Measuring Shock-Tube Ignition and Oxidation at Elevated Pressures

Eric L. Petersen
Department of Mechanical Engineering
Texas A&M University

Workshop on Techniques for High-Pressure Combustion
Argonne National Laboratory
Argonne, IL, 31 August 2011

Overview

1. High-Pressure Shock Tube Capabilities
2. Recent and On-Going Studies
3. Chemiluminescence Measurements
4. Challenges and Future Efforts at Elevated Pressures

High-Pressure
Shock Tube Capabilities

Shock Tubes

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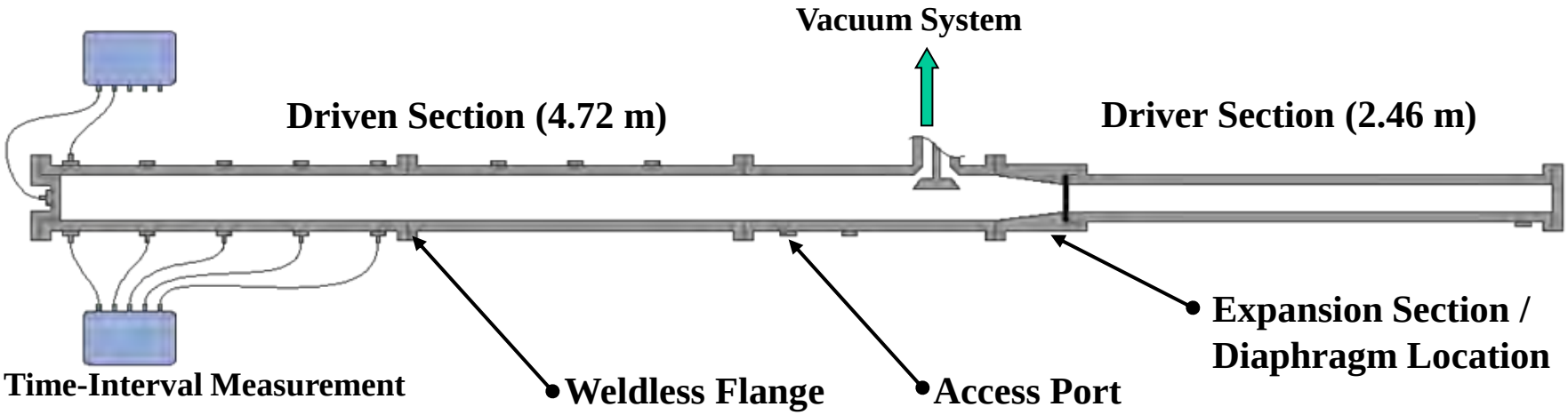
High-Pressure Shock-Tube Facility

- 1 – 100 atm Capability
- 600 – 4000 K Test Temperature
- Up to 20 ms Test Time
- 2.46 m Driver and 4.72 m Driven
- 15.24 cm Driven Inner Diameter



Shock Tubes

Low-Pressure Shock-Tube Facility is also Utilized

TAMU Low Pressure Shock-Tube Facility

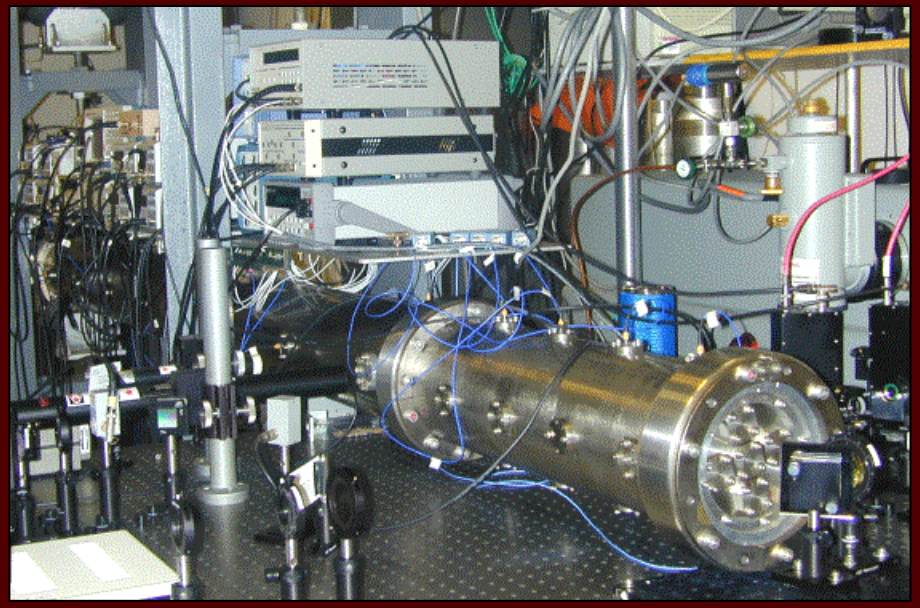
- 4 x 4 in Cross Sectional Driven Area
- 1 – 10 atm Capability
- 1-2 ms Test Time
- 6.1 m Length
- Heating Capability

Shock Tubes

Second High-Pressure Shock Tube Located at Aerospace Corp.

Aerospace High Pressure Shock-Tube Facility

- 1 – 100 atm Capability
- 600 – 4000 K Test Temperature
- 3 ms Test Time
- 3.5 m Driver and 10.7 m Driven Sections
- 16.2 cm Driven Inner Diameter



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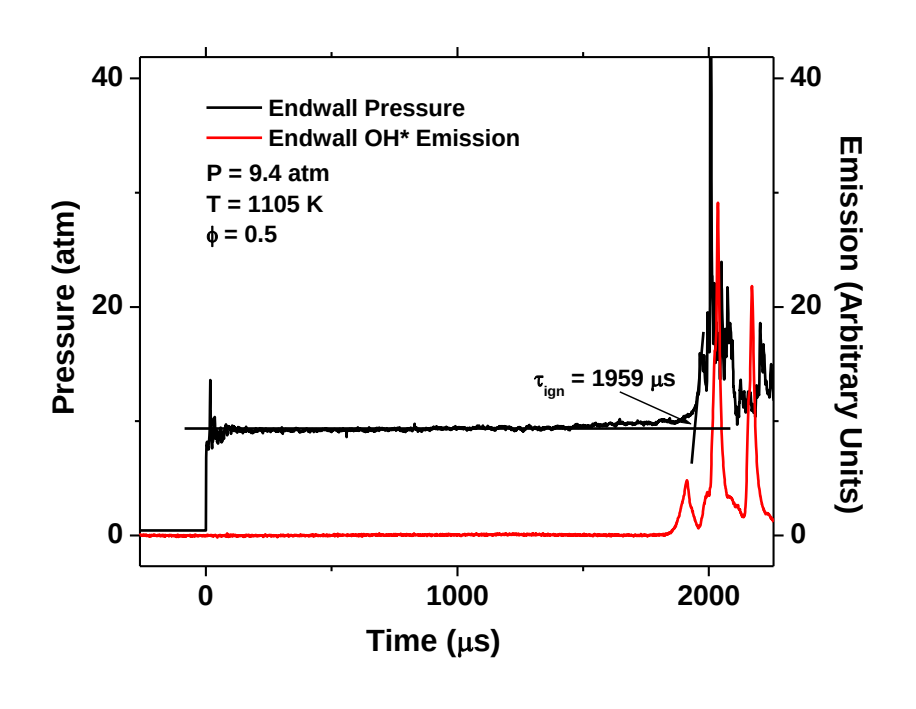
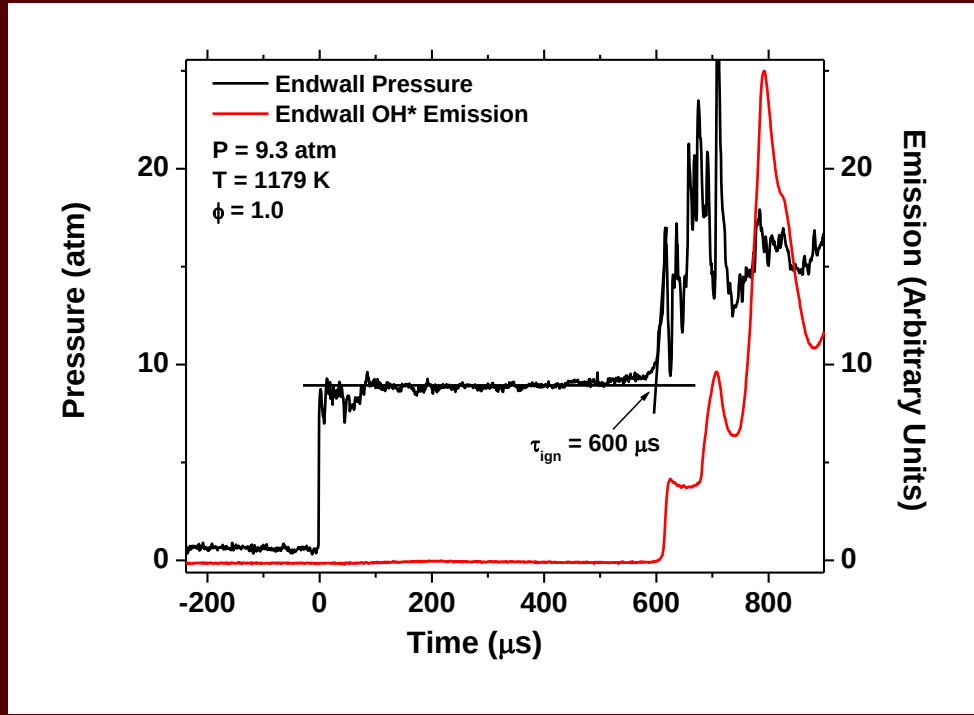
Experimental Apparatus

Ignition Delay Time from Pressure, Emission



Endwall Pressure Trace for long test time



Endwall Pressure Trace For typical test time

Recent and On-Going Studies

Recent Studies

1. C1 – C4 Ignition Delay Times
2. Pentane Experiments
3. Hydrogen and Syngas Experiments
4. Syngas with Impurities

C1-C4 Ignition Times

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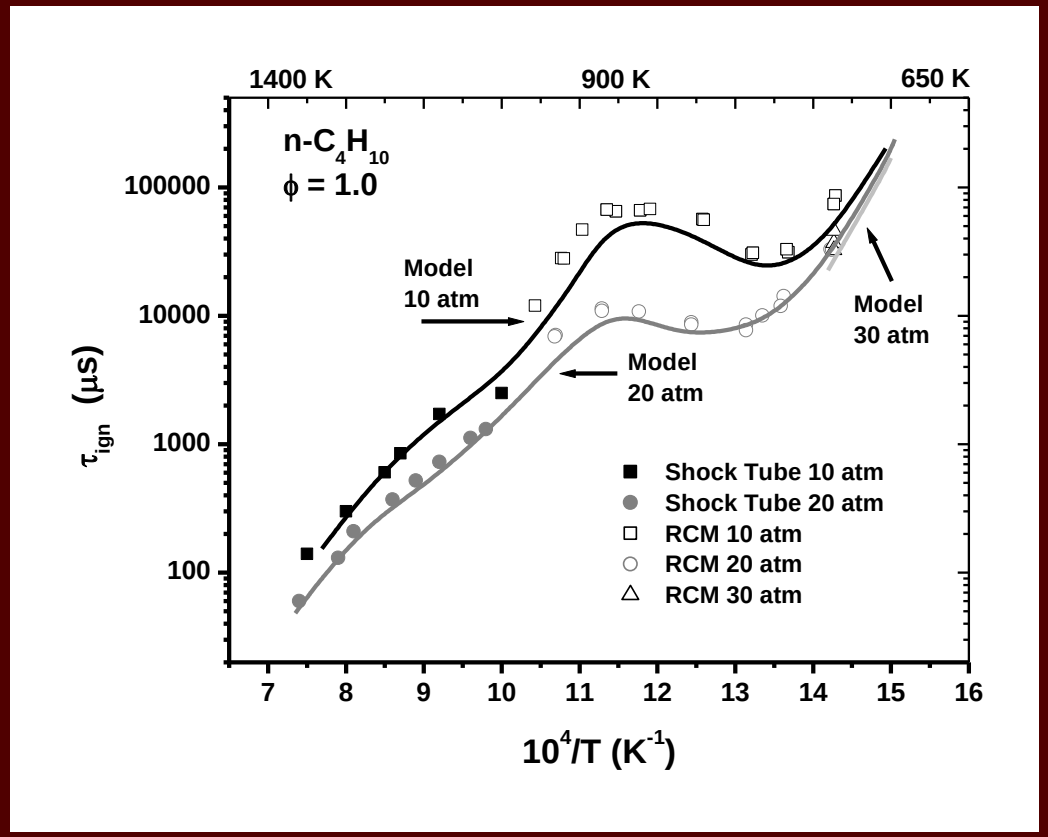
Recent Experiments and Modeling:

- Real Fuel-Air Mixtures at High Pressures
- 1 – 50 atm
- Shock Tube and RCM Temperatures



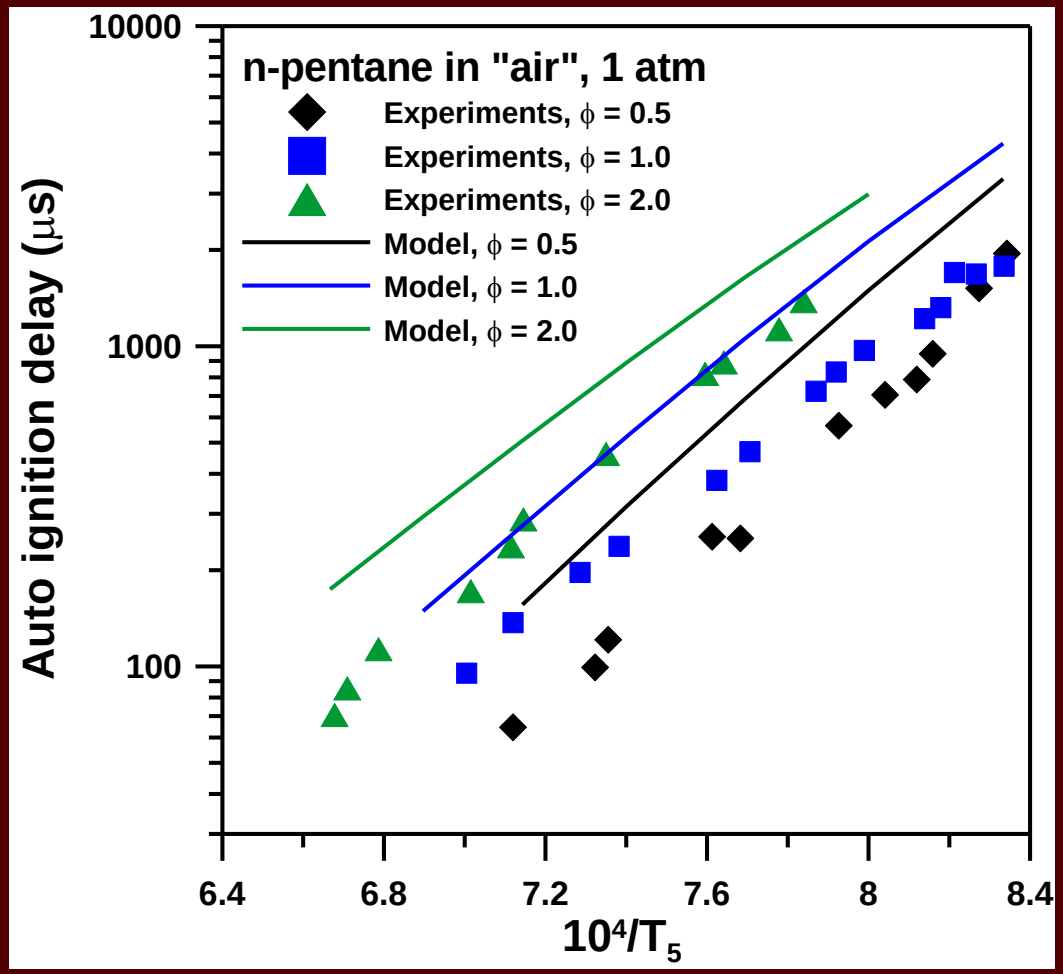
Validated with:

- CH_4, C_2H_6, C_3H_8
- CH_4/C_3H_8, CH_4/C_2H_6, CH_4/H_2, $CH_4/C_2H_6/C_3H_8$
- $CH_4/C_2H_6/C_3H_8/C_4H_{10}/C_5H_{12}$
- CH_4/DME, DME
- C_4H_{10}, iC_4H_{10}

