Large Eddy Simulation investigation of pressure and wall heat loss effects on rich ammonia-hydrogen-air combustion in a gas turbine burner

Kévin Bioche\textsuperscript{a,b,c,*}, Julien Blondeau\textsuperscript{a,b}, Laurent Bricteux\textsuperscript{c}

\textsuperscript{a}Thermo and Fluid Dynamics (FLOW), Faculty of Engineering, Vrije Universiteit Brussel (VUB), Pleinlaan 2, 1050 Brussels, Belgium
\textsuperscript{b}Brussels Institute for Thermal-fluid systems and clean Energy (BRITE), Vrije Universiteit Brussel (VUB) and Université Libre de Bruxelles (ULB), Belgium
\textsuperscript{c}Université de Mons (UMONS), Polytechnic Faculty, Belgium

Abstract

Ammonia is currently investigated as a sustainable energy source. Its mixture with hydrogen may present combustion characteristics that are similar to those of hydrocarbon, which motivates its use in gas turbines burners. Such similarities were discussed at atmospheric pressure in previous works for a fuel blend with a molar fraction of hydrogen $X_{\text{H}_2} = 0.46$, which is further studied here. The influences of pressure and wall heat loss on ammonia/hydrogen/air flames are for the first time investigated via LES. A first campaign is led at both 1 and 5 atm, to estimate the effect of pressure. It demonstrates that NO emissions are favoured by flame interactions with a hot wall, along which NO is convected. Accordingly, the flame length reduction observed at high pressure, due to higher heat release rates, leads to a more efficient NO consumption. Increasing the pressure shifts the equivalence ratio for optimal NH$_3$ and NO$_X$ emissions towards the lean side. It results in lower hydrogen emissions and therefore an increase of the combustion efficiency. Finally, the NH$_3$ and NO$_X$ emissions at optimal equivalence ratio are reduced from 450 ppmv for $\phi = 1.27$ at 1 atm, to $\sim$ 100 ppmv for $\phi = 1.20$ at 5 atm. A second campaign is led at both 1 and 5 atm, by varying the burner wall thermal boundary conditions. Lean combustion with cold walls presents high N$_2$O emissions of 607 ppmv while in rich cases, the higher gas temperatures and the excess of H radicals in the burned gases yield complete N$_2$O consumption. It is shown that heat loss effect on N$_2$O fractions distribution is reduced at high pressure due to weaker flame interaction with the cold walls. Finally, thermal boundary conditions are found to significantly affect NO$_X$, N$_2$O and NH$_3$ emissions, showing that heat losses should be considered when modeling such configuration.

Keywords: Large Eddy Simulation; Swirled burner; Ammonia; High pressure; Heat loss;
1. Introduction

Due to its propitious storage and transportation properties, ammonia (NH\textsubscript{3}) as an e-fuel was recently advanced as a renewable energy storage solution [1]. Still, its restitution in power plants is challenged by the low heating value and low flame speed of NH\textsubscript{3}, making it hard to burn in current gas turbines burners. Besides, its combustion with air might tend to emit large amounts of nitrogen oxides (NO\textsubscript{X}), or lead to NH\textsubscript{3} slip depending on the operating conditions [2]. The partial conversion of NH\textsubscript{3} into molecular hydrogen (H\textsubscript{2}) was shown to enhance combustion and provide stable flames in classic swirl burners [3]. Intents to provide stable combustion with low emissions are available in the literature. Rich ammonia-hydrogen blend combustion in a gas turbine burner was studied from RANS simulations in [4], showing NO\textsubscript{X} emissions under 400 ppm while heat loss to the wall were shown to impair the fuel conversion and thus combustion efficiency. Further experimental and numerical work was conducted on a similar fuel blend combined with humidification, it showed the possibility of injecting limited amounts of steam to reduce NO emissions while keeping low NH\textsubscript{3} emissions [5]. Yet, investigations on NH\textsubscript{3}/H\textsubscript{2}/air combustion are quite recent and further fundamental work is required to understand its dynamics in gas turbine burners. The present work aims at numerically investigating the effect of heat losses to the wall and pressure conditions, on NH\textsubscript{3}/H\textsubscript{2}/air combustion in a swirled burner.

