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Research Article

The Effect of Annealing on Nanothick Indium Tin Oxide Transparent Conductive Films for Touch Sensors

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This study aims to discuss the sheet resistance of ultrathin indium tin oxide (ITO) transparent conductive films during the postannealing treatment. The thickness of the ultrathin ITO films is 20 nm. They are prepared on B270 glass substrates at room temperature by a direct-current pulsed magnetron sputtering system. Ultrathin ITO films with high sheet resistance are commonly used for touch panel applications. As the annealing temperature is increased, the structure of the ultrathin ITO film changes from amorphous to polycrystalline. The crystalline of ultrathin ITO films becomes stronger with an increase of annealing temperature, which further leads to the effect of enhanced Hall mobility. A postannealing treatment in an atmosphere can enhance the optical transmittance owing to the filling of oxygen vacancies, but the sheet resistance rises sharply. However, a higher annealing temperature, above 250°C, results in a decrease in the sheet resistance of ultrathin ITO films, because more Sn ions become an effective dopant. An optimum sheet resistance of 336 Ω/sqr was obtained for ultrathin ITO films at 400°C with an average optical transmittance of 86.8% for touch sensor applications.

1. Introduction

Transparent conducting oxide (TCO) thin films have drawn a great deal of attention in recent years and have been widely applied in various optoelectronic devices such as solar cells [1, 2], flat panel displays [3, 4], organic light emitting devices (OLED) [5–7], and a variety of handheld devices. Until now, tin-doped In_2O_3 (indium tin oxide, ITO) has been the most widely used of the TCO materials because of its low resistivity (less than 10^{-3} Ω-cm) and good optical transmittance (more than 80%) in the visible region [8]. It is a degenerate n-type semiconductor with a wide energy band gap (3.7 eV) and possesses the qualities of high mechanical hardness and chemical inertness [9].

In previous studies, ITO films have been deposited using a variety of techniques such as ion beam assisted deposition [10], direct current (dc) magnetron sputtering [11–14], and chemical vapor deposition [15]. A pulsed dc magnetron sputtering method is the most common technology for the deposition of ITO films, because it is an easy way to get high quality thin films [16]. Using these techniques, the properties of ITO films are dependent on the process parameters like the oxygen partial pressure, substrate temperature, and postannealing temperature and different substrates, such as glass and PET [17–20].

The thickness of ITO films from 40 nm to 2.58 μm has been discussed in several studies. The results indicate that the electrical properties increase with increasing film thickness. However, the physical properties of the ITO film are notable when the thickness is less than 40 nm. In conclusion, we can say that the thickness is the most significant factor influencing the crystallization [21–27]. The main conductive mechanism of ITO films can be attributed to Sn-doping and oxygen vacancies to provide more free electrons. However, the leading contributor to the carrier concentration in amorphous ITO films is indistinct [28]. A postannealing treatment is an attractive way to improve the crystalline and other properties of ITO films, because it is a simple and low-cost process. According to a related study, an ITO film of 200 nm thickness can be achieved by dc magnetron sputtering at room temperature. The lowest resistivity after being treated by rapid thermal annealing (RTA) at 600°C in a vacuum is about 1.6×10^{-4} Ω-cm [29]. Gheidari et al. focused on the effect of the sputtering pressure and annealing temperature on the properties of ITO films. They found that the deposition rate decreases above 30 mTorr and the best conductivity and transmittance and larger grain size are achieved with an annealing temperature of 400°C [30].

Touch panels have become an important type of human-computer interface in recent years, and the ITO is the most commonly used material for this component. It functions as a transparent conductivity oxide, which requires an optical transmittance of more than 85% and high sheet resistance between 300 Ω/sqr and 500 Ω/sqr [31, 32]. However, in industrial practice ITO films are fabricated at room temperature with a postannealing treatment below 250°C in order to lower production cost. The fabrication cost can be reduced if ultrathin ITO films are used because less of the costly scarce rare element indium is used in its manufacture. However, the optical and electrical properties of ultrathin ITO films under an annealing temperature of 250°C are not well understood. This study focuses on postannealing treatment under the atmosphere for ultrathin ITO films deposited by dc pulsed magnetron sputtering at room temperature and what is needed to achieve high sheet resistance and high transmittance for touch sensor applications. The effect of the annealing temperature on the structure, electrical, and optical properties will be investigated, with a discussion of the properties of ultrathin ITO films.

