Manifold Methods for Energetic Materials

by

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Energetic Materials

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Outline

- Motivation
- Description of ILDM technique
- Results for unsteady $H_2 - O_2$ detonation
- Preliminary Results for HMX gas phase combustion
- Conclusions

Some Important Questions

- Do we have resolved, accurate solutions for HMX combustion?
- How can ILDM improve the calculation of HMX combustion?
- How can ILDM, derived for well-stirred systems, be corrected to account for convection and diffusion?

Preliminary HMX Calculations with Convection Diffusion

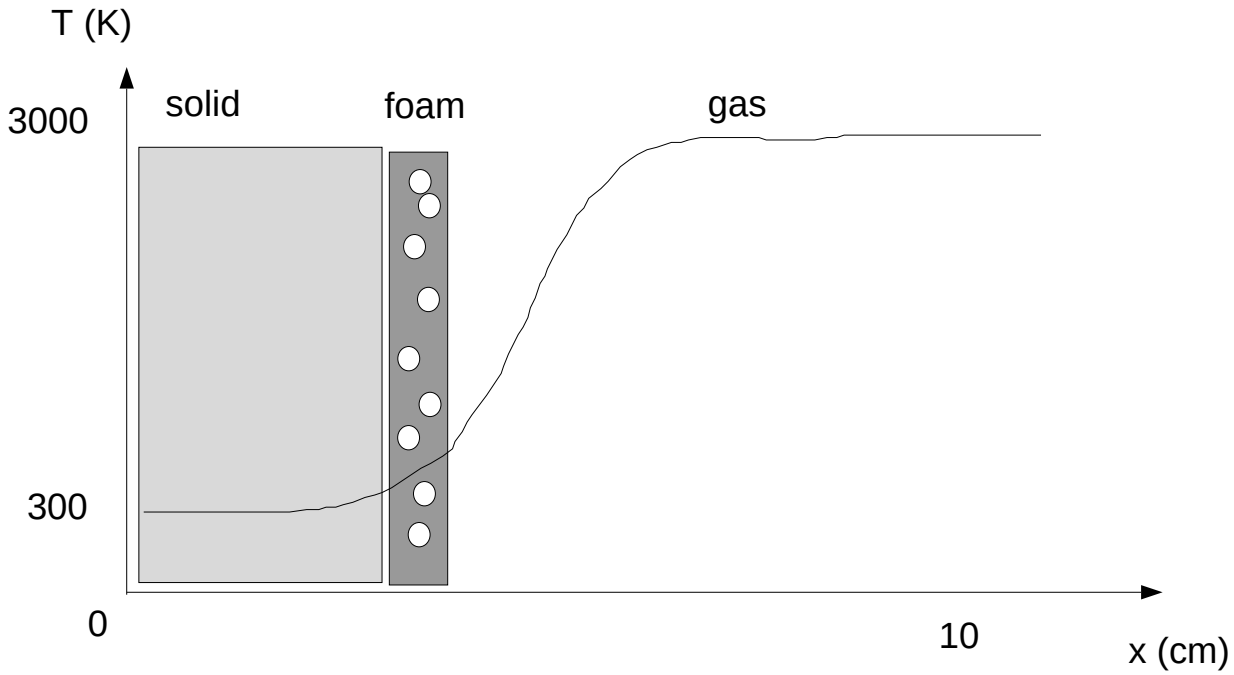
- Manifold methods not yet implemented
- We consider a constant pressure (0.75 atm) flame propagating into pure HMX pyrolysis product gas.
- This preliminary problem has invariant element concentration to facilitate construction of low-dimensional ILDM.
- Fundamental length scales dictated by flame speed which balances effects of reaction and diffusion
- Range of intrinsic length scales are severe for HMX flames: $\sim 10^{-14}\ m$ to $\sim 10^{-4}\ m$
- Typical geometric length scales of devices may be $\sim 10^{0}\ m$
- Preliminary calculations done on very small geometric domain with uniform grid (250 cells, so not all scales are resolved)
- Explicit convection diffusion step and implicit reaction step
- *For full problem, adaptive technique required with current computational resources.*

Motivation

- Detailed finite rate kinetics critical in reactive fluid mechanics:
  - Candle flames,
  - Atmospheric chemistry,
  - Internal combustion engines,
  - Gas phase reactions in energetic solid combustion.
- Common detailed kinetic models are computationally expensive.
  - 150 hr supercomputer time for calculation of steady, laminar, axisymmetric, methane-air diffusion flame (Smooke)
  - Expense increases with
    - number of species and reactions modeled (linear effect),
    - *stiffness*–ratio of slow to fast time scales, (geometric effect).
  - Fluid mechanics time scales: $10^{-5}\ s$ to $10^{1}\ s$.
  - Reaction time scales: $10^{-14}\ s$ to $10^{2}\ s$.
- Reduced kinetics necessary given current computational resources.
- Adaptive discretization necessary for fine spatial structures.
- Inclusion of *physical* diffusion necessary for *numerical* convergence.

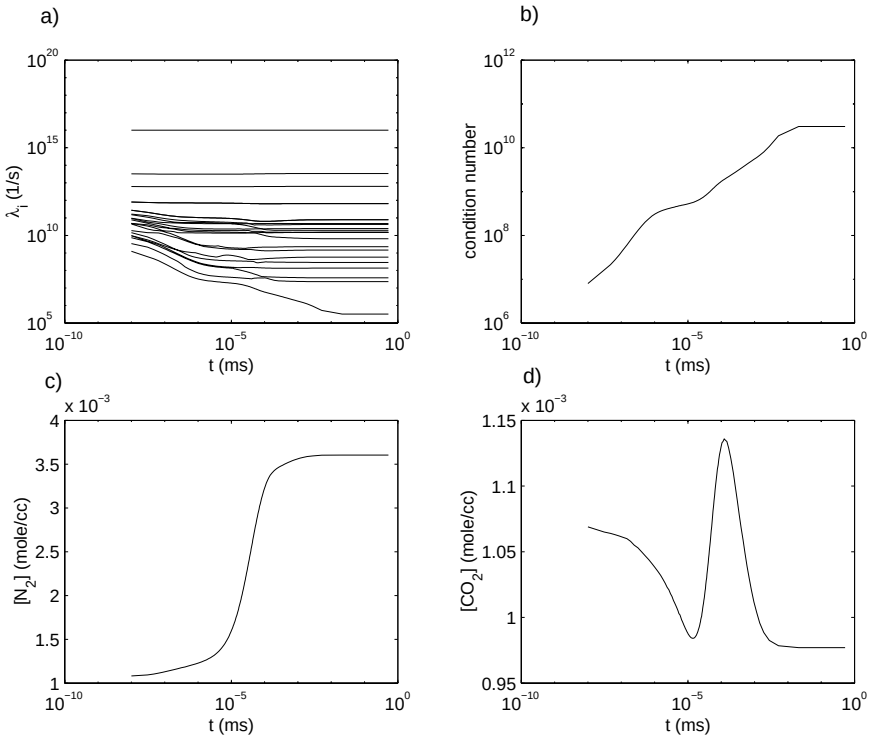
RDX/HMX COMBUSTION

- part of ongoing theoretical/experimental LANL study of low pressure $\sim 10\ atm$ combustion of explosives (Son, Liau, et al.),
- similar to solid propellant combustion,
- preheat zone in semi-infinite solid,
- two-phase bubbly liquid foam layer,
- gaseous flame region,
- gas phase reactions greatest computational burden in simulations.



RDX GAS PHASE COMBUSTION SIMULATION

- very similar to HMX
- Uses Yetter's 45 species, 232 reaction detailed kinetics mechanism,
- Constant pressure
- well-stirred
- fastest time scales $\sim 10^{-16}$ s!
- stiffness ratio (fastest time scale/slowest time scale) $\sim 10^{11}$



Goals

- Implement robust new reduced kinetic method (Intrinsic Low Dimensional Manifold-ILDM) of Maas and Pope (1992)
- Extend ILDM method to systems with time and space dependency while rationally accounting for convection and diffusion.
- Couple wavelet AMR (Paolucci & Vasilyev) and ILDM techniques.
- Applications:
  - premixed ozone flames
  - ignition delay in shock tubes
  - premixed HMX flames

Common Reduced Kinetics Strategies

- Fully frozen limit: no reaction allowed, *uninteresting*
- Fully equilibrated limit: commonly used in some problems
  - has value for events in which fluid time scales are slow with respect to reaction time scales,
  - misses events which happen on chemical time scales.
- Simple one and two step models
  - require significant intuition and curve fitting,
  - can give good first order results,
  - are often not robust.
- Partial equilibrium and steady-state assumptions
  - again require intuition,
  - are not robust.
- Sensitivity analysis
  - can remove need to include unimportant reactions,
  - not guaranteed to remove stiffness.

Intrinsic Low-Dimensional Manifold Method (ILDM)

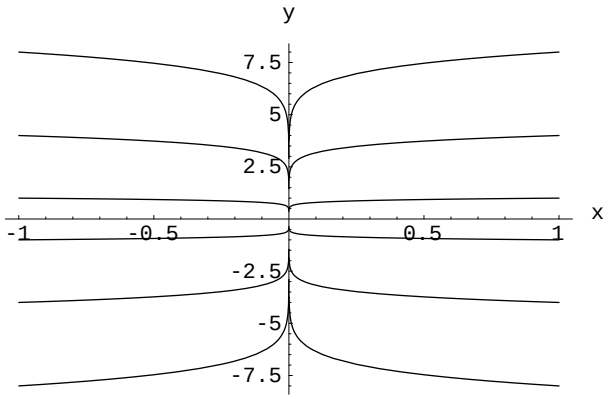
- Uses a dynamical systems approach,
- Does not require imposition of *ad hoc* partial equilibrium or steady state assumptions,
- Fast time scale phenomena are systematically equilibrated,
- Slow time scale phenomena are resolved in time,
- n-species gives rise to a n-dimensional phase space (same as composition space) for isochoric, isothermal combustion in well stirred reactors,
- Identifies m-dimensional subspaces (manifolds), $m < n$, embedded within the n-dimensional phase space on which slow time scale events evolve,
  - Fast time scale events rapidly move to the manifold,
  - Slow time scale events move on the manifold.
- Computation time reduced by factor of ~ 10 for non-trivial combustion problems; manifold gives much better roadmap to find solution relative to general implicit solution techniques (Norris, 1998)

Simplest Example

$$\frac{dx}{dt} = -10x, \qquad x(0) = x_o,$$
$$\frac{dy}{dt} = -y, \qquad y(0) = y_o.$$

- Stable equilibrium at $(x, y) = (0,0)$; stiffness ratio = 10.

- ILDM is $x = 0$



- Parameterization of manifold: $x(s) = 0; y(s) = s$.

$$\frac{dy}{dt} = \frac{dy}{ds}\,\frac{ds}{dt}, \qquad \text{chain rule}$$
$$-y(s) = \frac{dy}{ds}\,\frac{ds}{dt}, \qquad \text{substitute from ODE and manifold}$$
$$-s = (1)\frac{ds}{dt}, \qquad \text{no longer stiff!}$$
$$s = s_o e^{-t},$$
$$x(t) = 0; \qquad y(t) = s_o e^{-t}.$$

- Projection onto manifold for s_o, induces small phase error.

Formulation of General Manifolds

- A well stirred chemically reactive system is modeled by a set of non-linear ordinary differential equations:

$$\frac{d\mathbf{x}}{dt} = \mathbf{F}(\mathbf{x}), \qquad \mathbf{x}(0) = \mathbf{x}_o,$$

$$\mathbf{x}: \text{ species concentration}; \mathbf{x} \in \Re^n$$

- Equilibrium points defined by

$$\mathbf{x} = \mathbf{x}_{eq} \text{ such that } \mathbf{F}(\mathbf{x}_{eq}) = 0.$$

- Consider a system near equilibrium (the argument can and must be extended for systems away from equilibrium) with $\tilde{\mathbf{x}} = \mathbf{x} - \mathbf{x}_{eq}$.

- Linearization gives

$$\frac{d\tilde{\mathbf{x}}}{dt} = \mathbf{F}_{\mathbf{x}} \cdot \tilde{\mathbf{x}},$$

where $\mathbf{F}_{\mathbf{x}}$ is a *constant* Jacobian matrix.

