Discharge with a self-heated hollow cathode and a vaporizable anode in an inhomogeneous magnetic field

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Abstract. This work is devoted to studying the conditions of the stable burning of discharge with self-heated hollow cathode and anode placed in an inhomogeneous magnetic field. Such a discharge generates dense plasma in Ar-O\textsubscript{2} mixture, and compression of the discharge column by magnetic field allows efficient evaporation of the anode material. A new method of deposition of oxide coatings through reactive anode evaporation was implemented in such a system. The rate of this process, unlike the reactive magnetron sputtering, is not limited by the low-rate sputtering of oxidized target. In addition, this method provides intense ion bombardment of a coating surface. The terms of suppression of ion-sound oscillations have been determined, and a stable burning of discharge with current up to 40 A at a pressure of Ar-O\textsubscript{2} mixtures 0.1 Pa has been achieved. The results of the probe diagnostic of discharge plasma parameters are presented. Nanocrystalline Al\textsubscript{2}O\textsubscript{3} coatings have been deposited in the studied system on the area of 300-600 cm\textsuperscript{2} with rate 2-5 \textmu m/h at temperatures of 400-600\textdegree C.

1. Introduction
Deposition of thin films of metal oxides through reactive evaporation has a number of advantages over the ion sputtering method: a possibility to reach a high evaporation rate and the absence of local arcs on the molten surface. An effective method of deposition Al\textsubscript{2}O\textsubscript{3} films is the reactive electron-beam evaporation of Al [1]. The films were subjected to intense ion bombardment in order to reduce their crystallization temperature [2]. A known drawback of this method is the use of electron-beam guns that emit x-rays and require high vacuum pumping systems. Intense evaporation of metals combined with a high degree of vapor ionization can also provide a discharge with vaporizable anode [3]. Discharge with thermoemission cathode is the most widely used for these purposes; however, the possibility of oxide coatings deposition in such discharge is limited due to the fast degradation of hot cathodes [4].

The oxygen-containing plasma is successfully generated by a self-heated hollow cathode discharge (SHHC) in a flow of inert gas during reactive electron-beam deposition of Al\textsubscript{2}O\textsubscript{3} films [2]. The high density of power (up to 10\textsuperscript{5} W/cm\textsuperscript{2}) released on the anode at small electrode gap 1-5 cm allows to use the discharge for melting and welding of metals [5]. However, the use of SHHC for high-speed oxide coating deposition on large surfaces with intense ion assistance requires an increase in electrode gap length, which leads to the reduction of the power density at the anode and sharp decrease of a metal evaporation rate.

Preliminary experiments on the deposition of oxide coatings in SHHC with remotely located evaporable anode were carried out in electrode system with thermally insulated hollow anode-crucible located at a distance of 25 cm from the cathode, and the water-cooled hollow anode of the ionization...
system, through which O\textsubscript{2} was supplied into reaction volume [6]. A small portion of SHHC current (3–4 A) ensured the evaporation of the Al (1 g) placed inside the cavity of graphite crucible. A large part of the SHHC current (10-30 A) was focused on the anode of the ionization system, providing dissociation and ionization of oxygen particles and high density of ion current on the coating surface. In such a system, nanocrystalline Al\textsubscript{2}O\textsubscript{3} films were obtained at the rate up to 4 µ/h, and the parameters of ion flow required for the formation of single-phase α-Al\textsubscript{2}O\textsubscript{3} coating at 600°C were determined [6].

In this work the SHHC with water-cooled hollow anode-crucible placed in an inhomogeneous magnetic field was investigated. Forced cooling requires to increase heating power of the crucible, but allows avoiding problems caused by the interaction of molten Al with crucible material. The necessary high power density at the anode provides with a compression of discharge column in the non-uniform magnetic field.

The aim of the research was to study the conditions of stable burning of SHHC with remotely located evaporable anode placed in strongly inhomogeneous magnetic field, to determine the discharge plasma parameters, to measure the spatial distributions of particle flows in the volume, and to define the deposition rate of nanocrystalline Al\textsubscript{2}O\textsubscript{3} films at a low temperatures (400-600°C).

