

# The Viscosity of Carbon Dioxide, Methane, and Sulfur Hexafluoride in the Limit of Zero Density

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The paper contains accurate representations for the viscosity of the three polyatomic gases, carbon dioxide, methane, and sulfur hexafluoride, in the limit of zero density. These gases were studied because they possess permanent multipole moments of increasing order 4, 6, and 8, respectively. The correlations have associated uncertainties of  $\pm 0.3\%$  around room temperature rising to  $\pm 1.5\%$  at the low-temperature extreme and to a maximum of  $\pm 2.0\%$  at the high-temperature extreme. The correlating equation for carbon dioxide is valid for the temperature range 200–1500 K, that for methane from 110–1050 K and that for sulfur hexafluoride from 220–900 K. It is shown that a two-parameter law of corresponding states is inadequate for the representation of the data over these wide ranges of temperature.

Key words: carbon dioxide; corresponding states; methane; sulfur hexafluoride; transport properties; viscosity of gases.

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## 1. Introduction

The starting point for a correlation of the transport properties of fluids over a wide range of thermodynamic states must always be a description of the behavior of the properties in the limit of zero density. In this paper we extend our earlier studies of the viscosity of nitrogen and oxygen in this limit<sup>1</sup> to carbon dioxide, methane, and sulfur hexafluoride.

The most recent critical reviews and correlations of the viscosity of carbon dioxide and methane in the limit of zero density were published ten or more years ago,<sup>2-5</sup> and for sulfur hexafluoride no correlations are available in the literature. Since these publications, several new measurements of the transport properties of these systems have been made and are generally viewed with a higher level of confidence than the earlier measurements. In this paper we examine anew the viscosity of carbon dioxide, methane, and sulfur hexafluoride at the limit of zero density and over a wide range of temperatures. The aim has been to produce a correlation of a critically evaluated set of experimental data which is both accurate and simple.

For the monatomic gases simplicity follows directly from the near conformality of the spherically symmetric pair interactions among the various species. This is because such conformality implies a two-parameter law of corresponding states for the properties.<sup>6</sup> Although recent evidence<sup>7,8</sup> shows that the most accurate correlation requires modification of the simple two-parameter law, the changes are minor. For polyatomic gases the lack of spherical symmetry in the pair potential means that a two-parameter law of corresponding states cannot exist as a matter of principle. Nevertheless, over a limited range of temperature such a two-parameter correlation has proved accurate in practice, although there is evidence of its breakdown at extreme temperatures. In view of the usefulness of such a correlation for the reliable extrapolation of experimental data, we have used our high accuracy correlations for carbon dioxide, methane, and sulfur hexafluoride to investigate the scheme further.

## 2. Experimental Data

The methodology used in the formulation of the correlations reported here is similar to that used in an earlier publication<sup>1</sup> and we confine ourselves to an outline here.

The kinetic theory of monatomic gases clearly defines the relation of all of the bulk thermophysical and molecular properties, so that independent tests of consistency and accuracy can be performed. However, the theory for polyatomic gases has not been developed to this level of sophistication,<sup>7</sup> and the confirmation of the consistency of different measured thermophysical and molecular properties via theory is therefore generally impossible. As a result the quality

of any representation of the viscosity of these systems rests solely on the choice of the experimental data used in their formulation. In turn, the choice of experimental data must be based upon an assessment of the technique used in the measurements and the proven precision attained. To this end it has previously been found convenient to divide the experimental data into two categories.<sup>1,9</sup>

### (i) Primary data

These are the results of measurements made with a primary experimental apparatus of high precision, i.e., an instrument for which a complete working equation exists and for which a detailed knowledge of all corrections is available. Further, all principal variables should be measurable to a high degree of precision and the published work should include some demonstration of the precision of the measured data.

### (ii) Secondary data

These are the results of measurements which are of inferior accuracy to primary data. This inferior accuracy may arise from operation at extreme conditions or from incomplete characterization of the apparatus.

For each of the systems studied here, we have carried out a survey of the available literature data and assigned them to one of the above categories. In order to fulfill the desire that the final correlations be of high accuracy, we have only used data designated as primary in their formulation. A significance is attached to each datum determined by our estimate of the uncertainty. In the following sections the data selected as primary for each system are detailed, together with our estimate of their uncertainty, a summary of which is included in Table 1.

In most cases the viscosity data we employ have been acquired through measurements at, or below, atmospheric pressure. However, a few authors report measurements as a function of density at higher pressures. In order to employ values appropriate to the zero-density limit we have, in the latter case, made use of the results of the original statistical analyses of the data as a function of density. In the former case, the low-pressure data have been adjusted to the zero-density limit by means of a small correction based on the same analyses. In no case does the correction amount to more than  $\pm 0.2\%$ , which is usually within the uncertainty of the experimental data. In any event any error in the magnitude of the correction contributes insignificantly to the estimated uncertainty of the zero-density viscosity.

### 2.1. Carbon Dioxide

The most accurate measurements of the viscosity of carbon dioxide have been carried out with oscillating disk viscometers at Brown University<sup>13-25</sup> and at Tohoku University.<sup>26</sup> The measurements at Brown have been made with

Table 1. Primary experimental data for the viscosity of carbon dioxide, methane and sulphur hexafluoride near the zero-density limit

| Reference                            | Technique | Temp. Range (K) | Estimated Uncertainty (%) |
|--------------------------------------|-----------|-----------------|---------------------------|
| Carbon Dioxide                       |           |                 |                           |
| Harris et al. <sup>10</sup>          | C         | 203- 310        | ± 1.5 - ± 1.0             |
| Maitland and Smith <sup>11</sup>     | C         | 292-1497        | ± 1.0 - ± 2.0             |
| Bailey <sup>12</sup>                 | C         | 344- 719        | ± 1.0 - ± 2.0             |
| Kestin et al. <sup>13</sup>          | OD        | 298- 973        | ± 0.2 - ± 0.5             |
| Kestin et al. <sup>14</sup>          | OD        | 298- 773        | ± 0.2 - ± 0.4             |
| Kestin et al. <sup>15</sup>          | OD        | 296- 673        | ± 0.2 - ± 0.4             |
| Kestin et al. <sup>16</sup>          | OD        | 305             | ± 0.2                     |
| DiPippo and Kestin <sup>17</sup>     | OD        | 296- 775        | ± 0.2 - ± 0.4             |
| Kestin and Leidenfrost <sup>18</sup> | OD        | 293             | ± 0.2                     |
| Breestveld et al. <sup>19</sup>      | OD        | 293- 303        | ± 0.2                     |
| Kestin et al. <sup>20</sup>          | OD        | 296- 303        | ± 0.2                     |
| Kestin et al. <sup>21</sup>          | OD        | 296- 477        | ± 0.2 - ± 0.3             |
| Abe et al. <sup>22</sup>             | OD        | 298- 468        | ± 0.2 - ± 0.3             |
| DiPippo et al. <sup>23</sup>         | OD        | 293- 303        | ± 0.2                     |
| Kestin et al. <sup>24</sup>          | OD        | 293- 304        | ± 0.2                     |
| Kestin and Yata <sup>25</sup>        | OD        | 293- 304        | ± 0.2                     |
| Iwasaki and Takahashi <sup>26</sup>  | OD        | 298- 323        | ± 0.2                     |
| Methane                              |           |                 |                           |
| Clarke and Smith <sup>36</sup>       | C         | 114- 374        | ± 1.5 - ± 1.0             |
| Maitland and Smith <sup>37</sup>     | C         | 295-1022        | ± 1.0 - ± 2.0             |
| Dave et al. <sup>38</sup>            | C         | 294-1051        | ± 1.0 - ± 2.0             |
| Hellemans et al. <sup>39</sup>       | OD        | 298- 468        | ± 0.2 - ± 0.3             |
| Kestin and Leidenfrost <sup>42</sup> | OD        | 294             | ± 0.2                     |
| Kestin et al. <sup>20</sup>          | OD        | 296- 302        | ± 0.2                     |
| Kestin and Yata <sup>25</sup>        | OD        | 293- 303        | ± 0.2                     |
| Kestin et al. <sup>14</sup>          | OD        | 298- 473        | ± 0.2 - ± 0.3             |
| Abe et al. <sup>40</sup>             | OD        | 298- 468        | ± 0.2 - ± 0.3             |
| Kestin et al. <sup>15</sup>          | OD        | 296- 474        | ± 0.2 - ± 0.3             |
| Kestin et al. <sup>21</sup>          | OD        | 296- 477        | ± 0.2 - ± 0.3             |
| Kestin et al. <sup>41</sup>          | OD        | 296- 473        | ± 0.2 - ± 0.3             |
| Sulphur Hexafluoride                 |           |                 |                           |
| Harris et al. <sup>10</sup>          | C         | 218- 302        | ± 1.5 - ± 1.0             |
| Dave et al. <sup>38</sup>            | C         | 293- 873        | ± 1.0                     |
| Hellemans et al. <sup>39</sup>       | OD        | 298- 573        | ± 0.2 - ± 0.3             |
| Kestin et al. <sup>21</sup>          | OD        | 296- 477        | ± 0.2 - ± 0.3             |
| Kestin et al. <sup>14</sup>          | OD        | 298- 473        | ± 0.2 - ± 0.3             |
| Kestin et al. <sup>15</sup>          | OD        | 296- 474        | ± 0.2 - ± 0.3             |
| Kestin et al. <sup>20</sup>          | OD        | 296- 303        | ± 0.2 - ± 0.3             |
| Kestin et al. <sup>22</sup>          | OD        | 423- 468        | ± 0.3                     |

