Towards a predictive combustion chemistry model – Uncertainty propagation and minimization

Hai Wang and David A. Sheen

University of Southern California, Los Angeles, California, USA
Model Hierarchy

- **Aliphatics**
  - $\text{CH}_4$
  - $\text{CH}_x\text{O}_y$, $\text{C}_{2-4}\text{H}_x$ Oxidation
  - $\text{CO-H}_2$ Oxidation
  - $\text{H}_2$ Oxidation

- **Aromatics**

**Products**
- $\text{H}_2\text{O}$, $\text{CO}_2$
- $\text{H}_2$, $\text{CO}$
- $\text{C}_2\text{H}_2$, soot
- ....
Reaction Model Development

The Current Approach

proliferation of models

Critical review of literature kinetic data

Model compilation

“Tuning”

Validation

Publish model

The Current Approach

The Current Approach
Kinetic Rate Parameter Uncertainties

H + O₂ ⇌ OH + O (R1)

- Uncertainty factor ~1.25
- Logarithmic sensitivity coefficient = 0.24 (ethylene-air, f= 1, p = 1 atm)
- ±5% (±4 cm/s) uncertainty in predicted flame speed due to R1 alone
- Key question: How do we propagate uncertainties in rate constants in combustion simulations?

Baulch, et al. (2005)
Burke, et al. (2010)
Uncertainty

Rate parameter uncertainties

Physical problem

Data uncertainty

Fundamental reaction kinetics expt + calc

Fundamental combustion expt.
MUM-PCE

- Method of Uncertainty Minimization – Polynomial Chaos Expansions

- Model prediction presented as a (2-s) band of uncertainty resulting from kinetic parameter uncertainties.
- Model uncertainty may be constrained by experimental data (ignition delay, species-time history, flame speeds etc)
MUM-PCE: Methods

- **Stochastic Spectral Expansion**: express kinetic parameter $x_i$ as a polynomial expansion of basis random variables

  $$x_i = x_i^{(0)} + \sum_{j=1}^{m} \alpha_{ij} \xi_j + \sum_{j=1}^{m} \sum_{k=1}^{m} \beta_{ijk} \xi_j \xi_k + ...$$


- **Solution Mapping**: use polynomial response surface to express the relation between a combustion response $h$ and $x$

  $$\eta_r(x) \equiv \eta_{r,0} + \sum_{i=1}^{N} a_{r,i} x_i + \sum_{i=1}^{N} \sum_{j \geq i}^{N} b_{r,ij} x_i x_j$$
Forward Uncertainty Propagation

\[ \eta_r(x) = \eta_{r,0} + \sum_{i=1}^{n} a_i x_i + \sum_{i=1}^{n} \sum_{j \geq i}^{n} b_{ij} x_i x_j \]

Response surface from solution mapping

\[ x_i = \frac{1}{2} \xi_i \]

Spectral representation of uncertainty in x’s
(mean = 0, s = 0.5, each indep’t of others)

\[ \eta_r(x, \xi) = \eta_r \left( x^{(0)} \right) + \sum_{i=1}^{M} \hat{\alpha}_{r,i} \xi_i + \sum_{i=1}^{M} \sum_{j=i}^{M} \hat{\beta}_{r,ij} \xi_i \xi_j \]
Solution Mapping Method

• Fit a response surface to the model

G.E.P. Box, et al. (1978), Frenklach et al. (1992), S.G. Davis et al. (2004)
MUM-PCE – Application in H₂/O₂ Combustion

- High-pressure data sensitise kinetics of hydrogen oxidation.

- A large number of models outside experimental uncertainty at high pressures.

Burke, et al. (2010)

- 2σ uncertainty band calculated by MUM-PCE, based on rate parameter uncertainties.

- Models are statistical samples of parameter uncertainties.

