

Significance of Annealing Temperature on Transmission Properties of ZnO Thin Film by Hydrothermal Method

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Abstract

In this study Zinc oxide (ZnO) thin films were prepared using a low cost hydrothermal method and investigated for the effect of different annealing temperatures. The ZnO precursor solution was made using zinc acetate dehydrate $Zn(CH_3COO)_2 \cdot 2H_2O$ (ZAD) and hexamethylenetetramine $C_6H_{12}N_4$ (HMT). The samples were annealed at different temperatures (100°C, 200°C, 300°C, 400°C & 500°C) for 90 minutes and irradiated using refractory furnace. Thin films were analyzed using ultra violet-visible spectrophotometry, X-ray Diffraction (XRD) with Cu-K α radiation to determine the structure of the film, four probe and two probe method for electrical properties and Scanning Electron Microscopy (SEM) with Energy Dispersive X-ray Spectroscopy (EDX) for surface morphology and chemical composition. Result shows the 500°C annealing produce good thin films as well as the increasing deposition time from 4 to 24 hours leads to higher crystalline films with increased transmittance.

Keywords: Zinc Acetate Dihydrate (ZAD), Hexamethylene tetramine (HMT), Hydrothermal, Thin Film, Transmittance, Seed layer.

1. Introduction

Zinc oxide is a wide-bandgap semiconducting material with various applications due to its unique properties such catalytic [1], optoelectronic [2], and photochemical [3], Semiconducting ZnO thin films have been deposited using number of ways like spray pyrolysis method [4,5], thermal evaporation [6], sol-gel [7], chemical vapor deposition (CVD) [8], etching [9], hydrothermal [10,11]. Out of

these techniques hydrothermal is the easy for film deposition. ZnO has broad applications in the fields of optical sensing [2], piezoelectric [12, 13] photocatalysis [14], gas sensing [15], photodetectors [16, 17], sensors [18] also, as materials for room temperature ultraviolet (UV) laser diodes [19]. Due to nontoxic nature of ZnO, it is environment friendly so used as semiconducting material in world of solar cell [20]. Also various conditions like doping [21,22], control of deposition conditions like concentration, substrate used, deposition time etc. [11,23,24], and annealing [24,13] makes ZnO thin films non-conducting or high-resistivity which further can be used as insulating layers in devices [25,26], dielectric materials, and sensors [15].

Until now lot of experiments have been performed to explain how the factors like precursor source, concentration [23,27] precursor solution's nature, deposition temperature affects the optical, structural & electrical properties of prepared films [10,11]. Elif Peksu, Hakan Karaagac, had successfully fabricated a superstrate CZTS solar cell based on ZnO nanorods over ITO coated glass substrate [20]. Also recently Hussein M. Hussein, had studied about photosensitivity of ZnO thin films by doping of copper. [28] Zhonghao Liu Xuanpu Dong et al. had studied Effects of sputtering process and annealing on the microstructure, crystallization orientation and piezoelectric properties of ZnO films [13].

In the present work, ZnO thin films were prepared using HMT and ZAD via hydrothermal process. Initially the seed layer was prepared using drop casting method [23]. The significance of experimenting different annealing temperature and deposition time on the optical, structural and electrical properties of the prepared films was studied.

2. Methodology

All chemicals employed in this research were of analytical reagent grade, acquired from Loba Chemie, and utilized as received without further purification. Initially, a ZnO seed layer film was prepared using a drop-casting technique with a ZnO concentration of

0.025 M [7]. Subsequently, for the hydrothermal growth, the aqueous solutions were prepared by dissolving ZAD and HMT in deionized water at equimolar concentration of 0.025 M with continuous stirring for 30 minutes to ensure homogeneity. This homogenous solution was then transferred to a Teflon-lined autoclave for hydrothermal reaction.

In order to optimize and prepare thin film, annealing temperature and deposition time were used and two different experiments were carried out. In first experiment the hydrothermal process was carried out at 95°C for 24 hr followed by the meticulous rinsing with deionized water to eliminate any residual solution. Finally, the samples were dried in a refractory furnace at various annealing temperatures (100°C, 200°C, 300°C, 400°C, and 500°C) for 90 minutes. Second experiment was carried out by keeping the annealing temperature constant at 500°C, and deposition time was varied to at 4, 8, 12, and 24 hours at constant 95°C deposition temperature.

The synthesized ZnO nanostructures were comprehensively characterized using XRD (Bruker D8 Venture), SEM (TSCAN Vega II XMU), Energy dispersive X-ray spectroscopy (EDX), UV-visible spectrophotometer (Systronics UV-VIS spectrophotometer :119), four probe set (DFP 02), two probe set (High voltage power supply, Model EHT-11, DPM-11) to understand the properties.

