

HyDelta 2

WP9: Hydrogen in combustion use - NOx Effects

D9.1 & D9.2 - Literature research on low NOx hydrogen burners and developing design rules for low NOx burners

Status: Final Report

Document samenvatting

Corresponderende auteur

| | |
|--------------------------------|-----------------------|
| Corresponderende auteur | Sander Gersen |
| Verbonden aan | DNV |
| E-mailadres | sander.gersen@dnv.com |

Document historie

| Versie | Datum | Auteur | Verbonden aan | Samenvatting van de wijzigingen |
|--------|------------------|---|---------------|--|
| 1 | 9 november 2022 | Sander Gersen, Martijn van Essen Pieter Wolff | DNV | 1 ^e Conceptversie voor review rapport deel 1 door projectteam |
| 2 | 28 november 2022 | Sander Gersen, Martijn van Essen Pieter Wolff | DNV | Aanpassingen doorgevoerd |
| 3 | 27 maart 2023 | Sander Gersen, Martijn van Essen Pieter Wolff | DNV | 2 ^e Conceptversie voor review rapport deel 1 door projectteam |
| 4 | 17 april 2023 | Sander Gersen, Martijn van Essen Pieter Wolff | DNV | Aanpassingen doorgevoerd |

Verspreidingsniveau

| | | |
|-----------|---|---|
| PU | Publiek | X |
| RE | Beperkt tot <ul style="list-style-type: none"> Project partners inclusief Expert Assessment Group Externe entiteit met wie een geheimhoudingsplicht bestaat | |

Document review

| Partner | Naam |
|---|---------------------------|
| Gasunie | Jelle Lieffering |
| Stedin | Iman Pishbin |
| Enexis | Thijs Duisters |
| TNO | Nestor Gonzalez Diez |
| TNO | Mariusz Cieplik |
| DNV | Martijn van Essen |
| DNV | Pieter Wolff |
| NBNL, Gasunie, Kiwa, DNV, TNO, NEC, Hanze | HyDelta Supervisory Group |

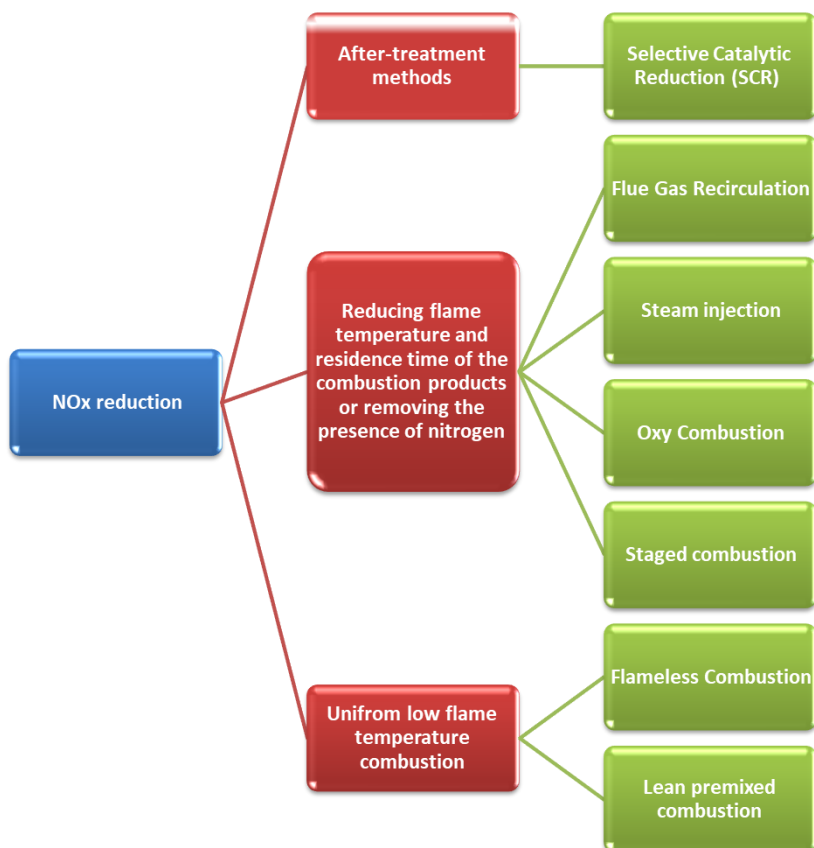
Samenvatting

De energie-intensieve industrie onderzoekt de mogelijkheid om met waterstof als brandstof de CO₂-intensiteit van hun productieprocessen drastisch te verminderen. Een van de uitdagingen is om een efficiënte verbranding te verkrijgen en tegelijkertijd de NO_x-emissies laag te houden. Dit onderzoek is gericht op de ontwikkeling van LowNO_x-waterstofbranders die nodig zijn voor grootschalige introductie van waterstof in de hoge temperatuurindustrie. Het project is opgedeeld in twee fasen. In dit rapport wordt in fase 1 een literatuurinventarisatie uitgevoerd van de verschillende brandercategorieën die in de industrie worden gebruikt met bijbehorende NO_x mitigerende strategieën. Verder zijn de verschillen tussen de NO_x-emissies van aardgas- en waterstofverbranding voor verschillende industriële hogetemperatuurbranders onderzocht. Met de informatie verkregen in fase 1 zijn nieuwe ontwerpregels voor lage NO_x waterstofbranders ontwikkeld in fase 2 van het project.

Uit de inventarisatie van de verschillende brandertechnologieën blijkt dat veel verschillende brandertypes worden gebruikt in industriële processen die elk zijn ontworpen voor een specifiek proces. Ter illustratie, branders zijn bijvoorbeeld ontworpen met het doel voornamelijk stralingswarmteoverdracht te genereren, terwijl andere zijn ontworpen om voornamelijk convectieve warmteoverdracht te genereren. In het algemeen kunnen de branders worden onderverdeeld in voorgemengde en niet-voorgemengde branders. Bij voorgemengde branders wordt de lucht en brandstof voorgemengd voor de brander en bij niet-voorgemengde branders vindt de menging pas na de brander plaats. Voorgemengde branders hebben bij overschakelen van aardgas naar waterstof als nadeel dat vlaminslag of oververhitting van de branderkop mogelijk kan plaatsvinden. Door de luchtvermaat te vergroten, kunnen deze problemen echter worden ondervangen met als bijkomend voordeel dat de NO_x-emissie wordt verminderd. Het merendeel van de branders die in de hogetemperatuurindustrie worden gebruikt zijn niet voorgemengde branders, ook wel diffusie- of nozzle mix branders genoemd. Uit de literatuurinventarisatie blijkt dat de diversiteit aan verschillende ontwerpen van niet voorgemengde branders groot is. Voorbeelden hiervan zijn swirlbranders, pijp-in-pijpbranders, FLOX-branders, regeneratieve branders, recuperatieve branders en stralingsbranders.

Toevoegen van waterstof aan aardgas leidt tot een hogere vlamtemperatuur, bredere ontvlambaarheidsgrenzen, snellere diffusie en hogere verbrandingssnelheid. Als gevolg hiervan komt de vlam bij toevoeging van waterstof dichter bij de branderkop te staan, wat leidt tot een lagere interne rookgasrecirculatie en (nog) hogere vlamtemperaturen. Voor het overgrote deel van de in de literatuur beschreven branders leidt waterstof toevoeging aan aardgas tot een verhoging van de NO_x-emissie. De NO_x-mitigerende maatregelen die geïdentificeerd zijn in de literatuur laten zien dat het verminderen van de verblijftijd, (lokale) temperatuur en de zuurstofconcentratie effectieve maatregelen zijn om de NO_x-emissie te verminderen tot het huidige of zelfs lagere niveau. Zo blijkt uit experimenten met een ventilator voorzet brander die is geplaatst in een industriële warmwaterboiler dat het overschakelen van aardgas naar waterstof resulteert in een drie keer hogere NO_x-emissie dan wettelijk toegestaan (70 mg/m³, 3% O₂). Door toepassing van externe rookgasrecirculatie is de NO_x-emissie bij gebruik van waterstof met succes teruggebracht tot ver onder de wettelijk toegestane NO_x-waarden. Voor branders die aanwezig zijn in hoge temperatuursprocessen is het echter nog een uitdaging om de NO_x-emissies drastisch te verlagen omdat de rookgastemperaturen vaak te hoog zijn voor externe rookgasrecirculatie en dus andere oplossingen gevonden moeten worden. Voor een aantal in de literatuur beschreven waterstofbrander ontwerpen wordt de NO_x emissie verlaagd door gebruik te maken van het creëren van voldoende interne rookgasrecirculatie door bijvoorbeeld de afstand tussen de brandstof- en luchtnozzles te vergroten, het aantal brandstof- en luchtnozzles te veranderen en de diameter van de brandstof- en luchtnozzles te veranderen. Andere branderontwerpen beschreven in de literatuur maken bijvoorbeeld gebruik van het Coanda-effect met

als doel het creëren van een onderdruk om meer rookgasrecirculatie te creëren. Andere technieken om tot lage NO_x emissie te komen zijn bijvoorbeeld FLOX verbranding, getrapte verbranding, waterinjectie en microverbranding. In de onderstaande figuur is een overzicht weergegeven van in de literatuur gevonden NO_x verlagende maatregelen.



Samenvattend blijkt uit de literatuurinventarisatie dat de wijze waarop de menging van waterstof, lucht en rookgassen plaatsvindt essentieel is om de NO_x-emissie te verminderen. Om een aantal algemene ontwerpregels voor branders te vinden, is het daarom belangrijk om een beter begrip te verkrijgen van de menging tussen waterstof, lucht en rookgassen. Hiertoe is in deze studie een jet-model ontwikkeld om de menging van waterstof, lucht en rookgassen te berekenen met verschillende branderontwerp parameters. De berekeningen uitgevoerd met het jet model zijn aangevuld met numerieke vlamberekeningen. De simulatieresultaten laten zien dat als gevolg van de bredere explosiegrenzen van waterstof en de hogere verbrandingssnelheid de verbranding veel dicht bij de branderkop plaatsvindt in vergelijking met methaan. Hierdoor ontstaan er hogere temperaturen dichtbij de branderkop en neemt de NO_x-vormingssnelheid toe bij het overschakelen van methaan naar waterstof. De numerieke vlamsimulaties laten zien dat interne rookgasrecirculatie en getrapte verbranding effectieve strategieën zijn om de NO_x te verminderen. Beide strategieën zijn gebaseerd op het verlagen van de verbrandingssnelheid en de vlamtemperaturen. De numerieke rookgasrecirculatie simulaties laten een goede overeenkomst zien met de gemeten waarden die nodig zijn om de NO_x-emissie terug te brengen tot die van methaanverbranding. Simulaties laten zien dat er ongeveer 16% rookgas in het H₂/luchtmengsel aanwezig moet zijn om bij een oventemperatuur van 1000 °C dezelfde NO_x-waarden te krijgen als voor methaan. Bovendien tonen simulaties aan dat verbranding onder brandstofrijke omstandigheden resulteert in lage NO_x-vorming. De berekende NO_x-vorming bij waterstof verbranding ligt in hetzelfde bereik als bij methaanverbranding in het geval dat de equivalentieverhouding hoger is dan $\phi=1.2$.

Op basis van de uitkomsten van deze studie zijn drie verschillende conceptuele branderontwerpen voorgesteld; 1) een pijp-in-pijpbrander, 2) een swirlbrander en 3) een getrapt verbrandingsontwerp. Alle drie de branderuitvoeringen zijn voorzien van een venturi om interne rookgasrecirculatie te bevorderen. De ontwerpparameters van de brander, zoals de diameter van de brandstof- en luchtinlaat, de diameter van de venturi en de luchtdruk die nodig is om voldoende rookgasrecirculatie en/of trapverbranding te creëren om de vorming van NO_x te onderdrukken, zijn berekend met behulp van het ontwikkelde jet-model.

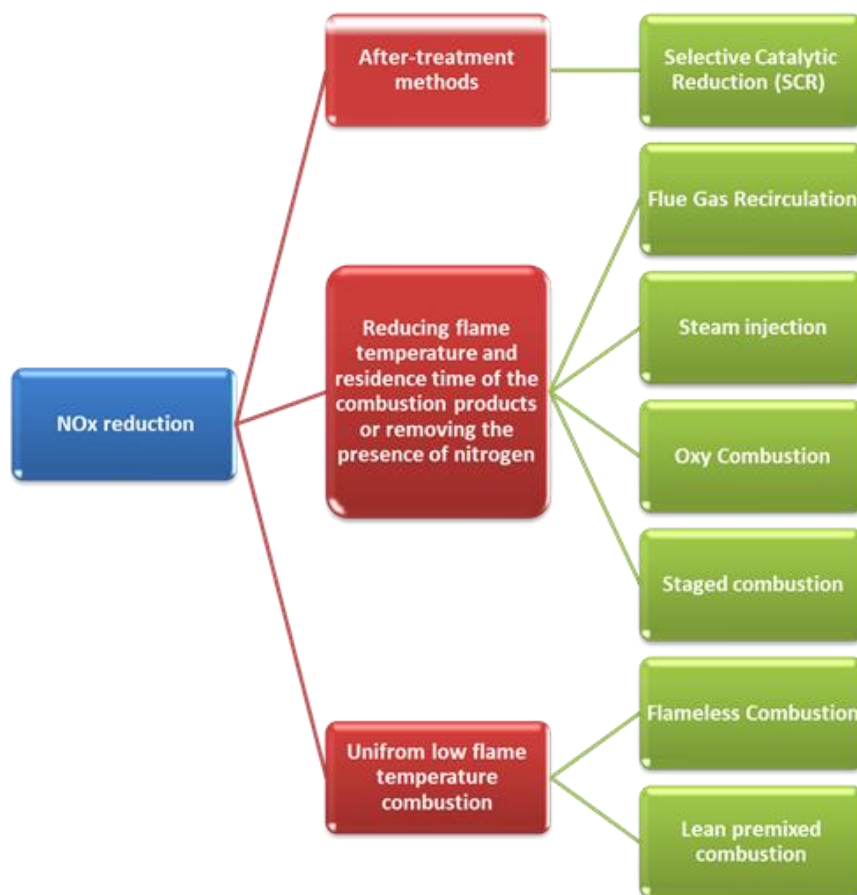
Het is aan te bevelen om de brander zodanig te construeren dat de branderkoppen flexibel verwisseld kunnen worden en om de brander te testen in een (semi)-industriële oven. Tijdens deze testen wordt het effect van verschillende branderkoppen en branderconfiguraties op de NO_x-emissie bestudeerd. De hierbij verkregen informatie zal worden gebruikt om de optimale configuratie te creëren voor de drie voorgestelde lage NO_x industriële branderontwerpen. Samen met de industrie en branderfabrikant(en) wordt één branderontwerp geselecteerd voor een veldtest bij één van de industriële eindgebruikers. Daarnaast zullen de test resultaten waardevolle inzichten opleveren voor het ontwerp van waterstofbranders voor diverse industriële toepassingen.

Summary

The energy-intensive industry is investigating the option to use hydrogen as a fuel to drastically reduce the carbon intensity of their manufacturing processes to meet the climate agreements. One of the challenges is to gain high efficient combustion while keeping the NO_x emissions low. Towards this end, this research is aimed to accelerate the development of LowNO_x hydrogen burners needed for the large-scale introduction of hydrogen in the high-temperature industry. The project is divided into two phases. In phase 1 a literature inventory was performed on the different burner types used in the industry and on the NO_x mitigating strategies that can be applied for these burner types. Furthermore, the differences between the NO_x emissions from natural gas and hydrogen combustion for various industrial high temperature burners were investigated. This information obtained in phase 1 was used for the development of new design rules for low NO_x hydrogen burners which was developed in phase 2 of the project.

The inventory on the different burner technologies reveal that many different burner types are used in industrial processes each designed for a specific process. For example, several burners are designed with the aim of generating mainly radiative heat transfer while other are designed to generate mainly convective heat transfer. Generally, the burners can be divided into premixed and non-premixed burners. In processes where the air and fuel are premixed prior entering the burner, switching to hydrogen can result in flashback and burner tip overheating. However, by increasing the combustion excess air, these issues can be overcome with the additional advantage that the NO_x emission will be reduced. Most burners used in the high temperature industry rely on the non-premixing concept referred to as diffusion or nozzle mix burners. The literature inventory show that a diversity of different designs of nozzle mix burners is large. Examples of nozzle mix concepts identified in the literature are swirl burners, pipe-in-pipe burners, FLOX burners, regenerative burners, recuperative burners and radiant heaters.

The literature inventory reveals the major challenges with blending hydrogen to natural gas (up to 100%) are higher flame temperature, wider flammability limits, faster diffusion and higher burning velocity. Altogether, upon hydrogen addition the flame moves closer to the burner tip causing lower internal flue gas recirculation and higher flame temperatures. As a result, it was found that for most of the burners tested, hydrogen addition to natural gas increases the NO_x emission. NO_x mitigating measures identified in the literature show that reducing the residence time, hot zones with high (local) temperature and oxygen concentration are effective measures to reduce the NO_x emission. For example, experiments performed on a forced draught burner present in an industrial boiler system show that switching from natural gas to hydrogen results in a three times higher NO_x emission than the legal NO_x limit allows. By applying external flue gas recirculation the NO_x emission, when using pure hydrogen, was successfully reduced below the Dutch legal NO_x limits (70 mg/m³, 3% O₂). However, for burners present in high temperature processes reducing the NO_x emissions is still a challenge to be solved, since the flue gas temperatures are often too high for external flue gas recirculation. Prototype hydrogen burner designs found in the literature aim to lower the NO_x emission by creating more internal flue gas recirculation by, for example, increasing the distance between the fuel and air nozzles, changing the number of fuel and air nozzles and changing the diameter of the fuel and air nozzles. Furthermore, several dedicated burner designs are making use of, for example, the Coanda effect with the aim of creating an under-pressure to create more flue gas entrainment. Other techniques identified in the literature used are mild combustion (FLOX), staged fuel combustion, water injection and micro combustion. The Figure below gives an overview of the NO_x mitigating measures identified in the literature.



The literature inventory reveals that the degree of mixing between hydrogen, air and flue gases is essential to reduce the NO_x emission. To find some general design rules for burners, it is important to have a better understanding of the mixing between hydrogen, air and flue gases. Towards this end a jet model was developed in this study to calculate the mixing of hydrogen, air and flue gases using different burner design parameters. The jet calculations are supplemented with numerical flame calculations. The simulation results reveal that due to the wider flammability range of hydrogen and faster burning velocity combustion takes place much closer to the burner head in comparison to methane and creating higher temperatures near the burner head. Consequently, the NO_x formation rate increases when switching from methane to hydrogen. Based on numerical flame simulations internal flue gas recirculation and staged combustion were found to be effective strategies in reducing NO_x. Both strategies are based on decreasing the burning velocity and flame temperatures. Comparison between the calculated amount of flue gas recirculation and measured values needed to reduce the NO_x emission back to that of methane combustion show excellent agreement. Simulations show that about 16% flue gas need to be present in the H₂/air mixture to get the same NO_x levels as that for methane for a furnace temperature of 1000 °C. Furthermore, simulations show that combustion under fuel rich conditions results in low NO_x formations, when the equivalence ratio is higher than $\phi=1.2$ the NO_x formation is in the same range as for methane combustion.

Based on the knowledge gained in the study three different conceptual burner designs are proposed; 1) a pipe-in-pipe burner, 2) a swirl burner and 3) a staged combustion design. All three burner designs are equipped with a venturi to create internal flue gas recirculation. The burner design parameters, such as the fuel- and air nozzle diameter, diameter of the venturi and air pressure

needed to create sufficient flue gas recirculation and/or staged combustion to suppress the NO_x formation were calculated using the jet model.

It is recommended to construct the burner in such a way that the burner heads can be flexibly exchanged and test the burner in a (semi)-industrial furnace. During these tests, the effect of different burner heads and burner configurations on the NO_x emission will be studied. The information acquired during these tests will be used to create the optimum configuration for the three proposed low NO_x industrial high-temperature hydrogen burner designs. Together with industry partner(s) and burner manufacture(s), one burner design will be selected for a field test. The test will also give valuable insights for the design of hydrogen burners for various industrial applications.

Content

| | |
|--|----|
| Document samenvatting..... | 2 |
| Samenvatting..... | 3 |
| Summary | 6 |
| Content..... | 9 |
| 1 Background..... | 11 |
| 2 Literature study on the implications of hydrogen in combustion use - NO _x effects | 12 |
| 2.1 Approach | 12 |
| 3 Physical properties of natural- and hydrogen gas..... | 13 |
| 3.1 Combustion properties..... | 13 |
| 3.2 NO _x formation | 17 |
| 3.3 References | 20 |
| 4 Overview of burner types and performance / Burner types and application area's | 21 |
| 4.1 Introduction..... | 21 |
| 4.2 Forced Draught Burners | 22 |
| 4.3 High-velocity burner..... | 24 |
| 4.4 Regenerative, recuperative and hot air burners | 27 |
| 4.4.1 Regenerative burners | 27 |
| 4.4.2 Hot air burners | 27 |
| 4.4.3 Recuperative burners | 30 |
| 4.5 Radiant IR burners..... | 30 |
| 4.6 Swirl burners..... | 31 |
| 4.7 FLOX or flameless oxidation burners..... | 33 |
| 4.8 Lance burners | 34 |
| 4.9 Flat flame burners | 35 |
| 4.10 Summary overview burner types / burner type and application area's | 36 |
| 4.11 References..... | 37 |
| 5 NO _x reduction methods..... | 38 |
| 5.1 (Partially) premixed combustion | 38 |
| 5.2 Non-premix combustion..... | 40 |
| 5.3 NO _x reducing strategies for gas turbines | 45 |
| 5.4 Oxy fuel combustion..... | 46 |
| 5.5 Post treatment NO _x reduction methods | 47 |
| 5.6 Summary strategies to reduce NO _x | 48 |

| | | |
|-------|---|----|
| 5.7 | References | 49 |
| 6 | Developing design rules for LowNO _x hydrogen high temperature burners..... | 51 |
| 6.1 | Effect flue gas recirculation and fuel staging on NO _x emission and temperature | 52 |
| 6.1.1 | Effect flue gas recirculation on the flame temperature and NO _x emission | 53 |
| 6.1.2 | Effect fuel staging on the flame temperature and NO _x emission | 55 |
| 6.1.3 | Conclusion and discussion:..... | 56 |
| 6.2 | Jet model | 58 |
| 6.2.1 | Brief description jet model | 58 |
| 6.3 | Proposed burner designs for low NO _x hydrogen HT burner | 61 |
| 6.4 | Burner design A | 62 |
| 6.4.1 | Effect changing fuel nozzle diameter on the hydrogen jet speed and jet composition distribution | 62 |
| 6.5 | Burner design B | 69 |
| 6.6 | Burner design C | 70 |
| 6.7 | Summary & outlook..... | 73 |
| 6.8 | References | 75 |

1 Background

One of the major challenges for the energy-intensive industry today is the drastic reduction in the carbon intensity of their manufacturing processes to meet climate agreements. A substantial part of the CO₂ emission from this industry is caused by burning fossil fuels for heating processes. A (sustainable) route to reduce the carbon intensity for industrial heating processes is to replace natural gas with renewable hydrogen.

The major advantage to use hydrogen as a fuel for heating processes is that the kilns, boilers and furnaces used for product manufacture and treatment themselves do not need to be changed drastically when switching fuels, resulting in only limited investment costs (CAPEX). In contrast, the alternative of electric heating often requires a complete redesign of the heating part of the production process and is often not technically feasible, since interaction with combustion products (reducing/oxidizing atmosphere) is needed for production. In summary, the major advantage of using hydrogen in (direct) heating processes is that ovens and furnaces do not drastically have to be adapted resulting in low investment costs as compared to the all-electric option.

The challenge for the transition of direct and indirect heating processes to a carbon-free, hydrogen-fueled, future is to maintain (or improve) product quality, optimize efficiency and keep the NO_x emissions within the prescribed limits for the range of fuel compositions encountered in the transition: from natural gas to pure hydrogen. To meet the challenge, tailoring heat transfer (including footprint), the composition of the kiln atmosphere and NO_x mitigating strategies, for the different product classes, all with a well-functioning burner system, must be achieved. As there is a need for experimental data and hydrogen equipment designs on all these aspects, applied research is necessary to facilitate this transition.

2 Literature study on the implications of hydrogen in combustion use - NO_x effects

An ambition of the HyDelta consortium is to accelerate the development of Low NO_x hydrogen burners needed for the large-scale introduction of hydrogen in the high-temperature industry. This project maps successful applied NO_x-reducing strategies and sets the basis for proper design rules for hydrogen burners applied in high-temperature processes. To reach this goal, the project is divided into two tasks: literature research for low NO_x strategies for H₂ burners, and the development of design rules for LowNO_x hydrogen burners used in high-temperature processes. In this report the results of the literature study are presented.

2.1 Approach

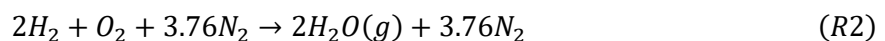
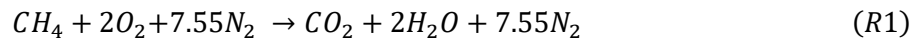
To prevent duplication of existing research a technical inventory is made. The points below are presented and addressed based on a literature study and knowledge available. In addition to hydrogen burner technology, the inventory also focuses on natural gas fueled burners so that the developed knowledge on NO_x reducing measures for natural gas burners can also be included to develop design rules for low NO_x hydrogen burners. The literature study includes the following activities:

- Inventory of different burner technologies used in the high-temperature industry
- Determining the fundamental combustion and flow properties of natural gas and hydrogen. How is NO_x produced when natural gas and hydrogen are burned?
- Investigating the differences between the NO_x emissions from natural gas and hydrogen combustion for various industrial high-temperature burners. In addition, what causes these differences in NO_x emissions?
- Providing an overview of successfully applied NO_x-reducing measures for natural gas burners and, if available, NO_x-reducing measures for hydrogen burners for high-temperature processes. This includes internal flue gas recirculation, changes in the flame surface, reduction of the residence time of the hot zones of the flame, etc.

3 Physical properties of natural- and hydrogen gas

3.1 Combustion properties

Natural gas and hydrogen have different chemical and physical properties and therefore show different combustion behavior. For example, upon burning natural gas, the main products produced by combustion are CO₂ and H₂O (R1) while, with hydrogen combustion the main combustion product will be H₂O (R2). Also, as can be seen from R1 two molecules of oxygen are needed to complete the combustion of methane while R2 shows that only ½ of a molecule of oxygen is needed for hydrogen combustion.



