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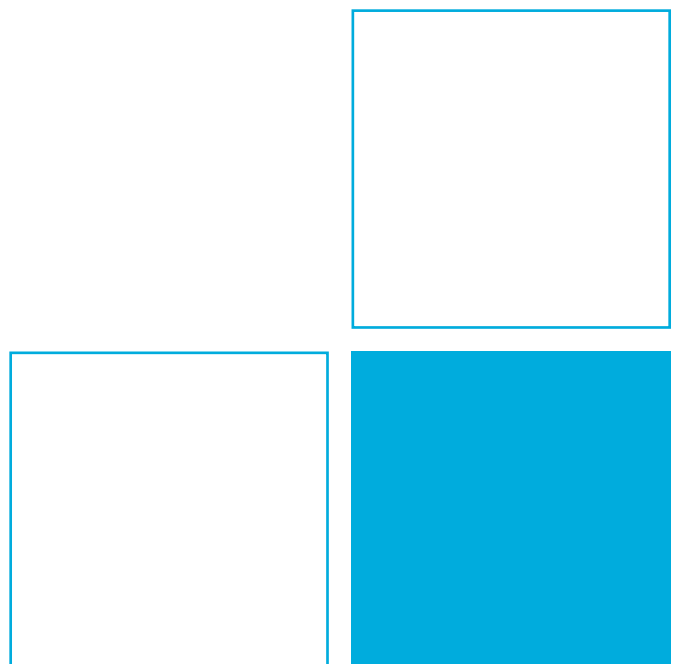
## Evaluation of a modified Hering – Zipperer method and the method by Schley et al. to compute the dynamic viscosity of natural gases

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# Evaluation of a modified Hering – Zipperer method and the method by Schley et al. to compute the dynamic viscosity of natural gases

A modified Hering-Zipperer method (H-Z-mod) is still used to calculate the viscosity of natural gas during calibrations. The method can calculate the dynamic viscosity of gas mixtures with a choice of 40 components. In the absence of literature references the method is re-documented in this paper. In addition, the H-Z-mod method has been compared with the RefProp software [3] in the range of -5 ~ 50°C and 1 ~ 101 bar. Using different natural gases, the deviations range between -6% and +7% depending on the gas composition. The viscosity calculation method based on Schley et al. [4] implemented in GasCalc [6], shows deviations ranging between -0.3% and 1.6%. The latter method to be used in the new Closed Loop Pigsar facility, is an order of magnitude more accurate than H-Z-mod. For both methods addition of 20% hydrogen results in an offset of more than 1% and a dispersion increase by more than a factor two.

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## Introduction

A so-called modified Hering-Zipperer method (H-Z-mod), can calculate the dynamic viscosity of gas mixtures with a choice of 40 components. The method was implemented in software available at PTB, e.g. in the control software used for calibrations with the High-Pressure Piston Prover at pigsar [2]. Unfortunately, a literature search did not result in any references to the H-Z-mod method. The documentation and source code of the software give a description of the method, however, without any references to the open literature. Below follows a description of the method.

## The algorithm

In their paper [1] Hering and Zipperer describe the mixture viscosity  $\eta_{mix}$  as the sum of the components' partial viscosities  $\eta_i$  weighted with the molar fractions  $x_i$  and the square root of the molar mass  $M_i$  and critical temperature  $T_{cr,i}$ .

$$\eta_{mix} = \frac{\sum x_i \eta_i \sqrt{T_{cr,i} M_i}}{\sum x_i \sqrt{T_{cr,i} M_i}} \quad (1)$$

The partial viscosities  $\eta_i$  are diluted gas viscosities converted to 0°C. The use of the critical temperature is based on the corresponding state principle, which means that all fluids behave similarly when described in terms of their reduced pressure and temperature.

In order to describe the temperature dependency of the viscosity a Sutherland constant  $C_S$  is introduced [2]. This parameter has the dimension of a temperature. The average  $C_S$  is derived from the  $C_{S,i}$  of the gas components  $i$ .

$$C_S = \sum x_i C_{S,i} \quad (2)$$

Table 1: Values of the Sutherland constants  $C_S$  [K], diluted viscosity  $\eta_i$  [ $\mu\text{Pa}\cdot\text{s}$ ], critical temperature  $T_{cr}$  [K] and molar mass  $M$  [kg/kmol] used in the modified Hering-Zipperer equation for determining the dynamic viscosity of a gas mixture.

