Comsol Multiphysics
在低溫電漿模擬之應用
~My Little Journal with Simulation

『多重物理CAE分析軟體COMSOL Multiphysics CONFERENCE』用戶研討會

Nov 9, 2012

台灣大學化工系
徐振哲

Start of "Plasma"

OSCILLATIONS IN IONIZED GASES

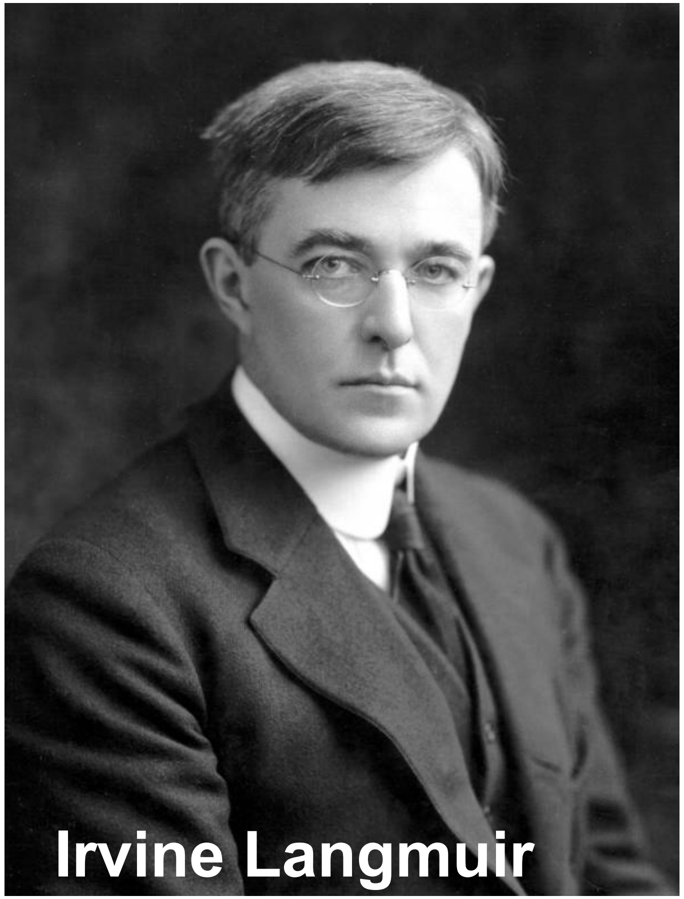
By Irving Langmuir

Research Laboratory, General Electric Co., Schenectady, N. Y.

Communicated June 21, 1928

gas contains ions and electrons in about equal numbers so that the re-
sultant space charge is very small. We shall use the name *plasma* to describe this region containing balanced charges of ions and electrons.

For purposes of calculation we may consider the plasma to consist of a



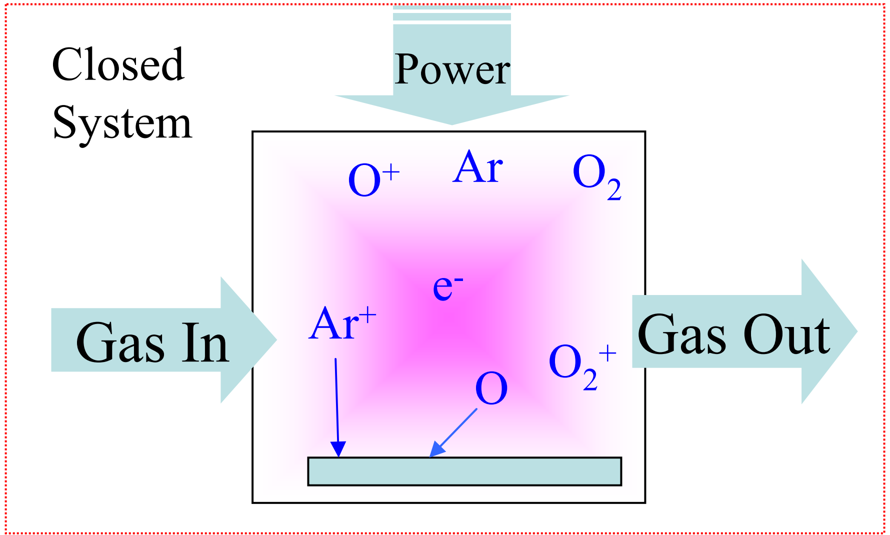
Plasma: ionized gas with equal amount of positive and negative charges.

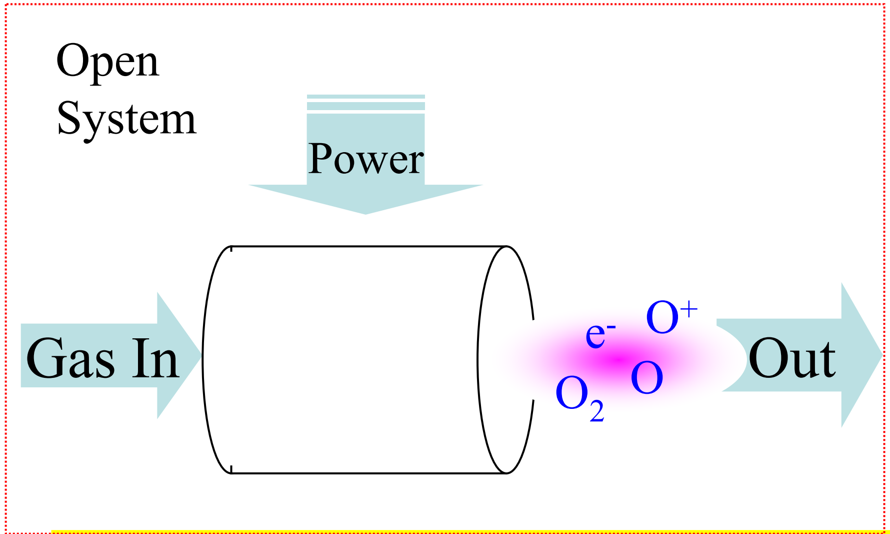
1928: Named plasma

1932: Nobel prize in Chemistry "for his discoveries and investigations in surface chemistry."

I. Langmuir, Proc. Natl. Acad. Sci. U. S. A. (1928)

Key Process Features





Kinetics Path

- e-: “heated” by the power, initiate reactions in parallel, e.g. ionization, dissociation, etc.
- Reactive neutral species: gas-surface or gas-gas reactions
- Ions: sometimes energetic.

