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ScienceDirect
Thin Solid Films 516 (2008) 1386 – 1390
www.elsevier.com/locate/tsf

Effects of oxygen concentration on the properties of sputtered SnO_2:Sb films deposited at low temperature

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Available online 24 May 2007

Abstract

Antimony doped tin oxide (SnO_2:Sb) films have been prepared by d.c. magnetron sputtering and the properties of the films depend on deposition conditions, such as O_2 gas ratio, were investigated. The gas composition was found to affect the properties of the films. With the incorporation additional oxygen, the electrical and optical properties of films significantly improved. The minimum value of resistivity of the films was 4.9×10^{-3} Ω cm at the oxygen concentration of 30% and the optical transmittance was over 80%.

Keywords: Tin oxide (SnO_2); Transparent conducting oxide (TCO); Sputtering; Oxygen gas ratio

1. Introduction

Doped tin oxide (SnO_2) thin films have been widely used as transparent electrodes in various fields such as solar cells, optoelectronic devices, flat panel displays, heat mirrors, and gas sensor owing to its high transmission and conductivity [1–7]. In addition, SnO_2 films are more chemically stable than other transparent conducting oxide (TCO) films such as zinc oxide (ZnO) and Sn-doped In_2O_3 (ITO) [8,9]. Various methods have been used to deposit these films. Particularly, sputtering techniques can be used to prepare large areas with high yields and competitive costs as compared with other deposition techniques. However, there is a little given in the literature about tin oxide films prepared by magnetron sputtering.

The sputtering method often requires heating the substrate at an elevated temperature during the film preparation or an additional post-annealing treatment. High temperature processes are unsuitable for some applications. For instance, the organic color filter coated substrates for flat panel displays, the flexible displays made with polyester, polyethylene terephthalate (PET) and other plastic foils are not compatible with a high temperature plasma process. Therefore the development of high quality SnO_2 films with at low processing temperatures is quite a challenge indeed. Lower-temperature SnO_2 thin films have been typically produced by d.c. (or r.f.) sputtering and vacuum reactive evaporation [10–14].

In the work, SnO_2:Sb films for the low temperature devices have been prepared by DC magnetron sputtering. The effects of O_2-to-Ar gas ratio during deposition process on the structural, electrical and optical properties of the SnO_2:Sb films have been investigated.

2. Experimental

The SnO_2:Sb films studied in this work were prepared by conventional d.c. magnetron sputtering in a versatile coater. Sintered SnO_2 disc with Sb_3O_5 content of 5 wt.% were used as a target. The substrates were Corning 7059 glass and were placed on a substrate holder having heating block. The distance between the substrate and target was 40 mm. Sputter deposition was carried out at pressure of 5 mTorr in pure Ar gas or a mixture of Ar and O_2. O_2-to-Ar gas ratio varied from 0% to 50% and the sputter gas was supplied via mass-flow-controlled gas inlets. Before the deposition of the films pre-sputtering was carried out for a sufficiently long time to remove the hydrolyzed surface layer of the target. The substrate temperature was measured by a thermocouple directly clamped onto the substrate holder and was varied from room temperature to 200 °C during the deposition. The film thickness, determined by surface profiler system, was about 100 nm.

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doi:10.1016/j.tsf.2007.05.027

X-ray diffraction (XRD) measurements were performed over a diffraction angle range of 20° to 70° to study the preferential orientation and crystallinity of SnO_2:Sb film. The films were analyzed by X-ray photoelectron spectrometer (XPS) using a XSAM800 (KRATOS). The sheet resistance of the samples was measured using a four-point probe and the resistivity of the film was calculated. Carrier concentration and Hall mobility were obtained from Hall-effect measurement by using the Van der Pauw technique. The optical transmittance and reflectance of the films were measured with an ultraviolet–visible-near-infrared spectrophotometer (Hitachi U-3410, Japan) and a Fourier-transform infrared (FTIR) spectrophotometer.

3. Results and discussion

Fig. 1 shows the effects of the gas composition on the deposition rate of SnO_2:Sb films deposited at the substrate temperatures of 100 °C and 200 °C. The maximum deposition rate achieved in pure argon, was about 4.5 nm/min. The deposition rate decreases with increasing oxygen concentration because of the lower sputtering yield and large polarizability of the oxygen ion [15]. The use of oxide target in place of metallic targets helps in controlling the stoichiometry of the films more precisely.

