Investigation of Autoignition and Combustion Stability of High Pressure Supercritical Carbon Dioxide Oxy-combustion

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UTSR Project: DE-FE0025174   PM: Seth Lawson

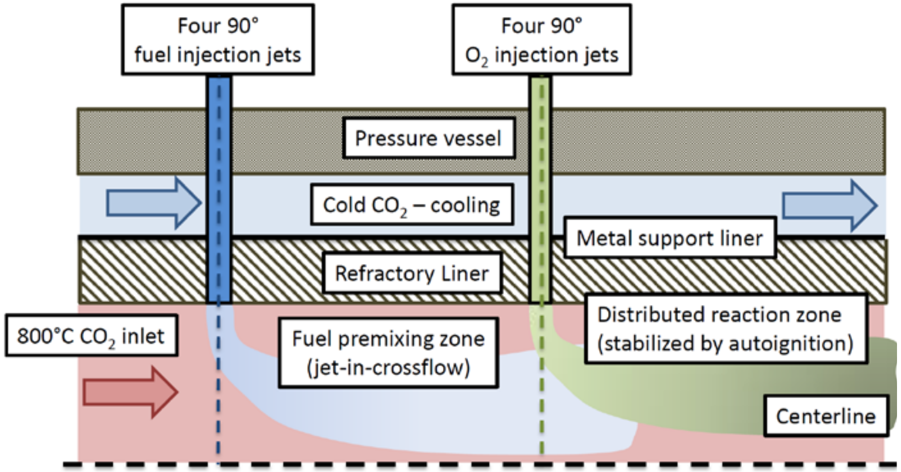
2017 UTSR Project Review Meeting

Overview of the Scientific Problem

- What fundamental combustion properties/knowledge we need in order to design combustor for SCO_2 oxy-combustion?
  - Kinetics and dynamics

Autoignition delays
and
flame dynamics of jet in crossflow

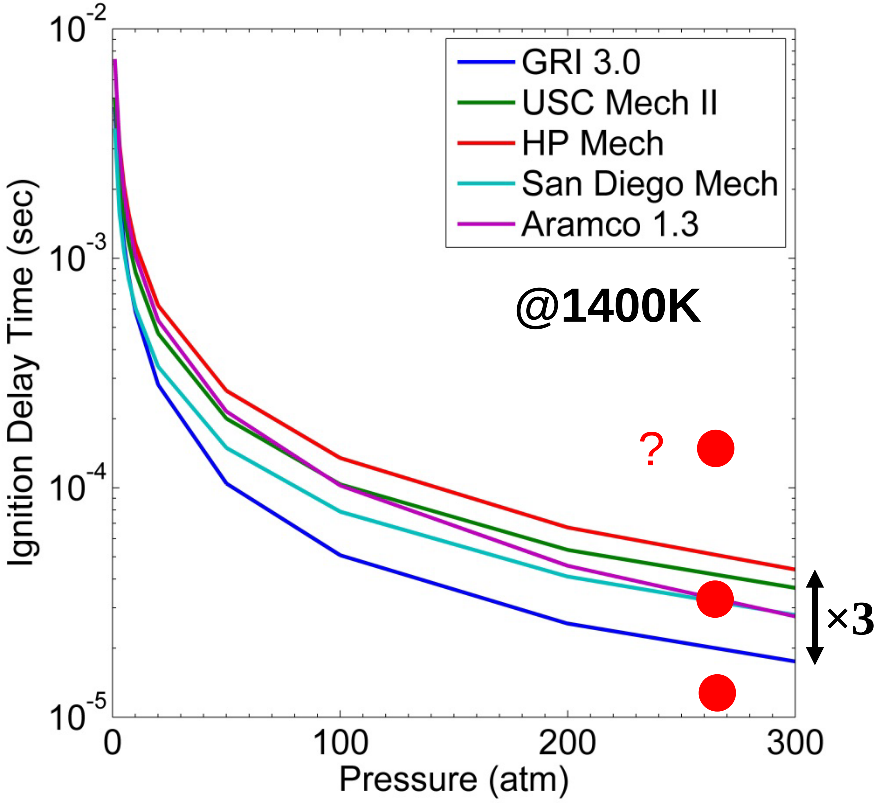
Conceptual combustor*



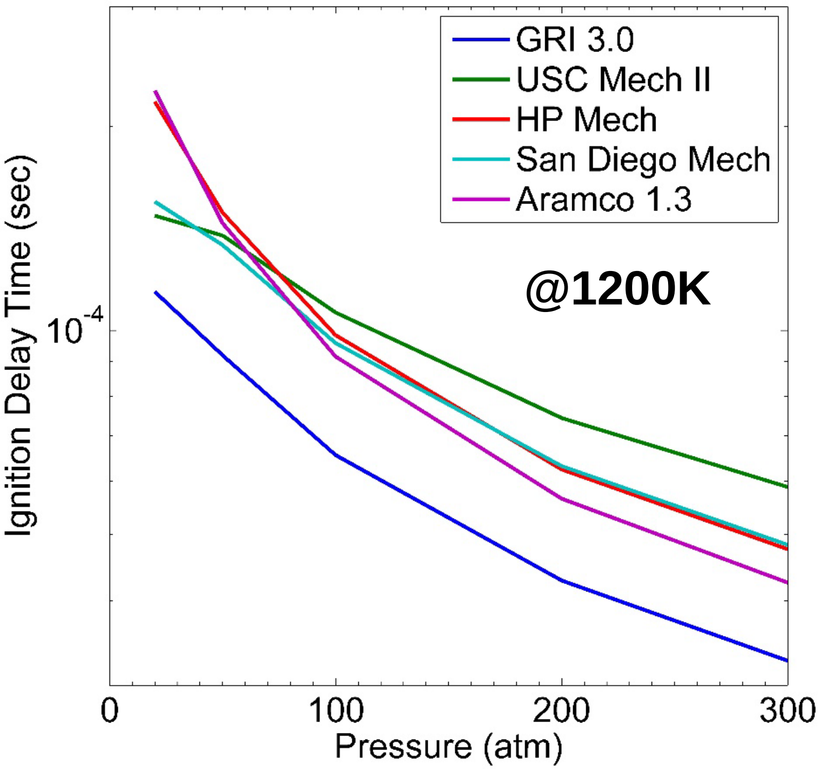
*J.Delimont, A. McClung, M. Portnoff, 2016 sCO2 symposium

Kinetic Challenges for SCO_2-fuel-O_2 Mixtures

Deviation increases with pressure: knowledge gap
Kinetic models must be validated at regime of interest



$CH_4/O_2/CO_2$ (9.5%:19%:71.48%)



$H_2/CO/O_2/CO_2$ (14.8%:14.8%:14.8%:55.6%)

Overview of the Scientific Questions and Proposed Work

- What is the fundamental combustion properties?
  - Experimental investigation of chemical kinetic mechanisms for SCO_2 Oxy-combustion (Task 1&2: Ranjan & Sun)

So what?

- How can we use the kinetic model to design combustors?
  - Development of a compact and optimized chemical kinetic mechanism for SCO_2 Oxy-combustion (Task 3: Sun)

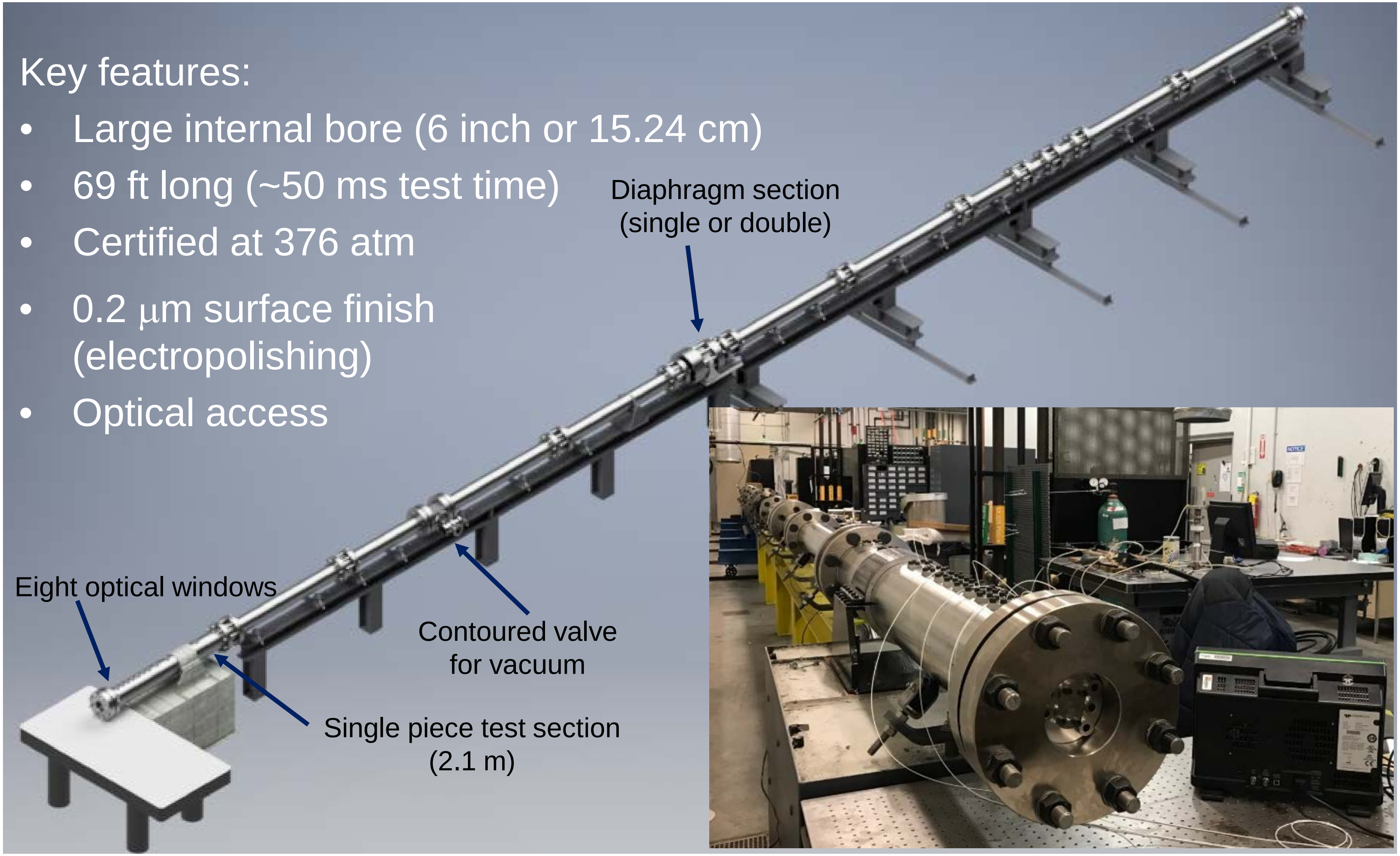
So what?

- What is the combustor dynamics at this new condition?
  - theoretical and numerical investigation of combustion instability for SCO_2 Oxy-combustion (Task 4&5: Lieuwen, Menon & Sun)

Task 1: Development of a High Pressure Shock Tube (complete)

Key features:

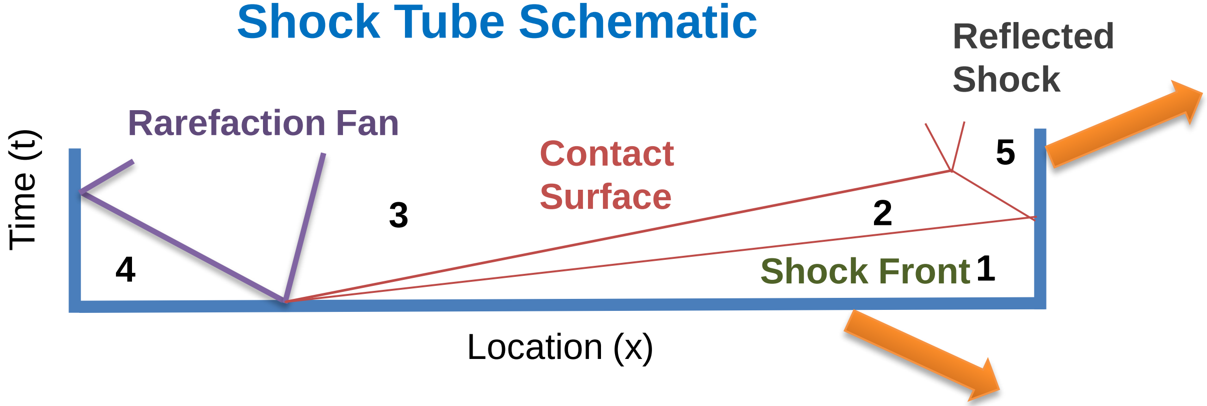
- Large internal bore (6 inch or 15.24 cm)
- 69 ft long (~50 ms test time)
- Certified at 376 atm
- 0.2 μm surface finish (electropolishing)
- Optical access



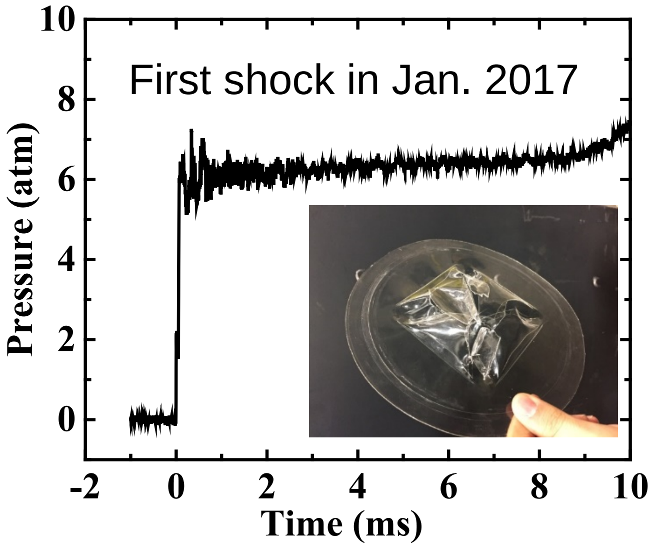
Task 1: Development of a High Pressure Shock Tube (complete)

