CHARGING OF A MACROPARTICLE IN CATHODIC ARC SHEATH

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The macroparticle (MP) contamination is the most important technological problem of cathodic vacuum arc deposition of coatings. The electrostatic reflection of MP from the substrate is considered as a possible reason for the reduction in number of MPs in the presence of reactive gas. It is shown that the probability of the electrostatic reflection increases with increasing the nitrogen pressure due to expansion of the region in the sheath where MP charge is negative. The MP charge in the collisional plasma sheath is calculated taking into account the dependence of the charge state of ions, ion fluxes and ion energies on the background gas pressure.

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INTRODUCTION

Vacuum arc deposition is a recognized technique for the formation of thin film and coatings [1]. For wide range of applications, including decorative coatings and hard coatings on cutting tools, reactive gas is added [2]. The vacuum arc sources generate highly ionized metal plasma with multiply charged ions [3]. The mean ion charge state is 2…3 and typically higher for materials with high melting point. The ions have supersonic velocities which correspond to ion energy in the range 20…200 eV, depending on the source material [4]. A disadvantage of cathodic arc deposition is the emission of macro-particles (MPs) during arcing. MPs are generally molten metal droplets (sometimes solid) generated by the action of the cathode spots [5]. The MPs occur in the range of size from a fraction to tens of microns. There is a strong dependence of MP production on the cathode material. The cathode erosion in the droplet phase decreases with increasing the cathode material melting temperature [6].

The incorporation of MPs into the coating degrades the quality of the films, e.g. produces surface roughening, protuberances, bumps and pinholes [7]. MPs may make a considerable portion of the coatings mass. This substantially limits the possibilities of vacuum arc plasma in coating technologies. Thus, MP contamination is regarded as the most important technological problem.

Several methods have been developed to eliminate MPs, such as magnetic filters [8], magnetically steered arc [9], background gas pressure [10 - 12], and substrate biasing [10]. The last effect has been theoretically investigated in our previous studies [13, 14]. The effect of background gas pressure on MPs in cathodic arc plasma deposition has been investigated by Keidar et al. [15]. They proposed a model for interaction between the negatively charged MP and floating surface. According to their model, the probability of electrostatic reflection of MP increases with background gas pressure as a consequence of the increased substrate potential. However, the MP charge used in this analysis was constant for a quasi-neutral plasma. In the present work, we propose the model of MP charging in collisional plasma sheath which takes into account the dependence of the charge state of ions, ion fluxes and ion energies on the background gas pressure. To calculate MP charge, the collisional plasma sheath model of Sheridan and Goree [16] is modified to account for multiple ion species and combined with orbital motion limited (OML) theory [17].

1. PLASMA CHEMISTRY MODEL

The main mechanism for the ion-neutral collisions is considered to be charge exchange, which corresponds to an electron transfer from a gas molecule to a multiply charged ion. Schematically the charge exchange reaction between a metal ion of charge state *Z* and nitrogen molecule N_2 can be written as

$$M^{Z+} + N_2 \Rightarrow M^{(Z-1)+} + N_2{}^{+},\ Z = 1, 2, 3. \quad (1)$$

Charge exchange collisions of metal ions with nitrogen molecules are relevant for the reduction of the average charge state of metal ions. The efficiency of different type of reactions is determined by cross section σ_k that changes the charge of metal ion from *Z* to *Z-1*. The cross section for charge exchange increases with the ion charge state number *Z* quadratically [18]

$$\sigma_k = \pi a_0^2 Z^2 \left(I_H / I\right)^2, \quad (2)$$

where $a_{0=}$ 0.529×10^{-8} cm is the radius of the first Bohr orbit of the hydrogen atom, I_H is the ionization potential of the hydrogen atom, and *I* is the ionization potential of the target particle.

