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04

Generation of highly ionized metallic plasma by anodic evaporation in a pulsed discharge

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The influence of the parameters of a pulse-periodic (1 ms, 60−450 Hz) discharge with a self-heating hollow cathode and an evaporated anode on the density and degree of ionization of the aluminum vapor stream is studied. It is shown that an increase in the current amplitude at a constant average discharge current of 12−16 A leads to a 3-4-fold increase in the film deposition rate, an increase in the proportion of Al^+ ions in the vapor stream to 100% at 80 A and increase of the proportion of Al^+ ions in the total ion current to 30% at 110 A. A method for determining the proportion of metal ions in a vapor stream by a grid probe with a retarding electrostatic field has been improved

Keywords: self-heating cathode, pulse discharge, anodic evaporation, degree of vapor ionization.

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An important characteristic of film deposition methods is the fraction of ions in the flow of deposited particles. Ion assistance improves the adhesion of the coating with the substrate and influences the growth processes and microstructure of the films, as well as the level of internal stresses. In recent years, IPVD [1] methods based on ion sputtering in a magnetron discharge have been rapidly developing, which allow achieving a high degree of ionization of the metal atom flux by imposing a high-frequency field (ICP MS) [2,3] or using a high-power pulsed discharge (HiPIMS) [1,4].

In discharges with a thermionic cathode and an evaporating anode [5], a high degree of vapor ionization is achieved at currents above 100 A [6]. A high-current discharge with a self-heated hollow cathode (SHHC) was used for ion-assisted deposition of Al_2O_3 films by electron-beam evaporation [7], as well as for melting of metals [8]. For stable operation of SHHC-discharge the flow of inert gas through the cathode cavity is necessary, so the plasma of such a discharge with vaporized anode contains along with metal ions and gas ions. When active gases are fed into the volume in such a discharge, the mode of reactive evaporation is realized and films of binary compounds [9] are deposited.

The degree of plasma ionization in SHHC-discharge is changed by adjusting the proportion of discharge current to the unevaporated anode, but the concentration of gas ions in the plasma also increases. An alternative method can be the use of repetitively-pulsed mode (RPM) of the discharge with vaporized anode. The growth of RPM discharge current will lead to an increase in ion current, while the change in the degree of vapor ionization will depend on the ratio of the rates of change of discharge current and vapor pressure, the fraction of gas ions in the plasma will also change. Studies of the dynamics of pulse evaporation in SHHC-discharge and the influence of RPM parameters on the degree of ionization and plasma composition have not been carried out before.

Methods of direct measurement of the fraction of metal ions in the vapor stream are based on measuring the mass gain on the collector after the metal stream has passed through an ion-retarding electric field [2,3] or an ion-deflecting magnetic field [10] and comparing the results with the value obtained without vapor stream filtration. However, the need for two measurements increases the measurement error.

The effect of the parameters of the RPM-discharge with SHHC and thermally insulated anode-crucible at a constant average discharge current on the rate of Al film deposition, the ionic current density from the plasma, and the degree of ionization of the metal vapor flux was investigated. Plasma parameters are measured and a device with a retarding electrostatic field and a method for determining the fraction of metal ions in the vapor stream through single measurements are described.

The electrode system of the discharge contains coaxially placed hollow cathode and anode-crucible, installed at a distance of 250 mm (Fig. 1). By changing the average current value in the anode-crucible circuit, a temperature of the vaporized substance and vapor flow. The cathode ($d = 6$ mm, $l = 50$ mm) was made of TiN, the heat-insulated crucible ($d = 14-20$ mm, mass $20-30$ g), in which pellets of vaporized metal (Al, 4 g) were loaded, was made of graphite. A two-electrode grid probe (TGP) for measuring the degree of vapor flow ionization (DI) was placed at a distance of 100 mm from the crucible at an angle

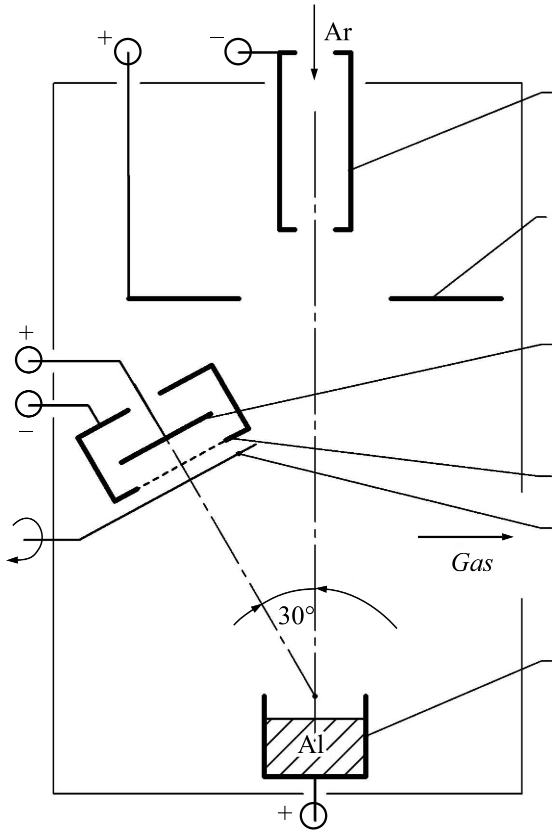

Figure 1. Experiment setup: self-heated cathode (*1*), crucible (*2*), auxiliary anode (*3*), two-electrode grid probe including grid (*4*) and collector (*5*), shutter (*6*).

