THE INFLUENCE OF HYDROGEN CONCENTRATION IN NATURAL GAS ON HEAT FLOWS IN A THERMAL AGGREGATE

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Natural gas is currently a hotly debated topic both from an environmental point of view due to the production of CO_2 and from an economic point of view. The article is focused on the analysis of selected energy factors when burning natural gas with a gradual increase in the hydrogen content in the fuel mixture. In the article, the authors focused on modelling combustion and the influence of hydrogen concentration on heat flows from flue gas in a thermal aggregate. The aim of the authors is to point out changes combustion in the thermal aggregate using document analysis and calculations. By gradually increasing hydrogen in natural gas, it is possible to prove the effect on heat flows during the exchange of heat from the flue gas to the heated material. According to the results, it can be concluded that increasing the content of methane in natural gas changes the nature of the combustion of the mixture. The combustion temperature increases, heat flow density, on the other hand, the volume of flue gas converted to 1 m^3 of fuel decreases. An important factor that positively decreases in this is the CO_2 emission factor.

KEYWORDS

natural gas, hydrogen, combustion, heat flow, flue gas

1 INTRODUCTION

The objectives of the EU Energy Policy clearly set priorities in all areas, both in industry and in the municipal sphere. The next control point is in 2030. An important goal is the reduction of CO_2 emissions and their complete elimination by 2050 [European Parliament 2021]. Reducing emissions is possible based on the modification of existing technologies or innovations to carbon-free technologies. According to the goals of the Energy Policy, the reduction of CO_2 emissions should also be carried out through energy savings, especially in large aggregates such as e.g. also heating furnaces.

A reduced consumption of chemical and electrical energy in the process of the charge heating in the furnace means lower total carbon dioxide emissions that negatively affect the natural environment [Bialik 2022, Gil 2016].

Reduction of energy consumption is currently possible with the help of optimization tools for more efficient use of energy and simultaneous monitoring of produced emissions. Such a tool is also smart systems for monitoring and optimizing the combustion process, as they were dedicated to, for example, authors [Holubčík 2022, Yeromin 2015].

In Slovakia, natural gas is mainly used in the municipal sector for water heating and heating. According to the latest SODB 2021 statistical census, 67.9% of houses in Slovakia are connected to gas and 66.2% of apartments use natural gas as the main source of heating [SODB 2021].

In industry, natural gas is used mainly in large heat aggregates such as heating and melting furnaces [Straka 2019], boilers in heating plants and power plants, etc. However, some heat aggregates also use other technical or synthetic gases. The use of technical gases depends on the technology where such gases can be used, such as in [Gil 2011].

According to the current situation of energy availability for consumers, from the point of view of more efficient use of natural gas, other energy devices are also important, namely cogeneration units, which are also used directly for biogas [Kocanova 2014] or directly in industry [Variny 2022].

Lively discussions are held on various discussion forums about the possibility of replacing natural gas supplied from Russia with other sources. Currently, these are mainly terminals where it is possible to import and gasify liquefied natural gas, which is then distributed to gas networks. These terminals are not yet enough for the EU, so other possibilities are being sought to supplement the missing capacities of natural gas from non-traditional sources.

The main component of natural gas in Slovakia is methane, with a content of more than 90 vol.%. If we look at the technologies where methane is found, it could also be gas obtained from gas degassing from mining activities, where it is possible to obtain more than 70 vol.% of methane in the gas [Skvarekova 2011].

Another option to replace or subsidize natural gas with an alternative is biomethane from biogas stations, where such gas contains up to 96 vol.% methane, which would positively reduce the CO_2 emission factor. Biogas stations would then have to include methane separators, which are well analyzed in [Bobak 2012, Izak 2014]. So far, only one biomethane station has been built in Slovakia in Jelsava [Potocar 2022]. According to expert discussions, other biomethane stations are being prepared. In any case, it can be concluded that there is potential for the construction and operation of such facilities in Slovakia.

In connection with the acquisition of alternative gaseous fuels, research was carried out in the field of underground gasification, which is addressed by the authors in [Kostur 2015, Zelenak 2021].

