Testing **Hy**drogen admixture for **G**as **A**pplications

Impact of hydrogen admixture on combustion processes – Part I: Theory

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Motivation and scope

Climate change is one of today's most pressing global challenges. Since the emission of greenhouse gases is often closely related to the use and supply of energy, the goal to avoid emissions requires a fundamental restructuring of the energy system including all parts of the technology chains from production to end-use. Natural gas is today one of the most important primary energy sources in Europe, with utilization ranging from power generation and industry to appliances in the residential and commercial sector as well as mobility. As natural gas is a fossil fuel, gas utilization is thus responsible for significant emissions of carbon dioxide (CO_2), a greenhouse gas. However, the transformation of the gas sector, with its broad variety of technologies and end-use applications is a challenge, as a fuel switch is related to changing physical properties.

Today, the residential and commercial sector is the biggest end user sector for natural gas in the EU, both in terms of consumption and in the number of installed appliances. Natural gas is used to provide space heating as well as hot water and is used in cooking and catering appliances, with in total about 200 million gas-fired residential and commercial end user appliances installed. More than 40 % of the EU gas consumption is accounted for by the residential and commercial sector.

The most promising substitutes for natural gas are biogases and hydrogen. The carbon-free fuel gas hydrogen may be produced e.g. from water and renewable electricity; therefore, it can be produced with a greatly lowered carbon footprint and on a very large scale. As a gaseous fuel, it can be transported, stored, and utilised in all end-use sectors that are served by natural gas today: Power plants, industry, commercial appliances, households, and mobility.

Technologies and materials however need to be suitable for the new fuel. The injection of hydrogen into existing gas distribution for example will impact all gas-using equipment in the grids, since these devices are designed and optimized to operate safely, efficiently and with low pollutant emissions with natural gas as fuel. The **THyGA project**[1] focusses on all technical aspects and the regulatory framework concerning the potential operation of domestic and commercial end user appliances with hydrogen / natural gas blends. The THyGA deliverables start with theoretical background from material science (D2.4) and combustion theory (this report), and extend to the project's experimental campaign on hydrogen tolerance tests as well as reports on the status quo and potential future developments on rules and standards as well as mitigation strategies for coping with high levels of hydrogen admixture. By this approach, the project aims at investigating which levels of hydrogen blending impact the various appliance technologies to which extent and to identify the regime in which a safe, efficient, and low-polluting operation is possible.

As this is in many ways a question of combustion, **this report** focuses on theoretical considerations about the impact of hydrogen admixture on combustion processes. The effects of hydrogen admixture on main gas quality properties as well as combustion temperatures, laminar combustion velocities, pollutant formation (CO, NO_x), safety-related aspects, and the impact of combustion control are discussed. This overview provides a basis for subsequent steps of the project, e.g. for establishing the testing program. A profound understanding of the impact of hydrogen on natural gas combustion is also essential for the development of mitigation strategies to reduce potential negative consequences of hydrogen admixture on appliances.

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[1] For more information, please visit https://thyga-project.eu/

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List of abbreviations

Air_{min}	minimum air requirement
α	thermal diffusivity
CARI	Combustion Air Index Requirement Index
CHP	combined heat and power
CH_4	methane
CO	carbon monoxide
CO_2	carbon dioxide
d	relative density
d_q	quenching diameter
GCV	gross calorific value
GRI	Gas Research Institute
H_2	hydrogen
H_S	higher heating value, gross calorific value
K_G	deflagration index
λ	air excess ratio
LEL	lower explosion limit
LOC	limiting oxygen concentration
MN	Methane Number
N_2	nitrogen
NCV	lower heating value, net calorific value
O_2	oxygen
Φ	equivalence ratio (= $1/\lambda$)
p	pressure
P	firing rate
PV	photovoltaics
ρ	density
s_L	laminar combustion velocity
T	temperature
UEL	upper explosion limit
u	flow velocity
V	volume
$\dot{V}$	volume flow
W_S	(superior) Wobbe Index, gross Wobbe Index

1 Introduction

Natural gas is today one of the most important primary energy sources in Europe, with combustion processes representing the dominant utilization path. Although natural gas also serves as a feedstock for a wide variety of products in the chemical industry (as well as in some metallurgical applications), the vast majority of the natural gas being distributed today is burned to provide heat, electricity or propulsion across many different sectors, from residential appliances to industrial furnaces, power plants or in natural gas vehicles. While combustion is used in different ways in these sectors, tailored to the specific application, the basic principles are the same.

The combustion of any hydrocarbon fuel will release carbon dioxide (CO_2), which has been identified as a greenhouse gas. While natural gas will form far less CO_2 per energy unit than other fuels such as coal or oil, measures must be taken to reduce the overall impact of gas combustion on the climate. Hydrogen admixture into the natural gas grids is being discussed as one potential way to reduce CO_2 emissions from natural gas combustion, across all sectors. Hydrogen admixture into the gas grids can also serve to store surplus renewable energy from wind turbines or PV panels in times of low power demand. Energy storage is one of the major challenges of the energy transition to renewable energies. While storing electricity is strongly limited in terms of storage time and scale, the long-term storage of energy in chemical form, i.e. as a fuel, is technically feasible on the large scale. Thus, power-to-gas can be a powerful tool to implement the storage of renewable energy, reduce curtailments of renewable power sources and thus make the existing gas grids a key component of a future low-carbon energy infrastructure.

From an end-use perspective, the admixture of hydrogen to natural gas means that the fuel properties and combustion characteristics of the new fuel (now a blend of natural gas and hydrogen) change, compared to “pure” natural gas. Considering that the chemical and physical properties of methane (the main component of natural gas) and hydrogen are significantly different, increasing hydrogen admixture into natural gas will lead to changing fuel characteristics and thus to changes in the combustion process.

Practically all residential and commercial gas appliances installed within the EU today were designed for operation with natural gas. This is also true for larger equipment in industry and power generation. Therefore, the impact of hydrogen admixture on these combustion processes will be an important criterion to assess the viability of feeding hydrogen into the natural gas grids. The residential and commercial sector, with an estimated population of about 200 million appliances within the EU [1], plays a major role in this discussion.

2 Basic combustion and gas quality characteristics

In a combustion process, a fuel (e. g. natural gas or hydrogen) and an oxidizer (usually air) react with one another to form reaction products. In this process, some of the energy chemically bound in the fuel is released as heat.

For the moment, this section will focus on a comparison of methane (CH_4, representing natural gas), hydrogen (H_2) and methane/hydrogen blends. While actually distributed natural gas contains other species such as higher hydrocarbons, carbon dioxide (CO_2) or nitrogen (N_2), methane is often used as a reference gas, since natural gas usually contains at least 90 vol.-% of CH_4. In the context of hydrogen admixture, the differences between various natural gases are in many ways negligible compared to the impact of hydrogen, as will be shown in a later section.

For methane, the combustion process with air can be described in a simplified manner as

$$1CH_4 + 2O_2 + 2 \cdot \frac{0.79}{0.21} N_2 \rightarrow 1CO_2 + 2H_2O + 2\frac{0.79}{0.21} N_2 + heat \qquad 1$$

For comparison, the reaction equation of a hydrogen-air combustion is as follows

$$1H_2 + \frac{1}{2} O_2 + \frac{1}{2} \cdot \frac{0.79}{0.21} N_2 \rightarrow 1H_2O + \frac{1}{2} \cdot \frac{0.79}{0.21} N_2 + heat. \qquad 2$$

The reaction equations given here describe a so-called stoichiometric combustion. A stoichiometric combustion process is a process in which the fuel is supplied with the minimum amount of oxidizer that is required to achieve complete fuel conversion.

While this is a very simplified way to describe the actual chemical processes taking place during combustion, it can already serve to determine a number of important fuel properties of methane and hydrogen. Properties such as minimum oxygen and air requirements, net and gross calorific values (NCV and GCV), Wobbe Indices and adiabatic combustion temperatures can easily be calculated using these reaction equations and their corresponding energy balances.

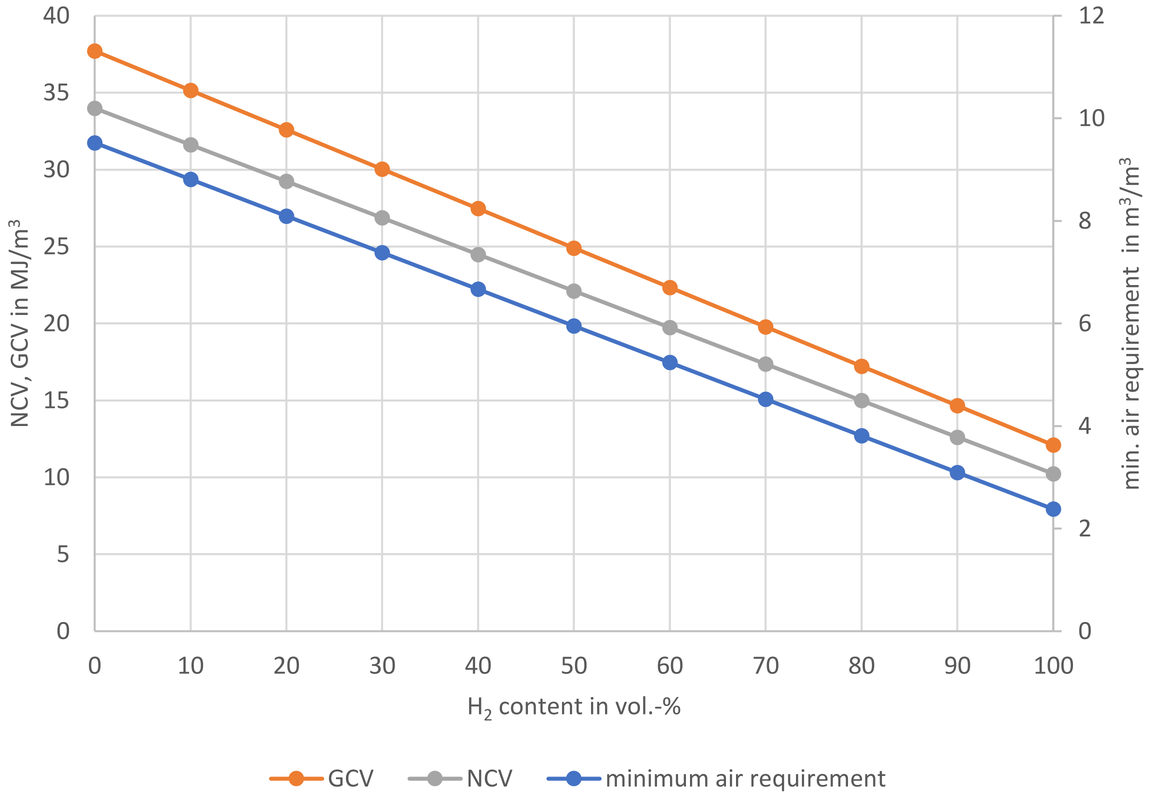

Figure 1: Impact of hydrogen content on minimum air requirement, net and gross calorific values. All values given in 15 °C/ 15 °C.

The first major difference between methane and hydrogen in terms of combustion is that hydrogen requires less oxygen to achieve complete fuel conversion than methane. The minimum oxygen requirement of H_2 is 0.5 mol O_2 / mol H_2, while methane requires 2 moles of O_2 per mole of CH_4. Correspondingly, the minimum air requirements are 2.381 mol air / mol H_2 and 9.524 mol air / mol CH_4, respectively.

This difference in oxygen requirements is also reflected in the volumetric net and gross calorific values of CH_4 / H_2 blends, as can be seen in **Figure 1** which shows the development of these properties with increasing hydrogen contents. With increasing hydrogen concentrations in the fuel gas blend, both air requirements and volumetric calorific values decrease in a linear manner. This also means that with increasing hydrogen concentrations in the CH_4/H_2 blend, more fuel gas is needed to provide the demanded heat input in an appliance.

It is worth pointing out that in mass-based terms, the calorific values actually increase with higher hydrogen contents, as the density of hydrogen is extremely low. This is shown in **Figure 2**.

Unless explicitly stated otherwise, calorific values will refer to volumetric properties in the following.

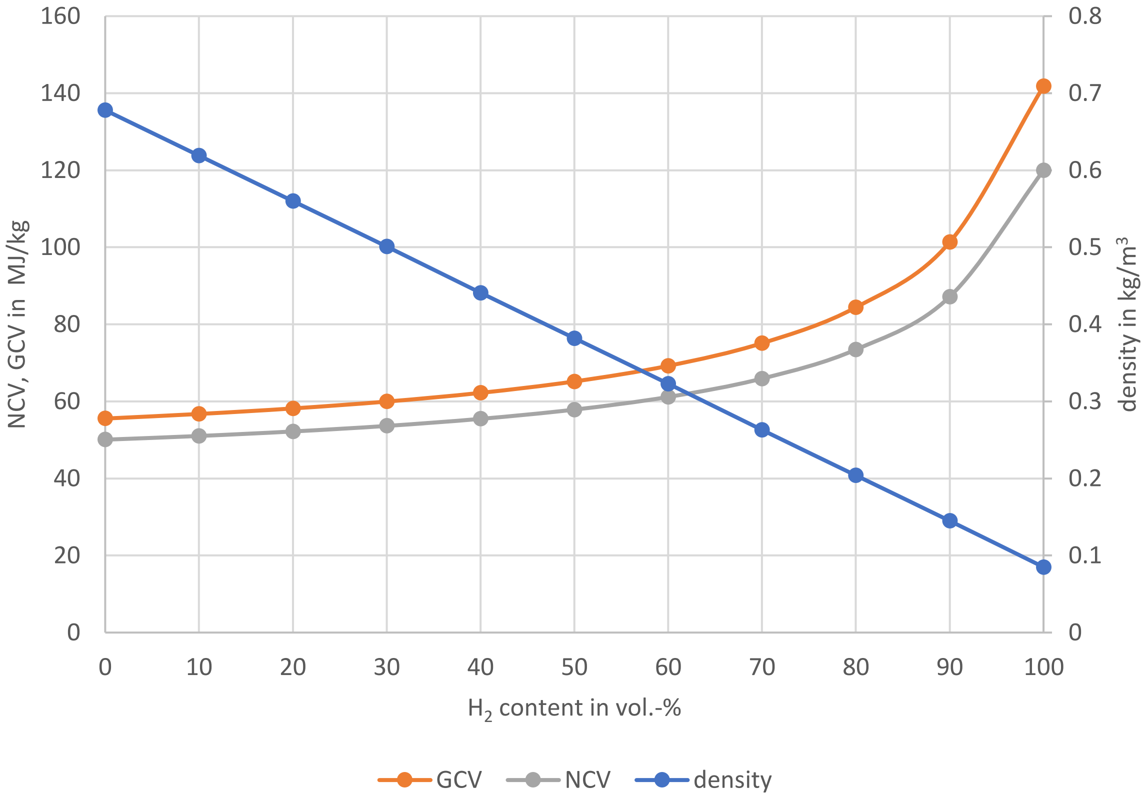

Figure 2: Mass-based calorific values and density of various CH_4/H_2 blends. All values given in 15 °C / 15 °C.

The impact of hydrogen admixture on the minimum air requirement is particularly relevant, as this property directly affects one of the most important process parameters of any combustion process, the air excess ratio λ.

