

Impact of hydrogen injection on thermophysical properties and measurement reliability in natural gas networks

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Abstract. In the context of the European decarbonization strategy, hydrogen is a key energy carrier in the medium to long term. The main advantages deriving from a greater penetration of hydrogen into the energy mix consist in its intrinsic characteristics of flexibility and integrability with alternative technologies for the production and consumption of energy. In particular, hydrogen allows to: i) decarbonise end uses, since it is a zero-emission energy carrier and can be produced with processes characterized by the absence of greenhouse gases emissions (e.g. water electrolysis); ii) help to balancing electricity grid supporting the integration of non-programmable renewable energy sources; iii) exploit the natural gas transmission and distribution networks as storage systems in overproduction periods. However, the hydrogen injection into the natural gas infrastructures directly influences thermophysical properties of the gas mixture itself, such as density, calorific value, Wobbe index, speed of sound, etc [1]. The change of the thermophysical properties of gaseous mixture, in turn, directly affects the end use service in terms of efficiency and safety as well as the metrological performance and reliability of the volume and gas quality measurement systems. In this paper, the authors present the results of a study about the impact of hydrogen injection on the properties of the natural gas mixture. In detail, the changes of the thermodynamic properties of the gaseous mixtures with different hydrogen content have been analysed. Moreover, the theoretical effects of the aforementioned variations on the accuracy of the compressibility factor measurement have been also assessed.

1 Introduction

The increasing penetration of non-programmable renewable energy sources (RES) (e.g. photovoltaic and wind) in electricity generation is impacting considerably on the power grid management system. Owing to the intermittent and random nature, power production from non-programmable RES generates critical issues in the balance between energy supply and demand, especially during overgeneration periods. Such issues can be tackled by employing large storage capacities, which are still not guaranteed by the storage systems installed on the electricity grid.

Currently, Power to Gas (PtG) represents the most suitable technology for storing excess electricity produced by non-programmable RES providing flexibility to the electrical system

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[2]. Exploiting the natural gas (NG) transmission and distribution networks, as well as the storage features of the existing NG infrastructure, the hydrogen produced from surplus renewable energy can be injected into the NG pipeline system allowing to store large amounts of energy on large-scale and for long-term period [3]. This will allow to decarbonize end-use sectors (e.g., buildings, transport, industry), since it is a zero-emission energy carrier, in addition to offer balancing and regulation services to the power grid. Blending hydrogen into the existing natural gas pipeline network is therefore a promising perspective for facilitating the development of an energy system based on a high share of non-programmable RES.

Nevertheless, the reliability of injection of hydrogen in natural gas (H₂NG) mixtures into existing infrastructures (e.g. pipelines, valves, compressor, boilers, measuring systems) is still a debated research topic. Indeed, increasing hydrogen content in NG lead to significant changes in thermo-physical properties of the resulting mixture. Stetsenko et al. [1] investigated how thermo-physical properties of different gas types of the "H" group of EN 437:2019 change with increasing hydrogen fraction. The authors have proved that all tested gases remain in group "H" when hydrogen is injected into natural gas with a content ranging from 2% to 23%. In detail, the properties of the mixtures vary as follows: i) the density (relative density) decreases in the range 1.7 – 20.5%; ii) the higher heating value decreases in the range 1.4 – 16%; iii) the Wobbe Index decreases in the range 0.49 – 5.7%; iv) the sound speed increases in the range 1 – 13.5%. The change of the thermodynamic properties leads to combustion and calorific characteristics changes. This could result in safety issues related to the volumetric percentage of hydrogen added (e.g. fugitive emissions are larger with hydrogen, flammability limits of hydrogen are wider than methane) [4]. Generally, only minor criticalities occur with blends of less than 5 – 15% hydrogen by volume, depending on site specific conditions and natural gas mixture compositions. Extensive studies and tests about the existing pipeline, valves, compressor, measuring systems and end-use appliances must be addressed for higher volume fractions of hydrogen in the range of 15 – 50% issues [5].

