Electron power absorption in electronegative capacitively coupled discharges of complex chemistry

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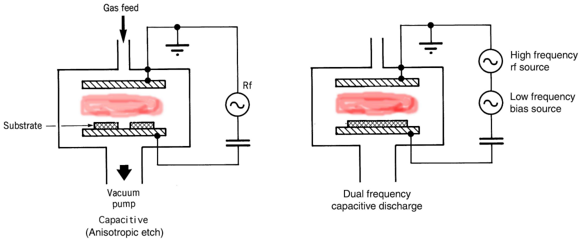
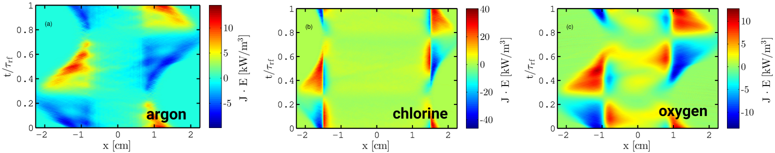
Introduction



Lieberman and Lichtenberg (2005), Principles of Plasma Discharges and Materials Processing, Wiley

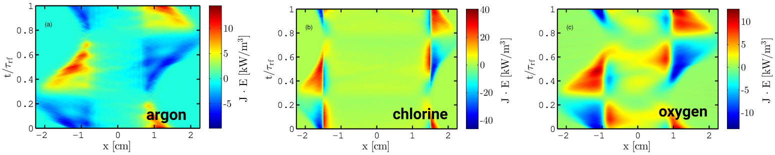
- The capacitive discharge is sustained by applying radio-frequency (rf) voltage or current between electrodes when a neutral gas at low pressure is injected
- The discharge is sustained by radio-frequency (rf) currents and voltages, introduced through capacitive sheaths

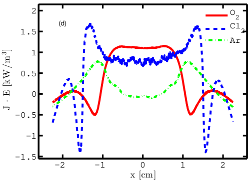
Introduction



- The two main electron power absorption mechanisms in these discharges are
  - momentum transfer due to the moving sheaths which leads to stochastic (pressure or collisionless) heating
  - currents in the main body of the plasma discharge lead to Ohmic (or collisional) heating in the bulk and sheath regions
- We explore this using 1D particle-in-cell/Monte Carlo collision simulation for three different chemistries
  - weakly electronegative oxygen discharge
  - highly electronegative chlorine discharge
  - electropositive argon discharge

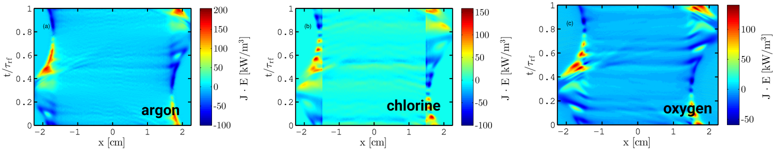
Introduction



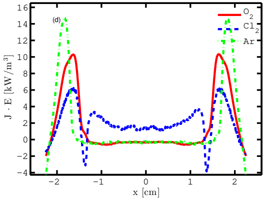


- The electron power absorption
  - $p = 1.33$ Pa
  - $f = 13.56$ MHz
  - $V_0 = 222$ V
  - gap = 45 mm
  - $\gamma_{see} = 0.0$
- For argon the discharge is in α-mode
- Chlorine and oxygen discharges operate in hybrid α- and drift ambipolar (DA)-mode

Introduction



- The electron power absorption
  - $p = 1.33$ Pa
  - $f = 27.12$ MHz
  - $V_0 = 222$ V
  - gap = 45 mm
  - $\gamma_{see} = 0.0$
- The argon and oxygen discharges are operated in α-mode
- The chlorine discharge operated in hybrid α- and drift ambipolar (DA)-mode



Outline

- **The 1D particle-in-cell/Monte Carlo collision simulation**
- **The low pressure oxygen discharge**
- **The low pressure chlorine discharge**
- **The intermediate pressure argon discharge**
- **Summary**