While it is, in theory, possible to provide just the minimum air required to a combustion process, this is not advisable in practice, as this would require a perfect mixing of fuel and oxidizer within the premix or combustion chamber in order to achieve complete fuel consumption. Therefore, almost all technologically relevant combustion processes are supplied with more air than theoretically necessary, in order to ensure that all fuel molecules find their respective reaction partners.

Another reason is that in a real-life application the composition of the supplied natural gas may change over time, as might the composition of the combustion air (e. g. due to humidity changes). A sufficient amount of excess air ensures that even with fluctuating fuel and oxidizer compositions, complete combustion can be achieved.

The ratio of the actually supplied amount of air to the theoretically required minimum amount of air is expressed as λ, the air excess ratio:

$$\lambda = \frac{\dot{V}_{air,actual}}{\dot{V}_{air,min}} = \frac{\dot{V}_{air,actual}}{Air_{min} \cdot \dot{V}_{fuel}} \tag{3}$$

In some contexts, especially in research, it is more common to express the stoichiometry of a combustion process by means of the equivalence ratio Φ, defined as

$$\Phi = \frac{\dot{V}_{air,,min}}{\dot{V}_{air,actual}} = \frac{1}{\lambda}. \tag{4}$$

If there is a change in the composition of the fuel, the impact of this change on the stoichiometry of the combustion system may, however, not only be dependent on the fuel characteristics, but also on the way how fuel and oxidizer are conveyed into the combustion chamber. This will be discussed in more detail in **Section 3**, since the admixture of hydrogen can have a profound impact on the stoichiometry in residential appliances.

As stated previously, from an end-use perspective, the impact of blending of natural gas with hydrogen on an appliance (or indeed any kind of combustion equipment) is basically a gas quality problem. The composition and properties of the fuel gas change, compared to the gas that the appliance was originally designed and adjusted for, and the consequences of this change on the performance of the combustion appliance, e. g. in terms of emissions or efficiency, have to be assessed.

Gas quality issues are often discussed using a property called the Wobbe Index (W_S). The (superior or gross) Wobbe Index is defined as

$$W_s = \frac{H_S}{\sqrt{\rho_{n,fuel}/\rho_{n,air}}} = \frac{H_S}{\sqrt{d}}, \tag{5}$$

with H_S as the volumetric gross calorific value and d as the relative density of the fuel. ρ_N stands for the standard densities of fuel and air respectively.

In theory, two fuels with the same Wobbe Index will release the same amount of heat per time in a combustion system, provided the nozzle geometry and pressure do not change. These assumptions are generally valid for residential appliances, although combustion systems for other applications, particularly thermal processing industries, can require a different approach [2], [3].

Due to the importance of the Wobbe Index, particularly for appliances in the residential sector, national and international gas quality regulations usually rely on this property as the main gas quality criterion to assess the interchangeability of fuel gases.

Figure 3 shows the impact of hydrogen admixture to methane on some combustion-related gas quality criteria: relative density, net and gross calorific values and the Wobbe Index. It can be seen that the volumetric energy content of the fuel gas (i. e. the calorific value) decreases linearly with increasing hydrogen concentrations. Compared to methane, pure hydrogen's NCV and GCV decrease to 30 % and 32 % respectively.

The impact of hydrogen admixture on the relative density is even more pronounced: the relative density of H_2 is about eight times lower than that of CH_4.

As a result, the change in the Wobbe Index from pure methane to pure hydrogen is far less severe, as the change in the calorific value is largely counteracted by the change in densities. Thus, the difference in Wobbe Indices between CH_4 and H_2 is only about 9.5 %, while the change in the gross calorific value amounts to about 68 %. Also, the Wobbe Index – in contrast to the calorific values or density - does not decrease monotonously over the entire range of the mixture space. Instead, it actually increases with increasing hydrogen concentrations for fuel blends with very high hydrogen contents (cf. **Figure 3**).

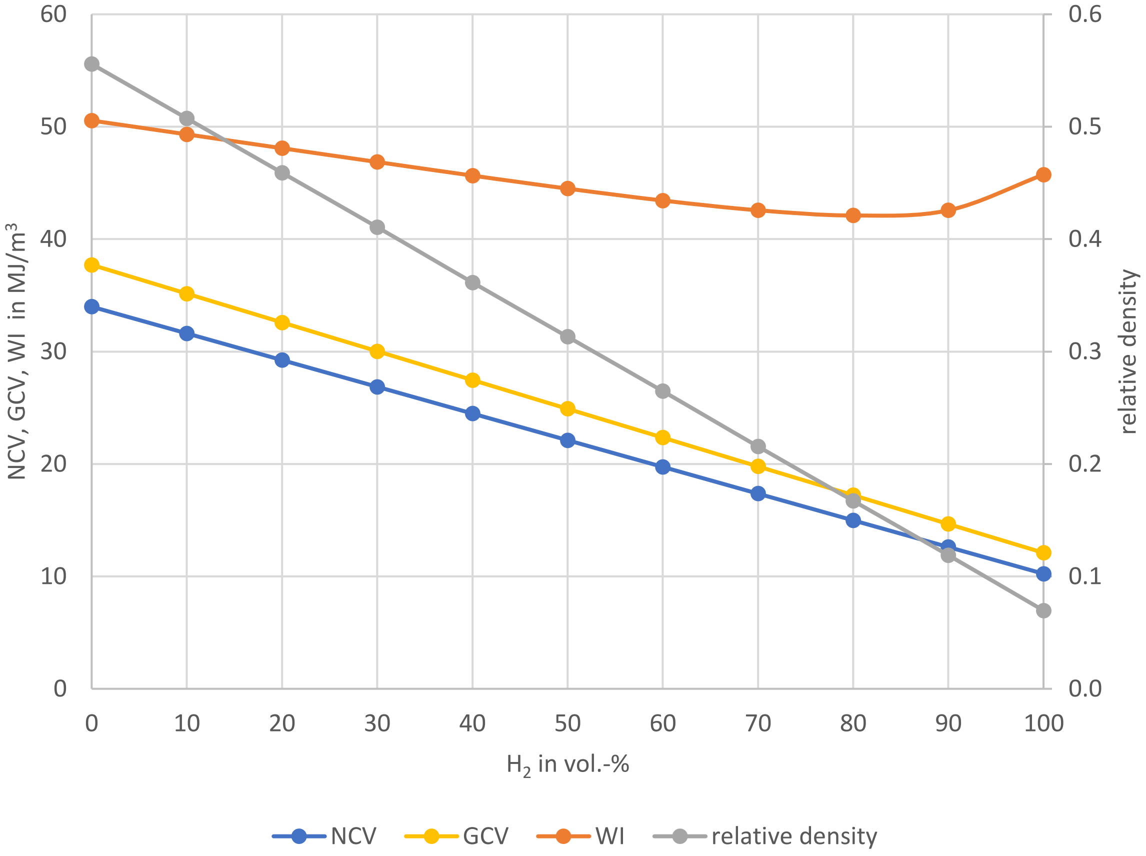

Figure 3: Change of main gas quality criteria as a function of hydrogen admixture to methane. All values given in 15 °C / 15 °C.

For any combustion process, the occurring temperatures are of paramount importance. Temperatures have a direct impact on heat transfer and process efficiency, but also on pollutant formation, e. g. of nitrogen oxides (NO_X).

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. The adiabatic combustion temperature is the theoretical maximum temperature that can be achieved in a combustion process, absent any losses. This means that the entire heat that is released by the chemical reaction is converted to thermal energy in the flue gas. The adiabatic combustion temperature is a function of the compositions and temperatures of fuel and oxidizer, pressure and the air excess ratio of the process. As such, it is an obvious first means to look at how hydrogen admixture will affect the temperatures in an appliance, even though the actual temperatures in the appliance will likely be significantly lower than the adiabatic temperatures.

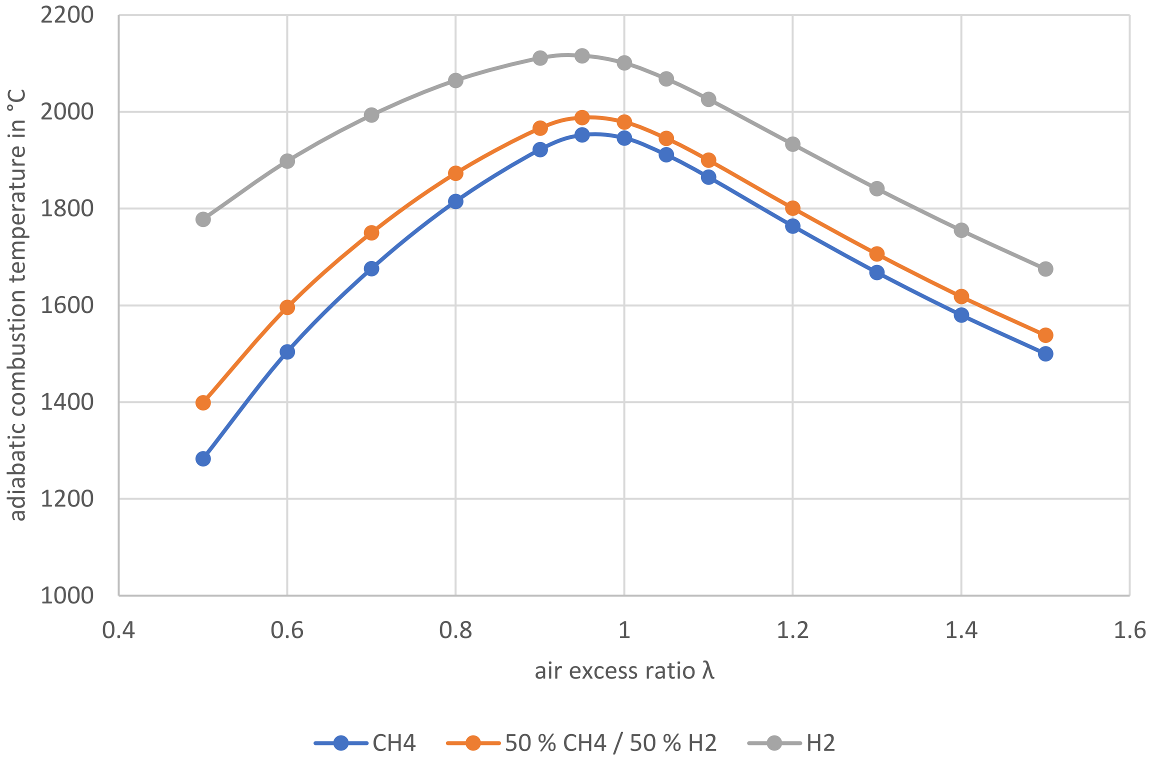

Figure 4: Adiabatic combustion temperatures of CH_4, 50 % CH_4 / 50 % H_2 and H_2 as functions of the air excess ratio, with air as oxidizer. T_{fuel} = 15 °C, T_{air} = 15 °C, p = 1 atm.

Figure 4 compares the adiabatic combustion temperatures of methane, a blend of 50 vol. % CH_4 and 50 % H_2 and pure H_2 over a wide range of air excess ratios. Hydrogen and methane / hydrogen blends 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 $\lambda = 1$). The difference between adiabatic combustion temperatures of methane and pure hydrogen, however, is significantly higher (more than 150 °C). This non-linear increase of the adiabatic combustion temperature with rising hydrogen concentrations can also be seen in **Figure 5**.

Another important gas quality criterion is the Methane Number (MN), primarily in the context of reciprocating engines. This is relevant for the residential end-use sector, as there are a number of micro-CHP appliances available on the EU market which use gas engines as prime movers to supply both heat and power in a decentralized manner. The Methane Number quantifies the propensity of a fuel gas to prematurely self-ignite in a spark-ignited reciprocating engine, which can lead to a situation called “knocking”. Knocking causes an uneven operation of the engine, reducing its efficiency, increasing pollutant emissions, and can even cause damage to the engine.
The Methane Number is determined by comparing the self-ignition propensity of a given fuel gas in a well-defined engine [4] to two extremes, pure methane and pure hydrogen. By definition, the very self-ignition-resistant CH_4 has a MN of 100, while the extremely reactive H_2 has a MN of 0. A fuel gas with a MN of 80 thus has the same knocking characteristic as a blend of 80 vol.-% CH_4 and 20 vol.-% H_2. This empirical approach is similar to the definition of Octane Numbers and Cetane Numbers for gasoline and diesel engines, respectively. There are algorithms available to calculate the Methane Number for a given fuel gas composition, e. g. the AVL-method [4] and MWM methods [5]. Both are based on empirical data and produce very similar Methane Numbers for a given fuel.

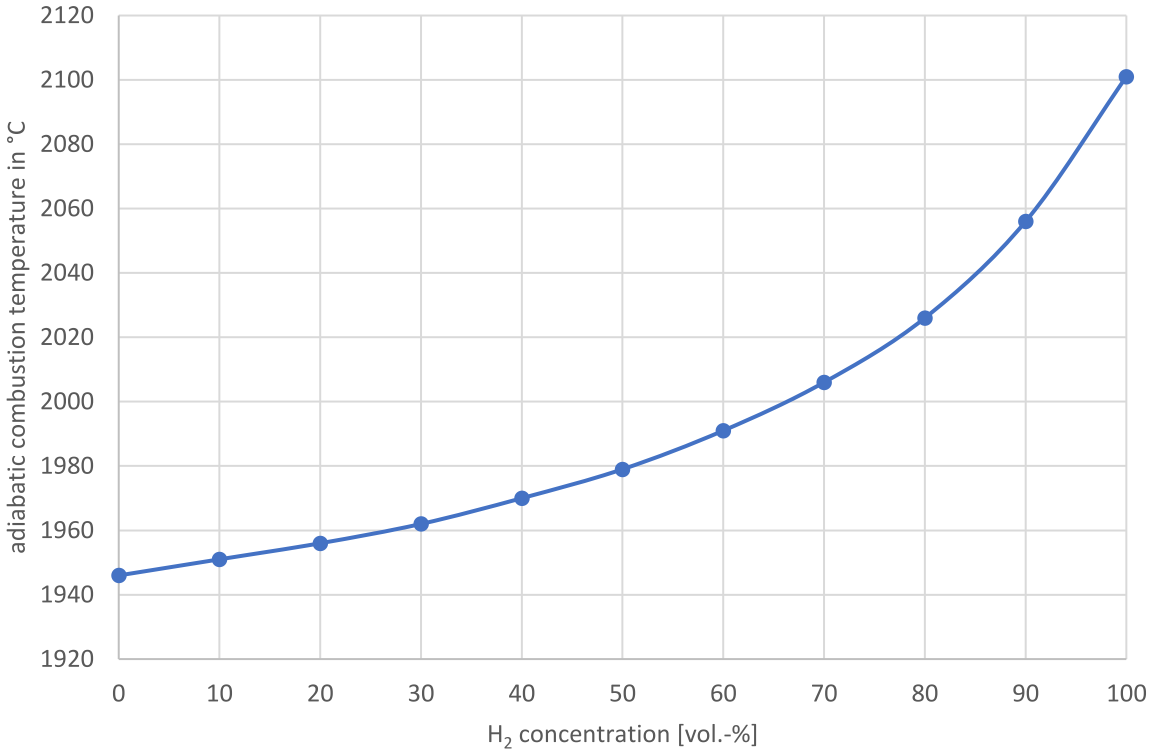

Figure 5: Effect of hydrogen admixture on the adiabatic combustion temperature at λ *=1. Oxidizer: air,* T_{gas} *= 15 °C,* T_{air} *= 15 °C, p = 1 atm.*

Higher hydrocarbons in the fuel gas reduce its Methane Number, i. e. the likelihood of knocking is increased, while carbon dioxide increases the MN and hence inhibits knocking. Other inert components such as nitrogen only have a very small impact on MN. For example, a mixture of 90 vol. % CH_4 and 10 vol.-% N_2 will result in a MN of 99.

