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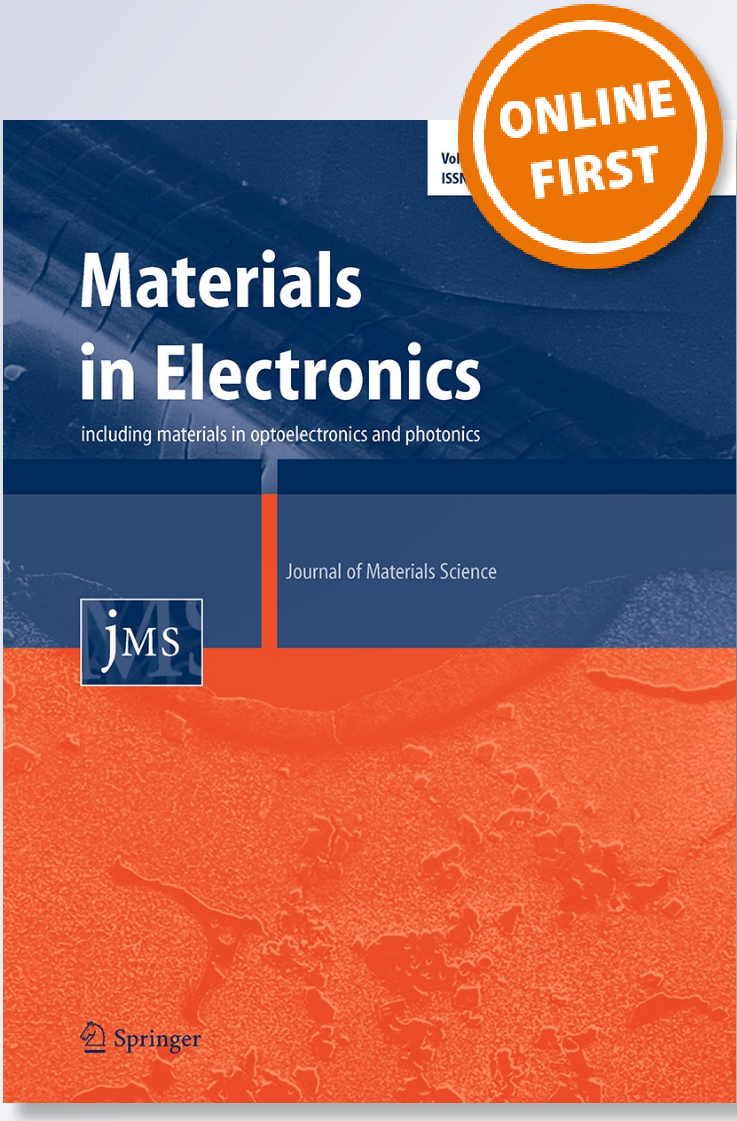
Journal of Materials Science: Materials in Electronics

ISSN 0957-4522

J Mater Sci: Mater Electron
DOI 10.1007/s10854-015-3314-7



Springer

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Effects of tin content on structure, properties, electrical repeatability, uniformity and stability of high sheet resistance ITO thin films for touch panels

Xianjie Zhou[1] · Jiwen Xu[2] · Ling Yang[3] · Guisheng Zhu[2] · Zhiming Yu[1]

Received: 15 March 2015 / Accepted: 2 June 2015

Abstract The Sn-doped In_2O_3 (ITO) thin films with high sheet resistance for touch panels application were deposited by magnetron sputtering process. The effects of tin content on the structure, optical properties, sheet resistance, electrical repeatability, uniformity and stability of ITO thin films were deeply investigated. The ultrathin ITO thin films showed polycrystalline structure with the (222) preferred orientation and smooth surface with low surface roughness. Furthermore, the ultrathin ITO thin films demonstrated strong visible transmittance of above 85 % and had no interference ripples. The ultrathin ITO thin films with ingredient ratio of 95/05 showed stable high sheet resistance, excellent repeatability and uniformity, and could resist the influence from external environment.

