Karlsruher Institut für Technologie

Rich premixed hydrogen/air oscillatory flames: detailed modelling and model reduction

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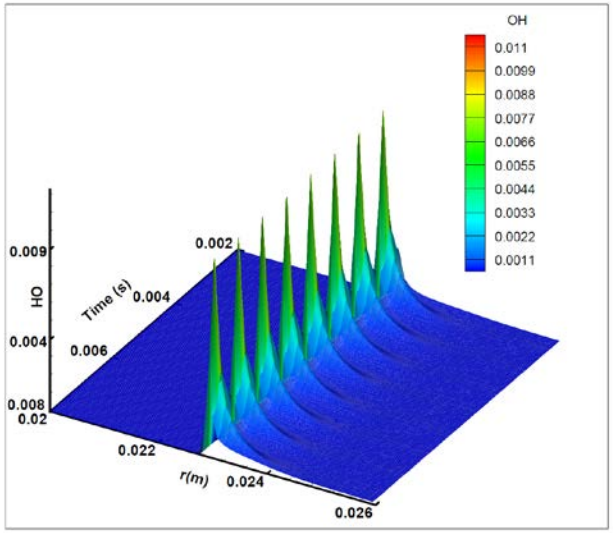
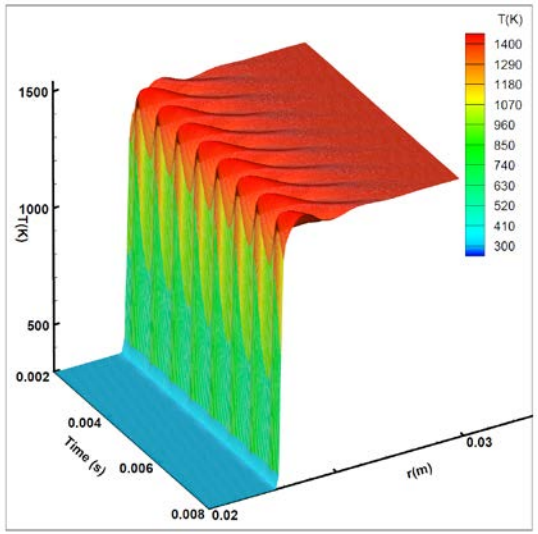




Motivation

- Diffusive-thermal instabilities indice appearance of different dissipative structures observed in experiments for various fuel-air mixtures mostly with Lewis numbers greater than one (see e.g., [1])

- Dissipative structures include pulsating axial, radial, drumhead modes, spirals, target, mixed mode and chaotic pulsations

- There are theoretical studies and quantitative modelling of such complex spatio-temporal regimes with the detailed reaction and diffusion mechanisms is a challenging task [2]

- Refs.:

[1] E. W. Christiansen, C. K. Law, C. J. Sung, Steady and pulsating propagation and extinction of rich Hydrogen/Air Flames at elevated pressures, Combustion and Flame 124 (2001) 35-49.

[2] G. Goyal, U. Maas, J. Warnatz, Simulation of the Behavior of rich hydrogen-air flames near the flammability limit, Combust. Sci. and Tech. 105 (1995) 183-193.

Motivation

- It has recently been shown [3] that even for a one-dimensional model characteristics of pulsating regimes predicted by using different reaction mechanisms are sensitive to the choice of mechanism and may differ significantly especially for elevated pressures

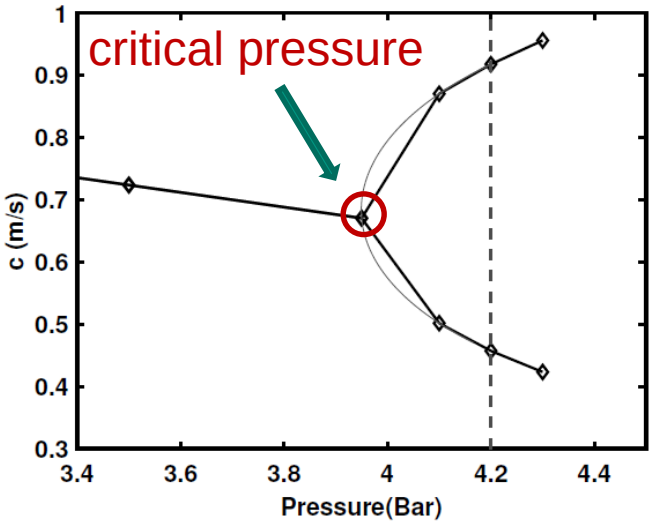
- This makes the phenomenon extremely important and can be considered for further development and validation of detailed and reduced reaction models. Such that the latter can be efficiently applied for numerical computational studies

