Chemical Kinetics and Transport of Combustion Processes

Hai Wang
B. Yang, J. Camacho, E. Dame, S. Lieb, S. Memarzadeh, S.-K. Gao and S. Koumlis

University of Southern California
Quick Summary

• Sooting properties of $n$- and $i$-butanol flames.
  – Provide an extended database for the oxidation kinetics of butanol isomers.
  – Complement the MBMS measurements of butanol flame structures.

• Methods of Uncertainty Minimization – Polynomial Chaos Expansions (MUM-PCE).
  – Application in foundational fuel model development.

• Molecule-particle crossed beams for diffusion cross section studies.
Sooting Behaviors of $n$- and $i$-Butanol Flames

Experimental Methods

- Premixed, burner-stabilized stagnation flame (BSSF) approach
- Probe sampling followed by mobility sizing of incipient soot particles
- Detailed temperature mapping and simulation

Burner-stabilized stagnation flame approach

Abid et al. (2009)
Sooting Behaviors of $n$- and $i$-Butanol Flames

$n$-C$_4$H$_9$OH  

$i$-C$_4$H$_9$OH  

$n$-C$_4$H$_{10}$  

$i$-C$_4$H$_{10}$

<table>
<thead>
<tr>
<th></th>
<th>n-C$_4$H$_9$OH</th>
<th>i-C$_4$H$_9$OH</th>
<th>n-C$<em>4$H$</em>{10}$</th>
<th>i-C$<em>4$H$</em>{10}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mole fraction (fuel/O$_2$/Ar)</td>
<td>0.118/0.282/0.600</td>
<td>0.109/0.290/0.600</td>
<td>0.096/0.304/0.600</td>
<td>0.096/0.304/0.600</td>
</tr>
<tr>
<td>C/O</td>
<td>0.69</td>
<td>0.63</td>
<td>0.63</td>
<td>0.63</td>
</tr>
<tr>
<td>Equivalence ratio, $\phi$</td>
<td>2.5</td>
<td>2.25</td>
<td>2.05</td>
<td>2.05</td>
</tr>
<tr>
<td>Cold gas velocity, $v_0$ (cm/s)</td>
<td>4.6</td>
<td>4.6</td>
<td>3.6</td>
<td>3.6</td>
</tr>
<tr>
<td>Flame temperature, $T_f$ (K)</td>
<td>1740</td>
<td>1790</td>
<td>~1750</td>
<td>~1750</td>
</tr>
</tbody>
</table>

- $n$-C$_4$H$_{10}$ and $i$-C$_4$H$_{10}$ flames as references.
- Keeping C/O and flame temperature constant.
- Measured temperature profiles show very similar flame conditions among flames studied.

![Graph showing temperature profiles](image-url)
Sooting Behaviors of $n$- and $i$-Butanol Flames

- Radiation correction by detailed modeling (OPPDIF/USC Mech II).

- Experimental data and model are in good agreement.
Sooting Behaviors of \( n \)- and \( i \)-Butanol Flames

- Incipient soot show a strong and persistent nucleation behavior well into the post flame.
- Under comparable condition, \( i \)-\( C_4H_9OH \) flame yields as much soot and at a rate the same as \( i \)-\( C_4H_{10} \) flame.
Sooting Behaviors of $n$- and $i$-Butanol Flames

- Incipient soot show a strong and persistent nucleation behavior well into the post flame.
- Under comparable conditions, $i$-$C_4H_9OH$ flame yields as much soot and at a rate the same as $i$-$C_4H_{10}$ flame.
- Chemical structure of the fuel has a bigger impact on soot chemistry than the presence of OH group in the fuel.
Burke, et al. (2010)
Burke, et al. (2010)
Uncertainty

rate parameter uncertainties

fundamental reaction kinetics
expt + calc

Physical problem

data uncertainty

fundamental combustion expt.
Uncertainty

rate parameter uncertainties

Physical problem

data uncertainty

fundamental reaction kinetics expt + calc

fundamental combustion expt.
• **Method of Uncertainty Minimization – Polynomial Chaos Expansions**


  • Model prediction presented as a \((2-\sigma)\) band of uncertainty resulting from kinetic parameter uncertainties.

  • Model uncertainty may be constrained by experimental data (ignition delay, species-time history, flame speeds etc)
rate parameter uncertainties

Physical problem

data uncertainty

fundamental reaction kinetics expt + calc

MUM-PCE

fundamental combustion expt.
MUM-PCE

- Method of Uncertainty Minimization – Polynomial Chaos Expansions

- Model prediction presented as a (2-σ) 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

• Method of Uncertainty Minimization – Polynomial Chaos Expansions

• Model uncertainty constrained by reducing the coupled uncertainty of rate parameters.
MUM-PCE – Application in $\text{H}_2/\text{O}_2$ Combustion

- High-pressure data sensitize kinetics of hydrogen oxidation.
- A large number of models outside experimental uncertainty at high pressures.

Burke, et al. (2010)

Sheen & Wang (2011)

- $2\sigma$ uncertainty band calculated by MUM-PCE, based on rate parameter uncertainties.
- Models are statistical samples of parameter uncertainties.
MUM-PCE – Application in H\textsubscript{2}/O\textsubscript{2} 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>No.</th>
<th>$P_0$, $P_5$ (atm)</th>
<th>$T_0$, $T_5$ (K)</th>
<th>$\phi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laminar Flame Speeds</td>
<td>12</td>
<td>1-15</td>
<td>298</td>
</tr>
<tr>
<td>Ignition Delay Times</td>
<td>13</td>
<td>0.5-33</td>
<td>1000-2600</td>
</tr>
<tr>
<td>Flow Reactor Profiles</td>
<td>9</td>
<td>1.0-16</td>
<td>915-1040</td>
</tr>
<tr>
<td>Laminar Flame Profiles</td>
<td>2</td>
<td>0.047</td>
<td>400</td>
</tr>
</tbody>
</table>

**Dataset 2:** From Burke, *et al.* (2010):

<table>
<thead>
<tr>
<th>No.</th>
<th>$P_0$, $P_5$ (atm)</th>
<th>$T_0$, $T_5$ (K)</th>
<th>$\phi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laminar Flame Speeds</td>
<td>18</td>
<td>15-25</td>
<td>298</td>
</tr>
</tbody>
</table>
MUM-PCE – Application in $\text{H}_2/\text{O}_2$ Combustion

Considering no experiments

Dataset 1 (knowledge prior to 2010)

Dataset 1+2 (latest knowledge)

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

H₂/O₂/He mixtures at equivalence ratio 1

H₂/O₂/Ar mixtures at equivalence ratio 2.5

Sheen & Wang (2011)
Dataset 1
Knowledge prior to 2010

Strong constraint by experiments

Weak constraint by experiments

Current knowledge

MUM-PCE – Application in $\text{H}_2/\text{O}_2$ Combustion
MUM-PCE – Application in H₂/O₂ Combustion

H₂/Air Perfectly-Stirred Reactor, 20 atm, T = 1400 K

Current Knowledge

No constraining
MUM-PCE – Future Work

• Apply the method to Dryer’s H$_2$/O$_2$ model – as the foundation of foundational fuel chemistry.
  • remaining uncertainties and their impact on C1 chemistry.

• Make the code available to the CEFRC community.

• Methodology development
  • Experimental design – condition space, uncertainty requirement, etc
  • Extend beyond A-factors – $k(T,P)$, transport coefficients, vibrational frequency, critical energy, $<E_{\text{down}}>$ etc