

High work function of Al-doped zinc-oxide thin films as transparent conductive anodes in organic light-emitting devices

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Abstract

Deposition of Al-doped ZnO (AZO) films with various film thicknesses on glass substrates was performed to investigate the feasibility of using AZO films as anode electrodes in organic light-emitting devices (OLEDs). The electrical resistivity of the AZO films with a 180-nm thickness was $4.085 \times 10^{-2} \Omega \text{ cm}$, and the average optical transmittance in the visible range was 80.2%. The surface work function for the AZO films, determined from the secondary electron emission coefficients obtained with a focused ion beam, was as high as 4.62 eV. These results indicate that AZO films grown on glass substrates hold promise for potential applications as anode electrodes in high-efficiency OLEDs.

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1. Introduction

Potential applications of organic light-emitting devices (OLEDs) have driven extensive efforts to fabricate various kinds of OLEDs with high brightness and high efficiency [1–5]. Various device structures, consisting of active layers and electrodes, have been designed to improve the efficiency of the OLEDs [6,7]. Among the several parts of OLEDs, transparent conducting oxide films have become particularly attractive due to their being promising candidates for anodes [8]. The materials of the device anodes for hole injection are particularly important for enhancing device efficiency. tin-doped indium oxides (ITOs) acting as anodes in OLEDs have been extensively used as anodes because of their high conductivity and transparency over the visible range and their high work

function. However, since the indium in the ITO thin films might diffuse into the organic layers, resulting in the degradation of the OLED efficiencies [9–11], the fabrications of the alternative anodes with thermal stabilities are necessary for improving the efficiency of OLEDs.

Among the alternative candidate thin films, Al-doped ZnO (AZO) thin films have been considered as suitable anodes because ZnO thin films are more stable in reducing ambient, more abundant, and less expensive in comparison with the ITO films which make them appropriate for potential use as anodes in OLEDs [12–16]. Since ZnO thin films are large bandgap oxide semiconductors with a large excitonic binding energy and a high chemical stabilization [17,18], their noble physical properties have stimulated applications in many promising optoelectronic devices, such as flat-panel displays. Since the utilization possibility of AZO thin films grown on glass substrates as anodes in OLEDs is strongly affected by the electrical, the optical, and the electronic properties, systematic studies concerning those properties are very important for

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improving the efficiencies of OLEDs. Even though some works on the electrical, the optical, and the electronic properties of the AZO thin films have been reported [13,19,20], systematic studies concerning the work functions with high magnitudes of the AZO thin films are still necessary for enhancement of the hole-injection efficiency in OLEDs.

The paper reports on the electrical, the optical, and the electronic properties of AZO thin films with various film thicknesses acting as anodes in OLEDs. The AZO thin films were grown on glass substrates by using a reactive radio-frequency sputtering system. The electrical, the optical, and the electronic properties of films were measured to investigate AZO thin films as promising candidates for use as anodes in OLEDs.

2. Experimental details

Atomic force microscopy (AFM) measurements were carried out in order to characterize the surface smoothness of the AZO layer, and Van der Pauw Hall effect measurements were performed in order to investigate the electronic parameters [21]. Transmittance measurements were performed in order to investigate the optical properties of the AZO layer, and the secondary electron emission coefficient (γ)-focused ion beam (FIB) measurements were performed to determine the γ values and the work functions of the AZO thin films.

The AZO films were grown on glass substrates at room temperature by using a reactive radio-frequency magnetron sputtering system. A target consisting of ZnO containing 5 wt.% Al was used. The thicknesses of the AZO thin films were 180, 220, and 600 nm, respectively. The film resistivity was determined from the sheet resistance measured by using the four-point probe technique. The Hall mobility and the carrier density measurements were made using the Van der Pauw method at 300 K. The optical transmittance measurements were made using a LAMDA 19 spectrophotometer (300–700 nm). The γ -FIB measurements were performed by using a home made equipment for determining the secondary electron emission coefficients (γ) and work functions of the AZO thin films [22].

