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Assessment of Adaptive chemistry via pre-partitioning of composition space and mechanism reduction for the Simulation of MILD Combustion

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ABSTRACT

Moderate or Intense Low-oxygen Dilution (MILD) combustion has become increasingly important as it ensures high combustion efficiency with low pollutant emissions. However, modelling this regime is much more difficult than for conventional flames due to the strong coupling between turbulent mixing and chemical kinetics. The use of detailed chemistry has therefore become an essential requirement in CFD (Computational Fluid Dynamics) simulations, although reducing the chemical mechanism is an effective way to dramatically reduce the computational cost. One globally reduce mechanism is suboptimal as it nees to be comprehensive for all the thermochemical conditions, yet the on-the-fly chemistry reduction carries also some computational overheard. Therefore the methodology SPARC (Sample-Partitioning Adaptive Reduced Chemistry) for the inclusion of detailed chemistry is investigated in this work. It alleviates the overhead associated to the on-the-fly reduction by building a library of locally reduced mechanisms in the pre-processing step.

The current paper focuses on an a-priori assessment of SPARC for simulating Delft jet in hot co-flow (DJHC) burner fed with natural gas. The analysis will assess the level of reduction reached in the preprocessing step with different chemical mechanism, Gri3.0_HT, POLIMIC1C3_HT and POLIMIC1C3HT_NOx. First the clustering algorithm is tested with datasets of different sizes. Then the impact of partitioning parameters on the degree of reduction is quantified.

INTRODUCTION

In the last decade there has been a growing interest for more efficient energy systems to fulfill the demand for heat and power in industry while respecting the today's emission reduction targets. Several solutions have been proposed in the past years. Among them Moderate or Intense Low-oxygen Dilution combustion (MILD) showed to be promising due to its high combustion efficiency with low pollutant emissions. Such regime can be achieved by preheating the reactants above their self-ignition temperature and by recirculating inert combustion products to the reaction zone in order to dilute the flame. As a consequence the temperature field is more uniform than in conventional combustors with no high temperature peaks. NO_x and soot formation are therefore suppressed, owing to the lean conditions and low temperatures. Further development of this new combustion technology would be supported by innovative CFD tools. However, modelling this combustion mode is much more difficult than for traditional regimes because to the strong coupling between turbulent mixing and chemical kinetics in which the chemistry plays a primary role.

The use of finite rate approaches has therefore become a fundamental prerequisite for CFD simulations. However, for realistic combustion systems it is still prohibitive, due to the large number coupled equations characterizing comprehensive mechanisms. In fact, the amount of operations required to solve the chemistry in CFD depends on the size of the oxidation mechanism, which reflects the level of complexity for integrating the system of nonlinear ODE (Ordinary Differential Equations), and the number of cells in the mesh, which indicates the number of time such system must be integrated (1).

A reduction in the computational cost for these two aspects is achieved by the TDAC method (2), which couples ISAT (3) and DAC (4). The ISAT algorithm aims to reuse previously computed results, thus reducing the effect of the number of cells, while DAC finds a skeletal mechanism at runtime, thus alleviating the effect of the mechanism size (1) allowing the inclusion of finite rate approaches in turbulent combustion modelling.
Several studies regarding the use of on-the-fly chemistry reduction techniques have been reported in the literature. Zhiyi Li et al. obtained a speed-up factor more than 10 when using TDAC with a comprehensive kinetic mechanism for simulating the DJHC burner operating in MILD conditions (5). Reduction models (DRG, DRGEP, and PFA) have been adapted for run-time reduction and results show that the contribution of tabulation is more important for small mechanisms, while reduction plays a major role for large ones (5). The time saving using TDAC with detailed chemistry is very promising, thus making it possible to be used for industrial or semi-industrial applications.

Since the present work focuses on the inclusion of detailed chemistry and since, for detailed mechanisms, the contribution to the speed-up given by reduction is more significant than the one by tabulation, the methodology called SPARC (Sample-Partitioning Adaptive Reduced Chemistry) for the inclusion of detailed chemistry in CFD is investigated in the present work. The methodology consists on building a library of reduced mechanisms in a pre-processing step, i.e. before carrying out the CFD simulation (6; 7). The training dataset is generated by means of low dimensional simulations (0-D and 1-D) using detailed chemistry. The corresponding composition space (species mass fractions and temperature) must adequately cover the composition space which is expected to be visited during the multidimensional CFD simulation. The composition space can be so pre-partitioned in a prescribed number of clusters, ensuring that each individual cluster is sufficiently homogeneous from the kinetic point of view, i.e. that is possible to correctly describe all the samples belonging to it with a single locally reduced mechanism (6).

