Predictability of existing chemical kinetics in MILD combustion

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Introduction

As the use of detailed chemical mechanisms in high fidelity simulations is becoming more approachable, due to increased computational power, the question still remains if existing chemical mechanisms are able to accurately predict these conditions. Considering that in MILD combustion the mixing between the fuel and oxidizer is very fast, with respect to conventional conditions, the ratio between the mixing and chemical time scale is close to unity, i.e the Damköhler number is close to unity, and the kinetics plays an important role in the simulations, more so than for conventional conditions where the mixing time scale usually is the limiting aspect. It is therefore important to consider detailed kinetics in MILD combustion. However, as most kinetic mechanisms have been developed for conventional conditions, their performance in MILD combustion is not necessarily adequate. An example of this can be seen in Figure 1, where the ignition delay time for biomass pyrolysis gas at stoichiometric conditions is plotted for different inlet temperatures, with different kinetic mechanisms. The number of kinetic parameters in a mechanism can roughly be estimated to 3 times the number of reactions in the mechanism. The number of kinetic parameters are therefore increasing linearly with the complexity of the mechanism, and as these kinetic parameters are determined through either experiments, theoretical calculations, or estimations, the number of uncertain parameters inside a mechanism is also linearly increasing with the size of the mechanism. For this reason, a large part of the numerical uncertainty in MILD combustion can be assigned to the kinetics. A way to tackle this is by using Optimization or Uncertainty Quantification (UQ) techniques [1] to use existing experimental data to determined which parameter combinations are the optimal ones or to reduce the uncertainty of the kinetic parameters in order to more accurately predict the experimental data. However, as detailed kinetic mechanisms generally consists of thousands of reactions, the optimization of all parameters would be unfeasible. The determination of the most impactful parameters is therefore crucial in the optimization process. In previous Optimization/UQ studies, rankings of the reactions were based on local sensitivity coefficients [2], impact factors [3, 4], or optimization potential [5]. In this work a new technique will be applied in order to determine which specific kinetic parameters that have the highest impact with respect to the experimental data. The second problem that we are faced with during optimization, is the amount of target experimental data. The more data used, the more validated and widely applicable the mechanism becomes. However, as the number of data points increase, also the computational effort increases. There is therefore a need for an effective optimization tool, which is able to consider many experimental data points, as well as the most impactful kinetic parameters. The focus of this work is to use such a tool, which consist of a coupling between OpenSMOKE++ [6] and Dakota [7], considering the most impactful parameters for a large set of experimental data in MILD conditions.

Methodology

This work was based on the experimental data from Sabia et al. [8] which consists of auto-ignition delay times for biomass pyrolysis gas in a Plug Flow Reactor (PFR). A 10 K increasing in temperature was used as criterion to detect the onset of ignition. The experiments were then numerically reproduced using a PFR model in OpenSMOKE++ [6]. The selection of the parameters for optimization was carried out with a new technique whith the following procedure; first, a local sensitivity analysis, with respect to temperature, was performed. Subsequently, the absolute values of the sensitivity coefficient for every reaction in the mechanism was extracted on the onset of ignition for each experimental data condition. Then, an average sensitivity vector based on the overall data set was derived, sorted and used to compute a Cumulative Sensitivity Function (CSF). By imposing a threshold value on this CSF, it's possible to associate a set of reactions with a certain percentage of the overall sensitivity. Secondly, a local brute force sensitivity analysis was performed in order to assess the direct impact of every single parameter on ignition delay time variations. The latter process was performed by coupling OpenSMOKE++[6] and Dakota[7], using a python interface. The parameters related uncertainty were computed by following the methodology reported in [4]. Similarly as for reactions, a CSF was derived for the considered parameters, although here the uncertainty range for each parameter was taken into account, resulting in a Cumulative Impact Function (CIF). A final subset of parameters was then obtained. Then the optimization was performed by coupling the two software OpenSMOKE++[6] and Dakota[7], using a C++ interface. The Dakota toolbox was developed by Sandia National Laboratories, and it is used to interface simulation codes with a number of different analysis tools, such as optimization, uncertainty quantification, sensitivity analysis, parameter studies, etc. The specific tool used for this study was the Evolutionary Algorithms optimization tool, which is suitable for strongly non-linear global optimization problems, such as the optimization of chemical kinetics. Evolutionary Algorithms uses the approach of "survival of the fittest", which is initially determined by a set of random samples based on the parameter space. The samples which provides the best objective function value, is then chosen and further mutations/combinations of these samples are used in order to find the globally optimal point in the parameter space.

