Validated modeling of plasma boundary physics coupled to surface response for atmospheric arcs

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Motivation: What is common between arc and divertor.

- Arc and divertor plasmas: heat fluxes and plasma-wall response play crucial role.
- Plasmas are longer than mean free path, but sheath is collisionless.
  - Both can be described by fluid equations with proper boundary conditions
- Wall ablation is strongly nonlinear phenomena.
  - Predict wall ablation is difficult task.
- Both processes need strong modeling validation and benchmarking to be predictive.
  - Talk will give such an example of validated modeling.

This work is funded by the Department of Energy, Office of Science, Fusion Energy Sciences.
Validation made possible by Laboratory for plasma nanosynthesis  http://nano.pppl.gov

Experimental details will be given by Yevgeny Raitses  NM9.00005 : Interaction of boron and nitrogen-rich plasmas with tungsten wall 11:00 AM–11:25 AM

Nano lab
In-situ characterization of plasma and nanoparticles

Laser-Induced Fluorescence (LIF) to measure T and n of C₂, C₃, B, BH, etc. Resolution: 10 ns, 100 µm

Laser-induced incandescence (LII) to detect nanoparticles of > 10’s nm Resolution: 10 ns, mm’s

Ex-situ characterization of nanomaterials by SEM, EDS, TEM, Raman spectroscopy, XRD, etc.

Experimental details will be given by Y. Raitses NM9.5 11:00 AM
Outline Arc model: towards fully self-consistent, non-equilibrium model with gas flow and chemical composition

- Benchmarking of the model
- Modeling results in comparison with experimental data
  - Arc column width
  - Gas flow pattern
  - Ablation rate
  - Chemical composition
  - Carbon nanoparticles growth

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Processes in the arc

Collisionless sheaths

(Debye radius < Mean free path ≈ 1 μm << Gap size)

Carbon deposit

Space-charge sheaths

Carbon transport

Anode ablation

Growth of nanoparticles

Convection in the chamber

Chamber

He 500 Torr

Graphite

50 A – 70 A

Anode

6 mm

Cathode

9 mm

1-4 mm
The non-equilibrium fluid arc model was implemented into ANSYS-CFX code. Model includes self-consistent electron current, sheaths, thermal diffusion, heat fluxes to electrodes and gas convection.

The biggest challenge: strongly-nonlinear coupling of the heat and particle fluxes are at the plasma-electrode interfaces, which makes fast convergence a problem.

For more details please attend:
- GEC TF2.04, Y. Raitses, *Towards understanding of plasma-based synthesis of carbon nanomaterials*, Friday, 10:30 AM, (invited)
Fluid arc plasma model

Fluid motion and species transport:
Momentum equation:
\[ \nabla \cdot (\rho \vec{v}) = -\nabla p + \nabla \cdot (\mu \nabla \vec{v}) + \rho \vec{g} \]
Continuity equation:
\[ \nabla \cdot (\rho \vec{v}) = 0 \]
Neutrals transport equation:
\[ \nabla \cdot (\rho n_a \vec{v}) = \nabla \cdot (D \nabla n_a) + S \]
Equation of state:
\[ p = (n_{neutrals} + n_i)kT + n_e kT_e \]

Ionization non-equilibrium:
Ions transport equation:
\[ \nabla \cdot (n_i \vec{v}) = \nabla \cdot \left( D_a \nabla n_i + D_{th \ diff} \nabla T + D_{th \ diff, e} \nabla T_e + \vec{j}_e \sigma_{e,i} \right) + S_i, \quad S_i = \alpha n_e n_a - \beta n_e^3 \]

Thermal non-equilibrium:
Energy balance of electrons:
\[ \nabla \cdot \left( (2.5 + A_e)kT_e \frac{\vec{j}_e}{e} \right) = \nabla \cdot (\lambda_e \nabla T_e) - S_i E_i - Q^{electrons-heavy} - Q^{rad} + \vec{j}_e \cdot \vec{E} \]
Energy balance of heavy particles:
\[ \nabla \cdot (\rho \vec{v}) = \nabla \cdot (\lambda \nabla T) + \frac{\partial p}{\partial t} + Q^{electrons-heavy} + \vec{j}_i \cdot \vec{E}, \quad h = \int C_p dT + \frac{v^2}{2} \]

Generalized Ohm’s law:
Transport of electrons:
\[ \frac{\vec{j}_e}{\sigma} = \vec{E} + \frac{k}{e} \left( C_e \nabla T_e + T_e \nabla \ln n_e \right) \]
Diffusion and thermal diffusion of electrons
Current conservation:
\[ \nabla \cdot (\vec{j}_e + \vec{j}_i) = 0 \]
Gauss’s law:
\[ e(n_e - n_i) = \varepsilon_0 \nabla \cdot \vec{E} \]
Quasi-neutrality (simplification):
\[ n_e = n_i \]
Conditions at plasma-electrode interfaces

\[ j_i = e n_i v_{th,i} \exp\left(-e \max(\Delta V_{sheath}, 0) / kT\right) \]

\[ j_e = e n_e v_{th,e} \exp\left(e \min(\Delta V_{sheath}, 0) / kT\right) \]

\[ j_{e,\text{emiss}} = A T^2 \exp\left(-e (V_w + \max(\Delta V_{sheath}, 0)) / kT\right) \]