1 Introduction

Recently, transparent and conducting oxide (TCO) thin films which could simultaneously resolve the contradictory concern of conductivity and transparency in one kind of material have been broadly studied [1]. Tin doped In_2O_3 (ITO) thin film is the most important TCO thin films because of its ultralow resistivity and high transmittance in the visible light range [2]. ITO thin films were used to make transparent conductive layer for displays such as liquid crystal displays, plasma displays and E-paper displays [3]. In recent years, ITO thin films were widely employed in touch panels as touch sensitive layer in resistive and capacitive touch panel technologies [4, 5]. However, the ITO thin films used as common and pixel electrodes in flat-panel displays demand an ultralow sheet resistance, and the thickness was usually above 100 nm [6]. During the touch panel fabrication process, the first step of glass or plastic based TCO thin films were deposited by magnetron sputtering process, and then etched the as-deposited thin films to become the detect units of the predetermined pixel sizes. Additionally, the etched patterns should be visually invisible in order to avoid disturbing the displayed images on the underlying monitor. Therefore, ITO thin films used in touch panels demand ultrathin thickness.

The dependence of sheet resistance on thickness shows that sheet resistance decreases when film thickness increases [7, 8]. The transmittance of ITO thin films increases with decreasing film thickness, but concomitantly decrease of conductivity due to its low crystallinity would happen [7]. The content of tin dopant is comparatively important for conductivity adjustment of ITO thin films system [9, 10], and the optimal doping content of 10 at.% tin was reported by Kostlin [11]. Therefore, the ITO targets with doping content of 10 wt% SnO_2 were usually used to deposit low resistivity ITO thin films. According to the specific requirements of touch panel, ITO thin films with different values of sheet resistance were designed and deposited. To obtain high sheet resistance ITO thin films, decreasing growth temperature, tin content and film thickness are proved to be feasible approach to increase

✉ Jiwen Xu
csuxjw@126.com

1 School of Material Science and Engineering, Central South University, Changsha 410083, People's Republic of China

2 Guangxi Key Laboratory of Information Materials, School of Materials Science and Engineering, Guilin University of Electronic Technology, No. 1 Jingji Road, Guilin 541004, People's Republic of China

3 Guangxi Experiment Center of Information Science, Guilin University of Electronic Technology, Guilin 541004, People's Republic of China

Published online: 10 June 2015

sheet resistance. However, decreasing growth temperature would affect the electrical stability of ITO thin films under multiple environmental conditions [12], and decreasing film thickness deteriorates the uniformity of both thickness and sheet resistance of ITO thin films. In recent years, a large number of works focused on how to prepare low resistivity ITO thin films with ultrathin thickness [5, 12–15]. The objective of the most work on ITO thin films is to maximize the conductivity while simultaneously maximizing the optical transmittance. To the best of our knowledge, ITO thin films with high sheet resistance, high transmittance, uniformity and stability are seldom investigated. Adjusting doping content of tin and deposition at high growth temperature may be possible for touch panel oriented ITO thin films with high sheet resistance and electrical stability.

In this work, ITO thin films with high sheet resistance of 210 ± 30 Ω/□ were investigated by adjusting the doping content of tin. The corresponding structure, electrical and optical properties, and repeatability, uniformity and stability of sheet resistance are examined in detail.

2 Experiments

The home-made 3-inches ITO ceramic targets with SnO_2 content of 3, 5, 7 and 10 wt% (named 97/03, 95/05, 93/07 and 90/10) were fabricated by sintering co-precipitationally synthesized powders and bonded with Cu backing plate. Ultrathin ITO thin film on touch panel glass (50 × 50 × 0.5 mm) were prepared by dc magnetron sputtering process. The base pressure of sputtering chamber was 5.0×10^{-6} Torr. During sputtering process, the working pressure was fixed at 6 m Torr, the power was 55 W, the oxygen content was 0.4 % by controlling the argon and oxygen flow, and the growth temperature heated by halogen lamp was controlled at 200 °C. The thickness of ultrathin ITO thin films is in range of 20–25 nm by controlling sputtering time, and the thickness decreases with increasing tin oxide content. The thickness of 90/10, 93/07, 95/05 and 97/03 ITO thin films was 20.4 nm, 22.3 m, 23.2 nm and 24.8 nm respectively. The stability of sheet resistance was investigated under high temperature, high temperature and high humidity, and alkaline conditions [12].

