Atmospheric Plasma Modelling Applied For Thermal Plasma Assisted Processes

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French company founded in 2019

Headquarters in La Turbine (95015, Cergy-Pontoise)
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2 Doctors with Ph.D. in Physics
1 Engineer in Electrotechnics

Expertises:
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Plasma Physics
CFD, turbulence and shock wave

On-going developments:
Neural Networks based methods

Fields of application:
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Wind/water fluid-structure interactions
EM/High-voltage environnements
Structural mechanics and Fatigue
Heat transfers

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Numerical modelling and Study
Standalone Application

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Plasma, the fourth state of matter

- Rare on Earth at the natural state: aurora borealis, lightning, flame

- Most abundant form of ordinary matter in the Universe: stars, intracluster medium, intergalactic medium

- Plasma contains electrons, ions and neutrals (atoms and molecules)

- Plasma can be artificially generated over a wide range of operating conditions: Low-pressure plasma, Atmospheric-pressure plasma, DC discharge, Arc discharge, RF/microwave plasma, …

- **3 categories of plasma:**
  - Cold plasmas (low pressure < 1 mbar, ambient temperature ~300 K)
  - Thermal plasmas (atmospheric pressure, medium temperature ~$10^{3\text{-}4}$ K)
  - Fusion plasmas (high pressure ≥ 1 atm, high temperature ~10^6 K)

⇩ The more the particle interactions increase, the more the challenges in simulation are met

Problem statement

- **Microwave plasmas (0.3-300GHz)** are used in industry for various applications such as in microelectronics or decomposition of greenhouse gases
- **High-pressure microwave plasmas** are still studied in laboratories since they are experimentally characterized by specific phenomena of contraction or filamentation [1]

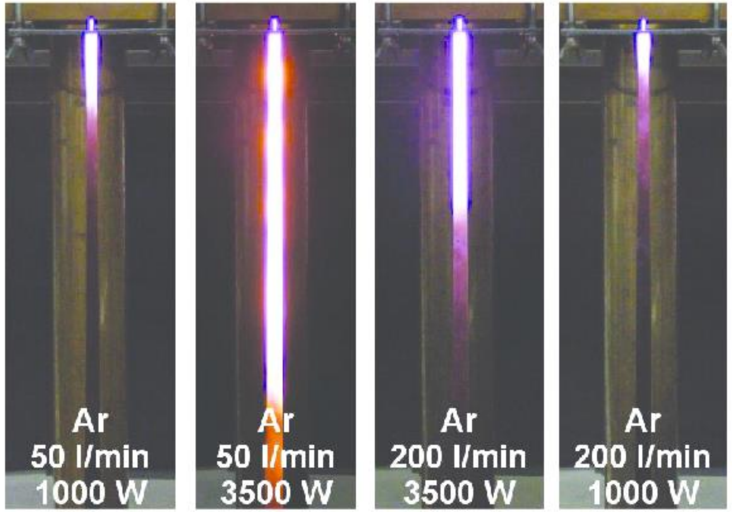
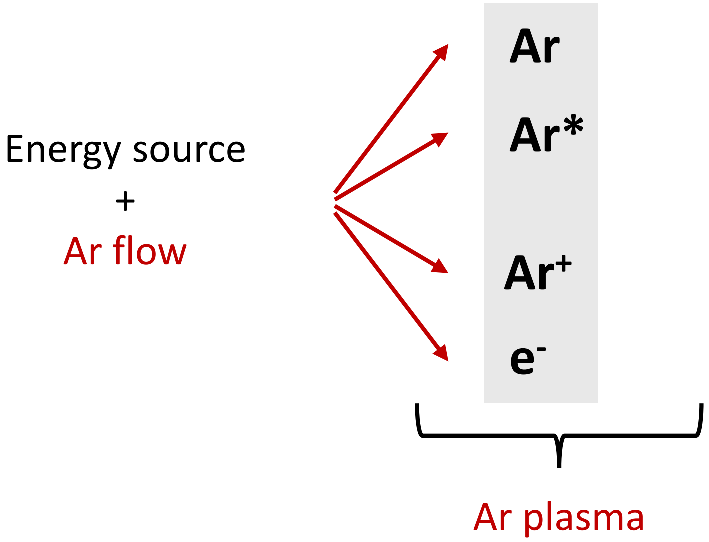


FIGURE 1. Cylindrical microwave plasma source operated with Argon at atmospheric pressure [2].



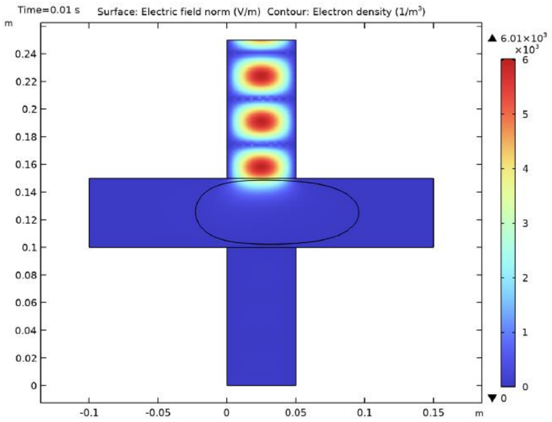
Energy source
+
Ar flow

Ar
Ar*
Ar$^+$
e$^-$

Ar plasma

[1] M. Moisan and J. Pelletier, Physique des Plasmas Collisionnels - Application aux décharges haute fréquence, EDP Sciences, 2006
[2] B. Hrycak, M. Jasinski and J. Mizeraczyk, Tuning characteristics of cylindrical microwave plasma source operated with argon, nitrogen and methane at atmospheric pressure, PRZEGLĄD ELEKTROTECHNICZNY (Electrical Review), ISSN 0033-2097, R. 88 NR 6/2012