In reality, the combustion does not occur via a single reaction but via a sequential process involving many reactive intermediate species. As a result, combustion pollutant emissions such as NO and NO₂ can be formed during combustion or when incomplete or 'unstable' combustion takes place components such as C_xH_y, CO, and H₂ can be present in the flue gases. Therefore, developers of combustion systems take into account the combustion behavior of the fuel blend when designing equipment such as burner systems. The challenge for burner designers is to optimize the mixing between fuel and air to maximize heat transfer while at the same time minimizing the CO and NO_x formations in the flame. One of the parameters that designers of combustion equipment use to optimize their systems is the fuel-to-air ratio. The fuel-to-air ratio at which combustion takes place is expressed by the equivalence ratio:

$$\phi = \frac{\text{Fuel}}{\text{Air}} \cdot \frac{1}{f_{st}} \quad (1)$$

Where f_{st} is the ratio of fuel and air under stoichiometric conditions. If there is an excess of combustion air available in the unburned fuel/air mixture, the mixture is fuel-lean ($\phi < 1$), while the mixture is called fuel rich ($\phi > 1$) when an excess of fuel is present. A mixture is called stoichiometric ($\phi = 1$) when the fuel-to-air ratio is such that all fuel and oxygen are burned as shown in the overall reactions R1 and R2 (reactions are balanced).

Industrial burners can generally be divided into two combustion modes 1) premixed and 2) non-premixed. In premixed burner systems, the fuel and air are premixed before entering the burner tip while for non-premixed systems the mixing of fuel and air takes place downstream of the burner. To design new burner concepts for hydrogen, the difference between the combustion properties of natural gas and hydrogen needs to be taken into account. As an illustration, a selection of the physical- and chemical combustion properties of methane and hydrogen (at stoichiometric conditions) are present in Table 3.1.

Table 3.1: Combustion properties of methane/hydrogen mixtures at $\phi = 1$ ($\lambda = 1$)

| CH ₄ (mole %) | H ₂ (mole %) | W ⁶ , MJ/m ³ | H _i ¹ , MJ/m ³ | H _s ² , MJ/m ³ | ρ, kg/m ³ | S _L , cm/s | T _{adiabatic} , K | LFL ¹ (vol.% in air) | UFL ² (vol.% in air) | Eignition, min, mJ |
|-----------------------------|----------------------------|---------------------------------------|--|--|-------------------------|--------------------------|----------------------------|------------------------------------|------------------------------------|--------------------|
| 100 | 0 | 53.5 | 35.9 | 39.8 | 0.717 | 36.6 | 2228 | 4.99 | 14.73 | 0.24 |
| 0 | 100 | 48.3 | 10.7 | 12.8 | 0.090 | 252.0 | 2384 | 4.07 | 74.24 | 0.03 |

¹ Net calorific value

² Gross calorific value

From Table 3.1 it can be seen that switching to hydrogen combustion increases the adiabatic flame temperature from 2228 to 2384K, which can result in an increase in the NO_x emission. Furthermore, the increase in laminar burning velocity (S_L) and the widening of the flammability limits (LFL³ and UEL⁴) when switching to hydrogen combustion can result in flash-back in premixed burner systems or a shift of the hot combustion zone of the flame closer to the burner tip for non-premixed (diffusion) burners which also enhances the NO_x formation. As shown in Figure 3.1 lowering the equivalence ratio changes the laminar combustion velocity substantially. Therefore, one of the strategies to prevent flashback or burner overheating used by burner designers of combustion systems is to decrease the equivalence ratio of fuel/air mixture.

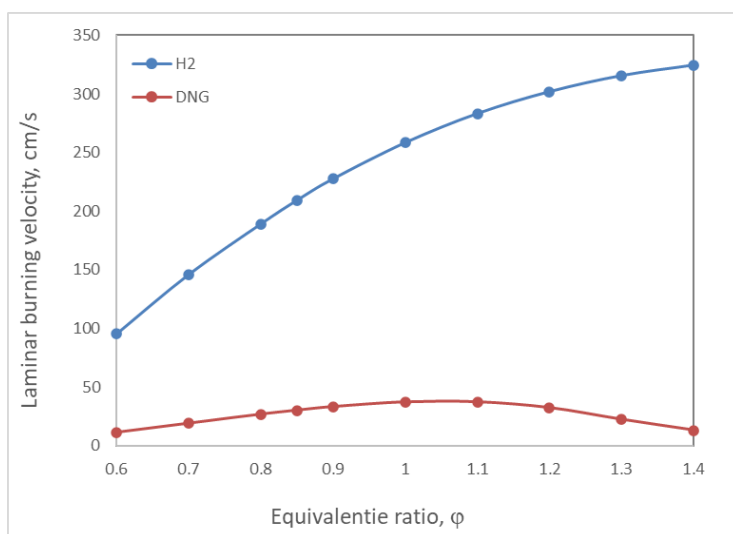


Figure 3.1: Numerically calculated Laminar burning velocities of Dutch natural gas (DNG) and hydrogen. for different equivalence ratios⁵ for hydrogen and Dutch Natural Gas (DNG⁶).

What changes to expect in the fuel (and) airflow when switching from natural gas and hydrogen combustion?

To gain insight into what extent the changes in the physical properties such as density and calorific value affect the burner power, fuel, air velocity and momentum flux when using hydrogen in a natural gas-designed burner system, a brief analysis has been conducted. Towards this, we use as an illustration that hydrogen is used in a natural gas burner system while keeping general parameters constant. When maintaining a constant fuel back pressure and a constant pressure drop across the fuel nozzle(s), the (volumetric) flow rate increases when switching from natural gas to hydrogen since the density decreases by about a factor of nine. As a result, according equation (2) the fuel velocity increases by a factor of three as shown in Figure 3.2. Here it should be noted that in equation (2) the bold assumption is made that the discharge coefficient of hydrogen and methane are identical.

³ LFL: Lower Explosion Limit

⁴ UFL: Upper Explosion Limit

⁵ Laminar burning velocities were calculated using the PREMIX program [2] from the CHEMKIN II package [1] and the USC MECH II chemical mechanism and thermodynamic data base [3]

⁶ Dutch Natural Gas (DNG), natural gas consisting of roughly 82.0 mole% CH₄, 2.7 mole% C₂H₆, 0.4 mole% C₃H₈, 0.9 mole% CO₂ and 14.0 mole% N₂

$$Q_v \sim \sqrt{\frac{2\Delta P}{\rho_{fuel}}} \quad (2)$$

Hence, although not shown when keeping the airflow constant when switching to hydrogen, the equivalence ratio increases (increases in the excess of combustion air). To keep a constant equivalence ratio the airflow needs to be decreased as shown in Figure 3.2.

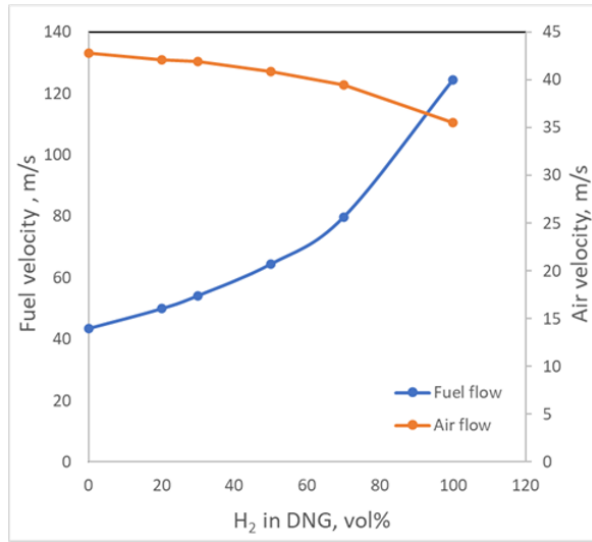


Figure 3.2. Impact of hydrogen addition to Dutch natural gas on the fuel velocity and air velocity at fixed fuel-to-air ratio.

The calorific value (H_s) and fuel flow (Q_v) determine the thermal load to the burner

$$Thermal\ burner\ load \sim Q_v H_s \quad (3)$$

Combining equations (2) and (3) at a constant back pressure of the fuel supply gives the following relation, which is 'similar' to the Wobbe index⁷;

$$Thermal\ burner\ load \sim \frac{H_s}{\sqrt{\rho_{fuel}}} \quad (4)$$

From equation 2 and Table 3.1 can be seen that hydrogen addition to natural gas causes a change in the gas flow rate (Q_v) and the calorific value of the fuel blend (H_s). Consequently, it results in a change in the thermal burner power as shown in Figure 3.3. However, switching to pure hydrogen combustion only has a minor effect on the burner load which suggests that (based on (only) the burner load) the same nozzle diameter in burners can be used for both natural gas and hydrogen combustion. If needed the fuel pressure can be adjusted by slightly closing the fuel valve to maintain a constant burner power.

⁷ Wobbe index = $\frac{H_s}{\sqrt{\rho_{relative\ density}}}$

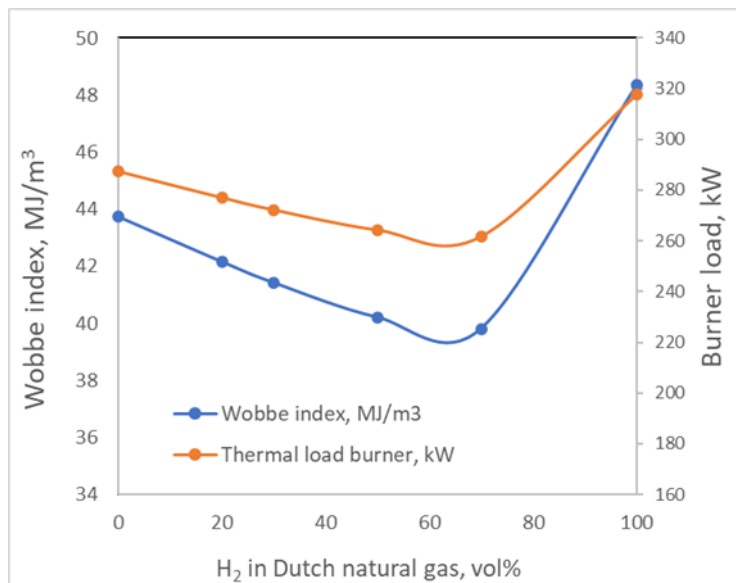


Figure 3.3: Changes in the Wobbe index and the burner power when adding hydrogen to natural gas at constant pressure drop across the fuel nozzle.

As discussed and shown in Figure 3.2 the fuel velocity increases upon hydrogen addition and the air velocity reduces when the air factor and thermal input are constant. The changes in the flow velocities result in a change of the mixing between fuel and air. More specifically, the mixing of the fuel jet with air and flue gases depends upon the difference in impulse momentum flux between the fuel and air jets and the surrounding. The momentum flux is defined as;

$$\Phi_{flux} = \rho \times v^2 \quad (5)$$

When adding hydrogen to natural gas, the momentum flux changes as a result of the change in density (ρ) as presented in Table 1, and the change in velocity (v) as shown in Figure 3.2.

As seen in Figure 3.4, the large increase in fuel jet velocity and the decrease in density results in little major changes in momentum flux between natural gas and hydrogen. This suggests that no large differences are expected between the mixing of the fuel, air, and flue gases in the 'cold' jets. However, due to the difference in the upper flammability limits (hydrogen needs 0.5 mole O₂ and CH₄ needs 2 moles O₂ for complete combustion), it is expected that hydrogen will burn faster, bringing the hot zone closer to the burner tip which can result in overheating and less internal flue gas recirculation and, consequently, for example, higher local flame temperatures.

To what extent these changes in physical and chemical properties between natural gas and hydrogen affect burner performance is investigated in this study by performing a literature inventory. The results of the literature study form the basis for the development of a new hydrogen burner system.

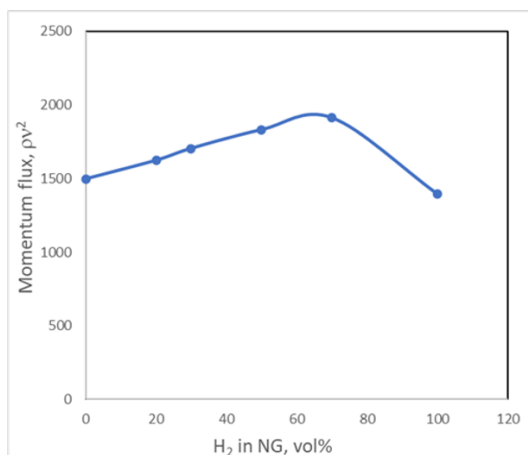


Figure 3.4: Effect of hydrogen blending to the momentum flux of the fuel jet

3.2 NO_x formation

NO is the primary nitrogen oxide formed in combustion processes and the precursor for the formation of other nitrogen oxides such as NO₂. The two main mechanisms of NO formation in flames are Zeldovich (thermal) NO formation and Fenimore (prompt) NO formation. As can be seen in the Figure below, the Zeldovich mechanism is dominant at fuel-lean and stoichiometric conditions, while the Fenimore mechanism is dominant at fuel-lean conditions.

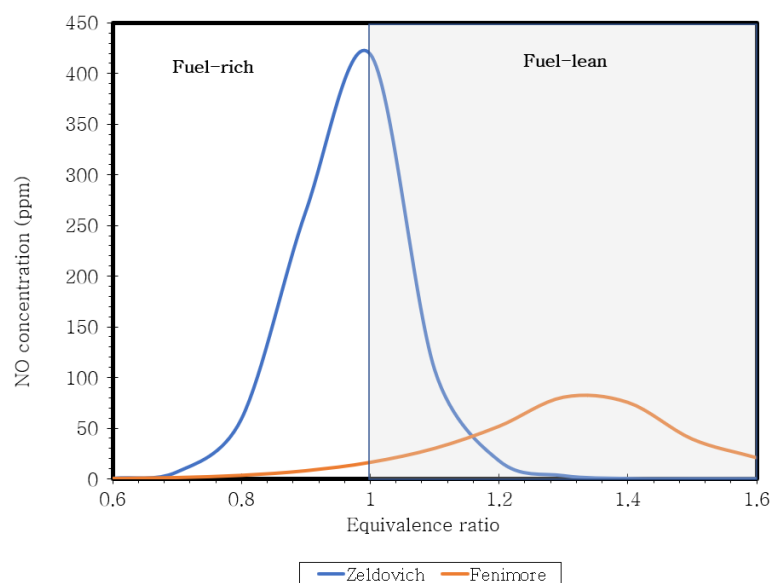
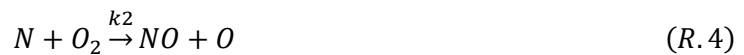
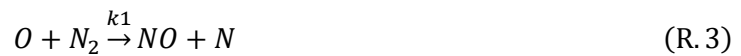


Figure 3.5: Calculated Zeldovich and Fenimore NO mole fractions as function of equivalence ratio. The calculations were performed using the Premix code [2] of the CHEMKIN II package [1] and GRI Mech 3.0 mechanism [4].

The principal reactions governing the formation of NO in the (extended) Zeldovich mechanism are:



Reaction (R.3) is considered to be the rate-determining step due to its high activation energy (needed to break open the triple nitrogen bond), which allows the reaction only to proceed efficiently at a relatively high temperature (typically above 1800K, hence the name “thermal NO formation”). The formation of NO via the Zeldovich mechanism is highly temperature dependent as illustrated in the Figure below. Since the flame temperature of hydrogen combustion is approximately 150 °C higher than natural gas combustion (at the same stoichiometry), NO_x formation will increase when switching from natural gas to hydrogen.

Assuming the initial NO concentration is low and that the atomic nitrogen concentration is at steady state, the following expression can be derived, which describes the overall NO formation via the Zeldovich mechanism:

$$\frac{d[NO]}{dt} = 2k_1(T)[O][N_2] \quad (R.6)$$

Where, $k_1(T)$ is the reaction rate constant for reaction R.3, and the brackets $[]$ denote the concentrations. In combustion processes where the air is used as the oxidant, the nitrogen concentration does not usually vary substantially, and the NO formation rate depends on the temperature and the atomic oxygen concentration as shown in equation R.6. Most NO_x mitigating measures are therefore aimed at lowering the temperatures and/or the oxygen concentration, e.g. by diluting the fuel-air mixture with inerts (flue gas recirculation) or upstream heat losses to reduce the flame temperature (i.e. the operating principle of a radiant burner).

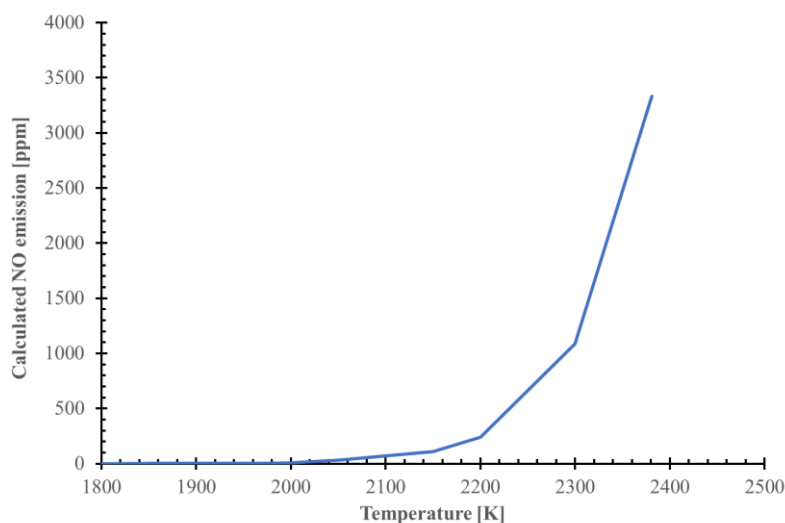


Figure 3.6: Calculated NO emission as function of temperature for a premixed adiabatic hydrogen/air mixture at stoichiometric conditions.

As can be seen in Figure 3.7 by adjusting the equivalence ratio towards fuel-lean conditions, the flame temperature drops and hence (see Figure 3.6) the NO formed will be reduced. At stoichiometric and fuel-rich conditions the temperature remains relatively high, which means that hotspots and fuel-rich zones should be avoided in order to reduce the (local) NO_x formation.

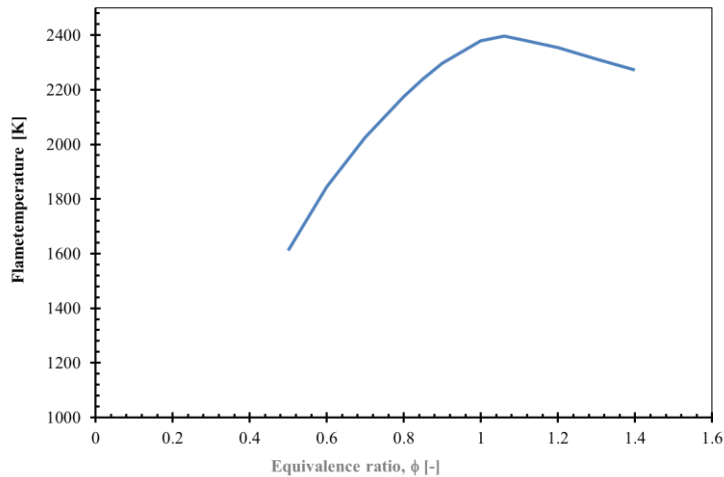


Figure 3.7: Calculated flame temperature as a function of the equivalence ratio for a premixed adiabatic hydrogen/air mixture at stoichiometric conditions.

As shown in Figure 3.5, the NO formation in fuel-rich flames proceeds primarily via the Fenimore mechanism, where NO is formed in the flame front via the following rate-determining step:



Both hydrogen cyanide and atomic nitrogen produced in this reaction will react to NO via a series of subsequent reactions. However, since carbon (CH) is needed for the reaction to proceed, the Fenimore mechanism is not relevant for pure hydrogen flames.

3.3 References

- [1] Kee, R.J., F.M. Rupley, J.A. Miller, "CHEMKIN II: A Fortran Chemical Kinetics Package for the Analysis of Gas-Phase Chemical Kinetics", Sandia National Laboratories, (1989).
- [2] Kee, R. J., J. F. Grcar, M. D. Smooke, J. A. Miller, Fortran program for modelling steady one-dimensional premixed flames. Sandia Report SAND85-8240. Sandia National Laboratories, 1985.
- [3] Wang, H., You, X., Joshi, A.V., Davis, S.G., Laskin, A., Egolfopoulos, F., Law, C.K., USC Mech Version II. "High-Temperature Combustion Reaction Model of H₂/CO/C₁-C₄ Compounds", http://ignis.usc.edu/USC_Mech_II.htm, May 2007
- [4] Smith, G. P., Golden, D. M., Frenklach, M., Moriarty, N. W., Eiteneer, B., Goldenberg, W., Bowman, C. T., Hanson, R., Gardiner, W. C, Lissianski, V., and Qin, Z., http://www.me.berkeley.edu/gri_mech/.

4 Overview of burner types and performance / Burner types and application area's

4.1 Introduction

Heating processes can be categorized in **direct**- and **indirect** heating. During direct heating, there is direct contact between the flame and the substance that has to be heated. Direct heating is used in industrial heating processes such as melting glass and baking ceramics. With indirect heating the flame does not come in contact with the substance to be heated. Instead, the heating energy is indirectly transferred to the product through a medium (like steam or hot water) in the form of conduction, convection, or radiation.

The performance of burners changes when hydrogen is used instead of natural gas. This change strongly depends upon the type of burner installed. There are two main types of burners, mentioned in chapter 3: (partially) premixed –, and non-premixed (diffusion) burners.

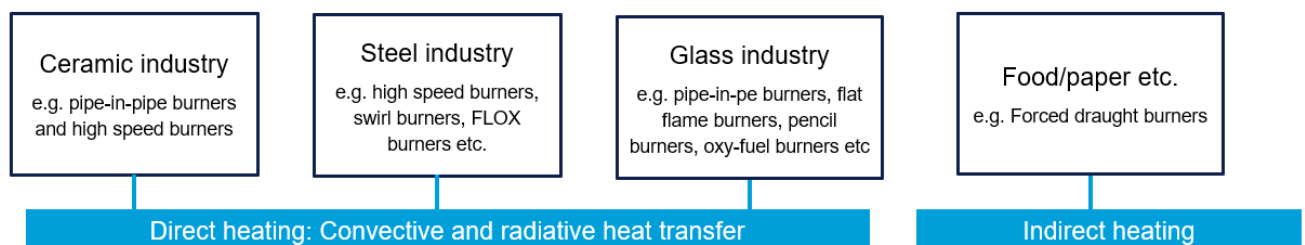


Figure 4.1: Many different burner types are used in the industry.

The (partially) premix burners blend the fuel and combustion air before entering the burner and before the ignition, as shown in Figure 4.2 below. Typical premix burner systems are modern domestic central heating boilers, pencil burners used in the forehearth of a glass melting bath, and burners present in dry low NO_x gas turbines. The main issue when switching to hydrogen combustion for premixed burners is the risk of overheating the burner tip and/or the occurrence of flashback as illustrated in Figure 4.3.

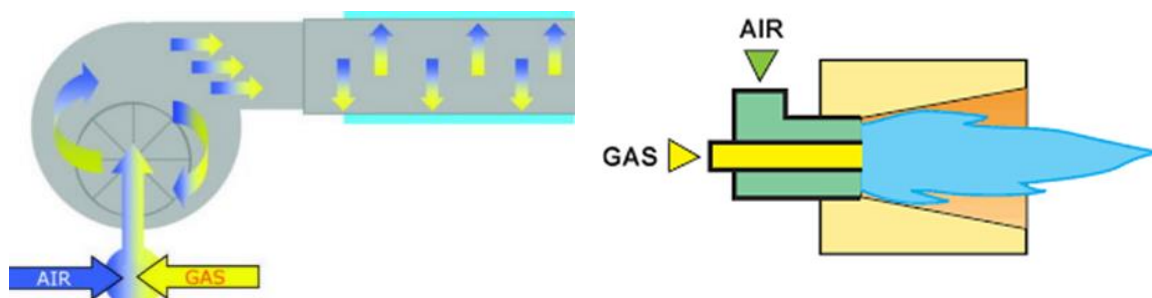


Figure 4.2. Left, fully premixed burner (air and natural gas are mixed before combustion [1]). Right: Nozzle mix burner (air and natural gas are mixed during combustion [2])

The flash-back in premixed burners can be prevented by increasing the fuel/air velocity and increasing the excess of air to lower the burning velocity, as illustrated in Figure 3.1. The advantage of increasing the air excess is the reduction of NO_x in premixed combustion systems, as shown in Figures 3.6 and 3.7. However, increasing the excess of air results in a decrease in thermal efficiency, which can be substantial for high-temperature processes.

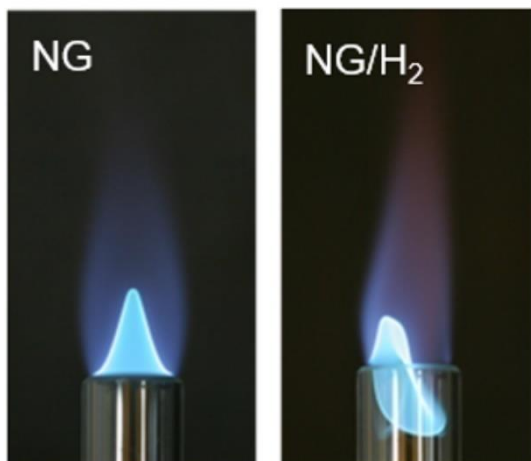


Figure 4.3: Example of flashback upon hydrogen addition to natural gas.

A burner is considered a non-premixed or nozzle mix (diffusion) burner when the fuel and combustion air are separately supplied at the burner tip and mix further downstream the burner tip. Fuel is injected via a fuel nozzle (or multiple nozzles) and the combustion air is supplied via natural- or forced draught feeding system, often using co-flow, swirl flow, etc. The advantage of nozzle mix burner systems is that they do not suffer from a potential flashback since there is no oxidizer inside the fuel nozzle. Nonetheless also the burner nozzles, burner block and refractory material may suffer from excessive temperatures if inadequately corrected for the presence of H₂ in the fuel blend. Nozzle mix burners work at different air/fuel ratios and their flame shape heavily depends on the nozzle diameter, supply angle, fuel and air back pressure, and the way the combustion air is supplied (e.g. via swirl, co-flow, or radial flow).