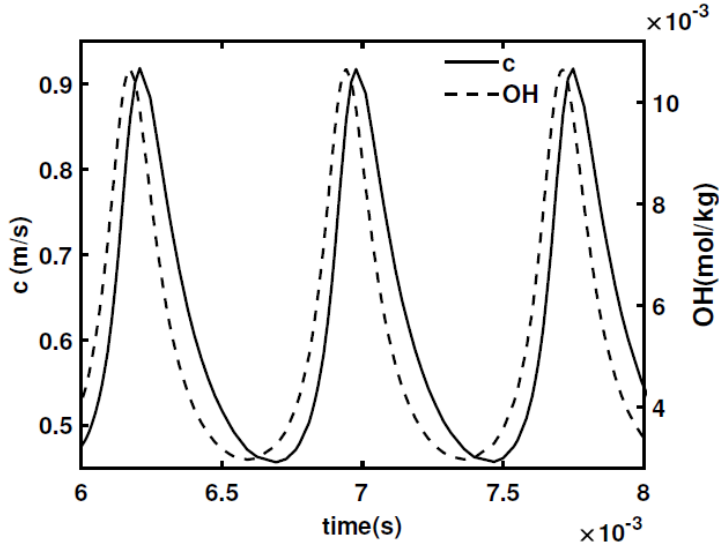
- Thus, one dimensional models to describe the onset of pulsating instabilities and influence of detailed kinetic models are in the focus of the current study

- Ref.: [3] A. Korsakova, V. Gubernov, A. Kolobov, V. Bykov, U. Maas, Stability of rich laminar hydrogen-air flames in a model with de-tailed transport and kinetic mechanisms, Combustion and Flame 163 (2016) 478-486.

Phenomenon

- The Warnatz mechanism (9 species, 18 reactions) for hydrogen/air system with Φ = 5, T = 298 K and variable pressure is considered
- The mixture composition is kept fixed at Φ = 5 and the pressure is increased from the ambient value until the pulsating solutions emerges





- Oscillatory flame front behavior: on the left the bifurcation diagram is shown in the pressure and laminar flame velocity C plane, on the right - evolution of flame speed with OH in specific mole numbers for p = 4.2 bar

Phenomenon

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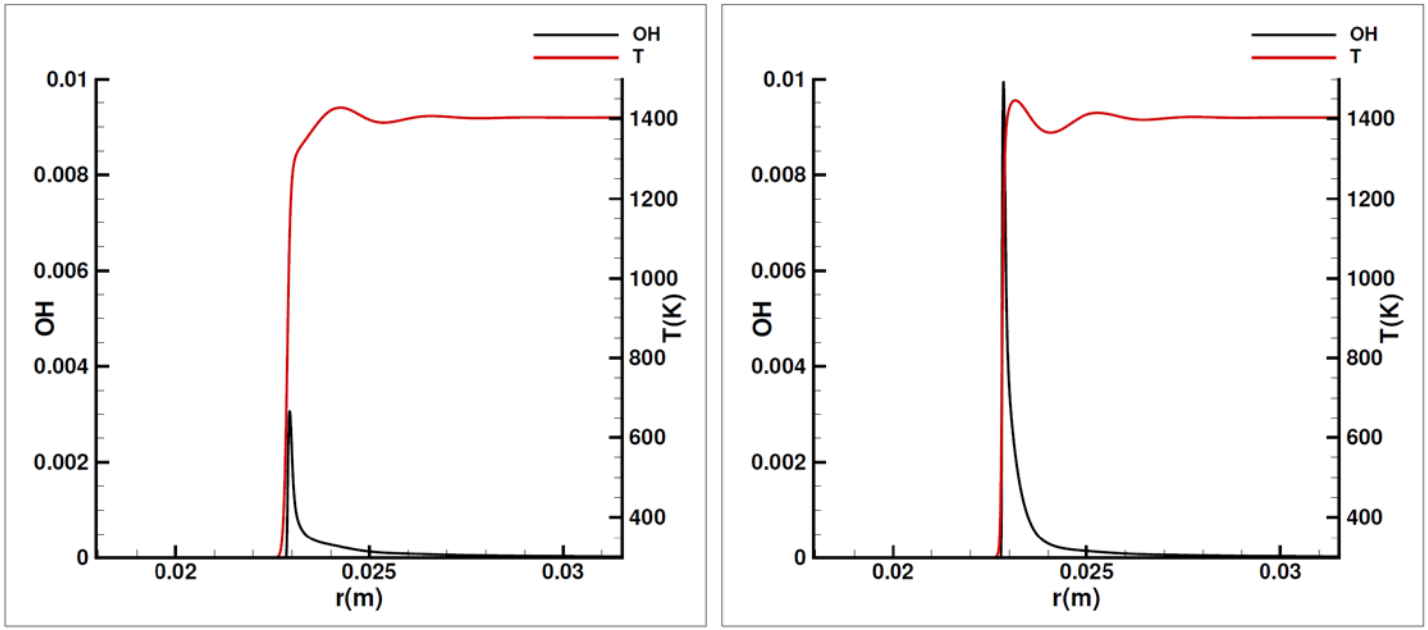

