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A REVIEW OF NATURAL GAS-HYDROGEN BLENDING IN PIPELINE SUPPLY AND DISTRIBUTION

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ABSTRACT

The reduction of greenhouse gases is an assumed objective of the European Community. Thus, in the future, natural gas transport and distribution pipelines will also have to deal with ensuring the transport of mixtures formed between hydrogen, biomethane and natural gas from fossil resources. Considering the increase in the amount of biomethane and the increase in the amount of green hydrogen produced, these fuels's use in natural gas distribution networks is expected. The article presents an analysis of hydrogen production methods as well as numerical models of hydrogen transportation and mixing with natural gas. Also, there are presented the results obtained at the international level regarding the thresholds for the use of hydrogen in the fossil fuel of natural gas type, as well as the effects produced by the presence of this gas on the transport and supply systems. Numerical models of mixing and dynamic behavior of the new fuel obtained are also analyzed.

Keywords: hydrogen, natural gas, blending, mathematical modelling.

INTRODUCTION

The European Union has introduced as a strategy to reduce greenhouse gas emissions, the year 2050 as the date from which Europe will be neutral from a climate point of view [1]. The transition to this stage is contained in the European Green Deal [2] and represents the implementation of the Paris Agreement on climate change and the reduction of greenhouse gases [3]. This strategy also represents an opportunity to build intelligent natural gas transport and distribution networks, ensuring in addition to an efficient control of fossil methane leaks into the atmosphere and the use of ecological fuels mixed with them (biomethane and hydrogen).

In this sense, 32 operators from the oil industry have created a research hub, The European Hydrogen Backbone (EHB) initiative [4], which analyzes the increase in the weight of hydrogen (considered as a fuel) in the formation of fuels necessary for the production of electricity and ensuring family comfort normal and having as the final goal, the reduction of polluting emissions. At the same time, the increase in biodegradable waste has also led to an increase in biogas production, which in the future will be able to provide an increased amount of hydrogen and also green methane mixed with fossil natural gases [5].

The purpose of using renewable resources (biomethane and bio-hydrogen) is to reduce the use of fossil methane in the supply and use of natural gas systems, and implicitly the reduction of gaseous pollutants in the form of NOx, COx and H_2S. It is also predicted that the use of hydrogen and green methane could provide the necessary fuel for the production of electricity through local cogeneration [6]. But the use of hydrogen in natural gas networks must be treated with caution, both because of the changes in the properties of the mixture gas, as well as its effects on transport systems and especially its danger as an explosive medium [7].

However, in the decarbonization plan of the European Union [8], it is foreseen that by 2050, hydrogen will play an important role in reducing greenhouse gas emissions, accounting for between 2 and 14% of the European energy mix. The use of hydrogen can be in its pure form in the production of electricity and in ensuring the autonomy of the movement of vehicles and railways. At the same time, following the mixture with methane (fossil or obtained from renewable sources), the use of hydrogen would lead to the provision of energy fuel and a decrease of more than 40% in greenhouse emissions by 2030 (reported data compared to the reference year 1990).

That is precisely why, in the last time, the scientific research has also been channeled into the development of a series of projects for the development of natural gas distribution networks mixed with hydrogen [9-15].

The present study analyzes the effect of hydrogen on oil installations, the maximum amount permitted to achieve a safe mixture in operation and supply, as well as the problems encountered in the implementation of the analyzed projects.

In the analysis carried out by Keele University, as a result of the implementation of the research project HyDeploy project, it was possible to supply methane gas mixed with hydrogen in 101 residential houses, 8 student dormitories, 17 administrative buildings on the university campus and 7 sports halls and swimming pools of swimming [9]. The project consisted in determining the maximum amount of hydrogen needed to be injected (mixed) into methane, under increased safety conditions (without causing technical and human accidents). Mixing hydrogen with methane was achieved by injecting it into the annular distribution network of the research area followed by a press campaign to create a favorable opinion for the use of this type of fuel. After 36 months from the start of the project, it was found that the use of an amount of 20 mol % of hydrogen mixed with the natural gas existing in the distribution pipelines, is safe both in the transport and supply systems and in the use of this mixture in the final installations. At the same time, the combustion plants that worked at this level of hydrogen mixture functioned normally, with the detectors stopping the supply in case of reaching the threshold of 200 ppm of CO_2 in the flue gases. The conclusions of this project consist in the need to ensure a mixture proportion of maximum 20% mol of hydrogen in methane gas. The mixture was also observed to have a lower flammability limit of 4.74% vol compared to 5% vol for methane gas.

