Acta Tecnología - International Scientific Journal about Technologies
Volume: 6 2020 Issue: 4 Pages: 111-117 ISSN 2453-675X

doi:10.22306/atec.v6i4.94 *Received: 19 Oct. 2020; Revised: 12 Nov. 2020; Accepted: 27 Nov. 2020*

REDUCTION OF POLLUTANTS IN THE RESIDENTIAL SECTOR BY MIXING HYDROGEN INTO THE NATURAL GAS NETWORK IN HUNGARY

István Szunyog
University of Miskolc, Miskolc - Egyetemváros, Miskolc, Hungary, szunyogi@uni-miskolc.hu (corresponding author)
Anna Bella Galyas
University of Miskolc, Miskolc - Egyetemváros, Miskolc, Hungary, gazgab@uni-miskolc.hu

Keywords: hydrogen in natural gas system, renewable energy comsumption, GHG reduction, hydrogen-natural gas mixture
Abstract: According to some forecasts, hydrogen will play a significant role throughout the world by 2030 as an energy source, the biggest benefits of which include not only being able to come from renewable sources, but thus storing the energy produced, which is not currently solved. The combustion of hydrogen does not produce CO_2, only negligible amounts of combustion air, unlike methane. This will reduce GHG emissions associated with end-user equipment. In this article, the authors examine the amount of hydrogen that can be fed into the Hungarian natural gas network in accordance with the current gas quality standard, and then carry out a comparative analysis of the methane, the main component that makes up hydrogen and natural gas. The authors will study the exact effect of hydrogen content on natural gas-regulated devices and estimate the theoretical CO_2 emissions available in the Hungarian residential sector at different rates of hydrogen.

1 Introduction

One of the most pressing global challenges of our time is to stop climate change. A significant group of impact gases emitted into the atmosphere can be linked to energy use and, as a means, to end-user combustion plants. At European level, the desire to increasingly replace conventional fossil fuels with renewable sources is growing, from industrial-scale electricity generation to the communal and residential sectors.

The highest GHG emissions from gas-fuelling gas-based wastes come from the carbon dioxide generated by the combustion. Renewable gases to be introduced into the gas grid are the most promising substitutes for natural gas, hydrogen being a legitimate priority on the part of the government, but it is necessary to examine exactly what impact this has on the devices regulated for natural gas.

In the European Union, we hear about a number of projects that aim to examine the issue, but there are no concrete results that can be introduced into domestic practice yet [1]. Based on the results of most research, about 15-20% hydrogen can be fed into the natural gas network without any or very small modification of gas user equipment. Even this amount of hydrogen is resulting in a significant reduction in GHG emissions. The authors set out to estimate the theoretical emissions of pollutants available in the residential sector in Hungary at different levels of gas network hydrogen mixing ratios.

One of the largest user segments of the natural gas sector is the residential sector in the EU and also in Hungary. In 2018, this sector supplied 3.35 billion m^3 of natural gas to the 3.26 million Hungarian residential customers in Hungary [2]. The numbers show the weight of the problem, as gas-supplying appliances in the residential sector are very large and cannot be switched, or only with significant economic investment, to fire up other gas quality. The question is therefore what is still permissible gas quality that can be fired with sufficient energy efficiency and safety in natural gas-controlled appliances.

2 Requirements for mixed hydrogen into natural gas system

Current European research [3,4] foresees that a maximum of 10% and possibly 20% is expected to be the rate that can still be safely mixed with the current gas equipment stock.

On the basis of the literature analysis, the following findings can be made for the retail sector:

- The residential sector has a large number of devices with an in homogenic distribution, type, age and technical condition.
- Most domestic and municipal gas-supplying appliances are likely to be able to handle the natural gas-hydrogen gas mixture without problems with a minimum (5-10%) in the case of an implication rate.
- The behaviour of atmospheric air pre-mixing and ventilation burners shall be tested separately.
- It can be concluded that the risk of natural gas consumption appliances with a hydrogen content of 10% depends essentially on two factors: the primary air volume and the Wobbe number of natural gas.
- The most sensitive to changes in hydrogen content are atmospheric burners.