Results

An initial evaluation of existing detailed kinetic mechanisms was performed in order to determine which one to use for further evaluation. The list of mechanisms, together with number of species, reactions and reference can be found in Table 1. The results at stoichiometric conditions are presented in Figure 1 below. It can clearly be seen that none of the mechanisms are able to accurately predict the ignition delay time, especially at high inlet temperatures. There is therefore a need for optimizing the kinetics with respect to these data, in order to accurately predict the onset of ignition in MILD combustion. The final choice of the mechanism will be based on a much larger set of experimental data, and the one that overall predicts the ignition delay times, will be chosen for the optimization.

Table 1: List of kinetic mechanisms used for the initial evaluation. The corresponding number of species, reactions and reference is also listed.

Mechanism	Number of Species	Number of Reactions	Reference
Aramco 1.3	124	766	[9]
Aramco 2.0	502	2716	[10]
GRI 2.11	49	279	[11]
GRI 3.0	53	325	[12]
POLIMI 1412	107	2642	[13]
POLIMI 1810	124	766	[13]
SanDiego	58	270	[14]

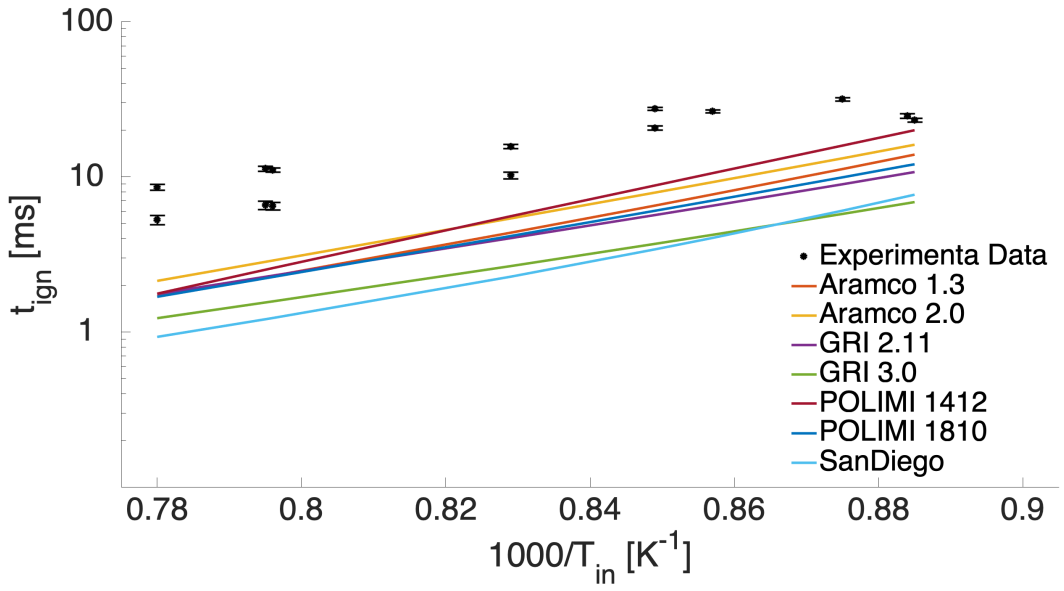

Figure 1: Ignition delay time at stoichiometric conditions vs one thousand over the inlet temperature. The experimental data from [8], are represented by the black dots and the simulation results are represented by the colored lines.

Conclusions

Existing detailed kinetic mechanisms are not always accurate in predicting key aspects of MILD combustion, as they have been developed and validated for conventional combustion. The usage of Optimization/UQ techniques are therefore a promising option, in order to improve the performance of existing detailed mechanisms with respect to MILD conditions. In this work only a preliminary study has been put forth, but the application of said methodology will be used on a large set of data point in order to improve the performance of an existing mechanism with respect to MILD combustion.

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