Modeling of radio frequency argon dielectric barrier discharge at atmospheric pressure in argon

A A Saifutdinova
A.N.Tupolev Kazan National Investigation Technical University
E-mail: aliya_2007@list.ru

Abstract. In this work modeling of atmospheric pressure barrier discharge in argon for rf-voltage signal applied to the electrodes was conducted. The model included balance equations for the densities of charged (electrons, ions) and the excited particles, the electron energy density, and the Poisson equation for the electric potential. The fluxes of charged and excited particles (electrons, ions) and electron energy were given in the drift-diffusion form. Distributions of spatiotemporal parameters of the discharge were obtained.

1. Introduction
Atmospheric pressure dielectric barrier discharges (DBD) have recently commanded much attention, fuelled by their promise to rival the widely used low-pressure glow discharges and their facilitation of numerous applications through the removal of the usually indispensable vacuum chamber [1,2]. They are capacitive nonthermal plasmas generated between two parallel electrodes that are either metallic [3,4] or coated with a dielectric layer[1,5], and their generation has been achieved over a very wide spectrum from kilohertz[1,5] and megahertz [3,7] to microwave [8]. They can also be generated as surface-wave discharges [9]. Today atmospheric pressure DBD are finding wide ranging applications including etching, deposition, surface modification, and sterilization [1,2]. The progress of their fundamental understanding, on the other hand, lags markedly behind their technological advancement largely due to comparatively few theoretical studies so far [10–13].

In this paper, atmospheric pressure (DBD) in argon initiated by the applied rf-voltage is numerically investigated.

2. Description of the Mathematical Model
The spatiotemporal characteristics of the DBD were determined using an extended hydrodynamic model describing the parameters of the gas-discharge plasma [17]. The model is based on the drift-diffusion equations and includes $k$ equations for the densities of charged (electrons, ions) and excited particles $n_k$ and the balance equation for the electron energy density $n_e$. The self-consistent electric field is determined from Poisson’s equation for the potential $\varphi$. The system of equations has the form

$$\frac{\partial n_k}{\partial t} + \nabla \cdot \Gamma_k + (\mathbf{u} \cdot \nabla) n_k = \sum_j c_{k,j} R_j$$

(1)
due to elastic collisions. Here, resonance.

corresponding inelastic process, and three positive ion species

temperature, which is assumed to be equal to room temperature (293.15 K), and δ = 2/

Here, q_e is the charge of an electron, e_0 is the permittivity of vacuum, Z_k is the charge of a particle of species k, E is the electric field, n_k = n_k<e> is the average electron energy density, and <e> is the average electron energy. The flux densities of charged particles Γ_k, as well as the electron energy flux density Q, are described in the drift-diffusion approximation,

\[ \frac{3}{2} \frac{\partial n_k}{\partial t} + \nabla \cdot Q + (u \cdot \nabla) n_k = -q_e \Gamma_e \cdot E - S_{el} - \sum_j \Delta \varepsilon_j R_j, \]  

\[ \Delta \varphi = -\frac{q_e}{e_0} \left( \sum_{k=1}^{N} Z_k n_k \right), \quad E = -\nabla \varphi. \]  

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\[ \Gamma_k = -D_k \nabla n_k + \varepsilon_0 \mu_k E n_k, \]  

\[ Q = -D_k \nabla n_k - \mu_k E n_k. \]  

Here, \( \mu_k \) and \( D_k \) are the mobilities and diffusion coefficients of charged particles, respectively; \( \mu_k = 5/3 \mu_e; \) and \( D_k = 2/3 \mu_e T_e \), where \( \mu_e \) is the electron mobility and \( T_e = 2/3 <e> \) is the electron temperature defined as 2/3 of the average electron energy.

