# TOWARDS THE DESIGN OF AN ULTRA LOW NO$_x$ COMBUSTOR FOR BIOMASS DERIVED LCV GAS

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## ABSTRACT

Fuel bound nitrogen (FBN), (mainly ammonia NH$_3$) contained in biomass derived LCV (Low Calorific Value) gas is one of the major problems related to the combustion of such gases; this is due to the high conversion ratio of this nitrogen to NOx during the combustion process.

Within Thermal Power Engineering Section of the TUDelft, a group of researchers is busy with reducing NOx emission to the lowest and acceptable levels. A 30% conversion of NH$_3$ to NO was already achieved within a modified Alstom combustor and the so-called TUD combustor designed within the section [Hoppesteyn 1999, Adouane et. al. 2001].

In this paper, we present the first experiments with a Winnox combustor designed for Low NOx emission. These experiments are in the framework of a project funded by Novem (A Dutch Governmental Agency for Energy and Environment), and in partnership with three industrial companies (Winnox Combustion Systems, HoSt, and Geveke Motoren). Natural gas doped with ammonia was the fuel used in all described experiments. In the second phase of this project it is planned to use a synthetic LCV gas from a mixing station simulating the real LCV gas from biomass gasification.

The main goal of this project is to achieve ultra low conversion of fuel nitrogen into NO$_x$ during the combustion of LCV gas in a newly designed combustor. The aim is to achieve this goal without using catalyst reduction or scrubbing but with only design and combustion process optimization.

The experiments described in this paper are part of the first experiments planned in the beginning of this project in order to determine the key parameters controlling the conversion of fuel nitrogen to NO$_x$. These parameters will be used later in the design of the "final" version of the combustor for Low NOx emission from biomass derived LCV gas combustion. This combustor is intended to push the limits further and to achieve a conversion ratio of ammonia to NO$_x$ of approximately 12%, with combustion efficiency ($\eta_{combustion}$) higher than 98%.

The results of the experiments in terms of ammonia to NO$_x$ conversion ratio and NO$_x$ and CO emissions are presented in this paper.

Key words: NO$_x$, biomass, combustion, LCV gas

## NOMENCLATURE

| | |
|---|---|
| $C_{fuel\,N \rightarrow NO}$ | [%] Fuel bound nitrogen conversion ratio |
| FBN or  FN | [%] Fuel Bound Nitrogen *or* Fuel Nitrogen |
| RL | [--] Rich Lean concept |
| $D_i$ | [mm] Diamter |
| L | [mm] length |
| LCV | [--] Low Calorific Value |
| TUD | [--] Reference to Technical University of Delft |
| $\eta_{combustion}$ | [%] Combustion efficiency |
| $\lambda$ | [--]Air Stoichiometry Number (The inverse of the Equivalence Ratio) |
| $\Phi_v$ | [nm$^3$/h]Volumetric flow rate |

# 1. INTRODUCTION

Biomass derived LCV gas is an important green fuel alternative susceptible to play a crucial role in the coming years. It is expected that worldwide primary energy consumption will nearly double in the next 20 years. During this period the contribution of renewable energy sources will increase [IEA 2001].

With more concern about emissions, although biomass derived LCV gas is $CO_2$ neutral; it has its own problems related to pollution. The major problem related to the combustion of biomass derived LCV gas is the high $NO_x$ emissions due to the high fuel nitrogen [AL-Shaikhly et al. 1994].

Many methods are used to decrease these emissions; including scrubbing, catalytic reduction, and system optimisation. In this paper we will concentrate on the reduction of $NO_x$ emissions by optimizing the combustion process; this is done by optimising the design and the process for the lowest possible emissions.

Due to the non-availability of the mixing station for the generation of the LCV gas at the time of these experiments, it was decided to use natural gas doped with ammonia. Although natural gas represents some disadvantageous regarding high flame temperatures and high methane content which could enhance in fact the conversion of $NH_3$ to $NO_x$, it was decided to use this fuel because it can still define the key factors for the conversion of ammonia to $NO_x$, which will be the basis of the final design, and with the fact that we are dealing with the worse case in conversion (due to high flame temperatures and methane content), we expect that the conversion ratio to obtain with real LCV gas (in the project second phase) will be significantly lower.

The combustor used in these experiments was designed to work in two modes; Rich-Lean and Lean-Lean modes. Starting from the RQL (Rich Quench Lean) concept for low $NO_x$ emissions from natural gas and fuels with high level of fuel bound nitrogen [Griebel 1997], we tried to optimise the combustor to simulate an RL (Rich-Lean) combustion system, trying to apply the same concept for LCV gas.

