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Radio frequency magnetron sputter-deposited indium tin oxide for use as a cathode in transparent organic light-emitting diode

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Abstract

Indium tin oxide (ITO) films were prepared by radio frequency magnetron sputtering at room temperature, for use as a cathode in a transparent organic light-emitting diode (TOLED). To minimize damage to the TOLED by the ITO sputtering process, the target-to-substrate distance was increased to 20 cm. An ITO film deposited at the optimum oxygen partial pressure exhibited an electrical resistivity as low as 4.06×10^{-4} Ω cm and a high optical transmittance of 91% in the visible range. The film was used as a transparent cathode for a TOLED with structure of an ITO coated glass substrate/Naphthylphenyldiamide (60 nm)/Tris–(8-hydroxyquinoline) aluminum (60 nm)/LiF (1 nm)/Al (2 nm)/Ag (8 nm)/ITO cathode (100 nm). A maximum luminance of 37,000 cd/m^2 was obtained. The device performance was comparable to a conventional OLED.

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1. Introduction

Indium tin oxide (ITO) thin films are in widespread use as transparent electrodes in optoelectronic and electro-optic devices such as solar cells and flat panel displays. This is largely due to their unique properties of both a high transmittance in the visible region and a low electrical resistivity. The majority of the methods used in their preparation involve a relatively high substrate temperature (>200 °C) because a reasonably low electrical resistivity of ITO films can be obtained by raising the substrate temperature during the deposition. Certain types of flat panel devices require that the ITO films are processed at a low temperature. Low-temperature (<100 °C) ITO films have been prepared by a variety of methods such as sputtering [1–11], evaporation [12], and pulsed laser deposition [13]. Among these, sputtering techniques including, direct current magnetron sputtering [1–3], radio frequency (RF) magnetron sputtering [4–9], facing target sputtering [10] and kinetic energy controlled sputter deposition [11] have been extensively reported because the methods are capable of being scaled-up and the processing variables are easily controlled [14]. Ishibashi et al. reported that sputtering with a low discharging voltage was very effective in reducing the resistivity of ITO films at low temperature because the lower sputtering voltage decreases the damage to an ITO film [1]. Hoshi et al. reported on an ITO film with a low electrical resistivity of 3.5×10^{-4} Ω cm at a sputtering voltage below 100 V [11]. It is well known that negative ions created in the sputtering process are accelerated into the substrate by the sheath electric field and the ions, which are mainly O^-, and bombard the ITO film, thus damaging it [15]. Therefore, in order to obtain a high quality ITO film at a low temperature, high energetic particles must be eliminated before they reach substrate.

When depositing an ITO film as a transparent cathode for a transparent organic light-emitting diode (TOLED) using

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sputtering, the performance of the device can be negatively affected by energetic particles created in the sputtering process [16,17]. An RF magnetron sputtering system has the potential for solving these problems because the method has the advantage of a low discharging voltage, that is, low energetic sputtered particles are produced, compared to conventional sputtering method. We recently reported the improvement in the performance of TOLED with increasing sputtering power in the deposition of an ITO cathode using an RF magnetron sputtering system [18].

In this study, we described the preparation of an ITO film by an RF magnetron sputtering method at room temperature for use as a cathode in a TOLED. In order to minimize the number of sputtered energetic particles arriving at a substrate during ITO sputtering, the target-to-substrate distance was increased to 20 cm. We also used this film as a transparent cathode for a TOLED having performance comparable to a conventional OLED.

2. Experimental details

ITO thin films (~200 nm) were prepared on glass by RF magnetron sputtering. When the base pressure reached 2.6×10^{-5} Pa, a 6 in. diameter ceramic plate of 90 wt.% In_2O_3 and 10 wt.% SnO_2 was sputtered in an $Ar–O_2$ mixture plasma at room temperature.

First, we prepared ITO films as a function of target-to-substrate distances at a fixed working pressure of 0.66 Pa to examine the dependence of structural properties of the films on the target-to-substrate distances. Then at a fixed target-to-substrate distance of 20 cm and a working pressure of 0.66 Pa, we varied the partial pressure of the oxygen (P_{O_2}) from 2.0×10^{-3} to 1.3×10^{-2} Pa to examine the dependence of optical and electrical of the films on P_{O_2}.

