

## Electrophysical and thermal properties of discharge with liquid (non-metallic) anode

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*The article presents the results of experimental and numerical studies of the properties of an electric discharge formed between a metal cathode and a liquid (non-metallic) anode at atmospheric pressure. The discharge is ignited by immersing the metal cathode (AMC-40 aluminum) into an electrolytic anode (3% NaCl in purified water). The electrophysical parameters of the discharge, including the volt-ampere characteristic, current pulsations and discharge voltage, are studied. The surface temperature of the electrodes in the discharge combustion zone is studied using infrared thermography. The article presents the results of numerical calculations of the evolution of the electron and ion components taking into account the plasma-chemical transformations occurring between them. The ionization and detachment processes are considered as sources of the electron component, and the attachment of electrons to neutrals and electron-ion recombination are considered as its sinks.*

**Key words:** *plasma-liquid systems, electric discharge, liquid electrodes, numerical methods, thermography.*

**Introduction.** Plasma-liquid systems that use non-metallic liquids, such as salt solutions in industrial, distilled or purified tap water, have valuable properties. Electric discharges formed in these systems are independent and can burn in a multi-channel or volumetric (diffuse) form when powered by direct current. As a rule, the range of atmospheric air pressures at which the discharge is studied varies from  $10^5$  to  $10^3$  Pa, since at lower pressure values, the electrolyte begins to boil and maintaining the discharge in a stationary form is difficult [1–3].

Electrical discharges with liquid (non-metallic) electrodes are a rapidly developing interdisciplinary field of research involving plasma science, heat and mass transfer, fluid dynamics, photolysis, and multiphase chemistry [4]. Such systems include various electrode arrangement configurations and can be divided into four main categories:

- Discharge in a liquid when a metal electrode is immersed in an electrolyte [5];
- Discharge in the gas phase with a metal electrode located above the electrolyte [6];
- Discharges between two liquid (flowing and non-flowing) electrodes [1];
- Discharges in multiphase media, including inside bubbles, vapor-gas mixtures, aerosols and foams [7].

In the electrode configuration where only one electrode is a liquid, the discharge properties are somewhat similar to the abnormal glow and arc discharge. In the variant where both electrodes are liquid, the discharge burns at a relatively high voltage  $U \geq 10^3$  V and a low current density  $j \sim 0.1\text{--}1.0$  A/cm<sup>2</sup>. Changing the composition and concentration of the electrolyte makes it possible to control the combustion mode over a wide range. The duration of the discharge burning, which is usually limited by their erosion when using metal electrodes, is practically unlimited for a discharge with a liquid (non-metallic) electrode. The discharge can be maintained by both direct current and HF and microwave currents [8–12].

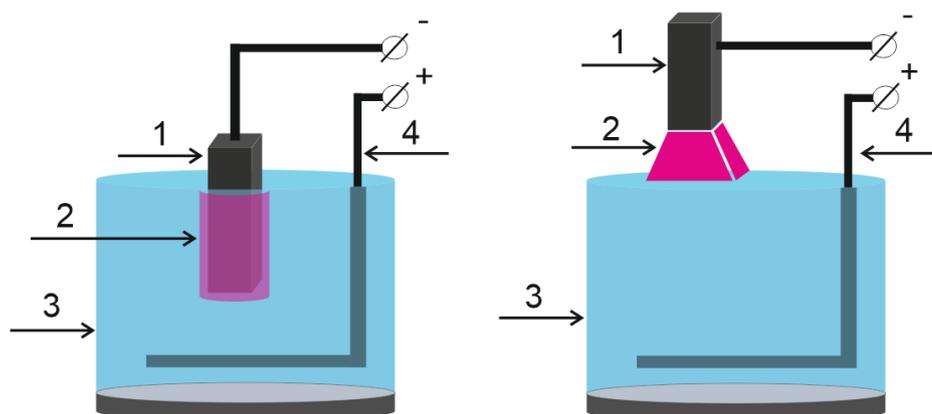
Discharges with a liquid (non-metallic) electrode are widely used to modify materials and products of various physical natures. The technology of electrolytic-plasma treatment of metal products is widely used in various industries. The presence in the radiation of a discharge with a liquid (non-metallic) electrode of spectral lines of elements dissolved in the liquid, the strong nonequilibrium of the plasma generated by the discharge, including when the discharge burns in an atmosphere of high-pressure gases, make it promising also for various technical applications in plasma chemistry (in particular, for cleaning contaminated gas flows), spectral analysis, etc. [13–15].

