Approaches to the incorporation of realistic chemical kinetics in multidimensional engine combustion simulations

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   - Challenges of simulation with detailed chemical kinetics

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   - The reduction-optimization approach
   - Some results
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Motivation and challenges

- After 4 000 centuries, **combustion** still accounts for more than 90% worldwide energy conversion.
- Burgeoning demand for energy supplies is urging research into more efficient combustion systems and sustainable alternatives to oil.
  - towards a **portfolio** of renewable energy sources.
  - **biofuels** can be candidate for gradually replacing petroleum-based fuels in transportation.
Motivation and challenges

- New combustion concepts (such as HCCI/PCCI, RCCI) show impressive improvements in conversion efficiency.
  - ICE indicated efficiency $\geq 50\%$
  - strong dependency on fuel chemistry and local mixture reactivity

- simple/phenomenological combustion models lack of resolution in modelling:
  - the whole range of operating conditions of practical systems
  - presence of exhaust gases in the mixture
  - simultaneous operation with multiple fuels
  - multi-component fuels chemistry
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Chemical Kinetics in CFD simulations

- Usually part of an operator-splitting scheme
- Each cell is treated as an adiabatic well-stirred reactor
  - embarassingly parallel problem
  - very stiff IVP
  - only the overall changes in species mass fractions and cell internal energy are passed to the flow solver

\[
\frac{\partial Y_i}{\partial t} = -\nabla \cdot (Y_i \mathbf{v}) - \nabla \cdot Y_i \mathbf{v}_{d,i} + \frac{1}{\rho_i} \dot{\omega}_i W_i
\]

\[
\frac{\partial E}{\partial t} = -\nabla \cdot (E \mathbf{v}) - \nabla \cdot (\mathbf{v} \cdot \mathbf{T}) - \nabla \cdot (\dot{Q} + \dot{Q}_r) + \mathbf{v} \cdot \sum_j m_j a_j + \sum_j \mathbf{v}_{d,i} \cdot m_j a_j
\]
Three levels of interaction

- Fluid Flow solver
- Grid
- ODE solver
- Kinetics
- Mechanism

Act on three levels
The reduction-optimization approach

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Reaction mechanism sizes for biofuels

- Number of reactions, \( n_r \)
- Number of species, \( n_s \)

\[ n_r = 5 \cdot n_s \]

- Ethanol (LLNL)
- Dimethyl carbonate (LLNL)
- Biodiesel (Golovitchev)
- DME (LLNL)
- Ethanol (Saxena)
- Methyl formate (LLNL)
- Small hydrocarbons (Konnov)
- MB (NUI)
- Ethers (NUI)
- Methyl esters (NUI)
- Methyl-9-decenoate (LLNL)
- Methyl-5-decenoate (LLNL)
- MD (LLNL)
- MD + nC\(_{16}\)H\(_{34}\)
The reduction-optimization approach

What amount of reduction?

- **Comprehensive mechanisms** aren’t viable for practical computations
  - 1000 to 3000 species, up to 10000 reactions
  - Jacobian matrix factorization scales with $\sim n_s^3$, diffusion iterations with $n_s^2$

- **Skeletal mechanisms** have very limited validity ranges
  - as few as 4 species, 10 reactions
  - they well predict only the main ignition event

**Idea**

**Reduced mechanisms** that contain the major reaction pathways
Optimized reaction rates to ensure desired validity ranges
The reduction-optimization approach

Key points for developing the reduced model

1. The reduced mechanism is derived from the detailed one as a **subset of species and reactions**

2. Methods for estimating subsets of ’important’ species are available in the literature (EF, DRG, ...)

3. An **error function** defines the accuracy of the reduced model in comparison to the full one

4. Exploiting average uncertainties in reaction rate constants, their values can be **optimized** to accomplish for the deleted reaction pathways
The reduction-optimization approach

An error function for the reduced mechanism

\[
f = -\log \left\{ 10^{-8} + \sum_{j=1}^{n_c} \left[ \sum_{k=1}^{n_s} \int_{\tau=0}^{\tau=t_j} W_k \frac{|X_{jk}^{\text{full}}(\tau) - X_{jk}^{\text{red}}(\tau)|}{X_{jk}^{\text{full}}(\tau)} d\tau \right] + \int_{\tau=0}^{\tau=t_j} \frac{|T_j^{\text{full}}(\tau) - T_j^{\text{red}}(\tau)|}{T_j^{\text{full}}(\tau)} d\tau \right\}
\]

**Task**: estimate the global error introduced into the mechanism by deletion of a subset of species and reactions

- Choice of \( n_c \) relevant cases that span the desired validity ranges
- Need **not to span orders of magnitude** for fitness-proportionate selection;
- Need to be both valid during the reduction and the optimization phases;
- Need to monitor **instantaneous time evolution** of the system;
- Need to evaluate **species concentrations** further than average thermal properties.
Setup as an optimization problem

- Independent variables: reaction rate coefficients $A_i, E_i$ in the Arrhenius formulation:
  $$k_{f,i} = A_i \ T^{b_i} \ \exp \left( \frac{E_i}{R \ T} \right), \quad i \in \{1, \ldots, n_r\}.$$

- Temperature exponent not included: most are zero.

