SELF-CONSISTENT NUMERICAL SIMULATION OF CARBON ARC FOR NANOPARTICLE SYNTHESIS

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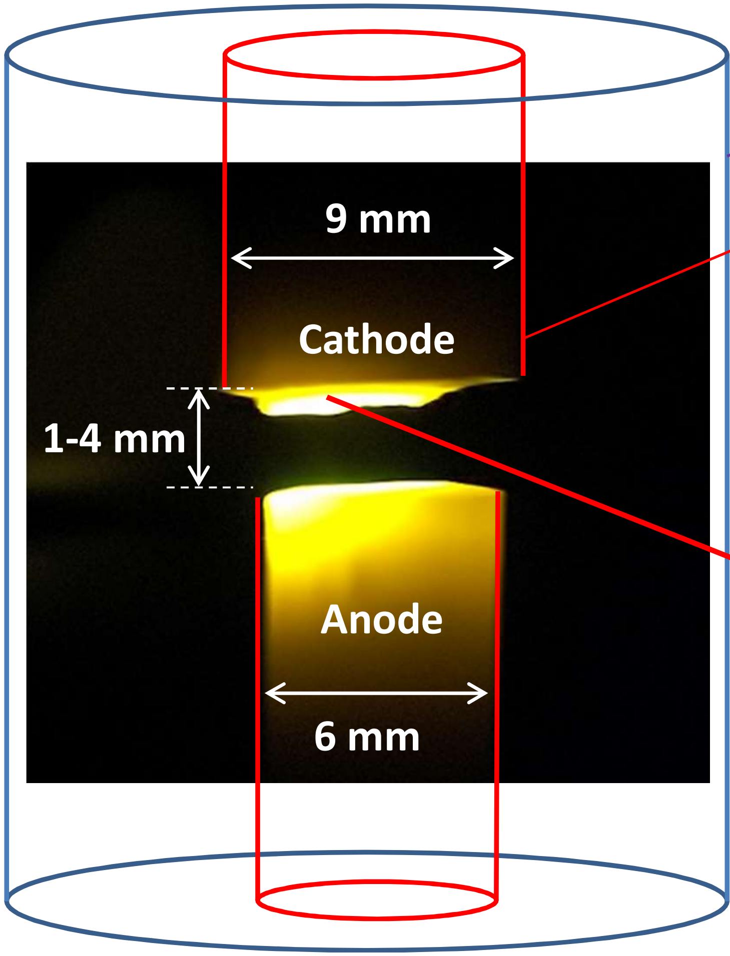
Carbon arc discharge for synthesis of nanomaterials

Simulations and experiments are conducted at Princeton Plasma Physics Lab. (nano.pppl.gov)



Helium filled chamber, 500 Torr

Graphite electrodes, long cylinders (10 cm)

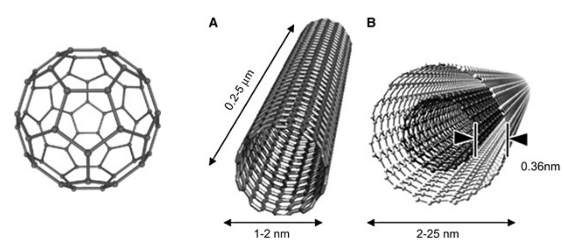
Arc current 50 A – 70 A

Anode ablates providing carbon into the arc region

(Helium does to ionize at these conditions)

Carbon deposit grows on a cathode
(low thermal conductivity)

Some part of carbon material escapes from the arc,
cools down and forms nanoparticles of different types:



Typical current density – 10^7 A/m^2

Typical voltage ≈ 20 V

Arc model, general features

- **Fluid model of plasma**
- **Short arc → near-electrode non-equilibrium effects are important:**
  - Non-equilibrium plasma ($T_e \neq T_g$, $n_e \neq n_{Saha}$)
  - Drift and diffusion of electrons
  - Near-electrode space-charge limited sheathes
- **Self-consistent arc model**
  - Conjugate heat transfer and current flow in gas and electrodes
  - Self consistent plasma-electrodes boundary conditions:
    - energy balance of electron gas, heavy particles, electrodes
    - unified ablation/deposition boundary condition

- **The arc model was implemented into a general-purpose code ANSYS-CFX**
  (ANSYS-CFX code was highly customized)
- **The model was benchmarked against 1D simulations of N. Almeida et.al. 2008 for argon arc**
  (1D code was developed)
- **The model was validated against analytical solutions**
  (which was developed for better understanding of the arc physics)

Governing equations in plasma

2D-3D model, C-He

Momentum equation: $\nabla \cdot (\rho \vec{v}\vec{v}) = -\nabla p + \nabla \cdot (\mu \nabla \vec{v}) + \rho \vec{g}$

Continuity equation: $\nabla \cdot (\rho \vec{v}) = 0$

Neutrals transport equation: $\nabla \cdot (\rho c_C \vec{v}) = \nabla (D \nabla (\rho c_C)) - S_i$

