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TEMPERATURE DYNAMICS OF THE MICRODROPLET FRACTION OF METAL PLASMA IN PLASMA-OPTICAL DEVICES WITH FAST ELECTRONS

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The computer simulation of a plasma filter with fast electrons shows that the main source of droplet heating is thermal electrons, while the role of fast electrons is reduced mainly to transferring energy to slow electrons. The processes of drop charging in the presence of an electron beam and the effective emission of electrons due are also considered.

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INTRODUCTION

The growing demand for nanoscale manufacturing in modern industries like integrated circuit production and surface layer modification necessitates efficient and reliable means of production. MEVVA-type ion plasma sources, utilizing vacuum-arc discharge, have proven effective for creating high-current heavy metal ion beams [1], which are widely used in applying protective and functional coatings and modifying surface properties. A significant limitation, however, is the presence of cathode material microdroplets in the ion-vapor flow, which hinder the creation of high-quality, nano-level uniform coatings.

Current filters and elimination methods for these microdroplets are based on various forms of selection, primarily mechanical and electrophysical filters [2]. While they can effectively remove microdroplets larger than 1 μm, using these filters for smaller droplets reduces the density of the metal plasma on the products considerably. This limitation restricts the full utilization of the high generation rate of the ion vapor flow of the erosion plasma source.

The Institute of Physics of the National Academy of Sciences of Ukraine has been developing axisymmetric cylindrical plasma-dynamic systems grounded on the principles of mean energy plasma optics [3]. This innovation opens up new possibilities for controlling a low-energy ion-plasma beam in MEVVA-type sources. A new approach using plasma-dynamic systems like electrostatic plasma lens (PL) and hollow cathode discharge systems (PC) has been proposed and investigated to eliminate microdroplets from dense metal plasma flow [4, 5]. These systems generate an energetic electron beam that can effectively vaporize and remove microdroplets.

Nonetheless, to fully harness the potential of these systems, there's an urgent need to understand the fundamental physical mechanisms affecting micro-inclusions in dense dust plasma during their passage through plasma-dynamic systems with fast electrons.

1. EQUAITONS OF DROP EVAPORATION

A device with a hollow cathode was taken as a prototype for our model of droplet evaporation. A detailed description of the setup and its parameters can be found in [6].

The rate of evaporation of a drop is determined by its temperature, therefore, to determine how quickly the evaporation process will occur, it is necessary to know the temperature of the drop, which changes during the stay of the droplet in the discharge. On the other hand, the temperature of the droplet is determined from the balance of energy absorbed and lost by the droplet. As our preliminary calculations show, one of the most important channels of energy loss in a droplet is precisely the evaporation process. Thus, to find the change in temperature and mass of a drop during its stay in a discharge, it is necessary to solve a system of two mutually related differential equations: the energy balance equation and the evaporation rate equation.

The general energy balance equation for a drop can be written as:

$$cm_{dr}\frac{dT}{dt} = A_{dr}\left(\sum_i f_i^{in} - \sum_i f_i^{out}\right), \qquad (1)$$

where c is the specific heat capacity of the drop material, m_{dr}, A_{dr}, and T_{dr} are the mass, surface area, and temperature of the drop, and f_i^{in} and f_i^{out} are the energy flow densities to and from the drop, respectively. In this equation, all three parameters characterizing the drop depend on time t, and are closely connected to each other. We will consider the relationship between mass and temperature later, and the surface area of a drop is determined by its radius r_{dr}, which, in turn, is determined by the mass of the drop $4/3\ \pi\ r_{dr}^3\ \rho_{dr} = m_{dr}$. Here ρ_{dr} is the density of the drop, which is the as the heat capacity, is considered constant in our approximation.

From our previous studies, it was established that the main sources of droplet heating in the discharge are the flow of fast and slow (“thermal”) electrons and ions on the droplets. On the other hand, the main processes through which drops lose energy are droplet evaporation, thermal radiation from the droplet surface, and thermoelectron emission.