In connection with the reduction of CO_2 emissions and at the same time with the use of RES, the possibilities of using excess amounts of electrical energy to store this energy in the form of hydrogen are being investigated. One possibility is to use gas networks to transport hydrogen in natural gas as a carrier and recover it in separation units for other uses. Another option is to subsidize existing natural gas from the gas network and mix it with hydrogen for use in existing appliances. In this context, it is necessary to ensure the interchangeability of such a mixture as a substitute for pure natural gas. The interchangeability of gases can be solved especially for devices that can also burn other gases, such as e.g. [Vlcek 2019]. The interchangeability solution is at the expense of the combustion characteristics of the combustible gas mixture created in this way.

Figure 1. Heating chamber furnace with heat input 5x100 kW

For the future use of new technologies with hydrogen, it is necessary to ensure enough sources of this energy carrier, respectively to ensure a sufficient amount of it. The current situation on the energy market in Slovakia and the possible development of future production were described by the authors in [Durcansky 2022].

The radiant energy of the flue gas can be used especially in the combustion chambers of boilers and heating furnaces, where it is possible to define a combustion space such as in Figure 1.

The shapes of the combustion chambers are different and therefore it is necessary to precisely define this specificity in the calculation models where the heat exchange is to be solved.

The problem of using hydrogen has already been solved by several authors, but rather with a focus on the combustion of hydrogen in the fuel mixture in gas turbines or combined cycles such as e.g. authors in [Skordoulias 2022]. When solving combustion and hydrogen production, they used modeling using Aspen Plus software.

In the presented article, the authors focus mainly on the case of heat exchange by radiation, where they mainly focused on the change in emissivity and radiant energy flow from flue gas depending on the increase in the concentration of methane in natural gas. The goal is to prove the effect of the increased concentration of hydrogen in natural gas on the combustion properties of the gas fuel mixture created in this way. The authors chose a calculation method with the aim of creating a mathematical model of the combustion of a gaseous fuel mixture. The goal is to analyse the combustion process using combustion characteristics in heating units.

2 PROPERTIES OF NATURAL GAS

Slovakia operates a gas network that transports and distributes natural gas to end customers. The gas network also includes underground reservoirs, which are filled at the time of low natural gas consumption (summer season) and at the time of increased natural gas consumption (winter season), natural gas is added from them to the gas network. The composition of natural gas depends on the source of gas extraction, i.e. origin of natural gas as well as recovery from underground reservoirs. The costs of such gas are also related to this, as is also pointed out in literature [Liptakova 2021].

CH_4	C_2H_6	C_3H_8	i-C_4H_{10}	n-C_4H_{10}
vol%	vol%	vol%	vol%	vol%
92.7202	3.8488	1.1272	0.1934	0.1934
i-C_5H_{12}	n,neo-C_5H_{12}	C_6H_{14}	CO_2	N_2
vol%	vol%	vol%	vol%	vol%
0.0452	0.0354	0.0567	0.9193	0,8879

Table 1. Natural gas composition (VIII. 2022, [SPP-Distribucia 2022]

Table 1 presents the composition of natural gas (VIII.2022), the composition of which is stated in terms of the obligation of the distributor SPP-Distribucia, a.s. [SPP-Distribucia 2022] publish the average monthly composition of supplied natural gas. The listed composition from Table 1 was used in the calculations in the numerical model.

Relative density	Density	Low heating value	High heating value	High Wobbe´s number	Sulphur content	Emission factor of CO_2
-	kg.m^{-3}	kWh.m^{-3}	kWh.m^{-3}	kWh.m^{-3}	mg.m^{-3}	t_{CO2}/TJ
0.6055	0.7421	9.854	10.912	14.02	0.0412	56.36

Table 2. Natural gas - properties (VIII. 2022, [SPP-Distribucia 2022])

Table 2 is a continuation of Table 1 and lists the average properties of natural gas. The values in Table 2 are given from [SPP-Distribucia 2022] and are recalculated to the business conditions of 15°C and 101325 Pa.

