EVAPORATION OF MICRO-DROPLETS IN CATHODE ARC PLASMA COATING UNDER FORMED ELECTRON BEAM

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Evaporation of micro-droplets in an arc plasma flow under the action of a self-consistency electron beam and the condition of direct heating of micro-droplets by fast electrons are considered. It is shown that the plasma is heated under the influence of the beam, even taking into account the fact that the electrons and ions of the plasma lose energy for the evaporation of micro-droplets. It is demonstrated that small micro-droplets evaporate more intensively. It is shown that the plasma electron density should be optimal. For the destruction of macro-particles in a plasma with a higher concentration, more powerful beams are required than in a case of plasma with a lower concentration.

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INTRODUCTION

In many cases, modern industrial technological processes require the transition of production means to work with nanometer scales. Such trends can be seen not only in the production of integrated circuits and devices on a chip but also in cases of application of modifying surface layers for various purposes. Fabrication quality and precision should also be complemented by maximum productivity.

It is known that ion plasma sources of the MEVVA type based on a vacuum-arc discharge are reliable and well-tested generators of ion plasma flow for creating high-current (several amperes) beams of heavy metal ions with particles of medium energy (1...100 keV) [1]. They are widely used in science and industry for deposition of various protective and functional coatings, and also to modify the surface properties of structural and decorative materials. However, the presence of microdroplets of cathode material (size from $\approx 0.01 \,\mu\text{m}$ to tens of µm) in the ion-vapor flow of erosion plasma sources restricted their use for creating high-quality coatings, especially with uniformity at the nano level. Existing filters and methods for eliminating the micro-droplet phase from the working flow are based on various methods of separation (removal) of the droplet phase from the ion-vapor flow [2]. Various mechanical and electrophysical filters are usually used to reduce the concentration of droplets. These filters effectively remove micro-droplets larger than 1 µm without significant loss of ion-vapor flow particles. At the same time, the use of existing filters to remove micro-droplets with smaller sizes leads to a significant (several times) decrease in the density of the metal plasma on the processed products. That is, modern filtration methods limit it the effective use of the high rate of ion-vapor flow generation inherent to an erosive plasma source. The Institute of Physics of the NAS of Ukraine has many years of experience in proposing and implementing ideas

for the creation of axially symmetrical cylindrical plasma-dynamic systems based on the fundamental principles of medium-energy plasma optics. Such systems are a well-developed means of focusing and manipulating ion beams in cases where the problem of compensation of the space charge of the beam is important [3]. The use of these systems in MEVVA-type sources creates new opportunities for predetermined manipulation of a low-energy ion-plasma beam propagating in the direction of the substrate (in the case of sputtering) or to the emission grid (in the case of a plasma source).

In previous works [4-7], a new approach to the elimination of micro-droplets from a dense flow of metal plasma using plasma-dynamic systems such as an electrostatic plasma lens (PL) and systems based on a discharge with a hollow cathode (HC) was proposed and investigated. These systems generate a beam of energetic electrons, which is formed in a self-consistent manner due to secondary ion-electron emission from the inner surface of the central electrode of the lens in the wall layer (in the case of PL) or from the surface of the cathode itself (in the case of HC). Preliminary evaluations and experiments have shown that this electron beam can provide effective evaporation and elimination of micro-droplets. However, for the most effective application of the proposed systems, an actual task is to determine the fundamental physical mechanisms of influence on the state of microinclusions in a dense dusty plasma during its passing through plasma-dynamic systems with fast electrons.

The article is devoted to the determination of the fundamental mechanisms of the action of fast electrons on the flow of a dense metallic dusty plasma.

1. EQUATIONS OF THE EVAPORATION OF MICRO-DROPLETS IN A PLASMA FLOW

Micro-droplets worsen the properties of the films that are formed. But the simple mechanical separation of

micro-droplets from the flow dramatically reduces the rate of film growth. Therefore, it is necessary to evaporate the micro-drops. Micro-droplets fly in an expanding cone [6] (Fig. 1). The flow of micro-droplets passes through the diaphragm, on which part of the micro-droplets remains. After the diaphragm, the flow spreads into the plasma-optical system. Therefore, for the evaporation of micro-droplets, it is enough to influence the near-surface hollow cylindrical flow layer of a certain thickness. We consider a plasma-optical system [4-8], in which, for the evaporation of micro-droplets, additional energy is pumped by a self-consistent electron beam. The electron beam is formed as a result of secondary ionelectron emission during the bombardment of the inner surface of the cylinder by peripheral plasma ions [4-8]. This group of high-energy electrons (electron beam) is accelerated by the applied potential difference approximately along the radius in the direction of the axis of the system.

