Predictability of existing chemical kinetics in MILD combustion

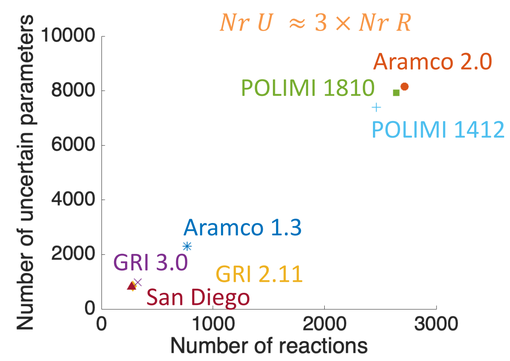
M. Fürst[1,2,3], A. Bertolino[1,2,3], A. Frassoldati[2] and A. Parente[1,3]
[1]Université Libre de Bruxelles, Ecole Polytechnique de Bruxelles, Aero-Thermo-Mechanics Laboratory
[2]Politecnico di Milano, Department of Chemistry, Materials, and Chemical Engineering
[3]comBUstion and Robust optimizatioN group (BURN), Université Libre de Bruxelles and Vrije Universiteit Brussel
Contact: magnus.furst@ulb.ac.be

Introduction

As computational power is drastically increasing, the use of detailed kinetics in high fidelity simulations, is becoming more feasible. However, with increasing mechanism size, the number of kinetic parameters are increasing, and therefore also the number of uncertain parameters.
As MILD combustion is characterized by low Damköhler numbers, the use of detailed kinetics is especially important.
Optimization techniques therefore presents an opportunity for improving the performance of chemical kinetics with respect to MILD combustion.

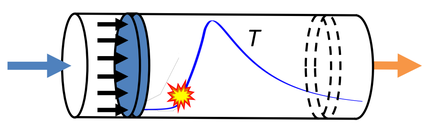
Table 1: List of kinetic mechanisms evaluated in this work, together with number of species, reactions and reference.

Mechanism	Number of Species	Number of Reactions	Reference
Aramco 1.3	124	766	[1]
Aramco 2.0	502	2716	[2]
GRI 2.11	49	279	[3]
GRI 3.0	53	325	[4]
POLIMI 1412	107	2642	[5]
POLIMI 1810	159	2463	[5]
San Diego	58	270	[6]



Methodology

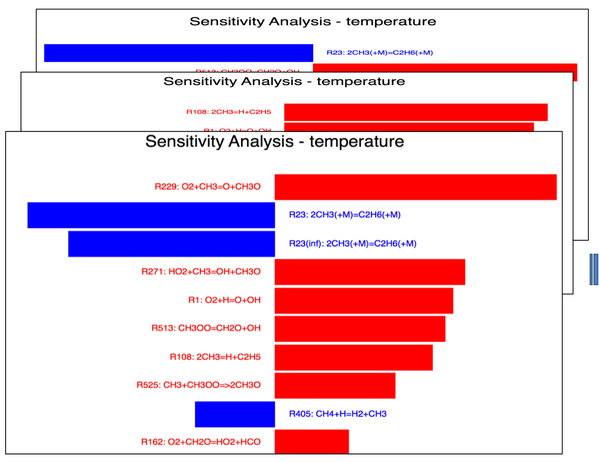
The experimental data used, were from Sabia et al. [7], where the ignition delay time for biomass pyrolysis gas was evaluated in a Plug Flow Reactor (PFR). The experiments were performed at different inlet temperatures, equivalence ratios and dilutions.

