LIB Thermal Runaway and Combustion Research at Texas A&M University

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Presentation Outline

1) Part 1: Chemical Equilibrium Analyses: *A priori* prediction of thermal runaway consequences in LIBs
   - Overview and potential of CEA for LIB TR analysis
   - Representative modeling results and validation

2) Part 2: Fundamental Combustion Studies: Improving the state-of-the-art chemical kinetics for LIB electrolytes
   - Experimental methods – shock tubes, laminar flame speed vessels, optical diagnostics
   - Representative fundamental data and chemical kinetic modeling

3) Part 3: Ongoing/Future Work: Pipeline LIB thermal runaway and combustion projects at TAMU

Part 1: Chemical Equilibrium Analyses

A priori prediction of thermal runaway consequences in LIBs

LIB CEA Motivation

The current approach to LIB TR hazard analysis is not a priori and can be expensive

- **Problem Statement:** Significant energy, toxic gases, and potentially combustible gases are released during thermal runaway of LIBs, which all represent potential hazards

- **Current Approach:** Evaluation of these hazards by inducing electrolyte decomposition or LIB thermal runaway in abuse experiments

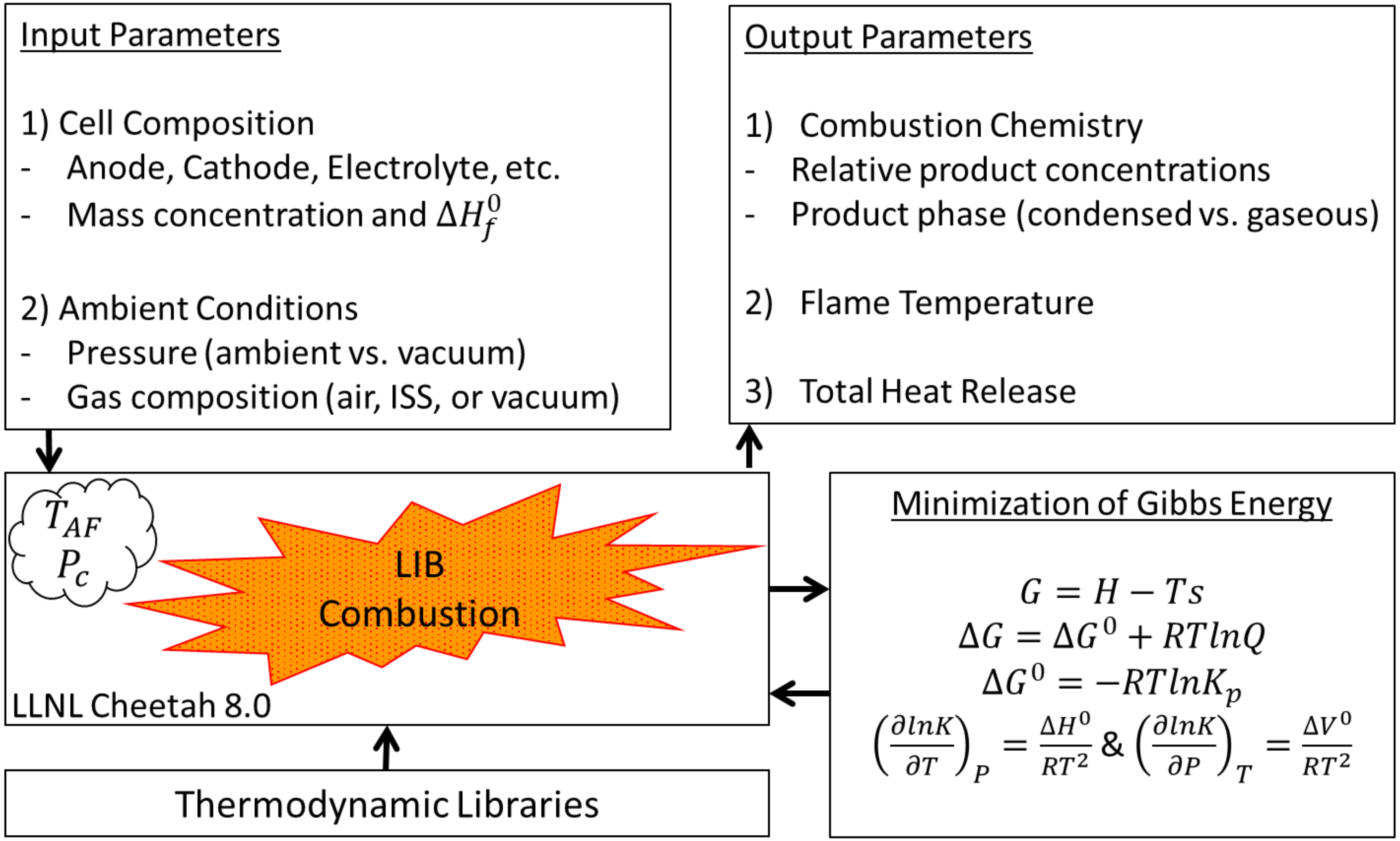
- **Objective:**
  1) Develop an a priori modeling approach
  2) Validate against existing experimental data
  3) Apply to various LIB chemistries, designs, conditions, etc.

Overview of Chemical Equilibrium Analysis (CEA)

CEA is utilized to predict reaction equilibrium conditions

Potential Impact:

- *a priori* modeling of TR events and consequences
- Inform experimental findings and conditions
- Aid in the design of LIB systems (chemistry, enclosures, atm, etc.)



Literature Summary

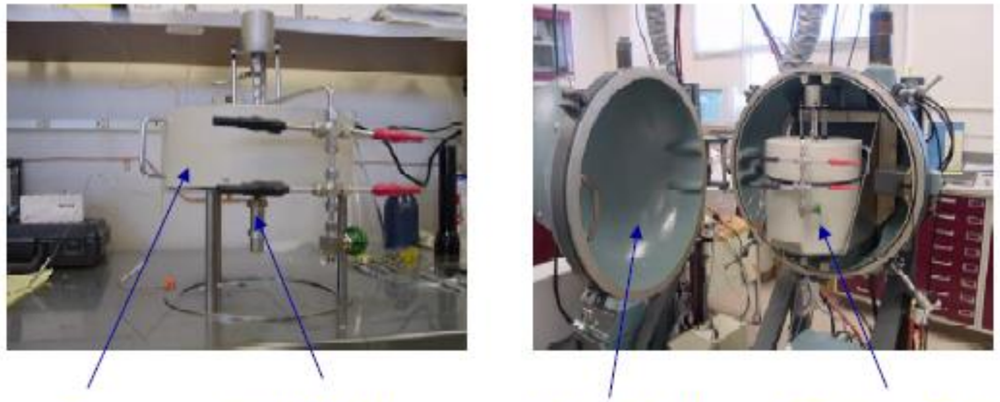
LIB TR hazard analysis is an active area of research

Literature Summary:

- Toxic gas release during LIB thermal runaway is a noteworthy hazard and an active area of research
- Several experimental approaches are currently being taken
  - ARC, cone calorimetry, closed vessel sampling
  - Current experimental data lacks ‘accurate’ cell compositions
  - Temperature ramp (dT/dt) is typically too slow
  - Very little modeling has been completed
  - [Golubkov et al., 2015] experiments are the best available

Sandia Experiments [Roth et al., 2004]
- DSC, ARC with temperature-transient gas sampling
- Reaction Gas Sampling
- Cell-level and sub-components

Shortcomings
- No ignition and combustion
- Gas sampling data is more useful for decomposition kinetics modeling



