Fundamental Studies to Enable Robust, Reliable, Low Emission Gas Turbine Combustion of High Hydrogen Content Fuels: experimental and computational studies

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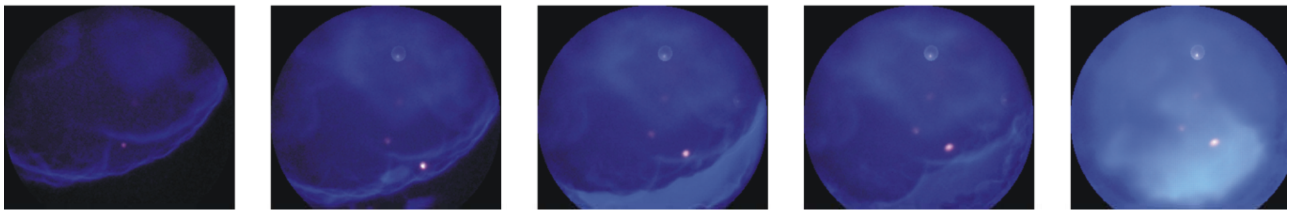
outline



Background and motivation

Low-temperature chemistry of syngas fuels

Experimental plan of work

Computational plan of work

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Syngas offers a clean alternative to coal and natural gas fired power plants

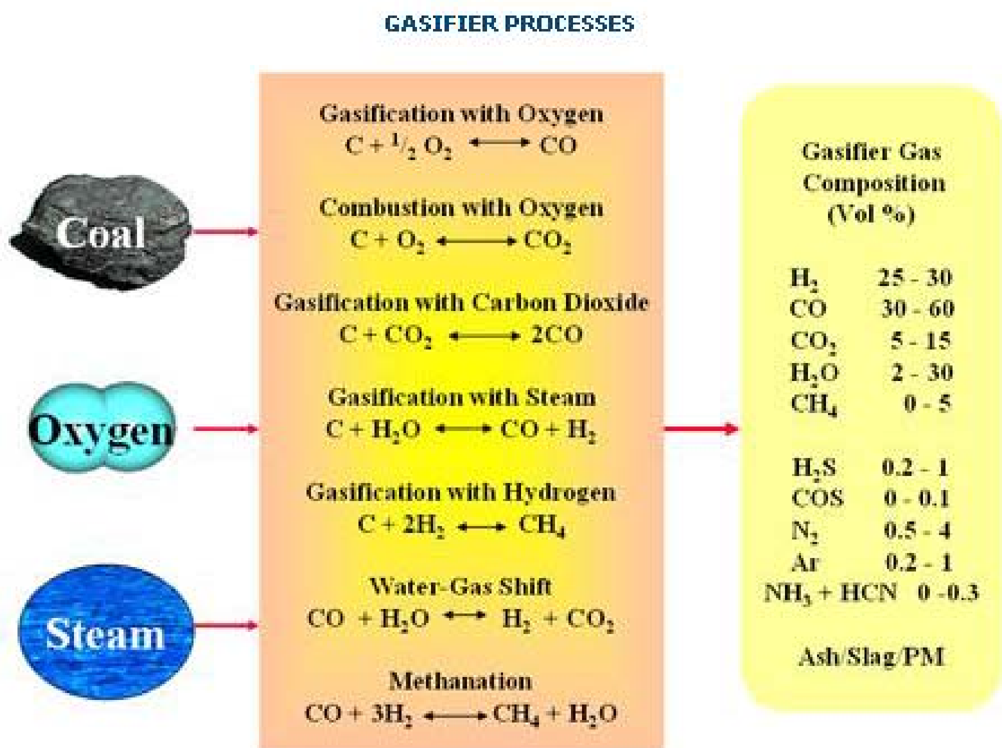
The successful operation of gas turbines using syngas (including hydrogen fuel concentrations >90%) has been demonstrated at numerous facilities in the U.S. and abroad, but challenges remain.

Lean premixed syngas/air offers potential for higher efficiencies while meeting NOx emissions requirements.

Combustor design is complicated by uncertainties in the HHC combustion kinetics that arise due to the high pressure gas turbine conditions.

These uncertainties combined with unique thermodynamic and transport characteristics of hydrogen further complicate accurate prediction and understanding of key combustion metrics, such as the laminar and turbulent burning velocities, and extinction and ignition limits.

These pose significant challenges in identifying effective control strategies for stable operation of gas turbines, especially in the presence of a wide variety in the syngas composition as well as its temporal and spatial fluctuations during the operation.



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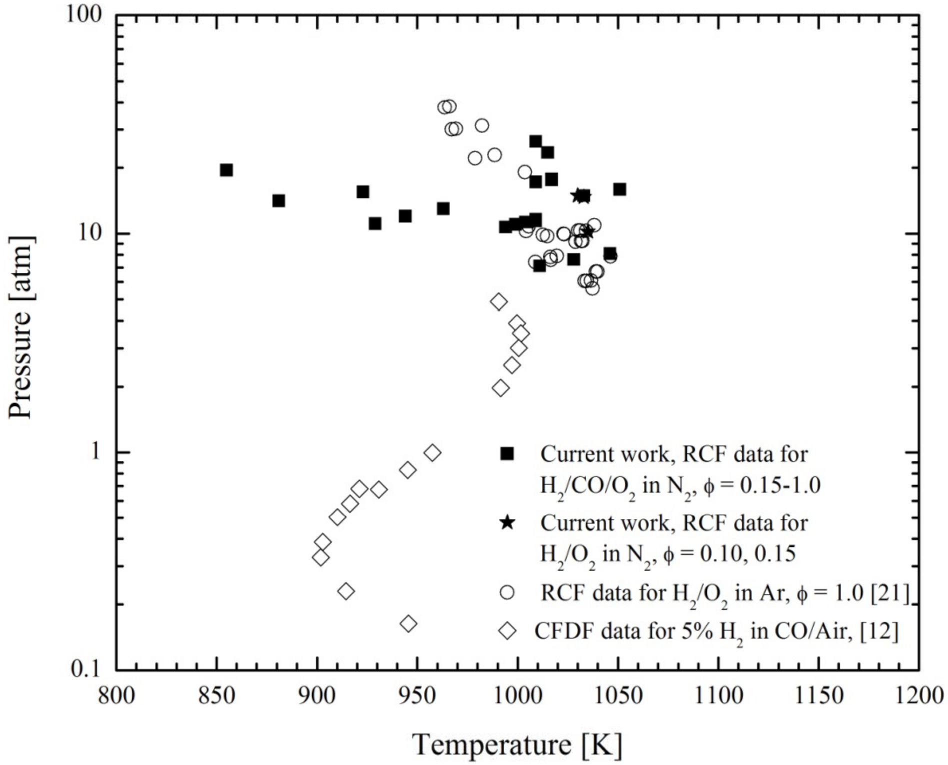
http://www.netl.doe.gov/technologies/coalpower/gasification/gasifipedia/4-gasifiers/4-3_syngas-detail.html

Syngas chemistry introduces particular challenges

- H_2/CO combustion chemistry experiments are highly sensitive to mixture composition including trace impurities.
- Recent studies indicate some reactions are missing from high temperature reaction mechanisms that are important at low temperatures and high pressures.
- 3rd body collisions efficiencies may need refinement.
- Experimental data and modeling approaches are convolved with uncertainties in initial conditions, boundary conditions, and process assumptions, to name a few important considerations.
- Until recently, there were few experimental data for validating syngas combustion kinetics at gas turbine conditions.



Circa 2006, Range of conditions of experimental H_2 and CO ignition studies. The CFDF data of [12] are results of a counterflow diffusion flame study, Walton et al. 2007.

UM fuel chemistry studies

Iso-octane (gasoline chemical surrogate)

- Quantified ignition delay time over a broad range of mixtures and conditions
  — X. He, M.T. Donovan, B.T. Zigler, T.R. Palmer, S.M. Walton, M.S. Wooldridge, A. Atreya, "An experimental and modeling study of iso-octane ignition delay times under homogeneous charge compression ignition conditions," *Combustion and Flame* 142 (2005) 266-275
- Quantified the reactions important in the formation of the radical pool via time-resolved OH measurements
  — X. He, B.T. Zigler, S.M. Walton, M.S. Wooldridge, A. Atreya, "A Rapid Compression Facility Study of OH Time Histories During Iso-octane Ignition," *Combustion and Flame* 145 (2006) 552-570.
- Quantified the reaction pathways controlling fuel consumption via gas-sampling and measurement of over 30 HC species
  — X. He, S.M. Walton, B.T. Zigler, M.S. Wooldridge, A. Atreya, "Intermediate species measurements of iso-octane ignition ," *International Journal of Chemical Kinetics* (2007), 498-517.
- Identified conditions of inhomogeneous reaction
  — S.M. Walton, X. He, B. T. Zigler, M. S. Wooldridge, and A. Atreya, 2007. "An experimental investigation of iso-octane ignition phenomena," *Combustion and Flame*, 150 (2007), 246-262.

Syngas (H_2/CO (product of biomass and coal gasification))

- Quantified ignition delay time over a broad range of mixtures and conditions
  — S.M. Walton, X. He, B.T. Zigler, M.S. Wooldridge, "An experimental investigation of the ignition properties of hydrogen and carbon monoxide mixtures for syngas turbine applications," *Proceedings of the Combustion Institute* 31 (2007), 3147-3154.

_n_-Heptane (diesel chemical surrogate), _n_-Dodecane, Farnesane ($C_{15}H_{32}$)

— Karwat, D. M. A., Wagnon, S., Lai, J. Y. W., Wooldridge, M. S., Westbrook, C. K. "An Experimental and Computational Investigation of n-Dodecane Ignition and Chemical Kinetics," AIAA Conference, Orlando, Florida, January 2011.

_n_-Butanol, Methanol, Acetylene, Indolene, Soy-Biodiesel

— Karwat, D. M. A., Wagnon, S., Teini, P. D., Wooldridge, M. S., (2011) "On the chemical kinetics of n-butanol: ignition and speciation studies," *Journal of Physical Chemistry A*, 115, pp. 4909-4921.

Esters: Methyl butanoate, Butyl methanoate, Ethyl Propanoate, Methyltrans3hexenoate, Methyl crotanate

- Walton, S. M., Karwat, D. M., Teini, P. D., Gorny, A., and Wooldridge, M. S., (2011) "Speciation Studies of Methyl Butanoate Ignition," *Fuel*, 90, pp. 1796-1804.
- S. M. Walton, and M. S. Wooldridge, C.K. Westbrook, "An experimental investigation of the structural effects on the auto-ignition properties of two C5 esters," *Proceedings of the Combustion Institute* 32 (2009), 255-262

Program objectives

The proposed research program focuses on three areas to advance syngas turbine design:

1. syngas chemistry
2. fundamental ignition and extinction limits of HHC fuels
3. data distillation for rapid transfer of knowledge to gas turbine design.

