Simulations of atmospheric pressure microwave plasma torch in Ar/H_2 mixture using Matlab and COMSOL Multiphysics API

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Abstract

This contribution presents a fluid plasma model for simulating high-frequency discharges. The model is implemented in Matlab with COMSOL Multiphysics API which proved to be a very good compromise between flexibility and work efficiency. The model has so far been used for simulations of an atmospheric pressure microwave plasma torch and a low-pressure microwave reactor with linear antennas. This contribution focuses only the application in the microwave plasma torch.

1 Introduction

Fluid plasma models are an established way of simulating macroscopic laboratory plasma. They allow the user to estimate the densities and mean energies of the ionized and excited species and those of electrons. These quantities are crucial for characterizing the plasma and for its applications in syntheses or surface treatment.

The model and algorithm presented in this contribution are used at the Department of Physical Electronics to model two types of discharges, an atmospheric pressure microwave plasma torch and a low-pressure microwave reactor with linear antennas. The former is used at the Dept. Phys. El. of the Masaryk University for fast synthesis of carbon nanotubes (CNTs) [1] and their applications in sensor devices. It has also been successfully used for fast synthesis of maghemite (γ-Fe_2CO_3) nanoparticles [2]. The low-pressure MW reactor is used at the Institute of Physics of the AS CR for synthesis of nanocrystalline diamond (NCD) and its bioapplications.

Even though the two systems differ greatly in the operating conditions (pressure/temperature) and geometry, there are common aspects to be found. Most importantly, this is the composition of the plasma since the MW plasma torch uses $Ar/H_2/CH_4$ for the CNT syntheses and the MW reactor with linear antennas uses H_2/CH_4 to synthesise NCD. This contribution describes only the application of the algorithm to the microwave plasma torch.

The model described below is implemented in Matlab with strong help of COMSOL Multiphysics API. COMSOL's Partial Differential Equation (PDE) module is used for all the equations except the gas flow equations in the microwave plasma torch.

2 Model

The underlying idea of fluid plasma models is to substitute the electrons, ionized and excited species by a massless charged fluid with given properties. In addition to the particle transport equations, the energy equations of the individual species have to be solved. Moreover, since the plasma is weakly ionized, the properties of the neutral gas must be taken into account. Therefore, the model consists of three parts described below. These three parts are linked together via an iterative loop illustrated in fig. 1. The individual parts of the loop are described in greater detail below.

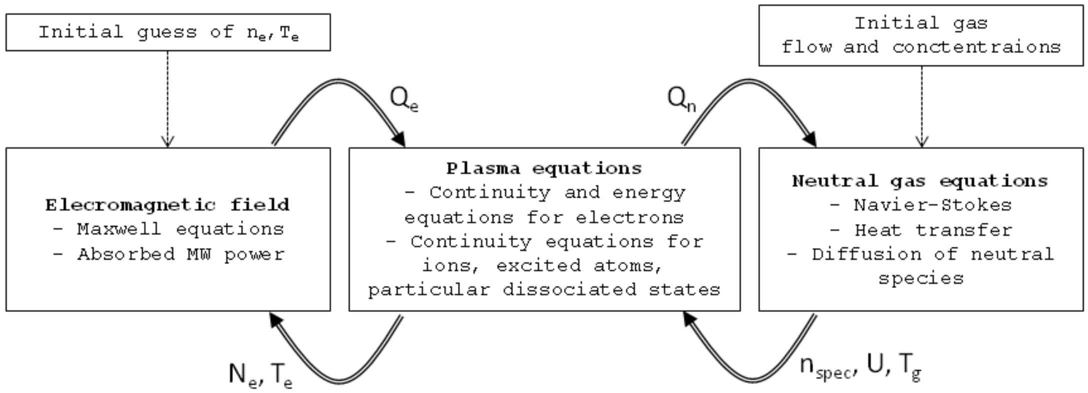


Figure 1: The iterative loop employed in the model

2.1 Electromagnetic field model

The electromagnetic field model solves the wave equation in the time-harmonic approximation

$$\nabla \times \left[\hat{\mu}^{-1} (\nabla \times \mathbf{E}_0)\right] - k_0^2 \left(\hat{\varepsilon_{\mathrm{r}}} - i\frac{\sigma}{\omega \varepsilon_0}\right) \hat{\sigma} \mathbf{E}_0 = 0. \tag{1}$$

