# Reduction of Complexity in Combustion Thermochemistry

Americo Barbosa da Cunha Junior
Advisor: Luís Fernando Figueira da Silva

Department of Mechanical Engineering
Pontifícia Universidade Católica do Rio de Janeiro
americo.cunhajr@gmail.com

Master Dissertation
August 19, 2010

# Outline



# The Combustion Phenomena

- Physicochemical phenomena characterized by a sequence of chemical reactions which convert reagents in products of combustion
- Large amount of heat release due to the exothermic reactions
- Application in several industrial devices (e.g. gas turbines, process furnaces, etc.)
- The processes of energy production which are based on combustion represent a significant share of the global ($> 90\%$) and of the Brazilian ($> 80\%$) energy matrix.

Figure: Flame.

# Fundamental Challenges

- Difficulties in the modelling due to the complex nature of the combustion phenomena
- Necessity of modelling combustion/turbulence interactions since most of chemically reactive flows with practical applications operate in the turbulent regime
- A detailed kinetic mechanism is necessary to correctly describe the combustion thermochemistry, which makes the solution of a computational model an expensive task

# Computational Model for Chemically Reacting Flows

A computational model to predict the behavior of chemically reacting flows must include a detailed reaction mechanism to describe the combustion thermochemistry and solve the following equations:

- Continuity
- Momentum
- Energy
- Transport for the Chemical Species
- State Variables Relations
- Constitutive Relations

It is necessary to solve *(# species + 5)* partial differential equations.

# Objectives of this Dissertation

- Study in details the available techniques for reduction of reaction mechanisms and for efficient computation of the chemically reactive flows models
- Identify a technique which may allow one to accommodate detailed combustion thermochemistry in LES calculations
- Implement and evaluate the potential of the chosen technique

# Reduction Approaches

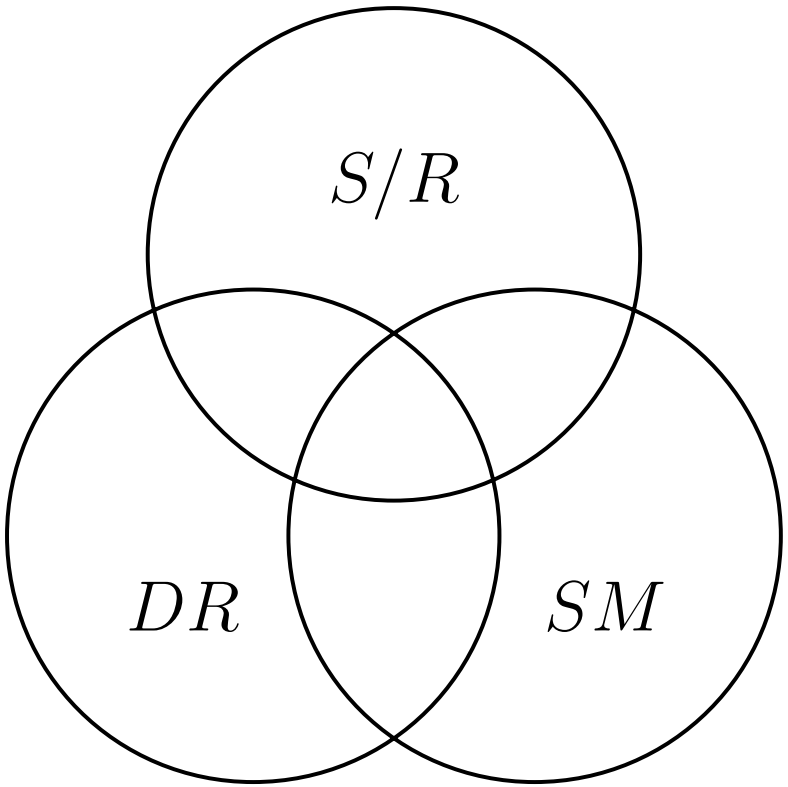


Figure: Classification of the different approaches.

The available techniques to reduce the complexity of combustion models can be classified in three categories:

- SM: Skeleton Mechanisms
- DR: Dimension Reduction
- S/R: Storage/Retrieval

Ren (2006)
Modelling Combustion with Detailed Chemistry
Ph.D. Thesis,
Cornell University

# Reduction to Skeleton Mechanisms

Eliminating species and reactions that have negligible influence on the combustion process results in the so-called skeletal mechanism.

- Sensitivity Analysis
- Proper Orthogonal Decomposition
- Directed Relation Graph

# SA - Sensitivity Analysis

- Determine the local behavior of the chemical system subjected to small disturbances.
- Small sensitivity to a perturbation is indicative that a specific reaction is redundant.
- Müller et al. (1992) reduce a mechanism for heptane with 171 species and 1011 reactions to one with 40 species and 79 reactions.

Müller et al. (1992) Global kinetics for n-heptane ignition at high pressures Symposium (International) on Combustion, v. 24 p. 777–784

# Dimension Reduction

Dimension Reduction approach is based on the assumption that a high-dimensional dynamical system can be parametrized in low-dimensional subset of the original phase space.

