

Presentation at NETL Ammonia Combustion Technology
Group Meeting, 12th Nov 2024

Dynamic Stability and Emissions in Premixed CH₄/NH₃ Swirling Flames with Nanosecond Pulsed Discharges

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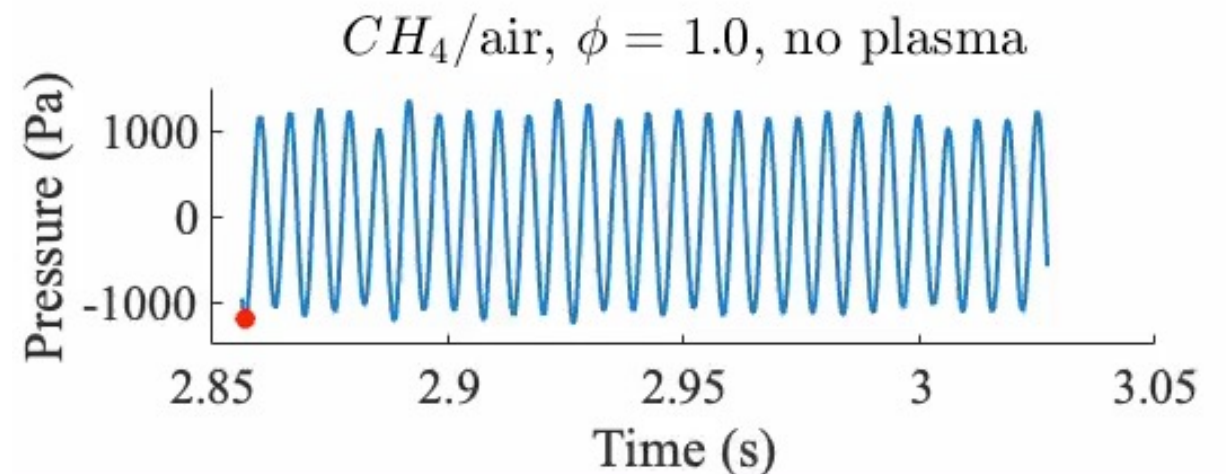
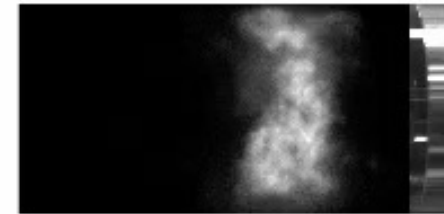
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Background - I

Dynamic Stability of NH₃/air flames : Definitions

- Dynamic Stability:
 - Occurrence of large pressure pulsations in the combustion chamber
 - Most dangerous of these is when the peak amplitude is a few percent of the chamber pressure
- All things being equal, how does substituting C_xH_y with NH₃ change characteristics of these oscillations
 - Can we link these to some flamelet characteristic or fuel chemistry/transport?
- How do we reduce/eliminate these oscillations?
 - (preferably in a way that affects the fuel chemistry/transport?)

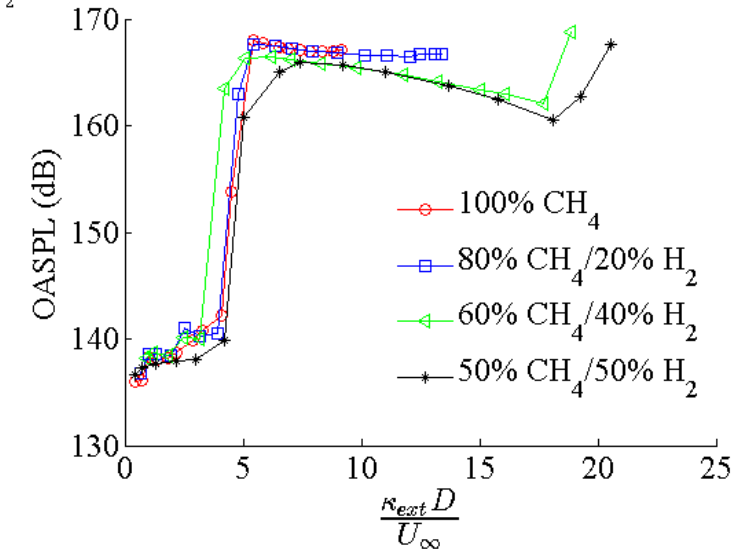
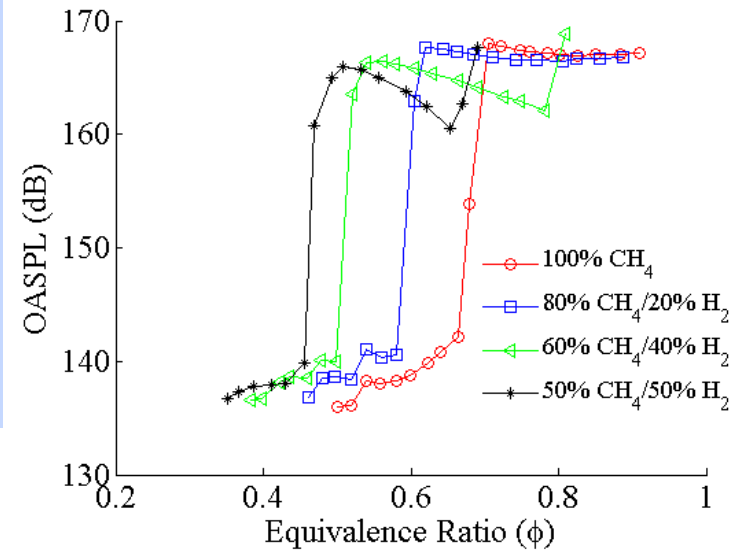


1000 Pa \equiv 154 dB

Background - I

Related: Dynamic Stability due to H₂ addition

- Related question, that we have addressed before:
 - How does switching from C_xH_y to H₂ change the dynamic stability of the combustor
- What changes:
 - Frequency, amplitude
 - Point of onset

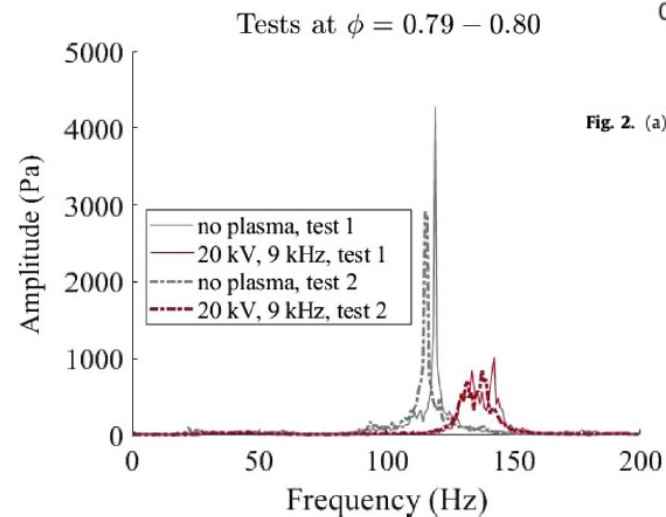


At stoichiometric conditions	NH ₃	CH ₄	H ₂
$T_{equil.}$ (K)	2075	2225	2400
S_u^o (cm/s)	7	35	175
K_{ext} (s ⁻¹)	~ 100	> 2200	> 30,000

Background – II

The effects of NRPD on Flame Stability and Emissions

- Prior work by us and others have shown that NRPDs can improve flame stability, but only for CH₄/air or other hydrocarbon flames
- Studies have shown that NRPDs can reduce NO_x in NH₃/air flames
- Can all the benefits of NRPDs be exploited for CH₄/NH₃/air flames? Improve stability and reduce NO_x?
 - This is a key motivation for the present work



Effect of NRPDs on instability amplitude reduction. Reproduced from Shanbhogue, ..., Ghoniem, 2023, J. Prop. Power

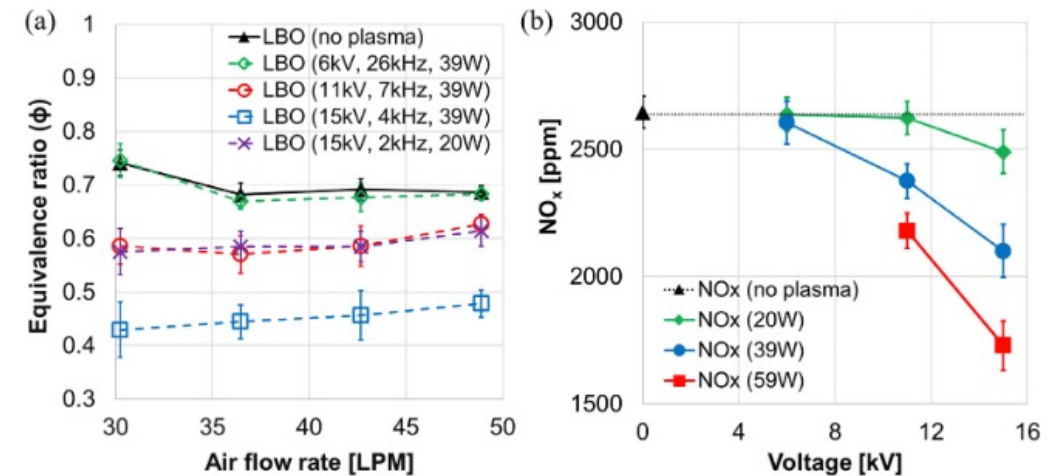
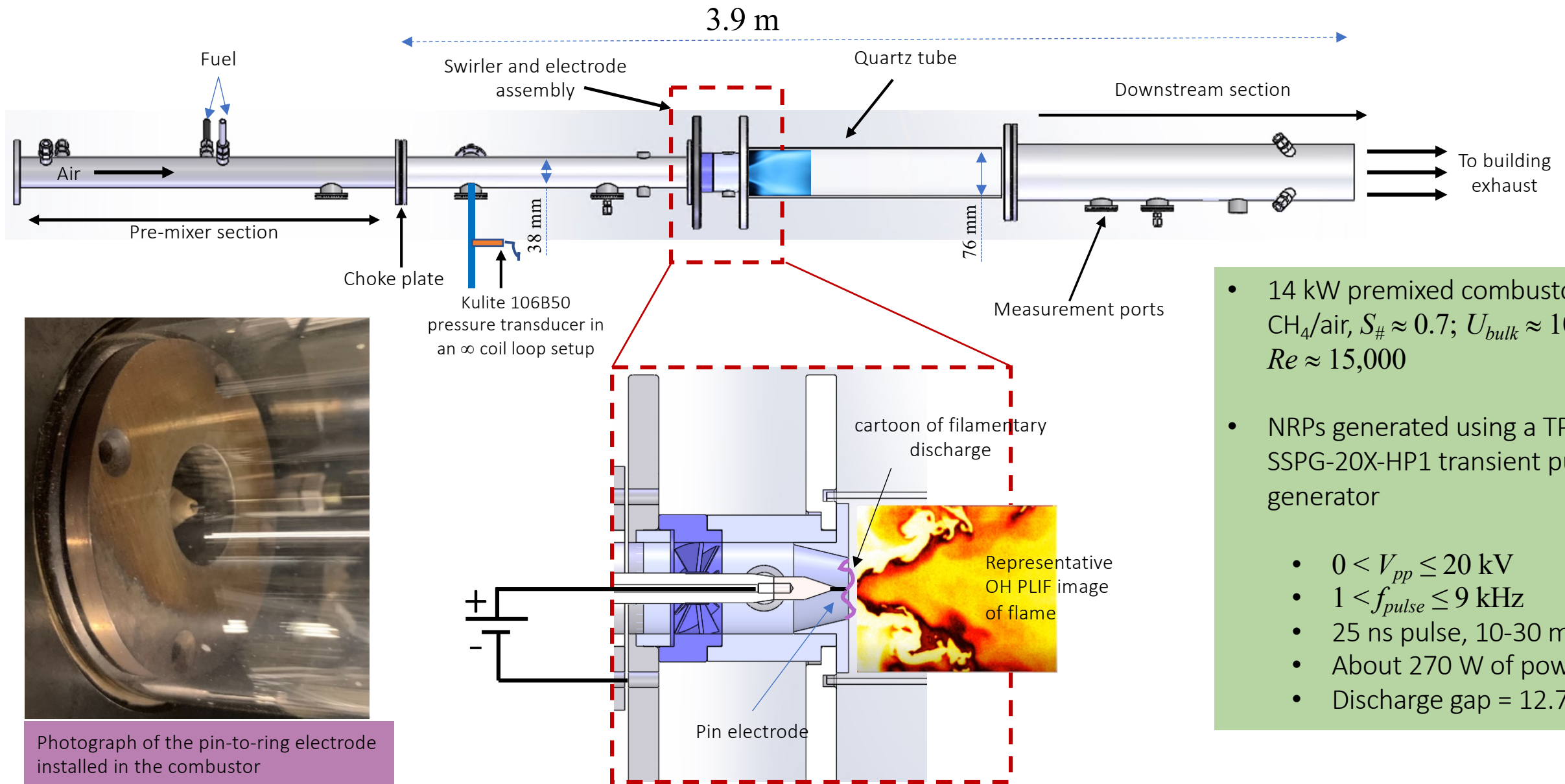