Several studies have investigated the H₂NG mixtures effects on the reliability of existing measurement systems. Stetsenko et al. [1] demonstrated that the injection of more than 10% hydrogen to natural gas allows to manage existing gas transportation and gas consumption systems without the need for reconstructions, modifications, changes in the algorithms for calculating physical parameters of the gas mixture and volumetric flow rate. On the other hands, further studies and the development of new algorithms are needed for higher hydrogen contents. In [6] no problems were recorded relating to the materials used in the construction of ultrasonic, membrane and turbine gas meters for hydrogen up to 30%. In particular, diaphragm, turbine and ultrasonic gas meters should be insensitive to this effect. However, they encourage field tests to determine the measurement accuracy of gas meters with H₂NG mixtures, as well as long-term metrological stability. Iskov et al. [7] analysed the medium-term effects (about one year) of 100% hydrogen on the metrological properties of traditional turbine, rotary piston and diaphragm gas meters at a pressure of 20 mbar, finding that hydrogen had no effect on the metrological characteristics of the gas meters tested, despite some issues with the turbine meter seal. Dehaeseleer [8] demonstrated that there are no operational issues with traditional gas meters (turbine, rotary piston and diaphragm) up to 10% of the hydrogen content in natural gas, however, above this value a new calibration could be necessary, together with the evaluation of changes to the design of the meters. Jaworski [9] experimentally evaluated the durability of diaphragm gas meters with hydrogen, demonstrating they are not significantly affected by injection of hydrogen up to 15%. The long-term drift has also been related to the wear of the internal parts of the meter, rather than to the hydrogen content in the gas mixture. In addition, no damage was found that could represent a threat to operational safety as all gas meters tested maintained unaltered tightness during and after the test. Jaworski and Dudek [10] found capillary thermal mass gas meters

comply with accuracy limits and only occasional values exceed the maximum permissible errors (MPE) for H2NG with 2, 4 and 5% of hydrogen; on the other hand, with hydrogen content of 10 and 15% they significantly exceeded the MPEs with error curves systematically shifted towards negative errors. No experimental evidences are available in the literature against the injection of H₂ up to 10% for ultrasonic meters, whereas for higher levels further experimental research is required [11].

Finally, as regards the effects on the volume conversion, Łach [12] investigated the effects of H2NG on the calculation of the compressibility factor. To this end, tests were carried out in the laboratory of the Polish National Research Oil and Gas Institute to compare SGERG-88 and AGA8-DC92 methods to determine the compressibility coefficient of gas with an increasing hydrogen content. In fact, both methods have application limitations in terms of hydrogen content in natural gas up to 10% (mol/mol). Research has shown that the AGA8-DC92 method provides satisfactory results when the hydrogen content is less than 40% (mol/mol).

This paper is aimed at presenting the results of a comprehensive study on the effects of hydrogen injection on the thermodynamic properties of the natural gas mixture. The main thermo-physical parameters of NG mixtures, including density, relative density, higher heating value, Wobbe Index, specific heat capacity and sound speed have been investigated. Furthermore, in order to assess the reliability of measurement systems installed on existing gas infrastructures, the impact of hydrogen injection on the accuracy of the compressibility factor measurement have been evaluated.

2 Methods

2.1 Calculation of the thermodynamic properties

The density of the real gaseous mixture at metering reference conditions is obtained using the ISO 6976:2016 [13] formula:

$$D(p, t) = \frac{D^0}{Z(p, t)} \quad (1)$$

where: i) $Z(p, t)$ is the compressibility factor of the gaseous mixture calculated in accordance with the ISO 6976:2016 [13]; ii) D^0 is the density of the ideal gas at the metering reference conditions evaluated through the following equation:

$$D^0(p, t) = \frac{M}{V^0} \quad (2)$$

where: i) V^0 is the ideal molar volume of the mixture calculated according to the ideal gas law; ii) M is the molar mass of the gas mixture calculated from:

$$M = \sum_{j=1}^N x_j M_j \quad (3)$$

where: i) x_j is the mole fraction of the j -th component in the gas mixture; ii) M_j is the molar mass of the j -th component.

The relative density is defined as the ratio of the density of the gas mixture to the density of dry air of reference composition at the same specified conditions of pressure and temperature.

