Assessment of On-the-Fly Chemistry Reduction and Tabulation Approaches for the Simulation of Moderate or Intense Low-Oxygen Dilution Combustion

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Supporting Information

ABSTRACT: The current paper focuses on the numerical simulation of the Delft jet in hot co-flow (DJHC) burner, fed with natural gas and biogas, using the eddy dissipation concept (EDC) model with dynamic chemistry reduction and tabulation, i.e., tabulated dynamic adaptive chemistry (TDAC). The central processing unit (CPU) time saving provided by TDAC is evaluated for various EDC model constants and chemical mechanisms of increasing complexity, using a number of chemistry reduction approaches. Results show that the TDAC method provides speed-up factors of 1.4–2.0 and more than 10 when using a skeletal mechanism (DRM19) and a comprehensive kinetic mechanism (POLIMIC1C3HT), respectively. The directed relation graph with error propagation (DRGEP), dynamic adaptive chemistry (DAC), and elementary flux analysis (EFA) reduction models show superior performances when compared to other approaches, such as directed relation graph (DRG) and path flux analysis (PFA). All of the reduction models have been adapted for run-time reduction. Furthermore, the contribution of tabulation is more important with small mechanisms, while reduction plays a major role with large ones.

INTRODUCTION

Moderate or intense low-oxygen dilution (MILD) combustion technology has gained increasing attention in the past few decades.1−3 It is characterized by very strong mixing between reactants and products. As a result, the fuel conversion rate is higher and the temperature field is more uniform than in conventional combustion, thereby reducing CO, soot, and NOx emissions.1,2 As a result of the intensive mixing and reduced temperature peaks, reactivity in MILD combustion is reduced to the point that the overall oxidation process is controlled by chemical kinetics.3 Thus, combustion models which consider finite-rate chemistry are required when modeling such a combustion regime. In the present work, the eddy dissipation concept (EDC) combustion model1 is used.

The number of species and reactions in a reaction mechanism grows with the number of carbon atoms in the fuel component. As a consequence, the central processing unit (CPU) time associated with chemistry resolution can be very significant for industrial applications involving realistic fuels. In this framework, the use of pre-reduced or pre-tabulated mechanisms may not be able to capture the dynamic phenomena, such as extinction and re-ignition, accurately. Therefore, on-the-fly/dynamic chemistry reduction/tabulation methods are essential to alleviate the calculation burden for finite-rate chemistry approaches. Various investigations based on the use of on-the-fly chemistry reduction techniques are reported in the literature. Tosatto et al.6 used a transport-flux-based directed relation graph (DRG) model in a two-dimensional simulation of axisymmetric co-flow flames. A speed-up factor of about 5 was reported for a steady case, while a factor ranging from 10 to 20 was obtained for a time-dependent oscillating flame. Yang et al.7 investigated the use of dynamic adaptive chemistry (DAC) for efficient chemistry calculations in turbulent reacting flow modeling. The incurred errors in the predicted temperature and species concentrations were effectively controlled, and a speed-up of 3−6 times was achieved. Zhang et al.8 and He et al.9 conducted homogeneous charge compression ignition (HCCI) engine simulations with the element flux analysis (EFA) approach. The size of the kinetic scheme was significantly reduced while maintaining reliable accuracy. A new adaptive reduction method that emphasizes solution error control was proposed by Oluwayemisi et al.10 and tested in combustion computational fluid dynamics (CFD) with autoignition [one-dimensional (1D), zero-dimensional (0D)] and two-dimensional (2D) laminar flame examples. They concluded that the proposed decoupled species and reaction reduction (DSRR) model is as fast as the directed relation graph (DRG) model. Moreover, DSRR offers improved solution accuracy control. In 2016, Xie et al.11 introduced a Jacobian-aided rate analysis (TSRA) approach. The chemical species are categorized through this approach plus time scale, and no problem-dependent parameters, such as starting species, are required. They drew the conclusion that the TSRA method yields
more accurate predictions of ignition delay time and composition compared to DRG based DAC approach.

The dynamic reduction method allows for significant CPU savings; however, the cost associated with the direct ordinary differential equation (ODE) integration can still be significant if the number of species in the reduced mechanisms is non-negligible. This is especially important in some non-conventional or engine combustion applications, where detailed chemistry is necessary and the number of species set is still large, even after reduction. The coupling with the dynamic tabulation approach provides a solution to this issue. Tang et al. combined chemical dimension reduction and tabulation with a methodology called in situ adaptive tabulation—rate-controlled constrained equilibrium (ISAT—RCCE). The chemistry reduction with RCCE is performed by specifying a set of represented species, and the selection of species is performed using a new greedy algorithm with local improvement (GALI). The ISAT species, and the selection of species is performed using a new RCCE calculations show good agreement with the direct strained equilibrium (ISAT) chemistry dimension reduction and tabulation with a method.

The present work is to provide an overall assessment of the TDAC approach under MILD conditions. All of the numerical results represent the mean of three replicates, and the number of species in the reduced mechanisms is non-negligible. This is especially important in some non-conventional combustion with the tabulated dynamic adaptive chemistry (ISAT) algorithm and chemical mechanism reduction methods, including DRG, directed relation graph with error propagation (DRGEP), DAC, elementary fluid analysis (EFA), and path flux analysis (PFA). The TDAC process is visualized in Figure 1. ISAT stores the initial composition and the solution of the ODE integration with so-called “leafs” and retrieves them with a linear approximation when the composition space is within an ellipsoid of accuracy (EOA), avoiding the need for a direct integration. When ISAT needs growth or addition instead of retrieve, it provides first the composition \( y^* \) to the mechanism reduction method, which simplifies the mechanism and provides a reduced set of species compositions \( y_{\text{red}} \) to the ODE solver. The solver computes the reaction mapping \( R(y_{\text{red}}) \), and then ISAT builds the full reaction mapping \( R(y^*) \) from \( R(y_{\text{red}}) \). If the differences between the existing and newly build reaction mappings are within the user-defined tolerance of \( \epsilon_{\text{ISAT}} \), the EOA will grow to include the new mapping. Otherwise, a new “leaf” will be added to include the newly computed mappings. Furthermore, a time-step-specific scale factor is defined, to give more control on the time-step variations.