There are many types of nozzle burner configurations, each designed for their specific purpose to provide optimal heating of the product or load. The heating occurs via radiation and/or convection depending on the heated substance and the production process. Different burner configurations are designed to provide optimal heating of the load. The burner design depends also on the type of heating needed for the desired process.

For example, melting glass is most effective when using radiant heating. Therefore, the burners are designed to produce highly radiant flames. If the process requires more convective heat transfer instead of radiant heat transfer (e.g. scrap metal, piles, bricks, water, oil, etc..) then the burner systems are designed such that the flame and combustion products produce dominantly (forced) convective heat transfer. Some common burner configurations used in the industry are described and shown in this chapter. Additionally, if available, we will also include the effect of hydrogen on the performance of the burner types described below.

4.2 Forced Draught Burners

Forced draught burners are primarily used for low-temperature processes, but are also occasionally used in medium and high-temperature processes. The process temperature of these nozzle mix burners varies between 50 and 1300 °C. Industrial low NO_x forced draught burners are used for example in asphalt production, drying processes, thermal oil heating, steam, hot water production, heat treatment such as the high temperature steel treatment processes, or medium temperature drying processes.

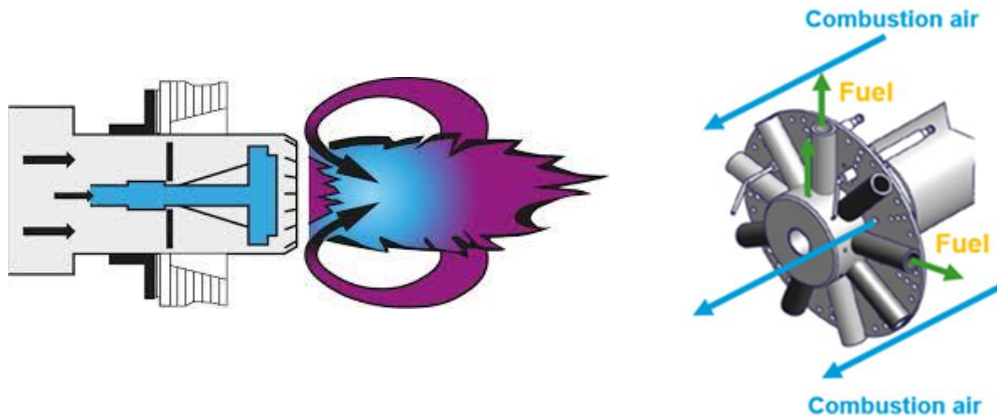


Figure 4.4: Working principle of forced draught burner

To create constant combustion within the boiler, the forced draft fan forces air into the burner. A part of the flue gases is (internally) recycled back by the under pressure created near the burner, due to the venturi effect of the high velocity of the gasses entering the combustion chamber. In the center of the burner plate, a 'pilot' flame burns that keeps the flame burning as shown in Figure 4.4.

At DNV [3] tests have been performed with a 500 kW forced draught burner in an industrial boiler system. The measurement results presented in Figure 4.5 show that the NO_x percentage measured increases significantly when adding hydrogen to natural gas. As an example, switching from natural gas to pure hydrogen results in an increase in the NO_x emission by about a factor of three; from ~70 to 200 mg/m³ (at 3% O₂ end point concentration). Within this burner, the fuel jets are radially mixed with the airflow downstream of the burner head (see also Figure 4.4). This creates rapid mixing between fuel and air. The faster combustion and the higher adiabatic flame temperature of hydrogen relative to that of natural gas results in a faster NO_x production mainly via the thermal NO_x mechanism as described above.

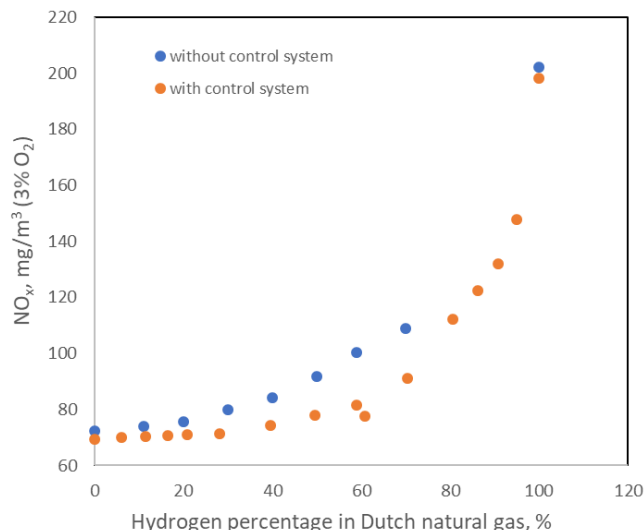


Figure 4.5: Effect of hydrogen blending to natural gas on the NO_x emission of a forced draught burner at constant thermal burner input [3]

4.3 High-velocity burner

High-velocity burners are typically used in the metal and ceramic industry to melt metal and to produce piles and brick from clay. The high flue gas velocities (~ 200 m/s) heat the product through forced convection. In the ceramic industry, high-velocity burners are dominantly used in the 'cold' part of the tunnel oven (700°C) and in the hot section mainly lance burners are used. High-velocity jet entrains (via momentum) surrounding air and flue gases [4], see also Figure 4.6. The flue gas cools the flame resulting in lower NO_x emission and homogeneous heat distribution.

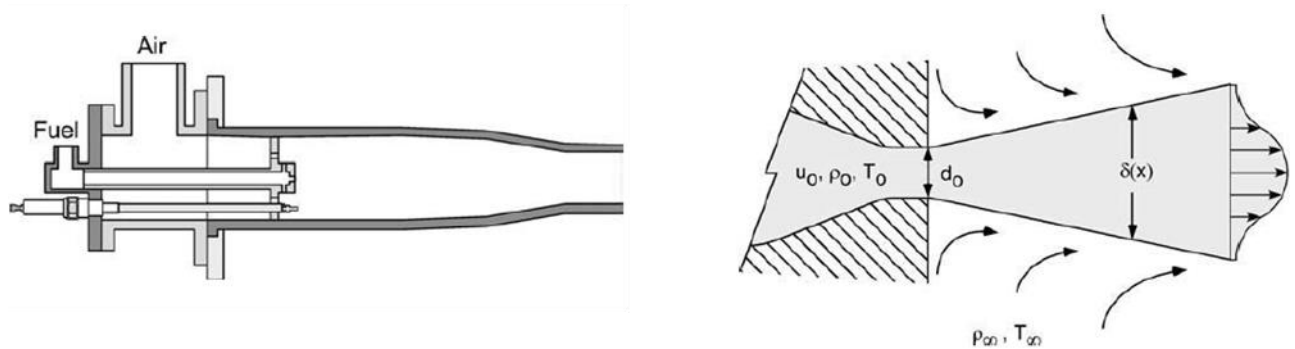


Figure 4.6: Schematic overview of a high-velocity burner (left) and a schematic overview of the entrainment of air and flue gasses into the jet (right).

There are currently a number of developments to reduce NO_x emissions in high-velocity heat burners. Two examples are the North American HiRAM burner and the North American Tempest™ burners [5] which are used in high-temperature applications such as forge furnaces, steel reheaters, and aluminum smelters. This burner design relies on Low NO_x Injection (LNI) technology which operates as a nozzle mix burner when the furnace temperature is below 760°C . When the temperature rises above 760°C beyond which the mixture is above the autoignition temperature, the fuel is switched from the center gas tube to strategically positioned nozzles adjacent to the burner tile port [5, 6]. Fuel from the nozzles and air from the burner port mix with the furnace gases (flue gases) and dilute the fuel/air mixture before combustion occurs. This causes a reduction in the flame temperature and the oxygen concentration, which reduces the NO_x emission (see equations R3 and R4).

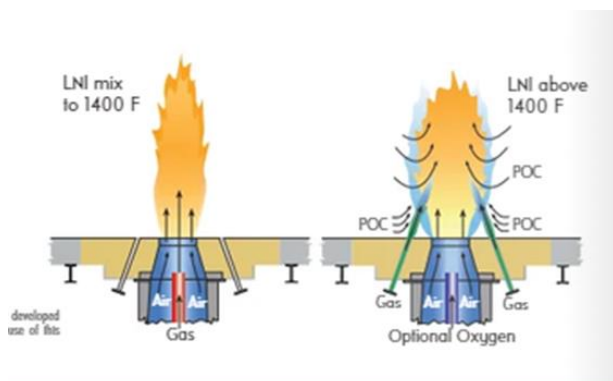


Figure 4.7: operating principle of LNI technology.

In a recent study, DNV investigated the effect of hydrogen addition to natural gas on the NO_x emission of a high-velocity burner (see Fig 4.8). The results in Figure 4.9 show that for the natural gas and the hydrogen (100%) cases, roughly the same NO_x emissions (3% O₂) were measured. However,

for mixtures of hydrogen and natural gas, up to 60% hydrogen in natural gas, the NO_x emission increases (from 23 to 35 g/GJ). Adding more hydrogen results in a decrease in NO_x emission as observed in Figure 4.9. The NO_x emission upon hydrogen addition differs from the behavior for other burners (see for example forced draught burner), where an increase in NO_x emission is observed upon hydrogen addition. The reduction in residence time, the degree of mixing between the fuel jet, combustion air, and the flue gases, and the higher diffusion rate of hydrogen plays an important role in the observed NO_x behavior.



Figure 4.8: High-speed velocity burner tested at DNV; flame image (left) and schematic of the burner (right) [7].

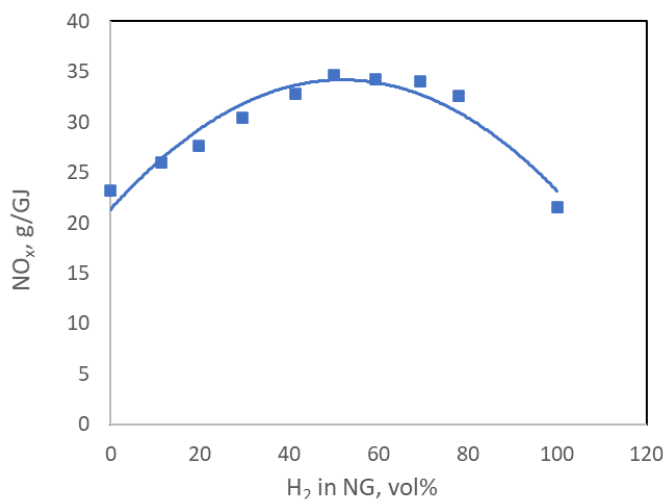


Figure 4.9: Effect of hydrogen addition to natural gas on the measured NO_x emission [7]

In ref [17] the effect of hydrogen addition (up to 50%) to natural gas for a 150 kW high velocity burner was experimentally investigated. The burner was fired in a unloaded kiln at constant energy input (firing rates). As can be seen in Figure 4.10, the addition of hydrogen to natural gas increases the NO_x emission similar in trend and values as shown in Figure 4.9. Additionally, hydrogen addition

to natural gas was found to lower the CO and CH₄ emissions probably caused by the flame moving closer to the burner tip upon hydrogen addition resulting in a lower amount of methane escaping the flame (and converting into CO due to incomplete combustion). The addition of hydrogen was not found to increase the burner tip (5 K temperature increase at 50 vol% hydrogen addition).

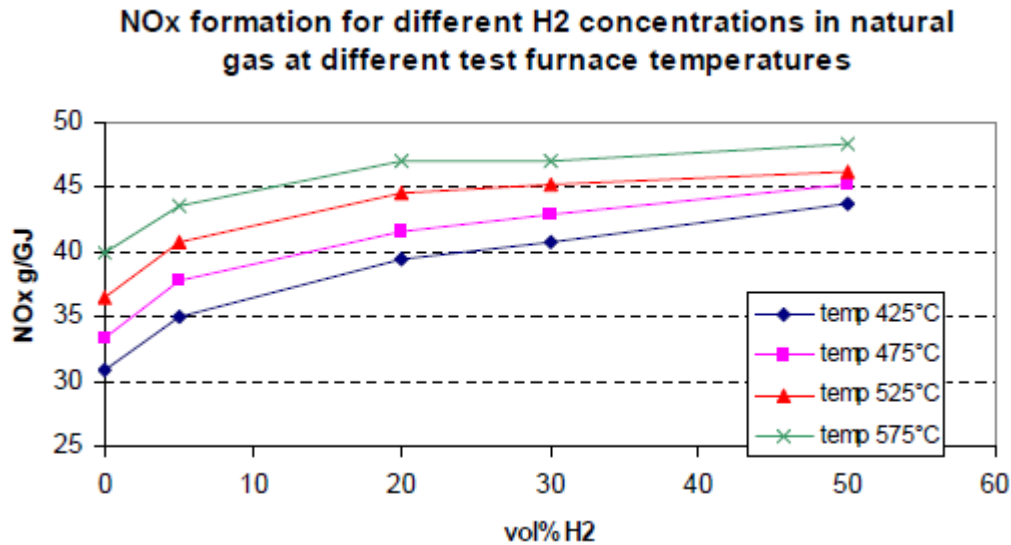


Figure 4.10. Measured NO_x emission for different volume percentages hydrogen in natural gas at different furnace temperatures.

4.4 Regenerative, recuperative and hot air burners

4.4.1 Regenerative burners

Regenerative burners recover the waste heat of a furnace's exhaust gas to pre-heat the combustion air of the furnace. The regenerative burners use heat reservoirs and dual heat-recovering generators at each burner. A regenerative burner system consists of (at least) two burners. The operating principle is as follows (see also Figure 4.10): when one burner is burning, the exhaust gas from that burner passes through and heats up the heat reservoir of the second burner that is turned off. Next, the burners switch, the combustion air of the second burner passes through the heat reservoir and is preheated. In this way, the energy from the exhaust gases is recovered resulting in significant energy savings. The burners are ignited at intervals of several seconds.

Regenerative burners are often used for glass melting where very high temperatures are needed. The flames are 'slowly' propagating highly radiating flames. Several attempts have been performed to lower the emission such as given in [8], where a >90% NO_x reduction is reported using preheated air and by separate and direct injection of fuel and air at high momentum (creating internal flue gas recirculation).

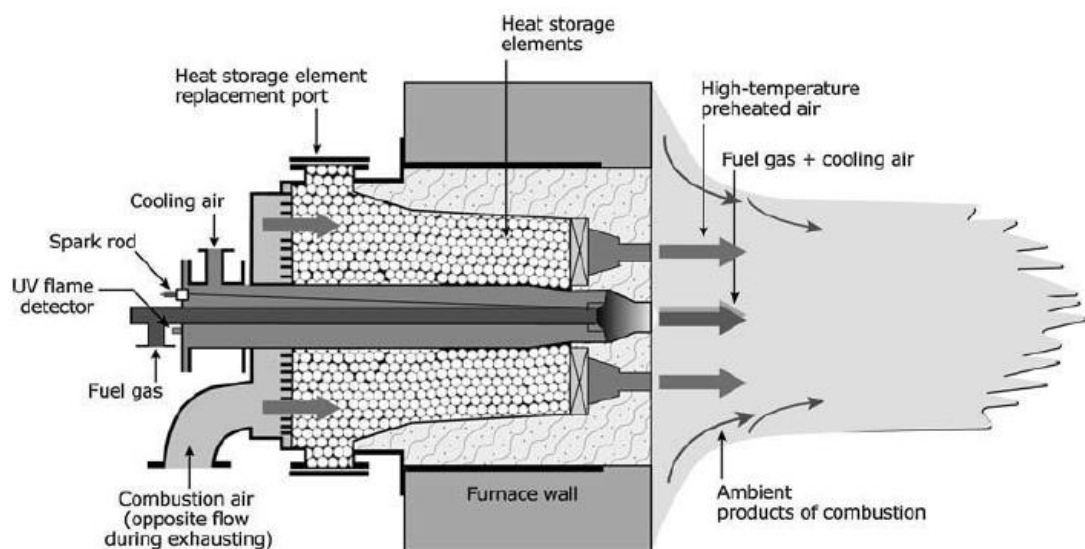


Figure 4.10: a schematic overview of a regenerative burner system [4].

4.4.2 Hot air burners

Hot air burners are used in for example the glass industry in glass melting furnaces where high temperatures are needed to keep the glass melted. To generate a high-temperature environment, the combustion air is externally preheated and mixed downstream of the burner with the fuel ('pipe-in-pipe', see also Figure 4.11a below).

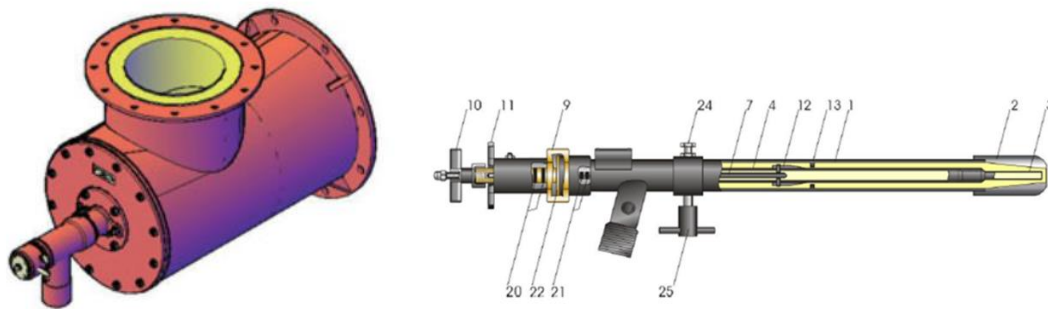


Figure 4.11a: Examples of a hot air burner, left [9] and right [10]

Due to the high temperature of the combustion air needed for glass melting, flames from hot air burners usually have a high temperature and hence a high NO_x emission (see also above). By injecting the air at a certain angle on top of the fuel jet (under port firing see burner in figure 4.11a on the right hand side), the high momentum of the air enhances the self-induced flue gas recirculation, which results in low thermal NO_x formation despite the high air temperature [9]. At GWI [11] NO_x measurements have been performed in the flue gases of a glass melt furnace at different hydrogen blends in natural gas as shown in Figure 4.11b. The results show that the NO_x emission increases from about 400 to 650 ppm NO_x emission when switching from natural gas to hydrogen combustion.

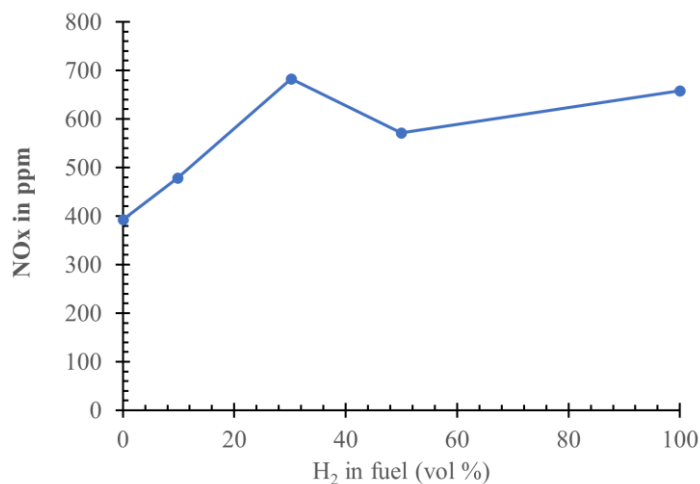


Figure 4.11b: NO_x emission measured using under-port firing in a glass melt [11]

Another strategy is to supply the air in co-flow ('pipe-in-pipe' burner) and allow an adjustment of the lance position and/or adjustment of the fuel jet impulse (momentum flux) as shown for the burner in Figure 4.11 on the left hand side. Some designs (as shown earlier) use dual fuel jets to optimize combustion and lower NO_x emissions. Optimal combustion, and thus optimal mixing between fuel and air is needed since when a reducing flame (incomplete combustion) comes into contact with molten glass this will result in degradation of the product quality.

DNV has measured the NO_x emission at different hydrogen concentrations in natural gas for a hot air burner. The results shown in Figure 4.12 below, show an (exponential) increase in NO_x emission upon hydrogen addition.

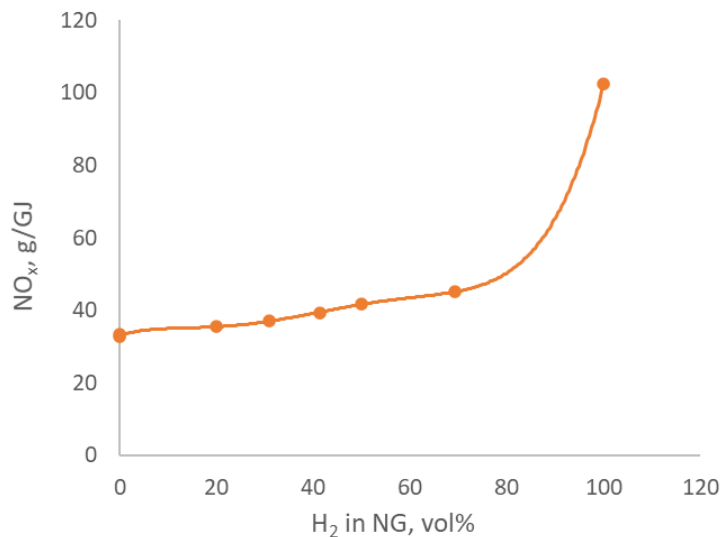


Figure 4.12: measured NO_x emission as a function of the hydrogen in natural gas for a hot air burner [7]

Figure 4.13 shows infrared images taken during the experiments at DNV. For natural gas (0% hydrogen) the red hot flame zone is located downstream of the burner (located at the right side of each image), which allows flue gases to be entrained by the unburned (cold) jet. However, upon hydrogen addition, the flame moves towards the burner surface and the red hot zone seems to become hotter (whiter) and larger. The flame moves towards the burner tip upon hydrogen addition as a result of faster mixing of fuel and air but also due to the widening of the flammability limits and the increase in the burning velocity. The movement of the flame towards the burner tip also affects the entrainment of flue gases into the jet, as the flame moves closer, less flue gas is entrained and as a result the flame temperature (and hence NO_x emission) increases rapidly as indicated by the appearance of large white areas in the infrared images. It should be noted that the movement of the flame towards the burner tip is not a unique feature of the hot air burner; experiments at DNV for different burner types reveal that in the majority of the burner types studied, the flame moves towards the burner tip [7].

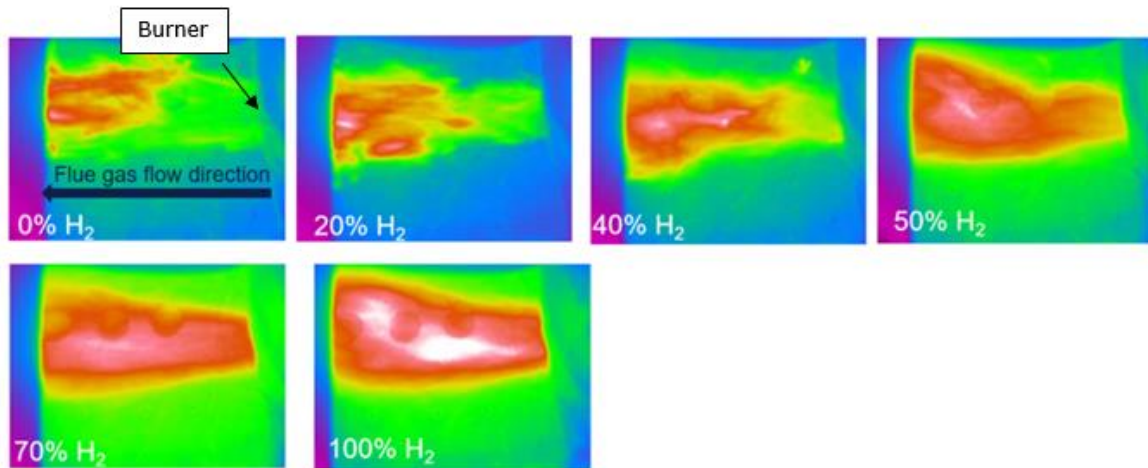


Figure 4.13: infrared images of hydrogen-natural gas flames for a hot air burner [7].

4.4.3 Recuperative burners

A recuperative burner is equipped with a heat exchanger that extracts heat from the exhaust gases to preheat the combustion air ('pipe-in-pipe' burner, see figure 4.14). These burners are often used for heat treatment processes where it is not desirable that the flue gases come into contact with the product such as metal annealing processes. The fuel is supplied via an inner fuel nozzle with a preheated co-flow of combustion air around it. For these types of burner systems, no data has been found on the effects of switching to hydrogen combustion.

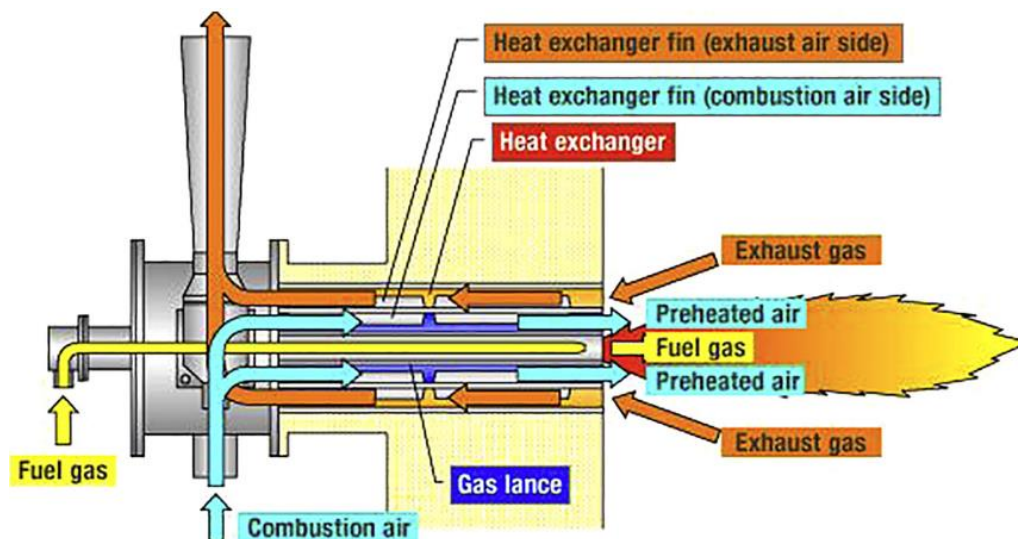


Figure 4.14: Schematic overview of a recuperative burner [12].