Considering the very definition of the Methane Number, it is obvious that hydrogen admixture will significantly reduce the Methane Number and hence increase the knocking propensity of a fuel gas. MN is a property dominated by chemical kinetics, and as such, not closely correlated to calorific values or Wobbe Indices. For example, two commonly distributed H-gases within the EU, Russian H-Gas and North Sea H-Gas have almost identical Wobbe Indices and vary in GCV by about 4 %, but differ significantly in Methane Numbers (Russian H: 90, North Sea H: 77).

Higher hydrocarbons will increase the Wobbe Index and calorific value of a fuel, but decrease the Methane Number. Hydrogen, on the other hand, will reduce calorific values, Wobbe Indices and Methane Numbers.

3 The impact of hydrogen admixture on stoichiometry

As was stated previously, the air excess ratio λ is one of the most important process parameters for any kind of combustion process, and closely related to the fuel gas composition. It will influence not only the temperatures occurring in the combustion system, but also energy efficiency and pollutant emissions. However, how exactly the stoichiometry is affected in a combustion process is not only dependent on the fuel composition, but on other factors as well, e. g. the way the fuel and oxidizer volume flows are controlled and whether there is some kind of active combustion control.

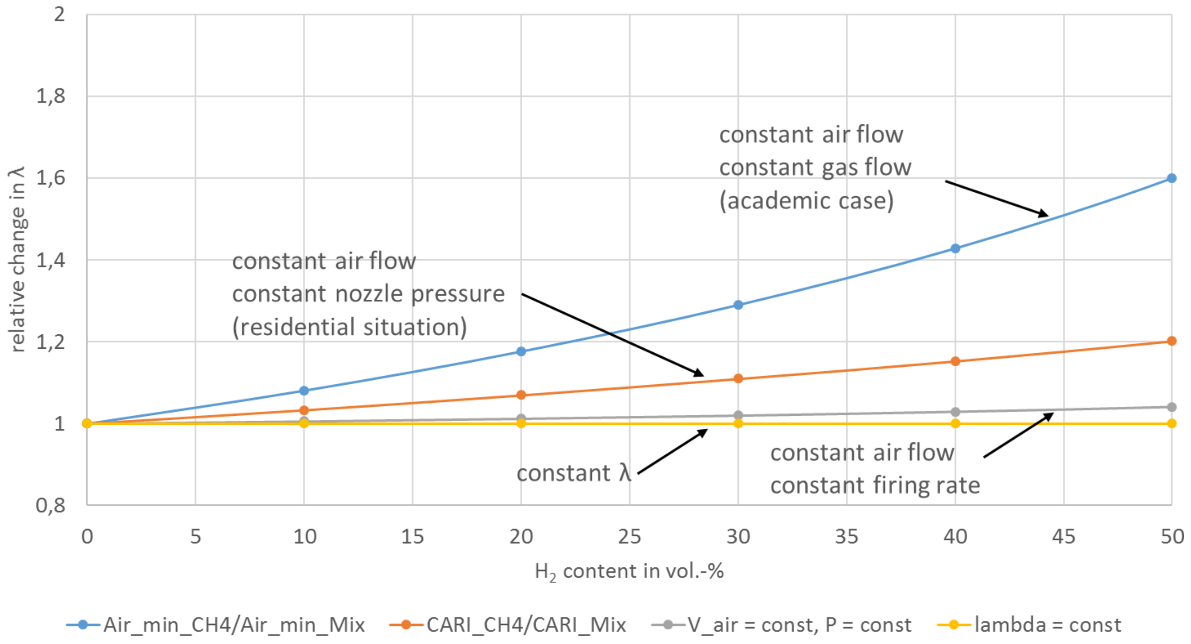

Figure 6: Impact of hydrogen admixture on the relative stoichiometry of a combustion system

As hydrogen has a lower minimum air requirement compared to methane, any blend of CH_4 and H_2 will also require less air per molar unit of fuel than pure methane for complete combustion. Thus, if only the chemistry of combustion is considered, the admixture of hydrogen to methane will result in a shift of the air excess ratio following the equation

$$\lambda_{mix} = \frac{Air_{min,CH4}}{Air_{min,mix}} \lambda_{CH4} \, . \qquad 6$$

This would correspond to a situation where only the fuel composition changes, but the volume flows of both fuel and oxidizer remain constant. This is, for the most part, an academic case that would also imply that the heat input into the combustion system is reduced as well, due to the reduced calorific value of the methane/hydrogen blend.

Another rather academic case would be if both the volume flow of air and the heat input into the system, i. e. the firing rate, were kept constant. In this case, the shift of the stoichiometry could be expressed by the equation

$$\lambda_{mix} = \frac{Air_{min,\,CH_4}}{Air_{min,mix}} \cdot \frac{H_{i,mix}}{H_{i,CH_4}} \, . \qquad 7$$

In such a system, the shift of stoichiometry would be significantly reduced as the changes in the minimum air requirements are largely compensated by the changing calorific values.

More appropriate for the situation in residential and commercial sector appliances without any kind of combustion control is a case where both the air flow and the pressure at the nozzle remain constant. In such a system, the volume flow of the fuel gas is a function of the Wobbe Index (which is based on the assumption of constant nozzle pressure and diameter), and thus, the shift in stoichiometry is significantly reduced (cf. **Figure 6**) since the gas volume flow also changes. This means that the firing rate of the burner changes as well.

For such systems, calculating the required amount of air by using the purely chemical minimum air requirement Air_{min} would be inappropriate. Instead, a property called the Combustion Air Requirement Index (CARI) is used, which is defined in analogy to the Wobbe Index:

$$CARI = \frac{Air_{min}}{\sqrt{d}} = \frac{Air_{min}}{\sqrt{\rho_{n,fuel}/\rho_{n,air}}} . \qquad 8$$

For such a situation (which is the case for most residential gas combustion appliances), the shift in air excess ratios will theoretically follow the ratio of the CARI values for the different fuels:

$$\lambda_{mix} = \frac{CARI_{CH4}}{CARI_{mix}} \lambda_{CH4} . \qquad 9$$

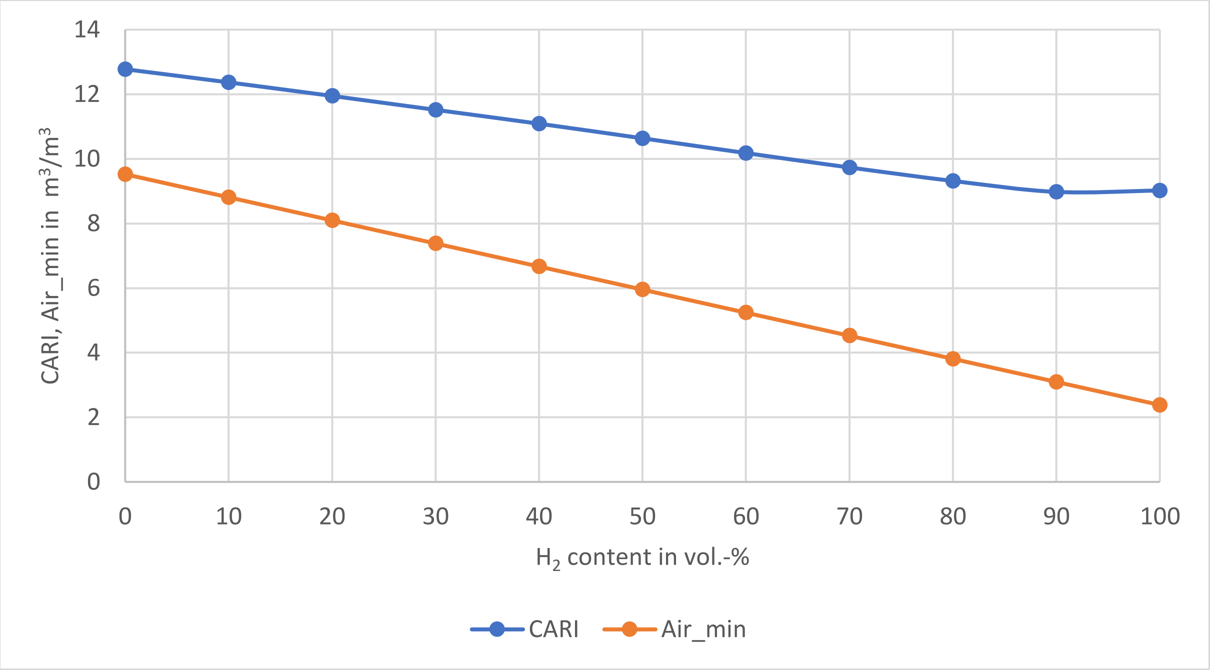

Figure 7: CARI and Air_{min} in CH_4/H_2 blends as functions of the H_2 concentration

Figure 7 compares the evolutions of both Air_{min} and CARI with increasing hydrogen concentrations. Similar to the Wobbe Index, the evolution of CARI is not linear, due to the effects of density. The final curve in **Figure 6** depicts a case were some of active combustion control maintains a constant air excess ratio, i. e.

$$\lambda_{mix} = \lambda_{CH_4} . \qquad 10$$

Such control systems are not in widespread use in residential and small-scale commercial appliances, but more common in larger combustion systems.

4 Laminar combustion velocities

Most residential heating appliances use premixed burners to generate heat, i.e. fuel and air are thoroughly mixed prior to being injected into the combustion space. In these burners, the laminar combustion velocity s_L plays an important role for flame stability and shape. It is defined as the velocity with which the flame front propagates into the unburned fuel/oxidizer mixture and is dependent on the composition of both fuel and oxidizer, temperature and pressure and the air excess ratio.

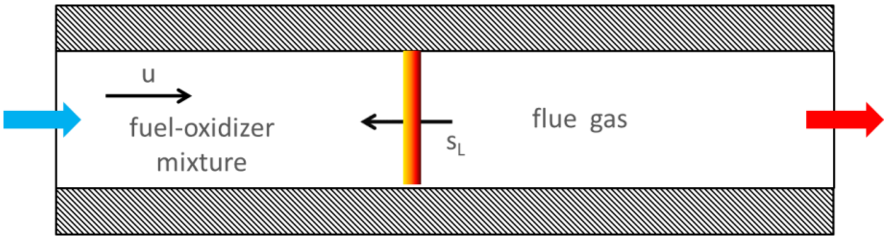

Figure 8: Flow velocity of the fuel-oxidizer mixture u and laminar combustion velocity s_L

Since flows in residential appliances are laminar, only the laminar combustion velocity will be discussed here.

The balance between the flow velocity of the fuel/oxidizer mixture u and the laminar combustion velocity s_L (see Figure 8) determines where the flame establishes itself in a stable manner:

1. If s_L is too small compared to u ($s_L << u$), the flame will be carried out of the combustion chamber towards to the flue gas duct, a so-called blow out.
2. When the flow velocity u and the laminar combustion velocity are of equal magnitude ($u =- s_L$), but opposite orientation, the flame is stable.
3. From the "stable flame" configuration, if s_L increases and becomes too close to an also changed u, the flame stabilizes closer to the burner, causing potential overheating.
4. If the combustion velocity becomes higher than the flow velocity ($s_L > u$), the flame will move upstream, find a stabilization point close to the injector and potentially damage the burner itself (flashback)

Although laminar flame speed is the predominant factor influencing flashback, thermal balance between the flame and the burner port also play a critical role. As there is a balance between flame and unburned gas velocity, there is also a thermal balance between the heat release and its dissipation upstream of the flame front. When a flame front is located near a surface, heat production in the front flame can be counterbalanced by heat dissipation to the surface and lead to an extinction of the flame. This phenomenon is known as flame quenching.

In the case of a premixed gas burner, when the laminar combustion velocity is gradually increased (by hydrogen addition, for example), the reaction zone stabilizes progressively closer to the burner and heat loss to the burner will begin to increase. If the port size of the burner is large, flash back occurs. But if the burner port is small enough, the flame is quenched due to heat loss, thus preventing flash back. This aspect is relevant in the context of hydrogen admixture, as the minimum quenching diameter for hydrogen combustion is significantly smaller as for methane combustion.

Minimum quenching diameters have been measured for several gases and for circular ports as reported in [6].

Table 4-1 Minimum quenching diameter for different fuel gases [6].

Gas	H_2	CH_4	C_2H_6	C_3H_8
Minimum quenching diameter (mm)	0.8	3.5	2.5	2.9

These minimum quenching diameters should be considered as theoretical because they are only valid for fully developed (parabolic) laminar flows. Moreover, reducing the flame port diameter to prevent flashback can have some side effects such as reducing the power range of the burner or causing blow off effect for some gases. However, it can be concluded that the smaller the flame ports, the more resistant to flash back the burner should be. On the other hand, burner nozzles that are sufficiently small to prevent flash backs in premixed methane-air combustion may be insufficient for hydrogen or methane-hydrogen blends.

For a particular air/gas mixture, there is a critical port diameter known as the quenching diameter, below which the flame will not light back. Several authors have produced theories on the calculation of the quenching diameter. Jones [6] cited the following formula:

$$\frac{s_L d_q}{\alpha} = constant \qquad 11$$

where s_L is the laminar velocity of the unburnt gas/air mixture, d_q is the quenching diameter and α is the thermal diffusivity of the mixture.

With this formula, it is not possible to calculate the quenching diameter for a new burner, but it allows to estimate its decrease when hydrogen is added to natural gas. In theory, according to this formula applied in **Table 2** for CH_4/H_2 mixtures, a stoichiometric burner having a quenching diameter of 1 mm for pure methane, should not have flash back with 40 % CH_4/60 % H_2 mixture if its port diameter is reduced to 0.68 mm. On top of having to increase the number of ports to the burner to compensate for their diameter decrease, there will be some other side effects like a change in the blow off limit and the heating power range.

Table 2 : Evolution of the quenching diameter for circular burner ports, starting from an arbitrary diameter of 1 mm, for CH_4-H_2 mixtures.

Quenching diameter evolution compared to an arbitrary reference of 1mm for CH4											
Primary air excess/%H_2	CH_4 (0% H_2)	10% H_2	20% H_2	30% H_2	40% H_2	50% H_2	60% H_2	70% H_2	80% H_2	90% H_2	**100% H_2**
0.6	1.00	0.95	0.88	0.81	0.74	0.65	0.55	0.43	0.27	0.10	**0.05**
1	1.00	0.96	0.92	0.87	0.82	0.76	0.68	0.60	0.51	0.41	**0.32**
1.3	1.00	1.46	0.91	0.86	0.81	0.74	0.67	0.59	0.50	0.40	**0.29**

As it can be seen in **Table 2**:

- The quenching diameter is still about 90 % of the initial diameter when 20 % of H_2 is added to natural gas.
- The quenching diameter decreases to about 60 % of the initial diameter when 60 % of H_2 added to natural gas. The quenching diameter decrease depends on the primary air excess of the burner. For partially premixed flames, the decrease is quicker than for fully premixed flames.