C Capillary viscometer  
OD Oscillating disc viscometer

two different instruments and the reproducibility of the instruments confirmed by independent measurements carried out over a number of years. These measurements extend over the temperature range 290-970 K with an accuracy of  $\pm 0.2\%$  at ambient temperature deteriorating to  $\pm 0.5\%$  at the highest temperature. The work of Iwasaki and Takahashi<sup>26</sup> has an accuracy of  $\pm 0.3\%$  but only extends over the range 298-323 K.

The remaining primary data are the results of measurements made with capillary viscometers, which are generally of inferior accuracy to those from oscillating disk viscometers. However, the results of Smith and his collaborators<sup>10,11</sup> and Bailey<sup>12</sup> enjoy a high level of confidence. In addition, because these results extend beyond the temperatures covered by the oscillating disk instruments they extend the range of the correlation substantially. The results of Smith and his group<sup>10,11</sup> are estimated to have an uncertainty of  $\pm 1.0\%$  at ambient temperatures, rising to  $\pm 1.5\%$  and  $\pm 2.0\%$  at the low- and high-temperature extremes, respectively. For the work of Bailey<sup>12</sup> it is estimated that the uncertainty of the data is  $\pm 1.0\%$  at the lowest temperature deteriorating to  $\pm 2.0\%$  at the highest temperature. For the reasons described in Sec. 3, of the 17 sets of data designated as primary, only six were used in the formulation of the correlation, and these are shown in Table 2. All of the primary data sets resulting from capillary viscometer measurements have been included in the formulation because of the large temperature range over which they extend. Of the results of

Table 2. Selected primary data for the viscosity employed in the correlation

| Reference                        | Technique | Temp. Range (K) | Estimated Uncertainty (%) | Relative Weight (w') |
|----------------------------------|-----------|-----------------|---------------------------|----------------------|
| Carbon Dioxide                   |           |                 |                           |                      |
| Harris et al. <sup>10</sup>      | C         | 203- 310        | ± 1.5 - ± 1.0             | 1.7 - 4              |
| Maitland and Smith <sup>11</sup> | C         | 292-1497        | ± 1.0 - ± 2.0             | 4 - 1                |
| Bailey <sup>12</sup>             | C         | 344- 719        | ± 1.0 - ± 2.0             | 4.1 - 1              |
| Kestin et al. <sup>13</sup>      | OD        | 298- 973        | ± 0.2 - ± 0.5             | 100 - 16.1           |
| Kestin et al. <sup>14</sup>      | OD        | 298- 773        | ± 0.2 - ± 0.4             | 100 - 30             |
| Kestin et al. <sup>15</sup>      | OD        | 296- 673        | ± 0.2 - ± 0.4             | 100 - 30             |
| Methane                          |           |                 |                           |                      |
| Clarke and Smith <sup>36</sup>   | C         | 114- 374        | ± 1.5 - ± 1.0             | 1.6 - 3.7            |
| Maitland and Smith <sup>37</sup> | C         | 295-1022        | ± 1.0 - ± 2.0             | 3.7 - 1              |
| Dave et al. <sup>38</sup>        | C         | 294-1051        | ± 1.0 - ± 2.0             | 3.7 - 1              |
| Kestin et al. <sup>14</sup>      | OD        | 298- 473        | ± 0.2 - ± 0.3             | 92 - 48              |
| Abe et al. <sup>40</sup>         | OD        | 298- 468        | ± 0.2 - ± 0.3             | 92 - 48              |
| Kestin et al. <sup>15</sup>      | OD        | 296- 474        | ± 0.2 - ± 0.3             | 92 - 48              |
| Kestin et al. <sup>41</sup>      | OD        | 296- 473        | ± 0.2 - ± 0.3             | 92 - 48              |
| Sulphur Hexafluoride             |           |                 |                           |                      |
| Harris et al. <sup>10</sup>      | C         | 218- 302        | ± 1.5 - ± 1.0             | 1 - 2.3              |
| Dave et al. <sup>38</sup>        | C         | 293- 873        | ± 1.0                     | 2.3                  |
| Hellemans et al. <sup>39</sup>   | OD        | 298- 573        | ± 0.2 - ± 0.3             | 56.3 - 29            |
| Kestin et al. <sup>14</sup>      | OD        | 296- 477        | ± 0.2 - ± 0.3             | 56.3 - 29            |
| Kestin et al. <sup>15</sup>      | OD        | 296- 473        | ± 0.2 - ± 0.3             | 56.3 - 29            |
| Kestin et al. <sup>15</sup>      | OD        | 296- 474        | ± 0.2 - ± 0.3             | 56.3 - 29            |

C Capillary viscometer  
OD Oscillating disc viscometer

measurements with oscillating disk viscometers, only those three sets were used that covered the largest temperature range.

Among the data for carbon dioxide categorized as secondary,<sup>27-34</sup> those measurements performed in early oscillating disk instruments are so classified owing to a lack of a complete working equation. The data of Kestin and Whitelaw<sup>30</sup> are classified as secondary due to failure of a thermal compensator mechanism in the instrument used for these measurements. The early capillary viscometer measurements<sup>31-34</sup> have been included in the category because of the unidentified systematic errors with which they are known to be burdened.<sup>35</sup>

## 2.2. Methane

The possibility of pyrolysis of pure methane at high temperatures has meant that the range of temperature over which its thermophysical properties have been measured is somewhat limited compared with carbon dioxide. Only data from the research centers at Brown University<sup>14,15,18,20,21,39-42</sup> using an oscillating disk viscometer and Oxford University<sup>36-38</sup> using a capillary viscometer are sufficiently accurate to be included in the primary data set. The data from Brown extend over the temperature range 290-480 K with an estimated uncertainty of  $\pm 0.2$  to  $\pm 0.4\%$ . The data of Smith and his collaborators extend the overall temperature range of 110-1050 K and the estimated uncertainty is  $\pm 1.0\%$  at ambient temperatures rising to  $\pm 1.5$  and  $\pm 2.0\%$  at low and high temperatures, respectively.

