Radio-Frequency Discharge at Low Pressure: a Non-Local Problem Statement Approach

V.T. Dubrovin\textsuperscript{a}, V.Ju. Chebakova\textsuperscript{a,}\textsuperscript{*}, V.S. Zheltukhin\textsuperscript{b}

\textsuperscript{a} Kazan Federal University, 18 Kremlyovskaya Street, Kazan 420008, The Russian Federation
\textsuperscript{b} Kazan National Research Technological University, 68 Karl Marx Street, Kazan 420015, The Russian Federation

Abstract

Self-consistent nonlocal nonlinear mathematical CCRF discharge model is constructed. The model includes the convection–diffusion equations for electronic and ionic gas, Poisson’s equation for electric field potential, the equation of balance of metastable as well as ground states neutral atoms, and stationary equation of heat conductivity for electronic and ionic gases. An algorithm for the numerical solutions of the model has been described. The results of test calculations of CCRF discharge characteristics at the interelectrode distance of 2.2 cm, the gas pressure of 13.3 Pa, the voltage amplitude of 65 V were obtained. Comparison of the results showed the accuracy of the results and proved the adequacy of the mathematical model and the method of calculation.

1. Introduction

Low-temperature plasma devices is used intensively in industries. Capacitive coupled radio-frequency (CCRF) discharge is effectively used in nanotechnologies for creating and modifying of nanostructures and for modification of natural polymeric materials, such as leather, fur and fabrics [1-3]. The characteristic features of the processing of this materials are a batch treatment, i.e. several samples must process at the same time. Thus an installation with

* Corresponding author. Tel.: +7-905-319-9652;
E-mail address: vchebakova@mail.ru
large-scale electrodes (~ 0.5x1.4 m²) as well as large-scale interelectrode distance (~ 0.2-0.3 m) should designing. CCRF discharges in pressure ranges below 13.3 Pa and from 133 Pa up to atmospheric pressure were examined [4-8]. Characteristics of CCRF discharge are very varied at different pressures. It is established that changes in gas and electronic temperatures lead to changes of other characteristics of plasma such as particle densities. The purpose of the present work is constructing a model of CCRF discharge at pressures in range from 13.3 to 133 Pa taking into account the electron and gas temperature.


The basic assumptions of the model are the following:
- The discharge is formed between two large-scale parallel electrodes; discharge properties is varied along the normal direction to the electrodes only (i.e., along x-axis).
- Plasma of CCRF discharge consists of four types particles such as neutral atoms in the main and excited states (metastable), electrons and positive one charge ions.
- Temperatures of atoms, metastable and ions is identical. Therefore the equations for metastable and positive ions temperature are not considered in this model.
- Magnetic field effects are not included.
- Negative ions are neglected.

The mathematical model include several initial and boundary value problems:
- Poisson's equation for electric field potential

\[-\frac{\partial^2 \phi(x,t)}{\partial x^2} = \frac{q_e}{\epsilon_0} \left( n_e(x,t) - n_i(x,t) \right), \quad 0 < x < l, t > 0, \tag{1}\]

Boundary conditions at x = 0, l are the following:

\[\phi(0,t) = 0, \quad \phi(l,t) = V_a \sin(2\pi ft). \tag{2}\]

Here l is the inter electrode distance, \(n_e, n_i\) is the electron and the positive ion densities, respectively, \(q_e\) is the electron charge and \(\epsilon_0\) is the electric constant, \(f = 13.56\) MHz, \(V_a\) is the RF voltage magnitude,
- The convection – diffusions equation for electronic gas:

\[\frac{\partial n_e}{\partial t} + \frac{\partial G_e}{\partial x} = R_1 n_e N + R_2 n_e^2 + R_3 n_m n_e - R_4 n_e n_i - R_5 n_i^2, \quad 0 < x < l, \quad t > 0, \tag{3}\]

Boundary conditions at x = 0, l are the following:

\[G_e = \begin{cases} \pm n_e v_e - \gamma \mu_e n_e E, & \text{when the electric field is directed into the electrode,} \\ \pm n_e v_e - \mu_e n_e E, & \text{when the electric field is directed from the electrode,} \end{cases} \tag{4}\]

Here \(G_e\) is the diffusion flux of electrons, \(N = \frac{P}{(kT_e)}\) is density of neutral gas, \(P\) is the gas pressure, \(k\) is the Boltzmann's constant, \(E = -\frac{\partial \phi}{\partial x}\) is the electric field, \(R_1 + R_2 n_e\) is the electron recombination coefficient, \(R_3 n_m\) describes the ionization process from the ground state, \(v_e\) is thermal velocity of electrons, \(\mu_e, \mu_i\) are mobility of electrons and ions, \(\gamma\) is the secondary electron emission coefficient, \(n_m\) is the metastable density, \(R_3, R_5\) is rate of step-wise ionization \(Ar^+ + e \rightarrow Ar^+ + 2e\) and pooling reactions \(Ar^+ + Ar^+ \rightarrow Ar^+ + Ar^+ + e\). Here \(Ar^+\) represents
the first excited state of argon, \( Ar^+ \) is positive ion, \( e \) designed the electron, \( G_e = -\frac{\partial D}{\partial x} n_e - \mu_e n_e E \), \( D_e \) is electron diffusion coefficient.

