ARCHIVES OF ELECTRICAL ENGINEERING VOL. 72(2), pp. 555–563 (2023)

DOI 10.24425/aee.2023.145425

Indium-gallium-zinc oxide thin-film preparation via single-step radio frequency sputter deposition using mixed-oxide powder targets

Takahiko Satake[1] ✉, **Hiroharu Kawasaki**[2], **Shin-Ichi Aoqiu**[1]

[1] *Graduate School of Engineering Sojo University*
Ikeda Nishi-ku Kumamoto City, Japan

[2] *Department of Electrical and Electronic Engineering,*
National Institute of Technology, Sasebo College
Okishin-machi, Sasebo City, Nagasaki Pref., Japan

e-mail: ✉*satake@sasebo.ac.jp*

(Received: 31.09.2022, revised: 02.03.2023)

Abstract: Indium gallium zinc oxide (In–Ga–Zn–O) thin films, which are transparent conductive films for liquid crystals and electroluminescent displays, were fabricated via single-step sputter deposition using one target containing different proportions of indium oxide, gallium oxide, and zinc oxide powders. Experimental results suggest that the In–Ga–Zn–O thin films can be prepared using the method of single-step radio frequency (RF) sputter deposition, applying a powder target containing indium oxide, gallium oxide, and zinc oxide. The In–Ga–Zn–O thin films were prepared on Si substrates, and the deposition rate depended on the target composition. In these plasma processes, electron density and temperature were essentially independent of target composition. The prepared films were very smooth with a root-mean-square roughness of less than 10 nm. The crystallinity of the ZnO peak was observed in all the films; whereas the In and Ga peaks were not observed in the films prepared. The X-ray photoelectron spectroscopy of the films also revealed that the elemental concentration ratio of In–Ga–Zn–O thin films could be prepared using one target, and that can be easily controlled by ratios in the In_2O_3/Ga_2O_3/ZnO composition in the powder target. The transmittances were > 75% at 800 nm for all the target mixtures, and increased with increasing In_2O_3 in the powder target.

Key words: In–Ga–Zn–O thin-film, plasma processes, powder target, sputtering deposition, transparent conductive film

1. Introduction

Indium gallium zinc oxide (In–Ga–Zn–O:IGZO) thin-film transistors (TFTs) are promising alternatives to Si TFT channels for next-generation active matrix panels used in display applications. IGZO TFTs have excellent features such as high electron mobilities, low threshold voltages, and facile control of carrier concentrations. In addition, IGZO TFTs have production advantages because existing Si:H TFT facilities can be used without modification. However, it is challenging to adjust the commercial sputter deposition conditions for fabrication of high-quality and high-performance IGZO thin films. Usually, separate targets are fabricated first, and then the film is deposited from each of the targets. Hence, high-quality IGZO thin films are costly, and optimum deposition conditions can change after many depositions [1, 2].

Sputtering is widely used to deposit thin films because it requires a simple apparatus, it is an easy process with an expansive choice of materials, and it produces high-uniformity films at high deposition rates [3–15]. In addition, it uses non-toxic gases and low energies to generate the processing plasma. It is thus safer than other plasma processing methods such as chemical-vapor deposition and pulsed-laser deposition. Sputtering has been used to deposit functional thin films composed of multiple materials [16, 17]. Preparing thin films consisting of multiple elements is difficult with conventional sputter deposition because high-density bulk targets (> 3 g/cm^3, > 95% density) are generally used. Therefore, it is necessary to make new targets by other methods, such as spark plasma sintering, and/or conduct sputter deposition using multiple targets [18, 19]. These target preparations are usually time-consuming and costly. As an alternative, plasma processes using multielement powder targets have been used to prepare multielement thin films in single-step processes. Several kinds of powders can be mixed and placed in a target holder in a vacuum chamber. Thin films are then deposited on a substrate mounted on the opposite side of the vacuum chamber with respect to the target holder. It is possible to obtain a few samples with different doping densities during one technological cycle [16]. This approach was also used to fabricate functional thin films containing multiple elements [19–34]. For example, bismuth iron garnet ($Bi_3Fe_5O_{12}$) films used for magneto-optics were fabricated by sputter deposition of mixed bismuth (Bi) and iron (Fe) powder targets. We prepared Sn-doped SiO_2 thin films using SnO_2 and SiO_2 mixed-powder targets [18, 22]. The elemental concentrations in the thin films could be controlled by the composition of the powder mixture. In this experiment, In-Ga-Zn oxide thin films were prepared via one-step sputter deposition using indium oxide, gallium oxide, and zinc oxide mixed-powder targets. The film properties, such as crystallinity, composition ratio, and surface roughness, were characterized.