- On the basis of the international test results so far, no general determination can be made on the maximum permissible hydrogen content!

The problem should essentially be examined from two perspectives, from the side of the device technique and the quality of the mains gas.

The technical side of the apparatus is the gas appliance tests carried out by certification and certification bodies on the basis of the test standards set before the gas appliances were placed on the market in that country. The gaseous apparatus tests shall be based on the principle that the operation of the apparatus shall be tested with reference gas of a specified quality and the associated boundary gases and test pressures indicating extreme gas quality. If the operation of the appliance with these gases is satisfactory, it may be placed on the market in the country of destination by actuating the gas of that group of gases. It should be mentioned here that this makes the issue more complex, since consumers also have devices that were put in place 20 or 30 years ago, and often we do not know their frequency of maintenance. So, the risk is a given. The increasing hydrogen share also means an increasing flame propagation rate, gradually increasing the risk of flame re-ignition into the burner. Unfortunately, there may also be old appliances for consumers that do not have or do not have a flame-protection, flame-guard unit.

Figure 1 shows the flame propagation rate values for a pure gas composed of methane, a gas of 70% methane and 30% hydrogen, and a gas with a hydrogen composition of 50% and 50%. It can be seen that the maximum values are around 10% excess air i.e., here the most ideal mixture of gas and air is in terms of combustion processes. It is a good thing that the maximum flame propagation rate that is developing is around 40 cm/s for methane, but for a 50-50% mixture, this is already 1.6 times that which the burner of the device can no longer tolerate and the flame can ignite. It can be concluded that testing the flame propagation rate is an essential requirement [5].