- Profiles of the oscillating solution in the physical coordinates at minimum and maximum amplitude of the mass flow rate

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Modelling - detailed reaction mechanisms

- Properties of mechanisms of detailed chemical kinetics
  - many species are involved in accurate modeling of auto-ignition problem

  participating in
  - “too many” (up to several thousand) elementary reactions with little known about their rate constants

  leading to

  - stiffness of system of governing equations and high dimensionality of thermo-kinetic state of combustion system

$$\boldsymbol{\psi} = \left(h, p, \frac{w_1}{M_1}, \frac{w_1}{M_1}, \cdots, \frac{w_{n_S}}{M_{n_S}} \right)^T \qquad \frac{d\boldsymbol{\psi}}{dt} = \mathbf{F}(\boldsymbol{\psi}), \qquad \boldsymbol{\psi} \in R^n$$

$$n_s >> 1$$

Which mechanism to choose?

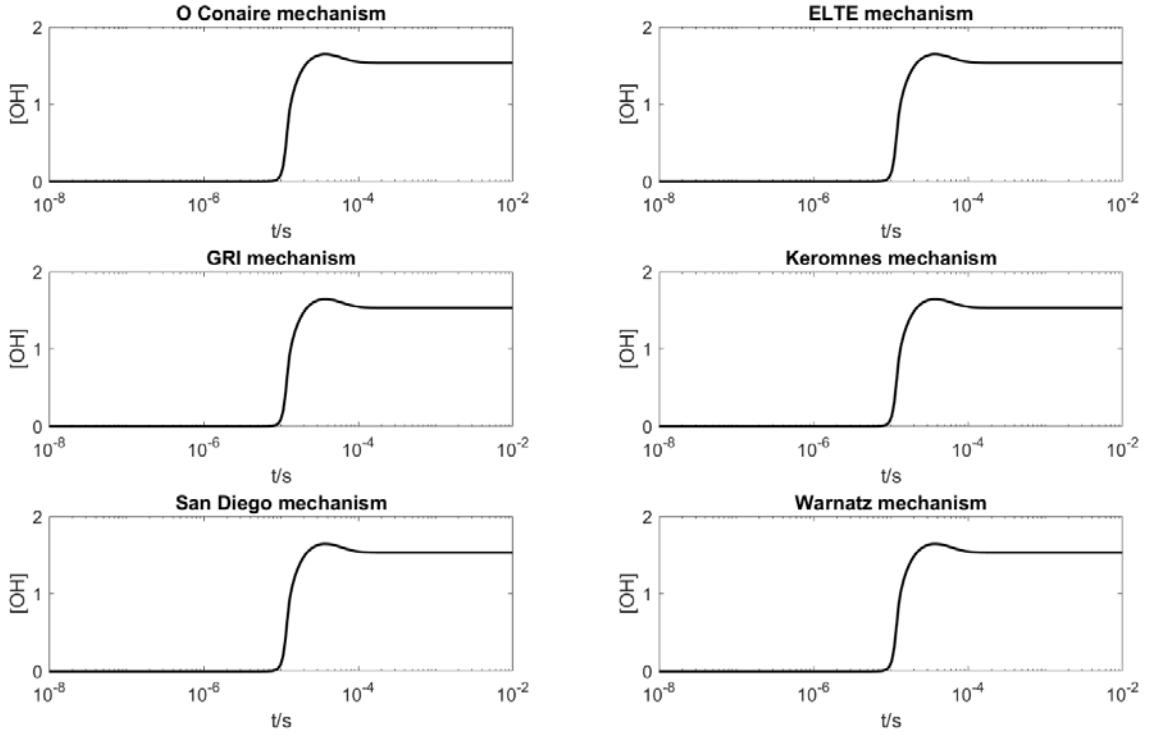
- Additionally, many different mechanisms have been developed for the same combustion system
  - For instance, about ~20 mechanisms exist for hydrogen/oxygen system
    - Ref.: Varga et al., Proc. Comb. Inst. 35 (2015) 589–596
  - Reaction rate parameters for e.g. for $H + O_2 = OH + O$ (units: cm^3, mol, s, kJ, K)

Mechanismons rate	Arrhenius Parameter		
	A	n	E
Ó Conaire	$1.915 \cdot 10^{14}$	0.00	68.831
ELTE	$1.378 \cdot 10^{13}$	0.24	60.403
GRI 3.0	$2.650 \cdot 10^{16}$	-0.67	71.301
Keromnes	$1.040 \cdot 10^{14}$	0.00	63.999
San Diego	$3.520 \cdot 10^{16}$	-0.70	71.468
Warnatz	$2.000 \cdot 10^{14}$	0.00	70.300

- Notable differences in reaction rate parameters, do they all describe the combustion system dynamics satisfactory?