- Schur decompose the Jacobian matrix:

$$\mathbf{F}_{\mathbf{x}} = \mathbf{Q} \cdot \mathbf{U} \cdot \mathbf{Q}^T$$

$$\mathbf{Q} = \begin{pmatrix} \vdots & \vdots & & \vdots \\ q_1 & q_2 & \cdots & q_n \\ \vdots & \vdots & & \vdots \end{pmatrix}, \quad \mathbf{U} = \begin{pmatrix} \lambda_1 & u_{12} & \cdots & u_{1n} \\ 0 & \lambda_2 & \cdots & u_{2n} \\ 0 & \cdots & \ddots & \vdots \\ 0 & \cdots & 0 & \lambda_n \end{pmatrix}, \quad \mathbf{Q}^T = \begin{pmatrix} \cdots & q_1^T & \cdots \\ \cdots & q_2^T & \cdots \\ & \vdots & \\ \cdots & q_n^T & \cdots \end{pmatrix}$$

Formulation of General Manifolds (cont.)

- $\mathbf{Q}$ is an orthogonal matrix with real Schur vectors q_i in its columns.
- $\mathbf{U}$ is an upper triangular matrix with eigenvalues of $\mathbf{F_x}$ on its diagonal, sometimes placed in order of decreasing magnitude.
- The Schur vectors q_i form an orthonormal basis which spans the phase space, $\Re^n$.
- We then define m slow time scales, $m \leq n$.
- Next define a non-square matrix $\mathbf{W}$ which has in its rows the Schur vectors associated with the fast time scales:

$$\mathbf{W} = \begin{pmatrix} \cdots & \cdots & q_{m+1}^T & \cdots & \cdots \\ \cdots & \cdots & q_{m+2}^T & \cdots & \cdots \\ & & \vdots & & \\ \cdots & \cdots & q_n^T & \cdots & \cdots \end{pmatrix}.$$

- Letting the fast time scale events equilibrate defines the manifold:

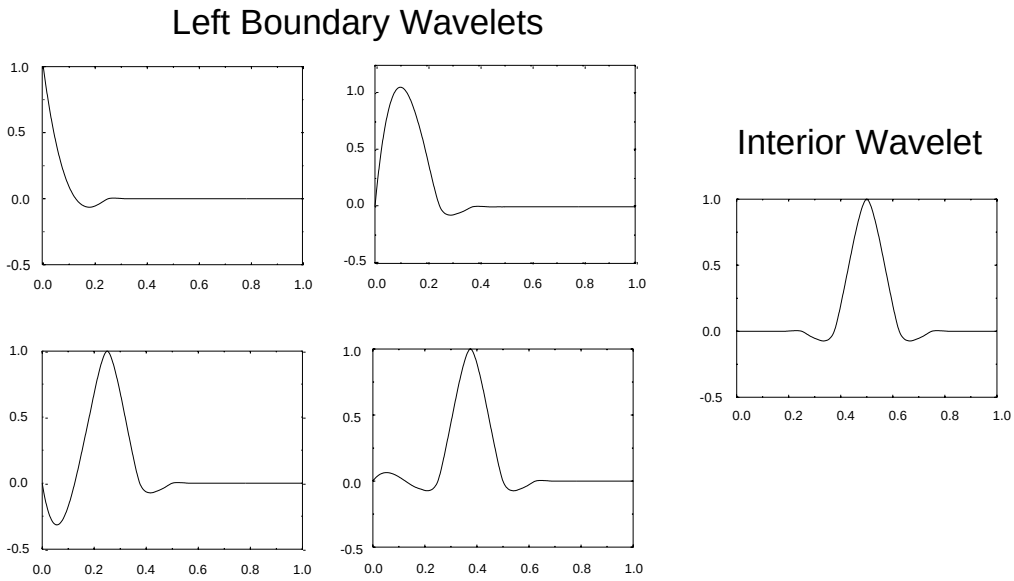
$$\mathbf{W} \cdot \mathbf{F}(\mathbf{x}) = 0.$$

- If $m = 0$, no slow time scales, $\mathbf{W} = \mathbf{Q}^T$, and $\mathbf{W} \cdot \mathbf{F}(\mathbf{x}) = 0$ implies $\mathbf{Q}^T \cdot \mathbf{F}(\mathbf{x}) = 0$, implies $\mathbf{F}(\mathbf{x}) = 0$: the equilibrium point is the low dimensional manifold!

Adaptive Multilevel Wavelet Collocation Technique

- Summary of standard spatial discretization techniques
  - Finite difference-good spatial localization, poor spectral localization, and slow convergence,
  - Finite element- good spatial localization, poor spectral localization, and slow convergence,
  - Spectral–good spectral localization, poor spatial localization, but fast convergence.
- Wavelet technique
  - See e.g. Vasilyev and Paolucci, "A Fast Adaptive Wavelet Collocation Algorithm for Multidimensional PDEs," *J. Comp. Phys.*, 1997,
  - Basis functions have compact support,
  - Well-suited for problems with widely disparate spatial scales,
  - Good spatial and spectral localization, and fast (spectral) convergence,
  - Easy adaptible to steep gradients via adding collocation points,
  - Spatial adaptation is automatic and dynamic to achieve prescribed error tolerance.

Wavelet Basis Functions



- Boundary-modified Daubechies autocorrelation functions and interior Daubechies autocorrelation function of order four
- Scaling function

$$\phi_{j,k}(x) = \phi(2^j x - k)$$

- Definition of the wavelet function on the first level

$$\psi_{1,0}(x) = \phi(2x - 1)$$

- Definition of the wavelet function on j level

$$\psi_{j,k+1}(x) = \psi(2^{j-1}x - k)$$

Algorithm Description

- Approximate initial function using wavelet basis,

$$\mathbf{P}^J u(x) = \sum_k u_{0,k}\phi_{0,k}(x) + \sum_{j=1}^{J}\sum_k d_{j,k}\psi_{j,k}(x)$$

- Discard non-essential wavelets if amplitude below threshold value (here we look only at P, T, u, and ρ, species could be included),

$$\mathbf{P}^J u(x) = u^J_{\geq}(x) + u^J_{<}(x)$$

$$u^J_{\geq}(x) = \sum_k u_{0,k}\phi_{0,k}(x) + \sum_{j=1}^{J}\sum_k d_{j,k}\psi_{j,k}(x), |d_{j,k}| \geq \epsilon$$

$$u^J_{<}(x) = \sum_{j=1}^{J}\sum_k d_{j,k}\psi_{j,k}(x), |d_{j,k}| < \epsilon$$

- Assign a collocation point to every essential wavelet,
- Establish a neighboring region of potentially essential wavelets,
- Discretize the spatial derivatives; five points used here (related to order of wavelet family),
- Integrate in time; linearized trapezoidal method (implicit) used here,
- Repeat

Ignition Delay in Premixed H_2-O_2

- Consider standard problem of Fedkiw, Merriman, and Osher, *J. Comp. Phys.*, 1996,
- Shock tube with premixed H_2, O_2, and Ar in 2/1/7 molar ratio,
- Initial inert shock propagating in tube,
- Reaction commences shortly after reflection off end wall,
- Detonation soon develops,
- Model assumptions
  - One-dimensional,
  - mass, momentum, and energy diffusion,
  - Nine species, thirty-seven reactions,
  - Ideal gases with variable specific heats.
  - Wavelet collocation technique used for AMR

Compressible Reactive Navier-Stokes Equations for H_2-O_2 Problem

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x}(\rho u) = 0, \qquad \text{mass}$$

$$\frac{\partial}{\partial t}(\rho u) + \frac{\partial}{\partial x}\left(\rho u^2 + P - \tau\right) = 0, \qquad \text{momentum}$$

$$\frac{\partial}{\partial t}\left(\rho\left(e + \frac{u^2}{2}\right)\right) + \frac{\partial}{\partial x}\left(\rho u\left(e + \frac{u^2}{2}\right) + u\left(P - \tau\right) + q\right) = 0, \qquad \text{energy}$$

$$\frac{\partial}{\partial t}(\rho Y_i) + \frac{\partial}{\partial x}(\rho u Y_i + j_i) = \sum_{j=1}^{M} a_j T^{\alpha_j} \exp\left(\frac{-E_j}{\Re T}\right) \nu_{ij} M_i \prod_{k=1}^{N}\left(\frac{\rho Y_k}{M_k}\right)^{\nu_{kj}}, \qquad \text{species}$$

$$P = \rho \Re T \sum_{i=1}^{N} \frac{Y_i}{M_i}, \qquad \text{thermal equation of state}$$

$$e = \sum_{i=1}^{N} Y_i \left(h_i^o + \int_{T_o}^{T} c_{pi}(\hat{T}) d\hat{T}\right) - \frac{P}{\rho}, \qquad \text{caloric equation of state}$$

$$\tau = \frac{4}{3}\mu \frac{\partial u}{\partial x}, \qquad \text{Newtonian gas with Stokes' assumption}$$

$$j_i = -\rho \sum_{j=1}^{N} \mathcal{D}_{ij} \frac{\partial Y_j}{\partial x}, \qquad \text{Fick's law}$$

$$q = -k\frac{\partial T}{\partial x} + \sum_{i=1}^{N} j_i \left(h_i^o + \int_{T_o}^{T} c_{pi}(\hat{T}) d\hat{T}\right) \qquad \text{augmented Fourier's law.}$$

$N = 9$ species: H_2, O_2, H, O, OH, H_2O_2, H_2O, HO_2, Ar

$M = 37$ reactions

Operator Splitting Technique

- Equations are of form

$$\frac{\partial}{\partial t}\mathbf{q}(x,t) + \frac{\partial}{\partial x}\mathbf{f}(\mathbf{q}(x,t)) = \mathbf{g}(\mathbf{q}(x,t)).$$

  where

$$\mathbf{q} = \left(\rho, \rho u, \rho\left(e + \frac{u^2}{2}\right), \rho Y_i\right)^T$$

- $\mathbf{f}$ models convection and diffusion
- $\mathbf{g}$ models reaction source terms
- Splitting
  1. Inert convection diffusion step:

$$\frac{\partial}{\partial t}\mathbf{q}(x,t) + \frac{\partial}{\partial x}\mathbf{f}(\mathbf{q}(x,t)) = 0,$$
$$\frac{d}{dt}\mathbf{q}_i(t) = -\Delta_x \mathbf{f}(\mathbf{q}_i(t)).$$

     Δ_x is either Godunov *or* wavelet discretization operator.

  2. Reaction source term step:

$$\frac{\partial}{\partial t}\mathbf{q}(x,t) = \mathbf{g}(\mathbf{q}(x,t)),$$
$$\frac{d}{dt}\mathbf{q}_i(t) = \mathbf{g}(\mathbf{q}_i(t)).$$

- Operator splitting with implicit stiff source solution can induce non-physical wave speeds! (LeVeque and Yee, *JCP* 1990)

ILDM Implementation in Operator Splitting

- Form of equations in source term step:

$$\frac{d}{dt}\begin{pmatrix} \rho \\ \rho u \\ \rho\left(e+\frac{u^2}{2}\right) \\ \rho Y_i \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ \omega \end{pmatrix}.$$

- Equations reduce to

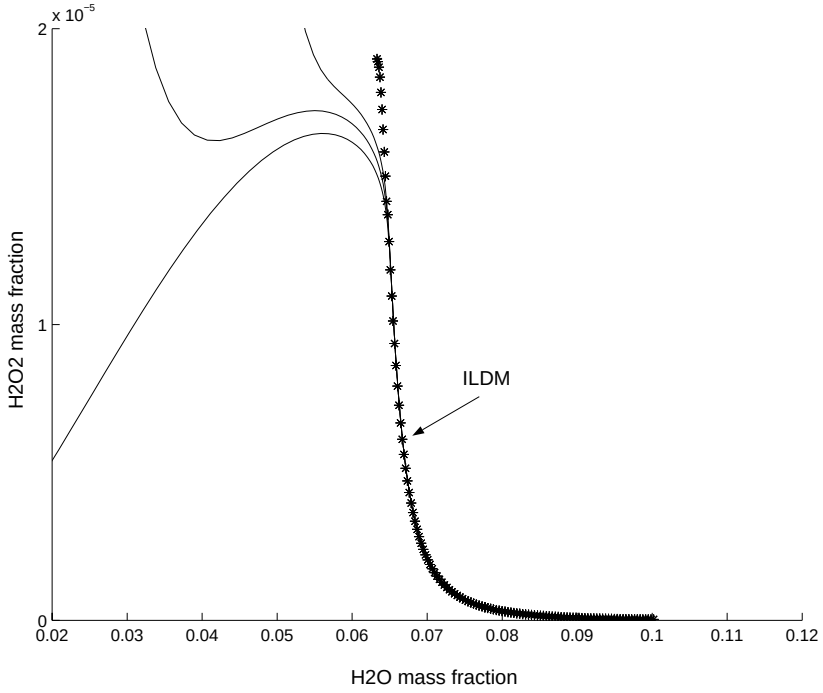
$$\rho = \rho_o, \qquad u = u_o, \qquad e = e_o,$$

$$\frac{dY_i}{dt} = \frac{\omega}{\rho_o}.$$

- ω has dependency on ρ, e, and Y_i
- ODEs for Y_i can be attacked with manifold methods when manifold with ρ, e, H and O parameterization is available.
- In premixed problem, H and O element concentrations are remarkably constant, reducing the dimension by two!
- Full equations integrated until sufficiently close to manifold
- Once on manifold, simple projection used to return to manifold following convection-diffusion step