It is possible to formulate a system of two equations that describe the evaporation of micro-droplets in the plasma flow under the action of a self-consistent electron beam. So, we obtain:

$$\frac{dT_{dr}}{dt} = \frac{3}{r_{dr}\rho_{dr}c} \left\{ kn_n V_{nth} (T_n - T_{dr}) - \alpha \sigma T_{dr}^4 + n_0 kT_e \sqrt{\frac{2kT_e}{\pi m_e}} \exp\left(\frac{-e\varphi_{dr}}{kT_e}\right) + 0.15n_0 \sqrt{\frac{kT_e}{m_i}} \left[4\sqrt{2}(1 + r_d^2/r_{dr}^2) + \gamma e\varphi_0\right] \right\},$$
(1)

$$\frac{dT_e}{dt} = 0.6 \,\gamma \,e \,\varphi_0 \sqrt{\frac{k \,T_e}{m_i}} \left(\frac{2}{R} - n_{dr} \,\pi \,r_{dr}^2\right). \tag{2}$$

The first equation is for the micro-droplet temperature T_{dr} . The second equation is for the plasma electron temperature T_{e} .

Here φ_0 – wall jump of electric potential; φ_{dr} – electric potential of a micro-drop which is equals $(T_e/e) \ln[0.6(2m_e/m_i)^{1/2}])$; $\varepsilon_i = e \varphi_{dr}$ is the energy of ions bombarding a micro-drople; c – heat capacity of the micro-droplet substance; m_{dr} , r_{dr} – mass and radius of micro-droplet; r_d – Debye radius; k – Boltzmann's constant; T_{dr} – temperature of micro-droplet; T_n – temperature of neutral particles; n_n , V_{nth} – density and thermal velocity of neutral particles; $j_b = n_b V_b$; n_b , V_b – the beam density and its speed; ρ_{dr} – the density of the substance of the micro-droplet; γ – the secondary emission coefficient; α – the emissivity of the microdroplet; σ – the Stefan-Boltzmann constant.

In (1), the coefficient $(1+r_d^2/r_{\rm dr}^2)$ approximately in the vicinity of $r_d \ge r_{dr}$ demonstrates the fact that the surface from which the ions are accelerated to the micro-drop can be larger than the surface of the micro-drop. Indeed, the ions are accelerated from the surface where the field of the micro-droplet penetrates. Then, if the Debye radius r_d of plasma electrons is less than the size of the micro-drop $r_d < r_{dr}$, then the surface that collects ions is approximately equal to the surface of the micro-drop. If the Debye radius of plasma electrons is larger than the size of a micro-droplet, then the surface that collects ions is approximately equal to the Debye surface πr_d^2 . For experimental parameters $(T_e = 3 \text{ eV},$ typical

 $n_e = 10^{12}$ cm⁻³), the Debye radius of electrons is equal to $r_d = (T_e/4\pi n_e e^2)^{\frac{1}{2}} \approx 13 \ \mu\text{m}.$

It can be seen that only for the largest micro-droplets $r_d \rightarrow r_{dr}$ may be achieved. That is, the class of large micro-droplets $r_d < r_{dr}$ includes only a small part of the largest micro-droplets. Thus, practically all micro-droplets belong to the class of small micro-droplets with $r_d >> r_{dr}$; as will be shown, they evaporate efficiently.





It is clear that the contribution to the heating of macroparticles from multi-charged ions is greater than from singly-charged ones. However, in this article, we consider the approximation of singly charged ions only.

It should be noted that the coefficient $(1+(r_d/r_{dr})^2)$ is obtained in the approximation of the absence of competition of micro-droplets. That is, it is obtained in approximation $n_{dr}^{-1/3} > r_d$.

It can be seen from equation (1) that the influence of the mass of the droplet M_{dr} remains in the denominator as the product of the density of the substance of the micro-droplet ρ_{dr} and its radius r_{dr} .