All droplet heating sources are related to the flow of charged particles per droplet, so to find their values we need to know the current densities of electrons and ions per droplet. Since with the plasma parameters we use in the calculations (plasma density $n_0 = 5\cdot10^{16}$ m^{-3}, electron temperature T_e = 5…20 eV), the Debye length λ_D is much greater than droplet radius r_{dr}, we can use the theory of L.O.M. (Low Orbital Motion). According to this theory [7], the current density on a spherical body of repulsive particles, which in our case are electrons, is given by the formula

$$j_e = -q\,n_0 \sqrt{\frac{k_b\,T_e}{2\pi\,m_i}}\exp\left(-\frac{q\,\varphi_{dr}}{k_b\,T_e}\right), \tag{2}$$

where k_b is the Boltzmann constant, q is the elementary charge, m_e is the electron mass, and φ_{dr} is the drop potential. The current density of ions, which in our case are particles attracted to the drop, according to L.O.M. is

$$j_i = q\,n_0 \sqrt{\frac{k_b\,T_i}{2\pi\,m_i}}\left(1-\frac{q\,\varphi_{dr}}{k_b\,T_i}\right), \tag{3}$$

where m_i and T_i are the ion mass and temperature, respectively.

As for fast electrons, due to the fact that their energy is much greater than the absolute value of the drop potential, in the first approximation it can be assumed that their current density per drop is equal to the current density in the discharge volume j_b.

To obtain the energy flow density, we simply replace the particle charge in the previous expressions with the average energy carried by this particle. Thus, for the energy flux density of slow electrons f_e, ions f_i, and fast electrons f_b, we have:

$$f_e = \tfrac{3}{2} k_b\,T_e\,n_0 \sqrt{\frac{k_b\,T_e}{2\pi\,m_e}}\exp\left(-\frac{q\,\varphi_{dr}}{k_b\,T_e}\right), \tag{4}$$

$$f_i = \tfrac{3}{2} k_b\,T_i\,n_0 \sqrt{\frac{k_b\,T_i}{2\pi\,m_i}}\left(1-\frac{q\,\varphi_{dr}}{k_b\,T_i}\right), \tag{5}$$

$$f_b = \left(\frac{\varepsilon_b}{q}-\varphi_{dr}\right) j_b\,. \tag{6}$$

Now consider the energy flows from the drop. As already mentioned, this is the evaporation of the droplet material, f_{ev}, the thermal radiation f_{rad}, and the energy carried by the thermal emission electrons, f_{th}. Evaporation energy losses are calculated based on the evaporation rate of the drop $\frac{d\,m_{dr}}{d\,t}$, and can be written as:

$$f_{ev} = \frac{1}{A_{dr}}\,r_{ev}\,\frac{d\,m_{dr}}{d\,t}, \tag{7}$$

where r_{ev} is the specific heat of vaporization of the drop material. It should be noted that the expressions for all energy flows are given without taking into account the direction of one or another flow. That is, all values have a positive value, and their influence on the total energy of the droplet is taken into account by the sign in equation (1).

The flow of radiation energy from a droplet is calculated by the Stefan-Boltzmann formula:

$$f_{rad} = \alpha\,\sigma\,T_{dr}^4, \tag{8}$$

where α is the emissivity of the drop material, and σ is the Stefan-Boltzmann's constant.

The last energy flow from the drop, which we take into account in our calculations, is determined by the flow of thermoemission electrons from the surface of the drop. The current density of such electrons is determined by the formula

$$j_{th} = A_0\,T_{dr}^2 \exp\left(-\frac{w}{k_b\,T_{dr}}\right), \tag{9}$$

where A_0 is the universal thermoemission constant, and w is the work of electron release from the droplet material. Since each electron flying out of the surface of the droplet spends energy w, the total density of the energy flow carried by thermoelectrons is

$$f_{th} = w\frac{A_0}{q}T_{dr}^2 \exp\left(-\frac{w}{k_b\,T_{dr}}\right). \tag{10}$$