The rate equation for the particle density of metal ions n_k in a state *Z* can be written in the form

$$\frac{dn_k}{dt} = nn_{k+1}\left\langle \sigma_{k+1} u_{k+1} \right\rangle - nn_k \left\langle \sigma_k u_k \right\rangle, \quad (3)$$

where *n* is the particle density of neutral background gas, u_k is the velocity of metal ion. Here, we don't take into account the losses caused by the diffusion to the wall. Therefore, equations (3) give upper limits to particle density of metal ions.

In contrast to metal ions, the gas ions are produced by charge exchange as well as by electron impact ionization [19]. The rate equation for the particle density of gas ions n_i is

$$\frac{dn_i}{dt} = k_i n_e n + \sum_1^3 nn_{k+1}\left\langle \sigma_{k+1} u_{k+1} \right\rangle - \sum_1^3 nn_k \left\langle \sigma_k u_k \right\rangle, \quad (4)$$

where n_e is the particle density of electrons, k_i is the rate coefficient of electron-impact ionization of gas [20].

Here, we don't account for secondary electrons from the substrate which can interact with the background gas. The ions, striking the substrate, may cause the electron emission. Because of the relatively low kinetic energy of the ions (below 1 keV), only the potential electron emission (PEE) can be of importance in the vacuum arcs. PEE requires that the ionization energy of incident ion E_i twice exceeds the work function of the material W [21]

$$E_i > 2W \, . \tag{5}$$

This inequality means that one electron is necessary to neutralize the arriving ion and the second is the one that is emitted. For multiply charged metal ions, the ionization energy is substantially less than E_i given by (5) since the work function of the target material is approximately 4.5 eV for most metals [21]. Thus, the singly charged metal ions do not cause PEE.

The inequality (5) is satisfied only for gas ions. For this case, the secondary electron yield is given by Baragiola et al. [22]

$$\gamma = 0.032(0.78E_i - 2W). \tag{6}$$

According to eq. (6), the resulting secondary electron yields for N_2^+ and N^+ ions are about 0.01. For example, at pressures above 0.1 Pa the titanium ions are almost singly ionized [23].Therefore, the potential electron emission is rather small, and the additional ionization is not important.

2. SHEATH MODEL

Let us consider a negatively biased substrate, immersed into a weakly collisional plasmas, when the mean free path for ion-neutral collisions λ_{jn} is much larger than Debye length $\lambda_D=(\varepsilon_0 T_e /n_0 e^2)^{1/2}$. According to our chemistry model the plasma consists of electrons and j $(j=k, i)$ positive plasma species: k species of metal ions (M^+, M^{2+}, M^{3+}), and i reactive gas species (N_2^+, N^+). The ions of j-th species have the temperature T_j and mass m_j. In our coordinate system a plasma-sheath interface (interface between essentially neutral and non-neutral region) is taken to be the origin, $x=0$. The production and destruction of ions occur in the plasma region, $x<0$. Thus, the relative shares of ions do not vary within the sheath. The position of the substrate is determined by the sheath thickness D. We neglect MP effect on the sheath structure.

The collisional plasma sheath model of Sheridan and Goree [16] can be generalized to multispecies plasmas by assuming that the positive plasma components are described by fluid theory. The continuity equations for each ion species have no source and sink terms

$$\frac{d}{dx}\left(n_j u_j\right) = 0 \, , \tag{7}$$

where n_j and u_j are the ion density and velocity, respectively.

The ion equations of motion in the fluid approximation are

$$m_j u_j \frac{du_j}{dx} = -q_j \frac{d\phi}{dx} - F_j \, , \tag{8}$$

where ϕ is the potential within the sheath, $q_j = ke$ is the ionic charge (k =1 for gas ions, e is the elementary charge). F_j is the ion drag force, σ_j is the cross section for collisions between ions of type j and neutrals. The cross section is assumed to be independent of the ion velocity. This case corresponds to constant mean free path ($\lambda_{jn} = 1/n\sigma_j$), thereby the ion drag forces are proportional to the square of the ion velocity, $F_j= m_j n\sigma_j u_j^2$.