The film thickness (t) was measured by observing cross-section of the film using scanning electron microscopy operating at 10 kV. The electrical resistivity (ρ) of the film was determined using the simple relation $\rho = R_s \cdot t$, where R_s is the sheet resistance measured by a four point probe. Hall mobility and carrier density were measured using the Van der Pauw's method at room temperature with a magnetic strength of 0.513 T. Ultra violet-visible-near infrared spectrometer was employed to measure the optical transmittance of the films. The crystal structure of the films was analyzed by X-ray diffraction using Cu Kα radiation.

We also fabricated a TOLED using an RF magnetron sputtered ITO thin film as a transparent cathode. Naphthylphenyldiamide (NBP), Tris-(8-hydroxyquinoline) aluminum (Alq_3), LiF, Al and Ag were sequentially deposited by a thermal evaporation on ITO-coated glass as the substrate. The individual layer thickness was 60, 60, 1, 2, and 8 nm for NPB, Alq_3, LiF, Al and Ag, respectively. A quartz crystal thickness monitor was used for the quantitative assurance of reproducible thickness. ITO film with a thickness of 100 nm was sputtered on the thin Ag layer. In the ITO deposition, the working pressure and the P_{O_2} was 0.66 and 5.3×10^{-3} Pa, respectively. Current–voltage–luminance was measured using a Keithley 238 source and a luminance meter (Minolta LS-100).

3. Results and discussions

Fig. 1 shows X-ray diffraction patterns of the ITO films as a function of target-to-substrate distances at a working pressure of 0.66 Pa. Studies on ITO films show that the different energy state of sputtered particles brings about different texture formation. High energetic sputtered particles prefer the (400) and (440) orientation, while relatively low energetic particles prefer the (222) orientation [19]. The energy of sputtered particles is dissipated through collisions with the sputtering gas such as Ar [20]. By examining the structures of an ITO film as a function of a target-to-substrate distance, one might indirectly classify the energy dissipation degree of the sputtered atoms. The films deposited at target-to-substrate distances smaller than 6 cm showed (222), (400) and (440) peaks. The appearance of the (400) and (440) peaks indicates that fairly high energetic particles were able to reach substrate. The (400) and (440) peaks disappeared with further increase in a target-to-substrate distance. The ITO films deposited at target-to-substrate distances of 20 cm were almost amorphous. This observation suggests that target-to-substrate distances of 20 cm is enough to fully dissipate the particle energy.

A strong correlation was observed between the P_{O_2} and the optical and electrical properties of the ITO films. Fig. 2 shows the optical transmittance of the ITO films deposited at a target-to-substrate distance of 20 cm and a working pressure of 0.66 Pa in the range from 400 to 700 nm. The optical transmittance increase rapidly with increasing

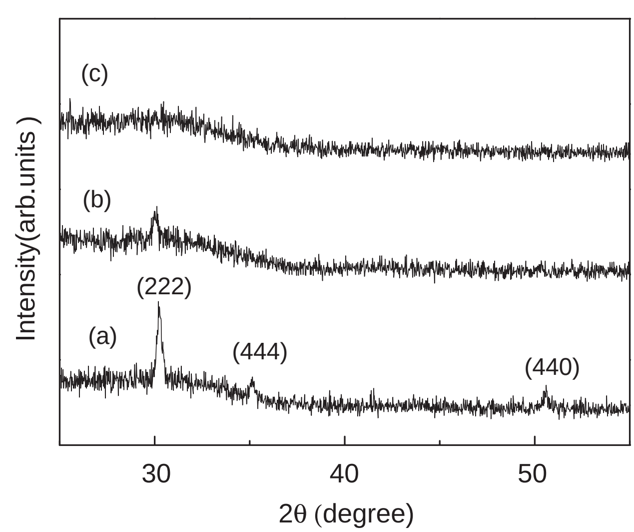

Fig. 1. X-ray diffraction patterns of ITO films on glass deposited at a different target-to-substrate distances of (a) 6 cm (b) 15 cm, and (c) 20 cm at a working pressure of 0.66 Pa.