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Optical Properties of Transparent Conducting Coatings Based on an Aluminum Grid Embedded in Anodic Aluminum Oxide

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Abstract—The optical properties of transparent conductive coatings based on an aluminum grid embedded in anodic aluminum oxide have been studied. The coatings were formed by local porous anodizing of an aluminum film deposited on a glass substrate. The conductivity of the coating samples produced reaches the equivalent of the conductivity of films with a surface resistance of 1.0 Ohm/square. The transmittance of the best sample of coatings averaged over the visible wavelength range reaches 89%, which is comparable with the best modern transparent conductive coatings.

Keywords: electrochemical anodizing, aluminum, transparent conductive coatings
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INTRODUCTION

Transparent conductive films are used as transparent electrodes in the production of touchscreens, LCD displays, top electrodes in solar cells and organic light-emitting diodes. They are two-dimensional conductors of electric current. Typically, continuous films of metal oxides (tin-doped indium oxide (ITO), aluminum-doped zinc oxide, calcium- or antimony-doped vanadium oxide) and conductive polymers, such as poly(3, 4-ethylenedioxythiophene) polystyrene sulfonate, PEDOT:PSS, are used as conductors.

ITO is currently one of the most widely used transparent conductive oxides due to its three main properties: high electrical conductivity, optical transparency, and the ease with which it can be obtained as a thin film. ITO is a solid solution of indium- and tin oxides. The optimal mass ratio is 90 : 10 for In_2O_3 and SnO_2. ITO films are transparent in the visible and near infrared range of the spectrum. The main characteristics of the ITO thin film coating, which is used by 90% of the display industry [1], are: 90% transparency at a wavelength of 550 nm and a surface resistance of 20 Ohm/square [2]. The disadvantages of this material include the high cost of indium and a decrease in the transparency of the material in the near infrared range: for wavelengths over 1000 nm, the transparency of the coating decreases sharply relative to the level for the visible spectrum. This is due to the plasma reflection of radiation by free electrons. The ceramic nature of the material also plays a negative role: ITO-based coatings crack when the substrate or a specific device is bent, due to which the specific resistance of the coating increases sharply without changing the optical properties. In addition, organic flexible optoelectronics assumes a low (up to 110°C) temperature of all manufacturing processes, and process temperatures over 300°C or high-temperature postprocessing are often required to obtain high-quality ITO with high transparency, electrical conductivity, and charge carrier mobility.

At the moment, the optoelectronic industry has a need to replace standard transparent conductive coatings based on ITO with a cheaper and more elastic analogue, comparable to ITO in optoelectronic properties and simplicity of production technology, which will allow the formation of flexible displays, cheap film solar cells, etc.

To meet the requirements of cost-effectiveness, transparency for infrared radiation, elasticity and low temperature of transparent conductive layers, the formation of coatings based on various micro- and nanostructured conductors with the structure of nanofabric and nanomesh have been developed and studied. Among the analyzed works, the coating based on silver nanowires demonstrates record parameters [3], namely, its own transparency of 95.2% and specific resistance of 4.2 Ohm/sq, while the resistance increased only two times after 2500 bending cycles. However, the nanofabric has a highly developed surface, since it consists of threads with a diameter of approximately 30 nm. In this case, the resistance of each thread is very sensitive to corrosion and, therefore, the coating requires a passivation layer.

In recent years, information has emerged on the possibility of using metal grids to form transparent electrodes for optoelectronic devices. An example is the creation of a 1 cm^2 solar cell with a front electrode in the form of a gold film with pores of 170 nm in diameter and a distance of 200 nm between them [4]. Compared to the same solar cell structure but using ITO as the front electrode, the authors claim an increase in short-circuit current by 41% and solar cell efficiency by 56%. There is also information about the advantage of using a nanomesh metal structure on the surface of the opaque back electrode of thin-film solar cells, which, due to plasmonic effects, increases the absorption of solar radiation by 50% [5]. Micro- and nanonetworks of metal conductors meet the requirements of elasticity, transparency, and electrical conductivity with a nanofabric structure consisting of contacting metal nanothreads or nanotubes, and a mesh structure containing continuous conductive paths and windows.

We propose an aluminum grid embedded in porous aluminum oxide as a transparent conductive coating. This article presents the results of a study on the optical characteristics of such coatings.

