Exploration of Hydrogen Influence on Physical Properties of Natural Gas and Metrological Characteristics of Its Metering Systems

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Abstract

Due to the predicted exhaustion of oil and natural gas resources by the end of this century, all industrialized countries are taking steps to find alternatives for hydrocarbon fuels. Nuclear energy is expected to be the main source of electricity production by the end of the XXI century. Electricity, however, has some disadvantages related to storage and losses in case of long-distance transmission. Among other sources of energy, the use of which can be possible in the nearest future, hydrogen is the most promising one. The advantages of using hydrogen as a fuel can be summarized as follows: using hydrogen as fuel can be one of the comprehensive solutions to the problem of environmental protection; hydrogen is an excellent energy carrier; as of today, society is largely dependent on fossil energy, while the contribution of sustainable fuel to global energy demand is limited.
Taking into consideration the above, and the possibility of using a mixture of hydrogen and natural gas as a source of energy, the research has been aimed in the following directions:
- Influence of hydrogen impurities on the physical properties of natural gas;
- the effect of adding hydrogen into natural gas on the metrological characteristics of metering systems.

1. Statement of the problem

To determine the effect of hydrogen on the above list of factors, research is aimed at solving the following problems:

1. Conducting studies on the effect of hydrogen injection in natural gas on the physical properties of the mixture requires determining the dependence of the following gas parameters on the percentage of hydrogen in the natural gas: density, Reynolds number, dynamic viscosity, compressibility factor, high heat value, Wobbe number, sound speed, adiabatic exponent.

2. Determining the permissible fraction (permissible concentration) of hydrogen in natural gas in modern gas-transport and gas-consuming systems.

3. Examining the effect of hydrogen injection on metrological characteristics of measuring equipment and gas custody transfer metering systems in the following areas:
- research and assessment of the possibility of using contemporary gas meters to measure the volumetric flow rate and volume of hydrogen and natural gas mixture;
- study of the effect of hydrogen injection on the results of measuring the volumetric flow rate and volume adjusted to basic conditions. Evaluation of the possibility of using existing flow computers and gas correctors to adjust the measured volumetric flow rates and mixture volumes to basic conditions;
- the influence of hydrogen injection on the calorific value of the mixture within the framework of the transition of gas metering systems of power plant units.

2. The main material

2.1 Conducting research on the effect of injecting hydrogen into natural gas on the physical properties of the mixture

Various samples of natural gas with different component mixtures were prepared to perform objective research and modeling.

Analysis of the following physical characteristics of these gas compositions and calculations of their physical criterion were carried out:
- adiabatic index and speed of sound;

- density;
- compressibility factor;
- higher heat of combustion;
- Wobbe index.

The calculation of the adiabatic index (the ratio of the relative change in pressure to the relative change in density without heat exchange with the environment) of gases was carried out according to the Kobza formula [1] for the component composition and density of each component at base conditions:

$$k = 1{,}556(1 + 0{,}074x_a) - 3{,}9 \cdot 10^{-4} T(1 - 0{,}68x_a) - \\ - 0{,}208\rho_b + + (p/T)^{1{,}43}\left[384(1 - x_a)(p/T)^{0{,}8} + 26{,}4x_a\right] \quad (1)$$

where: x_a - nitrogen content in gas;
p - pressure, MPa;
T - absolute temperature, K;
ρ_b - density of natural gas at base conditions, kg/m^3.

The speed of sound in natural gas is determined by the formula AGA 10 [2]:

$$u = 18{,}591\left(\frac{T \cdot Z \cdot K}{\rho_b}\right)^{0{,}5} \quad (2)$$

where: ρ_b - density of natural gas at base conditions, kg/m^3;
T - absolute temperature, K;
k - adiabatic index;
Z - compressibility factor.

