# Experimental and modelling study of the densities of the hydrogen sulphide + methane mixtures at 253, 273 and 293 K and pressures up to 30 MPa . 

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#### Abstract

Densities of three binary mixtures of hydrogen sulphide and methane $\left(\mathrm{xH}_{2} \mathrm{~S}+(1-\mathrm{x}) \mathrm{CH}_{4}\right)$, with mole fractions of $0.1315,0.1803$ and 0.2860 of acid gas, were determined experimentally at three temperatures $(253,273$ and 293$) \mathrm{K}$ and at pressures up to 30 MPa . Densities were measured continuously using a high temperature and high pressure Vibrating Tube densitometer (VTD), Anton Paar DMA 512. The SAFT-VR Mie, PR and GERG2008 equations of state (EoS) are used to describe the experimental data with different levels of success.


Keywords: $\mathrm{CH}_{4}, \mathrm{H}_{2} \mathrm{~S}$, Thermophysical properties, Acid gas injection

## 1. Introduction

Many sour natural gases reservoirs and sour gas condensates fields have been discovered in the last thirty years. This kind of natural hydrocarbon gas reservoirs represent about half of the known total worldwide resources ( 270 trillion $\mathrm{m}^{3}$ ) [1]. Acid gases are those raw natural
gases which contain significant amounts of carbon dioxide $\left(\mathrm{CO}_{2}\right)$ and hydrogen sulphide $\left(\mathrm{H}_{2} \mathrm{~S}\right)$, with compositions around of $0-80 \%$ moles $\mathrm{CO}_{2}$ and $0-30 \% \mathrm{H}_{2} \mathrm{~S}$ [2]. For this gas with high acid gas content carbon capture storage (CCS) technologies, such as oxy-fuel combustion, have the potential to become economically competitive with other zero-carbon and renewal sources of energy [3]. Thus, CCS may allow continuing to burn fossil fuels in power stations preventing $\mathrm{CO}_{2}$ emissions to the atmosphere. CCS refers to a large number of technologies and processes, such as combustion methods (pre-combustion, post-combustion and oxy-fuel), transport pipelines or injection systems in geological basins [4]. Pressure-Volume-Temperature (PVT) data and thermophysical property models are of principal importance to the design of process involved in the capture, transport and storage of $\mathrm{CO}_{2}$.

Since hydrogen sulphide is a toxic and corrosive compound, it is necessary to process sour gases in order to remove acid gases before their uses [5]. Different regulations require that $\mathrm{H}_{2} \mathrm{~S}$ contain of sweet gases must be less than 4 ppm . The sweetening and desulphurisation of acid gases, such as the SPREX process [6] or physical and chemical absorption with solvents, are costly processes that require accurate thermodynamic models to design natural gas processing plants. The methane $-\mathrm{H}_{2} \mathrm{~S}$ binary system is of interest to the oil and gas industry [7] because $\mathrm{CH}_{4}$ and $\mathrm{H}_{2} \mathrm{~S}$ are important components of reservoir fluids.

There are limited references available in the literature concerning single phase density ( $\rho-P T x$ ) of methane and hydrogen sulphide systems (Table 1). In 1951, Reamer et al [8] did a thorough study of this binary mixture, they were the first to measure vapour-liquid equilibria (VLE) and densities of several $\mathrm{H}_{2} \mathrm{~S}+\mathrm{CH}_{4}$ systems [9]. Reamer's density measurements cover a wide range of compositions (0.1-0.9 mol fraction $\mathrm{H}_{2} \mathrm{~S}$ ), temperatures ( $277-411 \mathrm{~K}$ ) and pressures (up to 69 MPa ) with 1140 data. Bailey et al. [10] studied the system $49 \% \mathrm{~mol}$ of $\mathrm{CH}_{4}$ and $51 \% \mathrm{~mol}$ of $\mathrm{H}_{2} \mathrm{~S}$ at higher temperature ( 501 K ) The main goal of the new measurements presented in this work is to complete the literature data at low temperature.

In this paper, new density data of $\mathrm{H}_{2} \mathrm{~S}+\mathrm{CH}_{4}$ mixtures are presented for the following compositions (mole fractions): $0.1315 \mathrm{H}_{2} \mathrm{~S}+0.8685 \mathrm{CH}_{4}, 0.1803 \mathrm{H}_{2} \mathrm{~S}+0.8197 \mathrm{CH}_{4}$ and $0.286 \mathrm{H}_{2} \mathrm{~S}+0.714 \mathrm{CH}_{4}$. The density of each system has been measured at three isotherms, 253,273 and 293 K , and at pressures up to 30 MPa . The data of the three measured systems are compared to density calculations using three types of equation of state: the Peng-Robinson EoS [11], the SAFT-VR Mie EoS [12] and the multiparameter GERG-2008 EoS [13] for
natural gas ). Finally, all the literature data were modelled using the three EoS proposed, in order to evaluate these models in a wider range of temperature, pressure and compositions.

## 2. Experimental

### 2.1 Sample preparation

The specification and sources of the chemicals used in this work are summarized in Table 2. Three mixtures of $\mathrm{CH}_{4}$ and $\mathrm{H}_{2} \mathrm{~S}$ were prepared volumetrically at laboratory temperature in $300 \mathrm{~cm}^{3}$ cylinder with its own pressure transducer to track the pressure of the sample. The final pressure target of the mixture in the vessel was approximately 40MPa in order to avoid any phase change and possible variations of composition during the injection of mixture into the densitometer set-up. To prepare the mixture, a volume of $\mathrm{H}_{2} \mathrm{~S}$ is first injected into the cylinder; thereby the injected volume will contain the number of moles calculated by an EoS to reach the expected final composition. Second, the methane is pumped in the cylinder until it the final pressure is reached. Finally, the composition of each mixture was validated by gas chromatography analysis (Varian model CP-3800). The resulting compositions of the prepared mixtures have been $0.1315,0.1803$ and 0.2860 mole fractions of $\mathrm{H}_{2} \mathrm{~S}$.

### 2.2 Equipment description

A full description of the experimental set-up and procedures are available in Bouchot and Richon [14], Coquelet et al. [15] and Nazeri et al. [16]. The density of the studied mixture have been measured in a Hastelloy Vibrating Tube Densitometer (VTD), Anton Paar model DMA 512, using the one fluid reference calibration method [17].

### 2.3 Experimental procedure

Three isotherms of each binary system were measured at 253 , 273 and 293 K . The mixture was gradually charged from the cylinder in the experimental set-up while the vibrating period, the temperature and the pressure were recorded continuously during the slow-increasing pressure up to 30 MPa and the slow decompression (the pressure in the vessel containing the mixture was maintained at pressure higher than 30 MPa during all the measurements). During the depressurisation, the acid outlet gas was neutralised by bubbling it through a column with a basic solution of sodium hydroxide.

### 2.4 Uncertainty of measurements

Uncertainties of the properties measured have been evaluated as standard uncertainties of type A and type B [18]. Type A of uncertainties is based on the statistical treatment of experimental data. The uncertainty of the composition measurements was evaluated as type A and it was calculated as [19]:

$$
\begin{equation*}
u\left(x_{H_{2} S}\right)=\sqrt{\frac{\sigma^{2}(x)}{N}} \tag{1}
\end{equation*}
$$

where $\sigma^{2}$ is the variance of the values measured and $N$ is the number of measurements. The expanded uncertainties ( $k=2$, in order to increase the confidence level up to $95 \%$ [19]) of the composition of the three systems are 0.0006 for $0.1315 \mathrm{H}_{2} \mathrm{~S}$ mole fraction system, 0.0008 for $0.1803 \mathrm{H}_{2} \mathrm{~S}$ mole fraction system and 0.0011 for $0.2860 \mathrm{H}_{2} \mathrm{~S}$ mole fraction system.

The uncertainties type B are evaluated by other means (not purely statistical), such as literature, manufacturer information, previous experience or, as in our case, calibrations. Then, uncertainty on temperature and pressure measurements was estimated as type B.

The temperature of the system in the set-up is regulated by two liquid thermoregulated baths, which temperatures are measured with two $\operatorname{Pt} 100$ ( $100 \Omega$ platinum resistance) probes. The probes calibration is done against a $25 \Omega$ reference thermometer (Tinsley Precision Instrument). From the calibrations, the expanded uncertainty ( $k=2$ ) in temperatures measurements was estimated to be 0.03 K .

The pressure was measured by two pressure transducers at two complementary ranges, one between $0-5 \mathrm{MPa}$ and other from 5 up to 30 MPa . The calibration of the transducers was done using a dead weight balance (Desgranges \& Huot, model 5202S). The expanded uncertainties $(\mathrm{k}=2)$ of the pressure measurements from the calibration are estimated to be less than 0.003 MPa for low pressure (up to 5 MPa ) and 0.005 MPa for higher pressures than 5 MPa (up to 30 $\mathrm{MPa})$.

Density uncertainty was evaluated as combined standard uncertainty, because density is expressed through the combination of two sources of expanded standard uncertainty: calibration and densitometer information. There is a mathematical relation ( $\rho=\mathrm{a} \tau+\mathrm{b}$ ) between these standard uncertainties, therefore applying the law of propagation of uncertainty [20], density uncertainty was obtained by:

$$
\begin{equation*}
u(\rho)=\sqrt{\left(\frac{\partial \rho}{\partial \tau}\right)_{a, b}^{2} u(\tau)^{2}+\left(\frac{\partial \rho}{\partial a}\right)_{\tau, b}^{2} u(a)^{2}+\left(\frac{\partial \rho}{\partial b}\right)_{\tau, a}^{2} u(b)^{2}} \tag{2}
\end{equation*}
$$

where $\tau$ is the vibrating period and ' $a$ ' and ' $b$ ' are the calibration parameters. From manufacturer's specifications, the standard uncertainty on the period of oscillation is $10^{-8} \mathrm{~s}$. Uncertainty on the parameters of the calibration has been estimated to be $0.5 \%$ over the investigated range of pressures; however, it is worth highlighting that, due to the used technique, average uncertainty at low pressure (below 0.5 MPa ) was $8.6 \%$. The combined standard uncertainties of the measurements are reported accompanying the density data in Tables 6-14.

The calculation of the compressibility factor $(Z)$ is function of the 3 measured properties $(Z=P / \rho R T)$ : temperature, pressure and density. Therefore, $\mathrm{u}(Z)$ gives an idea of the overall uncertainty of our experimental data because includes the uncertainty of every measurement. Then, following the same approach, the uncertainty of the Z was calculated using the law of propagation of uncertainty as:

$$
\begin{equation*}
u(Z)=\sqrt{\left(\frac{\partial Z}{\partial P}\right)_{\rho, T}^{2} u(P)^{2}+\left(\frac{\partial Z}{\partial \rho}\right)_{P, T}^{2} u(\rho)^{2}+\left(\frac{\partial Z}{\partial T}\right)_{P, \rho}^{2} u(T)^{2}} \tag{3}
\end{equation*}
$$

Further information about calculation of the combined uncertainties of our measurements is provided in Appendix A.

## 3. Modelling

### 3.1 Peng and Robinson

Despite hydrogen sulphide is a polar molecule, we have considered $\mathrm{H}_{2} \mathrm{~S}$ as a non-associating compound due to the good results that classical cubic models have shown to model the phase equilibrium of the $\mathrm{CH}_{4}+\mathrm{H}_{2} \mathrm{~S}$ system [7]. The critical properties ( $\mathrm{T}_{\mathrm{c}}$ and $\mathrm{P}_{\mathrm{c}}$ ) and acentric factors $(\omega)$ of methane and hydrogen sulphide are given in Table 3. The pure component parameters have been used to model the system with the Peng and Robinson (PR) EoS [11], which is one of the most used cubic EoS due its simplicity and reasonable accuracy for non associating compounds [21]. In this work the van der Waals (classical) mixing rules are used, i.e.:
$b=\sum_{i} x_{i} b_{i}$
$a=\sum_{i} \sum_{j} x_{i} x_{j} \sqrt{a_{i} a_{j}}\left(1-k_{i j}\right)$
where $a$ and $b$ are the parameters in the PR-EoS and $k_{i j}$ the binary interaction parameter (BIP).

The BIPs are coefficients introduced to describe the experimental phase behaviour. Thus, the binary parameters avoid wrong predictions of the two phase region during density calculations. A temperature independent $k_{i j}$ has been regressed by minimizing the objective function given in Equation 6, which is based on calculated bubble point pressures and experimental VLE data from the literature [7] [8] [22]. The regressed BIP is reported in Table 5.
$\min F=\frac{100}{N} \sum_{1}^{N}\left(\frac{P_{\text {bubbll }}^{\mathrm{exp}}-P_{\text {bubble }}^{\text {cal }}}{P_{\text {bubble }}^{\mathrm{exp}}}\right)$

The PR-EoS tends to underestimate experimental densities [23]. Volume translations can be used in order to improve the density calculations of liquid and dense fluid phases. The Peneloux volume correction [24] has been implemented here (Equation 7) and the results with and without Peneloux shift parameters are discussed in Section 4.
$V=V^{P R}+\sum_{i}^{\text {NComp }} x_{i} V_{i}^{c}$
where $V^{P R}$ is the molar volume calculated by the PR EoS, $x_{i}$ the mole fraction of each pure component $i$, and $V_{i}^{c}$ the volume correction parameter. The values of the parameters are reported in Table 3.

### 3.2 SAFT-VR Mie

The SAFT-VR Mie EoS proposed by Lafitte et al [12] is one of the latest updates of the SAFT family of EoS. In this equation of state, the attractive and repulsive interactions between the segments that build the molecules are described by the Mie potential that is defined as:
$u^{M i e}(r)=\frac{\lambda_{r}}{\lambda_{r}-\lambda_{a}}\left(\frac{\lambda_{r}}{\lambda_{a}}\right)^{\frac{\lambda_{r}}{\lambda_{r}-\lambda_{a}}} \varepsilon\left(\left(\frac{\sigma}{r}\right)^{\lambda_{r}}-\left(\frac{\sigma}{r}\right)^{\lambda_{a}}\right)$
where $r$ is the intersegment distance, $\sigma$ the temperature-independent segment diameter, and $\varepsilon$ the potential depth; $\lambda_{r}$ and $\lambda_{a}$ are the repulsive and attractive ranges, respectively.