The effect of an increase in pressure from 0.1 MPa to 0.5 MPa, on premixed and non-premixed pure NH\textsubscript{3}/air swirling flames, was investigated in studies [6–8]. These concluded that both nitric oxide (NO) emissions and NH\textsubscript{3} slip decrease with an increase in pressure. When operating with slightly rich mixtures to obtain equal NO and NH\textsubscript{3} emissions, these were of 700 ppmv at 0.1 MPa and 200 ppmv at 0.5 MPa. Besides, using a secondary air injection operated in rich-quench-lean (RQL) conditions, it was possible to reduce NO emissions down to 100 ppmv, while almost no NH\textsubscript{3} and H\textsubscript{2} were found in exhaust gases. When resorting to such a strategy, minimal final NO emissions were obtained by setting the first-stage equivalence ratio (E.R, $\phi$) to the slightly rich E.R minimizing both NO and NH\textsubscript{3} emissions.

Previous investigations on the effect of pressure elevation on NH\textsubscript{3}/H\textsubscript{2}/air swirl flames were conducted. Recent experiments [9, 10] showed that it is possible to stabilize such flames over a wide range of E.R and blend composition for pressures ranging from 0.1 to 0.5 MPa. These studies emphasized on emissions of very lean NH\textsubscript{3}/H\textsubscript{2}/air swirl flames and reached conclusion similar to that associated to NH\textsubscript{3}/air flames. While very lean conditions permits to produce low NO at high combustion efficiency, large amounts of nitrous oxide (N\textsubscript{2}O) (above 800 ppmv in the studied conditions) are observed in the exhaust gases. Numerical simulations of 1D flames also advanced the potential of pressure increase on NO emission reduction [11]. Other experiments [12] investigated the influence of steam addition and elevated pressure up to 0.184 MPa on a RQL NH\textsubscript{3}/H\textsubscript{2}/air burner with high inlet temperature (423 K). In this study, the molar fraction of H\textsubscript{2} in the fuel blend was set to $X_{\text{Fuel}}^{\text{H}_2} = 0.30$. By setting the first stage E.R for minimal NO and NH\textsubscript{3} emissions, and optimizing the humidity and secondary air injection at the highest pressure available, it was possible to achieve NO\textsubscript{X} emissions of 32 ppmv and NH\textsubscript{3} emissions of 50 ppmv at 15%O\textsubscript{2}.

The effect of heat losses to the combustors wall was investigated in [7, 8] for NH\textsubscript{3}/air swirl flames. Heat losses were shown to limit the NH\textsubscript{3} oxidation and increase emissions of unburnt NH\textsubscript{3} while lowering NO emissions. When operating at $\phi = 0.8$, the low temperature caused by heat loss led to the promotion of N\textsubscript{2}O. As a result, N\textsubscript{2}O emissions from this flame were found higher.
than NO, with 55 ppmv of NO, 580 ppmv of N₂O and 4457 ppmv of NH₃.

Large Eddy Simulation (LES) offered valuable insight on the study of pressure increase and heat loss effects on NH₃/air flames. Indeed, these two effects were shown to affect greatly the flame structure and species production locations of this in-homogeneous process, making LES a relevant tool. Still, these effects on NH₃/H₂/air swirl flames were never investigated using LES. In the present work, a first LES campaign is led at both 1 atm and 5 atm, to estimate the effect of pressure. A second LES campaigns is also led at both 1 and 5 atm, by varying the burner wall thermal boundary conditions (BC). The effect of heat loss on flame propagation and emissions is discussed for both atmospheric and higher pressure conditions.

2. Configuration

2.1. PRECCINSTA burner

The current research on NH₃/H₂ combustion in gas turbines burners focuses on laboratory-scale configurations [2, 13]. Based on previous work [14], the PRECCINSTA burner is selected for this study (see Fig. 1). Premixed NH₃/H₂/air enters the plenum and is directed toward the swirler. Twelve radial canals swirl and route the mixture toward the injection area composed of a converging nozzle and a central conical bluff-body. Combustion occurs in the chamber and products exit through a converging cone followed by a straight pipe.