3. Results and discussion

The as-grown AZO thin films had mirror-like surfaces without any indication of pinholes, which was confirmed by using Normarski optical microscopy and scanning electron microscopy. AFM images of AZO thin films with thicknesses of 180, 220, and 600 nm grown on glass substrates are shown in Fig. 1(a–c), respectively. The root mean squares of the surface roughnesses of the AZO thin films with thicknesses of 180, 220, and 600 nm, as determined from the AFM measurements, were 4.0, 2.8, and 10.5 nm, respectively. The images in Fig. 1 show that the surfaces of all the AZO thin films grown on glass substrates are smooth. Therefore, when the organic layer is grown on the AZO thin films with thicknesses of 180 and 220 nm, the AZO anode/organic heterostructure has an excellent heterointerface, resulting in high-efficiency OLEDs.

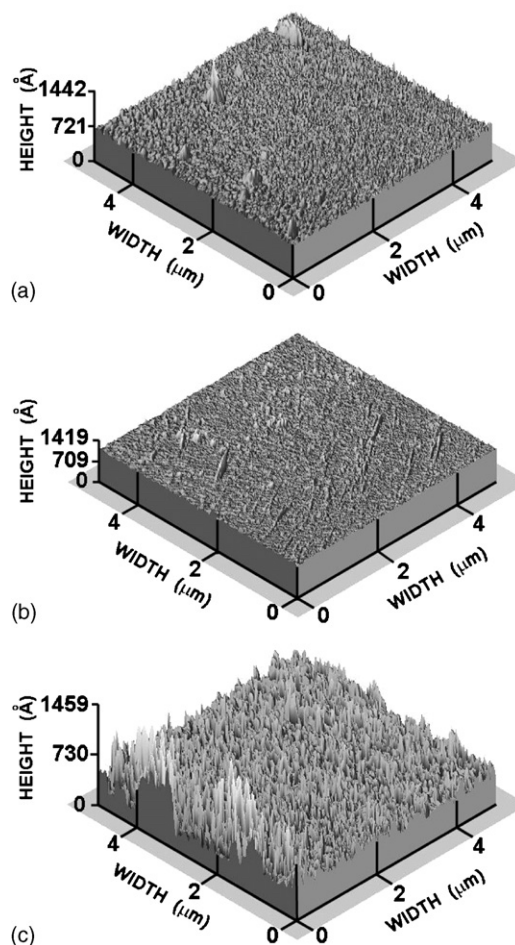


Fig. 1. Atomic force microscopy images of AZO films containing 5 wt.% of Al with film thicknesses of (a) 180, (b) 220, and (c) 600 nm.

The resistivities, the Hall mobilities, the carrier types, and the carrier concentrations for AZO films with thicknesses of 180, 220, and 600 nm, as determined from the Hall effect measurements at room temperature, are summarized in Table 1. The carriers for all of the AZO thin film were n-type. While the resistivity of the AZO thin films slightly decreased with increasing film thickness, their mobility significantly increased with increasing thickness. A relative higher resistivity might be caused by an increased incorporation of suboxides of Al–O, either in Al_2O_3 clusters or some intermediate materials [20]. The increase in the mobility of the AZO thin films with increasing thickness might be attributed to an increase of the grain size along the growth direction for thicker films [20]. Also, the significant higher resistivity values of the deposited

Table 1
Resistivities, Hall mobilities, and carrier concentrations of AZO films with thicknesses of 180, 220, and 600 nm, determined from Hall effect measurements at 300 K

Film thickness	180 nm	220 nm	600 nm
Resistivity ($\Omega \text{ cm}$)	4.085×10^{-2}	2.611×10^{-2}	1.215×10^{-2}
Hall mobility (cm^2/Vs)	6.507×10^{-1}	1.637	1.605×10^2
Carrier concentration (cm^{-3})	2.348×10^{20}	1.460×10^{19}	3.201×10^{18}

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