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INVESTIGATION OF THE HYDROGEN TOLERANCE OF THE HUNGARIAN NATURAL GAS SUPPLY SYSTEM

NEW SCIENTIFIC ACHIEVEMENTS OF PHD THESIS

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1 The aim of the research work

One of the most pressing problems today is the curbing of climate change, one of the major sources of which is the release into the atmosphere of greenhouse gases associated with energy use (GHG). There is also a growing demand at European level for renewable energy sources that can partially or fully replace traditional fossil fuels. Hydrogen promises to be suitable for this purpose. One of the biggest advantages of renewable hydrogen over current fossil fuels is that it produces water vapour when combusted, significantly reducing CO_2 emissions into the atmosphere.

According to the study "National Energy Strategy up to 2030 with an Outlook up to 2040" published by the Ministry of Innovation and Technology in January 2020, existing natural gas infrastructure could serve as "seasonal energy storage" in the future. This foreshadows the vision that one alternative to electricity and heat storage solutions could be to store energy in "molecular form", so that it can be stored for longer periods of time and used according to emerging needs. Using power-to-gas (hereinafter referred to as P2G) technology, which is already available on the market, hydrogen can be produced by photovoltaic and methane using carbon dioxide, which can then be fed directly into the natural gas network, eliminating daily and even annual seasonality issues *(Hungary's National Hydrogen Strategy, 2021)*.

But solving climate change is not that simple. The properties of hydrogen are significantly different from those of natural gas used in public services, posing a serious challenge to operators of the natural gas supply system (including transmission, storage, distribution and consumption infrastructures). For this reason, a lot of industrial research is currently underway to determine the maximum hydrogen content that natural gas networks and related gas user equipment can tolerate under safe operating conditions.

During my doctoral research I examined technical issues that can facilitate the utilization of hydrogen through the natural gas network in Hungary. In my thesis I examined the effect of hydrogen on the parameters defined by the Hungarian gas quality standards. The question of the change of the water dew point and the hydrocarbon dew point during the processing for the supply into the Hungarian natural gas system is significant, since the appearance of hydrogen may even require modification of the currently used technology. I examined the possibility of improving quality with commercial-grade propane, in which case my goal is to create a diagram with which the amount of propane that can be mixed into the methane-hydrogen gas mixture can be easily determined, meeting the domestic gas quality requirements.

In addition, a significant question arises to what extent the blending of hydrogen with natural gas modifies the energy capacity that can be transferred through the gas pipeline, for the quantification of which I developed an equation for the distribution and transmission networks, assuming constant operating conditions. In order to increase the complexity of the topic studied, I also aimed to determine the hydrogen potential produced from electricity on a renewable basis in Hungary, based on the available capacity resources. The presented results may also contribute to the elaboration of the technical-regulatory background of the domestic natural gas network of hydrogen.

2 Scientific preliminaries

During hydrogen feed-in, the use of the existing natural gas network has many advantages, the most significant of which is that the construction of new infrastructure can be avoided, which significantly reduces investment costs. In order to achieve climate neutrality, several international hydrogen projects have been launched in recent years, examining the possibility of generating, transporting and distributing hydrogen, as well as the hydrogen tolerance of gas-consuming devices currently in operation. When examining the feed-in of hydrogen into the natural gas grid, all research agrees that the operating natural gas network is suitable for receiving hydrogen to a small extent, but there are several critical questions arising from the effects of hydrogen on material quality and energy issues *(Scott et al., 2019; Liemberger et al., 2019; Quintino et al., 2021)*.

The permitted proportion of hydrogen that can be blended into the natural gas network is tested in many parts of the world, determining the amount that can be mixed in to safely operate the natural gas supply system. The table below summarises the most significant hydrogen feed-in projects in the European gas grid.

The most significant hydrogen projects in Europe *(Devinder et al, 2022)*

Project	Country	Year	Hydrogen [V/V%]	Project size range
HyDeploy	United Kingdom	2019	20	1500 households
East Neuk Power	United Kingdom	2020	20	15 GWh/year
Aberdeen Vision	United Kingdom	2020	20	300 households
HyNet Northwest	United Kingdom	2021	100	30 TWh/year
HyNTS Hydrogen Flow Loop	United Kingdom	2021	30	-
H21	United Kingdom	2018	100	6,4 TWh/year
Hy4Heat	United Kingdom	2018	100	-
HySpirit	United Kingdom	2019	100	-
Zero 2050 South Wale	United Kingdom	2020	100	-
Decarbonisation Pathway	United Kingdom	2020	100	-
GRHYD	France	2014	20	200 households
THyGA	EU	2019	10-100	100 households and commercial real estate
WindGas Falkenhagen	Germany	2013	2	-
WindGas Hamburg	Germany	2015	2	-

Note: The year shown in the table indicates the start date of the project.