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

The parameters are non-uniform at the electrode surfaces

Automatic determination of the sheath sign.
Benchmarking of the arc model: 1D modeling and analytical solution for a tungsten-argon arc
1D modeling of argon arc

Plasma density profiles at various current densities:
- $j = 2.5 \cdot 10^6$ A/m$^2$
- $j = 5 \cdot 10^6$ A/m$^2$
- $j = 7.5 \cdot 10^6$ A/m$^2$
- $j = 10^7$ A/m$^2$

Electric potential profiles:

Electric current composition:

- Non-equilibrium effects are crucial in major part of the arc
Benchmarking of 1D simulations


\[ P = 1 \text{ atm} \]
\[ j = 5 \cdot 10^6 \text{ A/m}^2 \]
\[ T_{\text{cathode}} = 3500\text{K} \]

Boundary conditions (collisionless sheath):

\[ j = j_{e,c}^P + j_{e,c}^{\text{emiss}} + j_{i,c} \]
\[ j_{i,c} = e n_i \sqrt{\frac{k(T_e + T)}{m_i}} \]
\[ j_{e,c}^P = e n_e \frac{1}{4} \sqrt{\frac{k T_e}{m_e}} e^{-\frac{V_{sh,c}}{k T_e}} \]
\[ q_{e,c} = j_{e,c}^{\text{emiss}} (V_{sh,c} + 2.5 T_e) - j_{e,c}^P (V_{sh,c} + 2.5 T_e) \]

The results are published in A. Khrabry et al., PoP 25, 013521.
Analytical solution for the near-cathode region

Energy balance:

**Cathode**
- Thermal conduction into the cathode body
  \[ q_{\text{c,h.cond.}} \]

**Sheath**
- Ionization layer
  \[ J_i (E_{\text{ion}} + 2T_c) \]
- Thermal conductivity

**Ionization layer**
- Electron emission – \( J V_w \)

**Plasma bulk**
- 3.2 JT

**3.2 J**

\[ q_{\text{to cathode}} = \varepsilon_{\text{ion}} J_i \]

\[ q_{\text{to cathode}} = q_{\text{c,h.cond.}} (T_c) + J V_w \]

**Cathodic voltage**

\[ j V_{\text{layer}} = q_{\text{to cathode}} + 3.2 k T_{\text{plasma}} \]

\[ j \approx j_{\text{emiss}} + j_i \]

\[ j_{\text{emiss}} = A T^2 \exp\left(-\frac{e V_w}{k T}\right) \]

Earlier:

\[ q_{\text{to cathode}} = E_{\text{ion}} J_i \]

\[ E_{\text{ion}} = 15.76 \text{V} \]

\[ E_{\text{ion}} = 40 \text{V} \]

\[ E_{\text{ion}} = 50 \text{V} \]

**1D simulations**

- Ion current to the cathode can be determined using ionization cost \( \varepsilon_{\text{ion}} = 50 \text{V} \)
Near-cathode region: thickness, temperature

Simplified ion transport equation:

\[
\frac{d}{dx} \left( D \frac{dn_e}{dx} \right) = k_r n_e^3 - k_i n_a n_e
\]

- \( k_r = \text{const}, \ k_i = \text{const}, \ D = \text{const}, \ n_a = \text{const} \)

Electron temperature in the layer:

\[
j_{i,e} = e k_i n_a \sqrt{\frac{D}{2k_r}} \quad \rightarrow \quad T_e = \frac{T_i - 0.5T_c}{\ln \left( \frac{j_{i,e} \sqrt{A_i}}{e A_i} \frac{8 m_e k \sigma_{\text{ion}}}{\pi} \frac{4}{p} \sqrt{T_c} \right)}
\]

- \( k_i(T) = A_i \exp(T_i / T) \)
- \( k_r(T) = A_r \exp(T_e / T) \)

(Benilov M.S., J. Phys D 33 (2000) 960)

Ionization layer width:

\[
L_i = \sqrt[2]{\frac{D}{2n_a k_i}} \text{atanh}(1 - \varepsilon)
\]

(Tolerance \( \varepsilon \approx 1\% \))

- Parameters of the near-cathode region are self-consistently determined analytically

Electron temperature near the cathode:

- Analytical model
- Simulations

Width of the near-cathode ionization layer:

- P = 1 atm
- P = 3 atm
Temperature profiles at various current densities:

- $j = 2.5 \times 10^6$ A/m²
- $j = 5 \times 10^6$ A/m²
- $j = 7.5 \times 10^6$ A/m²
- $j = 10^7$ A/m²

