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Dynamic Viscosity Measurements of Three Natural Gas Mixtures—Comparison against Prediction Models

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Abstract New and accurate viscosity measurements of three natural gas mixtures obtained directly at three different locations in the Norwegian gas transport network in the North Sea are presented. The viscosity measurements were performed using a vibrating-wire instrument with an uncertainty of ±1%, and covered a wide range of temperatures and pressures (263–303 K and 5.0–25 MPa). The predictive power of various models was also examined in relation to our new viscosity measurements. It was found that one of the Lee–Gonzalez–Eakin variants, as well as the NIST's SUPERTRAPP scheme, agreed very well with the present measurements. The correlative schemes reported by Schley et al. (Int J Thermophys 25:1623, 2004) and Vesovic–Wakeham (Int J Thermophys 22:415, 2001) were found to deviate significantly from the present measurements.

Keywords Measurement · Natural gas · Prediction · Vibrating-wire viscometer · Viscosity

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1 Introduction

Accurate prediction of gas viscosity is very important in modeling gas behavior at both micro and macro levels. While many measurements have been performed for artificial gas compositions of one to five components, very few measurements of real natural gases exist in the open literature. As a result, traditional viscosity prediction methods have not been tested extensively against such measurements. Efforts to develop a kinetic theory applicable to complex gas compositions have also failed so far. New and accurate viscosity measurements will consequently help to enhance knowledge and understanding in this field.

2 Experimental

The viscosity measurements were performed in the Thermophysical Properties Laboratory of Aristotle University at Thessaloniki, using a vibrating-wire viscometer because of its high precision and the availability of a full theoretical background for interpreting the results. The viscometer has already been employed for measuring the viscosity of gas mixtures of nitrogen with water, methane with water [1], refrigerant vapors [2], and a laboratory-prepared natural gas mixture [3]. As a result, the apparatus and methodology will only be briefly discussed here.

The viscometer, illustrated in Fig. 1, consists of a 7-μm diameter tungsten wire subject to a load of 1.5 g to keep it under constant tension. Two samarium-cobalt magnets provide a 1 T homogeneous magnetic field. Electrical connections to the wire are made through the upper chuck and the bottom weight. The viscometer is placed in a high-pressure vessel made of stainless steel for a maximum working pressure of 100 MPa. An ethylene glycol bath, externally regulated by a Julabo PID F81-MV circulator, keeps the temperature very stable. Two platinum resistance thermometers embedded along the pressure vessel wall measure the temperature, and were calibrated against a Tinsley Class 1 NPL thermometer with an uncertainty of ±20 mK. The pressure in the vessel filled from vacuum with natural gas is measured with a Druck PTX 520 pressure transducer with an uncertainty of ±0.0005 MPa.

Applying two pulses of opposite sign induces a symmetric oscillation in the wire. As a result, the wire performs a damped oscillating motion in the gas. This movement produces a voltage across the wire, which is amplified and sampled at a rate of 50 kHz by an analog-to-digital converter coupled to a computer. With a resolution of one part per 2×10^4, 3,000 data points are collected for every oscillation.

The instrument is filled with the natural gas sample at the highest pressure, and the temperature stabilized. A measurement is taken to yield 3,000 discrete points, and the pressure is lowered step-by-step. The procedure is repeated until the lowest pressure is reached. Starting at a new isotherm, the pressure is again raised to 25 MPa and the procedure repeated. Before and after each sample, the viscosity of nitrogen was measured to confirm that the viscometer was continuing to operate well.

The reproducibility and precision of the viscosity measurements is about ±0.1%. The viscosity value is also influenced by the logarithmic decrement in vacuum, which has been measured and found to be equal to 0.00068 ± 0.00001. This yields an error

