Slow Invariant Manifolds for Reactive-Diffusive Systems

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7th International Congress on Industrial and Applied Mathematics

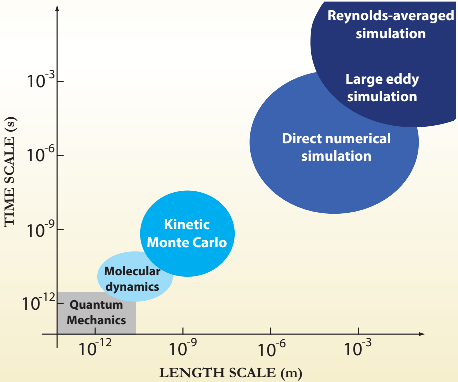
Vancouver, British Columbia, Canada
July 21, 2011

Outline

1. Motivation and Background
2. Model
3. Results
   - Oxygen Dissociation
   - Zel'dovich Mechanism
4. Conclusions

Motivation and Background

- Reactive systems induce a wide range of spatial and temporal scales, and subsequently severe stiffness
- DNS resolves all ranges of continuum physical scales present
- Under-resolved simulations attempt to account for missed physical phenomena with modeling
- Fully resolved simulations are expensive to compute

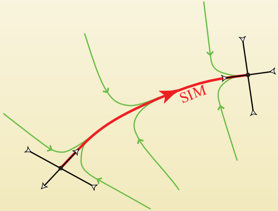


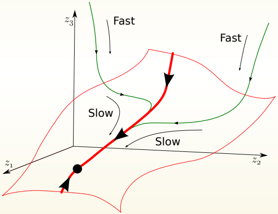
"Research needs for future internal combustion engines," *Physics Today*, Nov. 2008, pp 47-52.

Motivation and Background

- Manifold methods provide potential savings
- Most methods are for spatially homogeneous systems
- We employ the slow invariant manifold (SIM) model of Al-Khateeb, et al.

  (2009, *Journal of Chemical Physics*)





- We adjust for the dynamics of diffusion in the presence of weak spatial heterogeneity
- This is valid when diffusion is fast relative to reaction, i.e. thin regions of flames

Assumptions

Model a system of N species reacting in J reactions with diffusion in one spatial dimension

- Ideal mixture
- Calorically perfect
- Ideal gases
- Negligible advection
- Constant specific heat
- Single constant mass diffusivity
- Constant thermal conductivity

Balance Laws

- Evolution of species and energy

$$
\begin{aligned}
\rho \frac{\partial Y_i}{\partial t} + \frac{\partial j_i^m}{\partial x} &= \mathsf{M}_i \dot{\omega}_i(Y_n, T), \quad \text{for } i, n \in [1, N] \\
\rho \frac{\partial h}{\partial t} + \frac{\partial j^q}{\partial x} &= 0
\end{aligned}
$$

- Boundary conditions

$$
\begin{aligned}
\left.\frac{\partial Y_i}{\partial x}\right|_{x=0} = \left.\frac{\partial Y_i}{\partial x}\right|_{x=\ell} &= 0, \quad \text{for } i \in [1, N] \\
\left.\frac{\partial T}{\partial x}\right|_{x=0} = \left.\frac{\partial T}{\partial x}\right|_{x=\ell} &= 0
\end{aligned}
$$

- Initial conditions

$$
\begin{aligned}
Y_i(x, t=0) &= \tilde{Y}_i(x), \quad \text{for } i \in [1, N] \\
T(x, t=0) &= \tilde{T}(x)
\end{aligned}
$$

Constitutive Equations

- Simple diffusive flux terms

$$\begin{aligned} j_i^m &= -\rho \mathcal{D} \frac{\partial Y_i}{\partial x}, \quad \text{for } i \in [1, N] \\ j^q &= -k \frac{\partial T}{\partial x} + \sum_{i=1}^{N} h_i^f j_i^m \end{aligned}$$

- Caloric equation of state

$$h = \sum_{i=1}^{N} Y_i \left(c_{Pi}(T - T^o) + h_i^f \right)$$

- Ideal gas equation of state

$$P = \frac{\rho \bar{\mathfrak{R}} T}{\sum_{i=1}^{N} \frac{\mathsf{M}_i}{Y_i}}$$

