

Accurate experimental ( $p$ ,  $\rho$ ,  $T$ ) data for the introduction of hydrogen into the natural gas grid:  
Thermodynamic characterization of the nitrogen-hydrogen binary system from 240 K to 350 K and  
pressures up to 20 MPa

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## Abstract

The experimental density data of the binary system nitrogen-hydrogen available at the time of the development of the equation of state for natural gases and related mixtures, GERG-2008, were limited to hydrogen contents higher than 0.15 (amount-of-substance fraction) and temperatures above 270 K. This work provides accurate experimental ( $p$ ,  $\rho$ ,  $T$ ) data for three binary mixtures of nitrogen and hydrogen: (0.95 N<sub>2</sub> + 0.05 H<sub>2</sub>), (0.90 N<sub>2</sub> + 0.10 H<sub>2</sub>), and (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) at temperatures of (240, 250, 260, 275, 300, 325, and 350) K, thus extending the range of available experimental data to low hydrogen contents and low temperatures. The density measurements were performed by using a single-sinker densimeter with magnetic suspension coupling at pressures up to 20 MPa. Experimental data were compared with the corresponding densities calculated from the GERG-2008 and the AGA8-DC92 equations of state. The relative deviations of the experimental data from both equations of state were within the estimated uncertainty value of the equations. Therefore, the experimental data agree very well with the values estimated from the equations. The virial coefficients  $B(T,x)$ ,  $C(T,x)$ , and  $D(T,x)$  as well as the second interaction virial coefficient  $B_{12}(T)$  for the nitrogen-hydrogen binary system were also calculated from the experimental data set at temperatures from (240 to 350) K. The resulting values agree with those from literature.

Keywords: nitrogen; hydrogen; natural gas; density; single-sinker densimeter; GERG-2008 equation of state; virial coefficient.

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## 1. Introduction

The GERG-2008 equation of state (EoS)<sup>1</sup> currently serves as the ISO standard (ISO 20765-2) for the calculation of thermodynamic properties of natural gases<sup>2</sup>. The GERG-2008 EoS allows estimating the thermophysical properties in the entire fluid region for natural gases and related mixtures of up to 21 components from the following three elements that constitute the entire equation of state: pure-substance equations of state for all considered mixture components; composition-dependent reducing functions for the mixture density and temperature; and a departure function depending on the reduced mixture density, the inverse reduced mixture temperature, and the mixture composition. The 21 components considered in the GERG-2008 EoS generate 210 binary combinations with different types of functions for the description of the constituent binary mixtures. Consequently, the quality and the availability of experimental data determine and limit the achievable accuracy of the GERG-2008 calculations. Some data sets in the underlying framework of the GERG-2008 EoS are less substantiated, especially for mixtures containing the so-called “secondary components” in the GERG-2008. Hydrogen appears as a secondary component in the claims of the EoS and there is neither a binary specific nor a generalized departure function for the (N<sub>2</sub> + H<sub>2</sub>) binary system. Furthermore, the experimental data for this binary mixture available at the time of the development of the GERG-2008 EoS were restricted to hydrogen contents higher than an amount-of-substance fraction of 0.15 and temperatures above 270 K<sup>3-8</sup>.

Additionally, interest in hydrogen as an energy-storage medium is increasing. A practical alternative to transport and distribute hydrogen is using the existing infrastructure for natural gas. Experimental characterization of the thermodynamic behavior of binary gas mixtures of hydrogen with the main components of natural gas is of great importance for validation and improvement of the established reference equation of state for natural gas.

This work provides accurate experimental ( $p$ ,  $\rho$ ,  $T$ ) data for three nitrogen and hydrogen binary mixtures with nominal compositions of (0.95 N<sub>2</sub> + 0.05 H<sub>2</sub>), (0.90 N<sub>2</sub> + 0.10 H<sub>2</sub>), and (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>). Density measurements were performed by using a single-sinker densimeter with magnetic suspension coupling at temperatures of (240, 250, 260, 275, 300, 325, and 350) K and pressures up to 20 MPa. The experimental data were compared with the corresponding densities calculated from the GERG-2008 and the AGA8-

DC92 EoS.<sup>9</sup> Additionally, the second to fourth virial coefficients  $B(T, x)$ ,  $C(T, x)$ , and  $D(T, x)$  as well as the (composition-independent) second interaction virial coefficients  $B_{12}(T)$  for the (N<sub>2</sub> + H<sub>2</sub>) binary mixture at temperatures from (240 to 350) K were also evaluated from the experimental data set.

To achieve the highest accuracy in composition, the three binary mixtures were prepared by a gravimetric method.

## 2. Experimental

### 2.1. Mixtures preparation

Three (N<sub>2</sub> + H<sub>2</sub>) binary mixtures were prepared by the Federal Institute for Materials Research and Testing (Bundesanstalt für Materialforschung und –prüfung, BAM) in Berlin, Germany, according to the ISO 6142-1<sup>10</sup>. Table 1 shows the purity, supplier, molar mass, and critical parameters of the pure compounds (obtained from the reference equations of state for nitrogen<sup>11</sup> and hydrogen<sup>12</sup>). Table 2 shows the cylinder identifiers, composition, and the corresponding expanded uncertainty ( $k = 2$ ) of the mixtures. All substances were used without further purification, but the information on impurities from the specification was considered in the mixture preparation, according to the recommendations by CCQM (Consultative Committee for Amount of Substance: Metrology in Chemistry and Biology) and ISO. This is why there are some additional components in Table 2. However, the influence of those additional components are not considered in the comparison of the experimental results with the GERG-2008 and AGA8-DC92 equations of state. The prepared mixtures were supplied in aluminum cylinders of a volume of 10 dm<sup>3</sup>.

The preparation of the mixtures was carried out in two steps. First, the equimolar mixture of (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) was prepared by the consecutive introduction of pure nitrogen and pure hydrogen into an evacuated recipient cylinder (BAM no.: 96055000-160426, volume: 10 dm<sup>3</sup>). The filling was carried out by only working with the pressure difference between the cylinder which contained the pure compound and the recipient cylinder, thus avoiding the use of a compressor which could have introduced impurities. The mass of the gas portion was determined after each filling step using a high-precision mechanical gas balance (Volland model HCE 25, Volland Corp., New Rochelle NY, USA).

The other two binary mixtures were prepared by diluting a determined portion of the (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) parent mixture in a properly measured amount of nitrogen. The three mixtures were finally homogenized by a procedure of subsequent heating and rolling.

The samples of (0.95 N<sub>2</sub> + 0.05 H<sub>2</sub>) and (0.90 N<sub>2</sub> + 0.10 H<sub>2</sub>) were validated by gas chromatography (GC) on a multichannel GC-analyzer (Siemens MAXUM II, Siemens AG, Karlsruhe, Germany) using independently prepared gases (see Table 3) following a two-point bracketing method according to ISO 12963<sup>13</sup>. The analysis was performed isothermally at 333.15 K (60 °C). Hydrogen and nitrogen were analyzed on different channels equipped with packed columns and a separate TCD for each channel. Nitrogen was used as the carrier gas for hydrogen and helium for nitrogen, respectively. The equimolar (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) mixture was not directly analyzed due to the fact that its composition is beyond the recommended operational range of the GC-analyzer. However, it was possible to validate that composition indirectly from the results for the two other mixtures as these results were in concordance. The results of the GC analysis can be found in Table 3. These validations were carried out at BAM prior to the shipment of the bottles for density determination at the University of Valladolid.

## 2.2. Equipment description

The ( $p$ ,  $\rho$ ,  $T$ ) data were obtained by using a single-sinker densimeter with magnetic suspension coupling especially designed for density measurements of pure gases and gaseous mixtures. The equipment has been previously described in detail by Chamorro *et al.*<sup>14</sup> and has been further improved by Mondéjar *et al.*<sup>15</sup>. This method, originally developed by Brachthäuser *et al.*<sup>16</sup> and further improved by Klimeck *et al.*<sup>17</sup>, operates on the Archimedes' principle. The magnetic suspension coupling system allows measuring the buoyancy force on a sinker immersed in the gas without any contact between the sinker and a balance. Thus, accurate density measurements of fluids over wide temperature and pressure ranges can be performed<sup>18</sup>. The sinker used in this work was a silicon cylinder with a mass of  $61.59181 \pm 0.00016$  g and a volume of  $26.444 \pm 0.003$  cm<sup>3</sup> ( $k = 2$ ), at  $T = 293.05$  K and  $p = 0.101134$  MPa, determined by calibration at the Spanish National Metrology Institute (*Centro Español de Metrología*, CEM). Two calibrated masses

were used for reducing the ‘nonlinearity effect’ of the electronic balance and to improve the accuracy of the measurements. The calibrated masses are made of tantalum and titanium, respectively. They have different masses but approximately the same volume (4.9 cm<sup>3</sup>) and the difference in weight between the masses is similar to the sinker mass (of approximately 60 g). This load compensation system allows operating the balance near to its zero point and additionally makes the air buoyancy effect negligible. The two masses were provided by Rubotherm GmbH, Bochum, Germany, and their masses and volumes were determined at the Mass Division of the Spanish National Metrology Institute (CEM)<sup>19</sup>.

The density of the fluid can be calculated from the following expression:

$$\rho = \frac{(m_{s0} - m_{sf})}{V_s(T, p)}$$

(1)

where the difference between the sinker mass in a vacuum,  $m_{s0}$ , and the apparent sinker mass in the pressurized fluid,  $m_{sf}$ , is expressed in kg and refers to the buoyancy force that is exerted on by the sinker and is determined by a high-precision microbalance (Mettler Toledo AT261, Mettler Toledo GmbH, Gießen, Germany).  $V_s(T, p)$  is the volume of the sinker immersed in the fluid in m<sup>3</sup>, which is accurately known as a function of temperature and pressure.<sup>15</sup>

The temperature of the gas mixture inside the measuring cell is determined by two platinum resistance thermometers (S1059PJ5X6, Minco Products, Inc., Minneapolis MN, USA) connected to an AC comparator resistance bridge (F700, Automatic Systems Laboratories, Redhill, England). The pressure inside the cell is recorded by two pressure transducers which cover different pressures ranges: a Paroscientific 2500A-101 for pressures from (0 to 3) MPa and a Paroscientific 43KR-HHT-101 (Paroscientific Inc., Redmond WA, USA) for pressures up to 20 MPa.

The single-sinker densimeter is one of the most accurate devices for the measurement of fluids densities. However, there are two effects that must be considered: the force transmission error (FTE) due to the magnetic coupling<sup>20</sup> and the adsorption of gas molecules on the cell and sinker surfaces<sup>21</sup>. The FTE

consists of two terms: the apparatus effect and a fluid-specific effect. The apparatus effect of the FTE can be cancelled by an operational technique in which the sinker mass is also determined in a vacuum after the measurements of each isotherm have been finished. The fluid-specific effect depends on the magnetic behavior of the measured gas. The magnetic susceptibilities ( $\chi$ ) of the three (N<sub>2</sub> + H<sub>2</sub>) binary mixtures studied in this work were estimated by using the additive law proposed by Bitter<sup>22</sup>. The estimated values were  $\chi_{5\%} = 5.89 \cdot 10^{-9}$  for the (0.95 N<sub>2</sub> + 0.05 H<sub>2</sub>) mixture,  $\chi_{10\%} = 5.57 \cdot 10^{-9}$  for the (0.90 N<sub>2</sub> + 0.10 H<sub>2</sub>), and  $\chi_{50\%} = 3.02 \cdot 10^{-9}$  for the (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) mixtures. It is reported in the literature that the apparatus effect has a stronger influence than the fluid-specific effect, except for strongly paramagnetic fluids<sup>20</sup>. Since the values of magnetic susceptibility of the studied (N<sub>2</sub> + H<sub>2</sub>) mixtures are relatively low, a strong paramagnetic behavior is not perceived (e.g., the magnetic susceptibility of oxygen is bigger by more than two orders of magnitude:  $\chi_{O_2} = 1.78 \cdot 10^{-6}$ ). Therefore, a correction due to the fluid-specific effect was not considered in the measurements.

Regarding sorption effects inside the measuring cell, these effects have been previously discussed by other authors<sup>21</sup>, who estimate that they can be responsible for significant errors up to 0.1 % in density measurements, mainly when dealing with high densities or near the saturation line. According to the experimental procedure described in the next section, the measuring cell is evacuated at the end of each isotherm after a residence time of the fluid inside the cell lower than 40 hours. Fresh mixture is introduced in the cell for recording a new isotherm. Additionally, experimental points are far away from the saturation line of the mixture. The sorption tests carried out in previous experiments with this equipment show that the differences that manifest in a trend of the relative deviation in density from the GERG-2008 EoS along the period of recording an isotherm are one order of magnitude lower than the density uncertainty at the working pressure.<sup>23</sup> Therefore, sorption effects have not been taken into account in the measurements presented in this work.