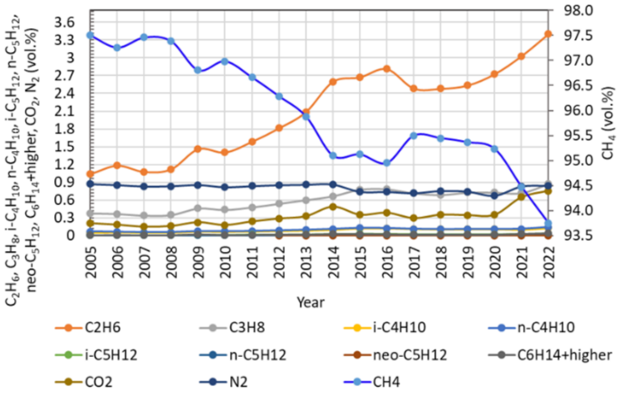

Figure 2. Comparison of average annual concentrations of individual components of natural gas in Slovakia (Adapted from [SPP-Distribucia 2022]

From the available information on [SPP-Distribucia 2022], an analysis of selected parameters of natural gas, which is used in Slovakia distribution network, was carried out. The analysis concerned information on natural gas in the years 2005-20022.

Figure 2 presents the course of the average annual data of the composition of natural gas since 2005.

From the given data, it is possible to observe a decrease in the content of the main component methane CH_4 in natural gas by 3.87% points compared to 2005, which corresponds to a total decrease of 3.97% points. On the other hand, there is a visible increase in heating components, especially ethane C_2H_6 by 242% compared to 2005, which represents a total increase of up to 233.04% points. Another significant change is also with propane C_3H_8, where the content increased by 0.52% compared to 2005, but overall, it represents an increase of 137.79% points. The other heating components have also increased, but they are small changes that do not represent a big value in the difference. An item that is not low heating value, but therefore affects the low heating value itself, is the amount of CO_2, which increased by 0.56% point compared to 2005, which in total represents up to 268.07% points. The graph shows relative changes in the composition of natural gas in the distribution network, which determine the energy properties of natural gas in Slovakia.

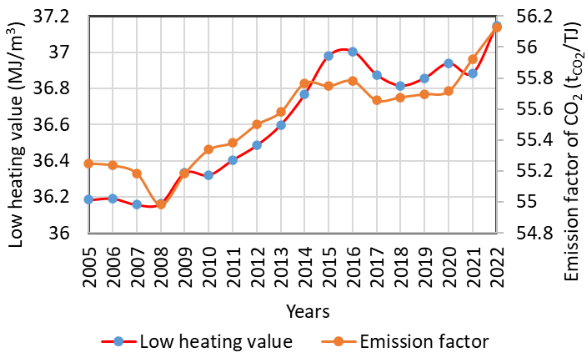

Figure 3. Average annual values of low heating value and emission factor for natural gas in Slovakia for the years 2005-2022 (Adapted from [SPP-Distribucia 2022])

From Figure 3, it is possible to state the average annual increase in low heating value by 0.0534 MJ/m^3 as well as the CO_2 emission factor by 0.048 t_{CO2}/TJ. Until 2007, these values changed minimally, but between 2008 and 2015, the average annual values began to increase for calorific value by 2.25% and the CO_2 emission factor by 1.39%, and from 2015, these average annual values changed only very minimally. Nevertheless, there is a tendency for growth depending on the changes that result from the provision of a enough natural gas from various energy sources.

3 HEAT TRANSFER

In furnace aggregates, heat is transferred by means of convection, conduction and radiation. Several authors agreed that heat transfer in industrial furnaces is carried out mainly by the radiant component of heat. Already in 1919, V.E. Grim-Gržimajlo drew attention to the irregularities in the heat exchange in the furnaces in connection with the removal of flue gas from the furnace. He did not know at that time that at a flue gas temperature above 800°C, heat transfer takes place mostly by radiation from the flue gas and to a lesser extent by convection. It was only in 1924 that Schack [Schack 1924] was the first to draw attention to the large share of the radiant component of heat transfer to the batch in technical fireplaces. The first calculation documents also come from this author. This fact was also supported experimentally in 1939 [Brunklaus 1962]. Using the already known Nusselt formulas for heat transfer by convection, A. Schack recalculated the heat transfer in the hearth of a steam boiler and was able to prove that only a quarter of the heat is transferred by convection and the rest is transferred by radiation. In flue gas heating furnaces, at low temperatures (less than 800°C), the share of gas radiation in heat exchange is small [Schack 1969].

