in the Critical Region

Viscosity and thermal conductivity of pure fluids diverge at the critical point due to long-range fluctuations. The critical enhancements can be described by a theoretical crossover model originally developed by Olchowy and Sengers<sup>72,73</sup> and recently modified by Luettmmer-Strathmann *et al.*<sup>74,75</sup> Unlike the critical enhancement in thermal conductivity, the enhancement in viscosity is small and becomes relevant only at temperatures and densities close to the critical point. For some fluids,<sup>14,15</sup> the ratio  $\Delta\eta_c(\rho, T)/\eta(\rho, T)$  exceeds 0.01 only within 1% of the critical temperature of the fluid. Experimental data for the viscosity of propane in the region around the critical point ( $\rho_c = 5.000 \text{ mol L}^{-1}$ ,  $T_c = 369.825 \text{ K}$ ,  $p_c = 4.24766 \text{ MPa}$ ) were determined by Starling *et al.*<sup>48</sup> and by Starling.<sup>50</sup> To test for the critical enhancement, a preliminary surface correlation without  $\Delta\eta_c$  in Eqs. (2) and (3) was performed excluding the data by Starling *et al.*<sup>48</sup> for the 370.1 K isotherm and all the data by Starling.<sup>50</sup> Their deviations from the preliminary correlation are illustrated in Fig. 3. In principle, these deviations might be identified as critical enhancement. Unfortunately, the number of points with densities between 4 and 6  $\text{mol L}^{-1}$  is small. Furthermore, the deviations are not positive with a systematic increase when approaching the critical point. On the contrary, there is a large decrease by more than 20% in the experimental data for the 370.0 K isotherm of Starling below the correlated values. In addition, the scattering of the deviations for neighboring points on the isotherms, somewhat more distant from the critical isotherm, is larger than 1%, so a systematic effect of this order of magnitude cannot

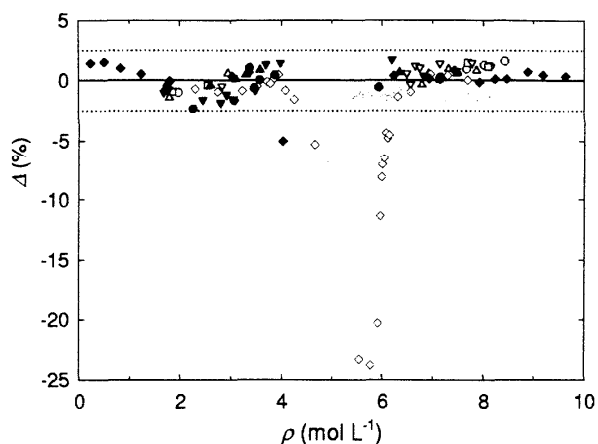


FIG. 3. Deviations ( $\Delta$ ) of the viscosity data by Starling *et al.* (1959) (Ref. 48) and by Starling (1960) (Ref. 50) from a preliminary correlation. Starling *et al.* (Ref. 48): ( $\blacklozenge$ ) 370.1 K; Starling (Ref. 50): ( $\circ$ ) 362.6 K, ( $\square$ ) 366.2 K, ( $\triangle$ ) 368.5 K, ( $\nabla$ ) 369.2 K, ( $\diamond$ ) 370.0 K, ( $\bullet$ ) 370.7 K, ( $\blacktriangle$ ) 371.5 K, ( $\blacktriangledown$ ) 373.7 K.  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$ .

be determined. We conclude that it is not possible to reconcile the experimental data with any expected critical enhancement. The strong decrease of the viscosity data by Starling near the critical point could be caused by experimental difficulties. Thus, 11 data of the 370.0 K isotherm by Starling<sup>50</sup> and one datum of the 370.1 K isotherm by Starling *et al.*<sup>48</sup> were excluded because of negative deviations  $>4\%$  from the preliminary correlation, whereas all other data were accepted for the primary data sets to begin with.

As a consequence of this analysis, we preferred to develop a surface correlation without considering the critical enhancement of the viscosity. There is a strong demand for highly accurate measurements in the critical region.

#### 4.2. Viscosity in the Zero-Density Limit

The viscosity  $\eta^{(0)}(T)$  of a fluid in the zero-density limit is not an experimentally accessible quantity. It is important to distinguish the viscosity in the true limit of zero density from values at low pressures, such as 0.1 MPa. Thus, it is necessary to extract values from the available experimental data in the low-density region. The best way to deduce zero-density values is to use experimental data measured along isotherms and to fit polynomials in density to them. But this extrapolation of isothermal values to the limit of zero density requires enough low-density points of low uncertainty. For propane the experimental data by Vogel<sup>11</sup> seem to be the only values suitable for such an exact evaluation, whereas those by Sage and Lacey<sup>28</sup> and even those by Carmichael *et al.*<sup>33</sup> are characterized by larger uncertainties and a limited number of points. In spite of the large uncertainties, these data and those from all other sources were corrected to the limit of zero density. For this purpose the initial density dependence of viscosity  $\eta^{(1)}(T)\rho$  was subtracted from the experimental  $\eta(\rho, T)$  values at densities  $\rho < 0.10 \text{ mol L}^{-1}$  according to the results of Sec. 4.3. The correction was

performed, although the effect of the initial density dependence is mostly small compared with the experimental uncertainties. But with regard to the intended surface correlation in large ranges of thermodynamic states, the effect was taken into account because it is systematic and  $\eta^{(1)}(T)$  amounts to large negative values at low temperatures instead of positive values at higher temperatures. As already mentioned all data considered are summarized in Table 1.

The zero-density viscosity  $\eta^{(0)}(T)$  can be analyzed independently of the other contributions in Eqs. (2) and (3). Since it is almost independent of the existence of the internal degrees of freedom and unaffected by inelastic collisions, the kinetic theory of dilute monatomic gases by Chapman and Enskog<sup>76</sup> could possibly be used in the analysis. But this theory cannot be applied for a highly accurate prediction of the viscosity in the limit of zero density because of the incomplete knowledge of the intermolecular potential energy surface of polyatomic molecules like propane. The development of the kinetic theory of polyatomic gases by Mc Court *et al.*<sup>77,78</sup> has only been employed for the practical evaluation of the transport properties of diatomic molecules like nitrogen.<sup>79</sup> Apart from polarization correction,<sup>80</sup> the viscosity of a pure polyatomic gas may be related to an effective collision cross section, which contains all the dynamic and statistical information about the binary collision. For practical purposes, this relationship is formally identical to that of monatomic gases and can be written as:

$$\eta^{(0)}(T) = \frac{0.021357(MT)^{1/2}}{\sigma^2 \mathfrak{S}_\eta^*(T^*)}, \quad (4)$$

where  $\mathfrak{S}_\eta^* = \mathfrak{S}(2000)/\pi\sigma^2 f_\eta$  is a reduced effective cross section,  $M$  is the molar mass in  $\text{g mol}^{-1}$ ,  $\sigma$  is a length scaling parameter in nm,  $f_\eta$  is a dimensionless higher-order correction factor according to Chapman and Cowling,  $\eta^{(0)}$  is in units of  $\mu \text{ Pa s}$ , and the temperature  $T$  is in K. The effective cross section is usually expressed in the functional form

$$\ln \mathfrak{S}_\eta^*(T^*) = \sum_{i=0}^4 a_i (\ln T^*)^i, \quad (5)$$

$$T^* = k_B T / \varepsilon, \quad (6)$$

where  $T^*$  is the reduced temperature and  $\varepsilon/k_B$  is an energy scaling parameter in K. Experimental  $\mathfrak{S}_\eta(\mathfrak{S}_\eta = \pi\sigma^2 \mathfrak{S}_\eta^*)$  values corresponding to the primary viscosity values in the limit of zero density have been calculated with the help of Eq. (4).

The fit could be carried out in two versions. The first method uses the assumption that the fluid under discussion obeys the extended principle of corresponding states, and the values of the scaling parameters  $\varepsilon/k_B$  and  $\sigma$  are determined using the known coefficients  $a_i$  of the functional  $\mathfrak{S}_\eta^*$  for the noble gases derived by Bich *et al.*<sup>38</sup> This adoption of a universal correlation has been used by Hendl *et al.*<sup>15</sup> in the case of ethane and by Vogel<sup>11</sup> for propane. In principle, propane does not conform to the extended principle of corresponding states for the rare gases because of the complexity of the molecule. The second approach uses an individual correla-

TABLE 3. Coefficients for the representation of the viscosity of propane.

Molar mass	
$M = 44.0956 \text{ g mol}^{-1}$	
Critical constants	
$T_c = 369.825 \text{ K}$	$\rho_c = 5.000 \text{ mol L}^{-1}$
Scaling factors	
$\varepsilon/k_B = 263.88 \text{ K}$	$\sigma = 0.49748 \text{ nm}$
Coefficients $a_i$ for $\mathcal{G}_\eta^*$ according to Eq. (5)	
$a_0 = 0.25104574$	$a_1 = -0.47271238$
$a_2 = 0$	$a_3 = 0.060836515$
$a_4 = 0$	
Coefficients $b_i$ for $B_\eta$ according to Eq. (11)	
$b_0 = -19.572881$	$b_1 = 219.73999$
$b_2 = -1015.3226$	$b_3 = 2471.01251$
$b_4 = -3375.1717$	$b_5 = 2491.6597$
$b_6 = -787.26086$	$b_7 = 14.085455$
$b_8 = -0.34664158$	
Coefficients $e_{ij}$ , $f_k$ and $g_l$ for $\Delta\eta_h$ according to Eqs. (22) and (23)	
$e_{20} = 35.9873030195$	$e_{21} = -180.512188564$
$e_{22} = 87.7124888223$	$e_{30} = -105.773052525$
$e_{31} = 205.319740877$	$e_{32} = -129.210932610$
$e_{40} = 58.9491587759$	$e_{41} = -129.740033100$
$e_{42} = 76.6280419971$	$e_{50} = -9.59407868475$
$e_{51} = 21.0726986598$	$e_{52} = -14.3971968187$
$f_1 = 1616.88405374$	
$g_1 = 2.50053938863$	$g_2 = 0.860516059264$

tion for propane. Then, the scaling factors determined for the universal correlation can be assumed to be fixed, and new coefficients  $a_i$  can be deduced by fitting Eqs. (4)–(6) to the quasi-experimental  $\mathcal{G}_\eta$  values. That procedure has been performed for nitrogen by Vogel *et al.*<sup>81</sup> and by Millat and Vesovic.<sup>82</sup> With regard to the fact that the scaling factors  $\varepsilon/k_B$  and  $\sigma$  are, in principle, arbitrary in such an individual correlation and in order to minimize the number of coefficients and parameters needed for the full surface correlation, we have adopted the same scaling factors, deduced in the treatment of the initial-density viscosity coefficient (see Sec. 4.3), for the representation of the zero-density viscosity values of propane. Fenghour *et al.*<sup>16</sup> applied this procedure to ammonia.

Table 3 contains the scaling factors  $\varepsilon/k_B$  and  $\sigma$  as well as the significant coefficients  $a_i$  for the individual correlation of propane taking into account the statistical weights  $w_k$  according to Eq. (1). The correlation is valid in the temperature range from 293 to 625 K and its uncertainty is estimated to be  $\pm 0.4\%$  according to the discussion in Sec. 5.1.

### 4.3. Initial Density Dependence of Viscosity

The temperature dependence of the linear-in-density coefficient of the viscosity  $\eta^{(1)}(T)$  in Eqs. (2) or (3) is remarkably large at subcritical temperatures and must be taken into account to obtain an accurate representation of the behavior of the viscosity in the vapor phase.  $\eta^{(1)}$  changes sign from positive to negative, when the temperature decreases. Therefore, the viscosity along an isotherm should first decrease in the vapor phase and subsequently increase with increasing density. The occurrence of a minimum depends on the range

until the phase boundary as has been observed for a number of polar fluids such as refrigerants,<sup>83,84</sup> ammonia,<sup>16</sup> and steam.<sup>85</sup> But Bich and Vogel<sup>86</sup> have shown that polar as well as nonpolar substances exhibit the same general behavior of the initial density dependence of viscosity, which can also be expressed by means of the second viscosity virial coefficient  $B_\eta(T)$ :

$$B_\eta(T) = \frac{\eta^{(1)}(T)}{\eta^{(0)}(T)}. \quad (7)$$

$B_\eta(T)$  follows from the slope and the zero-density limit of the experimental isotherms.

