Erosion of the sputtered electrodes in the surface barrier discharge

I Selivonin\textsuperscript{1,2}, A Lazukin\textsuperscript{2}, I Moralev\textsuperscript{1,2}, S Krivov\textsuperscript{2} and I Roslyakov\textsuperscript{3}

1. Joint Institute for High Temperatures of the Russian Academy of Sciences, Izhorskaya 13 Bldg 2, Moscow 125412, Russia
2. National Research University “Moscow Power Engineering Institute” Krasnokazarmennaya 14, Moscow 111250, Russia
3. Lomonosov Moscow State University, Leninskie Gory 1, 119991, Moscow, Russia

E-mail: inock691@ya.ru

Abstract. The paper is devoted to a phenomenological study of the modification of sputtered electrodes in a surface dielectric barrier discharge in atmospheric air. A decrease in the power supplied to the discharge in the case of using platinum and aluminum electrodes and an increase in the case of using a copper electrode were demonstrated. It is shown that during long-term operation of the electrode system, a decrease in its cold capacitance is observed. These effects are assumed to be due to the degradation of the electrode material, transfer of erosion products to the surface of the dielectric, as well as morphological changes at the electrode edge.

1. Introduction
The dielectric barrier discharge (DBD) is widely used in various applications, for example, surface treatment \cite{1,2}, plasma chemistry \cite{3–5}, plasma medicine and biology \cite{6,7}, plasma aerodynamics \cite{8–11}. In a variety of technological problems, two basic types of electrode configurations are used: with a pair of dielectric-covered electrodes (co-planar barrier discharge) and with one electrode exposed directly to the discharge (surface barrier discharge) \cite{12}.

One of the most important requirements for devices based on DBD is the long-term stability of their operational characteristics. The papers dedicated to the development of long-operated devices based on DBD describe two main processes of degradation of the electrode system: destruction of the dielectric barrier \cite{13,14}, and the erosion of electrodes, the latter taking place in surface configuration (sDBD) \cite{15–17}.

As was shown in \cite{16,17}, at long exposures foil electrodes significantly change their morphology. It was demonstrated that the degradation of the electrodes leads to a change in the integral characteristics (energy dissipated in the discharge, input power), determining the performance of the devices based on the barrier discharge. More subtle characteristics, such as the mode of the discharge operation and microdischarge statistics, are affected as well.

The rate and scenario of electrode degradation strongly depends on electrode thickness. In \cite{15}, the processes of degradation of the thin film deposited electrodes were considered. It has been shown that even with the use of oxidation-resistant materials, intense destruction of electrode edges is observed, which leads to the to a change in the capacity of the discharge cells and dissipated power level.
Thus, the correct choice of the material of the exposed electrode and the method of its manufacture is an important task in the development of devices based on sDBD, regardless of their intended purpose. Research in this direction will allow to create stable long-operated sDBD arrangements or, at least, to predict changes in their properties during the long-term operation.

2. Experimental setup and measurement methods

In this work, the classical configuration of the edge-plane surface barrier discharge was studied. The configuration and measurements organization are shown in Figure 1. The sputtered strips of aluminum, copper, and platinum were used as corona electrodes. The thickness of the electrodes was 0.5–2 μm. Before the exposure began, the edge quality was monitored visually using an MBS-10 optical microscope with a magnification of 100x. Alundum ceramics with a thickness of 1 mm and a dielectric constant of 10.4 was used as a dielectric barrier. Earlier experiments have shown that dielectric material is immune to plasma treatment in a barrier discharge. The buried electrode was sectioned in order to more accurately measure the electrical characteristics of the discharge.

The electrode system was powered by AC sinusoidal voltage with a frequency of 100 kHz and an amplitude of 3.4 kV. Exposure was carried out for 6 hours in room air at atmospheric pressure. Venting of the volume was organized to remove the chemical products of the discharge.

To measure the electrical characteristics of the DBD, the volt-coulomb curves (VCC) were analyzed. The method was described in detail in [18]. To exclude capacitive current through the electrode system from the measurements, a Wheatstone capacitor bridge was used, in the upper arm of which a tunable vacuum capacitor \( C_c = 3–50 \text{ pF} \) was installed. The bridge was balanced at the low voltages prior to the experiments. Measurement of the voltage on a capacitor \( C_m \), proportional to the charge transferred onto the dielectric barrier, was made with a differential voltage probe Pintek DP-150 (accuracy 5%, bandwidth 150 MHz). The high voltage measurement was made using a Tektronix P6015A high voltage probe (5% accuracy, 75 MHz bandwidth). The data were collected using an oscilloscope Tektronix TDS2024b with a bandwidth of 200 MHz. Recorded data were averaged across 128 periods of the supply voltage.

The energy dissipated in the discharge during one voltage period can be determined by estimating the VCC area. The cold capacitance of the discharge arrangement can be calculated as \( C_{cold} = dQ/dU \) in a silent part of the VCC when the discharge does not operate. Compensation procedure before the start of exposure led to exclusion of the displacement current, associated with \( C_{cold} \), from the measurements. Thus, the VCC had a zero slope at the silent part of the cycle (Fig. 2).

This method allows to obtain the values of the power dissipated into the discharge with an accuracy of 1 mW/mm, as well as to control changes in the discharge arrangement capacity without the discharge (cold capacitance) with an accuracy of 0.1 pF.
To obtain pictures of the morphology of the electrode edges, Olympus Lext OLS4000 laser confocal microscope with a spatial resolution better than 500 nm was used.