Only seven of the 12 primary data sets were used in the formulation of the primary correlation for methane, and these are shown in Table 2. The three sets of results measured with a capillary viscometer were included once again because of their large temperature ranges. Of the measurements made with an oscillating disk viscometer, five sets cover the approximate range 298-475 K with the other sets covering very limited temperature ranges about room temperature. Four of the five larger temperature sets were used

in the formulation of the correlation. The designation of data as secondary<sup>28,31,33,43-49</sup> for methane follows because of essentially the same reasoning as adduced for carbon dioxide, with the results of Gough *et al.*<sup>49</sup> classified as secondary because they are smoothed data.

### 2.3. Sulfur Hexafluoride

The relatively low temperature at which sulfur hexafluoride undergoes decomposition and the comparatively high temperature at which it boils reduces the accessible temperature range for this gas. For the same reasons as for methane, only data from two research centers are of sufficient accuracy to be included in the primary data set. The data from Brown<sup>14,15,20-22,39</sup> cover the temperature range 295–575 K with respective uncertainties in the range  $\pm 0.2$ – $\pm 0.4\%$ . The data from Oxford<sup>10,38</sup> cover the temperature range 215–875 K with uncertainties ranging from  $\pm 1.0\%$  at room temperature and above to  $\pm 1.5\%$  at low temperatures.

Of the eight primary data sets shown in Table 1, only six sets were used in the formulation of the primary correlation and they are listed in Table 2. The data obtained by Raw and Ellis,<sup>50</sup> and McCoubrey and Singh,<sup>51</sup> are assigned to the secondary category because they are burdened with the systematic errors typical of early capillary viscometer results.<sup>35,52</sup>

## 3. Methodology

For the representation of the viscosity data we have employed the kinetic theory relationship<sup>6</sup>

$$\eta = \frac{5}{16} \frac{(mkT/\pi)^{1/2}}{\sigma^2 \Omega_{\eta}^*(T^*)} \quad (1)$$

in which  $m$  is the molecular mass,  $T$  the absolute temperature, and  $k$  the Boltzmann's constant. The symbol  $\sigma$  represents a length scaling parameter, and  $\Omega_{\eta}^*$  is a functional of the pair potential for the interaction between the molecules of the gas. For spherically symmetric pair potentials between structureless particles, where the potential may be expressed in a universal reduced form

$$\frac{U(r/\sigma)}{\epsilon} = U^*(r^*), \quad (2)$$

it can readily be shown that the functional  $\Omega_{\eta}^*$  is a function only of the reduced temperature  $T^* = kT/\epsilon$  and that is universal among the same interactions. Thus a two-parameter law of corresponding states exists for the viscosity which is characterized by the values of the scaling parameters  $\epsilon$  and  $\sigma$  for each interaction. This result has been used to good effect by Kestin and his collaborators in their treatment of the zero-density properties of the noble gases.<sup>6-8</sup> However, it has been necessary to admit that the intermolecular pair potentials among these species are not exactly conformal in order to describe the experimental data with high accuracy.<sup>8</sup> For polyatomic gases interacting through nonspherically symmetric potentials,  $\Omega_{\eta}^*$  depends not only upon the intermolecular potential which is, in any event, nonspherically symmetric, but also upon the internal energy states of the molecules.<sup>7</sup> An exact two-parameter law of corresponding

states for the viscosity of such gases cannot, therefore, exist in principle. Nevertheless, Eq. (1) and the ideas of a corresponding states treatment remain a useful means of correlating the experimental data. Accordingly, all primary data pairs  $(T, \eta)$  have been reduced to the dimensionless values  $(T^*, \Omega_{\eta}^*)$  using values of  $\epsilon/k$  and  $\sigma$  determined in the manner described in a later section. The resulting body of data has been fitted to an equation of the form

$$\ln \Omega_{\eta}^* = \sum_{i=0}^4 a_i (\ln T^*)^i, \quad (3)$$

which has proved useful in earlier correlations.<sup>1,6,8</sup> Table 3 lists the selected values of  $\epsilon/k$  and  $\sigma$  together with the optimum values of the coefficients  $a_i$  for each gas.

In the fitting procedure each viscosity datum has been assigned a temperature-dependent statistical weight  $w_i$ . The weight itself is determined from our estimate of the uncertainty in the viscosity  $\Delta\eta$ , according to the equation

$$w_i = (3\eta/\Delta\eta)^2, \quad (4)$$

which assumes that the estimated precision is approximately three times the standard deviation of the data. The relative magnitudes of the weights  $w$  for the various sets of data used in the fit are included in Table 2.

If the weighting procedure described above is applied to all of the selected primary data a new difficulty emerges which must be considered. The experimental viscosity data of highest accuracy have most often been acquired near ambient temperatures. Such data have a high weight, not only because of their accuracy, but also because of the large number of them. This has the effect of biasing the fit to represent what is essentially a single datum while the remaining data are relatively poorly represented. An example of this effect

Table 3. Parameters and coefficients of the viscosity correlation

|                 | $\epsilon/k$<br>(K) | $\sigma$<br>(nanometres) | $a_i$ ( $i = 0, 4$ ) |          |
|-----------------|---------------------|--------------------------|----------------------|----------|
| CO <sub>2</sub> | 251.2               | 0.3751                   | $a_0$                | 0.45885  |
|                 |                     |                          | $a_1$                | -0.49676 |
|                 |                     |                          | $10a_3$              | 0.23436  |
|                 |                     |                          | $a_3$                | 0.10309  |
| CH <sub>4</sub> | 163.6               | 0.3709                   | $10a_4$              | -0.33775 |
|                 |                     |                          | $a_0$                | 0.45009  |
|                 |                     |                          | $a_1$                | -0.46460 |
|                 |                     |                          | $100a_2$             | -0.63653 |
| SF <sub>6</sub> | 204.5               | 0.5263                   | $a_3$                | 0.10925  |
|                 |                     |                          | $10a_4$              | -0.32954 |
|                 |                     |                          | $a_0$                | 0.42386  |
|                 |                     |                          | $a_1$                | -0.38220 |
|                 |                     |                          | $10a_2$              | -0.56997 |
|                 |                     |                          | $10a_3$              | 0.79345  |
|                 |                     |                          | $10a_4$              | -0.11127 |

can be seen in Fig. 1, which contains a plot of the deviations of the carbon dioxide primary data from a weighted fit to the entire set of primary data. The deviations clearly have a systematic character, beyond experimental uncertainty at high and low temperatures, that is unacceptable. To overcome this problem the data in the set used to construct the final correlation have been built up sequentially.

A fit of Eqs. (1) and (3) is first performed to a subset of the primary data spanning the entire temperature range. Subsequently, further data subsets of successively smaller extent are added and, after each addition, a new fit performed and the pattern of the deviations from it examined. As soon as the pattern begins to reveal systematic trends in the deviations of the data at the extreme temperatures, owing to the addition of too many data at ambient temperature, the process is terminated. The set of data built up in this fashion, which is used to produce the final correlation, usually then excludes some ambient-temperature data originally included in the primary category. It has been found that this procedure circumvents entirely the difficulty noted above.

#### 4. Primary Correlation

In this section we consider the correlation of the viscosity of the individual gases with the aid of Eqs. (1) and (3) and the parameters and coefficients of Table 3. Table 3 also includes the temperature range over which the correlations are valid.