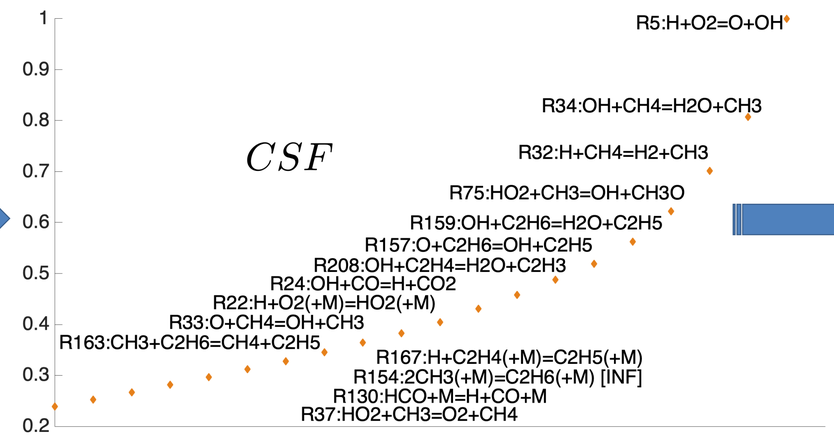


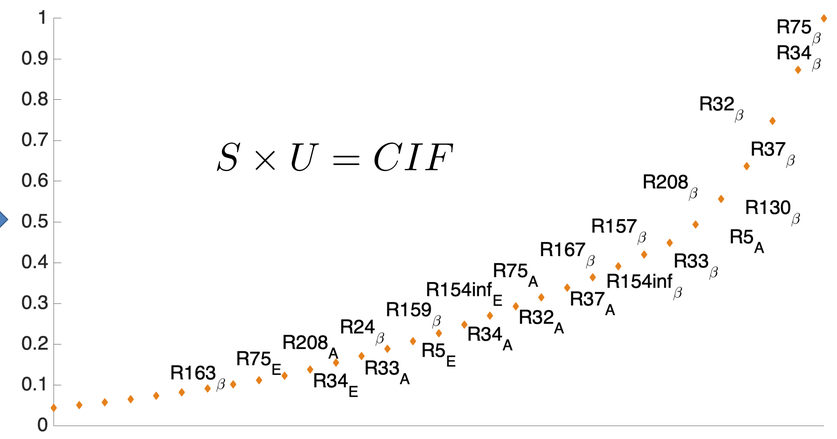
The optimization was performed using a coupling between the two software Dakota [8] and OpenSMOKE++ [9].

The choice of which kinetic parameters to optimize was done in the following steps:

- First local sensitivity analysis was performed
- These were then combined into a Cumulative Sensitivity Function (CSF)
- All kinetic parameters for the most sensitive reactions were then used in a local brute force sensitivity study
- The product of this sensitivity and the Uncertainty (U) of respective parameter then gave a Cumulative Impact Function (CIF)
- The most impactful parameters were then used in the optimization study







Results

Using the POLIMI 1810 mechanism, the kinetic parameters listed in Table 2 were used in the optimization.

Table 2: List of reactions and specific parameters used for the optimization.

Reaction	A	β	E
$H + O_2 = O + OH$	✓		✓
$OH + CO = H + CO_2$		✓	
$H + CH_4 = H_2 + CH_3$	✓	✓	✓
$O + CH_4 = OH + CH_3$	✓		✓
$OH + CH_4 = H_2O + CH_3$	✓	✓	✓
$HO_2 + CH_3 = O_2 + CH_4$	✓	✓	
$HO_2 + CH_3 = OH + CH_3O$	✓	✓	✓
$HCO + M = H + CO + M$		✓	
$2CH_3 (+M) = C_2H_6 (+M)$ [inf]		✓	✓
$O + C_2H_6 = OH + C_2H_5$		✓	
$OH + C_2H_6 = H_2O + C_2H_5$		✓	
$CH_3 + C_2H_6 = CH_4 + C_2H_5$		✓	
$H + C_2H_4 (+M) = C_2H_5 (+M)$		✓	
$OH + C_2H_4 = H_2O + C_2H_3$	✓	✓	

The optimization was done based on an objective function defined as:

Number of experimental conditions
Number of experimental points for condition i

$$\mathrm{Obj} = \sum_{i=1}^{N} \frac{1}{N_i} \sum_{j=1}^{N_i} \frac{\mid Exp_{ij} - Sim_{ij} \mid}{Exp_{ij}}$$