Fig. 2. (a) lean blowoff limits of ammonia/air flames with and without plasma, (b) NO_x emissions without and with plasma ($\phi = 0.94$).

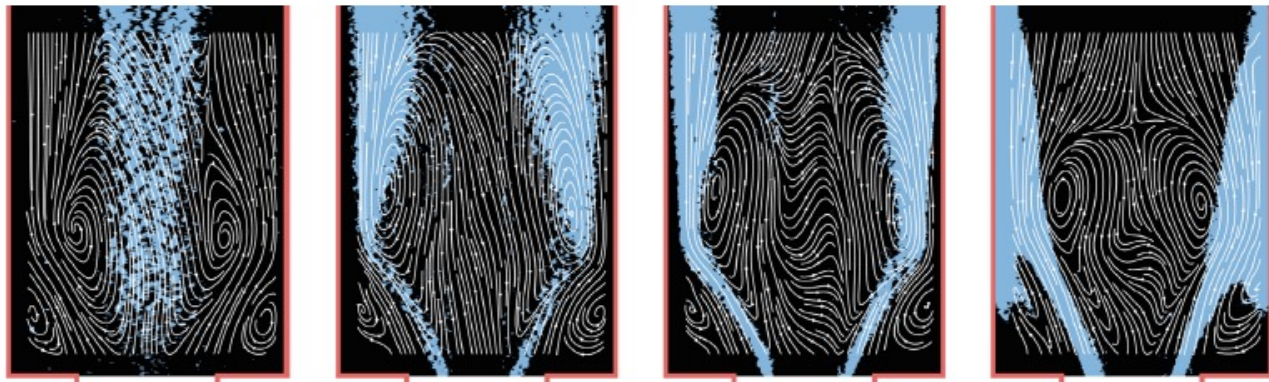
Effect of NRPDs NO_x reduction. Reproduced from Choon, ..., Sun, Comb. Flame, 2021

Experimental Setup I – RGD Combustor



- 14 kW premixed combustor, CH_4/air , $S_{\#} \approx 0.7$; $U_{bulk} \approx 10$ m/s; $Re \approx 15,000$
- NRPs generated using a TPS SSPG-20X-HP1 transient pulse generator
 - $0 < V_{pp} \leq 20$ kV
 - $1 < f_{pulse} \leq 9$ kHz
 - 25 ns pulse, 10-30 mJ/pulse
 - About 270 W of power
 - Discharge gap = 12.7 mm

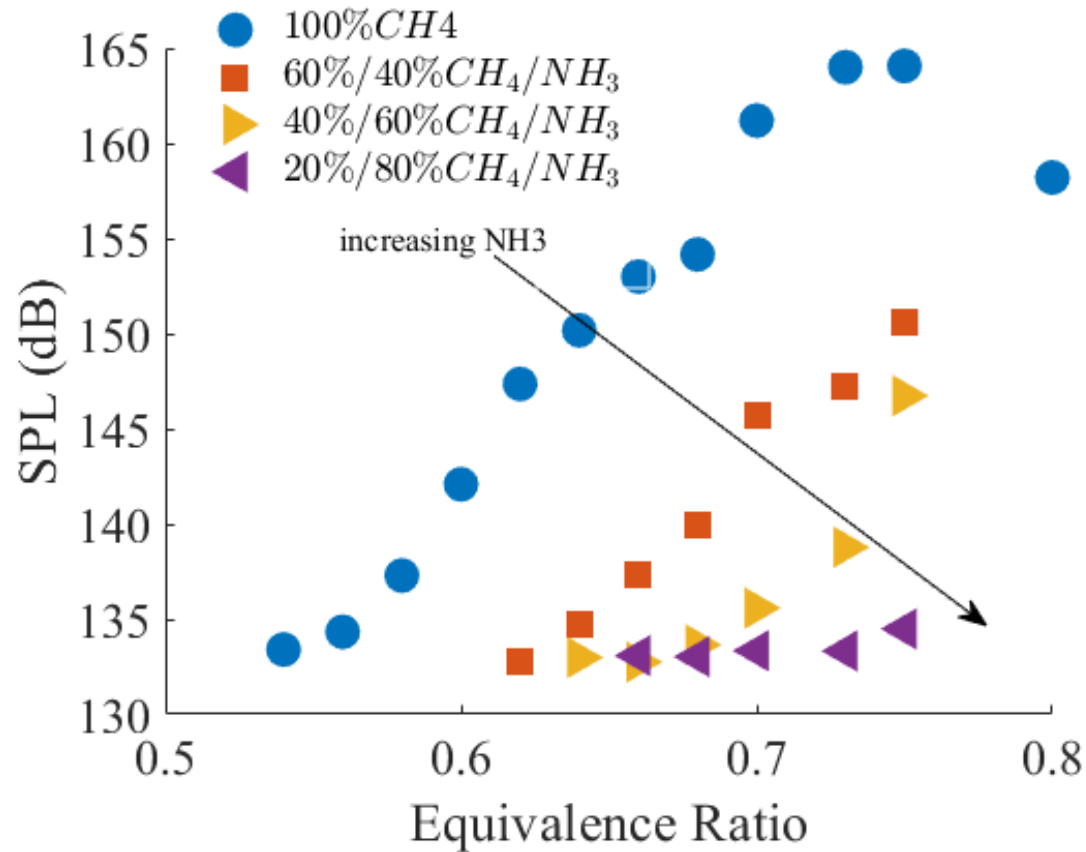
Results: Flame Macrostructure Transitions



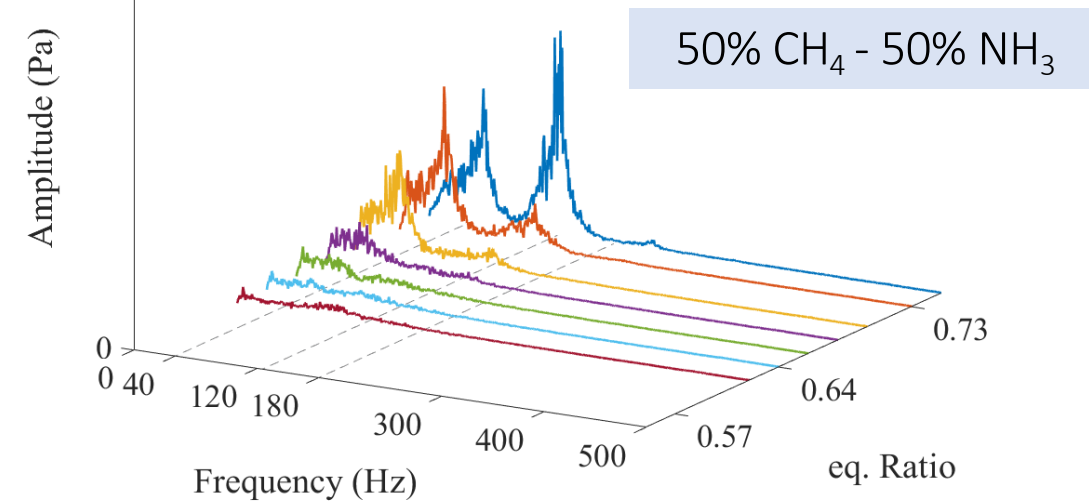
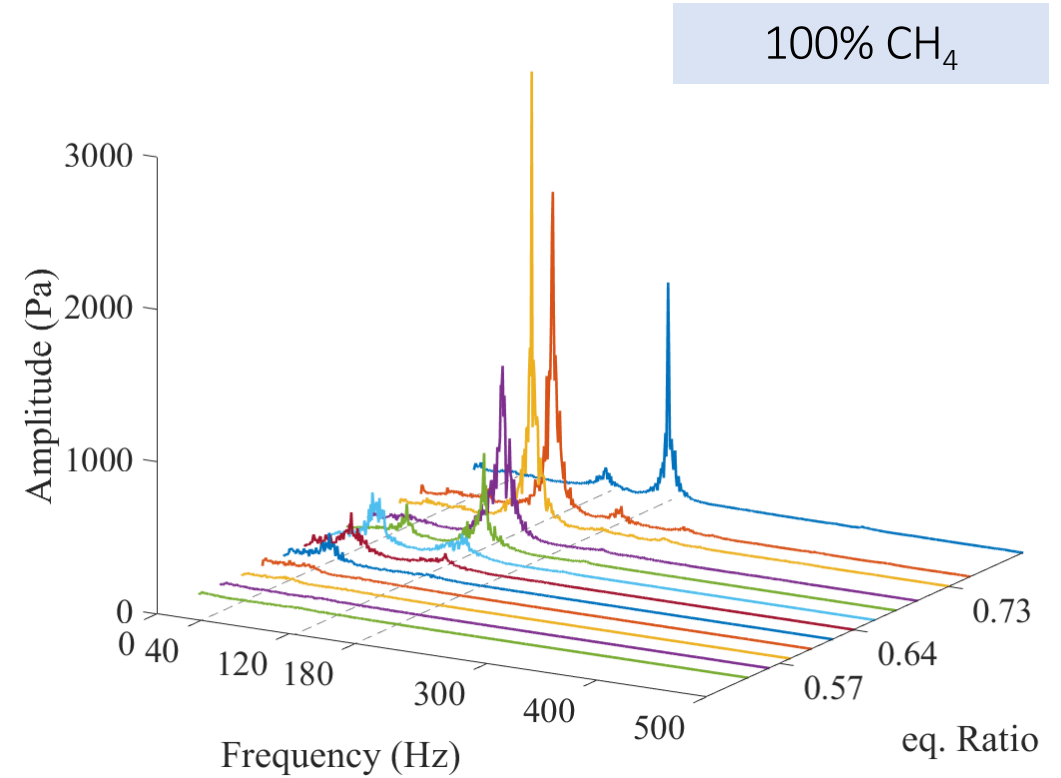
Ref: Taamallah, Shanbhogue & Ghoniem, 2016, Comb. & Flame