This theory has been extended to non-circular burners (cf. [6]).

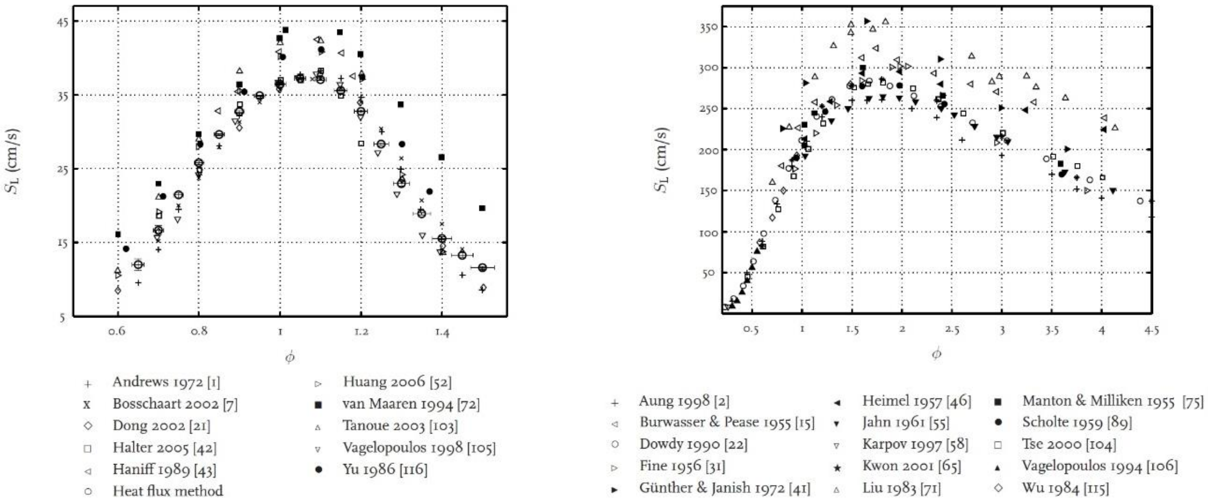

Figure 9: Laminar combustion velocities of CH_4 (left hand side) and H_2 (right hand side) [7]

Figure 9 shows a comparison of the laminar combustion velocities of methane (left hand side) and hydrogen (right hand side) over the equivalence ratio (i.e. 1/λ), based on a compilation of a large number of experiments and numerical simulations [7]. While there is a lot of scatter in these plots due to different measurement and simulation approaches, it is obvious that hydrogen has a significantly higher laminar combustion velocity over the entire relevant range of equivalence ratios. Also, while the maximum s_L is achieved at slightly sub-stoichiometric conditions for methane, this maximum is shifted significantly towards sub-stoichiometric regimes for pure hydrogen.

At the same time, the differences in the x-axes show that hydrogen is reactive over a far wider range of equivalence ratios.

These effects are highlighted in **Figure 10** which visualizes the laminar combustion velocities of methane, hydrogen and two CH_4/H_2 blends. The diagram is based on a series of one-dimensional simulations of freely propagating premixed flames, using the medium-sized reaction mechanism GRI 3.0 [8]. This mechanism contains 53 species and 325 reaction equations.

Figure 11 shows the same dataset, but focusing only on methane and the CH_4/H_2 blends. Both diagrams show the same trend: higher levels of hydrogen in the fuel increase the laminar combustion velocity, while the maximum is shifted towards sub-stoichiometry.

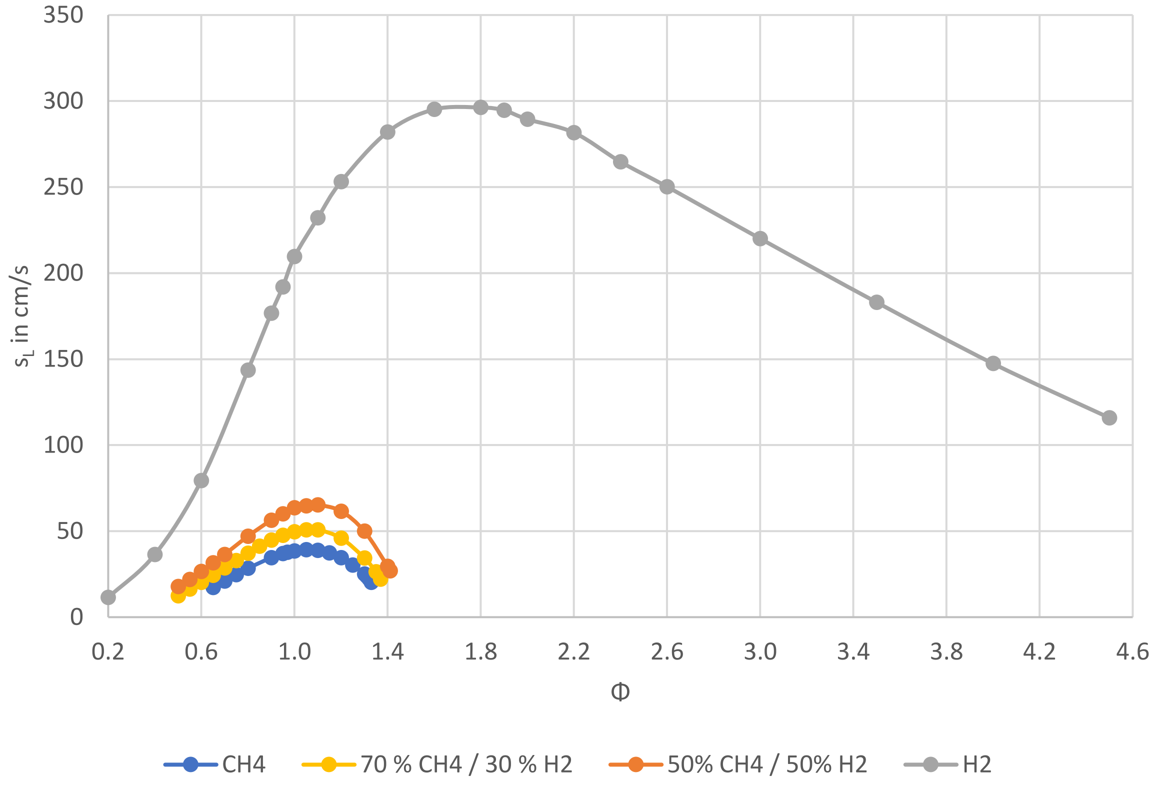

Figure 10: Comparison of laminar combustion velocities of methane, hydrogen and different CH_4/H_2 blends.

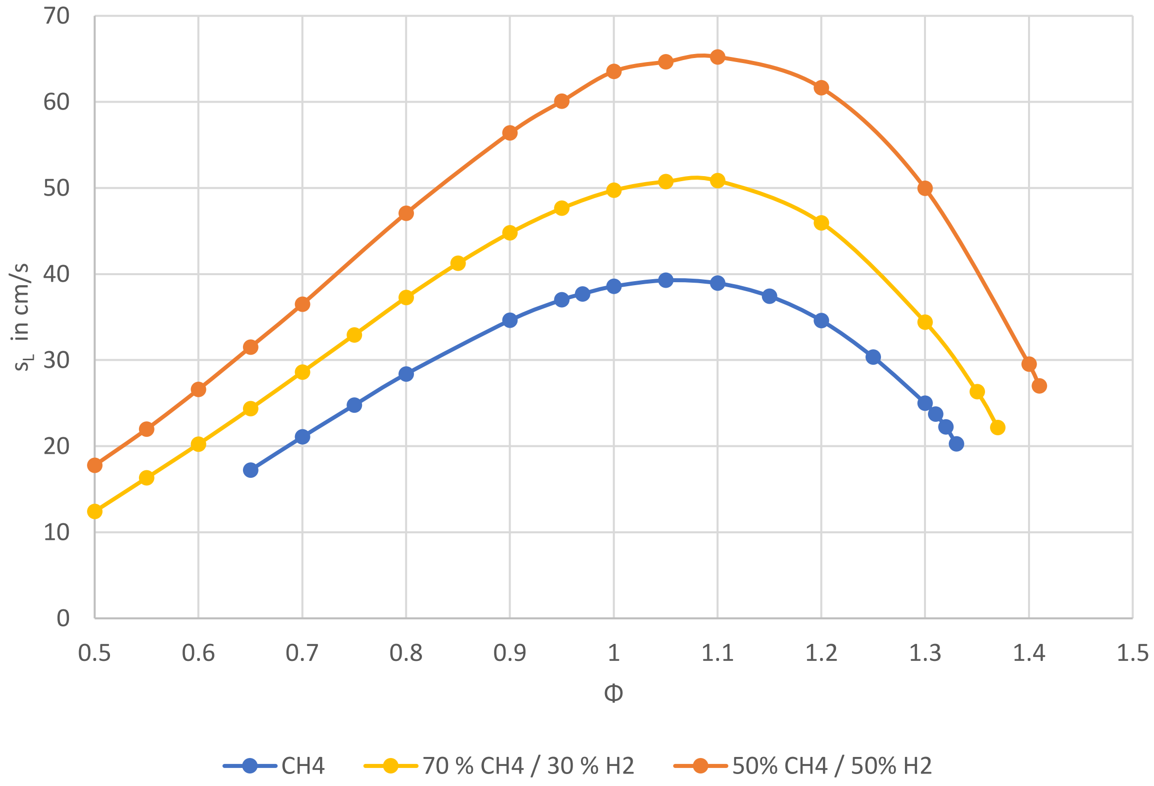

Figure 11: Comparison of laminar combustion velocities of methane, 70% CH_4 / 30 % H_2 and 50 % CH_4 / 50 % H_2

This is of particular concern for premixed or partially premixed burners since higher laminar combustion velocities increase the risk of overheating or even a flash back, as the balance between flow velocity and combustion velocity is changed.

In contrast to properties like densities, calorific values and Wobbe Indices, which can easily be calculated based on the fuel composition, the determination of the laminar combustion velocity of a fuel is more complex. It requires either a dedicated measurement device or the use of simulation methods.

Therefore, empirical approaches were developed to simplify these calculations (e. g. [9]) to avoid the need for a complex simulation. The approach described in [9] is based on a number of coefficients for key components of a fuel over a range of equivalence ratios, and calculates the ratio of s_L for a given natural gas composition to the laminar combustion velocity of pure methane. However, it was developed for the use with natural gases of various compositions. While a set of coefficients is included for hydrogen, it has to be checked whether these empirical approximations are reliable for natural gas / hydrogen blends, especially at higher levels of hydrogen.

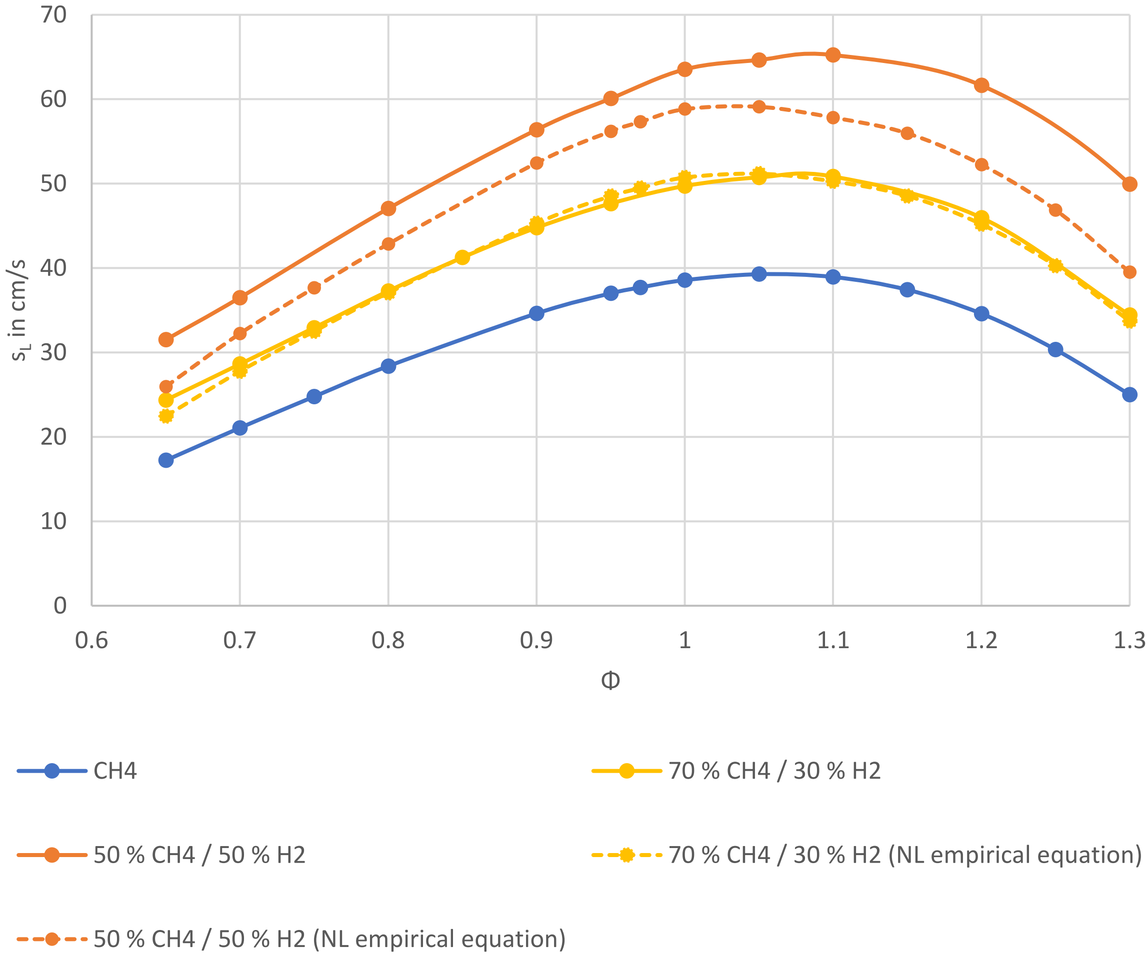

Figure 12: Comparison of laminar combustion velocities calculated by 1D simulation and the Dutch empirical approach [9]]

Figure 12 shows a comparison between the results of a 1D simulation based on a complex mechanism (GRI 3.0 in this case) and the approximation proposed in [9] for two CH_4/H_2 blends, with the dotted lines representing the empirical solution. While the approximation achieves a good agreement with the simulation data for the 70 % CH_4 / 30 % H_2 blend, (except in regions of high air excess), it severely underpredicts the laminar combustion velocity of the 50 % CH_4 / 50 % H_2 blend over the entire range, with deviations of up to -20 % at the extremes. Even at stoichiometry, the difference between simulation and empirical approximation amounts to about - 7 %.

