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Characteristics of indium zinc oxide/silver/indium zinc oxide multilayer thin films prepared by magnetron sputtering as flexible transparent film heaters

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ABSTRACT

Indium zinc oxide/silver/indium zinc oxide multilayer structures with a very low resistivity and high transmittance were deposited on a polyethylene terephthalate substrate by DC magnetron sputtering. The electrical, optical, mechanical, and thermal properties of the multilayer films were investigated at different Ag layer thicknesses (10–20 nm). The multilayers demonstrated relatively constant resistance change ($\Delta R/R_0$) over repeated bending tests with a radius of 8 mm. Efficient Joule heating could increase the operating temperature of the film heaters (7.3 Ω/□) to 73.7 °C in ~20 s at 3 V; moreover, the heat distribution remained uniform after bending tests. A high transmittance (84.7%) at a wavelength of 550 nm and low sheet resistance (7.3 Ω/□) of the optimized multilayer was obtained at an Ag layer thickness of 15 nm. These results indicate that the multilayers are promising as transparent film heaters for next-generation flexible applications.

1. Introduction

Transparent conductive oxide (TCO) films showing the particular characteristics of good conductivity and high transparency have attracted considerable research interest because of their potential technological applications, in particular, as transparent electrodes in optoelectronic devices, organic light emitting diodes, flat-panel displays, low emitting windows, and thin-film heaters (TFHs) [1–5].

Indium zinc oxide (IZO) films, in particular, have been attracting significant attention because of their good conductivity, high optical transparency, and low deposition temperature similar to indium tin oxide (ITO) films. In addition, crystalline ITO films are more brittle than amorphous IZO films due to their crystal structure, which renders them unsuitable for flexible applications. Recently, some researchers have proposed transparent conducting oxide TCO/Ag/TCO multilayer structures with much lower resistances as transparent conducting electrodes. The advantages of TCO/Ag/TCO multilayer structures, such as low sheet resistance, high optical transparency in the visible range, and relatively lower thickness in comparison to single-layered oxide films, favor their potential application with improved performance [6–9].

TFH is one application of transparent conductive films. Their controllable and stable Joule heating effect at low voltage renders such films suitable for safe and beneficial heater applications. The performance of TFHs, in terms of operating temperature and response time, depends on the characteristics of the thin films used. The flexibility of conventional TFHs is not sufficiently high, limiting potential applications. Although good electrical and optical properties of oxide/metal/oxide (OMO) multilayer electrodes including ITO/Ag/ITO, AZO/Au/AZO, IZO/Ag/IZO multilayers have been previously reported, [10–16] investigations of the reflectance characteristics in the near-infrared (NIR) region, heat distributions, and operating temperature performance after bending tests are lacking. Current research on TFHs is mainly focused on outdoor display panels and on windows for vehicles, airplanes, and buildings [17,18]. Research on properties after repeated bending tests is an essential part of the development of useful flexible TFHs.

In this work, we propose a scalable strategy to fabricate a flexible TFH consisting of an IZO/Ag/IZO multilayer grown on a polyethylene terephthalate (PET) substrate by magnetron sputtering. In particular, we focus on the study of the properties of flexible TFHs. Ag metal films of low resistivity were chosen in order to improve the electrical properties. Additionally, amorphous IZO layers were formed on both sides of the Ag film. We have demonstrated the possibility of using IZO/Ag/IZO multilayers as a replacement for conventional ITO thin films in windows for vehicles, aircraft, and building applications.

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2. Experimental procedure

IZO/Ag/IZO thin films were continuously sputter-deposited on polyethylene terephthalate (PET) substrates, using an IZO target (ZnO, 10 wt%) and an Ag metal target (purity 99.99%) at room temperature (RT, 300 K). Both targets to substrate distance were 100 mm. The 40 nm thick bottom IZO layer was deposited on the PET substrate with DC magnetron sputtering method using a single sintered target (3 in.). The base pressure was exhausted to 1.33×10^{-3} Pa and working pressure was maintained to 1 Pa during sputtering process. The DC power was fixed at 100 W and Ar gas flow rate of 20 sccm. After deposition the IZO layer, the Ag interlayer was continuously sputtered onto the bottom IZO layer as a function of Ag thickness at same sputtering conditions. The thickness of the Ag layer was controlled by deposition time, where Ag thickness is confirmed to 10, 15, and 20 nm for each deposition time 15, 22.5, and 30 s, respectively. After deposition of the Ag interlayer, a 40 nm-thick top IZO layer was also sputtered on the Ag interlayer. The thickness of the IZO layers was fixed at 40 nm, and for the Ag films, thicknesses of 10, 15, and 20 nm. Prior to deposition, each target was pre-sputtered for 10 min to remove impurities and to maintain the stability of the plasma.

