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Characteristics of indium tin oxide films deposited by r.f. magnetron sputtering

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Abstract

Highly conductive indium tin oxide (ITO) films were deposited by r.f. magnetron sputtering using ITO targets. The composition of the ITO targets was 90% indium oxide and 10% tin oxide. ITO films were deposited on 1 mm thick soda lime glass. Films deposited at substrate temperature of 300 °C, exhibited resistivities as low as 1.3×10^{-4} ohm cm^{-1}. Annealing of the ITO films in air for 2 h was necessary for achieving low resistivities. X-ray diffraction and transmissivity tests were carried out to study the effects of annealing. Lowest resistivity and highest transmission were found to occur at an annealing temperature of 350 °C. X-ray diffraction measurements revealed that the as deposited film had a strongly (222) oriented cubic structure. Annealing relieved the as deposited tensile strain and increased crystal perfection.

Keywords: Indium oxide; Resistivity; Sputtering; Tin oxide

1. Introduction

Indium tin oxide (ITO), a mixture of In_2O_3 and SnO_2, is a degenerate n-type semiconductor with low resistivity (less than 10^{-2} ohm cm) and a wide band gap (3.7 eV) which produces high transparency in the visible light region. Because of its high conductivity and transmissivity ITO finds various applications in optoelectronic devices like solar cells and flat panel displays [1-2].

Various deposition techniques, from low pressure to high vacuum, have been used to deposit ITO thin films [3-7]. Yao et al. fabricated ITO thin films by thermally evaporating indium and tin sequentially onto substrates kept at room temperature and subsequently annealing them in air to improve their transparency and conductivity [6]. Kulkarni et al. used a novel technique of layer-by-layer deposition of indium and tin by electron beam deposition and subsequent annealing in oxygen to form indium tin oxide [7]. However, films with the highest transmission and lowest resistivity have been prepared by sputtering. Kawada [8] has reported on ITO films deposited from metallic In-Sn alloy targets in a reactive sputtering process. Several researchers have investigated the deposition of ITO by d.c. sputtering [9-10], but very little data is available on ITO deposition by r.f. magnetron sputtering.

In this paper we report on the r.f. magnetron sputtering deposition of ITO on glass substrates using ITO targets and compare the resistivities of r.f. and d.c. sputtered films. Also the effects of annealing on the electrical and optical properties of ITO are presented.

2. Experimental procedure

Experiments were carried out in a Kurt J. Lesker sputtering system using magnetron cathodes of the type Torus-2C. Composition of the hot pressed ITO target was 90 wt% In_2O_3 and 10 wt% SnO_2. ITO target size was 2.0 in dia x 1/8 in thick. ITO films were deposited on 1 mm thick soda lime glass substrates. These substrates were precleaned with methanol and acetone. Before deposition the substrates were preheated to 300 °C. Listed below are the main process parameters that were

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used. Higher sputter pressure and sputter power were used for d.c sputtering as the plasma could not be maintained within the chamber at low pressure and power settings.

Deposition temperature: 300 °C; sputter pressure: 5.32 $\times$ 10^{-6} pascals (r.f.); 10.64 $\times$ 10^{-6} pascals (d.c.); sputter power: 40 W (r.f.); 75 W (d.c.); argon flow rate: 53 ml min^{-1}; oxygen flow rate: 0.0–2.4 ml min^{-1}.

After sputter deposition, films were annealed in air for 2 h. Film thickness was measured with Dektek™ surface profile measuring system and sheet resistance was measured with a VEECO FPP-100 four point probe. X-ray diffraction data were taken with a SCINTAG XDS-2000 diffractometer. Transmission was measured with ARC-275 monochromator.

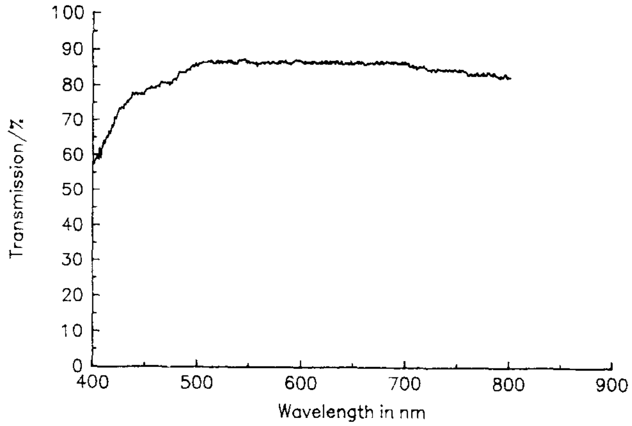

Fig. 2. Transmission vs. wavelength for r.f. sputtered ITO film; oxygen/argon flow rate was 3.58% during sputtering; annealing temperature was 350 °C; time of anneal was 2 h and ambient was air.