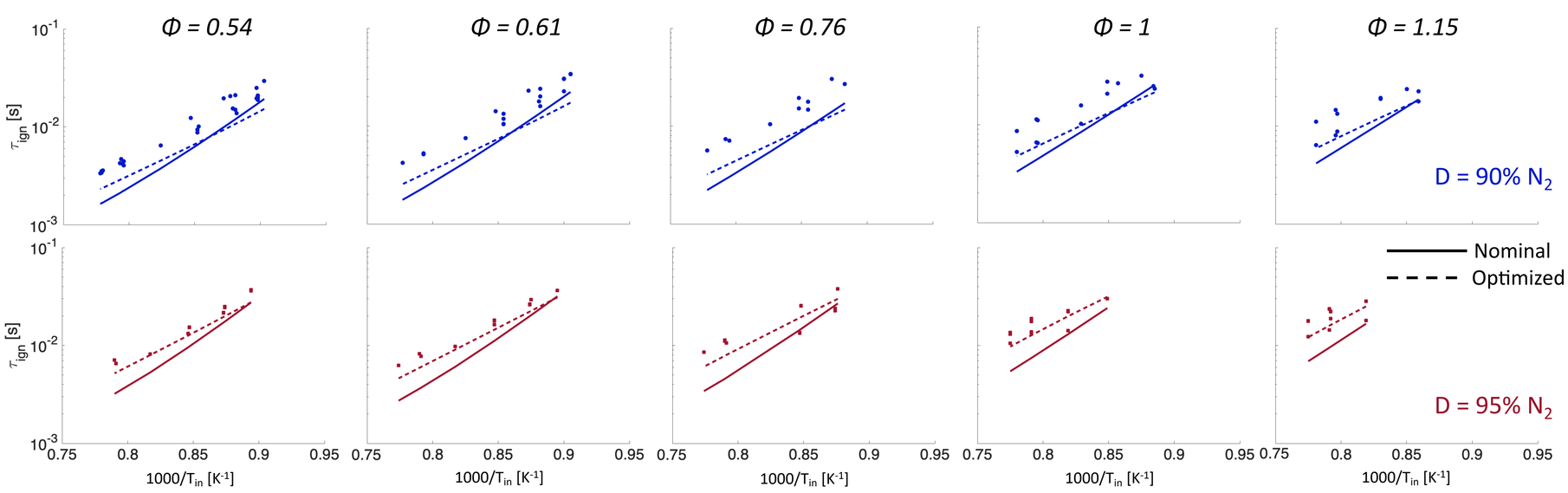


Figure 1: Ignition delay time vs the inverse of the inlet temperature for biomass pyrolysis gas at different equivalence ratios and dilutions.

Conclusions

In this work, the optimization of an existing detailed mechanism was performed. The following conclusions can be made:

- The most impactful parameters were determined using a new approach based on parameter specific local sensitivities and uncertainty ranges.
- A new tool for optimization, based on a coupling between Dakota and OpenSMOKE++, was evaluated and showed good performance.
- The optimized kinetics gave overall improvements for the prediction of ignition delay time, especially for the high dilution cases.

Acknowledgements

This work has been carried out in the framework of the Short Term Scientific Mission Program of SMARTCATs COST Action (CM1404, www.smartcats.eu), supported by COST (European Cooperation in Science and Technology, www.cost.eu) as well as it has received funding from the European Union's Horizon 2020 research and innovation program under the Marie Sklodowska-Curie grant agreement No 643134. The second Author acknowledges the support of Funds pour la Recherche Scientifique (FNRS) through a FRIA fellowship of the project "HOPTIMAL: Hierarchical development of OPTimised kinetic Mechanisms for Advanced combustion technoLogies", and the research of A. Parente was sponsored by the European Research Council, starting grant 714605.

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