**Analytical solution for the arc column:**

1. **Complete equilibrium:**
   Joule heating = radiation loss
   \[
   \frac{j^2}{\sigma} = Q_{rad}(T) \quad (1)
   \]
   \[
   \sigma = CT^{2.5}
   \]
   (Highly ionized plasma, L. Spitzer, 1962)

2. **Closer to the anode (local equilibrium):**
   Temperature decreases $\rightarrow$ radiation loss is low
   Temperature gradients are not too high $\rightarrow$ thermal conductivity and convective heat transfer are minor effects
   - **Electric current is driven merely by diffusion:**
   \[
   \frac{j}{\sigma} = \frac{k}{eT} \frac{dn_e}{n_e dx}
   \]
   With Saha relation for $n_e$:
   \[
   \frac{j}{\sigma} = \frac{j}{C T^{2.5}} = 0.5 E_{ion} \frac{dT}{T dx}
   \]
   **Solution:**
   \[
   T = \left( \frac{5j}{C E_{ion}} x + \text{const} \right)^{2/5} \quad (2)
   \]
Analytical solution for the near-anode region

Departure from ionization equilibrium

Ion diffusion + electron transport + energy balance

\[ \left( \frac{n}{n_{\text{Saha}}} \right)^2 \approx 1 + \frac{C_1}{k_i(T) PT^{2.5}} \]

\[ k_i(T) = A_i \exp \left( \frac{T}{T_i} \right) \]

Ion current to the anode:

\[ n_{\text{ref}} = \sqrt{2} n_{\text{Saha}}(T_{\text{ref}}) \]

\[ \Gamma_{i,a} \approx C_2 \frac{n_{\text{ref}}}{T_{\text{ref}}} \frac{2eE_{\text{ion}} - kT_i + 4kT_{\text{ref}}}{2eE_{\text{ion}} - kT_i - 6kT_{\text{ref}}} \]
Voltage in the the near-anode region

Electric potential profile (results of the simulations):

Anode region voltage. Numerical simulations VS analytical solution:

- **Anodic voltage, hot anode**
- **Anodic voltage, cold anode (1000 K)**

- Volt-Ampere characteristic of the region is dependent on the anode cooling mechanism
Validation against experiment

Argon arc between cylindrical tungsten electrodes 3mm in diameter

Experiment

Analytical model

1D simulations
2D simulations of C-He arc using customized ANSYS-CFX code

- GEC TF2.04, Y. Raitses, *Towards understanding of plasma-based synthesis of carbon nanomaterials*, Friday, 10:30 AM,
Flow pattern of electric current

Simulations:
- Electric current streamlines:
  - Arc contraction
  - Middle plane plasma density profile:
  - 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

Good agreement with experiments on the arc channel width
Transport of carbon in the arc

Flow pattern in the arc region

Mass fraction of carbon in C-He mixture

Temperature, K

Gravity

Gas flow in the camber (thermal convection)

Gas flow (convection)

Axis of symmetry

Anode

Cathode
Ablation/deposition rates

Constant inter-electrode gap width 1.5 mm, various currents:

- Rates of the anode ablation and carbon deposition at the cathode depend on both arc current and inter-electrode gap.
- The rates are strong function of the arc current; this behavior is captured by the simulations.
Carbon chemistry and growth of nanoparticles

Carbon mixture composition (from Gibbs free energy minimization):

Simple agglomeration model:
\[ \frac{dn}{dt} = -D^2 n^2 \sqrt{\frac{4\pi kT}{m}} \]

\[ D = 2r_0(n_0/n)^{1/3} \]

\( n_0 \) – initial density, \( r_0 \) – carbon lattice step.

Axis of symmetry

Cathode

Anode
Density profile of C$_2$ molecules

**Simulations:**
Carbon dimer profile - a bubble-like shape around the arc core.

**Experiments:**
(Vekselman, Khrabry et al., PSST 27 (2018))
Planar LIF technique.

- Arc current 50 A

- Good qualitative and quantitative agreement with the experimental data
Region of the nanoparticles growth

Location of the nanoparticles measured using planar LII

S. Yatom, A. Khrabry et al., MRS Communications (2018)

- Good qualitative and quantitative agreement with the experimental data
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 extensively verified and validated against previous simulation results and experimental data.
- Effective sheath boundary conditions were verified against sheath resolving approach.
- Analytical model of a short arc was developed. Useful relations for voltages and temperatures in various arc regions and heat fluxes to the electrodes were derived.
- Good agreement was obtained for simulated arc characteristics as: voltage, channel width, plasma density, ablation/deposition rates, $C_2$ molecules density profile, size and location of synthesized nanoparticles.

- GEC TF2.04, Y. Raitses, Towards understanding of plasma-based synthesis of carbon nanomaterials, Friday, 10:30 AM, (invited)
- DPP JP11.124, A. Khrabry, Self-consistent modeling of highly-collisional plasma interacting with electrodes, P. S. IV, Tuesday
- GEC LW1.1, A. Khrabry, Nanoparticles growth regions in carbon arc: simulations and experiments, Poster session II, Wednesday