Fuel →	100% CH ₄	60%CH ₄ , 40%NH ₃	20%CH ₄ , 80%NH ₃
Equiv. Ratio↓			
0.75	 M = IV, $\kappa_{ext} = 1395.61 \text{ s}^{-1}$	 M = IV, $\kappa_{ext} = 638.51 \text{ s}^{-1}$	 M = II, $\kappa_{ext} = 178.55 \text{ s}^{-1}$
0.7	 M = IV, $\kappa_{ext} = 1085.4 \text{ s}^{-1}$	 M = III, $\kappa_{ext} = 500.66 \text{ s}^{-1}$	 M = I, $\kappa_{ext} = 120.68 \text{ s}^{-1}$
0.66	 M = IV, $\kappa_{ext} = 847.13 \text{ s}^{-1}$	 M = II, $\kappa_{ext} = 371.84 \text{ s}^{-1}$	 M = I, $\kappa_{ext} = 76.981 \text{ s}^{-1}$
0.64	 M = III, $\kappa_{ext} = 724.92 \text{ s}^{-1}$	 M = II, $\kappa_{ext} = 312.66 \text{ s}^{-1}$	Flame blew off
0.60	 M = II, $\kappa_{ext} = 474.31 \text{ s}^{-1}$	 M = I, $\kappa_{ext} = 204.51 \text{ s}^{-1}$	Flame blew off
0.56	 M = II, $\kappa_{ext} = 299.77 \text{ s}^{-1}$	Flame blew off	Flame blew off
0.54	 M = I, $\kappa_{ext} = 237.39 \text{ s}^{-1}$	Flame blew off	Flame blew off

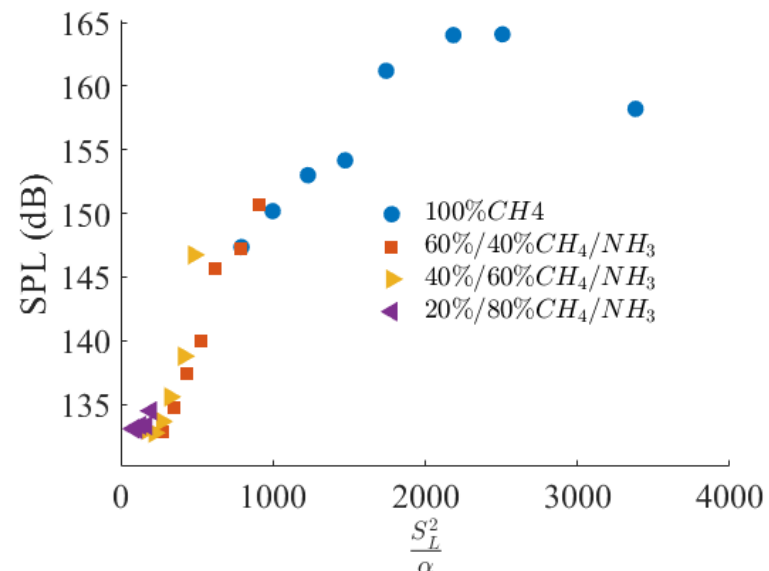
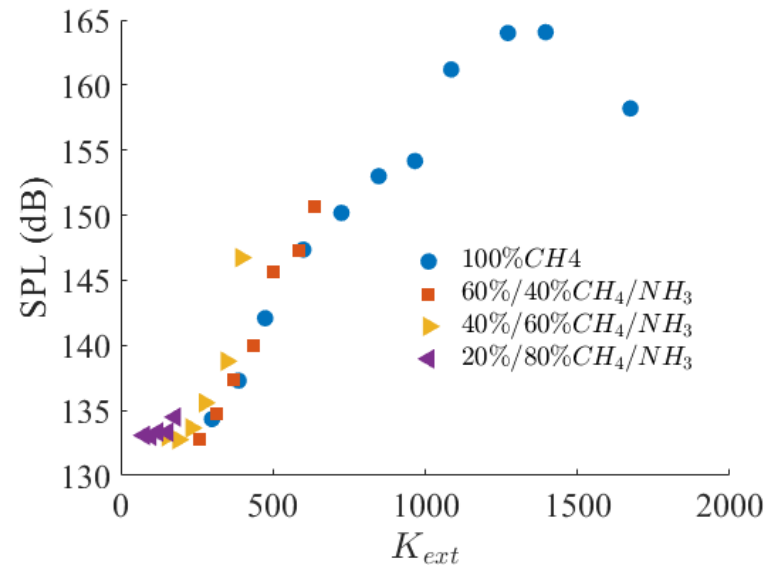
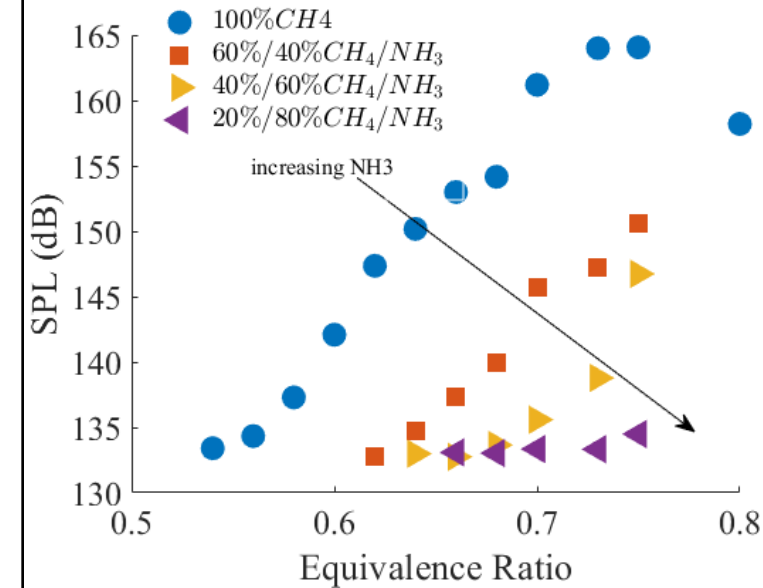
Results: Combustion Instabilities for different CH₄/NH₃ blends



As the percentage of NH₃ in increased, the oscillations are suppressed, and transitions delayed or gone



Scaling with flame time scale



- If the transitions and amplitudes as measured by SPL are a function of the Damköhler number

- Then should scale as $\left(\frac{1}{\tau_{flame}}\right)$

- K_{ext} : Extinction Strain Rate
 - Computed using CHEMKIN

- $\frac{\alpha}{S_L^2}$: Based on thermal diffusivity and burning velocity
 - Computed using Cantera

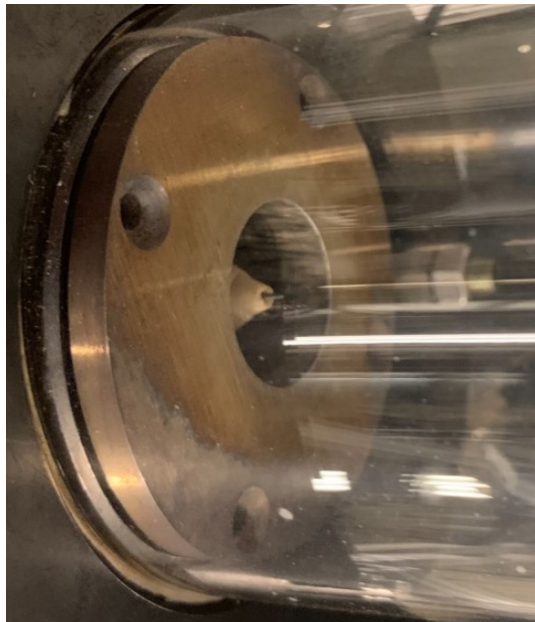
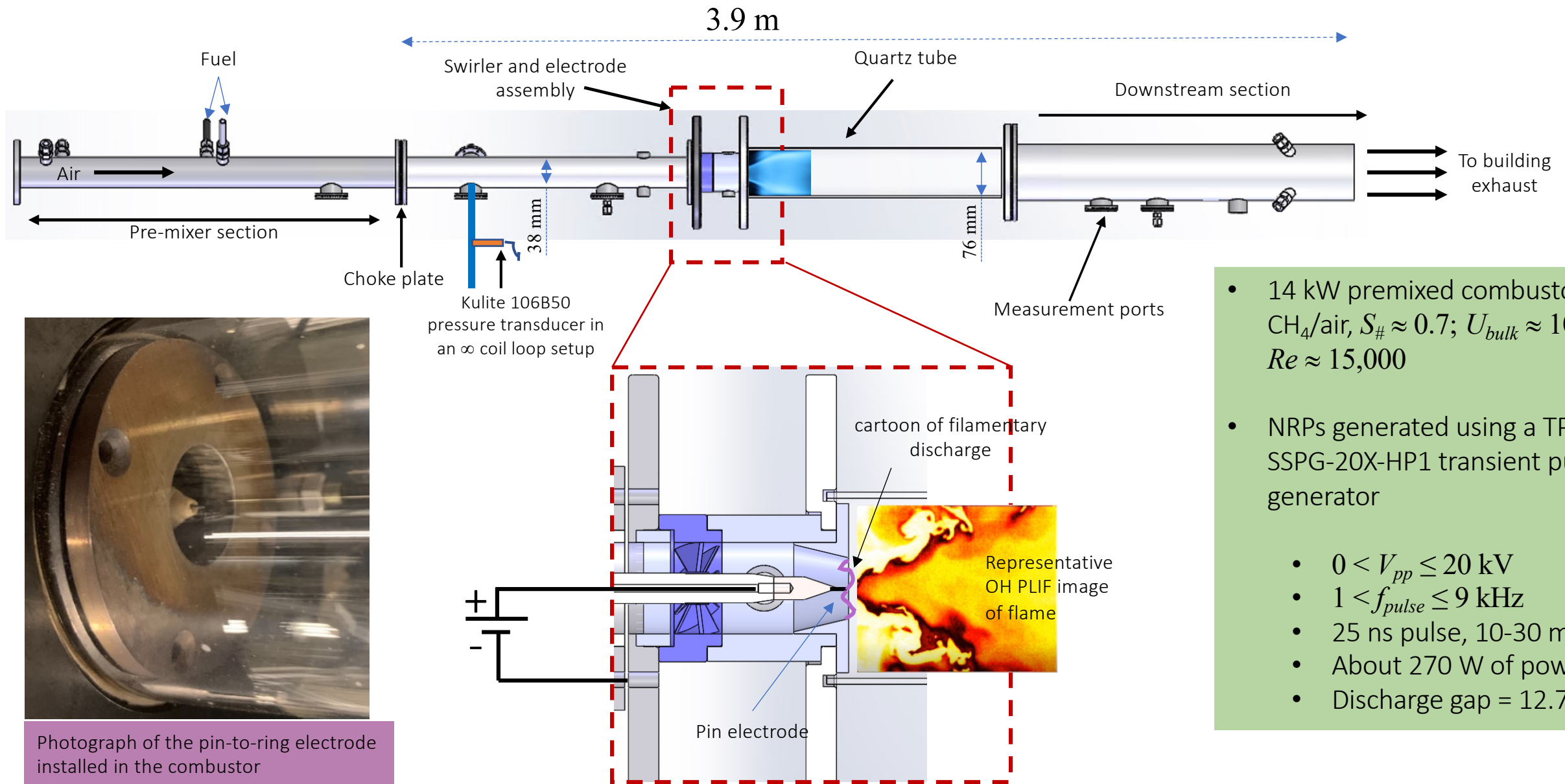
- For appropriate flow time scales, see Taamallah, Shanbhogue & Ghoniem, 2016, Comb. & Flame
 - $\tau_{flow} = 1/2\pi f_{ORZ}$