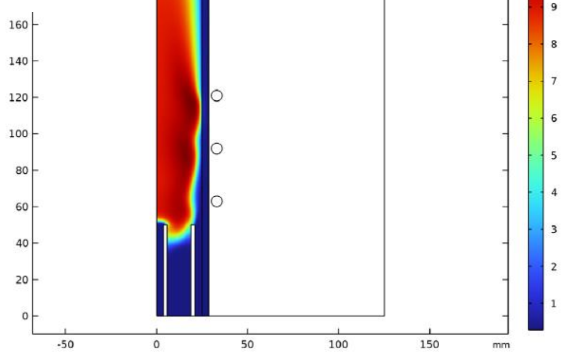
Overview of the existing models



- **In-Plane Microwave Plasma** - Application ID: 8664

  RF module + Plasma module

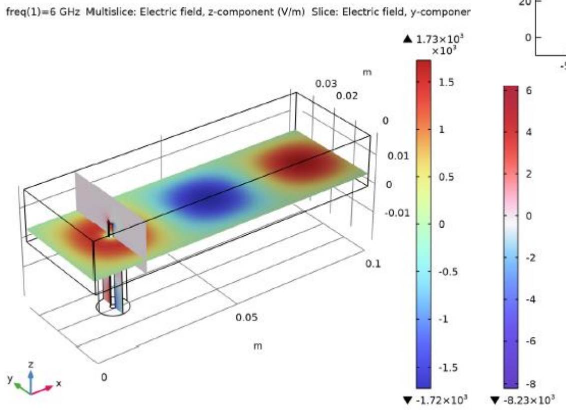
  → valid at low pressure, no increase in temperature



- **Inductively Coupled Plasma (ICP) Torch** - Application ID: 18125

  AC/DC module + Plasma module

  → valid at atmospheric pressure with an increase in temperature, no reaction set is considered, thermodynamic equilibrium vs T is assumed for the gas mixture properties



- **Coaxial to Waveguide Coupling** - Application ID: 1863

  RF module

  → waveguide-to-coaxial coupling is observed, no plasma

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Main goals and expected results

- In that context, **a fully-coupled microwave plasma model at atmospheric pressure** with COMSOL Multiphysics® is a necessary step to optimize the development of such a plasma reactor

- Four blocks of Physics must be considered to study this problem:
    - **Electromagnetics** for the microwave propagation and plasma interaction – RF module
    - **Fluid dynamics** for the gas mixture flow – CFD module
    - **Heat transfers** for the thermodynamic equilibrium – Heat transfer module
    - **Plasma physics** for the electron and heavy particule production – Plasma module

- The expected results are:
    - **Production of the plasma**
    - **Absorption of the microwave (skin effect)**
    - **Waveguide-to-coaxial coupling**
    - **Increase in the gas temperature**

Simulation of plasmas: Numerical assumptions [3]

- **Fluid approach:**
  - Continuum
  - Transport equations
  - Assumed Maxwellian EEDF

- **Limitations:**
  - Reduced electric field: $\frac{|\mathbf{E}|}{N} < 500\ \mathrm{Td}$
  - Electron density: $n_e \ll N$ (low degree of ionization)
  - Debye length: $\lambda_D \ll L$ (apparent charge neutrality)
  - Gas pressure: $p > 10^{-3}\ \mathrm{mbar}$

$$1\ \mathrm{atm@300K} \Rightarrow N \approx 10^{25}\ \mathrm{m}^{-3}$$

$$1\ \mathrm{Td} = 10^{-21}\ \mathrm{V.m}^2 \Rightarrow |\mathbf{E}|_{\mathrm{max}} \approx 5\ \mathrm{MV/m}$$

These assumptions are satisfied in the present case study.

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[3] J. Crompton and L. Gritter, Plasma modeling in COMSOL Multiphysics®, AltaSim Technologies - https://www.comsol.fr/video/modeling-plasmas-in-comsol-multiphysics

Simulation of plasmas

- **Electron density transport [4]:**

$$\frac{\partial n_e}{\partial t} + \boldsymbol{\nabla}.\boldsymbol{\Gamma}_e = \underbrace{R_e}_{\text{Production rate}} \qquad [1/(\mathrm{m}^3.\mathrm{s})]$$

$$\boldsymbol{\Gamma}_e = -\underbrace{(\mu_e \mathbf{E})n_e}_{\text{Convective flux}} - \underbrace{\nabla(D_e n_e)}_{\text{Diffusive flux}} \qquad [1/(\mathrm{m}^2.\mathrm{s})]$$

Convection of electrons due to fluid motion (**u**) is neglected

E is the electric field driven by the **Maxwell's equations**

$$R_{eX} = \pm n_e n_X k_{eX} \qquad k_{eX} = \int_0^{+\infty} \sigma_{eX}(u_e) 4\pi {u_e}^2 f(u_e) u_e du_e$$

$$f(u_e) = n_e \left(\frac{m_e}{2\pi k_B T_e}\right)^{\frac{3}{2}} \exp\left(-\frac{m_e|\mathbf{u}_e|^2}{2k_B T_e}\right)$$

Maxwellian EEDF [1]

#	Formula	Type
1	e+Ar → e+Ar	Elas.
2	e+Ar → e+Ar(4s)	Exc.
3	e+Ar → e+Ar(4p)	Exc.
4	e+Ar → 2e+Ar$^+$	Ion.
5	e+Ar(4s) → e+Ar(4s)	Elas.
6	e+Ar(4s) → e+Ar(4p)	Exc.
7	e+Ar(4s) → e+Ar	Exc.
8	e+Ar(4s) → 2e+Ar$^+$	Ion.
9	e+Ar(4p) → e+Ar(4p)	Elas.
10	e+Ar(4p) → e+Ar(4s)	Exc.
11	e+Ar(4p) → e+Ar	Exc.
12	e+Ar(4p) → 2e+Ar+	Ion.
13	2e+Ar$^+$ → e+Ar	Att.

Net electron production [5]

This is how electron density balance is computed.

