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Properties of IGZO Film Deposited by Ar/O_2 Inductively Coupled Plasma Assisted DC Magnetron Sputtering

Chul Hee Lee[1], Tae Hyung Kim[1], Seung Min Lee[1], Jeong Wun Bae[1], Kyong Nam Kim[2], and Geun Young Yeom[1,3,*]

[1] *School of Advanced Materials Science and Engineering, Sungkyunkwan University, Suwon, Gyeonggi-do, 16419, South Korea*
[2] *School of Advanced Materials Science and Engineering, Daejeon University, Yongun-dong, Dong-gu, Daejeon 34520, Republic of Korea*
[3] *SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University, Suwon, Gyeonggi-do, 16419, South Korea*

ABSTRACT

In this study, the effects of inductively coupled plasma (ICP) power added to the DC magnetron sputtering of indium gallium zinc oxide (IGZO) using Ar/O_2 on the plasma characteristics and IGZO thin film characteristics were investigated. The addition of ICP power decreased the magnetron voltage and increased the magnetron current due to the increased plasma density near the magnetron surface. The addition of ICP also increased the deposition rate but also increased the surface roughness of the deposited IGZO thin films. When the electrical characteristics of deposited IGZO thin films were measured, the increase of ICP power to 300 W not only increased the carrier concentration from 1.87×10^{19} to 2.59×10^{19} cm^{-3} but also increased carrier mobility from 14 to 16.7 cm^2/Vs possibly due to the decreased defects (decreased defect scattering) in the film even with the increased impurity scattering caused by increased carrier concentration. The further increase of ICP power to 500 W slightly decreased the carrier mobility while slightly increasing the carrier concentration due to both the increased impurity scattering by the increased carrier concentration and the increased surface scattering caused by the increased surface roughness. The optical transmittance was not significantly varied with the ICP power and was higher than 80% at for the visible wavelength and the structure of IGZO deposited at room temperature remained amorphous even with the ICP power up to 500 W.

KEYWORDS: Plasma, IGZO, Sputter, Mobility.

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1. INTRODUCTION

Amorphous oxide semiconductor thin film transistors (TFTs) have been widely investigated for possible replacement of conventional amorphous silicon (a-Si) TFTs and poly-Si TFTs in active matrix liquid crystal displays (AM-LCDs) and active matrix organic light-emitting diodes (AM-OLEDs).[1–3] Conventional a-Si TFTs exhibit uniform electrical characteristics on eighth generation substrate (2.2×2.5 m^2), however, their field-effect mobility is as low as 0.5 cm^2/Vs. On the contrary, low temperature poly-crystalline silicon TFTs exhibit the field-effect mobility higher than 50 cm^2/Vs and stable electrical performance. However, there are still significant issues such as poor uniformity over a large area, high process temperature over 500 °C during silicon crystallization, etc. Amorphous oxide semiconductors such as Indium gallium zinc oxide (IGZO) have been investigated to have 20 to 50 times faster mobility compared to a-Si, and it also has good transparency at the visible wavelength. In addition, IGZO TFTs can be fabricated on flexible substrates such as plastic substrates by low temperature processes.

Until now, various semiconducting oxide materials besides IGZO have been reported as a channel layer for transparent and flexible TFTs. In these studies, the semiconducting oxide materials were deposited by various methods such as direct current/radio frequency (DC/RF) magnetron sputtering,[4–6] pulsed laser deposition,[7] and chemical solution processing.[8] Among these methods, DC/RF magnetron sputtering deposition methods exhibited high deposition rate and ease of scaling. However, the

*Author to whom correspondence should be addressed.
Email: gyyeom@skku.edu
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deposited thin film can be easily damaged by the impact of high energy ions in the plasma.[9,10] Furthermore, in the case of DC/RF sputtering, the generation of reactive species and sputtered atom flux cannot be separately controlled for stoichiometric semiconducting oxide deposition since both sputtered atoms and reactive species are generated in the localized discharge at the vicinity of target surface and these species are transported to the substrate from the discharge. In a previous research, by using inductively coupled plasma (ICP) in front of DC/RF sputtering, fluxes of sputtered atoms and reactive species to the substrate could be separately controlled, and which enabled precise control of reactive species to the substrate for stoichiometric semiconductor oxide deposition.[11] However, no detailed studies on the material characteristics by the addition of ICP during the DC/RF magnetron sputtering are reported.

