Electron heating in electronegative capacitively coupled discharge of complex chemistry

Jón Tómas Guðmundsson[1,2], Davíð I. Snorrason[1], Andrea Proto[1], and Hólmfríður Hannesdóttir[1]

[1]Science Institute, University of Iceland, Reykjavík, Iceland
[2]Department of Space and Plasma Physics,
KTH – Royal Institute of Technology, Stockholm, Sweden
tumi@hi.is

45th IEEE International Conference on Plasma Science
Denver, Colorado
June 25, 2018

Introduction

- Oxygen forms a weakly electronegative 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
- We use the `oopd1` (objective oriented plasma device for one dimension) **particle-in-cell Monte Carlo collision** code to simulate the discharge
- It has 1 dimension in space and 3 velocity components for particles (1d-3v)
- 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., *Plasma Sources Sci. Technol.*, **22** 035011 (2013)

The oxygen discharge

- We assume a parallel plate capacitively coupled oxygen discharge at with electrode separation of 4.5 cm
- Nine species:
  - electrons, $O_2(X^3\Sigma_g^-)$, $O_2(a^1\Delta_g)$, $O_2(b^1\Sigma_g)$, $O(^3P)$, $O(^1D)$, O^-, O^+ and O_2^+
- The reaction set for the oxygen is comprehensive and for this study includes 67 reactions
- The discharge model includes energy dependent secondary electron emission yield
- We apply a global model[1] beforehand to calculate the partial pressure of the various neutrals

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

Gudmundsson and Lieberman, *Plasma Sources Sci. Technol.*, **24** 035016 (2015)

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

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

Outline

- Capacitively Coupled Oxygen Discharge at 13.56 MHz
  - including both $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g)$
  - including secondary electron emission
- Electron heating mechanism
  - Pressure dependence
  - Frequency dependence
  - Dependence on surface quenching of $O_2(a^1\Delta_g)$
- Summary

Capacitively Coupled Oxygen Discharge single frequency at 13.56 MHz

– pressure dependence –

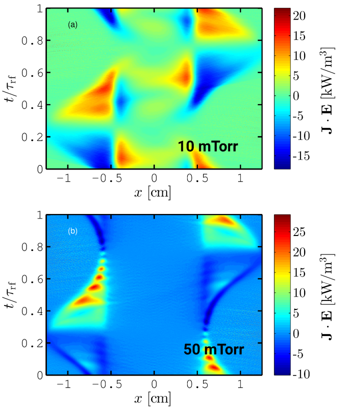
including $O_2(a^1\Delta_g)$, $O_2(b^1\Sigma_g)$ and $\gamma_{\rm see}(E)$

Oxygen CCP – pressure dependence

- The spatio-temporal electron heating $\mathbf{J}_e \cdot \mathbf{E}$ at 10 and 50 mTorr
- At 10 mTorr there is a significant electron heating within the electronegative core
- At 50 mTorr the electron heating occurs almost solely in the sheath region

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

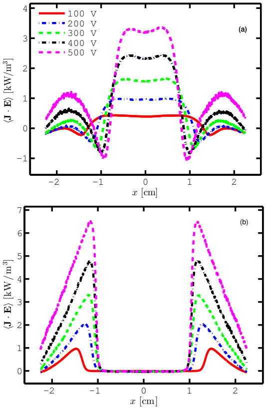
Gudmundsson and Ventéjou (2015) JAP **118** 153302



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

Oxygen CCP – pressure dependence

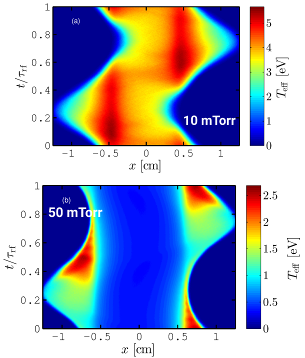
- The time averaged electron heating $\langle \mathbf{J}_e \cdot \mathbf{E} \rangle$ at 10 and 50 mTorr
- At 10 mTorr there is significant electron heating within the electronegative core
- At 50 mTorr, the heating rate in the electronegative core is roughly zero, and electron heating is almost entirely located in the sheath regions



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

Oxygen CCP – pressure dependence

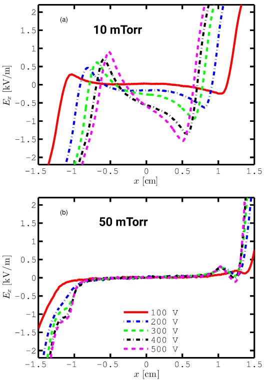
- At 10 mTorr the effective electron temperature is high within the plasma bulk (the electronegative core) throughout the rf period and peaks within the plasma bulk during the sheath collapse phase
- At 50 mTorr a peak in the effective electron temperature within the plasma bulk in the sheath expansion phase and is low within the plasma bulk throughout the rf period