### **2.3. Experimental procedure**

Experimental  $(p, \rho, T)$  data for the  $(0.95 \text{ N}_2 + 0.05 \text{ H}_2)$  and  $(0.90 \text{ N}_2 + 0.10 \text{ H}_2)$  binary mixtures were obtained at temperatures of (240, 250, 260, 275, 300, 325, and 350) K and pressures up to 20 MPa. The same isotherms, except at 260 K, were recorded for the  $(0.50 \text{ N}_2 + 0.50 \text{ H}_2)$  mixture. The pressure was decreasing in 1 MPa steps from (20 to 1) MPa for each isotherm and 30 measurements were recorded for each pressure step. The sinker mass in vacuum  $m_{s0}$  was determined at the end of each isotherm. This particular measurement, necessary for the fluid-density determination according to Eq. (1), also allows checking any misalignment suffered by the magnetic suspension coupling during the measurements and it cancels the apparatus effect of the FTE.

Before and after the measurements of the  $(\text{N}_2 + \text{H}_2)$  mixtures, test measurements using nitrogen as a reference fluid were carried out in the entire operational range of the apparatus. These results were compared with the densities calculated from the reference equation of state for nitrogen by Span *et al.*<sup>11</sup>. The relative deviations of the experimental data from the calculated densities remained within a  $\pm 0.02$  % band, with an absolute average deviation (AAD) of 0.0078 %.

#### 2.4. Uncertainty of the measurements

Uncertainties of the properties involved in the procedure of density determination by using the single-sinker densimeter of this work were previously evaluated by Mondéjar *et al.*<sup>15</sup>. The expanded uncertainty in temperature ( $k = 2$ ) is less than 4 mK. The pressure uncertainty depends on the individual pressure transducer and is given by Eq. (2) and Eq. (3) for the (3 to 20) MPa and (0 to 3) MPa transducers, respectively. The expanded uncertainty ( $k = 2$ ) in pressure is in both cases less than 0.005 MPa.

$$U(p)/\text{MPa} = 75 \cdot 10^{-6} \cdot p + 3.5 \cdot 10^{-3} \quad (2)$$

$$U(p)/\text{MPa} = 60 \cdot 10^{-6} \cdot p + 1.7 \cdot 10^{-3} \quad (3)$$

According to Eq. (1) and taking into account the law of propagation of uncertainties as displayed in the GUM<sup>24</sup>, density uncertainty depends on the uncertainty of the apparent sinker mass when the measuring



cell is evacuated  $m_{so}$ , and pressurized  $m_{sf}$ , and also depends on the uncertainty of the volume of the sinker,  $V_s(T, p)$ . The uncertainties of the apparent sinker masses are related with the balance readings and were calculated taking into account the balance calibration, resolution, repeatability, and drift as sources of uncertainty (both when the cell is pressurized and evacuated). The sinker volume changes with temperature and pressure due to its thermal and mechanical properties. The expanded uncertainty ( $k = 2$ ) in density  $\rho$  ( $\text{kg}\cdot\text{m}^{-3}$ ) is expressed as a function of density by Eq. (4).

$$U(\rho) / \text{kg}\cdot\text{m}^{-3} = 1.1 \cdot 10^{-4} \cdot \rho + 2.3 \cdot 10^{-2} \quad (4)$$

To calculate the overall expanded uncertainty in density  $U_T(\rho)$  ( $k = 2$ ), the uncertainties of density, temperature, pressure, and composition of the mixture must be considered, as expressed in Eq. (5).

$$U_T(\rho) = 2 \cdot \left[ u(\rho)^2 + \left( \left( \frac{\partial \rho}{\partial p} \right)_{T,x} \cdot u(p) \right)^2 + \left( \left( \frac{\partial \rho}{\partial T} \right)_{p,x} \cdot u(T) \right)^2 + \sum_i \left( \left( \frac{\partial \rho}{\partial x_i} \right)_{T,p,x_j \neq x_i} \cdot u(x_i) \right)^2 \right]^{0.5} \quad (5)$$

where  $p$  is the pressure,  $T$  is the temperature, and  $x_i$  is the amount-of-substance (mole) fraction of each of the mixture components. Partial derivatives were calculated from the GERG-2008 EoS by using the software REFPROP<sup>25</sup>.

Table 4 shows a summary of the uncertainty contributions of each property involved in the density determination and the overall uncertainty in density of the measurements for the three studied ( $\text{N}_2 + \text{H}_2$ ) binary mixtures.

### 3. Experimental results

Tables 5, 6, and 7 show the 399 experimental ( $p, \rho, T$ ) data measured for the three ( $\text{N}_2 + \text{H}_2$ ) binary mixtures in the course of this work. The relative deviation in density from the values estimated with the GERG-2008 and the AGA8-DC92 EoS are included in these tables. The state points on each isotherm were calculated as the arithmetic average of the last ten measures of the corresponding magnitude for each

pressure step. Tables 5, 6, and 7 also show the expanded uncertainty in density  $U(\rho_{\text{exp}})$  ( $k = 2$ ), calculated by Eq. (4), expressed in absolute density units and as percentage of the measured density.

Experimental data were compared with the corresponding densities calculated from the GERG-2008 and the AGA8-DC92 equations of state. Relative deviations of experimental density data from the densities obtained with the GERG-2008 and AGA8-DC92 equations of state are presented in Tables 5, 6, and 7, and in Figures 1 to 6.

The virial coefficients for the three ( $\text{N}_2 + \text{H}_2$ ) mixtures were calculated by fitting the experimental data to the virial equation of state (VEoS), as will be discussed in the next section.

## **4. Discussion of the results**

### **4.1. Relative deviation of the experimental data from the reference equations of state**

Figures 1 and 2 show the relative deviations for the ( $0.95 \text{ N}_2 + 0.05 \text{ H}_2$ ) mixture from the GERG-2008 and AGA8-DC92 EoS, respectively, over the entire  $p, T$ -range investigated. In an analogous manner, Figures 3 and 4 show the deviations for the ( $0.90 \text{ N}_2 + 0.10 \text{ H}_2$ ) mixture and Figures 5 and 6 for the ( $0.50 \text{ N}_2 + 0.50 \text{ H}_2$ ) mixture.

The uncertainty of the GERG-2008 in gas-phase over the temperature range from (250 to 450) K and at pressures up to 35 MPa is 0.1 % in density.<sup>1</sup> The relative deviations observed in the mixtures containing 5 % and 10 % of  $\text{H}_2$  are within this uncertainty value. The largest deviations of both mixtures were 0.032 % and 0.042 %, respectively. Therefore, it can be concluded that experimental data agree with the values estimated from the GERG-2008 EoS. Regarding the ( $0.50 \text{ N}_2 + 0.50 \text{ H}_2$ ) mixture, 81.5 % of measured points lie within a  $\pm 0.1$  % band. Nevertheless, the state points between (14 and 19) MPa in the 240 K isotherm present deviations slightly higher than 0.1 %. Here, the largest relative deviations for the equimolar mixture are observed at the lowest pressure. However, since the uncertainty of the measured density at such low density values is much larger than the deviations themselves, these deviations are not significant.

The relative deviations of experimental data from AGA8-DC92 EoS are very similar to the deviations from GERG-2008 for the three studied mixtures. A slightly smaller relative deviation is observed for the (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) mixture, especially at high pressures and low temperatures.

The relative deviations from the GERG-2008 EoS of the (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) mixture are compared to those from a mixture with a very similar composition reported by Jaeschke *et al.*<sup>3</sup> in Figure 7. As it can be observed, experimental data agree with those from literature at every corresponding temperature.

Other related works for the (N<sub>2</sub> + H<sub>2</sub>) binary system were found in the literature<sup>4,5,7,8</sup>. However, the compositions and/or the  $p, T$ -ranges investigated of these other mixtures deviated considerably from those of the three mixtures studied in this work, so they did not qualify for an explicit data comparison.

A statistical comparison of the relative deviation of the experimental data from the densities given by the GERG-2008 and AGA8-DC92 EoS is given in Table 8, together with the corresponding parameters calculated for literature mixtures with a similar composition. *AAD* is the average absolute deviation defined by Eq. (6), which indicates either a systematic or random difference between the data and the EoS; *Bias* is the average deviation defined by Eq. (7), which quantifies the average deviation of the data set; *RMS* refers to the root mean square defined by Eq. (8) and provides another indication of the systematic or random dispersion of the data from the EoS, and *MaxD* represents the absolute value of the maximum relative deviation in the considered data set. When all four statistical parameters are close to zero, data sets are accurately represented by the engaged equation of state.

$$AAD = \frac{1}{n} \sum_{i=1}^n \left| 10^2 \cdot \frac{\rho_{i,\text{exp}} - \rho_{i,\text{EoS}}}{\rho_{i,\text{EoS}}} \right| \quad (6)$$

$$Bias = \frac{1}{n} \sum_{i=1}^n \left( 10^2 \cdot \frac{\rho_{i,\text{exp}} - \rho_{i,\text{EoS}}}{\rho_{i,\text{EoS}}} \right) \quad (7)$$

$$RMS = \sqrt{\frac{1}{n} \sum_{i=1}^n \left( 10^2 \cdot \frac{\rho_{i,\text{exp}} - \rho_{i,\text{EoS}}}{\rho_{i,\text{EoS}}} \right)^2}$$

(8)

According to this statistical analysis, the relative deviation of experimental density data from values calculated from both EoS increases with the hydrogen content of the mixture, especially for deviations from the GERG-2008. The AAD from the GERG-2008 is 0.013 for the (0.95 N<sub>2</sub> + 0.05 H<sub>2</sub>) mixture, 0.027 for the (0.90 N<sub>2</sub> + 0.10 H<sub>2</sub>) mixture and 0.032 for the (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) mixture. The AAD from the AGA8-DC92 is 0.014 for the (0.95 N<sub>2</sub> + 0.05 H<sub>2</sub>) mixture, 0.017 for the (0.90 N<sub>2</sub> + 0.10 H<sub>2</sub>) mixture, and 0.018 for the (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) mixture.

#### 4.2. Virial coefficients

The virial coefficients for the three (N<sub>2</sub> + H<sub>2</sub>) mixtures were calculated by fitting the experimental data to the virial equation of state (VEoS).

$$Z = \frac{pM}{\rho RT} = \sum_{k=1}^N \left( \frac{B_k}{M^k} \right) \cdot \rho^k$$

(9)

where  $Z$  is the compressibility factor,  $p$  is the pressure,  $M$  is the molar mass,  $R$  is the ideal gas constant (8.31446 J·K<sup>-1</sup>·mol<sup>-1</sup>),  $T$  is the temperature,  $\rho$  is the mass density and  $B_k$  the virial coefficients, which have a physical meaning related to the interaction between groups of molecules, with  $B_1 = 1$ .  $N$  is the number of terms of the equation.

As the VEoS represents an infinite series, it must be truncated after a finite number of terms. The method proposed by Cristancho *et al.*<sup>26</sup> states that the number of terms to be fitted depends on the maximum experimental density,  $\rho_{\text{max}}$ , of the set of experimental data used for the adjustment. This method was used to determine the number of terms of the VEoS for processing the virial coefficients for the three (N<sub>2</sub> + H<sub>2</sub>) binary mixtures. The procedure consists of two consecutive fits: the first fit

determines the statistically significant combination of  $N$  and  $\rho_{\max}$  through determination of the molar mass  $M$  of the mixture as a parameter into the VEoS. In other words, this fit is used to estimate the number of virial coefficients which will give the best representation of experimental data. The second fit estimates those virial coefficients. Both fits were performed by using a least-squares fitting method implemented in MATLAB software<sup>27</sup>.

The results of the first fit, which were carried out with the experimental data obtained at 250 K, show that the final combinations for the studied mixtures were:  $N = 4$ ,  $\rho_{\max} = 145.769 \text{ kg}\cdot\text{m}^{-3}$  ( $p = 11 \text{ MPa}$ ) for the (0.95 N<sub>2</sub> + 0.05 H<sub>2</sub>) mixture;  $N = 4$ ,  $\rho_{\max} = 145.769 \text{ kg}\cdot\text{m}^{-3}$  ( $p = 14 \text{ MPa}$ ) for the (0.90 N<sub>2</sub> + 0.10 H<sub>2</sub>) mixture; and  $N = 3$ ,  $\rho_{\max} = 145.769 \text{ kg}\cdot\text{m}^{-3}$  ( $p = 12 \text{ MPa}$ ) for the (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) mixture.

The second fit to estimate the virial coefficients according to the results of the first fit was performed for each isotherm using the molar mass values from the gravimetric preparation of each mixture. Since experimental uncertainties of temperature, pressure, and density were taken into account, a normal random distribution term based on the estimated uncertainties for each magnitude was included into the fitting process with a coverage interval of 95 %. This distribution was performed following the Monte Carlo method as proposed by the GUM<sup>24</sup>. The estimated results for the second  $B(T, x)$ , third  $C(T, x)$ , and fourth  $D(T, x)$  virial coefficients with their corresponding uncertainties are shown in Table 9.

The nitrogen-hydrogen second interaction virial coefficient  $B_{12}(T)$  was also estimated from  $B(T, x)$  by Eq. (10). Virial coefficients of pure nitrogen ( $B_{11}$ ) and pure hydrogen ( $B_{22}$ ) were obtained from reference equations of state of nitrogen<sup>11</sup> and hydrogen<sup>12</sup> at corresponding temperatures by using REFPROP<sup>25</sup>. The uncertainty was estimated by applying the uncertainty propagation law. The results are also shown in Table 9.

$$B(T, x) = x_1^2 B_{11}(T) + 2x_1 x_2 B_{12}(T) + x_2^2 B_{22}(T)$$

(10)

The estimated values are compared with those calculated from the GERG-2008 in Figure 8. According to the theory, the interaction virial coefficients are independent of composition, therefore  $B_{12}(T)$  only depends on temperature. Taking into account the uncertainty estimated for  $B_{12}(T)$ , the values calculated from the experimental data agree with that.