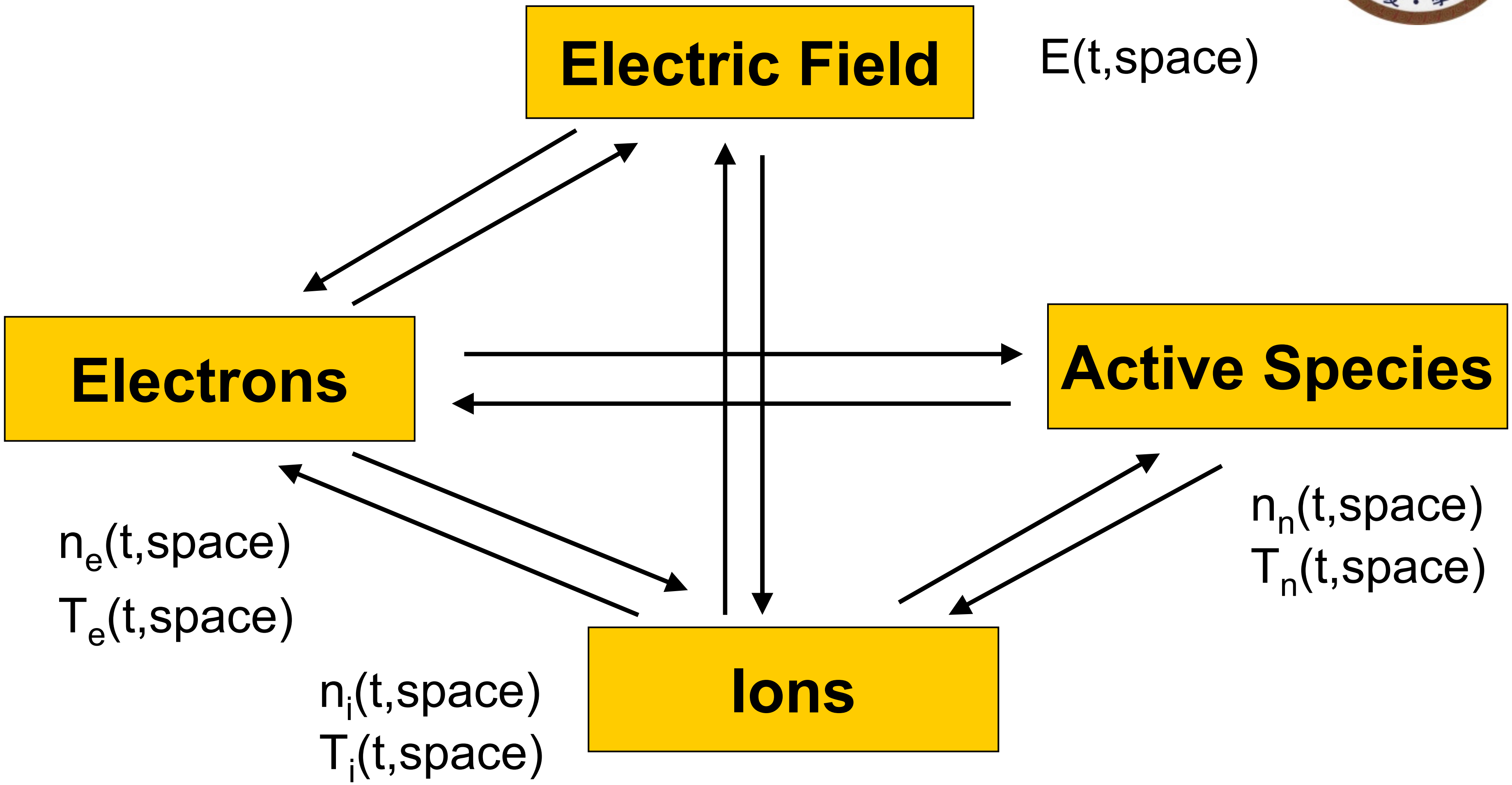
Key Process Parameters

- Operating pressure and atmosphere
- Type of power

Plasma Characteristics

- Species temperature, density and their distribution in time and in space.

Why Complicated

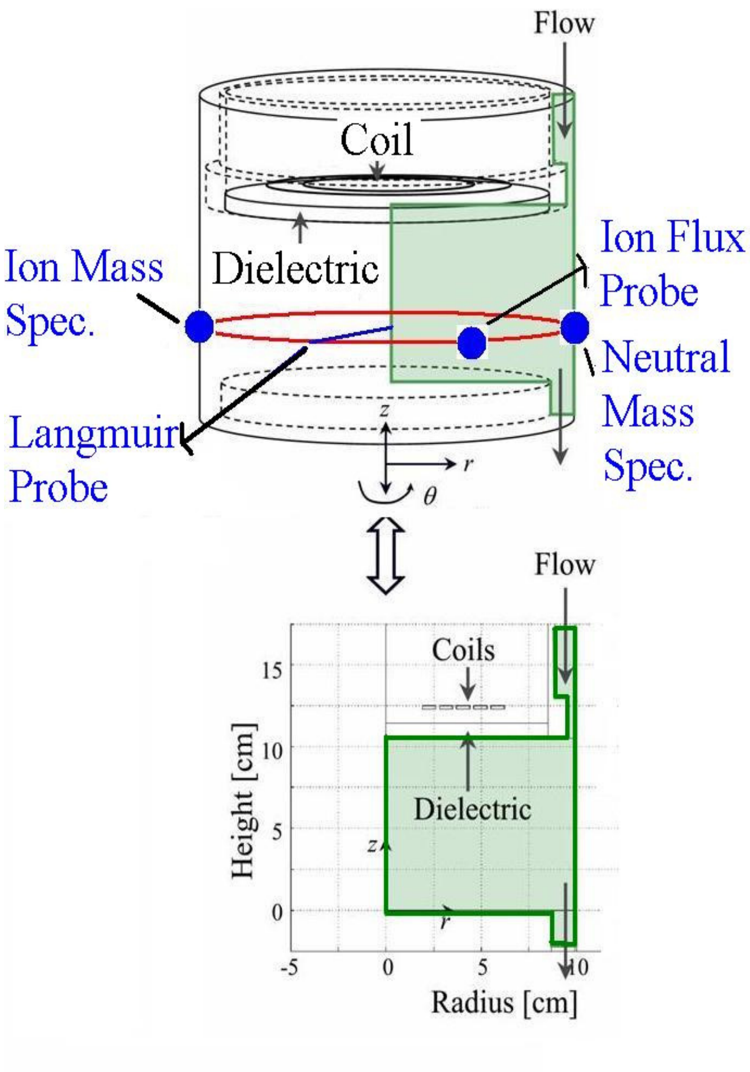


and Multiple species

My Little Journey of Simulation

2002~2006 @ UC Berkeley
Femlab 2.0 ~ Femlab 3.2
Modeling of Low Pressure Inductively Coupled Plasmas

System Description



Experimental System

- Diagnostic ICP
  - Multiple Diagnostics
  - Fits 6-in wafer
  - Well-defined boundaries
  - Stainless steel walls and dielectric top plate
  - Axisymmetric

