

# Deposition of Transparent IGZO Conducting Thin Films by Co-Sputtering of $Zn_2Ga_2O_3$ and $In_2O_3$ Targets at Room Temperature

Yu-Hsin Chen, Yuan-Tai Hsieh, Cheng-Shong Hong, Chia-Ching Wu, Cheng-Fu Yang, Yu-Jhen Liou

**Abstract**—In this study, we investigated  $(In,Ga,Zn)O_x$  (IGZO) thin films and examined their characteristics of using  $Ga_2O_3$ -2 ZnO (GZO) co-sputtered  $In_2O_3$  prepared by dual target radio frequency magnetron sputtering at room temperature in a pure Ar atmosphere. RF powers of 80 W and 70 W were used for GZO and pure  $In_2O_3$ , room temperature (RT) was used as deposition temperature, and the deposition time was changed from 15 min to 60 min. Structural, surface, electrical, and optical properties of IGZO thin films were investigated as a function of deposition time. Furthermore, the GZO co-sputtered  $In_2O_3$  thin films showed a very smooth and featureless surface and an amorphous structure regardless of the deposition time due to the room temperature sputtering process. We would show that the co-sputtered IGZO thin films exhibited transparent electrode properties with high transmittance ratio and low resistivity.

**Keywords**—IGZO, co-sputter,  $Ga_2O_3$ -2 ZnO,  $In_2O_3$ .

## I. INTRODUCTION

INDIUM gallium zinc oxide is a semiconducting material, consisting of indium (In), gallium (Ga), zinc (Zn) and oxygen (O), and often abbreviated as “IGZO”. Transparent field-effect transistors were developed by Professor Hideo Hosono's group at Tokyo Institute of Technology and Japan Science and Technology Agency (JST) in 2003 (crystalline IGZO-TFT) [1]. They used a single-crystalline thin-film transparent oxide semiconductor,  $InGaO_3(ZnO)_5$ , as an electron channel and amorphous hafnium oxide as a gate insulator. In 2004, they proposed to a novel transparent amorphous oxide semiconductor by using the In-Ga-Zn-O system ( $\alpha$ -IGZO) as the active channel in transparent thin-film transistors (TTFTs) [2]. The  $\alpha$ -IGZO was deposited on polyethylene terephthalate at room temperature and exhibited Hall Effect mobility exceeding  $10\text{ cm}^2V^{-1}s^{-1}$ , which is an order of magnitude larger

than for hydrogenated amorphous silicon.

Today, conventional active-matrix (AM) flat panel displays (FPDs) are based on amorphous or polycrystalline silicon thin-film transistor (TFT) technology. Limitations of amorphous silicon include visible light sensitivity and low field-effect mobility, which reduce the pixel aperture ratio and driving ability for some applications. Although polycrystalline silicon TFTs have larger field-effect mobility, their uniformity over large area might not acceptable for high yield manufacturing. Conventional metal oxide semiconductors such as zinc oxide (ZnO) are polycrystalline in nature, even at room temperature. The grain boundaries of such metal oxides could affect device properties, uniformity and stability over large areas. To overcome this issue, the IGZO thin films have been proposed for use as the channel layer in TFTs [2]. A protective layer is required in the IGZO TFTs with the inverse staggered type bottom-gate structure because the back channel of the IGZO active layer is damaged during the deposition and etching processes of the source/drain electrode. In the past, Cho et al. reported the effects of annealing and passivation on the electrical characteristics of transparent bottom-gate IGZO TFTs [3]. Among various oxide semiconductors, amorphous  $InGa(ZnO)_m$  film has been extensively investigated as a channel layer for transparent TFTs due to its high mobility, transparency and simple fabrication process using sputtering techniques [1]-[5]. However, Jeong and Kim used the co-sputtering of  $Ga:In_2O_3$  and  $Zn:In_2O_3$  targets to deposit the Ga and Zn co-doped  $In_2O_3$  electrode at room temperature [6]. In this study, we also used co-sputtering of  $Zn_2Ga_2O_3$  and  $In_2O_3$  targets to deposit the IGZO electrode at room temperature. This study examined the structural, surface, optical, and electrical properties of the IGZO electrodes on a glass substrate as a function of deposition time at a constant working pressure of  $3\times 10^{-3}$  torr, and a pure Ar flow rate of 50 sccm using a dual target sputtering system at room temperature.