Homogeneous system, ignition problem

- Simulated time histories (homogeneous reactor), $T_0 = 1500$ K, p = 1 bar, stoichiometric mixture



- In spite of differences, results obtained by different mechanisms agree quite well!?

Objectives

- Investigate sensitivity of the oscillatory flame front dynamics with respect to several well established detailed mechanisms
- Implement and verify reduced models based on the system decomposition approach (GQL)

Mechanisms	Comments	
	References	Changes
Ó Conaire	Ó Conaire et al., Int. J. Chem. Kin. 36 (2004) 603–622.	no changes
ELTE	Varga et al., Proc. Comb. Inst. 35 (2015) 589–596	OH(hv) was eliminated
GRI 3.0	Smith et al., Gas Research Institute, Des Plaines, IL, accessed Aug 21 (1999) 2017	species with C atoms were eliminated
Keromnes	K´eromn`es et al., Comb. Flame 160 (2013) 995–1011	C-... and OH(hv) were eliminated
San Diego	S´anchez and Williams, Prog. Energy Combust. Sci. 41 (2014) 1-55	no changes
Warnatz	U. Maas, J. Warnatz, Comb. Flame 74 (1988) 53–69	no changes

- Same thermo-chemical system state space (with 9 species) is analysed by characteristic time scales and using invariant subspaces of relatively fast and slow motions

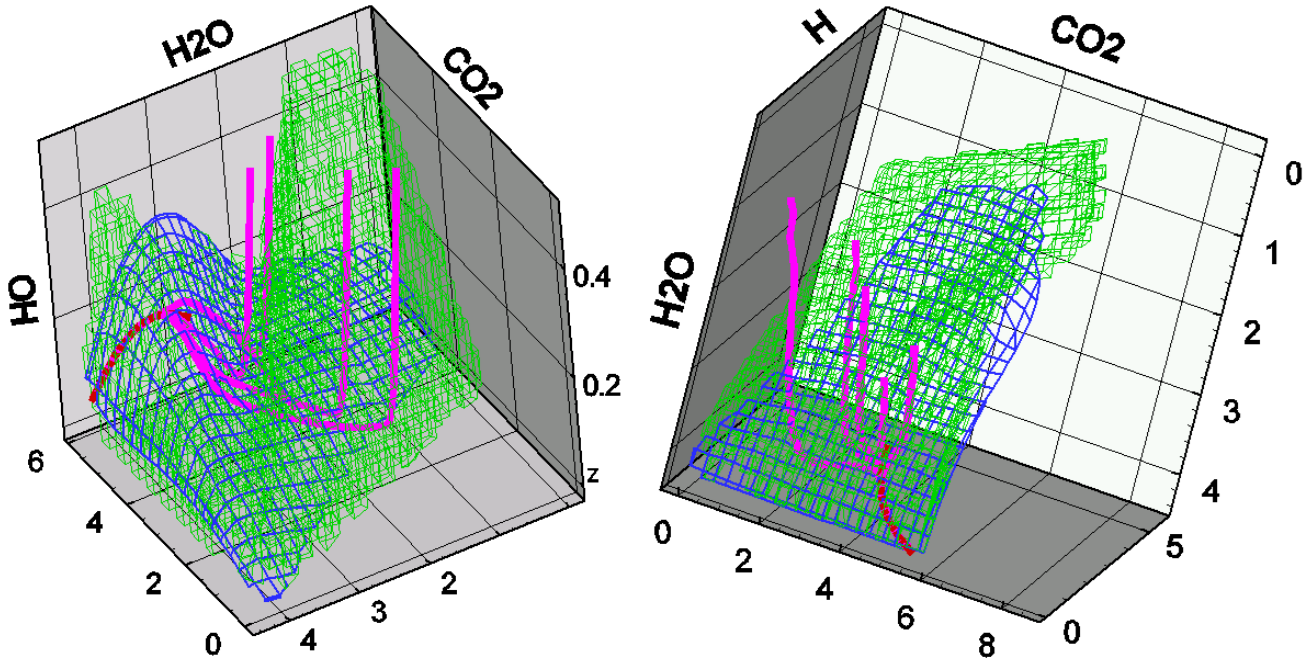
Methodology: relative fast and slow motions

- Reacting system state space is considered and invariant properties of the system are investigated