EXPERIMENTAL

The investigated transparent conductive coatings based on aluminum grids embedded in anodic aluminum oxide were formed by porous anodizing of an aluminum film deposited on a glass substrate using a mask [6].

The first series of samples were formed on 1.2-mm-thick glass substrates with an 0.8–1.0-μm-thick aluminum film and a 30-nm-thick niobium sublayer.

A photoresistive mask with a square-cell lattice pattern was formed using standard photolithography operations. The grid track width was 100 μm, and the grid window size was 1 mm. To form a transparent structure while preserving the conductive grid, the formed structures were then subjected to anodic oxidation in a two-electrode electrochemical cell with an aluminum cathode. Anodic oxidation converts the aluminum areas not covered by the mask into transparent aluminum oxide. The mask was then removed. After removing the mask, some of the samples were annealed in air for 1 h at 450°C. This operation was performed to oxidize metal residues at the oxide/substrate boundary in the areas of anodic oxide formation.

The second series of samples was formed on substrates with a structure similar to the structure used for the first series of samples but without a sublayer; 3-mm-thick glass was used as a substrate.

The subsequent processing stages for this series of samples are similar to the route for the first series of samples, with the exception of annealing, which was not carried out in this case.

In addition, selective etching of anodic aluminum oxide was performed on some of the samples in order to ensure maximum transparency of the structure while preserving the conductive grid.

The study of the optical characteristics of the formed transparent conductive films was carried out on four samples (two samples were selected from each series) (Fig. 1):

1. Sample no. 1 (series one): microgrid on 1.2-mm-thick glass, annealed (good transparency, iridescence is visually visible, haze is clearly visible when light is transmitted);
2. Sample no. 2 (series one): microgrid on 1.2-mm-thick glass, not annealed (reduced transparency);
3. Sample no. 3 (series two): microgrid on 3-mm-thick glass, not annealed (good transparency, visually uniform, no scattering is observed when light is transmitted);
4. Sample no. 4 (series two): microgrid on 3-mm-thick glass, not annealed (good transparency, visually nonuniform: there are areas of varying degrees of scattering).

The optical properties of the formed films were studied using a spectrophotometer (Fig. 2). In this work, we used the SF 1024USB spectrophotometer developed and assembled by specialists of the Izovac Company. The SF 1024USB spectrophotometer measures transmission and reflection spectra of optical details and coatings on them. The spectral range of the device is 360–1100 nm. The reflection (R) and transmission (T) spectra were measured directly, and the absorption- and scattering loss coefficient (k) was calculated by subtracting the sum of reflection and transmission from the 100% level. The Fourier infrared spectroscopy method was used to study the spectra of the samples in the infrared wavelength range from 2 to 5 μm. The transmission and reflection coefficients were measured at three different points on the sample surface (Fig. 1d).

RESULTS AND DISCUSSION

The spectra of transmission-, reflection-, and loss coefficients in the wavelength range from 360 to 1100 nm, measured on the mentioned samples, are shown in Fig. 3. On the samples of the first series, relatively low transparency is accompanied by high losses (scattering, absorption) and reflection. For the sample no. 1, this is probably due to crystallization and compaction of aluminum oxide after annealing and that for sample no. 2 due to the high content of underoxidized metal inclusions. Samples from the second series have a fairly good transmission coefficient. When analyzing the optical properties of the presented samples, it is necessary to take into account that the substrate made of the 3-mm-thick K8 glass has a transmission coefficient of 89% at a wavelength of 550 nm, a reflection coefficient of 8%, and an absorption coefficient of 3%.