## II. EXPERIMENTAL

$Ga_2O_3$  powder (99.99%) was mixed with ZnO powder (99.99%) to form the  $Ga_2O_3$ -2 ZnO composition (abbreviated as GZO). After being dried and ground, the GZO powder was calcined at  $800^\circ C$  for 1 h, then ground again. GZO powder and pure  $In_2O_3$  powder were mixed with polyvinylalcohol (PVA) as binder. The mixed powders were uniaxially pressed into pellets of 5 mm thickness and 54 mm diameter using a steel die. After debinding, the GZO and pure  $In_2O_3$  pellets were sintered at various  $1200^\circ C$  and  $1250^\circ C$ , respectively, for 2 h. Glass

Yu-Hsin Chen and Yu-Jhen Lio are with the Department of Chemical and Materials Engineering, National University of Kaohsiung, Taiwan, Kaohsiung, 81148, R.O.C. (e-mail: j1040404@gmail.com, sonic7838@hotmail.com).

Cheng-Fu Yang is with the Department of Chemical and Materials Engineering, National University of Kaohsiung, Taiwan, Kaohsiung, 81148, R.O.C. (corresponding author to provide phone: 886-7-5919283; fax: 886-7-5919277; e-mail: cfyang@nuk.edu.tw)

Yuan-Tai Hsieh is with the Department of Electronic Engineering, Southern Taiwan University, Tainan, Taiwan, 71005, R.O.C. (e-mail: ythsieh@mail.stust.edu.tw).

Cheng-Shong Hong is with the Department of Electronic Engineering National Kaohsiung Normal University, Kaohsiung, 82444, Taiwan, R.O.C. (e-mail: cshong@nkn.edu.tw).

Chia-Ching Wu is with the Department of Electronic Engineering, Kao Yuan University, Kaohsiung 82151, Taiwan, R.O.C. (e-mail: 9113718@gmail.com).

substrates (Corning 1737) with an area of  $2 \times 2 \text{ cm}^2$  were cleaned ultrasonically with isopropyl alcohol (IPA) and deionized (DI) water, and then dried under a blown nitrogen gas. Then, GZO and pure  $\text{In}_2\text{O}_3$  targets were used to deposit the thin films at the same time. RF powers of 80 W and 70 W were used for GZO and pure  $\text{In}_2\text{O}_3$ , respectively, room temperature (RT) was used as deposition temperature, and the deposition time was changed from 15 min to 60 min. The base pressure of the sputtering chamber was below  $5 \times 10^{-6}$  Torr and the working pressure was maintained at  $3 \times 10^{-3}$  Torr in pure Ar (99.99%) ambient (abbreviated as-deposited IGZO thin films). Thickness and surface morphology of the IGZO thin films were measured using a field emission scanning electron microscopy (FESEM), and their crystalline structures were measured using X-ray diffraction (XRD) patterns with Cu  $K\alpha$  radiation ( $\lambda=1.5418 \text{ \AA}$ ). The optical transmission spectrum was recorded using a Hitachi U-3300 UV-Vis spectrophotometer in the 300–1000 nm wavelength range. While the Hall-effect coefficient of the IGZO thin films was measured using a Bio-Rad Hall set-up.

