Generation of Diffuse Reactive Oxygen Plasma in an Atmospheric Pressure Plasma Jet

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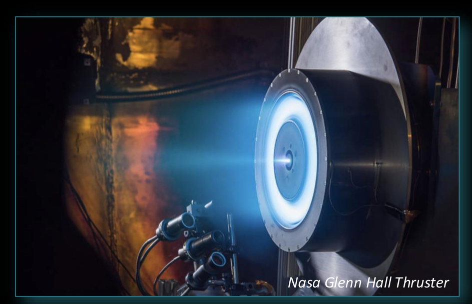
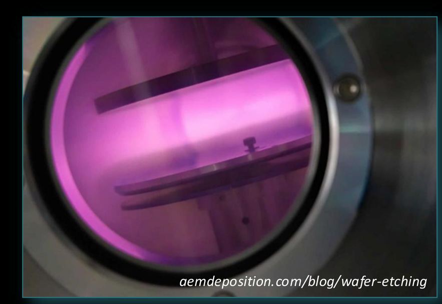
Background and Motivation

Low temperature plasma used in many applications and sectors:

- Industrial: Materials processing
- Aerospace: Thrusters
- Lighting: Fluorescent tube lights and plasma TVs
- Medical: Wound healing and dermal regeneration
- Water purification: Ozone generation

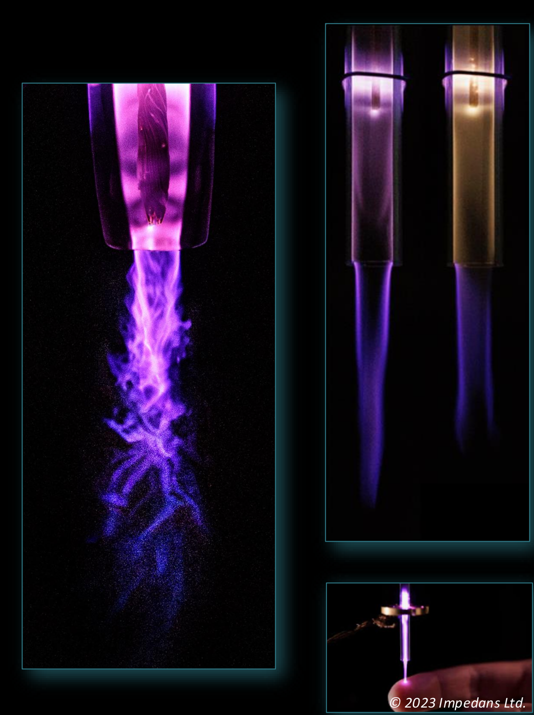
(Industrial, Aerospace, Lighting: Low pressure applications)





Plasma jets are used to create plasma at atmospheric pressure, which can simplify implementation and reduce energy cost

- Plasma jets can utilize a range of gases and power sources; choice depends on application
  - Commonly used gases are helium, argon, and air.
  - Can be generated with DC, RF, and AC power
- Challenges associated with plasmas at atmospheric pressure
  - Jets operate stably with helium gas (filament-free), which is expensive for practical applications
  - Operation with other gases (argon, nitrogen, air, oxygen) creates plasmas that are prone to filaments.
  - Filaments are self-organizing in nature. They are often hot and non-uniform; can cause damage to surfaces and variation in reactive species



Filament formation in plasma jets

100 ns snapshot of Argon filaments

- Avalanche → charge separation → localized E-field.
- Photons generated after previous ionization processes initiate additional avalanches and electrons, leading to a rapidly propagating ionization wave
- Gases with lower ionization potential are more prone to filamentation

Science question: *Is it possible to create a diffuse and reactive plasma at atmospheric pressure?*

New approach: Generation of highly reactive, diffuse atomic oxygen plasma at atmospheric pressure using Ar + trace O_2

Mechanisms for atomic oxygen production using argon gas with trace O_2

$Ar^+ + 2Ar \rightarrow Ar_2^+ + Ar$

$e + Ar_2^+ \rightarrow Ar^* + Ar$ (recombination)

Scenario 1

$Ar^* + O_2 \rightarrow 2O + Ar$

$e + O \rightarrow O(^1S) + e$ (electron impact)

