COMSOL Multiphysics as a tool for research works on plasma and plasma processing

Tatsuru Shirafuji
Innovative Collaboration Center, Kyoto University

A. Discharge and Plasma
1. Introduction
2. Governing Equations
3. Solver Parameters
4. Results and Discussion

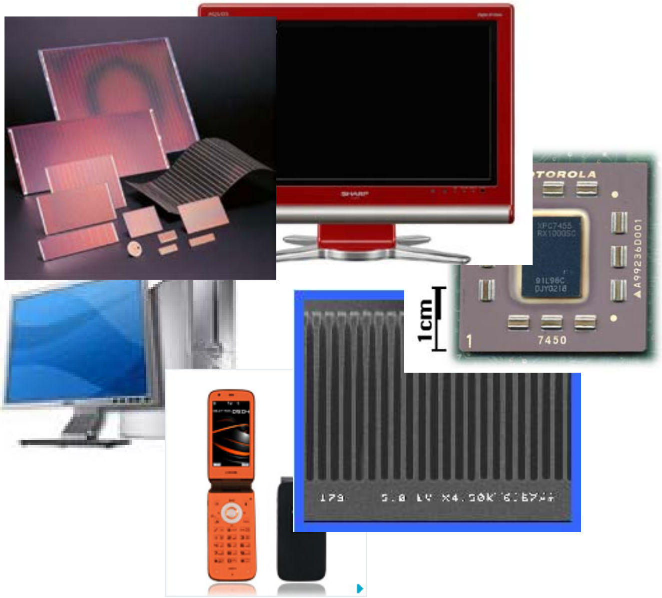
B. Chemical Reactions and Thin Film Deposition
1. Introduction
2. Governing Equations
3. Solver Parameters
4. Results and Discussion

Plasma Processing of Materials

Final Products

Thin Film Deposition

a-Si:H, uc-Si:H, poly-Si	Solar Cells, TFT
a-C:H, DLC	Hard Coating, Gas Barrier
SiOx(:CHx)	Dielectric Layer in ULSIs
SiNx	Passivation, Gas Barrier



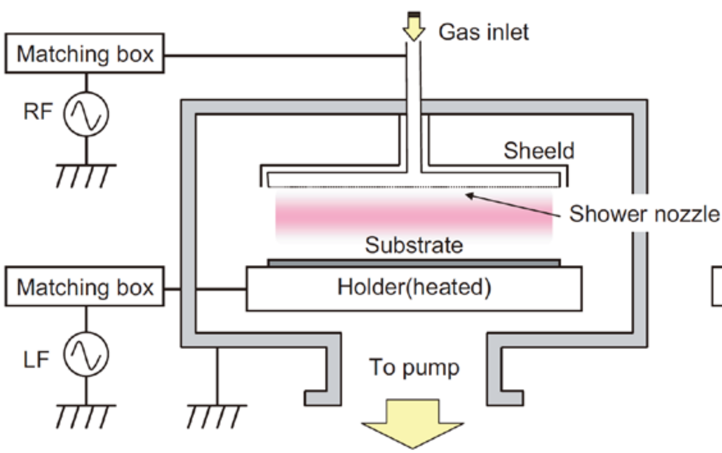
Dry Etching

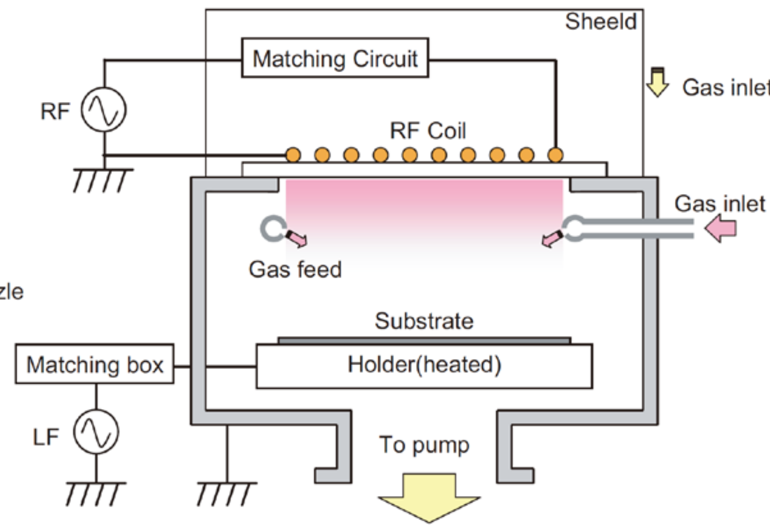
- Trench and Via in ULSIs
- Trench and Via in MEMS
- Ashing of photoresists
- Reactor Cleaning

Plasma Processing of Materials

Equipments for Processing

P Vacuum Chamber
Q Gas Feeding/Pumping Systems
V Electrodes
T Substrate Heater/Cooler



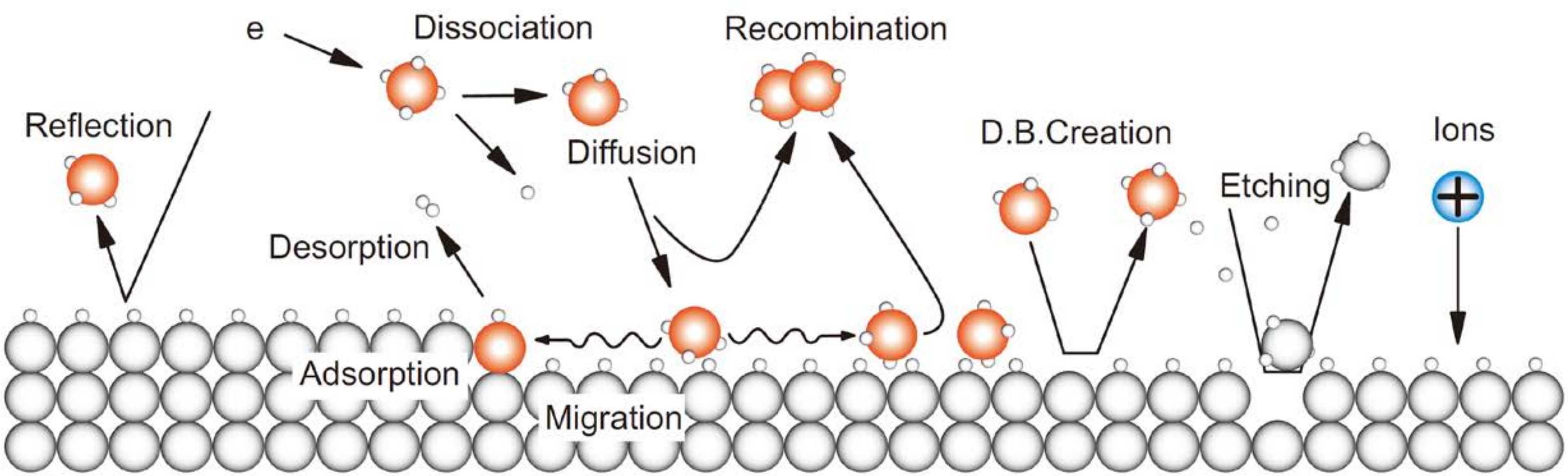


Plasma Processing of Materials

Inside the reactors

Discharge phenomena → time scale: ns or shorter
Transport phenonena → time scale: us or longer

Simultaneous solution is not realistic at this moment
→ Separated Treatment



Plasma Processing of Materials

Important Parameters

Electrical Discharge (Primary Processes)

Major interests =

Production rate of active species

Where and When?

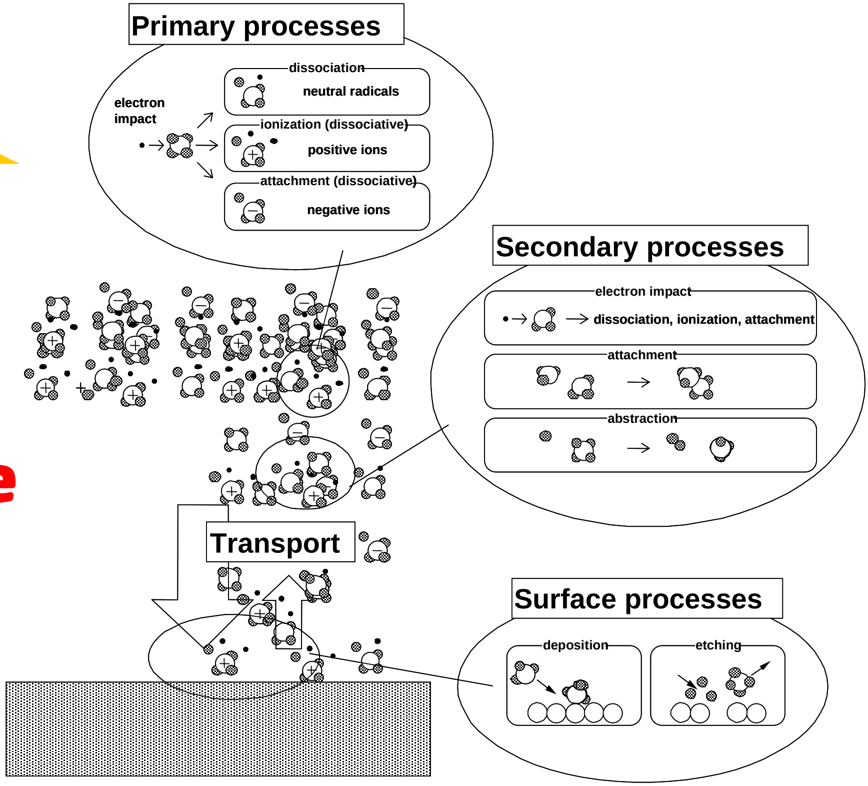
Feedback information if possible

Transport

Major interests =

Steady State Density Profile

Flux onto the Surface



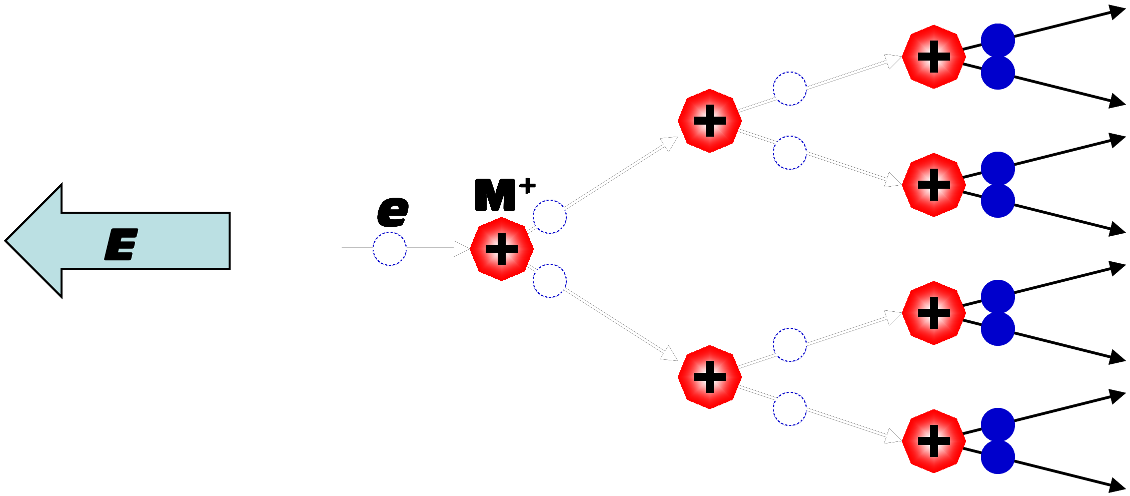
A. Discharge and Plasma

1. Introduction
2. Governing Equations
3. Solver Parameters
4. Results and Discussion

Electrical Discharge

Important Physics: Ionization
(Attachment)
(Recombination)

Chain reaction, or avalanche of electron and ion production



Electrical Discharge

Description of the Model

Transport of Electrons and Positive Ions

$$\frac{\partial n_e}{\partial t} - \nabla(D_e \nabla n_e - \upsilon_e n_e) = G_e - R_e$$

$$\frac{\partial n_i}{\partial t} - \nabla(D_i \nabla n_i - \upsilon_i n_i) = G_i - R_i$$

calculated (G_e, G_i)

neglect for simplicity
consider only diffusion loss (R_e, R_i)

Boundary Condition

Electrons	Density = Zero	
	Gamma Effects is not considered	
Ions	Convective Flux	

Electrical Discharge

Description of the Model (LFA*)

Ionization Rate

$$G_{\mathrm{e}} = G_{\mathrm{i}} = \alpha_{\mathrm{i}} \upsilon_{\mathrm{e}} n_{\mathrm{e}}$$

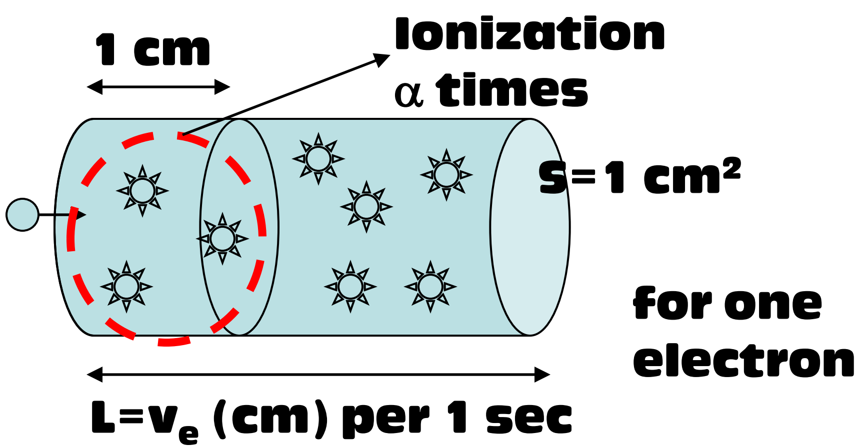
*Local Field Approx.

Townsend Ionization Coefficient α_i and v_e
← Obtained by solving Boltzmann Equation as a function of E/N or E/P

n_e ← Obtained from the data of previous time step

Townsend Ionization Coefficient α_i
= Ionization Frequency / unit length

UNIT: cm^{-1} or m^{-1}

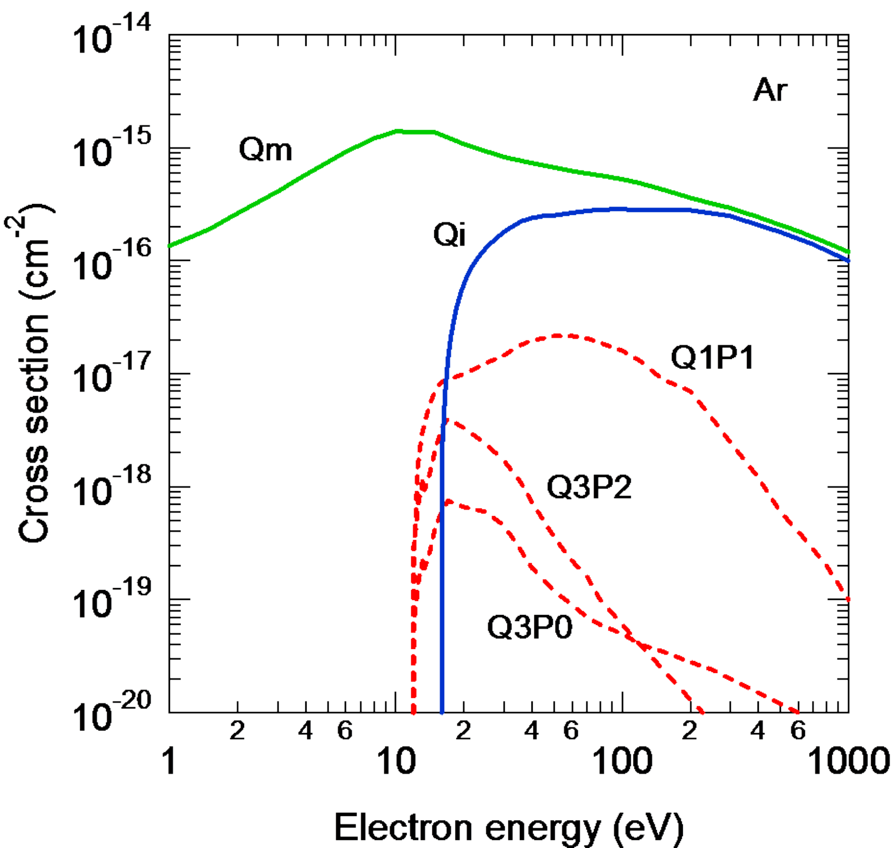


Electrical Discharge

Data for Solving Boltzmann Equation

Cross section data sets of the gas

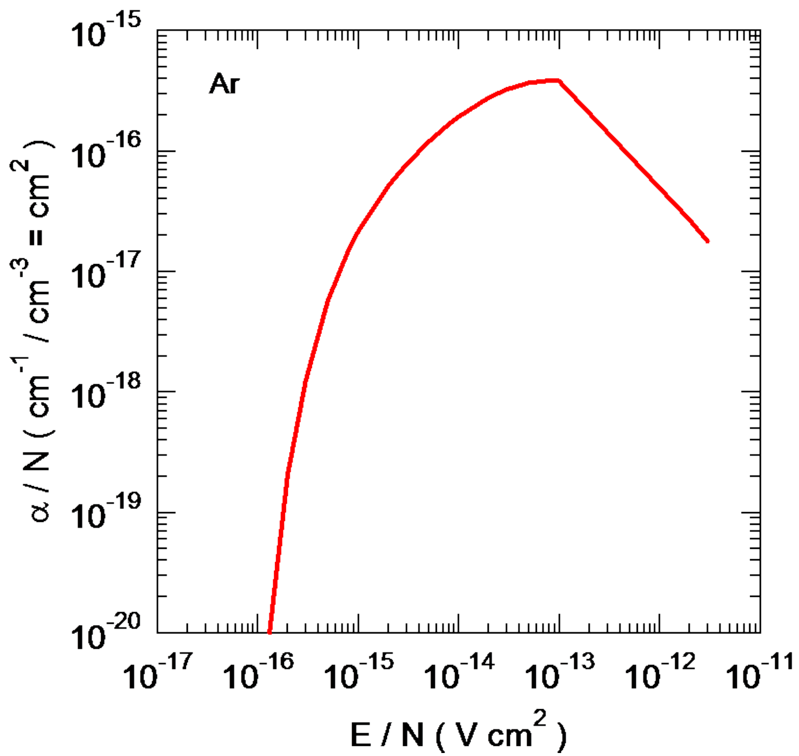
$$f = f(\boldsymbol{r}, \boldsymbol{\upsilon}, t) \qquad \frac{\partial f}{\partial t} + \boldsymbol{\upsilon} \bullet \nabla_r f + \boldsymbol{a} \bullet \nabla_\upsilon f = \left(\frac{\partial f}{\partial t} \right)_C$$

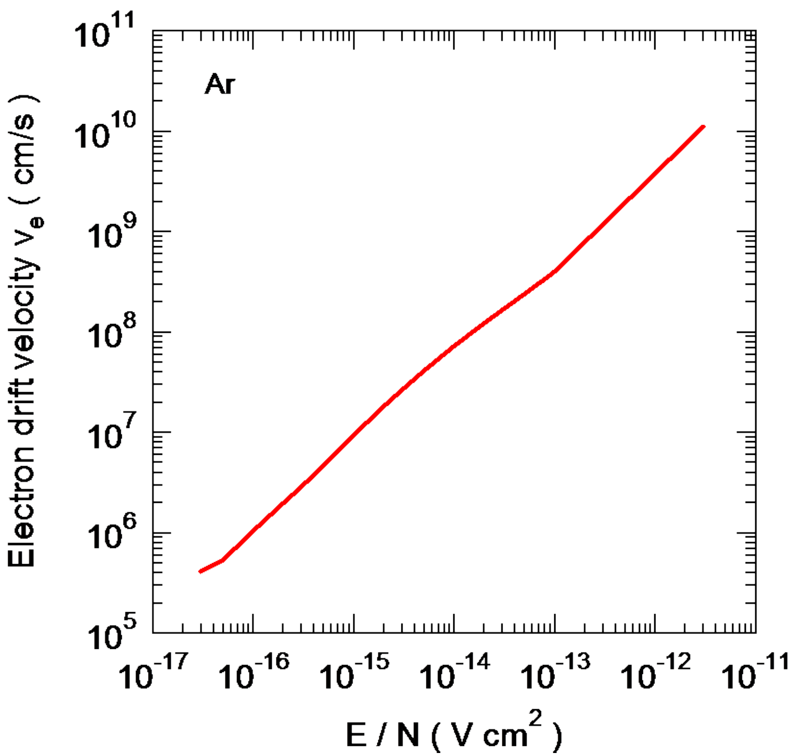


Electrical Discharge

Solution of Boltzmann Equation

- → **Ionization Coefficient vs. E/N**
- → **(Excitation/Attachment)**
- → **Electron Drift Velocity vs. E/N**
- → **Electron Diffusion Coefficient vs. E/N**





Electrical Discharge

Ionization Coefficient vs. E/N
(Validity check)

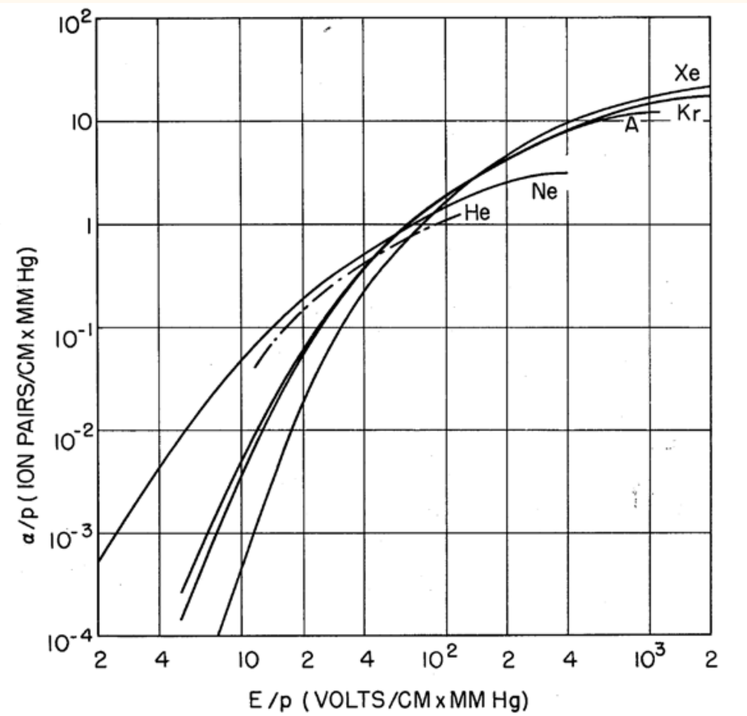


Fig. 4.34a. First Townsend ionization coefficients in noble gases.

A. von Engel, Handbuch der Physik, Springer Verlag, Berlin (1956) Vol. 21, p. 504.

Basic Data of Plasma Physics

SANBORN C. BROWN

Associate Professor of Physics
Massachusetts Institute of Technology

Published jointly by

The Technology Press of
The Massachusetts Institute of Technology

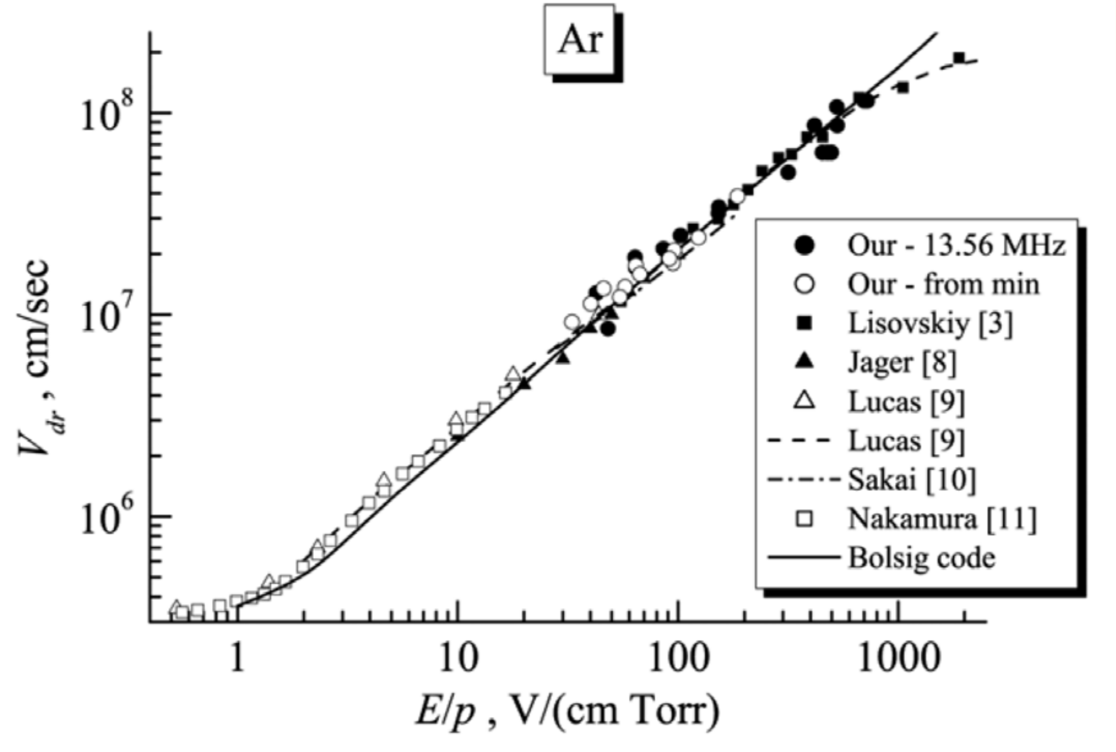
and

John Wiley & Sons, Inc., New York

Chapman and Hall, Limited, London

Electrical Discharge

Electron Drift Velocity vs. E/N
Validity Check



J. Phys. D: Appl. Phys. **39** (2006) 660–665

V Lisovskiy[1,3], J-P Booth[1], K Landry[2], D Douai[2], V Cassagne[2] and V Yegorenkov[3]

Electrical Discharge

Ion Transport Parameters

Evangelos Gogolides[a)] and Herbert H. Sawin

3971 J. Appl. Phys. **72** (9), 1 November 1992

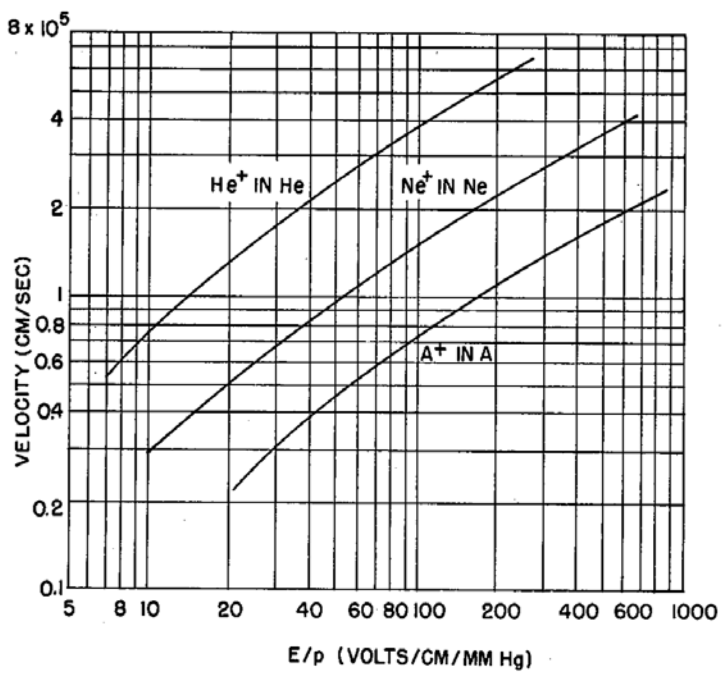
Parameters for Ar	Values
D_eP diffusivity * pressure	120×10^4 cm^2 Torr/s
μ_eP mobility * pressure	30×10^4 cm^2 Torr/V s
eD_e/μ_e characteristic energy	4.0 eV
Positive ion Ar^+	$m_+=40$ amu
D_+P diffusivity * pressure	40.0 cm^2 Torr/s
μ_+P mobility * pressure	14.44×10^2 cm^2 Torr/V s

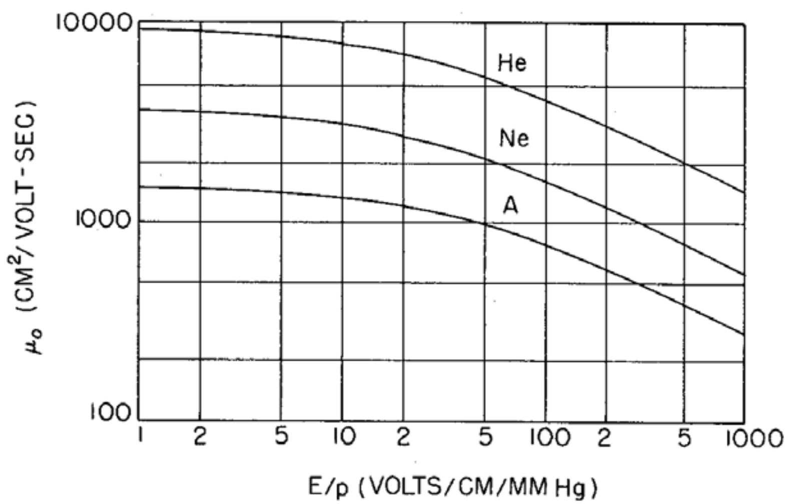
Basic Data of Plasma Physics

SANBORN C. BROWN
Associate Professor of Physics
Massachusetts Institute of Technology

Published jointly by
The Technology Press of
The Massachusetts Institute of Technology
and
John Wiley & Sons, Inc., New York

Chapman and Hall, Limited, London





ig. 3.24a. Ion mobility in helium, neon, and argon.