Let us now consider the equation for the evaporation of a drop. Since the concentration of metal atoms in the discharge volume is negligible, we can use the Hertz-Knudsen equation [8] for evaporation into a vacuum:

$$\frac{d\,m_{dr}}{d\,t} = -A_{dr}\sqrt{\frac{\mu}{2\pi\,k_b\,T_{dr}}}P_s(T_{dr}), \tag{11}$$

where μ is the mass of evaporating molecules or atoms, and $P_s(T)$ is the saturated vapor pressure. The pressure of saturated vapors depends not only on the temperature but also on the radius of curvature of the surface over which they are formed, but in the first approximation, we can ignore such discrepancies and use the experimental pressure data obtained for a flat surface.

One of the main parameters included in the formulas that determine energy flows to/from the drop is the value of the potential of the drop. This value can vary over a wide range, depending on the discharge conditions, and greatly influences energy flows. Therefore, it is advisable to obtain an agreed value of this parameter from the calculations. To do this, let's write another equation, which is determined by zero current per drop equality. The value of all these currents was found by us when finding energy flows. It is only necessary to take into account that fast electrons have sufficient energy to form secondary electron emission from the droplet, and thus their contribution to the total charge will be less by the amount γj_b, where γ is the coefficient of secondary emission. Thus, we have a mixed system of two differential and one algebraic equation:

$$\begin{cases} cm_{dr}\dfrac{dT_{dr}}{dt} = -r_{ev}\dfrac{dm_{dr}}{dt} + A_{dr}(-f_{rad} - f_{th} + f_e + f_b + f_i)\,, \\ \dfrac{dm_{dr}}{dt} = -A_{dr}\sqrt{\dfrac{\mu}{2\pi\,k_b\,T_{dr}}}P_s(T_{dr}), \\ j_i - (1-\gamma)\,j_b - j_e + j_{th} = 0\,, \end{cases} \tag{12}$$

where individual terms are determined by formulas (2)-(10).

The system of equations (12) is an explicit differential-algebraic system with three dependent variables m_{dr}, T_{dr}, and φ_{dr}. A distinctive feature of the system is its stiffness. That is, this system is poorly solved by traditional methods such as the Runge-Kutta method. Therefore, for the numerical solution of the system (12), an algorithm with a variable step and a variable order based on the inverse differentiation formulas of the first to the fifth order was chosen. The Jacobi matrix, which the algorithm needs for its work, was calculated by numerical differentiation. As research [9] shows, this algorithm effectively finds the solution of stiffness systems of equations and has good accuracy.

As initial conditions for solving the system of equations, the melting temperature of the metal of the droplet, the mass corresponding to the applied radius of the droplet, and the negative potential of the droplet whose absolute value was equal to the temperature of the plasma electrons were taken as initial conditions.

2. EVAPORATION OF A COPPER DROP

Fig. 1 displays the change in temperature of a 1 μm copper droplet over time spent in the discharge, considering various values of electron temperature. It is apparent that as time passes, the droplet's temperature eventually stabilizes. As electron temperature increases, so does the droplet's temperature. The time it takes for the droplet to reach a stable temperature decreases significantly with an increase in electron temperature.

Fig. 2 presents the change in droplet temperature over time for different values of another crucial discharge parameter, the current density of fast electrons. An increase in this parameter results in a greater flow of fast electrons per droplet, leading to more rapid and substantial droplet heating.

A comparison of these figures indicates that the electron temperature has a much stronger impact on droplet temperature than the current of fast electrons, meaning "thermal" electrons transfer more energy to the droplets than fast ones. This conclusion is corroborated by Fig. 3, which shows calculated energy flow densities per droplet from the three mentioned sources and the main channels of energy loss by the droplet for three sets of plasma parameters.

It's evident from Fig. 3 that an increase in plasma electron temperature leads to a concurrent rise in the heating contributions of slow electrons and ions to the droplet. At the same time, the heating power of fast electrons remains independent of this temperature, increasing only with the growth of fast electron current density. Conversely, an increase in the current of fast electrons results in only minor changes in the energy flow densities of slow electrons and ions. Worth noting is that these changes have opposite effects – increasing the energy of thermal electrons while decreasing the energy of ions. These shifts are attributable to the fact that as the droplet's temperature increases, its electrical potential decreases, consequently affecting the flow of electrons and ions.