$Ar^* + O \rightarrow ArO(^1S)$

$Ar_2^+ + O_2 \rightarrow O_2^+ + 2Ar$ (charge exchange)

Scenario 2 (low O_2 presence)

$e + O_2^+ \rightarrow O(^1S) + O$ (electron impact)

$O(^1S) \rightarrow O(^1D) + h\nu$ (557.7nm) (radiative loss)

$ArO(^1S) \rightarrow ArO(^1D) + h\nu$ (549.8 nm, 552.9 nm, 555.5 nm)

Acknowledgements

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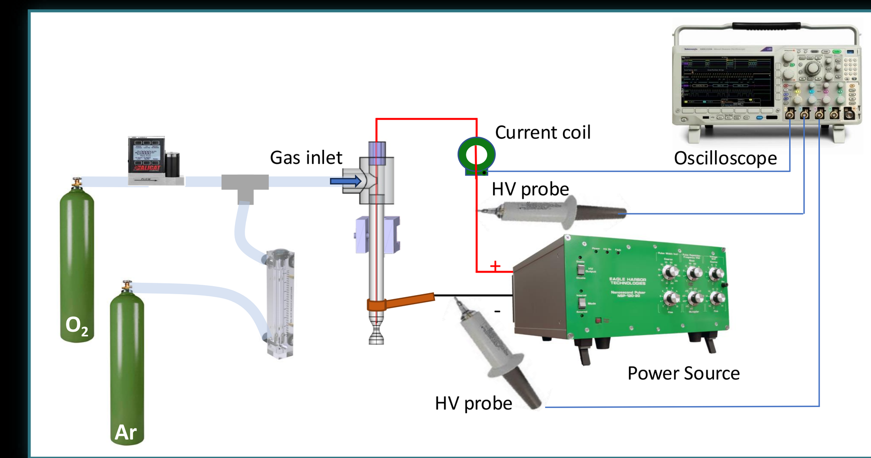
Experimental Setup

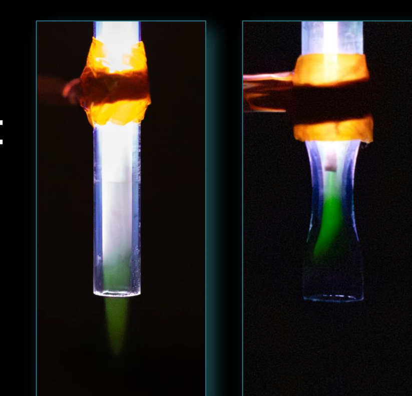
- **Power source:** EHT NSP 120-20F pulser capable of varying:
  - Voltage: 0-20 kV
  - ~ 4 kV required to make plasma
  - Frequency 0-10 kHz
  - Pulse width 0-260 ns
- **Gas flows:**
  - Rotameter: 0.7- 7 scfm Ar
  - MFC: 0-20 slpm O_2
- **Test plan:**
  - A parameter sweep of system variables, voltage, frequency, and pulse width
  - Optical emission spectroscopy for determination of species
  - Starch iodide tests to confirm reactivity
  - Two geometries studied: Cylindrical tube and cylindrical tube + CD nozzle



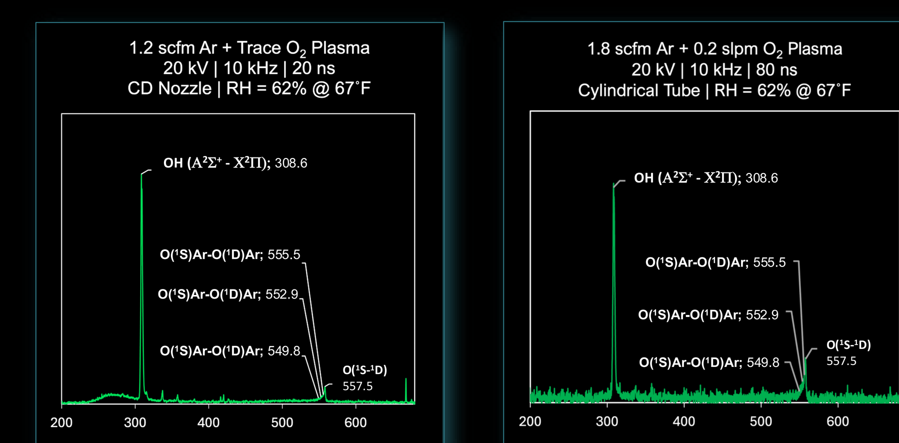
Experimental Results

Diffuse oxygen plasma was generated at atmospheric pressure using argon as carrier gas with two geometries: cylinder and cylinder + CD nozzle