In 2016, Northern Gas Networks in England started a project on the use of pure hydrogen in natural gas networks in the Leeds area, in order to use the current distribution and supply networks for the delivery of hydrogen (in proportion of 100 vol.%) [10]. The obtained results demonstrated that the effect of hydrogen on polyethylene pipes is insignificant. Only in the case of steel pipes is their fragility observed when operating at increased pressures. This inconvenience was solved by using X80 type steels [13,14].

The project developed by Engie in Dunkerque, France (GRHYD) concluded that the use of maximum 20 mol % hydrogen in methane gas provides a 12 % reduction in CO_2 emissions, 37 % in CO emission, 39 ppm % in NOx emissions and 43 % ratio CO/CO_2 emissions.

The Dutch Naturally project team determined that the use of 10% vol hydrogen injected into some elements of the gas transport and supply system would not create problems of stability and provision of the infrastructure for possible gas leaks, but in the storage systems and the use of the natural gas-hydrogen mixture in the case of its use as car fuel [11].

EON through the H2HoWi project, developed in the period 2020-2023, started from the use of no more than 10% volume of hydrogen mixed with natural gas in supply and distribution networks and the development of electrical cogeneration facilities that use 100% hydrogen for production of energy [12]. The purpose of the project was to ensure electricity in some points with high demand, without the need to create new natural gas supply networks, but only hydrogen production facilities.

The recent problems in the supply of fossil natural gas (the war in Ukraine, the alarming increase in supply prices, etc.) created the premises for a new strategy of using natural gas networks for the delivery of green hydrogen and methane mixtures, as well as the possibility of developing hydrogen production projects for the creation of electricity cogeneration plants.

Hydrogen production methods

Hydrogen has a high efficiency in terms of the amount of energy produced and insignificant carbon emissions, being a function of its production technique, classified in:

- Gray Hydrogen, produced by steam reforming,
- Blue Hydrogen, obtained by steam reforming with carbon capture,
- Green Hydrogen, obtained from the electrolysis process with the help of renewable techniques for obtaining energy and through biological processes using various green mass waste as a substrate for bio-photolysis, fermentation in the absence of light and photo-fermentation.
- Yellow Hydrogen, obtained from the thermolysis process with the help of energy obtained from solar panels,
- Turqoise hydrogen, obtained from gas flows from the refinery through thermal cracking, catalytic cracking or catalytic reforming,
- Pink Hydrogen, obtained by electrolysis of water with the help of nuclear energy,
- Black Hydrogen obtained from coal gasification,

- White Hydrogen is obtained by hydraulic fracturing of geological layers containing hydrogen (in volcanic areas),

Steam reforming (Grey Hydrogen) of any hydrocarbon involves the endothermic conversion of some hydrocarbon and water vapor or steam into hydrogen and carbon monoxide. From a commercial point of view, steam reforming of methane is the most extensive method to produce hydrogen with a conversion of 74-85 % [15].

$$CH_4+H_2O \leftrightarrow CO+3H_2 \qquad \Delta_R\, H^0_{298\,K} = 206{,}2\ kJ/mol \qquad (1)$$

$$CH_4+2H_2O \leftrightarrow CO+4H_2 \qquad \Delta_R\, H^0_{298\,K} = 165{,}13\ kJ/mol \qquad (2)$$

The raw material is mostly represented by natural gas, where the main component is methane. Natural gas often needs to be pretreated, which involves sulfur removal step. Process temperatures and pressures vary between 820 – 880°C and 20-25 bar respectively.

A nickel-based catalyst is used for maximum conversion. Moreover, to increase the production of hydrogen, the water-gas shift reaction or the reaction [16] is carried out.

$$CO+H_2O \leftrightarrow CO_2+3H_2 \qquad \Delta_R\, H^0_{298\,K} = -41{,}2\ kJ/mol \qquad (3)$$

We observe that the process does not capture carbon dioxide but releases it into the atmosphere. Another technique for obtaining hydrogen is through the partial oxidation of methane or carbonate compounds through an exothermic process (the raw material being heated to temperatures of 1250-1400ºC, in the presence of a stoichiometric amount of pure oxygen) [17].

$$CH_4+0{,}5\ O_2 \leftrightarrow CO+2H_2 \qquad \Delta_R\, H^0_{298\,K} = -35{,}7\ kJ/mol \qquad (4)$$