Damköhler number: $Da = \frac{\tau_{flow}}{\tau_{flame}}$

$$\tau_{flame} = \frac{1}{K_{ext}}, \frac{\alpha}{S_L^2}$$

$$Da = \tau_{flow} K_{ext} \text{ or } \tau_{flow} \frac{S_L^2}{\alpha}$$

Experimental Setup I – RGD Combustor

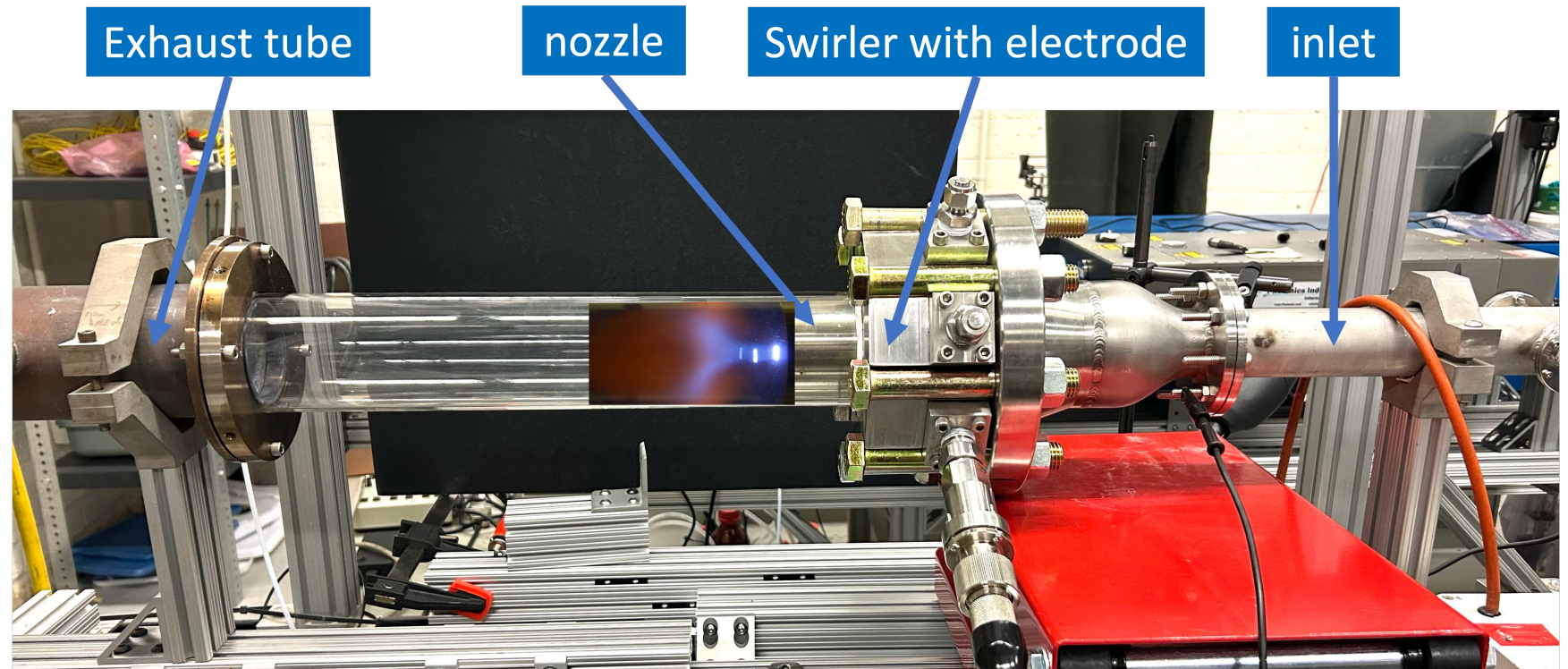


Photograph of the pin-to-ring electrode installed in the combustor

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 - $0 < V_{pp} \leq 20$ kV
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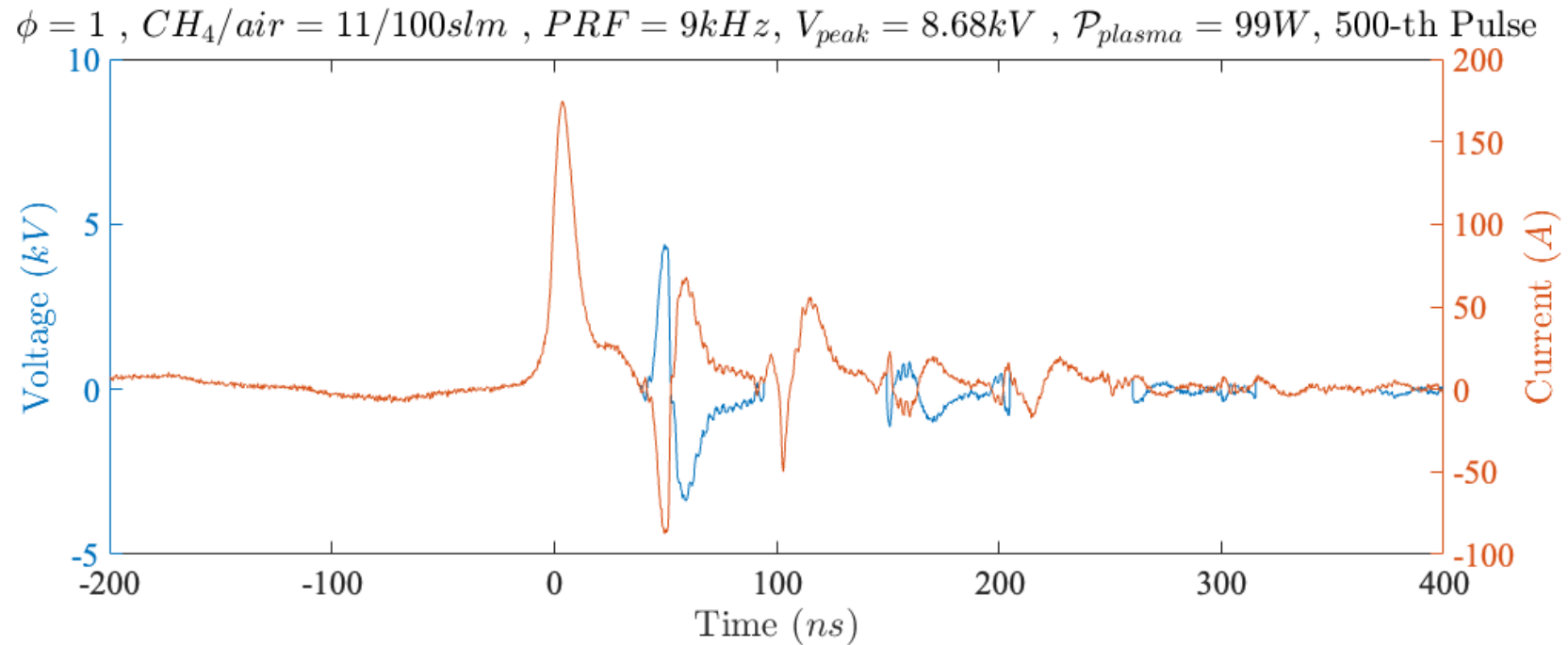
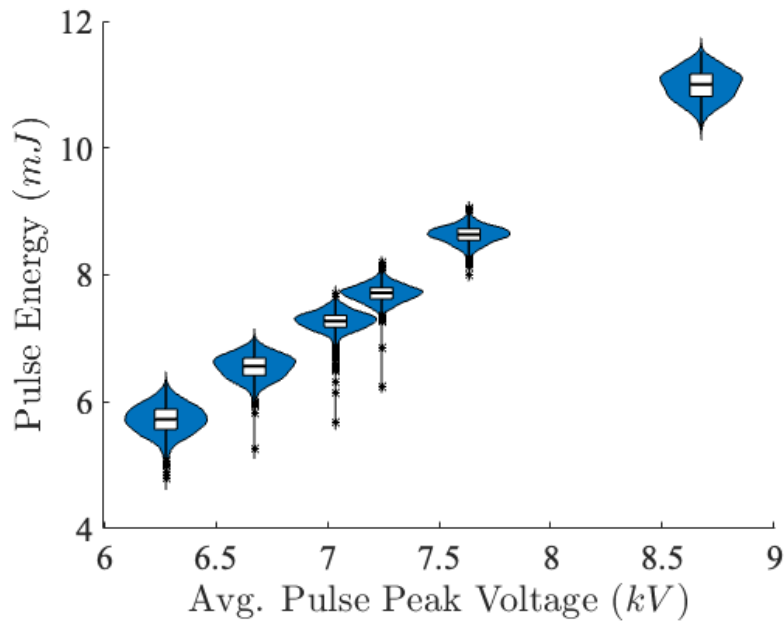
Experimental Setup II – Specter injector

- 6 kW, premixed, non-preheated combustor at atmospheric pressure
 - $Re \approx 20,000$
 - $U_\infty \approx 30$ m/s
 - CH_4 & NH_3 mixed well upstream of swirler
 - Entire setup ≈ 4 m long
 - Emissions tap at 2.66 downstream of dump plane
- The quartz tube in the picture is about 75 mm in diameter
- The flow flows into the quartz tube via a 9.1 mm nozzle
 - Center body with sharp tip functions as the anode
 - Discharge gap is thus 4.55 mm



- Key difference between this combustor and the RGD combustor
 - Discharge gap is 4.55 mm vs. 12.7 mm
 - Pulse energy for actuation is < 10 mJ/pulse vs.