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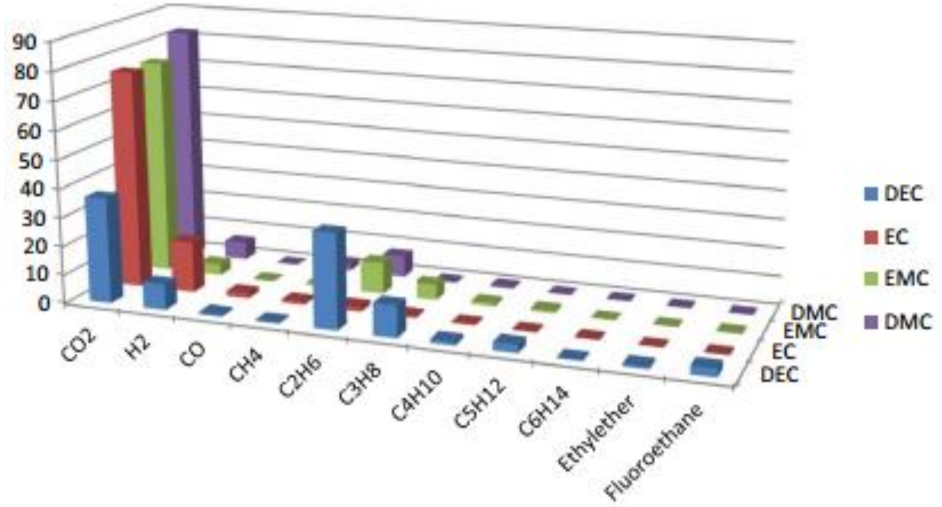
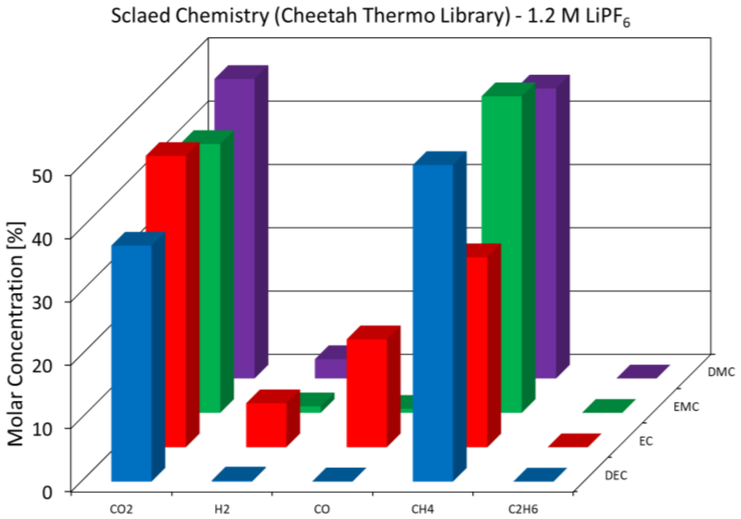
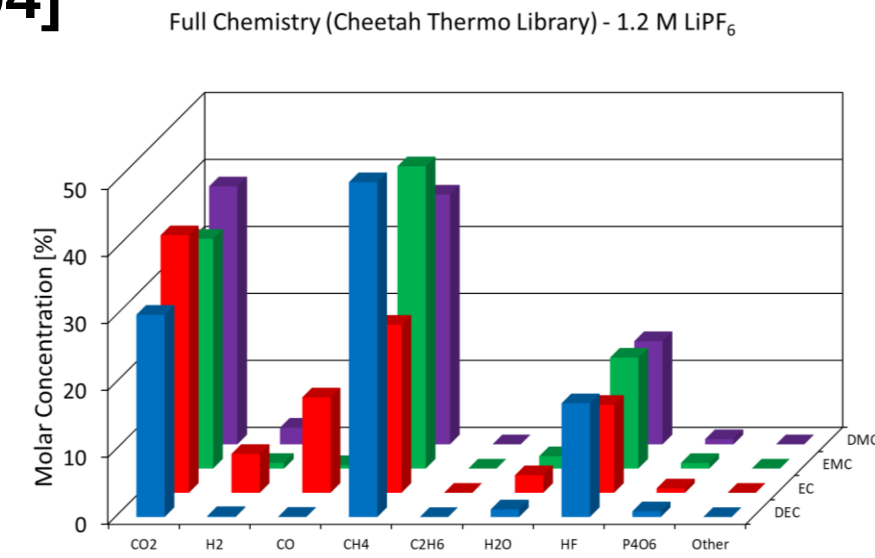
CEA Modeling – Example Results

Excellent agreement for plain electrolyte experiments, discrepancies for full LIB

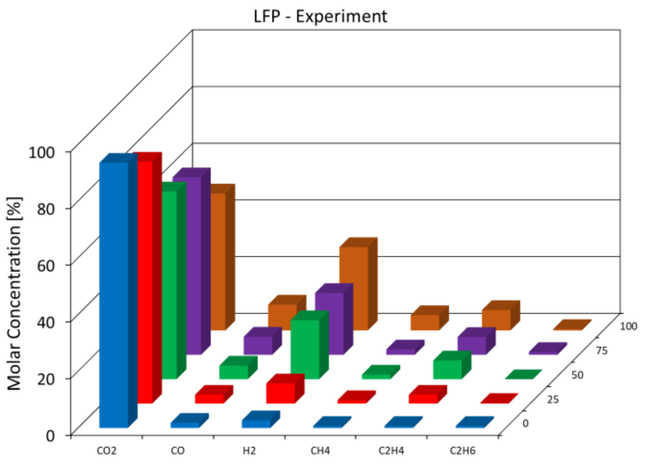
Electrolytes – Gas Production in ARC Experiments – [Roth et al., 2004]

