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Modelling the evaporation of vacuum arc macroparticles by a beam of energetic electrons

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The presence of macroparticles from cathode material in the plasma prevents the use of arc erosion sources in applications requiring high-quality coatings, especially those requiring low defect density and low roughness at the nanometer level. Here, we discuss an approach using energetic (keV) electrons injected into the arc plasma to introduce additional energy into the plasma which promotes evaporation of the macroparticles and may eventually lead to their complete destruction. By solving a self-consistent system of equations for the energy balance, the ion and electron currents to and from a macroparticle, and the macroparticle mass balance, it is shown that favorable conditions for the evaporation of macroparticles may exist.

Introduction

Arc erosion plasma sources are widely used in modern science and industry. Due to the high deposition rate, they are very productive tools for the synthesis of various functional and protective coatings, for modifying the surface properties of structural and decorative materials. However, they have a significant drawback - the presence of microdroplets of cathode material in the generated plasma flow, that limits the use of this type of source in the formation of high-quality coatings, especially those of submicron thickness. This is especially critical for optics and microelectronic devices, since the thickness of the deposited films is 0.01-1 μm , i.e. on the order of the size of the droplets. The modern filters, that are used for removing macroparticles, lead to significant losses of the plasma flow followed by a decrease in the deposition rate, which compromises one of the unique features of these sources, namely their high deposition rate. A filter concept that evaporates macroparticles without removing their mass from the plasma flow would improve the quality of the coating and increase the deposition rate. The energy of the erosion plasma source alone is not enough to evaporate microdroplets of the cathode material [1], so it is necessary to add energy to the plasma flow, for example, with a laser or an electron beam. They introduce additional energy into the plasma which promotes evaporation of the macroparticles and may eventually lead to complete destruction of macroparticles.

Base equations and results

The main parameters describing a drop in a plasma flow and influencing its evaporation

process are its size, charge and temperature. The rate of evaporation of the drop is determined by its temperature, which is determined from the balance of the energy absorbed and lost by the drop. The presence of an electron beam in the system significantly affects the charge and temperature of the drop in the plasma: it is the source of a flow of fast electrons to the droplet, which increases the charge of the droplet and brings additional energy, heating it, but also causes additional emission processes from the surface of the droplet, which carry away energy from the droplet and cool it. Thus, to describe the process of droplet evaporation, it is necessary to solve a system of three interconnected differential equations: the energy balance equation, the current balance equation, and the mass balance equation:

$$M_{dr} c \frac{dT_{dr}}{dt} = Q_e + Q_i + Q_b - Q_{rad} - Q_{vap} - Q_{hex} - Q_{ee} - Q_{e,th} , \quad (1)$$

$$\frac{dq_{dr}}{dt} = I_i + I_e + I_b - I_{ee} - I_{e,th} \quad (2)$$

$$\frac{dM_{dr}}{dt} = \frac{I_i}{e} m_i - 4\pi r_{dr}^2 \cdot \gamma_a m_a. \quad (3)$$

Here c – is the specific heat capacity of the drop material, M_{dr} , r_{dr} , q_{dr} and T_{dr} are the mass, radius, charge and temperature of the droplet respectively, Q_i , Q_e , Q_b and Q_{rad} , Q_{vap} , Q_{hex} , Q_{ee} , $Q_{e,th}$ – are the energy fluxes from plasma ions and electrons, and beam electrons, heating the droplet and from radiation, evaporation, heat exchange, secondary electron emission and thermal (field-electron) emission, cooling the droplet, respectively. I_i , I_e and I_b – are the currents plasma ions and electrons, and beam electrons to the droplet, I_{ee} , $I_{e,th}$ – are the currents of secondary electron emission and thermionic (field) emission from the droplet, respectively, m_i , m_a – ion and atom mass, respectively, γ_a – is the evaporation rate, which determines the number of atoms evaporated from a unit of surface during a unit of time, that has next form: $\gamma_a = \frac{\alpha_k p(T_{dr})}{\sqrt{2\pi m_a k T_{dr}}}$ [2], where α_k is the accommodation coefficient equal to the probability that when a vapor molecule hits the surface of the drop, it will not be reflected, $p(T_{dr})$ is the equilibrium saturated vapor pressure of the evaporating droplet material, which depends on the temperature T_{dr} on the surface of the droplet, for that can use the equation [3]: $\lg(p(T)) = A + \frac{B}{T} + C \lg(T) + DT$, where A , B , C , D – are material-dependent coefficients.

