

# Correlation for the Viscosity of Pentafluoroethane (R125) from the Triple Point to 500 K at Pressures up to 60 MPa<sup>†</sup>

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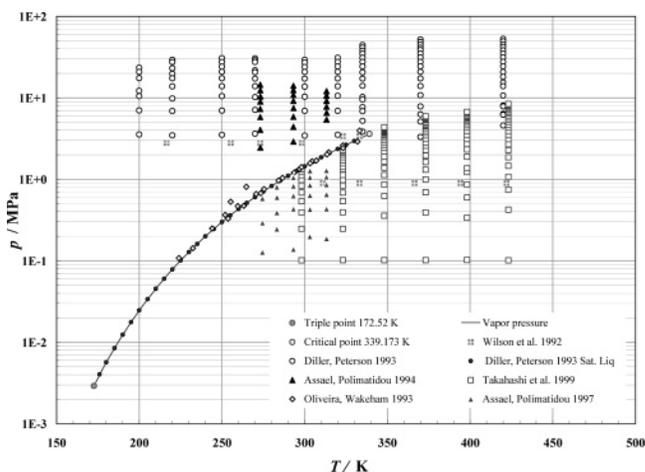
We present a correlation for the viscosity of pentafluoroethane (R125) based on a compilation and critical assessment of the available experimental data. The correlation covers a wide range of fluid states, including the supercritical region. It is applicable from the triple point at 172.52 to 500 K, with pressures varying up to 60 MPa. The formulation includes a zero-density contribution, initial density dependence based on the Rainwater–Friend theory, and a residual contribution for higher densities that combines virial terms with a free-volume term, both being temperature-dependent. The estimated uncertainty of the viscosity correlation (coverage factor of 2) is 3% along the liquid-phase saturation boundary, 3% in the compressed liquid phase at pressures to 60 MPa, and 0.8% in the vapor.

## Introduction

Concern over the negative environmental impact of chlorofluorocarbons (CFCs) has led to the adoption of the Montreal Protocol, an agreement which specifies a schedule for the phase-out of CFCs. This class of fluids has been widely used as refrigerants, solvents, and blowing agents; finding suitable environmentally friendly replacements is therefore an important task. One fluid that has been suggested as a potential replacement fluid is pentafluoroethane (R125). It has been proposed as a component in refrigerant mixtures such as R410A and R407C that are under consideration as replacements for the refrigerant chlorodifluoromethane (R22). Another application of R125, due to its reduction in global warming emissions<sup>1,2</sup> over perfluoro compounds, is for use as a dielectric etchant. In addition, the solvent properties of R125 under supercritical conditions are also under investigation.<sup>3–5</sup> The availability of accurate descriptions of the thermophysical properties of R125 aids the successful development of applications. Equations of state for the thermodynamic properties of R125 have been developed; two have appeared quite recently.<sup>6,7</sup> Transport properties such as thermal conductivity and viscosity are also very important for equipment design. Earlier work by this group<sup>8</sup> presented a corresponding states model for the representation of the transport properties of refrigerants, including R125. More recently, new data and a correlation for the thermal conductivity of R125 have been presented.<sup>9</sup> In this work, we survey the existing literature data for the viscosity of R125 and propose a fluid-specific correlation for the viscosity surface of R125 that is applicable over the entire fluid range, from dilute gas to compressed liquid, including supercritical conditions.

## Experimental Viscosity Data

We surveyed literature data and assembled a compilation from 18 references with viscosity data for R125 covering the gas, liquid, and supercritical fluid regions. Figure 1 illustrates the distribution of the data in the pressure–temperature plane and shows the phase boundary as determined by the equation of state of Lemmon and Jacobsen.<sup>6</sup> Table 1 summarizes the data



**Figure 1.** Distribution of the viscosity data for R125 in the pressure–temperature plane. For clarity, not all data sets for the saturated liquid or vapor are shown.

sets and includes the experimental method, the uncertainty, sample purity, and the temperature and pressure ranges of the experiments. In the following section, we present an assessment of the available data, including both data used in the regression and data used solely for comparison purposes.

## Data Selected for the Correlation

Criteria for primary data sets were the sample purity, the experimental method, the reported experimental uncertainty, and the data analysis. Preference was given to data sets that cover a wide range of pressure and temperature. In the gas phase, the most extensive data set was obtained by Takahashi et al.,<sup>25</sup> who measured the viscosity at sub- and supercritical temperatures from 298.15 to 423.15 K with pressures up to 9 MPa. The instrument was a proven oscillating-disk viscometer with an estimated uncertainty of 0.3%. These results were selected as the primary data set for the vapor and gas region. Other, less expansive gas-phase measurements were carried out by Assael and Polimatiidou<sup>22</sup> in a vibrating-wire instrument with an estimated uncertainty of 1% in the temperature range 273–333 K at pressures up to 1.3 MPa. Wilson et al.<sup>11</sup> measured with a constant-flow-rate capillary viscometer five points in the vapor phase between 311 and 422 K at 0.9 MPa, while Dunlop<sup>17</sup> reported a single point at 298 K and atmospheric pressure with