of 30°. The TGP electrode facing the plasma was made of a woven stainless steel mesh with a cell size in the light $30 \times 30\,\mu$m and a geometric transparency of 0.3; a stainless steel disk located at a distance of 2 mm from the grid served as the collector; the size of the input aperture of the device was 20 mm. This technique is used to measure the fraction of ions in the vapor flow rather than the degree of plasma ionization, since this is the key parameter in the process of film deposition.

The gas pressure in the volume was 0.4 Pa with Ar flow through the SHHC equal to 100 cm^3/min. A gas discharge with a frequency of 36 kHz and a pulse current of up to 14 A to an unevaporated anode located near the cathode was used to heat up the hollow cathode and accelerate the development of the pulse discharge with SHHC. The plasma and vapor flux parameters were measured after the cathode reached the operating mode and the crucible was heated up to $\sim 1200\,^\circ$C.

The discharge was operated in the RPM with an average discharge current of 12–16 A, the constancy of which $I_{av} = I_d f \tau$ was provided by simultaneous variation of the amplitude current I_d within 30–200 A and repetition frequency $f = 60{-}450$ Hz at a fixed pulse duration τ. The dependences of the RPM discharge voltage on the amplitude current at $I_{av} = \text{const}$ are linearly increasing [11]. Without Al loading into the crucible, the value of U is ~ 160 V for discharge in Ar with current 150 A, $f = 140$ Hz, $\tau = 0.6$ ms, in the mode with Al vaporization at the same discharge parameters $U$is lower by 40–60 V.

The pulse heating power of the material is defined as $I_d(U_a + 2kT_e/e + \varphi/e)$ [8], where φ — anode material work function (~ 4 eV for Al), U_a — anode potential drop, T_e — electron temperature. The estimates show that for a fourfold increase in current I_d (50–200 A) the pulsed thermal power at the anode increases by a factor of ~ 10. Variation of substance surface temperature under short-term high-energy exposure with power density F without regard to power lost to evaporation is described roughly by the following ratio [12]: $\Delta T = (2F/\lambda)(a\tau/\pi)^{1/2}$, where τ — pulse duration, λ — thermal conductivity, a — thermal diffusivity coefficient. At an anode power density of 6.6 kW/cm^2 and $\tau = 1$ ms the increase in ΔT is $\sim 100\,^\circ$C, which provides approximately a threefold increase in saturated vapor pressure over the pulse time at an average crucible temperature of $1200\,^\circ$C [13].

The electron temperature determined by the double probe method was 8–10 eV. In this method, the derivative of the probe characteristic is used to estimate $T_e$at a floating probe potential when the balance of currents per probe is provided by fast electrons. High T_e may be due to the presence of electron flux with energy $\sim 10{-}20$ eV [14] in the plasma of high-current discharges with SHHC. The double probe was mounted at a distance of 5 cm from the crucible with a 2.5 cm offset relative to the axis. Measurements were performed at $I_{av} = 10$ A and $\tau = 0.6$ ms. Value T_e increased as I_d increased from 50 A ($f = 350$ Hz) to 200 A ($f = 100$ Hz) and decreased as the probe was moved toward the crucible.

By selecting approximating functions for the probe characteristics of the single probe and their double differentiation, the value of the anodic potential drop U_a, was determined, which increased from +1.5 to +9.5 V as I_d increased from 50 to 200 A.

The total ion current from the plasma I_i estimated from the current in the circuit of the cylindrical electrode ($d = 160$ mm), inside which the discharge system was placed, was up to 20% of I_d (Fig. 2). The deposition rate of AI-films s at a constant average RPM-discharge current of 12 A increased linearly with increasing I_d (Fig. 2).

The usual way of using the TGP (Fig. 1) for measuring DI is to measure the collector mass gain without applying voltage between the electrodes and with ion cutoff. The DI value is calculated based on the results of measurements in these two modes. The method has an upper limit on the ion current density measurements, which is due to the disruption of the continuity of the space charge layer at the grid and depends on the grid cell size, so the DI measurements in the high-power ICP MS device were limited to a power level of 5 kW [3]. Another limitation is due to the occurrence of a virtual anode in the gap between the electrodes. The Bursian [15] j_b current density at which the virtual cathode occurs is $8j_c$, where j_c — the current density calculated based on the Child–Langmuir law. For Al^+ ions with a flux density of 25 mA/cm^2 accelerated in the ion layer at the grid to an energy of 40 eV, the thickness of the ion layer is ~ 0.1 mm, while the grid–collector gap should be < 0.3 mm long.

The proposed improvement of the method consists in measuring the mass gain of the grid and collector in one experiment with ion cutoff, which, given the known geometric transparency of the grid α allows calculating the DI value. This approach allows reducing the energy and increasing the density of measured ion fluxes.