3.1 *Radiant heat transfer*

In most industrial furnaces, the temperature of the flue gas in the furnace space is higher than 800°C, and therefore, in accordance with the findings of A. Schack, the calculation of the radiant component from the flue gas is very important. If we focus on the composition of the flue gas, it can be concluded that the main components are primarily CO_2 and H_2O, which in the heat exchange process can be defined as selectively glowing bodies [Kizek 2021]. Depending on the composition of the fuel, it is possible to add SO_2 to the mentioned glowing components and, in the case of solid fuels, also ash. Most large heating furnaces use gas as fuel and then these 2 components can be neglected.

Because gases emit and absorb heat energy throughout their volume, their ability to emit and absorb this energy will depend on the temperature, but also on the number of molecules through which the heat beam passes. The number of molecules depends on the thickness of the gas layer l_{vp} and on the partial pressure p_p (p_{CO_2}, p_{H_2O}). The emissivity or luminosity of the gas is therefore a function of temperature and the product $p_p.l_{vp}$ [Varga 1999]:

$$\varepsilon_g = f(T_g, p_p.l_{vp}) \tag{1}$$

T_g-thermodynamical gas temperature, (K),
p_p - partial gas pressure, (Pa),
l_{vp} - thickness of the gas layer, (m).

The dimensions of the combustion chamber are important for calculations of radiant energy from flue gases, from which the effective beam length l_{ef} is calculated according to the relation [Kizek 2021]:

$$l_{ef} = \eta.\frac{4.V}{F_V} \tag{2}$$

η- correction coefficient, (0,8 ≈ 0,9),
V- volume filled with glowing gas (flue gases), (m^3),
F_V- the area of the walls delimiting this space, (m^2).

Various mathematical models can be used to calculate flue gas emissivity, which are described e.g. in [Schack 1970] or others, which are listed e.g. in [Pronobis 1993, Kostowski 1987]. For the creation of the model, the approximation model according to E. Kostowski was used, which is described in [Kostowski 1987, Kizek 2021].

The area density of the heat flux by radiation from the flue gas can be calculated using the relation [Varga 1999, Ferstl 2016]:

$$q_r = \varepsilon_{fg}.c_o.\left(\frac{T_{fg}}{100}\right)^4 = c.\left(\frac{T_{fg}}{100}\right)^4 \tag{3}$$

ε_{fg}- Total emissivity of the flue gases [Kizek 2021], (-),
c_o- Absolute black body radiation coefficient = 5.67 W/(m^2.K^4),
T_{fg}- Thermodynamic temperature of flue gases, (K).

In the case of radiation from flue gas to the charge, the previous relation extends to the form [Varga 1999]:

$$q_r = c.\left[\left(\frac{T_{fg}}{100}\right)^4 - \left(\frac{T_m}{100}\right)^4\right] = \alpha_r.\Delta t \tag{4}$$

T_m- Thermodynamic temperature of the heated material, (K),
c- Grey body radiation coefficient, (W/(m^2.K^4)),
α_r- Radiation heat transfer coefficient, (W/(m^2.K),
Δt- The temperature difference of the flue gases and the body, (°C).

3.2 *Convection heat transfer*

The heat transferred by convection to the heated material ranges between 10-30% of the total heat transferred in furnaces with high temperatures. It is customary to sometimes replace this item with a correction factor of 1.1-1.3, which is then multiplied by the radiant component. A more accurate calculation is possible using Newton's relation for the surface heat flux density [Varga 1999, Ferstl 2016, Brodnianska 2018]:

$$q_c = \alpha_c.\Delta t \tag{5}$$

α_c- convection heat transfer coefficient, (W/(m^2.K),
Δt- the temperature difference of the flue gases and the body, (°C).

To determine α_c, it is possible to use the approximate formula recommended by M.A. Micheev for convection heat transfer in furnaces using the dimensionless Nusselt number [Varga 1999]:

$$Nu = \frac{\alpha_c.L}{\lambda_t} \tag{6}$$

L- characteristic dimension, (m),
λ_t- coefficient of thermal conductivity of the liquid, (W/(m.K).

- for laminar mode

$$Nu = 0.57.Re^{0.5} \tag{7}$$

- for turbulent mode

$$Nu = 0.032.Re^{0.8} \tag{8}$$

When creating the model, both components of heat transfer, such as radiation and convection, depending on the flue gas temperature, are considered.