##### 4.1. Carbon Dioxide

Figure 2 contains a plot of the deviations of the primary experimental viscosity data for carbon dioxide from the optimum correlation. The standard deviation of the fit is

$\pm 0.005 \mu\text{Pa s}$ , whereas the maximum deviation amounts to  $\pm 1.2\%$ . The estimates of the uncertainty given earlier and the deviation plot contained in Fig. 2 suggest that the uncertainty to be assigned to the correlated viscosity depends upon the temperature range considered. The solid line in Fig. 2 delineates the estimated accuracy over the entire temperature range.

Figure 3 contains a comparison of the present correlation for the viscosity of carbon dioxide with earlier correlations,<sup>2-5</sup> whereas Fig. 4 compares the present correlation with the secondary experimental data not included in its development as well as the primary data excluded from its formulation. The discrepancies between the various correlations reflect the benefit derived from the inclusion of new data in the present treatment and the "buildup" procedure used here, described in Sec. 3.

##### 4.2. Methane

The viscosity of methane is correlated by Eqs. (1) and (3), together with the parameters in Table 3. The deviations of the primary data from this representation are shown in Fig. 5. Again, the solid lines in Fig. 5 delineate the estimated uncertainty over the entire temperature range. The maximum deviation of the primary experimental data from the correlation amounts to  $\pm 1.0\%$  with a standard deviation of  $\pm 0.003 \mu\text{Pa s}$ .

Figure 6 contains a comparison of the present correlation with earlier correlations,<sup>2,4</sup> and Fig. 7 compares the secondary data and unused primary data with the present correlation, with the exception of the data of Rakshit *et al.*<sup>48</sup> as they are 10% lower than the present correlation. All earlier correlations are systematically higher, particularly in the low-temperature region. This reflects the inclusion of addi-

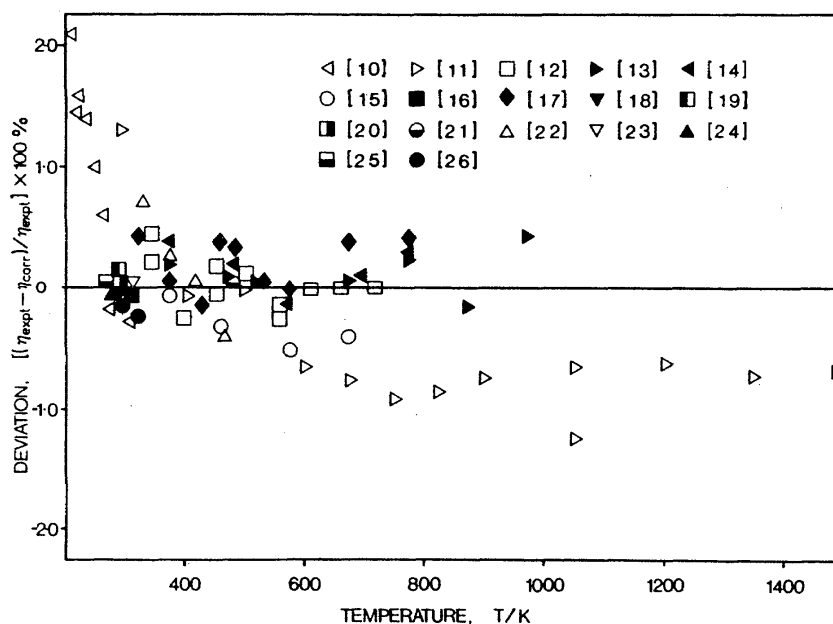


FIG. 1. Bias of the correlation of the viscosity of carbon dioxide resulting from the inclusion of an excessive number of data near ambient temperatures.

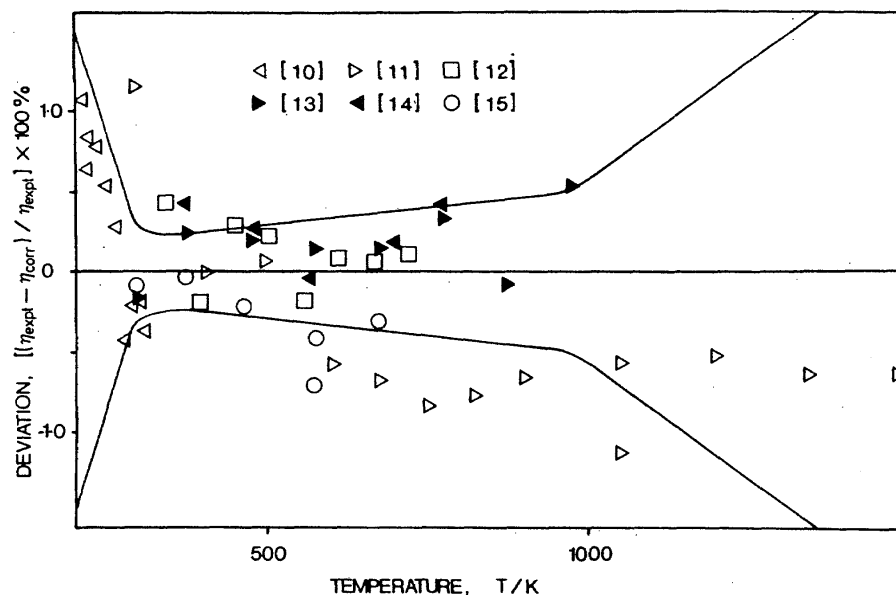


FIG. 2. Deviations of the primary experimental data for the viscosity of carbon dioxide from the correlation of Eqs. (1) and (3).

tional high-temperature data and the methodology used here, so that the present correlation is to be preferred.

#### 4.3. Sulfur Hexafluoride

The viscosity of sulfur hexafluoride is represented by Eqs. (1) and (3), together with the parameters in Table 3. The standard deviation from the fit is one of  $\pm 0.003 \mu\text{Pa s}$  and the maximum deviation is  $\pm 0.8\%$ . Figure 8 contains a plot of the deviation of the primary experimental data from the optimum correlation. The solid line delineates the estimated accuracy over the entire temperature range. Figure 9 shows a comparison of the secondary experimental data with the optimum correlation.

#### 5. Corresponding States

If a law of corresponding states characterized by two parameters were valid among the gases studied here, it follows that the collision integral  $\Omega_7^*$  would be a universal function<sup>6</sup> of the reduced temperature  $T^*$ . A suitable basis for a test of this proposition is provided by the functional  $\Omega^{(2,2)*}(T^*)$  found to be universal among the monatomic gases over an intermediate range of temperature.<sup>8</sup> For definiteness we have, in fact, selected the functional characteristic of argon over the entire temperature range. The parameters  $\epsilon/k$  and  $\sigma$  which yield the optimum representation of the primary, experimental viscosity data listed in Table 2 have been determined and these are the values quoted in Table 3.

