Reaction Kinetics and Investigation of Laminar Flames

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Introduction and motivation

- Laminar flames occur in many lab scale processes in combustion science and for nanoparticle synthesis
- Mass spectroscopy and laser based measurement techniques are widely used to provide spatially resolved species concentrations and temperature fields
- The knowledge of the flow field is essential for analysis and development of reaction kinetics mechanisms
- Numerical simulations of chemically reacting flows require a finite rate chemistry model
- Number of species and reactions is a limiting factor for 2D and 3D CFD simulations
- A reduced model still needs to be “detailed”
- Reduction strategies correspond to a search problem
- Mechanism reduction permits a better understanding of the model
- Mechanism reduction was implemented with genetic algorithms

Simulation of laminar flames

- The finite rate chemistry of the laminar flame was calculated using a reduced mechanism containing 15 species and 21 reactions [1].
- In order to obtain accurate temperature and velocity field of the reacting gas, a two-dimensional laminar flame was simulated in OpenFOAM [2].
- The reduced mechanism used here cannot predict a correct distribution of species concentration, therefore the simulated temperature fields from OpenFOAM were used to reconstruct the species concentration through one-dimensional flame simulations in Cantera [3] using GRI 3.0 mechanism [4].
- Complementary CFD simulation of the laminar flame is necessary to reconstruct accurate experimental conditions for making the measurement a more reliable source for model verification.

Reaction mechanism reduction

- An efficient reduction approach is based on genetic algorithms to find a relatively small subset of reactions from the detailed mechanism, taking into account the user’s demands for accuracy and cost.
- Genetic algorithm (GA) is a stochastic search method that mimic natural biological evolution and it is used for problems that do not have a well defined solution and where efficiency is important as well.
- Accuracy of the reduced mechanism is determined by comparing its performance to the performance of the detailed mechanism in predicting important chemical properties (e. g. temperature and species profiles, ignition delay time etc.).
- Aim of the algorithm is to find a reaction subset (reduced mechanism) for which this difference and the computational cost are minimal.
- Elements of the algorithm (initialization, crossover, mutation, selection) can be adapted to the problem
- Algorithm runs in parallel

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Mechanism reduction strategies

Genetic algorithm:

- Reactions are mapped onto the genome
- Homogenous reactor model chosen for evaluation
- Accuracy criteria are temperature, ignition delay time and mole fractions of selected species
- Cost criteria are CPU time and the size of the mechanism

Search results

- Mechanism could be reduced from 325 to 60 reactions
- Maximum error not determined by a strict value
- Search converged after 405 generations
- Growth of the problem depends weakly on mechanism size

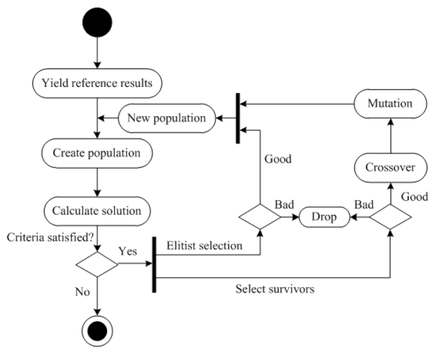


Fig. 1: Activity diagram of the GA search, [5]

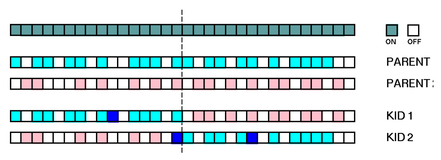


Fig. 2: Crossover between two individuals, mutation flips single genes (blue).

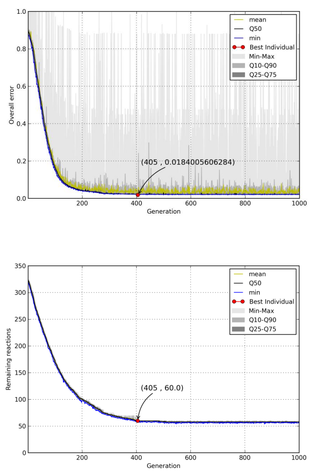


Fig. 3: Convergence plots of the GA search, [5]. The best performing genome was found in generation 405 with an overall error of 0.0184 and has 60 reactions.