In this study, to study the effect of ICP on material characteristics during the deposition of IGZO using ICP assisted DC magnetron sputtering more clearly, a spiral ICP coil was installed in front of IGZO magnetron target and the characteristics of IGZO film deposited as a function of ICP power were investigated in addition to the change of plasma characteristics.

2. EXPERIMENTAL DETAILS

Figure 1 shows the schematic diagram of the ICP assisted magnetron sputter system used in this study. As shown in Figure 1, 11.5 cm diameter and three-turn water-cooled copper ICP coil was installed in front of the 7.65 cm (3 inch) diameter magnetron. 13.56 MHz rf power was applied to the ICP coil while DC power was applied to the magnetron. As the magnetron sputter target, 7.65 cm diameter and 4 mm thick IGZO composed of In_2O_3: Ga_2O_3: ZnO = 1:1:1 was used.

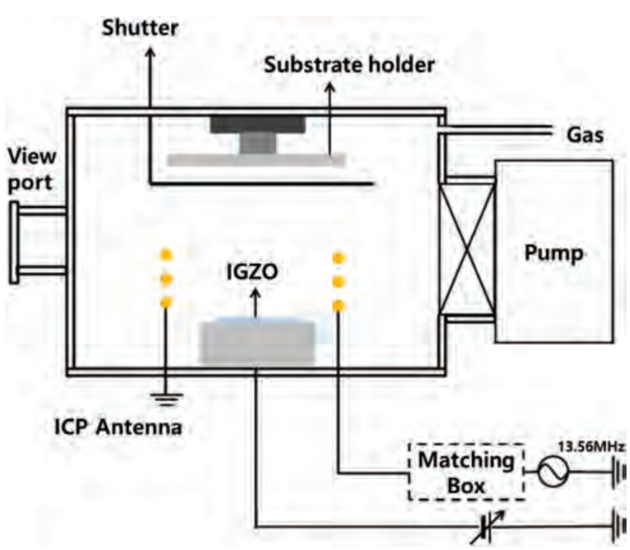

Fig. 1. The schematic diagram of ICP assisted magnetron sputter system used in this study.

The sputtering was carried out with the gas mixture composed of Ar (20 sccm)/O_2 (0.5 sccm) and at 5 mTorr of operating pressure. DC power was fixed at 200 W and the power to the ICP coil was varied from 0 to 500 W. The IGZO film was deposited on the glass substrate and, before the deposition, the magnetron target was pre-sputtered for 5 min. The deposition was carried out at room temperature and, for different deposition conditions, the deposition time was varied to maintain the deposited IGZO thickness to 100 nm.

The electrical characteristics of IGZO thin films were measured using a Hall effect probe (HMS-3000, Ecopia) and the surface roughness of the deposited film was investigated with Atomic force microscopy (AFM, Innova microscope, Bruker). The crystal structure of the deposited IGZO film was investigated using low angle X-ray diffraction (XRD, D8 Discover, Bruker), and the optical transmittance was measured using a Ultraviolet-visible spectrometer (UV-Vis Spectrometer, UV-3600, Shimadzu) for the wavelength range from 250 to 1000 nm. The chemical binding states of the deposited IGZO films were investigated using X-ray photoelectron spectroscopy (XPS, ESCA2000, VG Microtech Inc.).

3. RESULTS AND DICUSSION

To investigate the effect of ICP on the magnetron electrical characteristics during the sputter deposition of IGZO, the changes of voltage and current on the magnetron cathode during the operation of magnetron at the same DC power were measured as a function of ICP power and the results are shown in Figure 2. The ICP power was varied from 0 to 500 W while depositing IGZO film with the same DC power of 200 W, Ar (20 sccm)/O_2 (0.5 sccm) of gas mixture and 5 mTorr of operating pressure. As shown in Figure 2, the addition and increase of 13.56 MHz rf power to the ICP coil decreased the voltage from 465 V at no ICP power to 425 V at 500 W of ICP power while