When a premixed burner that was originally adjusted for methane is supplied with a methane/hydrogen mixture, the response of the combustion system in terms of the actual laminar combustion velocity is determined by a number of factors. The higher hydrogen content causes an increase in s_L, but at the same time may change the stoichiometry of the system, if there is no active control system to maintain a constant air excess ratio. The combustion temperatures change as well. In the case of super-stoichiometric combustion ($\lambda > 1$, common for heating systems), these effects will to a certain degree cancel out each other. The increase in s_L is largely compensated by the increasing air excess ratio, which tends to reduce the laminar combustion velocity (and temperature).
In the case of a sub-stoichiometric system ($\lambda < 1$), however, the effects of the shift in stoichiometry and higher hydrogen content will actually stack, leading to a significant increase of the laminar combustion velocity. This is a situation that could occur in a partially premixed cooking device, for example.

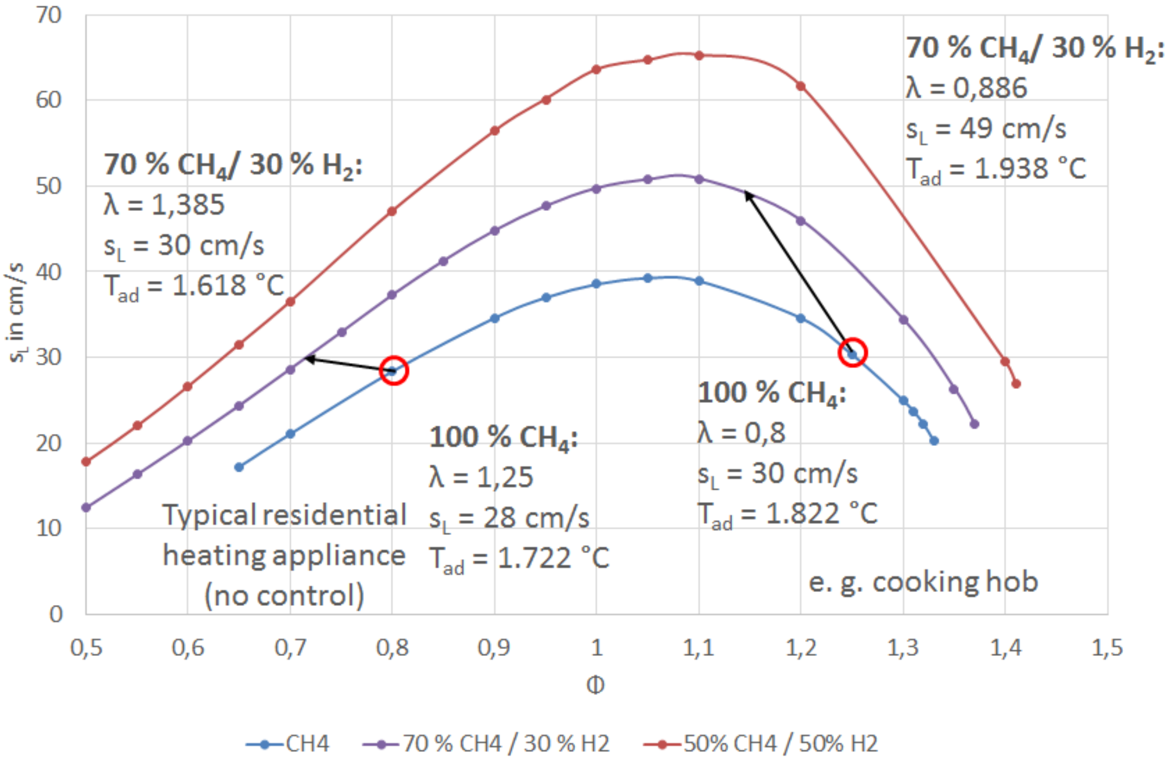

Figure 13: Impact of hydrogen admixture on the laminar combustion velocity for a system with constant air flow and nozzle pressure

Figure 13 visualizes this interaction between different phenomena for two examples. Both examples assume a constant nozzle pressure and air flow, which is a reasonable assumption for residential appliances. Also, no combustion control system is used in these two examples.
The first example represents a conventional residential heating system that was adjusted for pure methane to operate at an air excess ratio of 1.25 (i. e. an equivalence ratio Φ = 0.8). If this appliance is supplied with a fuel of 70 vol.-% CH_4 and 30 vol.-% H_2, the air excess ratio shifts towards a higher air

excess, in this case of $\lambda = 1.385$ ($\Phi = 0.722$). This shift is controlled by the ratio of the respective CARI values for both fuel compositions, as was discussed previously. Thus, the increase due to the higher hydrogen concentration of the fuel is largely compensated by the shift towards higher stoichiometry, resulting in an overall increase of s_L by about 7 %. At the same time, the adiabatic combustion temperature decreases as well, caused by the higher air excess ratio.

The second example in **Figure 13** assumes a sub-stoichiometric combustion process ($\lambda = 0.8$, i. e. $\Phi = 1.25$), for example as part of a partially premixed burner system in a cooker. Again, the fuel is switched from methane to a blend of 70 vol.-% CH_4/ 30 vol.-% H_2, and this time, both the change of fuel composition and the shift closer to stoichiometry will add up to a significant increase of the laminar combustion velocity by more than 31 %. The adiabatic combustion temperatures will also increase by more than 100 °C.

Figure 13 may also serve to show the impact of combustion control systems on the laminar combustion velocities. These systems generally use either flame ionization data or flue gas measurements in combination with a variable air flow to maintain a constant air excess ratio [10], [11]. This could mean for a heating system with combustion control that in the case of hydrogen admixture, laminar combustion velocities increase much stronger than in an uncontrolled system. For the first example, the switch from pure methane (at an air excess ratio of 1.25) to a blend of 70 vol.-% CH_4 / 30 vol.-% H_2 would result in an increase of the laminar combustion velocity of more than 30 %.

5 Pollutant and CO_2 emissions

In terms of combustion-related pollutant emissions, only carbon monoxide (CO) and nitrogen oxides (NO_X) emissions are relevant, as natural gas is basically sulphur-free.

Carbon monoxide is the result of the incomplete combustion of hydrocarbon fuels and as such, closely related to the air excess ratio λ. As was shown previously, the addition of hydrogen to methane (representing natural gas in this context) will shift the stoichiometry of a combustion process towards higher λ values, unless a constant air excess ratio is maintained by some kind of control system. This means that carbon monoxide emissions are less likely to occur. **Figure 14** shows the CO and NO emissions as functions of the air excess ratio. This diagram serves to highlight the principal interdependence between CO and NO emissions and the stoichiometry of the system. It can be seen that at air excess ratios greater than unity, the CO emission is practically unaffected by the level of hydrogen in the fuel.

It should be pointed out that **Figure 14** is based on a series of adiabatic chemical equilibrium calculations. In a real combustion system, there will also be an increase of CO emissions at high air excess ratios as the flame becomes increasingly unstable, leading to locally incomplete combustion and hence CO formation. This upper stability limit is, however, very much dependent on the actual burner design and cannot be easily calculated which is why it is neglected here. Measurements will be required to determine whether hydrogen admixture will affect these higher stability limits of burner systems.

Nitrogen oxides are a different matter, as the thermal NO_X formation process, the dominant formation pathway for gas combustion, is closely linked to temperatures and the availability of oxygen in the hot reaction zone. The admixture of hydrogen leads to higher combustion temperatures and higher air excess, hence more oxygen is available in the reaction zone. However, residential heating appliances are commonly operated at rather high air excess ratios. A further increase of λ due to the changing fuel composition will lead to a considerable reduction of local temperatures. The net effect is a reduction of NO_X emission when adding hydrogen, if the stoichiometry change is allowed to happen. In the case of a system operated at low air excess ($\lambda < 1.2$) or a system which uses some kind of combustion control to keep air excess ratios constant, higher NO_X emissions are to be expected, however, due to an increase in local temperatures. This would be similar to non-premixed burners, e. g. in industrial applications, where the main combustion processes always occurs at near-stoichiometric conditions in the flame front, independent of the global air excess ratio of the process [12], [13].

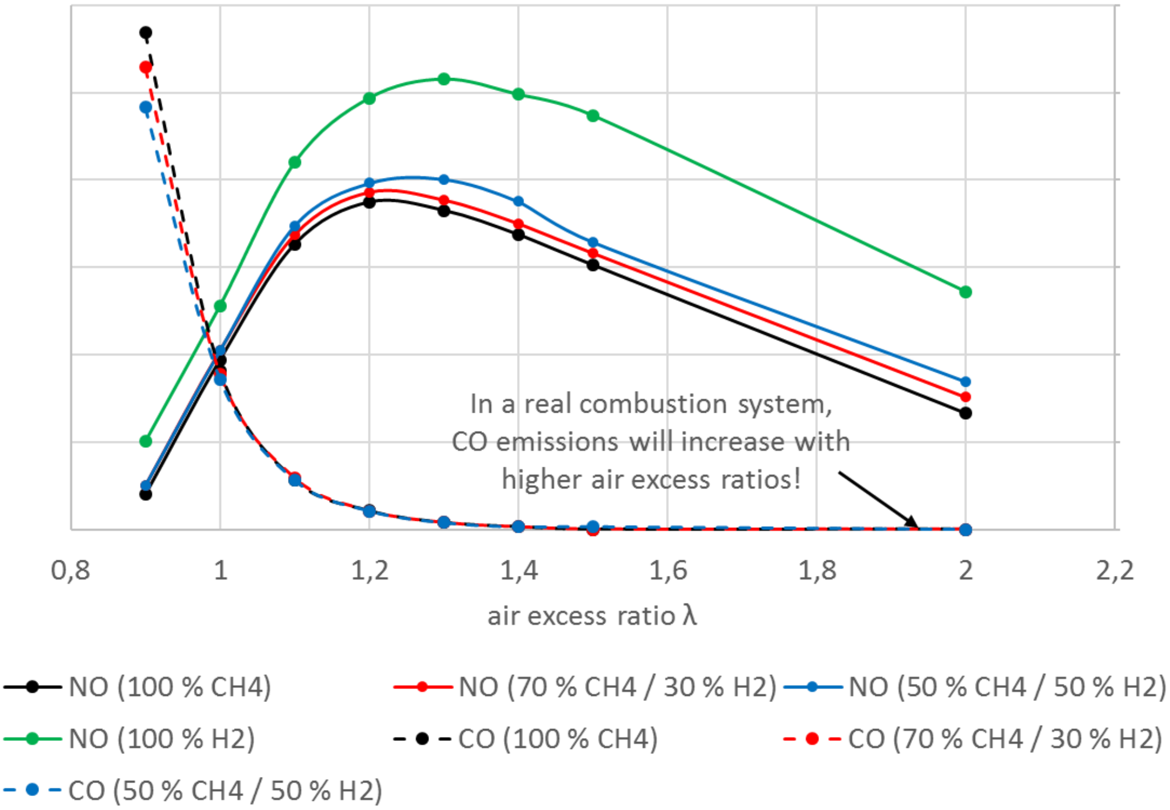

Figure 14: CO and NO emissions as a function of the air excess ratio for methane, hydrogen and CH_4/H_2 blends, corrected to 0 vol.-% O_2 in the flue gas.

One of the primary motivations to discuss hydrogen is the global need to reduce CO_2 emissions. **Figure 15** shows how the specific CO_2 emissions of methane / hydrogen blends evolve with increasing levels of hydrogen. The graph is based only on the combustion-related emissions, i. e. the additional CO_2 emissions due to production and transport of natural gas or hydrogen are not considered. It can be seen that from a combustion point-of-view, the potential to reduce CO_2 emissions by blending hydrogen with natural gas is rather limited. Thus, an admixture of 20 vol-% of H_2 into CH_4 will only reduce combustion-related CO_2 emissions by about 7 %, assuming the same heat output. With 40 vol. -% H_2 in the fuel gas, a CO_2 reduction of about 17 % can be achieved, while a hydrogen content of 60 vol.-% will result in a reduction of about 31 %.

This shows that hydrogen admixture alone will be unable to decarbonize gas utilization, but needs to be accompanied with a substitution of natural gas with either bio-methane or SNG.

Alternatively, switching to pure hydrogen would of course also be an option.

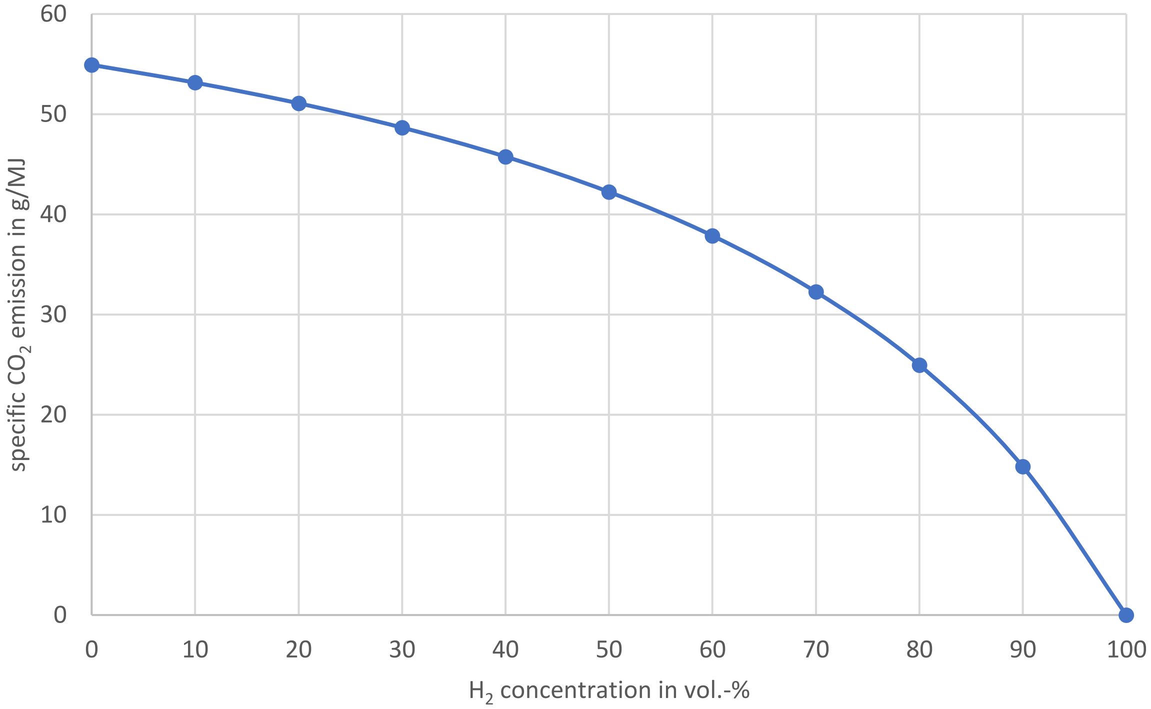

Figure 15: Evolution of the specific combustion-related CO_2 emissions of methane / hydrogen blends

6 Safety aspects

In addition to the impact of using natural gas/hydrogen blends on the combustion processes in residential or commercial appliances, additional aspects have to be considered as well. Safety obviously is of paramount importance, and some safety-related issues have already been addressed, e. g. CO emissions and flash back phenomena

Given the significantly different properties of hydrogen when compared to methane, other aspects have to be considered as well. Hydrogen has a minimum ignition energy that is about ten times lower than that of methane, while the self-ignition temperatures of both gases are rather similar (H_2: 560 °C, CH_4: 595 °C) [14]. As a consequence, CH_4/H_2 blends can be ignited much more easily than methane, with obvious safety-related consequences.

