Synthesis and study of transparent multicomponent metal oxide for use in multisensor system

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Abstract. The thin films on the basis of metal oxides of SnO₂-ZnO-SiO₂ were prepared by sol-gel techniques, and CVD. During the experiment optical and morphological characteristics of materials were investigated. According to the study we can say that this system is promising for use in transparent multisensor devices.

1. Introduction
Functional multi-metal oxides are widely used in various branches of micro- and nanoelectronics. Today the so-called transparent electronic is one of the most promising areas of manufacturing equipment, including the topical issue of the creation of transparent conductive materials. Metal micrometals, conducting polymers, graphite, carbon nanotubes, more often metal oxides are used as the transparent conductors [1-5]. For many years the most common transparent conductor is indium oxide doped with tin (ITO) due to their successful physical and processing characteristics. But due to the shortage of indium replacement ITO more affordable and inexpensive material is a topical issue. The tin dioxide, zinc oxide and titanium dioxide are a recognized competitors of indium oxide today.

2. Experiment.
Thin films based systems oxides of tin, zinc and silicon in different ratios, obtained by means of sol-gel and CVD for possible use as the transparent conductors, as well as a gas-sensitive multisensor systems compatible with transparent electronics were investigated [6-8]. Recent studies show that films of mixed oxides and oxide films, doped metal have the highest gas sensing properties.

Therefore morphological, optical, gas sensing characteristics of system SnO₂-ZnO-SiO₂ were investigated. As precursors were chosen chlorides and nitrides of the respective metals. Tetraethoxysilane (TEOS) was chosen to improve the adhesion to the substrate in a sol-gel method, annealing was carried out at a temperature 600°C. The substrate temperature during the pyrolysis was maintained at 500 °C.

Composition control of the obtained materials was conducted by X-ray phase analysis and diffraction of fast electrons (DFE). As an X-ray analytical equipment used desktop "DRN Farad" and small-angle electron diffraction registering EMR-102 (SELMI). Research film nanostructures morphology were conducted by top-mode vibrational technique of atomic force microscopy (AFM) by nanolaboratories NTegra Therma. Evaluation of the specific surface area was carried out by a known method with BET model. Optical properties were obtained using SF-56 spectrophotometer.
3. Results and Discussion.

Studies of the samples obtained by CVD demonstrated that formed films have a microcrystalline structure (Figure 1-6).

Figure 1. The diffraction pattern of a thin film of tin oxide produced by the CVD method.

Figure 2. Peaks position of a thin film of tin oxide produced by the CVD method.

Figure 3. The diffraction pattern of a thin film of zinc oxide produced by the CVD method.

Figure 4. Peaks position of a thin film of zinc oxide produced by the CVD method.

Figure 5. The diffraction pattern of a thin film of zinc stannat produced by the CVD method.

Figure 6. Peaks position of a thin film of zinc stannat produced by the CVD method.

Analysis of the morphology of the surface showed that the samples obtained using as carrier gas moistened air had a rougher surface compared with the samples obtained by using dried air. (Figure 7, 8).
Figure 7. Histogram distribution density values of Z for the sample obtained using as carrier gas humid air.

Figure 8. Histogram distribution density values of Z for the sample obtained using as carrier gas dried air.

The surface roughness of the samples was evaluated by calculating the fractal dimension of the surface by four methods: variational, by triangulation, counting cubes, by the power spectrum \[\text{Table 1, Figure 9}\]

**Table 1.** The values of the fractal dimension of the surface of the samples obtained by the CVD method with humidified and dry air.

| The synthesis conditions | The variational method | Method of counting of cubes | Triangulation method | Method of the power spectrum |
|--------------------------|------------------------|-----------------------------|----------------------|-----------------------------|
| With the participation of the humidified air | 2.45 | 2.28 | 2.36 | 1.95 |
| With the participation of the dried air | 2.66 | 2.41 | 2.51 | 2.30 |

As a result of the calculation we can say that despite the fact that the technology involving humidified air produces large crystallites with a greater surface roughness of thin films produced in the atmosphere of dry air have a developed surface, as evidenced by the large fractal dimension, calculated 4 ways.
For the purpose of complex characterizing of material the layers formed by sol-gel were investigated. The technology is described in detail in [9]. The single-layer and multi-layer samples with different relative amounts of oxides of tin and zinc were obtained. AFM studies have shown that increasing the relative proportion of zinc leads to a loosening of the porous structure of the layer. (Table 2, Figure 10).

**Table 2.** The value of the fractal dimension of porous hierarchical structures for a different time of maturing of the sol solution, calculated in several ways.

| The relative content of tin dioxide and zinc oxide | Method            | The values of fractal dimension |
|-------------------------------------------------|-------------------|--------------------------------|
|                                                | 150 h             | 360 h             | 1500 h            |
| 2:1                                             | decomposition     | 2,79              | 2,77              | 2,98             |
|                                                | counting cubes    | 2,51              | 2,38              | 2,60             |
|                                                | triangulation     | 2,60              | 2,39              | 2,69             |
|                                                | power spectrum    | 2,59              | 2,95              | 2,99             |
|                                                | decomposition     | 2,82              | 2,72              | 2,70             |
|                                                | counting cubes    | 2,37              | 2,39              | 2,50             |
| 1:1                                             | triangulation     | 2,46              | 2,50              | 2,60             |
|                                                | power spectrum    | 2,54              | 2,48              | 2,16             |
|                                                | decomposition     | 2,40              | 2,42              | 2,79             |
|                                                | counting cubes    | 2,18              | 2,17              | 2,60             |
| 1:2                                             | triangulation     | 2,15              | 2,16              | 2,69             |
|                                                | power spectrum    | 2,06              | 2,60              | 2,53             |
Analysis of the AFM images shows that increasing the proportion of the content of zinc oxide in the solution the number of pores decreases, and the pores become larger. Fractal dimension was calculated with a relative increase in the proportion of zinc oxide is reduced Figure 10, indicating that the downward trend in the developed surface of the samples with respect to the film with a predominant content of tin dioxide. As it can be noted, that dependence of the sol is not changed with increasing exposure time. These data correlate completely with the calculation of the specific surface obtained by the BET method. (Figure 11.)
Optical characteristics of thin films of zinc stannate investigated. From the measurements it was found that the transmittance of the material is close to 1 over the entire visible range. Thus, the results of the study we can say that the investigated thin films have a hierarchical surface morphology. The development of the surface can be controlled as a selection of process parameters and by varying the composition of the material. Investigated thin films have high transmittance, so promising for use in transparent electronics devices.

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