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Chemistry Reduction for Modelling Flameless Combustion of Ethylene

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Flameless combustion is a novel combustion technology able to ensure high combustion efficacies with low pollutant emissions thanks to the dilution of reactants, usually achieved through recirculation of combustion products. The technology has been successfully applied in several processes and has been found to be able to handle a large variety of fuels, including low grade fuels, industrial by-products and hydrogen. Further development of this innovative combustion technology would benefit of Computational Fluid Dynamics (CFD) tools; however, modelling flameless combustion is much more challenging than conventional flames, because of the strong coupling between turbulent mixing and chemical kinetics. In particular the chemical kinetics plays a fundamental role, even though there no common opinion on the degree a mechanism can be reduced. Some useful works may be found on flameless burners fed with methane, but there is lack of information on different fuels.

The present work describes the numerical modelling of an ethylene jet flame issuing in a hot coflow burner, emulating flameless combustion and fully characterised in literature, with the scope of investigating the potential for chemistry reduction in the context of flameless combustion. A Principle Component/Variable Analysis is used to investigate the most important species in the chemical mechanism and subsequently a dimension reduction technique based on the Rate-controlled constrained-equilibrium (RCEE) principle is applied to the detailed mechanism to be coupled to the CFD code, in order to make simulations more affordable. Results indicated that the use of Principle Component/Variable Analysis leads to a good choice of the variables to be retained as results with the reduced scheme were found to be consistent with those obtained with the full mechanism.

1. Introduction

Flameless combustion ensures high combustion efficiencies with low pollutant emissions thanks to the dilution of reactants. The technology has been successfully applied in several processes and has been found able to handle a large variety of fuels, including low-grade fuels, industrial by-products and hydrogen.

Further development would benefit of Computational Fluid Dynamics (CFD); however, flameless combustion modelling appears still very challenging, because of the strong coupling between turbulent mixing and chemical kinetics. Chemical kinetics plays a fundamental role in such a combustion regime due to the relevance of finite-rate chemistry. However, the degree of complexity required still represents an open question and the potential degree of simplification of comprehensive chemical mechanisms is an important area of investigation for such a regime. Some interesting works may be found on flameless burners fed with methane, but there is lack of information on different fuels.

The present work describes the numerical modelling of an ethylene jet flame issuing in a hot coflow burner, which is fully characterized in literature and emulates flameless combustion conditions (Medwell and Dally, 2008). Model uncertainties due to boundary conditions, turbulence and combustion model are discusses. Special attention is devoted to the possible degree of simplification of comprehensive chemical mechanisms, through a reduction technique based on constrained equilibrium (Keck, 1990). Two approaches are benchmarked for selecting represented species: a Principal Component/Principal Variable Analysis of the correlation structure of the thermo-chemical state of the system (Parente et al., 2009) which allows identifying

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the most energy-carrying variables, and an approach based on the separation of slow and fast reacting species, based on the spectral decomposition of the isothermal Jacobian of the state-space source terms (Isaac et al., 2009).

2. Geometry and numerical model

The Adelaide Jet in Hot Coflow burner modelled in this work has been experimentally studied by Medwell and Dally (2008) and it is shown for sake of clarity in Figure 1a. The burner consists of a fuel jet nozzle, which has an inner diameter of 4.6 mm located at the center of a perforated disc in an annulus, with inner diameter of 82 mm which provides nearly uniform composition of hot oxidizer coflow to the reaction zone. The entire burner is placed inside a wind tunnel introducing room temperature air at the same velocity as the hot coflow.

The fuel, injected at T = 305 K, is a mixture of ethylene and air, with a volumetric fraction of Y_{C2H4} = 25 %. The coflow is at T = 1,100 K and with O_2 volumetric fraction Y_{O2} = 3 % (and Y_{CO2} = 3 %, Y_{H2O} = 10 %). The available data consist of the mean and root mean square (rms) of temperature and some key species such as OH and CH_2O. Measurements errors are of the order of 10 % for temperature, while species measurements can be only employed for semi-qualitative validation.

