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The Effect of Adding Hydrogen to Natural Gas on Flue Gas Emissivity

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Abstract: The aim of the article is to analyse the impact of adding hydrogen to natural gas. The main goal is to analyze the influence of the increase of hydrogen during the combustion of the gas-air mixture on the emissivity of flue gases in the combustion chamber, applying calculation methods from known relations that can be used in the creation of mathematical models. The created mathematical model simulates the increase in the amount of hydrogen in natural gas. The results are graphical representations of the courses of the selected parameters depending on the increase of the hydrogen content in the mixture. The increase of hydrogen in the fuel mixture confirms a significant effect on the emissivity of exhaust gases, especially from 60% hydrogen content. Moreover, with already 45% of hydrogen in the fuel mixture, the composition and volume of the exhaust gases also changes significantly.

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1 Introduction

In connection with the transition to green energies, the main topics mostly focused on obtaining these energies from renewable energy sources. One of the topics discussed is the issue of obtaining and especially using hydrogen in various processes.

Hydrogen, as the fuel of the future, should serve as a tool for the decarbonization of transport and industry. From the point of view of the industry, it is important to know how to use hydrogen appropriately and especially efficiently in heat aggregates and smaller applications. Adding hydrogen to gaseous fuel is nothing new, and already in the early days of the gas industry, hydrogen was already present in light gas and other technological gases [1,2,3]. The problem was not the incinerators where these gases were burned, but the current smaller or larger heat aggregates and not only in industry but especially in households and the communal sphere. This results from the extent of gasification of the territory of Slovakia, where only natural gas is used to a large extent and it no longer contains hydrogen [4].

When burning natural gas, mainly CO_2 and H_2O emissions are produced, but when burning hydrogen, it is only H_2O [1,2,5,6,7]. From the combustion point of view, the goal of the current EU energy policy [8] is mainly focused on reducing the produced CO_2. However, from the point of view of combustion safety, the rate of hydrogen combustion is disproportionately higher, and therefore much attention is paid to the issue of hydrogen combustion in the fuel mixture.

In industry, large heat aggregates are mostly used, where the size and shape of the combustion chamber is important. The reason is the method of heat transfer to the batch [1,5,9,10].

For illustration, Figure 1 shows a view inside the heating chamber furnace, where burners with a total output of 5x100 kW are installed. With its performance, this furnace provides the power needed to heat the batch even to higher temperatures. At full power, the combustion chamber is completely filled with flue gases. During heat transfer to the batch, both the radiation component of the flue gas and the convective component flowing around the batch are used.

Currently, many scientific teams are dealing with the issue of using hydrogen in heat aggregates and monitoring selected parameters resulting from the work of the heat aggregate. So far, only a slight enrichment of the gas fuel is being considered so that the compatibility of the fuel of the replaceable mixture with the original fuel is maintained.

The authors of the article proposed a numerical model for calculating emissivity based on Kostowski's relations [11,12], where the effect of adding hydrogen to gaseous fuel was monitored. Simulations are performed for the entire range of adding hydrogen to the fuel up to 100% hydrogen as fuel.

Figure 1 Combustion chamber of heating furnace 5x100 kW

2 Methodology

2.1 Heat transfer by radiation

Radiation is one of the most important heat transfer mechanisms in industrial furnaces. No energy transfer medium is required During the transfer of heat by radiation, complex processes change the thermal energy inside the atoms into radiant energy. This energy is further spread by electromagnetic waves, and after impacting other bodies, it changes again into thermal energy. Electromagnetic radiation in the wavelength range of 0.4-40 μm is decisive for heat transfer by radiation. The total radiation consists of [1,2,5,9,13,14]:

a) visible radiation (0.4 – 0.8 μm),
b) infrared radiation (0.8 – 420 μm).

By radiation heat transfer recognized:

- *integral* radiation - in the entire range of wavelengths,
- *monochrome* radiation - in a narrow range of wavelengths.

