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Influence of annealing temperature on the morphology and crystal structure of Ga-doped ZnO thin films

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Abstract. Ga-doped ZnO (ZnO:Ga) thin films have been deposited on Corning glass substrates by handmade dc magnetron sputtering. The pressure and deposition time respectively were set on 500 mTorr and 60 minutes. The deposition temperature was fixed at 300ºC with 30 watts of plasma power. The deposited ZnO:Ga thin films were heated at 300 ºC, 350 ºC, and 400 ºC, respectively. The morphology and crystallinity of ZnO:Ga thin films have been observed with SEM and XRD. The observation with SEM shows that the film morphology is denser and the grain size is smaller when the temperature is increased. The crystallinity of the film increases as the annealing temperature is enhanced from 300 ºC to 350 ºC. However, the crystallinity of the ZnO:Ga films decreased when the annealing temperature is 400 ºC.

1. Introduction

Transparent conductive oxides (TCO) have been widely used as optoelectronic devices such as light emitting diodes, flat-panel displays, and solar cells [1]. Indium tin oxides (ITO) have been applied for TCO because the materials have good optical transmittance characteristics, wide band gap, and high conductivity [2]. Nevertheless, ITO is considered an expensive material and poses low electrical stability [3]. Alternatively, ZnO is proposed as a substitute material for ITO because it is cheap, non-toxic, stable, and has a wide band gap [4]. But it has a weakness in term of structural and electrical properties stability [5]. To improve its structure, some researchers have grown ZnO thin films by addition of doping materials [1,6,7]. Among all doping elements, gallium (Ga) is considered as one of the most suitable for ZnO material [6,7]. Some researchers also reported that the performances of ZnO thin film could be enhanced by performing of appropriate post-deposition treatment [8,9].

Several methods have been used to growth ZnO:Ga thin films, such as metal oxide chemical vapor deposition (MOCVD) system [1], atmospheric-pressure chemical vapor deposition (APCVD) [8], pulsed laser deposition (PLD) [9], sol-gel [10,11], RF magnetron sputtering [12] and dc magnetron sputtering [6]. In the previous study, the influence of Ga-doped concentrations on the structural and optical properties of ZnO thin films was deeply investigated. We obtained that the crystallinity of ZnO film significantly enhance while the band gap is slightly decreased by the increase of Ga-doped at low Ga concentration (1 % to 2%). Continuing our previous work, in this study, the improvement of structural ZnO:Ga thin film was carried out by conducting the annealing process as post-deposition

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treatment. The structural of a film was studied by X-Ray Diffraction spectroscopy (XRD) while the morphology of film was observed by Scanning Electron Microscopy (SEM). All the ZnO:Ga thin films were deposited on the corning glass substrates by using a handmade dc-magnetron sputtering system.

2. Methods

ZnO:Ga thin films were deposited on Corning glass substrates by using handmade dc-magnetron sputtering system [6]. The sputtering target was fabricated using high-purity Ga_2O_3 (99,999%) and ZnO (99,999%) powders. The Ga content in the target is 2% (wt). The target has about 25 mm diameter and 5 mm thickness. High purity (99,999%) Ar gas was used as sputtering gas. The ZnO:Ga thin films were prepared with plasma power, pressures of a chamber, temperature deposition, and deposition time are 30 W, 500 mtorr, 300 ^{o}C, 60 minutes, respectively. After deposition, the samples were annealed at the temperature of 300 ^{o}C, 350 ^{o}C and 400 ^{o}C for 20 minutes. SEM and XRD were used to investigate the effect of annealing temperature on the morphology and structure of the ZnO:Ga samples, respectively.

3. Result and Discussion

Effect of annealing temperature on the ZnO:Ga thin films morphology was studied by using SEM micrographs. Figure 1 shows that the surface morphology of ZnO:Ga thin films get denser with smaller grain sizes when the annealing temperature is increased from 300 ^{o}C to 400 ^{o}C.