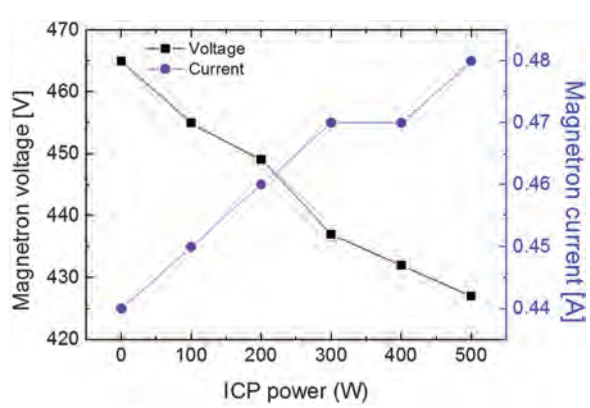

Fig. 2. Changes in the voltage and current on the magnetron cathode surface measured as a function of ICP power. The magnetron cathode was operated using 200 W of DC power, Ar (20 sccm)/O2 (0.5 sccm) of gas mixture, and 5 mTorr of operating pressure.

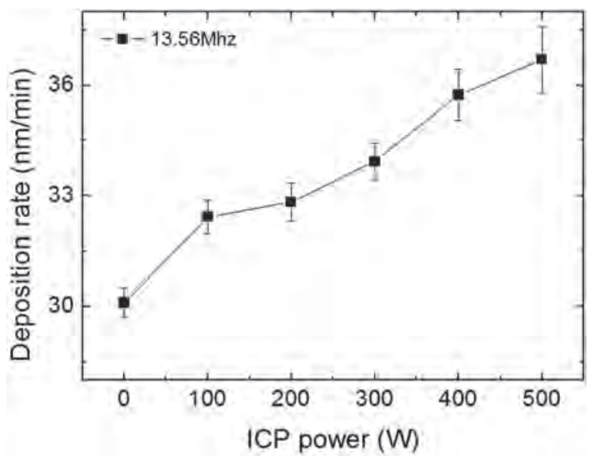

Fig. 3. Deposition rates of IGZO films as a function of ICP power while operating the magnetron cathode using 200 W of DC power, Ar (20 sccm)/O2 (0.5 sccm) of gas mixture and 5 mTorr of operating pressure.

increasing the current from 0.43 A to 0.48 A, respectively. The increase of current to the magnetron is related to the increase of ion density near the magnetron target surface by the addition of ICP power to the plasma. By the increase of ion current to the magnetron, the voltage on the magnetron was decreased to keep DC power = voltage × current at 200 W.

Figure 3 shows the deposition rate of IGZO measured as a function of power to the ICP coil while keeping the DC power to the magnetron at 200 W. The operating conditions were the same as those in Figure 2. As shown in Figure 3, the addition and increase of ICP power during the sputtering using the same DC power slightly increased the deposition rate from 30 nm/min at no ICP power to ~36 nm/min at 500 W of ICP power. The increase of sputter rate with the addition and increase of ICP power is due to the increased magnetron current, that is, due to the increased ion flux to the target by the increased ion density with the ICP in the plasma even though the ion energy to the target was decreased by the decreased cathode voltage at the constant DC power condition of the magnetron sputter source.

For the 100 nm thick IGZO film deposited for different ICP power conditions, the electrical characteristics of the deposited films were investigated using Hall effect measurement and the results are shown in Figure 4 for carrier concentration, mobility, and resistivity. The measured carrier type was *n*-type and the carrier concentration in the deposited IGZO film was increased with the increase of rf power up to about 300 W from 1.87×10^{19} to 2.59×10^{19} cm^{-3} and the further increase of rf power to 500 W increased the carrier concentration slightly. In the case of mobility, it also increased with the increase of ICP power from 14 to 16.7 cm^2/Vs until 300 W of ICP power and the further increase of ICP power to 500 W slightly decrease the mobility to about 16.2 cm^2/Vs. Therefore, the resistivity (ρ) which is related to the both