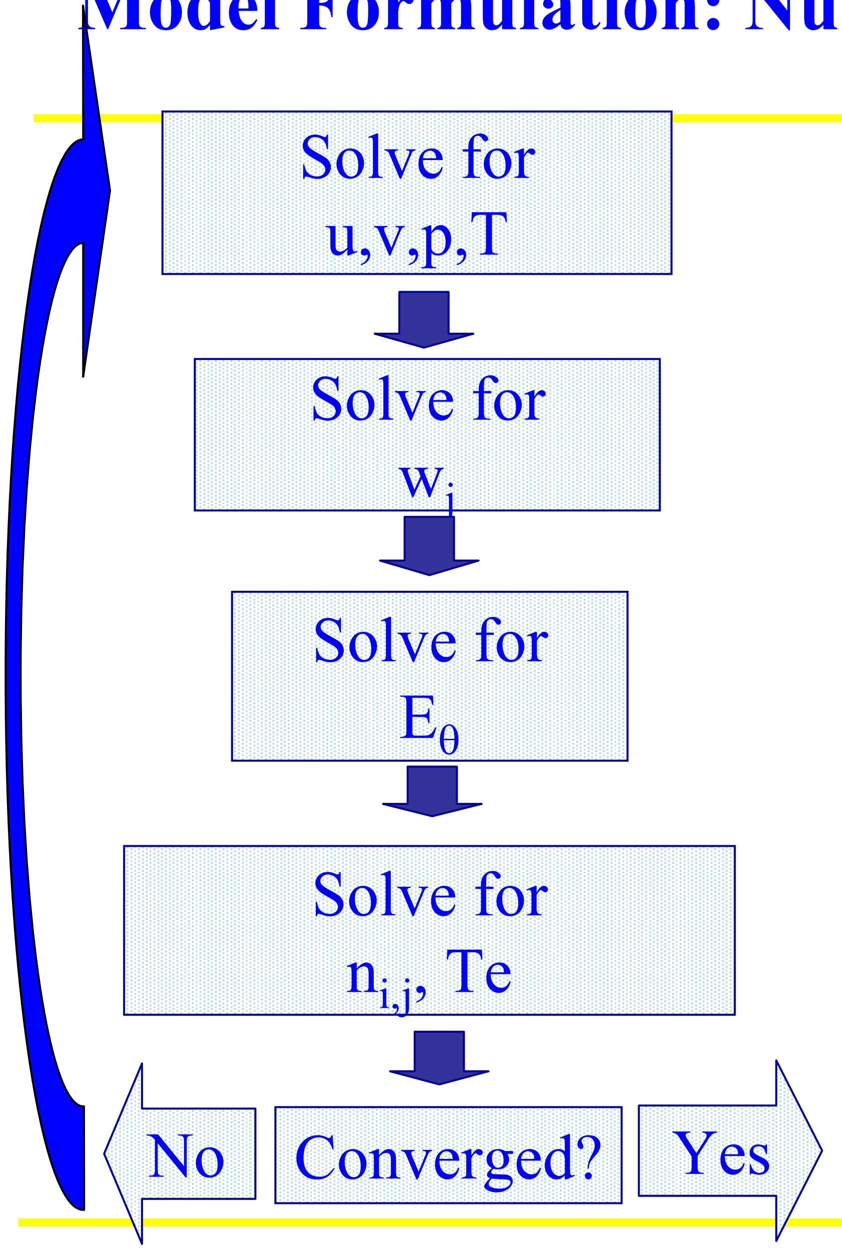
Model

- 2D, Fluid Model
- Ar/O_2 ICP as the preliminary test
- Femlab™ and Matlab™
  - Coupled neutral and plasma model
  - Easy to share / access
  - Easy to extend

Model Formulation: Equation System

Overall Neutral Continuity (p)	$\nabla\cdot(\rho\vec{v}) = R_n$
Neutral Species j mass balance (wj)	$\nabla\cdot\left(w_j\rho\vec{v} - \rho \mathrm{D_j}\nabla wj\right) = r_j$
Neutral Momentum Balance (u,v)	$\left[\nabla\cdot\bar{\bar{\boldsymbol{\tau}}}\right] = -\nabla p - \nabla\cdot(\rho\vec{v}\vec{v})$
Neutral Energy Balance (T)	$(\nabla\cdot\vec{q}) = -\nabla\cdot(\rho\vec{v}C_vT) - p(\nabla\cdot\vec{v}) + S_n$
Ion Continuity ($n_{i,j}$, $n_{ineg,j}$)	$\frac{\partial n_{ij}}{\partial t} + \nabla\cdot(-D_{ij} * (\nabla n_{ij} \pm \frac{\mathrm{T}e}{\mathrm{T}i} n_{ij}\frac{\nabla n_e}{n_e})) = r_{ij}$
Electron Energy (Te)	$\frac{\partial}{\partial t}\left(\frac{3}{2}n_e e\mathrm{T}_e\right) + \nabla\cdot\vec{\mathbf{Q}}_e = -e\vec{\mathbf{E}}\cdot\vec{\mathbf{\Gamma}}_e - E_e + P_{abs}$
Holmholtz Wave Equation (E_θ)	$(\nabla^2 E)_\theta = \frac{\omega^2}{c^2}K\cdot E_\theta - i\omega\mu_0 J_{ext,\theta}$

Model Formulation: Numerical Scheme



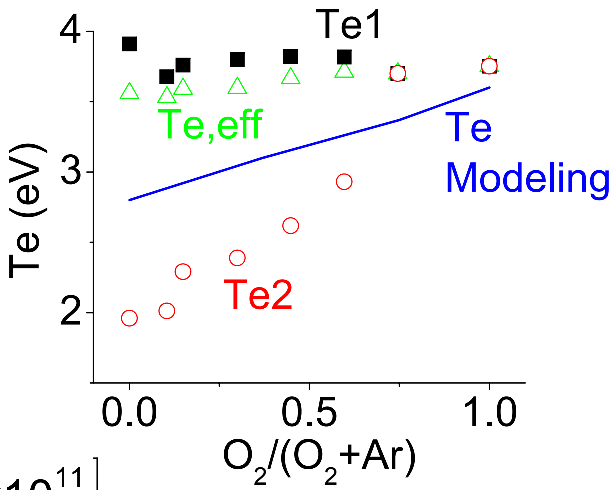
- Able to handle over 9 neutral species, and 8 charged species (totally 22 equations) with PC (~1GB memory).
  - 6 neutral ,4 ions species and 15 equations in current Ar/O_2 model.
- Convergence:
  - one iteration < 20min. Need < 10 iterations.
  - Robust, and easy to converge.

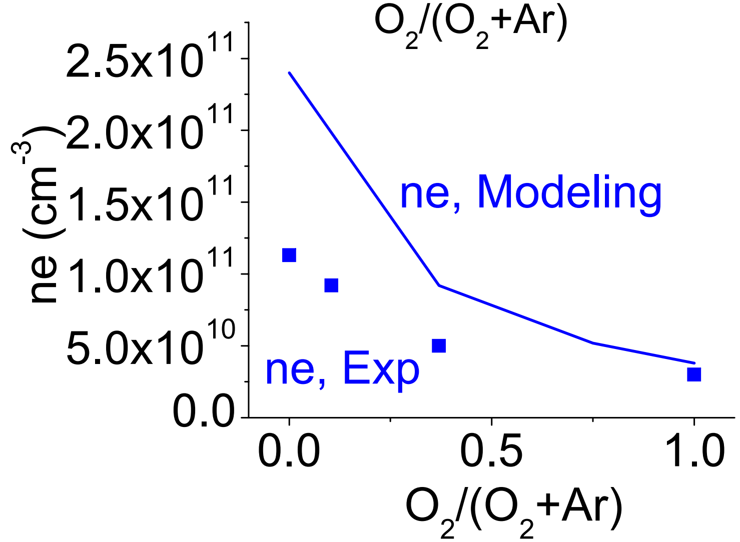
Model Formulation: Ar/O_2 ICP Details

- Chemistries
  - Neutral Species: ground state Ar, O_2, O, and metastable $O_2(a^1\Delta, b^1\Sigma)$ and $O(^1D)$
  - Ion Species: Ar^+, O_2^+, O^+, O^-
  - Major reactions: Ionization, excitation, dissociation, electron impact attachment, charge exchange. All cross sections and rate coefficients were taken from the literature.
- Model and Experiment Comparison
  - n_e profile, center n_e, and T_e,
  - n_O , total ion flux and composition at the wall.

Validation: n_e and T_e: Ar/O_2 Plasmas

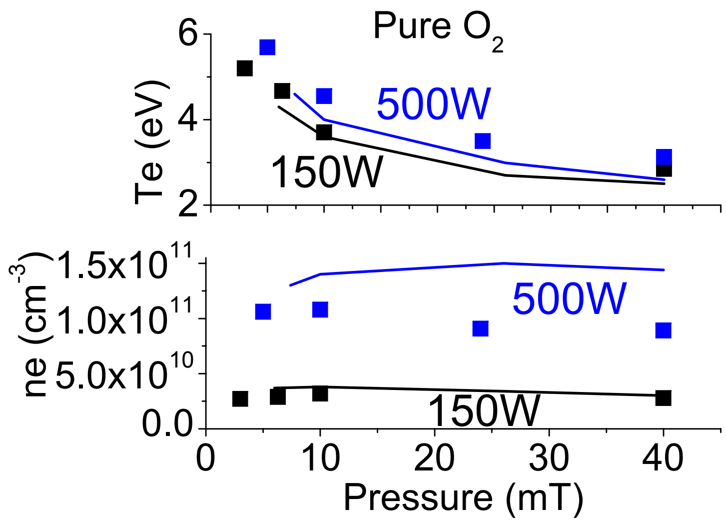
150W, 10mT





- Maxwellian EEPF at $O_2/(O_2+Ar) > 0.75$.
  - Better T_e prediction
  - Better n_e prediction

Good prediction in pure O_2 plasmas.