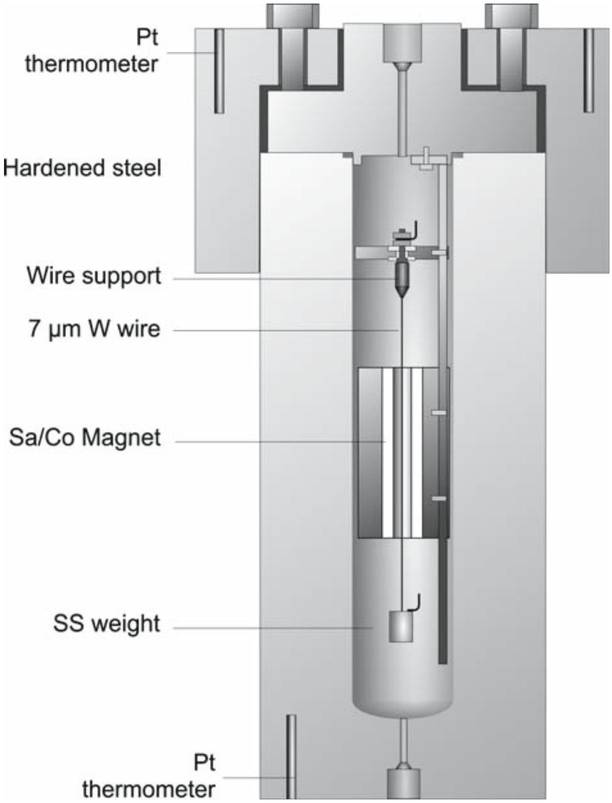

Fig. 1 Vibrating-wire viscometer

of ±0.04% in the viscosity. The uncertainty in the wire density evaluation is estimated to be ±0.2%, while the small uncertainty introduced by insufficiently accurate knowledge of the wire radius is minimized by measuring the viscosity of noble gases and nitrogen.

The temperature uncertainty is 0.04%, resulting from an uncertainty of ±20 mK in the calibration of the Pt thermometer. Pressure is measured with an uncertainty of ±0.0005 MPa, returning an uncertainty of ±0.25% for viscosity through the density calculation. Furthermore, the uncertainty in composition can result in a maximum error of ±0.3% in density, while the density correlation itself has an uncertainty of ±0.1–0.3%. Finally, the overall measurement uncertainty can be estimated to be about ±1%. Table 1 summarizes the uncertainty evaluations.

3 Measurement Results

The natural-gas samples were taken at three different locations in the Norwegian gas transport network in the North Sea, yielding three different compositions. The sample bottles were sent to E.ON Ruhrgas AG's central laboratory in Germany for

Table 1 Uncertainty evaluation of measurements

	Uncertainty (%)
Reproducibility/precision	±0.1
Log decrement	±0.04
Wire density	±0.2
Temperature	±0.04
Pressure	±0.25
Composition	±0.3
Density correlation	±0.1–0.3
Total measurement uncertainty	±1

Table 2 Composition of natural-gas samples

Components	Sample 1 (mol%)	Sample 2 (mol%)	Sample 3 (mol%)	Uncertainty (% rel)
Hydrogen	0.0000	0.0010	0.0005	
Helium	0.0068	0.0084	0.0168	
Water	0.0006	0.0000	0.0000	
Argon + oxygen	0.0514	0.0000	0.0000	1.5
Nitrogen	0.7587	0.6601	1.3916	1.5
Carbon dioxide	1.7947	2.1902	1.0030	1.5
Methane	90.1584	80.0079	92.2045	0.2
Ethane	6.3077	9.3063	4.3373	2.0
Propane	0.8010	4.9630	0.5396	2.5
n-Butane	0.0643	1.2791	0.0771	3
i-Butane	0.0446	0.7188	0.2562	3
n-Pentane	0.0044	0.2499	0.0198	5
i-Pentane	0.0054	0.2556	0.0468	5
neo-Pentane	0.0003	0.0055	0.0033	
Hexanes	0.0014	0.1793	0.0606	5
Heptanes	0.0005	0.1010	0.0364	15–20
Octanes	0.0001	0.0197	0.0038	15–20
Nonanes	0.0000	0.0086	0.0009	15–20
Decanes+	0.0000	0.0063	0.0002	15–20
Benzene	0.0002	0.0173	0.0007	10–15
Toluene	0.0000	0.0157	0.0006	10–15
Xylene	0.0000	0.0061	0.0003	10–15
Sum	100.0005	99.9998	100.0000	

composition analysis in a gas chromatograph. The resulting compositions are given in Table 2. It can be seen that Sample 1 has a medium methane content (about 90 mol%), Sample 2 has a low methane content (about 80 mol%), while Sample 3 has the highest methane content (about 92 mol%). The relative uncertainty in each component is also given in the same table. This uncertainty propagates to the uncertainty in the calculated density.

The density of the gas samples was calculated by using the AGA8-DX92 equation from Jaeschke and Schley [4]. The uncertainty of this equation is ±0.1% for the temperature range from 265 to 335 K at pressures below 12 MPa, rising to ±0.3% for pressures up to 25 MPa.