Rainwater and Friend<sup>69,70</sup> modeled the moderately dense gas as a mixture of monomers and dimers, which interact according to the Lennard-Jones (12-6) potential. Whereas the two-monomer contribution could be evaluated exactly and the three-monomer contribution approximately, the monomer-dimer contribution had to be derived by means of selected experimental second transport virial coefficients using two potential parameters ratios to relate the effective intermolecular potential for the monomer-dimer (MD) interaction to that of the monomer-monomer (M) interaction by appropriate scaling

$$\delta = \frac{\sigma_{MD}}{\sigma_M}, \quad \theta = \frac{\varepsilon_{MD}}{\varepsilon_M}. \quad (8)$$

Whereas Rainwater and Friend<sup>69,70</sup> recommended for these ratios  $\delta = 1.02$  and  $\theta = 1.15$ , Bich and Vogel<sup>12,71</sup> deduced somewhat improved values including  $B_\eta$  values for polyatomic gases and vapors:  $\delta = 1.04$  and  $\theta = 1.25$ . The representation of the reduced second viscosity virial coefficient  $B_\eta^*$  as a function of the reduced temperature  $T^*$  is based on the theorem of corresponding states and offers the opportunity of confirming the reliability of special experimental data and extending the experimental temperature range. Lennard-Jones (12-6) potential parameters  $\varepsilon/k_B$  and  $\sigma$  derived from the temperature function of the zero-density viscosity coefficient  $\eta^{(0)}(T)$  can be used for reducing the experimental  $B_\eta(T)$  values:

$$B_\eta^*(T^*) = \frac{B_\eta(T)}{N_A \sigma^3}, \quad (9)$$

where  $N_A$  is Avogadro's constant.  $B_\eta^*(T^*)$  values derived from the experimental  $B_\eta(T)$  data by Vogel<sup>11</sup> in this way are plotted as open circles in Fig. 4 together with the theoretical curves by Rainwater and Friend<sup>69,70</sup> and by Bich and Vogel.<sup>12,71</sup>

In accordance with the fact that propane does not obey the corresponding states principle for the monatomics as well as it does the Lennard-Jones (12-6) model, optimized scaling parameters  $\varepsilon/k_B$  and  $\sigma$  have been determined by fitting the representative correlation of the theoretical results by Bich and Vogel<sup>71</sup> to the experimental  $B_\eta(T)$  of Vogel.<sup>11</sup>  $\varepsilon/k_B = 263.88 \text{ K}$  and  $\sigma = 0.49748 \text{ nm}$  are the scaling factors given in Table 3 and already used in the development of the correlation for the viscosity in the limit of zero density in



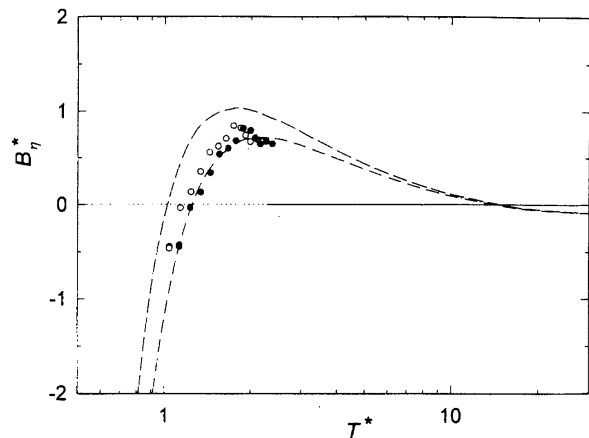


Fig. 4. Reduced second viscosity virial coefficient  $B_{\eta}^*$  as a function of the reduced temperature  $T^*$ . Vogel (1995) (Ref. 11): (○) Lennard-Jones (12-6) parameters from  $\eta^{(0)}(T)$ , (●) optimized parameters; (---) Rainwater-Friend theory ( $\delta=1.02$ ,  $\theta=1.15$ ); (-·-·-) Rainwater-Friend theory, Bich and Vogel ( $\delta=1.04$ ,  $\theta=1.25$ ); (····) Younglove and Ely (1987) (Ref. 4).

order to secure internal consistency. The filled circles in Fig. 4 demonstrate the improved representation of the experimental data.

Values of  $B_{\eta}^*$  were tabulated and correlated by Bich and Vogel<sup>71</sup> in the range  $0.5 \leq T^* \leq 100$  for practical applications by the empirical function

$$B_{\eta}^*(T^*) = \sum_{i=0}^{12} c_i (\sqrt{T^*})^{-i}. \quad (10)$$

This expression is unsatisfactory in its ability to be extrapolated to temperatures  $T^* < 0.5$ . Therefore, Vogel and Küchenmeister<sup>10</sup> used in their preliminary work the modified Enskog theory in the low-temperature range  $T^* < 0.75$  and the Rainwater-Friend theory for temperatures  $T^* > 0.75$  with different coefficients  $c_i$ . In this paper we recommend a new empirical equation in the complete reduced temperature range, which can safely be extrapolated down to  $T^* \approx 0.3$  with the coefficients  $b_i$  listed in Table 3:

$$B_{\eta}^*(T^*) = \sum_{i=0}^6 b_i (T^*)^{-0.25i} + b_7 (T^*)^{-2.5} + b_8 (T^*)^{-5.5}, \quad (11)$$

where  $B_{\eta}$  is in units of  $\text{L mol}^{-1}$  and  $T^* = k_B T / \epsilon$ .

#### 4.4. Higher-Density Terms of the Residual Viscosity

There is very little theoretical guidance for the functional form of  $\Delta \eta_h(\rho, T)$ . One methodological trend consists of representing the higher-density terms of residual viscosity by a power series expansion in the density including coefficients which are dependent on temperature.<sup>15,82</sup> In this procedure a structural optimization, using the SEEQ algorithm based on the stepwise linear least-squares technique,<sup>87</sup> is performed to determine the most suitable combination of terms from a bank of a relatively large number of double polynomials in density and reduced temperature. With respect to the strong

increase of the viscosity between vapor and liquid phases by 3 orders of magnitude at low temperatures in the case of propane, Vogel and Küchenmeister<sup>10</sup> included additional exponential terms in analogy to the MBWR equation of state for the thermodynamic properties:<sup>4</sup>

$$\Delta \eta_h(\rho, T) = \sum_{i=2}^n \sum_{j=0}^m e_{ij} \frac{\rho^i}{T^{*j}} + \sum_{i=5}^n \sum_{j=0}^m f_{ij} \frac{\rho^i}{T^{*j}} \exp(\gamma \rho^2), \quad (12)$$

where  $\gamma = 1/\rho_c^2$  and  $\rho_c$  is the critical density. In their preliminary work, Vogel and Küchenmeister found 11 polynomial and seven exponential terms to be necessary for representing the selected primary data sets within their uncertainties. But the full surface correlation by Vogel and Küchenmeister has been characterized by large loops in the two-phase region resulting in problems when propane is used as a reference fluid for the prediction of the viscosity of other fluids or fluid mixtures by the method of corresponding states. Therefore, the higher-density terms of a new correlation should take into account the dramatic viscosity rise in the liquid phase at low temperatures and show a reasonably smooth path in the two-phase region.

The required behavior seems to be modeled correctly by a free-volume term according to Batschinski<sup>88</sup> and Hildebrand.<sup>89</sup> Batschinski<sup>88</sup> proposed a formulation for the viscosity to be dependent on the difference between the specific volume  $v$  and a certain constant  $w$  similar to the van der Waals constant  $b$ :

$$\eta = \frac{c}{v-w}, \quad (13)$$

where  $c$  is another constant. Batschinski showed that plots of the fluidity  $1/\eta$  are linear functions of  $v$  for nonassociated liquids. Hildebrand<sup>89</sup> modified that relation to recognize that the fluidity depends on the relative expansion  $(V - V_{\infty})/V_{\infty}$ :

$$\frac{1}{\eta} = B \frac{V - V_{\infty}}{V_{\infty}}. \quad (14)$$

$V_{\infty}$  is the intrinsic molar volume, that is, the value when  $1/\eta = 0$ . If both constants of this expression are allowed to vary with temperature<sup>90</sup> and the molar volume  $V$  is replaced by the molar density  $\rho$ , the Batschinski-Hildebrand term may be expressed in reduced variables as

$$\eta_{\text{BH}}(\delta, \tau) = \beta_1(\tau) \frac{\delta}{\delta_0(\tau) - \delta}, \quad (15)$$

$$\delta = \rho/\rho_c, \quad \tau = T/T_c. \quad (16)$$

Here, the reduced close-packed density  $\delta_0(\tau)$  should be temperature dependent to account for the soft repulsive part of the intermolecular pair potential of real fluids. But details of the temperature dependence of the adjustable parameter  $\beta_1(\tau)$  remain to be determined.

A remarkably successful expression which is very similar to the Batschinski-Hildebrand term was introduced by Laesecke<sup>91</sup> and used in wide-ranging viscosity correlations:<sup>92-95</sup>

$$\eta_{AL}(\delta) = \frac{\beta_2}{\delta_0 - \delta} - \frac{\beta_2}{\delta_0} = \frac{\beta_2}{\delta_0} \frac{\delta}{\delta_0 - \delta}. \quad (17)$$

Here,  $\beta_2$  and  $\delta_0$  have been considered to be temperature independent. Apart from the difference in the temperature dependence both formulations are in accordance, identifying  $\beta_1(\tau) \approx \beta_2 / \delta_0$ .

The Batschinski–Hildebrand term has to be related to the residual viscosity in order to obtain correct behavior in the limit of zero density. In addition, if the Batschinski–Hildebrand term represents the higher-density terms of the residual viscosity, another linear term has to be subtracted to take into account that the linear-in-density term of viscosity has been treated separately:

$$\eta_{mBH}(\delta, \tau) = \beta_1(\tau) \left( \frac{\delta}{\delta_0(\tau) - \delta} - \frac{\delta}{\delta_0(\tau)} \right). \quad (18)$$

The temperature dependence of  $\delta_0(\tau)$  was chosen according to

$$\delta_0(\tau) = g_1 \left( 1 + \sum_{l=2}^5 g_l \tau^{(l-1)/2} \right), \quad (19)$$

whereas that of  $\beta_1(\tau)$  was modeled as

$$\beta_1(\tau) = \sum_{k=1}^{np} f_k / \tau^{k-1}. \quad (20)$$

The final correlation for the representation of the higher density terms of the residual viscosity consists of a combination of double polynomials in reduced density and reduced reciprocal temperature and of modified Batschinski–Hildebrand terms with a temperature-dependent reduced close-packed density including Eqs. (16) and (19):

$$\Delta \eta_h(\delta, \tau) = \sum_{i=2}^n \sum_{j=0}^m e_{ij} \frac{\delta^i}{\tau^j} + \sum_{k=1}^{np} \frac{f_k}{\tau^{k-1}} \times \left( \frac{\delta}{\delta_0(\tau) - \delta} - \frac{\delta}{\delta_0(\tau)} \right). \quad (21)$$

The SEEQ algorithm used in conjunction with the parameters  $e_{ij}$  and  $f_{ij}$  of Eq. (12) cannot be employed to deduce the parameters  $e_{ij}$ ,  $f_k$ , and  $g_i$  of Eqs. (19) and (21). Therefore, the NIST package ODRPACK by Boggs *et al.*,<sup>96</sup> based on a nonlinear least-squares regression, was used.

The uncertainties of the experimental data are too large to establish the temperature dependence of  $\delta_0(\tau)$  and  $\beta_1(\tau)$  unambiguously. Thus, we considered only that of the reduced close-packed density  $\delta_0(\tau)$ . ODRPACK also dropped the higher terms in the temperature dependence of  $\delta_0(\tau)$ . Furthermore, the double polynomials had to be restricted to  $n=5$  and  $m=2$ , so the parameters  $e_{ij}$  remained statistically significant. The best correlation of the selected primary data was obtained with the representation

$$\Delta \eta_h(\delta, \tau) = \sum_{i=2}^5 \sum_{j=0}^2 e_{ij} \frac{\delta^i}{\tau^j} - f_1 \left( \frac{\delta}{\delta_0(\tau) - \delta} - \frac{\delta}{\delta_0(\tau)} \right). \quad (22)$$

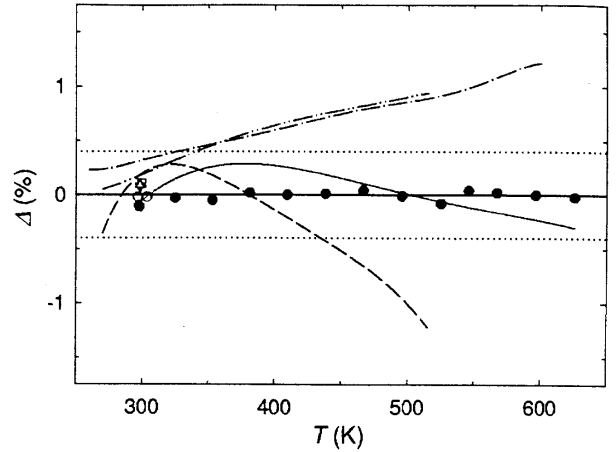


FIG. 5. Deviations ( $\Delta$ ) of the primary viscosity data and of correlations from the present individual zero-density correlation. (○) Kestin *et al.* (1971) (Ref. 19), (□) Kestin *et al.* (1977) (Ref. 20), (▽) Abe *et al.* (1978) (Ref. 21), (Δ) Abe *et al.* (1979) (Ref. 22), (●) Vogel (1995) (Ref. 11), (----) Tanaka and Makita (1976) (Ref. 1), (-·-·-·-) Holland *et al.* (1979) (Ref. 3), (- - - -) Younglove and Ely (1987) (Ref. 4), (—) preliminary universal correlation.  $\Delta = 100(\eta_{exp} - \eta_{cor}) / \eta_{exp}$ .

$$\delta_0(\tau) = g_1(1 + g_2 \tau^{1/2}) \quad (23)$$

of the higher density terms of the residual viscosity. Table 3 lists the significant coefficients which are based on the primary data and the statistical weights as derived from the relative experimental uncertainties of Table 2.

## 5. Results and Discussion

The viscosity of propane, as calculated by the correlation, using the coefficients of Table 3 and Eq. (3) with  $\Delta \eta_c(\rho, T) = 0$ , Eqs. (4)–(6) for  $\eta^{(0)}(T)$ , Eqs. (7), (9), and (11) for  $\eta^{(1)}(T)\rho$  as well as Eqs. (22) and (23) with Eq. (16) for  $\Delta \eta_h(\rho, T)$ , have been compared with primary and secondary experimental data. The performance of the correlation can be assessed by the extent of the agreement with the primary data used in its development. Furthermore, a comparison with the secondary data should reveal any systematic deviations or large scatterings of the experimental data. The comparison has been carried out for the zero-density viscosity coefficient, for the reduced second viscosity virial coefficient as a proof of the initial density dependence of viscosity, and for the total viscosity coefficient  $\eta(\rho, T)$  for the higher density ranges.