3. EVAPORATION OF A TITANIUM DROP

Titanium distinguishes itself from copper in several parameters. Foremost, titanium has a significantly lower saturated vapor pressure value than copper at the same temperature, implying that to achieve considerable evaporation of titanium, it must be heated to higher temperatures.

The second distinguishing parameter for titanium in our model is the work of electron extraction from the metal. Titanium has a significantly lower value for this parameter, causing the effect of thermal emission on the temperature and potential of a titanium droplet to occur at lower energy flow values. Fig. 4, which is analogous to Fig. 1, illustrates these discrepancies.

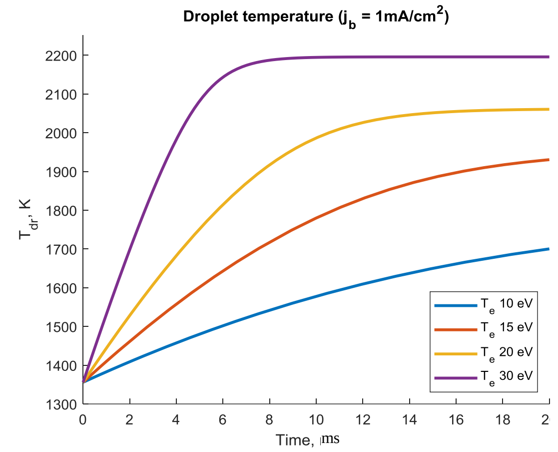

Fig. 1. Dependence of the temperature of a copper droplet with a diameter of 1 μm on the time it is in the discharge, for several values of the electron temperature

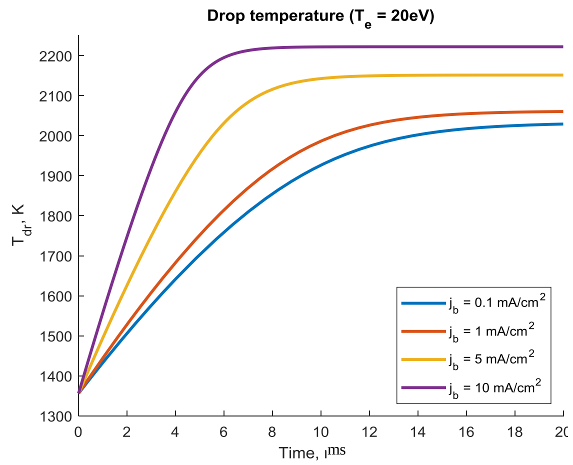

Fig. 2. Dependence of the temperature of a copper drop with a diameter of 1 μm on the time of its stay in the discharge, for several values of the current density of fast electrons

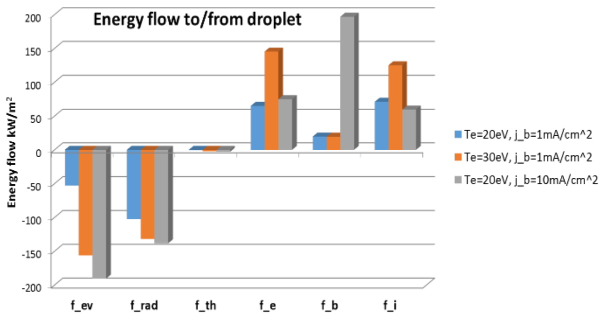

Fig. 3. Density ratio of energy fluxes to/from the drop, for some sets of plasma parameters. A drop of copper with a radius of 1 μm

From these figures, it's clear that a titanium droplet's temperature rises faster with an increasing power applied to the discharge than that of a copper droplet. Moreover, the primary cooling mechanism for the droplet is radiation. At temperatures below 2400 K, evaporation makes a minimal contribution to the droplet's energy loss. The share of thermal electron emission was also minor, but its contribution increased slightly compared to copper. However, as mentioned above, thermal emission primarily affects the droplet's potential value.