### III. RESULTS AND DISCUSSION

FESEM was used to examine the surface roughness and morphology of the IGZO electrodes grown on a glass substrate at different deposition time. Fig. 1 shows the FESEM surface images of the IGZO electrode with increasing deposition time, which indicates that as deposition time was changed, the surface morphologies apparently changed as well. As deposited at room temperature at 15 min and 30 min, as Figs. 1 (a) and (b) show, morphology of the IGZO thin films exhibited a very smooth surface regardless of deposition time. Surface morphology of 45min-deposited IGZO thin films, which also showed the nano-particle structure of the IGZO grains, had the similar result with those of 15 min- and 30 min-deposited IGZO thin films and it is not shown here. Although 60 min-deposited IGZO thin films exhibited some agglomerated surface images in Fig. 1 (c), it was not microcrystalline or a cluster of crystallines as being confirmed by XRD in Fig. 3. Therefore, all IGZO thin films regardless of deposition time showed stable amorphous surface features. In order to achieve high performance transparent oxide TFTs or memory devices, the preparation of source and drain electrodes with a smooth surface morphology is very important because surface roughness of the IGZO thin films will influence the leakage current between the semiconducting IGZO active layer and source/drain electrodes.

Fig. 2 shows the cross section observations of the IGZO thin films deposited at different deposition time. Calculated the results in Fig. 2, thickness of the IGZO thin films increased with increasing deposition time. Thickness of the IGZO thin films was around 108 nm, 275 nm, 460 nm, and 532 nm, respectively, as the deposition time was 15 min, 30 min, 45 min, and 60 min. As the cross-session micrographs shown in Fig. 2 are compared, there are different results as the deposition time is changed. As 15 min was used as deposition time, the IGZO thin films grew like a flat plane and no special morphology was observed; As 15 min and 45 min were used as deposition time, the deposited IGZO thin films had the

structure of highly oriented bars parallel to the substrate normal. As deposition time was increased 60 min, the nano-bar-aggregated growths were transformed into the irregular plate-shaped growths along the up direction. These results prove the IGZO thin films deposited at different time have different surface morphology, as Fig. 1 shows. In addition, there is no evidence of the segregation of the GZO and  $\text{In}_2\text{O}_3$  due to the uniform co-sputtering of the GZO and  $\text{In}_2\text{O}_3$  targets using tilted cathode guns.

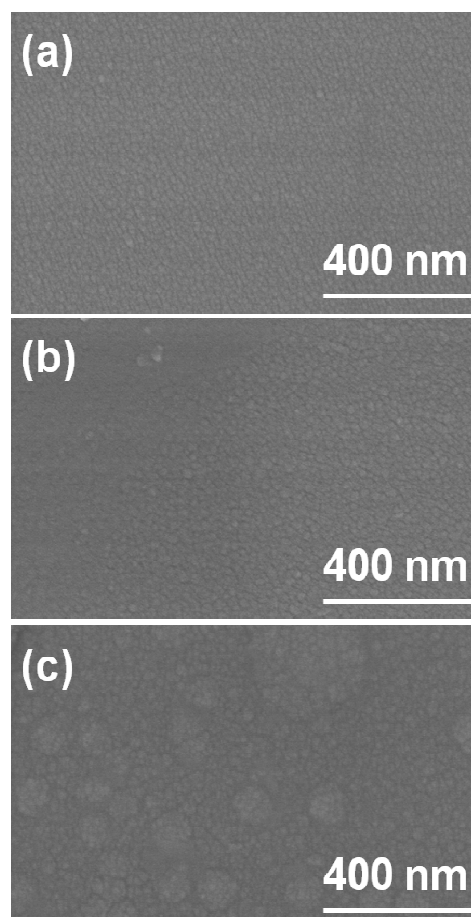


Fig. 1 Surface morphology of the IGZO thin films as a function of deposition time (a) 15min, (b) 30 min, and (c) 60 min, respectively

XRD was used to investigate the structural properties of the IGZO thin films grown at room temperature in a pure Ar atmosphere. Fig. 3 shows the XRD patterns of the 15 min-, 30 min-, 45 min-, and 60 min-deposited IGZO thin films with the different thicknesses. As expected from FESEM, XRD patterns of the 15 min- and 30 min-deposited IGZO thin films showed one weak and broad peak, and XRD patterns of the 45 min- and 60 min-deposited IGZO thin films show two weak and broad peaks. One weak and broad peak is assigned to the glass substrate and two weak and broad peaks are assigned to the glass substrate and IGZO thin films, respectively. Those results suggest that all the IGZO thin films exhibit the amorphous phase.