- Reactions involved in well-established low-order schemes are not included:
  $$N_{basic} = \{H, H_2, O, O_2, OH, H_2O, HO_2, H_2O_2, N_2, CO, CO_2\}$$

- Average experimental (NIST) variability ranges for the reactions of C1-C3 hydrocarbons:
  - $\epsilon_A = \Delta A_i / A_i \approx 80\%$,
  - $\epsilon_E = \Delta E_i / E_i \approx 15\%$. 
The reduction-optimization approach

Incorporation into an iterative procedure

- The reduced mechanism should compensate for deleted reaction pathways
- The reaction and species subset must include the most active reactions
- A unique, huge reduction would lead to an unmanageable search space

→ a progressive reduction and optimization algorithm
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**Some results: Ethanol**

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<th>$n_s$</th>
<th>$n_r$</th>
<th>Details</th>
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</thead>
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<td>LLNL</td>
<td>58</td>
<td>383</td>
<td>Marinov, 1999</td>
</tr>
<tr>
<td>ROO</td>
<td>33</td>
<td>155</td>
<td>Reduced and optimized against Curran, 1992</td>
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</table>
Some results: Ethanol (2)

- Reduced, 33 species
- Red. + Optimized, 33 species
- LLNL, 58 species
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**Observations**

- **Strengths**
  - The performance of the reduced mechanism is similar to that of the detailed one at a reduced computational cost.
  - No need to dynamically change the mechanism's dimensions allows for a tailored approach to the IVP solution.

- **Weaknesses**
  - The initial conditions chosen as suitable validity landscape are defined by the user → problem-dependent.
  - The possibility of considering **transport-driven** cases (e.g., 1-D laminar flames) is limited by their computational demand (unviable for genetic optimization).
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Motivation

Chemical kinetics IVPs in adiabatic environments

- For an arbitrary reaction mechanism,
\[ \sum_{i=1}^{n_s} \nu_k^i M_i \rightleftharpoons \sum_{i=1}^{n_s} \nu''_k^i M_i, \quad k = 1, \ldots, n_r \]

- Mass conservation:
\[ \frac{dY_i}{dt} = \frac{W_i}{\rho} \sum_{k=1}^{n_r} (\nu''_{k,i} - \nu'_{k,i}) q_k(Y, T), \quad i = 1, \ldots, n_s \]

- Energy conservation:
\[ \frac{dT}{dt}(Y, T) = -\frac{1}{\bar{c}_v(Y, T)} \sum_{i=1}^{n_s} \left( \frac{U_i(T)}{W_i} \frac{dY_i}{dt}(Y, T) \right) \]

- Integrated with stiff ODE solvers (VODE, LSODE, RADAU5...)

- Only species and internal energy sources are linked to the CFD solver
Motivation

Why develop an analytical Jacobian formulation?

- Reduce scaling of the computational demand for the Jacobian matrix, that is of the order of $n_s^2$ when using finite differences;
- Reduce dense matrix storage requirements, also of the order of $n_s^2$;
- Scaling of the computational costs for matrix factorization, of the order of about $n_s^3$ if dense matrix algebra is employed;
- Exploitation of mechanism sparsity, which is significant even on small ($n_s < 50$) reaction mechanisms;
- Quadratic convergence of Newton’s iterative method;
- No need to introduce automatic differentiation tools.
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Sparsity patterns of three reaction mechanisms for n-heptane oxidation:

1. ERC n-heptane
   - nz = 412
   - 54.2% sparsity

2. LLNL n-heptane
   - nz = 3570
   - 86.2% sparsity

3. LLNL PRF
   - nz = 22551
   - 97.9% sparsity
Jacobian sparsity: pressure-dependent reactions

- Species involved in pressure-dependent reactions have **dense** lines
- Simplifying assumption: $\partial C / \partial Y_j \approx 0$
The benefits of adopting the approximate, sparser formulation increase quasi-logarithmically with the number of species.

Number of non-zero elements can be halved at average dimensions (i.e. $n_s \approx 100$).

