Viscosity Measurements on Gaseous Methane: Re-evaluation

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Abstract
Previous experimental viscosity data for gaseous methane, published by Schley et al. (Int J Thermophys 25:1623, 2004) and originally obtained using a vibrating-wire viscometer in the temperature range between 260 K and 360 K, were re-evaluated after an improved re-calibration. For this purpose, a new reference value for argon at 298.15 K and at zero density, proposed by Vogel et al. (Mol Phys 108:3335, 2010) and further updated by Hellmann (Private Communication, 2020), was applied.

Keywords Methane · Re-evaluation · Viscosity

Previous measurements of the viscosity $\eta$ for methane, carried out by Schley et al. [1] using a vibrating-wire viscometer with freely suspended weight and additional measurements of temperature $T$ and pressure $p$ for calculating the required density $\rho$ with an equation of state by Setzmann, Wagner, and de Reuck [2, 3], have been re-evaluated. The re-evaluation concerns the specification of the wire radius by means of an improved calibration. The re-evaluated data, given as $\eta pT$ values, are to be used together with other new accurate $\eta pT$ data to generate a prospective viscosity correlation for methane.

The re-calibration of the applied vibrating-wire viscometer was performed in that way that the radius of the wire was newly specified using previous measurements on argon [4]. The original calibration employed an experimentally based reference value of Kestin and Leidenfrost [5] nowadays considered as obsolete. For the re-calibration, we used a today accepted value for the zero-density viscosity coefficient of argon, derived by Vogel et al. [6] from an ab initio potential on the basis of the kinetic theory of dilute gases and upgraded by Hellmann [7], to...
be \( \eta_{0,\text{Ar},298.15K} = 22.5534 \, \mu\text{Pa}\cdot\text{s} \) with a standard uncertainty of \( \pm 0.07 \% \). The wire radius amounts to 12.7548 \( \mu\text{m} \) using the new reference value for argon.

The results reported in Table II of the previous paper of Schley et al. [1] were restricted to \( \eta pp \) triples along the measured isotherms. In this new report, we include more details in order to make the information comparable to that given for the new viscosity measurements by Humberg et al. [8]. They recommended further a re-evaluation of the measurements by Schley et al. The individual points were not exactly measured at the nominal temperature of an isotherm \( T_{\text{nom}} \), but could be kept within small deviations from the nominal temperature each. The experimental viscosity data were adjusted to \( \eta_{T_{\text{nom}}} \) values at the nominal temperature using a Taylor series expansion restricted to the first power in temperature.

For this, the experimentally determined value of the initial density dependence \( (\partial \eta / \partial T)_p = (0.030 \text{ to } 0.034) \, \mu\text{Pa}\cdot\text{s} \cdot \text{K}^{-1} \) advised by Vogel [9] for methane was used. Further, it is supposed that the density values \( \rho_{\text{eos}(T,p)} \), computed from the measured data for \( T \) and \( p \) using the equation of state by Setzmann, Wagner, and de Reuck [2, 3], and those for the isotherms are the same. As a result of this, the pressures \( p_{T_{\text{nom}},\rho_{\text{nom}}} \) at the nominal temperature changed and were recalculated from the densities. The improved experimental \( \eta pp T \) data of this work for the previous measurements of Schley et al. [1] on methane (six isotherms at 260 K, 280 K, 300 K, 320 K, 340 K, and 360 K) are summarized in Tables 1, 2, 3, 4, 5, and 6, in which the data are given in the sequential arrangement of the original measurements.

The experimental data of each nominal isotherm for methane were correlated as a function of the reduced density \( \delta \) by means of a power-series representation restricted to the fourth power:

\[
\eta(\tau, \delta) = \sum_{i=0}^{4} \eta_i(\tau) \delta^i, \quad \delta = \frac{\rho}{\rho_{c,\text{CH}_4}}, \quad \tau = \frac{T}{T_{c,\text{CH}_4}}, \tag{1}
\]

with \( \rho_{c,\text{CH}_4} = 162.66 \, \text{kg}\cdot\text{m}^{-3}, T_{c,\text{CH}_4} = 190.564 \, \text{K} \).

Here, \( \delta \) is the reduced density, whereas \( \tau \) is the reduced temperature. The values of the critical density \( \rho_{c,\text{CH}_4} \) and of the critical temperature \( T_{c,\text{CH}_4} \) are those given by Setzmann, Wagner, and de Reuck [2, 3]. Weighting factors \( w_i = 100 \eta_{\text{exp},i}^{-2} \) were used in the multiple linear least-squares regression to minimize the weighted sum of squares \( \sigma = \sum_i w_i (\eta_{\text{cor},i} - \eta_{\text{exp},i})^2 \) as criterion for the quality of the representation of the considered isotherm. The coefficients \( \eta_i(\tau) \) of Eq. 1 including their standard deviations \( s.d._{\eta_i} \) and the weighted sum of squares \( \sigma \) for each isotherm are given in Table 7.
Table 1 Re-evaluated experimental $\eta ppT$ data for methane at 260 K