Bearing in mind that when dealing with biomass derived LCV gas, the main issue with emissions is from fuel $NO_x$ resulting from the conversion of fuel nitrogen. The question was how would the combustor behave with the new situation. It turned out that the RL (Rich-Lean) concept doesn't reduce fuel $NO_x$, significantly. This because the concept plays on the temperature factor which is not a big issue for fuel $NO_x$ from LCV gas, because the flame has relatively quite low temperature compared to natural gas. The issue is that, when ammonia is cracked in the first stage (even at low temperatures), it will immediately convert to $NO_x$ when it sees oxygen or OH radicals. From the RQL concept, it was believed that the rich stage will let the fuel bound nitrogen convert to $N_2$ followed by a quick quench by air and a last lean stage for burnout will lead to low $NO_x$ [Hoppesteyn 1999], this was not the case in these experiment with RL concept.

This paper presents the preliminary experiments prior the final design of the combustor which will be tested intensively with LCV gas from diluted natural gas and later with a more "real" LCV gas from a mixing station of the main gases contained in the biomass derived LCV gas. Of course it will not contain all contaminants contained in the biomass derived LCV gas, but we think it will simulate well the LCV gas.

Optimising the Winnox combustor by tuning the primary and secondary air for the lowest emissions with stable combustion regime shows a good and promising trend in minimising $NO_x$ emissions. It could get to very low conversion ratios for high ammonia contents and it could reach an optimal stoichiometric point with the lowest conversion ratio of ammonia. This was reproducible with different regimes and ammonia contents in the gas. This achievement was of a great importance for us in trying to understand the mechanism of fuel $NO_x$ formation. This key parameter controlling the conversion of fuel nitrogen to $NO_x$, is used in the design of the new version of the Winnox combustor which will be tested in the following phase of this project.

# 2. DESCRIPTION OF THE WINNOX COMBUSTOR

Winnox combustor (figure 1) was originally, designed to be mounted on a Rover gas turbine and to combust biomass derived LCV gas. This explains the presence of a bending in the construction. It has a diameter of ($D_1$=171mm) on the burner head side, and diameter ($D_2$=105mm) on the exhaust side and a total length of (L=685mm). The thermal power output at 3.2 bar is approximately 600kW. The combustion chamber exit temperature is approximately 1150 K at full load. (For the Rover gas turbine operational conditions).

The same Winnox combustor was selected to be used in the frame work of the current Novem funded project for a set of first experiments aiming to define the design parameters to be adopted in the newly version of a low $NO_x$ combustor for biomass derived LCV gas.

In these experiments, the combustor was operated between 50 and 100 $kW_{th}$ at atmospheric pressure, this is done in order to avoid high temperatures. The total airflow was between 80 and 110 $nm^3$/h.

Figure 1 shows a cross section of the Winnox combustor. The air is admitted through one air inlet then it is split to primary and secondary air. The primary air flows between the inner and outer liners, cooling the inner liner

and preheating the primary air to approximately 200 ºC before it enters the main swirling head of the burner. The secondary air enters the combustor through 9 holes on the circumference of the second stage with a diameter of 8mm.

The combustor contains a deflection plate mounted in between the primary and secondary stage, this plate plays a big role in separating the two regions, per consequence we make sure that we have definitely two flames in case of Rich Lean mode, and it also enhances the mixing in the firs stage.
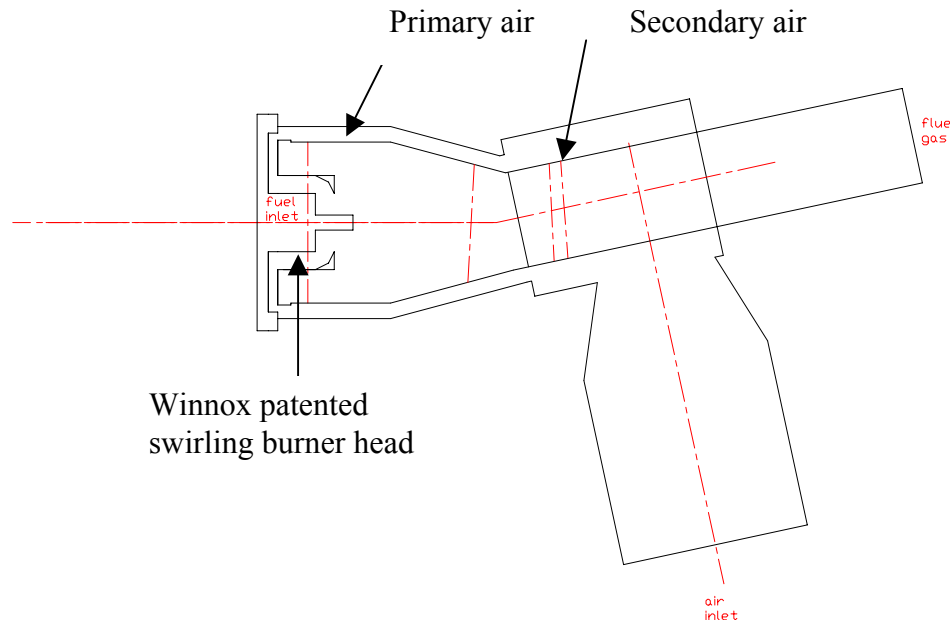