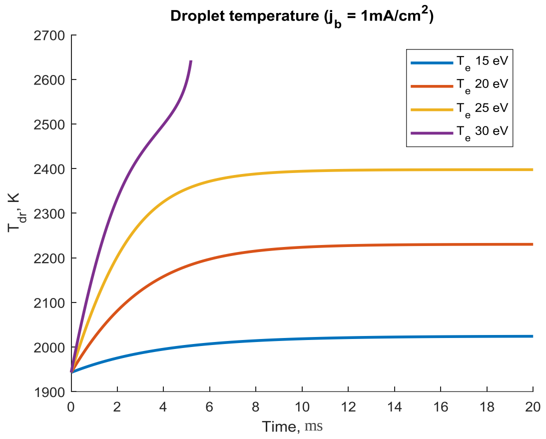

Fig. 4. Dependence of the drop temperature of titanium with a diameter of 1 μm on the time it is in the discharge, for several values of the electron temperature

As titanium droplet power increases, we observe a phenomenon not seen in our copper calculations. When the droplet reaches a certain critical temperature, there is a sudden further increase in its temperature, evident in Fig. 5 for an electron temperature T_e = 30 eV. Furthermore, the droplet's temperature rises to levels where our model stops functioning, i.e., the system of equations (12) no longer has solutions for such parameters. This phenomenon is tied to the fact that once the droplet reaches a certain temperature, the thermal emission current from the droplet begins to increase significantly. This increase in turn leads to a rise in the droplet's potential, and thus an increase in the energy flow density of plasma electrons.

Detailed examination of Fig. 5 shows the jump in temperature occurs when the droplet reaches a temperature of about 2483 K. It's possible to calculate the droplet's energy loss at this temperature. Calculations show the droplet loses approximately 20.8 kW per square meter of its surface to evaporation, 215.5 kW/m^2 to radiation, and 16.2 kW/m^2 to thermal emission. Therefore, the total energy loss of a droplet at the critical temperature T_{cr} = 2483 K is 252.5 kW/m^2.

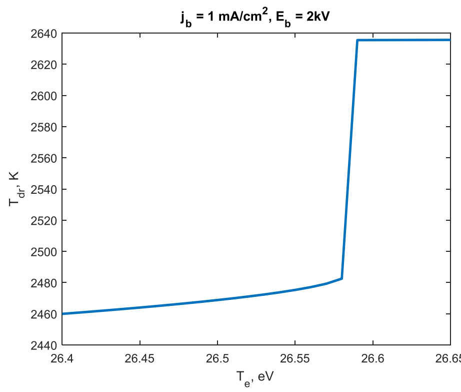

Fig. 5. Dependence of the temperature on the plasma temperature

Since slow plasma electrons are not the only heating source for the droplet, achieving this “explosive” droplet heating effect through other energy sources is feasible. Figs. 6 and 7 present calculations of droplet temperature and potential using fast electrons of varying energies. To minimize the energy supply to the droplet from other sources, particularly slow electrons, a low plasma temperature is required. However, the plasma temperature should not be too low that an increase in the flow of slow electrons caused by the increase in the droplet's potential fails to bring sufficient energy to compensate for its loss due to the increase in thermal emission current. In our calculations, we used a plasma temperature value of 5 eV, which is significantly lower than values measured in real devices.

Figs. 6 and 7 validate our assumption. As the energy of fast electrons increases, and hence the energy density they transmit to the droplet, both the droplet's temperature and potential increase. And at a fast electron energy of about 25 kV (which corresponds to an energy density per drop of 25 kW/m^2 at a fast electron current density of 1 mA/cm^2), the droplet's potential becomes zero, and our system loses its solution. The slightly higher droplet temperatures in this case are due to the fact that at low plasma temperatures, the droplet has a smaller (by absolute value) potential, therefore, the current of slow electrons is greater. As a result, the droplet must heat up more so that the thermoemission current begins to compensate for this larger current of slow electrons.