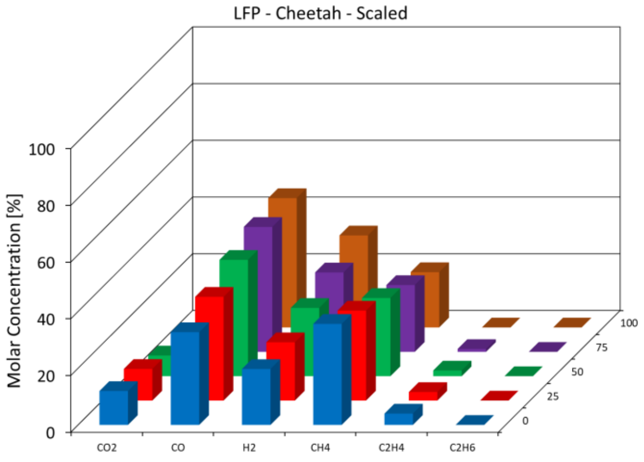


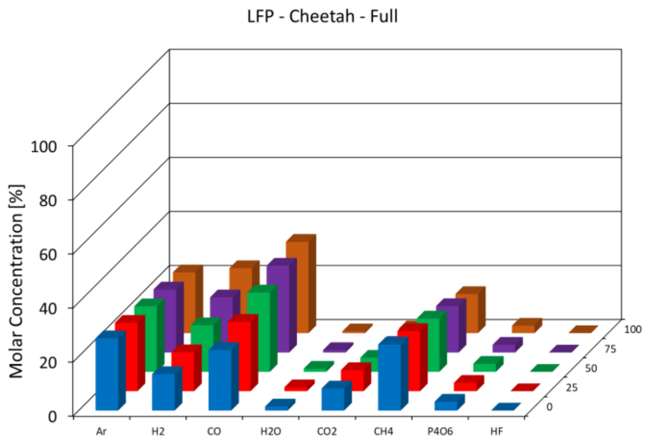


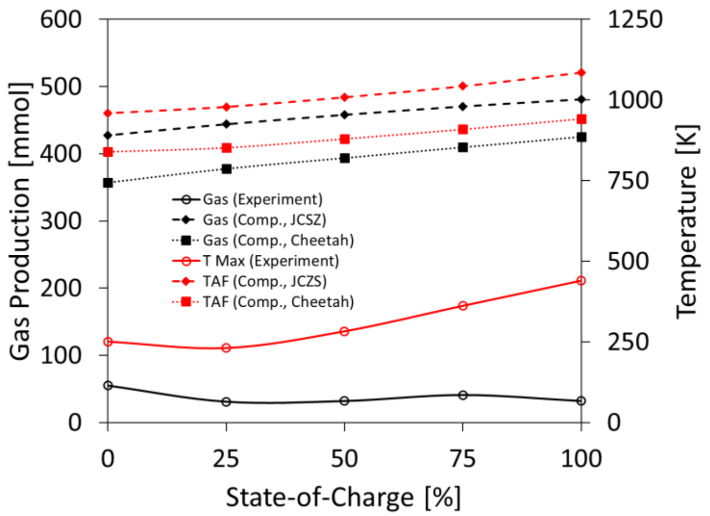


LFP LIB Cell – Gas Production in ARC Experiments – [Golubkov et al., 2015]









Summary and Future Work

- CEA modeling capability developed for TR of electrolytes and LFP batteries
  - Simple extension to other chemistries
  - Important computational products:
    - Product composition and chemistry
    - Reaction temperature
    - Total heat release
- Experimental and computational agreement is moderate
  - Need for rapid heating experiments with ‘well-characterized’ cells
  - Need for more restrictive modeling (cathode breakdown threshold)
- Future Work
  - Implementation of restrictive modeling inclusions
  - Validation via fast-heating experiments
  - Analyze effects of pressure, ambient composition, chemistry, etc.

Part 2: Fundamental Combustion Studies

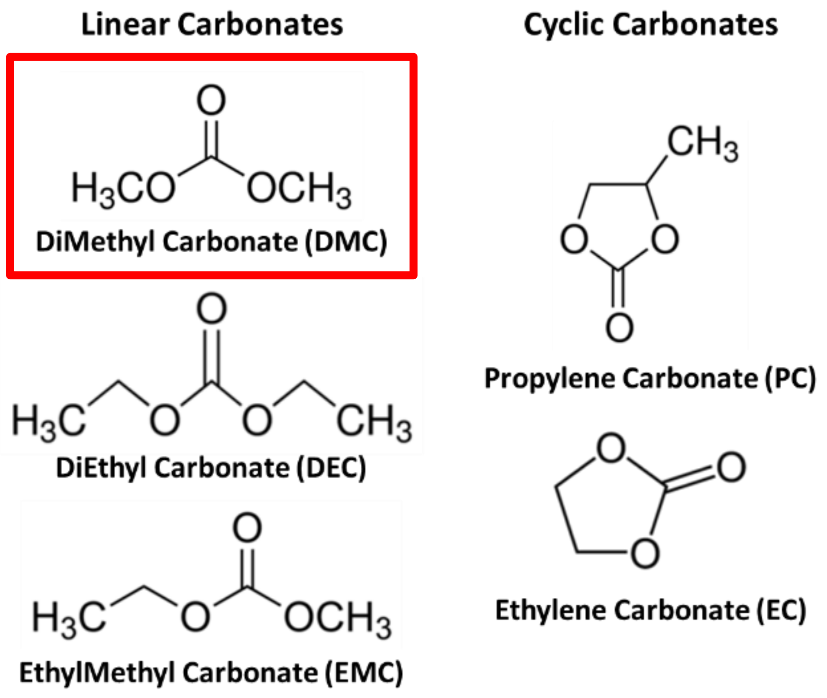
Improving the state-of-the-art chemical kinetics for LIB electrolytes

Motivation

The chemical kinetics of LIB electrolytes is an understudied topic

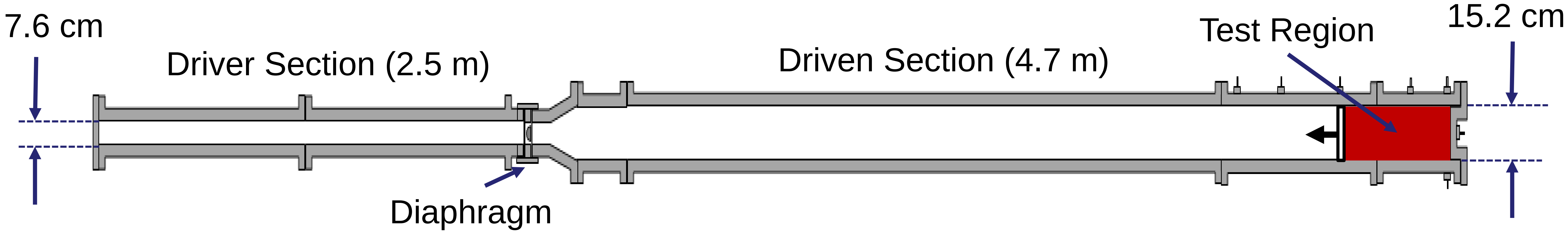
LIB Electrolytes:

- Essentially constituted of linear and cyclic carbonates
- DMC is a large component of electrolytes
- Most flammable/volatile component of LIBs
- LIB electrolyte chemical kinetics
  - Understudied topic, especially in plain form(s)
- Current Study: DMC chemical kinetics
  - pyrolysis (thermal decomposition) and oxidation (combustion)
  - Shock tube studies: $\phi = 0.5, 1.0, 2.0, and\ \infty; \mathrm{P} \sim 1.5\ atm$
    $T = 1230 - 2500\ K; Diluted\ (98 - 99.25\%)$
  - Laminar flame speed studies: $\phi = 0.7 - 1.5; \mathrm{P} = 1\ atm; T = 318, 363, and\ 423\ K$
  - Improved chemical kinetics model



Experimental Setup – Shock Tube

Shock tube experiments emulate extreme temperature/pressure conditions



<u>Shock Tube Fundamentals:</u>

- Diaphragm rupture (ΔP) produces shock wave
- Reflected shock wave yields stagnant gas at high pressure and/or temperature
- Highly-controllable conditions
- Experiment terminates when contact surface arrives (~ms)

<u>Laser Diagnostic(s):</u>

- Monochromatic light attenuation
- Beer's Law:

$$\frac{I}{I_0} = \exp(-k_\nu L P X_i)$$

- Absorption coefficient is computed

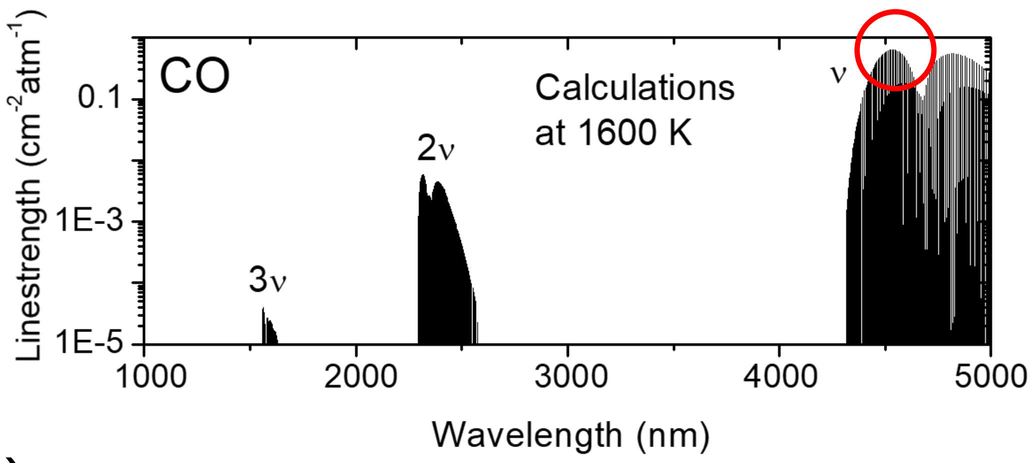
$$k_\nu = \underbrace{S}_{\substack{\text{Linestrength} \\ f(T)}} \times \underbrace{\phi(\nu - \nu_0)}_{\substack{\text{Lineshape} \\ f(T,P,\nu)}}$$