- Painting by Luigi Catani: Metaphor for system dynamics

Methodology: invariant manifolds of slow dynamics

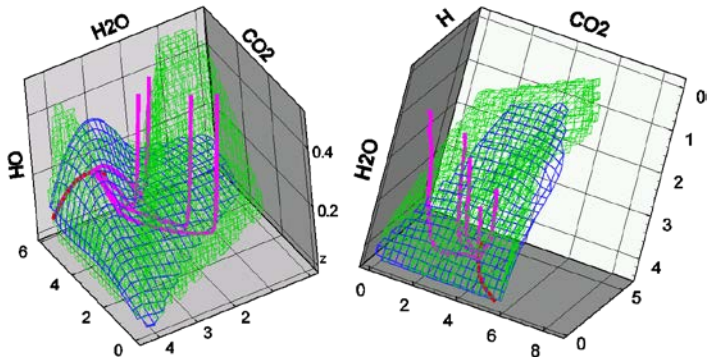
- Characteristic time scale analysis of system is implemented
- Global-Quasi-Linearization (GQL) approach is applied to study relatively fast and slow system dynamics



- Example: CH4 /air combustion reaction trajectories (red, magenta) together with 2D and 3D (blue and green) slow manifolds: Fast motion towards the manifold, slower motion on manifolds

System dynamics decomposition in the system state space: manifold of slow motions

- System solution trajectory can be decomposed into fast and slow stages



$$F(\boldsymbol{\psi}) \cong T \cdot \boldsymbol{\psi}$$

$$\lambda_i < \lambda_{i+1}$$

$$T = \left[Z_1 Z_2 \ldots Z_n\right] \begin{bmatrix} \lambda_1 & 0 & \ldots & 0 \\ 0 & \lambda_2 & \ldots & 0 \\ \ldots & \ldots & \ldots & \ldots \\ 0 & 0 & \ldots & \lambda_n \end{bmatrix} \begin{bmatrix} \tilde{Z}_1 \\ \tilde{Z}_2 \\ \ldots \\ \tilde{Z}_n \end{bmatrix}$$

- Matrix *T* is a global linearization of the source term $F(\boldsymbol{\psi})$
- Eigenvalue decomposition delivers
  - characteristic times ($1/\lambda_i$) and
  - slow relevant to (λ_1 , … , λ_m) and fast subspaces (spanned by Z_i) defines

$$\tilde{Z}_f F(\psi) \approx 0$$

  - Ref.: Bykov, Gol'dshtein, Maas, CTM, 12(2), 389 – 405 (2008).

Implementation: Implicitly decomposed form

- Dynamics of a reacting system can be decomposed and basis is identified by the GQL linearization and employed to define a mass matrix

$$T = \left[Z_1\, Z_2 \ldots Z_n\right] \begin{bmatrix} \lambda_1 & 0 & \ldots & 0 \\ 0 & \lambda_2 & \ldots & 0 \\ \ldots & \ldots & \ldots & \ldots \\ 0 & 0 & \ldots & \lambda_n \end{bmatrix} \begin{bmatrix} \tilde{Z}_1 \\ \tilde{Z}_2 \\ \ldots \\ \tilde{Z}_n \end{bmatrix} \Rightarrow Q_s = \left(Z_s\; Z_f\right) \cdot \begin{pmatrix} \mathrm{I}_s & \mathbf{0} \\ \mathbf{0} & \epsilon\,\mathrm{I}_f \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s \\ \tilde{Z}_f \end{pmatrix}$$

- System dynamics can now be decoupled and considered only on the “slow” manifold (ε → 0)

$$\tilde{Z}_s \frac{d\psi}{dt} = \tilde{Z}_s F(\psi)$$
$$0 = \tilde{Z}_f F(\psi) \Rightarrow Q_s \frac{d\psi}{dt} = F(\psi)$$

- The system is integrated on the slow manifold!

Computation: numerical integration

- Dynamics of H2/N2/O2 distributed system can be modelled in a wide range of system parameters - initial temperatures, pressures, equivalence ratios
- The suggested implementation treats the manifold in an implicit way both for reaction source term as well as for diffusion and advection

