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High rate deposition of tin-doped indium oxide films by reactive magnetron sputtering with unipolar pulsing and plasma emission feedback systems

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Abstract

Tin-doped indium oxide (ITO) films were deposited on the unheated alkali-free glass substrates (AN100) by reactive magnetron sputtering using an indium–tin alloy target with an unipolar pulsed power source feeding 50 kHz pulses and a plasma control unit (PCU) with a feed back system of oxygen plasma emission intensity at 777 nm. In order to achieve very high deposition rates, depositions were carried out in the 'transition region' between the metallic and the reactive (oxide) sputter modes where the target surface was metallic and oxidized, respectively. Stable depositions were successfully carried out in the whole 'transition region' with an aid of PCU. The lowest resistivity as the transparent ITO films deposited on unheated alkali-free glass was 7.5×10^{-4} Ωcm with the deposition rate of 650 nm/min. The electrical and optical properties of the films could be controlled systematically in the very wide range.

Keywords: Tin-doped indium oxide (ITO); Transparent conductive oxide (TCO); Reactive sputtering; Unipolar pulsing; Plasma emission; Transition region

1. Introduction

Several kinds of transparent conductive oxide (TCO) films have been widely used in the field of optoelectronics, such as in transparent metallization for various kinds of flat panel displays (FPDs) or solar cells. Sn-doped In_2O_3 (ITO) is an n-type, highly degenerate, wide-gap semiconductor which, due to its relatively low resistivity and high visible transmittance compared to the other TCO materials such as SnO_2 or ZnO, is at present overwhelmingly used as transparent electrodes in liquid crystal displays (LCDs), plasma displays and light emitting diodes (LEDs) [1–6]. Considerable effort has been focused on depositing ITO films of significantly low resistivity in order to accommodate the increasing technological demand for larger area flat panel displays with higher image quality [7–11].

The deposition method of ITO films employed so far for commercial productions has been the dc magnetron sputtering using an oxide ceramic target because of high stability and reproducibility. This method provides the low resistivity of about 10^{-4} to 10^{-3} Ωcm [7,8,11–14]. However, the deposition rate, one of the most important factors dominating the deposition cost, is not so high because of the low sputtering yield of the oxide. On the other hand, the reactive dc magnetron sputtering process using a metal or alloy target is considered to have higher potential for the large area coatings with very high deposition rates and low cost. The reactive sputtering process, however, is highly non-linear, where the deposition rate shows the 'hysteresis' with respect to the reactive gas flow rate [15–20]. Such a behavior originates from the oxidation state of the target surface, resulting in drastic changes in the sputtering yield. Therefore, in the 'transition region', the deposition rate decreases drastically with increasing oxygen gas flow rate depending on target surface conditions. Moreover, the reactive dc sputtering on the non-uniformly oxidized target surface causes the problem of abnormal discharges such as arcing during long time operations in the production. Dual magnetron sputtering (DMS) systems with mid-frequency pulsing of 50 kHz have been proposed for oxide or nitride films to reduce the arcing drastically even under high power density impression [21–23]. Two types of plasma control

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units (PCU) with a feedback system of either plasma emission intensity [21,22] or discharge impedance [23] have been reported in combination with the DMS systems for the very high rate deposition controlled stably in the 'transition region'. However, the DMS system needs two cathodes and two power supplies, which make it difficult to be reversible with a conventional single target production systems.

In this study, a reactive dc magnetron sputtering system with unipolar pulsed powering and the PCU was adopted to deposit ITO films, which is much simpler than the DMS system. The reactive sputter depositions in the 'transition region' were carried out in order to obtain high quality ITO films at around room temperature and at very high deposition rates. The structures and electrical properties were investigated in relation to various deposition parameters.