Table 3 Viscosity measurements of natural-gas sample 1

T (K)	P (MPa)	ρ ($\mathrm{kg \cdot m^{-3}}$)	η ($\mu\mathrm{Pa \cdot s}$)
263.345	24.820	265.12	31.58
263.345	22.490	252.46	29.63
263.390	20.110	236.97	26.94
263.394	17.490	215.70	24.07
263.394	15.260	192.62	21.49
263.400	12.490	156.21	17.95
263.400	10.010	118.22	15.01
263.410	7.520	80.94	13.05
263.410	4.730	45.47	11.43
278.200	24.150	238.99	28.09
278.200	22.580	229.42	26.89
278.200	20.010	211.25	24.19
278.145	17.570	190.43	21.80
278.145	14.990	163.82	19.50
278.070	12.280	131.36	16.60
278.070	9.950	101.68	14.60
278.020	7.500	71.75	13.25
278.000	5.170	46.13	12.10
283.450	25.020	236.42	27.93
283.455	22.490	221.06	26.10
283.455	20.010	203.20	23.85
283.480	17.600	182.55	21.43
283.488	14.970	156.00	18.75
283.489	13.100	134.81	17.40
283.489	11.555	116.46	15.94
283.430	7.690	71.20	13.36
283.828	4.460	37.78	11.96
288.260	25.110	230.37	27.43
288.260	22.490	214.26	25.38
288.250	19.720	193.98	22.94
288.240	17.310	173.13	20.69
288.241	15.180	152.07	18.99
288.241	12.500	122.76	16.50
288.190	9.990	94.26	14.70
288.185	7.580	67.83	13.50
288.170	5.170	43.60	12.50
303.425	24.910	210.07	25.20
303.420	22.620	195.69	23.75
303.416	18.980	168.89	20.63
303.196	17.580	157.46	19.66
303.107	15.100	135.15	18.07
303.054	12.680	111.78	16.51
303.054	10.000	85.33	14.89
303.045	7.620	62.49	13.70
303.045	5.130	40.13	12.70

For all samples, the viscosity was measured along five nominal isotherms: 263, 278, 283, 288, and 303 K. For each isotherm, the pressure ranged from 5 to 25 MPa at 2.5 MPa steps (Sample 1 and Sample 3). A total of 45 measurements were made for each of these samples. Sample 2 represented a slightly heavier gas and, in order to avoid liquid dropout, the pressure range was confined to 13–25 MPa in 2 MPa steps. Consequently, 35 measurement points were covered for this sample. The measurements are

Table 4 Viscosity measurements of natural-gas sample 2

T (K)	P (MPa)	ρ ($\mathrm{kg \cdot m^{-3}}$)	η ($\mu\mathrm{Pa \cdot s}$)
262.858	25.310	326.58	43.15
262.858	22.860	316.03	40.00
262.922	21.220	307.73	38.90
262.858	19.210	296.18	36.14
262.940	17.245	281.96	33.20
262.972	15.190	263.26	30.80
262.972	13.310	240.65	26.95
278.217	24.980	300.60	37.52
278.217	22.840	289.49	35.00
278.208	20.960	278.11	34.00
278.020	19.295	266.72	31.20
278.208	17.160	248.15	28.45
278.145	15.280	228.26	25.71
278.040	13.410	203.61	22.62
283.753	24.870	291.30	35.40
283.753	22.530	278.24	33.57
283.688	21.330	270.72	32.08
283.611	19.470	257.37	30.40
283.597	17.220	237.56	27.15
283.583	15.350	217.19	24.48
283.624	13.510	192.68	21.40
288.395	25.240	286.00	35.30
288.383	22.970	273.31	32.67
288.395	21.320	262.61	31.04
288.381	19.130	245.97	28.57
288.372	16.940	225.60	25.40
288.293	15.190	206.02	23.29
288.244	13.350	181.55	20.61
304.425	24.950	260.20	31.24
304.418	23.120	249.30	29.50
304.384	21.600	238.82	27.88
304.256	20.830	233.20	27.10
304.228	19.155	219.38	25.40
304.228	17.280	201.63	23.36

presented in Tables 3–5. The core measurements are presented—i.e., no smoothing procedure has been employed.

