H2AL

DECARBONISING ALUMINIUM RECYCLING WITH HYDROGEN

D3.3 – Technologies for optimized H_2 combustion:

Report on the designs and specifications of the systems layout and components integrations

VERSION 2.0

Clean Hydrogen Partnership

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Executive Summary

Climate change is one of the most pressing challenges of the 21st century. It is closely tied to anthropogenic greenhouse gas emissions, mostly resulting from providing and using energy across all fields of human activity. The energy-intensive basic materials industries are among the largest consumers of energy. Most of the energy is required as process heat, to manufacture materials and products ranging from steel and cement to non-ferrous metals or the products of the chemical industry. Process heat is today dominated by fossil fuels, particularly natural gas. As such, energy-intensive industries are responsible for significant greenhouse gas emissions, but are also essential for human society.

While electrification (using electricity from renewable sources) is one promising approach to decarbonize energy-intensive industries, electric process heating can also have drawbacks, especially in the context of high-temperature process heat. This is why alternative fuels, in particular hydrogen from renewable sources, are considered as valuable decarbonization options, especially for equipment and processes that are today operated with natural gas.

Aluminium recycling is one of those processes where hydrogen combustion for process heating is of particular interest. This deliverable discusses industrial combustion in general, and then highlights the differences between the combustion of natural gas, the most relevant fuel in secondary aluminium production today, and hydrogen.
The question of how industrial combustion processes and the corresponding equipment have to be adapted for optimum performance with the new fuel is therefore explored in detail. Given the high degree of specialization, the challenging requirements in terms of fitness-for-purpose, efficiency and pollutant emissions in industrial process heating, a profound understanding of the combustion of both natural gas and hydrogen is required to achieve the desired results, especially when it comes to retrofitting existing equipment. This deliverable thus aims at providing the necessary theoretical background to carry out adaptations and modifications of industrial combustion equipment.

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List of abbreviations

Air_{min}	minimum air requirement
ARF	Flue gas recirculation (German: Abgasrückführung)
CFD	computational fluid dynamics
CH_4	methane
CO	carbon monoxide
COG	coke oven gas
CO_2	carbon dioxide
FGR	flue gas recirculation
FLOX©	flameless oxidation
GHG	greenhouse gas
GCV	gross calorific value
d	relative density
η_{th}	thermal efficiency of a combustion process
H	atomic hydrogen
H_2	hydrogen
H_i	net calorific value or inferior heating value
$H_{i,m}$	net calorific value, mass-based
$H_{i,V}$	net calorific value, volume-based
H_S	gross calorific value or superior heating value
$H_{S,m}$	gross calorific value, mass-based
$H_{S,V}$	gross calorific value, volume-based
H_2O	water (vapour)
IR	infrared
ISO	International Standards Organization
λ	air excess ratio
LEL	lower explosion limit
LOC	limiting oxygen concentration
$\dot{m}_{fuel}$	mass flow of fuel
$\dot{m}_{O2}$	mass flow of oxygen
MIE	minimum ignition energy
MILD	Moderate or Intense Low-Oxygen Dilution
N	atomic nitrogen

N_2	nitrogen
NCV	net calorific value
NO	nitric oxide
NO_2	nitrogen dioxide
$NO_{X'}$	nitrogen oxides
O	atomic oxygen
O_2	oxygen
O_{2min}	minimum oxygen requirement
OH	hydroxyl
P	firing rate
$\dot{Q}_{flue}$	Energy flow in the flue gas
$\dot{Q}_{process}$	Process heat flow
$\rho_{n,airl}$	standard density of air
$\rho_{n,fuel}$	standard density of the fuel
S	Swirl Number
SCR	selective catalytic reduction
s_L	laminar combustion velocity
SNCR	selective non-catalytic reduction
s_t	turbulent combustion velocity
T_{ad}	adiabatic combustion temperature
T_{flue}	flue gas temperature
TRL	Technological Readiness Level
u	flow velocity
u_{fuel}	flow velocity of the fuel at the inlet
u_{O2}	flow velocity of the oxygen at the inlet
UEL	upper explosion limit
UV	ultraviolet
$\dot{V}_{air,actual}$	actual volume flow of air
$\dot{V}_{air,min}$	minimum volume flow of air (i.e. at stoichiometric conditions)
VDMA	German Association of Machinery and Equipment Manufacturers (Verband Deutscher Maschinen- und Anlagenbau e. V.)
$\dot{V}_{fuel,n}$	standard volume flow of fuel

$V_{flue,dry,min}$	minimum dry fly gas volume
$\mathbf{V_{n,flue,\ wet}}$	standard wet flue gas volume per unit of heat released
W_i	net or inferior Wobbe Index
W_S	gross or superior Wobbe Index
X_{NOx}	volume fraction of NO_X in the flue gas
$X_{O2,meas}$	measured volumetric O_2 concentration in the flue gas

1. Introduction

Climate change is one of the major global challenges of the 21st century. Anthropogenic emissions of greenhouse gases (GHG), primarily in the form of carbon dioxide (CO_2) and methane (CH_4), have been identified as a driving force for global warming. The majority of these emissions is connected to the combustion of fossil fuels such as coal, oil or natural gas, which provided for more than 80% of the global primary energy demand in 2023 [1]. Decarbonisation, *i.e.* the reduction of these GHG emissions by phasing out the use of fossil fuels with their inevitable CO_2 emissions, is considered as one of the most important measures to combat climate change and mitigate its consequences. At the same time, the ready availability of energy is a prerequisite for a modern society. Given a growing world population and improving standards of living world-wide, global energy demand is expected to grow [2].

Energy is required in different forms for different applications, ranging from electricity in household appliances or data centers, to heat in buildings and industrial manufacturing processes or mechanical energy for the propulsion of vehicles. For heat, in particular, the demand is substantial: in Germany, as an example for an advanced industrialized economy, residential heating and industrial process heat account for more than 50% of the country's final energy demand [3]. Fossil fuels are still the dominant energy source, both for residential and industrial heating. Both electrification of heat production and the combustion of alternative carbon free fuels such as hydrogen are expected to play significant roles in the decarbonisation of process heat, especially in high-temperature applications in energy-intensive industries [4-7].

Compared to a more conventional fuel like natural gas, hydrogen is different in many respects. These differences have to be considered when discussing converting established and mature industrial manufacturing processes to hydrogen. Questions arise not only in the context of combustion itself, but also in related topics such as process and energy efficiency, heat transfer, operational safety or pollutant emissions (in particular, nitrogen oxides (NO_X)). From the point of view of a manufacturer, product quality is of paramount importance.

In this report, some key concepts of industrial combustion for process heating will be explained, ranging from fuel properties like calorific values or combustion velocities to technologies typically found in the sector, such as waste heat recovery for air preheating, oxy-fuel combustion or flameless oxidation/MILD combustion. Safety-relevant properties like ignition limits or minimum ignition energies will be addressed as well. Natural gas and hydrogen will be compared with regards to their properties, and the effects of the changing fuel properties on industrial firing applications will be discussed, based on theoretical considerations and a literature review.
Some general guidelines will be provided on how to use hydrogen for industrial high-temperature process heating in a safe, efficient and environmentally friendly manner.

2. Industrial process heating: facts and figures

The use of heat is ubiquitous in human society, across many sectors. While heat is generally used directly in the residential sector, industrial process heat is an indirect form of heat utilisation: heat is required as part of a manufacturing process for many products, which may range from food and beverages to textiles or paper and to essential materials like steel, glass, ceramics, non-ferrous metals, cement or the many products of the chemical industry. Thus, industrial process heating is a means to an end, and product quality is of paramount importance.

Almost 70% of Germany's final energy demand in the industrial sector alone is accounted for by process heat demand, which translates into more than 20% of the entire country's final energy demand [3] (cf. **Figure 1**). Process heating is also clearly dominated by fossil fuels, natural gas in particular. Roughly 50% of the process heat demand in various industries is supplied by natural gas, while coal contributes another 24%. Renewable energies or electricity play only relatively minor roles today in the context of process heating, as is visualised in the pie chart of **Figure 2**. The situation is rather similar in other industrialised countries around the world [4, 8, 9]. While the industrial sector actually uses a significant amount of electricity (according to [3] almost 30% of its total energy demand, second only to natural gas in terms of energy carriers), electric power is mostly needed for drive applications, *e.g.* pumps, compressors or grinding mills, not for process heating.

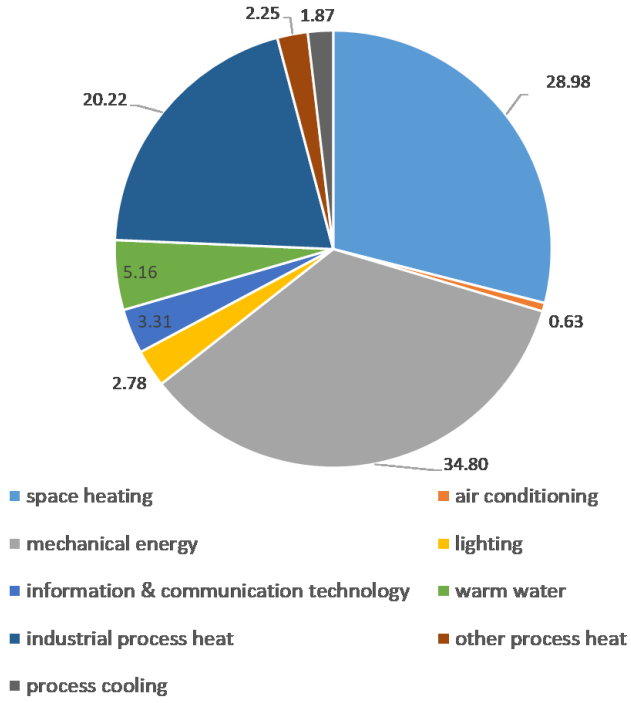


Figure 1: Energy use in Germany, 2021 (Data from [3])

On a global scale, almost 30% of the world's GHG emissions are caused by the industrial sector, the vast majority (> 80%) is produced by energy-related applications, while the rest is accounted for by product-related so-called process emissions, *e.g.* CO_2 formation due to calcination processes in the cement industry [10].

It is generally accepted that industrial process heating, especially high-temperature process heating, is one of the hard-to-abate sectors in the context of decarbonisation [7, 11]. There are several reasons why: first of all, it is a very diverse and heterogeneous sector. Given the wide variety of products, ranging from paper and textiles to energy-intensive basic materials such as steel, cement or glass, the processes, requirements and technologies are highly specialised and optimised for their specific purposes [12, 13].

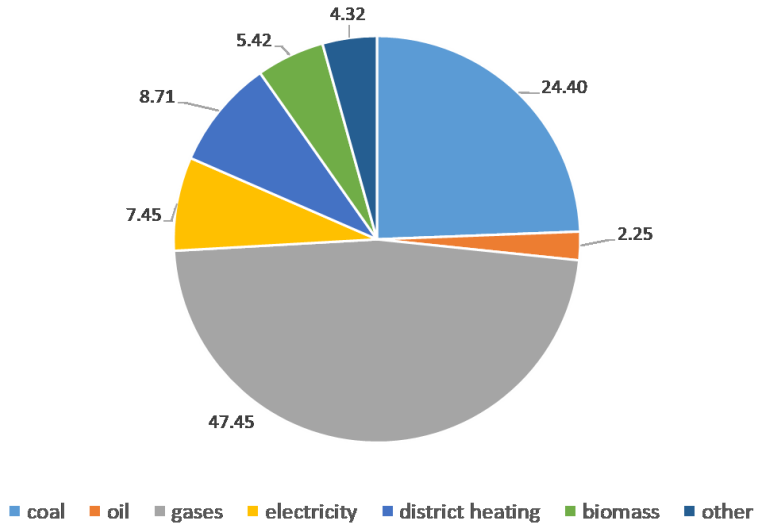


Figure 2: Contribution of energy carriers for process heating in Germany 2021. (Data from [3])

This heterogeneity of industrial process heating is highlighted by **Figure 3**, which shows the process heat demand in Germany as a function of the respective industries and also the required process temperatures [12]. While there are many processes in the low- and medium temperature range, *e.g.* for drying or steam generation, about 50% of industrial process heat is needed at temperatures of more than 400°C [8, 12]. This is particularly relevant in the context of decarbonisation as high-temperature process heat generally is harder to decarbonise than low-temperature applications, especially when high energy densities are required as well [4, 7, 8].

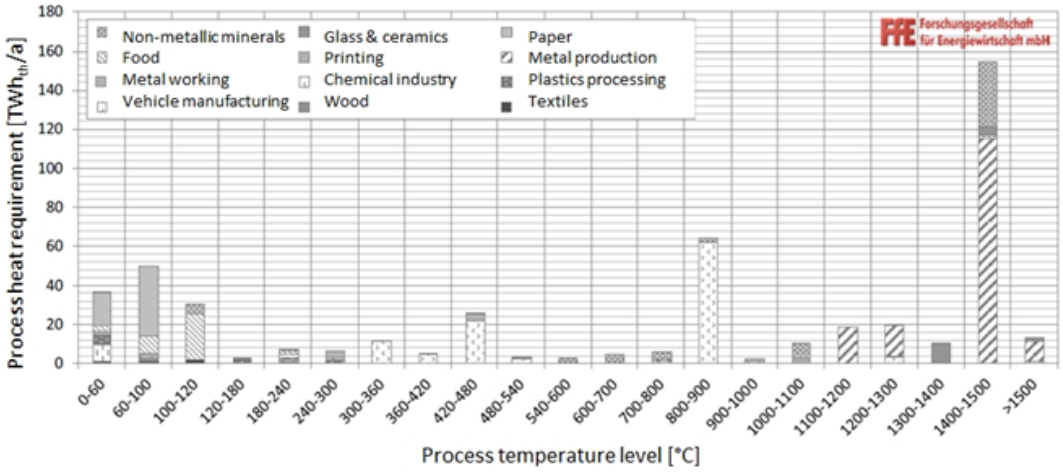


Figure 3: Process heat demand in Germany (translated from [12])

In the secondary aluminium industry for example, the focus of the H2AL project, process temperatures of about 800°C are required to re-melt aluminium scrap. It is worth noting

that recycling aluminium scrap is by itself a valuable contribution to combating climate change: not only does it promote a circular economy by re-using existing material, it is also far less energy-intensive than the primary aluminium route, as the energy demand per mass unit of aluminium is reduced by about 95% [14].

Industrial process heating is a means to an end, the focus is always on the product being manufactured. As a consequence, product quality is of paramount importance. Process heating is also often not about heat transfer alone. Frequently, additional phenomena like the interaction of the product with the furnace atmosphere or the exposure of the product to well-specified temperature-time profiles are crucial for achieving the desired physical properties in the materials [15]. Some products are more sensitive than others, and in some instances, even small deviations in process operation can cause unacceptable changes in product quality.
In the context of aluminium scrap recycling for example, one concern is the contamination of the scrap with organic residues from paints, lubricants or glues. These residues can interact with the aluminium surface when remelting, which can reduce both product quality and material yield, and hence, productivity of the process. As a consequence, electrically heated recycling furnaces tend to require higher scrap qualities than gas-fired furnaces, and are also smaller units in terms of production rates [16].

There are additional specifics to consider in the industrial sector, ranging from strict environmental requirements, *e.g.* in terms of air quality relevant pollutants such as nitrogen oxides (NO_X), to cost considerations, and international competition. Often, especially in high-temperature applications, equipment is operated continuously or quasi-continuously over long periods of time. With high-temperature equipment, an unanticipated shutdown can lead to damage to the furnace, which means that in addition to energy efficiency, a secure, uninterrupted supply of energy is often essential. This can limit the viability of demand side management plans, though this is very dependent on the specific process [17].
Energy costs, but also security of energy supply are locational factors, and in contrast to private citizens, industrial operators are more likely to shift their production elsewhere if local conditions are no longer economically tenable.

These specifics of the industrial sector have to be taken into account when considering decarbonisation strategies for this sector. Just like the products themselves, solutions will be highly specialized and tailored to the various industries and their specific demands. Given the huge amounts of energy involved and the critical need for continuous security of supply in many cases, the decarbonisation of energy-intensive industries will also have consequences for the expansion of the necessary low-carbon energy infrastructures. Studies [18] show that due to the high, generally continuous demand for energy in energy-intensive manufacturing processes, decentralised energy supply, *e.g.* with local wind farms or PV panels, will usually not suffice to meet the demand of these processes whose power demand can be in the double- or even triple-digit Megawatt range, depending on industry and application [19].

The two main pathways towards decarbonising high-temperature process heat are generally considered to be electrification or the use of hydrogen. This does not mean, however, that other decarbonisation pathways do not exist: the cement industry, for example, with its large share of process emissions [15, 20], is expected to focus on a combination of biomass and refuse-derived fuels (RDF) and CCS technology (CCS: carbon

capture and storage) to meet its emissions goals [11]. Ammonia (NH_3) with its favorable storageability may be a promising option as well, especially for industrial sites that are not directly connected to a hydrogen grid.
Generally speaking, however, electrification and hydrogen are two of the most promising options to decarbonise high-temperature process heating [4, 7]. In both cases, it is important that the energy carriers are produced from low-carbon sources, so that GHG emissions are not simply shifted, without an actual reduction of emissions, from the manufacturing process itself to a conventional power plant or, in the case of hydrogen, a steam methane reformer [21].
Both energy carriers have their specific advantages and disadvantages (cf. [6-8, 22]): direct electrification is generally more energy-efficient than using hydrogen produced from water electrolysis with renewable electricity, but often limited in the production rates that can be achieved, due to constraints of achievable heat flux densities. Existing manufacturing processes (and the corresponding equipment) can more easily be adapted and retrofitted to a new fuel than to a completely different heating technology. Wind and solar power are intermittent, and battery storage is limited in scale, while hydrogen as a chemical energy storage medium can in principle be stored in a similar manner as natural gas, though challenges remain.

Looking specifically at aluminum recycling, many of the advantages for the use of hydrogen can be found: inferior scrap qualities can be used as there is still a combustion process to burn off organic residues before they impact product quality and material yield, and existing plants can be retrofitted relatively easily, compared to switching to fully electrified systems. There are, however, also challenges: product quality is a concern as hydrogen is known to be easily soluble in aluminium, and there are also open questions about the interaction of both hydrogen and the higher water vapour content in the flue gas with typical refractory materials. Also, the necessary grid-level hydrogen infrastructures are still in the early stages of development.

As hydrogen is chemically a very different fuel compared to natural gas and given the high degree of heterogeneity in industrial process heating, existing processes and technologies will have to be adapted to the different fuel characteristics, while maintaining the high-performance level of industrial firing equipment. This report will describe some of the physical and technological aspects involved in the conversion of existing natural gas-fired equipment and processes to the use of hydrogen.

3. Key concepts and definitions in technical combustion

Combustion remains a key technology that is being utilised in all aspects of energy utilisation, from power generation to transportation to providing heat in both the residential and industrial sectors [23]. As the H2AL project investigates how to best switch from a conventional and well-established fuel like natural gas to a carbon-free fuel like hydrogen in order to decarbonise a hard-to-abate application such as aluminium scrap recycling, the impact of such a fuel change on the combustion processes has to be considered.

In this section, some fundamental aspects of combustion in a process heating context will be explored. This section serves both to introduce key concepts and definitions of combustion processes and the relevant fuel characteristics and process parameters such as calorific values, firing rates or air excess ratios, as well as to explain aspects such as thermal efficiency or pollutant formation (with a focus on nitrogen oxides (NO_X)).

The two fuels in the focus of this project, natural gas and hydrogen, will be compared in more detail in **Chapter 5**, while the consequences for a conversion of an industrial combustion process will be discussed in general terms in **Chapter 6**.

Basic combustion

In any combustion process, a fuel (*e.g.* natural gas or hydrogen) and an oxidiser (usually air) react with one another to form reaction products like carbon dioxide (CO_2) and water (H_2O). In this process, some of the energy chemically bound in the fuel is released as heat which then can be used, for example to melt metal.

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 or nitrogen (N_2), methane is often used as a reference gas, since natural gas usually consists of at least 90 vol.% of CH_4. The differences between various natural gases are in many ways negligible compared to the difference between natural gas and hydrogen.