2.2 Gas radiation

The intensity of gas radiation is not the same for all gases. Triatomic gases radiate in the widest range of wavelengths and with the greatest intensity. For diatomic gases, the range of wavelengths is much narrower, and the radiation intensity of these gases is negligible. In practice, these gases are considered superheated.

Gas radiation is characterized by the following properties:

- radiation and absorption of thermal energy takes place in the entire volume that the given gas fills,
- gases emit and absorb thermal energy only in a certain range of wavelengths,
- radiation and absorption of thermal energy in gases is about 10 times smaller compared to solid bodies,
- finely dispersed solid particles also influence gas radiation.

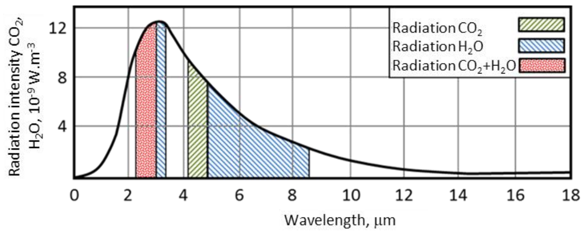

Figure 2 Radiation intensity of CO_2 and H_2O depending on the wavelength [1,15]

From the point of view of heat transfer by radiation, the components of CO_2 and H_2O are the most important. The range of wavelengths in which the emission and absorption of thermal energy of these gases occurs is shown in Figure 2. The wavelength ranges of CO_2 and H_2O overlap, so when these two gases radiate together, the total radiation is less than if they radiate separately [1,2,9,16].

Due to the fact that gases emit and absorb heat energy throughout their volume, their ability to emit and absorb this energy will depend not only on the temperature, but also on the number of molecules that the heat ray passes through during its path.

The number of molecules depends on the thickness of the gas layer and on the partial pressure (CO_2 and H_2O components). The emissivity, or luminosity of the gas is therefore a function of temperature and the product of the partial pressure and the thickness of the gas layer [1,2,9,11,12,15,16].

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

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

The thickness of the radiating layer in different directions of the workspace is different. Therefore, for technical calculations, the term effective beam length was introduced, which is determined according to the relationship [1,2]:

$$l_{ef} = \eta . \frac{4.V}{F_V} \quad (2)$$

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)

To quickly determine the emissivity of flue gases, it is possible to use emissivity graphs for CO_2, H_2O, from which, based on partial pressures and effective beam length, the emissivity of individual glowing gases can be determined as in [1,2,9,11,12].

Currently, however, such a calculation is rather one-time and insufficient for any optimization. When creating mathematical models, various models of approximation of these graphic dependencies are used. In many literatures it is possible to find different approximation models for flue gas emissivity [1,2,11,12,14,16]. The most famous model is the model created by Hottel and Egbert [11,12,16]. Another well-known flue gas emissivity model is from Schack [16]. Further descriptions of the models can be found in [9,11,12,16]

For the calculations, the model for calculating flue gas emissivity according to Kostowski [11,12] was chosen, which is described in the following relations and tables. For individual component in temperature range 200-2000°C and product $p_{p_i}.l_{ef}$ to 200 kPa.m it is true that:

$$\varepsilon_i = 1 - e^{-k_i.\left(p_{p_i}.l_{ef}\right)^n} \quad (3)$$

$$k_i = a + b.\frac{t_{fg}}{1000} \quad (4)$$

ε_i – emissivity of the respective component, (-),
k_i – correction coefficient for the effect of temperature on emissivity, (-),
$p_{p_i}.l_{ef}$ – the product indirectly defines the volume of the radiant component, (Pa.m),
n – exponent obtained from regression analysis, (-),
a, b – coefficients from regression analysis, (-),
t_{fg} – flue gas temperature, (°C).

The values of the coefficients a, b and the exponent n for the respective components of CO_2 and H_2O flue gases are determined depending on the product $p_{p_i}.l_{ef}$ from tables (*Table 1* and *Table 2)*. The radiation of gases is not directly proportional to the fourth power of temperature. For CO_2 is the exponent 3,5 ~ $T^{3,5}$ and for H_2O is the exponent 3 ~ T^3. For calculations, the power is chosen according to the Stefan-Boltzman law - T^4 [1,2].