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**Table 1. Summary of Viscosity Measurements of Pentafluoroethane and Comparisons with the Present Correlation, Equations 1–6<sup>c</sup>**

ref	year	method	uncert %	<i>T</i> (K)	<i>p</i> (MPa)	points	sample purity %	AAD %	BIAS %	RMS %	max dev %
Shankland <sup>10</sup>	1990	SGC	1	253.15–327.55	SL	15	not rep.	32	32	9.3	52
Wilson et al. <sup>11</sup>	1992	CFC	2	216.48–422.04	0.09–3.4	11	99.7	1.9	1.4	1.6	4.8
Bivens et al. <sup>12</sup>	1993	CFC	1.2	253.15–336.15	SL	6	not rep.	11	11	14	34
Diller and Peterson <sup>13</sup>	1993	TVC	3	176.00–420.00	SL-53.1	137	99.7 <sup>a</sup>	1.2	0.17	2.0	15
Oliveira and Wakeham <sup>14</sup>	1993	VW	0.5–1.	223.99–333.19	SL, SV	26	99.9	6.3	2.5	8.2	31
Ripple and Matar <sup>15</sup>	1993	SGC	3–5	250.55–301.95	SL	15	99.7	2.3	2.3	0.84	3.7
Assael and Polimatidou <sup>16</sup>	1994	VW	0.5	273.15–313.15	2.4–14.5	27	99.95 <sup>b</sup>	0.61	0.31	1.0	4.6
Dunlop <sup>17</sup>	1994	CFC	0.3	298.15	0.1	1	99.5	0.17	–0.17	0	–0.17
Geller et al. <sup>18</sup>	1994	CFC	1.2	253–338	SL	corr.	not rep.				
Assael et al. <sup>19</sup>	1995	VW	0.5	273.15–313.15	SL	3	99.95	2.0	2.0	1.9	4.6
Sun et al. <sup>20</sup>	1996	SGC	3	233.15–328.15	SL	20	99.95 <sup>b</sup>	6.9	–1.8	7.9	21
Heide and Schenk <sup>21</sup>	1996	RB	2	223.55–333.15	SL	12	99.9	3.3	0.52	5.0	15
Assael and Polimatidou <sup>22</sup>	1997	VW	1	273.15–313.15	0.1–1.3	29	99.95 <sup>b</sup>	0.82	–0.78	1.3	–6.6
Ripple and Defibaugh <sup>23</sup>	1997	SGC	2.40	255.95–303.07	SL	8	99.9 <sup>a</sup>	0.73	–0.73	0.28	–1.2
Oliveira and Wakeham <sup>24</sup>	1999	VW	2	223.99–331.99	SV	13	99.9	3.3	1.3	3.8	–11
Takahashi et al. <sup>25</sup>	1999	OD	0.3	298.15–423.15	0.1–8.4	131	99.9 <sup>a</sup>	0.30	–0.10	0.37	1.2
Fröba et al. <sup>26, 27</sup>	1999,2000	SLS	6, 4	233.15–333.15	SL	11	99.7	2.9	–1.6	4.3	–14

<sup>a</sup> Mole percent. <sup>b</sup> Mass percent. <sup>c</sup> SL saturated liquid; SV saturated vapor; VW vibrating wire viscometer; CFC const. flow rate capillary viscometer; OD oscillating disk viscometer; RB rolling ball viscometer; SLS surface light scattering; SGC sealed gravitational capillary viscometer; TVC torsionally vibrating crystal viscometer.

a similar instrument. Oliveira and Wakeham<sup>14,24</sup> reported viscosities of R125 vapor near saturation in 1993, measured with a vibrating-wire viscometer with an estimated uncertainty of “0.5 to 1%” in the temperature range 224–332 K. These results were reevaluated in 1999 with more accurate densities. While the revised data are slightly lower than the original values but agree within their estimated uncertainty, the revised viscosity value at the highest temperature of 332 K is 7.4% lower than the original value reported in 1993.

The most comprehensive viscosity measurements of R125 were carried out by Diller and Peterson<sup>13</sup> with two torsional crystal viscometers at NIST. The results cover saturated and compressed liquid states, as well as some vapor states, with pressures up to 53 MPa and over a wide temperature range from 176–420 K. Since a high-accuracy equation of state (EoS) was unavailable at the time of these experiments, an extended corresponding states model was used to obtain densities for the analysis of the experimental data. We have recalculated this data set using densities from the EoS of Lemmon and Jacobsen.<sup>6</sup> This resulted in the viscosities changing by up to almost 4%, with the largest deviations at the highest temperatures. The revised viscosities are lower than the original values and, in some cases, different by as much as 3.5%. We consider these revised values to be the most reliable, with uncertainties on the order of approximately 3%, and have selected them as primary data. This assessment is supported by later measurements in that instrument, which provided insights about its performance at low densities and the possible occurrence of electroviscous effects with conducting samples. Even though one would expect pentafluoroethane to act as a proton donor due to its chemical structure, no elevated conductances were observed in later torsional crystal viscometer measurements of mixtures containing this compound in various compositions.<sup>28</sup> Therefore, while Diller and Peterson did not record sample conductances, it is unlikely that their viscosity results contain electroviscous contributions that are not accounted for in the working theory of the instrument. Such contributions would lead to viscosities that are systematically higher than those measured in instruments where the sample is not exposed to an electric field. The comparison with the data of Ripple and Defibaugh,<sup>23</sup> that were measured in a sealed gravitational capillary viscometer, shows that this is not the case.