L.S.Frost, Phys. Rev. 105, 354 (1957).

Electrical Discharge (1D CCP)

Solution of 2 Fluids and Poisson Equations

1-Dimensional

Convection and Diffusion (for Electrons)

- ← Drift (Boltzmann)
- ← Diffusion (Constant)
- ← Ionization (Boltzmann)
- ← Boundary ($n_e = 0$)

$$\frac{\partial n_e}{\partial t} - \nabla(D_e \nabla n_e - \upsilon_e n_e) = G_e - R_e$$

Convection and Diffusion (for Ions)

- ← Drift (← constant mobility)
- ← Diffusion (Constant)
- ← Ionization (Boltzmann)
- ← Bounary ($dn_i/dx = 0$)

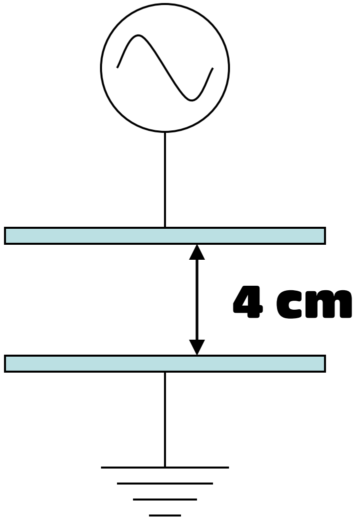
$$\frac{\partial n_i}{\partial t} - \nabla(D_i \nabla n_i - \upsilon_i n_i) = G_i - R_i$$

$$\nabla^2 V = -\frac{\rho}{\varepsilon_0}$$

$$\rho = q_0 (n_i - n_e) \times N_A$$

Poisson's Equation

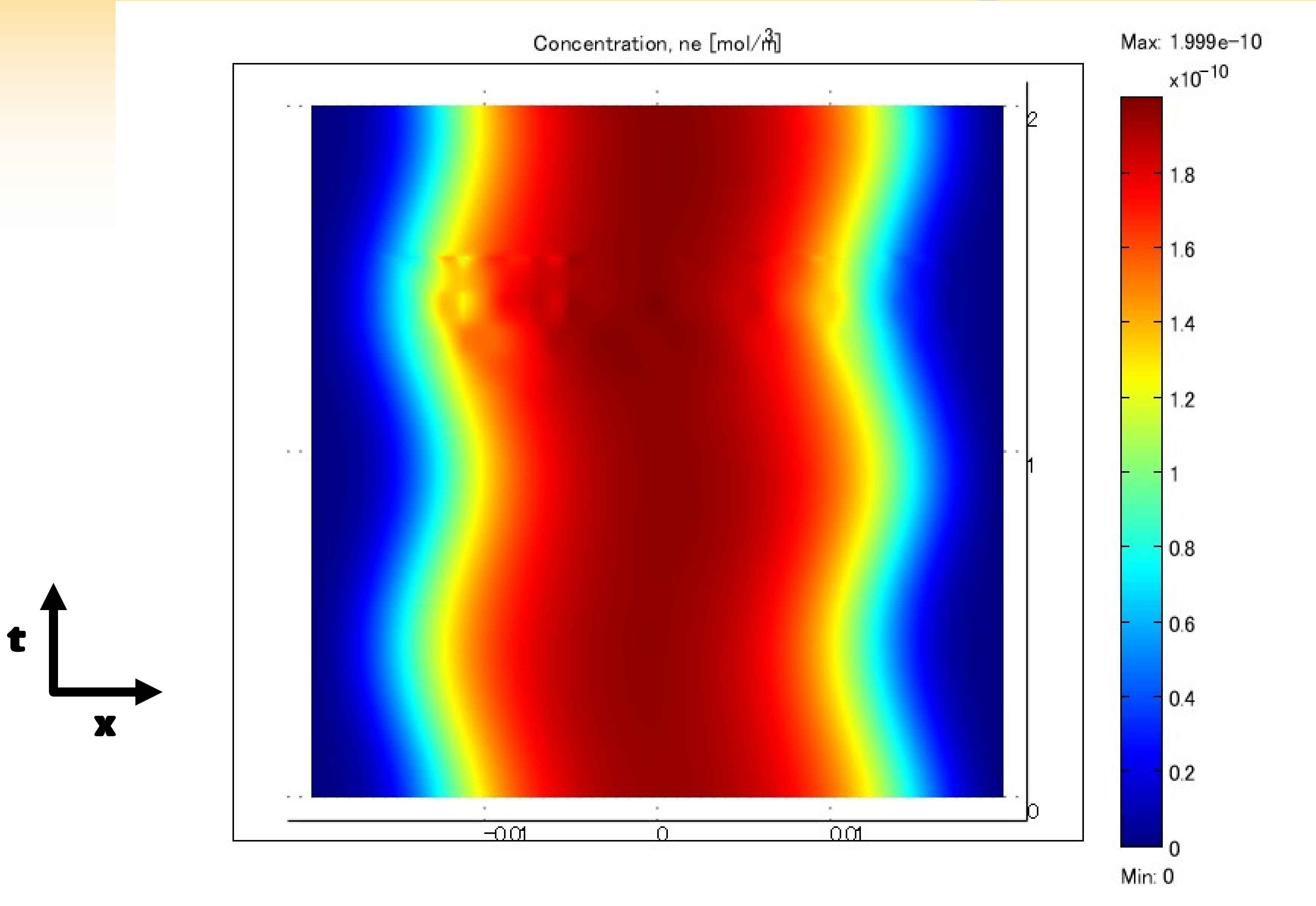
- ← Electron and Ion Density
- ← Boundary ($V(RF)=V_{app}*\sin(2\pi f t)$, $V(GND)=0$)



Electrical Discharge (1D CCP)

100 V 1 Torr 10 Cycles

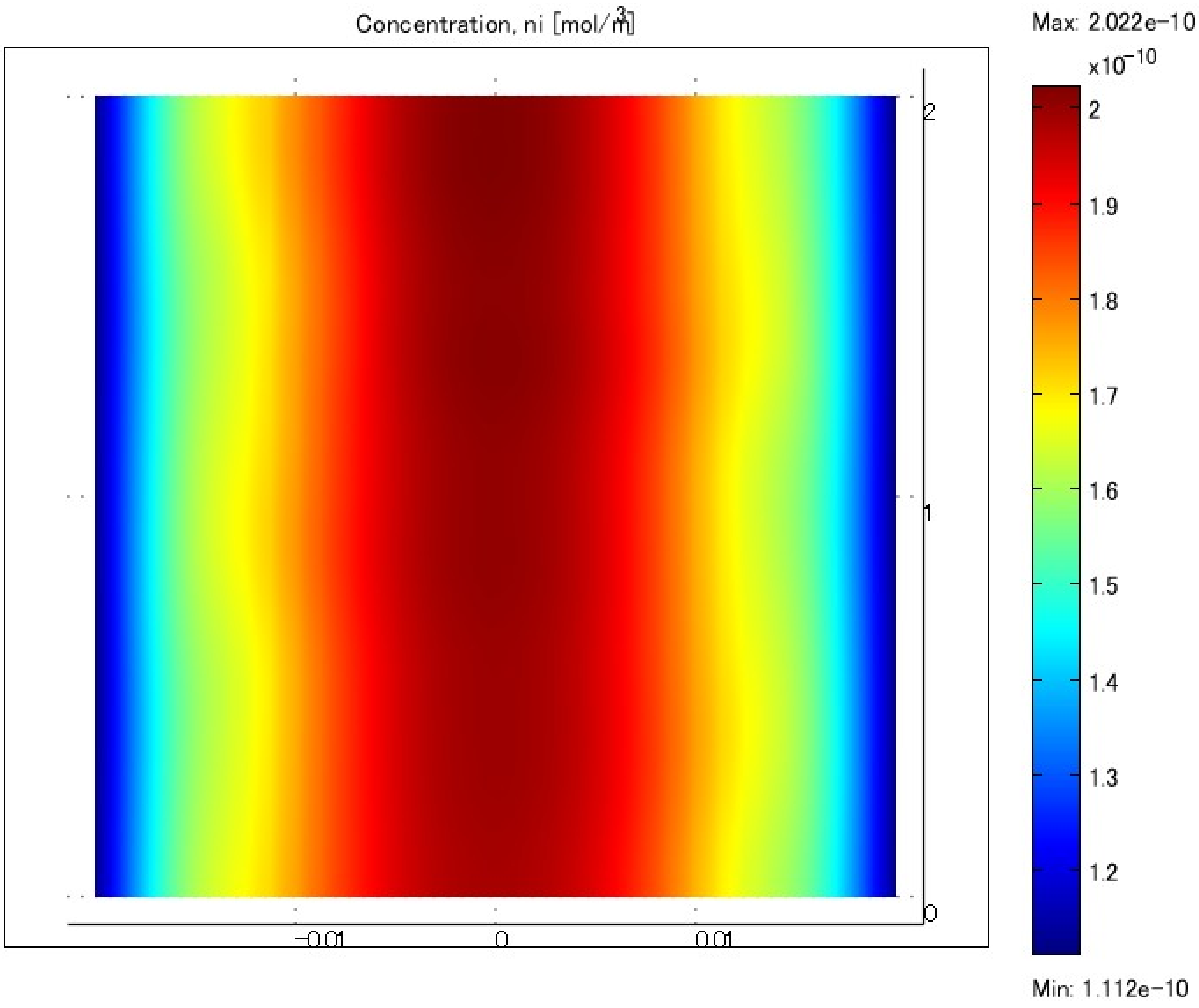
Electron density

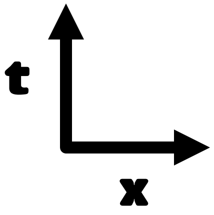


Electrical Discharge (1D CCP)

100 V 1 Torr 10 Cycles

Ion density

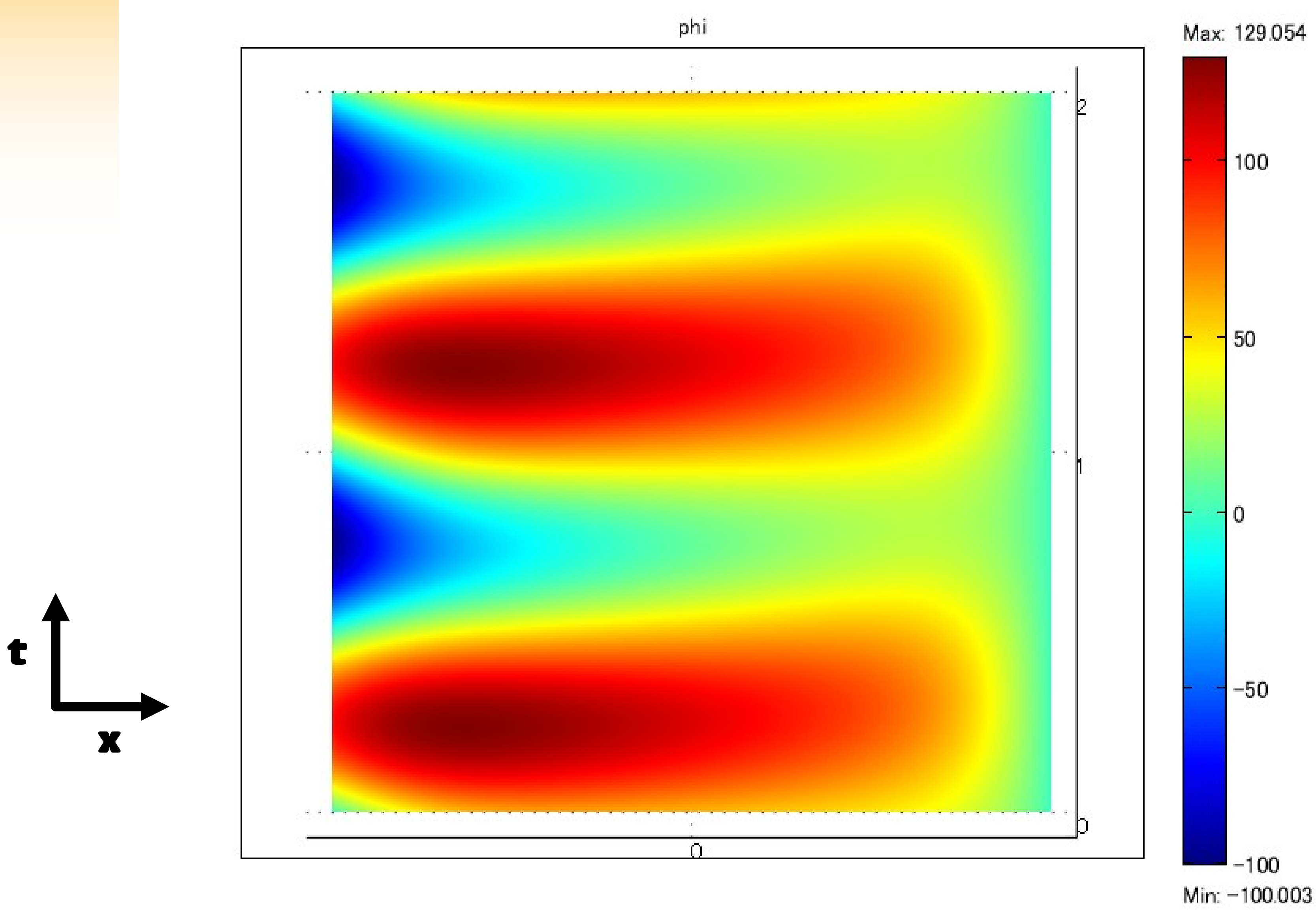




Electrical Discharge (1D CCP)

100 V 1 Torr 10 Cycles

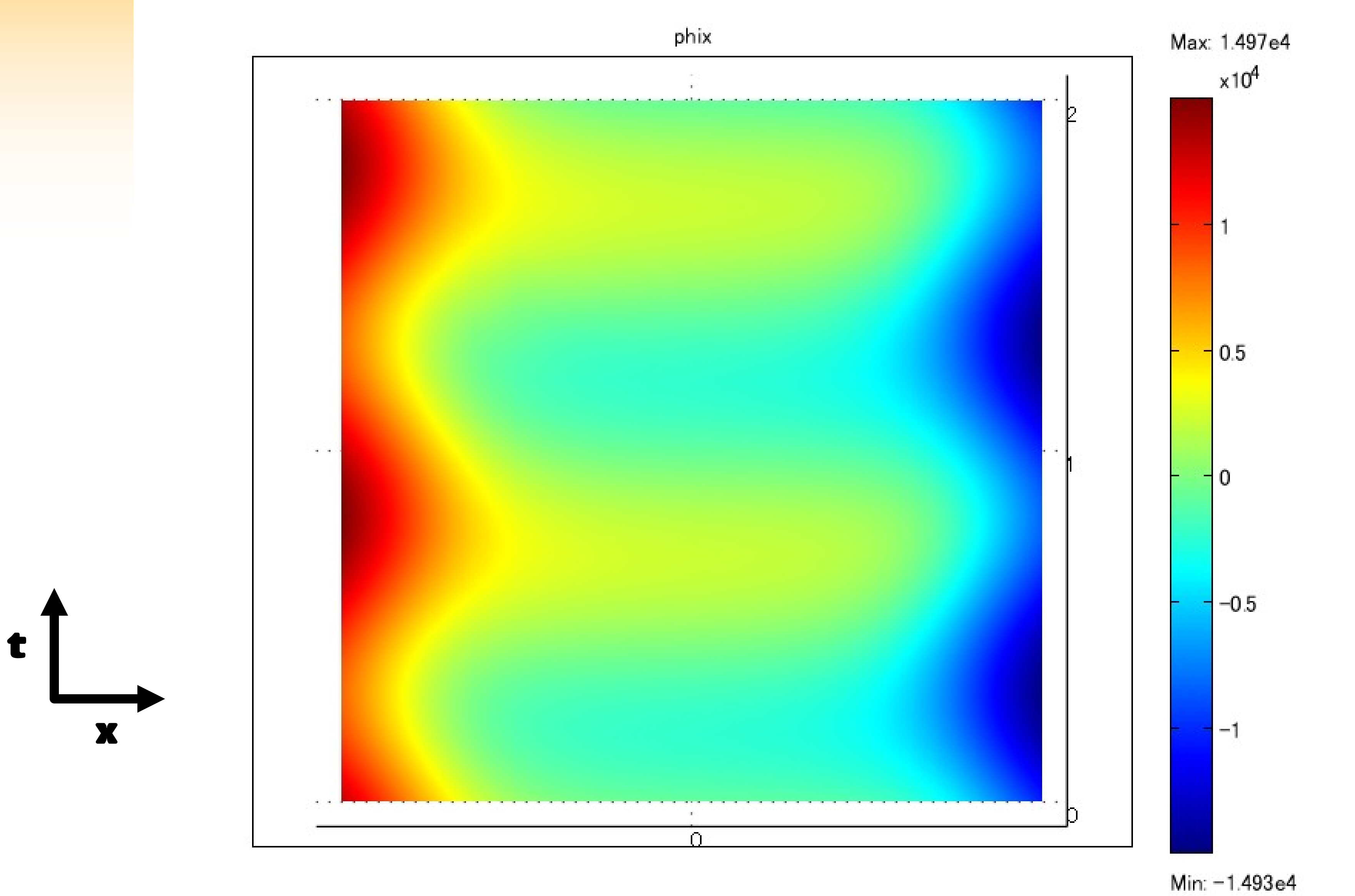
Electric Potential



Electrical Discharge (1D CCP)

100 V 1 Torr 10 Cycles

-Electric Field (dPhi/dx)

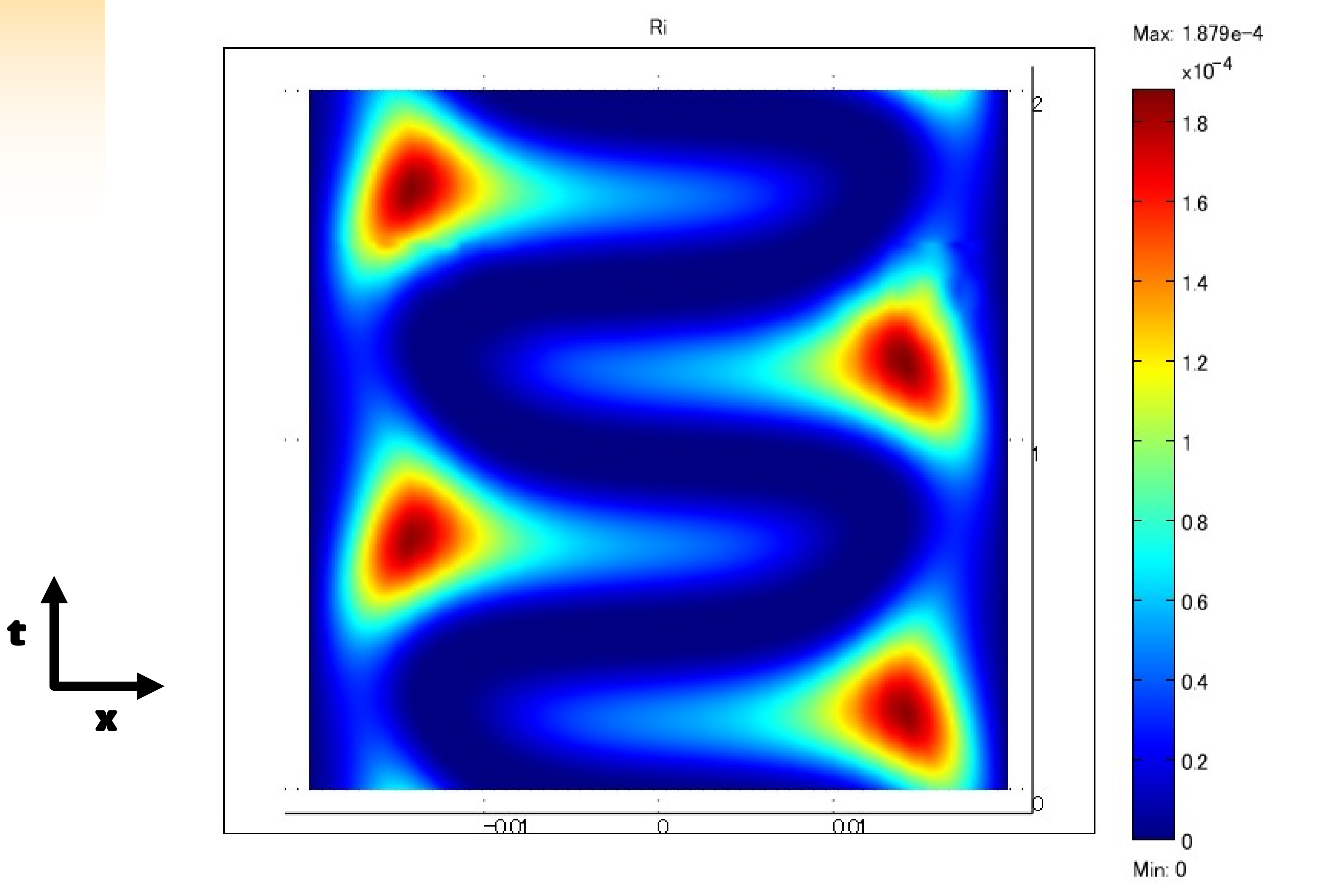


Electrical Discharge (1D CCP)

100 V 1 Torr 10 Cycles

Ionization Rate

$$G_e = G_i = \alpha_i \upsilon_e n_e$$

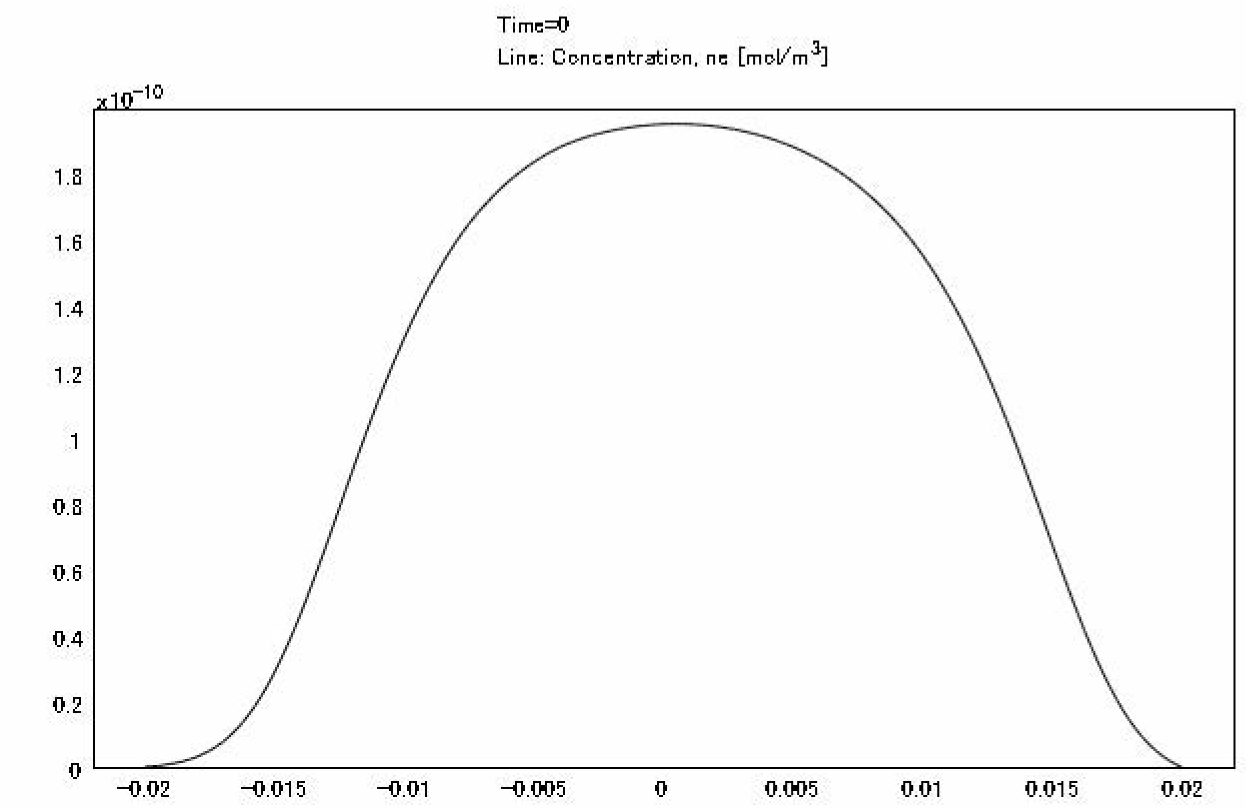


Electrical Discharge (1D CCP)

100 V 1 Torr 10 Cycles

Electron density

Spatio-Temporal Variation

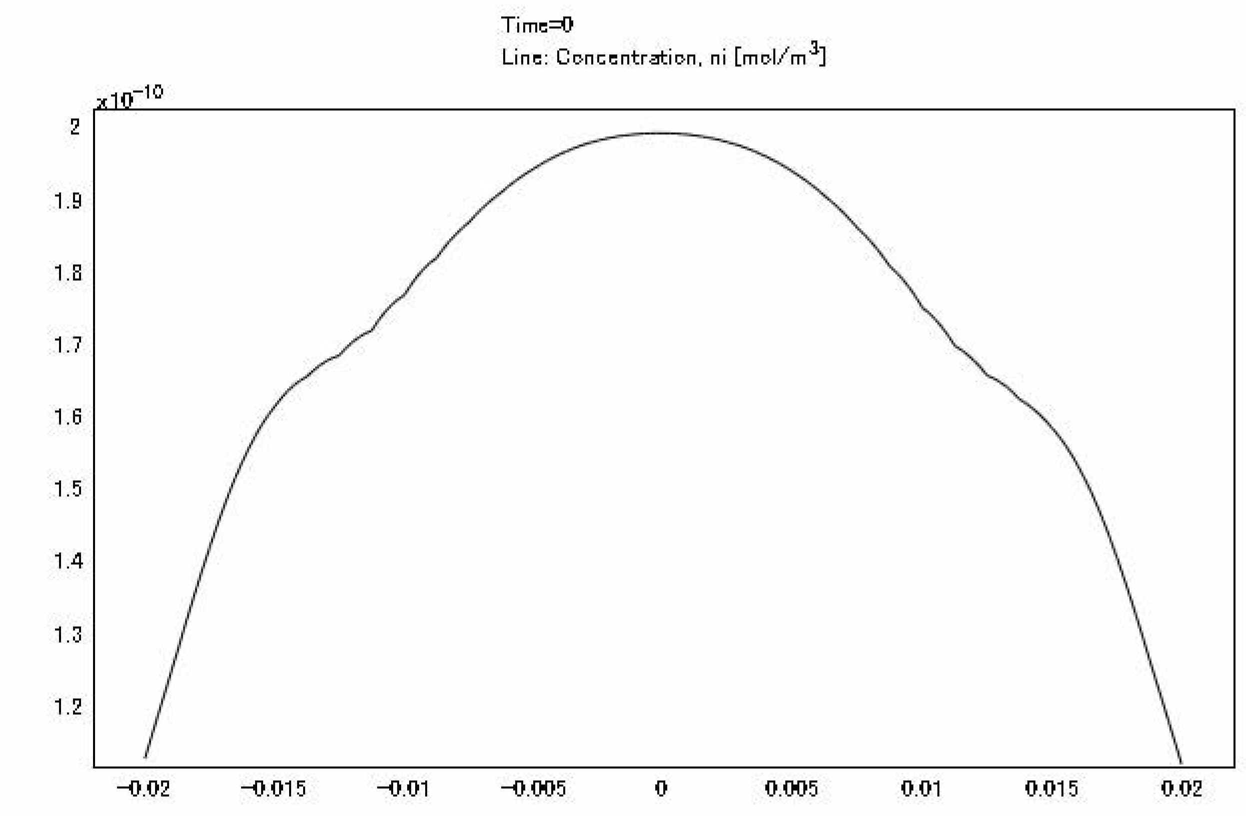


Electrical Discharge (1D CCP)