| $T$ (K) | $p$ (MPa) | $p_{260K,\text{eos}}$ (MPa) | $\rho_{\text{cont}T,p}$ (kg⋅m$^{-3}$) | $\eta$ (μPa⋅s) | $\eta_{260K}$ (μPa⋅s) |
|---------|-----------|-----------------------------|-------------------------------------|----------------|---------------------|
| 260.13  | 19.854    | 19.825                      | 205.16                              | 22.620         | 22.615              |
| 260.10  | 19.492    | 19.470                      | 202.55                              | 22.326         | 22.322              |
| 260.07  | 19.007    | 18.992                      | 198.92                              | 21.910         | 21.908              |
| 260.04  | 18.479    | 18.471                      | 194.80                              | 21.462         | 21.461              |
| 260.01  | 17.991    | 17.989                      | 190.81                              | 21.029         | 21.029              |
| 259.99  | 17.391    | 17.393                      | 185.65                              | 20.498         | 20.499              |
| 259.97  | 17.018    | 17.023                      | 182.32                              | 20.159         | 20.160              |
| 259.96  | 16.534    | 16.541                      | 177.81                              | 19.702         | 19.703              |
| 259.96  | 16.002    | 16.009                      | 172.61                              | 19.208         | 19.209              |
| 259.96  | 15.523    | 15.529                      | 167.73                              | 18.755         | 18.756              |
| 259.96  | 15.003    | 15.009                      | 162.23                              | 18.261         | 18.262              |
| 259.97  | 14.493    | 14.498                      | 156.61                              | 17.770         | 17.771              |
| 259.97  | 13.988    | 13.992                      | 150.85                              | 17.297         | 17.298              |
| 259.97  | 13.497    | 13.501                      | 145.09                              | 16.817         | 16.818              |
| 259.97  | 13.013    | 13.017                      | 139.25                              | 16.369         | 16.370              |
| 259.97  | 12.475    | 12.478                      | 132.60                              | 15.876         | 15.877              |
| 259.98  | 12.011    | 12.013                      | 126.76                              | 15.456         | 15.457              |
| 259.98  | 11.4752   | 11.4771                     | 119.942                             | 14.979         | 14.980              |
| 259.98  | 11.0092   | 11.0110                     | 113.968                             | 14.572         | 14.573              |
| 259.98  | 10.4966   | 10.4982                     | 107.385                             | 14.162         | 14.162              |
| 259.99  | 9.9936    | 9.9944                      | 100.939                             | 13.769         | 13.769              |
| 260.00  | 9.4550    | 9.4550                      | 94.099                              | 13.367         | 13.367              |
| 260.09  | 9.0135    | 9.0077                      | 88.499                              | 13.054         | 13.051              |
| 260.07  | 8.4848    | 8.4807                      | 82.010                              | 12.708         | 12.705              |
| 260.06  | 7.9975    | 7.9943                      | 76.144                              | 12.410         | 12.408              |
| 260.04  | 7.5015    | 7.4996                      | 70.312                              | 12.123         | 12.122              |
| 260.02  | 6.9988    | 6.9980                      | 64.550                              | 11.8522        | 11.8515             |
| 260.00  | 6.5024    | 6.5024                      | 59.013                              | 11.6044        | 11.6044             |
| 259.99  | 5.9959    | 5.9962                      | 53.520                              | 11.3753        | 11.3756             |
| 259.97  | 5.4974    | 5.4983                      | 48.279                              | 11.1632        | 11.1643             |
| 259.96  | 4.9940    | 4.9951                      | 43.144                              | 10.9688        | 10.9701             |
| 259.96  | 4.7449    | 4.7459                      | 40.661                              | 10.8774        | 10.8787             |
| 259.96  | 4.4832    | 4.4841                      | 38.094                              | 10.7878        | 10.7892             |
| 259.97  | 4.2613    | 4.2620                      | 35.949                              | 10.7148        | 10.7159             |
| 259.97  | 3.9886    | 3.9893                      | 33.356                              | 10.6234        | 10.6245             |
| 259.97  | 3.7393    | 3.7399                      | 31.024                              | 10.5494        | 10.5504             |
| 259.97  | 3.4825    | 3.4829                      | 28.659                              | 10.4806        | 10.4816             |
| 259.97  | 3.2477    | 3.2482                      | 26.532                              | 10.4108        | 10.4119             |
| 259.96  | 2.9859    | 2.9865                      | 24.196                              | 10.3439        | 10.3453             |
| 259.97  | 2.7329    | 2.7332                      | 21.972                              | 10.2837        | 10.2847             |
| 259.97  | 2.5032    | 2.5036                      | 19.985                              | 10.2308        | 10.2318             |
| 259.97  | 2.2622    | 2.2625                      | 17.929                              | 10.1784        | 10.1794             |
| $T$ (K) | $p$ (MPa) | $p_{260\,K,\mu}$ (MPa) | $p_{\text{out}}(T,p)$ (kg·m$^{-3}$) | $\eta$ (μPa·s) | $\eta_{260\,K}$ (μPa·s) |
|---------|-----------|-------------------------|-----------------------------------|----------------|------------------------|
| 259.97  | 1.9976    | 1.9979                  | 15.707                            | 10.1176        | 10.1186                |
| 259.97  | 1.7530    | 1.7532                  | 13.683                            | 10.0703        | 10.0714                |
| 259.97  | 1.5006    | 1.5008                  | 11.6262                           | 10.0250        | 10.0261                |
| 260.11  | 1.2526    | 1.2521                  | 9.6289                            | 9.9870         | 9.9833                 |
| 260.11  | 1.00295   | 1.00250                 | 7.6539                            | 9.9406         | 9.9369                 |
| 260.11  | 0.89975   | 0.89934                 | 6.8461                            | 9.9258         | 9.9220                 |
| 260.11  | 0.80037   | 0.80007                 | 6.0726                            | 9.9111         | 9.9073                 |
| 260.11  | 0.70073   | 0.70036                 | 5.3015                            | 9.8932         | 9.8895                 |
| 260.11  | 0.60083   | 0.60060                 | 4.5327                            | 9.8833         | 9.8795                 |
| 260.12  | 0.49743   | 0.49725                 | 3.7415                            | 9.8665         | 9.8625                 |
| 260.12  | 0.39841   | 0.39817                 | 2.9884                            | 9.8524         | 9.8484                 |
| 260.12  | 0.30137   | 0.30118                 | 2.2544                            | 9.8381         | 9.8341                 |
| 260.11  | 0.20117   | 0.20113                 | 1.5007                            | 9.8227         | 9.8189                 |
| 260.12  | 0.10188   | 0.10185                 | 0.75786                           | 9.8026         | 9.7985                 |
Table 2 Re-evaluated experimental $\eta_{pp}T$ data for methane at 280 K