The 1D particle-in-cell/Monte Carlo collision simulation

The oopd1 1d-3v PIC/MCC code

- We use the `oopd1` (objective oriented plasma device for one dimension) code to simulate the discharges
- The `oopd1` code was originally developed by the Plasma Theory and Simulation Group at UC Berkeley later Michigan State University
- It has 1 dimension in space and 3 velocity components for particles (1d-3v)
- The `oopd1` code is supposed to replace the widely used `xpdx1` series (`xpdp1`, `xpdc1` and `xpds1`)
- It is developed to simulate various types of plasmas, including processing discharges, accelerators and beams
  - Modular structure
  - Includes relativistic kinematics
  - Particles can have different weights

Gudmundsson et al. (2013) *Plasma Sources Sci. Technol.*, **22**(3) 035011

Wen et al. (2021) *Plasma Sources Sci. Technol.*, **30**(10) 105009

The low pressure oxygen discharge

Introduction

- Oxygen forms a weakly electronegative discharge
- The oxygen discharge is of vital importance in various materials processing applications such as
  - ashing of photoresist
  - etching of polymer films
  - oxidation and deposition of thin film oxides
- The presence of negative ions has a strong influence on the kinetics and dynamics of the oxygen discharge
- The oxygen chemistry is rather involved, in particular due to the presence of **metastable molecular and atomic** oxygen and their role in dissociative attachment and detachment processes

The oxygen discharge

- We consider a discharge that consists of:
  - electrons
  - the ground state oxygen molecule $O_2(X^3\Sigma_g^-)$
  - the metastable oxygen molecule $O_2(a^1\Delta_g)$
  - the metastable oxygen molecule $O_2(b^1\Sigma_g)$
  - the ground state oxygen atom $O(^3P)$
  - the metastable oxygen atom $O(^1D)$
  - the negative oxygen ion O^-
  - the positive oxygen ions O^+ and O_2^+
- The discharge model includes energy dependent secondary electron emission yield

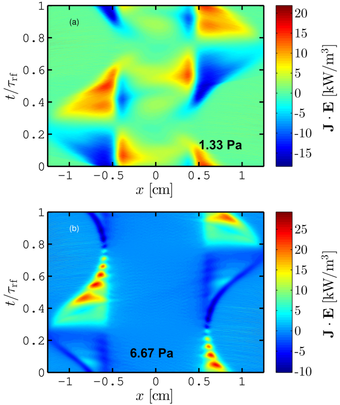
Hannesdottir and Gudmundsson, *Plasma Sources Sci. Technol.*, **25** 055002 (2016)

- We apply a global model beforehand to calculate the partial pressure of the various neutrals

Thorsteinsson and Gudmundsson, *Plasma Sources Sci. Technol.*, **19** 055008 (2010)

Oxygen CCP – pressure dependence

- A parallel plate capacitively coupled oxygen discharge at driving frequency of 13.56 MHz for gap separation of 45 mm
- The spatio-temporal electron heating $\mathbf{J}_\mathrm{e} \cdot \mathbf{E}$ at 1.33 and 6.67 Pa
- At 1.33 Pa there is a significant electron heating within the electronegative core
- At 6.67 Pa the electron heating occurs almost solely in the sheath region



Gudmundsson and Snorrason (2017) JAP **122** 193302

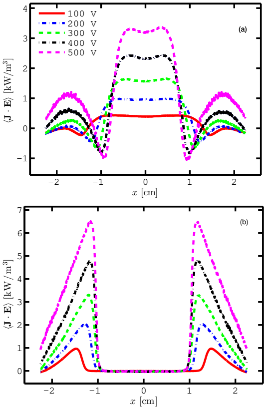
Hannesdottir and Gudmundsson (2016) PSST, **25** 055002

Gudmundsson and Ventéjou (2015) JAP **118** 153302

Oxygen CCP – pressure dependence

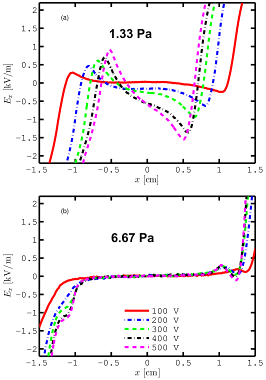
- The time averaged electron heating $\langle \mathbf{J}_{\rm e} \cdot \mathbf{E} \rangle$ at 1.33 and 6.67 Pa
- At 1.33 Pa there is significant electron heating within the electronegative core
- At 6.67 Pa, the heating rate in the electronegative core is roughly zero, and electron heating is almost entirely located in the sheath regions

