Direct numerical simulation of autoigniting mixing layers in MILD combustion

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Abstract

MILD combustion is a new combustion technology which promises an enhanced efficiency and reduced emission of pollutants. It is characterized by a high degree of preheating and dilution of the reactants. Since the temperature of the reactants is higher than that of autoignition, a complex interplay between turbulent mixing, molecular transport and chemical kinetics occurs. In order to reveal the fundamental reaction structures of MILD combustion, the process of a cold methane–hydrogen fuel jet issuing in a hot diluted coflow and the subsequent ignition process is modeled by direct numerical simulation of autoigniting mixing layers using detailed chemistry and transport models. Detailed analysis of one-dimensional laminar mixing layers shows that the ignition process is dominated by hydrogen chemistry and that non-unity Lewis number effects are of the utmost importance for modeling of autoignition. High scalar dissipation rates in mixing layers delay the autoignition time, but have a negligible effect on the chemical pathway followed during ignition. This supports the idea of using homogeneous reactor simulations for the construction of chemistry look-up tables. Simulations of two-dimensional turbulent mixing layers confirm the effect of scalar dissipation rate on autoignition time. The turbulence–chemistry interaction is limited under the investigated conditions, because the reaction layer lies at the edge of the mixing layer due to the very small value of the stoichiometric mixture fraction. When the oxidizer stream is more diluted, the autoignition time is delayed, allowing the developing turbulence to interact more with the ignition chemistry. The results of these direct numerical simulations employing a detailed reaction mechanism are expected to be used for the development of tabulated chemistry models and sub-grid scale models for large-eddy simulations of MILD combustion.

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1. Introduction

The focus of this research is on new combustion concepts, which produce energy from current and future fuels, with substantially enhanced efficiency and significantly reduced pollutant emissions. Going by names such as high efficiency combustion and flameless oxidation, these new methods allow the use of recuperated heat in high-temperature processes without the penalty of increased NOx emissions, and offer the possibility of substantially homogenizing the temperature field in combustion devices (see, e.g., [1,2]). In these
combustion systems a high degree of preheating of the reactants is coupled with a high degree of dilution. Following [2], we use the term MILD combustion to refer to these systems. Although MILD combustion systems have been successfully introduced in some industries, broad implementation is hampered by a lack of fundamental insight into this combustion regime.

In MILD combustion systems, combustion takes place in the midst of turbulent mixing of the reactants. Since the temperature of the reactants is above that of autoignition, a complex interplay between turbulent mixing, molecular transport and chemical kinetics occurs. In order to reveal the fundamental reaction structures of MILD combustion, high-fidelity numerical simulations are needed in which all scales of turbulent motion are resolved. Combined with detailed chemical reaction models, direct numerical simulation (DNS) is the appropriate tool to investigate these phenomena. The drawback of DNS, conversely, is that it is extremely expensive from a computational point of view.

In order to investigate MILD combustion systems experimentally, a number of interesting studies have been performed using turbulent jets in hot diluted coflow (JHC for short) as a model system (e.g., [3–5]). In addition to these experimental studies a number of DNS studies have been performed to investigate the stabilization mechanism of jet flames in hot coflows (e.g., [6,7]). Autoignition was found to be the key stabilizing mechanism. Ignition of turbulent non-premixed flames has been reviewed by Mastorakos [8]. Often, DNS studies employ simple one-step or hydrogen reaction mechanisms to reduce the computational cost. Another way to keep down the computational cost, is to employ large eddy simulation (LES) (see e.g., [9,10]). In LES, however, the interaction between turbulence, chemistry, and molecular diffusion is not resolved but needs to be modeled. DNS provides data and insight that can help in the development of such models.

In the present study, the process of a cold methane–hydrogen fuel jet issuing in a hot diluted coflow and the subsequent ignition process is modeled by DNS of autoigniting mixing layers using detailed chemistry and transport models. The conditions are taken to match the experiments of Dally et al. [3] as close as possible. First, the role of molecular diffusion and chemistry is examined in detail by analyzing the results of one-dimensional (1D) laminar mixing layers. The effect of differential diffusion is explained and the most important chemical reactions influencing the autoignition time are identified. Finally, the interaction between the ignition chemistry and the developing mixing layer is investigated by analyzing 2D DNS results.