Index	Component	$C_S$ [K]	$\eta_i$ [ $\mu\text{Pa}\cdot\text{s}$ ]	$T_{cr}$ [K]	$M$ [kg/kmol]
1	CO2	273	13.83	304.127	44.01
2	N2	102	16.58	126.24	28.0135
3	O2	125	19.23	154.58	31.9988
4	H2	72	8.44	32.976	2.0159
5	HE	83	18.5	5.1895	4.0026
6	NE	61	29.8	44.448	20.1797
7	AR	153	20.93	150.725	39.948
8	CO	102	16.58	132.92	28.01
9	H2O	650	8.75	647.13	18.0153
10	H2S	331	11.68	373.53	34.082
11	-	0	0	0	0
12	-	0	0	0	0
13	-	0	0	0	0
14	CH4	164	10.2	190.555	16.043
15	C2H6	252	8.6	305.33	30.07
16	C3H8	278	7.5	369.82	44.097
17	i-C4H10	330	6.9	408.13	58.123
18	n-C4H10	358	6.9	425.16	58.123
19	ne-C5H12	383	6.2	433.75	72.15
20	i-C5H12	383	6.2	460.39	72.15
21	n-C5H12	383	6.2	469.65	72.15
22	i-C6H14	436	5.9	499.93	86.177
23	n-C6H14	436	5.9	507.4	86.177
24	n-C7H16	490	4.99	540.2	100.204
25	n-C8H18	450	5.5	568.76	114.232
26	C9H20	450	5.5	594.56	128.259
27	C10H22	450	5.5	617.4	142.285
28	C11H24	450	5.5	638.73	156.312
29	C6H6	448	6.81	562.16	78.114
30	C7H8	400	6.79	591.72	92.141
31	C8H10	350	6.77	617.09	106.167
32	c-C5H10	400	6.5	511.6	70.134
33	c-C6H12	450	5.9	553.4	84.161
34	C7H14	450	5.5	572.12	98.188
35	C2H4	225	9.42	282.344	28.054
36	C3H6	322	7.75	365	42.081
37	1-C4H8	310	6.89	419.6	56.108
38	1-C5H10	400	6.5	464.74	70.134
39	C2H2	215	9.57	308.33	26.038
40	1,2-C4H6	300	6.88	449.3	54.092
41-52		0	0	0	0

Table II: Compositions in mol% of natural gases that are characteristic for the OGE network.

Component	NL_H-gas	NL_L-gas	No-gas	Ru-gas	Bio-gas
methane	88.853	84.343	90.60	96.24	96.15
nitrogen	3.220	9.494	0.88	0.41	0.75
CO <sub>2</sub>	1.208	1.504	1.80	0.34	2.90
ethane	5.078	3.874	5.78	2.71	
propane	1.133	0.512	0.68	0.20	
butane	0.221	0.089	0.09	0.03	
isobutane	0.154	0.085	0.10	0.05	
pentane	0.037	0.021	0.02	0.00	
isopentane	0.046	0.026	0.02	0.01	
hexane	0.050	0.052	0.02	0.01	
oxygen					0.20
neopentane			0.01		

For each component  $\eta_i$ ,  $T_{cr,i}$ ,  $M_i$  and  $C_{S,i}$  are listed in Table I. The index number has no relationship with ISO 6976 (any year) or any known compressibility algorithms. Compared with RefProp [3], the values of  $T_{cr}$  agree within 0.1% for common natural gas components.  $M_i$  values agree within 0.003% and  $\eta_i$  agrees for natural gas components up to hexane within 4%.

The temperature dependency of the viscosity is expressed by the following equation [2], in which  $t_c$  is the Celsius temperature of the gas mixture.

$$\eta_0 = \left(\frac{t_c}{T_0} + 1\right)^{3/2} \eta_{mix} \frac{T_0 + C_S}{T_0 + C_S + t_c} \quad (3)$$

Herning and Zipperer [1] do not describe a pressure dependency of the viscosity. This dependency is described by the following multiplication factor  $z_a$ .

$$z_a = a_0 + a_1 t_c + a_2 p_b t_c + a_3 p_b \quad (4)$$

where  $p_b$  is the absolute pressure in bar,  $a_0 = 0.91690348$ ,  $a_1 = 0.00042207 \text{ }^\circ\text{C}^{-1}$ ,  $a_2 = -0.00002207 \text{ }^\circ\text{C}^{-1} \text{ bar}^{-1}$  and  $a_3 = 0.00434531 \text{ bar}^{-1}$ .