Discharge Generation & Quantification (Specter Injector)



- NRP discharges generated using a TPS transient pulse generator (SSPG-20X-HP1)
- Typical FWHM pulse widths are about 12 ns in duration with a frequency of 9 kHz. Peak voltage, at the measurement location, is 6-9 kV
- Energy deposited in the discharge per pulse is 5-12 mJ
- Electrical waveforms measured using a single current shunt in the middle of a 10m coaxial cable
- Ref: C. Pavan, S. Rao, and C. Guerra-Garcia, "Tutorial: Electrical Measurements in Nanosecond Pulsed Plasma Reactors", J Phys D: Appl. Phys 2024 (<https://doi.org/10.1088/1361-6463/ad73e5>)

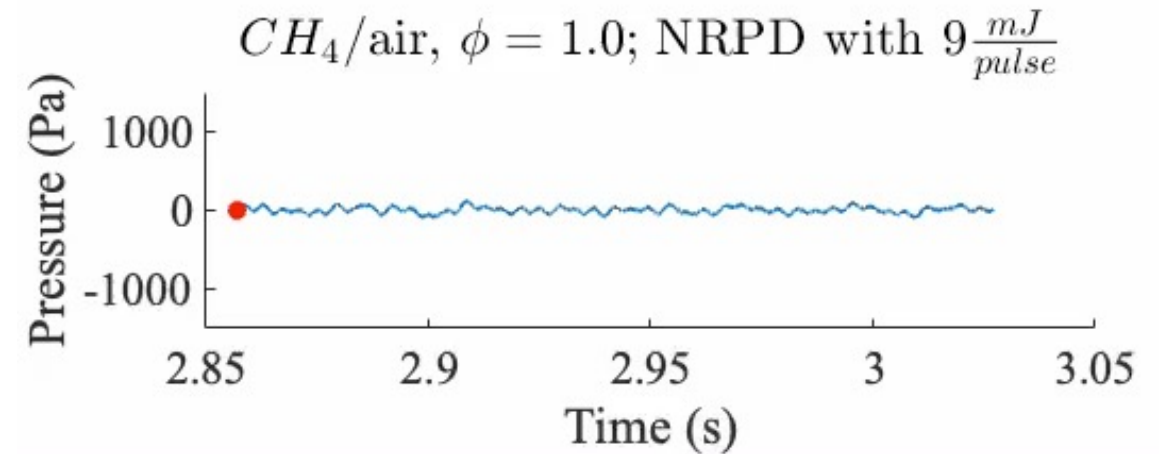
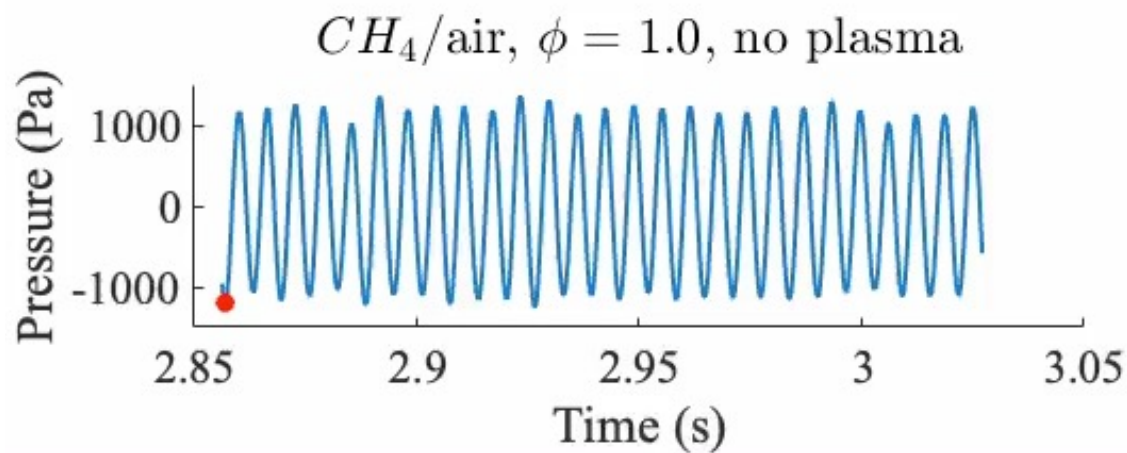
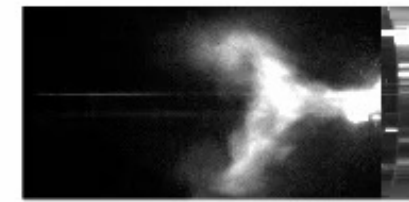
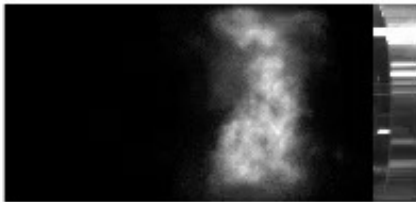
Flame Macrostructures

Equivalence Ratio	Flame Without Plasma	Flame With Plasma
1.0		
0.9		
0.8		
0.7		
0.6		

Equivalence Ratio	90% CH ₄ /10% NH ₃	20% CH ₄ /80% NH ₃
1.0		
0.9		

- Without plasma: Identical compact flames at a very broad range of equivalence ratios
 - Unlike many other studies no macrostructure transitions or very long flames
- With plasma: Clear anchored flame resembling the letter Y
 - For large amounts of NH₃, the flame is more distributed downstream, but still relatively compact (no tubular flames observed)

Effect of NRPD on Instabilities

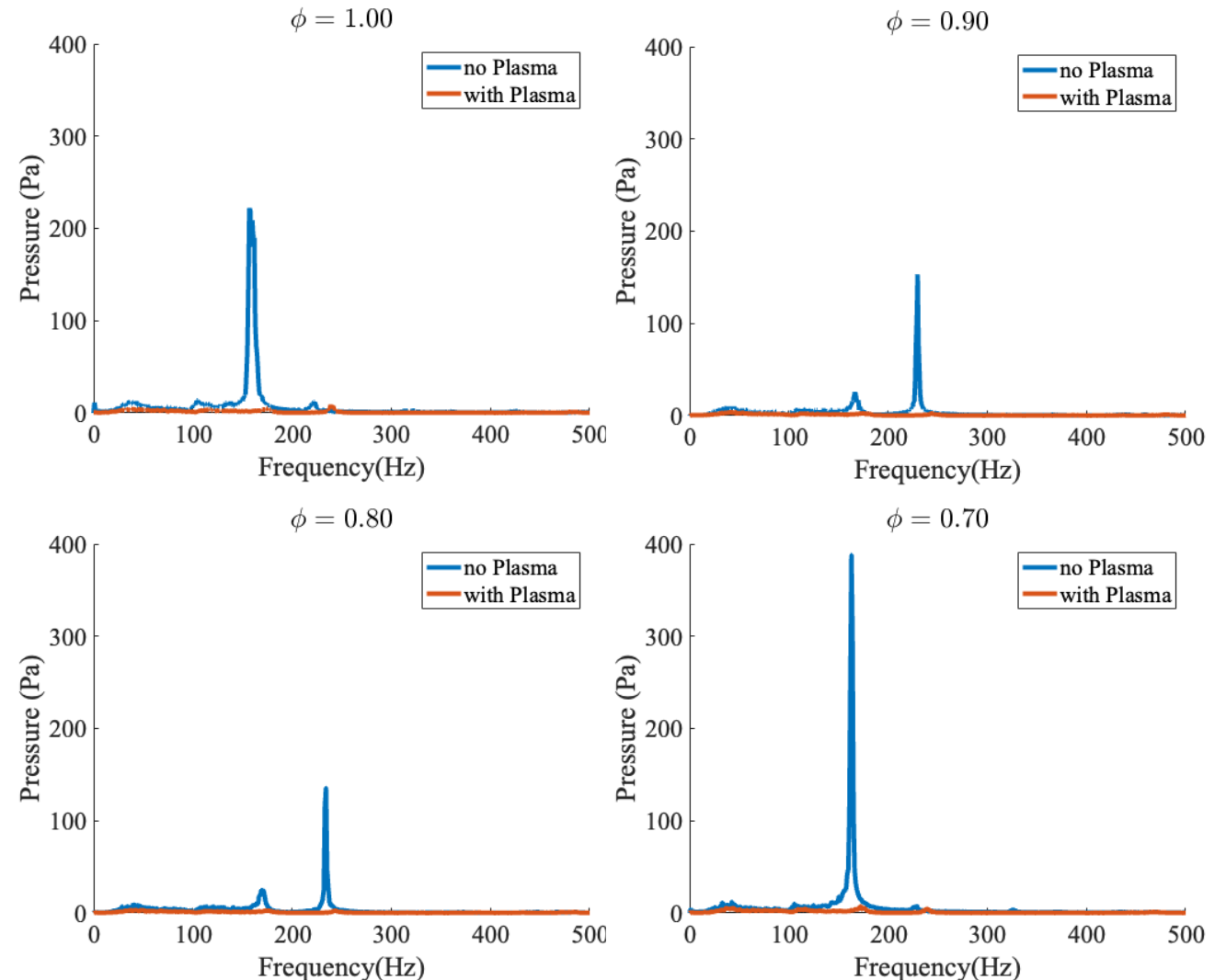


	Without plasma	With Plasma
Amplitude	$\approx 1000 \text{ Pa}, 154 \text{ dB}$	$< 50 \text{ Pa}, 133 \text{ dB}$
Flame Shape	Lifted, compact	Anchored, compact

Effect of NRPD on Instability Suppression

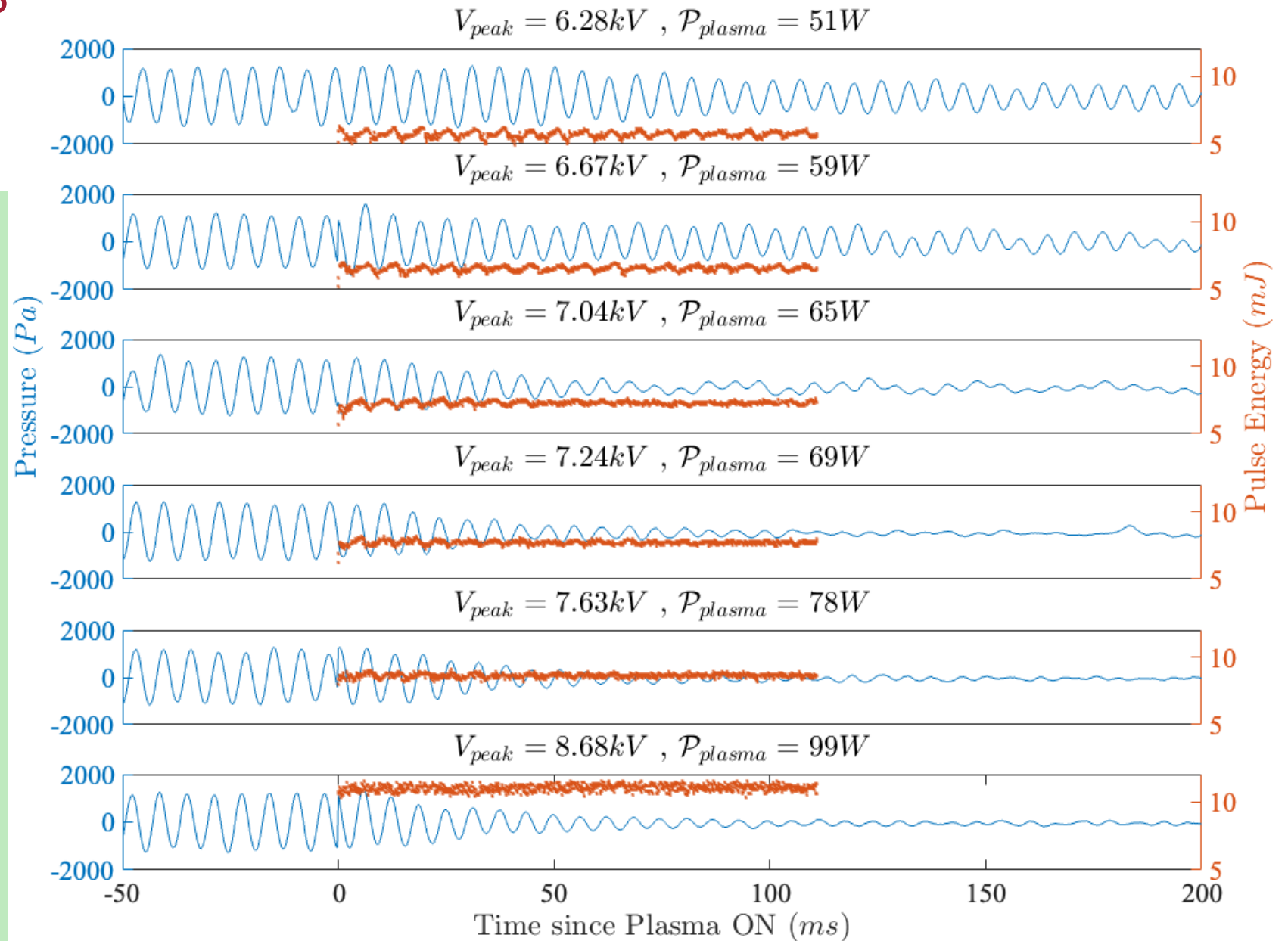
Spectra for various conditions

- We measured instability amplitudes and frequencies at various equivalence ratios
 - The instability frequency depends on equivalence ratio
 - Why the combustor chooses one mode over other discussed in Hong, Shanbhogue, Speth & Ghoniem, 2013, Comb. & Flame
- For all cases, with plasma, the instability vanishes with at sufficiently high pulse energies
 - By this we mean the spectra reverts to broadband flow noise
 - Peak noise reduction ≈ 25 dB
 - This is a significant improvement compared to our previous work (J. Prop. Power, 2023) where we saw only 3 dB or so reduction (amplitude is halved due to NRPD's)



NRPD Characteristics During Instability Suppression

- A question of great interest is how much plasma power is required to suppress the instability
 - This is measured in the figure
 - For all conditions in the figure, the flame was that of a stoichiometric CH₄/air mixture
- The plasma is turned on at $t = 0$ and stays on
 - Power measurements done only till 110 ms
 - Note that the instability dies out in a few milliseconds
 - At 69 W & above, the instability dies
 - The flame power is nearly 6 kW
- The required plasma power is 1-2% of the flame power
- The energy deposited doesn't remain constant but fluctuates due to the periodic motion of the flame, the "backward problem". See Pavan, ..., Guerra-Garcia, Plas. Sour. Sci. & Tech. 2024.