The five different reduction approaches (DRG, DRGEP, DAC, PFA, and EFA) implemented in the TDAC code are all based on a network

fine structures \( (\gamma_i) \) and the mean residence time of the fluid inside the fine structures \( (\tau^*) \) are described as

\[
y_i = C_i \left( \frac{k_i \epsilon}{\rho \gamma} \right)^{1/4}
\]

and

\[
\tau^* = C_i \left( \frac{k_i \epsilon}{\rho \gamma} \right)^{1/2}
\]

where \( k \) is the kinematic viscosity and \( \epsilon \) is the dissipation rate of kinetic energy, \( k_i \text{ and } C_i \) are two model parameters, set to 2.1377 and 0.4083, respectively. Finally, the mean reaction rate of species \( i \) is expressed as

\[
\omega_i = -\frac{\rho \gamma}{\tau^* (1 - \gamma_i)} (\bar{Y}_i - Y_i^*)
\]

This expression of the EDC model was proposed by Magnussen in 2005 and, therefore, denoted as EDC2005 in the present work. The term \( \bar{Y}_i \) in eq 3 is the mean mass fraction of species \( i \) between the fine structures, \( Y_i^* \) and the surrounding fluid, \( Y_i^{\text{ff}} \). The mass fraction \( Y_i^{\text{ff}} \) of species \( i \) inside the fine structures is estimated solving a PFR equation, with the residence time of \( \tau^* \). A detailed kinetic mechanism is used in the PFR calculation. In the present study, the DRM19 (19 species + N2 and AR, 58 reactions), GRI3.0 (53 species, 325 reactions), and POLIMIC1C3HT (107 species, 2642 reactions) mechanisms are used.

The EDC2005 model has been reported to overpredict peak temperatures in MILD combustion conditions. Adjustments of the \( C_i \) and \( C_\gamma \) constants have been proposed to correct this behavior. Among them, the adjustment proposed by De et al. focused on the DJHC flame, for which \( C_i \) was increased to 3.0 or \( C_\gamma \) decreased to 1.0.

The current investigation focuses on the simulation of MILD combustion with the tabulated dynamic adaptive chemistry (TDAC) approach, where several conventional reduction methods are adjusted to be dynamic and coupled with the dynamic tabulation method of ISAT. The study is motivated by the specific features of such a combustion regime: indeed, there is a general consensus about the need of detailed kinetics in MILD combustion, yet it is not clear to which extent a chemical mechanism reduction methods, including DRG, directed relation graph with error propagation (DRGEP), DAC, elementary fluid analysis (EFA), and path flux analysis (PFA). The TDAC process is visualized in Figure 1. ISAT stores the initial composition and

![Figure 1. TDAC method flowchart. This figure was adjusted with permission from ref 14. Copyright 2011 Elsevier.](image-url)
graph assumption, in which the relationship between various species or reactions are linked with weights. An error $\varepsilon$ is evaluated when a species or reaction is removed and compared to the maximum acceptable value $\varepsilon_{\text{max}}$ predefined by the user. If $\varepsilon$ is larger than the maximum error $\varepsilon_{\text{max}}$ this means that the species/reaction is important for the network graph and cannot be removed. The DRG, DRGEP, and PFA methods that were initially developed for mechanism preprocessing have been adapted for dynamic reduction in the TDAC method.\textsuperscript{14,38–32}

In DRG,\textsuperscript{34} the error $\varepsilon$ is expressed via an interaction coefficient $r_{AB}$ representing the contribution of species B to the production rate of species A

$$\varepsilon = r_{AB} = \frac{\sum_{i=1}^{n} \nu_{A,i} \mu_{i} \delta_{B,i}}{\sum_{i=1}^{n} \nu_{A,i} \mu_{i}}$$

where $\nu_{A,i}$ is the stoichiometric coefficient of species A, $\mu_{i}$ represents the reaction rate in each reaction, $n_{r}$ is the number of reactions, and $\delta_{B,i}$ is defined as

$$\delta_{B,i} = \begin{cases} 1, & \text{if the } i\text{th elementary reaction involves species } B \\ 0, & \text{otherwise} \end{cases}$$

Later, Pepiot-Desjardins and Pitsch\textsuperscript{35} proposed the DRGEP model. They argued that a more accurate way to consider the contribution of species B to species A is to use the net contribution, instead of evaluating a reaction rate in each reaction, production and consumption individually. Therefore, a new definition of the direct interaction coefficient is introduced

$$r_{AB} = \frac{\sum_{i=1}^{n} \nu_{A,i} \mu_{i} \delta_{B,i}}{\max(P_{A}, C_{A})}$$

in which $P_{A}$ and $C_{A}$ denote the production and consumption of species A, respectively. They are expressed as

$$P_{A} = \sum_{i=1}^{n} \max(0, \nu_{A,i} \mu_{i})$$

and

$$C_{A} = \sum_{i=1}^{n} \max(0, -\nu_{A,i} \mu_{i})$$

Furthermore, the effect of removing a group of species is also included, since the previously removed species are considered as well. Most importantly, the notion of error propagation is brought up in DRGEP. When evaluating the error $r_{AB}$, the length of the path the error has to propagate, from B to A, is considered.