The project objectives are:

1. To develop and validate an accurate and rigorous experimental and computational data base of HHC reaction kinetics, flame speeds and flammability limits of HHC fuels including mixtures with high levels of exhaust gases,
2. To develop detailed and reduced HHC chemical mechanisms that accurately reproduce the new experimental data as well as data in the literature,
3. To develop a quantitative understanding of the stability of HHC combustion to fluctuations in the flow field, including the opportunities and challenges of exhaust gas recirculation (EGR) on extinction, ignition and flame stability,
4. To develop domain maps which identify the range of conditions (e.g. % EGR) where HHC combustion can be effected in both positive and negative manners (e.g. expanded/restricted flammability limits).

Technical Approach:

Experimentally quantify ignition delay time as a function of state and mixture conditions

Experimentally quantify key radical species – OH during ignition

Experimentally evaluate flame/autoignition interactions

Computationally validate new reaction mechanisms to predict accurate ignition delay times.

Computationally reproduce and predict the RCF pressure-time histories.

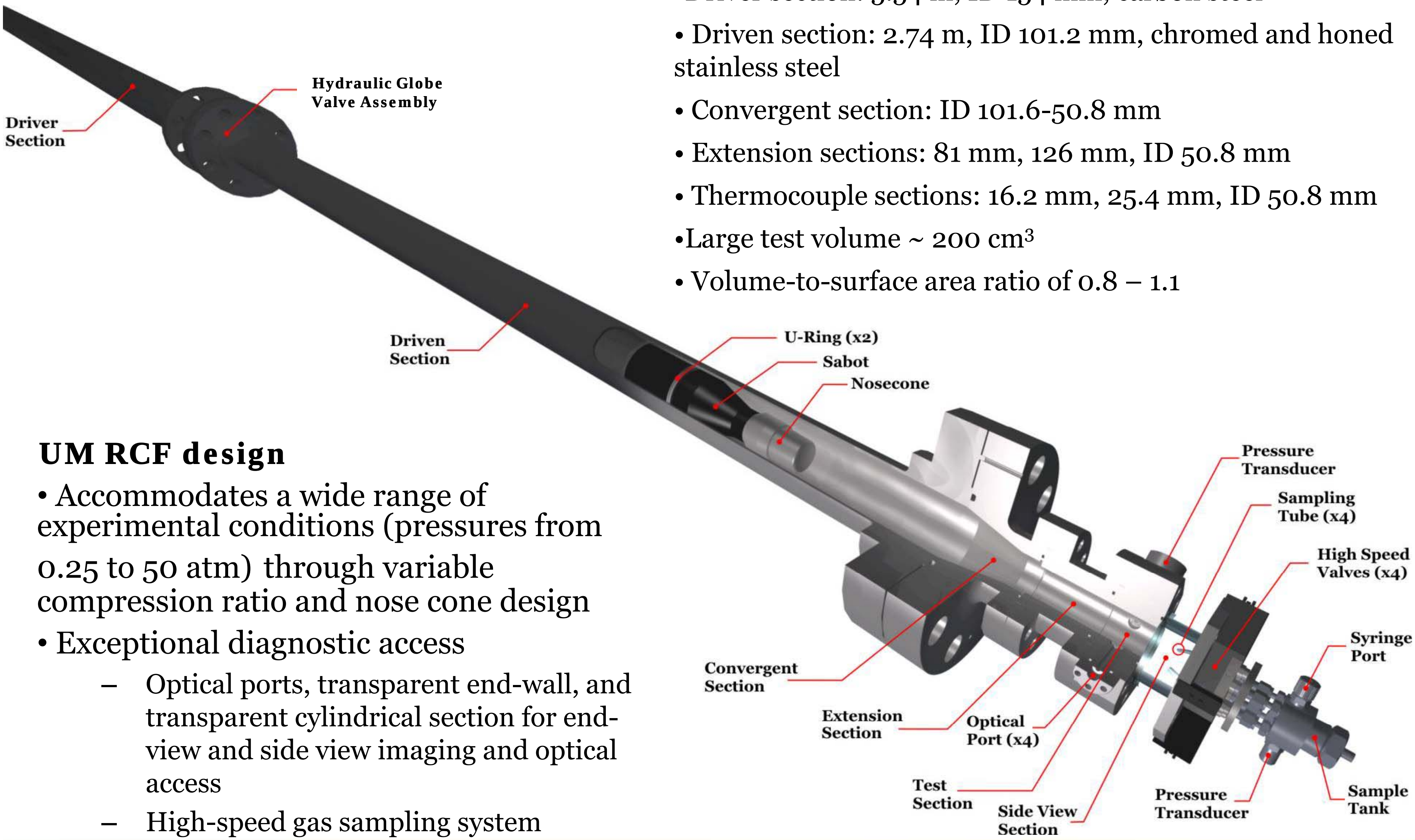
Computationally predict auto-ignition of HHC fuels in the presence of flow and scalar fluctuations.

Computationally predict flame instability dynamics in HHC fuels.

UM rapid compression facility

UM RCF Hardware:

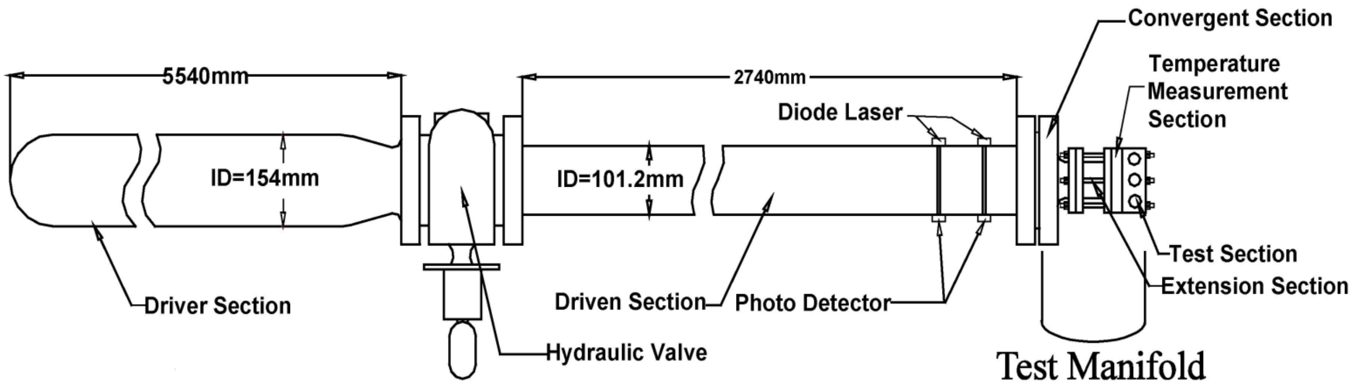
- Driver section: 5.54 m, ID 154 mm, carbon steel
- Driven section: 2.74 m, ID 101.2 mm, chromed and honed stainless steel
- Convergent section: ID 101.6-50.8 mm
- Extension sections: 81 mm, 126 mm, ID 50.8 mm
- Thermocouple sections: 16.2 mm, 25.4 mm, ID 50.8 mm
- Large test volume ~ 200 cm^3
- Volume-to-surface area ratio of 0.8 – 1.1

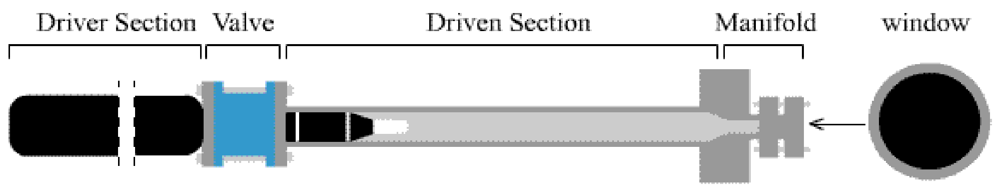


UM RCF design

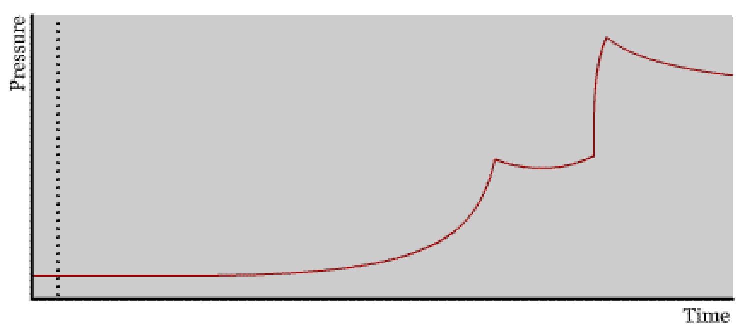
- Accommodates a wide range of experimental conditions (pressures from 0.25 to 50 atm) through variable compression ratio and nose cone design
- Exceptional diagnostic access
  - Optical ports, transparent end-wall, and transparent cylindrical section for end-view and side view imaging and optical access
  - High-speed gas sampling system

The UM rapid compression facility





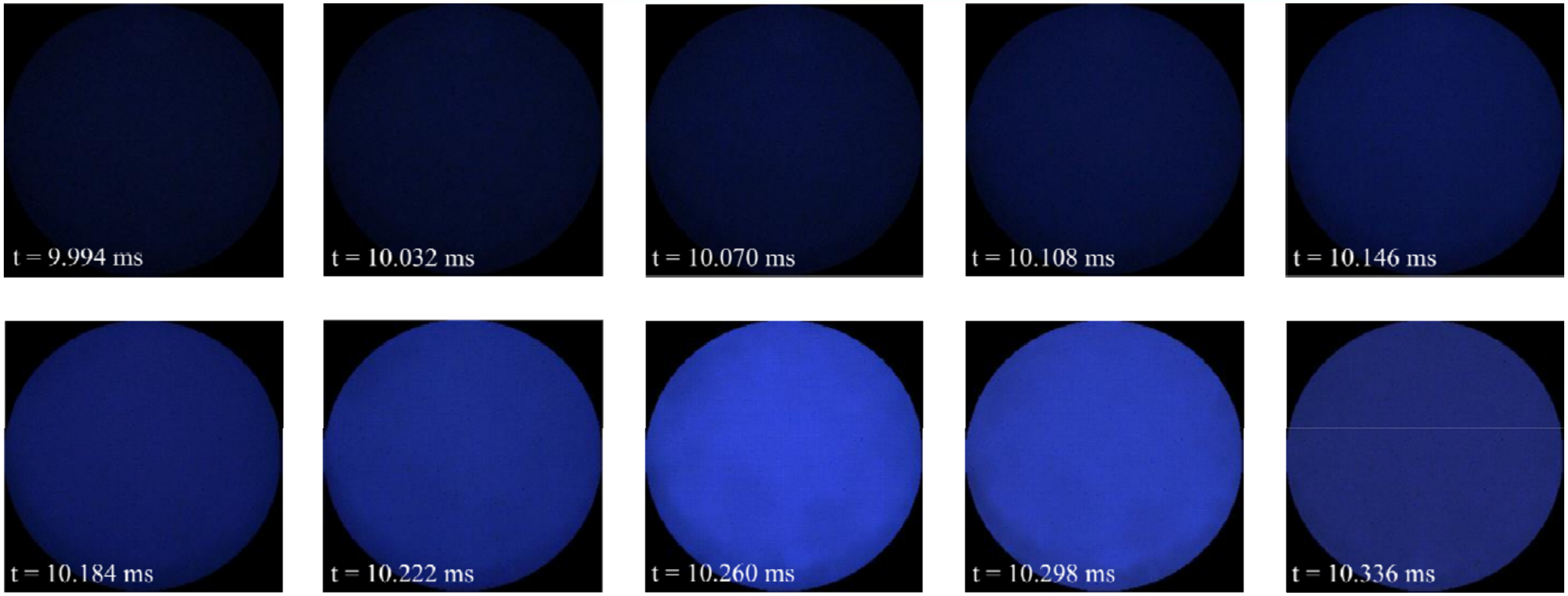
$$P_{eff} = \frac{1}{t_{\frac{dP}{dt}max} - t_{P_{max}}} \int_{t_{P_{max}}}^{t_{\frac{dP}{dt}max}} P\,dt$$