3. Results and Discussion

First experiment where the hydrothermal process was carried out at 95°C for 24 h with the varying annealing temperatures 100°C, 200°C, 300°C, 400°C, and 500°C for 90 minutes showed improper deposition of film at 100°C, 200°C, 300°C, and 400°C, whereas the annealing at 500°C showed proper thin film formation. Therefore, in the further experiment annealing temperature and annealing time was kept constant at 500°C for 90 min and the deposition time was varied at 4, 8, 12, and 24 hours at constant 95°C deposition temperature. The thin films produced at deposition time 4, 8, 12, and 24 hours then analyzed for their properties.

1. X-Ray Diffraction (XRD) Analysis

The phases of prepared ZnO thin films were confirmed by using XRD with 2θ ranges from 10° to 90° , $0.02^\circ \text{ s}^{-1}$ of scanning rate and Cu-K α radiation of 0.1540 nm. The XRD pattern observed in figure 1 depicts the five peaks at 31.80° , 34.50° , 36.40° , 56.60° and 67.47° shows orientations at (100), (002), (101), (110) and (200) respectively.

These orientations of the ZnO samples exhibits a polycrystalline hexagonal wurtzite structure (ICSD 01-074-0534) corresponds to PDF number 36-145. Figure 1 also depicts the relatively high intensity of peak (100) for all samples. The average crystallite size (d) of ZnO thin films, was estimated using Scherrer's formula $D = k \lambda / \beta \cos \theta$ where, where (D) is the average crystallite

size in nanometer, (k) is constant and equals (0.9), (λ) is wavelength for Cu-k α target for XRD instrument, (β) is full width at half maxima (FWHM) measured in radians of X-ray peak, (θ) is Bragg's angle. Values obtained were mentioned in table 1. illustrates that the value of FWHM goes on decreasing with the increasing deposition times and crystallinity of the film increased with increase in time of deposition. The average value of crystallite size (D) varied from 8.15 to 11.18 nm as the deposition time increased from 4 to 24 hr, respectively. This result can be attributed to longer hydrothermal deposition times which lead to recrystallization of the atoms into wurtzite lattice structure which further increases crystallinity due to production of the thermal energy produced by annealing process. [29] The strong XRD peaks confirms the high crystallinity of the ZnO films.

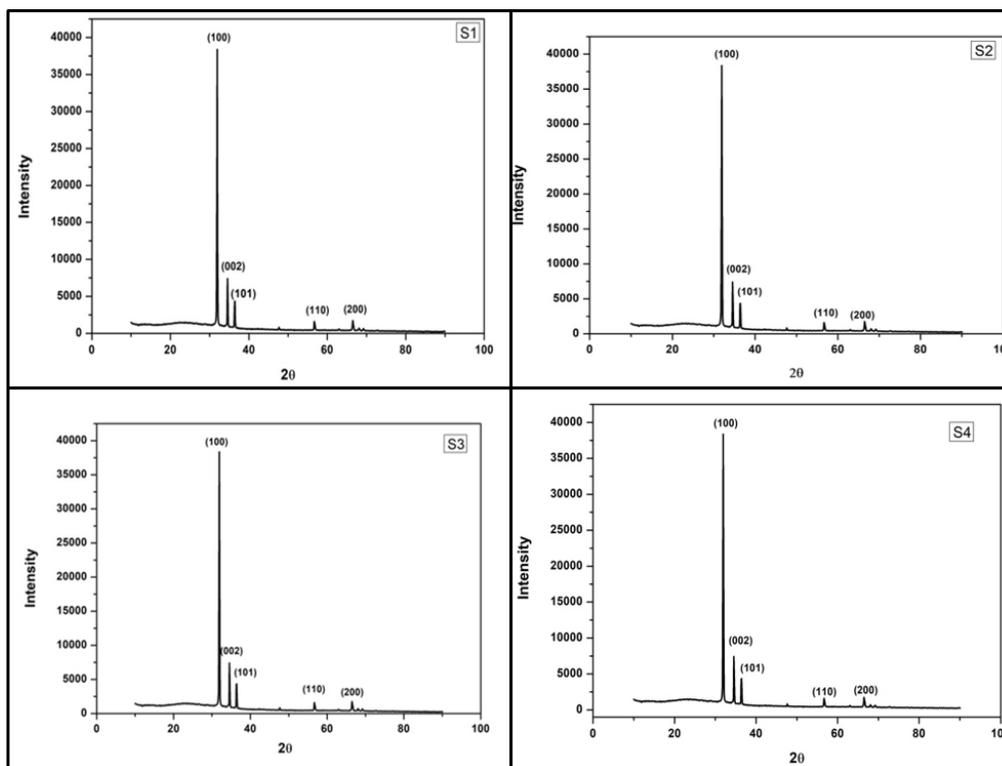


Fig.1. X-ray diffraction pattern of the obtained ZnO films using hydrothermal growth method via Zinc acetate at annealed temperature 500°C for different deposition time (S1- 4 hr, S2- 8hr, S3- 12 hr, S4- 24 hr)