Physically, the combustion of hydrogen with oxygen (O_2) is the simplest combustion process possible and can be described by the following equation:

$$1H_2 + \frac{1}{2}O_2 \rightarrow 1H_2O + heat.$$

For comparison, the combustion of CH_4 with O_2 is given by:

$$1CH_4 + 2O_2 \rightarrow 1CO_2 + 2H_2O + heat.$$

In technical combustion processes, combustion with air is much more common, as ambient air is easily available. Air consists of about 21 vol.% O_2 and 79 vol.% N_2 so that the reaction equations change as follows:

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

and

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

These equations already highlight two major differences between methane and hydrogen combustion: a hydrogen molecule needs fewer oxygen molecules to be completely converted to its reaction product than a methane molecule, while the flue gas composition (right-hand side of the equations) is also different since methane forms both CO_2 and H_2O as stable reaction products.
It should be noted that these equations sum up a large number of intermediate reaction steps. For a generalised description of a combustion process, such a simplified approach is sufficient. For the investigation of specific aspects such as ignition, flame stabilisation or pollutant formation, more detailed and comprehensive descriptions of the chemical processes are needed, generally in the form of chemical reaction mechanisms which may contain dozens of species even for relatively simple fuels, and hundreds of elementary chemical reactions.

Minimum O_2/air requirement, stoichiometry and air excess ratio

The minimum amount of oxidiser needed to completely convert a fuel is a characteristic property of the fuel. It is called minimum oxygen or air requirement (O_{2min} and Air_{min}, respectively), and it is only dependent on its chemical composition. For methane, O_{2min} is 2 mole O_2/mole CH_4, for hydrogen, it is 0.5 mole O_2/mole H_2. Assuming ideal gases (a valid assumption for most technical combustion processes), these values can also be given by standard volumes as 2 m^3 O_2/m^3 CH_4 and 0.5 m^3 O_2/m^3 H_2, respectively.

If a fuel is burned with exactly the amount of oxidiser needed for complete conversion, this is called a stoichiometric combustion. While theoretically, such a process is the ideal combustion process in many ways, it is hardly ever found in technical applications. Instead, technical combustion processes are usually operated with an excess of oxygen or air. There are several reasons for this: stoichiometric combustion requires perfect mixing of fuel and oxidiser at all times which can be difficult to ensure in real-life applications. If however, a carbon-containing fuel (which means almost all technically relevant fuels today) is not provided with sufficient oxygen during combustion, it will result in the formation of carbon monoxide (CO), a toxic and flammable pollutant species.
Supplying more oxidiser than theoretically necessary is thus an important and easy-to-implement safety feature found in most technical combustion processes, across all

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

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

with $\dot{V}_{air,actual}$ as the volume flow of air (or any other oxidiser), $\dot{V}_{air,min}$ the minimum volume flow of air required for complete combustion and $\dot{V}_{fuel}$ as the volume flow of fuel. Air_{min}, as stated previously, is the minimum air requirement.

It is worth pointing out that while Air_{min} (or O_{2min}, in the case of combustion with pure oxygen), are characteristic fuel properties which are only dependent on its chemical composition, the air excess ratio of a combustion process is not. Instead, it is an important process parameter, chosen by the operator of the process to be best suited for the specific purpose of the process. Therefore, λ values can be very different, depending on the specific application. In residential heating appliances, for example, air excess ratios are usually set around 1.2 to 1.4 [24, 25], *i.e.* these appliances are adjusted to run with 20 to 40% more air than theoretically necessary. Industrial furnaces, on the other hand, tend to be operated much closer to stoichiometry, with values of 1.1 or even less being quite common in industrial heating equipment [26, 27]. Some industrial processes are even intentionally set to sub-stoichiometric combustion (λ < 1), usually for process-related reasons. In copper recycling, for example, the copper scrap is very sensitive to any oxygen present in the flue gas as it will greatly affect product quality. Burners for such applications are therefore run sub-stoichiometrically to prevent the presence of any oxygen in the flue gas. The resulting flue gas will contain combustible components, so it will be conveyed to some kind of thermal post-combustion equipment to recover this energy of the not fully converted fuel and ensure operational safety.

The choice of an appropriate value for λ is often crucial for a given application since the air excess ratio can have a significant impact on a combustion process in terms of temperatures, flame shape, efficiency or pollutant performance, to name just a few aspects. There can also be an impact on product quality.

The differences between hydrogen and natural gas in terms of O_{2min} or Air_{min} also indicate that the oxidiser supply will have to be adapted if the fuel is changed.

Calorific values and firing rates

The purpose of technical combustion processes generally is to transform the energy chemically bound in the fuel into heat which can then be used. Therefore, the energy content of a fuel is obviously important. This energy density is usually expressed in the form of calorific values, a characteristic fuel property which is a function of the chemical fuel composition. Calorific values for gaseous fuels are often expressed volumetrically, *i.e.* in units such as [MJ/m^3] or [kWh/m^3], referenced to a standard state of temperature and pressure. Mass-based information can also be found, often in [MJ/kg]. **Table 1** gives the values the net and gross calorific values (NCV and GCV, H_i and H_S) in two common reference systems as well as in mass-based units.

Table 1: Calorific values of methane and hydrogen in various units and reference systems. Data calculated with [28]

Property	Unit	CH_4	H_2
$H_{i,vol}$ (@ 15 °C / 15 °C, 1.01325 bar)	MJ/m^3	34.06	10.24
$H_{i,vol}$ (@ 25 °C / 0 °C, 1.01325 bar)	MJ/m^3	35.89	10.79
$H_{S,vol}$ (@ 15 °C / 15 °C, 1.01325 bar)	MJ/m^3	37.80	12.10
$H_{S,vol}$ (@ 25 °C / 0 °C, 1.01325 bar)	MJ/m^3	39.83	12.75
$H_{i,m}$ (mass-based)	MJ/kg	50.03	120.01
$H_{S,m}$ (mass-based)	MJ/kg	55.51	141.78

The calorific values of a fuel can be directly calculated from the fuel composition, using an energy balance of the combustion process and the standard formation enthalpies of fuels and reaction products. As these standard formation enthalpies are referenced to a specified reference temperature, and to the standard volume needed to express calorific values in [MJ/m^3] also requires a reference temperature (and pressure), the values for NCV and GCV are themselves dependent on the choice of a reference system. Unfortunately, there are two competing systems within Europe. The official reference system, as specified in [29], works with 15°C and a pressure of 1.01325 bar, following ISO conventions. However, many major European countries such as Germany, the Netherlands and Poland use a system with two reference temperatures (25°C/0°C) (cf. for example the German gas quality specification [30]). The first temperature refers to the temperature that the standard formation enthalpies used for the calculation are referenced to, while the second temperature is the one used to calculate the standard volume. The reference pressure is still 1.01325 bar. In the ISO-based reference system, the same temperature is used both for the calculation of the caloric properties and to define a standard volume.
More important from a physical point of view, however, is the difference between gross and net calorific value. When calculating the net calorific value, it is assumed that the reaction product H_2O is present in the flue gas in gaseous form, while the calculation of GCV assumes that the water has condensed. This means that GCV and NCV differ by the condensation enthalpy of the water in the flue gas.

Based on the calorific value of a fuel, the firing rate of P a combustion process, *i.e.* the amount of energy delivered by the fuel per unit of time, can be determined. For most technical combustion processes, the net calorific value is the relevant fuel property as condensation of the water vapour in the flue gas is usually not desirable in industrial combustion processes. Hence, the firing rate P can be calculated as

$$P = \dot{m}_{fuel} \cdot H_{i,m} = \dot{V}_{fuel,n} \cdot H_{i,vol}$$

with [W] as SI unit. This means that in order to provide the same firing rate, *i.e.* energy input per unit of time, the quantities of fuel have to change if the fuel composition itself is changed.

Wobbe Index and fuel gas interchangeability

One obvious question that can arise for the use of combustion applications is how these devices respond to changes in the fuel. This is particularly relevant for the use of natural gas whose composition can differ significantly depending on the origin of the gas, pre-processing or mixing processes in the gas grid. The most popular criterion in the natural gas industry to assess the interchangeability of fuels is the so-called (superior) Wobbe Index W_S, defined as

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

with $\rho_{n,fuel}$ and $\rho_{n,air}$ as the standard densities of fuel gas and air respectively. d is called the relative density, or in the U.S., specific gravity. The inferior Wobbe Index W_i is more common from an end-user's perspective and is calculated using the net calorific value instead of the gross calorific value. This equation can be derived from Bernoulli's equation, assuming constant nozzle pressure and diameter [31].

In theory, two fuels with the same Wobbe Index will thus release the same amount of heat per time in a combustion system, provided the nozzle geometry (*i.e.* the diameter) and pressure do not change. Therefore, a given combustion device could be operated with both gases and still fulfill its purpose without the need to change it physically. Alternatively, if the Wobbe Index of the supplied gas changes (*e.g.* from an initial state 0 that the appliance was adjusted for to the current state 1), the firing rate in a combustion device will change with the ratio of the Wobbe Indices, following

$$\frac{P_1}{P_0} = \frac{W_{i,1}}{W_{i,0}}$$

These assumptions are generally valid for residential appliances, although combustion systems for other applications, particularly thermal processing industries, can require a different approach [26, 32]. If there is any kind of control for the firing rate, the Wobbe Index is no longer relevant, *e.g.* if the nozzle pressure is actively modulated to maintain a setpoint firing rate.

Due to the importance of the Wobbe Index 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 (see for example [30, 33, 34]).

It is worth pointing out that the Wobbe Index only considers heat release in a combustion process, while other important aspects such as combustion stability, pollutant emissions or efficiency are not taken into account. This is particularly relevant when looking at chemically very different fuels such as natural gas and hydrogen which will be discussed in more detail later.

Adiabatic combustion temperature

The adiabatic combustion temperature T_{ad} is closely related to the calorific values and can be calculated by means of an energy balance. Though it is not strictly speaking a fuel property (as it is also dependent on process parameters such as the pressure, the air

excess ratio or potential air preheat temperatures), it is nevertheless a useful criterion to assess the impact of a change in the combustion process (*e.g.* switching fuels or changing air excess ratios). The adiabatic combustion temperature is defined as the theoretical temperature of the flue gas in an adiabatic combustion process, *i.e.* the released heat is used exclusively to increase the temperature of the flue gas. It is thus the maximum temperature that can be achieved in a combustion process with given boundary conditions. It will generally be very high, compared to the actual temperatures occurring in real-life combustion applications, but can indicate how a change in operational settings such as a fuel change will affect the temperatures in the system, at least qualitatively.

Premixed and non-premixed combustion

In order for a combustion process to take place, fuel and oxidiser have to be mixed in a combustion space on a molecular level. There are two approaches to mixing fuel and oxidiser that can be found in the majority of combustion equipment, premixed and non-premixed combustion, along with an intermediate approach, partially premixed combustion (cf. **Figure 4**).

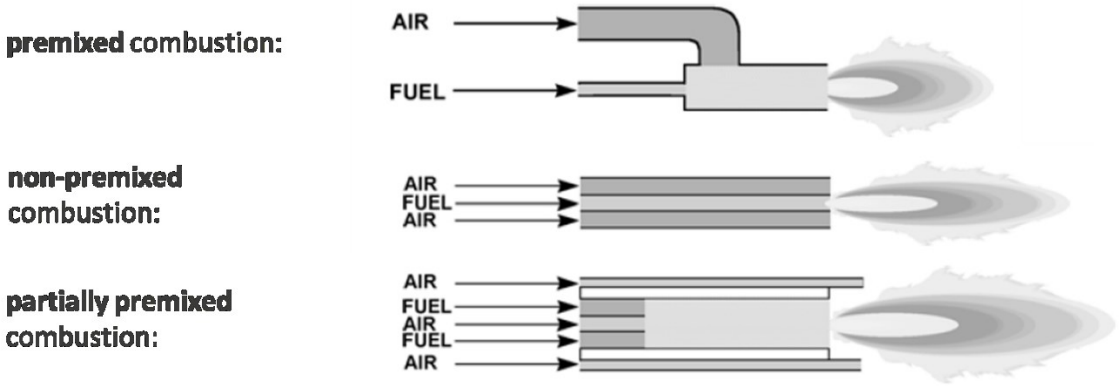


Figure 4: premixed, non-premixed and partially premixed forms of combustion [13]

In premixed combustion, fuel and oxidiser are thoroughly mixed in a mixing chamber before the fuel-oxidiser mixture is injected into the combustion space. This results in a short, compact flame with generally intense heat release. The combustion reactions occur with the global air excess ratio of the process, since mixing and chemical reaction are spatially decoupled. Flame stability and position are determined by the local balances of flow and combustion velocities (see next section for an explanation of combustion velocities). Premixed combustion processes are common in residential heating applications, but also in heavy-duty gas turbines for power generation.

Alternatively, there is the non-premixed combustion approach, sometimes also called diffusion combustion or nozzle-mixing combustion. Here, fuel and oxidiser are injected separately into the combustion space, so that both mixing and the chemical reactions take place in parallel there. This generally results in a longer flame which can be desirable if large spaces have to be heated in a more homogeneous manner, like in industrial furnaces. The separate injection of fuel and air also allows for preheating of combustion

air which is a common and very effective way to improve combustion efficiency in high-temperature applications, or the combustion with pure oxygen instead of air which is called oxy-fuel combustion [35]. Most burners in process heating applications are non-premixed systems.
As fuel and oxidiser mix in the combustion chamber itself, there is an inhomogeneous spatial distribution of local air excess ratios. The flame front will establish itself where there are locally stoichiometric conditions, while the local balance between flow and combustion velocities also has to be achieved. This makes non-premixed flames generally more stable than premixed flames, even when fuel compositions change drastically.
The intermediate approach is called partially premixed combustion: only some of the necessary combustion air is premixed with the fuel, with the remaining air injected or entrained further downstream to achieve complete fuel consumption. Domestic gas stoves are often designed as partially premixed appliances [36, 37].

(Laminar) combustion velocity

One important aspect of technical combustion processes is flame stabilisation. It is crucial to know where the flame will stabilise so that it can be monitored appropriately. This is important for the safe operation of combustion equipment because if the flame fails to stabilise where it is intended or at all, the fuel gas flow has to be cut immediately in order to prevent an unsafe state of the system.

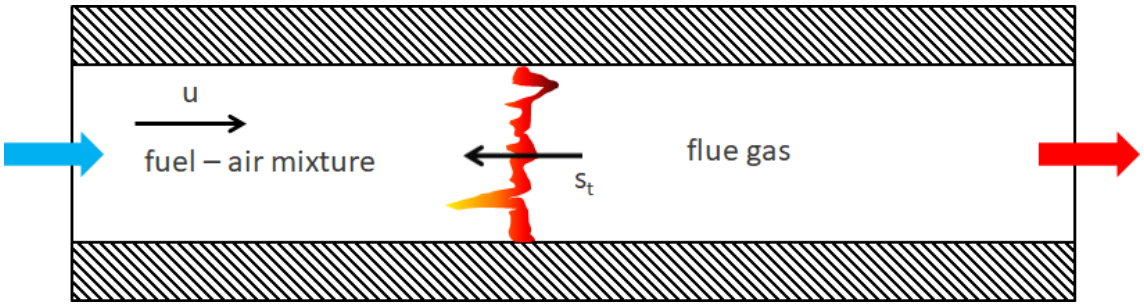


Figure 5: Flame stabilisation in an idealised premixed burner (Source: GWI)

The combustion velocity is essential in this context. In an idealised premixed one-dimensional combustion process, the flame front will stabilise where there is a local balance of the flow velocity of the fuel-oxidiser mixture heading downstream, and the combustion velocity of the flame front itself, which will propagate upstream where there is an unburned mixture (cf. **Figure 5**). If these two velocities are equal in magnitude, but opposite in direction, the flame's position remains unchanged for an external observer, and the flame is considered stable. In turbulent combustion processes, the relevant combustion velocity is the turbulent combustion velocity s_t. It is dependent on the fuel composition and the local air excess ratio in the flame front, but also on the turbulent flow field, *i.e.* this property is very much dependent on case-specific features such as the burner geometry or operational state. Although most technically relevant combustion processes are turbulent (the major exception being combustion processes in residential heating appliances [36, 38], it is therefore quite common to consider only the laminar combustion velocity s_L, in order to assess the impact of a fuel change on flame stabilisation. The laminar combustion velocity is only dependent on fuel composition, λ

and local temperature and pressure, and thus well-suited to analyse the impact of a fuel change on flame stabilisation behaviour.
Combustion velocities are an important consideration since they have a profound effect on flame stabilisation. If combustion velocities in the flame front are smaller than the local flow velocities, the flame will lift-off and stabilise itself elsewhere, usually in an undesirable location, *e.g.* a heat exchanger or flue gas duct. If, on the other hand, the combustion velocity in the flame front is higher than the local flow velocity, the flame will move upstream (a so-called flash-back), which can destroy a burner very quickly. Whether a burner system can experience a flash-back is also dependent on whether it is a premixed burner system or a non-premixed burner system (cf. the previous section). While premixed burners can flash-back, non-premixed burners cannot, as there is no combustible mixture upstream of the flame front.

Safety-related fuel aspects

Changing the fuel in any technical combustion process does not only have an impact on normal operation, but also has implications for the operational safety of combustion equipment. For any fuel, stable combustion can only occur within a certain range of mixtures between fuel and oxidiser. This range is specified by the lower and upper explosion limits (LEL and UEL) which, for gaseous fuels, are usually expressed as the volumetric concentrations of the fuel in the oxidiser for a standard temperature and pressure. If the volumetric concentration of the fuel is outside of this range, no stable combustion can occur as there is either not enough (LEL) or too much fuel (UEL) available in the mixture. The zones of stable combustion in a ternary mixture (fuel, oxygen and a diluent, usually nitrogen to account for combustion with air) are generally visualised using ternary diagrams, as depicted in **Figure 6** for methane.
Another important aspect in terms of safety is the minimum ignition energy (MIE). If the local energy state remains below this minimum ignition energy, even a per se combustible fuel-oxidiser mixture will not burn. As the minimum ignition energy of hydrogen is lower by about one order of magnitude compared to natural gas, a hydrogen-air mixture is much more easily ignited than a natural gas – air mixture, which has obvious safety implications. The auto-ignition temperature, *i.e.* the temperature at which a fuel-oxidiser mixture will ignite without an external ignition source, is relatively similar for both hydrogen and natural gas.

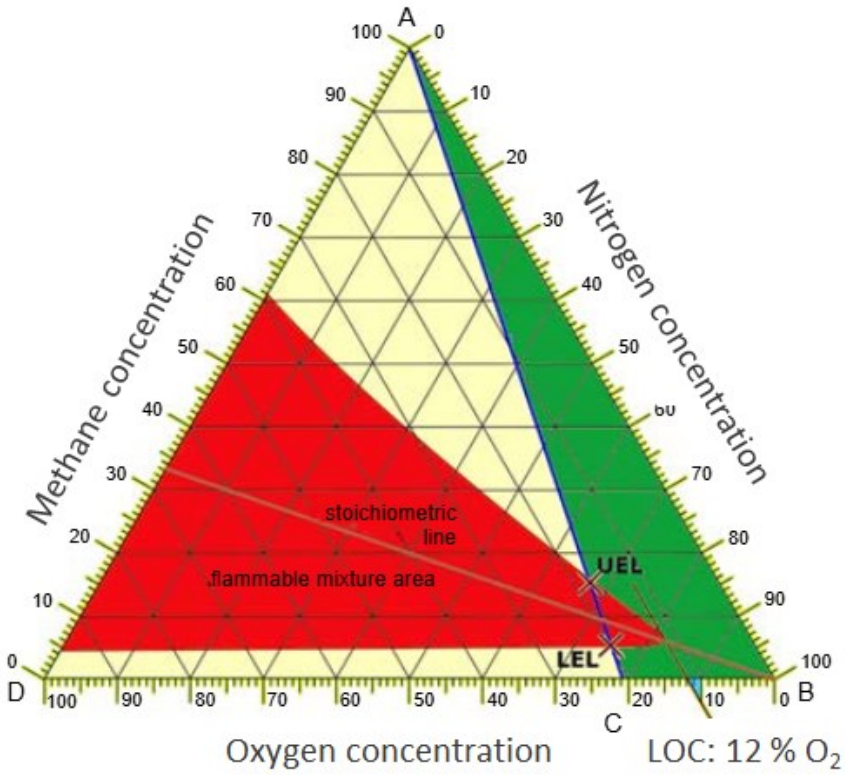


Figure 6: Ternary flammability diagram for CH_4, O_2 and N_2 (adapted from [39])

Efficiency of combustion processes

Efficiency is always an important consideration for any kind of technological application which relates the desired output to the necessary input. In the context of thermal processing applications by means of a combustion process, the output would be the process heat being used, while the input is the amount of energy being provided by the fuel. Thus, the thermal efficiency of such a process can be given as

$$\eta_{th} = \frac{\dot{Q}_{process}}{P} = 1 - \frac{\dot{Q}_{flue}}{P}$$

with $\dot{Q}_{process}$ representing the process heat (per unit of time), while $\dot{Q}_{flue}$ stands for the energy still contained in the flue gas flow, the primary source of energy losses in a combustion process. It is worth noting that that any heat fluxes going through the walls of a furnace are not seen as losses per se in this energy balance, as they are considered necessary in order to maintain the required furnace temperature [40].