Constitutive Equations

- Molar production rate

$$
\begin{aligned}
\dot{\omega}_i &= \sum_{j=1}^{J} \nu_{ij} r_j, \quad \text{for } i \in [1, N] \\
r_j &= k_j \left(\prod_{i=1}^{N} \left(\frac{\rho Y_i}{\mathsf{M}_i} \right)^{\nu'_{ij}} - \frac{1}{K_j^c} \prod_{i=1}^{N} \left(\frac{\rho Y_i}{\mathsf{M}_i} \right)^{\nu''_{ij}} \right), \quad \text{for } j \in [1, J] \\
k_j &= a_j T^{\beta_j} \exp\left(\frac{-\bar{E}_j}{\bar{\mathfrak{R}} T} \right), \quad \text{for } j \in [1, J] \\
K_j^c &= \exp\left(\frac{-\sum_{i=1}^{N} \bar{g}_i^o \nu_{ij}}{\bar{\mathfrak{R}} T} \right), \quad \text{for } j \in [1, J]
\end{aligned}
$$

Generalized Shvab-Zel'dovich

- Certain linear combinations of molar production rate sum to zero,

$$\frac{\partial}{\partial t}\left(\sum_{i=1}^{N}\varphi_{li}\frac{Y_i}{\mathsf{M}_i}\right)=\mathcal{D}\frac{\partial^2}{\partial x^2}\left(\sum_{i=1}^{N}\varphi_{li}\frac{Y_i}{\mathsf{M}_i}\right),\quad \text{for } l\in[1,L]$$

- In adiabatic systems, when the Lewis number is unity

$$\frac{\partial}{\partial t}\underbrace{\left(c_P(T-T^o)+\sum_{i=1}^{N}h_i^fY_i\right)}_{h}=\mathcal{D}\frac{\partial^2}{\partial x^2}\underbrace{\left(c_P(T-T^o)+\sum_{i=1}^{N}h_i^fY_i\right)}_{h}$$

- If initially spatially homogeneous, these PDEs can be integrated

$$\sum_{i=1}^{N}\varphi_{li}\frac{Y_i}{\mathsf{M}_i} = \sum_{i=1}^{N}\varphi_{li}\frac{\tilde{Y}_i}{\mathsf{M}_i},\quad \text{for } l\in[1,L]$$

$$c_P(T-T^o)+\sum_{i=1}^{N}h_i^fY_i = c_P(\tilde{T}-T^o)+\sum_{i=1}^{N}h_i^f\tilde{Y}_i$$

Reduced Variables

- Transform to specific mole concentrations

$$z_i = \frac{Y_i}{\mathsf{M}_i}, \quad \text{for } i \in [1, N-L]$$

- Evolution of remaining L species and temperature are coupled to these reduced variables by the algebraic constraints

$$\begin{aligned}
\frac{\partial z_i}{\partial t} &= \frac{\dot{\omega}(z_n, T)}{\rho} + \mathcal{D}\frac{\partial^2 z_i}{\partial x^2}, \quad \text{for } i, n \in [1, N-L] \\
T &= \begin{cases} \tilde{T}, & \text{if isothermal} \\ \dfrac{h - \sum_{i=1}^{N} \hat{z}_i(z_n)\bar{h}_i^f}{\sum_{i=1}^{N} \hat{z}_i(z_n)\bar{c}_{Pi}} + T^o, & \text{if adiabatic} \end{cases}
\end{aligned}$$

Galerkin Reduction to ODEs

- Assume a spectral decomposition

$$z_i(x,t) = \sum_{m=0}^{\infty} z_{i,m}(t)\phi_m(x), \quad \text{for } i \in [1, N-L]$$

- Orthogonal basis functions, $\phi_m(x)$, are eigenfunctions of diffusion operator that match boundary conditions

$$\phi_m(x) = \cos\left(\frac{m\pi x}{\ell}\right), \quad \text{for } m \in [0, \infty)$$

- Finite system of ODEs for amplitude evolution are recovered by taking the inner product with ϕ_n, and truncated at M

$$\frac{dz_{i,m}}{dt} = \frac{\langle \phi_m, \dot{\omega}_i\left(\sum_{m=0}^{\infty} z_{i,n}\phi_n\right)\rangle}{\langle \phi_m, \phi_m\rangle} - \frac{m^2\pi^2\mathcal{D}}{\ell^2} z_{i,m}, \qquad \text{for } i \in [1, N-L], \text{ and } m \in [0, M]$$

