

Calculation of the main parameters involved in the combustion process of CH₄-H₂ mixtures at different proportions

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Abstract. By setting clear targets for reducing pollutant emissions, the researchers in the field of combustion are pushed lately to find new alternatives for cleaner combustion. The partial or total transition to other types of fuels, such as hydrogen, involves substantial changes in the combustion process and possible necessary constructive changes. In the study of the combustion of CH₄-H₂ mixtures, both numerically and experimentally, preliminary calculations are required, which will help to easily establish the parameters and working regimes and then to use for verifying the results. This paper aims to find an easier method of calculating these parameters, depending on the percentage of gas in the fuel mixture. The calculated values resulted this way will lead to some logical estimates of important aspects of combustion, such as flame field and temperature variation, related to the variation of the amount of hydrogen in the mixture. The method can be a useful tool in the preliminary design of a combustion chamber for CH₄-H₂ mixtures.

1 Introduction

Starting from the environmental problems from the past decades, the scientific world is lately in a constant search for less pollutant and more efficient fuels. Hydrogen is studied as a possible alternative, since new ways for producing and transporting it developed in the past few years [1]. For example, from wind or solar energy, hydrogen can be produced on site by electrolysis and can be transported with the existing natural gas distribution network and then used for different existing or new applications. In the field of energy, it could be interesting to use this new fuel in gas turbines, at industrial level, in order to produce electricity and heat.

The new resulting gas, as a mixture of hydrogen and natural gas, depending on the proportions, can change significantly the combustion parameters of the final application, due to the different mixtures properties. From this perspective, numerical and experimental investigations are needed. Various studies and experiments were made with results pointing

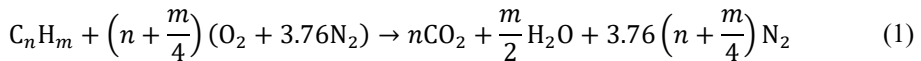
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that above 15-20% H₂, some structural modifications of the equipment are requested [2] [3] [4].

For both numerical simulations and experimental sessions, the preliminary calculations of the input values and nominal regimes are needed. In order to find the main operating parameters of the combustion, formulas and calculations are usually required, based on theory and literature. These calculations and the preliminary results obtained can be later used to define the input and output parameters in the numerical simulations and to establish the operating regimes, air and fuel flows, temperatures, pressures and stability limits in the experiments. The present paper will point below a method to easily calculate the excess air, the calorific value of the fuel mixture, adiabatic temperature and other important parameters in the combustion chamber and the variation of the products in the combustion gases, according to this mixture's different proportions between methane and hydrogen. Hence, the acquired formulas can be used for obtaining useful values that can be used both as starting data and also for verifying the experimental measured results.

2 Excess air - mixture ratio - gas analysis

The combustion process for a C_nH_m, hydrocarbon fuel, as a global stoichiometric reaction using air as an oxidizer, is written as [5] [6]:

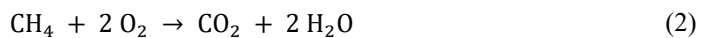


Although the air contains other compounds in small quantities, beside oxygen (20.95%) and nitrogen (78.08%) and other [5], here is simply considered to be composed only of oxygen (21%) and nitrogen (79%). Thus the molar ratio between the two compounds is considered to be 3.76 in the above relationship.

Therefore, for the combustion of each mole of fuel are required $(n+m/4)(1+3.76)$ moles of air, resulting in $4.76(n+m/4)+m/4$ moles of reaction products.

2.1 Combustion of CH₄

The overall reaction for stoichiometric combustion process in the case of CH₄ is written as follows [7]:



The molar mass of CH₄ is 16.043 g/mol . Thus, the fuel / stoichiometric air ratio is:

$$\left(\frac{\dot{m}_a}{\dot{m}_{f(CH_4)}}\right)_s = \frac{2 * \left(32 + \frac{79}{21} * 28\right)}{16.043} = 17.16 = L_{0CH_4} \quad (3)$$

To burn 1 kg of CH₄ requires 17.16 kg of air, an amount known in the literature as L_{minCH_4} , or L_{0CH_4} .

