Paper 21

Gerg Project: Wide-Range Reference Equation of State for Natural Gases

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SUMMARY

A group of European gas companies, GERG, supported the development of a new equation of state for the thermodynamic properties of natural gases covering the gas and liquid region including the vapour-liquid phase equilibrium. The new equation, GERG02, was developed on the basis of a multi-fluid approximation using pure substance equations for each component and experimental data for binary mixtures only. Therefore, the representation of multicomponent mixture data is predictive.

The results calculated with the new equation for thermal and caloric properties of natural gas mixtures show substantial improvements in comparison to the AGA8-DC92 and the GERG88 equation, which are known to be the current internationally accepted standard for density or compression factor calculations at pipeline conditions (see ISO 12213-part 2 and 3, 1997). The new reference equation allows high accuracy calculations for thermal and caloric properties in the homogeneous region (gas, liquid and supercritical) and also enables calculations for the vapour-liquid equilibrium. The reference equation can be used as a database or reference for technical applications and processes with natural gases, LNG, LPG, natural gas vehicles and hythane mixtures.

The uncertainty in density and in speed of sound at pipeline conditions is less than ±0.1%. As a result of the poor data situation, the description of the liquid phase and the vapour-liquid phase equilibrium is less accurate but still as good as possible compared to the uncertainty of the available experimental data.

Gas densities measured for rich natural gases containing high fractions of ethane (up to 20%), propane (up to 14%) and butane (up to 6%). are calculated by the new reference equation within less than ±0.1% to ±0.2%. The AGA8-DC92 equation does not accurately describe the densities of these gas mixtures.

For LNG like mixtures the new equation reproduces experimental saturated liquid densities to within ±0.2%. For comparison, the still widely used Peng-Robinson cubic equation deviate by more than 10%.

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1. INTRODUCTION

Thermodynamic properties of the natural gas main component methane and of natural gases have frequently been investigated because of their industrial importance. The most recent equations of state for thermodynamic properties were reported for methane by Wagner and de Reuck [1] and for natural gases by Jaeschke et al. [2, 3] (GERG88 equation) and by Starling and Savidge [4] (AGA8-DC92 equation), respectively. These equations for natural gases are now routinely applied in energy billing. Specifically with the help of these equations the volume metered under varying, actual field conditions are converted to well defined reference conditions.

The GERG88 equation is a thermal (volumetric) equation of state only. AGA8-DC92 designed as a caloric equation was assessed in an earlier project and shortcomings of the AGA8-DC92 equation were identified [5]. Especially for temperatures below 270 K, deviations of up to ±0.3%, ±1.0% and ±2% from experimental data occurred for density, speed of sound and isobaric enthalpy changes, respectively. Therefore, a consortium of European gas companies (Enagas, Gastransport Services, Gaz de France, Snam Rete Gas, Ruhrgas, Statoil) contracted Ruhr-Universität Bochum (RUB, W. Wagner) to develop a wide-range reference equation of state for natural gases. This paper discusses the structure of the new wide-range equation GERG02 being developed on the basis of a multi-fluid approximation using pure substance equations for each component and experimental data for binary mixtures only. The new reference equation is the first accurate natural gas equation that can be applied to the gas and liquid phases and also to vapour-liquid equilibrium calculations. The equation allows high accuracy calculations for thermal and caloric properties in the homogeneous region and also allows calculations for the vapour-liquid phase equilibrium. The results calculated with the new equation of state for thermal and caloric properties of natural gas mixtures are compared to results from the AGA8-DC92 equation, the current internationally accepted standard at pipeline conditions, as well as to results from the Klosek-McKinley model [6, 7] used for the calculation of LNG densities.

2. RANGE OF APPLICATION

The composition range of application of the new reference equation is given in Table 1. The new equation of state will allow the properties of natural gases to be calculated with the highest possible accuracy. The uncertainty in density and in speed of sound of typical natural gases is less than ±0.1% for temperatures from 250 K to 350 K and pressure up to 30 MPa.

Outside this range at lower temperatures, the thermodynamic properties are calculated within the experimental uncertainty of the best data available. The new mixture model also allows a reasonable description of properties in the vapour-liquid phase equilibrium. The limitations depend mainly on the available data base. Furthermore, the new equation GERG02 can accurately describe pure fluids (i.e. up to 100 mole percent). As the data available for mixtures is incomplete, the accuracy of the new equation cannot thoroughly be assessed for compositions outside the range given in Table 1.