1D model, Ar

$p = const$

$\vec{v} = 0$

$G_{Ar} = -G_i$

Equation of state: $p = (n_{neutrals} + n_i)kT + n_e kT_e$

Ions transport equation (non-LCE): $\nabla \cdot (n_i \vec{v}) = \nabla \left(D_a \nabla n_i + D_{th\,diff} \nabla T + D_{th\,diff,e} \nabla T_e + j_e \gamma_{e,i} \right) + S_i, \quad S_i = \alpha n_e n_{C,Ar} - \beta n_e^3$ *

Transport of electrons (generalized Ohm's law): $\nabla^2 V = -\frac{k}{e} \nabla \left(C_e \nabla T_e + T_e \nabla \ln n_e - \frac{\vec{j}_i}{\sigma} \right)$ | $\vec{E} = -\frac{k}{e} (C_e \nabla T_e + T_e \nabla \ln n_e) + \frac{\vec{j}_e}{\sigma}$

Energy balance of electrons (non-LTE): $\nabla \cdot \left((2.5 + A_e) k T_e \frac{\vec{j}_e}{e} \right) = \nabla \cdot (\lambda_e \nabla T_e) - S_i E_i - Q^{electrons-heavy} - Q^{rad} + \vec{j} \cdot \vec{E}$

Energy balance of heavy particles (non-LTE): $\nabla \cdot (\rho \varepsilon \vec{v}) = \nabla \cdot (\lambda \nabla (T)) + Q^{electrons-heavy} + \vec{j}_i \cdot \vec{E}$

Quasi neutrality: $n_e = n_i$ | Gauss's law: $e(n_e - n_i) = \varepsilon_0 \nabla \cdot \vec{E}$

** Ionization/recombination coefficients* *(J. Annaloro et.al., 2012, Physics of plasmas 19)*

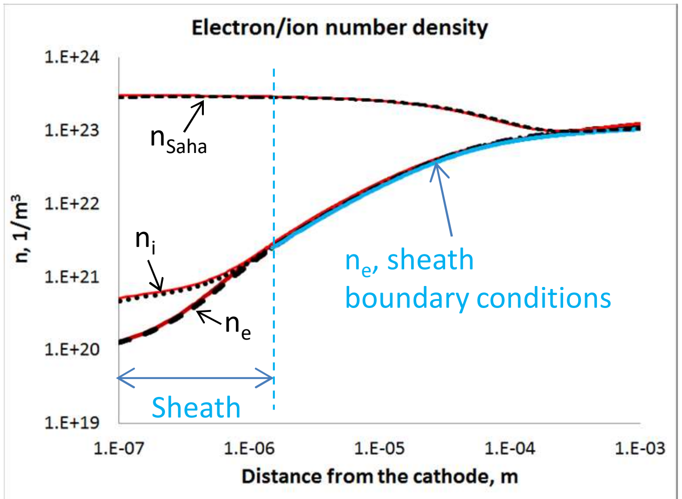
Transport coefficients $D_{...}, A_e, C_e, \sigma, \lambda_e, \lambda$ *and terms* $Q^{electrons-heavy}, Q^{rad}$ *are functions of* $T_e, T, n_e, n_a, Q_{e,i}, Q_{e,a}$, *derived from kinetic theory* *(see N. Almeida et.al., 2008, J. Phys. D: Appl. Phys. 41)*

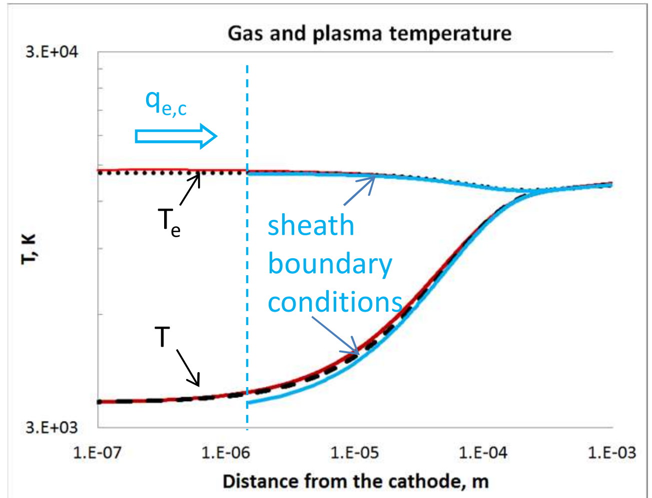
1D simulations of argon arc (benchmarking of the model)

Benchmarking of the model, 1D simulations

Argon arc, near-cathode layer.