Figure 9 shows  $B_{12}(T)$  as a function of temperature. The average of the calculated  $B_{12}(T)$  values for the system ( $N_2 + H_2$ ) was also compared to those data reported by other authors<sup>3,7,28-30</sup> at temperatures from (220 to 350) K. The plot in Figure 9 shows that there is good coincidence of the data with those from the literature, given the expected uncertainty of both data sets, except at lower temperatures for the values given by Zandbergen and Beenakker<sup>30</sup>.

## 5. Conclusions

Accurate ( $p, \rho, T$ ) experimental data for three binary mixtures of nitrogen and hydrogen, with nominal compositions of (0.95  $N_2 + 0.05 H_2$ ), (0.90  $N_2 + 0.10 H_2$ ), and (0.50  $N_2 + 0.50 H_2$ ), were obtained at temperatures between 240 K and 350 K and pressures up to 20 MPa using a single-sinker densimeter with magnetic suspension coupling. The mixtures were prepared gravimetrically at the Federal Institute for Materials Research and Testing (BAM) in Berlin, Germany. Experimental data were compared with the corresponding densities calculated from the GERG-2008 and the AGA8-DC92 EoS.

The relative deviations of the experimental data from both the GERG-2008 and the AGA8-DC92 EoS were within the  $\pm 0.1$  % band, which is the estimated uncertainty value of both equations at the temperature and pressure ranges evaluated, except for the (0.50  $N_2 + 0.50 H_2$ ) mixture at the lower temperature measured (240 K) and pressures from (14 to 19) MPa, which present deviations slightly larger than 0.1 % from the GERG-2008 EoS. Therefore, it can be concluded that experimental data agree very well with the values estimated by both equations of state.

Additionally, the second  $B(T, x)$ , the third  $C(T, x)$ , and the fourth  $D(T, x)$  virial coefficients of each mixture as well as the second interaction virial coefficient  $B_{12}(T)$  for the ( $N_2 + H_2$ ) binary system were

estimated, with their corresponding uncertainties, from the experimental data set at the studied temperature range.

### **Acknowledgments**

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## Figure captions

**Fig. 1.** Relative deviations in density of experimental ( $p, \rho, T$ ) data of the (0.94999954 N<sub>2</sub> + 0.05000022 H<sub>2</sub>) mixture  $\rho_{\text{exp}}$  from density values calculated from the GERG-2008 equation of state  $\rho_{\text{GERG}}$  versus pressure:  $\square$   $T = 240$  K;  $\diamond$   $T = 250$  K;  $\triangle$   $T = 260$  K;  $\times$   $T = 275$  K;  $+$   $T = 300$  K;  $\circ$   $T = 325$  K;  $*$   $T = 350$  K. Error bars on the 240 K isotherm indicate the expanded uncertainty ( $k = 2$ ) of the experimental density data calculated with Eq. (4).

**Fig. 2.** Relative deviations in density of experimental ( $p, \rho, T$ ) data of the (0.94999954 N<sub>2</sub> + 0.05000022 H<sub>2</sub>) mixture  $\rho_{\text{exp}}$  from density values calculated from the AGA8-DC92 equation of state  $\rho_{\text{AGA}}$  versus pressure:  $\square$   $T = 240$  K;  $\diamond$   $T = 250$  K;  $\triangle$   $T = 260$  K;  $\times$   $T = 275$  K;  $+$   $T = 300$  K;  $\circ$   $T = 325$  K;  $*$   $T = 350$  K. Error bars on the 240 K isotherm indicate the expanded uncertainty ( $k = 2$ ) of the experimental density data calculated with Eq. (4).

**Fig. 3.** Relative deviations in density of experimental ( $p, \rho, T$ ) data of the (0.90000685 N<sub>2</sub> + 0.09999291 H<sub>2</sub>) mixture  $\rho_{\text{exp}}$  from density values calculated from the GERG-2008 equation of state  $\rho_{\text{GERG}}$  versus pressure:  $\square$   $T = 240$  K;  $\diamond$   $T = 250$  K;  $\triangle$   $T = 260$  K;  $\times$   $T = 275$  K;  $+$   $T = 300$  K;  $\circ$   $T = 325$  K;  $*$   $T = 350$  K. Error bars on the 240 K isotherm indicate the expanded uncertainty ( $k = 2$ ) of the experimental density data calculated with Eq. (4).

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**Fig. 5.** Relative deviations in density of experimental ( $p, \rho, T$ ) data of the (0.50008195 N<sub>2</sub> + 0.49991771 H<sub>2</sub>) mixture  $\rho_{\text{exp}}$  from density values calculated from the GERG-2008 equation of state  $\rho_{\text{GERG}}$  versus pressure:  $\square$   $T = 240$  K;  $\diamond$   $T = 250$  K;  $\times$   $T = 275$  K;  $+$   $T = 300$  K;  $\circ$   $T = 325$  K;  $*$   $T = 350$  K. Error bars on the 240 K isotherm indicate the expanded uncertainty ( $k = 2$ ) of the experimental density data calculated with Eq. (4).

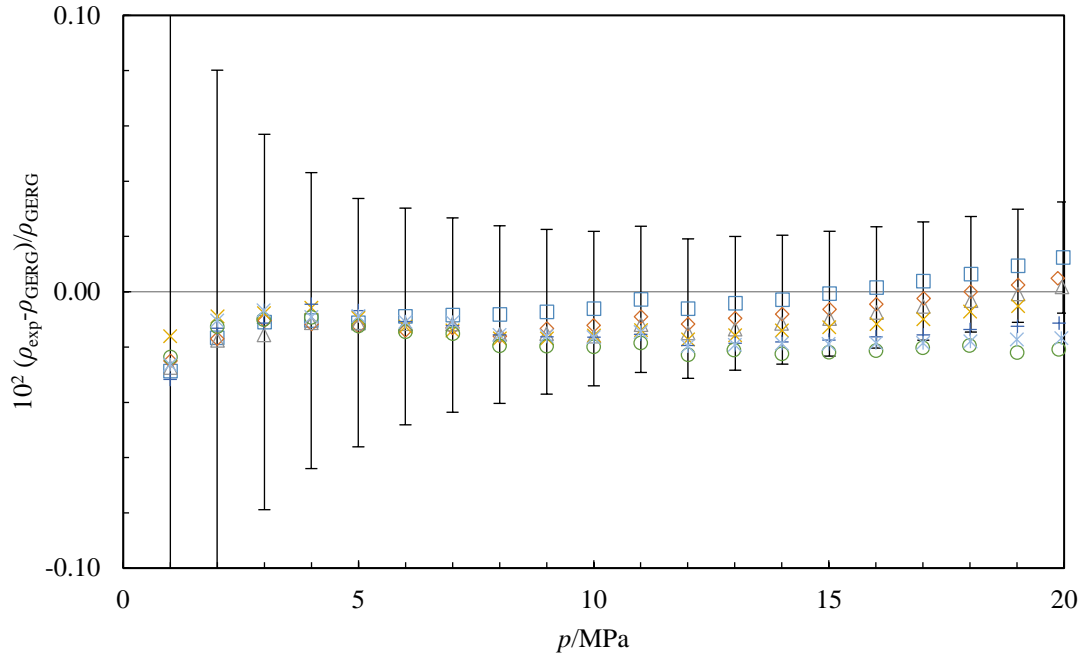
**Fig. 6.** Relative deviations in density of experimental ( $p, \rho, T$ ) data of the (0.50008195 N<sub>2</sub> + 0.49991771 H<sub>2</sub>) mixture  $\rho_{\text{exp}}$  from density values calculated from the AGA-DC92 equation of state  $\rho_{\text{AGA}}$  versus pressure:  $\square$   $T = 240$  K;  $\diamond$   $T = 250$  K;  $\times$   $T = 275$  K;  $+$   $T = 300$  K;  $\circ$   $T = 325$  K;  $*$   $T = 350$  K. Error bars on the 240 K isotherm indicate the expanded uncertainty ( $k = 2$ ) of the experimental density data calculated with Eq. (4).

**Fig. 7.** Comparison of the relative deviations in density from the GERG-2008 EoS of experimental ( $p, \rho, T$ ) data of the (0.50008195 N<sub>2</sub> + 0.49991771 H<sub>2</sub>) mixture and literature data of a (0.4998 N<sub>2</sub> + 0.5002 H<sub>2</sub>) mixture by Jaeschke *et al.*<sup>18</sup>. This work:  $\square$   $T = 240$  K;  $\diamond$   $T = 250$  K;  $\times$   $T = 275$  K;  $+$   $T = 300$  K;  $\circ$   $T = 325$  K;  $*$   $T = 350$  K. Jaeschke *et al.*:  $\blacksquare$   $T = 270$  K;  $\blacklozenge$   $T = 290$  K;  $\blacktriangle$   $T = 310$  K;  $\bullet$   $T = 330$  K.

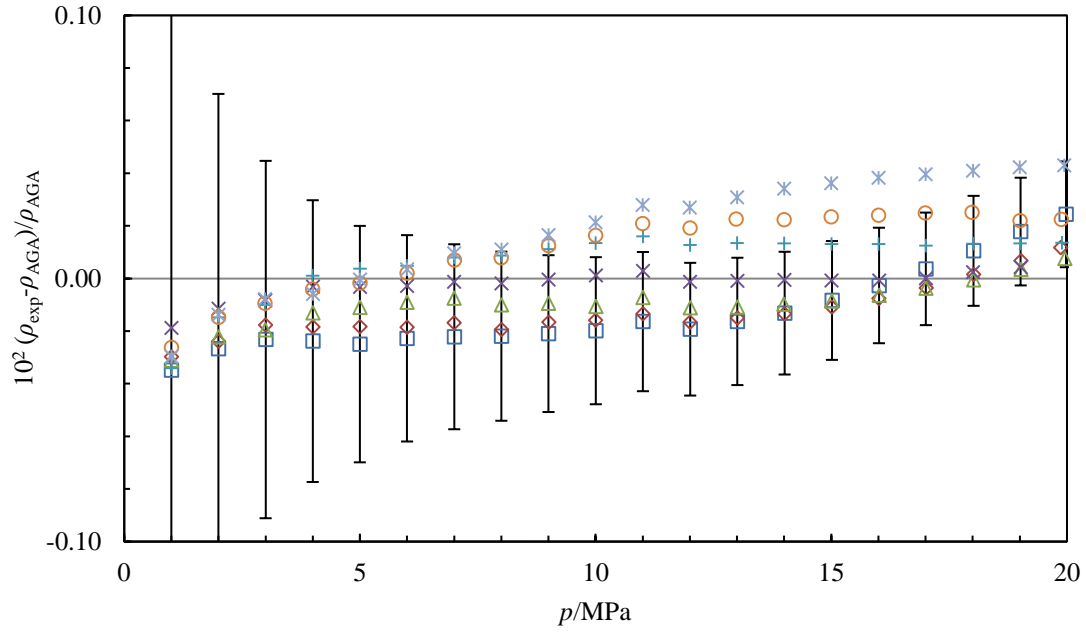
**Fig. 8.** The second interaction virial coefficient  $B_{12}(T)$  for the (N<sub>2</sub> + H<sub>2</sub>) binary system estimated from the experimental data:  $\square$   $T = 240$  K;  $\diamond$   $T = 250$  K;  $\triangle$   $T = 260$  K;  $\times$   $T = 275$  K;  $+$   $T = 300$  K;  $\circ$   $T = 325$  K;  $*$   $T = 350$  K. The dashed lines represent the  $B_{12}(T)$  values estimated from the GERG-2008 EoS at different temperatures.

