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Amorphous indium tin oxide films deposited on flexible substrates by facing target sputtering at room temperature

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ABSTRACT

Indium tin oxide (ITO) thin films were deposited on polyethylene terephthalate substrates using a DC facing target sputtering (DC-FTS) system at room temperature. The sputtering conditions including oxygen partial pressure and discharge current were varied from 0% to 4% and 0.5 A to 1.3 A, respectively. X-ray diffraction and scanning electron microscopy were used to study the structure and surface morphology of as-prepared films. All the films exhibited amorphous structures and smooth surfaces. The dependence of electrical and optical properties on various deposition parameters was investigated by a linear array four-point probe, Hall-effect measurements, and ultraviolet/visible spectrophotometry. A lowest sheet resistance of 17.4 Ω/square, a lowest resistivity of 3.61×10^{-4} Ω cm, and an average relative transmittance over 88% in the visible range were obtained under the optimal deposition conditions. The relationship between the Hall mobility (μ) and carrier concentration (n) was interpreted by a functional relation of $\mu \sim n^{-0.127}$, which indicated that ionized donor scattering was the dominant electron scattering mechanism. It is also confirmed that the carrier concentration in ITO films prepared by the DC-FTS system is mainly controlled by the number of activated Sn donors rather than oxygen vacancies.

1. Introduction

Indium tin oxide (ITO) thin films have been widely used in electronic devices such as solar cells, gas sensors, field effect transistors, flat panel displays, and organic light emitting diodes (OLEDs) owing to their high conductivity and high transparency in the visible range [1–7]. It is known that ITO thin films having a low resistivity of $1–4 \times 10^{-4}$ Ω cm can be easily fabricated at relatively high substrate temperatures above 200 °C [8]. However, the high fabrication temperatures limit the applications in some areas. Heterostructure solar cell manufacture demands the conservation of low substrate temperatures in order to avoid damages to the junction properties [9]. Plastic substrates such as polyethylene terephthalate (PET), polycarbonate, and polyethylene naphthalate used in flexible solar cells and flexible OLEDs cannot resist the high temperatures. Many attempts have been carried out to deposit ITO thin films on various substrates at low temperature utilizing various methods, such as different magnetron sputtering techniques [1,10–17], pulsed laser deposition [9,18,19], electron beam evaporation [8], thermal evaporation [20,21] and reactive evaporation [22].

Facing target sputtering (FTS) technology uses a different principle of plasma confinement compared with the conventional magnetron sputtering. In an FTS system, a pair of target planes is arranged to face each other in a vacuum chamber, and the magnetic fields are generated perpendicularly to the target planes in order to confine the plasma in the space between the facing target planes. The substrate is positioned outside the plasma. Consequently, less plasma damage is produced to the films and fragile substrates [23,24]. Moreover, the high energy particle bombardment to the substrate such as negative oxygen ions and secondary electrons can be effectively suppressed [24,25]. Therefore, the increment of the substrate temperature is low during the sputtering process and the corresponding film properties may be different from that obtained by conventional magnetron sputtering techniques.

In this work, ITO thin films were prepared on flexible PET substrates using a DC-FTS system at room temperature. The influence of sputtering parameters including the oxygen partial pressure and the discharge current on the structural, optical and electrical properties of the films was investigated. The relationship between the Hall mobility and carrier concentration was interpreted by a functional relation, and the dominant ionized donor scattering mechanism was suggested.

2. Experimental details

ITO thin films were deposited on commercial PET substrates at room temperature by DC-FTS system. Fig. 1 shows the schematic diagram of the sputtering system. Two rectangular ITO targets ($220 \times 50 \times 6$ mm^3 in size) which contain 90 wt.% In_2O_3 and 10 wt.% SnO_2 were placed 80 mm apart facing each other. The target–substrate distance was typically 80 mm.

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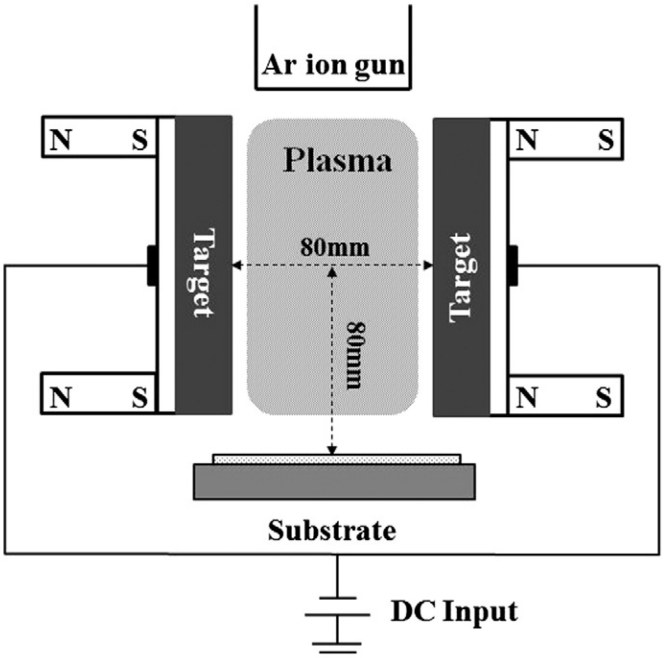

Fig. 1. The schematic diagram of the sputtering system with a confined plasma region.