4 Comparison with Prediction Models

A number of different viscosity prediction models are found in the literature. Some aim to predict the viscosity for a narrow range of composition, pressure, and temperature, while others claim to be valid for a broader range of conditions. The new measurements are compared with prediction models claimed to be valid and accurate for natural gases. Recent models based mainly on theory are included based on their expected accuracy, whereas older empirical models are included since they basically require less computational effort.

The Lee–Gonzalez–Eakin (LGE) equation was presented by Lee et al. [5] and is named for the people responsible for it, namely, Lee, Gonzalez, and Eakin. It is a

Table 5 Viscosity measurements of natural-gas sample 3

T (K)	P (MPa)	ρ ($\mathrm{kg \cdot m^{-3}}$)	η ($\mu\mathrm{Pa \cdot s}$)
262.963	25.240	259.98	31.42
262.986	22.530	245.39	28.96
262.989	20.070	229.29	26.54
263.018	17.550	208.75	23.81
263.033	15.070	183.20	20.83
263.025	12.460	149.73	17.61
263.016	10.010	114.05	14.97
263.032	7.550	79.03	12.99
263.023	5.030	47.84	11.61
279.134	25.120	235.89	28.08
279.146	22.610	221.02	25.85
279.117	19.870	201.65	23.46
279.084	17.470	181.28	21.30
279.081	14.610	153.40	18.46
279.069	12.390	126.82	16.38
279.093	10.190	100.42	14.68
279.098	7.530	69.59	13.04
279.120	5.030	43.40	11.99
283.720	24.870	228.24	27.05
283.565	22.590	214.63	25.40
283.825	19.970	195.61	22.86
283.596	17.620	176.21	20.70
283.541	15.200	152.77	18.70
283.498	12.650	125.05	16.41
283.589	10.140	96.33	14.57
283.538	7.620	68.50	13.14
283.525	4.990	42.40	12.07
288.192	25.230	224.36	26.62
288.188	22.610	208.48	24.38
288.174	20.040	190.20	22.50
288.246	17.470	168.67	20.30
288.307	15.160	146.58	18.22
288.334	12.410	117.67	15.98
288.343	10.060	92.11	14.48
288.337	7.410	64.29	13.15
288.292	4.960	40.67	12.17
303.198	25.120	205.50	24.64
303.212	22.630	190.15	22.70
303.244	19.980	171.50	20.95
303.225	17.410	151.09	19.20
303.242	15.000	130.75	17.43
303.397	12.570	107.20	15.91
303.569	10.090	84.55	14.45
303.607	7.640	60.99	13.55
303.636	4.950	38.51	12.57

semi-empirical scheme based on a database of 3,000 viscosity measurements of gas hydrocarbon mixtures. The LGE equation contains nine different parameters and two different parameter sets. These have been named LGE-1 and LGE-2. Both equations cover a range from 0.7 to 55 MPa, with an uncertainty of about ±3%. A variant of LGE-2 with an extended number of decimals was recently published in Whitson and Brule [6]. This is referred to as LGE-3. Another parameter set is suggested by Londono et al. [7]. These parameters are based on an extended set of 5,000 viscosity and density measurements, and resulted in an uncertainty of ±2.3% in the viscosity calculation. This version is referred to as LGE-4. Necessary inputs to the LGE equation are composition, temperature, and density.

The Lucas model is based on the corresponding-states principle and is not limited to hydrocarbon gases. It was published in the 1980s, and Poling et al. [8] present it as a model for predicting viscosity of gas mixtures at high pressure. Lucas' scheme requires composition, specific component parameters, temperature, and pressure as inputs.

SUPERTRAPP is a computer program developed by the National Institute of Standards and Technology (NIST) [9] which can be used to calculate the thermodynamic and transport properties of pure fluids and fluid mixtures. The whole scheme is based on the corresponding-states principle coupled with an extended set of shape factors produced by comparison with experiments. The SUPERTRAPP package produces the viscosity of natural gas mixtures with an uncertainty of about ±3% to ±4%. Necessary input parameters are composition, temperature, and pressure. The density is calculated by the program using a Peng–Robinson equation of state.