4.5 Radiant IR burners

Radiant heaters are (partially) premixed burner systems. These burners are used for drying processes in paper, food and plastic production industry. In radiant heaters, the flame is strongly stabilized on the burner tile. As a result of heat exchange between the flame and the burner tile, the temperature of the burner tile increases and the tile starts to radiate. The heat radiation is provided via mainly IR radiation. The heat transfer from the flame to the burner reduces the flame temperature (upstream heat loss), which lowers the NO_x emission. A disadvantage of these (partially) premixed burners is

their susceptibility towards flashback. Measurements performed at DNV show that these burner systems typically show flashback already between 30-50 vol% hydrogen in natural gas.

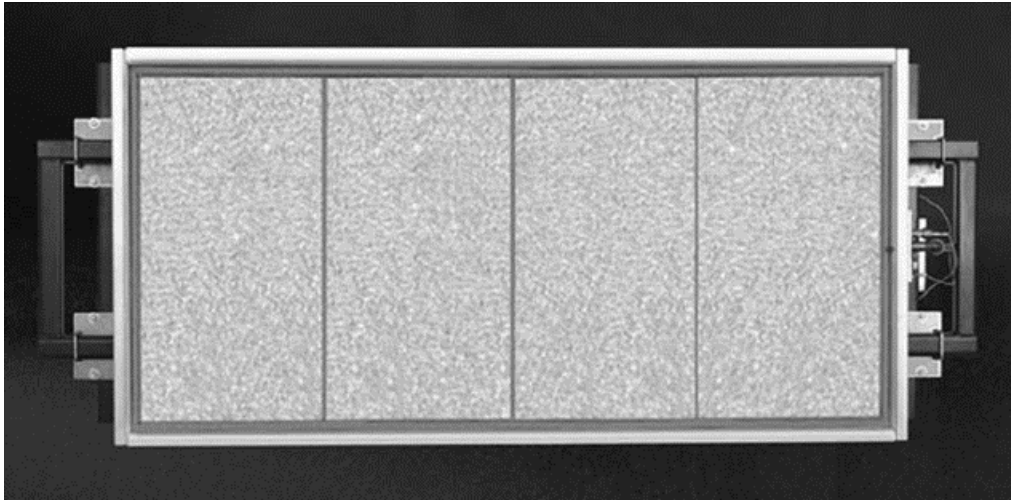


Figure 4.15: Radiant heater [4]

4.6 Swirl burners

Swirl burners are often used in industrial furnaces for incineration processes, metal treatment, and boiler systems. In a swirl burner, a set of guided vanes give the combustion air a rotating motion which improves flame stability and enhances the mixing process. The swirl motion also creates a recirculation of hot flue gases that are thus entrained in the fuel/air mixture. The size and position of this recirculation zone vary with the intensity of the swirl. This zone is an essential element for the stabilization of flames because it contains preheated (diluted with flue gas) fresh gases and allows better combustion and lower NO_x emission [4].

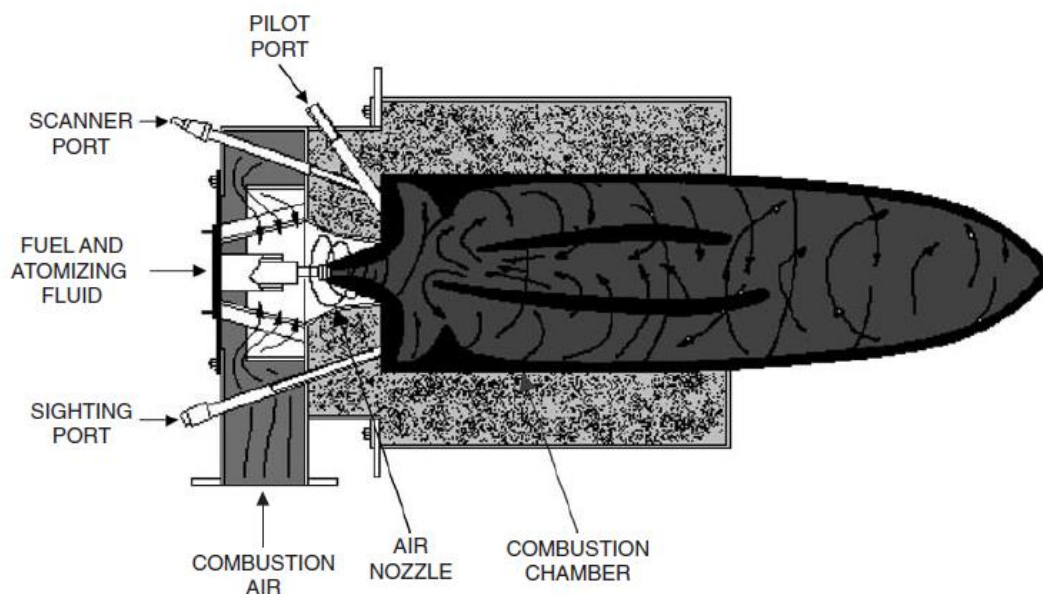


Figure 4.16: schematic view of a swirl burner [4]

At DNV two swirl burners are tested using hydrogen concentrations ranging from 0-100% hydrogen, one of these tested burners is presented in Figure 4.17.

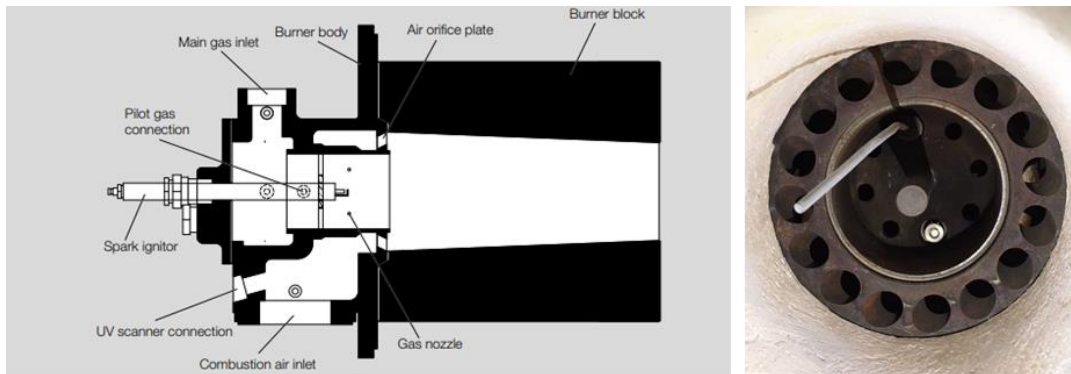


Figure 4.17: Swirl burner schematic (L) [18] and picture (R) tested at DNV using different hydrogen percentages in natural gas [13]

The measured NO_x emission for different hydrogen fractions in natural gas are shown in Figures 4.18 and 4.19. For both studied swirl burners the NO_x is shown to increase upon hydrogen addition. However, up to 70 vol% hydrogen only a minor increase is observed, while switching from natural gas to hydrogen increases the NO_x emission with more than a factor of two. This observed increase is attributed to the rapid mixing of fuel and air (faster combustion) that results in high (local) temperatures and hence NO_x emissions.

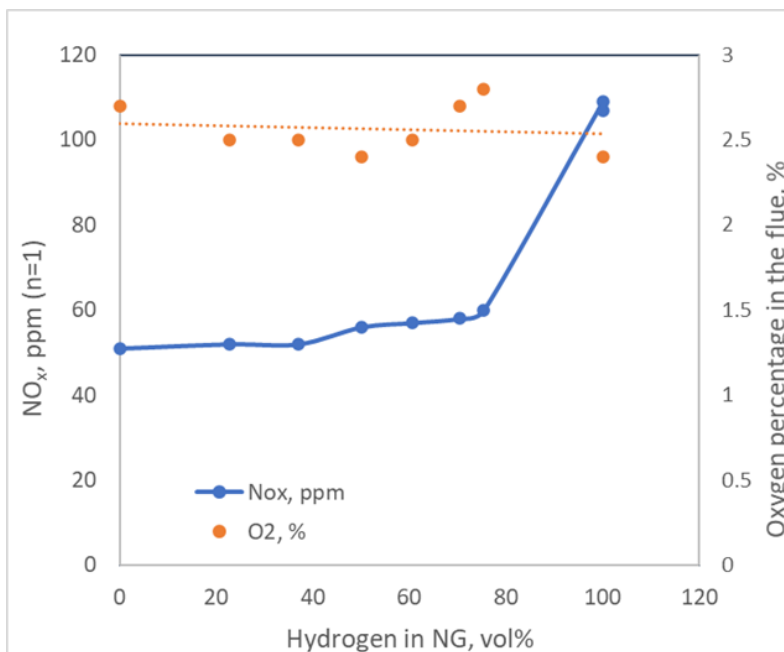


Figure 4.18: NO_x and O₂ concentration measurements in the flue gas for different hydrogen fractions in natural gas for Swirl Burner 1 [13]

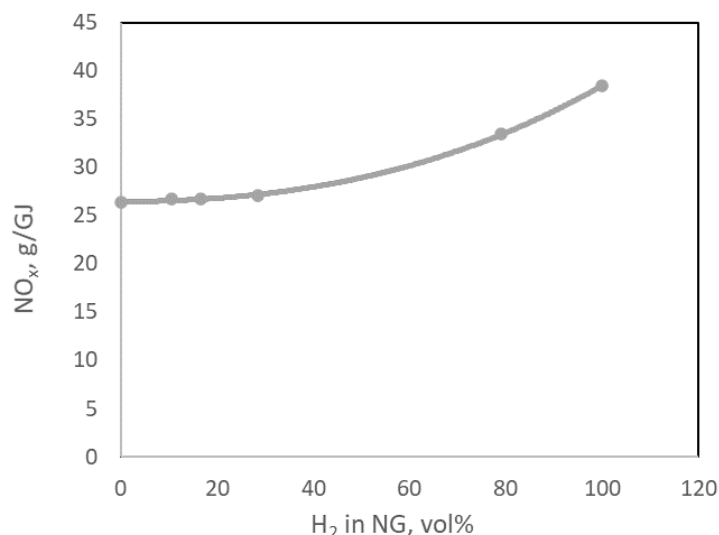


Figure 4.19: NO_x concentration measurements in the flue gas for different hydrogen fractions in natural gas for Swirl Burner 2 [7]

4.7 FLOX or flameless oxidation burners

In a FLOX burner fuel, air and fuel alongside the flue gasses are rapidly and intensely mixed in the combustion chamber before the onset of flame reactions can take place. The fuel and combustion air react hence in the whole, homogeneous gas volume downstream the burner nozzles, whereas in traditional flame combustion the reaction takes place in a highly intensive reaction zone attached to the burner nozzles. In these “flameless” burners, combustion reactions occur when the fuel and air are mixed with the hot combustion gases. A requirement is that the combustion chamber temperature is above the auto-ignition temperature. A high-temperature combustion (1200°C) [14, 15] is created without a visible flame. The temperature is however still below the minimum temperature for the thermal NO to occur which results in very low NO_x emissions for these burners, even when high air preheating or excess is applied, as the prompt NO_x formation is also suppressed due to the highly diluted form of combustion. Furthermore, it allows the burning of low calorific value gases without problems with flame stability. Due to this homogeneously distributed reaction, the temperature stress caused by peak temperatures is low and average temperatures can be increased significantly without changing the materials of the boiler.

In Ref [17] the effect of hydrogen addition to natural gas on the NO_x emissions and combustion behavior was investigated using a 150 kW flameless burner in a unloaded kiln. Up to 55 volume percentage hydrogen in natural gas, a flameless mode was achieved and the NO_x emissions were found to remain low (see Figure below). However, increasing the hydrogen volume percentage resulted in an higher NO_x emission and (at the same time) the occurrence of a visible flame. The authors conclude that the hydrogen addition (>55 vol%) alters the combustion mode of the burner because of stabilization of a flame at the end of the gas lance.

When using pure methane as a fuel, the burner fails to generate a flameless mode. In this case, the addition of hydrogen (15 volume percentage) was found to improve the flameless mode. Thus, where the addition of hydrogen (above 55 vol%) to natural gas was found to deteriorate the flameless mode, addition of hydrogen to methane improves the flameless mode. The authors attribute this difference in behavior to the presence of easily ignitable higher hydrocarbons in natural gas.

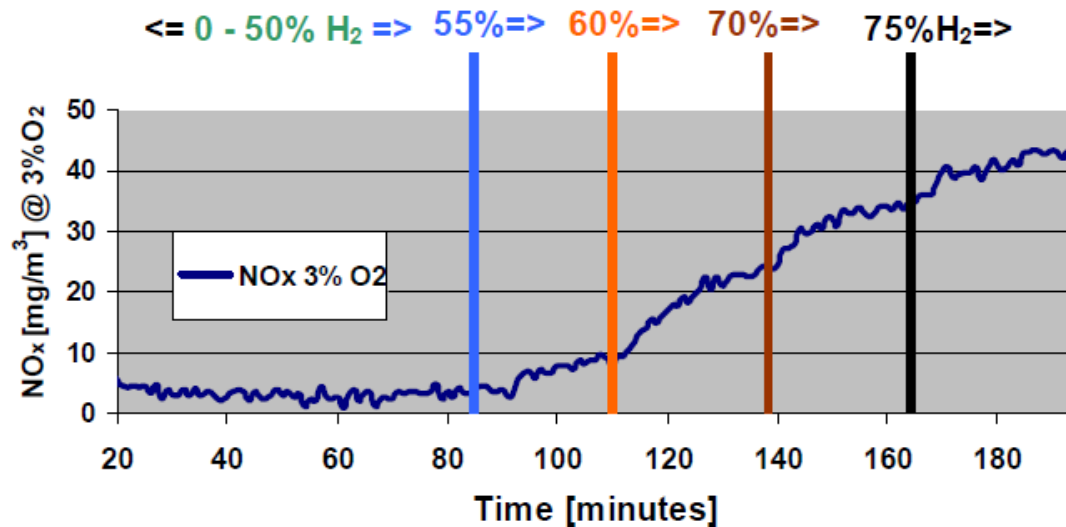


Figure 4.20. Measured NO_x emission for different volume percentages hydrogen in natural gas at constant furnace temperature [17].

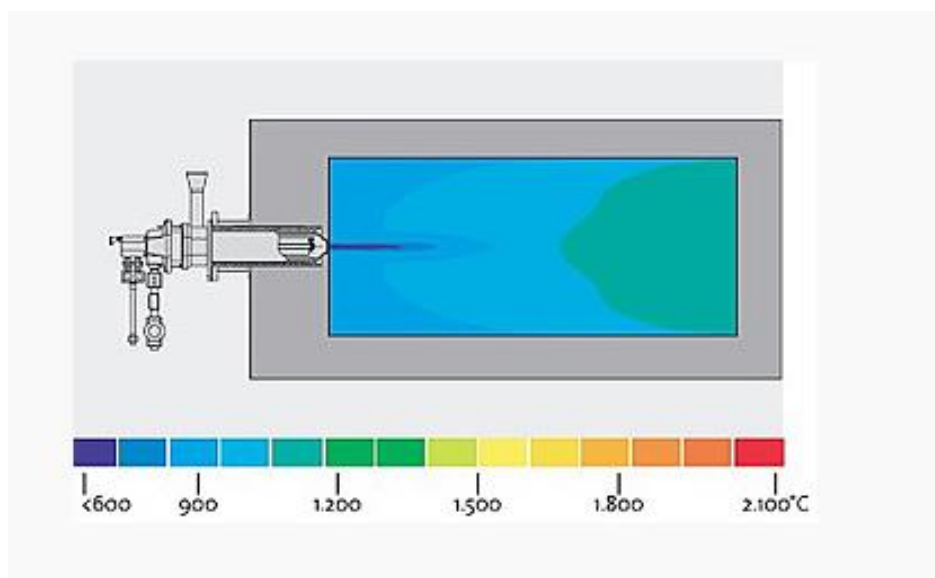


Figure 4.20: schematic view of a FLOX burner [19]

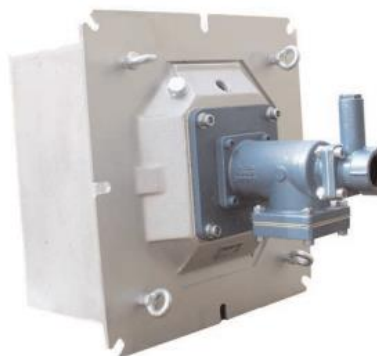
4.8 Lance burners

Lance burners are used in many high-temperature heating processes such as the ceramic industry. An example of a lance burner is presented below in which the fuel is supplied by an inner lance with a co-flow of air around it. No information has been found how these lance burners perform when using hydrogen as a fuel.



4.9 Flat flame burners

Flat flame burners are often nozzle-mixing units that have a high-velocity and spinning airflow. This creates a swirling air stream that produces an anticlockwise vortex inside the refractory block. Gas enters the vortex, mixing rapidly, and producing intense combustion. The inverted parabolic shape of the burner block port works with the vortex to pull flames flat to the furnace wall at firing rates and mixtures. High air excess operation allows for obtaining quite low flame temperatures (and hence low NO_x emissions) without changing the volumes of the fuel gas.



Pagina 35/75

4.10 Summary overview burner types / burner type and application area's

| | Forced draught burners | High speed burners | Lance burners | Flat flame burners | FLOX burners | Recuperative burners | Swirl Burners | Regenerative burners | IR burner |
|-------------------------------|-------------------------------|---------------------------|-------------------------|---------------------------|---|-----------------------------|------------------------------|-----------------------------|--------------------|
| <i>Mixing Type</i> | Radial Fuel Injection | parallel (pipe-in-pipe) | parallel (pipe-in-pipe) | multiple types | fuel injection homogeneous distributed hot oxygen | parallel (pipe-in-pipe) | fuel injection into a vortex | parallel (pipe-in-pipe) | (partial) premixed |
| <i>Metal /steel treatment</i> | X | X | X | X | X | X | X | | X |
| <i>Glass melting</i> | | | | X | | | | X | |
| <i>Ceramics</i> | | X | X | | | | | | |
| <i>Hot water /steam</i> | X | | | | | X | | | |
| <i>Asphalt production</i> | X | | X | | | | | | |
| <i>Calcination</i> | | | X | | | | | | |
| <i>Drying processes</i> | X | | | | | | X | | |
| <i>Incineration processes</i> | | | | | | | X | | |
| <i>Refinery</i> | | | | X | | | X | | |

The inventory on the different burner technologies reveal that many different burner types are used in industrial processes each designed for a specific process. For example, several burners are designed with the aim of generating mainly radiative heat transfer while other are designed to generate mainly convective heat transfer. As a result the diversity of different designs of nozzle mix burners is large. Examples of nozzle mix concepts identified in the literature are swirl burners, pipe-in-pipe burners, FLOX burners, regenerative burners, recuperative burners and radiant heaters. As can be seen in the Table above the majority of the burners make use of the swirl- or pipe-in-pipe burner concept.

4.11 References

- [1] <http://www.termolombarda.it/PdfProdotti/serie-x-generatori-aria-calda-a-condensazione-brochure-illustrativa.pdf>
- [2] <https://www.process-heating.com/articles/85945-burner-history-104-how-industrial-burners-evolved-for-modern-process-heating-applications>
- [3] S. Gersen, B. Slim, R. Zeijlmaker, M. Van Essen and R. Tichelaar, The Development of a Natural Gas/Hydrogen Boiler System, IGRC 2020
- [4] C.E. Baukal, Industrial burners handbook, 2003.
- [5] <https://www.fivesgroup.com/energy-combustion/burners-systems/north-american-hiram#:~:text=The%20North%20American%20HiRAM%C2%AE%20LNI%20burner%20technology%20takes%20advantage,oxygen%20concentration%20and%20residence%20time>
- [6] John N. Newby, 21 Years of Real-World Low NO_x Injection (“LNI”), AFRC 2013 Industrial Combustion Symposium – Kauai, September 22-25, 2013
- [7] Gersen, S., “Explore the use of sustainable hydrogen as a fuel for industrial heating processes (public summary)”, Netherlands Enterprise Agency (RVO), Report 22-1123, 07-15-2022
- [8] Nakamura, T. & Nakamachi, I. (2000). Development of Low NO_x Regenerative Burner System. Kagaku Kogaku Ronbunshu, 33, pp. 221-226.
- [9] <http://www.globalcombustion.com/product/vitroglo-rp/>
- [10] <https://www.hotwork.ag/products/glass/air-fuel-burners-injectors/>
- [11] J. Lechner, K. Gerner, Decarbonizing process heat in the glass industry with hydrogen and hydrogen / natural gas blends, Conference: 26th International Congress on GlassProject: HyGlass(2022)
- [12] Amisha Chauhan, Savvas A. Tassou, Waste heat recovery technologies and applications, Thermal Science and Engineering Progress 6 (2018) 268-289
- [13] Gersen, S., “Small Business Innovation Research (SBIR) project : Varigas: Een industrieel waterstof/aardgas brandersysteem,” 2018-2019, <https://www.rvo.nl/sites/default/files/2020/05/RVO%20SBIR%20gas%20projectenoverzicht.pdf>
- [14] Milani, Ambrogio & Wüning, J.. (2007). Flameless Oxidation Technology. 10.1007/978-1-4020-6515-6_26. <https://e-flox.de/en/frontend/2>
- [15] https://flox.com/documents/04_Mil.pdf
- [16] <https://www.esapyronics.com/wp-content/uploads/2014/12/E3503E.pdf>
- [17] H. B. Levinsky, B. Slim and G. Pieters, “Consequences of hydrogen addition to natural gas for industrial utilization systems”
- [18] Leicher, Jörg & Giese, Anne & Islami, Bledar & Görner, Klaus & Overath, Johann. Decarbonizing process heat in the glass industry with hydrogen and hydrogen / natural gas blends. (2022).
- [19] <https://e-flox.de/en/frontend/2>

5 NO_x reduction methods

In the past, various strategies have been developed for mitigating NO_x emissions. Which strategy can be applied depends strongly on the type of combustion system. Since NO_x emissions are dependent on the local temperatures inside the flame and the residence time, most of these reduction methods are based on lowering the local temperature and the residence time of the hot spots inside the flame. In this chapter the NO_x mitigating strategies are summarized.

5.1 (Partially) premixed combustion

Lean premix combustion

By using an excess of air (equivalence ratio < 1), the NO formed via the Zeldovich mechanism will be reduced as shown in Figure 3.6. These premix burner systems are used in both the industry where pencil burners and DrylowNO_x gas turbine burners are installed and in the domestic market where for example lean premixed boilers are used. At DNV a prototype hydrogen lean premixed burner system for hydrogen combustion in a domestic appliance was developed using lean combustion at an equivalence ratio of about $\phi=0.45$. The result shows that the NO_x emission from this appliance was lower than the European legal limits of 56 mg/kWh for natural gas. The company Bekaert developed a lean premixed LowNO_x hydrogen burner. Tests with a domestic boiler, shown in Figure 5.1, reveal that the NO_x emission using this burner system is much lower than the European NO_x limit.

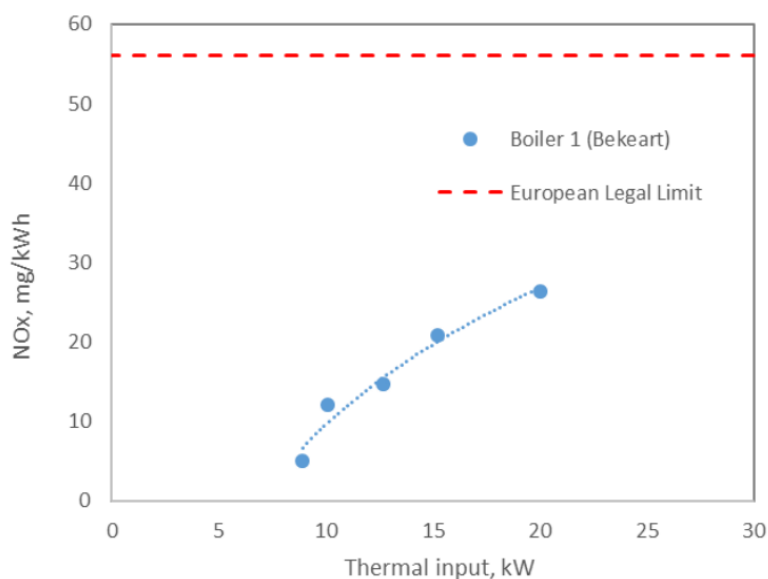


Figure 5.1: Measured NO_x emission from a domestic hydrogen appliance having a Bekaert premixed LowNO_x hydrogen burner [1].

Air staging

With flow-through water heaters and cooking stove burners, combustion takes place in two steps; initially, a premixed fuel-rich mixture is burned (under oxygen-deficient conditions). As a result, the NO emission is low (formed via the Fenimore mechanism, see Chapter 3). An excess of air is then added in the second combustion step. In both stages combustion takes place at relatively low flame temperatures (fuel-rich in the first stage, fuel-lean in the second stage), resulting in an overall low

NO_x emission. At DNV measurements with natural gas/hydrogen blends have been performed for these combustion systems but resulted in flame flashback when using hydrogen percentages above 50-70 vol% hydrogen. To our knowledge, no tests or developments for pure hydrogen combustion using partially premixed air staging combustion have been performed or initiated.

Flame stabilization

As shown above, radiant burner systems are widely used in the industry for drying processes. The working principle is based on creating upstream heat transfer to the burner deck by stabilizing the flame closer to the burner surface (also called burner stabilization). As a result, heat transfer takes place between the burner and the flame. The closer the flame is to the burner, the more heat is transferred to the burner; the burner starts to radiate. As a result, the flame transfers its energy to the burner deck and cools down (the flame temperature drops), and consequently, the NO_x emissions decrease. Experiments performed at DNV show that these burner systems containing a ceramic burner deck are prone to flashback when adding hydrogen to natural gas. Therefore new radiation burner systems need to be developed for pure hydrogen combustion. One of the innovative combustion designs found in the literature is the ceramic foam burner design developed in the past by ECN [2,3].

