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Electrical, Optical and Structure Properties of ITO Films Cosputtered with ZnO

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Abstract--The relationship between electrical, optical and material properties of the transparent and conductive films prepared by cosputtered ITO and ZnO targets has been investigated. The lowest resistivity and smoothest surface roughness at specific Zn impurity controlled by the cosputtered ZnO target are attributed to the formation of amorphous-like $Zn_kIn_2O_{3+k}$ ternary compound. As the cosputtered films with an atomic ratio of Zn / (Zn + In) is 0.60, a ZnO (001) phase with microcrystalline is observed and results in the degradations both on the resistivity and surface roughness. Furthermore, the optical bandgap calculates from the absorption edge of these cosputtered films is narrowed with the increase of Zn impurity that is controlled by a higher rf power supplied to the ZnO target.

1. Introduction

Transparent and conductive oxide (TCO) films have been widely used for optoelectronic devices [1-2]. A high optical transmittance in the visible region and low resistivity is the key requirements applied in these devices. Indium-tin oxide (ITO) prepared by the sputtering technology is the most commonly transparent electrodes in flat panel displays, such as LCD, PDP and OLED. To date, a new beginning for displaying pictures on flexible substrate is expanded. Since the glass transition temperatures of the flexible substrates are extreme low (70 ~ 150 ^{o}C), the high performance ITO films with post-annealing treatments are limited [3-4]. New materials prepared by a sputtering method, such as IZO and AZO, are promising to successfully deposit on flexible substrates [5-6]. In order to maintain the low resistivity properties and extend the additional applications on display devices, we prepared a transparent and conductive film by an rf magnetron cosputtering system at room temperature with ITO and ZnO targets [7]. The resistivity of these cosputtered films was not only comparable to the IZO films prepared by an In_2O_3-ZnO mixture target but achieved an optical film with multi-functions by using multilayer structures.

In this paper, the evolutions of the electrical and optical properties prepared by a cosputtering system with ITO and ZnO targets are investigation by X-ray diffraction (XRD). The microstructures conducted from XRD measurements of these cosputtered films are demonstrated from atomic force microscopy (AFM). Moreover, the tunable energy bandgap achieved from various ZnO impurities in the ITO film during cosputtering are also discussed

2. Experimental procedure

The configuration of an rf magnetron cosputtering system is shown in Fig. 1. The targets of ITO and ZnO materials with diameters of 50 mm were placed on the cosputtering gun. A dual rf power supply with synchronized phase was employed to control the rf power supplied to the ITO target and to vary the rf power supplied to the ZnO target. Detailed descriptions for the cosputtering system were reported elsewhere [7]. Electrical resistivity, carrier concentration, and Hall mobility were measured by the van der Pauw method. The crystalline of these cosputtered films was examined by X-ray diffraction (XRD). The surface morphologies were observed from an atomic force microscopy (AFM, DI3100). Optical properties were measured using an UV-Vis-NIR spectrophotometer over a wavelength range from 300 to 800 nm.

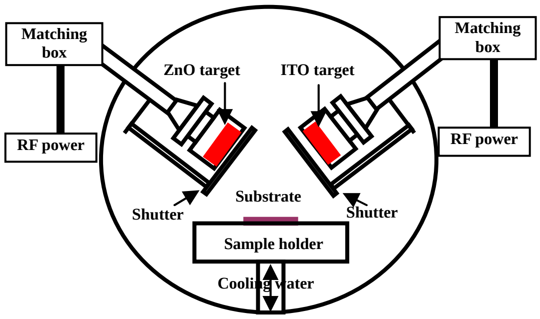

Fig. 1 Configuration of rf magnetron co-sputtering system.