Before performing the chemical step, each cell is classified using an unsupervised-learning algorithm suitable for pre-partitioning, in order to identify the cluster to which the cell belongs and to assign the corresponding reduced mechanism (6). This implies a double benefit on the simulation: on one side it allows to include locally reduce mechanisms able to reproduce local phenomena, on the other side it reduces the cost associated to the adaptive reduction step which can be significant when more complex reduction methods are adopted. A significant reduction in computational time can is achieved: results showed speed-up factors for the chemical step of ≈ 4 for laminar flames with a comprehensive mechanism (6), but the performances are expected if bigger mechanisms are adopted and unconventional regimes are investigated: since larger and stiffer kinetic mechanisms are required, a higher level of chemical reduction and speed-up factor, can be reached (5). Moreover, since the reduction takes place in the preprocessing step, it can be performed by using method characterized by high overhead which allow to monitor global observables of the mixtures, such as ignition delay time.

The current paper focuses on an a-priori assessment of SPARC methodology for simulating Delft jet in hot co-flow (DJHC) burner fed with natural gas. The analysis will assess the level of reduction reached in the preprocessing step with different chemical mechanism, Gri3.0_HT (53 species), POLIMIC1C3_HT (114 species) and POLIMIC1C3HT_NOx (159 species). First, the clustering algorithm is tested with data set of different sizes and then the effect of partitioning parameters on the degree of reduction is quantified.

METHODOLOGY

The methodology, called SPARC (Sample-Partitioning Adaptive Reduced Chemistry) (6; 7), consists of three steps in the pre-processing stage followed by the multi-dimensional CFD simulation (6) :

1. Dataset generation: by solutions of 0D and 1D simulations using detailed chemistry;
2. Clustering of the composition space via Local Principal Component Analysis (LPCA);
3. Generation of reduced mechanism for each cluster;
4. CFD simulation.

In the present work the preprocessing step is analysed (steps 1 - 3) giving an a-priori assessment of the clustering partition and level of reduction obtained.

Dataset generation

In order to produce a good classification of the flame regions, the dataset must properly cover the composition space which is expected to be visited during the simulation.
The training of LPCA algorithm and the chemical reduction can be done either using dataset from previous detailed simulations, or simple 0-D/1-D simulations replicating the combustion phenomenology.
Combustion datasets typically contain variables of different numerical ranges (in the present work both temperature and mass fractions of chemical species). Centering and scaling the datasets is crucial in order to balance the importance of all state-space variables (7). After centering and scaling, the original set of thermochemical variables ξ is standardized, and a new set of variables Φ is obtained and can be used as training data.

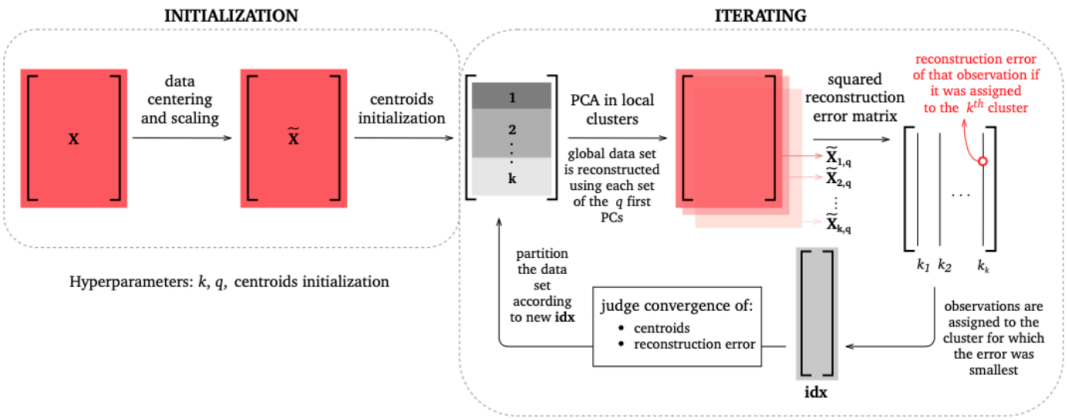

Figure 1 scheme of VQPCA algorithm. This figure was adjusted with permission from ref (8) .

Clustering via LPCA

In the context of dimensionality reduction, data driven approaches are gaining attention due to their ability of extracting important features from highly multidimensional datasets.