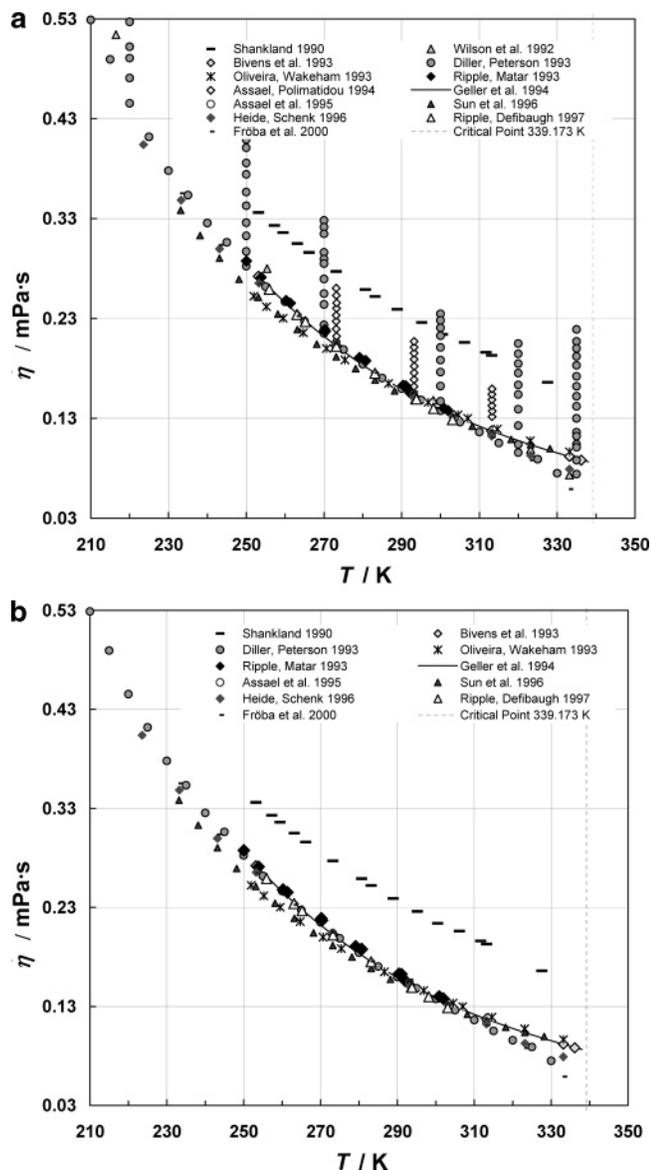
Diller and Peterson measured in their work<sup>13</sup> to lower densities than in their preceding studies with this instrument. It

was later found that resonance scans of the torsionally vibrating crystal in vacuo and at low external damping are subject to inherent difficulties.<sup>29</sup> These consist of (i) considerable ring-down times, which have to be considered after a frequency step before the admittance is measured, and (ii) an increased sensitivity to the drive voltage, which should be as low as possible to reduce the internal damping of the crystal. Diller and Peterson considered neither of these and used the highest drive voltage of 1.1 V that the impedance analyzer delivers. A comparison of their results at the lowest densities on the 370 and 420 K isotherms with the data of Takahashi et al. at 373.15 K and at 423.15 K indicates deviations that are consistent with these difficulties. The viscosities of Diller and Peterson are lower than those of the corresponding isotherms of Takahashi et al., although these are at 3.15 K higher temperatures. On the basis of these considerations, a higher uncertainty has to be assumed for the data of Diller and Peterson at 370 K, at 420 K, and with pressures below 10 MPa. Due to their large uncertainties, these data points were not used in the regression in this paper.

Assael and Polimatidou<sup>16</sup> used a vibrating-wire instrument with reported uncertainties of 0.5% to obtain the viscosity of liquid R125 from 273 to 333 K with pressures from near the saturation boundary to about 17 MPa. Here, this data set was designated primary as well, although the densities used in the data analysis were obtained by extrapolation for pressures above 6.3 MPa; therefore, we consider the uncertainties of the points obtained above 6.3 MPa to be higher than the 0.5% reported by the authors. A final data set included in the primary data set is that of Ripple and Defibaugh<sup>23</sup> who obtained the saturated liquid viscosity of R125 in a sealed capillary viscometer with a straight vertical capillary and reported uncertainties of 2.4%. In their data analysis, Ripple and Defibaugh<sup>23</sup> used experimental densities with an uncertainty of 0.05%.