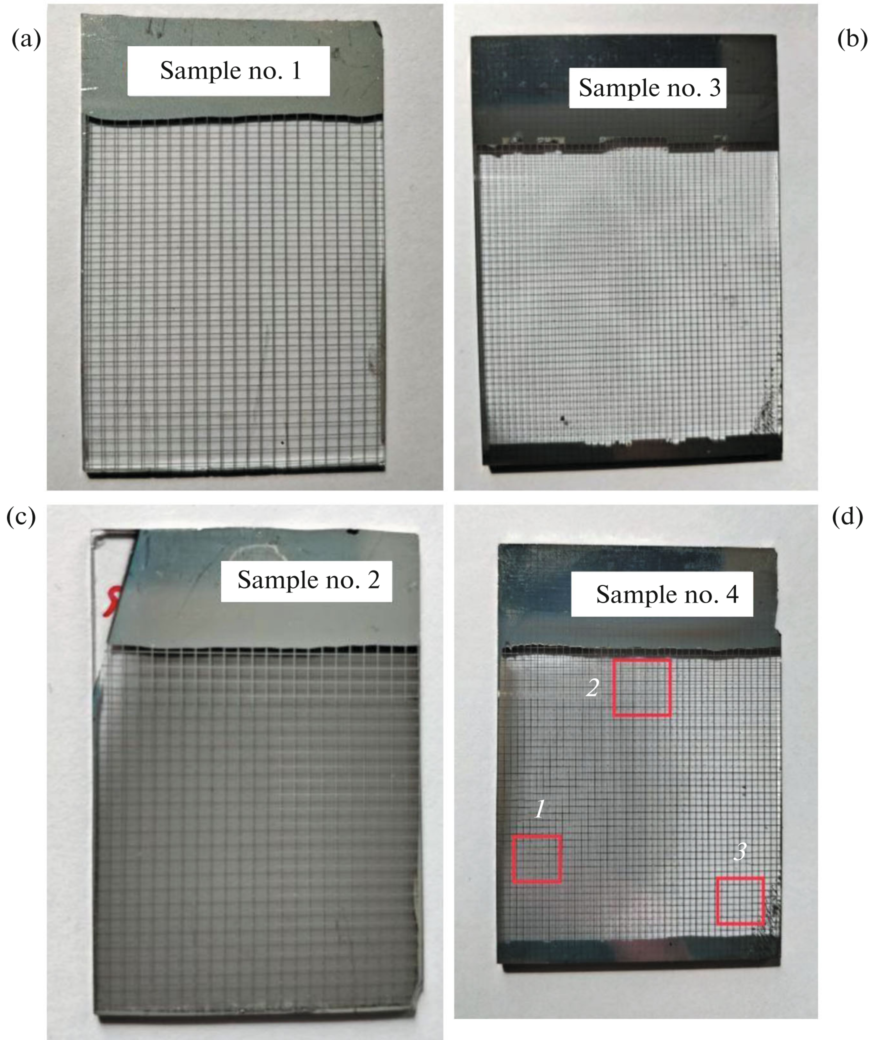

Fig. 1. Photographs of four samples of conductive coatings based on aluminum grids embedded in anodic aluminum oxide on a glass substrate.

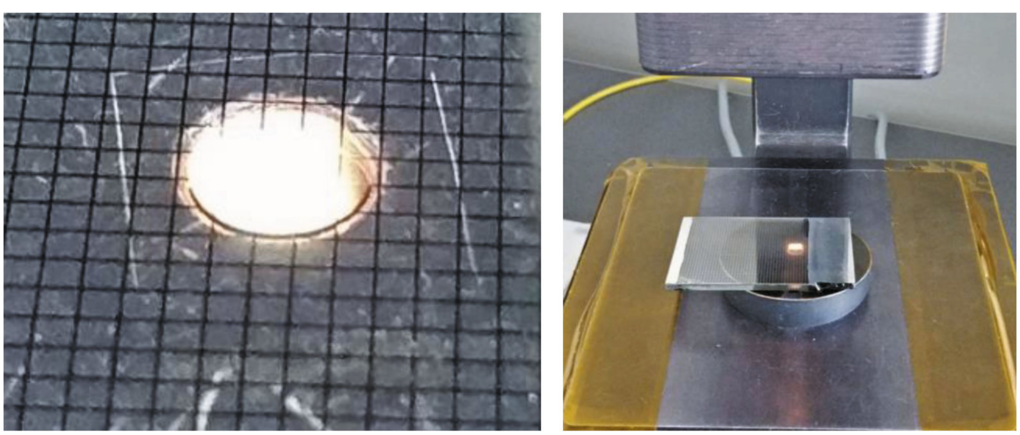

Fig. 2. Photographs of samples during the process of studying their spectra.

