Fundamental Mechanisms, Predictive Modeling, and Novel Aerospace Applications of Plasma Assisted Combustion

Overview of OSU research plan

Walter Lempert, Igor Adamovich, J. William Rich, and Jeffrey Sutton

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# Overview of OSU research plan

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Thrust 1. Experimental studies of nonequilibrium air-fuel plasma kinetics using advanced non-intrusive diagnostics

Task 1: Low-to-Moderate (T=300-800 K) temperature, spatial and time-dependent radical species concentration and temperature measurements in nanosecond pulse plasmas in a variety of fuel-air mixtures pressures (P=0.1 - 5 atm), and equivalence ratios (φ~0.1-3.0)

Goal: Generate an extensive set of experimental data on radical species concentrations and temperature rise; elucidate kinetic mechanisms of low-temperature plasma chemical fuel oxidation and ignition using kinetic modeling. Bridge the gap between room-temperature data (low-pressure gas discharges) and high-temperature data (shock tubes)
Test Bed #1: High-temperature, high-pressure nanosecond pulse discharge cell

High-pressure discharge cell inside a tube furnace (6 inch bore, up to $T=1200^\circ C$)
Premixed fuel-air flow (~1 m/s), preheated in the furnace, from 0.1 atm to a few atm
Repetitive nanosecond pulse discharge plasma: 20-40 kV, 5-25 nsec, 10 Hz to 100 kHz
Optical access (LIF, TALIF, CARS, CRDS) on the sides
Fuels: hydrogen, methane, ethylene, propane, pentane, methanol & ethanol vapor
Repetitive nanosecond pulse plasma for kinetic studies: Air, P=60 torr, ν=40 kHz, 40 msec burst, 1 μsec gate

- Some filamentary structure in pulses #1 and #2
- Uniform air plasma during subsequent pulses, at P=40-100 torr
Repetitive nanosecond pulse plasma for kinetic studies:
Ethylene-air, $P=40$ torr, $\varphi=1$, $\nu=40$ kHz

• Nearly uniform plasma during entire burst (except pulses #1 and #2)

• Ignition does not occur, likely due to rapid wall cooling

• Pressure is low – can this experiment be done at higher pressures?
Repetitive nanosecond pulse plasma for kinetic studies: Ethylene-air, $P=60$ torr, $\varphi=1$, $\nu=40$ kHz

- Uniform plasma during first few tens of pulses (except pulses #1 and #2)
- Well-defined filaments form in pulse #100, persist for several hundred pulses
- After ignition occurs, flame fills entire discharge volume, and plasma becomes uniform again
- Filamentation likely due to ionization / heating instability

- This is unacceptable: need to keep the plasma uniform during entire burst
- We know that preheating will improve plasma uniformity
- Sustaining plasma in a heated cell will allow measurements at higher pressures
Previous results: O atoms in air, methane-air, and ethylene-air at P=60 torr (single-pulse and burst mode, initially at T=300 K)

Objective: measure time-resolved O and H atoms in nsec pulse discharge plasmas in H$_2$-air and C$_x$H$_y$ air mixtures, at P ~ 0.1 - 1 atm, T=300-800 K

Outcome: kinetic mechanism of low-temperature plasma fuel dissociation and oxidation (specifically rates of O atom generation in the plasma and O atom reactions with fuel species)
Time-resolved species concentrations: OH (LIF with Hencken adiabatic burner calibration)

Work currently underway: OH in methane-air and ethylene-air at P=60 torr (single-pulse and burst mode, initially at T=300 K)

Objective: measure time-resolved OH in nsec pulse discharge plasmas in H₂-air and CₓHᵧ air mixtures, at P ~ 0.1 - 1 atm, T=300-800 K

Outcome: kinetic mechanism of low-temperature plasma fuel oxidation (specifically rates of H atom abstraction from fuel species)
Time-resolved species concentrations: NO
(LIF with calibration using known NO-N2 mixture)

Previous results: NO in air, methane-air and ethylene-air at P=60 torr (single-pulse, initially at T=300 K). State-of-the-art kinetic models cannot explain time-resolved data. Possible effect of N2(X,v) + O reaction.

Objective: measure time-resolved NO in nsec pulse discharge plasmas in H2-air and CxHy air mixtures, at P ~ 0.1 - 1 atm, T=300-800 K

Outcome: kinetic mechanism of low-temperature plasma fuel oxidation (specifically O2 dissociation vs. NO formation in N2* reactions)
Time-resolved, spatially resolved temperature
(purely rotational CARS)

Previous results: time-resolved temperature in air and ethylene-air at P=40 torr (burst mode, initially at T=300 K). Evidence of significant additional heat release in fuel-air, compared to air.