Despite the fact that discharge with liquid (non-metallic) electrodes has been studied for a long time, there is still no unified classification of plasma-liquid systems, which exists for discharges with traditional electrodes (glow, arc, spark, etc.). It is known that the classification of discharges is based on elementary processes. Therefore, conducting complex experimental and theoretical studies of such discharges, the results of which will form the basis of the knowledge base for the formation of classification parameters in the field of plasma-liquid systems, is an urgent task.

The previous work [16] presents the results of a study of an electric discharge between a metal anode and a liquid (non-metallic) cathode at atmospheric pressure. The types and forms of plasma structures formed in the interelectrode gap, electrophysical parameters are established. Data on the composition of the discharge plasma, electron concentration and heavy component are obtained. The results of numerical calculations of the current flow in an electrolytic cell and the mechanisms of the initial stage of the discharge are presented.

The aim of this work is to study the properties of the discharge when changing the polarity of the electrodes, i.e. when the discharge burns between the metal cathode and the liquid (non-metallic) anode. The results of the work can be used to develop physical and mathematical models of plasma-liquid systems, as well as for engineering methods for calculating plasma installations.

**Experimental setup.** Ignition and study of the electric discharge in the considered configuration of electrodes was carried out on the setup (Fig. 1), where: 1 - metal cathode; 2 - area with discharge; 3 - electrolytic bath; 4 - metal plate made of copper grade M1 for supplying positive potential to the electrolyte. A metal rod made of aluminum grade AMC - 40 was used as a metal cathode, and a 3% NaCl solution in purified tap water was used as a liquid (non-metallic) anode.



**Figure 1.** Functional diagram of a gas-discharge chamber for maintaining a discharge (a) – with a metal cathode immersed in the electrolyte (b) – with a metal cathode located above the surface of the electrolyte

The metal cathode was preliminarily immersed in the electrolyte and moved in the vertical plane at a distance of 10 mm using an automatic manipulator. A thermostat is provided to control the temperature of the electrolyte solution in the bath. Thermostating of the electrolyte was carried out using a circulating refrigeration cooler. The electrolyte in the bath is renewed using the electrolyte supply and pumping system. A coarse filter is provided in the system to clean the solution from impurities. Electrolyte vapors are removed from the discharge study area using a stationary exhaust hood and a fan.

The experimental setup is equipped with a high-voltage generator with a power of 40 kW and a variable voltage of up to 4 kV at a nominal current of up to 10 A, which provides power for the discharge current, diagnostic and auxiliary equipment. The power generator operates by converting and regulating the network voltage. The generator consists of high-voltage and low-voltage adjustable units, which provide the specified ranges of set voltages and currents. The setup is grounded. The current values of current and voltage were shown by pointer indicators on the source control panel, transmitted to the control computer and monitored by the operator.

The experiments were carried out at the set parameters of voltage  $U = 0.05\text{--}1.1$  kV, pressure  $p = 10^5$  Pa, diameter of the metal cathode  $d_k = 7$  mm, interelectrode distance between the metal cathode and electrolytic anode  $i = 5$  mm, immersion depth of the metal electrode in the electrolyte  $h = 3$  mm, specific conductivity of the electrolyte  $\sigma = 0,1\text{--}0,12$  Ohm<sup>-1</sup> cm<sup>-1</sup> and the temperature of the liquid (non-metallic) anode  $T_a = 19\text{--}23$  °C.