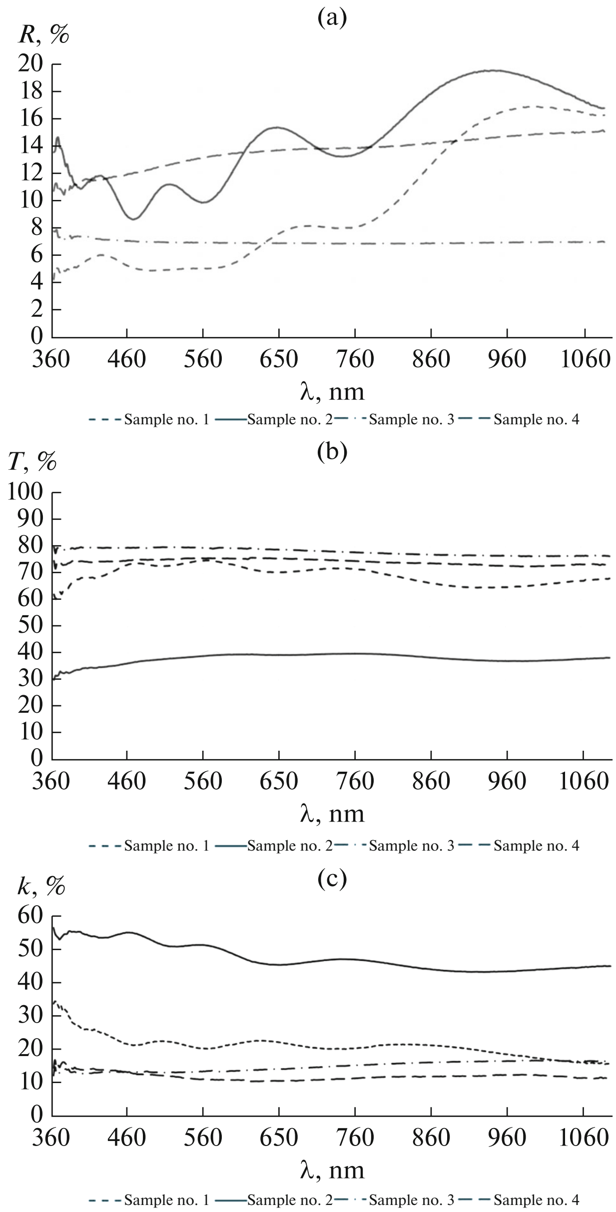

Fig. 3. Spectra of (a) reflection coefficients, (b) transmission coefficients, and (c) losses of the studied samples of transparent conductive coatings on glass substrates in the wavelength range from 360 to 1100 nm.

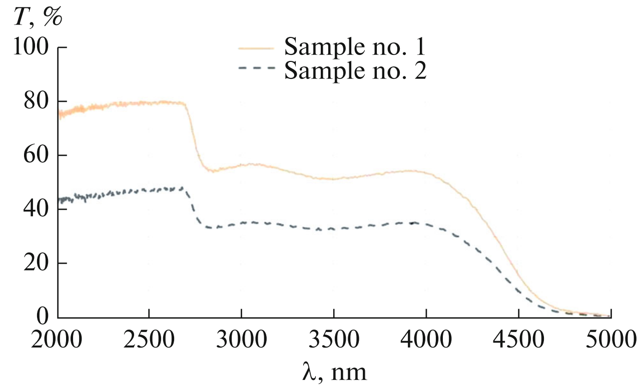

Fig. 4. Transmission spectra of sample nos. 1 and 2 in the infrared range.

The transmission spectra of the first series of samples in the infrared range are shown in Fig. 4. These spectra show a sharp drop in transmission coefficient at a wavelength of approximately 2700 nm, a plateau at 2800–4000 nm, and a cutoff of waves with a length of more than 4700 nm.

When comparing the transmission spectra of the sample no. 3 and pure glass (Fig. 5), it was found that the spectral characteristic of sample no. 3 is determined mainly by the substrate material and its thickness—the transmission level of the pure substrate in the range of 2800–4000 nm is 30% lower than the transmission coefficients of the microgrid samples on thin substrates, which indicates the absorption of infrared radiation in the volume of the glass used. Unlike ITO, the transparency of which drops sharply from 90% to a few percent in the near infrared range, the coatings based on the microgrid from the samples of the second series demonstrate a noninterference character and independence on the wavelength. A decrease in transparency by 8–10% relative to pure glass is due only to the fact that the conductor tracks occupy approximately 10% of the coating area.