100 V 1 Torr 10 Cycles

Ion density

Spatio-Temporal Variation

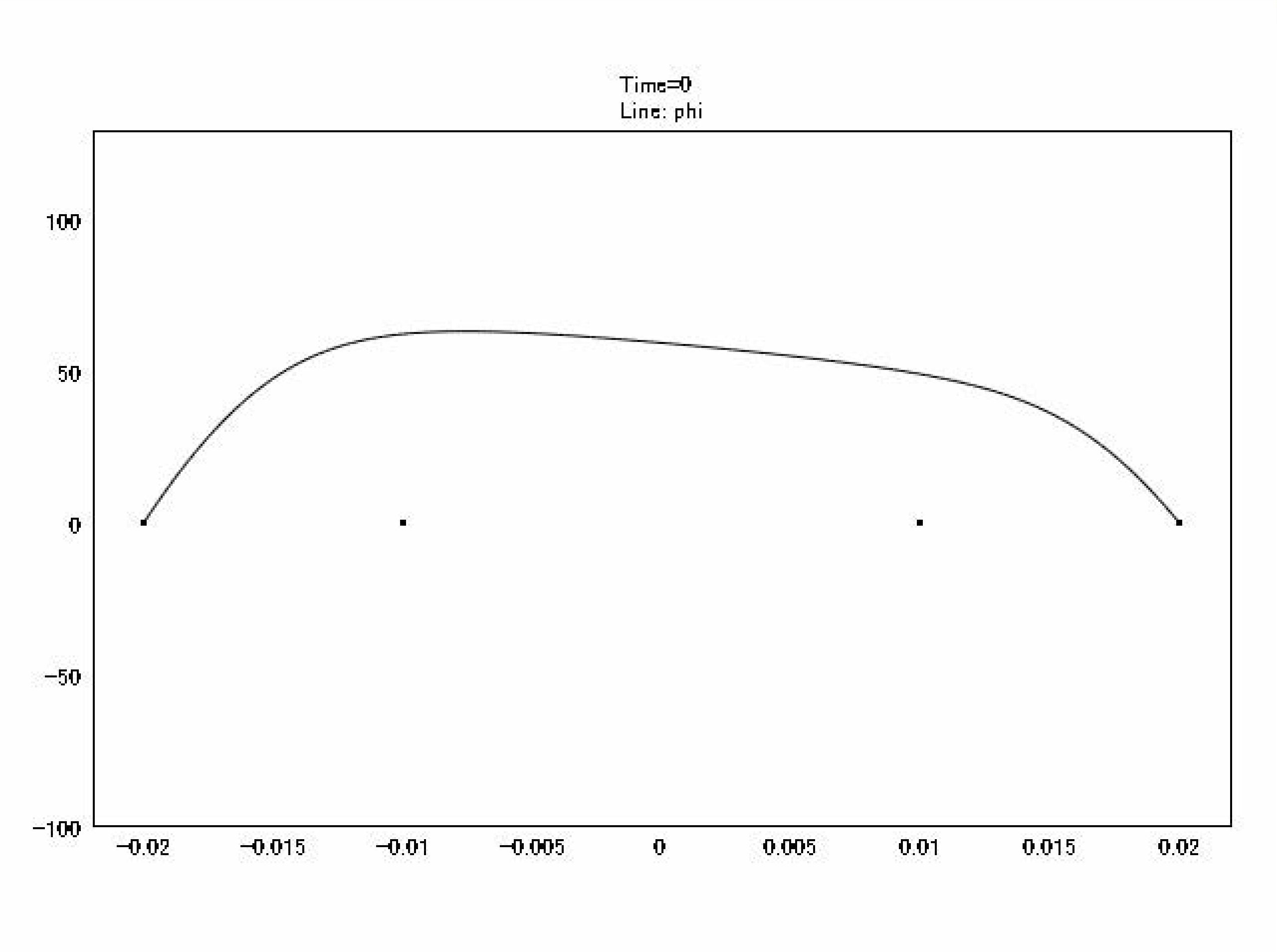


Electrical Discharge (1D CCP)

100 V 1 Torr 10 Cycles

Electric Potential

Spatio-Temporal Variation

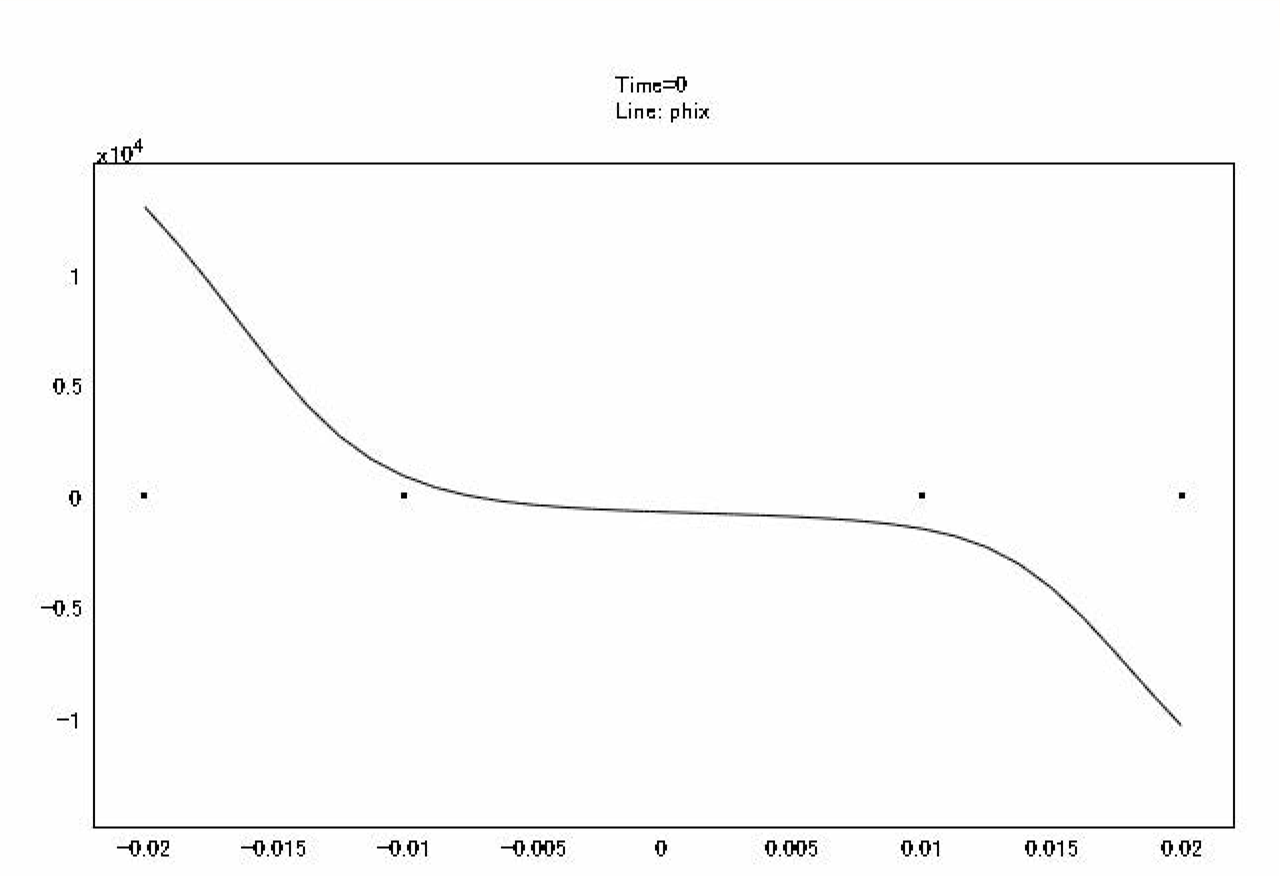


Electrical Discharge (1D CCP)

100 V 1 Torr 10 Cycles

- Electric Field (dPhi/dx)

Spatio-Temporal Variation

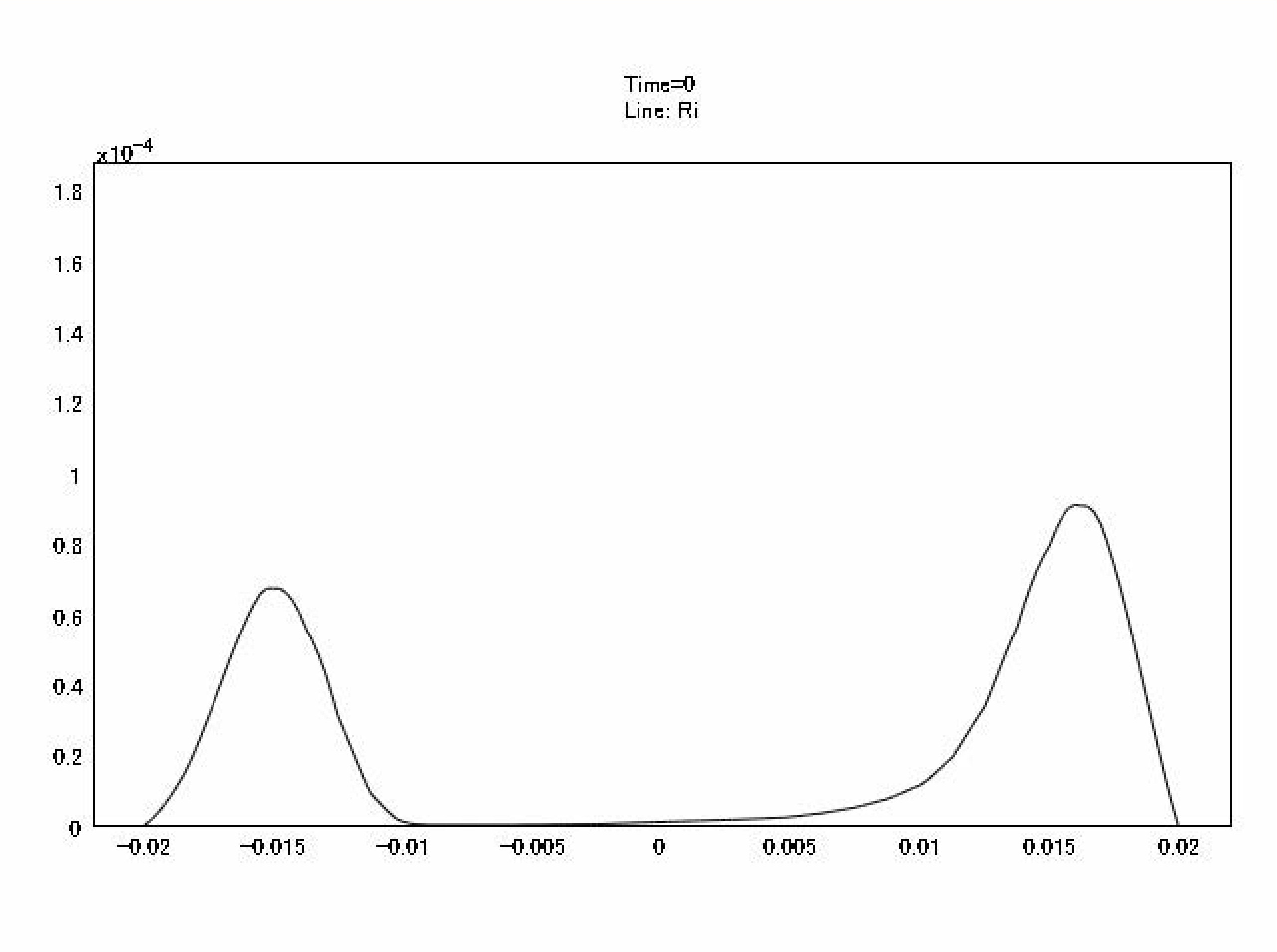


Electrical Discharge (1D CCP)

100 V 1 Torr 10 Cycles

Ionization Rate

Spatio-Temporal Variation



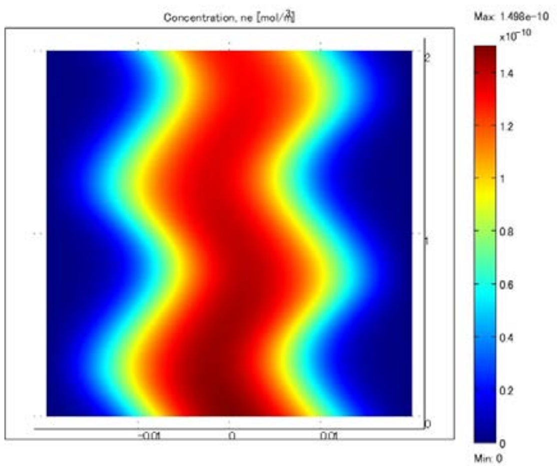
Electrical Discharge (1D CCP)

100 V 2 Cycles

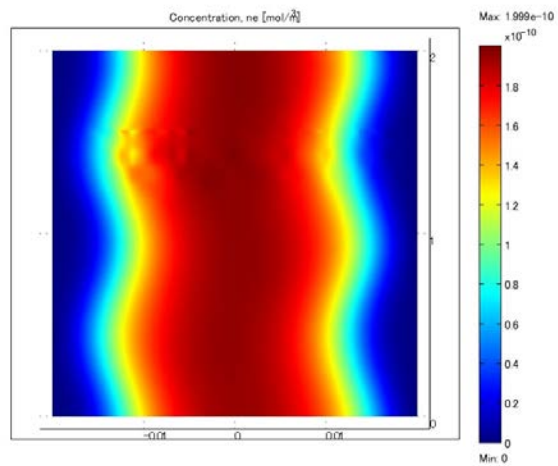
Pressure dependence

Electron density

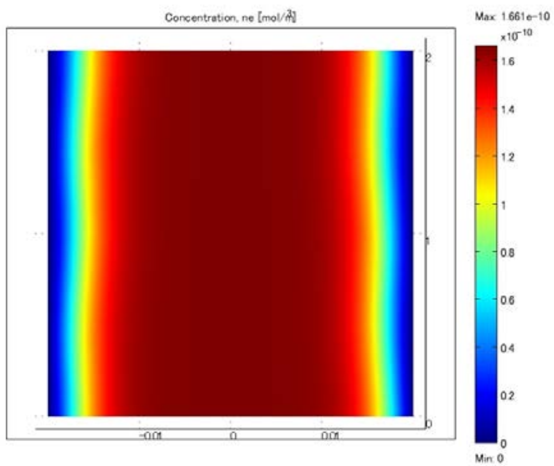
0.1 Torr

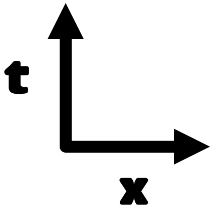


1 Torr



10 Torr





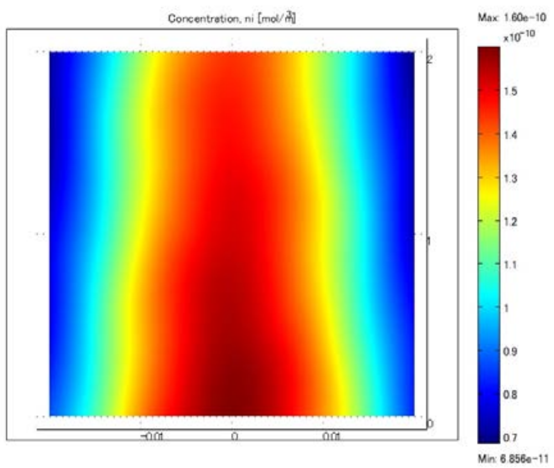
Electrical Discharge (1D CCP)

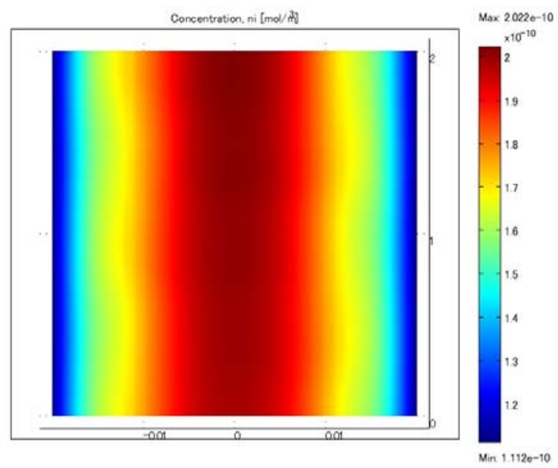
100 V 2 Cycles

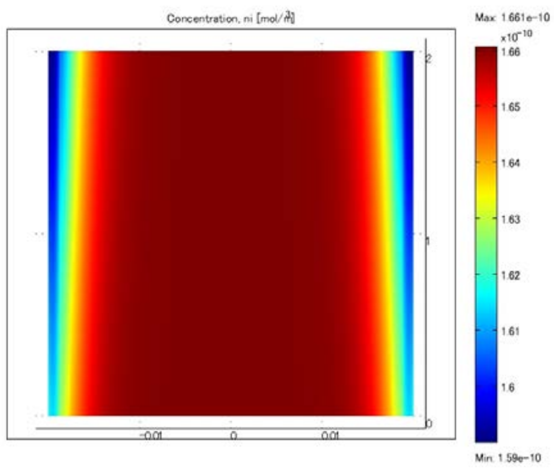
Pressure dependence

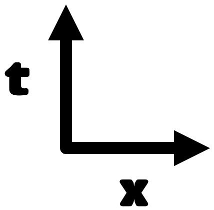
Ion density

0.1 Torr **1 Torr** **10 Torr**









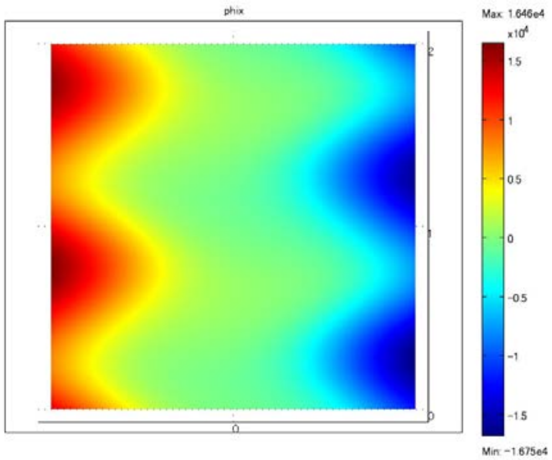
Electrical Discharge (1D CCP)

100 V 2 Cycles

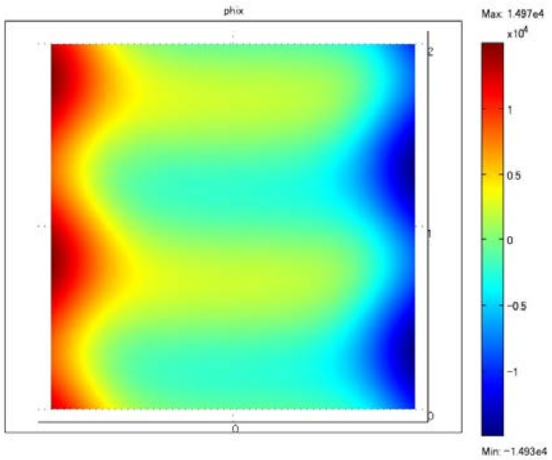
Pressure dependence

Electric Field

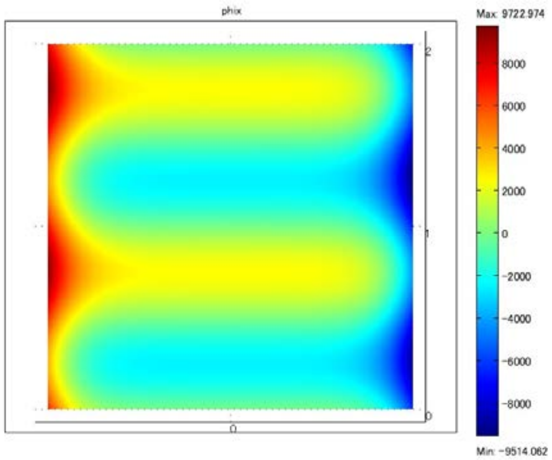
0.1 Torr

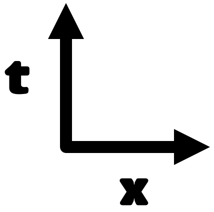


1 Torr



10 Torr



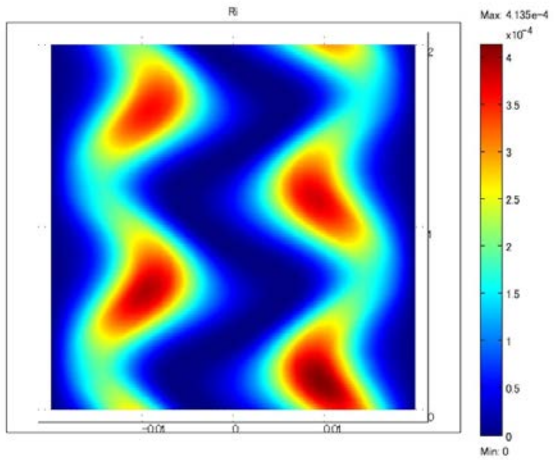


100 V 2 Cycles

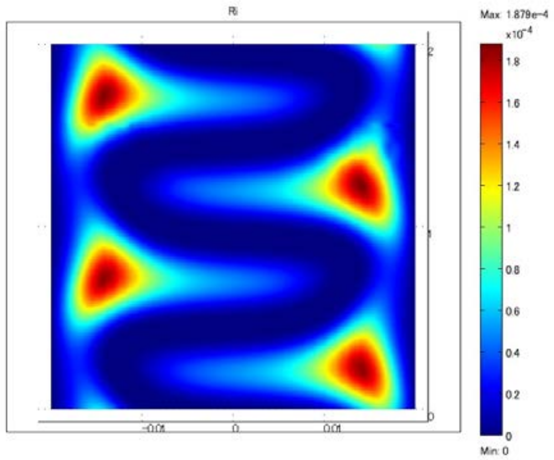
Pressure dependence

Ionization Rate

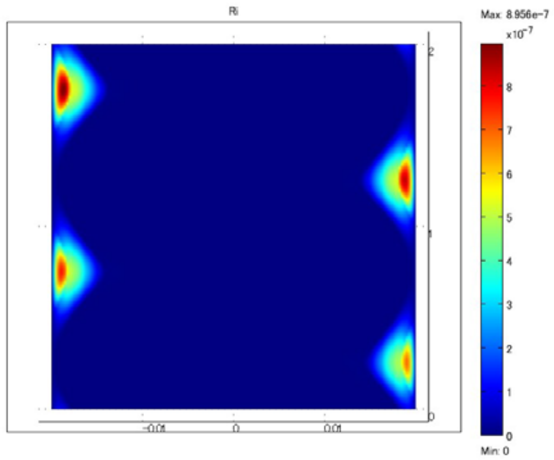
0.1 Torr

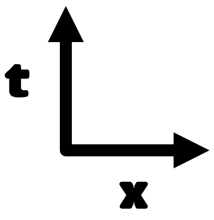


1 Torr



10 Torr

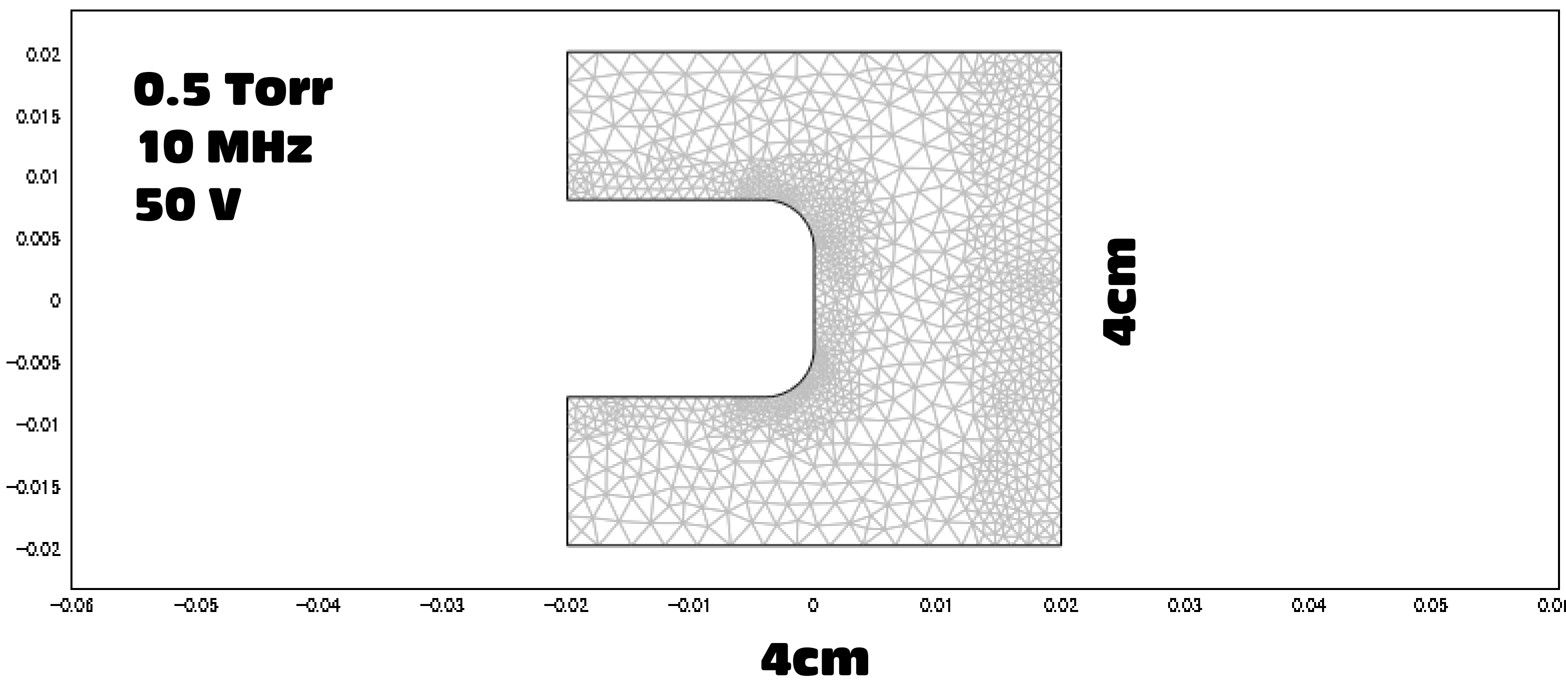




Electrical Discharge (2D CCP)

Easy to Extend from 1D to 2D

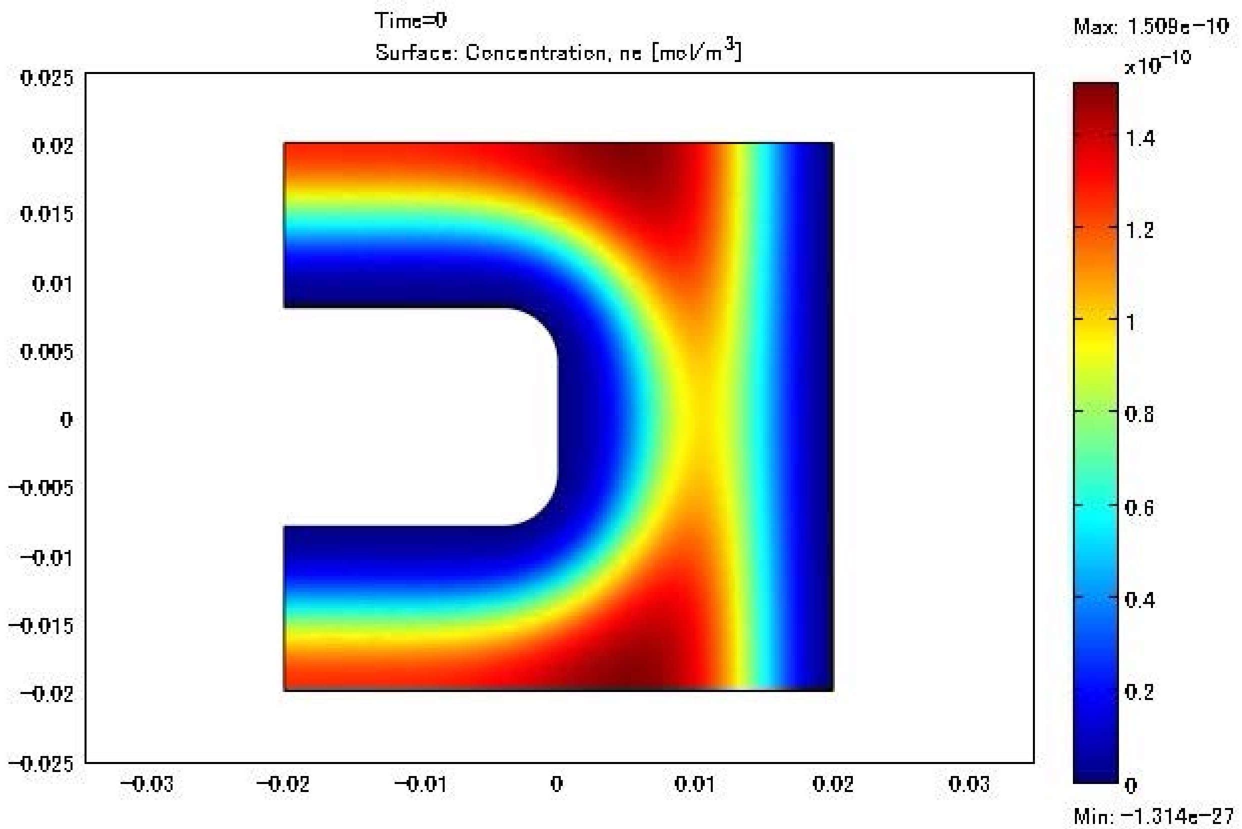
(but much memory is required)

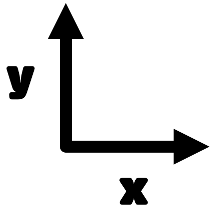


Electrical Discharge (2D CCP)

Electron Density

Motion of electrons can be understood

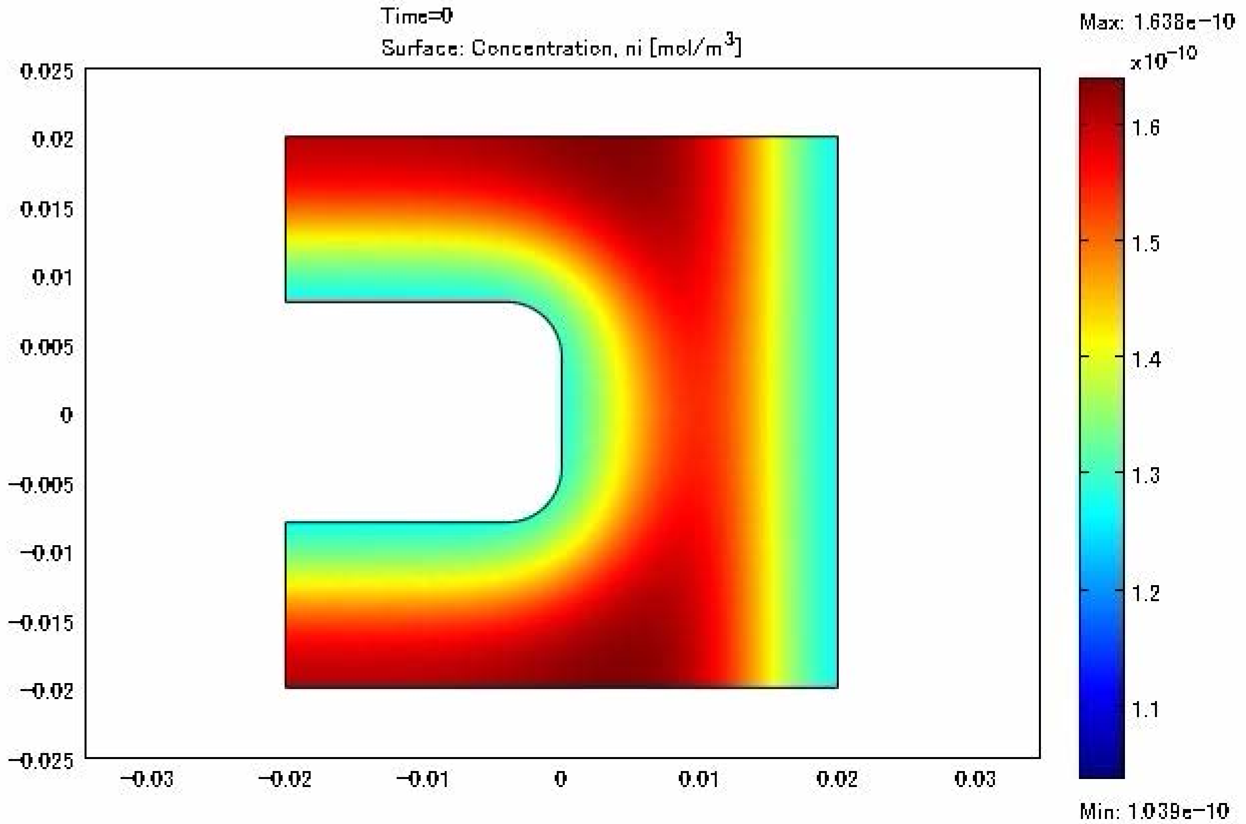


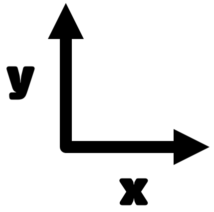


Electrical Discharge (2D CCP)

