Energy conversion in transient molecular plasmas:

Implications for plasma flow control and plasma assisted combustion

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Outline

I. Motivation: critical importance of energy transfer processes in nonequilibrium, high-pressure molecular plasmas

II. Electric field: discharge energy loading and partition

III. Electron density and electron temperature: discharge energy loading and partition

IV. Dynamics of temperature rise: “rapid” heating and “slow” heating

V. Air and fuel-air plasma chemistry: kinetics of plasma assisted combustion

VI. Air plasma kinetics and plasma flow control

VII. Summary and future outlook
I. Motivation
Energy Partition in Air Plasma vs. Electric Field

- Reduced electric field, $E/N$, controls input energy partition in the discharge
- Rates of electron impact processes: strongly (exponentially) dependent on $E/N$


(1) $O_2$ vibrational excitation:
Non-self-sustained discharges, very low $E/N$

(4) $N_2$ vibrational excitation:
Quasi-steady-state discharges, low $E/N$

(5,6) electronic excitation, dissociation,
(7) ionization:
Pulsed discharges, high $E/N$
Energy conversion in molecular plasmas: here is what we know

• Energy is coupled to electrons and ions by applied electric field

• Electric field in the plasma: controlled by electron and ion transport, and by surface charge accumulation

• Energy partition (vibrational and electronic excitation, dissociation, ionization): controlled by electron density and electric field (or electron temperature)

• Temperature rise in discharge afterglow: controlled by quenching of excited electronic states, vibrational relaxation

• Plasma chemical reactions, rates of radical species generation: controlled by populations of excited electronic states, e.g. $N_2^*$, excited vibrational states, e.g. $N_2(v)$

• Time-resolved measurements of $\vec{E}$, $n_e$, $T_e$, $N_2^*$, $N_2(v)$, and radical species ($O$, $H$, $OH$, $NO$, $CH$, $HO_2$, $CH_2O$): stable, reproducible, high-pressure ns pulse discharges

• Objective: quantitative insight into energy conversion mechanisms critical for plasma-assisted combustion and plasma flow control
II. Electric field in transient plasmas: 
insight into discharge energy loading and partition

Diagnostics: CARS-like 4-wave mixing
2-D Ns Pulse Discharge in Atmospheric Air

- Discharge sustained between a high-voltage electrode (razor blade) and grounded copper foil, covered with quartz plate 120 μm thick
- Discharge gap 600 μm
- Simple two-dimensional geometry, diffuse plasma
- Peak voltage 7.5 kV, peak current 7 A, coupled energy 2 mJ
- Two current peaks of opposite polarity: “forward” and “reverse” breakdowns
- Time-resolved electric field measured at several locations in the plane of symmetry
- Electric field distribution along the surface is also measured
“Curtain Plasma” Images, Negative Polarity Pulse

- Front view: near-diffuse plasma “curtain”
- Diffuse surface ionization wave detected, straight ionization front
- Wave speed ~ 0.03 mm/ns
- Surface plasma layer thickness ~150 μm
Time-resolved and Spatially-Resolved Electric Field Measurements

- **Initial field offset**: charge accumulation on dielectric surface from previous pulse
- **Field follows applied voltage rise**, increases until “forward breakdown”
- **After breakdown**, field reduced due to charge accumulation on dielectric surface
- **Field is reversed after applied voltage starts decreasing**
- **After discharge pulse**, field decays over several μs: surface charge neutralization by charges from plasma

- Field distribution measured at the moment when field reversal occurs near HV electrode (t=70 ns)
- “Snapshot” electric field distribution across the surface ionization wave front
III. Electron density and electron temperature in transient plasmas: insight into discharge energy loading and partition

Diagnostics: Thomson scattering
Filtered Thomson Scattering: $n_e$, $T_e$, and EEDF inference

- Electron density: area under Thomson scattering spectrum
- Electron temperature: spectral linewidth
- Raman scattering rotational transitions in N$_2$ used for absolute calibration
- Gaussian Thomson scattering lineshape: Maxwellian EEDF
Thomson Scattering Spectra

Sphere-to-sphere ns pulse discharge in H$_2$-He and O$_2$-He

5% H$_2$-He, P=100 torr, t=100 ns
ne = 1.5$\cdot$10$^{14}$ cm$^{-3}$, Te = 2.0 eV