Ions velocities are assumed to be large enough to satisfy Riemann's generalized Bohm criterion [24]

$$\sum_j \frac{q_j n_{js}}{m_j u_{js}^2} \le \frac{e n_{es}}{k_B T_e} \, , \tag{9}$$

where j runs over the ion species, and subscript s refers to values at the sheath edge. Gas ions have Bohm velocity, and velocities of metal ions are supersonic.

The total mean velocity of ions of j-th species (a combination of directed velocity u_j and mean thermal $\upsilon_{th,j} = (8k_B T_j /\pi m_j)^{1/2}$ velocity) and corresponding energy in the sheath are [25]

$$\upsilon_j(x) = \left(u_j^2 + \frac{8k_B T_j}{\pi m_j}\right)^{1/2} \, , \tag{10}$$

$$\frac{m_j \upsilon_j^2}{2} = \frac{4k_B T_j}{\pi} + \frac{m_j u_j^2}{2} . \tag{11}$$

The Poisson's equation for the potential ϕ is

$$\frac{d^2\phi}{dx^2} = -\frac{e}{\varepsilon_0}\left[\sum_j q_j n_j - n_e\right], \tag{12}$$

where ε_0 is the permittivity constant.

Applying the conservation of current density (7) for each individual ion species gives the ion densities

$$n_j = J_{js} / (q_j u_j), \tag{13}$$

where $J_{js}=q_j n_j \upsilon_{js}$ is the ion current density at the sheath edge.

The electron density obeys the Boltzmann relation

$$n_e = n_{es} \exp(e\phi / k_B T_e) \; . \tag{14}$$

By coupling Poisson's equation (12) with densities (13), (14) to the transport equation (8), the electric field spatial distribution can be obtained in a self-consistent manner. The governing equations can be written as

$$m_j u_j \frac{du_j}{dx} = -q_j \frac{d\phi}{dx} - m_j n\sigma_j u_j^2 \, , \tag{15}$$

$$\varepsilon_0 \frac{d^2\phi}{dx^2} = -\sum_j \frac{J_{js}}{u_j} + e n_{es} \exp\left(\frac{e\phi}{k_B T_e}\right). \tag{16}$$

The above equations can be made dimensionless by introducing the following variables:

$$\Phi \equiv \frac{e\phi}{k_B T_e}, z \equiv \frac{x}{\lambda_D}, \alpha_j \equiv \frac{\lambda_D}{\lambda_{jn}}, \tilde{u}_j \equiv \frac{u_j}{c_j}, j_{js} \equiv \frac{J_{is}}{I_{js}}, \tag{17}$$

where $c_j = (k_B T_e / m_j)^{1/2}$ is the sound velocity of j-th ion species, and $I_{js}=e n_{es} c_j$.

The degree of collisionality α_j is the number of collisions in a Debye length λ_D. It is the important parameter which quantifies the energy loss of ions in the sheath. The parameter α_j is related to the background gas pressure p

$$\alpha_j = \lambda_D n \sigma_j = \frac{\lambda_D p \sigma_j}{k_B T_g}, \tag{18}$$

where T_g is the gas temperature.

The dimensionless forms of governing equations (15) - (16) become

$$\tilde{u}_j \frac{d\tilde{u}_j}{dz} = -k\frac{d\Phi}{dz} - \alpha_j \tilde{u}_j^2 \, , \tag{19}$$

$$\varepsilon_0 \frac{d^2\Phi}{dz^2} = -\sum_j \frac{\tilde{j}_{js}}{\tilde{u}_j} + e n_{es} \exp\Phi \, . \tag{20}$$

This set of equations completely describes the plasma sheath. Adding the boundary conditions, we can numerically solve the system of differential equations. At the substrate ($z=d=D/\lambda_D$) the boundary condition is $\Phi(d)=V_b$, where V_b is applied bias voltage. At the sheath-plasma edge the boundary conditions are $\Phi(0)=0; d\Phi/dz=0$, and $u_j(0)=u_{js}$.