Ion Density

Ions do not move because of their higher mass

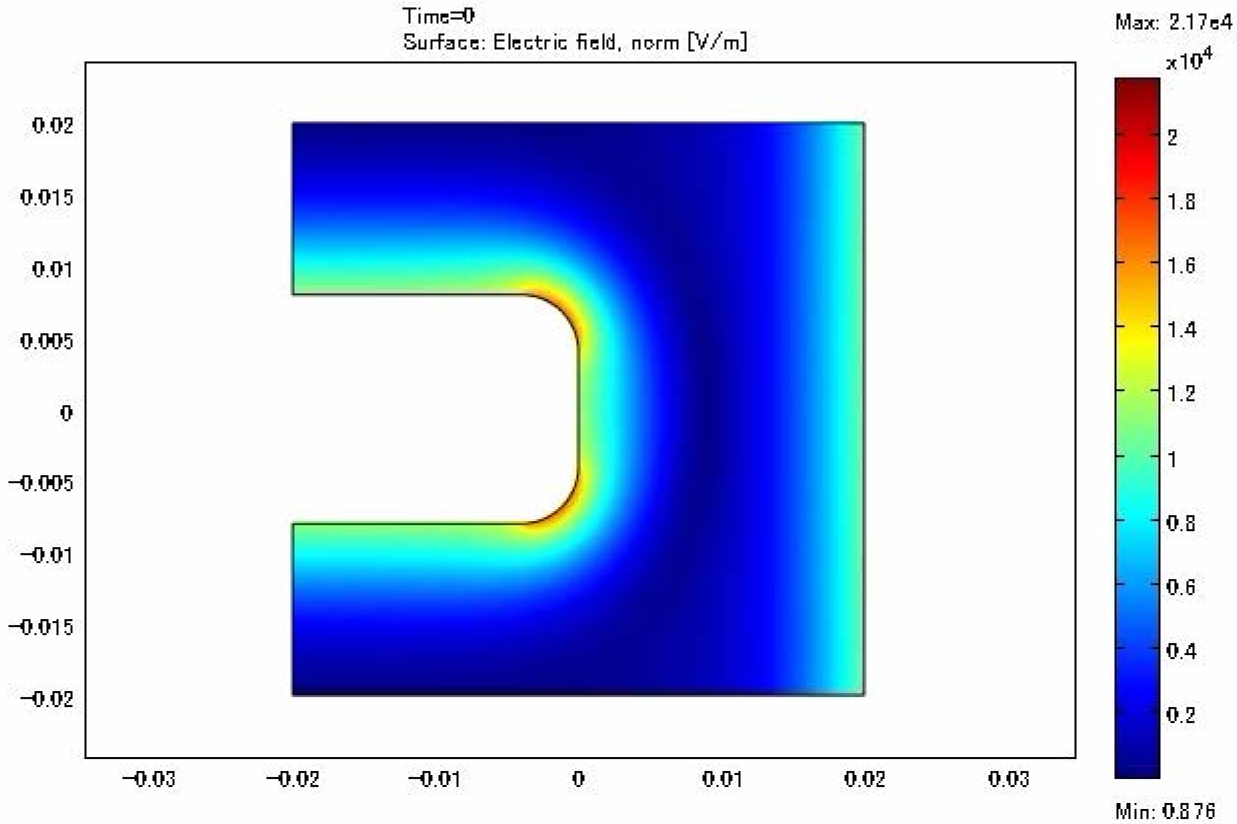


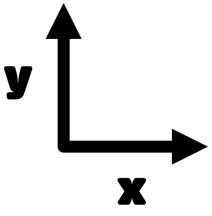


Electrical Discharge (2D CCP)

Electric Field (norm)

EF concentration at the electrode edges can be observed



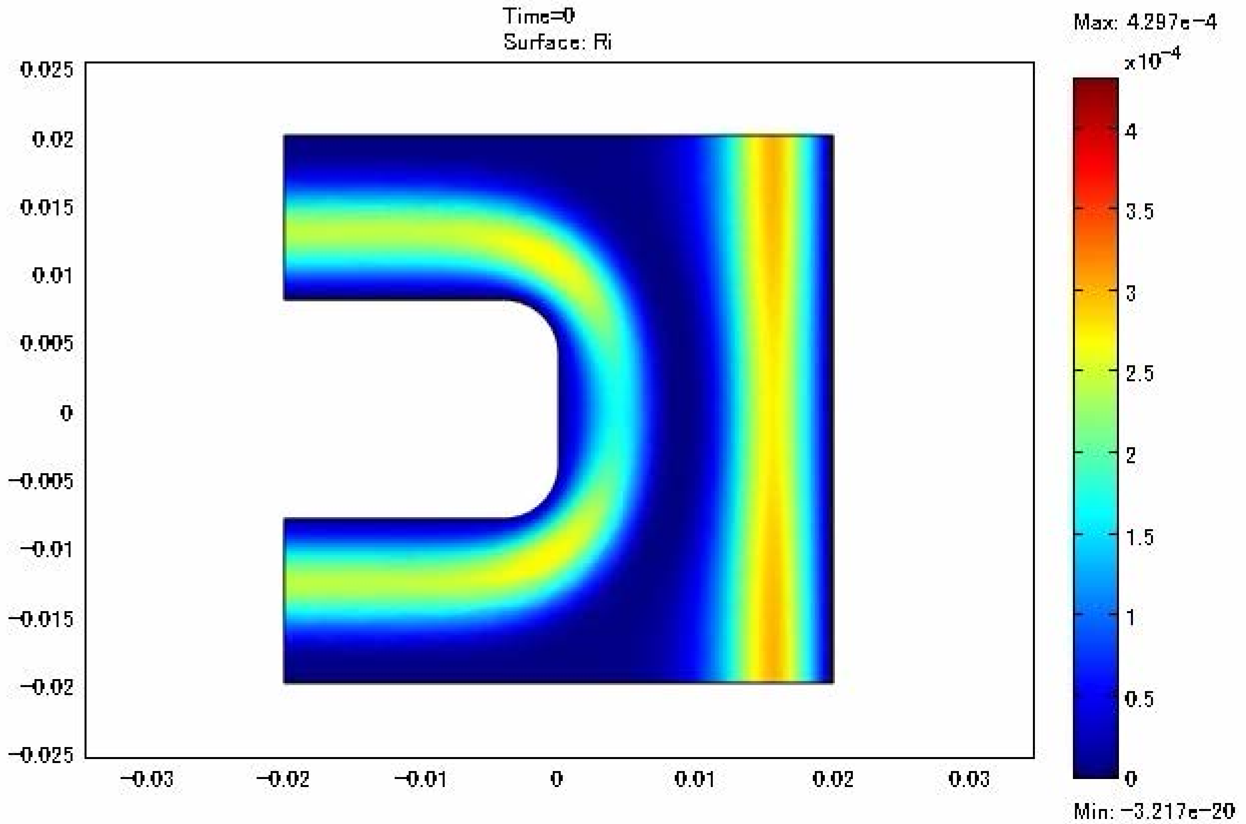


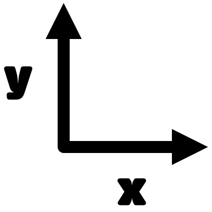
Electrical Discharge (2D CCP)

Ionization Rate

Important information: WHERE? and HOW MUCH?

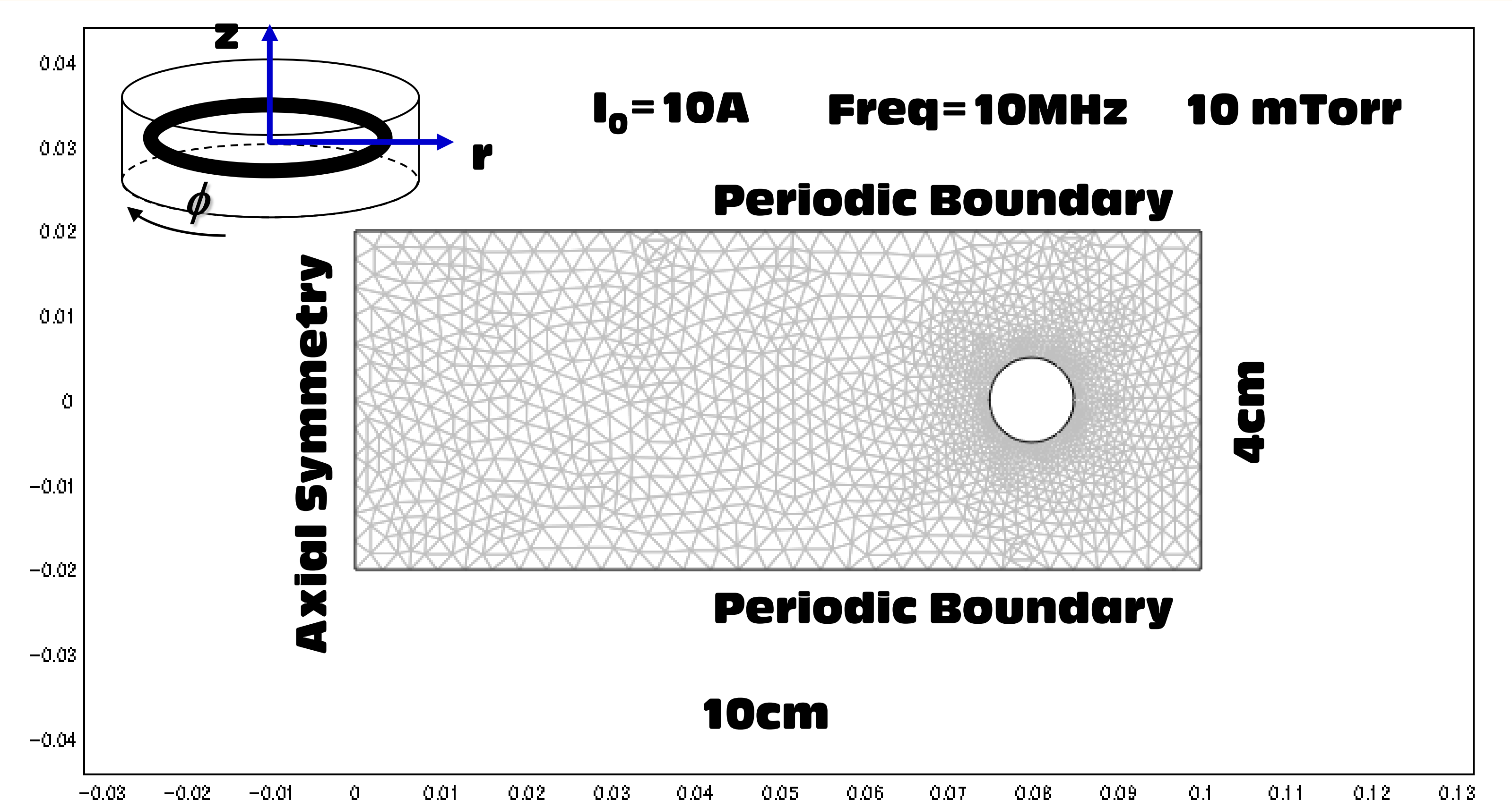
→ Time-averaged Ri profile can be used for Chemical Kinetics Simulation (ex. CVD)





Electrical Discharge (2D ICP Preliminary #1)

Extend from CCP to ICP



Electrical Discharge (2D ICP Preliminary #1)

Convection and Diffusion (for Electrons & Ions)

$$\frac{\partial n_\mathrm{e}}{\partial t} - \nabla(D_\mathrm{e}\nabla n_\mathrm{e} - \upsilon_\mathrm{e} n_\mathrm{e}) = G_\mathrm{e} - R_\mathrm{e} \qquad \frac{\partial n_\mathrm{i}}{\partial t} - \nabla(D_\mathrm{i}\nabla n_\mathrm{i} - \upsilon_\mathrm{i} n_\mathrm{i}) = G_\mathrm{i} - R_\mathrm{i}$$

Equation for Electric Potential V

$$\nabla^2 V = -\frac{\rho}{\varepsilon_0}, \quad \rho = q_0 (n_\mathrm{i} - n_\mathrm{e}) \times N_\mathrm{A}$$

V for Drive
rho for Guide

Equation for Magnetic Potential

$$\mu_0 \sigma \frac{\partial \boldsymbol{A}}{\partial t} + \nabla \times (\nabla \times \boldsymbol{A}) = \mu_0 \boldsymbol{J}^\mathrm{e}, \quad \sigma = (n_\mathrm{e}\mu_\mathrm{e} + n_\mathrm{i}\mu_\mathrm{i}) q_0 \times N_\mathrm{A}$$

J for Drive
sigma for Guide

Electric Field

$$E_r = -\frac{\partial V_r}{\partial r}, \quad E_z = -\frac{\partial V_z}{\partial z}, \quad E_\phi = -\frac{\partial A_\phi}{\partial t}$$

Ionization is assumed to be occurred only by E_ϕ

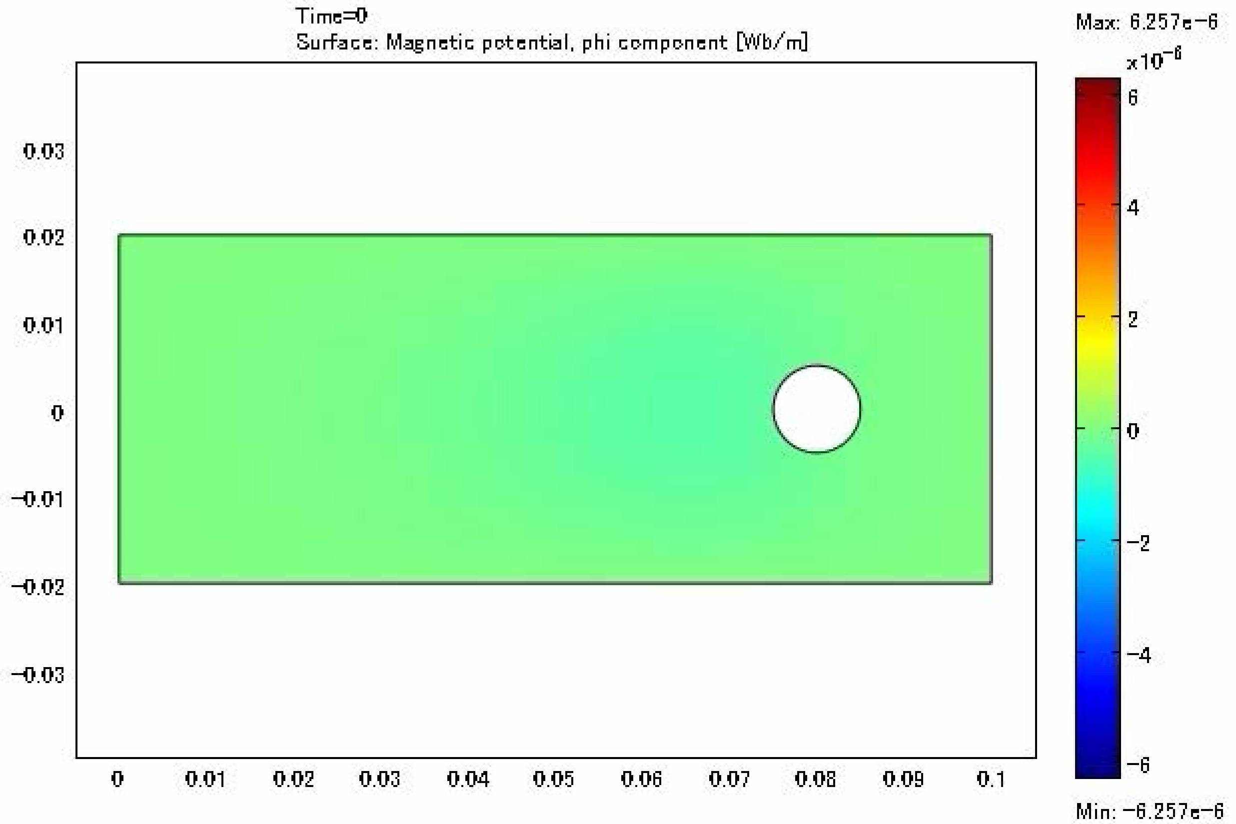
Magnetic Field

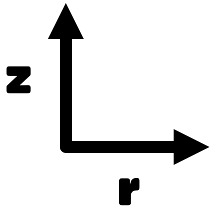
B/N: Not considered yet
ExB effects: Not considered yet

Electrical Discharge (2D ICP Preliminary #1)

Magnetic Potential Aϕ

Main driving force for ionization

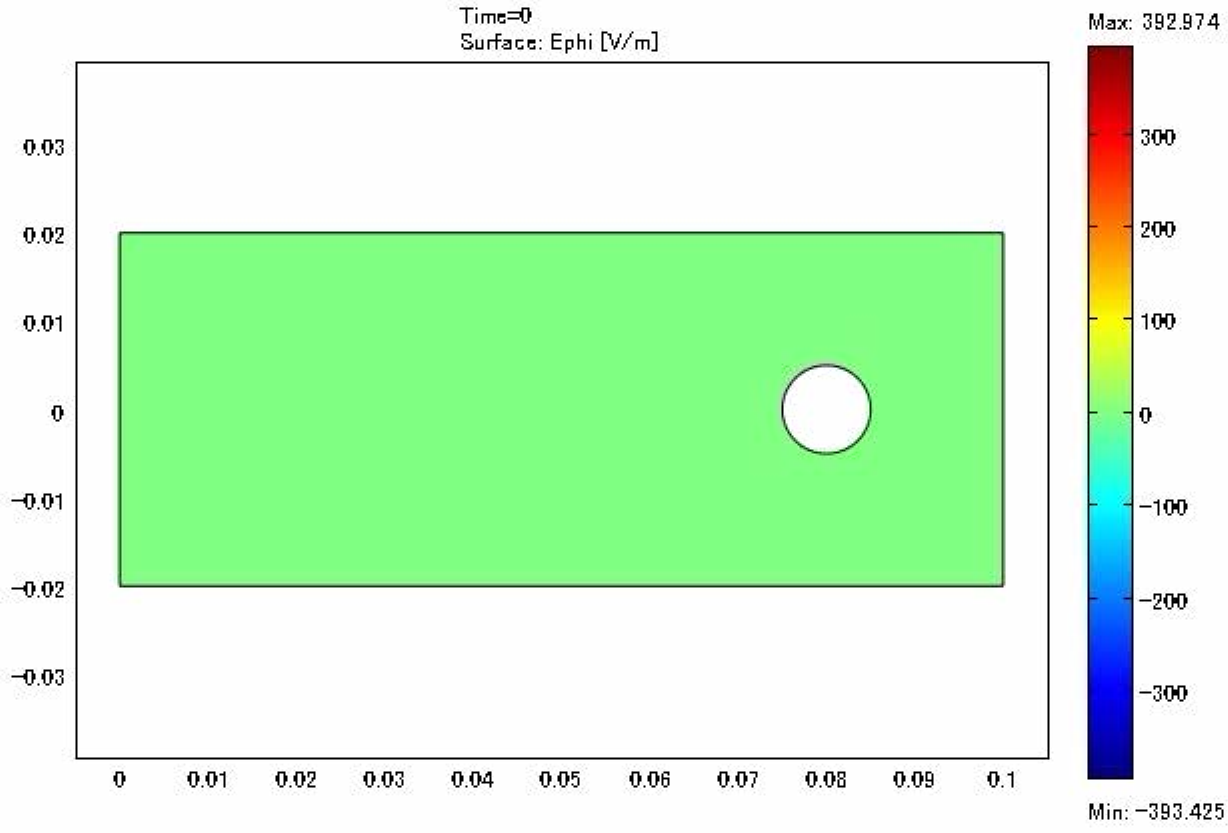


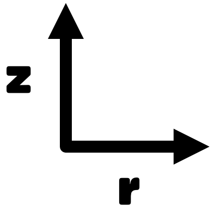


Electrical Discharge (2D ICP Preliminary #1)

Electric Field : ϕ component

$$E_\phi = -\frac{\partial A_\phi}{\partial t}$$



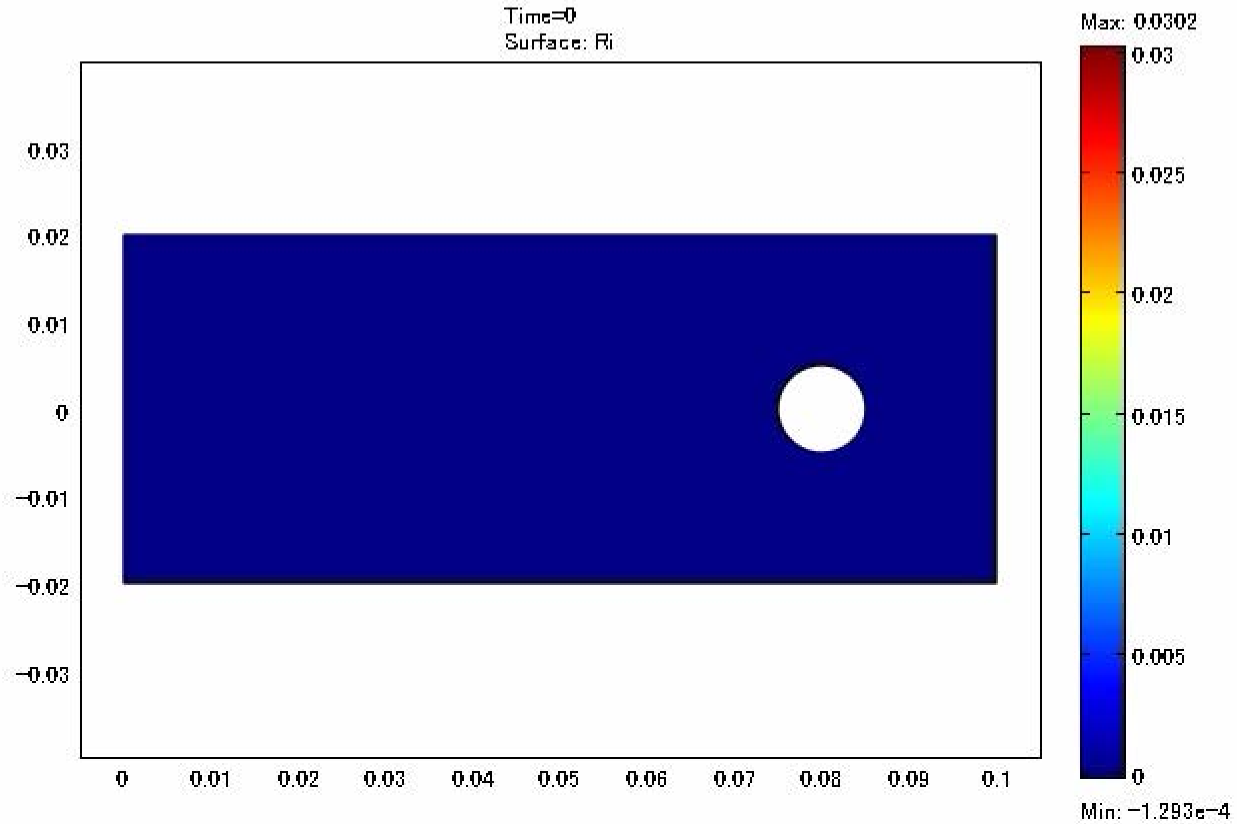


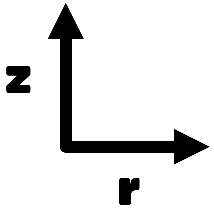
Electrical Discharge (2D ICP Preliminary #1)

Ionization Rate*

$$R_i = \alpha(E_\phi / P)\upsilon_e n_e$$

***only Eϕ is considered at this moment**

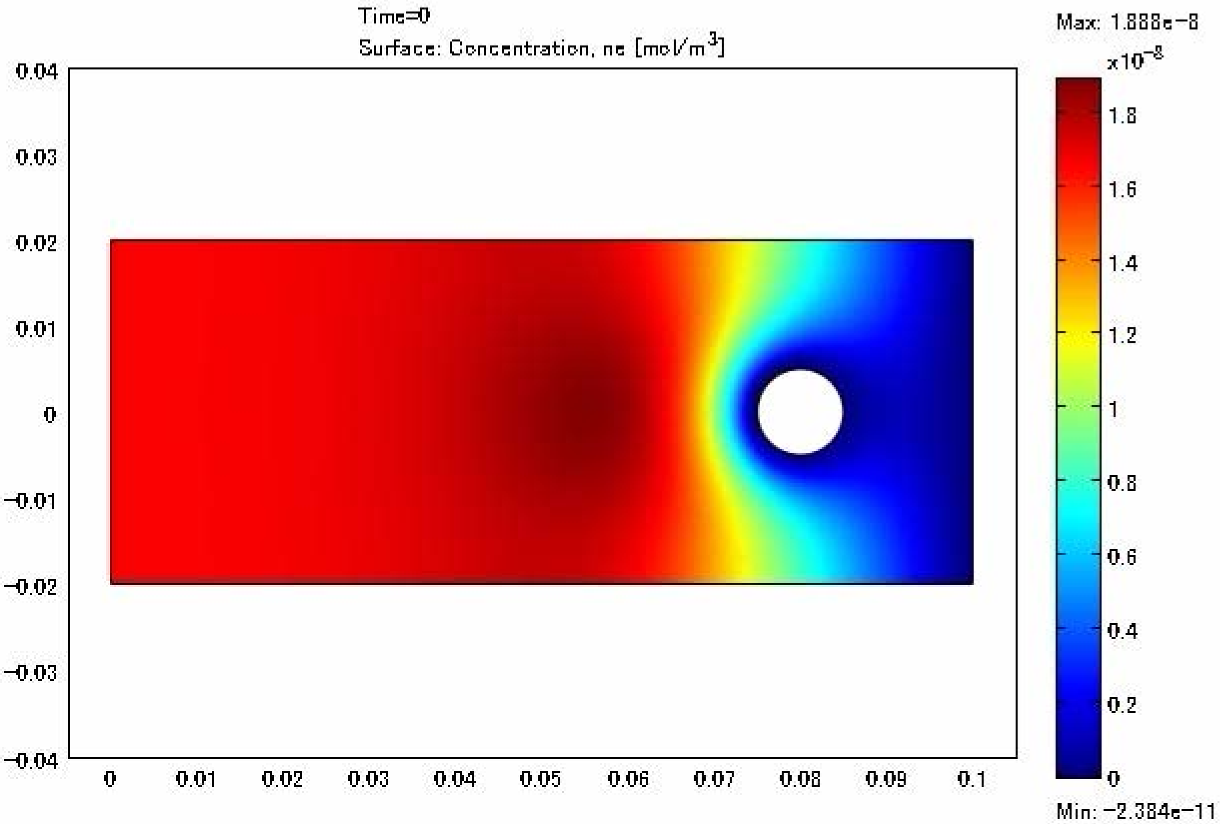


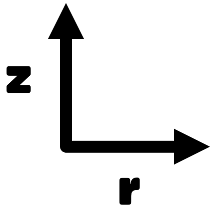


Electrical Discharge (2D ICP Preliminary #1)

Electron density

$2 \times 10^{-8}\ \mathrm{mol/m^3} = 1.2 \times 10^{16}\ /\mathrm{m^3} = 1.2 \times 10^{10}\ /\mathrm{cm^3}$