Table 1 Values of the coefficients a, b and the exponent n for the CO_2 flue gas component depending on $p_{p_i}.l_{ef}$, [11,12]

	a	*b*	*n*
$p_{CO_2}.l_{ef} \leq 0.93$	0.08697	-0.04108	0.614
$0.93 \leq p_{CO_2}.l_{ef} < 4.00$	0.07814	-0.03321	0.391
$4.00 \leq p_{CO_2}.l_{ef} < 70.00$	0.07613	-0.03038	0.374
$10.00 \leq p_{CO_2}.l_{ef} < 70.00$	0.07791	-0.02573	0.314
$p_{CO_2}.l_{ef} \geq 70.00$	0.07350	-0.02081	0.310

Table 2 Values of the coefficients a, b and the exponent n for the H_2O flue gas component depending on $p_{p_i}.l_{ef}$, [11,12]

	a	*b*	*n*
$p_{H_2O}.l_{ef} \leq 0.93$	0.04433	-0.02552	0.945
$0.93 \leq p_{H_2O}.l_{ef} < 4.00$	0.03892	-0.02027	0.814
$4.00 \leq p_{H_2O}.l_{ef} < 70.00$	0.04210	-0.01979	0.692
$10.00 \leq p_{H_2O}.l_{ef} < 70.00$	0.05729	-0.02375	0.530
$p_{H_2O}.l_{ef} \geq 70.00$	0.09700	-0.03809	0.395

The necessary exponent correction is then included in the emissivity. The correction factor β is determined as follows, [1,15]:

If $p_{p_{H_2O}}.l_{ef} > 1\ kPa.m$ (most cases), then:

$$\beta = 1 + \left[0.6225 - 0.1346.\log\left(p_{p_{H_2O}}.l_{ef}\right)\right].\left(\frac{p_{p_{H_2O}}}{100}\right)^{0,86} \quad (5)$$

If $p_{p_{H_2O}}.l_{ef} \leq 1\ kPa.m$, then:

$$\beta = 1 + 0.6225.\left(\frac{p_{p_{H_2O}}}{100}\right)^{0.86} \quad (6)$$

The overlap effects compensate between H_2O and CO_2 bands Δε is determined as:

$$\Delta\varepsilon = \varepsilon_{CO_2}.\varepsilon_{H_2O} \quad (7)$$

For the determination of heat transfer by radiation, it is very important to determine the emissivity of flue gases, which is determined from the following relationship [1,15]:

$$\varepsilon_{fg} = \varepsilon_{CO_2} + \beta.\varepsilon_{H_2O} + \varepsilon_{SO_2} - \Delta\varepsilon \quad (8)$$

In a gaseous fuel such as natural gas, sulphur occurs only in trace amounts, and therefore the glowing SO_2 component can be neglected. In lit. [7,15,17] the authors dealt with the enrichment of combustion air with oxygen and confirmed the increase in emissivity of flue gases with increasing concentration of oxygen in the oxidizing agent. It is caused by an increase in the partial pressures of the components (CO_2 and H_2O) and an increase in temperature.

3 Numerical model

The numerical simulation model of the addition of hydrogen to the mixture with natural gas is designed in such a way that it is possible to obtain results only within the radiation of flue gases. In the calculations, the methodology for calculating the composition of flue gas from lit. [1,2,5,6,18] was used.

In the model, the excess air coefficient m = 1.1 and the oxygen content in the combustion air of 21% were considered.

In heating furnaces, the temperature in the combustion chamber varies within a certain range, and to simplify the simulations, a constant flue gas temperature of 1000°C was determined for the calculation in the combustion chamber.

For the simulation, the simplified dimensions of the combustion chamber were designed based on Figure 1 in the shape of a cuboid and are shown in Figure 3. [3].