### 5.1. Zero-Density Limit and Initial Density Dependence

The deviations of the primary data from the final individual correlation are presented in Fig. 5 together with deviations for some other correlations. The correlations by Holland *et al.*<sup>3</sup> and by Younglove and Ely<sup>4</sup> were primarily based on the data by Kestin and collaborators<sup>20–22</sup> and therefore show large positive deviations with increasing temperature. The increasingly negative deviations of the correlation by Tanaka and Makita<sup>1</sup> result from the data of Trautz and

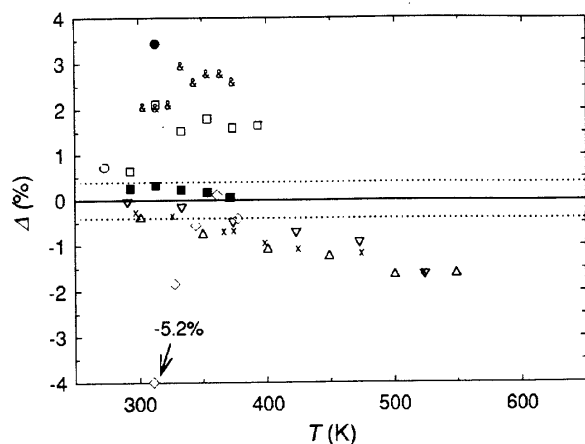


FIG. 6. Deviations ( $\Delta$ ) of older secondary viscosity data from the present individual zero-density correlation. (○) Klemenc and Remi (1923) (Ref. 23), (□) Titani (1930) (Ref. 24), ( $\Delta$ ) Trautz and Kurz (1931) (Ref. 25), ( $\nabla$ ) Trautz and Sorg (1931) (Ref. 26), (&) Adzumi (1937) (Ref. 27), ( $\diamond$ ) Sage and Lacey (1938) (Ref. 28), (■) Wobser and Mueller (1941) (Ref. 29), (●) Senftleben and Gladisch (1949) (Ref. 30), (×) Golubev and Petrov (1953) (Ref. 31).  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}}) / \eta_{\text{exp}}$ .

co-workers<sup>25,26</sup> on which it was based. In addition, the figure shows the differences between the individual correlation and the previous universal correlation based on the same primary data and on the universal functional for the monatomics by Bich *et al.*<sup>38</sup> The deviations amount to nearly  $\pm 0.3\%$  and justify using an individual correlation, whose uncertainty is estimated to be  $\pm 0.4\%$ , because only data from one laboratory could be accepted as primary data in the complete temperature range.

The deviations of the secondary zero-density viscosity data from the present correlation are presented in Figs. 6 and 7. Figure 7 illustrates that the data by Kestin and

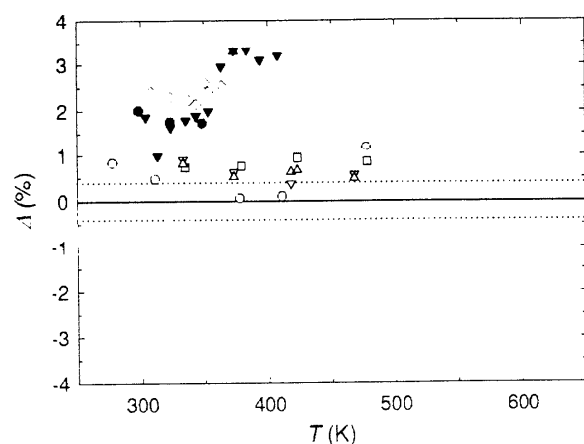


FIG. 7. Deviations ( $\Delta$ ) of more recent secondary viscosity data from the present individual zero-density correlation. (○) Lambert *et al.* (1955) (Ref. 32), (□) Carmichael *et al.* (1964) (Ref. 33), ( $\nabla$ ) Diaz Pena and Cheda (1975) (Ref. 34), ( $\blacktriangle$ ) Diaz Pena and Cheda (1975) (Ref. 35), (□) Kestin *et al.* (1977) (Ref. 20), (□) Abe *et al.* (1978) (Ref. 21), (□) Abe *et al.* (1979) (Ref. 22), (●) Nagaoka *et al.* (1986) (Ref. 36).  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}}) / \eta_{\text{exp}}$ .

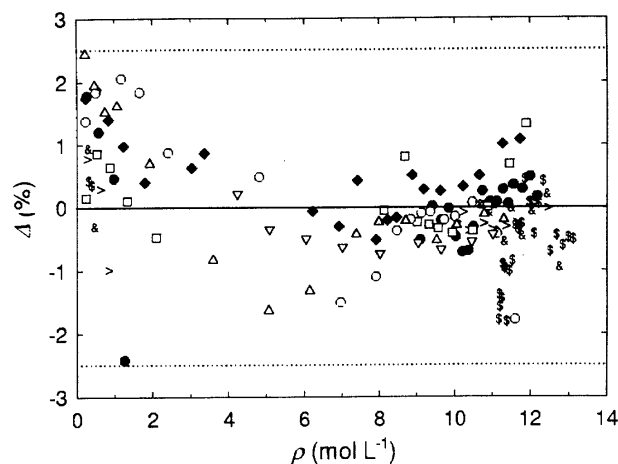


FIG. 8. Deviations ( $\Delta$ ) of the primary viscosity data by Starling *et al.* (1959) (Ref. 48) from the correlation. (S) 298 K, (&) 311 K, (>) 328 K, (●) 344 K, (□) 361 K, (◆) 370 K, (○) 378 K, ( $\Delta$ ) 394 K, ( $\nabla$ ) 411 K.  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}}) / \eta_{\text{exp}}$ .

collaborators<sup>20–22</sup> above room temperature systematically deviate from the individual correlation by about  $+0.7\%$  and that they are characterized by an additional scatter of up to  $\pm 0.3\%$ . These findings demonstrate that the data by Kestin and collaborators cannot be reconciled with the requirements for primary data. The very close agreement of the data by Wobser and Mueller<sup>29</sup> with the individual correlation in Fig. 6 seems to be accidental.

The extrapolation of the individual correlation to low temperatures is problematic, because there are no experimental data to define the correlation in that temperature range, even if a high uncertainty is accepted. But an extrapolation by means of the universal correlation would also be questionable, since a high reliability of the universal functional for the monatomics is limited to  $T^* \approx 1$ . The same is true for any intermolecular potential model, such as the Lennard-Jones model used by Younglove and Ely.<sup>4</sup> Thus, the uncertainty of the values resulting from the individual zero-density correlation of this paper could possibly increase up to  $\pm 20\%$  or more when approaching the triple-point temperature.

The initial density dependence of viscosity, which has been included in this paper, represents an essential improvement over that by Younglove and Ely.<sup>4</sup> In their correlation  $B_{\eta}^*(T^*)$  has been set to zero for all reduced temperatures, as is demonstrated in Fig. 4.

## 5.2. High-Density Region

The deviations of the primary experimental data by Starling *et al.*<sup>48</sup> from the present correlation along nine isotherms between 298 and 411 K are illustrated in Fig. 8. Figure 9 shows the deviations of the primary experimental data by Starling<sup>50</sup> along eight isotherms, which are relatively close to the critical isotherm. The deviations of those experimental data by Starling<sup>50</sup> and by Starling *et al.*<sup>48</sup> which had to be classified as secondary because of experimental problems close to the critical point, are shown in Fig. 10. Some outli-

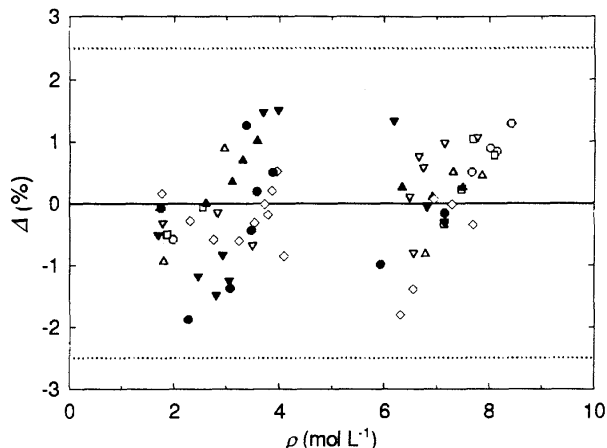


FIG. 9. Deviations ( $\Delta$ ) of the primary viscosity data by Starling (1960) (Ref. 50) from the correlation. ( $\circ$ ) 362.6 K, ( $\square$ ) 366.2 K, ( $\triangle$ ) 368.5 K, ( $\nabla$ ) 369.2 K, ( $\diamond$ ) 370.0 K, ( $\bullet$ ) 370.7 K, ( $\blacktriangle$ ) 371.5 K, ( $\blacktriangledown$ ) 373.7 K.  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$ .

ers from the paper by Starling *et al.*,<sup>48</sup> which have positive deviations of about 3%–5%, are included in this figure. Almost all of the primary data of both papers agree with the correlation within  $\pm 2\%$  in the complete density and temperature ranges of the measurements. In the moderately dense gas or vapor there is possibly a small systematic tendency to larger positive deviations with increasing density, as indicated by Fig. 8.

Figures 11 and 12 display the deviations of the primary experimental viscosity data by Carmichael *et al.*<sup>33</sup> and by Giddings *et al.*,<sup>52</sup> both along four isotherms between 278 and 478 K and between 278 and 378 K, respectively. Practically all these data are reproduced by the correlation within  $\pm 1.5\%$  in the dense gas as well as in the liquid region.

The liquid region is well represented by the new correlation. Figure 13 shows the deviations of the primary experi-

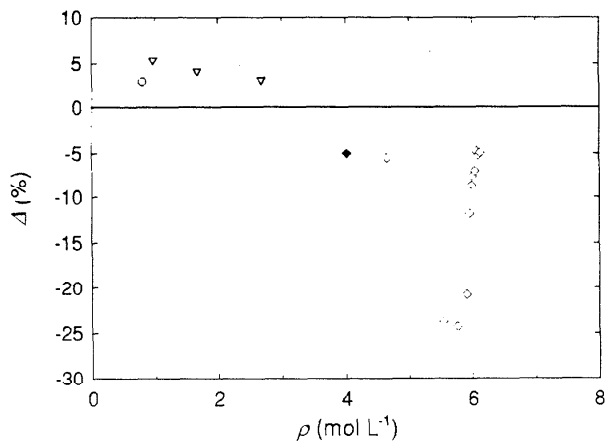


FIG. 10. Deviations ( $\Delta$ ) of the secondary viscosity data by Starling *et al.* (1959) (Ref. 48) and by Starling (1960) (Ref. 50) from the correlation. Starling *et al.* (Ref. 48): ( $\blacklozenge$ ) 370 K, ( $\circ$ ) 378 K, ( $\nabla$ ) 411 K; Starling (Ref. 50): ( $\diamond$ ) 370.0 K.  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$ .

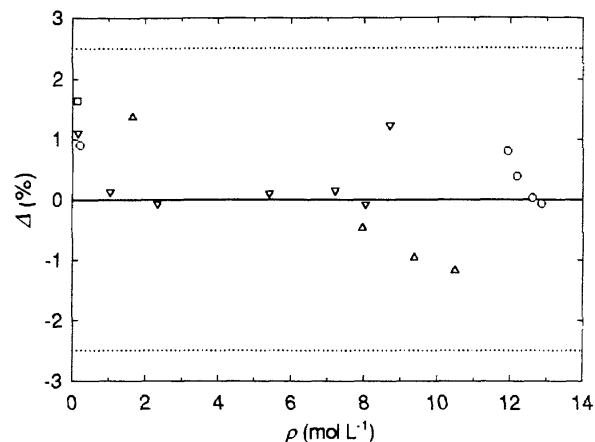


FIG. 11. Deviations ( $\Delta$ ) of the primary viscosity data of Carmichael *et al.* (1964) (Ref. 33) from the present correlation. ( $\circ$ ) 278 K, ( $\square$ ) 311 K, ( $\nabla$ ) 378 K, ( $\triangle$ ) 478 K.  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$ .

mental viscosity data by Huang *et al.*<sup>53</sup> along six isotherms between 173 and 273 K and that by Strumpf *et al.*<sup>54</sup> at 311 K. The deviations from the correlation are within  $\pm 1.5\%$ . The experimental data by Diller<sup>5</sup> for the compressed and saturated liquid are represented by the new correlation within  $\pm 2.5\%$  in the whole temperature range between 90 and 300 K. As indicated in Fig. 14, the deviations increase from  $\pm 1.0\%$  to  $\pm 2.5\%$  with increasing density or better with decreasing temperature of the isotherms.

The deviations of all these primary data sets from the new correlation fall within the ascribed relative uncertainties given in Table 2.

