# Thickened Flame model for multi-fuel multi-injection combustion– Pollutant analysis of an ammonia-hydrogen swirled flame

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## Abstract
An extension of the widely-used Thickened Flame model for Large Eddy Simulations (TFLES) to take into account multi-fuel multi-injection combustion processes is presented. The local variation in fuel composition and equivalence ratio due to differential diffusion and the mixing of differentiated fuel injections is computed from a transported mixture fraction passive scalar tracing the spatial evolution of each fuel stream. This allows to incorporate local fuel composition inhomogeneities into the combustion model and the flame sensor parameters. The proposed modeling is used to predict the ammonia-air swirling flame stabilized by multiple hydrogen injections operated at Cardiff University. To perform such simulations, a novel analytically reduced chemistry scheme for $NH_3$-$H_2$-$N_2$/air combustion is derived and validated at gas turbine operating conditions for multiple ammonia-hydrogen binary fuel blends as well as ternary fuel blends derived from ammonia decomposition. The results of the novel Multi-Fuel TFLES model (MF-TFLES) are compared against the conventional TFLES predictions and assessed via the experimental data at our disposal. The proposed modeling improves the flame shape prediction by assuring the correct application of the artificial flame thickening principles locally and coherently, taking into consideration the multi-fuel complex mixing process. Furthermore, thermal effects on $NO_X$ and flame topology are evaluated by comparing an adiabatic case against one considering wall heat losses.

## Introduction
Large Eddy Simulations (LES) have enabled accurate prediction of turbulent reactive flows by limiting the turbulence modeling to the small turbulent scales [1]. This work focuses on the Thickened Flame LES (TFLES) model [2] relying on the premixed flame theory. In this approach, the flame is artificially thickened by a factor $\mathcal{F}$, while conserving the flame speed ($S_L^0$) and the globally integrated heat release rate [3]. This model is, however, limited to case where (1) perfectly premixed combustion and (2) a mixture uniform unitary Lewis are assumed. The renewed interest in carbon-free fuels such as hydrogen ($H_2$) and ammonia ($NH_3$), highlighted limits of the approach previously dismissed, and new combustion modeling

challenges need to be considered for a proper use of the TFLES model in this new framework. Among many for such a vast challenge, two difficulties stand out: first, the correct modeling of unconventional burner technologies with multi-inlet multi-fuel injection systems, and second, the effect of differential diffusion introduced by the presence of $H_2$ in the fuel blend [4,5]. To the best of the authors' knowledge, none of the previous numerical studies [6,7] have considered the effect of differential diffusion issued by the presence of $H_2$ in the fuel blend. Furthermore, a configuration with differentiated injection of $NH_3$ and $H_2$, such as the one studied by Mashruk et al. [8], inevitably results in a spatially heterogeneous fuel composition in the burner, a feature overlooked by previous studies [6,9]. The present work aims, therefore, at developing a numerical methodology coupling a fuel-agnostic Multi-Fuel TFLES (MF-TFLES) model to a novel $NH_3$-$H_2$-$N_2$ Analytically Reduced Chemistry (ARC) scheme to perform high-fidelity LES of $NH_3$-$H_2$ partially-premixed flames with differentiated fuel injection in industrial gas turbine type applications.

**Chemical Kinetics Modeling**

The Stagni et al. [10] detailed mechanism is selected for reduction. The fully automatic reduction tool ARCANE [11] is used to derive a novel ARC scheme. The initial target fuel is a partially cracked ammonia ternary blend $X_{NH_3} = 0.4285$, $X_{H_2} = 0.4285$, and $X_{N_2} = 0.1430$ in moles. Quantities Of Importance (QOI) are evaluated under a wide range of operating conditions using the premixed freeflame canonical case. That is, 1-20 bar, 300 - 700 K and 0.6-1.6 for pressure, temperature, and equivalence ratio, respectively. The final mechanism, named ARC NH3H2N2_15_211_5_HV for future reference, has 15 species, 211 elementary steps (107 reactions), and 5 QSS species. Both reduced mechanisms showed accurate predictions compared to the basis mechanism and experimental data [12]. *A posteriori* validation of the mechanism is also performed for the whole range of ammonia-hydrogen binary blends ($0 < X_{H_2} < 1$), as well as for all ternary blends derived from ammonia decomposition ($0 < \gamma < 1$). For this exercise, with reported maximum error ($e_{rel}^{max}$) 10% for $S_L^0$, 1% for $T_{adb}$, and 15% for final $Y_{NO}$.