C.C. Hsu, et al. ,J. Phys. D-Appl. Phys., 39 (2006) 3272.

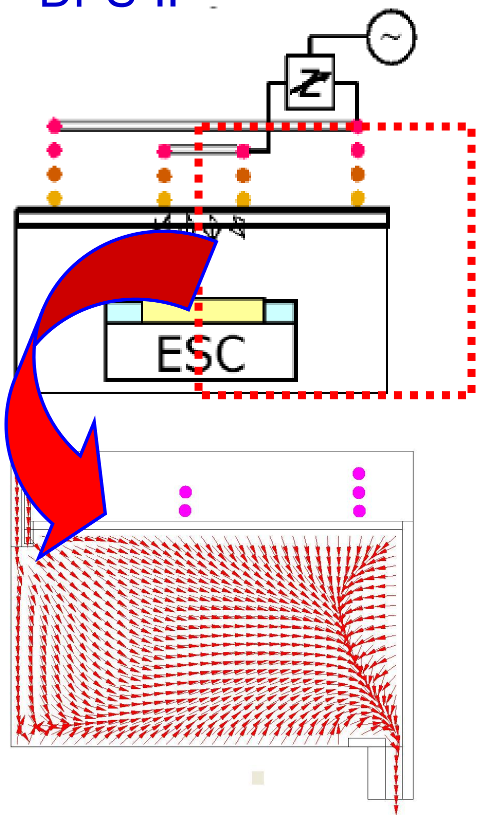
My Little Journey of Simulation

2006~2007 @ UCLA
Comsol Multiphysics 3.3
Solving an industrial problem

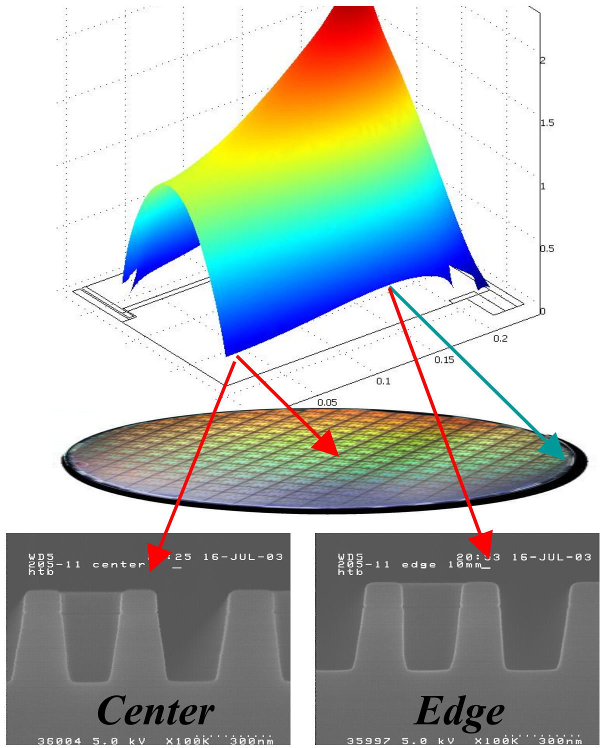
2006~2007 UCLA

Advisor: Dr. Jane P. Chang
Modeling Commercial Etching Tool to Explain Etching Behavior

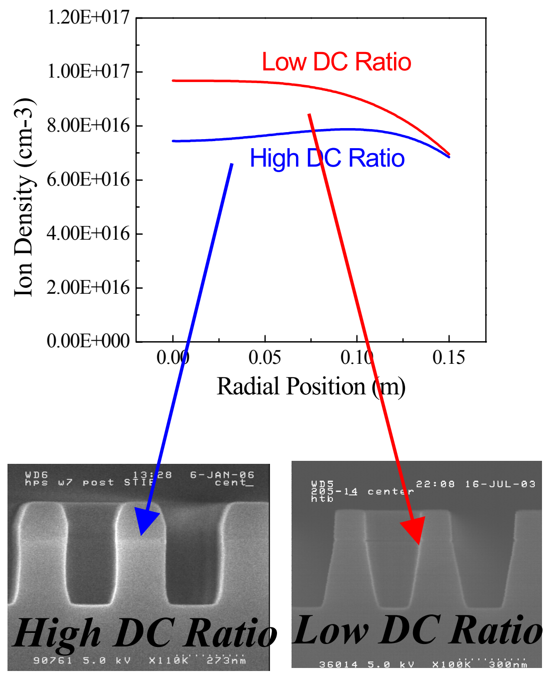
DPS-II



Spatial-Resolved Quantities: Neutral density, Ion flux, etc.



Identify Critical Parameters: Power, DC ratio….



Courtesy of Cypress Semiconductor

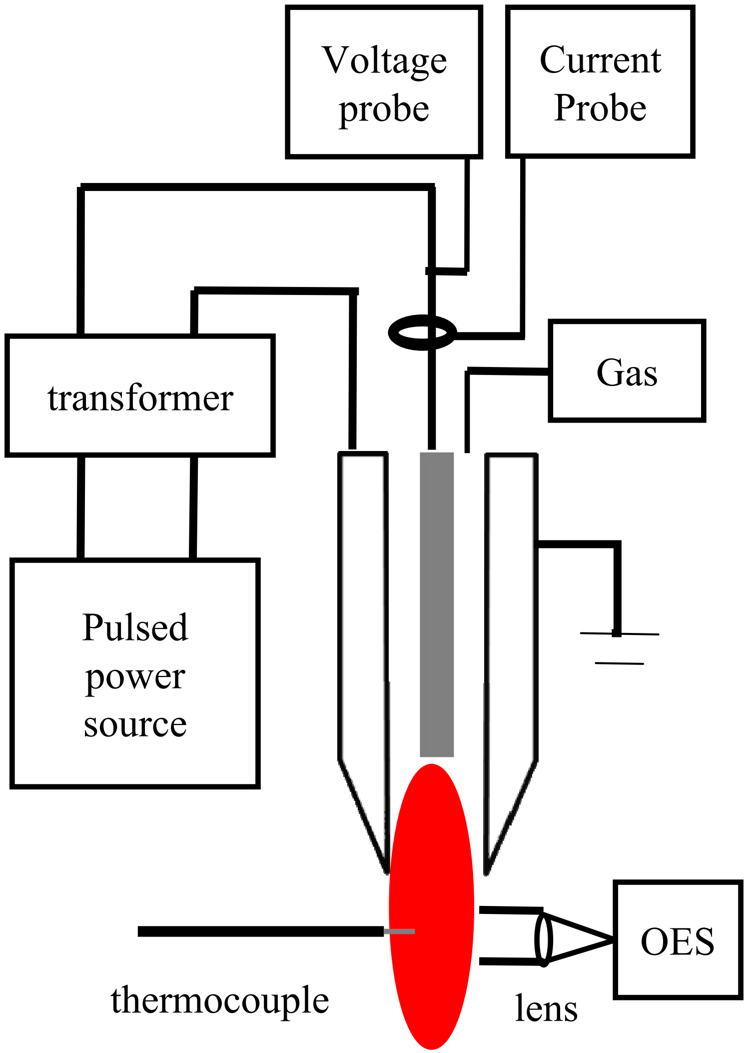
My Little Journey of Simulation

2007~ @ NTU

Comsol Multiphysics 3.5a

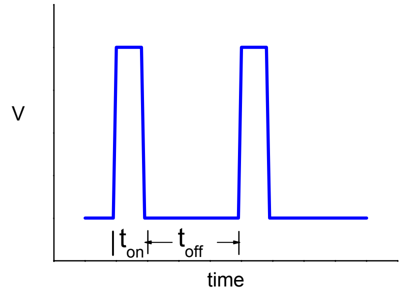
Modeling of Atmospheric Pressure Plasma Jet