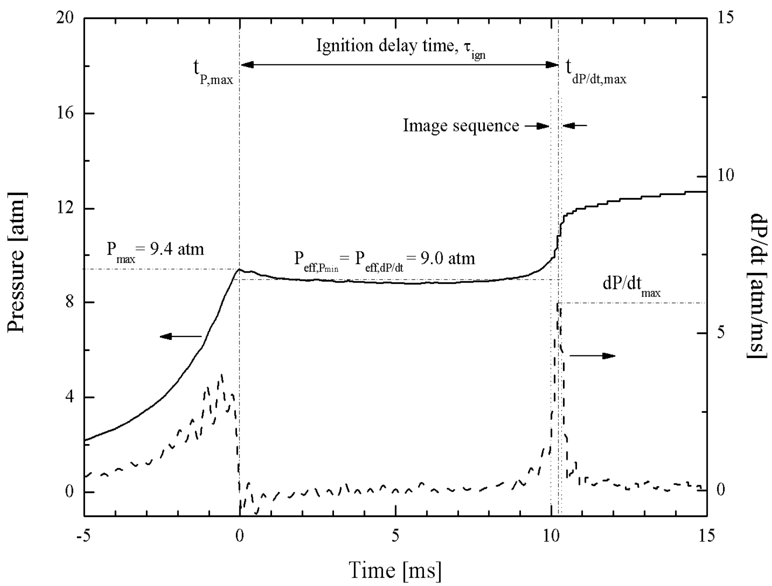


$$\int_{T_0}^{T_{eff}} \frac{\gamma}{\gamma - 1}\, d\ln T = \ln\left(\frac{P_{eff}}{P_0}\right)$$

Donovan, M. T.; He, X.; Zigler, B. T.; Palmer, T. R.; Wooldridge, M. S.; Atreya, A. *Combust. Flame* **2004**, *137*, 351-365.

Typical results for an iso-octane ignition experiment





Experimental conditions of volumetric ignition where ϕ = 0.30, inert/O_2 = 5.00, T_{eff} = 1020 K, P_{eff} = 9.0 atm

S.M. Walton, X. He, B. T. Zigler, M. S. Wooldridge, and A. Atreya, 2007. "An experimental investigation of iso-octane ignition phenomena," *Combustion and Flame*, 150 (2007), 246-262.

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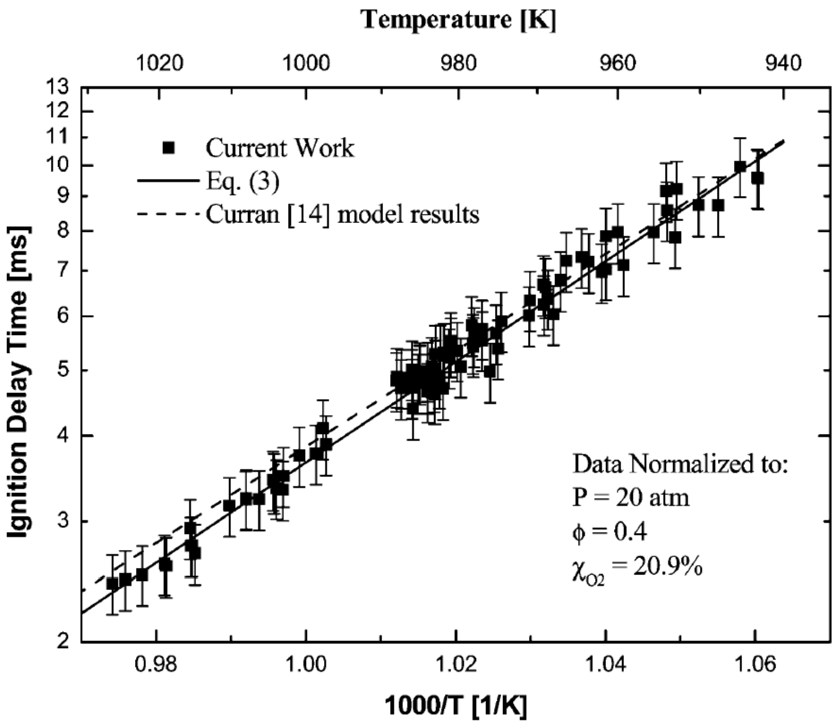
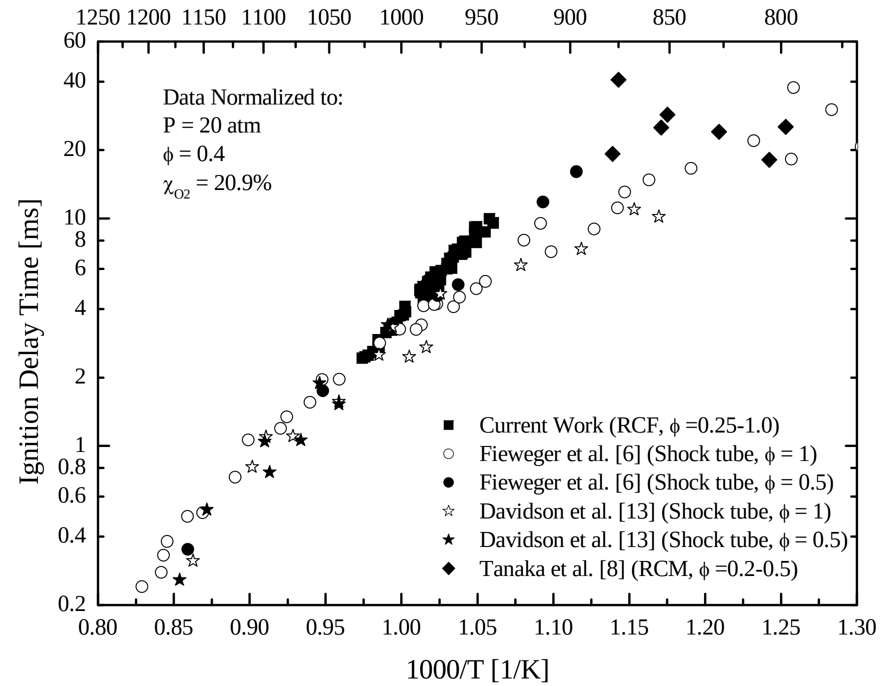
Some global metrics for iso-octane look great



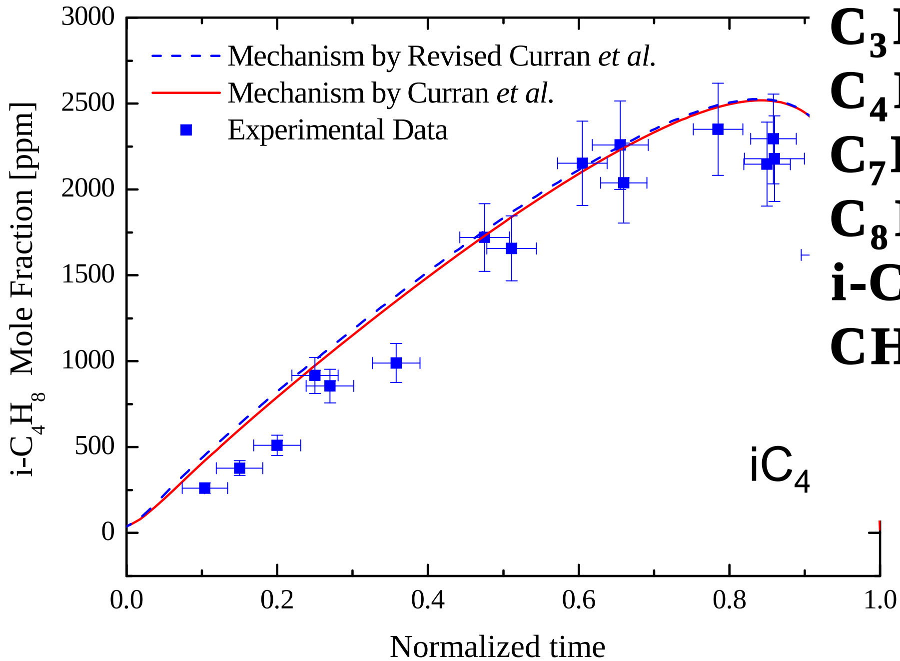


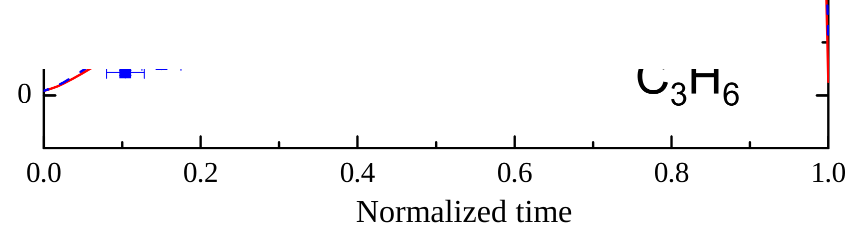
- **Iso-octane has limited negative temperature coefficient behavior**
- **Excellent experimental and computational agreement between vastly different conditions (dilution, pressure, etc.)**
- **RCF and other experimental approaches are highly complementary**

X. He, M.T. Donovan, B.T. Zigler, T.R. Palmer, S.M. Walton, M.S. Wooldridge, A. Atreya, “An experimental and modeling study of iso-octane ignition delay times under homogeneous charge compression ignition conditions,” *Combustion and Flame* 142 (2005) 266-275

And some other metrics are also in excellent agreement…

Measured 23 HC's: **CH_4, C_2H_6, C_2H_4, C_3H_6, a-C_3H_4, p-C_3H_4, i-C_4H_8, 1-C_4H_8, C_4H_6, a-C_5H_{10}, b-C_6H_{12}, o-C_7H_{14}, p-C_7H_{14}, x-C_7H_{14}, y-C_7H_{14}, i-C_8H_{16}, j-C_8H_{16}, i-C_8H_{18}, CH_3CHO, C_3H_6O1-2, i-C_3H_5CHO, i-C_3H_7CHO, t-C_4H_9CHO, CH_3COCH_3, C_3H_5OH, i-C_4H_7OH**





