Growth mechanism for soot primary particles in recirculating hydrocarbon flames

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Abstract

The project seeks to expand the growth mechanism for soot primary particles in recirculating hydrocarbon flames to include an additional step: the soot primary particle (SPP) is carbonized in the recirculating flame. This mechanism includes the roles of reevaporation, coalescence, diffusion, and coagulation. The SPP is then carbonized in the recirculating flame and captured as soot primary particles (SPRs).

Table 1. Experimental conditions for CH₄ diffusion flame

<table>
<thead>
<tr>
<th>Condition</th>
<th>Upright</th>
<th>IGFR with no Recirculation</th>
<th>IGFR with Recirculation</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH₄ Flow Rate (lpm)</td>
<td>0.13</td>
<td>0.24</td>
<td>0.13</td>
</tr>
<tr>
<td>Air Flow Rate (lpm)</td>
<td>1.8</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>Reactor Orientation</td>
<td>Upright</td>
<td>Inverted</td>
<td>Inverted</td>
</tr>
<tr>
<td>Flame Length (mm)</td>
<td>80.5</td>
<td>60.0</td>
<td>27.2</td>
</tr>
<tr>
<td>Residence Time (s)</td>
<td>0.06-0.01</td>
<td>0.3-0.8</td>
<td>0.5-3.0</td>
</tr>
</tbody>
</table>

Figure 2. Three methane diffusion flames studied. Soot at the base of the recirculating flame is observed flowing from the flame front back inside of the high temperature fuel rich region.

Table 2. Morphology of soot collected from the above methane diffusion flames

<table>
<thead>
<tr>
<th>Diameter (nm)</th>
<th>Number of Particles</th>
<th>Fractal Dimension</th>
</tr>
</thead>
<tbody>
<tr>
<td>33.4±3.9</td>
<td>150</td>
<td>1.68-1.69</td>
</tr>
<tr>
<td>25.2±4.1</td>
<td>240</td>
<td>1.68-1.70</td>
</tr>
<tr>
<td>68.0±7.9</td>
<td>354</td>
<td>1.69-1.70</td>
</tr>
</tbody>
</table>

Figure 3. SEM images of soot from the three flames shown in Figure 2. Recirculation increases d₅₃ for soot particles. Scale bars represent 500 nm.

Table 3. Morphology of soot collected from ethylene diffusion flame

<table>
<thead>
<tr>
<th>Diameter (nm)</th>
<th>Ethylene</th>
<th>IGFR</th>
<th>Upright</th>
</tr>
</thead>
<tbody>
<tr>
<td>39.3±5.5</td>
<td>53.7±8.4</td>
<td>53.7±8.4</td>
<td>53.7±8.4</td>
</tr>
<tr>
<td>Number of Particles</td>
<td>403</td>
<td>150</td>
<td></td>
</tr>
<tr>
<td>Fractal Dimension</td>
<td>1.66-1.67</td>
<td>1.58-1.61</td>
<td></td>
</tr>
</tbody>
</table>

Figure 4. Ethylene diffusion flame soot. A recirculating flame creates fractals with a consistently larger d₅₃. Left: SEM image of soot produced in upright ethylene flame with d₅₃, typically reported range. Right: SEM image of soot produced in ethylene IGFR recirculating flame.

Table 5. Resulting mole fraction of important growth species inside PSR1 for two mechanisms

<table>
<thead>
<tr>
<th></th>
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</tr>
</thead>
<tbody>
<tr>
<td>Mole Fraction C₂H₄</td>
<td>1.23E-02</td>
</tr>
<tr>
<td>Mole Fraction A²</td>
<td>5.01E-05</td>
</tr>
<tr>
<td>Mole Fraction A³</td>
<td>2.51E-06</td>
</tr>
</tbody>
</table>

Figure 6. Left: CFD showing recirculating pathlines and resulting residence time. Particles in the entrainment have residence times up to 20 seconds. Middle: Probability distribution of residence times. Right: The surrounding oxygen and temperature of a single particle entrained for 15 seconds inside the recirculation region. The particle oscillates between rich and lean regions of the reaction.

Figure 7. CRN created from experimental observations and CFD simulations. The PSR is composed of three main components, a PSR modeling the fuel rich region conducive to particle growth, a second PSR at stoichiometric conditions that carbonizes and solidifies soot particles, and a recirculation region that carries soot from the flame front back inside of the fuel rich region.

Conclusions

- Recirculation of soot particles from oxygen rich to fuel rich regions leads to larger d₅₃.
- Re-exposure to growth region inside flame front with C₂H₄ and PAHs after carbonization and restructuring at the flame front is the reason for larger d₅₃.
- Recirculation increases d₅₃ for multiple gas-phase hydrocarbon fuels (methane, ethylene, propane) and changes soot morphology.

Acknowledgments

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References


Chemical Reaction Network

- CRN based on CFD results and experimental observations for the qualitative representation of growth species
  - Recirculation region from fuel lean to fuel rich, nitrogen profile, stagnation point at flame front, and residence time
  - Ability to tweak results to match observable results
  - Composed of two perfectly stirred reactors (PSRs) which assume diffusion is infinitesimally small compared to chemical kinetics
  - Chemical kinetics follow Arrhenius relationships for C1 and C2 diffusion flames

Numerical

- Two dimensional axisymmetric CFD modeling provides gas flow, temperature, and species profiles in the combustor
- Basic chemistry to be expanded with additional CRN

Soot Formation Process

- Pyrolysis
- PAH Formation
- Clustering
- Coalescence/Surface Growth
- Carbonization at the flame front
- Coagulation

Project Objectives

Hypothesis: Repeated particle exposure to the gas and liquid growth species in the fuel rich zone after particle carbonization in the flame front leads to surface growth by formation of liquid hydrocarbon film on the surface followed by solidification of particle due to hydrogen abstraction.

1. Experimentally observe and measure flame conditions leading to particle growth with gaseous C₃H₈ fuels
2. Analyze size and structure of nanoparticles
3. Model IGFR to gain insight on variables that play a major role in particle growth
4. Distinguish between recirculating and other flame conditions on particle formation