Predicting NO Formation with Flamelet Generated Manifolds

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Abstract
With the increasing tightness of emission limits, models for pollutant formation become more and more important. Steady flamelet models have proven to be very efficient methods to model combustion. However, formation of nitrogen oxide and soot are relatively slow chemical processes, which cannot be assumed in quasi-steady state and therefore need additional treatment. In this study, two methods to model NO formation using steady flamelet tables are investigated. The results are compared with simulations using the full chemistry model. It appears that accurate results can be obtained from the table when \( \alpha \) is large enough \((\alpha > 10)\) to minimize interpolation errors. Solving an additional transport equation for NO is less sensitive to interpolation errors of the look-up procedure and results in slightly more accurate results than direct look-up.

Introduction
Reducing emissions of pollutants (e.g., \( \text{NO}_x \) and soot) is one of the major goals of designing new combustion equipment. Efficient and accurate numerical predictions of emissions can play an important role in the design process. In order to have reliable predictions detailed chemical models are needed, which usually include a large number of chemical species and reactions. However the use of such detailed chemical models is often prohibited due to the enormous numerical cost involved. Especially in industrial applications with complex geometries and turbulent flows, the need for efficient reduced chemical models is evident.

The goal of simplified chemical models is to reduce computational cost of combustion simulations without loosing (too much) accuracy. In order to realize this, different approaches are used. The first approach is the so-called chemistry based approach, in which a time scale analysis of the chemical source term appearing in the transport equations is performed. The chemical processes related to the fastest time scales are assumed to be in local equilibrium. This results in a reduced reaction system with a lot less degrees of freedom than the complete system. Examples of this approach are CSP [1], ILDM [2], and ICE PIC [3]. Usually these techniques compute the possible states of the reduced chemical system (low-dimensional manifold in the state space of the full system) in a pre-processing step and store this information in a look-up table as function of so-called controlling (or progress) variables.

The second approach to reduce computational cost of combustion simulations is the use of flamelet models. In laminar flamelet models it is assumed that the wildy moving flame in a combustion application can be regarded locally as a one-dimensional idealized flame front (flamelet). The complex structure of a flamelet is described by one or two degrees of freedom. A flame in a combustion system is then composed of a superposition of such flamelets, which “live” on the aerodynamic structure of the flow. In 1986, Peters [4] developed the basis of these flamelet models, which are still enormously popular in the combustion community. Usually these models are implemented by computing non-premixed flamelets and storing them in a look-up table as function of mixture fraction \( Z \) and scalar dissipation rate \( \chi \).

The third approach consists of flamelet-based manifold methods. These methods combine the advantages of the two above-mentioned approaches. In the time scale analysis not only reaction kinetics are considered, but transport processes as appearing in laminar flamelets are taken into account as well. This leads to a reaction model that can be applied like manifold methods, but with much manifolds of much smaller dimension. In practice a look-up table is constructed from premixed or non-premixed flamelets depending on the application. The manifold is parameterized by controlling variables consisting of linear combinations of species mass fractions. The FGM [5–8] and FPI [9] techniques are examples of this approach, which have been used successfully in many different flame studies. In this paper we will apply the FGM technique to model NO formation in unsteady laminar flame simulations.

Chemical reduction methods all share the same principle that there is a separation of time scales and that fast processes can be assumed to be in a quasi-steady state. However, since pollutant formation (especially \( \text{NO}_x \)) is related to “slow” chemical processes, it has to be treated with special care. For the chemistry based methods this usually means that additional degrees of freedom have to be used, which results in higher computational demands. For the flamelet models the problem is solved by using unsteady flamelets which are computed at runtime. This results in accurate NO predictions, but at an increased computational cost. In the FGM approach, a database is constructed from steady flamelets. Since the use of steady flamelets in standard flamelet models results in incorrect NO predictions, the question is whether it is possible to get accurate results with steady flamelets in the FGM approach.