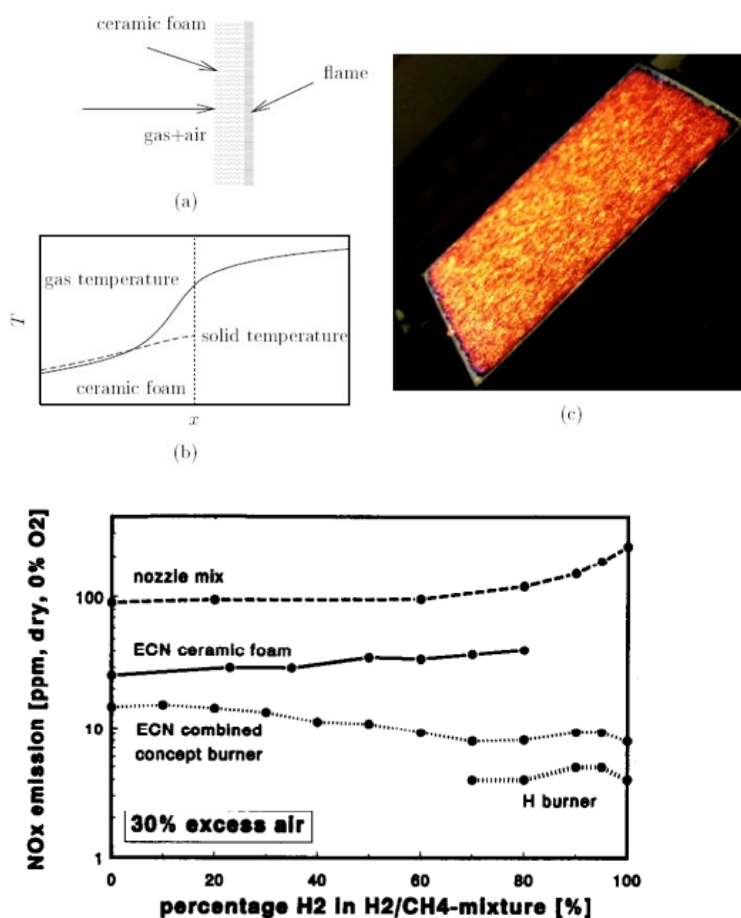


Figure 5.2 ceramic foam burners design developed by ECN [2, 3]

The ceramic foam burner is using a new combustion concept, where the flame is stabilized in the homogeneous foam structure, creating low NO_x and CO emission, large modulation width, and high efficiency. Furthermore, the glowing ceramic foam burner generates comparably intensive and homogenous thermal radiation whose wavelength can be precisely adapted to drying processes. The foam structure contains narrow channels through which the fuel/air mixture flows which prevents the occurrence of flame flashback in the burner deck. Ceramic foam burners turned out to be capable of burning hydrogen/natural gas mixtures with up to 70% hydrogen [3]. A new burner, the so-called H burner was developed to burn pure hydrogen. To ensure low NO_x emissions “internal flue gas recirculation” was applied. Testing this ceramic foam burner for industrial heating processes using hydrogen as a fuel would be interesting, given the potential to create low NO_x combustion.

5.2 Non-premix combustion

As discussed in Chapter 3 the majority of industrial burner systems are non-premixed burner systems in which the fuel and air are mixed downstream of the burner tip. These burners are often called diffusion or nozzle mix burners. Several NO_x mitigating strategies have been applied for these burner systems such as flue gas recirculation and staged combustion describe below.

External flue gas recirculation

One of the strategies to reduce the flame temperature and subsequently reduce the NO_x emission is by applying flue gas recirculation. For processes where the exhaust temperature is relatively low <500°C applying external flue gas recirculation is relatively easy. As shown in Figure 4.5 the NO_x emission of a forced draught burner present in a boiler fueled by hydrogen is three times higher than when using natural gas. To decrease NO_x emissions, external flue gas recirculation was applied [4]. When applying flue gas recirculation, a part of the flue gases was returned to the combustion air inlet. The dilution of air with (inert) flue gases reduced the adiabatic flame temperature and consequently reduced (thermal) NO_x formation. The amount of flue gas recirculation applied was controlled by a control valve that was connected to the Burner Management System. As can be seen in Figure 5.3, about 9 vol. % flue gases in the combustion air were needed to bring NO_x emissions below the Dutch legal NO_x limits. From this, it can be concluded that applying external flue gas recirculation is a very effective method to reduce the NO_x emission for ‘low’ temperature processes.

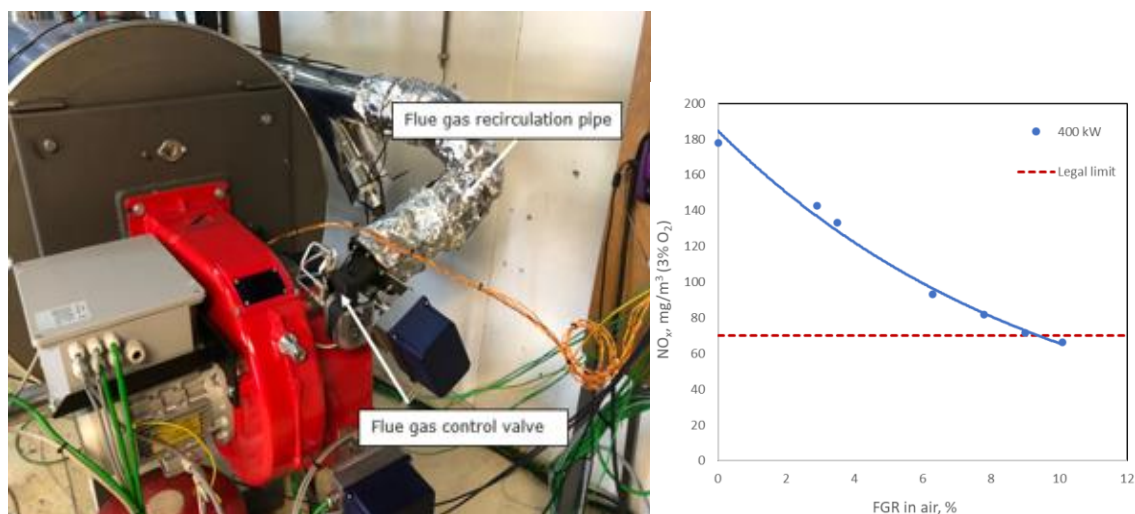


Figure 5.3: Effect of applying flue gas recirculation in the combustion air on NO_x emissions at $\lambda=1.2$ and a thermal input of 500 kW (left) and flue gas recirculation applied on the burner/boiler system (right) [4].

Internal flue gas recirculation

For high-temperature heating processes, applying external flue gas recirculation is more complex and expensive, since the flue gas temperature is high and thus cooling of the flue gases is required. For most natural gas burner designs, internal flue gas recirculation is applied. In such case flue gases are internally recirculated back to the burner by creating an area of lower pressure in the vicinity of the burner. This under-pressure can be created by for example using a swirler, a bluff body or flue gas entrainment into a high-velocity jet (impulse momentum). Several studies have been performed for hydrogen combustion to improve the internal flue gas recirculation using new designs as described below.

In [5] the study focuses on burning hydrogen in a burner for hydrocarbon fuels. The study experimentally evaluates the rates of NO_x production that are affected by changing the diameter and number of combustion air nozzles. A commonly used diffusion combustion burner with recirculation flow retention, shown in Figure 5.4, is used. The combustion air velocity is adjusted by replacing or plugging different air nozzles, as shown in Figure 5.5. Comparing Figure 5.5c and d, the gas/fuel velocity ratio is the same, however, when multiple jets are placed near each other, the space between the jets will become depressurized, which will stimulate recirculation flows.

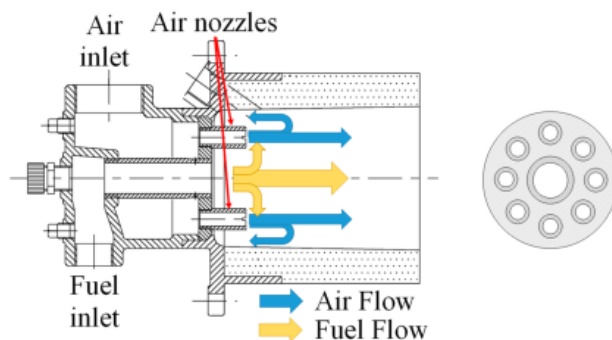


Figure 5.4: Burner system with a central fuel jet surrounded by multiple air nozzles [5]

This experiment showed that reducing the number of air nozzles increases the distance between air nozzles, resulting in a larger flue gas recirculation rate. In hydrocarbon combustion, the burner would misfire, but hydrogen will have maintain a stable flame without distorted combustion. When the number of air nozzles was reduced to one half, the flue gas recirculation rate was about 10%, and resulted in the confirmed low-NO_x combustion.

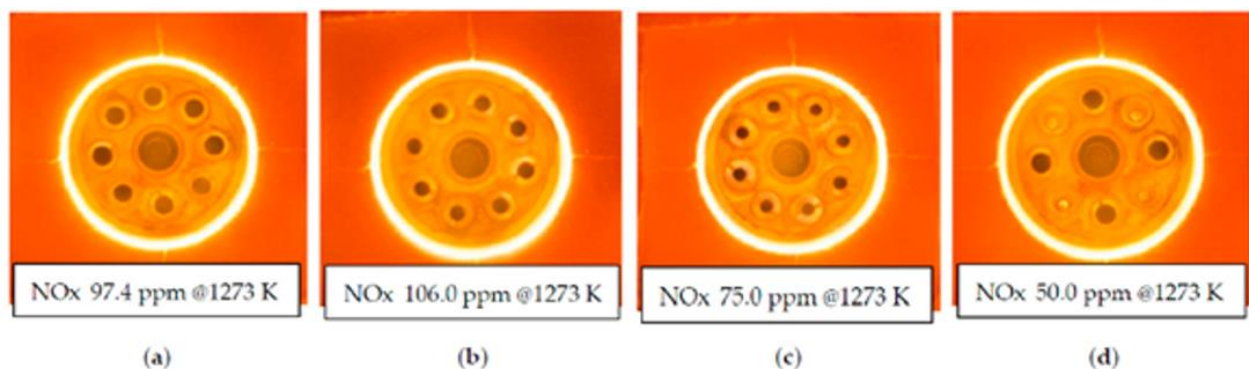


Figure 5.5: Photographs at 100% Hydrogen and 1273K at different amounts of nozzles and sizes: a) 11.9mmx8, b)10.2mmx8, c) 8.4mmx8 and d)11.9mmx4 [5]

In [6] a ultra-low NO_x round flame burner was developed and tested for the refinery industry. The COOLstar™ burner stages the fuel with raw gas tips and internally recirculates flue gases. As shown in Figure 5.6 the NO_x was kept low all fuel blends tested.

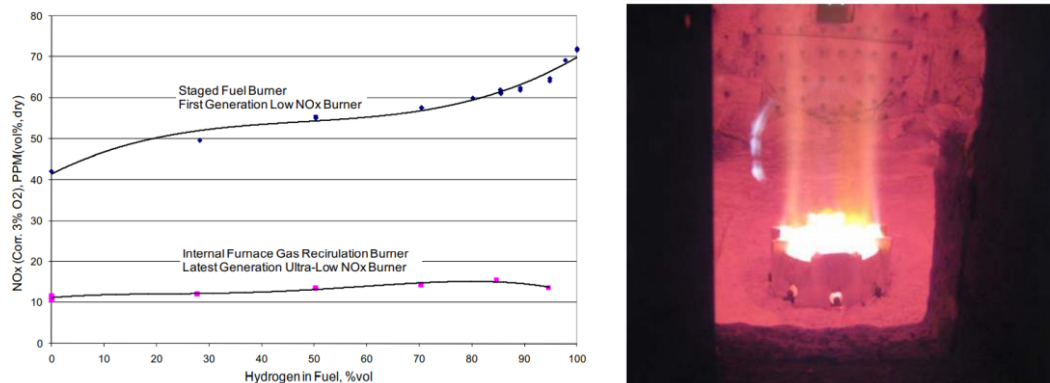


Figure 5.6: NO_x emissions for different hydrogen blends in an ultra-low NO_x round flame burner [6]

The low NO_x round burner [7] incorporates the Coanda effect for controlling fluid flow, mixing, and stability. As an illustration Figure 5.7 shows the Coanda principle in which a fluid is supplied to a chamber with a small opening. A curved surface with a designed profile causes the fluid to be drawn against the surface instead of being injected straight out of the nozzle to create an under-pressure region in which air and flue gases are entrained.

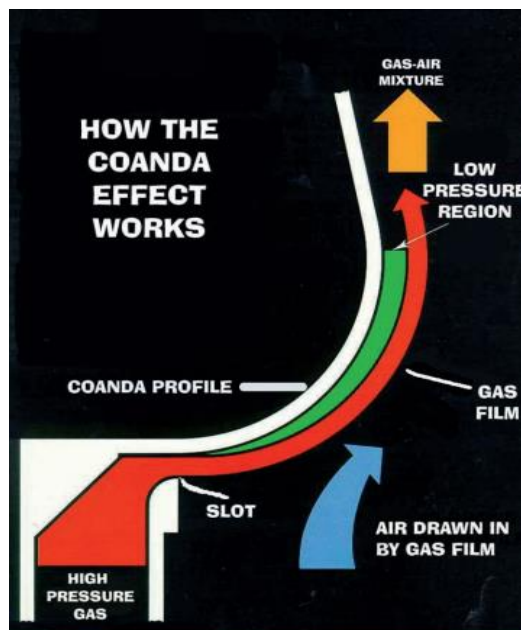


Figure 5.7: Illustration of the Coanda effect [7]

A preliminary burner design was created by Zeeco [8] based on their free-jet burner principle. The burner design illustrated in Figure 5.8 lowers the NO_x by using internal flue gas recirculation. The design aims to mix the flue gas before combustion starts to lower the flame temperature and NO_x emission. The flue gases are mixed with the fuel leaving the jet port. This is achieved when the flame is stabilized in a low-pressure recirculation zone created on a series of hot refractory ledges.

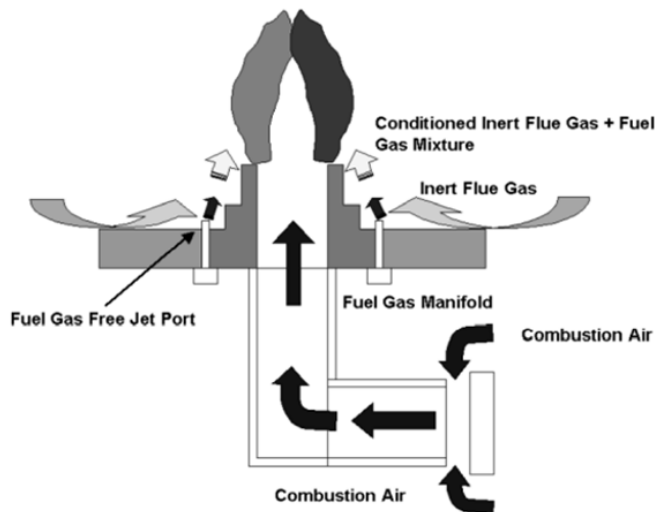


Figure 5.8: illustration of the free jet burner [8].

A second burner configuration designed by Zeeco [8] is by using a trapped vortex reaction zone to enhance flame stability and to entrain air and flue gases. The design illustrated in Figure 5.9 shows that a stabilization zone is created by diverting small portions of fuel and combustion air into an annular chamber within a tile. The hot flue gases and reactive species generated in this zone are transported to the top of the tile where they mix and react with traditional fuel and air in the second stabilization zone. The flue gases lower the flame temperature and the NO_x emission

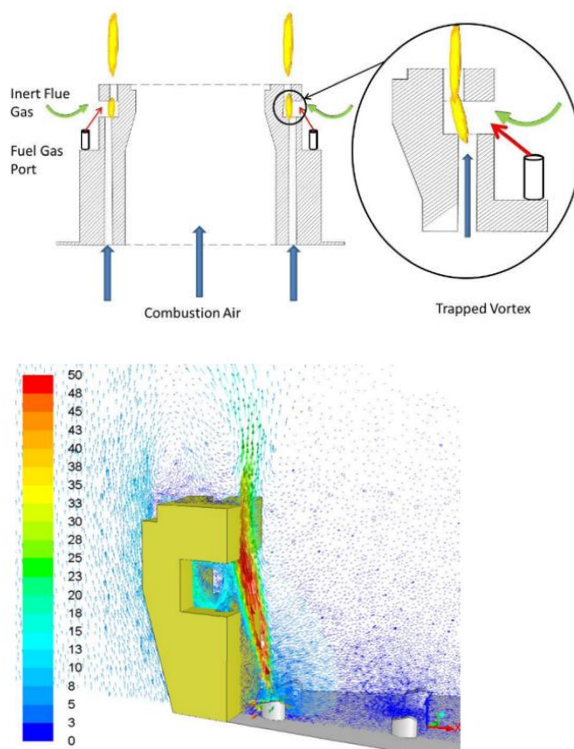


Figure 5.9: Illustration of the trapped vortex [8]

Staged Fuel burner

To achieve stable combustion with good flame shapes, and minimal NO_x emissions, a burner can be designed to ensure that the proper amounts of fuel and air are introduced in the right locations within the burner. When the fuel is introduced at different points of the airflow, the first portion of the fuel that encounters the combustion air forms a primary flame. Fuel that is injected further downstream from the primary fuel, is called staged fuel [9]. As illustrated in Figure 5.10 secondary or tertiary injectors can be used to inject a portion of the fuel and/or air into the flame to control heat transfer, produce longer flames, and reduce pollutant emissions such as NO_x. These longer flames typically have a lower peak flame temperature and more uniform heat flux distribution than nonstaged flames. Furthermore, flue gases produced in the primary flame mix with the secondary flame which reduces the flame temperature and NO_x formation. A challenge with staged combustion is that multiple longer flames might interact with each other and produce unpredictable consequences (shapes and mixing conditions) compared to single, shorter flames [9]. An example of a staged air burner is schematically shown in Figure 5.10.

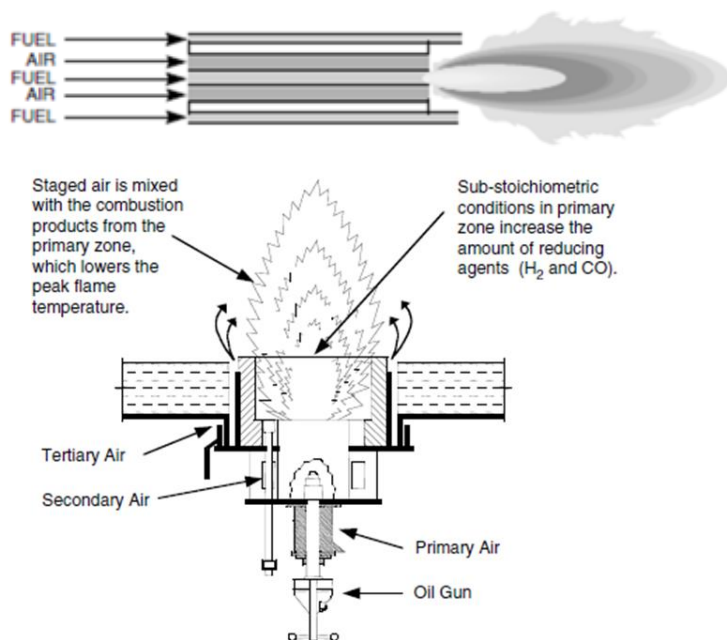


Figure 5.10: Staged combustion [9]

Mild combustion

The method of MILD (moderate or intense low-oxygen dilution) combustion is a successful combustion technology to lower NO_x emission [11]. In MILD combustion, in fact a specific form of FLOX combustion, the fuel is slowly oxidized in an environment where oxygen is highly diluted by recirculated exhaust gases and the furnace temperature needs to be above the auto-ignition temperature of the fuel. Due to the 'slow' combustion in a highly diluted environment, the temperatures are much lower than the adiabatic flame temperature and the temperature distribution is 'uniform'. At DNV a FLOX burner was developed that operated very well with natural gas. However, when hydrogen was introduced into this burner system the oxidation rate increased causing a flame structure instead of MILD/FLOX combustion. Currently, at TNO a new novel FLOX burner concept is developed, which is confirmed to operate well and with marginally higher NO_x.

emissions on pure hydrogen. In order to achieve this and also to provide higher load flexibility and load-following potential, this so-called HyFlexFIOx burner is also featuring electric combustion air preheating.

Recently, several companies developed MILD burners. For example the company Tenova [19] developed a MILD combustion methane/hydrogen air burner for heat treatment furnaces.

5.3 NO_x reducing strategies for gas turbines

Below the NO_x mitigating measures for gas turbines are described.

Water injection

For gas turbines with diffusion burners, no premixing of gas and air takes place. Combustion takes place on the stoichiometric (ideal combustion) plane (see also description of industrial burners) regardless of the gas/air ratio. Mixing hydrogen with methane will lead to a higher flame temperature and a corresponding increase in NO_x emissions. To control the NO_x emission upon hydrogen addition to natural gas, diffusion combustors typically use water or steam injection to quench the flame [35]. Generally, water injection provides a 60-70% reduction in NO_x emissions for small turbines, with large turbines percentages between 70-80% can be achieved. This nonetheless only works efficiently in combined cycle systems, as otherwise the cooler flames and the resulting loss of expansion results in a slightly lower mechanical system efficiency.

Premixed combustion systems (DLN: dry low NO_x)

Gas and air are premixed in the premixed gas turbines. This type of burner works with an excess of air - up to the limit of what is still stable in terms of combustion. In this way, the flame temperature is kept as low as possible, which is favorable for limiting NO_x formation (see also Chapter 3). With this type of gas turbine, the application of water/steam/nitrogen injection is often not necessary, which is beneficial for the efficiency and maintenance regime.

The addition of hydrogen to natural gas greatly increases the risk of undesired combustion technical effects, such as flame impaction/impingement and flame blow-off. In addition, unwanted vibrations and pulsations can occur. This can lead to failure and even serious damage to the turbine. For example, in consultation with the manufacturer, it can be decided to adjust burner settings such as larger air excess for burner stabilization or changing the burner system such as the micromix burner described below.

Micromix (hydrogen) combustion

Ref [13] focuses on the micromix combustion principle (MCP), which is based on the jet crossflow principle of miniaturized multiple injectors. Here, the fuel is injected perpendicularly into an air crossflow as shown in Figure 5.11. Together with this phenomenon, hydrogen segments and air-guiding panels act as bluff bodies, enhancing the formation of inner and outer vortices. Following the flame splitting method, the reaction zone is separated into multi-diffusion turbulent flames anchored between inner and outer vortices, reducing the high-temperature volumes where thermal NO_x rates increase and therefore reducing the overall thermal NO_x formation. Moreover, the resulting fast and

turbulent mixing between reactants reduces the residence time of the reactants and flue gas (hence also NO_x precursors) in the high-temperature regions, which also reduces the overall thermal NO_x formation.

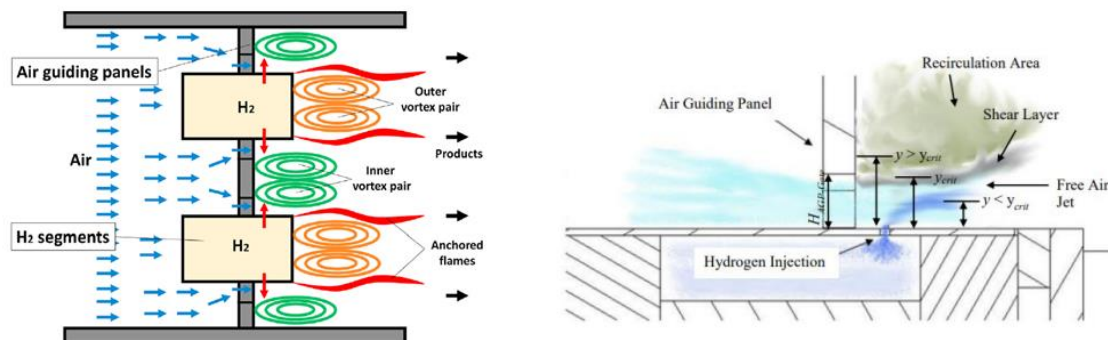


Figure 5.11: Micromix combustion left [13] and right [14]

5.4 Oxy fuel combustion

Oxyfuel combustion has the advantage of high efficiency and potentially near-zero NO_x emissions assuming that (nearly) no nitrogen is present, making it an attractive combustion technology. For high-temperature processes, such as glass melting, oxyfuel combustion is commonly used (using natural gas as a fuel) to create such high temperatures. The flame temperature of natural gas and H₂/O₂ flames are about 2800 °C. At DNV the ALGLASS Air Liquide burner was tested successfully for pure hydrogen. When there is no air leakage into the furnace and when there is no nitrogen impurity present in the hydrogen, there will be no production of NO_x. However, an even limited presence of nitrogen will produce high levels of NO_x due to the high adiabatic flame temperatures.



Figure 5.12: H₂/O₂ flame using the ALGLASS Air Liquide burner tested in the high-temperature furnace at DNV [15]

5.5 Post treatment NO_x reduction methods

If the combustion in burners produces too high NO_x levels, post combustion processes are needed to remove the NO_x emission. Industries, power plants, and gas turbine operators often use a catalyst with the addition of ammonia as an post-treatment or end-of-pipe system to convert the NO in flue gases to N₂ and water. This system called Selective Catalytic Reduction (SCR) is active over a wide range of exhaust temperatures [16] and may also be used in combination with oxidative catalysts to remove traces of unburnt hydrocarbons, CO or even micro-trace pollutants such as 'dioxins'. Another option is to use Selective Non Catalytic Reduction process (NSCR) for decomposition of thermal NO_x formed in the hottest zone of the installation. In this method ammonia (NH₃) is added to the flue gases. When the flue gases are at a sufficiently high temperature the NO reacts with OH to NH₂ and subsequently, NH₂ reacts with NO to form water and N₂. Although simpler and more thermally-efficient than SCR, this method has a drawback in the form of well-controlled temperature of about 1000 °C [16] and relatively long residence time (1-2 s). Other methods currently in development are for example using a plasma to convert the NO to N₂ and water but also activated carbon can be used to convert the NO to NO₂ and the NO₂ is absorbed by the activated carbon [17]. The efficiency of this method strongly depends upon the process conditions such as the temperature of the flue gases as shown in figure 5.13.