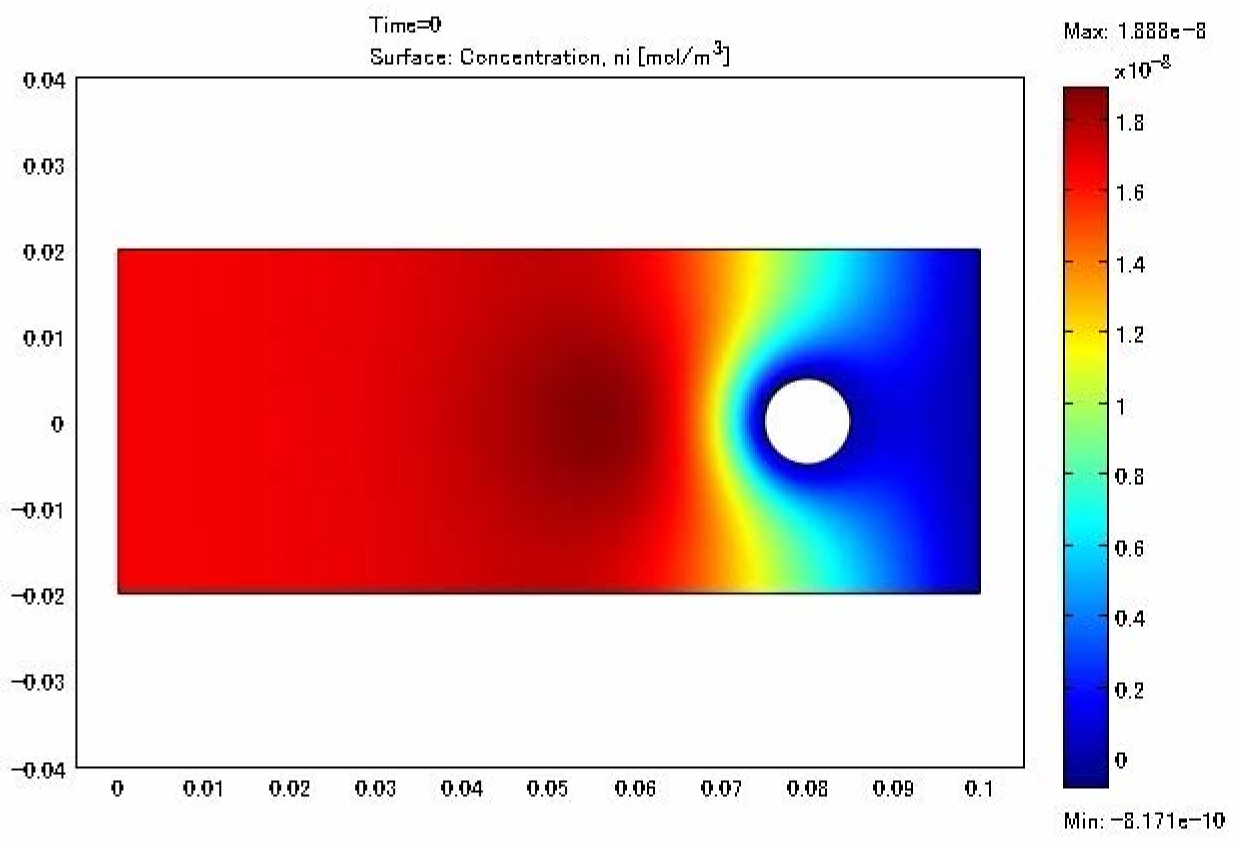


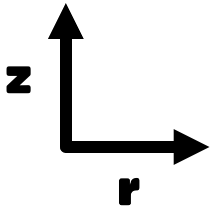


Electrical Discharge (2D ICP Preliminary #1)

Ion density

2×10^{-8} mol/m^3 = 1.2×10^{16} /m^3 = 1.2×10^{10} /cm^3

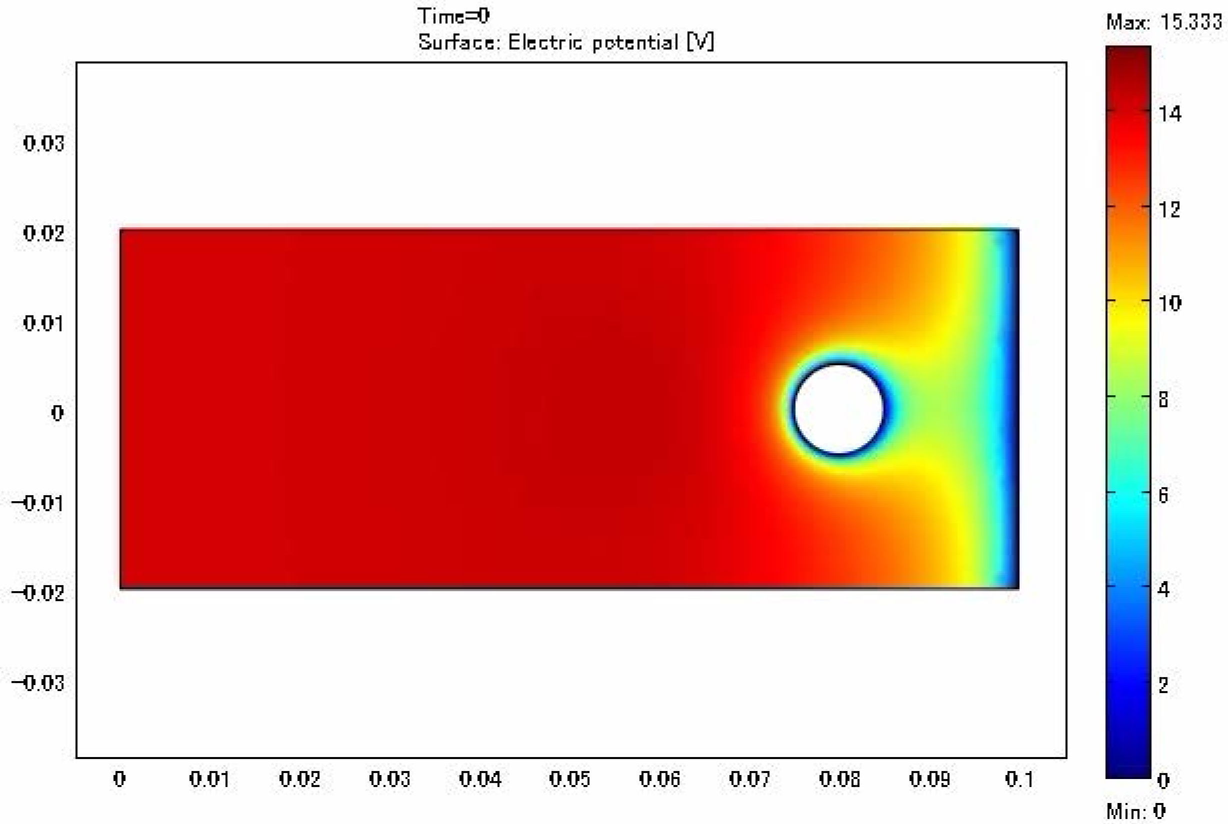


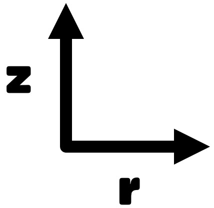


Electrical Discharge (2D ICP Preliminary #1)

Potential

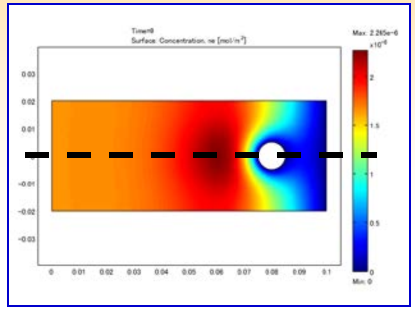
Vp = 15 V; Low plasma potential





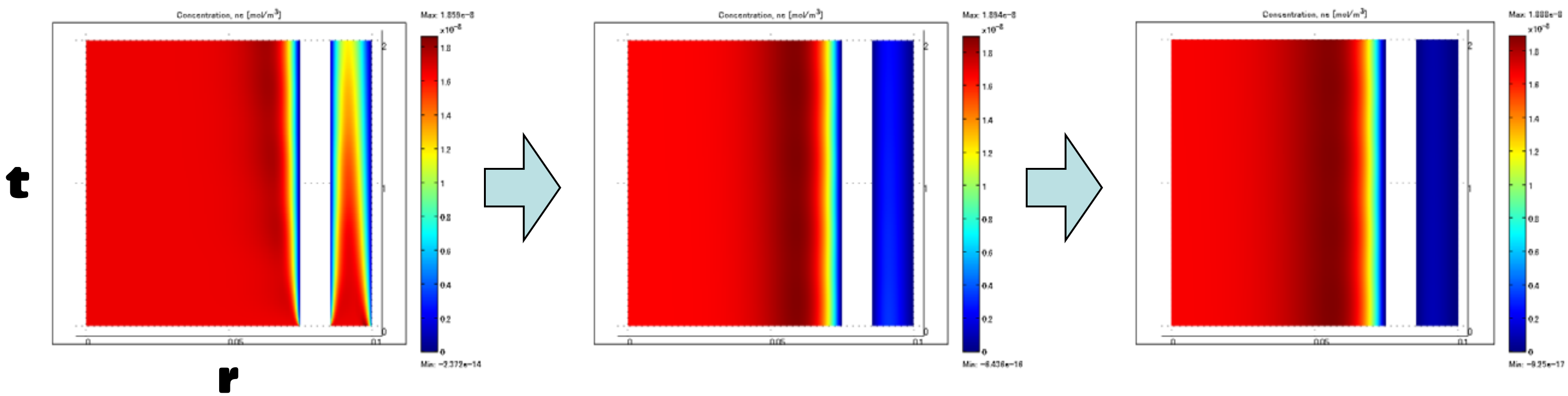
Electrical Discharge (2D ICP Preliminary #1)

Electron density



Initiation

Almost steady state

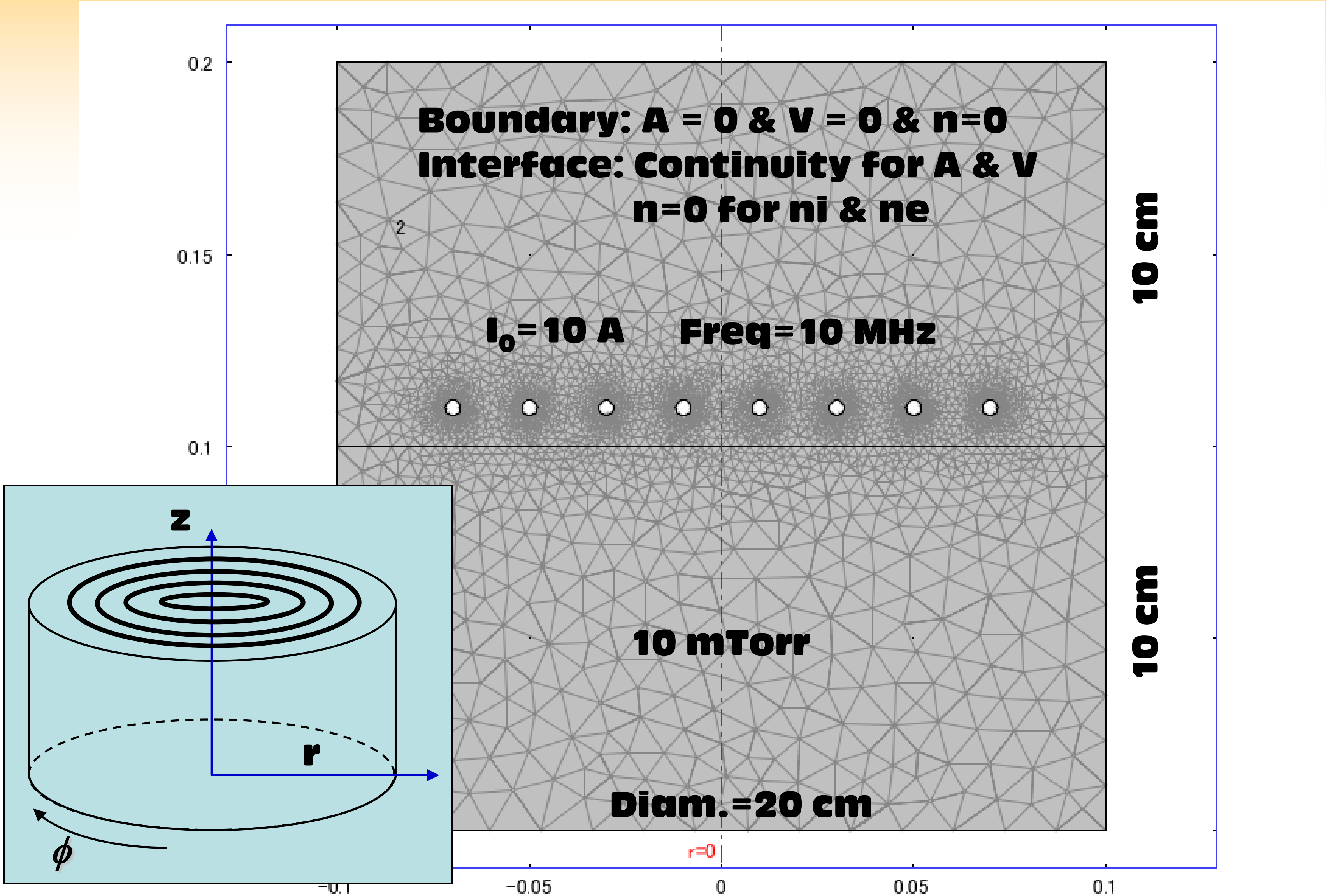


1st Cycle

5th Cycle

10th Cycle

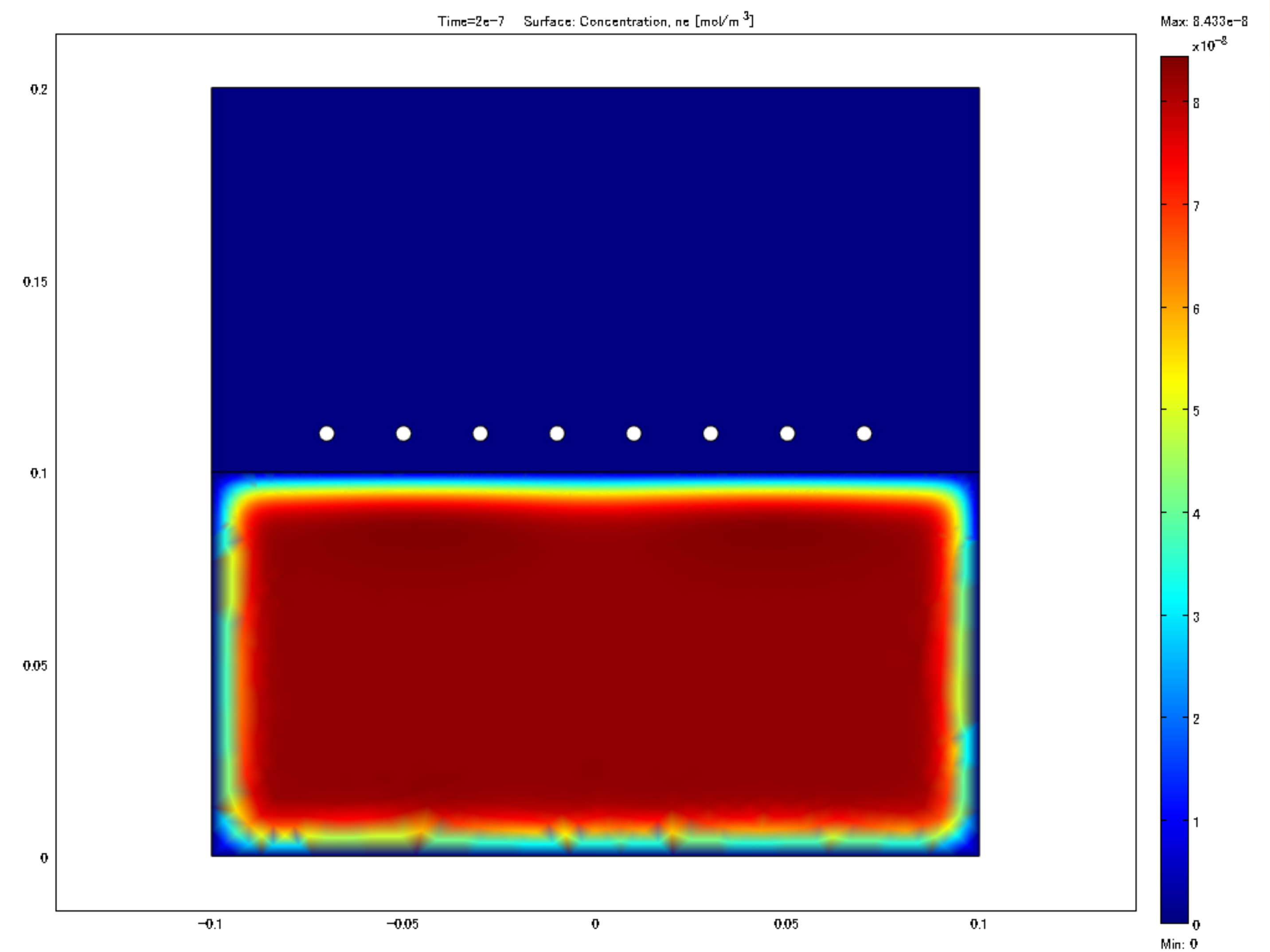
Electrical Discharge (2D ICP Preliminary #2)



Electrical Discharge (2D ICP Preliminary #2)

Electron Density

After 1 Cycle of RF
(almost same to initial profile)

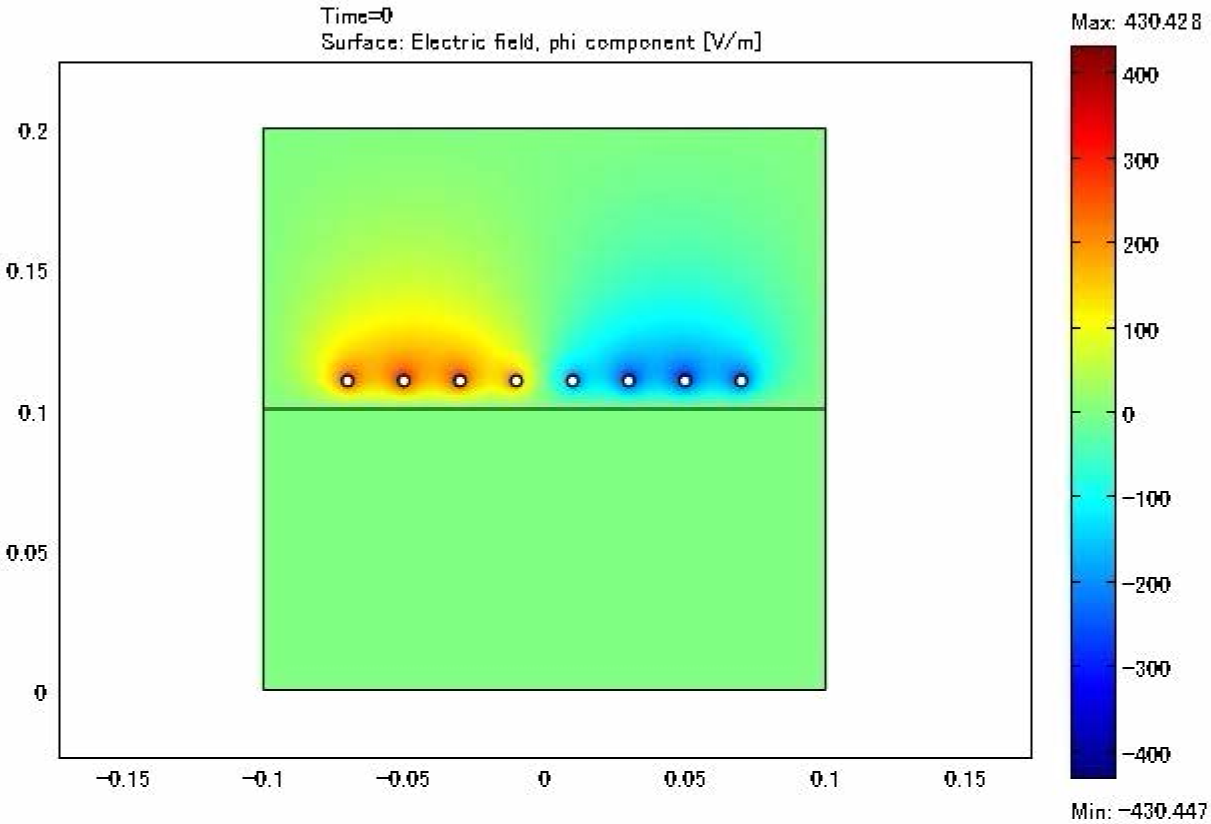


Electrical Discharge (2D ICP Preliminary #2)

Electric Field ϕ component

After 1 Cycle of RF

Explicit Skin Effect at the interface because of high density of n_e

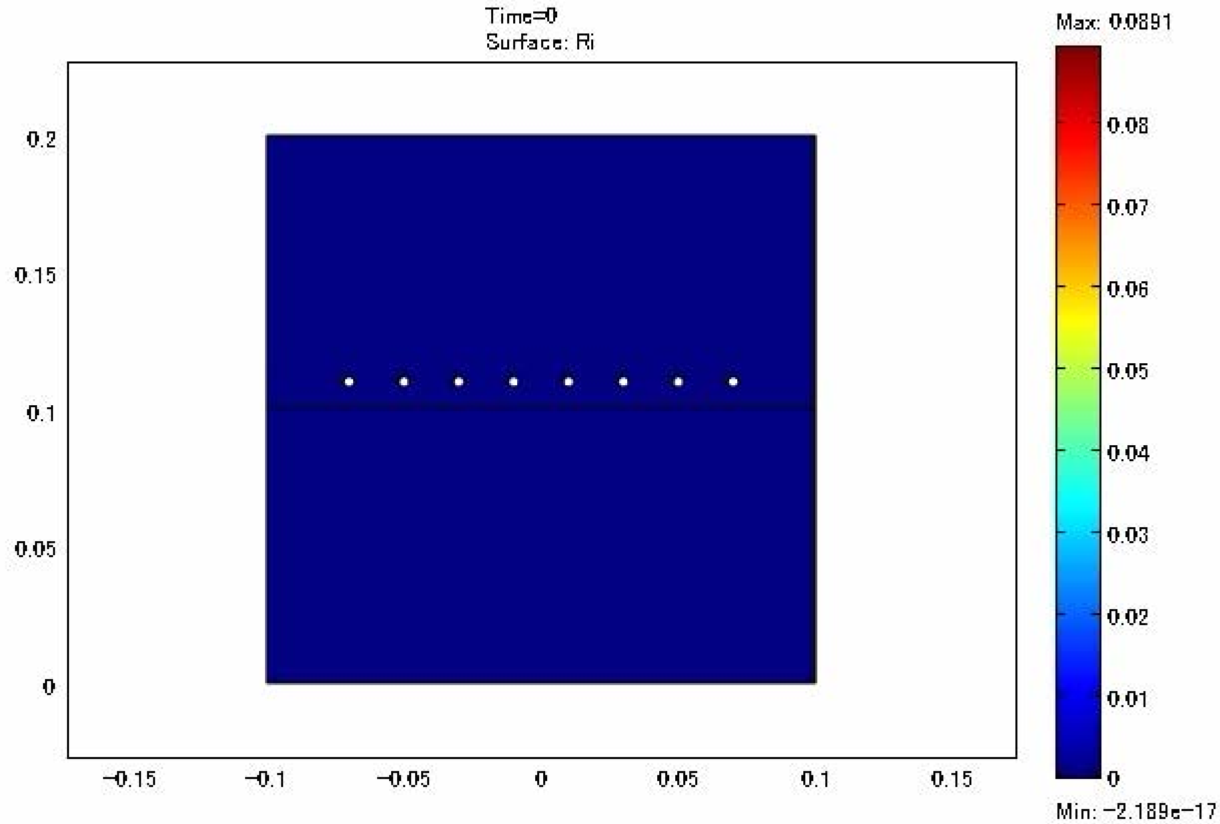


Electrical Discharge (2D ICP Preliminary #2)

Ionization Rate

After 1 Cycle of RF

Explicit Skin Effect at the interface

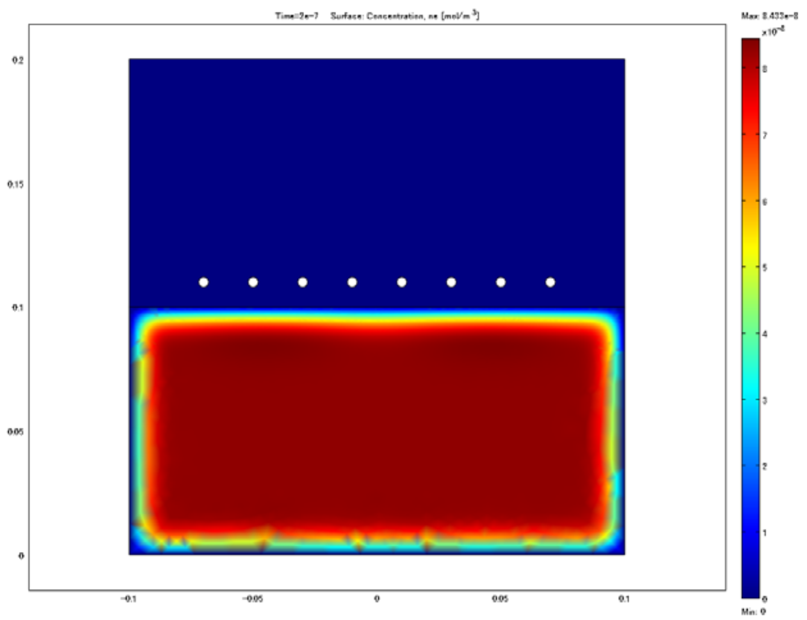


Electrical Discharge (2D ICP Preliminary #2)

Iteration until steady state

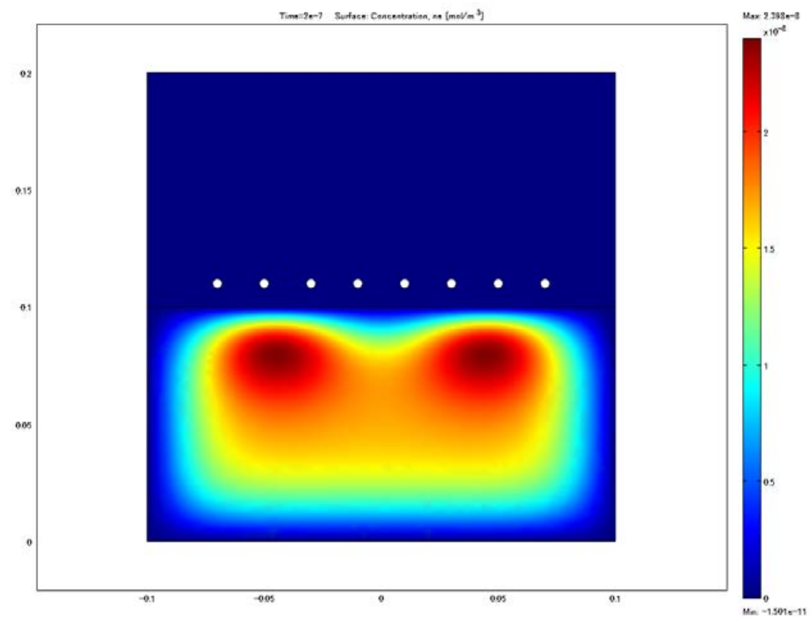
After 1 Cycle of RF

8.4×10^{-8} mol/m³



After 10 Cycle of RF

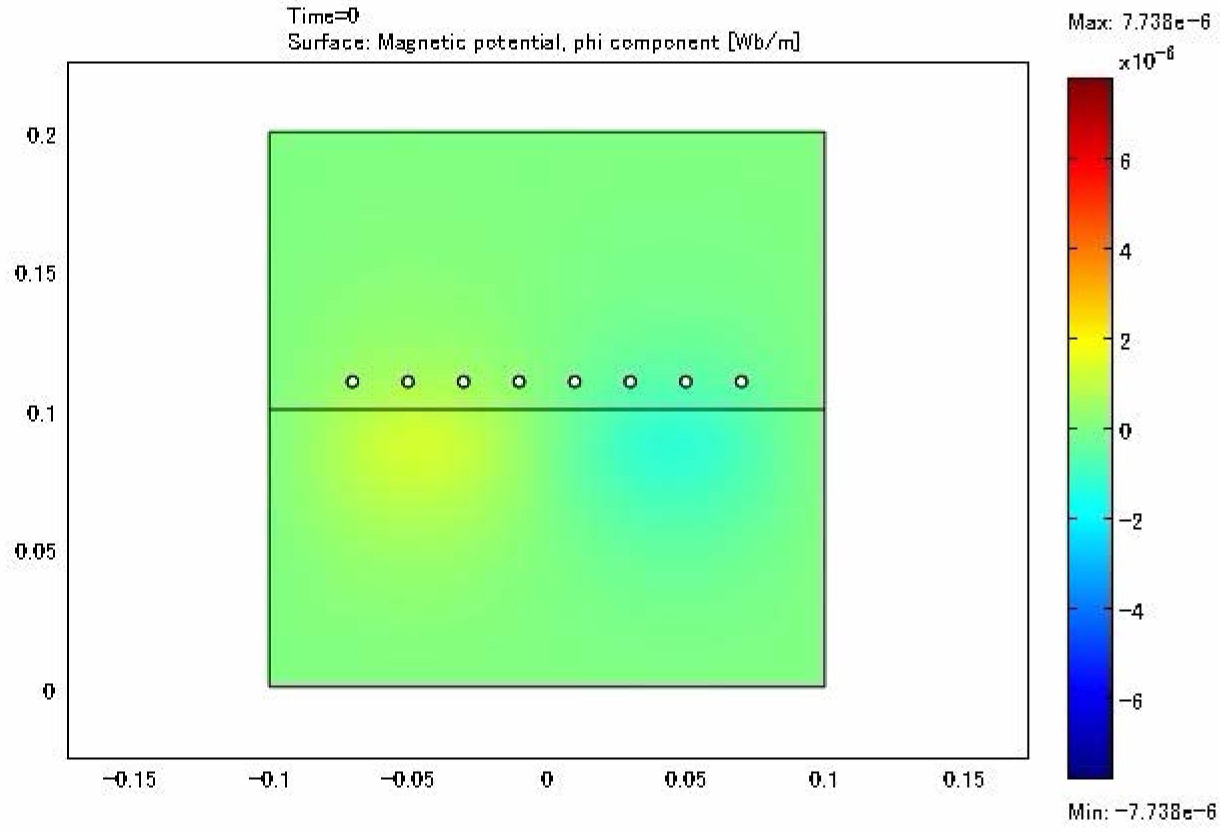
2.4×10^{-8} mol/m³

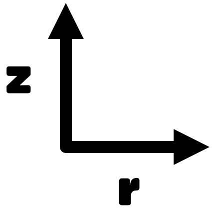


Electrical Discharge (2D ICP Preliminary #2)

