# STUDIES REGARDING THE ENERGETIC PROPERTIES OF HYDROGEN AND METHANE MIXTURES FOR DOMESTIC AND INDUSTRIAL APPLICATIONS

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*One of today's world challenges is represented by the implementation of greenhouse gases free technologies, in which the utilisation of hydrogen as energy vector is an essential element in order to obtain a sustainable global development. Natural gas (methane) represents the most important energy source in Europe, being used in electric energy production, in residential and commercial sectors, and also for transportation. Being responsible for a significant part of the carbon dioxide emissions, the elimination of its utilisation in these sectors represents a major challenge.*

*A viable solution for reducing the dependency on fossil fuels is to use different hydrogen and methane mixtures in energy production, having as result a decrease of greenhouse gases emissions into the atmosphere.*

*This work addresses some issues regarding of transition from the present system that utilises methane gas towards a system completely based on hydrogen. A literature review is done in view of identifying the properties of these mixtures for the characterisation of its combustion in a premixed flame burner. Properties like, laminar burning velocity, the behaviour of adiabatic flame temperature and the concentration limits of flame propagation, are evaluated for future use of hydrogen and methane mixture in the domestic and industrial sector. The evaluation is done by simulating the combustion in a premixed flame burner, providing data about the effect of adding hydrogen into methane.*

**Keywords**: Hydrogen addition in methane flux, Fuel energetic properties, Combustion process characteristics.

## 1. Introduction

The greenhouse gases emissions are caused especially by energy production, transport and utilisation in technological or domestic appliances. With the aim of gradual reduction, until total elimination, of these emissions, a fundamental reorganization of the energetic system is necessary, which must include the entire technological chain, from production until the end user. The natural gas (methane) represents the most important energy source in Europe, being

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used in electrical energy production, in residential and commercial sectors, and also in transports domain. Being responsible for a significant part of the carbon dioxide emissions, elimination of the natural gas from these sectors represents a major challenge [1].

Taking in consideration its very low carbon footprint, the fact that it can be transported, stored and utilised in all the domains that uses methane, hydrogen is the most promising candidate for replacement. Due to the high natural abundance, it represents an energy source with an outstanding potential for development and transformation of the global energetic system. The most important hydrogen source is water, which can be submitted to electrolysis using regenerable energy sources (wind, solar, etc.) [2]. Still, from the safety point of view, it is essential that the hazards associated with the exploitation to be minimized, and the costs associated to this transition to be as low as possible. In reference [3] the experiments have shown that hydrogen is more effective in reducing CO emissions than natural gas. Considering the economic aspects related to the use of hydrogen, it can be specified that due of its very low proportions, will not act as a financial inhibitor.

The transition towards a system that is completely based on hydrogen will be long and will necessitate significant research and development. In this regard, the technologies and the materials that will be used must be adapted to the new type of fuel. The use of hydrogen and methane mixtures significantly increase the thermal efficiency of combustion process, reduces greenhouse gas emissions, and ensures an efficient transition towards a hydrogen based economy [4, 5] Taking into account that the chemical and physical properties of methane and hydrogen are different, increasing hydrogen content into methane gas content will change the characteristics of the new fuel and the combustion process ( combustion temperature, laminar burning velocity, pollutants formation, flammability domain or autoignition temperature). In order to use the current infrastructure for a mixture of methane and hydrogen in different proportions, the hazards for population must be re-evaluated and the safety related aspects. In present, the international standards limit at 30% (volume) the hydrogen quantity that can be introduced in the existent infrastructure [6], for some appliances.

This work addresses some issues regarding of transition from the present system that utilises methane gas towards a system completely based on hydrogen. A literature review is done in view of identifying the properties of these mixtures for the characterisation of its combustion in a premixed flame burner. Properties like, laminar burning velocity, the behaviour of adiabatic flame temperature and the concentration limits of flame propagation, are evaluated for future use of hydrogen and methane mixture in the domestic and industrial sector. The evaluation is done by simulating the combustion in a premixed flame burner, providing data about the effect of adding hydrogen into methane.