The internal volume of the reactor is then perceived as a dielectric with spatially inhomogeneous complex dielectric function [3]

$$\varepsilon = \varepsilon_0 - \frac{\varepsilon_0 \omega_{pe}^2}{\nu_{en}^2 + \omega^2} - i\frac{\varepsilon_0 \omega_{pe}^2 \nu_{en}}{\omega\left(\nu_{en}^2 + \omega^2\right)} \tag{2}$$

In the equation (2), ε_0 denotes vacuum permittivity, $\omega = 2\pi \cdot 2.45 \cdot 10^9\ \mathrm{rad} \cdot \mathrm{s}^{-1}$ is the angular frequency of the EM radiation, ω_{pe} is the plasma frequency and ν_{ne} is the total electron-neutral collision frequency. The spatial inhomogeneity lies in the plasma frequency (which depends on the electron density) and in the collision frequency (which is a function of both electron energy and neutral gas density).

The most important input to the EM field model is the electron density n_{e} and temperature T_{e}. In the zeroth step, there is an initial condition of homogeneous electron density and temperature while in further iterations, n_{e} and T_{e} are taken from the plasma model. The most important output of the model is the amount of energy absorbed by the plasma, Q_{e} due to the complex dielectric function (2).

2.2 Gas flow

In the MW plasma torch, the gas is introduced to the discharge chamber through a nozzle with two inlets, each with diameter of around 1 mm (see figure 2c). At flow rates of 500 sccm for argon and 200 sccm for hydrogen, this leads to the Reynolds number of approx. 60 000. Therefore, the gas flow in the discharge chamber must be considered turbulent. In order to simulate the turbulent flow, the equations predefined in COMSOL's CFD library were used.

The model solves the so-called Reynolds-averaged Navier-Stokes (RANS) equation [4]. The main idea in this approach is separating the gas velocity into a steady component $\mathbf{U}(\mathbf{r})$ and a component $\mathbf{u}_{\mathrm{T}}(\mathbf{r}, t)$ oscillating around zero, accounting for the turbulence. Substituting this to the general Navier-Stokes equation and averaging over time leads to a formally similar equation with one extra term

$$\rho \frac{\partial \mathbf{U}}{\partial t} + \rho \left(\mathbf{U} \cdot \nabla\right) \mathbf{U} - \nabla \cdot \langle \rho (\mathbf{u}_{\mathrm{T}} \otimes \mathbf{u}_{\mathrm{T}}) \rangle = -\nabla \cdot \hat{P} - \nabla \cdot \mu \left[\nabla \otimes \mathbf{U} + (\nabla \otimes \mathbf{U})^T\right] + \mathbf{F}.$$

The third term on the left-hand side in the eq. (2.2) is modelled using the k–ε model, also included in COMSOL. Since the turbulence is generated only in a small region, the values of the turbulent kinetic energy k were adjusted manually. More information on the model are to be found for instance in [5]

In addition to the RANS equation, the heat equation and the diffusion equation for Ar and H_2 are solved. The heat equation takes the form

$$\rho C_p \frac{\partial T}{\partial t} + \rho C_p \mathbf{U} \cdot (\nabla T) = \nabla \cdot (\lambda \nabla T) + Q_{\mathrm{n}}, \tag{3}$$

where Q_{n} denotes the power transferred from the plasma to the neutral gas, as taken from the plasma model.