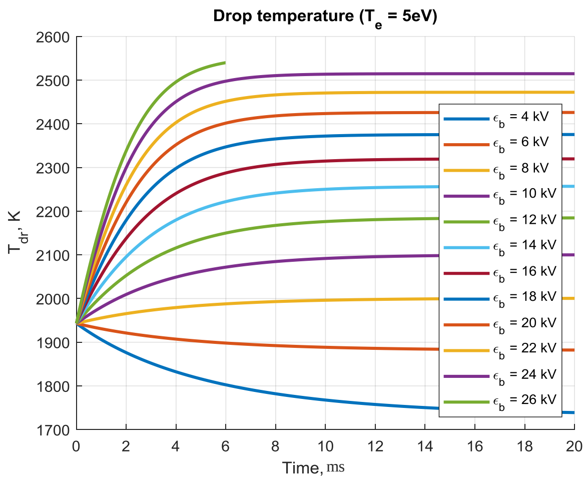

Fig. 6. Dependence of the temperature of a drop of titanium with a diameter of 1 μm on the time it is in the discharge, for several values of the energy of fast electrons

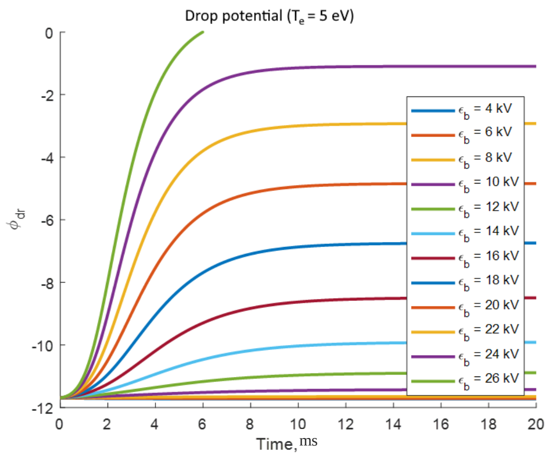

Fig. 7. Dependence of the potential of a drop of titanium with a diameter of 1 μm on the time it is in the discharge, for several values of the energy of fast electrons

4. DROP CHARGING

The fundamental characteristic of micro-droplets is the charge that the particle acquires in the plasma medium, and which depends on the plasma parameters. It is the charge that determines the movement of the droplet in the plasma and the possibility of its destruction and disappearance from the plasma flow [10]. In the absence of an electron beam, the main process of droplet charging is the absorption of electrons and plasma ions by the surface of the microdroplet; due to the high mobility of electrons compared to ions, they reach the droplet surface faster than ions, so the droplet absorbs more electrons than ions. As a result, the drop acquires a negative charge and has a negative floating potential φ_{dr} <0. The dynamics of droplet charging can be determined by the equation:

$$\frac{dQ}{dt} = \sum_n I_n(r_{dr}). \quad (13)$$

In the absence of an electron beam, this sum includes only the current of plasma electrons I_e and ions I_i per drop:

$$I_e(r_{dr}) = 4\pi r_{dr}^2 n_{0e} V_{Te} \exp\left(-\frac{e\varphi_{dr}}{kT_e}\right),$$

$$I_i(r_{dr}) = \pi r_{dr}^2 e n_{0i} V_{Ti}\left(1 - \frac{e\varphi_{dr}}{kT_i}\right) \quad (14)$$

in this case, the secondary emission of electrons from the surface of the droplet can be neglected, since the electron emission caused by plasma electrons and ions is much smaller than the current arriving at the droplet. But if an electron beam is present in the system, it significantly affects the charging of the droplet in the plasma. The electron beam in plasma systems also causes additional emission processes from the surface of the drop. The secondary electron emission is directly caused by the bombardment of the microdroplet surface with an electron beam. Thermoelectronic and autoelectronic emissions are the consequences of the bombardment of the droplet surface with an electron beam due to the increase, respectively, of the temperature and electric field of the droplet. That is, the presence of an electron beam, on the one hand, contributes to a high negative charge of the droplet, and on the other hand, additional emission processes lead to a decrease in the absolute value of the droplet's negative potential, and can even lead to its positive charge [7]. Thus, in the presence of an electron beam, taking into account the secondary electron emission, we have to add current of electron beam I_{eb} and emission current I_{ee}:

$$I_{eb}(r_{dr}) = \pi r_{dr}^2 e n_{eb} v_e\left(1 - \frac{e\varphi_{dr}}{\varepsilon_{eb}}\right),$$

$$I_{ee}(r_{dr}) = \delta I_b(r_{dr}), \quad (15)$$

where δ is the coefficient of secondary electron-electron emission, that has the form [11]:

$$\delta = 7.4\delta_m \frac{\varepsilon_{eb}}{\varepsilon_m} exp\left(-2\sqrt{\frac{\varepsilon_{eb}}{\varepsilon_m}}\right), \quad (16)$$

where δ_m is the maximum value of δ; ε_{eb} is the energy of electron beam; ε_m is the energy of the electron beam, for which $\delta = \delta_m$. Thus, the dynamics of droplet charging can be determined by the equation:

$$\frac{dQ}{dt} = I_e(r_{dr}) + I_i(r_{dr}) + I_{eb}(r_{dr})(1-\delta) = 0. \quad (17)$$

If the charging time can be neglected, this equation can be rewritten in the form:

$$exp\left(-\frac{e\varphi_{dr}}{kT_e}\right) = \sqrt{\frac{m_e/T_e}{M_i/T_i}}\left(1 - \frac{e\varphi_{dr}}{kT_e}\right) - \frac{1}{2}\frac{n_b}{n_0}\sqrt{\frac{\varepsilon_e}{T_e}}\left(1 - \frac{e\varphi_{dr}}{\varepsilon_e}\right)(1-\delta). \quad (18)$$

The numerical solution of this equation depending on the energy of the electron beam for cases when the beam density is comparable to and less than the plasma density is presented in Fig. 8.

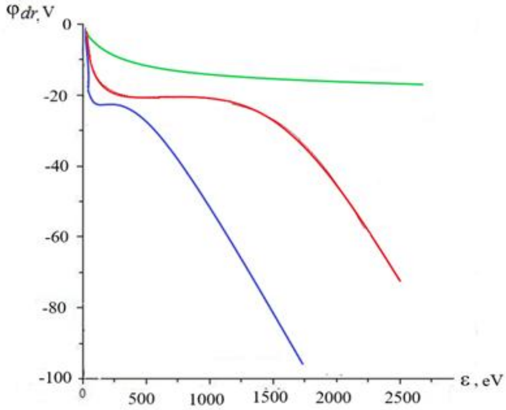

Fig. 8. Dependence of the droplet potential on the energy of the electron beam for different ratios of the electron beam density and plasma density: n_{eb}~n_0 (blue line); n_{eb}=0.1n_0 (red); n_{eb}=0.01n_0 (green)

It can be seen that if the plasma density is significantly higher than the density of the electron beam, the potential of the drop has a weak dependence on the energy of the electron beam. The potential of the drop gradually decreases to -20 V at an electron beam energy of -3 kV. In the case when the density of the beam becomes comparable to the density of the plasma, the potential of the drop drops rapidly, and it acquires a high negative potential when the energy of the electron beam increases.

If we know the potential of the drop, we can also find the charge of the drop depending on the energy of the electron beam. The relationship between the floating potential of a drop and its charge, as shown in [7] can be written as:

$$Q_{dr} = \pi\varepsilon_0\varphi_{dr} r_{dr}\left(1 + \frac{r_{dr}}{\lambda_D}\right) \quad (19)$$

thus, it is directly proportional to the potential. But, as can be seen from the formula, it is also dependent on the radius of the microdroplet. Fig. 9 shows the dependence of the charge of the droplet on the energy of the electron beam for different radius of the droplet and the beam density relative to the plasma density.