## 2. Energetic properties of hydrogen and methane mixtures

Hydrogen and natural gas have different physic-chemical characteristics, like: caloric power, burning and flowing properties, heat characteristics, density, flame propagation velocity and the interaction with the grid. The admixture of hydrogen in the methane flow induces small modifications in the mixture characteristics, but this process must not be avoided, taking into account the advantages that occur from the decarbonization. Hydrogen is considered to be the fuel of the future and it is viewed as a secondary energy that can be obtained from any kind of primary energy: fossil fuels, nuclear fuel, regenerable sources or electricity from the grid. On the other hand, natural gas, composed especially from methane and some other hydrocarbons like ethane, propane and butane in various proportions, and other gases (non-hydrocarbons), because its abundance, is the most utilised fuel [7]. Analysing the physical and chemical properties of the two gases (hydrogen and methane), one can observe challenges that must be approached regarding the utilisation of hydrogen in admixture with methane as fuel by utilising the current infrastructure for methane gas distribution in residential areas.

*Table 1*

**Physical, chemical and energetic properties of hydrogen compared with methane**. [6] [7] [8] [12] [13] [14] [15] [16]

| Characteristics | U.M. | Hydrogen | Methane | Comparison $H_2$ |
|---|---|---|---|---|
| Autoignition temperature | K | 813 | 763 | comparable |
| Minimum ignition temperature | mJ | 0,02 | 0,029 | 1/13 of GN |
| Flammability domain | %vol | 4,0 – 75,0 | 5 – 15 | *6 than GN |
| Laminar flame velocity | cm/s | 270 | 38 | *7 than GN |
| Volume upper calorific power | $MJ/m^3$ | 12,7 | 39,8 | 1/3 of GN |
| Volume lower calorific power | $MJ/m^3$ | 10.64 | 25.32 | 1/2,5 of GN |
| Mass upper calorific power | MJ/kg | 141,8 | 55,5 | *3 than GN |
| Mass lower calorific power | MJ/kg | 119.9 | 50 | *2,5 than GN |
| Flame temperature | K | 2374 | 2219 | 55K over GN |
| Isobar specific heat | kJ/kg*K | 14,29 | 2,232 | *6 than GN |
| Isochore specific heat | kJ/kg*K | 10,16 | 1,709 | *6 than GN |
| Minimum $O_2$ quantity | mol/mol | 0,5 | 2,0 | ¼ of GN |
| Minimum air quantity | mol/mol | 2,38 | 9,52 | ¼ of GN |
| Air-fuel stoichiometry | $Kg_{comb.}/kg_{aer}$ | 0,029 | 0,058 | ½ of GN |

The flammability domain is an important characteristic of these gases, and also of their mixture. Hydrogen is a very flammable gas; it ignites in oxygen at atmospheric pressure at concentrations in the domain between 4.65% – 93.9%, and in air atmosphere between 4% and 75% [9], the explosion domain being between

18.2% - 58.9% in air and between 15% - 90% in oxygen. The lower limit of flammability in air is approximately the same for hydrogen (4%) and methane (5%), while the difference between the upper flammability limit is significantly larger (75% for hydrogen and 14% for methane). For the mixtures of these two gases one can use Le Chatelier's principle to calculate the flammability limits of the mixture [10]. In Table 1 there are shown and compared some of the physical, chemical and energetic properties of hydrogen and methane. We can observe that, from volume point of view, hydrogen necessitates less air than methane for a stoichiometric combustion, whilst from mass point of view, things are opposite. Another effect of the low density of hydrogen can be observed by looking at the calorific power, observing that hydrogen contains a much larger energy quantity than methane per kilogram, whilst things are inverse from volume point of view.

Hydrogen has the autoignition temperature and the thermal conductivity higher than methane [11]. Because the mass of a methane molecule is 8 times bigger than a hydrogen molecule, by burning a mole of methane one can obtain 3 times more heat than burning a mole of hydrogen. From the study of the properties presented in previous table, there can be observed challenges regarding the admixture of hydrogen in the existent infrastructure for natural gas, as follows: mass energy content of hydrogen is larger than methane's, but hydrogen has a lower density, therefore a lower volume energy content, which means that we have to deliver a larger amount of hydrogen in order to obtain the same amount of energy; due to the differences in laminar flame velocities of the two gases, modifications of the faucets and burners will be necessary during transition from methane to pure hydrogen; because hydrogen is a colourless and odourless gas, and its flame is invisible, addition of supplements for elimination of the hazards caused by accidental leaks due to hydrogen higher solubility in metals.