Table 1. Range of mixture composition for natural gas

relative density [1] superior calorific value [1, 2]		0.55 to 0.90 19 to 48 MJ/m³
main components		
	mole percent methane	≥ 45
	mole percent nitrogen	≤ 50
	mole percent carbon dioxide	≤ 30
	mole percent ethane	≤ 20
secondary components		
	mole percent propane	≤ 10
	mole percent butanes	≤ 3
	mole percent pentanes	≤ 0.5
minor components		
	mole percent hexanes and above	≤ 0.3
	mole percent helium	≤ 0.2
	mole percent hydrogen	≤ 10
	mole percent carbon monoxide	≤ 3
	mole percent argon	≤ 0.05
	mole percent oxygen	≤ 0.05
	mole percent water	≤ 0.02 [3]

[1] *reference conditions: metering at 0°C, 0.101325 MPa*
[2] *reference conditions: combustion at 25°C, 0.101325 MPa*
[3] *and below water dew point*

3. EXPERIMENTAL WORK

The experimental part of the work included density measurements at Ruhr-Universität Bochum, Bochum [8], speed of sound measurements at Imperial College, London [9], and vapour-liquid equilibria (VLE) measurements at Technische Universität Braunschweig [10]. Additional density measurements have been made for ethane and propane. Speed of sound measurements have been carried out for methane + nitrogen and ethane + nitrogen mixtures and vapour-liquid equilibrium data including saturated densities and their compositions in the liquid and gas phase have been measured for systems containing methane, ethane, and nitrogen.

4. THEORETICAL WORK

The theoretical work and development of the new GERG02 equation of state have been carried out at Ruhr-Universität Bochum. This work also included the collection and examination of data available from publications to be used in the development or for testing the new equation of state.

4.1. Pure Fluid Equations

On the basis of accurate experimental data Ruhr-Universität Bochum [11] has developed equations of state for each of the main components methane, nitrogen, ethane and carbon dioxide, which describe the most important region in reference quality. The equations for methane, ethane, and nitrogen have an identical structure with 24 terms while the equation for carbon dioxide consists of 22 terms. Simple equations of states with 12 terms are used for secondary and minor components such as i.e. propane, n-butane and isobutane, n-pentane and isopentane, argon or oxygen.

4.2. Natural Gas Reference Equation

The natural gas reference equation is explicit in the reduced Helmholtz energy, $\boldsymbol{a} = A/(RT)$. The equation was developed on the basis of a multi-fluid approximation:

$$\boldsymbol{a}(\boldsymbol{d},\boldsymbol{t},X) = \boldsymbol{a}^{\circ}(\boldsymbol{d},\boldsymbol{t},X) + \sum_{i=1}^{k} x_i \boldsymbol{a}_{\mathrm{o},i}^{\mathrm{r}}(\boldsymbol{d},\boldsymbol{t}) + \sum_{i=1}^{k-1}\sum_{j>i}^{k} \Delta \boldsymbol{a}_{ij}^{\mathrm{r}}(\boldsymbol{d},\boldsymbol{t},X) \tag{1}$$

In a multi-fluid approximation the reduced Helmholtz energy $\boldsymbol{a}$ is the sum of three different parts, the ideal gas part, $\boldsymbol{a}^{\circ}$, the contribution of the pure fluid equations, $\sum x_i \boldsymbol{a}_{\mathrm{o},i}^{\mathrm{r}}(\boldsymbol{d},\boldsymbol{t})$, and the departure function, $\sum\sum \Delta \boldsymbol{a}_{ij}^{\mathrm{r}}(\boldsymbol{d},\boldsymbol{t},X)$. The residual parts $\boldsymbol{a}_{\mathrm{o},i}^{\mathrm{r}}$ of the pure fluids i are taken at the reduced density $\boldsymbol{d}$ and at the inverse reduced temperature $\boldsymbol{t}$ of the mixture. The variables are defined as:

- $\boldsymbol{r}$: density
- T: temperature
- X: vector of composition in natural gas
- $\boldsymbol{d}$: reduced density (= $\boldsymbol{r}/\boldsymbol{r}_{\mathrm{r}}(X)$)
- $\boldsymbol{t}$: reduced temperature (= $T_{\mathrm{r}}(X)/T$)
- x_i: mole fraction of component i
- k: number of components