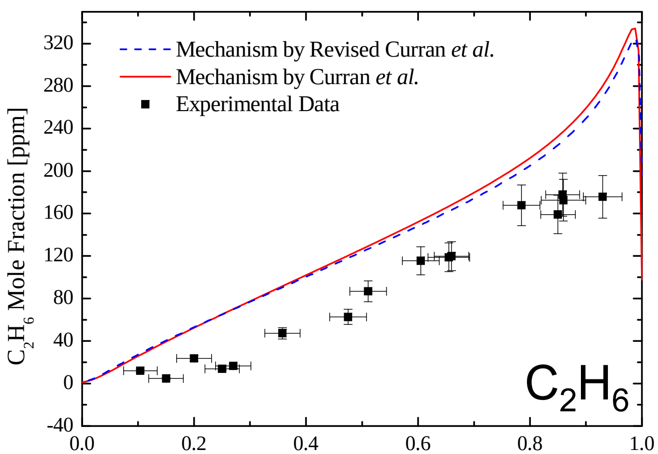
$$\text{i-}C_8H_{18} \longrightarrow \text{x-}\dot{C}_8H_{17} \xrightarrow[\beta\text{-scission}]{\text{Intermediate temperature}} \text{Olefin} + \dot{R}$$

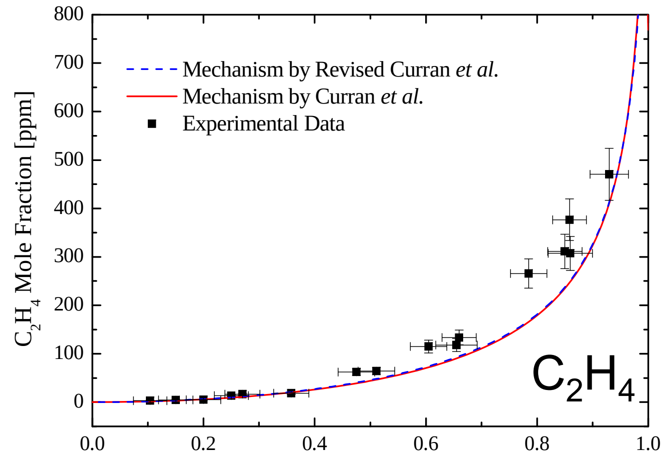
First experimental data to quantitatively demonstrate the reaction path sequence for iso-octane fuel consumption at lean, intermediate temperature conditions

- **iso-octane primarily consumed hydrogen abstraction reactions followed by fragmentation to yield iC_4H_8 and C_3H_6**

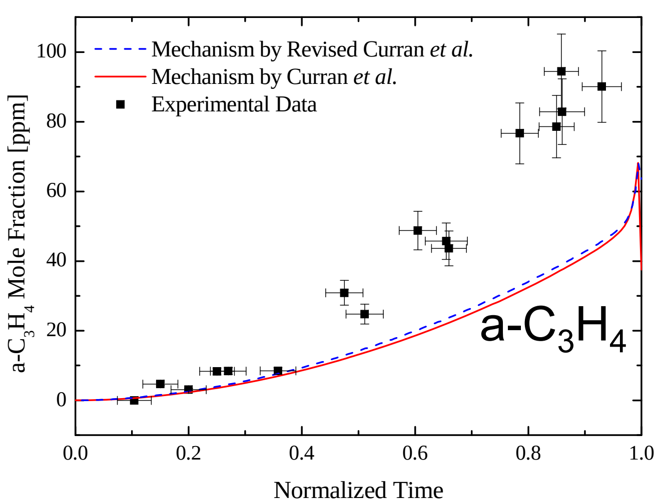
X. He, S.M. Walton, B.T. Zigler, M.S. Wooldridge, A. Atreya, "Intermediate species measurements of iso-octane ignition ," *International Journal of Chemical Kinetics* (2007), 498-517.

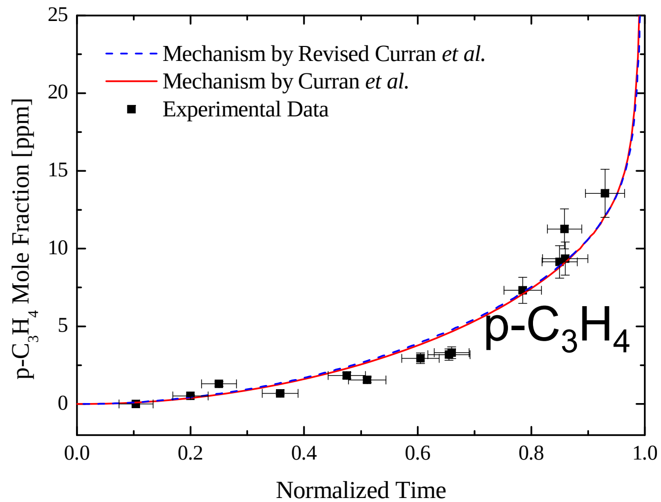
Comparison of iso-octane experimental and modeling results – C2's and C3's





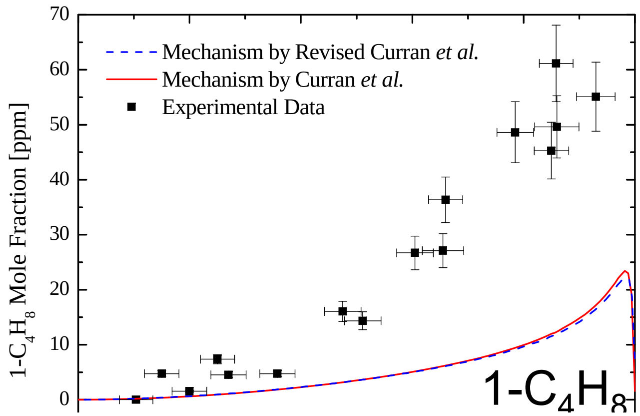
Curran mechanism showed excellent performance at reproducing C2-C3 hydrocarbons.

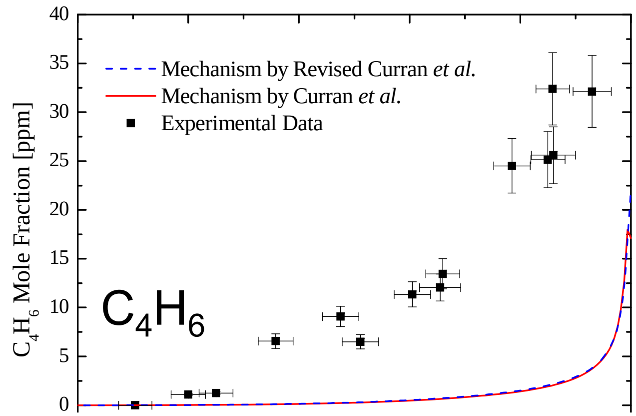




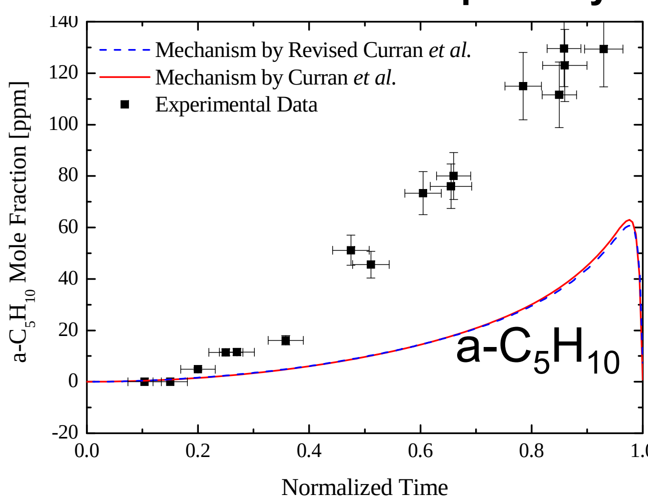
X. He, S.M. Walton, B.T. Zigler, M.S. Wooldridge, A. Atreya, "Intermediate species measurements of iso-octane ignition ," *International Journal of Chemical Kinetics* (2007), 498-517.

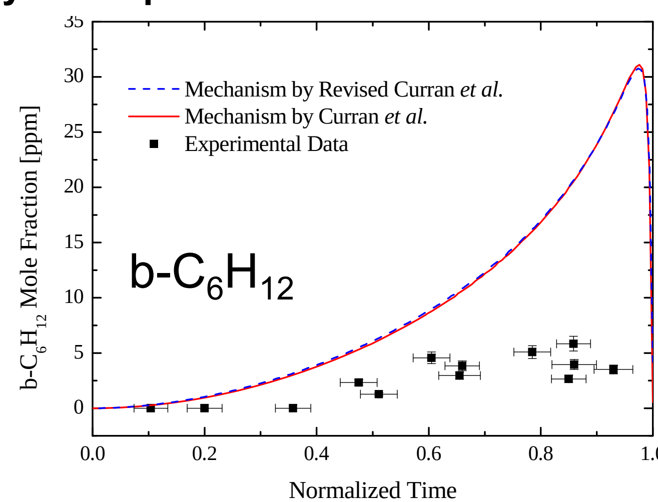
And some metrics differ by large (unacceptable?) margins





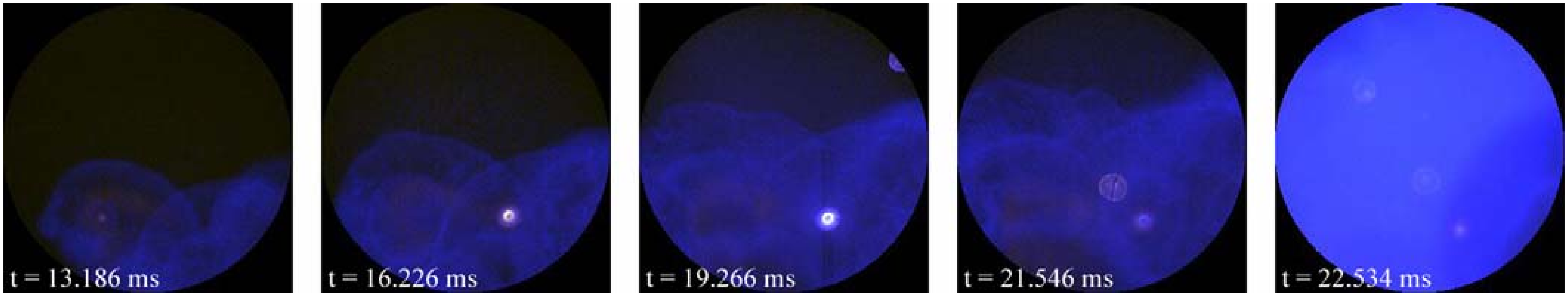
The behavior of some C4-C6 hydrocarbons are not well captured by the mechanism by Curran et al. ← some C7 isomer pathways may be responsible



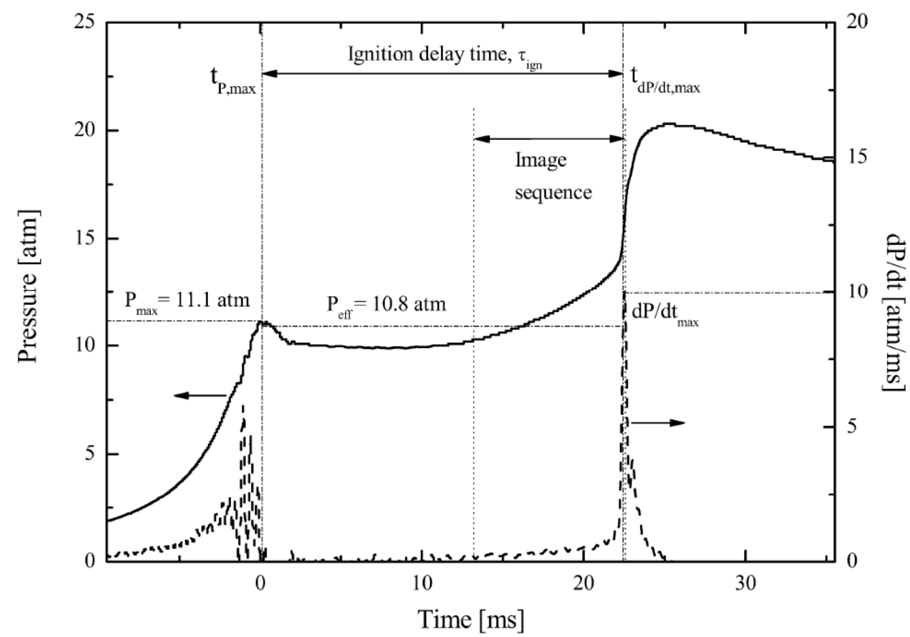


X. He, S.M. Walton, B.T. Zigler, M.S. Wooldridge, A. Atreya, "Intermediate species measurements of iso-octane ignition ," *International Journal of Chemical Kinetics* (2007), 498-517.