NUI Galway
OÉ Gaillimh

Pentane

***New Experiments** are Focusing on **Pentane** Mixtures*



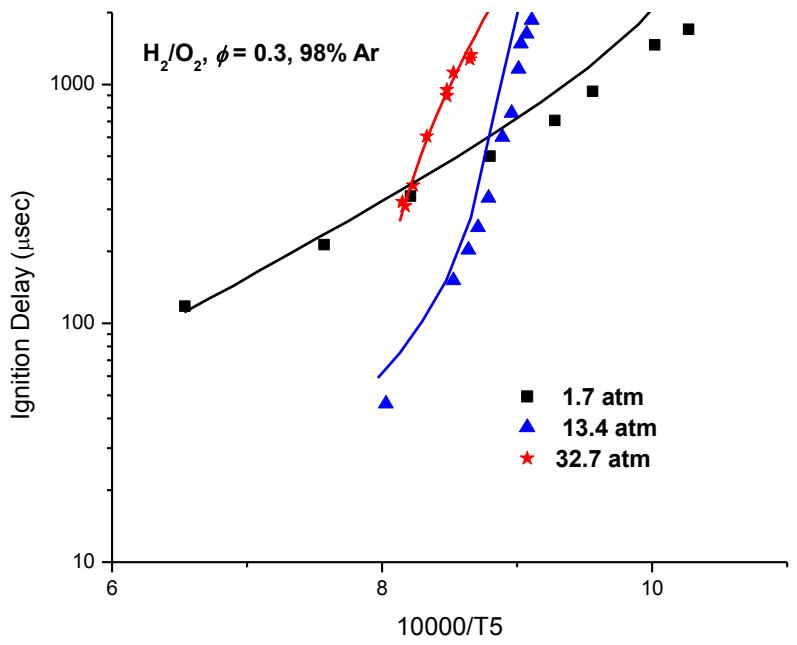
NUI Galway
OÉ Gaillimh

Rolls-Royce

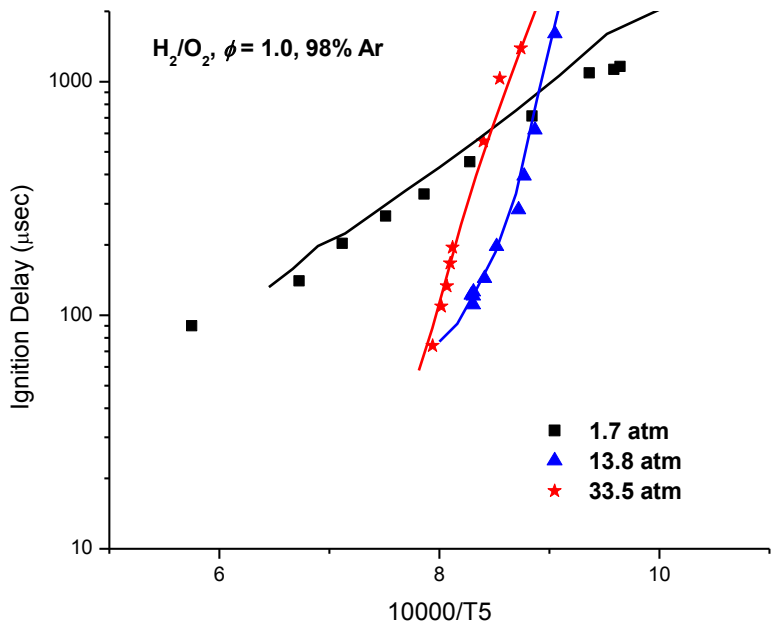
H_2 and Syngas

High-Pressure Experiments in H_2-O_2 Mixtures Show Good Agreement with Mechanism

ϕ = 0.3



ϕ = 1.0

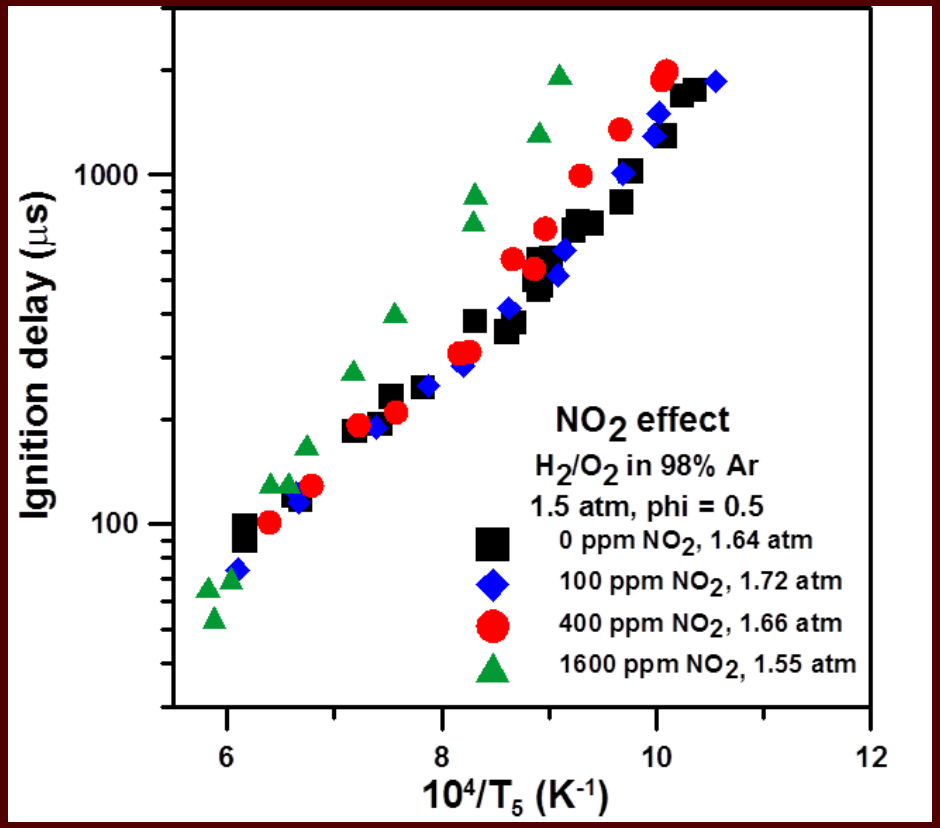


NUI Galway
OÉ Gaillimh

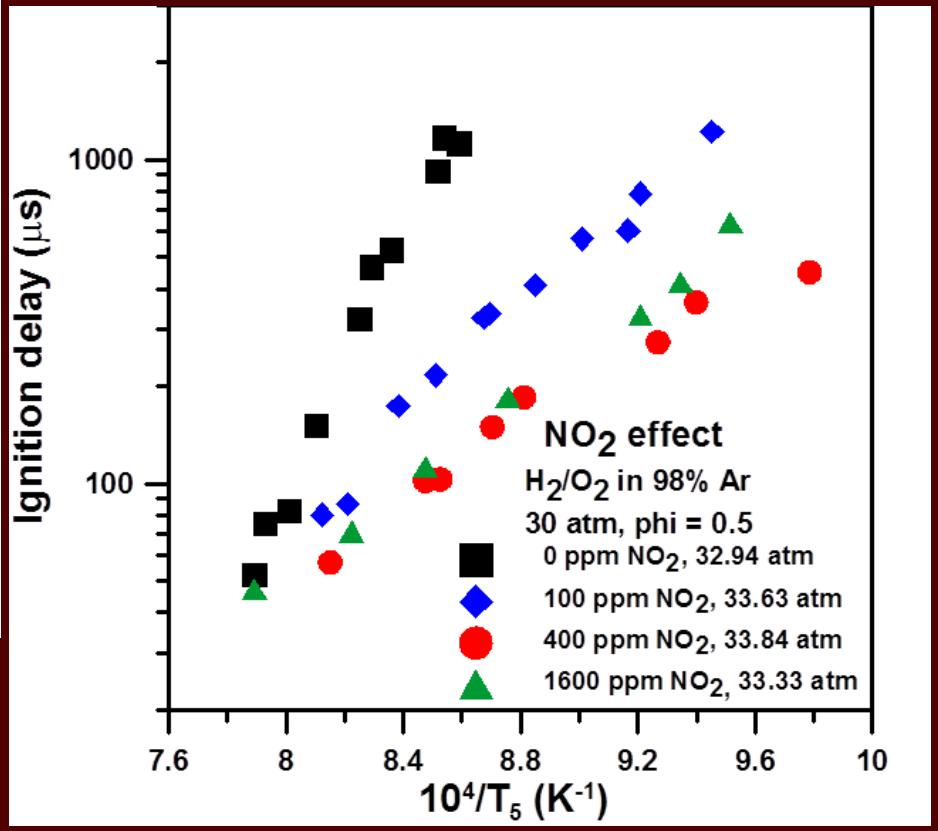
Syngas with Impurities

Effect of NO_2 on H_2-O_2 Mixtures is Being Studied

1.5 atm



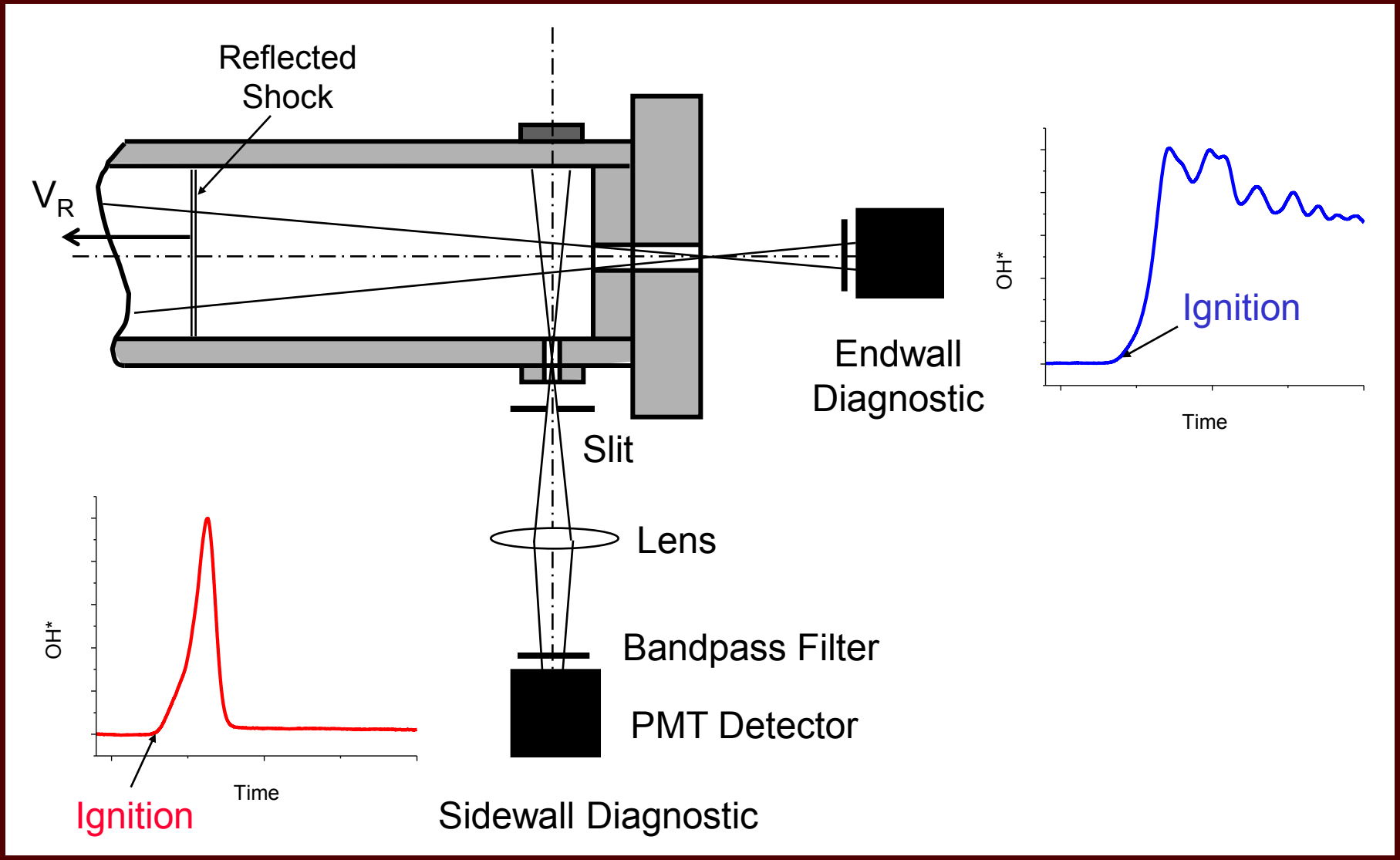
30 atm



Chemiluminescence

Chemiluminescence

***Emission** and **Pressure** Measured from Endwall and Sidewall*



Chemiluminescence

We Have Working Kinetics Models for OH, CH* at 1 atm, But* ***Not at Elevated Pressures***

Formation of OH*:

$CH + O_2 = CO + OH^*$
$O + H + M = OH^* + M$

Depletion of OH*:

$OH^* = OH + h\nu$
$OH^* + M = OH + M$

Formation of CH*:

$C_2H + O = CO + CH^*$
$C_2 + OH = CO + CH^*$
$C + H + M = CH^* + M$

Depletion of CH*:

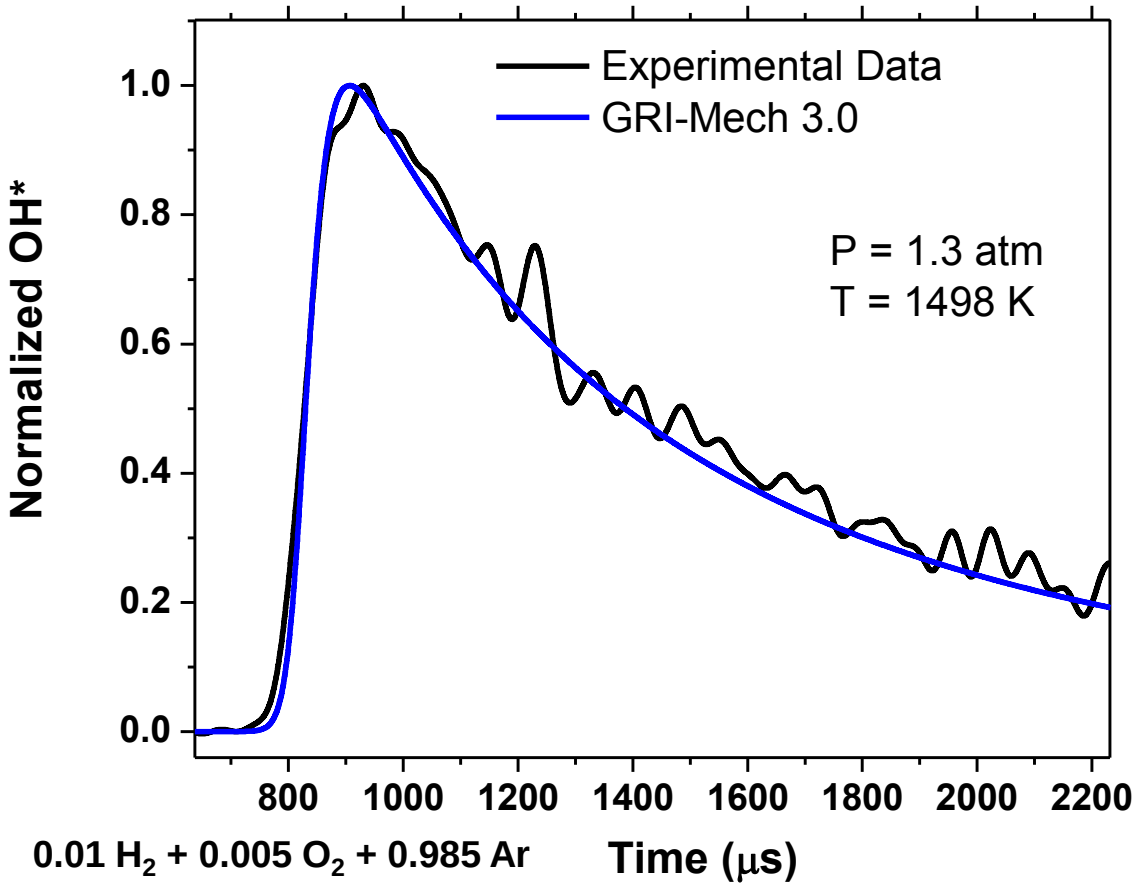
$CH^* = CH + h\nu$
$CH^* + M = CH + M$

Hall and Petersen, *IJCK* (2006)
Hall and Petersen, AIAA 2004-4164
Petersen et al., AIAA 2003-4493

Hall et al., AIAA 2005-1318
de Vries et al., *Co&Fl* (2007)

Chemiluminescence

Typical OH Time History at Dilute Conditions*



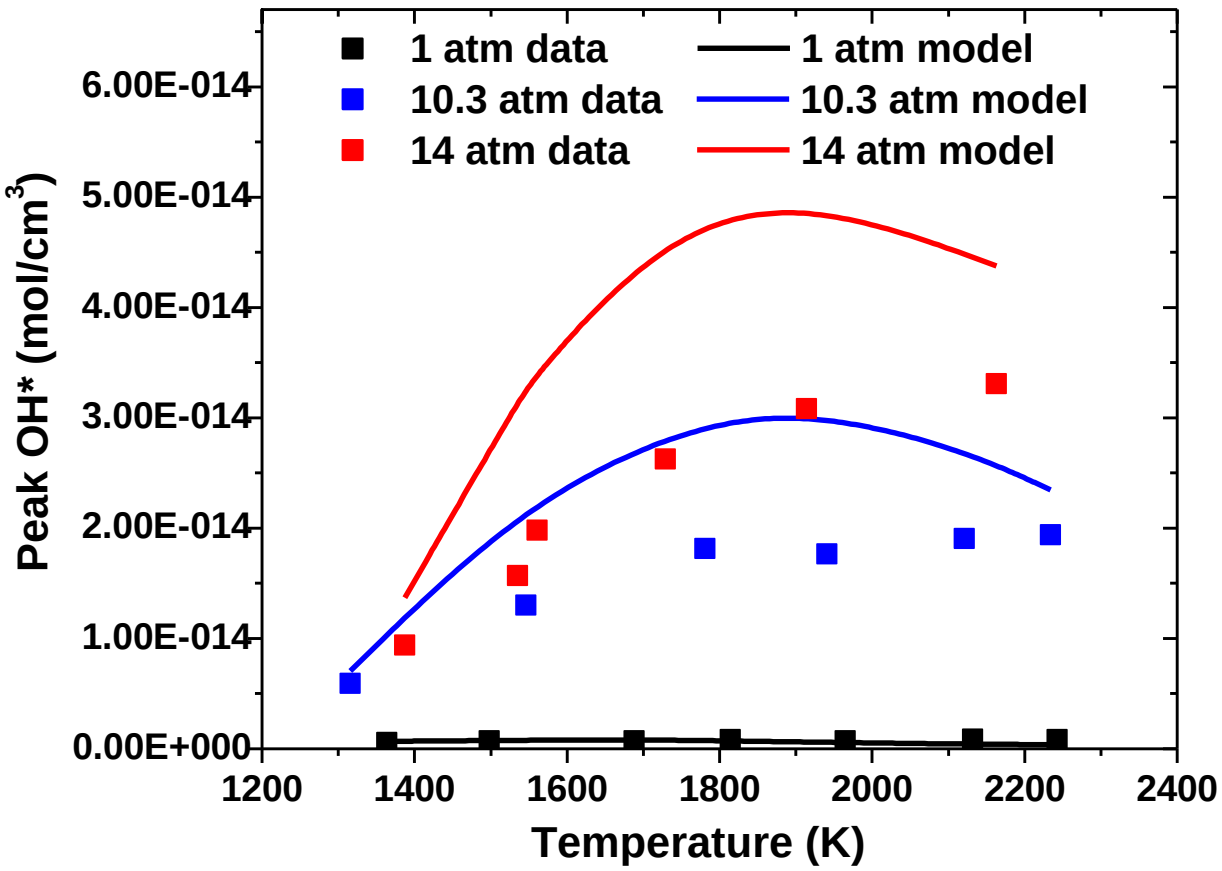
0.01 H_2 + 0.005 O_2 + 0.985 Ar

Well validated for dilute conditions.

