Characteristics of ITO/Ag/ITO Hybrid Layers Prepared by Magnetron Sputtering for Transparent Film Heaters

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Transparent film heaters (TFHs) based on Joule heating are currently an active research area. However, TFHs based on an indium tin oxide (ITO) monolayer have a number of problems. For example, heating is concentrated in only part of the device. Also, heating efficiency is low because it has high sheet resistance ($R_s$). To address these problems, this study introduced hybrid layers of ITO/Ag/ITO deposited by magnetron sputtering, and the electrical, optical, and thermal properties were estimated for various thicknesses of the metal interlayer. The $R_s$ of ITO(40)/Ag/ITO(40 nm) hybrid TFHs were 5.33, 3.29 and 2.15 Ω/□ for Ag thicknesses of 10, 15, and 20 nm, respectively, while the $R_s$ of an ITO monolayer (95 nm) was 59.58 Ω/□. The maximum temperatures of these hybrid TFHs were 92, 131, and 145°C, respectively, under a voltage of 3 V. And that of the ITO monolayer was only 32°C. For the same total thickness of 95 nm, the heat generation rate (HGR) of the hybrid produced a temperature approximately 100°C higher than the ITO monolayer. It was confirmed that the film with the lowest $R_s$ of the samples had the highest HGR for the same applied voltage. Overall, hybrid layers of ITO/Ag/ITO showed excellent performance for HGR, uniformity of heat distribution, and thermal response time.

Keywords : Indium tin oxide, Transparent film heater, Hybrid layer, Transparent conductive oxide, Infrared camera

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I. INTRODUCTION

With the recently increasing level of interest in transparent electronic devices, there has been much research on transparent conductive oxides (TCOs). Among these, SnO₂-doped In₂O₃ (ITO), a degenerate semiconductor with a wide band gap, has high electrical conductivity, qualifying it for numerous applications such as smart windows, solar cells, displays, and organic light emitting diodes (OLEDs) [1-4]. A particular focus of research has been on TFHs, which are based on Joule heating [5-9]. The HGR per area, ($q$), is given by Eq. (1) [10]

\[ q = \frac{V^2}{R_s} \] (1)

where $V$ is the voltage applied between the electrodes, and $R_s$ is the sheet resistance of the samples; TFHs generate more heat for lower values of $R_s$ [10]. There are numerous applications, such as outdoor display panels, periscopes, and windows for vehicles, airplanes and buildings [6, 7]. However, in the case of a TFH based on an ITO monolayer, heating can concentrate in a part of the device because of low electrical conductivity. To solve this problem, ITO films have to either be deposited above the crystallization temperature ($T_c$ about 170°C) or with a post-annealing process.

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II. METHODS

In this study, ITO/Ag/ITO thin films were continuously sputter-deposited on non-alkali glass substrates (Corning E-2000), using an ITO ceramic sintered target (SnO$_2$, 10 wt. %) and an Ag metal target (purity 99.99 %) at room temperature (RT). Deposition of hybrid multilayers was carried out with a DC- and RF-magnetron sputtering system equipped with Ag and ITO targets, respectively. The thickness of the ITO layers was fixed at 40 nm and for the Ag films, 10, 15, and 20 nm were used. Sputtering was performed under 20 sccm flowing high-purity Ar gas (purity 99.999%) at a working pressure of 1.0 Pa. Prior to deposition, each target was pre-sputtered for 10 min to remove impurities and to maintain stability of the plasma. The electrical and optical properties of films were measured using a Hall-effect measurement system (ECOPIA, HMS3000), and a UV-Vis spectrophotometer (UV-1800, SHIMADZU) in the range 200–1100 nm, respectively. Microstructure of the films was determined by X-ray diffractometer (XRD, CuK$_\alpha$ radiation at 40 kV – 40 mA, 2$\theta$ mode, GADDS, BRUKER). In addition, the thermal performance of the TFHs was evaluated using a surface-temperature infrared camera (IR camera, Nikon). DC voltage was applied from 0.5–3 V at constant current 1 A using a power supply (Keithly2400, USA), through two copper tapes that made contact at the edges as shown schematically in Fig. 1. We also tried to estimate the defrosting ability of the TFHs. The samples, before the defrosting test, were kept in the refrigerator for 1h to form frost on the sample surface.