[4] COMSOL Help Resources: Plasma Module > User's Guide > Plasma Interfaces > Plasma Reactors Theory

[5] W. Zhang, Recherche numérique et expérimentale sur les propriétés de décharge et les caractéristiques de propagation électromagnétique dans les torches à plasma micro-ondes, Toulouse INP, 2019

Simulation of plasmas

- **Electron energy density transport [6]:**

$$\frac{\partial n_\varepsilon}{\partial t} + \boldsymbol{\nabla}.\boldsymbol{\Gamma}_\varepsilon + \mathbf{E}.\boldsymbol{\Gamma}_\varepsilon = \underbrace{S_{en}}_{\text{Energy loss/gain (inelastic collisions)}} + Q \qquad [\mathrm{W/m^3}]$$

$$\boldsymbol{\Gamma}_\varepsilon = -(\mu_\varepsilon \mathbf{E}) n_\varepsilon - \nabla(D_\varepsilon n_\varepsilon) \qquad [\mathrm{W/m^2}]$$

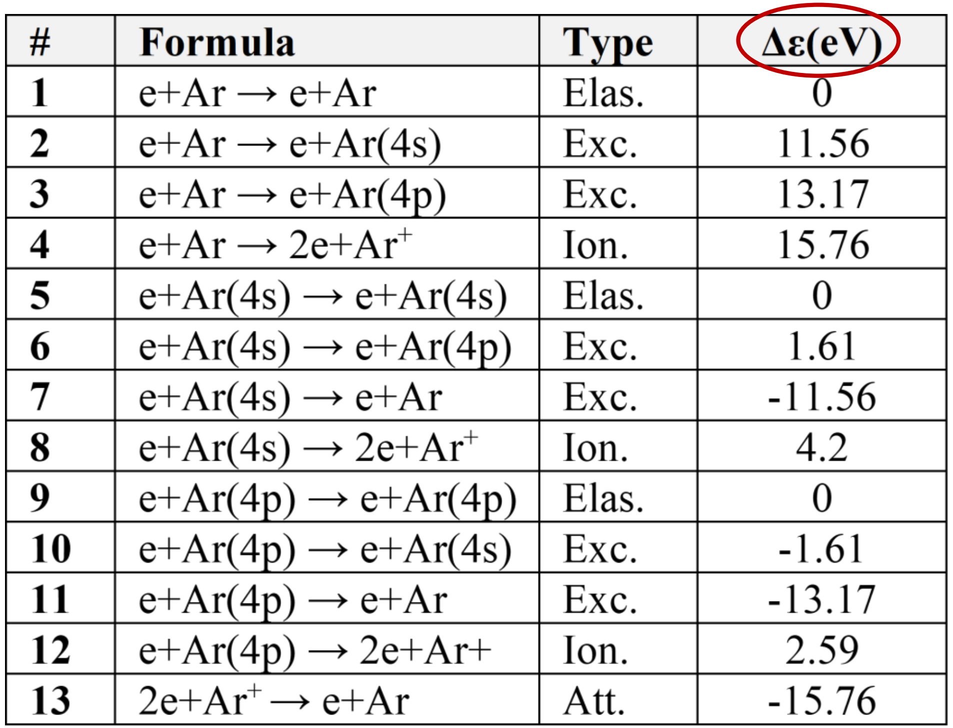
Convection of electrons due to fluid motion (**u**) is neglected

E is the electric field driven by the **Maxwell's equations**

Q is an external heat source driven by the **Electron heat source**

$$Q = \frac{1}{2}\Re(\mathbf{j}.\mathbf{E}^*) = \frac{n_e e^2}{m_e}\frac{\nu_m}{\nu_m{}^2 + \omega^2}\frac{E_0{}^2}{2}$$

Heat source for the electrons
(absorbed power density [1])



#	Formula	Type	Δε(eV)
1	e+Ar → e+Ar	Elas.	0
2	e+Ar → e+Ar(4s)	Exc.	11.56
3	e+Ar → e+Ar(4p)	Exc.	13.17
4	e+Ar → 2e+Ar^+	Ion.	15.76
5	e+Ar(4s) → e+Ar(4s)	Elas.	0
6	e+Ar(4s) → e+Ar(4p)	Exc.	1.61
7	e+Ar(4s) → e+Ar	Exc.	-11.56
8	e+Ar(4s) → 2e+Ar^+	Ion.	4.2
9	e+Ar(4p) → e+Ar(4p)	Elas.	0
10	e+Ar(4p) → e+Ar(4s)	Exc.	-1.61
11	e+Ar(4p) → e+Ar	Exc.	-13.17
12	e+Ar(4p) → 2e+Ar+	Ion.	2.59
13	2e+Ar^+ → e+Ar	Att.	-15.76

Electron energy transfers [5]

This is how microwave power is transferred to the electrons.

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[6] COMSOL Help Resources: Plasma Module > User's Guide > The Drift Diffusion Interface > Theory for the Drift Diffusion Interface > Electron Transport Theory

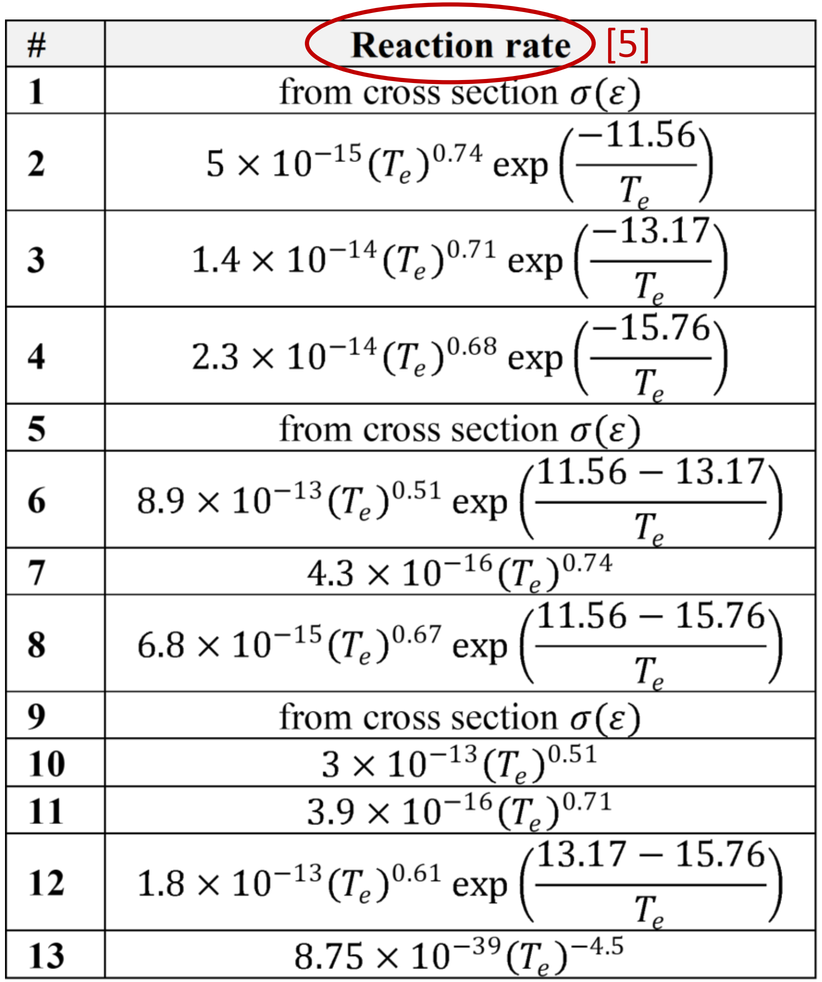
Simulation of plasmas

- **Heavy species mass fraction transport [7]:**

$$\rho\frac{\partial w_k}{\partial t}+\rho(\mathbf{u}.\nabla)w_k=\nabla.\underbrace{\mathbf{j}_k}_{\text{Diffusive flux vector}}+\underbrace{R_k}_{\text{Production rate}}\quad[\text{kg}/(\text{m}^3.\text{s})]$$