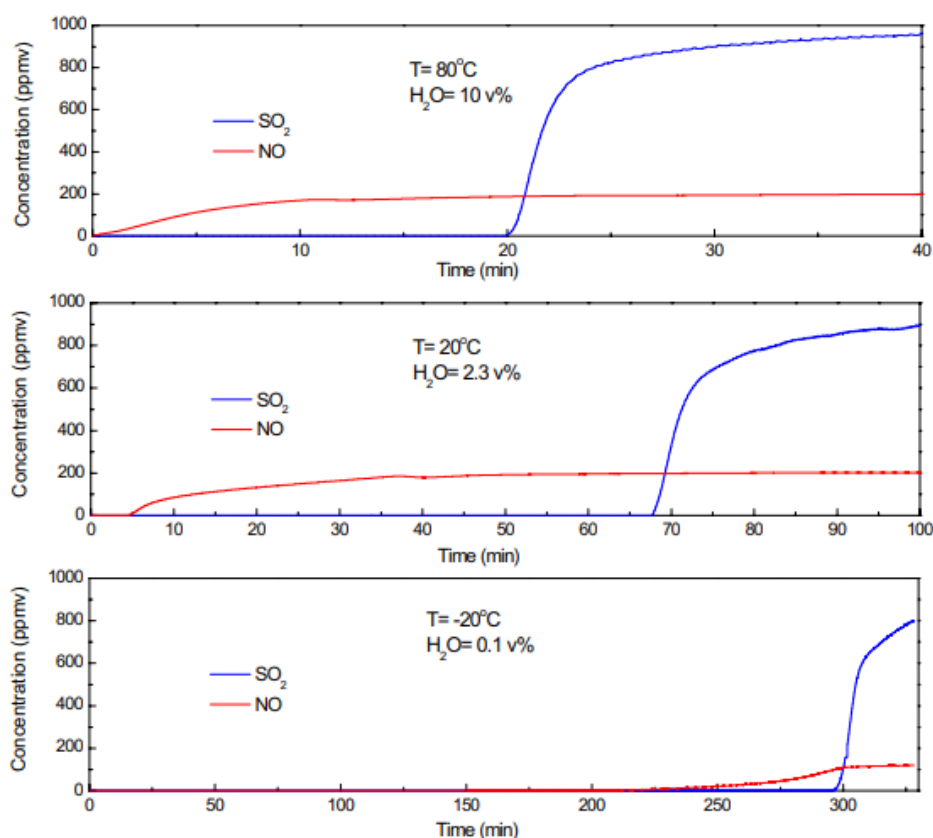


Figure 5.13 Breakthrough curves of SO₂ (blue) and NO (red) adsorption at 80, 20 and – 20°C (Simulated flue gas: SO₂=1000 ppmv, NO=200 ppmv, O₂=6 vol.%, CO₂=12 vol.%, space velocity=5000 h⁻¹) [17]

5.6 Summary strategies to reduce NO_x

In the previous Chapters, several NO_x-reducing combustion strategies have been discussed. The most commonly used strategies to reduce NO_x are summarized below.

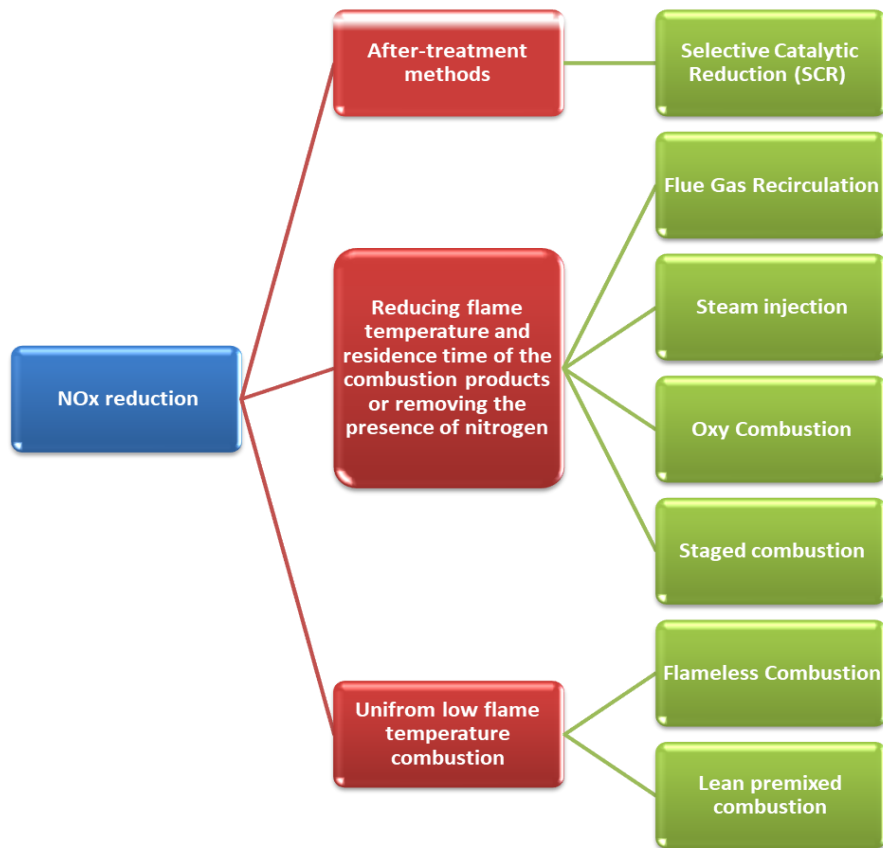


Figure 5.14: Summary of the NO_x mitigating strategies

This study aims to develop design rules for a low NO_x hydrogen burner to prevent or limit the need for NO_x after treatment. The focus of this project will be on air combustion. To summarize, designers of combustion systems use the following three factors in their design to lower the NO_x emission;

- Residence time (time scale to produce NO_x)
- Mixing rate with air (controlling temperature and residence time)
- Mixing rate with flue gases (controlling flame temperature and oxygen concentration)

Several combustion strategies have been described above using different designs to reduce NO_x emission. These strategies aim to control peak flame temperature and oxygen concentration by creating internal flue gas recirculation, reducing the residence time of the hot flame zone, and reducing hot spots. For hydrogen, the major challenge is the higher flame temperature, the wider flammability limits, faster diffusion, and higher burning velocity. Altogether, they move the flame closer to the burner tip causing lower internal flue gas recirculation and higher flame temperatures.

5.7 References

- [1] Gersen, S., Slim, B., Zeijlmaker, R., van Essen, M. et al. (eds.), “Domestic Hydrogen boiler in practice: Enabling the use of hydrogen in the built environment,” International Gas Research Conference (IGRC) 2020, Muscat, Oman, February 24-26, 2020.
- [2] Lammers, F. A.. Ceramic-foam surface burners in high-temperature environments. [Phd Thesis 1 (Research TU/e / Graduation TU/e), Mechanical Engineering]. Technische Universiteit Eindhoven (2001).
- [3] A. van der Drift, S.L. Tjeng, J.J. Beckers and J. Beesteheerde, Low-NO_x hydrogen burner, Iht. J. Hydrogen Energy Vol. 21, No. 6, pp. 445 449, 199
- [4] S. Gersen, B. Slim, R. Zeijlmaker, M. Van Essen and R. Tichelaar, The Development of a Natural Gas/Hydrogen Boiler System, IGRC 2020
- [5] Ref: Kenta Kikuchi, Tsukasa Hori and Fumiteru Akamatsu, Fundamental Study on Hydrogen Low-NO_x Combustion Using Exhaust Gas Self-Recirculation, Processes 2022, 10, 130.
<https://doi.org/10.3390/pr1001013>
- [6] Cliff Lowe, Nick Brancaccioa, Dan Battenb, Chris Leungb, Dick Waibel, Technology Assessment of Hydrogen Firing of Process Heaters, Energy Procedia 4 (2011) 1058–1065
- [7] D. Brown, C. Baukal, A variety of tests, Hydrocarbon Engineering, April 2010
- [8] C. Benson and R. Wilson, Fuel-Flexible Combustion System for Refinery and Chemical Plant Process Heaters, Project number DE-EE0000069
- [9] C.E. Baukal, Industrial burners handbook, 2003.
- [10] Tsuji H, Gupta A, Katsuki M. High temperature air combustion: from energy conservation to pollution reduction. Florida: CRC Press; 2003.
- [11] Wüning JA, Wüning JG. Flameless oxidation to reduce thermal no-formation. Progress in Energy and Combustion Science 1997;23(1):81e94.
- [12] Michael Welch, Brian M Igoe, Gas Turbine Fuel and Fuel Quality Requirements for use in Industrial Gas Turbine Combustion, Proceedings of the Second Middle East Turbomachinery Symposium 17 – 21 March 2013, Doha, Qatar
- [13] G. Lopez G, , I. Alava a, J.M. Blanco Study on the feasibility of the micromix combustion principle in low NO_x H₂ burners for domestic and industrial boilers: A numerical approach NO_x reduction methods, Energy, Volume 236, 1 December 2021, 121456.
- [14] Funke, Dr.-Ing. Harald et al. “Experimental and Numerical Characterization of the Dry Low NO_x Micromix Hydrogen Combustion Principle at Increased Energy Density for Industrial Hydrogen Gas Turbine Application. Vol. 1, June 2013.
- [15] S.Gersen, “Explore the use of sustainable hydrogen as a fuel for industrial heating processes (public summary)”, Netherlands Enterprise Agency (RVO), Report 22-1123, 07-15-2022
- [16] J. Warnatz · U. Maas · R.W. Dibble, Combustion, Physical and Chemical Fundamentals, Modeling and Simulation, Experiments, Pollutant Formation, 2006
- [17] ShiqingWang, Shisen Xu, , ShiwangGao, , Ping Xiao, , Minhua Jiang, He Zhao, Bin Huang, Lianbo Liu, Hongwei Niu, JinyiWang & DongfangGuo. Simultaneous removal of -SO₂ and -NO_x from fue gas by low-temperature adsorption over activated carbon, Nature , Scientific Reports | (2021) 11:11003
- [18] <https://process.honeywell.com/us/en/products/thermal-solutions/burners-and-heat-exchangers/direct-fired-furnace-burners/kinemax>

- [19] <https://furnaces-international.com/news/tenova-launches-hydrogen-ready-burner-with-integrated-industry-4-0-technologies-for-heat-treatment-furnaces>

6 Developing design rules for LowNO_x hydrogen high temperature burners

As discussed above, many different burner types are used in industrial processes, each designed for a specific process. The majority of the burners used for high temperature heating processes are nozzle mix (diffusion) burners in which fuel and air are not premixed before entering the burner and no sufficient LowNO_x technics are available yet when using hydrogen as a fuel. The aim of this study is to develop design rules for a nozzle mix LowNO_x hydrogen burner based on the findings from the literature inventory and performing flow field and combustion calculations.

From the literature inventory it was concluded that switching from natural gas to hydrogen combustion results in changes in the flow field, flue gas composition, heat distribution and local peak temperatures within the flame. As a result, hydrogen combustion on existing natural gas burners can result in overheating of the burner tip, burner block and/or ceramic burner tube and can result in a substantial increase in the NO_x formation.

As discussed in Chapter 5, the development of LowNO_x hydrogen burners for the high temperature industry requires more insights (design rules) to what extend changes in the 1) residence time of combustion, 2) mixing rate of hydrogen and air and 3) mixing rate with flue gases with hydrogen and combustion air will influence the burner performance. Towards this end, a numerical study was performed from which the insights will be used to understand how much internal flue gas recirculation is required. Furthermore, the amounts of primary air in the fuel jet needed at different position in the flame is calculated to keep the (local) oxygen concentration- and the flame temperature low to suppress the thermal NO_x formation. To gain understanding how changes in the burner configuration such as the jet diameter, fuel pressure and jet speeds influence the amount of flue gas recirculation and mixing between hydrogen and combustion air a jet model is developed. Simulations for both natural gas and hydrogen are performed to get a better understanding in the differences between the flow field when using hydrogen and natural gas in burner systems. Based on the insights of the simulations of the models three preliminary designs for LowNO_x hydrogen nozzle mix burners for the high temperature industry are proposed for further study.

6.1 Effect flue gas recirculation and fuel staging on NO_x emission and temperature

For non-premix flames the fuel and air are mixed downstream the burner via diffusion and (turbulent) mixing, as shown in Figure 6.1 [1]. As a result, different fuel-to-air ratio areas exist in the flame jet. Furthermore, a part of the combusted products are entrained back in the fuel and air jet. The local flame temperature and hence the amount of NO formed depends on the amount of flue gases that are entrained in the fuel and air jets and at which the fuel-to-air ratio the combustion takes place. Before discussing the techniques to improve the entrainment of flue gases (internal flue gas recirculation) in the flame and to influence the local fuel-to-air ratio in the flame (e.g. staged combustion) the impact of flue gas recirculation and changing the fuel-to-air ratio on the NO_x formation is discussed based on performing 1-D numerical flame calculations. Based on these calculations, first insights and guidelines are derived for the amount of flue gas recirculation that is needed to lower the NO_x emission from a hydrogen flame below that of a natural gas flame. Furthermore, the impact of using fuel lean (excess of air) and fuel (excess of fuel) on the NO_x emission will give insight which conditions are optimal for lowering the NO_x emission.

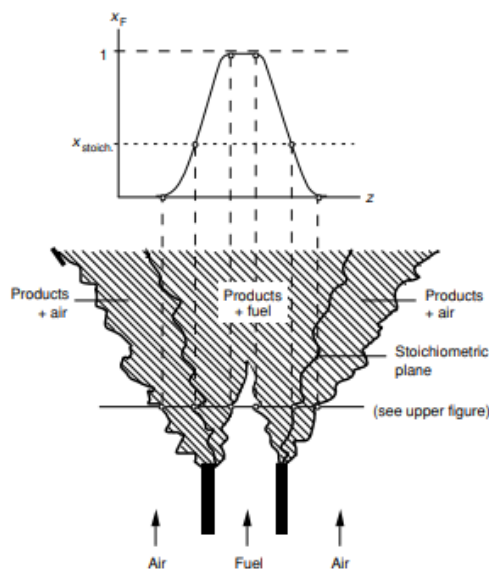


Figure 6.1. Schematic representation of a non-premixed jet flame [1]

6.1.1 Effect flue gas recirculation on the flame temperature and NO_x emission

To study the impact of flue gas recirculation on the NO_x formation 1-D numerical flame calculations have been performed for laminar premixed flames [2-4] for both natural gas and hydrogen. Here it is assumed that (worst-case) that combustion in non-premix flames take place in stoichiometric plane ($\phi=1$) as shown in Figure 6.1. For the flame calculations different percentage of flue gases in the total mixture are used as input and its impact on the flame temperature and NO_x formation is calculated. The calculations are based on recirculated flue gases consisting of N₂ and H₂O. Figure 6.2 shows that the temperature rise is much steeper for the hydrogen flame as a result of the much faster burning velocity in comparison to that of methane. Additionally, the adiabatic flame temperature of the hydrogen flame is substantially higher. The much faster burning velocity in combination with the higher flame temperature results in a steeper and faster calculated NO_x formation rate for the hydrogen flame as presented in Figure 6.2. As can be seen in Fig. 6.2 (left), the calculated flame temperature decreases when increasing the amount of flue gas recirculation. Additionally, the calculations reveal that flue gas recirculation also reduces the burning velocity which affects the residence time of combustion (see above). The combined effect of reduction in flame temperature and burning velocity results in lower NO_x production within the flame as shown in Figure 6.2 (Right). When about 7 mol% flue gases are present in the hydrogen/air gas mixture the NO_x formation rate is almost similar to that of methane. From this we conclude that about 7% flue gas recirculation is needed to lower the NO_x emissions from a hydrogen flame back to the NO_x emission of a natural gas flame.

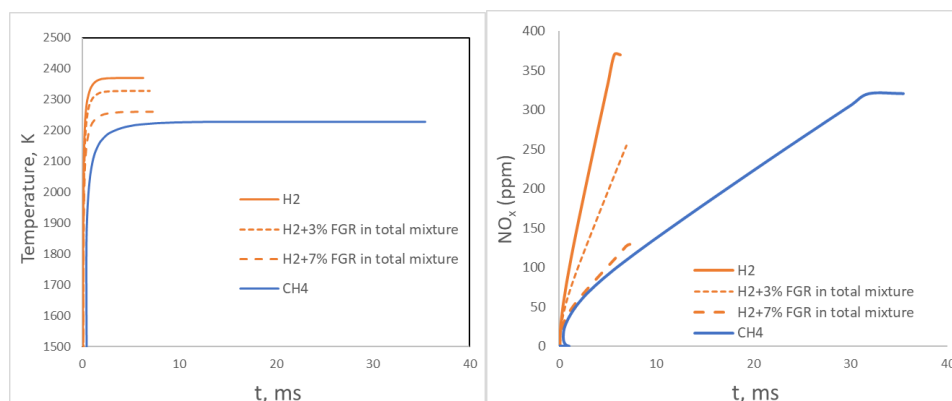


Figure 6.2. Calculated flame temperatures and NO_x emissions for a 1-D flame for different residence times and flue gas percentage in the total mixture (H₂/air/flue gas) assuming a flue gas temperature of 300K at stoichiometric conditions ($\lambda=1$)

To test the accuracy of the used method the results are compared with flue gas recirculation measurements in a 500 kW hot water boiler system using a forced draught burner system [5]. The tests show that about 6-7 mol% flue gases is required in H₂/air/flue gas mixture to lower the NO_x emission back to the NO_x emission measured in the flue gas when using natural gas. This result gives us confidence in the approach used for calculating the effect of flue gas recirculation on the NO_x emission.

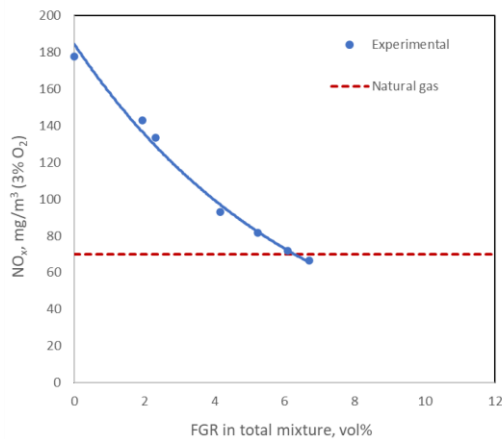


Figure 6.3. Measure NO_x emission for different percentage flue gases in the H₂/air/flue gas mixture

For high temperature direct heating processes, such as the ceramic industry the furnace and flue gas temperatures are substantially higher than used for indirect heating processes such as hot water production as shown above. When returning the hot flue gases to the cold unburned fuel/air mixture the initial temperature of the unburned mixture will rise and result in an increase in the adiabatic flue temperature. To estimate the effect of the flue gas temperature on the adiabatic flame temperature and the NO_x production numerical flame calculations have been performed by using as flue gas temperature 28, 500, 1000 and 1300 °C while keeping the amount of flue gases fixed to 7 mol% flue gases in the total mixture. As shown in Figure 6.4 the adiabatic flame temperature increases substantially with the flue gas temperature. Consequently, the effect of applying FGR on the NO_x reduction decreases when the flue gas temperature is higher which should be taken into account when designing low NO_x burners that make use of (internal) flue gas recirculation.

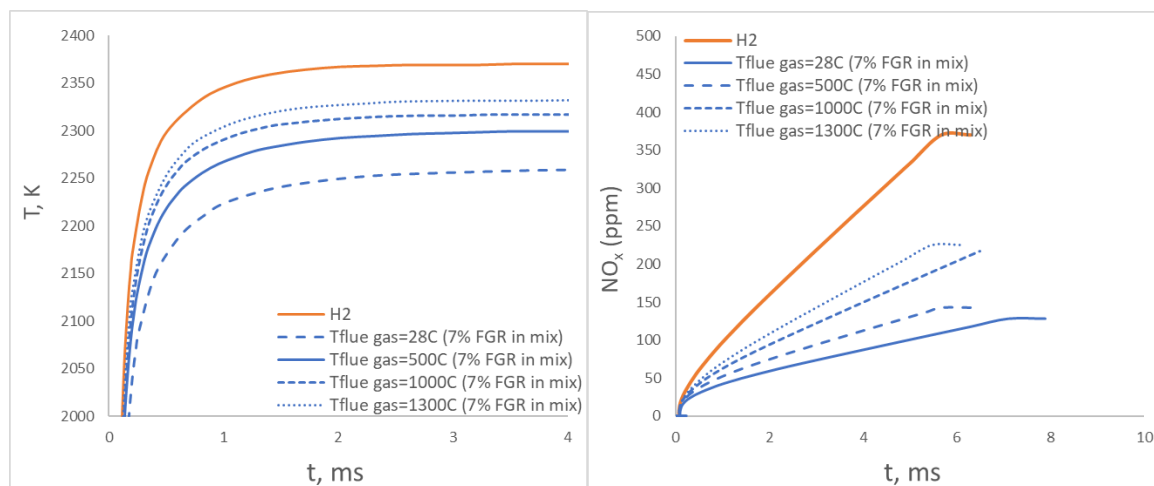


Figure 6.4. Calculated flame temperatures and NO_x emissions for a 1-D flame for different residence times and flue gas percentage in the total mixture (H₂/air/flue gas) assuming a flue gas temperature of 300K at stoichiometric conditions ($\lambda=1$)

To estimate the amount of flue gas recirculation needed for a furnace temperature of 1000 °C, which is a relevant temperature for the metal and ceramic industry numerical flame calculations were performed and presented in Figure 6.5. The results show that when about 16 mol% flue gases are present in the hydrogen/air mixture the NO_x emission is similar to the NO_x emission of methane.

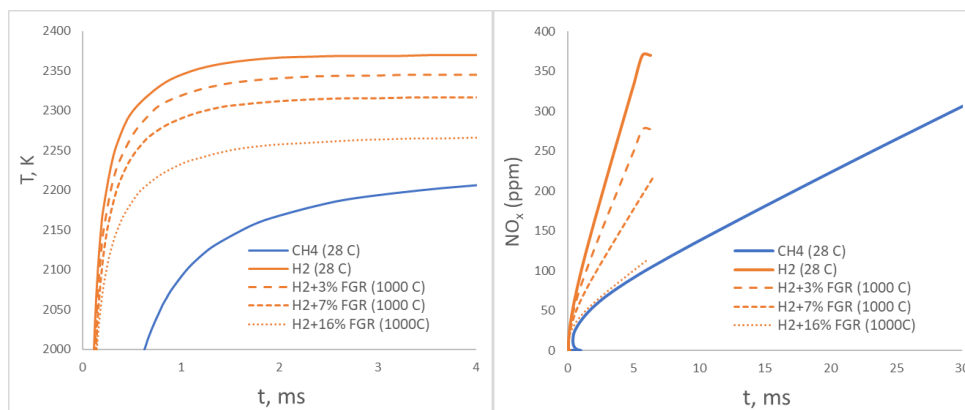


Figure 6.5. Calculated flame temperatures and NO_x emissions for a 1-D flame for different residence times and flue gas percentages in the total mixture (H₂/air/flue gas) using a flue gas temperature of 1000 °C and $\lambda=1$.

6.1.2 Effect fuel staging on the flame temperature and NO_x emission

Staged air combustion typically relies on slowing down the fuel-air mixing by supplying ‘small’ amounts of primary air at different position in the flame to the fuel in order to keep the local oxygen concentration- and the flame temperature low to suppress the thermal NO_x formation. To gain insights how the changes in equivalence ratio influence the flame temperature and NO_x emission 1-D numerical flame calculations have been performed at different air factors ranging from stoichiometric ($\phi=1$) to fuel rich (lack of air to complete the combustion, $\phi>1$) conditions.

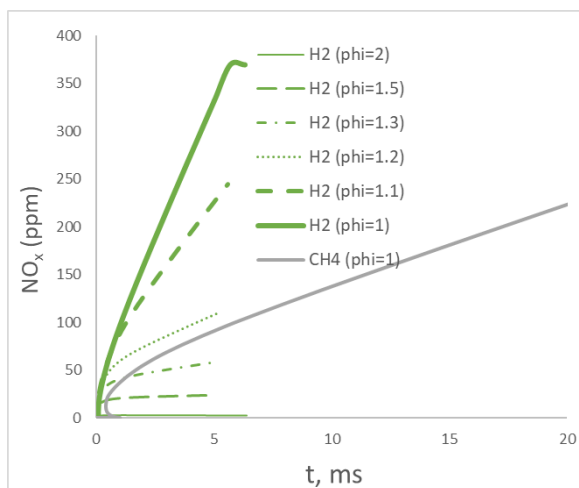


Figure 6.6. Calculated NO_x emissions for 1-D flames at different residence times and equivalence ratios (ϕ).

The results shown in Figure 6.6 show that increasing the equivalence ratio drastically reduces the NO_x formation. When increasing the equivalence ratio of the hydrogen/air mixture between $\phi=1.2$ -1.3 the NO_x formation is similar to that of methane. Here it should be noted that the NO_x formation for fuel rich hydrocarbon flames ($\phi>1$) differ from that of hydrogen flames. For example, for hydrocarbon flames the Fenimore mechanism ($\text{CH}+\text{N}_2=\text{HCN}+\text{N}_2$) is important for the NO formation in fuel rich flames while for hydrogen combustion other mechanisms are relevant such as the NNH and Zeldovich mechanism.

6.1.3 Conclusion and discussion:

Switching from methane to hydrogen combustion results in faster combustion (higher burning velocity) and higher adiabatic flame temperatures which causes an increase in the NO_x formation rate. In this study two NO_x mitigation strategies were numerically investigated 1) internal flue gas recirculation and 2) staged combustion. Both strategies are based on decreasing the burning velocity and flame temperature.

Applying internal flue gas recirculation

When applying internal flue gas recirculation a part of the flue gases present in the furnace are recirculated back to the fuel and/or air jet. The amount of flue gases that are entrained into the jet depends upon the burner design. As input for the burner design the effect of flue gases recirculation is addressed by performing numerical flame calculations:

- The numerical flame calculations performed in this study show that diluting the H₂/air mixture with flue gases (N₂ and H₂O) effectively decreases the burning rate and flame temperature of a hydrogen flame and consequently reduces the NO_x formation rate.
- The amount of (internal) flue gas recirculation needed to suppress the NO_x formation back to the NO_x production rate of a methane flame depends upon the flue gas temperature. When the flue gas temperatures increases, for instance at high temperature applications more flue gas recirculation is needed to lower the NO_x production rate, which should be taken into account for the design of the burner.
- As an example, in the table presented below the calculated amount of flue gas needed in the hydrogen/air blend to having the same NO_x emission levels as when using methane is calculated for a flue gas temperature of 28 °C and 1000 °C. The results show that about 7% flue gas needs to be present in the H₂/air mixture for low temperature processes, such as hot water boiler and about 16% is needed for high temperature processes with a furnace temperature of 1000 °C. The results are in good agreement with measurements performed in a boiler system.