## 3. Energetic properties of hydrogen and methane mixtures in variable proportions

Addition of hydrogen in natural gas flow modifies the combustion characteristics of the fuel, thus the new formed mixture presents, in stoichiometric conditions, extended flammability limits, raised laminar flame velocities [17] and burning temperatures [18], and pollution by emission of greenhouse gases is substantially reduced [17, 19]. In this regard, the study of the fundamental burning properties presents an essential importance in order to ensure a safe transition towards the utilisation of hydrogen in industrial and domestic applications. The main parameters that characterize the combustion process are laminar burning velocity, the behaviour of adiabatic flame temperature and the concentration limits of flame propagation.

For a first theoretical evaluation of the impact of hydrogen admixture on combustion processes designed for natural gas, it is helpful to look at the adiabatic combustion temperatures. This is an important characteristic of the combustion processes, because it can control the velocity of the chemical reaction. This parameter is defined as the maximum theoretical temperature developed in a combustion process which doesn't have any heat losses. In other words, the whole energy released during combustion process is transformed into thermal energy and it is used for the heating of the reaction products, and their temperature represents the adiabatic combustion temperature. This depends both on the concentrations and temperature of a fuel, and on the excess air [8]. On the other hand, it has an important influence on the different forms of burning, like flame propagation velocity, diminishing or even quenching of the flame, and ignition limit [9]. For any combustion process, the involved temperatures have a major impact, in the way that they influence not only the heat transfer and productivity of the process, but also the formation of the pollutants, like nitrogen oxides.

Therefore, it is important to analyse the way that hydrogen impacts the temperatures, even though the real temperatures will be lower than the adiabatic combustion temperature. For the adiabatic combustion temperature of methane, in air at atmospheric pressure, a value of 2220K [22] can be found in literature, while for hydrogen, in the same conditions, is significantly higher, with values between 2400 and 2483K [22, 23]. The new formed hydrogen methane mixtures produce higher burning temperatures throughout the whole domain of air excess. For any type of burning process, one of the most important parameters is the excess air ratio, which is strongly related to the fuel composition. This parameter influences both the temperatures from the burning system, and the energetic efficiency and pollutant emissions.

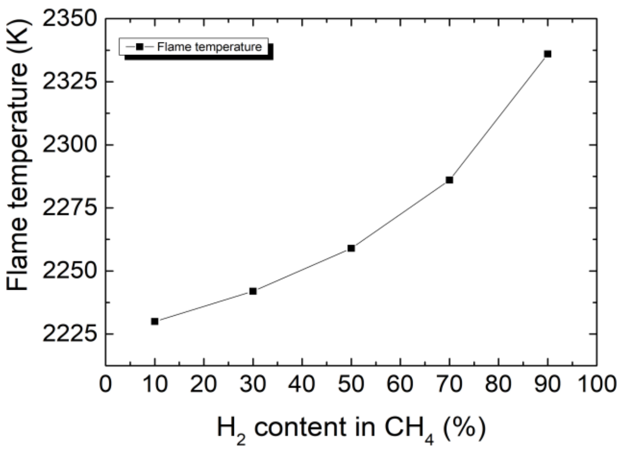



Fig. 1. Adiabatic flame temperature versus $H_2$ content in $CH_4$

For different proportions of hydrogen in methane, the values of the adiabatic burning temperature are plotted in figure 1, while in figure 2 these values are plotted against the equivalence ratio [10].

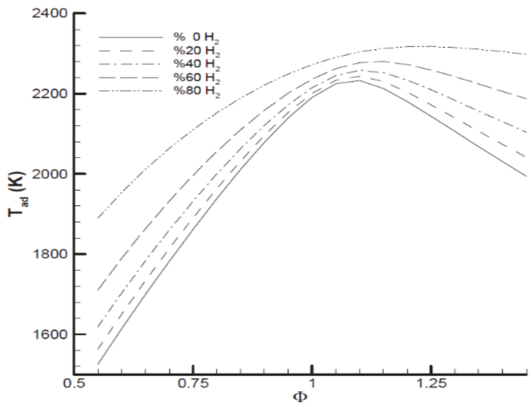



**Fig. 2.** The effect of hydrogen addition on adiabatic flame temperature [11]

We can observe an increase in the value of the adiabatic burning temperature from 2230K o 2336K with the increasing of the hydrogen concentration in methane, while pure methane's adiabatic burning temperature is 2226K [24].