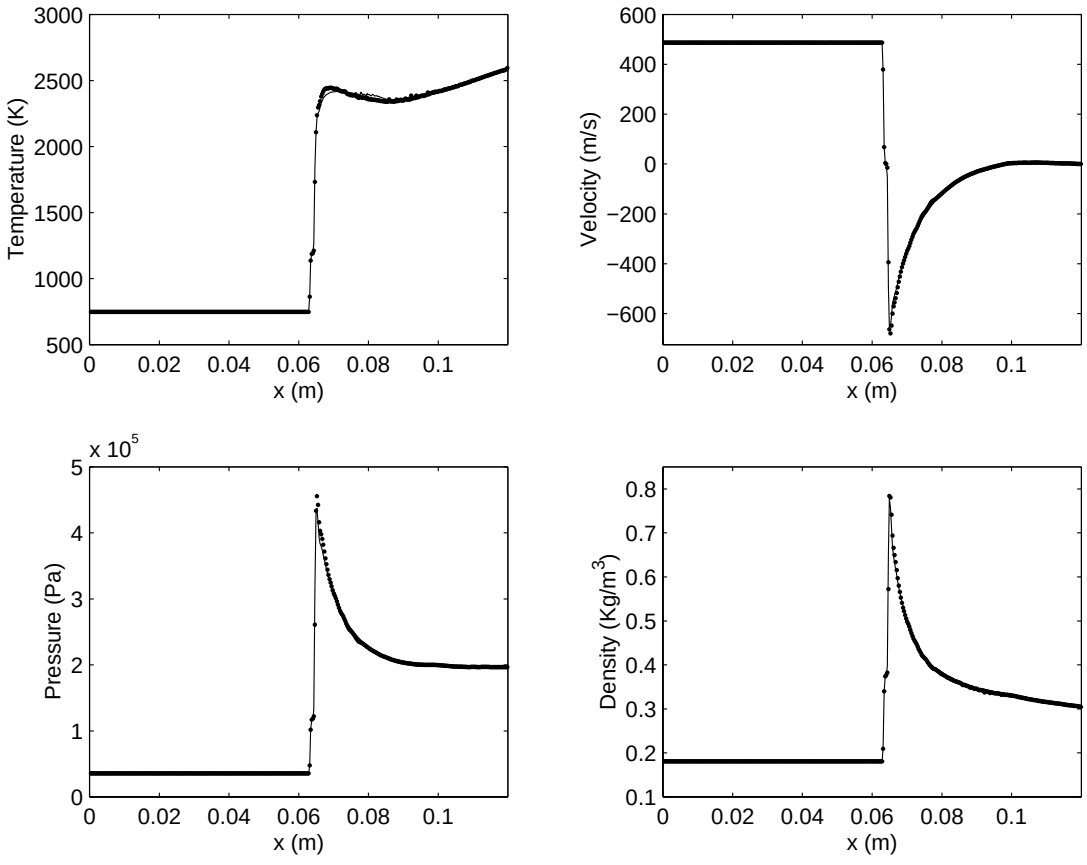
Sample ILDM for $H_2 - O_2$

- Projection of ILDM in H_2O, H_2O_2 plane,
- Adiabatic ($e = 525\ kJ/kg$), isochoric ($\rho = 0.25\ kg/m^3$), element concentrations of H and O constant,
- Complete manifold tabulated in three dimensions: ρ, e, Y_{H_2O},
- So we have e.g. $P\left(\rho, e, Y_{H_2O}\right), T\left(\rho, e, Y_{H_2O}\right), Y_H\left(\rho, e, Y_{H_2O}\right), \ldots$
- Linear interpolation used for points not in table,
- Captures $\sim 0.1\ \mu s$ reaction events.

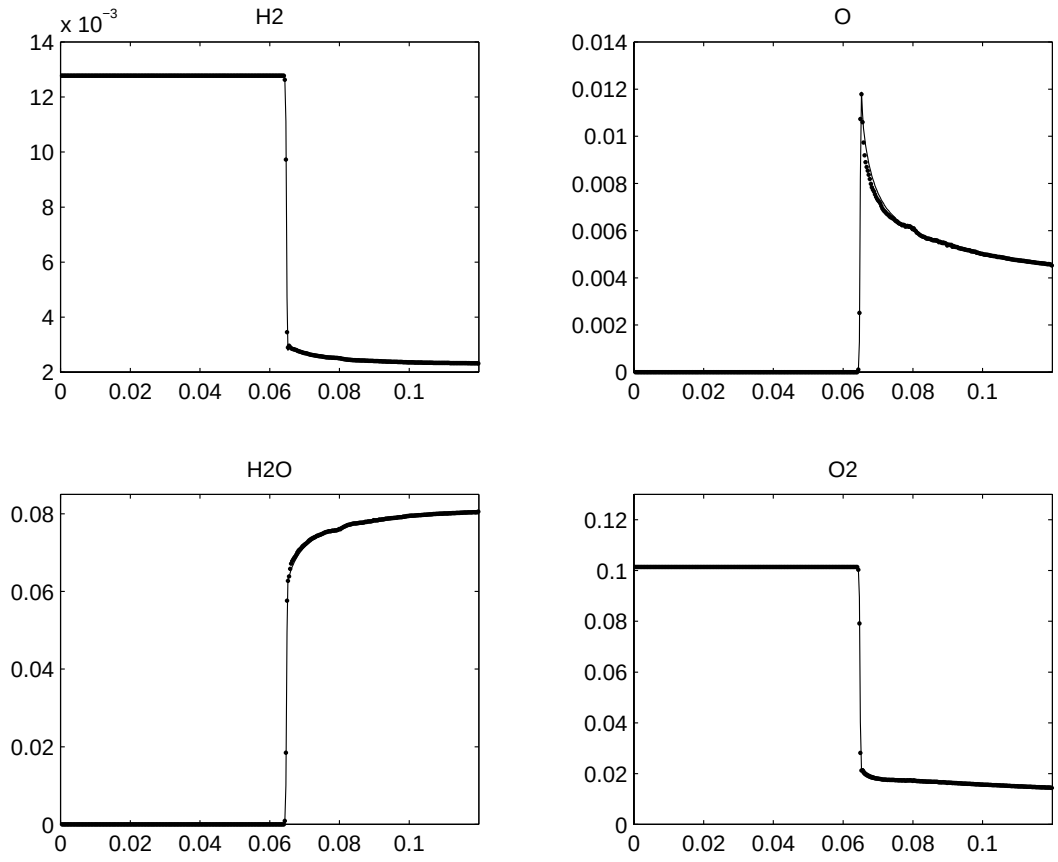


Inviscid $H_2 - O_2$ Ignition Delay with and without ILDM

- No diffusion,
- Godunov spatial discretization, 400 uniform finite difference cells,
- Implicit (trapezoidal) convection step; Implicit (dlsode) *or* ILDM reaction step,
- Correction of Fedkiw adopted to suppress artificial entropy layer after shock reflection (see Menikoff, 1994).

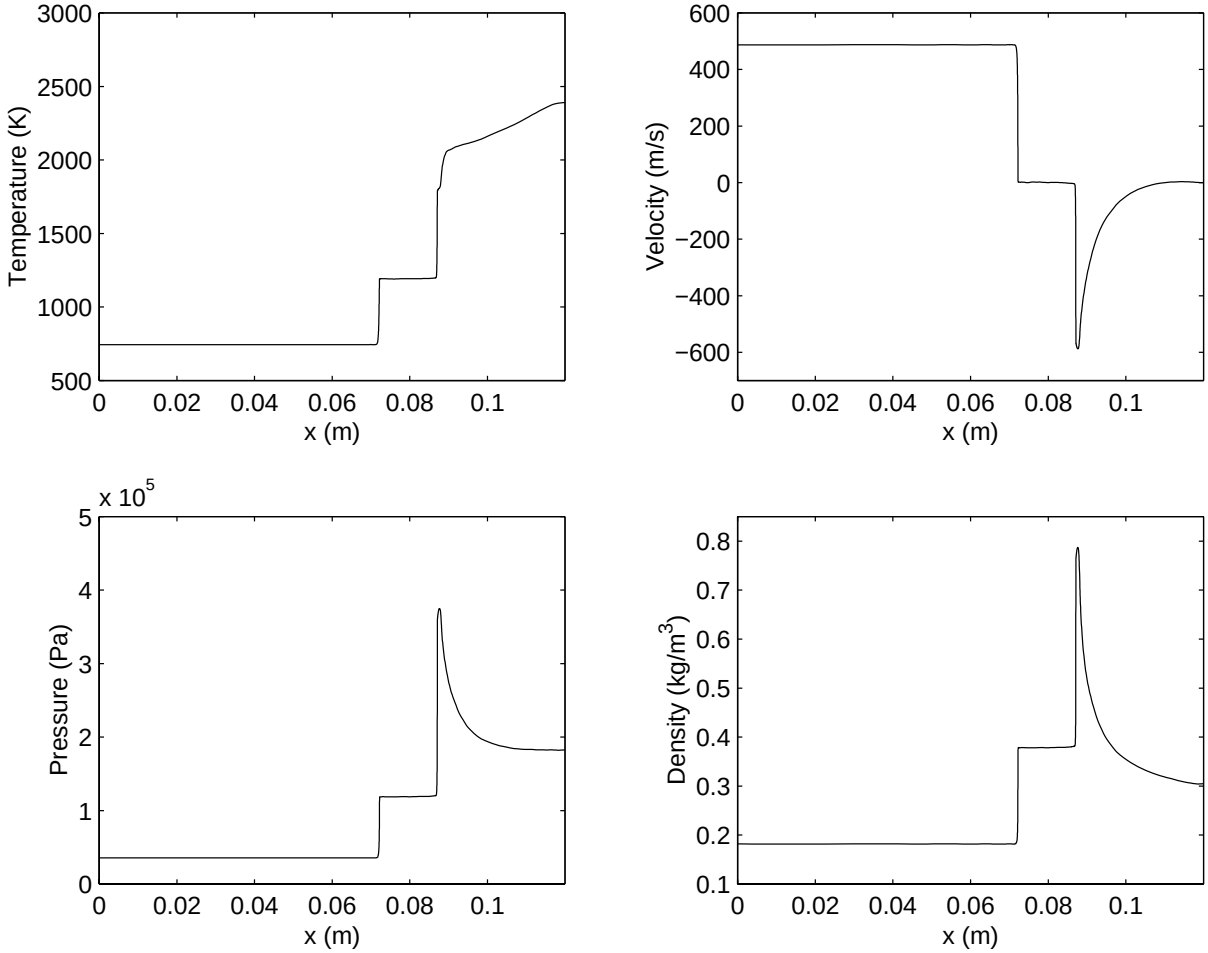


Inviscid $H_2 - O_2$ Ignition Delay with and without ILDM



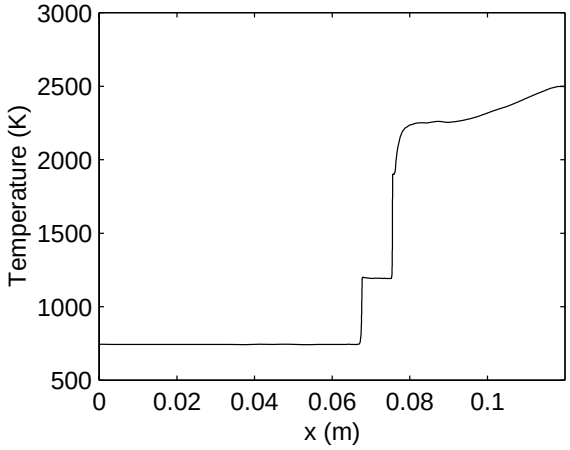
Viscous $H_2 - O_2$ Ignition Delay with Wavelets

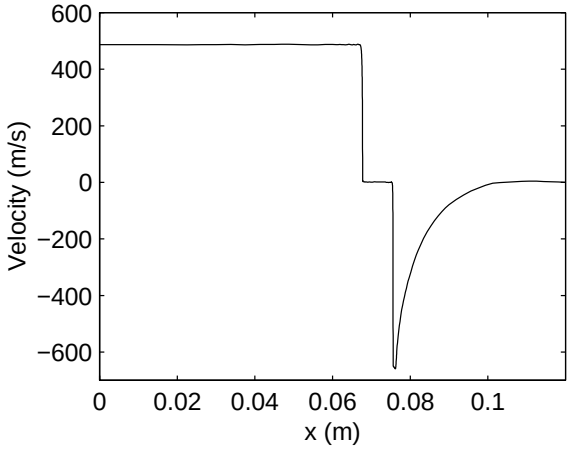
- Mass, momentum, and energy diffusion modelled,
- Wavelet spatial discretization, explicit convection-diffusion time stepping, implicit reaction time stepping,
- 300 collocation points, 15 wavelet levels,
- *Viscous shocks, induction zones, and entropy layers spatially resolved!*
- $t = 180\ \mu s$.