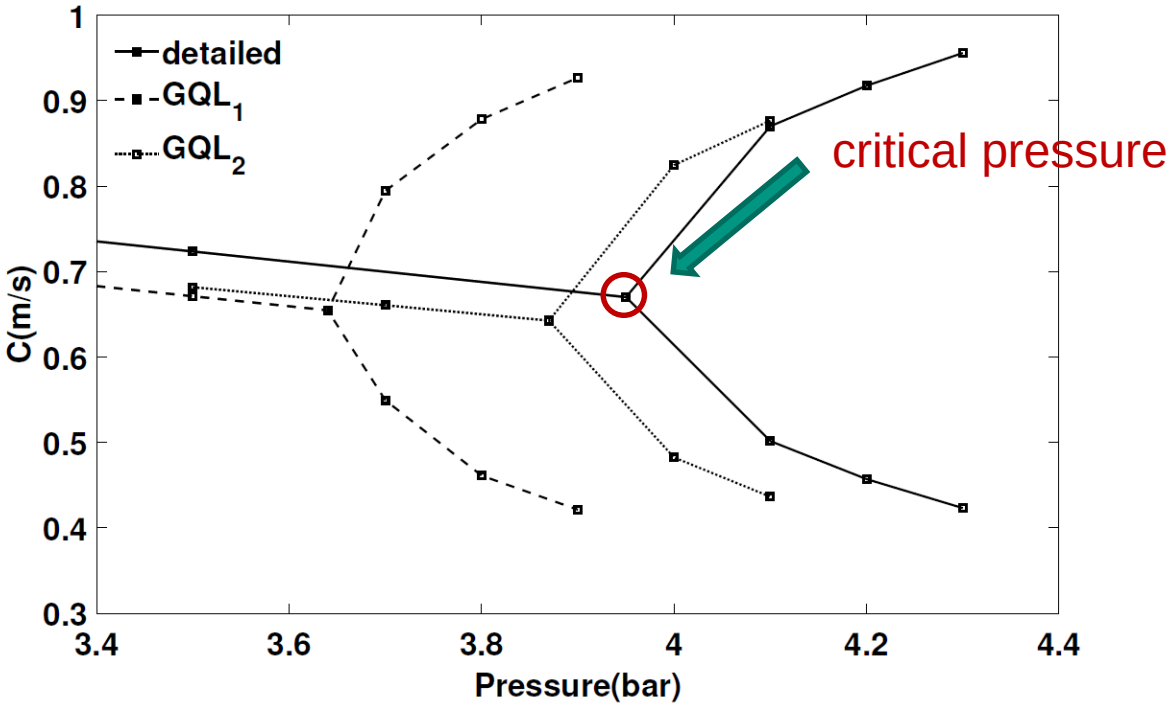
$$\mathrm{Q}_s = \left(Z_s \; Z_f\right) \cdot \begin{pmatrix} \mathrm{I}_s & \mathbf{0} \\ \mathbf{0} & \epsilon\,\mathrm{I}_f \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s \\ \tilde{Z}_f \end{pmatrix}$$

$$\mathrm{Q}_s \cdot \frac{\partial \psi}{\partial t} = F(\psi) - \mathrm{Q}_s \cdot v\,\mathrm{grad}\psi + \mathrm{Q}_s \cdot \frac{1}{\rho}\mathrm{div}\left(D\;\mathrm{grad}\psi\right).$$

- Both detailed and reduced models are integrated with DAEs system integrator LIMEX
- This framework allows us to compare any kind of implicitly defined reduced models

Onset of pulsations: performance of the linearization basis

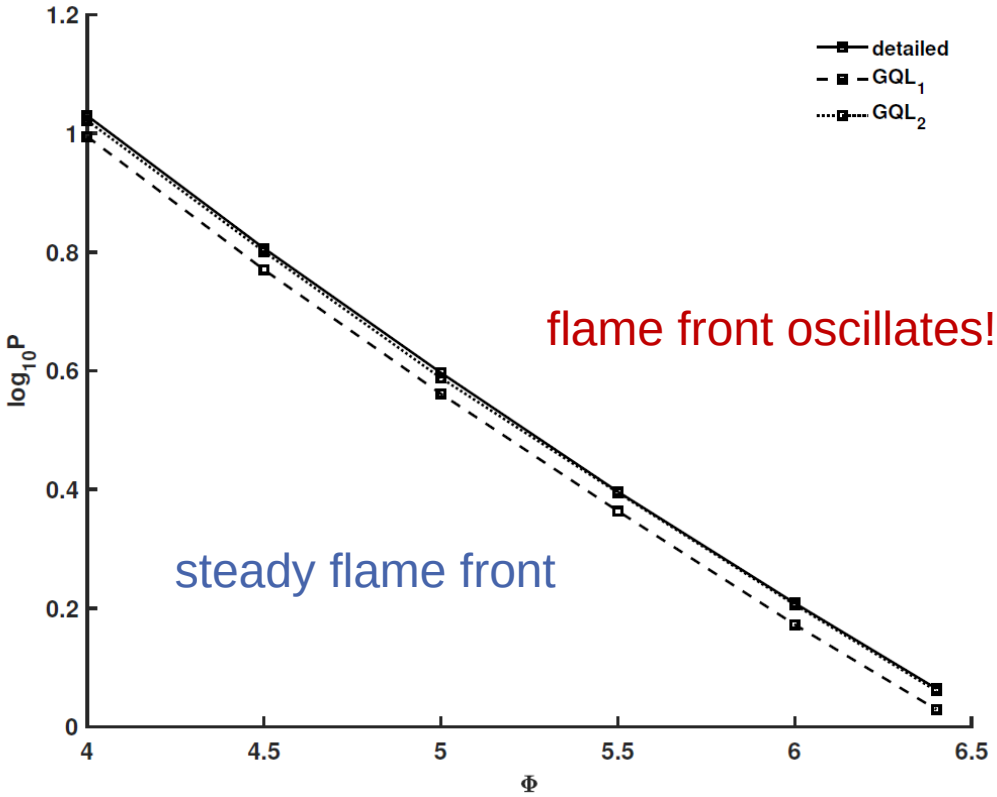
- Warnatz's and GQL reduced model integrations are compared. Here two linearization bases are used for Φ = 1 (GQL1) and Φ = 5 (GQL2) mixture compositions