$$\mathbf{j}_k=\rho\,w_k\mathbf{V}_k\qquad[\text{kg}/(\text{m}^2.\text{s})]$$

$$\underbrace{\mathbf{V}_k}_{\text{Diffusion velocity for species k}}=D_{k,m}\frac{\nabla w_k}{w_k}+D_{k,m}\frac{\nabla M_n}{M_n}+D_k^T\frac{\nabla T}{T}-z_k\mu_{k,m}\mathbf{E}$$



#	Reaction rate [5]
1	from cross section $\sigma(\varepsilon)$
2	$5\times10^{-15}(T_e)^{0.74}\exp\left(\frac{-11.56}{T_e}\right)$
3	$1.4\times10^{-14}(T_e)^{0.71}\exp\left(\frac{-13.17}{T_e}\right)$
4	$2.3\times10^{-14}(T_e)^{0.68}\exp\left(\frac{-15.76}{T_e}\right)$
5	from cross section $\sigma(\varepsilon)$
6	$8.9\times10^{-13}(T_e)^{0.51}\exp\left(\frac{11.56-13.17}{T_e}\right)$
7	$4.3\times10^{-16}(T_e)^{0.74}$
8	$6.8\times10^{-15}(T_e)^{0.67}\exp\left(\frac{11.56-15.76}{T_e}\right)$
9	from cross section $\sigma(\varepsilon)$
10	$3\times10^{-13}(T_e)^{0.51}$
11	$3.9\times10^{-16}(T_e)^{0.71}$
12	$1.8\times10^{-13}(T_e)^{0.61}\exp\left(\frac{13.17-15.76}{T_e}\right)$
13	$8.75\times10^{-39}(T_e)^{-4.5}$

This is how heavy species mass fraction balance is computed.

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[7] COMSOL Help Resources: Plasma Module > User's Guide > The Heavy Species Transport Interface > Theory for the Heavy Species Transport Interface > Multicomponent Diffusion Equations

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Simulation of plasmas

- **Thermodynamic properties: Total heat source for heavy species [8]**

$$Q=\sum_k Q_k+Q_{e,k}=\sum_k -H_k r_k+\underbrace{\left(2\frac{m_e}{m_k}\right)\frac{3}{2}\left(T_e-\frac{k_BT}{e}\right)Fr_k}_{\text{Electron impact reactions}}\qquad [\mathrm{W/m^3}]$$

$$h_k=R_g\left(a_1T+\frac{a_2}{2}T^2+\frac{a_3}{3}T^3+\frac{a_4}{4}T^4+\frac{a_5}{5}T^5+a_6\right)+F\Delta h$$

Enthalpy of reaction in J/mol from the NASA polynomials [9]

This is how reactions in a plasma heat the background gas.

- **Poisson's equation [4]**

$$\boldsymbol{\nabla}.(\varepsilon_0\varepsilon_r\mathbf{E})=\rho_q$$

$$\rho_q=q\left(\sum_k Z_kn_k-n_e\right)$$

Space charge density

$$\varepsilon_r(\omega)=1-\frac{\omega_p^2}{\omega^2+\nu_m^2}-i\frac{\nu_m}{\omega}\left(\frac{\omega_p^2}{\omega^2+\nu_m^2}\right)$$

$\mathrm{Im}(\varepsilon_r)$ → Absorption is expected in the plasma

This is how plasmas react with an external electric field.

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[8] Plasma Module > User's Guide > The Heavy Species Transport Interface > Theory for the Heavy Species Transport Interface > Thermodynamic Properties

[9] E. Goos, A. Burcat and B. Ruscic, "EXTENDED THIRD MILLENIUM IDEAL GAS AND CONDENSED PHASE THERMOCHEMICAL DATABASE," [Online]. Available: http://garfield.chem.elte.hu/Burcat/THERM.DAT

Simulation of electromagnetic wave propagation

- **Wave equation:**

$$\left(\nabla^2 + \mu_0\mu_r\sigma\frac{\partial}{\partial t} + \frac{\varepsilon_r\mu_r}{c^2}\frac{\partial^2}{\partial t^2}\right)\boldsymbol{E}(\boldsymbol{r},t) = 0$$

$$\boldsymbol{E}(\boldsymbol{r},t) = \boldsymbol{E}(x,y)e^{-i\omega t}e^{ikz}$$

Magnetic field is not considered here since its interaction with the electrons can be neglected (non-magnetized plasma)

  - **In a rectangular waveguide:**

$$E(\boldsymbol{r},t) = \begin{cases} E_x = 0 \\ E_y = i\omega B_0\left(\frac{a}{\pi}\right)\sin\left(\frac{\pi x}{a}\right)e^{-i\omega t}e^{ikz} \\ E_z = 0 \end{cases}$$

$$k^2 = {k_{10}}^2 = \mu\varepsilon\omega^2 - \frac{\pi^2}{a^2}$$

TE10 mode is expected in the rectangular waveguide

This is how EM field propagation is computed.

Simulation of heat transfers

- **Heat equation:**

$$\rho C_p \left(\frac{\partial}{\partial t} + \mathbf{u}.\boldsymbol{\nabla}\right) T + \boldsymbol{\nabla}. \underbrace{\mathbf{q}}_{\substack{\text{heat flux}\\ \text{by convection}}} = \underbrace{Q}_{\substack{\text{total heat source}\\ \text{(electrons and heavy species)}}} \quad [\mathrm{W/m^3}]$$

$$\mathbf{q} = -k\boldsymbol{\nabla}T$$

k is the thermal conductivity in W/m/K
C_p is the heat capacity in J/K/kg

$$C_{p,k} = R_g \left(\enclose{circle}{a_1} + \enclose{circle}{a_2} T + \enclose{circle}{a_3} T^2 + \enclose{circle}{a_4} T^3 + \enclose{circle}{a_5} T^4\right)$$

Heat capacity in J/mol/K from the NASA polynomials [9]

This is how thermal equilibrium is computed.