Solving the neutral gas flow equations is alone a non-trivial problem since the thermodynamic and transport properties of such a mixture are often non-linear and non-monotoneous. The detailed analysis of this problem has been previously carried out by one of the authors in [6]

2.3 Plasma model

The plasma model is the central part of the algorithm. It solves the continuity equations for ions, electrons and excited species in the form:

$$\frac{\partial n_j}{\partial t} + \nabla \cdot \mathbf{\Gamma}_j + (\mathbf{U} \cdot \nabla) n_j = R_j, \tag{4}$$

where n_j is the density of j-th species, $\mathbf{\Gamma}_j$ is the flux, $\mathbf{U}$ is the flow velocity (considered same for all species) and R_j is the source term. The flux of each species is assumed in the form proposed in [7]

$$\mathbf{\Gamma}_j = -\mu_j \mathbf{E} n_j - D_j \nabla n_j, \tag{5}$$

where μ_j is the mobility and D_j is the diffusivity. These quantities are obtained before the actual calculations from BOLSIG+ software, solving the Bolzmann kinetic equation in two-term approximation [7]. An equation formally similar to (4) is solved for the electron energy, as proposed in [7].

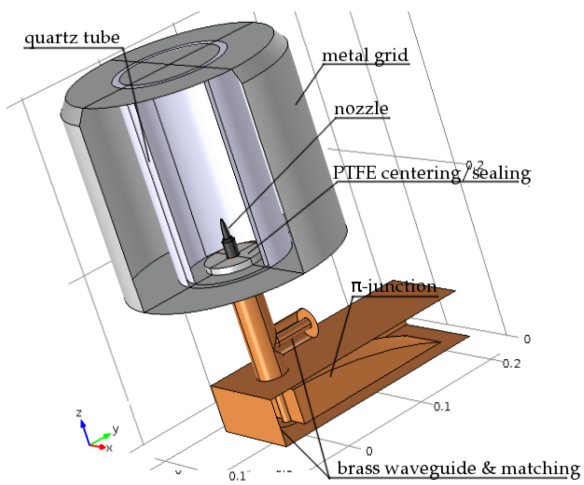
The plasma model contains 11 species, in particular Ar, Ar^+, Ar^*, H_2, H, H^+, H_2^+, H_3^+, H(n=2), H(n=3) and electrons. This results in 9 continuity equations to be solved self-consistently. The reactions that were considered, along with references, are shown in table 1. Apart from those, various rotational and vibrational collisions were considered in the electron energy equation. The plasma model takes data both from the EM field model (the absorbed

$e^- + Ar \rightarrow 2e^- + Ar^+$	[8]	$H_2 + H(n=2) \rightarrow H_3^+ + e^-$	[10]
$e^- + Ar \rightarrow e^- + Ar^*$	[8]	$H_2 + H(n=3) \rightarrow H_3^+ + e^-$	[10]
$e^- + Ar^+ \rightarrow Ar$	[8]	$H_2 + H_2^+ \rightarrow H_3^+ + H$	[10]
$e^- + H_2 \rightarrow 2e^- + H_2^+$	[9]	$e^- + H^+ \rightarrow H(n=2)$	[11]
$e^- + H_2 \rightarrow e^- + H + H(n=2)$	[9]	$e^- + H^+ \rightarrow H(n=3)$	[11]
$e^- + H_2 \rightarrow e^- + H + H(n=3)$	[9]	$e^- + H_2^+ \rightarrow H + H(n=2,3)$	[10]
$e^- + H \rightarrow e^- + H^+$	[9]	$e^- + H_3^+ \rightarrow H_2 + H(n=3)$	[10]
$e^- + H \rightarrow e^- + H(n=2)$	[9]	$2H + H_2 \rightarrow H_2 + H_2$	[12]
$e^- + H \rightarrow e^- + H(n=3)$	[9]	$3H \rightarrow H_2 + H$	[12]

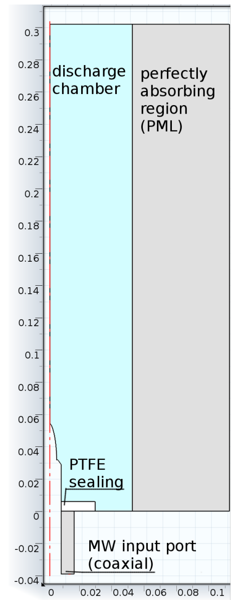
power Q_{e}) and from the gas flow model (the neutral gases' concentrations n_{Ar}, $n_{\mathrm{H_2}}$, neutral gas temperature T and neutral gas velocity $\mathbf{U}$). Its outputs are the electron density and temperature, which are used in the EM field model, and energy lost in elastic collisions Q_{n}, which heat the neutral particles.