The thermal efficiency of a combustion process can be influenced by a number of factors, *e.g.* the choice of air excess ratio in the process, as well as the use of technologies such as air preheating or oxy-fuel combustion. These will be explained in more detail in **Chapter 4.**

Nitrogen oxides (NO_X)

Formation of nitrogen oxides in the combustion of gaseous fuels

In addition to operational safety, fitness-for-purpose and high thermal efficiency, compliance with legal emission limits for pollutants is another constraint for the design and operation of technical combustion equipment. For industrial equipment, the focus is often on the emission of nitrogen oxides (NO_X), while other end-use applications, *e.g.* residential heating appliances, tend to focus on the emission of other pollutant species such as carbon monoxide (CO). Carbon monoxide is mostly a safety concern (CO is toxic), whereas NO_X emissions are primarily limited due to air quality concerns.
The term NO_X is a shorthand that includes both nitric oxide (NO) and nitrogen dioxide (NO_2). The majority of the NO_X emissions from technical combustion equipment is actually NO, which is then oxidised further in the atmosphere to NO_2.
In the combustion of gaseous fuels such as natural gas or hydrogen, the thermal formation pathway of NO_X usually is the most relevant one. This formation mechanism is well understood and follows the elemental equations:

$$N_2 + O \leftrightarrow NO + N$$
$$N + O_2 \leftrightarrow NO + O$$
$$N + OH \leftrightarrow NO + H$$

The first equation controls the kinetics of NO formation, as atomic N is a prerequisite for the second and third equation. Thermal NO_X formation is therefore primarily dependent on high local temperatures (> 1200°C) to disassociate the very stable N_2 molecules, the presence of oxygen and the residence time in the hot zone. Pressure also plays a role but is usually something that is outside the control of the combustion engineer as combustion processes generally take place either at ambient pressure (residential appliances or most industrial furnaces) or under pressurised conditions, *e.g.* in gas turbines or gas engines.
It is worth pointing out that combustion process actually only supplies the energy and temperature that are needed to initiate NO_X formation, as both components of NO, oxygen and nitrogen, are already present in air. This also means that in the context of oxy-fuel combustion, it is actually not the presence of oxygen which limits thermal NO_X formation, but the presence of nitrogen [35, 41]. Since the main source of nitrogen, air, is no longer being used, any nitrogen that is oxidised has to originate elsewhere, either as a species in the fuel itself, as a trace component in the oxygen or due to infiltration air [35, 42, 43].

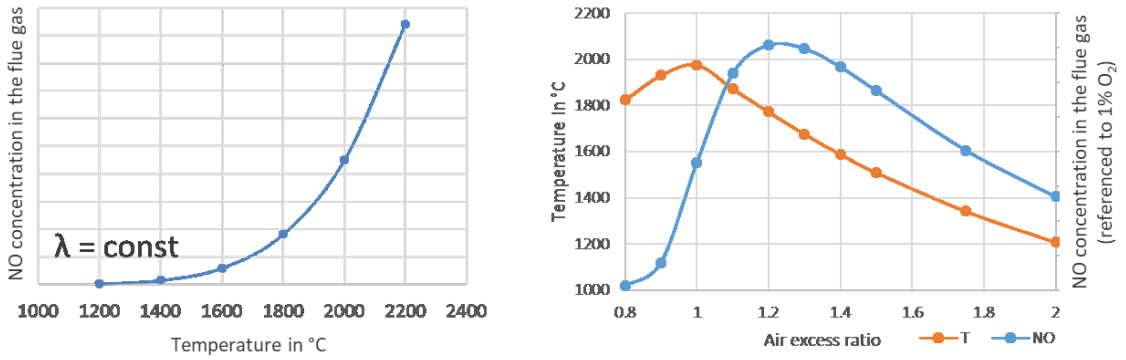


Figure 7: Effects of temperature (at λ = const) and air excess ratio on thermal NO formation (Source: GWI).

Figure 7 shows the principal effects of local temperature (represented in this theoretical consideration by the adiabatic combustion temperature) and air excess ratio on NO concentrations in the flue gas. The diagrams highlight two key features of the thermal NO_X formation mechanism: it is exponentially dependent on local temperatures, and the availability of oxygen (due to excess air) is a limiting factor. As the diagram on the right-hand side indicates the maximum NO concentrations does not coincide with the maximum temperatures at stoichiometry, but can be found in the leaner regime, at $\lambda \approx 1.2$. While the maximum temperature occurs at $\lambda \approx 1$, there is hardly any oxygen available to form NO, as it has already been consumed by the main oxidation reactions since, chemically speaking, thermal NO formation is relatively slow [44]. At around $\lambda \approx 1.2$, where the maximum of NO concentrations in the flue gas can be found, temperatures are still high enough to initiate NO formation, while there is also sufficient excess oxygen available to actually produce NO.

NO_X mitigation

As NO_X emissions from combustion processes are strictly regulated around the world, NO_X mitigation has been a driving force of combustion research across all sectors and applications for the last decades, and been very successful in reducing anthropogenic NO_X emissions overall. Even in challenging high-temperature applications such as glass melting, low NO_X emissions can be achieved [45], despite the high temperatures that are required. Given the wide variety of technical combustion processes, some of the solutions are highly specific, while others can be found in many different end-use applications across different sectors.

There are, in principle, two ways how NO_X emissions can be reduced in a combustion process: primary measures aim at preventing NO_X formation in the first place by changing the combustion process itself in some way.

Alternatively, secondary measures can be used to remove NO_X from the flue gas, usually in the form of selective catalytic or selective non-catalytic reduction techniques (SCR and SNCR, respectively). This involves the injection and mixing of a reducing agent (usually ammonia or urea) with the flue gas so that existing NO_X can be chemically reduced to N_2 and H_2O.

As visualised by **Figure 7**, most primary measures inhibit thermal NO formation by avoiding a state where high O_2 concentrations locally and temporarily coincide with high

temperatures over long periods of time (relatively speaking). For example, modern heavy-duty gas turbines operate lean premixed burners at high air excess ratios ($\lambda \approx 2$) to reduce temperature peaks in the flame and also reduce flame lengths, allowing for smaller combustion chambers and thus lower residence times in hot regions. On the other hand, industrial process heating equipment for high-temperature applications (*e.g.* for melting or heat treatment of materials), is usually equipped with non-premixed burners running at air excess ratios of 1.1 and below [26] in order to minimise the availability of oxygen to form NO, among other reasons. Often, long flames (another consequence of near-stoichiometric non-premixed combustion) are desirable anyway in these applications since the furnace spaces are large and a homogeneous temperature distribution is usually desirable for product quality reasons. A lean premixed combustion, as in gas turbines, would not be suitable here as the high temperatures are actually required to fulfill the purpose of the furnace, and high air excess ratios would lead to loss of efficiency and, in fact, strong NO_X formation as large quantities of oxygen are introduced into a very hot environment.

Wet and dry flue gas

Pollutant emissions, for example nitrogen oxides, are an important consideration for the design and operation any kind of combustion equipment. The question of pollutant emissions is, however, not only a question of physics and chemistry, but also a question of how pollutant emissions are actually measured. It is industrial practice in Europe that the concentration of pollutants in a flue gas is quantified for the “dry” flue gas. This means that when a flue gas sample is taken by a probe, this sample is rapidly cooled down so that the water vapour in the flue gas condenses to liquid water which is then removed. The reason for this procedure is mostly one of practicality: most industrially available sensor technologies are vulnerable to humidity, and there is also the risk of cross-sensitivity between water vapour and other, more relevant species. The most cost-effective way to monitor pollutant emissions is therefore to measure the pollutant concentration in the dry flue gas. Therefore, legal emission limits are also usually expressed in the same manner, using units such as [ppm] or [mg/m^3] as it is easier to measure a concentration than the actually emitted mass flow of a pollutant.
This common approach has serious implications when discussing a fuel switch from natural gas to hydrogen, which will be discussed in more detail in **Chapter 6**.

4. Combustion in industrial process heating: technologies and applications

Combustion is a key technology in many manufacturing industries, from steam generation and drying processes, *e.g.* in the food industry, to the production and treatment of essential materials such as steel, aluminium, glass or ceramics. Energy and temperature requirements can differ greatly, and often the success of a manufacturing process is not determined by heat transfer alone, but also for example by the interaction of the product with the furnace atmosphere. **Figure 8** gives an overview of typical industrial combustion processes alongside their respective temperature requirements.

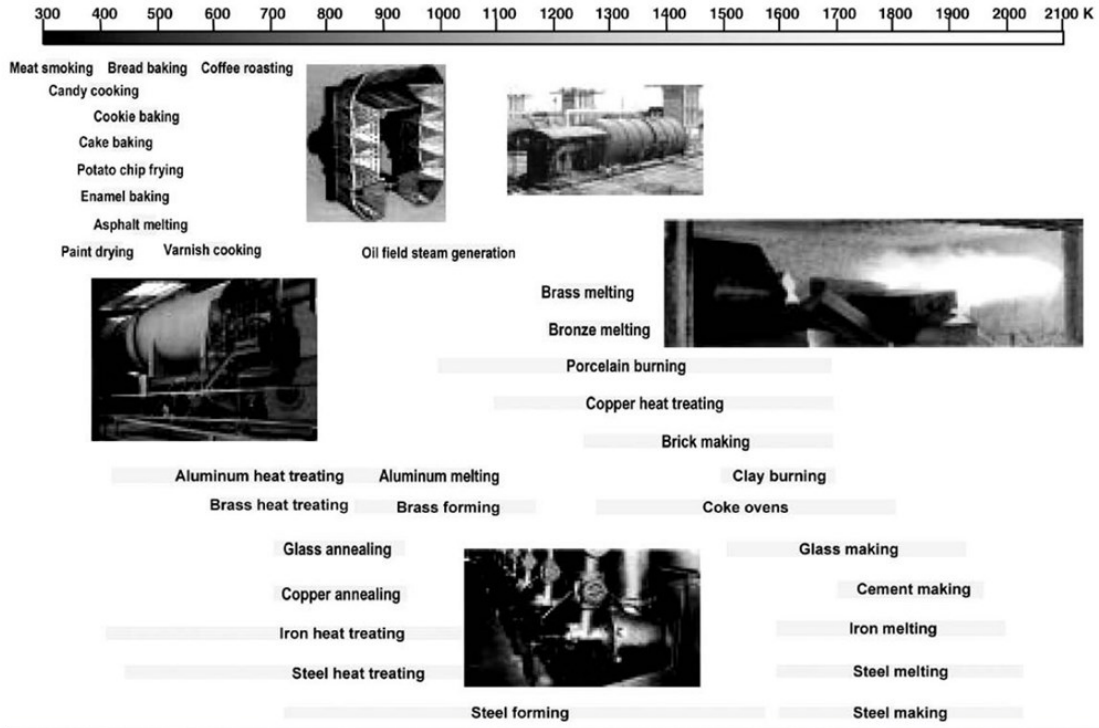


Figure 8: Typical industrial combustion processes and their required temperature levels [13]

Given the wide variety of processes and products, the respective combustion processes are very heterogeneous as well: in some applications, *e.g.* cement-making, the use of solid fuels such as coal or biomass is dominant, while others, such as the non-ferrous metals, glass or ceramics industries use gaseous fuels, usually natural gas. But even if the same fuel is used, the differences in the actual process demand a high degree of specialisation. Burners and combustion equipment for glass melting look very different from burners used for aluminium scrap recycling, for example. **Figure 8** gives an overview of combustion processes in typical manufacturing processes with their respective temperatures [13].

However, despite these many differences, there are also a lot of similarities in industrial combustion processes. Process heating always is a means to an end, so the focus of these processes is on achieving the desired product qualities.
Efficiency is also important as the amount of energy transformed in a furnace is usually substantial. Many furnaces operate in the double or even triple MW range continuously, so even small efficiency gains translate into cost savings (and CO_2 emissions reductions).

This is particularly relevant in countries where local energy costs are relatively high, like in the EU.
In addition to product quality and efficiency, environmental concerns usually are important for the operation of industrial combustion equipment as well. Traditionally this referred to compliance with environmental regulations for the emission of air quality-relevant pollutants such as nitrogen oxides. Emission limits became stricter over time, and industries had to adapt their processes to meet these requirements. In the context of decarbonization, however, this also implies the need to reduce and eventually completely eliminate greenhouse gas emissions such as carbon dioxide. While emissions of pollutants such as NO_X can usually be addressed by modifications to existing technologies (some primary measures for NO_X mitigation will be discussed later), the elimination of greenhouse gas emissions requires a more fundamental change.

On the more technical level, industrial combustion processes are usually non-premixed, while combustion applications in the residential sector or in power plant gas turbines usually are premixed.

In the following section, some peculiarities of industrial combustion applications will be explained as this is relevant for the subsequent discussion about how such processes will be affected by a switch from natural gas to hydrogen.

Near-stoichiometric combustion

One of the most distinguishing features of combustion processes for industrial process heating is that they are often operated very close to stoichiometry. A recent survey of industrial equipment operators in Germany found that the majority adjust their processes to run with air excess ratios of 1.1 or below, *i.e.* about 10% excess air [26]. This is very different compared to combustion applications in other sectors: in the residential sectors for example, appliances are usually set to air excess ratios between 1.2 and 1.4 [24, 25], while gas turbines for power generation tend to operate with high air excess ratios of about 2, in order to prevent overheating of turbine blades and to minimize NO_X formation in the combustor.

There is a number of reasons why industrial furnaces are set to run close to stoichiometry. First of all, industrial furnaces, especially in the energy-intensive basic materials industries, are often operated at very high temperatures. This is a prerequisite of the actual processes: for example, the melting temperature of aluminium alloys is about 700-800°C, while the heat treatment of steels can require temperatures of 1200°C and more. Glass melting requires temperatures of up to 1600°C.
These temperatures are significantly higher than what can be found in residential heating appliances or even large steam generators. At high furnace temperatures, a high air excess ratio translates into decreased thermal efficiency since the excess air has to be heated as well, although it does not contribute to the process. While the same effect, in principle, also applies to combustion in low-temperature applications, the efficiency losses at higher temperatures are much more significant. This is visualised in **Figure 9**, based on theoretical calculations of the thermal efficiencies of a combustion process for different air excess ratios and flue gas temperatures. The impact of high air excess ratios for low-temperature applications such as a residential condensing boiler with a typical

flue gas temperature of about 60°C is fairly negligible, but the effects of λ on the efficiency for a process with a flue gas temperature of 800°C are far more pronounced.

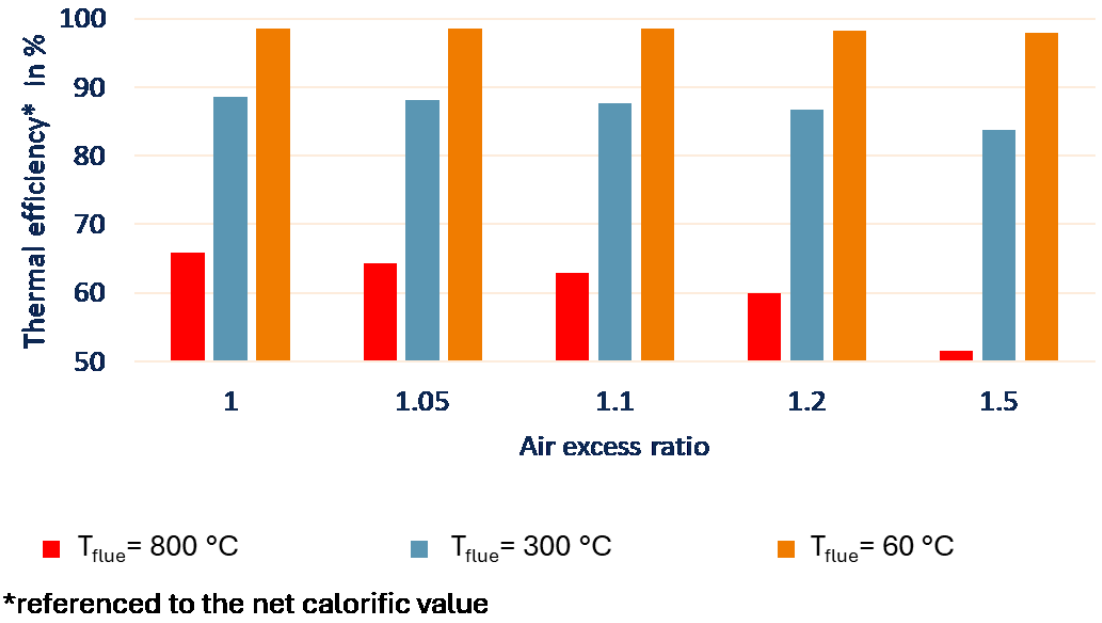


Figure 9: Thermal efficiencies of combustion processes (with CH_4) as functions of flue gas temperatures T_{flue} and air excess ratio λs. Source: GWI

It should be noted that in these calculations, waste heat recovery for air preheating, a common and effective method to improve the thermal efficiencies of high-temperature combustion processes, was not considered. This aspect will be discussed in the next section.

A low air excess ratio in combination with a non-premixed combustion process also creates long flames. While long flames are often not desirable in applications with limited available space, *e.g.* residential heating appliances or modern gas turbines, they can be beneficial in industrial furnaces which tend to be large, and often rather empty volumes. A long flame creates a more homogeneous heat and temperature distribution in a large furnace space which is usually advantageous in terms of product quality.

This is also relevant in the context of NO_X emissions and their mitigation. Premixed and non-premixed combustion processes behave fundamentally differently when it comes to the impact of the air excess ratio on NO_X formation as is highlighted by **Figure 10.** An increase in the air excess ratio in a premixed combustion process will usually result in reduced NO_X formation, as the higher λ will reduce the maximum flame temperatures. In a non-premixed combustion process, however, the local λ in the flame front will always be around unity, as these combustion processes are primarily stabilised by mixing. Here, the higher air excess ratio only serves to supply additional oxygen to a high-temperature environment, thus promoting NO_X formation even further.

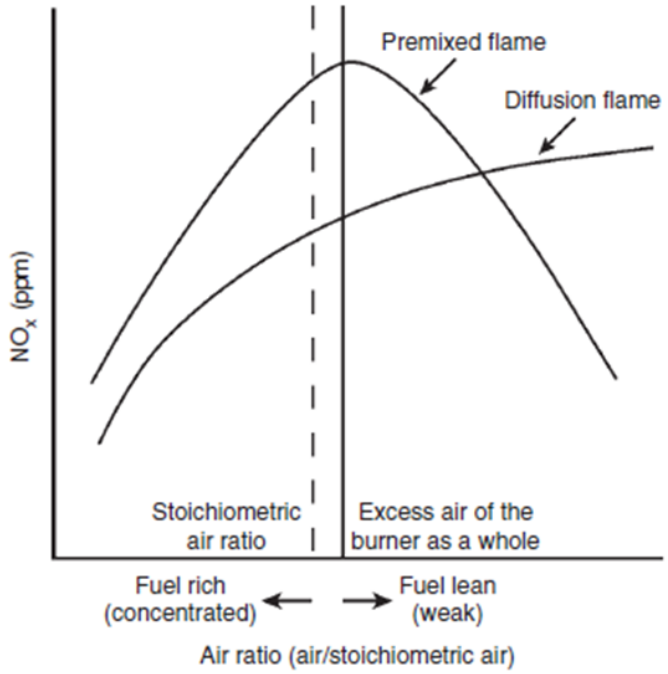


Figure 10: NO_X concentrations in premixed and non-premixed flames (labeled diffusion flames in the graphic)) as a function of the air excess ratio [46].