- Reaction Lumping
- Quasi-Steady-State Approximation
- Rate-Controlled Constrained Equilibrium
- Computational Singular Perturbation
- Intrinsic Low-Dimensional Manifold
- Proper Orthogonal Decomposition
- Invariant Constrained Equilibrium Edge Pre-image Curve

# QSSA - Quasi-Steady-State Approximation

- Assume steady state for some chemical species and partial equilibrium for some reactions which allow to replace an ODE by an algebraic equation in the system of governing equations.
- Hughes et al. (2009) reduce a n-heptane mechanism with 358 species and 2411 reactions to one with 81 species and 452 reactions.

Hughes et al. (2009)
The application of the QSSA via reaction lumping for the reduction of complex hydrocarbon oxidation mechanisms
Proceedings of the Combustion Institute, v. 32 p. 543–551

# ILDM - Intrinsic Low-Dimensional Manifold

- Automatically identify a low-dimensional manifold in which the chemical system can be described by a reduced number of parameters, which are stored in a multi-dimensional table to be recovered a posteriori via multi-linear interpolation.
- Maas and Pope (1992) create a 1-D and a 2-D manifolds that correctly parametrize a reaction mechanism with 13 species and 67 reactions in a spatially homogeneous, closed, adiabatic, isobaric reactor.

Maas and Pope (1992) Simplifying chemical kinetics: intrinsic low-dimensional manifolds in composition space. Combustion and Flame, v. 88 p. 239-264

# ICE-PIC - Invariant Constrained Equilibrium Edge Pre-image Curve

- Parametrize the reactive system in a low-dimensional manifold.
- Recover the original configuration using a pre-image curve technique.
- Ren et al. (2006) show that, for a fixed number of degrees of freedom, ICE-PIC is more accurate than QSSA and RCCE approaches.

Ren et al. (2006)
The invariant constrained equilibrium edge pre-image curve method for the dimension reduction of chemical kinetics
The Journal of Chemical Physics, v. 124 p. 114111

# Storage/Retrieval

Storage and Retrieval approach store the simulations results for a posteriori use (retrieval) via multilinear extrapolation.

- Look-Up Table
- Repro-Modelling
- Piece-Wise Reusable Implementation of Solution Mapping
- Artificial Neural Network
- *In Situ* Adaptive Tabulation

# ISAT - In Situ Adaptive Tabulation

- Requires less memory than a traditional look-up table method since the tabulation is done *in situ*
- It is extremely flexible to couple with other techniques
- Pope (1997) reports speed-ups of up to three orders of magnitude in the simulation time compared with the direct integration of the governing equations

Pope (1997)
Computationally efficient implementation of combustion chemistry using *in situ* adaptive tabulation.
Combustion Theory and Modelling, v.1 p. 41-63

# Hybrid Methodologies

Hybrid Methodologies are those which combine two or more existing approaches for reduction of chemical mechanisms. These sophisticated techniques envisage the best use of two or more well established techniques in order to achieve performance gains.

- ICE-PIC with ISAT

# Governing Equation

The thermodynamical state of a reactive mixture in a homogeneous transient reactor is determined by *composition* vector

$$\phi \equiv (h, p, Y_1, \cdots, Y_{n_s})^{\mathrm{T}},$$

which evolves according

$$\frac{d\phi}{dt} = -\mathbf{\Gamma}(t) + \mathbf{S}(\phi, t),$$

being

- $\mathbf{\Gamma}$ the rate of change due to micromixing transport
- $\mathbf{S}$ the rate of change due to the chemical reactions

# PMSR Characteristics

- Even number $n_p$ of particles
- Types of events: *inflow*, *outflow* and *pairing*,
- Time scales: $\tau_r$, $\tau_p$ and $\tau_m$
- System of evolution equations:

$$\frac{d\phi^{(j_1)}}{dt} = -\frac{\phi^{(j_1)} - \phi^{(j_2)}}{\tau_m} + \mathbf{S}(\phi^{(j_1)}, t),$$

$$\frac{d\phi^{(j_2)}}{dt} = -\frac{\phi^{(j_2)} - \phi^{(j_1)}}{\tau_m} + \mathbf{S}(\phi^{(j_2)}, t).$$

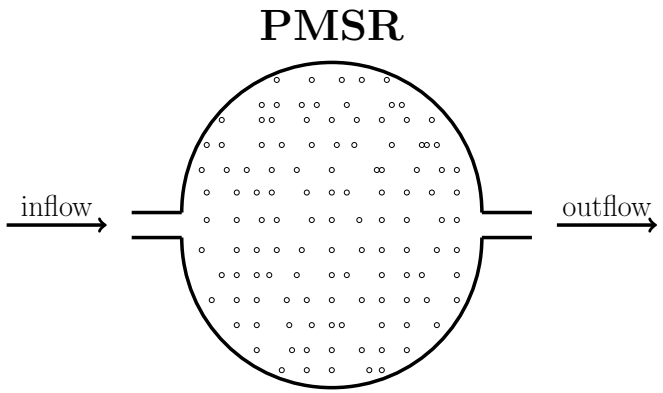


Figure: Sketch of a PMSR reactor.