2. Direct numerical simulation

Igniting temporally evolving mixing layers have been modeled by using DNS. The employed DNS code was developed initially for single-step chemistry [11]. Later, it was extended to deal with tabulated and detailed chemistry [12]. The code solves the governing equations in fully compressible form in terms of density $\rho$, velocity $u$, species mass fractions $Y_s$, and temperature $T$. The viscosity and conductivity of the mixture are fitted as functions of temperature. The diffusion of species is modeled by using a constant Lewis number approach. The Lewis numbers were determined by fitting the expressions to results of 1D simulations with more detailed transport models. The chemical source terms are computed by using the DRM19 reaction mechanism, which is a reduced reaction set derived from the GRI mechanism and contains 21 species and 84 reversible reactions [13].

The governing equations are discretized by using the sixth-order accurate compact finite difference method [14] for the diffusion terms and the fifth-order method [15] for the convective terms. Time integration is performed by an explicit compact-storage Runge–Kutta scheme of third order. The time step is chosen to be $10^{-8}$ s in order to satisfy the stability criteria. The governing equations are solved on a two-dimensional square domain with a length of 20 mm and 521 grid points uniformly distributed in each direction. This results in a mesh size of 38 $\mu$m. For one case, the results obtained with this mesh have been compared with results obtained with double the number of grid points. The largest deviations were found for the CH radical and these were smaller than 3% of the maximum value. For most other variables the deviations were smaller than 1%. The boundary conditions are modeled with the Navier–Stokes Characteristic Boundary Conditions (NSCBC) [16]. In the streamwise ($x$) direction, periodic boundary conditions are applied, while in the cross-stream ($y$) direction, non-reflecting outlet boundaries are used assuming atmospheric pressure at infinity. A more detailed description of the code can be found in Refs. [11,12].

The DNS consists of a temporally evolving, non-premixed, planar jet flame. A layer of fuel in the domain center is surrounded by countercflowing oxidizer. The composition of the fuel and the oxidizer is chosen to match the experiments in Ref. [3]. The fuel is a mixture of methane and hydrogen, equal in volume. The oxidizer consists of air diluted with combustion products and nitrogen. The temperature of the oxidizer is 1300 K. Three cases with different dilution levels are considered. Their fuel and oxidizer compositions are listed in Table 1.
The fuel core width, \( W \), is 2 mm, and the difference in the velocity of the streams, \( \Delta U \), is 67 m/s. Together with the viscosity \( \nu \) of the fuel stream, this leads to a Reynolds number of 3850, which is in the range of the experiments by Dally et al. [3]. The fuel core velocity is perturbed with homogeneous isotropic turbulence of low intensity \( u'/\Delta U = 1\% \) to trigger the instabilities of the shear layers between the fuel and oxidizer streams. The turbulent fluctuations develop in time. At the time of ignition in case HM1, the Reynolds number based on the transversal Taylor length scale \( \lambda \), \( Re_t = u'\lambda/\nu = 240 \), with \( u' \) the r.m.s. velocity fluctuation in the fuel jet region (\( |y| < 1 \text{ mm} \)). The Taylor length scale is determined from the autocorrelation function of the cross-stream velocity. The Kolmogorov length scale is estimated to be \( \eta = (v^3/\epsilon)^{1/4} = 30 \mu \text{m} \) with \( \epsilon \) the mean dissipation rate in the fuel jet region. Note that the grid resolution is fine enough to resolve these scales.

The chemical composition of the flow is initialized by top-hat profiles for the species mass fractions and enthalpy, which are smoothed in the cross-stream direction by applying a [121] filter 10 times. Temperature and density are computed from these smoothed profiles.

### 3. Results

#### 3.1. One-dimensional laminar mixing layers

Before the DNS results of the 2D mixing layers are presented, the results of laminar mixing layers with \( \Delta U = 0 \) are analyzed in order to reveal the effects of chemical kinetics and molecular diffusion in the absence of turbulence. In these simulations, the initial velocity is zero and the problem becomes one-dimensional because the solution is independent of \( x \). In Fig. 1, the evolution of the temperature profiles in these laminar mixing layers is shown for the three different cases. Because of symmetry, only the positive \( y \) part is shown. After a short period in which the large initial temperature gradients are reduced by conduction, the mixture ignites at the oxidizer side and the temperature rises. With increasing dilution level, the maximum flame temperature is reduced and ignition is delayed.