Table 6.1. Calculated amount of flue gas at different flue gas temperatures

| Flue gas temperature, °C | Calculated amount of flue gas, % | Measured amount of flue gas, % |
|--------------------------|----------------------------------|--------------------------------|
| 28 | 7 | 6-7 |
| 1000 | 16 | - |

Staged combustion

Staged combustion can effectively suppress the NO_x formation by delaying the mixing of air and fuel which creates fuel rich zones.

- Calculations show that when burning H₂/air mixtures with an equivalence ratio between $\phi=1.2-1.3$ 'similar' NO_x production levels are obtained as when burning natural gas.
- From this result it can be concluded that when designing LowNO_x hydrogen burners the equivalence ratio should be higher than $\phi>1.2$ for each staged but lower than the equivalence ratio of the upper flammability limit (UFL) of $\phi<6.9$.

From the results obtained it is recommended to develop a low NO_x burner that combines staged combustion with internal flue gas recirculation. Furthermore, the design should enable to flexibly control the amount of flue gas recirculation and the local equivalence ratio when using staged combustion.

6.2 Jet model

As base design a pipe-in-pipe burner is used. The fuel and/or air is supplied via nozzles creating a fuel (and air) jet downstream of the burner tip. Further downstream mixing takes place due to the differences in mass flux between the jet and its surroundings.

To get insights into the mixing of flue gas, air and fuel downstream of the burner while changing design parameters such as the nozzle size and the gas velocity a jet model was developed. The model is based on a jet model as developed by Naber and Siebers [6] and Musculus and Kattke [7]. As the gas leaves the burner nozzle, the gas expands and a low-pressure region is created downstream of the flow, which causes the surrounding gas to be pulled in. The model developed in this study is capable of simulating different burner configurations (nozzle diameter, back pressure, multiple fuel and air jets) with the aim to reduce the NO_x emissions.

6.2.1 Brief description jet model

For simplicity, the model will be described for a single fuel jet (in air). It should be noted that the model is capable of simulating multiple fuel and air jets, all based on the same principle as the single jet described here.

In the jet model, the jet is treated as a cone resting in the nozzle as shown in Fig. 6.7.

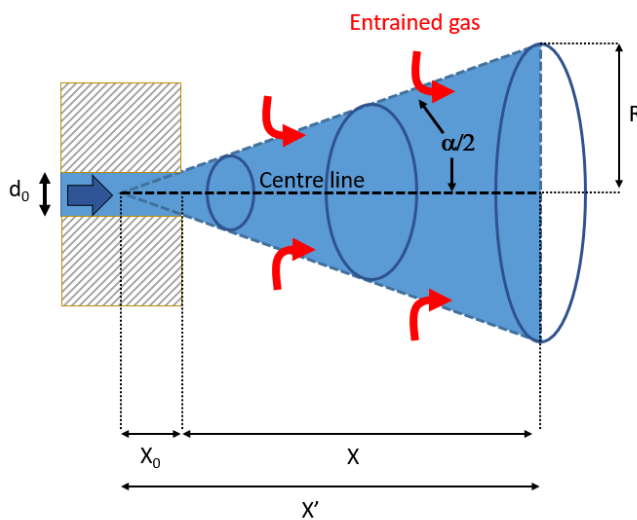


Figure 6.7. Basic principle of dispersion.

The expansion of the fuel leaving the nozzle is determined by the jet angle, which depends on the fuel characteristics (e.g. density, viscosity, and so on) and the nozzle diameter (d_0). In this study the jet angle is calculated using the model from Arai et al. [8]:

$$\theta = 0.025 \left(\frac{\rho_{surroundings} d_{spray}^2 (\rho_{spray} v_{spray}^2 - \rho_{surrounding} v_{surrounding}^2)}{2 \mu_{surrounding}} \right)^{0.25}$$

[degree] (1)

Where ρ_{jet} and $\rho_{surrounding}$ are the jet (e.g. fuel) and surroundings (e.g. air and flue gases) density [kg/m³], respectively, d_{jet} is the diameter of the jet nozzle [m] and $\mu_{surrounding}$ is the (temperature dependent) dynamic viscosity of the surrounding gas [kg/ms]. An expression for the jet velocity (v_{spray}) can be derived using the Bernoulli equation by assuming adiabatic flow of an ideal gas;

$$v_{spray} = \sqrt{\frac{2P_1}{\rho_f} \frac{k}{k+1} \left(1 - \left(\frac{P_2}{P_1}\right)^{\frac{k-1}{k}}\right)} \quad [\text{m/s}] \quad (2)$$

Where P_1 is the supply pressure (P_a) of the fuel or air, P_2 is the pressure in the furnace and k is ratio of the heat capacities: C_p/C_v . An expression for the surrounding gases ($v_{surrounding}$) can be found by taken into account the fuel-to-air ratio settings of the burner:

$$v_{surroundings} = \frac{0.25\pi d_{spray}^2 v_{spray} \cdot AIR}{0.25\pi d_{surroundings}^2 \varphi} \quad [\text{m/s}] \quad (2a)$$

Where AIR is the air requirement and $d_{surroundings}$ is the diameter of the nozzle for the surrounding gas [m]. If no nozzle is present for the surrounding gas then the surrounding velocity is taken as zero.

If the jet consists of the fuel and the velocity is known, the burner power can be calculated using the following equation;

$$P_{burner} = 250\pi d_0^2 v_{spray} H_i \quad [\text{kW}] \quad (3)$$

Where H_i is the net calorific value [MJ/m³] of the fuel.

In the model at each distance x from the nozzle, the jet volume fraction at the center line (X_{sprayc}) is calculated as follows:

$$X_{sprayc} = \frac{d_0}{2R} = \frac{d_0}{2 \tan\left(\frac{\theta}{2}\right) x'} \quad (4)$$

The radial profiles within the jet are given by

$$\frac{X_{spray(r)}}{X_{sprayc}} = \left(1 - \left(\frac{r}{R}\right)^\alpha\right)^2 \quad (5)$$

Where $X_{spray(r)}$ is the mole fraction at radial position r and R is the width of the jet given by

$$R = \tan\left(\frac{\theta}{2}\right) X' \quad (6)$$

Evaluation of the value of α reveals that this can be set at 1.5 [ref] shortly downstream of the nozzle corresponding to a fully developed jet [ref].

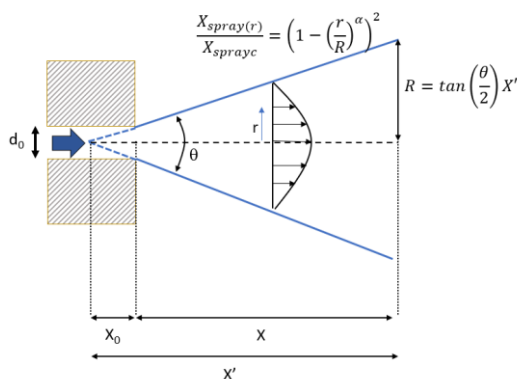


Figure 6.8. Schematic of axial and radial fuel fraction distribution

As an example, Figures 6.9 and 6.10 show the model results for a methane jet in air at different distances from the nozzle exit (X in Fig 7.8). At further distances from the nozzle, the fuel fraction lowers due to entrainment of the air into the methane spray. Consequently, the equivalence ratio

drops as more air is entrained. Mixing of air and fuel does not only takes place in axial direction, but also in radian direction as can be seen in Figure 6.10. The lefthand side of Figure 6.10 shows a steep decrease in fuel fraction and equivalence ratio at a distance close to the nozzle exit, whereas further away from the nozzle, the mixing of air and methane proceeds more gradually.

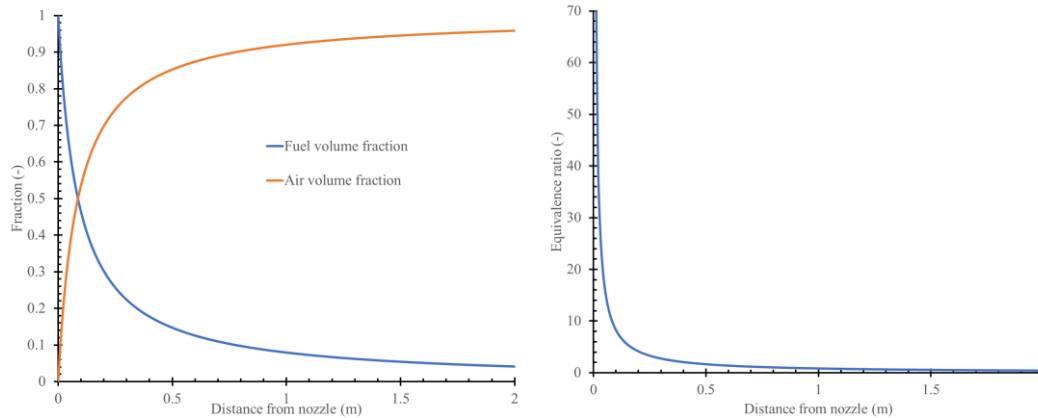


Figure 6.9. Axial fuel (methane) and air volume profiles for fuel jet in air ($\Delta p=2.7$ mbar, 261 kW, fuel and air at 298.15K and 1 atm) and equivalence ratio as function of the distance from the nozzle exit (distance X' in Figure 6.8).

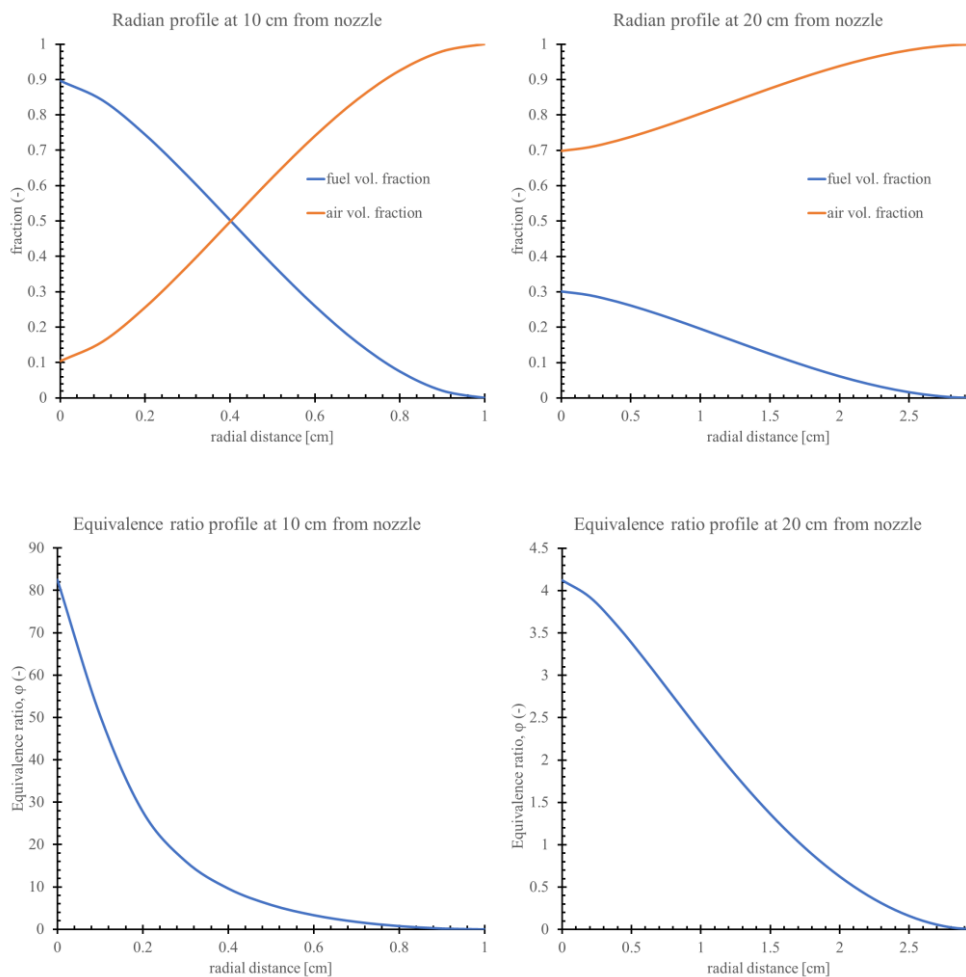


Figure 6.10. Radian profiles of fuel and air volume fractions at 0.1 m and 0.2 m from the nozzle exit (see also Fig. 6.9).

For multiple jets, the fuel and air volume fractions of each jet is individually calculated while taking into account the possibility of overlapping sprays as illustrated in the Figure below. The jet model is used to get insight how changing the design parameters will influence the mixing.

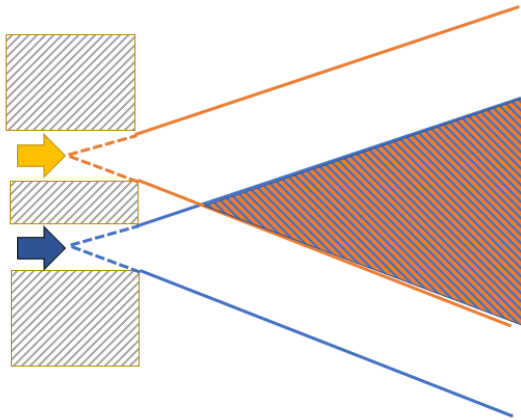


Figure 6.11: Schematic overview of a multiple spray

6.3 Proposed burner designs for low NO_x hydrogen HT burner

Based on the results of the literature study and preliminary calculations, three low NO_x burner designs concepts are proposed with a burner power of 250 kW. The three designs (A, B and C) are pipe-in-pipe burners and are shown in the Figure 6.11 below. All three burner designs are aimed at enhancing internal flue gas recirculation to reduce the NO_x emission. The first design (A in Figure 6.11) is a pipe-in-pipe burner with a central fuel jet surround by an air jet. In this design the flue gases are entrained by a venturi. Design B uses a swirl in the air to 1) improve mixing with the fuel and 2) improve mixing with the flue gasses from the surroundings. To create additional internal flue gas recirculation the swirl burner design is equipped with a venturi. Finally, design C uses the principle of staged combustion in combination with internal flue gas recirculation. In general, the burner designs have flexible configuration (nozzle diameter, back pressure, multiple fuel and air jets) to study the optimum settings for reducing the NO_x emission. Furthermore, the burner heads were designed is such a way that they are interchangeable. For example, the burner head for burner design A can be replaced by burner head design B. The next sections will discuss each burner design.

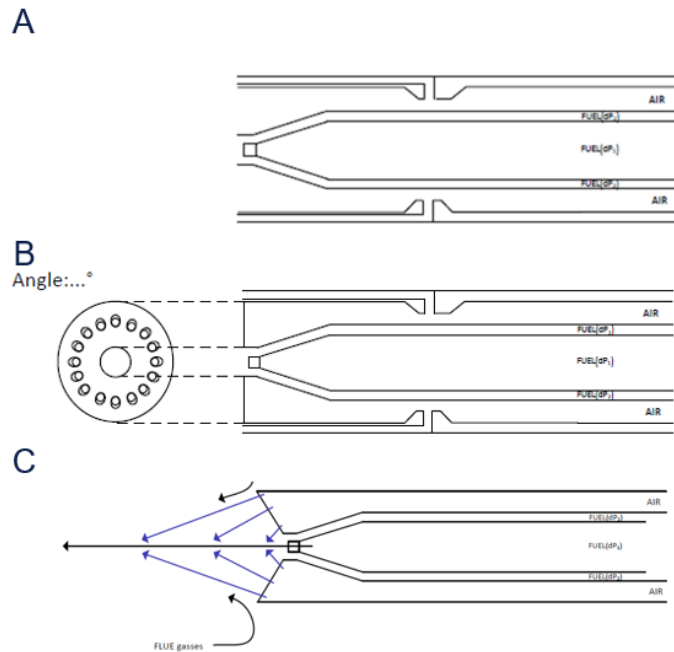


Figure 6.11. Three proposed burner designs (A, B and c) for a low NO_x hydrogen industrial burner

6.4 Burner design A

This chapter discusses burner design A which is a commonly used pipe-in-pipe burner with a central fuel flow surrounded by an air co-flow. The aim of the design is to create a low NO_x hydrogen burner by applying internal flue gas recirculation.

6.4.1 Effect changing fuel nozzle diameter on the hydrogen jet speed and jet composition distribution

To determine the optimal fuel and air nozzle size diameters for a 250 kW burner as illustrated in Figure 6.12 with an equivalence ratio of $\phi=0.9$ the effect of changing the fuel nozzle diameter on the mixing of fuel and air downstream the nozzle is calculated using the jet model. For the calculations an existing natural gas pipe-in-pipe burner with a fuel nozzle diameter of 18 mm and an air velocity of 9 m/s is used as starting point.

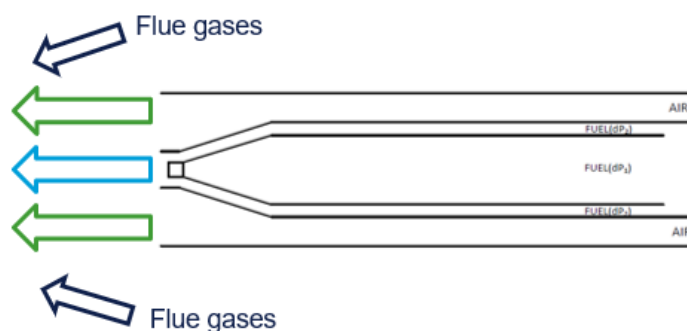


Figure 6.12. Illustration of air flow (green arrow), fuel flow (blue arrow) and flue gas flow (dark blue arrow) used for the jet model simulations.

To calculate the effect of changing the fuel nozzle diameter on the fuel jet speed leaving the nozzle, calculations in the jet model were performed at a constant air velocity for hydrogen while keeping the burner power constant at 250 kW. Figure 6.13 shows that decreasing the diameter of the fuel lance, for example from 18 to 15 mm, results in a strong increase in the needed fuel velocity. A further decrease to smaller diameters results in very high velocities and possible flame blow-off. To increase the velocity the pressure drop across the fuel nozzle is increased as shown in Figure 6.13. For fuel nozzle diameters larger than 25 mm the fuel- and air momentum flux are getting close to each other as shown in Figure 6.13, resulting in less mixing. Therefore, the nozzle diameter should be roughly between 15 and 25 mm to create sufficient mixing between fuel and air and prevent flame blow-off.

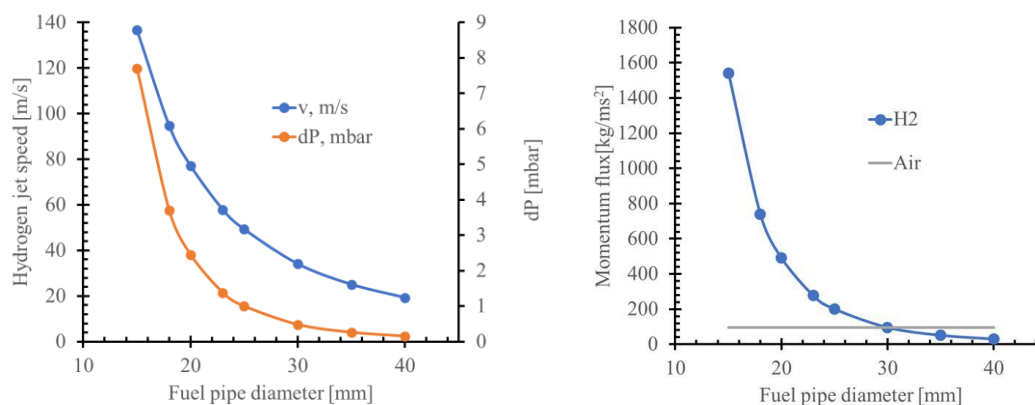


Figure 6.13. Calculated hydrogen jet velocities (left) and momentum (right) for hydrogen-air mixtures (250kW, $\phi=0.9$)

One of the differences when using hydrogen in a natural gas burner while keeping the power constant, is the increase of the hydrogen jet speed with about a factor of three. The combination of the change in fuel velocity and density of the fuel results in a change in momentum flux as shown in Figure 6.16. Furthermore, the flammability limits and the burning velocity differences between methane and hydrogen affect the flame structure of both flames. To get insights how the changes between the chemical and physical properties influence the mixing between air and fuel, calculations in the jet model were performed and presented in Figure 6.14. Furthermore, the effect of different nozzle diameters on the gas composition along the jet for a methane-air and hydrogen-air have been calculated using the jet model. The simulations in the jet model are performed for the situation that the air flow is surrounded by flue gas as illustrated in the Figure 6.12.

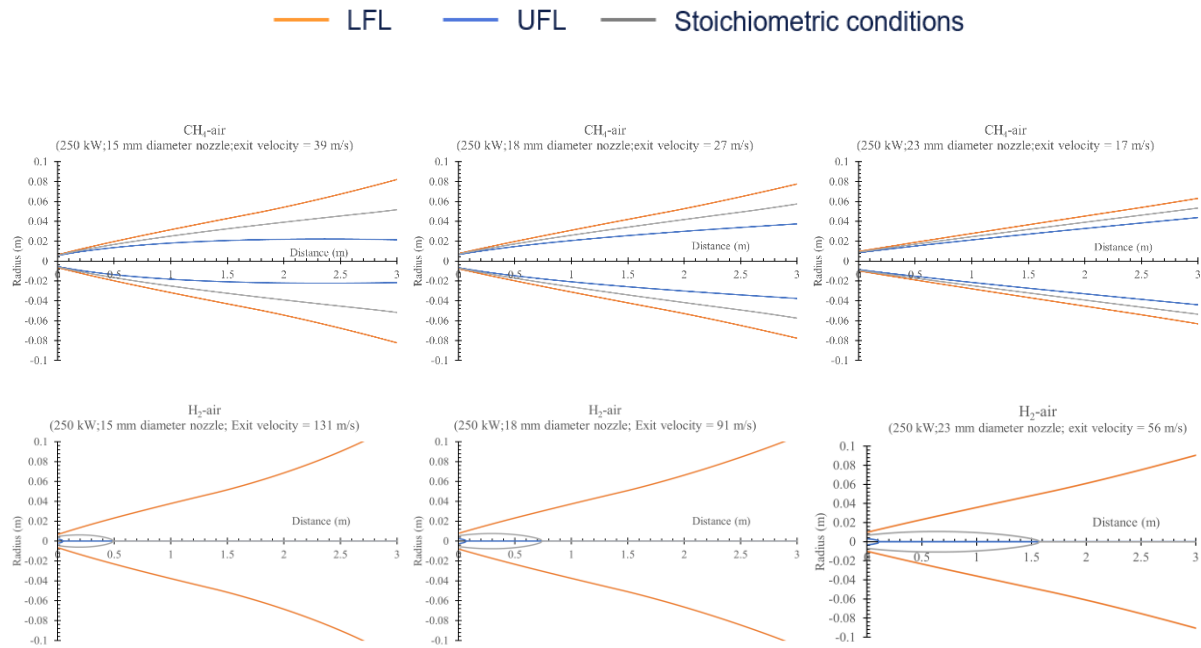


Figure 6.14. Calculated flammability limits (LFL and UFL) and stoichiometric ratio between fuel and air based on the composition distribution along the spray.

Figure 6.14 shows the results of the calculations, where the flammability limits and stoichiometric ratio contours along the jet for three nozzle diameters (15 mm, 18 mm and 23 mm) are shown. The jet model calculates for each position along the jet (X in Fig. 6.8) the radial position (r in Fig. 6.8) where the equivalence ratio of the fuel-air mixture corresponds to the values for the flammability limits (UFL and LFL) or the stoichiometric ratio (see also Table below). In the area between the UFL and the LFL, a combustible mixture of fuel and air (and flue gas) exists.

Table 6.2. Fuel to air ratios for the flammability limits and stoichiometric ratios for methane and hydrogen

| | H ₂ | CH ₄ |
|---------------------------------|----------------|-----------------|
| Upper flammability level | | |
| Equivalence ratio (ϕ) | 0.10 | 0.50 |
| Adiabatic flame temp. [K] | 632 | 1480 |
| Lower flammability level | | |
| Equivalence ratio (ϕ) | 6.88 | 1.65 |
| Adiabatic flame temp. [K] | 1191 | 1797 |
| Stoichiometric ratio | | |
| Equivalence ratio (ϕ) | 1 | 1 |
| Adiabatic flame temp. [K] | 2381 | 2226 |

Figure 6.15 shows that increasing the nozzle diameter results in lower (initial) fuel jet speeds at the nozzle for both methane-air and hydrogen-air mixtures which results in less mixing with the surroundings. After leaving the nozzle, the fuel expands (see e.g. Figure 6.8) and as a result the fuel jet speed decreases further downstream of the nozzle. Also, to maintain the same burner output

(250 kW), the hydrogen fuel jet speed at the nozzle needs to be approximately 3 times larger as compared to the fuel velocity for methane to compensate for the difference in heating value.

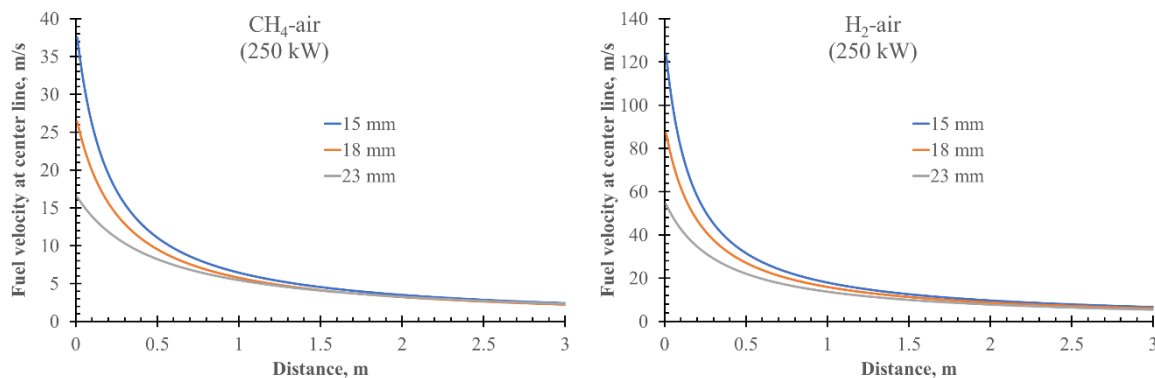


Figure 6.15 Fuel velocities at center line for methane (left) and hydrogen (right).

As a consequence of the lowering the fuel jet speed by increasing the internal diameter, the momentum fluxes of air and fuel approach each other as illustrated in Figure 6.16. Since there is no mixing between air and fuel when the momentum fluxes are equal, the results indicate that the increase of the internal diameter results in slower mixing as also shown in Figure 6.14.