| $T$ (K) | $p$ (MPa) | $p_{280K,\text{eos}}$ (MPa) | $\rho_{\text{out}(T,p)}$ (kg·m$^{-3}$) | $\eta$ (μPa·s) | $\eta_{280K}$ (μPa·s) |
|---------|-----------|----------------------------|-------------------------------------|----------------|---------------------|
| 280.00  | 19.837    | 19.837                     | 176.38                              | 20.236         | 20.236              |
| 280.01  | 19.469    | 19.467                     | 173.62                              | 19.975         | 19.974              |
| 280.02  | 19.016    | 19.013                     | 170.14                              | 19.655         | 19.654              |
| 280.02  | 18.514    | 18.511                     | 166.19                              | 19.301         | 19.300              |
| 280.02  | 17.996    | 17.993                     | 161.99                              | 18.931         | 18.931              |
| 280.01  | 17.504    | 17.503                     | 157.90                              | 18.567         | 18.566              |
| 280.01  | 17.005    | 17.004                     | 153.63                              | 18.219         | 18.218              |
| 280.01  | 16.510    | 16.509                     | 149.28                              | 17.849         | 17.849              |
| 280.00  | 16.005    | 16.005                     | 144.74                              | 17.495         | 17.495              |
| 280.00  | 15.504    | 15.504                     | 140.13                              | 17.140         | 17.140              |
| 279.99  | 15.003    | 15.004                     | 135.43                              | 16.780         | 16.780              |
| 280.06  | 14.481    | 14.475                     | 130.37                              | 16.407         | 16.405              |
| 280.08  | 13.943    | 13.934                     | 125.10                              | 16.036         | 16.033              |
| 280.07  | 13.505    | 13.498                     | 120.79                              | 15.744         | 15.741              |
| 280.05  | 13.006    | 13.001                     | 115.838                             | 15.417         | 15.415              |
| 280.03  | 12.485    | 12.483                     | 110.624                             | 15.075         | 15.074              |
| 280.01  | 12.000    | 11.9991                    | 105.734                             | 14.772         | 14.771              |
| 279.98  | 11.4938   | 11.4953                    | 100.625                             | 14.458         | 14.459              |
| 279.96  | 11.0031   | 11.0059                    | 95.662                              | 14.167         | 14.168              |
| 279.94  | 10.4991   | 10.5031                    | 90.573                              | 13.878         | 13.880              |
| 279.91  | 9.9988    | 10.0044                    | 85.551                              | 13.601         | 13.604              |
| 279.90  | 9.4946    | 9.5003                     | 80.512                              | 13.336         | 13.340              |
| 279.88  | 8.9919    | 8.9982                     | 75.540                              | 13.080         | 13.084              |
| 279.89  | 8.5157    | 8.5210                     | 70.868                              | 12.851         | 12.854              |
| 279.88  | 7.9776    | 7.9829                     | 65.670                              | 12.603         | 12.607              |
| 279.88  | 7.4998    | 7.5047                     | 61.121                              | 12.397         | 12.401              |
| 279.88  | 7.0039    | 7.0083                     | 56.475                              | 12.194         | 12.198              |
| 279.88  | 6.5016    | 6.5055                     | 51.851                              | 12.004         | 12.008              |
| 279.89  | 5.9960    | 5.9993                     | 47.283                              | 11.8222        | 11.8259             |
| 279.90  | 5.4944    | 5.4971                     | 42.840                              | 11.6536        | 11.6569             |
| 279.91  | 4.9837    | 4.9858                     | 38.410                              | 11.4929        | 11.4958             |
| 279.93  | 4.7424    | 4.7439                     | 36.347                              | 11.4196        | 11.4219             |
| 279.95  | 4.4872    | 4.4882                     | 34.189                              | 11.3484        | 11.3501             |
| 279.97  | 4.2319    | 4.2324                     | 32.054                              | 11.2760        | 11.2769             |
| 279.99  | 3.9980    | 3.9981                     | 30.119                              | 11.2164        | 11.2167             |
| 280.00  | 3.7364    | 3.7364                     | 27.981                              | 11.1446        | 11.1446             |
| 280.01  | 3.4973    | 3.4971                     | 26.047                              | 11.0899        | 11.0896             |
| 280.01  | 3.2471    | 3.2469                     | 24.047                              | 11.0308        | 11.0305             |
| 280.02  | 2.9988    | 2.9985                     | 22.083                              | 10.9727        | 10.9721             |
| 280.03  | 2.7428    | 2.7425                     | 20.081                              | 10.9225        | 10.9215             |
| 280.04  | 2.4965    | 2.4961                     | 18.176                              | 10.8711        | 10.8698             |
| 280.04  | 2.2264    | 2.2260                     | 16.111                              | 10.8162        | 10.8149             |
Table 2 (continued)

| $T$ (K) | $p$ (MPa) | $p_{280K_{\text{ref}}}$ (MPa) | $\rho_{\text{out}(T,p)}$ (kg·m$^{-3}$) | $\eta$ (μPa·s) | $\eta_{280K}$ (μPa·s) |
|--------|-----------|-----------------------------|-----------------------------|--------------|-----------------|
| 280.03 | 1.9909    | 1.9906                      | 14.332                      | 10.7728      | 10.7718         |
| 280.02 | 1.7459    | 1.7457                      | 12.500                      | 10.7254      | 10.7248         |
| 280.02 | 1.4976    | 1.4975                      | 10.6637                     | 10.6821      | 10.6814         |
| 280.01 | 1.2405    | 1.2404                      | 8.7835                      | 10.6391      | 10.6388         |
| 280.00 | 0.99793   | 0.99789                     | 7.0283                      | 10.6023      | 10.6023         |
| 280.00 | 0.90042   | 0.90043                     | 6.3279                      | 10.5874      | 10.5874         |
| 280.00 | 0.79968   | 0.79975                     | 5.6075                      | 10.5717      | 10.5717         |
| 279.99 | 0.70064   | 0.70073                     | 4.9025                      | 10.5570      | 10.5573         |
| 279.99 | 0.60094   | 0.60100                     | 4.1958                      | 10.5466      | 10.5469         |
| 279.99 | 0.49987   | 0.49983                     | 3.4824                      | 10.5300      | 10.5303         |
| 279.99 | 0.40047   | 0.40050                     | 2.7839                      | 10.5194      | 10.5197         |
| 279.99 | 0.30068   | 0.30074                     | 2.0857                      | 10.5018      | 10.5021         |
| 279.99 | 0.19854   | 0.19853                     | 1.3741                      | 10.4868      | 10.4871         |
| 279.99 | 0.10051   | 0.10049                     | 0.69416                     | 10.4666      | 10.4669         |
Table 3  Re-evaluated experimental $\eta_{ppT}$ data for methane at 300 K