4 NUMERICAL MODEL

The design of the numerical model was based on the parameters of the combustion chamber, which was used for the numerical model of the calculation of flue gas emissivity in [Kizek 2021]. Schematically, the furnace space is shown in Figure 4 and is based on the actual dimensions of the heating furnace in Figure 1.

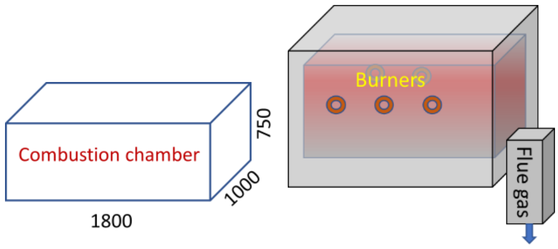

Figure 4. Dimension of the combustion chamber [Kizek 2021]

The mathematical model for the simulation of heat flows from flue gases was based on knowledge obtained during simulations [Kizek 2021]. However, in the new numerical model, the calculation algorithm will be expanded to include items for iterations of combustion temperatures and density of area heat fluxes from flue gases.

The calculation of combustion temperatures is well described in [Varga 1999, Travnicek 2020, Brodnianska 2018], so it will not be mentioned in the text.

Algorithm

The algorithm of the numerical model was based on the architecture of the model for flue gas emissivity calculations, which is described in [Kizek 2021]. The algorithm was extended by additional modules necessary for the calculation of the heat fluxes of flue gas to the lining and the charge and from the flue gas to the charge.

Input: Natural gas composition (Table 1); Excess combustion air m= 1.1; Combustion air temperature t_{air}=20°C; Combustion air Relative humidity = 60%; Barometric pressure = 101,325 Pa; Natural gas temperature t_{fuel}=15°C.

1- Combustion statics; amount of combustion air; volumes and composition of flue gases.

2- Natural gas low heating value Q_n; physical heat of the fuel Q^f_{fuel} and combustion airQ^f_{air};

3- Iterative calculation of adiabatic combustion temperature, t_adiabatic, (°C);

4- Iterative calculation of theoretical combustion temperature, t_theoretical, (°C);

5- Iterative calculation of dissociation combustion temperature, t_dissociation, (°C);

6- Emission flue gases, ε_{fg} [Kizek 2021];

7- Heat flow density - flue gases and flue gases on the charge;

8- Emission factor of CO_2, e_{CO_2} , (t_{CO2})/TJ),

$$e_{CO_2} = \frac{V_{CO_2}}{Q_n} . \rho_{o,CO_2} . 1000 \quad (8)$$

V_{CO_2}- The volume of CO_2 in the flue gas from combustion statics, (m^3/m^3fuel),

Q_n - Low heating value, (MJ/m^3_{fuel}),

ρ_{o,CO_2}- Density of CO_2 under the conditions (0°C; 101,325 Pa), (kg/m^3).

Output: combustion temperatures - adiabatic, theoretical, dissociation, Combustion air volume, Vol.% CO_2 and vol.% H_2O in flue gases, Flue gases volume, Heat flow density- flue gases and flue gases on the charge, emission factor of CO_2, Total input heat Q_n +Q^f_{fuel}+Q^f_{air}.

5 RESULTS

Graphical dependences of combustion temperatures and combustion air were obtained from the simulations, which are shown in Figure 5. The calculations confirmed knowledge from the literature [Varga 1999, Svinolobov 2002, Brodnianska 2018] that the highest combustion temperature is adiabatic, lower is theoretical and the lowest is with dissociation, where the dissociation heat of the flue gas components CO_2 and H_2O is taken into account. From Figure 5, it is possible to observe an increase in combustion temperatures depending on the addition of hydrogen to the mixture with natural gas. Up to a hydrogen content of 50 vol.%, there is an increase in combustion temperatures from 35 to 39.3°C, which corresponds to approximately an increase of 1.93% on average. From a content of 50 vol.% H_2, the increase is more significant, and this is an average of 184°C, which represents 7.32%. Overall, the average combustion temperatures increased by 184.1°C, which represents a 9.38% increase compared to combustion with pure natural gas.