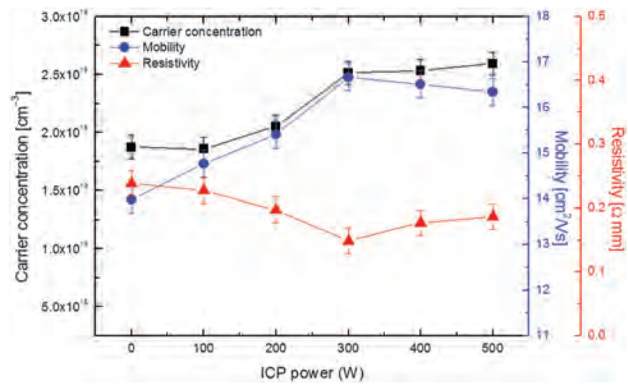

Fig. 4. Carrier concentration, mobility and resistivity of the IGZO thin films as a function of ICP power. The deposition conditions are the same as those in Figure 3. The thickness of IGZO was kept at 100 nm.

carrier concentration (n) and mobility (μ) of the carrier ($1/\rho = qn\mu$) was decreased from 2.38×10^{-2} $\Omega \cdot$cm at no ICP power to 1.422×10^{-2} $\Omega \cdot$ cm at 300 W of ICP power and the further increase of ICP power to 500 W slightly increased the resistivity to 1.82×10^{-2} $\Omega \cdot$cm.

One of the important factors affecting the carrier mobility of semiconductor materials is the crystallinity of the material. Using XRD, the possible change of crystallinity of the IGZO deposited as a function of ICP power was investigated and the results are shown in Figure 5. The deposition conditions are the same as those in Figure 3 and the thickness of the IGZO film was maintained at 100 nm. As shown in Figure 5, the IGZO deposited on glass substrate was amorphous structure and the addition and increase of ICP power up to 500 W did not change the crystal structure. A previous research by Takagi et al. also exhibited that the a-IGZO deposited at room temperature with magnetron sputtering maintains amorphous structure.[3]

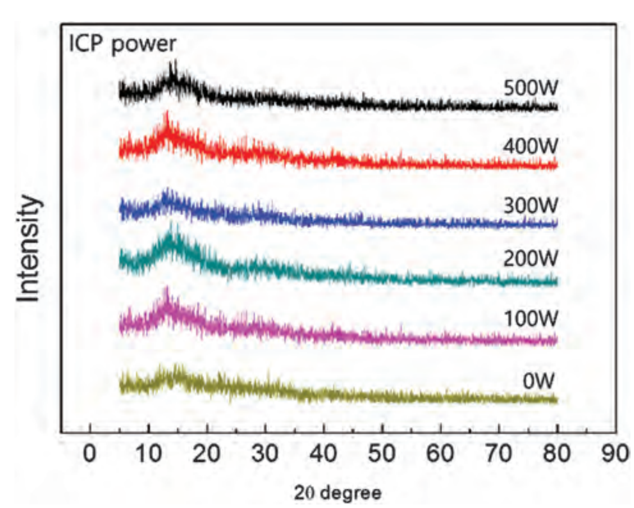

Fig. 5. X-ray diffraction intensity of the IGZO film measured as a function of ICP power. The deposition conditions are the same as those in Figure 3 and the thickness of the IGZO film was kept at 100 nm.

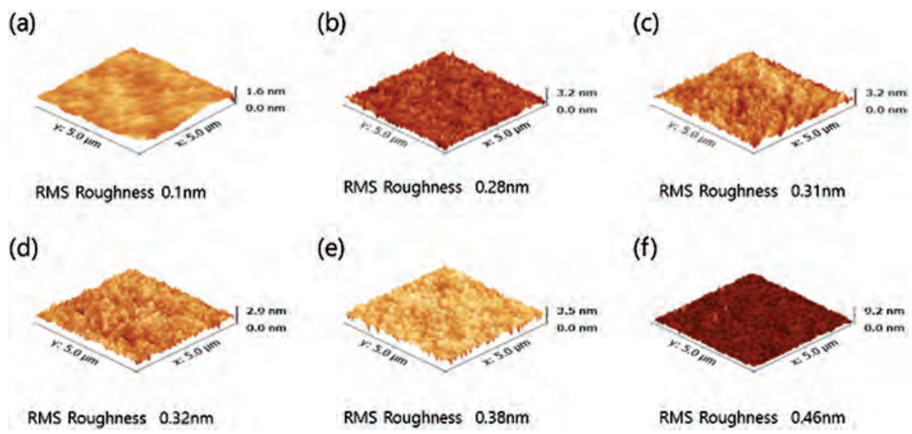

Fig. 6. AFM roughness images of the IGZO thin films as a function of ICP power (a) 0 W, (b) 100 W, (c) 200 W, (d) 300 W, (e) 400 W and (f) 500 W of ICP power. The deposition conditions are the same as those in Figure 3 and the thickness of the IGZO film was kept at 100 nm.