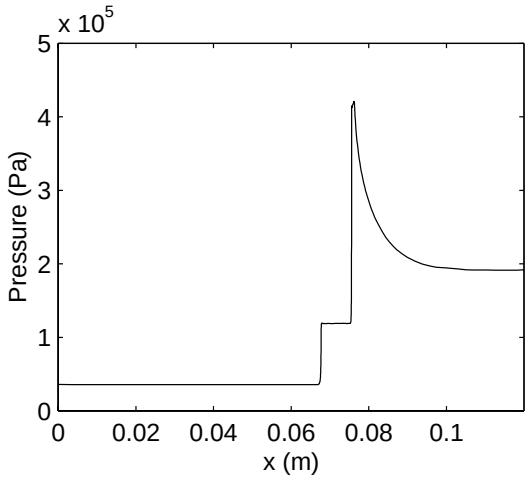


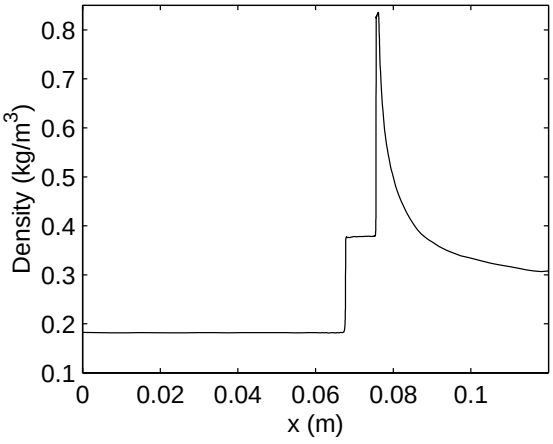
Viscous $H_2 - O_2$ Ignition Delay with Wavelets

- $t = 190\ \mu s$



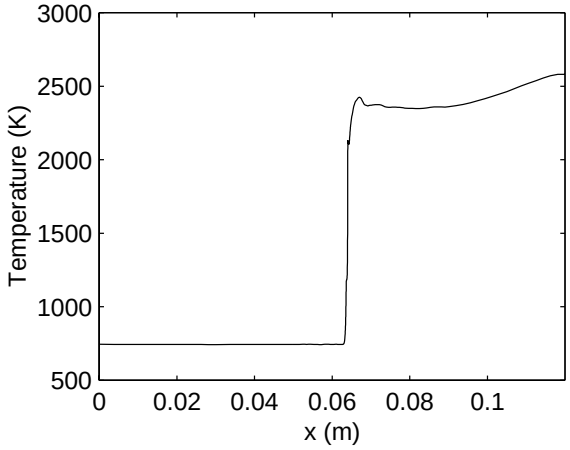


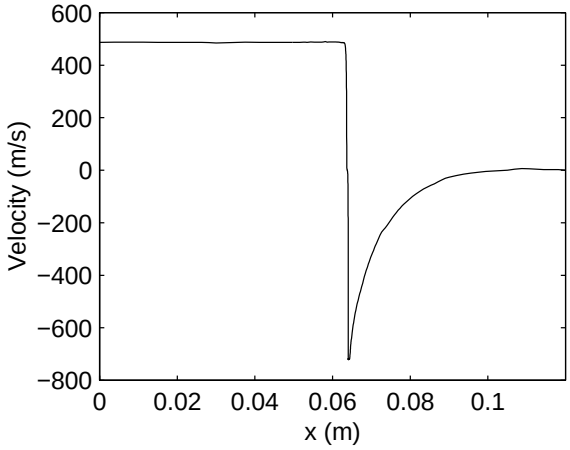


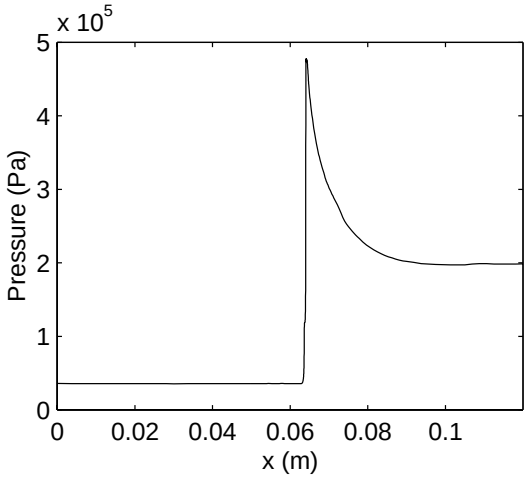


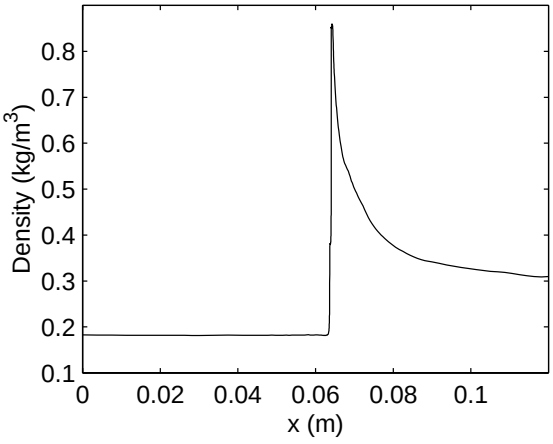
Viscous $H_2 - O_2$ Ignition Delay with Wavelets

- $t = 200\ \mu s$



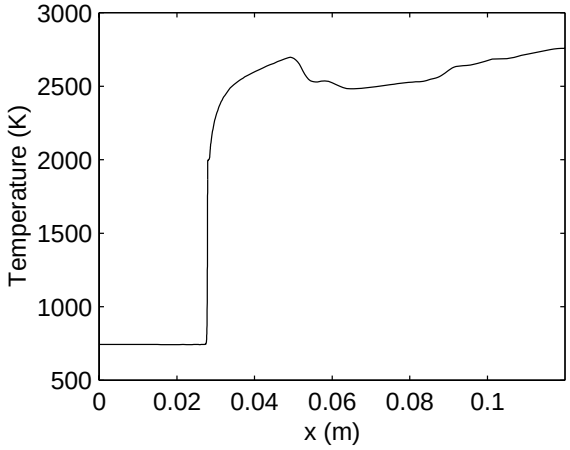


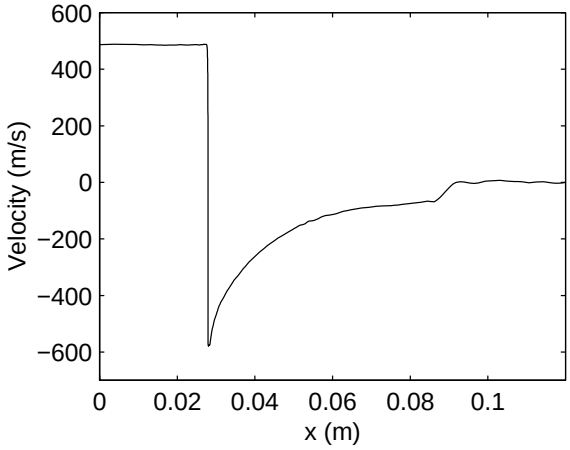


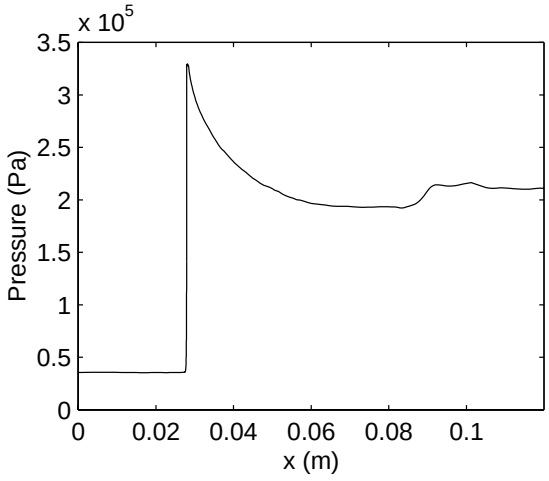


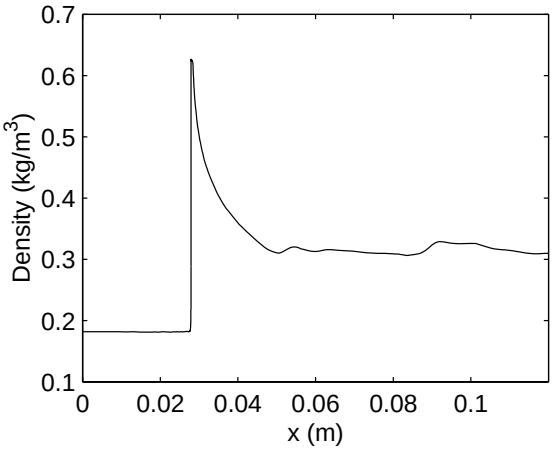
Viscous $H_2 - O_2$ Ignition Delay with Wavelets

- $t = 230\ \mu s$



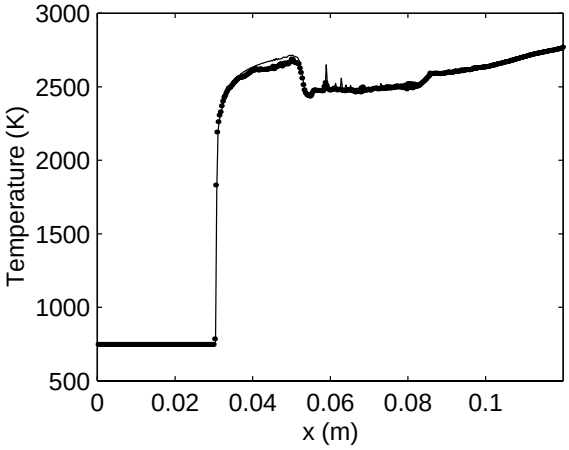


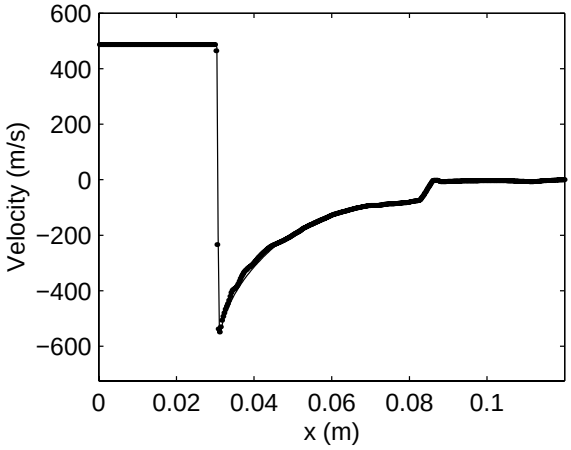


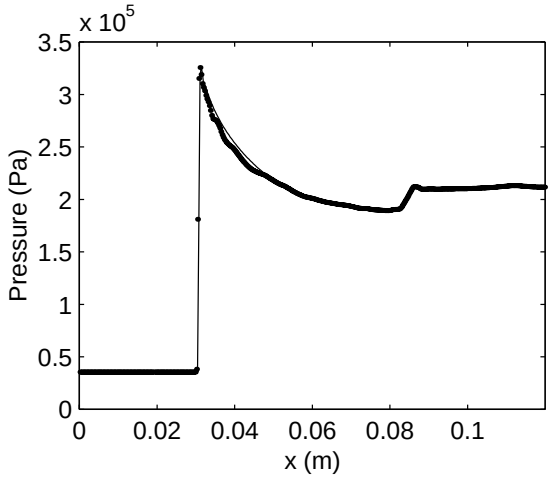


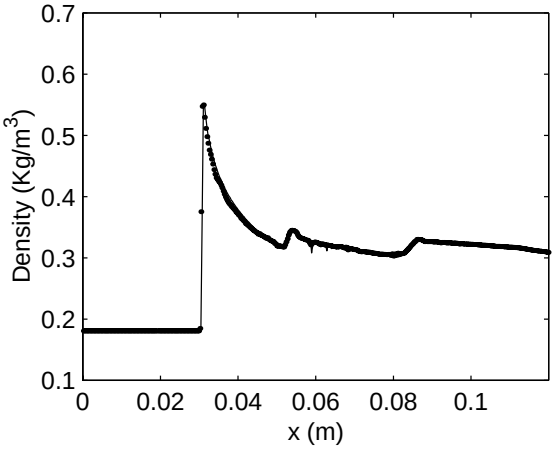
Comparison with Inviscid/ILDM Result at Same Time