**Turbulent Combustion Modeling**

Since the TFLES model is parameterized through 1D reference quantities ($\delta_{th}^0$, $S_L^0$, etc.), it may be used in partially-premixed configurations provided that the reference fresh state can be locally computed. Usually, the assumption of a mixture constant Lewis number allows TFLES input parameters, namely $\phi$, to be computed from the local composition of the reacting mixture using Bilger's mixture fraction definition [13]. While this simplification is generally considered acceptable for heavy carbonated/conventional fuels, it does not hold for lighter fuels with preferential diffusion. Furthermore, the assumption of a uniform fuel composition breaks down in multi-fuel applications with differentiated injection. Given the above limitations, the Multi-Fuel TFLES (MF-TFLES) model is proposed as an extension of the TFLES model for multi-fuel multi-injection partially-premixed combustion. The objective of these modifications is to cover four scenarios adequately: 1) a homogeneous fuel blend with species having similar diffusion properties; 2) a homogeneous fuel blend with distinct diffusion properties; 3) a differentiated injection of two fuel streams; 4) and the individual injection of each stream (first fuel, second fuel, and oxidizer). To address such scenarios, three reference tanks with assumed constant composition are considered: fuel tank one, fuel tank two, and a single oxidizer tank. A transport equation for a passive scalar per fuel tank is then explicitly resolved by the LES solver in parallel to the transport equations of the chemical scheme species. Each of the two transported passive scalars has, in such a case, the objective to track the evolution of its corresponding fuel mass fraction as if the flow were non-reactive. This work adds the notion of the so-called blend index as the fourth dimension of the reference value look-up table space, allowing for the first time to the best of the authors' knowledge, to correctly parameterize the thickening field in dual-fuel partially-premixed configurations.

**Application to a Swirl Burner**

The partially-premixed swirled burner experimentally studied by Mashruk et al. [8] is specifically addressed in this work for assessment of the proposed modeling strategy. This burner, displayed in Fig. 1, operates at atmospheric pressure using a fully premixed $NH_3$-air mixture injected at 288K. Further downstream, a partially-premixed regime is obtained by injecting pure hydrogen, as highlighted in blue in Fig. 1.b. A globally lean equivalence ratio of around 0.65 and a binary $NH_3$-$H_2$ fuel blend with $X_{H_2} = 0.3$ are prescribed.

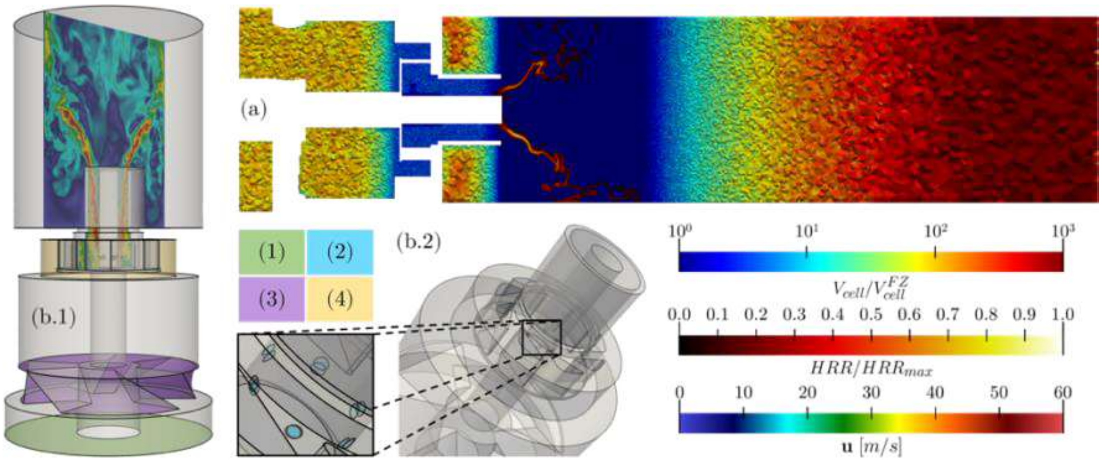


**Figure 1.** Numerical setup. (a) The cell size distribution normalized by the flame zone target cell volume is displayed. (b.1,b.2) The primary (1) and secondary (2) injection, as well as the axial (3) and the radial (4) swirler are highlighted.

