Numerical simulations of a dielectric barrier discharge in a mixture of argon, hydrogen and oxygen

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This contribution presents numerical simulations of gas dynamics and mixing and chemical balance in a volume dielectric barrier discharge. The discharge is ignited in a mixture of argon and hydrogen with a relatively small admixture of oxygen and it is typically used for decomposition of hydride compounds for analytic purposes. The numerical model presented is complemented by laser-induced fluorescence measurements of hydroxyl radicals and monoatomic oxygen radicals.

1. Introduction

The presented contribution focuses on volume dielectric barrier discharge (DBD) ignited in a mixture of argon, hydrogen and oxygen. The discharge is used for decomposition of hydride compounds for analytic purposes and enables sensitive detection of metals, such as arsenic, mercury or bismuth [1].

The device is characterized using a numerical model describing the gas dynamics in full 3D geometry and simplified plasma chemistry in a 2D cross section through the discharge. There is one-way coupling between the gas dynamics and the chemistry models since the background gas velocity and composition are a necessary input to the chemistry part of the model. The model is implemented using the COMSOL Multiphysics simulation platform, which uses the finite element method for the discretization of the differential equations.

2. Setup

The discharge is ignited in a silica glass vessel which is open to ambient atmosphere on the sides, as illustrated in figure 1.

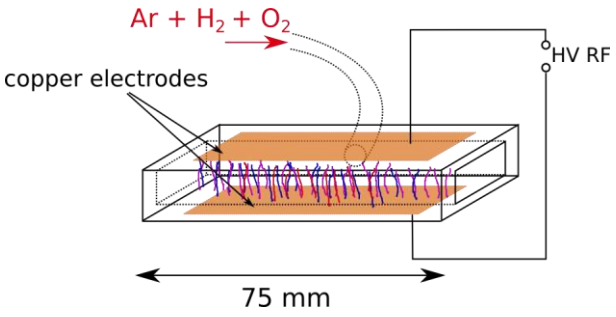


Figure 1: A schematic view of the dielectric barrier discharge. The setup is opened to ambient atmosphere on the sides.

The discharge is ignited in the mixture of argon, hydrogen and oxygen with argon being the dominant gas. The flow rate of argon is typically Q_{Ar} = 145 sccm, the flow rates of oxygen and hydrogen are varied in the experiment but the values adopted in the simulation are Q_{O2} = 5 sccm, Q_{H2} = 22 sccm. The gases are supplied through a circular inlet, as also shown in figure 1.The plasma is ignited using high-voltage electric field oscillating with 24 kHz frequency.

3. Model

As already mentioned, the numerical model consists of two parts, the background gas dynamics and the plasma-chemical reactions.

The background gas dynamics part solves the Navier-Stokes equations and diffusion equations for O_2, H_2 and air self-consistently, in full three-dimensional geometry. Ambient air is, in this case, considered a single species with given averaged properties [2]. Since the background-gas mixture is multi-component, the diffusion model that is used is Maxwell-Stefan diffusion with binary diffusion coefficients calculated from the Chapman-Enskog theory [2].

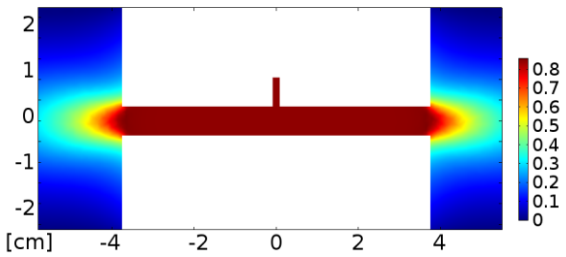
Given the gas flow velocities, turbulence does not have to be taken into account and the Navier-Stokes equation, therefore, takes the well-known form

$$\rho_m(\mathbf{u}\cdot\nabla)\mathbf{u} = \nabla\cdot\left[\mu_m(\nabla\mathbf{u}+(\nabla\mathbf{u})^T)-\frac{2}{3}\mu_m(\nabla\cdot\mathbf{u})\right]+\rho_m\mathbf{g}$$