Waste heat recovery and combustion air preheating

In an industrial combustion process, the flue gas is usually responsible for the biggest efficiency losses to the process, especially when high process temperatures are required. One very effective and common approach to recover some of the thermal energy still contained in the flue gas is to use it to preheat the combustion air. In this manner, some of the chemical energy conveyed by the fuel into the process can be substituted by recovered thermal energy from the flue gas which is transported into the furnace by the preheated air. **Figure 11** shows the principle of this approach.

Air preheating in industrial combustion processes can come in many different forms, depending on the actual process and also other constraints, for example the availability of space for the necessary heat exchangers.
Two heat exchange principles are commonly used, recuperative heat exchangers and regenerative heat exchangers. In a recuperative heat exchanger, the flue gas and the combustion air flow continuously, separated by a heat exchanger surface. Due to the temperature difference, heat is transferred to the combustion air in a flux through a separating surface.
Regenerative heat exchangers operate cyclically: the flue gas transfers the heat to a heat storage medium (usually some kind of ceramic material) and after a while, the ceramic is then exposed to the cold combustion air, which is then heated up. Typically, regenerative heat exchangers are operated in pairs: one is charged by the hot flue gas, while the other is used to preheat the combustion air at any given time. In this manner, a continuous supply of preheated air is guaranteed. Cycle times can vary between a few seconds for small regenerators and up to 20-30 minutes, for example in the glass or steel industry.

Generally, recuperative heat exchangers are made from steel and can thus be used with preheat temperatures up to about 800°C, though recuperators made from ceramics can withstand higher temperatures. Regenerative preheating can achieve air preheat temperatures up to 1400°C, for example in the glass industry.

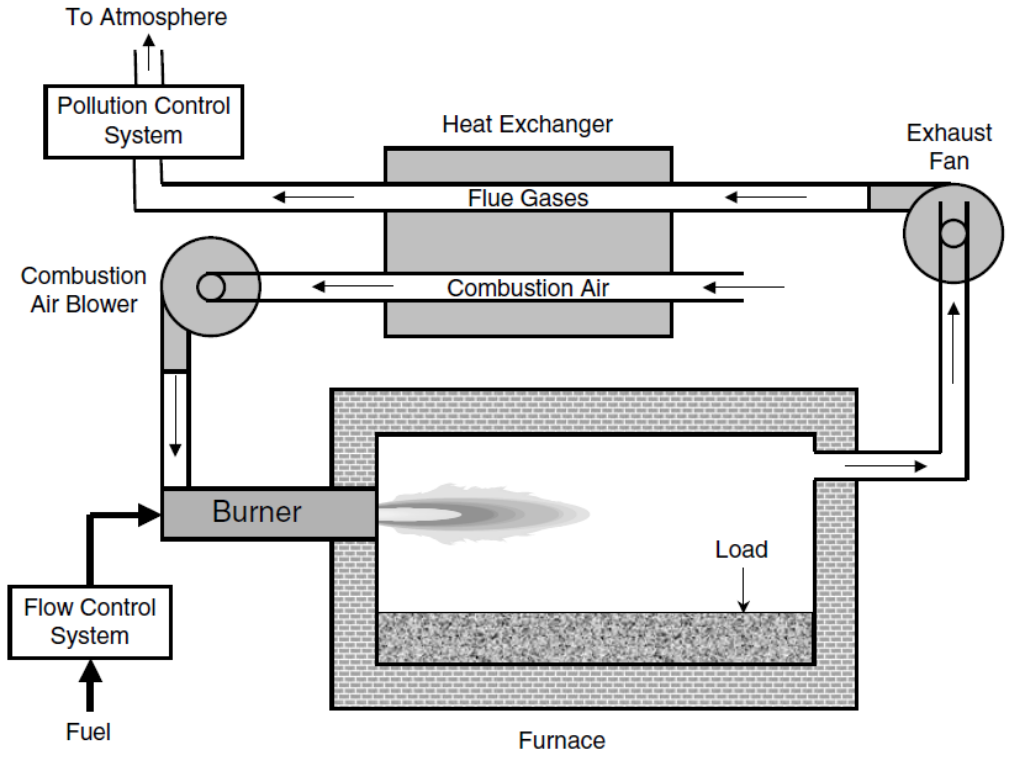


Figure 11: Principle of waste heat recovery by air preheating [47]

In some instances, central recuperators or regenerators are used, though both recuperative and regenerative heat exchangers can also be directly integrated into a burner, making for compact units. Central recuperative heat exchangers are less efficient than either self-recuperative or self-regenerative burners (*i.e.* burners where the heat exchangers are incorporated into the burner itself) while central regenerators are even more efficient than self-regenerative burners bur also require a lot of space.

The improvement of thermal efficiency (and hence energy savings) by air preheating can be significant, provided the available flue gas has a sufficiently high temperature, as is visualised by **Figure 12**. The theoretical thermal efficiency of a combustion process increases linearly with the air preheat temperature. There are two main reasons for this increase: the amount of energy lost by means of the flue gas is reduced and the recovered heat is used to replace some of the chemical energy provided by the fuel. Thus, fuel consumption is decreased. Also, air preheating leads to increased combustion temperatures (cf. **Figure 13**). Since radiative heat transfer is the dominant heat transfer mechanism in a high-temperature environment and highly dependent on local temperatures, these increases in flame temperatures intensify heat transfer, thus improving overall efficiency.

The potential of air preheating to improve process efficiency is limited by a number of factors. The first and obvious one is that sufficient quantities of hot flue gas are required. The air preheat temperature cannot rise above the temperature of the flue gas that

serves as a heat source. Thus, boilers or steam generators usually do not use air preheating to any significant extent as the flue gas temperature is too cold.

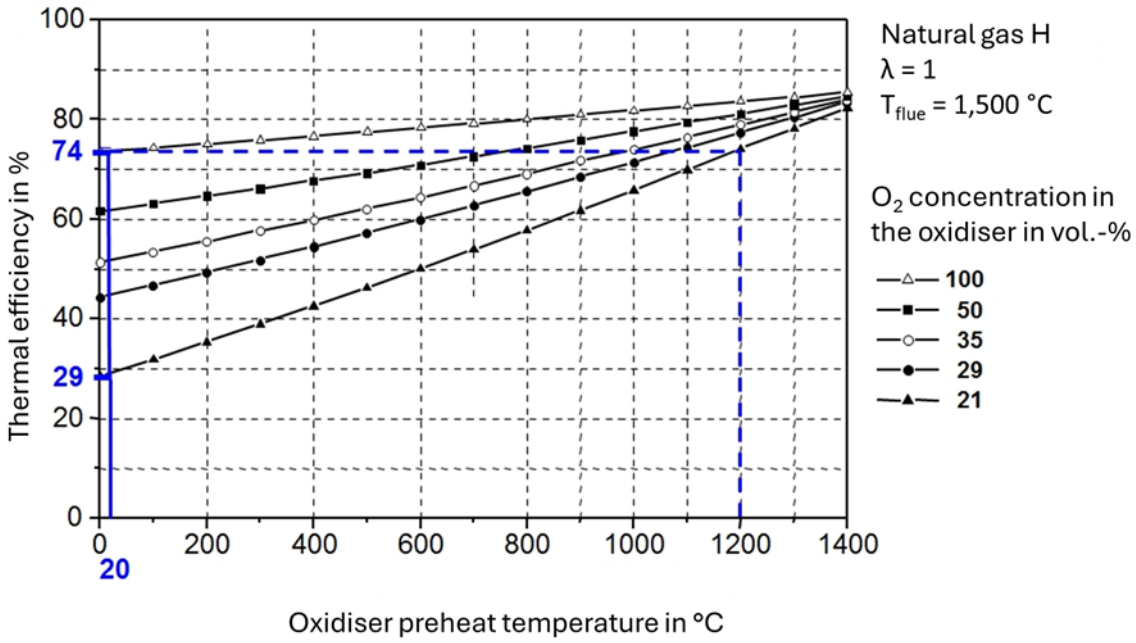


Figure 12: Thermal efficiency as a function of oxidiser preheat temperature and composition (translated from [28]).

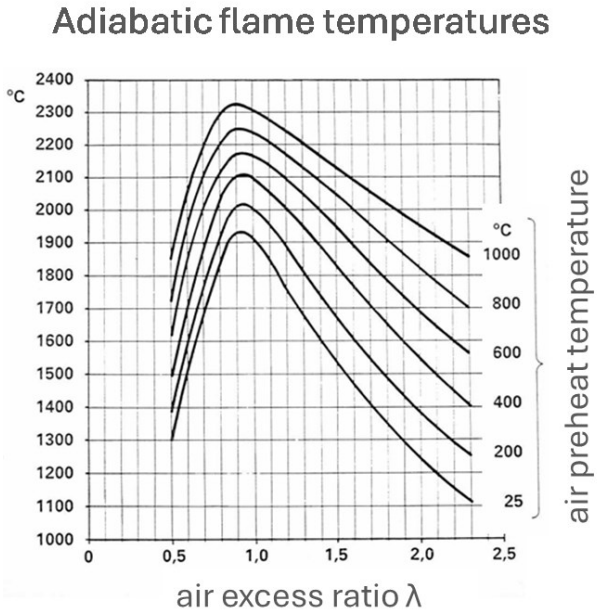


Figure 13: Adiabatic combustion temperatures of methane as a function of the air excess ratio and the air preheat temperature (translated from [28]).

Another aspect to consider are NO_X emissions. As previously described, NO_X formation in the combustion of gaseous fuels is strongly dependent on local temperatures. Thus, strong air preheating can potentially cause a significant increase in NO_X emissions which has to be considered when using air preheating. However, several primary measures are well-known and understood which can be used to achieve high-temperature air preheating (and thus efficiency) with low NO_X emissions, which will be discussed in the

following sections.
Finally, there is the question whether the flue gas contains aggressive trace components which could damage the heat exchangers during operation. Recuperative heat exchangers are particularly sensitive to this although this is very much dependent on the actual process being heated. It is, for example, a challenge for burners in aluminium recycling furnaces where the recycled scrap is often contaminated with various residues [48]. Burners in radiant tubes, on the other hand, are not affected by this at all since the combustion space and furnace space are physically separated.

Oxy-fuel combustion

An alternative approach to provide high-temperature process heat in a very efficient manner is the so-called oxy-fuel combustion. Here, (preheated) air is no longer used as oxidiser. Instead, almost pure oxygen serves as oxidiser [35]. This offers a number of advantages: oxy-fuel combustion can achieve much higher combustion temperatures than even combustion with strongly preheated air (the adiabatic combustion temperature of a stoichiometric natural gas – oxygen flame is about 2800°C), which, in combination with a flue gas that consists mostly of tri-atomic species CO_2 and H_2O (when burning natural gas or other hydrocarbons), results in excellent radiative heat transfer. This is shown by **Figures 14** and **15** which compare heat transfer from oxy-fuel combustion with other industrial combustion processes.

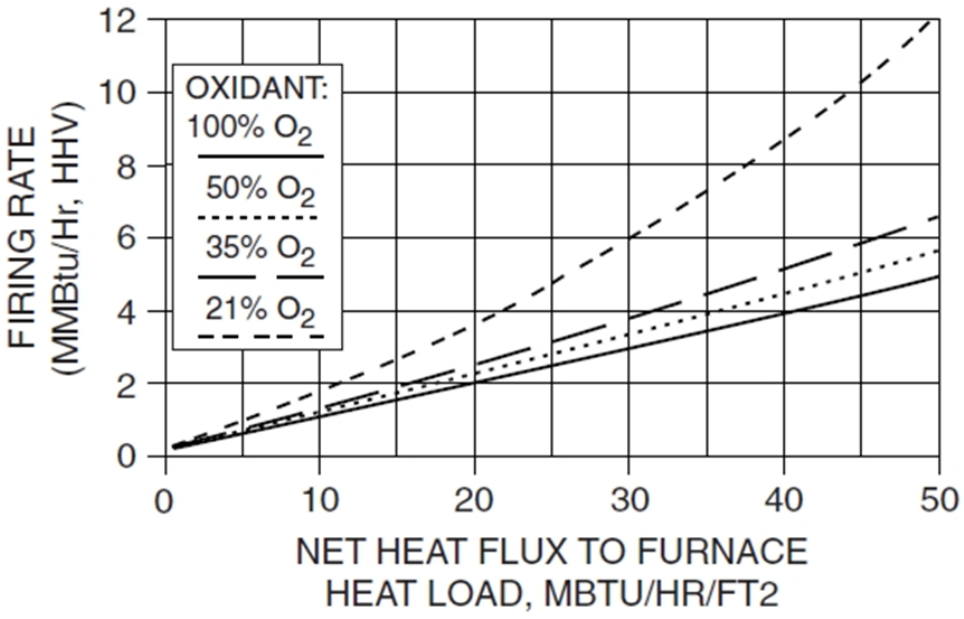


Figure 14: Firing rate and heat flux into the product for different oxidisers [47]

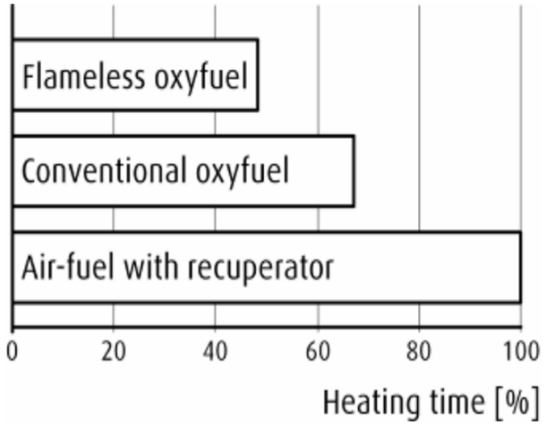


Figure 15: Heating times with different combustion technologies [49]

The lack of nitrogen is another aspect of oxy-fuel combustion which contributes to its overall efficiency. In a combustion process with air, nitrogen does not participate in the chemical reaction and only acts as a heat sink since it has to be heated along the reaction products. This explains the high flame temperatures and the high thermal efficiencies of oxy-fuel combustion, as shown for example in **Figures 12** and **14**. Additionally, this lack of nitrogen (air consists of about 79 vol.% N_2) can reduce NO_X formation significantly, especially in the near-stoichiometric combustion processes common for industrial process heating. Despite the overall high local temperatures, the slow NO_X formation cannot occur as the available oxygen has already mostly been consumed by the main combustion reactions. In conventional combustion processes, on the other hand, the nitrogen contained in the air usually is the primary nitrogen source. However, NO_X emissions can also be significant in oxy-fuel combustion if nitrogen is provided by other sources such as infiltration air or nitrogen content in the fuel or the oxidiser [42, 47, 50]. Many primary measures developed to reduce NO_X emissions from air-based combustion processes can be adapted to work with oxy-fuel combustion as well, *e.g.* staging [51] or flameless/MILD combustion [52, 53] (see next section).
From an economic perspective, the main drawback of oxy-fuel combustion is that oxygen has to be produced, while air is a free resource. Nevertheless, the advantages in terms of efficiency, increased productivity and pollutant reduction can outweigh the increase in operation or investment cost. Today, many industrial furnaces around the world use oxy-fuel combustion for process heating, primarily in high-temperature applications like melting of glass or metals, or the heat treatment of metals [35, 52, 54].

Flameless Oxidation/MILD Combustion

In addition to fitness for purpose and high efficiency, compliance with legally required NO_X emissions standards is another principal concern for operators of industrial furnaces as well as for manufacturers of the respective equipment. Two primary measures for NO_X reduction merit special consideration in the context of the H2AL project. The first one is the already mentioned oxy-fuel combustion. By removing air as the main source of nitrogen in the combustion process, oxy-fuel combustion, in theory, allows for extremely low NO_X emissions. However, given the high temperatures that can occur in oxy-fuel combustion, care must be taken to avoid any other nitrogen sources, *e.g.* as trace components in the fuel, as impurities in the oxygen or due to infiltration air [41, 42].

However, by combining oxy-fuel combustion with other conventional primary measures such as staging or flameless oxidation/MILD combustion, this sensitivity can be greatly reduced [47].

Flameless oxidation [55], often abbreviated FLOX© or alternatively called MILD combustion (MILD: Moderate or Intense Low-Oxygen Dilution) [56], is another very effective method to prevent NO_X formation in the first place. By mixing the reactants with large quantities of hot, but chemically inert flue gas, the temperature peaks in the reaction zone are drastically reduced, as is shown in **Figure 16,** while the local oxygen concentration is lowered as well.

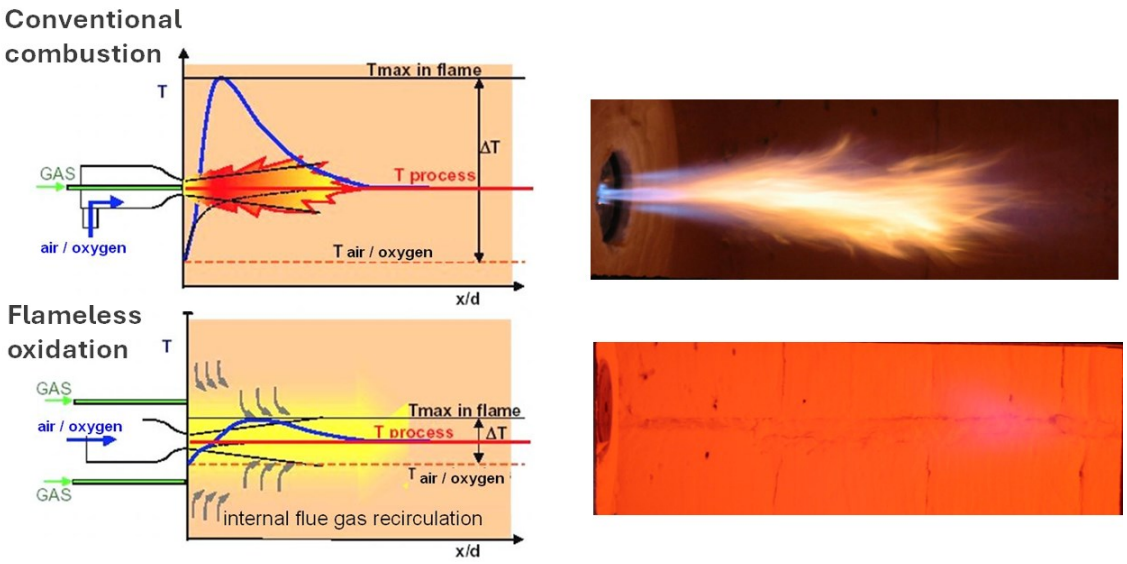


Figure 16: Comparison of the working principles of a conventional non-premixed combustion process and flameless oxidation [57]

Both effects inhibit the formation of thermal NO_X, so that very low emissions can be achieved even in high-temperature environments. In principle, flameless oxidation can be considered as an extreme form of internal or external flue gas recirculation. The consequences of the extreme dilution of the reactants are significant as the combustion reactions no longer take place in a classic thin flame front, but instead are distributed over a large reaction volume. One consequence is that the "flame" is no longer visible with the naked eye which gave it the name of "flameless oxidation". The reaction zone is still clearly visible in the UV spectrum though.

The intense mixing of flue gas into the reaction zone is typically achieved by injecting fuel and oxidiser at very high speeds (> 100 m/s), so that large quantities of chemically inert flue gas are entrained, disrupting the formation of a conventional flame sheet. At the same time, this hot flue gas serves to elevate local temperatures in the reaction zone above the self-ignition temperature of the fuel-oxidiser-flue gas mixture (cf. **Figure 17)**, so the chemical chain reactions are self-sustaining. In most cases, flameless oxidation is found in applications with strong air preheating and/or high flue gas temperatures.