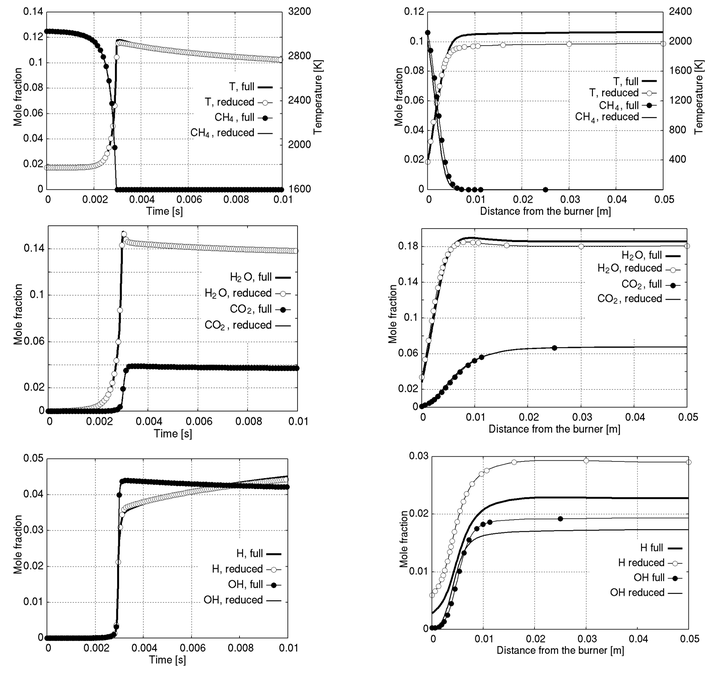


Fig. 4: Performance of the reduced GRI 3.0 for a homogeneous constant-pressure reactor and a burner-stabilized flame using the conditions from [5]

Simulation of mass spectroscopy measurements

Experimental configuration

- Sampling probe is an axisymmetric quartz cone with
  - Orifice diameter: 0.08mm, length: 20 mm
  - Inner angle: 40°, outer angle: 51°
- Premixed laminar flame composition: $CH_4/O_2/Ar$ (mole fractions: 0.06/0.15/0.79)
- Flow rate: 15 sccm
- Unburned gas temperature: 368 K, pressure: 1 atm

Simulation Results

- Perturbation due to this invasive probing technique is significant.
- Sampling zone is shown by the streamline pattern.
- A nearly perfect agreement was found in comparison to the measured CH_4 and H_2O.
- In the one-dimensional simulation strong acceleration of the gas was neglected, thus the simulation results at distances great than 0.5 mm show a better agreement with the experiment.

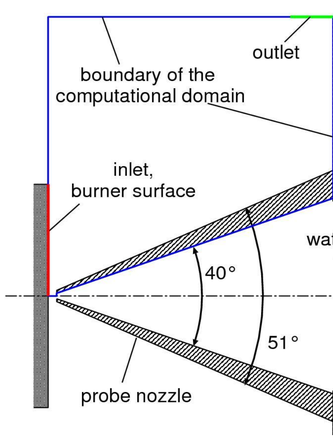


Fig. 5: Sketch of the experimental geometry and the computational domain [6, 7]

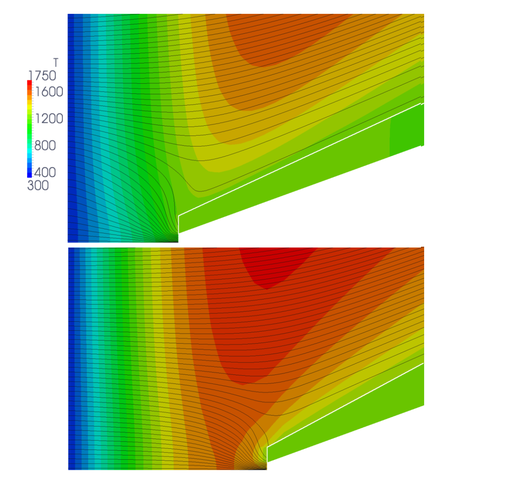


Fig. 7: Streamlines and temperature field at different probe positions (0.32 mm and 0.49 mm), [6]