Experimental Apparatus – Arc Plasma Jet



Power source:

- DC pulsed power, applied voltage (power source output) up to 350V.



t_{on} : pulse on time
t_{off} : pulse off time
Duty cycle : t_{on}/t_{off}
(25kHz, 8/32 μ s)

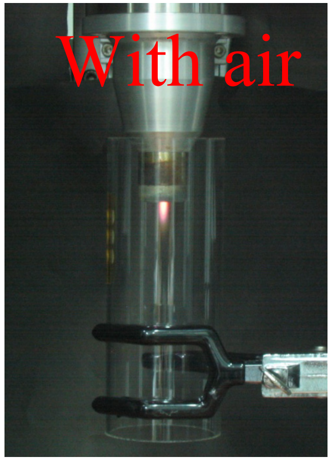
Process gases:

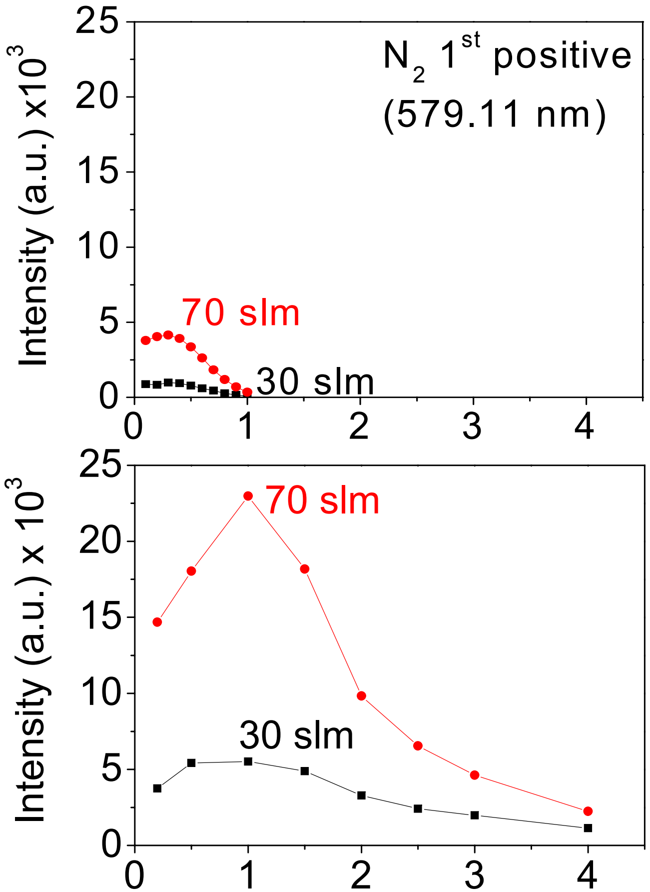
- N_2, (O_2, Air), with flow rate 10's slm.

Diagnostic tools:

- High voltage probe
- Current probe
- Optical Emission Spectrometer
- Thermocouple (Al_2O_3 covered)

Ambient Air Effects





- Without ambient air, the jet length and its width increase significantly, so does the emission intensity.
- Emission increases initially then decays with the increase of the distance
- High flow rate shows a higher reactivity regardless of the absence of the ambient air.

Reactivity: controlled by power density and the decay process
Ambient air diffusion significantly influence the plasma characteristics.

Equation System

Overall equation of continuity $\nabla \cdot \rho \vec{v} = R$

Momentum balance $\nabla \cdot \left(\rho \vec{v} \vec{v} \right) = -\nabla p - \nabla \cdot \vec{\tau}$

Species equation of continuity $\nabla \cdot \left(w_j \rho \vec{v} - \rho D_j \nabla w_j + w_j \rho \left(\sum_n D_j \nabla w_j \right) \right) = r_j$

Energy balance $-\nabla \cdot \left(\rho \vec{v} \hat{U} \right) = -\nabla \cdot \vec{q} - p \left(\nabla \cdot \vec{v} \right) + S_n$

Correction of the diffusivity $Sc^{(t)} = \frac{\nu^{(t)}}{D^{(t)}}$ $D_T \approx 0.009au$

Boundary conditions
mostly by convention: symmetric, no slip, convective flux, constant temperature

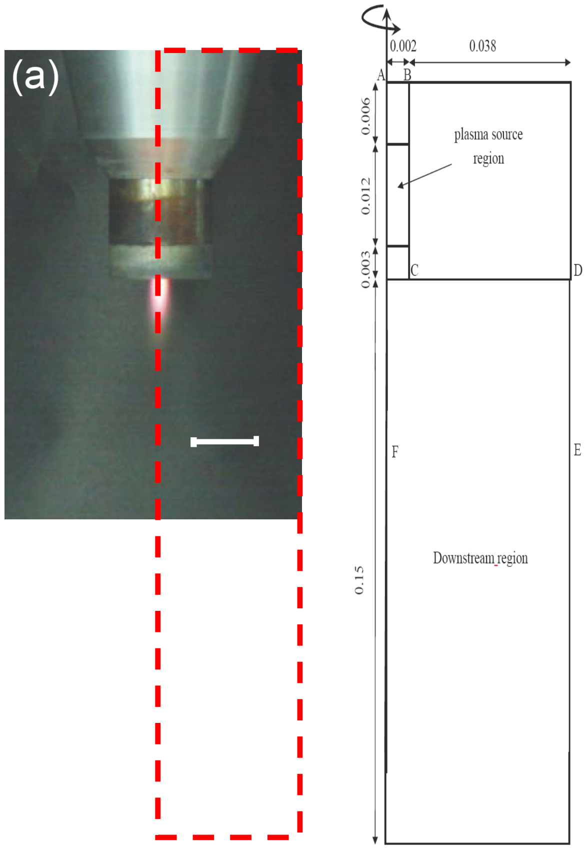
Model Chemistry

- **Upstream: electron-impact reactions and heavy particle reactions**
- **Downstream: heavy particle reactions – electrons are kinetically unimportant at the downstream.**