2. CONDITION OF MORE INTENSIVE EVAPORATION OF SMALL MICRO-DROPLETS

From equation (1), it can also be seen those small micro-droplets whose radius r_{dr} is smallerthan the Debye radius of plasma electrons $r_{dr} \ll r_d$ are most likely evaporate. The rate of energy pumping into a small micro-droplet (that is, the rate dT_{dr}/dt of its temperature T_{dr} increase) is proportional to

$$\frac{dT_{dr}^{(s)}}{dx} \sim \frac{1}{r_{dr}^3}.$$
(3)

While for large micro-droplets, the size of which is not less than the Debye radius of plasma electrons, we have

$$\frac{dT_{dr}^{(l)}}{dx} \sim \frac{1}{r_{dr}}.$$
(4)

From the comparison (3) and (4), we can conclude that energy pumping is more intense for small micro-droplets.

3. CONDITION FOR DIRECT HEATING OF LARGE DROPLETS BY HIGH-ENERGY ELECTRONS

Under certain conditions, direct heating of microdroplets by high-energy electrons exceeds indirect heating of micro-droplets, when high-energy electrons first heat plasma electrons. The plasma electrons then heat the micro-droplets directly, as well as by accelerating the plasma ions on the micro-droplets. This condition has the form $\gamma e \varphi_0 > 4\sqrt{2} \varepsilon_i (1 + r_d^2/r_{dr}^2)$. Based on the fact that this condition is more easily fulfilled for the largest micro-droplets, the radius of which is not less than the Debye radius of electrons. From this condition for the largest micro-droplets, we have $\gamma e \varphi_0 > 4\sqrt{2} \varepsilon_i$. If $\varepsilon_i \approx 40 \text{ eV}$ and $\gamma \approx 0.1$, then at $e \varphi_0 > 2260 \text{ eV}$ direct heating of micro-droplets by high-energy electrons exceeds heating of micro-droplets by electrons and plasma ions.

4. CONDITION FOR SIMULTANEOUS EVAPORATION OF MICRO-DROPLETS AND PLASMA HEATING

Now we will show that even taking into account the fact that plasma electrons and ions lose energy due to the evaporation of micro-droplets, the plasma heats up instead of cooling down. To do this, let's compare the flow of energy pumped into the plasma by the beam and the flow to all micro-droplets of plasma ion energy I_{ε_i} in the approximation that the volume of all micro-droplets is a small part of the plasma volume,

$$I_{\varepsilon b} = 2\pi R L \gamma n_i V_s e \varphi_0,$$

$$I_{\varepsilon i} = \pi (r_{dr}^2 + r_d^2) n_i V_s \pi R^2 L n_{dr} \varepsilon_i,$$

$$I_{\varepsilon b} / I_{\varepsilon i} = 2\gamma e \varphi_0 (r_{dr}^2 + r_d^2) \pi R n_{dr} \varepsilon_i.$$
 (5)

For $\varepsilon_i \approx 40 \text{ eV}$, $\varepsilon_b \approx 1500 \text{ eV}$, $\gamma \approx 0.1$ and large microdroplets from (5) we obtain

$$\frac{I_{\varepsilon b}}{I_{\varepsilon i}} = \frac{2.4}{r_{dr}^2} R \ n_{dr} > 1.$$
(6)

Condition (6) is fulfilled if the intersection of all microdroplets is less than the surface area of the beam injection.

Thus, the plasma is heated under the influence of the beam, and not cooled, even taking into account the fact that the electrons and ions of the plasma lose energy for the evaporation of micro-droplets.

5. HEATING SPEEDS OF PLASMA ELECTRONS AND MICRO-DROPLETS

In order to clearly see the ratio of heating rates of plasma electrons and micro-droplets, consider small times after the start of the electron beam exposure, when the change in the radii of the micro-droplets can be neglected. Consider equations (1), and (2) in the simplest case of neglecting the contribution to the evaporation of micro-droplets by gas and the direct evaporation of micro-droplets by high-energy electrons. We also neglect the contribution to the evaporation of micro-droplets of plasma electrons compared to the contribution of plasma ions. This is determined by the fact that the currents of electrons and ions per micro-drop are equal (Fig. 2), but the energy $e\varphi_{dt} \gg T_e$ which is brought by the ions to the micro-drop is greater than that of the electrons.



