Creating ceramic electrically insulating coating on metal surface

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Abstract. The present work deals with the problem of creating dielectric (ceramic) coating on the surfaces of conductive materials for endowing them with insulating properties. The coatings have been created using the electron-beam evaporation of aluminum oxide ceramics. A plasma electron source operating in the forevacuum range of pressure (5–100 Pa) have been used as a source of electrons. It has enabled an effective transport of the electron beam onto the ceramic non-conductive surface and its melting and evaporation. During this process, the electric charge brought by the electron beam onto the surface of the ceramic target is completely neutralized by the ions of gas discharge plasma generated by the electron beam along its transport path towards the target.

1. Introduction
Metal and dielectric materials, finding ubiquitous applications, are electrically polar substances. Metals are electrically conductive materials with high plasticity and a relatively low melting temperature. Dielectrics, such as ceramics, as they lack free electrons in the conducting band, are characterized by high electric insulating and thermal conductive properties. Additionally, thanks to their specific structure, they exhibit an enhanced brittleness and high melting and evaporation temperatures. The enticing idea of creating dielectric materials with such opposing properties explains the undertaking of the present investigation.

For evaporated targets, we used pressed alumina powder (Al₂O₃) with the particle size of about 100 µm that included modifying additives, such as talc (Mg₃Si₄O₁₀(OH)₂) and magnesia powders (MgSO₄) with the particle size 30–50 µm. The main reason for using these modifiers was to compensate for the difference in the thermal expansion between the substrate material (copper, stainless steel) and the aluminum oxide being deposited and the substrate.

2. Experimental setup and techniques
The experiments were carried out on a vacuum installation with a forevacuum plasma electron source [1]. The working gas was air of the residual atmosphere at 5–6 Pa. The experimental setup is shown in figure 1.
Figure 1. Experimental setup: 1 – hollow cathode; 2 – hollow cathode plasma; 3 – anode; 4 – perforated electrode; 5 – accelerating electrode (extractor); 6 – focusing coil; 7 – magnetic deflection system; 8 – electron beam; 9 – evaporated sample; 10 – graphite crucible; 11 – sample holder; 12 – sample.

Emission plasma 2 was generated by a glow discharge with hollow cathode 1. Electron beam 8 was formed by extraction electrons from the plasma boundary through the holes in perforated electrode 4 installed in source anode 3. Accelerated electron beam 8 was focused by magnetic system 6 and aimed at target 9 by magnetic deflection system 7. Technical specifications of the plasma electron source are given in more details in [2].

Sample 12, onto which the dielectric film was coated, was fixed in holder 11 so that the distance along the normal line from sample 12 to target 9 did not exceed 50 mm. The angle between the sample surface normal and the beam propagation direction was 45°.

A sintered aluminium oxide with modifying additives was used for the evaporated target. The modifiers were added to the target material to balance the coefficients of thermal linear expansion of the sample material and the dielectric film during the coating process and its service life. The evaporated sample was fabricated by hot pressing at the temperature of 300–400°C. The evaporated sample had a length 10–12 mm and a diameter 10 mm. The following powders were used to make the evaporated samples: 1) talc with the particle size 30–50 µm; 2) pure aluminium oxide with the particle size ~100 µm; 3) aluminium oxide with talc additives of 10, 30, and 50 wt.%; 4) aluminium oxide with magnesium additives of 10, 30, 50 wt.% and the particle size 20–60 µm; 5) a complex composition with talc in the upper part of the evaporated target (figure 2).

Figure 2. Scheme evaporated sample.
Additionally, we investigated the samples with thermally sprayed coatings of aluminum and titanium (0.6–1 µm thick) whose coefficients of thermal linear expansion had intermediate values as compared with those of copper, stainless steel and aluminum oxide.

Deposition of the dielectric film on the sample was performed as follows. The focused electron beam was guided to the upper butt of the evaporated target. The evaporation process was preceded by the stages of heating (for degassing) and preliminary sintering of the ceramic target, which were performed by gradual increase of the electron beam power for 20–25 minutes. During the evaporation, the accelerating voltage was 18–20 kV, the beam current was 20–30 mA, and the power was maintained at 450–500 W. The evaporation lasted for 30–60 minutes. The electron beam diameter at the target upper butt was fixed using a single magnetic lens and sustained so that the forming melting pool, from which the evaporation occurred, took up the entire upper part of the target. As a result, the growth rate of the dielectric film was from 35 to 160 nm·min\(^{-1}\).

After depositing the dielectric film on the sample by vacuum thermal spraying, there were deposited aluminum electrodes, ~ 1 µm thick. A layout of the obtained sample is shown in figure 3.

![Figure 3. Layout of the sample and its view from above.](image)

After the sample fabrication, the following investigations were conducted: 1) SEM Hitachi S-3400N examinations of the surface morphology and elemental composition of the deposited dielectric films; 2) thickness measurements of the deposited films using a micro-interferometer MII-4 (for thickness 0.1 to 0.8 µm) and an interference microscope profilometer MNL-1 (for thickness up to 50 µm); 3) dependence of capacitance, volume and surface resistance, imittance, and the loss tangent on frequency using an LCR-Meter AMM-3148 (frequency range from 50 Hz to 100 kHz) and LCR-meter E7-29 (frequency range from 50 kHz to 50 MHz).

