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Viscosity Measurements on Natural Gas: Re-evaluation

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Abstract
Previous experimental viscosity data for natural gas, 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 320 K, were re-evaluated after an improved re-calibration. For this purpose, a new reference value for the viscosity of 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, University of the Federal Armed Forces Hamburg, Hamburg, 2020), was applied. In addition, the density computed from the measured temperatures and pressures was determined using the equation of state by Kunz and Wagner (J Chem Eng Data 57:3032, 2012) instead of employing a calculation according to the International Standard ISO 12213 nowadays out of date.

Keywords Natural gas · Re-evaluation · Viscosity

1 Introduction

We appreciate the collaboration with Professor Span, particularly when we worked together on the viscosity correlation for ethane [1]. Professor Span's group performed measurements and modeling of the viscosity of nitrogen-carbon dioxide [2] and methane-ethane mixtures [3] which we used to derive diffusion coefficients. Previous measurements of the viscosity η for two samples of natural gas with different compositions, carried out by Schley et al. [4] using a vibrating-wire viscometer with freely suspended weight and additional measurements of temperature T and pressure p for the calculation of the required density ρ applying the International Standard ISO 12213 [5], have been re-evaluated. The re-evaluation

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concerns the determination of the wire radius by means of an improved calibration and the density calculation. In this work, the density is calculated using the equation of state by Kunz and Wagner [6].

During and since the investigation by Schley et al. [4], viscosity measurements on natural gases or on mixtures with at least three components of natural gases were performed by Nabizadeh and Mayinger [7], Assael et al. [8], Langelandsvik et al. [9], Atilhan et al. [10], Kashefi et al. [11], Jarrahian et al. [12], Nazeri et al. [13], and Al Ghafri et al. [14–16]. The re-evaluated data of this work are reported as $\eta\rho pT$ values and can be used together with the other data to update the database for natural gases. Moreover, mixture models using the current database for natural gases were discussed in the literature, e.g., by Chichester and Huber [17], by Heidaryan and Jarrahian [18], by Yang et al. [19], by Theran-Becerra et al. [20], or by Xiong et al. [21], and should enable to generate a prospective viscosity correlation for natural gases.

2 Re-evaluation of the Data

The re-calibration of the applied vibrating-wire viscometer was performed such that the radius of the wire was newly determined using previous measurements on argon [22]. The original calibration employed an experimentally based reference value of Kestin and Leidenfrost [23] nowadays considered as obsolete. For the re-calibration, we applied a today accepted value for the zero-density viscosity coefficient of argon, derived by Vogel et al. [24] from an ab initio potential on the basis of the kinetic theory of dilute gases and upgraded by Hellmann [25], to be $\eta_{0,\mathrm{Ar},298.15\mathrm{K}} = 22.5534$ μPa·s with a standard uncertainty of 0.07 %. The wire radius amounts to $R = 12.7548$ μm using the new reference value for argon.

The results reported in Tables III and IV of the previous paper of Schley et al. [4] were restricted to $\eta\rho p$ triples along the measured isotherms. In this work, we include more details in order to make the information comparable to recent viscosity measurements. The individual points were not exactly measured at the nominal temperature of an isotherm T_{nom}, but were kept within small deviations from the nominal temperature each. The experimental viscosity data were adjusted to $\eta_{T_{\mathrm{nom}}}$ values at the nominal temperature employing a Taylor series expansion restricted to the first power in temperature. For this, the experimentally determined value of the temperature coefficient $(\partial\eta/\partial T)_\rho = (0.020\ \text{to}\ 0.038)$ μPa·s·K^{-1} calculated from the fit of a double polynomial in temperature and in density to the re-evaluated experimental viscosity data for natural gas was used. In addition, it is supposed that the density values $\rho_{\mathrm{eos}}(T,p)$ computed from the measured data for p and T applying the equation of state by Kunz and Wagner [6] via their implementation in REFPROP (Lemmon et al. [26]) and those for the isotherms are the same. As a result of this, the pressures $p(T_{\mathrm{nom}},\rho_{\mathrm{eos}})$ at the nominal temperature changed and were recalculated from the

Table 1 Components and their mole fractions of the natural gas samples H and L given by Schley et al. [4] and used for the re-evaluated viscosity measurements of this work

Component	Mole fractions by Schley et al. mol%	Mole fractions used in this work mol%
Natural gas H	(Molar mass $M = 17.993$ kg·kmol^{-1}	
Methane	89.5669	89.5669
Ethane	6.1464	6.1464
Nitrogen	1.5324	1.5324
Propane	1.2532	1.2532
Carbon dioxide	0.7740	0.7740
Isobutane	0.2857	0.2857
Butane	0.1924	0.1924
Isopentane (+Neopentane)[a]	0.0565	0.0597
Hexane	0.0572	0.0572
Heptane	0.0340	0.0340
Pentane	0.0324	0.0324
Oxygen (+Argon)[b]	0.0419	0.02095
Argon[c]	0.0000	0.02095
Helium	0.0137	0.0137
Octane	0.0038	0.0038
Benzene	0.0021	0.0021
Toluene (+Xylenes)[a]	0.0009	0.0016
Nonane	0.0010	0.0010
Decane	0.0009	0.0009
Hydrogen (normal)	0.0007	0.0007
Xylenes[d]	0.0006	0.0000
Neopentane[e]	0.0032	0.0000
Natural gas L	(Molar mass $M = 17.993$ kg·kmol^{-1}	
Methane	84.3322	84.32343
Ethane	3.4085	3.40815
Nitrogen	9.7520	9.75099
Propane	0.6023	0.60224
Carbon dioxide	1.4523	1.45215
Isobutane	0.1033	0.10329
Butane	0.1282	0.12819
Isopentane (+Neopentane)[a]	0.0357	0.04130
Hexane	0.0388	0.03880
Heptane	0.0174	0.01740
Pentane	0.0350	0.03500
Oxygen (+Argon)[b]	0.0100	0.00500
Argon[c]	0.0000	0.00500
Helium	0.0520	0.05199
Octane	0.0041	0.00410

Table 1 (continued)

Natural gas L	(Molar mass $M = 17.993$ kg·kmol^{-1}	
Benzene	0.0250	0.02500
Toluene (+Xylenes)[a]	0.0031	0.00410
Nonane	0.0021	0.00210
Decane	0.0013	0.00130
Hydrogen (normal)	0.0005	0.00050
Xylenes[d]	0.0010	0.00000
Neopentane[e]	0.0056	0.00000

[a]In Ref. [4] separately treated

[b]In Ref. [4] treated together with argon

[c]In this work separately treated

[d]In this work treated together with toluene

[e]In this work treated together with isopentane

densities. Furthermore, it has to be stated that the original composition of the two samples natural gas H and L, given by Schley et al. in Table I of their paper including the uncertainties of the underlying gas analysis, were used in this work, but with some combinations of components to yield 20 components as input parameters for the mixture in REFPROP. Table 1 shows the mole fractions of the components of the two natural gas samples H and L that means the composition stated by Schley et al. and that used for the re-evaluation of the density and viscosity data in this work.

Schley et al. [4] summed up the mole fractions of oxygen and argon, whereas Table 1 reveals that we split the mole fractions in two parts with equal values and treated separately these components. To meet the requirement of REFPROP (Lemmon et al. [26]) to limit the components to the maximum number of 20, we added the mole fractions of the xylenes (0.0006 mol% for natural gas H and 0.0010 mol% for natural gas L) to that of toluene (an additional 0.0001 mol% was added to the mole fraction of toluene in the case of natural gas H to obtain unity) and the mole fraction of neopentane to that of isopentane (0.0032 mol% for natural gas H and 0.0056 mol% for natural gas L). Moreover, the composition for the sample of natural gas L given by Schley et al. does not yield unity. Therefore, we normalized the mole fractions of all components of this sample as shown in Table 1. Finally, the molar masses of each sample were recalculated, but the numbers do not change compared with the values given by Schley et al.

