

NO Formation in MILD Combustion: Sensitivity to Finite-rate Chemistry Combustion Models and Kinetic Mechanisms

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Moderate and Intense Low-Oxygen Dilution (MILD) combustion, also known as Flameless [1] or HiTAC [2] combustion, provides higher combustion efficiency and lower NO_x emissions compared to traditional combustion. Numerical simulations of MILD combustion burners are crucial for addressing the modelling challenges related to this rather new combustion technology. One of the challenges to address is the prediction of NO_x emissions: the thermal mechanism [3], the major contributor to NO formation in most of the conventional combustion systems, is suppressed by the lower temperatures and the absence of large fluctuations in MILD combustion, so that NO formation is controlled by other routes, such as the prompt NO [4] and/or N₂O intermediate [5]. Moreover, in the presence of H₂-containing fuels, the NNH pathway is relevant, as shown for a lab-scale burner [6] and an H₂-enriched semi-industrial scale [7].

The current work presents a numerical study about the role of kinetic mechanisms and mixing models on the prediction of NO formation in Moderate or Intense Low oxygen Dilution (MILD) combustion. The impact of the uncertainty associated with the choice of these specific sub-models is assessed. The KEE [8] and GRI2.11 [9] mechanisms are employed for the chemistry. The Eddy Dissipation Concept (EDC) [10] and the Partially-Stirred Reactor (PaSR) [11] models are used for turbulence-chemistry interactions. These mixing models assume that the chemical reactions take place in confined regions of the computational cell. In the PaSR model, the volume fraction of the reacting structures depends both on mixing and chemical time scales. Particularly, the factor κ provides the partially stirred condition, being the volume fraction of the reactive zone, defined as the ratio between the chemical time scale τ_c and the sum of the chemical time scale and the mixing time scale τ_{mix} : $\kappa = \tau_c / (\tau_c + \tau_{mix})$. Therefore, the appropriate choice of these time scales becomes essential to ensure predictivity for the numerical simulations. The PaSR model allows the calculation of different chemical time scales for diverse species. Results show that the use of an engineering approach that employs different chemical time scales in the PaSR model leads to significant improvements in the prediction of NO formation.

The measurements of NO mass fractions in the Adelaide Jet in Hot Co-flow (AJHC) burner are used for validation. The Adelaide Jet in Hot Co-flow (AJHC) burner is a jet with a simple geometry that emulates MILD conditions via the injection of a heated and vitiated co-flow [12]. Measurement data for experiments with a central jet fuel of CH₄ and H₂ (in equal proportions on a molar basis), different co-flow oxygen levels (3%, 6% and 9% as mass fractions), and different fuel jet Reynolds numbers (5k, 10k and 20k) are available. The AJHC burner with 3% co-flow oxygen content and Re=10k is chosen for assessing the performances of different sets of sub-models. The experimental profiles used for comparison include both the mean values and the error bar with 99.99% confidence interval associated with a Student's distribution for the true mean value [13]. Along with the NO mass fraction profiles, temperature and OH mass fraction profiles are considered since they have an impact on the chemistry and thus the emissions of NO.

Steady and unsteady numerical simulations of the burner have been performed with the commercial CFD code Ansys Fluent 19. Turbulence is modelled via the RANS approach, using the κ - ϵ model with a modified value of the parameter $C_{1\epsilon}$, as put forth by Dally et al. [14]. Li et al. [15] showed that both EDC and PaSR models are suitable for high-fidelity numerical simulations of the AJHC. Therefore, these mixing models are used to treat turbulence-chemistry interactions. In particular, two versions of the EDC model with modified values of the time scale constant C_τ ,

i.e., 1.5 and 0.82, are used, and the PaSR model is employed with two different approaches for the estimation of the mixing time-scale (τ_{mix}), i.e., static [16] and dynamic [17].

Two chemical mechanisms are considered: i) the KEE58 skeletal mechanism, with 17 species and 58 reversible reactions; and ii) the GRI2.11 detailed mechanism, with 49 species and 279 reactions. The KEE scheme does not include the NO_x formation pathways. Therefore, in this case, the NO_x modelling is handled by the post-processing tool of Ansys Fluent, which assumes that NO_x chemistry has negligible influence on the predicted flow field, temperature, and major combustion product concentrations. Thermal NO formation is modelled using a Finite Rate (FR) approach with a simplified one-step mechanism obtained from the Zeldovich scheme by assuming a steady state for the N radicals and taking the O and OH radical concentrations from the local O and OH radical mass fractions available from the KEE scheme. Prompt NO formation from methane is modelled according to De Soete [18]. For MILD combustion regimes, the description of NO formation at low-temperatures requires the incorporation of additional mechanisms, i.e., the N₂O intermediate mechanism [19] and the NNH route [20]. The NNH route is not directly available in Fluent; therefore, it has been implemented by means of a UDF following Konnov et al. [21]. For all the above NO formation kinetic rates the Arrhenius equation is integrated over a probability density function (PDF) for temperature to account for the effect of temperature fluctuations on the mean reaction rates. The assumed PDF shape is that of a beta function [22] and is evaluated through the temperature variance, which is solved by means of a transport equation.