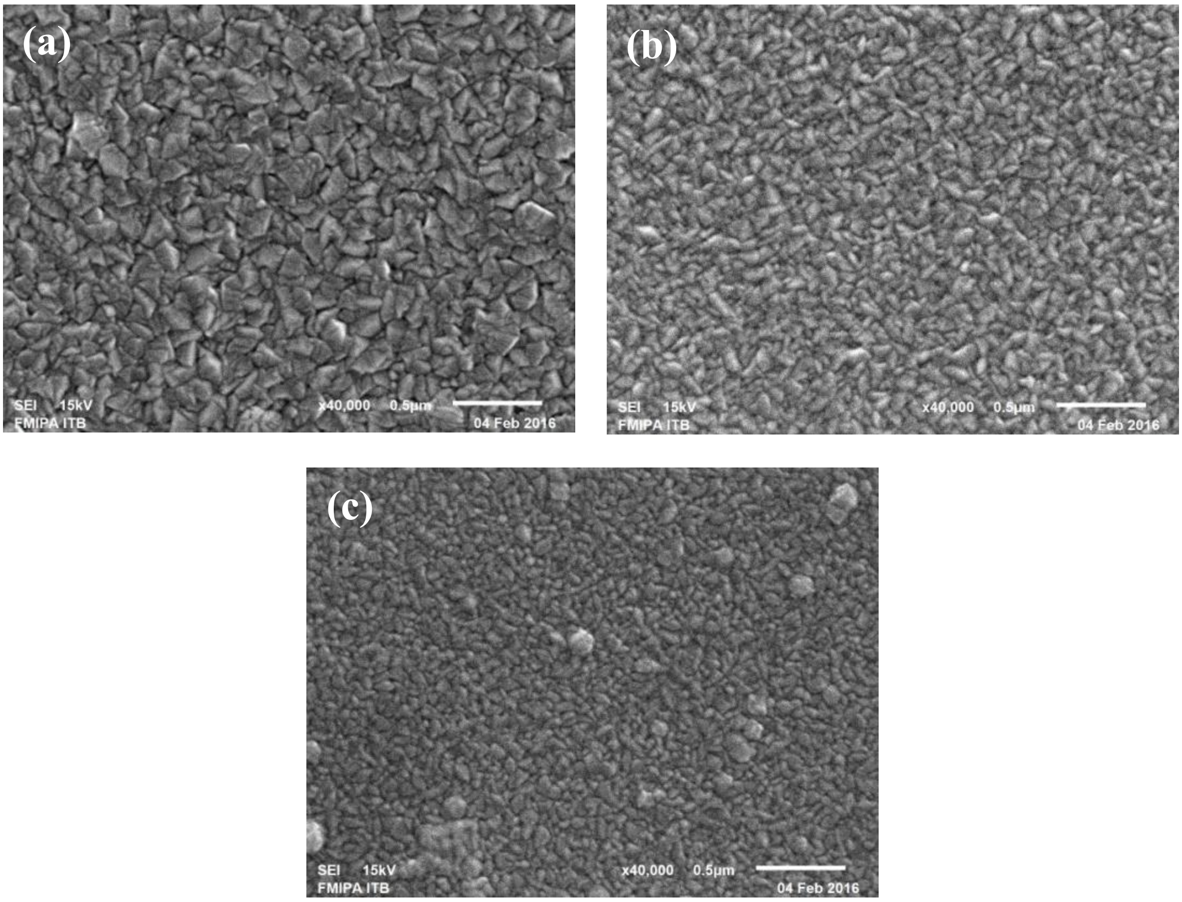

Figure 1. SEM images of ZnO:Ga thin films with different annealing temperatures: (a) 300 ^{o}C, (b) 350 ^{o}C, and (c) 400 ^{o}C.

It is clearly shown that the annealing temperature has a great influence on ZnO:Ga film surface structures, which surface roughness decreased when the annealing temperature was increased. The surface morphology of the ZnO:Ga film is denser when the annealing temperature is increased from 300 ºC to 400 ºC. It can be seen that, by increasing the annealing temperatures, the roughness of the films is reduced. Decreasing the surface roughness of ZnO:Ga thin films due to annealing treatment was probably caused by the relaxation of defects [3]. It is possible that some oxygen atoms from ZnO:Ga are released when the annealing temperature is enhanced so that the grain size of the films decreases.