| $T$ (K) | $p$ (MPa) | $p_{300K, eos}$ (MPa) | $\rho_{eos(T,p)}$ (kg·m$^{-3}$) | $\eta$ (μPa·s) | $\eta_{300K}$ (μPa·s) |
|---------|-----------|-----------------------|-----------------------------|--------------|-------------------------|
| 300.36  | 20.079    | 20.028                | 155.47                      | 19.028       | 19.016                  |
| 300.35  | 19.572    | 19.525                | 151.95                      | 18.735       | 18.723                  |
| 300.33  | 18.984    | 18.941                | 147.77                      | 18.400       | 18.390                  |
| 300.31  | 18.512    | 18.473                | 144.35                      | 18.131       | 18.121                  |
| 300.29  | 18.000    | 17.965                | 140.57                      | 17.839       | 17.830                  |
| 300.28  | 17.508    | 17.476                | 136.86                      | 17.560       | 17.551                  |
| 300.27  | 17.008    | 16.978                | 133.02                      | 17.276       | 17.267                  |
| 300.26  | 16.499    | 16.471                | 129.05                      | 16.983       | 16.974                  |
| 300.36  | 15.996    | 15.959                | 124.98                      | 16.702       | 16.690                  |
| 300.34  | 15.512    | 15.479                | 121.12                      | 16.441       | 16.430                  |
| 300.33  | 15.006    | 14.975                | 117.009                     | 16.162       | 16.151                  |
| 300.32  | 14.545    | 14.517                | 113.233                     | 15.923       | 15.912                  |
| 300.30  | 14.048    | 14.023                | 109.131                     | 15.656       | 15.646                  |
| 300.28  | 13.479    | 13.457                | 104.400                     | 15.364       | 15.355                  |
| 300.27  | 12.999    | 12.979                | 100.374                     | 15.119       | 15.110                  |
| 300.27  | 12.519    | 12.500                | 96.330                      | 14.882       | 14.873                  |
| 300.27  | 12.016    | 11.9977               | 92.073                      | 14.642       | 14.634                  |
| 300.26  | 11.5266   | 11.5101               | 87.939                      | 14.411       | 14.403                  |
| 300.25  | 10.9621   | 10.9474               | 83.171                      | 14.157       | 14.149                  |
| 300.25  | 10.5010   | 10.4872               | 79.280                      | 13.953       | 13.945                  |
| 300.24  | 9.9986    | 9.9862                | 75.059                      | 13.740       | 13.732                  |
| 300.24  | 9.5112    | 9.4996                | 70.981                      | 13.540       | 13.532                  |
| 300.23  | 9.0100    | 8.9997                | 66.817                      | 13.342       | 13.335                  |
| 300.23  | 8.4735    | 8.4640                | 62.390                      | 13.141       | 13.133                  |
| 300.36  | 7.9941    | 7.9804                | 58.430                      | 12.969       | 12.958                  |
| 300.35  | 7.5080    | 7.4957                | 54.499                      | 12.803       | 12.792                  |
| 300.34  | 7.0117    | 7.0008                | 50.528                      | 12.638       | 12.627                  |
| 300.33  | 6.4804    | 6.4709                | 46.328                      | 12.471       | 12.461                  |
| 300.32  | 5.9811    | 5.9728                | 42.431                      | 12.322       | 12.312                  |
| 300.31  | 5.5018    | 5.4945                | 38.738                      | 12.186       | 12.176                  |
| 300.30  | 5.0006    | 4.9944                | 34.929                      | 12.053       | 12.043                  |
| 300.30  | 4.7591    | 4.7532                | 33.112                      | 11.9902      | 11.9805                 |
| 300.30  | 4.5009    | 4.4954                | 31.184                      | 11.9285      | 11.9189                 |
| 300.30  | 4.2541    | 4.2490                | 29.356                      | 11.8682      | 11.8585                 |
| 300.31  | 3.9947    | 3.9898                | 27.447                      | 11.8102      | 11.8002                 |
| 300.31  | 3.7562    | 3.7517                | 25.707                      | 11.7569      | 11.7469                 |
| 300.31  | 3.5015    | 3.4972                | 23.862                      | 11.7009      | 11.6910                 |
| 300.31  | 3.2523    | 3.2484                | 22.072                      | 11.6473      | 11.6373                 |
| 300.31  | 2.9960    | 2.9925                | 20.246                      | 11.5949      | 11.5849                 |
| 300.31  | 2.7408    | 2.7375                | 18.442                      | 11.5479      | 11.5379                 |
| 300.31  | 2.4972    | 2.4944                | 16.735                      | 11.5006      | 11.4906                 |
| 300.31  | 2.2534    | 2.2508                | 15.039                      | 11.4581      | 11.4481                 |
Table 3 (continued)