Figure 1. Winnox combustor for LCV gas combustion

## 3. EXPERIMENTS DESCRIPTION

All the experiments were performed using natural gas doped with $NH_3$. This because of the non availability of the mixing station for synthetic LCV gas generation at the time of these experiments. Although, natural gas can present some disadvantageous because of the high flame temperature and the effect of high methane concentration, but, as these first experiments are meant to define the key parameters controlling the conversion of $NH_3$ to $NO_x$, we judged acceptable the generated results. The mixing station will be used in the second phase of the project. We are convinced that we are dealing with the worse case of fuel nitrogen and if we could optimise our combustor to the lowest possible emissions using natural gas doped with ammonia, there is a big hope that the achieved conversion ratio will be lowered further when using the real synthetic LCV gas. This was the driving force for going further with those experiments.

The combustion chamber was operated with a capacity ranging from 50 to 100 kW.

The strategy in these experiments was to look at the effect of ammonia concentration in fuel gas on the emissions in terms of $NO_x$, and to investigate the effect of different operational regimes and equivalence ratios on the emissions.

The combustor, in each testing day is first fired without ammonia injection, set to it its optimum regime in terms of emissions and capacity, and then we start ammonia injection, and tuning the combustor regime by adjusting air and gas to find the optimal operational regime giving the lowest emission in terms of CO and NOx and thus the lowest conversion ratio of ammonia to $NO_x$.

## 4. NO FORMATION

NO is the dominant constituent of $NO_x$ in diffusion flames, this was confirmed in early experiments with an integrated combustion system with a PFBG (Pressurized Fluidized Bed gasifier) [Hoppesteyn, 1999], where we found out that $NO_2$ in the flue gas was 5 to 8 orders of magnitude lower than the concentration of NO: Other nitrogen containing species (with the exception of molecular nitrogen), such as $N_2O$, $NH_3$ and HCN, were not detected in the exhaust gas. [Adouane et al. 2003].

Because of the relatively low flame temperature in the case of LCV gas combustion, the thermal NO is negligible compared to the fuel NO resulting from the fuel bound nitrogen contained in the fuel gas. The major

mechanism responsible for the NO emission in the combustion of LCV fuel gas is the fuel NO mechanism (mainly conversion of $NH_3$ and HCN to NO). This hypothesis is still valid for our experiments since the combustor is set to its lowest $NO_x$ emission before ammonia injection, then we start $NH_3$ injection, so we could correct our $NO_x$ emission to only fuel $NO_x$ by subtracting the initial $NO_x$ emission from ammonia-free gas, which corresponds to thermal $NO_x$.

Since the concentration of NO is zero in the fuel gas and the concentration of $NH_3$ in the flue gas is zero, the conversion of FBN to NO can be defined as:

$$C_{fuel\ N \to NO} = \frac{[NO]_{flue\ gas} \cdot \Phi_{v,\ flue\ gas}}{[NH_3]_{LCV\ fuel\ gas} \cdot \Phi_{v,LCV\ fuel\ gas}} \cdot 100\ \%$$

(1)

In this definition it is assumed that all NO is attributed to the fuel NO mechanism.