The improved experimental $\eta\rho pT$ data of this work for the previous measurements of Schley et al. [4] on the two samples of natural gas at four isotherms

Table 2 Re-evaluated experimental $\eta\rho pT$ data for natural gas H at 260 K

T K	p MPa	$p(260\,\mathrm{K}, \rho_{\mathrm{eos}})$ MPa	$\rho_{\mathrm{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\mathrm{eos}})$[a] μPa·s	$\eta(260\,\mathrm{K}, \rho_{\mathrm{eos}})$[a] μPa·s
260.11	0.094613	0.094572	0.78965	9.7275[b]	9.7238[b]
260.09	0.19927	0.19920	1.6691	9.7489	9.7459
260.12	0.31257	0.31243	2.6279	9.7660	9.7619
260.14	0.40275	0.40253	3.3962	9.7803	9.7755
260.12	0.49874	0.49850	4.2198	9.8012	9.7971
260.12	0.60688	0.60659	5.1539	9.8196	9.8155
260.12	0.70137	0.70103	5.9758	9.8309	9.8269
260.09	0.80069	0.80040	6.8465	9.8513	9.8482
260.04	0.89967	0.89952	7.7212	9.8668	9.8654
260.00	1.01443	1.01443	8.7429	9.8885	9.8885
259.98	1.2499	1.2500	10.8642	9.9397	9.9404
259.96	1.5032	1.5034	13.187	9.9963	9.9976
259.94	1.7473	1.7477	15.468	10.055	10.057
259.93	1.9980	1.9986	17.853	10.119	10.121
259.93	2.2519	2.2526	20.315	10.198	10.200
259.94	2.4971	2.4978	22.739	10.272	10.274
259.95	2.7646	2.7653	25.435	10.361	10.363
259.96	3.0003	3.0009	27.858	10.409	10.410
259.97	3.2525	3.2530	30.503	10.495	10.496
259.98	3.4920	3.4924	33.065	10.581	10.581
259.99	3.7517	3.7519	35.900	10.678	10.678
260.01	3.9946	3.9944	38.606	10.768	10.767
260.02	4.2431	4.2426	41.434	10.862	10.861
260.01	4.4950	4.4947	44.368	10.964	10.963
260.01	4.7424	4.7422	47.311	11.066	11.066
259.98	4.9915	4.9921	50.350	11.179	11.180
259.97	5.4940	5.4950	56.669	11.422	11.423
259.95	5.9944	5.9963	63.254	11.693	11.695
259.92	6.5106	6.5140	70.366	12.019	12.022
259.93	6.9892	7.0078	77.448	12.318	12.321
259.94	7.4869	7.4902	84.646	12.708	12.710
259.96	8.0035	8.0060	92.624	13.123	13.124
259.99	8.5061	8.5067	100.634	13.562	13.563
260.04	9.1950	9.1919	111.94	14.247	14.245
260.07	9.5437	9.5378	117.76	14.595	14.592
260.09	9.9894	9.9812	125.28	15.060	15.057
260.11	10.4881	10.4771	133.71	15.613	15.609
260.11	11.0035	10.9916	142.40	16.205	16.201
260.10	11.4809	11.4692	150.32	16.787	16.784
260.09	12.004	11.9926	158.76	17.429	17.426
260.09	12.509	12.497	166.61	18.060	18.057

Table 2 (continued)

T K	p MPa	$p(260\,\text{K}, \rho_{\text{eos}})$ MPa	$\rho_{\text{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\text{eos}})^{\text{a}}$ μPa·s	$\eta(260\,\text{K}, \rho_{\text{eos}})^{\text{a}}$ μPa·s
260.09	12.995	12.982	173.83	18.661	18.658
260.11	13.500	13.483	180.96	19.296	19.293
259.96	14.139	14.145	189.81	20.183	20.184
259.99	14.688	14.690	196.60	20.855	20.855
259.99	14.993	14.995	200.23	21.225	21.225
259.98	15.520	15.524	206.23	21.866	21.866
259.98	15.996	16.000	211.32	22.434	22.435
259.96	16.659	16.667	218.01	23.204	23.206
259.95	16.992	17.003	221.19	23.579	23.581
259.96	17.498	17.507	225.75	24.122	24.124
259.98	18.019	18.024	230.17	24.675	24.676
260.00	18.510	18.510	234.12	25.197	25.197
260.00	19.069	19.069	238.42	25.760	25.760
260.00	19.508	19.508	241.65	26.197	26.197
260.00	20.017	20.017	245.22	26.694	26.694

[a] $U_{\text{c,r}}(\eta) = 1.0\,\%$

[b] Influenced by slip

(260 K, 280 K, 300 K, and 320 K) are summarized in Tables 2, 3, 4, and 5 for natural gas H (highly caloric) and in Tables 6, 7, 8, and 9 for natural gas L (lowly caloric), in which the data are given in the sequential arrangement of the original measurements. The experimental points at pressures below 0.19 MPa are influenced by the slip effect, they are marked in Tables 2, 3, 4, 5, 6, 7, 8, and 9. The relative combined expanded ($k = 2$) uncertainty $U_{\text{c,r}}(\eta)$ is estimated to be 1.0 % for the 260 K isotherms and 0.8 % for the 280 K, 300 K, and 320 K isotherms of the natural gas samples H and L. We note that the uncertainty reported by Schley et al. (0.5 %) was a standard uncertainty ($k = 1$) and could be slightly improved in this work due to the application of a more accurate calibration value and an improved density calculation, except for the 260 K isotherms influenced by the retrograde behavior.

The experimental data of each nominal isotherm for natural gas samples H and L were correlated as a function of the reduced density δ by means of a power series representation restricted to the fourth power:

Table 3 Re-evaluated experimental $\eta\rho pT$ data for natural gas H at 280 K

T K	p MPa	$p(280\,\mathrm{K}, \rho_{\mathrm{eos}})$ MPa	$\rho_{\mathrm{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\mathrm{eos}})$[a] μPa·s	$\eta(280\,\mathrm{K}, \rho_{\mathrm{eos}})$[a] μPa·s
280.12	0.096173	0.096132	0.74485	10.391[b]	10.387[b]
280.12	0.20018	0.20009	1.5546	10.413	10.409
280.11	0.30012	0.30000	2.3370	10.427	10.423
280.11	0.39810	0.39794	3.1080	10.440	10.436
280.10	0.50025	0.50007	3.9162	10.456	10.452
280.09	0.60028	0.60007	4.7119	10.471	10.468
280.09	0.70000	0.69977	5.5094	10.481	10.478
280.09	0.80021	0.79994	6.3150	10.499	10.496
280.08	0.89970	0.89943	7.1194	10.513	10.511
280.08	1.00170	1.00140	7.9484	10.532	10.529
280.08	1.2511	1.2507	9.9945	10.571	10.568
280.08	1.5012	1.5008	12.075	10.616	10.613
280.08	1.7462	1.7457	14.141	10.662	10.659
280.09	1.9982	1.9975	16.294	10.711	10.708
280.09	2.2493	2.2485	18.470	10.767	10.764
280.09	2.5276	2.5267	20.919	10.830	10.828
280.09	2.7614	2.7603	23.006	10.885	10.882
280.09	2.9993	2.9982	25.158	10.942	10.939
279.94	3.2462	3.2471	27.443	11.004	11.006
279.95	3.5044	3.5051	29.846	11.074	11.076
279.95	3.7405	3.7414	32.077	11.142	11.144
279.96	3.9960	3.9968	34.523	11.221	11.222
279.96	4.2461	4.2469	36.954	11.299	11.300
279.97	4.4958	4.4964	39.414	11.383	11.384
279.96	4.7432	4.7442	41.892	11.469	11.470
279.96	4.9978	4.9988	44.475	11.558	11.559
279.97	5.4951	5.4959	49.627	11.750	11.751
279.97	5.9892	5.9902	54.893	11.958	11.959
279.97	6.4978	6.4989	60.463	12.185	12.186
279.96	6.9907	6.9923	66.008	12.425	12.426
279.97	7.4914	7.4927	71.773	12.681	12.682
279.97	7.9948	7.9962	77.708	12.966	12.967
279.97	8.4881	8.4897	83.648	13.260	13.261
279.98	9.0301	9.0313	90.294	13.602	13.603
279.98	9.4894	9.4907	96.019	13.909	13.910
279.99	10.0024	10.0031	102.481	14.271	14.272
280.00	10.4896	10.4896	108.670	14.632	14.632
279.87	10.9988	11.0093	115.310	15.018	15.022
279.92	11.4839	11.4908	121.47	15.410	15.412
279.98	11.9962	11.9980	127.92	15.838	15.839
280.02	12.480	12.478	133.98	16.248	16.248