These are also typical results for iso-octane ignition



Imaging sequence of weak iso-octane ignition where reaction fronts are present prior to volumetric ignition, ϕ= 0.20, T_{eff} = 917 K, P_{eff} = 10.8 atm, inert/O_2 = 1.38, τign = 22.5 ms, 26,000 fps (color adjusted for clarity). Not all frames in the imaging sequence are presented.

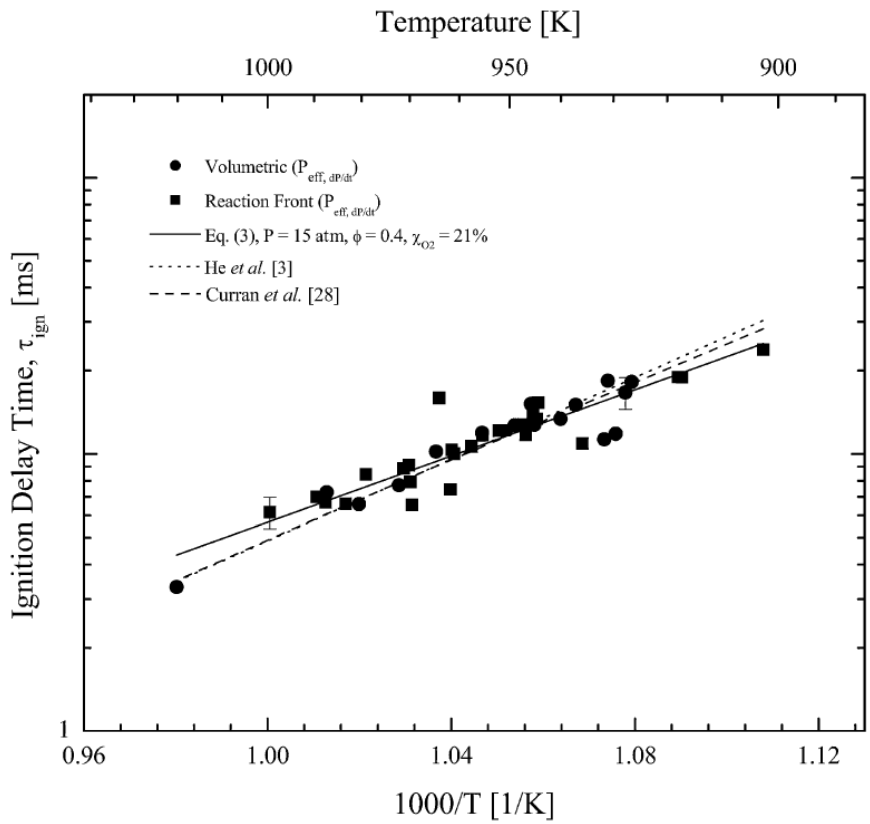


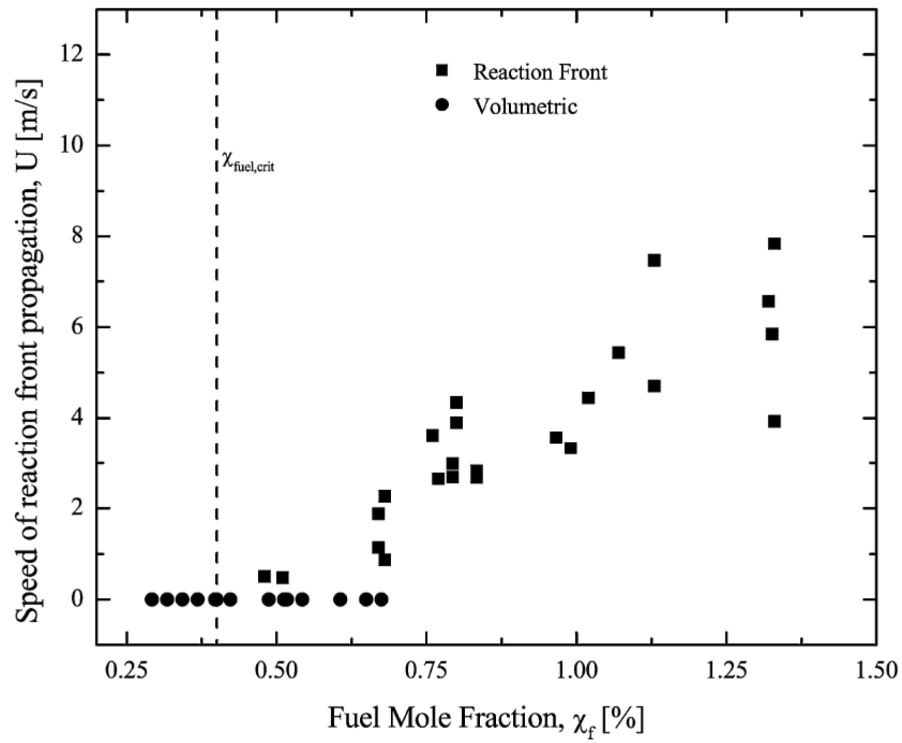
S.M. Walton, X. He, B. T. Zigler, M. S. Wooldridge, and A. Atreya, 2007. "An experimental investigation of iso-octane ignition phenomena," *Combustion and Flame*, 150 (2007), 246-262.

What causes the transition from weak to strong ignition conditions...?

And how much does it matter?

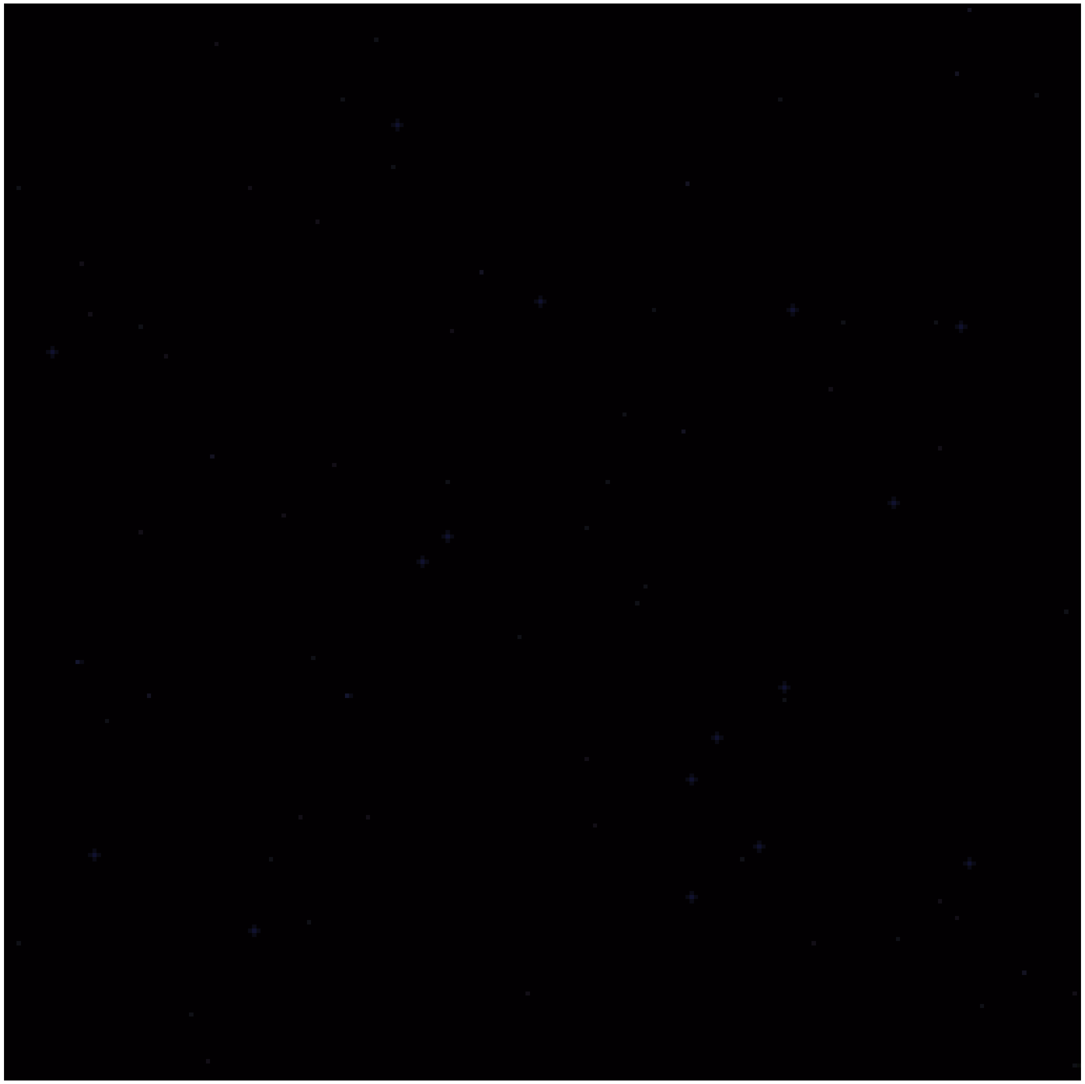
S.M. Walton, X. He, B. T. Zigler, M. S. Wooldridge, and A. Atreya, 2007. "An experimental investigation of iso-octane ignition phenomena," *Combustion and Flame*, 150 (2007), 246-262.





For iso-octane: 1. ignition delay time can be captured using time integrated state conditions. 2. The most critical parameter appears to be the fuel mole fraction, not fuel concentration, ϕ, or O_2 mole fraction.