Background - IV

NO_x emissions from NH₃/air flames

- For conventional hydrocarbon fuels in gas-turbines, NO_x is in single digits, even without post-combustion emissions controls
 - This is even when there is substantial N₂ in the gas-mixture (e.g. process gas)
- For NH₃, NO_x can easily reach 1000 ppm (in laboratory flames) without any post-combustion emissions controls

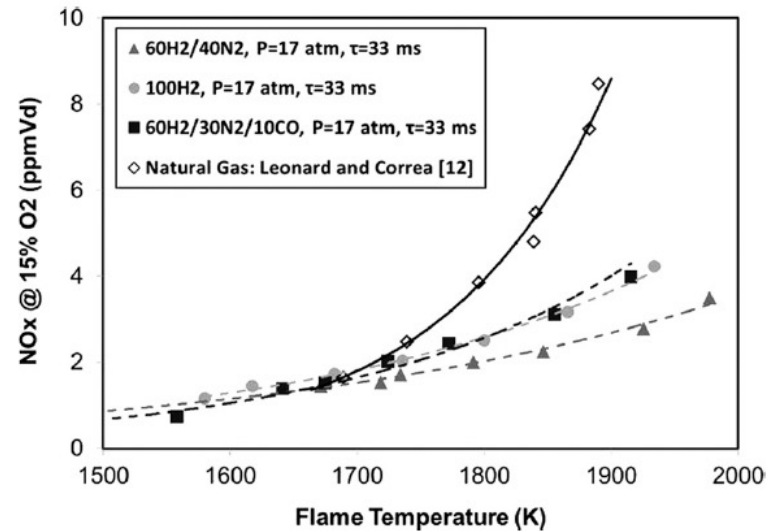


Fig. 7 Perfectly premixed NO_x measurements for high hydrogen fuels taken at $P = 17$ atm and $\tau = 33$ ms

Baseline: NO_x emissions from hydrocarbon flames. Reproduced from York, ..., Yilmaz, 2013, ASME TurboExpo

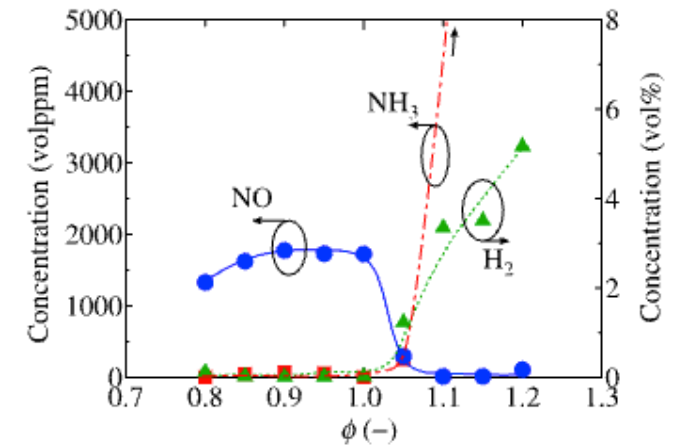


Fig. 7 – Concentrations of NO, NH₃ and H₂ for cylindrical liner with 200 mm liner length in the case of the swirl number of $S = 0.736$.

NO_x emissions from laboratory scale NH₃/air flames. Reproduced from Hayakawa, ..., Kobayashi 2017. Int J. H₂ energy

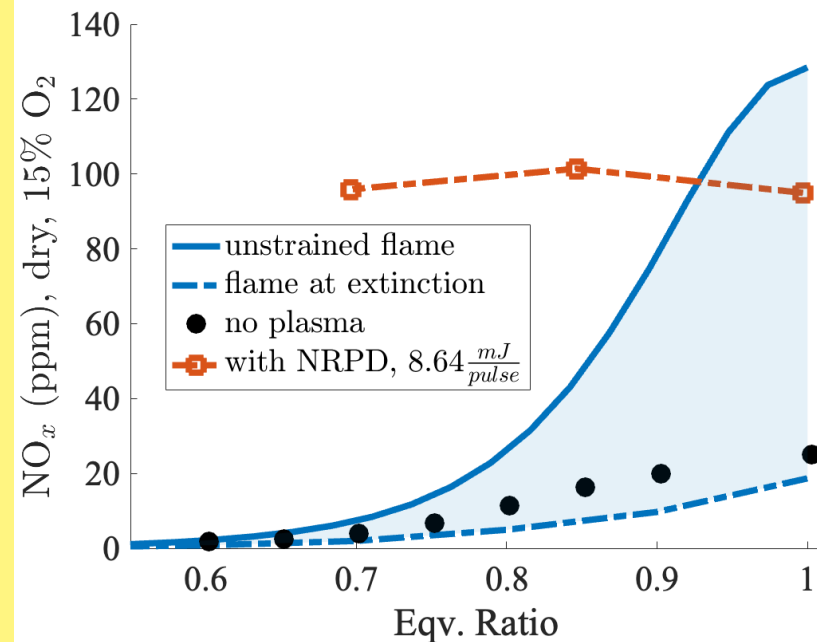
Another key question is what kind of kinetic models and flamelet models can model NO_x from these combustors. Kinetic models that reproduce ignition delays and burning velocities are available in literature, but struggle with NO_x prediction.

Effect of NRPD on NO_x Emissions

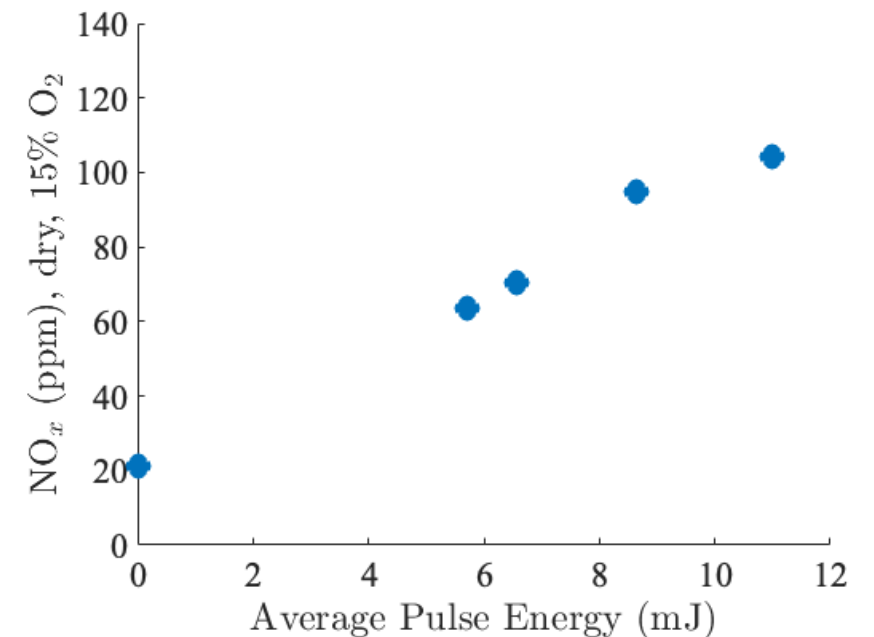
CH₄/air flames

Basic question: What kind of flamelet model is appropriate for NO_x predictions in CH₄/air flames (without NRPD)

- Blue shaded area shows the NO_x emissions for strained flames
 - Lower limit – unstrained flame
 - Upper limit – flame at extinction strain rate
- Calculations done using GRI 3.0
 - There are more comprehensive and validated mechanisms such as NUIGMech1.3, but these are computationally expensive
- Measurements indicate that the emissions in this combustor are consistent with a strained flame



- With NRPD the NO_x emissions jump to a constant value, apparently independent of the equivalence ratio

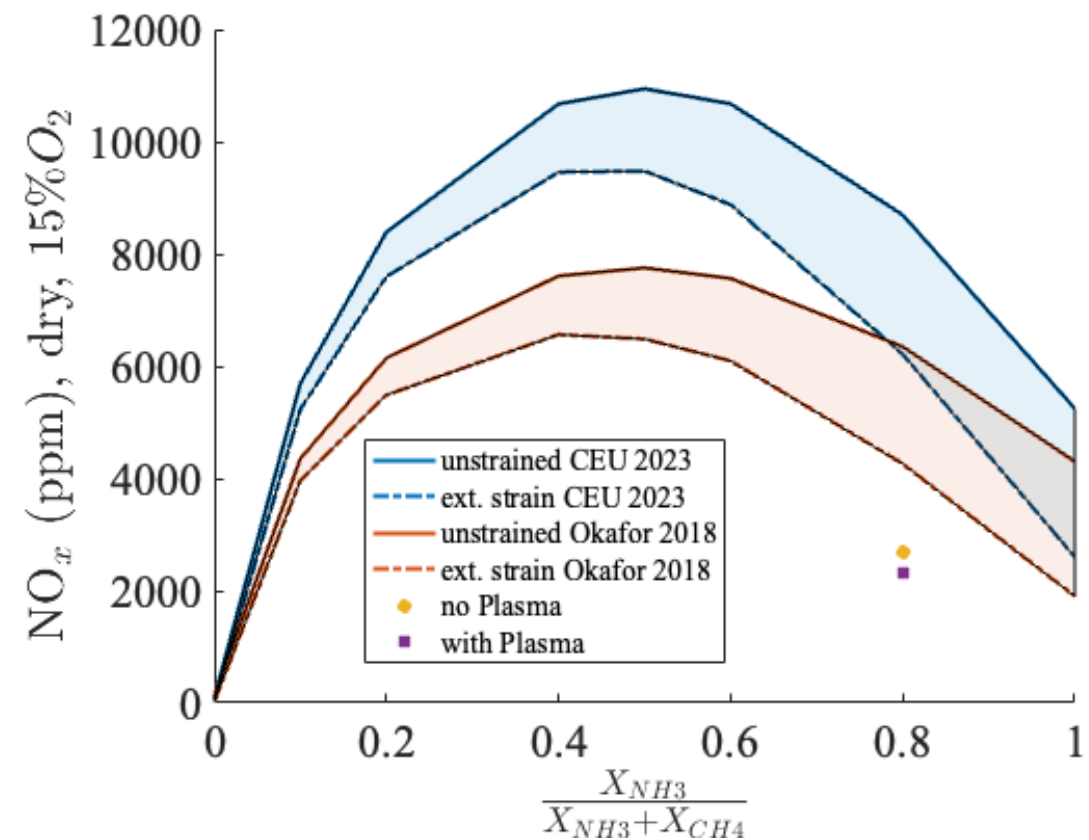


- The emissions are dependent on the pulse energy deposited in the flow
- The highest pulse energies guarantee complete suppression but produce more NO_x
- Optimal pulse energy a balance between allowable instability amplitude and excess NO_x