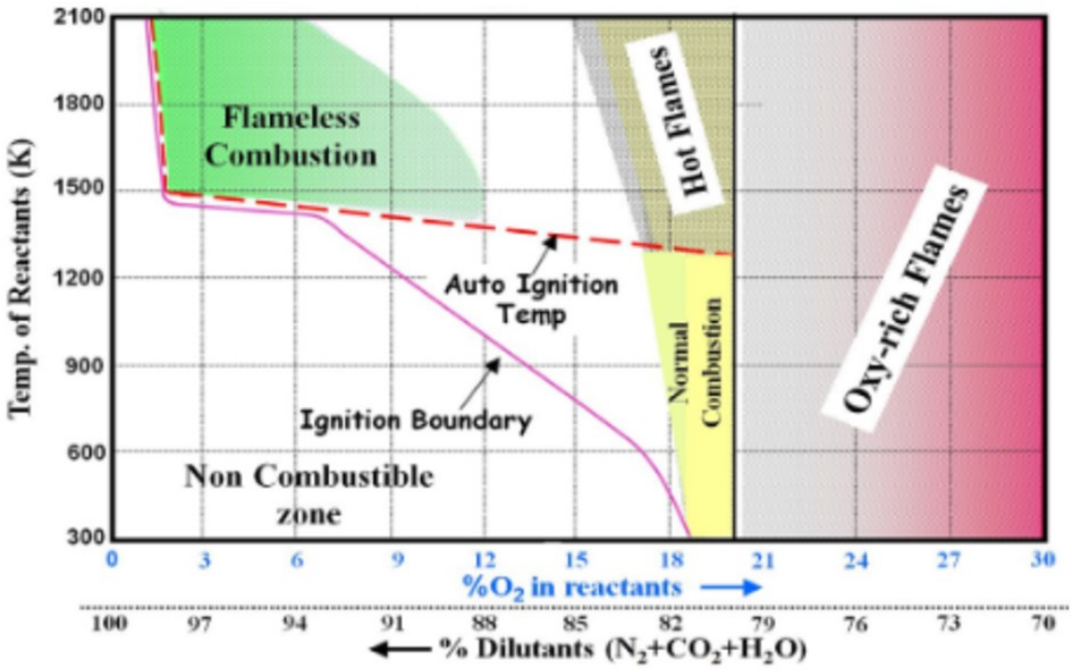


Figure 17: Schematic of various combustion regimes. Flameless combustion is characterised by a low concentration of oxygen in the reaction zone at temperatures higher than the auto-ignition temperature [58]

Flameless oxidation/MILD combustion has been established as one way to achieve extremely low NO_X emissions even in high-temperature environments such as industrial furnaces, and can be found in applications in the metals [40], and even the glass industry [59]. While this technology was originally developed for the combustion with air, the same principles also apply for flameless oxidation with oxy-fuel [52, 60], as the NO_X formation mechanisms remain the same both in the combustion of natural gas (or hydrogen) with air or with oxygen.

Another advantage of flameless oxidation is that it generates very homogeneous temperature distributions in the combustion space which is beneficial as it avoids local overheating in components or the materials being heated.

Industrial furnaces

As stated previously, industrial process heat is an important aspect of global energy utilisation, across many different industries, and as such, also an important source of industrial greenhouse gas emissions, as process heat mostly produced from fossil fuels today, in particular natural gas. Much of this gas-generated process heat is used in industrial furnaces, although other applications also exist, *e.g.* steam generators or boilers that can be found in the chemical, food or paper industries in large quantities.
According to VDMA (the German Association of Machine and Equipment Manufacturers), an industrial furnace or more broadly, a thermal processing plant is defined as an enclosed space in which a material is subjected to heat to carry out certain processes within the material or on its surface [61].
The key feature which distinguishes an industrial furnace from a boiler or steam generator is that the operating temperatures in furnaces tend to be much higher, *e.g.* in order to melt metals or glass, or to carry out a heat treatment: temperatures inside the

furnace can go up to 1800°C (cf. **Figure 8)**. Therefore, the inside of the furnace walls is lined with insulating heat resistant ceramics, the so-called refractory, while the inside walls of a boiler or steam generator are usually made from steel or other metallic materials.
The refractory serves two purposes: it protects the metallic structural components of the furnace from the high temperatures inside, while also reducing heat loss through the walls, thus improving process efficiency.

Given the wide variety of products and processes, industrial furnaces come in many different forms and sizes. Some of them are operated with solid fuels like coal or biomass, many use natural gas. **Figure 18** shows a furnace design typical for aluminium recycling, especially for aluminium casting. This setup, often called a tower furnace [48] or melting tower [62], is in principle a combination of two very common furnace types, a shaft furnace and a reverbatory furnace. This is the furnace type operated by project partner 2A which in the course of the H2AL project is to be converted from a conventional combustion of natural gas with air to a hydrogen-oxy-fuel process.

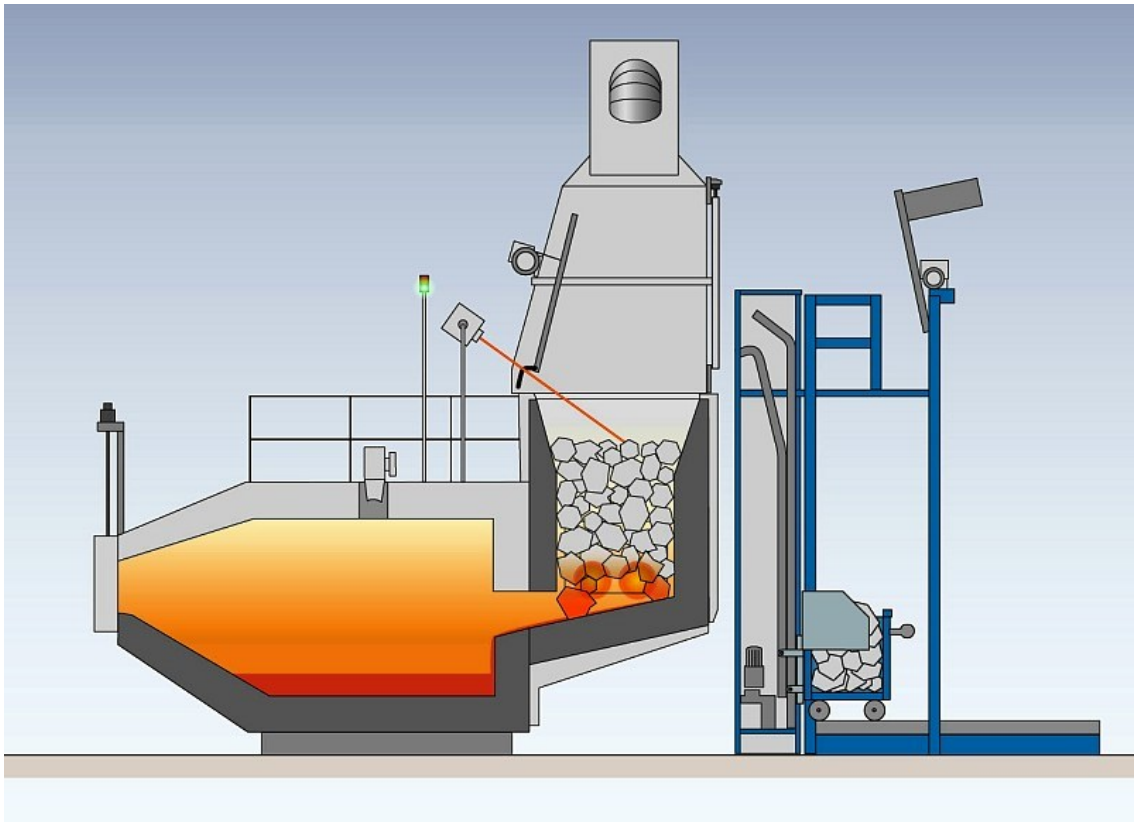


Figure 18: Schematic of an aluminium melting tower [62]

This furnace consists of a vertical shaft into which the aluminium scrap is introduced, and a holding furnace where the molten aluminium is kept until it is discharged from the furnace. Burners are mounted at the base of the shaft and in the holding furnace to provide the necessary heat. The burners at the base of the shaft are bigger than those in the holding section.

The shaft acts as a counterflow heat exchanger: the flue gas from the burners moves upwards towards the chimney, while the charge is first heated and then liquefied as it

slowly passes through the shaft section into the melting and holding furnace where it can be tapped.

Figure 19: Images of a melting tower for aluminium recycling (Source: MARCONI).

This working principle makes melting towers energetically more efficient than other furnaces such as reverbatory hearth or bath furnaces [48, 63]. Waste of metallic aluminium is also lower than with other furnace types. The finished aluminum melt is characterised by a reduced content of hydrogen and non-metallic inclusions. This is achieved by separating the melting zone and the preheating zone. Moisture and other contaminants evaporate or burn off while still in the shaft, and a dry charge with a minimum of polluting materials enters the melting zone. However, these furnaces require a lot of space, and refractory materials are subjected to significant mechanical stresses [63].

The burners used in these furnaces are usually non-premixed natural gas burners with no or only very little air preheating. There are two main reasons: on the one hand, aluminium scrap is often contaminated with aggressive trace components which would quickly damage heat exchangers, On the other, hand, waste heat recovery is already an integral part of the furnace setup since the vertical shaft serves as a counterflow heat exchanger by itself.

5. Characteristic fuel properties of natural gas and hydrogen

Any technical combustion process has to take the specific properties of the intended fuel into account and be designed and adjusted accordingly. Changes in fuel composition will affect the performance of combustion equipment, with potential consequences ranging from changes in pollutant emissions or combustion efficiency even to safety-relevant aspects such as the formation of toxic species in the flue gas (*e.g.* carbon monoxide) or a flash-back event. In industrial manufacturing processes, product quality is an additional yet crucially important aspect to consider.

In this chapter, the main differences in terms of fuel properties of both natural gas and hydrogen will be discussed in general terms. In most comparisons, natural gas will be represented here by methane, although natural gas is actually a mixture of methane, higher hydrocarbons such as ethane or propane, and inert components like carbon dioxide or nitrogen. The actual composition of natural gas can vary significantly depending on the origin of the gas or any pre-processing [64], and also fluctuate over time (cf. for example [32, 65, 66]). However, compared to hydrogen, these variations in fuel properties due to different natural gas compositions can often be neglected. Depending on the sensitivity of the application, even changes within a well-defined family of chemically similar fuels such as natural gases can have a negative impact on the performance of combustion equipment [64, 66, 67].
Hydrogen is usually not a species found in natural gas, although there are discussions about blending hydrogen into natural gas to reduce the carbon footprint and create a demand for hydrogen from sustainable sources [68]. Considering that natural gas and hydrogen are, in fact, chemically very different, it is to be expected that the effects of such a fuel change on existing combustion equipment can be severe, so that modifications of the equipment will be needed so that the device or process can still fulfill its intended purpose, while also being operated in a safe, efficient and environmentally friendly manner.
Table 2 gives a comparison of some of the key fuel properties of methane and hydrogen. These figures already give a first inclination of what can happen in a combustion process when switching from natural gas to hydrogen, and how combustion equipment like burners or control systems have to be adapted to maintain optimum performance.

The first properties to note are the volumetric net and gross calorific values of methane and hydrogen. The net calorific value of hydrogen is about 1/3 of the net calorific value of methane in volumetric terms, which means that a roughly three times higher volume flow of fuel is needed to produce the same amount of heat in a combustion process in any given time period. At the same time, the minimum oxygen or air requirements (O_{2min} and Air_{min}, respectively) to achieve complete combustion of a hydrogen molecule is 75% lower, so the overall demand of oxidiser to release the same amount of heat is reduced by about 20%. Consequently, the velocity and turbulence fields in the combustion space can change significantly if the burner is not adapted accordingly, with repercussions for aspects such as mixing of fuel, oxidiser and flue gas, flame shape, heat release distribution and pollutant formation.

Table 2: Selected fuel properties of methane (CH_4, representing natural gas) and hydrogen. All values given in (25 °C/ 0 °C, 1.01325 bar). Data calculated with [28]

	Unit	100 % CH_4	100 % H_2
$H_{i,vol}$	MJ/m^3	35.89	10.79
$H_{i,m}$	MJ/kg	50.03	120.01
d	-	0.5571	0.0698
W_S	MJ/m^3	53.37	48.24
O_{2min}/Air_{min}	m^3/m^3	2.0 / 9.524	0.5 / 2.381
T_{ad} (λ = 1, O_2 / air)	°C	2,765 / 1,951	2,784 / 2,106
s_L (λ = 1, air)	cm/s	38.57	209
$V_{n,flue, wet}$ (λ = 1, air)	m^3/MWh	1,055	961
LEL with air	vol.-%	5	4
UEL with air	vol.-%	15	75
MIE [69]	mJ	0.28	0.017

Though the Wobbe Index W_S is a widely used fuel property, particularly in the gas industry, to evaluate the interchangeability of different fuel gases in an application [31], its usefulness as an interchangeability criterion is very limited in the context of hydrogen. The difference in W_S between pure methane and pure hydrogen is about 10%, while for all other properties given, the differences are significantly larger. Also, the Wobbe Index by its very definition only takes into account the heat release without any form of active combustion control to moderate the energy input into the process. Aspects such as flame stability, combustion efficiency or pollutant formation are not considered by this property, yet are crucially important when discussing hydrogen combustion [70]. These limitations are particularly relevant in the context of industrial combustion processes which tend to be more demanding than residential heating appliances [36], for example.

The adiabatic combustion temperature of hydrogen is significantly higher than that of natural gas under identical conditions, at least as long as air is used as oxidiser. This means that hydrogen flames tend to be hotter in a real application as well, so that local overheating of components can be a concern. Another immediate consequence of these higher temperature peaks is that there is a potential for higher NO_X emissions with hydrogen combustion than with natural gas, as NO_X formation is highly dependent on local temperatures. The main pathway towards NO_X formation is still the thermal pathway though, which means that most of the well-established primary measures to reduce NO_X formation, for example fuel and/or air staging, flue gas recirculation, flameless oxidation/MILD combustion or oxy-fuel combustion, have been proven to be effective with hydrogen combustion as well, though they may require some adaptations

(cf. for example [71-73]). It is worth noting that contrary to the combustion with air, the differences in adiabatic temperatures in the oxy-fuel combustion of methane and hydrogen are relatively small, although the values are very high. This has consequences for the NO_X emissions when switching from natural gas to hydrogen, which will be discussed in a later chapter (cf. **Chapter 6)**.

One of the key differences in the combustion of natural gas and hydrogen is the combustion velocity: the laminar combustion velocity of H_2 is about five times higher than that of CH_4 in stoichiometric conditions. At the same time, the minimum ignition energy (MIE) of hydrogen is also lower by about one order of magnitude which means that it is far easier to ignite hydrogen than natural gas. As a consequence, there is a much higher risk for flame flash-backs with hydrogen in premixed burners, unless the burners have been specifically designed or modified to be used with hydrogen. This is, however, less of an issue with the non-premixed burners which are commonly used in industrial process heating. In these combustion processes, flame stabilisation is controlled mostly by mixing, not determined by the balance of chemical kinetics and the flow field. As there is no combustible mixture upstream of the flame, a flash-back cannot occur in a non-premixed combustion process as the flame cannot physically propagate upstream. Thus, many industrial burners originally designed to be used with natural gas can safely be operated with hydrogen [74], though usually with a reduced performance in terms of efficiency or emissions, compared to a bespoke burner for hydrogen.

Another quite literally obvious difference between the combustion of natural gas and hydrogen is that hydrogen flames (with air) are barely visible with the naked eye. This is visualised by **Figure 20** which shows a comparison of a natural gas flame and a hydrogen flame in a lab-scale non-premixed burner, operated with the same firing rate P, air excess ratio λ and Swirl Number S. While the natural gas flame is easily visible with the naked eye (left-hand side), the hydrogen flame is not (center image). However, with an increased exposure time, it can be seen that hydrogen flames actually emit a reddish hue in the visible spectrum, albeit with very low luminosity (right hand side).

From a safety perspective, there are also significant differences between natural gas/methane and hydrogen that need to be taken into account. While the lower explosion limits of both fuels are quite similar (5 and 4 vol.% in air, respectively), the upper explosion limit of hydrogen is much higher (75 vol.% in air, compared to 15 vol.% for CH_4). As previously mentioned, the minimum ignition energy of hydrogen is lower by one order of magnitude than that of methane [69], though at low concentrations (< 10 vol.%), it is in the same range as with natural gas [75].

Given the much lower density but greater diffusivity of hydrogen, tightness of installations, especially joints, is a safety-related concern as well.

The flame ionisation characteristics of hydrogen flames are another important safety-related aspect, especially when compared to natural gas flames. Ionisation probes in industrial burners serve to ensure the operation of the burner. A small current flows through the flame being conducted by the ions within the flame. If the current is disrupted, the burner management system assumes that the flame is extinguished and cuts the fuel gas flow in order to prevent flooding the furnace with a combustible gas, an inherently unsafe state. The formation of the relevant ions requires the presence of carbon, which means that this approach does not work with hydrogen flames.

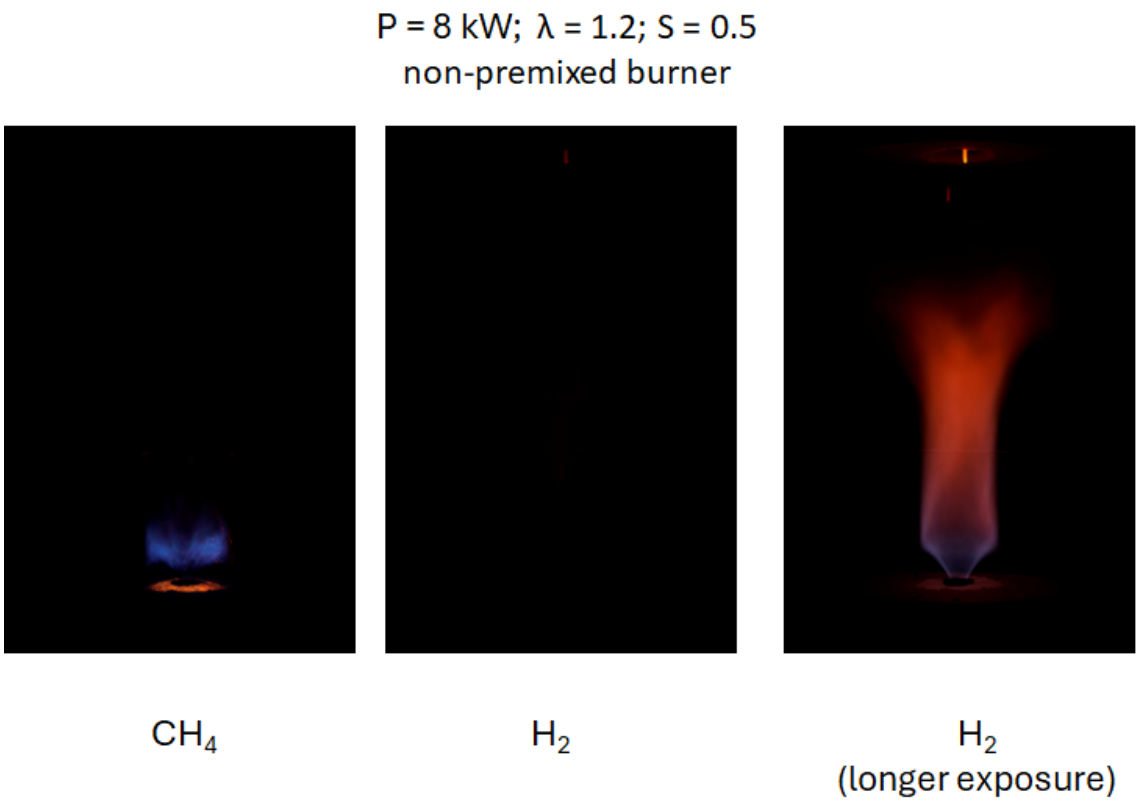


Figure 20: Images of a methane flame (left hand side) and a hydrogen flame (center and right hand side) with identical burner settings (Source: GWI).

Figure 21 shows experiments where the ionisation current in a non-premixed flame was measured for fuel gases ranging from natural gas to pure hydrogen. While the measured currents are sufficient for a wide range of fuel blends, the signal is insufficient for pure hydrogen combustion. The figure also shows results from two other flame monitoring techniques that are commonly found in technical combustion processes, infrared or ultraviolet sensing (IR and UV respectively). IR sensing is in principle well-suited to detect the presence of a hydrogen flame, but only in a relatively cold environment such as a boiler. In a high-temperature application like an industrial furnace where the refractory from the furnace wall will also emit IR radiation, an IR-based monitoring system will be unable to distinguish the different IR sources and reliably detect the presence of a flame. UV sensing has been proven to work with hydrogen flames even in a hot environment, though the sensitivity of the sensor has to be adjusted as the UV signal is weaker than with a natural gas flame, especially when flue gas recirculation (FGR) is used to prevent NO_X formation as this will reduce the signal strength even further.