2. Experimental methods

A schematic of the sputter deposition apparatus is shown in Fig. 1 The stainless-steel deposition chamber had a diameter of 400 mm and a height of 450 mm. A powder target was placed in a 50.8-mm-diameter stainless-steel target holder. The spherical shape of In_2O_3 (40–300 µmϕ, 2 g), Ga_2O_3 (100–500 µmϕ, 2 g), and ZnO (20–200 µmϕ, 2 g) powders were mixed mechanically and then placed in the target holder ((1) in Fig. 1). The mixed powder target was placed in a sealed container, and then the container was rotated 60 times per minute for 24 hours to mix the targets

((2) in Fig. 1). The weight ratios of the powders were changed to vary the target composition ((3) in Fig. 1). The chamber was vacuumed to a base pressure of $5 \cdot 10^{-3}$ Pa using turbomolecular and rotary pumps ((4) in Fig. 1). The total deposition pressure was 30 Pa with argon gas (99.99%) introduced at a 10-sccm flow rate ((5) in Fig. 1). Sputtering deposition was performed by radio frequency (13.56 MHz) plasmas. The plasma was generated by applying 100 W discharge power to the powered electrode ((6) in Fig. 1). Prior to loading in the deposition chamber, substrates were cleaned in an ultrasonic agitator several times, followed by washing with high-purity deionized water. The surface of the substrate was at room temperature and the deposition time was 100 min.

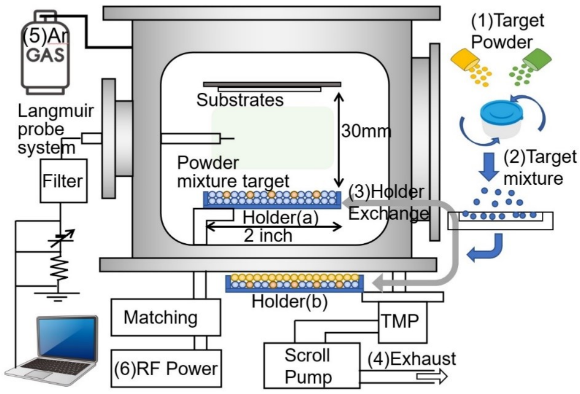

Fig. 1. A schematic of the sputter deposition apparatus

The electron density and temperature of the processing plasma were measured using a Langmuir probe (ARIOS LMP-100). The probe was composed of a tungsten wire with a diameter of 0.3 mm and a length of 0.5 mm. The surface morphology and roughness of the film was measured via atomic force microscopy (AFM; JEOL JSTM-4100). The film thickness was measured using an alpha-step profilometer (Kosaka Lab. Surfcorder ET4000A). The crystalline structure and composition of the films were measured, respectively, via X-ray diffraction (XRD; RIGAKU RINT2100V) and XPS (JEOL JPS9010).

The light transmittance of the thin films was measured using an ultra violet-visible spectrophotometer (UV-Vis; Shimadzu UV-mini 1240). The resistivity of the thin films was measured by a hand-made four-probe resistivity measurement system.

3. Results and discussion

Figure 2 shows the dependence of the electron density and temperature in the processing plasma on the powder target mixture. Both affected the deposition process of the thin films and were measured using a Langmuir probe during sputter deposition. Here, the target powder mixture was in terms of the weight ratios $In_2O_3/Ga_2O_3/ZnO$, and the plasma measurements were collected 15 mm from the substrate surface. The electron density was approximately 10^9–10^{10} cm^{-3} and almost independent of the powder mixture. For example, In_2O_3:Ga_2O_3:ZnO ratios of 1:1:1, 2:1:1, and 3:1:1 had electron densities of $2.0 \cdot 10^9$, $2.4 \cdot 10^9$, and $2.8 \cdot 10^9$ cm^{-3}, respectively, as shown in Fig. 2 The electron densities for the ratios 2:1:1, 1:2:1, and 1:1:2 at 100 W were $2.4 \cdot 10^9$, $2.0 \cdot 10^9$, and $1.8 \cdot 10^9$ cm^{-3}, respectively. The electron temperature was approximately 1–2 eV in all cases. Therefore, the electron density and temperature were essentially independent of target composition.