To calculate the currents, we used the Orbital-motion-limited theory (OML) [4], according to which the currents from (2) arriving at the droplet are described as follows: $I_i(r_{dr}) = \pi r_{dr}^2 e n_i V_{ib} \left(1 - \frac{e\varphi_{dr}}{\varepsilon_{ib}}\right)$, $I_e(r_{dr}) = \pi r_{dr}^2 e n_e V_{Te} \exp\left(-\frac{e\varphi_{dr}}{kT_e}\right)$, $I_b(r_{dr}) = \pi r_{dr}^2 e n_{eb} v_{eb} \left(1 - \frac{e\varphi_{dr}}{\varepsilon_{eb}}\right)$. They bring energy into the droplet, heating it, and for the corresponding energy fluxes from (1) we can write: $Q_i = I_i (2k_B T_i + (\varepsilon_{vap} + \varepsilon_{ex} + \varepsilon_i) + e\varphi_{dr} + \Phi_{Cu})/e$, $Q_e = I_e (2k_B T_e + \Phi_{Cu})/e$, $Q_b = I_b (\varepsilon_b + \Phi_{Cu})/e$.

For the emission currents from the droplet, we can use the Sternglass equation [5] for the secondary electron emission I_{ee} and the Murphy-Good equation [6] for the thermo-field emission $I_{e,th}$ in the region of dominating thermionic emission, that is suitable without

limitation on the values of electric field strength and temperature: $I_{ee}(r_{dr}) = \delta_{ee} I_b(r_{dr})$, $\delta_{ee} = 7.4 \delta_m \frac{\varepsilon_{eb}}{\varepsilon_{max}} \exp\left(-2\sqrt{\frac{\varepsilon_{eb}}{\varepsilon_{max}}}\right)$; $I_{e,th} = 4\pi r_{dr}^2 T_{dr}^2 \frac{em_e}{2\pi^2 \hbar^3} \frac{\pi h_0}{\sin(\pi h_0)} \exp\left(-\frac{e(\Phi - \sqrt{\frac{eE}{4\pi\varepsilon_0}})}{k_B T_{dr}}\right)$. They carry away

energy fluxes and cool the droplet: $Q_{ee} = I_{ee}(k_B T_e - \Phi_{Cu})/e$, $Q_{e,th} = I_{e,th}(2k_B T_{dr} - \Phi_{Cu})/e$.

Besides this, there are other processes that cool the droplet: radiation, evaporation and heat exchange with surrounding gas: $Q_{rad} = 4\pi r_{dr}^2 \alpha \sigma_{SB} T_{dr}^4$, $Q_{vap} = 4\pi r_{dr}^2 \gamma_a (\varepsilon_{vap} + 2k_B T_{dr})/e$, $Q_{hex} = 4\pi r_{dr}^2 k n_g v_{T,a} (T_{dr} - T_g)$. In these expression Φ_{Cu} is the work function of material, ε_{vap} is the energy of vaporization of one atom, ε_{ex} is the energy of excitation bound electrons, ε_i is the ionization energy, α is the emissivity of the droplet material, and σ_{SB} is the Stefan-Boltzmann constant, n_g , T_g are surrounding gas density and temperature, respectively.

Typical parameters of the plasma flow formed by the vacuum arc source are used as parameters for solving the equations (1) - (3): the energy and temperature of the ions in the flow are $\varepsilon_{ib} \sim 20-60$ eV, $T_i = 1$ eV, respectively. The electron temperature is $T_e = 3$ eV, the plasma density is $n_0 = 10^{10} \text{ cm}^{-3}$ to 10^{12} cm^{-3} . Assuming the distance between the cathode and the substrate to be 20 cm and assuming a microparticle velocity of $v_{dr} \sim 100$ m/s, we calculate a residence time of the microparticle in the plasma of $t_{res} \sim 2$ ms.

There are two main mechanisms that can affect the destruction of a droplet under the action of an electron beam - these are Rayleigh decay and evaporation. Evaporation occurs at any temperature, but at a higher temperature it occurs faster, it is important that it exceeds the temperature at which Q_{vap} becomes greater than Q_{rad} (for copper this is about 1800 K). Rayleigh decay - when the droplet receives a charge exceeding the Rayleigh limit ($q_R = 8\pi\sqrt{\varepsilon_0 \sigma r_{dr}}$). It is important to understand which mechanism and under what conditions will play a decisive role. As can be seen from Fig. 1, the droplet charge depends significantly on the beam energy and increases rapidly with it. The charge also depends on the beam density: if it is $n_b < 0.01 n_0$, then the droplet very quickly acquires an equilibrium charge (blue, green curve) that is not much exceed its equilibrium charge without the beam.