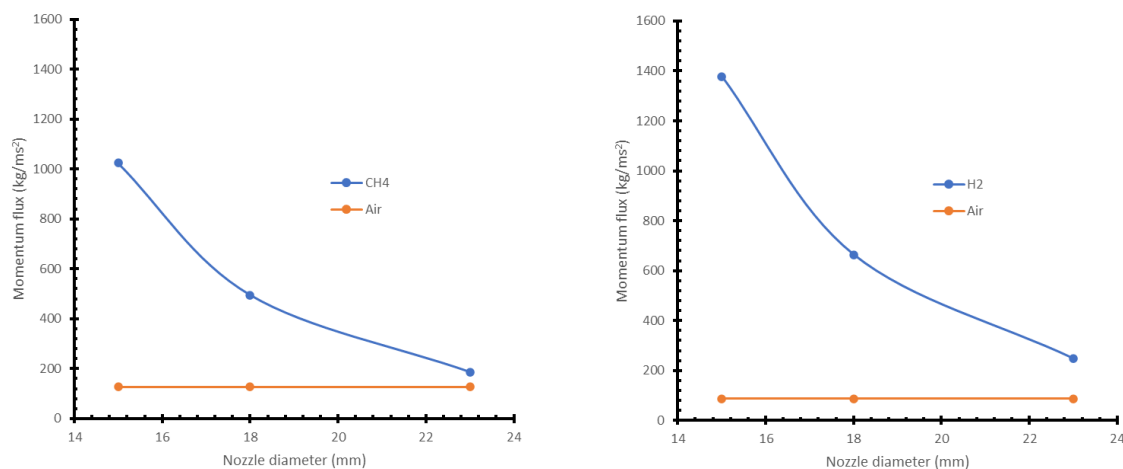


Figure 6.16. Calculated momentum flux for methane-air (left) and hydrogen-air (right) as function of the nozzle diameter (250 kW, $\phi=0.9$).

Furthermore, the results in Figure 6.14 shows that the envelope at which combustion can take place (between the LFL and the UFL contours in Figure 6.14) is much larger for hydrogen. Assuming that combustion primarily takes place in the stoichiometric plane, the results in Figure 6.14 indicate that the flame is positioned much closer to the fuel exit when using hydrogen as a fuel which is in agreement with the observation in the literature, see for example Figure 4.13. This result suggests high temperatures and NO_x emission are present already close to the burner tip which is expected to slow down the internal flue gas recirculation needed to cool the flame. Increasing the fuel jet speed in an effort to move the hydrogen flame away from the burner surface, does not result in significantly delaying the combustion, as indicated by Figures 6.14-15. In fact, at higher fuel jet speeds mixing occurs faster due to the larger momentum flux difference between hydrogen and air resulting in a flame located nearer the burner surface.

In general, the results show that hydrogen combustion will occur much faster than methane combustion which allows larger parameters variations, such as jet velocities than for methane flames. Under the conditions used in the jet model we expect that the methane flame will blow-off since the flame velocity is expected to be lower than the exit velocities especially at the fuel lean and rich parts of the flame. However, for hydrogen which has a factor of 8 higher burning velocity as compared to methane, the stoichiometric condition where the highest flame speed is reached is already close to the burner tip. Based on this result, it is expected that hydrogen as compared to methane will result in much more stable combustion under identical conditions which allows potentially more flue gas recirculation in hydrogen flames. This was also observed during the experiments performed at DNV using an industrial pipe-in-pipe burner with an 18 mm fuel diameter and similar conditions (fuel and air flows) as in the simulations. Additionally, experiments were performed at DNV using an in-house fabricated pipe-in-pipe burner with a configuration similar to the one used in the model. In case methane was used as a fuel the flame blew off at powers above 3 kW, while the hydrogen flame was capable of producing a stable flame at higher powers (see images below).



Figure 6.17. Tested pipe-in-pipe burner

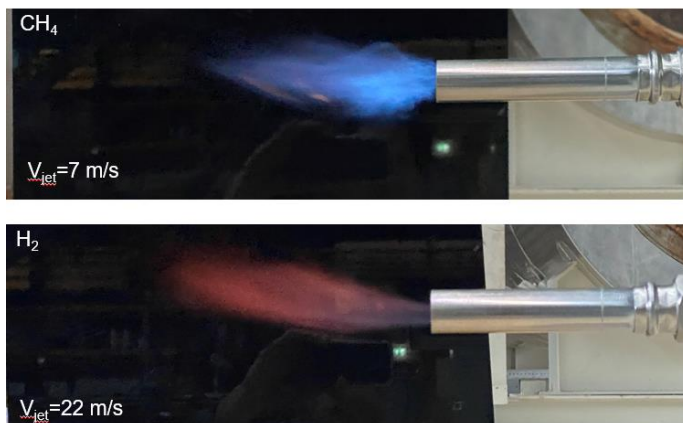


Figure 6.18. Difference in combustion between hydrogen and methane at 3 kW at $\phi=0.9$

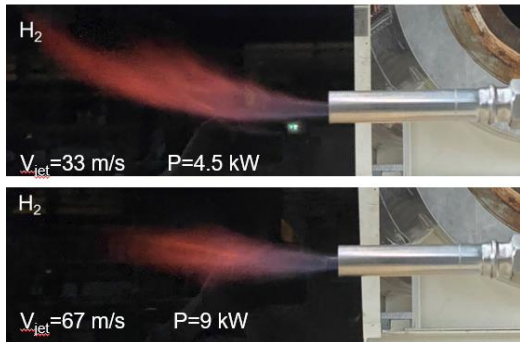


Figure 6.19. effect increasing hydrogen jet velocity at $\phi=0.9$

The experiments using the pipe-in-pipe burner (see Fig 6.17-19) and the simulation results of the jet model indicate that it is difficult to slow down the hydrogen combustion by changing the fuel jet speed. An effective way of slowing down the hydrogen combustion rate and the NO_x formation rate is to entrain more flue gases. To create more internal flue gas recirculation in the base design A, an alternative design was proposed as illustrated in Figure 6.20.

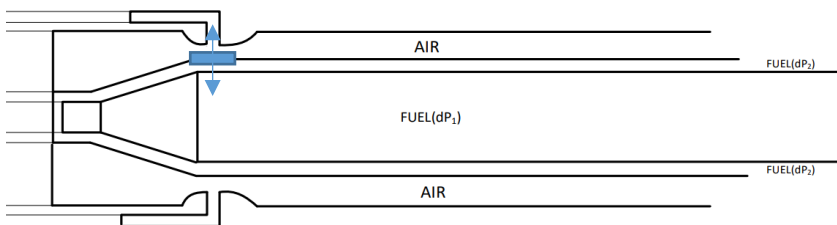


Figure 6.20. Cross section of burner design with venturi in the air section.

In the above Figure a constricted section is placed in the air section, which forces the air velocity to increase when passing through the constriction. As a consequence of the venturi effect, an under pressure is created in the constriction (venturi) which entrains the flue gases from the outside via four flue gas pipes.

To determine the parameters for the venturi in the air section, shown in Figure 6.20, the effect of the flue gas pipe diameter on the amount of flue gas entrained in the air was calculated. The calculations were performed assuming 250 kW burner power, an air outer diameter of 85 mm and a furnace temperature of 1000°C. The results shown in Fig. 6.21 reveal that a diameter of approximately 15 mm for the four flue gas pipes is required to achieve 16% flue gases in the air mixture using a venturi diameter of 72 mm.

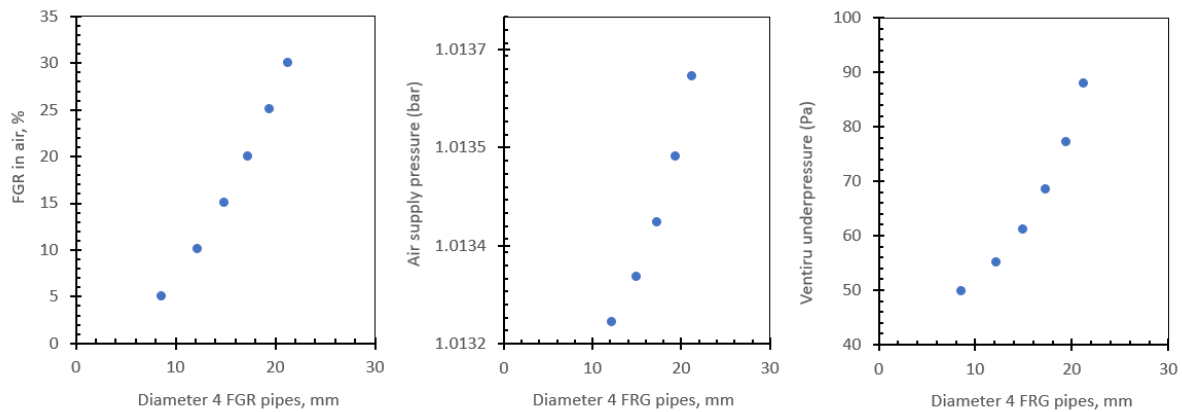


Figure 6.21. Effect of the diameter of the flue gas pipe on the flue gas percentage in air (left), air supply required (middle) and on the pressure in the constriction (venturi) having a diameter of 71 mm.

A smaller flue gas pipe diameter can be used to achieve 16% flue gas in air if the venturi diameter is also reduced as shown in Figure 6.22 to create more under pressure in the venturi throat. However, this also requires a higher air supply pressure as shown in the right-hand side of Fig. 6.22.

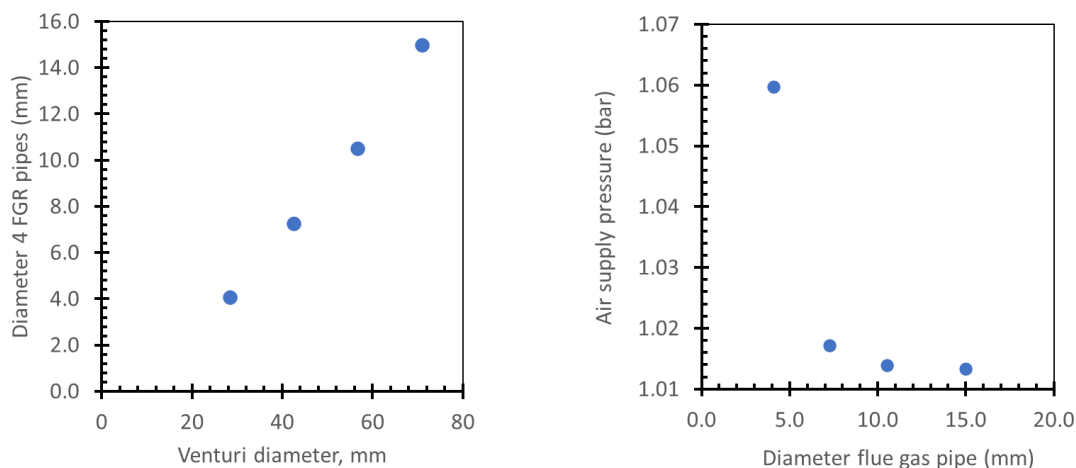


Figure 6.22. Effect of changing the diameter of the flue gas pipe on venturi diameter (left) and the air supply pressure (right) for a fixed amount of flue gases (16%).

In general, the results of the calculations show that the new design should allow for sufficient flue gasses to be entrained in the air to reduce the NO_x emissions. An example for the design parameters to create 16 % flue gas recirculation are shown in the Table 6.3 and the burner characteristics are shown in Table 6.4. The burner will be designed with an adjustable configuration to change the venturi diameter and the flue gas diameter independently from each other. Changing the flue gas diameter can be done by using removable flue gas pipes with different diameters. Changing the venturi size can be done by placing inserts on the fuel lance (see Figure 6.20 with different sizes to create a larger or smaller restriction).

Table 6.3. Example design parameters for creating flue gas recirculation

| Fuel nozzle diameter, mm | Flue gas diameter, mm | Air nozzle outer diameter, mm | Venturi diameter, mm | Air supply pressure before venturi, bar |
|--------------------------|-----------------------|-------------------------------|----------------------|---|
| 18 | 4 | 85 | 28 | 1.06 |

Table 6.4. The burner characteristics

| Power, kW | Air velocity, m/s | H ₂ jet speed leaving nozzle, m/s | Flue gas entrainment in total mixture, mole% |
|-----------|-------------------|--|--|
| 250 | 9 | 91 | 16 |

6.5 Burner design B

The literature study identified swirling burners as a possible option to reduce the NO_x. In this burner design, guiding vanes are installed on the air side to give a swirling motion to the combustion air. This motion creates a recirculation zone between the air and fuel which helps to sustain the flame.

Additionally, the swirling motion helps accelerate the process of mixing internal flue gases with air (and fuel) to reduce the NO_x. The angle of the vanes determines the amount of swirling. The design of the burner allows different burner heads to be installed prior to starting the experiments. For this reason, different burner heads will be designed with a (different) fixed angle of the vanes to study the effect of swirling on the NO_x emission. The swirl number characterizes the swirl and is defined as the ratio of angular momentum to the axial momentum. Typical swirl numbers range between 0.3-2. A swirl number of 0.3 creates moderate swirl, 0.6 creates considerable swirl, a swirl number of 1 creates high swirl and a swirl number of 2 gives very high swirl. To create moderate swirl the angle is about 25° and to create very high swirl a 75 ° angle is required based on calculations [9].

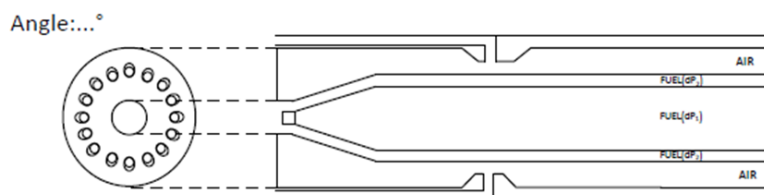


Figure 6.23. Cross section of burner design with a swirl burner head.

To create additional flue gas recirculation the burner will be equipped with the venturi design.

6.6 Burner design C

As shown in Figure 6.14 the mixing of hydrogen and air by using a co-flow creates a flammable region and a high temperature zone near the burner tip. Consequently, the NO_x production rate is high when not enough flue gas recirculation is applied. To make use of the fast combustion of hydrogen and the wide flammability range the concept of staged combustion is applied to minimize the NO_x emissions (see also above) while maintaining stable combustion. Figure 6.24 illustrated the principle of this burner design.

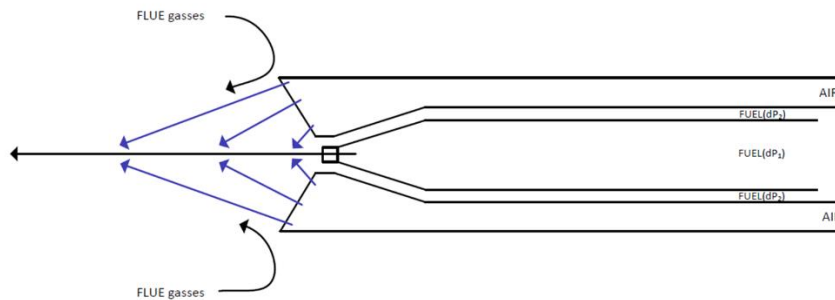


Figure 6.24. Principle of burner design C (staged combustion)

The working principle of this burner is that air is introduced at different positions in the fuel flow. The first portion of air encounters the fuel at fuel rich conditions (low NO_x), while air that is injected further downstream encounters the fuel at nearly stoichiometric conditions and high flame temperatures. Under these conditions, NO_x production may be high, but as the air is injected further downstream of the fuel more flue gas is entrained reducing the flame temperature and hence the NO_x formation. Additionally, this concept may produce longer flames that typically have low peak flame temperatures.

In this burner design, the air nozzles are located at 12 positions as illustrated in Figure 6.25. To illustrate the effect of flue gas entrainment and air injection on the flame temperature along the center fuel line, jet calculations were performed using the jet model for a 200-kW burner and a fuel nozzle diameter of 18 mm. Three air nozzle diameters of 20 (d_1), 25 (d_2) and 30 (d_3) mm were considered for the air nozzles (see also Figure below).

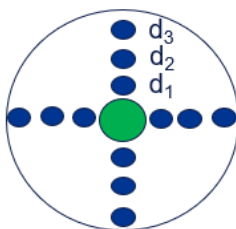


Figure 6.25. Schematic of air and fuel nozzle positions (viewed in front of the burner). Position of air nozzles indicated by blue circles; the fuel nozzle is indicated by the green circle.

As illustrated in Figures 6.24-6.26, the first nozzle (d_1) is located close to the fuel nozzle exit and injecting air in the fuel jet will result in a local fuel-rich mixture, while the other nozzles d_2 and d_3 are located further downstream of the fuel flow. It is expected that for the first nozzle (d_1) no flue gas is

entrained due to the short jet length and its close location to the fuel nozzle. For air coming from the other nozzles (d_2 and d_3), the jet length is longer as compared to nozzle d_1 , and the nozzles are located further away from the fuel nozzle. This allows for more flue gas to be entrained in the air jet when it arrives at the center fuel line. To calculate the amount of flue gas entrainment in the air jet the jet length for air coming from nozzles d_2 and d_3 is calculated using the geometry equations shown in Figure 6.25. Next, for each jet length the jet model calculates the amount of flue gases entrained in air for each nozzle and the volume flows of air, flue gas and fuel at the center line. Based on this information, the equivalence ratio and the effect of flue gas entrainment on the adiabatic flame temperature was calculated within the spray. The calculations were performed for different nozzle sizes and jet lengths and the most optimal burner design parameters are presented in Table 6.5. The results show that the calculated equivalence ratios fall within the flammability values (φ between 0.1 and 6.88), hence a combustible mixture is formed for each air injection. Also, the results in Table 6.5 show that flue gas entrained results in a significant reduction in flame temperature and hence lower NO_x emissions. The addition of a venturi in the air section is expected to increase even further the amount of flue gases entrained in the air flow, and thus decreasing the NO_x values to even lower values. Therefore, the burner head can be attached to venturi section as shown in Figure 6.20.

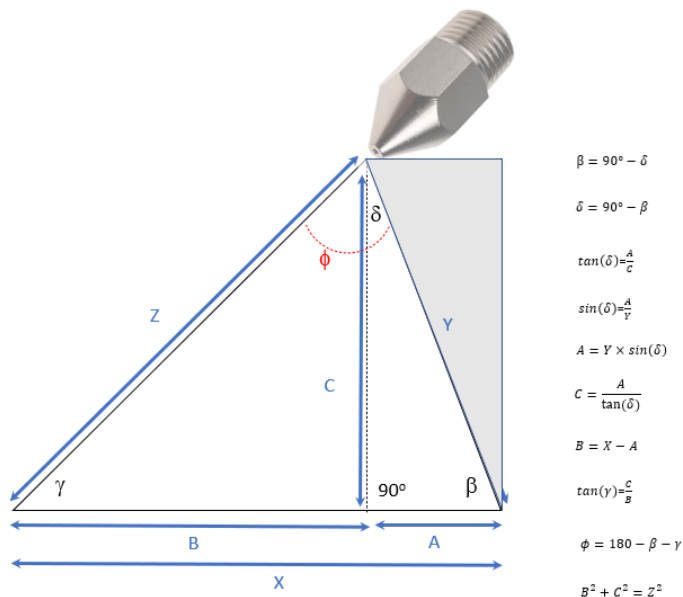


Figure 6.26. Schematic of jet calculations

Table 6.5. Results of jet calculations for air nozzles d₁, d₂ and d₃.

| | d1 | d2 | d3 |
|------------------------------------|-----------|-----------|-----------|
| Number of nozzles | 4 | 4 | 4 |
| Diameter, mm | 20 | 25 | 30 |
| Air velocity, m/s | 8 | 8 | 8 |
| δ (with plane), ° | 30 | 30 | 30 |
| φ (on centre line fuel spray), ° | 68 | 114 | 114 |
| Y, mm | 17 | 30 | 45 |
| X, mm | 24 | 250 | 400 |
| Z (jet length), mm | 19 | 236 | 380 |
| % FGR in air | 0% | 15% | 23% |
| Phi (mix) | 4.6 | 1.8 | 1.0 |
| Adiabatic flame temperature | | | |
| Without FGR, K | 1461 | 2131 | 2347 |
| With FGR, K | 1461 | 1954 | 2018 |

6.7 Summary & outlook

This study shows that internal flue gas recirculation and staged combustion can be effectively applied to reduce the NO_x emission from hydrogen combustion in high temperature industrial burner. Three different pipe-in-pipe burner designs have been proposed and evaluated by using numerical simulations and an in-house developed burner jet model. The results indicate that when using hydrogen as a fuel the combustion is fast and is creating high temperatures near the burner head. To slow down the combustion, internal flue gas recirculation is enhanced by applying a venturi in the air supply section of the pipe-in-pipe burner designs. Additionally, to further improve internal flue gas circulation a swirl burner design and a staged combustion burner are proposed to create a fuel rich combustion zone to suppress NO_x formation.

The basic design is shown in Figure 6.27 (design A) and consists of a central fuel flow surrounded by an air flow. In this flexible burner design the burner head can be replaced by the swirl burner head (design B) or a staged combustion burner head (design C). Additionally, the burner has an adjustable configuration (nozzle diameter, back pressure, multiple fuel and air jets) with the aim to find the optimum configuration for the NO_x emissions reduction.

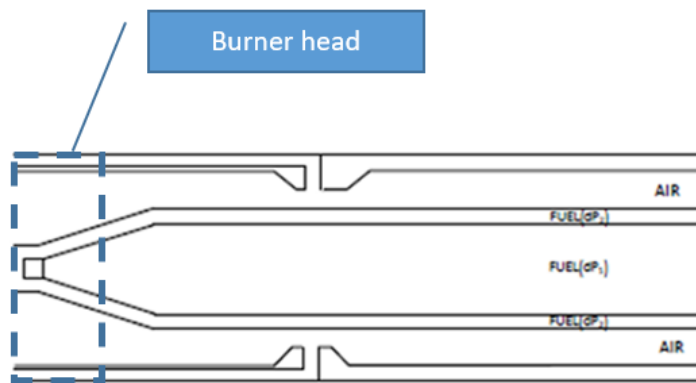
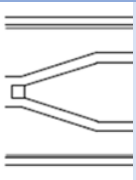
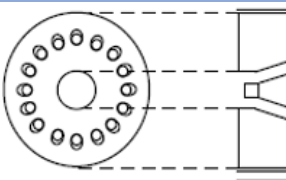
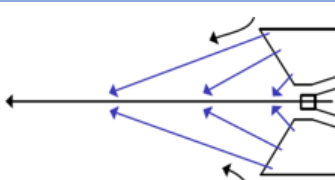


Figure 6.27. Basic design for a low-NO_x high temperature hydrogen pipe-in-pipe burner.

Based on the results found in this study, the following design parameters shown in Table 6.6 are proposed in support of the development of the low-NO_x high temperature hydrogen burner system.

Table 6.6. Burner designs and design parameters for a 250-kW burner with an equivalence ratio $\phi=0.9$

| | Design A (basis) | Design B (swirl) | Design C (staged combustion) |
|-------------------------------------|---|--|---|
| |  |  |  |
| Number of nozzles | | | |
| Air | 1 | To be determined | 4 x 3 |
| Fuel | 1 | 1 | 1 |
| Nozzle diameter [mm] | | | |
| Air | 85 | - | 20, 25 and 30 |
| Fuel | 15-23 | 15-23 | 15 - 23 |
| Jet speed range [m/s] | | | |
| Air | 8-12 | To be determined | 8-12 |
| Fuel | 50-125 | 50-125 | 50-125 |
| Angle of injection [degrees] | | | |
| Air | 0° | 25-70° | 68°, 114° and 114° |
| Fuel | 0° | 0° | 0° |

It is recommended to construct the burner in such a way that the burner heads can be flexibly exchanged and to test the burner in a (semi)-industrial furnace. During these tests, the effect of different burner heads and burner configurations on the NO_x emission will be studied. The information acquired during these tests will be used to create the optimum configuration for the three proposed low NO_x industrial high-temperature hydrogen burner designs. Together with industry partner(s) and burner manufacture(s), one burner design will be selected for a field test at one of the industry partners. The test will also give valuable insights for the design of hydrogen burners in general.

6.8 References

- [1] J. Warnatz, U. Maas, R.W. Dibble, *Combustion, Physical and Chemical Fundamentals, Modelling, and Simulations, Experiments, Pollutant Formation*. 3rd Edition
- [2] Kee, R. J., Rupley, F. M., and Miller, J. A., "CHEMKIN II: A Fortran Chemical Kinetics Package for the Analysis of Gas-Phase Chemical Kinetics", Report No.SAND89-8009, Sandia National Laboratories, 1989
- [3] R.. J. Kee, J. F. Grcar, M. D. Smooke, J. A. Miller, *Fortran program for modelling steady one-dimensional premixed flames. Sandia Report SAND85-8240. Sandia National Laboratories*, (1985).
- [4] Smith, G. P., Golden, D. M., Frenklach, M., Moriarty, N. W., Eiteneer, B., Goldenberg, W., Bowman, C. T., Hanson, R., Gardiner, W. C, Lissianski, V., and Qin, Z., http://www.me.berkeley.edu/gri_mech/.
- [5] S.Gersen, "Explore the use of sustainable hydrogen as a fuel for industrial heating processes (public summary)", Netherlands Enterprise Agency (RVO), Report 22-1123, 07-15-2022
- [6] J. D. Naber and D. L. Siebers. "Effect of gas density and vaporization on penetration and dispersion of Diesel sprays". In: SAE Technical Paper 105.412 (1996), pp. 82–111. ISSN: 0148-7191. DOI: 10.4271/960034.
- [7] Musculus, M. and Kattke, K., "Entrainment Waves in Diesel Jets," *SAE Int. J. Engines* 2(1):1170-1193, 2009, <https://doi.org/10.4271/2009-01-1355>.
- [8] Arai M., Tabata M., Hiroyasu H., Shimizu M. (1984) Disintegrating Process and Jet Characterization of Fuel Jet Injected by a Diesel Nozzle, SAE International, SAE paper 840275.
- [9] Spangelo, Øystein, "Experimental and Theoretical Studies of a Low NO_x Swirl Burner", doctoral thesis, Norwegian University of Science and Technology, September 2004, <https://ntnuopen.ntnu.no/ntnu-xmlui/handle/11250/231210>