The ignition timing is more clearly visible in Fig. 2. In this figure the temperature rise \( \Delta T \) is shown as function of time \( t \). \( \Delta T \) is defined as the maximum temperature rise in mixture fraction space with respect to the initial temperature profile:

\[
\Delta T(t) = \max_Z \{T(Z,t) - T(Z,0)\}. \tag{1}
\]

In (1) the mixture fraction \( Z \) is defined as a linear combination of the element mass fractions \( Z_j \) following Ref. [17]:

\[
Z = \frac{Z^* - Z_{\text{mix}}}{Z^*_{\text{fu}} - Z_{\text{ox}}^*} \quad \text{with} \quad Z^* = \frac{Z_C}{M_C} + \frac{1}{2} \frac{Z_H}{M_H} - \frac{Z_O}{M_O},
\]

in which \( M_j \) is the molar mass of element \( j \) and the subscripts \( \text{fu} \) and \( \text{ox} \) refer to the fuel and oxidizer.

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**Table 1.**

<table>
<thead>
<tr>
<th>Case</th>
<th>Fuel</th>
<th>Oxidizer</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( T (K) )</td>
<td>( Y_{\text{CH}} )</td>
</tr>
<tr>
<td>HM1</td>
<td>305</td>
<td>0.889</td>
</tr>
<tr>
<td>HM2</td>
<td>305</td>
<td>0.889</td>
</tr>
<tr>
<td>HM3</td>
<td>305</td>
<td>0.889</td>
</tr>
</tbody>
</table>

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Fig. 1. Temperature profiles in laminar mixing layers at \( t = 0, 0.2, \ldots, 1 \text{ ms} \) for the cases HM1 (left), HM2 (middle), and HM3 (right).
Figure 2 shows that the most diluted case (HM1) ignites at 0.32 ms, when the time of ignition is defined at the maximum gradient of $\Delta T$, whereas cases HM2 and HM3 ignite at 0.20 and 0.15 ms, respectively.

Another interesting observation from Fig. 2 is the small temperature increase that occurs during the first 0.1 ms of the simulation. This increase is not related to chemistry, but rather to differential diffusion effects. This conclusion can be drawn from the results presented in Fig. 3, in which the temperature rise is shown for case HM1 computed with different diffusion models. The constant Lewis number approach is compared with more detailed models such as the mixture-averaged approach, the multi-component model, and the simplified unity Lewis number approach in which there are no differential diffusion effects. While the ignition time is similar for the first three methods, it is more than 10 times longer for the unity Lewis number approach. Also, the initial temperature rise prior to ignition does not occur in the unity Lewis number case. The reason for this behavior is explained in Fig. 4. There the temperature and mass fraction of hydrogen ($\text{H}_2$) and oxygen ($\text{O}_2$) are plotted as a function of mixture fraction at $t = 0$ and at $t = 0.1$ ms for the constant Lewis number and unity Lewis number cases. At $t = 0$, the species profiles follow from the initial condition, resulting in a linear relation between $Y_z$ and $Z$. For the unity Lewis number case, this linear relation remains as long as chemistry can be neglected. However, for the constant Lewis number case, differential diffusion results in an increased temperature, hydrogen and oxygen mass fraction near the stoichiometric region, while the mass fraction of methane is reduced (not shown). These changes lead to a more reactive mixture with a 10 times shorter ignition delay. The small difference between the constant Lewis number approach and the two more detailed models can be attributed to thermodiffusion (Soret effect), which is lacking in the former approach.

To better understand how chemistry affects the ignition process, homogeneous reactor simulations are performed. Often, the initial conditions for such simulations at a given $Z$ are taken as a linear combination of the fuel and oxidizer stream corresponding to a unity Lewis number assumption. This will lead to ignition delay times which are not representative for non-homogeneous systems because of the preferential diffusion effects which change the initial condition on a shorter time scale than the chemistry does. Therefore, the initial conditions for the homogeneous simulations are taken from the solution of a steady non-reacting counterflow mixing problem including differential diffusion. In mixture fraction space, this solution is similar to the profiles observed in Fig. 4 for the non-unity Lewis number case at $t = 0.1$ ms.