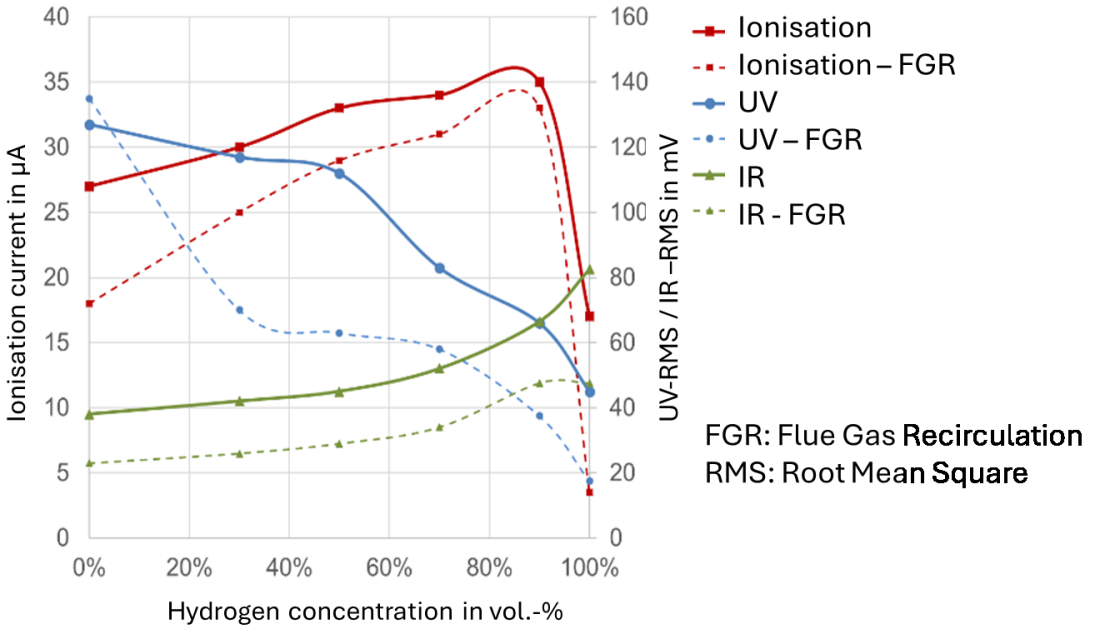


Figure 21: Effects of hydrogen admixture into natural gas on different flame monitoring techniques in a non-premixed flame [71]

6. Switching fuels: consequences and considerations for industrial combustion processes

Given the wide variety of technical combustion processes across all sectors, it is evident that the consequences of a fuel change may be quite different, depending on the application. While this is particularly relevant for comparisons between applications in different sectors, it is also true for many industrial heating applications, as there is a high degree of specialization and heterogeneity in the industrial sector alone. Thus, while there are similarities between the effects that a switch to hydrogen combustion can have in different applications and sectors, there is also a need for case-by-case analysis.

Control strategies and physical adaptations of the burner geometry

The first major difference to be accounted for when switching from natural gas to hydrogen relates to the required quantities of fuel and oxidiser. The net calorific value (in volumetric terms) of hydrogen is lower by about a factor of 3, compared to methane. At the same time, hydrogen requires less oxygen (or air) per molecule than natural gas for complete combustion.

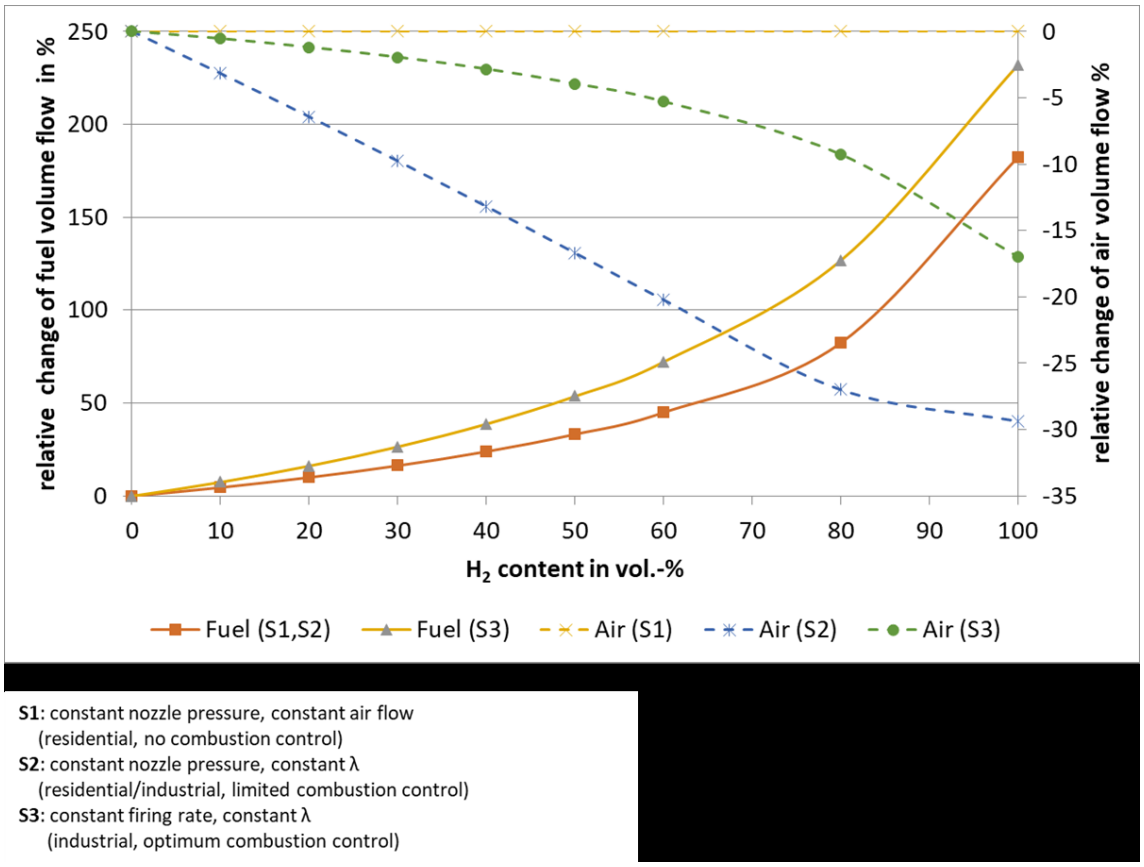


Figure 22: Impact of different combustion control strategies and hydrogen admixture rate on volume flows of fuel and air (Source: GWI)

From an end-user perspective, there are three main options how to deal with this change of the fuel properties if the application was originally adjusted to natural gas. These options are visualised in **Figures 22** and **23**.

The first option (Scenario 1, S1) is to do nothing at all which means that due to the reduced Wobbe Index of the hydrogen/natural gas blend, the firing rate will be reduced while the fuel volume flow will actually increase. This scenario also assumes that the supplied amount of air will remain constant so that the reduced minimum air requirement of the fuel blend will also lead to increasingly higher air excess ratios. The effects of this scenario can be seen in **Figure 23**, based on calculations for a reference case with a firing rate of 100 kW and an air excess ratio of 1 with methane. This approach would be representative of a residential gas heating appliance without combustion control [36] and without any physical changes to the burner to accommodate hydrogen, *e.g.* by changing the fuel nozzles. Note than in real life, the appliance would not be adjusted to stoichiometric combustion, but to a significant air excess.

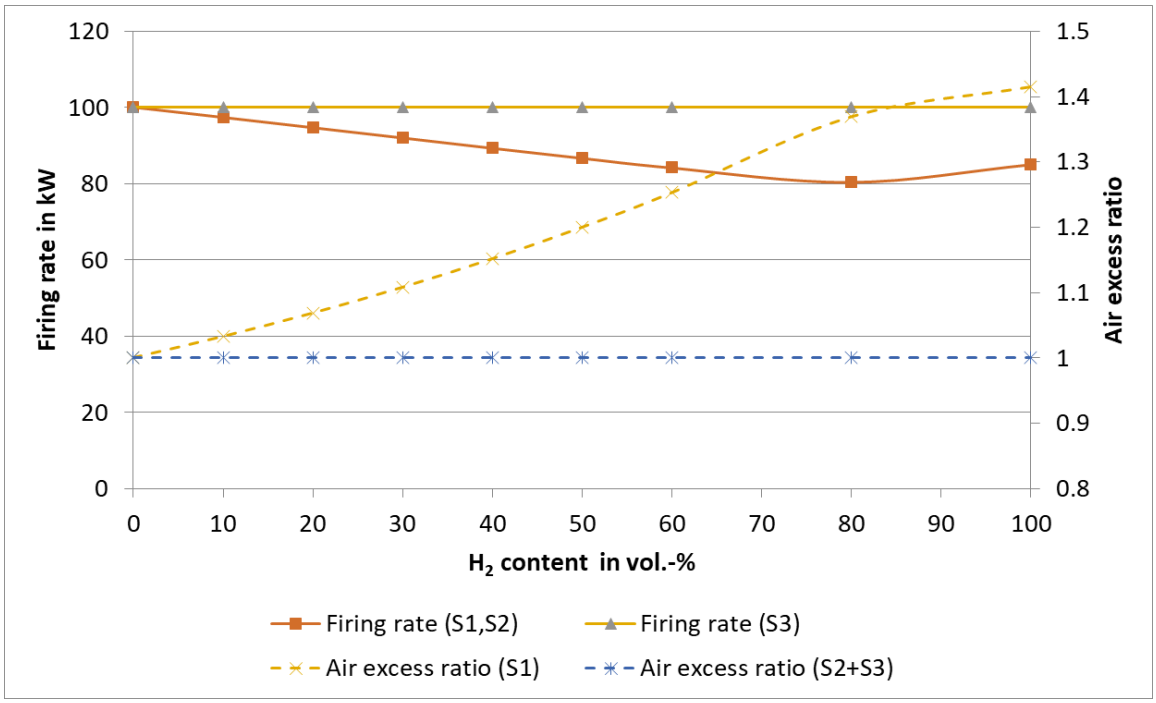


Figure 23: Impact of different combustion control strategies and hydrogen admixture rates on firing rate and air excess ratio (Source: GWI).

The second strategy (S2) shown in the diagram assumes that at least the air excess ratio is controlled by a burner management system, based, for example, on input from an oxygen sensor in the flue gas duct. The volume flow of air is reduced with increasing levels of hydrogen in the fuel to maintain the O_2 level in the flue gas, while the volume flow of the fuel gas is free to change like in Scenario 1. This approach gives the operator a limited control over the combustion process and can still be found in some industrial installations.

The third scenario (S3) presumes that both fuel and air volume flows can be controlled independently from one another to always maintain the intended firing rates and air excess ratios. This is a control strategy that is found in more advanced industrial firing systems. Here, the volume flow of fuel is determined by the calorific value of the fuel blend, while the volume flow of air is a function the fuel's minimum air requirement, both properties which are only dependent on the fuel composition. This represents an optimum response to the change in the fuel by minimising the impact of the fuel change on the process as a whole, as can be shown both by CFD simulations and experimental investigations (cf. for example [76-79]). In the case of a complete conversion from natural gas to hydrogen, this approach results in an increase of the required volume flow of fuel by about a factor of 3.3 (*i.e.* a relative change of 230%, as indicated in **Figure 22**), while the necessary volume flow of air drops by almost 20%.

In the context of burner control in thermal processing applications, there is another important control strategy that needs to be discussed, the so-called fixed-ratio approach. In this still very common control approach, the volume flows of fuel and oxidiser are always in a fixed ratio, *i.e.* one flow follows the other in a linear fashion. This simple control approach allows burner operators to respond to changes in the load of the furnace, *e.g.* for going from full load to half load. But this control approach inherently assumes that the chemical composition of the fuel does not change. This is, however, not the case when discussing hydrogen admixture into natural gas or even the full

conversion to hydrogen. Given the differences in the minimum oxygen and air requirements, switching from methane (as a stand-in for natural gas) to hydrogen would lead to an increase of the air excess ratio by a factor of approximately 4. Even for cases where a furnace is switched from one natural gas to another, this approach can already lead to significant operational and even safety-related problems [80]. The consequences of such a control approach for a fuel switch from methane to hydrogen are visualised for a combustion process with an assumed constant firing rate of 1 MW in **Figure 24.**

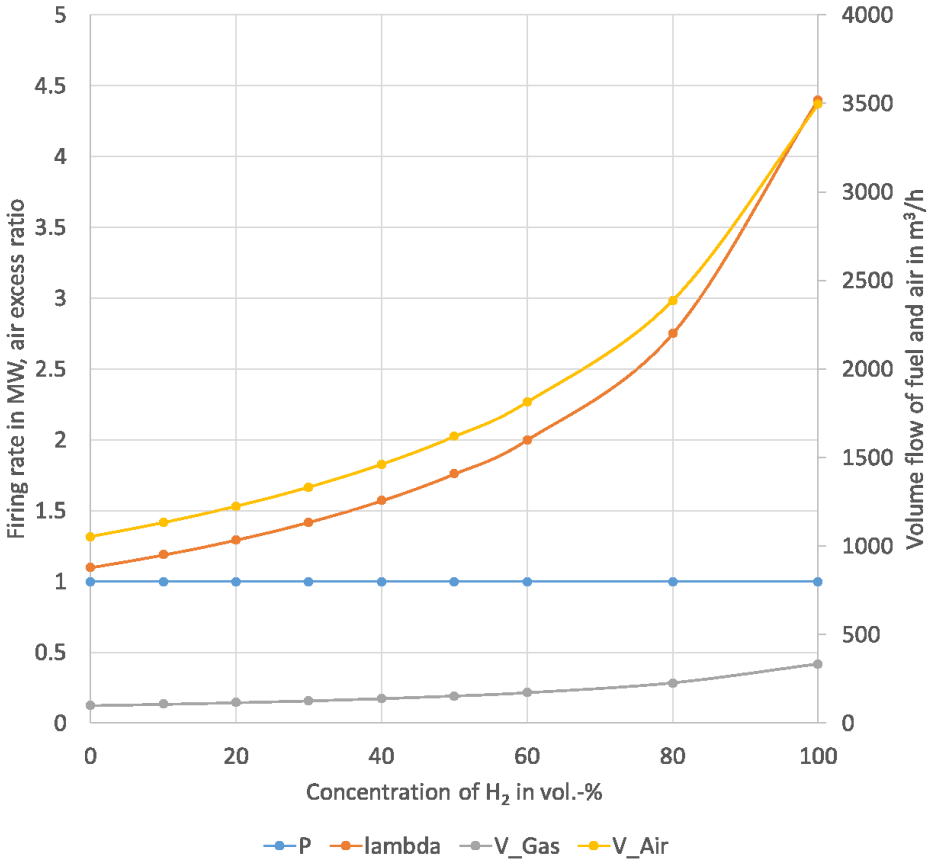


Figure 24: Effects of a fixed-ratio control approach on firing rate and air excess ratio for different methane-hydrogen blends. (Source: GWI)

These theoretical considerations already point to a crucial requirement of any burner management system that is supposed to be able to operate with natural gas, hydrogen or any blends of the two: it is essential that at least the calorific value of the currently available fuel is known, and that the volume flows of fuel and oxidiser can be controlled independently from one another.

Volume and mass flows of both fuel and oxidiser will inevitably change when switching fuels. This will have an impact on local velocities and flow patterns in the combustion space, which in turn can affect flame shape, heat transfer or pollutant formation. One effective approach to address this in non-premixed burners is by adapting the nozzles for fuel and oxidiser. In [43], a case is presented where an existing natural gas oxy-fuel burner in a rotary aluminium recycling furnace was physically converted to be used with hydrogen, using CFD analyses. As the volume and mass flows have to change to maintain firing rate and oxygen excess ratio despite the different fuels, it was decided to modify the diameters of both the fuel and oxygen nozzles in such a way that the momentum

flux ratios of the streams remained roughly constant when switching from natural gas to hydrogen:

$$\frac{\dot{m}_{fuek} \cdot u_{fuel}}{\dot{m}_{O2} \cdot u_{O2}} \approx const.$$

The flame shape in non-premixed combustion is primarily dependent on the turbulent mixing of fuel and oxidiser in the combustion space, which is in turn dependent, among other things, on the momenta of fuel and oxidiser streams. Consequently, the diameter of the fuel nozzles was increased by 14%, while the diameter of the oxygen nozzle was reduced by 10%, in order to maintain a roughly similar ratio of the two momentum fluxes.

The CFD simulations indicate that this strategy is valid: flame shapes and thermal efficiencies remained almost identical when switching from natural gas to hydrogen while NO_X emissions decreased by about 40% (cf. **Figure 25**).

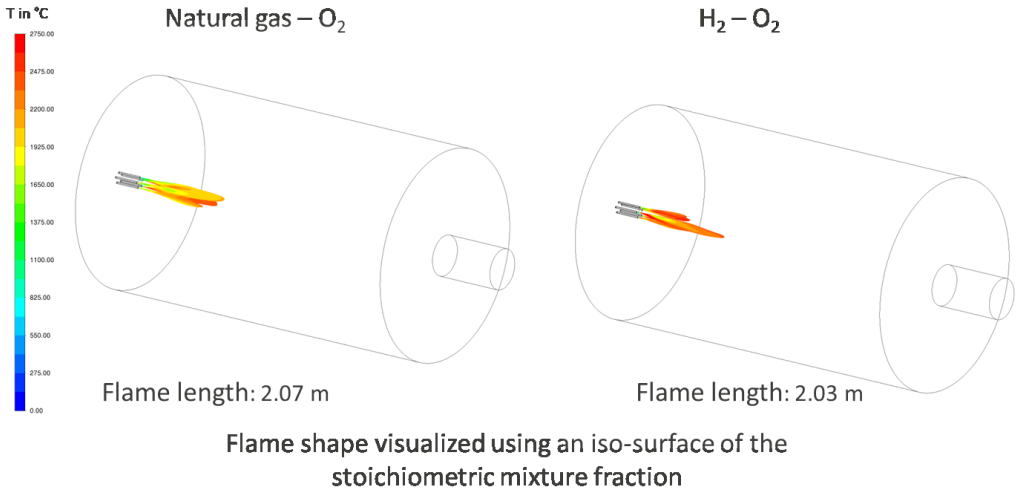


Figure 25: Visualisation of the flame shape in an oxy-fuel-fired aluminium recycling furnace using natural gas (left hand side) and hydrogen (right hand side) [43]

It may not always be possible to geometrically adapt the nozzles of the burner when switching from natural gas to hydrogen, for example if a burner system is intended to run both with both fuels, depending on the supply situation. In this case, there are two options: either the delivery pressure of the fuel gas is increased to compensate for the reduced calorific value or Wobbe Index, or the burner is a priori designed as a dual-fuel system, with two independent fuel nozzle systems, one for natural gas and one optimised for hydrogen. **Figure 26** shows an example of one such system: despite the very different fuel characteristics, the optimisation of the different nozzles allows for very similar flame shapes [81].