3. MP CHARGING

We consider the MP with a radius a as a spherical probe immersed in the cathodic arc plasma sheath. The charging of a MP in the cathodic arc can be described in the framework of the OML approach [17]. The OML theory is applicable for MP radius a much less than the Debye length λ_D, which in turn, is less than the mean free path of electron-neutral and ion-neutral collisions

$$a << \lambda_D << \lambda_{e(j)n}. \quad (21)$$

The MP is charged through collection of the electrons and both metal and gas ions from the plasma. The floating potential of MP is determined by the balance of electron current I_e and sum of ion currents I_j:

$$I_e(\phi_d) = \sum_{j=k,i} I_k(\phi_d). \quad (22)$$

The MP charge Q is defined by the MP potential with respect to the local sheath potential

$$Q(x) = C(\phi_s(x) - \phi(x)) = C\phi_d(x), \quad (23)$$

where $C=4\pi\varepsilon_0 a$ is the capacitance of the MP.

In the case of Maxwellian electrons and negatively charged MP

$$I_e = \pi a^2 e n_{es}(x)\upsilon_{Te} \exp\left(\frac{e\phi_d(x) + e\phi(x)}{k_B T_e}\right). \quad (24)$$

The velocity distribution function of ions is approximated by a shifted Maxwellian distribution. The currents of gas and metal ions can be written as:

$$I_j = \pi a^2 q_j n_j(x)\upsilon_j(x)\sum_j j_j\left(1 - \frac{2q_j\phi_d}{m_j\upsilon_j^2}\right). \quad (25)$$

Substituting (24) and (25) into MP charging equation (22), the latter can be written in dimensionless form

$$\exp(V_d(z) + \Phi(z)) = \sum_j j_j\sqrt{\frac{\pi m_e}{8m_j}}\left(1 - \frac{ZV_d(z)}{E(z)}\right), \quad (26)$$

where new variables are $V_d=e\phi_d/(k_BT_e)$ and $E=m_j\upsilon_j/(2k_BT_e)$. This balance equation (26) allows one to compute the potential difference V_d and, in turn, the dust charge $Q(z)$ as a function of normalized distance from the sheath edge.

4. RESULTS AND DISCUSSION

The experimental observations reveal that the presence of background gas reduces both the number and size of the MPs in coatings [10 - 12]. As pointed out, for example, in [12], MP sizes for Ti range up to 40 μm in the absence of background gas and 30 μm in the presence of nitrogen. The log-log presentation of the size distribution of Ti MPs is shown in Fig. 1.

These results can be explained by the higher melting point of the surface layer of TiN on the cathode (3203 K) in comparison with that of Ti (1941 K). MPs are formed as a result of the plasma pressure on the liquid pool at the arc spot [26], and their mass is proportional to the cathode spot volume. The reduction in the size of MPs is related to the reduction in the cathode spot size, which is the result of increasing the melting point of the compound layer formed at the cathode surface.

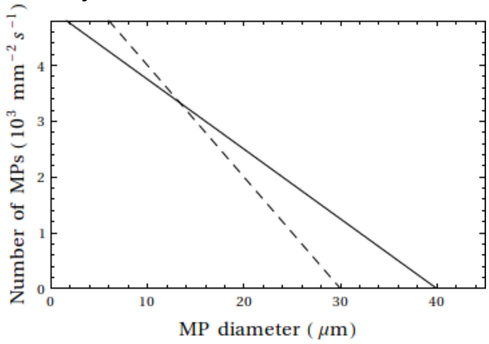

Fig. 1. MP size distribution for Ti, normalized as MP number per area and time deposition for pressures 10^{-3} Pa (solid line) and 1Pa (dashed line). Experimental results were taken from [12]

It has previously been reported that the average number of MPs reduced linearly with chamber pressure [10, 11]. These results can be explained by the higher melting point of the nitrides. Another possible reason for this decrease can be attributed to the nitridation of MPs. The metallic MPs react with nitrogen during their motion to the substrate. As a result, the thin layer of compound is formed on the MP surface, and the MP is expected to reflect from the substrate [7]. We study this effect which is additional factor of the MP reduction.