The thickness of ITO thin films was jointly measured by reflectometer (ST2000-DLXn, K-MAC) and step profiler (XP-100, Ambios). The phase structure and preferred orientation were confirmed by X-ray diffractometer (XRD, D8-ADVANCE, Bruker). The surface morphology and roughness were analyzed by a scanning electron microscope (SEM, S4800, Hitachi) and atomic force microscopy (AFM, D3100, Vecco). The sheet resistance was measured by four-point probe platform (MCP-T610, Mitsubishi) at room temperature. The optical transmittance in the visible light range was measured by spectrophotometer (V-600, Jasco).

3 Results and discussion

Figure 1 shows the XRD patterns of the ultrathin ITO thin films as a function of tin content. It can be seen that the as-deposited ITO thin films exhibits three peaks of indices (222), (400) and (440), which matches well with the bixbyite tin substituted In_2O_3 structure [16]. The intensity of all the diffraction peaks is relatively weak because of ultra small film thickness. The intensity of the (222) peak is significantly higher than that of the (400) and (440) peaks, which means that the ultrathin ITO thin films still exhibit the (222) preferred orientation.

As shown in Fig. 1b, all the (222) diffraction peaks of four kinds of ITO targets shift toward higher angle as compared with those of the cubic In_2O_3 [$2\theta_{(222)} = 30.58°$], which is an indication of the replacement of the smaller Sn^{4+} ions (Sn^{4+}: 0.69 Å) in the cubic In_2O_3 lattices (In^{3+}: 0.80 Å) and, thus, the formation of the smaller ITO lattice. The 90/10 ITO target shows a large angle shifting because of the high doping content of tin. However, all the (222) diffraction peaks of four kinds of ITO thin films shift toward lower angle as compared with those of the ITO targets. This decrease in 2θ value indicates an increase of the film lattice constant, which is probably related to the strong compressive stress in films due to the ultra small film thickness [17].

The surface morphologies of SEM, 2D and 3D AFM images of the 95/05 ITO thin films are shown in Fig. 2a–c, respectively. As shown in Fig. 2a, the 95/05 ITO thin film shows very smooth surface morphology, and contains massive nanocrystals. The 2D and 3D AFM images illustrate that the 95/05 ITO thin film also shows very flat and tiny grains over the entire surface (20 × 20 μm^2). It is known that the root mean square roughness (R_q) and mean roughness (R_a) have been used to describe the surface morphology. R_q is the standard deviation of the surface height within the given area, and R_a is defined as the mean value of the surface height relative to the center plane. The R_q and R_a surface roughness of the 95/05 ITO thin films is 0.236 and 0.178 nm, respectively. The surface roughness values of R_q and R_a is smaller than 1 nm, which illustrates that the ultrathin ITO thin films have a flat and homogeneous surface morphology. The 3D AFM image illustrates a number of columnar bulges on the surface.

Figure 3 shows the optical transmittance spectra of all the ITO thin films (including glass absorption) in the wavelength range of 300–800 nm as a function of tin content. It can be seen that undulating ripples of the transmittance curves that frequently occurred in the thicker