Table 1. The X-ray data ZnO thin films at different deposition time

Sample	Annealing temperature ($^\circ\text{C}$)	Deposition time (hours)	hkl	2θ (deg)	FWHM (deg)	D (nm)
S1	500°C	4	(100)	31.80°	1.0591	8.15
S2	500°C	8	(100)	31.80°	1.0555	8.18
S3	500°C	12	(100)	31.80°	0.7804	11.06
S4	500°C	24	(100)	31.80°	0.7722	11.18

2. Scanning Electron Microscopy (SEM) Analysis

Typical scanning electron micrographs (SEM) for the ZnO samples prepared with deposition time 4, 8, 12, 24 hours by annealing at 500°C are shown ZnO thin film has randomly shaped porous spot-like structures (figure 2). It also depicts the average size for samples S1, S2, S3 and S4 (deposition time 4, 8, 12, 24 h respectively) as 87 nm, 91 nm, 210 nm, 783 nm, respectively. This morphology states that with increase in deposition time grain size increases.

3. Energy dispersive X-Ray (EDX) Analysis

Furthermore, the investigation of the ZnO film performed using the EDX scans. The peaks obtained after EDX analysis of some of the selected area of the SEM image displayed in figure 3 shows Zinc (Zn) and

Oxygen (O) were the basic elements present. The other elements were present due to atmospheric impurity, shown in the table 2. (Chlorine(Cl) due to impurity mixed with distilled water during solution preparation, Tin(Sn) due to prepared film was coated with reflective tin coating during EDX scans and Silicon(Si) peak present due to film deposited on glass substrate).

4. Optical Analysis

For optical study of prepared film the transmittance was found using UV-visible spectrophotometer (Systronics UV-VIS spectrophotometer: 119). During the experimentation air medium was used as reference cell and the corresponding reading of transmittance versus wavelength was taken in the range 200 to 900 nm.

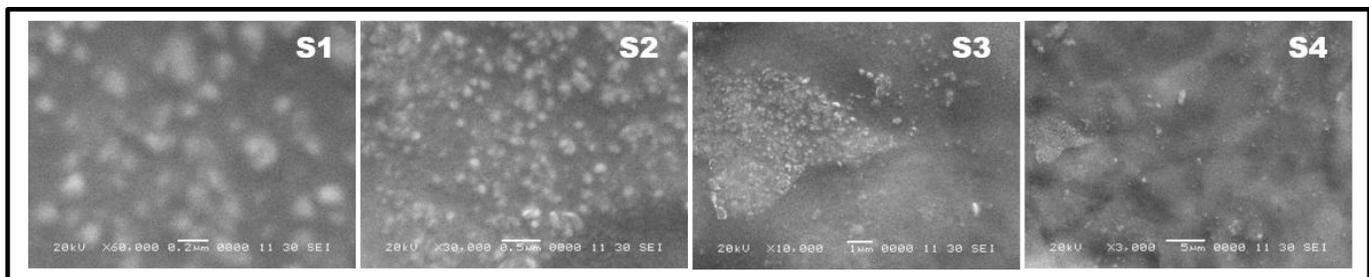


Fig. 2 SEM surface morphology for the ZnO films using Acetate at annealed temperature 500°C for different deposition time (S1- 4 hr, S2- 8hr, S3- 12 hr, S4- 24 hr)