2. Experimental technique
A tubular cathode \textit{1} with an internal diameter of 12 mm made by magnetic pulse pressing of TiN powder [7] was located at a distance of 250 mm from a water-cooled hollow anode-crucible \textit{2} made of stainless steel and located inside short magnetic coil \textit{3} (figure 1). Induction of the magnetic field on the coil axis reached 20 mT at coil current 10 A. Crucible diameter was 25 mm, volume – 6 cm\textsuperscript{3}. Crucible was filled with Al granules. Ar (10-20 sccm) was supplied through the cathode cavity and O\textsubscript{2} (30 sccm) fed into the anode area of the discharge. Pumping was done with the turbomolecular pump, the working pressure of the gas mixture was ~0.1-0.15 Pa.

![Figure 1. Experimental scheme](image)

Energy spectrum of fast electrons from the hollow cathode was measured through retarding potential technique using triple-grid energy analyzer with output disc-shaped collimator. Interception of electrons with large lateral velocity component by collimator provides analysis of the longitudinal velocity of electrons and reduces the influence of space charge of electrons in the retarding field. The energy range of the analyzer was limited at ~10 eV. Plasma diagnostics of the SHHC near anode area was done using a single collecting probe and a dual Langmuir probes.

Profiles of the current density of electrons and ions and the floating potential of the probe were measured with moving Langmuir probe 0.2 mm in diameter, which moved in a plane distanced from the anode by 10 mm. Spatial distribution of a metal vapor flow was determined according to the rate of deposition of films onto the samples located at different distances from the anode-crucible. Dependencies of ion current density on discharge current and magnetic field were measured using the sample holder \textit{4} with the area of 30 cm\textsuperscript{2} installed at a distance of 70 mm from the crucible. Heater \textit{5} located on the back side of the holder provided the radiation heating of samples up to 400-600°C.

Microhardness of the films was determined with ultra-microhardness tester DUH-211/211S (Shimadzu), thickness of the films was measured through the ball abrasion technique on the unit
“Calotest” (CSM Instruments), XRD analysis was conducted using X-ray diffraction meter XRD-7000 (Shimadzu).

3. Experimental results

Energy spectra of fast electrons for the hollow cathode discharge currents 5-25 A are shown in figure 2. Distribution maxima are shifted into the lower energy levels with increasing currents, which can be attributed to the reduced voltage of discharge burning (figure 3).

![Figure 2](image1.png) ![Figure 3](image2.png)

Figure 2. Energy spectra of the electrons flow from the hollow cathode. SHHC current: 1 – 5, 2 – 10, 3 – 25 A.

Figure 3. Dependencies of discharge burning voltage $U$ and electron energy corresponding to the maximum of the energy spectrum $E_m$ on the discharge current $I$.

Voltage-current characteristics (VAC) of the collecting probe obtained in the range of magnetic field induction 0–20 mT were qualitatively identical. Electron temperature estimated according to the slope angle of the straight-line segment of the probe semilog scale VAC decreased from ~6 to ~4 eV along with increasing of arc current from 10 to 40 A. Measurements of the anode potential drop through the dual probe technique were carried out in the absence of metal vapor. With current more than 30 A the positive anode potential drop was a little over 40 V (figure 4).

Radial profiles of electron current on the probe were obtained at the arc current 40 A, magnetic coil current 0 and 10 A under conditions of intensive evaporation of Al (figure 5). Full width at half height of profiles under application of the magnetic field reduces from 7.5 to 2.6 mm. The presence of metallic vapor ensures compression of the discharge column by magnetic field for all values of the discharge current, whereas in absence of metal vapor compression was observed only at currents no more than ~10 A.

