

Investigation of temperature stability of ITO films characteristics

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Abstract. The paper represents research of thermal stability of optical and electro-physical parameters of ITO films deposited using various techniques. Variation of optical and electro-physical parameters was recorded using spectroscopy, and Hall's and four-probe measurements. The best thermal stability was demonstrated by ITO films deposited by metal target sputtering In(90%)/Sn(10%) in mixture of gases O₂ (25%) + Ar (75%) with further annealing in air atmosphere. This enables to apply this technique for production of thin film transparent resistive elements capable to heat the translucent structures up to 100°C without deterioration of their parameters.

Introduction

ITO (Indium tin oxide, In₂O₃:Sn) thin films have recently gained much more attention among research teams [1-7], due to their unique combination of high electrical conductivity, high transmission coefficient in the visible portion of the spectrum and reflective power in IR-range.

These films are extensively used in electronics and microelectronics, in transportation industry and architecture. Research in [2] describes how ITO films are used as a layer of current distribution for GaN-based blue light-emitting diodes. The authors demonstrated in their study that application of these ITO films enabled growth of light-emitting diodes efficiency coefficient by 2.5 times. Review paper [3] reports on successful production of ITO-based thin film oxide transistors. Application of ITO films in transportation industry and architecture was discussed in [4]. C.G. Granqvist and A. Hultaker present results of proving out the technology of ITO films deposition on sheet glass for their further use in optimized electrochromic devices for smart windows production. Smart windows find their challenging application as electronic tinted coating of airplane portholes [5].

Apart from the applications mentioned above, ITO films can also be prospectively used as transparent heating elements. Some research and production companies [6, 7] manufacture heated glass used in temperature-proof houses for surveillance cameras, heated sighted devices, observation windows, transparent heating elements of LCD displays enabling to broaden the temperature range of functional devices operation up to -60°C.

Due to large interest in ITO films application as heating elements, its properties are needed to be kept unchanged during long-term high temperature impact. However, the data

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of research papers referring to this issue generally provide the dependences of ongoing variations of films electrical conductivity during their heating in order to define its mechanisms [8]. Variations of electro-physical properties after heating were not considered in previous studies.

The given paper aims to study the stability of optical and electro-physical parameters of ITO films deposited by magnetron sputtering after high temperature treatment.

Experimental

ITO films under study were 130-150 nm thick; they were deposited on preliminary polished sitall substrate by magnetron sputtering technique according to different modes, as specified in Table 1. Films deposition was performed by reactive magnetron sputtering with various working gas composition (modes 1 and 2) according to the technique described in [9]; and also films were obtained by oxide target sputtering in argon atmosphere.

Table 1. Deposition modes of samples under study.

Mode No.	Applied target	Working atmosphere composition	Deposition parameters	In-process annealing of samples
1	In (90%) / Sn (10%)	O ₂ (25%) + Ar (75%)	Working pressure $8 \cdot 10^{-3}$ torr; discharge current 0.3 A; temperature of substrate during deposition 25 °C	Annealing in air atmosphere at 600 °C for 20 min.
2	In (90%) / Sn (10%)	O ₂ (29%) + Ar (71%)		Annealing in nitrogen atmosphere at 600 °C for 20 min.
3	In ₂ O ₃ (90%) / SnO ₂ (10%)	Ar (100%)	Working pressure $6 \cdot 10^{-3}$ torr; discharge current 1.85 A; temperature of substrate during deposition 250 °C	Without annealing

Temperature stability of ITO films parameters under high temperature treatment was investigated as described below. Samples with deposited ITO films and the formed metallic islands were placed into muffle furnace and kept for 15 and 30 min at 600 °C in air atmosphere. After temporary exposure the surface resistance of films was measured, as well as concentration and charge carrier mobility and transmission coefficient.

Surface resistance measurements were conducted using four-probe method. Parameters of charge carriers in the films were measured using Hall Effect. Both methods of thin films parameters checkout were described in details in [10].

Transmission spectrum of ITO films was recorded by UV-spectrometer USB 2000. The facility is intended for conducting various analytical investigations of optical absorption spectrum, transmission spectrum, reflection and lumescent spectrum in UV and visible range.

Results

Electro-physical parameters of ITO films were measured using the above mentioned methods right before and after high temperature exposure of samples at 600 °C for 15 and 30 min under atmospheric pressure. Measuring results can be found in Tables 2, 3 and 4. Figures 1, 2 and 3 illustrate dependence graphs for transmission coefficient and wavelength during annealing for films obtained using all three modes.

Table 2. Variation of electro-physical parameters of samples obtained using the Mode No.1 depending on the time of exposure in the furnace at 600°C.

