Reduced chemical kinetics for combustion and ignition

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Overview

• introduction
• ILDM
• problems and solutions
  – numerical calculation
  – manifolds of stationary states higher order
  – reduced ignition chemistry
  – implementations of reduced chemistry
• summary
Simulation of combustion processes

- combustion chemistry
- complete description by elementary reactions
  --> detailed mechanisms

- flow field
- conservation equations for every chemical species
  --> small mechanisms

Reduced mechanisms
- accuracy of detailed mechanisms
- small number of reaction progress variables
Reduction of detailed reaction mechanisms

**conventional methods**
- assumption of *partial equilibrium for reactions*
- *stationary state of species*

**disadvantage:** physical motivation of assumptions restrict the reduced mechanism to specified conditions (e.g. high temperatures, lean conditions)

**ILD M-method**
- reduction to the rate limiting slow part of the mechanism
- mathematical/numerical method
- only input: number of reaction progress variables
Homogeneous reactor in state space

fast reaction to Intrinsic Low-Dimensional Manifolds (ILDM) in state space
Idea: use ILDM as a basis for a mechanism reduction method (Maas, Pope 1992)
----> identification of rate limiting part of reaction mechanism
Tabulation of ILDM

- calculation of ILDM is computationally expensive
- solution: tabulation and use of fast table look-up instead of slow calculation
Application: homogeneous reaction

- initial conditions: partially burnt stoichiometric mixture of dodecane-air (temperature of unburnt mixture 300 K, 1 bar)
- lines: detailed mechanism; symbols: ILDM
- 2 reaction progress variables CO₂ and H₂O
- very good comparison both for main species CO₂ and CO as for the radicals O and CH

--- mechanism reduction with ILDM-method is highly accurate
Application: flame 2d-ILDM

- laminar premixed flame methane-air (stoichiometric, 300 K, 1 bar)
- lines: detailed mechanism; symbols: ILDM
- 2 reaction progress variables CO$_2$ and H$_2$O
- good comparison for main species CO$_2$, H$_2$O and O$_2$
- deviations with O-atoms

--- influence of diffusion on the relaxation of fast chemical time scales

--- improvement by additional reaction progress variable O$_2$
Application: flame 3d-ILDM

- laminar premixed flame methane-air (stoichiometric, 300 K, 1 bar)
- lines: detailed mechanism; symbols: ILDM
- 3 reaction progress variables CO₂, H₂O and O₂
- very good comparison for main species CO₂, H₂O and O₂
- small error for O-atoms

--->
3 reaction progress variables are sufficient for laminar premixed flames
Application: flame velocities

- laminar premixed flame heptane-air (800 K, 80 bar)
- lines: detailed mechanism; detailed transport
- white symbols: ILDM; Le=1
- filled symbols: measurements
Application: Non-premixed flame

• Laminar non-premixed flame heptane-air (stoichiometric, 300 K, 1 bar)
• 2 reaction progress variables CO$_2$, H$_2$O (temperature)
• Very good comparison for main species CO$_2$, H$_2$O and O$_2$
• Good comparison for minor species
Application: turbulent jet-flame

- non-premixed flame methane-air
- turbulence-chemistry interaction: PDF, Monte-Carlo-method
- 2 reaction progress variables $\text{CO}_2$, $\text{H}_2\text{O}$, temperature
Application: Diesel engine

- Turbulence-chemistry-interaction: Presumed PDFs
- 3-dimensional ILDM table (temperature, CO$_2$, H$_2$O)

Correa (2000)
Is ILDM the perfect method for mechanism reduction?

- All slides look fine!
- All relevant applications have been done!
- Where is the problem?
There still exist major problems!

- manifold
  - numerical calculation often fails
  - bug in theory? Strange results in homogeneous reactions
  - existence only in the burnt region: Ignition chemistry is not available

- tabulation
  - needs too much space

- implementation
  - needs experts
Definition of the manifold

- Chemistry:
  \[ \dot{x} = S(x) \]

- Local linearization
  \[ J = \frac{\partial S(x)}{\partial x} \]

- Eigenvalue decomposition
  - Order in fast and slow
  \[ J = E\Lambda \tilde{E} = \begin{pmatrix} E_s \, E_f \end{pmatrix} \begin{pmatrix} \Lambda_s \cr \Lambda_f \end{pmatrix} \begin{pmatrix} \tilde{E}_s \\ \tilde{E}_f \end{pmatrix} \]

- Definition of the manifold:
  \[ \tilde{E}_f(x)S(x) = 0 \]
Calculation of the manifold I

• Solution of algebraic equation system is difficult
  – nonlinearity and
  – stiffness of reaction mechanism

• Reason for stiffness:
  – thermodynamics
  – higher alcanes are extremely unstable at equilibrium temperatures

• Stiffness is increasing with growing numbers of C-atoms in fuels
Calculation of the manifold II

- Conversion of
  - algebraic equation system \( \tilde{E}_f(x)S(x) = 0 \)
  - with parameters
  - into an ODE system

\[
\begin{align*}
\dot{x} &= S(x) + \tilde{E}_s(x)q \\
\dot{q} &= f(x, p)
\end{align*}
\]

- Solution with standard stiff-stable ODE-solvers
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Bug in theory?