- - - - **our code,** **——** **simulations by N. Almeida et.al.** *(N. Almeida et.al., 2008, J. Phys. D: Appl. Phys. 41)*





P = 1 atm

j = 5·10⁶ A/m²

$T_{cathode}$ = 3500K

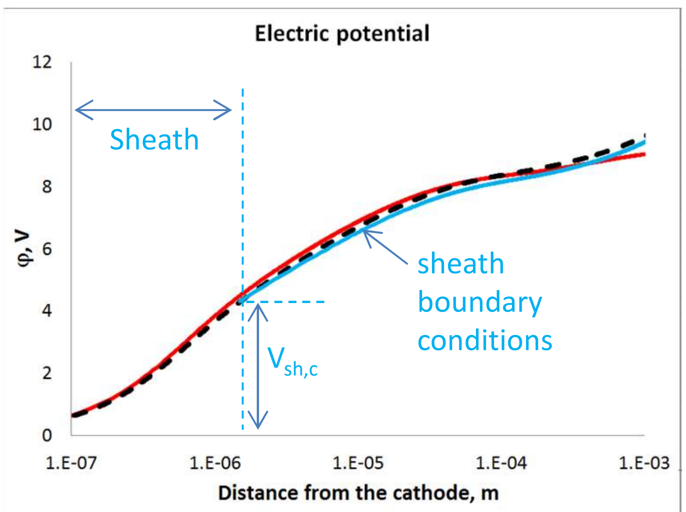
Boundary conditions (collisionless sheath):

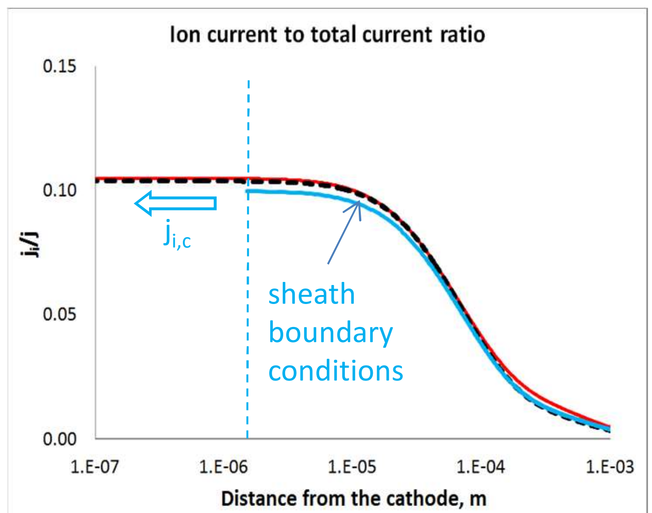
$$j = j_{e,c}^{p} + j_{e,c}^{emiss} + j_{i,c}$$

$$j_{i,c} = q_e n_i \sqrt{\frac{k(T_e + T)}{m_a}}$$

$$j_{e,c}^{p} = q_e n_e \frac{1}{4}\sqrt{\frac{kT_e}{m_e}} \cdot e^{-\frac{V_{sh,c}}{kT_e}}$$

$$q_{e,c} = j_{e,c}^{emiss}\left(V_{sh,c} + 2.5T_c\right) - j_{e,c}^{p}\left(V_{sh,c} + 2.5T_e\right)$$