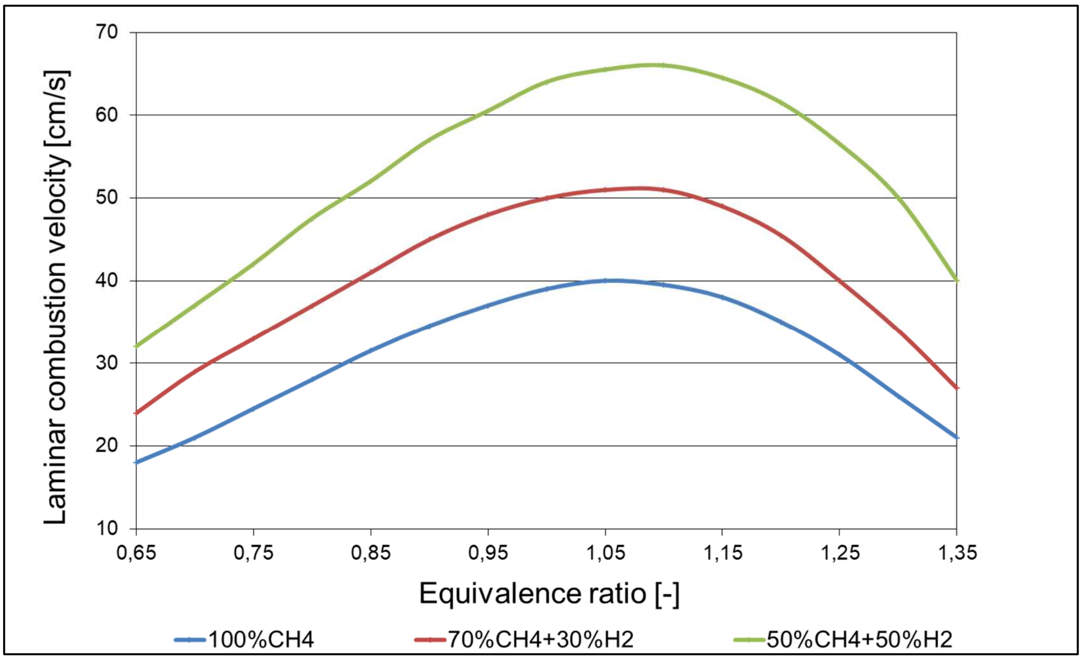

Figure 1 Laminar combustion velocity of methane-hydrogen gas mixture

On the gas quality standards side, the question can be concluded that keeping the quality of gas supplied through the natural gas network within narrow limits is in the fundamental interest of system operators. Disturbing changes in gas quality would put into question the safe and energy-efficient operation of end-user equipment. When regulating the feed of combustible gases of a quality other than natural gas into the natural gas system, the quality parameters of the gas supplied should remain at the heart of the set of requirements. The standards and specifications most used in Europe are based on EN 437 i.e., compliance with Wobbe number [6]. The standard follows the principle that only a change in the burning characteristics of natural gas within a group of natural gas may be allowed which does not affect the operation of the gas appliances and does not require the conversion of the appliance or burner. Gas other than natural gas belonging to the original quality group may only be used after the proper adjustment of the appliance (component replacement, reassembly, adjustment). In the case of the H gas group, which is also crucially available in Hungary, the standard has provided for a test with a hydrogen content of 23% in the border gas of re-ignition since 1999, but this does not provide a close guarantee that the devices installed thereafter would be able to wither this effect continuously and over the long term.

Volume: 6 2020 Issue: 4 Pages: 111-117 ISSN 2453-675X

If a pure methane-hydrogen gas mixture is used, a hydrogen content of not more than 25,45 mol% is allowed in the mixture, so that the mixture still meets the heat value for gas group H, and 41,09% hydrogen may still meet the requirements of the Wobbe number i.e., it may remain within the permitted ranges for the gas group. (Figure 2) It follows that, for an average natural gas composition, the maximum proportion of hydrogen that can be fed into the gas group H is approximately 25% i.e., there is no group change. This is confirmed by the maximum share of 20 mol% and 23 mol% respectively of the limit gases in EN 437. We note that this theoretical number is only approximated from the gas quality side, it does not provide information about the operating side of the device, its behaviour (in particular the changing flame propagation rate).