Table 3 (continued)

T K	p MPa	$p(280\,\mathrm{K}, \rho_{\mathrm{eos}})$ MPa	$\rho_{\mathrm{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\mathrm{eos}})^{\mathrm{a}}$ µPa·s	$\eta(280\,\mathrm{K}, \rho_{\mathrm{eos}})^{\mathrm{a}}$ µPa·s
280.04	13.016	13.011	140.63	16.716	16.715
280.06	13.450	13.443	145.92	17.104	17.102
280.08	13.989	13.980	152.36	17.592	17.589
279.93	14.536	14.545	158.97	18.154	18.156
279.93	15.032	15.042	164.60	18.607	18.609
279.91	15.485	15.498	169.61	19.019	19.022
280.10	15.986	15.971	174.67	19.440	19.437
280.09	16.474	16.460	179.71	19.896	19.893
279.96	16.987	16.993	185.01	20.384	20.386
279.99	17.551	17.553	190.35	20.892	20.893
279.99	17.994	17.996	194.42	21.304	21.304
279.99	18.488	18.490	198.78	21.742	21.742
279.97	18.995	19.001	203.13	22.198	22.199
279.95	19.485	19.495	207.17	22.632	22.634
279.96	20.064	20.072	211.70	23.125	23.127

[a] $U_{\mathrm{c,r}}(\eta) = 0.8\,\%$

[b] Influenced by slip

$$\eta(\tau,\delta) = \sum_{i=0}^{4} \eta_i(\tau)\,\delta^i, \qquad \delta = \frac{\rho}{\rho_{\mathrm{c},j}}, \qquad \tau = \frac{T}{T_{\mathrm{c},j}}, \tag{1}$$

$$\begin{aligned} &\text{with } j = \mathrm{H} \quad \text{for natural gas H,} \quad j = \mathrm{L} \quad \text{for natural gas L,} \\ &\text{and } \rho_{\mathrm{c,H}} = 208.45\ \mathrm{kg\cdot m^{-3}}, \qquad T_{\mathrm{c,H}} = 213.26\ \mathrm{K} \quad \text{for natural gas H,} \\ &\rho_{\mathrm{c,L}} = 186.21\ \mathrm{kg\cdot m^{-3}}, \qquad T_{\mathrm{c,L}} = 191.88\ \mathrm{K} \quad \text{for natural gas L,} \end{aligned}$$

where δ is the reduced density, whereas τ is the reduced temperature. The values of the critical densities $\rho_{\mathrm{c,H}}$ and $\rho_{\mathrm{c,L}}$ as well as of the critical temperatures $T_{\mathrm{c,H}}$ and $T_{\mathrm{c,L}}$ were determined using REFPROP (Lemmon et al. [26]). Weighting factors $w_i = 100\,\eta_{\mathrm{exp},i}^{-2}$ were applied in the multiple linear least-squares regression to minimize the weighted sum of squares $\sigma = \sum_i w_i(\eta_{\mathrm{cor},i} - \eta_{\mathrm{exp},i})^2$ 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 σ for each isotherm are given in Table 10 for natural gas H and in Table 11 for natural gas L.

Table 4 Re-evaluated experimental $\eta\rho pT$ data for natural gas H at 300 K

T K	p MPa	$p(300\,\mathrm{K}, \rho_{\mathrm{eos}})$ MPa	$\rho_{\mathrm{eos}}(T, p)$ $\mathrm{kg \cdot m^{-3}}$	$\eta(T, \rho_{\mathrm{eos}})^{\mathrm{a}}$ μPa·s	$\eta(300\,\mathrm{K}, \rho_{\mathrm{eos}})^{\mathrm{a}}$ μPa·s
300.07	19.863	19.852	182.30	20.818	20.816
300.01	19.351	19.350	178.34	20.449	20.448
300.00	19.004	19.004	175.54	20.201	20.201
300.00	18.500	18.500	171.37	19.843	19.843
299.99	17.974	17.975	166.90	19.457	19.458
299.98	17.486	17.488	162.65	19.100	19.101
299.98	17.002	17.005	158.32	18.749	18.750
299.97	16.504	16.508	153.78	18.394	18.395
299.97	15.997	16.000	149.04	18.025	18.026
299.97	15.490	15.493	144.20	17.671	17.672
299.97	14.996	14.999	139.41	17.324	17.325
299.97	14.463	14.466	134.14	16.954	16.955
299.97	13.998	14.001	129.49	16.637	16.638
299.97	13.500	13.502	124.44	16.298	16.299
299.98	13.005	13.006	119.366	15.970	15.970
299.97	12.495	12.497	114.129	15.650	15.651
299.96	11.9982	12.001	108.997	15.339	15.340
299.95	11.4909	11.4943	103.745	15.029	15.031
299.94	10.9880	10.9919	98.538	14.736	14.738
299.94	10.4970	10.5006	93.462	14.456	14.458
300.03	10.0029	10.0012	88.325	14.179	14.178
300.02	9.4957	9.4946	83.150	13.914	13.913
300.01	9.0001	8.9997	78.139	13.669	13.669
300.01	8.4987	8.4983	73.116	13.430	13.430
300.02	8.0022	8.0014	68.200	13.207	13.206
300.02	7.4999	7.4992	63.299	12.986	12.986
300.01	6.9969	6.9965	58.468	12.786	12.785
300.01	6.4993	6.4990	53.764	12.595	12.594
300.01	5.9956	5.9953	49.085	12.413	12.412
300.01	5.4946	5.4944	44.514	12.242	12.242
300.01	5.0013	5.0011	40.099	12.086	12.086
300.02	4.7494	4.7490	37.874	12.008	12.007
300.02	4.5006	4.5002	35.701	11.934	11.934
300.02	4.2460	4.2457	33.500	11.862	11.862
300.02	4.0000	3.9997	31.393	11.799	11.799
300.02	3.7498	3.7495	29.273	11.736	11.735
300.02	3.4992	3.4989	27.171	11.670	11.669
300.02	3.2489	3.2486	25.093	11.607	11.606
300.01	3.0002	3.0001	23.051	11.552	11.552
300.06	2.7467	2.7461	20.985	11.494	11.492
300.06	2.4965	2.4959	18.972	11.451	11.449

Table 4 (continued)