4.3. Reducing Functions

$\boldsymbol{r}_{r}(X)$ and $T_{r}(X)$ are reducing functions which depend only on composition [11].

$$\frac{1}{\boldsymbol{r}_{\mathrm{r}}(X)} = \sum_{i=1}^{k}\sum_{j=1}^{k} x_i x_j \boldsymbol{b}_{v,ij} k_{v,ij} \frac{x_i + x_j}{\boldsymbol{b}_{v,ij}^2 x_i + x_j} \frac{1}{8}\left(\frac{1}{\boldsymbol{r}_{\mathrm{c},i}^{1/3}} + \frac{1}{\boldsymbol{r}_{\mathrm{c},j}^{1/3}}\right)^3 \tag{2}$$

$$T_r(X) = \sum_{i=1}^{k} \sum_{j=1}^{k} x_i x_j \boldsymbol{b}_{T,ij} k_{T,ij} \frac{x_i + x_j}{\boldsymbol{b}_{T,ij}^2 x_i + x_j} \left(T_{c,i} \cdot T_{c,j}\right)^{0.5} \qquad (3)$$

Each reducing function contains two adjusted parameters, $k_{v,ij}$, $\boldsymbol{b}_{v,ij}$, and $k_{T,ij}$, $\boldsymbol{b}_{T,ij}$, respectively. These binary parameters were fitted to data for binary mixtures. If their values are set to 1, their influence disappears and only the combining rules determine the reducing values of the respective mixture. For the pure fluid i the reducing functions are equal to the critical density $\boldsymbol{r}_{c,i}$ and the critical temperature $T_{c,i}$ of the fluid.

4.4. Departure Functions

Two different types of binary departure functions are used, namely binary specific departure functions for certain binaries and a generalised departure function. A binary specific departure function can be written as:

$$\Delta \boldsymbol{a}_{ij}^r(\boldsymbol{d},\boldsymbol{t},X) = x_i x_j F_{ij} \sum_{m=1}^{M_{ij}} n_{m,ij} \boldsymbol{d}^{d_{m,ij}} \boldsymbol{t}^{t_{m,ij}} \exp\left(-\boldsymbol{d}^{c_{m,ij}}\right) \qquad (4)$$

The departure function is equal to zero at the limit of a pure fluid. The summation depends on reduced density and temperature and contains polynomial and exponential terms. In the binary specific departure functions the parameters c, d, n and t have specific values for the mixture ij and the parameter F_{ij} is equal to one. The generalised departure function adopts the same structure for the density and temperature-dependant part for different binaries. Thus, the values of the parameters c, d, n and t are equal for a group of binary mixtures, but the parameter F_{ij} is fitted to data of this binary system ij. Such departure functions used for the binary systems of natural gas mixtures typically consist of 5 to 9 terms and were developed using binary mixture data only. If the information on binary mixtures is even more limited, the departure function is set to zero. Then, only adjusted reducing functions are used, propagating the information on those binary mixtures into the equation of state through the second term of equation (1). Fortunately, this is only the case for less important mixtures.

5. RESULTS

Natural gas data were not used in the development of the new equation of state and, therefore, the calculations for multicomponent mixtures are predictive.

5.1. Pure Fluids

With the new equations for the pure fluids, typical deviations of calculated densities from experimental data are approx. ±0.03 %, ±0.1 % and ±0.2 % for the main, secondary and minor components, respectively (see Table 2). For speed of sound data the situation in the gas phase is similar. As a result of the lack of accurate speed of sound data in the liquid and super-critical regions for the secondary and minor components, the uncertainties here are higher than for densities.

Table 2. Typical deviations of density calculated with the new reference equation from accurate experimental data for pure components

	gas and super critical (*dense fluid*)	liquid
highly accurate EOS (22 – 24 terms, *main components*)	±0.03%	±0.05%
simple EOS (12 terms, *secondary components*)	±0.1%	±0.1%
simple EOS (12 terms, *minor components*)	±0.1% to ±0.2%	±0.1% to ±0.2%

5.2. LNG like mixtures

For LNG like mixtures, the new equation reproduces experimental saturated liquid densities [12,13] to within ±0.2%. For comparison, the still widely used Peng-Robinson cubic equation [14] deviates by more than 10% (see Figure 1). The Klosek-McKinley equation [6, 7], which was correlated to these saturated liquid densities, also reproduces the data within ±0.2%. But it does not allow the calculation of the other thermodynamic properties or of the phase boundary or of the homogeneous gas region. The AGA8-DC92 equation [4] is not applicable to liquid densities.