- $t = 230\ \mu s$



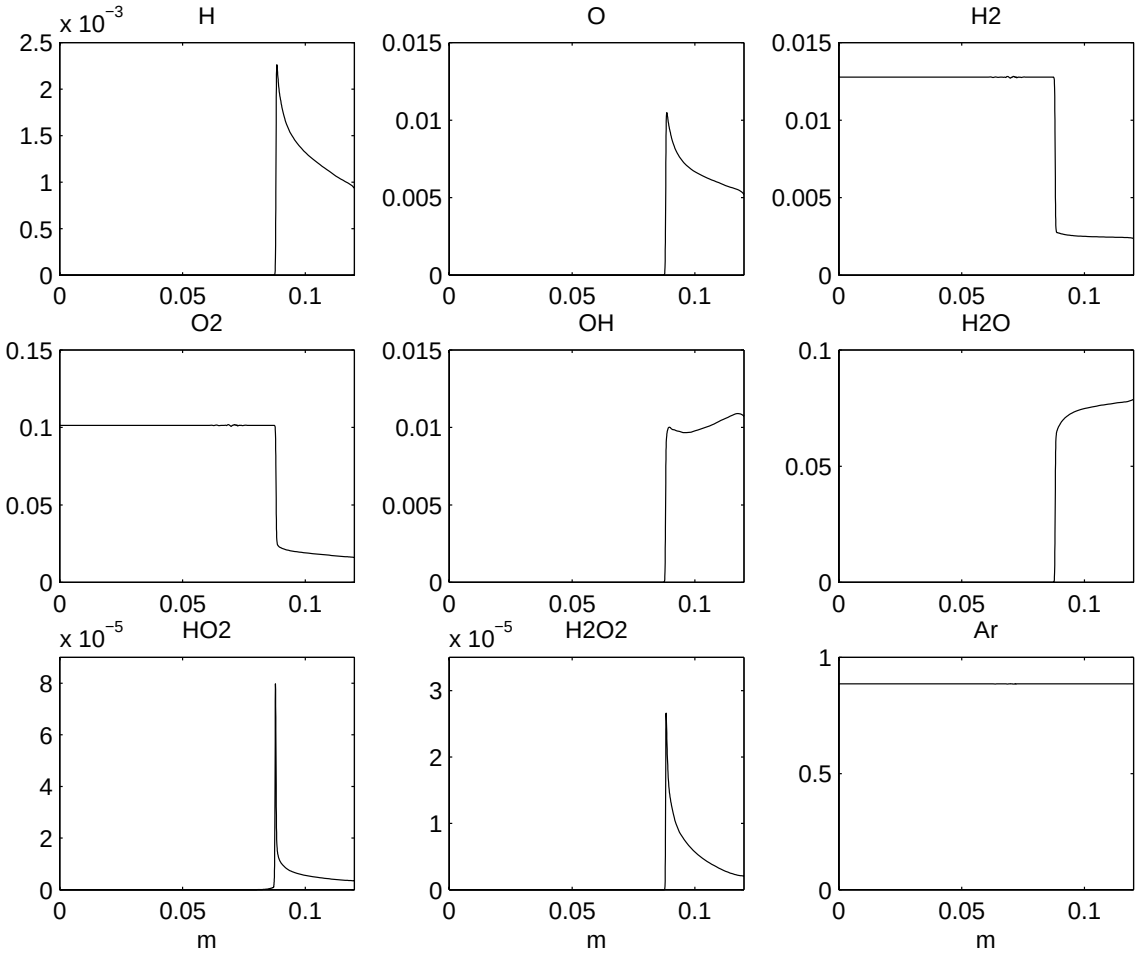






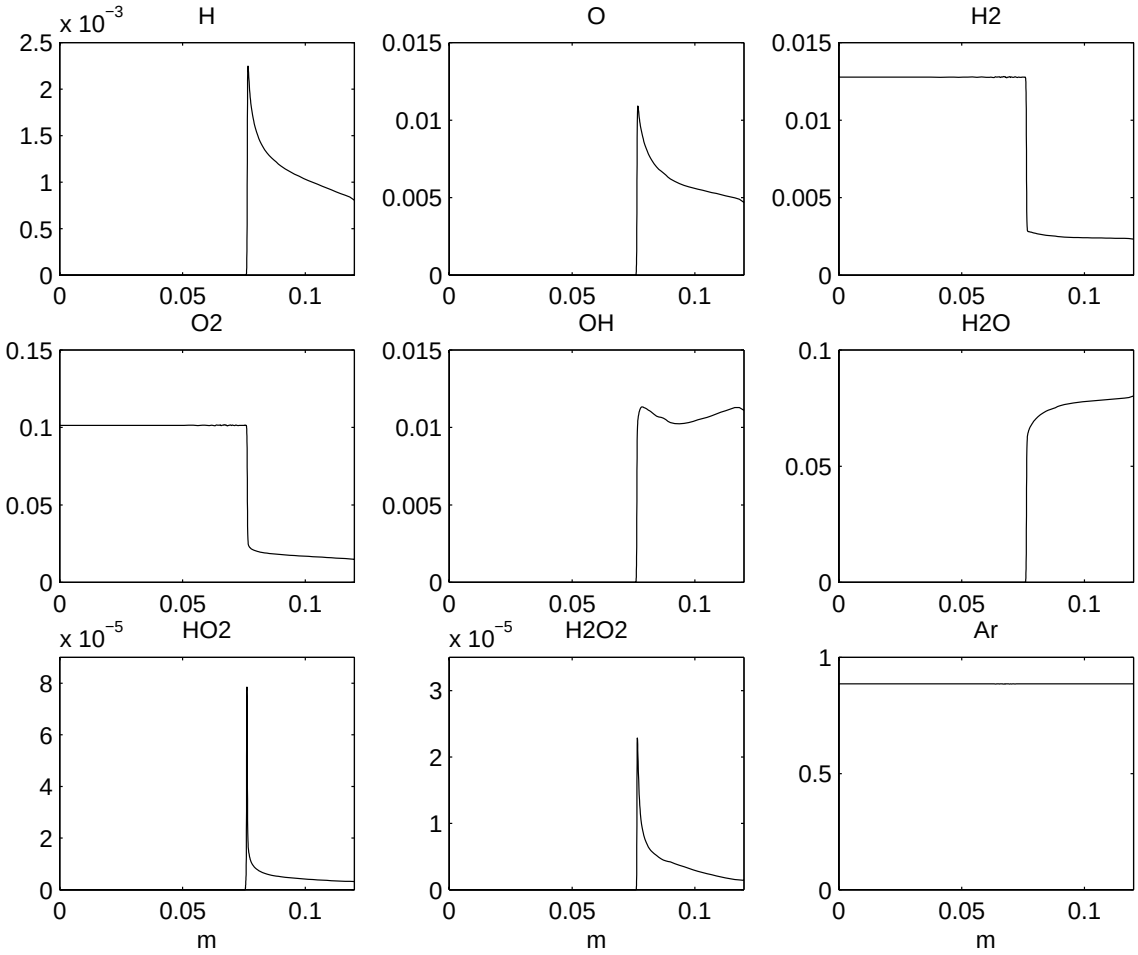
Viscous $H_2 - O_2$ Ignition Delay with Wavelets

- $t = 180\ \mu s$
- species mass fractions plotted vs. distance



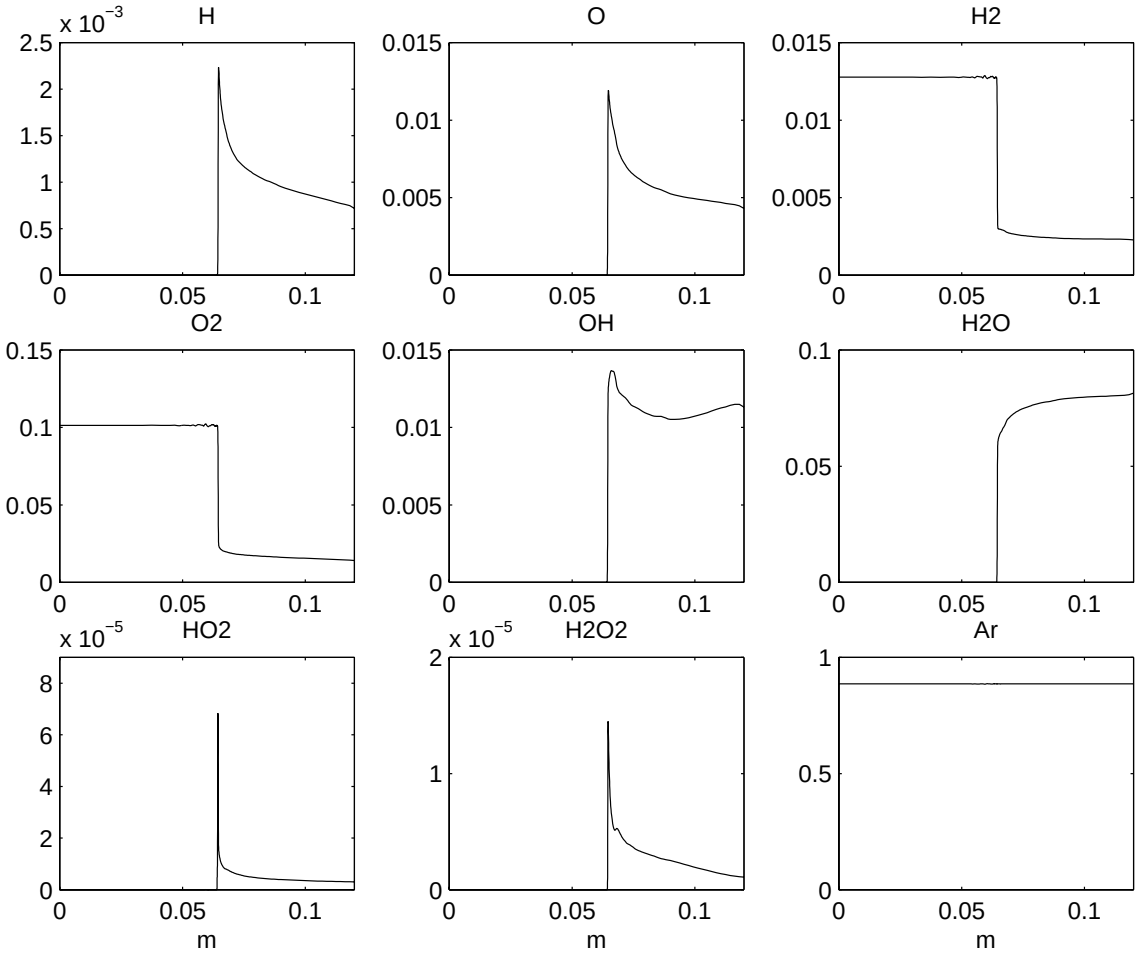
Viscous $H_2 - O_2$ Ignition Delay with Wavelets