The SAFT-VR Mie EoS can be expressed in terms of the reduced Helmholtz energy as the sum of several contributions:

$$
\begin{equation*}
a=\frac{A}{n R T}=a^{\text {IDEAL }}+a^{\text {MONOMER }}+a^{\text {CHAIN }}+a^{\text {ASSOCIATION }} \tag{9}
\end{equation*}
$$

The molecular parameters for $\mathrm{CH}_{4}$ and $\mathrm{H}_{2} \mathrm{~S}$ are presented in Table 4. Hydrogen sulphide parameters were regressed using experimental saturation pressures $\left(P_{\text {sat }}\right)$ and liquid densities $\left(\rho_{\text {sat }}\right)$, thereby the fitting consisted in the minimization of the following objective function [12]:

A temperature independent binary interaction parameter was regressed following the same approach and VLE data as for the PR-EoS. The resulting $k_{i j}$ is also reported in Table 5.

### 3.3 GERG

GERG-2008 is a wide-range EoS for natural gases and other similar mixtures of gases [13]. It is an empirical equation of state based on pure component EoS and correlations for the binary systems [25]. Like the SAFT EoS, GERG is expressed in terms of the Helmholtz free energy, as the sum of an ideal gas contribution and a residual part.

In this work, the PR, SAFT-VR Mie and GERG-2008 [13] EoSs were employed to predict the densities of mixtures and were implemented into the thermodynamic package HWPVT [16] [26] [27] [28] [29].

## 4. Result and discussion

Densities of three binary systems of $\mathrm{CH}_{4}$ and $\mathrm{H}_{2} \mathrm{~S}$ with $0.1315,0.1803$ and 0.2860 mol fractions of hydrogen sulphide have been measured continuously using a high temperature and high pressure VTD Anton Paar. First, the densitometer calibration was done using pure ethane with a first-order polynomial calibration at 253,273 and 293 K and pressures up to 31 MPa. Previous to the measurements, the phase envelops of the three binary systems have been studied in order to forecast at which pressure the two phase behaviour is found. The phase
diagrams of the three measured systems calculated by the PR-EoS and other three systems comparing calculations against literature data [22] are shown in the Figure 1. According to Scott and van Konynenburg, the phase diagram of this system is classified as type III [7]. This means that the curves presented in the phase envelops of Figure 1 are dew lines, therefore no density was measured in the liquid region. The studied binary mixtures are systems with no critical point [30] [31], thus all our $\rho-P T x$ measurements have been done in the gas phase. The P-T envelops show that there is no phase change for any of the studied isotherms for the 0.1315 mol fraction of $\mathrm{H}_{2} \mathrm{~S}$ system. However, it is possible to enter the two phase region at 253 K for the 0.1803 mol fraction of $\mathrm{H}_{2} \mathrm{~S}$ system and at 253 and 273 K in the 0.2860 mol fraction of $\mathrm{H}_{2} \mathrm{~S}$ system.

The experimental density data for the measured systems are presented in Table 6 to Table 14 . The compressibility factors derived from the measured temperature, pressure and density data, as well as their uncertainty, are also reported in the Tables 6-14.

The PR, SAFT-VR Mie and GERG-2008 equations of state have been used to model the density of the methane and hydrogen sulphide binary systems. The modelling results are presented by comparing the average deviations (\%AAD) between the models and the experimental data. The AAD is the absolute average deviation and is defined as:

$$
\begin{equation*}
A D D(\%)=\frac{1}{N} \sum_{i=1}^{N}\left(\left|\rho_{\text {exp }}-\rho_{E o S}\right| / \rho_{\text {exp }}\right) \cdot 100 \tag{11}
\end{equation*}
$$

The resulting SAFT-VR Mie parameters for pure hydrogen sulphide wi are reported in Table 4 , as well as the $\operatorname{AAD}(\%)$ of the equation of state from the experimental vapour pressure $P_{\text {sat }}$, saturated-liquid densities $\rho_{\text {sat }}$ (Figure 2) and enthalpy of vaporization $\Delta H_{v}$ [32]. The pure $\mathrm{H}_{2} \mathrm{~S}$ densities in the gas, liquid and supercritical regions have been evaluated at 13 temperatures between 200 to 500 K and pressures up to 100 MPa , reporting an \%AAD of $2.3 \%$. However, considering the $\mathrm{H}_{2} \mathrm{~S}$ single phase densities at the pressure and temperature ranges of our measurements, the absolute average deviation is $0.9 \%$ (Figure 3).

BIPs for the $\mathrm{CH}_{4}-\mathrm{H}_{2} \mathrm{~S}$ system have been regressed for both the SAFT-VR Mie EoS ( $k_{i j}=0.0314$ ) and the PR EoS $\left(k_{i j}=0.0807\right)$ models using VLE data from the literature [7] [8] [22] (Figure 4). Using these BIPs, the PR EoS is slightly more accurate to correlate the phase behaviour of the $\mathrm{CH}_{4}-\mathrm{H}_{2} \mathrm{~S}$ system than the SAFT-VR Mie EoS, with bubble point pressure AADs of $4.4 \%$ and $4.8 \%$, respectively.

The deviations of the PR, SAFT-VR Mie and GERG-2008 models for the three measured systems at each temperature are reported in Table 15. Generally, the SAFT-VR Mie EoS shows lower overall deviations $(\mathrm{AAD}=2.5 \%)$ than the other two models, PR EoS $(3.0 \%)$ and GERG-2008 (3.4\%). The experimental densities measured in this work and the predicted densities for the system $0.2860 \mathrm{H}_{2} \mathrm{~S}$ mol fraction systems by using the GERG-2008 and SAFT-VR Mie are illustrated in Figures 5. As the GERG EoS is an empirical model developed for natural gases whose parameters for the $\mathrm{CH}_{4}+\mathrm{H}_{2} \mathrm{~S}$ binary system have been fitted to experimental data over wide ranges of temperature ( 189 to 501 K ), pressure ( 0.048 to 68.9 MPa ) and methane mol fraction ( 0.1 to 0.9993 ) [13], it was unexpected that the GERG2008 EoS reported the highest deviations for these systems, particularly for the 0.2860 mol fraction of $\mathrm{H}_{2} \mathrm{~S}$ system (Figure 5) with an AAD of $4.9 \%$. However, the GERG-2008 EoS presents better results than SAFT and PR EoSs for the binary mixture with smaller amount of $\mathrm{H}_{2} \mathrm{~S}$.

Density data available in the literature (1748 points) and those measured in this work have been modelled with the PR, PR + Peneloux volume correction, SAFT-VR Mie and GERG2008 equations of state, and the deviations are listed in Table 16. As can be observed, the SAFT-VR Mie EoS reports the lower deviation between experimental and calculated densities with an AAD of $4.3 \%$, while GERG-2008 shows similar level of agreement (4.6\%). The experimental densities of $50 \mathrm{~mol} \% \mathrm{H}_{2} \mathrm{~S}+50 \mathrm{~mol} \% \mathrm{CH}_{4}$ and the predicted densities using the PR + Peneloux, GERG-2008 and SAFT-VR Mie EoSs are presented in Figure 6, as an example of the modelling result of this system at high pressure (up to 70MPa).

The PR-EoS is the studied model that has reported the largest total deviation (AAD $=7.8 \%$ ), especially when PR calculations are compared against Reamer et al. [8] data at pressures over 30 MPa . The results of the PR -EoS with volume correction (VC) have not been presented in Table 6 because no better density predictions were observed after applying the Peneloux VC to model our measured densities. However, remarkable improvements in the density calculations with the PR-EoS can be noticed in the results of modelling the full Reamer's data set using the PR-Peneloux, decreasing by half the ADD. The AAD reduces from 7.8\% to $5.4 \%$ when the complete density data set is modelled using the PR with Peneloux shift model. Figure 7 shows the comparison of PR and PR-Peneloux calculations against literature data of the 0.3 mol fraction $\mathrm{H}_{2} \mathrm{~S}$ system, in order to illustrate the large deviations of PR EoS at high pressure.

The uncertainties for each measured density are reported in Tables 6-14. The uncertainties were evaluated as combined standard uncertainty with $95 \%$ level of confidence. Despite the low average uncertainty ( $0.5 \%$ ), the uncertainty of the density measurements below 0.5 MPa is $8.3 \%$ with a maximum value of $10.1 \%$. This high uncertainty was expected due to the fact that the vibrating tube densitometer is not the most appropriate technique for the determination of the densities of gases at low pressure (i.e. low mass in the VTD) [33]. The elevated uncertainty of density at low pressure is propagated to compressibility factor calculations (Figure 8).

The volumetric behaviour of the $\mathrm{CH}_{4}+\mathrm{H}_{2} \mathrm{~S}$ binary mixture at low pressure has been studied with the virial expansion truncated after the second coefficient. A virial equation provides a simple way to describe thermodynamic properties, and the virial coefficients can be determined by different experimental methods or correlations [34]. The form for our virial expansion is given by:

$$
\begin{equation*}
Z=1+\rho B\left(T, x_{1}\right) \tag{12}
\end{equation*}
$$

where $B$ is the second virial coefficient which is calculated as a function of temperature and mol fraction of methane $\left(x_{I}\right)$ as:

$$
\begin{equation*}
B\left(T, x_{1}\right)=a+\frac{b}{T}+\frac{c}{T^{3}}+d x_{1} \tag{13}
\end{equation*}
$$

where $a, b, c$ and $d$ are the parameters reported in Table 17. These parameters have been correlated using our density measurements and the low pressure $\rho-P T x$ data sets from the literature [8] [10]. The correlated virial equation of state has an AAD of $1.5 \%$ with a maximum deviation of $5.7 \%$ in a wide range of compositions, pressures up to 1.9 MPa and temperatures between 253 and 500 K (Figure 9). Several isotherms calculated with our $\mathrm{CH}_{4}+\mathrm{H}_{2} \mathrm{~S}$ virial equation of state are illustrated in Figure 10.

## 5. Conclusion

Density data measurements for the three binary mixtures of methane with $0.1315,0.1803$ and 0.286 mol fraction of hydrogen sulphide were performed at three temperatures between 253 and 293 K and pressures up to 30 MPa by using a vibrating tube Anton Paar densitometer. The average uncertainty of the measurements at the $95 \%$ confidence level is $0.5 \%$, reporting the highest average uncertainty of $8.3 \%$ at low pressures (below 0.5 MPa ).

The measured experimental data were compared with the densities predicted with the PR, SAFT-VR Mie and GERG-2008 equations of state. For these measurements the SAFT-VR Mie EoS has the lowest absolute average deviation ( $2.5 \%$ ) among the three investigated models. However deviations for all models are of the same order ( 3.4 \% for the GERG EoS and for $3.0 \%$ the PR EoS without volume correction $3.0 \%$ ). However, theGERG-2008 is slightly superior to describe the density of the 0.1315 mol fraction of $\mathrm{H}_{2} \mathrm{~S}$ system ( $\mathrm{AAD}=2.8 \%$ ) than the SAFT-VR Mie $(\mathrm{AAD}=3.0 \%)$ and $\mathrm{PR}(\mathrm{AAD}=2.9 \%)$ EoSs.

Furthermore, the literature data have been studied with the different models. When compared against the full density dataset, the lowest deviations are observed for the SAFT-VR Mie (4.3 \%). The deviations with GERG-2008, PR, PR-Peneloux EoSs were 4.6, 7.8 and 5.4 \%, respectively. Significant improvement in density predictions are observed when volume correction are used with the PR-EoS, due to the overestimated densities predicted by the PR EoS at pressures over 30MPa.

Finally, the measured densities and the $\rho-P T x$ data from the literature at low pressure were used to correlate the second virial coefficient as a function of temperature and $\mathrm{CH}_{4} \mathrm{~mol}$ fraction. The absolute average deviation of the compressibility factor calculated from our virial EoS in a wide range of compositions and at pressures up to 1.9 MPa and temperatures between 253 and 500 K is $1.5 \%$ with a maximum deviation of $5.7 \%$.