In the 1950s-1960s, autothermal reforming was also used to obtain hydrogen as a combination of steam reforming and partial oxidation [18]:

$$4CH_4+O_2 + 2H_2O \leftrightarrow 4CO+10H_2 \qquad (5)$$

By capturing carbon dioxide, blue hydrogen is obtained, the obtaining process consisting of a combination of the steam reforming process and the partial oxidation process, in order to use CO_2 [19]:

$$CH_4+CO_2 \rightarrow 2CO+2H_2 \qquad \Delta_R\, H^0_{298\,K} = 247\ kJ/mol \qquad (6)$$

Plasma reforming is a process that generates temperatures above 2000 ºC but requires large amounts of energy to produce black hydrogen. Methane, diesel, gasoline, fuel oil can be used as raw material, obtaining hydrogen and carbon black by direct dissociation [20]. As an example, the dissociation of methane is presented in Eq.7:

$$CH_4 \rightarrow C+2H_2 \qquad \Delta_R\, H^0_{298\,K} = 74{,}6\ kJ/mol \qquad (7)$$

Coal gasification is a process of thermochemical transformation of coals, coke and other carbon-containing compounds. Depending on the process, solid coal is partially oxidized with air or a mixture of oxygen and water vapor or carbon dioxide necessary to produce hydrogen black, carbon monoxide, synthetic gas, carbon dioxide, water vapor and ash.

$$C+O_2 \rightarrow CO_2 \qquad (8)$$

$$C+H_2O \rightarrow H_2+CO \qquad (9)$$

$$C+H_2 \rightarrow CH_4 \qquad (10)$$

Turqoise hydrogen is obtained from refinery gas streams through thermal cracking, catalytic cracking or catalytic reforming. During catalytic reforming, naphthenes are dehydrogenated and converted to aromatics. For example, hydrogen and benzene are obtained by dehydrogenation of cyclohexane. The reaction is exothermic, taking place at temperatures of 300-450ºC in the presence of a platinum catalyst.

$$C_6H_{12} \rightarrow C_6H_6 + 3H_2 \qquad \Delta_R\, H^0_{298\,K} = 206{,}2\ kJ/mol \qquad (11)$$

Yellow Hydrogen is obtained by dissociating the water molecule at approximately 1800 ºC [15], the yield of thermolysis increasing with temperature (at 5000 ºC the maximum threshold of 50 % is reached). The reaction is reversible at low temperatures.

Green Hydrogen is a process of obtaining hydrogen through the electrolysis of water which consists of passing a continuous electric current (obtained from renewable resources) through two electrodes immersed in water.

At the cathode we will have the chemical reaction:

$$2H_2O \rightarrow H_2 + 2OH^- - 2e, \qquad (12)$$

and at the anode the electrons are released and they are captured further by the cathode:

$$2OH^- \rightarrow ½\ O_2 + H_2O + 2e \qquad (13)$$

By summing the two partial processes, the reaction of separating oxygen from hydrogen is obtained at a potential difference of 1.23 volts [16].

$$2H_2O \rightarrow H_2(gaz) + 2O_2\ (gaz) \qquad (14)$$

Electrolysis is the best performing process, reaching up to 60% efficiency in hydrogen production and having oxygen as a by-product, but it requires expensive electrocatalytic metals (platinum) and has a high corrosive effect at the cathode.

Pink Hydrogen is obtained from the dissociation of water with the help of nuclear energy.

Through bio-photolysis, the dissociation of the H-O bond is achieved under the action of light or ultraviolet radiation, with the help of chemical catalysts (zinc oxide - ZnO, tin oxide - SnO, zirconium dioxide - ZrO_2 and other semiconductor sulfur oxides).

The reduction of pollution and therefore the use of biodegradable green mass in the production of hydrogen, is one of the most promising alternatives, due to the diversity of raw materials that could be processed and the possibility of using microorganisms such as microalgae, cyanobacteria, fermentative bacteria, photosynthetic bacteria, which behave as and bio hydrogen plants.

Also, the use of bacteria does not require increased energy consumption, the process being carried out at normal atmospheric temperatures and pressures.

Biomass is the most renewable source of hydrogen production and uses the solar energy. This is captured in the process of photosynthesis (in growing plants), chlorophyll absorbing solar energy and then using it in the following reaction [23]:

$$x\ CO_2 + x\ H_2O \rightarrow (CH_2O)x + x\ O_2 \qquad (15)$$

The organic compounds produced during this stage of the process are carbohydrates and lignin. The existence in biomass of cellulose, hemicellulose, lignin, lipids, proteins, simple sugars, starch, water and hydrocarbons can influence the thermal or biochemical conversion process, the final products obtained being qualitatively and quantitatively different.