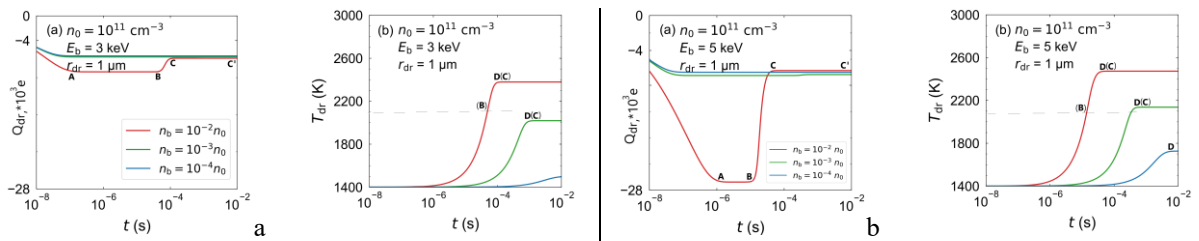


Fig. 1. Charge and temperature dynamics of a copper droplet with radius $r_{dr}=1 \mu\text{m}$ ($n_0=10^{11} \text{ cm}^{-3}$, note the use of a logarithmic time-axis): a) $E_b=3\text{keV}$, b) $E_b=5\text{keV}$

If $n_b \geq 0.01 n_0$, the drop very quickly acquires a large negative charge (that, at $n_b > 0.1 n_0$, can exceed the Rayleigh limit) and retain it for some time (section AB) until the drop temperature reaches a critical value at which thermionic emission begins, droplet lost charge (line BC) and

reached equilibrium state (CC'). The temperature also depends on the energy and density of the beam and increases with their increase and under $n_b > 10^{-2} n_0$ and energy 3-5 keV is possible to reach temperatures 2400 - 2600 K, thus it can be assumed that evaporation is the main process leading to the removal of droplets from the plasma flow if $n_b \leq 0.1 n_0$. Fig. 2a shows the results of solving the equations (1) - (3) for $n_0 = 10^{11} \text{ cm}^{-3}$ and 10^{12} cm^{-3} , $E_b = 5 \text{ keV}$ and different beam density for droplets with an initial radius $r_{dr} = 1 \text{ }\mu\text{m}$ for a typical residence time of the droplet in the plasma flow $t_{res} \sim 2 \text{ ms}$. We see that increasing the beam density significantly reduces the evaporation time.

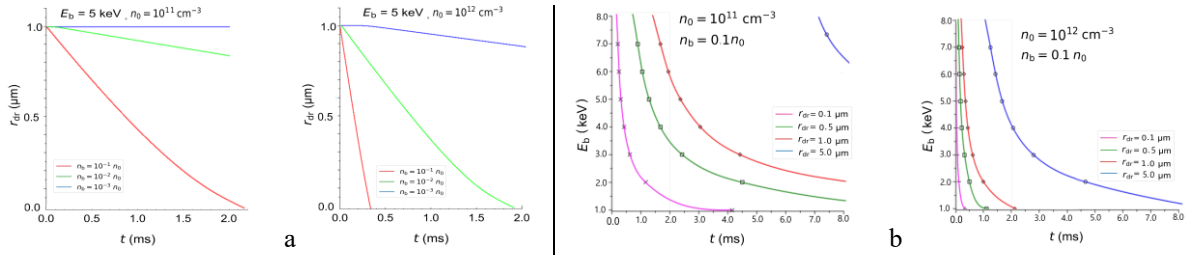


Fig. 2. a) Dynamic of the copper droplet's radius with initial size $r_{dr} = 1 \text{ }\mu\text{m}$: $E_b = 5 \text{ keV}$, $n_0 = 10^{11} \text{ cm}^{-3}$ (on the left) 10^{12} cm^{-3} (on the right); b) beam energy, that need for evaporation droplet with different size in giving time: $n_0 = 10^{11}$ (left), $n_0 = 10^{12} \text{ cm}^{-3}$ (right)

Fig. 2 b) shows what beam energy E_b is required for complete evaporation in a certain time of a droplet with initial radii of 5, 1, 0.5 and 0.1 μm at a plasma density of $n_0 = 10^{11} \text{ cm}^{-3}$ and 10^{12} cm^{-3} if the beam density $n_b = 0.1 n_0$. We see that smaller droplets evaporate much faster than larger ones, but for specific conditions possible also evaporate big droplet with $r_{dr} = 5 \text{ }\mu\text{m}$ during residence time. But drops with $r_{dr} \leq 1 \text{ }\mu\text{m}$ can be evaporated during this time by an electron beam with an energy of 1-2 keV, if its density is 10^{11} cm^{-3} .

To summarize, the main parameters that play a role in droplet evaporation are the energy and density of the electron beam, as well as the plasma density. At a higher plasma density, an electron beam with a lower density and lower energy can be used to evaporate droplets, and at a higher beam energy, a lower beam density can be used, and vice versa. It is possible to select the optimal beam density and its energy for effective droplet evaporation, as well as the location of the filter.

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