Also, increasing levels of hydrogen in methane lead to a wider range of flammability, as can be seen in **Figure 16**. While the lower explosion limit (LEL) is hardly affected by hydrogen admixture, the upper explosion limits (UEL) rise drastically with higher hydrogen concentrations [15]. At the same time, the limiting oxygen concentration (LOC), i. e. the minimum oxygen concentration below which combustion is not possible, decreases from 13.6 vol.-% O_2 for methane to 5.1 vol.-% O_2 for pure hydrogen.

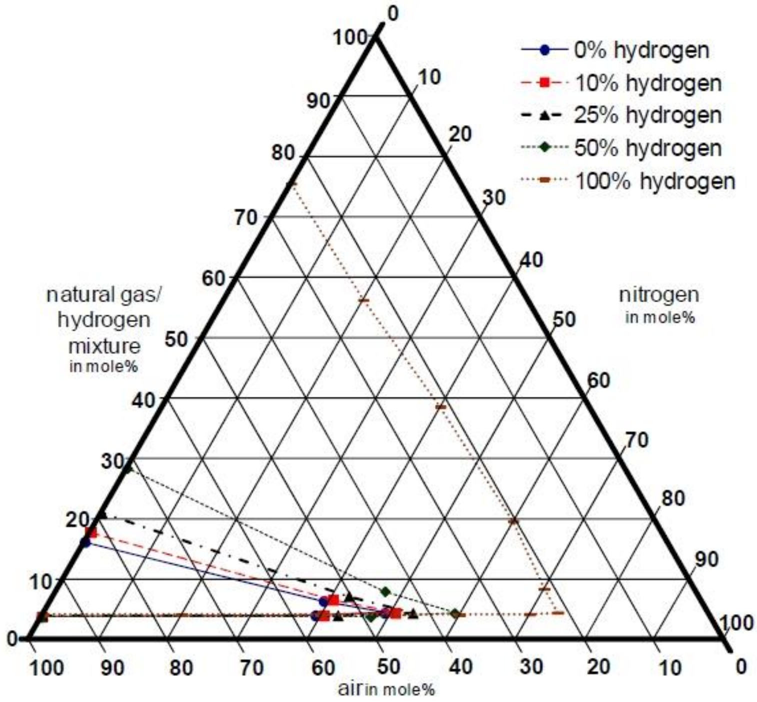

Figure 16: Ignition limits of methane, hydrogen and natural gas / hydrogen blends with air [15]

Explosions are always a concern when it comes to operate any kind of combustion equipment. Two important properties in this regard are the maximum explosion pressure and the deflagration index K_G. The maximum explosion pressure (also called deflagration pressure) is the maximum pressure that can occur due to heat release when a combustible mixture is ignited in a closed vessel. It is worth pointing out that this is only true for deflagrations, i. e. a sub-sonic propagation of a flame and a corresponding pressure increase due to heat release from combustion. This is physically very different from a detonation where the pressure wave moves at supersonic speeds and there is a strong interaction between the chemistry and pressure effects [16].

Figure 17 shows the impact of hydrogen admixture on the maximum explosion pressure as a function of the air excess ratio, calculated using an iso-volume adiabatic chemical equilibrium in combination with the GRI 3.0 thermo-chemical data base [8]. It can be seen that higher levels of hydrogen in the fuel blend reduce the maximum explosion pressure. The maximum pressure levels always occur at about $\lambda \approx 0.9$ and are independent of the fuel gas composition.

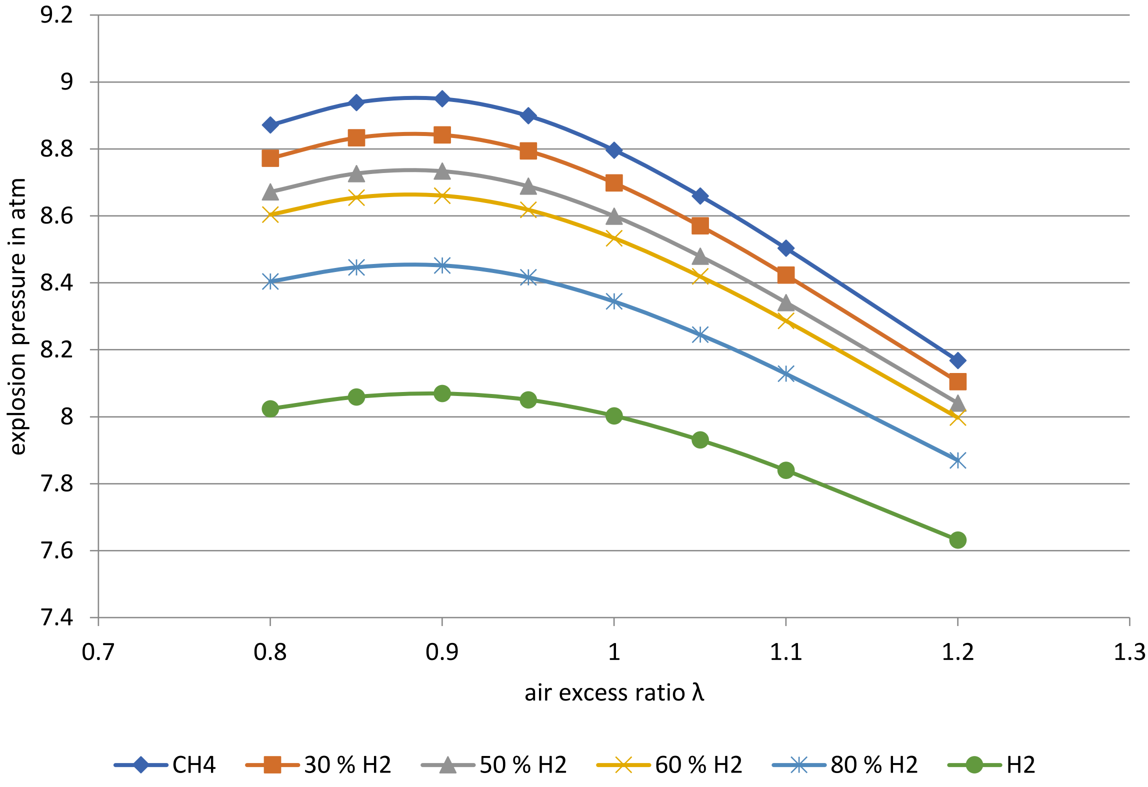

Figure 17: Maximum explosion pressures of various CH_4/H_2/air blends as functions of the air excess ratio

In addition to the pressure levels of a deflagration, the intensity of the deflagration also has to be considered. This intensity can be quantified by the so-called deflagration index K_G [17], defined as

$$K_G = \left(\frac{dp}{dt}\right)_{max} \cdot \sqrt[3]{V}\,, \qquad 12$$

with V as the volume of the vessel in the experiment and $\left(\frac{dp}{dt}\right)_{max}$ as the maximum pressure gradient over time. It can be shown that K_G is independent of the size of the vessel [17].

Figure 18 shows a comparison of experimental and simulation results to determine K_G as a function of the equivalence ratio Φ (the inverse of the air excess ratio) for methane (a) and hydrogen (b). It is obvious that hydrogen deflagrations yield a much higher deflagration index than methane deflagrations.

At the same time, the stoichiometry range at which significant K_G values can be found extends much farther into the sub-stoichiometric regime for hydrogen than for methane.

Thus, while maximum pressures are lower with hydrogen/air deflagrations than with methane/air deflagrations, the deflagration of the hydrogen/air mixture is much more forceful.

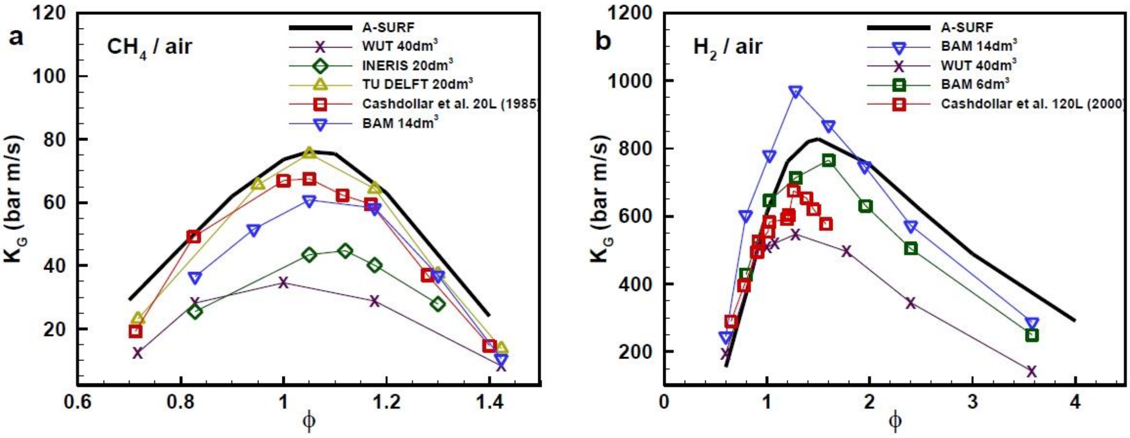

Figure 18: Deflagration indices of methane/air and hydrogen/air mixtures as functions of the equivalence ratios, based on experimental and simulation data [17].

7 Open issues

There are some aspects about the impact of hydrogen admixture to natural gas on combustion processes that theoretical considerations as explained in this report cannot entirely address.

One aspect would be the color of the flame, which may be relevant for the monitoring of the flame, but also for aesthetic purposes, i.e. in decorative fireplaces. To the best of the authors' knowledge, there is no way how flame colors can be predicted by combustion theory in a satisfactory manner. Instead, this topic may be addressed both as part of the literature review in the forthcoming deliverable D.2.3 and in the experimental campaigns of work package 3 of the THyGA project. A few fundamental research studies (e. g. [18], [19]) on the impact of hydrogen on flame colors were found, however, it has yet to be evaluated to what extent these findings can be transferred to residential gas fired appliances. Most of these experiments were conducted using very different burner designs compared to typical technology used for residential appliances.

Another important topic is flame ionization. This physical phenomenon is often used to monitor the flame. Flame monitoring is an important safety feature to ensure that gas is not conveyed into the combustion chamber if there is no flame present. Some research [13] shows that the presence of hydrogen in the base gas increases the ionization current, however, these experiments were carried out for industrial non-premixed burner systems at constant firing rate and air excess ratio. A further review of existing literature is required in this context.

8 Potential challenges due to hydrogen admixture and mitigation options

Considering that gas appliances in the market today were designed and optimised for the operation with natural gas (which does not usually contain any hydrogen at all), it is to be expected that a switch from natural gas to natural gas / hydrogen blends may pose challenges to the safe, efficient and environmentally friendly operation of such appliances. The severity of these challenges will depend not only on the type and design of an appliance, but of course also on the level of hydrogen in the blend. While many of these aspects have already been discussed elsewhere in this report, it may be helpful to point out the main points of concern and also highlight potential mitigation options in this chapter. It should be noted though, that there is a dedicated Work Package (Work Package 5) in the THyGA project which will explore mitigation options in greater detail.

One of the most common concerns when operating a residential or commercial gas appliance relates to CO emissions, as carbon monoxide is both toxic and flammable. It is therefore crucial that hydrogen admixture into the existing gas infrastructure should not lead to increased CO emissions from appliances.
As was shown previously, CO formation is closely tied to the air excess ratio λ and thus the adjustment of the appliance (cf. Figure 14). The presence of varying levels of hydrogen in the distributed natural gas means, however, that common adjustment practices for residential and commercial appliances may have to be reconsidered.

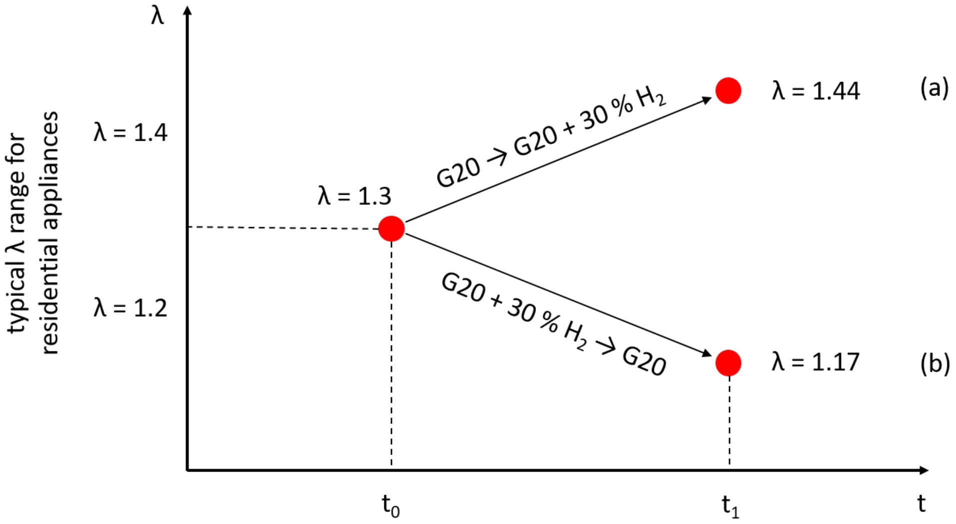

Figure 19: The impact of the initial appliance adjustment on the air excess ratio λ. Case (a) initial adjustment to λ = 1.3 with G20 (i. e. CH_4) at t = 0, then a switch to a blend with 30 % H_2. Case (b) initial adjustment to λ = 1.3 with a blend (70 vol. % CH_4 / 30 vol.-% H_2), then a switch to pure CH_4.

In theory, appliances covered by the Gas Appliance Regulation [20] and its predecessor, the Gas Appliance Directive [21], are to be adjusted by the manufacturer to a proper operational point, using a well-defined reference gas. For H-gas, this reference gas is defined in EN 437 [22] to be pure methane, called G 20. If an appliance adjusted in such a manner is then supplied with a natural gas / hydrogen blend, air excess ratios will increase if the air excess ratio is not actively controlled. Thus, the appliance will move towards an inherently safer state in terms of CO (cf. Eq. 9 and Figure 6), unless the initial air excess ratio is very high, so that a shift to even higher λ-values would cause flame instabilities.

Statistical surveys both in Germany [3], [23] and in France [24] indicate, however, that in practice, installers today generally adjust an appliance on-site to the local, usually unknown gas quality during commissioning, based on OEM-recommended values for either CO_2 or O_2 concentrations in the flue gas. This approach can already cause problems with fluctuating local natural gas qualities, but if higher and fluctuating levels of hydrogen are also present in the distributed gas, the question of how to adjust appliances properly becomes even more relevant.

If, for example, an appliance were to be adjusted with the locally available gas at the time of adjustment (which may contain a significant H_2 concentration) to a setpoint air excess ratio and is then supplied with a gas which contains less or even no hydrogen at a later stage, λ will actually decrease, hence potentially increasing the risk of CO formation.

Figure 19 visualizes how the gas that the appliance was initially adjusted to determines how the air excess ratio will shift in a gas quality change. In case (a), the appliance was adjusted to an air excess ratio of 1.3 with the reference gas G20, i.e. pure methane, as it is stipulated in EU regulations. If this appliance is then supplied with a blend of methane and 30 vol.-% hydrogen, λ will shift to higher values, in this case to about 1.44. In terms of CO emissions, this should be an even safer state.