XRD pattern of ZnO:Ga thin films deposited on Corning glass substrate with different annealing temperature is shown in figure 2. All the films exhibit a strong (002) peak, and a weak (101) peaks as previously reported [6]. The ZnO:Ga films are polycrystalline with peak oriented along the (002) plane. The same diffraction peak (002) shows that the films mostly grow along the c axis [12]. It indicated that the grains of the films were strongly oriented along the *c*-axis of the wurtzite structure [13]. The diffraction patterns show that increasing of annealing temperature influence the diffraction intensity. When the temperature annealing increased from 300 ºC to 350 ºC, the intensity of the (002) peak is increased, but the (001) peak intensity is decreased. The decrease of (002) peak intensity at 400 ºC annealing temperature indicated the occurring structural degradation of the ZnO thin films. This is showed that over annealing temperature leads to the decreasing of crystallinity.

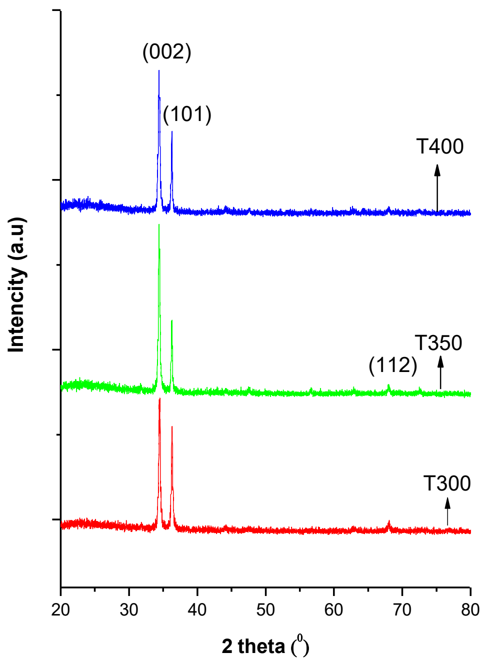

Figure 2. XRD pattern of ZnO:Ga thin films with different annealing temperatures.

Table 1 shows the structural parameters of ZnO:Ga thin films. The crystallite size of ZnO:Ga films have been determined from the Full Width at Half Maximum (FWHM) of diffraction peak using the Scherrer formula. It can be showed that the annealing temperature has a significant contribution to the change of the crystal size of the deposited films. The crystal size increases from 59.40 nm up to 66.56 nm, while the annealing temperature increase from 300ºC to 350ºC. However, the crystal size decreases from 66.56 nm to 57.38 nm, while the annealing temperature increased from 350ºC to

400ºC. The lowest FWHM is obtained at 350ºC annealing temperature. This shows that this condition is the optimum annealing temperature.

Table 1. Structure parameters of ZnO:Ga thin films with different annealing temperature

Annealing temperature (˚C)	2θ (º)	FWHM (º)	*d* spacing (Å)	*c* lattice (Å)	crystal size (nm)	*a* lattice (Å)
300	34.44	0.28	2.600	5.200	59.40	7.654
350	34.40	0.25	2.601	5.202	66.56	7.663
400	34.41	0.29	2.604	5.208	57.38	7.661

4. Conclusion

The influence of annealing temperature on the morphology and crystalline structure of ZnO:Ga thin films deposited by handmade dc-sputtering method were analyzed using SEM micrograph and XRD diffractometer. The annealing temperature has a great influence on ZnO:Ga film surface morphology. The observations with SEM show that the film morphology is denser and the grain size is smaller when the annealing temperature is increased. The crystallinity of the film increases as the annealing temperature is enhanced from 300 ºC to 350 ºC. However, the crystallinity of the ZnO:Ga thin films decreased when the annealing temperature at 400 ºC.

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