The Vesovic–Wakeham methodology is presented in detail by Vesovic [10]. The methodology acts as an interpolator between the viscosities of each pure component, and therefore relies heavily on the equation used to calculate the viscosity for methane. Two different methane equations were employed here—the one by Friend et al. [11] (prediction calculations are designated VW-F) and the other by Schley et al. [12] (calculations designated VW-S). All the calculations were performed by Vesovic. Following a sensitivity analysis, it was decided to represent the different compositions as a mixture of methane, ethane, propane, *n*-butane, iso-butane, nitrogen, carbon dioxide, and a C_{5+} fraction. The Vesovic–Wakeham calculations are only available for Samples 2 and 3. Input parameters to the model are composition, a viscosity equation for each gas component, temperature, and density.

The measurements were also compared to viscosity values calculated using the correlation scheme presented by Schley et al. [12]. This is a theoretically based scheme, developed recently, and tuned against the authors' own natural gas viscosity measurements. Necessary input parameters are composition, specific component parameters, temperature, and density.

5 Discussion

Figures 2–4 present the percentage deviations of the measured viscosity and the predicted viscosity values for the aforementioned prediction models for a typical isotherm (283 K). The average absolute deviation (AAD) is shown in Table 6, while the maximum deviation is shown in Table 7.

It is clear that the LGE-3 equation and the SUPERTRAPP model perform well for all the samples and across the whole range of pressure and temperature. For LGE-3, the AAD's are 1.6, 1.2, and 0.8% for Samples 1, 2, and 3, respectively. For SUPERTRAPP, the equivalent numbers are 2.1, 2.3, and 1.4%. As discussed above, the LGE equation is an older and simple correlation, empirical, based on many viscosity mea-

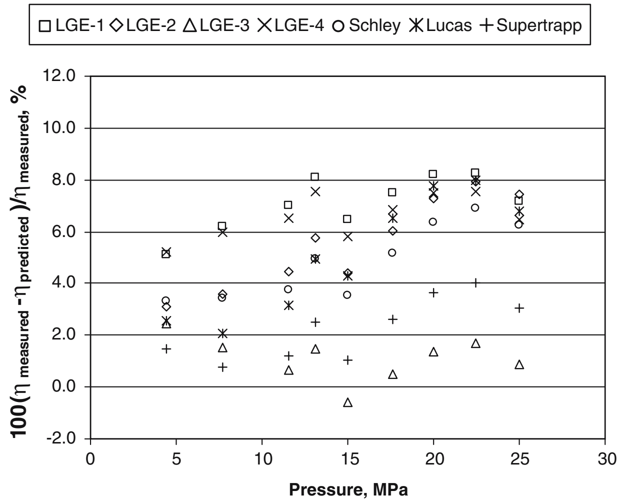

Fig. 2 Percentage deviations, $100(\eta_{\text{measured}} - \eta_{\text{predicted}})/\eta_{\text{measured}}$, of the experimental viscosity data of Sample 1 from other prediction models, at 283 K

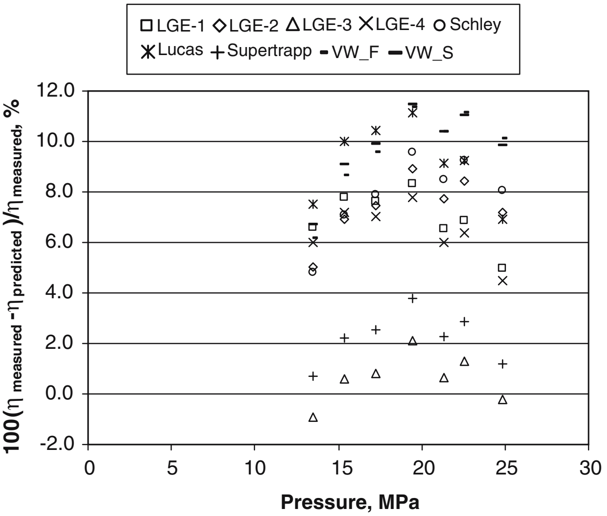

Fig. 3 Percentage deviations, $100(\eta_{\text{measured}} - \eta_{\text{predicted}})/\eta_{\text{measured}}$, of the experimental viscosity data of Sample 2 from other prediction models, at 283 K

surements, and derived to perform well across the large range of compositions, pressures, and temperatures present here. It is also seen that the decimal variant (LGE-3) performs considerably better than the other variants of the LGE formula investigated. The SUPERTRAPP model, based on corresponding-states principles and corrected with experimental data, seems to perform equally well.