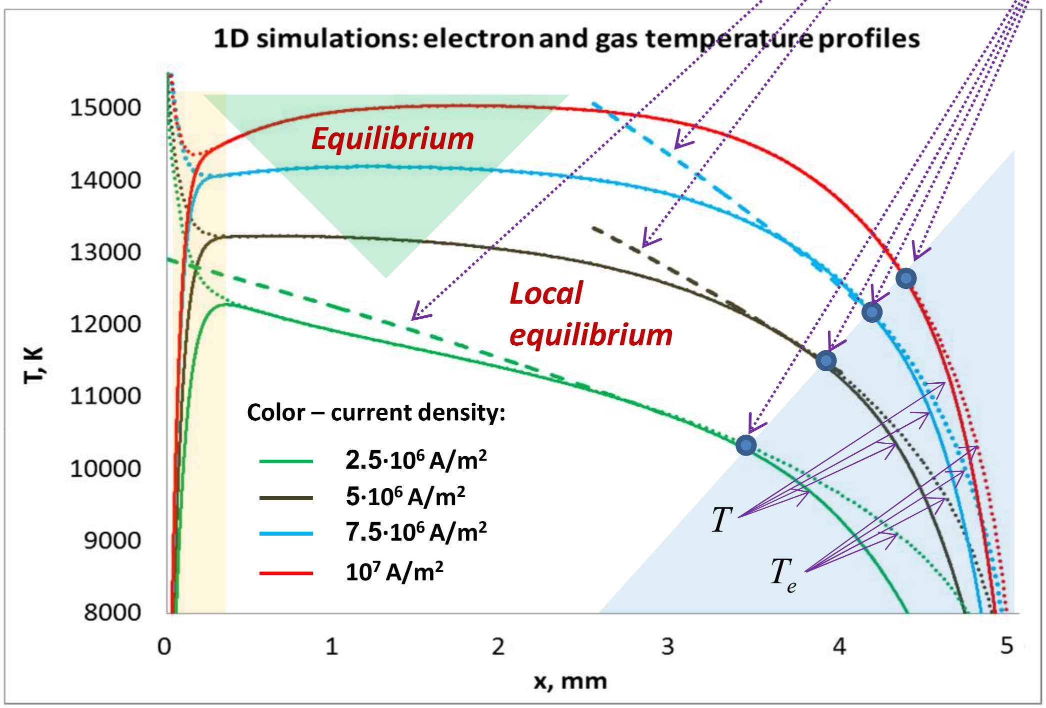


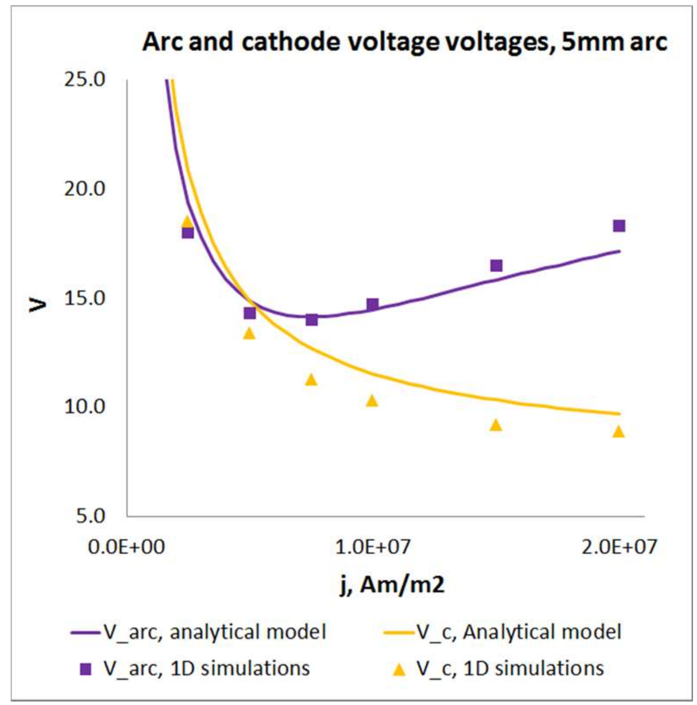
Analytical arc model

Cathodic region: $V_{cathode} = \frac{q_{c,loss}(T_c)}{j} + V_w + 3.2\frac{k}{e}T_{e,plasma}$ $\quad T_{e,plasma} = f(j_{ion}, k_i) \approx 15000K$

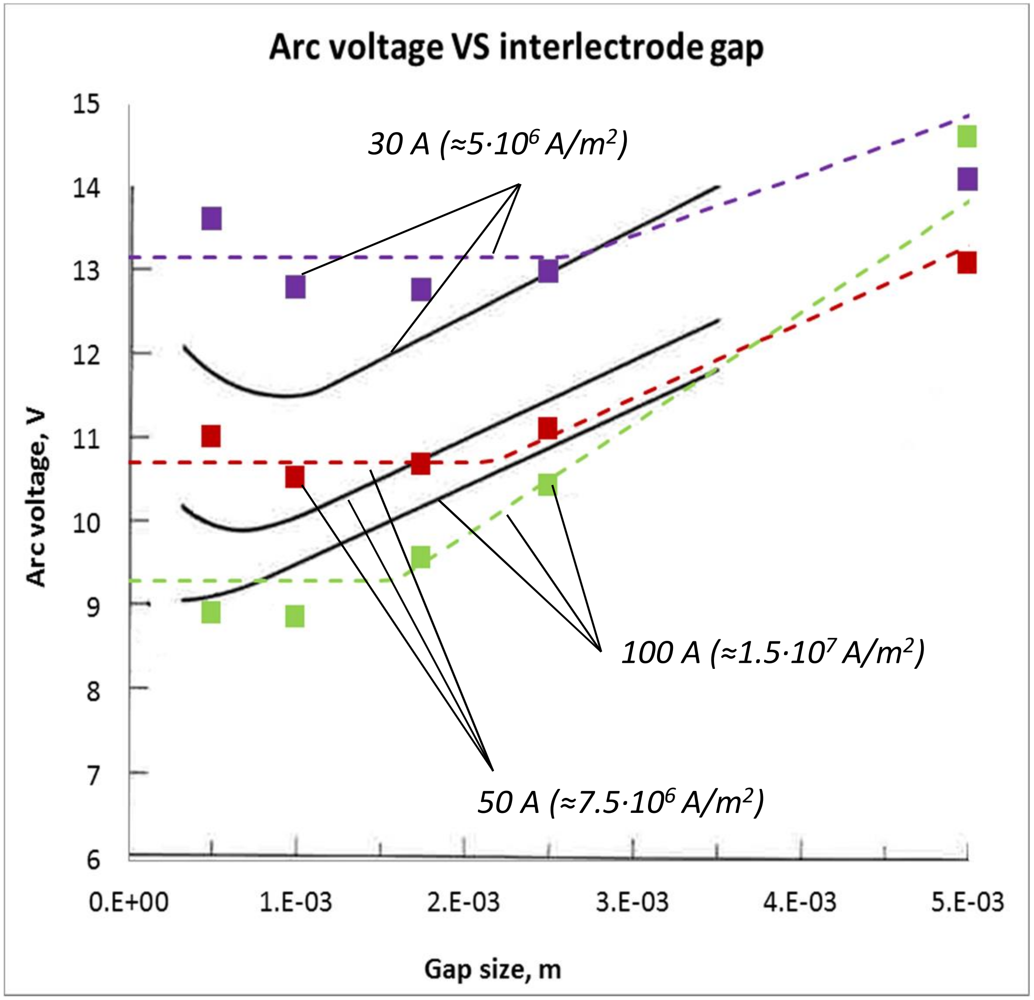
Arc column: $\frac{dV}{dx} = \frac{\rho_0}{T_e^{2.5}} \quad \frac{\rho_0 j}{T_e^{2.5}} = Q^{rad}(T_e, p)$ $\quad \rho_0 j \left(\frac{T_{e,0}}{T_e}\right)^{2.5} = 0.5\frac{E_{ion}}{T_e}\nabla T_e \quad \frac{dV}{dx} = 3.2\frac{k}{e}\frac{dT_e}{dx}$