The performance of the representation has been tested using the secondary experimental data of Table 2. The deviations from the correlation of the data by Sage and Lacey,<sup>28</sup> by Smith and Brown,<sup>60</sup> by Bicher and Katz,<sup>61</sup> and by Comings *et al.*<sup>62</sup> are illustrated in Fig. 15. Most of these data agree with the correlation within  $\pm 10\%$  with the exception of that by Sage and Lacey, which show a systematic trend at

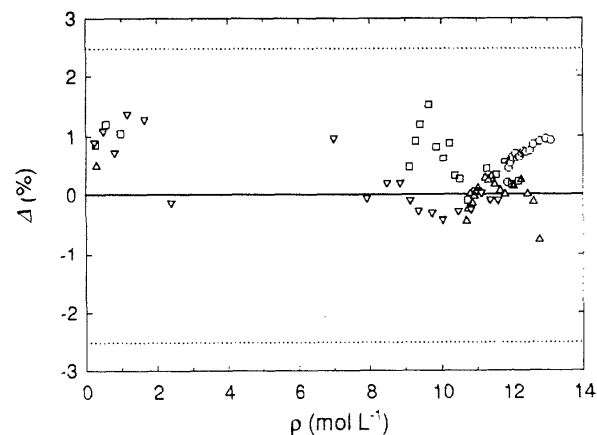


FIG. 12. Deviations ( $\Delta$ ) of the primary viscosity data of Giddings *et al.* (1966) (Ref. 52) from the present correlation. ( $\circ$ ) 278 K, ( $\triangle$ ) 311 K, ( $\square$ ) 344 K, ( $\nabla$ ) 378 K.  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$ .

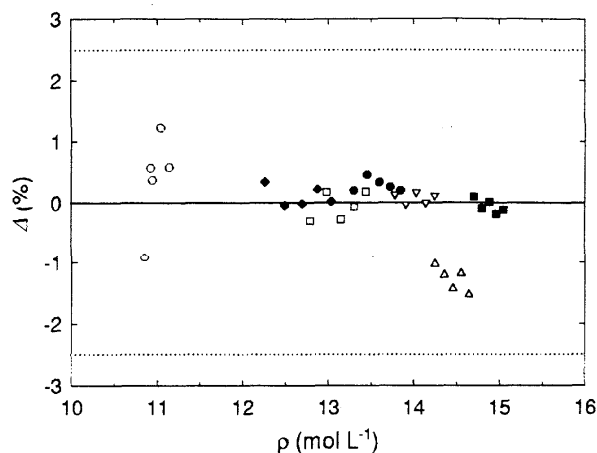


FIG. 13. Deviations ( $\Delta$ ) of the primary viscosity data of Huang *et al.* (1966) (Ref. 53) and of Strumpf *et al.* (1974) (Ref. 54) from the present correlation. Huang *et al.* (Ref. 53): (■) 173 K, ( $\Delta$ ) 193 K, ( $\nabla$ ) 213 K, (●) 233 K, (□) 253 K, (◆) 273 K. Strumpf *et al.* (Ref. 54): (○) 311 K.  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$ .

moderate densities. Figure 16 shows the deviations from the correlation of the data in the liquid region by van Wijk *et al.*,<sup>56</sup> by Gerf and Galkov,<sup>57</sup> by Galkov and Gerf,<sup>58</sup> by Lipkin *et al.*,<sup>59</sup> by Swift *et al.*,<sup>64,65</sup> and by Babb and Scott.<sup>66</sup> The agreement with the correlation is again within  $\pm 10\%$  except for a systematic trend of the data by Swift *et al.*<sup>64</sup> at low temperatures, one outlier by Swift *et al.*<sup>65</sup> in the neighborhood of the critical point, and two outliers by Gerf and Galkov.<sup>57</sup> Remarkably, the data by Babb and Scott<sup>66</sup> deviate by less than 8% from the correlation, although they are outside the pressure range of the MBWR equation of state. As indicated in Fig. 17, the data by Golubev and Petrov,<sup>31</sup> Baron *et al.*,<sup>63</sup> and Nasarenko and Golubev<sup>67</sup> agree with the correlation within  $\pm 5\%$  apart from some data by Golubev and Petrov.<sup>31</sup> Larger deviations of the data by Nasarenko and Golubev<sup>67</sup> at higher densities are due to the fact that the pressures of these experimental points exceed the pressure

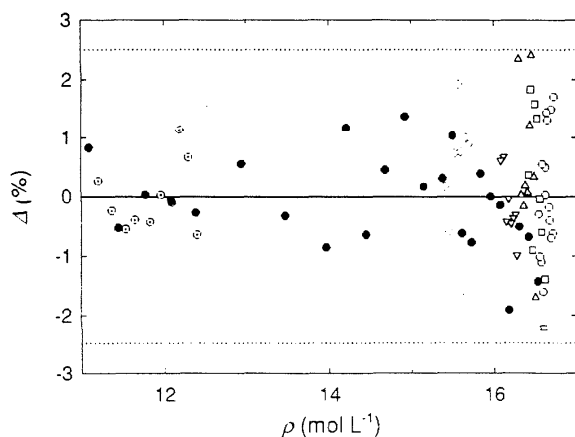


FIG. 14. Deviations ( $\Delta$ ) of the primary viscosity data of Diller (1982) (Ref. 5) from the present correlation. (○) 90 K, (□) 95 K, ( $\Delta$ ) 100 K, ( $\nabla$ ) 110 K, (◇) 110 K, (⊙) 300 K, (●) saturated liquid.  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$ .

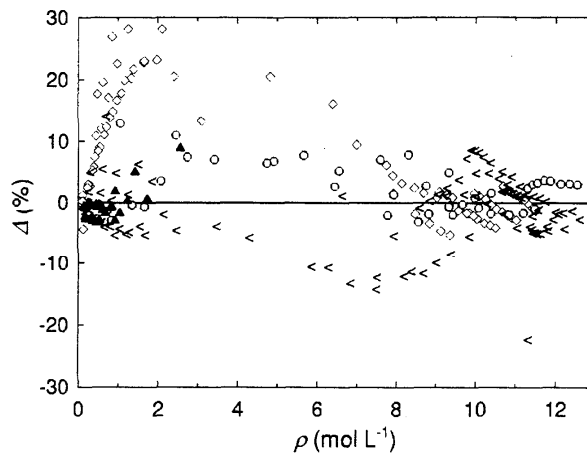


FIG. 15. Deviations ( $\Delta$ ) of secondary viscosity data from the present correlation (Part I). (◇) Sage and Lacey (1938) (Ref. 28), (<) Smith and Brown (1943) (Ref. 60), (○) Bicher and Katz (1943) (Ref. 61), (▲) Comings *et al.* (1944) (Ref. 62).  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$ .

range of the MBWR equation of state. Because the differences are not dramatic, it should be possible to extrapolate moderately beyond the range of validity of the equation of state.

### 5.3. Comparison with the Younglove–Ely Representation

As remarked in the introduction, the most recent propane viscosity correlation by Younglove and Ely<sup>4</sup> was largely undocumented with regard to the data base and the development of the functional form. A comparison between the Younglove–Ely representation and the present correlation is shown along a number of selected isotherms in Fig. 18. The isotherms have specifically been chosen to be representative of the behavior in the vapor phase and at low densities, be-

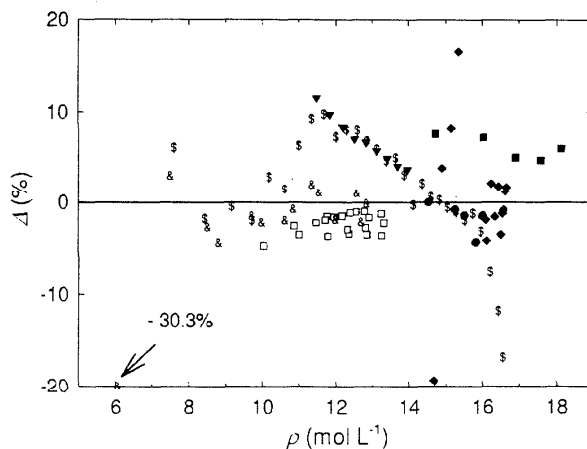


FIG. 16. Deviations ( $\Delta$ ) of secondary viscosity data from the present correlation (Part II). (□) van Wijk *et al.* (1940) (Ref. 56), (◆) Gerf and Galkov (1940) (Ref. 57), (●) Galkov and Gerf (1941) (Ref. 58), (▼) Lipkin *et al.* (1942) (Ref. 59), (S) Swift *et al.* (1959) (Ref. 64), (&) Swift *et al.* (1960) (Ref. 65), (■) Babb and Scott (1961) (Ref. 66).  $\Delta = 100(\eta_{\text{exp}} - \eta_{\text{cor}})/\eta_{\text{exp}}$ .

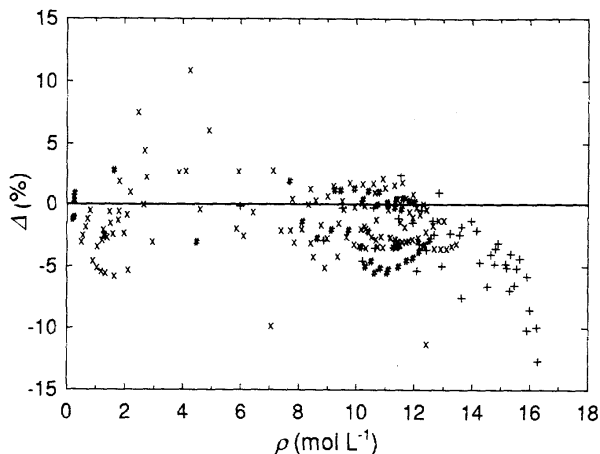


FIG. 17. Deviations ( $\Delta$ ) of secondary viscosity data from the present correlation (Part III). (x) Golubev and Petrov (1953) (Ref. 31), (#) Baron *et al.* (1959) (Ref. 63), (+) Nasarenko and Golubev (1976) (Ref. 67).  $\Delta = 100(\eta_{exp} - \eta_{cor})/\eta_{exp}$ .

cause the inclusion of a new source of low-density data<sup>11</sup> and of a new way of predicting data in the vapor phase in the present work may have a significant effect. Surprisingly, the deviations between the two correlations are within  $\pm 2.5\%$  at zero density in the temperature range from 100 to 200 K, as illustrated by different open symbols. This agreement is accidental, since neither correlation is based on any experimental data in this thermodynamic range. In fact, the uncertainty of an extrapolation to low temperatures corresponds to  $\pm 20\%$ , as already discussed in Sec. 5.1. Systematic deviations between the two correlations are obvious in the vapor phase. Thus, the viscosity at 300 K is overestimated by up to 5% using the Younglove–Ely representation. The reason is that Younglove and Ely did not take into account the initial

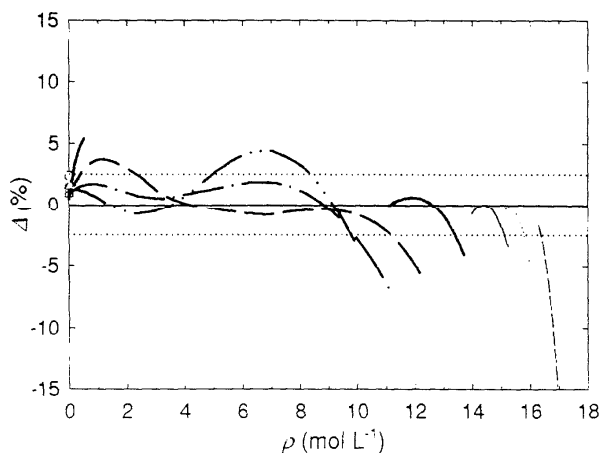


FIG. 18. Comparison with the correlation by Younglove and Ely (1987) (Ref. 4). (· · · · ·) 100 K, (— · — · —) 150 K, (—) 200 K, (— — —) 300 K, (— · — · —) 400 K, (— · — · — · —) 500 K, (— · — · — · — · —) 600 K.  $\Delta = 100(\eta_{Y\&E} - \eta_{res.cor})/\eta_{Y\&E}$ .

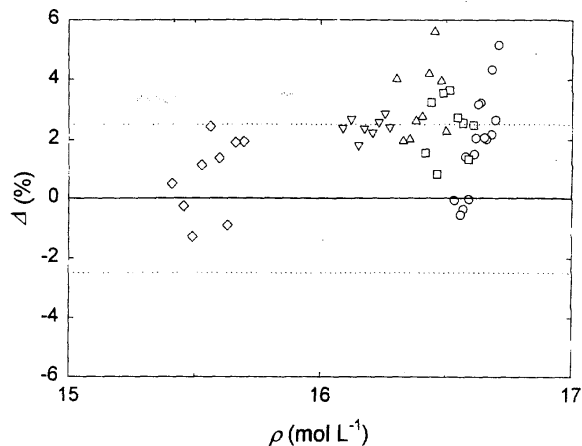


FIG. 19. Deviations ( $\Delta$ ) of the viscosity data of Diller (1982) (Ref. 5) from the Younglove–Ely correlation (Ref. 4). (○) 90 K, (□) 95 K, (Δ) 100 K, (▽) 110 K, (◇) 140 K.  $\Delta = 100(\eta_{Diller} - \eta_{Y\&E})/\eta_{Diller}$ .

density dependence of viscosity. Therefore, we think that in the vapor phase the present representation is an improvement over the previous one.

In the supercritical region and in the liquid phase, both correlations seem to have been based on nearly the same sets of experimental data. Remarkably, the deviations show a systematic trend for each isotherm with increasing density leading to an underestimation of about 5% for the highest density by the use of the Younglove–Ely correlation, for which no uncertainty bounds were reported. For the lowest isotherm of 100 K the deviations reach up to 15%, but at the highest densities there are no experimental data on which the correlations could be based. The differences between both correlations are caused by the choice of functional forms for the high-density region. Their suitability for the representation of the viscosity at high densities could be tested by a comparison with the experimental values by Diller,<sup>5</sup> the only data close to this region. Deviations between the Younglove–Ely correlation and the data by Diller are displayed for the five lowest isotherms in Fig. 19. They reveal that the Younglove–Ely correlation underestimates the viscosity systematically with decreasing temperature and increasing density. Therefore, we conclude that the new representation is also better in the high-density region.