Profiles agree with experimental results.

OH* Results

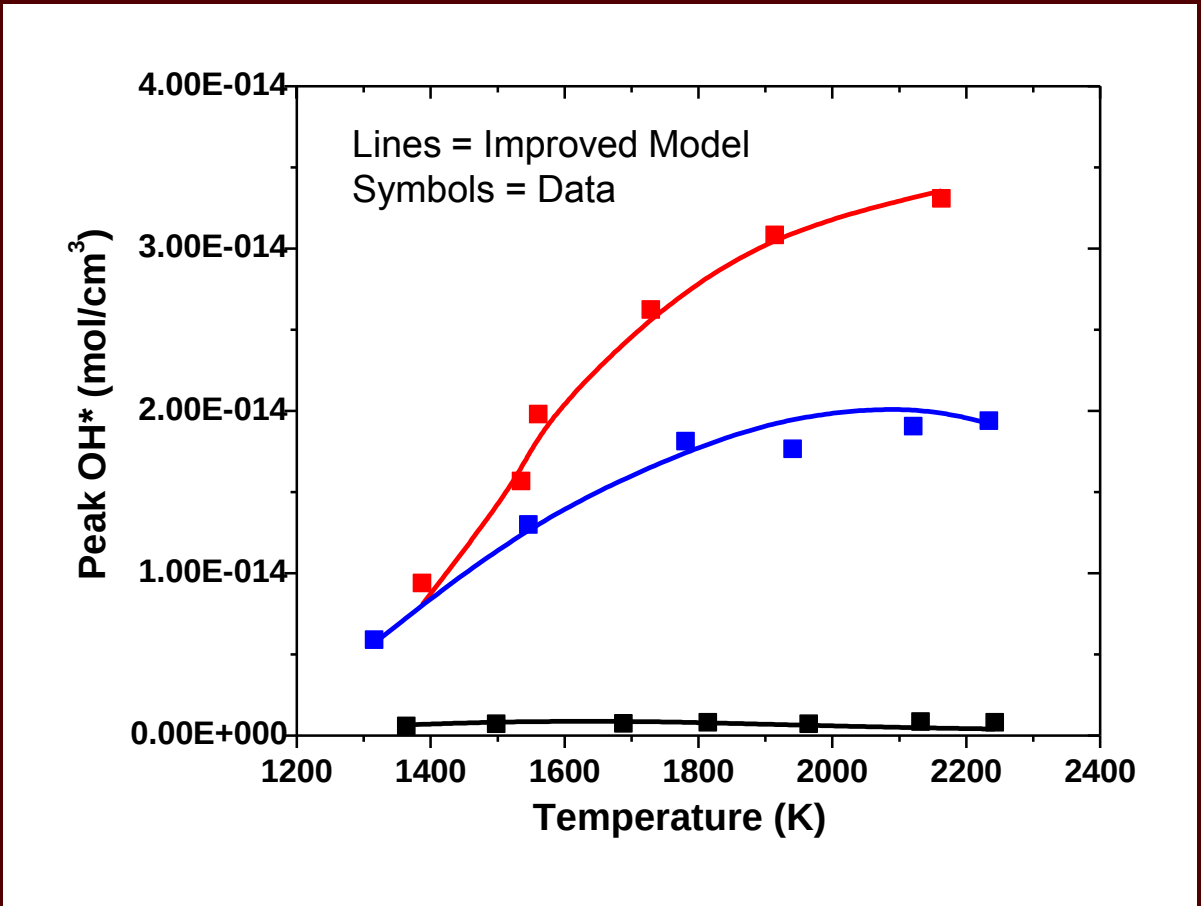
*Current **Model Overpredicts OH*** at Elevated Pressures*



OH* Results

Improved Model Results

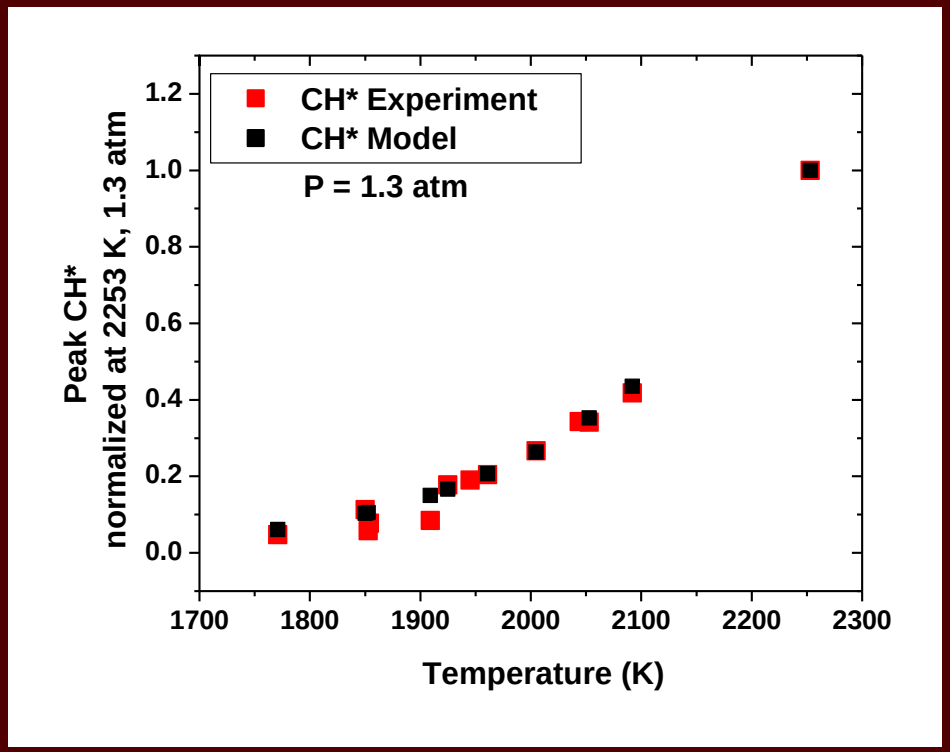
Using new pressure dependent reaction rate.



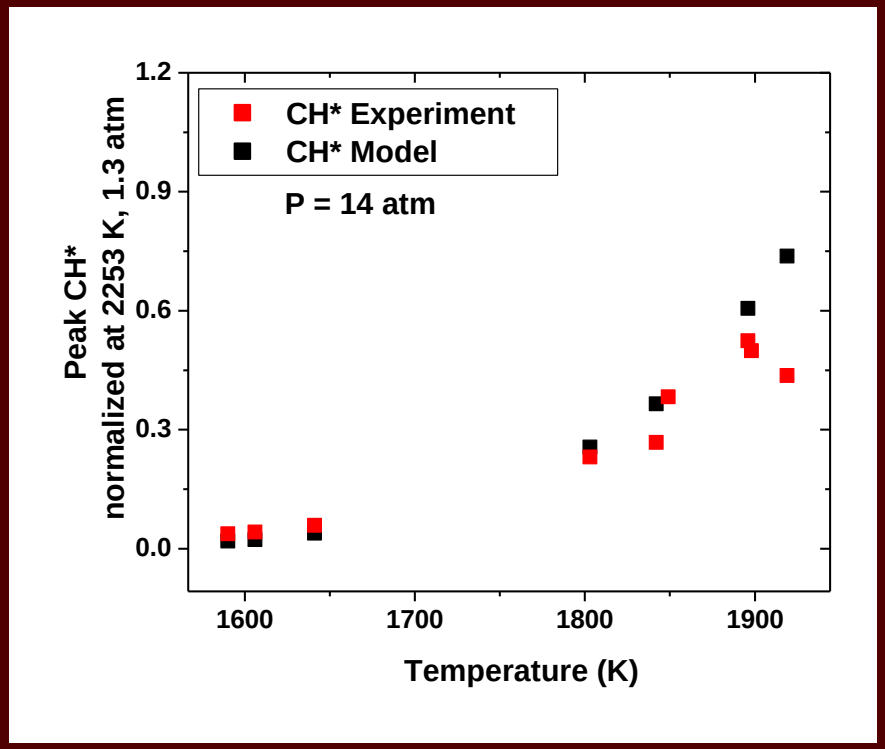
CH* Results

Experimental Peak Concentration of CH is **Accurately Predicted** by Model at Low and High Pressure*

P = 1.3 atm

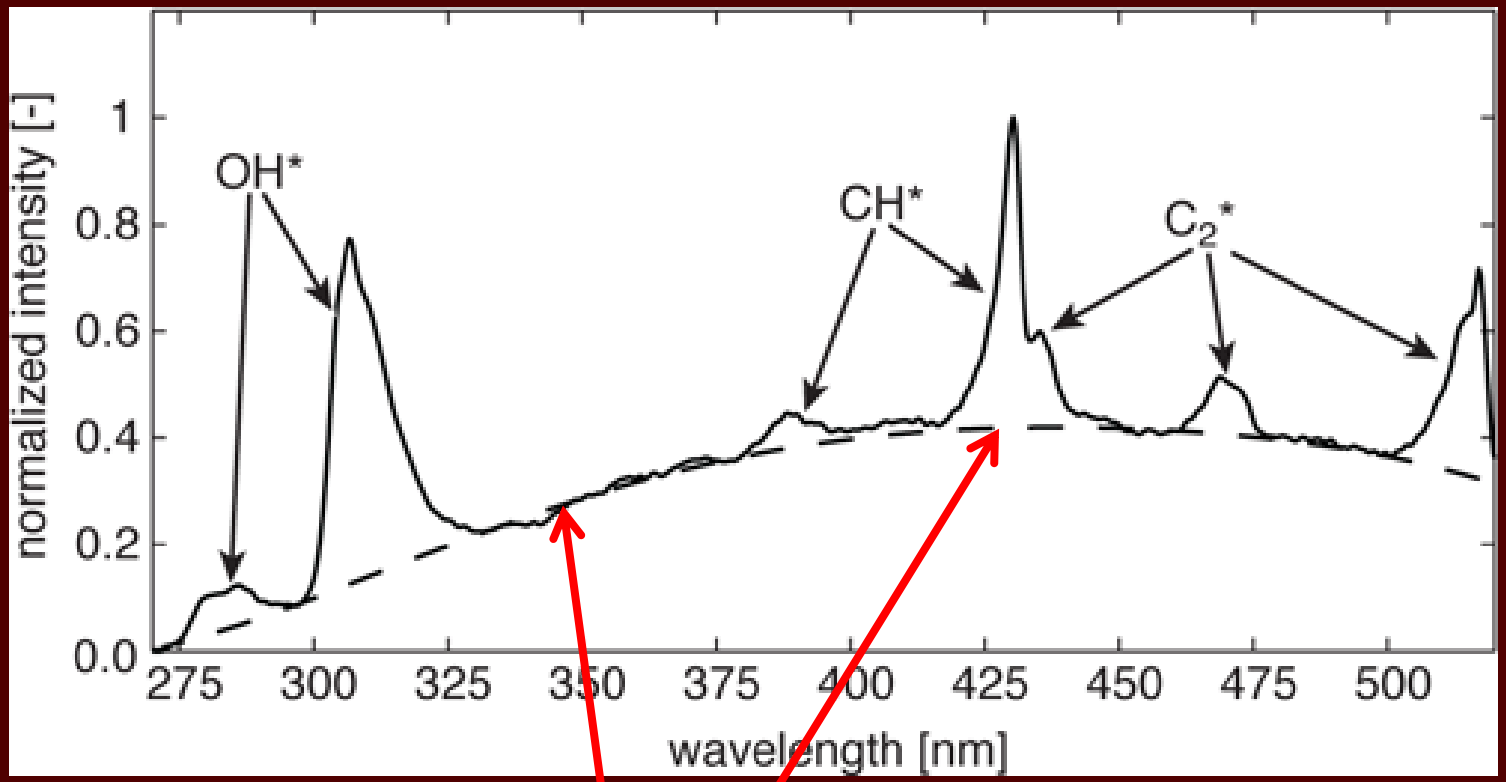


P = 14 atm



Chemiluminescence

Possible Controversy Over Background Emission



CO_2* or CH_2O*, HCO*?