- $t = 190\ \mu s$
- species mass fractions plotted vs. distance



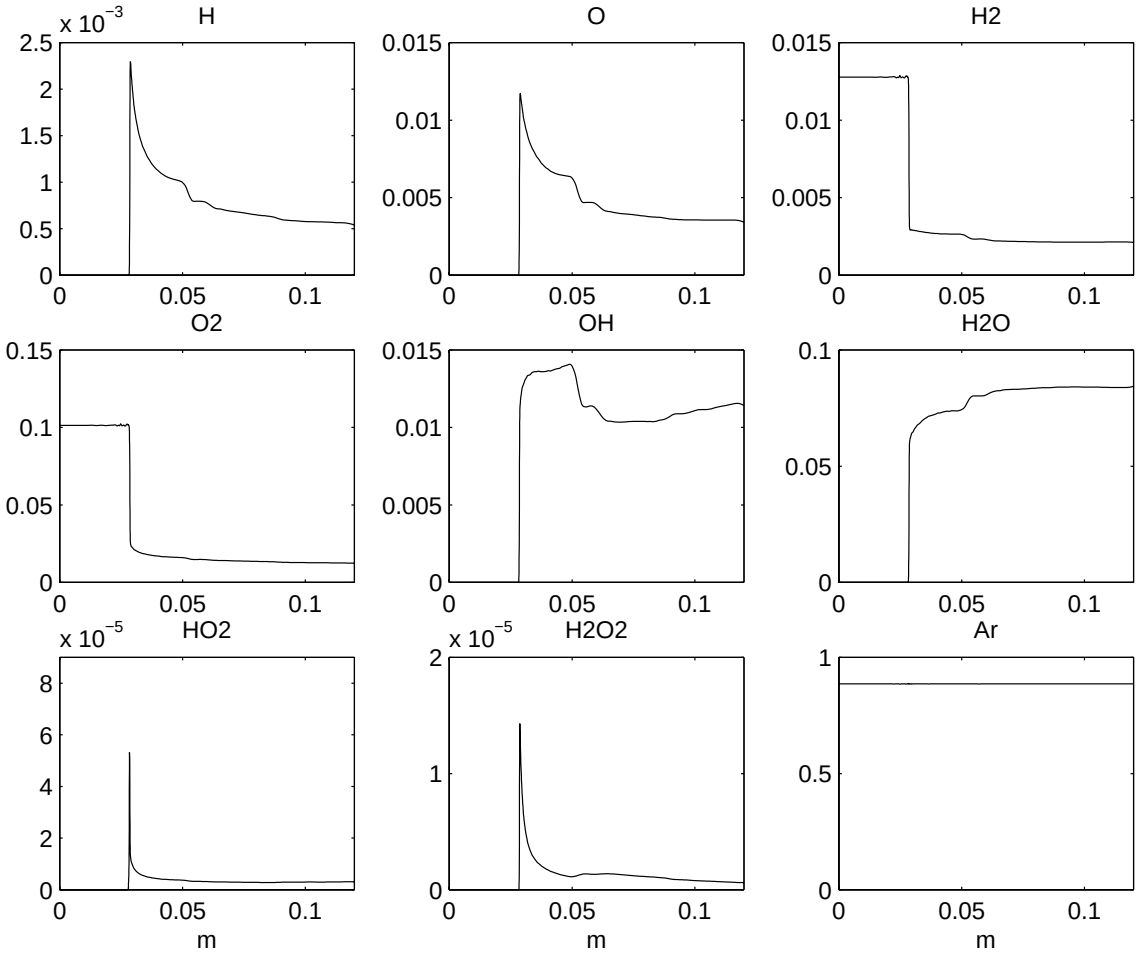
Viscous $H_2 - O_2$ Ignition Delay with Wavelets

- $t = 200\ \mu s$
- species mass fractions plotted vs. distance



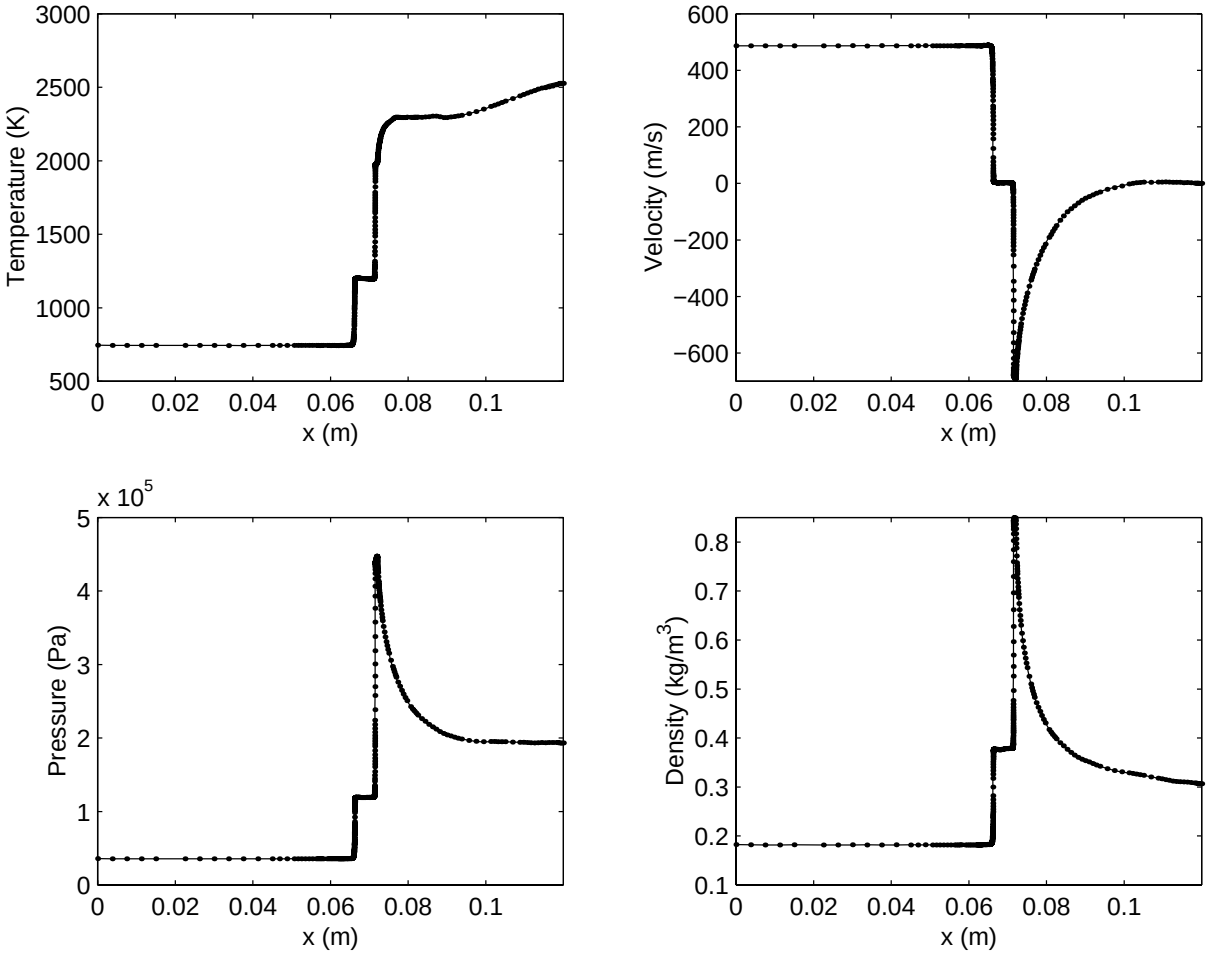
Viscous $H_2 - O_2$ Ignition Delay with Wavelets

- $t = 230\ \mu s$
- species mass fractions plotted vs. distance



Viscous $H_2 - O_2$ Ignition Delay with Wavelets and ILDM

- $t = 195\ \mu s$
- ILDM gives nearly identical results as full chemistry
- computational time $\sim$ 10 hours on 330 MHz Sun Ultra10 for full chemistry and ILDM !?
- ILDM *will improve dramatically* when inefficient I/O is eliminated (factor of at least 10 for simpler problems)



Viscous $H_2 - O_2$ Ignition Delay with Wavelets and ILDM

- $t = 195\ \mu s$
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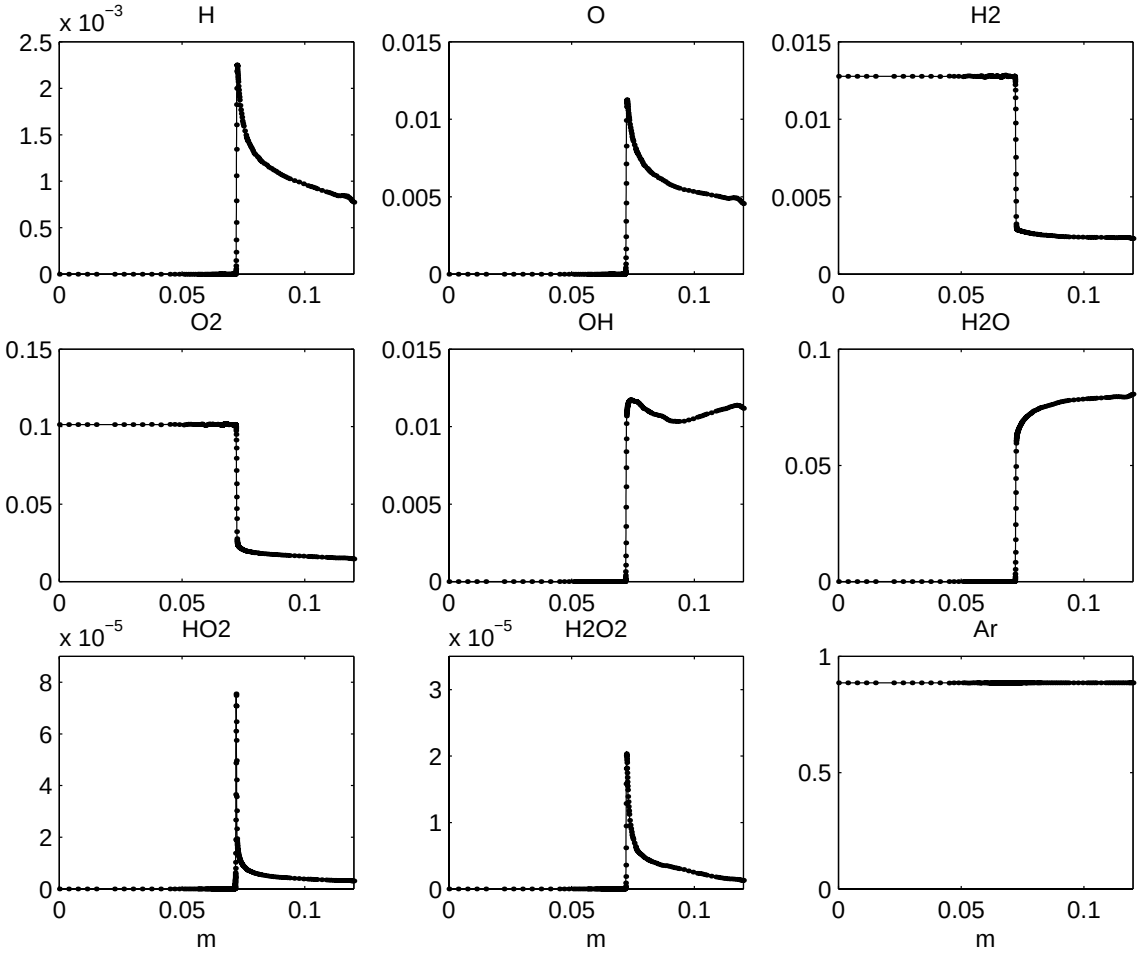
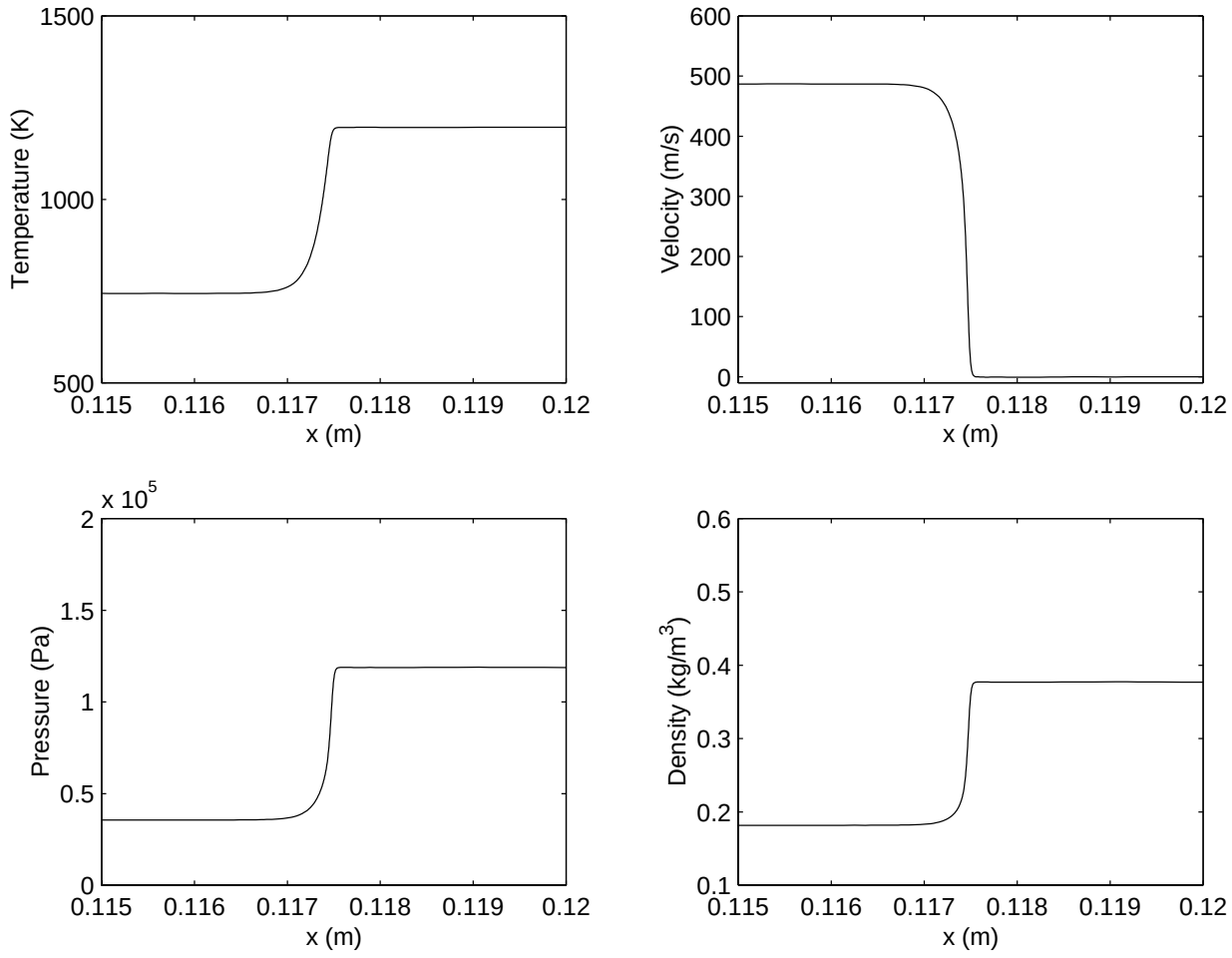