# 5. RESULTS AND DISCUSSION

These experiments were performed in order to define the key parameters controlling the conversion ratio of ammonia to $NO_x$. Although the fuel used is natural gas, which differs a lot from LCV gas from biomass gasification, it is judged that it could still be used to define those key parameters dominating the conversion of fuel nitrogen to $NO_x$

Figure 2 shows clearly the trend of ammonia conversion ratio versus ammonia concentration in the fuel gas, in this experiment, the combustor was operated at its lowest optimal conversion ratio, with $O_2$ in the exhaust between 3.4 and 4.7%, which was found to be the optimal range of $O_2$ for the lowest conversion of $NH_3$. Decreasing ammonia concentration affects drastically the conversion ratio inversely. Increasing ammonia concentration 20 times results in a decrease of the conversion ratio of about 4 orders of magnitude.
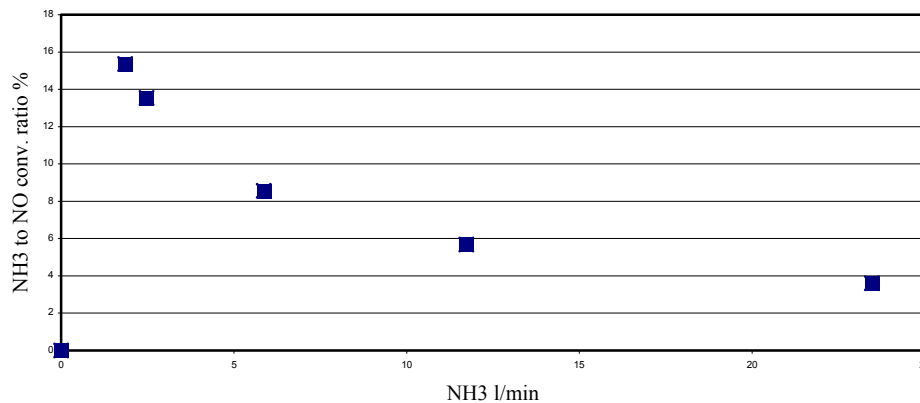


Figure 2: Effect of ammonia concentration on the conversion ratio of $NH_3$ to $NO_x$.

In figure 3, we see the trend of $NH_3$ to $NO_x$ conversion ration versus $O_2$ in the exhaust at high and low ammonia concentration in the gas. The deep is very clear at high ammonia concentration in the fuel gas and relatively flat at low ammonia concentration. The deep is seen at $O_2$ about 4% in the exhaust. This finding is also of a very big importance for the determination of the conversion of ammonia to $NO_x$ controlling parameters. It shows clearly a competing effect between the first and second stage which ends by an optimal point for the conversion ratio at about 4% $O_2$ in the exhaust.

From the design characteristics, we know that at about 4% $O_2$ in the exhaust, the first stage is at about stoichiometry, ($\lambda \approx 1$). It was surprising when we found out that the combustor gives the lowest conversion ratio when the first stage is at about stoichiometry. This explains the competing effect inside the combustor between $NO_x$ production and destruction. At this point ($O_2$=4%), the combustor is operating at its optimal point leading to the lowest conversion ratio of $NH_3$.

The curve demonstrates also clearly the factor of 10 to 100 orders of magnitude between ammonia concentration and conversion ratio respectively. This finding in addition the deep described above is the corner stone for the optimisation of the combustor for the lowest ammonia to $NO_x$ conversion.

In figure 3, we see that the behaviour of the combustor for low and high ammonia concentration in the fuel gas. At 4 % O2, we got a lowest conversion of about 4% which is acceptable for high $NH_3$ containing fuel gas. We

don't know exactly what are the chances to get this conversion ratio very low than this in order to reduce further $NO_x$ emissions.

For low NH3 content (1300 to 2000ppm), Figure 3 shows that the lowest conversion ratio was at about 50% which is relatively high compared to the results obtained with Alstom UK swirling combustor [Hoppesteyn 1999] where 30% conversion ratio was obtained. But it is comparable to the results obtained with a TUD combustor [Adouane, 2003], [van der Wel, Adouane 2002]. We believe that this relatively high conversion ratio at low $NH_3$ content in the gas is very related to the fuel used in these experiment, which is natural gas leading to high flame temperature enhancing the conversion of $NH_3$ to $NO_x$ and also without excluding the effect of thermal $NO_x$ (in spite of the correction done at the beginning for thermal $NO_x$), in addition to the role that may play $CH_4$ acting as a catalyst in favour of $NO_x$ production.

From what is said we believe that our Winnox combustor under investigation now will perform much better with real LCV gas from biomass gasification because of the low concentration of methane and also the low temperature, which when reducing both of them would enhance obtaining lower conversion ratios of $NH_3$ to $NO_x$.