$$r_{AB,p} = \prod_{i=1}^{n_{p}} r_{S_{i+1}}$$

Instead, only the weakest contribution is taken in DRG

$$r_{AB,p} = \min_{i=1}^{n_{p}} r_{S_{i+1}}$$

In eqs 9 and 10, $S_{j} = A, S_{k} = B$, and $p$ denotes a certain path that links two species A and B. Finally

$$\varepsilon = r_{AB} = \max_{\text{all paths}} r_{AB,p}$$

The DAC method implemented in the TDAC code is very similar to the DRGEP approach discussed above, except that the contribution of removing a group of species is neglected.\textsuperscript{29,36} With regard to the PFA model based on the DRG and DRGEP methods, both the directly linked (first generation) and indirectly linked (higher generation) species contributions are evaluated,\textsuperscript{14} resulting in an interaction coefficient $r_{AB}$ with information from multiple generations. For the EFA approach, the error evaluation is focused on removing individual reactions, expressed as

$$\varepsilon = \frac{\sum_{i=1}^{n_{r}} (r_{i} - \bar{r})^2}{s^2}$$

where $r_{i}$ and $\bar{r}$ are the individual reaction rates before and after removing a reaction $k$ from the network. The variance $s^2$ is defined as

$$s^2 = \frac{\sum_{i=1}^{n_{r}} (r_{i} - \bar{r})^2}{n_{r} - 1}$$

where $\bar{r}$ is the averaged reaction rate of the remaining reactions after reaction $k$ is removed.

Table 1 shows the differences and similarities of the five reduction models explained.

| Table 1. Differences and Similarities of the Reduction Models in TDAC |
|------------------|-------|-------|-----|-----|-----|
| reduction model  | DRG   | DRGEP | DAC | PFA | EFA |
| graph network    | yes   | yes   | yes | yes | yes |
| species group effect | no    | yes   | no  | yes | yes |
| error propagation | no    | yes   | no  | yes |     |
| error evaluation based on | species | species | species | species | reaction |

## VALIDATION CASES

The DJHC burner\textsuperscript{15,17} is chosen as a validation case to test the EDC2005 model and the TDAC potential for CPU time saving. 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 the inner diameter of 82.8 mm. A schematic 2D drawing of the DJHC burner is shown in Figure 2. A detailed description of the DJHC jet can be found in the studies of Oldenhof et al.\textsuperscript{15} and Sarras et al.\textsuperscript{17}

![Figure 2. 2D schematic drawing of the DJHC burner. This figure was adjusted with permission from ref 15. Copyright 2011 Elsevier.](image)

The mean and variance of the temperature and velocity experimental values are available for validation. Both Dutch natural gas\textsuperscript{15} and biogas\textsuperscript{17} are used as central jet fuel. The Dutch natural gas case is classified as “DJHC-I” with $Re = 4100$ and biogas as “biogas” with $Re = 4000$ in the research work of Sarras et al.\textsuperscript{17} The differences on the fuel component are listed in Table 2.

Further properties of the fuel and co-flow streams are presented in Table 3.

| Table 2. Central Jet Fuel Content for the Dutch Natural Gas and Biogas Cases |
|-----------------|-------|-------|-------|
| molar content (%) | CH$_4$ | N$_2$ | C$_2$H$_6$ | CO$_2$ |
| Dutch natural gas | 81   | 15   | 4   |
| biogas           | 56.7 | 10.5 | 2.8 | 30 |

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toward the radial direction (r). Kinetic mechanism: DRM19. TDAC with DAC reduction model. Fuel: Dutch natural gas. All spatial discretization. The DRM19, 19 GRI3.0, 20 and POLI-MIC1C3HT21 mechanisms are selected, because they are characterized by an increasing complexity, i.e., number of species and reactions. According to Contino,29 the value of 10−4 is commonly used for the ISAT tabulation tolerance if the test case is not highly inhomogeneous. Bourgeois tested several choices of reduction tolerance of DAC,39 showing that the cases with reduction tolerances of 1 × 10−4 and 1 × 10−5 show almost identical ignition delay time as the case with DAC turned off. However, using 1 × 10−4 is able to largely reduce the active species. Therefore, TDAC is used with a tolerances for both reduction and tabulation of 1 × 10−4.

The seulex solver is used for ODE integration, and the absolute tolerance of 1 × 10−11 and relevant tolerance of 1 × 10−8 are chosen. Seulex uses an extrapolation algorithm, and it is based on the linearly implicit Euler method with step size control and order selection.31 The target species of CO, CH₄, and HO₂ are chosen for reduction methods after a sensitivity analysis.

The standard EDC parameters are first used for the simulation of both natural gas and biogas cases. Looking at eq 2, it is clear that increasing Cᵣ leads to an increased residence time in the fine structures. This results in a higher CPU time required for the ODE integration. Preliminary simulations confirmed this, indicating that setting Cᵣ to 3.0 results in a simulation time 2.6 times higher than with standard constants. Therefore, we chose to adjust first the value of Cᵣ to 1.0, as suggested by De et al.,25 and then modify the constant Cₓ to improve the prediction. This approach was chosen since the focus of the present work is on dynamic chemistry reduction and tabulation. However, more advanced EDC extensions have been proposed for MILD combustion, such as the use of functional expressions accounting for local flow conditions or using a variable reacting fraction of the fine structures.40

### NUMERICAL SETTINGS

Unsteady Reynolds-averaged Navier–Stokes (URANS) simulations with local time stepping (LTS) were performed, using the standard k—ε turbulence model. Previous work conducted by Lewandowski and Ertesvåg40 has shown that the standard k—ε gives the best prediction on jet mean velocity decay among the other modified k—ε model variations for DJHC flames with a low Reynolds number. The simulation domain is extended 22S mm axially (z) downstream of the burner exit and 80 mm toward the radial direction (r). The whole domain is discretized with a structured 2D axisymmetric mesh. After a grid independence study,40 the mesh with 14 400 cells is chosen. The inlet boundary conditions for the temperature and velocity were taken from experimental values and tabulation of 1 species. Therefore, TDAC is used with a tolerances for both reduction methods on TDAC performances is also discussed.