Anodic region: $V_{anode} = V_{pre-sheath,a} + V_{sh,a}$ $\quad \frac{\sigma_{i,a}\sqrt{m_a}p^2}{j^2}T_{eq,a}^5 e^{eE_i/kT_e} = const$





Validation against experiment



Experiment

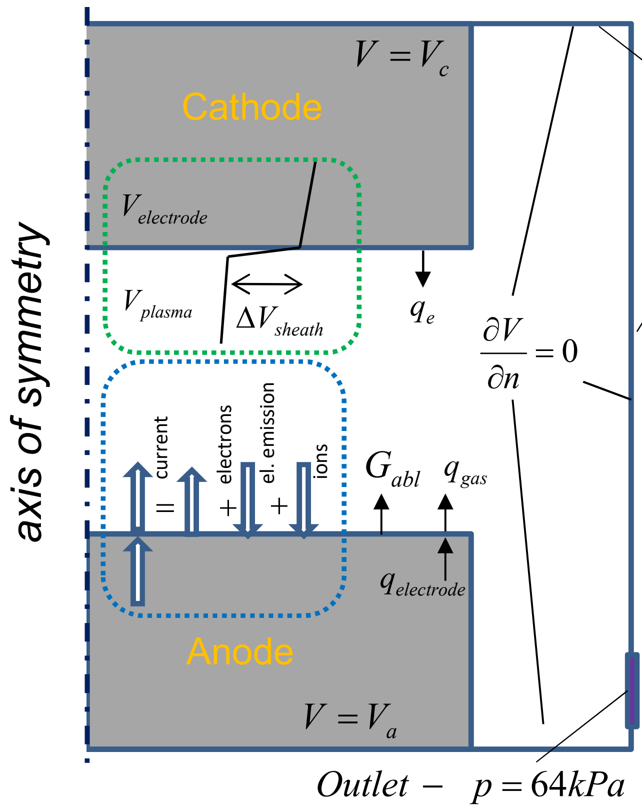
(Lancaster J.F., 1954, Brit. Weld. J., Vol. 1)

Analytical model

1D simulations

2D-axisymmetric simulations of carbon-helium arc, validation against experiments

2D setup: boundary and interfacial conditions



Chamber walls:

$$\vec{v} = 0,\ \frac{\partial V}{\partial n} = 0,\ T = 350\,K,\ \frac{\partial T_e}{\partial n} = 0$$

Plasma-electrode interfaces:

Ablation-deposition:

$$G_{abl} = \left(p_{satur,C}(T) - p_C\right)\sqrt{M_C/(2\pi RT)},\quad p_C = n_C kT \text{ – partial pressure}$$

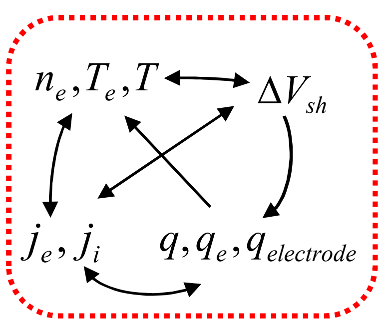
Electric current and potential:

$$V_{electrode} - V_{plasma} = \Delta V_{sheath}$$

$$j_{n,plasma} = j_{n,electrode}$$

$$j_{n,plasma} = j_e^{emission} - j_e^{plasma\ to\ electrode} + j_i = f(\Delta V_{sheath}, T, T_e, n_e)$$

Self-consisted model, parameters coupling at electrode surfaces:



Heat transfer:

Heat fluxes are computed with accounting for:

- ablation/deposition;
- emission;
- radiation;
- sheath contribution;
- work function;
- ionization energy.

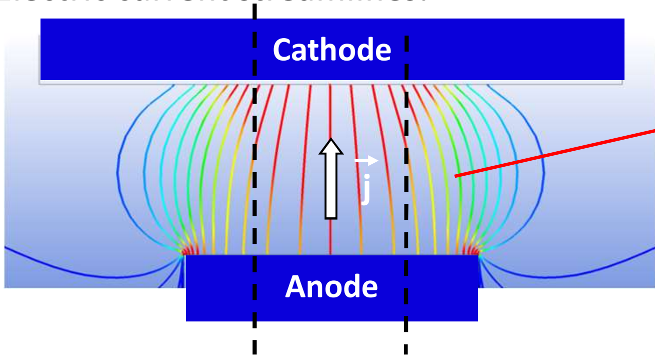
Automatic determination of the sheath sign.