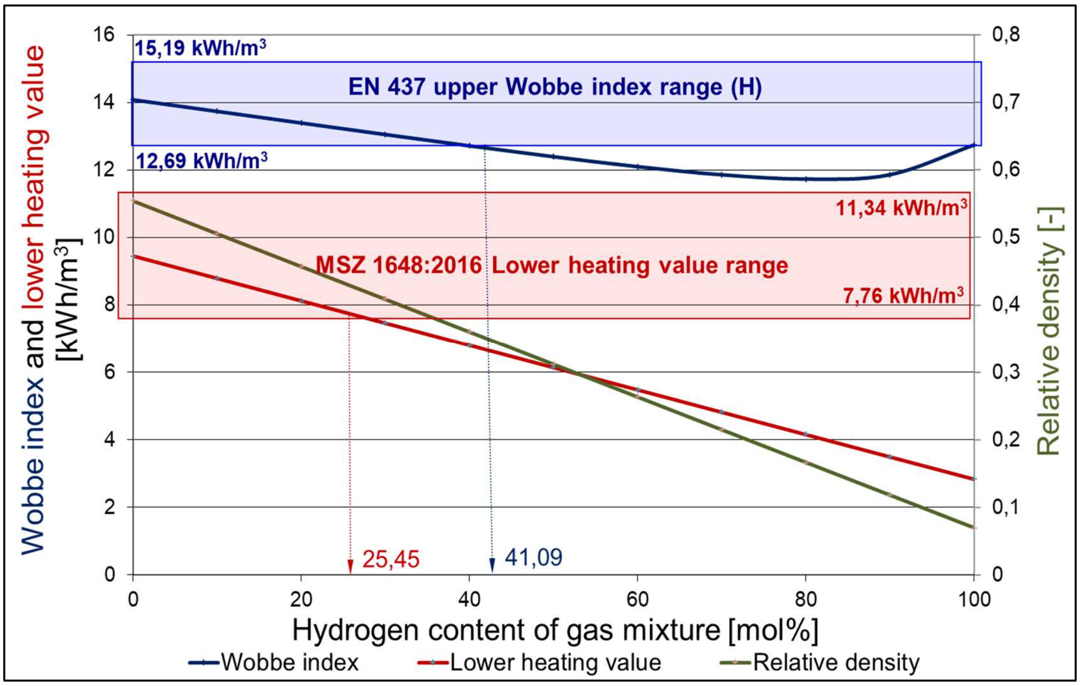

Figure 2 Combustion characteristics of methane-hydrogen gas mixture

3 Gas quality characteristics of hydrogen and natural gas

Table 1 shows the physical and combustion properties of hydrogen and a typical Russian natural gas. The parameters that can be the source of the problem during the fire are clearly shown in the table. Because hydrogen has a very low relative density compared to air (Figure 2), therefore, in case of leakage, it is much easier to separate from methane and form explosive medium in the setting room.

Table 1 Comparison of combustion properties of hydrogen and natural gas [7-9]

Properties	Unit	Hydrogen (H_2)	Natural gas (2H russian)
Molar weight	kg/kmol	2.016	16.409
Density (15 °C; p_n)	kg/m^3	0.090	0.695
Relative density	-	0.070	0.567
Lower heating value (15/15 °C)	kWh/m^3	2.840	9.501
Upper heating value (15/15 °C)	kWh/m^3	3.362	10.548
Lower Wobbe index	kWh/m^3	10.496	12.613
Upper Wobbe index	kWh/m^3	12.744	14.002
Laminar ombustion velocity	cm/s	~267	~34
Flammability limits (20 °C)	V/V%	4.0-77.0	4.3-15.6
Theoretical oxygen volume	m^3/m^3	0.499	2.014
Theoretical air volume	m^3/m^3	2.383	9.614
CO_2 emission	m^3/m^3	0,001	1.009
Methane number	-	0,0	92.5

Acta Tecnología - International Scientific Journal about Technologies
Volume: 6 2020 Issue: 4 Pages: 111-117 ISSN 2453-675X

Its energy content per unit volume is only a third of that of natural gas, but its Wobbe number, one of the most characteristic parameters of gas mixture interchangeability, is only 9.5%. The most important difference is the rate of flame propagation of gases and the ranges of ignition concentrations, as hydrogen burns 10.5 times faster in the air than natural gas. When fired in gas appliances, this will be one of the most decisive variables, which will severely limit the allowable mixable quantities.

A very significant shift in the ignition concentration limit towards the upper limits indicates the danger of hydrogen gas flowing into the environment or fire field. (Figure 3) It can be seen that the lower ignition concentration limit is almost unchanged, but the upper limit increases exponentially with the enrichment of hydrogen.

The minimum ignition energy of hydrogen is about 10 times lower than methane, i.e. much easier to ignite, but their self-ignition temperature is quite similar (H_2: 560 °C; CH_4: 595 °C) [1]. As a result, natural gas-hydrogen mixtures can become more easily ignited, thus increasing a higher risk of explosion.