R_1	$e^- + N_2 \rightarrow 2N + e^-$	R_{15}	$N_2(A) + N_2(A) \rightarrow N_2(C) + N_2$	R_{29}	$N + NO_2 \rightarrow N_2 + O_2$
R_2	$N + N + M \rightarrow N_2 + M$	R_{16}	$e^- + N_2 \rightarrow N_2(a) + e^-$	R_{30}	$N + NO_2 \rightarrow 2NO$
R_3	$e^- + N_2 \rightarrow N_2(A) + e^-$	R_{17}	$N_2(a) + N_2 \rightarrow N_2(B) + N_2$	R_{31}	$O + NO \rightarrow NO_2 + h\nu$
R_4	$N_2 + N + N \rightarrow N_2(A) + N_2$	R_{18}	$N_2(a) + O_2 \rightarrow N_2 + O + O$	R_{32}	$O + NO_2 \rightarrow NO + O_2$
R_5	$N_2(A) + N_2 \rightarrow N_2 + N_2$	R_{19}	$N_2(a) + NO \rightarrow N_2 + N + O$	R_{33}	$O + NO + M \rightarrow NO_2 + M$
R_6	$N_2(A) + N \rightarrow N_2 + N$	R_{20}	$N + NO \rightarrow O + N_2$	R_{34}	$NO_2 + NO_2 \rightarrow 2NO + O_2$
R_7	$e^- + N_2 \rightarrow N_2(B) + e^-$	R_{21}	$N + O_2 \rightarrow O + NO$	R_{35}	$N + O_3 \rightarrow NO + O_2$
R_8	$N + N + M \rightarrow N_2(B) + M$	R_{22}	$N + O + M \rightarrow NO + M$	R_{36}	$O + O_3 \rightarrow O_2 + O_2$
R_9	$N_2(A) + N_2(A) \rightarrow N_2(B) + N_2$	R_{23}	$N_2(A) + O_2 \rightarrow N_2 + O + O$	R_{37}	$NO + O_3 \rightarrow O_2 + NO_2$
R_{10}	$N_2(B) \rightarrow N_2(A) + h\nu$	R_{24}	$N_2(A) + NO \rightarrow N_2 + NO$	R_{38}	$O + O_2 + M \rightarrow O_3 + M$
R_{11}	$N_2(B) + N_2 \rightarrow N_2(A) + N_2$	R_{25}	$N_2(B) + NO \rightarrow N_2(A) + NO$	R_{39}	$+ NO_2 \rightarrow N_2O + O$
R_{12}	$e^- + N_2 \rightarrow N_2(C) + e^-$	R_{26}	$N_2(B) + O_2 \rightarrow N_2 + O + O$	R_{40}	$N_2(A) + O_2 \rightarrow N_2O + O$
R_{13}	$N_2(C) \rightarrow N_2(B) + h\nu$	R_{27}	$O + O + M \rightarrow O_2 + M$	R_{41}	$N_2(A) + N_2O \rightarrow N_2 + N + NO$
R_{14}	$N_2(C) + N_2 \rightarrow N_2(a) + N_2$	R_{28}	$N + NO_2 \rightarrow 2O + N_2$	R_{42}	$O + N_2O \rightarrow NO + NO$

AP Jet Downstream Modeling

Model Geometry



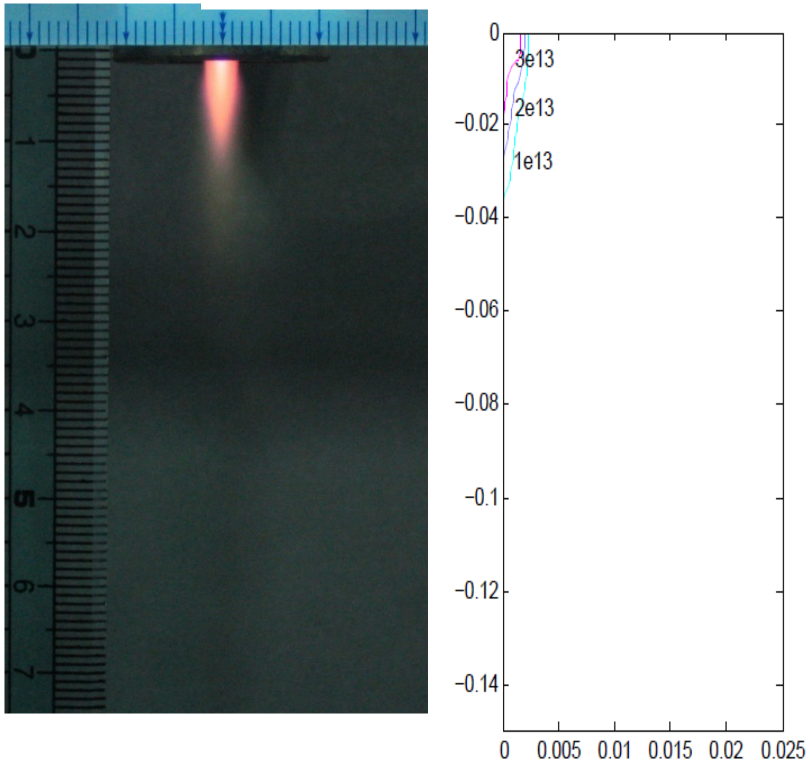
- Use fluid model to simulate each individual species. Each species has its own continuity equation, and exchanges mass through collision.
- Species included: N_2, N, 4 excited N_2, O_2, O, NO, NO_2 and N_2O.
- Over 10 coupled different equations.
- Use MatlabTM and Comsol Multiphysics™ to solve the equation system.

- Assuming constant ne and Te at the upstream.

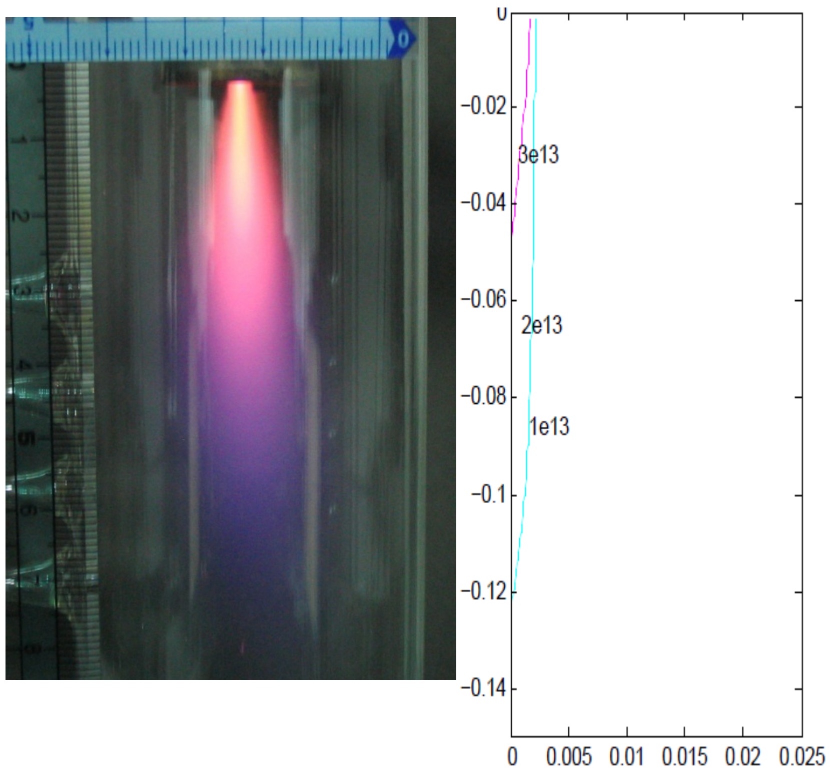
- Cases with and without ambient air diffusion.
- Cases with high and low flow rate.

Model Validation

With air



Without air



- Clearly demonstrate the effect of the ambient air diffusion, mostly due to

$$N_2^* + O_2 \rightarrow N_2 + 2\,O$$

I.H. Tsai, C.C. Hsu, IEEE Trans. Plasma Sci., 38 (2010) 3387

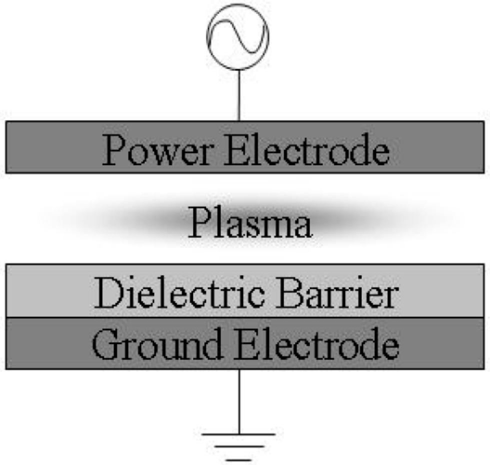
My Little Journey of Simulation

2007~ @ NTU

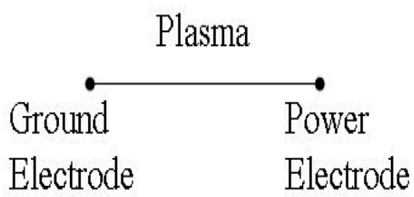
Comsol Multiphysics 3.5a

Modeling of Dielectric Barrier Discharge

Model Formulation



Model Geometry



Equation System

- Continuity Equation (e, N_2^+, N_4^+, N_2^*)

$$\frac{\partial n_i}{\partial t} + \nabla \cdot \Gamma_i = S_i$$

- Poisson Equation

$$e\sum_i q_i n_i = -\varepsilon_0 \nabla^2 V$$

- Surface charging

$$\varepsilon_0 \nabla \phi + \frac{\varepsilon_d}{l_{db}} \phi = \sigma_S$$

$$\sigma_S = \int \sum \pm e\Gamma_j dt$$

- Dielectric barrier at both electrodes.
- Not solving the dielectric barrier.
- Consider surface charging using Gauss law.