- The equation of convection–diffusion of ions gas

\[
\frac{\partial n_i}{\partial t} + \frac{\partial G_i}{\partial x} = R_i n_e N + R_{ii} n_m n_e + R_{ii} n_m n_e - R_i n_e n_e - R_{ii} n_m n_e, \quad 0 < x < l, \quad t > 0,
\]

(5)

Boundary conditions at \( x = 0, l \) are the following:

\[
G_i = \begin{cases} 
\pm n_i v_i + \mu_i n_i E, & \text{when the electric field is directed on the electrode,} \\
\pm n_i v_i, & \text{when the electric field is directed from the electrode,}
\end{cases}
\]

(6)

Here \( G_i \) is diffusion flux of ions, \( v_i \) is thermal velocity of ions, \( D_i \) is electron diffusion coefficient.

- The metastable density balance equation:

\[
\frac{\partial n_m}{\partial t} - \frac{\partial G_m}{\partial x} = R_m n_e N - R_m n_m n_e - R_{ii} n_m n_e - R_m n_e n_e - R_{ii} n_m n_e, \quad 0 < x < l, \quad t > 0.
\]

(7)

Boundary conditions at \( x = 0, l \) are the following:

\[
G_m = \pm n_m v_a
\]

(8)

Here \( G_m = \frac{\partial n_m}{\partial x} \), \( D_m \) is the metastable atom diffusion coefficient, \( R_m i = 6, 7, 8, 9 \) are coefficients of processes such as ground excitation to metastable \( Ar + e \rightarrow Ar^+ + e \), superelastic collision \( Ar^+ + 2Ar \rightarrow 2Ar^+ \), \( Ar^+ + 2Ar \rightarrow Ar_2 + Ar \), radiative decay \( Ar^+ \rightarrow Ar + hv \), further excitation \( Ar^+ + e \rightarrow Ar^++e \), \( v_a \) is thermal velocity of atoms.

The electron temperature balance

\[
\frac{3}{2} k \frac{\partial (n_e T_e)}{\partial t} + \frac{\partial}{\partial x} \left( \frac{5}{2} k T_e G_e - \lambda_e \frac{\partial T_e}{\partial x} \right) = n_e q_e E^2 - Q_{el} n_e - I R_i N_n e - I_i R_i n_m n_e, \quad 0 < x < b, t > 0
\]

(9)

Temperature on borders is equal to temperature of electrodes.
Here \( I \) is the ionization energy of 15.76 eV, \( I_i \) is the step ionization energy, \( V_e \) is the electron speed, \( \lambda_e \) is the thermal conductivity of electrons, the term \( Q_{el} \) is exchange energy in collisions of elastic scattering.

- The atoms temperature balance

\[
-\partial (\lambda_a \frac{\partial T_a}{\partial x}) = j_i E + Q_{el} N_n e
\]

(10)

Here \( \lambda_a \) is the thermal conductivity of atoms, \( j_i = q_i G_e \). Temperature on borders is equal to temperature of electrodes.

The dependences of equation coefficients on the electron temperature are presented in [9]. The initial values of variables were constant.
3. Analysis of model formulation.

The system is characterized by several features which are complicated of formulation of an algorithm and a numerical method of solution. These features are the following:
- the system include differential equation of several types: the boundary value problem which parametrically depend from time, initial and boundary value problem of parabolic type;
- quickly periodic oscillation of the equation solution;
- large gradients of solutions near boundaries;
- multilevel nonlinearities of the problem


The implicit finite-difference scheme on uniform grid was used for numerical realization of the problem. The convective transfer term was approximated by the directed differences. The Newton method was applied for linearization of nonlinear terms in right hand of the equations. The integro-interpolation method was used for creation of the conservative finite-difference scheme. The iterative method of nonlinearity removal to the previous layer was applied for linearization of parabolic equations and the whole system. Recalculation of gas temperature was made once during hundred temporary periods. Calculations were carried until the full charge balance on electrodes was reached: the electron charge removing from positive column is compensated by a positive ion charge exactly during one fluctuation of an electromagnetic field. Numerical methods for solving nonlinear systems of mathematical physics can be found in the works [14-20]

5. Results of the simulations.

The results of test calculations of CCRF discharge in the plasma torch at the interelectrode distance of 2.2 cm, the gas pressure of 13.3 Pa, the voltage amplitude of 65 V were obtained. Results were compared with experimental data [21]. Comparison of the results showed the tolerable accuracy of the results and the adequacy of the mathematical model and the method of calculation to experimental data (see Figure 1 and Figure 2, where square and diamond points correspond to the experimental data).

Figure 1. The average ion density in the interelectrode space in comparison with experimental datas [21](dashed line – calculation, squares – experiment).

Acknowledgements

This work was supported by the Russian Science Foundation (project 16-11-10299)
Figure 2. Distribution of the electron temperature in the interelectrode space at different times in comparison with experimental datas [21]. The electron temperature given in eV.

References