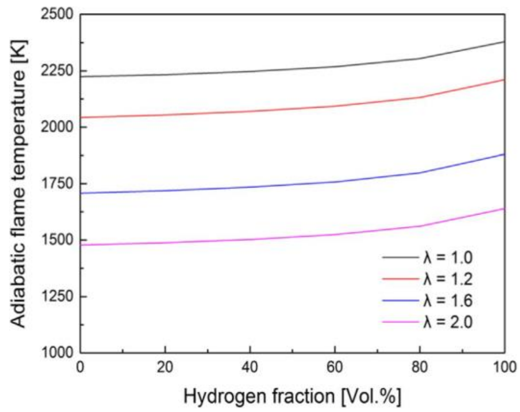



Fig. 3. Variations of adiabatic flame temperature in air as a function of excess air ratio and hydrogen fraction

This behaviour of adiabatic flame temperature, in the same conditions of stoichiometric fuel-air mixture, it is reported in the article of Boushaki et al. [11] where the adiabatic flame temperature increase with 10K by adding 20% hydrogen, while for 50% Hydrogen in methane this value was 33K. In figure 3. a similar behaviour can be observed by decreasing of λ the adiabatic flame temperature is increasing [12]. Figure 4. compares the adiabatic combustion temperatures of 50% $CH_4$ and 50% $H_2$, pure hydrogen and pure methane over a wide range of air excess ratios. Hydrogen and methane / hydrogen mixture produce higher combustion temperatures over the entire λ-range, although the temperature increase from methane to the blend of 50% methane and 50% hydrogen is relatively moderate (about 30°C at λ = 1). The difference between adiabatic combustion temperatures of methane and pure hydrogen, however, is significantly higher (more than 150 °C). [13]

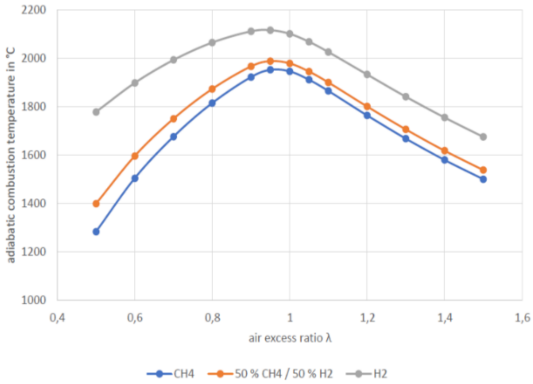



Fig. 4. Adiabatic combustion temperatures of $CH_4$, 50% $CH_4$/50% $H_2$ and $H_2$ as functions of the air excess ratio, with air as oxidizer [14]

In table 1 we can observe that hydrogen necessitates, for a complete burn, less air, in comparation with methane, meaning that any hydrogen-methane mixture will require less air per fuel molar unit than pre methane. Taking into account only the chemical reaction of the burning process, the hydrogen addition in methane flux will result in a change of excess air ratio. This observation represents an advantage of hydrogen-methane mixtures utilisation in both residential and commercial sectors, where there is no control on the burning process parameters like the air flux or pressure.

Most of the residential heating devices use premixed burners in order to generate heat, meaning fuel and air are completely mixed before introduction in the combustion room. Thus, the laminar combustion velocity plays an important role for flame stability and shape burners. In figures 5 and 6 the dependence of methane - air and hydrogen–air mixtures laminar burning velocity under normal conditions on the equivalence ratio (1/λ) is presented. From the experimental data and numerical simulations, it was observed that hydrogen has a significantly higher laminar combustion velocity over the entire range of equivalence ratios. Also, we can notice that the maximum laminar burning velocity is achieved at slightly sub-stoichiometric conditions for methane, while for pure hydrogen this maximum is shifted significantly towards sub-stoichiometric regimes. [14]

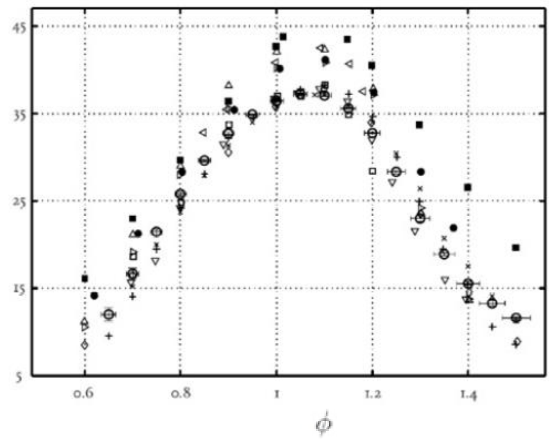



Fig. 5. Laminar combustion velocities of $CH_2$ [15]