Simulation of fluid dynamics

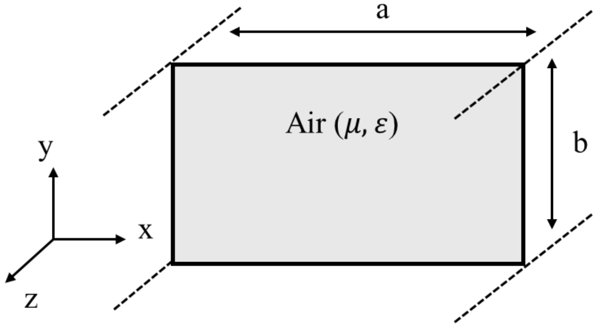
- **Navier-Stokes equation:**

$$\rho\frac{\partial \mathbf{u}}{\partial t}+\rho(\mathbf{u}.\boldsymbol{\nabla})\mathbf{u}=\boldsymbol{\nabla}.\Big[-\underbrace{p}_{\text{pressure}}\mathbf{I}+\underbrace{\mathbf{K}}_{\text{viscous stress tensor}}\Big]+\mathbf{F} \qquad [\mathrm{N/m^3}]$$

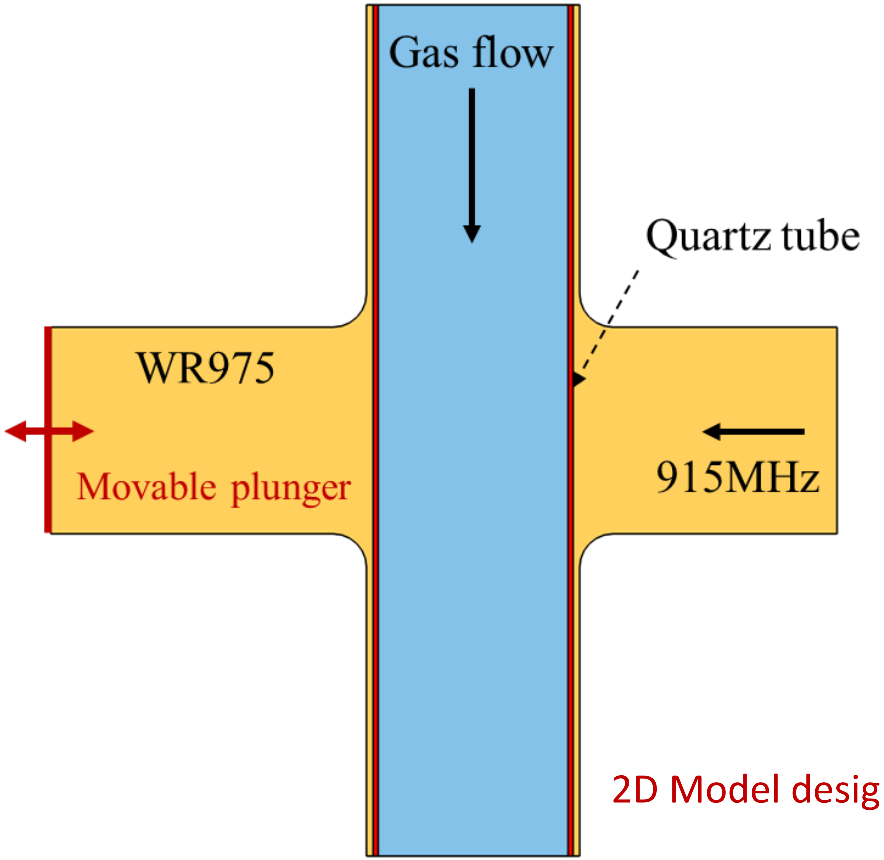
$$\frac{\partial \rho}{\partial t}+\boldsymbol{\nabla}.(\rho\mathbf{u})=0$$

This is how fluid flow is computed.

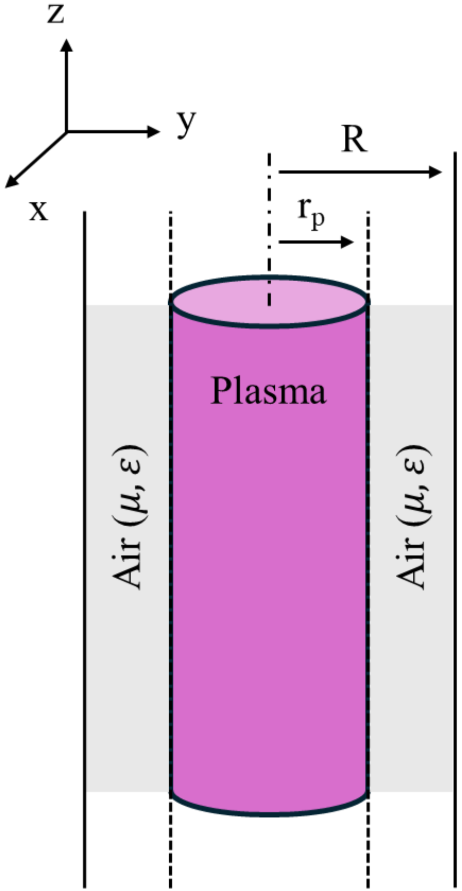
Experimental set-up (2D model)