## Appendix A. Uncertainty in density measurements and compressibility factors

Density uncertainty was evaluated as combined standard uncertainty [18] [20]. Density measurement uncertainty can be expressed through the combination of two sources of expanded standard uncertainty, calibration and densitometer information, due to the mathematical relation used to do the calibration:

$$
\begin{equation*}
\rho=a \tau+b \tag{A1}
\end{equation*}
$$

Therefore, applying the law of propagation of uncertainty [20], density uncertainty was obtained by:

$$
\begin{equation*}
u(\rho)=\sqrt{\left(\frac{\partial \rho}{\partial \tau}\right)_{a, b}^{2} u(\tau)^{2}+\left(\frac{\partial \rho}{\partial a}\right)_{\tau, b}^{2} u(a)^{2}+\left(\frac{\partial \rho}{\partial b}\right)_{\tau, a}^{2} u(b)^{2}} \tag{A2}
\end{equation*}
$$

where $\tau$ is the time of oscillation and both ' $a$ ' and ' $b$ ' are the calibration parameters. Subsequently, the partial derivatives required for calculating $u(\rho)$ are derived from equation A1 as:

$$
\begin{equation*}
\left(\frac{\partial \rho}{\partial \tau}\right)_{a, b}=a \quad\left(\frac{\partial \rho}{\partial a}\right)_{\tau, b}=\tau \quad\left(\frac{\partial \rho}{\partial b}\right)_{a, \tau}=1 \tag{A3}
\end{equation*}
$$

Finally, the combined standard uncertainty of our density measurements can be expressed by

$$
\begin{equation*}
u(\rho)=\sqrt{a^{2} u(\tau)^{2}+\tau^{2} u(a)^{2}+u(b)^{2}} \tag{A4}
\end{equation*}
$$

The calculation of the compressibility factor $(Z)$ is function of the 3 properties measured: temperature, pressure and density. The compressibility factor can be formulated as

$$
\begin{equation*}
Z=\frac{P}{\rho R T} \tag{A5}
\end{equation*}
$$

Consequently, uncertainty in compressibility factor calculation, $u(Z)$, gives an idea of the overall uncertainty of our experimental data due to the consideration of the uncertainties of every measurement. Thus, following the same approach for the propagation of the uncertainty, the uncertainties of the compressibility factors were calculated using the law of propagation as:

$$
\begin{equation*}
u(Z)=\sqrt{\left(\frac{\partial Z}{\partial P}\right)_{\rho, T}^{2} u(P)^{2}+\left(\frac{\partial Z}{\partial \rho}\right)_{P, T}^{2} u(\rho)^{2}+\left(\frac{\partial Z}{\partial T}\right)_{P, \rho}^{2} u(T)^{2}} \tag{A6}
\end{equation*}
$$

where the partial derivatives are obtained from deriving the equation A5 as follows:

$$
\begin{equation*}
\left(\frac{\partial Z}{\partial P}\right)_{\rho, T}=\frac{1}{\rho R T} \quad\left(\frac{\partial Z}{\partial \rho}\right)_{P, T}=-\frac{P}{\rho^{2} R T} \quad\left(\frac{\partial Z}{\partial T}\right)_{\rho, P}=-\frac{P}{\rho R T^{2}} \tag{A7}
\end{equation*}
$$

Table 1 - Available experimental $\rho$-PTx data in the literature with their uncertainties for the methane and hydrogen sulphide system.

| Reference | Year | No of Data | $\mathrm{H}_{2} \mathrm{~S}$ mol fraction |  | Pressure |  | Temperature |  | Density |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  | Range (\%) | $\begin{gathered} \mathrm{u}\left(x_{H 2 S}\right) \\ (\%) \end{gathered}$ | $\begin{aligned} & \text { Range } \\ & \text { (MPa) } \end{aligned}$ | $\begin{gathered} \hline \mathrm{u}(P) \\ (\mathrm{MPa}) \\ \hline \end{gathered}$ | Range (K) | $\begin{gathered} \mathrm{u}(T) \\ (\mathrm{K}) \\ \hline \end{gathered}$ | $\begin{gathered} \text { Range } \\ \left(\mathrm{kg} \cdot \mathrm{~m}^{-3}\right) \end{gathered}$ | $\begin{gathered} \mathrm{u}(P) \\ (\%) \end{gathered}$ |
| Reamer et al. [8] | 1951 | 1140 | 10-90 | 0.3 | 1-69 | 0.001 | 277-411 | 0.01 | 1-69 | 0.5 |
| Bailey et al. [10] | 1987 | 80 | 50.7 | 0.01 | 0.2-38 | 0.003 | 299-501 | 0.01 | 1-230 | 1 |

Table 2 - Details of the chemicals, suppliers and purities of the components used in this study.

| Chemical Name | Source | Initial Purity $^{\text {a }}$ | Certification | Analysis Method $^{\text {b }}$ |
| :---: | :---: | :---: | :---: | :---: |
| Hydrogen sulphide | Air Liquide | 0.995 vol | Air Liquide Certified | SM |
| Methane | Air Liquide | 0.99995 vol | Air Liquide Certified | SM |

a No additional purification is carried out.
${ }^{\mathrm{b}}$ SM: Supplier method

Table 3 - Critical parameters, acentric factors and volume correction parameters for methane and hydrogen sulphide.

| Compound | $\mathbf{T}_{\mathbf{c}} / \mathbf{K}$ | $\mathbf{P}_{\mathbf{c}} / \mathbf{M P a}$ | $\boldsymbol{\omega}$ | $\mathbf{V}^{\mathbf{C}} / \mathbf{m}^{3 \cdot} \mathbf{m}^{-\mathbf{1}}$ |
| :---: | :---: | :---: | :---: | :---: |
| $\mathbf{C H}_{\mathbf{4}}$ | 190.58 | 4.604 | 0.01083 | -5.02 |
| $\mathbf{H}_{\mathbf{2}} \mathbf{S}$ | 373.20 | 8.963 | 0.081 | -4.02 |

Table 4 -Mie molecular parameters for pure methane and hydrogen sulphide, and average absolute deviation (\%AAD)from pure experimental data [32] for the vapour pressure $\left(P_{\text {sat }}\right)$, the saturated-liquid density $\left(\rho_{\text {sat }}\right)$ and the enthalpy of vaporization $\left(\Delta H_{v}\right)$.

| Compound | $\mathbf{m}_{\mathbf{s}}$ | $\boldsymbol{\sigma} / \AA$ | $(\boldsymbol{\varepsilon} / \mathbf{k}) / \mathbf{K}$ | $\boldsymbol{\lambda}_{\mathbf{r}}$ | $\boldsymbol{\lambda}_{\mathbf{a}}$ | AAD (\%) |  |  |  |
| :--- | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  | $\mathbf{H}_{\mathbf{2}} \mathbf{S}$ (this work) | 1 |  | 387.28 | 22.451 | 6 | 2.2 | 0.4 | $\boldsymbol{\rho}_{\text {sat }}$ |
| $\mathbf{C H}_{4}[12]$ | 1 | 3.7412 | 153.36 | 12.650 | 6 |  | 0.7 |  |  |

Table 5 - Regressed $\mathrm{k}_{\mathrm{ij}}$ values for PR and SAFT-VR Mie EoSs and their absolute average deviations (\%AAD) in bubble point pressure and vapour phase composition calculations for the methane + hydrogen sulphide binary system.

|  | $\mathbf{k}_{\mathrm{ij}}$ | \%AAD P $^{\text {bubble }}$ | ${\text { \%AAD } \mathbf{y}_{\mathbf{1}}}^{\text {PR }}$ |
| :--- | :---: | :---: | :---: |
| SAFT-VR Mie | 0.0807 | 4.4 | 1.9 |

Table 6 - Experimental results of the $\mathbf{0 . 8 6 8 5}$ mole $\mathbf{C H}_{4}+\mathbf{0 . 1 3 1 5}$ mole $\mathbf{H}_{2} \mathrm{~S}$ system at $253 \mathrm{~K}, \mathbf{u}\left(\mathbf{x}_{\mathbf{H} 2 \mathrm{~S}}\right)=0.0006$, $\mathbf{u}(\mathbf{T})=\mathbf{0 . 0 3} \mathrm{K}, \mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 3} \mathrm{MPa}$ for pressures up to 5 MPa and $\mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 5} \mathrm{MPa}$ for pressures from 5 to 30 MPa.

| No | T <br> [K] | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\begin{aligned} & \mathbf{u}(\mathbf{Z}) \\ & 10^{-2} \end{aligned}$ |  | $\mathbf{T}$[K] | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\begin{aligned} & u(Z) \\ & 10^{-2} \end{aligned}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |
| 1 | 253.28 | 0.188 | 1.76 | 0.18 | 10.1 | 0.93 | 9.4 | 37 | 253.46 | 12.710 | 199.55 | 1.00 | 0.5 | 0.56 | 0.3 |
| 2 | 253.29 | 0.508 | 4.80 | 0.22 | 4.7 | 0.92 | 4.3 | 38 | 253.47 | 13.026 | 204.72 | 1.03 | 0.5 | 0.56 | 0.3 |
| 3 | 253.30 | 1.250 | 12.25 | 0.33 | 2.7 | 0.89 | 2.4 | 39 | 253.48 | 13.361 | 209.99 | 1.06 | 0.5 | 0.56 | 0.3 |
| 4 | 253.31 | 1.824 | 18.07 | 0.41 | 2.3 | 0.88 | 2.0 | 40 | 253.49 | 13.557 | 213.18 | 1.07 | 0.5 | 0.56 | 0.3 |
| 5 | 253.33 | 2.475 | 24.75 | 0.50 | 2.0 | 0.87 | 1.8 | 41 | 253.50 | 13.904 | 217.77 | 1.10 | 0.5 | 0.56 | 0.3 |
| 6 | 253.35 | 3.335 | 33.57 | 0.52 | 1.5 | 0.87 | 1.3 | 42 | 253.53 | 14.217 | 222.31 | 1.13 | 0.5 | 0.56 | 0.3 |
| 7 | 253.36 | 4.114 | 41.57 | 0.54 | 1.3 | 0.87 | 1.1 | 43 | 253.53 | 14.605 | 227.86 | 1.16 | 0.5 | 0.56 | 0.3 |
| 8 | 253.38 | 4.487 | 45.79 | 0.55 | 1.2 | 0.86 | 1.0 | 44 | 253.54 | 15.328 | 236.23 | 1.23 | 0.5 | 0.57 | 0.3 |
| 9 | 253.34 | 4.807 | 49.57 | 0.56 | 1.1 | 0.85 | 1.0 | 45 | 253.56 | 16.209 | 246.09 | 1.31 | 0.5 | 0.58 | 0.3 |
| 10 | 253.33 | 5.174 | 53.92 | 0.57 | 1.1 | 0.84 | 0.9 | 46 | 253.58 | 16.612 | 250.04 | 1.33 | 0.5 | 0.58 | 0.3 |
| 11 | 253.32 | 5.258 | 55.98 | 0.58 | 1.0 | 0.82 | 0.8 | 47 | 253.59 | 17.129 | 255.23 | 1.26 | 0.5 | 0.59 | 0.3 |
| 12 | 253.33 | 5.895 | 64.88 | 0.60 | 0.9 | 0.79 | 0.7 | 48 | 253.60 | 17.589 | 259.35 | 1.21 | 0.5 | 0.59 | 0.3 |
| 13 | 253.32 | 6.503 | 73.25 | 0.62 | 0.9 | 0.78 | 0.7 | 49 | 253.61 | 18.036 | 263.28 | 1.15 | 0.4 | 0.60 | 0.3 |
| 14 | 253.34 | 6.994 | 80.75 | 0.65 | 0.8 | 0.76 | 0.6 | 50 | 253.61 | 18.480 | 267.15 | 1.10 | 0.4 | 0.60 | 0.2 |
| 15 | 253.36 | 7.255 | 84.54 | 0.66 | 0.8 | 0.75 | 0.6 | 51 | 253.62 | 18.822 | 269.80 | 1.06 | 0.4 | 0.61 | 0.2 |
| 16 | 253.35 | 7.572 | 90.00 | 0.67 | 0.7 | 0.74 | 0.6 | 52 | 253.63 | 19.390 | 274.47 | 1.00 | 0.4 | 0.62 | 0.2 |
| 17 | 253.35 | 7.961 | 99.59 | 0.69 | 0.7 | 0.70 | 0.5 | 53 | 253.64 | 19.853 | 277.53 | 0.96 | 0.3 | 0.63 | 0.2 |
| 18 | 253.36 | 8.383 | 108.50 | 0.72 | 0.7 | 0.68 | 0.4 | 54 | 253.66 | 21.081 | 286.30 | 0.84 | 0.3 | 0.64 | 0.2 |
| 19 | 253.37 | 8.539 | 112.16 | 0.72 | 0.6 | 0.67 | 0.4 | 55 | 253.66 | 21.605 | 289.87 | 0.80 | 0.3 | 0.65 | 0.2 |
| 20 | 253.38 | 8.660 | 115.11 | 0.73 | 0.6 | 0.66 | 0.4 | 56 | 253.67 | 22.087 | 292.82 | 0.76 | 0.3 | 0.66 | 0.2 |
| 21 | 253.38 | 8.850 | 119.77 | 0.74 | 0.6 | 0.65 | 0.4 | 57 | 253.68 | 22.512 | 295.31 | 0.73 | 0.2 | 0.67 | 0.2 |
| 22 | 253.39 | 9.030 | 124.20 | 0.75 | 0.6 | 0.64 | 0.4 | 58 | 253.68 | 22.918 | 297.52 | 0.70 | 0.2 | 0.67 | 0.2 |
| 23 | 253.40 | 9.215 | 128.92 | 0.76 | 0.6 | 0.63 | 0.4 | 59 | 253.56 | 23.487 | 300.68 | 0.66 | 0.2 | 0.68 | 0.2 |
| 24 | 253.40 | 9.484 | 135.48 | 0.78 | 0.6 | 0.61 | 0.4 | 60 | 253.29 | 24.245 | 303.67 | 0.62 | 0.2 | 0.70 | 0.1 |
| 25 | 253.43 | 9.711 | 140.67 | 0.79 | 0.6 | 0.60 | 0.3 | 61 | 253.29 | 24.733 | 306.89 | 0.59 | 0.2 | 0.70 | 0.1 |
| 26 | 253.43 | 9.944 | 145.46 | 0.81 | 0.6 | 0.60 | 0.3 | 62 | 253.30 | 25.670 | 310.08 | 0.55 | 0.2 | 0.72 | 0.1 |
| 27 | 253.41 | 10.167 | 150.17 | 0.82 | 0.5 | 0.59 | 0.3 | 63 | 253.57 | 26.154 | 313.25 | 0.53 | 0.2 | 0.73 | 0.1 |
| 28 | 253.42 | 10.405 | 155.11 | 0.84 | 0.5 | 0.59 | 0.3 | 64 | 253.72 | 26.573 | 314.90 | 0.51 | 0.2 | 0.74 | 0.1 |
| 29 | 253.44 | 10.674 | 160.71 | 0.85 | 0.5 | 0.58 | 0.3 | 65 | 253.53 | 27.036 | 316.40 | 0.50 | 0.2 | 0.75 | 0.1 |
| 30 | 253.42 | 10.879 | 164.94 | 0.87 | 0.5 | 0.58 | 0.3 | 66 | 253.32 | 27.359 | 317.71 | 0.49 | 0.2 | 0.75 | 0.1 |
| 31 | 253.42 | 11.161 | 170.62 | 0.89 | 0.5 | 0.57 | 0.3 | 67 | 253.32 | 27.986 | 320.59 | 0.48 | 0.1 | 0.76 | 0.1 |
| 32 | 253.45 | 11.408 | 175.60 | 0.90 | 0.5 | 0.57 | 0.3 | 68 | 253.33 | 28.601 | 322.89 | 0.47 | 0.1 | 0.77 | 0.1 |
| 33 | 253.46 | 11.691 | 181.06 | 0.93 | 0.5 | 0.56 | 0.3 | 69 | 253.33 | 29.015 | 324.42 | 0.46 | 0.1 | 0.78 | 0.1 |
| 34 | 253.46 | 11.951 | 186.09 | 0.94 | 0.5 | 0.56 | 0.3 | 70 | 253.31 | 29.542 | 326.32 | 0.46 | 0.1 | 0.79 | 0.1 |
| 35 | 253.46 | 12.221 | 190.99 | 0.96 | 0.5 | 0.56 | 0.3 | 71 | 253.33 | 30.002 | 327.17 | 0.46 | 0.1 | 0.80 | 0.1 |
| 36 | 253.45 | 12.503 | 195.90 | 0.99 | 0.5 | 0.56 | 0.3 |  |  |  |  |  |  |  |  |