The pressure and temperature dependent viscosity of the gas is now described by equation (3) multiplied by the maximum of equation (4) and 1.

$$\eta_{HZ}(p_b, t_c) = \max(1, z_a) \cdot \eta_0 \quad (5)$$

## Evaluation of the H-Z-mod method

The operating range and the accuracy of this Herning-Zipperer Mod algorithm was neither documented in the spreadsheet nor in the comments of the source code [2]. In order to determine the accuracy a systematic comparison with RefProp [3] was performed using natural gas compositions that are characteristic for the OGE network. These are listed in Table II.

For NL\_L-gas the relative deviation  $\eta_{HZ}/\eta_{RP} - 1$  is shown in Figure 1 in the temperature range of  $-5 \sim 50^\circ\text{C}$  and the pressure range of  $1 \sim 101$  bar. The deviations range between  $-3\%$  and  $+7\%$ . At pressures below 41 bar there is

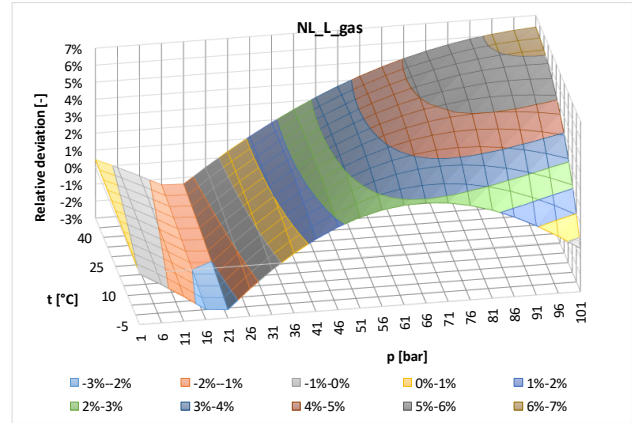


Figure 1: Relative deviation  $\eta_{HZ}/\eta_{RP} - 1$  [%] of the H-Z-mod versus RefProp as a function of temperature  $t$  [ $^\circ\text{C}$ ] and pressure  $p$  [bar] for low calorific Dutch gas.

hardly any temperature dependence of the deviation. The other gases show a similar behaviour. Table III shows the average deviation and its double standard deviation, the minimum and the maximum. If 20% hydrogen is added to NL\_H-gas the dispersion of values is almost twice the dispersion of natural gases.

## Evaluation of the viscosity calculations according to Schley et al.

A second method used in pigsar for the calculation of viscosities is a method developed by Schley et al. [4]. This method uses the mass density of the gas mixture which is obtained using the AGA-8 algorithm [5]. This method has been implemented in the GasCalc 2.6 software [6] and will be used in the control software of the new Closed Loop

Table IV: Deviations of GasCalc versus RefProp  $\eta_{GC}/\eta_{RP} - 1$  for different gases. The average (Avg), the double standard deviation (2-s), the minimum (min) and the maximum (max) are shown. H2NL-Hmix is NL\_H-gas with 20% hydrogen added.

	Avg	2-s	Min	Max
NL_H-gas	0.37%	0.46%	0.02%	1.00%
NL_L-gas	1.25%	0.36%	0.93%	1.59%
No-gas	0.32%	0.55%	-0.28%	0.96%
Ru-gas	0.20%	0.31%	-0.20%	0.64%
Biogas	0.25%	0.54%	-0.25%	0.88%
H2NL-Hmix	2.09%	1.38%	1.37%	3.92%

Table III: Deviations of H-Z-mod versus RefProp  $\eta_{HZ}/\eta_{RP} - 1$  for different gases. The average (Avg), the double standard deviation (2-s), the minimum (min) and the maximum (max) are shown. H2NL-Hmix is NL\_H-gas with 20% hydrogen added.