The results are presented in Fig. 5 in terms of ignition time $\tau_{ig}$, which is the time after which the maximum temperature gradient $dT/dr$ is reached. As expected, $\tau_{ig}$ increases with increasing dilution level due to the reduced oxygen concentration. The curves in Fig. 5 show a minimum ignition time, $\tau_{ref}$, at the most reactive mixture fraction $Z_{MR}$. For the HM1 case, $Z_{MR}$ almost coincides with the stoichiometric value $Z_{st}$. For the other cases, $Z_{MR}$ is slightly larger than for HM1, but the ratio $Z_{MR}/Z_{st}$ is much smaller because the stoichiometric mixture fraction has increased significantly. The minimum ignition times, $\tau_{ref}$, are 0.19, 0.11 and 0.08 ms for HM1, HM2 and HM3, respectively. These values are almost 50% lower than those of the ignition delay times found in the mixing layer. The cause of these differences is diffusion, which retards the ignition time in non-homogeneous systems (see e.g., [8]).
The relative sensitivity of the autoignition time to the reaction rate constants $A_k$ is defined as

$$S_{\tau_{ig}} = \frac{\partial \ln \tau_{ig}}{\partial \ln A_k}, \quad (3)$$

and it has been computed at $Z_{MR}$ by evaluating the change in $\tau_{ig}$ due to a small perturbation in $A_k$. Similarly, one can define the relative sensitivity $S_{\tau_{init}}$ of $\tau_{ig}$ to the initial conditions in terms of major species’ mass fractions and temperature. Results for the HM1 case are shown in Figs. 6 and 7. The results clearly indicate that the chain-branching reactions in the H$_2$–O$_2$ mechanism play a dominant role together with the chain-initiation reaction between H$_2$ and O$_2$. These results are also reflected in the sensitivity to the initial condition. Besides the initial temperature, the autoignition time is also sensitive to the mass fraction of H$_2$ and O$_2$. Methane, H$_2$O and CO$_2$ have almost no influence on $\tau_{ig}$. It can be concluded that the ignition time is controlled by hydrogen chemistry.
Therefore, differential diffusion, which results in a faster diffusion of $H_2$ into the hot oxidizer stream, has a large impact on the ignition time.

3.2. Two-dimensional mixing layers

The DNS results of the two-dimensional turbulent mixing layers are now presented. In Figs. 8 and 9, the evolution of $Y_{H_2}$ and $Y_H$ is shown for the three cases. The contour plots of $H_2$ show the growth of the shear layer instability. This instability forms vortical structures which interact and merge. Due to the very small values of $Z_{st}$, the stoichiometric isocontours lie at the outside of the mixing layer and appear to enclose the turbulent region. As a result, the reaction layers are only mildly influenced by turbulence. The Karlovitz numbers $Ka = \tau_{ref}/\tau_\eta$ vary between 4 and 10 for these cases, indicating a much stronger interaction. Note, however, that the turbulence time scale $\tau_\eta = (v/\epsilon)^{1/2}$ is estimated from the velocity fluctuations in the core of the jet. In the core of the jet, mixing is dominated by turbulent eddies. In the reaction zone, however, the velocity fluctuations are much weaker, making molecular diffusion the main mixing process. Comparing the results of the three cases, it can be seen that the dilution level hardly influences the flow pattern. The different oxidizer mixtures have only very small differences in density and viscosity, leading to almost the same hydrodynamic behavior.

The autoignition time, however, differs significantly between the three cases. This can be seen in the contour plots of $Y_{H_2}$. The $H$ radical plays a central role in the chain-branching reactions and is therefore an excellent marker for the ignition process. As seen in Fig. 10, ignition delay times in the perturbed mixing layer are similar to those found in the laminar mixing layers. Furthermore, ignition appears to start near the stoichiometric isocontours. Distinct ignition spots at regions of small scalar dissipation rates as found, for example in Refs. [18,19], are not observed here. For the HM2 and HM3 cases, ignition appears to start everywhere along a $Z_{st}$ contour at the same time. For the HM1 case, small variations in $Y_{H_2}$ can be observed during ignition.

