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P-type tin–indium oxide films prepared by thermal oxidation of metallic InSn alloy films

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Abstract

P-type transparent conducting tin–indium oxide (TIO) films were successfully fabricated on quartz substrates by thermal oxidation of InSn alloy (In / Sn = 0.2) films that were deposited by magnetron sputtering at room temperature (R.T.). Structural and electrical properties of TIO films were investigated. X-ray diffraction studies showed that all TIO films were polycrystalline with an orthorhombic structure. The surface morphology of TIO films viewed by field emission scanning electron microscope (SEM) revealed that the films are composed of uniformly distributed submicron grains. Hall effect measurement results indicated that hole concentration as high as 9.61×10^{18} cm^{-3} was achieved. It's found that 600 °C was the optimum thermal oxidation temperature to get p-type TIO films with highest hole concentration.

Keywords: Indium tin oxide; Magnetron sputtering; P-type conductivity; Transparent conducting oxide

1. Introduction

Tin dioxide (SnO_2) is a kind of n-type wide band gap semiconductor material with E_g = 3.6–4.0 eV [1–4]. Due to its low resistivity, high optical transparency, thermal and chemical stability, and low cost, SnO_2 is extensively used in many applications such as transparent electrodes, liquid crystal displays (LCD), gas sensors, solar cells, etc. [5–11]. Until now, most of the transparent conducting oxide (TCO) films are n-type, such as indium tin oxide (ITO) [12–14], fluorine doped tin oxide (FTO) [15,16], aluminum doped zinc oxide (AZO) [17,18]. Recently, great interests have been paid to the research of p-type TCO films. Kawazoe et al. reported p-type conductivity from copper aluminium oxide ($CuAlO_2$), but the hole concentration was low (10^{17} cm^{-3}) and the transmittance was only about 50% in the visible region [19]. Recently, a few papers reported on p-type conducting from indium–tin oxide films [20,21]. In these papers, Ji et al. prepared p-type transparent conducting indium tin oxide films (ITO) by sol–gel method, and found that the optimal In / Sn ratio for p-type conducting films was about 0.2, but the results were not so

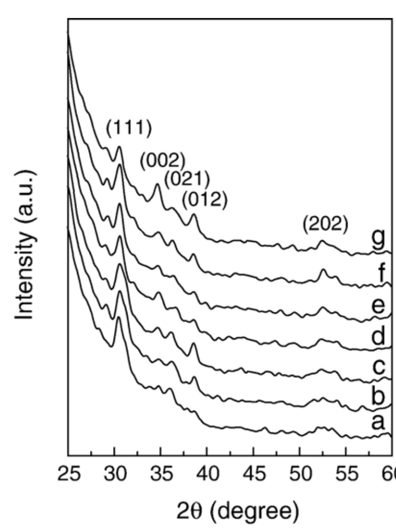

Fig. 1. XRD spectra of the films after thermal oxidation at a = 450 °C, b = 500 °C, c = 550 °C, d = 600 °C, e = 650 °C, f = 700 °C, g = 750 °C.

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satisfying as expected because of the poor film quality and thin thickness as limited by sol–gel method.

In order to improve the quality of the p-type TIO films, structural and electrical properties of the p-type conducting tin–indium oxide films prepared by thermal oxidation of InSn alloy films were studied, and improved crystal quality and electrical properties than the p-type ITO films reported by Ji et al. were achieved [20].

2. Experimental

The metallic InSn alloy films were deposited on quartz glass by DC magnetron sputtering. The target of $\phi = 75$ mm was fabricated by melting tin (purity 99.999%) and indium (purity 99.999%, In/Sn atomic ratio 0.2) together followed by subsequent machining. Before deposition the substrates were cleaned ultrasonically in acetone, rinsed with deionized water and dried by nitrogen blow. The base pressure of the deposition system was 1×10^{-3} Pa, during sputtering, the pressure increased to 1 Pa as the argon gas (purity 99.999%) was admitted into the chamber, and no oxygen was supplied. The sputtering was carried out at a cathode voltage of 240 V and an ion beam current of 100 mA at the target–substrate distance of 100 mm. The substrate was not heated intentionally during deposition, and no temperature rise of the substrate during the 5-min deposition was observed. After deposition, the alloy films were oxidized in air between 400 and 750 °C to form the stable TIO oxide films.