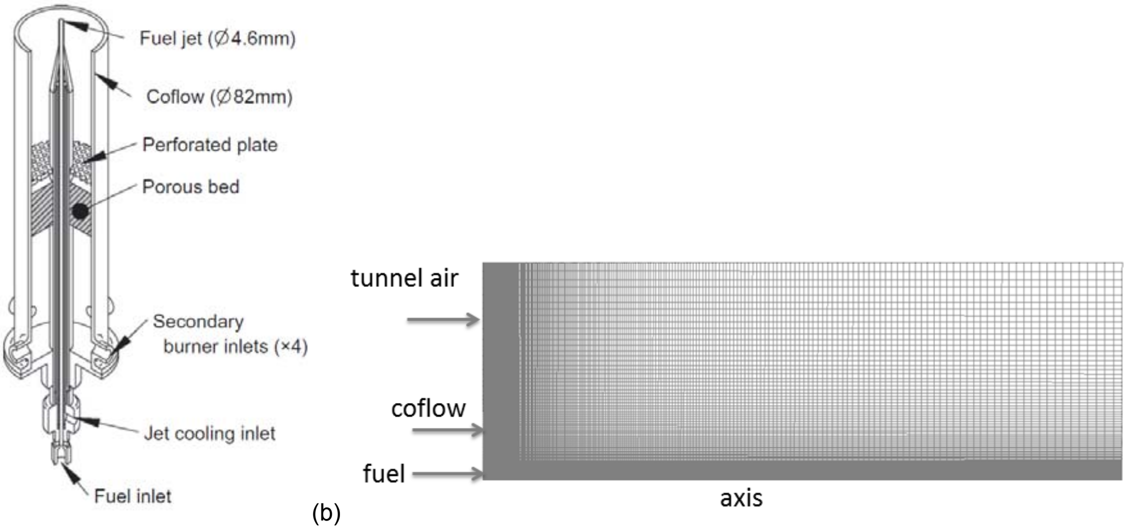

Figure 1: (a) Scheme of the burner (from Medwell and Dally, 2012) and (b) computational grid.

The geometry of the JHC burner allows to use a 2D axisymmetric domain (Figure 1b), constructed starting from the burner exit. The computational grid was chosen from a grid independency study and was structured with 25,800 cells. Richardson extrapolation was applied to determine the discretization error, which was estimated below 5 % for both velocity and temperature measurements.

Steady-state FANS equations were solved with a finite volume scheme using the commercial CFD code ANSYS Fluent® v. 15. Several turbulence models (TMs) were investigated: the standard k-ε model, two k-ε model variants for round jets consisting in a modified value of the first constant of ε equation (i,e. $C_{\varepsilon 1}$ =1.6) Morse, 1981) or a modified ε equation (Pope, 1978) and the Explicit Algebraic Reynolds Stress Model (EASM) (Wallin, 2000).

The GRI-3.0 oxidation mechanism (Smith et al., www.me.berkeley.edu/gri_mech), without the NO subset, thus with 219 reversible reaction and 36 species, was used to treat ethylene oxidation. The interaction between turbulence and chemistry was handled through the Eddy Dissipation Concept (EDC) model (Magnussen, 2005). A discussion of the model capability is also provided by Magnussen et al. (2013). However different values of the fine structure residence time constant C_t were employed as recently it was suggested by both Aminian et al. (2012) and De et al. (2011) to use larger C_t values that the standard C_t =0.41 for MILD combustion conditions. Aminian et al (2012) suggested to use C_t = 1.5 for the Adelaide Jet in Hot Coflow fed with CH_4/H_2 mixture, whereas De et al. (2011) suggested C_t = 3 for a similar burner, i.e. Delft Jet in Hot Coflow, The discrete ordinate (DO) method together with the Weighted-Sum-of-Gray-Gases (WSGG) model with coefficients taken from Smith et al. (1982) was employed to solve the radiative transfer equation (RTE) in 16 different directions across the computational domain.

A zero-shear stress wall was adopted at the side boundary, instead of a more realistic pressure inlet/outlet condition, to facilitate calculations. However, as the tunnel air was considered wide enough, the choice of the boundary condition does not affect the flame structure (Aminian et al., 2011)

Uniform velocities were set for air, fuel jet and coflow oxidizer. The turbulence levels of all three inlet streams was adapted to better capture the development of the mixing layers using values suggested by Christo and Dally (2005) for the fuel inlet, by Frassoldati et al. (2010) for the coflow and Aminian et al. (2011) for the tunnel air.