Surface roughness of the deposited IGZO films deposited as a function of ICP power was measured using AFM and the results are shown in Figure 6. The deposition conditions are the same as those in Figure 3 and the thickness of the IGZO film was also maintained at 100 nm. The scan size was 5 μm $\times$ 5 μm. The RMS surface roughness was increased with the addition and increase of ICP power from 0.1 nm at no ICP power to 0.46 nm at 500 W of ICP power. The increased surface roughness with the increase of ICP power could be partially due to the higher deposition rate at the higher ICP power or partially related to the nanocrystal line formation with the assist of ICP power even though we were not able to find the crystalline peak by XRD as shown in Figure 5.

Using XPS, atomic composition of IGZO deposited as a function of different ICP power from 0 to 500 W were investigated and the results are shown in Figure 7. As shown in the figure, even though we did not change the oxygen gas flow rate during the operation, the oxygen percentage in the deposited IGZO was decreased from 39.3% at no ICP power to 34.5% at 500 W of ICP power and In was also slightly decreased possibly due to the different sputter yields of oxides and the transport of materials to the substrate at different ICP power (need further investigation).

Using XPS, the chemical binding states of oxygen with metals were also investigated and Figure 8 shows the O1*s* narrow scan data of the deposited 100 nm thick IGZO film measured as a function of ICP power (0, 100, 200, 300, 400, and 500 W of ICP power for a, b, c, d, e, and f, respectively). The O1*s* peak could be deconvoluted into two different peaks at 530.5 eV (O1 peak) and 532 eV (O2 peak). The O1 peak at 530.5 eV is related to oxygen bonded to metals such as In, Ga, and Zn around O^{2-} ion[12–14] that is, related to the stoichiometric oxide while the O2 peak at 532 eV is related to the nonstoichiometric oxide which is caused by oxygen vacancy in the oxide.[15] As shown in Figure 8, the ratio of O2/(O1 + O2) was increased from 9.67% for no ICP power (17.76% at 100 W, 24.39% at 200 W, 30.99% at 300 W and 34.76% at 400 W) to 37.37% for 500 W ICP power which is caused by the decreased oxygen incorporation in the film with the increase of ICP power as shown in Figure 7. The increased oxygen vacancy in the IGZO thin film is known to be related to the increased carrier concentration,[16,17] therefore, the increased carrier concentration with the increase of ICP power shown in Figure 4 appears to be related to the increased oxygen vacancy in the film.

In general, the mobility of semiconductor materials is related to various scattering factors as follows;

$$\frac{1}{\mu} = \frac{1}{\mu_{\text{lattice}}} + \frac{1}{\mu_{\text{defect}}} + \frac{1}{\mu_{\text{impurity}}} + \cdots$$

where, μ_{lattice} is related to the phonon scattering (higher the temperature decrease the mobility), μ_{defect} is related to the scattering by various defects in the semiconductor materials such as lattice defect, surface defects, etc., and μ_{impurity} is related to the electrostatic scattering of electron

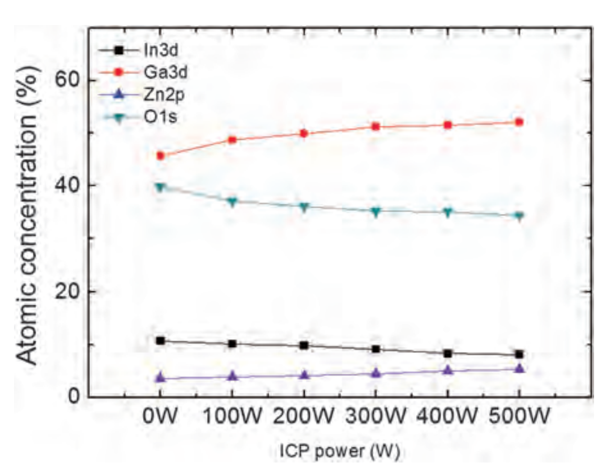

Fig. 7. Atomic composition of IGZO deposited with different ICP power. The deposition conditions are the same as those in Figure 3 and the thickness of the IGZO film was kept at 100 nm.