![Fig. 1: Simulated geometry of the PRECCINSTA burner.](image)

2.2. Operating conditions

Following previous works [14, 15], the inlet mass flow rate is set to 770 g.min⁻¹, the inlet temperature to 300 K and the blend composition to X_{H₂}^{Fuel} = 0.46. The targeted operating blend (ϕ, X_{H₂}^{Fuel}) was determined in [14] from a 1D preliminary study, based on two criteria. First, to meet the well-known trade-off in NOₓ and NH₃ emissions. Second, to mimic the basic flame characteristics (adiabatic flame speed, thickness and temperature) of a lean methane/air combustion taken as a reference in this burner configuration [15]. From this starting point, obtained from low-order modelling, the E.R. was varied in the large eddy simulations to recover the NOₓ/NH₃ emissions at the trade-off point and in its vicinity.

To investigate the effect of pressure, series of simulations are performed at 1 atm and 5 atm. In both series, the E.R is varied to approach the optimal emission point where X_{NOX}/X_{NH₃} = 1. Indeed, NO and NH₃ emissions were shown to increase dramatically on the lean and rich side of this optimum point, respectively. Besides, this E.R and slightly leaner mixtures were found optimal for the first stage in RQL configurations [6]. The series of simulations are summarized in Table 1.
The effect of heat loss are then investigated at both 1 atm and 5 atm. The optimal E.R simulations, at both pressure, are repeated for four wall thermal conditions. The precise setting of boundary conditions are described later and are named BCadiab, BC1800, BC1000 and BC300. The series of simulations are summarized in Table 2.

Table 1: Simulations parameters and results for the campaign on pressure influence. Inlet mass flow rate: 770 g.min$^{-1}$. Inlet temperature: 300 K. Fuel blend composition: $X_{\text{H}_2}=0.46$.

<table>
<thead>
<tr>
<th>$P$ [atm]</th>
<th>$\phi$ [-]</th>
<th>$X_{\text{NO}_x}$ [ppmv]</th>
<th>$X_{\text{NH}_3}$ [ppmv]</th>
<th>$X_{\text{H}_2}$ [%]</th>
<th>CE [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1.15</td>
<td>3278</td>
<td>1</td>
<td>4.9</td>
<td>84.8</td>
</tr>
<tr>
<td></td>
<td>1.25</td>
<td>904</td>
<td>63</td>
<td>7.4</td>
<td>78.0</td>
</tr>
<tr>
<td></td>
<td>1.27</td>
<td>480</td>
<td>409</td>
<td>7.9</td>
<td>76.8</td>
</tr>
<tr>
<td></td>
<td>1.30</td>
<td>197</td>
<td>1851</td>
<td>8.4</td>
<td>75.0</td>
</tr>
<tr>
<td></td>
<td>1.15</td>
<td>632</td>
<td>5</td>
<td>6.0</td>
<td>85.6</td>
</tr>
<tr>
<td></td>
<td>1.20</td>
<td>147</td>
<td>318</td>
<td>6.8</td>
<td>81.9</td>
</tr>
<tr>
<td></td>
<td>1.23</td>
<td>74</td>
<td>848</td>
<td>7.2</td>
<td>79.7</td>
</tr>
<tr>
<td></td>
<td>1.25</td>
<td>59</td>
<td></td>
<td></td>
<td>78.3</td>
</tr>
</tbody>
</table>

Table 2: Simulations parameters and results for the campaign on heat losses influence. Inlet mass flow rate: 770 g.min$^{-1}$. Inlet temperature: 300 K. Fuel blend composition: $X_{\text{H}_2}=0.46$.