The floating potential of the probe on the discharge axis in the absence of metal vapor was modulated by ion-sound oscillations (30-120 kHz), amplitude of which increases up to 220 V along with increase in discharge current. Increase of magnetic field up to 20 mT results in fluctuation amplitude drop to ~40 V at discharge current 40 A (figure 6). In evaporation mode at discharge currents over 20 A potential fluctuations are absent, at discharge current 40 A the floating potential is about 30 V.

Dependencies of average ion current density from SHHC current were measured in the sample holder after supply of the pulsed negative bias to the collector (200 V, 50 kHz). The highest average density of ion current in evaporation mode at the maximum magnetic field amounted to 5 mA/cm$^2$ (figure 7).

The spatial distribution of the metal vapor flow was estimated according to the deposition rate of amorphous Al$_2$O$_3$ films onto the stainless steel samples with dimensions 100×10 mm at a low temperature (200°C) and low (~50 V) bias potential (figure 8). Samples were installed vertically on the distances 70, 110 and 150 mm from the discharge axis. Axisymmetric placing of samples at radial...
distance 150 mm can give a coating with uneven distribution of thickness of ~20% on the area of ~600 cm² at coating rate up to 0.4 µm/h.

![Figure 4](image1.png)  ![Figure 5](image2.png)  ![Figure 6](image3.png)

**Figure 4.** Dependencies of the anode potential drop on the magnetic coil current. SHHC current: 1 – 10, 2 – 20, 3 – 30, 4 – 40 A.

**Figure 5.** Radial distribution profiles of the electron current. SHHC current 40 A. Magnetic coil current: 1 – 0, 2 – 2, 3 – 8, 4 – 10 A.

**Figure 6.** Dependencies of the fluctuation amplitude of the floating potential probe on discharge axis with evaporating – Al (1, 2) and non-evaporating – C (3) anode. SHHC current: 1 – 10; 2, 3 – 40 A.

Dependency between the heat output $W_1$ due to water cooling of the anode-crucible walls and the full power $W_d$ consumed by the discharge is shown in figure 9. The value of $W_1$ was measured through calorimetric method, the full power $W_d$ was calculated based on the values of SHHC current and burning voltage. Dependency $W_1 = f(W_d)$ obtained at constant voltage of discharge burning 60 V is close to linear and $W_1 \sim 0.45 W_d$. Stable burning of discharge with voltage 60 V in the Al evaporation mode was sustained at minimum Ar flow through the cathode cavity of 10 sccm. The aluminum melting zone had a diameter of 6-8 mm, the surface of melt was stable during evaporation process, and there were no bursts of liquid metal.

$\text{Al}_2\text{O}_3$ films were deposited to the stainless steel sample mounted at a distance of 70 mm from the anode at a temperature of 400 °C, discharge current 40 A and bias voltage 40-100 V. The average density of ion current was 5 mA/cm², the flow rates of $\text{O}_2$ and Ar – 50 and 10 sccm, respectively. Coating deposition rate in this mode was 4 µm/h. Diffraction pattern of the coating contains peaks of nanocrystallites $\alpha$-$\text{Al}_2\text{O}_3$ with size 7 nm, and peaks of $\alpha$-$\text{Al}_2\text{O}_3$ with a fraction of ~5%. (figure 10).

4. **Discussion**

The occurrence of the ion-sound oscillations is characteristic of SHHC, different conditions resulted in build-up of oscillations of plasma potential with increasing current [9] and in stabilization of discharge with current growth [5]. In our experiments, increase in discharge current with TiN cathode over 20 A,
compression of the discharge with magnetic field in the near anode area and the presence of metal vapors in the discharge gap stabilized the discharge, which is explained by increased ion flow into cathode discharge area.

**Figure 7.** Dependencies of the ion current density in discharge burning mode with evaporating – Al (1) and non-evaporating – C (2, 3) anode from the discharge current. The magnetic coil current: 1, 2 – 10; 3 – 0 A.

**Figure 8.** Dependencies of coating thickness on vertical samples from the distance to the anode plane. SHHC current 40 A. Magnetic coil current 10 A. Distance from the anode axis: 1 – 70, 2 – 110, 3 – 150 mm.