Magnetic Potential Aϕ

does not fully penetrate into the plasma region because of skin effect

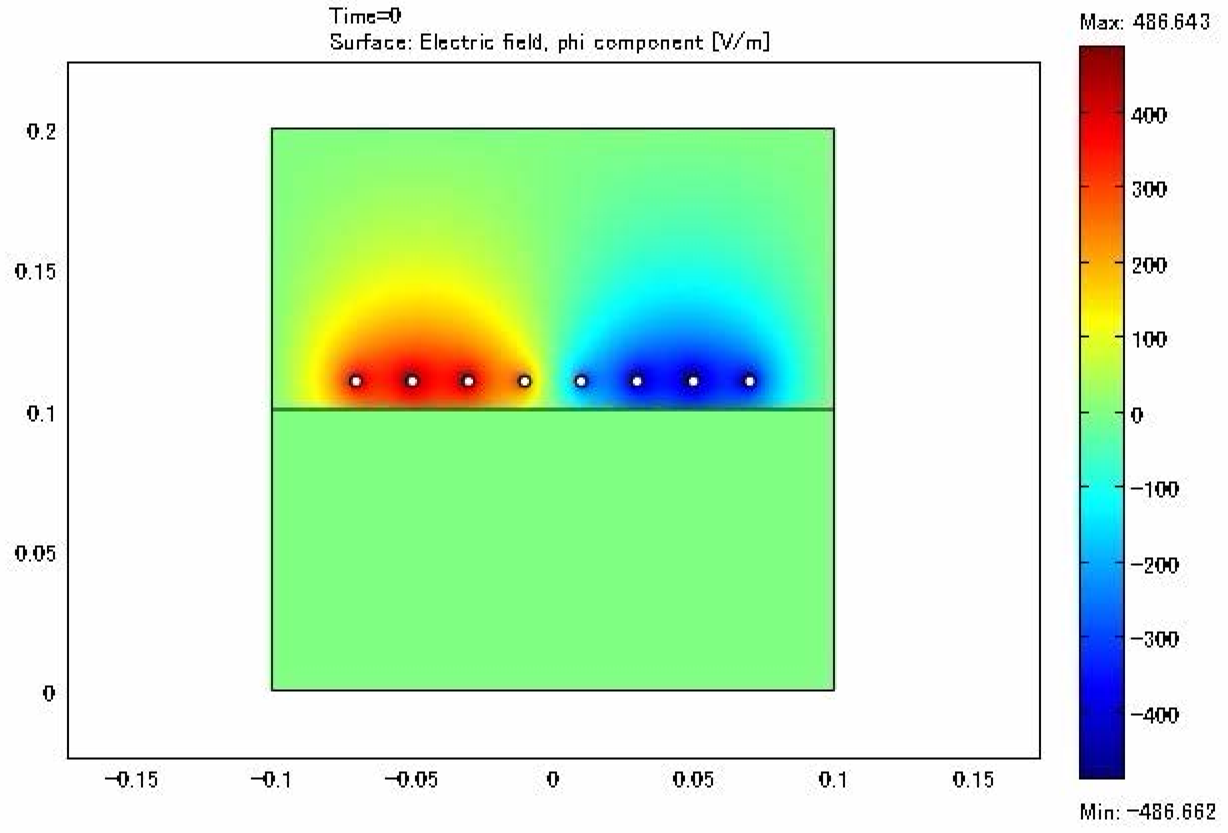


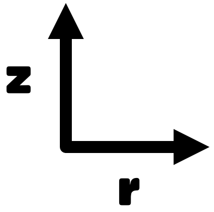


Electrical Discharge (2D ICP Preliminary #2)

Electric Field ϕ component

EF = - dA/dt
A = sin(wt) → EF = cos(wt) ; Phase shifted



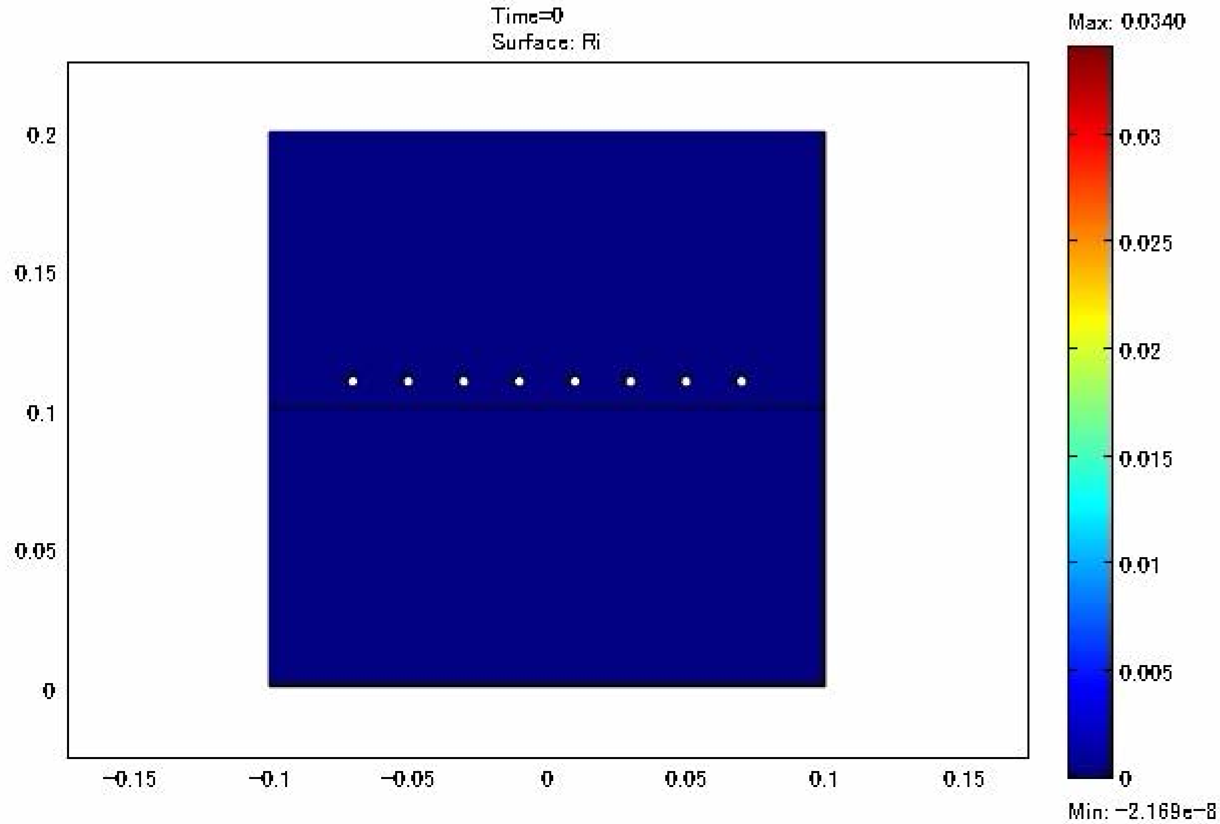


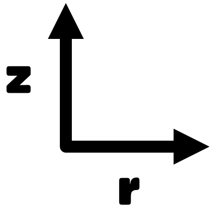
Electrical Discharge (2D ICP Preliminary #2)

Ionization Rate*

$R_i = \alpha \, v_e \, n_e \qquad v_e = \mu E$

Ignition at the interface where EF is high and ne is moderate. After that, with reduction of EF, Ri peak position moves down to the position which has higher ne.

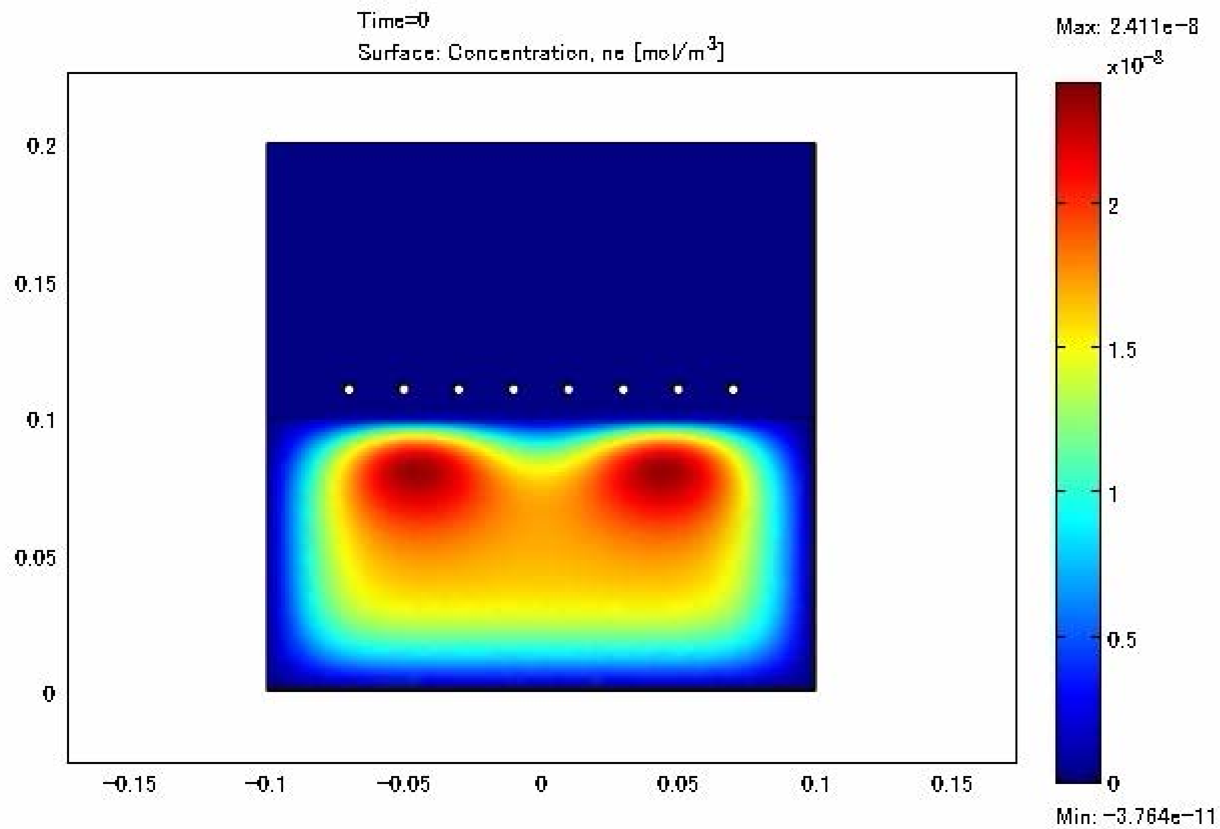


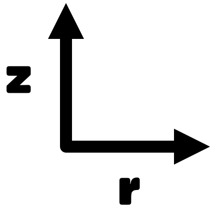


Electrical Discharge (2D ICP Preliminary #2)

Electron density

8×10^{-8} mol/m³ = 4.8×10^{16} /m³ = 4.8×10^{10} /cm³

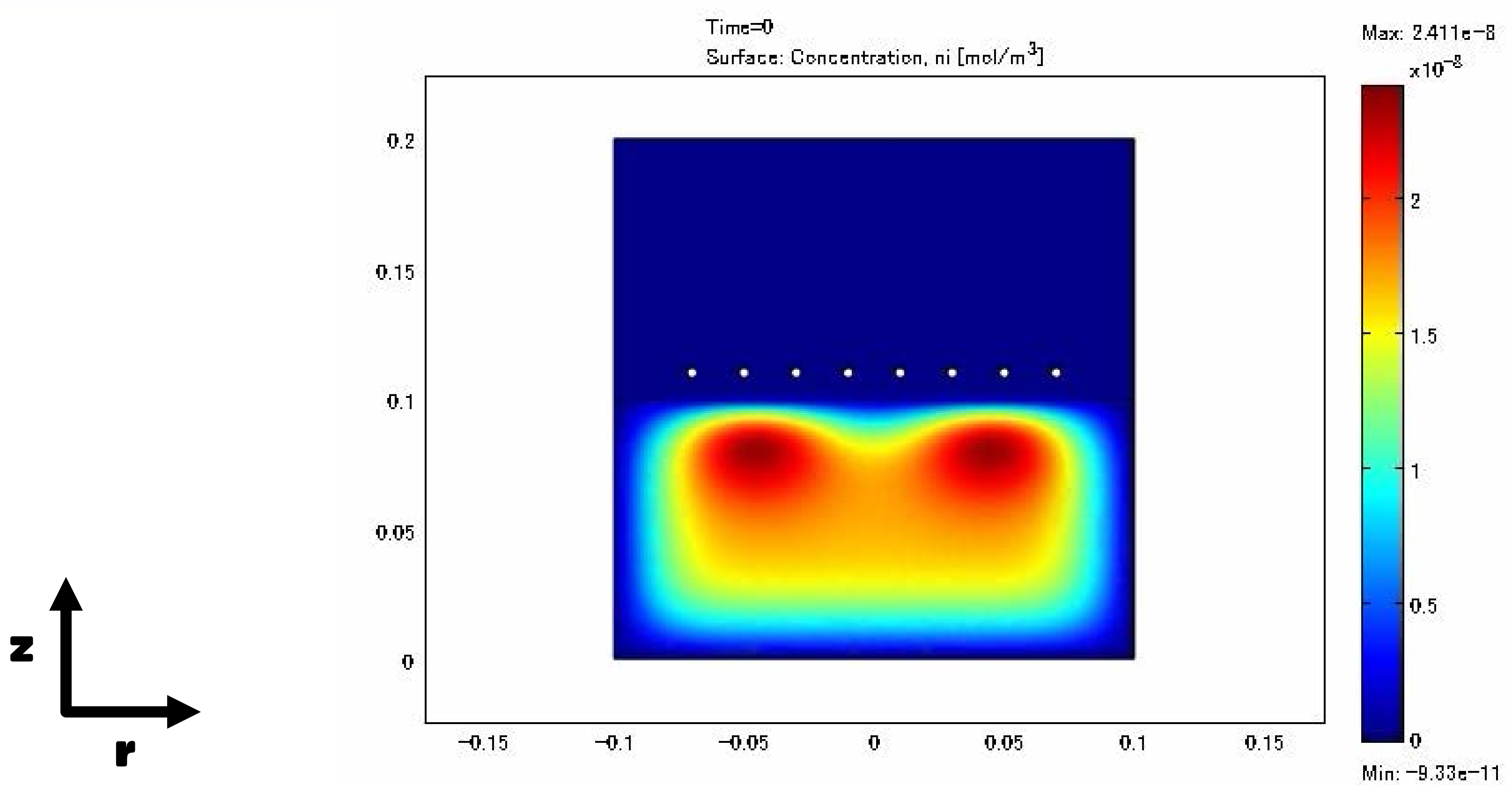




Electrical Discharge (2D ICP Preliminary #2)

Ion density

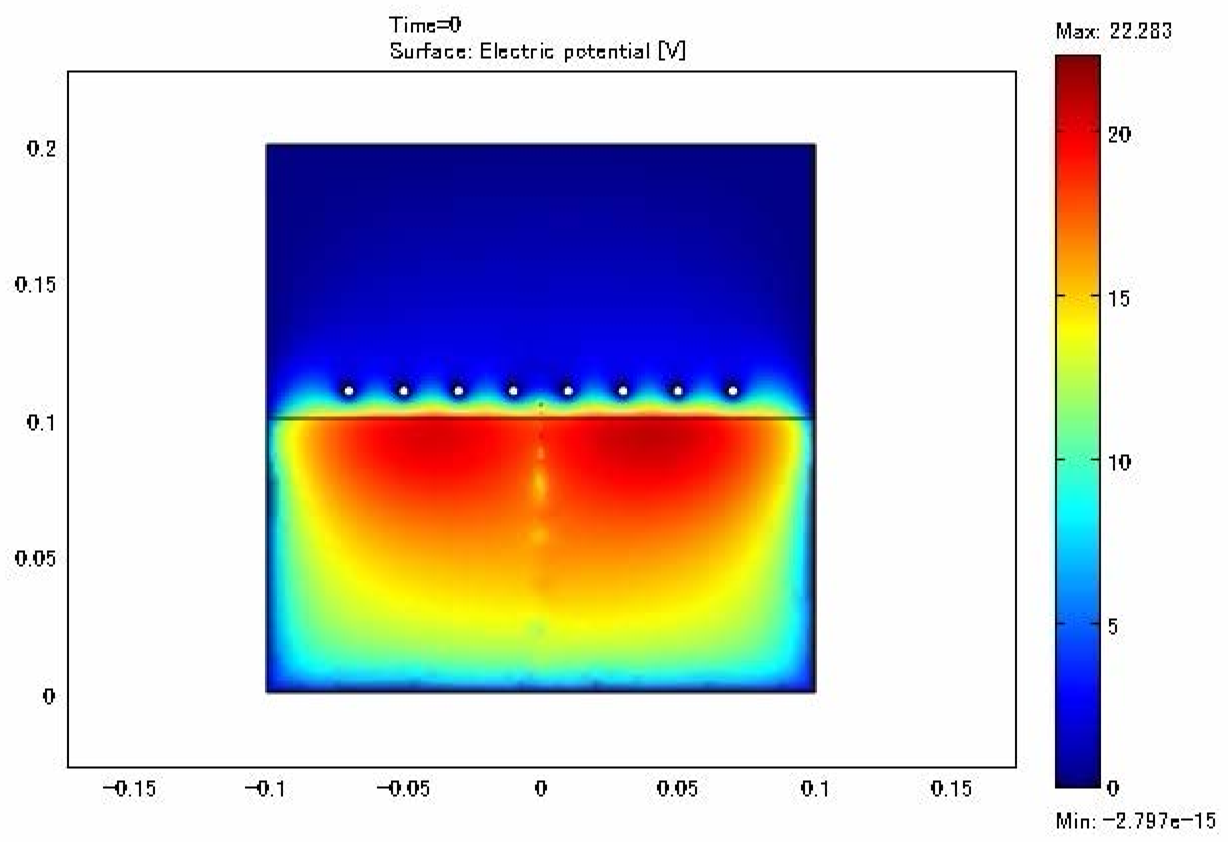
8×10^{-8} mol/m³ = 4.8×10^{16} /m³ = 4.8×10^{10} /cm³

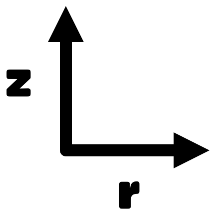


Electrical Discharge (2D ICP Preliminary #2)

Potential

approx. 22 V
represents low plasma potential





Limitation of LFA model (QTE, RCT must be tried)

But,

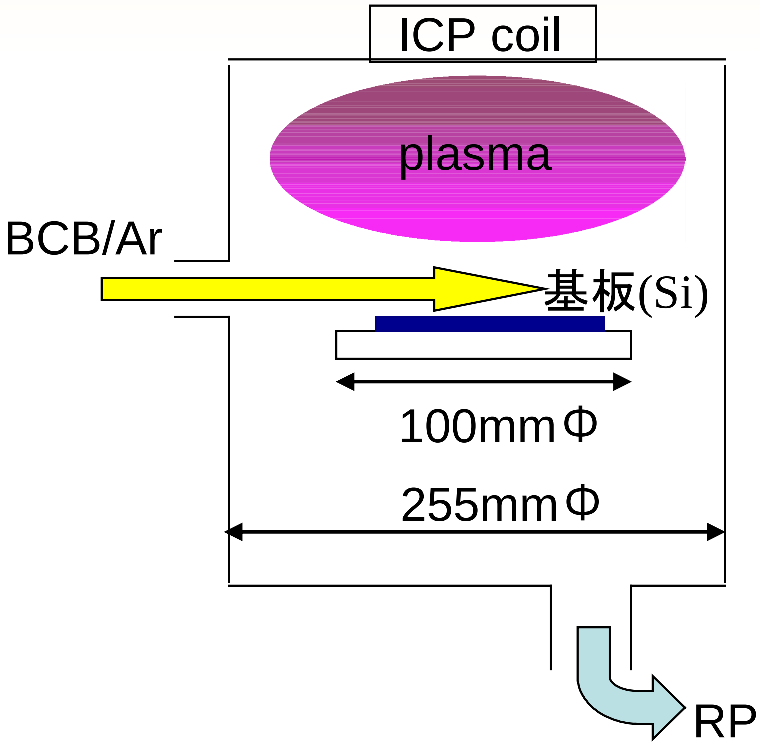
Good for Qualitative Understanding of the behavior of Ne, Ni and EF in plasmas (Educational Purpose)

B. Chemical Reactions and Thin Film Deposition

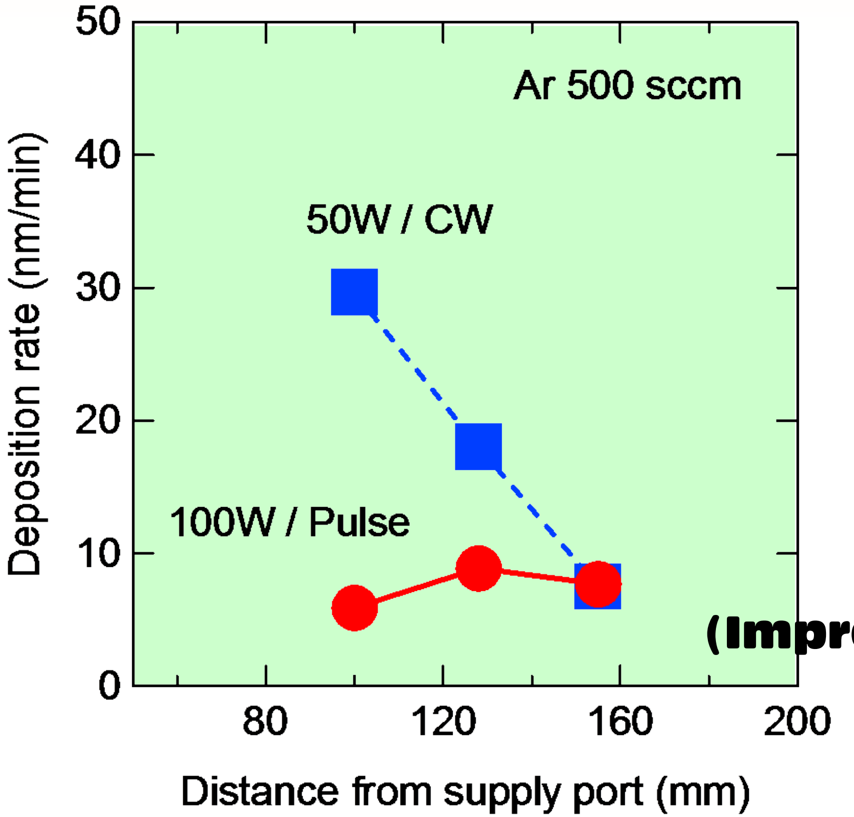
1. Introduction
2. Governing Equations
3. Solver Parameters
4. Results and Discussion

Experimental Results

Spatial Non-Uniformity



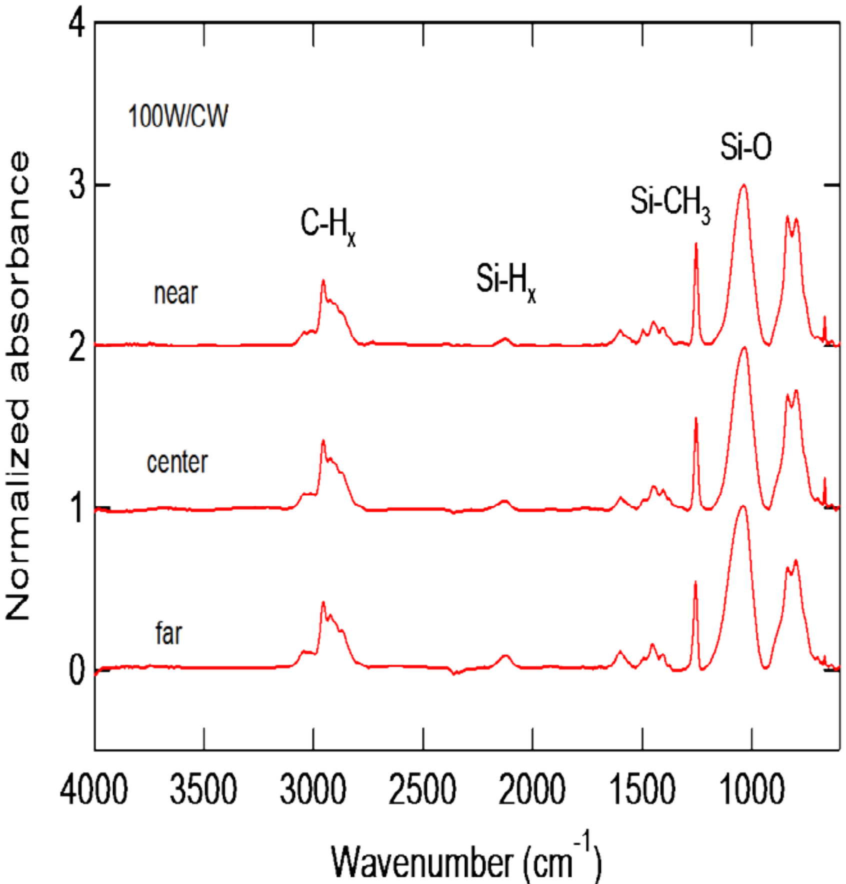
Non Uniformity of Deposition Rate



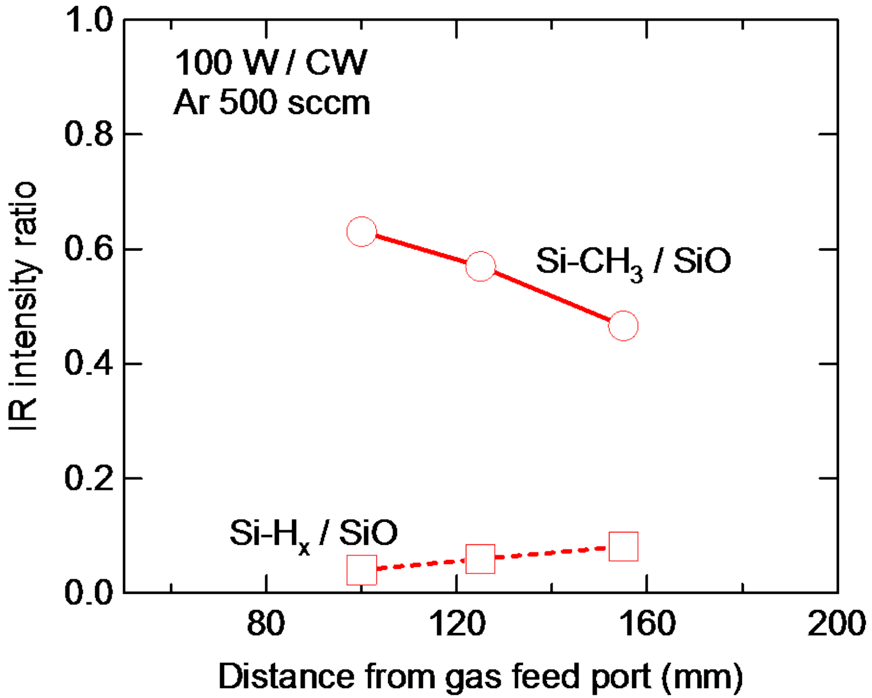
(Improved by Pulsing)

Non Uniformity of Film Composition

← Variation of Deposition Precursors
(Explained by Cumulative (or Multi-Step) Dissociation and production of different precursors at each stage)



Reduction of Methyl Group
→ Increase of k-value



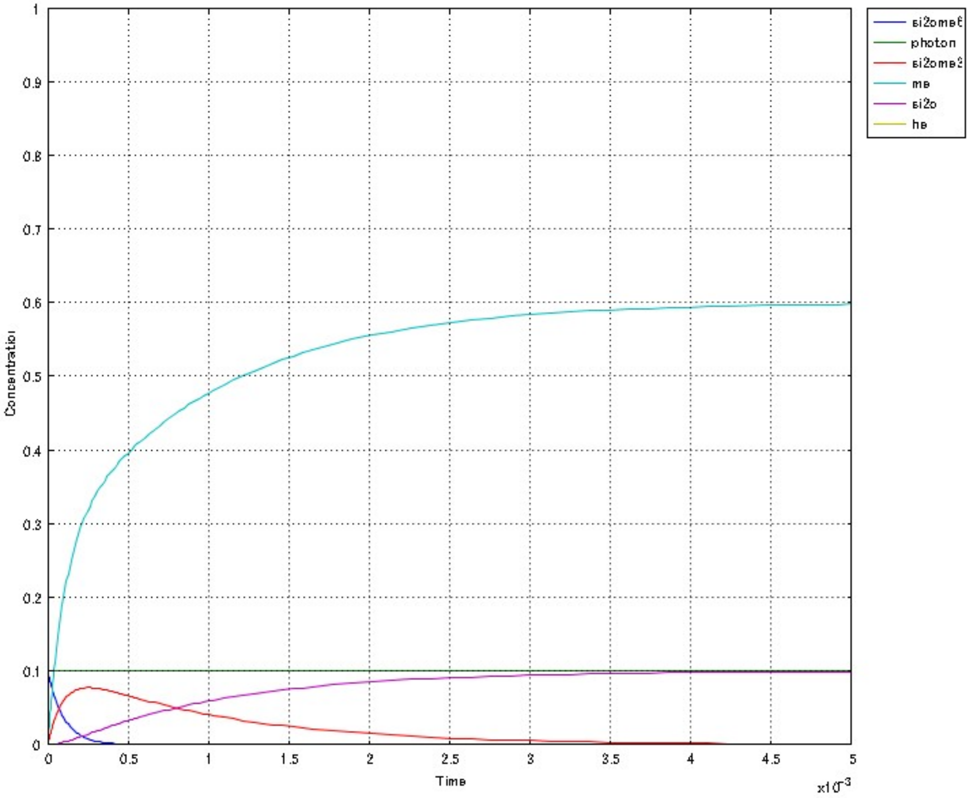
Simple Model

HMDSO: $(CH_3)_3$-Si-O-Si-$(CH_3)_3$

1st Step
si2ome6 + e
→ si2ome3 + 3me + e
(with methyl group)