Fig. 2. Flows of high-energy electrons, as well as plasma electrons and ions on a micro-droplet

Then there remains the evaporation of micro-droplets only by plasma ions when they are accelerated in the electric potential of the micro-droplet created by plasma electrons. And so, the equations (1), (2) take the form

$$\frac{dT_{dr}}{dt} = \frac{3}{r_{dr}\rho_{dr}c} \left\{ -\alpha\sigma T_{dr}^{4} + 0.6\sqrt{2}n_{0}\varepsilon_{i}\sqrt{\frac{T_{e}}{m_{i}}} \left(1 + r_{d}^{2}/r_{dr}^{2}\right) \right\},$$
(7)

$$\frac{dT_e}{dt} = 1.2 \,\gamma \, e \,\varphi_0 \sqrt{\frac{k \,T_e}{m_i}}/R. \tag{8}$$

Integrating (8), it can be obtained that T_e grows approximately according to

$$T_e = t^2 \cdot 0.36 \left(\frac{\gamma e \varphi_0}{R}\right)^2 / m_i \tag{9}$$

that is, proportional to the square of time $T_e \sim t^2$.

From equation (7) at a short time after the beginning of the impact of the electron beam, when the change in the radii of the micro-droplets can be neglected, it follows, taking into account the expression $\varepsilon_i = T_e \ln[0.6(2m_e/m_i)^{1/2}]$, that the temperature $T_{dr}^{(l)}$ of largemicro-droplets grow according to

$$T_{dr}^{(l)} \approx 0.1\sqrt{2}n_0 t^4 \left(\frac{\gamma e\varphi_0}{R}\right)^3 \frac{\ln[0.6 (2m_e/m_i)^{1/2}]}{r_{dr}\rho_{dr}c m_i^2}.$$
 (10)

The temperature $T_{dr}^{(s)}$ of small micro-droplets increases even faster

$$T_{dr}^{(S)} \approx 0.01t^6 \left(\frac{\gamma e \varphi_0}{R}\right)^5 \frac{\ln[0.6 (2m_e/m_i)^{1/2}]}{\sqrt{2}\pi e^2 r_{dr}^3 \rho_{dr} c m_i^3}.$$
 (11)

That is according to (10) $T_{dr}^{(l)}$ grows proportional to t^4 , whereas from (11) $T_{dr}^{(s)}$ is proportional to t^6 .

When the thermal radiation of micro-droplets becomes significant, their temperature stabilizes. The temperature $T_{dr}^{(l)}$ of large micro-droplets stabilizes at

$$T_{dr}^{(l)} = T_e^{3/8} \left\{ \frac{0.6\sqrt{2}n_0 \ln[0.6(2m_e/m_i)^{1/2}]}{\alpha\sigma\sqrt{m_i}} \right\}^{1/4}.$$
 (12)

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The temperature $T_{dr}^{(s)}$ of small micro-droplets stabilizes at

$$T_{dr}^{(S)} = T_e^{5/8} \left\{ \frac{0.3n_0 \ln[0.6 (2m_e/m_i)^{1/2}]}{\sqrt{2}\pi n_0 e^2 r_{dr} \alpha \sigma \sqrt{m_i}} \right\}^{1/4}.$$
 (13)

The stationary temperature of small micro-droplets is greater than that of large micro-droplets because, as it is seen from (12) and (13)

$$\frac{T_{dr}^{(s)}}{T_{dr}^{(l)}} \approx \sqrt{\frac{r_d}{r_{dr}^{(s)}}} \gg 1.$$
(14)

Thus, from (14) it can be concluded that small microdroplets evaporate more intensively. So, the largest micro-drops can be separated, and the small micro-drops can be evaporated by the new device.

6. TWO APPROXIMATIONS FOR THE ENERGY BALANCE EQUATION OF THE ELECTRON BEAM AND PLASMA ELECTRONS

Until now, we have used the approximation that all the energy of the beam is transmitted to the plasma electrons, except for that transmitted by the beam directly to the drops:

$$\frac{dT_e^{(1)}}{dt} = 0.6\gamma e \varphi_0 \sqrt{\frac{kT_e}{m_i}} \left(\frac{2}{R} - n_{dr} \pi r_{dr}^2\right).$$
(15)

Equation (15) for the plasma electron temperature T_e .