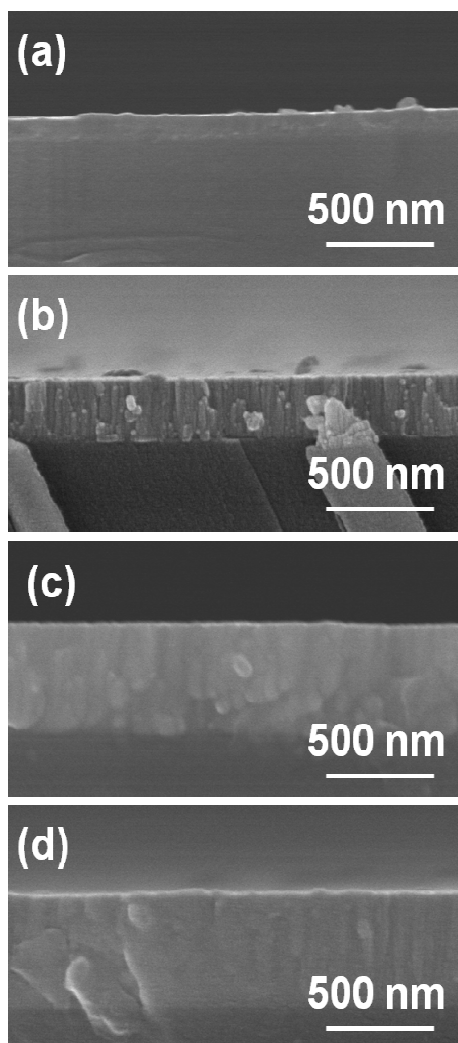


Fig. 2 Cross section observations of the IGZO thin films as a function of deposition time (a) 15min, (b) 30 min, (c) 45 min, and (d) 60 min, respectively

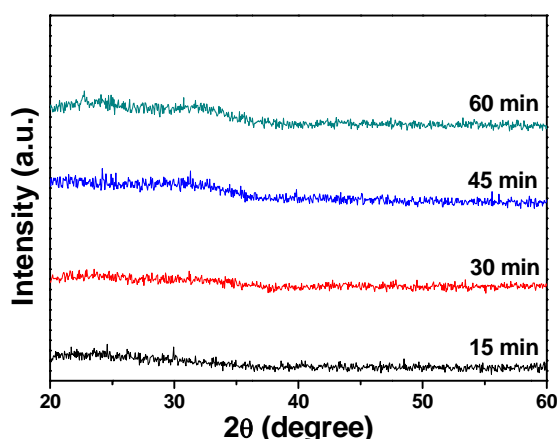


Fig. 3 XRD patterns of the IGZO thin films as a function of deposition time

Fig. 4 shows the transmission ratios of the IGZO thin films plotted against wavelengths in the region of 300–1000 nm, with