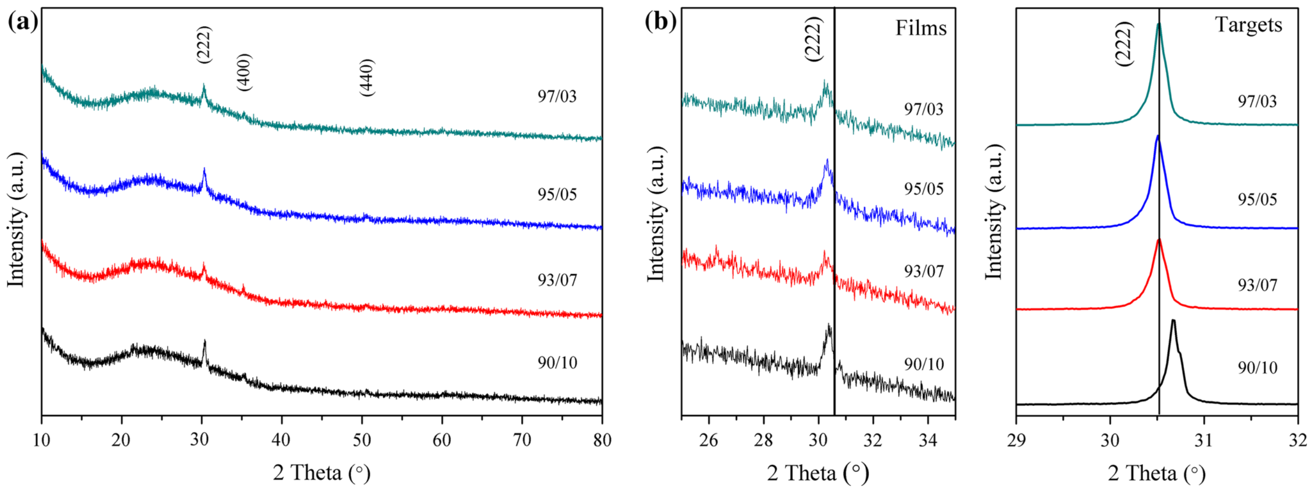

Fig. 1 **a** XRD patterns of ITO thin films as a function of tin content, and **b** the magnification of the (222) peak of ITO thin films and targets