It has been calculated according to the ISO 6976:2016 formula for the real gaseous mixture [13]:

$$G(t, p) = \frac{G^0 \cdot Z_{air}(t, p)}{Z(t, p)} \quad (4)$$

where: i) $Z(p, t)$ is the compressibility factor of the gaseous mixture calculated in accordance with the ISO 6976:2016 [13]; ii) $Z_{air}(p, t)$ is the compressibility factor of dry air of reference composition; iii) G^0 is the relative density of the ideal gas, which is calculated through the following formula:

$$G^0 = \sum_{j=1}^N x_j \frac{M_j}{M_{air}} \quad (5)$$

where: i) x_j is the mole fraction of the j-th component in the gas mixture; ii) M_j is the molar mass of the j-th component; iii) M_{air} is the molar mass of dry air of reference composition.

The calculation of the ideal higher heating value $(Hv)_G^0$ on volume basis, at a temperature t_1 , has been carried out basing on the ISO 6976:2016 standard [13] through the following equation:

$$(Hv)_G^0(t, p) = \frac{(Hc)_G^0(t)}{V^0} \quad (6)$$

where: i) $(Hc)_G^0(t)$ is the ideal higher molar-basis calorific value; ii) V^0 is the ideal molar volume of the mixture calculated according to the ideal gas law.

The Wobbe Index is defined in accordance with ISO 6976:2016 [13] as the ratio of the higher heating value, at specified reference condition, to the square root of the relative density:

$$W_G^0(t, p) = \frac{(Hv)_G^0(t, p)}{\sqrt{G^0}} \quad (7)$$

where: i) $(Hv)_G^0$ is the higher heating value on volume basis of the ideal mixture; ii) G^0 is the relative density the ideal mixture.

A crucial parameter for ultrasonic gas meters accuracy is represented by the speed of sound, which can be estimated through the AGA 10 formula [14]:

$$u = 18.591 \left(\frac{T \cdot Z \cdot k}{\rho_s} \right)^{0.5} \quad (8)$$

where: i) T is the absolute temperature; ii) Z is the compressibility factor of the gaseous mixture calculated employing the ISO 6976:2016 standard [13]; iii) k is the adiabatic coefficient of gas mixture (i.e. the ratio between heat capacities at constant pressure and volume, respectively); iv) ρ_s is the density of the gas mixture at reference conditions.

On the other hand, the product $D \cdot c_p$ strongly affects thermal mass gas meters. The correlations depending on temperature and gaseous mixture composition available in literature can be employed estimate to the specific heat capacity variation as a function of the hydrogen content. The specific heat capacity at constant pressure has been evaluated as the

weighted average of the specific heat capacities at constant pressure of the components constituting the gaseous mixture:

$$c_p(t_1) = \sum_{j=1}^N x_j \cdot c_{p,j} \tag{9}$$

where: i) x_j is the mass fraction of the j -th component in the gas mixture; ii) $c_{p,j}$ is the specific heat capacity at constant pressure of the j -th component calculated according to the equation in [15]:

$$c_{p,j}(T) = A_j + B_j T + C_j T^2 + D_j T^3 + E_j T^4 \tag{10}$$

where i) T is the absolute temperature; ii) A_j, B_j, C_j, D_j, E_j are the regression coefficients for the j -th component of the gaseous mixture (see Table 1).

Table 1. Regression coefficients for chemical compound [16].