Table 7 - Experimental results of the $\mathbf{0 . 8 6 8 5}$ mole $\mathrm{CH}_{4}+0.1315$ mole $\mathbf{H}_{2} \mathrm{~S}$ system at $273 \mathrm{~K}, \mathbf{u}\left(\mathrm{x}_{\mathrm{H} 2 \mathrm{~S}}\right)=\mathbf{0 . 0 0 0 6}$, $\mathbf{u}(\mathbf{T})=\mathbf{0 . 0 3} \mathrm{K}, \mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 3} \mathrm{MPa}$ for pressures up to 5 MPa and $\mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 5} \mathrm{MPa}$ for pressures from 5 to 30 MPa.

| No | T <br> [K] | $\mathbf{P}$$[\mathrm{MPa}]$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\mathbf{u}(\mathbf{Z})$$10^{-2}$ | No | T[K] | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\begin{aligned} & u(Z) \\ & 10^{-2} \end{aligned}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |
| 1 | 273.21 | 0.209 | 1.70 | 0.17 | 10.0 | 0.99 | 9.6 | 31 | 273.26 | 11.726 | 146.08 | 0.69 | 0.5 | 0.65 | 0.3 |
| 2 | 273.21 | 0.648 | 5.39 | 0.22 | 4.1 | 0.97 | 4.0 | 32 | 273.26 | 12.002 | 150.33 | 0.70 | 0.5 | 0.65 | 0.3 |
| 3 | 273.24 | 1.167 | 10.00 | 0.26 | 2.6 | 0.95 | 2.5 | 33 | 273.26 | 12.322 | 155.19 | 0.72 | 0.5 | 0.64 | 0.3 |
| 4 | 273.24 | 1.761 | 15.44 | 0.30 | 2.0 | 0.92 | 1.8 | 34 | 273.26 | 12.637 | 159.86 | 0.74 | 0.5 | 0.64 | 0.3 |
| 5 | 273.25 | 2.240 | 19.97 | 0.34 | 1.7 | 0.91 | 1.5 | 35 | 273.26 | 13.040 | 165.72 | 0.76 | 0.5 | 0.64 | 0.3 |
| 6 | 273.26 | 2.770 | 25.12 | 0.37 | 1.5 | 0.89 | 1.3 | 36 | 273.27 | 13.466 | 171.70 | 0.78 | 0.5 | 0.64 | 0.3 |
| 7 | 273.27 | 3.280 | 30.25 | 0.40 | 1.3 | 0.88 | 1.1 | 37 | 273.27 | 13.771 | 175.85 | 0.80 | 0.5 | 0.64 | 0.3 |
| 8 | 273.25 | 3.776 | 35.41 | 0.42 | 1.2 | 0.86 | 1.0 | 38 | 273.30 | 14.847 | 189.62 | 0.87 | 0.5 | 0.63 | 0.3 |
| 9 | 273.27 | 4.207 | 40.03 | 0.44 | 1.1 | 0.85 | 0.9 | 39 | 273.31 | 15.329 | 195.37 | 0.90 | 0.5 | 0.64 | 0.3 |
| 10 | 273.27 | 4.225 | 40.23 | 0.44 | 1.1 | 0.85 | 0.9 | 40 | 273.32 | 15.811 | 200.85 | 0.93 | 0.5 | 0.64 | 0.3 |
| 11 | 273.26 | 4.666 | 45.13 | 0.45 | 1.0 | 0.84 | 0.8 | 41 | 273.32 | 16.120 | 204.24 | 0.96 | 0.5 | 0.64 | 0.3 |
| 12 | 273.26 | 5.086 | 49.93 | 0.47 | 0.9 | 0.83 | 0.8 | 42 | 273.32 | 16.817 | 211.55 | 1.00 | 0.5 | 0.64 | 0.3 |
| 13 | 273.28 | 5.512 | 54.98 | 0.48 | 0.9 | 0.81 | 0.7 | 43 | 273.33 | 17.352 | 216.85 | 1.04 | 0.5 | 0.65 | 0.3 |
| 14 | 273.28 | 5.933 | 60.12 | 0.48 | 0.8 | 0.80 | 0.6 | 44 | 273.34 | 17.978 | 222.75 | 1.09 | 0.5 | 0.65 | 0.3 |
| 15 | 273.30 | 6.333 | 65.17 | 0.49 | 0.8 | 0.79 | 0.6 | 45 | 273.34 | 18.233 | 225.06 | 1.11 | 0.5 | 0.66 | 0.3 |
| 16 | 273.31 | 6.712 | 70.10 | 0.51 | 0.7 | 0.78 | 0.6 | 46 | 273.35 | 18.773 | 229.81 | 1.05 | 0.5 | 0.66 | 0.3 |
| 17 | 273.31 | 7.200 | 76.68 | 0.52 | 0.7 | 0.76 | 0.5 | 47 | 273.35 | 19.531 | 236.12 | 0.97 | 0.4 | 0.67 | 0.3 |
| 18 | 273.31 | 7.483 | 80.60 | 0.52 | 0.6 | 0.75 | 0.5 | 48 | 273.37 | 20.048 | 240.23 | 0.83 | 0.3 | 0.68 | 0.2 |
| 19 | 273.31 | 7.817 | 85.33 | 0.53 | 0.6 | 0.74 | 0.5 | 49 | 273.36 | 20.692 | 245.12 | 0.68 | 0.3 | 0.68 | 0.2 |
| 20 | 273.29 | 8.136 | 89.96 | 0.54 | 0.6 | 0.73 | 0.4 | 50 | 273.33 | 21.405 | 250.29 | 0.54 | 0.2 | 0.69 | 0.1 |
| 21 | 273.28 | 8.496 | 95.31 | 0.55 | 0.6 | 0.72 | 0.4 | 51 | 273.31 | 22.905 | 260.37 | 0.36 | 0.1 | 0.71 | 0.1 |
| 22 | 273.29 | 8.809 | 100.05 | 0.56 | 0.6 | 0.71 | 0.4 | 52 | 273.29 | 23.690 | 265.29 | 0.34 | 0.1 | 0.72 | 0.1 |
| 23 | 273.27 | 9.149 | 105.29 | 0.57 | 0.5 | 0.70 | 0.4 | 53 | 273.27 | 24.569 | 270.52 | 0.31 | 0.1 | 0.74 | 0.1 |
| 24 | 273.28 | 9.596 | 112.28 | 0.59 | 0.5 | 0.69 | 0.4 | 54 | 273.27 | 25.361 | 275.02 | 0.29 | 0.1 | 0.75 | 0.1 |
| 25 | 273.27 | 9.851 | 116.33 | 0.60 | 0.5 | 0.69 | 0.4 | 55 | 273.27 | 26.319 | 280.21 | 0.26 | 0.1 | 0.76 | 0.1 |
| 26 | 273.28 | 10.137 | 120.88 | 0.61 | 0.5 | 0.68 | 0.3 | 56 | 273.26 | 27.255 | 285.03 | 0.24 | 0.1 | 0.77 | 0.1 |
| 27 | 273.29 | 10.446 | 125.82 | 0.63 | 0.5 | 0.67 | 0.3 | 57 | 273.26 | 28.276 | 290.05 | 0.23 | 0.1 | 0.79 | 0.1 |
| 28 | 273.29 | 10.749 | 130.66 | 0.64 | 0.5 | 0.67 | 0.3 | 58 | 273.26 | 29.372 | 295.19 | 0.21 | 0.1 | 0.81 | 0.1 |
| 29 | 273.26 | 11.062 | 135.65 | 0.65 | 0.5 | 0.66 | 0.3 | 59 | 273.25 | 30.481 | 300.13 | 0.21 | 0.1 | 0.82 | 0.1 |
| 30 | 273.25 | 11.361 | 140.37 | 0.67 | 0.5 | 0.66 | 0.3 |  |  |  |  |  |  |  |  |

Table 8 - Experimental results of the $\mathbf{0 . 8 6 8 5}$ mole $\mathrm{CH}_{4}+0.1315$ mole $\mathbf{H}_{2} \mathrm{~S}$ system at $293 \mathrm{~K}, \mathbf{u}\left(\mathrm{x}_{\mathrm{H} 2 \mathrm{~S}}\right)=\mathbf{0 . 0 0 0 6}$, $\mathbf{u}(\mathbf{T})=\mathbf{0 . 0 3} \mathrm{K}, \mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 3} \mathrm{MPa}$ for pressures up to 5 MPa and $\mathbf{u}(\mathbf{P})=0.005 \mathrm{MPa}$ for pressures from 5 to 30 MPa.

|  | T <br> [K] | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\mathbf{u}(\mathbf{Z})$$10^{-2}$ |  | T <br> [K] | $\begin{gathered} \mathbf{P} \\ {[\mathbf{M P a}]} \end{gathered}$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\begin{aligned} & \mathbf{u}(\mathbf{Z}) \\ & 10^{-2} \end{aligned}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |  |  |  |  | $\left.\frac{k g}{m^{3}}\right]$ | $\%$ |  |  |
| 1 | 293.18 | 0.239 | 1.86 | 0.18 | 9.5 | 0.97 | 9.2 | 31 | 293.19 | 13.230 | 140.02 | 0.71 | 0.5 | 0.71 | 0.4 |
| 2 | 293.18 | 0.469 | 3.71 | 0.19 | 5.3 | 0.96 | 5.0 | 32 | 293.18 | 13.726 | 145.96 | 0.74 | 0.5 | 0.71 | 0.4 |
| 3 | 293.19 | 0.792 | 6.46 | 0.22 | 3.4 | 0.93 | 3.2 | 33 | 293.20 | 14.242 | 152.42 | 0.76 | 0.5 | 0.71 | 0.4 |
| 4 | 293.21 | 1.117 | 9.20 | 0.24 | 2.7 | 0.92 | 2.4 | 34 | 293.20 | 14.579 | 156.48 | 0.78 | 0.5 | 0.70 | 0.4 |
| 5 | 293.19 | 1.341 | 11.22 | 0.26 | 2.3 | 0.90 | 2.1 | 35 | 293.20 | 14.924 | 160.62 | 0.81 | 0.5 | 0.70 | 0.4 |
| 6 | 293.20 | 1.820 | 15.41 | 0.29 | 1.9 | 0.89 | 1.7 | 36 | 293.20 | 15.365 | 165.51 | 0.83 | 0.5 | 0.70 | 0.4 |
| 7 | 293.19 | 2.300 | 20.14 | 0.32 | 1.6 | 0.86 | 1.4 | 37 | 293.20 | 15.806 | 170.31 | 0.86 | 0.5 | 0.70 | 0.4 |
| 8 | 293.18 | 2.890 | 25.81 | 0.36 | 1.4 | 0.85 | 1.2 | 38 | 293.19 | 16.228 | 174.96 | 0.89 | 0.5 | 0.70 | 0.4 |
| 9 | 293.19 | 3.396 | 30.35 | 0.38 | 1.3 | 0.85 | 1.1 | 39 | 293.19 | 16.699 | 180.07 | 0.92 | 0.5 | 0.70 | 0.4 |
| 10 | 293.19 | 3.988 | 35.82 | 0.41 | 1.1 | 0.84 | 1.0 | 40 | 293.20 | 17.183 | 185.14 | 0.95 | 0.5 | 0.70 | 0.4 |
| 11 | 293.19 | 4.428 | 40.25 | 0.42 | 1.0 | 0.83 | 0.9 | 41 | 293.19 | 17.682 | 190.29 | 0.98 | 0.5 | 0.70 | 0.4 |
| 12 | 293.20 | 4.887 | 44.78 | 0.44 | 1.0 | 0.82 | 0.8 | 42 | 293.20 | 18.163 | 195.16 | 1.01 | 0.5 | 0.70 | 0.4 |
| 13 | 293.20 | 5.494 | 50.99 | 0.45 | 0.9 | 0.81 | 0.7 | 43 | 293.23 | 18.686 | 200.27 | 0.99 | 0.5 | 0.71 | 0.3 |
| 14 | 293.20 | 5.964 | 55.59 | 0.46 | 0.8 | 0.81 | 0.7 | 44 | 293.29 | 19.219 | 205.31 | 0.87 | 0.4 | 0.71 | 0.3 |
| 15 | 293.20 | 6.411 | 60.21 | 0.46 | 0.8 | 0.80 | 0.6 | 45 | 293.23 | 19.770 | 210.30 | 0.73 | 0.3 | 0.71 | 0.2 |
| 16 | 293.19 | 6.909 | 65.40 | 0.47 | 0.7 | 0.80 | 0.6 | 46 | 293.22 | 20.355 | 215.54 | 0.58 | 0.3 | 0.71 | 0.2 |
| 17 | 293.19 | 7.335 | 70.05 | 0.48 | 0.7 | 0.79 | 0.5 | 47 | 293.20 | 20.932 | 220.42 | 0.43 | 0.2 | 0.72 | 0.1 |
| 18 | 293.19 | 7.786 | 74.96 | 0.49 | 0.7 | 0.79 | 0.5 | 48 | 293.20 | 21.495 | 224.99 | 0.39 | 0.2 | 0.72 | 0.1 |
| 19 | 293.18 | 8.263 | 79.99 | 0.50 | 0.6 | 0.78 | 0.5 | 49 | 293.23 | 22.173 | 230.34 | 0.37 | 0.2 | 0.73 | 0.1 |
| 20 | 293.18 | 8.722 | 85.68 | 0.51 | 0.6 | 0.77 | 0.5 | 50 | 293.23 | 22.860 | 235.56 | 0.34 | 0.1 | 0.73 | 0.1 |
| 21 | 293.19 | 9.097 | 90.09 | 0.53 | 0.6 | 0.76 | 0.4 | 51 | 293.21 | 23.505 | 240.20 | 0.32 | 0.1 | 0.74 | 0.1 |
| 22 | 293.19 | 9.534 | 95.16 | 0.54 | 0.6 | 0.76 | 0.4 | 52 | 293.22 | 24.254 | 245.19 | 0.30 | 0.1 | 0.75 | 0.1 |
| 23 | 293.18 | 9.972 | 100.39 | 0.56 | 0.6 | 0.75 | 0.4 | 53 | 293.22 | 25.052 | 250.52 | 0.28 | 0.1 | 0.76 | 0.1 |
| 24 | 293.19 | 10.368 | 105.20 | 0.57 | 0.5 | 0.74 | 0.4 | 54 | 293.21 | 25.865 | 255.77 | 0.26 | 0.1 | 0.76 | 0.1 |
| 25 | 293.19 | 10.794 | 110.28 | 0.59 | 0.5 | 0.74 | 0.4 | 55 | 293.21 | 26.669 | 260.49 | 0.24 | 0.1 | 0.77 | 0.1 |
| 26 | 293.19 | 11.183 | 115.06 | 0.61 | 0.5 | 0.73 | 0.4 | 56 | 293.20 | 27.629 | 265.96 | 0.22 | 0.1 | 0.79 | 0.1 |
| 27 | 293.19 | 11.608 | 120.16 | 0.63 | 0.5 | 0.73 | 0.4 | 57 | 293.20 | 28.496 | 270.59 | 0.21 | 0.1 | 0.80 | 0.1 |
| 28 | 293.18 | 12.003 | 124.94 | 0.65 | 0.5 | 0.73 | 0.4 | 58 | 293.21 | 29.415 | 275.27 | 0.20 | 0.1 | 0.81 | 0.1 |
| 29 | 293.18 | 12.415 | 130.08 | 0.67 | 0.5 | 0.72 | 0.4 | 59 | 293.21 | 29.995 | 278.16 | 0.20 | 0.1 | 0.81 | 0.1 |
| 30 | 293.18 | 12.830 | 135.13 | 0.69 | 0.5 | 0.72 | 0.4 | 60 | 293.21 | 30.419 | 280.17 | 0.19 | 0.1 | 0.82 | 0.1 |