III. RESULTS AND DISCUSSION

Figure 2 shows (a) $R_s$ and resistivity ($\rho$), carrier density ($n$), and Hall mobility ($\mu$) of the ITO monolayer and the hybrid layers with different Ag thicknesses. In Fig. 2 (a), it is seen that the $R_s$ of the hybrid layers was remarkably reduced in comparison to the ITO monolayer: for the ITO monolayer the $R_s$ was 59.58 $\Omega/\Box$; for the ITO/Ag/ITO hybrid layers, 5.33, 3.29, and 2.15 $\Omega/\Box$ for Ag thicknesses of 10, 15, and 20 nm, respectively. The $R_s$ of the hybrid layers further decreased with increasing Ag layer thickness. The resistivity of the hybrid layers, as seen in Fig. 2 (b), was also clearly reduced: $5.11 \times 10^{-5}$, $3.26 \times 10^{-5}$, and $2.23 \times 10^{-5} \Omega \cdot cm$ for Ag 10, 15, and 20 nm, respectively, compared to $5.58 \times 10^{-4} \Omega \cdot cm$ for the ITO monolayer, as a result of increased carrier density. However, with increased carrier density, the Hall mobility decreased, as a result of ionized-impurity scattering. And the resistivity is related to these properties through Eq. (2) [14]:

$$\rho = \frac{e \cdot n \cdot \mu}{q}$$
\[
\rho = \frac{1}{\sigma} = \frac{1}{ne\mu}
\]  

(2)

where: \(\rho\) denotes resistivity; \(\sigma\), electrical conductivity; \(n\), carrier density; \(e\), electronic charge; and \(\mu\), Hall mobility.

The \(\rho\) of the hybrid layers remarkably improved, compared to the ITO monolayer, because the increase of \(n\) was larger than the decrease of \(\mu\).

With an increase in thickness of metal in the hybrid layers, \(n\) and \(\mu\) increased. This could result from an increase in free electrons per unit volume. On the other hand, the \(\mu\) of hybrid layers increased from 17.4 to 23.3 cm²/V·s as the Ag interlayer thickness increased from 10 to 20 nm. This behavior is attributed to the transition from a formation of islands to a continuous film. The grain size in the Ag layer increased with increasing thickness (see Fig. 3), which could have led to a decrease in grain-boundary scattering [15]. As a result, it was shown that the \(\sigma\) of the multilayer was affected by the Ag interlayer [16]. It has been reported that an ITO monolayer requires an additional step of post-annealing or other high temperature process to decrease \(R\) [11]. However, hybrid layers deposited at RT exhibit superior conductivity without an extra process.

Figure 3 shows XRD patterns of the ITO monolayer and hybrid layers deposited at RT; analysis was carried out in the 2θ mode over 24–48°. The ITO film shows a (222) peak near 2θ =30° and a (400) diffraction peak near 2θ =35° [17]. The ITO monolayer (Fig. 3(a)) reveals a halo-pattern indicating an amorphous structure. With the hybrid layers, the ITO (222) peak and the Ag (111) peak are clearly observed. It is thought that the upper ITO layer may have resulted from epitaxial growth caused by the (111) peak of the Ag interlayer [18]. On the other hand, the peak intensity of Ag (111) monotonically increased with Ag thickness. Therefore, this increased Ag layer crystallinity could have led to the improvement of the hybrid layer’s electrical properties seen in Fig. 2.

Figure 4 shows the optical transmittance of the ITO monolayer and the hybrid layers with different Ag layer thicknesses, for wavelengths 200–1100 nm and with air as reference. The optical transmittance at 550 nm for the ITO monolayer (95 nm thickness) is 81.6%, and for the ITO(40)/Ag/ITO(40 nm) hybrid layers with Ag thicknesses 10, 15 and 20 nm, are 86.9, 81.7, and 66.5%, respectively. The ITO(40)/Ag(15)/ITO(40 nm) hybrid layer shows nearly the same transmittance as the 95 nm ITO monolayer. Notably, the ITO(40)/Ag(10)/ITO(40 nm) layer shows a clearly higher transmittance (86.9%) than the ITO monolayer (81.6%), as a result of index-matching of the OMO structure [19, 20]. These data confirm that the hybrid layer is a key factor determining both the electrical and optical properties. The optical transmittance of hybrid layers decreased with increasing Ag thickness in the Near Infrared (NIR) region, which is due to the increase in reflectance caused by surface plasmon resonance (SPR) phenomena. By Drude theory [21], plasma frequency (\(\omega_p\)) depend on free electron density, that is, it shifts to short wavelength with increasing free electron density. As shown in Fig. 2, the \(n\) of ITO/Ag/ITO layer increased with increasing Ag layer thickness. Therefore, decrease of transmittance for ITO/Ag/ITO layer could be

![FIG. 3. XRD patterns of ITO monolayer and hybrid layer deposited with various thicknesses of Ag layer. (a) ITO monolayer (95 nm), (b) ITO(40)/Ag(10)/ITO(40 nm), (c) ITO(40)/Ag(15)/ITO(40 nm), (b) ITO(40)/Ag(20)/ITO(40 nm).](image1)

![FIG. 4. Optical transmittance of the ITO monolayer and the hybrid layers as a function of the Ag layer thickness.](image2)
attributed to increase in $n$ [22].