<table>
<thead>
<tr>
<th>$P$ [atm]</th>
<th>$\phi$ [-]</th>
<th>Thermal BC</th>
<th>$X_{\text{NO}_x}$ [ppmv]</th>
<th>$X_{\text{NH}_3}$ [ppmv]</th>
<th>$X_{\text{H}_2}$ [%]</th>
<th>CE [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1.15</td>
<td>BCadiab</td>
<td>894</td>
<td>11</td>
<td>12.7</td>
<td>65.5</td>
</tr>
<tr>
<td></td>
<td>1.25</td>
<td>BC1800</td>
<td>315</td>
<td>300</td>
<td>12.6</td>
<td>65.7</td>
</tr>
<tr>
<td></td>
<td>1.27</td>
<td>BC1000</td>
<td>86</td>
<td>1517</td>
<td>12.4</td>
<td>65.8</td>
</tr>
<tr>
<td></td>
<td>1.30</td>
<td>BC300</td>
<td>40</td>
<td>3326</td>
<td>12.1</td>
<td>65.9</td>
</tr>
<tr>
<td></td>
<td>1.15</td>
<td>BCadiab</td>
<td>275</td>
<td>5</td>
<td>6.0</td>
<td>81.8</td>
</tr>
<tr>
<td></td>
<td>1.20</td>
<td>BC1800</td>
<td>147</td>
<td>29</td>
<td>6.0</td>
<td>81.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>BC1000</td>
<td>52</td>
<td>509</td>
<td>5.9</td>
<td>81.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>BC300</td>
<td>15</td>
<td>2224</td>
<td>5.7</td>
<td>81.9</td>
</tr>
</tbody>
</table>

3. Modelling

3.1. Numerics

Large-Eddy Simulations of the PRECCINSTA burner are performed with the finite-volume code YALES2 [16], following the same modelling strategy presented in [14]. The low-Mach number Navier-Stokes equations, with variable density, are solved on an unstructured mesh. Transport properties are modelled with the mixture-average approach [17]. The dynamic Smagorinsky model [18] is used for sub-grid scale modelling. Turbulent combustion is modelled with the dynamic TFLES model [19] (imposing 5 points in the flame front) and the efficiency function of Charlette [20].

Two different mesh resolutions are adopted to discretize the geometry displayed in Fig. 1. Previous work led at 1 atm showed with a mesh refinement study that a 9 million elements mesh, refined down to $\Delta = 1$ mm in the combustion area, provides accurate results [14]. When increasing the pressure to 5 atm, the flame thickness diminishes from 669 $\mu$m to 213 $\mu$m at $X_{\text{H}_2}=0.46$ and $\phi = 1$. Finer meshes are thus required to capture precisely the combustion process. Accurate results were obtained with $\Delta = 0.3$ mm in the combustion area. With such resolution, the thickening factor used in the TFLES model is limited to $\sim 6$ in all cases, which was found not to impact predicted emissions in [14]. As discussed later, the combustion area is smaller at high
pressure resulting in a reasonable increase of meshes size, ranging from 30 to 38 million number of elements.

3.2. Kinetics

In a reference work at 1 atm, a kinetic mechanism was selected based on available experiments [14]. Typical kinetic mechanisms were assessed on the basis of the flame speed and ignition delay time measurements closest to predicted operating conditions, and Zhang mechanism [21] was selected. A similar preliminary study was presently conducted at 5 atm. At $X_{\text{H}_2} = 0.46$, it points out that the operating E.R should approach $\phi = 1.35$ to yield equal NOX and NH3 emissions (see Fig. S1 in supplementary material).

Experiments by Wang [22] and Chen [23] are selected to assess the kinetic mechanisms for flame speed and ignition delay time, respectively (see Figs. S2 and S3 in supplementary material). The following objective function $E$ is used to evaluate the models predictions for both ignition delay time (IDT) and laminar flame speed ($S_L$) measurements:

$$E = \frac{1}{N} \sum_{i=1}^{N} \left( \frac{1}{N_i} \sum_{j=1}^{N_i} \frac{(Y_{ij}^{\text{sim}} - Y_{ij}^{\text{exp}})^2}{Y_{ij}^{\text{exp}}} \right),$$

where $N$ is the number of datasets, $N_i$ is the number of data points in the $i$-th dataset and $Y_{ij}$ is the $j$-th data point in the $i$-th dataset. The superscripts sim and exp refer to the model simulation and reference experiment, respectively. The expression of the data point $Y_{ij}$ depends on the nature of the experiment. For flame velocities experiments $Y_{ij} = S_L$, while for auto-ignition delay experiments $Y_{ij} = \log(\text{IDT})$. Results are gathered in Table S1 in supplementary material. Only Okafor [24] mechanism places itself in the three best mechanisms both on flame speed and ignition delay time. It is thus selected for the 5 atm campaigns. For the sake of comparison, the 1 atm campaign is repeated with Okafor mechanism, although it was found that is not best suited for these simulations [14]. This process allows identifying the effect of pressure alone, aside from the discrepancies typically observed between kinetic mechanisms [25]. Still, Zhang mechanism is conserved for the campaign varying the thermal boundary conditions at 1 atm.