- Mechanism of operation

Shock Tube Schematic



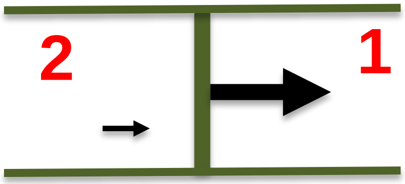
Measured P, calculated T



Lab-Frame Reflected Shock

T_5 = 1000 – 4000 K
$P_5 > P_2$

Lab-Frame Incident Shock



T_2 = 500 – 2000 K
$P_2 > P_1$

Study of High Pressure Autoignition - Facilities: mixture preparation

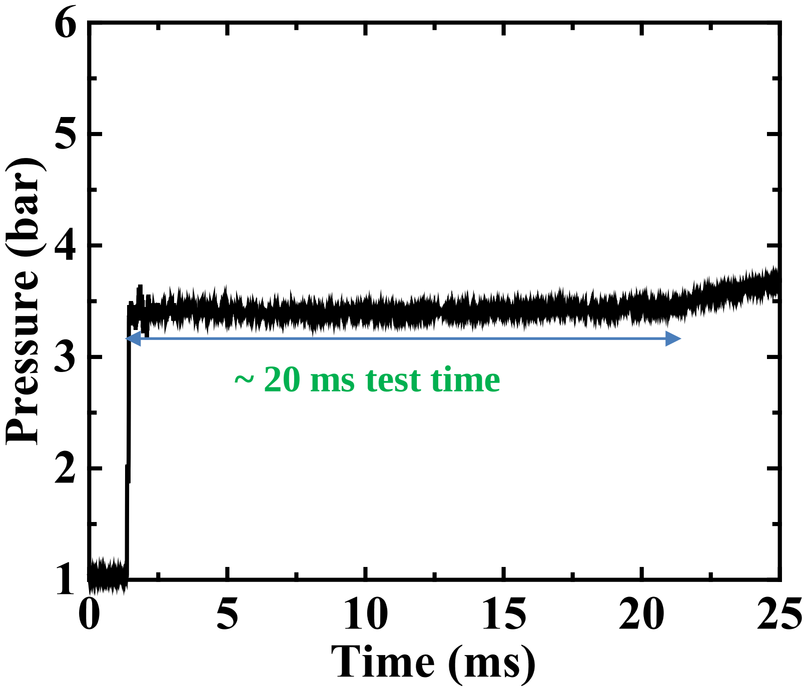
High accuracy Baratrons (**0.05%**) to measure partial pressure for mixture preparation

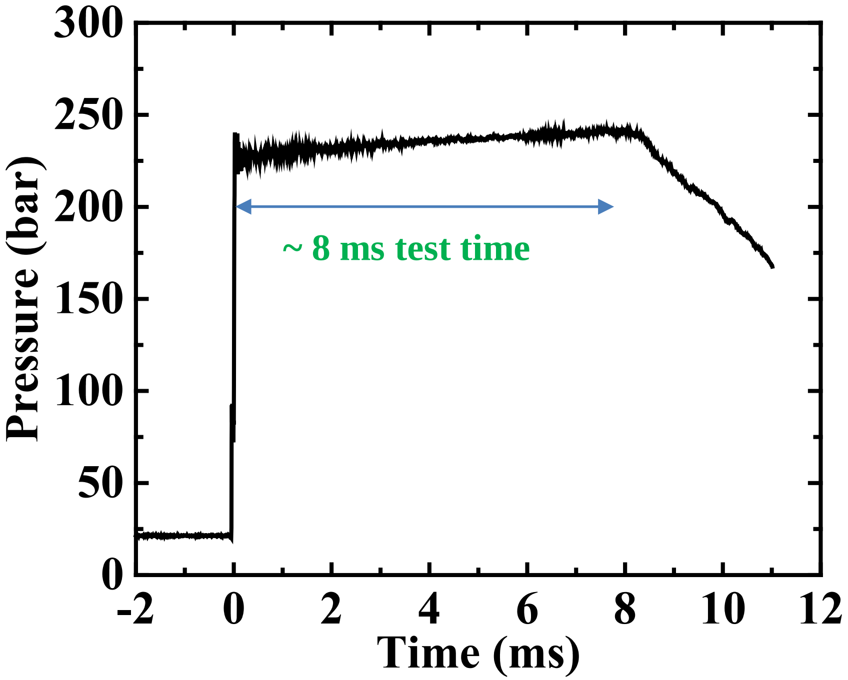
Turbo molecular pump

MicroGC to monitor compositions

Magnetic stir to promote mixing

Example of Pressure Traces



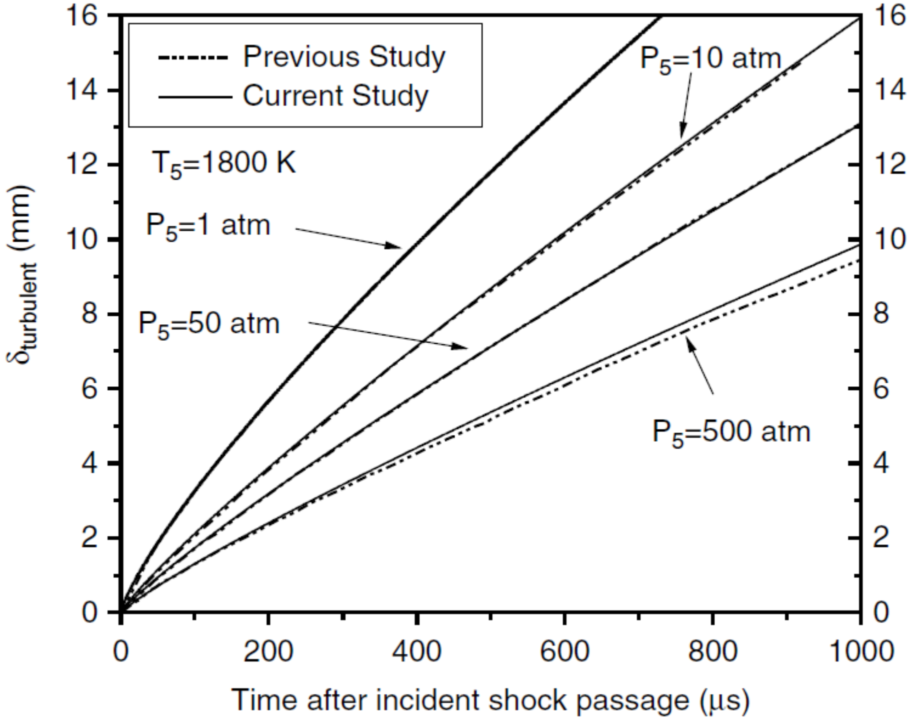


Unique features for high quality data

Challenges of High Pressure Shock Tube

- **Shock tube is not just a tube**
- Boundary layer
  - – from moving shock
- For polyatomic gases, BL is much thicker
- ID of shock tube must be large
  - – 150 mm

Argon as test gas

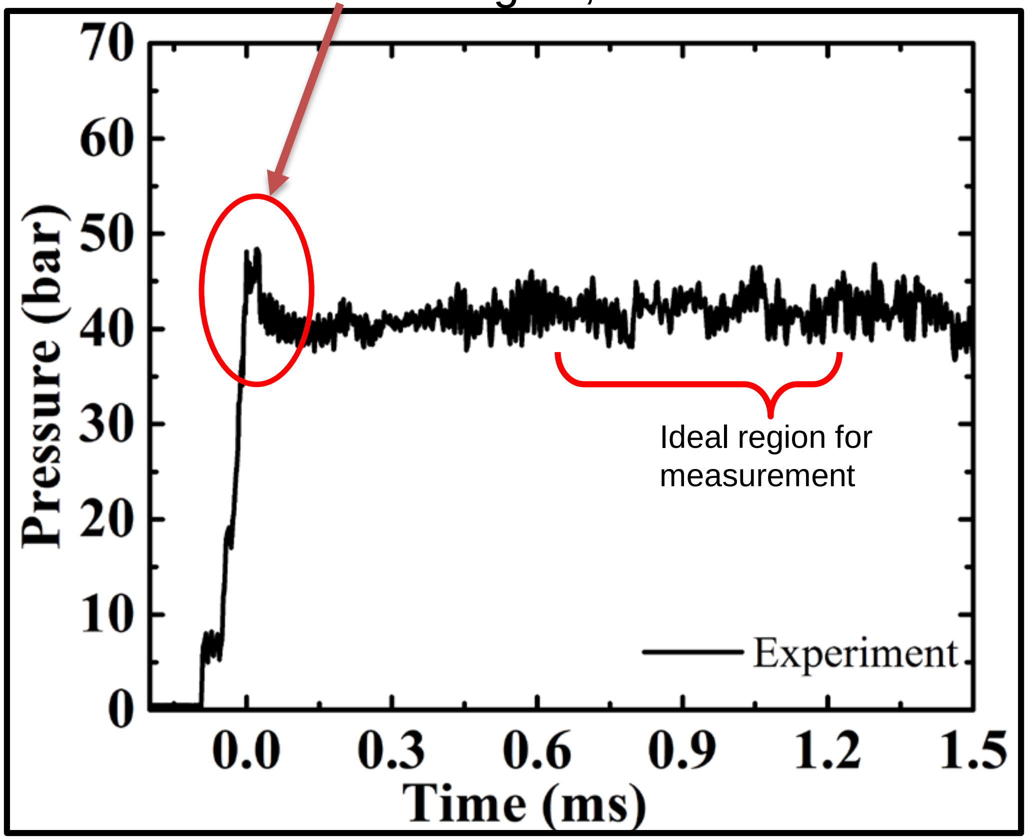


J. Hargis & E. Petersen, AIAA J, 2017

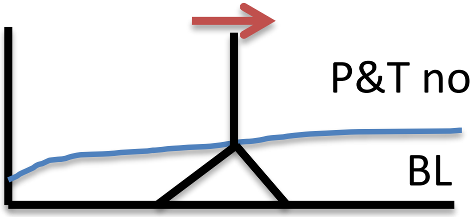
Challenges of High Pressure Shock Tube

- Test time needs to be long
  - Long enough to capture autoignition
  - Avoid bifurcation region
  - Longer tube, longer test time (21 m)

- A failed example
  - $CH_4/O_2/Ar/CO_2$=1:4:16:79
  - P=40 bar, T=1488 K
  - No autoignition captured


Shock bifurcation region, avoid

Ideal region for measurement


Reflected shock

P&T not constant

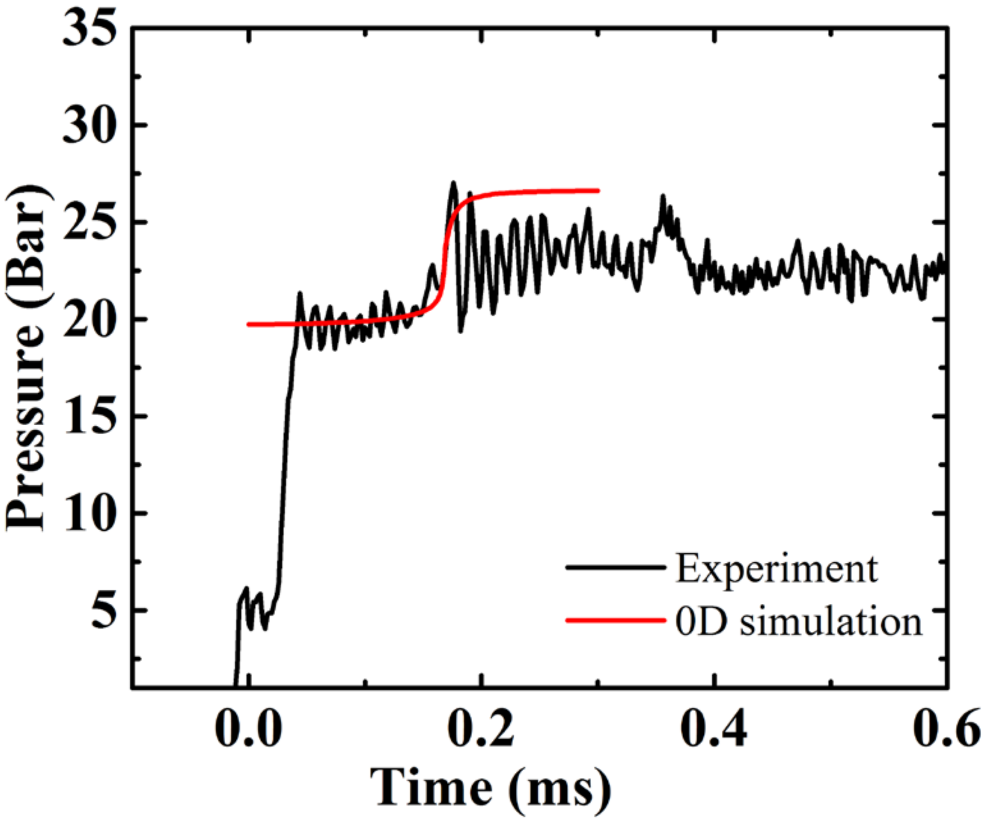
Facility Validation

- Low pressure autoignition measurement and validation
  - $P = 20$ atm, $T = 1641$ K
  - $CH_4/O_2/Ar=1:4:95$