Effect of NPRD on NO_x Emissions

CH₄/NH₃/air flames

- We were able to test the effect of NPRD on NO_x for just one condition – 80% NH₃ and 20% CH₄ + air
 - For higher ammonia fractions, we couldn't stabilize the flame at these velocities
 - For lower ammonia fractions, the NO_x values exceed 5000 ppm, which is the detection limit of our analyzer.
- But NPRD reduces NO_x by nearly 200 ppm at these conditions
- Just like for CH₄/air flames, we attempted to model the emissions using different kinetic model, but none of these match our experimental observations



Conclusions

Dynamic Stability:

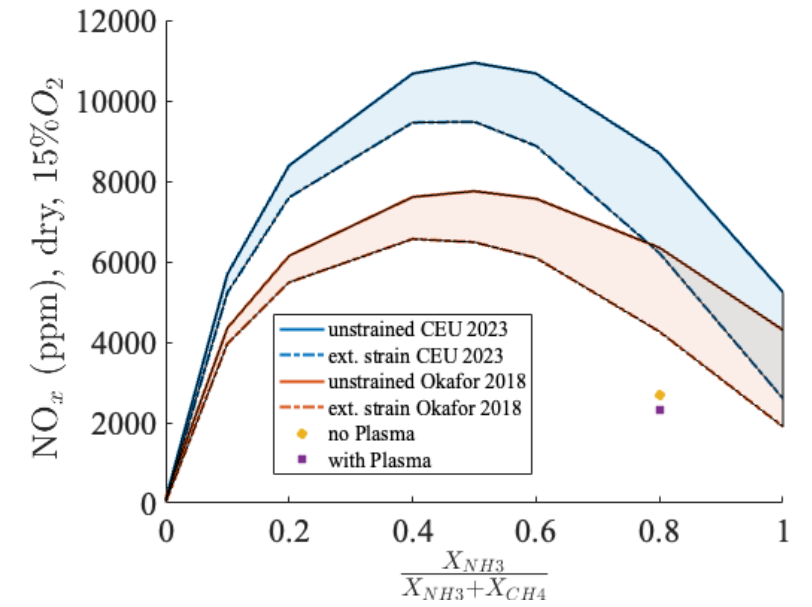
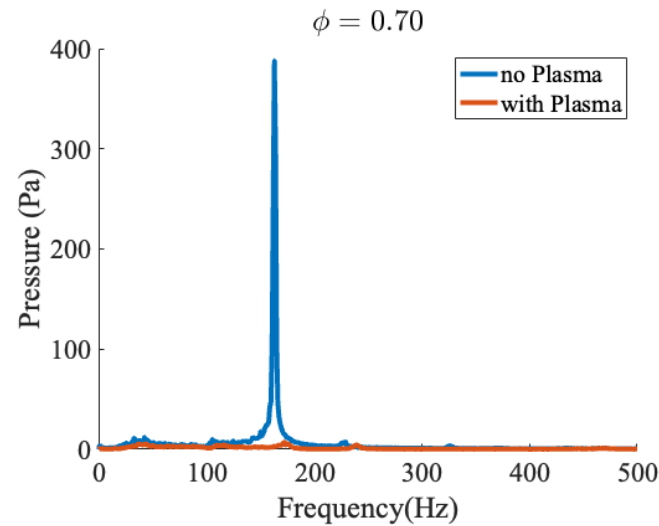
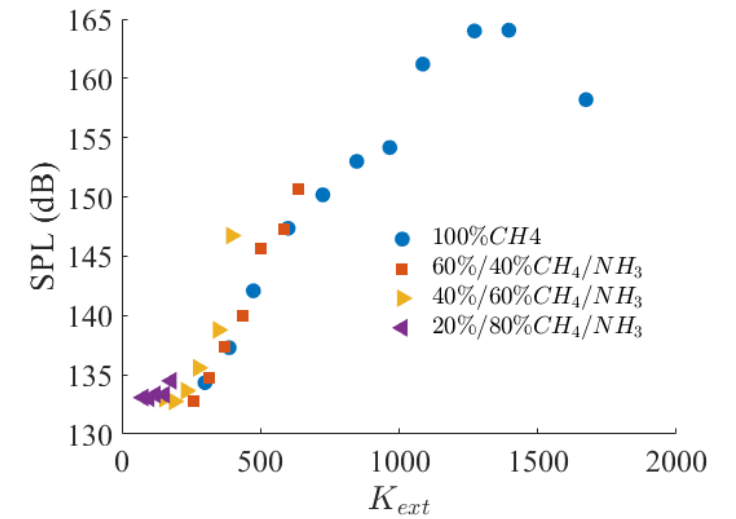
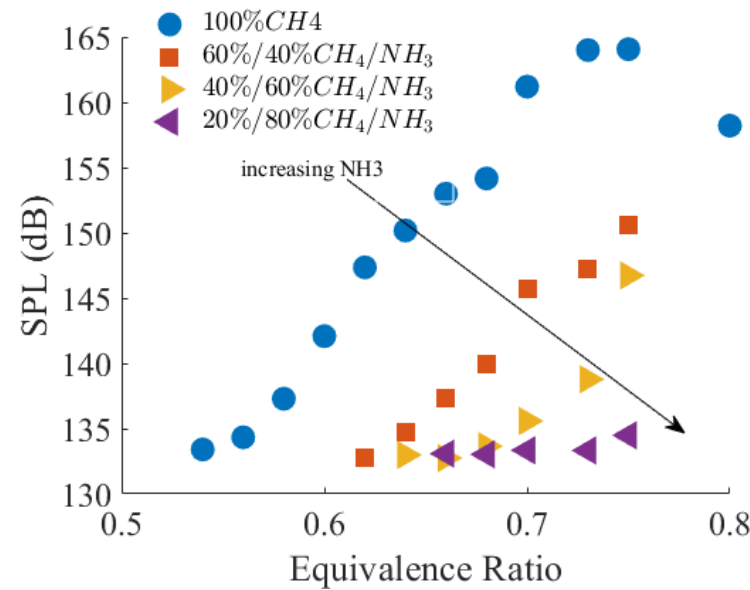
- What does ammonia addition do to a combustor nominally fired with hydrocarbons:
 - It makes it more stable, -- narrow window of pulsations
 - This is the opposite of what addition of hydrogen does to the combustor, i.e. makes it more unstable – wider window of conditions where pulsations occur
- Data scales with $Da = \tau_{flow} K_{ext}$ or $\tau_{flow} \frac{S_L^2}{\alpha}$ suggesting a direct link to chemistry/transport at the flamelet level

Effect of NRPD (plasmas) on dynamic stability:

- Plasmas can suppress instabilities by up to 25 dB across a wide range of conditions
- Electrode design/arrangement is important here – we found success with a pin-to-ring discharge

Effect of NRPD on NO_x

- For CH_4 /air flames, plasmas increase NO_x – at least at the levels required to suppress combustion oscillations
- For CH_4/NH_3 /air flames, plasmas reduce NO_x



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Questions?

Feel free to email us: santosh1@mit.edu

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- Office of Naval Research (ONR), award number N00014-21-1-2571
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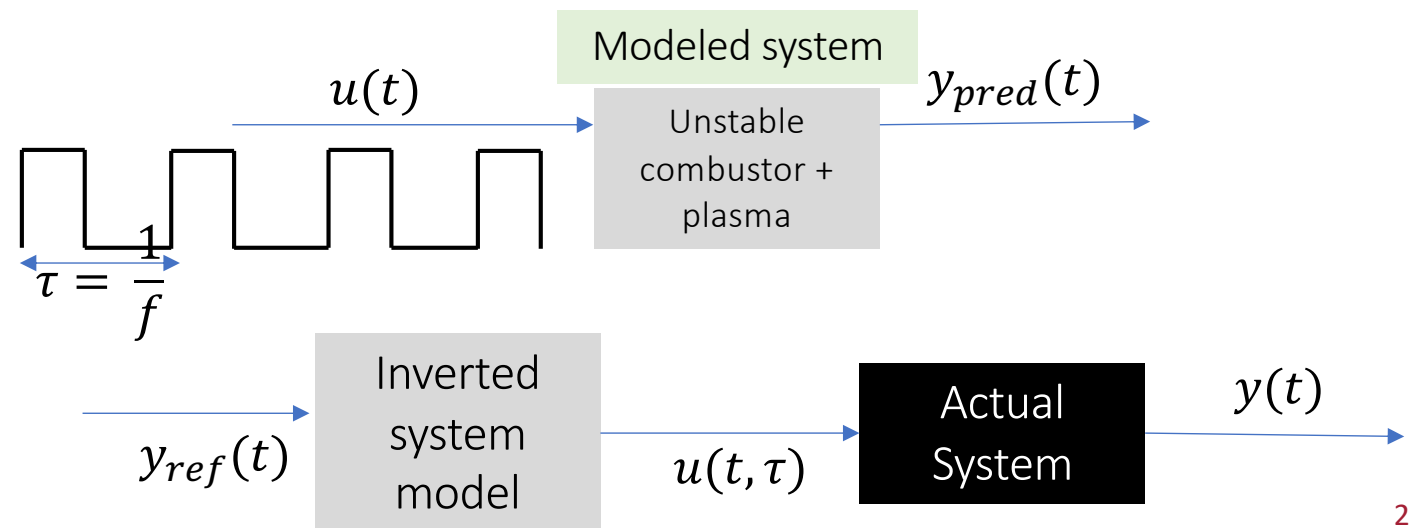
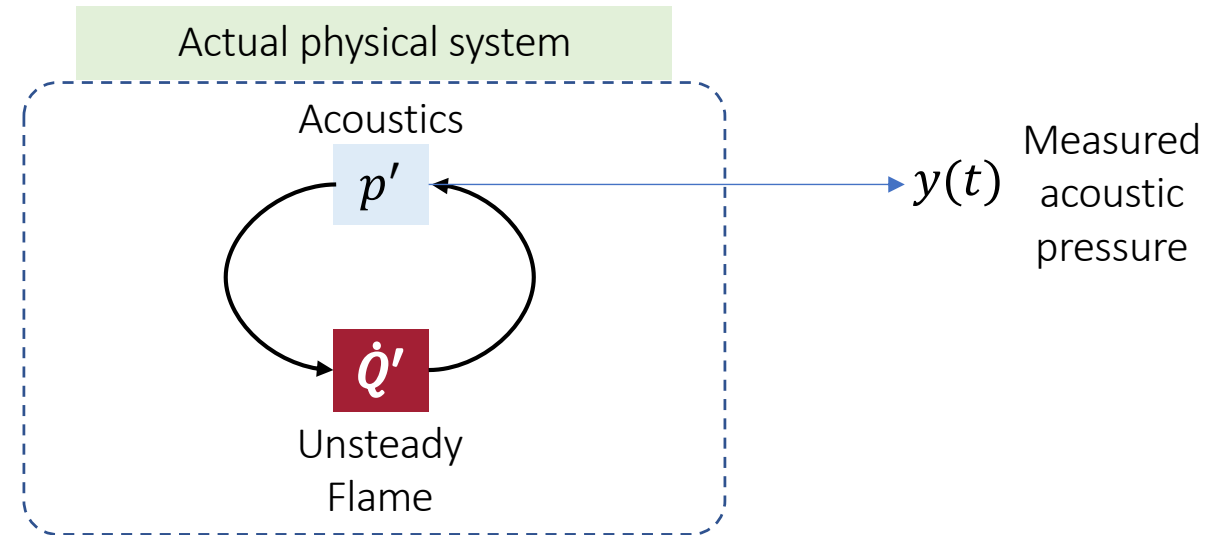
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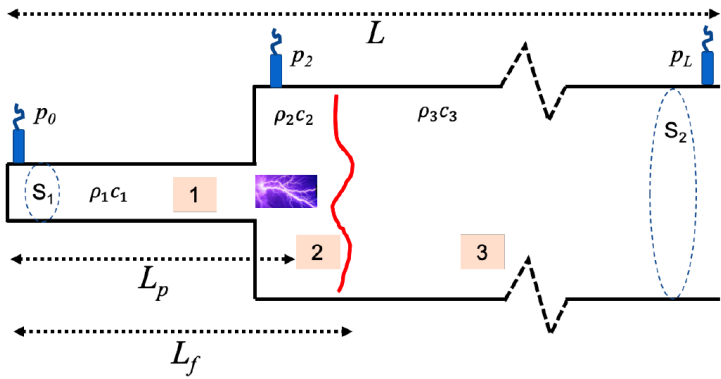
³Specter Aerospace, Peabody, MA, USA