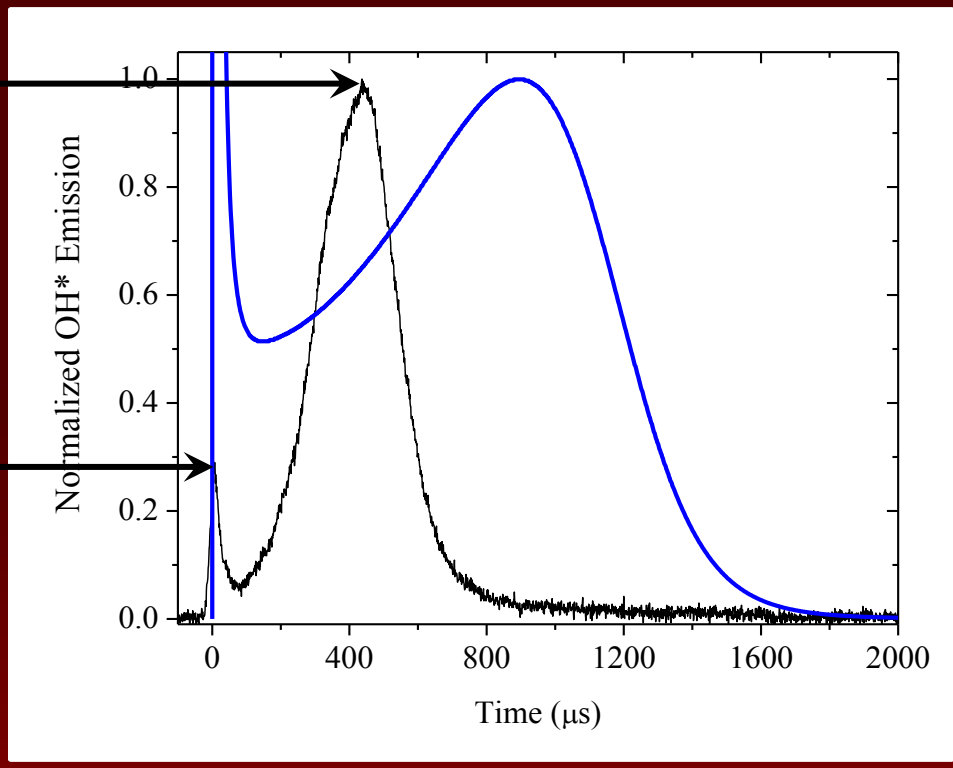
Base Image courtesy of Sattelmayer et al.

Chemiluminescence

Can be Used as Tuning Parameters for Model Improvement

Ignition Peak

Incipient Peak



OH* Kinetics Present Near Time-Zero

Objective 1: Improve OH* Incipient Peak-to-Ignition Peak Ratio

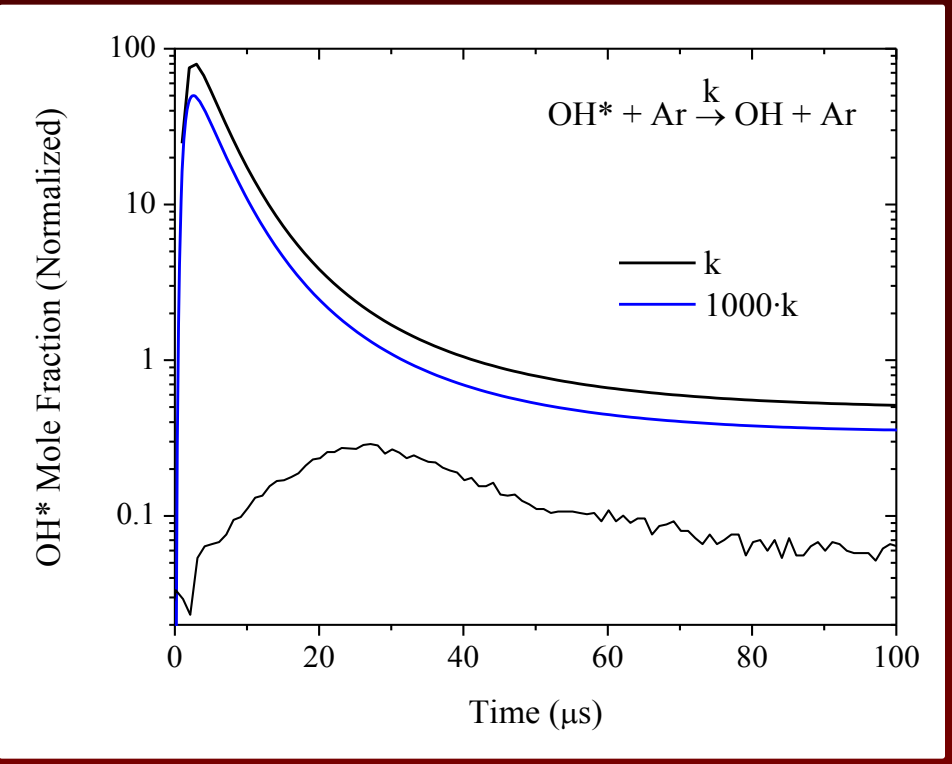
Objective 2: Improve Time-Dependent OH* Kinetics (i.e. Profile Width)

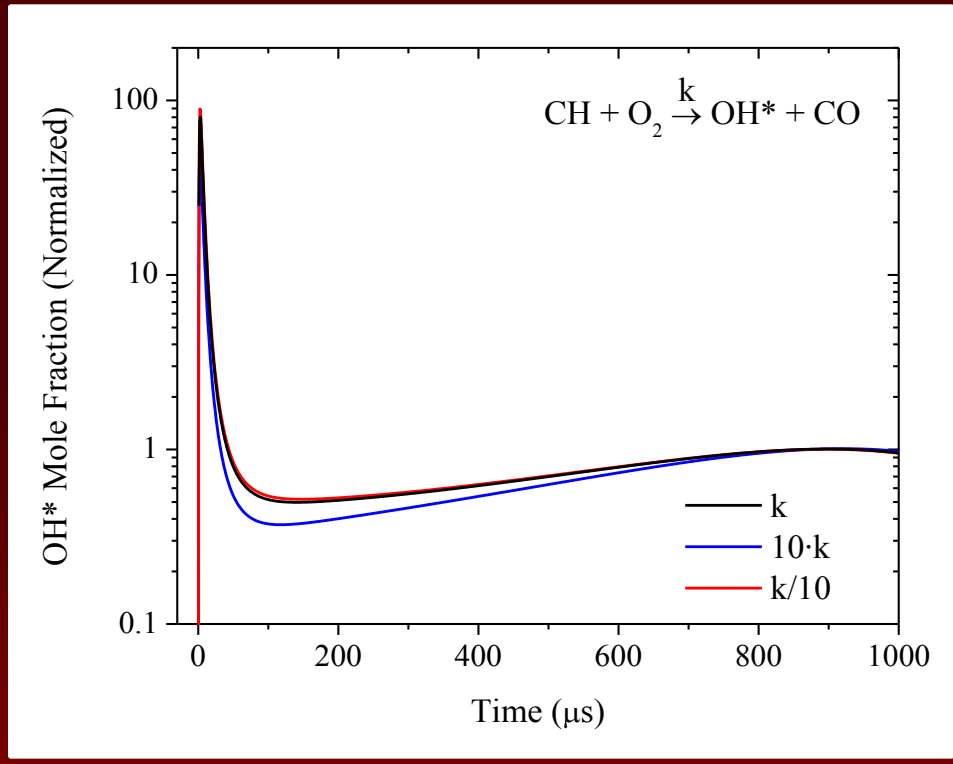
ϕ = 2.0, 1.5 atm

0.10% n-C_9H_{20} + 0.70% O_2 + 99.2% Ar

Chemiluminescence

Incipient Oxidation Insensitive to Primary Chemilum. Reactions





Negligible Reduction of Incipient OH* from Increased Rate of De-excitation by Ar:

$$OH^* + Ar \rightarrow OH + Ar$$

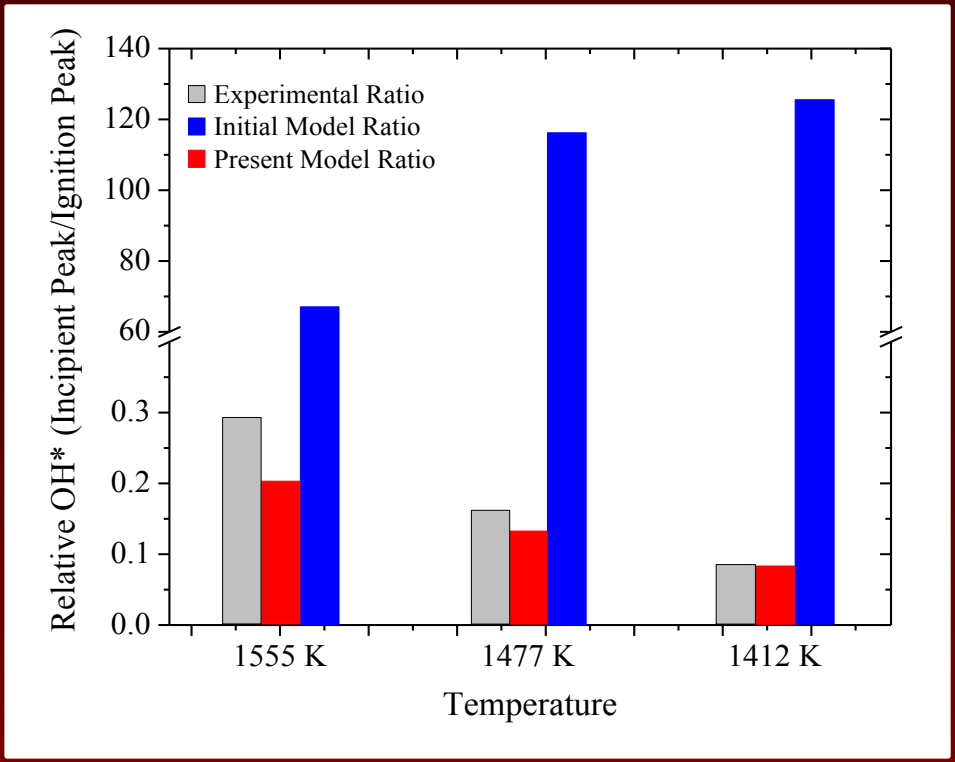
Relative Peak Magnitude Insensitive to Change in Reaction Rate of Primary OH* Production Channel:

$$CH + O_2 \rightarrow OH^* + CO$$

ϕ = 2.0, 1.5 atm, 1555 K, 0.10% n-C_9H_{20} + 0.70% O_2 + 99.2% Ar

Chemiluminescence

Incipient Reactions Leading Toward Model Improvement



Methyl production: $n\text{-}C_9H_{20} \rightarrow C_8H_{17} + CH_3$ $sCH_2 + H_2 \rightarrow CH_3 + H$

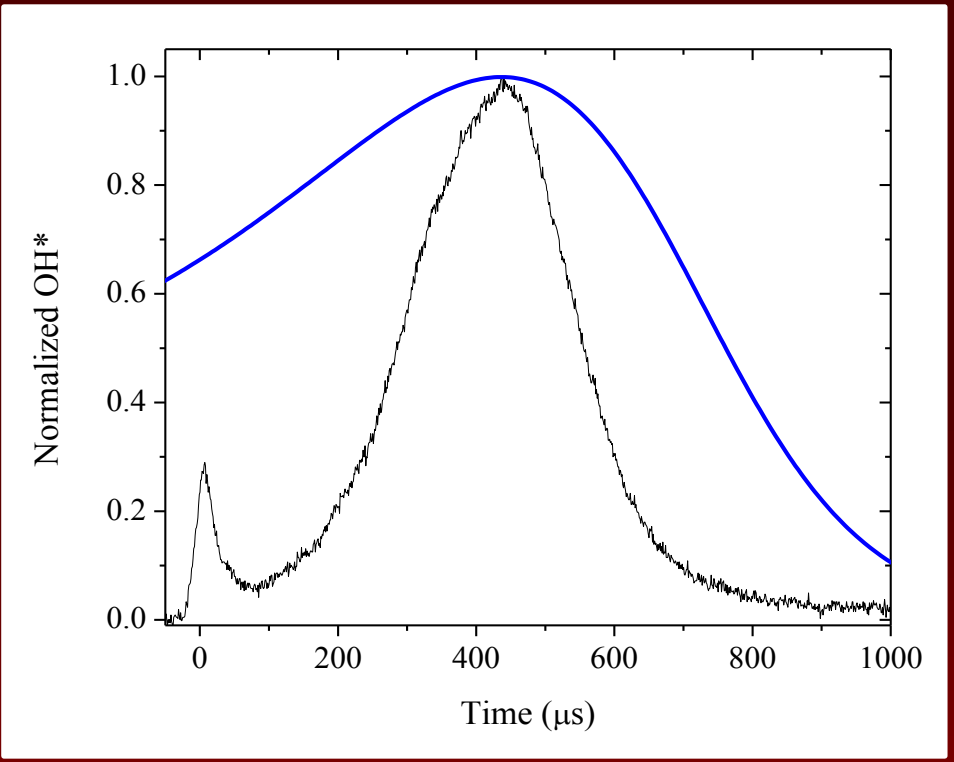
Methylene production: $CH_3 + OH \rightarrow CH_2 + H_2O$ $CH_3 + H \rightarrow CH_2 + H_2$