According to the results of measuring the values of grid mass gain M_g and collector mass M_c the DI value was determined as $\{M_g - M_c[(1-\alpha)/\alpha]\}/(M_g + M_c)$. The measurements were carried out using Class I scales with a discreteness of 0.1 mg. To improve the accuracy of measurements, the exposure time of TGP in the plasma flow was increased to 3 h. The dependence of the fraction of Al^+ ions in the vapor flow on the discharge current amplitude at an average current of 12.6 A is shown in Fig. 3 (curve *1*). At $I_d \geqslant 80$ A the degree of flux ionization reaches 100%.

The results of calculating the pulse current density of metal ions I_m^+ by the value of the grid mass weight gain minus the contribution of neutral atoms $I_m^+ = e\{M_g - M_c[(1-\alpha)/\alpha]\}/(M_i t\tau f)$, where M_i — ion mass [kg], t — measurement duration, e — elementary charge, are shown in Fig. 3 (curve *2*). The total current density of gas and metal ions I_t^+ was determined by the value of the current on the negatively biased (−100 V) TGP collector without a grid. The obtained dependence of I_t^+ on I_d is shown in Fig. 3 (curve *3*). The dependence of the gas ion current density I_g^+, calculated as the difference $(I_t^+ - I_m^+)$ for curves *2* and *3*, is shown by the curve *4* in Fig. 3.

The fraction of metal ions in the total ion flux increases monotonically with current I_d and reaches $\sim 30\%$ at

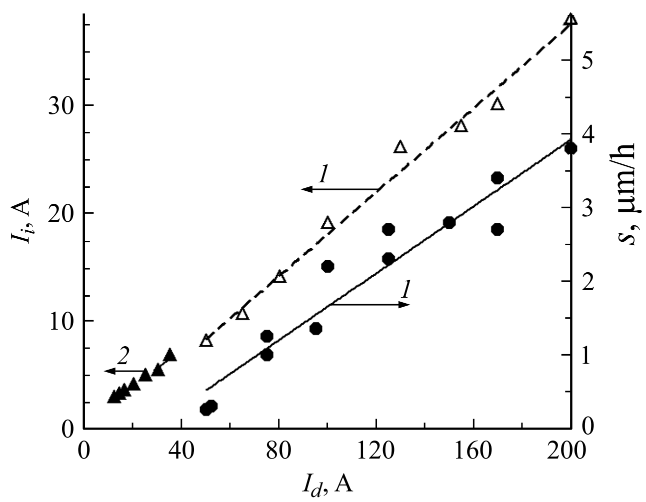

Figure 2. The dependence of the total ion current from the plasma I_i and the rate of aluminum film deposition s on the discharge current. *1* — in pulse-periodic mode, *2* — in continuous mode. The average discharge current is 12 A. In continuous mode, $s = 0.5\,\mu$m/h.

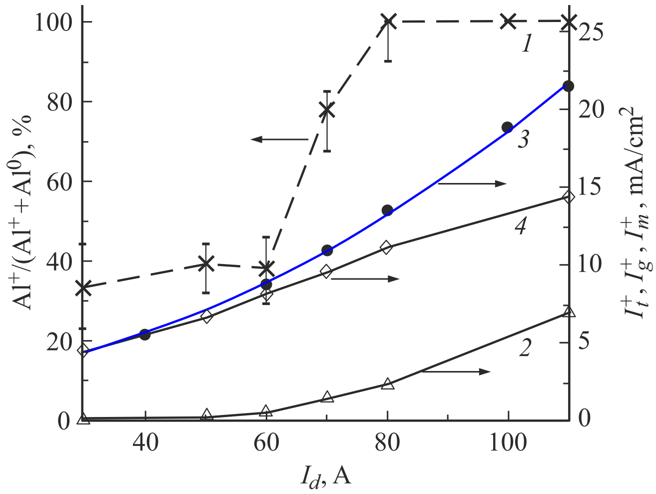

Figure 3. Dependence of the fraction of ions Al^+in the vapor flow (*1*), pulse current density of metallic ions I_m^+ (*2*), total ion current I_t^+ (*3*) and gas ions I_g^+ (*4*) on the discharge current amplitude. The average RPM current of the discharge is 12.6 A.

$I_d = 110$ A. At $I_d = 80$ A the DI value reaches 100%. The increase in the metal ion current at 100% ionization is due to the increase in the vapor pressure with the increase in I_d.

Thus, by changing the parameters of the repetitively-pulsed discharge mode with a self-heated hollow cathode and an evaporated anode, the degree of ionization of the metal vapor flow and the generation of fully ionized vapor flows at relatively small average discharge currents (12–16 A) are controlled. The rapid growth of the melt surface temperature during the pulse and the increase in the saturated vapor pressure lead to a multiple increase in the coating deposition rate compared to the continuous discharge mode at the same average current. Adaptation of the method for measuring the degree of ionization of metal vapor flows by a grid probe based on the measurement of the mass gain of two probe electrodes under conditions of ion flow cutoff to the collector provides measurements in dense flows of low-energy ions.

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Conflict of interest

The authors declare that they have no conflict of interest.

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