2.2 Combustion of H₂

The overall reaction for stoichiometric combustion in the case of H₂ is:



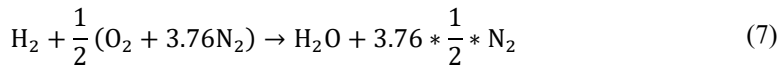
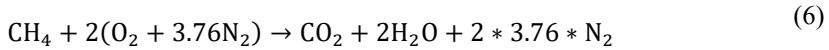
The molar mass of H₂ is 2.016 g/mol. Thus the fuel/stoichiometric air ratio is:

$$\left(\frac{\dot{m}_a}{\dot{m}_{f(H_2)}}\right)_s = \frac{0.5 * (32 + \frac{79}{21} * 28)}{2.016} = 34.32 = L_{0H_2} \quad (5)$$

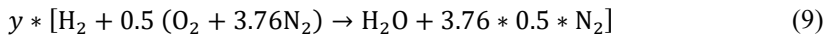
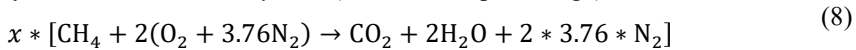
To burn 1 kg of H₂ requires 34.32 kg of air, an amount known in the literature as L_{minH_2} or $L_{O H_2}$.

2.3 Combustion of CH₄ + H₂ mixtures

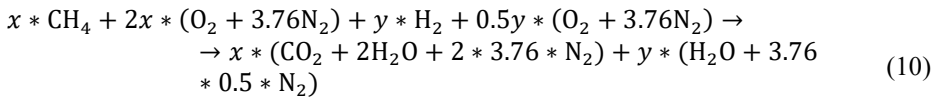
The classical stoichiometric method of calculation is adopted, writing the individual equations for CH₄ and H₂ based on the relation (1): [8]



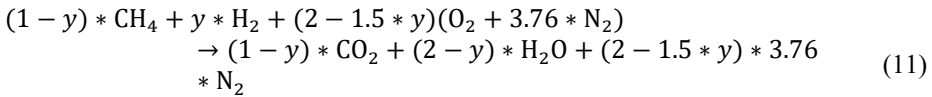
Then the stoichiometric equations are multiplied by the molar fractions of the fuel mixture. For this paper, the following notations and rule are adopted: the molar fraction (or the volume percentage by multiplying by 100) for CH₄ is noted with x , and for H₂ it is noted with y . The condition is $x + y = 1$; (1 = 100% as percentage). In these conditions:



By mixing the above results in the stoichiometric equation of combustion for the mixture CH₄ - H₂ :



By transformations the stoichiometric equation is obtained:



From the relation (11), depending on the molar (volumetric) participations, the stoichiometric air is further defined – L_s [Kmol_{air}/Kmol_{fuel}] ($[m^3_{N_{air}}/ m^3_{N_{fuel}}]$)

$$L_s = \frac{(2 - 1.5y)(1 + 3.76)}{(1 - y) + y} = (2 - 1.5 * y) * 4.76 \quad [m^3_{N_{air}}/ m^3_{N_{fuel}}] \quad (12)$$

The minimum theoretical air required [8] [kg_{air}/kg_{fuel}] :

$$L_0 = L_s * \frac{M_{air}}{M_{fuel}} = \frac{4.76 * (2 - 1.5 * y) * M_{air}}{M_{fuel}} \quad [kg_{air}/kg_{fuel}] \quad (13)$$

Where: $M_{air} = 28.84$ [kg/kmol] is the molar mass of air;

$M_{fuel} = (1-y)*16+y*2$ [kg/kmol] is the molar mass of the fuel mixture

It results:

$$L_0 = \frac{8.5799 * (2 - 1.5 * y)}{1 - 0.875 * y} \quad [\text{kg}_{\text{air}}/\text{kg}_{\text{fuel}}] \quad (14)$$

The excess air is further defined by an excess air coefficient, known also as air–fuel equivalence ratio [5] [9]:

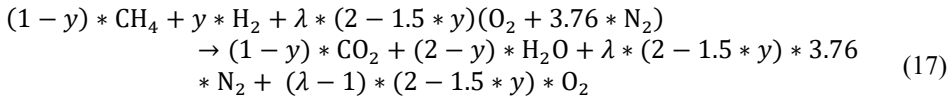
$$\lambda = \frac{\dot{m}_a/\dot{m}_{fuel}}{(\dot{m}_a/\dot{m}_{fuel})_s} = \frac{\dot{m}_a}{\dot{m}_{fuel} * L_0} \quad (15)$$

where: \dot{m}_a - air flow
 \dot{m}_{fuel} - fuel mixture flow

Fuel–air equivalence ratio, is defined as:

$$\phi = \frac{\dot{m}_{comb}/\dot{m}_a}{(\dot{m}_{comb}/\dot{m}_a)_s} = \frac{1}{\lambda} \quad (16)$$

Thus, for lean fuel combustion ($\phi \leq 1$; $\lambda \geq 1$) the relation (11), can be redefined, obtaining the global equation in the form:



Starting from the above, the excess air and dosage are defined (required for comparisons with data from the literature, where dosage is used as a basis):

Stoichiometric dosage:

$$f_0 = \frac{1}{L_0} \quad [\text{kg}_{\text{fuel}}/\text{kg}_{\text{air}}] \quad (18)$$

$$\varphi_0 = \frac{1}{L_s} \quad [\text{m}^3_{\text{fuel}}/\text{m}^3_{\text{air}}] \quad (19)$$

The table below shows the variations of these parameters, depending on the composition of the mixture:

Table 1. Calculated values for various percentages of H₂ in the mixture

H ₂ (y) % vol	H ₂ % mass	M _{fuel}	f ₀ [kg/kg]	L ₀ [kg/kg]	φ ₀ [m ³ _N /m ³ _N]	L _s [m ³ _N /m ³ _N]
0	0.00%	16	0.058	17.160	0.105	9.520
0.2	3.03%	13.2	0.057	17.680	0.124	8.092
0.4	7.69%	10.4	0.054	18.480	0.150	6.664
0.6	15.79%	7.6	0.050	19.869	0.191	5.236
0.8	33.33%	4.8	0.044	22.880	0.263	3.808
1	100.00%	2	0.029	34.320	0.420	2.380

The flue gas analysis results from the relation (17), depending on the volume fraction of H₂ and on the excess air:

$$\text{CO}_2 = \frac{(1 - y)}{(3 - 2y) + \lambda * (2 - 1.5 * y) * 3.76 + (\lambda - 1) * (2 - 1.5 * y)} \quad (20)$$

$$H_2O = \frac{(2 - y)}{(3 - 2y) + \lambda * (2 - 1.5 * y) * 3.76 + (\lambda - 1) * (2 - 1.5 * y)} \quad (21)$$

$$N_2 = \frac{\lambda * (2 - 1.5 * y) * 3.76}{(3 - 2y) + \lambda * (2 - 1.5 * y) * 3.76 + (\lambda - 1) * (2 - 1.5 * y)} \quad (22)$$

$$O_2 = \frac{(\lambda - 1) * (2 - 1.5 * y)}{(3 - 2y) + \lambda * (2 - 1.5 * y) * 3.76 + (\lambda - 1) * (2 - 1.5 * y)} \quad (23)$$

Considering that in practice, when using the gas analyzer, water is condensed on the well path, or at the inlet through the special installation in the analyzer, the analysis of dry flue gases, respectively CO₂, is considered as the main reference element:

$$CO_2 = \frac{(1 - y)}{(1 - y) + \lambda * (2 - 1.5 * y) * 3.76 + (\lambda - 1) * (2 - 1.5 * y)} \quad (24)$$

It results:

$$\lambda = \frac{1 - y + CO_2 - 0.5 * y * CO_2}{4.76 * CO_2 * (2 - 1.5y)} \quad (25)$$