2. Experimental Details

2.1. Film Preparation. ITO thin films were coated onto B270 substrates that were $25.4 \times 25.4\,\text{mm}^2$ and 1 mm thick by a pulsed dc magnetron sputtering system at room temperature. Figure 1 shows a schematic representation of the pulsed DC magnetron sputtering system. This system consisted of a deposition chamber with two magnetron sputtering cathodes. There was a pulse generator with a frequency of 20 kHz located between the dc power supply and the sputtering cathode. The pulse generator can help decrease the arcing and maintain stable plasma. The sputtering target was made of In_2O_3 mixed with 10 wt.% SnO_2 powder. An ITO target 75 mm in diameter was mounted on one cathode and set about 80 mm below the substrate.

The sputtering chamber was pumped down to a base pressure of less than 8×10^{-6} Torr by a cryopump. During deposition, argon (Ar) was directly injected as the working gas and the working pressure was set to $2\text{-}3 \times 10^{-3}$ Torr. The major goal of this paper is to study the effects of different annealing temperatures on the optical and electrical properties of ultrathin ITO thin films. The ITO films were prepared with 100 W of sputtering power at room temperature for the same amount of time after which the films were annealed at various temperatures (200–500°C) in air for 1 h.

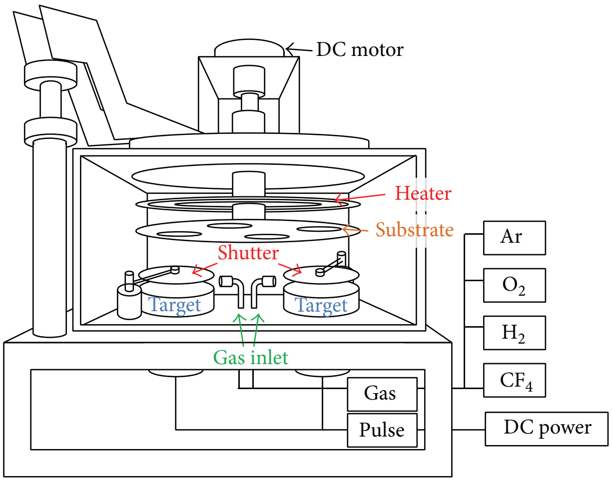

Figure 1: Schematic drawing of the experimental setup for pulsed dc magnetron sputtering.

2.2. Film Characterization. The thickness of all samples was about 20 nm as measured using a surface profiler (Dektak 8). The optical transmittance of thin films on B270 substrates was measured with a Hitachi U4100 spectrometer in the wavelength range from 300 to 700 nm. The crystalline structure was then examined through X-ray diffraction (XRD). The resistivity, Hall mobility, and carrier concentration of the films were determined by Hall measurements (HEM-300) at room temperature.

3. Results and Discussion

Ultrathin 20 nm thick ITO films were deposited on B270 substrates at room temperature by a pulsed dc magnetron sputtering system. To compare the effect of annealing on the properties of ultrathin ITO films, the as-deposited films were annealed in air at various temperatures of 200, 250, 300, 350, 400, 450, and 500°C. Figure 2 shows the carrier density, Hall mobility, and sheet resistance of the ultrathin ITO films at the various annealing temperatures, as obtained from the Hall Effect measurements carried out at room temperature. The electrical properties depend on the postannealing treatment. The crystallite size in the ITO films becomes larger as the annealing temperature increases, resulting in a decrease in boundary scattering and increase of the carrier lifetime [30]. However, the sheet resistance of the ultrathin ITO films increases with an increased annealing temperature, reaching a maximum value of 1075 Ω/sqr at an annealing temperature of 250°C. After this, the sheet resistance decreases with increasing temperature. This is an interesting result which differs from the results for ITO films of more than 40 nm. When as-deposited films are annealed in air, the free oxygen easily fills up oxygen vacancies in the lattice of the In_2O_3 structure, because of the ultrathin thickness of the ITO films. This reaction will decrease the oxygen vacancies and carrier concentration, which leads to an increase in the sheet resistance of the ultrathin ITO films. This also contributes to oxidation, resulting in an increase of the transmittance, as