The solution to this goal is achieved by using modern diagnostic equipment, methods and research approaches:

1. Video recording of the processes occurring in the discharge combustion zone, as well as the plasma structures formed in the process, was carried out using a high-speed video camera of the Casio EX-F1 brand. Due to the high dynamism of the processes occurring in the discharge combustion zone, the shooting speed was chosen to be 1200 and 600 frames per second. The camera was mounted on a tripod at a distance of 300 mm from the discharge combustion zone, which transmitted the received information to a computer with an operator. The received data were processed on a personal computer with the HX software installed. Link " and "Movavi Video Editor 14 Plus".

2. To analyze the temperature distribution of the studied surface of the metal and electrolytic electrodes during the discharge combustion process, a FLIRA6500SC thermal imaging camera with a detector spatial resolution of 640 x 512 pixels with an operating spectral range of 3.6–4.9 μm was used. The thermal imager ensured recording the electrode surface temperature in the calibrated range of 4–2400 °C. A multiwave pyrometer was used to calibrate the thermal imaging camera. The pyrometer was used because an oxide film and scale may form during discharge combustion, which may lead to errors in the measured temperature. The obtained values were processed on a computer with ALTAIR v5.91.010 software.

3. The study of pulsations, fluctuations of current and discharge voltage was carried out by a set of digital oscilloscopes of various brands "GDS - 806S" and brand "GOS-6030". In order to ensure control of electrophysical parameters at the moment of ignition and maintenance of the discharge, a device for recording optical radiation of the discharge on photodiodes with a microcircuit was connected to the oscilloscopes.

4. Numerical calculations were performed in the MATLAB environment. A system of equations consisting of balance equations for the concentrations of electrons, positive and negative ions, and the Poisson equation for the electric field potential was solved. The system of equations was solved by the finite element method in a two-dimensional formulation with axial symmetry.

**Discussion of results.** The discharge is formed by immersing the metal cathode in the electrolyte to a depth of 3 mm at atmospheric air pressure. When a potential of up to 290 V is applied to the electrodes, the met-

al cathode begins to heat up due to the direct current flowing in the circuit. The process of electron emission from the cathode surface and their movement towards the anode occurs. In this case, the electrolyte around the metal cathode begins to boil and passes into a gaseous state. This process is accompanied by intense evaporation of the electrolyte and the release of convective vapor-gas flows. At the electrode-electrolyte interface, a process characteristic of electrolysis occurs.

With an increase in the applied voltage of more than 300 V, a breakdown is recorded with a discharge burning in the form of microchannels that periodically appear in the vapor-gas shell around the metal cathode. The discharge burns in the form of current pulses  $I = 2.5\text{--}11$  A. The combustion process is accompanied by strong acoustic pops and disturbances of the electrolyte surface with abundant evaporation of the electrolyte, which is due to the periodic contact of the liquid with the surface of the highly heated metal electrode.

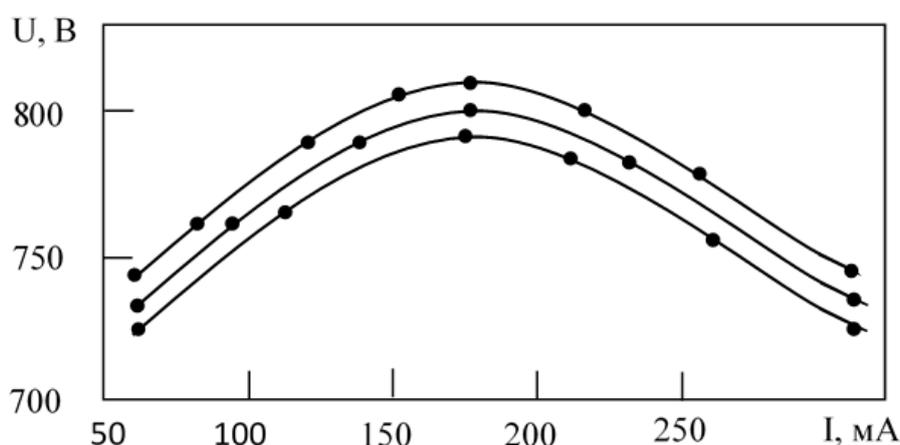
Next, the metal cathode is raised by an automatic manipulator to a distance of 5 mm above the surface of the liquid anode. As a result, the discharge is stabilized in a volumetric (diffuse) form of yellow color. The yellow shade of the discharge is due to the presence of the element Na in the electrolyte solution.