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

Oxygen CCP – pressure dependence

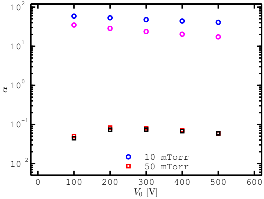
- The axial electric field at $t/\tau_{\text{rf}} = 0.5$ for both 10 and 50 mTorr
- At 10 mTorr there is a significant electric field strength within the electronegative core
- This strong electric field within the plasma bulk (the electronegative core) indicates a drift-ambipolar (DA) heating mode
- This electric field is a combination of a drift field and an ambipolar field
- At 50 mTorr the electric field is zero within the electronegative core

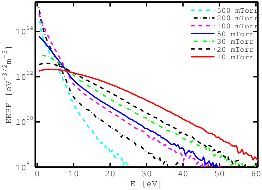


Gudmundsson and Snorrason (2017) JAP **122** 193302

Oxygen CCP – pressure dependence

- The electronegativity is significantly higher when operating at 10 mTorr than when operating at 50 mTorr
- At 10 mTorr, the discharge is operated in a combined drift-ambipolar (DA) and α-mode
- At 50 mTorr, the discharge is in a pure α-mode and sheath heating dominates
- The transition from the combined DA-α-mode to the pure α-mode coincides with a significant decrease in the electronegativity





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

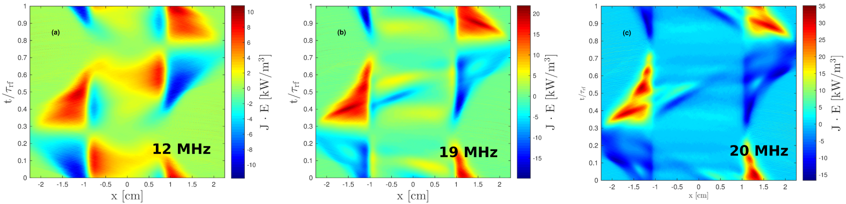
Gudmundsson and Ventéjou (2015) JAP **118** 153302

Capacitively Coupled Oxygen Discharge single frequency at 10 mTorr

– driving frequency dependence –

including $O_2(a^1\Delta_g)$, $O_2(b^1\Sigma_g)$ and $\gamma_{\rm see}(E)$

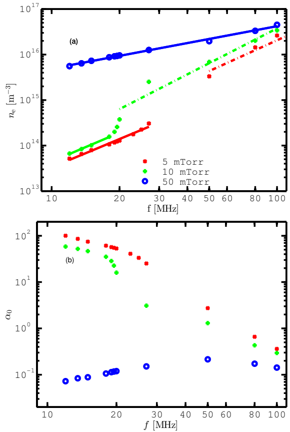
Oxygen CCP – frequency dependence



- 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 – frequency dependence

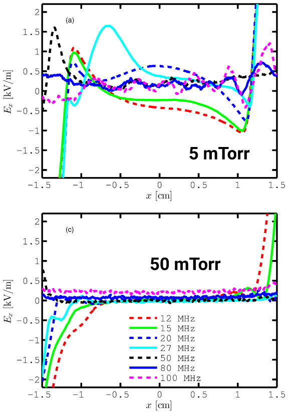
- At 10 mTorr there is a jump in the center electron density between 20 and 27 MHz
- At 10 mTorr $n_e \propto f^{2.11}$ at low frequency, below 18 MHz, and $n_e \propto f^{2.00}$ at higher frequencies, 27.12 MHz and above
- At 50 mTorr $n_e \propto f^{1.16}$ over the entire frequency range explored and no transition is observed
- We see that at 5 and 10 mTorr the electronegativity decreases with increasing driving frequency



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

Oxygen CCP – frequency dependence

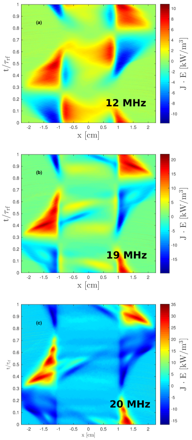
- The electric field profile at $t/\tau_{\mathrm{rf}} = 0.5$ for discharges operated at 5 and 50 mTorr
- We see a significant electric field strength within the electronegative core at low driving frequency and low pressure
- The strong electric field within the plasma bulk (the electronegative core), at low pressure and low driving frequency, indicates a drift-ambipolar (DA) heating mode



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

Oxygen CCP – frequency dependence

- At a low driving frequency and low pressure (5 and 10 mTorr), a combination of stochastic (α-mode) and drift ambipolar (DA) heating in the bulk plasma (the electronegative core) is observed
- The DA-mode dominates the time averaged electron heating
- As the driving frequency is increased, the heating mode transitions into a pure α-mode
- At low pressure (5 and 10 mTorr), this transition coincides with a sharp decrease in electronegativity