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Two approaches can be distinguished when using steady flamelets:

1. Include NO in definition of reaction progress variable \( \mathcal{Y} \)
2. Solve a transport equation for NO with the source term from the look-up table

With mixed results both approaches have been applied in simulations of turbulent flames [10, 11]. It is not easy to draw conclusions from these simulations because the predictions are strongly influenced by the turbulence modeling and the accuracy of the NO measurements is not always high enough. Therefore, we will compare both approaches in simulations of unsteady one-dimensional laminar flames. This allows us to validate the reduced models against simulations with detailed chemistry.

In the following section we will explain the FGM method and how it is adopted to model NO formation. After that the one-dimensional test problem is described and the results are presented. The paper ends with conclusions.

**Flamelet generated manifolds**

Flamelet generated manifolds are constructed from solutions of steady laminar flamelet simulations with detailed chemistry. In accordance with the application, premixed methane-air flamelets are used here. Since the equivalence ratio is not fixed in the test problem, a series of premixed flamelets is computed for mixtures of different stoichiometry. This set of flamelets forms a two-dimensional manifold, which can be parameterized by mixture fraction \( Z \) and a progress variable \( \mathcal{Y} \). Usually the progress variable is defined as a combination of major species. In this case

\[
\mathcal{Y} = \frac{Y_{H_2}}{M_{H_2}} + \frac{Y_{H_2O}}{M_{H_2O}} + \frac{Y_{CO_2}}{M_{CO_2}},
\]

where \( Y_i \) and \( M_i \) are the mass fraction and the molar mass of species \( i \), respectively. Since the chemical state is now described by these two controlling variables, only transport equations for these variables have to be solved and not for all chemical species. All thermochemical data that is needed to solve these transport equations and the flow equations are stored in a look-up table. All species mass fractions are stored in a table as well, which is used during post-processing of the results.

Since the formation of NO is a rather slow chemical process, the progress variable has almost reached its maximum value, while the mass fraction of NO is still increasing. This results in extremely large gradients in the database \( \partial Y_{NO}/\partial \mathcal{Y} \rightarrow \infty \) and, hence, in large interpolation errors during data retrieval. This effect can be observed in Fig. 1. In this figure the NO mass fraction is shown versus progress variable at stoichiometric conditions \( (Z = Z_{st}) \). It can be seen that the mass fraction of NO increases sharply when the progress variable has nearly reached its maximum value \( \mathcal{Y} = \mathcal{Y}_{\text{max}} \). Obviously, this choice of parametrization would result in huge interpolation errors. One way to solve this problem is to add the mass fraction of NO to the definition of the progress variable, viz.

\[
\mathcal{Y} = \mathcal{Y}^0 + \alpha Y_{NO}/M_{NO}.
\]

The resulting parametrization is also shown in Fig. 1 for different values of \( \alpha \). It can be seen that the gradient \( \partial Y_{NO}/\partial \mathcal{Y} \) becomes smaller for increasing \( \alpha \). Due to the very small values of \( Y_{NO} \) compared to the major species, a rather large value of \( \alpha \) is needed to get a significant contribution of \( Y_{NO} \) in the progress variable.

A similar observation can be made for the chemical source term \( \dot{\rho}_{NO} \) of NO, which is shown in Fig. 2. Although the effect is less dramatic then for the NO mass fraction, also interpolation errors in the source term may occur. Since this source term is used to solve the transport equation for NO, it also results in errors in the NO predictions.

In this work, a two-dimensional manifold is generated from premixed methane-air flamelets. The mixture fraction is varied between \( Z = 0.025 \) and \( Z = 0.09 \) by
changing the stoichiometry of the reactants. The manifold is parameterized by $Z$ and $\mathcal{Y}$ and it is stored in a table using a grid of $151 \times 301$ points, which are equidistantly distributed in each direction. In Fig. 3 the chemical source term $\omega_Y$ of the progress variable is shown as function of the controlling variables $Z$ and $\mathcal{Y}$.