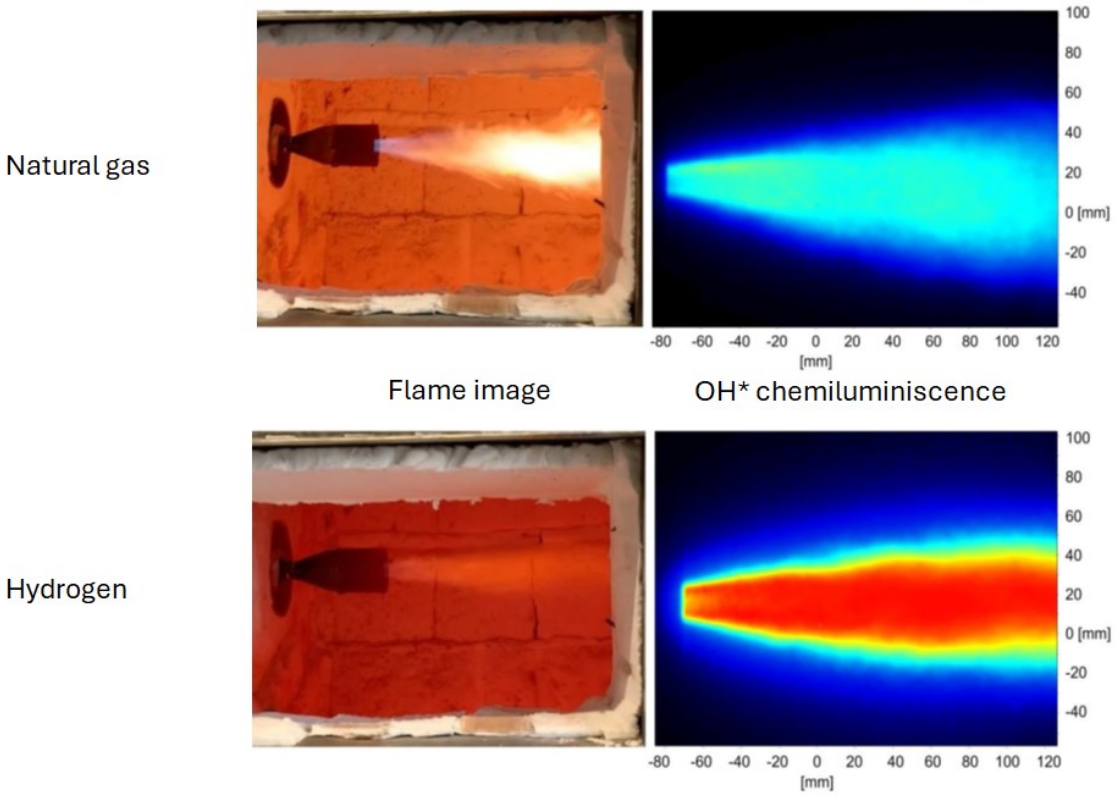


Figure 26: Dual-fuel burner system operating with natural gas and hydrogen. The images on the right hand side show flame visualisations in the UV spectrum, using OH chemiluminescence [81].*

The change in volume flows affects not only the burner itself, but also the plant infrastructure dedicated to transport fuel and oxidiser to the furnace as well as the flue gas system which conveys the flue gas from the furnace to the flue gas treatment and eventually the stack. For a constant firing rate and air excess ratio, the changes in the volume flows of oxidiser and flue gas are not that significant (about 20% and 10%, respectively), but the volume flow of fuel roughly triples when switching from natural gas to hydrogen. This has to be considered when planning the conversion of an industrial site, as have safety-related aspects.

Efficiency and heat transfer

Efficiency is an obvious concern when discussing any fuel change, especially in energy-intensive industries where large quantities of energy are being transformed and energy cost make up a significant part of the operational cost. In terms of thermal efficiencies, the differences between the combustion of methane and hydrogen are, for the most part, relatively minor, as is visualised in **Figure 27**.

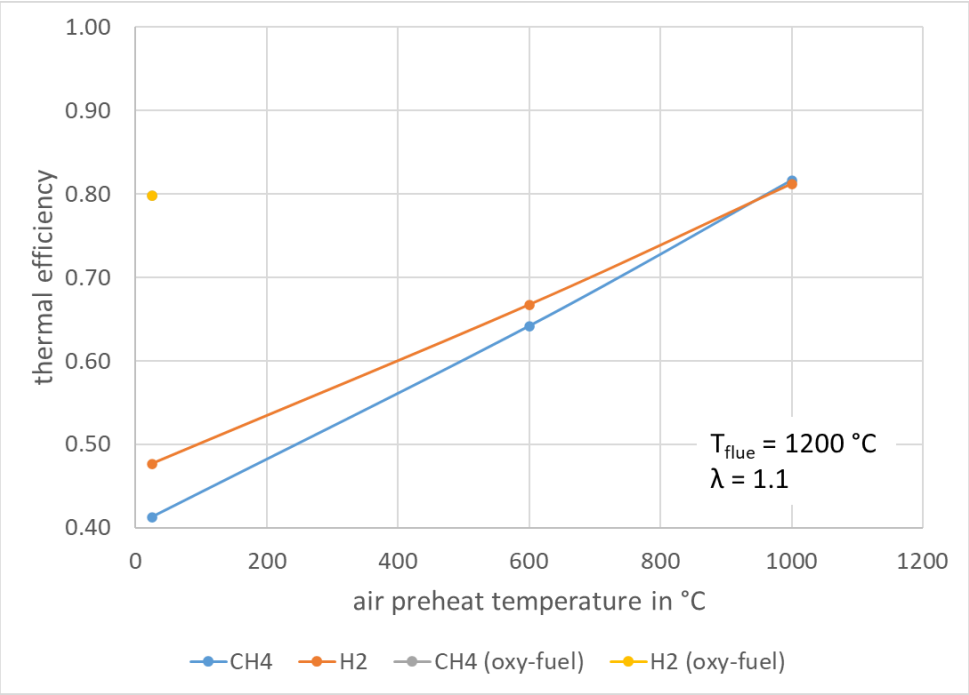


Figure 27: Thermal efficiencies of methane and hydrogen combustion as functions of the air preheat temperature and oxy-fuel combustion. (Source: GWI).

For a given air excess ratio and flue gas temperature (chosen to be similar to an aluminium scrap recycling furnace), the thermal efficiency of the combustion of hydrogen is better than with methane, though the difference shrinks with increasing air preheat temperature. At a preheat temperature of about a 1000°C (the exact number depends on the chosen air excess ratio and flue gas temperature), the situation flips and methane combustion becomes slightly more efficient, since more air at a high temperature is introduced into the system than with hydrogen combustion, so the heat recovery from the flue gas becomes more effective in this case.
In the case of oxy-fuel combustion, the thermal efficiencies of the combustion of both fuels are virtually identical. It should be noted though that this efficiency consideration only covers the energy-related aspects of the furnace operation itself: it does take into account neither the energy requirements to produce oxygen nor the improved heat transfer due to oxy-fuel combustion (cf. **Figure 15**).

Another energy-related concern about the switch from natural gas to hydrogen is about how it might affect heat transfer, in particular radiative heat transfer, in an industrial furnace. Radiative heat transfer is the most dominant heat transfer mechanism in high-temperature environments, and both CO_2 and H_2O as tri-atomic species in the flue gas participate in it. Since the flue gas composition changes when switching from natural gas to hydrogen, radiative heat transfer and thus the overall efficiency of the furnace process could be impacted. However, CFD simulations indicate that the overall energy efficiencies of industrial furnaces are not much affected by the fuel change (cf. for example [43, 79]). These findings are, to some degree, corroborated by experimental results in the semi-industrial or even fully industrial scale, albeit only for hydrogen admixture into natural gas (up to 50 vol.% in the case of semi-industrial tests [77] (cf. **Figure 28**), up to 15 vol.% in full-scale tests in a float glass furnace the UK [82]).

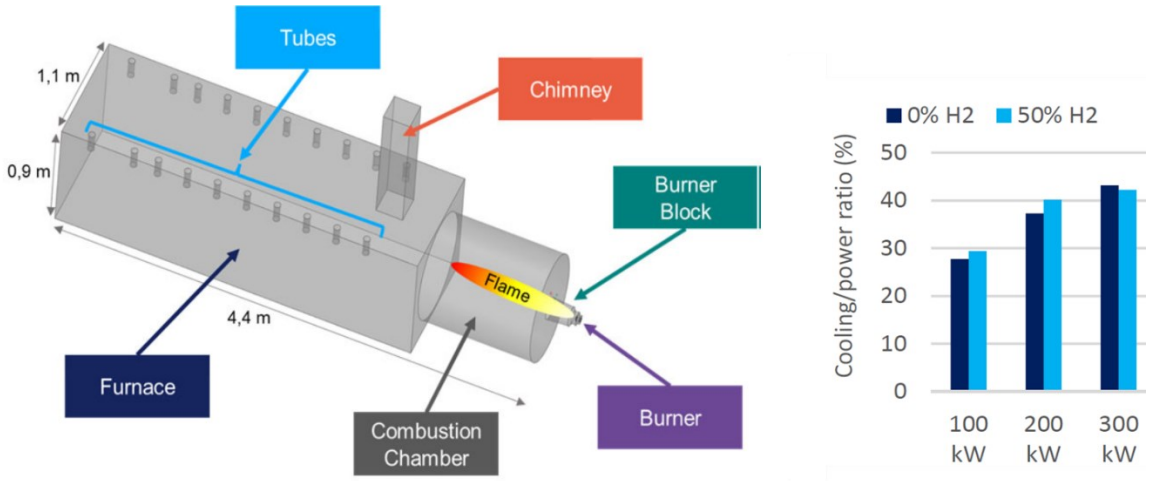


Figure 28: Experimental setup of a semi-industrial combustion test rig (left hand side) and comparison of cooling to firing rate ratios for different hydrogen admixture rates (right-hand side, [77])

Figure 28 shows the layout of a semi-industrial combustion test rig in which a burner was tested under realistic conditions. The burner was operated with different fuel blends (natural gas and up to 50 vol.% hydrogen) at firing rates between 100 and 300 kW. Air excess ratios and furnace temperatures were kept constant. Among other things, the heat flux needed to keep the furnace at the target temperature was measured, which can be interpreted as the heat load in a real-life industrial furnace. As the diagram on the right-hand side of the figure indicates the ratio of cooling to firing rate remained almost constant over the whole range of investigated burner settings.

Flue gas composition and product quality

One common aspect of any fuel change is that the composition of the flue gas will change. Compared to the combustion of natural gas, hydrogen combustion will result in a total elimination of CO_2 emissions (unless there are process emissions of carbon dioxide), but also in a higher concentration of water vapour in the flue gas (assuming a constant air excess ratio).

Table 3 shows the volumetric concentrations of the main components of the flue gas from the combustion of methane and hydrogen, for both air and pure oxygen as oxidisers, based on an elementary combustion calculation. The air excess ratios λ were chosen to be typical values for industrial combustion processes. Since oxy-fuel processes are usually run closer to stoichiometry, a lower excess ratio was used for the oxy-fuel case, to reflect industrial practice.

Table 3: Concentrations of the main flue gas species when burning methane and hydrogen for typical air excess ratios.

		combustion with air (λ =1.1)					**combustion with oxygen (λ = 1.02)**			
		CH_4		**H_2**			**CH_4**		**H_2**	
	unit	**wet**	**dry**	**wet**	**dry**		**wet**	**dry**	**wet**	**dry**
CO_2	**vol.-%**	8.71	10.55	0.00	0.00		32.89	96.15	0.00	0.00
H_2O	**vol.-%**	17.43	-	32.06	-		65.79	-	99.01	-
O_2	**vol.-%**	1.74	2.11	1.60	2.36		1.32	3.85	0.99	100.00
N_2	**vol.-%**	72.12	87.34	66.34	97.64		0.00	0.00	0.00	0.00

It can be seen that in the case of combustion with air, the water vapour concentration in the flue gas of a hydrogen combustion is almost two times higher than in the combustion of methane (assuming a constant air excess ratio). In the case of oxy-fuel combustion, the flue gas of hydrogen combustion consists almost entirely of water vapour.

These different compositions of the furnace atmospheres and the flue gas can have an impact on the overall energy balance of the furnace process due to changes in the isobaric heat capacities, but there are also concerns that the switch to hydrogen combustion may have a negative impact on product quality or the refractory materials (which will be discussed in more detail in a separate report D4.4). Product quality is obviously of paramount importance for any manufacturing process. In the context of industrial process heating, product quality can be affected by changes in the heat transfer characteristics, but also by changes in the furnace atmosphere which may interact with the product itself.

However, research in other industries can already provide some insights. Based on existing literature, the effects can be very specific and also depend very much on the material in question. Some investigations show [83, 84] no significant impact of hydrogen combustion on the properties of the investigated aluminium alloy samples, while other groups [85, 86] reported that for other aluminium alloys, effects were detectable. [85] also reports that the choice of combustion concept (hydrogen with air, pure oxygen or oxygen enrichment) can affect the formation of dross and the hydrogen content in the aluminium.

Similarly to the aluminium recycling industry, the glass industry is an energy-intensive basic materials industry which today relies heavily on natural gas and thus faces the challenge of decarbonisation. Process temperatures in glass manufacturing, particularly during the melting process, are significantly higher than in the aluminium industry, and furnaces are often also much larger, with firing rates up to 100 MW in the case of float glass furnaces, *e.g.* for glass for the automotive sector or windowpanes. Along with an increasing share of electrification, hydrogen is considered as one pathway towards decarbonization, and several projects have been investigating this approach over the last few years. In Germany, one such project is the HyGlass project [79, 87], which, among other aspects, also investigated the effects of the fuel change on glass quality. It was

found that the fuel change from natural gas to hydrogen did in fact cause changes in the colouration of the investigated glass samples [88], but that this effect is – for the most part – due to the change in the water vapour concentrations in the flue gas which removed certain trace components from the melt. Similar effects had previously been observed when oxy-fuel combustion was first introduced into glass melting. They were counteracted by modifications of the raw material composition. Another project in the UK glass industry explored the impact of hydrogen admixture in a full-scale float glass furnace, both by globally admixing up to 15 vol.% H_2 into natural gas for the entire furnace, and by switching one port completely to hydrogen [82]. In both cases, no significant impact on glass quality was reported. A similar project but with higher hydrogen admixture rates (30% by energy, which corresponds to about 70% by volume) is currently underway in Germany [89]. So far, no negative impact of the fuel change on product quality has been reported.
In the context of the aluminium recycling industry, a recently concluded project in Germany [83] found no significant impact of hydrogen combustion on the mechanical properties of the investigated aluminium alloys, based on measurement of densities and soluble hydrogen in the aluminium melt as well as mechanical analysis of the material properties. [84] gives an overview over recent high-TRL activities to introduce hydrogen combustion into the aluminium recycling industries.

The use of hydrogen-rich fuels in thermal process heating is not new per se. Prior to the introduction of first heavy fuel oil and later natural gas, many industrial processes were heated with so-called “town gas” or “producer gas”, *i.e.* gasified coal, which consists mainly of hydrogen and carbon monoxide [90]. Even today, some large glass melting furnaces with production rates of up to 700 t/d are operated with producer gas in China [91]. In Germany and the UK, the use of coke oven gas (COG), a by-product of coke production for the steel industry, is being explored and in some cases even already commercialised to provide process heat for other industries, *e.g.* in the glass, aluminium or steel refinement industries [92-94]. COG also consists of up to about 60 vol.% of hydrogen and can thus be considered as a precursor to the conversion to pure hydrogen. Given that some of these plants have been in full production for some time, the operators do not seem to expect quality-related problems from using this hydrogen-rich fuel.
Similar questions arise about the impact of the fuel switch on the refractory material. Again, this will be explored in more detail in a dedicated deliverable within the project, but other studies, *e.g.* [95, 96], show that while some refractory materials can be affected, there are often alternatives already available. This is just one more aspect to be considered when discussing the use of hydrogen for high-temperature process heating.

Nitrogen oxides (NO_X)

NO_X emissions usually are concern for equipment operators when discussing a change from natural gas to hydrogen in technical combustion systems. NO_X formation in the combustion of gaseous fuels is dominated by the thermal formation mechanism, unless the fuel contains non-molecular nitrogen, *e.g.* in the form of ammonia (NH_3). As hydrogen has a higher adiabatic combustion temperature than natural gas when burning

with air (cf. **Table 2**), stronger NO_x formation is to be expected, especially in a non-premixed combustion process. Various references indicate higher NO_x concentrations in the flue gas when admixing hydrogen to natural gas or switching entirely to hydrogen [74, 77, 79], unless measures are taken to compensate for the stronger propensity of hydrogen combustion to produce NO_X. (cf. [71, 97], for example). **Figure 29** gives an example of how a non-premixed industrial process burner [74], designed and optimised for use with natural gas, typically responds to hydrogen admixture in terms of NO_X formation. Air preheating exacerbates the trend toward increased emissions even further, due to higher local temperatures in the flame core.

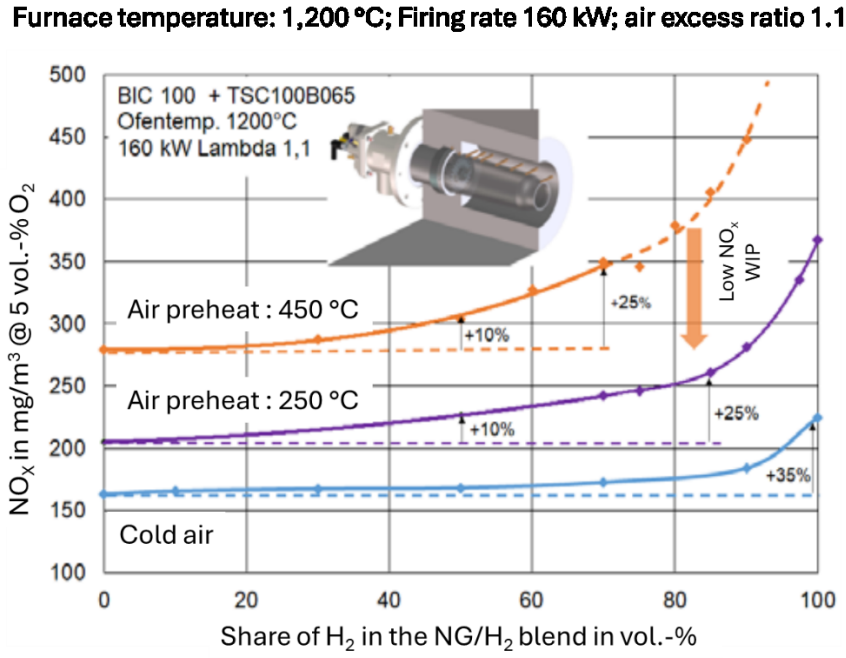


Figure 29: Impact of H_2 admixture and air preheating on NO_X emissions for a typical industrial process burner. Translated and adapted from [74]

The higher adiabatic combustion temperatures of hydrogen in comparison to natural gas do not automatically lead to higher NO_X emissions, however. Both for natural gas and hydrogen, thermal NO_X formation is the dominant formation mechanism, which is well understood. Many of the measures to reduce NO_X emissions work just as well with H_2 as with natural gas, they just have to be adapted to the fuel. **Figure 30** provides an example from a commercially available burner designed for use in a steel reheating furnace [73].