One may look at the less favorable case when the beam transfers energy to plasma electrons only due to collisions: $\frac{dT_e^{(2)}}{dt} = \frac{dT_e^{(1)}}{dt} v_{eb} \frac{R}{3V_b}$; v_{eb} – frequency of beam collisions with plasma electrons. The model using collisions can lead to a lower intensity of energy exchange of the beam with plasma electrons, that is $\frac{dT_e^{(1)}}{dt} > \frac{dT_e^{(2)}}{dt}$ only in the weakly collisional case $v_{eb} < \frac{R}{3V_b}$.

However, the reality is somewhere in between. Namely, beam-plasma instability can play a big role in the transfer of electron beam energy to plasma electrons.

7. OPTIMUM PLASMA DENSITY

It should be noted that the higher the plasma electron density, the faster the droplets evaporate, and the energy absorbed by the plasma from high-energy electrons is also proportional to the plasma electron density. Then less energy reaches the drops, especially taking into account the fact that plasma electrons have a finite lifetime. That is, plasma electrons take energy, and then it is not used for evaporation of microdroplets. Therefore, there must be an optimal density of plasma electrons. That is, for the destruction of macro-particles in a plasma with a higher concentration, more powerful beams are needed than in a plasma with a lower concentration.

For effective droplet evaporation, a significant density of the energy reserve n_eT_e , which is proportional to the plasma density n_e , must be transferred to the plasma. But if the density of the plasma is too low, then ions must be collected from the larger volume of the

droplet's plasma so that the droplet evaporates. If the competition starts, then the drop cannot get more ions. That is, the maximum number of ions that can fall on a droplet is determined by the size of $n_{dr}^{-1/3}$. That is, the energy of the ions collected from the volume with a radius of $n_{dr}^{-1/3}$ should be sufficient for the evaporation of the drop $\frac{4\pi}{3}R^3n_ie\varphi_0 > \frac{4\pi}{3}n_{ss}r_{dr}^3\varepsilon_{ev}$, $R = n_{dr}^{-1/3}$. The plasma density necessary for evaporation follows from the latter $\frac{n_i}{n_{dr}}e\varphi_0 > n_{ss}r_{dr}^3\varepsilon_{ev}$ that is $\frac{n_i}{n_{dr}} > \frac{n_{ss}r_{dr}^3\varepsilon_{ev}}{e\varphi_0}$. This is the optimal plasma density and the optimal droplet density $\frac{n_i^{(opt)}}{n_{dr}} > \frac{n_{ss}r_{dr}^3\varepsilon_{ev}}{e\varphi_0}$. That is, the plasma density should be no less than optimal, and the droplet density should be no more optimal.

8. EVAPORATION RATE OF DROPLETS

Depending on the parameters, two droplet evaporation modes can be implemented. During droplet evaporation, one mode can change the other. One mode is realized if the density of drops n_{dr} in the arc plasma flow is sufficiently large. This mode is implemented if the next inequality holds $n_{dr}^{-1/3} \ll r_d (r_d$ the Debye radius of plasma electrons), This condition corresponds to the fact that there are many drops in a sphere whose radius is equal to the Debye radius $n_{dr}r_d^3 \gg 1$. In this case, the drops are not independent. Their competition for plasma ions, which accelerate to the droplets and determine their evaporation, becomes particularly significant. The size of the volume from which plasma ions are accelerated to drops is approximately equal to $n_{dr}^{-1/3}$. Then the energy flow per drop, which is determined by plasma ions, is proportional to the large parameter $n_{dr}^{-1/3}$ or r_{dr} , where r_{dr} is the radius of the droplet.

Initially or in the process of evaporation of droplets in the arc plasma flow, the density of droplets n_{dr} may become small. Then the first condition may become invalid. If the inequality holds $n_{dr}^{-1/3} > r_d$ in a sphere whose radius is equal to the Debye radius, there is no more than one drop. In this case, the drops are independent and they do not compete. The radius of the sphere from which plasma ions are accelerated to drops is equal to r_d . Then the energy flow per drop, which is determined by the plasma ions, is proportional to the larger parameter from r_d and r_{dr} .