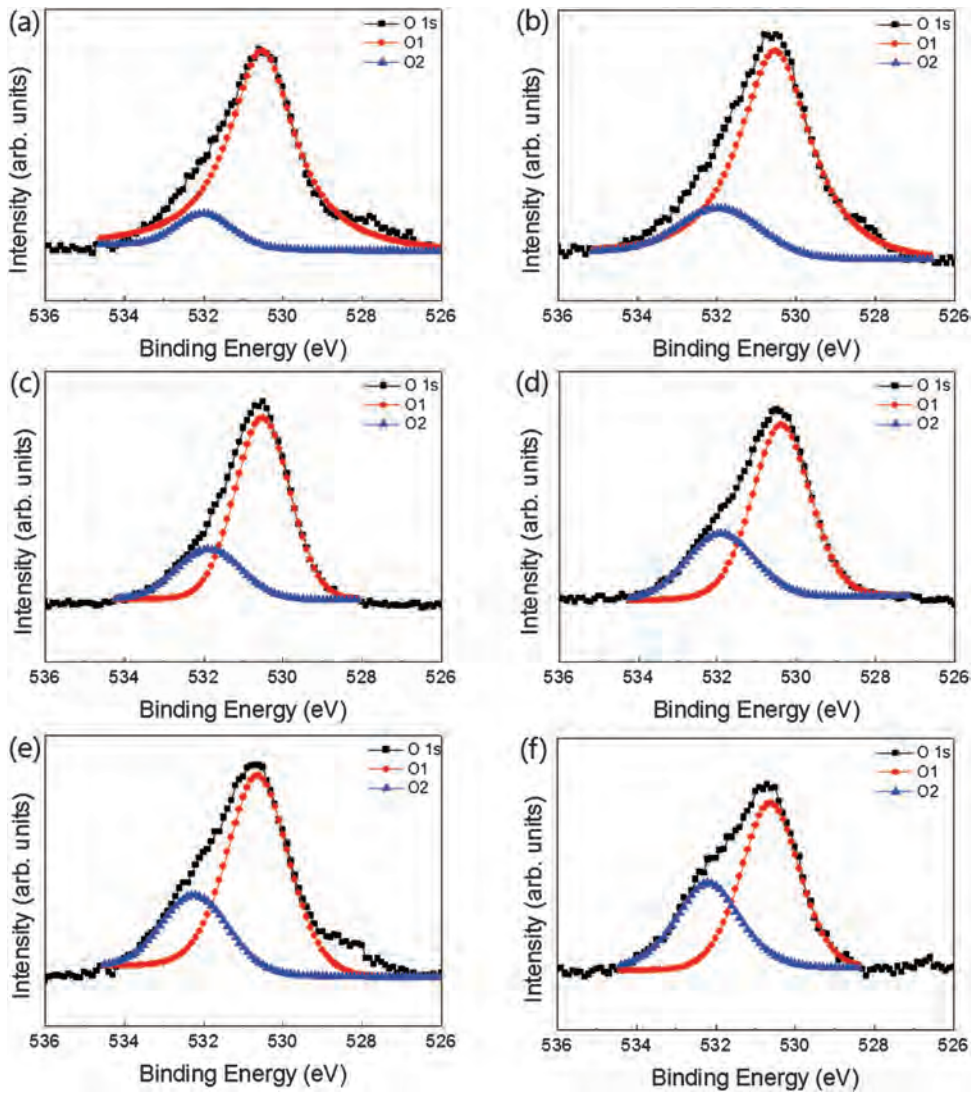

Fig. 8. XPS O1*s* narrow scan data of the IGZO films as a function of ICP power; (a) 0 W, (b) 100 W, (c) 200 W, (d) 300 W, (e) 400 W and (f) 500 W of ICP power. The deposition conditions are the same as those in Figure 3.