In the studied samples, the best coating in terms of optoelectronic properties was found in sample no. 3 with an intrinsic transparency of 89% (Fig. 6) and a specific resistance of 2.1 Ohm/sq. If conductivity is considered as a more important parameter, then the best coating is for sample no. 4 with an intrinsic transparency of 84% and a specific resistance of 1.0 Ohm/sq. Intrinsic transparency is understood as the transparency of the directly formed coating without taking into account the substrate. These parameters compete with the best modern coatings based on new materials. For example, a coating based on silver nanowires [3] with a transmittance of 95.2% and a specific resistance of 4.2 Ohm/sq. transmits 6.2% more visible light but has a specific resistance that is two times higher.

The disadvantage of the presented samples is the low resolution of the used photomask, due to which the grid is visible to the naked eye, and it is impossible to form a display structure on such an electrode due to the nonuniform distribution of the electric field. Nevertheless, the proposed technology allows the use of photomasks with an arbitrary cell size and track width in the range of values available in modern photolithography. The use of a stronger mask made of refractory metal can increase the anisotropy of etching and prevent oxidation of the metal grid due to the photoresist lifting by the growing porous oxide.

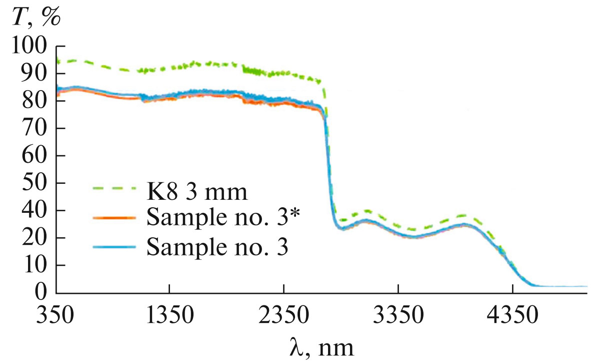

Fig. 5. Transmission spectra of the substrate (K8 glass, 3 mm thick), sample no. 3 (with porous anodic aluminum oxide) and no. 3* (the same sample after removing the porous anodic aluminum oxide).

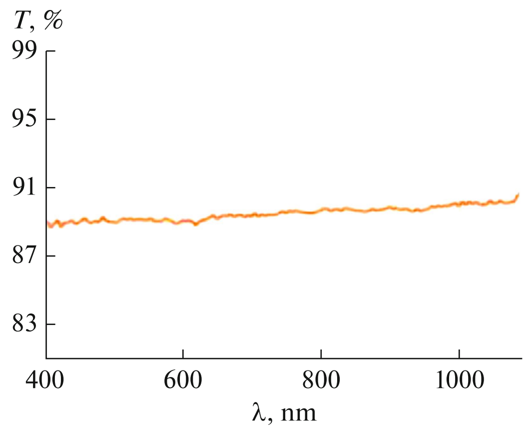

Fig. 6. Intrinsic transparency of sample no. 3.

CONCLUSIONS

Transparent conductive coatings based on an aluminum grid embedded in anodic aluminum oxide have been formed with an intrinsic transmittance of 89% in the wavelength range of 360–1100 nm at a specific resistance of 2.1 Ohm/sq. Such a coating transmits 6.2% less visible light but has a resistance that is two times lower than the best similar coating based on silver nanowires. At the same time, the resulting coating is a layer protected from chemical and mechanical actions by porous anodic aluminum oxide, while the above-mentioned silver coating requires a passivation layer, which can also reduce its transparency.

Thus, using inexpensive materials and due to the oxidation anisotropy and self-organization of the porous structure characteristic of electrochemical anodizing, it is possible to produce inexpensive, mechanically and chemically stabilized transparent electrodes with optoelectronic parameters that compete with the best known world conductive transparent coatings in the visible wavelength range.

In the future, work on developing this technology will be aimed at reducing the size of the grid cells while maintaining electrical conductivity and transparency. For this purpose, it is planned to use photolithography on the surface of the aluminum film to form a mask of a refractory metal, such as niobium, with greater adhesion to the film. At the same time, it is expected that the mask will be prevented from peeling off due to the volumetric growth of porous anodic aluminum oxide and the interruption of conductive paths. Another interesting direction is the formation of similar coatings on a polyamide film with subsequent study on the elasticity of the coating. Based on mathematical modeling, it is possible to optimize the grid topology to achieve optimal operating parameters.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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