Fig. 2 gives the X-ray diffraction patterns of SnO_2:Sb films deposited at different O_2-to-Ar gas ratio. Films grown in pure argon have no diffraction peaks, regardless of substrate temperature. With the addition 5% oxygen, the SnO_2:Sb films exhibit two peaks corresponding to (101) and (211) plane of SnO_2 phase. Especially, for the substrate temperature of 200 °C, the (101) peak is dominant. A preferred (101) orientation of SnO_2:Sb films at low oxygen concentration has been reported by Suzuki and Mizuhashi [15]. They explained that this preferred (101) orientation may be a characteristic structure of the magnetron sputtering. However, Ma et al. [16] reported the (110) preferential orientation for their sputter deposited SnO_2:Sb films on glass and polyimide substrates. The intensity of (101) diffraction peak decreases with increasing oxygen concentration and then almost completely disappear at high O_2 concentration. In addition to (101) and (211) peaks, new (110) and (200) diffraction lines are observed at 10% O_2. The (110) diffraction line increases in intensity with oxygen concentration and becomes very strong at high O_2-to-Ar gas ratio, while (200) peak sharply decreases. This indicates that the degree of the (110) preferential orientation of SnO_2:Sb film increases as the oxygen concentration is raised. It should be noted that the (102) diffraction peak of SnO phase is observed at low O_2 concentration. This peak, however, highly decreases with increasing oxygen concentration and disappears at the O_2-to-Ar gas ratio more than 20%, as shown in Fig. 2. It must be noted that for films deposited at T_s=200 °C, the (102) peak of the SnO phase appeared again at a very high O_2 concentration (50% O_2). However, the substrate temperature didn't have a marked influence on the structure of the films.

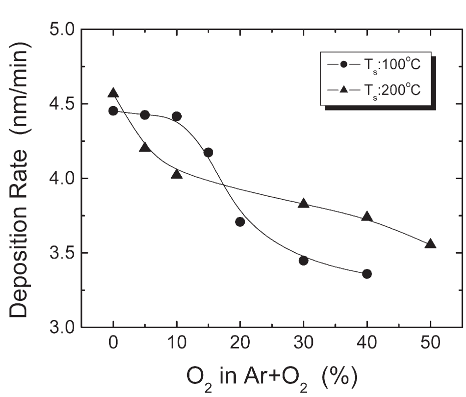

Fig. 1. Deposition rate of SnO_2:Sb films as a function of oxygen concentration in the sputtering gas.

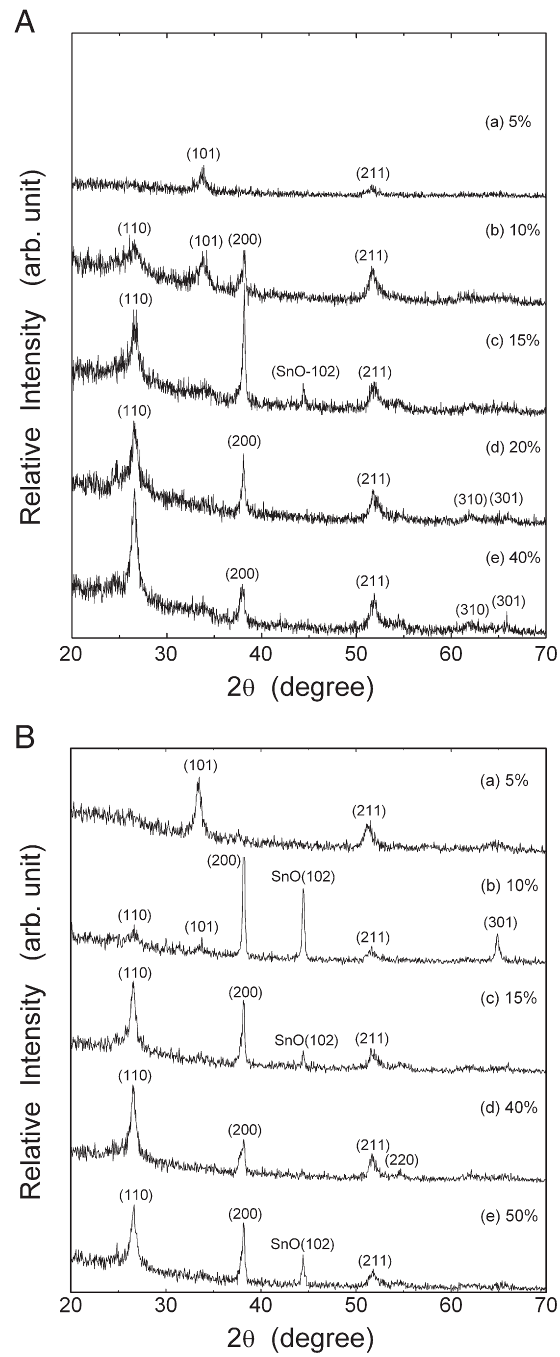

Fig. 2. X-ray diffraction patterns of SnO_2:Sb films deposited at various gas compositions: (A)T_s=100 °C; (B)T_s=200 °C.