with the ionized impurities in the semiconductor materials which is related to doping. Therefore, in general, the increase of carrier concentration in the semiconductor material decreases carrier mobility. However, in Figure 4, the initial increase of carrier concentration was accompanied by the increased carrier mobility. The increase of both carrier concentration and carrier mobility of the IGZO film deposited with the increase of ICP power up to 300 W is believed to be related to the decreased defect formation ($1/\mu_{defect}$) in the IGZO film with the increase of ICP power compared to the decreased carrier mobility by the increased impurity scattering ($1/\mu_{impurity}$) because the IGZO film can be less damaged during the deposition at the higher ICP power due to the lower cathode voltage during the deposition and shorter exposure time to plasma for the deposition of the same thickness as shown in Figure 3. The decrease of carrier mobility after 300 W of ICP power in Figure 4 is believed to be related to the increased surface roughness, that is, by the increased surface scattering ($1/\mu_{defect}$) as shown in Figure 6 in addition to the increased carrier concentration, that is, by the increased impurity scattering ($1/\mu_{impurity}$).

Optical transmittance of the IGZO films deposited as a function of ICP power was measured using a UV-Vis spectrometer and the results are shown in Figure 9(a). The operating conditions were the same as those in Figure 3 and the thickness was maintained at 100 nm. As shown in Figure 9(a), the optical transmittance of the deposited film at 550 nm was higher than 80%. Figure 9(b) shows the optical bandgap energy estimated by Tauc relation with the optical transmittance measured using the UV-Vis spectrometer.[18] The optical bandgap energy was increased from 3.58 eV at no ICP power to 3.65 eV at 500 W of ICP power possibly due to the Burstein moss effect which is related to the electron filling of conduction band by the increased carrier concentration.[19, 20]

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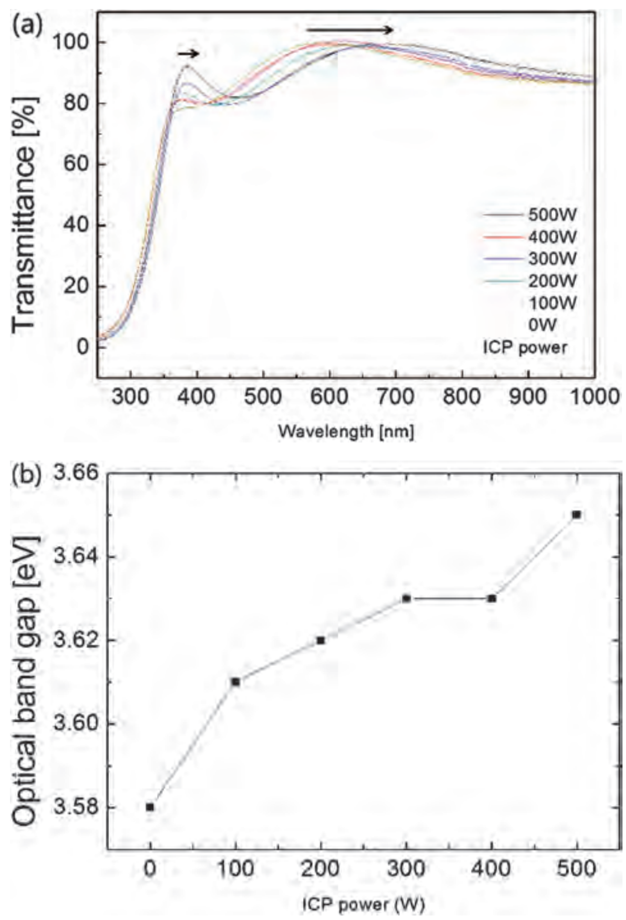

Fig. 9. (a) Optical transmittance and (b) optical band gap of IGZO thin film as a function of ICP power. The operating conditions are the same as those in Figure 3.