Gudmundsson and Snorrason (2017) JAP **122** 193302



Oxygen CCP – pressure dependence

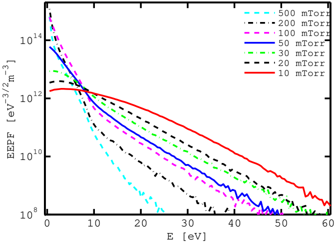
- The axial electric field at $t/\tau_{\mathrm{rf}} = 0.5$ for both 1.33 and 6.67 Pa
- At 1.33 Pa is a significant electric field strength within the electronegative core while at 6.67 Pa it is zero
- This strong electric field within the plasma bulk (the electronegative core) indicates a drift-ambipolar (DA) heating mode
- The electronegativity is significantly higher when operating at 1.33 Pa $\alpha > 20$ than when operating at 6.67 Pa $\alpha < 0.1$



Gudmundsson and Snorrason (2017) JAP **122** 193302

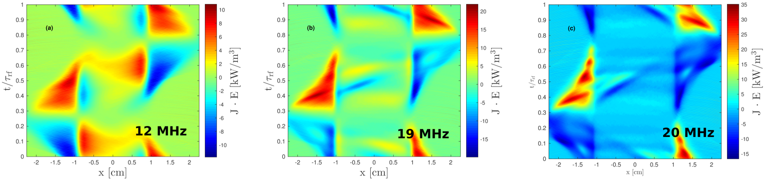
Oxygen CCP – pressure dependence

- At low pressure (< 4 Pa) the EEPF is Druyvesteyn like and becomes bi-Maxwellian as the pressure is increased
- These results contradict what is commonly found for the capacitively coupled argon discharge where the EEPF evolves from being bi-Maxwellian at low pressure to being Druyvesteyn like at high pressure



Gudmundsson and Ventéjou (2015) JAP **118** 153302

Oxygen CCP – frequency dependence

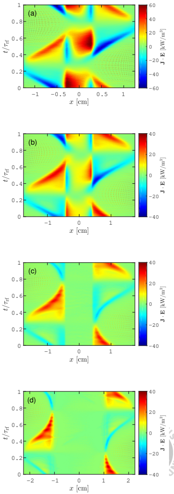


Gudmundsson et al. (2018), PSST **27**(2) 025009

- Oxygen discharge at 1.33 Pa
- At 12 MHz significant heating is observed in the plasma bulk but also in the sheath region
- At 19 MHz the heating and cooling in the sheath regions has increased, however there is contribution to the electron heating in the bulk region (note the change in scale)
- At 20 MHz there is almost no electron heating in the plasma bulk

Oxygen CCP – electrode spacing

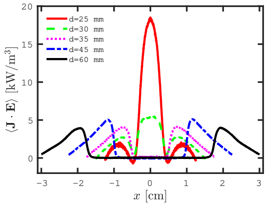
- A parallel plate capacitively coupled oxygen discharge at 6.67 Pa driven by a 400 V voltage source at 13.56 MHz as the gap separation is varied
- At small electrode spacing a combination of stochastic (α-mode) and drift ambipolar (DA) heating in the bulk plasma (the electronegative core) is observed
- As the electrode spacing is increased, the heating mode transitions into a pure α-mode

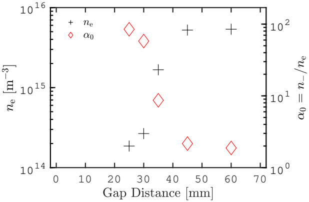


Gudmundsson and Proto (2019) PSST **28** 045012

Oxygen CCP – electrode spacing

- The time averaged electron power absorption $\langle \mathbf{J}_e \cdot \mathbf{E} \rangle$
- The electron density in the discharge center (left *y* axis) and the electronegativity in the discharge center (right *y* axis) as a function of the gap separation
- The heating mode transition coincides with a sharp decrease in electronegativity
- This agrees with recent experimental findings of You et al. (2019)