**Figure 2.** Thermogram of the surface of a metal cathode and a liquid (non-metallic) anode under discharge combustion conditions

Based on the analysis of thermography results, it follows that the temperature of the metal cathode in the range of 9–208 px increases and reaches  $T_k = 445$  °C. Then  $T_k$  in the range from 209 to 310 px does not

change (Fig. 2). This is explained by the fact that the electric field strength at the surface of the metal cathode near the discharge has a maximum value. Then  $T_k$  in the range from 311 to 365 px drops to 100 °C, which corresponds to the hot gas zone near the metal cathode. Also in the range of 330–345, temperature jumps up to 200 °C are recorded, which corresponds to anode spots on the surface of the liquid (non-metallic) anode. The interval on the graph corresponding to the interval 365–383 px refers to the liquid anode, the temperature of which changes in the range  $T_a = 89\text{--}99$  °C.



**Figure 3.** Current-voltage characteristic of the discharge between a metal cathode and a liquid (non-metallic) anode.

The volt-ampere characteristic (VAC) of the discharge after the metal cathode is positioned above the surface of the liquid anode is plotted (Fig. 3). Based on the VAC analysis, it follows that in the current range of 75–150 mA the value of  $U$  increases, and with an increase in current from 150 to 300 mA the VAC of the discharge has a decreasing character.

This development of the discharge VAC is explained by the fact that with an increase in current, the nature of the processes of electrons leaving the discharge changes. Instead of diffusion processes, volume processes begin to play a significant role. In this case, the growth of current can only be caused by an increase in the electric field intensity of the gas discharge.

The gas temperature, which increases with increasing current, leads to its rarefaction in the axial region and, accordingly, to an increase in the local value of the reduced electric field strength  $E/N$ . In connection with this, the gas ionization frequency increases sharply.

The characteristic lifetime of a free electron in a discharge depends on the gas temperature less strongly. Therefore, to maintain a stationary con-

centration of electrons in the positive column, the electric field strength of the gas discharge must decrease. Thus, heating of the gas in the vapor-gas gap between the electrodes leads to a falling current-voltage characteristic.

The model of discharge between a pin cathode and an electrolytic anode is considered. The equations of balance of concentrations of electrons and ions are considered taking into account the plasma-chemical transformations occurring between them. As sources of the electronic component the processes of ionization and detachment are considered, and as its sinks - the attachment of electrons to neutrals and electron-ion recombination. The same processes are responsible for the evolution of the concentrations of positive and negative ions. Taking into account also the processes of diffusion and drift of charged particles, we arrive at the following system of drift-diffusion equations describing the evolution of the concentrations of charge carriers in the discharge region

$$\frac{\partial n_e}{\partial t} = (\alpha_i - \alpha_a)n_e + \alpha_d n_n - \beta_{ei} n_e n_p - \operatorname{div} \mathbf{J}_e + q, \quad (1)$$

$$\frac{\partial n_p}{\partial t} = \alpha_i n_e - \beta_{ep} n_e n_p - \beta_{np} n_n n_p - \operatorname{div} \mathbf{J}_p + q, \quad (2)$$

$$\frac{\partial n_n}{\partial t} = \alpha_d n_e - \alpha_d n_n - \beta_{np} n_n n_p - \operatorname{div} \mathbf{J}_n, \quad (3)$$

In (1)–(3)  $n_e$ ,  $n_p$ ,  $n_n$  and  $\mu_e$ ,  $\mu_p$ ,  $\mu_n$  are the concentrations of electrons, positive and negative ions, respectively;  $\alpha_i$ ,  $\alpha_a$ ,  $\alpha_d$  are the frequencies of ionization, attachment and detachment of electrons;  $\beta_{ei}$ ,  $\beta_{np}$  are the coefficients of electron-ion and ion-ion recombination;  $q$  is the frequency of external ionization.