If we now consider the dynamics of the droplet charging in the presence and absence of an electron beam, we will see that in the presence of an electron beam the droplet is charged faster than in its absence. Fig. 10 shows the change in charge of the drop over time, where the time step is $\tau_0 = \frac{\lambda_D}{r_{dr}\omega_{pe}}$.

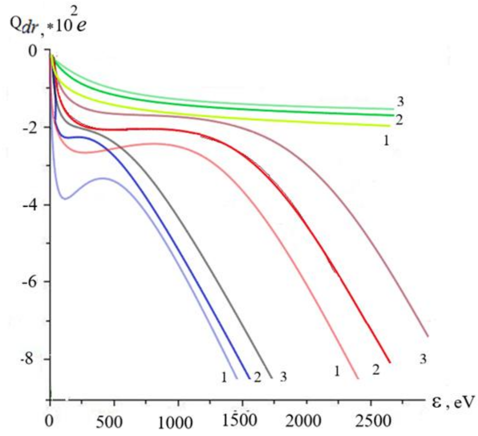

Fig. 9. Dependence of the charge on the droplet on the energy of the electron beam for different radius and density ratios of the electron beam and plasma: n_{eb}~n_0 (blue lines); n_{eb}=0.1n_0 (red); n_{eb}=0.01n_0 (green), 1 – r_{dr}=2 μm; 2 – r_{dr}=1 μm; 3 – r_{dr}=0.5 μm

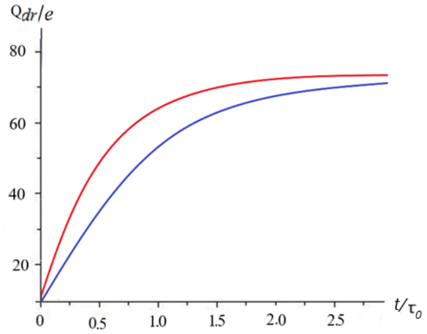

Fig. 10. Droplet charging dynamics in the presence of an electron beam with ε_{eb}=2 keV (red line) and in its absence (blue)

CONCLUSIONS

By solving the system of equations for the balance of energy and current per droplet, it is shown that in plasmodynamic devices with hollow cathodes, there may be favorable conditions for the evaporation of microdroplets. It also follows from these calculations that the main sources of energy on the drop in such discharges are the flow of slow electrons and ions.

It is shown that heating a microdroplet to a certain temperature leads to a rapid increase in the potential of the droplet due to the thermal emission current, which in turn causes its further “explosive” heating due to an increase in the electron current from the plasma to the droplet.

It is shown that in the presence of fast electrons in the system, the drop is charged faster, while the charge that the drop accumulates depends not only on its radius, but also on the ratio of the beam and plasma densities. If the concentration of fast electrons is comparable to the plasma density, the drop is capable of accumulating a large charge, and then the Rayleigh decay mechanism of the drop is possible.

We also note that when the droplet is heated to high temperatures, the kinetic energy of the thermal motion of electrons increases and, at a certain temperature, thermionic emission from the droplet surface begins; therefore, it is necessary to consider a self-consistent solution of the system of equations for the droplet temperature and charge.

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ТЕМПЕРАТУРНА ДИНАМІКА МІКРОКРАПЕЛЬНОЇ ФРАКЦІЇ МЕТАЛЕВОЇ ПЛАЗМИ У ПЛАЗМООПТИЧНИХ ПРИЛАДАХ ЗІ ШВИДКИМИ ЕЛЕКТРОНАМИ

О.А. Гончаров, І.В. Літовко, А.В. Рябцев

Комп’ютерне моделювання плазмового фільтра зі швидкими електронами показує, що основним джерелом нагріву краплі є теплові електрони, тоді як роль швидких електронів зводиться, в основному, до передачі енергії повільним електронам. Розглянуто також процеси заряджання краплі в присутності електронного пучка та ефективну емісію електронів за рахунок цього.