| $T$ (K) | $p$ (MPa) | $p_{300\text{K},eos}$ (MPa) | $\rho_{eos(T,p)}$ (kg·m$^{-3}$) | $\eta$ ($\mu$Pa·s) | $\eta_{300\text{K}}$ ($\mu$Pa·s) |
|---------|-----------|----------------------------|-------------------------------|----------------|----------------|
| 300.31  | 2.0059    | 2.0037                     | 13.332                        | 11.4137        | 11.4037        |
| 300.30  | 1.7534    | 1.7514                     | 11.6043                       | 11.3724        | 11.3627        |
| 300.30  | 1.4930    | 1.4914                     | 9.8376                        | 11.3321        | 11.3224        |
| 300.30  | 1.2532    | 1.2520                     | 8.2245                        | 11.2955        | 11.2859        |
| 300.30  | 0.99661   | 0.99556                    | 6.5121                        | 11.2574        | 11.2477        |
| 300.30  | 0.89862   | 0.89767                    | 5.8620                        | 11.2424        | 11.2327        |
| 300.31  | 0.79654   | 0.79568                    | 5.1870                        | 11.2284        | 11.2184        |
| 300.31  | 0.69734   | 0.69653                    | 4.5334                        | 11.2177        | 11.2077        |
| 300.31  | 0.60073   | 0.60009                    | 3.8990                        | 11.2062        | 11.1962        |
| 300.31  | 0.50093   | 0.50043                    | 3.2458                        | 11.1940        | 11.1840        |
| 300.31  | 0.40241   | 0.40197                    | 2.6031                        | 11.1802        | 11.1702        |
| 300.31  | 0.30029   | 0.29995                    | 1.9392                        | 11.1622        | 11.1522        |
| 300.32  | 0.20117   | 0.20097                    | 1.2968                        | 11.1503        | 11.1400        |
| 300.32  | 0.099667  | 0.099496                   | 0.64141                       | 11.1309        | 11.1206        |
Table 4  Re-evaluated experimental $\eta p p T$ data for methane at 320 K