- Using AC power source
- Quantities vary with time.

Boundary Conditions

Continuity Equations:

$$\Gamma_e \cdot n = \frac{1}{4} n_e \cdot v_{th,e} - h_s \sum_j (\gamma_{i,j} \cdot \Gamma_{i,j}) - \sum_j (\gamma_{n,j} \cdot \Gamma_{n,j})$$

$$\Gamma_i \cdot n = h_s \cdot \mu_i \cdot n_i \cdot E + \frac{1}{4} n_i \cdot v_{th,i}$$

$$\Gamma_n \cdot n = \frac{1}{4} n_n \cdot v_{th,n}$$

where $h_s = \begin{cases} 1 & E \cdot n > 0 \\ 0 & E \cdot n \le 0 \end{cases}$

γ: secondary electron emission coefficient
v_{th}: thermal velocity
h_s: switching function, considers the direction of the E-field

Poisson Equation

$$\nabla \phi = \left(\sigma_s - \frac{\varepsilon_d}{l} (\phi - V) \right) \Big/ \varepsilon_0$$

where $\sigma_s = \int \sum (q_i \cdot \Gamma_i) dt$

$V = 0 \quad or \quad V_0 \sin(\omega t)$

σ_s: surface charging
ε_d: dielectric constant of the dielectric barrier
ε_0: vacuum vacuum permittivity
l: dielectric layer thickness (2 mm on both sides)

Plasma Chemistry

Species included:

e^-, N_2^+, N_4^+, excited N_2($A^3\Sigma_u^+$ $B^3\Pi_g$ $C^3\Pi_u$ $a'^1\Pi_u^-$)

Not include: N and N^+ due to small amount and unimportant kinetically.

Electron-Impact Reaction

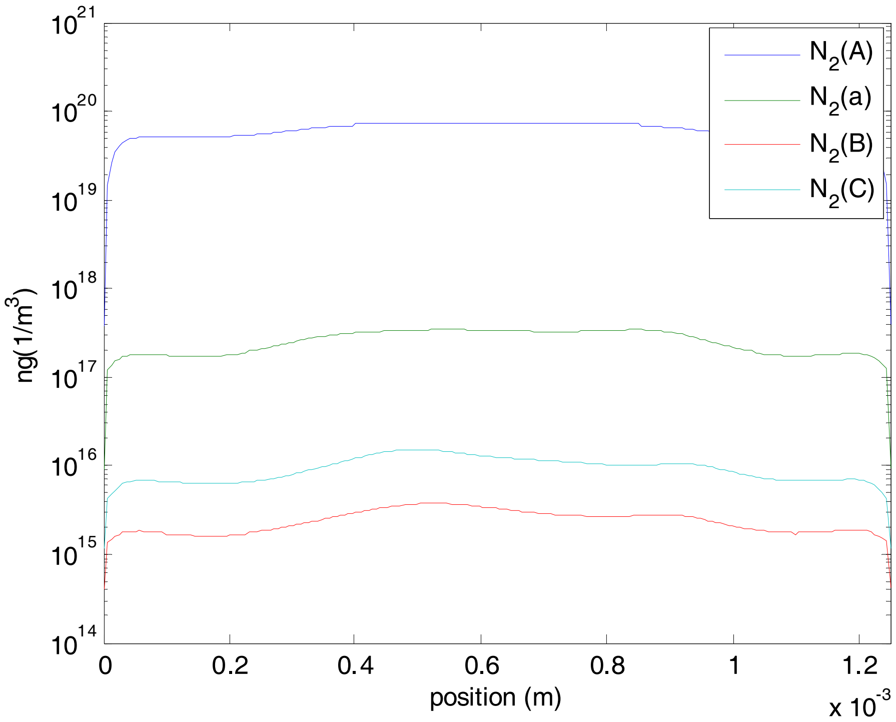
R10	$N_2 + e \rightarrow N_2^+ + 2e$
R11	$N_2 + e \rightarrow N_2(A) + e$
R12	$N_2 + e \rightarrow N_2(a) + e$
R13	$N_2 + e \rightarrow N_2(B) + e$
R14	$N_2 + e \rightarrow N_2(C) + e$
R20	$N_2^+ + e \rightarrow N_2$
R21	$N_2^+ + e \rightarrow N_2$
R25	$N_4^+ + e \rightarrow N_2(C) + N2$
R32	$N_2(a) + N_2(a) \rightarrow N_4^+ + e$
R33	$N_2(a) + N_2(A) \rightarrow N_4^+ + e$

Heavy Particle Reactions

R30	$N_2(a) + N_2 \rightarrow 2N_2$
R31	$N_2(a) + N_2 \rightarrow N_2(B) + N_2$
R34	$N_2(A) + N_2(A) \rightarrow N_2(C) + N_2$
R35	$N_2(A) + N_2(A) \rightarrow N_2(B) + N_2$
R36	$N_2(C) \rightarrow N_2(B)$
R37	$N_2(C) + N_2 \rightarrow N_2(a) + N_2$
R38	$N_2(B) \rightarrow N_2(A) + hv$
R39	$N_2(B) + N_2 \rightarrow N_2(A) + N_2$

Neutral Kinetics – Time-Averaged Densities

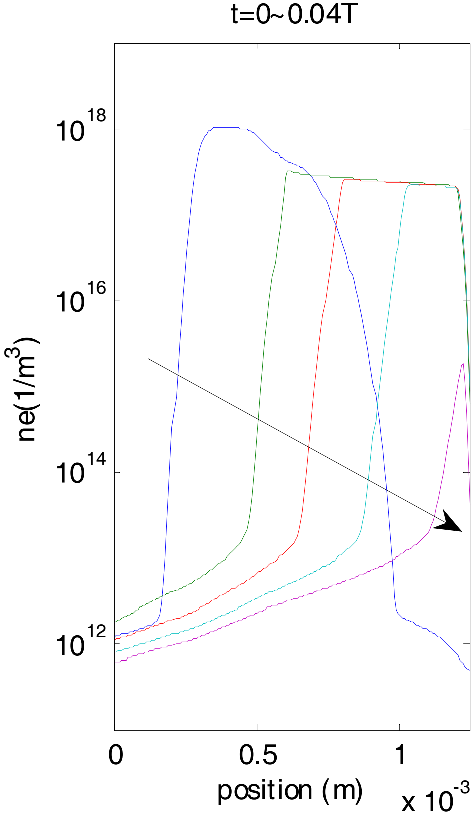
Center line condition: 15 kV, 30 kHz, 1.25 mm Gap