Several researches deal with the effects and simulation modelling of hydrogen feed-into the natural gas network. A significant question is what kind of energy changes the appearance of hydrogen in the natural gas network causes in relation to the fuel flowing in the pipeline, which has already been examined by several authors analysing the issues to be solved during the feed into the natural gas supply network. Based on their research, they all concluded that the appearance of hydrogen in the natural gas network impairs the combustion characteristics, therefore this issue should be prioritized during planning *(Tabkhi et al., 2008; Di Lullo et al, 2021; Abd et al., 2021).*

Guandalini et al. developed a model for a section of gas pipeline assuming a non-steady-state state to analyse the dynamic effects of hydrogen feed-into the natural gas grid. In their investigations, a comparison was made for a section of pipeline running on natural gas and then natural gas mixed with hydrogen. During the analysis, the change in combustion characteristics was pointed out as a key evaluation criterion and it was shown that the proportion of hydrogen that can be mixed is significantly limited, one of the reasons for which is the gas appliances located at the end point of the system.

Abeysekera et al. investigated a theoretical low pressure gas network for hydrogen and biomethane mixing. The steady-state model presents two approaches: taking into account the gas volume flows required by gas consumers, and following the energy demand of consumers and, if necessary, increasing the volume flow. Their results showed that renewable gases have a significant impact on gas quality and fluid dynamics, especially flow rate and pressure, but if properly regulated, they can reduce import dependency and help the operation of the electricity grid.

Cheli et al. simulated a representative urban gas network with industrial and residential customers and an associated P2G system for which a steady-state model was developed. Based on their results, the hydrogen blended causes a significant decrease in Wobbe index. An algorithm has been developed to predict pressure loss and flow rate at each node.

Hafsi et al. studied numerical simulation of the flow of a transient hydrogen-natural gas mixture in a looped network based on a mathematical model that takes into account the change in the compressibility coefficient of the gas mixture according to pressure under isothermal conditions.

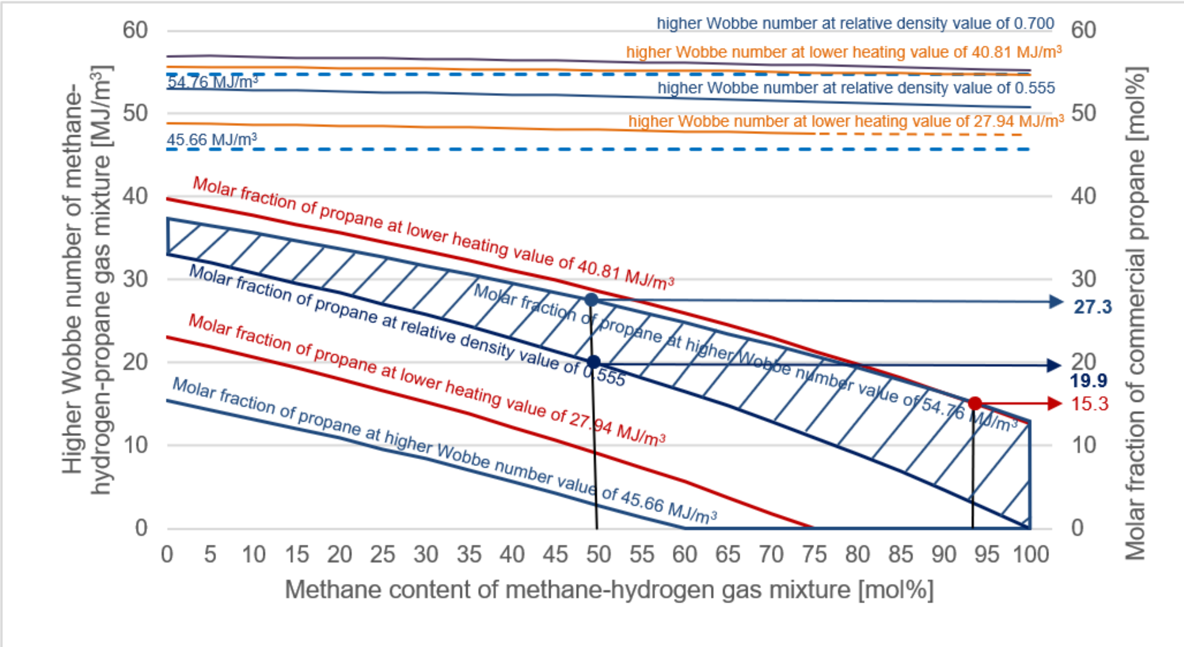
3 New scientific results

The new scientific results of the paper can be summarised as follows.