In the present section, numerical simulations of the DJHC burner fed with natural gas and biogas are validated against experimental data. The reduction and tabulation efficiency, which is represented with CPU time saving, provided by TDAC is demonstrated. The influence of the chemical mechanism and reduction methods on TDAC performances is also discussed. Finally, reduction and tabulation in TDAC are tested separately, to identify their contributions to speed-up.

#### Natural Gas Flame Modeling.

For the natural gas case, the mean radial and axial temperature and velocity profiles are used for validation. The axial locations for the radial profiles are z = 15/30/60/120/150 mm. The mean temperature profile is first presented in Figure 3. The corresponding values of Cᵣ and Cₓ are listed in Table 4.

![Figure 3](Image)

**Table 3. Physical Properties of the Natural Gas and Biogas Flames**

| case               | V_fuel,mean (nL/min) | T_start (K) | V_0-fuel,O2 | R_fuel |
|--------------------|----------------------|-------------|-------------|--------|
| Dutch natural gas  | 16.1                 | 1540        | 0.076       | 4100   |
| biogas             | 15.3                 | 1436        | 0.095       | 4000   |

**Table 4. Standard and Adjusted EDC Constants**

| constant   | standard | adjust 1 | adjust 2 | adjust 3 |
|------------|----------|----------|----------|----------|
| Cₓ         | 2.1377   | 1.0      | 1.0      | 3.0      |
| Cᵧ         | 0.4083   | 0.4083   | 1.47     | 3.0      |

In Figure 3, the effect of the EDC model parameters become clear at axial locations above 30 mm. At z = 60 mm, a pronounced temperature peak, by over 300 K higher than the experimental value, is obtained using the standard and first set of modified constants.
constants (adjust1). With the second set of modified constants (adjust2), this overprediction is alleviated to 100 K. Further downstream, at \( z = 120 \) and 150 mm, a slightly shifted temperature peak is observed for all of the cases, with the case using the adjust2 set closer to the measured values. For the centerline profile, applying the second set of constants helps correct the overprediction of the temperature downstream (\( z \geq 100 \) mm) of the jet. Overall, the second set of constants better capture the experimental temperatures for the DJHC natural gas flame case. Using the third set of modified constants (adjust3) leads to an extinguished flame. Thus, the corresponding profiles are not shown here. The reduced peak temperature value from standard constants to first set of constants is due to the decrease of \( C_m \) which leads to the reduction of the mass fraction of the reacting zone in each computational cell, as shown in eq 1. Going from the first to the third set of constants, \( C_m \) is kept constant and \( C_r \) is increased. As a result, a higher residence time \( \tau \) and lower mean reaction rates are observed, as indicated by eq 3. Therefore, the temperature peak is further alleviated with the second set of constants, and the flame is extinguished with the third.

The velocity profiles are presented in Figure 4. Using different parameter sets results in negligible differences at almost all locations. However, the case with standard constants shows earlier decay, as indicated by the centerline profile. Generally, the prediction of the radial velocity profiles is quite accurate, at all axial locations. The centerline profile shows, however, an underprediction of velocity after \( z = 30 \) mm, by 13.5% at \( z = 60 \) mm. This means that the jet downstream decay rate is slightly overpredicted by the model.

As far as the computational savings are concerned, the relative, normalized CPU time of the simulations, with and without TDAC, is shown in Table 5. The normalization is based on the CPU time consumption of the base case, defined as the simulation with standard EDC and without TDAC. A speed-up of 1.4−2 times is obtained using the DRM19 mechanism, depending upon the set of adjusted constants used for the natural gas flame. The mean temperature and velocity profile obtained using TDAC are virtually identical to the results obtained without TDAC; therefore, they are not shown here.

### Table 5. Normalized Time Consumption with/without TDAC and Using Various EDC Sets of Constants

| constant set          | standard | adjust1 | adjust2 | adjust3 |
|-----------------------|----------|---------|---------|---------|
| without TDAC          | 1.00     | 0.72    | 1.94    |         |
| with TDAC             | 0.51     | 0.50    | 1.26    |         |
| speed-up              | 1.96     | 1.44    | 1.54    |         |

**TDAC with DAC reduction model applied. Fuel: Dutch natural gas.**

### Biogas Flame Modeling

For the biogas case, the radial profiles at axial locations \( z = 65/110/140/170 \) mm are used for temperature validation, while the experimental data at \( z = 30/60/90/120 \) mm are available to compare to computed velocities. In Figure 5, the mean numerical and experimental temperature profiles are compared, using several EDC2005 parameter settings. The trend is very similar to what is observed for the natural gas case. A clear overprediction of temperature levels is obtained using the standard and adjust 1 sets of constants. The level of agreement is significantly improved using the second and third sets of constants, although some overpredictions (10−20%) can still be observed at \( z = 140/170 \) mm using the third set. Figure 6 shows the comparison between the measured and computed velocity profiles. The agreement is very satisfactory, with the maximum difference always being below 1.0 m/s, and no distinctive difference can be observed between the profiles provided by the different EDC constant sets.

The normalized time consumption factor with and without TDAC is shown in Table 6. Together with the data from the natural gas case in Table 5, a conclusion can be drawn that the total CPU time is indeed increased with the increased \( C_r \) value. Furthermore, the speed-up factor still lies in the range between 1.4 and 2.0. Almost no visual difference can be observed between the results obtained with and without TDAC.