- Diffusion time scale identified, $\tau_{\mathcal{D}} = \dfrac{\ell^2}{\pi^2\mathcal{D}}$

Oxygen Dissociation

$$O + O + M \quad \leftrightharpoons \quad O_2 + M$$

- $N = 2$ species
- $J = 1$ reactions
- $L = 1$ constraints
- $N - L = 1$ reduced variables
  $z = z_O$

- Isochoric,
  $\rho = 1.6 \times 10^{-4}\ g/cm^3$
- Isothermal, $T = 5000\ K$

Partial differential equation governing evolution

$$\frac{\partial z}{\partial t} = 249.84130 - 74734.78\ z^2 - 172406.48\ z^3 + \mathcal{D}\frac{\partial^2 z}{\partial x^2}$$

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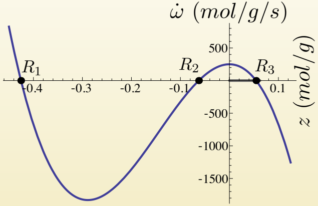


Spatially homogeneous evolution equation

$$\frac{dz_0}{dt} = 249.84130 - 74734.78\ z_0^2 - 172406.48\ z_0^3$$

Diffusion-Correction – Galerkin Projection

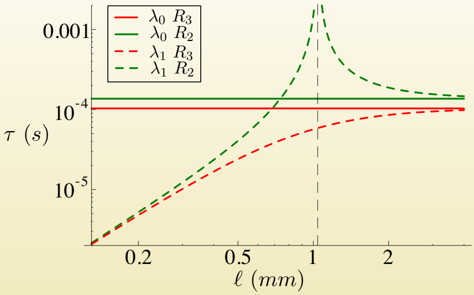
- One spatial mode ($M = 1$) evolution equation

$$\frac{dz_0}{dt} = 249.84130 - 74734.78\left(z_0^2 + \frac{z_1^2}{2}\right) - 172406.48\left(z_0^3 + \frac{3z_0 z_1^2}{2}\right)$$

$$\frac{dz_1}{dt} = -74734.78\,(2z_0 z_1) - 172406.48\left(3z_0^2 z_1 + \frac{3z_1^3}{4}\right) - \frac{\pi^2 \mathcal{D}}{\ell^2} z_1$$

- Spatially homogeneous evolution when $z_1 = 0$
- Spatially homogeneous equilibria retained
- Eigenvalues about these equilibria are modified

$$\lambda_1 = \lambda_0 - \frac{\pi^2 \mathcal{D}}{\ell^2}$$



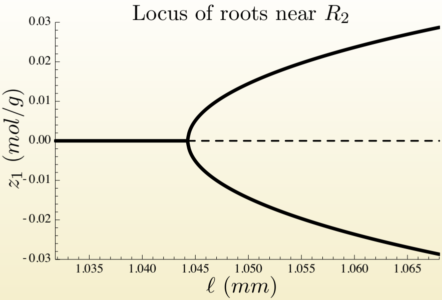
Bifurcation

- Change in sign of modified eigenvalue, $\lambda_1 = \lambda_0 - \frac{\pi^2 \mathcal{D}}{\ell^2}$, identifies a critical length where SIM origin changes character

- Bifurcation occurs at R_2 equilibrium

$$\frac{\pi^2 \mathcal{D}}{\ell^2} = \lambda_0 \quad = \quad 7321.5 \ s^{-1}$$
$$\ell \quad = \quad 1.04 \ mm$$

- Diffusion-corrected SIM origin shifts to bifurcated branches



- Bold branches are saddles; dashed branch is source

Poincaré Sphere

Map variables into a space where infinity is on the unit circle

$$\eta_0 = \frac{\alpha z_0}{\sqrt{1+\alpha^2 z_0^2 + \alpha^2 z_1^2}}$$

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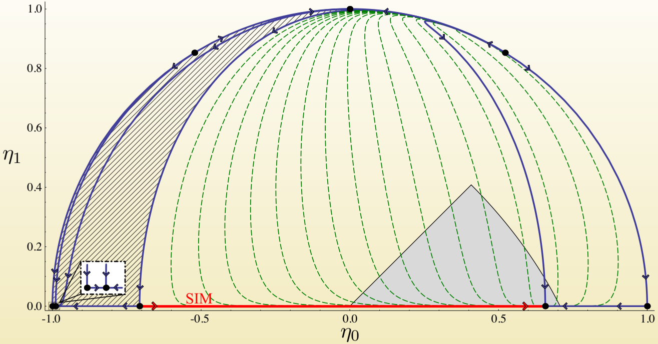
$\ell = 0.334\ mm$