The compressibility factor (the ratio of the actual volume of real gas at a certain pressure and temperature to the volume of an ideal gas under the same conditions) is calculated by the formula (3) ISO 6976:1995/Cor.2:1997, Cor.3:1999 [3]:

$$z_{mix}(t_2, p_2) = 1 - \left[\sum_{j=1}^{N} x_j \cdot \sqrt{b_j}\right]^2 \quad (3)$$

where: x_j - the content of the j-th component in the gas;
$\sqrt{b_j}$ - summation of the coefficient for j-th component of natural gas.

The relative density (the ratio of the density of the gas to the density of dry air of standard composition at the same pressure and temperature) is calculated by the formula (14) ISO 6976:1995/Cor.2:1997, Cor.3:1999 [3]:

$$d(t, p) = \frac{d^\circ \cdot Z_{air}(t, p)}{Z_{mix}(t, p)} \quad (4)$$

where: d° - the relative density of an ideal gas;
$Z_{air}(t, p)$ - compressibility factor of dry air of standard composition;
$Z_{mix}(t, p)$ - gas compressibility factor.

The relative density of an ideal gas is calculated by the formula (11) ISO 6976:1995/Cor.2:1997, Cor.3:1999 [3]:

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

where: x_j - the content of the j-th component in the gas;
M_j - the molar mass of the j-th component;
M_{air} - molar mass of dry air compressibility standard composition.

The density is calculated by the formula (15) ISO 6976:1995/Cor.2:1997, Cor.3:1999 [3]:

$$\rho(t, p) = \frac{\rho^\circ(t, p)}{Z_{mix}(t, p)} \quad (6)$$

where: $\rho^\circ(t, p)$ - ideal gas density;
$Z_{mix}(t, p)$ - gas compressibility factor.

The ideal gas density is calculated by the formula (11) ISO 6976:1995/Cor.2:1997, Cor.3:1999:

$$\rho^\circ(t, p) = \left(\frac{p}{RT}\right)\sum_{j=1}^{N} x_j \cdot M_j \quad (7)$$

where: x_j - the content of the j-th component in the gas;
M_j - the molar mass of the j-th component;
p - pressure, MPa;
T - absolute temperature, K;

R - universal gas constant, equal to 8,314510 J·mol^{-1}·K^{-1}.

The determination of the values of the lowest and highest calorific values of gas is carried out in accordance with the formula (10) ISO 6976:1995/Cor.2:1997, Cor.3:1999:

$$\overline{H}[t_1, V(t_2, p_2)] = \frac{\overline{H}°(t_1)}{Z_{mix}(t_2, p_2)} \qquad (8)$$

where: $\overline{H}°(t_1)$ - the value of the ideal heat of combustion (higher or lower);
$Z_{mix}(t_2, p_2)$ - gas compressibility factor under base conditions.

The determination of the values of the lowest and highest ideal heat of combustion of a gas is made in accordance with the formula (4) ISO 6976:1995/Cor.2:1997, Cor.3:1999:

$$\overline{H}°(t_1) = \sum_{j=1}^{N} x_j \cdot \overline{H}_j^{\circ}(t_1) \qquad (9)$$

where: x_j - the content of the j-th component in the gas;
$\overline{H}_j^{\circ}(t_1)$ - the value of the ideal heat of combustion of the j-th component (higher or lower).

The calculation of the Wobbe number was performed according to the formula (16) ISO 6976:1995/Cor.2:1997, Cor.3:1999 [3]:

$$W[t_1, V(t_2, p_2)] = \frac{\overline{H}_S[t_1, V(t_2, p_2)]}{\sqrt{d(t_2, p_2)}} \qquad (10)$$

where: $\overline{H}_S[t_1, V(t_2, p_2)]$ - the value of the highest calorific value;
d - relative density.

The results of the analysis of the physical parameters of the four natural gas samples are shown in Tables 1 - 4.