The structural characteristics were examined with a high resolution X-ray diffractometer of Bede plc D1 system that used a CuKα radiation ($\lambda = 1.54056$ Å). The surface morphology was taken with a SIRION-100 field emission scanning electron microscope (SEM) of FEI Company. The thickness of the film was measured from the cross-sectional view of the SEM image.

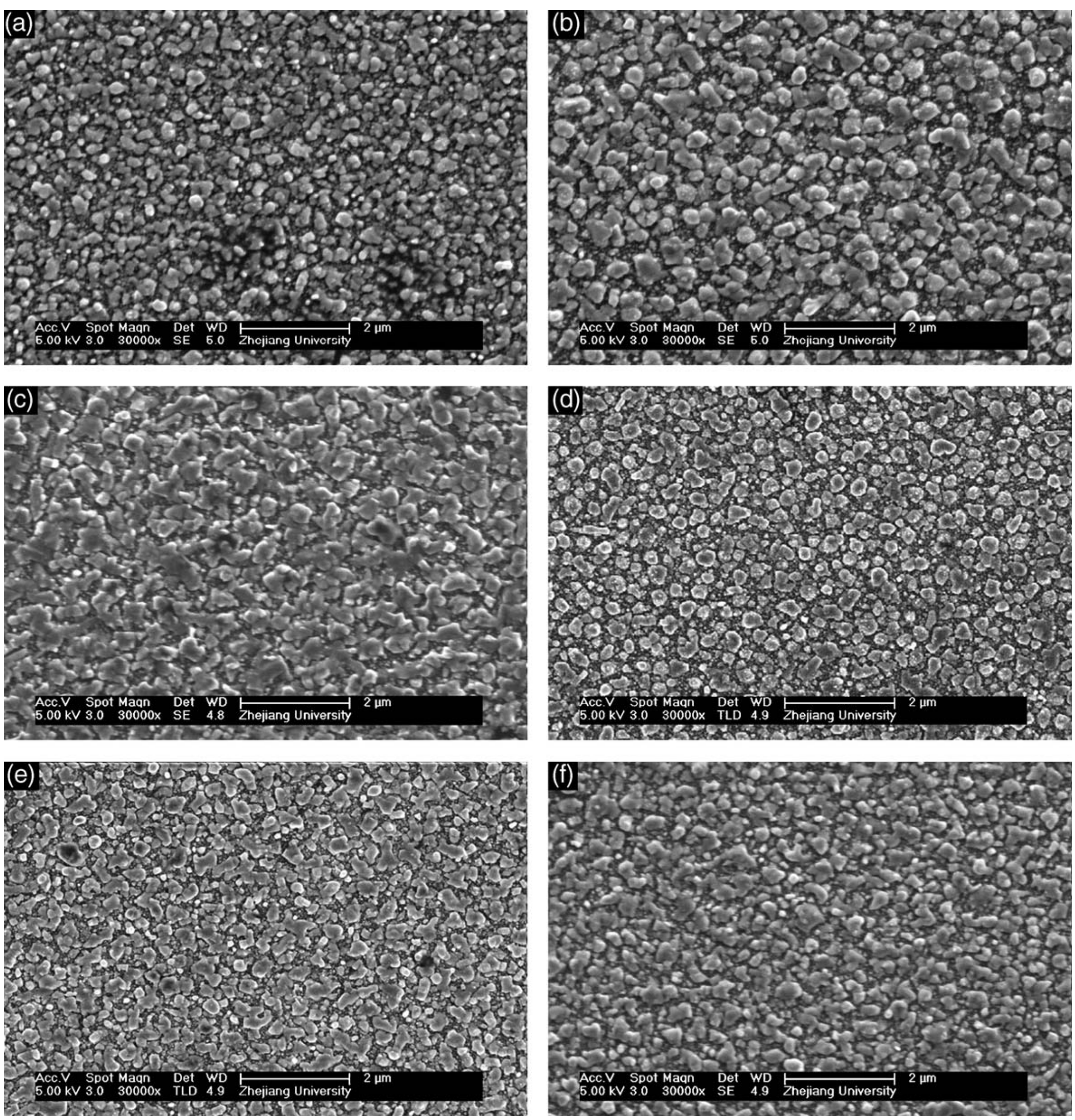

Fig. 2. SEM micrographs of the TIO films prepared at (a)=450 °C, (b)=500 °C, (c)=550 °C, (d)=600 °C, (e)=650 °C, (f)=700 °C.