### Other Literature Data

Since R125 is used primarily as a working fluid in refrigeration cycles, the very first viscosity measurements were reported for the saturated liquid state. The normal boiling point<sup>6</sup> of R125 at 225.06 K requires the use of sealed viscometers to avoid evaporation of the sample liquid at higher temperatures. The first experimental viscosity data were reported by Shankland<sup>10</sup> in 1990 in the temperature range 253.15–327.55 K. Their uncertainty was estimated at 1%. Shankland used the sealed



**Figure 2.** Viscosity data for saturated and compressed liquid R125 as a function of temperature.

gravitational flow viscometer with a coiled capillary that had been used earlier by Phillips and Murphy<sup>30,31</sup> for measurements of chlorofluorocarbon refrigerants. The results of Phillips and Murphy had been found to systematically deviate from those of other researchers, but the cause of the deviations had not been identified. Subsequent measurements of R125 showed similar deviations of the data that Shankland<sup>10</sup> had measured with this coiled capillary viscometer. For instance, the next published viscosity data of Wilson and co-workers in 1992<sup>11</sup> were systematically lower in the liquid phase from 216.48 to 333.15 K. The deviations exceeded the estimated uncertainty of the data of 2% and increased with temperature. At the highest temperature measured by Shankland, 327.55 K, the viscosity was 90% higher than the corresponding value in the results of Wilson et al., Figure 2a. The third published data set was measured from 253.15 to 336.15 K by Bivens et al.<sup>12</sup> in a sealed capillary viscometer where the gravitational flow of the saturated liquid sample was accomplished by a mercury column in contact with the sample. The uncertainty of these data was reported as 1.2%. They agreed with those of Wilson et al.<sup>11</sup> within their combined uncertainties up to 313.15 K but were systematically higher above that temperature. At the highest temperature

(333.15 K) measured by Wilson et al.,<sup>11</sup> the viscosity values of Bivens et al.<sup>12</sup> were 28% higher.

The fourth data set for the viscosity of R125 was the already mentioned contribution by Diller and Peterson<sup>13</sup> in 1993. These results provided further evidence for a systematic error in the data of Shankland and lent additional support to those of Wilson et al. because they were obtained with the torsionally vibrating crystal technique and not with a capillary viscometer, as in the earlier studies. However, the saturated liquid viscosities by Diller and Peterson<sup>13</sup> were systematically lower than the comparable data by Wilson et al.,<sup>11</sup> ranging from  $-7.3\%$  at 216.48 K to  $-7.6\%$  at 330 K, the deviations exceeded the combined uncertainties of the two data sets.

The subsequently published experimental data continued to cluster alongside the measurement results of Diller and Peterson<sup>13</sup> and Wilson et al.<sup>11</sup> They were consistent in that they were about the same order of magnitude lower than the viscosities reported by Shankland.<sup>10</sup> Ripple and Defibaugh<sup>23</sup> were not aware of the data of Shankland<sup>10</sup> but suggested neglect of vapor buoyancy and the curvature of the capillary as possible causes for the deviations of the R152a viscosity data of Phillips and Murphy. Laesecke et al.<sup>32</sup> demonstrated for the case of R134a that ref 10 had indeed neglected corrections of the raw experimental data to account for the vapor buoyancy and for the radial acceleration in a sealed gravitational viscometer with a coiled capillary. At the highest temperature where R134a was measured by Shankland, these two corrections lowered the data by 14% and 17%, respectively. Shankland's viscosity data for R134a could be brought into agreement with other experimental results when these two corrections were applied.

While the systematic deviation of Shankland's data could be rationalized, the smaller but significant inconsistencies among the remaining viscosity data for liquid R125 are unresolved. Taking the saturated liquid data of Diller and Peterson as a baseline, it can be seen in Figure 2b that the results of Oliveira and Wakeham<sup>14</sup> and those of Sun et al.<sup>20</sup> are systematically lower below a temperature of approximately 293 K. At 233.15 K, the deviation is about 5%. Above 293 K, these data as well as those of Bivens et al.<sup>12</sup> are systematically higher. The deviation of the data of Bivens et al.<sup>12</sup> may be due to the lack of a correction for the increasing compressibility of the test liquid when the temperature approaches the critical point. Bivens et al.<sup>12</sup> evidently did not use such a correction, which had been worked out by van den Berg et al.<sup>33</sup> A similar trend is observed in the deviations of the data of Heide and Schenk.<sup>21</sup> The positive deviations of the data by Ripple and Matar,<sup>15</sup> slightly increasing with temperature, may appear to be negligible when considering the combined uncertainties of their data and the data of Diller and Peterson.<sup>13</sup> Nevertheless, these deviations are systematic and may be due to the insufficient correction for the radial acceleration of the test liquid in the coiled capillary of Ripple and Matar's capillary viscometer. To avoid this correction, Ripple and Defibaugh<sup>23</sup> remeasured R125 in a viscometer with a straight vertical capillary, and these data agree with those of Diller and Peterson<sup>13</sup> and were therefore included in the data set used to develop the correlation. Fröba et al.<sup>26,27</sup> reported data sets for the viscosity of saturated liquid R125 that were obtained with the surface light scattering technique. The data in the first report<sup>26</sup> are identical to those in the later journal article;<sup>27</sup> however in the more recent work, they revised the estimate for the maximum uncertainty of the kinematic viscosity from 6% to 4%. A comparison with the results of Diller and Peterson revealed deviations of a systematic nature that exceeded these uncertainty estimates up to a maximum of  $-12\%$ .

Therefore, the results of Fröba et al.<sup>26,27</sup> were not considered in the regression of the correlation.