The electrical and optical properties of the samples were measured using a Hall-effect measurement system (ECOPIA, HMS3000) and a UV–Vis-NIR spectrometer (PerkinElmer, Lambda 750), respectively. Cross-sectional views were obtained using transmission electron microscopy (TEM). An infrared camera (IR camera, Nikon) was used to measure the surface temperatures of the transparent film heaters and their heat ray reflectance. A DC voltage of 3 V was applied for heat generation using a power supply (Keithley2400, USA). To evaluate the heat reflectance of the multilayers, we used a custom-made instrument. This instrument was used to monitor the temperature changes of the sample, which was kept in a heat insulating box as shown in Fig. 1. The light source used in this instrument was a Philips BR125, emitting light in the wavelength range 900–2000 nm under a source power of 250 W. The heat insulating box was made up of Styrofoam in order to prevent heat loss. The sample was placed at the center of the box and was at a distance of 20 cm from the NIR source. In order to concentrate the NIR light, we made a 150 mm long, 50 mm × 50 mm rectangular channel using Al plates because of the emissivity property of Al. The base temperature of the box (without heating) was 27 °C. The temperature change of the box was monitored by an IR camera for 10 min at intervals of 1 min.

3. Results and discussion

We used the Essential Macleod optical simulating program in order to design the NIR reflective coating. The thickness of each layer was determined to obtain the desired optical transmittance and reflectance for the multilayer films. Durrani et al. [19] suggest using the same thickness for the first and third layers in OMO structures. In addition, OMO structure multilayers with 40 nm-thick TCO have the best optical performance which also has be proved by our before research [2]. Multilayers also show higher transmittance compared to a single layer. Therefore, the thickness of the oxide layer was fixed at 40 nm, while that of the Ag layer was varied to obtain the best optical properties.

The simulated optical properties of the multilayers with 40 nm-thick IZO layers and at varying thicknesses of the Ag layer are shown in Fig. 2(a). All samples demonstrated a high transmittance and low reflectance in the visible range. Furthermore, the IZO single layer also has low reflectance in the NIR region. However, the reflectance of multilayers increased significantly with an increase in the Ag layer thickness in the NIR range. The reflectance of IZO/Ag/IZO multilayers was found to increase from 72.5 to 91.6% as the Ag layer thickness increased from 10 to 20 nm at a wavelength of 1500 nm. Therefore, on the basis of the obtained transmittance and reflectance values, multilayers can be expected to be suitable for NIR reflective coatings.

Fig. 2(b) shows the measured transmittance and reflectance of the IZO single layer and IZO multilayers at different Ag layer thicknesses. The optical transmittance at a wavelength of 550 nm was 75.4%, 83.7%, 84.7%, and 78.2%, respectively, for the IZO single layer and IZO multilayers(with 10, 15, and 20 nm-thick Ag layer). It is noteworthy that the IZO/Ag/IZO multilayers showed a clearly higher transmittance than the single layer, reaching a peak of 84.7%, when the thickness of Ag was 15 nm, as a consequence of index-matching of the OMO structure [20]. The optical transmittance of the multilayers decreased and the reflectance increased with increasing Ag thickness in the NIR region, which was due to the increase in reflectance caused by surface plasmon resonance phenomena [21].

Fig. 3(a) shows the resistivity (ρ), carrier density (n), and Hall mobility (μ) of a series of IZO multilayers with different Ag layer thicknesses. The resistivity of the multilayers was lower than that of the IZO single layer and continuously decreased with increasing Ag layer thickness because of the increase of n donated mainly by the Ag layer. However, although the Hall mobility decreased due to ionized-impurity scattering, it is obvious that the effect of increasing n was larger than that of decreasing μ. Further, as we increased the thickness of the Ag interlayer from 10 to 20 nm in the multilayers, both n and μ were found to increase slightly. This behavior was attributed to the increased n. Fig. 3(b) presents the sheet resistance (R_s) as a function of the IZO layer. The R_s of the IZO single layer was 102 Ω/□, and 13.7 Ω/□, 7.3 Ω/□, and 4.5 Ω/□ for multilayers with 10, 15, 20 nm-thick Ag layers, respectively. The R_s of the multilayer sharply decreased compared to the single layer and further decreased as the Ag thickness increased from 10 to 20 nm, due to the low resistivity of the multilayers.

The temperature variations inside the box are shown in Fig. 4. Images on the right side of the figure show the temperature inside the

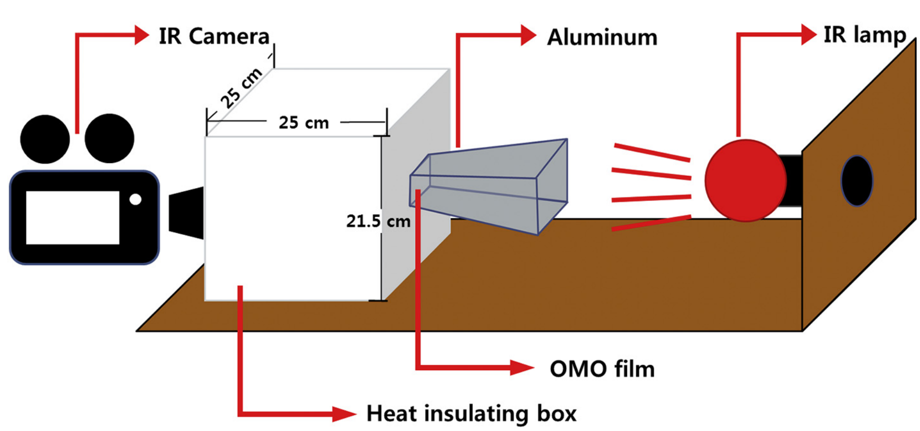

Fig. 1. Schematic of heat-reflecting measurement system.