The model's predictions are tested for a titanium cathodic vacuum arc operated in a nitrogen atmosphere allowing us a direct comparison with the experimental data [10, 11]. The specific plasma parameters are used as typical values from experiments: the substrate bias V_b= –20 V, the electron temperature T_e =2 eV, the plasma bulk density $n=10^{16}$ m^{-3}, the temperature of titanium ions is about 0.3 eV, and both temperatures of nitrogen ions and neutrals are kept at room temperature (0.026 eV). The ion energies used in computation have been taken from the experiment [19]. The investigated pressure range is 0.0001…1.33 Pa with three different pressures selected for analysis. According to eq. (2), the cross sections of the charge exchange reactions of Ti^+, Ti^{2+}, Ti^{3+} ions in nitrogen are $\sigma_1= 6.5\times10^{-17}$ cm^2, $\sigma_2= 2.6\times10^{-16}$ cm^2, $\sigma_3= 5.9\times10^{-16}$ cm^2, respectively. The cross sections of the charge exchange reactions of N_2^+ and N^+ ions in nitrogen are 3.5×10^{-15} cm^2 and 5×10^{-16} cm^2, respectively [27]. According to eq. (18), the resulting degrees of collisionality for Ti^+, Ti^{2+}, Ti^{3+}, N_2^+ and N^+ ions are $\alpha_1= 1.6\times10^{-4}\,p$, $\alpha_2= 6.3\times10^{-4}\,p$, $\alpha_3= 1.4\times10^{-3}\,p$, $\alpha_4= 7.3\times10^{-3}\,p$, and $\alpha_5= 1.2\times10^{-3}\,p$ (p in Pa).

The solutions of the set of rate equations (3) give the dependence of the ion fluxes on the background pressure. As one can see from the Fig. 2, the flux of Ti^{3+} ions decreases practically to zero. The flux of Ti^{2+} ions decreases more slowly. The flux of Ti^+ ions increases with increasing pressure due to charge transfer reactions of the Ti^{2+} ions. The corresponding average charge state of Ti ions decreases from 1.98 to 1.7 with increasing the pressure.

Knowledge of the ion fluxes allows one to compute the MP charge. We consider, as an example, MP with the radius of 0.25 μm. The profile of MP charge is shown in Fig. 3 for different values of pressure. It is seen that the sheath width decreases with ion-neutral collisions. MP charge is positive near the substrate. The point, where MP charge equals to zero, moves towards the substrate with increasing the pressure. This means that the probability of the electrostatic reflection of MP increases with background pressure. These results can be explained by the decreasing of the ion current density.

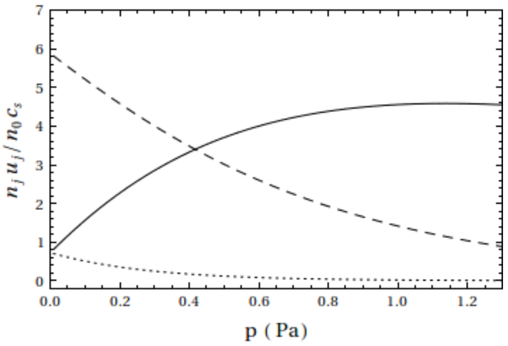

Fig. 2. Dependence of the normalized ion flux on the pressure at the distance 0.25 m from the cathode: Ti^{+} (solid line), Ti^{2+} (dashed line), Ti^{3+} (dotted line)