Methylidyne production: $CH_2 + H \rightarrow CH + H_2$

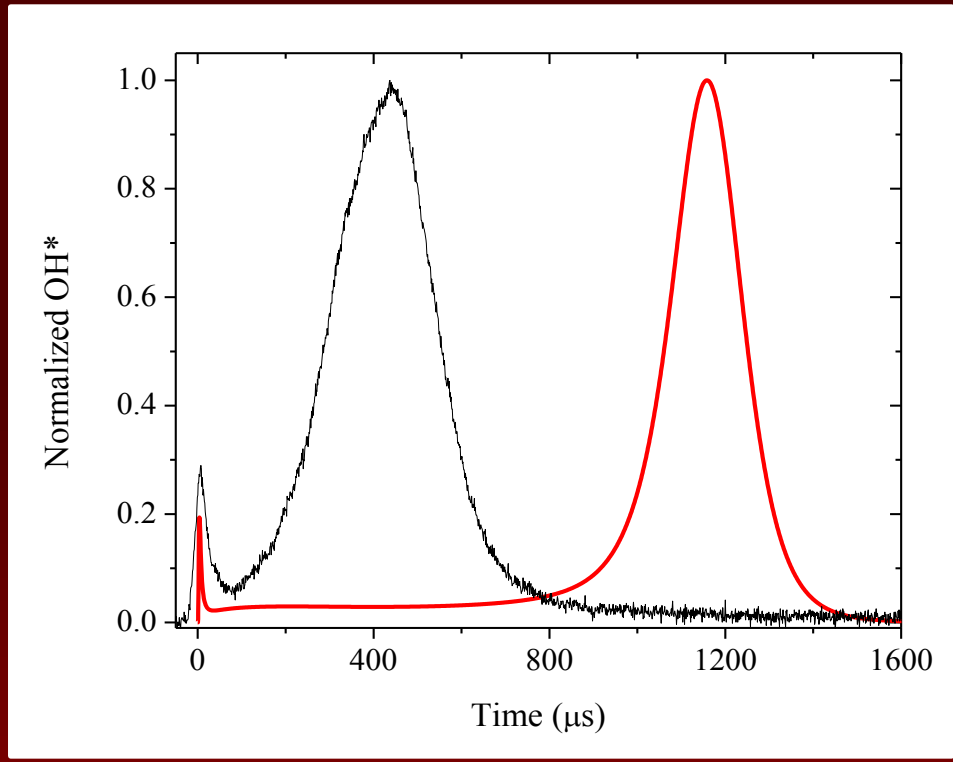
OH* production: $CH + O_2 \rightarrow OH^* + CO$

Chemiluminescence

Fuel-Rich (ϕ = 2.0) OH Chemiluminescence Profile Comparison



Initial Model Prediction of OH* Compared to Experimental Profile



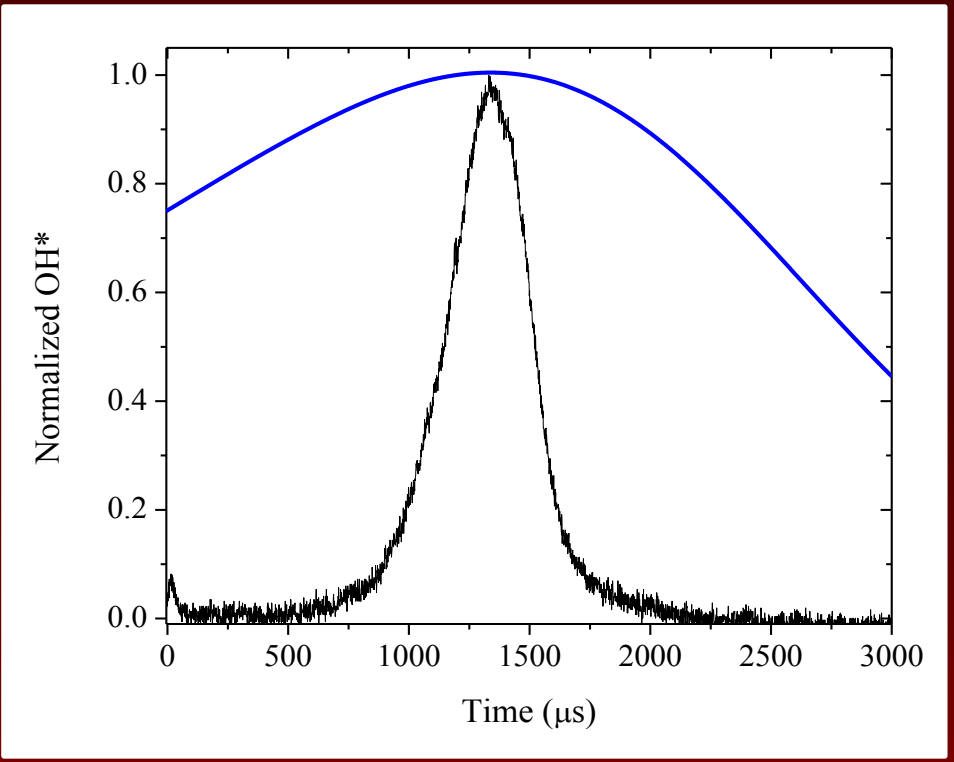
Present Model Prediction of OH* Compared to Experimental Profile

ϕ = 2.0; 0.1% n-C_9H_{20}, 0.7% O_2, 99.2% Ar

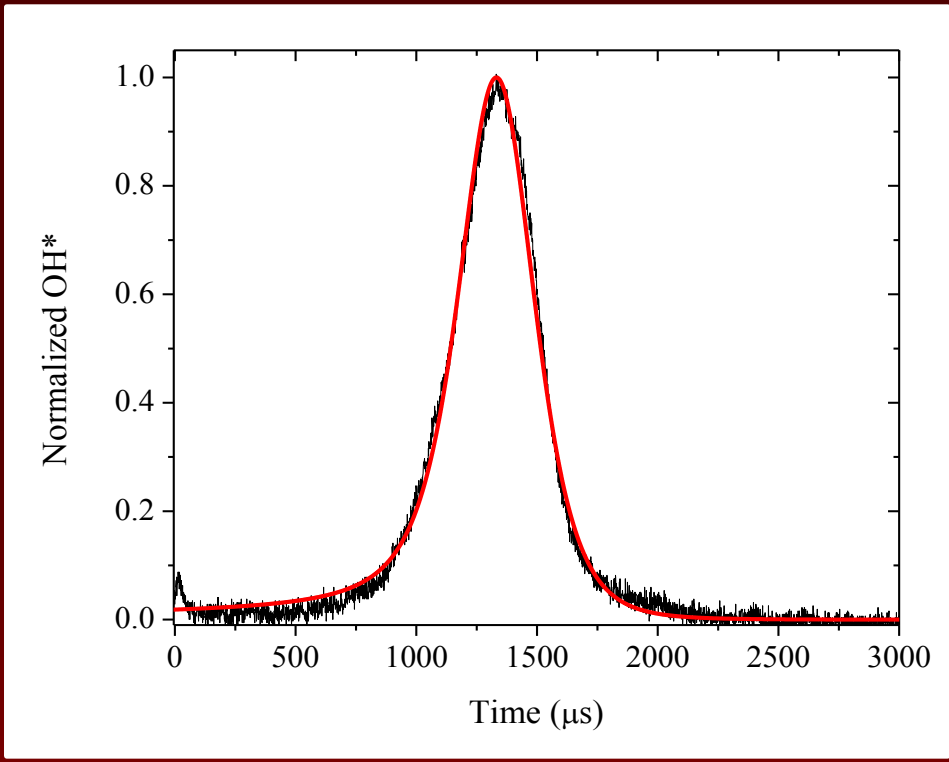
T = 1555 K, P = 1.5 atm

Chemiluminescence

Fuel-Rich (ϕ = 2.0) OH Chemiluminescence Profile Comparison



Initial Model Prediction of OH* Compared to Experimental Profile



Present Model Prediction of OH* Compared to Experimental Profile

ϕ = 2.0; 0.1% n-C_9H_{20}, 0.7% O_2, 99.2% Ar

T = 1412 K, P = 1.5 atm

Challenges and Future Efforts

Challenges

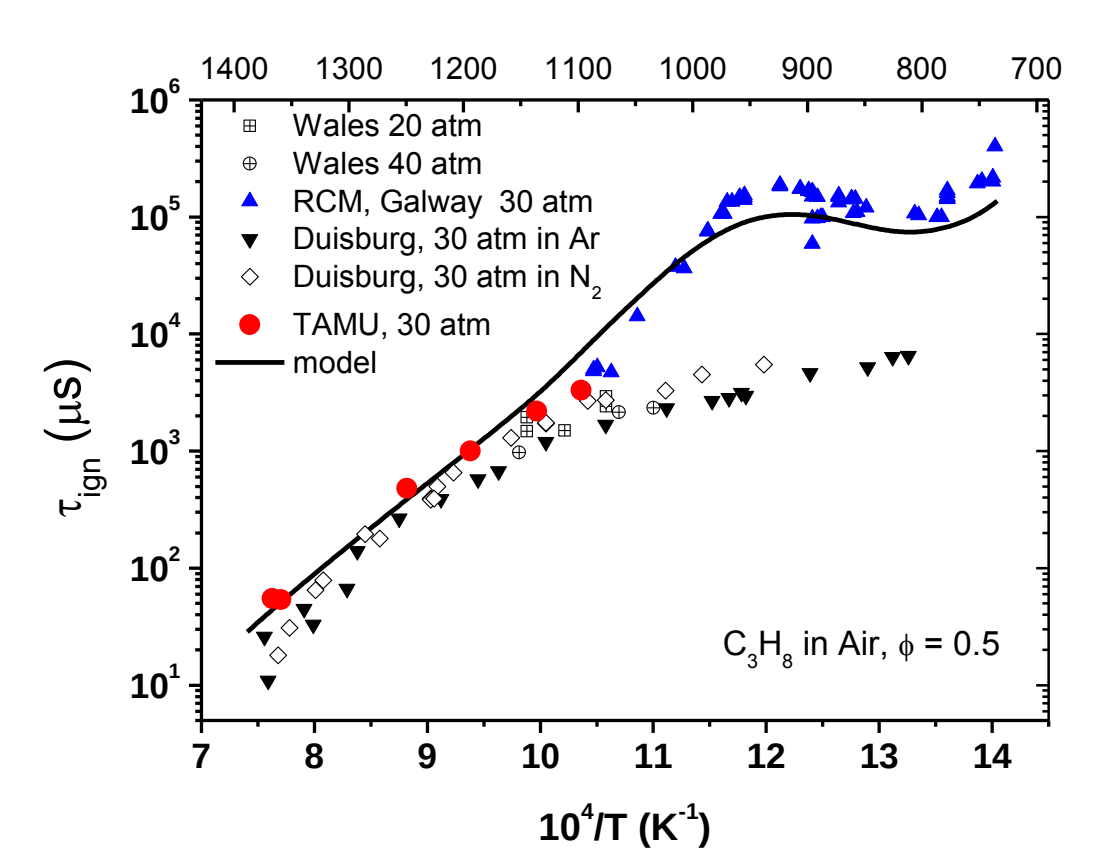
There are Some Challenge Areas for Future High-Pressure Shock Tube Experiments