### Influence of Chemical Mechanisms

The natural gas flame is chosen for the current and following sections, to demonstrate the influence of chemical kinetics and reduction algorithm on the simulation speed-up as well as to clarify the role of tabulation and reduction in TDAC. In the study of De et al., the DRM19 mechanism was found to perform satisfactorily for the DJHC.
However, to test the potential speed-up of the TDAC method, two additional mechanisms of increasing size are chosen, namely, GRI3.0 and POLIMIC1C3HT. At the same time, the effect of the mechanism on the accuracy of the predictions is assessed as well. Figure 7 compares the temperature profiles provided by the different mechanisms plotted against the experimental data. The second set of constants (adjust2) is employed for all simulations. The profiles provided by the different mechanisms do not show major differences. Nevertheless, using a more detailed mechanism at downstream axial distances, z = 120 mm, helps reduce the temperature overprediction by 30−40 K.

In Table 7, a trend of increasing speed-up is observed when more complicated mechanisms are chosen. The CPU time consumption normalization is based on the case with the DRM19 mechanism and without TDAC. Using the POLIMIC1C3HT mechanism requires 27.43 times the CPU required for the DRM19 case when neither reduction nor tabulation is applied. Using the TDAC method reduces the gap to 4.12 times.

Table 6. Normalized Time Consumption with/without TDAC and Using Various EDC Sets of Constants

| constant set   | standard | adjust1 | adjust2 | adjust3 |
|----------------|----------|---------|---------|---------|
| without TDAC   | 1.0      | 1.34    | 1.58    | 1.88    |
| with TDAC      | 0.55     | 0.78    | 1.03    | 1.14    |
| speed-up       | 1.82     | 1.72    | 1.53    | 1.65    |

aTDAC with DAC reduction model applied. Fuel: biogas.
which corresponds to roughly 10 times speed-up. The use of a mechanism as complicated as POLIMIC1C3HT might not be necessary for the present case. However, the significant CPU time saving achieved using TDAC is very promising for the application of the method to more complex cases and regimes, with no a priori knowledge about the appropriateness of a pre-reduced mechanism (as DRM19).

Finally, the number of active species distribution for the various chemical mechanisms in the domain is presented in Figure 8. The number of active species indicates the species left after removing unimportant species. We can observe that almost 100% of the species are used in both the reaction zone and the downstream ($z \geq 100$ mm) area of the simulation domain when

![Figure 7. Mean temperature profiles obtained with the EDC2005 model using the DRM19, GRI3.0, and POLIMIC1C3HT mechanisms compared to the experimental data. EDC2005 constant: adjust2. TDAC with DAC reduction model. Fuel: Dutch natural gas.]

**Table 7. Normalized Time Consumption with/without TDAC and Various Chemical Mechanisms**

| mechanism   | DRM19 | GRI3.0 | POLIMIC1C3HT |
|-------------|-------|--------|--------------|
| without TDAC| 1.0   | 4.01   | 27.43        |
| with TDAC   | 0.65  | 2.10   | 2.68         |
| speed-up    | 1.54  | 1.91   | 10.24        |

“TDAC with DAC reduction model. Fuel: Dutch natural gas.

![Figure 8. Contour plot of the number of active species (nActiveSpecies) obtained using the EDC2005 model with the DRM19, GRI3.0, and POLIMIC1C3HT mechanisms. EDC2005 constant: adjust2. TDAC with DAC reduction model applied. Fuel: Dutch natural gas. Axis unit: m.]

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the DRM19 mechanism is chosen. Away from these regions, the number of active species drops, thus saving computational resources. This effect is magnified with two other chemical mechanisms. With GRI3.0, around 75% of the species are necessary in the active zone close to the centerline. Upstream and far from the centerline region, the number of active species is further reduced to around 20–30. With the POLIMIC1C3HT mechanism, approximately 56% of the species set is retained, leading to the observed speed-up factor of 10.24, as indicated in Table 7. The reason for the low reduction potential on the DRM19 mechanism is that it is already a small (skeletal) mechanism with less than 20 species (inert species excluded). In the MILD non-conventional combustion regime, most of the species and reactions are necessary to include to account for the complex turbulence/chemistry interactions. However, for a large mechanism, such as POLIMIC2C3HT, there are much more species and reactions. Some of them are important only for a certain area of the domain; thus, more reduction on CPU time is obtained.

**Influence of the Chemistry Reduction Approach.** In the present section, the speed-up factor of various chemistry reduction models (DRG, DRGEP, DAC, PFA, and EFA) are compared using both the skeletal and comprehensive mechanisms. The mean temperature and species mass fraction profiles obtained from the five different reduction models with the DRM19 mechanism show only minor differences, as indicated in the Supporting Information. In Table 8, the speed-up factors are listed and compared. Minor differences are observed for the DRM19 case, using the different chemistry reduction approaches. This can be explained with the help of Figure 8. Indeed, the DRM19 mechanism is already a reduced mechanism, and this implies that the margins for further reduction are tight. On the other hand, using the POLIMIC1C3HT mechanism, larger differences are identified between the five methods. The DRGEP, DAC, and EFA models have speed-up factors around 3 times higher than those obtained using DRG and PFA models. Using a more complicated mechanism helps to better evaluate the efficiency of the various approaches.

In Figure 9, a contour plot of the number of active species in the simulation domain is again presented when the POLIMIC1C3HT mechanism is applied. The reduction ability of DRG, DRGEP, DAC, PFA, and EFA models can thus be analyzed. The DRG and PFA reduction models provide very weak reduction ability. Approximately only 10% of the whole species set are removed for the reaction area and regions nearby, whereas the other three approaches, DRGEP, DAC, and EFA, present a significantly reduced set of species, around 40% by estimation. Among them, the DRGEP model provides the most reduced set of species in the whole domain. Moreover, the reactive area is well-identified with DRGEP, DAC, and EFA models as well.