Biomass pyrolysis is the process of its thermal decomposition in the absence of oxygen, this being an endothermic process.

$$\text{Biomass+ energy} \rightarrow \text{bio} - \text{oil} + \text{charcoal} + \text{syngaz} \quad (16)$$

$$\text{Bio} - \text{oil} + H_2O \rightarrow CO + H_2 \quad (17)$$

$$CO + H_2O \rightarrow CO_2 + H_2 \quad (18)$$

Another bio-technological process is the biomass gasification, which represents the transformation of organic compounds into gaseous fuel (thermal degradation of biomass in the presence of an oxidizing agent and at temperatures above 1000 K).

Hydrogen production through biomass gasification consists of biomass gasification per se, catalytic reforming of synthesis gas, hydrogen separation and purification [23].

Biomass+steam→temperature→H_2+CO+CO_2+CH_4+ other hydrocarbons + Charcoal

Numerical models for the flow of hydrogen-natural gas mixtures through pipelines

To write the equations for the flow of blends formed by hydrogen and natural gas through pipes, the energy conservation equation (Bernoulli) was used [24]:

$$-Vdp = d\left(\frac{v^2}{2g}\right) + \lambda \frac{dL}{D}\frac{v^2}{2g} \quad (19)$$

Equation 19 can be written for conditions close to normal (T_0 and p_0) in the form:

$$Q_0 = Q\frac{p_1}{p_0}\frac{T_0}{T} \quad (20)$$

$$Q_0 = \frac{\pi\sqrt{g}}{4}\frac{T_0}{p_0}\sqrt{\frac{D^5(p_1^2-p_2^2)R_a}{\lambda l \delta T}} \quad \left(\frac{m_N^3}{s}\right) \quad (21)$$

If we substitute the gravitational acceleration g=9.81 m/s^2 and the universal constant of gases relative to air, R_a= 8314 J/mol K =286,9 J/Kg K in the Equation 21, and take into consideration higher values for p_1 and p_2, we get the flow equation as:

$$Q_{CH_4} = 13{,}302\frac{T_0}{p_0}\sqrt{\frac{D^5(p_1^2-p_2^2)}{\lambda\delta_{CH_4}Z_{CH_4}LT}}\left(\frac{m^3}{s}\right) \quad (22)$$

Where: Q_0 is the gas flow rate at T_0 and p_0,

p_0, p_1, p_2 represent the normal, initial and final pressure, kgf/m^3,

L is the length of the pipe, m,

D is the pipe diameter, m,

δ is the density of the gas relative to air,

λ represents the coefficient of friction,

T_0 and T represent the absolute temperatures, ºK,

Z is the deviation (non-ideal) coefficient of gases.

To observe the effect of hydrogen on the initial flow we will rewrite equation 22 for hydrogen:

$$Q_{mixH_2} = 13{,}302 \frac{T_0}{p_0} \sqrt{\frac{D^5(p_1^2 - p_2^2)}{\lambda \delta_{mixH_2} Z_{mixH_2} LT}} \left(\frac{m^3}{s}\right) \tag{23}$$

And dividing equation 23 by 22 we get:

$$\frac{Q_{mixH_2}}{Q_{CH_4}} = \sqrt{\frac{\delta_{CH_4}}{\delta_{mixH_2}}} \cdot \sqrt{\frac{Z_{CH_4}}{Z_{mixH_2}}} \tag{24}$$

Equation 24 can also be written in the form:

$$\frac{Q_{mixH_2}}{Q_{CH_4}} = \sqrt{\frac{M_{CH_4}}{M_{mixH_2}}} \cdot \sqrt{\frac{Z_{CH_4}}{Z_{mixH_2}}} \tag{25}$$

Where M_{CH_4} and M_{mixH_2} is the molecular mass of pure methane and hydrogen mixture.

The influence of hydrogen on the natural gas mixture was studied on a natural gas extracted from the Transylvania area and which had the following composition: methane CH_4=87% by volume, ethane C_2H_6=8% by volume, propane C_3H_8=2% by volume and nitrogen N_2=3%.

The equations for the variation of the properties of the methane gas mixture depending on the hydrogen content in the mixture are given in Table 1 and are obtained by processing the experimental and calculation data.