**Test problem and results**

We will apply the two approaches for NO modeling as described above to a simple test problem. It consists of a one-dimensional, adiabatic, premixed methane-air flame, which is subjected to sudden change in stoichiometry. The initial solution consists of a steady lean premixed flame ($Z = 0.5Z_{\text{st}}$). At $t = 0$ the equivalence ratio of the inlet mixture is changed to rich conditions ($Z = 1.7Z_{\text{st}}$). This is done by prescribing the composition of the unburnt mixture at the inlet of the computational domain ($x = -1$ cm). The flame is stabilized in the domain by adapting the inlet gas velocity in such a way that the temperature at $x = 0$ remains constant, i.e. $T(x = 0) = 1500$ K. At $t = 0.25$ s the unburnt mixture composition is switched back to lean conditions.

In Fig. 4 the profiles of mixture fraction $Z$, temperature $T$, and NO mass fraction $Y_{\text{NO}}$ are shown as function of space $x$ and time $t$. These results have been computed using detailed chemistry, i.e. the GRI 3.0 reaction mechanism. In the plot for mixture fraction, it can be seen that the inlet composition suddenly changes at $t = 0$ and $t = 0.25$ s. While this sudden change in mixture fraction travels with the gas velocity towards the flame front, it is smoothed by molecular diffusion. When this mixture fraction "wave" travels through the flame front, its wavelength is increased by the increase in gas velocity due to expansion of the reaction products.

In the temperature plot, it can be seen that when the mixture fraction wave hits the flame front the temperature rises, because the stoichiometry changes from lean to stoichiometric. When the wave has passed the reaction front, the temperature decreases again since the conditions have changed from stoichiometric to rich. When the mixture fraction is changed back to lean conditions at $t = 0.25$ s, a similar but reversed process can be observed.

The profiles of NO mass fraction closely follow the temperature profiles. When the mixture fraction is close to stoichiometry and the temperature is high, a lot of NO is produced. This results in a growing NO peak which is convected away from the flame front by the burnt gases. This can also be observed in Fig. 5, in which snapshots of the NO profile are shown at three time instants. In the
Figure 5: Profiles of NO mass fraction $Y_{NO}$ at three different times computed with detailed chemistry and four different FGM tables and two different approaches: direct look-up (left) and solving a transport equation for NO (right).
top figures the mixture fraction has just increased, in the middle figures a steady rich condition is obtained, and in the bottom figures the mixture fraction has just changed back to lean conditions.

In Fig. 5 also the results of FGM simulations with the two different approaches and different values of $\alpha$ are shown. The results with $\alpha = 10$ indicate that adding a small amount of NO to the progress variable does not prevent large interpolation errors. In fact, for this value of $\alpha$ the profile of NO is still not resolved by the grid spacing used in the look-up table. The interpolation errors result in an overestimation of the mass fraction of NO, but an underestimation of its chemical source term. For larger values of $\alpha$, the results become more accurate. The steady situation at $t = 0.2$ s is then very well captured by both approaches. However, the unsteady parts of the simulation are more accurately predicted by the approach in which a transport equation for NO is solved. The direct look-up approach gives only acceptable results for $\alpha = 10^3$. Note that in the limit $\alpha \to \infty$, both approaches are the same because then the progress variable $\gamma$ is equivalent to $Y_{NO}$.

Conclusions

The conclusions of this work can be summarized as follows:

1. Both approaches to predict NO (direct look-up and solving a transport equation) suffer from severe interpolation errors due to large gradients in the manifold for small values of $\alpha$.

2. Adding NO to the definition of the progress variable results in a smoother mapping of NO mass fraction and its chemical source term and reduces the interpolation errors.

3. Solving a transport equation yields more accurate results than direct look-up for transient simulations.

4. Despite the use of steady flamelets to generate the look-up table, accurate and efficient simulations of NO$_x$ formation are possible using FGM.

References