# Reaction Mapping

The solution of the governing equation of the PMSR from an initial time $t_0$ to a time $t$ is given by

$$\mathbf{R}(\phi_0, t) \equiv \phi_0 - \int_{t_0}^{t} \mathbf{\Gamma}(t')dt' + \int_{t_0}^{t} \mathbf{S}(\phi, t')dt'.$$

which corresponds to a trajectory in *composition space*.

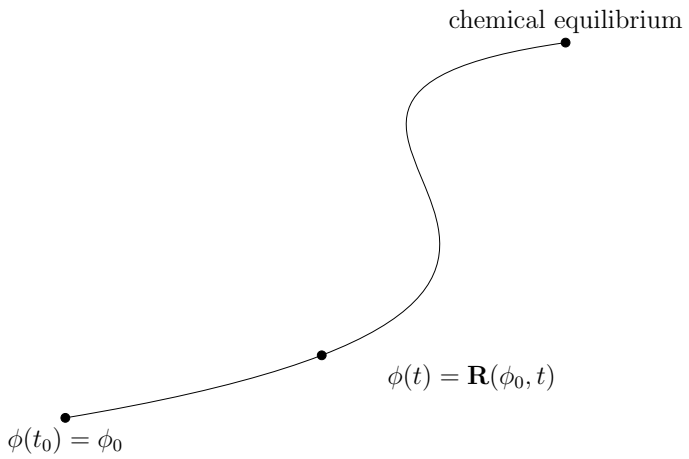


Figure: Sketch of a trajectory in composition space departing from $\phi_0$ until the chemical equilibrium.

# Operator Splitting Technique

The system of governing equations is splited into two subsystems: pure mixing and pure chemical reaction systems.

$$\frac{d\phi}{dt} = -\mathbf{\Gamma}(t) + \mathbf{S}(\phi, t) \begin{cases} \dfrac{d\phi}{dt} = -\mathbf{\Gamma}(t) \\ \\ \dfrac{d\phi}{dt} = \mathbf{S}(\phi, t) \end{cases}$$

The overall process of integration can be represented as

$$\phi(t) \overset{mixing}{\longrightarrow} \phi_{mix}(t + \Delta t) \overset{reaction}{\longrightarrow} \phi(t + \Delta t).$$

- analytical solution for pure mixing system (linear)
- BDF for pure chemical reaction system (nonlinear and stiff)

# Linearized Reaction Mapping

The *linear approximation* for the reaction mapping of $\phi$ around an initial composition $\phi_0$ is given by

$$\mathbf{R}^l(\phi, t) \equiv \mathbf{R}(\phi_0, t) + \mathbf{A}(\phi_0, t)\delta\phi,$$

where

$$\delta\phi \equiv \phi - \phi_0,$$

$$A_{ij}(\phi_0, t) \equiv \frac{\partial R_i}{\partial \phi_{0_j}}(\phi_0, t).$$

# Ellipsoid of Accuracy (EOA)

The *local error* of the linear approximation is defined as

$$\varepsilon \equiv ||\mathbf{R}(\phi, t) - \mathbf{R}^{l}(\phi, t)||.$$

The *region of accuracy* of the linear approximation is approximated by a hyper-dimensional ellipsoid centered at $\phi_0$

$$\delta\phi^{\mathsf{T}}\mathbf{L}\mathbf{L}^{\mathsf{T}}\delta\phi \leq \varepsilon_{tol}^{2},$$

where $\mathbf{L}$ is a Cholesky lower triangular matrix and $\varepsilon_{tol}$ is a positive *error tolerance* heuristically chosen.

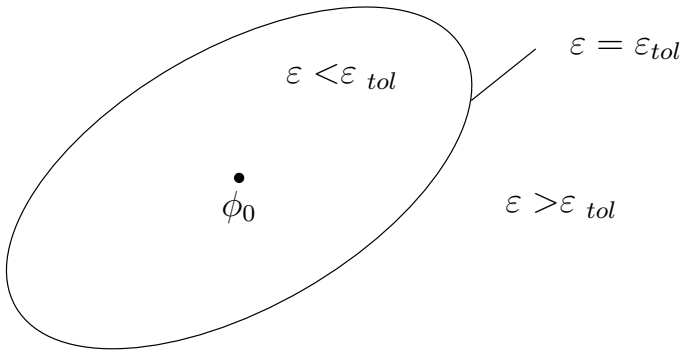


Figure: Sketch of the approximation for the region of accuracy.