- Bifurcation diagram of H2 / O2 / N2 combustion system in the pressure and flame velocity C plane for the Warnatz model (Pressure axis scaled to highlight differences)

Implementation results – neutral stability

- GQL with dimension m=4 was computed based on the Warnatz mechanism, for $T_0 = 1500$ K, p=1 bar, $\Phi = 1$ and $\Phi = 5$

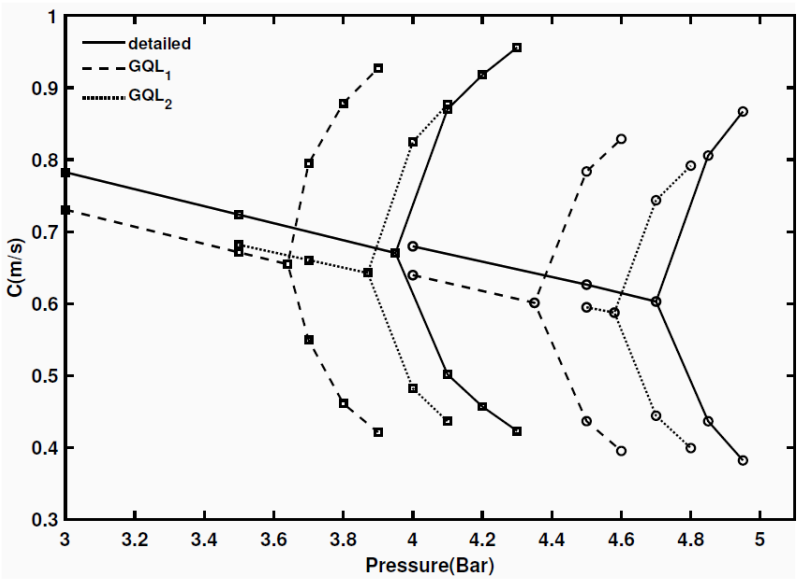


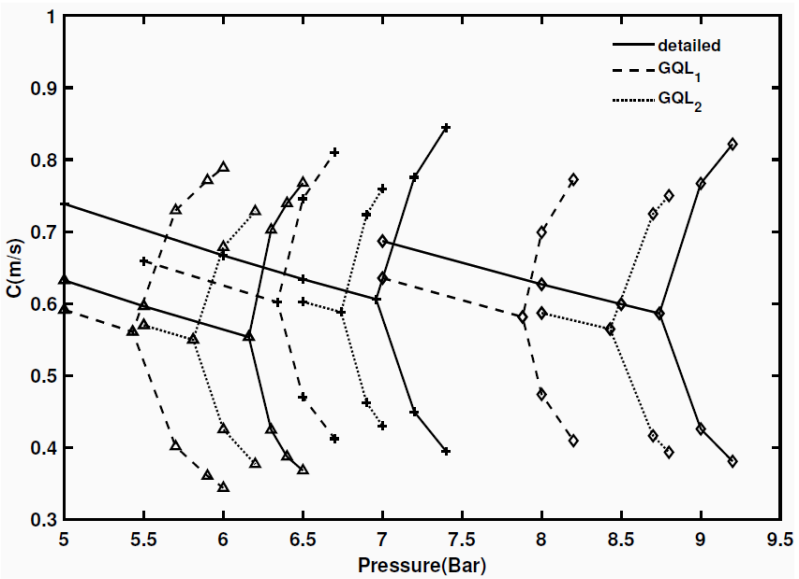
- Neutral stability boundary for Warnatz mechanism is shown in the log10 P vs Φ plane with results of GQL1 and GQL2 linearizations

Mechanisms comparison

- Bifurcation plots for detailed and reduced model (with the same mass matrix) integrations are compared