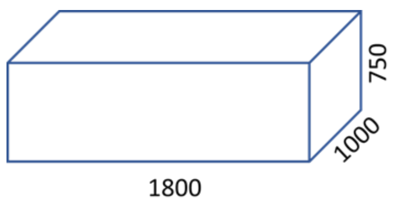

Figure 3 Dimensions of the combustion chamber for calculation in the numerical model.

The mathematical model is composed of several calculation blocks in which the addition of hydrogen to the known composition of gaseous fuel is simulated. Natural gas was used as gas fuel, the composition of which was taken from lit. [4].

Table 3 Natural gas composition (XII. 2021, SPP[4])

CH_4	C_2H_6	C_3H_8	i-C_4H_{10}	n-C_4H_{10}
[vol.%]	[vol.%]	[vol.%]	[vol.%]	[vol.%]
94.4946	3.0131	0.7037	0.1002	0.1117
i,n,neo-C_5H_{12}	C_6H_{14}	CO_2	N_2	
[vol.%]	[vol.%]	[vol.%]	[vol.%]	
0.0466	0.0301	0.6466	0.8533	

3.1 Calculation procedure

Figure 4 shows the development diagram of the simulation model for calculating flue gas emissivity. Individual blocks are subsequently described under Figure 4.

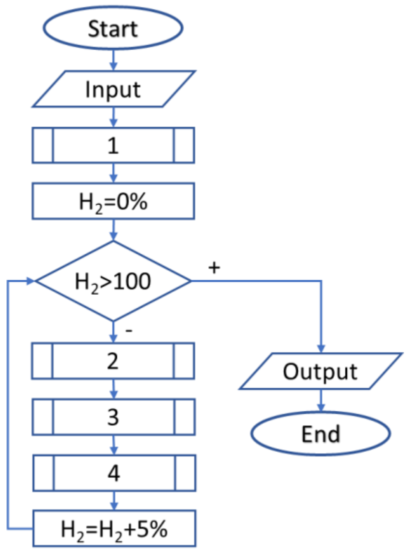

Figure 4 Iterative procedure for emissivity calculation

Input: Loading input data: composition of natural gas, dimensions of the combustion chamber, coefficient of excess air, temperature in the combustion chamber, correction coefficient $\eta = 0.85$

1. Calculation l_{ef}, eq (2)

2. Recalculate fuel gas composition

$$x_1 = \frac{x.(100 - H_2)}{100} \quad (9)$$

3. Calculation combustion statics – CO_2, H_2O, V_{fg}

4. Calculation emissivity ε_{CO_2}, ε_{H_2O}, correlation factor: Δε, β

Output: Output of results to a file

4 Results and discussion

Graphical dependencies were created from the obtained results, which are shown in the following images. Figure 5 shows the effect of adding hydrogen to natural gas on the amount and composition of flue gas, such as the concentration of CO_2 and H_2O. The biggest impact is manifested from 50% hydrogen in the mixture, where with CO_2 the content in the flue gas decreases by 1.861%, which represents a decrease by 20.95% from the original CO_2 content without adding hydrogen. At 60% hydrogen, this is a drop of another 0.666%, which is a drop of another 7.5% from the original CO_2 content.

Up to 50% hydrogen content in the fuel, the trend of decreasing CO_2 content in the flue gas is moderate, but from 50% hydrogen content in the fuel, there is a rapid decrease in the CO_2 content in the flue gas to zero content at 100% hydrogen.

The more dominant glowing component in the flue gas is water vapor H_2O, which increases parabolically with increasing hydrogen content in the fuel. Up to a content of 50% hydrogen in the fuel, the content of the H_2O component will increase by 3.114% of the original content, which represents an increase of 18.11%, and at a content of hydrogen of 60%, it is by another 1.114%, which represents an increase of 6.48% from the original of the H_2O content in the flue gas. In total, the glowing H_2O component in the flue gas increased by 14.865%, which is an increase of 86.44% compared to the initial state

The simulations showed a linear dependence of the decrease in the amount of flue gas. As a result of adding hydrogen to the fuel at a content of 50%, the volume of flue gases decreased by 4.325 m^3/m^3_{fuel}, which represented a decrease of 36.75% from the original volume. The total decrease in the volume of flue gases represents 8.651 m^3/m^3_{fuel}, which is up to 73.5% of the original volume as when burning pure natural gas without added hydrogen.