- Agrees well with simulation using Aramco 2.0 (as expected)

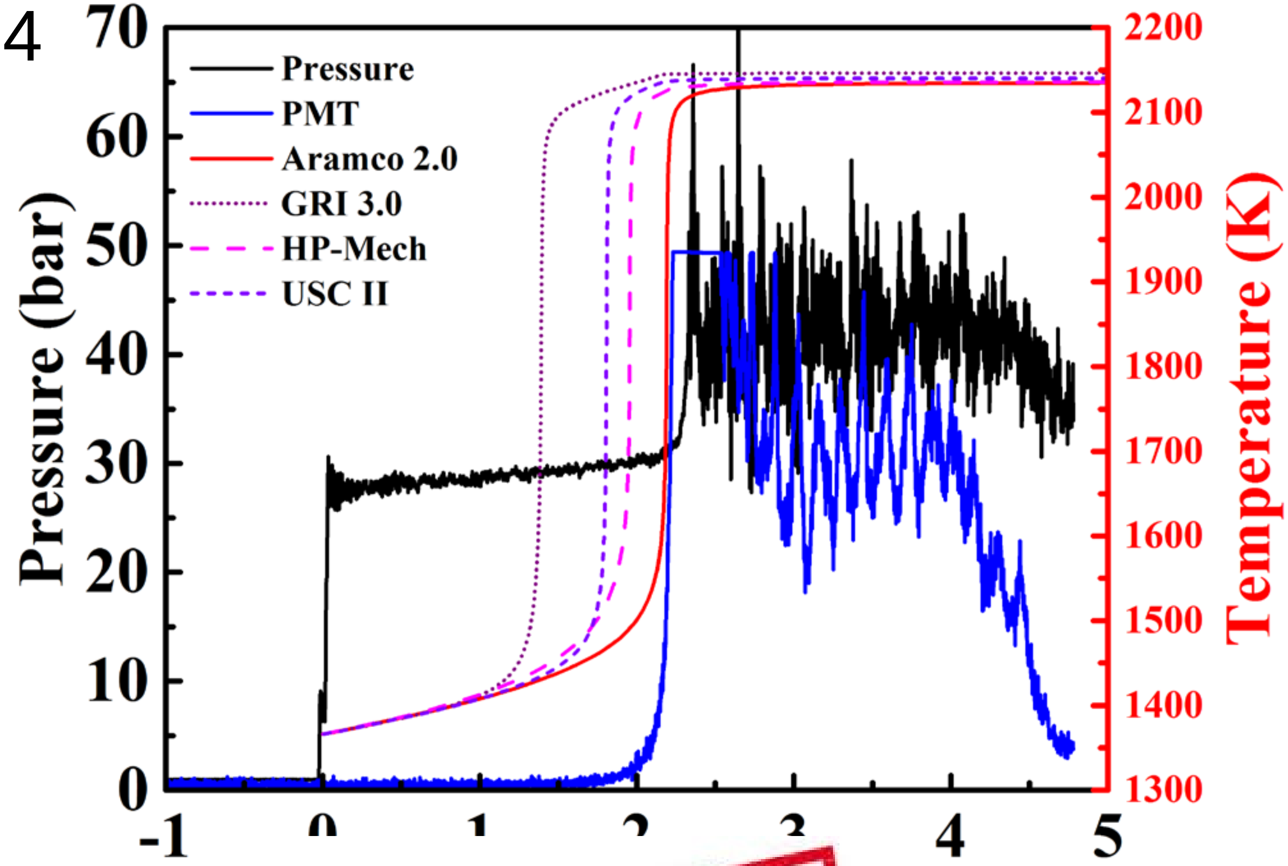
- Experiments vs. Stanford results
  - Agreed at similar conditions
  - e.g., $CH_4/O_2/Ar$ (2/4/96)
  - Stanford: 13.19 bar 1760 K $\tau_{ig}= 67\ \mu s$
  - GT: 16.5 bar 1737 K $\tau_{ig}= 57\ \mu s$

Good agreement between expt. and sim.



Facility Validation

- $CH_4/O_2/Ar=2:4:94$
- P=30 bar
- T= 1366 K
- Excellent between PMT signal (OH* emission) and simulation with Aramco 2.0



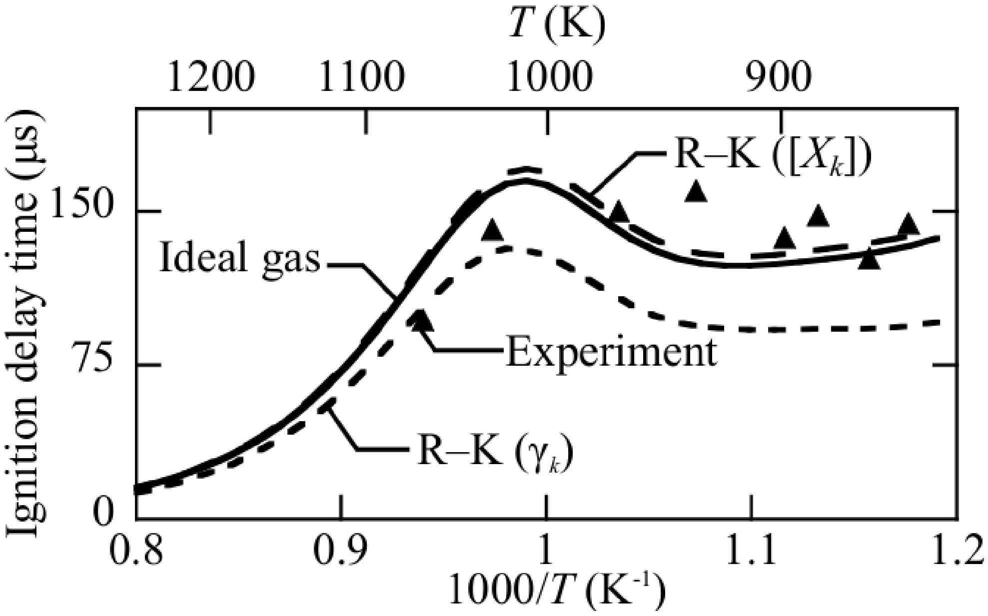
READY TO GO

Headaches from SCO_2 !?

Real Gas Effect in Shock Tube

- Negligible effect on thermodynamic properties (P, T) in region of interest
  - Small difference (<10 K) in high T (>1000 K) region
  - Kogekar et al., CNF 2017; Tang et al., IJCK 2006; Davidson et al., IJC 1996;

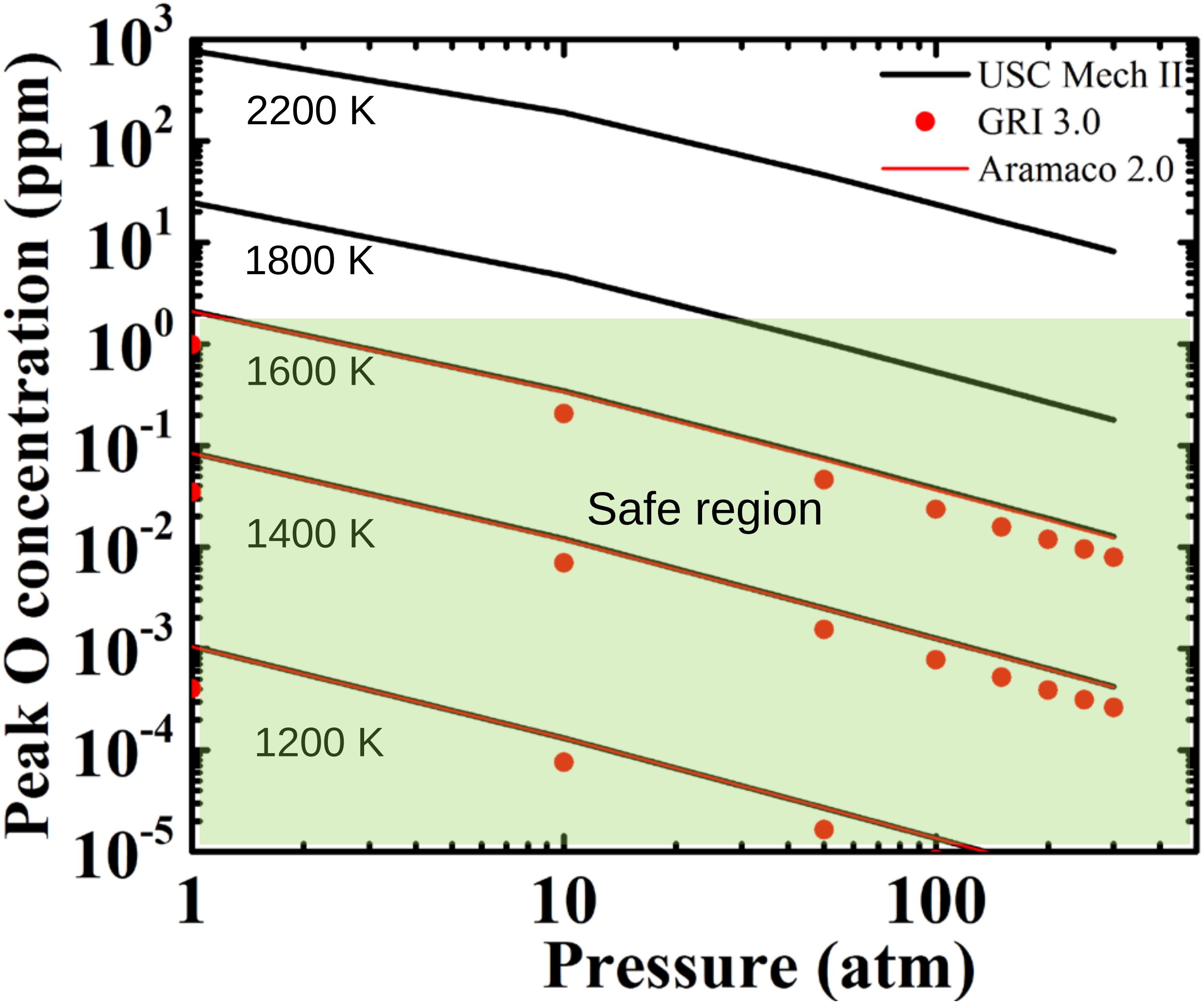
- It does NOT mean negligible effect on chemical reactions
  - Real gas non-unity activity coefficient (or fugacity) (negligible above 1100 K)
  - unknowns



n-dodecane/air at 80 atm

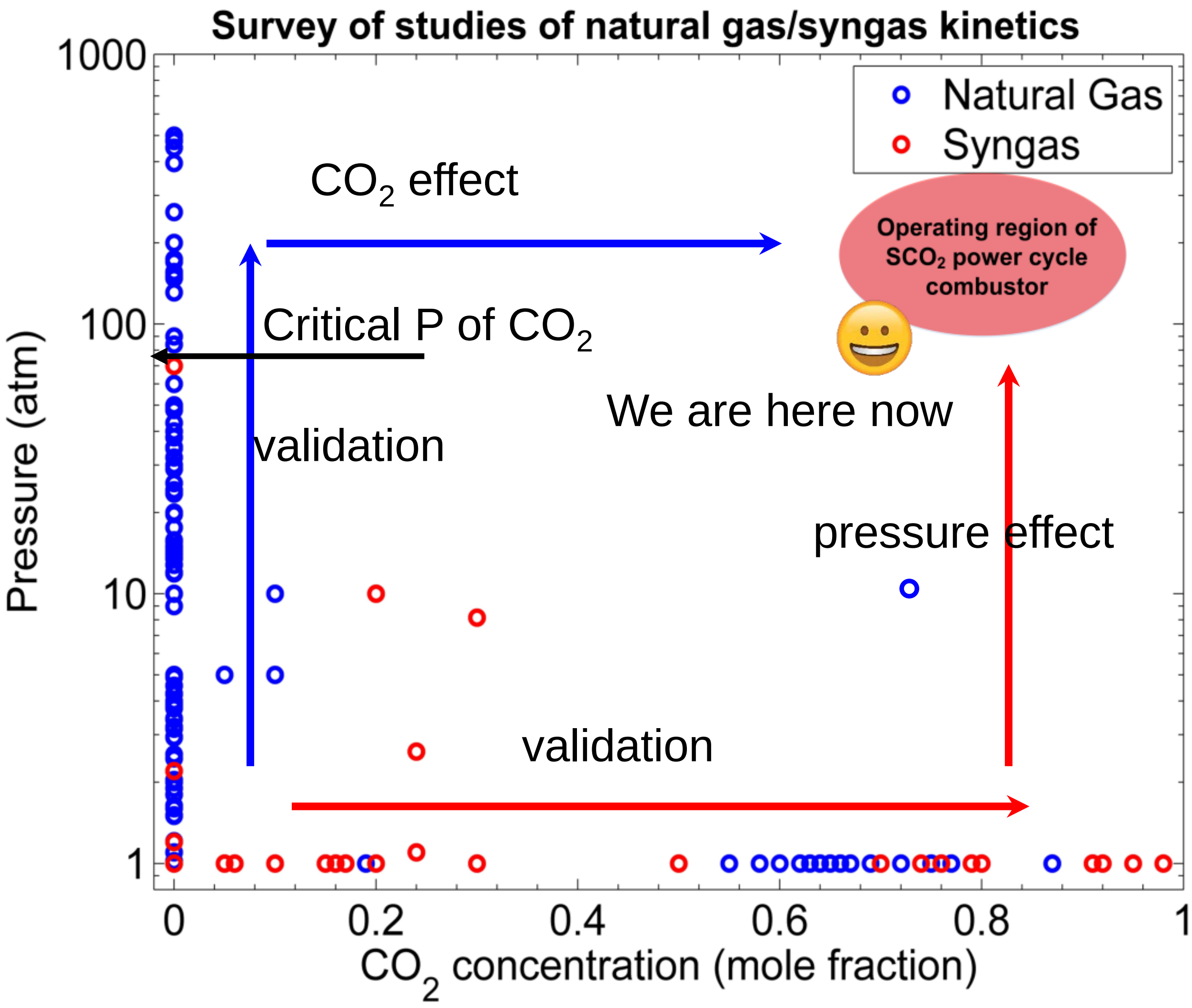
CO_2 Decomposition

- CO_2 decomposition
  - $CO_2 \rightarrow CO+O$
- Favored at high T, low P
- May affect autoignition measurement
  - Loose demarcation



Task 2: Investigation of Natural Gas and Syngas Autoignition in sCO_2 Environment

- No study before in region of interest
- A new regime to explore!
- CO_2 has negligible chemical effect
  - Based on 1 to 15 atm results and simulation using GRI 3.0 and Aramco 1.3
  - GT 17 atm expt. Agreed with Aramco 1.3 using same mixture with Hargis et al.