The differences in the x-axis show that hydrogen has a flammability domain over the range of equivalent ratios that represent an advantage of using it in comparison with methane. The obtained values of laminar burning velocity for hydrogen–air is mixtures are in the range from 260 to 350 cm/s, while for methane-air mixture is 36 cm/s. [14]

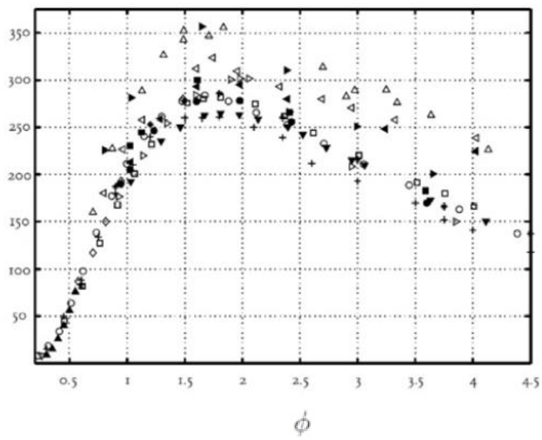



Fig. 6. Laminar combustion velocities of $H_2$ [15]

Therefore, the laminar burning velocity of hydrogen - air mixture is from 7 to 9 times higher than the corresponding value for methane–air mixtures under normal conditions, that represent the main differences in the combustion processes of hydrogen and methane. In figures 7 and 8 the laminar flame velocity of hydrogen

- methane–air mixtures were determined, using the heat flux method, under normal conditions and the equivalence ratio from 0.6 to 1.5 with additions of hydrogen up to 40%. The results for all the mixtures show the same behaviour, namely the addition of hydrogen increases the normal flame velocity. [14, 15]

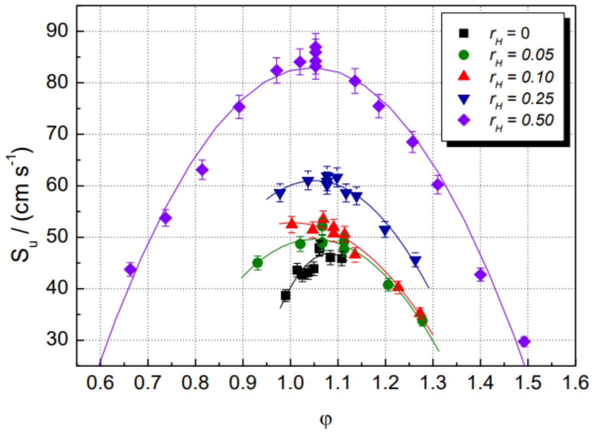



Fig. 7. Laminar burning velocities for $CH_4$–$H_2$–air mixture at ambient initial pressure and temperature [17]

In addition, in references [15, 16] the variation of laminar burning velocity as function of the hydrogen content follows a linear trend in the range up to 40%.

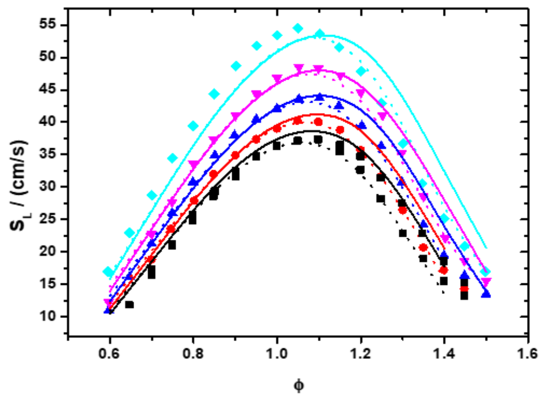



Fig. 8. Laminar flame speeds for 100% $CH_4$/air at 1 atm and 298 K
■100%$CH_4$, ●90%$CH_4$/10%$H_2$, ▲80%$CH_4$/20%$H_2$, ▼70%$CH_4$/30%$H_2$, ♦ 60% $CH_4$/40%$H_2$ [15]

In references [17, 18, 19], the experimental values of the normal velocity of the laminar flame of methane–hydrogen–air mixtures were compared with the values obtained by simulation. The simulation was carried out using kinetic mechanisms, GRI-Mech 3.0 and Konnov [20], which showed good agreement with experimental results, especially for the Konnov mechanism, and also a comparative study between OpenFOAM and Ansys Fluent [19].