## 6. Tabulations and the Overall Representation

Tabulations of the viscosity and density of propane are given in Tables 4 and 5 over the entire range of the correlation including the saturation line. The tabulations have been generated directly from the representative equations (3)–(7), (9), (11), (16), (22), and (23) including the coefficients of Table 3 as functions of pressure and temperature using the MBWR equation of state.<sup>4</sup> The tables provide useful reference values as well as assistance in the validation of computer codes.

TABLE 4. Viscosity and density of propane

Temperature	90 K		100 K		110 K	
	Pressure (MPa)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )
0.01	16.52	7388.0	16.29	3783.0	16.06	2266.0
0.05	16.52	7392.0	16.29	3785.0	16.06	2267.0
0.10	16.52	7397.0	16.29	3787.0	16.06	2269.0
0.15	16.52	7401.0	16.29	3789.0	16.06	2270.0
0.20	16.52	7406.0	16.29	3791.0	16.06	2271.0
0.25	16.52	7410.0	16.29	3793.0	16.06	2272.0
0.30	16.52	7415.0	16.29	3795.0	16.06	2273.0
0.35	16.52	7419.0	16.29	3797.0	16.06	2274.0
0.40	16.52	7424.0	16.29	3799.0	16.06	2276.0
0.50	16.52	7433.0	16.29	3803.0	16.06	2278.0
0.60	16.52	7442.0	16.29	3808.0	16.06	2280.0
0.80	16.53	7460.0	16.29	3816.0	16.07	2285.0
1.00	16.53	7478.0	16.30	3824.0	16.07	2290.0
1.50	16.53	7524.0	16.30	3845.0	16.07	2301.0
2.00	16.53	7570.0	16.30	3867.0	16.08	2313.0
2.50	16.54	7616.0	16.31	3888.0	16.08	2325.0
3.00	16.54	7663.0	16.31	3909.0	16.08	2337.0
3.50	16.54	7709.0	16.31	3931.0	16.09	2348.0
4.00	16.55	7756.0	16.32	3952.0	16.09	2360.0
5.00	16.56	7851.0	16.33	3995.0	16.10	2384.0
6.00	16.56	7947.0	16.33	4039.0	16.11	2409.0
8.00	16.58	8143.0	16.35	4128.0	16.13	2457.0
10.00	16.59	8342.0	16.36	4218.0	16.14	2507.0
15.00	16.62	8862.0	16.40	4450.0	16.18	2634.0
20.00	16.66	9411.0	16.44	4692.0	16.22	2766.0
25.00	16.69	9993.0	16.47	4945.0	16.26	2903.0
30.00	16.72	10610.0	16.51	5208.0	16.30	3044.0
35.00	16.76	11260.0	16.54	5483.0	16.33	3190.0
40.00	16.79	11950.0	16.58	5769.0	16.37	3342.0
50.00	...	...	16.64	6379.0	16.44	3660.0
60.00	...	...	16.71	7040.0	16.51	4000.0
80.00	...	...	16.83	8535.0	16.64	4749.0
100.00	...	...	16.94	10300.0	16.75	5599.0

Temperature	120 K		130 K		140 K	
	Pressure (MPa)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )
0.01	15.83	1510.0	15.60	1087.0	15.37	827.5
0.05	15.83	1511.0	15.60	1087.0	15.37	827.8
0.10	15.83	1512.0	15.60	1088.0	15.38	828.2
0.15	15.83	1512.0	15.60	1088.0	15.38	828.5
0.20	15.83	1513.0	15.60	1089.0	15.38	828.9
0.25	15.83	1514.0	15.61	1089.0	15.38	829.2
0.30	15.83	1515.0	15.61	1090.0	15.38	829.6
0.35	15.83	1515.0	15.61	1090.0	15.38	830.0
0.40	15.83	1516.0	15.61	1091.0	15.38	830.3
0.50	15.84	1517.0	15.61	1092.0	15.38	831.0
0.60	15.84	1519.0	15.61	1093.0	15.38	831.8
0.80	15.84	1522.0	15.61	1095.0	15.38	833.2
1.00	15.84	1525.0	15.61	1097.0	15.39	834.7
1.50	15.84	1532.0	15.62	1102.0	15.39	838.3
2.00	15.85	1539.0	15.62	1107.0	15.40	841.9
2.50	15.85	1547.0	15.63	1112.0	15.40	845.5
3.00	15.86	1554.0	15.63	1117.0	15.41	849.2
3.50	15.86	1562.0	15.64	1122.0	15.41	852.9
4.00	15.87	1569.0	15.64	1127.0	15.42	856.5
5.00	15.88	1584.0	15.65	1137.0	15.43	863.9
6.00	15.89	1599.0	15.66	1147.0	15.44	871.3
8.00	15.90	1630.0	15.68	1168.0	15.46	886.3
10.00	15.92	1660.0	15.70	1189.0	15.48	901.4
15.00	15.96	1739.0	15.75	1242.0	15.53	939.8

TABLE 4. Viscosity and density of propane—Continued

Temperature	120 K		130 K		140 K	
Pressure (MPa)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)
20.00	16.01	1821.0	15.79	1297.0	15.58	979.3
25.00	16.05	1905.0	15.84	1353.0	15.63	1020.0
30.00	16.09	1991.0	15.88	1411.0	15.67	1061.0
35.00	16.13	2080.0	15.92	1471.0	15.72	1104.0
40.00	16.17	2172.0	15.96	1532.0	15.76	1147.0
50.00	16.24	2364.0	16.04	1659.0	15.85	1238.0
60.00	16.31	2567.0	16.12	1792.0	15.93	1332.0
80.00	16.45	3008.0	16.26	2080.0	16.08	1534.0
100.00	16.57	3498.0	16.40	2396.0	16.22	1753.0
Temperature	150 K		160 K		170 K	
Pressure (MPa)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)
0.01	15.14	656.9	14.91	537.7	14.68	450.3
0.05	15.15	657.1	14.91	537.9	14.68	450.5
0.10	15.15	657.4	14.91	538.1	14.68	450.7
0.15	15.15	657.6	14.92	538.3	14.68	450.9
0.20	15.15	657.9	14.92	538.5	14.68	451.0
0.25	15.15	658.2	14.92	538.8	14.68	451.2
0.30	15.15	658.5	14.92	539.0	14.68	451.4
0.35	15.15	658.7	14.92	539.2	14.69	451.6
0.40	15.15	659.0	14.92	539.4	14.69	451.8
0.50	15.15	659.6	14.92	539.9	14.69	452.1
0.60	15.15	660.1	14.92	540.3	14.69	452.5
0.80	15.15	661.2	14.92	541.2	14.69	453.2
1.00	15.16	662.3	14.93	542.1	14.69	454.0
1.50	15.16	665.1	14.93	544.3	14.70	455.8
2.00	15.17	667.9	14.94	546.5	14.71	457.7
2.50	15.17	670.7	14.95	548.8	14.72	459.5
3.00	15.18	673.5	14.95	551.0	14.72	461.4
3.50	15.19	676.3	14.96	553.2	14.73	463.3
4.00	15.19	679.1	14.96	555.5	14.74	465.1
5.00	15.20	684.7	14.98	560.0	14.75	468.9
6.00	15.21	690.4	14.99	564.5	14.76	472.6
8.00	15.24	701.8	15.01	573.6	14.79	480.2
10.00	15.26	713.3	15.04	582.8	14.82	487.7
15.00	15.31	742.6	15.10	606.1	14.88	507.0
20.00	15.37	772.6	15.16	629.9	14.94	526.5
25.00	15.42	803.2	15.21	654.1	15.00	546.4
30.00	15.47	834.6	15.26	678.9	15.06	566.6
35.00	15.52	866.6	15.32	704.1	15.12	587.2
40.00	15.56	899.4	15.37	729.8	15.17	608.1
50.00	15.66	967.0	15.46	782.8	15.27	651.1
60.00	15.74	1037.0	15.56	837.7	15.37	695.5
80.00	15.90	1187.0	15.73	953.9	15.55	788.9
100.00	16.05	1349.0	15.88	1079.0	15.71	888.6
Temperature	180 K		190 K		200 K	
Pressure (MPa)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)
0.01	14.44	383.7	0.006 377	5.258	0.006 051	5.526
0.05	14.45	383.8	14.21	331.4	13.96	289.1
0.10	14.45	384.0	14.21	331.5	13.97	289.2
0.15	14.45	384.2	14.21	331.7	13.97	289.3
0.20	14.45	384.3	14.21	331.8	13.97	289.4
0.25	14.45	384.5	14.21	332.0	13.97	289.6
0.30	14.45	384.6	14.21	332.1	13.97	289.7
0.35	14.45	384.8	14.21	332.2	13.97	289.8
0.40	14.45	385.0	14.21	332.4	13.97	289.9
0.50	14.45	385.3	14.21	332.7	13.97	290.2



TABLE 4. Viscosity and density of propane—Continued

Temperature	180 K		190 K		200 K	
	Pressure (MPa)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )
0.60	14.45	385.6	14.22	332.9	13.97	290.4
0.80	14.46	386.2	14.22	333.5	13.98	290.9
1.00	14.46	386.9	14.22	334.0	13.98	291.4
1.50	14.47	388.4	14.23	335.4	13.99	292.7
2.00	14.48	390.0	14.24	336.8	14.00	294.0
2.50	14.48	391.6	14.25	338.2	14.01	295.2
3.00	14.49	393.2	14.26	339.6	14.02	296.5
3.50	14.50	394.8	14.26	341.0	14.03	297.7
4.00	14.51	396.4	14.27	342.4	14.04	299.0
5.00	14.52	399.6	14.29	345.2	14.06	301.5
6.00	14.54	402.8	14.31	348.1	14.07	304.0
8.00	14.56	409.2	14.34	353.7	14.11	309.1
10.00	14.59	415.7	14.37	359.3	14.14	314.1
15.00	14.66	432.0	14.44	373.6	14.22	326.8
20.00	14.73	448.6	14.52	388.0	14.30	339.6
25.00	14.79	465.4	14.58	402.5	14.38	352.4
30.00	14.86	482.4	14.65	417.3	14.45	365.4
35.00	14.91	499.7	14.71	432.2	14.51	378.5
40.00	14.97	517.3	14.78	447.2	14.58	391.7
50.00	15.08	553.2	14.89	478.0	14.70	418.6
60.00	15.19	590.2	15.00	509.6	14.82	446.1
80.00	15.38	667.6	15.20	575.2	15.03	503.0
100.00	15.55	749.6	15.39	644.4	15.22	562.6
Temperature	220 K		240 K		260 K	
	Pressure (MPa)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )
0.01	0.005 491	6.062	0.005 028	6.599	0.004 637	7.135
0.05	0.027 95	6.033	0.025 47	6.580	0.023 42	7.123
0.10	13.47	225.1	0.051 84	6.558	0.047 45	7.110
0.15	13.47	225.2	12.94	179.0	0.072 15	7.098
0.20	13.47	225.3	12.95	179.1	0.097 57	7.088
0.25	13.47	225.4	12.95	179.2	0.123 8	7.080
0.30	13.47	225.5	12.95	179.3	0.150 8	7.074
0.35	13.47	225.6	12.95	179.4	12.39	144.5
0.40	13.47	225.7	12.95	179.5	12.39	144.6
0.50	13.48	225.9	12.95	179.7	12.39	144.7
0.60	13.48	226.2	12.96	179.9	12.40	144.9
0.80	13.48	226.6	12.96	180.3	12.41	145.3
1.00	13.49	227.0	12.97	180.7	12.41	145.7
1.50	13.50	228.1	12.98	181.6	12.43	146.6
2.00	13.51	229.2	13.00	182.6	12.45	147.5
2.50	13.52	230.2	13.01	183.6	12.47	148.5
3.00	13.53	231.3	13.02	184.5	12.49	149.4
3.50	13.54	232.4	13.04	185.5	12.50	150.3
4.00	13.55	233.4	13.05	186.5	12.52	151.2
5.00	13.58	235.6	13.08	188.4	12.55	153.0
6.00	13.60	237.7	13.10	190.3	12.59	154.8
8.00	13.64	242.0	13.16	194.1	12.65	158.3
10.00	13.68	246.2	13.20	197.8	12.71	161.8
15.00	13.78	256.8	13.32	207.1	12.85	170.4
20.00	13.87	267.3	13.43	216.3	12.98	178.7
25.00	13.95	277.9	13.53	225.4	13.10	186.8
30.00	14.04	288.5	13.62	234.5	13.21	194.9
35.00	14.11	299.1	13.71	243.6	13.31	202.9
40.00	14.19	309.8	13.80	252.6	13.41	210.8
50.00	14.33	331.3	13.96	270.7	13.59	226.6
60.00	14.46	353.2	14.10	288.9	13.75	242.3
80.00	14.70	397.9	14.36	325.8	14.04	273.8
100.00	14.91	444.3	14.59	363.7	14.29	305.9