Survey of studies of natural gas/syngas kinetics

CO_2 effect

Operating region of SCO_2 power cycle combustor

Critical P of CO_2

We are here now

validation

pressure effect

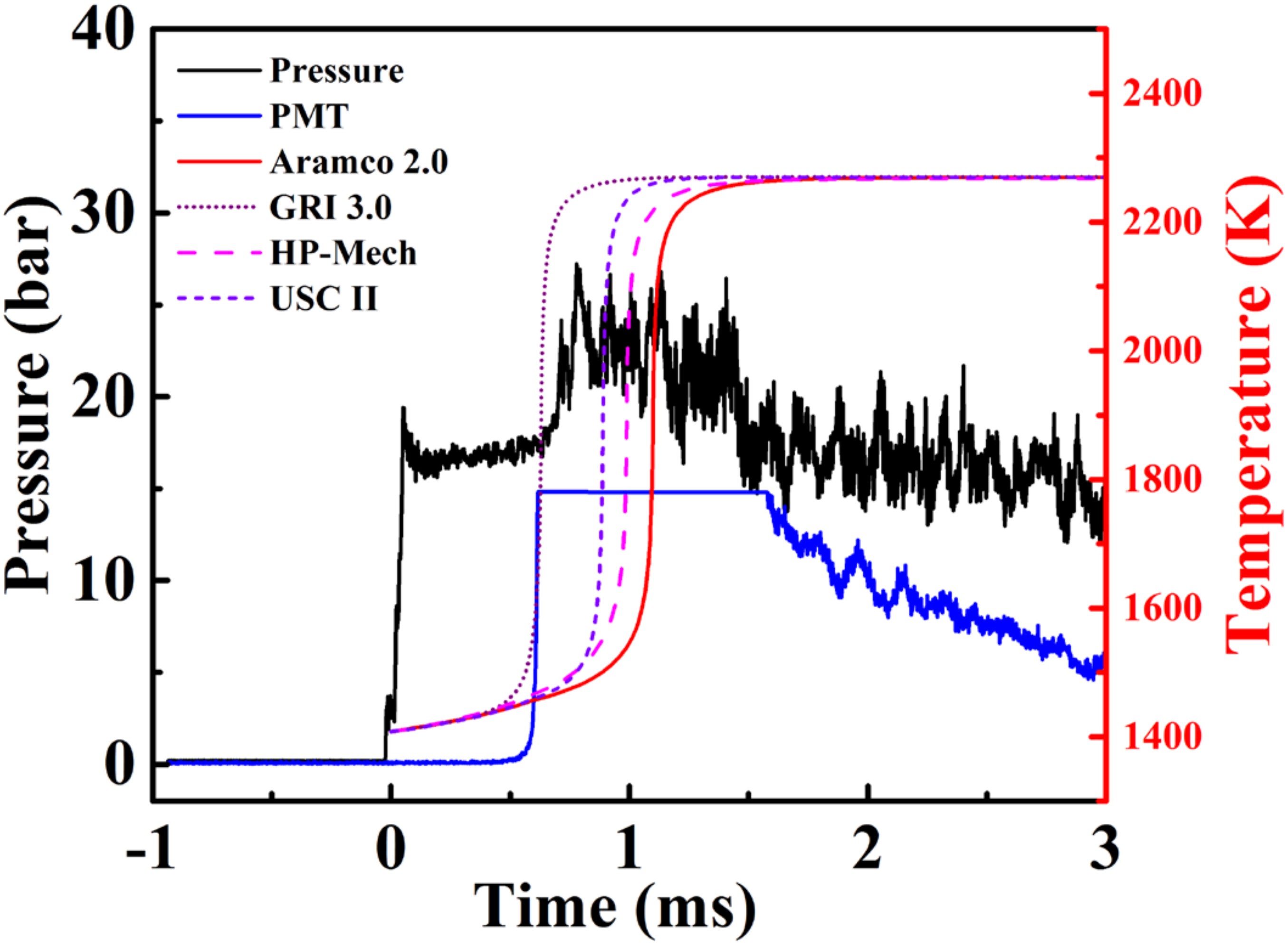
validation

e.g.:
J.W. Hargis, E.L. Petersen, Energy & Fuels, (29) 2015
S. Vasu, D.F. Davidson, R.K. Hanson, Energy & Fuels, (25) 2011

Too early to make conclusion

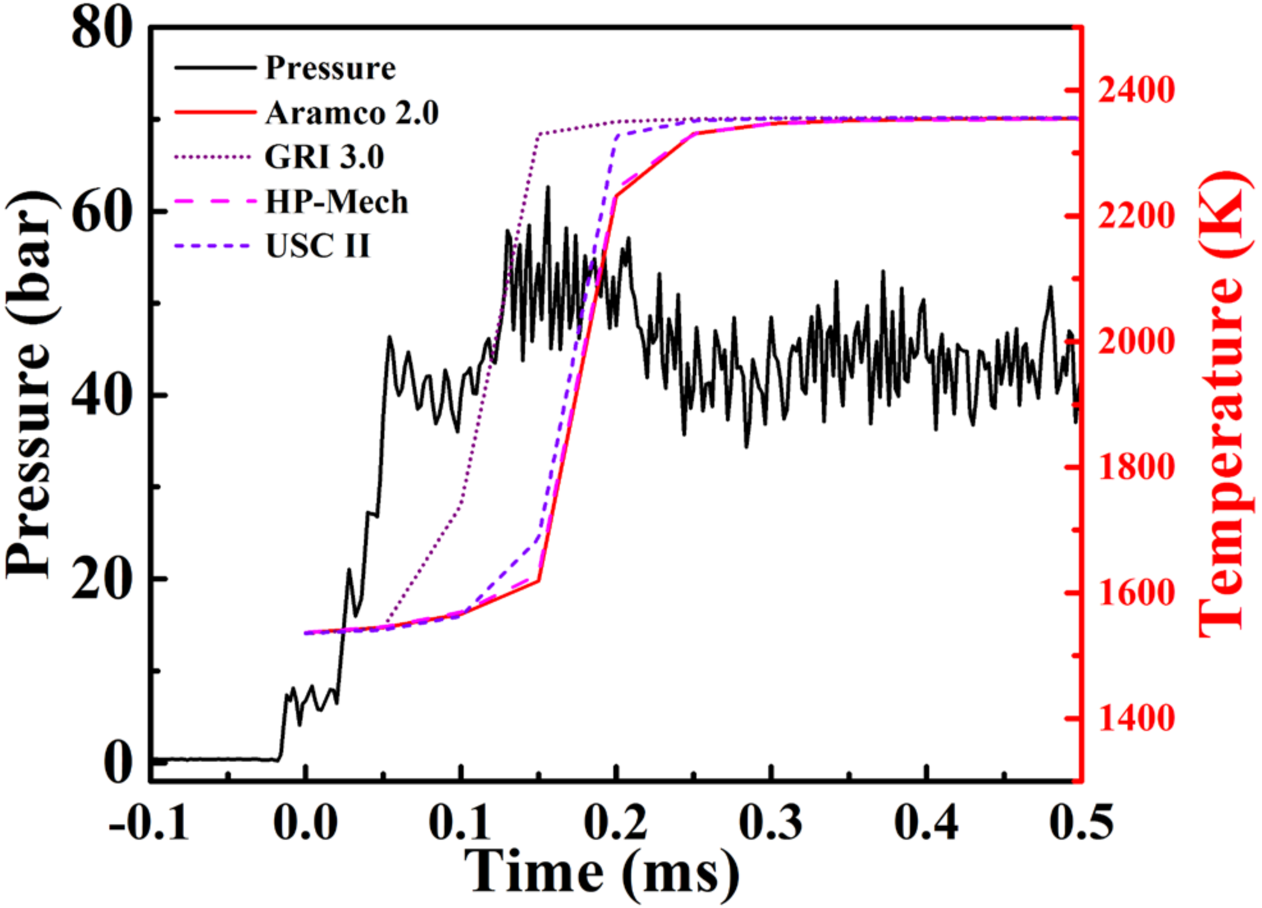
Autoignition with high CO_2 concentration: 15 bar

- $CH_4/O_2/Ar/CO_2$= 5:10:40:45
- P=15 bar
- T= 1409 K



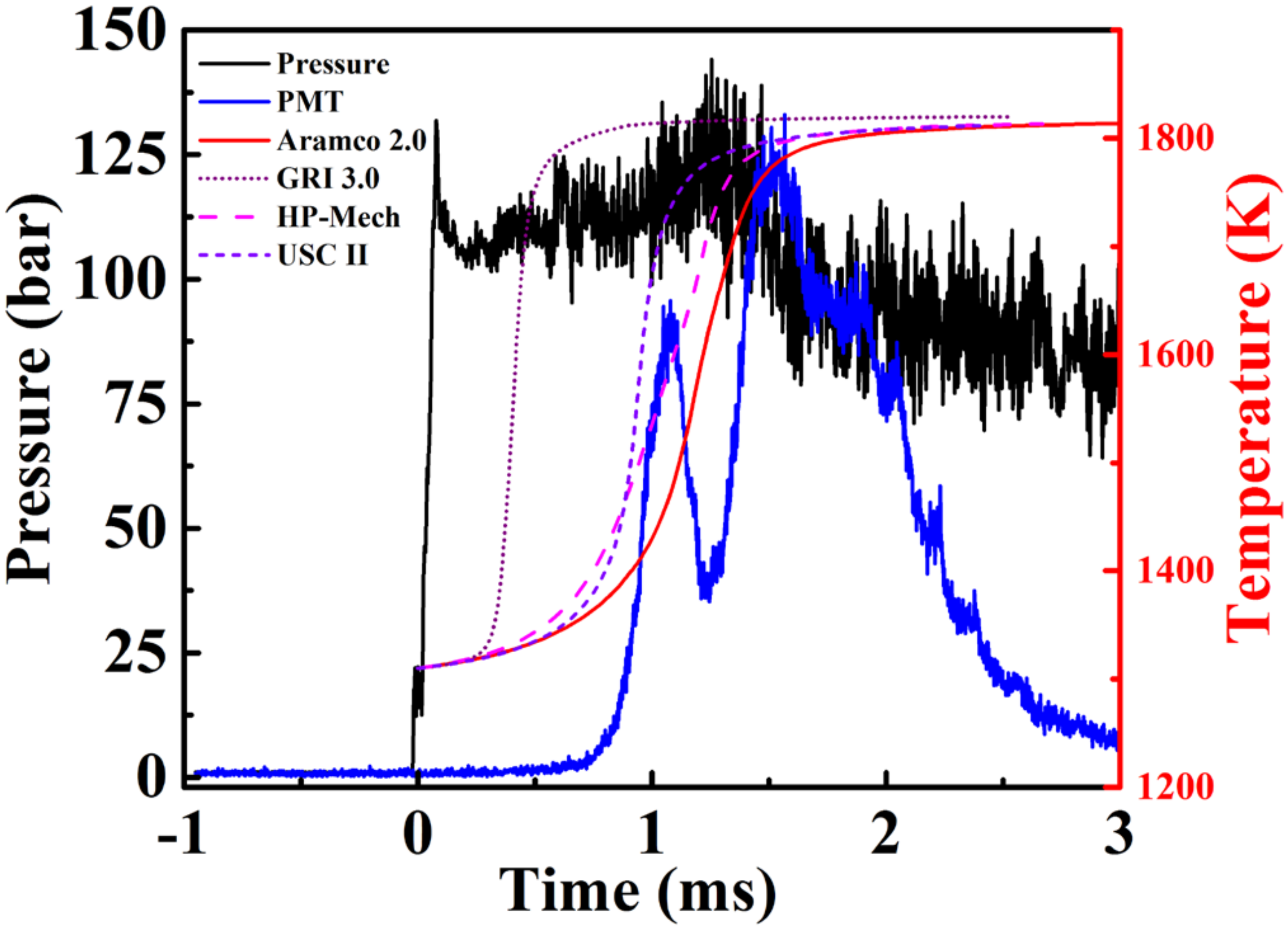
Autoignition with high CO_2 concentration: 41 bar