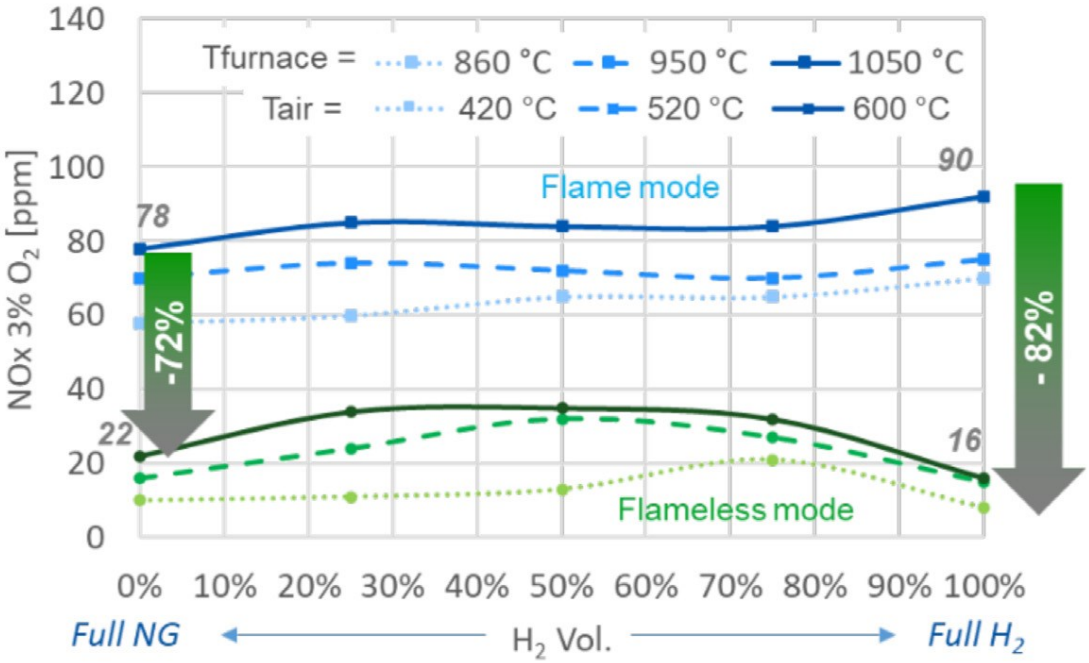


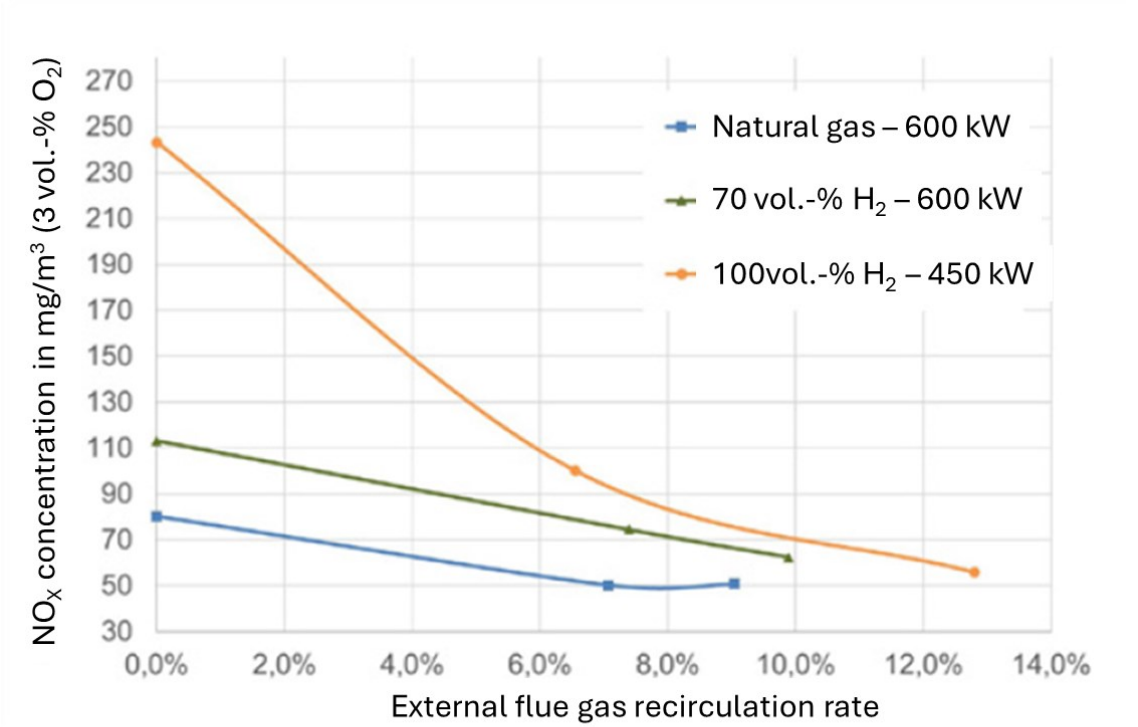
Figure 30: NO_X concentrations for various natural gas / hydrogen blends and air preheat temperatures in flame and flameless mode [73]

This burner system is designed to operate in flameless mode, but can also switch to a conventional flame mode. The diagram shows comparable NO_X concentration levels in both modes for all fuel blends as well as a significant overall decrease of emissions once the burner is switched to flameless mode.

Other examples from different applications also demonstrate that most primary measures to reduce NO_X formation work well with both hydrogen and natural gas.

Figure 31 shows results of experimental investigations looking into the efficacity of external flue gas recirculation to reduce NO_X emissions of a forced-draught burner [71]. These burners are usually found in applications with lower process temperatures, *e.g.* steam generation, and external flue gas recirculation is a common method to mitigate pollutant emissions in such applications. If no flue gas recirculation is used, the measured NO_X concentrations in the flue gas are significantly higher for hydrogen than for natural gas, a situation which is further acerbated if the burner is operated in partial load. Once flue gas recirculation is used however, very similar concentrations can be achieved for both fuels although the recirculation rate has to be higher when hydrogen is used.

In an oxy-fuel combustion process, the situation changes somewhat anyway: the adiabatic combustion temperatures of both natural gas and hydrogen with oxygen are almost identical, and the availability of molecular nitrogen (either due to impurities in the fuel or oxidiser, or due to infiltration air) in the reaction zone becomes the limiting factor [35, 41, 42]. Natural gas usually contains some nitrogen, and in industrial firing systems, oxygen is often not of the highest purity either, nor does it need to be. Both factors create a situation in which there can be more nitrogen available in an oxy-fuel combustion process with natural gas than with hydrogen, so NO_X emissions can actually be reduced when switching to hydrogen [43] (cf. **Figures 33** and **34**).



$$ARF\ Rate = \frac{\dot{V}_{recirculated}}{\dot{V}_{flue}} \cdot 100\%$$

Figure 31: NO_X concentrations in a forced-draught burner with external flue gas recirculation[71]

There is another aspect to consider besides the chemical kinetics of thermal NO_X formation. As previously discussed, pollutant emissions (and their respective legal limit values) are today usually expressed as species concentrations in the dry flue gas, referenced to a fixed excess oxygen value to ensure comparability. Typically, units such as [ppm] or [mg/m^3] are used to quantify emissions, although they actually only provide concentrations of the pollutant. The flue gas sample is dried during the measurement process in order to protect the sensor from humidity. As long as the fuel composition does not change significantly, this is a perfectly valid approach to quantifying and regulating pollutant emissions in a technical combustion process. However, the concentration of H_2O in the flue gas of a hydrogen combustion process is much higher than in the flue gas of a CH_4 (or more general, natural gas) combustion process (cf. **Table 3**), as long as the air excess ratios are the same, while the flue gas volume per unit of energy release is lower.

As a consequence, the emissions of NO_X (or any other pollutant species) from hydrogen and natural gas combustion processes are not directly comparable. Even if the same amount of NO_X were formed in both cases in absolute terms (*e.g.* expressed in [mg/s]), the resulting concentration of NO_x in the dry flue gas the hydrogen combustion process would be higher, as more water vapour is removed.

Figure 32 visualises this issue based on experimental data obtained in the German "HyGlass" project [79, 87] where a typical burner system used in glass melting furnaces was investigated with both natural gas and hydrogen in a semi-industrial test rig.

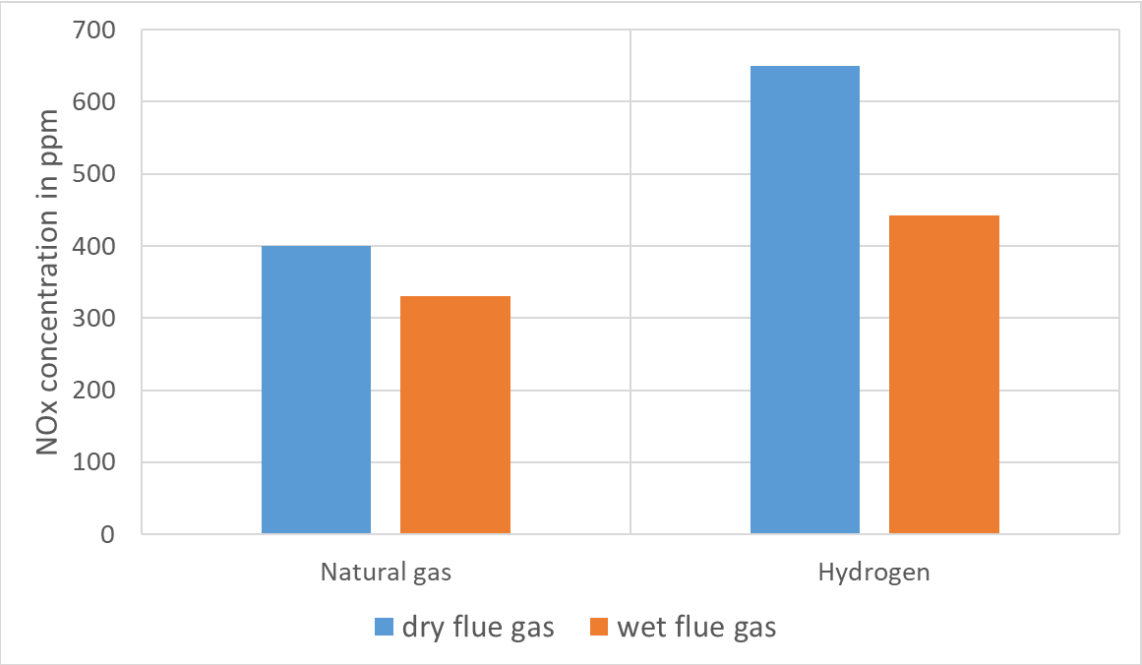


Figure 32: Measured NO_x concentrations from natural gas and hydrogen combustion in a semi-industrial test rig [79]

Operational conditions were constant (firing rate: 500 kW, air excess ratio 1.1, air preheat temperature 1150°C), only the fuel was changed from natural gas to hydrogen. The burner itself was not modified in any way. As expected, the measured NO_X concentrations increase when switching from natural gas to hydrogen. With the conventional measurement approach using a dried flue gas sample, a NO_X concentration of 400 ppm is measured for natural gas, while hydrogen combustion yields 650 ppm, *i.e.* concentrations rise by more than 60%. However, if the wet concentrations are considered (which can easily be calculated when the fuel and air excess ratio are known), values of 330 ppm for natural gas and 442 ppm for hydrogen are obtained, *i.e.* an increase of “only” 34%. Just by the technically necessary drying of the flue gas sample, the difference between the measured values for natural gas and hydrogen almost doubled. Using dried concentrations as a metric for pollutant emissions, it becomes very difficult to differentiate between the actual increase of NO_X due to the changed fuel and the artificial increase due to an ill-suited criterion.

In the context of oxy-fuel combustion, this problem becomes even more critical as the flue gas of a H_2-O_2 combustion consists almost entirely of water vapour. **Figure 32** shows results from CFD simulations carried out within the COSIMa project ([50, 98]) which, among other things, investigated the use of hydrogen-oxygen combustion for glass melting applications. For both simulations, the firing rate was set to 320 kW and the oxygen excess ratio to 1.02.

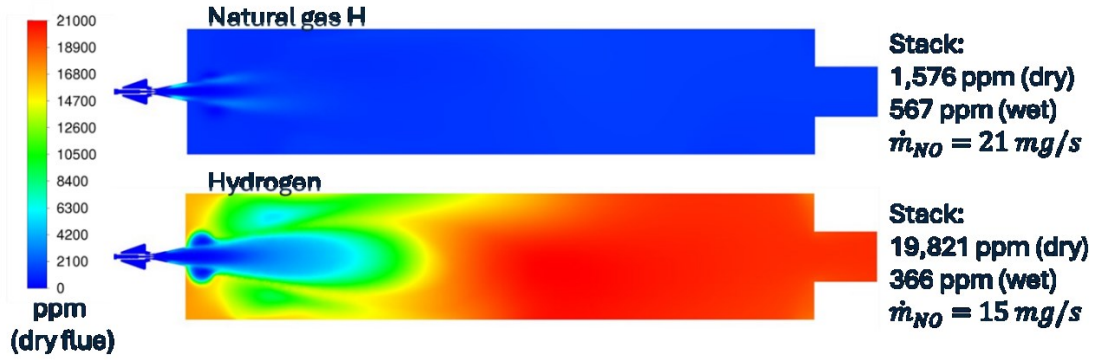


Figure 33: CFD-simulated NO concentrations in the oxy-fuel combustion of natural gas and hydrogen in a semi-industrial test rig (translated from [50])

The comparison of the contour plots of the dry concentrations in the figure indicates that hydrogen combustion leads to a drastic increase in NO formation. However, this is actually not true as shown by both the wet NO concentrations and the calculated NO mass flows at the exit of the computational domain. Both decrease when switching from natural gas to hydrogen. This can be explained physically: NO formation in oxy-fuel combustion is primarily limited by the availability of nitrogen. The natural gas used in these simulations contained some nitrogen (about 1 vol.%) and the oxygen used was also doped with 1 vol.% of N_2 in order to have a nitrogen source for NO formation even in the case of H_2 combustion (and also to mimic the effects of infiltration air). Both factors result in a higher quantity of nitrogen in the combustion space in the case of natural gas combustion, which causes higher emissions in this case.
Other metrics are better suited to capture the impact of a fuel change on the emissions performance. Often, an energy-based unit such as [mg/MJ] or [mg/kWh] is chosen as a more physically reasonable metric (see for example [50, 99-101]).
In the case of the HyGlass experiments (cf. **Figure 31**), the natural gas case yields a NO_X emission of 217 mg/MJ vs. 262 mg/MJ for hydrogen, *i.e.* an increase of only about 20%, compared to the 34% and 63% when using the wet and dry NO_X concentrations respectively. For the oxy-fuel case shown in **Figure 32**, the NO emissions decrease by about 29% when switching to hydrogen: from 66 mg/MJ to 47 mg/MJ.

Calculating the energy-referenced emissions based on dry flue gas concentrations is straightforward. The industrial standard EN 676 [102] gives the following equation:

$$NO_x = X_{NOx} \cdot 2.056 \left(\frac{21}{21 - X_{O2,meas}} \right) \cdot \frac{V_{flue,dry,min}}{H_i}$$

with NO_X as the energy-referenced emission in [mg/MJ] or [mg/kWh], X_{NOx} as the measured dry NO_X concentration, $X_{O2,}$meas as the measured excess oxygen in the dry flue gas, $V_{flue,dry,min}$ as the minimum dry flue gas volume in $\left[\frac{m^3_{flue}}{m^3_{fuel}}\right]$ and H_i as the net calorific value in either [MJ/m^3] or [kWh/m^3].

The potential of the oxy-fuel combustion of hydrogen to reduce NO_X emissions is corroborated by experimental results in a combustion test rig, which were obtained in a measurement campaign of the COSIMa project. **Figure 34** shows the measured NO emissions given in [mg/MJ]. Clearly, the fuel switch causes a significant reduction of NO formation, as was predicted by the CFD study, with reductions of 76% and 70% for λ

values of 1.05 and 1.1 respectively. During these experiments, the natural gas contained about 0.972 vol.% of nitrogen, according to a gas chromatograph used to monitor the fuel composition. Also, it seems as if the simulations' assumption of 1 vol.% nitrogen in the oxygen was rather too high.

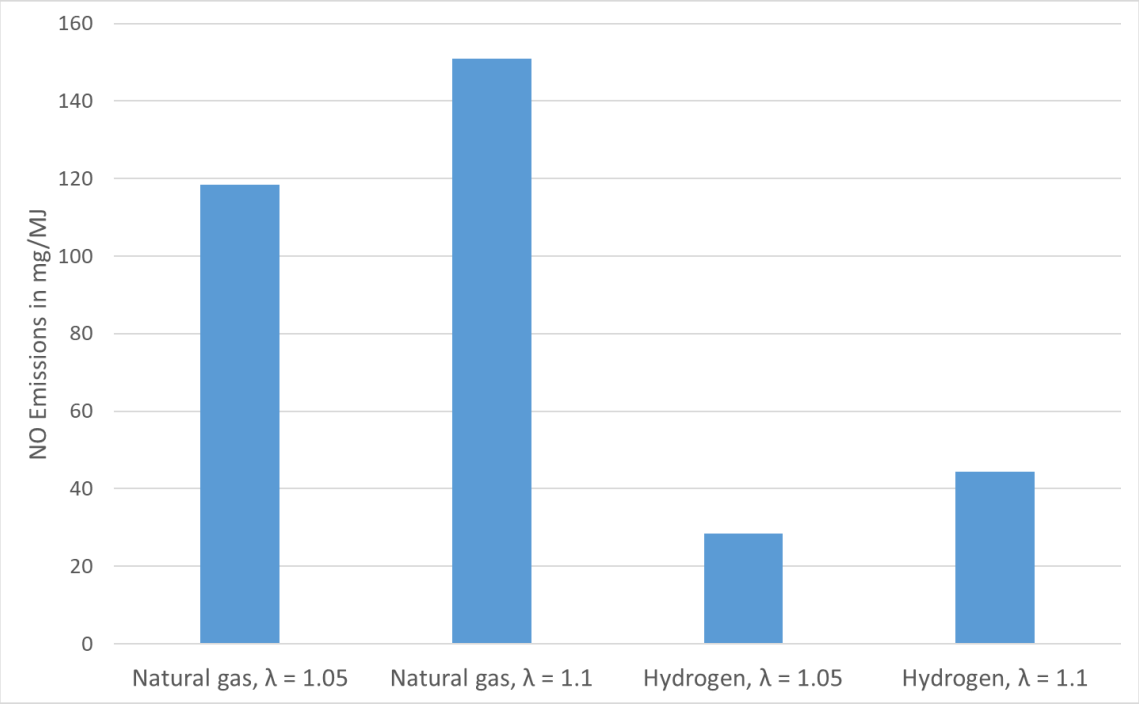


Figure 34: Experimentally obtained NO emissions from oxy-fuel combustion processes with natural gas and hydrogen (P =320 kW, (Source: GWI).

The measurements indicate that the oxygen excess ratio λ and hence, the local availability of oxygen, still plays a certain role, even in a high-temperature near-stoichiometric environment. Higher values for λ lead to an increase in NO formation both with natural gas and hydrogen which matches theoretical considerations (cf. also **Figure 10**).

These examples highlight that in order to evaluate the impact of a fuel change appropriately, it is insufficient to just look species concentrations in the dried flue gas. This approach is valid as long as the fuel does not change much in its composition, but leads to significant loss of comparability, both quantitatively and qualitatively, when applied to very different fuels like natural gas and hydrogen. Other metrics, *e.g.* the energy-referenced emissions in units such as [mg/MJ] or [mg/kWh] are better suited for this task.

7. Open issues

This report is intended to provide an overview of typical combustion processes in industrial process heating and how they may be affected by a fuel change from natural gas to hydrogen. As such, some open questions remain, especially when the H2AL project as a whole focuses on one specific application in one production site, in this case the aluminium recycling furnace operated by the project partner 2A s.p.a. in northern Italy. In this case, both the fuel and the oxidiser are to be substituted: the fuel from natural gas to hydrogen for decarbonisation purposes, and the oxidiser from air to oxygen in order to improve process efficiency and reduce NO_X emissions.
Thus, this report can only deduce consequences of these changes based on theoretical considerations and a literature review while the actual impact on the specific plant will have to be explored during the retrofit and the subsequent trial runs within the project.

One crucial aspect in this context is the impact of the fuel change on product quality and refractory: while this topic is addressed to some extent in this report, the “H2AL” project has a dedicated Deliverable (D4.4) dealing with this crucial issue which goes into much more detail.

8. Conclusions

Decarbonising industrial process heat, in particular high-temperature process heat, is a fundamental challenge in the energy transition as many energy-intensive materials like metals, glass, ceramics or cement are on the one hand essential for a modern society. On the other hand, the required high-temperature heat in these manufacturing processes is responsible for significant greenhouse gas emissions today, as the furnaces rely heavily on fossil fuels, most often natural gas.
Hydrogen from renewable sources is one promising pathway to cut greenhouse gas emissions from these applications, along with electrification, again with electricity from sustainable sources. While electrification tends to be more energy efficient overall, other aspects such as product quality or the retrofit of existing equipment and furnaces can be in favour of a fuel switch. Aluminium recycling is one industrial application where hydrogen combustion is generally seen as an important tool for decarbonisation.

From a technical perspective, hydrogen is a very different fuel, compared to natural gas. Given the high degree of specialisation and optimisation in many industries, these fuel-related specifics have to be taken into account when converting existing processes to hydrogen combustion. This refers not only to aspects such as operational safety and the combustion process itself, but just as importantly also to aspects such as product quality, heat transfer, efficiency or the formation of other pollutants, *e.g.* nitrogen oxides (NO_X).

This report provides an overview of some key concepts of industrial combustion and how they may be affected by a fuel switch from natural gas to hydrogen. Fuel characteristics such as calorific values, minimum oxygen/air requirements or Wobbe Indices are explained, as are important process parameters such as the firing rate or the air excess ratio. The fuel-specific differences between natural gas and hydrogen are discussed and the consequences for industrial combustion processes in terms of efficiency, heat transfer or pollutant formation are explored. An extensive literature review, drawn from various energy-intensive high-temperature basic materials industries, is also provided.
Based on these theoretical and fundamental considerations, guidelines of how to retrofit a technical process which is designed and adjusted for natural gas to the use of hydrogen are developed, which will provide input for both further investigations within the H2AL project and inform the retrofit strategies for an existing aluminium recycling furnace which is the ultimate goal of the project.

9. Acknowledgements and disclaimer

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Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the Clean Hydrogen Partnership. Neither the European Union nor the granting authority can be held responsible for them.

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