### Viscosity Correlation

The viscosity  $\eta$  of a pure fluid is considered as a sum of three contributions,<sup>34</sup>

$$\eta(\rho, T) = \eta^\circ(T)[1 + B_\eta(T)\rho] + \Delta\eta_h(\rho, T) + \Delta\eta_c(\rho, T) \quad (1)$$

The term  $\eta^\circ(T)$  represents the viscosity in the limit of zero density,  $B_\eta(T)$  is the second virial coefficient for viscosity based on the Rainwater–Friend theory,<sup>35</sup>  $\Delta\eta_h(\rho, T)$  is the residual contribution that represents the higher-order density terms as a function of the absolute temperature  $T$  and density  $\rho$ , and the term  $\Delta\eta_c(\rho, T)$  represents the critical enhancement of the viscosity. In this work, we set the critical enhancement term to zero, since it is significant only in the immediate vicinity of the gas–liquid critical point,<sup>36</sup> where the viscosity of R125 has not been measured. For reasons of theory, eq 1 is expressed in terms of density and temperature. Thus, densities have to be associated to experimental pressures and temperatures. In this work, the fundamental equation of state (EoS) of Lemmon and Jacobsen<sup>6</sup> was used for all density calculations. This EoS is valid from the triple point<sup>37</sup> at 172.52 K to 500 K with pressures up to 60 MPa.

### Zero-Density Limit

The temperature-dependent viscosity of a fluid in the zero-density limit,  $\eta^\circ(T)$ , cannot be measured directly. Generally, it is necessary to extrapolate values from the available experimental data at low densities to zero density, which can entail significant errors. Kinetic theory of gases<sup>38</sup> gives for spherical particles the relationship

$$\eta^\circ(T) = 5\sqrt{mkT/\pi}/(16\sigma_0^2\Omega^{(2,2)*}(T)) \quad (2)$$

where  $k = 1.380\,650\,5 \times 10^{-23} \text{ J}\cdot\text{K}^{-1}$  is the Boltzmann constant,<sup>39</sup>  $m = M/N_A$  is the molecular mass with  $M$  being the molar mass and  $N_A = 6.022\,141\,5 \times 10^{23} \text{ mol}^{-1}$  being Avogadro's constant,<sup>39</sup> the collision diameter  $\sigma_0$  is inserted in nanometers, and the viscosity has units of  $\mu\text{Pa}\cdot\text{s}$ . The collision diameter  $\sigma_0$  is defined as the separation distance where the intermolecular potential function is equal to zero, and  $\Omega^{(2,2)*}(T)$  is a collision integral that depends on the potential function. If sufficient experimental data are available, one may use an empirical relationship for the collision integral similar to that developed for the reference correlation for the viscosity of propane.<sup>34</sup> Alternatively, one may assume that a particular intermolecular potential function, such as the Lennard–Jones (for nonpolar fluids) or the Stockmayer (for polar fluids), applies. This will be discussed in more detail later.

### Initial Density Dependence

At very low densities, the density dependence of the viscosity is initially linear, and the temperature variation is represented by the second viscosity virial coefficient  $B_\eta(T)$ . Rainwater and Friend<sup>35,40</sup> calculated the second viscosity virial coefficient of the Lennard–Jones potential theoretically. For this two-parameter force field model,  $B_\eta(T)$  is obtained from its dimensionless form according to

$$B_\eta(T) = N_A\sigma^3 B_\eta^*(T^*) \quad (3)$$

**Table 2. Parameters for the Correlation of the Second Viscosity Virial Coefficient,<sup>34</sup> Equation 4**

$i$	$b_i$	$t_i$
0	−19.572 881	0
1	219.739 99	−0.25
2	−1015.322 6	−0.50
3	2471.012 5	−0.75
4	−3375.171 7	−1.00
5	2491.659 7	−1.25
6	−787.260 86	−1.50
7	14.085 455	−2.50
8	−0.346 641 58	−5.50

where  $T^* = kT/\epsilon$  is the dimensionless temperature and  $\epsilon/k$  is an energy scaling parameter in kelvin. The results of Rainwater and Friend for  $B_\eta^*(T^*)$  were later adjusted by Bich and Vogel<sup>41</sup> for better agreement with experimental data and revised values were tabulated in the range  $0.5 \leq T^* \leq 100$ . We use the correlation that was developed by Laesecke as a component of the reference correlation for the viscosity of propane by Vogel et al.<sup>34</sup>

$$B_\eta^*(T^*) = \sum_{i=0}^8 b_i (T^*)^{t_i} \quad (4)$$

with parameters  $b_i$  and the exponents  $t_i$  from ref 34 that are given in Table 2. Equation 4 may be safely extrapolated to temperatures as low as  $T^* \sim 0.3$ , which corresponds to a point well below the triple point of R125.