Thesis No. 1

a) I have developed a graphically plotted procedure for determining the commercial propane quantities required for natural gas network feed-in to the two-component methane-hydrogen gas mixture in case of gas quality 2H.

Presentation of the thesis:
Within the hatched area enclosed by boundary lines, the gas mixture formed by methane-hydrogen commercial propane satisfies the parameters of standard MSZ 1648:2016, which defines the requirements for natural gas network feed-in, in case of 2H gas quality.



Mixing limit values applicable when mixing the methane-hydrogen gas mixture with commercial propane depending on the upper Wobbe number range and relative density (2H) allowed by the MSZ 1648:2016 standard

In the case of any methane-hydrogen gas mixture of known composition, the minimum and maximum commercial propane quantity can be read from the figure, which can be mixed with the two-component gas mixture to meet the gas quality requirements for 2H gas quality, which are required for feeding into the natural gas network.

b) **I have defined the boundary conditions under which the gas mixture formed by the quantity of commercial propane required for natural gas network feed-in can be mixed with the two-component methane-hydrogen gas mixture and correspond to the gas quality 2H according to the Hungarian standard.**

Presentation of the thesis:
Based on the results of these tests, it can be concluded that a methane-hydrogen mixture of known composition requires the addition of at least a quantity of commercial propane to reach a relative density of 0.555, since in this case it satisfies the requirements for lower heating value, upper Wobbe number and relative density. At most, the quantity of commercial propane, which improves quality, is required to reach the upper limit of the Wobbe number range (54.76 MJ/m^3). In the case of a methane-hydrogen gas mixture with a CH_4 content of 93.0 mol%, the maximum amount of commercial propane that can be miscible is determined on the basis of the upper limit of the lower standard heating value (40.81 MJ/m^3) instead of the higher Wobbe number, since this upper limit is reached earlier.

Boundary conditions	<93,0 mol% CH_4	≥93,0 mol% CH_4
lower limit according to compliance	the methane-hydrogen-propane gas mixture reaches the lower limit of the relative density standard (0.555)	
upper limit according to compliance	the methane-hydrogen-propane gas mixture reaches the lower limit of the relative density standard (54.76 MJ/m^3)	the methane-hydrogen-propane gas mixture reaches the lower limit of the relative density standard (40.81 MJ/m^3)

Boundary conditions for compliance of gas mixture formed by methane-hydrogen commercial propane with the Hungarian standard for 2H gas quality