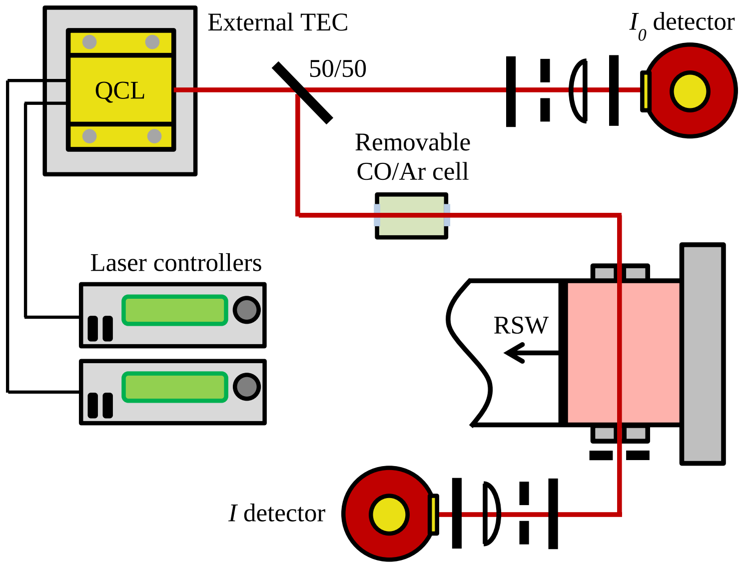
Experimental Setup – Laser Diagnostics

CO and H_2O laser diagnostics implemented on the shock tube

CO Laser Diagnostic

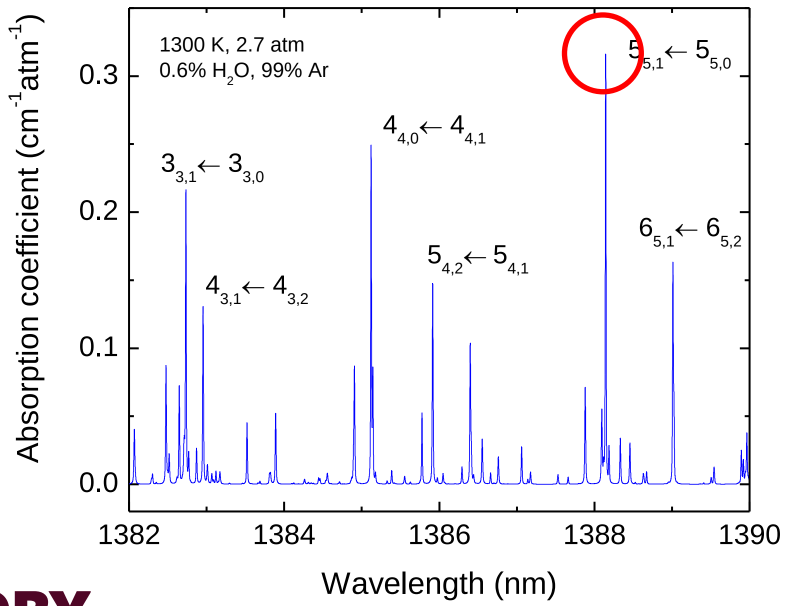
- Quantum cascade laser (QCL)
- Matched InSb photodetectors
- Removable CO/Ar cell
- Fundamental ($\Delta\nu = 1$) CO band
- R(12), $\nu'' = 0$ transition (4566.17 nm)