Objective: measure temperature in nsec pulse discharge plasmas in H_2-air and C_xH_y air mixtures, at P ~ 0.1 - 1 atm, T=300-800 K.

Outcome: kinetic mechanism of low-temperature plasma chemical energy release in exothermic fuel oxidation reactions with radicals.
Test Bed #2: Flat flame McKenna burner with nanosecond pulse discharge

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Flame

HV electrode 5-15mm ~1-5 mm

Wire mesh HVE

Burner surface

McKenna burner

Flat flame burner inside a six-arm cross vacuum chamber (8 inch bore)
Premixed fuel-air flow (~0.1-1.0 m/s) with N₂ co-flow, P=10-40 torr
Repetitive nanosecond pulse discharge plasma: 20-40 kV, 5-25 nsec, 10 Hz to 100 kHz
Optical access (LIF, TALIF, CRDS) on two perpendicular axes
Fuels: hydrogen, methane, ethylene, propane, pentane, methanol & ethanol vapor
Steady, laminar, low-pressure flat flames allow spatially-resolved measurements of temperature and species concentrations.

Minimize transport influence; isolate kinetic effects.

Can investigate full range of temperature conditions (from below 500 K to 2000 K) by adjusting measurement position (i.e. height above burner).

Typical spatial scale ~5-20 mm, spatial resolution <200 µm.

Straightforward integration of nsec discharge plasma into a low-pressure flame facility and study of plasma effects (i.e. measurements with plasma “off” and “on”).
Previous low-pressure flame results (LIF): P=10-40 torr; CH$_4$, C$_2$H$_6$, C$_3$H$_8$, C$_4$H$_{10}$; φ=0.6 -1.4

Flame temperature from rotational structure of OH A-X (1,0) band near 282 nm

Spectral features used for profiles of flame species:
- **CH** A-X (0,0) at 435 nm
- **NO** A-X (0,0) at 226 nm
Previous low-pressure flame results (LIF):

\( P = 10 - 40 \) torr; \( CH_4, C_2H_6, C_3H_8, C_4H_{10}; \varphi = 0.6 - 1.4 \)

Spatially-resolved measurements of radicals to understand high-temperature flame chemistry, help kinetic model development

Kinetic modeling: GRI-Mech 3.0

We will look at the region upstream of the flame where coupling between plasma kinetics and flame chemistry is most important
Objective: Examine coupling of plasma and combustion kinetics in a 1-D low-pressure flame. Use spatially-resolved species concentration and temperature measurements by LIF (OH, H, O, and CH) and CRDS (HO₂, HCO, CH₃) to study the effect of quasi-steady (RF) and repetitively pulsed nsec discharge plasmas on low-temperature chemistry and coupling with the flame zone.

Outcome: Kinetic mechanism of low-temperature plasma chemical fuel oxidation and energy release, and its effect on flame speed and burn rate. Specifically, boundary between “low-T” and “high-T” chemistry by measuring HO₂ radical concentration, at the conditions when O₂ is electronically excited:

\[ \text{O}_2 + \text{H} \rightarrow \text{OH} + \text{O} \quad \text{(high temperatures)} \]
\[ \text{O}_2 + \text{H} + \text{M} \rightarrow \text{HO}_2 + \text{M} \quad \text{(low temperatures)} \]

CRDS diagnostics will be used in both “test bed” experiments, (I) high-T, high-P nsec discharge plasma cell, and (II) low-P flame / plasma cell.
Task 8: Development and validation of a predictive kinetic model of non-equilibrium plasma fuel oxidation and ignition, using experimental results of Thrust 1

Goal: Identify key mechanisms, reaction, and rates of plasma chemical fuel oxidation processes for a wide range of fuels, pressures, temperatures, and equivalence ratios. This is absolutely essential to predictive capability of the model.
Air plasma model: equations for ground state species (**N**, **N**<sub>2</sub>, **O**, **O**<sub>2</sub>, **O**<sub>3</sub>, **NO**, **NO**<sub>2</sub>, **N**<sub>2</sub>**O**), charged species (electrons and ions), and excited species (**N**<sub>2</sub>(**A**<sup>3Σ</sup>), **N**<sub>2</sub>(**B**<sup>3Π</sup>), **N**<sub>2</sub>(**C**<sup>3Π</sup>), **N**<sub>2</sub>(**a**<sup>1Σ</sup>), **O**<sub>2</sub>(**a**<sup>1Δ</sup>), **O**<sub>2</sub>(**b**<sup>1Σ</sup>), **O**<sub>2</sub>(**c**<sup>1Σ</sup>), **N**(2**D**), **N**(2**P**), **O**(1**D**)) produced in the plasma.