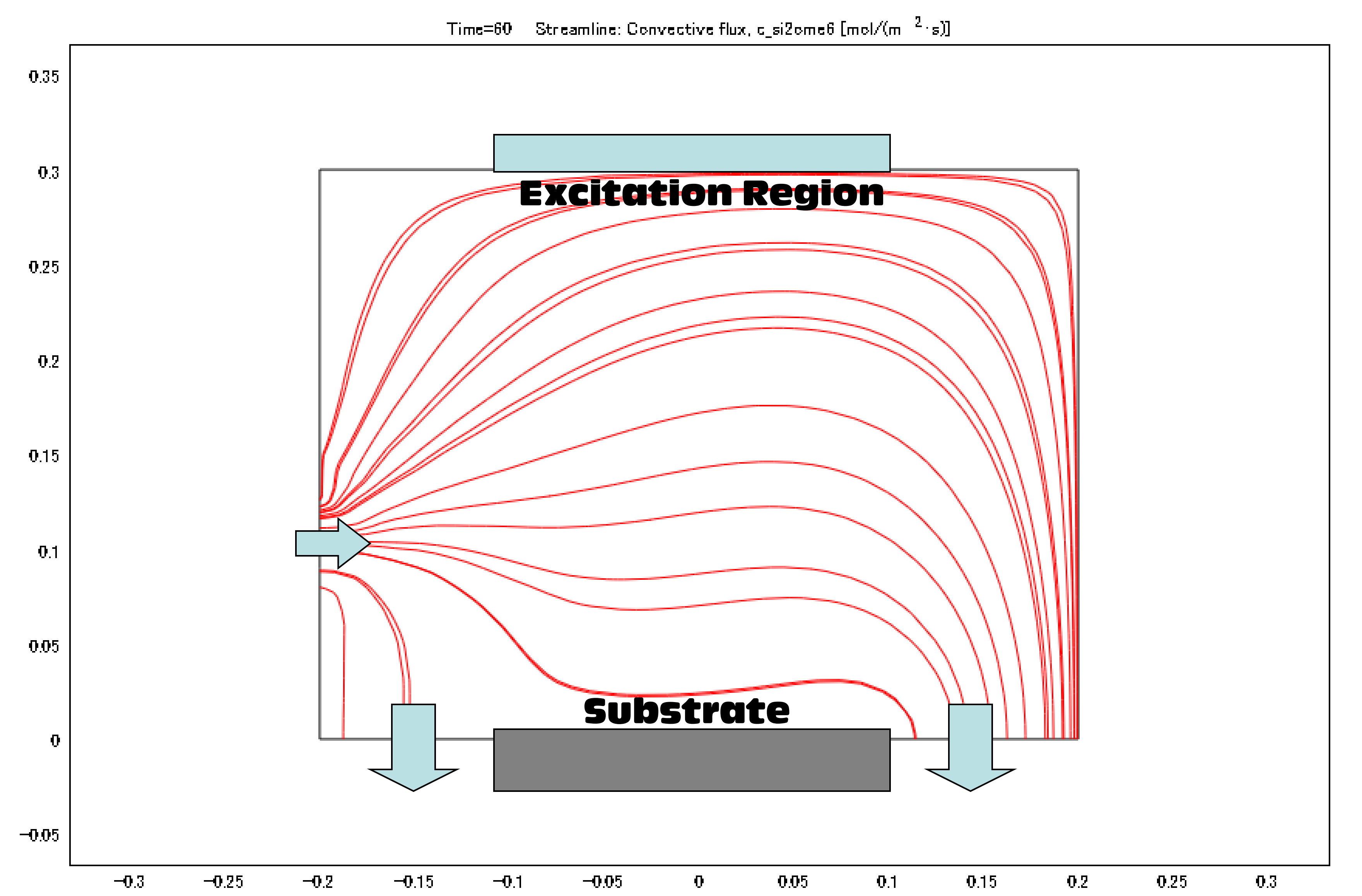
2nd Step
si2ome3 + e
→ si2o + 3me + e
(without methyl group)

- dN/dt = k N ne → k' N



Adjust k' so as to match the experimental results

Transport



Time=60 Streamline: Convective flux, c_si2ome6 [mol/(m 2·s)]
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
−0.05
−0.3
−0.25
−0.2
−0.15
−0.1
−0.05
0
0.05
0.1
0.15
0.2
0.25
0.3
Excitation Region
Substrate

Boundary Conditions

Depends on Sticking Probability β

β = 0 (not deposition precursors)

→ Insulation / Symmetry

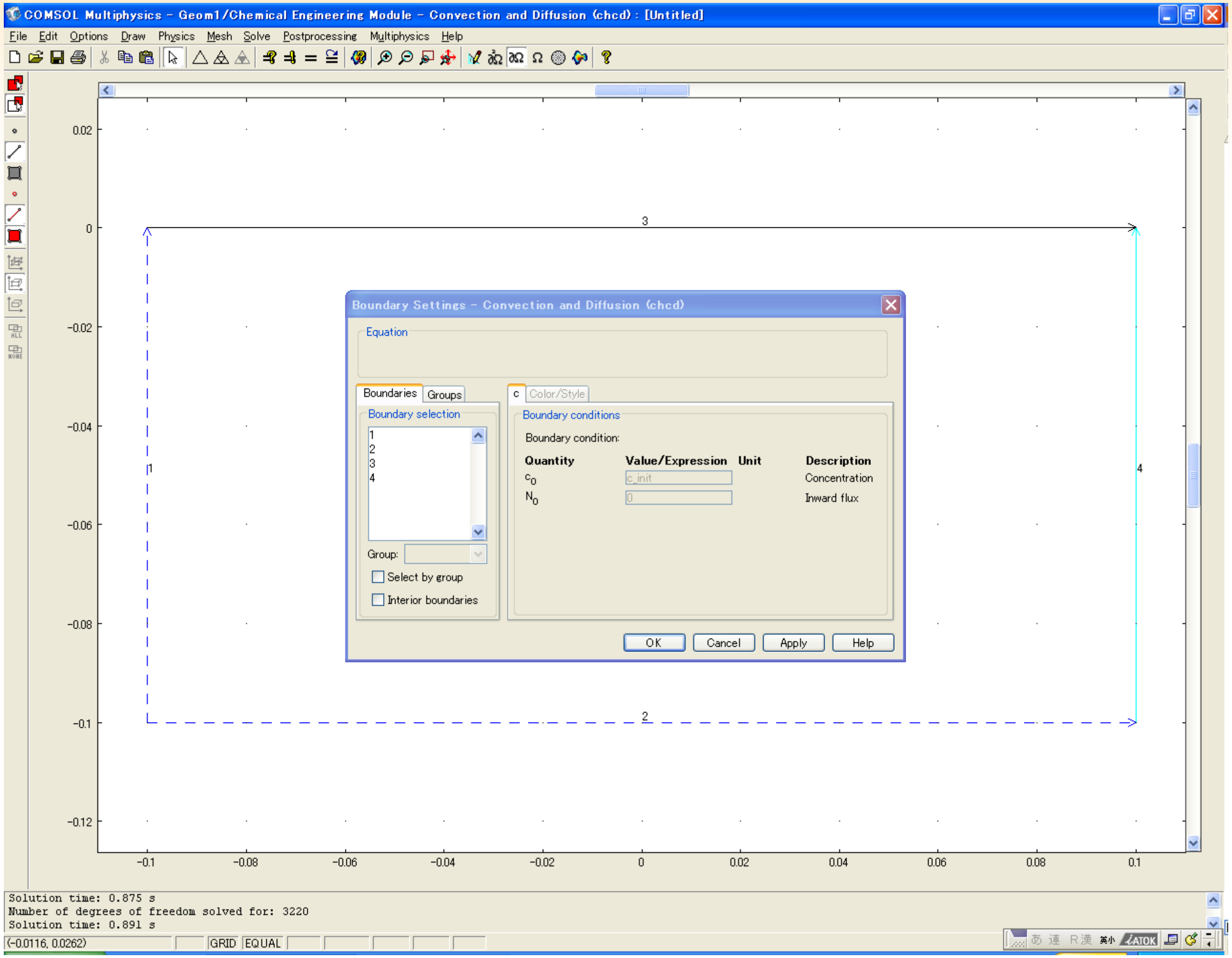
$\beta > 0$ (can be deposition precursors)

```
Flux =
-b_si2ome3*c_si2ome3*(Rg*T_gas)^0.5/(2*pi*M_si2ome3)^0.5
```

β

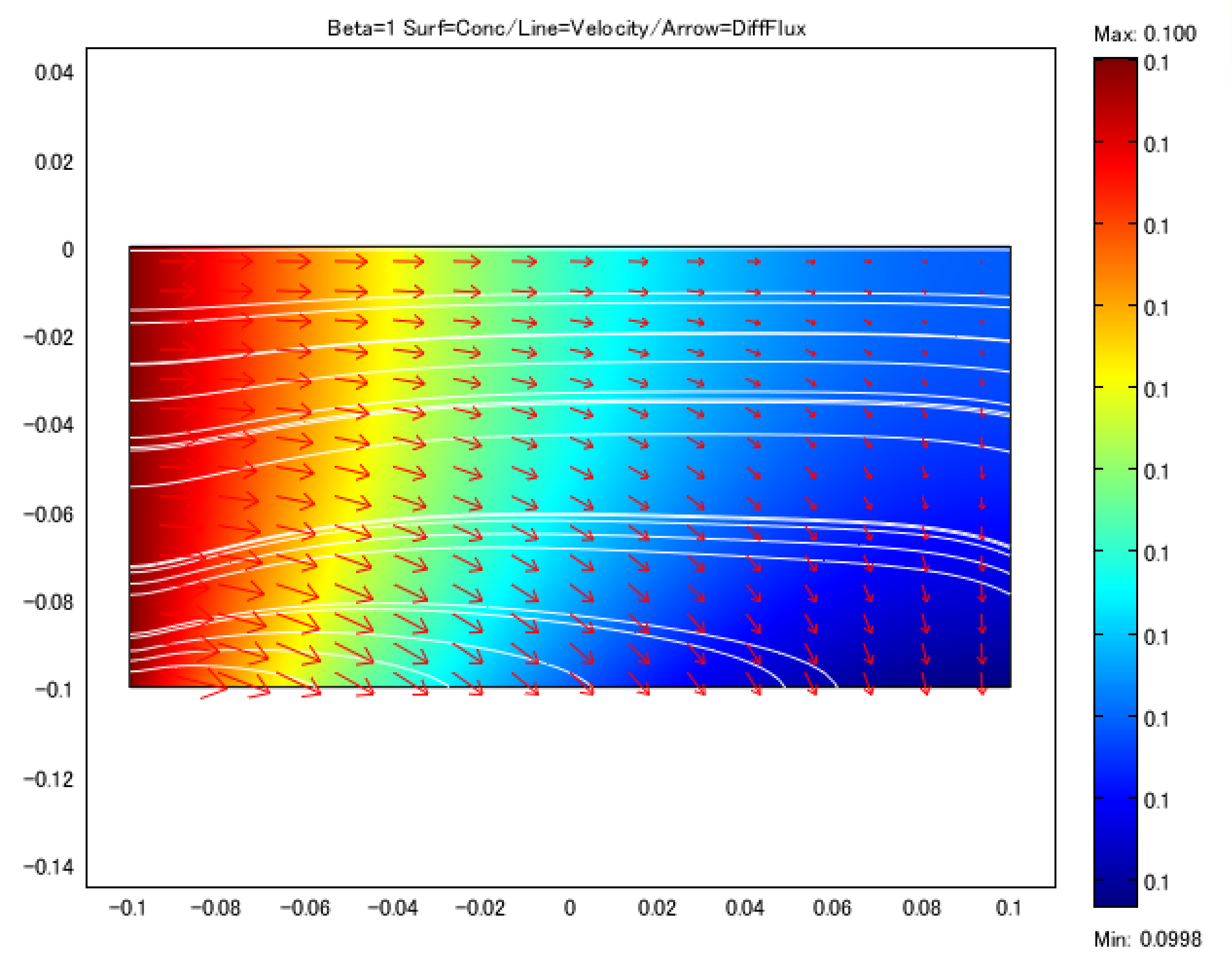
Transport

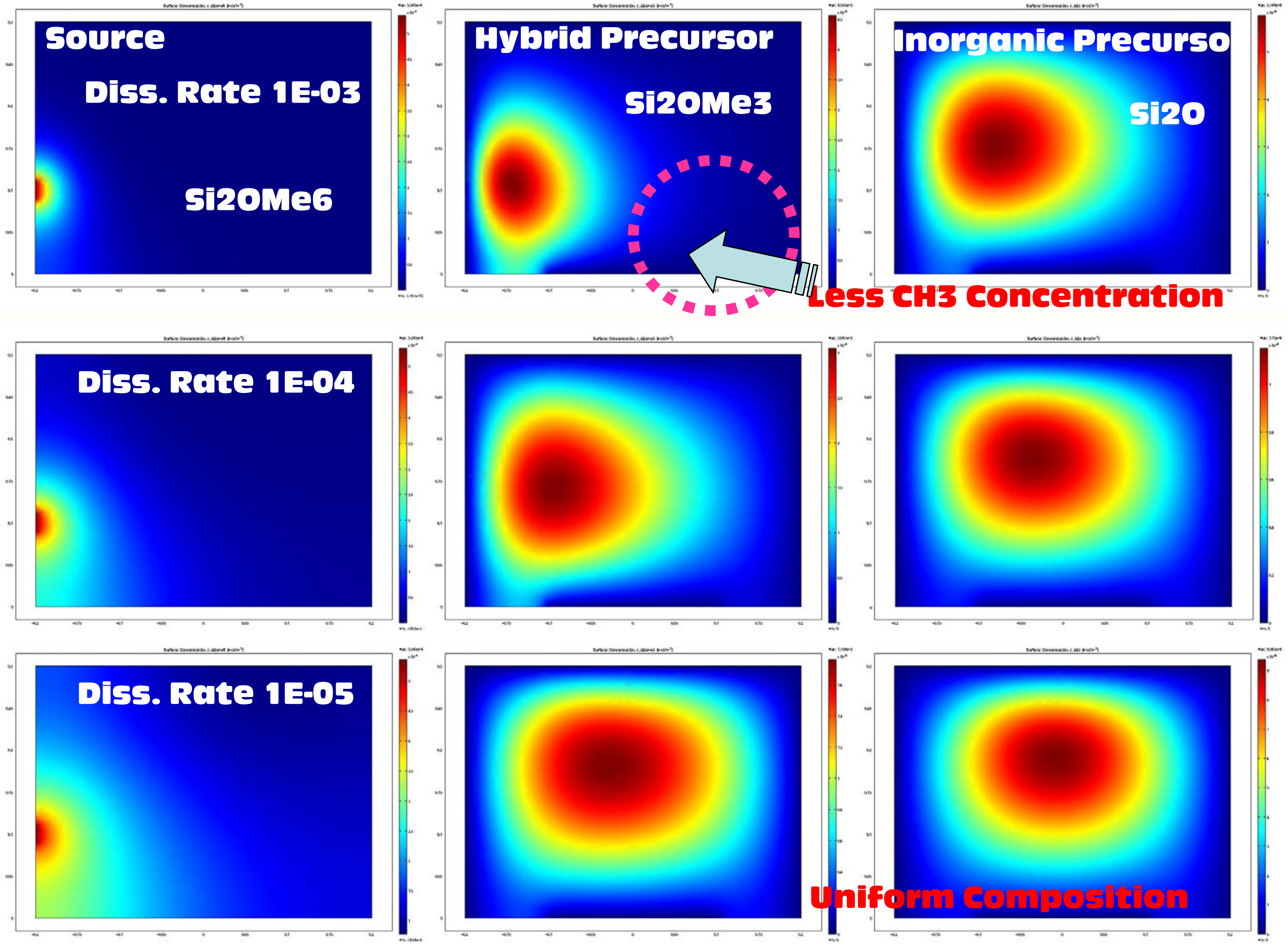
Check Effects of Sticking Probability



Transport

Check Effects of Sticking Probability Results

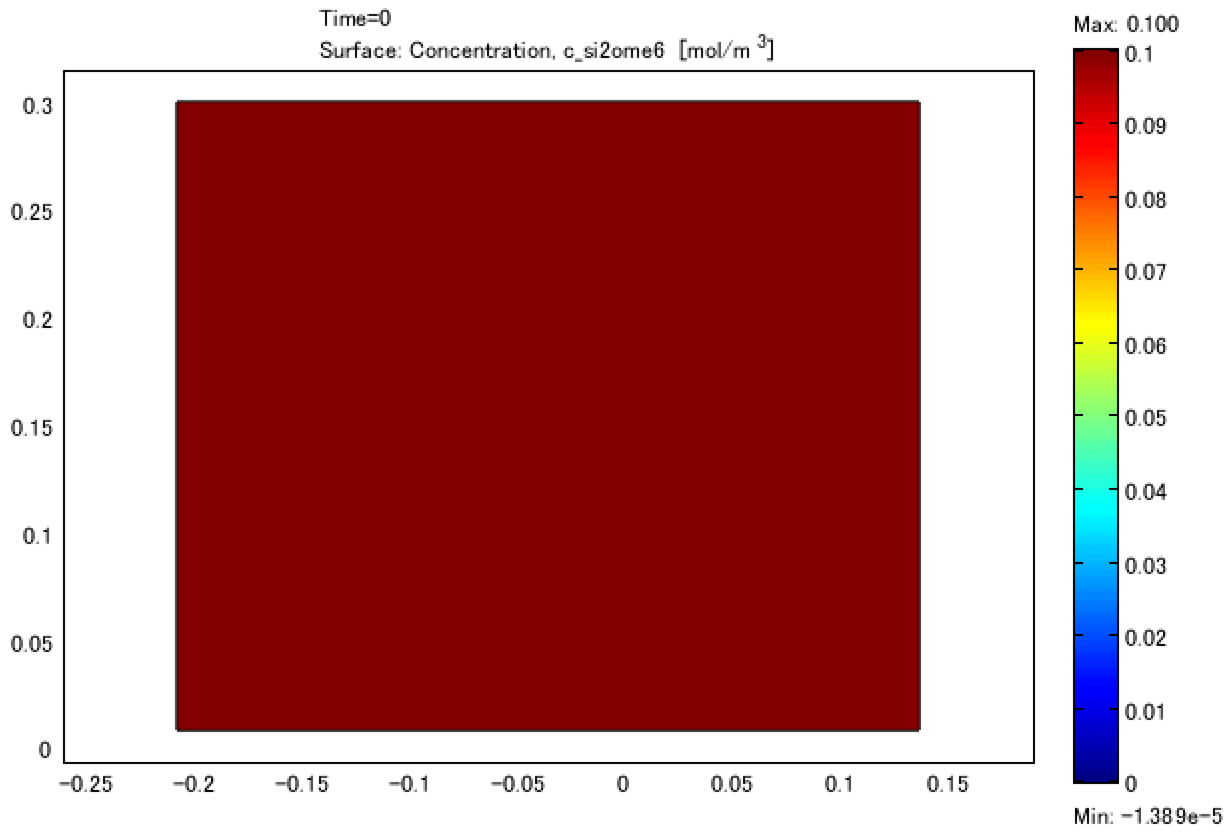



Source
Diss. Rate 1E-03
Si2OMe6
Hybrid Precursor
Si2OMe3
Inorganic Precurso
Si2O
Less CH3 Concentration
Diss. Rate 1E-04
Diss. Rate 1E-05
Uniform Composition