If, on the other hand, the appliance was originally adjusted according to the current practice today, with the gas available on-site at the time of adjustment, the situation can be reversed. In case (b) of **Figure 19**, for example, the appliance is initially also adjusted to an air excess ratio of 1.3 but this time with a blend of 70 vol.-% CH_4 and 30 vol.-% H_2. The appliance is then supplied with pure methane at a later point in time. In this case, the air excess ratio of the combustion process will be reduced, and the appliance is more likely to produce CO.

It is recommended that the issue of appliance adjustment and its sensitivity to H_2 admixture should be explored in more detail in Work Package 5 of the THyGA project, which looks into mitigation strategies in more depth.

Another issue in this context is how the adjustment is actually carried out. The adjustment procedure via recommended CO_2 values causes increased uncertainty in the adjustment process as can be deduced from **Figure 20**. The diagram plots the evolution of the (dry) CO_2 concentrations in the flue gas of various fuel blends as a function of the air excess ratio λ. If, for example, an OEM recommends adjusting an appliance to a (dry) CO_2 concentration of 8 vol.-%, this would correspond to a λ of about 1.4 for pure methane, but only about 1.3 for a blend with 30 % H_2.

Adjusting appliances to recommended O_2 concentrations instead of CO_2 is far less sensitive in this regard, as is highlighted by **Figure 21**, which shows the relationship between λ and oxygen concentrations in the flue gas for various fuels. The excess oxygen in the flue gas is almost completely independent of the fuel composition, so that an appliance adjusted to 6 vol.-% excess oxygen will run at an air excess ratio of about 1.35, independent whether the adjustment was carried out with pure methane, a natural gas or a natural gas / hydrogen blend. The curves for the various fuel gases in Figure 21 are almost indistinguishable. Thus, adjustment via set point O_2 concentrations in the flue gas should be the preferred procedure, to reduce uncertainties in the settings.

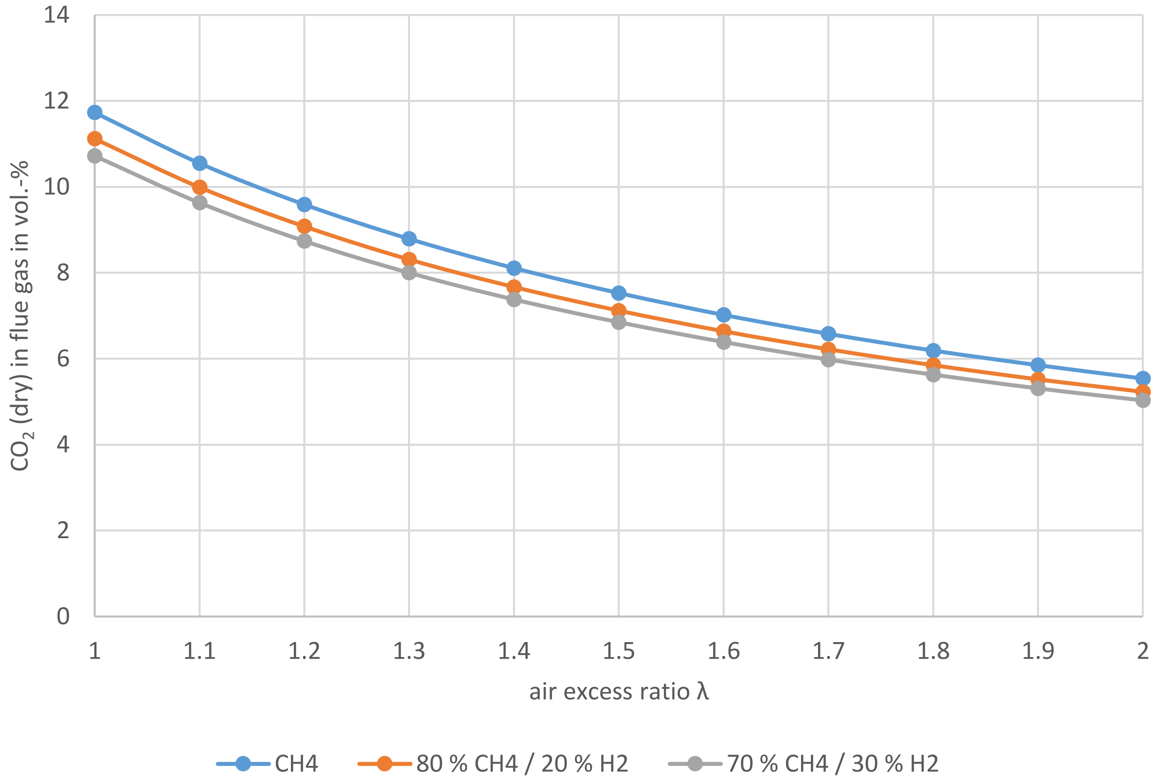

Figure 20: (Dry) CO_2 concentration in the flue gas of various fuel blends as a function of the air excess ratio.

The issue of the air excess ratio is also closely connected to the question whether a combustion control system is installed in a given appliance. Such a system, at least in the context of residential and commercial appliances maintains a constant air excess ratio in the appliance. Often, a flame ionization signal is used as input, since flame ionization probes are installed as a safety feature anyway to ensure that a flame is actually present in the combustion chamber.

While the combination of a flame ionization sensor to detect a change in the air excess ratio with a variable-frequency fan to adapt the air flow accordingly works very well to compensate for fluctuations of natural gas qualities (see for example [25], cf. **Figure 22**), both these investigations as well as the first analyses of the THyGA project show this approach is less successful when it comes to compensating gas quality changes due to hydrogen admixture.

Figure 22 gives an example of how an appliance with combustion control (based on a flame ionization signal) responds to various levels of H_2 in the fuel gas. For these measurements, the system was originally adjusted to an air excess ratio of about 1.38 at minimum load with pure methane, and then supplied with in increasingly higher levels of hydrogen in the fuel gas, up to 40 vol.-%. Fuel volume flows and oxygen concentrations in the flue gas were measured to determine the actual air excess ratio (λ, yellow line). The same procedure was then repeated, but at full load (right hand side of the diagram). The measurements indicate that while the combustion control system is able to maintain a constant air excess ratio for various levels of hydrogen at minimum load, it is unable to do so at full load. Instead, the air excess ratio (determined by a measurement of the oxygen concentration in the flue gas) increases with higher levels of H_2. A step-wise increase of the λ-value can clearly be seen. There is, however, at least some effect as the rise in λ is less pronounced as if this were an uncontrolled system.

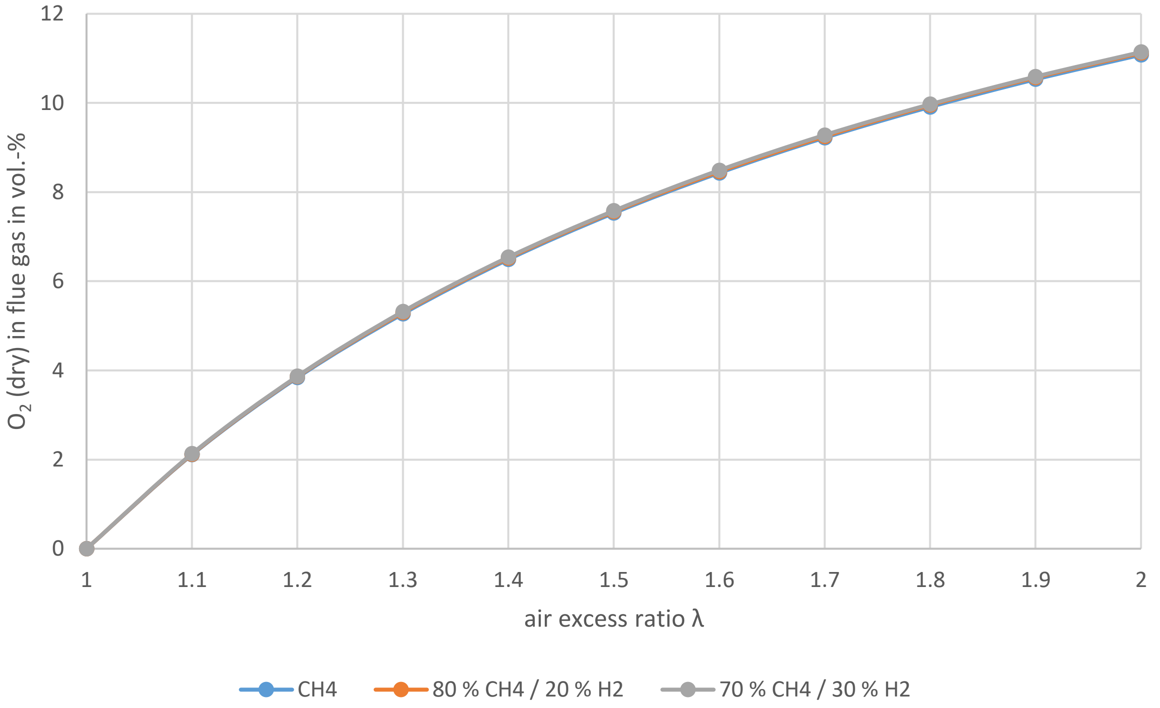

Figure 21: (Dry) O_2 concentrations in the flue gas of various fuel blends as a function of the air excess ratio. Data curves are almost indistinguishable, since the O_2 concentration is almost independent from the three chosen fuel compositions.

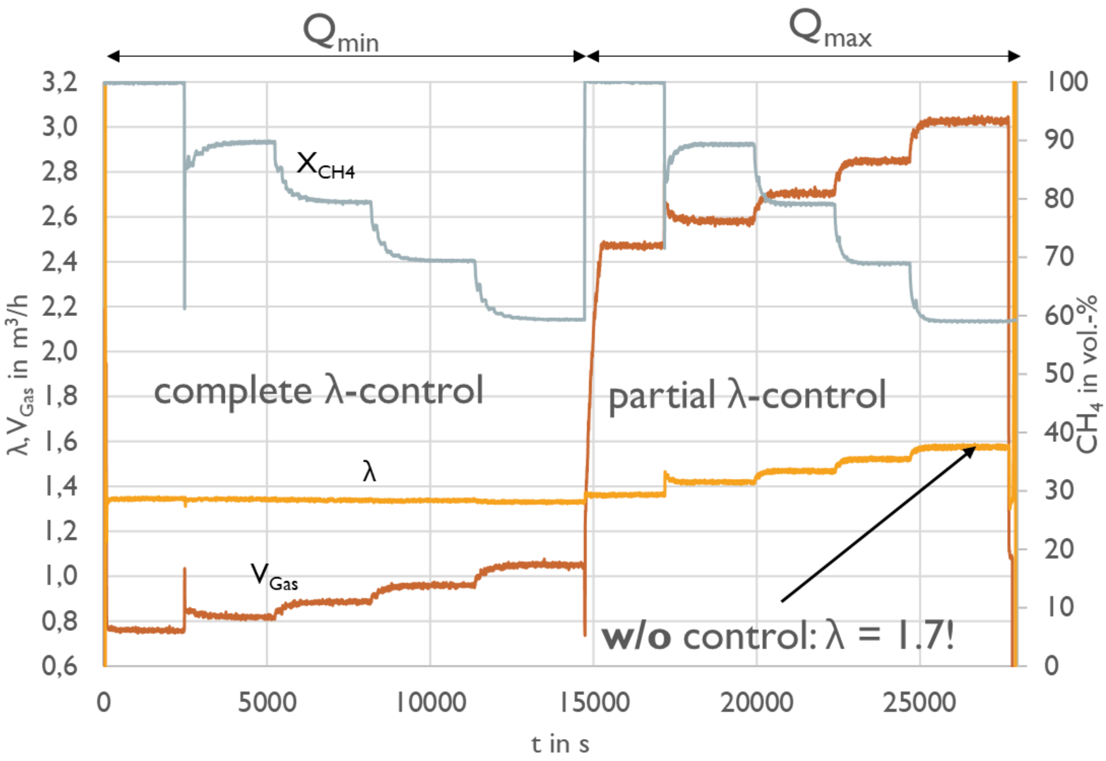

Figure 22: Measurements of the response of an appliance with combustion control to different levels of hydrogen in the fuel gas at different loads.

Other combustion-controlled appliances show a similar behaviour, both in the measurement campaigns of the THyGA project so far, but also in other studies [25], cf. **Figure 24**.

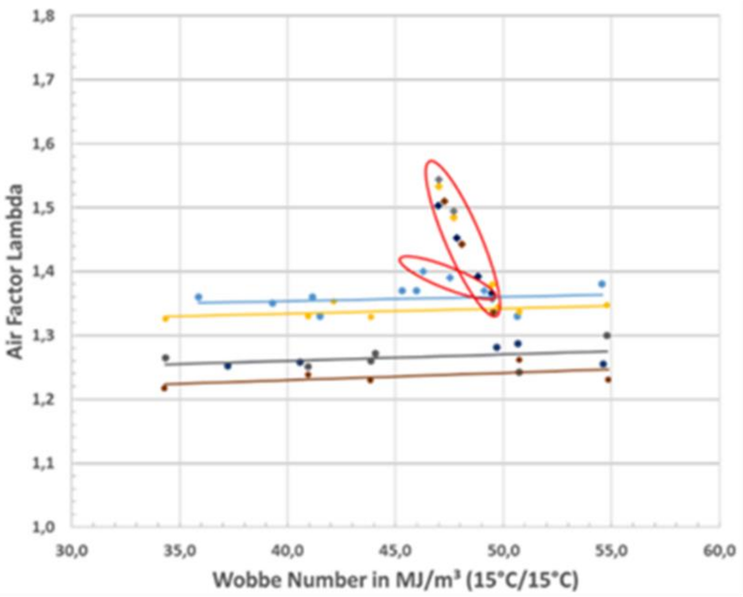

Figure 23: Tests of condensing boilers with combustion control for variable fuel gas qualities. The different colours indicate different appliances. The measurements highlighted in red are measurements where the Wobbe Index change is induced by hydrogen admixture [25].

The reason for this failure to maintain a constant λ-value at certain load points is likely connected to the principle of flame ionization measurement itself. In theory, the flame ionization current will have a maximum in a stoichiometric flame, i.e. $\lambda = 1$. The control system can then use the measured flame ionization current to re-adjust the combustion process to the setpoint air excess ratio. The flame ionization signal is, however, not only dependent on the stoichiometry and the composition of the fuel of the combustion process, but also on the relative position of the flame to the probe [26]. The very different combustion characteristics of hydrogen, compared to natural gas, are likely to cause a very different flame shape and position, at least in some load states, which leads to the inability of the measurement device to adequately determine the actual λ, resulting in an inadequate response of the control system. Thus, if combustion control based on flame ionization signal is to be used in the future, the existing systems will have to be adapted with hydrogen admixture in mind.

This possible explanation for the failure of ionisation-based measurement and control systems is corroborated by measurements with non-premixed burners [13] which showed an ionization current which was almost independent of the level of hydrogen in the fuel (experiments were carried out up to 50 vol.-% H_2). As was discussed previously, non-premixed flames are usually longer than premixed flames and the main reactions always take place at roughly stoichiometric conditions, independent of the global air excess ratio. Thus, the sensor is much more likely to be in an appropriate position to detect a flame in a non-premixed burner system and provide a good, unambiguous signal. A premixed flame, whose shape and position are much more sensitive to changing fuel compositions, may well establish itself in an unsuitable location, leading to ambiguous flame signals. It is worth pointing out that with extremely high H_2 levels in the fuel, the flame ionization signal will become too weak to be useful, even in non-premixed flames [27].