- $CH_4/O_2/Ar/CO_2$= 5:10:40:45
- P=41 bar
- T= 1535 K



Autoignition with high CO_2 concentration: 105 bar

- $CH_4/O_2/Ar/CO_2$= 3:6:24:67
- P=105 bar
- T= 1310 K

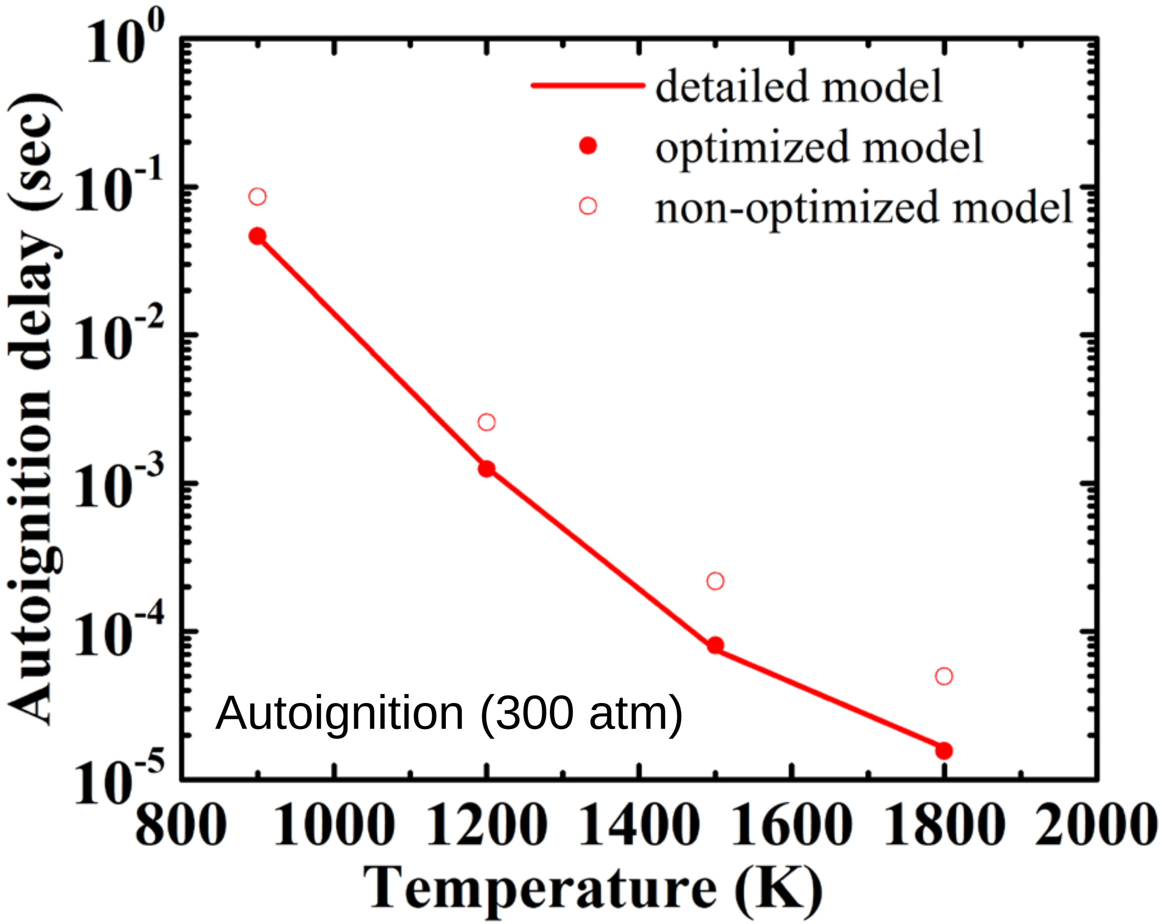


More data needed, simulation vs. expt. for comparison only
No conclusion, no recommendation yet

Task 3: Development of a Compact and Optimized Chemical Kinetic Model for SCO_2 Oxy-combustion

- USC Mech II (111 species) is used as a starting point for future optimized mechanism
- A 27 species reduced mechanism[1] for natural gas and syngas is developed (still too large for CFD)
- A new 13 species model was developed with optimization
  - Covers 900 K to 1800 K, 150 atm to 300 atm
  - Max 12% deviation

$CH_4/O_2/CO_2=0.031/0.062/0.907$ $\phi=1$



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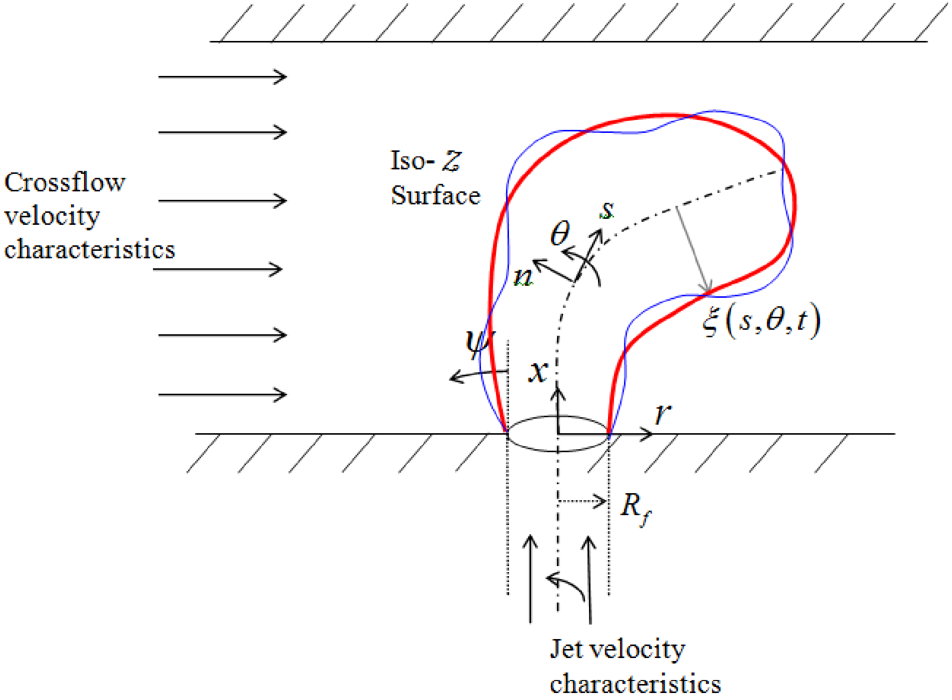
[1] S. Coogan, X. Gao, W. Sun, Evaluation of Kinetic Mechanisms for Direct Fired Supercritical Oxy-Combustion of Natural Gas, TurboExpo 2016

Task 4: Analytical modeling of Supercritical Reacting Jets in Crossflow

- Analytical framework for reacting jets in cross-flow
  - connect flow dynamics to flame dynamics
  - Modeling explicit flame position dynamics
  - Modeling spatially integrated heat release dynamics as a function of flame position

- Understanding flow dynamics of a jet in cross-flow
  - provide key inputs to the velocity field used in the analytical model



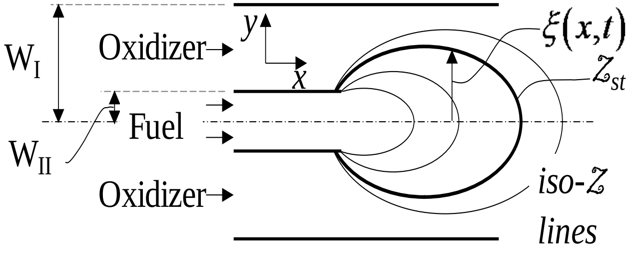
Analytic model of jet in crossflow

Position Dynamics PDE

$$\boxed{\frac{\partial \xi}{\partial t}+\left(u-u_D(x,\xi)\right)\frac{\partial \xi}{\partial x}-v=\mathscr{D}\frac{\partial^2 \xi}{\partial x^2}-s_D(x,\xi)\left[1-\left(\frac{\partial \xi}{\partial x}\right)^2\right]}$$

- Non-linear wrinkle convection
  - Flow based convection as well as position-coupled diffusion based convection
- Linear term from “Diffusion” of wrinkles
  - Similar to stretch effects in premixed flames (i.e. stretch correction to flame speed)
- Non-linear propagation-like term from diffusion
- Decompose all quantities into a steady time-average and time-dependent perturbation

$$\boxed{\begin{array}{ll} \xi=\xi_0(x)+\xi_1(x,t) & \\ u=u_0(x)+u_1(x,t) & v=v_0(x)+v_1(x,t) \\ u_D=u_{D,0}+u_{D,1} & s_D=s_{D,0}+s_{D,1} \end{array}}$$



Flame Position Dynamics

$$\frac{\partial \xi_1(x,t)}{\partial t} + \left(u_0(x) - u_{D,0}(x) - 2s_{D,0}(x)\right)\frac{\partial \xi_1(x,t)}{\partial x} - \mathscr{D}\frac{\partial^2 \xi_1}{\partial x^2}$$

$$= \left(v_1(x,t) - u_1(x,t)\frac{d\xi_0(x)}{dx}\right) + \xi_1 \underbrace{\left(\frac{\partial u_{D,0}}{\partial \xi_0}\frac{d\xi_0(x)}{dx} - \frac{\partial s_{D,0}}{\partial \xi_0}\left[1-\left(\frac{d\xi_0(x)}{dx}\right)^2\right]\right)}_{\text{Reaction Coefficient}}$$

- Governing Physics
  - Wrinkle convection
  - Diffusion, similar to premixed flame stretch
  - Reactive type dynamics
- High Pe limit
  - Diffusion time-scale large compared to convection time-scale
  - Diffusion based convection – $1/Pe^2$
  - Diffusion based propagation – $1/Pe$

Global Flame Dynamics

- For acoustically compact flames, spatially integrated heat release is the dynamics relevant quantity

$$\dot{Q}(t) = \int_{flame} \dot{m}''_F \, h_R \, dA = \int_{flame} \rho_u \mathscr{D} \left|\nabla Z\right|_{Z_{st}} h_R \sqrt{1+\left(\frac{\partial \xi}{\partial x}\right)^2}\, dx$$

$$= \int_{flame} \dot{m}''_{F,0} \, h_R \, dA_0 + \int_{flame} \dot{m}''_{F,0} \, h_R \, dA_1 + \int_{flame} \dot{m}''_{F,1} \, h_R \, dA_0$$

- Time-average heat release

$$\dot{m}''_{F,0} = \rho_u \mathscr{D} \frac{1}{\sqrt{1+\left(\xi_{0,x}\right)^2}} \left.\frac{\partial Z_0}{\partial y}\right|_{Z_{st}} \qquad dA_0 = \sqrt{1+\left(\xi_{0,x}\right)^2}$$

- Weighted Area Dynamics
  - Note that for premixed flame with constant flame speed, this weighting was constant = flame speed

$$dA_1 = \frac{\xi_{0,x}}{\sqrt{1+\left(\xi_{0,x}\right)^2}} \xi_{1,x}$$

- Mass burning rate dynamics

$$\dot{m}''_{F,1} = -\rho_u \mathscr{D} \frac{1}{\sqrt{1+\left(\xi_{0,x}\right)^2}} \left[\left(\frac{1+\left(\xi_{0,x}\right)^2}{\xi_{0,x}}\right) \left(\frac{\partial Z_0}{\partial y}\right)_{Z_{st}} \xi_{1,x} + \left.\frac{\partial^2 Z_0}{\partial y^2}\right|_{y=\xi_0(x)} \xi_1 \right]$$

Experiment Data Processing

- Vortex Tracking
- Extract Phase roll-off from experimental data
  - Further data reduction and smoothing required to get meaningful information
- Physical parameters
  - Convection speed
  - Differences in leeward and windward side