4. CONCLUSION

In this study, by installing a spiral ICP coil in front of IGZO magnetron target, the effect of ICP power on the plasma characteristics and thin film characteristics during the deposition of IGZO using ICP assisted DC magnetron sputtering was investigated. The addition and increase of ICP power during the IGZO sputtering at constant DC power of 200 W decreased magnetron cathode voltage while increasing the magnetron current, that is, increasing the deposition rate due to the increased plasma density with the increase of ICP power. The IGZO deposited with the increase of ICP power up to 500 W exhibited the increased carrier concentration but also exhibited increased surface roughness. The increase of carrier concentration with the increase of ICP power was related to the increased oxygen vacancy in the deposited film. Up to 300 W of ICP power, both the carrier concentration (from 1.87×10^{19} to 2.59×10^{19} cm^{-3}) and the carrier mobility (from 14 to 16.7 cm^2/Vs) of the deposited IGZO thin film were increased with the increase of ICP power possibly due to the decreased defects in the deposited film even with the increase of carrier concentration (impurity scattering). The further increase of ICP power to 500 W decreased the carrier mobility to due to both the increased carrier concentration (impurity scattering) and increased surface roughness (surface scattering).

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References and Notes

1. K. Nomura, H. Ohta, A. Takagi, T. Kamiya, M. Hirano, and H. Hosono, *Nature* 432, 488 (**2004**).
2. H. Chiang, J. Wager, R. Hoffman, J. Jeong, and D. A. Keszler, *Appl. Phys. Lett.* 86, 013503 (**2005**).
3. A. Takagi, K. Nomura, H. Ohta, H. Yanagi, T. Kamiya, M. Hirano, and H. Hosono, *Thin Solid Films* 486, 38 (**2005**).
4. H. Yabuta, M. Sano, K. Abe, T. Aiba, T. Den, H. Kumomi, K. Nomura, T. Kamiya, and H. Hosono, *Appl. Phys. Lett.* 89, 2123 (**2006**).
5. J. K. Jeong, J. H. Jeong, H. W. Yang, J. Park, Y. Mo, and H. D. Kim, *Appl. Phys. Lett.* 91, 3505 (**2007**).
6. Y. Moon, S. Lee, D. Kim, D. Lee, C. Jeong, and J. Park, *Japanese Journal of Applied Physics* 48, 031301 (**2009**).
7. A. Suresh, P. Wellenius, A. Dhawan, and J. Muth, *Appl. Phys. Lett.* 90, 123512 (**2007**).
8. K. Banger, Y. Yamashita, K. Mori, R. Peterson, T. Leedham, J. Rickard, and H. Sirringhaus, *Nat. Mater.* 10, 45 (**2011**).
9. D. K. Kim, J. Lee, D. Kim, K. Shin, M. Kim, S. Choi, and C. Kang, *Thin Solid Films* 519, 6645 (**2011**).
10. T. Wang, X. Diao, and X. Wang, *Appl. Surf. Sci.* 257, 9773 (**2011**).
11. K. Takenaka, K. Cho, Y. Ohchi, H. Otani, G. Uchida, and Y. Setsuhara, *Japanese Journal of Applied Physics* 54, 06GC02 (**2015**).
12. L. K. Rao and V. Vinni, *Appl. Phys. Lett.* 63, 608 (**1993**).
13. R. Cebulla, R. Wendt, and K. Ellmer, *J. Appl. Phys.* 83, 1087 (**1998**).
14. M. Chen, Z. Pei, C. Sun, L. Wen, X. Wang, and J. Cryst. *Growth* 220, 254 (**2000**).
15. J. C. Fan and J. B. Goodenough, *J. Appl. Phys.* 48, 3524 (**1977**).
16. P. Carcia, R. McLean, M. Reilly, and G. Nunes, Jr., *Appl. Phys. Lett.* 82, 1117 (**2003**).
17. R. Martins, P. Barquinha, I. Ferreira, L. Pereira, G. Goncalves, and E. Fortunato, *J. Appl. Phys.* 101, 044505 (**2007**).
18. T. Tansley and C. Foley, *J. Appl. Phys.* 59, 3241 (**1986**).
19. E. Burstein, *Physical Review* 93, 623 (**1954**).
20. T. Moss, *Proceedings of the Physical Society. Section B* 67, 775 (**1954**).