Two-term expansion Boltzmann equation for plasma electrons

Fuel-air plasma: model combined with GRI Mech 3.0 **C**<sub>x</sub>**H**<sub>y</sub> oxidation mechanisms, supplemented with fuel dissociation by electron impact and in reactions with electronically excited nitrogen

Peak E/N adjusted for pulse energy to be same as predicted by the nanosecond pulse discharge model

We have absolutely no reason to trust the model predictions: GRI Mech 3.0 (or any other combustion mechanism) is not designed to work at low temperatures (starting at **T**=300 K)

Confidence in the model can be provided **only by detailed kinetic measurements** such as discussed in Thrust 1 plan
Here is what we know so far: dominant radical and energy release processes in \( \text{C}_2\text{H}_4 \)-air predicted by the model

**O atom generation**

\[
\begin{align*}
\text{N}_2 + e^- & = \text{N}_2(\text{A}^3\Sigma) + e^- \\
\text{N}_2 + e^- & = \text{N}_2(\text{B}^3\Pi) + e^- \\
\text{N}_2 + e^- & = \text{N}_2(\text{C}^3\Pi) + e^- \\
\text{N}_2 + e^- & = \text{N}_2(\text{a}^1\Sigma) + e^- \\
\text{O}_2 + e^- & = \text{O}(^3\text{P}) + \text{O}(^3\text{P,1D}) + e^- \\
\text{N}_2(\text{C}^3\Pi) + \text{O}_2 & = \text{N}_2(\text{a}^1\Sigma) + \text{O}_2 \\
\text{N}_2(\text{a}^1\Sigma) + \text{O}_2 & = \text{N}_2(\text{B}^3\Pi) + \text{O}_2 \\
\text{N}_2(\text{B}^3\Pi) + \text{O}_2 & = \text{N}_2(\text{A}^3\Sigma) + \text{O}_2 \\
\text{N}_2(\text{A}^3\Sigma) + \text{O}_2 & = \text{N}_2 + \text{O} + \text{O}
\end{align*}
\]

**Fuel dissociation**

\[
\begin{align*}
\text{C}_2\text{H}_4 + e^- & = \text{products} + e^- \\
\text{N}_2(\text{A}^3\Sigma) + \text{C}_2\text{H}_4 & = \text{N}_2 + \text{C}_2\text{H}_3 + \text{H} \\
\text{N}_2(\text{B}^3\Pi) + \text{C}_2\text{H}_4 & = \text{N}_2 + \text{C}_2\text{H}_3 + \text{H} \\
\text{N}_2(\text{C}^3\Pi) + \text{C}_2\text{H}_4 & = \text{N}_2 + \text{C}_2\text{H}_3 + \text{H} \\
\text{N}_2(\text{a}^1\Sigma) + \text{C}_2\text{H}_4 & = \text{N}_2 + \text{C}_2\text{H}_3 + \text{H}
\end{align*}
\]

**O atom decay**

\[
\begin{align*}
\text{O} + \text{C}_2\text{H}_4 & = \text{CH}_3 + \text{HCO} \\
\text{O} + \text{C}_2\text{H}_4 & = \text{H} + \text{CH}_2\text{CHO} \\
\text{C}_2\text{H}_3 + \text{O}_2 & = \text{HCO} + \text{CH}_2\text{O} \\
\text{C}_2\text{H}_3 + \text{O}_2 & = \text{O} + \text{CH}_2\text{CHO} \\
\text{O} + \text{O}_2 + \text{M} & = \text{O}_3 + \text{M} \\
\text{O} + \text{O}_3 & = \text{O}_2 + \text{O}_2
\end{align*}
\]
Model validation summary: so far so good…

... but no surprise if the model fails at some point

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Need a lot more data from Thrust 1 for extensive model validation

Outcome: a self-consistent low-temperature fuel-air plasma chemical mechanism
Task 10: Characterization and modeling of nsec pulse discharges

Goal: Prediction of E/N and electron density in the plasma, individual pulse energy coupled to the plasma, and their scaling with pressure, temperature, pulse waveform, and mixture composition
Two-pronged approach to plasma assisted ignition modeling