Controlled parameter	Duration of high-temperature exposure		
	Before heating	15 minutes exposure	30 minutes exposure
Surface resistance, Ohm/□	5.9	2.5	4.9
Charge carrier concentration, m ⁻³	5·10 ²⁵	4.6·10 ²⁵	1.1·10 ²⁵
Charge carrier mobility, m ² /(V·s)	0.16	0.26	0.05

Comparison of the obtained experimental data of the samples under study allowed revealing the following tendencies. During 15 min exposure of samples obtained in accordance with the Mode No.1 in the furnace at 600°C the decrease of surface resistance of the films occurs due to increased charge carrier mobility. It is most probably conditioned by formation of more expressed films crystalline structure. Further exposure results in the decrease in surface resistance of ITO films compared to the films that were not subject to annealing in the furnace. This fact is attributed to the increase in charge carrier concentration originating from activation of the introduced additive and formation of oxygen vacancies.

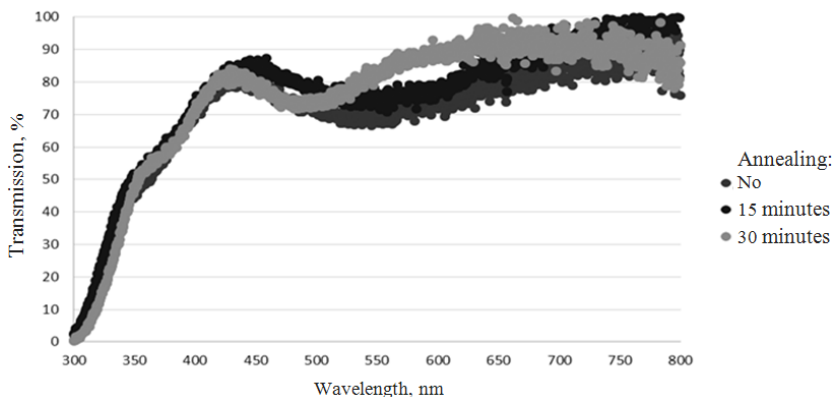


Fig. 1. Dependence graph of transmission spectrum of ITO film on the wavelength obtained using Mode No. 1.

As can be seen from Figure 1, during high temperature treatment of films the average increase of the transmission coefficient by 10% is observed.

Table 3. Variation of electro-physical parameters of samples obtained using the Mode No.2 depending on the time of exposure in the furnace at 600°C.

Controlled parameter	Duration of high-temperature exposure		
	Before heating	15 minutes exposure	30 minutes exposure
Surface resistance, Ohm/□	8.1	21.8	18.9
Charge carrier concentration, m ⁻³	1.7·10 ²⁵	7.2·10 ²⁴	1.3·10 ²⁵
Charge carrier mobility, m ² /(V·s)	0.21	0.19	0.12

Comparison of experimental data of samples obtained according to the Mode No.2 showed that the increase of surface resistance is observed during 15 min exposure in the furnace. The increase in surface resistance is attributed to the decrease in charge carrier concentration in ITO film after heating, which probably occurs due to oxygen diffusion from the film into atmosphere during heating. Further exposure leads to continuing decrease in surface resistance. This fact is conditioned by the increased concentration of charge carriers during activation of the introduced stannic additive.

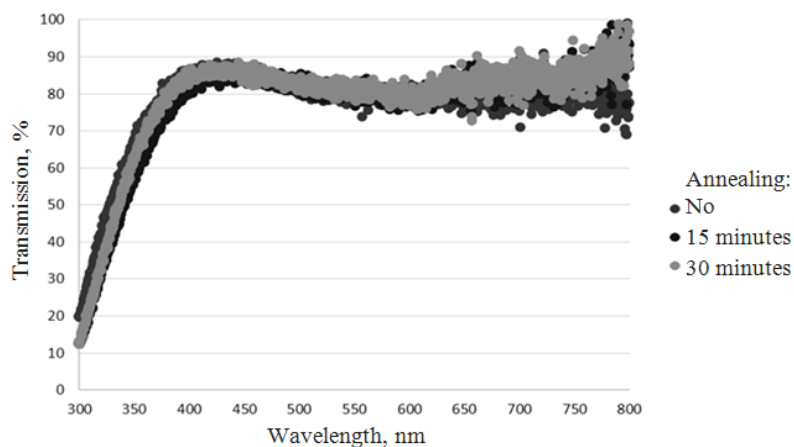


Fig. 2. Dependence graph of transmission spectrum of ITO film on the wavelength obtained using the Mode No. 2.

Figure 2 demonstrates stable dependence of the transmission coefficient on the wavelength irrespective of the exposure duration in the furnace.

Table 4. Variation of electro-physical parameters of samples obtained using the Mode No.3 depending on the time of exposure in the furnace at 600°C.

Controlled parameter	Duration of high-temperature exposure		
	Before heating	15 minutes exposure	30 minutes exposure
Surface resistance, Ohm/□	315.8	25.7	19.9
Charge carrier concentration, m ⁻³	6·10 ²⁴	1.3·10 ²⁵	7.67·10 ²⁴
Charge carrier mobility, m ² /(V·s)	0.04	0.11	0.15

Experimental data of samples obtained using the Mode No. 3 shows decrease in surface resistance during high temperature exposure. It is attributed to the increase in concentration and mobility of charge carriers due to formation of more expressed crystalline structure of films and by the activation of dopant stannic atoms.

