Mathematical simulation of the low-temperature plasma at the interaction with oil products

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Abstract. Self-consistent mathematical models of a non-equilibrium low-temperature plasma of a radio frequency capacitive discharge in argon in different pressure ranges have been created. These mathematical models allow us to calculate the internal structure and discharge parameters for controlling parameters of the plasma torches at solving the target problems in order to optimize their functioning. The methods for numerical realization of the proposed models are developed. The results of numerical calculations for test problems are analyzed.

1. Introduction

At present, the methods for studying the environmental safety problems with oil pollution are rapidly developing. The significant scale of oil inflow into the environment when developing oil and gas resources leads to the fact that this type of pollution is the main one for a lot of oil production areas. Currently, the most promising is the technology of joint processing of oil-containing wastes and/or heavy oil fractions using plasma processing methods applying a low-temperature plasma [1-4]. Plasma-chemical methods provide a higher degree of processing (conversion of raw materials is 96-98% of mass), increase the depth of processing into unsaturated hydrocarbons (more than 75%), allow using heavy oil fractions (kerosene and diesel) as raw materials. These methods also reduce the number of stages and branching of chemical processes.

Plasma technologies have also been found wide application in other areas of environmental protection. For example, in [5] a method is given for controlling algae cyanobacteria Microcystis aeruginosa in treatment plants. In [6], a breakthrough system is developed by assisting the acid hydrolysis with two-step microwave treatment to improve the more efficient saccharification process of starch and fiber from sago pith. In [7], the safe use of materials in contact with food in microwave discharges is investigated. When creating plasma plants are used in the petrochemical and chemical industries, it is usually difficult to carry out field experiments, since their carrying out requires considerable resource costs, and besides, experimental methods do not allow obtaining detailed information on internal plasma-chemical processes [8, 9]. Therefore, experimental and computational methods were used for the research. They mutually complement each other, allow solving a lot of problems of physics and chemistry of low-temperature plasma. This approach allows us to relate the internal and external parameters of the discharges. This problem is especially important for optimizing the operation of plasma plants [10-16].

The aim of this work is to create self-consistent mathematical models of the non-equilibrium low-temperature plasma of a radio frequency capacitive coupled (RFCC) discharge in argon at various pressures. They allow calculating the structure and internal parameters of the RFCC discharges for controlling the parameters of the RFCC discharge when solving target problems.
effective numerical algorithms implemented in the form of applied software have been developed to analyze the change in the components of the ionized inert gas as a function of pressure ranges and to identify the main factors affecting internal processes in RFCC discharges. The constructed self-consistent mathematical model describes a capacitive RF discharge between two plane-parallel electrodes, one of which is grounded and the other is connected to an RF generator (the frequency of the RF generator $f = 13.56$ MHz) with the inter-electrode distance which is smaller, than the dimensions of the electrodes themselves. Under these conditions, the electric field is close to the potential field and the discharge is homogeneous along the electrodes. It allows one-dimensional approximation to be used. Estimates of the time and distance over which electrons lose energy acquired from the electric field show that when simulating a low-pressure RF discharge we can use a non local approximation in which the parameters of the electronic component of the plasma depend on the electron temperature, whereas at higher pressures from the local value of the reduced electric field.

2. Statement of the problem
When constructing the model, we denoted $b$ as the distance between the electrodes. For the coordinate $x=0$ we take the grounded electrode, for the coordinate $x=b$ we take the loaded one (the $x$ axis is directed perpendicular to the surface of the electrode). When setting the boundary conditions, it is assumed that at $x=0$ the field is directed to the electrode, if $E<0$, and from the electrode, if $E \geq 0$, and at $x=b$ the field is directed to the electrode, if from the electrode, if $E<0$. A self-consistent model of the RFCC discharge at low pressures in the non local approximation contains the following equations with boundary conditions.

- Ion gas balance equation

$$
\frac{d}{dt}\left(n_+ \frac{d n_+}{dx}\right) + \frac{d}{dx}\left(-D_+ n_+ + \mu_+ n_+ E\right) = R_1 n_e N + R_2 n_m^2 + R_3 n_m n_e - R_4 n_e n_+ - R_5 n_e^2 n_+, \quad 0 < x < b, t > 0,
$$

$$
G_+(0, t) = \begin{cases} 
(-n_+ \nu_+ + \mu_+ n_+ E)_{(0+, t)}, & E < 0 \\
(-n_+ \nu_+ )_{(0+, t)}, & E \geq 0 
\end{cases},

G_+(b, t) = \begin{cases} 
(n_+ \nu_+ + \mu_+ n_+ E)_{(b-, t)}, & E \geq 0 \\
(n_+ \nu_+ )_{(b-, t)}, & E < 0 
\end{cases}.
$$