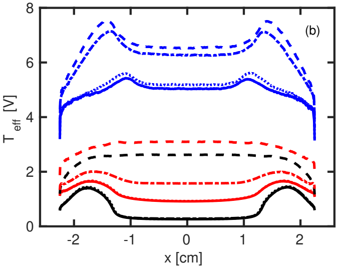


Gudmundsson and Proto (2019) PSST **28** 045012

Oxygen CCP – pressure dependence

- The effective electron temperature drops as the pressure is increased
- **1.33 Pa**, **6.67 Pa** and **26.67 Pa**
- When the metastable singlet oxygen molecule $O_2(a^1\Delta_g)$ is added to the discharge model the effective electron temperature drops
- This is due to detachment by the metastable $O_2(a^1\Delta_g)$ molecule

$$O^- + O_2(a^1\Delta_g) \rightarrow \text{products}$$

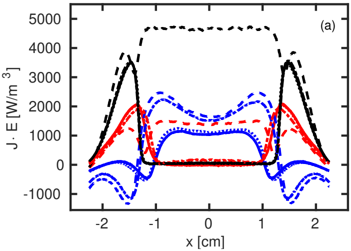


Gudmundsson and Lieberman (2015) PSST **24** 035016

Gudmundsson and Hannesdottir, AIP Conf. Proc. (2017) **1811** 120001

Oxygen CCP – pressure dependence

- At **1.33 Pa** excluding the metastable states in the simulation has very small influence on the heating mechanism
- At **6.67 Pa** the metastable states have a significant influence on the heating mechanism
- The role of the metastables is even more significant at **26.67 Pa**



Gudmundsson and Ventéjou (2015) JAP **118** 153302

Gudmundsson and Lieberman (2015) PSST **24** 035016

Gudmundsson and Hannesdottir, AIP Conf. Proc. (2017) **1811** 120001

Electron power absorption – Bolzmann term analysis

- To determine the electron power absorption mechanisms we apply Boltzman term analysis

Surendra and Dalvie (1993) PRE **48**(5) 3914 and Schulze et al. (2018) **27**(5) 055010

- The electric field within the electronegative core constitutes contributions through various different phenomena
- The electric field is composed of six terms

$$E = \underbrace{\underbrace{-\frac{m_\mathrm{e}}{e}\frac{\partial u_\mathrm{e}}{\partial t}}_{\mathrm{I}} + \underbrace{\frac{m_\mathrm{e}}{e}\frac{u_\mathrm{e}^2}{n_\mathrm{e}}\frac{\partial n_\mathrm{e}}{\partial x}}_{\mathrm{II}} + \underbrace{\frac{m_\mathrm{e}}{e}\frac{u_\mathrm{e}}{n_\mathrm{e}}\frac{\partial n_\mathrm{e}}{\partial t}}_{\mathrm{III}}}_{\text{inertia}} \underbrace{- \underbrace{\frac{T_\mathrm{e}}{n_\mathrm{e}}\frac{\partial n_\mathrm{e}}{\partial x}}_{\mathrm{IV}} - \underbrace{\frac{\partial T_\mathrm{e}}{\partial x}}_{\mathrm{V}}}_{\text{pressure}} \underbrace{- \underbrace{\frac{m_\mathrm{e} u_\mathrm{e} \nu_c}{e}}_{\mathrm{VI}}}_{\text{Ohmic}}$$

Electron power absorption – Bolzmann term analysis

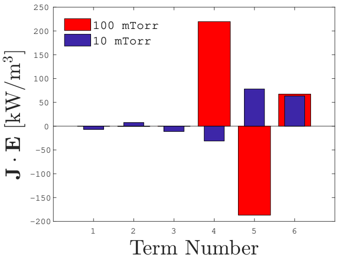
- The electron absorbed power can be determined as follows

$$J_e \cdot E = \underbrace{m_e u_e n_e \frac{\partial u_e}{\partial t}}_{\text{I}} - \underbrace{m_e u_e^3 \frac{\partial n_e}{\partial x}}_{\text{II}} - \underbrace{m_e u_e^2 \frac{\partial n_e}{\partial t}}_{\text{III}} + \underbrace{e u_e T_e \frac{\partial n_e}{\partial x}}_{\text{IV}} + \underbrace{e n_e u_e \frac{\partial T_e}{\partial x}}_{\text{V}} + \underbrace{m_e n_e \nu_c u_e^2}_{\text{VI}} \qquad (1)$$

  - Terms I and III – power absorption due to electron inertia
  - Term II – the electron density gradient
  - Term IV – ambipolar field – electron density gradient
  - Term V – the electron temperature gradient
    - Terms IV and V are pressure or collisionless heating
  - Term VI – electron neutral collisions or Ohmic heating