| $T$ (K) | $p$ (MPa) | $p_{320K-eos}$ (MPa) | $\rho_{eos(T,p)}$ (kg⋅m$^{-3}$) | $\eta$ (μPa⋅s) | $\eta_{320K}$ (μPa⋅s) |
|---------|-----------|----------------------|-------------------------------|----------------|----------------------|
| 320.06  | 19.936    | 19.929               | 137.78                        | 18.266         | 18.264               |
| 320.04  | 19.508    | 19.503               | 135.04                        | 18.065         | 18.063               |
| 320.02  | 19.019    | 19.017               | 131.86                        | 17.832         | 17.831               |
| 320.01  | 18.525    | 18.524               | 128.60                        | 17.594         | 17.594               |
| 320.00  | 18.011    | 18.011               | 125.15                        | 17.357         | 17.357               |
| 319.99  | 17.539    | 17.540               | 121.96                        | 17.131         | 17.131               |
| 319.98  | 17.033    | 17.035               | 118.488                       | 16.900         | 16.901               |
| 319.97  | 16.525    | 16.528               | 114.963                       | 16.677         | 16.678               |
| 319.97  | 16.017    | 16.020               | 111.394                       | 16.437         | 16.438               |
| 319.96  | 15.495    | 15.498               | 107.700                       | 16.205         | 16.206               |
| 319.95  | 14.998    | 15.002               | 104.152                       | 15.983         | 15.984               |
| 319.94  | 14.500    | 14.505               | 100.573                       | 15.775         | 15.777               |
| 319.96  | 14.000    | 14.003               | 96.939                        | 15.563         | 15.564               |
| 319.95  | 13.493    | 13.496               | 93.254                        | 15.353         | 15.355               |
| 319.95  | 12.977    | 12.980               | 89.484                        | 15.142         | 15.144               |
| 319.95  | 12.482    | 12.485               | 85.852                        | 14.940         | 14.942               |
| 320.08  | 11.8703   | 11.8657              | 81.305                        | 14.712         | 14.710               |
| 320.07  | 11.5003   | 11.4964              | 78.591                        | 14.565         | 14.563               |
| 320.06  | 11.0000   | 10.9969              | 74.921                        | 14.381         | 14.379               |
| 320.05  | 10.4933   | 10.4909              | 71.208                        | 14.201         | 14.199               |
| 320.03  | 10.0023   | 10.0009              | 67.621                        | 14.030         | 14.029               |
| 320.02  | 9.4991    | 9.4983               | 63.952                        | 13.858         | 13.858               |
| 320.01  | 8.9947    | 8.9943               | 60.289                        | 13.693         | 13.693               |
| 320.00  | 8.4947    | 8.4947               | 56.676                        | 13.538         | 13.538               |
| 319.99  | 8.0047    | 8.0050               | 53.155                        | 13.389         | 13.389               |
| 319.98  | 7.4801    | 7.4807               | 49.410                        | 13.235         | 13.236               |
| 319.97  | 6.9969    | 6.9977               | 45.986                        | 13.103         | 13.104               |
| 319.96  | 6.4848    | 6.4859               | 42.386                        | 12.965         | 12.966               |
| 319.95  | 5.9993    | 6.0005               | 39.002                        | 12.837         | 12.839               |
| 319.95  | 5.4892    | 5.4902               | 35.477                        | 12.713         | 12.714               |
| 319.94  | 4.9997    | 5.0008               | 32.130                        | 12.599         | 12.600               |
| 319.94  | 4.7526    | 4.7538               | 30.453                        | 12.543         | 12.544               |
| 319.95  | 4.5079    | 4.5087               | 28.798                        | 12.492         | 12.493               |
| 319.96  | 4.2485    | 4.2491               | 27.055                        | 12.434         | 12.436               |
| 319.96  | 4.0085    | 4.0091               | 25.452                        | 12.385         | 12.387               |
| 319.96  | 3.7591    | 3.7597               | 23.795                        | 12.333         | 12.334               |
| 319.97  | 3.4921    | 3.4925               | 22.031                        | 12.283         | 12.284               |
| 319.97  | 3.2600    | 3.2604               | 20.507                        | 12.240         | 12.241               |
| 319.97  | 2.9995    | 2.9997               | 18.806                        | 12.190         | 12.191               |
| 319.98  | 2.7428    | 2.7431               | 17.141                        | 12.147         | 12.148               |
| 319.98  | 2.4932    | 2.4933               | 15.531                        | 12.103         | 12.104               |
| 319.98  | 2.2531    | 2.2533               | 13.993                        | 12.064         | 12.065               |
| $T$ (K) | $p$ (MPa) | $p_{320K}$ (MPa) | $\rho_{\text{out}(T,p)}$ (kg·m$^{-3}$) | $\eta$ (μPa·s) | $\eta_{320K}$ (μPa·s) |
|-------|---------|-----------------|---------------------------------|--------------|------------------|
| 319.98 | 2.0005  | 2.0007          | 12.384                          | 12.025       | 12.025           |
| 319.98 | 1.7492  | 1.7492          | 10.7924                         | 11.9839      | 11.9845          |
| 319.98 | 1.5026  | 1.5026          | 9.2415                          | 11.9473      | 11.9480          |
| 319.98 | 1.2501  | 1.2501          | 7.6633                          | 11.9125      | 11.9131          |
| 319.98 | 1.00008 | 1.00021         | 6.1106                          | 11.8788      | 11.8795          |
| 320.06 | 0.90159 | 0.90137         | 5.5003                          | 11.8655      | 11.8636          |
| 320.06 | 0.80028 | 0.80017         | 4.8757                          | 11.8545      | 11.8526          |
| 320.06 | 0.69957 | 0.69952         | 4.2565                          | 11.8418      | 11.8399          |
| 320.06 | 0.60029 | 0.60023         | 3.6477                          | 11.8293      | 11.8274          |
| 320.06 | 0.50009 | 0.50003         | 3.0348                          | 11.8181      | 11.8162          |
| 320.05 | 0.40013 | 0.40006         | 2.4251                          | 11.8044      | 11.8028          |
| 320.05 | 0.30001 | 0.29998         | 1.8159                          | 11.7916      | 11.7901          |
| 320.05 | 0.20142 | 0.20146         | 1.2175                          | 11.7783      | 11.7767          |
| 320.04 | 0.097179| 0.097227        | 0.58664                         | 11.7561      | 11.7549          |
| \(T\) (K) | \(p\) (MPa) | \(p_{340K,eos}\) (MPa) | \(\rho_{eos(T,p)}\) (kg⋅m\(^{-3}\)) | \(\eta\) (\(\mu\)Pa⋅s) | \(\eta_{340K}\) (\(\mu\)Pa⋅s) |
|---|---|---|---|---|---|
| 340.04 | 28.773 | 28.767 | 170.47 | 21.614 | 21.612 |
| 340.02 | 27.800 | 27.797 | 166.02 | 21.211 | 21.210 |
| 340.01 | 26.949 | 26.948 | 162.00 | 20.855 | 20.855 |
| 340.04 | 25.964 | 25.959 | 157.19 | 20.448 | 20.447 |
| 340.02 | 24.966 | 24.963 | 152.19 | 20.023 | 20.022 |
| 339.99 | 23.882 | 23.883 | 146.59 | 19.571 | 19.572 |
| 339.97 | 22.945 | 22.949 | 141.60 | 19.188 | 19.189 |
| 339.95 | 21.981 | 21.987 | 136.32 | 18.789 | 18.790 |
| 339.93 | 20.982 | 20.990 | 130.70 | 18.378 | 18.380 |
| 340.04 | 20.089 | 20.085 | 125.48 | 18.012 | 18.011 |
| 340.02 | 19.023 | 19.021 | 119.187 | 17.584 | 17.583 |
| 340.00 | 17.991 | 17.991 | 112.958 | 17.175 | 17.175 |
| 339.97 | 17.019 | 17.022 | 106.987 | 16.798 | 16.799 |
| 339.95 | 15.995 | 15.999 | 100.573 | 16.412 | 16.413 |
| 339.92 | 15.002 | 15.008 | 94.274 | 16.048 | 16.051 |
| 339.92 | 14.496 | 14.501 | 91.024 | 15.867 | 15.870 |
| 339.94 | 14.010 | 14.014 | 87.890 | 15.695 | 15.697 |
| 339.97 | 13.494 | 13.496 | 84.535 | 15.511 | 15.512 |
| 339.98 | 12.971 | 12.972 | 81.138 | 15.331 | 15.331 |
| 339.98 | 12.494 | 12.495 | 78.033 | 15.172 | 15.173 |
| 339.98 | 11.8825 | 11.8835 | 74.042 | 14.970 | 14.971 |
| 339.98 | 11.4908 | 11.4918 | 71.484 | 14.849 | 14.850 |
| 339.98 | 10.9859 | 10.9868 | 68.186 | 14.691 | 14.691 |
| 339.98 | 10.4666 | 10.4675 | 64.795 | 14.533 | 14.534 |
| 339.98 | 9.9898 | 9.9906 | 61.686 | 14.390 | 14.391 |
| 339.98 | 9.4977 | 9.4984 | 58.481 | 14.248 | 14.249 |
| 339.98 | 9.0019 | 9.0026 | 55.260 | 14.109 | 14.110 |
| 339.99 | 8.4909 | 8.4912 | 51.949 | 13.973 | 13.974 |
| 340.00 | 8.0050 | 8.0050 | 48.812 | 13.847 | 13.847 |
| 340.00 | 7.9406 | 7.9405 | 48.397 | 13.828 | 13.828 |
| 340.00 | 7.4842 | 7.4843 | 45.467 | 13.715 | 13.715 |
| 340.00 | 7.0060 | 7.0060 | 42.410 | 13.596 | 13.596 |
| 340.00 | 6.4721 | 6.4720 | 39.015 | 13.470 | 13.470 |
| 340.00 | 5.9912 | 5.9912 | 35.977 | 13.358 | 13.358 |
| 340.00 | 5.5006 | 5.5007 | 32.897 | 13.254 | 13.254 |
| 340.00 | 4.9156 | 4.9157 | 29.251 | 13.136 | 13.136 |
| 340.00 | 4.7412 | 4.7411 | 28.169 | 13.100 | 13.100 |
| 340.06 | 4.4865 | 4.4857 | 26.591 | 13.050 | 13.048 |
| 340.06 | 4.2523 | 4.2515 | 25.150 | 13.007 | 13.005 |
| 340.06 | 4.0025 | 4.0017 | 23.619 | 12.959 | 12.957 |
| 340.05 | 3.7476 | 3.7469 | 22.063 | 12.913 | 12.911 |
| 340.05 | 3.5061 | 3.5056 | 20.596 | 12.871 | 12.870 |
Table 5 (continued)

