Formation of fractal microstructures in conductive layers of indium-tin oxides and zinc oxide

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Abstract. In this paper, we have studied the fractal microstructures that form within conducting layers of oxide compositions and the effects that happen during electrical breakdown. We have determined that application of additional polymer films on top of the layers of oxide compositions allows one to visualize the breakdown processes taking place in the structure. The enlarged "polymer photography" that results in this makes it possible to assess the quality of contact layers without using high-resolution optical equipment.

1. Introduction

Despite positive outlook of the development of flexible electronics and photonics, there are still problems that slow down the progress in this area. In particular, there are difficulties in creating contact areas that can withstand multiple bends. Contacts with a fractal structure can solve this problem. However, the creation of such contacts using conventional methods of microelectronic engineering is a complex and expensive task. Therefore, it was logical to search for a way of creation of this fractal structure during a physical process. The effects that appeared during the flow of high-density currents through conventional film materials made it possible to study the possibility of creation of fractal film micro- and nanostructures. The film materials used for these experiments are normally used in transparent conductive electrode coating and are made on the bases of oxide compositions.

2. Samples and techniques

Structures based on indium-tin oxide and zinc oxide served as test samples (Figure 1). The thickness of the substrate was 0.5 - 3 mm. The thickness of the oxide compositions films varied in the range of 1-10 μm. Some of the samples were coated with organic polymeric layers based on polycarbonate (PC) and polymethyl methacrylate (PMMA) with a thickness of 100-200 nm.

The setup used to study the effects of high density currents (Figure 2) had a nanoelectrode based on the eutectic composition Ga / In. This electrode allowed creation of high local field intensities and maintained diameter of contact area between the electrode and the film close to ~ 60 μm [1].
The electrode was fed with step voltage of positive polarity with an amplitude of 20 to 300 V. A layer of the oxide composition served as the second electrode. The current was limited by a resistor that had its resistance values range from 1 to 20 kΩ. A measuring resistor of 100 Ω was connected in series with the sample to control the amount of flowing current. A digital oscilloscope recorded the time dependences of the voltage drop. The shape and the size of formed microfractal structures were recorded using a digital video camera with a resolution of ~5 μm.

The setup also had a spectrometric unit that was used to measure spectral characteristics of subsequent luminescence. These measurements were performed at high resolution both over the wavelength and in time [2].

Additionally, the setup allowed to carry out experiments with magnetic fields of various intensity.

Finally, created fractal micro- and nanostructures were studied with optical methods using high-resolution microscopes, metallographic computer analysis, and atomic force microscopy.

3. Experiment.

The research of high-density current flow in layers of indium-tin oxide compositions revealed that ITO layer destructions are localized in current channels (tracks).

The shape of these tracks depended on the thickness of the ITO layer. In thin ITO layers (thickness of 100-300 nm), destructions took the form of individual short tracks with a length of 200-400 microns. In films with thickness of more than 300 nm, the tracks had the shape of a multi-turn helix [3]. The shape of the emerging fractal formations was determined primarily by the properties of the oxide layer, in particular, its resistance and crystal structure. Figure 2 shows that the propagation path of the breakdown had a spiral shape with numerous branches that were mainly directed to the outer part of the spiral. Under normal conditions, the breakdown trajectory is very symmetrical and represents a spiral that expands with time. The observed distortions in Figure 2a (the selected regions) were most likely associated with defects and inhomogeneities in the original ITO layer. As can be seen from Figure 2b, when the formation of several nearby fractal spirals, the distribution law of the fractal regions changes: the spirals grew without intersecting each other. When a magnetic field was applied, the breakdown was oriented in the direction specified by the intensity vector of this field.