**Fig. 9.** The second interaction virial coefficient  $B_{12}(T)$  for the (N<sub>2</sub> + H<sub>2</sub>) binary mixture:  $\blacksquare$  This work;  $\circ$  Jaeschke *et al.*<sup>3</sup>;  $\times$  Verschoyle<sup>7</sup>;  $\diamond$  Brewer and Vaughn<sup>28</sup>;  $\triangle$  Edwards and Roseveare<sup>29</sup>;  $+$  Zandbergen and Beenakker<sup>30</sup>. The solid line represents the linear fit to experimental data of this work:

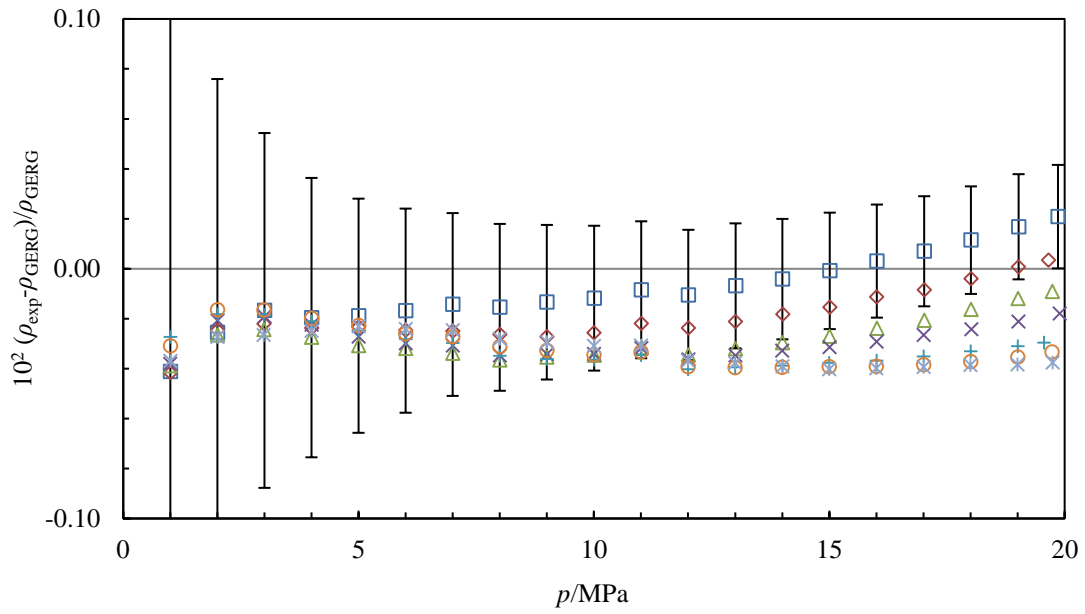
$$B_{12}(T)/\text{cm}^3 \cdot \text{mol}^{-1} = 0.063 \cdot T - 5.94.$$



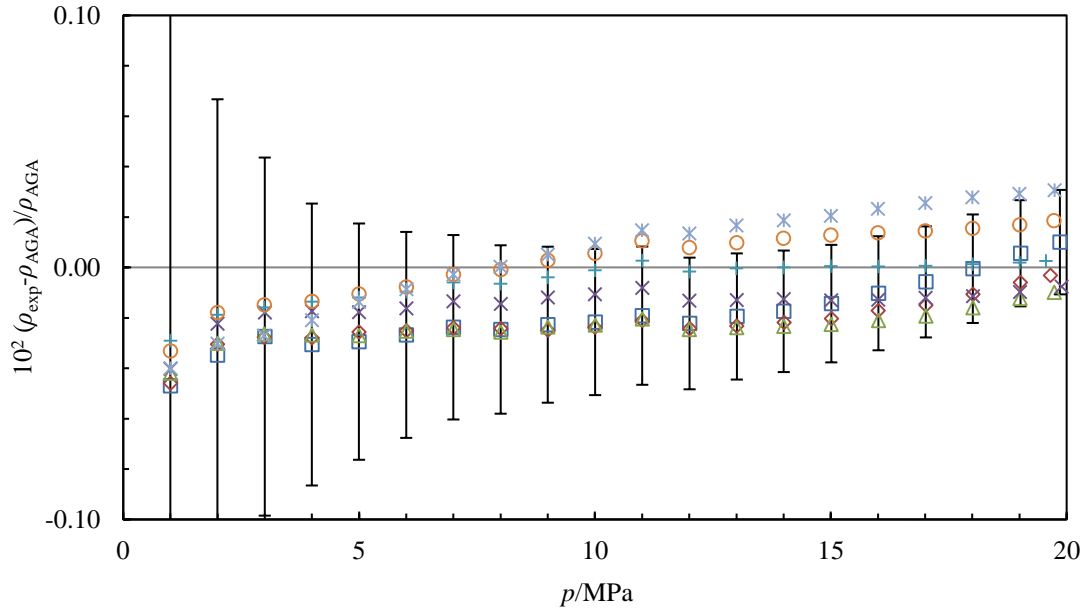
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**Fig. 2.** Relative deviations in density of experimental ( $p, \rho, T$ ) data of the (0.94999954 N<sub>2</sub> + 0.05000022 H<sub>2</sub>) mixture  $\rho_{\text{exp}}$  from density values calculated from the AGA8-DC92 equation of state  $\rho_{\text{AGA}}$  versus pressure:  $\square$   $T = 240$  K;  $\diamond$   $T = 250$  K;  $\triangle$   $T = 260$  K;  $\times$   $T = 275$  K;  $+$   $T = 300$  K;  $\circ$   $T = 325$  K;  $*$   $T = 350$  K. Error bars on the 240 K isotherm indicate the expanded uncertainty ( $k = 2$ ) of the experimental density data calculated with Eq. (4).

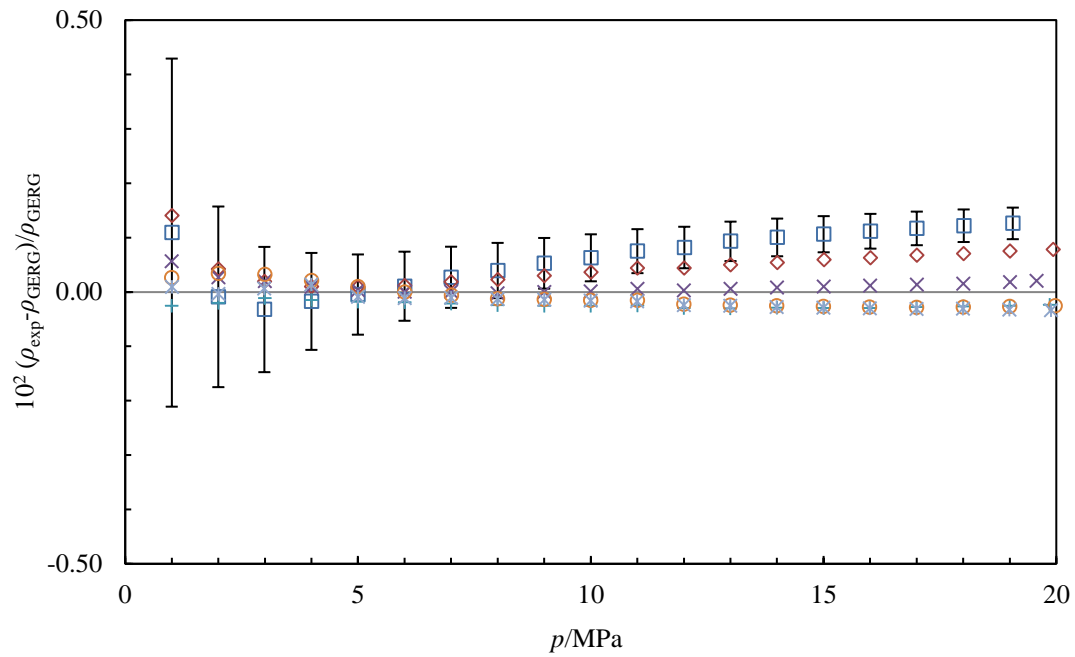


**Fig. 3.** Relative deviations in density of experimental ( $\rho$ ,  $\rho$ ,  $T$ ) data of the (0.90000685 N<sub>2</sub> + 0.09999291 H<sub>2</sub>) mixture  $\rho_{\text{exp}}$  from density values calculated from the GERG-2008 equation of state  $\rho_{\text{GERG}}$  versus pressure:  $\square$   $T = 240$  K;  $\diamond$   $T = 250$  K;  $\triangle$   $T = 260$  K;  $\times$   $T = 275$  K;  $+$   $T = 300$  K;  $\circ$   $T = 325$  K;  $*$   $T = 350$  K. Error bars on the 240 K isotherm indicate the expanded uncertainty ( $k = 2$ ) of the experimental density data calculated with Eq. (4).

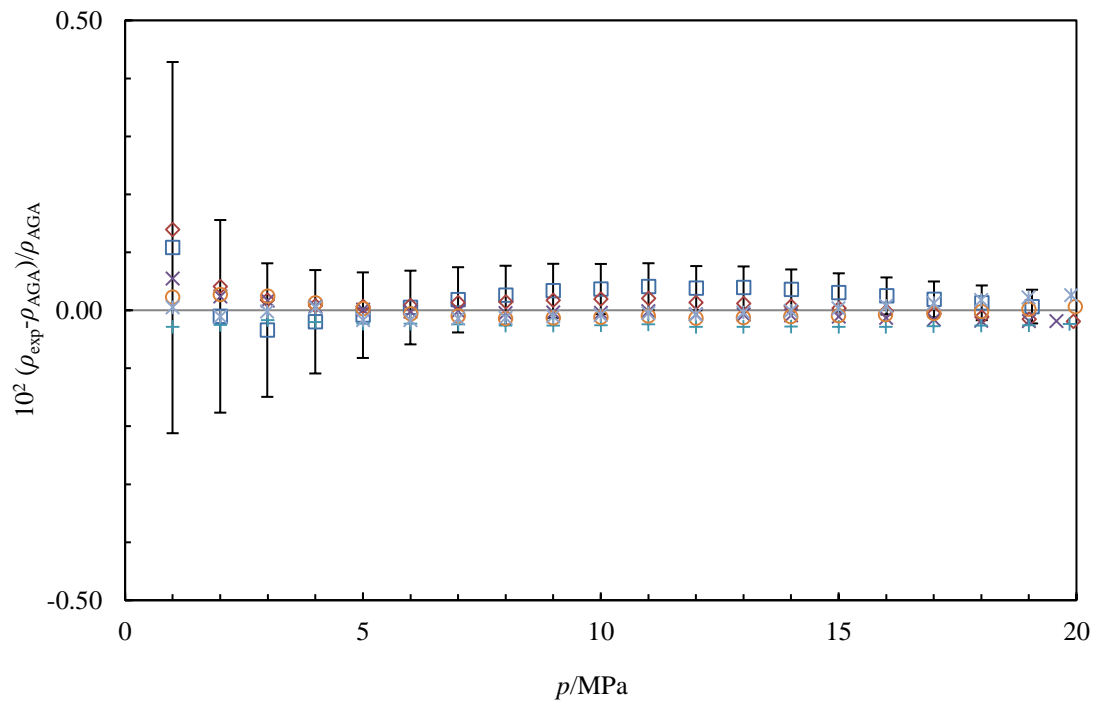


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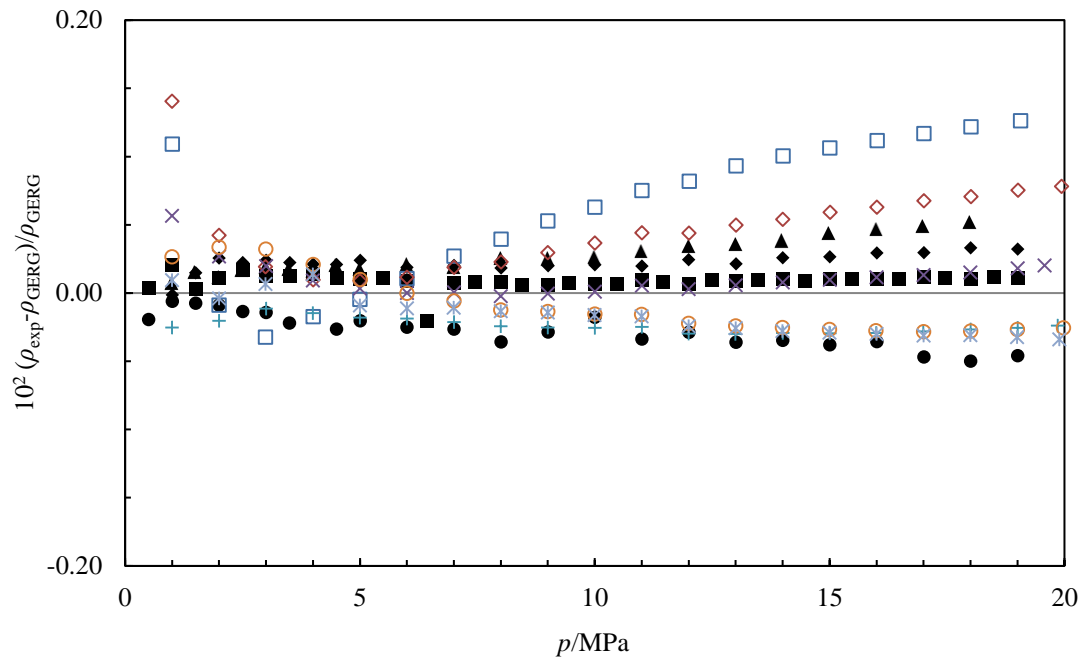