3.3. Wall thermal boundary conditions

Simulations in Table 1 are performed with the realistic thermal boundary conditions BC1800. These were set at 1 atm pressure in [14] to mimic the heat transfer through the injector and the burner walls, based on previous work on a lean methane case [15]. It consists in a temperature Dirichlet boundary condition ranging from 300 K in the injector area to 1800 K in the second half of the burner walls.

Simulations in Table 2 use three other thermal boundary conditions. To study enhanced thermal losses, BC1000 and BC300 yield temperature Dirichlet boundary condition with 1000 K and 300 K in the second half of the burner walls, respectively. Adiabatic conditions are also investigated and named BCadiab. Although BCadiab and BC300 are not realistic conditions, they shed light on the limit behaviour of the system versus heat losses.

4. Results
4.1. Effect of pressure

With a pressure increase from 1 atm to 5 atm, the E.R. yielding equal NO\textsubscript{X} and NH\textsubscript{3} emissions present an emissions reduction from 450 ppmv to \sim 100 ppmv (see Fig. 2). This observation is consistent with previous work on pure NH\textsubscript{3} combustion [6], in which emissions were lowered by a factor 3.5 for a similar pressure increase. In a previous study on NH\textsubscript{3}/H\textsubscript{2} combustion [12], NO\textsubscript{X} emissions were reduced by nearly an order of magnitude for a fixed rich E.R across the experimental pressure range (0.105 MPa-0.184 MPa), while NH\textsubscript{3} emissions increased, shifting the optimal point towards the lean side. Consistently, the present optimal emissions E.R is shifted towards the lean side from $\phi = 1.27$ to slightly above 1.20, while the pressure rise yields e.g. NO\textsubscript{X} emissions reduction from 904 ppmv to 59 ppmv at $\phi = 1.25$. Besides, H\textsubscript{2} emissions are not much impacted by the pressure. Accordingly, when comparing pollutant emissions optimums at both pressure, the shift of the optimal point towards the lean conditions results in lower H\textsubscript{2} emissions and thus higher combustion efficiency defined as (see Fig. 3),

$$CE = 1 - \frac{Y_{\text{products}}^{\text{NH}_{3}} \cdot \text{LHV}_{\text{NH}_{3}} + Y_{\text{products}}^{\text{H}_{2}} \cdot \text{LHV}_{\text{H}_{2}}}{Y_{\text{reactants}}^{\text{NH}_{3}} \cdot \text{LHV}_{\text{NH}_{3}} + Y_{\text{reactants}}^{\text{H}_{2}} \cdot \text{LHV}_{\text{H}_{2}}}.$$  

![Fig. 2: Pressure effect on NO\textsubscript{X} and NH\textsubscript{3} emissions.](image)

Fig. 4 shows time averaged fields of the chamber right half plane for simulations with realistic thermal boundary conditions, at optimal emissions E.R. At high pressure, the flame exhibits a much higher mean heat release rate $\dot{HHR}$ causing a drastic flame length reduction. As discussed in [6] from 1D simulations, the combustion characteristic time $\tau_{c} = \frac{D_{th}}{S_{c}^{2}}$, with $D_{th}$ the fresh gas thermal diffusivity, is reduced when the pressure augments, especially for the present pressure range. 1D freely propagating premixed flames computed with Cantera [26] yield that $\tau_{c}$ is reduced by a factor 3, from 502 \mu s at 1 atm and $\phi = 1.27$, to 174 \mu s at 5 atm and $\phi = 1.20$. With this flame length reduction, the combustion process occurs closer to the bluff body and a stronger flame anchoring is observed at the burner basis. Besides, the mean longitudinal velocity $U_{X}$ slices show
a central recirculation zone (CRZ) slightly larger at high pressure, yet having similar shapes in both cases ($\bar{U}_X=0$ is marked by a white line). In the presence of the CRZ, the flame develops conically towards the burner wall. Both configurations present a small outer recirculation zone (ORZ) at the right wall vicinity, but only the high pressure yields two ORZ at the burner wall basis, participating in the stabilization of the external flame envelope. While a weak external envelope was observed on the 1 atm flame yielding almost a V-shape, the $HHR$ is more evenly spread at high pressure giving a clear M-shape.