![Figure 2. VCC evolution during sDBD operation](image)

3. Results and discussion

During the first hour of operation, a decrease in the discharge power to the level of ~ 60% of the initial value is observed on the aluminum electrode (Fig. 3, left). Such a change in power is accompanied by a significant decrease in the cold capacity of the discharge cell (Fig. 3, right). The reduction of the cold capacity reaches 0.013 pF/mm after 2 hours of cell operation while the initial capacity of the electrode system is 0.5 pF/mm electrode length. In the case of using a platinum electrode, similar trends are observed, although less pronounced. In the case of a copper electrode, an increase in the power deposited in the discharge reaches 30-50% of the initial value. No change in the capacity of the discharge system is observed.

While the power dynamics qualitatively looks the same as in the case of foil electrodes [16,17], a change in the capacity of the discharge cell is a new effect, appearing only for thin exposed electrodes. The decrease in the cold capacity of the electrode system is obviously associated with a decrease in the area of the exposed electrode as a result of the effect of the sDBD plasma. Knowing the change in the capacitance of the electrode system and assuming that electrode material is removed from the surface or replaced by the oxide layer, one can estimate the width of the oxidized strip. To do this, one can use the formula:

\[
\Delta C = \varepsilon \varepsilon_0 S \frac{\Delta d}{d},
\]

where \( \Delta C \) is the reduction in the capacitance of the electrode configuration, \( \varepsilon \) and \( d \) are the dielectric constant and thickness of ceramics, \( S \) is the width of the oxidized strip.

The result of this estimate is shown in Figure 4 on the left. In the case of using an aluminum electrode, an edge degradation is observed at distances up to 140 μm for 2 hours exposure. To verify this result, images of the edge were taken using a confocal microscope (Figure 4, right). They clearly show a blackened oxide band along the entire edge. At the 2 hours exposure time, the width of this band is about 250 μm, that somehow exceeds the estimate obtained from a decrease in the capacity of the discharge system. Obviously, in the blackened area, the electrode oxidizes unevenly; in some areas, oxidation does not occur across the entire thickness of the electrode, and there is a conductive metal layer under a thin oxide layer. This fact is also confirmed visually - during long-term discharge operation, the attachment points of the microdischarges move from the edge of the electrode, however, they are localized in the far region of the oxide band.

In the case of using a platinum electrode, edge degradation can also be counted and observed in images. At 2 hours of exposure, it is about 100 microns. However, the edge patterns obtained with a microscope show that the erosion process of the electrode differs from the processes on the aluminum edge. In addition to the oxidation of the strip, active transfer of erosion products to the surface of the
barrier in the discharge region is observed. As a result of these processes, the properties of the surface of the dielectric change, which also leads, apparently, to the difficulty of developing the discharge.

![Figure 3. The dynamics of the discharge power on the electrodes of various materials (left) and the reduction of electrode system capacitance (right)](image)

**Figure 3.** The dynamics of the discharge power on the electrodes of various materials (left) and the reduction of electrode system capacitance (right)

![Figure 4. Width of the oxidized area of the electrodes (left) and images of the edges of these electrodes at different exposure times (right)](image)

**Figure 4.** Width of the oxidized area of the electrodes (left) and images of the edges of these electrodes at different exposure times (right)

In the case of a copper electrode, no bulk degradation of the electrode is observed. On the electrode edge, craters with a size of the order of 100 μm, evenly distributed along the edge, are formed. These craters correspond to the conservative positions of microdischarges. In the vicinity of these craters, the intensive formation of copper oxides is observed. However, inside the craters the clean nonoxidized electrode material can be seen. Previous studies have shown that cathode erosion cleaning at the locations of stationary microdischarges can be responsible for the formation of the removal of the oxides front the crater center. The initial increase in discharge power seems to be related to the formation of the crater and stabilization of the microdischarges positions. In general, the behavior of the copper sputtered electrode is qualitatively similar to the behavior of the copper foil electrode, which was described in [16].
Summarizing the above, we can describe the processes of modification of the sputter ed electrodes as follows.

In the first tens of minutes, the edge of the aluminum electrode is modified by discharge. At the same time, there significant decrease in power occurs. At the completion of this stage, the edge acquires a certain structure with a gradient of the thickness of the oxide strip. Further, this once formed structure moves into the depth of the electrode. This process leaves the power nearly constant, while reduction of the electrode area reduces the cold capacitance of the system. Obviously, the decrease in power and degradation rate of the edge are determined by electrode thickness.

The modification of the platinum electrode can also be divided into 2 stages - a fast one, in which a structure with an oxide band on the edge and contamination by products of erosion of the barrier surface are formed; and slow, in which the electrode spots of the microdischarges move deeper into the electrode from the edge. Accordingly, during the first stage, the dynamics of discharge power is observed, and during the second one it is stable.

Microdischarges on the copper electrode are initially practically stationary. In the places of attachment, the formation of craters is observed, with their inner surface intensively cleaned by discharge from the oxide layer, which leads to an increase in power. The formation of such a structure leads to the establishment of an even more stable quasi-filamentary mode of discharge operation.

4. Conclusions

An unexpected conclusion of this work is that due to the peculiarities of the processes of oxidation, noble metals are not definitely the best choice of material for the corona electrode.

The durability of the aluminum and platinum electrodes is limited by their oxidizing by the discharge products. In addition, it is necessary to pay attention to the reduction in the capacity of the discharge cell. When using resonant power sources, reducing the load on several pF may be sufficient to significantly change the power supply mode of the cell. Copper appears to be the most resistant of the deposited electrodes. However, it is worth noting that the features of the discharge mode on such an electrode (quasi-filamentation) may strongly affect the applications where the discharge homogeneity is required.

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