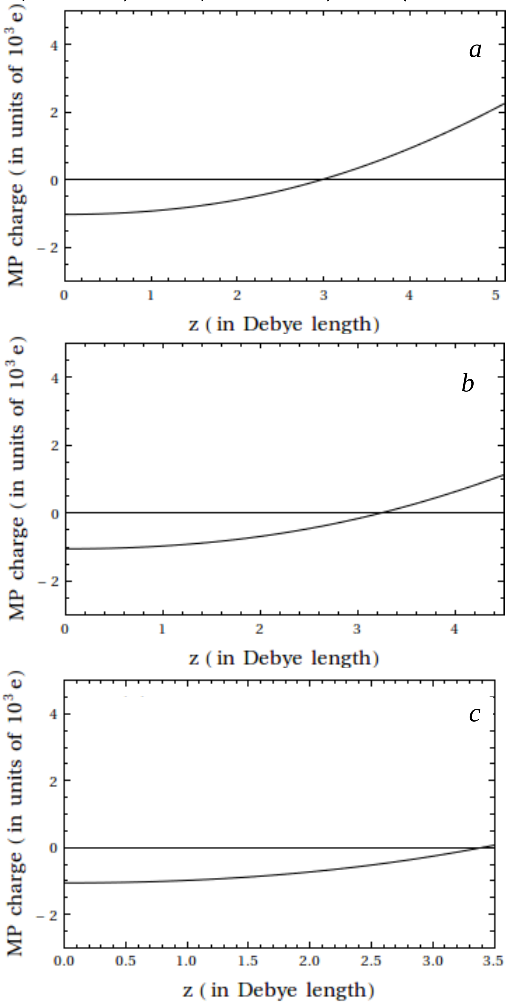

Fig. 3. Dependence of the charge of the MP of radius 0.25 μm on its position z for different pressures: p=0.0001 Pa (a); p=0.1 Pa (b); p=1 Pa (c)

CONCLUSIONS

The presence of a reactive gas leads to enhanced metal plasma-gas interaction which affects the chemistry of the plasma, the ion composition, the ion energies and ion currents. An increase in gas pressure causes a decrease of metal ion charge states, and an increase in gas ion fraction. In turn, the charge of MP is governed by local plasma parameters in the sheath.

The major factor affecting the reduction in number of MPs is processes on the cathode surface in the presence of a reactive gas. The electrostatic reflection of MPs from the substrate is additional factor which is responsible for MP reduction. It is shown that the probability of the electrostatic reflection increases with increasing nitrogen pressure due to expansion of the region in the sheath where MP charge is negative.

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ЗАРЯДКА МАКРОЧАСТИЦЫ В СЛОЕ КАТОДНОЙ ДУГИ

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Наиболее важной технологической проблемой вакуумно-дугового осаждения покрытий является загрязнение макрочастицами (МЧ). Одной из возможных причин уменьшения числа частиц при наличии реактивного газа является электростатическое отражение МЧ от подложки. Показано, что вероятность электростатического отражения увеличивается с давлением азота благодаря расширению области в плазменном слое, где заряд МЧ отрицателен. Заряд МЧ рассчитан с учетом зависимости зарядового состава ионов, потоков и энергий ионов от давления газа.

ЗАРЯДЖЕННЯ МАКРОЧАСТИНКИ В ШАРІ КАТОДНОЇ ДУГИ

О.В. Ромащенко, О.А. Бізюков, І.О. Гірка

Найбільш важливою технологічною проблемою вакуумно-дугового осадження покриттів є забруднення макрочастинками (МЧ). Однією з можливих причин зменшення кількості частинок у присутності реактивного газу є електростатичне відбиття МЧ від підкладки. Показано, що ймовірність електростатичного відбиття зростає з тиском азоту завдяки розширенню ділянки плазмового шару, де заряд МЧ є від'ємним. Заряд МЧ розраховано з урахуванням залежності зарядового складу іонів, потоків та енергій іонів від тиску газу.