3. Experimental results

Figure 2 showed the resistivity, carrier concentration and Hall mobility of the individual ITO film at an rf power of 150 W and the cosputtered films at rf power of 70, 165 and 250 W, respectively. The doped atomic concentrations [Zn / (Zn + In)] of these cosputtered films were 0, 0.34, 0.60 and 0.71 evaluated from the deposited rates of the individual ZnO and ITO films. The resistivity was obvious reduce with an additional ZnO impurities and reached to a lowest resistivity of 3.0×10^{-4} Ω when the rf cosputtered power was 70 W. As an rf cosputtered power reached 250 W, an obvious degradation on resistivity was found. In theory, there was an inverse relationship between carrier

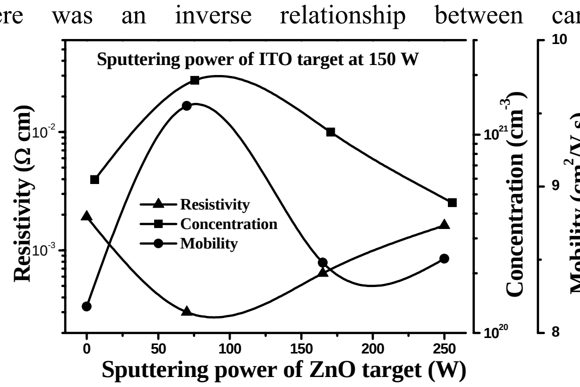

Fig. 2 Resistivity, carrier concentration and Hall mobility as functions of rf power at ZnO target while rf power of ITO target is 150 W.

concentration and Hall mobility for an ITO film at a specific temperature. However, the carrier concentration shown in Fig. 2 was proportion to Hall mobility as the additional ZnO rf cosputtered powers at 70 and 160 W except for the cosputtered rf power at 250 W. Therefore, these cosputtered films must experience the microstructure changes at various ZnO doped in the ITO films by cosputtering.

Figure 3 showed the XRD measurements of the individual ITO film and cosputtered films described in Fig.2. The crystalline structure of individual ITO films was poly-crystalline with obvious peak of (222) and (400).

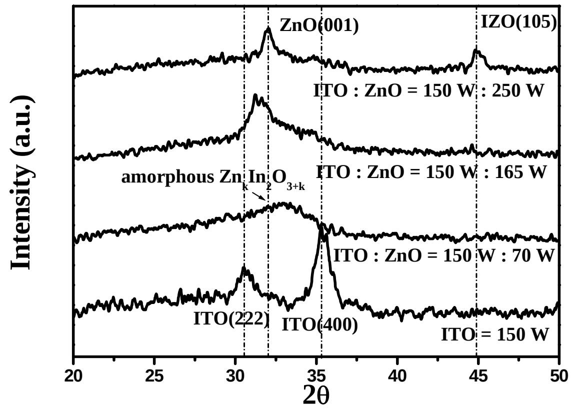

Fig. 3 X-ray diffraction pattern as functions of rf power at ZnO target while rf power of ITO target is 150 W.

However, the crystallinity of cosputtered films with ITO and ZnO targets at 150 and 70 W was amorphous. This amorphous structure was identified as amorphous-like $Zn_kIn_2O_{3+k}$ compound (k was integer) [8]. The carrier concentration and Hall mobility shown in Fig. 2 were markedly increase with the formation of $Zn_kIn_2O_{3+k}$ compound. However, as the rf cosputtered power of ZnO target reached 165 W, an additional microcrystalline phase with ZnO (001) superposed on the amorphous-like $Zn_kIn_2O_{3+k}$ phase. This indicated excess ZnO impurities were appeared in the cosputtered film. The excess Zn^{2+} ions were considered to suppress the carrier concentration of an ITO film [9]. Hall mobility was also reduced from the appearance of the microcrystallinity. At an rf cosputtered power of 250 W, a higher Hall mobility compared to that of 165 W was obtained attributed to the crystalline growth with narrow FWHM show in Fig. 3. The evidences of the amorphous structure and crystalline growth at various rf cosputtered powers were also observed from surface roughness observations. The amorphous-like $Zn_kIn_2O_{3+k}$ at an rf cosputtered power of 70 W had a smoothest surface roughness of 1.18nm while the cosputtered films with an additional phase of ZnO (001) presented a rough surface roughness higher than 3.0 nm.