The particle flux densities are given by the equations

$$\mathbf{J}_s = n_s \mathbf{v}_s - D_s \operatorname{grad} n_s, \quad (4)$$

where the index  $s$  takes the values  $e$ ,  $p$  and  $n$ ;  $D_e$ ,  $D_p$ ,  $D_n$  are the diffusion coefficients. Drift velocities of particles

$$\mathbf{v}_e(\mathbf{r}, t) = -\mu_e \mathbf{E}(\mathbf{r}, t), \quad (5)$$

$$\mathbf{v}_n(\mathbf{r}, t) = -\mu_n \mathbf{E}(\mathbf{r}, t), \quad (6)$$

$$\mathbf{v}_p(\mathbf{r}, t) = \mu_p \mathbf{E}(\mathbf{r}, t), \quad (7)$$

$\mu_e$ ,  $\mu_p$ ,  $\mu_n$  — mobilities of electrons, positive and negative ions, respectively;  $\mathbf{E}$  — electric field strength.

The Poisson equation is used to calculate the distribution of the electric field potential.

$$\Delta\varphi = -\frac{e}{\varepsilon_0}(n_p - n_e - n_n). \quad (8)$$

Electric field strength vector

$$\mathbf{E}(\mathbf{r}, t) = -\text{grad}\varphi(\mathbf{r}, t), \quad (9)$$

where  $\varphi$  is the electric field potential;  $\alpha_i$ ,  $\alpha_a$ ,  $\alpha_d$  are the frequencies of ionization, attachment and detachment of electrons;  $\beta_{ei}$ ,  $\beta_{np}$  are the coefficients of electron-ion and ion-ion recombination;  $p$ ,  $T$ ,  $\rho$ ,  $N$  are the pressure, temperature, mass and volume densities of the gas.

The condition of cylindrical symmetry relative to the  $z$  axis is adopted. Calculations were made for pressures  $p = 100$  kPa under the following boundary conditions. On a metal cathode

$$\varphi|_K = -V, \quad J_e|_K = \gamma J_p|_K, \quad \left. \frac{\partial n_e}{\partial \mathbf{n}} \right|_K = 0, \quad \left. \frac{\partial n_p}{\partial \mathbf{n}} \right|_K = 0, \quad n_n|_K = 0.$$

Here  $\gamma$  is the coefficient of secondary electron emission,  $\mathbf{n}$  is the normal vector to the boundary.

At the plasma-dielectric boundary, the condition of continuity of current density is applied, at the rest of the boundary

$$n_e = n_p = n_n = 0, \quad \left. \frac{\partial \varphi}{\partial \mathbf{n}} \right| = 0.$$

Condition of axial symmetry

$$\left. \frac{\partial n_e}{\partial r} \right| = \left. \frac{\partial n_p}{\partial r} \right| = \left. \frac{\partial n_n}{\partial r} \right| = 0, \quad \left. \frac{\partial \varphi}{\partial r} \right| = 0 \text{ at } r = 0.$$

The electrolyte was considered as a homogeneous conducting medium with a given electrical conductivity  $\sigma$ .