Mechanism	Ref.	Comment	symbol/Fig. 7
O'Conaire	(Ó Conaire et al., 2004)	no change needed	+
ELTE	(Varga et al., 2015)	OH($h\nu$) is eliminated	○
Keromnes	(Kéromnès et al., 2013)	C-.., OH($h\nu$) removed	◇
San Diego	(Sánchez and Williams, 2014)	no change needed	△
Warnatz	(Warnatz et al., 2012)	no change needed	□



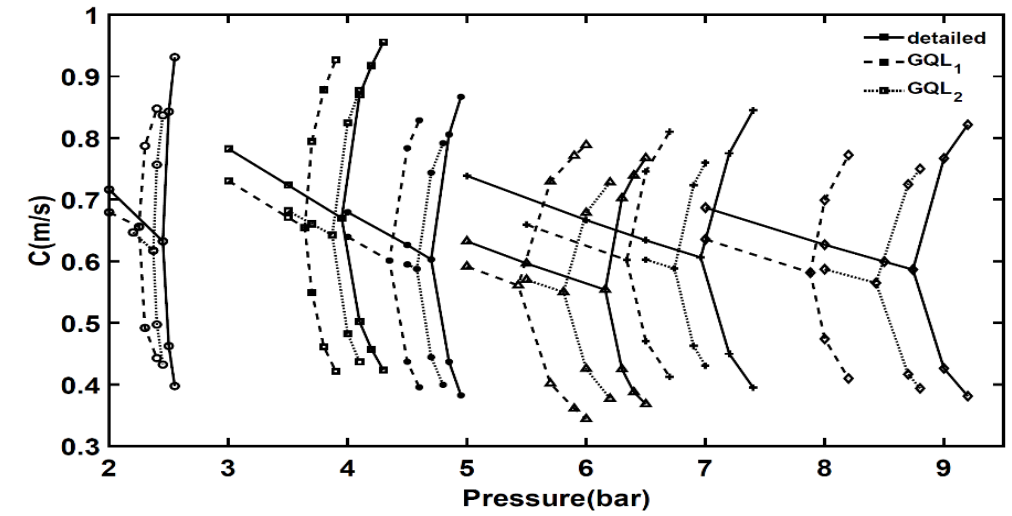


- Bifurcation diagrams of H2 /O2/N2 combustion system in the pressure and flame velocity C plane with all detailed and reduced models for the onset of flame front pulsations

Error assessment

- Critical pressure with errors of different detailed mechanisms using two linearization bases calculated on the basis of the Warnatz det. model

Mechanism	Warnatz	SanDiego	Keromnes	O'Conaire	ELTE
Critical Pressure, P_h	3.95	6.15	8.74	6.95	4.73
Deviation, GQL_1,%	7.89	11.81	9.87	8.90	8.26
Deviation, GQL_2,%	2.07	5.64	3.58	3.15	3.18



- Ref.: V. Bykov, C. Yu, V. Goldshtein, U. Maas, Model reduction and mechanism comparison of hydrogen/oxygen auto-ignition, Proceedings of the Combustion Institute 37 (2019) 781-787

Conclusions

- In this study an extension of the GQL reduced model to treat rich hydrogen-air flames under the critical conditions near the onset of the thermal-diffusion instability was suggested.

- It was shown how the 4D GQL reduced chemistry slow manifold designed for a homogeneous system performs to address hydrogen/air oscillatory flames.

- The manifold was constructed for an auto-ignition problem for one mechanism, namely, for the War-natz mechanism and for two sets of initial conditions and system parameters, namely, $\Phi = 1$ and $\Phi = 5$ with $T = 1800$ K and $p = 1$ bar constant.

- These GQL based reduced models then were applied to study critical values for the onset of pulsating flames and characteristics of the oscillatory flame fronts with a number of well established and validated hydrogen combustion mechanisms. For all mechanisms a scatter of 100% for the critical pressure was reported.

Conclusions

- The 4D GQL reduced chemistry optimized for a stoichiometric case predicts the critical pressure for the onset of instabilities very accurately (about 10% relative error). The optimized GQL model for the same equivalence ratio, where the onset was observed improves the situation (relative error of order of 5%).

- This observation is very important and signifies that the GQL reduced chemistry is capable of accurately predicting near limit behavior in rich hydrogen/air systems in a wide range of system parameters that shows good extrapolation power of the approach.

- The demonstrated successful applicability of this method to describe on a uniform basis different flame phenomena such as ignition, flame propagation, and the onset of flame instabilities for an unprecedented large scale of mixture compositions and pressures opens up a new perspective to develop new and effective approaches to simulate various types of combustion configurations. We look forward to test this method in our future work on different types of combustion systems and fuels.

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