| $T$ (K) | $p$ (MPa) | $p_{340\text{K-}\rho_{\text{ua}}}$ (MPa) | $\rho_{\text{ua}(T,p)}$ (kg·m$^{-3}$) | $\eta$ (µPa·s) | $\eta_{340\text{K}}$ (µPa·s) |
|---------|-----------|--------------------------------------|-------------------------------------|---------------|-----------------|
| 340.04  | 3.2440    | 3.2436                               | 19.010                              | 12.825        | 12.824          |
| 340.04  | 3.0040    | 3.0035                               | 17.563                              | 12.785        | 12.784          |
| 340.04  | 2.7531    | 2.7527                               | 16.058                              | 12.744        | 12.743          |
| 340.03  | 2.4972    | 2.4969                               | 14.530                              | 12.709        | 12.708          |
| 340.03  | 2.2541    | 2.2539                               | 13.085                              | 12.671        | 12.670          |
| 340.02  | 1.9946    | 1.9944                               | 11.5491                             | 12.635        | 12.634          |
| 340.02  | 1.7492    | 1.7491                               | 10.1040                             | 12.597        | 12.596          |
| 340.02  | 1.4925    | 1.4924                               | 8.5991                              | 12.565        | 12.564          |
| 340.02  | 1.2636    | 1.2634                               | 7.2635                              | 12.538        | 12.538          |
| 340.02  | 1.00122   | 1.00114                              | 5.7401                              | 12.502        | 12.502          |
| 340.02  | 0.90554   | 0.90556                              | 5.1866                              | 12.490        | 12.489          |
| 340.02  | 0.80276   | 0.80270                              | 4.5931                              | 12.476        | 12.476          |
| 340.02  | 0.70130   | 0.70118                              | 4.0084                              | 12.465        | 12.465          |
| 340.02  | 0.60212   | 0.60207                              | 3.4380                              | 12.453        | 12.452          |
| 340.01  | 0.50192   | 0.50189                              | 2.8631                              | 12.442        | 12.441          |
| 340.00  | 0.40440   | 0.40448                              | 2.3045                              | 12.432        | 12.432          |
| 339.99  | 0.30053   | 0.30056                              | 1.7109                              | 12.419        | 12.419          |
| 339.99  | 0.19776   | 0.19783                              | 1.12459                             | 12.401        | 12.401          |
| 340.00  | 0.099605  | 0.099633                             | 0.56584                             | 12.383        | 12.383          |
| T (K) | p (MPa) | p_{360 K rer} (MPa) | \rho_{\text{eos(T,p)}} (kg \cdot m^{-3}) | \eta (\mu Pa \cdot s) | \eta_{360 K} (\mu Pa \cdot s) |
|------|--------|---------------------|---------------------------------|-------------------|---------------------|
| 360.01 | 29.185 | 29.183 | 158.93 | 21.191 | 21.191 |
| 360.01 | 28.029 | 28.028 | 153.89 | 20.767 | 20.766 |
| 360.02 | 26.906 | 26.903 | 148.82 | 20.348 | 20.347 |
| 360.03 | 25.978 | 25.975 | 144.53 | 20.021 | 20.020 |
| 360.03 | 24.996 | 24.993 | 139.87 | 19.659 | 19.658 |
| 360.04 | 24.027 | 24.023 | 135.15 | 19.298 | 19.297 |
| 360.04 | 22.887 | 22.883 | 129.46 | 18.900 | 18.899 |
| 360.03 | 21.983 | 21.980 | 124.85 | 18.576 | 18.575 |
| 360.03 | 20.985 | 20.982 | 119.640 | 18.220 | 18.219 |
| 360.02 | 19.942 | 19.940 | 114.086 | 17.854 | 17.853 |
| 360.01 | 18.990 | 18.989 | 108.918 | 17.522 | 17.522 |
| 360.00 | 18.015 | 18.015 | 103.537 | 17.195 | 17.195 |
| 359.99 | 17.036 | 17.036 | 98.044 | 16.869 | 16.869 |
| 359.99 | 16.004 | 16.005 | 92.174 | 16.535 | 16.535 |
| 359.99 | 14.993 | 14.994 | 86.356 | 16.214 | 16.215 |
| 359.99 | 14.494 | 14.495 | 83.458 | 16.056 | 16.056 |
| 359.98 | 13.983 | 13.984 | 80.480 | 15.901 | 15.901 |
| 359.99 | 13.481 | 13.481 | 77.541 | 15.753 | 15.753 |
| 359.99 | 12.985 | 12.986 | 74.635 | 15.607 | 15.607 |
| 359.99 | 12.488 | 12.488 | 71.707 | 15.462 | 15.462 |
| 360.00 | 11.9849 | 11.9849 | 68.740 | 15.322 | 15.322 |
| 360.00 | 11.504 | 11.504 | 65.901 | 15.189 | 15.189 |
| 360.00 | 10.9925 | 10.9925 | 62.878 | 15.051 | 15.051 |
| 360.00 | 10.4821 | 10.4821 | 59.860 | 14.917 | 14.917 |
| 359.98 | 9.9973 | 9.9980 | 56.998 | 14.790 | 14.791 |
| 359.98 | 9.4823 | 9.4829 | 53.955 | 14.660 | 14.661 |
| 359.98 | 8.9782 | 8.9788 | 50.980 | 14.537 | 14.538 |
| 359.98 | 8.5100 | 8.5106 | 48.222 | 14.427 | 14.427 |
| 359.97 | 8.0209 | 8.0218 | 45.348 | 14.309 | 14.310 |
| 359.97 | 7.4950 | 7.4959 | 42.264 | 14.193 | 14.194 |
| 359.98 | 7.0014 | 7.0018 | 39.376 | 14.089 | 14.090 |
| 359.97 | 6.5000 | 6.5007 | 36.457 | 13.981 | 13.981 |
| 359.97 | 6.0060 | 6.0065 | 33.590 | 13.882 | 13.883 |
| 359.96 | 5.5031 | 5.5038 | 30.686 | 13.782 | 13.783 |
| 359.96 | 5.0028 | 5.0035 | 27.810 | 13.692 | 13.693 |
| 359.96 | 4.7515 | 4.7520 | 26.370 | 13.649 | 13.650 |
| 359.96 | 4.5021 | 4.5027 | 24.946 | 13.600 | 13.602 |
| 359.97 | 4.2508 | 4.2511 | 23.513 | 13.557 | 13.558 |
| 359.98 | 3.9983 | 3.9986 | 22.079 | 13.517 | 13.517 |
| 359.98 | 3.7513 | 3.7515 | 20.680 | 13.474 | 13.475 |
| 359.99 | 3.5007 | 3.5007 | 19.264 | 13.432 | 13.433 |
| 359.99 | 3.2510 | 3.2510 | 17.859 | 13.394 | 13.394 |
### Table 6 (continued)

