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Effects of Annealing on Structural and Optical Characterization of Nanostructured ZnS Thin Films

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Abstract:

Nanostructured ZnS films were deposited via chemical spray pyrolysis (CSP) method. These films were deposited at annealing (T_a) of (300, 400 and 500) ˚C. XRD analysis assures film polycrystallinity with predominant orientation along (200). The peak intensity was increased with increase of annealing. Microstrain and the dislocation density decreased with increase of annealing, indicating the enhancement of crystal quality. The optical transition was direct allowed to band gap energies between 3.19 and 3.06 eV depending on annealing. AFM results confirm that average particle size increase from (50 to 80) nm and average roughness decrease with increase of T_a.

Keywords: ZnS, spray pyrolysis, annealing, structural, Optical.

1-Introduction

ZnS is wide bandgap semiconductor, belongs to II–VI group. ZnS occur in two crystallographic shapes that possess a cubic zinc blend or hexagonal wurtzite structure [1]. The band gap of cubic lies between 3.54–3.6 eV, while for hexagonal was in the range of 3.74–3.87 eV [2]. ZnS films have many applications in optoelectronic device such as LED [3, 4], solar cells [5], gas sensors [6], antibacterial [7]. Various techniques were used to deposited ZnS such as sol-gel [8], Successive ionic layers adsorption and reaction [9], CVD [10], CSP [11-13], MBE [14], hydrothermal [15], RF Reactive Sputtering [16], wet chemistry processes [17]. Many researchers are working on the fabrication of ZnS films by CSP, because of it is simple, low cost and better control over the substrate. Besides the films have acceptable homogeneity. [40-60]This study was focused on the relationship between annealing temperature and surface morphology of nanostructured ZnS thin films by CSP.

Experimental

ZnS films are grown via CSP method. A solution containing 0.1 M $ZnCl_2$ and SC(NH2)2 resolved in redistilled water to get ZnS. The nozzle was put at 29 cm over the base. flow rate 5ml/ min, spraying time was 8 s, waiting 2 min to restrain cooling, transporter gas was air keeping on 10^5 Pascal. Film thickness is measured by gravimetric method, their values were in the domain of 350 ± 35 nm. Substrate temperature was utilized 300 ºC. Transmittance is recorded via double beam spectrophotometer. High accuracy X-ray (Shimadzu, model: XRD-6000, Japan) is used to measure film structure, AFM was applied to find out surface topography.

Result and Discussion

Fig.1 offers XRD styles of nanostructured ZnS. The film was revealed with high intensity peak oriented along (200) plane. This was compatible with the zinc blend structure according to (JCPDS card No.55-0566). Table 1 shows the average crystallite size (D) is increasing with T_s increase from 31.27 to 36.49nm, resulting in grain growth, which agrees well with Haque et al. [17], D was calculated by using Scherrer's formula [18-21]:

$$D = \frac{0.94\lambda}{\beta cos\theta} \qquad (1)$$

Where $\lambda = 1.54184$Å, θ is Bragg's angle and β FWHM. The dislocation density (δ), is calculated by [22-25]:

$$\delta = \frac{1}{D^2} \qquad (2)$$

The microstrain (ε) was predestined using the equations [26-29]:

$$\varepsilon = \frac{\beta cos\theta}{4} \qquad (3)$$

These structural parameters are represented in Table 1. Fig. 2 indicate the increment of *D* with the decrease of microstrain and dislocation density.