3. Results

3.1. Oxygen flow rate

Fig. 1 shows the specific resistivity versus the oxygen flow rate for 100 nm thick ITO films deposited by r.f. sputtering and d.c. sputtering. Oxygen flow rate was varied between 0–2.4 ml min^{-1} and its effect on the resistivity of the film was observed. All these films were annealed in air at 350 °C. The influence of oxygen flow rate on resistivity can be explained in terms of two competing effects [10]. Increase in oxygen flow rate enhances the growth of crystalline phases of ITO films which leads to higher mobility of carriers. On the other hand increased oxygen content decreases the carrier density. It is seen in Fig. 1 that lowest resistivity was achieved at the oxygen/argon flow rate of 3.58%. Fig. 2 shows transmission versus wavelength for a r.f. sputtered ITO film which was annealed at 350 °C, and the oxygen/argon flow rate was 3.58% during sputtering. Similar measurements were carried out for different

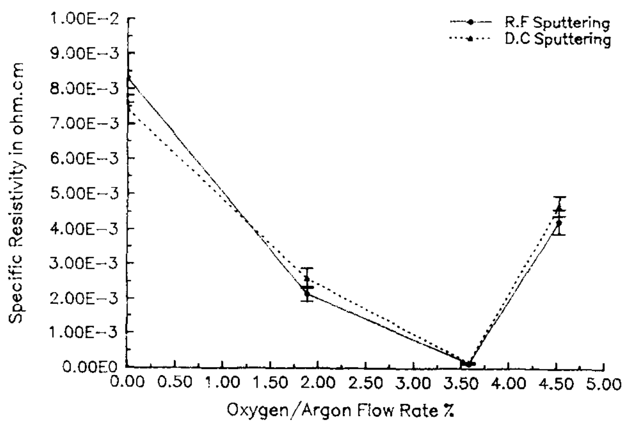

Fig. 1. Specific resistivity of sputtered ITO films annealed in air for 2 h at 350 °C as a function of oxygen/argon flow rate percentage during sputtering.

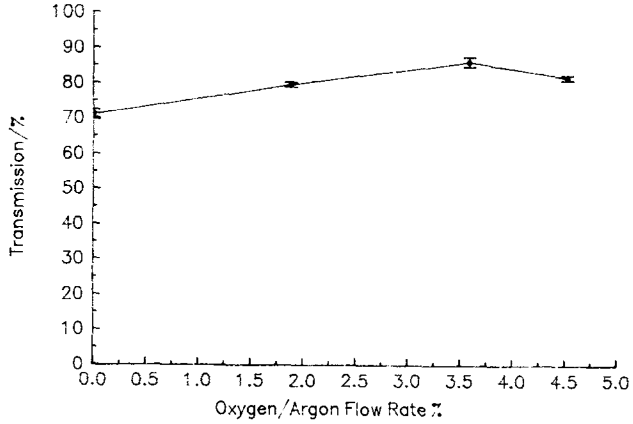

Fig. 3. Maximum transmission (400–700 nm range) vs. oxygen/argon flow rate percenatge for r.f. sputtered ITO films.

oxygen flow rate values and the maximum value of transmission is plotted against the flow rate in Fig. 3. It is clear that transmissivity of the films improves as oxygen flow rate is increased and films with maximum transmission of 87% (400–700 nm range) were produced at oxygen/argon flow rate of 3.58%.

3.2. Annealing study

Annealing temperature was varied from 0 °C to 450 °C and its effect on specific resistivity and transmission was studied. Fig. 4 shows how specific resistivity varies with increase in annealing temperature; lowest resistivity was achieved at 350 °C. Fig. 5 shows the effect of annealing temperature on transmission and it is observed that with increase in annealing temperature transmission improves.

3.3. X-ray diffraction study

X-ray diffraction measurements using Cu Kα radiation were performed to study the effect of annealing

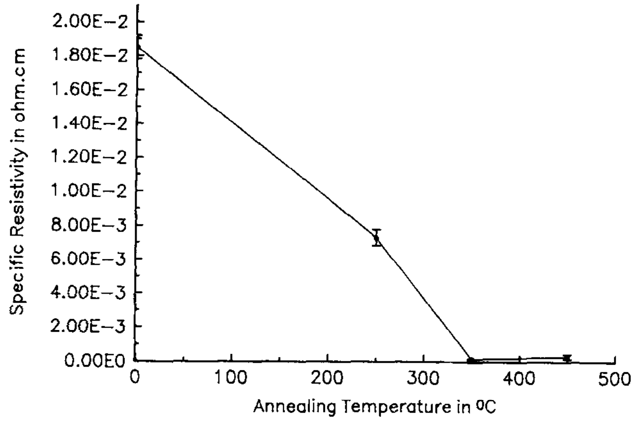

Fig. 4. Specific resistivity vs. annealing temperature for r.f. sputtered ITO films; oxygen/argon flow rate was 3.58% during sputtering; time of anneal was 2 h and ambient was air.