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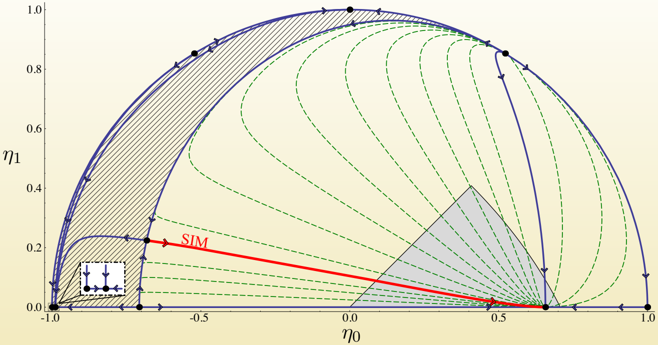
$\ell = 1.05\ mm$



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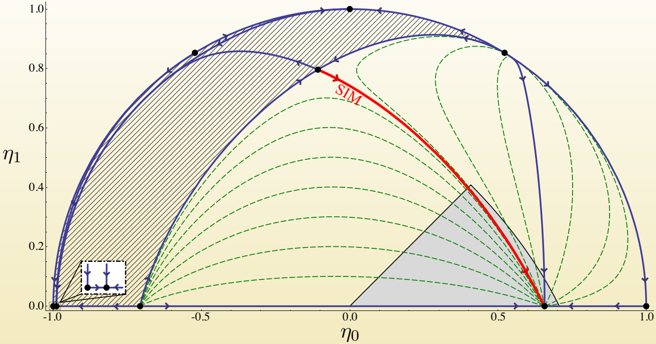
$\ell = 3.34\ mm$



Zel'dovich Mechanism – Isothermal

$$N + NO \leftrightharpoons N_2 + O$$
$$N + O_2 \leftrightharpoons NO + O$$

- $N = 5$ species
- $J = 2$ reactions
- $L = 3$ constraints
- $N - L = 2$ reduced variables
  $z_1 = z_{NO},\ z_2 = z_N$

- Isochoric, $\rho = 1.2002\ g/cm^3$
- Isothermal, $T = 4000\ K$
- Bimolecular, isobaric, $P = 1.6629 \times 10^6\ dyne/cm^2 = 1.64\ atm$

Partial differential equation governing evolution

$$\frac{\partial z_1}{\partial t} = 250 - 9.97\times10^4 z_1 + 1.16\times10^7 z_2 - 3.22\times10^9 z_1 z_2 + 6.99\times10^8 z_2^2 + \mathcal{D}\frac{\partial^2 z_1}{\partial x^2}$$
$$\frac{\partial z_2}{\partial t} = 250 + 8.47\times10^4 z_1 - 1.17\times10^7 z_2 - 1.84\times10^9 z_1 z_2 - 6.98\times10^8 z_2^2 + \mathcal{D}\frac{\partial^2 z_2}{\partial x^2}$$

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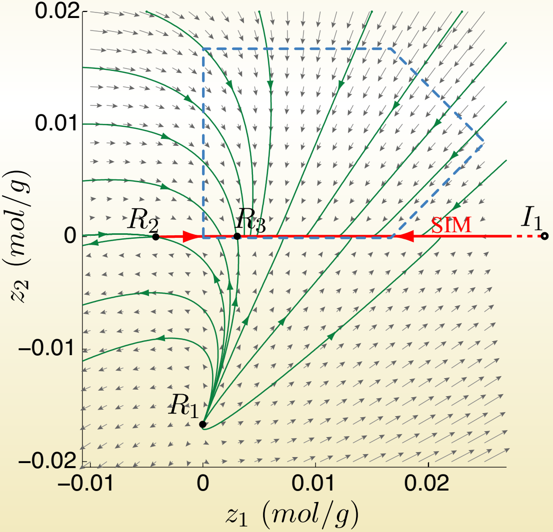
Spatially homogeneous evolution equations – second order polynomials.