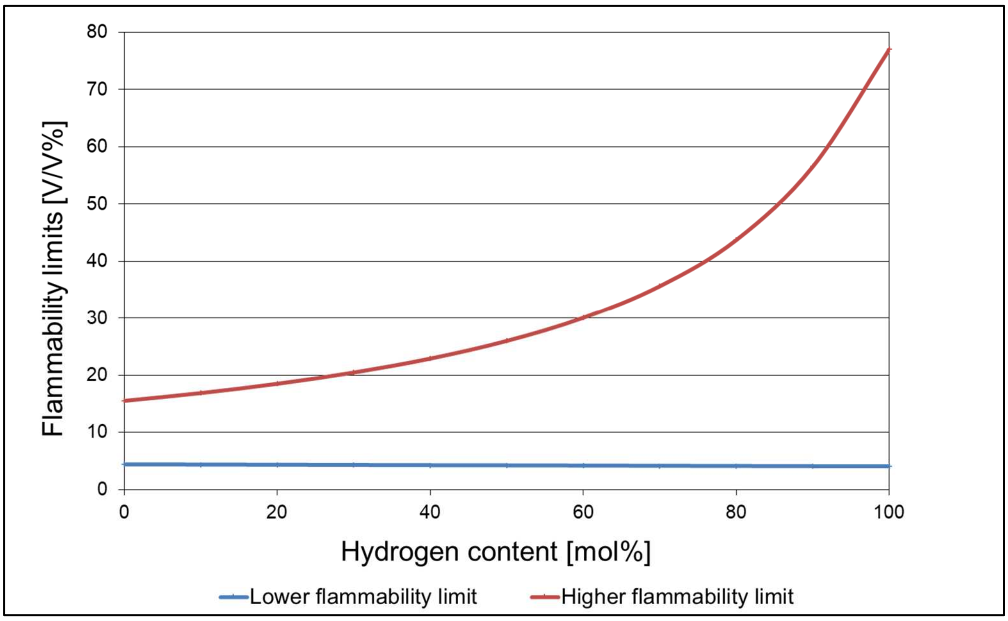

Figure 3 Flammability limits of methane-hydrogen mixture

4 Pollutant emissions

In the operation of the devices, it would be advisable to use a natural gas-hydrogen mixture in order to reduce the amount of carbon dioxide that is going into the atmosphere from the burning of fossil natural gas. Figure 4 shows CO_2 emissions in g/MJ calculated on the basis of the theoretical air volume and the lower heat value of the gas. The CO_2 emission value of pure methane 58.03 g/MJ (15 °C; pn) may be reduced to 0.13 g/MJ using pure hydrogen. Note: the minimum carbon dioxide content of the hydrogen combustion product is not zero due to the carbon dioxide content of the air used for combustion. For the maximum blending rate of 25% tested by us, a value of 52.73 g/MJ can be achieved, resulting in a saving of 5.3 g/MJ. In Hungary, in 2018, the quantity of natural gas transferred to residential customers during domestic sales was 115,084,720 GJ, equivalent to 3.36 billion m^3 for the average annual lower heat value of natural gas at a combustion and measurement reference state of 15/15 °C. The average lower heat value is calculated on the basis of MEKH statistics on energy content (GJ) and volume unit (em^3) [2].

During the research, we conducted studies on how much CO_2 emission reductions can be achieved during the replacement of natural gas with hydrogen, based on residential natural gas use in 2018. In the year under consideration, as a result of gas consumption in the residential sector, 6,623 thousand tonnes of CO_2 were added to the atmosphere, accounting for 14.7% of the country's total CO_2 emissions that year [10]. Chart 5 shows the extent to which CO_2 emissions can be saved by increasing the supply of hydrogen to the natural gas network and its use in residential gas-supplying appliances. In 2018, the replacement of 1 mol% of the amount of natural gas consumed by the residential sector to hydrogen (in terms of energy content) will save 19.8 thousand tonnes of CO_2 emissions, which represents 0.04% of the carbon dioxide emissions generated by Hungary in the year under review. The use of natural gas with a hydrogen content of 5 mol% is already 102.1 thousand tonnes, while gas with a hydrogen content of 10 mol% results in a decrease in CO_2 emissions of 211.8 thousand tonnes based on residential natural gas consumption in 2018. If the 25% hydrogen content of natural gas had been tested earlier, the CO_2

emissions measured in 2018 would have already decreased by 9%.