### Residual Contribution

As mentioned before, the terms  $\Delta\eta_h(\rho, T)$  for higher density of eq 1 are formulated in terms of the reduced density  $\delta = \rho/\rho_c$  and the reduced temperature  $\tau = T/T_c$  because these independent variables are suggested by theory. After systematic consideration of a variety of functional forms, the final correlation contains the following combination of polynomial terms and a Batschinski–Hildebrand free-volume term:

$$\Delta\eta_h(\rho, T) = 1000 \left( \sum_{j=2}^3 \sum_{k=1}^2 \alpha_{jk} \frac{\delta^j}{\tau^k} + c_1 \delta \left( \frac{1}{\delta_0 - \delta} - \frac{1}{\delta_0} \right) \right) \quad (5)$$

In this equation, the term  $\Delta\eta_h(\rho, T)$  is in  $\mu\text{Pa}\cdot\text{s}$ , the individual terms are constrained to be zero at  $\rho = 0$ , and its leading-order density dependence is of higher order than linear. The free-volume term is one used successfully for other fluids.<sup>34,42–44</sup> A term, linear-in-density, arising from a Taylor-series expansion of the free-volume term about zero density, is subtracted, since the linear-in-density term has already been accounted for in the second viscosity virial coefficient term discussed earlier. No linear-in-density polynomial terms were permitted for this reason as well. The temperature dependence of the reduced close-packed density  $\delta_0(\tau)$  is written as

$$\delta_0 = c_2 + c_3\sqrt{\tau} \quad (6)$$

A total of eight adjustable parameters are present in the dense-fluid contributions in eqs 5–6. These parameters are determined by fitting the experimental data, and the final fit may contain fewer parameters depending upon the statistical significance of the coefficients obtained from regression.

### Results and Discussion

Data in the primary data set at densities less than  $0.1 \text{ mol}\cdot\text{L}^{-1}$  were used in the regression to determine parameters for the

**Table 3. Parameters for the Viscosity Correlation**

coefficient	value	std dev
$\sigma_0$ (nm)	0.5235	$\pm 6.163 \times 10^{-4}$
$\epsilon_0/k$ (K)	237.077	$\pm 1.295$
$\alpha_{21}$	0	
$\alpha_{22}$	$5.677\,448 \times 10^{-3}$	$\pm 5.2 \times 10^{-4}$
$\alpha_{31}$	$-5.096\,662 \times 10^{-3}$	$\pm 1.3 \times 10^{-4}$
$\alpha_{32}$	0	
$c_1$	$1.412\,564 \times 10^{-1}$	$\pm 7.0 \times 10^{-3}$
$c_2$	3.033 797	$\pm 3.9 \times 10^{-2}$
$c_3$	$2.992\,464 \times 10^{-1}$	$\pm 6.6 \times 10^{-2}$

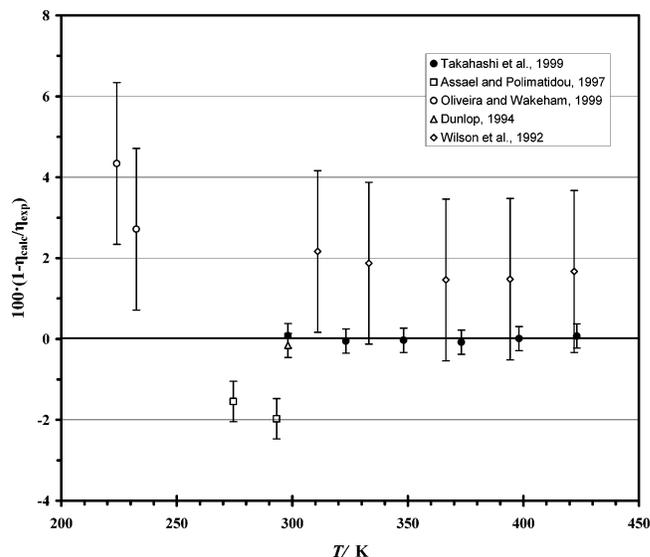
dilute gas contribution to the viscosity of R125. Since R125 is a polar molecule, with a gas-phase dipole moment of  $1.563 \pm 0.005$  D,<sup>45</sup> the interactions among such particles are approximated more realistically by the Stockmayer potential function, which is a superposition of the often-used spherical nonpolar Lennard–Jones (6-12) force field with the angle-dependent interaction of two point dipoles. The collision integral  $\Omega^{(2,2)*}(T)$  of the Stockmayer potential was calculated by Monchick and Mason,<sup>46</sup> and a correlation of their results was developed in our study of the viscosity of methanol.<sup>47</sup> This correlation contains three parameters: a collision diameter  $\sigma_0$ , an energy-well depth  $\epsilon_0$ , and the reduced dipole moment  $\delta_D$ . However, the three parameters are related by the expression<sup>46</sup>

$$\sigma_0^3 = 3.6220\mu^2/(\delta_D\epsilon_0/k) \quad (7)$$

where  $\mu$  is the dipole moment in debye, resulting in only two adjustable parameters to be determined by fitting data. We fit the data of Takahashi et al.<sup>25</sup> and obtained  $\sigma_0 = 0.5230$  nm,  $\epsilon_0/k = 233.539$  K, and from eq 7 found  $\delta_D = 0.2649$  with an average absolute deviation between the data and the correlation of 0.052%. The linear density term in eq 1 was evaluated with the expression in eq 4, which should be viewed as an approximation since eq 4 is based on the results of Rainwater and Friend for  $B_{ij}^*(T^*)$  of the Lennard–Jones potential. Theory has not been advanced to  $B_{ij}^*(T^*)$  of the Stockmayer potential nor to any other force fields. For comparison, we also applied the Lennard–Jones 6-12 potential, using the correlation for  $\Omega^{(2,2)*}$  developed by Neufeld et al.,<sup>48</sup>