Component	Temperature range [K]	A	B	C	D	E
Methane, CH ₄	50-1500	34.942	-3.9957E-02	1.9184E-04	-1.5303E-07	3.9321E-11
Ethane, C ₂ H ₆	100-1500	28.146	4.3447E-02	1.8946E-04	-1.9082E-07	5.3349E-11
Propane, C ₃ H ₈	200-1000	31.986	4.27E-02	5.00E-04	-6.56E-07	2.56E-10
n-Butane, n-C ₄ H ₁₀	200-1500	20.056	2.8153E-01	-1.3143E-05	-9.4571E-08	3.4149E-11
iso-Butane, iso-C ₄ H ₁₀	200-1500	6.772	3.1447E-01	-1.0271E-04	-3.6849E-08	2.0429E-11
n-Pentane, n-C ₅ H ₁₂	200-1500	26.671	3.2324E-01	4.2820E-05	-1.6639E-07	5.6036E-11
iso-Pentane, iso-C ₅ H ₁₂	200-1000	16.288	3.1754E-01	2.0237E-04	-4.3027E-07	1.8001E-10
n-Hexane, C ₆ H ₁₄	200-1500	25.924	4.1927E-01	-1.2491E-05	-1.5592E-07	5.8784E-11
Nitrogen, N ₂	50-1500	29.342	-3.5395E-03	1.0076E-05	-4.3116E-09	2.5935E-13
Carbon dioxide, CO ₂	50-5000	27.437	4.2315E-02	-1.9555E-05	3.9968E-09	-2.9872E-13
Hydrogen, H ₂	250-1500	25.399	2.0178E-02	-3.8549E-05	3.1880E-08	-8.7585E-12

2.2 Calculation of the compressibility factor

The compressibility factor at metering conditions, Z , has been estimated basing on the ISO 12213-2:2010 [17]. The latter uses the AGA8-DC92 equation, developed by Starling e Savidge in 1992 [18]. The equation is an extended virial-type formulation and, which is only capable to calculate the properties in the gas phase. Moreover, it employs a detailed molar-composition in which all constituents present in amounts exceeding a mole fraction of 0.00005 should be represented. It may be written as:

$$Z = 1 + B\rho_m - \rho_r \sum_{n=13}^{18} C_n^* + \sum_{n=13}^{58} C_n^*(b_n - c_n k_n \rho_r^{k_n}) \rho_r^{k_n} \exp(-c_n \rho_r^{k_n}) \tag{14}$$

where: i) B is the second virial coefficient; ii) ρ_m is the molar density; iii) ρ_r is the reduced density; iv) b_n, c_n, k_n are constants; v) C_n^* are the coefficient which depend on the temperature and NG mixture composition. The reduced density ρ_r is correlated to the molar density ρ_m through the below relation:

$$\rho_r = K^3 \rho_m \tag{15}$$

where: i) K is a mixture size coefficient; ii) ρ_m is the molar density calculated using the following equation:

$$\rho_m = \frac{p}{ZRT} \tag{16}$$

where: i) p is the absolute pressure; ii) R is the universal gas constant; iii) T is the absolute temperature.

The calculation method of compressibility factor is critical in NG industries especially for large flow metering plants [19]. In order to assess its reliability, the calculation of the compressibility factor has been carried out also using the ISO 12213-3, AGA NX 19 and AGA NX 19 Mod. standards, which are still used in NG industries.

3 Results and discussion

3.1 Analysis of the thermodynamic properties of H2NG mixtures

Aimed at evaluating the effects of hydrogen injection in natural gas mixtures, the authors investigated 5 NG mixtures (i.e. the ones without presence of H₂) used as example calculations in Annex C of ISO 12213-2:2010 [17], as described in Table 2.

Table 2. Molar composition of the investigated gas mixtures.

Substance	GAS 1	GAS 2	GAS 3	GAS 5	GAS 6
Methane, CH ₄	96.50	90.70	85.90	81.20	82.60
Ethane, C ₂ H ₆	1.80	4.50	8.50	4.30	3.50
Propane, C ₃ H ₈	0.45	0.84	2.30	0.90	0.75
n-Butane, n-C ₄ H ₁₀	0.10	0.15	0.35	0.15	0.12
iso-Butane, iso-C ₄ H ₁₀	0.10	0.10	0.35	0.15	0.12
n-Pentane, n-C ₅ H ₁₂	0.03	0.04	0.05	0.00	0.04
iso-Pentane, iso-C ₅ H ₁₂	0.05	0.03	0.05	0.00	0.04
n-Hexane, C ₆ H ₁₄	0.07	0.04	0.00	0.00	0.02
Nitrogen, N ₂	0.30	3.10	1.00	5.70	11.70
Carbon dioxide, CO ₂	0.60	0.50	1.50	7.60	1.10

In Figure 1 the results of the analyses have been graphically reported. Where available, in the graphs the dashed lines represent the allowable limit according to the related applicable standards and national regulations as below reported:

- relative density and higher heating value [17];
- Wobbe Index [20];
- speed of sound for ultrasonic domestic gas meters [21];
- D·c_p product for thermal mass gas meters [22].