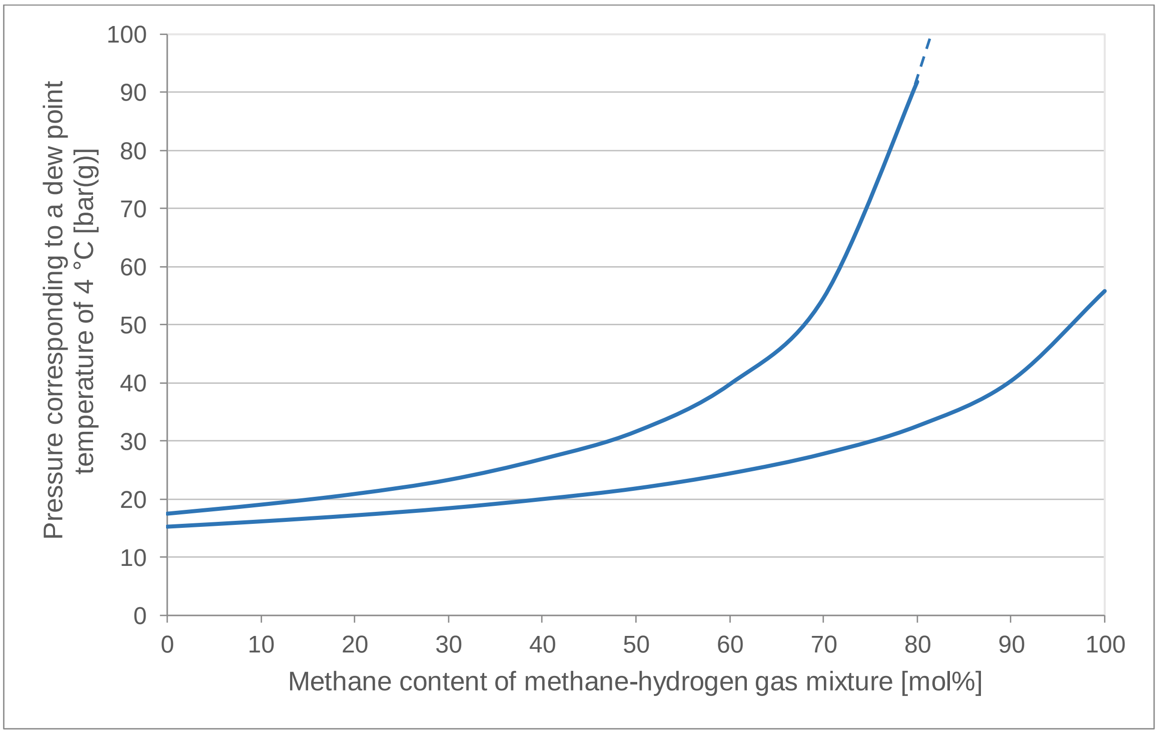
Thesis No. 2

According to standard MSZ 1648:2016, the phenomenon of hydrocarbon condensation can be predicted in the case of quality improvement of a methane-hydrogen gas mixture of known composition with commercial propane at the dew point temperature of 4 °C used for natural gases, in case of 2H gas quality.

Presentation of the thesis:
The phenomenon of hydrocarbon condensation in the natural gas system is not permissible. The relationship between pressure and temperature of hydrocarbon condensation is given by the hydrocarbon dew point curve corresponding to the given gas composition. The problem typically arises when components with higher carbon numbers occur, in the case under consideration, during quality improvement with commercial propane, which should be treated as a priority. The tests were performed with the simulation software called Aspen HYSYS. The

lower curve is the methane hydrogen boom produced by quality improvement with propane to the lower limit of relative density, while the upper curve is the upper limit of the Wobbe number range. In the case of a propane gas mixture (see demarcation lines of the area enclosed by five points in the preceding figure), show the pressure at which hydrocarbon condensation can be predicted at a dew point temperature of 4 °C.

Furthermore, it was established that the appearance of hydrogen in the natural gas network and its incorporation into natural gas without commercial-grade propane-incorporation does not increase the phenomenon of hydrocarbon condensation, but reduces the hydrocarbon dew point of the natural gas originally supplied at a given pressure, since, due to its composition, it does not increase the hydrocarbon content of the natural gas composition with a higher carbon number.



Hydrocarbon dew point curves of methane-hydrogen-propane gas mixture at 4 °C set to lower and upper limits of compliance according to MSZ 1648:2016 standard for 2H gas quality

Thesis No. 3

Based on the results of my simulation tests, it can be stated that in the case of 20 mol% hydrogen part of the natural gas network – taking into account the water saturation change – no technological change is required when setting the water dew point of the gas qualities in the domestic natural gas system.

Presentation of the thesis:
Simulation studies have shown that the water dew point modifying effect of hydrogen when mixed with 2H natural gas used in public services is negligible. According to the results, at atmospheric pressure, hydrogen reduces the maximum amount of water vapour that can be miscible with natural gas, but at higher pressures this effect is reversed, and the gas mixture can keep more water in a vapor phase at a given pressure and temperature than in the case

of the hydrogen-free sample gas composition. As a rule, with increasing pressure, this deviation increases. In Hungary, the water dew point of natural gas network quality gases is set during gas processing at a maximum pressure of 70 bar, where in the presence of 20 mol% hydrogen content, the value of the change in water saturation is not more than 3.09% at the minimum temperature tested, the value of the deviation decreases with increasing temperature compared to the original gas composition. It is very important to note that the determination of hydrogen resistance of material grades used in gas processing technology was not included in this study.

Thesis No. 4

In addition to the pressure conditions occurring in the Hungarian natural gas distribution network, I determined a relationship describing the change in the energy content that can be transferred through the pipeline, which occurs as a result of the hydrogen content, assuming a constant pressure loss.