Here $n_e$, $n_+$, $n_m$ are the densities of electrons, ions and metastables, respectively, $\mu_+$ is the mobility of ions, $G_+ = -D_+ n_+/dx + \mu_+ n_+ E$ is the ion flow, $D_+$ is the ion diffusion, factor $E$ is an electric field intensity, $(E = -\partial \varphi / \partial x)$, $\varphi$ is the electric field potential, $n_e = \sqrt{8kT_e/(\pi m_e)} / 4$ is the average thermal velocity of ions, $m_+$ is atomic ion mass, $k$ is the Boltzmann’s constant, $T_e$ is a temperature of atoms. Hereinafter $R_i = 1 \ldots 9$ are the factors of plasma chemical reaction velocities ($Ar+e \rightarrow Ar^++2e$; $Ar^+Ar \rightarrow Ar^++Ar^+$; $Ar^+e \rightarrow Ar^+e$; $Ar^+2e \rightarrow Ar^++e$; $Ar+e \rightarrow Ar^+e$; $Ar^+Ar \rightarrow 2Ar$; $Ar^+e \rightarrow Ar^++e$) [17–21].

- Electron gas balance equation:

$$
\frac{d}{dt}\left(n_+ \frac{d n_+}{dx}\right) + \frac{d}{dx}(D_e n_e - \mu_e n_e E) = R_1 n_e N + R_2 n_m^2 + R_3 n_m n_e - R_4 n_e n_+ - R_5 n_e^2 n_+, \quad 0 < x < b, \quad t > 0
$$

$$
G_e(0, t) = \begin{cases} 
(-n_e \nu_e - \gamma \mu_e n_e E)_{(0+, t)}, & E < 0 \\
(-n_e \nu_e )_{(0+, t)}, & E \geq 0 
\end{cases},

G_e(b, t) = \begin{cases} 
(n_e \nu_e - \gamma \mu_e n_e E)_{(b-, t)}, & E \geq 0 \\
(n_e \nu_e - \mu_e n_e E)_{(b-, t)}, & E < 0 
\end{cases}.
$$

Here $G_e = -D_e n_e /dx - \mu_e n_e E$ is the electrons flow, $\mu_e$ is the mobility of electrons, $D_e$ is the electrons diffusion, $\gamma$ is the secondary electron emission factor, $n_e = \sqrt{8kT_e/(\pi m_e)} / 4$ is the average thermal velocity electrons, $T_e$ is temperature of electrons, $m_e$ is mass of an electron.

- Poisson’s equation for the electric field:

$$
-\frac{d^2 \varphi}{dx^2} = \frac{2e}{\varepsilon_0} (n_e - n_+), \quad a(r - 1) < x < b, \quad t > 0, \quad \varphi(0, t) = 0, \quad \varphi(b, t) = V_0 \sin(\omega t)
$$
where \( q_e \) is the electron charge, \( e_0 \) is the electrical constant, \( \omega \) is the angular frequency of electric field, \( V_a \) is voltage peak-to-peak amplitude on charged electrode.

- The balance equation of metastable atoms:

\[
\frac{dn_m}{dt} + \frac{d}{dx} \left( -D_m \frac{dn_m}{dx} \right) = R_8 n_e N - R_2 n_m^2 - R_3 n_m n_e - R_7 n_m - R_8 N_m - R_9 n_m n_e, \quad 0 < x < b, t > 0
\]

\[
G_m(0,t) = (-n_m \sqrt{8kT_a / (m_n / 4)}) \bigg|_{(0+b,t)}, \quad G_m(b,t) = (-n_m \sqrt{8kT_a / (m_n / 4)}) \bigg|_{(b-t)}
\]

where \( G_m = -D_m \frac{dn_m}{dx} \) is density of flow of metastable atoms, \( D_m \) is diffusion factor of metastable argon atoms.

- Balance equation for the electron temperature:

\[
\frac{3}{2} \frac{k}{\lambda_e} \frac{dn_e T_e}{dt} + \frac{d}{dx} \left( \frac{5}{2} kT_e G_e - \lambda_e \frac{dT_e}{dx} \right) = q_e n_e \left( \mu_e E^2 \right) - Q_{el} N_n e - I R_n e N_e - I_1 R_3 n_m n_e,
\]

\[
T_e(0,t) = T_e(b,t) = T_e \text{w}
\]

where \( \lambda_e \) is heat transfer ration of electron gas, \( \text{Here } l \) is the ionization energy of 15.76 eV, \( l_1 \) is the step ionization energy, the term \( Q_{el} \) is the exchange energy in collisions of elastic scattering.

- Balance equation for the atomic ion gas temperature:

\[
\frac{d}{dx} \left( -\lambda_a \frac{dT_a}{dx} \right) = j(E) + Q_{el} N_n e, \quad T_a(0,t) = T_a(b,t) = T_a \text{w}.
\]

Here \( \lambda_a \) is the thermal conductivity of atoms, \( J = q_e \mu_e n_e \) is the temperature on borders and is equal to temperature of electrodes.