CH$_2$O is a key precursor in the initiation process of the reaction for fuel molecules with carbon atoms, especially in MILD combustion. Therefore, it is adopted here in combination with the temperature to identify the reactive region in the flame. The regions with a high temperature and high CH$_2$O mass fraction are highlighted in Figure 10. When DRGEP, DAC, or EFA methods are chosen, a larger number of active species is identified in these regions (Figure 9), which is not the case when the DRG or PFA methods are used. The latter identifies a large number of active species far from the centerline, after $z = 100$ mm, where low temperature and CH$_2$O levels are observed. This proves that the DRG and PFA models are not as efficient and accurate as the other reduction approaches in selecting the appropriate active species in the relevant regions of the flame.

**Influence of Reduction/Tabulation.** Because the TDAC method is a combination of chemistry tabulation and reduction, it is interesting to quantify how much they contribute to the speed-up separately. In Table 9, the normalized CPU time is listed considering both tabulation and reduction, only reduction, only tabulation, and without reduction/tabulation. Both DRM19 and POLIMIC1C3HT mechanisms were used. The base case for normalization is the case with both tabulation and reduction for a given mechanism used. A vertical comparison of normalized CPU time is not possible. From Table 9, it can be observed that, using a small mechanism, such as DRM19, the tabulation contribution is more significant, whereas with a large mechanism, such as POLIMIC1C3HT, the reduction step plays the major role. For a detailed mechanism, such as POLIMIC1C3HT, the reduction of the number of species significantly speed-up the ODE integration step. When it comes to the DRM19 case, the mechanism is already very light (the number of species smaller than 20, inert species excluded) and the reduction potential is much lower. Therefore, tabulation is the key step under such conditions.

A contour plot of the averaged tabulation results (TabulationResultsMean) is shown in Figure 11. Because it is averaged data, the results are presented in decimals between 0 and 2, instead of integers of 0 (add), 1 (grow), and 2 (retrieve). The retrieve (solution directly recovered with linear approximation), grow (ellipsoid of accuracy expanded to include the new mapping), and add (new “leaf” added) in each computational cell can be identified with red, green, and blue colors. Most regions that are far away from the centerline are marked with retrieve for both mechanisms. In the reactive region, as identified from Figure 10, it is characterized mostly by green (grow), indicating an expansion of the EOA. One can also appreciate that the POLIMIC1C3HT mechanism is able to capture the interaction of co-flow and air streams in the upstream area, which is colored with green, whereas the DRM19 mechanism predicted the same area to be closer to retrieve. Using two mechanisms, the reaction system is described with different reactions and species. Therefore, the contributions of reactions and dominating phenomenon in local regions are different as well, showing inconsistency on the tabulation results.

| Table 8. Simulation Speed-up with Various Reduction Methods
|-------------------|--------|--------|--------|--------|
| reduction model   | DRG    | DRGEP  | DAC    | PFA    |
| speed-up with DRM19| 1.57   | 1.55   | 1.55   | 1.58   | 1.55   |
| speed-up with POLIMIC1C3HT | 3.9    | 10.90  | 10.22  | 3.2    | 9.6    |

"Tabulation turned on. Fuel: Dutch natural gas."
PFA, and EFA). Moreover, the contributions of tabulation and reduction are investigated separately. The following conclusions are summarized: (1) The current EDC model implementation in OpenFOAM with a modified set of constants ($C_\gamma = 1.0$ and $C_\tau = 1.47$) provide satisfactory predictions for mean temperature profiles of the natural gas flame, while the lower reactivity of the biogas flame make the use of $C_\gamma = 1.0$ and $C_\tau = 3.0$ more accurate. The mean velocity distribution is well-predicted for both flames with all of the model settings. A speed-up factor of 1.4−2.0 is obtained using the TDAC method with the DRM19 mechanism. (2) Using a more complicated chemical mechanism, such as GRI3.0 or POLIMIC1C3HT, slightly improves temperature predictions and leads to significantly increased CPU time with respect to the DRM19 case (27.4 times for POLIMIC1C3HT) when TDAC is turned off. However, with TDAC, a 10 times speed-up is obtained for the POLIMIC1C3HT mechanism, reducing the CPU time gap between POLIMIC1C3HT and DRM19 to 4.16 times. (3) The DRGEP, DAC, and EFA reduction approaches are more efficient and accurate compared to the DRG and PFA models. (4) When a small mechanism is used (DRM19), tabulation is the main contributor to computational saving. With more complex mechanisms (POLIMIC1C3HT), chemistry reduction plays the main role.

The current investigation validates the application of the EDC model in combination with the TDAC method under MILD regime using the open-source software OpenFOAM. The time saving using TDAC with complex chemistry is very promising.

Table 9. Normalized Time Consumption with/without Reduction and Tabulation

| model     | no TDAC | TDAC | only reduction | only tabulation |
|-----------|---------|------|----------------|-----------------|
| DRM19     | 1.55    | 1.0  | 1.41           | 1.03            |
| POLIMIC1C3HT | 10.22   | 1.0  | 2.38           | 6.33            |

*DAC reduction model. Fuel: Dutch natural gas. Comparison between the normalized times is possible only horizontally.

Figure 9. Contour plot of the number of active species (nActiveSpecies) obtained using the EDC2005 model with the DRM19, GRI3.0, and POLIMIC1C3HT mechanisms. EDC2005 constant: adjust2. TDAC with DAC reduction model applied. Fuel: Dutch natural gas. Axis unit: m.