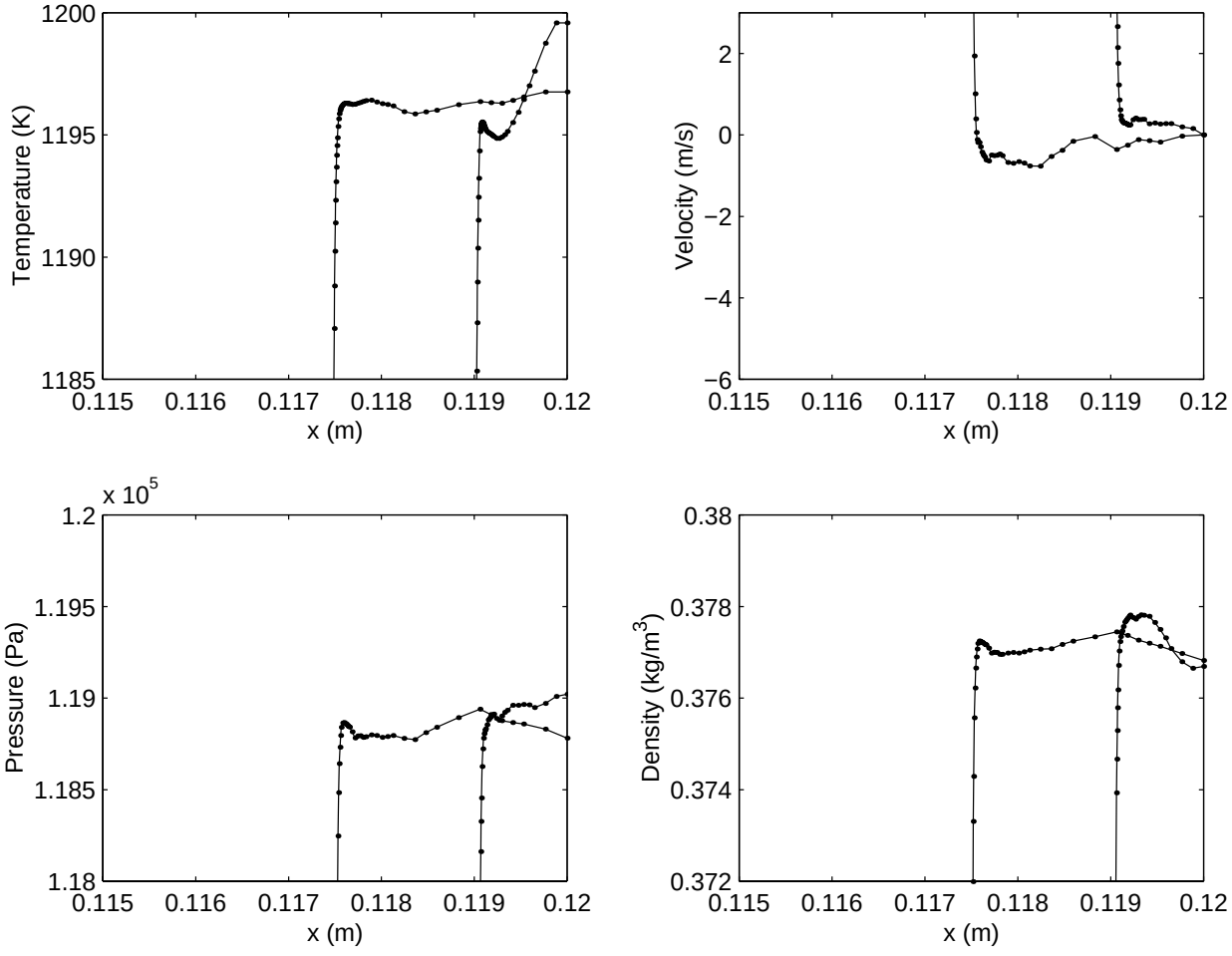
Post Reflection Entropy Layer?: Viscous Wavelet Results

- No significant entropy layer evident on macroscale after shock reflection when resolved viscous terms considered,
- Inviscid codes with coarse gridding introduce a larger entropy layer due to numerical diffusion,
- Unless suppressed, unphysically accelerates reaction rate.



Post Reflection Entropy Layer: Viscous Wavelet Results

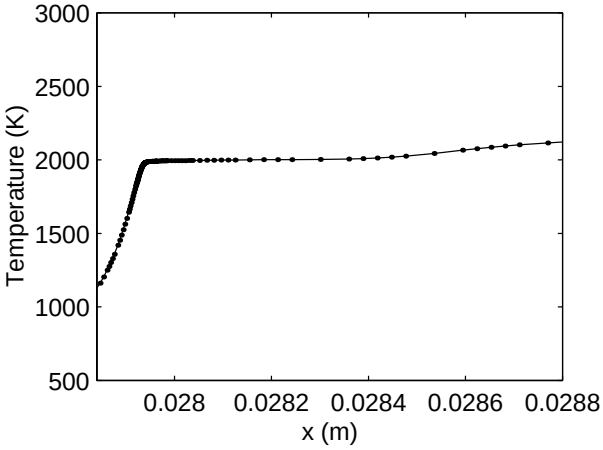
- small entropy layer evident on finer scale,
- temperature rise $\sim 5\ K$; dissipates quickly,
- inviscid calculations before adjustment give persistent temperature rise of $\sim 20\ K$; reaction acceleration small.

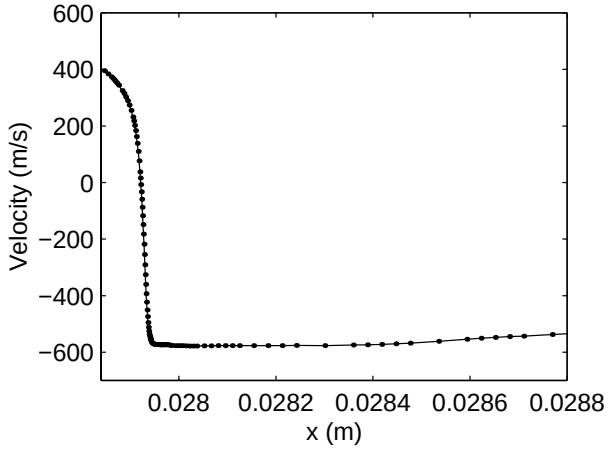


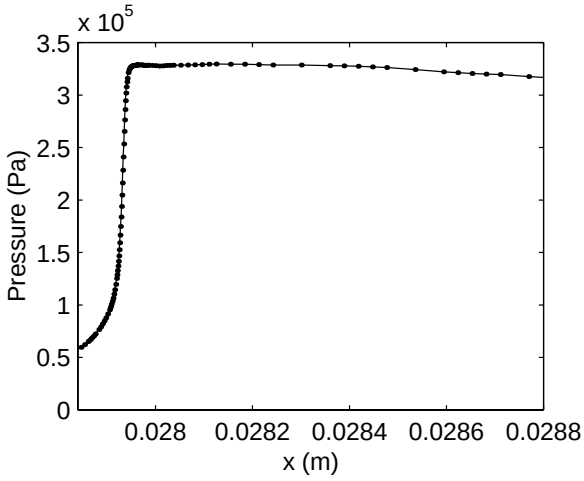
Viscous $H_2 - O_2$ Ignition Delay with Wavelets

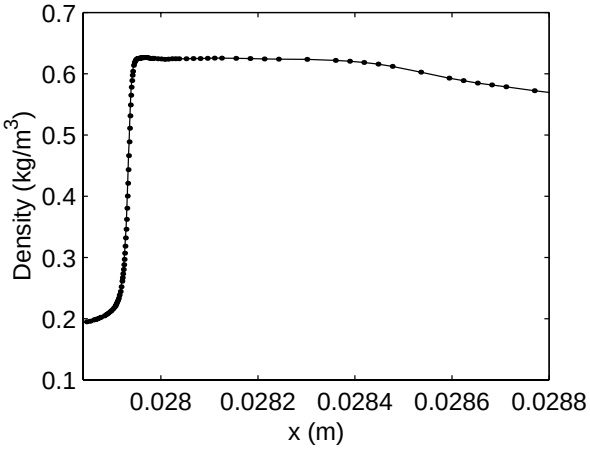
Close-up: Viscous Shock Stucture and Induction Zone

- $t = 230\ \mu s$,
- Induction zone length: $\sim 470\ \mu m$,
- No significant reaction in viscous shock zone.