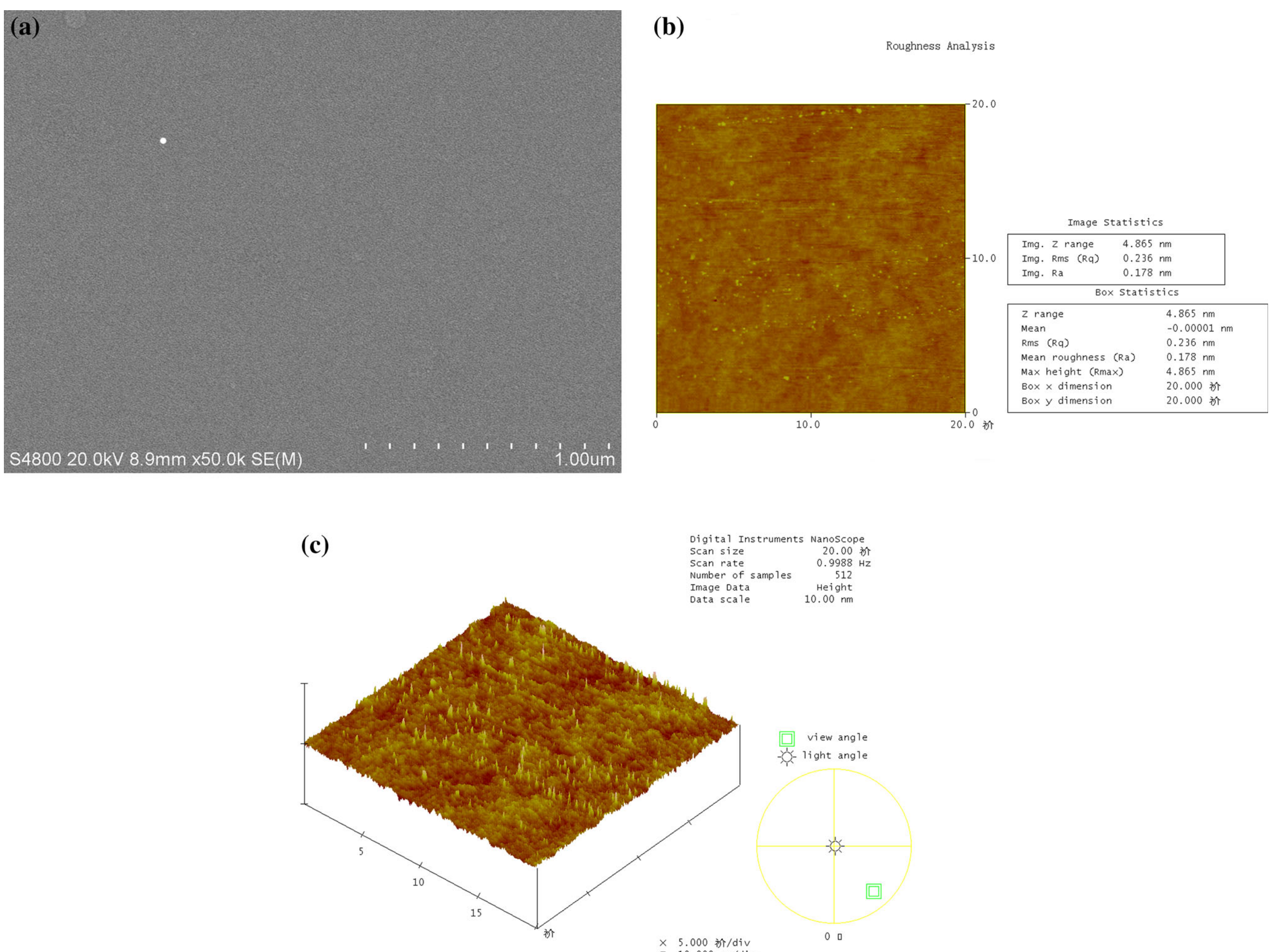

Fig. 2 Images of the 95/05 ITO thin film analyzed by **a** SEM, **b** 2D and **c** 3D AFM

ITO thin films are invisible in the ultrathin ITO thin films. The straight curves in 400–800 nm indicate that all the ITO thin films do not generate the interference phenomenon in the interface between the ITO thin film and the glass substrate. The very small film thickness can not meet the requirement of optical interference in the visible light range

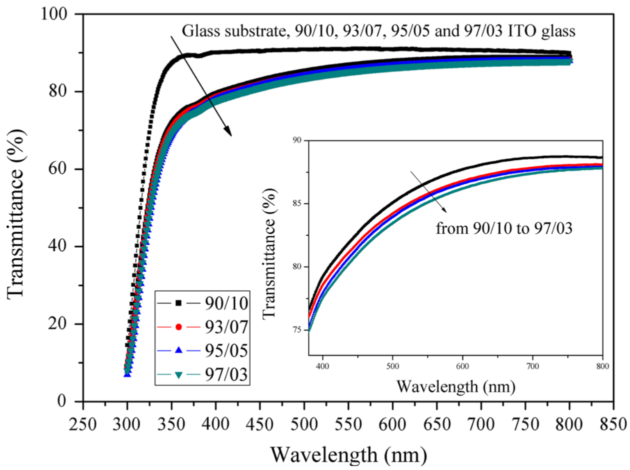

Fig. 3 Optical transmittance of ITO thin films as a function of tin content in visible light range, *inset* shows the magnification at the range of 380–800 nm

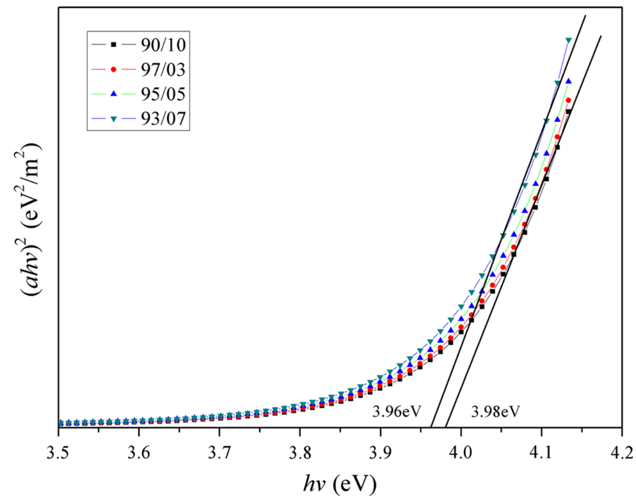

Fig. 4 Band gap of ITO thin films as a function of tin content

as compared with those of thicker ITO thin films [18]. The inset clearly indicates that the transmittance slightly increases with decreasing film thickness. The transmittance of all the ITO thin films is close to 90 % in the visible light range. The absorption edges shift towards the short wavelength side (blue shift) and the band gaps increase with decreasing film thickness.