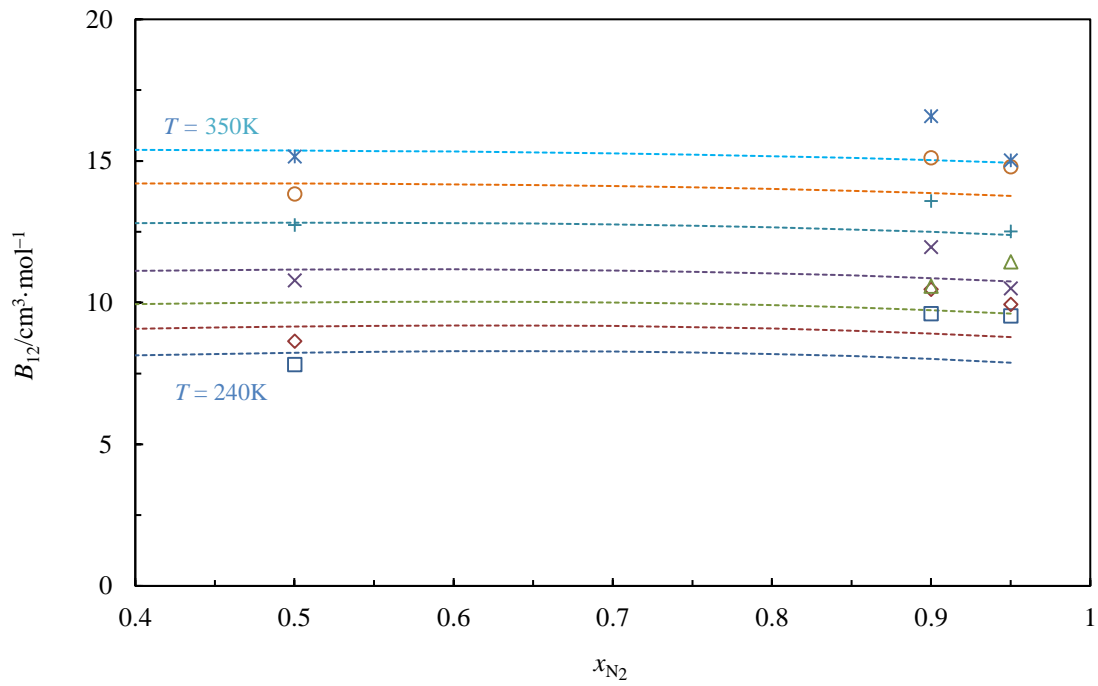
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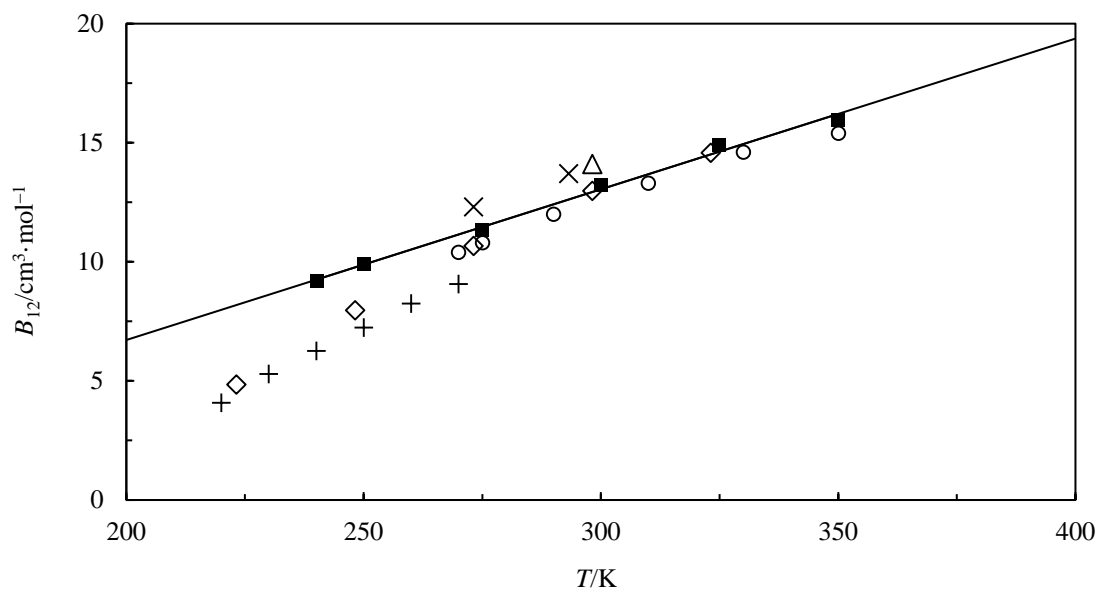
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**Fig. 7.** Comparison of the relative deviations in density from the GERG-2008 EoS of experimental  $(p, \rho, T)$  data of the  $(0.50008195 \text{ N}_2 + 0.49991771 \text{ H}_2)$  mixture and literature data of a  $(0.4998 \text{ N}_2 + 0.5002 \text{ H}_2)$  mixture by Jaeschke *et al.*<sup>18</sup>. This work:  $\square$   $T = 240 \text{ K}$ ;  $\diamond$   $T = 250 \text{ K}$ ;  $\times$   $T = 275 \text{ K}$ ;  $+$   $T = 300 \text{ K}$ ;  $\circ$   $T = 325 \text{ K}$ ;  $\ast$   $T = 350 \text{ K}$ . Jaeschke *et al.*:  $\blacksquare$   $T = 270 \text{ K}$ ;  $\blacklozenge$   $T = 290 \text{ K}$ ;  $\blacktriangle$   $T = 310 \text{ K}$ ;  $\bullet$   $T = 330 \text{ K}$ .



**Fig. 8.** The second interaction virial coefficient  $B_{12}(T)$  for the  $(N_2 + H_2)$  binary system estimated from the experimental data:  $\square$   $T = 240$  K;  $\diamond$   $T = 250$  K;  $\triangle$   $T = 260$  K;  $\times$   $T = 275$  K;  $+$   $T = 300$  K;  $\circ$   $T = 325$  K;  $*$   $T = 350$  K. The dashed lines represent the  $B_{12}(T)$  values estimated from the GERG-2008 EoS at different temperatures.



**Fig. 9.** The second interaction virial coefficient  $B_{12}(T)$  for the  $(\text{N}_2 + \text{H}_2)$  binary mixture: ■ This work; ○ Jaeschke *et al.*<sup>3</sup>; × Verschoyle<sup>7</sup>; ◇ Brewer and Vaughn<sup>28</sup>; △ Edwards and Roseveare<sup>29</sup>; + Zandbergen and Beenakker<sup>30</sup>. The solid line represents the linear fit to experimental data of this work:

$$B_{12}(T)/\text{cm}^3 \cdot \text{mol}^{-1} = 0.063 \cdot T - 5.94.$$

## Table captions

**Table 1.** Purity, supplier, molar mass, and critical parameters of the constituting components of the studied ( $\text{N}_2 + \text{H}_2$ ) mixtures.

**Table 2.** Composition of the studied ( $\text{N}_2 + \text{H}_2$ ) binary mixtures including information on impurities from the supplier.

**Table 3.** Results of the GC analysis and relative deviation between gravimetric composition and GC analysis for the ( $0.95 \text{ N}_2 + 0.05 \text{ H}_2$ ) and the ( $0.90 \text{ N}_2 + 0.10 \text{ H}_2$ ) binary mixtures.

**Table 4.** Contributions to the expanded overall uncertainty in density ( $k = 2$ )  $U_T(\rho_{\text{exp}})$  for the three studied ( $\text{N}_2 + \text{H}_2$ ) binary mixtures in the temperature range from (240 to 350) K.

**Table 5.** Experimental ( $p, \rho, T$ ) measurements for the ( $0.95 \text{ N}_2 + 0.05 \text{ H}_2$ ) mixture (actual composition given in Table 2), absolute and relative expanded uncertainty in density ( $k = 2$ )  $U(\rho_{\text{exp}})$ , and relative deviations from the GERG-2008 and AGA8-DC92 EoS; where  $T$  is the temperature (ITS-90),  $p$  the pressure,  $\rho_{\text{exp}}$  the experimental density, and  $\rho_{\text{GERG}}$  and  $\rho_{\text{AGA}}$  the densities calculated from the GERG-2008 and the AGA8-DC92 EoS.

**Table 6.** Experimental ( $p, \rho, T$ ) measurements for the ( $0.90 \text{ N}_2 + 0.10 \text{ H}_2$ ) mixture (actual composition given in Table 2), absolute and relative expanded uncertainty in density ( $k = 2$ )  $U(\rho_{\text{exp}})$ , and relative deviations from the GERG-2008 and AGA8-DC92 EoS; where  $T$  is the temperature (ITS-90),  $p$  the pressure,  $\rho_{\text{exp}}$  the experimental density, and  $\rho_{\text{GERG}}$  and  $\rho_{\text{AGA}}$  the densities calculated from the GERG-2008 and the AGA8-DC92 EoS.

**Table 7.** Experimental ( $p, \rho, T$ ) measurements for the ( $0.50 \text{ N}_2 + 0.50 \text{ H}_2$ ) mixture (actual composition given in Table 2), absolute and relative expanded uncertainty in density ( $k = 2$ )  $U(\rho_{\text{exp}})$  and relative deviations from the GERG-2008 and AGA8-DC92 EoS; where  $T$  is the temperature (ITS-90),  $p$  the pressure,  $\rho_{\text{exp}}$  the experimental density, and  $\rho_{\text{GERG}}$  and  $\rho_{\text{AGA}}$  the densities calculated from the GERG-2008 and the AGA8-DC92 EoS.

**Table 8.** Statistical parameters of the data set with respect to the GERG-2008 and AGA8-DC92 EoS for the (N<sub>2</sub> + H<sub>2</sub>) mixtures and literature comparison.

**Table 9.** Least-squares fit results for the three (N<sub>2</sub> + H<sub>2</sub>) mixtures. Virial coefficients (*B*, *C*, and *D*) and the second interaction virial coefficient (*B*<sub>12</sub>) with the corresponding expanded uncertainties (*k* = 2).

**Table 1.** Purity, supplier, molar mass, and critical parameters of the constituting components of the studied (N<sub>2</sub> + H<sub>2</sub>) mixtures.

Components	Purity	Supplier	$M/\text{g}\cdot\text{mol}^{-1}$	Critical parameters	
				$T_c/\text{K}$	$p_c/\text{MPa}$
Nitrogen	$\geq 99.9999$ mol-%	Linde <sup>a</sup>	28.013 <sup>b</sup>	126.192 <sup>b</sup>	3.396 <sup>b</sup>
Hydrogen	$\geq 99.9999$ mol-%	Linde <sup>a</sup>	2.016 <sup>c</sup>	33.145 <sup>c</sup>	1.296 <sup>c</sup>

<sup>a</sup> Linde AG, Unterschleißheim, Germany.

<sup>b</sup> Span *et al.* [11].

<sup>c</sup> Leachman *et al.* [12].



**Table 2.** Composition of the studied ( $\text{N}_2 + \text{H}_2$ ) binary mixtures including information on impurities from the supplier.

Component	(0.95 $\text{N}_2$ + 0.05 $\text{H}_2$ ) BAM no.: 96054 968-160517		(0.90 $\text{N}_2$ + 0.10 $\text{H}_2$ ) BAM no.: 96054 970-160501		(0.50 $\text{N}_2$ + 0.50 $\text{H}_2$ ) BAM no.: 96055 000-160426	
	$x_i$	$U(x_i) (k = 2)$	$x_i$	$U(x_i) (k = 2)$	$x_i$	$U(x_i) (k = 2)$
	( $10^{-2}$ mol·mol $^{-1}$ )	( $10^{-2}$ mol·mol $^{-1}$ )	( $10^{-2}$ mol·mol $^{-1}$ )	( $10^{-2}$ mol·mol $^{-1}$ )	( $10^{-2}$ mol·mol $^{-1}$ )	( $10^{-2}$ mol·mol $^{-1}$ )
Nitrogen	94.999954	0.001330	90.000685	0.001277	50.008195	0.000950
Hydrogen	5.000022	0.001583	9.999291	0.002738	49.991771	0.013201
Oxygen	0.000013	0.000013	0.000014	0.000012	0.000019	0.000016
Carbon dioxide	0.000005	0.000005	0.000006	0.000005	0.000008	0.000007
Carbon monoxide	0.000005	0.000005	0.000006	0.000005	0.000008	0.000007

**Table 3.** Results of the GC analysis and relative deviation between gravimetric composition and GC analysis for the (0.95 N<sub>2</sub> + 0.05 H<sub>2</sub>) and the (0.90 N<sub>2</sub> + 0.10 H<sub>2</sub>) binary mixtures.

Component	(0.95 N <sub>2</sub> + 0.05 H <sub>2</sub> ) BAM no.: 96054 968-160517			(0.90 N <sub>2</sub> + 0.10 H <sub>2</sub> ) BAM no.: 96054 970-160501		
	Concentration		Relative deviation between gravimetric preparation and GC analysis	Concentration		Relative deviation between gravimetric preparation and GC analysis
	$x_i$ (10 <sup>-2</sup> mol·mol <sup>-1</sup> )	$U(x_i)$ (k = 2) (10 <sup>-2</sup> mol·mol <sup>-1</sup> )	%	$x_i$ (10 <sup>-2</sup> mol·mol <sup>-1</sup> )	$U(x_i)$ (k = 2) (10 <sup>-2</sup> mol·mol <sup>-1</sup> )	%
Nitrogen	94.9961	0.0188	-0.004	90.0063	0.0190	0.006
Hydrogen	4.9973	0.0232	-0.055	9.9921	0.0483	-0.072
	Validation mixture BAM no.: 96054 946-160530			Validation mixture BAM no.: 96054 985-160524		
Nitrogen	94.999948	0.002679		90.001044	0.002699	
Hydrogen	5.000028	0.002370		9.998931	0.004413	
Oxygen	0.000013	0.000012		0.000013	0.000011	
Carbon dioxide	0.000005	0.000005		0.000006	0.000005	

Carbon monoxide

0.000005

0.000005

0.000006

0.000004

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**Table 4.** Contributions to the expanded overall uncertainty in density ( $k = 2$ )  $U_T(\rho_{\text{exp}})$  for the three studied ( $\text{N}_2 + \text{H}_2$ ) binary mixtures in the temperature range from (240 to 350) K.