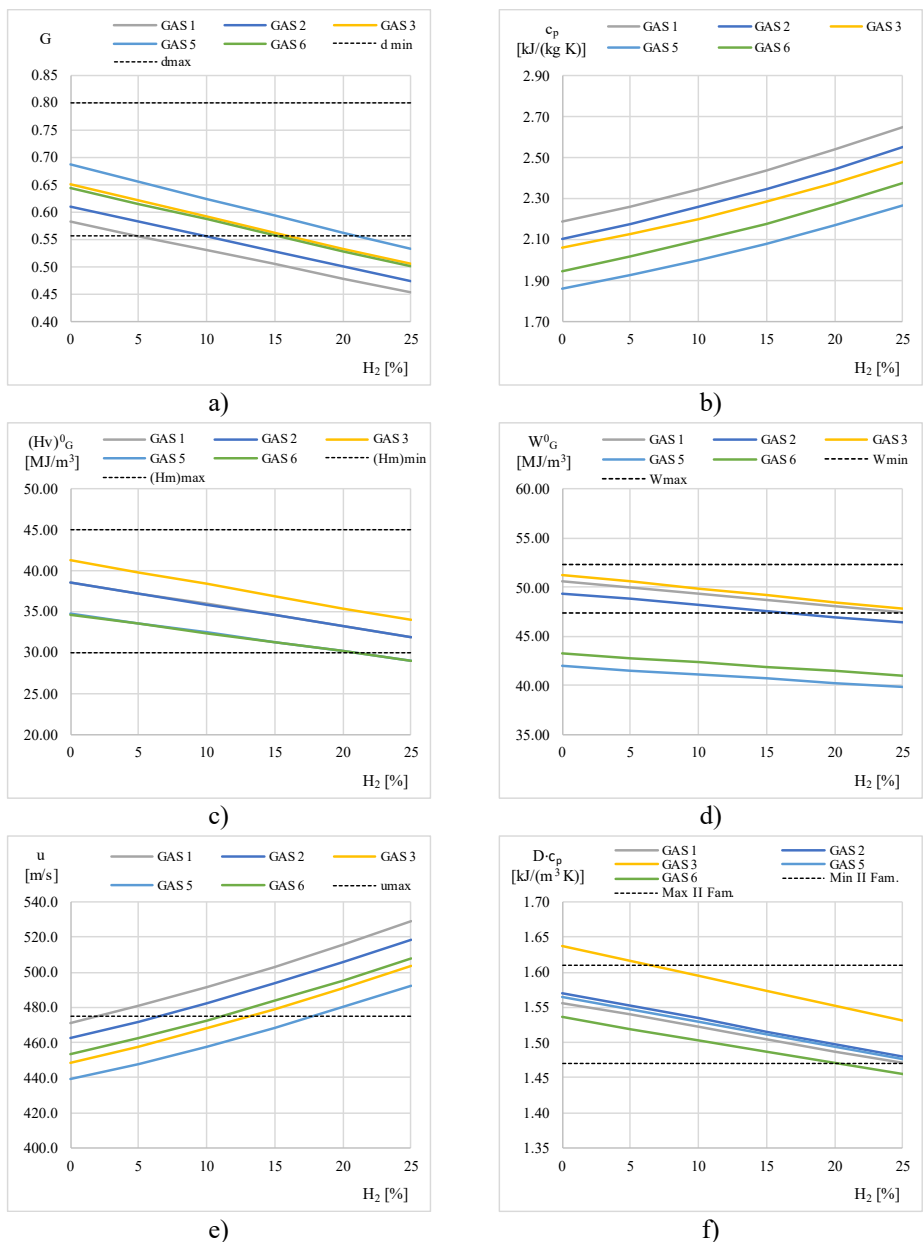


Fig. 1. Trend of thermophysical properties of H₂NG mixtures as a function of the H₂ content: a) relative density, b) specific heat capacity, c) higher heating value, d) Wobbe Index, e) speed of sound and f) D·c_p product.