In terms of the computational domain, the burner is described using an 87 million cell unstructured tetrahedral mesh. The primary injection inlet is highlighted in green in Fig. 1.b. The secondary injection system is represented by short-length pipes keeping the injecting tube section untouched. The simulations have been performed using the parallel explicit cell-vertex code AVBP [14]. The MF-TFLES turbulent combustion model previously detailed is implemented. A 9-point discretization is chosen for a correct flame front resolution. The walls upstream from the burner are no-slip and adiabatic. Heat losses are prescribed for the quartz tube walls.

A reference simulation using the TFLES model (Fig. 2.a) was used to reconstruct the *a priori* effects of the MF-TFLES model (Fig. 2.b). Next, another simulation using the MF-TFLES approach allowed for an *a posteriori* comparison (Fig. 2.c). Misidentification of the of the flame location and misestimation of the required thickening led to the flame being attached to the injector ring.

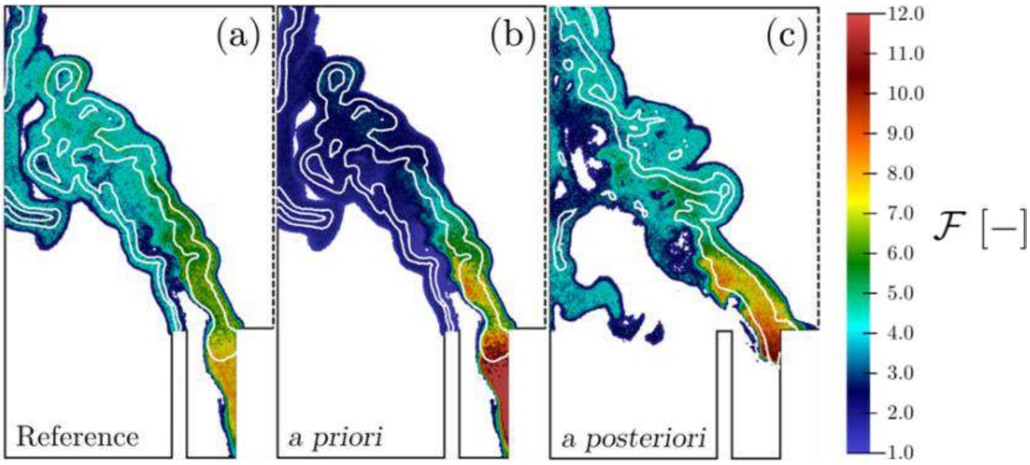


**Figure 2.** Impact of the MF-TFLES model. TFLES reference appears attached to the injector ring, this branch disappears on the MF-TFLES case (right).

$NO_X$ emissions are highly dependent on the temperature of the burner walls. Therefore, a third simulation considering adiabatic wall for the quartz tube was performed to evaluate heat loss effects on flame topology and $NO_X$ emissions.

