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Structural and Optical Properties of Zinc Oxide (ZnO) based Thin Films Deposited by Sol-Gel Spin Coating Method

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Abstract. The structural and optical properties of ZnO thin films deposited by sol-gel spin coating method have been investigated. The ZnO thin films were deposited on corning glass 7059 substrate from zinc acetate (ZnAc), ethanol, and stearic acid as starting material, solvent and stabilizer were used, respectively. The characteristics of the film were analysed from transmittance spectra obtained by UV-Vis spectroscopy measurement, while the atomic structure of the films analysed from XRD and AFM measurement. The results showed that the characteristics of ZnO thin films were strongly depend on annealing temperature. The XRD measurement indicated that deposited film shows a type of nanocrystalline peak broadening with hexagonal structure and preferential orientation along (002) crystal plane. The transmittance value of the ZnO films was relatively high (75 - 80 %). The optical band gap of ZnO films decreases with increasing in annealing temperature. AFM images showed the annealing temperature effects surface morphology of the films.

1. Introduction

Zinc oxide (ZnO) has attracted much attention as one of the most promising material for electronic applications. The interest comes from the wide direct bandgap (3.37 eV) and a large binding energy (60 meV) of this material. With such characteristics, it makes ZnO become a potential useful in various optoelectronic applications such as optical sensors and light emitter, solar cells, etc. [1]. One area of great interest is the application of ZnO as a transparent conducting oxide (TCO) for solar cells applications replacing traditional indium tin oxide [2].

ZnO thin films have been prepared by various techniques (vacuum and non vacuum) such as sputtering [3], chemical vapor deposition (CVD) [4], molecular beam epitaxy (MBE) [5], pulse laser deposition (PLD) [6], spray pyrolysis [7], and sol-gel process [8,9]. Among these methods, sol-gel process using spin coating techniques is particularly attractive because its simplicity, flexibility, easy to dope, offers the possibility or preparing large area coating, and acceptable cost [10]. However, the crystalline quality of ZnO thin films might be inferior [1,2]. Several significant challenges remain, including difficulty in preparation of stable dispersion, poor uniformity in coating process, and poor conductivity of ZnO thin films.

In this study, we investigate the structural and optical properties of ZnO thin films prepared by solgel spin coating techniques. The effect of annealing temperature on the optical transmittance, optical

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absorption, and optical bandgap would be discussed. The surface morphology and roughness was also studied due to annealing temperature variation.

2. Experimental Procedure

ZnO thin films were deposited on corning glass 7059 substrate by sol-gel spin coating process. Zinc Acetate powder, ethanol, and stearic acid were used as starting material, solvent, and stabilizer, respectively. The ZnAc solution was stirred and heated to 80 $^{\circ}$ C for 1 hour. The corning glass substrate was cleaned in methanol and de-ionized water for 10 minutes each and dried. Coating solution was drop onto glass substrate and spun at 2500 rpm for 20 seconds by using home-made spin coater. After coating process, the films were dried at 200 $^{\circ}$ C for 15 minutes in a furnace in order to evaporate the solvent and remove organic residuals. The ZnO films were then annealed at temperature of 300 – 700 $^{\circ}$ C for 1 hour. The preparation procedure of ZnO thin films deposition is shown in Figure 1.



Figure 1. Flow chart for ZnO thin films deposition procedure

The optical characteristics of ZnO thin films were analyzed from ultraviolet-visible (UV-Vis) spectrometer measurement. The optical band gap was determined using Touch's plot method hv versus $(\alpha hv)^{1/2}$, where α and hv denote the optical absorption coefficient and photon energy, respectively [11]. The optical absorption coefficient (α) were calculated from transmittance spectra obtained by UV-Vis measurement using the relation: $\alpha = -\frac{1}{d} lnT(\lambda)$, where d is the thickness and T

is optical transmittance of the films. The structure and surface morphology of films were analyzed from X-ray diffraction (XRD) and Atomic Force Microscopy (AFM) measurement.

3. Results and Discussion

Figure 2 showed the XRD patterns of ZnAc, ZnO powder, and ZnO thin film annealed at 500 ^oC for 15 minutes. The ZnO samples were corresponded to hexagonal structure. Although, the intensity of diffraction peaks for both ZnO samples were different. The difference in intensity of diffraction peak might be indicated the difference in crystal size. Miller Indices of diffraction peaks of ZnO samples are shown in Table 1.