TABLE 4. Viscosity and density of propane—Continued

Temperature Pressure (MPa)	280 K		300 K		320 K	
	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)
0.01	0.004 304	7.670	0.004 015	8.203	0.003 763	8.732
0.05	0.021 69	7.663	0.020 20	8.199	0.018 91	8.731
0.10	0.043 81	7.655	0.040 73	8.196	0.038 07	8.731
0.15	0.066 41	7.649	0.061 59	8.193	0.057 48	8.732
0.20	0.089 50	7.644	0.082 81	8.192	0.077 15	8.733
0.25	0.113 1	7.640	0.104 4	8.191	0.097 10	8.735
0.30	0.137 3	7.638	0.126 4	8.192	0.117 3	8.738
0.35	0.162 1	7.637	0.148 8	8.194	0.137 9	8.741
0.40	0.187 5	7.637	0.171 6	8.196	0.158 7	8.746
0.50	0.240 6	7.644	0.218 7	8.205	0.201 4	8.757
0.60	0.278	7.649	0.267 9	8.219	0.245 4	8.772
0.80	0.353	7.663	0.373 8	8.265	0.338 4	8.815
1.00	0.428	7.677	0.480 1	8.333	0.439 0	8.877
1.50	0.574	7.711	0.636 4	8.437	0.740 0	9.148
2.00	0.720	7.745	0.792 7	8.571	1.036	9.419
2.50	0.866	7.779	0.949 0	8.735	1.332	9.690
3.00	1.012	7.813	1.105 3	8.929	1.628	9.961
3.50	1.158	7.847	1.261 6	9.153	1.924	10.232
4.00	1.304	7.881	1.417 9	9.407	2.220	10.503
5.00	1.640	7.949	1.744 1	9.881	2.812	10.944
6.00	1.976	8.017	2.070 3	10.385	3.404	11.385
8.00	2.644	8.117	2.746 5	11.259	4.396	11.926
10.00	3.312	8.217	3.422 7	12.133	5.388	12.467
15.00	4.640	8.317	4.748 9	13.507	7.380	13.008
20.00	5.968	8.417	6.075 1	14.881	9.372	13.549
25.00	7.296	8.517	7.401 3	16.255	11.364	14.090
30.00	8.624	8.617	8.727 5	17.629	13.356	14.631
35.00	9.952	8.717	10.053 7	19.003	15.348	15.172
40.00	11.280	8.817	11.379 9	20.377	17.340	15.713
50.00	14.312	8.967	14.341	22.085	21.332	16.754
60.00	17.344	9.117	17.303	23.793	25.324	17.795
80.00	23.712	9.317	23.665	26.801	33.316	19.336
100.00	30.080	9.517	30.027	29.809	41.308	20.877

Temperature Pressure (MPa)	340 K		360 K		370 K	
	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)
0.01	0.003 541	9.258	0.003 344	9.779	0.003 253	10.04
0.05	0.017 78	9.259	0.016 78	9.782	0.016 32	10.04
0.10	0.035 75	9.261	0.033 70	9.786	0.032 77	10.05
0.15	0.053 91	9.264	0.050 78	9.791	0.049 35	10.05
0.20	0.072 27	9.268	0.068 01	9.796	0.066 07	10.06
0.25	0.090 84	9.271	0.085 40	9.801	0.082 93	10.06
0.30	0.109 6	9.276	0.103 0	9.807	0.099 94	10.07
0.35	0.128 6	9.281	0.120 7	9.813	0.117 1	10.08
0.40	0.147 9	9.287	0.138 6	9.820	0.134 4	10.08
0.50	0.187 0	9.301	0.174 9	9.836	0.169 5	10.10
0.60	0.227 2	9.317	0.212 0	9.853	0.205 2	10.12
0.80	0.310 9	9.359	0.288 5	9.896	0.278 7	10.16
1.00	0.399 6	9.414	0.3685	9.948	0.355 2	10.21
1.50	0.649 8	9.627	0.587 2	10.13	0.561 9	10.39
2.00	0.963 6	9.999	0.841 4	10.41	0.796 6	10.64
2.50	1.335	59.93	1.151	10.85	1.071	11.01
3.00	1.745	61.63	1.561	11.55	1.408	11.56
3.50	2.192	63.19	2.057	13.06	1.859	12.44
4.00	2.671	64.64	2.553	14.57	2.329	13.32
5.00	3.687	67.31	3.549	17.08	3.201	14.20
6.00	4.743	69.74	4.545	19.59	4.073	15.08
8.00	6.441	74.11	6.041	24.60	5.505	16.96
10.00	8.139	78.02	7.537	29.61	6.937	18.84
15.00	11.54	86.56	10.533	39.62	9.369	21.72

TABLE 4. Viscosity and density of propane—Continued

Temperature	340 K		360 K		370 K	
	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)
20.00	11.02	94.05	10.47	81.38	10.19	75.77
25.00	11.27	100.9	10.77	88.24	10.52	82.67
30.00	11.48	107.3	11.02	94.56	10.79	88.95
35.00	11.66	113.4	11.24	100.5	11.02	94.81
40.00	11.83	119.3	11.43	106.1	11.23	100.4
50.00	12.12	130.5	11.75	116.8	11.57	110.8
60.00	12.37	141.3	12.03	127.0	11.86	120.7
80.00	12.79	162.0	12.49	146.3	12.34	139.5
100.00	13.13	182.3	12.86	165.0	12.72	157.5
Temperature	380 K		400 K		420 K	
	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)
0.01	0.003 167	10.30	0.003 008	10.81	0.002 865	11.31
0.05	0.015 88	10.30	0.015 08	10.81	0.014 36	11.32
0.10	0.031 88	10.30	0.030 25	10.82	0.028 79	11.33
0.15	0.048 00	10.31	0.045 53	10.82	0.043 30	11.33
0.20	0.064 25	10.32	0.060 89	10.83	0.057 89	11.34
0.25	0.080 61	10.32	0.076 36	10.84	0.072 55	11.35
0.30	0.097 11	10.33	0.091 93	10.85	0.087 30	11.36
0.35	0.113 7	10.34	0.107 6	10.86	0.102 1	11.37
0.40	0.130 5	10.35	0.123 4	10.86	0.117 0	11.38
0.50	0.164 4	10.36	0.155 2	10.88	0.147 1	11.39
0.60	0.198 9	10.38	0.187 6	10.90	0.177 6	11.41
0.80	0.269 7	10.42	0.253 5	10.94	0.239 5	11.46
1.00	0.343 0	10.48	0.321 5	11.00	0.302 9	11.51
1.50	0.539 3	10.64	0.500 8	11.15	0.468 6	11.66
2.00	0.758 4	10.88	0.695 8	11.36	0.645 7	11.85
2.50	1.008	11.21	0.910 1	11.63	0.836 1	12.09
3.00	1.300	11.66	1.149	11.99	1.042	12.38
3.50	1.658	12.32	1.418	12.44	1.266	12.75
4.00	2.131	13.34	1.728	13.04	1.511	13.19
5.00	4.768	22.12	2.549	14.96	2.085	14.40
6.00	7.123	36.52	3.902	19.19	2.813	16.27
8.00	8.149	46.12	6.554	32.80	4.772	23.30
10.00	8.660	52.02	7.559	40.84	6.282	31.49
15.00	9.416	62.50	8.683	52.84	7.905	44.62
20.00	9.900	70.57	9.303	61.30	8.690	53.39
25.00	10.26	77.52	9.744	68.34	9.218	60.48
30.00	10.56	83.78	10.09	74.58	9.620	66.68
35.00	10.81	89.59	10.38	80.30	9.946	72.31
40.00	11.02	95.07	10.62	85.65	10.22	77.53
50.00	11.39	105.3	11.03	95.58	10.67	87.17
60.00	11.70	115.0	11.37	104.8	11.04	96.06
80.00	12.19	133.2	11.90	122.1	11.62	112.5
100.00	12.59	150.6	12.32	138.5	12.07	128.0
Temperature	440 K		460 K		480 K	
	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)
0.01	0.002 735	11.81	0.002 615	12.31	0.002 506	12.79
0.05	0.013 70	11.82	0.013 10	12.31	0.012 55	12.80
0.10	0.027 46	11.83	0.026 25	12.32	0.025 14	12.81
0.15	0.041 28	11.84	0.039 45	12.33	0.037 77	12.82
0.20	0.055 17	11.84	0.052 70	12.34	0.050 45	12.83
0.25	0.069 12	11.85	0.066 01	12.35	0.063 17	12.84
0.30	0.083 14	11.86	0.079 37	12.36	0.075 94	12.84
0.35	0.097 22	11.87	0.092 78	12.36	0.088 75	12.85
0.40	0.111 4	11.88	0.106 3	12.37	0.101 6	12.86
0.50	0.139 9	11.90	0.133 4	12.39	0.127 4	12.88

TABLE 4. Viscosity and density of propane—Continued

Temperature	440 K		460 K		480 K	
	Pressure (MPa)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )
0.60	0.168 7	11.92	0.160 7	12.42	0.153 5	12.90
0.80	0.227 1	11.96	0.216 0	12.46	0.206 1	12.95
1.00	0.286 7	12.01	0.272 3	12.51	0.259 4	13.00
1.50	0.441 1	12.16	0.417 3	12.65	0.396 2	13.13
2.00	0.604 2	12.34	0.568 8	12.82	0.538 2	13.30
2.50	0.776 8	12.55	0.727 5	13.02	0.685 6	13.48
3.00	0.960 0	12.81	0.893 9	13.25	0.838 8	13.70
3.50	1.155	13.12	1.069	13.53	0.997 9	13.95
4.00	1.363	13.48	1.252	13.84	1.163	14.22
5.00	1.826	14.40	1.648	14.59	1.514	14.88
6.00	2.361	15.65	2.086	15.56	1.892	15.68
8.00	3.683	19.58	3.096	18.26	2.727	17.76
10.00	5.090	25.31	4.221	22.09	3.642	20.55
15.00	7.110	37.92	6.346	32.80	5.660	29.14
20.00	8.073	46.73	7.467	41.27	6.889	36.91
25.00	8.693	53.78	8.177	48.14	7.678	43.44
30.00	9.153	59.90	8.694	54.09	8.249	49.16
35.00	9.520	65.41	9.102	59.45	8.696	54.32
40.00	9.827	70.50	9.440	64.40	9.063	59.10
50.00	10.32	79.85	9.981	73.45	9.646	67.84
60.00	10.72	88.44	10.41	81.74	10.10	75.83
80.00	11.34	104.2	11.07	96.92	10.80	90.44
100.00	11.81	119.0	11.57	111.0	11.33	103.9

Temperature	500 K		520 K		540 K	
	Pressure (MPa)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )
0.01	0.002 406	13.27	0.002 313	13.75	0.002 228	14.21
0.05	0.012 04	13.28	0.011 58	13.75	0.011 15	14.22
0.10	0.024 12	13.29	0.023 19	13.76	0.022 32	14.23
0.15	0.036 24	13.30	0.034 82	13.77	0.033 52	14.24
0.20	0.048 39	13.31	0.046 49	13.78	0.044 74	14.25
0.25	0.060 58	13.32	0.058 19	13.79	0.055 99	14.26
0.30	0.072 80	13.32	0.069 92	13.80	0.067 26	14.26
0.35	0.085 06	13.34	0.081 67	13.81	0.078 55	14.28
0.40	0.097 35	13.34	0.093 46	13.82	0.089 88	14.28
0.50	0.122 1	13.36	0.117 1	13.84	0.112 6	14.30
0.60	0.146 9	13.38	0.140 9	13.86	0.135 4	14.32
0.80	0.197 1	13.43	0.188 9	13.90	0.181 4	14.37
1.00	0.247 8	13.48	0.237 3	13.95	0.227 7	14.41
1.50	0.377 5	13.61	0.360 7	14.08	0.345 4	14.54
2.00	0.511 2	13.77	0.487 3	14.23	0.465 7	14.69
2.50	0.649 2	13.94	0.617 2	14.40	0.588 7	14.85
3.00	0.791 6	14.15	0.750 6	14.59	0.714 4	15.03
3.50	0.938 5	14.37	0.887 4	14.80	0.842 8	15.23
4.00	1.090	14.63	1.028	15.04	0.973 8	15.45
5.00	1.407	15.21	1.319	15.56	1.244	15.94
6.00	1.743	15.90	1.623	16.18	1.523	16.49
8.00	2.466	17.62	2.267	17.66	2.107	17.80
10.00	3.241	19.80	2.945	19.47	2.715	19.36
15.00	5.078	26.67	4.600	25.06	4.212	24.03
20.00	6.354	33.55	5.872	31.02	5.445	29.16
25.00	7.205	39.61	6.763	36.53	6.355	34.10
30.00	7.823	45.01	7.418	41.55	7.038	38.71
35.00	8.305	49.93	7.930	46.19	7.576	43.03
40.00	8.699	54.50	8.349	50.53	8.015	47.11
50.00	9.322	62.89	9.009	58.55	8.708	54.72
60.00	9.807	70.59	9.520	65.93	9.244	61.78
80.00	10.54	84.65	10.29	79.46	10.05	74.78
100.00	11.10	97.61	10.87	91.93	10.65	86.78