10% O$_2$-He, P=100 torr, t=100 ns
ne = 1.7$\cdot$10$^{13}$ cm$^{-3}$, Te = 1.6 eV, T=350 K
Electron Density and Electron Temperature
Sphere-to-sphere ns pulse discharge in O$_2$-He

- $n_e = 10^{13} - 3 \cdot 10^{14}$ cm$^{-3}$, $T_e = 0.3 - 5.5$ eV (0-10% O$_2$)

- “Double maxima” in $n_e$, $T_e$: two discharge pulses
  $\approx$400 ns apart

- Electron temperature in the afterglow $T_e \approx 0.3$ eV
  Superelastic collisions prevent electron cooling

- Modeling predictions in good agreement with data
IV. Dynamics of temperature rise in transient plasmas: “rapid” heating and “slow” heating

Diagnostics: vibrational and pure rotational ps CARS
Sphere-to-sphere ns pulse discharge in air, P=100 Torr
Discharge pulse waveforms and CARS spectra

Pulse energy coupled to plasma $\sim 0.5$ eV

$N_2(v=0-9)$ bands during and after discharge pulse
$N_2(v=0)$ band (without plasma)
Temperature rise in ns pulse discharge and afterglow: air vs. nitrogen

Air, P=100 Torr

- Compression waves formed by “rapid” heating, on sub-acoustic time scale, $\tau_{acoustic} \sim r / a \sim 2 \mu s$
- Strong effect on high-speed flows
- Strong vibrational excitation in the discharge, $N_2(v=0-8)$
- $T_v(N_2)$ rise in early afterglow: V-V exchange, $N_2(v) + N_2(v=0) \rightarrow N_2(v-1) + N_2(v=1)$
- $T_v(N_2)$ decay in late afterglow: V-T relaxation, $N_2(v) + O \rightarrow N_2(v-1) + O$, radial diffusion
- “Rapid” heating: quenching of $N_2$ electronic states, $N_2(C,B,A,a) + O_2 \rightarrow N_2(X) + O + O$
- “Slow” heating: V-T relaxation, $N_2(X,v) + O \rightarrow N_2(X,v-1) + O$
- “Rapid” heating: pressure overshoot on centerline, compression waves detected in experiments
- NO formation: dominated by reactions of $N_2$ electronic states, $N_2^* + O \rightarrow NO + N$, also in good agreement with [NO], [N] measurements

Comparison with modeling predictions in air: vibrational kinetics and temperature rise
Adding CO$_2$ (rapid V-T relaxer) to air: accelerating energy thermalization rate

Rapid N$_2$ relaxation and temperature rise. Mechanism of accelerated heating:

- V-V energy exchange between N$_2$ and CO$_2$(v$_3$) mode: N$_2$(v=1) + CO$_2$(000) $\leftrightarrow$ N$_2$(v=0) + CO$_2$(001)
- CO$_2$ energy re-distribution among vibrational modes: CO$_2$(001) + M $\leftrightarrow$ CO$_2$(100,020,010) + M
- V-T relaxation of bending mode: CO$_2$(010) + M $\rightarrow$ CO$_2$(100) + M
- Strong effect on nonequilibrium compressible flows
V. Fuel-air chemistry in transient plasmas: kinetics of plasma assisted combustion

Diagnostics: Rayleigh scattering, LIF, CARS
Plasma chemical reactions and transport: ns pulse discharge in $\text{H}_2 - \text{O}_2 - \text{Ar}$, $P=40$ torr

**Hot central region:**
OH production dominated by chain branching

\[ \text{H} + \text{O}_2 \rightarrow \text{OH} + \text{O} ; \quad \text{O} + \text{H}_2 \rightarrow \text{OH} + \text{H} \]

**Colder peripheral region:**
OH accumulation in HO$_2$ reactions
radial diffusion of H atoms;

\[ \text{H} + \text{O}_2 + \text{M} \rightarrow \text{HO}_2 ; \quad \text{H} + \text{HO}_2 \rightarrow \text{OH} + \text{OH} \]
[OH] kinetics in preheated fuel-air mixtures after ns pulse discharge burst

“Near 0-D” diffuse plasma, a burst of 50-100 pulses used to couple sufficient energy

Comparison with modeling predictions: plasma assisted combustion kinetic mechanism validation
Plasma chemical reactions reduce ignition temperature in H$_2$-air