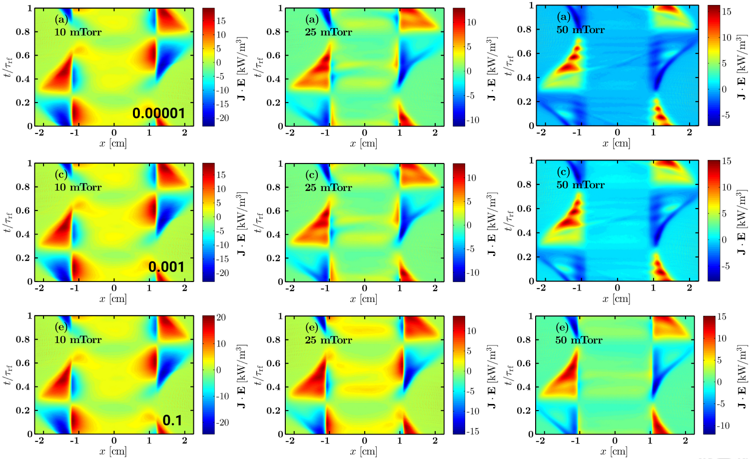
Gudmundsson et al. (2018) PSST **27** 025009

Capacitively Coupled Oxygen Discharge

– surface quenching of $O_2(a^1\Delta_g)$ –

including $O_2(a^1\Delta_g)$, $O_2(b^1\Sigma_g)$ and $\gamma_{\rm see}(E)$

Oxygen CCP – surface quenching of $O_2(a^1\Delta_g)$

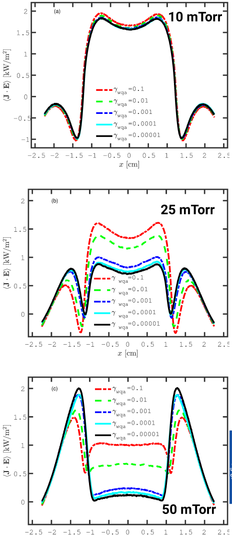


Proto and Gudmundsson (2018) PSST accepted for publication

Oxygen CCP – surface quenching of $O_2(a^1\Delta_g)$

- At 10 mTorr almost all the electron heating occurs in the plasma bulk (the electronegative core) and the electron heating profile is independent of the surface quenching coefficient
- At 50 mTorr only for the highest surface quenching coefficients 0.1 and 0.01 there is some electron heating observed in the bulk region
- Typical value is 0.007 for iron (Sharpless and Slanger, 1989)

Proto and Gudmundsson (2018) PSST accepted for publication



Summary

Summary

- We demonstrated particle-in-cell/Monte Carlo collision simulation of a capacitively coupled discharge
- Including the detachment processes by the singlet metastable states has a strong influence on the effective electron temperature and electronegativity in the oxygen discharge
- At low pressure the discharge is operated in a combined drift-ambipolar (DA) and α-mode, and at higher pressure it is operated in the pure α-mode
- At low operating frequency the discharge is operated in a combined drift-ambipolar (DA) and α-mode, and at higher frequency it is operated in the pure α-mode
- The transition in heating mechanism from DA-α to α-mode is accompanied by a drop in electronegativity

Acknowledgments

Thank you for your attention

The slides can be downloaded at
`http://langmuir.raunvis.hi.is/~tumi/ranns.html`

The work was funded by Icelandic Research Fund grants nos. 130029 and 163086 and the Swedish Government Agency for Innovation Systems (VINNOVA) contract no. 2014-04876

References

Gudmundsson, J. T., E. Kawamura, and M. A. Lieberman (2013). A benchmark study of a capacitively coupled oxygen discharge of the oopd1 particle-in-cell Monte Carlo code. *Plasma Sources Science and Technology 22*(3), 035011.

Gudmundsson, J. T. and M. A. Lieberman (2015). On the role of metastables in capacitively coupled oxygen discharges. *Plasma Sources Science and Technology 24*(3), 035016.

Gudmundsson, J. T. and D. I. Snorrason (2017). On electron heating in a low pressure capacitively coupled oxygen discharge. *Journal of Applied Physics 122*(19), 193302.

Gudmundsson, J. T., D. I. Snorrason, and H. Hannesdottir (2018). The frequency dependence of the discharge properties in a capacitively coupled oxygen discharge. *Plasma Sources Science and Technology 27*(2), 025009.

Gudmundsson, J. T. and B. Ventéjou (2015). The pressure dependence of the discharge properties in a capacitively coupled oxygen discharge. *Journal of Applied Physics 118*(15), 153302.

Hannesdottir, H. and J. T. Gudmundsson (2016). The role of the metastable $O_2(b^1\Sigma_g^+)$ and energy-dependent secondary electron emission yields in capacitively coupled oxygen discharges. *Plasma Sources Science and Technology 25*(5), 055002.

Proto, A. and J. T. Gudmundsson (accepted 2018). The role of surface quenching of the singlet delta molecule in a capacitively coupled oxygen discharge. *Plasma Sources Science and Technology*.

Thorsteinsson, E. G. and J. T. Gudmundsson (2010). The low pressure Cl_2/O_2 discharge and the role of ClO. *Plasma Sources Science and Technology 19*(5), 055008.

Sharpless, R. L. and T. G. Slanger (1989). Surface chemistry of metastable oxygen. II. Destruction of $O_2(a^1\Delta_g)$. *Journal of Chemical Physics 91*(12), 7947 – 7950.