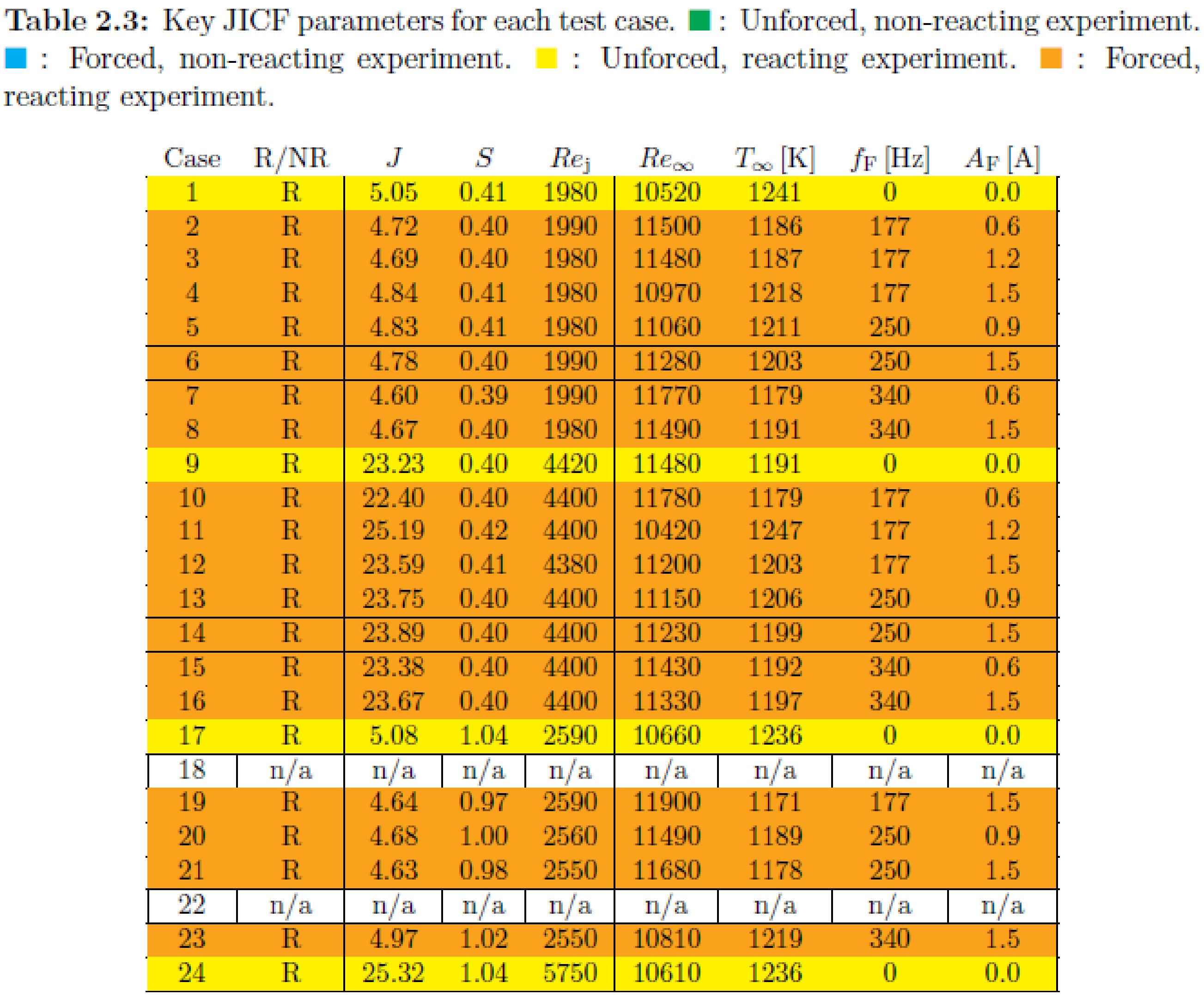
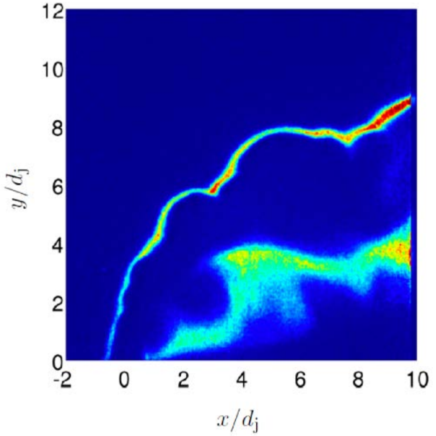


Table 2.3: Key JICF parameters for each test case. 🟩 : Unforced, non-reacting experiment. 🟦 : Forced, non-reacting experiment. 🟨 : Unforced, reacting experiment. 🟧 : Forced, reacting experiment.

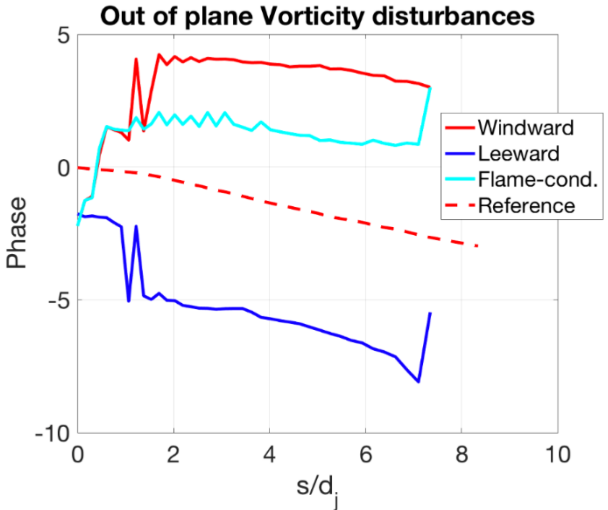
Case	R/NR	J	S	Re_j	Re_∞	T_∞ [K]	f_F [Hz]	A_F [A]
1	R	5.05	0.41	1980	10520	1241	0	0.0
2	R	4.72	0.40	1990	11500	1186	177	0.6
3	R	4.69	0.40	1980	11480	1187	177	1.2
4	R	4.84	0.41	1980	10970	1218	177	1.5
5	R	4.83	0.41	1980	11060	1211	250	0.9
6	R	4.78	0.40	1990	11280	1203	250	1.5
7	R	4.60	0.39	1990	11770	1179	340	0.6
8	R	4.67	0.40	1980	11490	1191	340	1.5
9	R	23.23	0.40	4420	11480	1191	0	0.0
10	R	22.40	0.40	4400	11780	1179	177	0.6
11	R	25.19	0.42	4400	10420	1247	177	1.2
12	R	23.59	0.41	4380	11200	1203	177	1.5
13	R	23.75	0.40	4400	11150	1206	250	0.9
14	R	23.89	0.40	4400	11230	1199	250	1.5
15	R	23.38	0.40	4400	11430	1192	340	0.6
16	R	23.67	0.40	4400	11330	1197	340	1.5
17	R	5.08	1.04	2590	10660	1236	0	0.0
18	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
19	R	4.64	0.97	2590	11900	1171	177	1.5
20	R	4.68	1.00	2560	11490	1189	250	0.9
21	R	4.63	0.98	2550	11680	1178	250	1.5
22	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
23	R	4.97	1.02	2550	10810	1219	340	1.5
24	R	25.32	1.04	5750	10610	1236	0	0.0

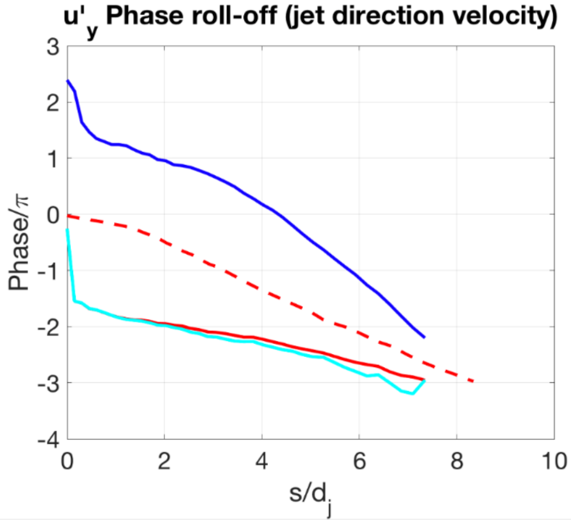
Modeling Velocity Disturbances

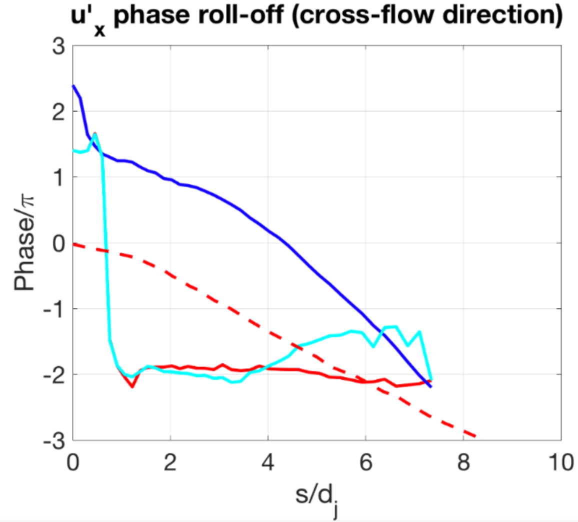
- Using experiment database
- Data Sampling
  - w.r.t. jet centerline co-ordinate
  - at windward and leeward vortex centerlines
  - conditioned to flame location
    - Leeward flame was too diffuse
- Spatial variation of phase roll-off from Fourier modes



$$u'(x,t) = \mathrm{Re}\left[\left\{\hat{A}(x)\exp\left(\frac{-i\omega x}{c_0}\right) + \hat{B}(x)\exp\left(\frac{-i\omega x}{u_0} + i\varphi(x)\right)\right\}\exp(-i\omega t)\right]$$


Out of plane Vorticity disturbances

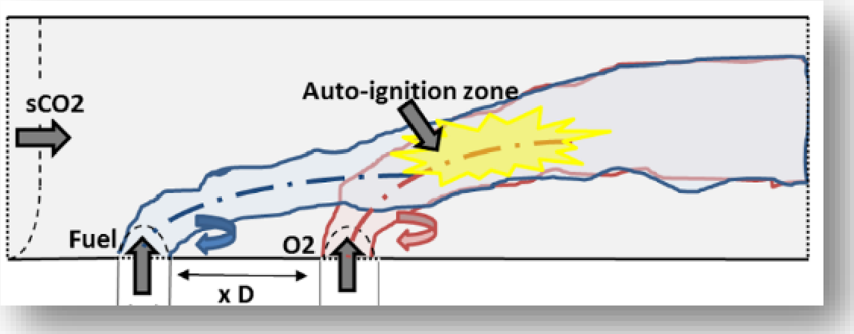

u'$_y$ Phase roll-off (jet direction velocity)


u'$_x$ phase roll-off (cross-flow direction)

Key Takeaways from Task

- PDEs for steady state and fluctuating flame position
  - Reduce the need for a full-field mixture fraction solution
- Global dynamics through spatially integrated heat release expressed in terms of flame position dynamics
  - Simplified expression for combustion dynamics modeling
- Identification of control parameter
  - From previously measured JICF data
  - Vortex tracking
  - Phase roll-off convection speed
  - Differences in speed between windward and leeward side

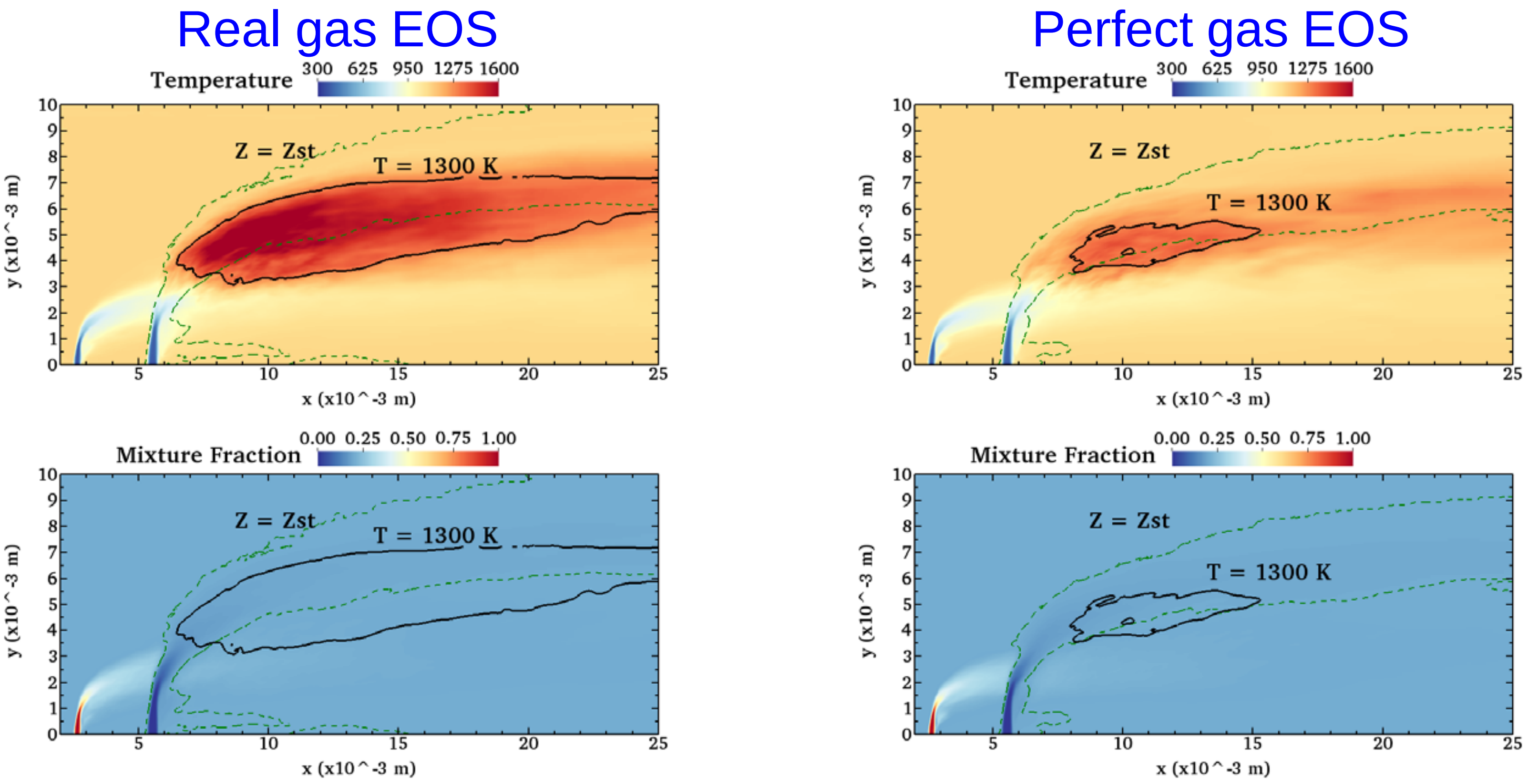
Task 5: LES Studies of Supercritical Mixing and Combustion



Baseline model
NOT actual design

- Mixing and flame stability
- Systematic variation of design parameters
  - Momentum ratios for fuel and oxygen, flow rate, number of sets
  - Size, spacing, and locations of injectors
- Computational modeling may be more cost effective but include its own challenges
  - Autoignition kinetics (large uncertainty, maybe wrong)
  - Turbulence-chemistry closure
  - Real gas effects