WR975 rectangular waveguide

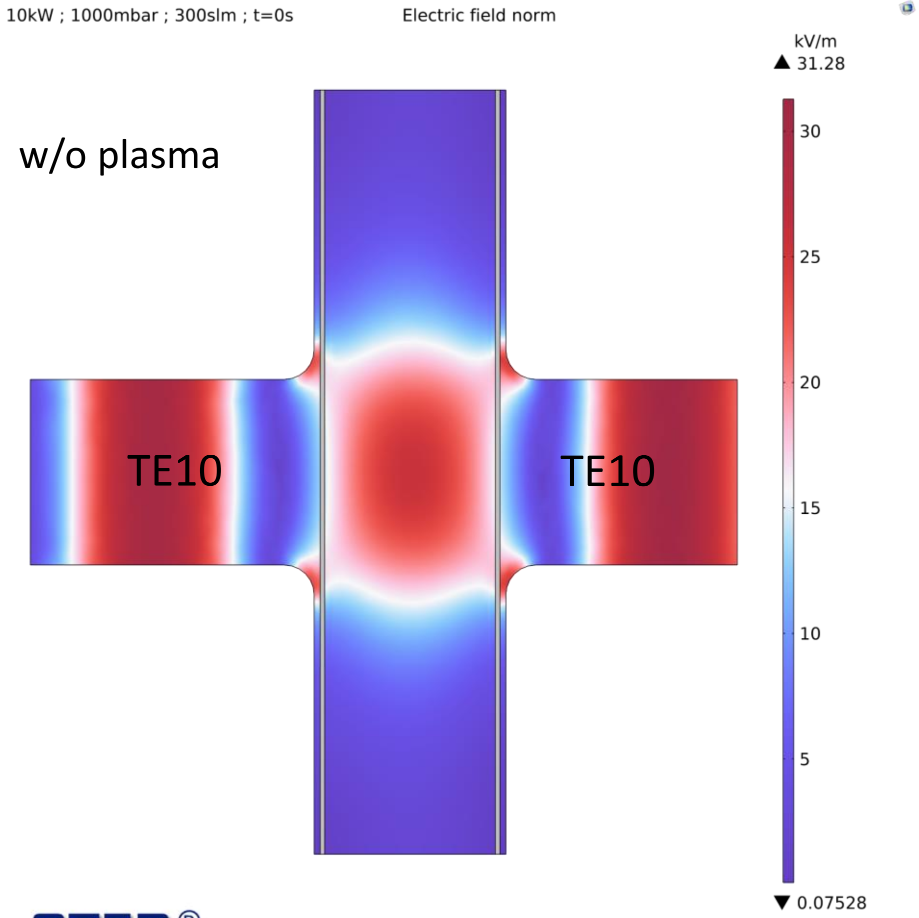


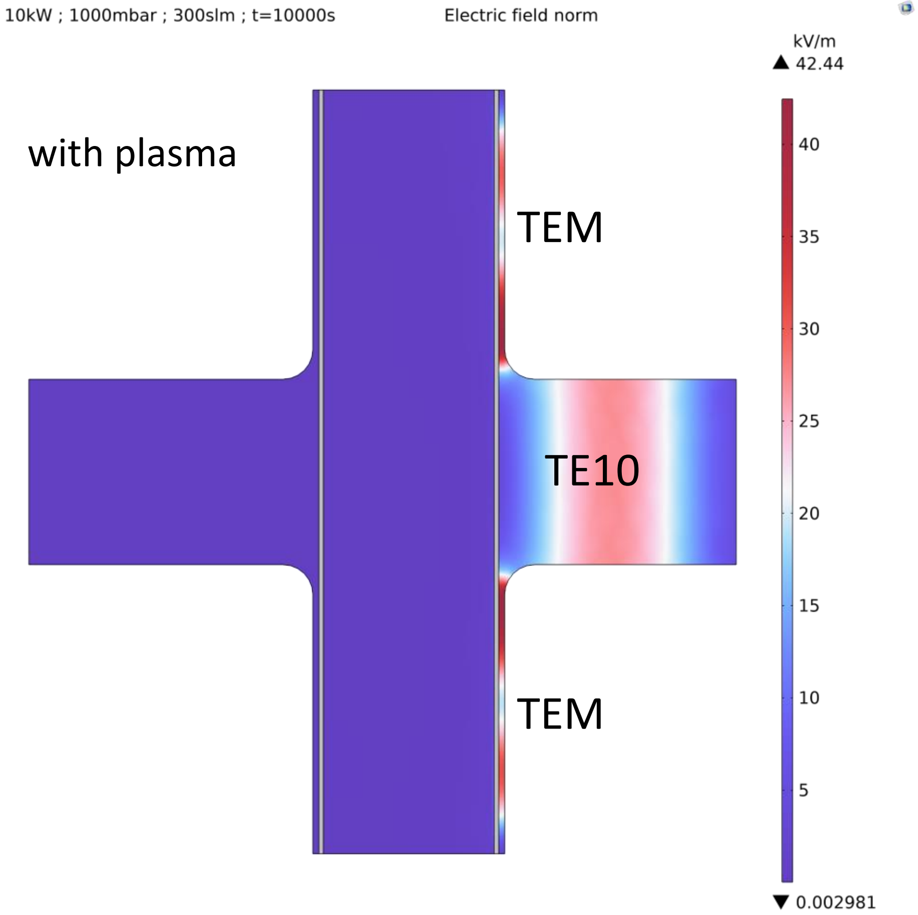
2D Model design



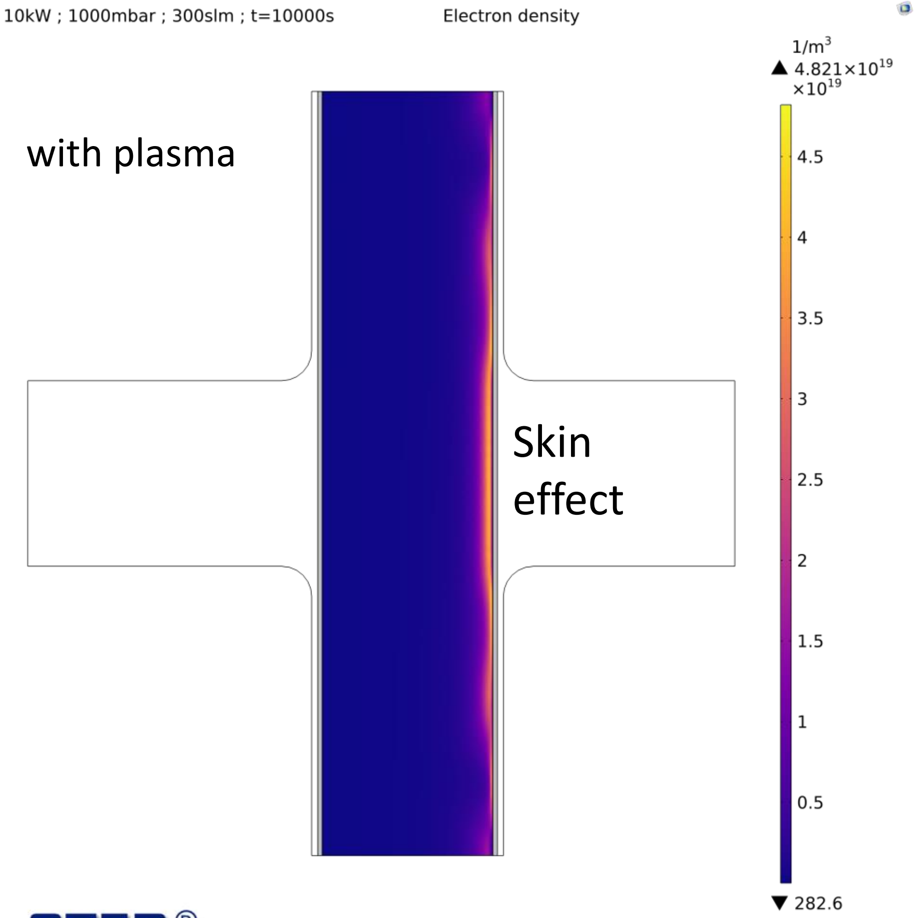
Cylindrical plasma

Results: Waveguide-to-coaxial coupling



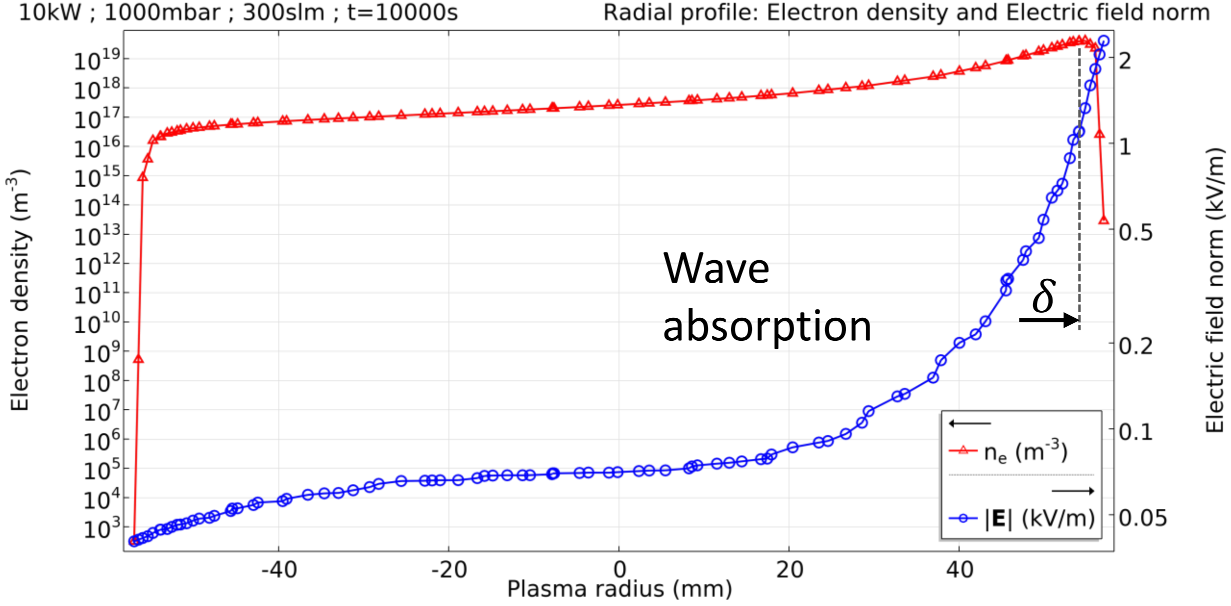


Results: Wave absorption and skin effect



with plasma

Skin effect



Wave absorption

δ

δ: skin depth

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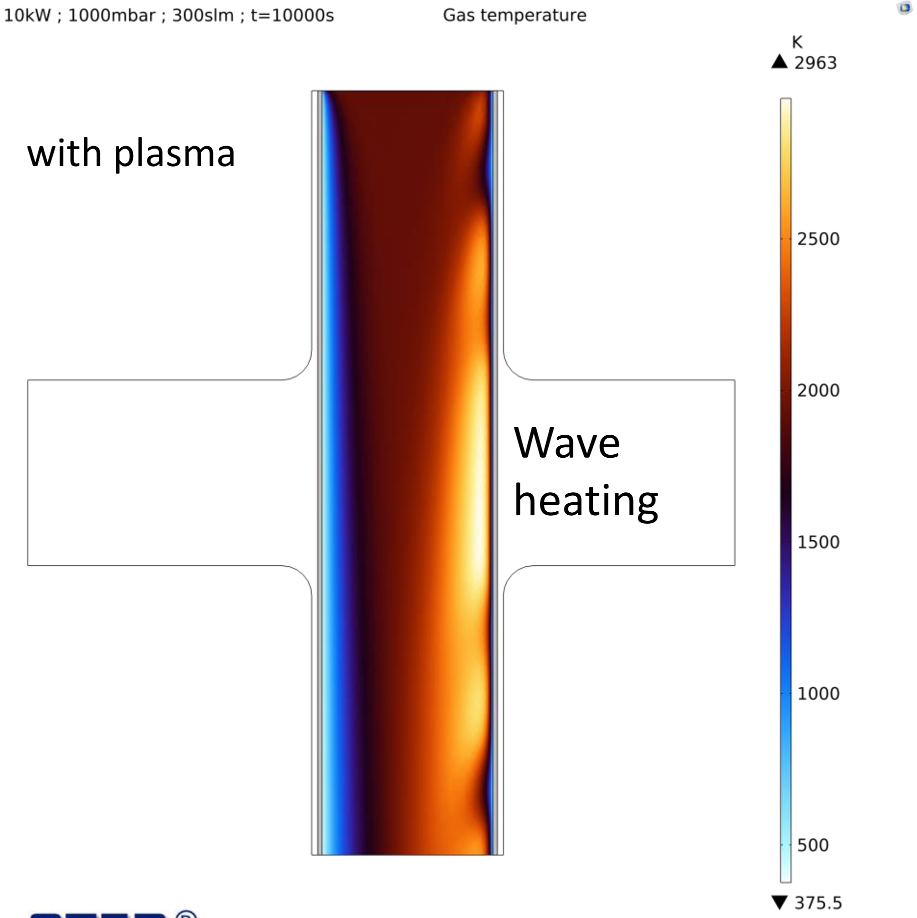
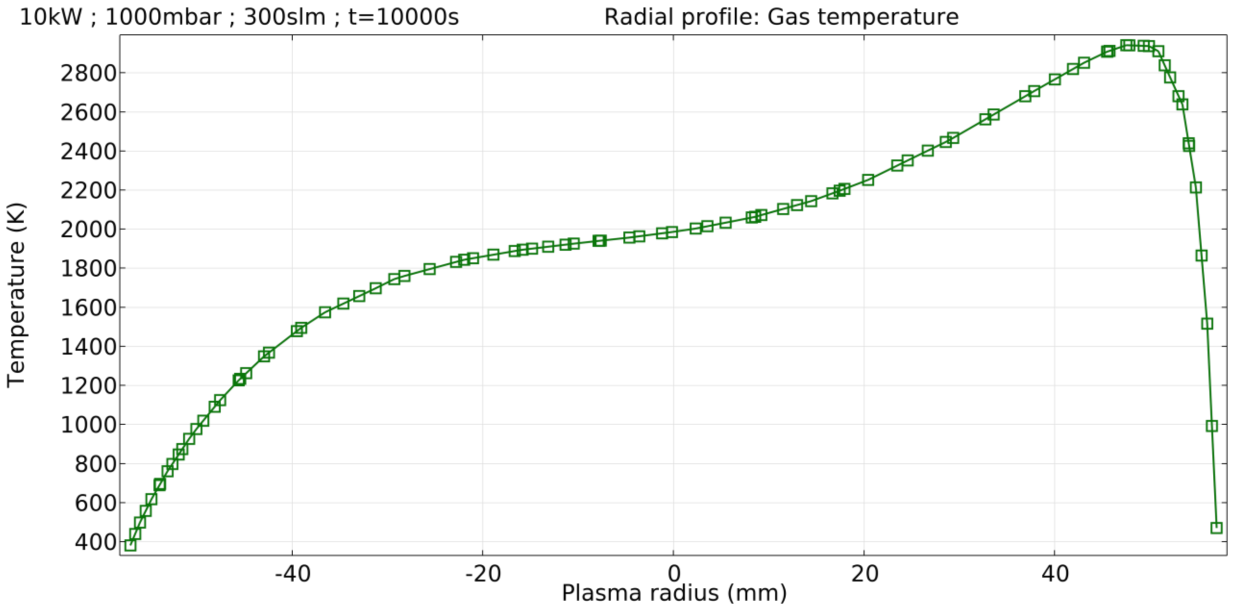
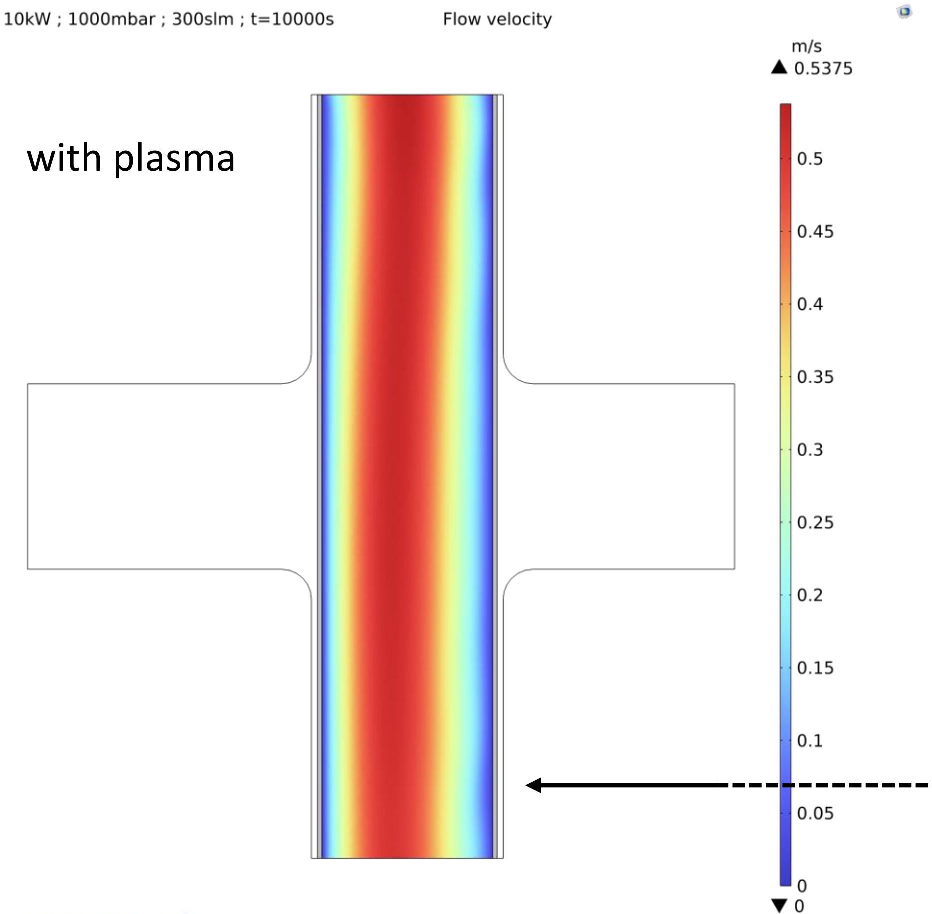
Results: Gas mixture temperature





Results: Gas mixture flow velocity



The change of the thermodynamic and fluid properties of the gas mixture with the gas temperature may affect the flow velocity.

Conclusions

- **A fully-coupled microwave plasma model at atmospheric pressure** has been successfully achieved in Ar with COMSOL Multiphysics®

- **Waveguide-to-coaxial coupling** has been recovered in the presence of a cylindrical plasma crossing a rectangular waveguide as expected from the theory of the transmission lines

- **Skin effect** has been observed as expected from the high-pressure plasma theory

- **A rise of the gas mixture temperature** has been observed according the thermodynamic properties and the wave-heating due to the electrons in a resistive plasma

Next steps

- **Next works** will focus on:
    - The EEDF’s when they are computed from the Boltzmann equation
    - The operating conditions and design
    - The gas flow regime at higher mass flow rates
    - Other feed gases with more by-products
    - Heavy-heavy particle collisions
    - Radiative heat transfers

Thank you for your attention