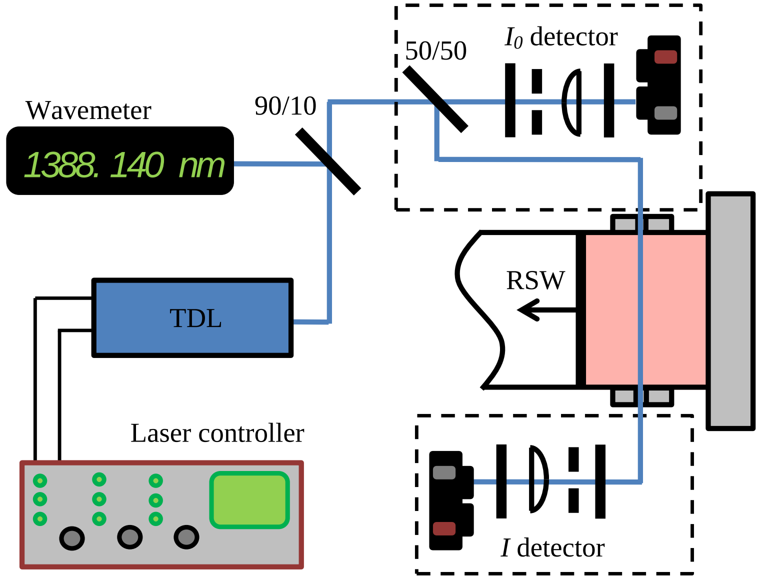




H_2O Laser Diagnostic

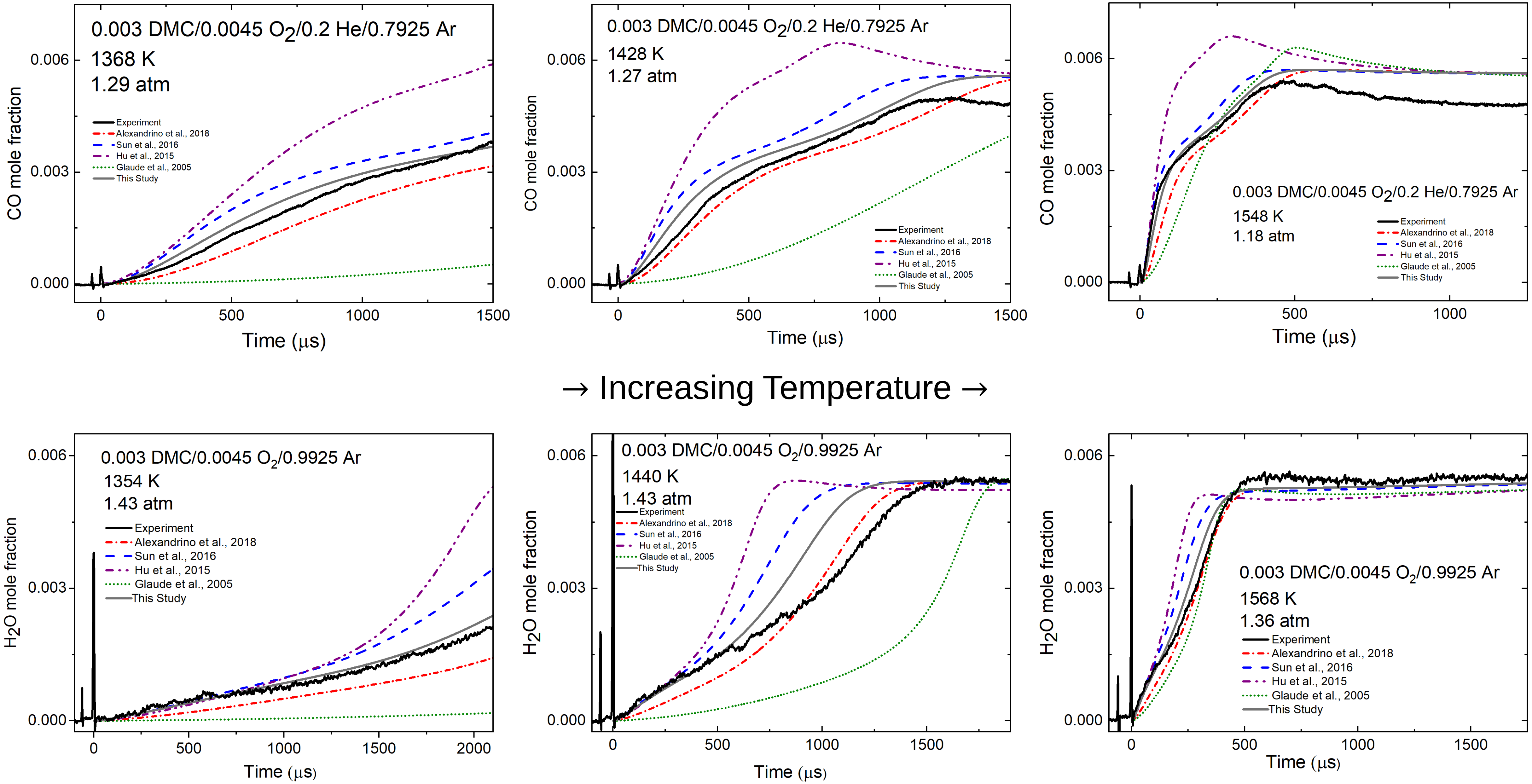
- Tunable diode lase (TDL)
- Matched InSb photodetectors
- Lexan enclosures (N_2, < 0.1% RH)
- The $\nu_1 + \nu_3$ combination band
- $5_{5,1} \leftarrow 5_{5,0}$ transition (1388.139 nm)
- No interference from CO or CO_2





Results – DMC ST Combustion (ϕ=2) – Species

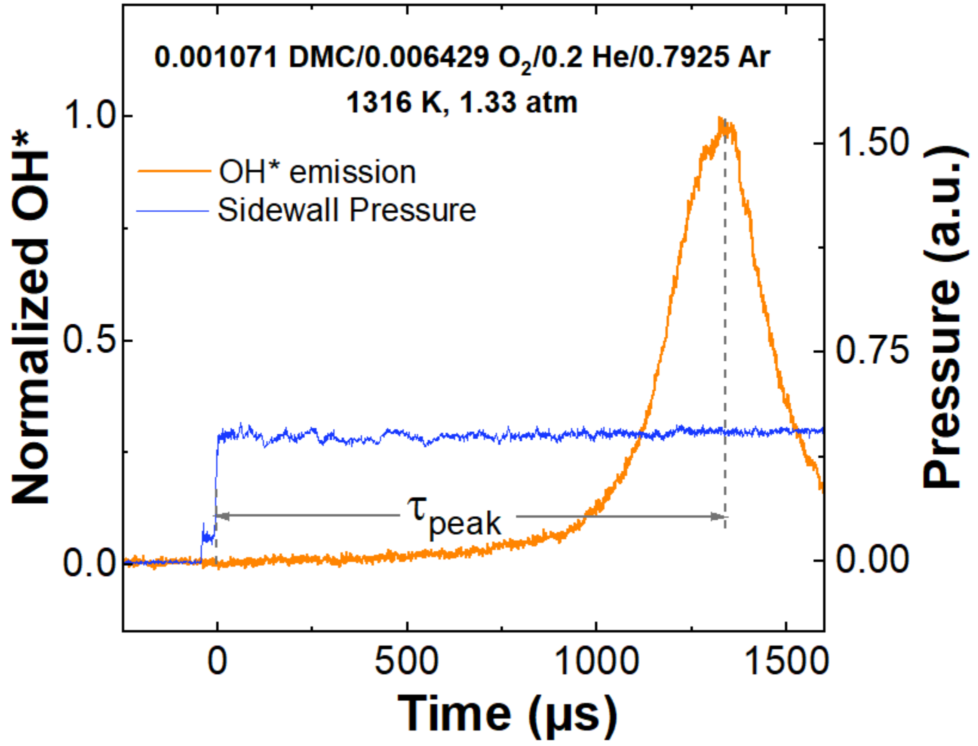
Established database of transient species profiles