The change in flame length impacts NO$_X$ production. In both cases, final emissions of nitrogen dioxide (NO$_2$) stay below $\sim 1$ ppmv, leaving NO species as the main component of NO$_X$ by far. N$_2$O emissions also stay below $\sim 1$ ppmv. Highest concentrations of NO are observed at the flame tip, followed by a consumption in the burned gas. Still, at 1 atm this consumption appears to be limited by the hot wall and NO pockets are transported along it [14]. The shorter flame observed at high pressure allows an efficient consumption of NO before the burned gases are pushed towards the wall, although the relatively high temperature observed at the flame tip led locally to very high NO concentrations.

The influence of pressure on NO emissions is further analysed from 1D simulations of freely propagating premixed flames. NO mass fraction $Y_{NO}$ and mass fraction production rate $\dot{\omega}_{NO}/\rho$ are analysed for NH$_3$/H$_2$/air flames with $\phi = 1.25$ and $X_{H_2}^{\text{Fuel}} = 0.46$ at 1 atm and 5 atm (see Fig. 5). The flame thickness is notably reduced from $\delta_F^{1\text{atm}} = 736 \mu m$ at 1 atm to 194 $\mu m$ at 5 atm. When increasing the pressure, $Y_{NO}$ yields both a lower peak at the flame front center vicinity and a sharper decrease toward the burned gas side. Indeed, the higher NO fraction production rate at the flame front center does not compensate for the reduced flame thickness. The NO consumption in the flame front burned gas side and post combustion zone is enhanced by the high pressure and $Y_{NO}$ is reduced by a factor 4.3 from its peak value when reaching $x = 2\delta_F^0$, while a factor 1.6 is observed at 1 atm. The conservation equation for species $k$ yields,

$$\rho u \frac{\partial Y_k}{\partial x} = - \frac{\partial j_k}{\partial x} + \dot{\omega}_k,$$

with $j_k$ the diffusive mass flux of species $k$. Interestingly, once scaled by the flame thickness, the relative effects of convection, diffusion and production on the NO species conservation equation
show small discrepancies at 1 and 5 atm (see Fig. 5). At high pressure, the mass diffusion away from the flame core is slightly more intense, while NO consumption is enhanced in the post-combustion zone. The convective term is finally consistently lower across the flame front at high pressure.

At both pressures, the main kinetic route leading to NO production passes through HNO, with the reaction $H + HNO \leftrightarrow H_2 + NO$ playing the first order role (see Fig. 6). In the high pressure flame, the reaction $HNO + M \leftrightarrow H + NO + M$ plays a much stronger role than for its low pressure counterpart, which constitutes the main difference between the two flames. In both cases, second order chemical routes for NO production include NH and N via reactions $NH + O \leftrightarrow H + NO$, $NH + NO \leftrightarrow H + N_2O$ and $N + O_2 \leftrightarrow NO + O$. Nitrogen reaction pathways graphs available in the supplementary material support this analysis (see Figs. S4-S5).

4.2. Effect of heat loss

Results in Table 2 point out that increased heat loss have for effect to reduce NOX emissions and increase NH3 slip, for 1 atm and 5 atm rich configurations. Moving from BCadiab to BC300 reduces NOX emissions by a factor $\sim 20$ in both cases. Figs. 7 and 8 show that NO is mainly produced in the reaction zone, with peaks in concentrations located at the flame tip, and much higher concentrations observed in the adiabatic cases, in association to high $HRR$. Further, low wall temperatures favour an efficient NO consumption, while NO is convected along hot walls.
In the 5 atm adiabatic case, the flame length reduction allows for a consequent NO consumption before it reaches the hot wall, while the long flame observed at 1 atm does not. NO consumption far from the burner walls is strongly correlated to relatively low gas temperatures. Accordingly, much higher NO concentrations are observed in the CRZ and ORZ for the adiabatic cases.