Source of uncertainty	Units	Contribution ( $k = 2$ )	Estimation in density ( $k = 2$ )	
			$\text{kg}\cdot\text{m}^{-3}$	%
(0.95 $\text{N}_2$ + 0.05 $\text{H}_2$ )				
Temperature $T$	K	0.004	< 0.008	< 0.004
Pressure $p$	MPa	0.005	< 0.064	(0.004 - 0.11)
Density $\rho$	$\text{kg}\cdot\text{m}^{-3}$	(0.024 - 0.054)	(0.024 - 0.054)	(0.02 - 0.27)
Composition $x_i$	$\text{mol}\cdot\text{mol}^{-1}$	< 0.0001	< 0.032	< 0.006
Overall uncertainty $U_T(\rho_{\text{exp}})$			(0.024 - 0.078)	(0.027 - 0.33)
(0.90 $\text{N}_2$ + 0.10 $\text{H}_2$ )				
Temperature $T$	K	0.004	< 0.005	< 0.002
Pressure $p$	MPa	0.005	< 0.056	(0.006 - 0.18)
Density $\rho$	$\text{kg}\cdot\text{m}^{-3}$	(0.024 - 0.052)	(0.024 - 0.052)	(0.021 - 0.28)
Composition $x_i$	$\text{mol}\cdot\text{mol}^{-1}$	< 0.0001	< 0.027	< 0.011
Overall uncertainty $U_T(\rho_{\text{exp}})$			(0.024 - 0.079)	(0.032 - 0.44)
(0.50 $\text{N}_2$ + 0.50 $\text{H}_2$ )				
Temperature $T$	K	0.004	< 0.002	< 0.002
Pressure $p$	MPa	0.005	< 0.030	(0.006 - 0.18)
Density $\rho$	$\text{kg}\cdot\text{m}^{-3}$	(0.024 - 0.038)	(0.024 - 0.038)	(0.03 - 0.46)
Composition $x_i$	$\text{mol}\cdot\text{mol}^{-1}$	< 0.0001	< 0.018	< 0.014
Overall uncertainty $U_T(\rho_{\text{exp}})$			(0.024 - 0.052)	(0.039 - 0.56)

**Table 5.** Experimental ( $p$ ,  $\rho$ ,  $T$ ) measurements for the (0.95 N<sub>2</sub> + 0.05 H<sub>2</sub>) mixture (actual composition given in Table 2), absolute and relative expanded uncertainty in density ( $k = 2$ )  $U(\rho_{\text{exp}})$ , and relative deviations from the GERG-2008 and AGA8-DC92 EoS; where  $T$  is the temperature (ITS-90),  $p$  the pressure,  $\rho_{\text{exp}}$  the experimental density, and  $\rho_{\text{GERG}}$  and  $\rho_{\text{AGA}}$  the densities calculated from the GERG-2008 and the AGA8-DC92 EoS.

$T/\text{K}^a$	$p/\text{MPa}^a$	$\rho_{\text{exp}}/\text{kg}\cdot\text{m}^{-3}$	$U(\rho_{\text{exp}}) / \text{kg}\cdot\text{m}^{-3}$	$10^2 U(\rho_{\text{exp}}) / \rho_{\text{exp}}$	$10^2 (\rho_{\text{exp}} - \rho_{\text{GERG}}) / \rho_{\text{GERG}}$	$10^2 (\rho_{\text{exp}} - \rho_{\text{AGA}}) / \rho_{\text{AGA}}$
240.046	19.977	266.493	0.054	0.020	0.012	0.024
240.047	19.018	255.783	0.052	0.020	0.009	0.018
240.047	18.019	244.268	0.051	0.021	0.006	0.011
240.047	17.007	232.236	0.050	0.021	0.004	0.004
240.047	16.011	220.048	0.048	0.022	0.002	-0.003
240.046	15.014	207.511	0.047	0.023	-0.001	-0.008
240.047	14.010	194.555	0.045	0.023	-0.003	-0.013
240.047	13.008	181.315	0.044	0.024	-0.004	-0.016
240.047	12.006	167.817	0.042	0.025	-0.006	-0.019
240.048	11.004	154.093	0.041	0.026	-0.003	-0.016
240.047	10.004	140.187	0.039	0.028	-0.006	-0.020
240.048	9.001	126.079	0.038	0.030	-0.007	-0.021
240.049	8.000	111.894	0.036	0.032	-0.008	-0.022
240.049	7.000	97.671	0.034	0.035	-0.008	-0.022
240.049	5.999	83.412	0.033	0.039	-0.009	-0.023
240.048	4.998	69.190	0.031	0.045	-0.011	-0.025
240.049	3.998	55.055	0.029	0.054	-0.010	-0.024
240.049	2.999	41.055	0.028	0.068	-0.011	-0.023
240.051	1.997	27.150	0.026	0.097	-0.017	-0.027
240.051	0.998	13.460	0.025	0.184	-0.029	-0.035

250.023	19.863	251.120	0.052	0.021	0.005	0.012
250.021	19.020	242.075	0.051	0.021	0.002	0.007
250.021	18.015	231.002	0.050	0.021	< 0.001	0.001
250.021	17.012	219.644	0.048	0.022	-0.002	-0.004
250.021	16.008	207.967	0.047	0.023	-0.004	-0.008
250.020	15.009	196.059	0.046	0.023	-0.006	-0.011
250.020	14.006	183.836	0.044	0.024	-0.008	-0.013
250.020	13.002	171.330	0.043	0.025	-0.010	-0.015
250.020	12.002	158.648	0.041	0.026	-0.012	-0.017
250.019	11.003	145.769	0.040	0.027	-0.009	-0.013
250.018	9.998	132.641	0.038	0.029	-0.012	-0.016
250.019	9.002	119.485	0.037	0.031	-0.013	-0.017
250.018	8.001	106.147	0.035	0.033	-0.016	-0.019
250.019	6.999	92.739	0.034	0.036	-0.013	-0.017
250.018	5.999	79.318	0.032	0.041	-0.014	-0.019
250.018	4.999	65.908	0.031	0.047	-0.013	-0.018
250.018	3.999	52.525	0.029	0.056	-0.012	-0.018
250.018	2.999	39.211	0.028	0.071	-0.010	-0.018
250.019	1.998	25.981	0.026	0.101	-0.017	-0.024
250.018	0.998	12.909	0.025	0.191	-0.025	-0.030
260.020	19.954	239.529	0.051	0.021	0.002	0.008
260.019	19.020	229.930	0.049	0.021	-0.001	0.003
260.019	18.018	219.361	0.048	0.022	-0.003	-0.001
260.020	17.003	208.387	0.047	0.023	-0.005	-0.004
260.020	16.012	197.415	0.046	0.023	-0.007	-0.006
260.019	15.009	186.058	0.044	0.024	-0.010	-0.009
260.021	13.989	174.256	0.043	0.025	-0.012	-0.010
260.019	13.007	162.680	0.042	0.026	-0.013	-0.011

260.020	12.004	150.650	0.040	0.027	-0.015	-0.011
260.019	11.004	138.476	0.039	0.028	-0.012	-0.007
260.018	10.003	126.121	0.038	0.030	-0.016	-0.011
260.018	8.999	113.598	0.036	0.032	-0.015	-0.009
260.018	8.004	101.083	0.035	0.034	-0.015	-0.010
260.020	6.997	88.348	0.033	0.038	-0.012	-0.007
260.020	5.998	75.652	0.032	0.042	-0.012	-0.009
260.020	4.998	62.921	0.030	0.048	-0.011	-0.011
260.020	3.998	50.206	0.029	0.058	-0.011	-0.013
260.021	2.987	37.390	0.027	0.073	-0.016	-0.019
260.019	1.998	24.915	0.026	0.105	-0.018	-0.022
260.020	0.999	12.396	0.025	0.199	-0.027	-0.031
274.995	20.152	225.084	0.049	0.022	-0.002	0.008
274.995	19.017	214.131	0.048	0.022	-0.005	0.004
274.995	18.001	204.074	0.046	0.023	-0.007	0.002
274.993	17.006	194.006	0.045	0.023	-0.010	< 0.001
274.993	16.011	183.720	0.044	0.024	-0.012	-0.001
274.993	15.002	173.076	0.043	0.025	-0.013	-0.001
274.993	14.000	162.308	0.042	0.026	-0.014	-0.001
274.992	13.002	151.394	0.040	0.027	-0.016	-0.001
274.992	12.003	140.289	0.039	0.028	-0.017	-0.001
274.991	11.002	129.012	0.038	0.029	-0.014	0.003
274.991	10.001	117.586	0.037	0.031	-0.016	0.001
274.991	9.000	106.023	0.035	0.033	-0.017	< 0.001
274.990	8.000	94.385	0.034	0.036	-0.017	-0.002
274.990	6.998	82.636	0.033	0.039	-0.014	-0.001
274.990	5.998	70.840	0.031	0.044	-0.013	-0.003
274.991	4.997	58.999	0.030	0.051	-0.010	-0.003

274.991	3.997	47.147	0.029	0.061	-0.006	-0.003
274.991	2.986	35.164	0.027	0.077	-0.008	-0.008
274.991	1.998	23.478	0.026	0.110	-0.009	-0.012
274.990	0.997	11.688	0.025	0.210	-0.016	-0.019
299.938	19.889	200.449	0.046	0.023	-0.011	0.014
299.939	19.003	192.638	0.045	0.023	-0.012	0.013
299.940	18.000	183.626	0.044	0.024	-0.014	0.013
299.940	17.005	174.510	0.043	0.025	-0.016	0.013
299.939	16.000	165.148	0.042	0.025	-0.016	0.013
299.939	14.999	155.650	0.041	0.026	-0.017	0.013
299.940	14.000	146.008	0.040	0.027	-0.018	0.013
299.940	13.002	136.232	0.039	0.028	-0.019	0.013
299.940	12.000	126.269	0.038	0.030	-0.019	0.013
299.940	10.999	116.189	0.036	0.031	-0.015	0.016
299.939	10.000	106.004	0.035	0.033	-0.016	0.013
299.940	9.000	95.695	0.034	0.036	-0.016	0.011
299.940	7.999	85.271	0.033	0.039	-0.016	0.009
299.938	6.999	74.769	0.032	0.042	-0.012	0.008
299.938	5.998	64.189	0.031	0.048	-0.011	0.005
299.940	4.997	53.549	0.029	0.055	-0.007	0.004
299.939	3.997	42.862	0.028	0.066	-0.005	0.001
299.939	2.984	32.013	0.027	0.084	-0.011	-0.010
299.939	1.998	21.430	0.026	0.120	-0.013	-0.015
299.929	0.997	10.683	0.024	0.229	-0.032	-0.034
324.938	19.880	182.728	0.044	0.024	-0.021	0.022
324.939	19.001	175.603	0.043	0.025	-0.022	0.022
324.940	17.986	167.256	0.042	0.025	-0.019	0.025



324.943	16.993	158.937	0.041	0.026	-0.020	0.025
324.945	15.998	150.466	0.040	0.027	-0.021	0.024
324.945	14.997	141.813	0.039	0.028	-0.022	0.023
324.947	13.998	133.041	0.038	0.029	-0.022	0.022
324.947	12.979	123.971	0.037	0.030	-0.021	0.023
324.945	12.002	115.150	0.036	0.032	-0.023	0.019
324.947	10.996	105.963	0.035	0.033	-0.019	0.021
324.946	9.999	96.736	0.034	0.035	-0.020	0.016
324.947	8.999	87.379	0.033	0.038	-0.020	0.012
324.946	7.997	77.913	0.032	0.041	-0.020	0.008
324.948	7.000	68.408	0.031	0.045	-0.015	0.007
324.947	5.997	58.767	0.030	0.051	-0.014	0.002
324.947	4.997	49.080	0.029	0.059	-0.012	-0.002
324.947	3.997	39.338	0.028	0.070	-0.009	-0.004
324.947	2.984	29.414	0.027	0.090	-0.010	-0.009
324.946	1.998	19.717	0.025	0.129	-0.013	-0.015
324.946	0.998	9.855	0.024	0.247	-0.024	-0.026
349.935	19.940	168.761	0.042	0.025	-0.017	0.043
349.935	18.992	161.661	0.042	0.026	-0.017	0.042
349.936	18.002	154.124	0.041	0.026	-0.018	0.041
349.935	16.996	146.347	0.040	0.027	-0.018	0.040
349.934	15.998	138.524	0.039	0.028	-0.018	0.038
349.934	14.993	130.518	0.038	0.029	-0.019	0.036
349.934	13.999	122.495	0.037	0.030	-0.019	0.034
349.934	12.999	114.314	0.036	0.032	-0.019	0.031
349.935	11.995	105.989	0.035	0.033	-0.020	0.027
349.935	10.998	97.623	0.034	0.035	-0.015	0.028
349.935	9.999	89.144	0.033	0.037	-0.016	0.021

349.935	8.999	80.553	0.032	0.040	-0.016	0.017
349.935	7.998	71.871	0.031	0.044	-0.016	0.011
349.936	6.996	63.099	0.030	0.048	-0.011	0.010
349.936	5.998	54.273	0.029	0.054	-0.011	0.004
349.935	4.998	45.368	0.028	0.063	-0.009	< 0.001
349.935	3.997	36.385	0.027	0.075	-0.009	-0.006
349.934	2.982	27.214	0.026	0.097	-0.007	-0.008
349.936	1.998	18.271	0.025	0.138	-0.010	-0.014
349.935	0.997	9.137	0.024	0.265	-0.026	-0.030

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<sup>a</sup> Expanded uncertainties ( $k = 2$ ) in temperature and pressure are  $U(T) = 0.004$  K and  $U(p) = 0.005$  MPa, respectively.