T K	p MPa	$p(300\,\mathrm{K}, \rho_{eos})$ MPa	$\rho_{eos}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{eos})$[a] μPa·s	$\eta(300\,\mathrm{K}, \rho_{eos})$[a] μPa·s
300.05	2.2500	2.2496	17.011	11.399	11.398
300.05	1.9986	1.9983	15.030	11.351	11.349
300.04	1.7500	1.7497	13.092	11.305	11.303
300.04	1.4992	1.4990	11.1564	11.260	11.259
300.03	1.2502	1.2501	9.2553	11.221	11.220
300.03	1.00043	1.00033	7.3675	11.180	11.179
300.03	0.90001	0.89992	6.6140	11.166	11.165
300.02	0.79941	0.79935	5.8626	11.154	11.153
300.02	0.69983	0.69978	5.1216	11.137	11.137
300.01	0.59996	0.59994	4.3818	11.125	11.124
300.01	0.50002	0.50000	3.6443	11.114	11.114
300.01	0.40006	0.40004	2.9097	11.099	11.099
300.01	0.29994	0.29993	2.1770	11.084	11.084
300.01	0.19962	0.19961	1.4458	11.070	11.070
300.00	0.10064	0.10064	0.72745	11.048[b]	11.048[b]

[a] $U_{c,r}(\eta) = 0.8\,\%$

[b] Influenced by slip

3 Discussion of the Results

Humberg et al. [3] recently stated that the experimental values published by Schley et al. [4] for methane are assumed to be about 0.15 % too high due to the use of the former reference value for the viscosity of argon given by Kestin and Leidenfrost [23] for the calibration of the viscometer used by Schley et al. The same calibration value was employed for the measurements of the two natural gas samples discussed herein. Therefore, the re-evaluated values reported in this work are expected to be about 0.15 % lower than the older ones. Figures 1 and 2 show deviations of the experimental viscosity data for the two samples natural gas H and L, respectively, given by Schley et al. from the re-evaluated viscosity data reported in this work.

In connection with Figs. 1 and 2, it has to be noted that the density values of the re-evaluated data were re-calculated from the experimental data for pressure and temperature employing the equation of state by Kunz and Wagner [6] via their implementation in REFPROP (Lemmon et al. [26]). Figure 1, which illustrates the deviations between the original Schley data and the re-evaluated data as function of

Table 5 Re-evaluated experimental $\eta\rho pT$ data for natural gas H at 320 K

T K	p MPa	$p(320\,\text{K}, \rho_{\text{eos}})$ MPa	$\rho_{\text{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\text{eos}})^{a}$ μPa·s	$\eta(320\,\text{K}, \rho_{\text{eos}})^{a}$ μPa·s
319.96	19.996	20.001	161.95	19.699	19.700
319.99	19.499	19.500	158.25	19.413	19.413
319.99	18.995	18.996	154.48	19.106	19.106
320.00	18.493	18.493	150.63	18.815	18.815
320.00	17.998	17.998	146.77	18.515	18.515
320.00	17.496	17.496	142.79	18.237	18.237
320.01	17.004	17.003	138.82	17.939	17.939
320.01	16.497	16.496	134.68	17.653	17.652
320.00	15.997	15.997	130.55	17.373	17.373
320.00	15.498	15.498	126.37	17.091	17.091
319.99	14.992	14.993	122.09	16.811	16.811
320.00	14.497	14.497	117.84	16.542	16.542
320.00	14.003	14.003	113.576	16.280	16.280
320.00	13.501	13.501	109.217	16.015	16.015
320.00	13.000	13.000	104.833	15.761	15.761
320.00	12.500	12.500	100.447	15.513	15.513
320.01	12.005	12.004	96.092	15.265	15.265
320.01	11.5007	11.5001	91.656	15.031	15.031
320.00	11.0053	11.0053	87.305	14.802	14.802
319.99	10.4946	10.4951	82.827	14.572	14.572
319.98	9.9878	9.9888	78.399	14.354	14.354
319.97	9.5046	9.5060	74.196	14.151	14.152
319.97	9.0035	9.0047	69.859	13.949	13.950
319.96	8.4887	8.4902	65.439	13.750	13.751
319.95	7.9876	7.9894	61.173	13.563	13.564
319.95	7.4953	7.4969	57.017	13.390	13.391
320.10	6.9684	6.9654	52.579	13.214	13.210
320.08	6.4852	6.4830	48.596	13.061	13.058
320.06	5.9931	5.9917	44.587	12.913	12.911
320.04	5.4864	5.4855	40.509	12.764	12.763
320.03	4.9816	4.9811	36.500	12.625	12.624
320.02	4.7470	4.7466	34.655	12.563	12.563
320.01	4.4957	4.4955	32.693	12.502	12.502
320.01	4.2479	4.2478	30.771	12.444	12.444
320.01	3.9997	3.9996	28.859	12.385	12.385
320.01	3.7508	3.7507	26.957	12.326	12.326
320.00	3.4923	3.4923	24.996	12.264	12.264
320.00	3.2449	3.2449	23.134	12.208	12.208
320.00	2.9977	2.9977	21.286	12.158	12.158
320.00	2.7474	2.7474	19.430	12.110	12.110
320.00	2.4981	2.4981	17.596	12.071	12.071

Table 5 (continued)

T K	p MPa	$p(320\,\text{K}, \rho_{\text{eos}})$ MPa	$\rho_{\text{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\text{eos}})^{a}$ μPa·s	$\eta(320\,\text{K}, \rho_{\text{eos}})^{a}$ μPa·s
319.99	2.2491	2.2491	15.779	12.024	12.025
319.99	1.9974	1.9975	13.956	11.978	11.978
319.99	1.7502	1.7503	12.180	11.936	11.937
320.00	1.4983	1.4983	10.3834	11.897	11.897
320.00	1.2516	1.2516	8.6387	11.858	11.858
320.00	1.00032	1.00032	6.8763	11.822	11.822
320.01	0.90004	0.90001	6.1766	11.810	11.810
320.02	0.79905	0.79900	5.4743	11.793	11.793
320.02	0.69914	0.69910	4.7821	11.781	11.780
320.03	0.60100	0.60094	4.1041	11.770	11.769
320.03	0.49595	0.49590	3.3809	11.760	11.759
320.03	0.39591	0.39587	2.6945	11.749	11.748
320.04	0.30029	0.30025	2.0405	11.732	11.731
320.04	0.19939	0.19937	1.3527	11.719	11.718
320.03	0.098437	0.098427	0.66670	11.699[b]	11.698[b]

[a] $U_{c,r}(\eta) = 0.8\,\%$

[b] Influenced by slip

density for the four isotherms of natural gas sample H, does not show constant values near to − 0.15 % as expected due to the re-calibration of the radius of the wire. The isotherm at 260 K is characterized by the largest oscillations in the deviations around − 0.15 %. The oscillations decrease with increasing temperature. The maximum and minimum values occur at $\rho \approx 75\,\text{kg·m}^{-3}$ and $\rho \approx 180\,\text{kg·m}^{-3}$, respectively. They are certainly caused by the change in the calculation of the densities from the ISO 12213 [5] to the equation of state by Kunz and Wagner.

The corresponding deviations between the original Schley data and the re-evaluated data for the natural gas sample L are displayed for the four isotherms in Fig. 2. The figure shows, similar to Fig. 1, also oscillations of the deviations around − 0.15 %. These oscillations are less at lower densities ($\rho < 100\,\text{kg·m}^{-3}$) and comparable at higher densities ($\rho > 120\,\text{kg·m}^{-3}$) compared with natural gas H. The oscillations in the deviations decrease again with increasing temperature.