While varying BC, all cases yield final emissions of NO$_2$ below the order of 1 ppmv, leaving NO as the main component of NO$_X$. While in these cases N$_2$O emissions are below the order of 1 ppmv, previous works on lean NH$_3$/air flames showed that heat loss inhibited NO but promoted N$_2$O emissions, up to 600 ppmv for similar configurations [8, 27]. These works predicted that this powerful greenhouse gas is mainly consumed via thermal decomposition and reactions with hydrogen (H) radicals.
Fig. 7: Effect of heat loss on swirled flames at 1 atm and $\phi = 1.46$. Time averaged fields.

Fig. 8: Effect of heat loss on swirled flames at 5 atm and $\phi = 1.20$. Time averaged fields.
To analyse further N₂O emissions process in NH₃/H₂/air flames, 1D freely propagating flames were computed with Cantera. Like previously, the flames yield \( X_{\text{H}_2}^{\text{Fuel}} = 0.46 \) and were computed at 1 atm and 5 atm, for both rich (\( \phi = 1.25 \)) and lean (\( \phi = 0.7 \)) conditions. Rates of progress of N₂O are displayed in Fig. 9. H radicals play the main role on N₂O consumption, via reactions \( \text{H} + \text{N}_2\text{O} \leftrightarrow \text{N}_2 + \text{OH} \) and \( \text{NH} + \text{NO} \leftrightarrow \text{H} + \text{N}_2\text{O} \). For the lean flame at high pressure, the third body reaction \( \text{N}_2\text{O} + \text{M} \leftrightarrow \text{N}_2 + \text{O} + \text{M} \) plays a role of similar importance, probably due to the low availability of H radicals in lean flames, further pronounced at high pressure (see Fig. 10).

To investigate the effect of heat loss on N₂O emissions, the temperature profile of the adiabatic lean atmospheric 1D flame \( T_{\text{ad}}(x) \) is extracted and used to compute a 1D burner flat flame with the library Cantera. A first flame is computed with the adiabatic flame temperature profile \( T_{\text{ad}}(x) \). A second flame is computed with a modified temperature profile mimicking heat losses. With the flame front located at \( x = 0 \), the profile with heat loss follows the following rule. For \( x \geq 0 : T(x) = \max \left( T_{\text{ad}}(x) - 100 \frac{x}{\phi}; 300 \right) \), while \( T(x) = T_{\text{ad}}(x) \) for \( x < 0 \), resulting in the temperature profile shown in Fig. 11(a). Corresponding N₂O and H profiles are displayed in Figs. 11(b) and 11(c), respectively. The two cases yield a N₂O peak of same magnitude in the
flame front, but the presence of heat loss results in a lower H peak. For these flames, N\textsubscript{2}O is here again mainly consumed by combination with H radicals (see Fig. 11(d)). Accordingly, while H radicals are available in sufficient amounts to yield quasi zero N\textsubscript{2}O emissions for the adiabatic flame, significant emissions ($\approx$ 600 ppmv) are observed for the flame submitted to heat loss.

![Figures](image1.png)

Fig. 11: Properties across 1D NH\textsubscript{3}/H\textsubscript{2}/air flames computed with a fixed temperature profile. $\phi = 0.7$ and $X_{\text{H}_2} = 0.46$. Computed with Cantera.

Consistently, in the LES configuration, Fig. 7 shows that low wall temperatures limit drastically N\textsubscript{2}O decomposition compared to high temperatures. Still, in all rich cases, complete N\textsubscript{2}O consumption is finally completed before the burner outlet. Presently, a lean configuration LES was done and predicted 607 ppmv of N\textsubscript{2}O at the burner exhaust with parameters: $P = 1$ atm, $\phi = 0.7$ and BC300. Figs. 12 and 13 shows averaged and instantaneous fields of this simulation, respectively. It reveals that the zones of N\textsubscript{2}O consumption are located where H radicals are present, and are strongly dependent on local gas temperature. As advanced from the 1D investigation presented above, H radicals are almost absent in the burned gas, which combined with the low wall temperature inhibits N\textsubscript{2}O consumption. Along the wall higher part, N\textsubscript{2}O is convected in the cold flow areas, while a slight consumption is still observed in the hot gas away from the wall. In the rich cases, the higher gas temperatures and the excess of H radicals in the burned gases yield complete
$N_2O$ consumption (see Figs. 14 and 15). This constitutes an advantage of rich over lean combustion. Besides, Fig. 8 shows that the effect of heat loss on $N_2O$ fraction distribution is reduced at high pressure due to the flame length reduction and consequent lower interaction with the cold walls.