PCA is a statistical technique that allows a large number of interdependent variables to be reduced to a smaller number of uncorrelated variables, while preserving as much of the original data variance as possible (9). Given a dataset $\mathbf{X}$ of n rows (observations) and p columns (variables), the covariance matrix $\mathbf{S} = \frac{1}{n-1}\mathbf{X}^T\mathbf{X}$ can be calculated and then to decomposed: $\mathbf{S} = \mathbf{ALA}^T$. The eigenvectors of the covariance matrix, that represented the columns of the $\mathbf{A}$ matrix, are referred to as Principal Components (PCs) while the eigenvalues, located on the principal diagonal of the $\mathbf{L}$ matrix, represent the portion of variance they account for. Dimensionality reduction is achieved by considering only a subset of the PCs, whose size is chosen as a function of the variance considered (6). Since each PC is associated to a certain percentage of variance by the magnitude of its associated eigenvalue, it is possible to quantify the variance explained by a subset of *PCs* by calculating the ratio: $t_q = \sum_{j=1}^{q} j_j / \sum_{j=1}^{p} l_j$, where q is the chosen size of the subset and l_j is the $j-th$ eigenvalue obtained from the decomposition of the covariance matrix $\mathbf{S}$ (6). If the reconstruction process leads to errors that are comparable with the ones obtained for the training data, we can expect that PCA captures the essence of the physical processes underlying the system (9).

In case of strongly non-linear systems, such as reactive flows, a large number of PCs may be needed to describe the system correctly. To overcome this limitation, it is possible to use a locally linear approach called Local Principal Component Analysis (LPCA) (10). LPCA is an unsupervised-learning algorithm suitable for partitioning the data-space in different groups where the variables share common features. Subsequently PCA is performed in each cluster to find a reduced representation. When the dataset is partitioned and PCA is performed locally in each cluster, a different set of eigenvectors is tied to each identified cluster. Those local PCs can better represent the physics or the underlying features of a particular cluster by capturing the local variance instead of the global one (11). Data clustering, prior to applying local PCA, can be performed with any algorithm of choice. The techniques adopted in this paper is the Vector Quantization PCA (VQPCA) algorithm (12; 8) where each observation of the dataset is assigned to the cluster in which that observation has the minimum reconstruction error $\varepsilon_{GRSE,k} = ||\tilde{x}_i - \tilde{x}_{i,q}||$. Vectors $\tilde{x}_i$ and $\tilde{x}_{i,q}$ are, respectively, the original and the reconstructed, via PCA, i-th centered and scaled observation of the dataset.
The VQPCA process is visualized in Fig. [1] and algorithm summarized as follow:

1. Initialization: for each cluster the initial centroids $\bar{x}_k$ are chosen;
2. Partition: by means of the calculation of $\varepsilon_{GRSE,k}$ each observation is assigned to a cluster;
3. Update: the cluster centroids are updated on the basis of the partitioning performed in step 2;
4. LPCA is performed in each cluster;
5. Steps 2. to 4. are iterated until one of convergence criteria is met.

Mechanism Reduction

A reduced mechanism is generated for each cluster following the partition process described above: a single reduced mechanism for cluster $k-th$ is formed by the merging the reduced mechanisms of the single observations belonging to cluster $k-th$. If the partitioning (through LPCA) is performed correctly, it is expected that the mechanisms generated for each sample of a cluster are very similar, since their compositions are also similar. Therefore, their union is expected to result in a final mechanism that is not significantly larger than the mechanism of the individual samples (6), both in terms of number of species and number of reactions in which those species are involved.

In the present work chemistry reduction is performed using `pyMARS` (Python-based Model Automatic Reduction Software) (13), a software package that implements and

applies methods from the literature to reduce chemical kinetic models for combustion applications. pyMARS implements three "skeletal" reduction methods that select and remove species and reactions with negligible contribution: DRG, DRGEP and PFA represent the kinetic system as a graph, where nodes are species and directed, weighted edges represent the dependence of one species on another, through their participation in reactions (13). To eliminate species, a cutoff threshold is applied to eliminate negligible connections. For all three methods, the algorithm iteratively increases the cutoff threshold to remove species until reaching a user-specified error limit on the ignition delay time. This results in the algorithm including a minimum number of species/reactions that ensures the replication of global observables of the mixture, rather the production of individual species (13).