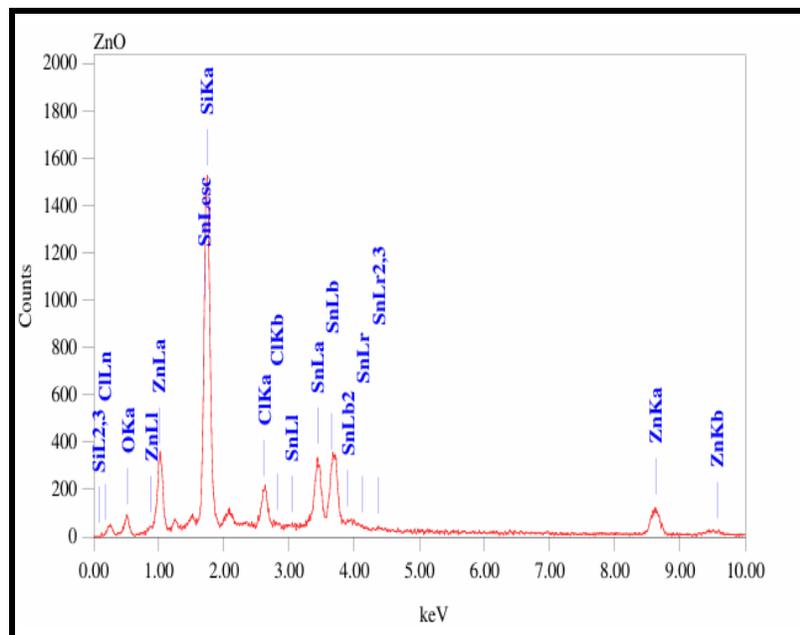


Fig.3 EDX Spectra for the ZnO film using ZAD deposited on glass substrate at annealing temperature 500°C.

Table 2. Elemental composition

Element	Mass (%)
O	5.10
Si	28.73
Cl	4.70
Zn	26.35
Sn	35.12

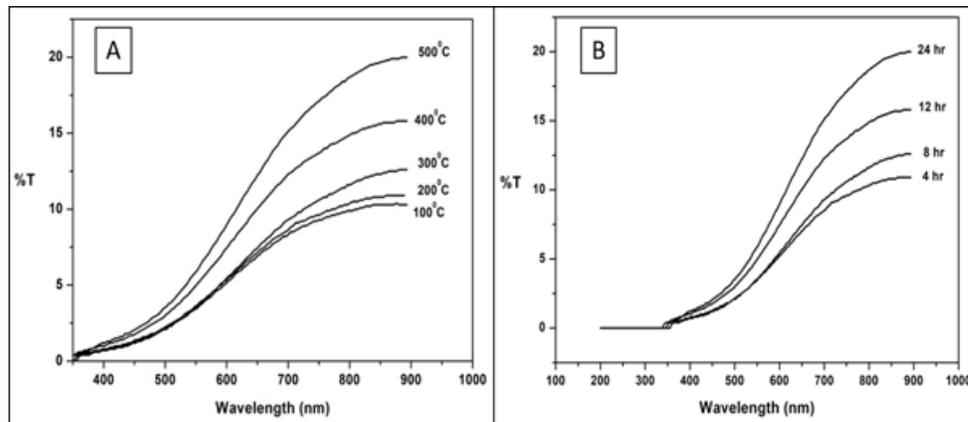


Fig. 4: Transmission spectra of the obtained ZnO films using hydrothermal growth method via Zinc acetate at different annealing temperatures and for different deposition time.

The transmission spectra of ZnO thin films prepared hydrothermally at annealing temperatures 100°C, 200°C, 300°C, 400°C, and 500°C shows 8, 11, 12, 14, 20 percent transmittance respectively (Figure 4A). As the percent transmittance at the annealing temperature 500°C was found 20% which was relatively more as compared to the temperatures other than 500°C. So temperature 500°C was used for further experiments. Whereas the films prepared with deposition time 4, 8, 12 and 24 hours at 500°C shown in transmittance of 11, 12, 14, 20 percent transmittance (Figure 4B). The increase in transmittance at higher annealing temperature that is 500°C and at the higher deposition time 24 h can be attributed to the increased crystallinity with increase in annealing temperature and deposition time which also enhance grain size. (As smaller grain size leads to increased scattering and reduced transmittance.)

5. Electrical Analysis

The prepared films does not shows any readings of current and voltage in four probe and two probe method, this indicates the prepared films are non-conductive.

Conclusion

In the presented work, firstly ZnO seed layer deposited by drop casting method and then ZnO films were deposited on glass substrates by hydrothermal method. The structural analysis was carried out using XRD patterns, where the ZnO had good quality (crystallite size in nanoscale range) and preferred orientations (100), (002), (101), (110) and (200). The EDX was used to perform compositional study. SEM technique was used to illustrates morphology growth using surface images. Optical properties were studied by UV-Visible Spectroscopy which revealed that the film deposited for 24 hr at 500°C shows the highest transmittance. The prepared films do not show current and voltage activity under four and two probe electrical analysis indicates the prepared thin films are non-conductive. In this way as we go on increasing deposition time at higher annealing temperature prepared ZnO film shows more optical transmittance due to high crystallinity and highest grain size.

Conflicts of interest: The authors stated that no conflicts of interest.

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