Modeling for active control

- Feedback models and some feedforward models need a reduced order model of the system for choosing the right controller
 - We've combined the thermoacoustic system into a single model
- The plasma is actually triggered with a frequency f – this is the input variable
 - $1 < f \text{ (kHz)} < 9$
 - We set the voltage to the highest value, say 20 kV, which sets the energy per pulse



Swirl Combustor Simplified



Canonical state-space form

$$\dot{x} = Ax + Bu$$

$$y = Cx + Du$$

Start with conservation equations

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho u}{\partial x} = 0$$

$$\rho \frac{\partial u}{\partial t} + \rho u \frac{\partial \rho u}{\partial x} = -\frac{\partial p}{\partial x}$$

$$\rho c_p \frac{\partial T}{\partial t} + \rho c_p u \frac{\partial T}{\partial x} = \frac{\partial p}{\partial t} + u \frac{\partial p}{\partial x} + \dot{E}$$

Split terms into mean + fluctuations

$$p(x, t) = \bar{p} + p'(x, t)$$

$$u(x, t) = \bar{u} + u'(x, t)$$

$$\rho(x, t) = \bar{\rho} + \rho'(x, t)$$

$$\dot{E}(x, t) = \bar{\dot{E}} + \dot{E}'(x, t)$$

Point source assumption

$$\dot{E}'(x, t) = \dot{E}'_{flux}(t) \delta(x - x_p)$$

Wave equation

$$\frac{\partial^2 p'}{\partial t^2} - c^2 \frac{\partial^2 p'}{\partial x^2} = (\gamma - 1) \frac{\partial \dot{E}'}{\partial t}$$

$$p'(x, t) = \sum_{j=1}^N \psi_j(x) \eta_j(t)$$

Galerkin expansion

$$\begin{Bmatrix} \dot{\eta}_j \\ \ddot{\eta}_j \end{Bmatrix} = \begin{bmatrix} 0 & 1 \\ -\omega_j & 0 \end{bmatrix} \begin{Bmatrix} \eta_j \\ \dot{\eta}_j \end{Bmatrix} + \begin{bmatrix} 0 \\ K \cdot \frac{(\gamma - 1)}{E} \psi_j^*(x) \end{bmatrix} (f_{max} - f)$$

$$y = [\psi_j(x_{transducer}) \quad 0] \begin{Bmatrix} \eta_j \\ \dot{\eta}_j \end{Bmatrix}$$

$$\ddot{\eta}_j(t) + \omega_j^2 \eta_j(t) = \frac{(\gamma - 1)}{E} \psi_j^*(x) \frac{\partial \dot{E}'_{flux}}{\partial t}$$

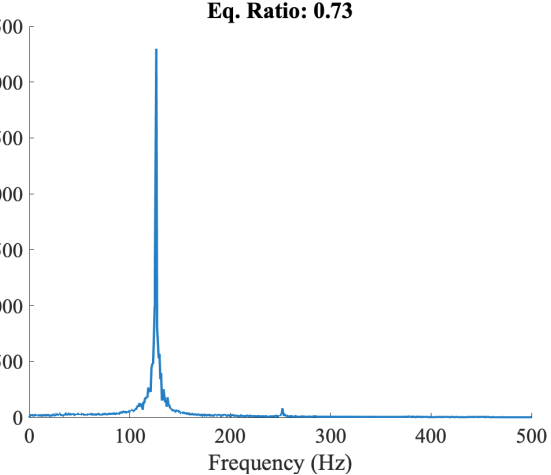
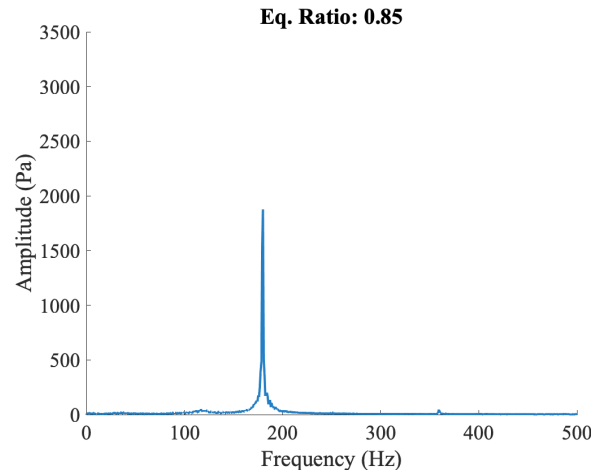
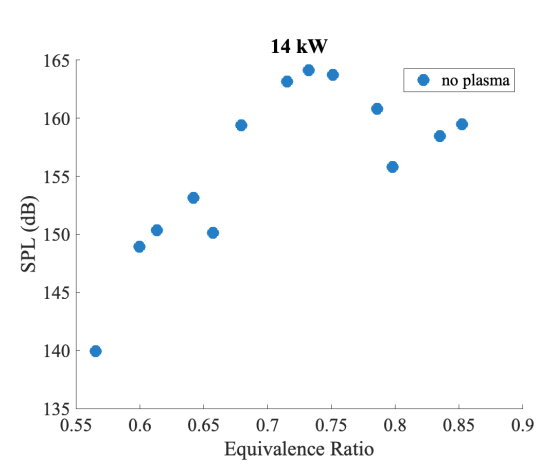
$$\frac{\partial \dot{E}'_{flux}}{\partial t} = K(f_{max} - f)$$

Model: Excitation term linearly decreases with plasma PRF

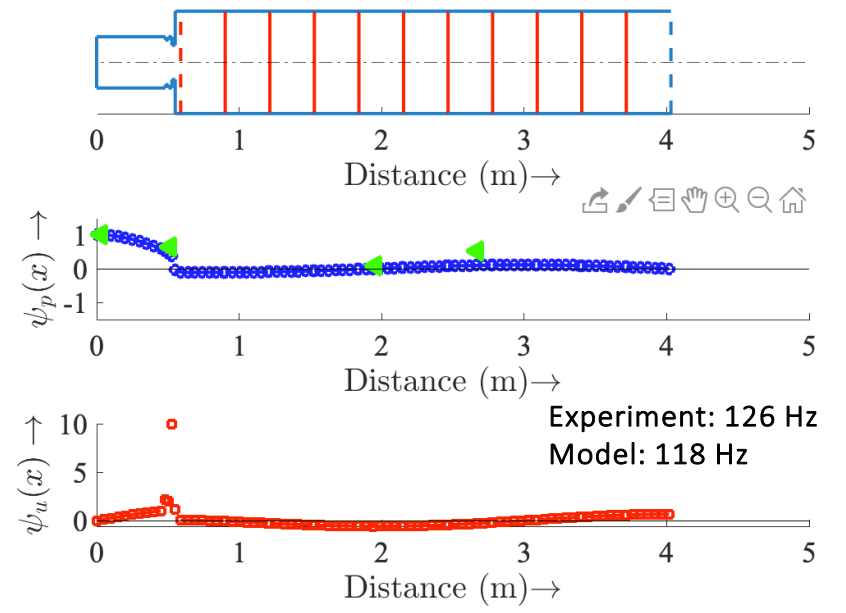
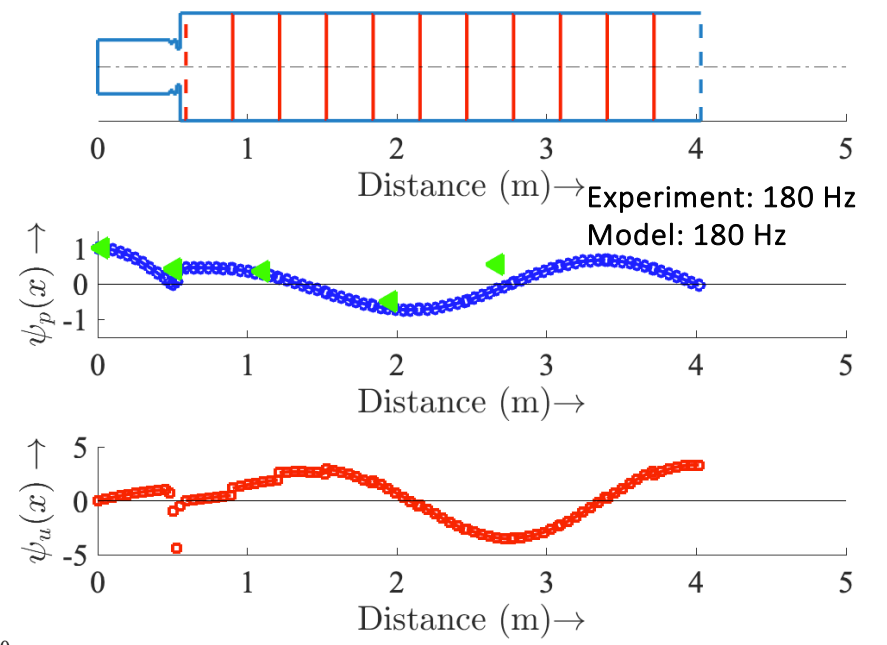
System model described in detail in the paper

Model Validation

Mode shapes: $\psi_j(x)$ & frequencies: ω_j



- The model requires temperature measurements and pressure measurements at different points in the combustor
- We tested this at two frequencies 130 and 180 Hz and in both cases, the model reasonably matched the data



Open-loop control

- In this experiment, the operator sat down and adjusted the knob between 1000Pa and 3000 Pa
- The model computes the PRF and sends it to the plasma system
- Not a perfect match
 - This will happen only when there is feedback