Table 1

Hydrogen	0	2	10	23
Methane	93.031	91.170	83.728	71.634
Ethane	3.541	3.470	3.187	2.727
Propane	0.835	0.818	0.752	0.643
i - Butane	0.105	0.103	0.095	0.081
n - Butane	0.136	0.133	0.122	0.105
neo - Pentane	0.013	0.013	0.012	0.010
i - Pentane	0.029	0.028	0.026	0.022
n - Pentane	0.023	0.023	0.021	0.018
Hexan	0.008	0.008	0.007	0.006
Nitrogen	1.125	1.103	1.013	0.866
Carbon Dioxide	1.149	1.126	1.034	0.885
Oxygen	0.008	0.008	0.007	0.006
Calorific value MJ/m3	38.650	38.116	35.980	32.516
Shift, %	-	-1.38	-6.91	-15.87
Wobbe index MJ/m3	49.8571	49.611	48.623	47.015
Shift, %	-	-0.49	-2.48	-5.70

Table 2

Hydrogen	0	2	10	23
Methane	90.779	88.963	81.701	69.900
Ethane	4.555	4.464	4.100	3.507
Propane	1.056	1.035	0.950	0.813
i - Butane	0.110	0.108	0.099	0.085
n - Butane	0.174	0.171	0.157	0.134
neo - Pentane	0.002	0.002	0.002	0.002
i - Pentane	0.045	0.044	0.041	0.035
n - Pentane	0.038	0.037	0.034	0.029
Hexan	0.034	0.033	0.031	0.026
Nitrogen	1.519	1.489	1.367	1.170
Carbon Dioxide	1.684	1.650	1.516	1.297
Oxygen	0.008	0.008	0.007	0.006
Calorific value MJ/m3	38.811	38.273	36.124	32.638
Shift, %	-	-1.39	-6.92	-15.90
Wobbe index MJ/m3	49.428	49.184	48.205	46.613
Shift, %	-	-0.49	-2.47	-5.70

Table 3

Hydrogen	0	2	10	23
Methane	89.425	87.637	80.483	68.857
Ethane	5.179	5.075	4.661	3.988
Propane	1.191	1.167	1.072	0.917
i - Butane	0.115	0.113	0.104	0.089
n - Butane	0.185	0.181	0.167	0.142
neo - Pentane	0.003	0.003	0.003	0.002
i - Pentane	0.047	0.046	0.042	0.036
n - Pentane	0.040	0.039	0.036	0.031
Hexan	0.054	0.053	0.049	0.042
Nitrogen	1.646	1.613	1.481	1.267
Carbon Dioxide	2.111	2.069	1.900	1.625
Oxygen	0.007	0.007	0.006	0.005
Calorific value MJ/m3	38.904	38.364	36.208	32.709
Shift, %	-	-1.39	-6.93	-15.92
Wobbe index MJ/m3	49.156	48.913	47.940	46.359
Shift, %	-	-0.49	-2.47	-5.69

Table 4

Hydrogen	0	2	10	23
Methane	89.213	87.428	80.291	68.694
Ethane	5.114	5.012	4.603	3.938
Propane	1.163	1.140	1.047	0.895
i - Butane	0.118	0.115	0.106	0.091
n - Butane	0.209	0.205	0.188	0.161
neo - Pentane	0.003	0.003	0.003	0.002
i - Pentane	0.059	0.058	0.053	0.046
n - Pentane	0.047	0.046	0.042	0.036
Hexan	0.05	0.049	0.045	0.039
Nitrogen	1.509	1.479	1.358	1.162
Carbon Dioxide	2.510	2.460	2.259	1.933
Oxygen	0.0055	0.005	0.005	0.004
Calorific value MJ/m3	38.809	38.271	36.122	32.636
Shift, %	-	-1.39	-6.92	-15.91
Wobbe index MJ/m3	48.905	48.665	47.701	46.135
Shift, %	-	-0.49	-2.46	-5.66

The results of calculations allow to draw conclusions that when adding hydrogen to the natural gas in an amount from 2 % to 23 % physical parameters of the obtained mixture vary accordingly in the following ranges:
- the speed of sound increases by (1 - 13.5) %;
- density (relative density) of the mixture is reduced by (1,7 - 20,5) %;
- compressibility factor at atmospheric pressure varies within the limits (0,01 - 0,1) % at atmospheric pressure;
- the high heat value decreases by approximately (1.4 - 16) %;
- the Wobbe index decreases in the range (0.49 - 5.7) %.