Table 9 - Experimental results of 0.8197 mole $^{C_{H}}+\mathbf{0 . 1 8 0 3}$ mole $\mathbf{H}_{2} \mathrm{~S}$ system at $253 \mathrm{~K}, \mathbf{u}\left(\mathrm{x}_{\mathrm{H} 2 \mathrm{~S}}\right)=0.0008$, $\mathbf{u}(\mathbf{T})=\mathbf{0 . 0 3} \mathrm{K}, \mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 3} \mathrm{MPa}$ for pressures up to 5 MPa and $\mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 5} \mathrm{MPa}$ for pressures from 5 to 30 MPa.

| No | T[K] | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\mathbf{u}(\mathbf{Z})$$10^{-2}$ | No | T[K] | P [MPa] | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\begin{gathered} u(Z) \\ 10^{-2} \end{gathered}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ |  |  |  |  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |
| 1 | 253.03 | 0.225 | 2.10 | 0.18 | 8.7 | 0.98 | 8.6 | 31 | 252.98 | 16.314 | 285.18 | 1.32 | 0.5 | 0.53 | 0.2 |
| 2 | 253.05 | 0.472 | 4.43 | 0.22 | 4.9 | 0.97 | 4.8 | 32 | 252.99 | 16.702 | 288.37 | 1.32 | 0.5 | 0.53 | 0.2 |
| 3 | 253.07 | 1.105 | 10.58 | 0.31 | 2.9 | 0.96 | 2.8 | 33 | 253.01 | 17.275 | 293.02 | 1.24 | 0.4 | 0.54 | 0.2 |
| 4 | 253.08 | 1.496 | 14.53 | 0.36 | 2.5 | 0.94 | 2.4 | 34 | 253.02 | 17.701 | 296.11 | 1.19 | 0.4 | 0.55 | 0.2 |
| 5 | 253.08 | 2.006 | 20.39 | 0.43 | 2.1 | 0.90 | 1.9 | 35 | 253.03 | 18.070 | 299.00 | 1.15 | 0.4 | 0.55 | 0.2 |
| 6 | 253.25 | 2.607 | 28.21 | 0.50 | 1.8 | 0.85 | 1.5 | 36 | 253.02 | 18.537 | 302.29 | 1.10 | 0.4 | 0.56 | 0.2 |
| 7 | 253.21 | 2.947 | 32.50 | 0.51 | 1.6 | 0.83 | 1.3 | 37 | 253.02 | 19.009 | 305.49 | 1.04 | 0.3 | 0.57 | 0.2 |
| 8 | 253.16 | 3.517 | 38.95 | 0.52 | 1.3 | 0.83 | 1.1 | 38 | 253.02 | 19.564 | 309.06 | 0.99 | 0.3 | 0.58 | 0.2 |
| 9 | 253.19 | 4.054 | 46.42 | 0.54 | 1.2 | 0.80 | 0.9 | 39 | 253.01 | 20.084 | 312.27 | 0.93 | 0.3 | 0.59 | 0.2 |
| 10 | 253.18 | 4.507 | 52.31 | 0.55 | 1.1 | 0.79 | 0.8 | 40 | 253.01 | 20.574 | 315.19 | 0.89 | 0.3 | 0.60 | 0.2 |
| 11 | 253.12 | 4.809 | 59.33 | 0.56 | 0.9 | 0.74 | 0.7 | 41 | 253.00 | 21.010 | 317.91 | 0.85 | 0.3 | 0.61 | 0.2 |
| 12 | 253.13 | 5.304 | 67.27 | 0.58 | 0.9 | 0.72 | 0.6 | 42 | 253.00 | 21.473 | 320.52 | 0.81 | 0.3 | 0.61 | 0.2 |
| 13 | 252.91 | 9.089 | 146.00 | 0.75 | 0.5 | 0.57 | 0.3 | 43 | 253.00 | 21.973 | 323.20 | 0.77 | 0.2 | 0.62 | 0.1 |
| 14 | 252.91 | 9.296 | 153.90 | 0.77 | 0.5 | 0.55 | 0.3 | 44 | 253.00 | 22.531 | 326.01 | 0.73 | 0.2 | 0.63 | 0.1 |
| 15 | 252.91 | 9.508 | 162.60 | 0.78 | 0.5 | 0.54 | 0.3 | 45 | 253.00 | 22.822 | 327.46 | 0.71 | 0.2 | 0.64 | 0.1 |
| 16 | 252.91 | 9.727 | 172.12 | 0.79 | 0.5 | 0.52 | 0.2 | 46 | 253.00 | 23.125 | 329.05 | 0.69 | 0.2 | 0.64 | 0.1 |
| 17 | 252.90 | 9.948 | 181.75 | 0.81 | 0.4 | 0.50 | 0.2 | 47 | 253.01 | 23.774 | 332.07 | 0.64 | 0.2 | 0.66 | 0.1 |
| 18 | 252.90 | 10.181 | 191.68 | 0.82 | 0.4 | 0.49 | 0.2 | 48 | 253.01 | 24.145 | 333.77 | 0.62 | 0.2 | 0.66 | 0.1 |
| 19 | 252.90 | 10.440 | 200.58 | 0.84 | 0.4 | 0.48 | 0.2 | 49 | 253.02 | 24.553 | 335.64 | 0.60 | 0.2 | 0.67 | 0.1 |
| 20 | 252.89 | 10.727 | 209.51 | 0.86 | 0.4 | 0.47 | 0.2 | 50 | 253.02 | 25.031 | 337.77 | 0.58 | 0.2 | 0.68 | 0.1 |
| 21 | 252.89 | 11.063 | 217.25 | 0.88 | 0.4 | 0.47 | 0.2 | 51 | 253.03 | 25.582 | 340.15 | 0.55 | 0.2 | 0.69 | 0.1 |
| 22 | 252.89 | 11.429 | 224.63 | 0.91 | 0.4 | 0.47 | 0.2 | 52 | 253.04 | 26.249 | 342.94 | 0.52 | 0.2 | 0.70 | 0.1 |
| 23 | 252.90 | 11.825 | 231.88 | 0.93 | 0.4 | 0.47 | 0.2 | 53 | 253.07 | 26.760 | 345.19 | 0.51 | 0.1 | 0.71 | 0.1 |
| 24 | 252.90 | 12.251 | 239.02 | 0.97 | 0.4 | 0.47 | 0.2 | 54 | 253.07 | 27.152 | 346.63 | 0.50 | 0.1 | 0.72 | 0.1 |
| 25 | 252.91 | 12.717 | 246.10 | 1.00 | 0.4 | 0.47 | 0.2 | 55 | 253.06 | 27.510 | 348.27 | 0.49 | 0.1 | 0.72 | 0.1 |
| 26 | 252.92 | 13.223 | 253.09 | 1.04 | 0.4 | 0.48 | 0.2 | 56 | 253.07 | 28.062 | 350.41 | 0.48 | 0.1 | 0.73 | 0.1 |
| 27 | 252.92 | 13.782 | 260.02 | 1.09 | 0.4 | 0.49 | 0.2 | 57 | 253.08 | 28.543 | 352.25 | 0.47 | 0.1 | 0.74 | 0.1 |
| 28 | 252.94 | 14.392 | 266.89 | 1.14 | 0.4 | 0.50 | 0.2 | 58 | 253.08 | 28.966 | 353.85 | 0.46 | 0.1 | 0.75 | 0.1 |
| 29 | 252.95 | 15.070 | 273.89 | 1.20 | 0.4 | 0.51 | 0.2 | 59 | 253.08 | 29.496 | 355.79 | 0.46 | 0.1 | 0.76 | 0.1 |
| 30 | 252.97 | 15.829 | 280.91 | 1.28 | 0.5 | 0.52 | 0.2 | 60 | 253.08 | 30.039 | 357.73 | 0.46 | 0.1 | 0.77 | 0.1 |

Table 10 - Experimental results of the 0.8197 mole $\mathrm{CH}_{4}+0.1803$ mole $\mathbf{H}_{2} \mathrm{~S}$ system at $273 \mathrm{~K}, \mathbf{u}\left(\mathrm{x}_{\mathbf{H} 2 \mathrm{~S}}\right)=$ $0.0008, \mathbf{u}(\mathbf{T})=0.03 \mathrm{~K}, \mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 3} \mathrm{MPa}$ for pressures up to 5 MPa and $\mathbf{u}(\mathbf{P})=0.005 \mathrm{MPa}$ for pressures from 5 to 30 MPa .