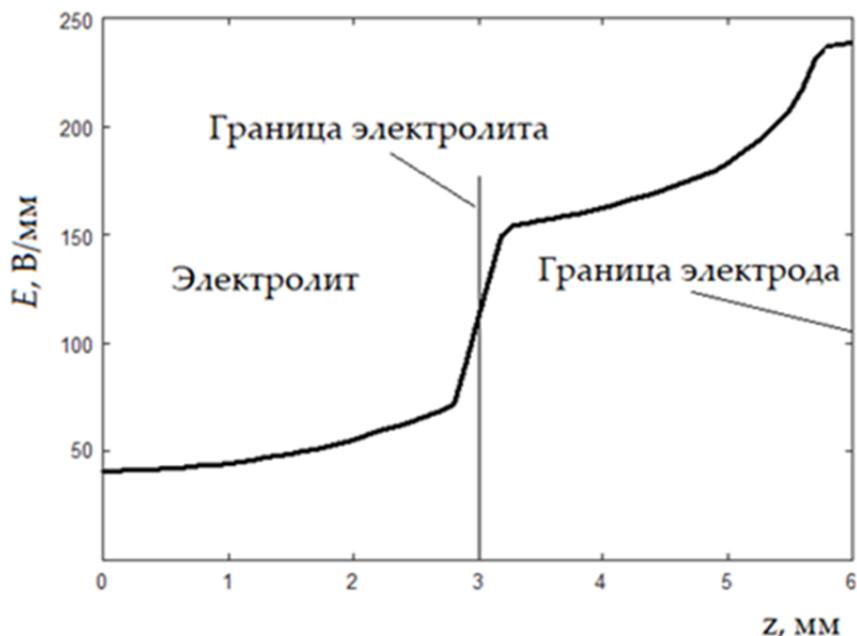
Ionization coefficient  $\alpha_i = \mu_e E \alpha_T$ . The dependence of the Townsend ionization coefficient  $\alpha_T$  on  $E/p$  for air is taken as

$$\frac{\alpha_T}{p} = A \exp\left(-\frac{B}{E/p}\right),$$

where  $A = 15 \text{ cm}^{-1} \text{ Torr}^{-1}$ ,  $B = 365 \text{ V cm}^{-1} \text{ Torr}^{-1}$ .

Calculations using the equations were carried out until stationary values of the concentrations of charged particles were obtained.

Fig. 4 shows the change in the electric field strength along the discharge axis with a voltage applied to the electrode of  $U = 750$  V. The distance between the electrode and the electrolyte surface was taken to be 3 mm.



**Figure 4.** Change in electric field strength along the discharge axis ( $r = 0$ )

The electric field is highly non-uniform near the electrode. The distance on the graph from 0 to 3 mm corresponds to the electrolyte, where the electric field strength smoothly changes from 40 to 60 V/mm, this is due to the fact that the electrolyte is a homogeneous conducting medium.

At the boundary between the electrolyte and the discharge in the range from 2.8 mm to 3.2 mm, the electric field strength increases sharply, which is explained by the difference in electrical conductivity of the electrolyte and plasma in the region adjacent to the electrolyte.

The range from 3.2 mm to 6 mm corresponds to the region of gas discharge plasma, where the electric field strength smoothly increases from 150 to 240 V/mm, as a result of the non-uniformity of the electric field created by the pin cathode.

In the near-cathode region at a distance of 5.8 to 6 mm, the electric field strength has a practically constant value, since in the small near-cathode region along the central line the electric field will be approximately uniform.

Fig. 5 shows the change in intensity along the lower edge of the electrode. As can be seen from the figure, the electric field intensity increases at the edges of the metal electrode to 275 V/mm, due to the strong non-uniformity of the electric field.

The electron concentration changes along the discharge axis from  $8 \cdot 10^{13} \text{ mm}^{-3}$  near the metal cathode to  $\sim 10^{13} \text{ mm}^{-3}$  near the electrolytic anode (Fig. 6). Immediately near the cathode, electrons are formed due to

the bombardment of the surface by ions, then the electrons are accelerated in the electric field and ionization of the vapor-gas gap occurs, which leads to an increase in the electron concentration. Then the electron concentration drops due to a decrease in the electrolytic field strength in the direction of the electrolyte (Fig. 4).