! The parameters are non-uniform at the electrode surfaces

Flow pattern of electric current

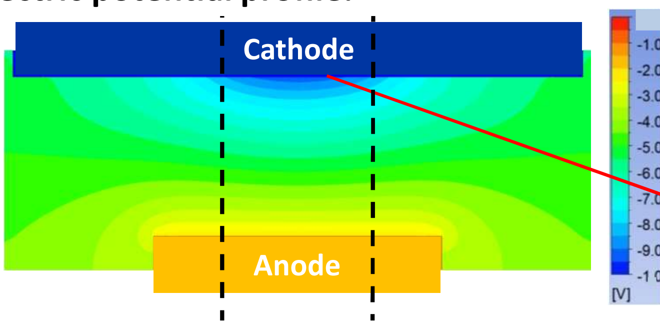
Simulations:

Electric current streamlines:



Arc contraction

Electric potential profile:

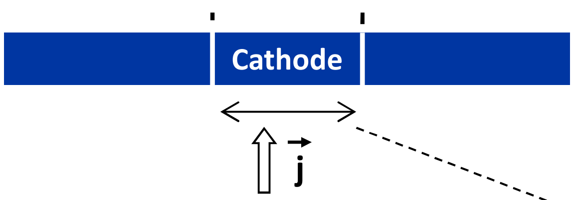


Arc channel

Non-uniform potential distribution along the cathode tip

Experiments with a segmented cathode:

(Y.-W. Yeh et.al., 2016, Carbon 105)

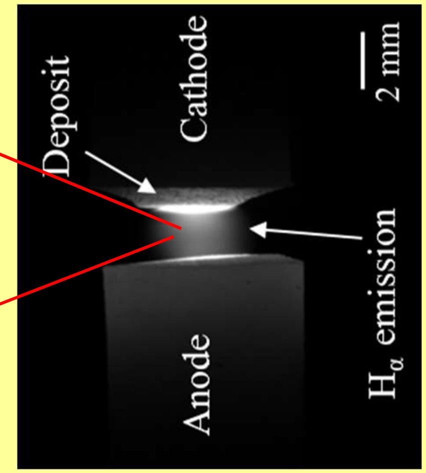


2 radial segments, central segment d=3.2mm, **95%** of current

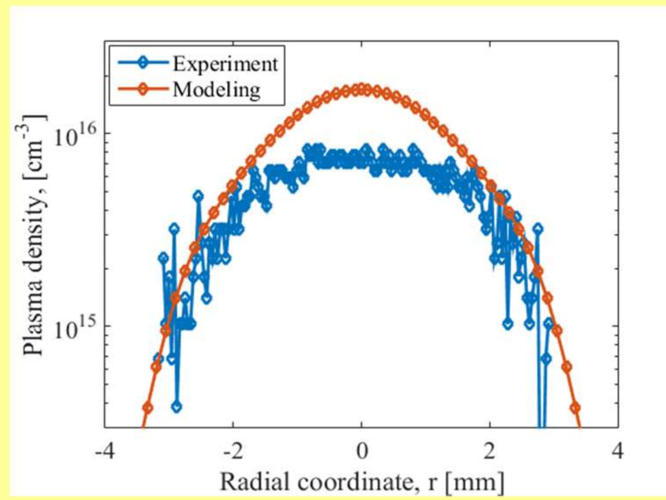
Simulations: **75%** of current went through 3.2mm segment

Experimental data:

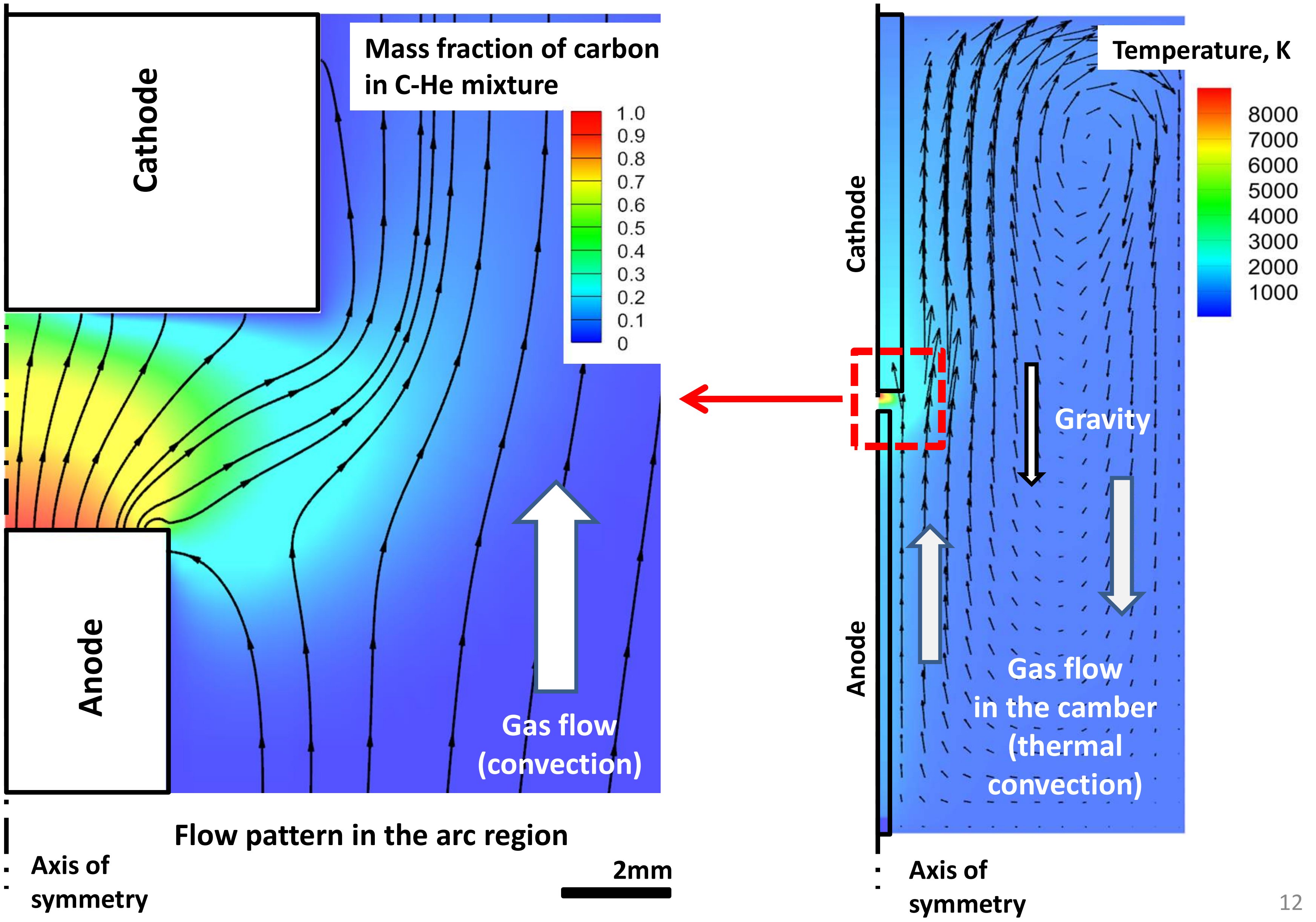
(V. Vekselman, ICOPS-2017, Session TU 2.3)



Middle plane plasma density profile:

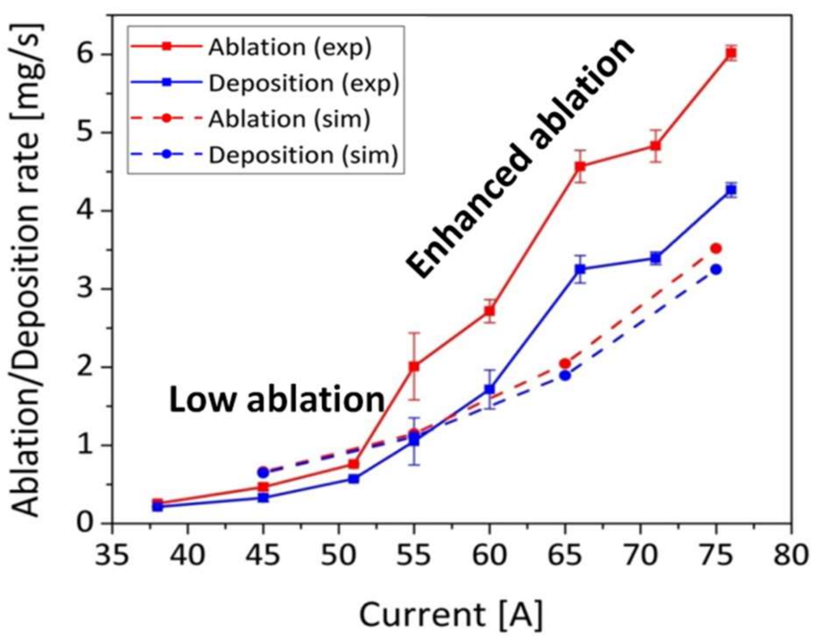


Transport of carbon in the arc



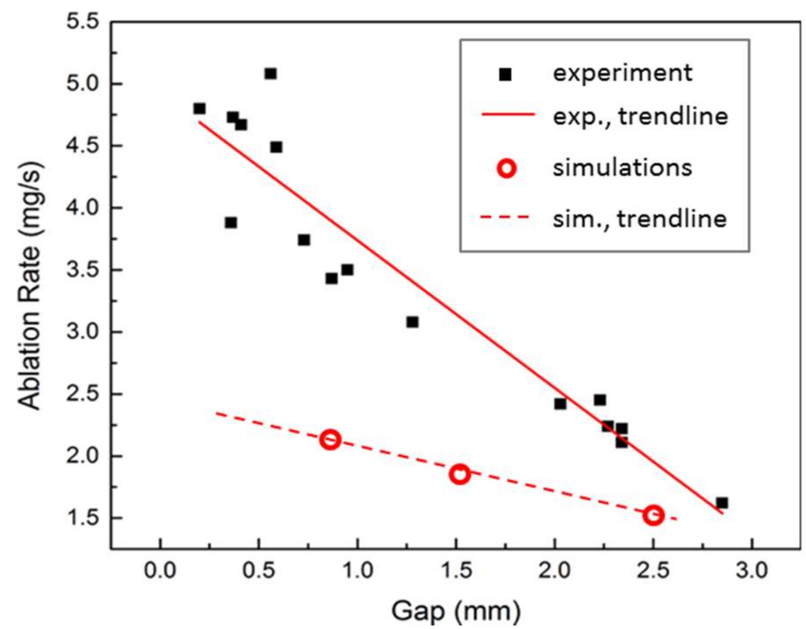
Ablation/deposition rates

Constant inter-electrode gap 1.5 mm, various currents:



V. Vekselman et.al.,
ICOPS-2017, Session TU 2.3

Constant current 65 A, various inter-electrode gap sizes:



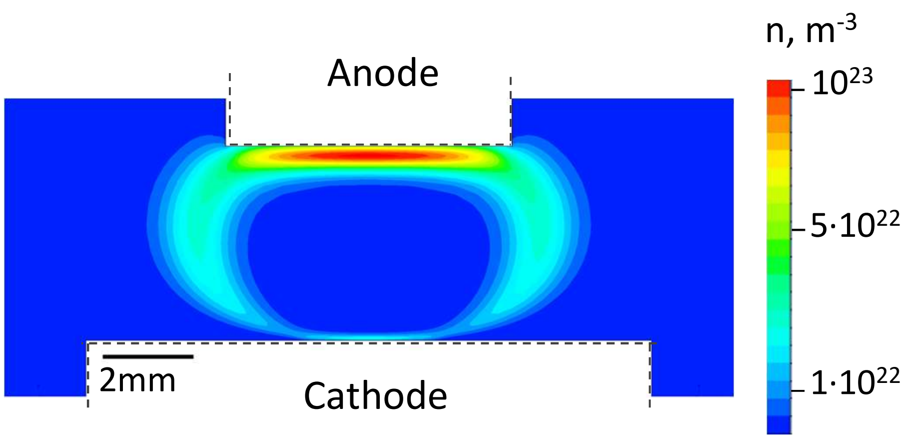
T. Huang et.al.,
Session TU Posters: TU P3

Density profile of C_2 molecules

Simulations:

Equilibrium carbon chemistry
(Wang et.al., 2011, J. Phys. D: Appl. Phys. 44),
transport coefficients for C_1

Profile of number density of C_2 molecules:

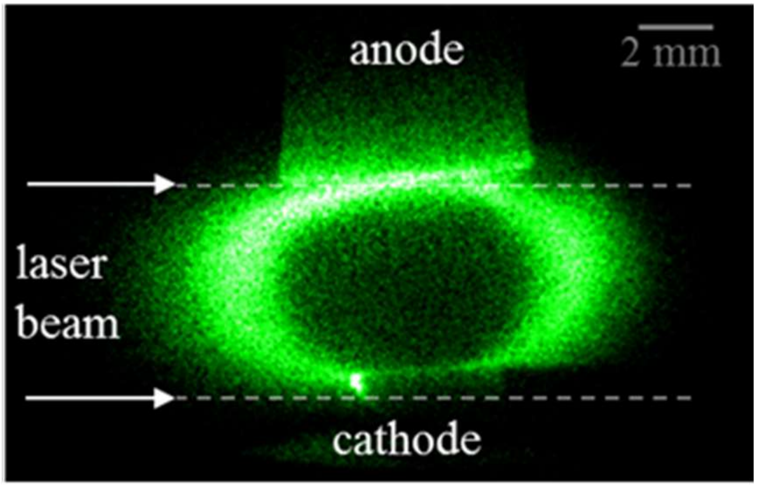


Experiments:

(V. Vekselman et.al., ICOPS-2017, Session TU 2.3)

Planar LIF: spectral image of carbon dimer (C_2)

Emission at 470 nm (laser at 437 nm)



- **Carbon dimer distribution has a bubble-like shape around the arc core**

Summary

Self-consistent model of carbon arc discharge in helium atmosphere was implemented into a general purpose code ANSYS-CFX which was highly customized for this purpose.

The arc model was benchmarked against simulations of N. Almeida et.al. 2008 and analytical solution.

2D-axisymmetric simulations of the discharge at various arc currents and inter-electrode gap sizes were performed and compared with experimental data.

Good quantitative agreement on the width of arc channel, plasma density and qualitative agreement on ablation rate were obtained.

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Thank you!