The DRGEP method was selected, as it showed better performances in MILD combustion in term of accuracy and computational cost (5). Lu and Law (14) first developed the DRG method as a way to use graphs to represent kinetic models, where species are nodes and directed edges represent dependence. Pepiot and Pitsch (15) first presented the DRGEP method as an improved version of DRG that better represents the indirect dependence of species down graph pathways. In DRGEP, the weight of the directed edges, which represents the dependence of one species on another, is given by the direct interaction coefficient r_{AB} in eq. (1):

$$
\begin{aligned}
r_{\mathrm{AB}} &= \frac{\left|\sum_{i=1}^{N_{\mathrm{reactions}}} \nu_{A,i}\omega_i\delta_{Bi}\right|}{\max(P_{\mathrm{A}}, C_{\mathrm{A}})} \\
\delta_{\mathrm{B},i} &= \begin{cases} 0, & \text{if } i \text{ reaction involves B} \\ 1, & \text{otherwise} \end{cases} \\
P_{\mathrm{A}} &= \sum_{i=1}^{N_{\mathrm{reactions}}} \max(0, \nu_{\mathrm{A},i}\omega_i) \\
C_{\mathrm{A}} &= \sum_{i=1}^{N_{\mathrm{reactions}}} \max(0, -\nu_{\mathrm{A},i}\omega_i)
\end{aligned}
\tag{1}
$$

where N_{reaction} is the number of reactions, $\nu_{\mathrm{A,i}}$ is the overall stoichiometric coefficient for species A in reaction i, PA and CA denote the production and consumption of species A respectively. ω_i is the overall reaction rate of the reaction i. Following DRGEP, the mechanism is further reduced thought Sensitive Analysis. Although sensitivity analysis (16) becomes computationally expensive when applied to the large kinetic models (e.g., 100-1000 of species). Instead, we can use the above graph-based methods to first remove a significant fraction of species, and then analyze a portion of the remaining species using the interaction coefficient values to sort and identify which to consider (13). In particular, the DRGEP-based sensitivity analysis (DRGEPSA) has shown great efficacy for reduction. In the DRGEPSA approach (17), an upper threshold value ε^* is applied to the overall interaction coefficients, and any species that would be removed (but were remaining following the DRGEP reduction) are considered "limbo" species. All of the limbo species are then considered for removal one-by-one (13) though one of the two sensitivity analysis algorithms implemented: *initial* and *greedy* (18). In the *initial* algorithm, the errors induced by the removal of the limbo species are evaluated only once, initially, and then the species are considered for removal in ascending order. In contrast, the *greedy* algorithm reevaluates the induced error of remaining limbo species after each removal, and chooses the species with the lowest induced error at that point (13). In the present work the *greedy* algorithm is selected.

Validation Case and Numerical Setup

The DJHC burner is chosen to test the SPARC methodology potential for CPU time saving in CFD of MILD combustion. The DJHC burner has a central fuel jet with an inner diameter of 4.5 mm. The hot co-flow is provided by a secondary burner mounted in an outer tube with an inner diameter of 82.8 mm (5). A detailed description of the DJHC can be found in the studies of Oldenhof (19) and Sarras (20). Experimental values of temperature and velocity values are available for validation. Dutch natural gas (19) is used as central jet fuel with Re = 4100. Other properties of the fuel and co-flow jets are repoerted in Tab. 1.

Unsteady Reynolds-averaged Navier−Stokes (URANS) simulations with local time step (LTS) were performed using the standard k−ε turbulence model (21). The simulation domain extends 225 mm axially (z) downstream the burner outlet and 80 mm toward the radial direction (r). The entire domain is discretized with a structured axisymmetric 2D mesh.
The inlet boundary conditions for temperature and velocity were taken from experimental values measured 3 mm downstream of the jet exit, as suggested by De (22). The reactingFoam solver is used. The GRI3.0 (23), POLIMIC1C3HT and POLIMIC1C3HT_NOx (24) mechanisms are considered, as they are characterized by increasing complexity, i.e. number of species and reactions.

$\dot{V}_{\mathrm{fuel}}$ [nL/min]	$T_{\mathrm{co\text{-}flow,\,max}}$ [K]	$Y_{\mathrm{co\text{-}flow},O_2}$	Re_{fuel}
16.1	1540	0.076	4100

Table 1 Physical Properties of considered Natural gas.

RESULTS AND DISCUSSION

In this section the impact of different parameters on the LPCA clustering solution and the level of reduction reached for each cluster is assessed.

Clustering of the composition space

In this work, since the goal is to provide an assessment in term of reduction level and the size of the problem is not prohibitive, the training data are chosen from three CFD solutions using the three detailed mechanisms described above.