Presentation of the thesis:

Using the characteristic equation for describing pipeline gas flows, derived from the frictional Bernoulli equation, I determined the relationship that gives the amount of energy capacity reduction compared to the pure methane content on a pipeline under pressure conditions in the natural gas distribution network. The significance of this is that under an operating pressure of 10 bar, the flowing gas can be considered ideal, so the value of the compressibility factor can be taken into account as 1, i.e. it does not affect the value of the KF conversion factor. Apparently, the conversion factor is determined by the heating value (H_a) and relative density (ρ_{rel}) of the flowing medium. During the derivation of the equation, constant operating conditions were assumed, so the most important parameter is the unchanged pressure loss during pipeline flow. Based on the results, when mixing a 10 mol% H_2 content, the transferable energy content relative to the pure methane content decreases by 2 % (KF=0.98), while at 20 mol% H_2 this value decreases by 5 % (KF=0.95).

$$\mathbf{KF} = \frac{\mathbf{E_{mix}}}{\mathbf{E_{CH_4}}} = \frac{\mathbf{H_{a_{mix}}}}{\mathbf{H_{a_{CH_4}}}} \cdot \sqrt{\frac{\boldsymbol{\rho}_{\mathbf{rel_{CH_4}}}}{\boldsymbol{\rho}_{\mathbf{rel_{mix}}}}} = \frac{\dfrac{\mathbf{H_{a_{mix}}}}{\sqrt{\boldsymbol{\rho}_{\mathbf{rel_{mix}}}}}}{\dfrac{\mathbf{H_{a_{CH_4}}}}{\sqrt{\boldsymbol{\rho}_{\mathbf{rel_{CH_4}}}}}}$$

Where

- KF – conversion factor [-];
- E_{mix} – energy content of the gas mixture [kWh/m^3];
- E_{CH_4} – energy content of methane [kWh/m^3];
- $H_{a_{mix}}$ – lower heating value of the gas mixture [kWh/m^3];
- $H_{a_{CH_4}}$– lower heating value of methane [kWh/m^3];
- $\rho_{rel_{CH_4}}$ – relative density of methane [-];
- $\rho_{rel_{mix}}$ – relative density of the gas mixture [-].

Thesis No. 5

I have determined a relationship valid under pressure conditions on the Hungarian natural gas transmission network, to calculate the change in transferable energy content due to hydrogen content for a pipeline section assuming a constant pressure loss.

Presentation of the thesis:
In contrast to the distribution network, in the case of a high-pressure transmission network, the value of the compressibility factor (z) should not be neglected when determining the KF conversion factor.

$$\mathbf{KF} = \frac{\mathbf{E_{mix}}}{\mathbf{E_{CH_4}}} = \frac{\mathbf{H_{a_{mix}}}}{\mathbf{H_{a_{CH_4}}}} \cdot \sqrt{\frac{\boldsymbol{\rho}_{\mathbf{rel_{CH_4}}}}{\boldsymbol{\rho}_{\mathbf{rel_{mix}}}} \cdot \frac{\overline{\mathbf{z_{CH_4}}}}{\overline{\mathbf{z_{mix}}}}}$$

Where

- KF – conversion factor [-];
- E_{mix} – energy content of the gas mixture [kWh/m^3];
- E_{CH_4} – energy content of methane [kWh/m^3];
- $H_{a_{mix}}$ – lower heating value of the gas mixture [kWh/m^3];
- $H_{a_{CH_4}}$– lower heating value of methane [kWh/m^3];
- $\rho_{rel_{mix}}$ – relative density of the gas mixture [-];
- $\rho_{rel_{CH_4}}$ – relative density of methane [-];
- $\overline{z_{mix}}$ – average compressibility factor of the gas mixture [-];
- $\overline{z_{CH_4}}$ – average compressibility factor of methane [-].

Furthermore, simulation studies have shown that while the compressibility factor of methane decreases with increasing pressure, hydrogen increases, i.e. the development of the compressibility factor of the methane-hydrogen gas mixture is affected by the opposite behavior of the two components. Another significant difference is that the compressibility factor of hydrogen is almost independent of temperature, the curves corresponding to different temperatures coincide.

Thesis No. 6

Using linear regression, I determined the relationship for calculating the KF conversion factor in the pressure range of 20-75 bar(a) and temperature range of 0-50 °C, 0-100 mol% hydrogen content in case of CH_4-H_2 gas mixture.

Presentation of the thesis:
The KF conversion factor was determined over a wide test range, where the curves could be approximated by a straight line. Based on this, the conversion factor of methane-hydrogen gas mixtures of different composition in the pressure and temperature ranges studied can be determined by the following relationship, which is described using linear regression:

$$\mathbf{KF=\propto \cdot p+\beta}$$

Where

p – average pressure [bar(a)];

α, β – parameters [-].