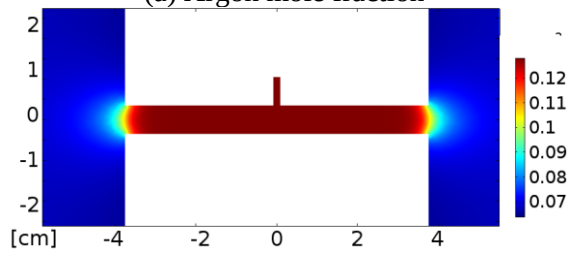
In the equation above, **u** is the mixture velocity ρ_m is the mixture density and μ_m is the mixture viscosity. The mixture density and viscosity are functions of the local gas composition and are calculated using Wilke's mixture rules [3]. The properties of individual components are taken from the NIST database [4].

The 3D background gas dynamics and mixing model is an input to the chemical model describing the transport and balance of various species in the plasma. The plasma chemistry model is still work-in-progress and the reaction scheme is largely based on that proposed in [5,6]. It solves multiple transport equations for the reactive species in two-dimensional horizontal cross sections through the discharge. In order to achieve reasonable computation times, the list of species and reactions has to be reduced substantially compared, for instance, to the 1D model presented in [5]. Luckily, the reference also identifies some of the most important reaction pathways, which serves as a good starting point for the development of the 2D model.

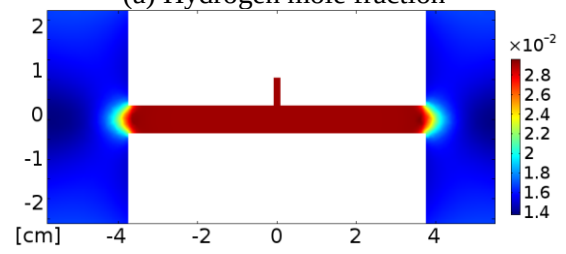
Since laser-induced fluorescence experiment suggests that the number densities of the reactive species do not exceed 10^{22} m^{-3}, it is reasonable to assume that the dissociation and ion formation induced by the plasma does not influence the gas dynamics. Therefore, the background gas dynamics and plasma chemistry models do not have to be solved self-consistently.



(a) Argon mole fraction



(a) Hydrogen mole fraction



(c) Oxygen mole fraction

Figure 2: Mole fractions of the background gases plotted in a horizontal cross-section through the discharge chamber for Q_{Ar} = 145 sccm, Q_{H2} = 22 sccm and Q_{O2} = 5 sccm.

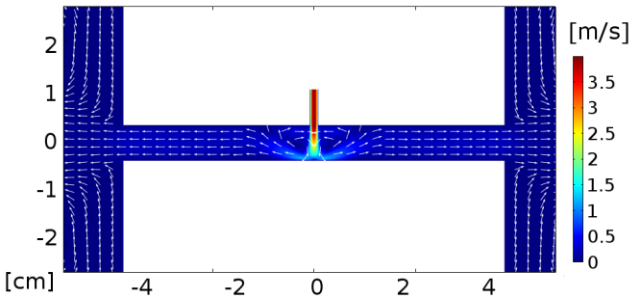


Figure 3: Gas velocity magnitude and direction plotted in a horizontal cross-section through the discharge chamber for Q_{Ar} = 145 sccm, Q_{H2} = 22 sccm and Q_{O2} = 5 sccm.

4. Results

Figure 2 shows the mole fractions of the background gases within the discharge chamber and in its close proximity. It shows that there is no substantial back diffusion of ambient air to the discharge chamber and, therefore, the reaction scheme inside the discharge chamber does not have to include nitrogen and water vapour impurities. Figure 3 shows the gas velocity and direction within the discharge chamber. Apparently, some re-circulation occurs near the gas inlet but the flow stabilizes within approx. 1 cm.

5. References

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