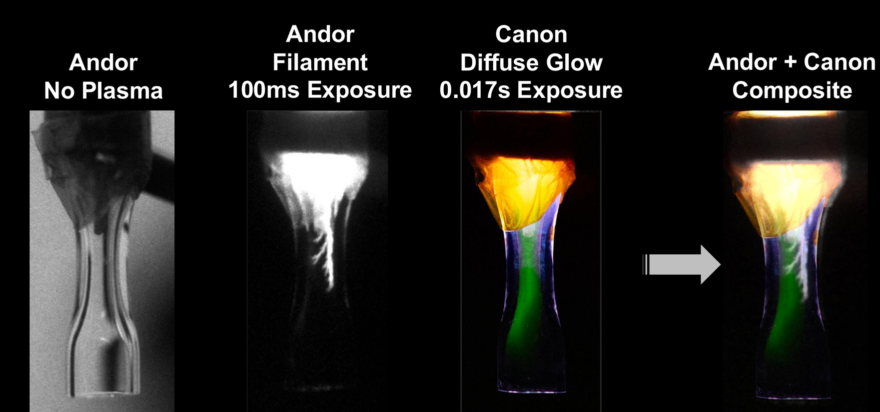
- Cylindrical tube requires higher gas flow and higher O_2, O plume exits tube; higher energy species reach substrate
- Tube + Nozzle generates brighter O discharge and requires less gas flow and O_2 in the feed gas, but the O plume does not exit the tube



Optical emission spectroscopy highlights the production of reactive species O and OH in both tube geometries



Verification of diffusivity condition



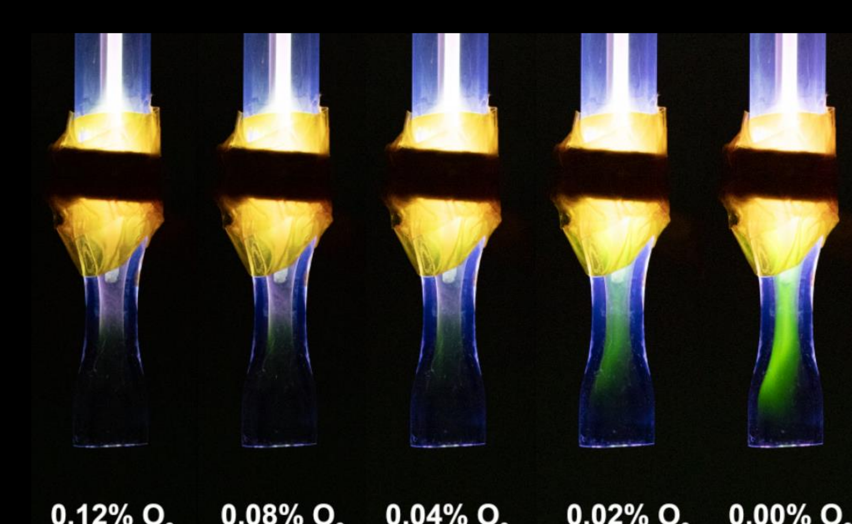
Argon plasma at 1.7 scfm | 20 kV | 10 kHz | 20 ns width | 0.0001 sec period

- High-speed camera image (Andor iStar) superimposed on traditional image (Canon R3) demonstrates a large portion of filament-free O plasma
- Green part of plume is diffuse because the $O(^1S)$ state is only 4.19 eV above ground level

Stable diffuse green plasma requires only trace O_2

- Better to flow Ar+O_2 and then turn off O_2
- Green plasma diminishes with O_2 flowing
- Once O_2 is shut off, green O plasma continues with only Ar gas flowing
- Trace O_2 sources:
  - Impurities in argon cylinder
  - Electronegative O_2 sticks to jet surfaces
- In presence of high O_2:

$Ar_2^+ + O_2 \rightarrow O_2^+ + Ar + Ar$
$O_2^+ + O_2 + Ar \rightarrow O_4^+ + Ar$
$O_4^+ \rightarrow O_2 + O_2$



Like Aurora Borealis, atomic O plasma in jet is an afterglow

Einstein coefficients, A_{21}:

- $A_{O(1S)}$ ~ 1.25 s^{-1}
- A_{Ar} ~ 10^6 - 10^7 s^{-1}
- $A_{ArO(1S)}$ ~ 576 · $A_{O(1S)}$ ~ 720 s^{-1}

$$\tau = \frac{1}{A_{21}}$$

Pulse period = 0.0001 s

Radiative lifetime τ:

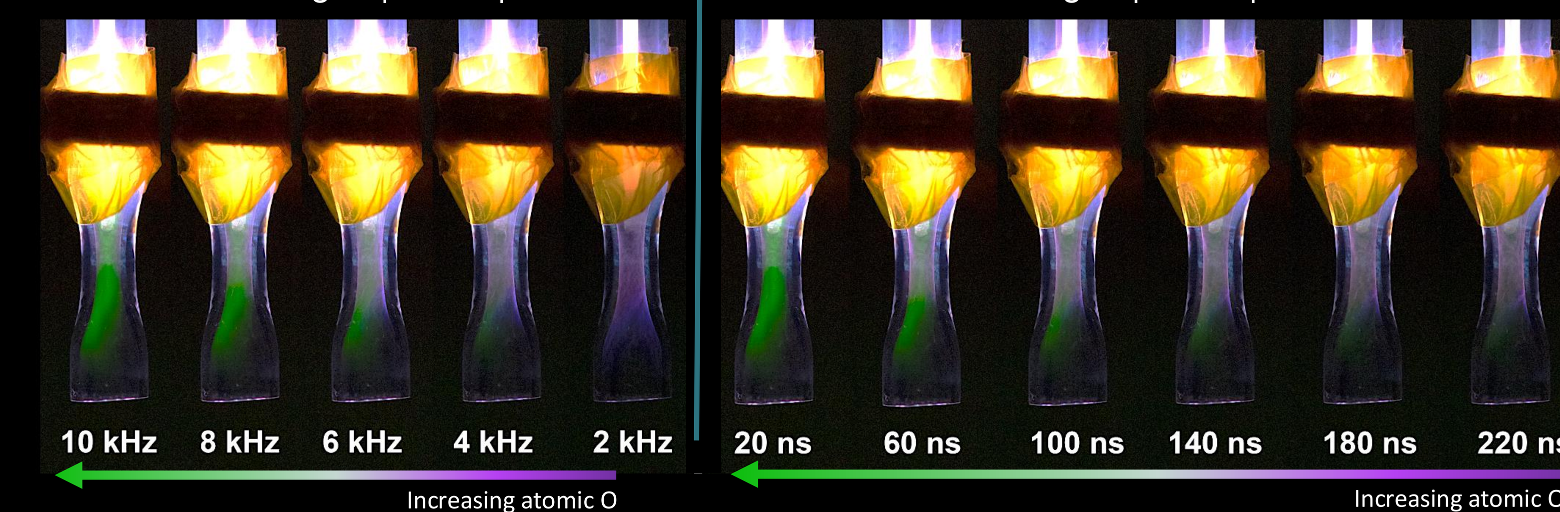
- $\tau_{O(1S)}$ ~ 0.75 s
- τ_{Ar} ~ 8 -16 ns
- $\tau_{ArO(1S)}$ ~ 1.4 ms