$$\frac{dz_{1,0}}{dt} = 250 - 9.97 \times 10^4 z_{1,0} + 1.16 \times 10^7 z_{2,0} - 3.22 \times 10^9 z_{1,0} z_{2,0} + 6.99 \times 10^8 z_{2,0}^2$$
$$\frac{dz_{2,0}}{dt} = 250 + 8.47 \times 10^4 z_{1,0} - 1.17 \times 10^7 z_{2,0} - 1.84 \times 10^9 z_{1,0} z_{2,0} - 6.98 \times 10^8 z_{2,0}^2$$

Spatially Homogeneous Isothermal Phase Space

- Identify equilibria
- Characterize equilibria by eigenvalues of their Jacobian matrix
- Classify time scales as fast and slow
- Identify SIM as a heteroclinic orbit from saddle to sink



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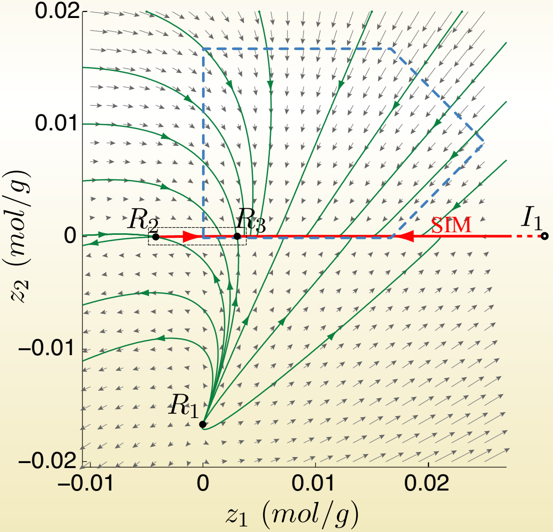

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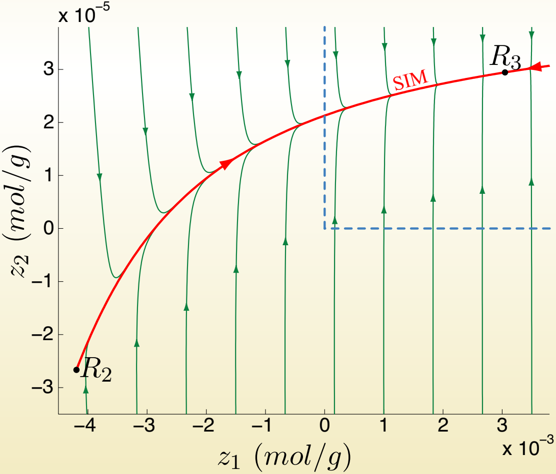
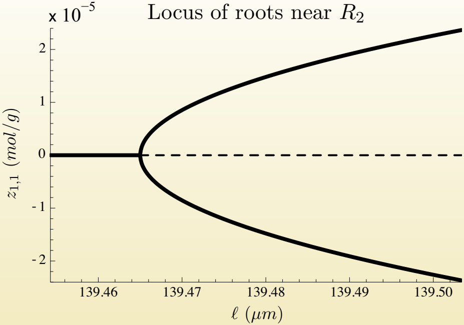
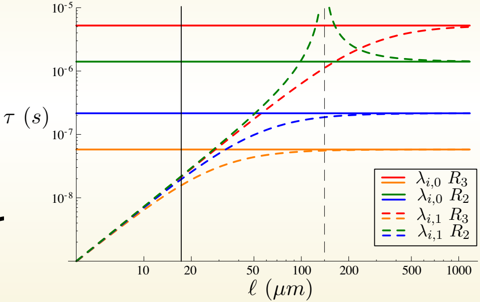


Diffusion-Correction – Galerkin Projection

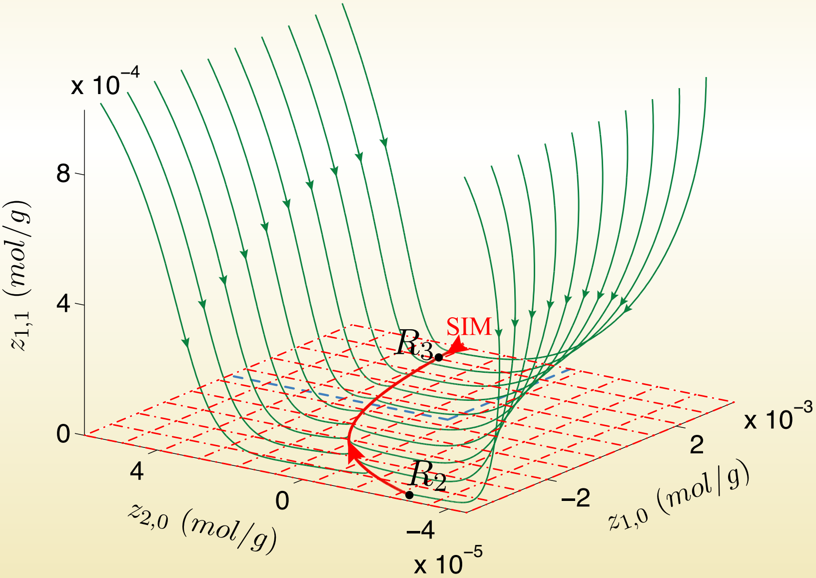
- First diffusion mode adds modified time scale
- Positive eigenvalue identifies critical length scale



