Influence of Baking Temperature on Physical Properties of ZnO Transparent Thin Films by Sol-gel Dip Coating Method

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Abstract

ZnO thin films were deposited on glass substrates by sol-gel dip coating method under different baking temperatures of 80, 100, 150, 200, and 250 °C. The influence of baking temperature on structural, morphological, and optical properties of ZnO transparent thin films were investigated by X-ray diffraction, Scanning Electron Microscope, and UV-Visible spectroscopy. X-Ray Diffraction results show crystal structure of ZnO wurtzite structure. The optical transmittance spectra of ZnO thin films shows high optical transmittance in visible range. At high baking temperature condition, the XRD peak intensities of the deposited thin films were higher than the low baking temperature condition. The results imply that baking temperature has significant influence on both structural and optical properties of sol-gel derived ZnO thin films.

Keywords: ZnO thin film, Sol-gel dip coating, baking temperature

1. Introduction

In general, transparent conducting oxide films (TCOs) play important roles in the optoelectronic devices because of its suitable properties such as high transparency and excellent electrical conductivity. ZnO is one of interesting candidate materials due to its exceptional properties such as high transparency in visible region, low cost, resource ability, and nontoxicity. Moreover, the optical and electrical properties of ZnO films can be enhanced to meet desired features by doping Group III and IV element (Group III element such as B or Al, Ga, In, etc. and Group IV element such as Si or Ge, Sn, Ti, Zr, etc.) [1, 2] into ZnO lattice. Based on previous works, it could be advised that proper deposition technique with specific details during coating process is mandatory in order to prepare thin films with high quality and well-defined properties. Among well-known deposition processes, sol-gel based deposition processes possess considerable advantages including ease of processing equipment and deposition, ability of large area coating with homogeneity and readiness of doping. During typical sol-gel based coating process, mild heating during each coating is one of significant processing factor that may affect the quality of the films. Enigochitra et al. reported that the substrate temperature during the sol-gel spray pyrolysis process of ZnO thin films had significant effect on crucial properties of the films [3]. Guo et al. reported that baking temperature during coating was a key processing parameter for depositing good quality ZnO thin films via sol-gel spin coating method [4].

In this work, ZnO thin films were deposited onto glass substrate by sol-gel dip coating method. The investigation of baking temperature during the coating process on structural, and important optical properties of ZnO thin films has been conducted and reported.

2. Experimental Details

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To prepare starting precursor, 0.2M zinc acetate dihydrate ($(CH3COO)_2Zn \bullet 2H_2O$) was dissolved in absolute ethanol (C_2H_6O) solvent and diethanolamine (DEA,$HN(CH_2CH_2OH)_2$) used as stabilizer. The solution was stirred at 75°C for 3 h and aged for 24 h at room temperature. ZnO thin films were deposited on the glass substrates by sol–gel dip coating at drawing speed of 60 mm/min. During each dip-coating the films were baked at different temperatures of 80, 100, 150, 200, and 250°C for 5 min in air to evaporate the solvent. The coating was repeated several times before annealing process in air at 500 °C for 2 h.

The crystal structures of ZnO thin films were measured by X-ray diffraction (JEOL: JSM-634OF) while their surface topography and thickness was monitored by a field emission scanning electron microscope (JEOL: JSM-634OF). Optical properties of the films were investigated by a UV–Vis spectrophotometer (Thermo Electron: Helios Alpha).

3. Results and Discussion

Fig.1 shows the XRD patterns of ZnO thin films deposited on glass substrates by sol-gel dip coating method under different baking temperatures with variation of 80, 100, 150, 200, and 250°C, and annealed in air at 500°C. The XRD patterns show characteristic diffracted peaks of ZnO hexagonal wurtzite structure with (100), (002), (101), (102), (110), and (103) [JCPDS: 36-1451]. From XRD peaks, it is clearly noticed that peak intensities were increased with increase of baking temperature, indicating the generation of well-defined crystallinity of the films. The average crystallite size (D) of the films can be calculated by Scherrer's equation [5].

$$D = \frac{0.9\lambda}{\beta\cos\theta} \quad (1)$$

Where λ is the wavelength of incident X-ray source CuK_α (λ = 0.15405 nm), β is the FWHM measured in radians and θ is the Bragg angle of diffraction peak. The FWHMs of three major peaks (100), (002), and (101) exhibit considerable decrement with increasing baking temperature until 250°C implying the increase in its crystallite size of the prepared films. Their crystallite sizes were evaluated from the major (100), (002), (101) peaks. The average crystallite size seems to be increased from 4 nm to 17 nm as the baking temperature increases. Based on Pecharapa *et al.* report [4], the TGA-DTA result of zinc acetate sol-gel precursor in air was significant change at 258.29°C. The increase in average crystallite size and enhancement in crystallinity of the films may be associated to the hydrolysis and thermal decomposition of zinc acetate during the process [5]. Other mechanism that may play major role on this feature is good interfacial matching between deposited layers of intermediate species of sol-gel precursor after each baking that can act as seeding layer for the next deposited layer. The good formation of this intermediate product at higher baking temperature could initiate the nice interfacial matching leading to the well-developed thin films.

Fig.2 shows surface morphologies of deposited films monitored by SEM. It is indicated that the ZnO thin films were polycrystalline structure in nature. The grain size of thin film obviously increases with increasing baking temperature that is in harmony with XRD result. The average thickness of deposited film is approximately 200 nm as seen from the cross section image in Fig 2(f).