When applying this equation, it is advisable to substitute the average pressure on the line in order to take into account the average values along the length of the line. The range of absolute error of the equation shall be between 0.3 % and 0.5 % for 20 mol% hydrogen content, with a maximum deviation at 50 mol% H_2 but not exceeding 0.8 %.

a) I defined the parameter ∝ in the context used to calculate the KF conversion factor.

Presentation of the thesis:
Based on the tests, it became apparent that the parameter depends on both temperature and hydrogen content. The curves describing the temperature dependence can be approximated by a quadratic polynomial, so the relationship is as follows:

$$\propto = \boldsymbol{\gamma} \cdot \mathbf{T}^2 + \boldsymbol{\delta} \cdot \mathbf{T} + \boldsymbol{\varepsilon}$$

Where

$\gamma, \delta, \varepsilon$ – parameters expressing temperature dependence [-];

T – temperature [°C].

The hydrogen content-dependent behaviour of the parameters γ, δ and ε describing temperature dependence can be described with sufficient accuracy by a third-degree polynomial:

$$\boldsymbol{\gamma} = -\mathbf{2.847 \cdot 10^{-13} \cdot (H_2)^3 + 5.601 \cdot 10^{-11} \cdot (H_2)^2 - 3.576 \cdot 10^{-9} \cdot (H_2) - 7.357 \cdot 10^{-9}}$$

$$\boldsymbol{\delta} = \mathbf{3.932 \cdot 10^{-11} \cdot (H_2)^3 - 7.951 \cdot 10^{-9} \cdot (H_2)^2 + 5.485 \cdot 10^{-7} \cdot (H_2) + 7.216 \cdot 10^{-7}}$$

$$\boldsymbol{\varepsilon} = -\mathbf{2.280 \cdot 10^{-9} \cdot (H_2)^3 + 5.003 \cdot 10^{-7} \cdot (H_2)^2 - 3.894 \cdot 10^{-5} \cdot (H_2) - 1.865 \cdot 10^{-5}}$$

Where

$\gamma_1, \gamma_2, \gamma_3, \gamma_4, \delta_1, \delta_2, \delta_3, \delta_4, \varepsilon_1, \varepsilon_2, \varepsilon_3, \varepsilon_4$ – parameters expressing hydrogen content [-];

H_2 – hydrogen content [mol%]

b) I defined the parameter β in the context used to calculate the KF conversion factor.

Presentation of the thesis:

Based on the studies, it can be stated that the β parameter is indifferent from temperature, influenced solely by H_2 content. The course of the parameter with a third-degree polynomial can be described with sufficient precision as follows:

$$\boldsymbol{\beta = 5.807 \cdot 10^{-7} \cdot (H_2)^3 - 6.361 \cdot 10^{-5} \cdot (H_2)^2 - 9.050 \cdot 10^{-4} \cdot (H_2) + 9.900 \cdot 10^{-1}}$$

4 Utilisation of the results

During the preparation of my dissertation based on scientific research, I managed to show results in a number of issues that can greatly contribute to the cooperation of the domestic hydrogen and natural gas industry, and can facilitate the elaboration of technical-regulatory issues necessary for the integration of hydrogen produced in Hungary into the natural gas network as soon as possible. The results to be conveyed by the thesis are suitable for solving the technical issues arising during the preparation for the hydrogen natural gas network feed-in, providing a basis for industrial professionals, thus creating the safe and continuous operation of the natural gas network. Furthermore, the research results presented in the thesis may be suitable for the preparation of the technical and regulatory regulations of the Hungarian natural gas industry to be developed against hydrogen, which are still to be elaborated.

The results of my thesis can be used well in the undergraduate and postgraduate education of gas industry professionals, as there is currently a great lack of knowledge about the supply and transportation of hydrogen into the natural gas network, which is currently the basis of many research projects in Europe. Hungary's National Hydrogen Strategy mentions the development of educational programs, the development of teaching materials and/or the adaptation of EU curricula as priority actions regarding the development of knowledge about hydrogen and hydrogen technology. I think my dissertation can help develop these learning materials.

As one of the possible directions for the further development of the scientific results presented by the dissertation, the development of a computer software is offered that gives the maximum amount of hydrogen mixed with natural gas supplied in the given area so that it still meets the Hungarian gas quality requirements.

My further goal was to make the topic understandable for all interested professionals with the help of the displayed results and the related theoretical and literature examples. I hope that with the results presented I have succeeded in arousing the interest of some players in both the natural gas and hydrogen industries.

5 List of related publications in time line

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Notes