However, the other correlations/prediction models investigated clearly underpredict the viscosity measurements presented in this work, and the deviations are significant

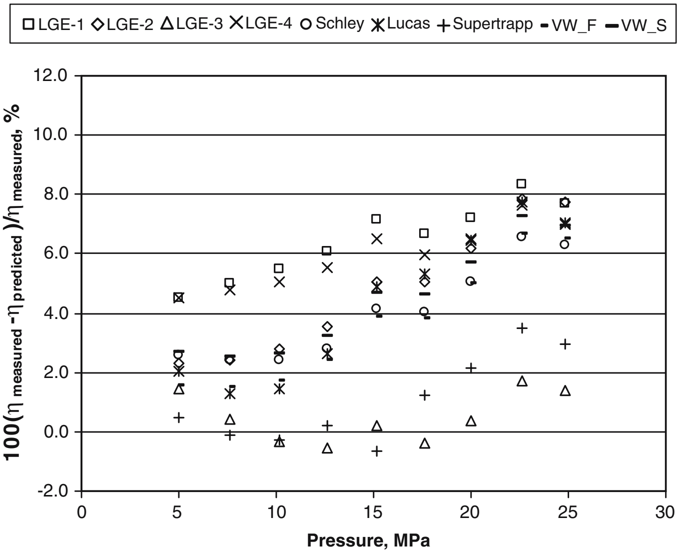

Fig. 4 Percentage deviations, $100(\eta_{\text{measured}} - \eta_{\text{predicted}})/\eta_{\text{measured}}$, of the experimental viscosity data of Sample 3 from other prediction models, at 283 K

Table 6 Average absolute deviation (AAD) in %, across all isotherms

	LGE-1	LGE-2	LGE-3	LGE-4	Schley	Lucas	SUPERTRAPP	VW-F	VW-S
Sample 1	6.91	5.16	1.63	6.39	4.57	4.75	2.08		
Sample 2	7.03	7.43	1.20	6.47	8.63	8.78	2.34	9.99	10.10
Sample 3	6.47	4.64	0.82	5.94	4.04	4.28	1.37	3.67	4.45

Table 7 Maximum deviation in %, across all isotherms

	LGE-1	LGE-2	LGE-3	LGE-4	Schley	Lucas	SUPERTRAPP	VW-F	VW-S
Sample 1	8.52	8.07	3.63	7.57	6.89	8.01	4.27		
Sample 2	10.33	9.82	2.81	9.50	10.98	11.30	4.49	13.04	12.76
Sample 3	9.73	8.14	1.99	8.78	7.54	8.69	3.48	8.11	8.26

in some cases. While all the schemes seem to converge at low pressures, deviations generally increase with pressure—which accords in essence with expectations.

Furthermore, the deviations are larger for Sample 2, which has a methane content of 80 mol%, lower than for the other samples. This corresponds well with the fact that most of the existing natural-gas viscosity measurements have a higher methane content, typically around 90 mol%. Prediction models have not been extensively verified against measurements from natural-gas samples with such a low methane content. It is particularly noted that the Sample 2 composition deviates considerably from the two gas compositions employed by Schley et al. [12]. Their heaviest gas contained around 84 mol% methane, 3.4 mol% ethane, 0.6 mol% propane, and 10 mol% nitrogen, while

Sample 2 contained 80 mol% methane, 9 mol% ethane, 5 mol% propane, and only 0.7 mol% nitrogen.

However, it should be pointed out that the deviations between our measurements and those reported by Schley et al., which were also obtained in a vibrating-wire viscometer and employed the same density equation, far exceed the quoted mutual uncertainties. Viscosity measurements in the two laboratories have agreed very well in the past, see, for example, Assael et al. [3] where measurements on an artificial natural gas mixture are seen to be predicted well by the Vesovic–Wakeham model in the investigated pressure range up to 15 MPa. The deviations reported here therefore needs further work to be explained, and more measurements on real gas samples would probably be particularly useful.

6 Conclusion

The extensive set of dynamic viscosity measurements of real natural-gas samples presented in this article makes a good contribution to the development of more accurate viscosity prediction models for gas mixtures at high pressure. The three compositions covered a wide range of pressures and temperatures.

The predictive power of various models was also examined in relation to our new viscosity measurements. It was found that one of the Lee–Gonzalez–Eakin variants, the LGE-3 scheme, and the NIST SUPERTRAPP scheme agreed very well with the present measurements. The correlative schemes reported by Schley et al. [12] and Vesovic [10] were found to deviate significantly from the present measurements.

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