# Growth Process of the EOA

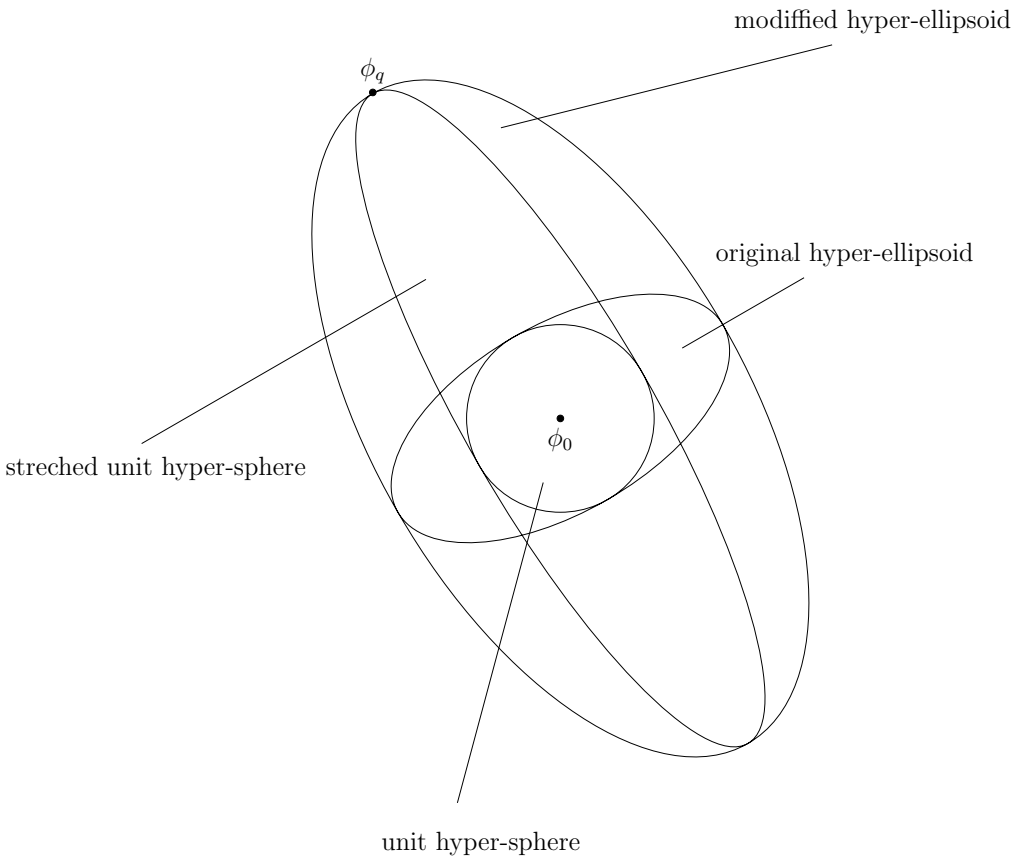


Figure: Hyper-ellipsoid growth process.

The adaptive step of ISAT algorithm has the following problem associated:

**Growth Problem**

Given a hyper-ellipsoid and a point $\phi_q$ outside of it, determine a new hyper-ellipsoid of minimum hyper-volume, which encloses $\phi_q$ and the original hyper-ellipsoid.

# Binary Search Tree

Some of the computed values are stored in a binary search tree. This process is known as *in situ* tabulation.

Each leaf stores:

- $\phi_0$
- $\mathbf{R}(\phi_0, t)$
- $\mathbf{A}(\phi_0, t)$
- $\mathbf{L}$

Each node stores:

- $\mathbf{v}$
- $a$

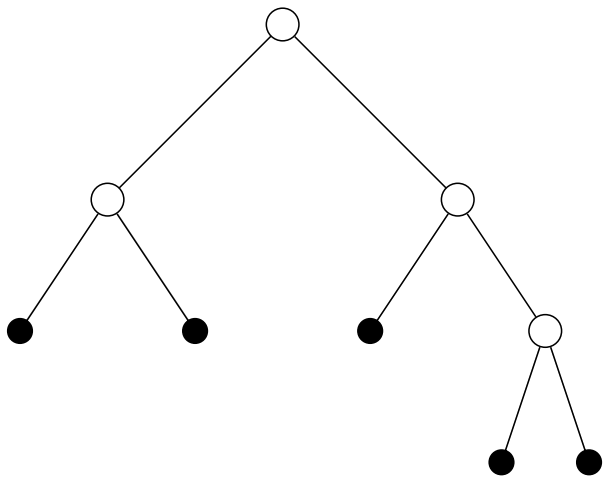

Figure: Sketch of the binary search tree.

# Addition and Search in the Binary Search Tree

Addition criterion:

- $\mathbf{v} \equiv \phi_q - \phi_0$
- $a \equiv \mathbf{v}^{\mathrm{T}} \left( \frac{\phi_q + \phi_0}{2} \right)$
- $\phi_0 \longrightarrow$ left
- $\phi_q \longrightarrow$ right

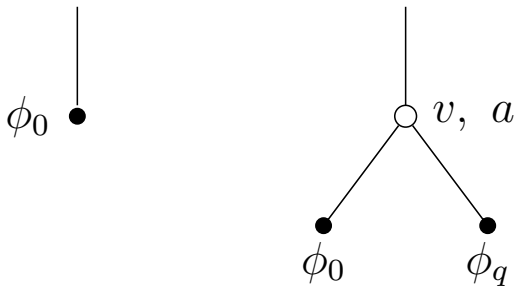


Figure: Sketch of the addition of a new node.