Peng & Robinson (1976)
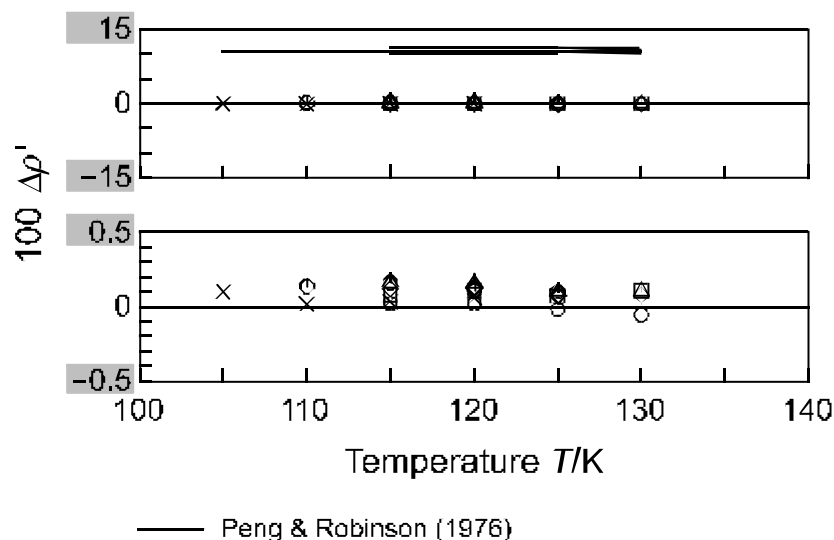
Figure 1. Percentage deviations of experimental saturated liquid densities for LNG like mixtures containing up to 8 components (Hiza et al. 1980, Haynes 1982); baseline: new equation

5.3. Natural gases

Speed of sound

In the temperature range from 250 K to 350 K the new equation describes the speed of sound data of typical natural gases [15, 16] to within ±0.1%. Figure 2 shows some results of the Younglove et al. data [15]. For the AGA8-DC92 equation deviations of up to –0.5% occur at 250 K and above 5 MPa.
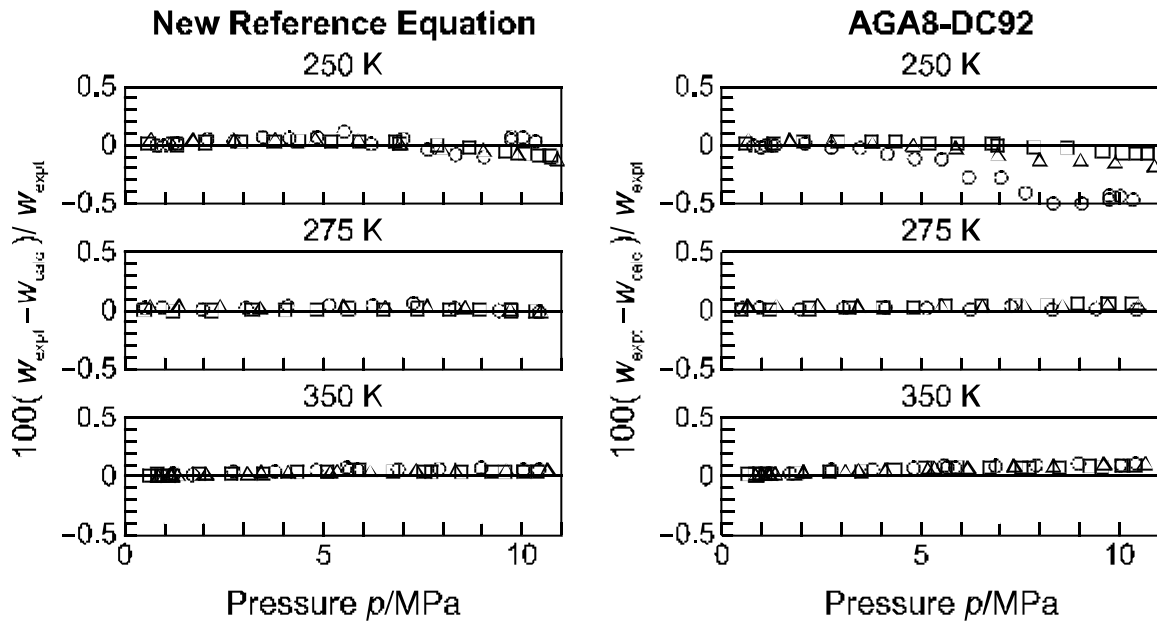