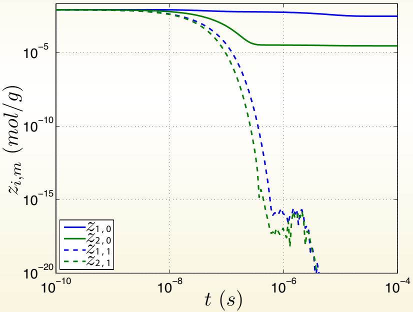


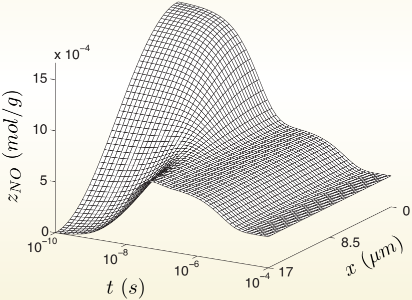
- Bifurcation occurs at this length scale
- Let us examine a length below this critical length scale, $\ell = 17\ \mu m$

Diffusion-Correction Isothermal Phase Space



Diffusion-Correction Isothermal Evolution





- Two additional fast time scales from diffusion
- Spatially inhomogeneous amplitudes decay earlier than either reaction time scale

Zel'dovich Mechanism – Adiabatic

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- $N = 5$ species
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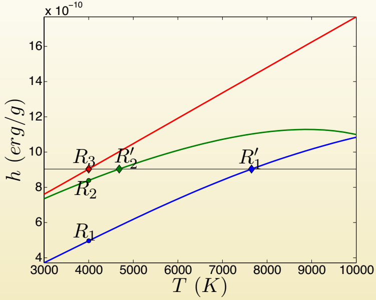
- $L = 3$ constraints
- $N - L = 2$ reduced variables
  $z_1 = z_{NO}, \; z_2 = z_N$
- $T = T(z_1, z_2)$

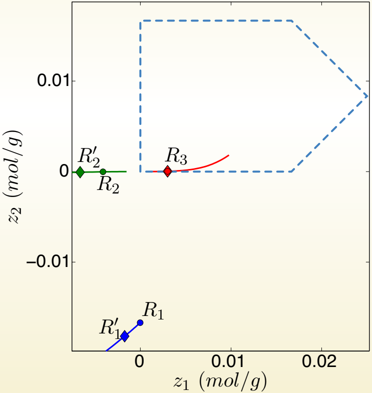
- Isobaric,
  $P = 1.6629 \times 10^6 \; dyne/cm^2$
  $= 1.64 \; atm$
- Adiabatic,
  $h = 9.0376 \times 10^{-10} \; erg/g$
  chosen to keep chemical equilibrium at same point

Evolution equations highly nonlinear due to temperature-dependance in exponentials

Isothermal Equilibria as a Function of Temperature

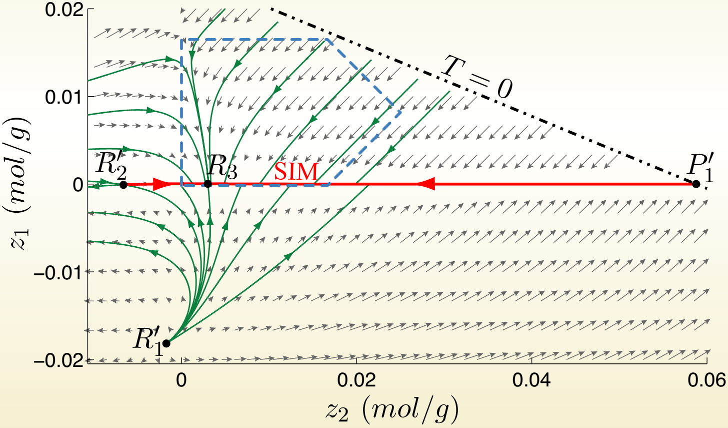
- Equilibria of adiabatic system difficult to identify
- Find isothermal equilibria for various temperatures





