Particle in cell Monte Carlo collision simulations of capacitive Ar/Cl_2 discharges

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The oopd1 1d-3v PIC/MCC code

- We use the `oopd1` (objective oriented plasma device for one dimension) code to simulate the discharge
- The `oopd1` code was originally developed at the Plasma Theory and Simulation Group at UC Berkeley
- It has 1 dimension in space and 3 velocity components for particles (1d-3v)
- Modular structure
- Includes relativistic kinematics

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

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

The oopd1 1d-3v PIC/MCC code

- We consider a chlorine 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^+

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

- We consider an argon discharge that consists of:
  - electrons
  - the ground state argon atom Ar
  - the metastable argon atom Ar^m (two levels as one species)
  - the radiative levles of the argon atom Ar^r (two levels as one species)
  - the excited argon levels Ar(4p) (all as one species)
  - the argon ion Ar^+

Gudmundsson et al. (2021) *Plasma Sources Sci. Technol.*, **30**(12) 125011

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

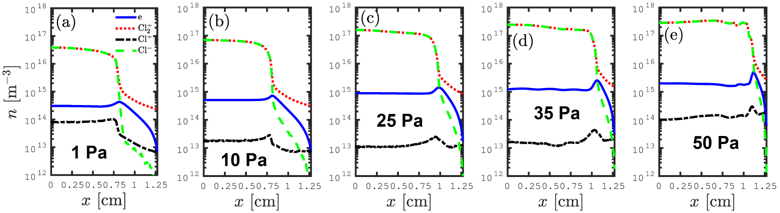
Electron power absorption
Chlorine

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

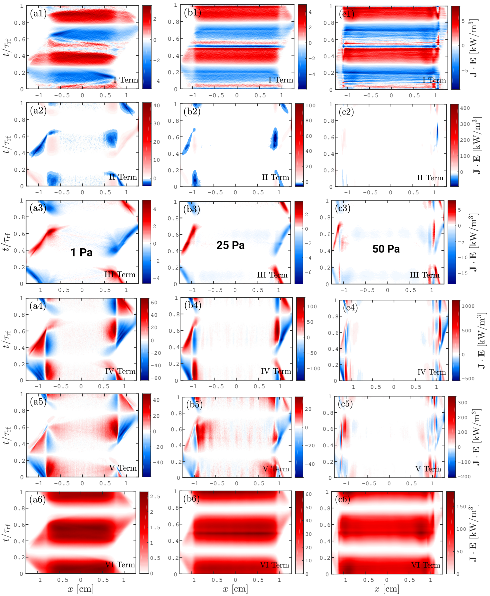
Electron power absorption – Chlorine



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 – Chlorine



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

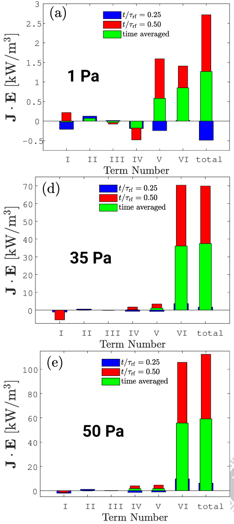
Proto and Gudmundsson (2021) PSST **30**(6) 065009

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

Electron power absorption – Chlorine

- The space averaged electron power absorption profile terms
  - $t/\tau_{\mathrm{rf}} = 0.25$ blue bar
  - $t/\tau_{\mathrm{rf}} = 0.5$ 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



Surface effects
Chlorine

Voltage source operated at a single frequency

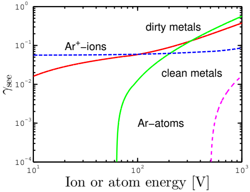
$$V(t) = V_{\mathrm{rf}} \sin(2\pi f t)$$

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

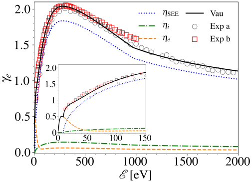
Secondary electron emission – Chlorine

Secondary electron emission due to

- ion bombardment of the electrodes
- neutral bombardment of the electrods
  - Using fits for argon bombardent developed by Phelps and Petrović (1999)
- electron bombardment of the electrodes
  - Using the modified Vaughan method as described by Wen et al. (2023)



based on Phelps and Petrović (1999) PSST **8** R21



Wen et al. (2023) PSST **32** 064001

Secondary electron emission – Chlorine

- The electron density, the Cl^- density, and the electronegativity in the discharge center (α_0) versus pressure
- With increasing pressure the electronegativity in the discharge center α_0 increases before it stabilizes at a relatively constant level
- Secondary electron emission from the electrodes leads to a reduction in electronegativity, which is especially noticeable at higher pressures