Figure 2. Percentage deviations of experimental speed of sound data for three natural gas mixtures (Younglove et al. 1993); baseline: left new equation, right AGA8-DC92 equation

Isobaric enthalpy differences

For the isobaric enthalpy differences the new equation agrees with the data [17] within their experimental uncertainty which is supposed to be of ±0.2% to ±0.5%. For the AGA8-DC92 equation at the lower temperature around 250 K the deviations are larger than +1.5% (see Figure 3). Thus a substantial improvement can be seen, especially at lower temperatures (250 K) and predominantly for caloric properties.

Thermal or volumetric properties

Thermal properties for natural gases such as density or compression factor are improved and calculated by the new equation in the gas phase at lower temperatures (250 K) and at pressures up to 30 MPa within ±0.1%. The AGA8-DC92 equation does not accurately describe the data at the lowest temperature especially for nitrogen rich gases, where differences of –0.2% occur. This statement is based on measurements performed in a GRI-GERG round-robin test [18, 19] and data sets D22, D23, D47 and D48 of [20] (see Figure 4). For a quaternary natural gas like mixture, data set D18 of [20], the percentage deviations are greater than –0,3% for the AGA8-DC92 equation(see Figure 5).
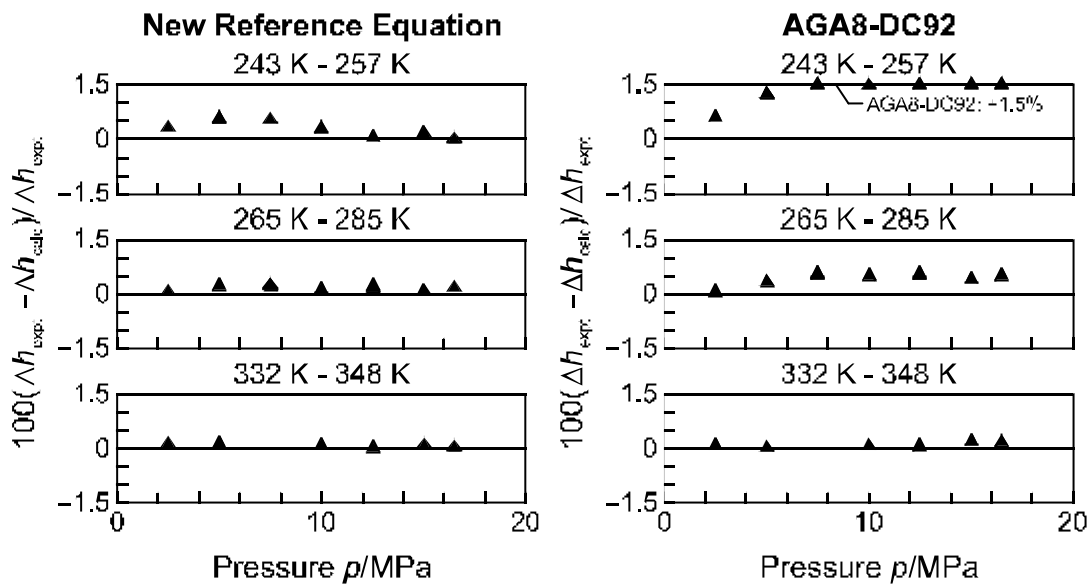
Figure 3. Percentage deviations of experimental isobaric enthalpy differences of a quintic natural gas like mixture (Owren et al. 1996); baseline: left new equation, right AGA8-DC92 equation
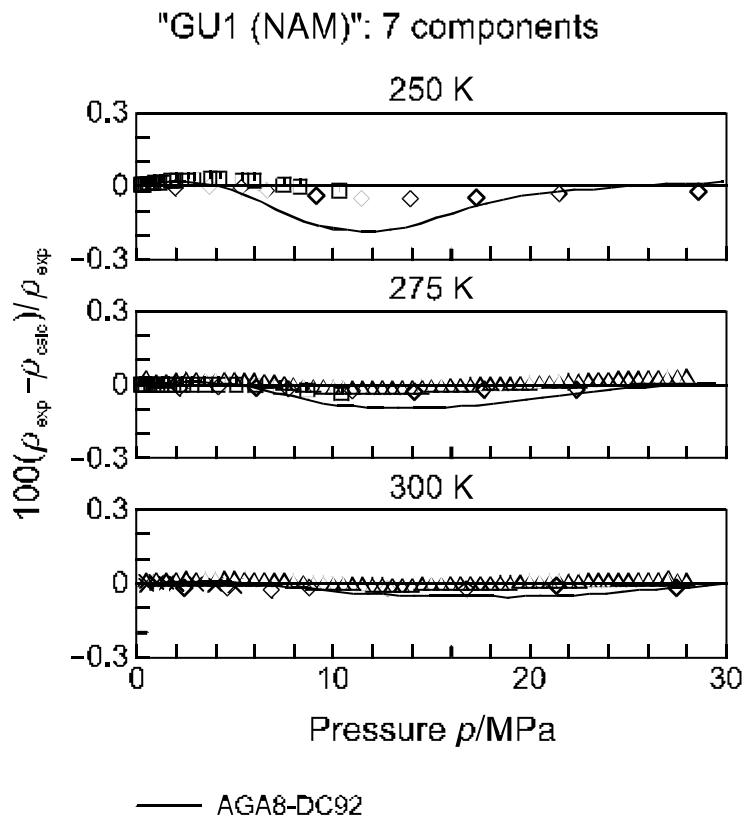