Another obvious approach to mitigate this effect of hydrogen admixture is not to use flame ionization as an input signal for combustion control at all, but to utilise another signal instead. In many combustion applications, λ control via excess oxygen measurement is very common and known to be insensitive to changing fuel compositions (cf. Figure 20). Another approach could be to deploy onboard gas quality sensors in appliances and use their output to properly control the combustion process. In both cases, the technology exists, but cost can be a restriction for their deployment in residential and commercial appliances.

There is, however, also another question to consider: is combustion control actually beneficial in the context of hydrogen admixture? Given the extremely high reactivity of hydrogen, one of the main safety-related concerns is the potential for flash-backs in residential and commercial appliances. Since most appliances use fully premixed laminar burners and the laminar combustion velocity increases with higher levels of hydrogen (cf. Figure 10), hydrogen admixture in principle increases the risk for flash-backs in burners that were originally designed for use with natural gas.

However, as previously shown, there are competing phenomena at play if the air excess ratio is allowed to shift towards higher values. Thus, in an uncontrolled combustion process (or in an appliance where the combustion control system is unable to maintain a constant air excess ratio), the higher air excess ratio largely compensates the effect of hydrogen admixture in the fuel gas. Higher values for λ reduce the laminar combustion velocity, at least for super-stoichiometric combustion processes, so the net change of the laminar combustion velocity (and hence the risk for flash-back) is relatively small, at least for moderate levels of hydrogen.

A similar effect can also be identified with regards to NO_X emissions. Although hydrogen admixture into natural gas leads to higher combustion temperatures (and hence potentially stronger formation of thermal NO_X) at a given air excess ratio, the change in stoichiometry counteracts this development to a large degree in a premixed burner. This, in addition to a shift of the flame position towards the burner (leading to increased heat flux into the burner material and thus lowering local temperatures [28]) tends to lead to an overall reduction of NO_X formation in residential appliances.

Given these effects, the necessity and benefits of combustion control systems for residential and commercial appliances should be discussed in more detail in Work Package 5, also taking into account the results of the experimental investigations.

It is worth pointing out that non-premixed burner systems behave very differently in this regard. Here, H_2 admixture usually leads to increased NO_X emissions [12] even if the global air excess ratio shifts towards higher values since the main heat release always takes place in a stoichiometric regime so that the higher local combustion temperatures cause increased NO_X formation.

Another mitigation strategy is to replace critical components like nozzles in appliances, similar to the conversion kits used for appliances in the market transition from L-Gas to H-Gas. In this manner, aspects like flash-backs and reduced firing rates due to H_2 admixture could be easily addressed. There are other things to consider here, however. It is estimated that there are more than 200 million gas appliances, of many different types and categories, in the residential and commercial sector in the European Union [29], so the logistical and organizational effort to convert the entire appliance population all across Europe is significant. Lessons could however be learnt from the successful realisation of the L-Gas → H-Gas market transition, e.g. concerning time spans needed, percentage of appliances to be changed, personnel deployment and costs monitored.

Also, on a more technical level, the question remains for which fuel gas and which hydrogen concentrations new components and appliances should be designed. The situation would become even more complex if manufacturers had to adapt to locally fluctuating hydrogen levels. Components would have to be designed in such a way that fluctuations of H_2 concentration would not result in safety-relevant consequences if the H_2 level in the supplied gas (or the gas quality of the natural gas that H_2 is admixed to) changes significantly from the gas that components were designed for.

The topic of mitigating hydrogen blending impact on appliances is also addressed in the upcoming work package 5 of the THyGA project.

9 Conclusion

The impact of different levels of hydrogen in methane/hydrogen blends on combustion and gas quality-related properties were discussed, using combustion theory, with a focus on residential and commercial appliances.

Given the significant differences between the physical and chemical properties of natural gas (represented here by pure methane, CH_4) and hydrogen, switching from natural gas to natural gas /hydrogen blends or even pure hydrogen can affect combustion processes in residential and commercial appliances in terms of performance, but also in terms of safety. It is obvious that the consequences will become more pronounced with higher levels of hydrogen in the fuel gas.

It can be seen that the response of a combustion process to higher levels of hydrogen is not only dependent on the change in the fuel properties, but to a large degree also on its concrete technological implementation. A premixed combustion process will react differently than a non-premixed system, and the presence of a combustion control system plays an important role as well. Thus, in a system without combustion control, the admixture of hydrogen will shift the stoichiometry of a premixed combustion process into the leaner region, which compensates the effects of hydrogen on flame temperatures, laminar combustion and NO_X formation to a large extent. A notable exception here is a combustion process intentionally run at sub-stoichiometric conditions, for example in the primary zone of a partially premixed cooking device. Here, the effects of the sub-stoichiometric combustion and hydrogen admixture will actually stack, causing increases in local temperatures and laminar combustion velocities.

The situation is different in combustion processes where the local air excess ratio is kept constant, either due to some kind of combustion control, or due to their very nature, for example in non-premixed combustion processes. Here, the hydrogen-induced shift of stoichiometry cannot occur, and as a consequence hydrogen admixture increases local temperatures, NO_X emissions and laminar combustion velocities.

In terms of safety, combustion theory shows that admixing hydrogen to natural gas should not lead to increased carbon monoxide emissions, except possibly for high air excess ratios due to flame instabilities. The maximum explosion pressure of hydrogen is also lower than that of methane, while the deflagration index is significantly higher.

One main finding for many common situations found in residential appliances is, that different effects of hydrogen compensate each other to a certain degree. For example, in uncontrolled residential combustion systems (which comprise the large majority of the residential appliance population in the EU), hydrogen admixture will result in a shift of the air excess ratio towards higher values which will largely counteract the increase of the laminar combustion velocity and combustion temperatures due to the presence of hydrogen.

Based on the theoretical considerations of the combustion of natural gas and natural gas / hydrogen blends and the expected impacts on the operation of residential and commercial gas appliances, a number of key issues were identified and potential mitigation options outlined.

In general, the distribution of hydrogen-natural gas blends is a viable option to decarbonise the combustion processes in the very heterogeneous natural gas sector to a certain extent. The theoretical background presented here must be taken into account when developing suitable equipment technology as well as the necessary rules and standards for hydrogen / natural gas blends. Typical existing technologies in the field may be used up to certain hydrogen admixture levels without causing safety risks, while some technologies, especially at high hydrogen additions, will have to be replaced. Detailed insight into the findings on the appliance technology level will be given in the future reports from the THyGA project[2].

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[2] Please visit https://thyga-project.eu/

11 References

[1] ‘GASQUAL Deliverable Approved by CEN/BT WG 197 “Gas Quality”’, CEN/AFNOR/WG 197 N 231, 2010 [Online]. Available: www.gasqual.eu

[2] Ourliac, M., ‘Deal with gas quality variations and melt glass with syngas from gasification’, Essen, Germany, 2017.

[3] Leicher, J., Giese, A., Görner, K., Werschy, M., Krause, H., Dörr, H, ‘Statistical Surveys on the Sensitivities of Industrial End-Users to Gas Quality Fluctuations in Germany’, presented at the International Gas Union Research Conference (IGRC), Rio de Janeiro, Brasilien, 2017.

[4] Leiker, M., Christoph, K., Rankl, M., Cartellieri, W., Pfeifer, U., ‘Evaluation of anticknocking property of gaseous fuels by means of methane number and its practical application to gas engines’, presented at the 9th International Congress on Combustion Engines, Stockholm, Sweden, 1971.

[5] Zepf, P., Stellwagen, K., ‘MWM MN Calculation Method’, presented at the European Sustainable Shipping Forum, 2014.

[6] Jones, H.R.N., *The Application of Combustion Principles to Domestic Burner Design*. Taylor & Francis, 1989.

[7] Hermanns, R.T.E., ‘Laminar Burning Velocities of Methane-Hydrogen-Air Mixtures’, Technische Universiteit Eindhoven, Eindhoven, The Netherlands, 2007.

[8] Smith, G. P., Golden, D. M., Frenklach, M., Moriarty, N. W., Eiteneer, B., Goldenberg, M., Bowman, C. T., Hanson, R. K., Song, S., Gardiner Jr., W. C., Lissianski, V. V., Quin, Z., 2000. [Online]. Available: www.me.berkeley.edu/gri_mech

[9] ‘Netherlands Technical Agreement NTA 8837 - Gas group K of the second gas family: gas composition, gas pressures and conditions for 1) normal use of the gas appliances; 2) testing of gas appliances’, NEN, 2012.

[10] Dzubiella, M., Hack, S., Gleim, E., Hesse, W., Vogt, A., Brämer, R., ‘Entwicklungsstand gasadaptiver Verbrennungsregelungssysteme für den Bereich der Gebäudebeheizung’, presented at the 28. Deutscher Flammentag, Darmstadt, Germany, 2017.

[11] Carpentier, S., Milin, P., Mostefaoui, N., Nitschke-Kowsky, P., Schweitzer, J., Sadegh, N., Thibaut, O., ‘Self-regulated gas boilers able to cope with gas quality variation - State of the art and performances’, 2018 [Online]. Available: https://www.dgc.dk/sites/default/files/filer/publikationer/R1804_self_regulated_boilers.pdf

[12] Leicher, J., Nowakowski, T., Giese, A., Görner, K., ‘Hydrogen in natural gas: how does it impact industrial end users?’, presented at the World Gas Conference 2018, Washington DC, USA, 2018.

[13] Nowakowski, T., Leicher, J., Giese, A., ‘Untersuchungen der Auswirkung von Wasserstoff-Zumischung ins Erdgasnetz auf industrielle Feuerungsprozesse in thermoprozesstechnischen Anlagen’, Gas- und Wärme-Institut Essen e.V., Essen, Schlussbericht IGF-Vorhaben Nr. 18518/1, 2017 [Online]. Available: https://www.gwi-essen.de/fileadmin/dateien/abschlussberichte/2017/18518N_H2_Subs_abschluss_korr.pdf

[14] Schröder, V., Askar, E., Tashqin, T., Habib, A.K., ‘Sicherheitstechnische Eigenschaften von Erdgas-Wasserstoff-Gemischen’, Bundesanstalt für Materialforschung und -prüfung, Berlin, Abschlussbericht zum Forschungsvorhaben 2539, 2016 [Online]. Available: https://www.bgetem.de/redaktion/arbeitssicherheit-gesundheitsschutz/dokumente-und-dateien/brancheninformationen/energie-und-wasserwirtschaft/gasversorgung/abschlussbericht-zum-forschungsvorhaben-2539-sicherheitstechnische-eigenschaften-von-erdgas-wasserstoff-gemischen

[15] Askar, E., Schröder, V., Schütz, S., Seemann, A., ‘Power-to-Gas: Safety Characteristics of Hydrogen/Natural Gas Mixtures’, *Chemical Engineering Transactions*, vol. 48, 2016, doi: DOI: 10.3303/CET1648067.

[16] Liberman, M.A., *Introduction to Physics and Chemistry of Combustion - Explosion, Flame, Detonation*. Springer-Verlag Berlin Heidelberg New York, 2008.

[17] Faghih, M., Chen, Z., ‘Deflagration Index of Methane/Hydrogen/Air Mixtures’, presented at the 10th Asia-Pacific Conference on Combustion, Beijing, China, 2015.

[18] Schefer, R.W., Kulatilaka, W.D., Patterson, B.D., Settersten, T.B., ‘Visible emission of hydrogen flames’, *Combust. Flame*, vol. 156, pp. 1234–1241, 2009.

[19] El-Ghafour, S.A.A., El-dein, A.H.E., Aref, A.A.R., ‘Combustion characteristics of natural gas-hydrogen hybrid fuel turbulent diffusion flame’, *International Journal of Hydrogen Energy*, vol. 35, no. 6, pp. 2556–2565, 2010, doi: 10.1016/j.ijhydene.2009.12.049.

[20] ‘Regulation (EU) 2016/426 of the European Parliament and of the Council of 9 March 2016 on appliances burning gaseous fuels and repealing Directive 2009/142/EC’, Brussels, Belgium, Regulation (EU) 2016/426, 2016 [Online]. Available: https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32016R0426&from=EN

[21] ‘Directive 2009/142/EC of the European Parliament and the Council of 30 November 2009 relating to appliances burning gaseous fuels (codified version)’, European Union, Brussels, Belgium, Directive 2009/142/EC, 2009 [Online]. Available: http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2009:330:0010:0027:DE:PDF

[22] ‘EN 437 Test gases -Test pressures - Appliance categories’, Comité Européen de Normalisation, Brussels, Belgium, European Norm EN 437:2003, 2003 [Online]. Available: http://www.sarm.am/docs/437.pdf

[23] Krause, H., Giese, A.., Dörr, H., Brückner, H.-J., ‘Hauptstudie zur Analyse der volkswirtschaftlichen Auswirkungen von Gasbeschaffenheitsschwankungen auf die Sektoren des Gasverbrauchs und deren Kompensation Phase 1 (Hauptstudie Gasbeschaffenheit)’, DVGW Deutscher Verein des Gas- und Wasserfaches e. V., Bonn, Germany, final report G1/01/15, 2016 [Online]. Available: http://www.dvgw-innovation.de/fileadmin/innovation/pdf/g1-01-15-phase1.pdf

[24] Ruillard, R., ‘L’ajustement sur site par les artisans chauffagistes’, Paris, France, 2012.

[25] Schweitzer, J., ‘Self-regulated gas boilers able to cope with gas quality variation: State of the art and performances’, Groningen, NL, 2019 [Online]. Available: https://www.en-trance.org/custom/uploads/2019/11/P2.6-Schweitzer.pdf

[26] Ding, Y., Durox, D., Darabiha, N., Schuller, T., ‘Combustion state monitoring of premixed heating appliances with flame ionization current and chemiluminiscence’, *Combust. Sci. and Tech.*, vol. 191, no. 2, pp. 382–401, 2019.

[27] Huber, A., ‘Auswirkungen von Wasserstoff und Erdgas-Wasserstoffgemischen auf Gasgebläsebrenner’, *Prozesswärme*, no. 4, pp. 31–37, 2020.

[28] Hinrichs, J., Hellmuth, M., Meyer, F., Kruse, S., Plümke, M., Pitsch, H., ‘Investigation of nitric oxide formation in methane, methane/propane and methane/hydrogen flames under condensing gas boiler conditions’, *Applications in Energy and Combustion Science*, vol. 5, p. 100014, 2021, doi: https://doi.org/10.1016/j.jaecs.2020.100014.

[29] Flayyih, M., Schaffert, J., Burmeister, F., Albus, R., Görner, K, Milin, P., Carpentier, S., Krishnaramanujam, K., Endisch, J., de Wit, K., Geerts E., Schweitzer, J.., ‘Market segmentation of domestic and commercial natural gas appliances’, GWI, ENGIE, EBI, gas.be, DGC, 2020 [Online]. Available: https://thyga-project.eu/wp-content/uploads/20200703-D2.2-Impact-of-hydrogen-admixture-on-combustion-processes-%E2%80%93-Part-I_Theory.pdf