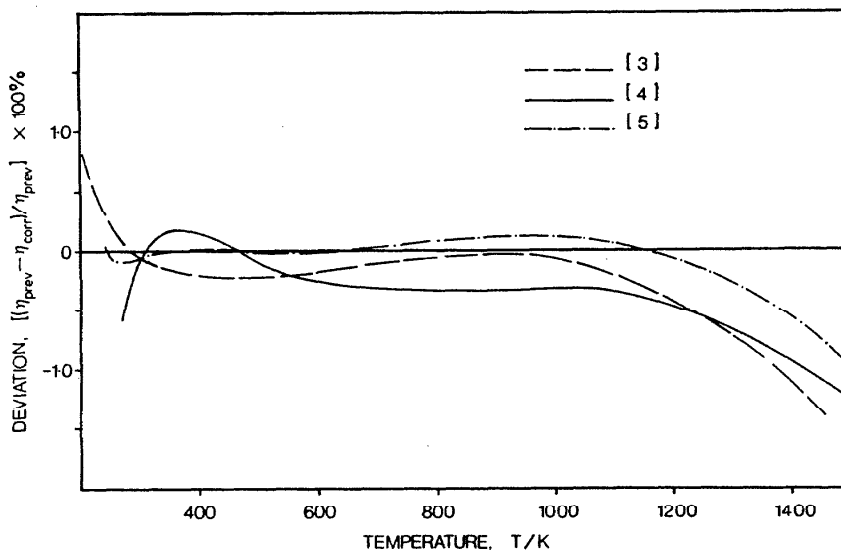


FIG. 3. A comparison of the present correlation for the viscosity of carbon dioxide with that of earlier work.

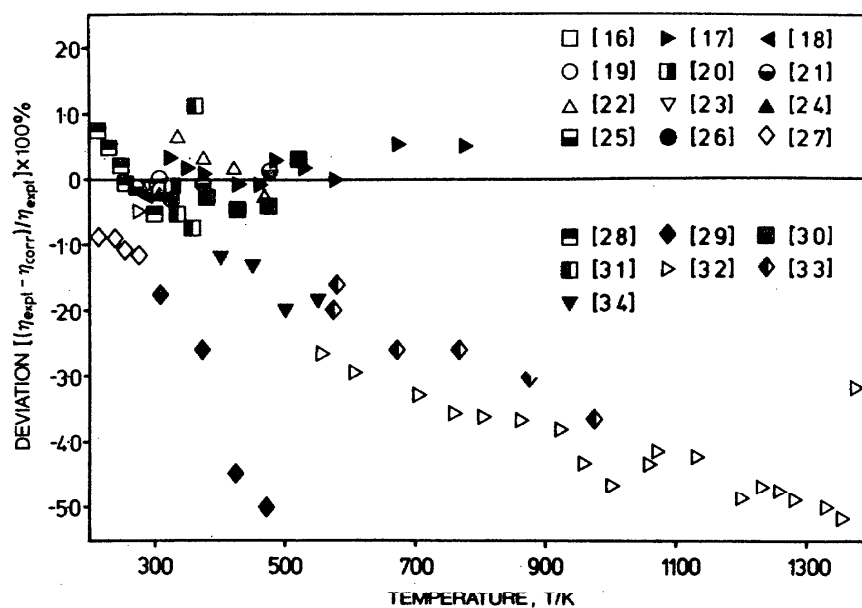


FIG. 4. Deviations of the secondary and excluded primary experimental data for the viscosity of carbon dioxide from the correlation of Eqs. (1) and (3).

Figure 10 shows the deviations of the present individual correlations of the gases  $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{SF}_6$  from the universal correlation. The deviations are shown as continuous curves for clarity. It is clear that there is a systematic trend in the deviations from the argon correlation which amounts to as much as 6% at the lowest temperatures. Furthermore, the data for the three gases deviate substantially from each other. Thus a universal representation of the viscosity of these three gases cannot secure even a moderately good fit to the

experimental data, particularly at low temperatures. These discrepancies are borne out more clearly by the plot of the quantity  $(T^*)^{1/3} \Omega_\eta^*$  as a function of  $T^*$ . The group is a constant for the inverse sixth power potential characteristic of the long-range pair potential of the monatomic gases.<sup>7</sup> Such a plot is shown in Fig. 11 for argon, xenon, and the three gases studied here. In the intermediate temperature range the curves for all five gases are quite similar. Because it is in just this range that the most accurate results from the

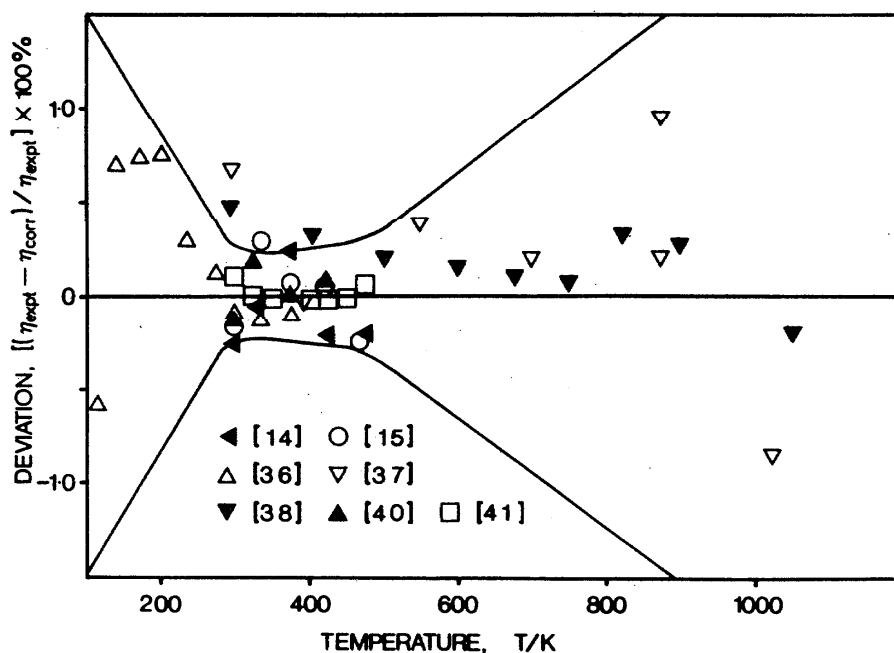


FIG. 5. Deviations of the primary experimental data for the viscosity of methane from the correlation of Eqs. (1) and (3).

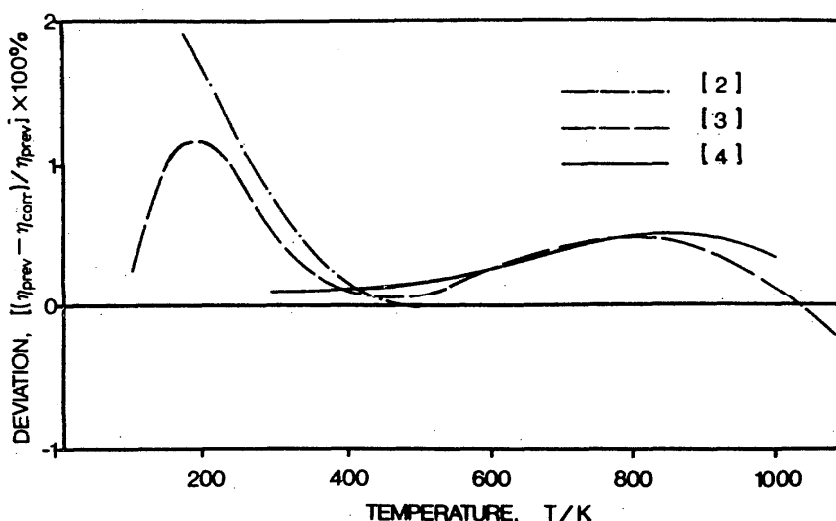


FIG. 6. A comparison of the present correlation for the viscosity of methane with that of earlier work.

oscillating disk viscometers lie, it is not surprising that the two-parameter corresponding states correlation should be observed for them. However, over a wider range of temperatures, particularly at low temperatures, significant departures from correspondence can be discerned between the three polyatomic gases and between these gases and the monatomic systems. The differences are much larger than are observed between the monatomic gases themselves. This result means that the two-parameter law of corresponding states cannot be used to extend the range of the correlations given here in a reliable fashion.