**Table 6.** Experimental ( $p$ ,  $\rho$ ,  $T$ ) measurements for the (0.90 N<sub>2</sub> + 0.10 H<sub>2</sub>) mixture (actual composition given in Table 2), absolute and relative expanded uncertainty in density ( $k = 2$ )  $U(\rho_{\text{exp}})$ , and relative deviations from the GERG-2008 and AGA8-DC92 EoS; where  $T$  is the temperature (ITS-90),  $p$  the pressure,  $\rho_{\text{exp}}$  the experimental density, and  $\rho_{\text{GERG}}$  and  $\rho_{\text{AGA}}$  the densities calculated from the GERG-2008 and the AGA8-DC92 EoS.

$T/\text{K}^a$	$p/\text{MPa}^a$	$\rho_{\text{exp}}/\text{kg}\cdot\text{m}^{-3}$	$U(\rho_{\text{exp}}) / \text{kg}\cdot\text{m}^{-3}$	$10^2 U(\rho_{\text{exp}}) / \rho_{\text{exp}}$	$10^2 (\rho_{\text{exp}} - \rho_{\text{GERG}}) / \rho_{\text{GERG}}$	$10^2 (\rho_{\text{exp}} - \rho_{\text{AGA}}) / \rho_{\text{AGA}}$
240.067	19.857	248.956	0.052	0.021	0.021	0.010
240.067	19.021	240.134	0.051	0.021	0.017	0.006
240.067	18.009	229.140	0.049	0.022	0.011	-0.001
240.067	17.016	218.039	0.048	0.022	0.007	-0.006
240.067	16.012	206.497	0.047	0.023	0.003	-0.010
240.067	15.010	194.680	0.045	0.023	-0.001	-0.014
240.069	14.004	182.528	0.044	0.024	-0.004	-0.017
240.068	13.007	170.235	0.043	0.025	-0.007	-0.019
240.067	12.006	157.633	0.041	0.026	-0.011	-0.022
240.069	11.003	144.806	0.040	0.027	-0.008	-0.019
240.069	10.003	131.826	0.038	0.029	-0.012	-0.022
240.070	9.001	118.665	0.037	0.031	-0.013	-0.023
240.069	8.000	105.416	0.035	0.033	-0.015	-0.025
240.069	6.999	92.089	0.034	0.037	-0.014	-0.024
240.069	5.999	78.743	0.032	0.041	-0.017	-0.027
240.070	4.998	65.397	0.031	0.047	-0.019	-0.029
240.070	3.998	52.100	0.029	0.056	-0.020	-0.031
240.070	3.000	38.902	0.028	0.071	-0.017	-0.027
240.071	1.998	25.761	0.026	0.101	-0.026	-0.035
240.072	0.998	12.789	0.025	0.193	-0.041	-0.047

250.045	19.653	234.021	0.050	0.021	0.003	-0.003
250.047	19.014	227.523	0.049	0.022	0.001	-0.006
250.044	18.011	217.113	0.048	0.022	-0.004	-0.011
250.044	17.014	206.478	0.047	0.023	-0.008	-0.015
250.046	16.004	195.446	0.045	0.023	-0.011	-0.017
250.045	15.008	184.307	0.044	0.024	-0.015	-0.020
250.047	14.008	172.872	0.043	0.025	-0.018	-0.022
250.044	13.005	161.179	0.042	0.026	-0.021	-0.023
250.042	12.002	149.273	0.040	0.027	-0.024	-0.024
250.042	11.001	137.200	0.039	0.028	-0.022	-0.021
250.043	9.995	124.895	0.037	0.030	-0.026	-0.024
250.041	9.001	112.600	0.036	0.032	-0.027	-0.025
250.044	8.000	100.114	0.035	0.035	-0.026	-0.024
250.043	6.999	87.557	0.033	0.038	-0.025	-0.024
250.042	5.999	74.953	0.032	0.042	-0.025	-0.025
250.043	4.998	62.325	0.030	0.049	-0.023	-0.026
250.041	3.998	49.722	0.029	0.058	-0.024	-0.028
250.041	2.987	37.024	0.027	0.074	-0.022	-0.028
250.039	1.998	24.659	0.026	0.105	-0.025	-0.030
250.042	0.998	12.260	0.025	0.201	-0.042	-0.046
260.042	19.736	223.429	0.049	0.022	-0.009	-0.010
260.044	19.010	216.361	0.048	0.022	-0.012	-0.012
260.041	18.009	206.415	0.047	0.023	-0.016	-0.016
260.041	17.014	196.279	0.046	0.023	-0.021	-0.019
260.040	16.007	185.789	0.044	0.024	-0.024	-0.021
260.040	15.004	175.116	0.043	0.025	-0.027	-0.023
260.039	14.004	164.255	0.042	0.026	-0.030	-0.023

260.039	13.005	153.211	0.041	0.027	-0.032	-0.024
260.039	12.001	141.918	0.039	0.028	-0.035	-0.025
260.038	11.002	130.507	0.038	0.029	-0.032	-0.020
260.040	10.001	118.929	0.037	0.031	-0.035	-0.023
260.038	9.000	107.209	0.035	0.033	-0.035	-0.024
260.037	7.999	95.394	0.034	0.036	-0.037	-0.026
260.036	6.999	83.502	0.033	0.039	-0.034	-0.025
260.037	5.998	71.547	0.031	0.044	-0.032	-0.025
260.035	4.998	59.561	0.030	0.050	-0.031	-0.027
260.035	3.998	47.566	0.029	0.060	-0.027	-0.027
260.035	2.987	35.462	0.027	0.077	-0.024	-0.026
260.034	2.000	23.676	0.026	0.109	-0.027	-0.030
260.033	0.998	11.773	0.025	0.208	-0.039	-0.042
275.002	19.891	209.964	0.047	0.022	-0.018	-0.008
275.000	19.012	201.918	0.046	0.023	-0.021	-0.010
274.998	18.013	192.591	0.045	0.023	-0.024	-0.011
274.996	17.005	182.969	0.044	0.024	-0.027	-0.012
274.996	16.004	173.220	0.043	0.025	-0.029	-0.013
274.996	15.005	163.298	0.042	0.026	-0.031	-0.013
274.995	13.999	153.123	0.041	0.027	-0.033	-0.013
274.992	13.002	142.866	0.039	0.028	-0.035	-0.013
274.992	12.001	132.405	0.038	0.029	-0.036	-0.013
274.991	11.001	121.821	0.037	0.030	-0.032	-0.008
274.991	9.998	111.047	0.036	0.032	-0.034	-0.011
274.989	9.000	100.212	0.035	0.035	-0.034	-0.012
274.988	7.998	89.232	0.033	0.037	-0.035	-0.014
274.987	6.998	78.183	0.032	0.041	-0.031	-0.013
274.986	5.998	67.065	0.031	0.046	-0.030	-0.016

274.985	4.998	55.901	0.030	0.053	-0.027	-0.018
274.985	3.997	44.699	0.028	0.063	-0.022	-0.017
274.985	2.997	33.490	0.027	0.081	-0.019	-0.018
274.984	1.997	22.285	0.026	0.116	-0.021	-0.022
274.982	0.997	11.103	0.024	0.220	-0.038	-0.040
299.925	19.557	186.761	0.044	0.024	-0.030	0.003
299.927	19.002	182.110	0.044	0.024	-0.031	0.002
299.929	18.004	173.629	0.043	0.025	-0.033	0.001
299.930	17.004	164.981	0.042	0.025	-0.035	0.001
299.931	16.001	156.149	0.041	0.026	-0.037	< 0.001
299.930	14.998	147.162	0.040	0.027	-0.038	0.001
299.930	14.000	138.079	0.039	0.028	-0.039	< 0.001
299.929	12.999	128.823	0.038	0.029	-0.039	< 0.001
299.930	11.999	119.448	0.037	0.031	-0.040	-0.002
299.930	10.998	109.939	0.036	0.033	-0.035	0.003
299.930	9.998	100.316	0.035	0.035	-0.036	-0.001
299.930	8.998	90.589	0.034	0.037	-0.036	-0.004
299.930	7.998	80.755	0.032	0.040	-0.035	-0.006
299.929	6.998	70.842	0.031	0.044	-0.030	-0.006
299.930	5.997	60.838	0.030	0.050	-0.028	-0.010
299.929	4.997	50.784	0.029	0.057	-0.025	-0.012
299.930	3.997	40.678	0.028	0.068	-0.021	-0.014
299.932	2.984	30.399	0.027	0.088	-0.019	-0.016
299.930	1.999	20.375	0.026	0.125	-0.018	-0.019
299.929	0.998	10.171	0.024	0.240	-0.027	-0.029
324.945	19.731	171.908	0.043	0.025	-0.033	0.019
324.947	19.001	166.298	0.042	0.025	-0.035	0.017

324.948	18.003	158.509	0.041	0.026	-0.037	0.016
324.948	17.000	150.561	0.040	0.027	-0.038	0.014
324.948	15.998	142.496	0.039	0.028	-0.039	0.014
324.948	14.996	134.301	0.039	0.029	-0.039	0.013
324.950	13.997	126.005	0.038	0.030	-0.040	0.012
324.949	13.000	117.611	0.037	0.031	-0.040	0.010
324.950	11.999	109.068	0.036	0.033	-0.039	0.008
324.950	10.997	100.420	0.035	0.035	-0.033	0.011
324.949	9.998	91.672	0.034	0.037	-0.035	0.006
324.950	8.998	82.834	0.033	0.039	-0.033	0.003
324.949	7.998	73.894	0.032	0.043	-0.031	-0.001
324.949	6.997	64.863	0.031	0.047	-0.027	-0.003
324.949	5.997	55.762	0.030	0.053	-0.026	-0.008
324.949	4.998	46.597	0.029	0.061	-0.023	-0.011
324.948	3.999	37.374	0.027	0.073	-0.020	-0.013
324.950	2.983	27.940	0.026	0.094	-0.017	-0.015
324.951	1.999	18.750	0.025	0.135	-0.016	-0.018
324.950	0.998	9.371	0.024	0.259	-0.031	-0.033
349.937	19.744	158.626	0.041	0.026	-0.038	0.031
349.938	18.992	153.266	0.041	0.027	-0.038	0.029
349.938	17.997	146.084	0.040	0.027	-0.039	0.028
349.937	17.001	138.788	0.039	0.028	-0.039	0.026
349.938	15.993	131.294	0.038	0.029	-0.040	0.023
349.938	14.999	123.797	0.037	0.030	-0.040	0.020
349.936	14.000	116.157	0.036	0.031	-0.039	0.019
349.937	12.994	108.364	0.036	0.033	-0.038	0.017
349.936	11.997	100.534	0.035	0.034	-0.037	0.013
349.937	10.996	92.584	0.034	0.036	-0.031	0.015

349.937	9.996	84.544	0.033	0.039	-0.031	0.009
349.937	8.998	76.424	0.032	0.042	-0.029	0.005
349.937	7.998	68.207	0.031	0.045	-0.028	< 0.001
349.938	6.997	59.903	0.030	0.050	-0.025	-0.003
349.938	5.998	51.533	0.029	0.056	-0.024	-0.009
349.937	4.998	43.083	0.028	0.065	-0.023	-0.014
349.937	3.998	34.568	0.027	0.079	-0.025	-0.021
349.938	2.979	25.835	0.026	0.101	-0.027	-0.027
349.938	1.999	17.375	0.025	0.145	-0.027	-0.030
349.939	0.998	8.697	0.024	0.278	-0.037	-0.040

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<sup>a</sup> Expanded uncertainties ( $k = 2$ ) in temperature and pressure are  $U(T) = 0.004$  K and  $U(p) = 0.005$  MPa, respectively.



**Table 7.** Experimental ( $p$ ,  $\rho$ ,  $T$ ) measurements for the (0.50 N<sub>2</sub> + 0.50 H<sub>2</sub>) mixture (actual composition given in Table 2), absolute and relative expanded uncertainty in density ( $k = 2$ )  $U(\rho_{\text{exp}})$  and relative deviations from the GERG-2008 and AGA8-DC92 EoS; where  $T$  is the temperature (ITS-90),  $p$  the pressure,  $\rho_{\text{exp}}$  the experimental density, and  $\rho_{\text{GERG}}$  and  $\rho_{\text{AGA}}$  the densities calculated from the GERG-2008 and the AGA8-DC92 EoS.