| No | T <br> [K] | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\mathbf{u}(\mathbf{Z})$$10^{-2}$ | No | T$[\mathbf{K}]$ | P [MPa] | $\left[\frac{\mathrm{kg}}{\mathrm{~m}^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\begin{gathered} u(Z) \\ 10^{-2} \end{gathered}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |
| 1 | 273.04 | 0.220 | 1.87 | 0.18 | 9.6 | 1.00 | 9.5 | 32 | 273.13 | 15.510 | 224.73 | 0.91 | 0.4 | 0.59 | 0.2 |
| 2 | 273.05 | 0.512 | 4.39 | 0.21 | 4.8 | 0.99 | 4.6 | 33 | 273.13 | 16.041 | 231.85 | 0.95 | 0.4 | 0.59 | 0.2 |
| 3 | 273.06 | 1.024 | 8.86 | 0.25 | 2.8 | 0.98 | 2.3 | 34 | 273.13 | 16.504 | 237.74 | 0.98 | 0.4 | 0.59 | 0.2 |
| 4 | 273.06 | 1.546 | 13.54 | 0.29 | 2.1 | 0.97 | 2.0 | 35 | 273.11 | 17.004 | 243.74 | 1.02 | 0.4 | 0.59 | 0.2 |
| 5 | 273.06 | 1.998 | 17.83 | 0.32 | 1.8 | 0.95 | 1.7 | 36 | 273.08 | 17.522 | 249.57 | 1.06 | 0.4 | 0.60 | 0.2 |
| 6 | 273.07 | 2.472 | 22.64 | 0.35 | 1.5 | 0.93 | 1.4 | 37 | 273.10 | 18.017 | 254.80 | 1.09 | 0.4 | 0.60 | 0.2 |
| 7 | 273.08 | 3.006 | 28.12 | 0.38 | 1.4 | 0.91 | 1.2 | 38 | 273.08 | 18.504 | 259.61 | 1.13 | 0.4 | 0.61 | 0.2 |
| 8 | 273.08 | 3.496 | 33.34 | 0.41 | 1.2 | 0.89 | 1.1 | 39 | 273.07 | 19.004 | 264.22 | 1.12 | 0.4 | 0.61 | 0.2 |
| 9 | 273.07 | 4.037 | 39.48 | 0.43 | 1.1 | 0.87 | 0.9 | 40 | 273.05 | 19.490 | 268.43 | 0.98 | 0.4 | 0.62 | 0.2 |
| 10 | 273.06 | 4.568 | 45.89 | 0.45 | 1.0 | 0.85 | 0.8 | 41 | 273.06 | 19.898 | 271.74 | 0.87 | 0.3 | 0.62 | 0.1 |
| 11 | 273.02 | 5.020 | 51.69 | 0.46 | 0.9 | 0.83 | 0.7 | 42 | 273.05 | 20.545 | 276.64 | 0.71 | 0.3 | 0.63 | 0.1 |
| 12 | 273.04 | 5.526 | 58.55 | 0.48 | 0.8 | 0.80 | 0.7 | 43 | 273.07 | 21.088 | 280.47 | 0.60 | 0.2 | 0.64 | 0.1 |
| 13 | 273.05 | 6.060 | 66.20 | 0.49 | 0.7 | 0.78 | 0.6 | 44 | 273.08 | 21.490 | 283.16 | 0.52 | 0.2 | 0.64 | 0.1 |
| 14 | 273.05 | 6.535 | 73.36 | 0.49 | 0.7 | 0.76 | 0.5 | 45 | 273.07 | 22.026 | 286.58 | 0.44 | 0.2 | 0.65 | 0.1 |
| 15 | 273.06 | 7.007 | 80.78 | 0.51 | 0.6 | 0.74 | 0.5 | 46 | 273.10 | 22.561 | 289.86 | 0.37 | 0.1 | 0.66 | 0.1 |
| 16 | 273.06 | 7.503 | 88.86 | 0.52 | 0.6 | 0.72 | 0.4 | 47 | 273.11 | 23.102 | 293.06 | 0.36 | 0.1 | 0.67 | 0.1 |
| 17 | 273.08 | 8.037 | 97.86 | 0.54 | 0.5 | 0.70 | 0.4 | 48 | 273.12 | 23.507 | 295.40 | 0.34 | 0.1 | 0.68 | 0.1 |
| 18 | 273.09 | 8.509 | 106.04 | 0.55 | 0.5 | 0.68 | 0.4 | 49 | 273.13 | 24.081 | 298.69 | 0.32 | 0.1 | 0.69 | 0.1 |
| 19 | 273.09 | 8.697 | 109.34 | 0.56 | 0.5 | 0.68 | 0.3 | 50 | 273.14 | 24.511 | 301.13 | 0.31 | 0.1 | 0.69 | 0.1 |
| 20 | 273.11 | 9.417 | 122.17 | 0.58 | 0.5 | 0.66 | 0.3 | 51 | 273.15 | 25.182 | 304.96 | 0.29 | 0.1 | 0.70 | 0.1 |
| 21 | 273.11 | 10.159 | 135.60 | 0.61 | 0.5 | 0.64 | 0.3 | 52 | 273.15 | 25.661 | 307.41 | 0.28 | 0.1 | 0.71 | 0.1 |
| 22 | 273.14 | 10.580 | 143.25 | 0.63 | 0.4 | 0.63 | 0.3 | 53 | 273.14 | 26.076 | 309.49 | 0.27 | 0.1 | 0.72 | 0.1 |
| 23 | 273.13 | 10.896 | 148.97 | 0.65 | 0.4 | 0.62 | 0.3 | 54 | 273.14 | 26.577 | 312.08 | 0.26 | 0.1 | 0.72 | 0.1 |
| 24 | 273.13 | 11.490 | 159.68 | 0.68 | 0.4 | 0.61 | 0.3 | 55 | 273.14 | 26.982 | 314.11 | 0.25 | 0.1 | 0.73 | 0.1 |
| 25 | 273.13 | 12.024 | 169.16 | 0.70 | 0.4 | 0.60 | 0.3 | 56 | 273.13 | 27.492 | 316.71 | 0.24 | 0.1 | 0.74 | 0.1 |
| 26 | 273.17 | 12.519 | 177.81 | 0.73 | 0.4 | 0.60 | 0.2 | 57 | 273.13 | 28.026 | 319.37 | 0.23 | 0.1 | 0.75 | 0.1 |
| 27 | 273.16 | 13.305 | 191.11 | 0.77 | 0.4 | 0.59 | 0.2 | 58 | 273.13 | 28.568 | 321.92 | 0.22 | 0.1 | 0.75 | 0.1 |
| 28 | 273.13 | 13.513 | 194.52 | 0.79 | 0.4 | 0.59 | 0.2 | 59 | 273.13 | 29.088 | 324.13 | 0.22 | 0.1 | 0.76 | 0.1 |
| 29 | 273.13 | 14.007 | 202.45 | 0.82 | 0.4 | 0.59 | 0.2 | 60 | 273.13 | 29.546 | 325.75 | 0.21 | 0.1 | 0.77 | 0.1 |
| 30 | 273.11 | 14.299 | 207.01 | 0.83 | 0.4 | 0.59 | 0.2 | 61 | 273.13 | 29.999 | 327.01 | 0.21 | 0.1 | 0.78 | 0.1 |
| 31 | 273.13 | 14.904 | 216.11 | 0.87 | 0.4 | 0.59 | 0.2 |  |  |  |  |  |  |  |  |

Table 11 - Experimental results of the 0.8197 mole $\mathbf{C H}_{4}+0.1803$ mole $\mathbf{H}_{2} \mathrm{~S}$ system at $293 \mathrm{~K}, \mathbf{u}\left(\mathbf{x}_{\mathbf{H} 2 \mathrm{~S}}\right)=$ $0.0008, \mathbf{u}(\mathrm{~T})=0.03 \mathrm{~K}, \mathbf{u}(\mathbf{P})=0.003 \mathrm{MPa}$ for pressures up to 5 MPa and $\mathbf{u}(\mathbf{P})=0.005 \mathrm{MPa}$ for pressures from 5 to 30 MPa .

|  | T <br> [K] | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\mathbf{u}(\mathbf{Z})$$10^{-2}$ | No | T$[\mathbf{K}]$ | P [MPa] | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\mathbf{u}(\mathbf{Z})$$10^{-2}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |
| 1 | 293.27 | 0.193 | 1.54 | 0.15 | 9.7 | 0.99 | 9.5 | 32 | 293.45 | 15.532 | 182.32 | 0.84 | 0.5 | 0.67 | 0.3 |
| 2 | 293.27 | 0.498 | 4.03 | 0.21 | 5.2 | 0.98 | 5.1 | 33 | 293.41 | 16.017 | 188.26 | 0.87 | 0.5 | 0.67 | 0.3 |
| 3 | 293.35 | 0.936 | 7.74 | 0.23 | 3.0 | 0.96 | 2.9 | 34 | 293.45 | 16.485 | 193.92 | 0.90 | 0.5 | 0.67 | 0.2 |
| 4 | 293.33 | 1.434 | 12.03 | 0.27 | 2.2 | 0.94 | 2.1 | 35 | 293.40 | 17.009 | 200.00 | 0.94 | 0.5 | 0.67 | 0.2 |
| 5 | 293.36 | 1.962 | 16.56 | 0.30 | 1.8 | 0.94 | 1.6 | 36 | 293.41 | 17.526 | 205.67 | 0.97 | 0.5 | 0.67 | 0.2 |
| 6 | 293.35 | 2.422 | 20.50 | 0.33 | 1.6 | 0.93 | 1.4 | 37 | 293.42 | 18.033 | 211.20 | 1.01 | 0.5 | 0.68 | 0.2 |
| 7 | 293.38 | 2.915 | 26.63 | 0.36 | 1.4 | 0.87 | 1.1 | 38 | 293.42 | 18.536 | 216.41 | 1.02 | 0.5 | 0.68 | 0.2 |
| 8 | 293.33 | 3.440 | 30.85 | 0.38 | 1.2 | 0.88 | 1.0 | 39 | 293.41 | 19.019 | 221.14 | 0.92 | 0.4 | 0.68 | 0.2 |
| 9 | 293.42 | 3.941 | 35.88 | 0.40 | 1.1 | 0.87 | 1.0 | 40 | 293.41 | 19.519 | 225.91 | 0.79 | 0.4 | 0.68 | 0.2 |
| 10 | 293.35 | 4.502 | 41.72 | 0.43 | 1.0 | 0.85 | 0.9 | 41 | 293.41 | 20.054 | 230.93 | 0.65 | 0.3 | 0.69 | 0.1 |
| 11 | 293.32 | 5.019 | 46.91 | 0.44 | 0.9 | 0.85 | 0.8 | 42 | 293.42 | 20.511 | 234.95 | 0.54 | 0.2 | 0.69 | 0.1 |
| 12 | 293.32 | 5.508 | 52.59 | 0.45 | 0.9 | 0.83 | 0.8 | 43 | 293.41 | 21.059 | 239.83 | 0.40 | 0.2 | 0.69 | 0.1 |
| 13 | 293.34 | 6.006 | 58.11 | 0.46 | 0.8 | 0.82 | 0.6 | 44 | 293.35 | 21.512 | 243.53 | 0.39 | 0.2 | 0.70 | 0.1 |
| 14 | 293.31 | 6.513 | 63.51 | 0.46 | 0.7 | 0.81 | 0.5 | 45 | 293.34 | 22.090 | 248.06 | 0.37 | 0.1 | 0.70 | 0.1 |
| 15 | 293.32 | 7.004 | 68.50 | 0.47 | 0.7 | 0.81 | 0.5 | 46 | 293.37 | 22.488 | 251.05 | 0.36 | 0.1 | 0.71 | 0.1 |
| 16 | 293.32 | 7.520 | 74.89 | 0.48 | 0.6 | 0.79 | 0.4 | 47 | 293.39 | 22.971 | 254.58 | 0.34 | 0.1 | 0.71 | 0.1 |
| 17 | 293.33 | 8.008 | 80.89 | 0.49 | 0.6 | 0.78 | 0.4 | 48 | 293.41 | 23.490 | 258.21 | 0.32 | 0.1 | 0.72 | 0.1 |
| 18 | 293.32 | 8.507 | 87.23 | 0.51 | 0.6 | 0.77 | 0.4 | 49 | 293.39 | 24.024 | 261.83 | 0.31 | 0.1 | 0.73 | 0.1 |
| 19 | 293.33 | 9.072 | 94.80 | 0.53 | 0.6 | 0.76 | 0.4 | 50 | 293.39 | 24.659 | 266.01 | 0.29 | 0.1 | 0.73 | 0.1 |
| 20 | 293.32 | 9.526 | 100.87 | 0.54 | 0.5 | 0.75 | 0.3 | 51 | 293.42 | 25.034 | 268.44 | 0.28 | 0.1 | 0.74 | 0.1 |
| 21 | 293.37 | 10.013 | 107.38 | 0.56 | 0.5 | 0.74 | 0.3 | 52 | 293.41 | 25.538 | 271.56 | 0.27 | 0.1 | 0.74 | 0.1 |
| 22 | 293.32 | 10.518 | 114.24 | 0.58 | 0.5 | 0.73 | 0.3 | 53 | 293.37 | 26.028 | 274.53 | 0.26 | 0.1 | 0.75 | 0.1 |
| 23 | 293.33 | 10.922 | 121.63 | 0.60 | 0.5 | 0.71 | 0.3 | 54 | 293.42 | 26.523 | 277.36 | 0.25 | 0.1 | 0.76 | 0.1 |
| 24 | 293.35 | 11.685 | 130.63 | 0.63 | 0.5 | 0.71 | 0.3 | 55 | 293.41 | 27.022 | 280.18 | 0.24 | 0.1 | 0.76 | 0.1 |
| 25 | 293.36 | 12.068 | 136.11 | 0.65 | 0.5 | 0.70 | 0.3 | 56 | 293.37 | 27.520 | 283.00 | 0.23 | 0.1 | 0.77 | 0.1 |
| 26 | 293.36 | 12.506 | 142.15 | 0.67 | 0.5 | 0.70 | 0.3 | 57 | 293.38 | 27.986 | 285.56 | 0.22 | 0.1 | 0.78 | 0.1 |
| 27 | 293.36 | 13.036 | 149.41 | 0.70 | 0.5 | 0.69 | 0.3 | 58 | 293.38 | 28.437 | 287.96 | 0.21 | 0.1 | 0.78 | 0.1 |
| 28 | 293.36 | 13.500 | 155.72 | 0.72 | 0.5 | 0.69 | 0.3 | 59 | 293.38 | 28.979 | 290.66 | 0.21 | 0.1 | 0.79 | 0.1 |
| 29 | 293.34 | 14.027 | 162.75 | 0.75 | 0.5 | 0.68 | 0.3 | 60 | 293.37 | 29.405 | 292.65 | 0.20 | 0.1 | 0.80 | 0.1 |
| 30 | 293.34 | 14.501 | 169.12 | 0.78 | 0.5 | 0.68 | 0.3 | 61 | 293.37 | 29.932 | 295.13 | 0.20 | 0.1 | 0.80 | 0.1 |
| 31 | 293.37 | 15.006 | 175.59 | 0.81 | 0.5 | 0.68 | 0.3 |  |  |  |  |  |  |  |  |

Table 12 - Experimental results of the 0.714 mole $\mathbf{C H}_{4}+\mathbf{0 . 2 8 6}$ mole $\mathbf{H}_{2} \mathrm{~S}$ system at $253 \mathrm{~K}, \mathbf{u}\left(\mathbf{x}_{\mathbf{H} 2 \mathrm{~S}}\right)=0.0011$, $\mathbf{u}(\mathbf{T})=0.03 \mathrm{~K}, \mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 3} \mathrm{MPa}$ for pressures up to 5 MPa and $\mathbf{u}(\mathbf{P})=0.005 \mathrm{MPa}$ for pressures from 5 to 30 MPa.