In view of the considerable advantages of the principle of corresponding states, the introduction of a third parameter in order to retain these would seem a natural step. Recent model calculations<sup>53</sup> show that some of the behavior reported in this section can be reproduced by the addition of multipole moments to a spherically symmetric pair poten-

tial. This suggests that the third parameter of a corresponding treatment might be a suitably reduced multipole moment. On the other hand, such a treatment would neglect any repulsive anisotropy in the potential which other studies reveal may be equally significant.<sup>54</sup>

## 6. Tabulations

The correlations described in the preceding sections have been employed to generate a table of recommended viscosities for carbon dioxide over the temperature range 200–1500 K, for methane over the temperature range 110–1050 K, and for sulfur hexafluoride over the temperature range 220–900 K. The calculated values are listed in Table 4. The tabulated values are subject to the uncertainties ascribed to the corresponding correlations, thus Table 4 should be read in conjunction with Figs. 2, 5, and 8.

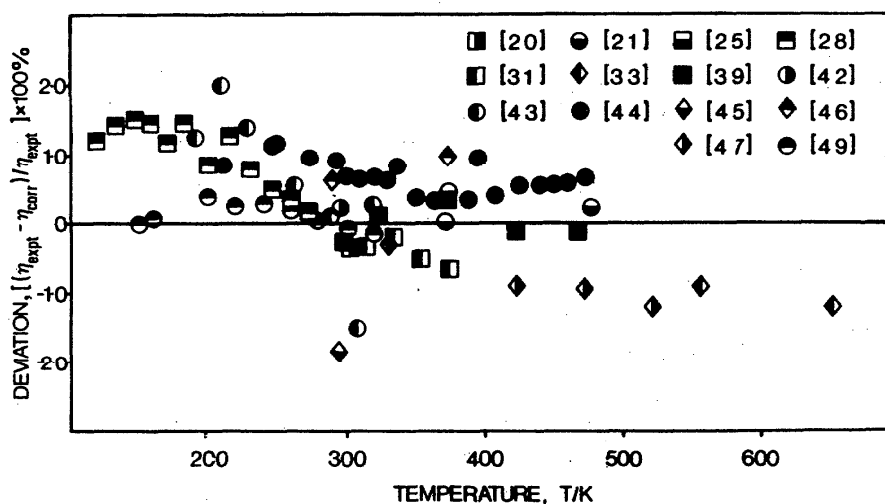


FIG. 7. Deviations of the secondary and excluded primary experimental data for the viscosity of methane from the correlation of Eqs. (1) and (3).



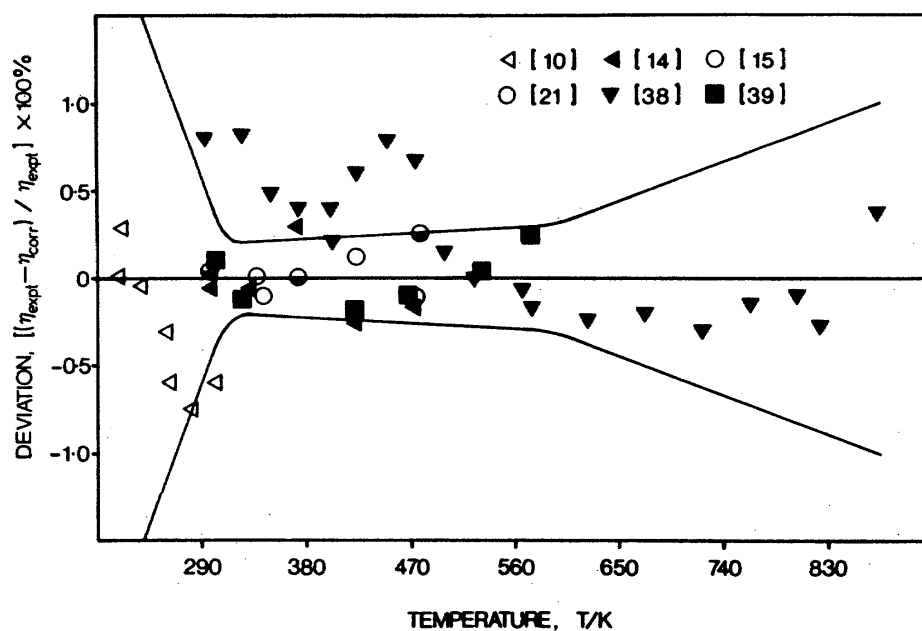


FIG. 8. Deviations of the primary experimental data for the viscosity of sulfur hexafluoride from the correlation of Eqs. (1) and (3).

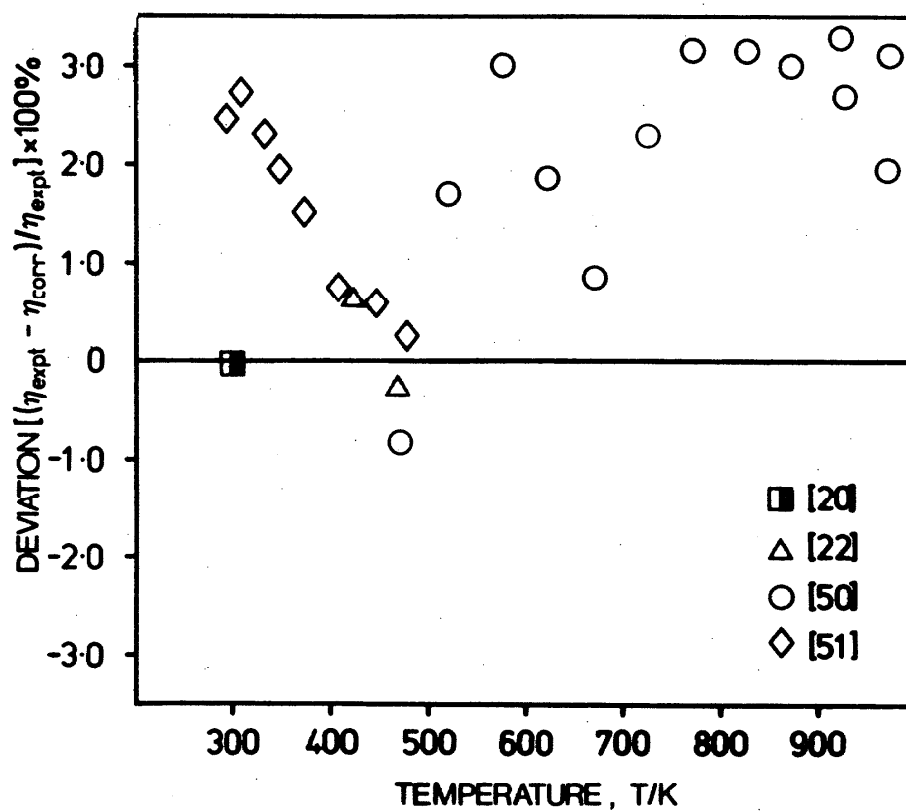


FIG. 9. Deviations of the secondary and excluded primary experimental data for the viscosity of sulfur hexafluoride from the correlation of Eqs. (1) and (3).