Both figures indicate that the differences of the original Schley data and the re-evaluated data amount to extra ± 0.2 % due to the changed density calculation dependent on the density itself. Figures 1 and 2 show that the re-calibration of the

Table 6 Re-evaluated experimental $\eta\rho pT$ data for natural gas L at 260 K

T K	p MPa	$p(260\,\mathrm{K}, \rho_{\mathrm{eos}})$ MPa	$\rho_{\mathrm{eos}}(T, p)$ $\mathrm{kg{\cdot}m^{-3}}$	$\eta(T, \rho_{\mathrm{eos}})$[a] μPa·s	$\eta(260\,\mathrm{K}, \rho_{\mathrm{eos}})$[a] μPa·s
259.25	0.10208	0.10238	0.87717	10.368[b]	10.393[b]
259.24	0.20515	0.20576	1.7681	10.386	10.412
259.25	0.30014	0.30102	2.5939	10.397	10.422
259.27	0.40768	0.40886	3.5341	10.412	10.437
259.30	0.50242	0.50383	4.3671	10.428	10.451
259.26	0.63368	0.63557	5.5304	10.442	10.467
259.25	0.69939	0.70151	6.1160	10.449	10.474
259.25	0.80580	0.80827	7.0689	10.466	10.492
259.22	0.90237	0.90526	7.9399	10.477	10.503
259.21	1.00535	1.00864	8.8739	10.497	10.524
259.21	1.2472	1.2514	11.0899	10.541	10.568
259.20	1.4955	1.5007	13.399	10.579	10.606
259.20	1.7468	1.7528	15.772	10.644	10.671
259.20	2.0318	2.0390	18.510	10.695	10.722
259.19	2.2472	2.2554	20.611	10.746	10.773
259.22	2.5421	2.5512	23.532	10.815	10.841
259.20	2.7406	2.7508	25.533	10.866	10.893
259.24	2.9885	2.9993	28.060	10.932	10.958
259.28	3.2546	3.2659	30.817	11.003	11.028
259.31	3.5060	3.5179	33.465	11.086	11.109
259.32	3.7818	3.7947	36.425	11.174	11.197
259.33	3.9821	3.9957	38.607	11.233	11.256
259.34	4.2492	4.2637	41.562	11.335	11.357
259.35	4.4886	4.5040	44.255	11.412	11.434
259.21	4.7747	4.7950	47.573	11.524	11.550
259.22	5.0296	5.0512	50.546	11.626	11.653
259.24	5.5115	5.5354	56.300	11.830	11.856
259.24	6.0015	6.0284	62.344	12.058	12.083
259.24	6.5077	6.5381	68.786	12.313	12.339
259.22	7.0017	7.0365	75.277	12.585	12.612
259.22	7.5000	7.5387	82.001	12.889	12.916
259.23	7.9783	8.0204	88.614	13.187	13.213
259.13	8.4882	8.5409	95.923	13.544	13.574
259.15	9.0172	9.0742	103.556	13.930	13.958
259.17	9.5155	9.5762	110.852	14.321	14.349
259.20	10.0236	10.0875	118.357	14.740	14.767
259.22	10.4783	10.5454	125.11	15.113	15.140
259.25	10.9932	11.0632	132.75	15.584	15.610
259.42	11.5658	11.6246	140.97	16.116	16.136
259.45	11.9812	12.040	146.99	16.502	16.521
259.44	12.471	12.535	154.04	16.982	17.001

Table 6 (continued)

T K	p MPa	$p(260\,\mathrm{K}, \rho_{\mathrm{eos}})$ MPa	$\rho_{\mathrm{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\mathrm{eos}})^{a}$ μPa·s	$\eta(260\,\mathrm{K}, \rho_{\mathrm{eos}})^{a}$ μPa·s
259.43	12.991	13.061	161.37	17.502	17.522
259.44	13.516	13.590	168.53	18.040	18.059
259.38	13.998	14.084	175.01	18.540	18.561
259.38	14.494	14.585	181.36	19.032	19.053
259.37	15.019	15.117	187.85	19.577	19.598
259.36	15.480	15.583	193.31	20.049	20.071
259.35	15.983	16.093	199.05	20.556	20.579
259.34	16.510	16.628	204.80	21.080	21.102
259.18	16.961	17.114	209.80	21.560	21.588
259.20	17.514	17.669	215.27	22.082	22.109
259.22	18.026	18.183	220.10	22.571	22.598
259.23	18.514	18.675	224.52	23.034	23.060
259.24	19.018	19.182	228.90	23.491	23.517
259.22	19.510	19.684	233.05	23.950	23.977
259.22	20.048	20.228	237.35	24.434	24.461

[a] $U_{c,r}(\eta) = 1.0\,\%$

[b] Influenced by slip

radius of the vibrating wire results in a reduction in the viscosity of − 0.15 %, particularly at low densities.

In addition, a remark concerns the outliers in Figs. 1 and 2. The reason for these deviations are typing errors in Tables III and IV of Schley et al. [4]. In Table III (natural gas H) at the isotherm 260 K, the viscosity value at $\rho = 4.3077$ mol·L^{-1} ($\rho = 77.4$ kg·m^{-3}) should read 12.315 μPa·s instead of 12.335 μPa·s and in Table IV (natural gas L) at the isotherm 320 K, the viscosity value at $\rho = 5.3865$ mol·L^{-1} ($\rho = 99.3$ kg·m^{-3}) should read 15.910 μPa·s instead of 15.900 μPa·s.

Moreover, we compare our re-evaluated data with the extended corresponding states (ECS) model [27] implemented in REFPROP [26]. This is done in Figs. 3 and 4, which present deviations between the re-evaluated data for the two samples of natural gas H and L and the calculated values for the ECS model. The zero line corresponds to the ECS model developed by Ely and Hanley. The calculation of the ECS values results from the experimental temperature and pressure data. We note that for natural gas sample H as well as sample L no viscosity values applying the ECS model could be calculated at 260 K in low density ranges, for sample H at $4 < \rho/\mathrm{kg{\cdot}m^{-3}} < 84$ and for sample L at $2 < \rho/\mathrm{kg{\cdot}m^{-3}} < 103$, respectively. This is due to the retrograde behavior of the samples at low temperatures.