Sheen & Wang (2011)
rate parameter uncertainties

Physical problem

data uncertainty

fundamental reaction kinetics expt + calc

fundamental combustion expt.
MUM-PCE

rate parameter uncertainties

Physical problem

data uncertainty

fundamental reaction kinetics expt + calc

fundamental combustion expt.
Method of Uncertainty Minimization

\[ x = x_0 + \alpha \xi \]

Chemical model + associated uncertainty

\[ \eta_r(x) \equiv \eta_{r,0} + \sum_{i=1}^{N} a_{r,i} x_i + \sum_{i=1}^{N} \sum_{j \geq i}^{N} b_{r,ij} x_i x_j \]

Physics model

\[ \eta_r(x, \xi) = \eta_r(x^{(0)}) + \sum_{i=1}^{m} \alpha_{r,i} \xi_i + \sum_{i=1}^{m} \sum_{j=i}^{m} \beta_{r,ij} \xi_i \xi_j \]

Predictions + associated uncertainty

\[ \Phi(x_0^*) = \min_{x_0} \left\{ \sum_{r=1}^{M} \left[ \eta_{r,0} - \eta_r(x_0) \right]^2 \left( \frac{\sigma_{x_0}}{\sigma_r} \right)^2 + \sum_{n=1}^{N} \left( x_{0,n} \right)^2 \right\} \]

\[ \Sigma = \left[ \sum_{r=1}^{n} \frac{1}{(\sigma_r)^2} \left( bx_0^* b + ax_0^* a + b^T \xi_0 a^T + a^T \right) + 4I \right]^{-1} \]

\[ \alpha^* = \Sigma^{1/2} \]
MUM-PCE – Application in H₂/O₂ Combustion

- Model uncertainty constraining
- JetSurF 2.0 H₂/CO submodel
  - 14 species, 41 reactions

**Dataset 1:** From Davis, *et al.* (2005):

<table>
<thead>
<tr>
<th></th>
<th>No.</th>
<th>( P_0, P_5 ) (atm)</th>
<th>( T_0, T_5 ) (K)</th>
<th>( f )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laminar Flame Speeds</td>
<td>12</td>
<td>1-15</td>
<td>298</td>
<td>1.0-3.0</td>
</tr>
<tr>
<td>Ignition Delay Times</td>
<td>13</td>
<td>0.5-33</td>
<td>1000-2600</td>
<td>1.0-6.1</td>
</tr>
<tr>
<td>Flow Reactor Profiles</td>
<td>9</td>
<td>1.0-16</td>
<td>915-1040</td>
<td>0.3-1.0</td>
</tr>
<tr>
<td>Laminar Flame Profiles</td>
<td>2</td>
<td>0.047</td>
<td>400</td>
<td>1.9</td>
</tr>
</tbody>
</table>

**Dataset 2:**

<table>
<thead>
<tr>
<th></th>
<th>No.</th>
<th>( P_0, P_5 ) (atm)</th>
<th>( T_0, T_5 ) (K)</th>
<th>( f )</th>
</tr>
</thead>
</table>
MUM-PCE – Application in H₂/O₂ Combustion

Considering no experiments

Dataset 1 (knowledge prior to 2010)

Dataset 1+2 (latest knowledge)

Sheen & Wang (2011)
MUM-PCE – Application in $\text{H}_2/\text{O}_2$ Combustion

$\text{H}_2/\text{O}_2/\text{He}$ mixtures at equivalence ratio 1

$\text{H}_2/\text{O}_2/\text{Ar}$ mixtures at equivalence ratio 2.5

Sheen & Wang (2011)
MUM-PCE – Application in H₂/O₂ Combustion

Dataset 1
Knowledge prior to 2010

+ Burke, et al. (2010)
Current knowledge

Weak constraint by experiments
Strong constraint by experiments
JetSurF – A Jet Surrogate Fuel Model

JetSurF is a detailed chemical reaction model for the combustion of jet-fuel surrogate. The model is being developed through a multi-university research collaboration and is funded by the Air Force Office of Scientific Research. Project participants include:

F. N. Egolfopoulos, Hai Wang  
University of Southern California

R. K. Hanson, D. F. Davidson, C. T. Bowman, H. Pittsch
Stanford University

C. K. Law
Princeton University

N. P. Cernansky, D. L. Miller
Drexel University

W. Tsang
National Institute of Standards and Technology

R. P. Lindstedt
Imperial College, London

A. Violi
University of Michigan

New Release: JetSurF Version 2.0 – A working model for the combustion of n-alkane up to n-dodecane, cyclohexane, and mono-alkylated cyclohexane up to n-butyl-cyclohexane  
(Release Date: September 19, 2010)

Old Releases: JetSurF Version 1.1 – A interim model for the combustion of n-butyl-, n-propyl-, ethyl-, and methyl-cyclohexane and cyclohexane  
(Release Date: September 15, 2009)
JetSurF Validation – Species Concentrations behind reflected shock waves


Plot stolen from Ron Hanson. Solid line: experiments; dashed line: JetSurF
Prediction Uncertainties in As-Compiled Model

Good nominal prediction with significant uncertainty!
Chemistry Model & Experimental Targets

- Modified JetSurF 1.0
  - 196 species, 1478 reactions

<table>
<thead>
<tr>
<th></th>
<th>No.</th>
<th>$P_0$, $P_5$ (atm)</th>
<th>$T_0$, $T_5$ (K)</th>
<th>$f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laminar Flame Speeds</td>
<td>4</td>
<td>1</td>
<td>353</td>
<td>0.8-1.4</td>
</tr>
<tr>
<td>Ignition Delay Times</td>
<td>11</td>
<td>1-4</td>
<td>1000-2600</td>
<td>0.5-2</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>No.</th>
<th>$P_5$ (atm)</th>
<th>$T_5$ (K)</th>
<th>$f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>OH, H$_2$O, CO$_2$, C$_2$H$_4$, CH$_3$ Species Profiles</td>
<td>11</td>
<td>1.6-2.4</td>
<td>1365-1545 K</td>
<td>1</td>
</tr>
</tbody>
</table>

No. = Number of simulations, $P_0$, $P_5$ (atm) = Initial and final pressures (atm), $T_0$, $T_5$ (K) = Initial and final temperatures (K), $f$ = Range of values.
Predictions of As-Compiled and Uncertainty-Minimized Models

Unconstrained

Constrained

Mole Fraction

Time (s)

Mole Fraction

Time (s)

Mole Fraction

Time (s)

Mole Fraction

Time (s)

Mole Fraction

Time (s)
Effect on Flame Speed Predictions

Considering no experiments

Model constrained by species profiles

Model constrained by species profiles + flame speeds
Effect on Flame Speed Predictions

Uncertainty in Species Value, $2\sigma^{\text{obs}}$

- $\sigma_s$ (cm/s)
- $1/(2\sigma^{\text{obs}})$

- CH$_3$ (Series 2) only
- OH (Series 1) only
- All multi-species (Series 1 & 2)
What did uncertainty minimization do?

Model constrained by species profiles

Model constrained by flame speeds

CH₃, CH₂, secondary chain branching, fuel breakup

H chain branching
What did uncertainty minimization do?
"Our" Approach

Review of literature kinetic data

Model compilation

Electronic structure calculations

Reaction rate theory

New experiments

Sensitivity analysis

Response surface development

Uncertainty minimization

Validation

Take home messages:

• Consistency is more critical than predictability!
• Need a comprehensive model to truly reflect the inherent model hierarchy and uncertainty.
Acknowledgements

Previous students/postdocs
• Xiaoqing You
• Baptiste Sirjean

Current students/postdocs
• David Sheen
• Enoch Dames
• Bing yang

Collaborators
• Stephen Klippenstein (ANL)
• Chung-King Law (Princeton)
• Fokion Egolfopoulos (USC)
• Elke Goos (DLR)

The JetSurF team
• Ron Hanson (Stanford)
• Tom Bowman (Stanford)
• Heinz Pitsch (Stanford)
• Wing Tsang (NIST)
• Angela Violi (UMich)
• Peter Lindstedt (Imperial Col.)
• Nick Cernansky (Drexel)
• David Miller (Drexel)

Financial Support
AFOSR, AFRL, SERDP, DOE, NSF