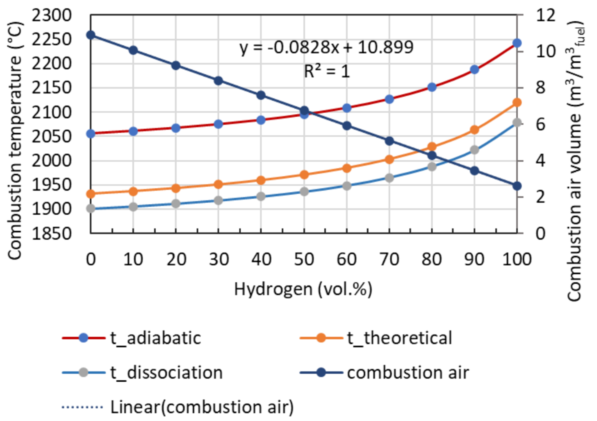

Figure 5. Course of combustion temperatures and amount of combustion air depending on the concentration of hydrogen in the natural gas

On the other hand, there is a visible decrease in the required amount of combustion air by 8.28 m^3/m^3_{fuel}, which represents a total of 75.97%. A linear dependence of the decrease is visible from the graphic display.

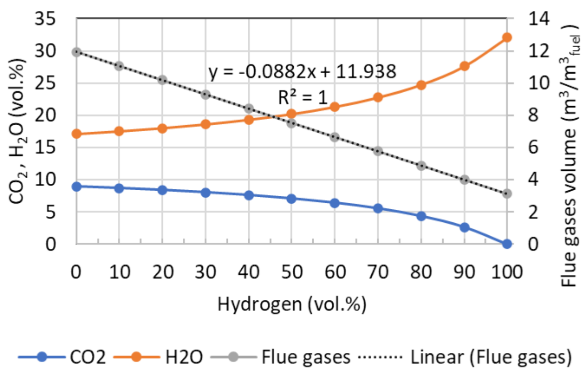

Figure 6. Course of the content of CO_2 and H_2O flue gas components and the amount of actual flue gas depending on the addition of hydrogen to natural gas

From Figure 6, it is possible to observe a non-linear decrease and increase in the CO_2 and H_2O components of the flue gas while maintaining the linear dependence of the decrease in the flue gas volume with increasing hydrogen content in the mixture with natural gas. In the case of CO_2, it can be observed that up to the content of 50 vol.% H_2 there is a smooth decrease of 1.85 vol.% in the flue gas, which represents but 20.72% compared to the initial value. The H_2O component shows the opposite trend. Here, with increasing hydrogen, the content of this component increases, up to 50 vol.% H_2 by 3.1 vol.%, which represents an 18.12% increase compared to the original value. Subsequently, the share of both components changes significantly parabolically. In relation to the total amount of flue gas from 1 m^3 of fuel, a linear dependence of the decrease in the volume of flue gas emerged. In total, the volume of flue gas decreased by 8.82 m^3/m^3_{fuel}, which represents a total decrease of 73.87%. This significant decrease has an effect mainly on heat transfer by convection, where the velocity of flue gases is recalculated in the dimensionless Reynolds criterion (Re).

The obtained results were subsequently used for other modules of the numerical model, where the supporting part was the calculation of heat flow density from flue gases. The obtained calculation results are shown in Figure 7.

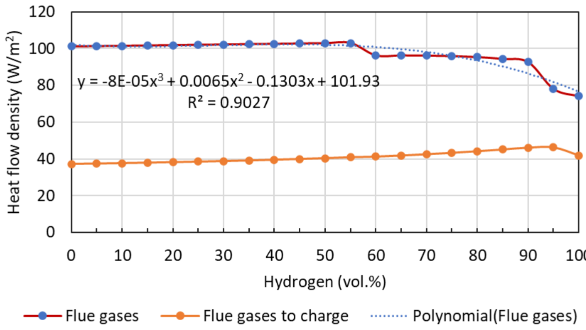

Figure 7. Heat flow density of flue gas

From Figure 7, it can be concluded that the total heat flow density from the flue gas does not change significantly up to the content of 55 vol.% H_2 in the fuel or increases by 1.72%. The decrease in total heat flow density from flue gas occurs only from a content of 60 vol.% H_2 (6.53%), but it is a minimal change to a content of 90 vol.% H_2, by another 3.8%. From 90 vol.% H_2, the total heat flow density from flue gases will be reduced by another 19.91%. The course of values can best be described by a 3rd-order polynomial regression, which is presented in Figure 7. The shown step changes at the hydrogen content of 60 and 95 vol.% are caused by transitions when changing the regressions of the mathematical model according to [Kostowski 1987].