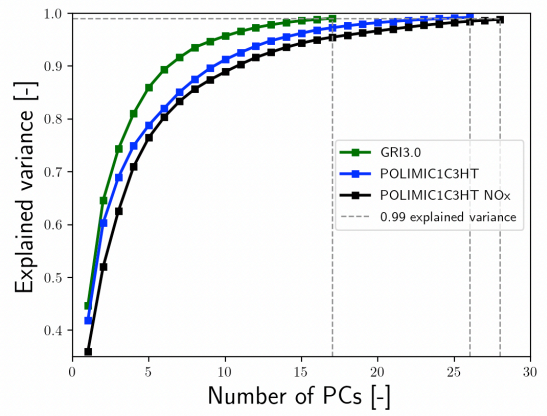

Figure 2 Explained global variance as function of the PCs considered in PCA method for th three considered training datasets.

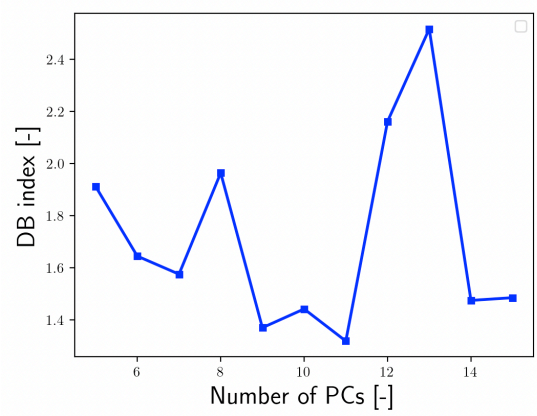

Figure 3 DB index as function of number of Principal Components for Gri30 mechanism, k=6.

The parameters needed to perform LPCA are the number of PCs considered (n_{PCs}) and the number of clusters (k). The effect of both parameters are investigated in this section.

Following a rule of thumb of data analysis, once selected the dataset, the number of PCs needed to explain a sufficient portion of variance by applying LPCA (i.e. performing PCA locally in each cluster), is in between a third and a half of the number of PCs needed to explain the same variance. Hence, a first global PCA analysis (i.e. considering the whole computational field) has been performed on the starting data set. The trend of the explained global variance in function of number of PCs for the three different mechanisms is reported in Fig. 2. Results highlight how, increasing the size of the data set, a larger number of PCs is needed to explain 99% of the global variance in global PCA. According to this it is possible to establish a range of PCs which have to be taken in consideration for further evaluation.

To determine the number of clusters k, the Davies-Bouldin (DB) index (25) is computed in order choose the optimal number of clusters (k) for partitioning. The Davies-Bouldin index is the ratio between the scatter within each cluster and the distance between clusters. It measures how well the clustering separates the dataset into subgroups. Moreover, the DB index evaluation can be used as criterion to better determine other parameters for LPCA, as the n_{PCs} itself. In fact, even though the reconstruction error $\varepsilon_{GRSE,k}$ is monotone decreasing with respect to n_{PCs}, the dimentinality adopted for the recontruction have to be chosen properly in order to reproduce just the important features of the data, so that the observations can be effectively divided in groups. Both k and n_{PCs} can be so investigated and the optimum parameters couple is chosen as the one which leads to the minimum of DB index as shown is Figure 3. For each dataset two solutions, with different number of k, are selected to asset different reduction levels after the partition. Tab. 2 collects the selected couples of parameters (k and n_{PCs}) for the three different data set and the corresponding DB index.

Mechanism	k	n_{PCs}	*DB index*
GRI3.0	6	11	1.3704
	9	8	1.3831
POLIMIC1C3HT	6	5	1.4741
	8	10	1.6530
POLIMIC1C3HT_NOx	8	12	1.7354
	10	12	1.736

Table 2 Selected number of PCs and clusters for the three considered mechanism.

The solution of the clustering algorithm is reported in Figures 4, 5 and 6 for the three datasets. The plot shows the map of the flame where each point is assigned to a cluster according to the local conditions.