Recap of Last Year: Real Gas Effect



Real gas EOS

Perfect gas EOS

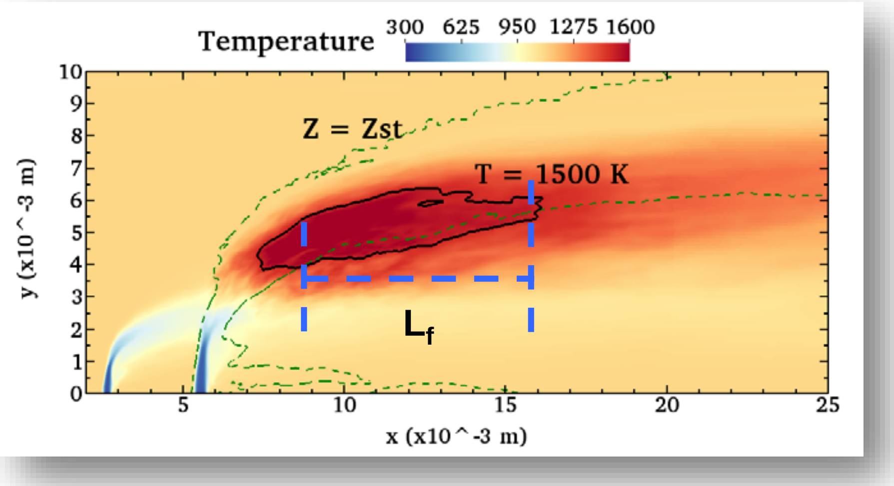
- Global (highly simplified) kinetic model
- Reduced jet penetration with perfect gas EOS in comparison to Peng Robinson EoS – clearly shows RG effects
- Heat release also decreased with perfect gas EOS

Recap of Last Year:
Flame Length and Combustion Efficiency

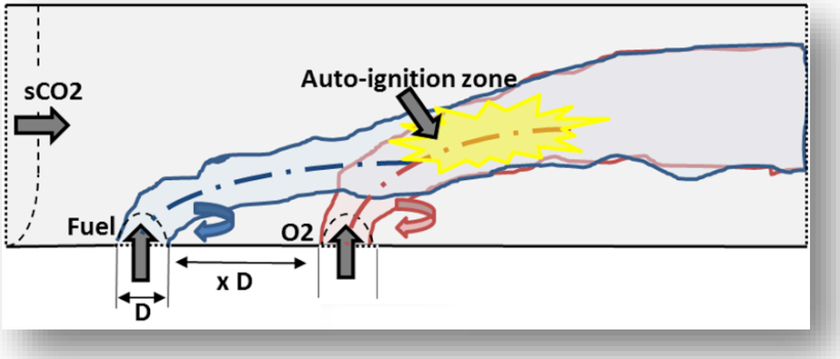
- **Combustion is not efficient**
- Combustion efficiency estimated as:

$$\eta = 100 \times \frac{\dot{m}_{f,in} - \dot{m}_{f,out}}{\dot{m}_{f,in}} \sim 49\%$$

- Flame length, $L_f \sim 14.5\ D_{ox}$
  - estimated as intersection of $Z = Z_{st}$ and T = 1500 K
- η needs to be improved
  - Inflow realistic turbulence
  - Modify J and jet spacing
  - Mass flow rate changes
  - Jet-staging and distributed mixing
  - **Inflow swirling**
- Mixing is the key

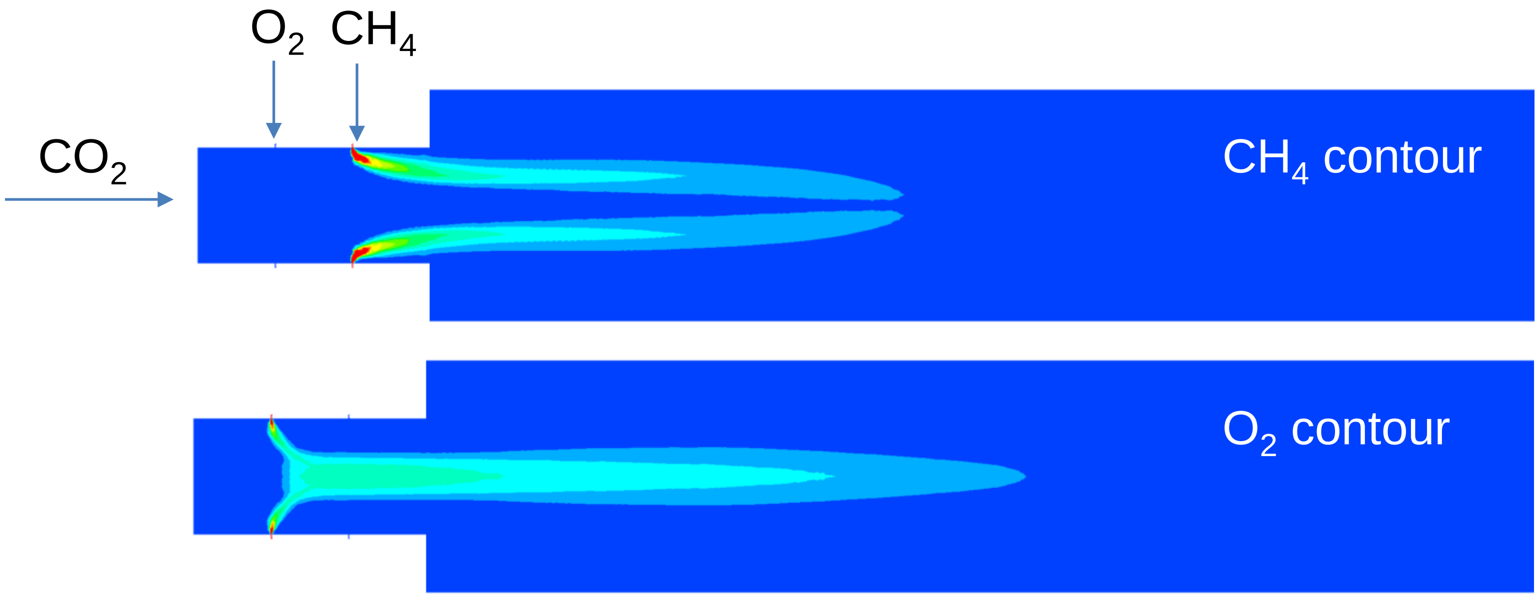


Temperature overlaid with stoichiometry line



FLUENT Simulation

- Fluent simulation with circumferential injections
- Mixing is challenging

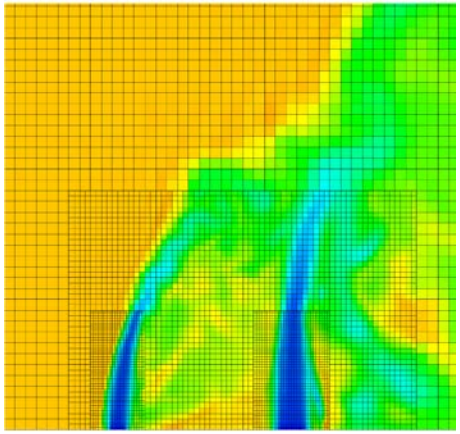


Summary of Progress for Numerical Investigation

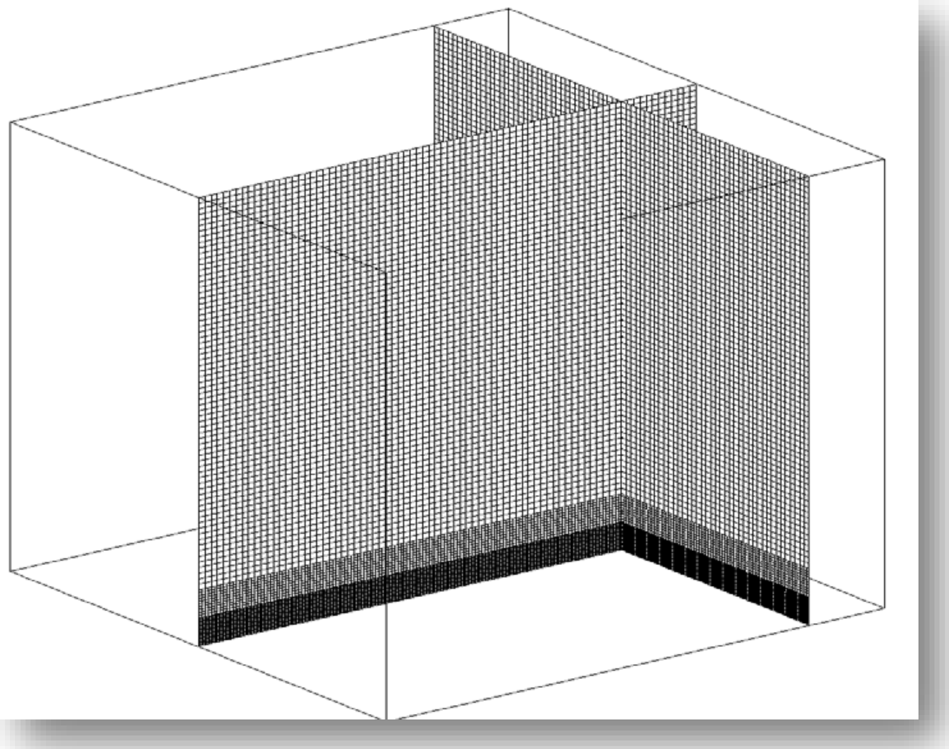
- Focus on jet mixing, LES of non-reacting mixing to identify where stoichiometric surface appear, then identify autoignition regions
  - Case 1: fuel jet behind O_2 jet by 28 mm
  - Case 2: O_2 behind fuel jet by 28 mm; Case 3: 14 mm
- LES using compressible adaptive-mesh-refinement (AMR)
  - Reduced finite-rate kinetics (from Task 3) used
  - Implemented in a PSR based network model
- Studies of reacting spatial mixing layer (SML) configuration
  - Canonical problem with some known features
  - CH_4-O_2 mixing and reactions in CO_2 background
  - Study effect of pressure, details of the kinetics

LES using AMR: Mixing in JICF

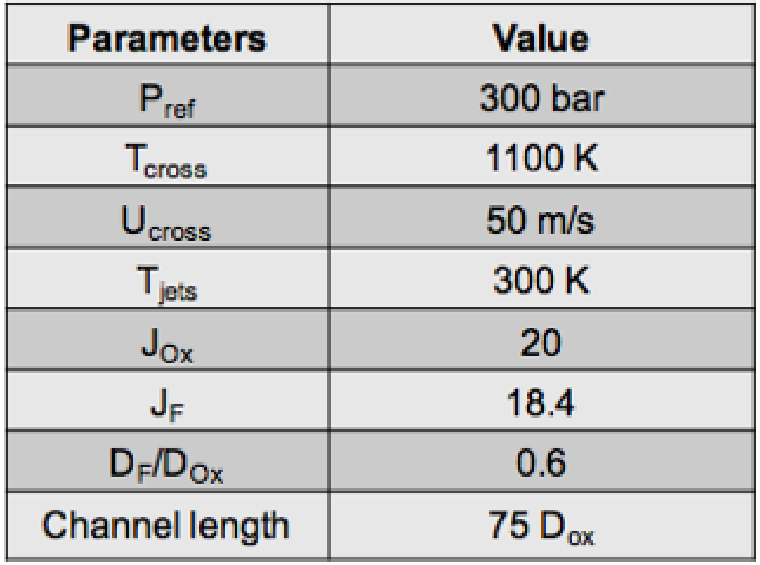
- AMR refines grid near the jet inlets.
- SGS closure accounts for AMR[1]



Temperature (K)



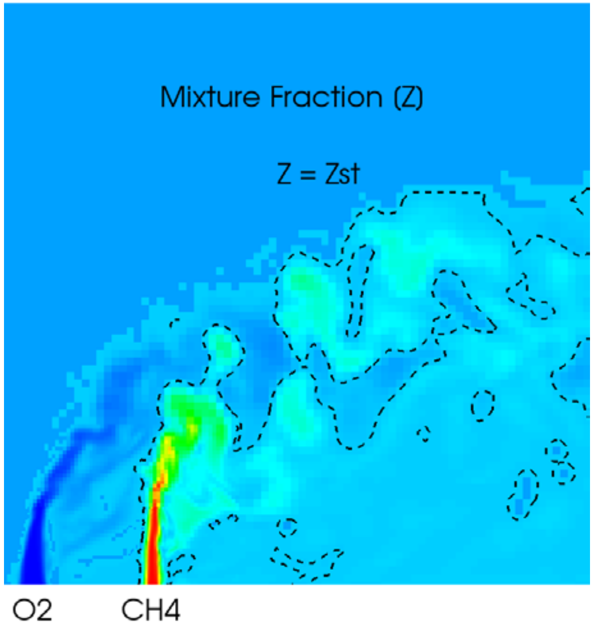
Dynamic AMR[1]