Or by transforming the relationship (24) :

$$CO_2 = \frac{1}{1 + \lambda * (2 + 0.5 * \frac{y}{1 - y}) * 4.76 - (2 + 0.5 * \frac{y}{1 - y})} \quad (26)$$

$$CO_2 = \frac{1}{4.76 * \lambda * (2 + 0.5 * \xi) + 0.5 * \xi - 1} \quad (27)$$

Where: $y/(1-y) = \xi$ represents the molar ratio H₂/CH₄

In this way, it turns out that:

$$\lambda = \frac{\frac{1}{CO_2} + 1 + 0.5 * \xi}{4.76 * (2 + 0.5 * \xi)} \quad (28)$$

These latter relationships will be used during the experiments to determine the excess air, as an additional, and more accurate, verification of the measured CH₄, H₂ and air flow ratios. Based on the measurements recorded with the gas analyzers, obtaining values for CO₂, CO, O₂, the excess air resulting from the measurements can be calculated and then it can be compared as a verification with the calculated one. There are similar formulas obtained and used by the literature in the calculation of excess air based on the concentrations of the components in the flue gases. [10] [6].

3 Adiabatic temperature - mixture ratio

It starts from the relationship known for defining the efficiency of combustion: [11]

$$\zeta * Hi = (1 + \lambda * L_0) * h_3 - \lambda * L_0 * h_2 \quad (29)$$

- where: - ζ – combustion efficiency [%];
 - Hi – lower calorific value (fuel) [kJ/kg];
 - h₃ – flue gas enthalpy [kJ/kg];
 - h₂ – enthalpy of the air at the entrance to the combustion chamber [kJ/kg];

The enthalpy can be defined by the following relationship [12]:

$$\Delta h = C_p * (T - T_{ref}) \quad (30)$$

It was considered that the reference temperature is $T_{ref} = 0^\circ\text{C}$. So, it can be written:

$$\zeta * H_i = (1 + \lambda * L_0) * C_{p_{flgas}} * T_3 - \lambda * L_0 * C_{p_{air}} * T_2 \quad (31)$$

It results:

$$T_3 = \frac{\zeta * H_i + \lambda * L_0 * C_{p_{air}} * T_2}{(1 + \lambda * L_0) * C_{p_{flgas}}} \quad (32)$$

where:

- $C_{p_{air}}$ – average specific heat for air [kJ/kg K]; [8]
- $C_{p_{flgas}}$ - average specific heat for flue gases [kJ/kg K];
- T_3 – the temperature at the outlet of the combustion chamber (flue gases temperature) [K]
- T_2 – the temperature at the inlet of the combustion chamber [K]
- $T_0 = T_{ref}$ – ambient temperature, reference [K]; value is considered $0^\circ\text{C} = 273\text{ K}$
- λ - air–fuel equivalence ratio (equation (15));
- L_0 – the minimum theoretical air required [$\text{kg}_{air}/\text{kg}_{fuel}$] (capitolul 2):
- H_i – lower calorific value [kJ/m_N^3]

For the calculation of the calorific value, it is possible to calculate by volume or by mass.

If we start from the **calorific value by volume**, we can write the relationship [6]:

$$H_{i_v} = (1 - y) * H_{i_{vCH_4}} + y * H_{i_{vH_2}} \quad [\text{kJ}/\text{m}_N^3] \quad (33)$$

where: $H_{i_{vCH_4}}, H_{i_{vH_2}}$ the volumetric calorific values of CH_4 respectively H_2 .

$$H_{i_v} = \frac{H_{i_{vH_2}} * (\frac{H_{i_{vCH_4}}}{H_{i_{vH_2}}} * (1 - y) + y)}{M_{fuel}} * V_{mol} \quad [\text{kJ}/\text{kg}] \quad (34)$$

- where: - $V_{mol} = \text{Molar volume} = 22.4 \text{ m}_N^3/\text{kmol}$
 - $M_{fuel} = (1-y)*16+y*2$ [kg/kmol] is the molar mass of the fuel mixture

Values considered [13]: $H_{i_{vCH_4}} = 35800 \text{ kJ}/\text{m}_N^3$; $H_{i_{vH_2}} = 10700 \text{ kJ}/\text{m}_N^3$;

It results:

$$H_{i_v} = \frac{10700 * (3.345 * (1 - y) + y)}{M_{fuel}} * V_{mol} \quad [\text{kJ}/\text{kg}] \quad (35)$$