The optical properties of these cosputtered films were shown in Fig. 4. The average optical transmittance obtained from these cosputtered films in the visible spectrum was over 80 %. Furthermore, the related optical bandgap (E_g) was estimated by the absorption coefficient against photon energy (inserted figure in Fig. 4). The optical bandgap was found to decrease with increasing rf cosputtered power of ZnO target from 3.49 eV (70 W) to 3.38 eV (250 W)..

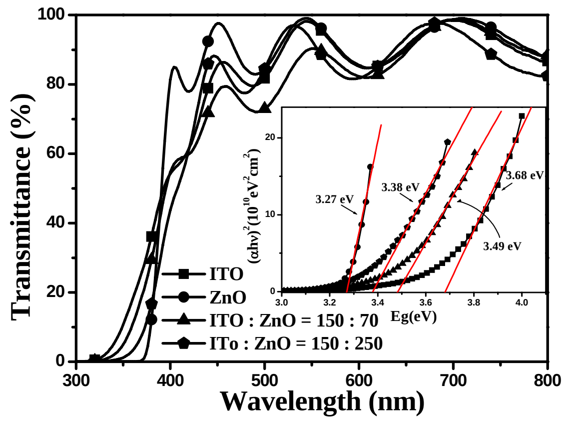

Fig. 4 Optical transmittance as functions of rf power at ZnO target while rf power of ITO target is 150 W (inserted figure is optical bandgap).

4. Conclusions

The XRD measurements indicated that an amorphous-like $Zn_kIn_2O_{3+k}$ ternary compound formed from the ITO cosputtered with ZnO was responsible for the superior carrier concentration and Hall mobility to the individual ITO films. A smooth surface roughness was also obtained form the amorphous structure. The degradation mechanisms were attributed to excess ZnO (ZnO (001)) impurities appeared during cosputtering and also increased the film resistivity. At a cosputtered power of 250 W, the grain size of the ZnO (001) phase was found to increase and resulted in an improved Hall mobility. The surface roughness of these cosputtered films with microcrystalline ZnO (100) phase was inferior to the amorphous one. Moreover, the optical bandgap decreased from 3.49 eV to 3.38 eV with increasing the rf cosputtered powers supplied to the ZnO target.

Acknowledgments

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References

[1]Y. H. Tak, K. B. Kim, H. G. Park, K. H. Lee and J. R. Lee, Thin Solid Films. **411** (2002) 12.

[2]J. Pla, M. Tamasi, R. Rizzoli, M. Losurdo, E. Centurioni, C. Summonte and F. Rubinelli,, Thin Solid Films **425** (2003) 185.

[3]B. T .Tang, Q. X. Yu, H. Y. Lee and C. T. Lee: Mater. Sci. Eng. B **82** (2001) 259.

[4]L.R. Cruza, C. Legnani, I.G. Matoso, C.L. Ferreira and H.R. Moutinho, Mater. Res. Bull. **39** (2004) 993.

[5]H. M. Kim, S. K. Jung, J. S. Ahn, Y. J. Kang and K. C. Je, Jpn, Appl. Phys. **42** (2003) 223.

[6]H. Kim, C. M. Gilmore, J. S. Horwitz, A. Pique, H. Murata, G. P. Kushto, R. Schlaf, Z. H. Kafafi and D. B. Chrisey, Appl. Phys. Lett. **76** (2002) 259.

[7]D. S. Liu, C. C. Wu and C. C. Lee, accepted by Jpn. Appl. Phys. (2005).

[8]N. Naghavi, C. Marcel, L. Dupont, A. Rougier, J. B. Leriche and C. Guery, J. Mater. Chem. **10** (2000), 2315.

[9]T. Minami, T. Yamamoto, Y. Toda and T. Miyata, Thin Solid Films **373** (2000) 189.