Figure 4. Percentage deviations of experimental density (prT) data for the natural gas "GU1" (Hwang et al. 1997, Magee et al. 1997 and data set Ruhrgas D22, Jaeschke et al. 1997); baseline: new equation
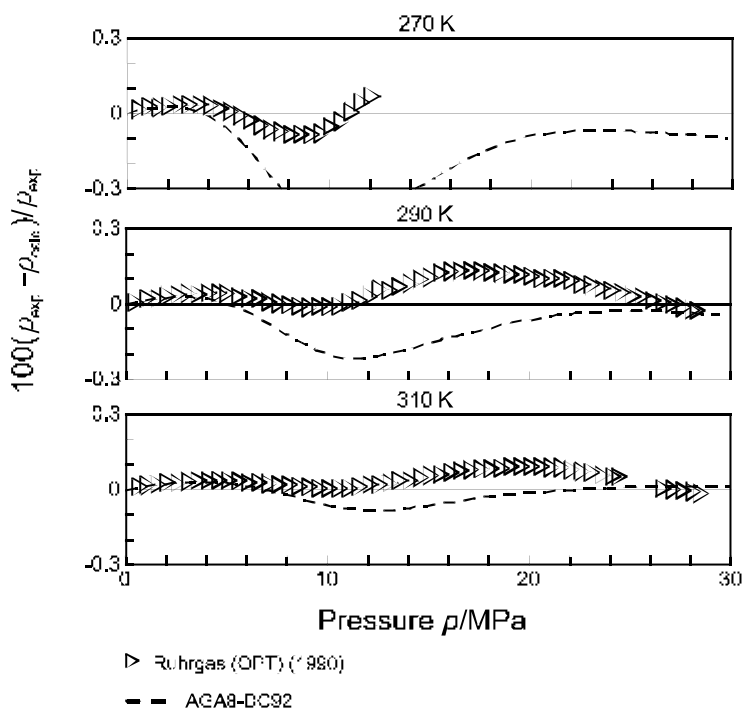
Figure 5. Percentage deviations of experimental density (prT) data for a quaternary mixture with 61% CH_4, 13% N_2, 13% CO_2 and 13% C_2H_6 (data set Ruhrgas D18, Jaeschke et al. 1997); baseline: new equation

5.4. Special gases

Rich natural gases

Rich natural gas mixtures are natural gases containing high fractions of ethane (up to 20%), propane (up to 14%) and butane (up to 6%). Gas densities measured for rich natural gases in the temperature range of 310 K to 360 K and pressures up to 30 MPa and at lower temperatures below the two-phase regime are calculated by the new reference equation within less than ±0.1% to ±0.2%. The AGA8-DC92 equation does not accurately describe the densities of these gas mixtures. The differences to the experimental data of Watson and Millington [21] or Jaeschke and Schley [22] are between +0.5% and +1% (see Figures 6 and 7).