![Fig. 12: Swirled flame at 1 atm and $\phi = 0.7$, BC300. Time averaged fields.](image)

Passing from BCadiab to BC300, the reduction of the $HHR$ leads to an increase of unburned NH$_3$ by more than 2 orders of magnitude in both cases (see Table 2 and Figs. 7 and 8). Combined with the NO emissions reduction associated to wall heat loss, it tends to shift the low NO/NH$_3$ emission window towards leaner E.R. At both low and high pressure, the lower gas temperatures and H$_2$ recirculation observed in the ORZ with BC300 do not allow for the flame stabilization in this area. The flames pass from M-shapes to V-shapes, sign of weaker stability. With the lower fuel consumption, less H$_2$ present in the fuel is consumed, but also less H$_2$ is produced from NH$_3$ dissociation. Overall, H$_2$ concentrations are slightly reduced with heat loss, consistently with work on NH$_3$/air flames [28]. Still, this effect is limited, and combustion efficiency is fairly stable across the various boundary conditions.

![Fig. 13: $N_2O$ production and consumption at 1 atm, $\phi = 0.70$ and BC300. Computed with Zhang mechanism. Instantaneous fields.](image)
Fig. 14: $N_2O$ production and consumption at 1 atm and $\phi = 1.46$. Instantaneous fields.

Fig. 15: $N_2O$ production and consumption at 5 atm and $\phi = 1.20$. Instantaneous fields.
5. Conclusions

Investigating the effect of pressure variation via LES in a range from 1 atm to 5 atm showed that increasing the pressure shifts optimal emissions E.R. towards the lean side, resulting in lower \( \text{H}_2 \) emissions and therefore in an increased combustion efficiency. Besides, high pressure flames exhibits a much higher mean heat release rate causing flame length reduction, leading to a more efficient NO consumption. Finally, the \( \text{NH}_3 \) and \( \text{NO}_X \) emissions at optimal equivalence ratio are reduced from 450 ppmv for \( \phi = 1.27 \) at 1 atm, to \( \sim 100 \) ppmv for \( \phi = 1.20 \) at 5 atm. The numerical investigation of heat loss effect showed that low wall temperatures are favourable to an efficient NO consumption. Lean combustion with cold walls showed high \( \text{N}_2\text{O} \) emissions of 607 ppmv while in rich cases, the higher gas temperatures and the excess of \( \text{H} \) radicals in the burned gases yield complete \( \text{N}_2\text{O} \) consumption. Besides, heat loss effect on \( \text{N}_2\text{O} \) fraction distribution is reduced at high pressure due to weaker flame interaction with the cold walls. Finally, thermal BC can significantly vary \( \text{NO}_X \), \( \text{N}_2\text{O} \) and \( \text{NH}_3 \) emissions, showing that heat loss should be accounted for.

Although an increase of pressure was shown to offer a higher combustion efficiency, this last is still relatively low, due to high hydrogen emissions. Future work will focus on rich-quench-lean combustion to obtain a complete fuel conversion, as suggested in recent work [25]. Further work will include typical constraints, such as the cracking of ammonia into hydrogen and the combustion of \( \text{NH}_3/\text{H}_2/\text{N}_2/\text{air} \) blends [29].

Acknowledgments

This research received the support of the Energy Transition Fund of Belgian Federal Government (BEST project) and benefited from computational resources made available on the Tier-1 supercomputer of the Fédération Wallonie-Bruxelles, infrastructure funded by the Walloon Region under the grant agreement n°1117545. The authors thank V. Moureau, G. Lartigue and P. Bénard for sharing the YALES2 code.

Supplementary material

Supplementary material for this draft can be found in a joined file.

References