1. Non-Ideal and Non-Homogeneous Effects
2. Aerosol Shock Tube
3. Laser Measurements at Elevated Pressure
4. Other?

Non-Homogeneous Effects

C_3H_8/Air Data - Discrepancy between Model and Shock Tube

- Clear offset from shock-tube data at T < 1100 K
- Data from different shock tubes tend to agree
- RCM data agree with model at lower temperatures
- ***Further analysis is needed***



Non-Homogeneous Effects

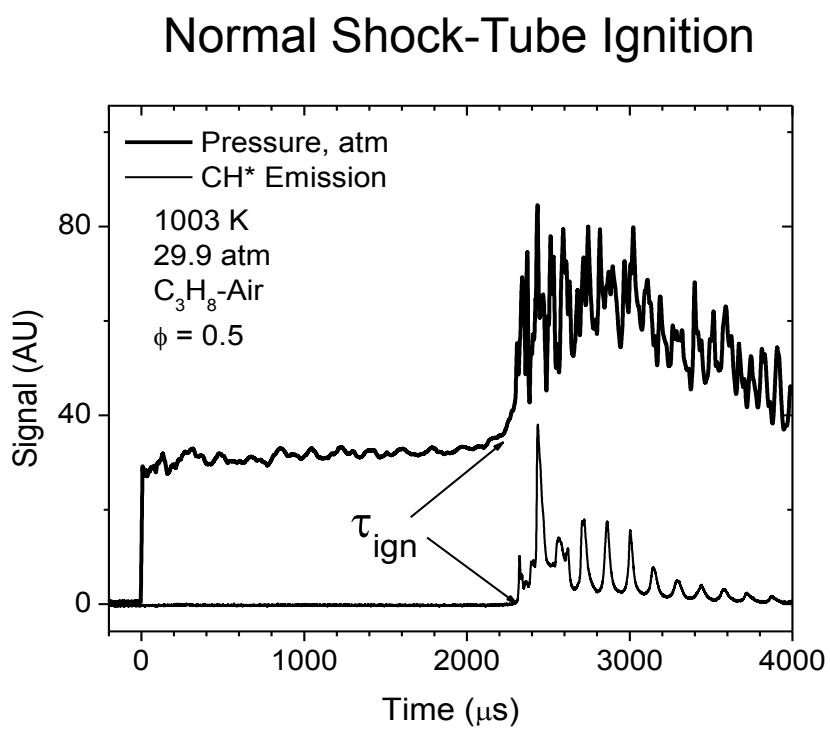
Non-Ideal Pressure Rise Can Come from 2 Sources

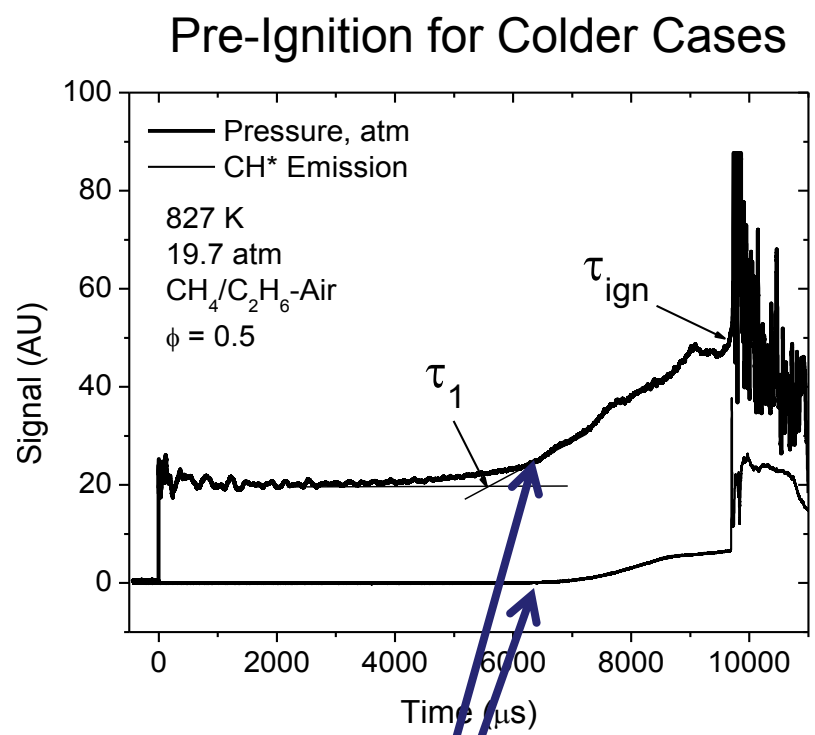
1. Boundary Layer Effects ⇨ <u>Gradual Increase in P</u>

2. Compression Due to Non-Homogeneous Reaction
   ⇨ <u>Marked Increase in Pressure</u>

Non-Homogeneous Effects

Key to the Discrepancy Lies in Observed Pressure Trace

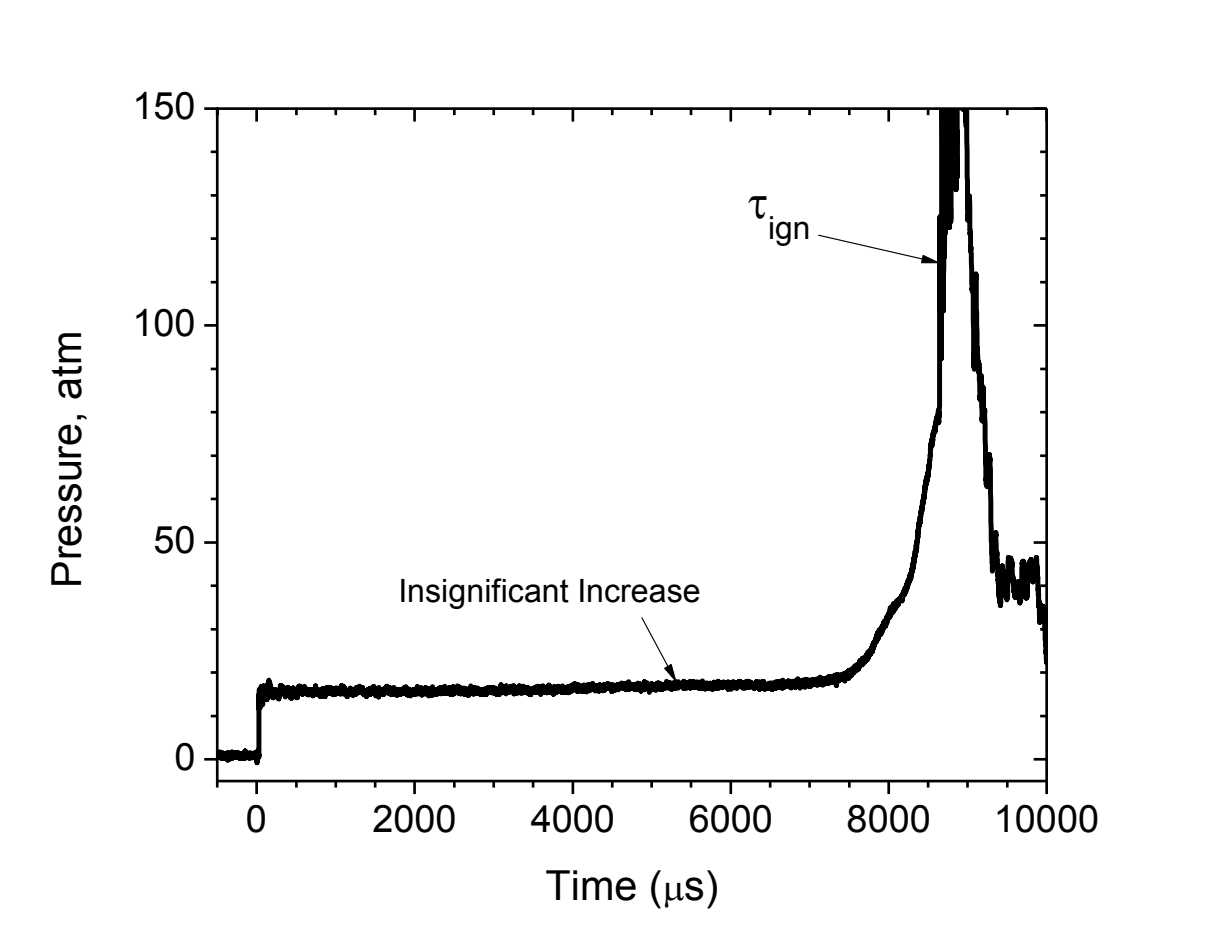




Early Reactivity
Leading to Pressure Rise

Non-Homogeneous Effects

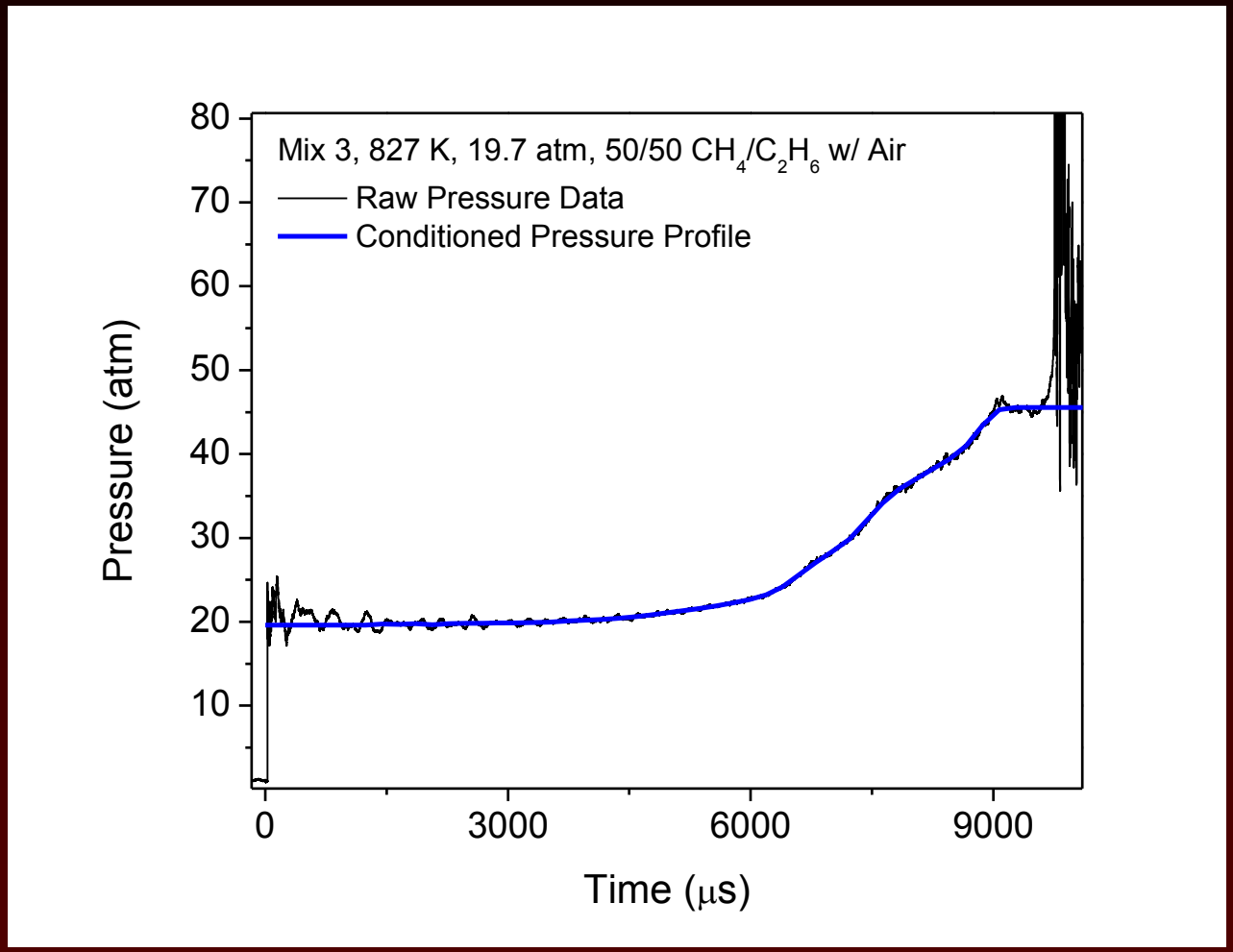
Long-Time Pressure Behavior for Delayed Early Reaction