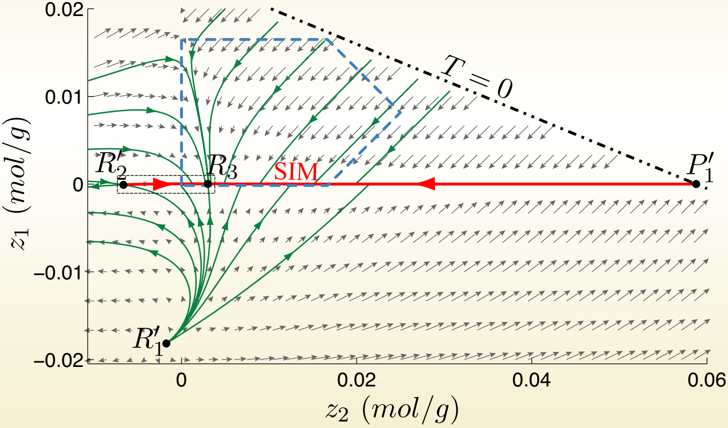
- Adiabatic equilibria where enthalpy constraint is met
- Method is not exhaustive

Spatially Homogeneous Adiabatic Phase Space



- Equilibria and dynamics remain similar to isothermal case
- SIM is heteroclinic orbit connecting analogous points

Spatially Homogeneous Adiabatic Phase Space



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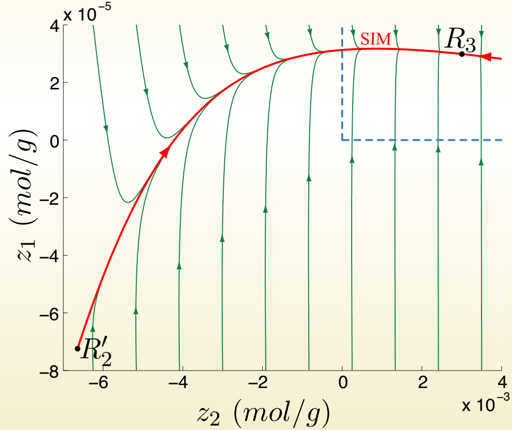

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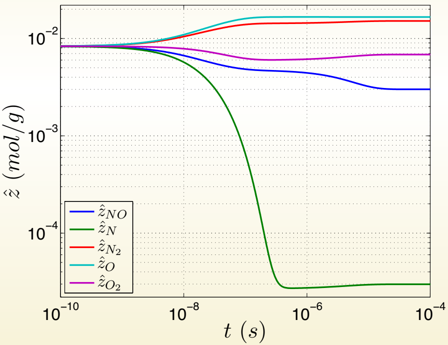
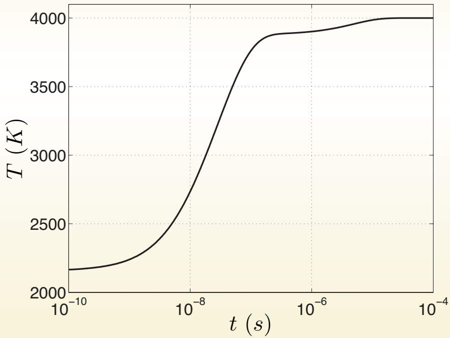
Spatially Homogeneous Adiabatic Evolution





- Species and temperature evolution exhibit fast and slow time scales, consistent with equilibrium eigenvalues
- Adiabatic reactive-diffusive systems have yet to be analyzed

Conclusions

- The SIM isolates the slowest dynamics, making it ideal for a reduction technique
- A critical length scale has been identified where the diffusion time scale matches a reaction time scale; at this length a bifurcation occurs that affects the slow dynamics of the system
- For sufficiently short length scales, diffusion time scales are faster than reaction time scales, and the system dynamics are dominated by reaction
- When lengths are near or above the critical length, diffusion plays a more important role
- Extension of SIM to spatially homogeneous adiabatic systems is shown to be feasible

Acknowledgments

Partial support provided by NSF Grant No. CBET-0650843,
Notre Dame ACMS Department Fellowship, and SIAM travel grant