Figure 2. The XRD patterns of ZnAc powder, ZnO powder, and ZnO thin film

In order to understand the crystalline ZnO particles, the grain size of ZnO particles has been estimated from the FWHM of (002) the diffraction peak using the Scherrer formula [12]:

$$d = \frac{0.9\lambda}{\beta \cos\theta}$$

Where λ is the x-ray wavelength of 1.54 Å, θ is the Bragg diffraction angle of (002) peak, and β is the full-width at half-maximum (FWHM) of θ (002). We found that the grain size of ZnO particle is about 23.8 nm.

Table 1. Indice Miller of ZnO samples.		
No.	Diffraction angle, 20	Indices Miller (hkl)
1	31.80	100
2	34.59	002
3	36.22	101
4	47.54	102
5	56.59	110
6	62.84	103
7	67.93	201

Figure 3 showed the transmittance spectra of ZnO thin films annealed at different temperature as a function of photon wavelength. These spectra were obtained by Uv-Vis spectrometer measurement. The transmittance value of ZnO films relatively high (up to 80%). This result indicated that ZnO thin films have good probability for transparent conducting oxide (TCO) layer application. The transmittance value of ZnO thin films slightly decrease with increasing in annealing temperature. We

also obtained that the optical response of ZnO films shift to longer wavelength with increasing in annealing temperature. This tendency showed the improvement of the structure of the films caused by annealing process.



Figure 3. The transmittance spectra of ZnO thin films deposited by spin coating and annealed at different temperature

The optical absorption coefficient was also affected by annealing temperature as shown in Figure 4. The optical absorption coefficient value of ZnO thin films was relatively high (up to 5 x 10^4 cm⁻¹). With a high absorption coefficient makes this material able to absorb more photon.



Figure 4. The optical absorption coefficient (α) of ZnO thin films deposited by spin coating and annealed at different temperature

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Figure 5 showed the optical band gap of ZnO thin films annealed at various temperatures. The optical band gap value was obtained using Touc's Plot method. We found that the optical band gap of ZnO films decrease from 3.82 - 3.69 eV with increasing in annealing temperature from 300 - 700 °C. The optical band gap of the films was determined by film structure and the atomic composition of the film. In this research, the optical band gap of the films was strongly depended on annealing temperature. The decreasing of the optical band gap was due to the improvement of the crystallization of ZnO.



Figure 5. The optical band gap (E_g) of ZnO thin films deposited by spin coating and annealed at different temperature, (a) $T_a = 300 \ ^{\circ}C$, (b) $T_a = 400 \ ^{\circ}C$, (c) $T_a = 500 \ ^{\circ}C$, (d) $T_a = 600 \ ^{\circ}C$, and (e) $T_a = 700 \ ^{\circ}C$

Surface morphology of ZnO films annealed at different temperature was shown in Figure 6. These images were obtained from atomic force microscopy (AFM) measurement. We observed that the surface morphology and the roughness of the ZnO films were also improved with the increasing of annealing temperature. Surface morphology of the film annealed at temperature of 700 ^oC seem more uniform with small granular.

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Figure 6. AFM images of ZnO thin films deposited by spin coating and annealed at different temperature, (a) $T_a = 300$ °C, (b) $T_a = 400$ °C, (c) $T_a = 500$ °C, (d) $T_a = 600$ °C, and (e) $T_a = 700$ °C

4. Conclusions

In this research, ZnO thin films have been successfully deposited from zinc acetate by using spin coating method and annealed at different temperature. We found that the characteristics of ZnO thin films were strongly depend on annealing temperature. The ZnO thin film annealed at 500 $^{\circ}$ C for 15 minutes was corresponded to hexagonal structure with that the grain size of ZnO particle is about 23.8 nm. The transmittance value of the ZnO films was relatively high (up to 80 %). Annealing temperature also influents the optical absorption coefficient of ZnO films. We found that the optical coefficient value of ZnO films is up to 5 x 10^4 cm⁻¹. The optical band gap of ZnO films are about 3.69 – 3.82 eV. The optical bandgap of ZnO films decreases with increasing in annealing temperature. We also obtained that annealing temperature affects the surface morphology of the ZnO films.

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