The optical absorption edge was determined by the optical absorption, a simple method that provides explanation for the features concerning the band structure of the ITO thin films. The optical absorption coefficient, α, of a direct band gap semiconductor near the band edge, for photon energy $h\nu$ greater than the band gap energy E_g of the semiconductor, is given by the relation [19].

$$\alpha h\nu = C(h\nu - E_g)^{1/2} \quad (1)$$

Where h is Planck's constant and ν is the frequency of the incident photon. The Tauc plot of $(\alpha h\nu)^2$ versus energy $h\nu$ for all the ITO thin films was shown in Fig. 4. The band gap energy was obtained by extrapolating the linear part of the Tauc plot curves to intercept the energy axis (at $\alpha h\nu = 0$). The estimated values of E_g for the ITO thin films increase from 3.96 to 3.98 eV with an increase of tin content in the In_2O_3 lattice. As shown in Fig. 4, the ultrathin ITO thin films exhibit an expansion of band gap energy with decreasing film thickness, which can be mainly attributed to the one-dimensional quantum confinement (ODQC) effect in the ultrathin ITO thin films, the similar results was observed in ultrathin amorphous IGZO films [20]. The allowed direct transitions were found previously for different ITO thin films and the direct optical band gap, E_g, values ranging from 3.5 to 4.5 eV were also reported [21, 22]. However, the shift of band gap for thicker ITO thin films is Burstein–Moss (BM) effect rather than ODQC effect [20].

Figure 5 shows the values and corresponding distribution of sheet resistance of all the ITO thin films as a function of tin content. It can be seen that the desired high sheet resistance can be obtained by controlling film thickness and using four kinds of ITO targets before three reproducible experiments. However, the repeatability of high sheet resistance deposited by using four kinds of ITO targets shows bigger difference. The ITO thin films deposited by using the 90/10 and 93/07 ITO targets show poor repeatability of high sheet resistance at three reproducible experiments. The values of sheet resistance at some batches and locations are out of range of 210 ± 30 Ω/□. The sheet resistance of two 90/10 ITO thin films is a little bigger. One of 93/07 ITO thin films is a little larger sheet resistance than 240 Ω/□, and the other two ITO thin films are a little smaller than 180 Ω/□. The ITO thin films deposited by using the 95/05 and 97/03 ITO targets show excellent repeatability of high sheet resistance at three reproducible experiments, which mainly attributes to the influence of film thickness. As is known to all, the sheet resistance of ITO thin films increases with decreasing film thickness. The ITO thin films with high tin content deposited at the same condition show better conductivity. The increase of sheet resistance of ITO thin films with high tin content in this work was achieved by reducing film thickness. The decrease of film thickness deteriorates the uniformity of film thickness, which results in bad repeatability and uniformity of sheet resistance.

The effect of tin content on the uniformity of sheet resistance is evaluated by the equation [23]:

$$UNI\,(\%) = \frac{Rs_{\max} - Rs_{\min}}{2 \times Rs_{avg}} \times 100\% \quad (2)$$

where UNI (%) is the sheet resistance uniformity; $Rs_{\max}$, $Rs_{\min}$ and Rs_{avg} are the highest, lowest, and average