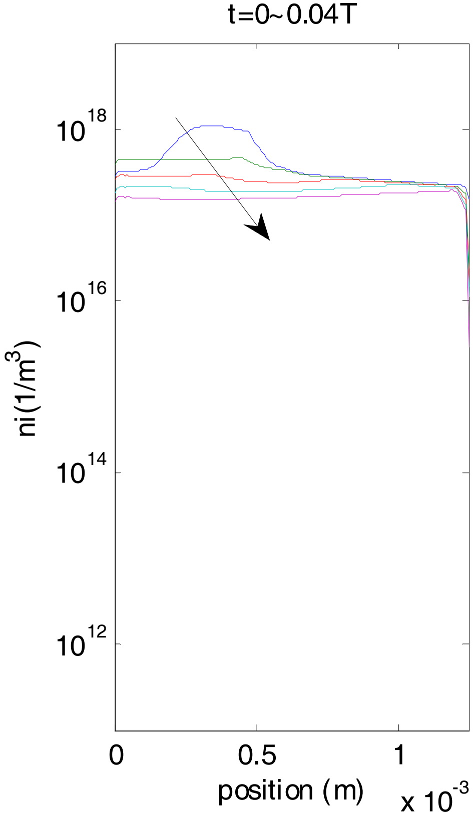


- $N_2(A)$ appears to be the dominant excited state N_2, as reported in the literature – same trend for most conditions.
- Major formation mechanism are electron-impact excitation.
- Rather flat profile in the bulk as a result of high pressure (AP)
- Rapidly drop at the electrode surface due to surface quenching reaction
- Simulation shows the density is more sensitive to the bulk reaction rates than to the surface quenching coefficient.

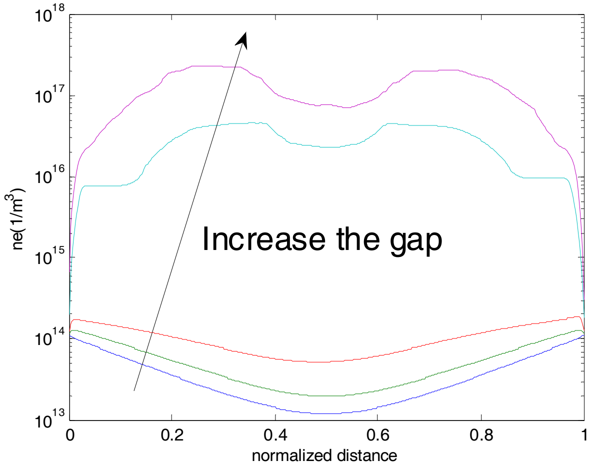
Plasma Structure – Charged Species

Center line condition: 15 kV, 30 kHz, 1.25 mm Gap

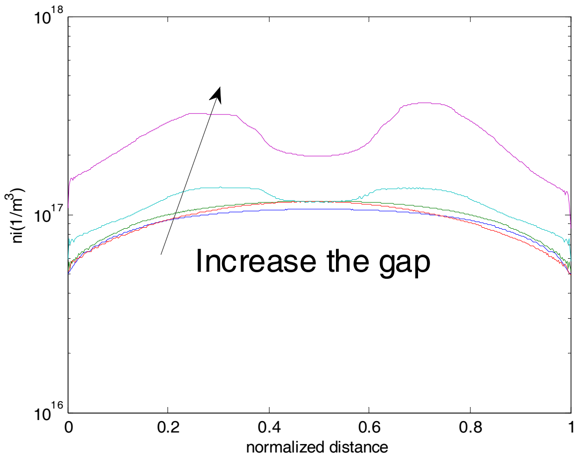




Averaged n_e, gap=0.75~1.5 mm



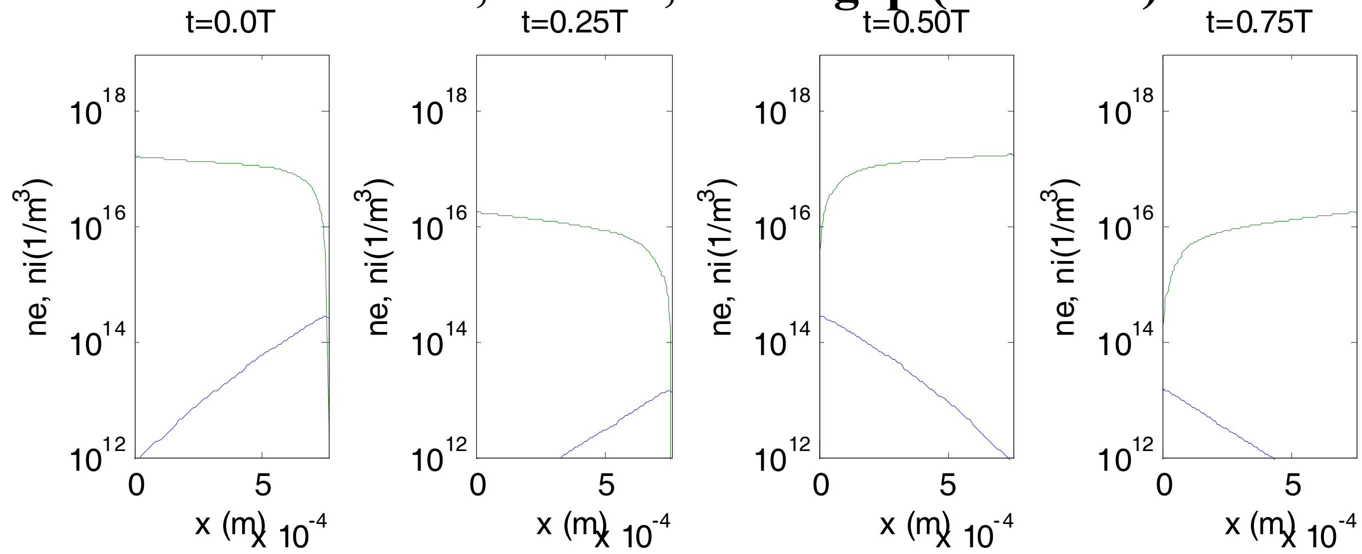
Averaged n_i, gap=0.75~1.5 mm

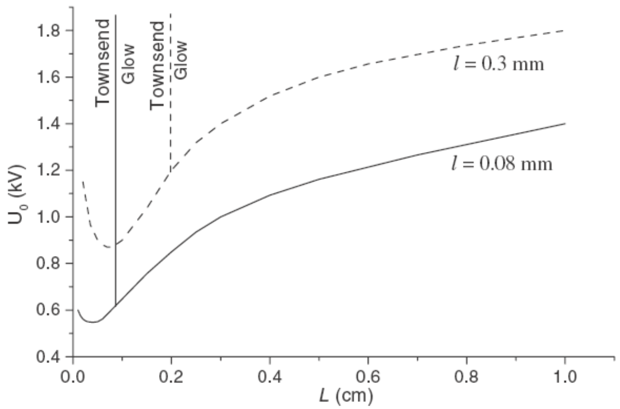


- Non-smooth averaged n_e due to its rapid with time.
- More significant change in n_e than in n_i
- Oscillation of ne is clearly seen.

Plasma to Non-Plasma Transition

15 kV, 30 kHz, Small gap (0.75 mm)





Y. B. Golubovskii, et al., J. Phys. D-Appl. Phys. **36**, 39-49 (2003).

- At small gap (<0.75 mm), no quasi-neutral region is seen throughout the power period.
- Similar behavior is seen in the literature for He plasmas.
- Non-plasma region sometimes terms “Townsend discharge” in the literature.
- Suggesting smaller gap does not necessarily lead to a higher density (reactivity).

Conclusion

- Comsol Multiphysics is a viable PDE solver to handle complex systems.
  - Able to simulate low/high pressure, steady state/ transient behavior of low temperature plasmas
  - Numerical simulation allows for better understanding and clearer observation of phenomena and physics, especially those not accessible by experimental work.
  - Having a tool (computer program) in hand is very important to scientific researchers/students.