| $T$ (K) | $p$ (MPa) | $p_{360 K, \rho_{\text{max}}}$ (MPa) | $p_{\text{eos}(T,p)}$ (kg⋅m$^{-3}$) | $\eta$ (μPa⋅s) | $\eta_{360 K}$ (μPa⋅s) |
|---------|-----------|---------------------------------|---------------------------------|----------------|----------------------|
| 359.98  | 3.0017    | 3.0018                          | 16.461                          | 13.355         | 13.356               |
| 359.98  | 2.7436    | 2.7438                          | 15.018                          | 13.318         | 13.319               |
| 359.98  | 2.4999    | 2.5001                          | 13.660                          | 13.289         | 13.289               |
| 359.97  | 2.2470    | 2.2471                          | 12.255                          | 13.252         | 13.253               |
| 359.97  | 1.9996    | 1.9998                          | 10.886                          | 13.216         | 13.217               |
| 359.96  | 1.7506    | 1.7508                          | 9.5131                          | 13.184         | 13.186               |
| 359.95  | 1.4988    | 1.4991                          | 8.1296                          | 13.154         | 13.155               |
| 359.95  | 1.2512    | 1.2515                          | 6.7736                          | 13.122         | 13.123               |
| 359.96  | 1.00063   | 1.00068                         | 5.4064                          | 13.092         | 13.093               |
| 359.96  | 0.90014   | 0.90031                         | 4.8596                          | 13.077         | 13.078               |
| 359.99  | 0.79961   | 0.79961                         | 4.3131                          | 13.071         | 13.071               |
| 359.99  | 0.69939   | 0.69949                         | 3.7695                          | 13.058         | 13.058               |
| 359.99  | 0.59989   | 0.59996                         | 3.2307                          | 13.046         | 13.046               |
| 359.98  | 0.50016   | 0.50026                         | 2.6916                          | 13.036         | 13.037               |
| 359.97  | 0.39974   | 0.39986                         | 2.1495                          | 13.021         | 13.022               |
| 359.97  | 0.29870   | 0.29874                         | 1.6049                          | 13.009         | 13.010               |
| 359.93  | 0.19670   | 0.19671                         | 1.05611                         | 12.994         | 12.997               |
| 359.92  | 0.10261   | 0.10272                         | 0.55056                         | 12.972         | 12.974               |

### Table 7 Coefficients of Eq. 1 for the re-evaluated viscosity measurements on methane

| $T$ (K) | $n$ | $\rho_{\text{max}}$ (kg⋅m$^{-3}$) | $\eta_0$ (μPa⋅s) | $\eta_1$ (μPa⋅s) | $\eta_2$ (μPa⋅s) |
|---------|-----|---------------------------------|-----------------|-----------------|-----------------|
| 260     | 4   | 205.16                          | 9.796 ± 0.001   | 2.712 ± 0.015   | 7.028 ± 0.060   |
| 280     | 4   | 176.38                          | 10.465 ± 0.001  | 2.858 ± 0.016   | 6.863 ± 0.070   |
| 300     | 4   | 155.47                          | 11.120 ± 0.001  | 2.965 ± 0.014   | 6.724 ± 0.070   |
| 320     | 4   | 137.78                          | 11.755 ± 0.001  | 3.034 ± 0.017   | 6.804 ± 0.096   |
| 340     | 4   | 170.47                          | 12.386 ± 0.001  | 2.996 ± 0.015   | 6.995 ± 0.069   |
| 360     | 4   | 158.93                          | 12.979 ± 0.001  | 3.122 ± 0.024   | 6.557 ± 0.119   |

| $T$ (K) | $\eta_3$ (μPa⋅s) | $\eta_4$ (μPa⋅s) | $\sigma$ |
|---------|-----------------|-----------------|---------|
| 260     | −2.586 ± 0.080  | 1.349 ± 0.034   | 0.023   |
| 280     | −2.504 ± 0.108  | 1.299 ± 0.053   | 0.020   |
| 300     | −2.514 ± 0.122  | 1.339 ± 0.068   | 0.015   |
| 320     | −2.978 ± 0.186  | 1.691 ± 0.115   | 0.015   |
| 340     | −3.144 ± 0.111  | 1.680 ± 0.057   | 0.017   |
| 360     | −2.556 ± 0.205  | 1.409 ± 0.113   | 0.025   |
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Declarations

Conflict of interest  The authors declare that they have no conflict of interest.

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