Another advantage of hydrogen addition, up to 40%, in methane flux is represented by the diminished effect on the upper and lower flammability limits. This allows such mixtures to be used on the same equipment and under the same safety regulations that have long been well established for working with natural gas. It is also necessary to take into account that H2 enrichment of $CH_4$/air premixed flames affects the flow field in both quantitative and qualitative terms. With the increase of the hydrogen content in the methane flux, the upper flammability limit increases. This means that the flammable range of the new fuel (hydrogen - methane - air mixtures) is extended when there is more hydrogen inside the fuel. On the other hand, changing the hydrogen content has a limited influence on the value of the lower flammability limit. [21]

In conclusion, methane-hydrogen mixture can be used as an alternative energy source in different applications that use fossil fuels. Thereby, the properties of hydrogen-methane mixtures are important parameters in the design of hydrogen-methane premixed burners. When hydrogen is blended with methane, the new fuel formed present different properties compared with those of pure methane or pure hydrogen. The density of the new fuel is lower than pure methane, and one of the most important advantages of the density difference is an increase in gas leakage volumetric flow rate.

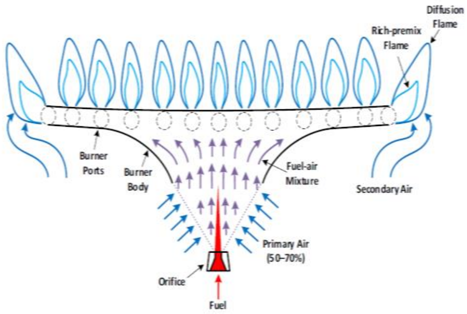
The hydrogen addition in methane flux, represent a transition step in the process of replacement of fossil fuels and could have immediate applications. Principally, the hydrogen produced from renewable energy sources (wind, solar, hydro, biomass) increases the calorific value to the existing energy supply. Another impact of hydrogen using is the reduces greenhouse gas emissions. In reference [22], is analyzed the combustion of various types of solid biomass like sawdust, chopped wood, straw briquette, ropes of wine, cobs corn, and energy willow with and without hydrogen enriched gas. The experimental results reveal the reduction by 40% of SO2 concentration, while the NOx concentration increased by 10%, and leads to the conclusion that by injecting hydrogen in the primary air in the co-combustion process of biomass, the flame temperature increased by 10%, the CO concentration decreased by 60e80% for specific ratio 15 L/kg, and the combustion efficiency raised by 2 - 4%. There are several challenges that need to be managed when a burner designed for methane is supplied with a hydrogen-methane mixture. The higher the hydrogen content in the mixture, the higher the laminar burning velocity increases. However, at the same time, it can alter the ratio if there is no control system. Additionally, the adiabatic flame temperature also increases, and in the case of a lean burn process

($\lambda > 1$), these effects tend to cancel each other out, meaning that an increase in the air level leads to a decrease in the laminar burning velocity and temperature. Under sub-stoichiometric conditions ($\lambda < 1$), due to the accumulation of unburned hydrogen, the laminar burning velocity tends to increase.

## 4. Simulation of a premixed combustion burner using $H_2$+$CH_4$ mixtures

In this chapter, a series of simulations were conducted regarding the addition of hydrogen to the methane flux for use in traditional household burners. Since the intention is to use the existing infrastructure for $H_2$+$CH_4$ mixtures and to maintain explosion risk at an acceptable level [23, 24], the simulations were carried out for hydrogen concentrations of up to 20% in methane. Through these simulations, various parameters will be determined related to flame characterization and the resulting products of the combustion process, such as CO, CO2, Nox. Figure 9 illustrates the operation of a classic household burner. The simulation aims to reproduce combustion through a slot of the classic household burner, taking into account the assumption that the air-fuel mixture is homogeneous and evenly distributed to all slots of the burner.

a) Actual view of a burner




b) Schematic illustrations of a burner [42]

Fig. 9. Classic household burners

A 4.5 kW burner (calculated with $CH_4$) with 20 slots was considered, supplied with 0.45 m3/h of fuel at a pressure of 20 mbar. This burner belongs to the category of self-aspiration burners, where the air required for combustion enters the burner body due to the pressure drop created by the fuel jet, inducing an air flow. As a result, the combustion is characterized as partially premixed, with up to 70% of the air required for complete combustion being entrained by the fuel towards the base of the flame, while the remaining air is drawn from outside the burner body.

**Computational domain and mesh**: The combustion simulation within such a burner was realised using Fluent 2D for a single slot (3 x 1 mm), supplied with 0.2 m3/h of fuel at a pressure of 20 mbar, taking into account the assumption that the air-fuel mixture is homogeneous and uniform distributed to all burner slots. Figures 11 and 12 represents the computational domain and the 2D mesh, that are relevant for the obtaining the results.