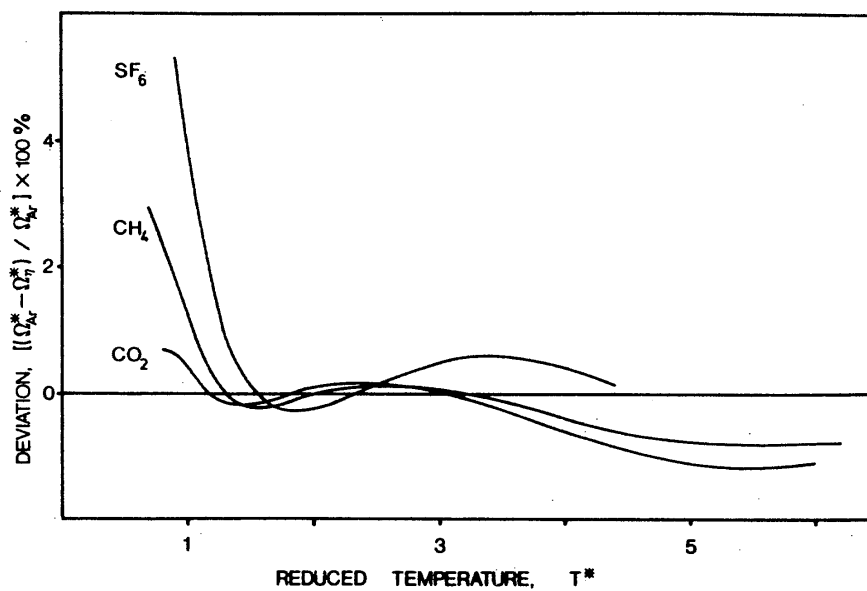


FIG. 10. Deviations of the present correlations of the functional  $\Omega_7^*$  for carbon dioxide, methane, and sulfur hexafluoride from that for  $\Omega^{(2,2)*}$  of argon ( $\Omega_{Ar}^*$ ).

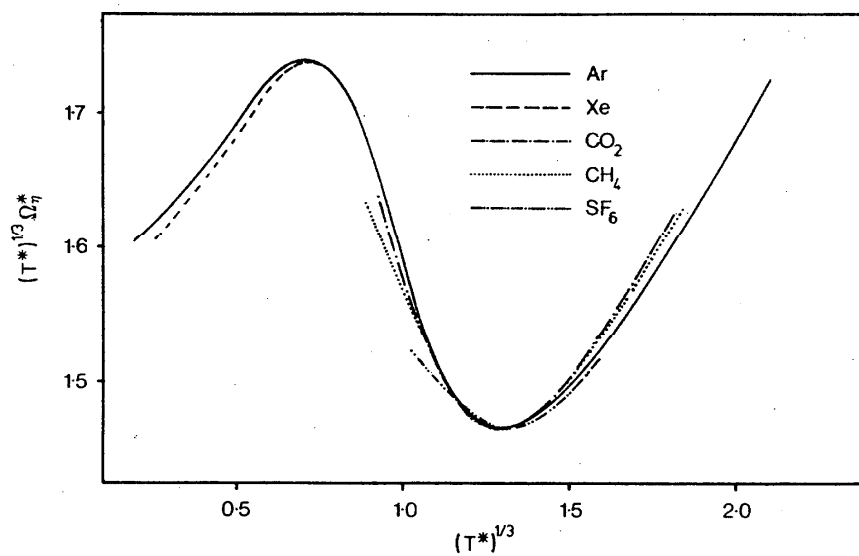


FIG. 11. A plot of  $(T^*)^{1/3} \Omega_7^*$  for argon, xenon, carbon dioxide, methane, and sulfur hexafluoride.

Table 4. The viscosity of carbon dioxide, methane, and sulfur hexafluoride—continued.

| Temperature |                 | Viscosity ( $\mu\text{Pa s}$ ) |                 |       | Temperature     |                 | Viscosity ( $\mu\text{Pa s}$ ) |  |  |
|-------------|-----------------|--------------------------------|-----------------|-------|-----------------|-----------------|--------------------------------|--|--|
| T (K)       | CO <sub>2</sub> | CH <sub>4</sub>                | SF <sub>6</sub> | T (K) | CO <sub>2</sub> | CH <sub>4</sub> | SF <sub>6</sub>                |  |  |
| 110.        |                 | 4.36                           |                 |       |                 |                 |                                |  |  |
| 115.        |                 | 4.54                           |                 | 310.  | 15.52           | 11.51           | 15.81                          |  |  |
| 120.        |                 | 4.72                           |                 | 315.  | 15.76           | 11.67           | 16.03                          |  |  |
| 125.        |                 | 4.90                           |                 | 320.  | 16.01           | 11.83           | 16.26                          |  |  |
| 130.        |                 | 5.09                           |                 | 325.  | 16.25           | 11.99           | 16.48                          |  |  |
| 135.        |                 | 5.27                           |                 | 330.  | 16.49           | 12.15           | 16.71                          |  |  |
| 140.        |                 | 5.46                           |                 | 335.  | 16.73           | 12.30           | 16.93                          |  |  |
| 145.        |                 | 5.64                           |                 | 340.  | 16.97           | 12.46           | 17.16                          |  |  |
| 150.        |                 | 5.83                           |                 | 345.  | 17.21           | 12.61           | 17.38                          |  |  |
| 155.        |                 | 6.02                           |                 | 350.  | 17.44           | 12.77           | 17.60                          |  |  |
| 160.        |                 | 6.20                           |                 | 355.  | 17.68           | 12.92           | 17.82                          |  |  |
| 165.        |                 | 6.39                           |                 | 360.  | 17.92           | 13.07           | 18.04                          |  |  |
| 170.        |                 | 6.58                           |                 | 365.  | 18.15           | 13.23           | 18.26                          |  |  |
| 175.        |                 | 6.76                           |                 | 370.  | 18.39           | 13.38           | 18.48                          |  |  |
| 180.        |                 | 6.95                           |                 | 375.  | 18.62           | 13.52           | 18.70                          |  |  |
| 185.        |                 | 7.14                           |                 | 380.  | 18.85           | 13.67           | 18.92                          |  |  |
| 190.        |                 | 7.32                           |                 | 385.  | 19.08           | 13.82           | 19.14                          |  |  |
| 195.        |                 | 7.51                           |                 | 390.  | 19.31           | 13.97           | 19.35                          |  |  |
| 200.        | 10.05           | 7.69                           |                 | 395.  | 19.54           | 14.11           | 19.57                          |  |  |
| 205.        | 10.30           | 7.87                           |                 | 400.  | 19.77           | 14.26           | 19.78                          |  |  |
| 210.        | 10.55           | 8.06                           |                 | 405.  | 20.00           | 14.40           | 19.99                          |  |  |
| 215.        | 10.80           | 8.24                           |                 | 410.  | 20.22           | 14.54           | 20.21                          |  |  |
| 220.        | 11.05           | 8.42                           | 11.63           | 415.  | 20.45           | 14.68           | 20.42                          |  |  |
| 225.        | 11.30           | 8.60                           | 11.87           | 420.  | 20.67           | 14.83           | 20.63                          |  |  |
| 230.        | 11.55           | 8.78                           | 12.10           | 425.  | 20.90           | 14.96           | 20.84                          |  |  |
| 235.        | 11.80           | 8.96                           | 12.33           | 430.  | 21.12           | 15.10           | 21.05                          |  |  |
| 240.        | 12.05           | 9.13                           | 12.57           | 435.  | 21.34           | 15.24           | 21.26                          |  |  |
| 245.        | 12.30           | 9.31                           | 12.80           | 440.  | 21.56           | 15.38           | 21.47                          |  |  |
| 250.        | 12.55           | 9.48                           | 13.04           | 445.  | 21.78           | 15.52           | 21.67                          |  |  |
| 255.        | 12.80           | 9.66                           | 13.27           | 450.  | 22.00           | 15.65           | 21.88                          |  |  |
| 260.        | 13.05           | 9.83                           | 13.50           | 455.  | 22.22           | 15.79           | 22.08                          |  |  |
| 265.        | 13.30           | 10.00                          | 13.74           | 460.  | 22.43           | 15.92           | 22.29                          |  |  |
| 270.        | 13.55           | 10.18                          | 13.97           | 465.  | 22.65           | 16.05           | 22.49                          |  |  |
| 275.        | 13.80           | 10.35                          | 14.20           | 470.  | 22.86           | 16.19           | 22.69                          |  |  |
| 280.        | 14.04           | 10.51                          | 14.43           | 475.  | 23.08           | 16.32           | 22.90                          |  |  |
| 285.        | 14.29           | 10.68                          | 14.66           | 480.  | 23.29           | 16.45           | 23.10                          |  |  |
| 290.        | 14.54           | 10.85                          | 14.89           | 485.  | 23.50           | 16.58           | 23.30                          |  |  |
| 295.        | 14.78           | 11.01                          | 15.12           | 490.  | 23.71           | 16.71           | 23.50                          |  |  |
| 300.        | 15.03           | 11.18                          | 15.35           | 495.  | 23.92           | 16.84           | 23.70                          |  |  |
| 305.        | 15.28           | 11.34                          | 15.58           | 500.  | 24.13           | 16.97           | 23.89                          |  |  |