2.2 Finding out the permissible rate (concentration) of hydrogen in natural gas in existing gas transmission and gas consuming facilities.

Determination of the permissible fraction (permissible concentration) of hydrogen in natural gas was performed to satisfy the following requirements and limitations:
- the possibility of using the mixture in existing gas transmission and gas-consuming systems without additional reconstructions, modifications;
- requirements and limitations of regulatory and metrological documentation;
- safety and explosion protection requirements.

Analysis of regulatory documentation and modeling showed the following:
a) According to the European standard EN 437 Tested gases - Test pressures - Appliance categories [4], natural gas is classified into “H” and “L” groups depending on the Wobbe index values. Simulations showed that with the addition of hydrogen in a volume of up to 23%, all four samples of natural gas remained in the “H” group;
b) In accordance with the requirements of the international standard ISO 12213-3: 2006 [5], restrictions are imposed on the use of natural gas for domestic purposes according to the following parameters:
- absolute pressure (from 0 to 12 MPa);
- the molar fraction of hydrogen (from 0 to 10%);
- relative density (from 0.55 to 0.8).

The simulation results carried out in the first section showed that, by the criterion of permissible values of the relative density, the fraction of hydrogen in natural gas should be in the range from 0 to 10%. Further increase in the proportion of hydrogen (up to 23%) depends on the quality (Wobbe number) of natural gas and requires the search and use of other algorithms for calculating the physical parameters of gas (described by other regulatory documents).

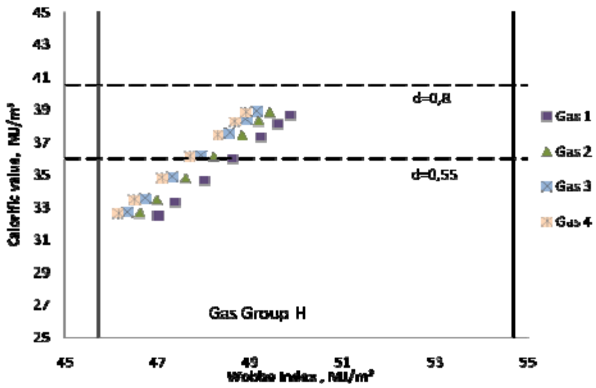

Figure 1

c) The algorithms for calculating the compressibility factor carried out according to GERG 88, also introduce a limit on the proportion of hydrogen in natural gas not more than 10%.

d) Analysis of the hydrogen explosion hazard showed that hydrogen ignites at a concentration of 4 to 74.5% (methane ignites at concentrations of 2.1 to 9.5%), although explosive concentrations for hydrogen are much higher than for natural gas (between 18.3 and 59% in concentration with air for hydrogen, compared with 6.3-14% for methane). This means that adding up to 59% of hydrogen to natural gas does not require changing the degree of explosion-proof, therefore, does not require additional modifications and certification of existing ex-proof equipment.

2.3 Analysis of the effect of hydrogen injection on the metrological characteristics of measuring instruments and gas metering systems

Currently, the most frequent systems (facilities, components) of fiscal metering of gas consumption include: a gas meter, a gas volume corrector (flow computer), pressure and temperature transducers, a measuring pipeline with straight pipe runs before and after the gas meter, communication lines between component parts of the system (complex, node) and auxiliary devices.