The right-hand side of Eq. (2) is determined by the reactions occurring in the discharge. Each reaction makes a positive contribution to the source function if it results in the production of the corresponding particle species and a negative contribution if the particles of this species disappear. In this equation, \( j \) is the reaction index (28 plasma chemical reactions in total were taken into account) and \( c_{kj} \) is the number of particles of the \( k \)th species produced in the \( j \)th reaction (\( c_{kj} \) can be either positive or negative). The reaction rates \( R_j \) are determined by the rate constants \( k_j(T_e) \) of the corresponding processes and are proportional to the product of the densities of the reacting species,

\[ R_j = k_j(T_e) \prod_{k=1}^{N} n_k, \]  

where \( N = 2 \) for reactions involving two species and \( N = 3 \) for reactions involving three species. The first summand on the right-hand side of Eq. (2) describes Joule heating of electrons in the electric field. The second summand, \( S_{el} = (3/2) 3 \varepsilon_0 m_e n_e(T_e - T) \) describes energy exchange between electrons and neutral gas particles due to elastic collisions. Here, \( T \) is the gas temperature, which is assumed to be equal to room temperature \( (T = 293.15 \text{ K}) \), and \( \delta = 2m_e/M_a \) is the doubled ratio of the electron mass \( m_e \) to the gas atom mass \( M_a \). The third summand takes into account inelastic energy losses of the electron gas. Here, \( \Delta \varepsilon \) is the energy lost (or acquired if \( \Delta \varepsilon < 0 \)) by an electron in a given reaction and \( R_j \) is the reaction rate, which is determined by the rate constant of the corresponding inelastic process, \( R_j = k_j(T_e) n_e n_k \), where \( n_e \) is the electron density and \( n_k \) is the density of neutral particles.

The set of plasmachemical reactions used to describe a discharge in argon is presented in Table 1. It takes into account three effective excited atomic levels (metastable Ar^+, resonance Ar^+, and excited Ar* ones), two effective excimer levels (Ar_2^+ and Ar_3^+), and three positive ion species (Ar^+, Ar_2^+, and Ar_3^+). In addition, the effective process \( e + Ar \leftrightarrow e + Ar^+ \) with a threshold energy of \( \Delta \varepsilon = 13.9 \text{ eV} \) was introduced to correctly take into account inelastic energy losses of the electron gas in Eq. (2). The cross section for this process was found similarly to [18] by subtracting the cross sections for individual processes of excitation of high-lying levels (above 13.9 eV) [17] from the total excitation cross section taken from.

The coefficients of electron mobility and diffusion, as well as the rate constants of elastic and inelastic electron-impact processes, were calculated using the electron energy distribution function (EEDF). The EEDF was found by solving the Boltzmann kinetic equation in the two-term \( (f_0, f_1) \) approximation under the assumption of a local dependence on the electric field strength. The kinetic equation took into account electron heating in the longitudinal electric field and the change in the electron energy due to elastic, electron–electron, and inelastic collisions.

All constants, transport coefficients (6), and the effective temperature were first calculated as
functions of $E/N$. Then, by virtue of the monotonic dependence $T_{e}(E/N)$, the constants and transport coefficients were redefined as functions of the effective temperature. The distribution of the latter was found from the electron thermal balance equation (2), in which not only volumetric processes but also heat conduction transfer were taken into account. Therefore, there was a spatial shift between the distributions of the field, electron density, and electron temperature. In other words, the profile $T_{e}(x)$ and, accordingly, the profile of the electron-impact ionization rate were spread over the electron thermal relaxation length; i.e., there appeared a nonlocal dependence of the discharge parameters on the electric field.

The ion mobilities $\mu_{p} = 6.86 \times 10^{-4} \text{ m}^{2}/(\text{V} \cdot \text{s})$, $\mu_{n} = 5.14 \times 10^{-4} \text{ m}^{2}/(\text{V} \cdot \text{s})$, and $\mu_{t} = 4.84 \times 10^{-4} \text{ m}^{2}/(\text{V} \cdot \text{s})$, where $p$ is the gas pressure, were taken from, while the diffusion coefficients were found from the Einstein relation.