Search criterion:

- $\mathbf{v}^{\mathrm{T}}\phi > a \longrightarrow$ right
- $\mathbf{v}^{\mathrm{T}}\phi \leq a \longrightarrow$ left

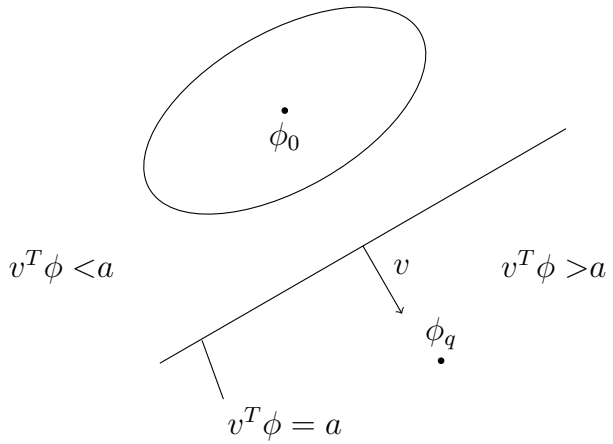


Figure: Sketch of cutting plane in relation to EOA position.

# ISAT Algorithm Overview

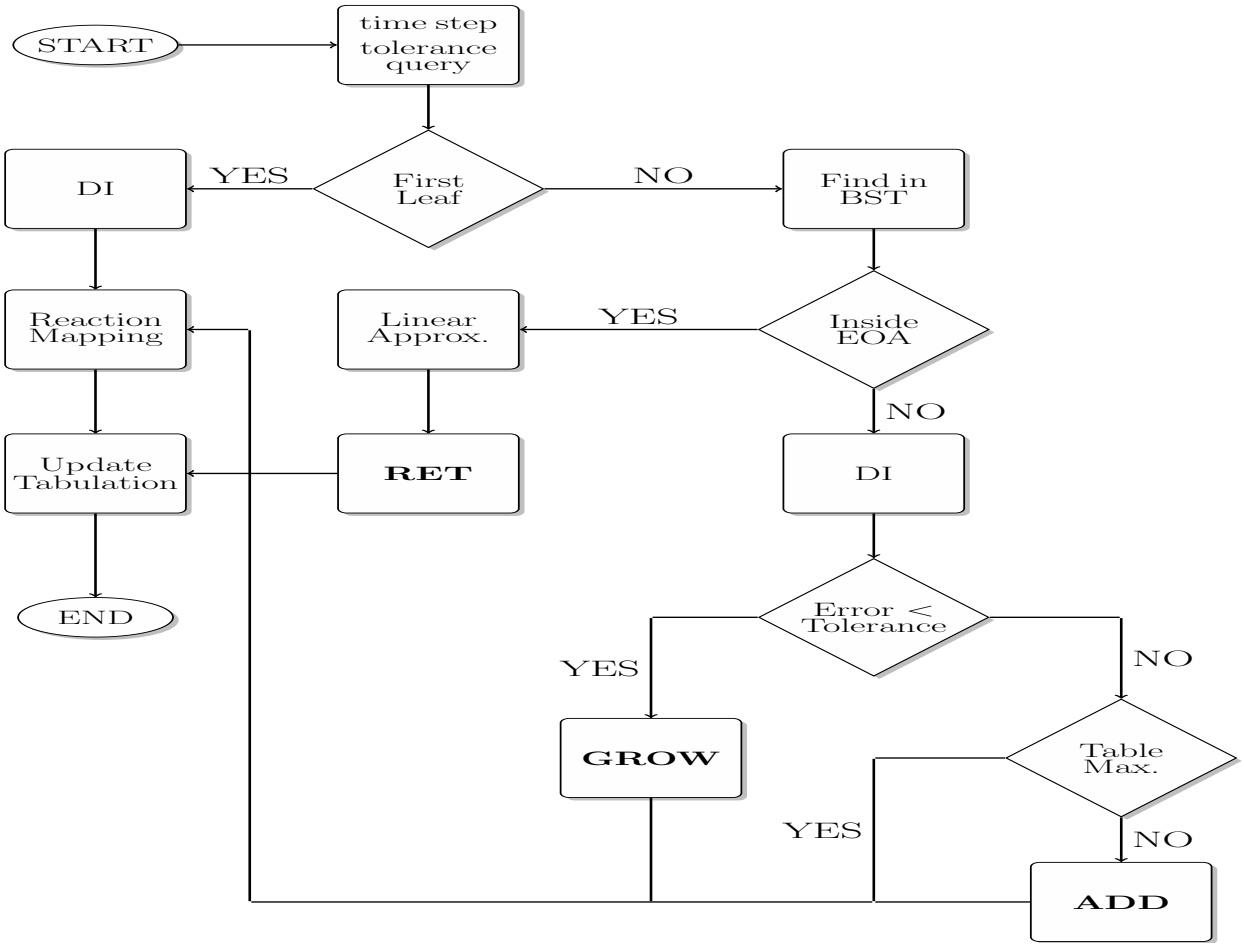


Figure: ISAT algorithm flowchart.

