Quantitative Thermochemical Measurements in High-Pressure Gaseous Combustion

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Abstract
We present our strategic experiment and thermochemical analyses on combustion flow using a subframe burst gating (SBG) Raman spectroscopy. This unconventional laser diagnostic technique has promising ability to enhance accuracy of the quantitative scalar measurements in a point-wise single-shot fashion. In the presentation, we briefly describe an experimental methodology that generates transferable calibration standard for the routine implementation of the diagnostics in hydrocarbon flames. The diagnostic technology was applied to simultaneous measurements of temperature and chemical species in a swirl-stabilized turbulent flame with gaseous methane fuel at elevated pressure (17 atm). Statistical analyses of the space-/time-resolved thermochemical data provide insights into the nature of the mixing process and its impact on the subsequent combustion process in the model combustor.

“Advanced Engine Diagnostics”
July 12-13, 2012
GE Global Research Center
Niskayuna, NY
Quantitative Thermochemical Measurements in High-Pressure Gaseous Combustion

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July 12-13, 2012 @ GE Global Research Center
**GOAL**
Provide accurate quantitative scalar data for “benchmark tests” for CFD combustor code.

**STRATEGY**
- Advance a point-wise single-shot laser Raman diagnostics
- A series of experiments:
  1. Calibration on reference burners;
  2. Testing on realistic burner;
  3. Increase chemical complexity.

**TECHNICAL CHALLENGE**
- Simultaneous measurements: temperature, major species, mixture fraction.
- Accuracy: uncertainty <5%.
- Optically-harsh environment: high pressure, geometric limitation, optical interference; two-phase flows.
Facility

SE-5 High-Pressure Turbulent Combustion Facility
- Pressure up to 30 atm
- Gaseous and liquid fueled combustion
- Advanced laser diagnostics
- Air can be preheated to 1000F

APCD Combustion Diagnostics Facility
- Diagnostic development & calibration
Linear Raman Diagnostics

- Multi-Scalar Detection w/ Single Laser System
- Weak Scattering
- Raman Spectroscopy
- Quantitative
- Non-Intrusive

HIGH-POWER PULSED LASER
- Sensitivity
- Signal visibility
- Plasma sparking
- Optics damage
Developed a patent-pending optical gating scheme: *Subframe Burst Gating (SBG)*.

First-ever single-shot polarization-resolved Raman spectroscopy in liquid-fueled combustion, that enabled *interference (noise)-free* scalar measurements.

Significantly improved signal visibility (5 times) in combustion while eliminating a need of a conventional mechanical shutter for gating.
On-Chip Subframe Burst Gating*

Sensor area \( (N \times N \text{ pixels}) \)

Illuminated area \( (n \times N \text{ pixels}) \)

No illumination

Charge flow direction

Readout

1st laser pulse (vert. pol.)

2nd laser pulse (horiz. pol.)

\( t = t_1 \)

\( t = t_1 + \Delta t_{\text{shift}} \)

Shift (<5 \( \mu \text{s} \))

Raman scattering + Optical background

Subtraction

Optical background only (flame emission, LIFs, etc)

‘True’ Raman signal

*Patent Application #12/893,627 (filed on 09-29-10)
Single-shot background-free measurement

- A fuel-rich n-heptane flame
- A pair of orthogonally-polarized Nd:YAG 532-nm pulsed lasers (650 mJ/pulse)
Calibration Experiment (I)

**Raman Spectra**

CH$_4$/air flames, fuel-rich, Hencken burner, 500-shot averaged

**Species:**

*Super-pixel Integration*

**Temperature:**

Stokes/anti-Stokes ratio of N$_2$

$$ R_{SAS} = \left( \frac{v_0 + v_s}{v_0 - v_s} \right) \exp \left( - \frac{h \cdot c \cdot v_s}{k \cdot T} \right) $$

$n_0$: Excitation frequency (cm$^{-1}$); $n_s$: Raman shift (cm$^{-1}$); $h$: Planck's constant (J/s); $c$: Speed of light (cm/s); $k$: Boltzmann constant (J/K); $T$: Temperature (K)

<table>
<thead>
<tr>
<th>Species</th>
<th>Integration limits (nm)</th>
<th>Integration Width (nm)</th>
<th>Integration Width (pixels)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO$_2$</td>
<td>565.5, 578.0</td>
<td>12.5</td>
<td>48</td>
</tr>
<tr>
<td>O$_2$</td>
<td>578.0, 581.7</td>
<td>3.7</td>
<td>14</td>
</tr>
<tr>
<td>CO</td>
<td>596.7, 601.7</td>
<td>5.0</td>
<td>19</td>
</tr>
<tr>
<td>N$_2$</td>
<td>601.8, 608.4</td>
<td>5.4</td>
<td>24</td>
</tr>
<tr>
<td>CH$_4$ ($v_1$)</td>
<td>625.4, 631.9</td>
<td>6.5</td>
<td>25</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>644.9, 662.1</td>
<td>17.2</td>
<td>66</td>
</tr>
<tr>
<td>H$_2$</td>
<td>662.2, 685.1</td>
<td>22.9</td>
<td>84</td>
</tr>
</tbody>
</table>
Calibration Experiment (II)