3 Geometry

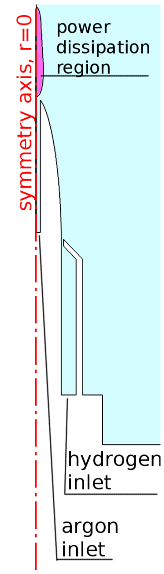
The plasma was simulated in 2D axisymmetrical geometry. Even though the device does not in fact possess such geometry (see fig. 2), only the discharge chamber with the coaxial waveguide were considered and it was assumed that all the input power is at all times matched to the discharge chamber.



(a) The whole MW plasma torch



(b) The computational domain



(c) Nozzle detail

Figure 2: The geometry

4 Results

This section presents the initial results obtained from the model. Initially, the gas flow was simulated for plasma of a fixed shape (determined experimentally). The results for typical conditions are shown in fig. 3. The results have shown, among other things, that the plasma

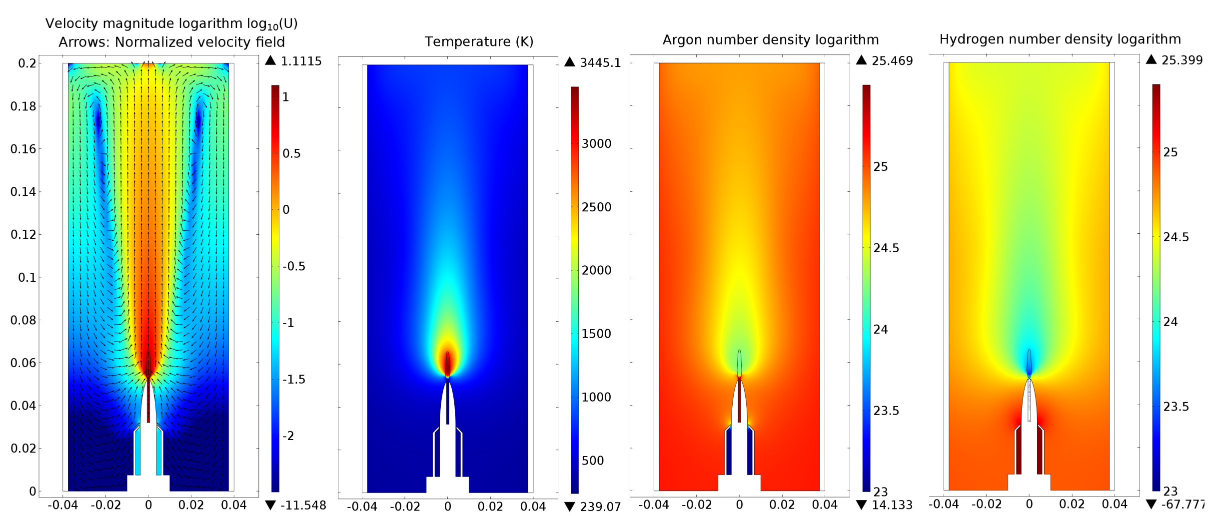
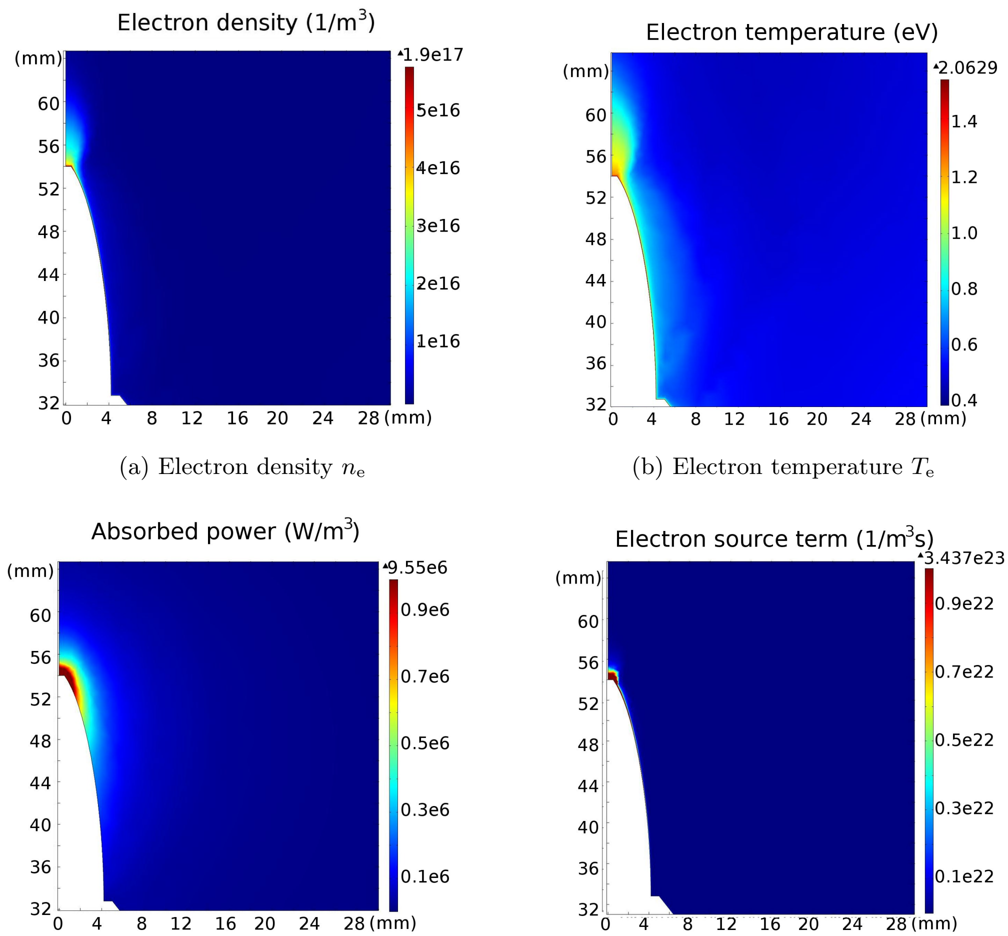