Flexible PET substrates with a thickness of 100 μm were used in this work. They have a maximum working temperature of about 90 °C. The transmittance is around 85% in the visible region and the refractive index is 1.65. Prior to the deposition of the films, the PET substrates were ultrasonically cleaned in acetone, ethanol, and deionized water successively. Then they were dried with N_2 gas. Before sputtering of the ITO films on the PET substrate, the surface of the PET substrate was pretreated by Ar ion beam from a Kaufman ion source for 15 min to remove surface contaminations and improve adhesion between the ITO and PET substrate. For this study, the base pressure was 5×10^{-4} Pa evacuated by a turbo molecular pump combined with a rotary pump. The flow rates of Ar (99.99%) and O_2 (99.99%) were controlled individually by mass flow controllers. The sputtering pressure was kept constant at 0.3 Pa. The oxygen partial pressure ratio was defined as $PO_2 = P(O_2) / [P(O_2) + P(Ar)]$, where $P(O_2)$ and P(Ar) were the gas pressures. PO_2 was adjusted from 0% to 4% and the discharge current (I_d) was varied from 0.5 A to 1.3 A while the sputtering voltage was approximately 505 V. The deposition time was adjusted in order to maintain the thickness of the films at approximately 230 nm so that the measured properties were comparable. All the films were deposited at room temperature.

The film thickness was determined by spectroscopic ellipsometry using a SENpro variable angle ellipsometer. The covering energy region of the ellipsometry measurements was 1.181 eV to 3.263 eV (wavelength range: 380 nm to 1050 nm) and the angles of incidence light used were 65°, 70° and 75°. The structural properties were analyzed by using X-ray diffraction (XRD) with a Cu Kα source, and the surface morphology was imaged by scanning electron microscopy (SEM). The sheet resistance was measured by using a DS-510 four-point probe (NAGY Company). The optical transmittance spectra were measured by a HITACHI U-3010 ultraviolet/visible spectrophotometer over a wavelength range of 300–900 nm. Hall-effect measurement using the Van der Pauw geometry with a permanent magnet of 0.55 T was carried out to determine the resistivity (ρ), carrier concentration (n) and Hall mobility (μ) of the ITO thin films.

3. Results and discussion

All the ITO films obtained in this study showed good adherence to the PET substrates. No peel-off or crack on the surface was observed after the samples were bended repeatedly for 50 times. The stability is attributed to the improvement of the adhesion between the ITO and PET substrate and creation of new adsorption sites resulting from the Ar ion beam pretreatment of the PET substrate [13].

Fig. 2 shows the XRD pattern and SEM image of the ITO film prepared at oxygen partial pressure of 3% and discharge current of 1.3 A. Other samples show similar patterns and surface morphology regardless of different PO_2 and I_d. Only a sharp diffraction peak at about $2\theta = 26°$ and humps in the 2θ ranges 46° ~ 48° and 53° ~ 56° are observed which are attributed to the PET substrate. No peaks of ITO are found in the XRD patterns of all the as-deposited films. As previously reported, compared to a conventional sputtering technique, the plasma is effectively confined in the FTS system. The substrate is located outside the high-density plasma region. Therefore, the elevation of temperature is relatively low during the sputtering process [26]. The low temperature prevents the crystallization of the ITO films. No certain morphologies except a smooth surface can be observed from the SEM image in Fig. 2. This result is in a good agreement with the analysis of XRD characteristics that all the ITO thin films are of an amorphous nature.

An overview of the deposition time (t_d), sheet resistance (R_s), average relative transmittance (T_{avg}), figure of merit (Φ_{TC}), and other parameters of the samples is shown in Table 1. Deposition time (t_d) was adjusted to limit the thickness of all the samples to approximately 230 nm. It is found that the deposition rate ranges from 31.6 nm/min to 91.8 nm/min as I_d increases from 0.5 A (sputtering power = 252.5 W) to 1.3 A (sputtering power = 656.5 W). However, the influence of PO_2 on the deposition rate is negligible. The figure of merit Φ_{TC} defined by Haacke [27] was calculated to evaluate the performance of the films as shown in Eq. (1).

$$\Phi_{TC} = \frac{T_{avg}^{10}}{R_s}. \tag{1}$$

Where T_{avg} is the average relative transmittance in the visible range and R_s is the sheet resistance. In this work, we took the values of uncoated PET transmittance as 100% to calculate the relative transmittance for ITO films. As displayed in Table 1, the highest value of the Φ_{TC} is $17.3 \times 10^{-3}\ \Omega^{-1}$ for sample 9 which exhibits the lowest R_s of 17.4 Ω/Sq. (Ohm/Square) and a transmittance of 88.7%. It is observed that R_s decreases at the beginning and increases after it reaches a minimum value of 21.3 Ω/Sq. with PO_2 increasing from 0% to 4%. The optimum PO_2 is 3% which yields the highest value of the Φ_{TC} where I_d is kept at 0.7 A. A continuous decrease in R_s with I_d increasing from 0.5 A

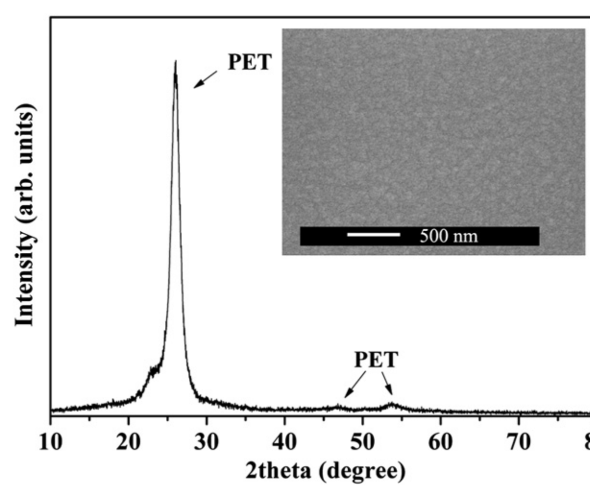

Fig. 2. XRD pattern and SEM image of the ITO film. PO_2 and I_d were fixed at 3% and 1.3 A, respectively.