# Cases 1 and 2 (Simple Kinetics)

- binary search tree with a maximum of 50,000 entries
- $\Delta t = 10\ \mu\mathrm{s}$ and $\varepsilon_{tol} = 10^{-3}$
- Mixture of $CO/O_2$ (4 species and 3 reactions)

Gardiner Mechanism (2000)

- initial mixture: $\Phi = 0.7,\ T = 2948.5\ \mathrm{K}$ and $p = 1\ \mathrm{atm}$
- inflow mixture: $\Phi = 0.7,\ T = 300\ \mathrm{K}$ and $p = 1\ \mathrm{atm}$
- time scales (Case 1): $\tau_m/\tau_r = 1/2$ and $\tau_p/\tau_r = 1/2$
- time scales (Case 2): $\tau_m/\tau_r = 1/10$ and $\tau_p/\tau_r = 1/10$

# Case 3 (Complex Kinetics)

- binary search tree with a maximum of 60,000 entries
- $\Delta t = 0.1$ ms and $\varepsilon_{tol} = 10^{-3}$
- Mixture of $CH_4/Air$ (53 species and 325 reactions)

   GRI Mechanism 3.0 (1999)
- initial mixture: $\Phi = 1$, $T = 2100$ K and $p = 1$ atm
- inflow mixture: $\Phi = 1$, $T = 300$ K and $p = 1$ atm
- time scales: $\tau_m/\tau_r = 1/4$ and $\tau_p/\tau_r = 1/4$

# Evolution of $\langle T \rangle^*$

$$\tau^* \equiv \frac{t}{\tau_r} \qquad \langle T \rangle^* \equiv \frac{1}{n_p} \sum_{j=1}^{n_p} [T^*]^{(j)}$$

There is a good qualitative agreement between ISAT and DI results for $\langle T \rangle^*$.

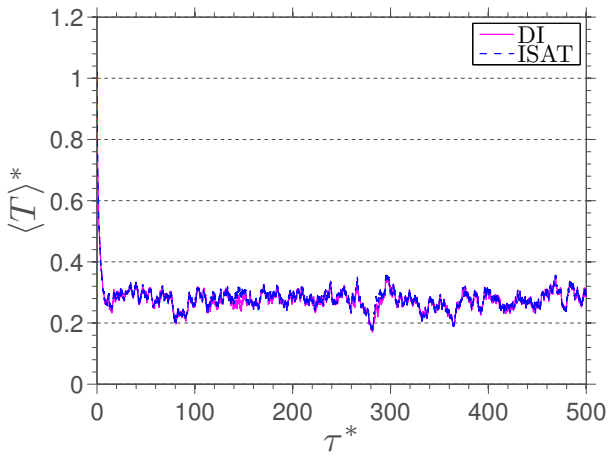


Figure: Case 1

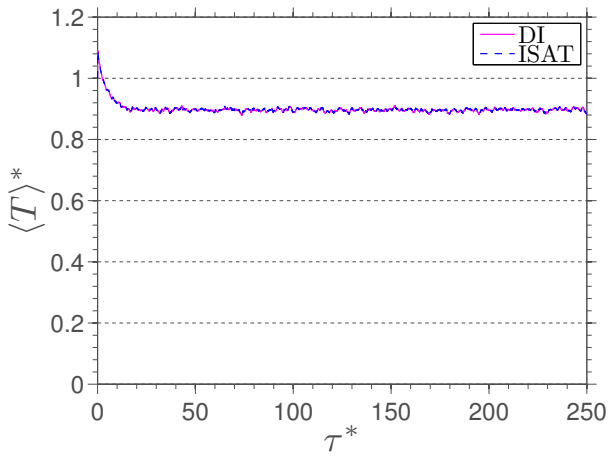


Figure: Case 2

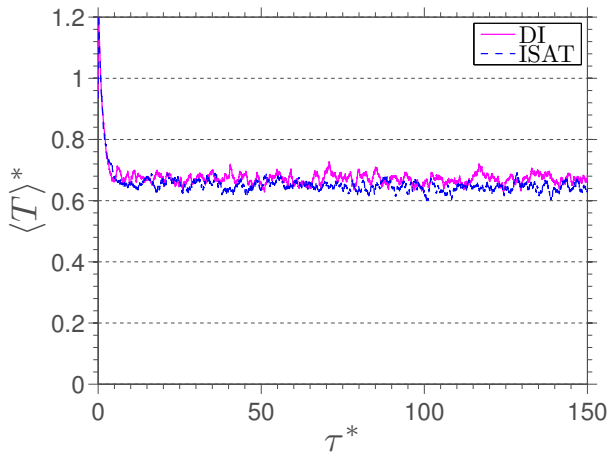


Figure: Case 3

# Evolution of Relative Local Error of $\langle T \rangle^*$

$$\varepsilon_{r,\langle T \rangle^*} \equiv \frac{|\langle T \rangle^*_{DI}(t) - \langle T \rangle^*_{ISAT}(t)|}{\langle T \rangle^*_{DI}(t)}$$

There is a large statistical variation due to the stochastic nature of the PMSR model.