deposition time as the parameter. As deposition time was 15 min, 30 min, and 45 min, the average transmittance ratio of the IGZO thin films in the range of 400 nm~700 nm was 86.3%, 87.4%, and 85.9%, respectively. Those results suggest that the transmittance ratios of the IGZO thin films are almost unchanged as the deposition time is equal and less 45 min. however, as the deposition time was 60 min, the average transmittance ratio of the IGZO thin films in the range of 400 nm~700 nm was 77.1%. From the results shown in Fig. 1, the agglomerated surface images are the reason to cause the decrease in average transmittance ratio. For the transmission spectra shown in Fig. 4, as longer deposition time was used, the optical band edge shifted to a longer wavelength was observable and a greater sharpness was noticeable in the curves of the absorption edge, which suggest a decrease in the  $E_g$  values. Fig. 4 also shows that the IGZO thin films deposited on glass substrates have high transmittance ratio of over 86.1%, 92.6%, 92.6%, and 85.4% in the near-infrared region (700 nm ~ 1000 nm) as deposition time was 15 min, 30 min, 45 min, and 60 min, respectively.

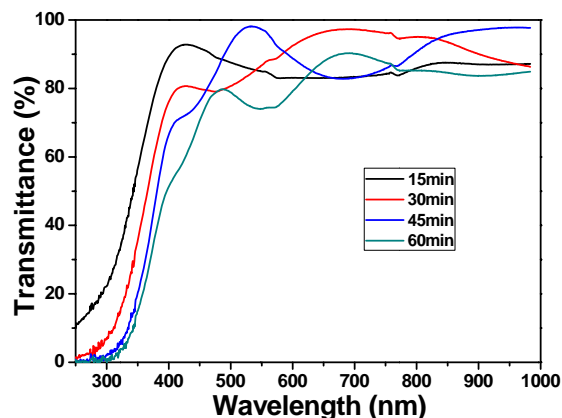


Fig. 4 Transmittance spectrum of the IGZO thin films as a function of deposition time

The results of the carrier mobility, carrier concentration, and resistivity shown Fig. 5 indicate that the electrical properties of the IGZO thin films were dependent on deposition time. When plasma molecules are deposited on a glass substrate, many defects result and inhibit electron movement. As thin films are deposited at room temperature, two factors are believed to influence the carrier mobility of the IGZO thin films. First, depositing at room temperature cannot provide enough energy to enhance the motion of plasma molecules, which will improve the crystallization and grain size growth of the IGZO thin films, the defects in the IGZO thin films will generate during the deposition process. Second, as deposition time is 60 min, because the agglomerated particles in the GZO thin films increases, and that will cause the decrease in the inhibiting of the barriers electron transportation [7] and the mobility will increase. However, as Fig. 5 shows, both the carrier concentration and carrier mobility have no apparent variation as the deposition time increases from 15 min to 60 min. As the deposition time was changed from 15 min to 60 min, the carrier mobility increased from 15.2  $\text{cm}^2/\text{V}\cdot\text{s}$  to 27.4  $\text{cm}^2/\text{V}\cdot\text{s}$  and the

carrier concentration increased from  $1.91 \times 10^{20} \text{ cm}^{-3}$  to  $6.56 \times 10^{21} \text{ cm}^{-3}$ , respectively. The resistivity of the TCO thin films is proportional to the reciprocal of the product of carrier concentration  $N$  and mobility  $\mu$ :

$$\rho = 1 / Ne\mu \quad (1)$$

Both the carrier concentration and the carrier mobility contribute to the conductivity. As deposition time was changed from 15 min to 60 min, the resistivity of the IGZO thin films changed from  $9.62 \times 10^{-3} \Omega\text{-cm}$  to  $1.74 \times 10^{-3} \Omega\text{-cm}$ . The minimum resistivity of the IGZO thin films at a deposition time of 30 min is mainly caused by the carrier concentration at its maximum.

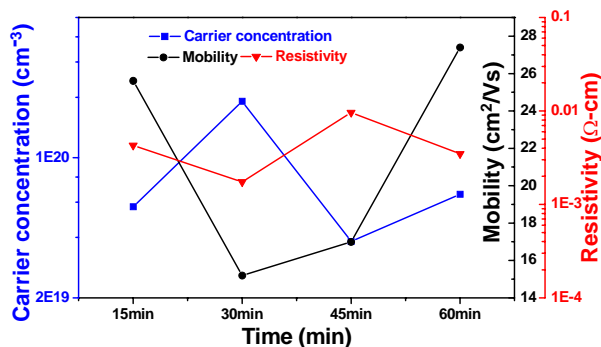


Fig. 5 Hall mobility, carrier concentration, and resistivity of the IGZO thin films as a function of deposition time.