After transformations it results:

$$H_{i_v} = 239680 * \frac{[3.345 * (1 - y) + y]}{[(1 - y) * 16 + y * 2]} \quad [\text{kJ}/\text{kg}] \quad (36)$$

If we start from the **calorific value by mass**, the mass fractions are taken into account and thus the following relation results. [6]:

$$H_{i_m} = \frac{(1 - y) * 16}{(1 - y) * 16 + y * 2} * H_{i_{mCH_4}} + \frac{y * 2}{(1 - y) * 16 + y * 2} * H_{i_{mH_2}} \quad [\text{kJ}/\text{kg}] \quad (37)$$

$$Hi_m = Hi_{mCH_4} * \frac{[(1 - y) * 16 + \frac{Hi_{mCH_4}}{Hi_{mH_2}} * y * 2]}{[(1 - y) * 16 + y * 2]} \quad [kJ/kg] \quad (38)$$

The following values are considered: $Hi_{mCH_4}=50000$ kJ/kg; $Hi_{mH_2} = 120000$ kJ/kg (the values presented in the literature differ by +/- 0.5%, preferring these values to simplify the relationship).

Therefore:

$$Hi_m = 50000 * \frac{[(1 - y) * 16 + 2.4 * y * 2]}{[(1 - y) * 16 + y * 2]} \quad [kJ/kg] \quad (39)$$

With the two calculation relations for the calorific values by mass, the differences are:

Table 2. Comparison between the values obtained for the calorific value by mass

	y	0	0.1	0.2	0.4	0.6	0.8	1
Rel (36)	Hi [kJ/kg]	50108	51063	52221	55472	61118	73352	119840
Rel (39)	Hi [kJ/kg]	50000	50959	52121	55385	61053	73333	120000
	Differences [%]	-0.22%	-0.20%	-0.19%	-0.16%	-0.11%	-0.03%	0.13%

Relation (39) was chosen, and the calculations are presented in the following table:

Table 3. Calculated values of calorific value, for different percentages of H₂ in the mixture

Y (molar fraction) (H ₂ % by vol)	0 (0%)	0.1 (10%)	0.2 (20%)	0.4 (40%)	0.6 (60%)	0.8 (80%)	0.9 (90%)	1 (100%)
Hi [kJ/kg]	50000	50959	52121	55385	61053	73333	87059	120000
M _{fuel}	16	14.6	13.2	10.4	7.6	4.8	3.4	2
Mass fraction H ₂ by mass [%]	0	0.0137 1.37%	0.0303 3.03%	0.0769 7.69%	0.1579 15.79%	0.3333 33.33%	0.5294 52.94%	1 100%
Hi increment [%]	0%	2%	4%	11%	22%	47%	74%	140%

It can be observed that over 40% H₂ (y=0.4) the rising of the lower calorific value is increasing, which is also observed in the diagram below, in which the slope of the curve changes essentially after this value. This observation correlates with the reports in the literature, in which experiments say that up to 10-15% by volume H₂, there are not special problems for the combustion process [14]. This should be monitored during experiments. In addition to this, the variation of the fuel mixture's flow for CH₄-H₂ will be considered, which will be adjusted with the change of the percentage of H₂, in order to obtain the same constant thermal power.

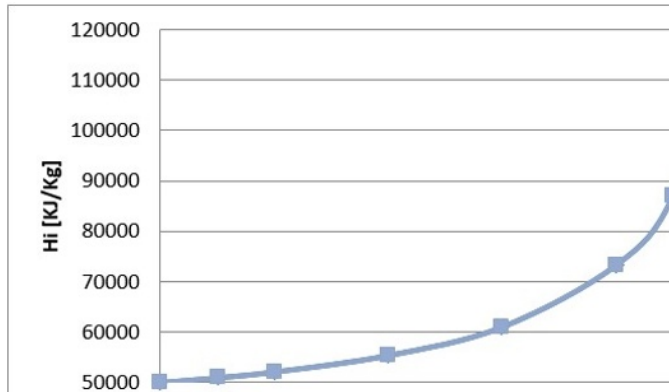


Fig. 1 Variation of the calorific value and of H₂ by mass, depending on the volume fraction of H₂