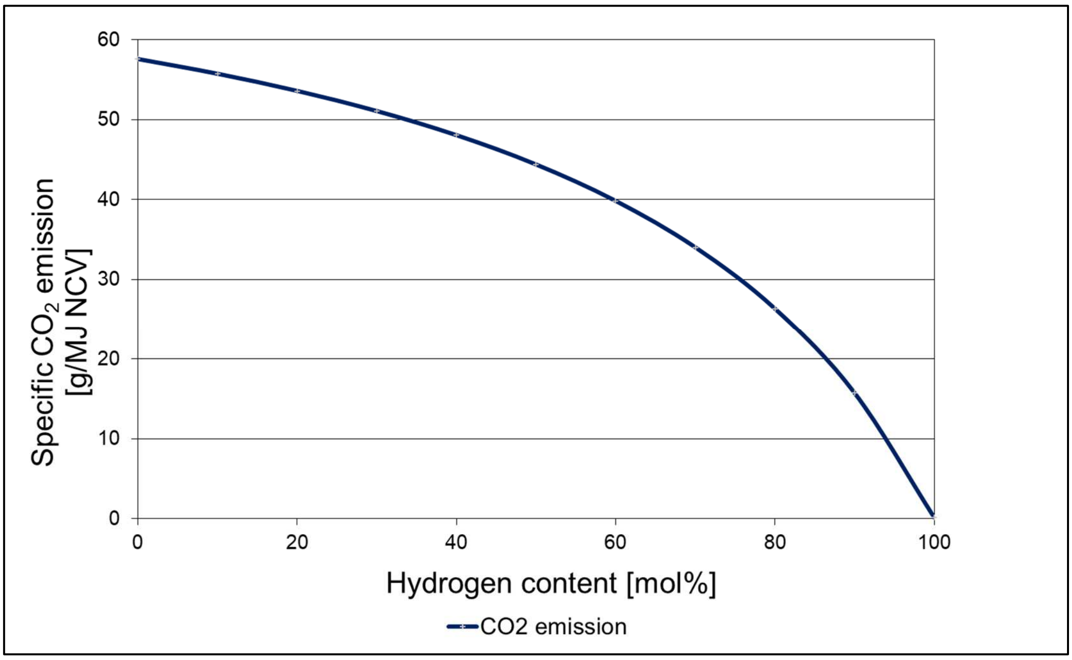

Figure 4 Evolution of the specific combustion-related CO_2 emissions of methane-natural gas mixture

In addition, it is important to note that hydrogen has a significantly lower energy density than natural gas – approximately only 30 % – and therefore requires a higher amount of methane-hydrogen gas mixture to achieve the same output. As can be seen in Table 2, the replacement of natural gas supplied to the residential sector in 2018 to 1 mol% hydrogen (in terms of energy content) will increase residential consumption in that year by 79 million m^3, while for 10 mol% hydrogen content it will mean 790 million m^3 of additional gas, which is nearly 24% higher than the amount of residential natural gas consumption in m^3 unit in 2018. Natural gas with a hydrogen content of 25 % increases the consumption of natural gas by approximately 2 billion m^3 in the year under consideration, and a hydrogen molecule would constitute 52.8 V/V% of the resulting 5.33 billion m^3 natural gas-hydrogen mixture. In the year under consideration, the total consumption of natural gas by hydrogen requires the production of 11.26 billion m^3 of hydrogen gas, which is 3.35 times the volume of natural gas originally supplied.

In addition to CO_2, only carbon monoxide and nitrogen oxide emissions are relevant for combustion-related emissions. Carbon monoxide (CO) is basically formed due to imperfect combustion i.e., too little combustion air surplus, or too high a fire field temperature. If these factors can be eliminated from the process, the amount of formation will not be decisive. If the air supply of the appliance is not re-regulated when hydrogen gas is fed, excess air will not allow carbon monoxide to form. The temperature of the fire field is proportional to the flame temperature, at which we saw that it does not increase significantly with up to 25% hydrogen mixing i.e., this factor is not really relevant either. However, it should not be ignored that excess air that is too high cools the fire area, making the flame increasingly unstable, which can lead to locally imperfect combustion i.e., carbon monoxide formation.

In the case of natural gas burning, the formation of nitrogen oxides due to thermal i.e., high fire field temperatures, can be decisive. The appearance of hydrogen in the mixture results in higher combustion temperatures and higher excess air, so more oxygen is available in the reaction zone. Since the increase in the hydrogen share also reduces the energy content of the gas per unit, the energy released, and with it the fire room temperature, will be lower if we do not change the amount of gas that led to the burner. It can be a typical situation in the residential sector. This also reduces the likelihood of nitrogen oxides forming.