- Trajectories starting on the ILDM are leaving and deviating from the ILDM!
- Error of 40 K
- ILDM: black
- Trajectory: grey
Why there is a deviation?

- Small spectral gap
- Definition of ILDM: Local non-linearity of reaction is neglected
Investigation on a theoretical mechanism

- Reaction system

\[ A + A \rightarrow C \]
\[ B \rightarrow A \]

\[
\begin{pmatrix}
\dot{a} \\
\dot{b}
\end{pmatrix}
=
\begin{pmatrix}
-a^2 \\
+b \\
-b
\end{pmatrix}
\]

- bundling trajectories
Series of manifolds of stationary states higher order

- ILDM is not a good approximation for the trajectory
- Series is converging to the trajectory
Proof: Series of manifolds of stationary states higher order is attractor for trajectories.
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Reduced ignition chemistry

- Idea of ILDM: All chemical scales are in equilibrium
  - except the very slow ones

- Ignition: All chemical scales are frozen
  - except the very fast ones

- ILDM can not be used in ignition prediction!
Is a reduced ignition chemistry possible?

- Problem of dimensionality with ignition:
  - Equilibration of scales leads to a dimension reduction → no influence of initial conditions!
  - Ignition occurs with the dimension of the chemical state space (size of mechanism) → influence of initial conditions!

- No rigorous model with generally applicable reaction progress variables can be found for ignition
Reduced ignition chemistry

• But: In practical applications (e.g. CI engine) dimensionality of initial conditions is much smaller:
  – unreacted fuel-air mixture
  – variable temperature
  – variable pressure
• Idea: Use one reaction progress variable from trajectories
• Result: Table of reduced ignition chemistry with a few variables (mixture, temperature, pressure, one reaction progress variable)
Applied Reduced ignition chemistry in a CI engine

- Reduced ignition model + 3-dimensional ILDM (temp., CO$_2$, H$_2$O)
- Turbulence-chemistry-interaction: Presumed PDFs

Correa (2000)
Applied reduced ignition chemistry in a CI engine

Correa ((2000)
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Tabulation

Dimensionality of reduced chemistry in a CI engine
- Temperature/enthalpy
- Pressure
- Mixture
  - Mixture fraction
  - More rigorous: 3 element mole fractions
- Reaction progress variables
  - 1: seems to work not too badly
  - More are better!

Result:
- 4 axis are the minimum
- In-situ tabulation is the only way to handle that big tables
Application of in-situ tabulation in a CI engine
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CFD-code

• How to implement tabulated manifold chemistry?

• Problems:
  – Reaction progress variables (RPV) are very different from species
  – Different RPV for ignition and combustion
  – Variable number of RPV $\rightarrow$ variable number of conservation equations
Conventional implementation of ILDM

- Both RPV and species required
- Small reduction in space and computation time
- Diffent #RPV is a hard problem
Idea of skeleton mechanism implementation

- Manifold chemistry should be usable like a skeleton mechanism
  - small number of species
  - Low stiffness
  - Details of the manifold (different models for ignition and combustion, variable dimension, ...) should be hidden from the user
  - Physical interface layer
Idea of skeleton mechanism implementation

<table>
<thead>
<tr>
<th>chemischer Quellterm</th>
<th>nichtchemischer Transportterm</th>
</tr>
</thead>
</table>

klassische Implementierung

<table>
<thead>
<tr>
<th>( \tilde{S}(\theta) )</th>
<th>( \Xi )</th>
</tr>
</thead>
</table>

| neue Implementierung mit Skelett-Spezies |

\[
\begin{align*}
\dot{S}_s & \quad \dot{S}_f \\
\Xi_t & \quad \Xi_f
\end{align*}
\]
Skeleton mechanism implementation

- CFD-code is unchanged except the chemical source term
- Details of the manifold chemistry hidden in the chemical source term
Are there more problems?

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• implementation
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Summary: Reduced chemical kinetics for combustion and ignition

Combined Reduced Chemistry
- Manifold chemistry (ILDM, stationary states higher order) for the combustion/equilibration zone
- Reduced ignition chemistry
- In-situ tabulation
- Skeleton mechanism implementation