Finally, for the calculations, the relation (39), will be preferred, which together with the relation (14), by introduction in the relation (32), results:

$$T_3 = \frac{\zeta * 50000 * \frac{[(1 - y) * 16 + 2.4 * y * 2]}{[(1 - y) * 16 + y * 2]} + \lambda * \frac{8.5799 * (2 - 1.5 * y)}{1 - 0.875 * y} * Cp_{air} * T_2}{\left(1 + \lambda * \frac{8.5799 * (2 - 1.5 * y)}{1 - 0.875 * y}\right) * Cp_{flgas}} \quad (40)$$

To determine the specific heats, the literature and the common methods indicates various ways, or charts to calculate. One example is presented and used in the program and calculation procedures for combustion chambers used in 1968 by NASA [15]. After transformations, the specific heats can be written:

$$Cp_{air} = [0.2419 - 0.8181 * 10^{-5} * T + 17.91 * 10^{-9} * T^2 - 2.743 * 10^{-12} * T^3] * 4.186 \quad [\text{kJ/kg K}] \quad (41)$$

$$Cp_{flgas} = \frac{(0.431 + 52.558 * 10^{-5} * T - 38.24 * 10^{-9} * T^2 + 0.25 * 10^{-12} * T^3)}{8.5799 * (2 - 1.5 * y) * \lambda} * \frac{(1 - 0.875 * y)}{8.5799 * (2 - 1.5 * y) * \lambda} \quad (42)$$

The procedure is iterative (2-3 iterations), in which we start from a specific heat value and depending on the resulting temperature, the calculation is repeated with the value of the specific heat corresponding to the temperature resulting in the first iteration. The method is often used in the field, with different calculation relationships [8] [15] [16].

The presented relations do not take into account the thermal dissociation.

In order to make the dissociation correction, we will start from the correction concept according to which the lower calorific value is diminished by the energy lost by dissociation [15] [16]:

$$Hi_{corrected} = Hi - Hi_{dissociation} \quad [\text{kJ/kg}] \quad (43)$$

where $Hi_{dissociation}$ is approximated with the relation [15]:

$$Hi_{dissociation} = \frac{3 * 10^{-26} * T_3^{7.5}}{f} * 2.326 \quad [\text{kJ/kg}] \quad (44)$$

where: f – fuel-air ratio; T_3 – flue gases temperature [K];

After transformations and adaptation to the CH₄-H₂ mixture, the relationship becomes:

$$H_{i\text{dissociation}} = \frac{581 * 10^{-26} * T_3^{7.5} * (1 - 0.875 * y)}{8.5799 * (2 - 1.5 * y) * \lambda} \quad [\text{kJ/kg}] \quad (45)$$

$H_{i\text{dissociation}}$ is shown graphically in the next figure below, in values [kJ/kg]:

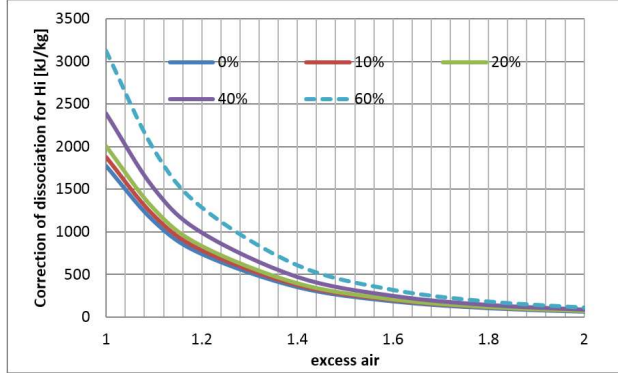


Fig. 2 Corrections of lower calorific value for different values of H_2 %

In the end, the dissociation correction is applied. Calculations can also be done directly taking into account the dissociation correction, the final relationship becoming:

$$T_3 = \frac{\zeta * (50000 * \frac{[(1 - y) * 16 + 2.4 * y * 2]}{[(1 - y) * 16 + y * 2]} - \frac{581 * 10^{-26} * T_3^{7.5} * (1 - 0.875 * y)}{8.5799 * (2 - 1.5 * y) * \lambda})}{(1 + \lambda * \frac{8.5799 * (2 - 1.5 * y)}{1 - 0.875 * y}) * C_{p\text{flgas}}} + \frac{\lambda * \frac{8.5799 * (2 - 1.5 * y)}{1 - 0.875 * y} * C_{p\text{air}} * T_2}{(1 + \lambda * \frac{8.5799 * (2 - 1.5 * y)}{1 - 0.875 * y}) * C_{p\text{flgas}}} \quad [\text{K}] \quad (46)$$

where: T_2 – air temperature; T_3 – adiabatic temperature of combustion (in the equation above, T_3 is iterative, as in the case of specific heats $C_{p\text{flgas}}$).

Results of calculations using these relationships are presented in the following graphs:

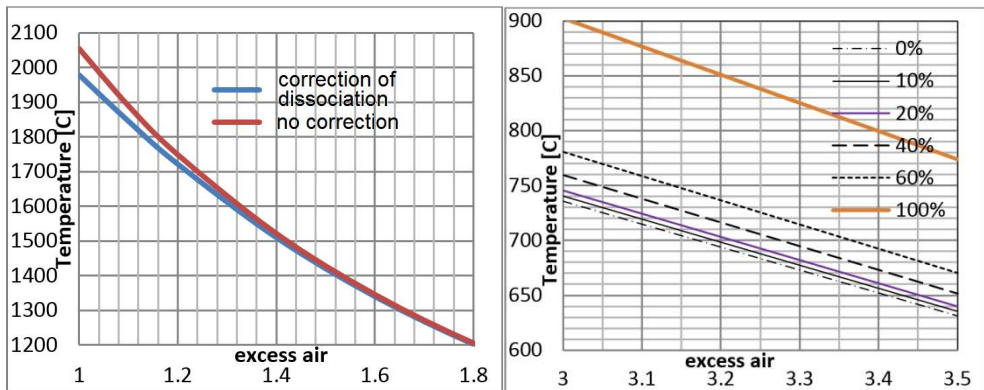


Fig. 3 Corrected combustion temperature
 a) for CH_4 100%, b) for the mixture CH_4-H_2 , at various % of H_2

Fig. 3b shows the combustion temperatures for the mixtures at values of interest for the experimental sessions.

Verification was performed with data from the known literature [15] [16] [17] [18] [19] [20]. The error margin $(T_{calc}-T_{lit})/T_{calc}$ [%] is in between the limits of 1.32 % and -1.27%, the compared cases being methane and pure hydrogen, at different air temperatures. Some examples are given in the table below.

Table 4. Comparison of calculated values with the literature

	y=0	y=1	calculation	Deviation from the literature ($T_{calc}-T_{lit}$)/ T_{calc} [%]	
K	2251	2494			
C	1978	2221			
C	1963	2254	literature	-0.69%	1.32%
C	2004	2210		1.13%	-0.44%
K	2225	-		-1.18%	
K	2223	2483		-1.27%	-0.44%

As it can be seen, the deviations are small and of both positive and negative values, not only in one direction (which would suggest a methodological error).

4 Conclusions

The work here does not aim in this phase to bring special innovations in the field of combustion for CH₄-H₂ mixtures, it starts from known formulas, specific to the physical and chemical processes that take place in the combustion chambers. However, going deeper into the calculation details for the peculiarities of burning CH₄ and H₂ mixtures, new aspects come to light, highlighting specific tendencies that should direct numerical or experimental research for a certain course. For example, a clear picture of the variation in lower calorific value, depending on the variation in the percentage of hydrogen in the fuel mixture, can lead to some logical estimates of important aspects of combustion, such as flame field and temperature variation, related to the variation of the amount of hydrogen.

Moreover, in this paper very useful formulas have been developed for establishing the starting parameters for possible numerical calculations, as well as for establishing operating regimes for experiments. Finally, the data obtained with these calculations can be compared with those obtained by CFD analysis, or from experiments, as a very useful, correct and easy verification.

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