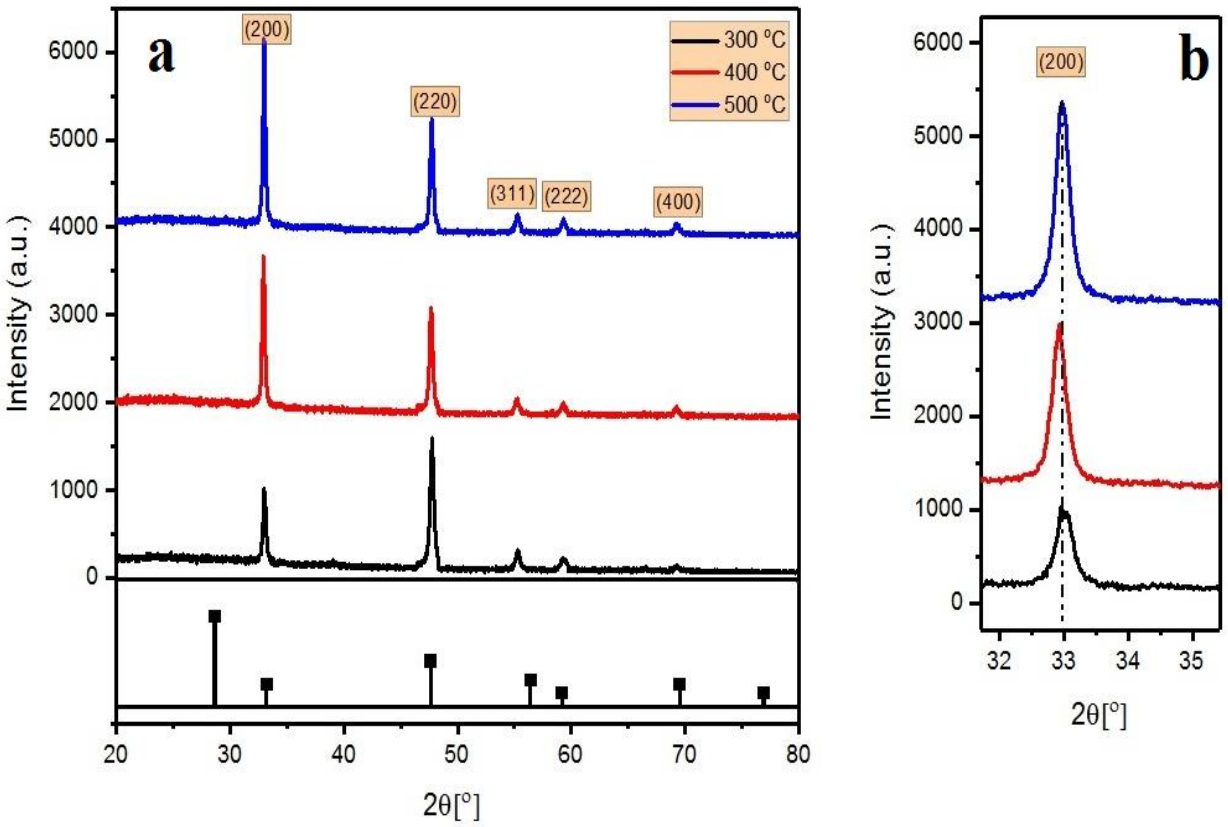

Figure 1. XRD styles of the growing films.

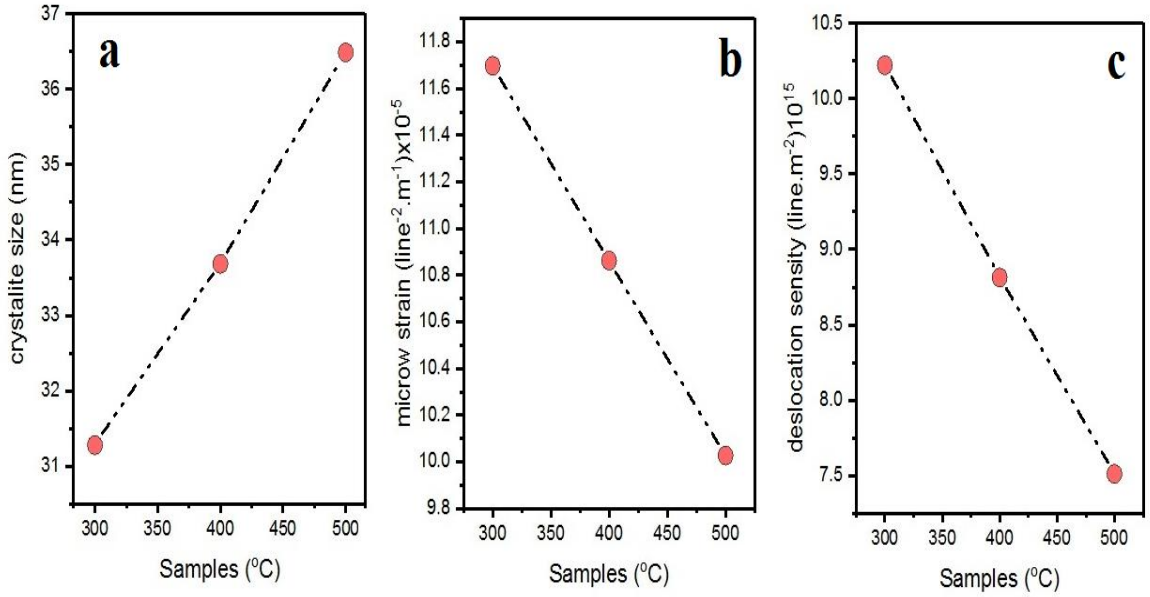

Figure 2. Structural parameter of growing films deposited at various Ta.