	Avg	2-s	Min	Max
NL_H-gas	0.05%	3.96%	-5.62%	3.19%
NL_L-gas	1.86%	4.72%	-2.51%	6.30%
No-gas	-0.36%	3.63%	-5.97%	2.94%
Ru-gas	0.55%	4.30%	-3.07%	4.38%
Biogas	1.53%	4.99%	-2.75%	5.69%
H2NL-Hmix	3.19%	9.03%	-3.30%	10.03%

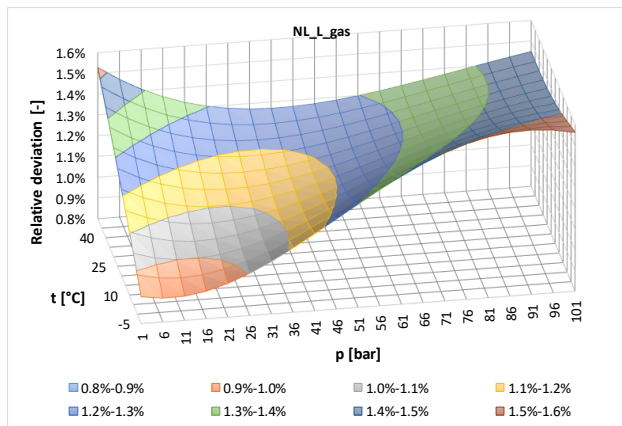


Figure 2: Relative viscosity deviation  $\eta_{GC}/\eta_{RP} - 1$  [%] of the GasCalc versus RefProp calculation as a function of temperature  $t$  [°C] and pressure  $p$  [bar] for low calorific Dutch gas.

Pigsar (CLP) test facility.

The method by Schley et al. [4] is also compared with RefProp [3] in the same way as has been done for H-Z-mod. Figure 2 displays the relative deviation  $\eta_{GC}/\eta_{RP} - 1$  between the GasCalc and RefProp calculations based on low calorific Dutch gas.

In the temperature range of  $-5 \sim 50^\circ\text{C}$  and the pressure range of  $1 \sim 101$  bar, the deviations range between 0.9% and 1.6%, which is an order of magnitude better than the deviation of the H-Z-mod algorithm. Table IV shows the average deviation and its double standard deviation, the minimum and the maximum. The high-calorific gases show a smaller average deviation than the low-calorific gas. The 2s values have the same magnitude. If 20% hydrogen is added to NL\_H-gas the dispersion of values is more than twice the dispersion of natural gases.

In general, the viscosities from the GasCalc calculations are more accurate than the viscosities calculated from H-Z-mod.

## Conclusion

In this paper the modified Hering-Zipperer method for calculating dynamic viscosities has been redocumented and evaluated using the RefProp software [3] in the range of  $-5 \sim 50^\circ\text{C}$  and  $1 \sim 101$  bar. Using different natural gases, the deviations range between  $-6\%$  and  $+7\%$  depending on the gas composition.

The GasCalc calculations based on Schley et al. [4] are an order of magnitude more accurate than H-Z-mod. The observed deviations range between  $-0.3\%$  and  $1.6\%$ .

For both methods addition of 20% hydrogen results in an offset of more than a percent and a dispersion increase by more than a factor two. Except for hydrogen enriched gas, the viscosity calculation method by Schley et al. [4] is within  $-0.3\%$  and  $+1.6\%$  of RefProp 10.0. For the calculation of Reynolds numbers this is adequate.

## Symbol list

$a_{0..3}$	constant	[...]
$C_S$	Sutherland constant	[K]
$i$	component number	[-]
$M$	Molar mass	[kg/kmol]
$p_b$	pressure	[bar]
$T_0$	thermodynamic temperature at $0^\circ\text{C}$	[K]
$T_{cr}$	critical temperature	[K]
$t_c$	temperature	[°C]
$x$	molar fraction	[-]
$z_a$	constant	[-]
$\eta_0$	dynamic viscosity at $t_c$	[ $\mu\text{Pa}\cdot\text{s}$ ]
$\eta_i$	diluted dynamic viscosity at $0^\circ\text{C}$	[ $\mu\text{Pa}\cdot\text{s}$ ]
$\eta_{mix}$	mixture viscosity at $0^\circ\text{C}$	[ $\mu\text{Pa}\cdot\text{s}$ ]
$\eta_{HZ}$	viscosity calculated using Hering-Zipperer	[ $\mu\text{Pa}\cdot\text{s}$ ]
$\eta_{GC}$	viscosity calculated using GasCalc	[ $\mu\text{Pa}\cdot\text{s}$ ]
$\eta_{RP}$	viscosity calculated using RefProp	[ $\mu\text{Pa}\cdot\text{s}$ ]

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