- $\phi=0.4$, 120-pulse burst, $T_0=500$ K, $P=80-90$ torr
- Model predictions: good agreement with time-resolved temperature measurements
- Ignition temperature with plasma, $T_i \approx 700$ K, lower than autoignition temperature, $T_a \approx 900$ K
VI. Air plasma kinetics and plasma flow control
Every nanosecond discharge pulse produces a robust spanwise vortex. Enhanced mixing with free stream → boundary layer reattachment. Same effect detected up to $u=96$ m/sec ($M=0.28$, $Re_x \sim 1.5 \cdot 10^6$). Consistently outperform AC DBD actuators.
Localized Arc Plasma Flow Actuators (LAPFA): Exciting instabilities in transonic and supersonic flows (M=0.9-2.0)

- BN nozzle extensions, tungsten wire electrodes
- Circular nozzle, 1 inch diameter recently
- Multiple channels controlled by fast HV switches
- Independent control of frequency, phase, and duty cycle → excitation of different instability modes
LAPFA: Formation of coherent structures in a M=0.9 circular jet

- High amplitude perturbations (localized heating in arc filaments)
- Every discharge pulse results in vortex formation
- Flow responds to forcing near jet column instability frequency
**Plenum:** overlapped ns pulse / DC sustainer discharge for vibrational loading of $N_2$

- $P_0 = 300$ torr, $T_v=2000$, $T=500$ K, 2-D nozzle, top wall contoured, bottom wall plane
- Condition at nozzle exit: $M = 2.5$, $P_{exit} = 15$ torr
- Subsonic flow below expansion corner: injection of $N_2$ or $CO_2$
- Optical access for schlieren, CARS, and NO PLIF in subsonic and supersonic flows
Effect of vibrational relaxation of shear layer: 
$\text{N}_2 / \text{N}_2$ (left) vs. $\text{N}_2 / \text{CO}_2$ (right)

- Time delay between frames 5 ms, $t = 0-80$ ms
- $\text{N}_s$ pulse / DC discharge (2.3 kW) is turned on at $t = 10-45$ ms, to excite main $\text{N}_2$ flow
- No perturbation of shear layer detected in $\text{N}_2 / \text{N}_2$ flow
- In $\text{N}_2 / \text{CO}_2$ flow, shear layer expansion angle decreases, approaching $\theta = 0^\circ$
- No change observed if main $\text{N}_2$ flow is not excited
• Top flow: vibrationally excited N$_2$, $T_V=1900$ K, estimated $T_{rot}=240$ K

• Bottom flow: CO$_2$ bleeding through backstep, static pressure 7 torr

• CO$_2$ bleeding reduces $T_V(N_2)$, increases $T_{trans/rot}$ and static pressure

• Consistent with time-resolved measurements in ns pulse discharge in quiescent N$_2$-CO$_2$

• Static pressure increase pushes up shear / mixing layer
• Growing body of time-resolved, spatially-resolved data characterizing transient, high-pressure air and fuel-air plasmas

• Measurements of electric field, electron density, and electron temperature necessary for insight into discharge energy coupling and partition

• Measurements of temperature, N$_2$(v) populations, and excited electronic states of N$_2^*$ necessary for insight into temperature dynamics

• Measurements of N$_2^*$ and key radicals (O, H, OH, and NO) critical for quantifying their effect on fuel-air plasma chemistry

• Comparing measurement results with kinetic modeling predictions provides confidence in the models, assesses their predictive capability
Summary: nonequilibrium plasma flow control

- Surface and volumetric ns pulse discharges: energy thermalization on sub-
  acoustic time scale, high-amplitude compression wave generation

- Mechanism of energy thermalization (“rapid heating” and “slow heating”) is well understood

- NS-DBD surface plasma actuators: large-scale coherent flow structures; significant flow control authority in subsonic flows (up to \( M = 0.3 \)) at low actuator powers; scalable to large dimensions (~1 m)

- LAFPA actuators: large-scale coherent structures; excitation of flow instability modes; significant control authority in transonic and supersonic flows (\( M = 0.9-2.0 \)) at low actuator powers; scalable to large phased arrays

- Flow control by vibrational relaxation: injection of “rapid relaxer” species into nonequilibrium flow at desired location; temperature and pressure rise due to accelerated relaxation; strong effect in supersonic shear layer
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