Table 7 Re-evaluated experimental $\eta\rho pT$ data for natural gas L at 280 K

T K	p MPa	$p(280\,\mathrm{K}, \rho_{\mathrm{eos}})$ MPa	$\rho_{\mathrm{eos}}(T, p)$ $\mathrm{kg \cdot m^{-3}}$	$\eta(T, \rho_{\mathrm{eos}})$[a] μPa·s	$\eta(280\,\mathrm{K}, \rho_{\mathrm{eos}})$[a] μPa·s
280.00	0.095008	0.095008	0.75525	11.077[b]	11.077[b]
279.93	0.19866	0.19871	1.5832	11.099	11.102
279.95	0.30067	0.30072	2.4015	11.119	11.121
279.97	0.39994	0.39998	3.2013	11.134	11.135
279.99	0.50193	0.50195	4.0266	11.148	11.149
280.01	0.60215	0.60213	4.8411	11.161	11.160
280.04	0.70640	0.70630	5.6918	11.174	11.172
280.06	0.80550	0.80532	6.5043	11.191	11.189
280.07	0.90389	0.90365	7.3148	11.208	11.205
280.07	1.00140	1.00113	8.1217	11.224	11.221
280.05	1.2549	1.2547	10.2372	11.268	11.266
280.05	1.5112	1.5109	12.400	11.311	11.309
280.05	1.7513	1.7509	14.449	11.358	11.357
280.04	1.9987	1.9984	16.585	11.404	11.403
280.04	2.2562	2.2558	18.833	11.458	11.457
280.03	2.4981	2.4978	20.970	11.509	11.508
280.02	2.7526	2.7524	23.244	11.565	11.564
280.02	3.0148	3.0146	25.613	11.619	11.618
280.01	3.2505	3.2503	27.768	11.680	11.680
280.01	3.5013	3.5011	30.086	11.747	11.747
280.00	3.7404	3.7404	32.321	11.812	11.812
279.99	4.0048	4.0050	34.821	11.882	11.882
279.98	4.2454	4.2458	37.123	11.952	11.953
279.98	4.5034	4.5038	39.616	12.027	12.028
279.97	4.7491	4.7498	42.019	12.103	12.104
279.97	5.0045	5.0052	44.542	12.187	12.188
279.97	5.5022	5.5030	49.538	12.359	12.360
279.97	5.9830	5.9839	54.463	12.530	12.531
279.98	6.4882	6.4888	59.735	12.730	12.731
279.99	6.9885	6.9888	65.056	12.936	12.936
279.99	7.4883	7.4887	70.471	13.155	13.155
279.99	8.0002	8.0006	76.108	13.398	13.398
280.00	8.4771	8.4771	81.431	13.628	13.628
280.00	9.0069	9.0069	87.426	13.905	13.905
280.01	9.4855	9.4849	92.895	14.170	14.169
280.01	9.9745	9.9738	98.534	14.444	14.443
280.02	10.4708	10.4694	104.285	14.742	14.741
280.03	10.9850	10.9828	110.265	15.058	15.057
280.04	11.4844	11.4813	116.078	15.377	15.376
280.05	11.9779	11.9737	121.810	15.707	15.705
280.06	12.482	12.477	127.64	16.039	16.037

Table 7 (continued)

T K	p MPa	$p(280\,\mathrm{K}, \rho_{\mathrm{eos}})$ MPa	$\rho_{\mathrm{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\mathrm{eos}})^{a}$ µPa·s	$\eta(280\,\mathrm{K}, \rho_{\mathrm{eos}})^{a}$ µPa·s
280.06	12.994	12.988	133.51	16.396	16.394
280.06	13.488	13.482	139.14	16.750	16.748
280.06	13.988	13.982	144.75	17.119	17.117
280.06	14.491	14.485	150.30	17.489	17.487
280.05	14.992	14.986	155.74	17.865	17.863
280.06	15.481	15.473	160.93	18.215	18.213
280.05	15.976	15.969	166.09	18.597	18.595
280.04	16.486	16.480	171.28	18.980	18.978
280.04	16.982	16.976	176.18	19.357	19.355
280.04	17.483	17.477	181.01	19.740	19.739
280.04	17.989	17.982	185.74	20.125	20.124
280.04	18.475	18.469	190.16	20.493	20.492
280.04	18.982	18.976	194.63	20.884	20.883
280.04	19.513	19.507	199.17	21.285	21.284
280.04	20.007	20.000	203.25	21.652	21.650

[a] $U_{c,r}(\eta) = 0.8\,\%$

[b] Influenced by slip

Figure 3, in which the deviations of the re-evaluated data for the natural gas sample H from the values calculated for the ECS model [27] are illustrated, makes evident that for the higher isotherms 280 K, 300 K, and 320 K the deviations are between +0.09 % at the lowest densities and ≈− 0.45 % up to high densities. For the isotherm 260 K, the largest deviation amounts to +0.84 % at $\rho = 85$ kg·m^{-3}. The deviations decrease with increasing density for this isotherm, pass through zero, and become negative with a minimum of − 0.45 % at $\rho = 181$ kg·m^{-3}. The deviations at $\rho > 181$ kg·m^{-3} decrease to − 0.15 % on the average, corresponding to the re-calibration of the wire.

Figure 4, which displays the deviations of the re-evaluated data for the natural gas sample L from the values calculated for the ECS model [27], shows that larger deviations occur compared to natural gas sample H. For the isotherms 280 K, 300 K, and 320 K, the deviations at the lowest densities amount to + 0.80 %, +1.23 %, and +1.44 %, respectively. They decrease with increasing density down to +0.60 % (280 K) and to ≈ 1.0 % (300 K and 320 K) at $\rho \approx 100$ kg·m^{-3}, followed by a further decrease to +0.67 % (320 K) and to +0.33 % (300 K) at the highest density each. The deviation of the 260 K isotherm at $\rho = 104$ kg·m^{-3} amounts to +0.63 %, whereas the further deviations and those for the isotherm 280 K show the similar course with increasing density. They pass through

Table 8 Re-evaluated experimental $\eta\rho pT$ data for natural gas L at 300 K

T K	p MPa	$p(300\,\text{K}, \rho_{\text{eos}})$ MPa	$\rho_{\text{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\text{eos}})$[a] µPa·s	$\eta(300\,\text{K}, \rho_{\text{eos}})$[a] µPa·s
299.90	0.12261	0.12266	0.91004	11.791[b]	11.795[b]
299.91	0.21131	0.21137	1.5707	11.807	11.810
299.91	0.30425	0.30434	2.2652	11.823	11.826
299.91	0.40345	0.40357	3.0089	11.837	11.839
299.91	0.50327	0.50342	3.7599	11.850	11.853
299.90	0.60661	0.60681	4.5402	11.865	11.868
299.91	0.71841	0.71863	5.3873	11.876	11.879
299.92	0.81927	0.81949	6.1542	11.891	11.894
299.92	0.90292	0.90317	6.7924	11.900	11.903
299.93	1.0155	1.01575	7.6540	11.919	11.921
299.93	1.2613	1.2616	9.5471	11.961	11.963
299.94	1.5072	1.5076	11.4575	12.006	12.008
299.93	1.7548	1.7553	13.398	12.041	12.044
299.94	2.0019	2.0023	15.349	12.090	12.092
299.97	2.2596	2.2599	17.401	12.131	12.132
299.98	2.5381	2.5383	19.639	12.188	12.188
299.98	2.7572	2.7574	21.415	12.224	12.224
299.99	3.0032	3.0033	23.424	12.280	12.280
300.00	3.2544	3.2544	25.492	12.336	12.336
300.00	3.5301	3.5301	27.783	12.395	12.395
299.97	3.7481	3.7485	29.612	12.443	12.444
299.97	3.9983	3.9988	31.723	12.504	12.505
299.97	4.2531	4.2536	33.891	12.565	12.566
299.97	4.5161	4.5167	36.146	12.633	12.634
299.97	4.7497	4.7503	38.164	12.701	12.702
299.98	4.9971	4.9976	40.316	12.767	12.767
299.99	5.4906	5.4908	44.656	12.911	12.912
300.00	5.9919	5.9919	49.127	13.062	13.062
300.00	6.4942	6.4942	53.669	13.226	13.226
300.00	6.9864	6.9864	58.176	13.395	13.395
300.00	7.4941	7.4941	62.881	13.576	13.576
299.98	8.0123	8.0130	67.742	13.765	13.766
299.99	8.4902	8.4906	72.260	13.963	13.964
300.01	8.9870	8.9866	76.990	14.163	14.163
300.02	9.4927	9.4918	81.842	14.375	14.375
300.02	9.9885	9.9875	86.631	14.598	14.597
300.01	10.4865	10.4859	91.468	14.825	14.825
300.01	11.0013	11.0007	96.478	15.077	15.076
300.02	11.5124	11.5111	101.451	15.327	15.327
300.02	12.008	12.007	106.278	15.576	15.576
300.00	12.498	12.498	111.052	15.834	15.834

Table 8 (continued)