Table 1. Properties of the mixture of natural gas and hydrogen (values determined by calculation and from experiments)

Properties	**The behavior equation, where *y* is the property and *x* is the volume content of hydrogen**	**Coefficient of determination, R^2**
Molecular mass	$y = -0.15x + 17.66$	1
The non ideal factor, *Z*	$y = 6E\text{-}05x + 0.975$	1
$\sqrt{\frac{M_{CH_4}}{M_{mixH_2}}}$	$y = 0.0244x + 0.9496$	0.9983
$\sqrt{\frac{Z_{CH_4}}{Z_{mixH_2}}}$	$y = -0.0002x + 1.0003$	1
$\frac{Q_{mixH_2}}{Q_{CH_4}}$	$y = 0.0048x + 0.9983$	0.9983
Relative density	$y = -0.0061x + 0.6012$	0.9998
Gross caloric value (MJ/Nm^3)	$y = -0{,}1376x + 50.451$	0.9925
Wobbe index of blending (MJ/Nm^3)	$y = -0.2968x + 39.227$	0.9974
Boiling time of one litre of water, s	$y = -0.1442x^2 + 5.3091x + 348.77$	0.9924

The effect of hydrogen on the mixture is shown in figure 1.

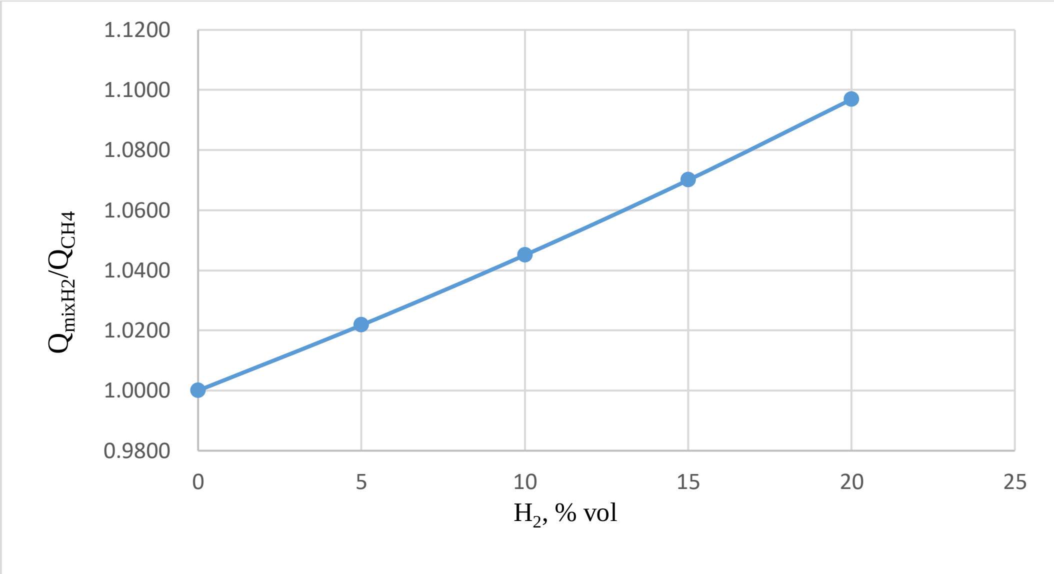

Figure 1. Effect of hydrogen on increasing gas flow rate

Table 2. Some values of the mixture of natural gas and hydrogen (values processed from laboratory data and calculation)

Hydrogen blending whit natural gas % hydrogen volume	Molecular mass	The non ideal factor, Z	Relative density	Gross caloric value (MJ/Nm^3)	Wobbe index of blending (MJ/Nm^3)	Boiling time of one litre of water, s
0	17,66	0,975	0,60	50,45	39,23	348
5	16,91	0,9753	0,57	49,76	37,74	372
10	16,16	0,9756	0,54	49,08	36,26	387
15	15,41	0,9759	0,51	48,39	34,78	396
20	14,66	0,9762	0,48	47,70	33,29	397

CONCLUSION

The analysis of the industrial experiments carried out and also correlating these data with those obtained in the laboratory entitle us to conclude the following:

a. Hydrogen can be used in gas distribution networks and in natural gas transport pipelines with up to 20 % volume percentages.

b. Effect of hydrogen on polyethylene pipes is insignificant.

c. effect of hydrogen of steel pipes is the active fragility of steels when operating at increased pressures.

d. the use of a maximum of 20 mol % of hydrogen in methane gas provides a 12 % reduction in CO_2 emissions, 37 % in CO emission, 39 ppm % in NOx emissions and 43 % ration CO/CO_2 emissions.

e. The effect of hydrogen on the mixture increase of the natural gas flow rate by a maximum of 1.96 is observed.

f. Gross caloric value and Wobbe index of blending decrease with increasing hydrogen content (with a maximum of 1.178), but the effect on the atmosphere leads to a reduction in pollutant emissions.

g. The boiling time of a liter of water increases by 0.877.

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