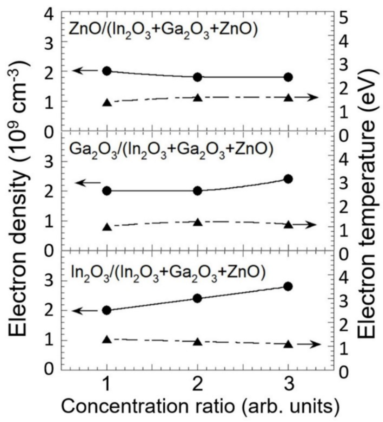

Fig. 2. Dependence of the electron density and temperature in the processing plasma on the powder target mixture

Figure 3 shows the surface morphology of the sputtered film using a 1:1:1 target mixture of In_2O_3, Ga_2O_3, and ZnO powders, as imaged with atomic force microscopy. The surface was smooth, with a root-mean-square roughness of less than 10 nm. The film deposition rate

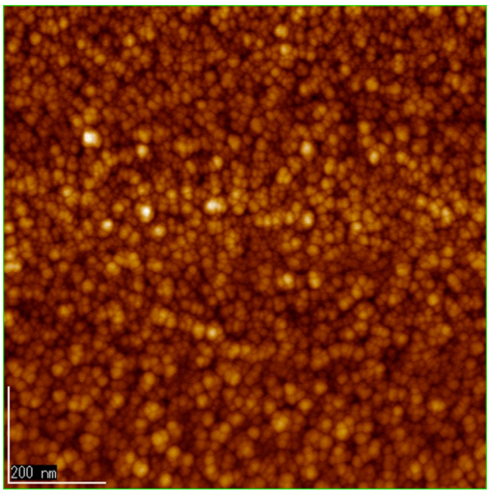

Fig. 3. Surface morphology of the sputtered film using a 1:1:1 target mixture of In_2O_3, Ga_2O_3, and ZnO powders

was approximately 5–15 nm/min. These results indicated that the deposition rate was almost independent of the composition of the powder target.

Figure 4 shows X-ray photoelectron spectra (XPS) of films prepared using a 1:1:1 mixed target. $In3d_{5/2}$, Ga2d, Zn3d, and O1s peaks verified that In-Ga-Zn-oxide thin films were prepared. Figure 5 plots narrow-band $In3d_{5/2}$ XPS spectra of films prepared with 1:1:1, 2:1:1, and 3:1:1 target mixtures of In_2O_3, Ga_2O_3, and ZnO powders, respectively. The $In3d_{5/2}$ signal increased accordingly with the increasing In_2O_3 ratio in the powder target.

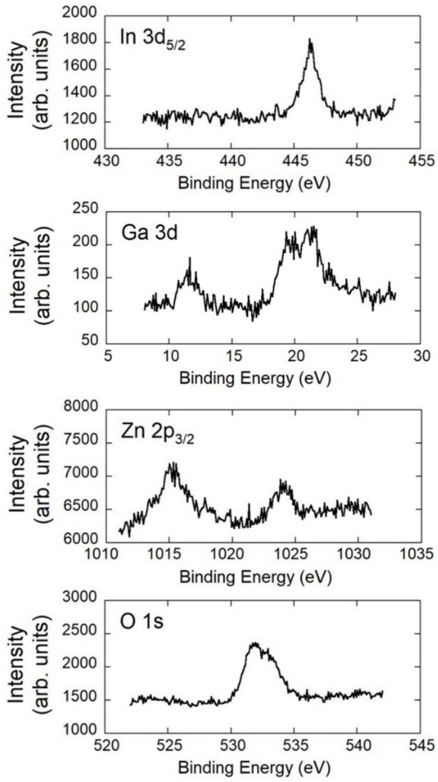

Fig. 4. X-ray photoelectron spectra (XPS) of films prepared using a 1:1:1 mixed target

Figure 6 shows the X-ray diffraction data of In-Ga-Zn-oxide films prepared using targets consisting of 1:1:1, 2:1:1, 1:2:1, 1:1:2 mixtures of In_2O_3, Ga_2O_3, and ZnO powders, respectively. The ZnO peak was observed in all the films; whereas the In and Ga peaks were not observed in the films prepared with the 1:1:1, 1:2:1, and 1:1:2 mixtures. The In peak was observed in the films prepared with the 2:1:1 mixed powder; however, Ga peaks never appeared in any of these films.

Figure 7 plots the optical transmittances of films prepared with 1:1:1, 2:1:1, and 3:1:1 mixed-powder targets. The transmittances were $> 75\%$ at 800 nm for all the target mixtures, and increased with increasing In_2O_3 in the powder target.