T K	p MPa	$p(300\,\mathrm{K}, \rho_{\mathrm{eos}})$ MPa	$\rho_{\mathrm{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\mathrm{eos}})^{\mathrm{a}}$ µPa·s	$\eta(300\,\mathrm{K}, \rho_{\mathrm{eos}})^{\mathrm{a}}$ µPa·s
300.00	12.999	12.999	115.905	16.093	16.093
300.01	13.490	13.489	120.63	16.366	16.366
300.01	13.998	13.997	125.48	16.641	16.641
300.02	14.496	14.495	130.20	16.921	16.921
300.02	14.999	14.997	134.91	17.203	17.202
300.03	15.499	15.496	139.54	17.489	17.488
300.02	15.994	15.992	144.07	17.773	17.772
300.00	16.503	16.503	148.68	18.081	18.081
300.00	17.013	17.013	153.21	18.391	18.391
299.99	17.492	17.493	157.39	18.670	18.671
299.99	17.999	18.000	161.73	18.987	18.987
300.00	18.488	18.488	165.83	19.286	19.286
300.00	18.990	18.990	169.96	19.595	19.595
299.99	19.505	19.506	174.12	19.914	19.914
299.97	20.003	20.007	178.07	20.218	20.219

[a] $U_{\mathrm{c,r}}(\eta) = 0.8\,\%$

[b] Influenced by slip

zero at $\rho \approx 180$ kg·m^{-3} and become negative (− 0.13 % for 280 K and − 0.18 % for 260 K at the highest density each).

4 Conclusion

A new reference value for argon at 298.15 K and at zero density, which was derived by Vogel et al. [24] and further updated by Hellmann [25], was used to re-evaluate viscosity measurements of Schley et al. [4] for two natural gas samples H (highly caloric) and L (lowly caloric) for the four isotherms each. Schley et al. carried out relative measurements using a vibrating-wire viscometer and calibrated their measuring device employing a nowadays outdated reference value of Kestin and Leidenfrost [23].

Furthermore, Schley et al. [4] applied the international standard ISO 12213 [5] for calculating the densities from the experimental pressure and temperature data. In this work, the densities were computed employing the equation of state

Table 9 Re-evaluated experimental $\eta\rho pT$ data for natural gas L at 320 K

T K	p MPa	$p(320\,\text{K}, \rho_{\text{eos}})$ MPa	$\rho_{\text{eos}}(T, p)$ kg·m^{-3}	$\eta(T, \rho_{\text{eos}})$[a] μPa·s	$\eta(320\,\text{K}, \rho_{\text{eos}})$[a] μPa·s
320.03	0.10845	0.10844	0.75379	12.467[b]	12.466[b]
320.03	0.20925	0.20923	1.4564	12.488	12.487
320.03	0.30621	0.30618	2.1340	12.498	12.497
320.03	0.42389	0.42385	2.9589	12.511	12.510
320.04	0.53558	0.53551	3.7440	12.531	12.530
320.04	0.60543	0.60535	4.2362	12.534	12.532
320.02	0.69890	0.69885	4.8967	12.544	12.543
320.01	0.81154	0.81151	5.6947	12.562	12.562
320.00	0.90354	0.90354	6.3483	12.575	12.575
319.98	0.99951	0.99957	7.0321	12.582	12.583
319.98	1.2525	1.2525	8.8416	12.622	12.623
319.98	1.5045	1.5046	10.6564	12.660	12.660
319.99	1.7479	1.7479	12.420	12.695	12.695
319.99	2.0045	2.0046	14.292	12.737	12.737
320.00	2.2514	2.2514	16.104	12.783	12.783
320.01	2.5191	2.5191	18.082	12.825	12.825
320.02	2.7871	2.7870	20.074	12.874	12.873
320.02	3.0007	3.0005	21.673	12.915	12.915
319.99	3.2938	3.2939	23.881	12.963	12.963
319.99	3.5271	3.5273	25.649	13.013	13.013
319.99	3.7526	3.7527	27.367	13.061	13.061
319.99	4.0028	4.0029	29.283	13.117	13.117
320.00	4.2477	4.2477	31.169	13.164	13.164
320.00	4.4974	4.4974	33.103	13.223	13.223
320.01	4.7525	4.7523	35.088	13.288	13.287
320.02	4.9960	4.9956	36.993	13.339	13.339
320.01	5.5105	5.5103	41.055	13.471	13.470
320.01	6.0140	6.0138	45.068	13.602	13.602
320.01	6.5005	6.5002	48.982	13.732	13.731
320.02	6.9828	6.9822	52.892	13.875	13.874
320.02	7.5051	7.5045	57.163	14.036	14.035
320.02	8.0147	8.0140	61.359	14.192	14.192
320.01	8.4869	8.4865	65.277	14.345	14.345
320.01	8.9939	8.9935	69.501	14.519	14.519
320.01	9.5108	9.5104	73.829	14.698	14.698
320.01	10.0627	10.0622	78.467	14.900	14.899
320.01	10.4787	10.4782	81.973	15.058	15.058
320.01	11.0099	11.0093	86.455	15.260	15.260
319.98	11.5481	11.5492	91.014	15.474	15.475
319.97	11.9901	11.9918	94.750	15.653	15.653
319.98	12.536	12.537	99.345	15.885	15.886

Table 9 (continued)

T K	p MPa	$p(320\,\mathrm{K}, \rho_{eos})$ MPa	$\rho_{eos}(T, p)$ $\mathrm{kg \cdot m^{-3}}$	$\eta(T, \rho_{eos})^a$ μPa·s	$\eta(320\,\mathrm{K}, \rho_{eos})^a$ μPa·s
319.99	12.995	12.996	103.199	16.076	16.077
320.00	13.505	13.505	107.463	16.302	16.302
320.00	13.989	13.989	111.494	16.516	16.516
320.00	14.492	14.492	115.661	16.741	16.741
320.00	14.990	14.990	119.758	16.971	16.971
320.00	15.493	15.493	123.86	17.206	17.206
320.01	16.002	16.001	127.97	17.450	17.450
320.01	16.492	16.491	131.89	17.688	17.687
320.02	16.997	16.995	135.88	17.928	17.927
320.02	17.527	17.525	140.03	18.197	18.196
320.01	18.010	18.009	143.76	18.428	18.427
320.01	18.570	18.569	148.02	18.709	18.709
320.01	19.005	19.004	151.28	18.932	18.931
320.01	19.524	19.523	155.11	19.195	19.194
320.01	20.015	20.013	158.69	19.450	19.450

[a] $U_{c,r}(\eta) = 0.8\,\%$

[b] Influenced by slip

Table 10 Coefficients of Eq. 1 for the re-evaluated viscosity measurements on the sample of natural gas H

T K	n	ρ_{max} $\mathrm{kg \cdot m^{-3}}$	η_0 μPa·s	η_1 μPa·s	η_2 μPa·s
260	4	245.22	9.730 ± 0.003	3.916 ± 0.069	9.632 ± 0.305
280	4	211.70	10.403 ± 0.002	3.300 ± 0.038	11.024 ± 0.187
300	4	182.30	11.064 ± 0.001	3.392 ± 0.023	11.187 ± 0.130
320	4	161.95	11.715 ± 0.001	3.462 ± 0.030	11.393 ± 0.185
T K			η_3 μPa·s	η_4 μPa·s	σ
260			− 4.205 ± 0.453	3.096 ± 0.211	0.100
280			− 5.007 ± 0.317	3.093 ± 0.169	0.046
300			− 5.644 ± 0.252	3.491 ± 0.154	0.023
320			− 6.444 ± 0.398	4.056 ± 0.272	0.025