Extreme Pressure rise before ignition is not from facility-induced boundary layer effects

Non-Homogeneous Effects

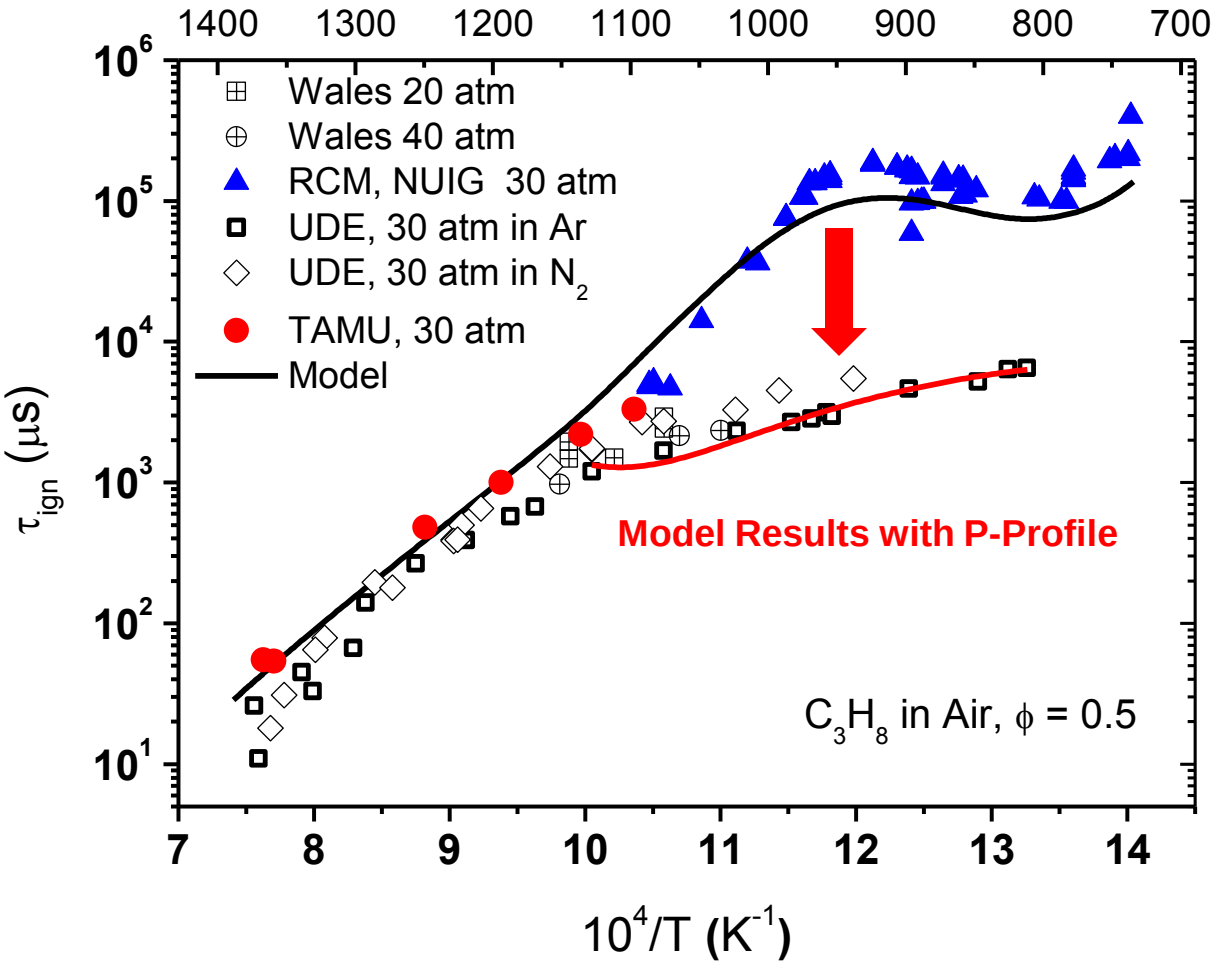
Imposing Pressure Trace on Kinetics Mechanism



Apply Smoothing and Remove Ignition Feature

Non-Homogeneous Effects

Excellent Agreement Found with Imposed P-Profile



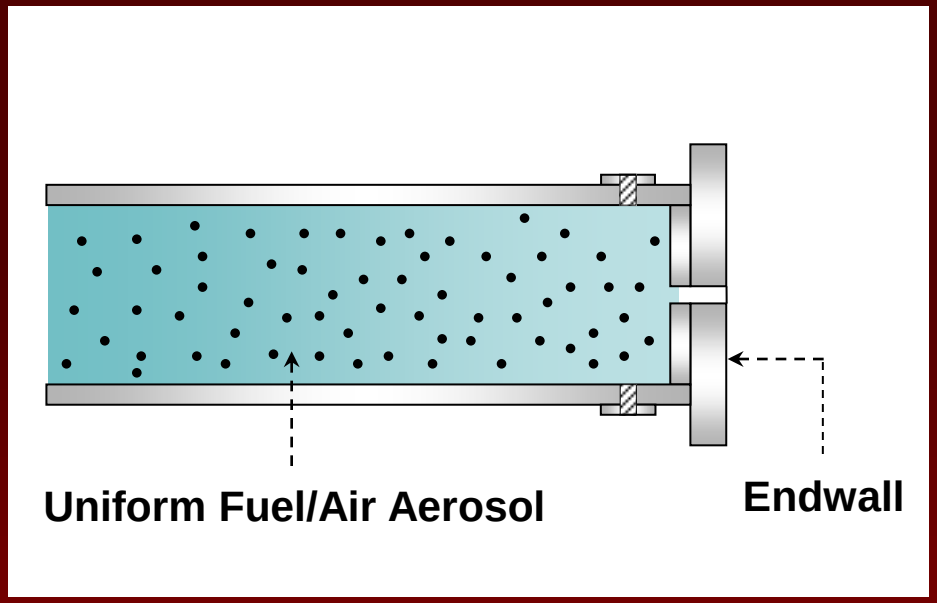
Non-Homogeneous Effects

Challenges for Future Progress in Reactive, High-Pressure Shock Tube Experiments

- Longer Test Times with Negligible (BL) Pressure Rise
- Sources of Early, Non-Homogeneous Ignition?
- Can We Extend Shock Tube to RCM Conditions?

Aerosol Shock Tube

Aerosol Shock Tube Method of Heavy Hydrocarbon Combustion



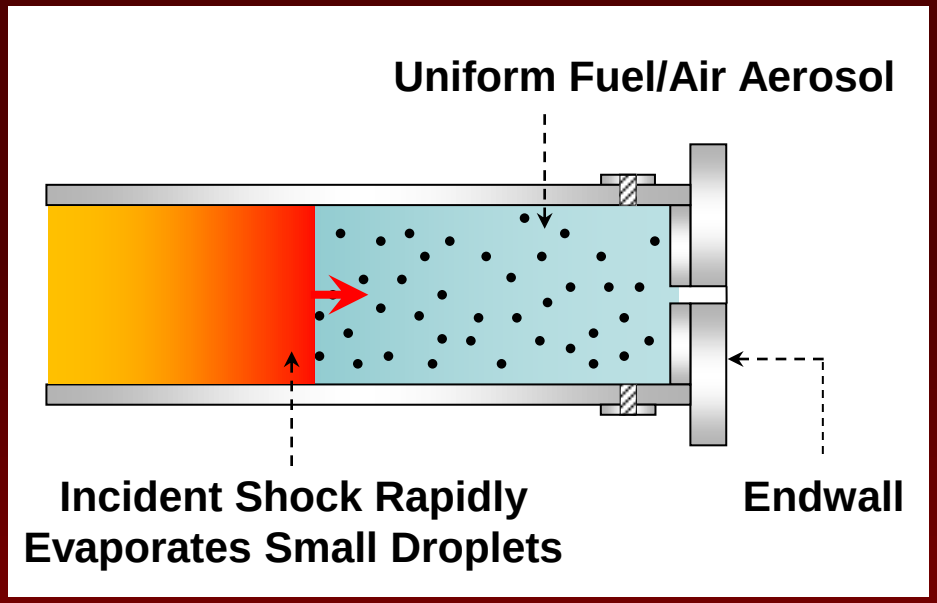
Heterogeneous Shock Tube Experiment
(Gaseous Oxidizer, Liquid Fuel)

Related Issues using Aerosol Approach:

- **Requires Narrow Size Distribution**
- **Small-Diameter Droplets Necessary**
- ***in situ* Measurement of [Fuel]**
- **Low Mass Loading**
- **Concern Over Endwall Fuel-Film Deposition**

Aerosol Shock Tube

Aerosol Shock Tube Method of Heavy Hydrocarbon Combustion



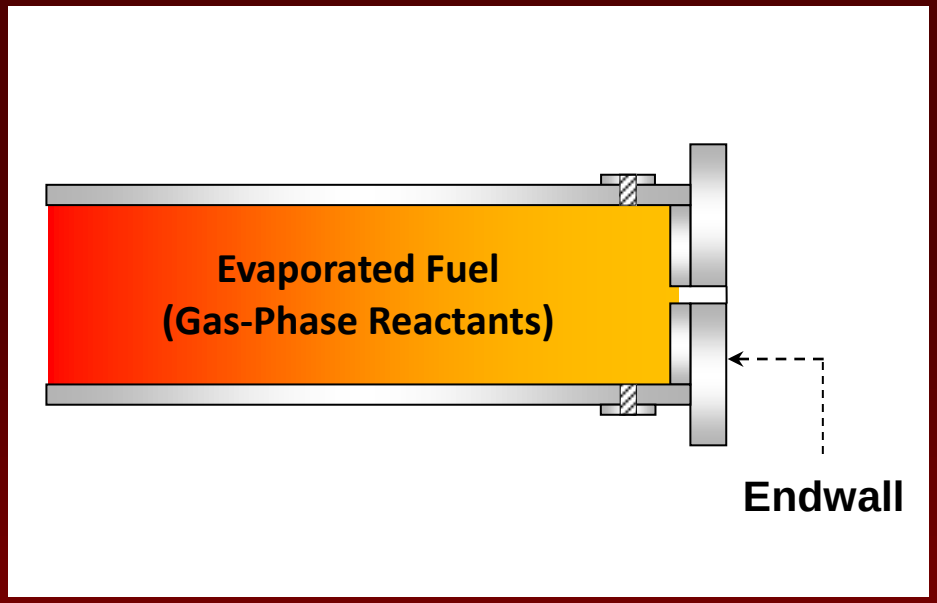
Condensed Fuel Evaporates from High Temperature Created by Shock Wave

Related Issues using Aerosol Approach:

- **Requires Narrow Size Distribution**
- **Small-Diameter Droplets Necessary**
- ***in situ* Measurement of [Fuel]**
- **Low Mass Loading**
- **Concern Over Endwall Fuel-Film Deposition**

Aerosol Shock Tube

Aerosol Shock Tube Method of Heavy Hydrocarbon Combustion



Heterogeneous Experiment becomes Traditional Shock Tube Experiment

Related Issues using Aerosol Approach:

- **Requires Narrow Size Distribution**
- **Small-Diameter Droplets Necessary**
- ***in situ* Measurement of [Fuel]**
- **Low Mass Loading**
- **Concern Over Endwall Fuel-Film Deposition**

Aerosol Shock Tube

Challenges for Future Progress in Aerosol Shock Tubes

- Test-to-Test Repeatability
- Homogeneous Dispersion of Condensed Phase
- Certainty on Test Conditions and Concentrations
- Impact of Particle Size Distribution

Laser Measurements

Challenges for Future Progress in Laser Absorption Measurements at Elevated Pressure

- Isolation of Noise Sources (windows, turbulence, etc.)
- Increased Signal-to-Noise Ratio Needed
- Certainty on Test Conditions and Concentrations

Summary

1. High-Pressure Shock Tube Capabilities
2. Recent and On-Going Studies
3. Chemiluminescence Measurements
4. Challenges and Future Efforts at Elevated Pressures

Shock Tube Group

Dr. Olivier Mathieu

John Pemelton

Christopher Aul

Marissa Brower

Brandon Rotavera

Brian Hogan

Madeleine Kopp

Anthony Levacque

Carmen Osorio

Brandon Marks

Aerospace Corp: Mark Crofton, Andrea Hsu

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