Hythane mixtures

Hythane mixtures are natural gases containing a high fraction of hydrogen. These mixtures may serve in the future as alternative fuel enabling the reduction of the emission of CO, CO2 and NOx. For natural gases containing a high fraction of hydrogen (10% hydrogen), see data sets N72 to N74 of [20], or methane - hydrogen binary mixtures with up to 75% of hydrogen, see data sets B25-1 to B25-7 of [20], the new equation reproduces the experimental data in the temperature range of 270 K to 350 K and pressures up to 30 MPa within ±0.1%. Moreover, for the special gas mixture N116 of [20] containing high fractions of hydrogen, nitrogen and carbon monoxide the agreement is within ±0,1% whereas the AGA8-DC92 equation deviates by more than +0,3% up to +0,7% (see Figure 8). Thus, the new equation enables for the first time to accurately describe hythane mixtures.
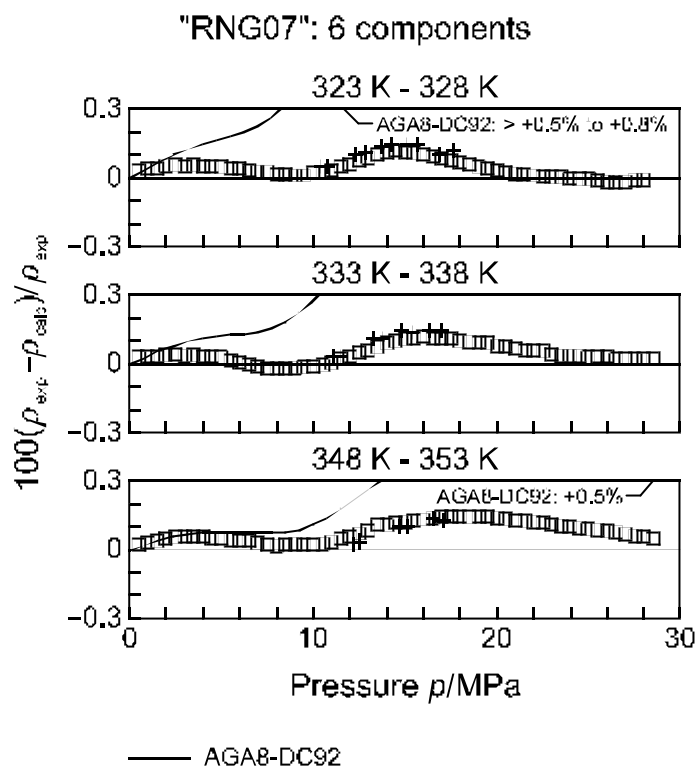
Figure 6. Percentage deviations of experimental density (*prT*) data for the rich natural gas "RNG07" with 59% CH_4, 10% C_2H_6, 14% C_3H_8 and 6% n-C_4H_{10} (Watson et al. and Jaeschke et al, both 1998); baseline: new equation