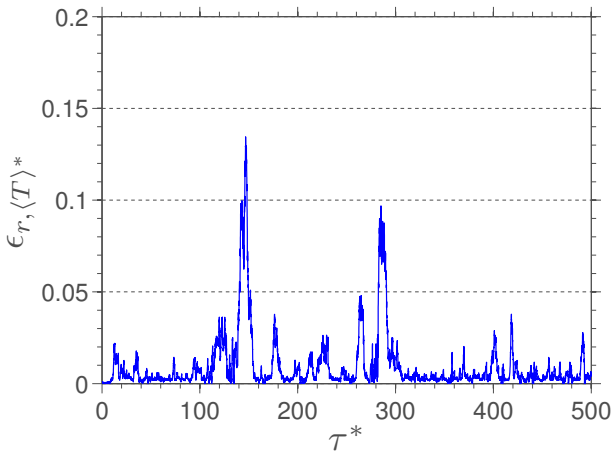


Figure: Case 1

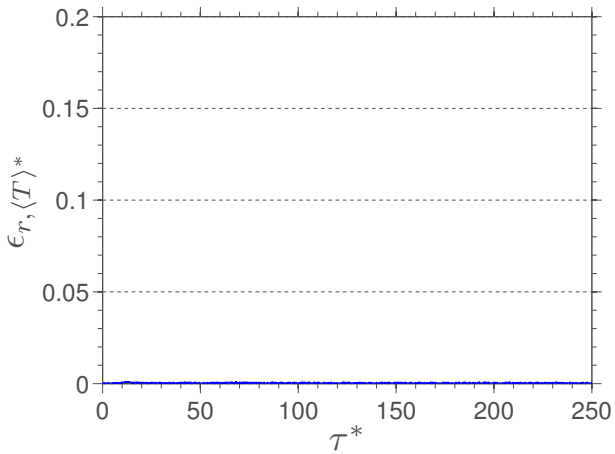


Figure: Case 2

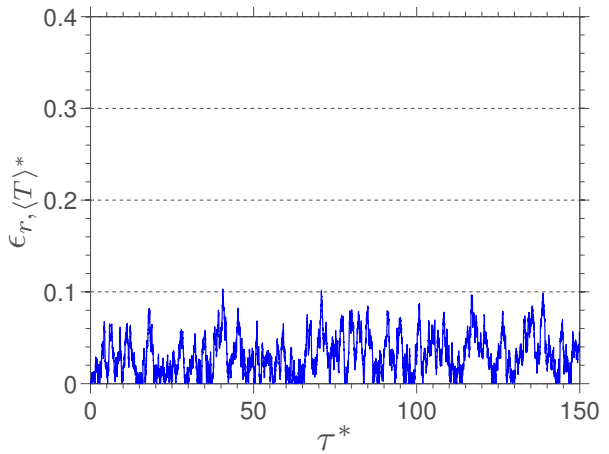


Figure: Case 3

# Absolute Global Error vs Error Tolerance

$$\varepsilon_g \equiv \frac{1}{\Delta\tau}\int_t^{t+\Delta\tau} ||\langle\phi\rangle(t')_{DI} - \langle\phi\rangle(t')_{ISAT}||dt'$$

For all values of $\varepsilon_{tol}$ tested, one can observe a small value for the absolute global error.

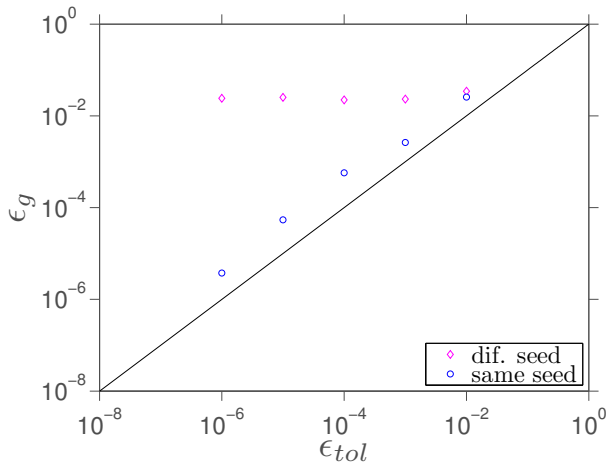


Figure: Case 1

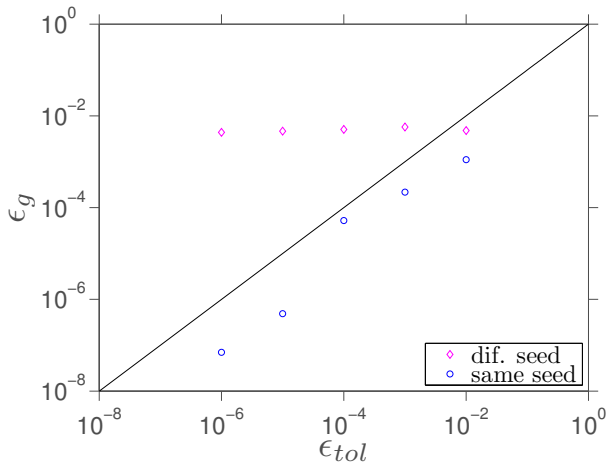


Figure: Case 2

# Evolution of ISAT Outputs

First, the binary tree is completely filled. Then # retrieves exceeds # additions, which ensures the efficiency of ISAT algorithm.