Table 4. The viscosity of carbon dioxide, methane, and sulfur hexafluoride —continued

| Temperature |                 | Viscosity ( $\mu\text{Pa s}$ ) |                 |       | Temperature     |                 | Viscosity ( $\mu\text{Pa s}$ ) |  |
|-------------|-----------------|--------------------------------|-----------------|-------|-----------------|-----------------|--------------------------------|--|
| T (K)       | CO <sub>2</sub> | CH <sub>4</sub>                | SF <sub>6</sub> | T (K) | CO <sub>2</sub> | CH <sub>4</sub> |                                |  |
| 510.        | 24.54           | 17.22                          | 24.29           | 1010. | 41.46           | 27.84           |                                |  |
| 520.        | 24.95           | 17.47                          | 24.68           | 1020. | 41.75           | 28.03           |                                |  |
| 530.        | 25.36           | 17.72                          | 25.06           | 1030. | 42.03           | 28.22           |                                |  |
| 540.        | 25.76           | 17.97                          | 25.44           | 1040. | 42.32           | 28.42           |                                |  |
| 550.        | 26.16           | 18.21                          | 25.82           | 1050. | 42.60           | 28.61           |                                |  |
| 560.        | 26.56           | 18.45                          | 26.20           | 1060. | 42.88           |                 |                                |  |
| 570.        | 26.95           | 18.69                          | 26.57           | 1070. | 43.16           |                 |                                |  |
| 580.        | 27.34           | 18.93                          | 26.94           | 1080. | 43.43           |                 |                                |  |
| 590.        | 27.72           | 19.17                          | 27.31           | 1090. | 43.71           |                 |                                |  |
| 600.        | 28.10           | 19.40                          | 27.67           | 1100. | 43.99           |                 |                                |  |
| 610.        | 28.48           | 19.63                          | 28.03           | 1110. | 44.26           |                 |                                |  |
| 620.        | 29.85           | 19.86                          | 28.39           | 1120. | 44.54           |                 |                                |  |
| 630.        | 29.23           | 20.08                          | 28.75           | 1130. | 44.81           |                 |                                |  |
| 640.        | 29.59           | 20.31                          | 29.10           | 1140. | 45.08           |                 |                                |  |
| 650.        | 29.96           | 20.53                          | 29.44           | 1150. | 45.35           |                 |                                |  |
| 660.        | 30.32           | 20.76                          | 29.79           | 1160. | 45.62           |                 |                                |  |
| 670.        | 30.68           | 20.98                          | 30.13           | 1170. | 45.89           |                 |                                |  |
| 680.        | 31.04           | 21.19                          | 30.47           | 1180. | 46.16           |                 |                                |  |
| 690.        | 31.39           | 21.41                          | 30.81           | 1190. | 46.42           |                 |                                |  |
| 700.        | 31.74           | 21.63                          | 31.14           | 1200. | 46.69           |                 |                                |  |
| 710.        | 32.09           | 21.84                          | 31.47           | 1210. | 46.96           |                 |                                |  |
| 720.        | 32.43           | 22.05                          | 31.80           | 1220. | 47.22           |                 |                                |  |
| 730.        | 32.77           | 22.26                          | 32.12           | 1230. | 47.49           |                 |                                |  |
| 740.        | 33.11           | 22.48                          | 32.44           | 1240. | 47.75           |                 |                                |  |
| 750.        | 33.45           | 22.68                          | 32.76           | 1250. | 48.01           |                 |                                |  |
| 760.        | 33.78           | 22.89                          | 33.08           | 1260. | 48.27           |                 |                                |  |
| 770.        | 34.12           | 23.10                          | 33.39           | 1270. | 48.54           |                 |                                |  |
| 780.        | 34.45           | 23.30                          | 33.70           | 1280. | 48.80           |                 |                                |  |
| 790.        | 34.77           | 23.51                          | 34.01           | 1290. | 49.06           |                 |                                |  |
| 800.        | 35.10           | 23.71                          | 34.31           | 1300. | 49.32           |                 |                                |  |
| 810.        | 35.42           | 23.92                          | 34.61           | 1310. | 49.58           |                 |                                |  |
| 820.        | 35.74           | 24.12                          | 34.91           | 1320. | 49.84           |                 |                                |  |
| 830.        | 36.06           | 24.32                          | 35.21           | 1330. | 50.09           |                 |                                |  |
| 840.        | 36.37           | 24.52                          | 35.50           | 1340. | 50.35           |                 |                                |  |
| 850.        | 36.69           | 24.72                          | 35.80           | 1350. | 50.61           |                 |                                |  |
| 860.        | 37.00           | 24.92                          | 36.09           | 1360. | 50.87           |                 |                                |  |
| 870.        | 37.31           | 25.12                          | 36.37           | 1370. | 51.12           |                 |                                |  |
| 880.        | 37.62           | 25.31                          | 36.66           | 1380. | 51.38           |                 |                                |  |
| 890.        | 37.92           | 25.51                          | 36.94           | 1390. | 51.63           |                 |                                |  |
| 900.        | 38.23           | 25.71                          | 37.22           | 1400. | 51.89           |                 |                                |  |
| 910.        | 38.53           | 25.90                          |                 | 1410. | 52.14           |                 |                                |  |
| 920.        | 38.83           | 26.10                          |                 | 1420. | 52.40           |                 |                                |  |
| 930.        | 39.13           | 26.29                          |                 | 1430. | 52.65           |                 |                                |  |
| 940.        | 39.43           | 26.49                          |                 | 1440. | 52.91           |                 |                                |  |
| 950.        | 39.72           | 26.68                          |                 | 1450. | 53.16           |                 |                                |  |
| 960.        | 40.02           | 26.88                          |                 | 1460. | 53.41           |                 |                                |  |
| 970.        | 40.31           | 27.07                          |                 | 1470. | 53.67           |                 |                                |  |
| 980.        | 40.60           | 27.26                          |                 | 1480. | 53.92           |                 |                                |  |
| 990.        | 40.89           | 27.46                          |                 | 1490. | 54.17           |                 |                                |  |
| 1000.       | 41.18           | 27.65                          |                 | 1500. | 54.43           |                 |                                |  |

## 7. Conclusions

A concise representation of the viscosity of carbon dioxide, methane, and sulfur hexafluoride in the limit of zero density has been presented. For each gas the accuracy of the correlations and associated tabulations is estimated as one of  $\pm 0.3\%$  in the ambient temperature region, rising to  $\pm 2\%$  outside this region.

It has not proved possible to give a unified representation of all three gases since a two-parameter law of corresponding states is not obeyed within an acceptable tolerance.

## 8. Acknowledgments

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