Predictive modeling of energy release rate and ignition delay time in low-temperature, repetitive nanosecond pulse fuel-air plasmas requires:

- E/N in the plasma, individual pulse energy coupled to the plasma, and their scaling with pressure, temperature, pulse waveform, and mixture composition

- Air plasma and fuel-air plasma chemistry: reactions among ground state species, excited species and radicals generated in the plasma, and their effect on energy release rate

These two problems require separate analysis:

- Nsec pulse plasma / sheath models cannot incorporate detailed reactive plasma chemistry: too many species (~100) and reactions (~1,000)

- Detailed plasma chemistry models (quasi-neutral) cannot incorporate repetitive, nsec time scale sheath dynamics and plasma shielding

Approach:

- Predict plasma E/N and coupled pulse energy using nsec pulse plasma / sheath model
- Incorporate results into fuel-air plasma chemistry model
Previous results:
Repetitive nsec discharge pulse energy measurements

Nitrogen, P=300 torr, v=100 kHz

Nitrogen, P=350 torr, v=100 kHz
0.3 seconds after start (pulse # 30,000)

Pulse energy 11 mJ/pulse
Discharge power 110 W

What are the electric field and the electron density?
Previous results:
Analytic nsec pulse discharge plasma / sheath model

- Equations for electron and ion number density
- Poisson equation for the electric field
- Plane-to-plane discharge geometry
- Voltage pulse: Gaussian fit to experimental waveform
- Dielectric plate charging / plasma shielding

Analytic solution: time-dependent electron density and electric field in the plasma, coupled pulse energy
Excellent agreement with numerical solution, experimental data
Previous results:

Analytic nsec pulse discharge plasma / sheath model

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**Power density, kW/cm³**

- **Numerical model**
- **Analytic solution**

**Breakdown**

**Shielded plasma**

**Coupled pulse energy, mJ**

- **Value inferred from TALIF O atom measurements**

- **P*=100 torr**
- **P*=80 torr**
- **P*=60 torr**
- **P*=40 torr**

\[
Q_{\text{total}} = Q_{\text{break}} + Q_{\text{after}} \approx \frac{1}{2} C_{\text{load}} V_{\text{peak}}^2 \left( \frac{V_0}{V_{\text{peak}}} \right)^2 + \frac{\sqrt{2\pi}}{V_{\text{RC}} T_{\text{pulse}}} \]

- **Coupled pulse energy scales with the number density, can be increased by increasing peak voltage, reducing pulse duration**

- **Excellent agreement with numerical solution, experimental data**
Objective: measure time- and space-resolved electric field and electron density in nsec pulse discharge plasmas using psec CARS and Thomson scattering; comparison with the model

Outcome: predictive capability for electron impact kinetic processes in the plasma
Thrust 4. Studies of diffusion and transport of active species in representative 2-D reacting flow geometries

Task 12: Ignition and flameholding in nonequilibrium plasma cavity flows at low static temperatures

Goal: Determine viable approaches to flameholding in high-speed flows using low-temperature plasmas. We simply cannot process the entire flow with the plasma!
Previous results: cavity ignition in premixed ethylene-air flows by nsec plasma (25 kV, 20 nsec, 40 kHz)

Fuel-air, 200 torr, 50 m/s

Air, 200 torr, 50 m/s

Fuel-air, 150 torr, 25 m/s

Air, 150 torr, 25 m/s

Diffuse plasma in air, filamentation in fuel-air during ignition, diffuse plasma after ignition
Previous results: cavity ignition and flameholding in premixed and non-premixed ethylene-air flows by nsec plasma

Ignition and stable flameholding in both premixed and non-premixed flows up to 100 m/sec (global $\phi=1$ in both cases)

- 80-90% burned fuel fraction
- Plasma power $\sim$100 W, combustion energy release 35 kW

After ignition, plasma needs to be “on” at all times (flame blow-off without plasma)
Objectives:

- Further studies of cavity ignition and flameholding by repetitive nsec pulse plasmas in fuel injection flows (hydrogen and hydrocarbons)
- High frame rate (10-20 kHz) NO and OH PLIF imaging of ignition process using burst mode laser
- Increasing flow velocity beyond 100 m/sec, operating at low global equivalence ratios ($\phi=0.1-0.2$)
- Comparison with kinetic modeling calculations using reduced plasma chemical ignition mechanism. Plasma flameholding mechanism after ignition – thermal or not?

Outcome: Demonstration of true predictive capability of the model