$$\Omega_{LJ}^{(2,2)*} = 1.16145/T^{*0.14874} + 0.52487e^{-0.77320T^*} + 2.16178e^{-2.4378T^*} \quad (8)$$

and obtained  $\sigma_0 = 0.5235$  nm and  $\epsilon_0/k = 237.077$  K, with an average absolute deviation of 0.054%. The zero-density viscosities calculated with these two parameter sets are virtually indistinguishable; the viscosity over the temperature range 170–500 K is differing by less than 0.01%. The data set is limited, containing only six points; therefore, since the Lennard–Jones expression is the simpler of the two methods and we have theory for the linear density regime for this potential, we selected the LJ representation for the zero-density region. Table 3 summarizes the coefficients obtained from the fit using eq 8 and the Lennard–Jones 6-12 potential function, along with the standard deviation of the coefficients. Figure 3 shows the deviations (here defined as  $[100(1 - \eta_{\text{calc}}/\eta_{\text{exp}})]$ ) between the calculated and experimental values of viscosity for all data at densities less than  $0.1 \text{ mol}\cdot\text{L}^{-1}$  and the uncertainty associated with the measurements. The data used in the regression are shown with filled symbols, while those used only for comparison are shown with open symbols. The data of Takahashi et al.<sup>25</sup> are represented to well within their reported uncertainty of 0.3%. The single data point of Dunlop<sup>17</sup> was not used in the fit but agrees well with the data of Takahashi et al.<sup>25</sup> and is represented



**Figure 3.** Deviations of the viscosity data from the present correlation in the dilute gas region.

to within its reported uncertainty of 0.3%. However, the other data sets of Assael and Polimaitidou,<sup>22</sup> Oliveira and Wakeham,<sup>24</sup> and Wilson et al.<sup>11</sup> are not represented to within their reported uncertainties, nor do they agree with each other to within their reported uncertainties. Unfortunately, this problem is often seen when examining experimental data for viscosity.

The primary data were used to obtain the coefficients of the high-density contribution (eqs 5 and 6), which are presented in Table 3. The primary data were weighted equally, and the regression was performed with the statistical package ODRPACK.<sup>49</sup> Initially, all polynomial terms in eq 5 were included in the regression, but some were later discarded due to lack of statistical significance, resulting in a total of five parameters in the final representation of the residual viscosity. Figure 4a shows the percent deviations of the primary data from the present correlation as a function of density. Figure 4b shows the percent deviations of the primary data from the extended corresponding states model developed earlier.<sup>8</sup> Significant improvement is shown, especially for the data of Takahashi et al.<sup>25</sup> in the density range  $1\text{--}4 \text{ mol}\cdot\text{L}^{-1}$  and for the compressed liquid data of Diller and Peterson.<sup>13</sup> Table 1 presents a tabular summary of the results of comparisons of the correlation with available experimental data, using the following definitions for average absolute deviation (AAD), bias, and root-mean-square (RMS) deviation:

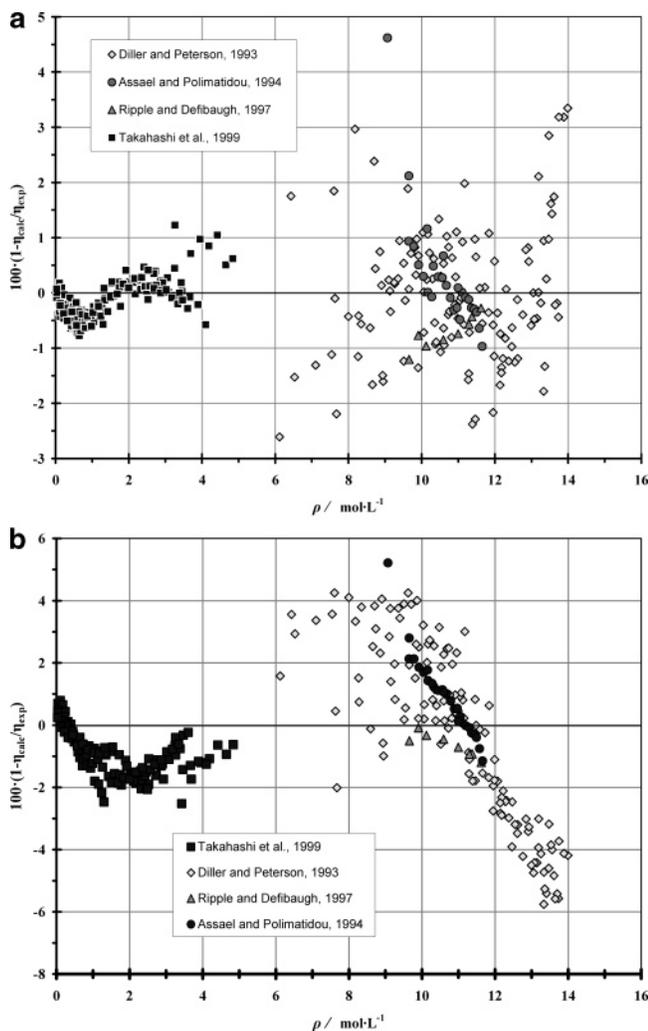
$$\text{AAD} = \frac{100}{n} \sum_{i=1}^n \left| 1 - \frac{\eta_i^{\text{calc}}}{\eta_i^{\text{exp}}} \right| \quad (9)$$

$$\text{BIAS} = \frac{100}{n} \sum_{i=1}^n \left( 1 - \frac{\eta_i^{\text{calc}}}{\eta_i^{\text{exp}}} \right) \quad (10)$$