## 5. Results and discussions

The simulation foregoes an experimental regarding the investigation of the combustion of $CH_4$ and $H_2$ mixtures in existing household burners in order to identify the possibility of using the existing infrastructure at the time of $H_2$ addition.

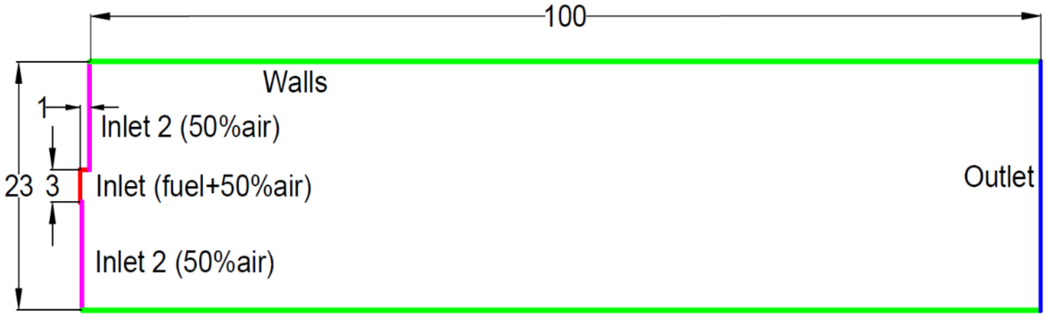



Fig. 10. Computational domain and dimensions

The laminar burning velocity and temperature flame (figures 12 and 13)

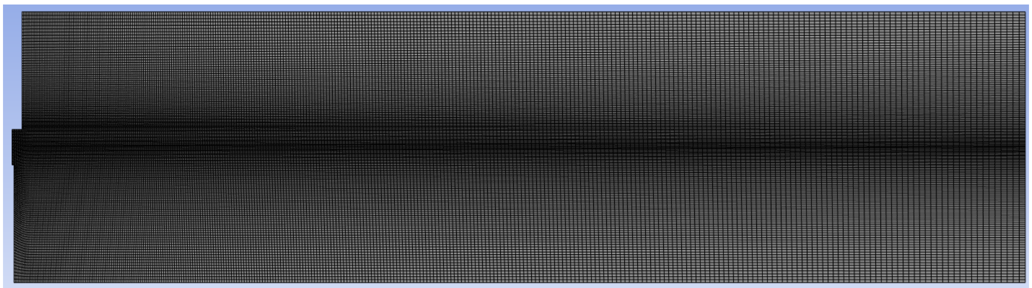


Fig. 11. Visualisation of the two-dimensional mesh

were determined for different concentrations of $H_2$ in $CH_4$ (0 - 30% by mass). Both temperature and velocity increase with the mass concentration of $H_2$ in the mixture.

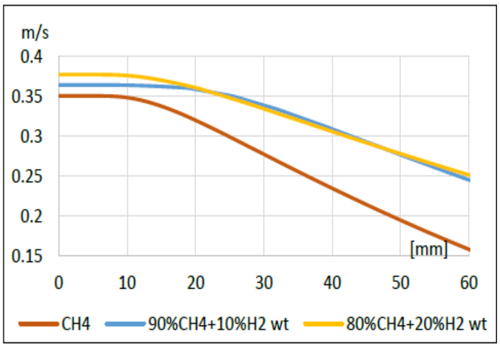



Fig. 12. Laminar burning velocity of different CH4 and H2 mixtures

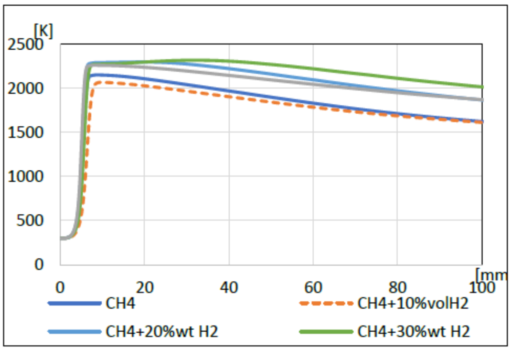



Fig. 13. Flame temperature of different CH4 and H2 mixtures

As the temperature increasing at higher $H_2$ concentrations, NOx emissions also increase (figure 14). However, they can be reduced by increasing the excess air coefficient, while NOx decreases.