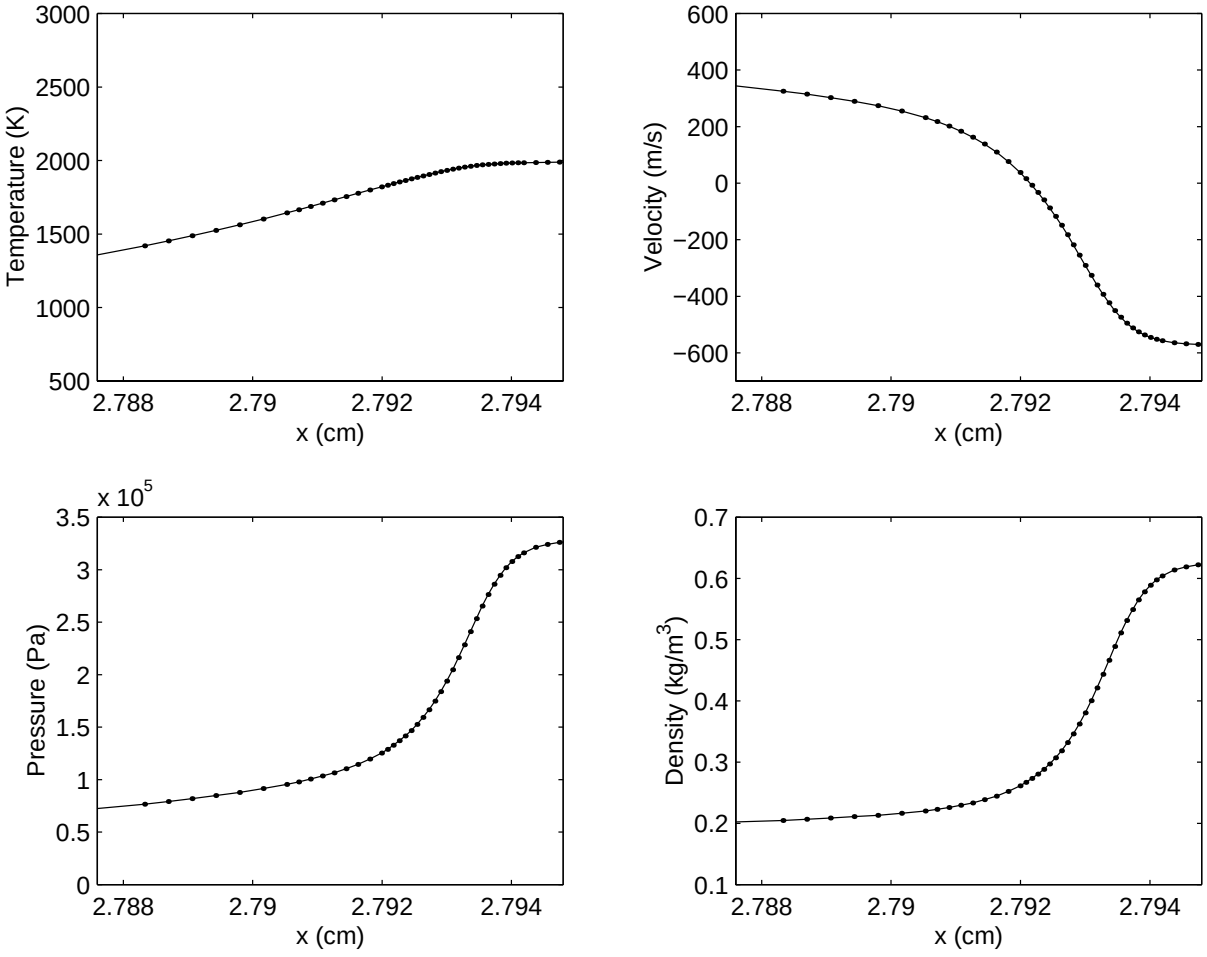




Viscous $H_2 - O_2$ Ignition Delay with Wavelets

Closer-up: Viscous Shock Stucture Only

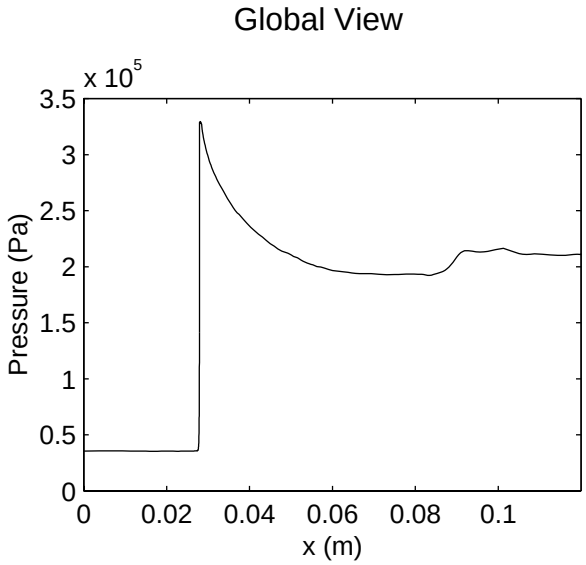
- $t = 230\ \mu s$
- predicted shock thickness: $\sim 50\ \mu m$.

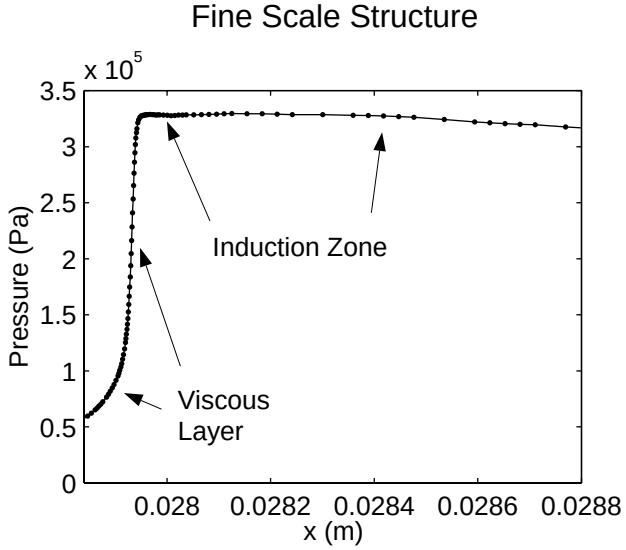


Viscous $H_2 - O_2$ Ignition Delay with Wavelets

Global and Fine Scale Structures

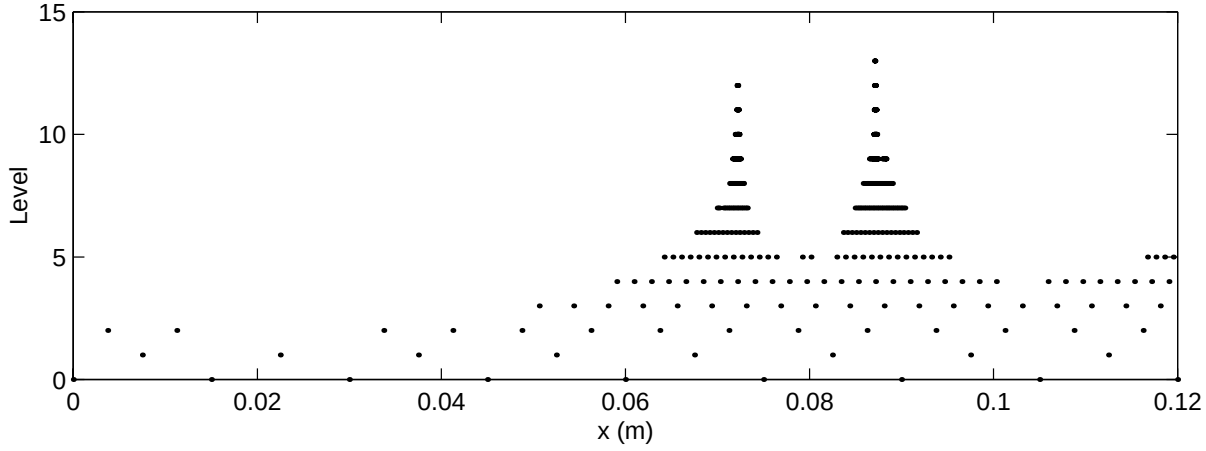
- $t = 230\ \mu s$

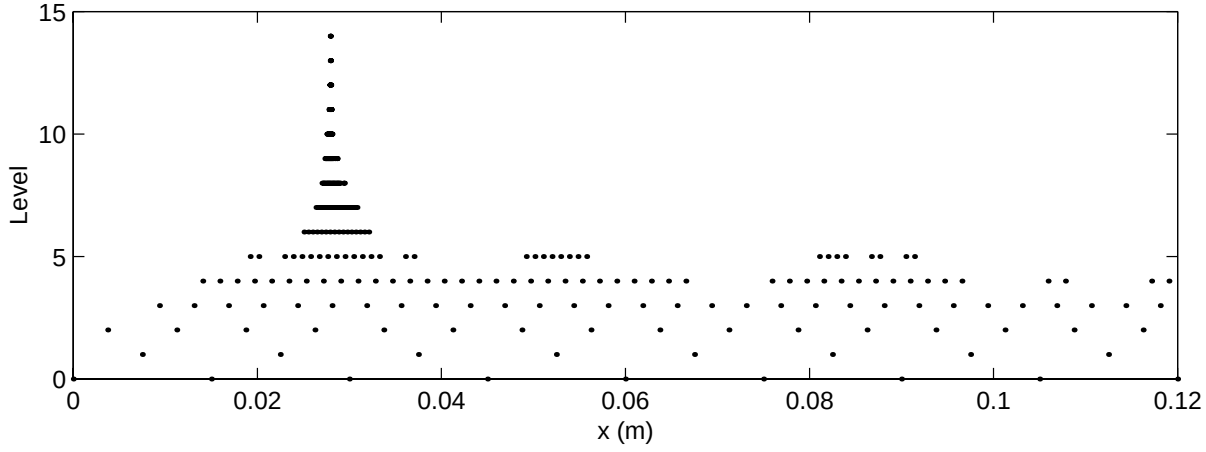




Viscous $H_2 - O_2$ Ignition Delay with Wavelets, Instantaneous Distributions of Collocation Points

- $t = 180\ \mu s$, two-shock structure with consequent collocation point distribution,
- $t = 230\ \mu s$, one-shock structure with evolved collocation point distribution.





Application to Gas Phase HMX System

- Equations for gas phase combustion of HMX are of form

$$\frac{\partial}{\partial t}\mathbf{q}(x,t) + \frac{\partial}{\partial x}\mathbf{f}(\mathbf{q}(x,t)) = \mathbf{g}(\mathbf{q}(x,t)),$$

- For *non-premixed* problem, higher dimension ($\geq$ 8?!) manifolds necessary!

- Will need to parameterize by ($h, \rho, H, O, N, C, Ar,$ $\geq$ one free parameter)

$$10^7 < h < 10^{11}\, erg/g; 10^{-5} < \rho < 10^{-3}\, g/cm^3; 10^{-32} < \chi_{Ar} < 10^{-2};$$

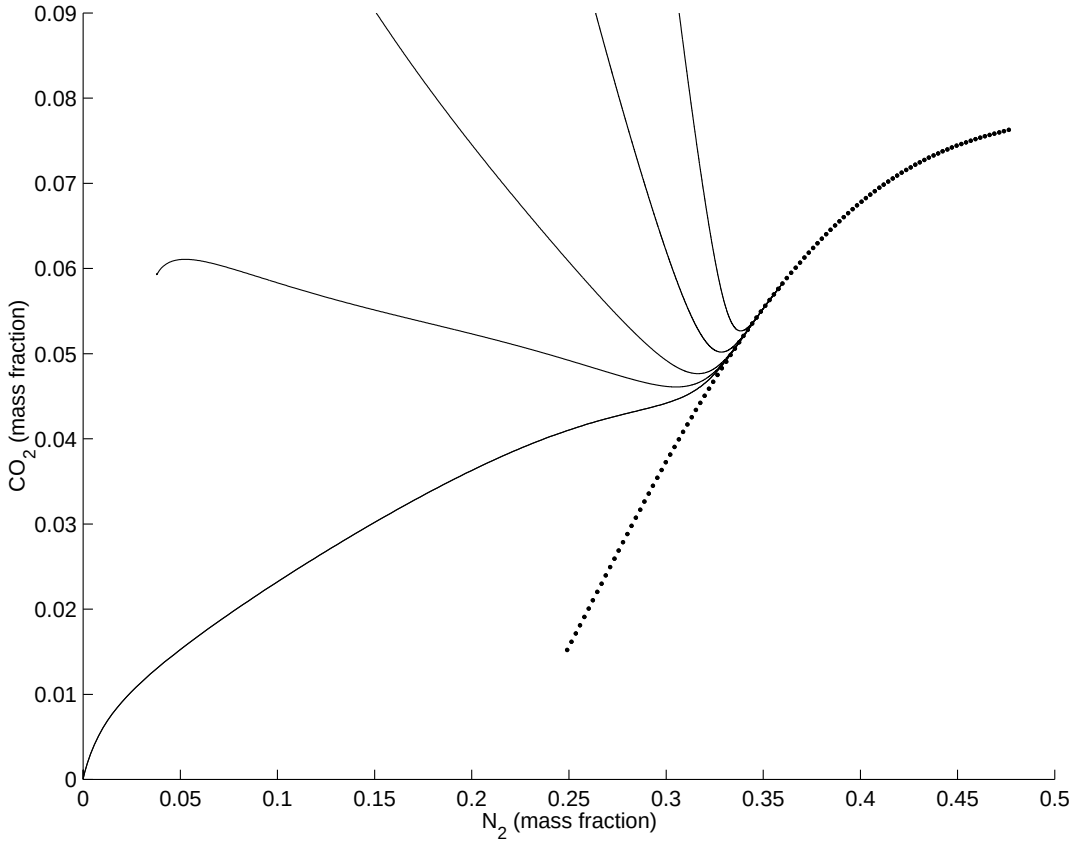
$$0 < \chi_C < 10^1; 0 < \chi_H < 10^1; 0 < \chi_N < 10^1; 0 < \chi_O < 10^1.$$

(Liau, 1999)

- We will start with a premixed problem with $Le = 1$ to maintain constant element concentration.

ILDM for Gas Phase HMX System

- Based on 45 species, 232 step mechanism of Yetter, et al.,
- Adiabatic ($h = 62 \times 10^9\ erg/g$) , isobaric ($P = 32\ bar$),
- projection in Y_{N_2}, Y_{CO_2} plane.



Conclusions

- Accurate solutions for HMX gas phase chemistry with detailed kinetics will require high dimensional ILDMs coupled with an adaptive multilevel scheme for spatial resolution.
- Such schemes have been demonstrated on simpler systems and are being applied currently to HMX.