The boundary conditions for the electron number density, electron energy density, and densities of ions and excited particles were specified at the boundaries of the discharge volume (i.e., at the inner surfaces of the dielectrics) as follows:

$$
-n \cdot \Gamma_{e} = -\alpha n_{e} \mu_{e} E \cdot n + \frac{1}{2} \sum_{p} \gamma_{p} (\Gamma_{p} \cdot n),
$$

$$
-n \cdot Q = -\alpha n_{e} \mu_{e} E \cdot n + \frac{5}{6} v_{e,th} n_{e} (1 - \alpha) \sum_{p} \gamma_{p,\bar{E}} (\Gamma_{p} \cdot n),
$$

$$
-n \cdot \Gamma_{k} = \left( \frac{1}{4} v_{e,th} n_{e} \right) - \alpha n_{k} \mu_{k} E \cdot n,
$$

where $n$ is the unit normal vector; $\gamma_{p} = 0.02$ is the secondary electron emission yield from the dielectric surface under bombardment by particles of the $p$th species; $\Gamma_{p}$ is the flux of particles of the $p$th species onto the electrode; $\bar{E}_{p}$ is the average energy of electrons emitted due to bombardment by particles of the $p$th species; and $v_{e,th} = \sqrt{8k_{b}T_{e}/\pi m_{e}}$ and $v_{i,th} = \sqrt{8k_{b}T_{i}/\pi M}$ are the thermal velocities of electrons and heavy plasma particles, respectively (here, $T_{e}$ is expressed in eV and it is set $T_{k} = T_{g}$).

It is assumed that $\alpha = 1$ if the drift component of the flux of charged particles of a given species, $z_{d} n_{i} \mu_{i} E$, is directed to the wall (i.e., if $z_{d} n_{i} \mu_{i} \cdot E > 0$) and $\alpha = 0$ in the opposite case. Here, the heavy particle (an ion or an excited particle) of a given species is assumed to lose its charge and decay (in our case, into Ar atoms) on the dielectric surface, i.e., a set of surface reactions is specified.

The potentials of the metal electrodes (adjacent to the outer boundaries of the dielectric) were specified as the boundary conditions for Poisson’s equation:

$$
\varphi = 0
$$

at the grounded electrode and

$$
\varphi = V(t)
$$

at the powered electrode.

At the interface between the plasma and dielectric, the discharge is accumulated at the latter. For the jump of the normal component of the electric induction at the dielectric surface, $D_{1} - D_{2}$, we have

$$
n \cdot (D_{1} - D_{2}) = \varepsilon_{1} \varepsilon_{0} n \cdot E_{1} - \varepsilon_{2} \varepsilon_{0} n \cdot E_{2} = \sigma,
$$

where $\varepsilon_{1}$ and $\varepsilon_{2}$ are the relative permittivities of the plasma and dielectric, respectively; $\varepsilon_{0}$ is the permittivity of vacuum; $E_{1}$ and $E_{2}$ are the normal components of the electric field in the dielectric and plasma, respectively; and $\sigma$ is the surface charge density accumulated on the barrier surface (it is defined only at the plasma–dielectric interface; in the one-dimensional case, it consists of two points).

For the surface charge density $\sigma$, the following equation is solved separately:
\[
\frac{d\sigma}{dt} = -n_e \sum_k z_k \frac{\Gamma_k - \Gamma_e}{\Gamma}
\]  
(13)

where the sum over \(k\) is extended to all ion species.

3. Numerical simulation and results
To model the spatiotemporal behavior of a dielectric barrier rf-discharge, a one-dimensional computation domain was defined. The discharge gap length was specified to be equal to \(L = 1\) mm, while the thicknesses of the dielectric barriers were assumed to be \(d = 1\) mm. The relative permittivity was set at 4, which corresponds to quartz. The feeding voltage amplitude was 1.5 kV.

Figure 1 \(a\) and \(b\) present the spatial distribution of the electron number density and electron temperature (along the horizontal axis) extruded in time for one period of the external excitation along the vertical axis. These results show that this discharge is similar to lower pressure RF capacitively coupled discharges. The electron density is static in time in the discharge center. However, near the wall the electron density evolves in time driven by the applied time-varying electric field. The electron temperature near the wall is also strongly time modulated attaining peak electron temperatures of 6.9 eV. In the plasma bulk the electron temperature is much cooler than near the walls and is much less modulated since the electric field is much less intense than in the sheaths.

![Figure 1](image)

**Figure 1.** \(a\) Electron number density and \(b\) electron temperature spatial and time evolution.

4. Conclusions
Based on the one-dimensional extended fluid model, the characteristics of dielectric barrier discharge excited rf voltage are numerically investigated in atmospheric pressure argon. All the main discharge characteristics were obtained.

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