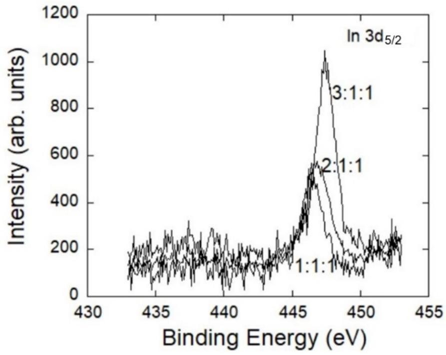

Fig. 5. Narrow-band $In3d_{5/2}$ XPS spectra of films prepared with 1:1:1, 2:1:1, 3:1:1 target mixtures of In_2O_3, Ga_2O_3, and ZnO powders

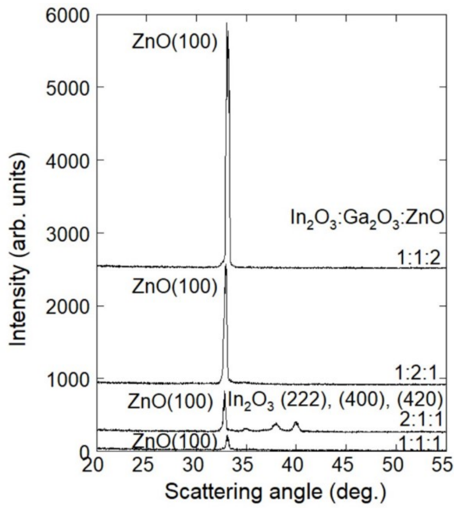

Fig. 6. X-ray diffraction data of In-Ga-Zn-oxide films prepared using targets consisting of 1:1:1, 2:1:1, 1:2:1, 1:1:2 mixtures of In_2O_3, Ga_2O_3, and ZnO powders

However, the crystallinity of the films changed with the amount of In_2O_3 in the powder mixture. As noted above, the In X-ray diffraction peak was not observed for the 1:1:1 mixture, and only appeared in the 2:1:1 mixture. Previously, for Sn-doped SiO_2 thin films prepared using targets containing Sn and SiO_2 powders [21–25], the Sn/SiO_2 ratio was difficult to control. This may have been because of the much lower melting temperature of Sn (504 K) compared with that of SiO_2 (1 923 K) [26–30]. Here, the melting points of In_2O_3 (2 183 K), Ga_2O_3 (2 173 K), and ZnO (2 248 K) were comparable; whereas, those of In and Zn are 430 K and 693 K, respectively. Therefore, In atoms in the plasma may affect the high-energy electrons and/or ions in the processing plasma, and the crystallinity could be changed by the powder mixture. The experimental results revealed that the processing plasma and elemental concentration ratio could be controlled

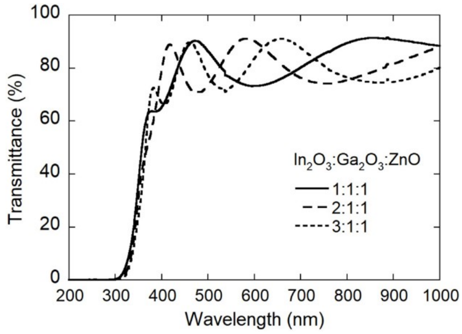

Fig. 7. Optical transmittances of films prepared with 1:1:1, 2:1:1, and 3:1:1 mixed-powder targets

by using metal-oxide powder targets. The controllability of these factors strongly depended on the properties of the powder materials used in the target, such as melting points.

4. Conclusions

In–Ga–Zn–O thin films were prepared via sputter deposition using targets consisting of mixed In_2O_3, Ga_2O_3, and ZnO powders. The processing plasma and elemental concentration ratios of the films were determined by the In_2O_3/Ga_2O_3/ZnO composition in the powder target. The properties of the powder, such as a melting point, strongly affected the ability to control the plasma parameters and the composition of the resulting thin films.

Acknowledgements

This study was supported in part by Grant-in-Aid for Scientific Research in Priority Areas (A) (No. 18H03848) and (C) (Nos. 23340181, 16K04999, 19K03045 and 20K03264), Kato Foundation for Promotion of Science; the Center for Low-temperature Plasma Science, Nagoya University; The Joint Usage/Research Center on Joining and Welding, Osaka University; Cooperation Promotion Center of Toyohashi University of Technology; Hatakeyama Culture Foundation; Research Institute for Applied Mechanics, Kyushu University. The authors wish to thank Prof. Shiratani and Prof. Higashida of Kyusyu University, Prof. Setsuhara of Osaka University, and Prof. Inoue and Dr. Nakamura of Toyohashi University for their helpful discussions.

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