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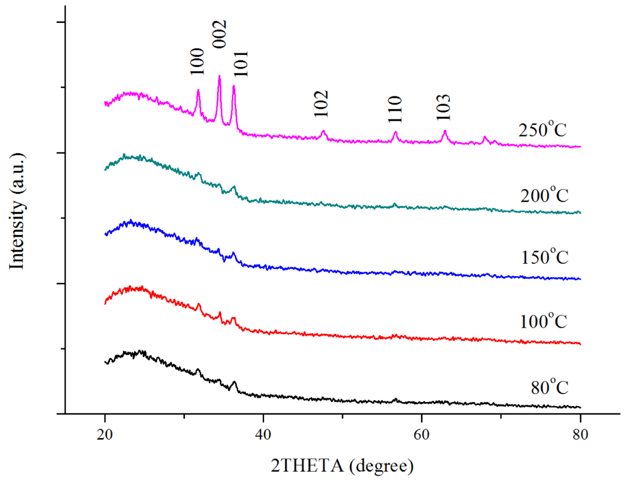

Fig.1. XRD patterns of sol-gel dip-coated ZnO thin films prepared at different baking temperatures.

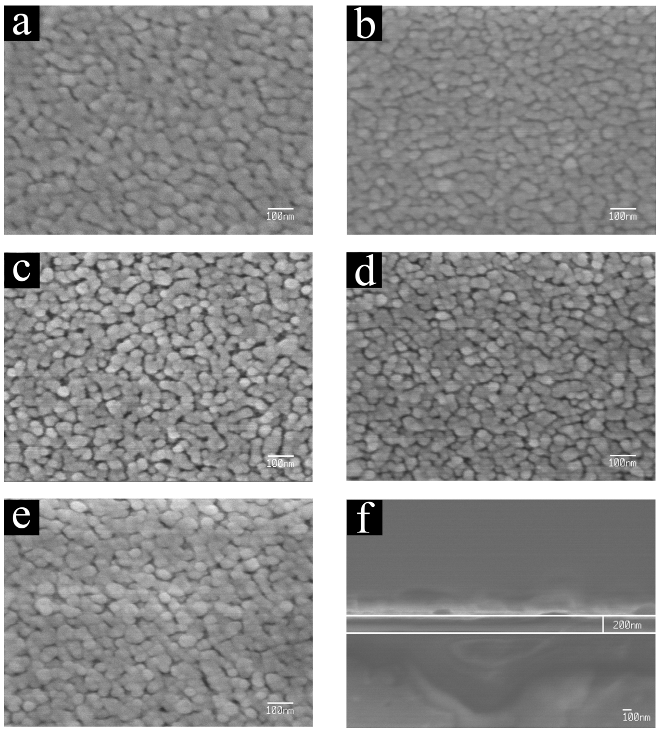

Fig. 2. SEM micrographs of ZnO thin films prepared at different baking temperature; (a) 80°C, (b) 100°C, (c) 150°C, (d) 200°C, (e) 250°C, and (f) cross-section of the film.

Fig.3 shows the optical transmittance spectra of the ZnO thin films prepared at different baking temperatures of 80, 100, 150, 200, and 250°C, and annealed in air at 500°C. In the range of 350 nm to 700 nm, the deposited films have average optical transmittance of 97%, 96%, 96%, 96% and 92%, respectively. In the condition of baking temperature 250°C, the absorption edge is prominent and well-defined. The optical band gap of deposited films was estimated by The Tauc's method represented as following equation [6]:

$$(\alpha h\upsilon) = A(h\upsilon - E_g)^{1/2} \qquad (2)$$

Where α is the absorption coefficient, $h\upsilon$ is the photon energy, A is a constant, E_g is the optical band gap. Inset of Fig. 3 represents the optical band gap of the deposited thin film baked at 250°C from the extrapolation of the straight section to the energy axis of the plot of $(h\alpha\upsilon)^2$ versus $h\ \upsilon$. The optical band gap of the deposited thin film shows the value of 3.26 to 3.27 eV. The influence of baking temperature on the optical band gap may be due to the degraded crystallinity and reduced grain size of thin films as shows in the low temperature conditions [6].

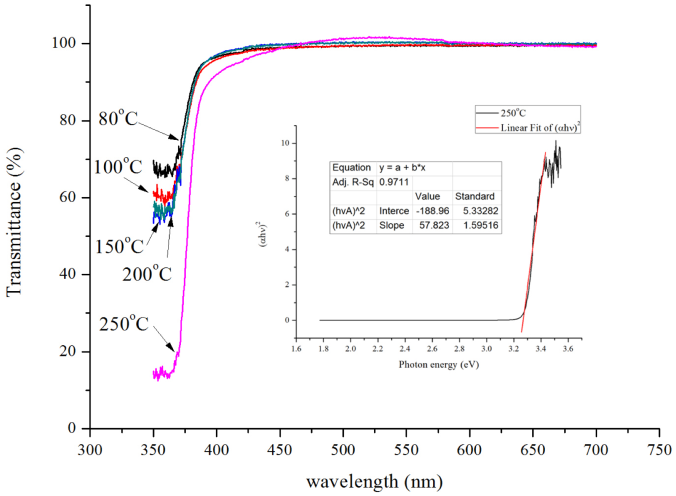

Fig. 3. Optical transmittance spectra of ZnO thin films with different baking temperature and (inset) Tauc's plot.

4. Conclusion

In summary, ZnO thin films were deposited on glass substrates by sol-gel dip coating method under different baking temperatures of 80, 100, 150, 200, and 250°C, and annealed at 500 °C. The ZnO thin films have hexagonal wurtzite structure and show high optical transmittance more than 90%. With baking temperature of 250 °C, the deposited film exhibit superiority in structural, morphological and optical properties. From this present research work, it could be deduced that baking temperature during sol-gel dip coating is one of key parameter affecting the crucial properties of ZnO thin films.

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