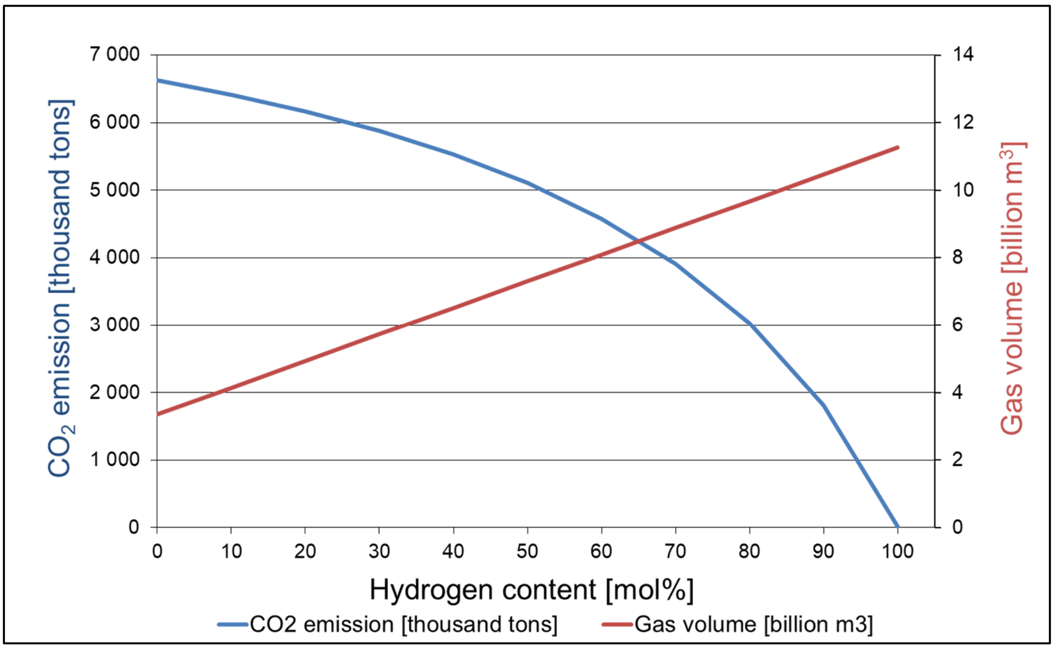

Figure 5 Theoretical CO_2 emissions savings by blending hydrogen into natural gas system in Hungary

Table 2 Replacement of natural gas with hydrogen in terms of energy content

	Domestic sales to residential users	1 mol% H_2 replacement	5 mol% H_2 replacement	10 mol% H_2 replacement	25 mol% H_2 replacement	100 mol% H_2 replacement	Unit
Natural gas	3 355 631 000	3 322 074 690	3 187 849 450	3 020 067 900	2 516 723 250	0	m³
Hydrogen	0	112 571 726	562 858 629	1 125 717 259	2 814 293 146	11 257 172 585	m³
Gas mixture	3 355 631 000	3 434 646 416	3 750 708 079	4 145 785 159	5 331 016 396	11 257 172 585	m³
Average lower heating value	9.527	9.460	9.192	8.858	7.855	2.840	kWh/m³

5 Conclusion

Hydrogen mixed into the natural gas network may be an appropriate environmentally friendly alternative to the partial replacement of natural gas, but the physico-chemical and combustion technical properties and safety conditions of the mixture, even significantly different from natural gas, should not be ignored. Combustion theory tests are forward-looking in terms of applicability, but on their own they can only provide support and guidance, they cannot replace the actual device tests. It is clear that the question must be answered before it is actually used in the residential gas consumer sector.

We need to distinguish between gas equipment techniques equipped with an automatic combustion control system, i.e. they can be adapted to varying gas quality to a certain extent, as well as the large number of gas appliance parks, mainly residential, in which, on the one hand, there is no automatic control system and, on the other hand, it contains a significant number of partial pre-mix burners. In this case, in addition to the changing gas composition, the excess air does not, or only minimally, change, while the energy content of the fuel is even more significant.