A self-consistent model of the RFCC discharge at low pressures in the non local approximation contains the following equations with boundary conditions.

- The diffusion-drift equation for atomic ions:

\[
\frac{\partial n_a}{\partial t} + \frac{\partial G_a}{\partial x} = R_1 n_e N + R_2 n_m^2 + R_3 n_m n_e - R_4 n_e n_n - R_5 n_e^2 n_e + R_12 n_e n_{2n} - R_{11} n_m^2 + R_{20} N_{2n}, 0 \leq x < b, t > 0,
\]

\[
G_a(0,t) = \begin{cases} 
(n_e u_e + \mu_a n_a E)_{(0+b,t)} < 0, \\
(n_e u_e)_{(0+b,t)} \geq 0,
\end{cases} \quad G_a(b,t) = \begin{cases} 
(n_e u_e + \mu_a n_a E)_{(b-t)} < 0, \\
(n_e u_e)_{(b-t)} \geq 0,
\end{cases}
\]

where \( n_2 \) are densities of molecules positive ions, hereinafter \( R_n, \ i = 10...20 \) are velocity factors of plasma-chemical reactions: \( 2Ar + Ar^{2+} \rightarrow Ar^{2+} + Ar^{+} + Ar, Ar^{+} + Ar^{2+} \rightarrow Ar^{2+} + e, Ar^{2+} + e \rightarrow Ar + Ar, Ar^{2+} + Ar^{2+} + h v, Ar^{2+} + 2Ar \rightarrow Ar^{2+} + Ar + Ar^{2} + e, Ar^{2+} + Ar + Ar^{2+} \rightarrow e + 2Ar + Ar^{2+}, Ar^{2+} + e \rightarrow e + Ar^{2+} + Ar^{2}, e + Ar^{2} \rightarrow 2Ar^{2} + e, Ar^{2} + 2Ar \rightarrow 3Ar^{2} + h v, Ar^{2} + Ar \rightarrow 2Ar + Ar^{2} + Ar^{2} + e \rightarrow e + Ar^{2} + Ar^{2} + e, Ar^{2} + Ar \rightarrow 2Ar^{2} + e, Ar^{2} \rightarrow 2Ar + Ar^{2} + Ar^{2} + Ar \rightarrow 2Ar + Ar^{2} (\text{see } 22-24).

- The diffusion-drift equation for molecular ions:

\[
\frac{\partial n_{2+}}{\partial t} + \frac{\partial (G_{2+})}{\partial x} = R_{10} n_m^2 + R_{11} n_n N^2 - R_{12} n_e n_{2+} - R_{13} n_m n_{2+} + R_{16} n_{2+}^2 + R_{17} n_m n_{2+} - R_{20} N_{2n},
\]

\[
\begin{cases}
G_{2+}(0,t) = (-n_{2+} u_{2+} + \mu_{2+} n_{2+} E)_{(0+b,t)} < 0, \quad G_{2+}(0,t) = (-n_{2+} u_{2+} + \mu_{2+} n_{2+} E)_{(0+b,t)} < 0,
\end{cases}
\]

\[
\begin{cases}
G_{2+}(b,t) = (-n_{2+} u_{2+})_{(b-t)} \geq 0, \quad G_{2+}(b,t) = (-n_{2+} u_{2+})_{(b-t)} \geq 0,
\end{cases}
\]

where \( G_{2+} = -D_{2+} \frac{dn_{2+}}{dx} + \mu_{2+} n_{2+} E \) is the density of molecular ion flow, \( \mu_{2+} \) is the mobility of molecular ions, \( D_{2+} \) is the diffusion coefficient of molecular ions, \( u_{2+} = \sqrt{8kT_a / (m_{2+} / 4)} \) is the average thermal velocity of molecular ions, \( m_{2+} \) is the mass of atomic ion.

- The diffusion-drift equation for electrons:

\[
\frac{\partial n_e}{\partial t} + \frac{\partial G_e}{\partial x} = R_1 n_e N + R_2 n_m^2 + R_3 n_m n_e - R_4 n_e n_n - R_5 n_e^2 n_e + R_{10} n_m^2 - R_{13} n_m n_{2+} + R_{16} n_{2+}^2 + R_{17} n_m n_{2+},
\]
where $n_{2s}$ is the density of argon dimmers.