3. Dimensional reduction

Numerical simulations are carried out with the objective of investigating the potential for chemistry reduction in the context of MILD combustion. To this purpose, full chemistry simulations were compared to reduced ones, performed using a dimension reduction technique based on the Rate-controlled constrained-equilibrium (RCEE) principle (Keck, 1990). In RCCE, a reduced number of species transport equations is solved, for the so-called represented species or constraints. The rest of the chemical state space is reconstructed assuming that they are in a chemical equilibrium, constrained by the concentration of the transported species. The main steps involved in the RCCE approach can be summarized as follows:

- Resolution of a smaller number of species transport equations than the number of species in the full chemical mechanism.
- Reconstruction of the remaining species in the mechanisms by solving a free Gibbs energy minimization problem for chemical equilibrium.
- Equilibration of the species subject to the constraint given by the constrained species mass fractions.

In ANSYS FLUENT, the RCCE approach is referred to as Dimension Reduction (DR). The combination of DR with the ISAT algorithm allows achieving significant CPU time reduction with respect to full calculations.

However, the accuracy of the approach strongly depends on the choice of the constraints for the selection of the species for which transport equations are solved.

In the present work, an approach based on the use of Principal Variables (Parente et al., 2009) is investigated for the selection of the constraints. The Principal Variable (PV) approach is directly linked to Principal Component Analysis (PCA) (see Figure 2). In PCA, the objective is to optimally reduce the dimensionality of a data set, consisting of several observations of a large number of variables (in this case the variables identifying the chemical state space of the system, temperature, pressure and composition). Through an eigenvalue problem, PCA allows finding (Jolliffe, 1986) a few (<< *p*) derived variables, called principal components (PCs), which preserve most of the information given by the original data. The PCs are linear combinations of the original variables, which are uncorrelated and derived so that the preserved variance is maximal. PV represents an attempt to gain a physical understanding from principal component analysis. PV algorithms try to link the PC back to a subset of the original variables, which satisfies the optimality property of PCA, namely that retained variance is maximized. For additional details on PCA and PV approaches, refer to Parente (2009).

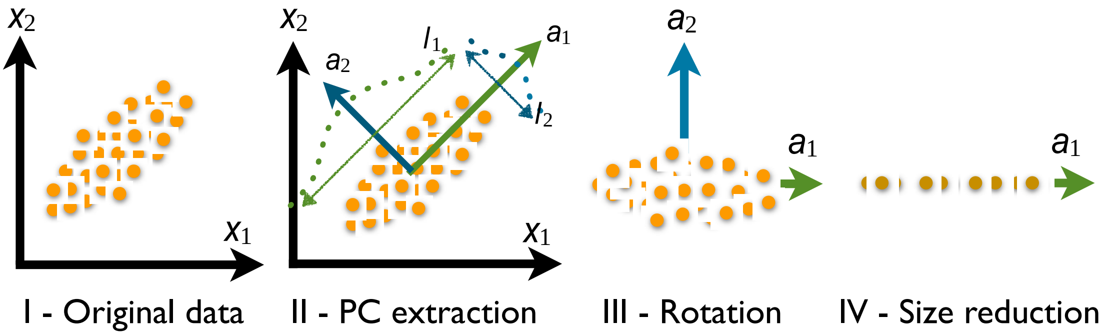

Figure 2: PCA reduction process

The application of the PV approach to the data sets obtained from the full chemistry calculation allows determining the constraints or represented species for the application of the RCCE approach. In the following such species will be indicated as principal variables.

4. Results

Figure 3a compares the experimental radial profile of temperature taken at *z* = 35 mm and those obtained with the full GRI.30 mechanism but using different constants for the fine structures residence times, C_t , of the EDC model. It can be noticed that the standard constant (C_t = 0.41) leads to an overestimation of temperature by of approximately 70 K, as also observed in literature for other MILD combustion conditions (Aminian et al., 2012). Increasing the constant up to C_t = 1.5 or C_t = 3.0 largely improves the predictions. As for OH concentration, the standard EDC was found to over predict the OH peak at *z* = 35 mm by approximately 7 times, whereas the C_t = 1.5 or C_t = 3.0 leads to peak values consistent with experimental ones. For that reason it was decided to retain the C_t = 1.5, in accordance with Aminian et al. (2012) for subsequent calculations.