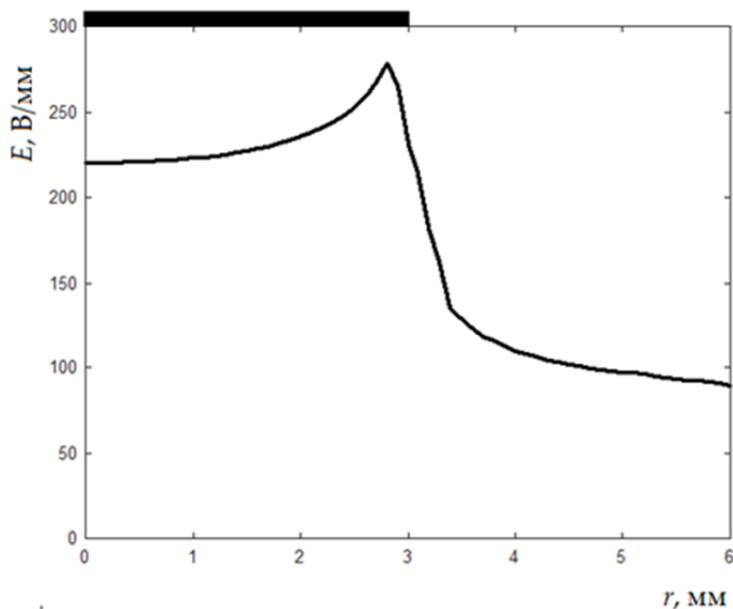


Figure 5. Change in electric field strength along the lower edge of the electrode

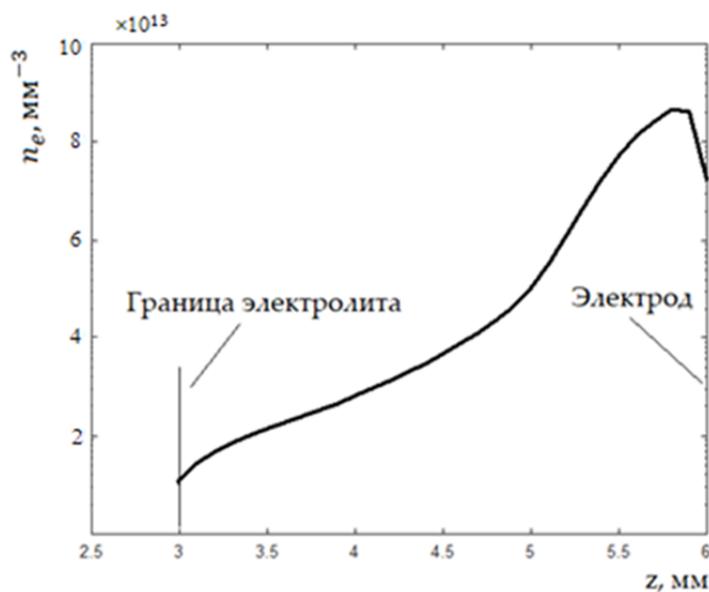
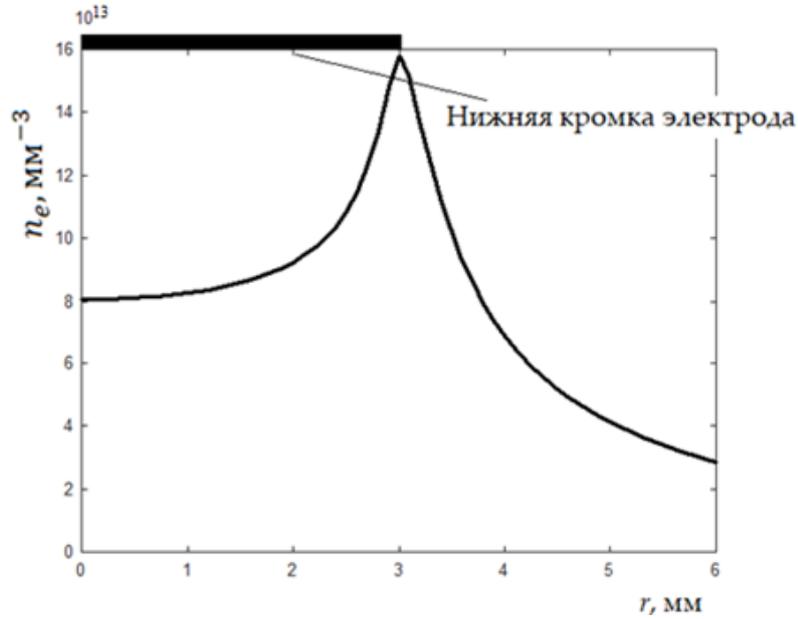
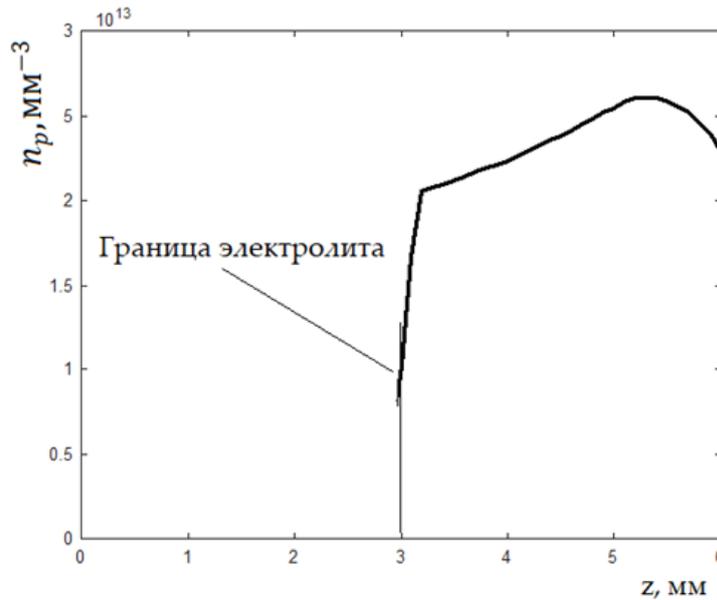