- Poisson's equation for the electric field:
  \[-\varphi''/\varphi = q_e (n_e + n_{2s} - n_i)/\varepsilon_0 \quad 0 \leq x \leq b, t > 0, \quad \varphi(0,t) = 0, \varphi(b,t) = V_a \sin(\omega t),\]

- Balance equation of metastable atoms:
  \[\frac{\partial n_m}{\partial t} - \frac{\partial (g_m)}{\partial x} = R_6 N_n e + R_{18} n_e n_{2s} - R_2 n_m^2 - R_3 n_m n_e - R_4 n_m - R_5 n_n -
  - R_9 n_m e - R_{10} n_m^2 - R_{15} N_n^2 n_m - R_{17} n_m n_{2s} - R_{19} n_m N^2,
  \]
  \[G_m(0,t) = (-n_m \sqrt{8kT_a/(\pi m_a)/4}) \bigg|_{(0+,t)}, \quad G_m(b,t) = (-n_m \sqrt{8kT_a/(\pi m_a)/4}) \bigg|_{(b-,t)},\]

- Kinetic equation for argon dimmers:
  \[\frac{\partial n_{2s}}{\partial t} = R_{19} n_m N^2 + R_{15} N^2 n_m - R_{14} n_{2s} - R_{16} n_{2s}^2 - R_{17} n_m n_{2s} - R_{18} n_m n_{2s} + R_{19} n_m N^2,\]

- Kinetic equation for neutral argon atoms:
  \[\frac{\partial N}{\partial t} = -R_1 n_e N + R_{21} n_m^2 + R_4 n_e n_s + R_5 n_s^2 n_s - R_6 N_n e + R_7 n_m + R_8 N_m n_m + R_9 n_m n_e - R_{11} n_s N^2 +
  + R_{12} n_e n_{2s} + 2R_{13} n_e n_{2s} + R_{14} n_{2s} - R_{15} N^2 n_m + 2R_{16} n_{2s} + R_{17} n_m n_{2s} + R_{19} n_m N^2,\]
  the initial conditions are based on ideal gas equation: $P/(kT_a(x,0)) = N(x,0), \quad n_{2s}(x,0) = 0$.

- Balance equation atomic ion gas temperature:
  \[-\frac{d}{dx} \left( -\lambda_a \frac{dT_a}{dx} \right) = j_i E + Q_{el} N_n e,
  \]
  \[-\lambda_a \frac{dT_a}{dx} \bigg|_0 = -\chi(T_a(0,t) - T_w), \quad -\lambda_a \frac{dT_a}{dx} \bigg|_b = \chi(T_a(b,t) - T_w),\]
  where $\lambda_a$ is the heat transfer ration of atom-ion gas.

3. Approximate method

The systems of boundary and initial boundary value problems are characterized by several features (nonlinearity, rapid growth of coefficients in narrow domains, different scales of processes, etc.) complicating its numerical solution and making it impossible to use traditional methods [25-28].

To solve the nonlinear systems proposed in the present paper, we used an approximate method based on preliminary finite-dimensional approximation of the problem by means of difference schemes with subsequent application of an iterative process for its implementation [29–32]. The finite-difference approximation for convection-diffusion equations for charged particles is constructed by the integro-interpolation method using the method of directed differences.

To solve the Cauchy problem, an implicit Eulerian scheme was used. When the quadratic nonlinearity is approximated, the Newtonian linearization is used in the right-hand sides of the equations. The flux densities for ion and electron gas were calculated using the Hummel-type method based on the solution found by the implicit scheme.

The numerical algorithm is based on the demolition of nonlinearity with respect to incoming coefficients (diffusion coefficients, mobility and rates of plasma-chemical reactions) on the lower layer. The linearization of the system was carried out using the Seidel-type method.
4. Results of calculations
The results of the numerical solution of the model problem revealed changes in the composition of the gas during the development of the discharge. If molecular ions first predominate at a small gas temperature and the concentrations of excited atoms and dimers are approximately equal, then when the gas is heated, the concentration of molecular ions and dimers decreases with increasing concentration of atomic ions and dimer reduction. The concentration of molecular ions is greater in the near-electrode layers, where the gas temperature is lower. In this case, a quasineutral region is conserved in the discharge. It was shown that in the case of significant gas heating, the change in the gas temperature in the interelectrode space begins to affect significantly the ratio of contributions to the formation and death of particles of various plasmochemical processes, and, consequently, affects the distribution as well as the fraction of charged (electrons, Molecular ions), and excited particles in the discharge gap, thereby determining the development of the discharge. In conclusion, we note that the results obtained in this paper can be used to develop models for the interaction of low-temperature plasma with various substances. These models play an important role in solving environmental safety problems, which arise, in particular, when processing pipelines for oil and gas and engineering infrastructure from composite materials in order to improve the operational reliability of oilfield pipeline systems. In addition, these models can be used to develop technologies for the joint processing of oily waste and/or heavy oil fractions using low-plasma systems.

Acknowledgments
The work is performed according to the Russian Government Program of Competitive Growth of Kazan Federal University. The publication was carried out with the financial support of the Russian Science Foundation (project No 16-11-10299).

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