3. Experimental results

Figure 4 shows the experimental results of the surface morphology of the deposited dielectric films. Figure 5 shows the elemental composition of the created films.

Durability of coatings is one of its important characteristics and one of the main factors that determines this property of coating is adhesion. There are many methods for measurements of adhesion between films and their substrates. The grinding technique [3–4] is among the simplest qualitative methods to do so. The film is being removed from the surface by grinding. The film resistance is determined by the results of its processing by a sandpaper. To obtain a qualitative estimate, we used a sandpaper P2500 with the grain size 3–5 µm. As a result, the films of pure aluminum oxide deposited onto aluminum and titanium sublayers, as well as the films of complex composition, have proved to have the highest adhesion properties (resistance to wear). Comparison between stainless steel and copper substrates has shown that the films on stainless steel are more wear resistant.
Figure 4. The surfaces of the films deposited onto stainless steel. a) Talc, film thickness ~ 1 µm. b) Aluminium oxide with a magnesium additive of 10 wt.%, film thickness ~ 2 µm. c) Titanium sublayer, ~ 1 µm, and pure aluminium oxide, film thickness less than 1 µm. d) Complex composition: talc, thickness ~ 1 µm, talc + aluminium oxide and pure aluminium oxide, total thickness ~ 6 µm. A droplet of sintered dielectric in the right-upper corner.

The electrical measurements of the deposited films included measurements of dielectric permeability $\varepsilon$, the loss tangent $\tan\delta$, volume $\rho$ and full (emittance) $Z$ resistance as functions of frequency. The results are shown in figures 6 and 7.

4. Discussion
In order to produce highly insulating coatings on metal surfaces, it is required to create a solid, non-porous dielectric film with good adhesion properties and a high specific electrical resistance. The investigations carried out previously [5] have demonstrated that the deposited alumina films prove to be sufficiently uniform and non-porous, but the entire surface is covered with micro-cracks created during the growth of the dielectric film at the substrate temperature about 700°C.

We believe that the micro-cracks appear due to different coefficients of thermal linear expansion of the substrate material and the material of the deposited film. In our experiment, we used substrates made of stainless steel 12X18H9T and electrochemical copper. The thermal expansion coefficients of these metals at the temperature 100°C are $16.6 \times 10^{-6} \text{ K}^{-1}$ (stainless steel) and $17.18 \times 10^{-6} \text{ K}^{-1}$ (copper). This coefficient is less for aluminum oxide, $(7–10) \times 10^{-6} \text{ K}^{-1}$ [6]. This difference brings about mechanical stress in the dielectric film and induces micro cracking.

As known [7, 8], the use of talc as an additive to ceramics decreases its brittleness and inhibits cracking. The use of talc as a sublayer between the metal substrate and the alumina film with a smooth talc-alumina transition boundary enabled us to obtain a solid dielectric coating on the metal surface without formation of micro cracking. It was possible due to unique properties of talc: low hardness, plasticity, high thermal expansion coefficient, and lower melting temperature (1547°C) [7] as compared with aluminium oxide (2327°C) [6].

Measurements of capacitance (relative dielectric permeability), volume and full (emittance) resistance, and the loss tangent as functions of frequency in the range from 50 Hz to 50 MHz have shown the following: a) the volume resistance has proved to be several orders of magnitude less than the known values; b) the order of magnitude of the loss tangent is two or three times greater than that of the alumina
films [9]. The dielectric losses are important electrical characteristics of dielectric materials. The losses depend on the nature of dielectric defects and the types of additives. The influence of reach-through conductivity is especially noticeable at lower frequency. There are also observed relaxation losses. Figure 8 shows dependence of the loss tangent on the frequency logarithm. In the frequency range 50–100 kHz, there is seen a distinct peak, typical for ceramic materials [10].

The presence of excessive reach-through conductivity in the dielectric film can be caused by several factors. It has been established in [11] that during alumina evaporation, a vapour over the melting pool contains gases of $\text{Al}, \text{O}, \text{Al}_2\text{O}, \text{Al}_2\text{O}_2, \text{AlO}_2$, and $\text{Al}_2\text{O}_3$. For this reason, the deposited film may contain a surplus of aluminium. Since the evaporation occurs from the graphite crucible, there is also a possibility of aluminium deoxidization in the reaction with carbon and its subsequent penetration to the dielectric film. The composition analysis of the deposited film (figure 5) shows that the film contains carbon, which can lead to an increased conductivity.

![Figure 5](image_url)

**Figure 5.** Elemental composition of the created films. a) Titanium sublayer, ~ 1 µm, and pure aluminum oxide, the film thickness less than 1 µm. b) Complex composition: talc, thickness ~ 1 µm, talc + aluminum oxide and pure aluminum oxide, total thickness ~ 6 µm.

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5. Conclusion

The investigations have demonstrated a possibility of creating an alumina insulating coating on the metal surface using the electron beam evaporation of a dielectric target by a plasma electron source in the forevacuum range of pressure. This technique enables the dielectric film growth on the metal surface at the rate of 35–160 nm·min⁻¹. Investigations of the surface morphology of dielectrics coated on metal have shown that deposition of pure aluminium oxide produces strong mechanical stress in the film that results in the appearance of micro-cracks. The use of talc powder as a modifier in the evaporated target has allowed obtaining a solid (without micro-cracks) dielectric film on the metal surface with good adhesive properties.

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