Results

A comparison between measured and simulated temperatures, OH and NO mass fractions at several axial locations, i.e., 60/120/200 mm, in radial direction is performed for the test case at Re=10k with a 3% O₂ in the co-flow. In Figure 1, the comparison is shown for the simulation cases employing the KEE scheme with the post-processing for NO and four different TCI models, namely: i) EDC with $C_\tau=1.5$; ii) EDC with $C_\tau=0.82$; iii) PaSR with the static approach for the mixing time-scale; iv) PaSR with the dynamic approach for the mixing time-scale.

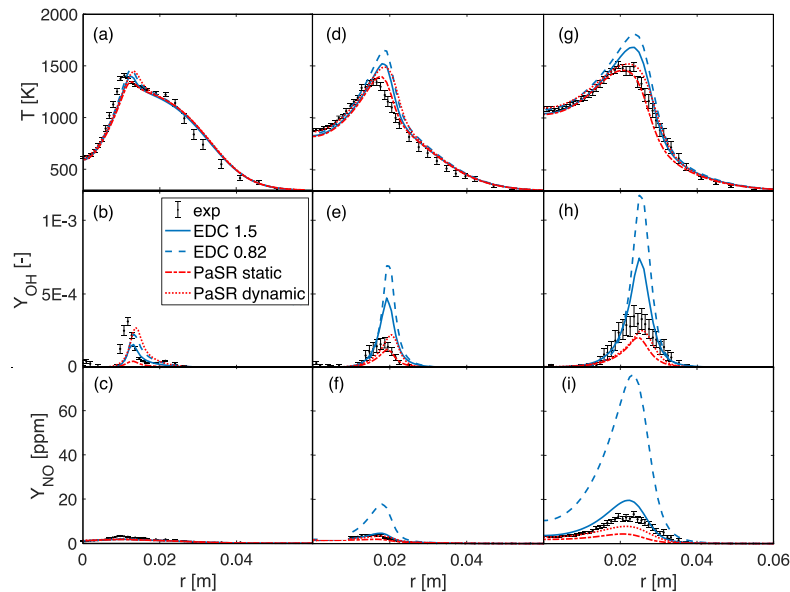


Figure 1. Sensitivity analysis of mixing models using the KEE scheme on temperature, OH and NO mass fraction profiles, at axial locations $z=60$ mm (a,b,c), 120 mm (d,e,f), and 200 mm (g,h,i).

The results show that better predictions of temperatures, OH and NO mass fractions are achieved by the dynamic PaSR model, despite an overestimation of the peak temperature at 120 mm and an overall underestimation of the NO profiles.

In Figure 2, the comparison is shown for the simulations employing the GRI2.11 scheme and four different TCI models, same as listed above.

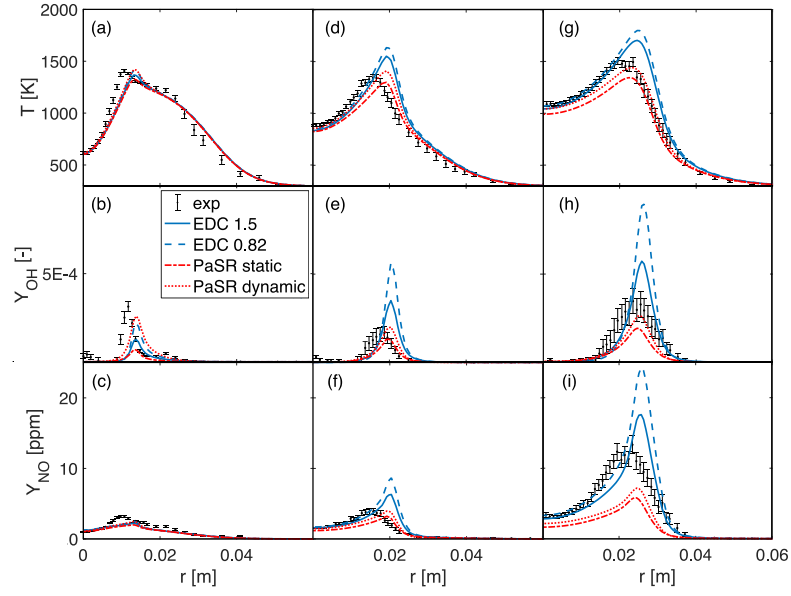


Figure 2. Sensitivity analysis of mixing models using the GRI2.11 scheme on temperature, OH and NO mass fraction profiles, at axial locations $z=60$ mm (a,b,c), 120 mm (d,e,f), and 200 mm (g,h,i).

The results show that better predictions of temperatures, OH and NO mass fractions are obtained by the dynamic PaSR model. The overprediction of the peak temperature at 120 mm is reduced, although an overall underestimation of the NO profiles still persists.