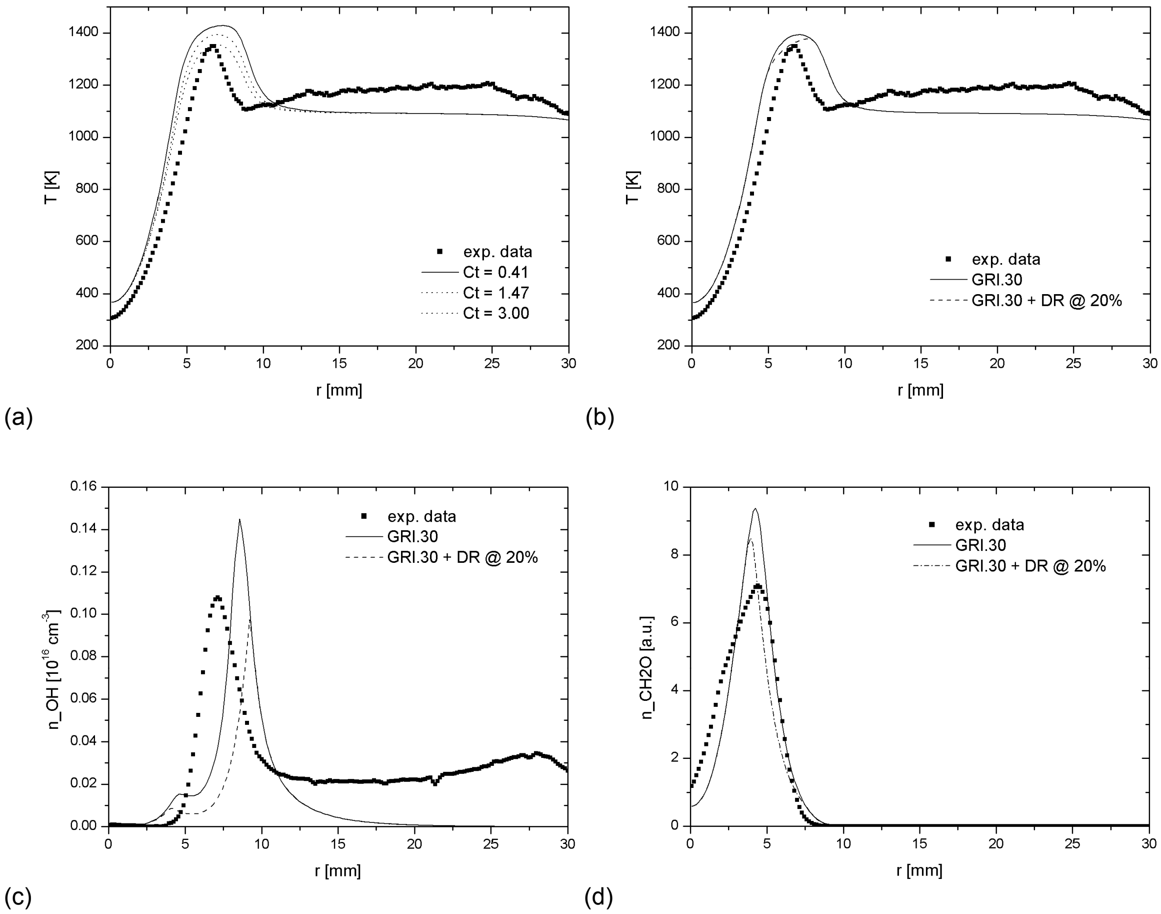

Figure 3: Comparison between experimental radial profiles at z = 35 mm of (a, b) temperature, (c) OH and (d) CH2O concentration and those predicted with: (a) different EDC C_t constants and full GRI.30 mechanisms; (b,c,d) full and reduced GRI.30 and EDC C_t = 0.41.