| No | T$[\mathbf{K}]$ | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{\mathrm{kg}}{\mathrm{~m}^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\mathbf{u}(\mathbf{Z})$$10^{-2}$ | No | T$[\mathbf{K}]$ | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\begin{gathered} u(Z) \\ 10^{-2} \end{gathered}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |  |  |  |  | $\left[\frac{\mathrm{kg}}{\mathrm{~m}^{3}}\right]$ | \% |  |  |
| 1 | 252.96 | 0.508 | 5.18 | 0.23 | 4.4 | 0.99 | 4.3 | 21 | 253.11 | 20.550 | 389.58 | 0.89 | 0.2 | 0.53 | 0. |
| 2 | 252.94 | 1.010 | 10.45 | 0.30 | 2.9 | 0.97 | 2.7 | 22 | 253.10 | 21.049 | 391.70 | 0.85 | 0.2 | 0.54 | 0.1 |
| 3 | 252.95 | 1.513 | 16.11 | 0.37 | 2.3 | 0.95 | 2.2 | 23 | 253.15 | 21.583 | 393.97 | 0.80 | 0.2 | 0.55 | 0.1 |
| 4 | 253.00 | 2.008 | 22.87 | 0.43 | 1.9 | 0.88 | 1.7 | 24 | 253.17 | 22.039 | 395.89 | 0.76 | 0.2 | 0.56 | 0.1 |
| 5 | 253.02 | 2.407 | 28.16 | 0.48 | 1.7 | 0.86 | 1.5 | 25 | 253.15 | 22.523 | 397.90 | 0.73 | 0.2 | 0.57 | 0.1 |
| 6 | 253.27 | 13.000 | 342.99 | 1.03 | 0.3 | 0.38 | 0.2 | 26 | 253.09 | 23.043 | 399.91 | 0.69 | 0.2 | 0.58 | 0.1 |
| 7 | 253.23 | 13.535 | 348.43 | 1.07 | 0.3 | 0.39 | 0.2 | 27 | 253.07 | 23.585 | 402.01 | 0.66 | 0.2 | 0.59 | 0.1 |
| 8 | 253.17 | 14.005 | 352.44 | 1.11 | 0.3 | 0.40 | 0.2 | 28 | 253.09 | 24.033 | 403.73 | 0.63 | 0.2 | 0.60 | 0. |
| 9 | 253.25 | 14 | 356.43 | 1.15 | 0. | 0.41 | 0.2 | 29 | 253.12 | 24.495 | 405.58 | 0.60 | 0.1 | 0.61 | 0. |
| 10 | 253.16 | 15.038 | 360.04 | 1.20 | 0.3 | 0.42 | 0.2 | 30 | 253.14 | 25.004 | 407.49 | 0.58 | 0.1 | 0.62 | 0. |
| 11 | 253.22 | 15.498 | 363.19 | 1.25 | 0.3 | 0.43 | 0.2 | 31 | 253.11 | 25.528 | 409.34 | 0.55 | 0.1 | 0.63 | 0.1 |
| 12 | 253.14 | 16.004 | 366.38 | 1.29 | 0.4 | 0.44 | 0.3 | 32 | 253.09 | 25.963 | 410.91 | 0.54 | 0.1 | 0.64 | 0.1 |
| 13 | 253.21 | 16.513 | 369.43 | 1.34 | 0. | 0.45 | 0.3 | 33 | 253.06 | 26.459 | 412.74 | 0.52 | 0.1 | 0.65 | 0 |
| 14 | 253.18 | 17.054 | 372.46 | 1.27 | 0.3 | 0.46 | 0.2 | 34 | 253.05 | 26.980 | 414.51 | 0.50 | 0.1 | 0.66 | 0.1 |
| 15 | 253.13 | 17.511 | 374.95 | 1.22 | 0.3 | 0.47 | 0.2 | 35 | 253.07 | 27.494 | 416.29 | 0.49 | 0.1 | 0.67 | 0.1 |
| 16 | 253.19 | 18.047 | 377.75 | 1.15 | 0.3 | 0.48 | 0.2 | 36 | 253.10 | 28.034 | 418.14 | 0.48 | 0.1 | 0.68 | 0.1 |
| 17 | 253.16 | 18.491 | 380.05 | 1.10 | 0.3 | 0.49 | 0.2 | 37 | 253.13 | 28.568 | 420.00 | 0.47 | 0.1 | 0.69 | 0.1 |
| 18 | 253.12 | 19.058 | 382.70 | 1.04 | 0.3 | 0.50 | 0.2 | 38 | 253.16 | 29.087 | 421.82 | 0.46 | 0.1 | 0.69 | 0.1 |
| 19 | 253.15 | 19.521 | 384.91 | 0.99 | 0.3 | 0.51 | 0.2 | 39 | 253.16 | 29.450 | 423.08 | 0.46 | 0.1 | 0.70 | 0.1 |
| 20 | 253.16 | 20.070 | 387.42 | 0.94 | 0.2 | 0.52 | 0.1 | 40 | 253.16 | 30.025 | 425.02 | 0.46 | 0.1 | 0.71 | 0.1 |

Table 13 - Experimental results of the 0.714 mole $\mathbf{C H}_{4}+\mathbf{0 . 2 8 6}$ mole $\mathbf{H}_{2} \mathrm{~S}$ system at $273 \mathrm{~K}, \mathbf{u}\left(\mathbf{x}_{\mathbf{H} 2 \mathrm{~S}}\right)=0.0011$, $\mathbf{u}(\mathbf{T})=0.03 \mathrm{~K}, \mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 3} \mathrm{MPa}$ for pressures up to 5 MPa and $\mathbf{u}(\mathbf{P})=0.005 \mathrm{MPa}$ for pressures from 5 to 30 MPa.

| No | $\mathbf{T}$$[\mathbf{K}]$ | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{\mathrm{kg}}{\mathrm{~m}^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\mathbf{u}(\mathbf{Z})$$10^{-2}$ | No | T$[\mathbf{K}]$ | P <br> [MPa] | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\begin{gathered} \mathbf{u}(\mathrm{Z}) \\ 10^{-2} \end{gathered}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  |  | $\left[\frac{\mathrm{kg}}{\mathrm{m}^{3}}\right]$ | \% |  |  |  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | $\%$ |  |  |
| 1 | 272.97 | 0.448 | 4.32 | 0.22 | 5.1 | 0.97 | 5.0 | 25 | 273.10 | 18.931 | 331.97 | 1.16 | 0.3 | 0.53 | 0.2 |
| 2 | 272.98 | 0.925 | 9.10 | 0.25 | 2.7 | 0.95 | 2.5 | 26 | 273.16 | 19.608 | 336.94 | 0.94 | 0.3 | 0.54 | 0.2 |
| 3 | 272.99 | 1.499 | 15.62 | 0.29 | 1.8 | 0.89 | 1.6 | 27 | 273.23 | 20.100 | 340.42 | 0.81 | 0.2 | 0.55 | 0.1 |
| 4 | 272.98 | 1.959 | 21.08 | 0.32 | 1.5 | 0.86 | 1.4 | 28 | 273.20 | 20.623 | 344.02 | 0.69 | 0.2 | 0.56 | 0.1 |
| 5 | 272.99 | 2.470 | 27.39 | 0.35 | 1.3 | 0.84 | 1.2 | 29 | 273.19 | 21.199 | 347.78 | 0.57 | 0.2 | 0.57 | 0.1 |
| 6 | 273.07 | 2.991 | 34.29 | 0.38 | 1.1 | 0.81 | 1.0 | 30 | 273.21 | 21.506 | 349.84 | 0.52 | 0.1 | 0.57 | 0.1 |
| 7 | 273.06 | 3.515 | 41.26 | 0.41 | 1.0 | 0.80 | 0.9 | 31 | 273.14 | 21.825 | 351.99 | 0.47 | 0.1 | 0.58 | 0.1 |
| 8 | 273.02 | 4.006 | 49.21 | 0.43 | 0.9 | 0.76 | 0.7 | 32 | 273.16 | 22.152 | 354.18 | 0.42 | 0.1 | 0.58 | 0.1 |
| 9 | 273.02 | 4.495 | 57.70 | 0.45 | 0.8 | 0.73 | 0.6 | 33 | 273.16 | 22.483 | 356.27 | 0.38 | 0.1 | 0.59 | 0.1 |
| 10 | 273.00 | 5.009 | 66.37 | 0.46 | 0.7 | 0.71 | 0.6 | 34 | 273.18 | 22.828 | 358.14 | 0.37 | 0.1 | 0.60 | 0.1 |
| 11 | 273.03 | 5.418 | 73.50 | 0.47 | 0.6 | 0.69 | 0.5 | 35 | 273.20 | 23.590 | 362.19 | 0.34 | 0.1 | 0.61 | 0.1 |
| 12 | 273.01 | 12.511 | 252.68 | 0.73 | 0.3 | 0.46 | 0.2 | 36 | 273.37 | 24.085 | 364.70 | 0.32 | 0.1 | 0.62 | 0.1 |
| 13 | 273.00 | 13.055 | 265.48 | 0.76 | 0.3 | 0.46 | 0.2 | 37 | 273.25 | 24.591 | 367.01 | 0.31 | 0.1 | 0.63 | 0.1 |
| 14 | 273.00 | 13.551 | 273.70 | 0.79 | 0.3 | 0.46 | 0.2 | 38 | 273.19 | 24.981 | 368.95 | 0.30 | 0.1 | 0.63 | 0.1 |
| 15 | 273.00 | 14.019 | 280.64 | 0.82 | 0.3 | 0.47 | 0.2 | 39 | 273.23 | 25.533 | 371.48 | 0.28 | 0.1 | 0.64 | 0.1 |
| 16 | 273.00 | 14.532 | 287.50 | 0.85 | 0.3 | 0.47 | 0.2 | 40 | 273.25 | 26.031 | 373.67 | 0.27 | 0.1 | 0.65 | 0.1 |
| 17 | 273.01 | 14.977 | 293.17 | 0.88 | 0.3 | 0.48 | 0.2 | 41 | 273.22 | 26.410 | 375.25 | 0.26 | 0.1 | 0.66 | 0.1 |
| 18 | 273.00 | 15.457 | 298.99 | 0.91 | 0.3 | 0.48 | 0.2 | 42 | 273.24 | 27.020 | 377.85 | 0.25 | 0.1 | 0.67 | 0.1 |
| 19 | 272.99 | 15.974 | 304.94 | 0.95 | 0.3 | 0.49 | 0.2 | 43 | 273.05 | 27.474 | 379.67 | 0.24 | 0.1 | 0.68 | 0.1 |
| 20 | 272.99 | 16.531 | 310.78 | 0.98 | 0.3 | 0.50 | 0.2 | 44 | 273.06 | 27.956 | 381.54 | 0.23 | 0.1 | 0.68 | 0.1 |
| 21 | 273.03 | 16.977 | 315.19 | 1.02 | 0.3 | 0.50 | 0.2 | 45 | 273.10 | 28.471 | 383.53 | 0.23 | 0.1 | 0.69 | 0.1 |
| 22 | 273.08 | 17.450 | 319.61 | 1.05 | 0.3 | 0.51 | 0.2 | 46 | 273.05 | 29.015 | 385.58 | 0.22 | 0.1 | 0.70 | 0.1 |
| 23 | 273.11 | 17.956 | 324.07 | 1.09 | 0.3 | 0.52 | 0.2 | 47 | 273.03 | 29.571 | 387.72 | 0.21 | 0.1 | 0.71 | 0.1 |
| 24 | 273.09 | 18.521 | 328.70 | 1.13 | 0.3 | 0.53 | 0.2 | 48 | 273.03 | 30.070 | 389.49 | 0.21 | 0.1 | 0.72 | 0.1 |

Table 14 - Experimental results of the 0.714 mole $\mathrm{CH}_{4}+0.286$ mole $\mathbf{H}_{2} \mathrm{~S}$ system at $293 \mathrm{~K}, \mathbf{u}\left(\mathrm{x}_{\mathrm{H} 2 \mathrm{~S}}\right)=0.0011$, $\mathbf{u}(\mathbf{T})=\mathbf{0 . 0 3} \mathrm{K}, \mathbf{u}(\mathbf{P})=\mathbf{0 . 0 0 3} \mathrm{MPa}$ for pressures up to 5 MPa and $\mathbf{u}(\mathbf{P})=0.005 \mathrm{MPa}$ for pressures from 5 to 30 MPa.