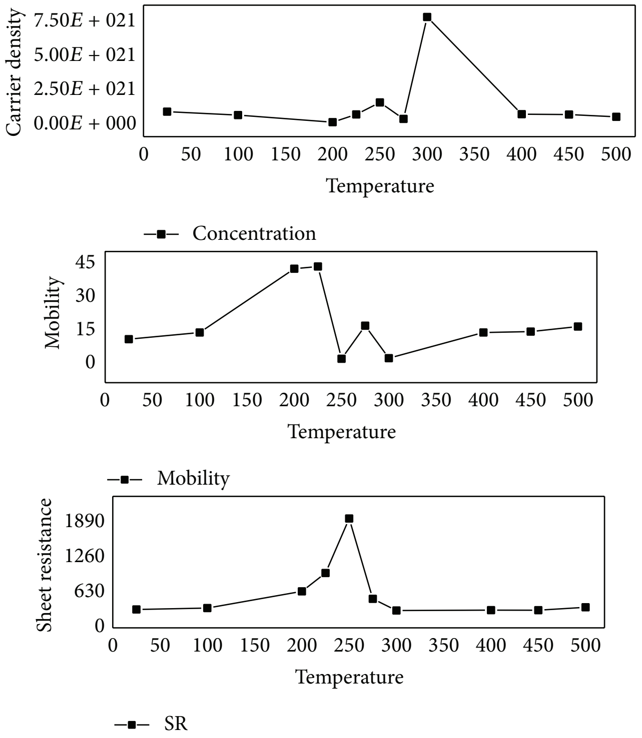

Figure 2: Comparison of the electrical properties of ultrathin ITO films as a function of the annealing temperature.

shown in Figure 3. Figure 3 shows the transmittance spectra of ultrathin ITO films before and after annealing at different temperatures. The average transmittance in the visible region (400–700 nm) before annealing is 83.84%, but this increases to over 85% after annealing at over 250°C. The inset to the figure shows an obvious increase in the transmittance with the increase in annealing temperature. This result corresponds with the aforementioned Hall measurements.

In addition to oxidation to fill up the oxygen vacancies, there is another possibility for the improvement of transmittance in ITO films which is also attributed to good crystallinity. However, the ultrathin ITO films do not easily crystallize at low annealing temperatures. The crystallization of the ultrathin ITO films is strongly dependent on the higher annealing temperature and the phase change from amorphous to crystalline during the annealing treatment, as shown in Figure 4. The transmittance of the crystalline structure is higher than that of the amorphous structure and this implies that the transmittance in the visible light range of ultrathin ITO films is closely related to the film structure. The grain-boundary scattering mechanism in polycrystalline ITO films has been discussed in a previous report [30]. The crystalline structure of the films can be improved and the crystallite size increased after annealing, which can decrease the scattering of incident light by decreasing the number of grain-boundaries and enhancing the transmittance. The crystallinity of ultrathin ITO films is higher, which further results in the effect of enhanced Hall mobility.

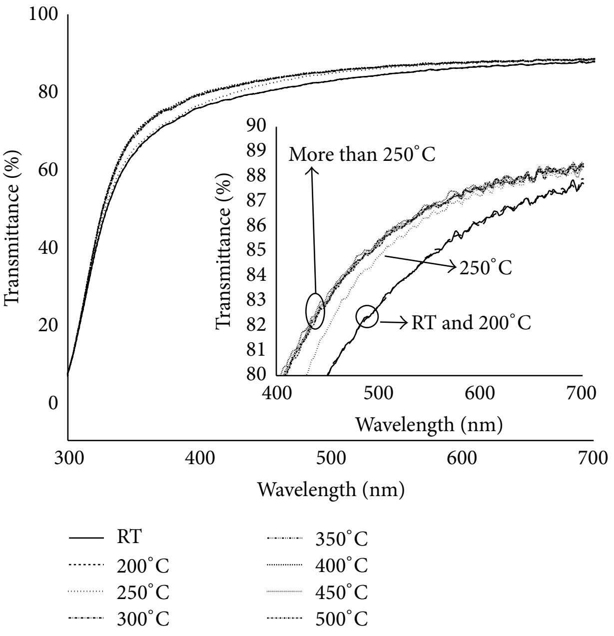

Figure 3: Optical transmittance of ultrathin ITO films during annealing treatment with various annealing temperatures.