From the above list, only the gas meter and the volume corrector (calculator) are sensitive to hydrogen injections. Therefore, the study of the effect of the addition of hydrogen on the metrological characteristics of measuring instruments and systems for the fiscal accounting of natural gas was conducted in the following directions:
- research and evaluation of the possibility of using existing (ultrasonic) gas meters to account for the volumetric flow rate and the volume of the mixture of hydrogen and natural gas;
- learning the influence of hydrogen additives on the results of taking into consideration the volumetric flow rate and the volume reduced to base conditions. Evaluation of the possibility of using existing gas calculators and proofreaders to bring measured volumetric flow rates and mixture volumes to basic conditions;
- studying the influence of hydrogen additives on the heat of combustion of the mixture within the framework of the transition of gas metering systems to energy units.

Estimating the possibility of the use of existing gas meters was carried out only for measuring devices based on ultrasonic technology, whose functioning is based on measuring the propagation time of acoustic waves in the current medium. The main physical parameters of gas that affect the measurement results of meters are its density and acoustic velocity.

Analysis of the simulation results showed that when hydrogen is added to natural gas, the specified parameters change within the permissible limits, at which the metrological characteristics of the meter remain unchanged.

The calculation of the volume of gas in basic conditions on the values of volume in the measurement conditions is performed by the formula:

$$V_b = \frac{T_b \cdot P_m \cdot V_m \cdot Z_m}{T_m \cdot P_b \cdot Z_b} \quad (11)$$

where: V_m - volume in measurement conditions, m^3/h;
T_b - temperature at base conditions, K;
T_m - temperature in measurement conditions, K;
P_b - base pressure, MPa;
P_m - measurement pressure, MPa;
Z_b - compressibility factor under base conditions;
Z_m - compressibility factor under measurement conditions.

The proper reduction of volume (volumetric flow rate) of a mixture of natural gas and hydrogen and the influence of the compressibility factor to the percentage of hydrogen is of great importance.

The simulation results showed a slight change (from 0.01% to 0.1%) of the compressibility factor when hydrogen is added to 23% at atmospheric pressure.
According to ISO 12213-3: 2006 [5], the expected uncertainty of the compressibility factor with hydrogen content up to 10% does not exceed 0.1% for pressures up to 10 MPa and 0.2% for pressures from 10 to 12 MPa.

The evaluation of the changeability of the calculation of compression index factors outside the limits of factors specified in ISO 12213-3: 2006 [5] requires additional calculations and research.

Taking into consideration current trends in the transition of gas metering systems in power plants, i.e., switching to natural gas metering based on its energy value (caloric content), the effect of hydrogen additives on the heat of combustion of the mixture was analyzed.

The results of the analysis of the calculations showed that with the addition of hydrogen up to 10%, the higher heat of combustion of the mixture decreases by about 7%, while the Wobbe index decreases by no more than 3%.

This allows us to conclude about the possibility of adding hydrogen to natural gas in the amount of up to 10% without any modifications of both gas transmission and gas-consuming systems and algorithms for calculating the physical parameters of gas.

3. Conclusion

Adding hydrogen to natural gas in a volume of up to 10% allows to operate existing gas-transport and gas-consumption systems without any reconstructions, modifications, changes in algorithms for calculating the physical parameters of the gas mixture and (volume) volumetric flow rate for custody transfer purposes.

Adding hydrogen in the volume of more than 10%, and also, the use of the mixture at high pressures requires additional study and the development of new algorithms.

References

[1] Kobza Z., Dobrovolska B., Gontarek J. (Polish Higher Engineering School). Analysis of the impact of the sources of adiabatic index determination on the error in calculating the flow rate.

[2] AGA REPORT NO. 10, Speed of sound in natural gas and other related hydrocarbon gases

[3] ISO 6976:1995/Cor. 2:1997, Cor. 3:1999 Natural gas - Calculation of calorific values, density, relative density and Wobbe index from composition

[4] EN 437:2018 Test gases – Test pressures – Appliance categories

[5] ISO 12213-3:2006 Natural gas – Calculation of compression factor – Part 3: Calculation using physical properties