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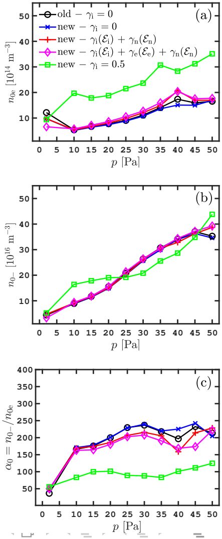
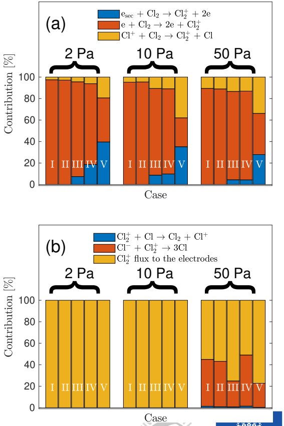


Secondary electron emission – Chlorine

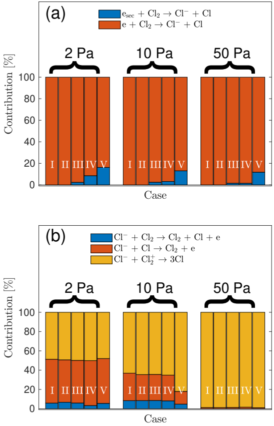
- The creation and loss of Cl_2^+ ions
- Electron impact ionization creates the Cl_2^+ ions
- At the lower pressures the Cl_2^+ ions are almost entirely lost as they bombard the electrodes
- With increasing pressure to 50 Pa the role of ion-ion mutual recombination increases

Mahdavipour and Gudmundsson (2024) PSST **33**(6) 065006



Secondary electron emission – Chlorine

- The creation and loss of Cl^- ions
- The creation of the negative ion Cl^- is almost entirely due to dissociative attachment
- The dominating loss processes for the negative ion Cl^- at 50 Pa is ion-ion mutual neutralization $Cl^- + Cl_2^+$
- Lowering the pressure associative detachment by the Cl atom and detachment by the Cl_2 molecule play an increasing role, with over 50 % contribution at 2 Pa



Mahdavipour and Gudmundsson (2024) PSST **33**(6) 065006

Argon diluted by chlorine

Current source operated at a single frequency

$$J(t) = J_{\mathrm{rf}} \sin(2\pi f t)$$

gap = 2.54 cm, J_{rf} = 50 A/m^2 and f = 13.56 MHz, p = 6.67 Pa

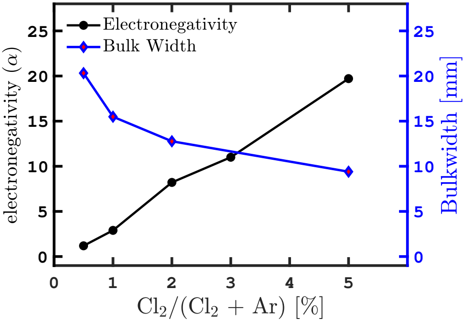
Argon diluted by chlorine

Case	exited species	ions	neutrals	electrons
I	0.0	0.0	0.0	0.0
II	included	0.0	0.0	0.0
III	included	$\gamma_{\mathrm{see,i}}(\mathcal{E}_{\mathrm{i}})$	$\gamma_{\mathrm{see,n}}(\mathcal{E}_{\mathrm{n}})$	$\gamma_{\mathrm{see,e}}(\mathcal{E}_{\mathrm{e}}, \theta)$

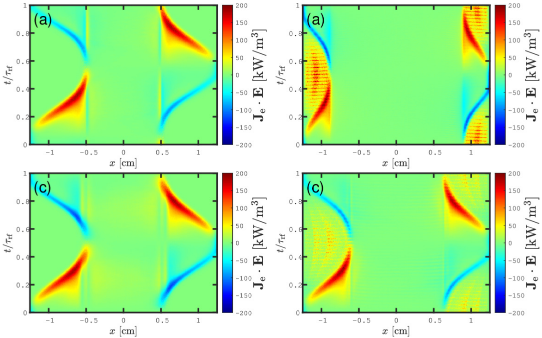
- We explored three cases
  - Neglecting excited states of argon and all secondary electron emission
  - Including excited states of argon and neglecting all secondary electron emission
  - Including excited states of argon and all secondary electron emission

