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Optical and Electrical Properties of Oxide Multilayers

Sangmin Han, Jiao Long Yu, and Sang Yeol Lee†
Department of Semiconductor Engineering, Cheongju University, Cheongju 28503, Korea

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Oxide/metal/oxide (OMO) thin films were fabricated using amorphous indium-gallium-zinc-oxide (a-IGZO) and an Ag metal layer on a glass substrate at room temperature. The optical and electrical properties of the a-IGZO/Ag/a-IGZO samples changed systemically depending on the thickness of the Ag layer. The transmittance in the visible range tends to decrease as the Ag thickness increases while the resistivity, carrier concentration, and Hall mobility tend to improve. The a-IGZO/Ag (13 nm)/a-IGZO thin film with the optimum Ag thickness showed an average transmittance (T_{av}) of 71.7%, resistivity of 6.63×10^{-5} Ω·cm and Hall mobility of 15.22 $cm^2V^1s^{-1}$.

Keywords: a-InGaZnO, Multilayer, Ag thickness, Hall measurement, Transparent conductive oxide

1. INTRODUCTION

Transparent conductive oxide (TCO) materials have attracted a significant amount of attention due to their potential for use in transparent and flexible displays [1,2]. Of these, indium-tin-oxide (ITO) is the material that has been most extensively used. ITO thin films exhibit a high conductivity and a high transparency in the visible range [3]. However, indium is an expensive rare earth material [4]. As a result, aluminum-zinc-oxide (AZO) and gallium-zinc-oxide (GZO) materials could become good alternatives to ITO because they have a low resistivity in the order of 10-4 Ω·cm despite being indium-free materials [5,6]. However, these materials still cannot be applied in large area, flexible substrates due to their polycrystalline structure [7,8].

Multilayer oxide-metal-oxide (OMO) thin films have been extensively studied for use as the TCO material in transparent electronics, mainly due to their high transparency in the visible range and metal-like electrical conductivity. Previous studies have reported that polycrystalline oxide such as ITO and AZO have been applied as top and bottom oxide layers in OMO multilayer structures [9,10]. However, it is difficult to apply flexible substrates in this case due to several problems, including cracking, delamination and brittleness when subjected to bending stress test. Recently, many researchers have studied amorphous oxide thin films applied as top and bottom layers of OMO structures to obtain flexible TCO [11,12].

Amorphous oxide thin films, such as amorphous indium-gallium-zinc-oxide (a-IGZO), are expected to be a suitable material for fabrication on flexible substrates due to their high transparency, low temperature process, flexibility and high electrical properties, despite their amorphous structure [13].

In this work, we investigate the characteristics of multilayer a-IGZO/Ag/a-IGZO according to the thickness of the Ag layer. The results indicate that the thickness of the metal layer determines the conductivity and transmittance of the OMO multilayer structures. We expected that the thickness of the Ag layer in the OMO multilayer need to be optimized to realize flexible TCO.

2. EXPERIMENTS

We have fabricated a-IGZO/Ag/a-IGZO thin films on a glass substrate (Corning1737). The a-IGZO thin films were used as top and bottom oxide layers and were deposited via radio frequency (RF) magnetron sputtering at room temperature. RF sputtering was conducted at a working pressure of 3 mTorr with an Ar-only atmosphere, by supplying an RF power of 30 W. The Ag film inserted between top and bottom oxide layers was deposited via DC magnetron sputtering at room temperature. DC sputtering was conducted at a working pressure of 4 mTorr with an Ar-only atmosphere by supplying a DC power of 30 W. The thickness of the Ag layer was systematically changed, ranging from 7 nm to 15

† Author to whom all correspondence should be addressed:
E-mail: sylee@cju.ac.kr

nm with 2 nm steps. We have used a UV-VIS spectrometer (Cary 5000 UV-Vis-NIR, Agilent) and Hall measurements to analyze the optical and electrical properties of the a-IGZO/Ag/a-IGZO thin films as the thickness of the Ag layer varied.