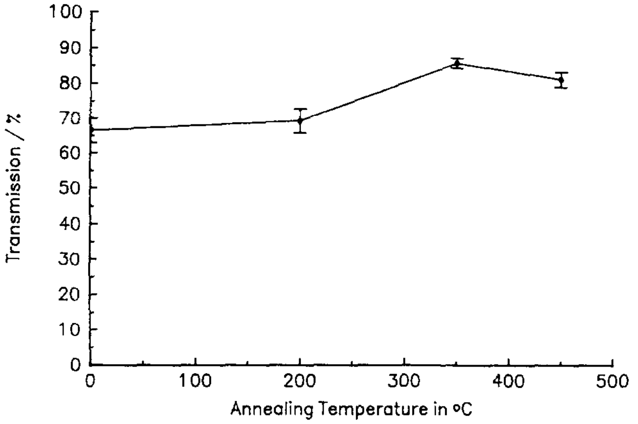

Fig. 5. Transmission vs. annealing temperature for r.f. sputtered ITO films; oxygen/argon flow rate was 3.58% during sputtering; time of anneal was 2 h and ambient was air.

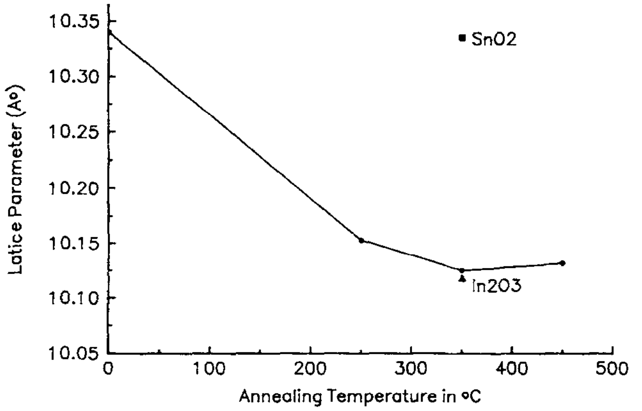

Fig. 6. Lattice parameter vs. annealing temperature.

temperature on the ITO films. For as deposited films, only the (222) peak was observed indicating a strong preferred orientation. The lattice parameter was 2% larger than that of In_2O_3 but decreased (Fig. 6) to that of In_2O_3 after annealing at 350 °C indicating stress relaxation. Annealing slightly decreased the preferred

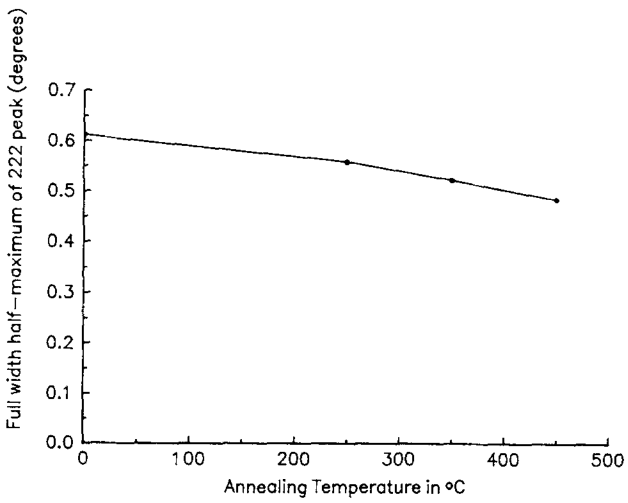

Fig. 7. Full width half maximum for 222 peak vs. annealing temperature.

orientation as indicated by the appearance of a (400) line. The full width at half maximum (FWHM) of the (222) peak is plotted as a function of annealing temperature in Fig. 7; FWHM decreased as crystallite perfection improved.

4. Conclusions

Indium tin oxide films of low resistivity (1.3×10^{-4} ohm cm^{-1}) and high transmission (87%) were deposited on glass by r.f. magnetron sputtering in argon and oxygen. Film characteristics were very sensitive to the oxygen flow rate during sputter deposition and to the annealing temperature. Annealing in air for 2 h at 350 °C reduced the resistivity of the ITO films and increased the visible transmittance.

Specific resistivity values of the ITO films fabricated by d.c. and r.f. sputtering methods were quite close to each other.

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