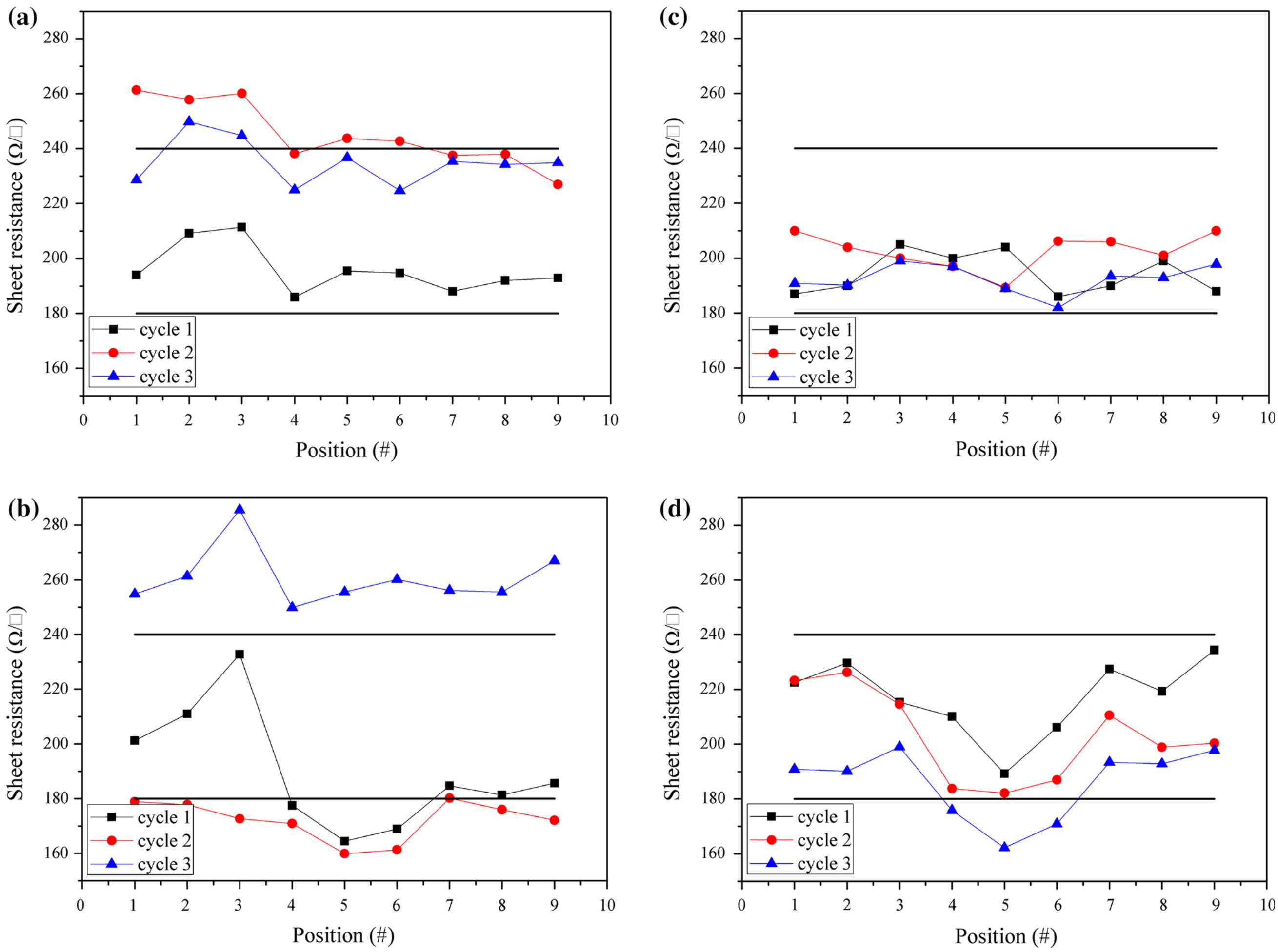

Fig. 5 Values and distribution of sheet resistance of ITO thin films as a function of tin content and batches

measured sheet resistance, respectively. Therefore, the smaller *UNI* (%) is preferable for ITO thin films. Figure 6 shows the calculated uniformity values of sheet resistance of all the ITO thin films at three reproducible experiments. It can be seen that the repetition of the uniformity of sheet resistance at different batches and locations is quite excellent except for the first repetition of the 93/07 ITO thin film. The uniformity of the abnormal 93/07 ITO thin film has a large value of 18 %, and that of others ITO thin films is lower than 11 %. The 95/05 ITO thin film can obtain the preferable uniformity of lower 6 %. The higher *UNI* (%) represents a higher ranging value between Rs_{max} and Rs_{min}, which means that the ITO thin films have poor sheet resistance uniformity. In terms of repeatability and uniformity of sheet resistance, high sheet resistance ITO thin films preferentially select the ingredient ratio of 95/05.

The change rate of sheet resistance of ITO thin films under high temperature (HH), high temperature and high humidity (HT&HH), and alkaline (AK) environments was calculated by the equation [12]:

$$\frac{Rs - Rs_0}{Rs_0} \times 100\% \tag{3}$$

where the Rs and Rs_0 is the sheet resistance of the post-tested and as-deposited ITO thin films. The change rate of sheet resistance as a function of tin content is shown in Fig. 7. It can be seen that the change rate of sheet resistance of all the ITO thin films under HT, HT&HH and AK conditions is lower than 12 %, which indicates that ITO thin films with different tin content have stable conductivity after suffering HT, HT&HH and AK surroundings. The better stability of sheet resistance attributes to the crystallization of the as-deposited ITO thin films at high growth temperature of 200 °C and a certain film thickness. The ITO thin films deposited at lower growth temperature show amorphous or weak crystallization, which results in instability of conductivity at HT, HT&HT and AK conditions [24]. When the thickness of ultrathin ITO thin films is larger than a certain value, the sheet resistance can keep better stability under HT, HT&HH and AK conditions [12].