Figure 6. Change in electron concentration along the discharge axis ( $r = 0$ )

The electron concentration is maximum near the edge of the electrode, where the electric field strength is greatest and reaches a value of  $16 \cdot 10^{13} \text{ mm}^{-3}$  (Fig. 7).



**Figure 7.** Change in electron concentration along the lower edge of the electrode. The electrode radius is 3 mm.



**Figure 8.** Change in the concentration of positive ions along the discharge axis

Fig. 8 shows the distribution of the concentration of positive ions along the discharge axis. Calculations have shown that in the main discharge region the quasi-neutrality condition is approximately satisfied  $n_p - n_e - n_n \approx 0$ . Near the cathode surface the concentration of positive ions increases due to the ionization of the vapor-gas mixture by electron impact. Then the concentration of positive ions decreases in accordance with the fact that the concentration of electrons in the direction of the electrolyte decreases and the ionization processes are reduced.

**Conclusion.** 1. The combustion of an electric discharge in the form of microchannels in a vapor-gas mixture around a metal cathode at a voltage of 300 V was established. The discharge burns in the form of current pulses  $I = 2.5\text{--}11$  A. When the metal cathode rises above the surface of the liquid (non-metallic) anode, the discharge stabilizes in a volumetric (diffuse) form of yellow color. The yellow tint of the discharge is due to the presence of the element Na in the electrolyte solution.

2. The volt-ampere characteristic (VAC) of the discharge after the metal cathode is placed above the surface of the liquid anode is plotted. In the current range from 75 to 150 mA, the value of  $U$  increases, and with an increase in current from 150 to 300 mA, the VAC of the discharge has a decreasing character. This development of the VAC of the discharge is explained by the fact that with an increase in current, the process of electron loss in the positive plasma column changes. Instead of diffusion processes, volume processes begin to play a significant role.

3. It was found that the electric field strength increases at the edges of the metal electrode to 275 V/mm, due to the strong non-uniformity of the electric field. The electron concentration changes along the discharge axis from a value of  $8 \cdot 10^{13} \text{ mm}^{-3}$  near the metal cathode to  $\sim 10^{13} \text{ mm}^{-3}$  near the electrolytic anode.

4. The distribution of the concentration of positive ions along the discharge axis is given. Calculations have shown that in the main region of the discharge the quasi-neutrality condition is approximately satisfied  $n_p - n_e - n_n \approx 0$ .

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