TABLE 4. Viscosity and density of propane—Continued

Temperature	560 K		580 K		600 K	
	Pressure (MPa)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )
0.01	0.002 148	14.67	0.002 074	15.13	0.002 005	15.57
0.05	0.010 75	14.68	0.010 38	15.13	0.010 03	15.58
0.10	0.021 52	14.69	0.020 77	15.14	0.020 07	15.58
0.15	0.032 31	14.70	0.031 18	15.15	0.030 13	15.59
0.20	0.043 12	14.71	0.041 61	15.16	0.040 20	15.60
0.25	0.053 95	14.72	0.052 05	15.17	0.050 29	15.61
0.30	0.064 80	14.72	0.062 52	15.18	0.060 39	15.62
0.35	0.075 67	14.73	0.072 99	15.19	0.070 51	15.63
0.40	0.086 56	14.74	0.083 49	15.20	0.080 64	15.64
0.50	0.108 4	14.76	0.104 5	15.21	0.100 9	15.66
0.60	0.130 3	14.78	0.125 7	15.23	0.121 3	15.68
0.80	0.174 5	14.83	0.168 1	15.28	0.162 2	15.72
1.00	0.218 9	14.87	0.210 8	15.32	0.203 3	15.76
1.50	0.331 5	15.00	0.318 8	15.44	0.307 1	15.88
2.00	0.446 3	15.14	0.428 5	15.58	0.412 3	16.01
2.50	0.563 2	15.29	0.540 0	15.73	0.518 9	16.16
3.00	0.682 2	15.47	0.653 2	15.90	0.626 9	16.32
3.50	0.803 3	15.66	0.768 0	16.08	0.736 1	16.49
4.00	0.926 5	15.86	0.884 4	16.27	0.846 7	16.68
5.00	1.179	16.32	1.122	16.70	1.071	17.08
6.00	1.438	16.83	1.365	17.17	1.300	17.53
8.00	1.975	18.01	1.864	18.26	1.767	18.53
10.00	2.528	19.38	2.373	19.49	2.242	19.66
15.00	3.895	23.40	3.632	23.01	3.411	22.80
20.00	5.073	27.81	4.749	26.85	4.467	26.18
25.00	5.984	32.20	5.648	30.73	5.346	29.62
30.00	6.685	36.39	6.358	34.52	6.056	33.02
35.00	7.241	40.38	6.928	38.17	6.635	36.35
40.00	7.699	44.19	7.399	41.71	7.117	39.60
50.00	8.420	51.37	8.145	48.44	7.883	45.88
60.00	8.978	58.09	8.723	54.81	8.478	51.89
80.00	9.816	70.55	9.589	66.72	9.371	63.25
100.00	10.44	82.10	10.23	77.83	10.03	73.92

Figure 20 indicates the range of applicability of the present representation as well as the uncertainty in various thermodynamic regions derived from a comparison with the critically assessed experimental data. In the temperature range 293–600 K and for densities below 0.05 mol L<sup>-1</sup> the uncertainty is ±0.4%; in the temperature range 278–600 K and for densities up to 0.1 mol L<sup>-1</sup> the uncertainty is ±1%. In all other temperature and pressure ranges, in which the correlation is based on primary experimental data, its uncertainty is estimated to be ±2.5% corresponding to three standard deviations. The uncertainty increases up to ±4%, when extrapolations are performed in most of the other temperature and pressure ranges of the MBWR equation of state. Finally, the uncertainty increases further at the lowest temperatures and highest densities as well as in the zero-density limit and in the vapor phase below 230 K.

The viscosity surface of propane is shown in Fig. 21. The saturation boundary indicates that an essential part of the surface represents metastable and unstable states, which are experimentally not accessible or only partly accessible. Computations in the two-phase region may be necessary, if propane is used as reference fluid in corresponding states

calculations for other fluids or fluid mixtures. Figure 21 also includes the upper pressure limit of the equation of state used. A fair extrapolation beyond the applicability range of the equation of state can be expected due to the smoothness of the viscosity surface. Such findings have already been discussed for the experimental results by Babb and Scott<sup>66</sup> and by Nasarenko and Golubev.<sup>67</sup>

## 7. Conclusion

A critical assessment was made of the available experimental data for the dynamic viscosity of propane. The representation of the viscosity surface of propane was developed, guided by theoretical considerations for the zero-density limit and the initial density dependence and to a certain degree for the high-density terms. The correlation represents the experimental data within their ascribed uncertainties as well as the phenomenological behavior of the viscosity over a wide range of temperatures and pressures. The uncertainty ascribed to the viscosity surface is within ±2.5%, when based on experimental viscosity data, or within ±4% in the remaining thermodynamic ranges of the

TABLE 5. Viscosity and density of propane along the saturation line

Temperature (K)	Pressure (MPa)	Saturated liquid		Saturated vapor	
		Density (mol L <sup>-1</sup> )	Viscosity (μPa s)	Density (mol L <sup>-1</sup> )	Viscosity (μPa s)
85.48 <sup>a</sup>	1.691 × 10 <sup>-10</sup>	16.64	11020.0	2.381 × 10 <sup>-10</sup>	2.639
90.0	9.666 × 10 <sup>-10</sup>	16.53	7529.0	1.291 × 10 <sup>-9</sup>	2.744
95.0	5.400 × 10 <sup>-9</sup>	16.41	5250.0	6.830 × 10 <sup>-9</sup>	2.861
100.0	2.507 × 10 <sup>-8</sup>	16.30	3841.0	3.014 × 10 <sup>-8</sup>	2.979
105.0	9.937 × 10 <sup>-8</sup>	16.18	2920.0	1.139 × 10 <sup>-7</sup>	3.099
110.0	3.439 × 10 <sup>-7</sup>	16.07	2291.0	3.762 × 10 <sup>-7</sup>	3.220
115.0	1.058 × 10 <sup>-6</sup>	15.95	1846.0	1.108 × 10 <sup>-6</sup>	3.343
120.0	2.940 × 10 <sup>-6</sup>	15.84	1521.0	2.948 × 10 <sup>-6</sup>	3.467
125.0	7.463 × 10 <sup>-6</sup>	15.72	1278.0	7.183 × 10 <sup>-6</sup>	3.591
130.0	1.750 × 10 <sup>-5</sup>	15.61	1092.0	1.619 × 10 <sup>-5</sup>	3.717
135.0	3.825 × 10 <sup>-5</sup>	15.49	946.1	3.408 × 10 <sup>-5</sup>	3.843
140.0	7.856 × 10 <sup>-5</sup>	15.38	829.9	6.749 × 10 <sup>-5</sup>	3.970
145.0	1.526 × 10 <sup>-4</sup>	15.26	735.7	1.266 × 10 <sup>-4</sup>	4.097
150.0	2.821 × 10 <sup>-4</sup>	15.15	658.2	2.262 × 10 <sup>-4</sup>	4.226
155.0	4.985 × 10 <sup>-4</sup>	15.03	593.3	3.869 × 10 <sup>-4</sup>	4.354
160.0	8.461 × 10 <sup>-4</sup>	14.92	538.5	6.365 × 10 <sup>-4</sup>	4.483
165.0	1.385 × 10 <sup>-3</sup>	14.80	491.5	1.011 × 10 <sup>-3</sup>	4.612
170.0	2.193 × 10 <sup>-3</sup>	14.68	450.9	1.555 × 10 <sup>-3</sup>	4.741
175.0	3.372 × 10 <sup>-3</sup>	14.56	415.4	2.324 × 10 <sup>-3</sup>	4.871
180.0	5.043 × 10 <sup>-3</sup>	14.45	384.2	3.385 × 10 <sup>-3</sup>	5.000
185.0	7.360 × 10 <sup>-3</sup>	14.33	356.4	4.812 × 10 <sup>-3</sup>	5.129
190.0	0.010 50	14.21	331.6	6.696 × 10 <sup>-3</sup>	5.258
195.0	0.014 67	14.09	309.4	9.135 × 10 <sup>-3</sup>	5.386
200.0	0.020 11	13.97	289.3	0.012 24	5.515
205.0	0.027 09	13.84	271.0	0.016 13	5.643
210.0	0.035 91	13.72	254.3	0.020 94	5.771
215.0	0.046 89	13.59	239.1	0.026 82	5.898
220.0	0.060 41	13.47	225.1	0.033 91	6.026
225.0	0.076 83	13.34	212.2	0.042 38	6.154
230.0	0.096 59	13.21	200.2	0.052 40	6.282
235.0	0.120 1	13.08	189.2	0.064 17	6.411
240.0	0.147 8	12.94	178.9	0.077 87	6.540
245.0	0.180 2	12.81	169.3	0.093 73	6.670
250.0	0.217 8	12.67	160.4	0.112 0	6.802
255.0	0.261 1	12.53	152.0	0.132 8	6.936
260.0	0.310 7	12.38	144.2	0.156 6	7.073
265.0	0.366 9	12.24	136.8	0.183 5	7.212
270.0	0.430 5	12.09	129.8	0.213 9	7.355
275.0	0.502 0	11.93	123.3	0.248 1	7.502
280.0	0.581 9	11.77	117.1	0.286 6	7.655
285.0	0.670 9	11.61	111.2	0.329 7	7.814
290.0	0.769 5	11.45	105.6	0.378 0	7.980
295.0	0.878 3	11.27	100.2	0.431 9	8.155
300.0	0.998 0	11.09	95.14	0.492 2	8.341
305.0	1.129	10.91	90.24	0.559 6	8.539
310.0	1.273	10.72	85.53	0.635 0	8.752
315.0	1.429	10.52	80.98	0.719 4	8.982
320.0	1.599	10.31	76.58	0.814 1	9.234
325.0	1.783	10.09	72.28	0.920 9	9.512
330.0	1.983	9.850	68.08	1.042	9.822
335.0	2.198	9.599	63.93	1.180	10.17
340.0	2.432	9.327	59.81	1.339	10.58
345.0	2.683	9.028	55.67	1.525	11.06
350.0	2.955	8.694	51.44	1.747	11.63
355.0	3.248	8.307	47.03	2.022	12.36
360.0	3.566	7.835	42.24	2.384	13.37
365.0	3.912	7.182	36.52	2.929	14.98
369.825 <sup>b</sup>	4.286	5.000	22.89	4.835	22.12

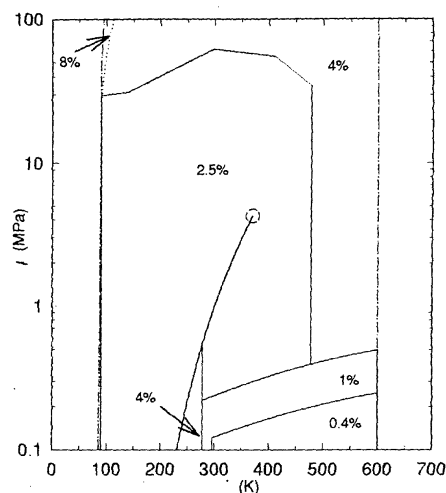
<sup>a</sup>Triple point.<sup>b</sup>Critical point.

FIG. 20. The extent of the viscosity representation and its estimated uncertainty.

MBWR equation of state apart from the lowest temperatures. This uncertainty should be adequate for many engineering purposes. Nevertheless, it would be desirable to have more measurements in the vapor phase, particularly at low temperatures down to 200 K, along the saturation line in the vapor phase, in the whole density range at supercritical temperatures above 420 K, and in the vicinity of the critical point. Owing to the complete absence of experimental data in the critical region, the expected enhancement in the viscosity could not be taken into account. Tables 4 and 5 give values of viscosity and density as functions of temperature and pressure to provide easy reference and to enable validation of computer codes.

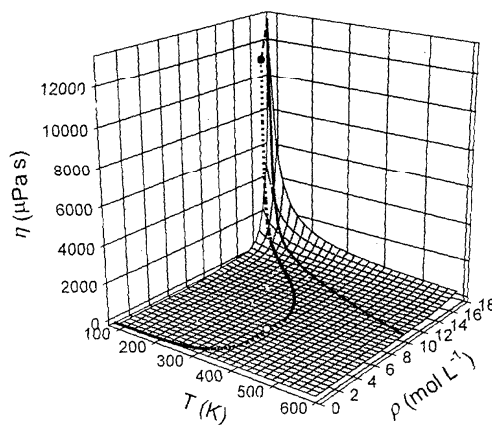


FIG. 21. Viscosity surface correlation of propane. (●) triple point, (○) critical point, (—) range of EOS (100 MPa), (·····) saturation line, (---) melting line.

## 8. Acknowledgments

One of the authors, C.K., is grateful to the Ministry of Culture of Mecklenburg-Vorpommern, Germany, for the Award of the Landesgraduiertenförderung. The research at the University Rostock was supported 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. The authors thank Marcia Huber for helpful remarks in the course of the work.