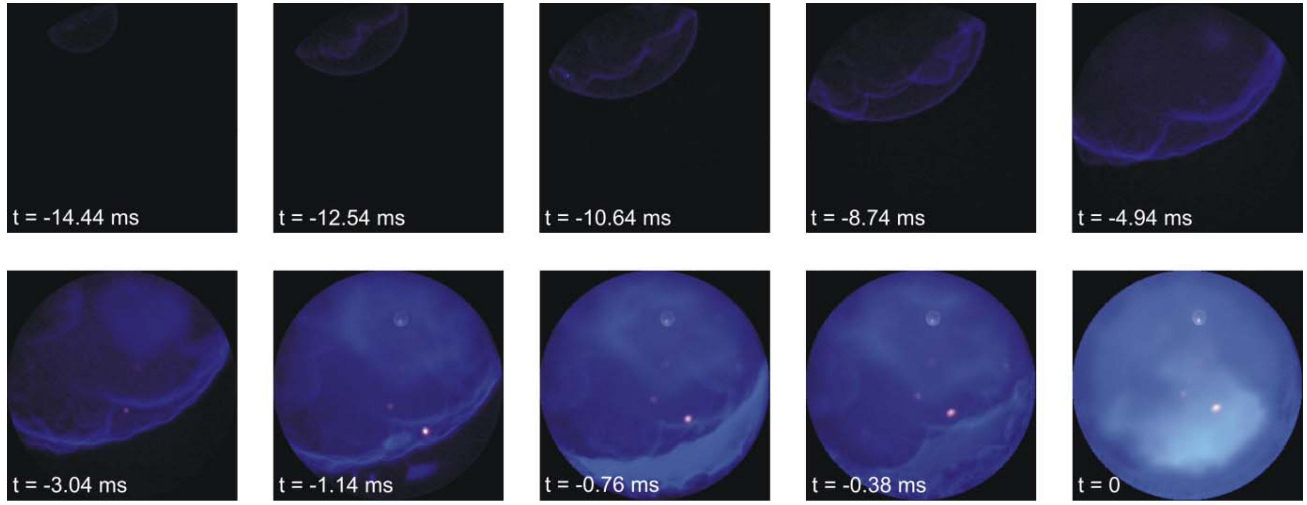
High speed imaging of syngas ignition



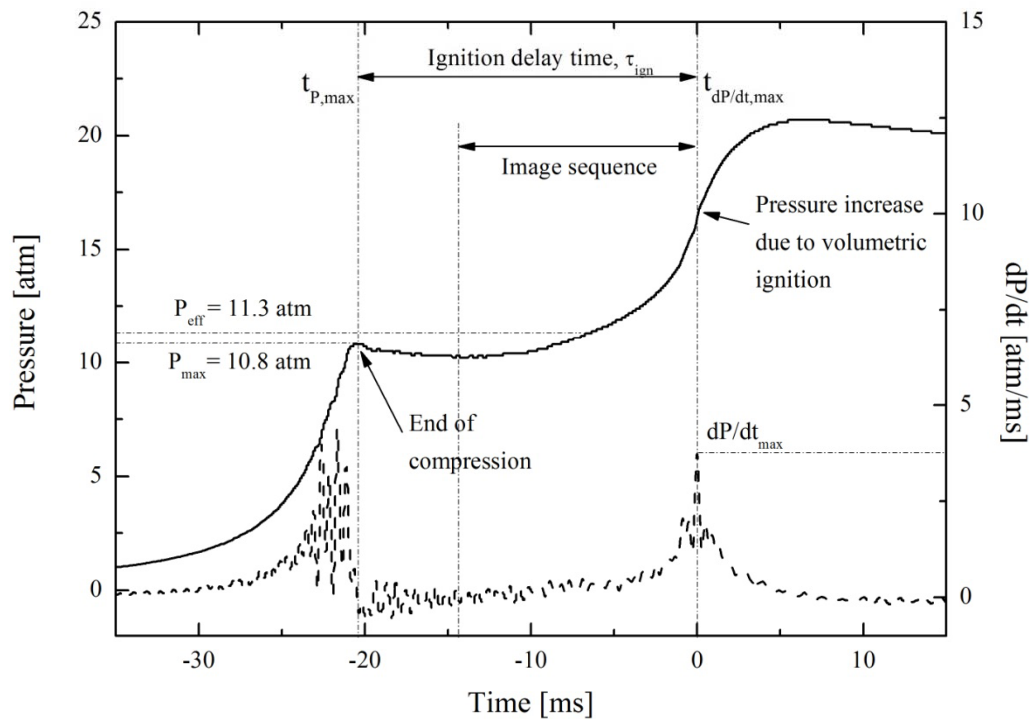
Digital imaging, high-speed color digital video camera: Vision Research, Phantom V7.1, SR-CMOS 48 bit color array, 26,000 fps, 256× 256 pixels, exposure time of 38 μs, each pixel in the CMOS array imaging focused light from a volume with a height × width × depth of approximately 198 μm × 198 μm × 2 mm.

Results of previous UM RCF ignition studies of syngas mixtures

High speed imaging data document syngas ignition characteristics

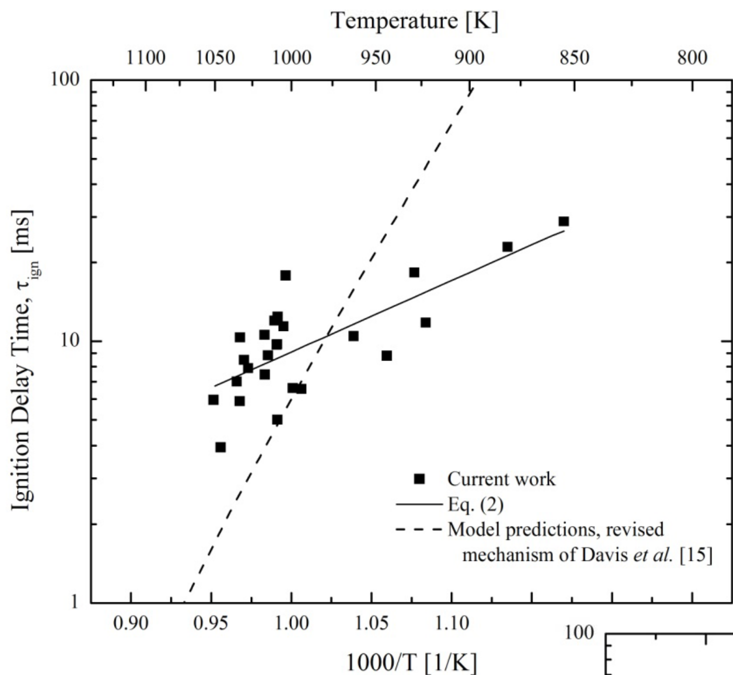


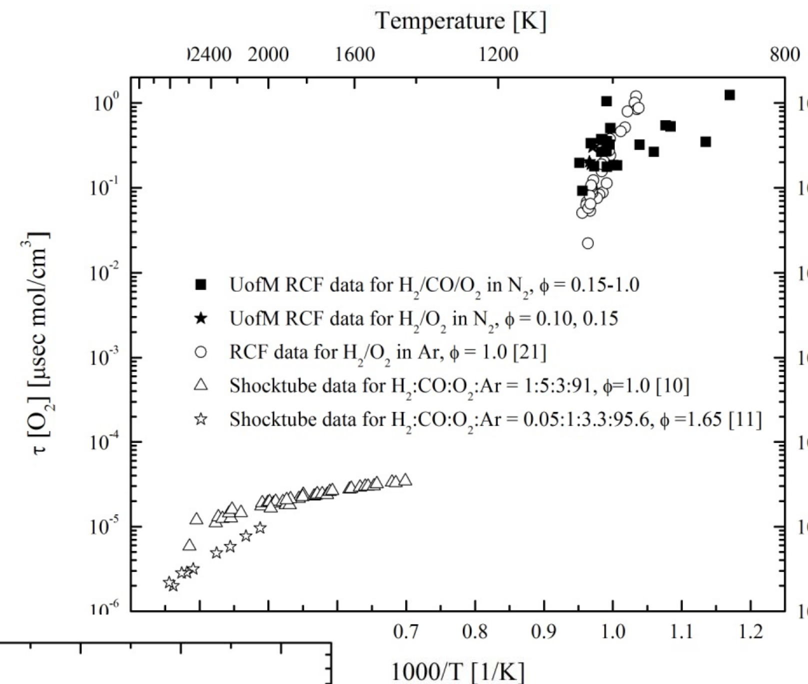
Typical experimental results
T_{eff} = 1004 K, P_{eff} = 11.3 atm,
ϕ= 0.4
inert:O_2 ratio = 3.76
fuel = 20% H_2/80% CO

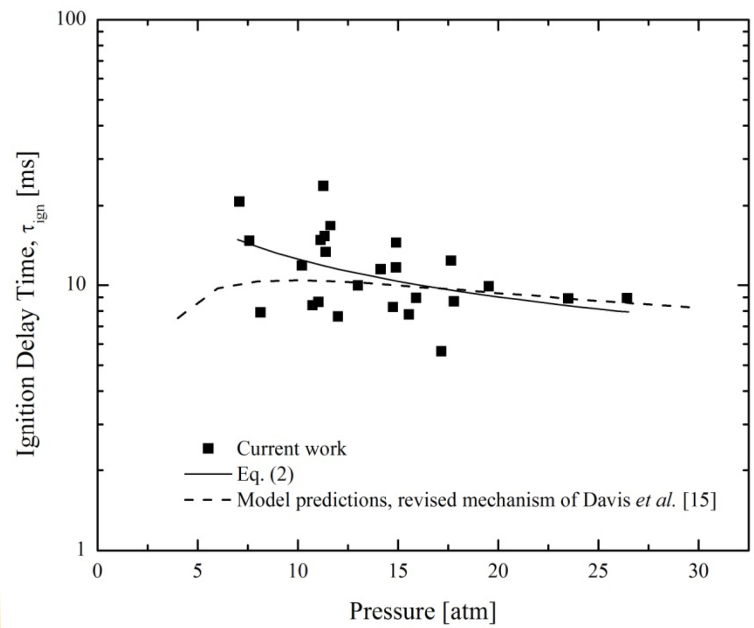


S.M. Walton, X. He, B.T. Zigler, M.S. Wooldridge, "An experimental investigation of the ignition properties of hydrogen and carbon monoxide mixtures for syngas turbine applications," *Proceedings of the Combustion Institute* 31 (2007), 3147-3154.

Summary of previous syngas UM RCF ignition data







S.M. Walton, X. He, B.T. Zigler, M.S. Wooldridge, "An experimental investigation of the ignition properties of hydrogen and carbon monoxide mixtures for syngas turbine applications," *Proceedings of the Combustion Institute* 31 (2007), 3147-3154.