→ **Gas dynamics play significant role in plasma transport**

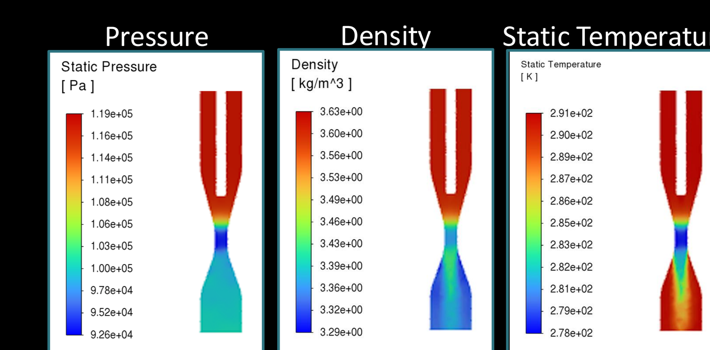
Key parameters leading to production of atomic O plasma

Pulse frequency effect: 1.7 scfm Argon | 18 kV | 20 ns

Pulse width effect: 1.7 scfm Argon | 18 kV | 10 kHz

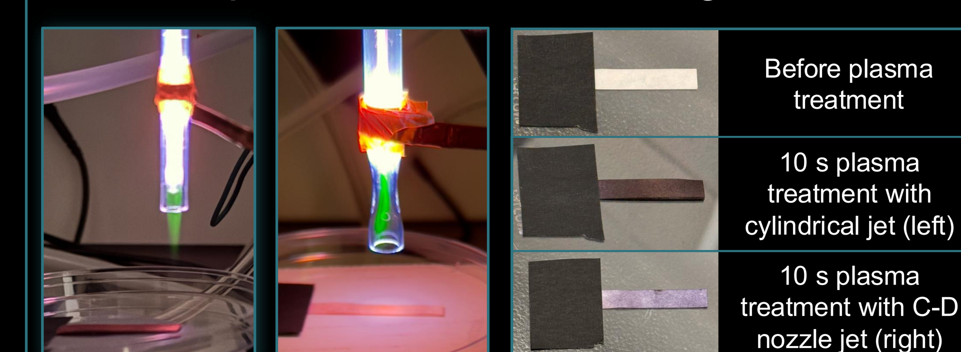


- **Pulse width**: Longer power deposition into the discharge fuels development of streamers, creating hotter discharge, which inhibits atomic O plasma processes
- **Pulse frequency:** Increased frequency of pulses for afterglow plasma leads to multiplicative effects



CFD simulations of C-D nozzle in Ansys Fluent demonstrate changes in discharge conditions (pressure, density) that are conducive to production of the atomic O plasma

Starch iodide tests confirm strong presence of reactive species after 10 seconds, when placed downstream of glow



Surface changes due to prolonged exposure to atomic O in space

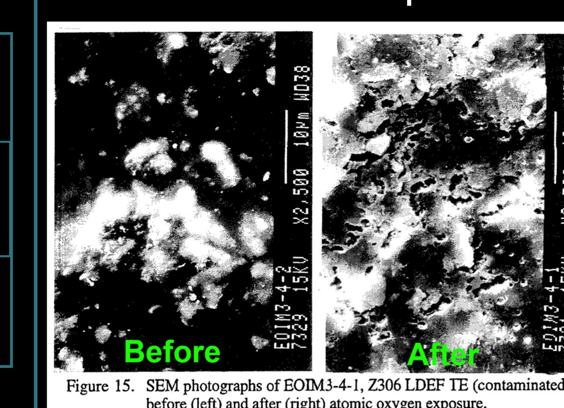


Figure 15. SEM photographs of EOIM-3-4-1, Z306 LDEF TE (contaminated), before (left) and after (right) atomic oxygen exposure.

Discussion / Conclusion

- We have generated a diffuse and reactive atomic oxygen plasma in an atmospheric pressure argon plasma jet with trace amounts of O_2
  - $O(^1S)$ state requires low-energy electrons; does not meet Meek's condition for filament formation
    → Keeping the pulse width low keeps electrons from gaining too much energy
  - Production of $O(^1S)$ requires trace concentrations of O_2 due to quenching
  - Metastable $O(^1S)$ has a lifetime of approximately 0.75 s and $ArO(^1S)$ excimers have a lifetime of approximately 1.4 ms; pulse width = 0.0001 s
    → Green plasma is an afterglow in which gas dynamic effects also impact transport

Future Work

- Determination of reactive species flux in filamentless state using LIF
- Investigation of polarity effect on O plasma using AC power source, with ring-to-ring electrode configuration

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