Figure 5(a) shows the temperature reached by the TFHs as a result of ohmic heating, as a function of heating and cooling time after 3 V was continuously applied and then removed. Voltage was applied for 100 sec, and then removed between 100 and 300 sec. For the ITO monolayer, the temperature increased to 31.3°C within 30 sec and by then was effectively equilibrated, with a maximum temperature ($T_{\text{max}}$) of 32°C arrived at by 100 sec. For the hybrid layers, the temperature reached 86°C (Ag, 10 nm), 122°C (Ag, 15 nm), and 135.5°C (Ag, 20 nm) after 30 sec, clearly indicating higher HGR than that of the ITO monolayer. Furthermore, the $T_{\text{max}}$ of the hybrid layers were 92°C (Ag, 10 nm), 131°C (Ag, 15 nm) and 145°C (Ag, 20 nm) after 100 sec. Especially, the hybrid layers (Ag, 15 and 20 nm) show $T_{\text{max}}$ almost 100°C higher than the ITO monolayer. This result follows from the much lower $R_s$ of the hybrid layers [10].

Figure 5(b) shows a comparison of the average and maximum temperatures $T_{\text{ave}}$ and $T_{\text{max}}$ during the heating time (100 sec), in order to evaluate uniformity of heat distribution. For the hybrid layers, the HGR for 3 V applied requires approximately 30 sec to reach the equilibrium temperature, and natural cooling to RT then occurs in approximately 50 sec.

It is confirmed that the hybrid layers consisting of ITO/Ag/ITO showed higher heating rates compared to previous studies about TFHs [22, 23].

Figure 6 shows photographs indicating transmittance for an ITO (95 nm) monolayer and an ITO(40)/Ag(15)/ITO(40 nm) hybrid layer of the same overall thickness. Those transmittances at 550 nm were 81.6% and 81.7% for the ITO monolayer and hybrid layer, respectively, as shown in Fig. 4. These results confirm that both the ITO monolayer and the ITO(40)/Ag(15)/ITO(40 nm) hybrid layer are suitable for TFHs, because the transmittances are virtually identical in the visible range.

Figure 7 shows the spatial heat distribution measured by

FIG. 5. (a) Change in the temperature reached by the TFHs as a function of heating time for 3 V applied; (b) summarized $T_{\text{ave}}$ and $T_{\text{max}}$ for the samples.

FIG. 6. Photographs for comparison of transparencies, (a) ITO (95 nm) monolayer, (b) ITO(40)/Ag(15)/ITO(40 nm) hybrid layer.

FIG. 7. Contours of the temperature distributions measured by IR camera for various applied voltages after 60 sec. Samples were (a) ITO monolayer (95 nm), and ITO(40)/Ag/ITO(40 nm) hybrid layer structures for Ag thicknesses (b) 10 nm, (c) 15 nm, and (d) 20 nm, respectively.
an infrared surface temperature camera for various voltages applied to the samples for 60 sec. For the ITO monolayer (a), the temperature reached was 32°C for 3 V applied and increased only to 97°C when the voltage was increased to 12 V, indicating a large heating variation across the sample. However, the hybrid layers showed both excellent HGR (temperatures of 92°C and above for only 3 V applied) and uniformity of heat distribution, which likely result from high electrical and thermal conductivity of the hybrid structure. The highest HGR and uniformity of heat distribution were obtained for the sample of ITO(40)/Ag(20)/ITO(40 nm). However, this sample showed a lower optical transmittance in the visible region (see Fig. 4). These data confirm that a metal thickness of 15 nm is most appropriate for TFHs applications. Figure 8 shows the defrosting efficiency as a function of time under the same applied power (voltage: 3 V, current: 1A) for (a) an ITO monolayer (95 nm) and (b) ITO(40)/Ag(15)/ITO(40 nm) hybrid layers. With the ITO monolayer TFH, defrosting appeared at 5 sec, but the moisture was not completely removed within 30 sec. However, a TFH with hybrid layer structure was defrosted completely within only 5 sec. Yoon et. al reported that a CNT TFH took 1 min for defrosting with a power of 0.12 W (12 V) [8]. The hybrid layer TFHs in the current study also performed better than TFHs of different design in another previous study [6]. Therefore, TFHs with hybrid structure are expected to have high potential for industrial and commercial applications.

IV. CONCLUSION

TFH performance of OMO structure in relation to Ag thickness was investigated. In the case of an ITO monolayer (95 nm), the sample temperature increased to only 32°C under 3 V during 100 sec and then it was not changed with increasing time. On the other hand, the hybrid layers ITO(40)/Ag/ITO(40 nm) showed excellent heating efficiency under the same condition indicating that the sample temperatures increased to 92°C (Ag 10 nm), 131°C (Ag 15 nm) and 145°C (Ag 20 nm). The hybrid layer [ITO(40)/Ag(15)/ITO(40 nm)] shows nearly the same transmittance (81.7%, @550 nm) as the ITO monolayer (81.6%, @550 nm) of identical thickness, as a result of index-matching by the OMO structure. The ITO monolayer was not completely defrosted within 30 sec, while the hybrid layer [ITO(40 nm)/Ag(15 nm)/ITO(40 nm)] defrosted in only 5 sec under 3 V. This indicates superior performance in comparison to other previous TFH reports. Therefore, it is confirmed that the hybrid structure, ITO/Ag/ITO, has high potential for TFH applications, as shown in optical transmittance, HGR, uniformity of heat distribution, and defrosting test results.

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