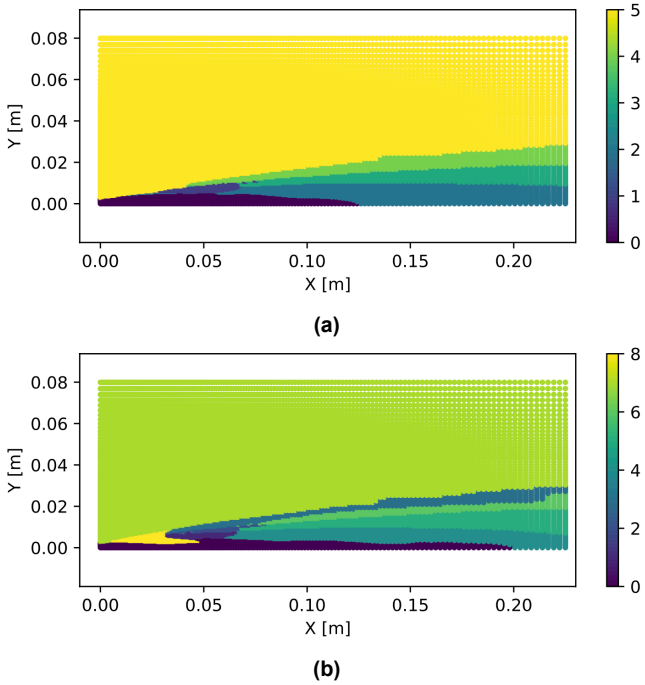

Figure 4 Clustering Solution of the VQLPCA algorithm plotted on mesh with Gri3.0 chemical mechanism (53 species): (a) k=6 and nPCs=11, (b) k=9 and nPCs=8.

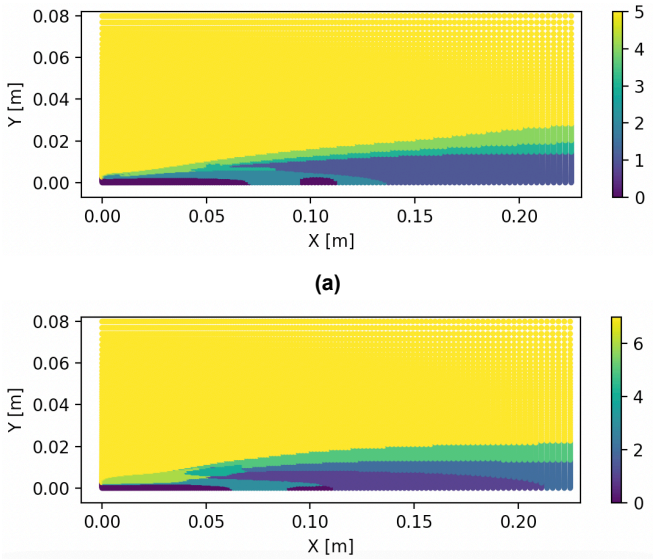

Figure 5 Clustering Solution of the VQLPCA algorithm plotted on mesh with POLIMIC1C3HT chemical mechanism (114 species): (a) k=6 and nPCs=5, (b) k=8 and nPCs=10.

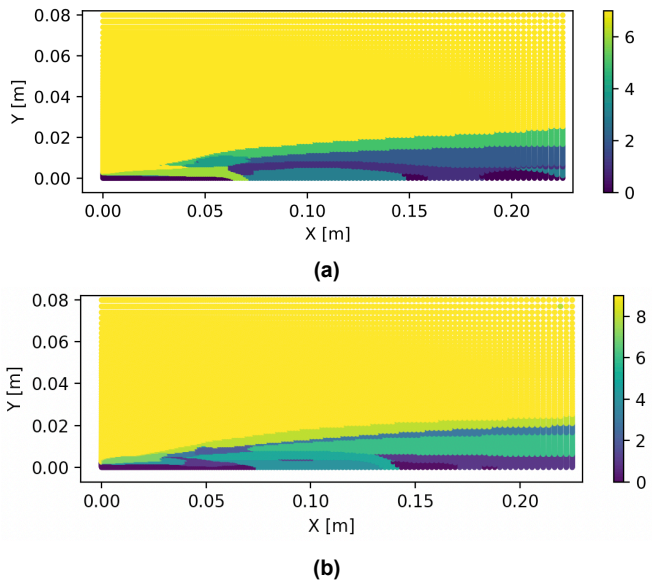

Figure 6 Clustering Solution of the VQLPCA algorithm plotted on mesh with POLIMIC1C3HT_NOx chemical mechanism (159 species): (a) k=8 and nPCs=12, (b) k=10 and nPCs=12.

CFD results are also reported for a comparison: Figures 7a, 7b and 7c show the temperature, the heat released and the OH fields respectively.