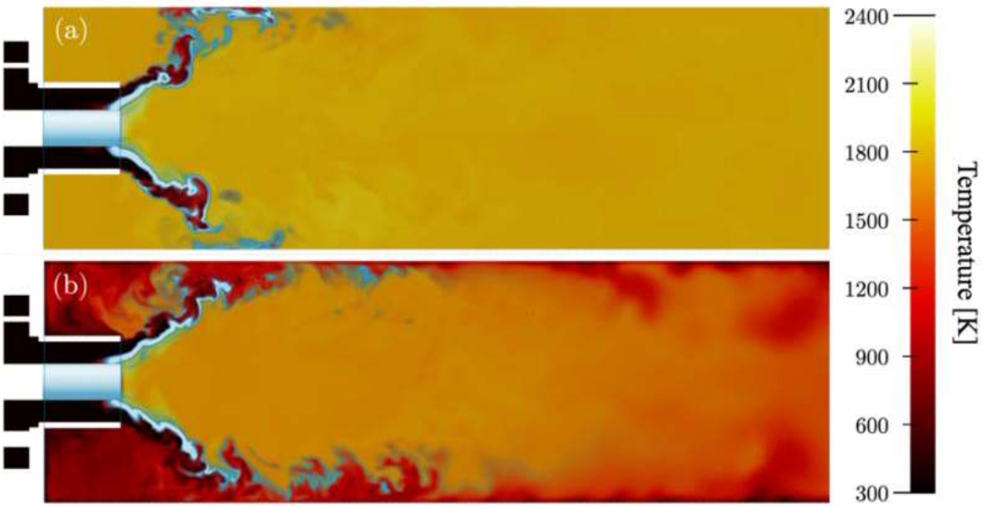


**Figure 3.** Longitudinal slice of the domain colored by temperature for (a) the adiabatic case and (b) the heat loss case. The flame position is highlighted in blue.

In both simulations, the differentiated injection of hydrogen results in a higher temperature in the central region where the main flame branch attaches to the central rod as illustrated by Fig. 3. This behavior is reinforced by the preferential diffusion of the $H_2$ molecule. The first observable effect of the heat loss treatment is a temperature reduction near the outer ring of the injector (Fig. 3.b). This ultimately leads to the detachment of the outer branch of the flame which 'unfolds' downstream in the quartz tube. Since the outer branch is detached, unburned fuel leaks downstream through the low temperature zone near the wall.

As expected, $NO_X$ production is also affected by heat losses. Time-averaged solutions considering 20 ms of physical time were used to investigate pollutant concentration. Two sampling methods allowed to evaluate the homogeneity of the pollutant concentration in the radial direction (Fig. 4). In the first approach, pollutant concentrations were sampled using point-probes distributed along the longitudinal axis of the quartz tube. In the second approach, the cross-section integral was computed for 30 planes situated in the same abscissa positions as the point-probes. This analysis revealed strong gas composition heterogeneities in the heat loss case.

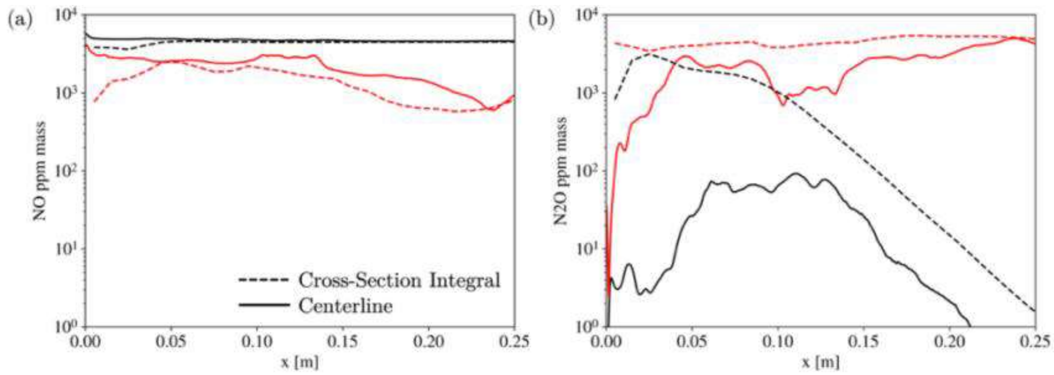


**Figure 4.** Concentration profiles for (a) NO and (b) $N_2O$ along the quartz tube. Black lines and red lines represent the adiabatic and heat loss case, respectively.

## Conclusion

In this work, an ARC for $NH_3$-$H_2$-$N_2$ combustion is coupled to an extension of the TFLES model dealing with differential diffusion and space-inhomogeneous fuel blends in multi-fuel, multi-injection partially-premixed flames. The premixed ammonia-air flame with differentiated hydrogen injection operated at Cardiff University is chosen as a simulation test case. The effect of the MF-TFLES model is evaluated against a reference TFLES model, flame topology effects were observed. Thermal effects are evaluated by comparing an adiabatic case and a heat loss case. Heat losses modify the flame topology resulting in strong exhaust composition heterogeneities and therefore pollutant concentration highlighting the importance of probe placing in experimental configurations. These results may further be used to describe the impact of the flame-turbulence interaction on pollutant formation pathways in decarbonized combustion.

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