Focus of Gen2 UM RCF syngas ignition studies

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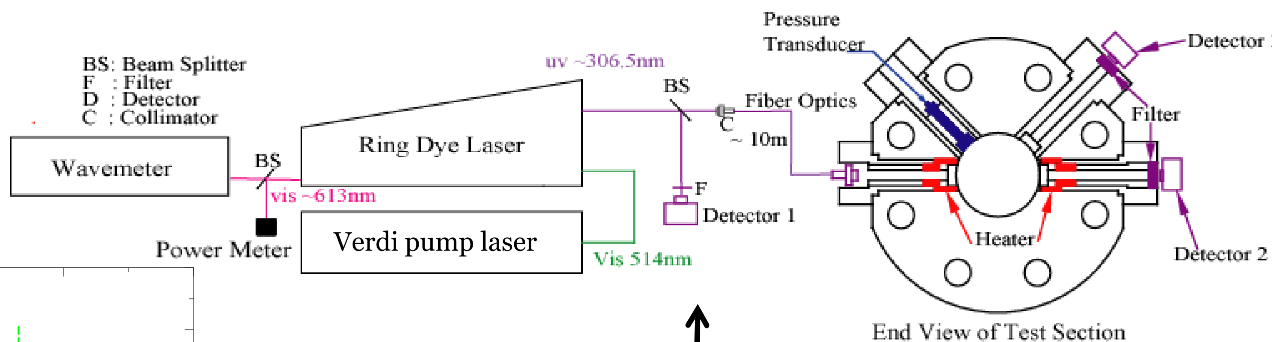
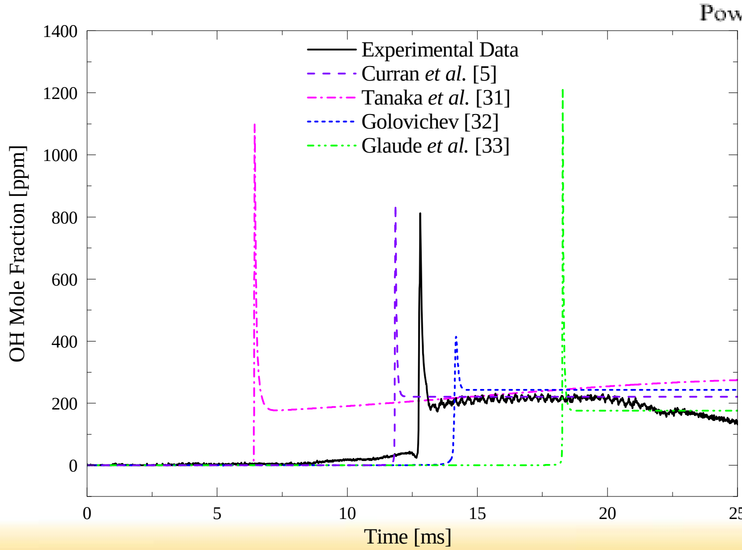
Range of experimental conditions of interest

Experimental parameter	Range of interest
temperature	640 – 1730 K
pressure	10-25 atm
H_2:CO ratio	0.3 - 2, 100% H_2
CH_4 content	0-10%
trace impurities	0-100 ppm H_2S, 0-100 ppm SiH_4, 0-100 ppm NH_3, etc
ϕ	0.2 – 1.0
H_2O addition	0.1 – 10%*
CO_2 addition	0.5 – 30%
N_2 dilution of H_2	0 – 50%

1. Expand syngas ignition data and quantify strong and weak ignition regimes.
2. Acquire more rigorous chemical kinetic targets of OH time histories and stable intermediates



Schematic of the OH laser absorption diagnostic used in UM-RCF studies



Typical OH ignition data (iso-octane)

X. He, B.T. Zigler, S.M. Walton, M.S. Wooldridge, A. Atreya, "A Rapid Compression Facility Study of OH Time Histories During Iso-octane Ignition," *Combustion and Flame* 145 (2006) 552-570

Parametric studies and validation using high-fidelity simulation

Objectives

- To gain insights into fundamental aspects of autoignition characteristics under HCCI-like conditions
- Study the effects of local temperature (T) and equivalence ratio (Φ) inhomogeneities with different T- Φ correlations
- Understand the role of different heat release modes
- Develop automated diagnostic tools to identify different ignition regimes

Computational Capabilities: Direct Numerical Simulation

- Full compressible Navier-Stokes, species and energy equations solved
- Higher order time integration and spatial differencing for efficient simulation
- Detailed/reduced reaction mechanism, thermodynamic and transport processes
- Structured grid with algebraic stretching
- Robust Navier-Stokes characteristic boundary conditions (NSCBC) for inert, reactive, and solid boundary treatment

Previous work: 2D ignition of hydrogen/air mixture

Bansal and Im, Combustion and Flame (2011)

2D domain: 4.1mm x 4.1mm (960x960)

P = 41 atm

Hydrogen-air detailed mechanism (Mueller et al., 1999)

Prescribed random turbulence

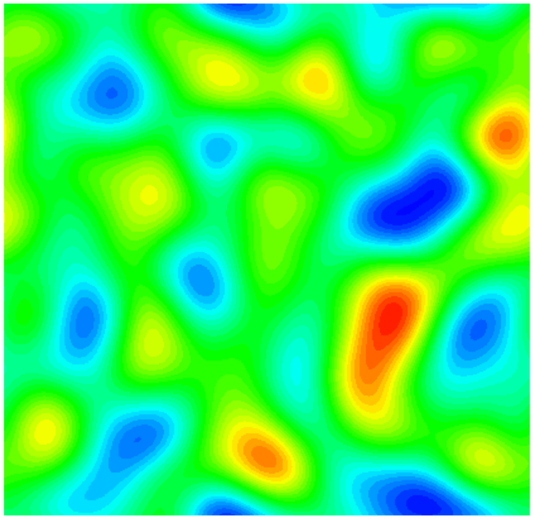
- u' = 0.5m/s, L_{11} = 0.34mm

Prescribed random temperature field

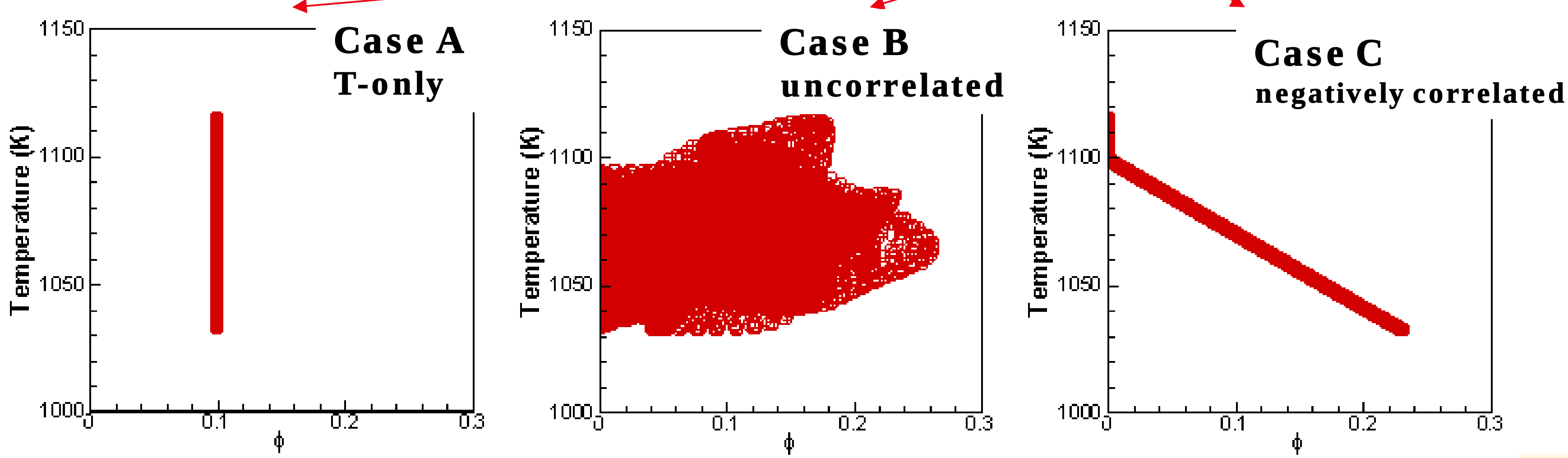
- T_{mean}=1070K, T'=15K

Prescribed random Φ field

- Φ_{mean} = 0.1, Φ' = 0.0



Initial T field (blue: 1033 K, red:1116 K)