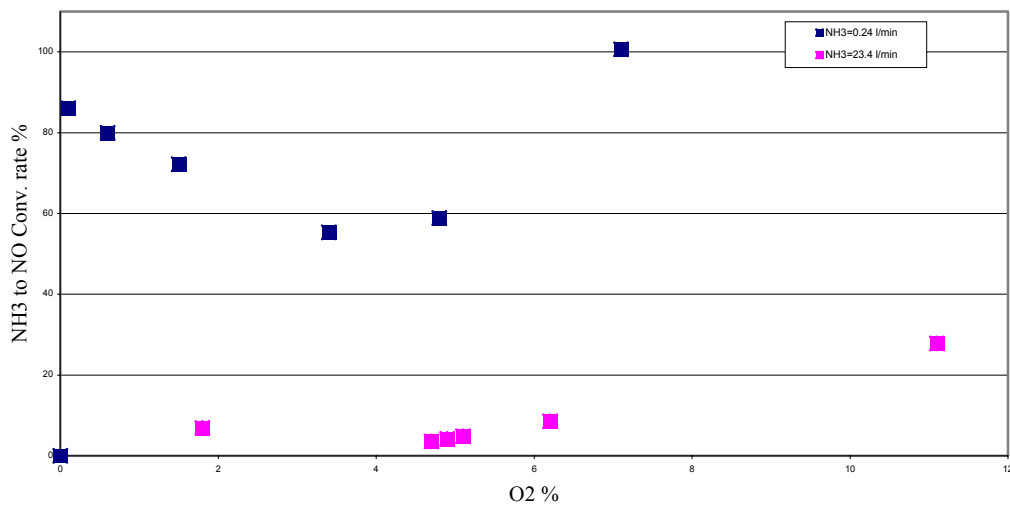Figure 3: NH3 to NOx conversion ratio at two high and low ammonia concentration

Figure 4 shows the conversion ratio of $NH_3$ to NO in addition to CO emissions. Oxygen on the x-axis is ranging from 1 to 3.5%. From the design characteristics of the Winnox combustor, it is known that throughout all this range (1 to 3.5% O2) the first stage is under-stoichiometric, which means that the first stage continue its production/destruction of $NO_x$ all along this range of $O_2$. It is clearly seen that going from 1% to 3.5% O2, the conversion ratio of $NH_3$ to NO has decreased by one order of magnitude (from 30 to less than 15%), which is quite important. Arriving 3.5% O2 in the exhaust, CO was already in the acceptable level.
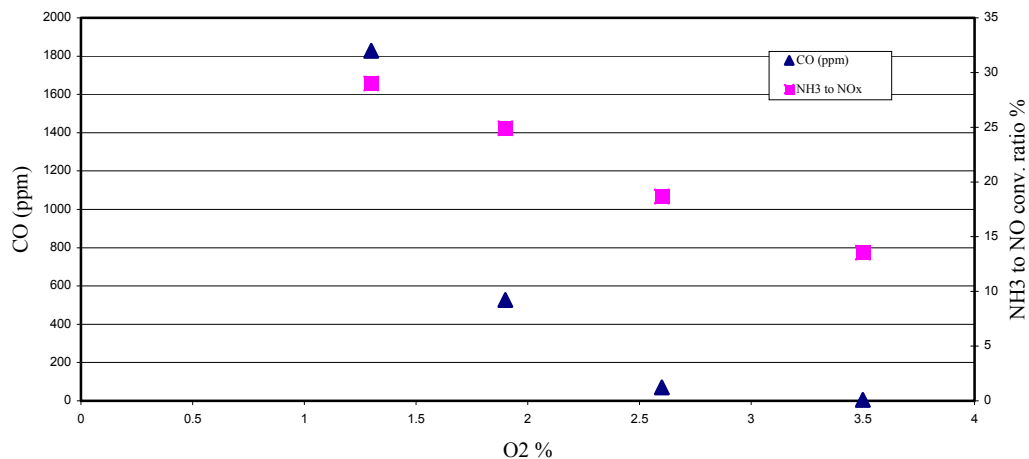
Figure 4: NH$_3$ to NOx conversion and CO emission versus O$_2$ in the exhaust (NH$_3$=2.47 l/min)