Figure 10. Contour plot of the mean CH$_2$O mass fraction and mean temperature obtained using the EDC2005 model. Kinetic mechanism: POLIMIC1C3HT. EDC2005 constant: adjust2. Tabulation turned on. DAC reduction models. Fuel: Dutch natural gas. Axis unit: m.
thus making it feasible to extend it to industrial or semi-industrial applications.

■ ASSOCIATED CONTENT

* Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.energyfuels.8b01001.

Mean temperature and species mass fraction profiles from the five cases with different reduction methods (DRG, DRGEP, DAC, EFA, and PFA) compared to the case in which the reduction is turned off (PDF).

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■ REFERENCES

(1) Winning, J. A.; Winning, J. G. Flameless oxidation to reduce thermal NO formation. Prog. Energy Combust. Sci. 1997, 23, 81−94.
(2) Cavaliere, A.; de Joannon, M. MILD combustion. Prog. Energy Combust. Sci. 2004, 30, 329−366.
(3) de Joannon, M.; Sorrentino, G.; Cavaliere, A. MILD combustion in diffusion-controlled regimes of hot diluted fuel. Combust. Flame 2012, 159, 1832−1839.
(4) Ihme, M.; Zhang, J.; He, G.; Dally, B. Large-eddy simulation of a jet-in-hot-coflow burner operating in the oxygen-diluted combustion regime. Flow, Turbul. Combust. 2012, 89, 449−464.
(5) Magnusson, B. F. Eddy dissipation concept: A bridge between science and technology. Proceedings of the ECCOMAS Thematic Conference on ComputationalCombustion; Lisbon, Portugal, June 21−24, 2005.
(6) Tosatto, L.; Bennett, B. A. V.; Smooke, M. D. A transport-flux-based directed relation graph method for the spatially inhomogeneous instantaneous reduction of chemical kinetic mechanisms. Combust. Flame 2011, 158, 820−835.
(7) Yang, H.; Ren, Z.; Lu, T.; Goldin, G. M. Dynamic adaptive chemistry for turbulent flame simulations. Combust. Theory Modell. 2013, 17, 167−183.
(8) Zhang, S.; Broadbelt, L. J.; Androulakis, I. P.; Ierapetritou, M. G. Comparison of biodiesel performance based on HCCI engine simulation using detailed mechanism with on-the-fly reduction. Energy Fuels 2012, 26, 976−983.
(9) He, K.; Androulakis, I. P.; Ierapetritou, M. G. Numerical investigation of homogeneous charge compression ignition (HCCI) combustion with detailed chemical kinetics using on-the-fly reduction. Energy Fuels 2011, 25, 3369−3376.
(10) Oluwole, O. O.; Ren, Z.; Petre, C.; Goldin, G. Decoupled Species and Reaction Reduction: An error-controlled method for Dynamic Adaptive Chemistry simulations. Combust. Flame 2015, 162, 1934−1943.
(11) Xie, W.; Lu, Z.; Ren, Z.; Hou, L. Dynamic adaptive chemistry via species time-scale and Jacobian-aided rate analysis. Proc. Combust. Inst. 2017, 36, 645−653.
(12) Tang, Q.; Pope, S. B. Implementation of combustion chemistry by in situ adaptive tabulation of rate-controlled constrained equilibrium manifolds. Proc. Combust. Inst. 2002, 29, 1411−1417.
(13) Ren, Z.; Liu, Y.; Lu, T.; Lu, L.; Oluwole, O. O.; Goldin, G. M. The use of dynamic adaptive chemistry and tabulation in reactive flow simulations. Combust. Flame 2014, 161, 127−137.
(14) Contino, F.; Jeannart, H.; Lucchini, T.; D’Errico, G. Coupling of in situ adaptive tabulation and dynamic adaptive chemistry: An effective method for solving combustion in engine simulations. Proc. Combust. Inst. 2011, 33, 3057−3064.
(15) Oldenhof, E.; Tummers, M. J.; van Veen, E. H.; Roekaerts, D. J. E. M. Role of entrainment in the stabilisation of jet-in-hot-coflow flames. Combust. Flame 2011, 158, 1553−1563.
(16) Oldenhof, E.; Tummers, M. J.; van Veen, E. H.; Roekaerts, D. J. Transient response of the Delft jet-in-hot-coflow flames. Combust. Flame 2012, 159, 697−706.
(17) Sarras, G.; Mahmoudi, Y.; Arteaga Mendez, L. D.; van Veen, E. H.; Tummers, M. J.; Roekaerts, D. J. E. M. Modeling of turbulent natural gas
and biogas flames of the Delft Jet-in-Hot-CoFlow burner: Effects of coflow temperature, fuel temperature and fuel composition on the flame lift-off height. *Flow, Turbul. Combust.*, 2014, 93, 607–635.

(18) Kulkarni, R. M.; Poliube, W. LES of Delft-Jet-In-Hot-CoFlow (DJHC) with tabulated chemistry and stochastic fields combustion model. *Fuel Process. Technol.* 2013, 107, 138–146.

(19) Kazakov, A.; Frenklach, M. GRI-Mech 1.2; http://combustion.berkeley.edu/drm/.

(20) Smith, G. P.; Golden, D. M.; Frenklach, M.; Moriarty, N. W.; Eitteneer, B.; Goldenberg, B.; Bowman, C. T.; Hanson, R. K.; Song, S.; Gardiner, W. C.; Jr.; Lissianski, V. V.; Qin, Z. GRI-Mech 3.0; http://www.me.berkeley.edu/gri_mech/.

(21) Frassoldati, A.; Faravelli, T.; Ranzi, E. Kinetic modeling of the interactions between NO and hydrocarbons at high temperature. *Combust. Flame* 2003, 135, 97–112.

(22) Evans, M. J.; Medwell, P. R.; Tian, Z. F. Modelling lifted jet flames in a heated coflow using an optimized eddy dissipation concept model. *Combust. Sci. Technol.* 2015, 187, 1093–1109.