Figure 3: Gas flow simulation results for 500 sccm Ar flow rate (central inlet) and 200 sccm H_2 flow rate (outer inlet)

normally referred to as atmospheric-pressure plasma does actually operate at pressures one order lower than the atm. pressure, due to the high temperature in the discharge chamber (3000 - 4000 K)

Subsequently, first simulations were carried out using the complete model. The preliminary results are show in fig. 4. Due to high computational costs, it was not yet possible to perform a sufficient number of iterations. It is estimated (based on [12]) that convergence would be reached after 10 - 15 iterations.

The calculated electron temperature is in very good agreement with measurements performed on similar devices [13]. The calculated electron density on the other hand is of the order 10^{17} m^{-3} while the expected value was of the order 10^{19} m^{-3} [13]. This discrepancy can be explained by the insufficient number of iterations. The simulations have also brought very important qualitative informatiomation such as that most of the power gets absorbed at the very tip of the nozzle and the electrons are subsequently transported upwards, together with the neutral gas.



(a) Electron density n_e

(b) Electron temperature T_e

(c) Power absorption Q_e

(d) Electron production term R_e

Figure 4: Plasma simulation results

4.1 Conclusion and acknowledgements

A fluid model for simulating high-frequency discharges was implemented using Matlab and COMSOL Multiphysics API. The model was applied to the experimental setup of a microwave plasma torch and first results were obtained. These results have shown partial agreement to the experiment, but it is reasonable to expect, that increasing the number of iterations of the algorithm would improve the agreement. It will also be necessary to perform sevelar verification experiments (e.g. optical emmision spectroscopy, Schlieren fotografie).

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