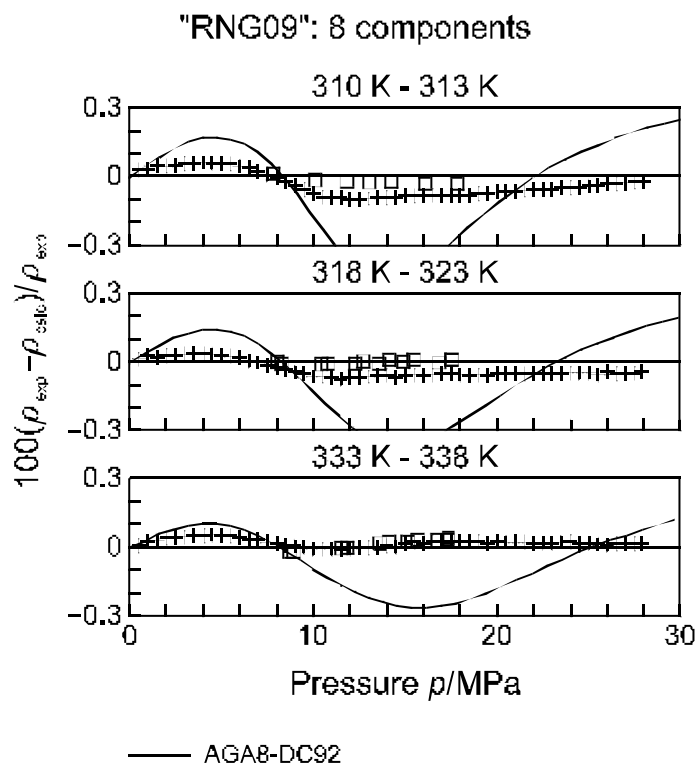**Figure 7.** Percentage deviations of experimental density (*prT*) data for the rich natural gas "RNG09" with 59% CH_4, 18% C_2H_6, 8% C_3H_8 and 0,5% n-C_4H_{10} (Watson et al. and Jaeschke et al, both 1998); baseline: new equation
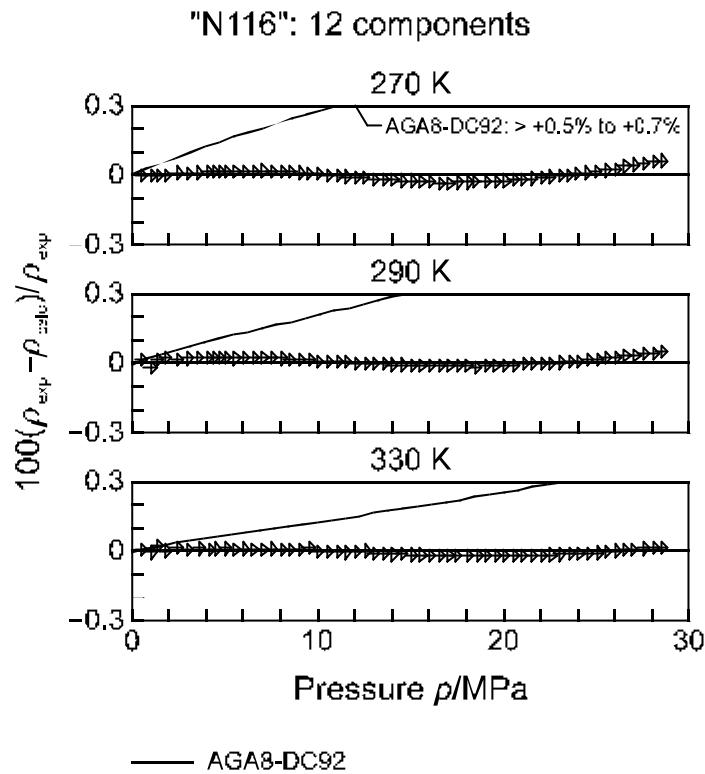
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"N116": 12 components

Figure 8. Percentage deviations of experimental density (*prT*) data for a natural gas containing 12 components with high fractions of 28% N_2, 27% H_2 and 13% CO (data set Ruhrgas N116, Jaeschke et al. 1997); baseline: new equation

6. CONCLUSIONS

The new reference equation describes caloric and thermal properties of pipeline quality natural gases in the gas phase within the experimental uncertainty of the data and more accurately than the AGA8-DC92 equation. The uncertainty in density and in speed of sound at pipeline conditions is less than ±0.1%. Furthermore, the new equation has a wider range of validity with respect to composition and therefore allows special gases, rich natural gas or hythane gas mixtures, to be handled very accurately.

The reference equation allows high accuracy calculations for thermal and caloric properties in the homogeneous region and also allows calculations for the vapour-liquid phase equilibrium. Furthermore, in the liquid phase and at saturation conditions the new equation is valid and reproduces the data within their experimental uncertainty where other equations fail or differ by more than +10 to +20%. As a result of the poor data situation, the description of the liquid phase and the vapour-liquid phase equilibrium is less accurate compared to the uncertainty given for density and speed of sound in the gas phase, but still as good as possible.

The reference equation can be used as a database or reference for present and future technical applications and processes with natural gases, rich natural gases (RNG), liquefied natural gases (LNG), liquefied petroleum gases (LPG), compressed natural gases (CNG) and hythane mixtures.

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7. ACKNOWLEDGEMENTS

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