Argon diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The electronegativity increases with increased chlorine fraction
- The plasma bulk width decreases with increased chlorine fraction
- Gap width 2.5 cm, driving current density 50 A/m^2 and frequency 13.56 MHz
- The argon model includes excited argon atoms



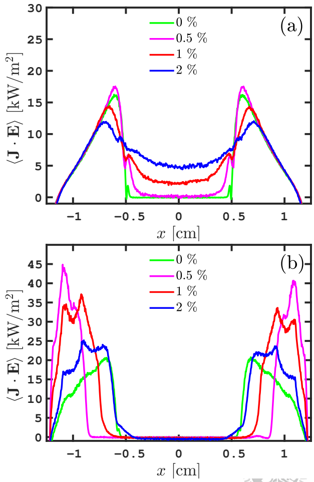
Argon diluted by chlorine



- The spatio-temporal behavior of primary electron power absorption at (a) 0.5 % and (c) 2 % chlorine
  - Left: neglecting excited argon atoms
  - Right: including excited states of the argon atom

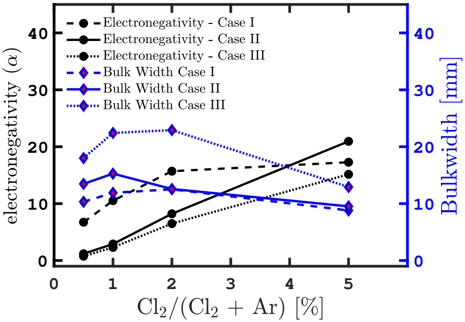
Argon diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The time-averaged electron power absorption for varying chlorine dilution
  - (a) neglecting excited argon states
  - (b) including excited argon states



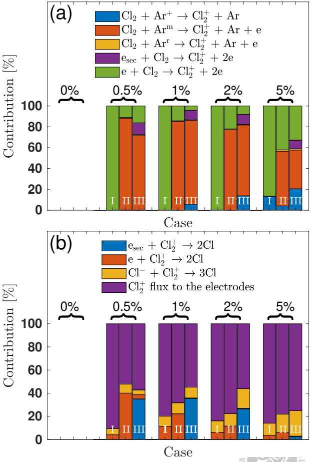
Argon diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The electronegativity decreases with addition of excited argon states
- The electronegativity decreases with addition of secondary electron emission from electrodes
- The plasma bulk width increases with addition of secondary electron emission from electrodes



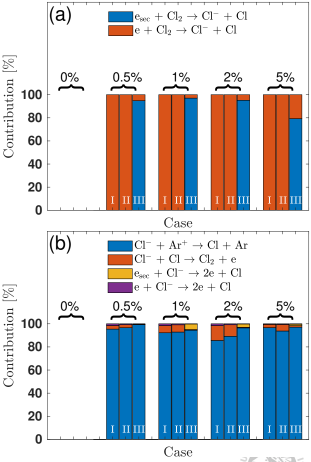
Argon diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The creation and loss of the Cl_2^+ ion
- The role of Penning ionization becomes rather significant as excited states of argon are included in the discharge model
- Gap width 2.5 cm, driving current density 50 A/m^2 and frequency 13.56 MHz



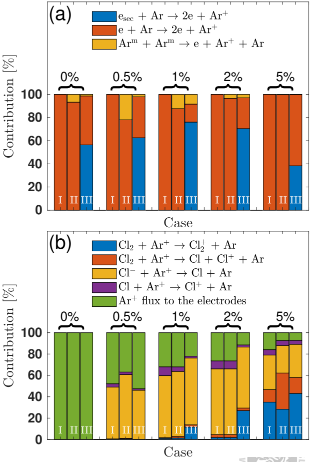
Argon diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The creation and loss of the negative Cl^- ion
- The creation of the negative ion Cl^- is almost entirely due to dissociative attachment as for pure chlorine discharge
- The role of secondary electrons becomes significant as they are included in the discharge model
- Ion-ion recombination with Ar^+ is the main loss process



Argon diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The creation and loss of the Ar^+ ion
- The contribution of secondary electrons becomes significant as they are included in the discharge model



Summary

Summary

- The chlorine discharge exhibits high electronegativity
- At the lowest pressure (1 Pa) the electron power absorption is due to both the pressure and the Ohmic terms and at higher pressure Ohmic terms dominate (drift-ambipolar (DA) mode)
- Penning ionization of the chlorine molecule is rather significant source of Cl_2^+
- The role of secondary electrons becomes significant for the dissociative attachment to create Cl^- as they are included in the discharge model

Thank you for your attention

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