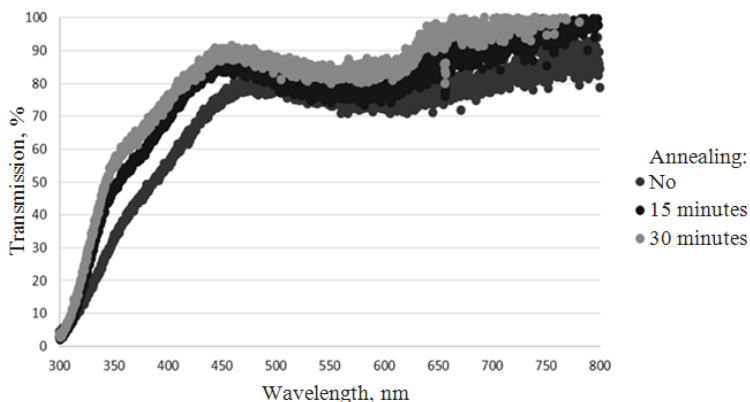


Fig. 3. Dependence graph of transmission spectrum of ITO film on the wavelength obtained using the Mode No. 3.

Figure 3 illustrates the increase of the films transmission coefficient. This is attributed to the increased content of oxygen in the film.

Thus, the Mode No.1 appeared to be the best technique of ITO films deposition on transparent substrates for their further use as a transparent heating element. High temperature heating almost does not alter the surface resistance of these films. At the same time, the transmission coefficient after temperature treatment of films increased from 75 % to 85 %.

Application of this deposition technique for optical transparent electrically conductive ITO films stipulated the design elaboration of thin film transparent heating elements (Fig. 4a). The elements are capable to heat the surface up to 100°C without deterioration of its parameters. The obtained temperature dependence of a heating element based on ITO film (resistance 40 Ohm/□) on the applied power normalized per surface unit is shown in Figure 4b.

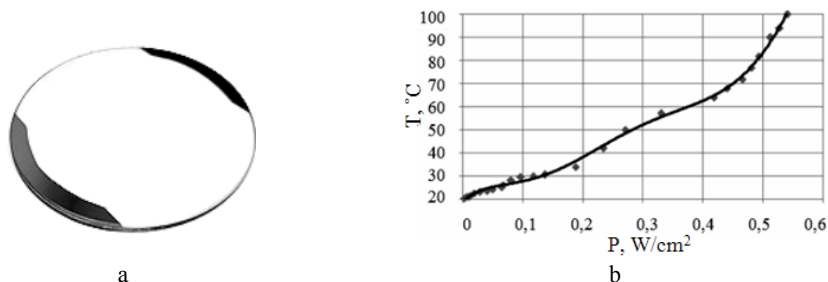


Fig. 4. Heating element based on resistive ITO film (a) and dependence of the temperature of heating on the consumed current power (b).

Conclusion

Therefore, the most rational technique of ITO films deposition for their further use as a transparent heating element is reactive magnetron sputtering technique of metal target In (90%) / Sn (10%) in oxygen-containing atmosphere O₂ (25%) + Ar (75%) with further annealing in air atmosphere. This is the exact mode when the parameters of deposited films under high temperature treatment are not deteriorated.

These results contribute to the more thorough selection of the deposition technique of ITO films further applied in design of reliable thin film heating elements.

The extension of research will be connected with defining the optimal sheet resistance of ITO films providing better effectiveness of heating element.

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References

1. Review of the market for indium-tin oxides and materials for their production in the world, available at: <http://www.infomine.ru/order-demo/458> (in Russian)
2. I.P. Smirnova, L.K. Markov, A.S. Pavlyuchenko, M.V. Kukushkin, Semiconductors, **46**(3), 384 (2012) (in Russian)
3. E. Fortunato, P. Barquinha, R. Martins, Advanced Materials, **24**(22), 2945 (2012)
4. C.G. Granqvist, A. Hultaker, Thin Solid Films, **411**, 1 (2002)
5. A.I. Cheban, The first European "Plane of Dreams" arrived in Ukraine, available at: <http://alexcheban.livejournal.com/102426.html> (in Russian)
6. "Polytech" research and production company website, available at: <http://www.carmirror.ru/catalogue.php?part=24> (in Russian)
7. "NIKOL" group of companies website, available at: <http://www.aonikol.ru> (in Russian)
8. V. Brinzari, M. Ivanov, B.K. Cho, M. Kamei, G. Korotcenkov, Sensors Actuators B Chem., **148**, 427 (2010)
9. Y.S. Zhidik, P.E. Troyan, Y.V. Sakharov, Reports of TUSUR, **26**(2) part 2, 169 (2012) (in Russian)
10. S.V. Smirnov, *Metody issledovaniya materialov i struktur jelektroniki* [Methods for studying materials and structures of an electron] (TUSUR, Tomsk, 2007) (in Russian)