Figure 4 shows NO$_x$ and CO emissions versus O$_2$ in the exhaust at low ammonia content in the fuel gas. Here we see the same deep in the NO$_x$ curve at O$_2$ about 4% in the exhaust.
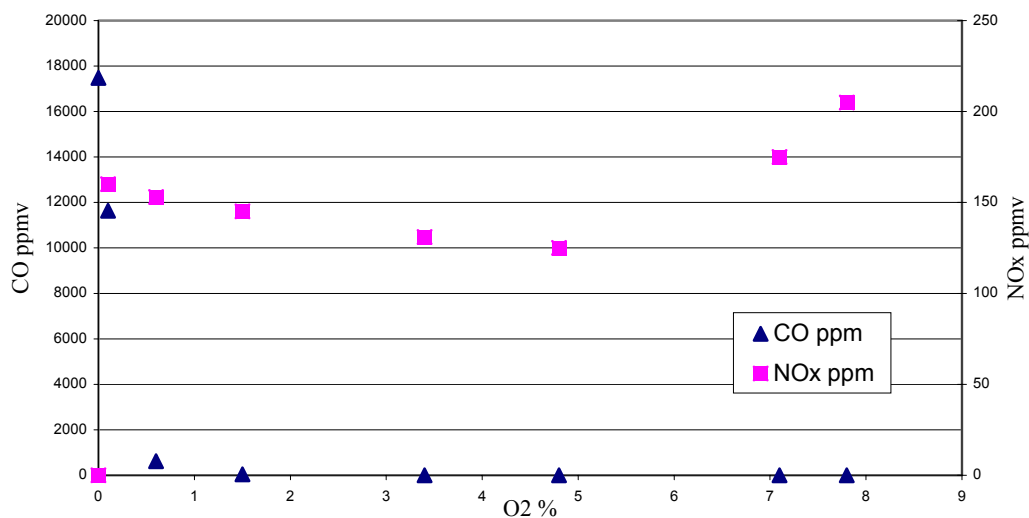


Figure 4: CO and NO$_x$ versus O$_2$ in the exhaust (NH$_3$= 1300-2000 ppm)

## CONCLUSIONS

After the set of experiments performed on the Winnox combustor, the main key parameters controlling the conversion ratio of NH$_3$ to NO$_x$ were determined

- The RL (Rich Lean) concept doesn't reduce the NH$_3$ conversion to NO$_x$ (fuel NO$_x$) as it does for thermal NO$_x$ from natural gas combustion.
- The conversion ratio depends strongly on the concentration of NH$_3$ in the fuel gas, this controversial to the finding of [Hoppesteyn 1999] of being independent.
- The combustor with two stages has an optimal regime resulting in the lowest possible conversion ratio, this was achieved at O$_2$ about 4 % in the exhaust, this corresponds to an optimal production/destruction of NOx in the first and second stage
- At 4% O2 in the exhaust, and from the design information, the first stage is operating at about stoichiometry ($\lambda \approx 1$), and this corresponds to the lowest conversion, this means that at this stoichiometry, and because it is not deeply rich in the first stage, the combustor produces

locally NO$_x$ but not to the point to convert totally all NH$_3$, and part of this NO$_x$ formed will be reduced either by ammonia itself acting as a catalyst or by other NO$_x$ precursors namely NH$_i$.

- The first stage continue reducing the conversion ratio in a relatively large understoichiometric range, this indeed prove the fact the stoichiometry level of the first stage in combination with the second stage plays a key role in the production/destruction of NO$_x$.
- Comparing the curves of low and high NH$_3$ content in the fuel gas, we see that the difference in conversion ratio is in the order of magnitude of 10, while the difference of ammonia concentration was in the order of 100 (23.4 to 0.24 l/min). This explains the mechanism of the conversion of fuel nitrogen (FN) to NO$_x$, increasing NH$_3$ in the fuel gas reduces the conversion ratio but not linearly, because on the other hand there is the mixing effect, the system is far from being ideally mixed to the molecular level.

## 6. THE NEWLY DESIGNED COMBUSTOR FOR ULTRA LOW NO$_X$

Based on the results described in this paper and in addition to the modelling results obtained by Fluent code simulations and calculations using Chemkin software, the design of the newly version of the Low NO$_x$ combustor has been defined and it is, while writing this paper, under construction. The new combustor will be tested using synthetic LCV gas from a mixing station and modelled using Fluent CFD commercial package.

The experiments in the second phase of this project using the newly designed combustor will be more detailed, and a large range of parameters will be measured, including flame temperature, walls temperature, species measurement inside the combustor at different positions and at the exhaust. After gathering all the results from the intensive measurement campaign, the last adjustments will be defined towards the optimisation of the design and the combustion process for an ultra low NO$_x$ emission from fuel bound nitrogen contained in the LCV gas.

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