- Diagnostic thermometry cross-check (premixed CH\textsubscript{4}/air flat-flame)
- Stokes/anti-Stokes (SAS) ratio of N\textsubscript{2} vibrational Q-branch band vs. CARS (Meier, DLR)
Calibration Experiment (III)

Temperature profiles of the reference standard (calibration) gaseous flames: $\text{H}_2/\text{air}$ flat-flame; $\text{CH}_4/\text{air}$ flat-flame; $\text{C}_7\text{H}_{16}/\text{air}$ flat-flame.
Calibration Experiment (IV)

- Complete transferable Raman calibration matrix: empirical/theoretical calibration coefficients to determine the major species concentration.
- Corrections of crosstalk between O₂ and CO₂.
Calibration Experiment (V): calibration matrix

\[ S_i = \Lambda E_L k_{i,j}(T) N_i \]

\[
k_{i,j} = \begin{pmatrix}
    f(T)_{O_2} & 0 & f(T)_{H_2 \rightarrow O_2} & 0 & f(T)_{CO_2 \rightarrow O_2} & 0 \\
    0 & f(T)_{N_2} & f(T)_{H_2 \rightarrow N_2} & 0 & 0 & 0 \\
    0 & 0 & f(T)_{H_2} & 0 & 0 & 0 \\
    0 & f(T)_{N_2 \rightarrow CO} & f(T)_{H_2 \rightarrow CO} & f(T)_{CO} & 0 & 0 \\
    f(T)_{O_2 \rightarrow CO_2} & 0 & f(T)_{H_2 \rightarrow CO_2} & 0 & f(T)_{CO_2} & 0 \\
    0 & 0 & f(T)_{H_2 \rightarrow H_2O} & 0 & 0 & f(T)_{H_2O} \\
\end{pmatrix}
\]

\[ f(T)_i = a_0 + a_1 T + a_2 T^2 + a_3 T^3 + \ldots + a_n T^n, \text{ or} \]
\[ f(T)_i = b_0 \exp(b_1 T) \]

- \( N_i \): Number density, species \( i \)  
- \( \Lambda \): Optical throughput/efficiency  
- \( E_L \): Excitation laser energy  
- \( k_{i,j} \): Raman calibration matrix element  
- \( S_i \): Raman scattering signal (super-pixel count)

<table>
<thead>
<tr>
<th>( k_{ij} )</th>
<th>( a_0 )</th>
<th>( a_1 )</th>
<th>( a_2 )</th>
<th>( a_3 )</th>
<th>( a_4 )</th>
<th>( a_5 ) or ( b_0 )</th>
<th>( a_6 ) or ( b_1 )</th>
<th>Cal. flame</th>
</tr>
</thead>
<tbody>
<tr>
<td>( CO_2 )</td>
<td>-9.6516E1</td>
<td>1.1611E2</td>
<td>-2.9599E-2</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>CH₄(rich)</td>
</tr>
<tr>
<td>( O_2 )</td>
<td>3.0887E4</td>
<td>-3.4452E1</td>
<td>1.2742E1</td>
<td>-1.9918E-4</td>
<td>1.6531E-7</td>
<td>-6.8439E-11</td>
<td>1.1196E-14</td>
<td>H₂(lean)</td>
</tr>
<tr>
<td>( CO )</td>
<td>2.5515E4</td>
<td>-1.3450E-1</td>
<td>7.0400E-4</td>
<td>-3.9523E-7</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>CH₄(rich)</td>
</tr>
<tr>
<td>( N_2 )</td>
<td>2.4826E4</td>
<td>-3.8542E-1</td>
<td>1.8054E-3</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>H₂</td>
</tr>
<tr>
<td>( CH_4 )</td>
<td>2.224E5</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>cold gas</td>
</tr>
<tr>
<td>( H_2O )</td>
<td>6.6698E4</td>
<td>-1.1590E1</td>
<td>6.7770E-3</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>H₂</td>
</tr>
<tr>
<td>( H_2 )</td>
<td>1.0072E5</td>
<td>2.0440</td>
<td>-1.5094E-2</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>H₂(rich)</td>
</tr>
<tr>
<td>( CO_2 \rightarrow O_2 )</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>4.2732E1</td>
<td>2.0524E-3</td>
<td>CH₄(rich)</td>
<td></td>
</tr>
<tr>
<td>( O_2 \rightarrow CO_2 )</td>
<td>-2.0669E3</td>
<td>1.1764E1</td>
<td>-1.9805E-2</td>
<td>1.2227E-5</td>
<td>-2.1984E-9</td>
<td>-</td>
<td>-</td>
<td>H₂(lean)</td>
</tr>
</tbody>
</table>
Calibration Experiment (V)

\[ N_i = \left( \frac{1}{\Lambda E_L} \right) k_{i,j}^{-1}(T) S_i \]