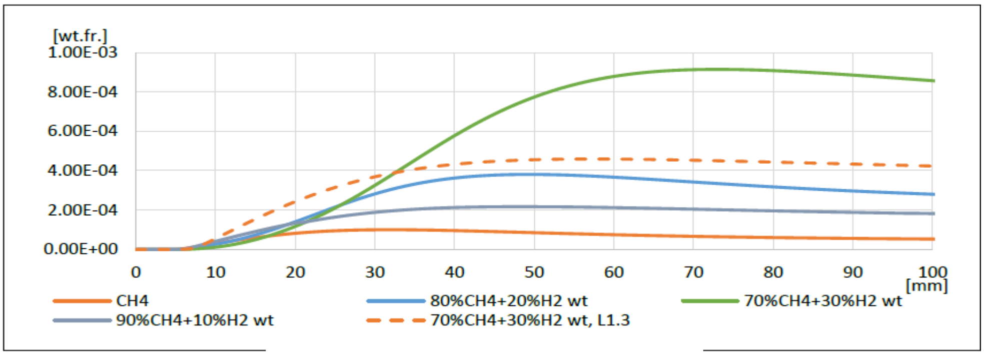



Fig. 14. Mass fraction of NOx

In the present simulation conditions, the species obtained from the combustion of $O_2$, $CO_2$, $H_2O$ were also obtained and can be seen in figures 15 and 16).

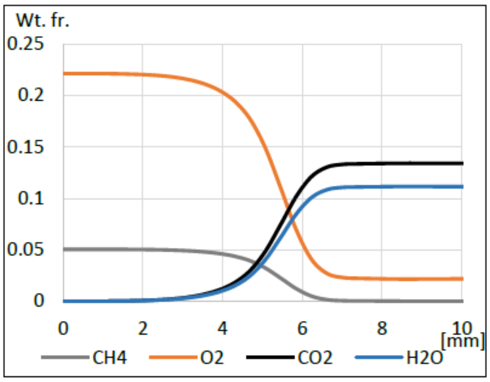



Fig. 15. The resulted species after combustion 100%$CH_4$

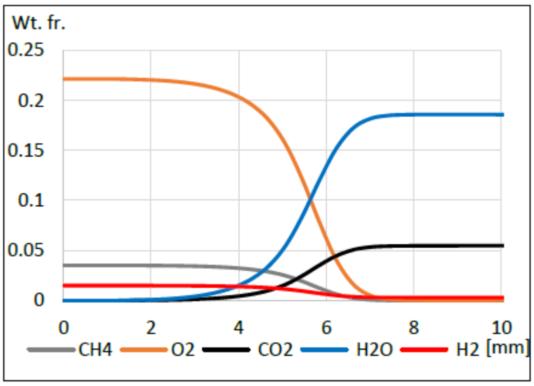



Fig. 16. The resulted species after combustion 70%$CH_4$+30%$H_2$wt.

$CO_2$ decreases with the addition of $H_2$ in the mixture, while the concentration of $H_2O$ increases.

## 6. Conclusions

Using hydrogen as a "vector energetic" offers numerous advantages but involves several scientific and technical challenges, especially concerning its storage for later use. Hydrogen plays a role in both nuclear fission, tidal energy, wind energy, hydropower, solar energy, and nuclear fusion reactions, and its use as an energy source has the advantage of eliminating harmful emissions. In principle, using hydrogen could address the global issue of the greenhouse effect. From the study of the physical and chemical properties presented above, challenges related to injecting hydrogen into the existing methane infrastructure become apparent:

1. Higher Mass Energy Content: Hydrogen has a higher mass energy content than methane. However, hydrogen has a lower density, resulting in a lower volumetric energy content. This means that a larger quantity of hydrogen must be delivered to provide the same amount of energy.
2. Differences in Flame Speed: Due to differences in laminar burning velocities between the two gases, modifications to valve and burner designs will be necessary when transitioning from using methane to 100% hydrogen.
3. Safety Concerns: Hydrogen is colourless, odourless, and its flame is invisible. This necessitates the addition of additives to mitigate the risks of undetected accidental leaks. Hydrogen is more soluble in materials, so leak rates in distribution systems are expected to be higher than those of methane. Safety considerations take precedence over economic concerns in this context.
4. Addressing these challenges will be essential for a successful transition to hydrogen as an energy carrier and for realizing its potential benefits in reducing greenhouse gas emissions and advancing clean energy technologies.

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