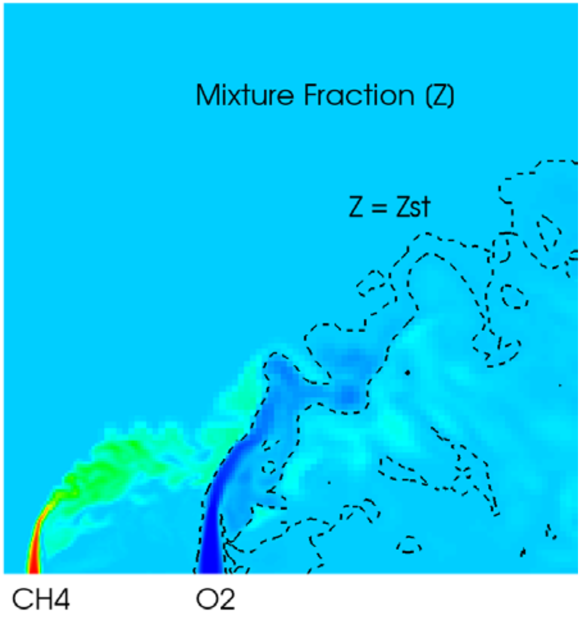
Parameters	Value
P_{ref}	300 bar
T_{cross}	1100 K
U_{cross}	50 m/s
T_{jets}	300 K
J_{Ox}	20
J_F	18.4
D_F/D_{Ox}	0.6
Channel length	75 D_{ox}

[1] Muralidharan & Menon, AIAA-0825-2014

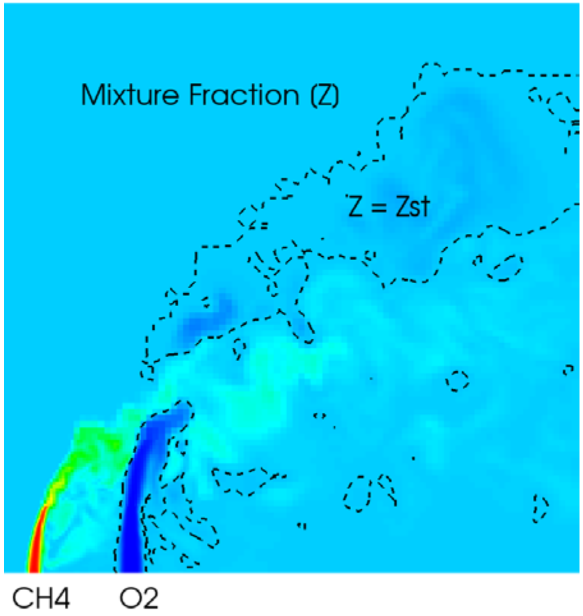
Mixing Studies Using LES



Case 1



Case 2

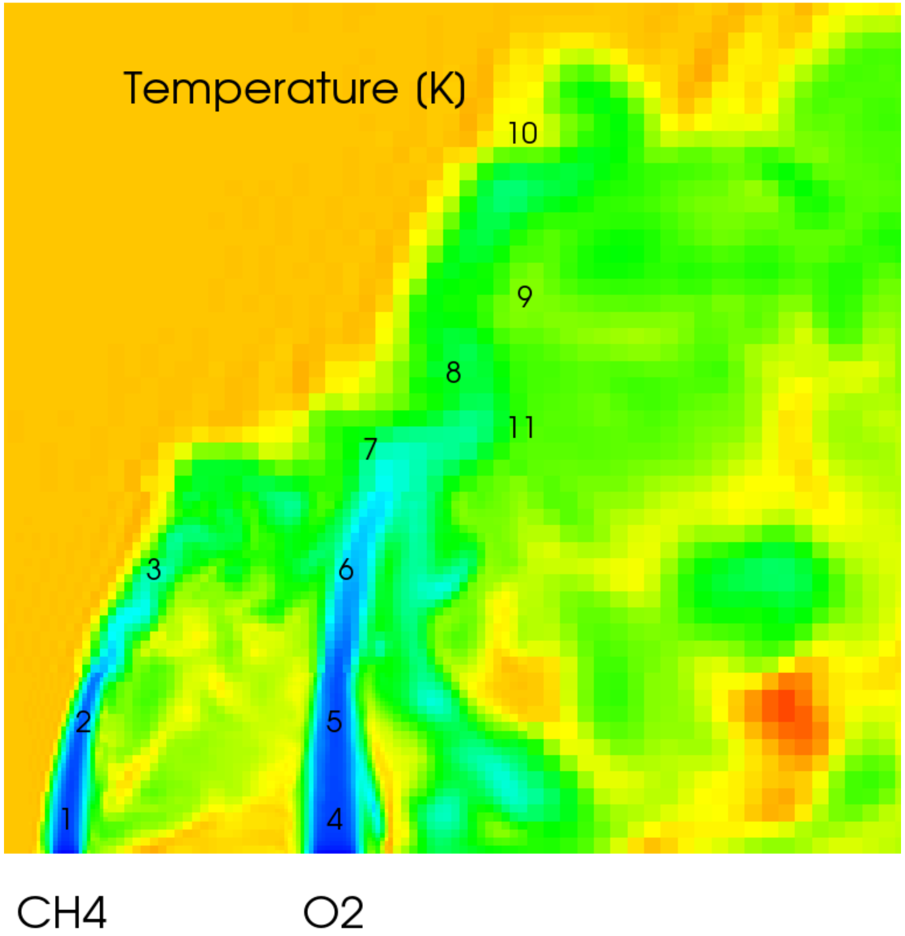


Case 3

- Z= 1 (fuel), Z = 0 (oxidizer); dashed black line: stoichiometric mixture fraction
- Cross flow: sCO_2 at 300 atm; Fuel jet diameter is 3 mm, O2 jet diameter is 5 mm
- Case 1 and 3 - distance between jets is 28 mm; Case 2 - 14 mm
- A bigger and continuous zone of stoichiometry is visible when the two jets are closer indicating enhanced mixing
- Mixing dependent on injection locations & conditions – difficult to optimize

Task 5: LES Studies of Supercritical Mixing and Combustion

Equilibrium Calculations using PSR



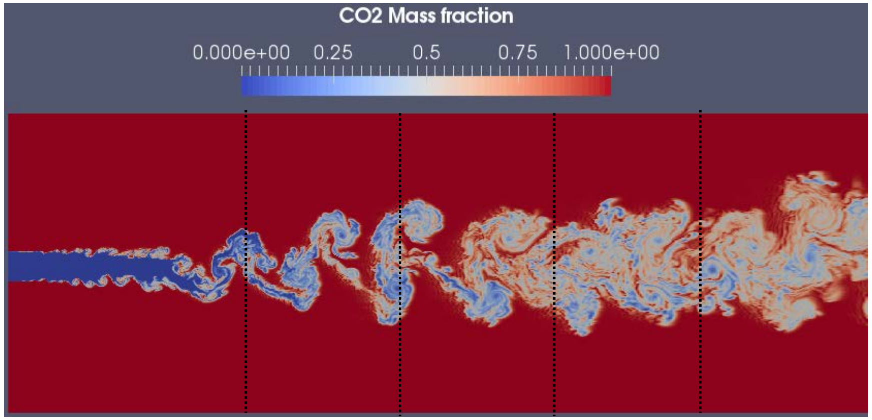
- Points were selected from the LES as an input to PSR
- Initial concentrations of species and temperature were selected at these points.
- The equilibrium temperature, species concentrations are tabulated in the next slide.
- From the table we see that the points 7, 8 and 9 where the oxidizer and fuel have mixed we get combustion
- Shown for Case 2

Task 5: LES Studies of Supercritical Mixing and Combustion

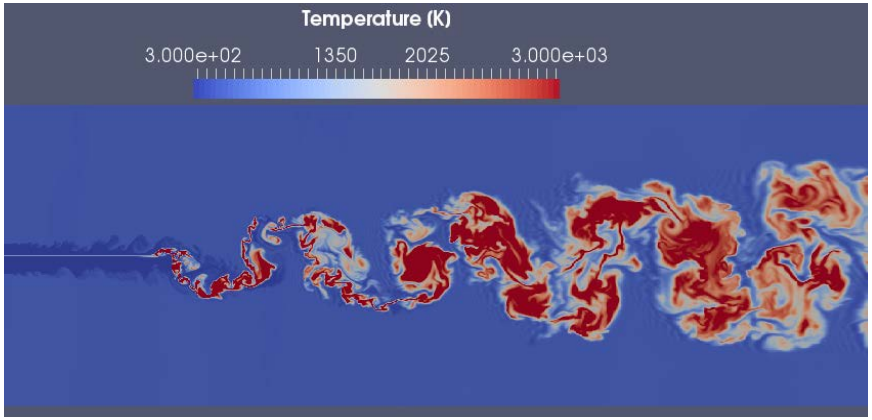
Point	Temp. Tin{K}	CH4 Conc.	O2 Conc.	CO2 Conc.	Temp. Eq.{K}	CH4 Eq.	O2 Eq.	CO2 Eq.
1	361.22	0.989	0.0	0.011	361.22	0.989	0.0	0.011
2	385.95	0.980	0.0	0.02	385.93	0.980	0.0	0.020
3	695.79	0.325	0.0	0.675	645.30	0.313	0.0	0.687
4	363.14	0.0	0.999	0.001	363.14	0.0	0.999	0.001
5	364.06	0.0	0.998	0.002	364.06	0.0	0.998	0.002
6	474.49	0.0	0.948	0.052	483.78	0.0	0.947	0.053
7	654.53	0.028	0.545	0.427	1184.11	0.0	0.546	0.454
8	697.67	0.044	0.414	0.542	1446.9	0.0	0.325	0.675
9	807.32	0.042	0.236	0.722	1464.7	0.0	0.151	0.849
10	1026.6	0.005	0.076	0.919	1104.5	0.0	0.064	0.936
11	859.22	0.100	0.015	0.885	762.65	0.059	0.0	0.941

2D Spatial Mixing Layer

- Splitter plate: 1.2 mm
- CH_4 jet of 3mm, 30 m/s, 300K
- O_2 jet of 5 mm,30 m/s, 300 K
- Outer jets of CO_2 at 50 m/s, 500k
- 1 atm, 200 atm and 300 atm cases
- 5-species reduced kinetics from Task 3
- New analysis shows that vapor-liquid equilibrium (VLE) can occur under supercritical conditions



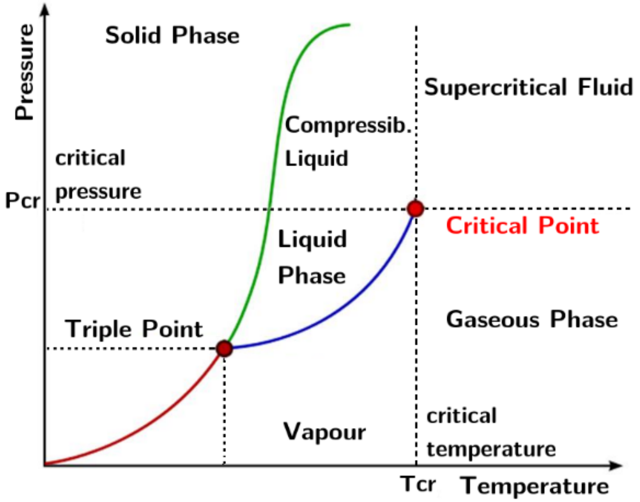
Mixing Studies, CO_2 Contours



Reacting Studies, Temp Contours

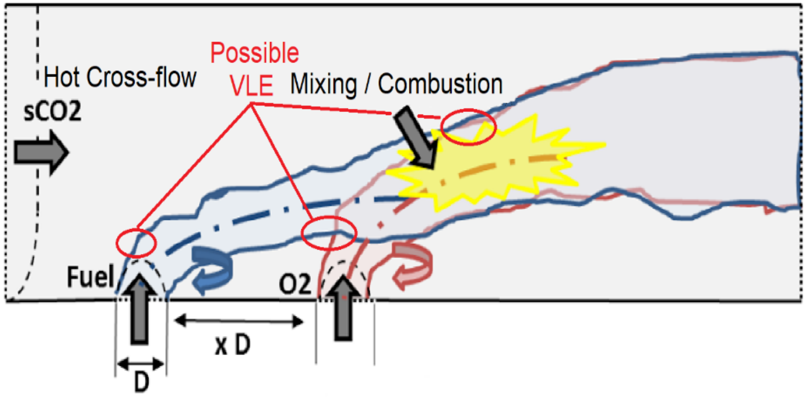
Vapor-Liquid Equilibrium in Supercritical Mixtures

- Single species: the phase is uniquely defined by the equilibrium diagram
- Subcritical regime: jet exhibits atomization, droplets, and sharp gas/liquid interface
- Supercritical regime: Interface is diffused and no droplet formation
- Mixtures: VLE exists at interface for given (p, T) and composition z_i.
- JICF can have local VLE regions in
    - CH_4-CO_2, O_2-CO_2 interfaces
    - CH_4-O_2-CO_2-H_2O regions
- Critical properties of each component play a crucial role to determine VLE
- Need to include VLE effects to account for mixture effects



Sub-critical jet

Super-critical jet



Future of Task 5

- Revisit the earlier supercritical JICF mixing case, accounting for presence of VLE to reassess the problems seen in the past.
- Continue spatial mixing layer studies with different conditions
  - Binary mixing under supercritical conditions
  - Reacting cases under supercritical conditions
- Autoignition studies will require more detailed kinetics
  - 19 species chemistry from Task 3 available

Summary of Year 2 Achievement

- High pressure shock tube commissioned
  - System validation (vs simulation, previous work)
  - Measurement of autoignition delays with high CO_2 concentration (above critical pressure of CO_2)
- Different optimized reduced kinetic models developed and implemented in CFD
- Governing equation developed for theoretical frame work
- LES investigation of JICF
  - Not efficient on mixing
  - Sensitive to kinetic models
  - Jet mixing, quick estimation of autoignition location
  - Vapor-liquid equilibrium plays important role

Thank you!
&
Questions?

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