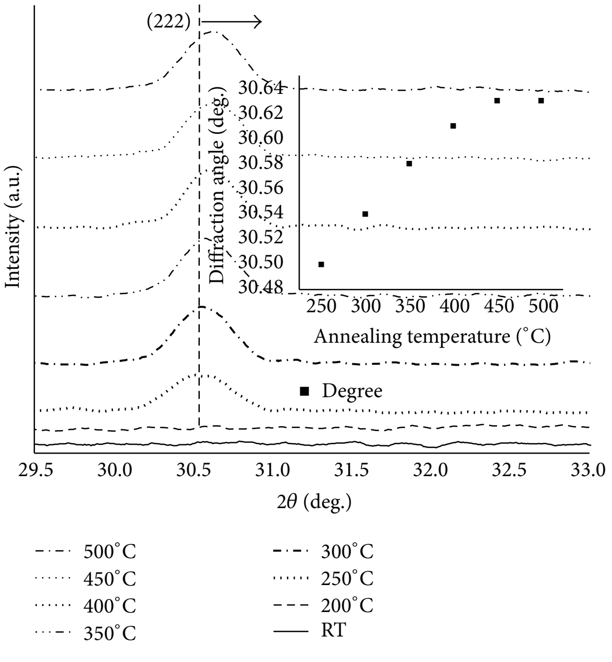

Figure 4: X-ray diffraction peaks and diffraction angles of ultrathin ITO films during annealing treatment with various annealing temperatures.

The results discussed above seem to contradict the sheet resistance of the Hall measurement when the annealing temperature is more than 250°C. This is a point worthy of discussion. A higher annealing temperature leads to a decrease in the sheet resistance of ultrathin ITO films. This is due to the fact that the annealing treatment rearranges the structure of the ITO films, causing more Sn ions to become an effective dopant. This is demonstrated by the XRD measurements. It can be seen in the inset to Figure 4 that the (222) peak intensity of the ITO films becomes higher with the increase of annealing temperature. Consequently, there is a decrease in the sheet resistance because of the replacement of the In^{3+} by Sn^{2+} when the annealing temperature exceeds 250°C. According to Bragg's law, the Bragg angle (θ) increases due to a decrease in the d spacing. The length of the d spacing is expected to be shorter if the In ions are replaced by Sn ions, due to the smaller ionic radius of Sn (the ionic radii of In^{3+} and Sn^{4+} are 80 and 69 pm, resp.). This is also consistent with the results obtained for the electrical properties and the increase in carriers owing to the more effective dopant. Furthermore, the more effective dopant acts as a donor source to increase the mobility of ITO films. In this study, the best conditions for ultrathin ITO films for touch sensor applications offer a sheet resistance of 336 Ω/sqr and an optical transmittance of 86.8% in the visible region when the annealing temperature is 400°C.