| No | $\mathbf{T}$$[\mathbf{K}]$ | $\begin{gathered} \mathbf{P} \\ {[\mathrm{MPa}]} \end{gathered}$ | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\mathbf{u}(\mathbf{Z})$$10^{-2}$ | No | T[K] | P [MPa] | $\left[\frac{k g}{m^{3}}\right]$ | $\mathbf{u}(\mathrm{p})$ |  | Z | $\begin{gathered} u(Z) \\ 10^{-2} \end{gathered}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |  |  |  |  | $\left[\frac{k g}{m^{3}}\right]$ | \% |  |  |
| 1 | 293.27 | 0.227 | 1.99 | 0.19 | 9.5 | 0.99 | 9.3 | 34 | 293.30 | 15.503 | 239.94 | 0.84 | 0.4 | 0.56 | 0.3 |
| 2 | 293.27 | 0.560 | 4.92 | 0.21 | 4.3 | 0.99 | 4.2 | 35 | 293.31 | 16.108 | 249.28 | 0.88 | 0.4 | 0.56 | 0.3 |
| 3 | 293.28 | 1.101 | 9.72 | 0.25 | 2.6 | 0.98 | 2.5 | 36 | 293.34 | 16.524 | 253.44 | 0.91 | 0.4 | 0.57 | 0.3 |
| 4 | 293.28 | 1.547 | 14.1 | 0.29 | 2.1 | 0.95 | 2.0 | 37 | 293.34 | 17.035 | 260.09 | 0.94 | 0.4 | 0.57 | 0.3 |
| 5 | 293.28 | 2.065 | 19.16 | 0.31 | 1.6 | 0.94 | 1.5 | 38 | 293.35 | 17.740 | 267.95 | 0.99 | 0.4 | 0.57 | 0.3 |
| 6 | 293.30 | 2.511 | 23.63 | 0.34 | 1.4 | 0.92 | 1.2 | 39 | 293.33 | 18.012 | 271.26 | 1.00 | 0.4 | 0.58 | 0.3 |
| 7 | 293.29 | 2.976 | 28.53 | 0.37 | 1.3 | 0.91 | 1.1 | 40 | 293.33 | 18.543 | 276.99 | 1.02 | 0.4 | 0.58 | 0.3 |
| 8 | 293.29 | 3.489 | 33.95 | 0.40 | 1.2 | 0.89 | 1.0 | 41 | 293.33 | 19.012 | 281.75 | 0.92 | 0.3 | 0.59 | 0.2 |
| 9 | 293.42 | 3.851 | 38.01 | 0.41 | 1.1 | 0.88 | 1.0 | 42 | 293.33 | 19.476 | 286.23 | 0.80 | 0.3 | 0.59 | 0.2 |
| 10 | 293.43 | 4.386 | 46.59 | 0.42 | 0.9 | 0.82 | 0.8 | 43 | 293.34 | 20.081 | 291.58 | 0.65 | 0.2 | 0.60 | 0.1 |
| 11 | 293.44 | 4.823 | 51.27 | 0.43 | 0.8 | 0.82 | 0.7 | 44 | 293.34 | 20.504 | 295.67 | 0.54 | 0.2 | 0.60 | 0.1 |
| 12 | 293.39 | 5.407 | 58.83 | 0.45 | 0.8 | 0.80 | 0.7 | 45 | 293.31 | 21.026 | 300.53 | 0.41 | 0.1 | 0.61 | 0.1 |
| 13 | 293.32 | 5.943 | 66.82 | 0.46 | 0.7 | 0.77 | 0.6 | 46 | 293.33 | 21.863 | 306.80 | 0.38 | 0.1 | 0.62 | 0.1 |
| 14 | 293.33 | 6.464 | 75.29 | 0.46 | 0.6 | 0.75 | 0.4 | 47 | 293.32 | 22.053 | 308.34 | 0.37 | 0.1 | 0.62 | 0.1 |
| 15 | 293.31 | 6.885 | 80.49 | 0.47 | 0.6 | 0.74 | 0.4 | 48 | 293.31 | 22.118 | 308.67 | 0.37 | 0.1 | 0.62 | 0.1 |
| 16 | 293.27 | 7.261 | 85.92 | 0.48 | 0.6 | 0.74 | 0.4 | 49 | 293.31 | 22.512 | 311.24 | 0.36 | 0.1 | 0.63 | 0.1 |
| 17 | 293.27 | 7.749 | 94.83 | 0.49 | 0.5 | 0.71 | 0.4 | 50 | 293.30 | 22.979 | 314.55 | 0.34 | 0.1 | 0.64 | 0.1 |
| 18 | 293.28 | 8.092 | 103.64 | 0.50 | 0.5 | 0.68 | 0.4 | 51 | 293.30 | 23.460 | 317.83 | 0.32 | 0.1 | 0.64 | 0.1 |
| 19 | 293.29 | 8.523 | 108.80 | 0.51 | 0.5 | 0.68 | 0.4 | 52 | 293.31 | 23.712 | 319.93 | 0.32 | 0.1 | 0.64 | 0.1 |
| 20 | 293.30 | 9.149 | 119.96 | 0.53 | 0.4 | 0.66 | 0.3 | 53 | 293.29 | 23.988 | 321.16 | 0.31 | 0.1 | 0.65 | 0.1 |
| 21 | 293.30 | 9.741 | 129.36 | 0.55 | 0.4 | 0.66 | 0.3 | 54 | 293.30 | 24.512 | 324.48 | 0.29 | 0.1 | 0.66 | 0.1 |
| 22 | 293.33 | 10.169 | 136.99 | 0.57 | 0. | 0.65 | 0.3 | 55 | 293.29 | 25.012 | 327.50 | 0.28 | 0.1 | 0.66 | 0.1 |
| 23 | 293.29 | 10.516 | 145.41 | 0.58 | 0.4 | 0.63 | 0.3 | 56 | 293.30 | 25.515 | 330.43 | 0.27 | 0.1 | 0.67 | 0.1 |
| 24 | 293.29 | 11.236 | 161.66 | 0.61 | 0.4 | 0.60 | 0.3 | 57 | 293.30 | 25.988 | 333.07 | 0.26 | 0.1 | 0.68 | 0.1 |
| 25 | 293.30 | 11.789 | 174.33 | 0.64 | 0.4 | 0.59 | 0.3 | 58 | 293.31 | 26.506 | 335.86 | 0.24 | 0.1 | 0.69 | 0.1 |
| 26 | 293.32 | 12.220 | 183.56 | 0.66 | 0.4 | 0.58 | 0.3 | 59 | 293.33 | 26.879 | 337.31 | 0.24 | 0.1 | 0.69 | 0.1 |
| 27 | 293.32 | 12.521 | 189.88 | 0.67 | 0.4 | 0.57 | 0.3 | 60 | 293.64 | 27.145 | 339.08 | 0.23 | 0.1 | 0.70 | 0.1 |
| 28 | 293.31 | 12.964 | 198.31 | 0.69 | 0.4 | 0.57 | 0.3 | 61 | 293.53 | 27.520 | 340.85 | 0.23 | 0.1 | 0.70 | 0.1 |
| 29 | 293.30 | 13.528 | 208.17 | 0.72 | 0.3 | 0.57 | 0.2 | 62 | 293.43 | 28.085 | 343.33 | 0.22 | 0.1 | 0.71 | 0.1 |
| 30 | 293.29 | 13.780 | 213.21 | 0.74 | 0.3 | 0.56 | 0.2 | 63 | 293.42 | 28.586 | 345.52 | 0.21 | 0.1 | 0.72 | 0.1 |
| 31 | 293.30 | 14.426 | 223.09 | 0.78 | 0.3 | 0.56 | 0.2 | 64 | 293.40 | 29.019 | 347.54 | 0.21 | 0.1 | 0.73 | 0.1 |
| 32 | 293.31 | 14.523 | 224.65 | 0.78 | 0.3 | 0.56 | 0.2 | 65 | 293.33 | 29.517 | 349.82 | 0.20 | 0.1 | 0.73 | 0.1 |
| 33 | 293.30 | 15.049 | 233.30 | 0.81 | 0.3 | 0.56 | 0.2 | 66 | 293.33 | 29.998 | 352.02 | 0.20 | 0.1 | 0.74 | 0.1 |

Table 15 - Absolute average deviations (\%AAD) between the studied models and the densities measured in this work.

| $\mathbf{x}_{\text {H2 } \mathbf{S}}$ | T/K | SAFT-VR Mie | PR | GERG 2008 |
| :--- | :---: | :---: | :---: | :---: |
| $\mathbf{x = 0 . 1 3 1 5}$ | 253 | 3.3 | 4.8 | 2.1 |
|  | 273 | 1.6 | 1.4 | 2.0 |
|  | 293 | 3.9 | 2.2 | 4.3 |
|  | Total | 3.0 | 2.9 | 2.8 |
| $\mathbf{x = 0 . 1 8 0 1}$ | 253 | 1.6 | 3.4 | 2.5 |
|  | 273 | 1.5 | 2.0 | 2.6 |
|  | 293 | 2.5 | 2.0 | 3.3 |
|  | Total | 1.9 | 2.5 | 2.8 |
|  | 253 | 1.8 | 4.1 | 4.6 |
|  | 273 | 3.2 | 3.8 | 5.3 |
|  | 293 | 3.1 | 3.1 | 4.8 |

Table 16 - Absolute average deviation (\%AAD) between the available experimental data and the studied models.

| Author | No Data | PR | PR+Peneloux | SAFT-VR Mie | GERG 2008 |
| :--- | :---: | :---: | :---: | :---: | :---: |
| Bailey et al. | 80 | 5.1 | 3.9 | 4.6 | 3.3 |
| Reamer et al. | 1140 | 10.2 | 5.5 | 5.0 | 5.3 |
| This work | 526 | 3.0 | 5.2 | 2.5 | 3.4 |
| Total | 1748 | 7.8 | 5.4 | 4.3 | 4.6 |

Table 17 - Regressed parameters for the calculation of the second virial coefficient $B$, condition ranges and AAD deviations between the correlation and experimental compressibility factors.

| Parameters | $\frac{\mathbf{a}\left(\mathbf{m}^{3} \cdot \mathbf{m o l}^{-1}\right)}{1.5385 \times 10^{-4}}$ | $b\left(\mathbf{m}^{3} \cdot K \cdot \mathrm{~mol}^{-1}\right)$ |  | $\mathrm{c}\left(\mathrm{m}^{3} \cdot \mathrm{~K}^{3} \cdot \mathrm{~mol}^{-1}\right)$ |  | d ( $\mathrm{m}^{\mathbf{3}} \cdot \mathrm{mol}^{-1}$ ) |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  | $-6.6201 \times 10^{-2}$ |  | $-1.5017 \times 10^{2}$ |  | $9.1010 \times 10^{-5}$ |
|  | $\mathrm{CH}_{4}$ mole fraction |  | Pressure (MPa) |  | Temperature (K) |  |
|  | 0.1-0.9 |  | $0.17-1.88$ |  | 253-500 |  |
|  | \%AAD |  |  | Max.Dev. |  |  |
|  | $1.5 \%$ |  |  | 5.7\% |  |  |



Figure 1. Predicted phases envelopes with the PR EoS of the methane + hydrogen sulphide system with $0.1101,0.1315,0.1803,0.248,0.286$ and $\mathbf{0 . 4 5 8} \mathbf{~ m o l}$ fractions of $\mathbf{H}_{2}$ S. The solid lines are the compositions studied in this work and the dashe lines are other phase diagrams compared against Kohn and Kurata data [22]: (■) $\mathbf{0 . 1 1 0 1}$, ( $\bullet$ ) $\mathbf{0 . 2 4 8}$ and ( $\mathbf{( 1 )} \mathbf{0 . 4 5 8} \mathbf{~ m o l}$ fractions of $\mathbf{H}_{2} S$.


Figure 2. Temperature-density coexistence envelop for pure $\mathbf{H}_{\mathbf{2}} \mathbf{S}$. Comparison between experimental data (ㅁ) [32] and SAFT-VR Mie calculation.


Figure 3. Predicted and experimental densities of pure $\mathbf{H}_{2} \mathrm{~S}$. The solid lines are isotherms calculated with the SAFT-VR Mie and the dashed line is the calculated coexistence curve. The symbols denote experimental data: from the literature [32] at $T=250 \mathrm{~K}(\Delta), T=300 \mathrm{~K}(\square), T=350 \mathrm{~K}(\circ), \mathrm{T}=400 \mathrm{~K}(\diamond)$ and $T=450 K(\times)$ and at saturation (■).


Figure 4. P -x diagrams of $\mathrm{CH}_{4}(\mathbf{1})+\mathrm{H}_{2} \mathrm{~S}(2)$ system at a) $\mathrm{T}=223.17 \mathrm{~K}$, b) $\mathrm{T}=273.54 \mathrm{~K}$ and c$) \mathrm{T}=310.93$. Symbols: ( $\square$ ) Coquelet et al. [7] and ( $\Delta$ ) Kohn and Kurata [22]. Solid line: calculated bubble and dew lines using PR model with $\mathbf{k}_{\mathrm{ij}}=\mathbf{0 . 0 8 0 7}$. Dashed line: calculated bubble and dew lines using SAFT-VR Mie EoS with $\mathrm{k}_{\mathrm{ij}}=\mathbf{0 . 0 3 1 4}$.


Figure 5. Experimental and predicted densities of the 0.714 mole $\mathrm{CH}_{\mathbf{2}}+\mathbf{0 . 2 8 6}$ mole $\mathbf{H}_{2} \mathrm{~S}$ system.
Experimental results: ( $\square \mathbf{2 5 3} \mathbf{~ K , ~ ( O ) ~} 273$ K, ( $\triangle$ ) 293 K. Lines: Predictions using the SAFT-VR Mie EoS. Dashed lines: Predictions using the GERG-2008 EoS. Dotted lines: Predictions using the PR EoS.


Figure 6. Experimental and predicted densities of the 0.5 mole $\mathbf{C H}_{\mathbf{4}} \boldsymbol{+} \mathbf{0 . 5} \mathbf{~ m o l e} \mathbf{H}_{\mathbf{2}} \mathrm{S}$ system. Experimental results: ( $\square$ ) $311 \mathrm{~K},(\triangle) 344 \mathrm{~K}$, ( O ) 377 K and $(\diamond) 411 \mathrm{~K}$ [8]. Lines: Predictions using the SAFT-VR Mie EoS. Dashed lines: Predictions using the GERG-2008 EoS. Dotted lines: Predictions using the PR+Peneloux EoS.


Figure 7. Experimental and predicted densities of the 0.7 mole $^{\mathbf{C H}_{\mathbf{4}}+\mathbf{0 . 3}} \mathbf{~ m o l e} \mathbf{H}_{\mathbf{2}} \mathrm{S}$ system. Comparison of PR+Peneloux (continuous curve), PR (dashes) and literature data at $T=311 \mathrm{~K}$ ( $\square$ ), $\mathrm{T}=\mathbf{3 4 4 K}$ ( $O$ ) and $\mathrm{T}=411 \mathrm{~K}(\triangle)$ [8].


Figure 8. Compressibility factor of the $0.8197 \mathrm{~mol} \mathrm{CH}_{\mathbf{4}} \mathbf{+ 0 . 1 8 0 3} \mathbf{~ m o l ~}_{\mathbf{H}}^{\mathbf{2}} \mathbf{S}$ system. Comparison betwen the SAFT-VR Mie calculation and experimental data measured in this work: $T=253 \mathrm{~K}$ ( $\square$ ), $\mathrm{T}=\mathbf{2 7 3 K}$ ( $O$ ) and T=293K ( $\triangle$ ).


Figure 9. Deviations between this work ( $\triangle$ ), Reamer et al. data ( $\square$ ) [8] and Bailey et al. data ( $O$ ) [10] and predictions using low pressure virial EoS from


Figure 10. $\mathrm{P}-\rho$ diagram of several systems of $\mathrm{H}_{2} \mathrm{~S}+\mathrm{CH}_{4}$ at 8 isotherms. Comparison of predictions with low pressure virial EoS and experimental data: $T=253 \mathrm{~K}$ and $\mathrm{x}_{\mathrm{CH} 4}=0.87(\triangle), \mathrm{T}=273 \mathrm{~K}$ and $\mathrm{x}_{\mathrm{CH} 4}=0.82$ ( $\square$ ), $T=293 \mathrm{~K}$ and $\mathrm{x}_{\mathrm{CH} 4}=0.71(O), T=344 \mathrm{~K}$ and $\mathrm{x}_{\mathrm{CH} 4}=0.4(*), T=377 \mathrm{~K}$ and $\mathrm{x}_{\mathrm{CH} 4}=0.5 \quad(-), T=411 \mathrm{~K}$ and $\mathrm{x}_{\mathrm{CH} 4}=0.6$ $(+), T=444 \mathrm{~K}$ and $\mathbf{x}_{\mathbf{C H} 4}=\mathbf{0 . 8}(\times)$ and $T=500 \mathrm{~K}$ and $\mathrm{x}_{\mathrm{CH} 4}=\mathbf{0 . 5}(\diamond)[8][10]$.

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