Beneficial especially for its factorization.
Analytical Jacobian approach

Interpolation of temperature-dependent quantities

- Species thermodynamic potentials are polynomial functions of temperature
  - in JANAF format, e.g.
    \[ U_i = R_{mol} \left[ (a_i - 1) T + \frac{b_i}{2} T^2 + \frac{c_i}{3} T^3 + \frac{d_i}{4} T^4 + \frac{e_i}{5} T^5 + f_i \right] ; \]

- Equilibrium constant is an exponential function of the reaction’s free energy delta:
  - \[ K_{c_{eq,k}} (T) = \exp \left( -\Delta g_{k}^{0} \right) \left( \frac{p_{atm}}{R T} \right)^{\sum_{i=1}^{n_{s}} \left( \nu''_{k,i} - \nu'_{k,i} \right)} ; \]

- Reaction rates are exponentials too:
  - \[ \kappa_{f,k} (T) = A_{k} T^{b_k} \exp \left( -\frac{E_{k}}{R T} \right) . \]
Interpolation of temperature-dependent quantities

- Then, the interpolation errors can be very low e.g. at degree-4 interpolation
- Fixed temperature steps make storage simpler and data contiguous
- CPU time reduction of more than 1 order of magnitude with `-O3`
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Reference conditions

- 18 IVP cases, at $p_0 \in \{2.0; 20.0\} \text{ bar}$,
  $T_0 \in \{750; 1000; 1500\} \text{ K}$, $\lambda \in \{0.5; 1.0; 2.0\}$
- Integration intervals subdivided into 100 subcycles
Some results

- Almost linear speedup in comparison with a reference code that uses FD
- About one order of magnitude at typical CFD-tailored mechanism dimensions
60-degree engine sector grids with 16950, 42480 cells

Time spent for the fluid flow solution becomes almost negligible
Conclusions

- The computational efficiency allowed by such approach is significant
  - Speedups of about 2x also at almost skeletal mechanisms
  - About one order of magnitude for average mechanism sizes

- The combination of analytical formulation + \textit{sparse} matrix algebra is the key point

- Degree-4 interpolation helps drop the CPU times and does not affect the integrator performance

- Potential can be improved if the development of a tailored \textit{sparse stiff ODE solver} is addressed
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When/where does chemistry need to be solved in a computational domain?

Is it worthwhile to solve it in each single cell?

On which basis can reacting cells be regarded as 'similar' or 'different'?
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Concluding remarks
The idea of clustering cells with similar reactivity is not new

- Usually based on $\phi - T$ maps for engine calculations
- Search for similar cells based on proximity (neighbors, ROI) or on clustering ($k$-means)
- Chemistry is integrated for each cluster, and then conservatively redistributed

**Why develop a different approach?**

- The $\phi - T$ is problem-specific
- Performances can deteriorate when in presence of multiple fuels or large mechanisms with many intermediate species
The clustering space is defined as the $d$-dimensional cell positions in the state space (temperature, mass fractions):

- $x_{1,j} = T_j$;  
- $x_{2:d,j} = Y_{k,j}, \forall k \in S$

Normalized to a unity hyperbox
Bounding-box clustering

- Cluster initialisation as a **structured grid**
  - unique indexing
- Each point is contained in a bounding box of $2^d$ cluster centers
- Clusters have to **stay local** (**bounding-box-constrained** $k$-means algorithm)
- Reduced computational efforts than $k$-means: evaluate $2^d$ distances per point
Why have the clusters to ’stay local’?

- Bounding-box-constrained $k$-means vs. $k$-means w/ random cluster initialisation
- $n = 20$
Why have the clusters to ’stay local’?

- Bounding-box-constrained $k$-means vs. $k$-means w/ random cluster initialisation
- $n = 100$
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Some results

- Fiat 1.3l DI diesel engine, operated with multiple injections

Grid size: 24780 cells at BDC
Dimensionality: \( d = 5 \ (T, C_7 H_{16}, O_2, CO_2, HO_2, H_2O) \)
Cluster initialisation resolution: \( \varepsilon_T = 20K, \varepsilon_Y = 0.005 \)
Some results

- Local species distributions appear to be consistent
- Sensitivity analyses have shown no better accuracy at stricter resolutions
Conclusions

- Unsupervised cell clustering can be much beneficial (3-4x speedup), independent on the reaction mechanism used.
- The bounding-box approach allows:
  - distributed final arrangement of the clusters
  - reduced scaling with increasing number of clusters
  - it’s still proportional to the number of points
  - unsupervised approach to clustering in high-dimensional spaces

To be done

- Test the algorithm with huge grids / run it in parallel
- Assess its accuracy in presence of multiple or multi-component fuels
In order to correctly simulate a multidimensional problem that has reacting behaviour:

- Identification of the phenomena that need to be modeled by the reaction mechanism
- A correct choice of the mechanism dimensions to avoid unnecessary calculations
- Tailored approach to the solution is beneficial in case the mechanism doesn’t undergo on-the-fly reduction

- Much can still be achieved
Thank you!

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