Oxygen CCP – Bolzmann term analysis

- The pressure terms are important – significant part of the electron power absorption at both 1.33 Pa and 13.3 Pa is due to the pressure terms
- The Terms IV (ambipolar) and V (electron temperature gradient) flip signs and are sharply smaller in the absolute value at 1.33 Pa
- The Ohmic term's magnitude (Term VI) is similar at both pressures
- Note that Ohmic power absorption is important even at low pressure



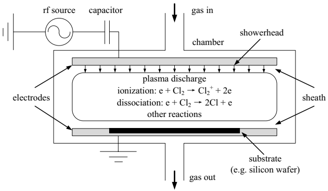
Proto and Gudmundsson (2020) JAP **128** 113302

The low pressure chlorine discharge

The chlorine discharge

- Chlorine is an electronegative diatomic gas that is widely used in plasma etching of both semiconductors and metals, in particular poly-silicon gate and aluminum interconnects
- Chlorine atoms are believed to be the primary reactant in plasma etching
- The chlorine molecule has
  - a low dissociation energy (2.5 eV)
  - a near-zero threshold energy for dissociative attachment
- All electronic excitations of the molecule appear to be dissociative, and no metastable molecular states are of importance

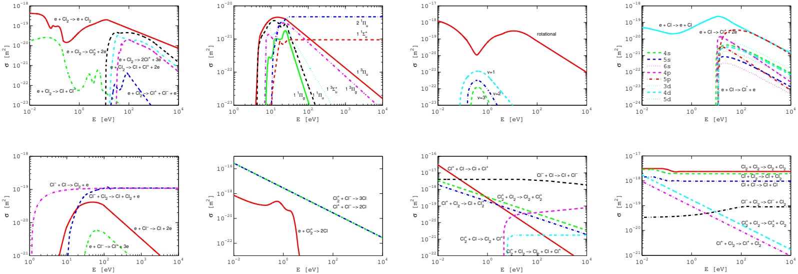
The chlorine discharge



- We consider a discharge that consists of:
  - electrons
  - the ground state chlorine molecule $Cl_2(X\,^1\Sigma_g^+, v = 0)$,
  - the ground state chlorine atom $Cl(3p^5\,^2P)$
  - the negative chlorine ion Cl^-
  - the positive chlorine ions Cl^+ and Cl_2^+
- We apply a global model beforehand to calculate the fraction of Cl atoms

Thorsteinsson and Gudmundsson (2010) *Plasma Sources Sci. Technol.*, **19**(1) 015001

The chlorine discharge



- The reaction set for the chlorine is comprehensive and includes 44 reactions

Huang and Gudmundsson (2013) *Plasma Sources Sci. Technol.*, **22**(5) 055020

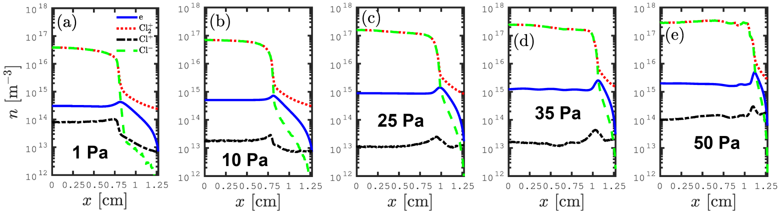
Electron power absorption

Voltage source operated at a single frequency

$$V(t) = V_{\rm rf} \sin(2\pi f t)$$

gap = 2.54 cm, $V_{\rm rf}$ = 222 V and f = 13.56 MHz

Electron power absorption



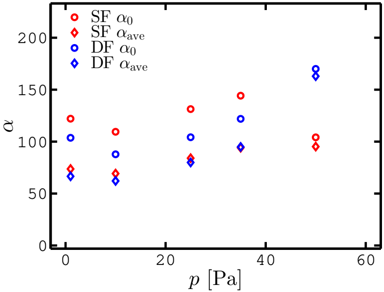
Proto and Gudmundsson (2021) PSST **30**(6) 065009