- True scalar value (compared w/ adiabatic chemical equilibrium): \( C_7H_{16} \) flat-flame test.
- Measurement accuracy:

<table>
<thead>
<tr>
<th>Species</th>
<th>( \text{CO}_2 )</th>
<th>( \text{O}_2 )</th>
<th>( \text{N}_2 )</th>
<th>( \text{CO} )</th>
<th>( \text{H}_2\text{O} )</th>
<th>( \text{H}_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uncertainty</td>
<td>2.1%</td>
<td>6.9%</td>
<td>1.4%</td>
<td>8.5%</td>
<td>1.0%</td>
<td>5.6%</td>
</tr>
</tbody>
</table>

Inverse matrix formula for N.D. determination.
Single-Cup LDI Burner for Validation

- Modified angled 6-jet (1 mm dia. each) gaseous fuel Lean-Direct Injection
- CFD friendly design
- Interchangeable between gaseous and liquid fuel injectors
- Swirler (1” cup): 60-deg and 45-deg.
- Latest run: $\Phi = 0.74$, $P = 250$ psia, 940 slm air.

Simulation Credit: C. Wey (RTB)
Raman Diagnostics Setup (point-wise):
New Dual-SBG Detection

**Anti-Stokes**
spectrometer (446-524 nm)

**Stokes**
spectrometer (559-694 nm)

- Nd:YAG (II) pulsed laser
  - 532nm, 8ns, 10Hz, <700 mJ/pulse
- Unpolarized pulse stretcher
  - ~75 ns
- Quartz Sphere
- Ant-Stokes spectrometer (446-524 nm)
- Stokes spectrometer (559-694 nm)
- e2v EMCCD (low-noise, ultrafast frame-transfer mode)
- Spectrograph
- Optical fiber
- Beam dump
- Shortwave-pass filter
- Longwave-pass filter
- Lens
- Burner
- e2v EMCCD
- Spectrograph
- Optical fiber
Raw spectral data (10-Hz repetition rate).

- Raman frequency signature → Chemical composition
- Random changes in intensity → Turbulent mixing
- Simultaneous measurements of multiple variables → Combustor code validation
Scalar Contour (averaged)

- Temp: ave. low 700 K; ave. high 1400 K.
- Side-spreading, low-profile flame (indicated by the temp).
- Majority of mixing within 8 mm height.
- Almost no residual fuel above 15 mm height.
- Predictable CO$_2$, H$_2$O (combustion product) profiles in post-flame zone.
Scalar Contour (standard deviation)

- Temp RMS shows the region with swirl-induced turbulent-chemistry interaction.
- Mixture fraction and fuel RMS support the idea.
Thermochemical Analysis (I)

Near fuel exit plane: 

$(x, r) = (2, 0) – (2, 16)$

- Highly turbulent (mixing) region indicated by the largest scatter in mixture fraction (scatter plots).
- Reaction incomplete (3-scalar correlation).
- Co-existence of cold fuel and oxidizer – unburnt pockets.
Thermochemical Analysis (II)

- Reaction zone: reached at the highest temp (approx. 2000K with the widest temp distribution (PDF/scatter plot).
- Unique bimodal distribution (PDF) – recirculation zone.
- Little fuel residual (3 scalar correlation).
Post-flame zone:
- homogeneous, well-reacted region indicated by normal (narrower) distribution.
- No residual fuel.
- Very little scatter in mixture fraction (end of fuel-air mixing)
Thermochemical Analysis (IV)

- Overall profile agreement with laminar flame calculation (UC Berkely) – precision of the measurement.
- Large number of samples (mixing and reaction zone) out of adiabatic equilibrium condition – nature of non-premixed turbulent flames.
- Dominant partially premixed combustion regime.
- A majority of the measured samples (at the post flame zone) indicated complete or near-complete reaction.
- Mixing-only conditions (i.e., preheated to below-ignition-point temperature) following the global equivalence ratio of the flame: Evidence of fast premixing.
High-Speed Raman Scattering Measurements

- Diagnostics for combustion dynamics and instability.
- Demonstration in a fuel-lean H₂-air flat-flame.
- Single-shot Raman spectra at 1 kHz data rate with a 527-nm DPSS Nd:YLF laser (30 mJ/pulse, 30W) and a high-speed image-intensified CCD camera.
- Trade-off: data rate vs. accuracy
- 10-kHz system under development.
Summary

✓ Significant upgrade to SE5 high-pressure turbulent combustion validation facility (nasa grc): available for code-validation experiments.
✓ Unconventional laser Raman diagnostics (double-SBG) is invented for routine operations.
✓ Generated one-of-the-kind quantitative multi-scalar data for swirl-stabilized combustion at 250 psia (17 atm).
✓ Our thermochemical analysis explored nature of the turbulent flame structure.