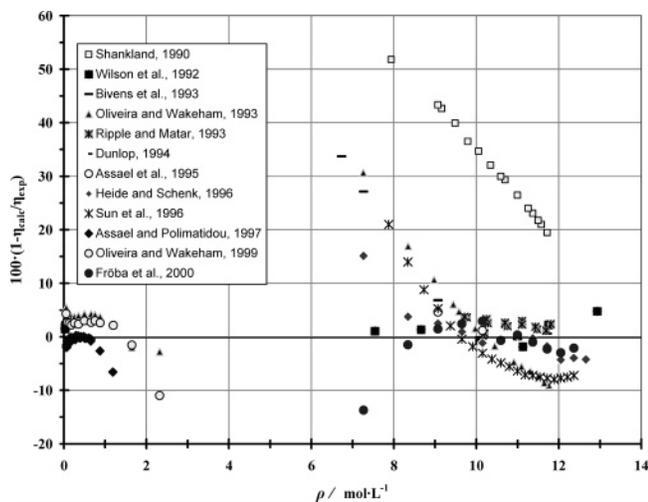
and

$$\text{RMS}^2 = \frac{100}{n} \left( \sum_{i=1}^n \left( 1 - \frac{\eta_i^{\text{calc}}}{\eta_i^{\text{exp}}} \right)^2 \right) - \text{BIAS}^2 \quad (11)$$

The gas phase and supercritical data of Takahashi et al.<sup>25</sup> are represented very well, with an AAD of 0.3%, while the saturated and compressed liquid data of Diller and Peterson<sup>13</sup> have an AAD of 1.2%. The other primary data sets are also represented



**Figure 4.** (a) Deviations of the primary viscosity data from the present correlation as a function of density. (b) Deviations of the primary viscosity data from the extended corresponding states model<sup>8</sup> as a function of density.



**Figure 5.** Deviations of the secondary viscosity data from the present correlation as a function of density.

well; the saturated liquid data of Ripple and Defibaugh<sup>23</sup> and the liquid-phase data of Assael and Polimatidou<sup>16</sup> have AADs of less than 1%. Figure 5 shows deviations between the correlation and the data not used in the development of the correlation (the secondary viscosity data). The data of Ripple and Matar<sup>15</sup> display an AAD of 2.3% which is within their

reported uncertainty of 3–5%. The vapor-phase measurements of Assael and Polimatidou<sup>22</sup> have a reported uncertainty of 1%, and the deviations exceed this value both at their lowest and their highest densities; however, the representation from 0.1 to 0.6 kg·m<sup>-3</sup> is within the reported uncertainty. Except for their lowest temperature (216 K) point, the liquid-phase data of Wilson et al.<sup>11</sup> also agree well with the correlation, to within 3%. The other secondary sets display much larger deviations, the largest being those of Shankland.<sup>10</sup> As discussed earlier, the data of Shankland<sup>10</sup> do not include corrections for both vapor buoyancy and radial acceleration and display very large positive deviations from the correlation that decrease in magnitude as the density increases. The 1993 data of Oliveira and Wakeham<sup>14</sup> and the data of Sun et al.,<sup>20</sup> although obtained from different types of instruments, display a similar deviation pattern. The saturated liquid data of Bivens et al.,<sup>12</sup> at higher temperatures, also display a similar deviation pattern; as discussed earlier, some of this may be attributed to the lack of a correction for the increasing compressibility of the liquid as the temperature approaches the critical point. Finally, test points for validating computer calculations with the new correlation are  $T = 300$  K,  $\rho = 10.596\,999\,8$  mol·L<sup>-1</sup>,  $\eta = 177.37$   $\mu\text{Pa}\cdot\text{s}$  (corresponds to  $p = 10$  MPa) and  $T = 400$  K,  $\rho = 0.030\,631$  mol·L<sup>-1</sup>,  $\eta = 17.070$   $\mu\text{Pa}\cdot\text{s}$  (corresponds to  $p = 0.101\,325$  MPa).

## Conclusion

On the basis of experimental data for the viscosity of pentafluoroethane (R125), a correlation has been developed for this property that is valid from the triple point<sup>37</sup> at 172.52 K to 500 K with pressures up to 60 MPa. Data comparisons support an estimated uncertainty of 3% along the liquid-phase saturation boundary. In addition, we estimate the uncertainty in the liquid phase at pressures to 60 MPa to also be 3% at a coverage factor of 2. In the gas region, the correlation represents the data to within 0.8%, which includes a coverage factor of 2. The new fluid-specific correlation presents an improvement over an earlier, general model based on extended corresponding states<sup>8</sup> and provides a simple-to-use correlation for engineering applications.

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