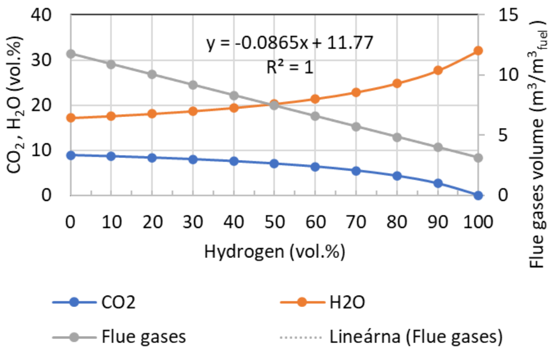

Figure 5 The course of the composition of CO_2 and H_2O in the flue gas as well as the total volume of the flue gas depending on the increasing content of hydrogen in the fuel

Figure 5 shows the composition of flue gas in relation to 1 Nm^3 of gaseous fuel, but in order to maintain the heat input and maintain a constant temperature, the calculations need to be supplemented with additional calculation modules. However, this was not the goal of the authors in this publication.

From Figure 6, it is possible to observe only a small change in the total emissivity of flue gases up to a hydrogen content of 85% in the fuel. The total emissivity increased slightly by 3.33%. From a hydrogen content of 85% in the fuel, the total emissivity began to drop sharply, up to 20.67%. The dominant component for the total emissivity is the H_2O component, which increases slightly parabolically with the increasing proportion of hydrogen in the fuel. Its total increase is nevertheless 23.72% compared to the initial value. Although the radiant H_2O component increased, the total flue gas emissivity was also significantly affected by another CO_2 component. Up to a content of 60% hydrogen in the fuel, the drop in emissivity of the CO_2 component is only 15.37%. With the further addition of hydrogen, the proportion of the glowing CO_2 component in the flue gas starts to decrease significantly until it gradually disappears.

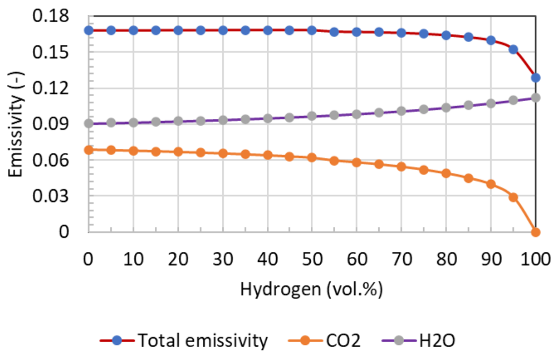

Figure 6 The course of CO_2, H_2O and total flue gas emissivity depending on the increasing hydrogen content in the fuel

From the mentioned simulation results, it is possible to observe the effect of adding hydrogen to the gaseous fuel and how this mixture affects the selected flue gas parameters. The results of the simulations show a minimal effect on flue gas emissivity up to 85% hydrogen content in the fuel. When the hydrogen content is further increased, the character of the flue gas will change significantly and with it the total emissivity of the flue gas.

5 Conclusions

In heat aggregates, heat transfer by radiation is an integral part of the heat transfer from the radiant medium (body) to the charge. In the case of gas-fired heating devices, the radiation from the flue gas produced in the heating process is part of the radiation. Solving the radiant component of the gaseous medium is an important component of all simulation programs, or mathematical models that only solve specific tasks.

The simulation model solved the issue of flue gas emissivity in the case of adding hydrogen to gaseous fuel. The results obtained from the simulation model show how the selected flue gas parameters change depending on the increasing amount of hydrogen in the gaseous fuel.

Due to the simplifications introduced it is not yet possible to assess the impact on the overall heat transfer to the charge or to the combustion chamber as a whole. This will be the subject of analysis using the created additional numerical models.

The created model can be used in the analysis of thermal systems [20] or in post-combustion chambers [21].

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