- The time averaged charged particle density profiles of a parallel plate capacitively coupled chlorine discharge
- At low pressures, the profile for Cl_2^+ ions is cosine-like or parabolic since Cl_2^+ ions are lost mainly due to diffusion to the walls
- As the pressure increases, the recombination between Cl_2^+ and Cl^- ions becomes the major loss mechanism for Cl_2^+ ions and the density profile for Cl_2^+ and Cl^--ions becomes flat in the bulk region

Electron power absorption

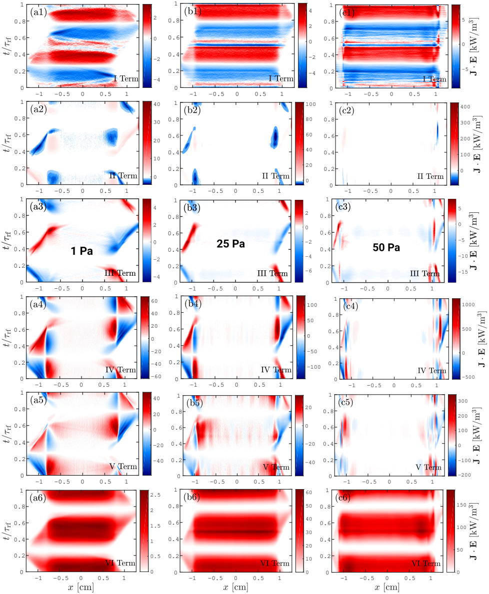
- The electronegativity in the discharge center is very high and increases with increased operating pressure
- Negative ion creation proceeds via electron impact dissociative attachment whose cross-sections are high
- Drift-ambipolar (DA) power absorption is expected within the electronegative core

Schulze et al. (2011) PRL **107**(27) 275001



Skarphedinsson and Gudmundsson (2020) PSST **29**(8) 084004

Electron power absorption



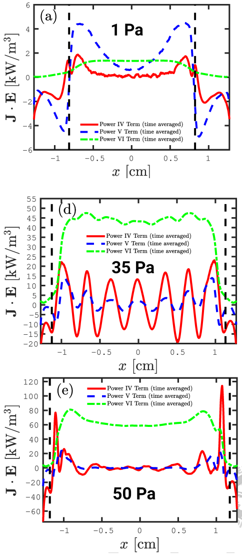
- The spatio temporal behavior of the various terms that constitute the electron power absorption
  - Terms I and III – electron inertia
  - Term I – electron density gradient
  - Terms IV and V – pressure (collisionless) heating
  - Term VI – Ohmic heating

Proto and Gudmundsson (2021) PSST **30**(6) 065009

Electron power absorption

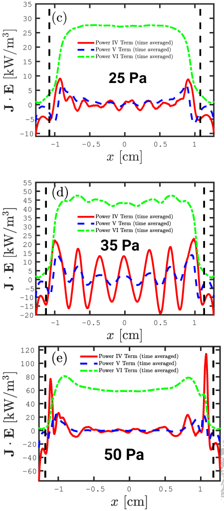
- The time averaged electron power absorption profile of
  - term IV (red line)
  - term V (blue dashed line)
  - term VI (green dot dashed line)
- At 1 Pa the pressure terms and the Ohmic term contribute to the electron power absorption
- At higher pressures Ohmic power absorption dominates
- At 35 Pa striations are observed

Proto and Gudmundsson (2021) PSST **30**(6) 065009



Electron power absorption

- Striations are known to appear in electronegative discharges when two conditions are simultaneously fulfilled:
  - high enough electronegativity
  - driving frequency comparable to the ion plasma frequency
- If fulfilled, positive and negatively charged ions oscillate and electric field is superimposed onto the drift field
- The appearance of the striations depends heavily on the ion-neutral collision frequency and therefore on the pressure