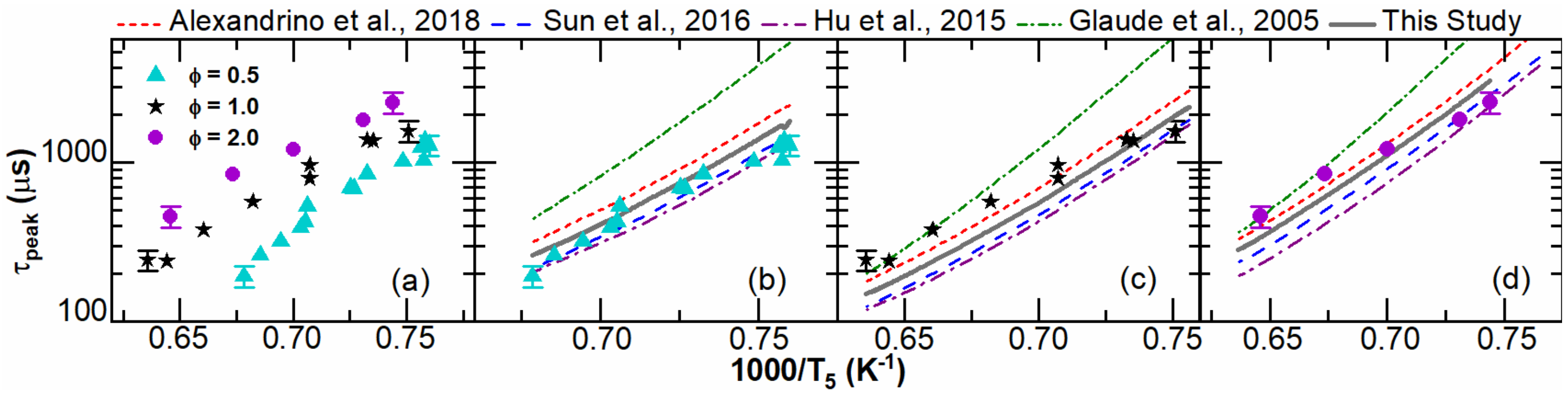


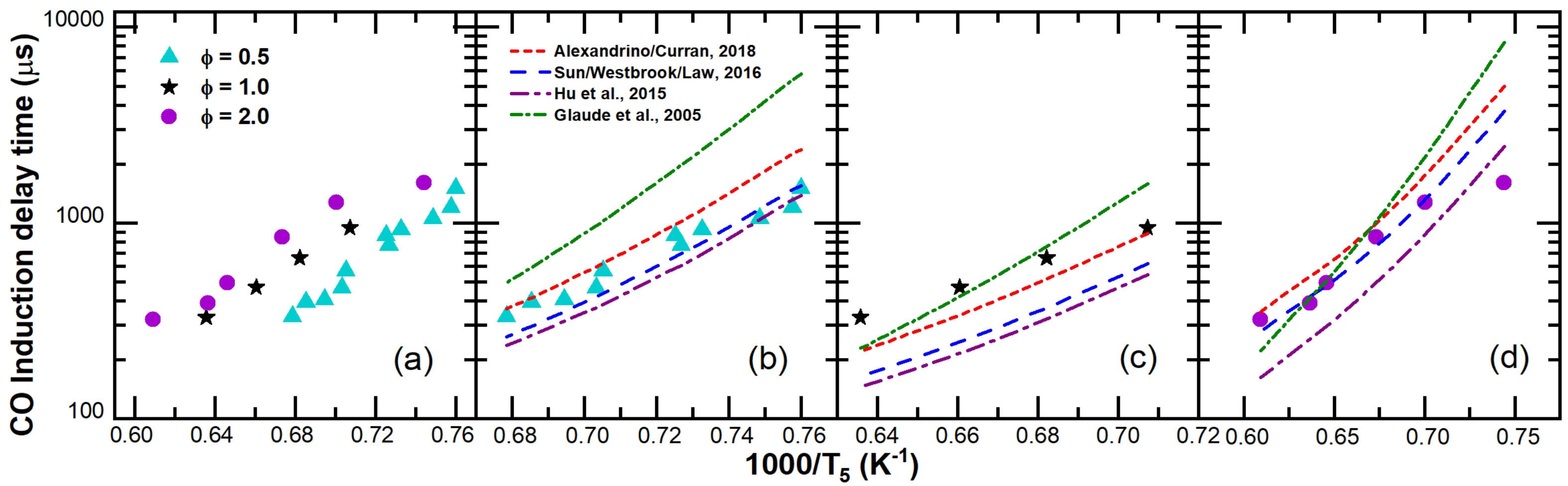
[Atherley et al., 2020]

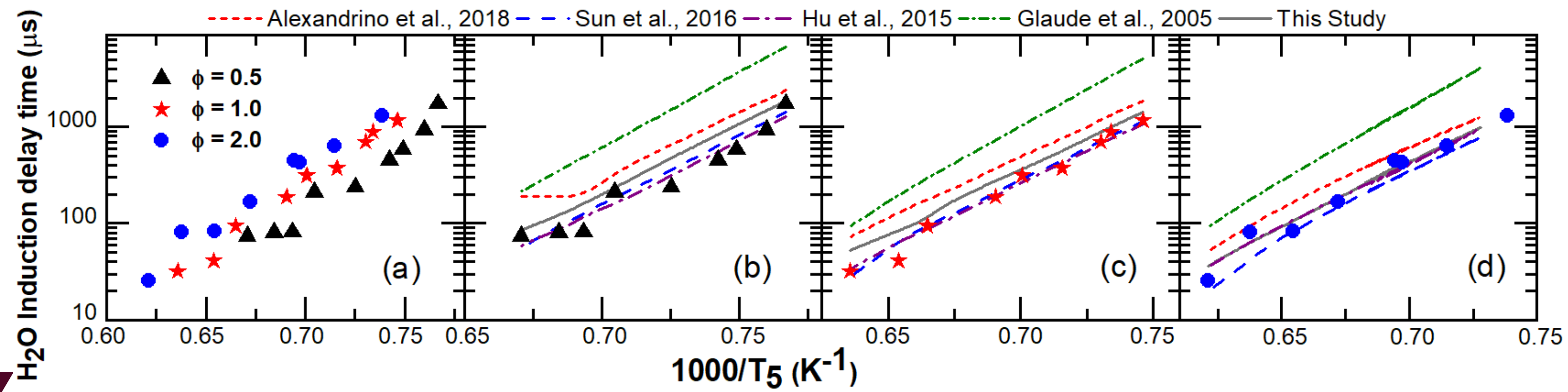
Results – DMC ST Combustion – Delay Times

Established database of ignition and induction delay times









Key Findings:

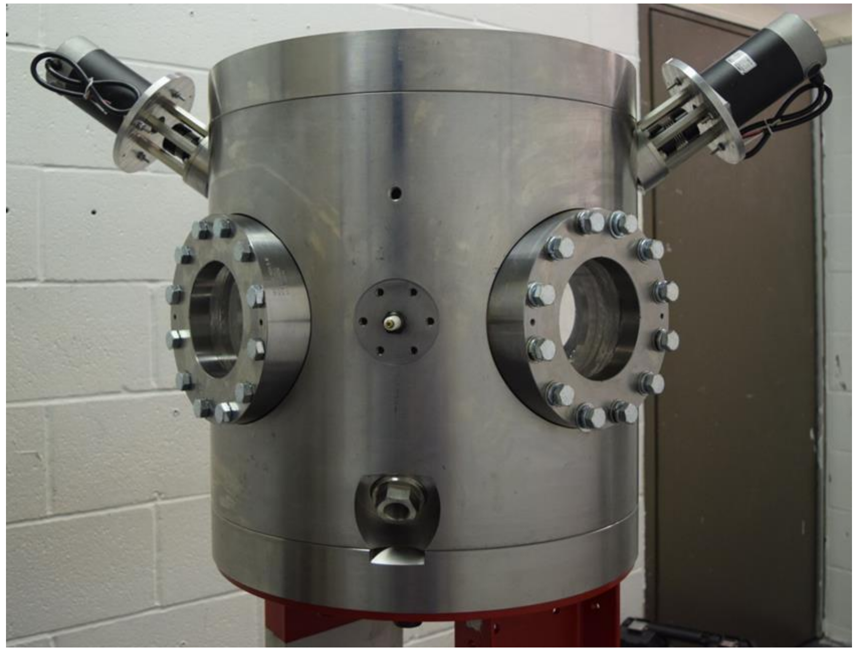
- Established fundamental database(s)
  - Transient species profiles
  - Ignition/induction delay times
- Improved state-of-the-art models

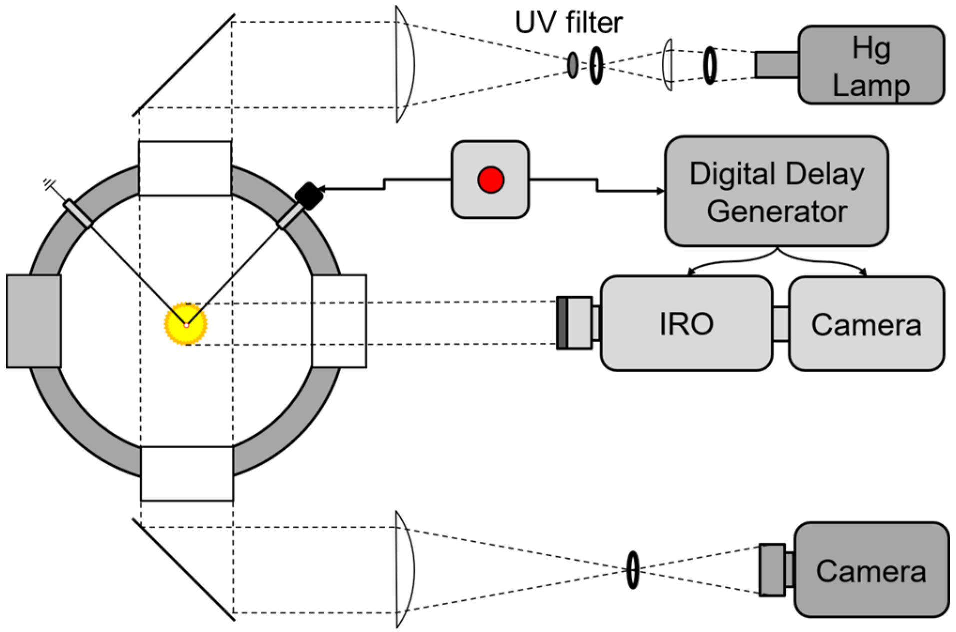
Experimental Setup – Flame Speed Measurements