From the above reported results, it can be highlighted that:

- the relative density of the H₂NG mixture decreases as the H₂ content increases leading to values lower than the corresponding limit EN ISO 12213-2 at about 10% H₂;
- the heat capacity at constant pressure significantly increases as the hydrogen content increases (up to +21.8% for Gas 6 at x_{H₂}=25%);

- the higher heating value per volume significantly decreases as the hydrogen content increases (up to -17.8% for Gas 3 at $x_{H_2}=25\%$) and for investigated gases 5 and 6 it becomes lower than the corresponding limit of EN ISO 12213-2 at $x_{H_2}=25\%$);
- the Wobbe Index smoothly decreases as the H_2 content increase (up to - 6.6% $x_{H_2}=25\%$);
- the speed of sound increases up to 12.3% and it becomes tendentially higher than the accepted limit of 475 ms^{-1} of the ISO 14236 standard for ultrasonic gas meters even at low H_2 contents;
- the $D \cdot c_p$ product decreases as the H_2 content increases (up to - 6.7% for Gas 3); this can lead to unavoidable drift of the error curve of thermal mass gas meters unless an effective gas recognition system is present with the consequent need for design changes.

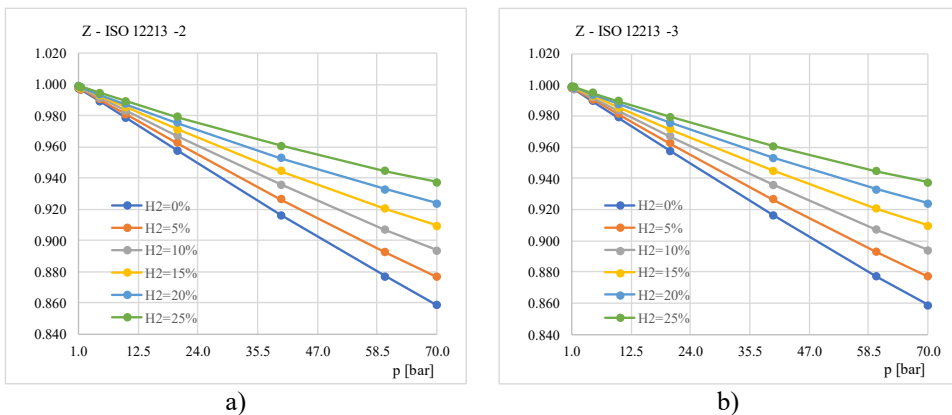
3.2 Analysis of the compressibility factor of H2NG mixtures

The effects of hydrogen injection in natural gas mixtures have been also evaluated in terms of accuracy of the compressibility factor measurement. To this aim, a typical NG mixture distributed in Italy has been considered (see Table 3).

Table 3. Molar composition of the investigated gas mixture.

Component	(% mol/mol)
Methane, CH_4	95.99
Ethane, C_2H_6	2.292
Propane, C_3H_8	0.639
n-Butane, n- C_4H_{10}	0.097
iso-Butane, iso- C_4H_{10}	0.102
n-Pentane, n- C_5H_{12}	0.012
iso-Pentane, iso- C_5H_{12}	0.018
n-Hexane, C_6H_{14}	0.012
Nitrogen, N_2	0.650
Carbon dioxide, CO_2	0.188

Figure 2 shows the effects of the increase of H_2 content up to 25% in the NG mixture on the compressibility factor Z , calculated employing the ISO 12213-2, ISO 12213-3, AGA NX 19 and AGA NX 19 Mod algorithms. Calculations have been performed at different gas pressures considering a constant gas temperature $T=15\text{ }^\circ\text{C}$.