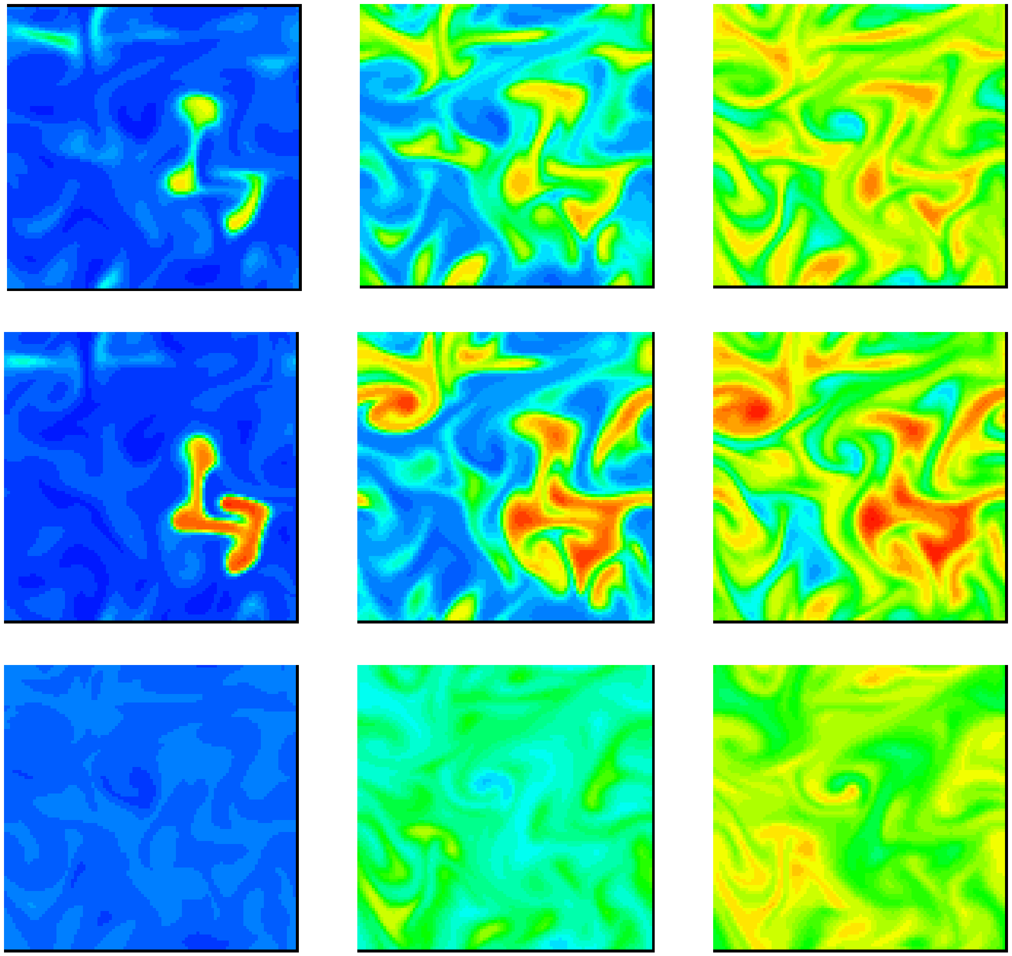
Evolution of temperature

10% | 50% | 90%

Case A

Case B

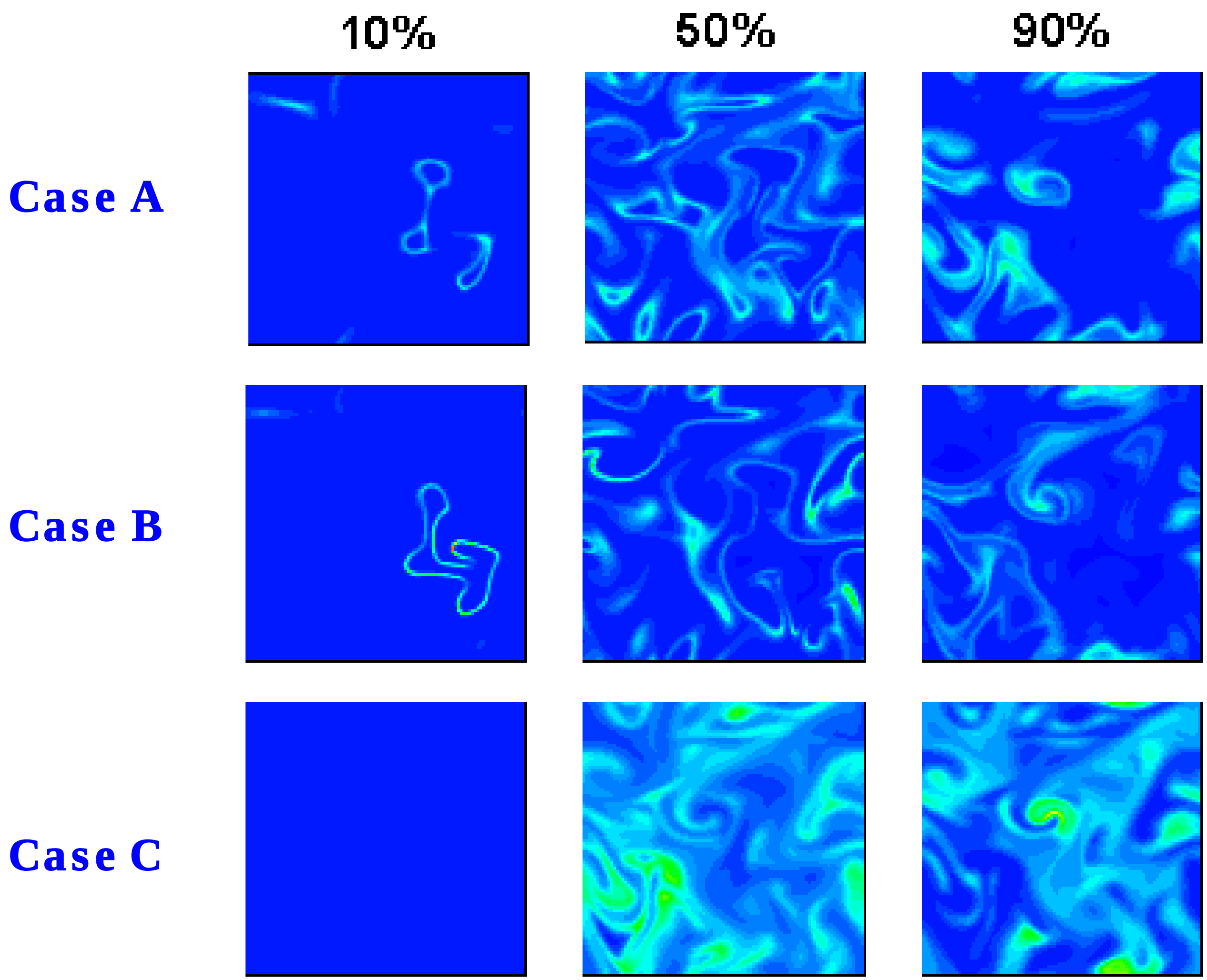
Case C



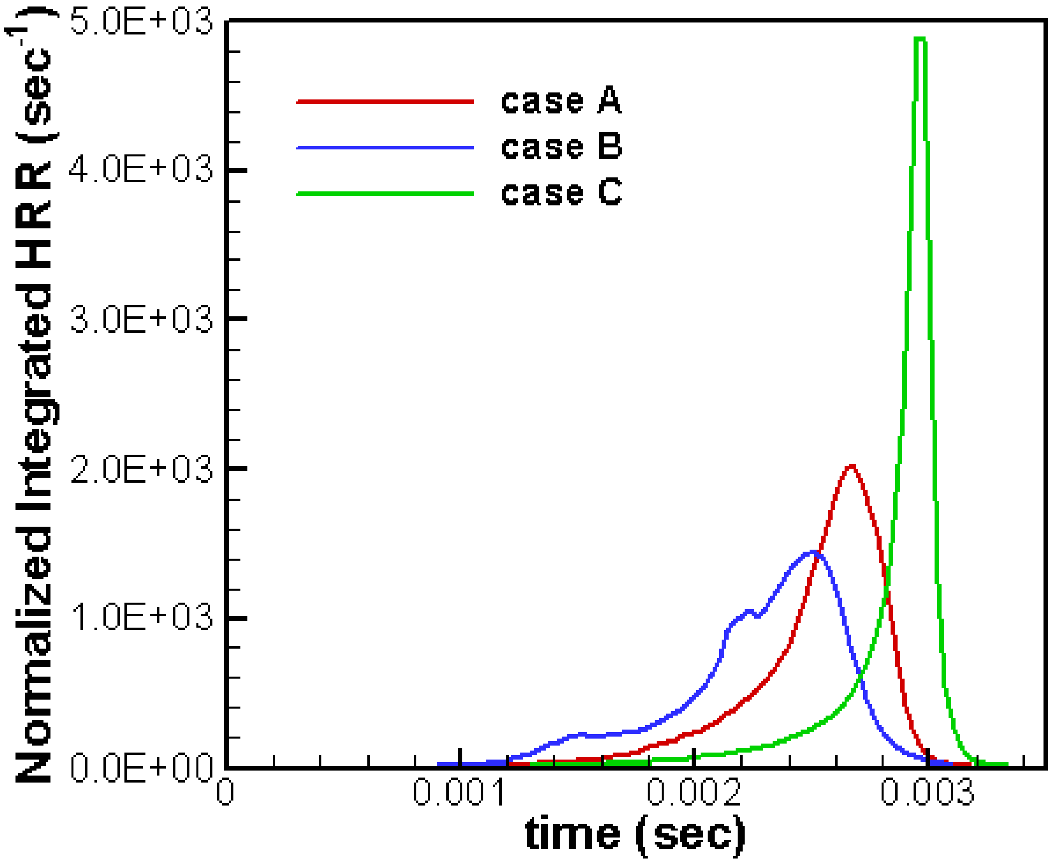
Blue: 1050K

Red: 1620K

Evolution of heat release rate



Time history of integrated heat release rate



Ignition delay:	Case C > Case A > Case B
Peak heat release:	Case C > Case A > Case B

Identifying ignition regimes using computational singular perturbation

Gupta, Im, and Valorani, Proc. Comb. Inst. V. 33 (2011)

Goal: characterize ignition regimes in non-uniform combustion systems relevant to new turbulent combustion sub-models

Approach: Use CSP analysis of timescales in a reacting flow field to determine Importance Index

$$I^T = \left| I^T_{(T-\text{Diffusion})_{slow}} \right| + \left| I^T_{(T-\text{Convection})_{slow}} \right|$$

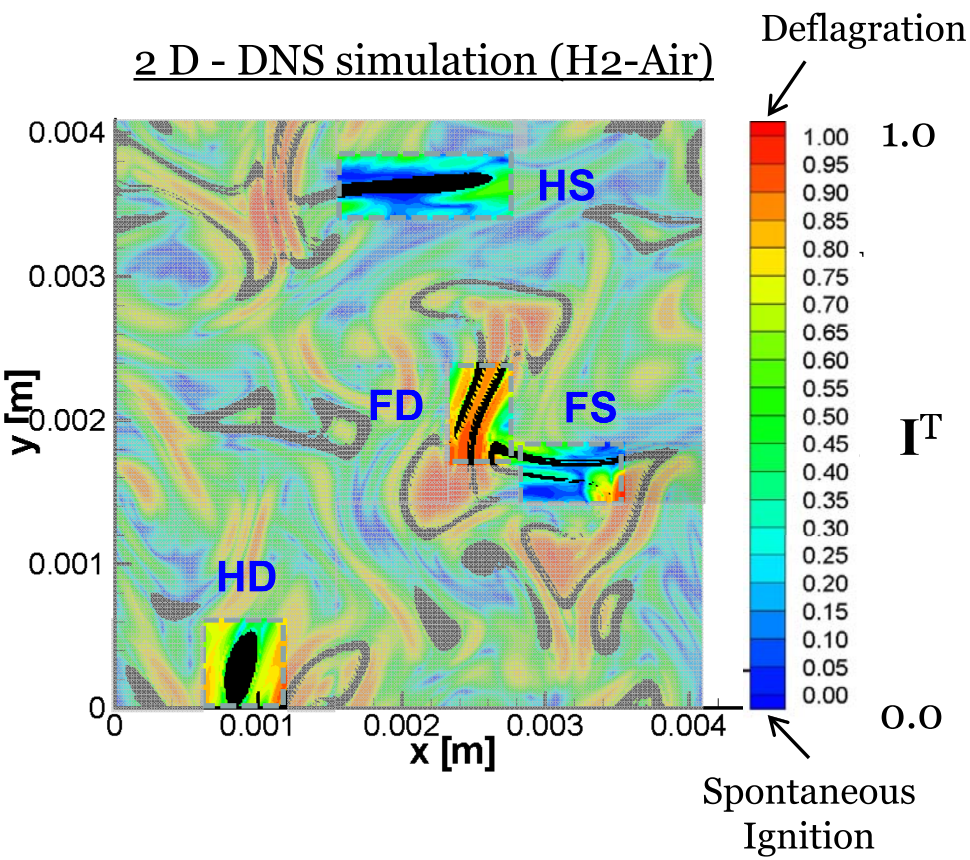
Results:

- New diagnostic tool identifies regions of interest in multi-mode combustion regimes

Future work:

- Application of method to HHC and syngas fuels
- Compositional stratification

2 D - DNS simulation (H2-Air)



M = 1 contour in black [denotes active reaction zone]

H: Homogeneous kernel, **F**: Front

S: Spontaneous Ignition, **D**: Deflagration

Summary

Project objectives: emphasizing the experimental and computational strengths at the UofM and beyond

1. to develop an accurate and rigorous experimental and computational data base of HHC reaction kinetics, flame speeds and flammability limits of HHC fuels including mixtures with high levels of exhaust gases

2. to develop detailed and reduced HHC chemical mechanisms that accurately reproduce the new experimental data as well as data in the literature, and

3. to develop a quantitative understanding of the stability of HHC combustion to fluctuations in the flow field, including the opportunities and challenges of exhaust gas recirculation (EGR) on extinction, ignition and flame stability,

4. to develop domain maps which identify the range of conditions (e.g. % EGR) where HHC combustion can be effective in both positive and negative manners (e.g. expanded/restricted flammability limits).

thank you