3. RESULTS AND DISCUSSION

Figure 1 shows the schematic diagram and equivalent circuit model of the a-IGZO/Ag/a-IGZO thing film. It is interesting to note that the total resistance of the OMO multilayer structure was consistent with the parallel-connected resistance of the top/bottom oxide layer and metal layer. This model can be used to define the resistivity on the OMO multilayer as follows [14]:

$$\frac{1}{R_{OMO}} = \frac{1}{R_{oxide\_t}} + \frac{1}{R_{metal}} + \frac{1}{R_{oxide\_b}} \quad (1)$$

where R_{oxide_t} and R_{oxide_b} are the resistances of the top and bottom oxide layers, respectively. The total resistivity of the a-IGZO/Ag/a-IGZO thin film was the dominant property according to the thickness of the Ag layer, and this could be mainly attributed to: (1) the thickness of the top and bottom oxide layers being fixed during the change in the Ag thickness; (2) the main current path occurred through the Ag layer due to lower resistivity for Ag than for the oxide layer.

Figure 2 shows the transmittance in the visible range for the a-IGZO/Ag/a-IGZO thin film according to the thickness of the Ag layer. It is well known that in an OMO multilayer structure, the transmittance increases according to the decrease in the thickness of the Ag layer. However, the a-IGZO/Ag (7 nm) /a-IGZO thin films showed a relatively low transmittance despite the thinner Ag layer when compared to other a-IGZO/Ag/a-IGZO thin films. This result can be explained by the scattering effect that is attributed to discontinuous Ag island formation [15].

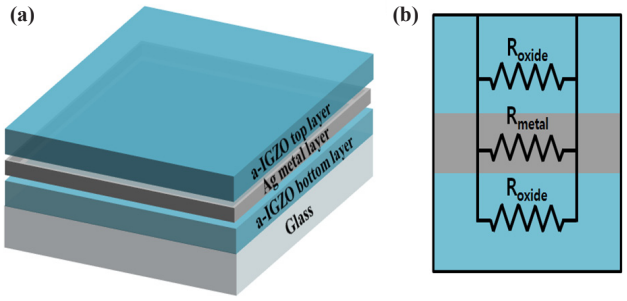

Fig. 1. (a) Schematic diagram and (b) equivalent circuit model of the multilayer a-IGZO/Ag/a-IGZO thin films.

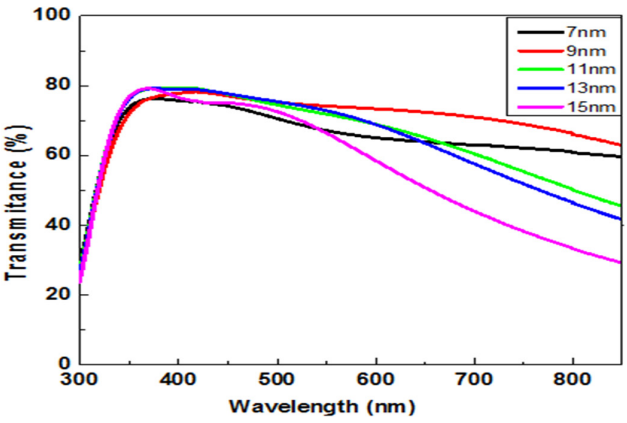

Fig. 2. Optical transmittance of a-IGZO/Ag/a-IGZO thin films depending on the thickness of the Ag layer.

Table 1. Average transmittance of a-IGZO/Ag/a-IGZO thin films depending on the thickness of the Ag layer.

Ag thickness (nm)	7	9	11	13	15
T_{av} (%)	67.1	74.1	71.2	71.7	64.5

Table 1 shows the average transmittance (T_{av}) of the a-IGZO/Ag/a-IGZO multilayers in the wavelength range from 380 nm to 780 nm. The T_{av} was calculated using the following relation [16]:

$$T_{av} = \frac{\int V(\lambda)T(\lambda)d\lambda}{\int V(\lambda)d\lambda}, (380\,nm < \lambda < 780\,nm) \quad (2)$$

where V(λ) is the luminous spectral efficiency, and T(λ) is the transmittance measured for the film system. We confirmed that the a-IGZO/Ag (9 nm)/a-IGZO thin film exhibited a higher T_{av} of 74.1% than the others. A further increase in the thickness of Ag to 15 nm resulted in a decrease of T_{av} to 64.5%. The behavior of T_{av} with an Ag thickness from 9 nm to 15 nm can be explained by the Drude model. In the Drude model, the permittivity of TCO is a complex dielectric function that relates the optical properties of a solid to the underlying electronic structure, allowing the use of optical measurements to understand the underlying electronic properties of materials. That is, the imaginary part (ε") denotes the optical losses, as shown in Eq. 3. [17]