2. Experimental details

ITO films were deposited on unheated alkali-free glass substrates (AN100, Asahi Glass) by reactive sputtering with unipolar pulsing unit and plasma control unit (PCU, FEP: Fraunhofer Institut fur Elektronenstrahl- und Plasmatechnik). The schematic illustration of this system is shown in Fig. 1. This system consists of one magnetron cathode (RM400, FEP), specially designed hidden anodes, dc power source (pinnacle 6 kW, Advanced Energy) with mid-frequency pulse unit (UBS-C2, FEP) and PCU with feedback system of plasma emission intensity on the reactive gas flow. A planar In–Sn alloy target (Sn: 10 at.%, 130 mm × 400 mm in size) was connected with the UBS-C2 which was operated in the unipolar pulse mode. The mid-frequency pulse unit possesses the approximate shape of a square wave with frequency of 50 kHz and duty cycle of 80%, which make it possible to impress very high power density without arcing at the cathode. The hidden anode was designed to be an effective anode to keep the discharge stably without any surface poisoning during the depositions. Reactive and sputtering gases were O_2 (purity: 99.999%) and Ar (purity: 99.999%), respectively. The reactive gas flow was precisely controlled by piezoelectric valves under the control of PCU. The plasma emission of the atomic O* line at 777 nm was led into the optical emission detector (containing a photomultiplier and optical filter) to transform the optical intensity into the photovoltage. The PCU adjusted the piezoelectric valve to control O_2 flow precisely and rapidly until the photovoltage is equal to the set point value for the optical emission intensity (OEI). The set points could be chosen in the 'transition region' between the metallic and the reactive (oxide) sputter mode in order to achieve high deposition rate. The chamber was evacuated down to a pressure of less than 9.0×10^{-4} Pa. The second control loop kept the total gas pressure (P_{tot}) constant at 0.5 Pa by adjusting the argon gas flow. The 'pre-pre sputtering' was carried out only with argon for 20 min in order to remove the oxide layer at the target surface and the 'pre-sputtering' was carried out with reactive gas for 10 min under the same conditions during deposition. The set points of the OEI of the O* line at 777 nm were selected between the photovoltage of 2 and 9 V. The deposition parameters are listed in Table 1.

All the depositions were carried out with dc power of 5 kW on the unheated glass substrate which was fixed in front of the target (Fig. 1). The substrate surface temperature after the depositions was lower than 70 °C confirmed both by thermo-couple fixed at the substrate surface and by the radiation thermometer, where the both measurements indicated the same values.

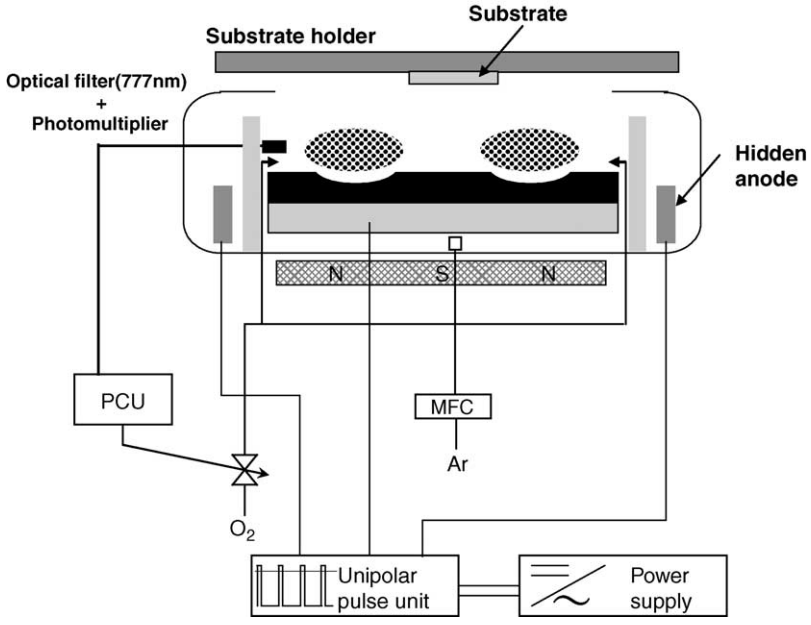

Fig. 1. Schematic illustration of the reactive magnetron sputtering system with unipolar puls unit and plasma control unit (PCU), where the plasma emission of the atomic O* line at 777 nm was led into a photomultiplier through optical filters and the optical emission intensity (OEI) was used to control reactive gas (O_2) flow.