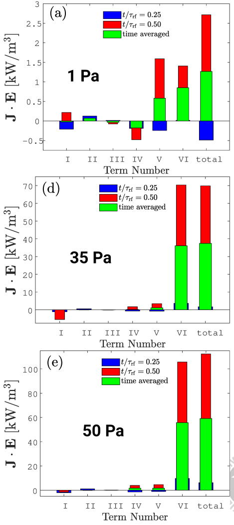


Liu et al. (2017) PSST **26**(5) 055024

Electron power absorption

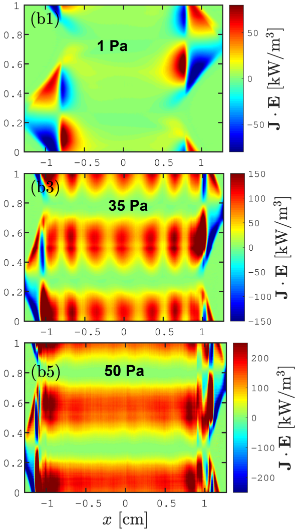
- The space averaged electron power absorption profile terms
  - $t/\tau_{\rm rf} = 0.25$ blue bar
  - $t/\tau_{\rm rf} = 0.5$ red bar
  - time averaged green bar
- At 1 Pa the pressure terms and the Ohmic term contribute to the electron power absorption
- At higher pressures Ohmic power absorption dominates

Proto and Gudmundsson (2021) PSST **30**(6) 065009



Electron power absorption

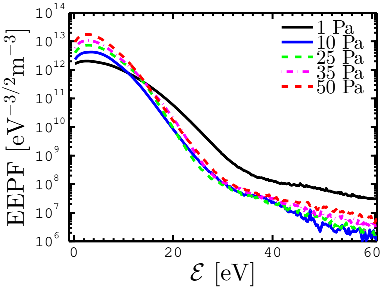
- The spatio temporal behavior of the total electron power absorption $\mathbf{J}_\mathrm{e} \cdot \mathbf{E}$ over the full gap length for a capacitively coupled chlorine discharge
- At 1 Pa there is clear sign of drift ambipolar heating (DA-mode) and stochastic heating (α-mode)
- At 35 and 50 Pa there are indications of striations in addition to drift ambipolar heating (DA-mode) and stochastic heating (α-mode)



Proto and Gudmundsson (2021) PSST **30**(6) 065009

Electron power absorption

- The electron energy probability function (EEPF) in the discharge center is Druyvesteyn like at all pressures
- This is expected when there is significant Ohmic heating in the plasma bulk



Tailored voltage waveforms

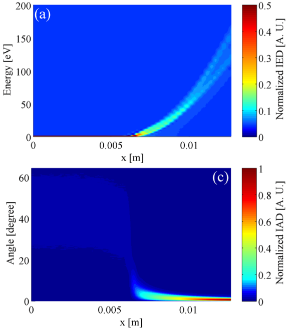
Dual frequency voltage source

$$V(t) = \frac{V_0}{2}\sin(2\pi ft) + \frac{V_0}{2}\sin(4\pi ft + \theta)$$

gap = 2.54 cm, V_0 = 222 V and f = 13.56 MHz

Tailored voltage waveforms

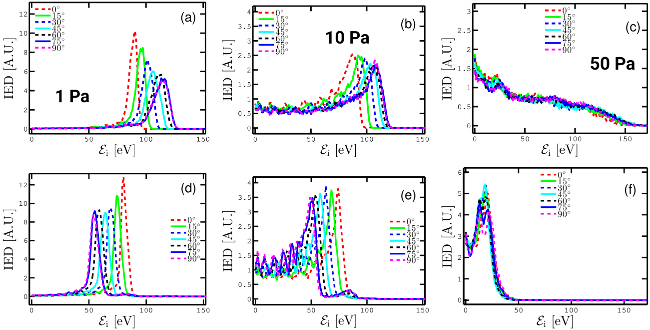
- By applying voltage at two or more frequencies, a fundamental frequency and its harmonics the ion bombarding energy can be controlled
- By adjusting the phase angle θ between the fundamental and the second harmonic a dc self-bias voltage can be generated
- This gives separate control of the ion flux and ion energy in a capacitively coupled discharge
- Electrically asymmetric discharge is formed in otherwise a geometrically symmetric reactor



Huang and Gudmundsson (2014)

TPS **42**(10) 2854

Tailored voltage waveforms



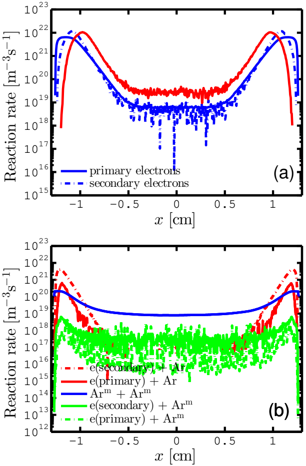
Skarphedinsson and Gudmundsson (2020) PSST **29**(8) 084004