$T/\text{K}^a$	$p/\text{MPa}^a$	$\rho_{\text{exp}}/\text{kg}\cdot\text{m}^{-3}$	$U(\rho_{\text{exp}}) / \text{kg}\cdot\text{m}^{-3}$	$10^2 U(\rho_{\text{exp}}) / \rho_{\text{exp}}$	$10^2 (\rho_{\text{exp}} - \rho_{\text{GERG}}) / \rho_{\text{GERG}}$	$10^2 (\rho_{\text{exp}} - \rho_{\text{AGA}}) / \rho_{\text{AGA}}$
240.083	19.066	131.423	0.038	0.029	0.126	0.006
240.084	18.010	125.090	0.037	0.030	0.122	0.013
240.084	17.004	118.935	0.037	0.031	0.117	0.019
240.085	16.007	112.717	0.036	0.032	0.112	0.025
240.085	15.003	106.346	0.035	0.033	0.106	0.031
240.086	14.003	99.881	0.035	0.035	0.100	0.036
240.086	13.002	93.298	0.034	0.036	0.093	0.039
240.086	12.008	86.648	0.033	0.038	0.082	0.038
240.080	11.003	79.831	0.032	0.040	0.075	0.041
240.077	10.002	72.937	0.032	0.043	0.063	0.037
240.076	8.999	65.940	0.031	0.047	0.053	0.034
240.076	7.999	58.867	0.030	0.051	0.039	0.026
240.075	6.999	51.715	0.029	0.056	0.027	0.018
240.075	5.999	44.484	0.028	0.064	0.011	0.005
240.075	4.998	37.187	0.027	0.074	-0.005	-0.008
240.075	3.999	29.835	0.027	0.089	-0.017	-0.020
240.076	2.989	22.354	0.026	0.115	-0.032	-0.034
240.075	2.000	14.997	0.025	0.166	-0.009	-0.010
240.075	0.999	7.515	0.024	0.320	0.109	0.108

250.044	19.934	130.700	0.038	0.029	0.078	-0.019
250.042	19.005	125.441	0.038	0.030	0.075	-0.015
250.041	18.007	119.684	0.037	0.031	0.071	-0.012
250.043	17.004	113.790	0.036	0.032	0.068	-0.006
250.040	16.003	107.801	0.035	0.033	0.063	-0.002
250.041	15.005	101.724	0.035	0.034	0.059	0.003
250.039	14.001	95.510	0.034	0.036	0.054	0.007
250.039	13.003	89.231	0.033	0.037	0.050	0.011
250.039	12.001	82.831	0.033	0.039	0.044	0.013
250.038	11.002	76.353	0.032	0.042	0.044	0.021
250.038	9.999	69.753	0.031	0.045	0.036	0.019
250.038	8.999	63.085	0.030	0.048	0.030	0.017
250.038	7.999	56.336	0.030	0.053	0.023	0.015
250.036	6.999	49.504	0.029	0.058	0.019	0.014
250.036	5.999	42.600	0.028	0.066	0.011	0.008
250.035	4.998	35.626	0.027	0.077	0.009	0.007
250.035	3.998	28.596	0.026	0.093	0.009	0.008
250.033	2.988	21.436	0.026	0.120	0.019	0.018
250.032	1.999	14.386	0.025	0.173	0.042	0.041
250.032	0.999	7.213	0.024	0.333	0.140	0.139
274.998	19.574	116.461	0.036	0.031	0.020	-0.019
274.998	19.000	113.476	0.036	0.032	0.018	-0.019
274.996	17.996	108.187	0.036	0.033	0.015	-0.018
274.996	17.000	102.851	0.035	0.034	0.013	-0.016
274.996	15.997	97.397	0.034	0.035	0.012	-0.013
274.994	14.998	91.878	0.034	0.037	0.010	-0.011
274.994	13.998	86.267	0.033	0.038	0.008	-0.009
274.994	13.000	80.586	0.032	0.040	0.006	-0.007

274.994	11.996	74.789	0.032	0.042	0.003	-0.007
274.993	10.998	68.948	0.031	0.045	0.005	-0.002
274.994	9.998	63.009	0.030	0.048	0.001	-0.004
274.992	8.998	57.003	0.030	0.052	< 0.001	-0.004
274.992	7.997	50.911	0.029	0.057	-0.002	-0.005
274.992	6.997	44.758	0.028	0.063	0.001	-0.002
274.992	5.997	38.535	0.028	0.072	< 0.001	-0.002
274.991	4.997	32.248	0.027	0.083	0.003	0.001
274.991	3.997	25.901	0.026	0.101	0.009	0.006
274.990	2.998	19.501	0.025	0.130	0.020	0.017
274.992	1.998	13.041	0.025	0.189	0.027	0.024
274.992	0.998	6.537	0.024	0.366	0.057	0.055
299.941	19.854	107.886	0.035	0.033	-0.024	-0.024
299.942	18.998	103.792	0.035	0.034	-0.026	-0.026
299.941	17.997	98.934	0.034	0.035	-0.027	-0.027
299.942	16.992	93.982	0.034	0.036	-0.028	-0.028
299.941	15.992	88.984	0.033	0.037	-0.029	-0.029
299.942	14.998	83.948	0.033	0.039	-0.030	-0.029
299.941	13.996	78.800	0.032	0.041	-0.029	-0.028
299.942	12.994	73.577	0.032	0.043	-0.030	-0.029
299.941	11.998	68.317	0.031	0.045	-0.030	-0.029
299.942	10.997	62.968	0.030	0.048	-0.025	-0.024
299.942	9.998	57.555	0.030	0.052	-0.026	-0.026
299.942	8.996	52.057	0.029	0.056	-0.025	-0.026
299.941	7.997	46.510	0.029	0.061	-0.024	-0.026
299.940	6.997	40.897	0.028	0.068	-0.021	-0.024
299.940	5.998	35.223	0.027	0.077	-0.019	-0.023
299.941	4.998	29.485	0.027	0.090	-0.018	-0.023

299.941	3.998	23.691	0.026	0.109	-0.015	-0.020
299.940	2.999	17.846	0.025	0.141	-0.011	-0.017
299.940	1.998	11.937	0.025	0.206	-0.020	-0.025
299.941	0.998	5.984	0.024	0.399	-0.025	-0.029
324.948	19.974	100.115	0.035	0.035	-0.025	0.006
324.950	18.996	95.768	0.034	0.036	-0.027	0.002
324.950	17.998	91.271	0.034	0.037	-0.028	-0.002
324.949	16.998	86.706	0.033	0.038	-0.028	-0.006
324.947	15.985	82.016	0.033	0.040	-0.028	-0.008
324.951	14.993	77.366	0.032	0.041	-0.027	-0.009
324.950	13.992	72.609	0.031	0.043	-0.025	-0.011
324.951	12.995	67.810	0.031	0.046	-0.024	-0.013
324.951	11.997	62.949	0.030	0.048	-0.022	-0.014
324.951	10.996	58.014	0.030	0.051	-0.016	-0.010
324.951	9.998	53.025	0.029	0.055	-0.016	-0.012
324.952	8.997	47.967	0.029	0.060	-0.014	-0.013
324.951	7.996	42.850	0.028	0.066	-0.012	-0.014
324.953	7.002	37.712	0.027	0.073	-0.006	-0.010
324.951	6.001	32.482	0.027	0.083	< 0.001	-0.006
324.951	5.000	27.194	0.026	0.097	0.010	0.003
324.952	3.999	21.856	0.026	0.118	0.021	0.013
324.951	2.996	16.446	0.025	0.152	0.032	0.024
324.952	1.998	11.018	0.024	0.222	0.034	0.027
324.953	0.998	5.527	0.024	0.431	0.026	0.023
349.939	19.885	92.681	0.034	0.036	-0.034	0.026
349.938	18.991	88.967	0.033	0.037	-0.032	0.022
349.937	17.991	84.762	0.033	0.039	-0.031	0.018

349.937	16.996	80.519	0.032	0.040	-0.031	0.012
349.937	15.994	76.196	0.032	0.042	-0.030	0.008
349.937	14.992	71.820	0.031	0.044	-0.029	0.003
349.937	13.990	67.389	0.031	0.046	-0.028	-0.001
349.937	12.996	62.939	0.030	0.048	-0.026	-0.004
349.936	11.994	58.400	0.030	0.051	-0.024	-0.007
349.936	10.995	53.825	0.029	0.055	-0.017	-0.005
349.936	9.995	49.185	0.029	0.059	-0.016	-0.008
349.936	8.997	44.504	0.028	0.064	-0.014	-0.011
349.935	7.995	39.749	0.028	0.070	-0.013	-0.013
349.936	6.996	34.959	0.027	0.078	-0.011	-0.014
349.936	5.998	30.119	0.027	0.088	-0.011	-0.017
349.936	5.000	25.225	0.026	0.103	-0.009	-0.017
349.936	3.995	20.259	0.026	0.126	0.013	0.005
349.939	2.984	15.198	0.025	0.164	0.006	-0.002
349.940	1.998	10.223	0.024	0.238	-0.004	-0.011
349.940	0.999	5.133	0.024	0.463	0.009	0.005

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<sup>a</sup> Expanded uncertainties ( $k = 2$ ) in temperature and pressure are  $U(T) = 0.004$  K and  $U(p) = 0.005$  MPa, respectively.

**Table 8.** Statistical parameters of the data set with respect to the GERG-2008 and AGA8-DC92 EoS for the (N<sub>2</sub> + H<sub>2</sub>) mixtures and literature comparison.

$(p, \rho, T)$ data	$x$ (H <sub>2</sub> )	Covered ranges		$N$	Experimental vs. GERG-2008				Experimental vs. AGA8-DC92			
		$T/K$	$p/\text{MPa}$		$AAD$	$Bias$	$RMS$	$MaxD/\%$	$AAD$	$Bias$	$RMS$	$MaxD/\%$
This work (2017)	0.05	240 - 350	1 - 20	140	0.013	-0.012	0.014	0.032	0.014	-0.0003	0.018	0.043
This work (2017)	0.10	240 - 350	1 - 20	140	0.027	-0.026	0.028	0.042	0.017	-0.011	0.020	0.047
This work (2017)	0.50	240 - 350	1 - 20	119	0.032	0.011	0.043	0.140	0.018	0.0009	0.025	0.139
Jaeschke <i>et al.</i> (1997) [3]	0.50	270 - 350	0.5 - 28.9	184	0.028	0.002	0.036	0.104	0.021	0.007	0.025	0.062
Verschoyle (1926) [7]	0.50	273 - 293	3.7 - 20.8	21	0.104	0.081	0.124	0.238	0.093	0.068	0.109	0.195
Wiebe and Gaddy (1938) [8]	0.52	273 - 573	2.5 - 101	46	0.210	0.205	0.260	0.658	0.552	0.538	0.935	3.254

**Table 9.** Least-squares fit results for the three (N<sub>2</sub> + H<sub>2</sub>) mixtures. Virial coefficients (*B*, *C*, and *D*) and the second interaction virial coefficient (*B*<sub>12</sub>) with the corresponding expanded uncertainties (*k* = 2).

<i>T</i> /K <sup>a</sup>	<i>B</i> / cm <sup>3</sup> ·mol <sup>-1</sup>	<i>U</i> ( <i>B</i> )/ cm <sup>3</sup> ·mol <sup>-1</sup>	<i>C</i> / cm <sup>6</sup> ·mol <sup>-2</sup>	<i>U</i> ( <i>C</i> )/ cm <sup>6</sup> ·mol <sup>-2</sup>	<i>D</i> / cm <sup>9</sup> ·mol <sup>-3</sup>	<i>U</i> ( <i>D</i> )/ cm <sup>9</sup> ·mol <sup>-3</sup>	<i>B</i> <sub>12</sub> / cm <sup>3</sup> ·mol <sup>-1</sup>	<i>U</i> ( <i>B</i> <sub>12</sub> )/ cm <sup>3</sup> ·mol <sup>-1</sup>
(0.95 N <sub>2</sub> + 0.05 H <sub>2</sub> )								
240.048	-16.63	0.86	1400	400	26 000	45 000	10.0	1.5
250.019	-13.81	0.99	1400	490	26 000	57 000	10.5	1.4
260.019	-11.2	1.1	1300	550	27 000	69 000	11.1	1.3
274.992	-7.8	1.2	1400	620	20 000	82 000	11.1	1.4
299.939	-2.8	1.4	1300	820	24 000	130 000	13.2	1.6
324.945	1.3	1.6	1200	1100	32 000	170 000	15.5	1.7
349.935	4.6	1.8	1200	1300	21 000	220 000	15.8	1.9
(0.90 N <sub>2</sub> + 0.10 H <sub>2</sub> )								
240.069	-13.91	0.55	1300	210	32 000	19 000	9.8	1.3
250.043	-11.25	0.58	1200	230	32 000	22 000	10.7	1.1
260.038	-8.79	0.62	1200	260	32 000	26 000	11.7	1.0
274.991	-5.64	0.68	1200	310	31 000	33 000	12.2	1.0
299.930	-1.05	0.82	1100	410	32 000	50 000	13.9	1.1
324.949	2.76	0.94	1100	510	36 000	67 000	15.4	1.2
349.937	6.0	1.1	1000	620	44 000	89 000	16.9	1.3
(0.50 N <sub>2</sub> + 0.50 H <sub>2</sub> )								
240.080	2.21	0.31	890	62	-	-	7.7	1.2
250.038	3.48	0.33	900	70	-	-	8.53	0.96
274.994	6.38	0.39	870	90	-	-	10.68	0.81
299.941	8.83	0.45	830	120	-	-	12.66	0.84
324.951	10.58	0.51	850	140	-	-	13.76	0.87
349.937	12.24	0.55	820	170	-	-	15.09	0.90

<sup>a</sup> *T* is the average temperature of each isotherm.