Thanks to the scaling of the training data, the clustering does not reflect just the temperature distribution. In fact, because the partition is made on the whole composition space, it is possible to identify points which are close under the thermo-chemical point of view, i.e. described by the same physics phenomena, which means same locally reduced mechanism. As a consequence both the inlets of air and co-flow, which have really different temperature (Fig 7a), are collected in the same region where mixing is predominant and the reactivity is still low (as it can been seen in Fig 7b). Altogether clustering results can be considered satisfactory as, for all the considered configurations, the methodology was able to identify main regions of the flame : inlet of fuel, inlet of co-flow and cold gases, interaction region between fuel and coflow, and the hot reacting region where the main part of the heat is released.

It as to be noticed that the effective size and shape of those clusters is strongly dependent from the dataset of choice: for bigger mechanism more detailed flame regions are identified in the interaction zone between fuel and the fast co-flow jet. How to correctly generate local mechanisms for those regions would represent an important step forward in the understanding of CFD of MILD combustion. In fact modelling this regime is much more challenging than conventional flames just because of the chemical kinetics which plays a fundamental role respect to turbulent mixing.

The level of reduction obtained for the configurations reported above is presented in the next section.

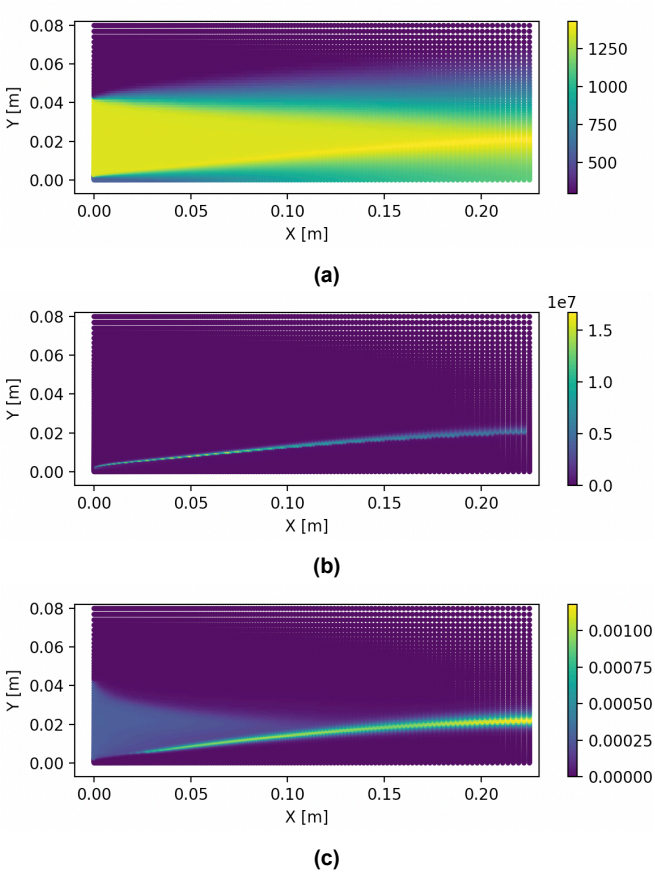

Figure 7 CFD solution for DJHC case: (a) Temperature [K] (b) Heat released through combustion [J/kg/s], (c) OH mass fraction [-].