When passing from the first regime (first condition) to the second regime (second condition) as the droplets evaporate, the flow of ions per drop increases, and their energy also increases (because the potential of the droplets increases (without taking into account thermal emission and secondary electron-electron emission)). It should be noted that as the temperature of the droplets increases, thermal emission from the droplets increases, and secondary electron-electron emission also grows when the droplets evaporate. So, the second regime may be the most favorable for the evaporation of droplets. In this mode, the highest droplet temperature and the most significant rate of their evaporation are achieved.

9. SIMULATION RESULTS

As was shown earlier, when a plasma flow passes through a plasma lens with a magnetic field, if a negative potential of 2...3 kV is applied to its central electrode, a radial beam of fast electrons is self-consistently formed, the energy of which is sufficient to destroy the drop [8]. Also, this beam heats the plasma electrons, which in turn interact with the drops, this is described by bursts (1), (2). Numerical results of solving these equations for the case of a plasma lens with a magnetic field are presented in Figs. 3, 4. During the calculation, it was assumed that the potential of the drop is calculated according to the expression: $\varphi_{dr}=(T_e/e) \ln[0.6(2m_e/m_i)^{1/2}]$.



Fig. 3. Heating of plasma electrons by a beam of fast electrons



Fig. 4. Heating of the drop by electrons over time depending on the size of the drop: $1 - r_{dr} = 0.5 \ \mu m; \ 2 - r_{dr} = 1 \ \mu m; \ 3 - r_{dr} = 5 \ \mu m$

On Fig. 3 shows the heating of plasma electrons over time. It can be seen that, taking into account the heating of the plasma by a beam of fast electrons, the temperature of the plasma electrons also rapidly increases and they also begin to heat the droplets.

On Fig. 4 shows the change over time in the temperature of the drop depending on the radius. As you can see, for small drops it grows rapidly and reaches the boiling point, and they begin to evaporate. For large drops, its temperature rises more slowly and it takes more

time to reach the boiling point, and evaporation has begun.

CONCLUSIONS

A system of two equations describing the evaporation of micro-droplets in an arc plasma flow under the action of a self-consistent electron beam and the condition of direct heating of micro-droplets by fast electrons was formulated. It was shown that the plasma is heated under the influence of the beam, even taking into account the fact that the electrons and ions of the plasma lose energy for the evaporation of micro-droplets. The heating rates of plasma electrons and micro-droplets at short exposure times of a self-consistent electron beam were obtained. It was shown that small micro-droplets evaporate more intensively.

It should be noted that the higher the plasma electron density, the faster the droplets evaporate, and the energy absorbed by the plasma from high-energy electrons is also proportional to the plasma electron density. Then less energy reaches the droplets, especially taking into account the fact that plasma electrons have a finite lifetime. That is, plasma electrons take energy, and then it is not used for evaporation of microdroplets. Therefore, there must be an optimal density of plasma electrons. That is, for the destruction of macro-particles in a plasma with a higher concentration, more powerful beams are needed than in a plasma with a lower concentration.

The magnitude of the energy contribution of the fast electrons flow to the overall heating of micro-droplets depends on the temperature of the plasma electrons and the potential of the droplet. As the calculations show, during the stay of the drop in the volume of the plasma device, it has the opportunity to heat up to the temperature necessary for its evaporation.

It is clear that the contribution to the heating of macroparticles from multi-charged ions is greater than from singly-charged ones.

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ФОРМУВАННЯ ЕЛЕКТРОННОГО ПУЧКА І ЙОГО РОЛЬ У НОВІЙ ПЛАЗМООПТИЧНІЙ СИСТЕМІ ДЛЯ ВИПАРОВУВАННЯ КРАПЕЛЬ У ДУГОВІЙ ПЛАЗМІ

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Розглядаються випаровування мікрокрапель у потоці дугової плазми під дією самоузгодженого електронного пучка та умова прямого нагріву мікрокрапель швидкими електронами. Показано, що плазма гріється під дією пучка, навіть з урахуванням того, що електрони і іони плазми втрачають енергію на випаровування мікрокрапель. Встановлено, що дрібні мікрокраплі випаровуються інтенсивніше. Показано, що має бути оптимальна щільність електронів плазми. Для руйнування макрочастинок у плазмі з більшою концентрацією потрібні потужніші пучки, ніж у плазмі з меншою концентрацією.