Transport

Spatio-Temporal Bahvior of Source Monomers

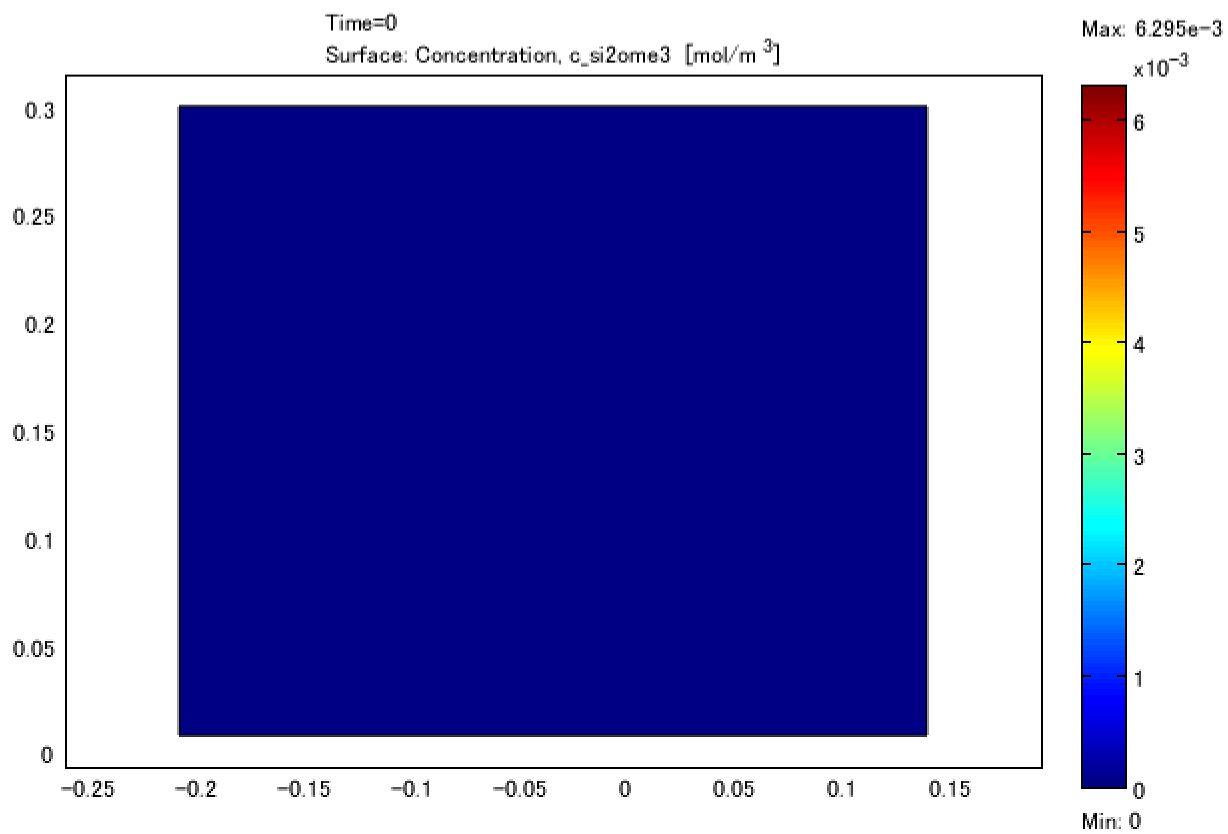
Si2OMe6: Me3-Si-O-Si-Me3



Transport

Spatio-Temporal Bahvior of Hybrid Precursors

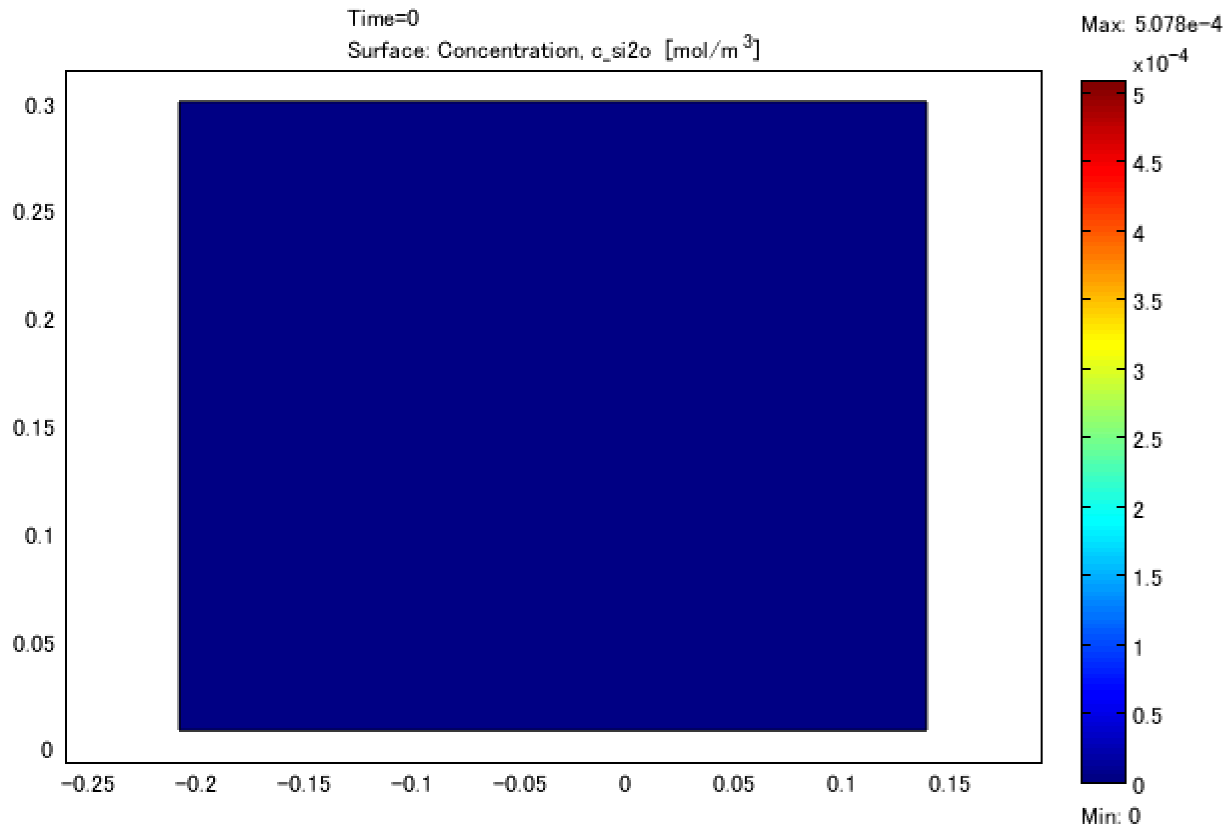
Si2OMe3: Me3-Si-O-Si-



Transport

Spatio-Temporal Bahvior of Inorganic Precursors

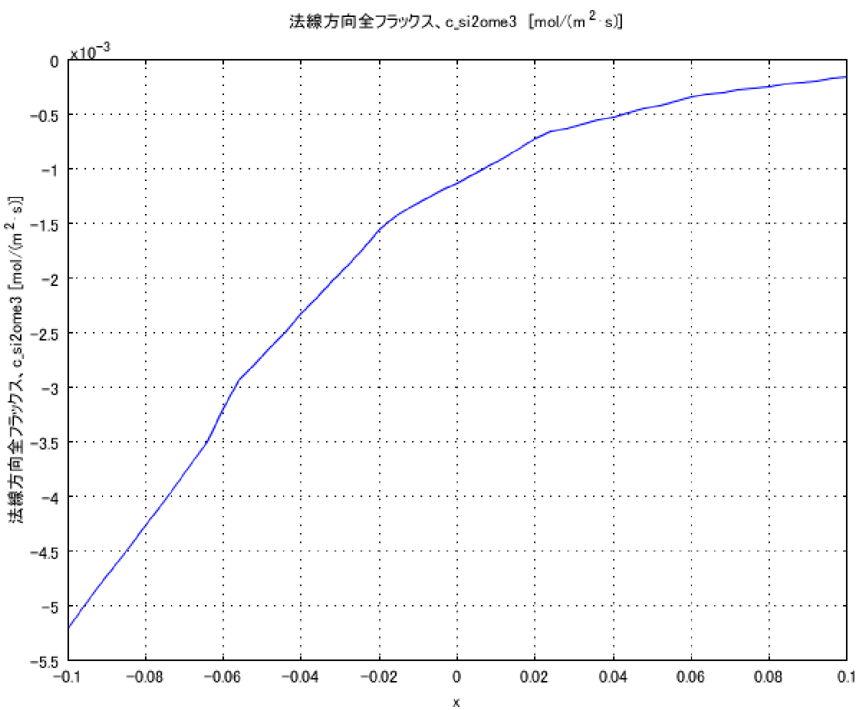
Si2OMe3: -Si-O-Si-



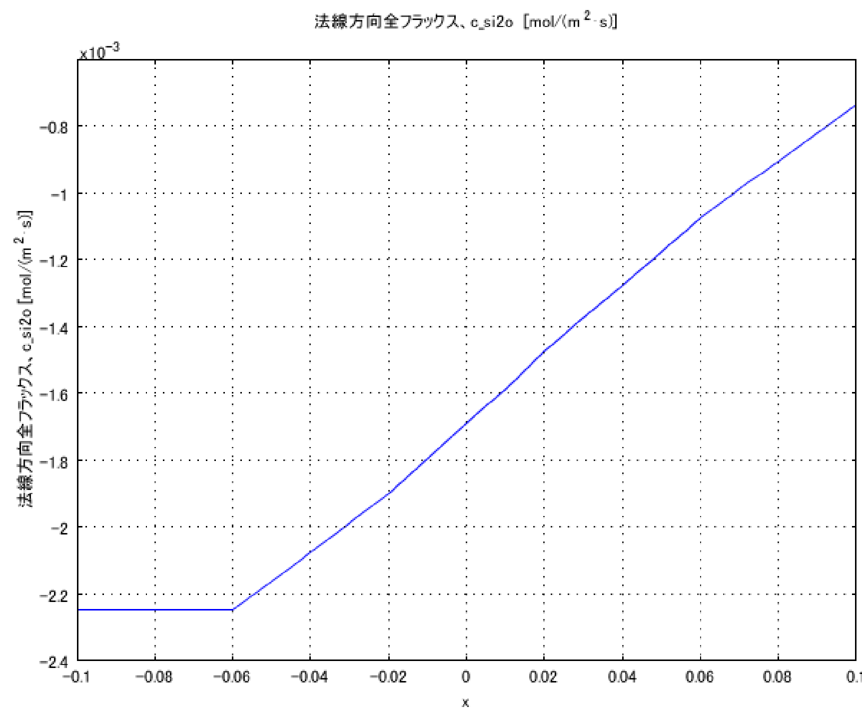
Transport

Flux onto the substrate

Si2OMe3: Me3-Si-O-Si-



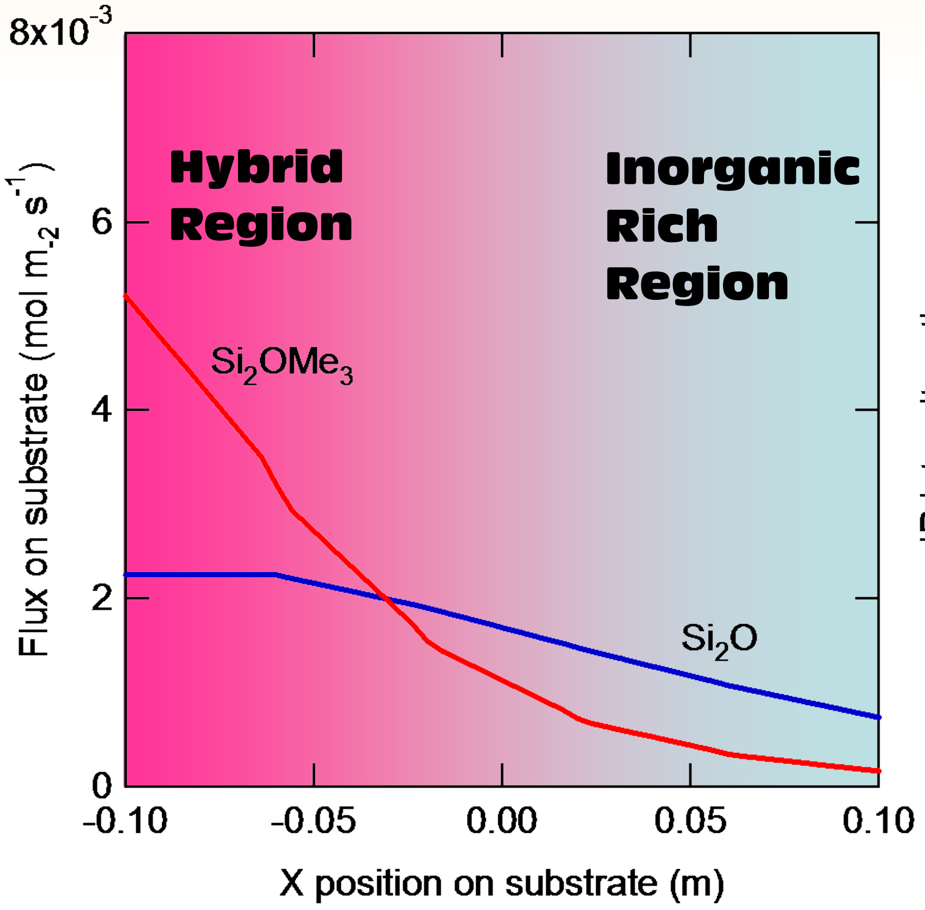
Si2OMe3: -Si-O-Si-

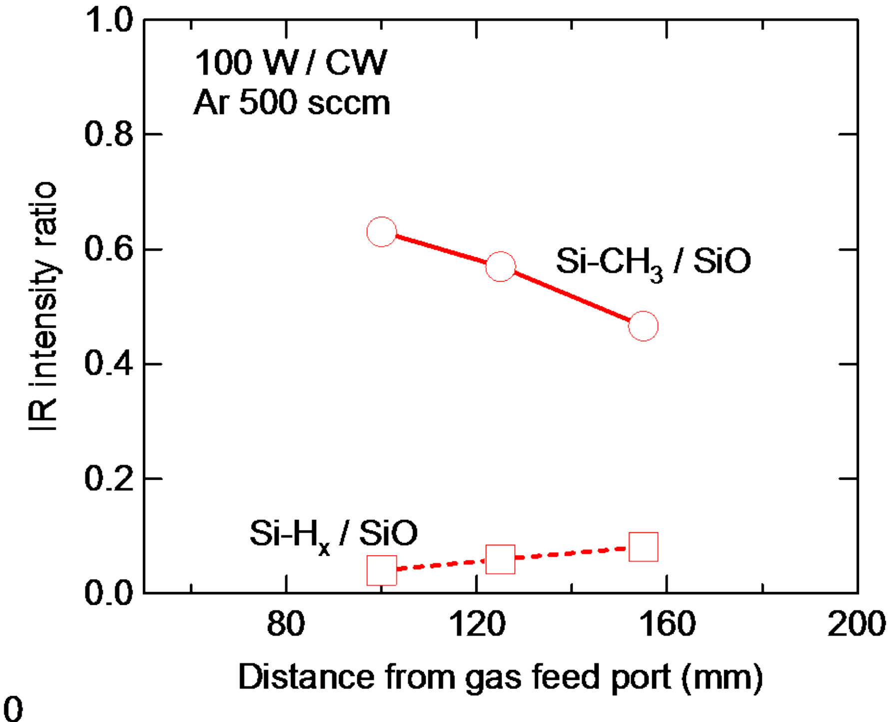


Transport

Flux onto the substrate

Composition variation can be qualitatively explained





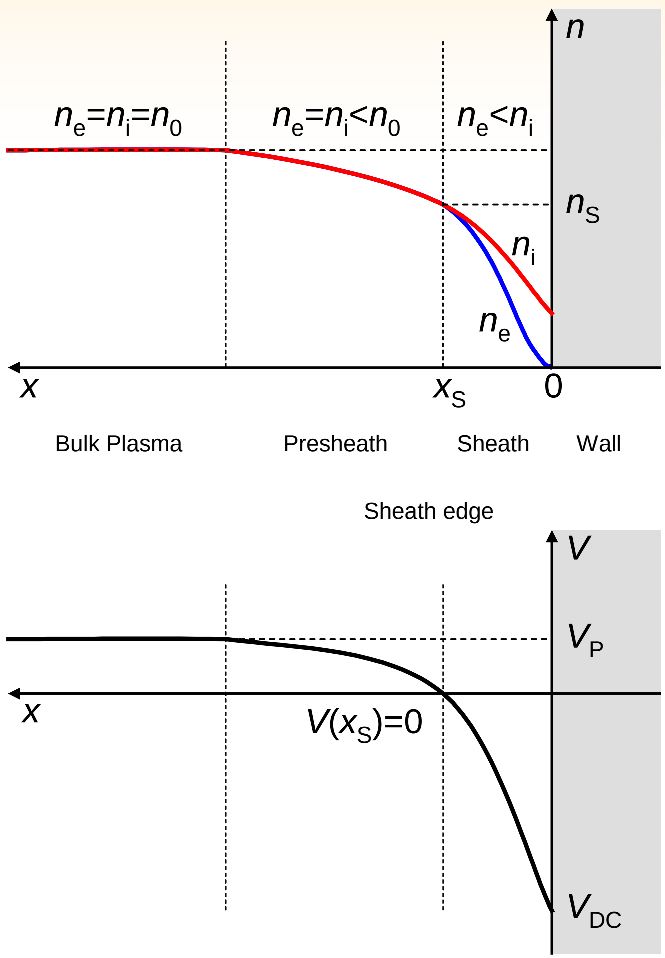
Transport

Combination of Chem. Eng. Module and Reac. Eng. Lab. provides visualization of spatio-temporal behavior of chemical species.

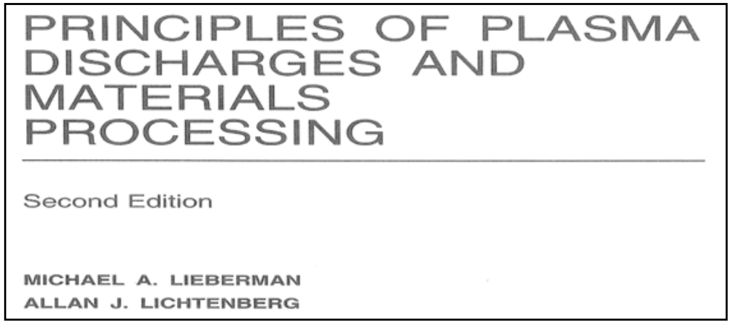
Useful tools for understanding the experimental results, and may be also for designing the reactor structure.

Sheath Analysis

Sheath on the structured surface
→ Trajectory of ions



1D → described on the text book


PRINCIPLES OF PLASMA DISCHARGES AND MATERIALS PROCESSING

Second Edition

MICHAEL A. LIEBERMAN
ALLAN J. LICHTENBERG

2D → depends on actual surface

Calculation for a given structure is required

Sheath Analysis

Physics

Ion energy conservation

$$\left.\frac{1}{2}M_{\mathrm{i}}u^2(x)\right|_{x>x_S} = \left.\frac{1}{2}M_{\mathrm{i}}u^2(x) - q_0 V(x)\right|_{x<x_S}$$

Ion flux continuity

$$\left.n_{\mathrm{is}}u_{\mathrm{s}}\right|_{x=x_S} = \left.n_{\mathrm{i}}(x)u(x)\right|_{x<x_S}$$

Bohm velocity

$$n_{\mathrm{i}}(x) = n_{\mathrm{is}} \quad \text{for} \quad x > x_S \quad V(x) > \frac{1}{2}M_{\mathrm{i}}u_S^2$$

$n_i(x)$ ← Solution of these equation

$$n_{\mathrm{i}}(x) = n_{\mathrm{is}}\left(1 - \frac{2q_0 V(x)}{M_{\mathrm{i}}u_S^2}\right)^{-1/2}$$

$n_e(x)$ ← Boltzman relation

$$n_{\mathrm{e}}(x) = n_{\mathrm{es}}\exp\left(\frac{q_0 V(x)}{kT_{\mathrm{e}}}\right)$$

Sheath Analysis

What we want to know?

- n_e profile … given by

$$n_e(x,y) = n_{es}\exp\left(\frac{q_0 V(x,y)}{kT_e}\right)$$

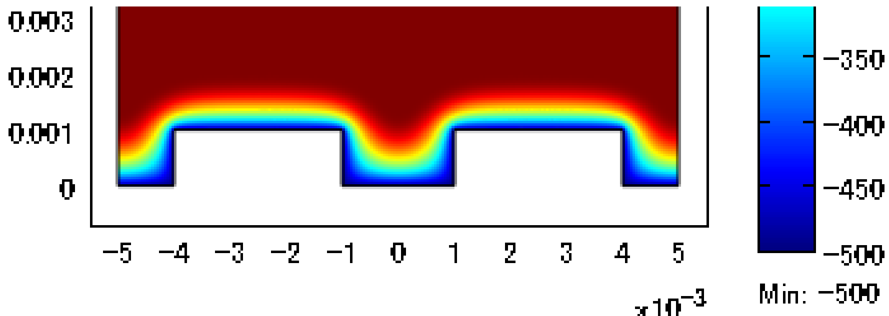
- n_i profile … given by

$$n_i(x,y) = n_{is}\left(1 - \frac{2q_0 V(x,y)}{M_i u_S^2}\right)^{-1/2}$$

Following two parameters are important for understanding ion trajectory in the Sheath (ex. direction of ion bombardment).

(1) Sheath Thickness (or profile) (x_s, y_s)
(2) 2D EF (or V) profile in the Sheath

But analytical solution is not available, because n_e & n_i is given as a function of V(x,y) (not (x,y)!).
→ Numerical Approach



Sheath Analysis

Governing equations

$$\frac{\partial^2 V}{\partial x^2} = -\frac{q_0}{\varepsilon_0}(n_{\mathrm{i}} - n_{\mathrm{e}})$$

$$n_{\mathrm{i}}(x) = n_{\mathrm{is}}\left(1 - \frac{2q_0 V(x)}{M_{\mathrm{i}} u_{\mathrm{S}}^2}\right)^{-1/2}$$

$$n_{\mathrm{e}}(x) = n_{\mathrm{es}} \exp\left(\frac{q_0 V(x)}{kT_{\mathrm{e}}}\right)$$

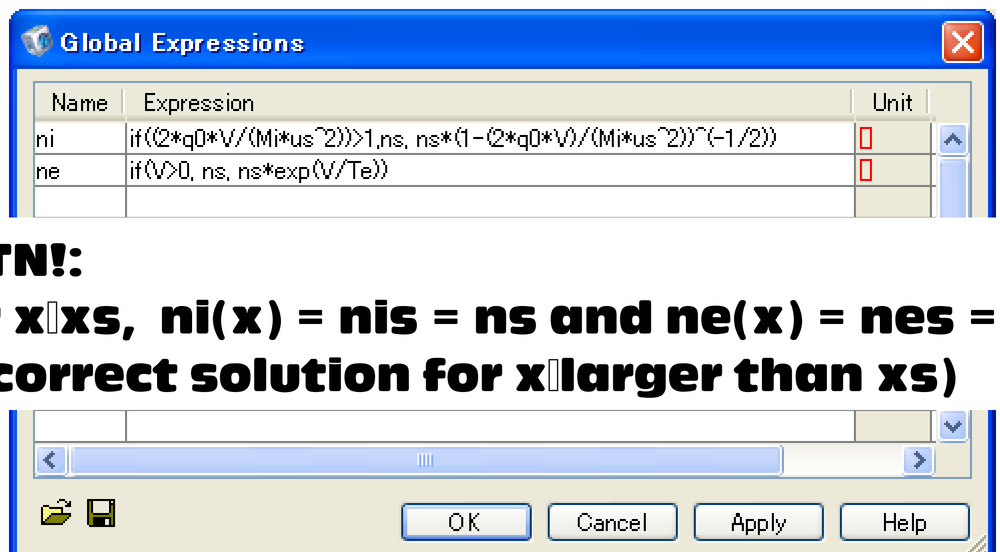
What we should do.

(1) Find self-consistent V(x,y) profile which satisfy these 3 Eqs.

(2) Find (x,y) position where V(x,y)=0

→ ***Sheath Edge profile***

Implementation to COMSOL



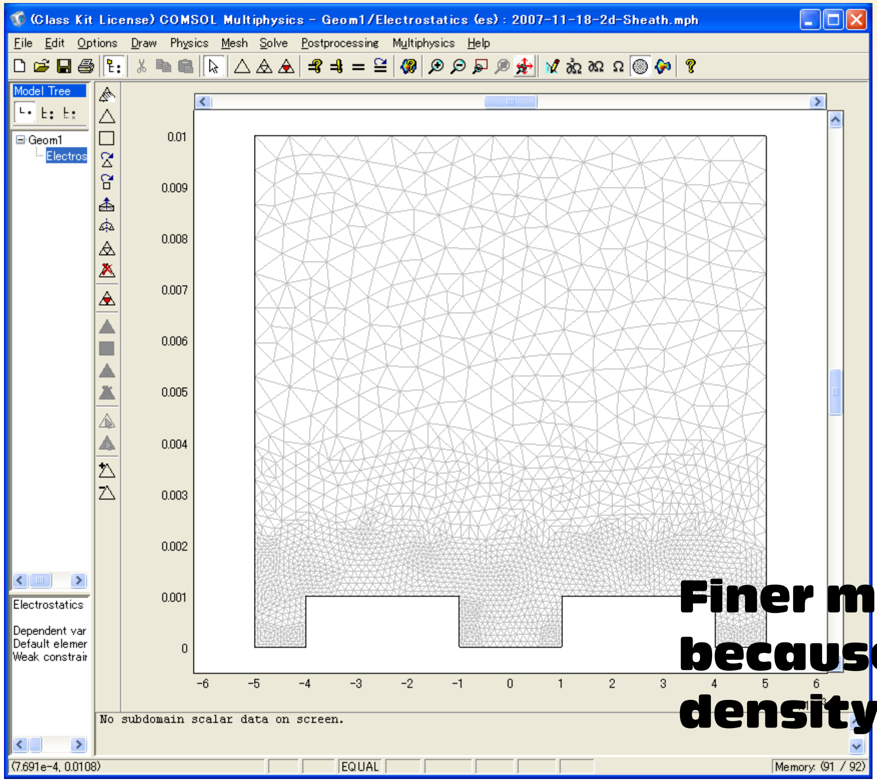
ATTN!:

For x□xs, ni(x) = nis = ns and ne(x) = nes = ns.

(Incorrect solution for x□larger than xs)

Sheath Analysis

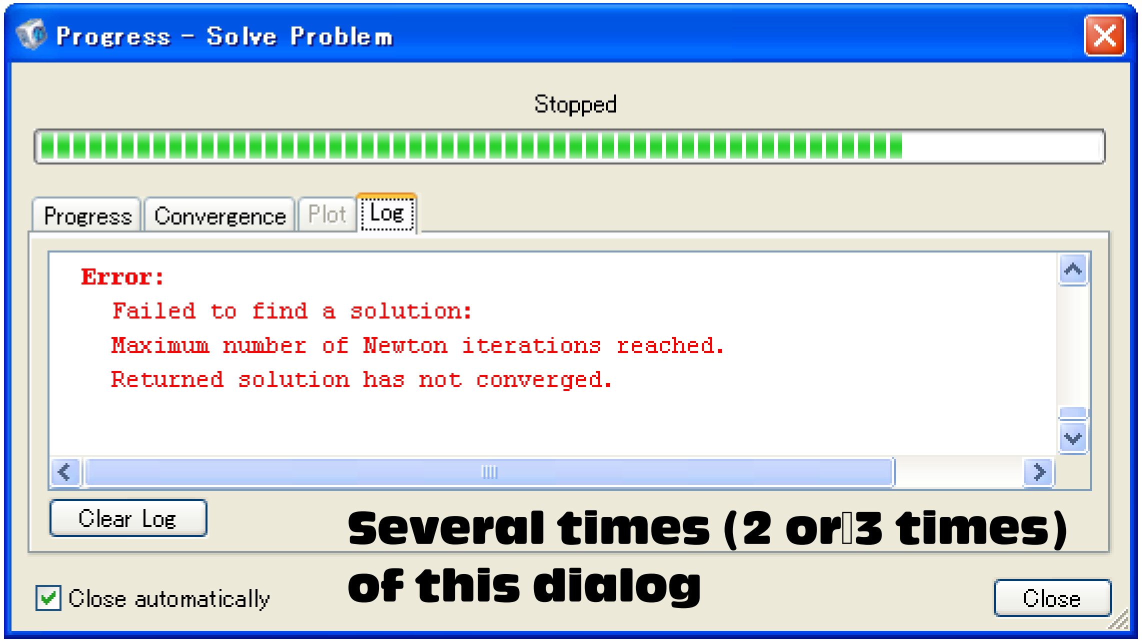
Meshing in COMSOL



Finer mesh near the substrate because of steep potential and density drop

Sheath Analysis

Requires several sets of iteration for obtaining the self-consistent result



Several times (2 or 3 times) of this dialog

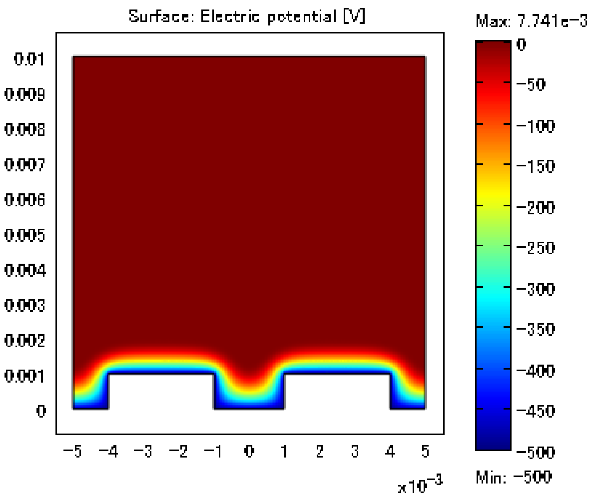
Sheath Analysis

Sheath on the structured surface

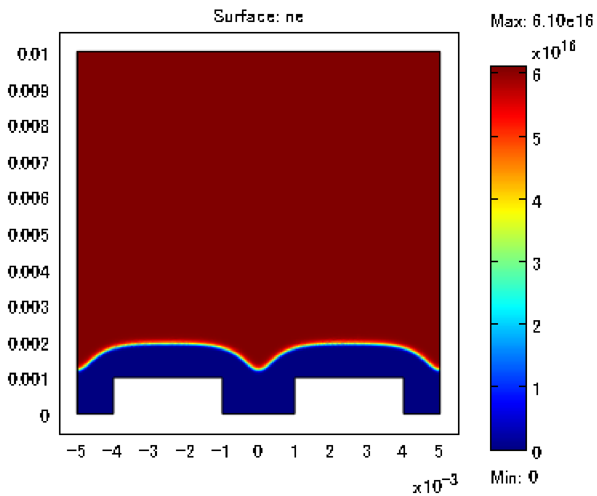
T_e = 2eV, V_{dc} = -500V, n_0 = 10^{17} /m^3

M_i for Fluorine = $19\times10^{-3}/N_A$ kg

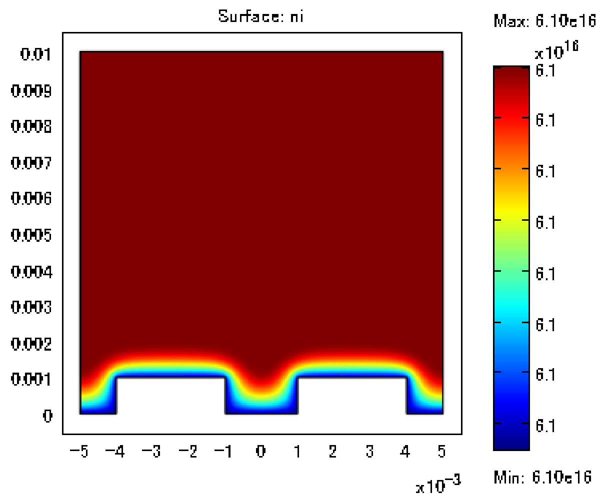
V(x)



ne



ni



$$\frac{\partial^2 V}{\partial x^2} = -\frac{q_0}{\varepsilon_0}(n_i - n_e)$$

$$n_e(x) = n_{es}\exp\left(\frac{q_0 V(x)}{kT_e}\right)$$

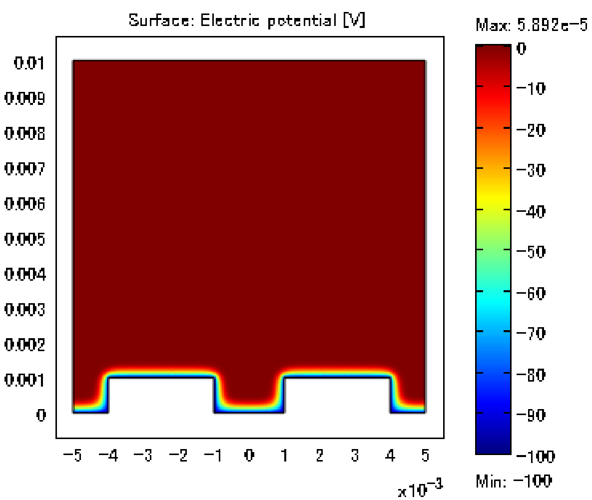
$$n_i(x) = n_{is}\left(1 - \frac{2q_0 V(x)}{M_i u_S^2}\right)^{-1/2}$$

$$n_s = n_0 \exp\left(-\frac{V_p}{T_e}\right), \quad V_p = \frac{T_e}{2} \quad \Rightarrow n_s = 0.61 n_0 \qquad u_s = \left(-\frac{q_0 V_p}{M_i}\right)$$

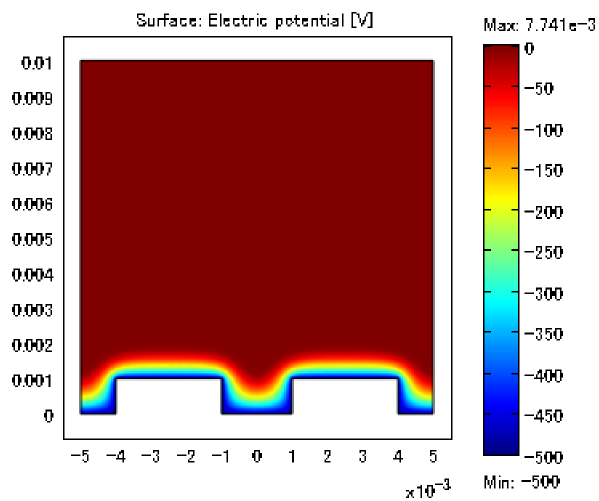
Sheath Analysis

Effect of Vdc

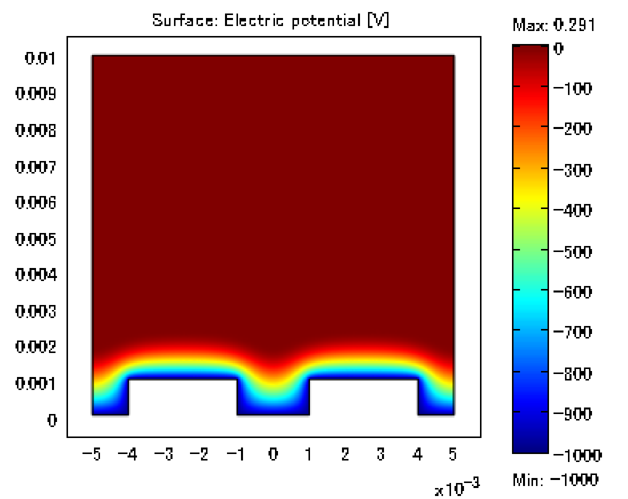
Vdc=-100V



Vdc=-500V



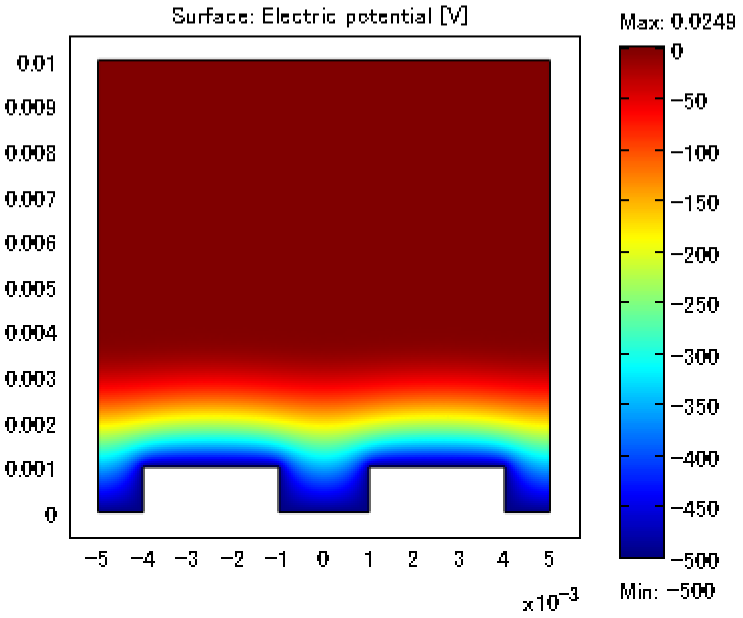
Vdc=-1000V



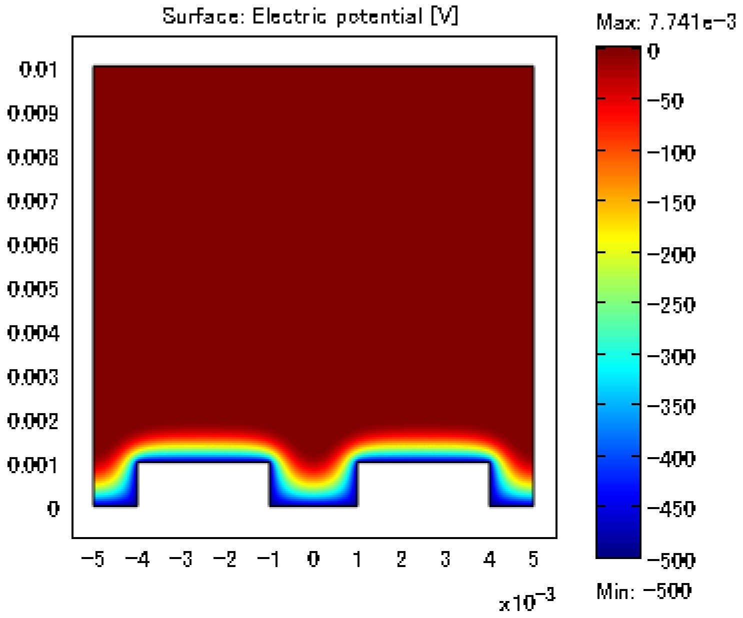
Sheath Analysis

Effect of n_o

$n_o = 10^{16}$ /m^3



$n_o = 10^{17}$ /m^3



Conclusions

1D Simulation

Written in the text book, but animation of the temporal behavior of plasma parameters is better than figures on the text book

2D Simulation

1D analytical model does not tell us "shape" of the plasma parameters.

Actual shape of the objects (e.g. substrate, reactor, ...) is reflected in the results of 2D simulation.

Valid for rough design of reactors or for understanding effects of the shape of reactors (or substrate)

Thank you.