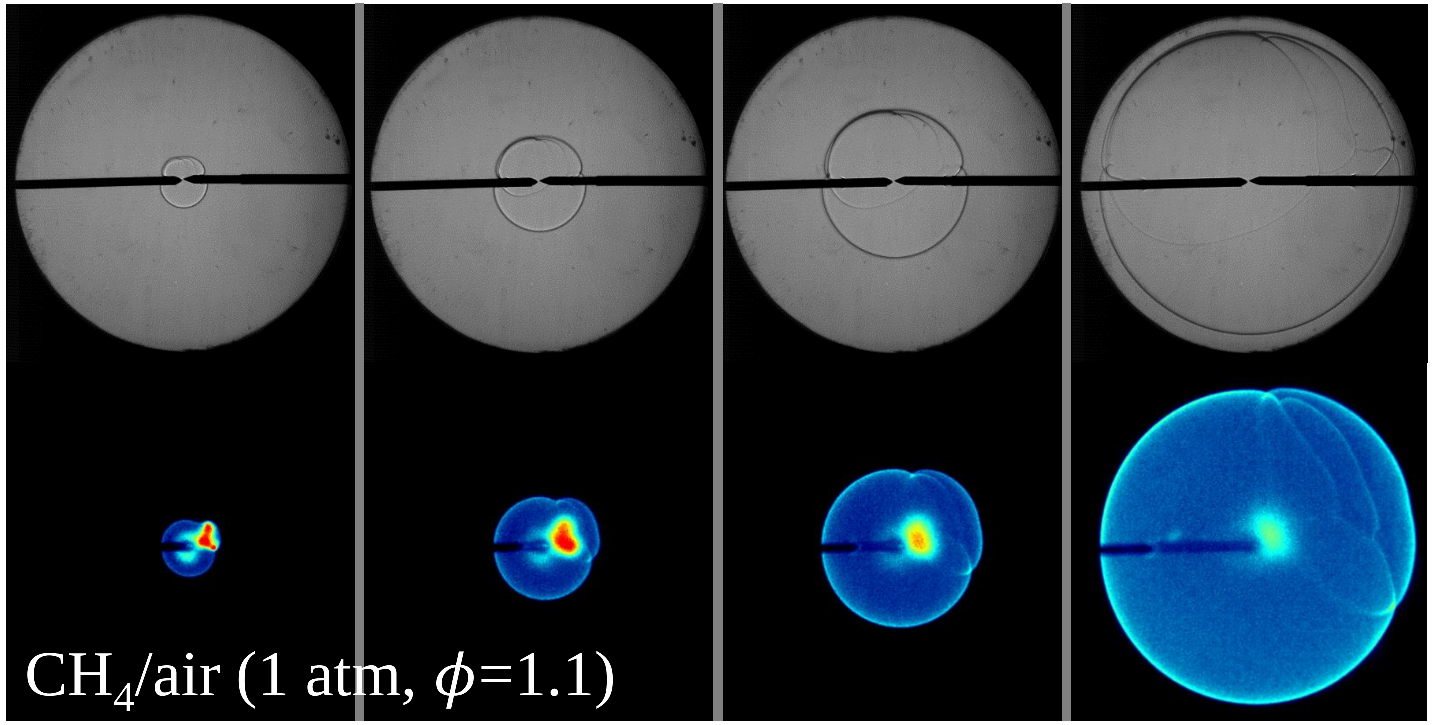
Laminar flame speed measurement provide fundamental combustion data

Laminar Flame Speed Vessel:

- Constant-volume bomb (14” ID x 16” H)
- High pressure (P < 150 psia) and temperature (T_0 < 400 K)
- Spherically expanding flame
- Optical diagnostics
  - Schlieren photography
  - Chemiluminescence (CH* and OH*)





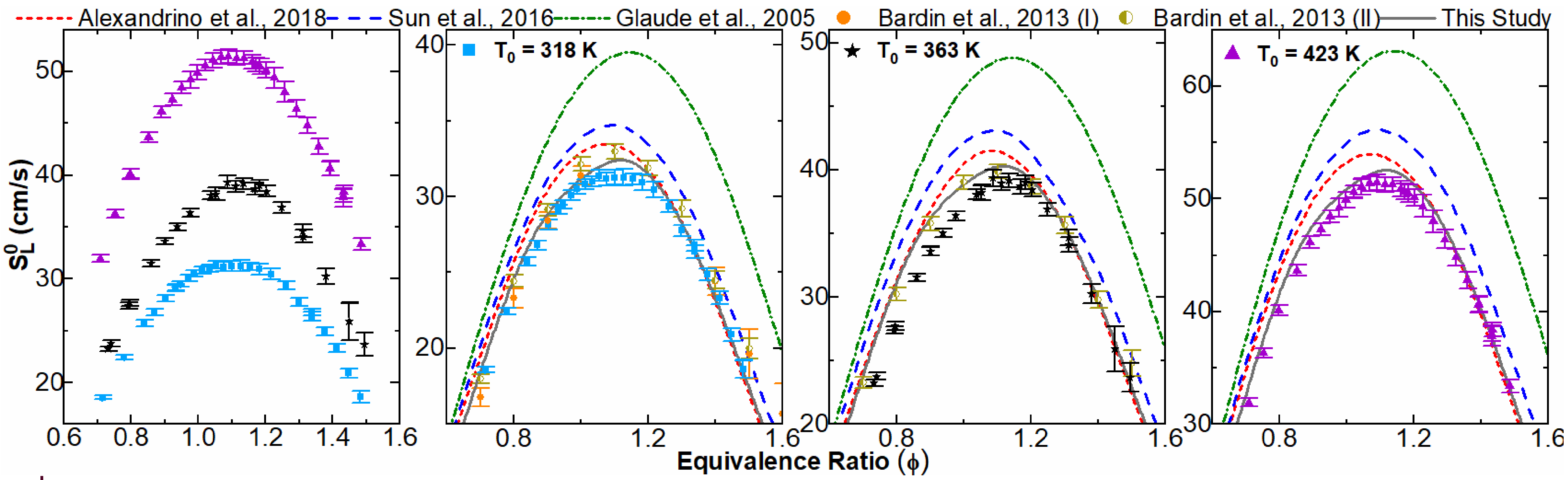


Results – Flame Speed Measurements

Established database laminar flame speeds

Key Findings:

- DMC flame speed measurement database established over wide range of temperatures (completed at CNRS ICARE facility)
- Good agreement with available literature
- State-of-the-art chemical kinetics models improved



[Atherley et al., 2020]

Summary and Future Work

Summary

- Shock tube and laminar flame speed experiments conducted with dimethyl carbonate (DMC)
- Pyrolysis and combustion (w/ O_2) conditions over a wide range of conditions
- Experimental data included species (CO & H_2O) time histories, ignition/induction delay times, and laminar flame speeds
- State-of-the-art kinetics models compared to the collected dataset and improved

Future Work

- Further data is required with other electrolytes and electrolyte mixtures
  - Key reactions need to be revisited
- Evaluation of candidate fire suppressant additives