(23) Shahbanian, S. R.; Medwell, P. R.; Rahimi, M.; Frassoldati, A.; Cuoci, A. Kinetic and fluid dynamic modeling of ethylene jet flames in diluted and heated oxidant stream combustion conditions. *Appl. Therm. Eng.* 2013, 52, 538–554.

(24) Parente, A.; Malik, M. R.; Contino, F.; Cuoci, A.; Dally, B. B. Extension of the Eddy Dissipation Concept for turbulence/chemistry interactions to MILD combustion. *Fuel* 2016, 163, 98–111.

(25) De, A.; Oldenhof, E.; Sathiah, P.; Roekaerts, D. Numerical simulation of Delft-Jet-in-Hot-CoFlow (DJHC) flames using the eddy dissipation concept model for turbulence-chemistry interaction. *Flow, Turbul. Combust.* 2011, 87, 537–567.

(26) Aminian, J.; Galletti, C.; Shahhosseini, S.; Tognotti, L. Numerical investigation of a MILD combustion burner: Analysis of mixing field, chemical kinetics and turbulence-chemistry interaction. *Flow, Turbul. Combust.* 2012, 88, 597–623.

(27) Li, Z.; Cuoci, A.; Sadiki, A.; Parente, A. Comprehensive numerical study of the Adelaide Jet in Hot-CoFlow burner by means of RANS and detailed chemistry. *Energy* 2017, 139, 555–570.

(28) Pope, S. B. Computational efficient implementation of combustion chemistry using in situ adaptive tabulation. *Combust. Theory Model.* 1997, 1, 41–63.

(29) Contino, F. Combustion in homogeneous charge compression ignition engines: Experimental analysis using ethyl esters and development of a method to include detailed chemistry in numerical simulations. Ph.D. Thesis, Université Catholique de Louvain, Louvain-la-Neuve, Belgium, 2011.

(30) Contino, F.; Lucchini, T.; D’Errico, G.; Duynslaeger, C.; Dias, V.; Jeannart, H. Simulations of advanced combustion modes using detailed chemistry combined with tabulation and mechanism reduction techniques. *SAE International Journal of Engines* 2012, 5, 185–196.

(31) Contino, F.; Foucher, F.; Dagaut, P.; Lucchini, T.; D’Errico, G.; Mounain-Rousselle, C. Experimental and numerical analysis of nitric oxide effect on the ignition of iso-octane in a single cylinder HCCI engine. *Combust. Flame* 2013, 160, 1476–1483.

(32) Contino, F.; Masurier, J.-B.; Foucher, F.; Lucchini, T.; D’Errico, G.; Dagaut, P. CFD simulations using the TDAC method to model iso-octane combustion for a large range of oxygen seeding and temperature conditions in a single cylinder HCCI engine. *Fuel* 2014, 137, 179–184.

(33) Bourgeois, N.; Goldsborough, S. S.; Vanhoeve, G.; Duponcheel, M.; Jeannart, H.; Contino, F. CFD simulations of rapid compression machines using detailed chemistry: Impact of multi-dimensional effects on the auto-ignition of the iso-octane. *Proc. Combust. Inst.* 2017, 36, 383–391.

(34) Lu, T.; Law, C. K. A directed relation graph method for mechanism reduction. *Proc. Combust. Inst.* 2005, 30, 1333–1341.

(35) Pepiot-Desjardins, P.; Pitsch, H. An efficient error-propagation-based reduction method for large chemical kinetic mechanisms. *Combust. Flame* 2008, 154, 67–81.

(36) Liang, L.; Stevens, J. G.; Farrell, J. T. A dynamic adaptive chemistry scheme for reactive flow computations. *Proc. Combust. Inst.* 2009, 32, 527–534.

(37) Karst, F.; Maestri, M.; Freund, H.; Sundmacher, K. Reduction of microkinetic reaction models for reactor optimization exemplified for hydrogen production from methane. *Chem. Eng. J.* 2015, 281, 981–994.

(38) Sun, W.; Chen, Z.; Gou, X.; Ju, Y. A path flux analysis method for the reduction of detailed chemical kinetic mechanisms. *Combust. Flame* 2010, 157, 1298–1307.

(39) Bourgeois, N. Numerical study of multi-dimensional effects in rapid compression machines. Ph.D. Thesis, Université Catholique de Louvain and Vrije Universiteit Brussel, Brussels, Belgium, 2017.

(40) Lewandowski, M. T.; Ertesvåg, I. S. Analysis of the Eddy Dissipation Concept formulation for MILD combustion modeling. *Fuel* 2018, 224, 687–700.

(41) Hairer, E.; Wanner, G. *Solving Ordinary Differential Equations II: Stiff and Differential-Algebraic Problems*, 2nd ed.; Springer: Berlin, Germany, 1996; Springer Series in Computational Mathematics, Vol. 14, DOI: 10.1007/978-3-642-05221-7.

(42) Gordon, R. L.; Masri, A. R.; Pope, S. B.; Goldin, G. M. Transport budgets in turbulent lifted flames of methane autoigniting in a vitiated co-flow. *Combust. Flame* 2007, 151, 495–511.

(43) Medwell, P. R. Laser diagnostics in MILD combustion. Ph.D. Thesis, The University of Adelaide, Adelaide, Australia, 2007.

(44) Duwig, C.; Li, B.; Li, Z. S.; Aldén, M. High resolution imaging of flameless and distributed turbulent combustion. *Combust. Flame* 2012, 159, 306–316.

(45) Minamoto, Y. Physical aspects and modelling of turbulent MILD combustion. Ph.D. Thesis, University of Cambridge, Cambridge, U.K., 2013.