The PV approach described in section 3 was applied to the calculations carried out with full chemistry and the modified model constant C_t = 1.5. Species concentrations and temperature in each cell of the computational domain were extracted to a matrix for the subsequent application of PCA and PV. In the present work, the PV approach is carried out with the objective of selecting a subset of species, which minimizes the reconstruction error of the low-dimensional representation. The analysis provides two sets of variables, the retained and the discarded ones, respectively, which identify the constraints and the constrained species for the application of the dimension reduction technique. Several analyses were performed, to achieve various reduction degrees. Only the results provided by a reduction of 20 % with respect to the original number of species is presented here. For this case, the PCA/PV approach lead to the identification of the following constrained specie: C_3H_8, C_2H_2, C_2H_3, CH_2CO, C_2H_5, CH_2<s>, C and CH_4. Calculations were then performed transporting only 24 principal variables, i.e. the constraints.

Figure 3b compares the temperature profiles obtained with the full GRI.30 mechanism with that obtained through dimensional reduction.

The estimated peak temperature is the same for the two mechanisms even though the high temperature region is larger with the dimensional reduction. This is confirmed by the distribution of temperature which is shown in Figure 4 for the two mechanisms.

Measured and predicted OH concentrations at *z* = 35 mm are reported in Figure 3c and indicate lower OH levels obtained with the reduced model. Some differences can be also noticed from the OH distribution shown in Figure 5.

The evaluation of CH_2O is also very satisfactory and both models give results in good agreement with experimental data (see Figure 3d).

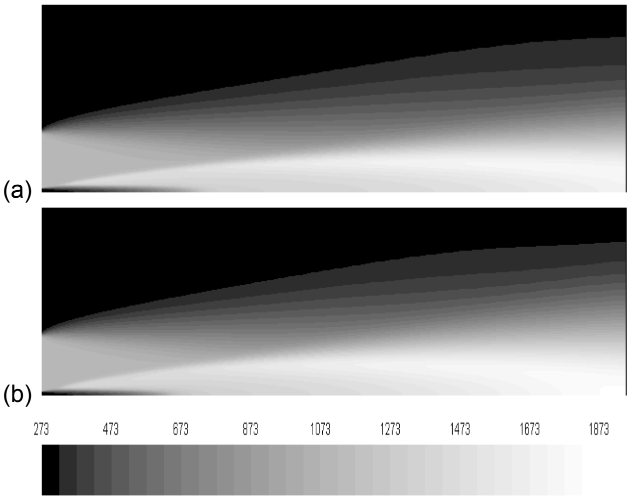

Figure 4: Distribution of temperature evaluated with (a) GRI.30 and (b) dimensional reduction.

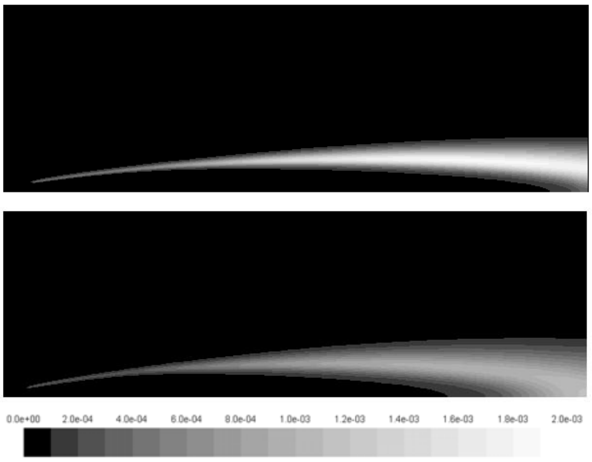

Figure 5: Distribution of temperature evaluated with (a) GRI.30 and (b) dimensional reduction.

4. Conclusions

A methodology based on preliminary Principle Component/Variable Analysis and subsequent reduction of detailed oxidation mechanisms based on Rate-Controlled Constrained-Cquilibrium (RCEE) principle is proposed for the modelling of a flameless burner fed with ethylene. Preliminary results indicate the potential of the proposed approach for the selection of an appropriate subset of principal variables controlling the evolution of the system. Indeed, results obtained with a 20 % reduction of the original number of species transport equations lead to numerical results consistent with those obtained with the full kinetic mechanism.

Future work will focus on the benchmark of different techniques for the selection of the constraints for the RCCE approach as well as on the maximum degree of simplification achievable without significant accuracy loss.

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