4. Conclusion

Ultrathin ITO films were prepared on B270 glass substrates at room temperature by the dc pulsed magnetron sputtering method. The effect of the annealing temperature on ultrathin ITO films was investigated. As the annealing temperature is increased, the structure of the ultrathin ITO film changes from amorphous to polycrystalline. The postannealing treatment in the atmosphere can enhance the optical transmittance owing to a filling up of the oxygen vacancies, but there is also a sharp rise in the sheet resistance. A higher annealing temperature of above 250°C results in a decrease in the sheet resistance of the ultrathin ITO films because of more Sn ions, to become an effective dopant. The crystallinity of ultrathin ITO films becomes higher with an increase of annealing temperature, which further leads to the effect of enhanced Hall mobility. In terms of the sheet resistance and transmittance, an annealing temperature of 400°C is the best for our ultrathin ITO samples. A suitable sheet resistance of 336 Ω/sqr was obtained for ultrathin ITO films at 400°C with an average optical transmittance of 86.8% making it suitable for applications in touch sensors.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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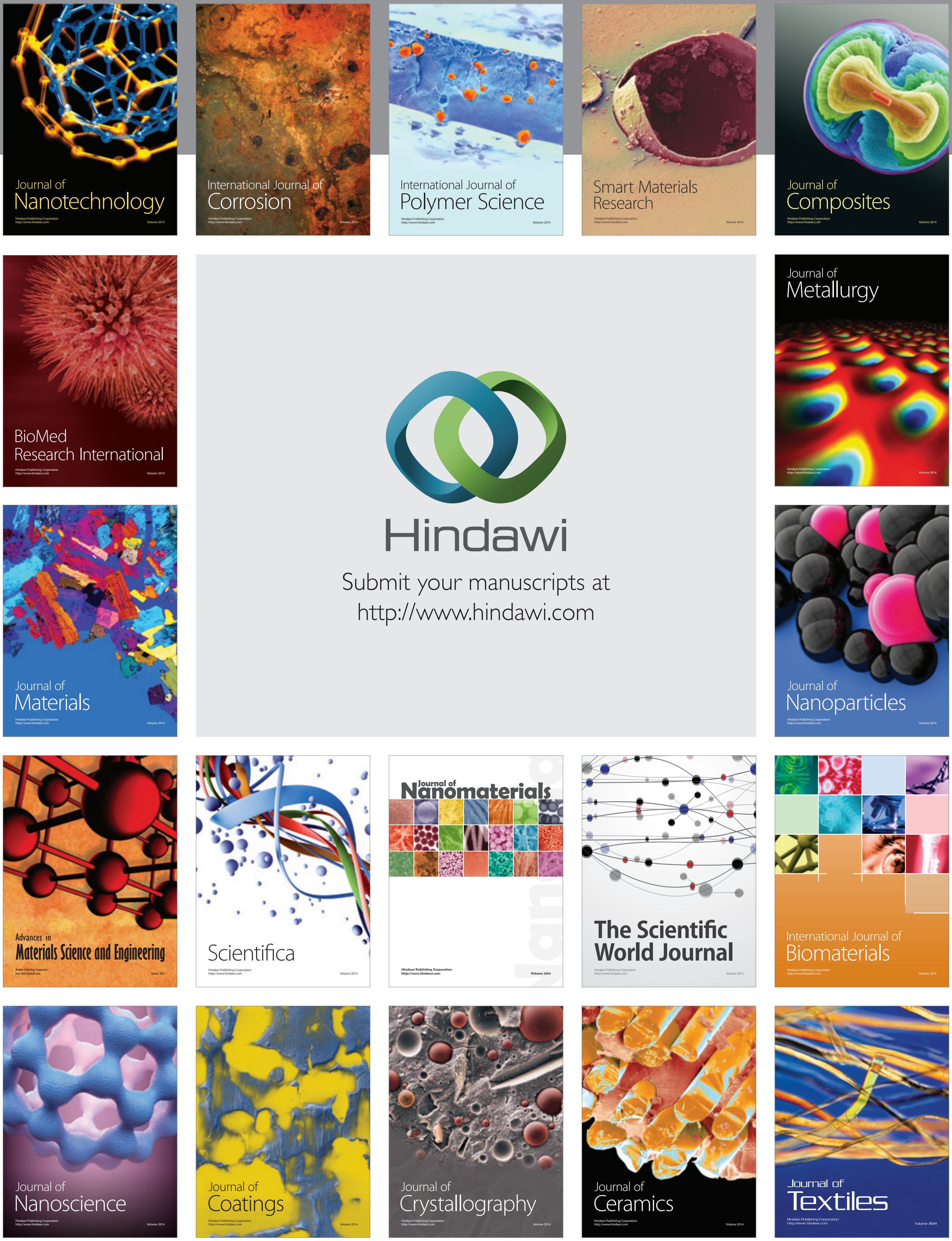
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