**Figure 9.** Dependencies between the power retracted by water from the anode-crucible walls and the total power consumed by the discharge. The magnetic coil current: 1 – 0, 2 – 10 A.

**Figure 10.** XRD spectra of Al₂O₃ coating obtained at a temperature of 400°C, discharge current 40 A and offset voltage 40-100 V.

The discharge column is compressed in curvilinear magnetic field, because when the Larmor radius of electrons and screw trajectory pitch are considerably smaller than the area of the filed heterogeneity, the electron motion occurs as superposition of the Larmor rotation, directional movement along the lines of the magnetic field and the azimuth drift across the magnetic field [10]. For example, the Larmor radius of the electron with thermal energy ~5 eV in the induction field 20 mT is about 0.4 mm.

In order to assess thermal flux from plasma to anode at changing of discharge current in a wide range, the term “volt-equivalent” is used. It is defined by the ratio of power $W_a$ released at the anode and the current to the anode $I$. $W_a$ is determined by the known relation:

$$W_a = I(U_a + \phi/e + \epsilon/e),$$
where $U_a$ is the positive anode drop, $\phi$ – cathode material work function, $\bar{\varepsilon}$ – average energy of thermalized electrons. The measured value of volt-equivalent with accuracy to $\phi$ was equal to $U_a$ in vacuum arcs with evaporating hot anode [11]. In contrast, for SHHC where energy distribution of electrons in plasma differs significantly from the Maxwell model, the anode heating was provided mainly by fast electrons [5].

In our experiments, the loss of $W_1$ as a result of heat removing by water accounted for 45% of $W_d$. According to [3], melting and evaporation loss at power density over 1 kW/cm$^2$ constitute 30–45% of $W_d$. Ignoring radiation losses, the power released at the anode $W_a \sim 75$–90% of $W_d$. So, at a discharge voltage of 60 V the volt-equivalent was 45-54 V. Even taking into account the existing of fast electrons with an energy of $\sim 10$ eV in the energy spectrum, the energy balance at the anode holds for the presence of the positive anode drop of potential $\sim 20$–30 V.

Numerical calculation of the temperature regime of the vaporized aluminum target in the water-cooled crucible was conducted using ELCUT program [14] and experimental electron density profiles. Integration of electron current profile has shown that the power density in distribution maximum at the discharge current 40 A and the maximum magnetic field is close to 1 kW/cm$^2$. Calculations have shown that in the absence of the magnetic field, the temperature of Al surface is insufficient for its efficient evaporation. Application of the magnetic field, the maximum calculated temperature exceeded 1000°C, which provides the experimentally achieved rates of Al evaporation and deposition of coatings.

It has been shown [7] that the formation of nanocrystalline single-phase $\alpha$-Al$_2$O$_3$ film at low temperature (600°C) requires ion current density 2–8 mA/cm$^2$ and ion energy 25-200 eV on the film surface. The ratio of ion fluence $F$ and the number of particles $N$ forming the unit area coating must be $\sim 1$–2. Calculations show that if the average density of ion current is 5 mA/cm$^2$ and $F/N \sim 2$, the rate of nanocrystalline coating formation can reach 5 µm/h, which was experimentally confirmed.

5. Conclusion
It has been shown that arc discharge with self-heated hollow cathode and remotely located anode-crucible placed in an inhomogeneous axisymmetric magnetic field stably operates in Ar-O$_2$ mixtures at currents up to 40 A and gas pressure 0.1 Pa. Power density on the surface of the anode at the discharge compression by magnetic field reaches 1 kW/cm$^2$, which ensures the effective evaporation of aluminum from the water-cooled anode-crucible. The average density of ion current on the substrates located at a distance of 70 mm from the crucible reaches 5 mA/cm$^2$, which reduces the temperature of coating crystallization. It has been shown that the nanocrystalline coating of aluminum oxide can be obtained in the studied system in the area of 300-600 cm$^2$ at 400-600°C with a rate of 2-5 µm/h.

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