#### IV. CONCLUSION

This study examined the characteristics of IGZO thin films prepared by  $\text{Ga}_2\text{O}_3\text{-2 ZnO}$  (GZO) co-sputtered  $\text{In}_2\text{O}_3$  as an alternative method to investigate the characteristics of the IGZO thin films. Thickness of the co-sputtered IGZO thin films was around 108nm, 275 nm, 460 nm, and 532 nm, as the deposition time was 15 min, 30 min, 45 min, and 60 min. As deposition time was 15 min, 30 min, and 45 min, the average transmittance ratio of the IGZO thin films in the range of 400 nm~700 nm was 86.3%, 87.4%, and 85.9%, and the high average transmittance ratio of over 86.1%, 92.6%, 92.6%, and 85.4% in the near-infrared region (700 nm ~ 1000 nm), respectively. As the deposition time was changed from 15 min to 60 min, the carrier mobility increased from  $15.2 \text{ cm}^2/\text{V-s}$  to  $27.4 \text{ cm}^2/\text{V-s}$ , which are higher than those of the most reported IGZO thin films.

#### ACKNOWLEDGMENT

The authors acknowledge financial supports of Nanowin Technology Co. Ltd., NSC 102-2622-E-390 -002-CC3 and NSC 102-2221-E-390-027.

#### REFERENCES

[1] K. Nomura, H. Ohta, K. Ueda, T. Kamiya, M. Hirano, and H. Hosono, "Thin-Film Transistor Fabricated in Single-Crystalline Transparent Oxide Semiconductor," *Science*, vol. 300, no. 5623 pp. 1269-1272, May 2003.

[2] K. Nomura, H. Ohta, A. Takagi, T. Kamiya, M. Hirano, and H. Hosono, "Room-temperature fabrication of transparent flexible thin-film transistors using amorphous oxide semiconductors," *Nature*, vol. 432, no. 7016, pp. 488-492, November 2004.

[3] D. H. Cho, S. H. Yang, J.H. Shin, C. W. Byun, M. K. Ryu, J. I. Lee, C. S. Hwang, and H. Y. Chu, "Passivation of Bottom-Gate IGZO Thin Film Transistors," *J. Korean Phys. Soc.* vol. 54, no. 1, pp. 531-534, January 2009.

[4] H. Yabuta, M. Sano, K. Abe, T. Aiba, T. Den, H. Kumomi, K. Nomura, Toshio Kamiya, and H. Hosono, "High-mobility thin-film transistor with amorphous  $\text{InGaZnO}_4$  channel fabricated by room temperature rf-magnetron sputtering," *Appl. Phys. Lett.* 90 (2007) 22114.

[5] C.C. Lo and T.E. Hsieh, "Preparation of IGZO sputtering target and its applications to thin-film transistor devices," *Ceram. Inter.* vol. 38, no. 5, pp. 3977-3983, 2012.

[6] J.A. Jeong and H.K. Kim, "Transparent Ga and Zn co-doped  $\text{In}_2\text{O}_3$  electrode prepared by co-sputtering of Ga: $\text{In}_2\text{O}_3$  and Zn: $\text{In}_2\text{O}_3$  targets at room temperature," *Thin Solid Films*, vol. 519, no. 10, pp. 3276-3282, 2011.

[7] Y. Igasaki and H. Saito, "Substrate temperature dependence of electrical properties of ZnO:Al epitaxial films on sapphire (1210)," *J. Appl. Phys.* vol. 69, no. 4, pp. 2190-2195, 1991.