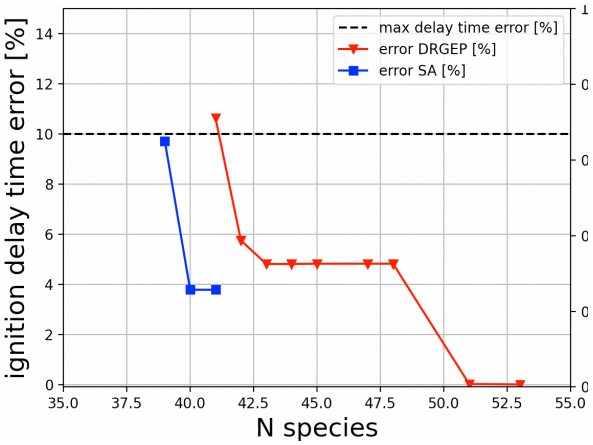

Figure 8 DRGEP and SA reduction loops for cluster 7, 53sp k=9 Pcs=8

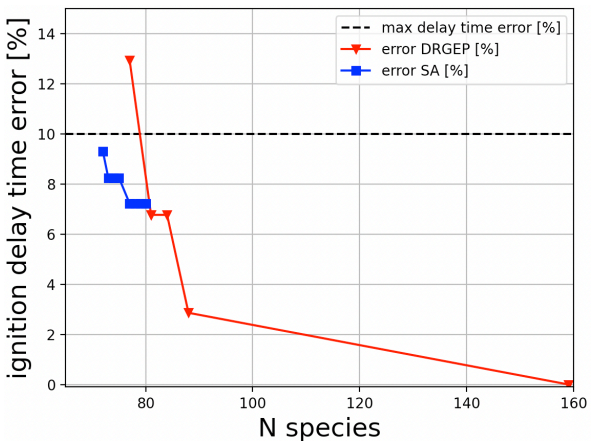

Figure 9 DRGEP and SA reduction loops for cluster 6, 159sp k=8 Pcs=12

Mechanism Reduction

For each cluster, the reduction is performed using DRGEP and sensitive analysis. The max error on the ignition delay time of the mixture err_{max} is set at 10%. The initial threshold is set 0.01 and two species for fuel and oxidizer (CH4 and O2) are defined as targets species for the DRGEP method. Subsequently also Sensityve Analisys (with upper-threshold equal to 0.4) is performed to remove other species while respecting the constraint on the ingnition delay time.

Figures 8 and 9 show the DREGP and SA reduction loops for cluster 5 for 53 species dataset and cluster 7 for 159 species dataset respectively. The ignition delay time error at any iteration (*err*) is reported in function of the number of species for the two subsequent reduction loops. In the first case (Fig. 9), after the DRGEP stage 19% of the species is removed and, after SA the mechanism accounts of 39 species respect 53 in the original mechanism (-26 %). This level of reduction grows with the size of the mechanism. In the case of comprehensive mechanism with 159 species (Fig. 8) a great number of species has been removed by DRGEP ($\approx$ 49%) and the further SA stage selected other six species to be neglected. 54% of species the original species have been removed and the final locally reduced mechanism counts of 73 species. Figures 9 and 8 also show the advantage of having different reduction methods working subsequently: more species are selected for evaluation through different algorithms and this combined investigation leads to strongly reduced mechanisms which, at the same time, are able to respect the constraint on global observable. It must be notice that the targets represent just a criterion which leads to the investigation of species to remove, while checking that the condition on the ignition delay time is respected. Hence, if the focus is only on such global observable, the set of target which leads to a minimum number of species in the reduced mechanism is preferred.

Following this criterion the active species after the two reduction stages are reported for each clusters for the three datasets in Tab 3.

N Global	N Locally Reduced										
	k	0	1	2	3	4	5	6	7	8	9
53	6	34	38	30	0	0	42				
53	9	28	26	26	0	0	0	0	39	34	
114	6	44	0	53	0	57	0				
114	8	43	0	0	38	0	0	52	0		
159	8	48	0	0	0	29	49	73	0		
159	10	46	0	0	50	0	0	0	80	30	0

Table 3 Number of species in the local reduced mechanism in each cluster for the selected configurations.

The obtained level of reduction is very promising with a view of performing CFD simulation. In fact, the number of active species is directly related to the number of ODE which need to be solved. An important result is the identification of clusters with zero species: groups of observation for which no ignition has be detected from autoignition simulations performed during the reduction loops. Those clusters can be defined non reactive and the species can be just transported during CFD without solving any ODE. Moreover using big mechanisms (114 and 159 species) helps identifying a big non reactive region in the mixing zone between air and co-flow where no chemistry needs to be solved.

CONCLUSIONS

In the current paper, the DJHC burner is numerically investigated by means of RANS simulations using three different detailed mechanisms. The aim is to give an assessment of the SPARC methodology, in the prepocessing step, in term of degree of reduction obtained by focusing on global observables, such as the ignition delay time. The composition space covered during the simulation is analyzed thought LPCA data-driven clustering method and the algorithm's ability of extracting important features from high dimensional dataset has been proven.

The effect of parameters necessary for applying LPCA has been investigated. The DB index is a good indi-

cator to evaluate the clustering solution a posteriori and it offers a guideline to choose the best couple of parameters k and n_{PCs}.

The locally reduced mechanism for each cluster have been generated by a combination of DRGEP and SA methods. For the graph-based method the set of target species is chose as the one which leads to a more reduced local mechanism. The further reduction stage through SA offers a valuable additional option to remove species not investigated in the previous step. Therefore a combination of the two methods is always preferred.

The obtained degree of the reduction is bigger when larger detailed mechanism are considered. Moreover the identification of non-reactive zones makes possible not to resolve the chemistry in some parts of the domain. Those results make the methodology very promising for reducing computational cost in detailed CFD simulations.

Further development of the proposed methodology will comprehend the generation of the training data though 1-D simulations and the assessment on the speed up obtained for CFD simulations.

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