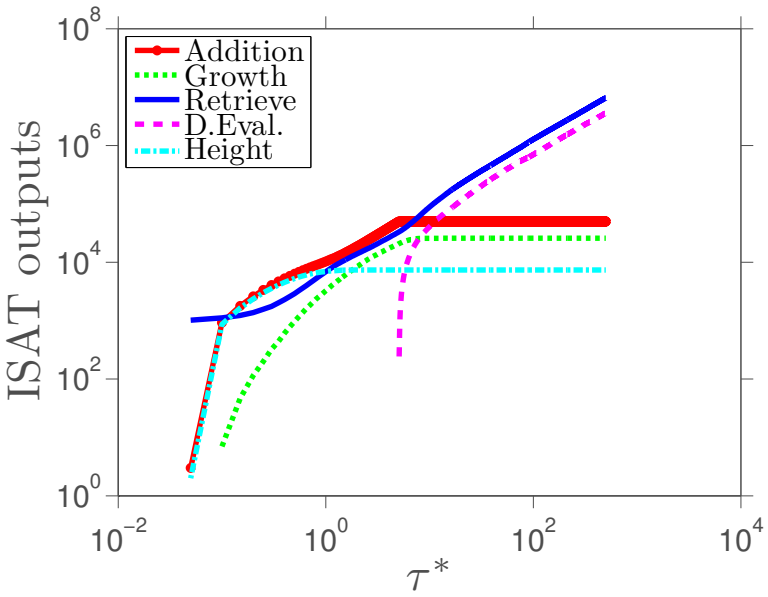


Figure: Case 1

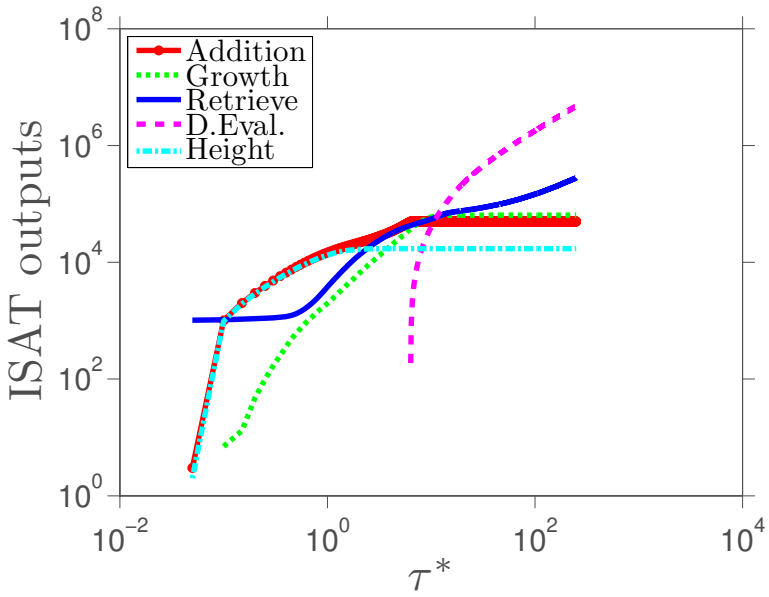


Figure: Case 2

# Computational Time Spent

Table: Comparison between the computational time spent by DI and ISAT and the corresponding speed-up factors.

| | time DI (ks) | time ISAT (ks) | speed-up | time saves |
|---|---|---|---|---|
| Case 1 | 4.0 | 2.3 | $\sim 1.7$ | $\sim 43\%$ |
| Case 2 | 2.0 | 2.1 | $\sim 1.0$ | $\sim 0\%$ |
| Case 3 | 689.3 | 454.5 | $\sim 1.5$ | $\sim 34\%$ |

# Computational Memory Spent

Table: Comparison between the total memory cost of ISAT using two different implementations.

| | `GSL` (Mbytes) | `ANSI C` (Mbytes) | difference |
|---|---|---|---|
| Case 1 | 40 | 22 | $\sim 45\%$ |
| Case 2 | 40 | 22 | $\sim 45\%$ |
| Case 3 | 3209 | 1744 | $\sim 47\%$ |

# Contributions of this Dissertation

- A detailed literature review about available techniques to produce reduced models for combustion thermochemistry
- Analysis of the accuracy, performance and memory usage of ISAT technique
- Concerning the accuracy, the ISAT shows good results from a global point of view and errors of up to 20% for the local error
- In terms of performance, the ISAT algorithm allows to reduce the computational time of the simulations in up to 80%
- Regarding the memory usage, the ISAT technique shows to be very demanding. In particular, the present implementation, spends up to 3.3 $\mathrm{Gbytes}$.

# Suggestions for Further Works

- Characterize ISAT technique in the simulation of reactors that use reaction mechanisms different than those used in this work
- Incorporate the improvements of ISAT algorithm proposed by

  Lu & Pope (2009)
  An improved algorithm for in situ adaptive tabulation
  Journal of Computational Physics,
  v. 228 p. 361–386
- Coupling of a detailed reaction mechanism, using the ISAT technique, with the hybrid LES/PDF model
- Validate the models studied in this work using data from carefully designed direct numerical simulations

# Thanks for your attention!