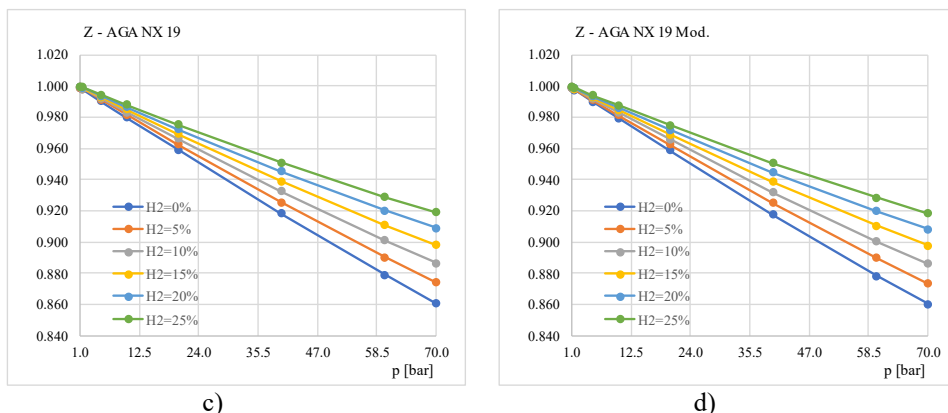


Fig. 2. Trend of compressibility factor, Z , as a function of the H_2 content: a) ISO 12213-2, b) ISO 12213-3, c) AGA NX 19 and d) AGA NX 19 Mod..

From the obtained results it can be observed that the compressibility factor increases as H_2 content increases. Furthermore, at high pressures H_2 content affects the compressibility factor more than at low pressures (e.g. using the ISO 12213-2 standard an increase in the range 0.10 – 0.44% at 5 bar and 2.1 – 9.1% at 70 bar have been found). Moreover, the obtained results show that pressure more significantly influence the compressibility factor at low H_2 concentrations.

Finally, the compressibility factor decreases as pressure increases. As for example, a decrease of Z has been found in the range 0.25 – 14% at $H_2 = 0\%$ together with at $H_2 = 25\%$ a lower decrease in the range 0.05 – 0.2% at $H_2 = 25\%$.

4 Conclusions

The reliability of hydrogen and natural gas mixtures in existing infrastructures (e.g. pipelines, valves, compressor, measuring systems) is a crucial issue the current energy transition in European Union. Injection of hydrogen into natural gas networks, in fact, directly affects thermodynamic properties of the gas itself. As a consequence, changes in the metrological performance especially in terms of volume and gas quality measurements are expected.

In the present paper the change of the main thermo-physical parameters of H_2NG , including density, relative density, higher heat value, Wobbe Index, specific heat capacity and sound speed have been analysed when H_2 content in the mixture varies up to 25%.

The obtained results show that H_2 injection in the NG mixture significantly affects the relative density, heat capacity and higher heating value, whereas the Wobbe Index is smoothly influenced. In particular, the relative density decreases as H_2 content increases and for GAS 6 just at 10 % H_2 sufficient the corresponding lower limit is exceeded. Similarly, the higher heating value significantly decreases as the H_2 content increases. As for example, for gases characterized by a low content of CH_4 , the higher heating value decreases up to 20% at 25% H_2 exceeding the lower limit of EN ISO 12213-2.

Furthermore, the H_2 concentration considerably affects both the sound speed and $D \cdot c_p$ product. In particular, the speed of sound increases up to 12.3% becoming tendentially higher than the accepted limit of 475 ms^{-1} of the ISO 14236 standard for ultrasonic gas meters starting from $H_2 = 5\%$. On the other hand, the $D \cdot c_p$ product decreases up to 6.7% for Gas 3 as the H_2 concentration increases.

The compressibility factor trend as a function of H₂ content has been also investigated basing on the ISO 12213-2, ISO 12213-3, AGA NX 19 and AGA NX 19 Mod. algorithms. The obtained results show the compressibility factor at high metering pressures is higher affected by H₂ content than at low pressures. In fact, the compressibility factor evaluated using the ISO 12213-2 standard increases as H₂ content increases in the range 0.10 – 0.44% at 5 bar, whereas it increases in the range 2.1 – 9.1% at 70 bar. Furthermore, gas pressure shows a more significant effect on the compressibility factor at low H₂ concentrations.

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