Table 11 Coefficients of Eq. 1 for the re-evaluated viscosity measurements on the sample of natural gas L

T K	n	ρ_{max} kg·m^{-3}	η_0 μPa·s	η_1 μPa·s	η_2 μPa·s
260	4	237.35	10.354 ± 0.001	2.683 ± 0.023	8.204 ± 0.093
280	4	203.25	11.077 ± 0.001	3.002 ± 0.024	7.505 ± 0.107
300	4	178.07	11.785 ± 0.001	2.965 ± 0.029	8.054 ± 0.149
320	4	158.69	12.462 ± 0.001	2.995 ± 0.023	7.958 ± 0.127
T K			η_3 μPa·s	η_4 μPa·s	σ
260			− 3.671 ± 0.125	1.869 ± 0.053	0.035
280			− 3.021 ± 0.166	1.606 ± 0.082	0.029
300			− 4.053 ± 0.261	2.121 ± 0.145	0.031
320			− 4.144 ± 0.247	2.312 ± 0.153	0.020

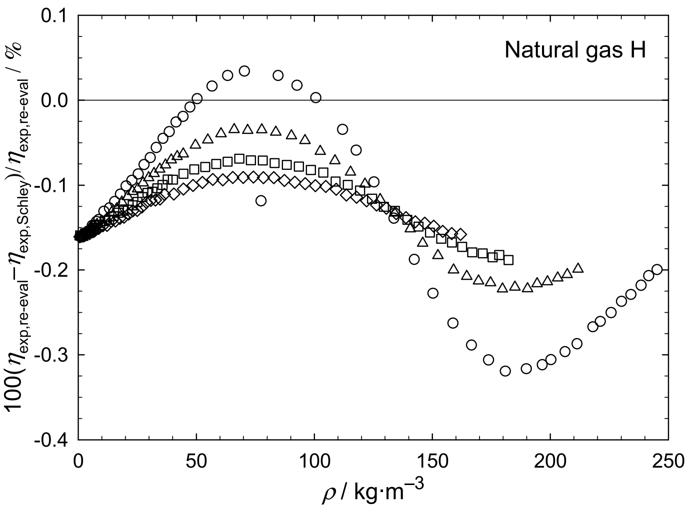

Fig. 1 Comparison of the re-evaluated viscosity data of this work $\eta_{exp,re-eval}$ for the sample natural gas H with the data $\eta_{exp,Schley}$ for the four isotherms, as a function of density ρ. Data: ○, 260 K; △, 280 K; □, 300 K; and ◇, 320 K

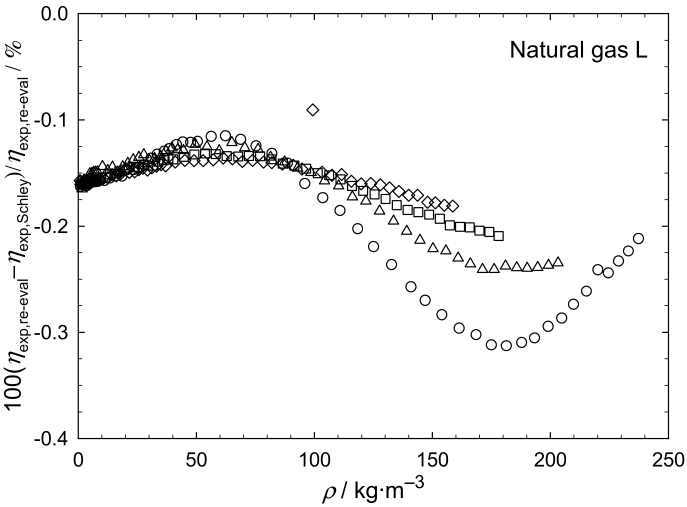

Fig. 2 Comparison of the re-evaluated viscosity data of this work $\eta_{exp,re-eval}$ for the sample natural gas L with the data $\eta_{exp,Schley}$ for the four isotherms, as a function of density ρ. Data: ○, 260 K; △, 280 K; □, 300 K; and ◇, 320 K

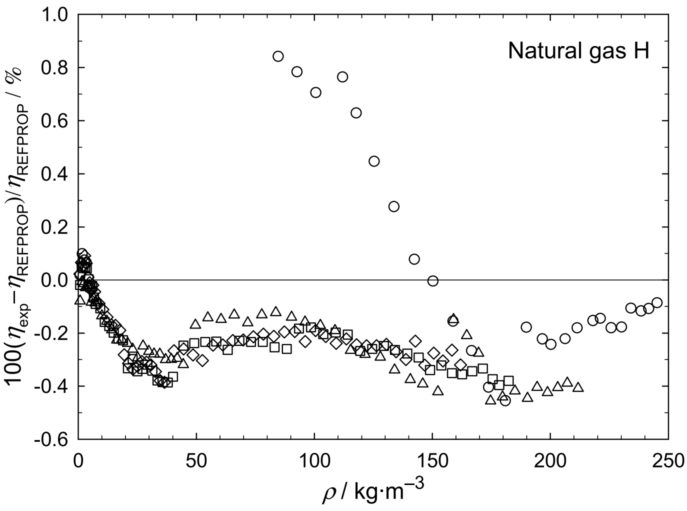

Fig. 3 Comparison of the re-evaluated viscosity data of this work η_{exp} for the sample natural gas H with values $\eta_{REFPROP}$ calculated from the approach of the ECS model developed by Ely and Hanley [27] using REFPROP [26] for the four isotherms, as a function of density ρ. Data: ○, 260 K; △, 280 K; □, 300 K; and ◇, 320 K

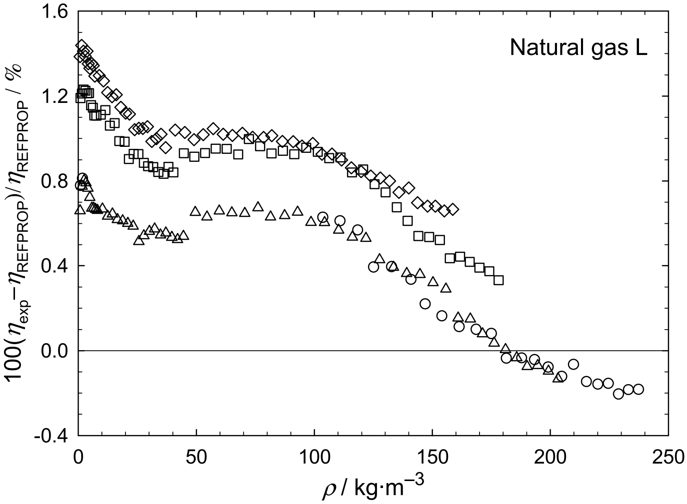

Fig. 4 Comparison of the re-evaluated viscosity data of this work η_{exp} for the sample natural gas L with values $\eta_{REFPROP}$ calculated from the approach of the ECS model developed by Ely and Hanley [27] using REFPROP [26] for the four isotherms, as a function of density ρ. Data: ○, 260 K; △, 280 K; □, 300 K; and ◇, 320 K

of Kunz and Wagner [6] from the pressure and temperature data reported by Schley et al.

The re-evaluated experimental data of each nominal isotherm for the two natural gas samples H and L were correlated as a function of the reduced density δ by means of a power-series representation restricted to the fourth power. Finally, the re-evaluated viscosity data were discussed in comparison with values calculated using the approach of the ECS model by Ely and Hanley [27] via its implementation in REFPROP [26]. The re-evaluated data of this work reported as $\eta\rho pT$ data should be employed to update the database for natural gases.

Author Contributions SH performed the calculations regarding the re-evaluation and prepared the tables and figures. All authors wrote and reviewed the manuscript.

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Declarations

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

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