Hungary natural gas use, the retail sector plays a prominent role. As a consequence, significant attention should be paid to end-user equipment prior to the release of hydrogen on the natural gas network. The studies have clearly shown that the appearance of hydrogen on the natural gas network results in a not insignificant reduction in CO_2 emissions. Looking at the consumption of natural gas by the public in 2018 with the replacement of 25 mol% hydrogen content, the annual emissions of CO_2 Hungary have been reduced by nearly 10%. In addition, it should be noted that due to the lower energy content of hydrogen compared to natural gas, a higher gas volume is required to achieve the same output.

Hungary natural gas use, the retail sector plays a prominent role. As a consequence, particular attention should be paid to end-user equipment prior to the release of hydrogen on the natural gas network. In 2018, the replacement of 1% of natural gas consumed by the residential sector to hydrogen will result in a reduction in CO_2 emissions of 0.3% and 10% hydrogen by 3.2%.

It can be said with great probability that hydrogen mixed in natural gas is strongly suited to the decarbonisation of the residential energy sector, but the restrictive requirements dictated by the existing gas appliance park should also be strongly taken into account. The mixture does not pose a safety and fire risk up to a certain composition limit, but the high hydrogen share will clearly only be available by replacing the existing equipment fleet.

Acknowledgement

The described article was carried out as part of the EFOP-3.6.1-16-2016-00011 "Younger and Renewing University – Innovative Knowledge City – institutional development of the University of Miskolc aiming at intelligent specialisation" project implemented in the framework of the Szechenyi 2020 program. The realization of this project is supported by the European Union, co-financed by the European Social Fund."

References

[1] SCHRÖDER, V., ASKAR, E., TASHQIN, T., HABIB, A. K.: *Sicherheitstechnische Eigenschaften von Erdgas-Wasserstoff-Gemischen,* Bundesanstalt für Materialforschung und -prüfung, Berlin, Abschlussbericht zum Forschungsvorhaben 2539, 2016.

[2] MEKH Magyar Energetikai és Közmű-szabályozási Hivatal: *A magyar földgázrendszer 2018, évi adatai*; Available: http://www.mekh.hu/a-magyar-foldgazrendszer-2018-evi-adatai [18 Oct 2020], 2018. (Original in Hungarian)

[3] SMITH, N., BYRNE, N., COATES, M., LINTON, V., VAN ALPHEN, K.: *Identifying the commercial, technical and regulatory issues for injecting renewable gas in Australian distribution gas networks,* Research Report, 2017.

[4] ALTFELD, K., PINCHBECK, D.: *Admissible Hydrogen Concentrations in Natural Gas Systems, HIPS – the paper,* 2013. Available: https://www.gerg.eu/wp-content/uploads/2019/10/HIPS_Final-Report.pdf [18 Oct 2020], 2013.

[5] THYGA PROJECT *Testing Hydrogen admixture for Gas Applications: Impact of hydrogen admixture on combustion processes – Part I: Theory Deliverable: D2.2 Status: Final,* 27th of June 2020, 2020.

[6] EN 437:2019 *Test gases – Test Pressures – Appliance categories,* 2019.

[7] SZUNYOG, I.: *Quality Requirements of the Application of Biogases in Natural Gas Public Utility Services in Hungary,* PhD dissertation, 2009.

[8] LEE, S. W., LEE, H. S., PARK, Y. J., CHO, Y.: Combustion and emission characteristics of HCNG in a constant volume chamber, *Journal of Mechanical Science and Technology,* Vol. 25, No. 2, pp. 489-494, 2011.

[9] MAZLOOMI, K., GOMES, C.: Hydrogen as an energy carrier: Prospects and challenges, *Renewable and Sustainable Energy Reviews,* Vol. 16, No. 5, pp. 3024-3033, 2012.

[10] eurostat Data Browser, Greenhouse gas emissions by source sector (source: EEA): https://ec.europa.eu/eurostat/databrowser/view/env_air_gge/default/table?lang=en [18 Oct 2020], 2020.

Review process

Single-blind peer review process.