Ignition behavior can be studied in more detail by looking at Fig. 10, in which the maximum mass fraction of $H$ in the domain is shown as function of time. The exponential growth due to chain-branching reactions during the induction time can be clearly observed. Furthermore, the results for the laminar and turbulent mixing layers
almost coincide. Only for the most diluted case (HM1) does the turbulent mixing layer ignite 0.03 ms earlier. This effect can be attributed to variations in the scalar dissipation rate $\chi$, as is explained in the following. Since the ignition time of HM1 is the longest, there is more interaction of the developing turbulence with the chemistry for this case, leading to the largest differences between the laminar and turbulent mixing layer.

Fig. 11 also shows the maximum mass fraction of H for HM1, but these results are now compared with results from a homogeneous reactor simulation and a simulation of an igniting counterflow flame with a strain rate of 100 s$^{-1}$. The scalar dissipation rate of the counterflow geometry is in between that of the homogeneous simulation ($\chi = 0$) and that of the mixing layers. It can be seen that diffusion processes quantified by $\chi$ retard the ignition time.

In Fig. 12, scatter plots of $Y_H$ and $\log \chi$ are shown together with their conditional (on mixture fraction) means during the induction phase and they are compared with results of the laminar mixing layer. Although the conditional mean scalar dissipation is higher in the turbulent case, many flow locations in the turbulent simulation have $\chi$ values that are smaller than in the laminar case. At these points the growth of $Y_H$ proceeds faster than at points with a higher $\chi$. This results in a large scatter in the $Y_H$ plot and finally it leads to a faster ignition of the 2D turbulent mixing layer. The rather low value of $\chi_{st}$ leads to a large flame Damköhler number $Da = (\tau_{ref}/\chi_{st})^{-1} \approx 10^4$.

Although the ignition time is changed significantly by diffusion effects (see Fig. 11), the ignition chemistry itself is less affected. In Fig. 13, the maximum source term of H is plotted as function of the maximum value of H for the same flame configurations as in Fig. 11. It can be seen that the
profiles coincide during the induction time. This means that the chemistry in simple 1D or 0D flame configurations can be used to efficiently compute more complex configurations by using tabulated chemistry with a progress variable approach [10,12]. Furthermore, it indicates that \( \chi \) is not needed as an additional parameter for the tabulation of ignition chemistry in these mixtures.

4. Conclusions

DNS of igniting mixing layers have been performed using detailed chemistry at conditions similar to those of the jet-in-hot-coflow experiments in Ref. [3]. In order to investigate what kind of reaction–diffusion structures arise in these flows, the DNS results are analyzed in terms of chemistry and molecular diffusion. The main conclusions can be summarized as follows:

1. The ignition process of this CH\(_4\)–H\(_2\) mixture is dominated by hydrogen chemistry and its associated high-temperature chain-branching reactions.
2. Non-unity Lewis number effects are of the utmost importance for modeling of autoignition in mixing layers with a fuel stream containing hydrogen.
3. High scalar dissipation rates delay the ignition time, but have a negligible effect on the ignition kinetics.
4. Due to the very small values of \( Z_{st} \), the reaction layer lies at the edge of the turbulent mixing layer, resulting in rather weak turbulence–chemistry interaction.

5. With increasing dilution the autoignition time is increased, which allows the developing turbulence to interact more with the ignition chemistry.

In 3D mixing layers the turbulence is expected to develop faster, probably leading to a stronger interaction. Also, the presence of turbulence in the oxidizer stream could have this effect. For the 3D case, larger fluctuations in the scalar dissipation rate are to be expected in the turbulent region. However, since the stoichiometric iso-surface lies at the edge of the turbulent region, the scalar dissipation at \( Z_{st} \) will be affected to a much lesser extent. Therefore, 3D effects are not expected to change the conclusions. They will change the results quantitatively, but not qualitatively.

Future research utilizing the presented data will encompass the development of tabulated chemistry models and sub-grid scale models for LES of MILD combustion.

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References