$$i\varepsilon'' = i\frac{\omega_p^2\gamma_p}{(\omega^2+\gamma_p^2)\omega}, \omega_p = \sqrt{\frac{ne^2}{m^*\varepsilon_0}} \quad (3)$$

where ω_p is the plasma frequency, γ_p is the Drude relaxation rate, and n and m* are the carrier concentration and mass of the electron, respectively. In conventional plasmonic materials, Naik et al. reported that the imaginary part of the Drude model (ε", denoted the optical losses) in the visible range increased according to an increase in the carrier concentration. When the Ag layer is well formed, the carrier concentration (n) increase proportionally to the Ag thickness. There is a corresponding increase in the plasma frequency (ω_p), and therefore, the imaginary part, which relates the optical loss, increases. Namely, the transmittance will decreases depending on an increase in the carrier concentration. We confirm this mechanism by investigating the electrical properties of the a-IGZO/Ag/a-IGZO thin films by obtaining Hall measurements.

Figure 3 shows the electrical properties of the a-IGZO/Ag/a-IGZO thin films measured using a four point probe method. This method measures the resistivity of thin film using two parameters as follows [16]:

$$\rho = 1/ne\mu \quad (4)$$

where is the resistivity, n is the number of charge carriers, e is the charge of the carrier, and μ is the carrier mobility. The resistivity decreases according to an increase in the thickness of the Ag layer, while the bulk concentration and mobility are increased proportionally with the increase in the thickness of the Ag layer. These results confirm that the optical and electrical properties of the a-IGZO/Ag/a-IGZO thin films depending on the thickness of the Ag layer are consistent with the Drude model mechanism de-

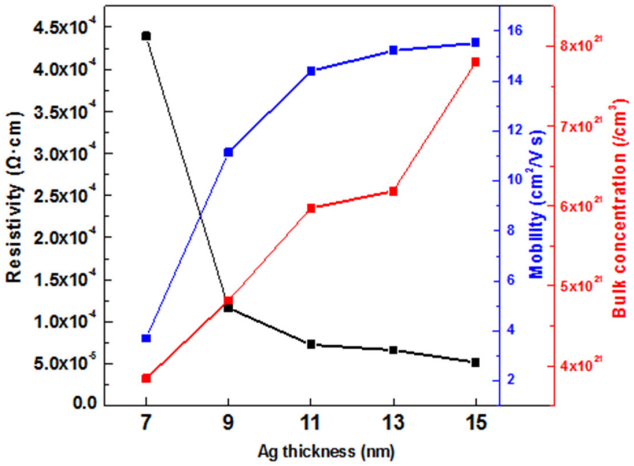

Fig. 3. Hall measurement results for the a-IGZO/Ag/a-IGZO thin films as a function of Ag thickness.

scribed above. In addition, the optimum Ag thickness is 13 nm, which shows a high transmittance and low resistivity. Our results show the systematic change in both the electrical and optical properties and are comparable to previous reports presented by other groups [18,19].

Therefore, the proposed a-IGZO/Ag/a-IGZO thin films can be implemented in TCO applications to control the optical and electrical properties by changing the thickness of the Ag layer.

4. CONCLUSIONS

In summary, we have investigated the influence that the thickness of the Ag layer has on the optical and electrical properties of a-IGZO/Ag/a-IGZO thin films by measuring the visible transmittance and taking Hall measurements. The a-IGZO/Ag/a-IGZO thin films were fabricated at room temperature with various Ag thicknesses ranging from 7 nm to 9 nm, and a systemic change was observed in the optical and electrical properties, including T_{av}, resistivity, mobility and bulk concentration. In particular, we confirmed that our results are consistent with the Drude model through the relation between T_{av} and the bulk concentration. When the optimum thickness of 13 nm of the Ag interlayer is used, the resulting a-IGZO/Ag/a-IGZO thin film shows a T_{av} of 71.7%, resistivity of 6.63×10^{-5} Ω·cm, mobility of 15.22 $cm^2V^{-1}s^{-1}$ and bulk concentration of 6.2×10^{21} cm^{-3}. These results indicate that the OMO multilayer with an amorphous oxide thin film is a promising candidate to replace conventional TCO electrodes, including ITO, AZO, carbon nanotubes and Ag nanowires.

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