The heat flow from the flue gas to the charge has an increasing tendency up to a content of 95 vol.% H_2, with an increase of up to 24.21% with increasing content of H_2 in the fuel. When the H_2 content is further increased, however, the heat flow density will decrease by 9.47%.

The numerical model also included the calculation of the Emission factor of CO_2, which was obtained from combustion statics and was converted to the calorific value of the fuel.

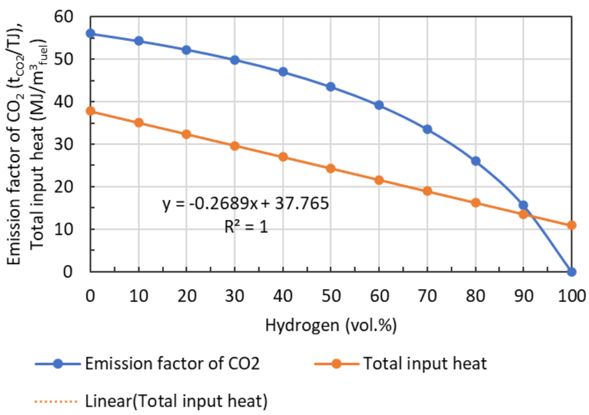

Figure 8. Evolution of the emission factor depending on the addition of hydrogen to natural gas

From Figure 8, it is possible to observe a non-linear decrease of the CO_2 emission factor depending on the addition of hydrogen to the fuel mixture. At 50 vol.% H_2, the drop in the CO_2 emission factor is 22.35% (12.52 t_{CO2}/TJ) and at 60 vol.% H_2 the drop is by another 7.81% (4.37 t_{CO2}/TJ). By further increasing the hydrogen content in natural gas, a significant decrease in the CO_2 emission factor is visible. On the other hand, the total heat, as the sum of the low heating value and the physical heat of fuel and air, shows a linear decrease in relation to 1 m^3 of fuel. The calculations showed that the total physical heat of fuel and air in the results represented from 0.813-0.803% of the total incoming heat. A more significant change would be with higher preheating of the air, i.e. higher physical heat of the combustion air, but that was not the goal of the calculations in this case.

6 CONCLUSIONS

The properties of natural gas can be influenced by mixing with other gaseous fuels while maintaining the required properties. Diversification of sources requires alternative solutions in case of lack of primary source of natural gas. Plans to use renewable energy sources, either in the form of biomethane or in the form of hydrogen as a carbon-free fuel, can fulfil this goal.

The advantage of renewable gas sources is that it can be produced not only in Europe but also directly in Slovakia. The International Energy Agency (IEA) has included this target among the priorities [REPowerEU 2022], the main objective of which is to move away from Russian gas much earlier than 2030.

The carried out numerical simulations proved that when enriching natural gas with hydrogen, it is necessary to monitor the combustion characteristics of the created gas fuel mixture because:

1. Subsidizing natural gas from other and alternative sources affects the overall characteristics of the gas distributed to the gas network. The use of biomethane stations or separation processes from alternative gas sources would reduce the content of higher hydrocarbons and thus could partially reduce the CO_2 emission factor of the mixture thus created.
2. By increasing the hydrogen content in the mixture, the combustion temperature increases, but the required amount of combustion air and thus the total amount of flue gas from 1 m^3 of the fuel mixture decreases. This finding has an impact on the design of the burners as well as on changing the batch heating mode.
3. The areal density of the heat flux from the flue gas changed positively when the hydrogen content in the fuel mixture increased. The increase in the areal density of the heat flow was mainly influenced by the H_2O component in the flue gas up to a content of 95 vol.% hydrogen.
4. In connection with the increase of hydrogen in the fuel, the CO_2 emission factor also decreased non-linearly. This insight, in connection with point 2, proves the suitability of using hydrogen for reducing the produced CO_2 emissions in thermal aggregates.

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