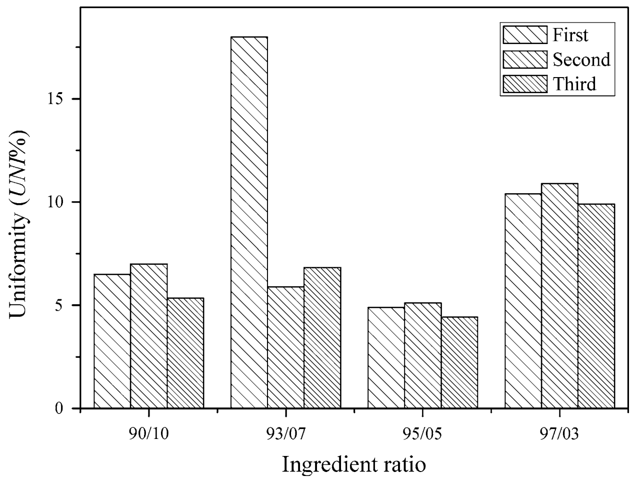

Fig. 6 Uniformity of sheet resistance of ITO thin films as a function of tin content and batches

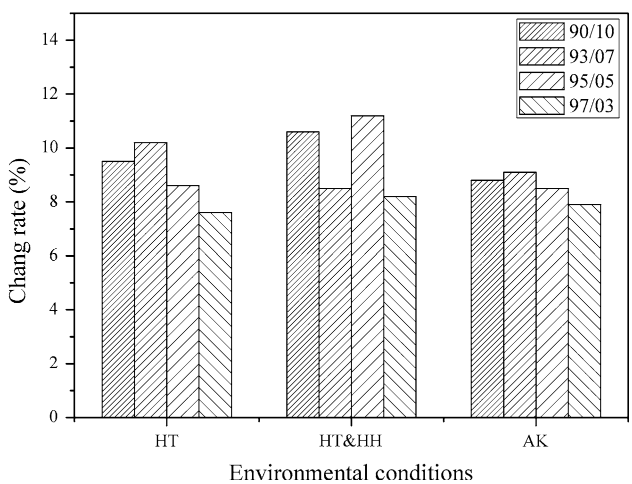

Fig. 7 Stability of sheet resistance of ITO thin films as a function of tin content under high temperature (HT), high temperature and high humidity (HT&HH), and alkaline (AK) conditions

The growth of ultrathin films is dominated by the initial film growth mode, which involves different stages of tunneling, percolation and linear ohmic growth. Isolated small islands grow gradually and coalesce, leading to the formation of continuous conduction paths at the percolation stage [25]. Therefore, the sufficient film thickness can obtain complete film structure and resist the influence from the external environment.

4 Conclusions

The touch panels oriented ITO thin films with high sheet resistance were deposited onto glass substrates by magnetron sputtering process. The influence of tin content on the structure, transmittance, sheet resistance, and repeatability, uniformity and stability of sheet resistance were investigated. The ultrathin ITO thin films show polycrystalline structure with the (222) preferred orientation and smooth surface with small surface roughness. The ultrathin ITO thin films results in ultrahigh transmittance of above 85 % in visible light range, and has flat transmittance curves without interference fringes. The high sheet resistance of 210 ± 30 Ω/□ can be obtained by using four kinds of ingredient ratio. ITO thin films with ingredient ratio of 95/05 show excellent repeatability and uniformity of sheet resistance, and can keep the electrical stability under HT, HT&HH and AK environmental conditions. The 95/05 ITO thin films can obtain the desired sheet resistance, and its uniformity and stability are lower than 6 and 12 %, respectively.

Acknowledgments This work is jointly supported by the National Science Foundation of China (21176051, 61361004), Guangxi Key Laboratory of Information Materials Foundation (1210908-204-Z, 131024-Z) and Guangxi Experiment Center of Information Science.

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