In Figure 3, the comparison between experiments and simulations is shown for the following cases: i) static PaSR with KEE scheme; ii) dynamic PaSR with KEE scheme; iii) static PaSR with GRI2.11 scheme; iv) dynamic PaSR with GRI2.11 scheme.

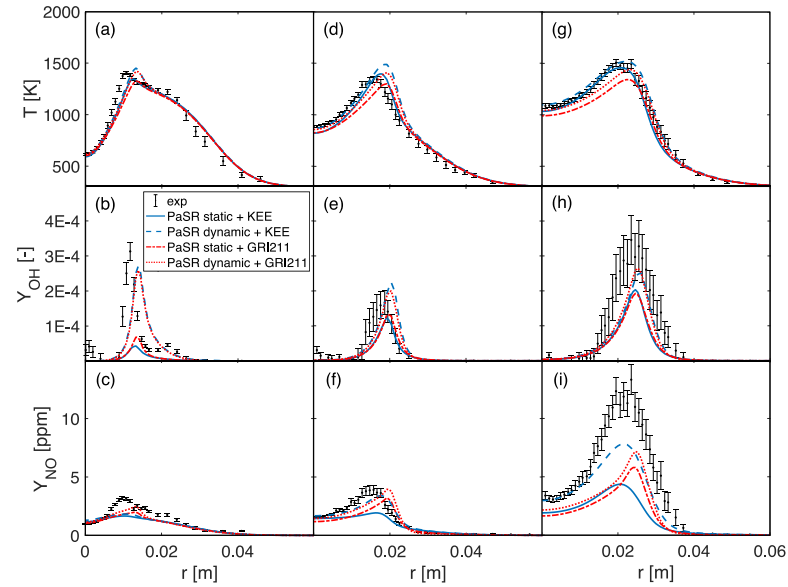


Figure 3. Sensitivity analysis of PaSR models and kinetic mechanisms on temperature, OH and NO mass fraction profiles, at axial locations $z=60$ mm (a,b,c), 120 mm (d,e,f), and 200 mm (g,h,i).

Similar predictions of temperatures, OH and NO mass fractions are achieved by the dynamic PaSR model with either KEE or GRI2.11. To overcome the underestimation of the NO profiles, an engineering adjustment of the PaSR model is put forth and accounts for the different time scales of the fuel-oxidizer reactions and NO_x formation pathways, such as thermal and NNH. The κ factor is set to 0.75 for NO species. This adjustment is applied to the case that employs dynamic PaSR and GRI2.11, and the obtained results are shown in Figure 4.

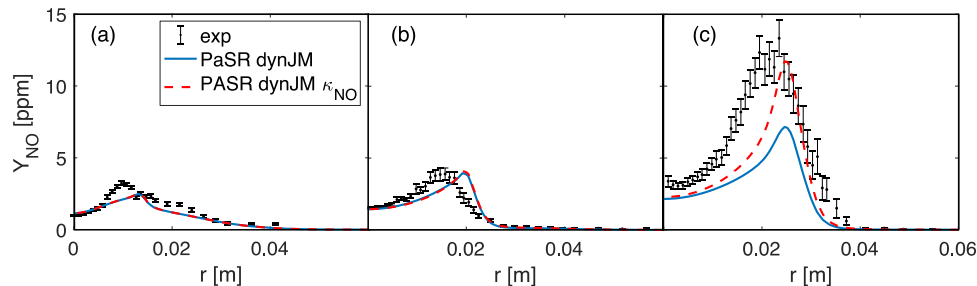


Figure 4. Experimental and numerical NO mass fraction profiles at axial locations $z=60$ mm (a), 120 mm (b), and 200 mm (c).

Preliminary conclusion

The role of selected finite-rate chemistry combustion models and kinetic mechanisms on the prediction of NO emissions in the Adelaide Jet in Hot Co-flow (AJHC) burner has been studied. The choice of a combustion model has a higher impact on NO predictions than the choice of a kinetic mechanism. The PaSR model with a dynamic estimation of the mixing time scale gives the best predictions of temperature, OH mass fractions and NO emissions. Comparable results are obtained by employing either KEE or GRI2.11 as chemical mechanisms. They both provide an overall underestimation of NO concentrations. An adjustment of the dynamic PaSR model has been introduced to overcome the underprediction of NO by accounting for the different time scales of the oxidation reactions and the reactions involved in the NO formation pathways. An elevated chemical time scale results in the reactive volume fraction κ of the computational cell approaching a unitary value. A value of κ of 0.75 for NO induces a better estimation of the NO mean source term, which comes from the PaSR closure, and eventually results in better predictions of NO emissions in the AJHC burner. A subsequent analysis will show that the proposed adjustment of the PaSR model is beneficial for the thermal NO and the NNH intermediate routes, being these two pathways the slowest ones within the NO_x chemistry. This approach shows its usefulness when detailed mechanisms, accounting for the NO_x chemistry, are employed in numerical simulations of burners operating under combustion regimes like MILD combustion, where the overlap between chemistry and mixing time scales occurs.

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