- <sup>1</sup> Y. Tanaka and T. Makita, *Rev. Phys. Chem. Jpn.* **45**, 93 (1976).
- <sup>2</sup> K. Stephan and K. Lucas, *Viscosity of Dense Fluids* (Plenum, New York, 1979), p. 219.
- <sup>3</sup> P. M. Holland, H. J. M. Hanley, K. E. Gubbins, and J. M. Haile, *J. Phys. Chem. Ref. Data* **8**, 559 (1979).
- <sup>4</sup> B. A. Younglove and J. F. Ely, *J. Phys. Chem. Ref. Data* **16**, 577 (1987).
- <sup>5</sup> D. E. Diller, *J. Chem. Eng. Data* **27**, 240 (1982).
- <sup>6</sup> J. F. Ely and H. J. M. Hanley, *Ind. Eng. Chem. Fund.* **20**, 323 (1981).
- <sup>7</sup> J. F. Ely and H. J. M. Hanley, *Ind. Eng. Chem. Fund.* **22**, 90 (1983).
- <sup>8</sup> J. F. Ely, *Proceedings of the 63rd Annual Convention* (Gas Processors Association, Tulsa, OK, 1984), p. 9.
- <sup>9</sup> M. Huber, *NIST Standard Reference Database 4—Thermophysical Properties of Hydrocarbon Mixtures Database (SUPERTRAPP) Version 1.04* (NIST Standard Reference Data Program, 1990).
- <sup>10</sup> E. Vogel and C. Küchenmeister, *High Temp.-High Press.* **29**, 397 (1997).
- <sup>11</sup> E. Vogel, *Int. J. Thermophys.* **16**, 1335 (1995).
- <sup>12</sup> E. Bich and E. Vogel, *Int. J. Thermophys.* **12**, 27 (1991).
- <sup>13</sup> M. E. Baltatu, R. A. Chong, M. L. Huber, and A. Laesecke, *Int. J. Thermophys.* (in press).
- <sup>14</sup> 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).
- <sup>15</sup> S. Hendl, J. Millat, E. Vogel, V. Vesovic, W. A. Wakeham, J. Luettmer-Strathmann, J. V. Sengers, and M. J. Assael, *Int. J. Thermophys.* **15**, 1 (1994).
- <sup>16</sup> A. Faghri, W. A. Wakeham, V. Vesovic, J. T. R. Watson, J. Millat, and E. Vogel, *J. Phys. Chem. Ref. Data* **24**, 1649 (1995).
- <sup>17</sup> J. Kestin and W. Leidenfrost, *Physica* **25**, 1033 (1959).
- <sup>18</sup> J. Kestin, M. Sokolov, and W. Wakeham, *Appl. Sci. Res.* **27**, 241 (1973).
- <sup>19</sup> J. Kestin, S. T. Ro, and W. A. Wakeham, *Trans. Faraday Soc.* **67**, 2308 (1971).
- <sup>20</sup> J. Kestin, H. E. Khalifa, and W. A. Wakeham, *J. Chem. Phys.* **66**, 1132 (1977).
- <sup>21</sup> Y. Abe, J. Kestin, H. E. Khalifa, and W. A. Wakeham, *Physica A* **93**, 155 (1978).
- <sup>22</sup> Y. Abe, J. Kestin, H. E. Khalifa, and W. A. Wakeham, *Ber. Bunsenges. Phys. Chem.* **83**, 271 (1979).
- <sup>23</sup> A. Klemenc and W. Remi, *Monatsh. Chem.* **44**, 307 (1923).
- <sup>24</sup> T. Titani, *Bull. Chem. Soc. Jpn.* **5**, 98 (1930).
- <sup>25</sup> M. Trautz and F. Kurz, *Ann. Phys. (Leipzig)* **9**, 981 (1931).
- <sup>26</sup> M. Trautz and K. G. Sorg, *Ann. Phys. (Leipzig)* **10**, 81 (1931).
- <sup>27</sup> H. Adzumi, *Bull. Chem. Soc. Jpn.* **12**, 199 (1937).
- <sup>28</sup> B. H. Sage and W. N. Lacey, *Ind. Eng. Chem.* **30**, 829 (1938).
- <sup>29</sup> R. Wobser and F. Mueller, *Kolloid-Beih.* **52**, 165 (1941).
- <sup>30</sup> H. Sentfleben and H. Gladisch, *Z. Physik* **125**, 653 (1949).
- <sup>31</sup> I. F. Golubev and V. A. Petrov, *Tr. Gos. Inst. Azotnoi Prom.* **2**, 5 (1953).
- <sup>32</sup> J. D. Lambert, K. J. Cotton, M. W. Pailthorpe, A. M. Robinson, J. Scrivins, W. R. F. Vale, and R. M. Young, *Proc. R. Soc. London Ser. A* **231**, 280 (1955).
- <sup>33</sup> L. T. Carmichael, V. M. Berry, and B. H. Sage, *J. Chem. Eng. Data* **9**, 411 (1964).
- <sup>34</sup> M. Diaz Pena and J. A. R. Cheda, *An. Quim. (Esp.)* **71**, 34 (1975).
- <sup>35</sup> M. Diaz Pena and J. A. R. Cheda, *An. Quim. (Esp.)* **71**, 257 (1975).
- <sup>36</sup> K. Nagaoka, Y. Yamashita, Y. Tanaka, H. Kubota, and T. Makita, *J. Chem. Eng. Jpn.* **19**, 263 (1986).
- <sup>37</sup> R. Di Pippo, J. Kestin, and J. H. Whitelaw, *Physica* **32**, 2064 (1966).
- <sup>38</sup> E. Bich, J. Millat, and E. Vogel, *Wiss. Z. W.-Pieck-Univ. Rostock* **36**, 5 (1987).
- <sup>39</sup> E. Bich, J. Millat, and E. Vogel, *J. Phys. Chem. Ref. Data* **19**, 1289 (1990).
- <sup>40</sup> J. Kestin, S. T. Ro, and W. A. Wakeham, *Physica* **58**, 165 (1972).
- <sup>41</sup> J. Kestin and E. A. Mason, *AIP Conf. Proc.* **11**, 137 (1973).
- <sup>42</sup> B. Najafi, E. A. Mason, and J. Kestin, *Physica A* **119**, 387 (1983).
- <sup>43</sup> J. Kestin, K. Knierim, E. A. Mason, B. Najafi, S. T. Ro, and M. Waldman, *J. Phys. Chem. Ref. Data* **13**, 229 (1984).
- <sup>44</sup> J. Kestin, S. T. Ro, and W. A. Wakeham, *J. Chem. Phys.* **56**, 4119 (1972).
- <sup>45</sup> E. Vogel, *Ber. Bunsenges. Phys. Chem.* **88**, 997 (1984).
- <sup>46</sup> L. v. Körtvelyessy, *Thermoelement-Praxis* (Vulkan, Essen, 1981), p. 447.
- <sup>47</sup> B. E. Eakin and R. T. Ellington, *Trans. Am. Inst. Min. Metall. Pet. Eng.* **216**, 85 (1959).
- <sup>48</sup> 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 DC, 1959).
- <sup>49</sup> K. E. Starling, B. E. Eakin, and R. T. Ellington, *AIChE J.* **6**, 438 (1960).
- <sup>50</sup> K. E. Starling, Master's thesis, Illinois Institute of Technology, Chicago, Illinois, 1960.
- <sup>51</sup> K. E. Starling, B. E. Eakin, J. P. Dolan, and R. T. Ellington, *Proceedings of the 2nd Symposium on Thermophysical Properties*, edited by P. F. Masi and D. H. Tsai (American Society of Mechanical Engineers, New York, 1962), p. 530.
- <sup>52</sup> J. G. Giddings, J. T. F. Kao, and R. Kobayashi, *J. Chem. Phys.* **45**, 578 (1966).
- <sup>53</sup> E. T. S. Huang, G. W. Swift, and F. Kurata, *AIChE J.* **12**, 932 (1966).
- <sup>54</sup> H. J. Strumpf, A. F. Collings, and C. J. Pings, *J. Chem. Phys.* **60**, 3109 (1974).
- <sup>55</sup> D. E. Diller, *Proceedings of the 8th Symposium on Thermophysical Properties*, edited by J. V. Sengers (American Society of Mechanical Engineers, New York, 1982), Vol. 1, p. 219.
- <sup>56</sup> W. R. van Wijk, J. H. van der Veen, H. C. Brinkman, and W. A. Seeder, *Physica* **7**, 45 (1940).
- <sup>57</sup> S. F. Gerf and G. I. Galkov, *Zh. Tekh. Fiz.* **10**, 725 (1940).
- <sup>58</sup> G. I. Galkov and S. F. Gerf, *Zh. Tekh. Fiz.* **11**, 613 (1941).
- <sup>59</sup> M. R. Lipkin, J. A. Davison, and S. S. Kurtz, Jr., *Ind. Eng. Chem.* **34**, 976 (1942).
- <sup>60</sup> A. S. Smith and G. G. Brown, *Ind. Eng. Chem.* **35**, 705 (1943).
- <sup>61</sup> L. B. Bicher, Jr. and D. L. Katz, *Ind. Eng. Chem.* **35**, 754 (1943).
- <sup>62</sup> E. W. Comings, B. J. Mayland, and R. S. Egly, *Univ. Ill. Eng. Exptl. Sta. Bull.* **354**, 1 (1944).
- <sup>63</sup> J. D. Baron, J. G. Roof, and F. W. Wells, *J. Chem. Eng. Data* **4**, 283 (1959).
- <sup>64</sup> G. W. Swift, J. A. Christy, and F. Kurata, *AIChE J.* **5**, 98 (1959).
- <sup>65</sup> G. W. Swift, J. Lohrenz, and F. Kurata, *AIChE J.* **6**, 415 (1960).
- <sup>66</sup> S. E. Babb, Jr. and G. J. Scott, *J. Chem. Phys.* **40**, 3666 (1964).
- <sup>67</sup> V. A. Nasarenko and I. F. Golubev, *Viscosity of ethane, propane and ethylene at pressures up to 5000 kg/cm<sup>2</sup> and at different temperatures* Report No. 916/76 deposited (Res. Inst. Sci. Tech. Inform., Tscherkassi, 1976) (in Russian).
- <sup>68</sup> J. H. Dymond, E. Bich, E. Vogel, W. A. Wakeham, V. Vesovic, and M. J. Assael, in *Transport Properties of Fluids. Their Correlation, Prediction and Estimation*, edited by J. Millat, J. H. Dymond, and C. A. Nieto de Castro (Cambridge University Press, Cambridge, 1996), Chap. 5, p. 66.
- <sup>69</sup> D. G. Friend and J. C. Rainwater, *Chem. Phys. Lett.* **107**, 590 (1984).
- <sup>70</sup> J. C. Rainwater and D. G. Friend, *Phys. Rev. A* **36**, 4062 (1987).
- <sup>71</sup> E. Bich and E. Vogel, in *Transport Properties of Fluids. Their Correlation, Prediction and Estimation*, edited by J. Millat, J. H. Dymond, and C. A. Nieto de Castro (Cambridge University Press, Cambridge, 1996), Chap. 5.2, p. 72.
- <sup>72</sup> G. A. Olchowy and J. V. Sengers, *Phys. Rev. Lett.* **61**, 15 (1988).
- <sup>73</sup> G. A. Olchowy and J. V. Sengers, *Int. J. Thermophys.* **10**, 417 (1989).
- <sup>74</sup> J. Luettmer-Strathmann, J. V. Sengers, and G. A. Olchowy, *J. Chem. Phys.* **103**, 7482 (1995).
- <sup>75</sup> J. Luettmer-Strathmann and J. V. Sengers, in *Transport Properties of Fluids. Their Correlation, Prediction and Estimation*, edited by J. Millat, J. H. Dymond, and C. A. Nieto de Castro (Cambridge University Press, Cambridge, 1996), Chap. 6, p. 113.
- <sup>76</sup> G. C. Maitland, M. Rigby, E. B. Smith, and W. A. Wakeham, *Intermolecular Forces: Their Origin and Determination* (Clarendon, Oxford, 1987).

- <sup>77</sup>F. R. W. Mc Court, J. J. M. Beenakker, W. E. Köhler, and I. Kuscer, *Nonequilibrium Phenomena in Polyatomic Gases* (Clarendon, Oxford, 1990), Vol. I.
- <sup>78</sup>F. R. W. Mc Court, J. J. M. Beenakker, W. E. Köhler, and I. Kuscer, *Nonequilibrium Phenomena in Polyatomic Gases* (Clarendon, Oxford, 1991), Vol. II.
- <sup>79</sup>E. L. Heck and A. S. Dickinson, *Mol. Phys.* **81**, 1325 (1994).
- <sup>80</sup>J. Millat, V. Vesovic, and W. A. Wakeham, in *Transport Properties of Fluids. Their Correlation, Prediction and Estimation*, edited by J. Millat, J. H. Dymond, and C. A. Nieto de Castro (Cambridge University Press, Cambridge, 1996), Chap. 4, p. 29.
- <sup>81</sup>E. Vogel, T. Strehlow, J. Millat, and W. A. Wakeham, *Z. Phys. Chem. (Leipzig)* **270**, 1145 (1989).
- <sup>82</sup>J. Millat and V. Vesovic, in *Transport Properties of Fluids. Their Correlation, Prediction and Estimation*, edited by J. Millat, J. H. Dymond, and C. A. Nieto de Castro (Cambridge University Press, Cambridge, 1996), Chap. 14.2, p. 332.
- <sup>83</sup>M. Takahashi, C. Yokoyama, and S. Takahashi, *Kagaku Kogaku Ronbunshu* **11**, 155 (1985).
- <sup>84</sup>M. Takahashi, C. Yokoyama, and S. Takahashi, *J. Chem. Eng. Data* **32**, 98 (1987).
- <sup>85</sup>J. T. R. Watson, R. S. Basu, and J. V. Sengers, *J. Phys. Chem. Ref. Data* **9**, 1255 (1980).
- <sup>86</sup>E. Vogel, E. Bich, and R. Nimz, *Physica A* **139**, 188 (1986).
- <sup>87</sup>K. M. de Reuck and B. Armstrong, *Cryogenics* **19**, 505 (1979).
- <sup>88</sup>A. J. Batschinski, *Z. Phys. Chem.* **84**, 643 (1913).
- <sup>89</sup>J. H. Hildebrand, *Science* **174**, 490 (1971).
- <sup>90</sup>J. H. Dymond, in *Transport Properties of Fluids. Their Correlation, Prediction and Estimation*, edited by J. Millat, J. H. Dymond, and C. A. Nieto de Castro (Cambridge University Press, Cambridge, 1996), Chap. 5.4, p. 90.
- <sup>91</sup>A. Laesecke, *Viskosität und Wärmeleitfähigkeit als thermodynamische Zustandsgrößen und ihre Darstellung durch Zustandsgleichungen*, Fortschritt-Berichte VDI, Reihe 3: Verfahrenstechnik (VDI, Düsseldorf, 1986), No. 117.
- <sup>92</sup>K. Stephan, R. Krauss, and A. Laesecke, *J. Phys. Chem. Ref. Data* **16**, 993 (1987).
- <sup>93</sup>A. Laesecke, R. Krauss, K. Stephan, and W. Wagner, *J. Phys. Chem. Ref. Data* **19**, 1089 (1990).
- <sup>94</sup>R. Krauss, J. Luettmer-Strathmann, J. V. Sengers, and K. Stephan, *Int. J. Thermophys.* **14**, 951 (1993).
- <sup>95</sup>R. Krauss, V. C. Weiss, T. A. Edison, J. V. Sengers, and K. Stephan, *Int. J. Thermophys.* **17**, 731 (1996).
- <sup>96</sup>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, MD, 1992).