- The IED for Cl_2^+ ions bombarding the electrodes while varying the phase angle, between the fundamental and the second harmonic
- The grounded (upper row) and the driven (lower row) electrode

The intermediate pressure argon discharge

The intermediate pressure argon discharge

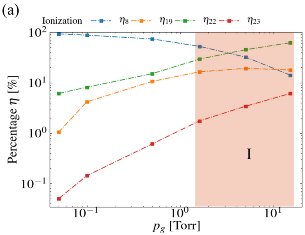
- Capacitive argon discharge at 213 Pa with 2.54 cm gap
- The reaction rates for ionization
  - neglecting excited states (upper) and (neglecting) and (including) secondary electrons
  - including excited state kinetics (lower), energy dependent secondary electron emission due to ion and atom bombardment of the electrodes, and electron reflection
- Most of the ionization occurs near the plasma-sheath interfaces, with little ionization within the bulk region



Gudmundsson et al. (2021) *Plasma Sources Sci Technol.*, accepted

The intermediate pressure argon discharge

- The ionization processes versus pressure neglecting secondary electrons
  - Electron impact ionization
  - Step-wise ionization
  - Metastables pooling ionization
  - Metastables pooling ionization
- The region labeled I ($p_g > 200$ Pa) indicates where the contributions of metastable pooling and step-wise ionization exceed that of electron impact ground state ionization



Wen et al. (2021) *Plasma Sources Sci. Technol.*, **30**(10) 105009

The intermediate pressure argon discharge

- When secondary electrons are included – transition from α-mode to γ-mode
  - electron impact by secondary 76 %
  - electron impact by primary 11 %
  - metastable Penning ionization 13 %
  - multi-step ionization, about 0.3 %
- Excited states are very effective in producing secondary electrons, with approximately 14.7 times the contribution of ion bombardment

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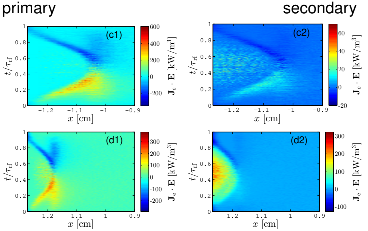
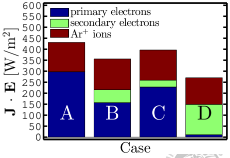
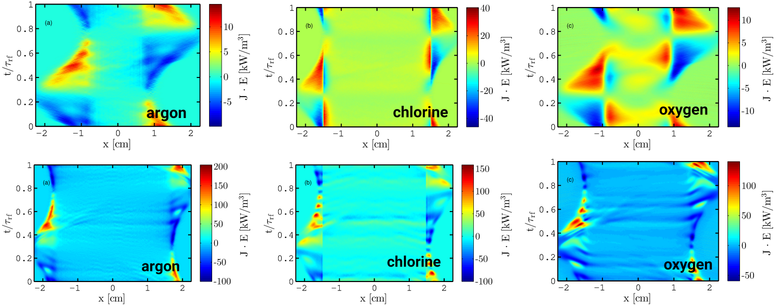




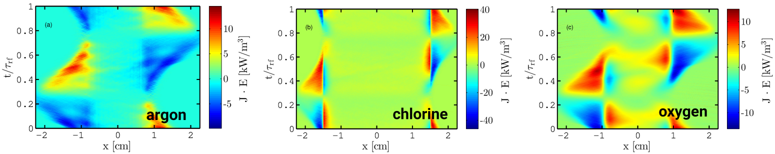
Summary

Summary



- Capcacitively coupled argon, oxygen and chlorine discharges were studied by particle-in-cell/Monte Carlo collision simulations

Summary



- In a chlorine discharge DA electron power absorption dominates and becomes increasingly more Ohmic with increased pressure
- Including the detachment processes by the singlet metastable states has a strong influence on the electronegativity in the oxygen discharge
- A heating mode transition, from hybrid drift-ambipolar (DA) and α-mode to pure α-mode, is observed as the pressure, driving frequency, or electrode gap spacing are increased, or the quenching coefficient of the oxygen metastable $O_2(a^1\Delta_g)$ is varied

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Thank you for your attention

The slides can be downloaded at

`http://langmuir.raunvis.hi.is/~tumi/ranns.html`

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