Part 3: Ongoing/Future Work

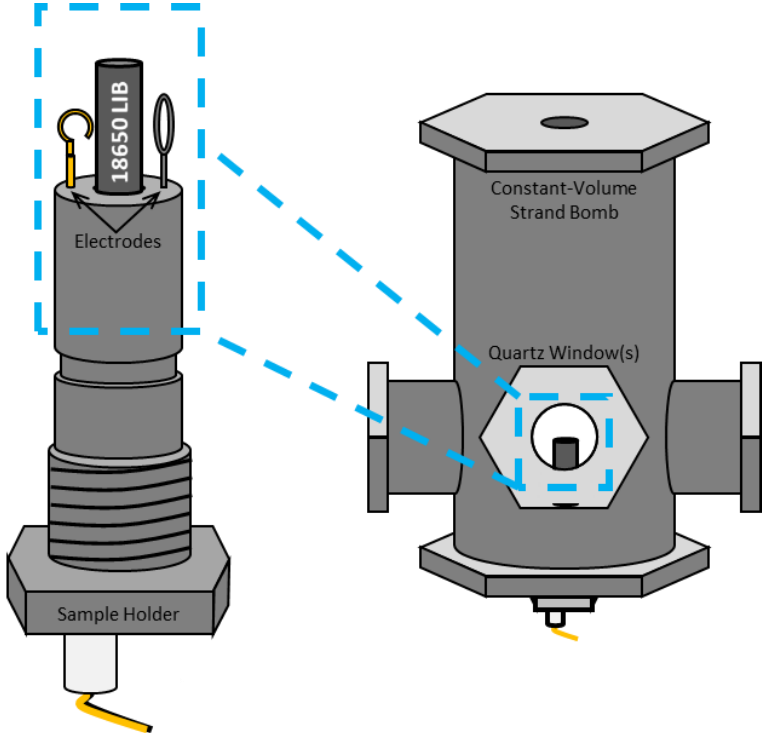
Pipeline LIB thermal runaway and combustion projects at TAMU

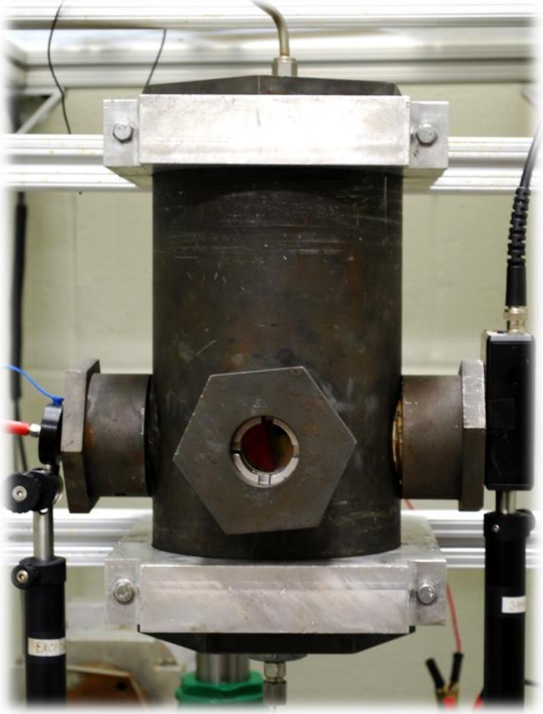
Optical Diagnostics for LIB TR and Combustion

High-fidelity diagnostics can improve our fundamental understanding of LIB TR

Experimental Approach:

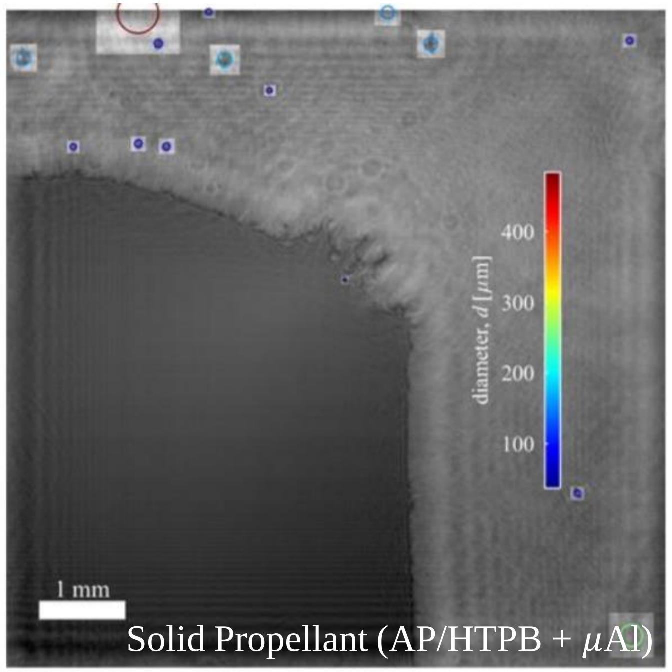
- LIB TR in an atmosphere-controlled bomb
- Initiation via heating, overcharge, laser
- Application of high-fidelity diagnostics
- Future: NASA JSC calorimeter





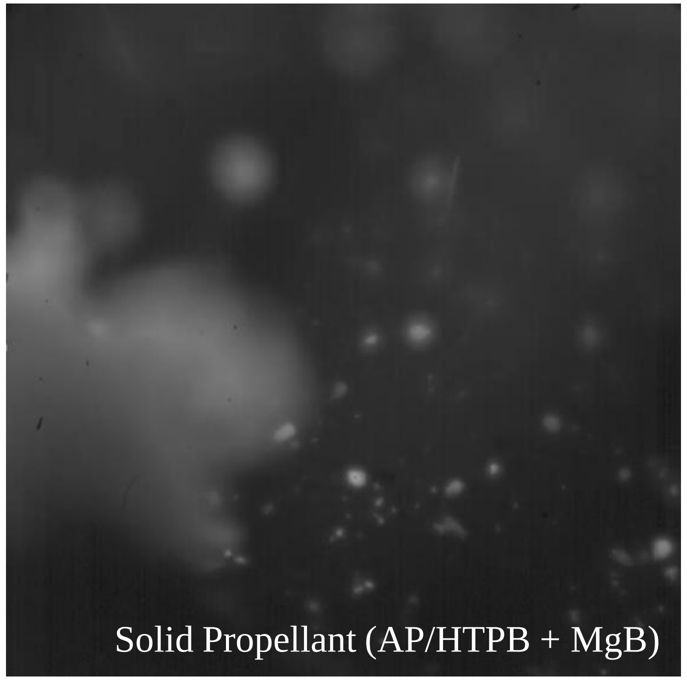
Potential Diagnostics:

- High-speed video optics
  - High-magnification (< 3 μm/pixel)
  - Emission imaging
  - Chemiluminescence
  - Digital holography (Dr. W. Kulatilaka)
- Laser-based species measurements



Solid Propellant (AP/HTPB + μAl)

[Niemiec., 2018]



Solid Propellant (AP/HTPB + MgB)

Combustion Behavior of LIB TR Vent Gas

Vent gases from LIB TR events can pose significant combustion hazards

UL 9540A: Test Method for Evaluating Thermal Runaway Fire Propagation in Battery Energy Storage Systems

- Cell Level Testing: TR propensity and characteristics
  Vent gas composition and combustion characteristics
- Module, Unit, and Installation Level Testing

Experimental Approach:

- LIB TR initiation in a controlled-atmosphere bomb
- Gas capture via catch tank and subsequent compositional analysis (GC-MS)
- Fundamental combustion measurements with: a) captured TR vent gas
  b) emulated gas compositions
- Experimental measurements: flammability limits, flame speed, explosion pressure, etc.

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