![Figure 2](image-url)
The features of current flow processes and the appearance of fractal tracks in the studied structures were largely determined by the fact that the liquid gallium-indium eutectic was used as the lead electrode. When an electric current passed through this liquid contact, it performed self-oscillatory motions along a spiral trajectory during the breakdown. This happened due to the interaction of the current flowing through the probe and the electric field. This trajectory was supposedly associated with the probe's aspiration to return to its initial position at the center of the spiral. At the initial moment, a tunnel breakdown of the ITO particles takes place. Then a sharp increase in the current led to evaporation and sublimation of its material. However, the gallium-indium liquid-contact electrode did not burn out unlike conventional electrodes. Its contact point moved from the region of the evaporated material to the neighboring region almost along the circular path due to the tendency of the elongated electrode to return to the most advantageous vertical position. When the rotation stopped, the contact area of the discharge moved to the next coil of the spiral. The step size of the full turn was determined by the amplitude of the voltage pulse applied to the structure.

Emission spectrums arising from the flow of high-density currents through the layers of oxide compositions were analyzed. A linear spectral characteristic that appeared during the course of this experiment has a dominant peak at a wavelength of 583 nm in the formation of spiral fractal structures (Figure 3a) and additional maximums depending on the parameters of the oxide layer. During formation of the beam structures, peaks at the wavelength of 410 and 450 nm showed the greatest intensity (Figure 3b).

![Figure 3](image-url)

**Figure 3.** Radiation spectrum arising from the breakdown of an ITO layer 3 μm thick and a resistance of 100 Ω per square (a) and 20 Ω per square (b)

When the needle with applied voltage was in contact with the surface of the substrate, it was possible to observe combustion of the substrate material. Figure 4 shows a set of spectral characteristics of radiation corresponding to breakdowns with and without combustion of the material. In addition to the main lines, a large number of weaker peaks are present in the spectrum. These weak peaks corresponded to the radiative transitions that appeared during the processes of thermal shock, sublimation of the material, and formation of fractal microstructures.
Figure 4. Radiation spectrums that appear from the breakdown of the ITO layer with and without combustion of the material.

The most obvious were destructions of thin ITO layers when a thin polymeric layer that increased the contrast of the image were applied to the ITO surface. In addition, in this case the electrical contact between the ITO and the filamentary electrode occurred only after electrical breakdown and destruction of the polymer dielectric film in the region of contact of the threadlike electrode. The polymer film broke through at a voltage of ~ 100 V. Hence, the voltage after the breakdown was applied to the ITO. That consequently allowed a significant current flow through the ITO. Its flow through the ITO layer was localized in a narrow (about 10 μm) track. Its heating led to destruction and removal of the polymer from the track and eventually to the destruction of the ITO in the track region.

Thus, the application of additional polymer films over transparent oxide layers allows visualization of the breakdown processes occurring in the structure. The resulting "polymer photography" makes it possible to assess the quality of the oxide layer.

4. Conclusion

The focus of this research was to study processes that occur during the formation of fractal microstructures in conductive oxide compositions formed on glass substrates. A setup was developed for this research. It allowed usage of high-intensity local field intensity and a nanoelectrode based on a Ga / In eutectic composition to provide a micrometer-sized contact spot diameter. The created fractal micro- and nanostructures were studied using the methods of optical spectrometry, metallurgical computer analysis, and atomic force microscopy. This research has shown that in thin ITO layers (thickness of 100-300 nm) the destructions have the form of individual short tracks with lengths of 200-400 μm. In ITO with thickness of more than 300 nm the destructions have the shape of a multi-turn helix with a diameter of ~ 1 mm. The specific features of current flow processes in the structures studied were
largely determined by the fact that the liquid gallium-indium eutectic was used as the lead electrode. This liquid contact performed self-oscillatory motions along a spiral trajectory when an electric current passed through it. Unlike conventional electrodes, the liquid-contact electrode did not burn out, and the contact point moved from the region of the evaporated material to the neighboring region practically along a circular path because of the tendency of the elongated electrode to return to the most advantageous vertical position. The step size of the turn was determined by the amplitude of the voltage pulse applied to the structure. In most structures, the spiral trajectory of breakdown was supplemented by numerous branches, which in turn have additional processes at the next level of the fractal hierarchy. Analysis of the emission spectrum that occur during the formation of spiral fractal structures shows that a line spectral characteristic with dominant peaks appears, whose position is determined by the parameters of the oxide layer.

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