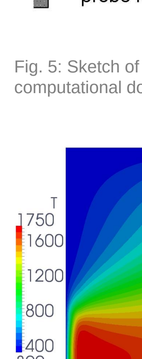


Fig. 6: Temperature field for a probe tip position at 1.06 mm [6]

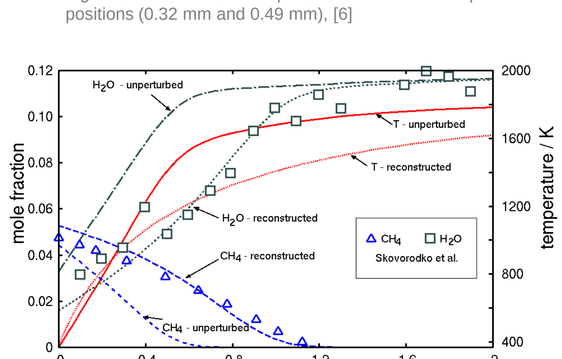
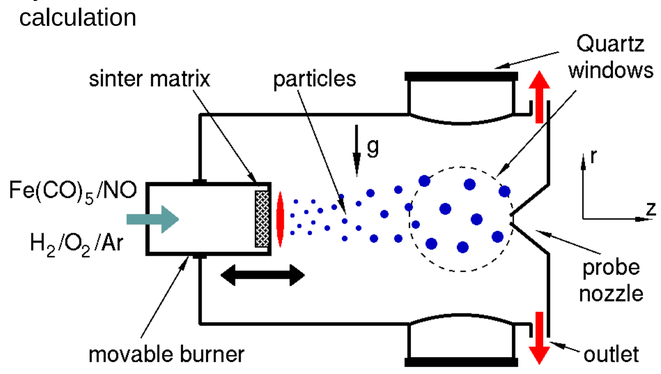


Fig. 8: Mole fractions of CH4 and H2O, measured (symbols), calculated for the undisturbed flame and calculated for the CFD flame reconstruction [6, 7]

Simulation of nanoparticle synthesis from a premixed laminar flame

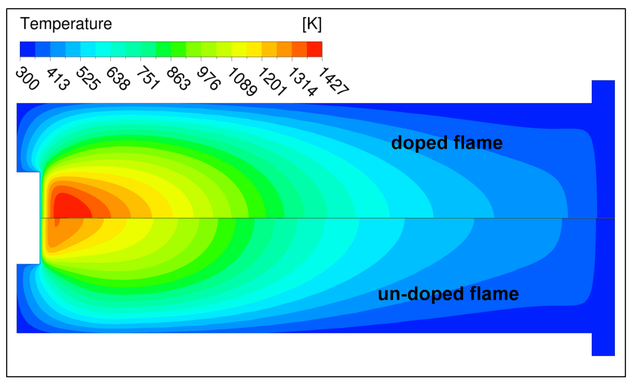
CFD “reconstruction” of a reactor flow

- CFD model assumed rotational symmetry and negligible buoyancy
- CFD calculations with and without $Fe(CO)_5$ correspond well to experimental observations [8]
- Simulations were performed with the reduced mechanism [9]
- Transport velocity and temperature for the 1D particle dynamics simulation were extracted from the CFD calculation



Case	Flow rate in sccm			$Fe(CO)_5$ in ppm	Data source
	H2	O2	Ar		
Hecht [4]	770	1040	575	35, 70, 170	LIF
This study	400	400	600	300, 2500	PMS

Fig. 9: Experimental setup of nanoparticle synthesis reactor (Pressure p = 3000 Pa)



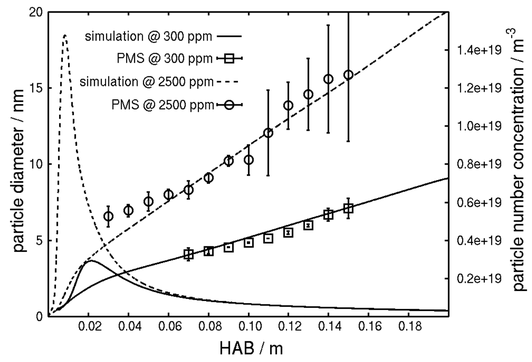


Fig. 10: Particle diameter and number concentrations from population balance equation model calculations in comparison to measurements [9, 10]