Table 1. S_P of the intended films

Sample (°C)	**(hkl) Plane**	**2theta (Deg.)**	**Lattice constant a (Å)**	**FWHM (Rad)**	**D (nm)**	**ε (line^{-2}.m^{-1})× 10^{-3}**	**Dislocation density(δ) (line.m^{-2})×10^{15}**
300	(200)	32.95	5.406	0.28	31.2776	11.69	10.22
400	(200)	32.95	5.406	0.26	33.6835	10.86	8.81
500	(200)	32.95	5.406	0.24	36.4905	10.02	7.51

Fig. (3, 4, 5) shows the AFM used to determine the distribution of the particle size and calculate the particle size of ZnS films grown at 300, 400 and 500 °C annealing. Fig. (3a, 3b, and 3c), show the 2 and 3dimensional AFM images of ZnS Films. uniform particles and densely packed with average particle size (50.16-80.97) nm and average roughness (6.19-1.49) nm was noticed and the RMS was in the zone of (7.22−1.72) nm. AFM result illustrates that average particle size increase and average roughness decrease with the increase of substrate temperature indicating that films improved in crystallinity and decrease in defects and these values proved the film nanocrystalline.

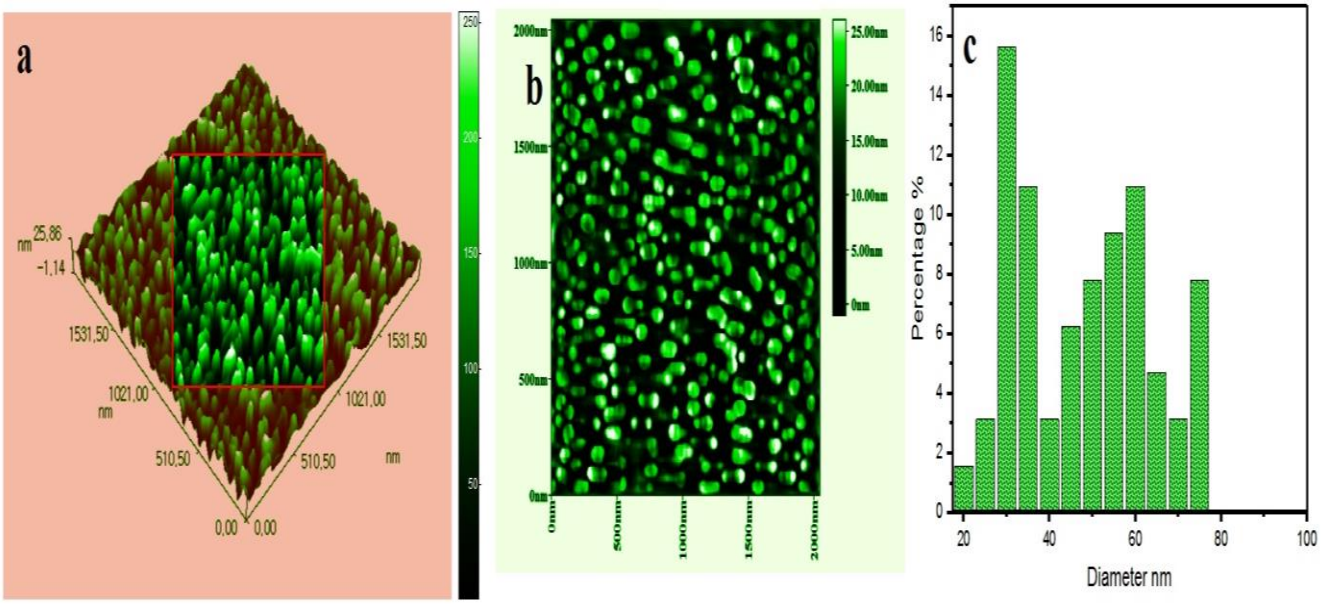

Figure 3. AFM Images of growing films at 300 ºC.

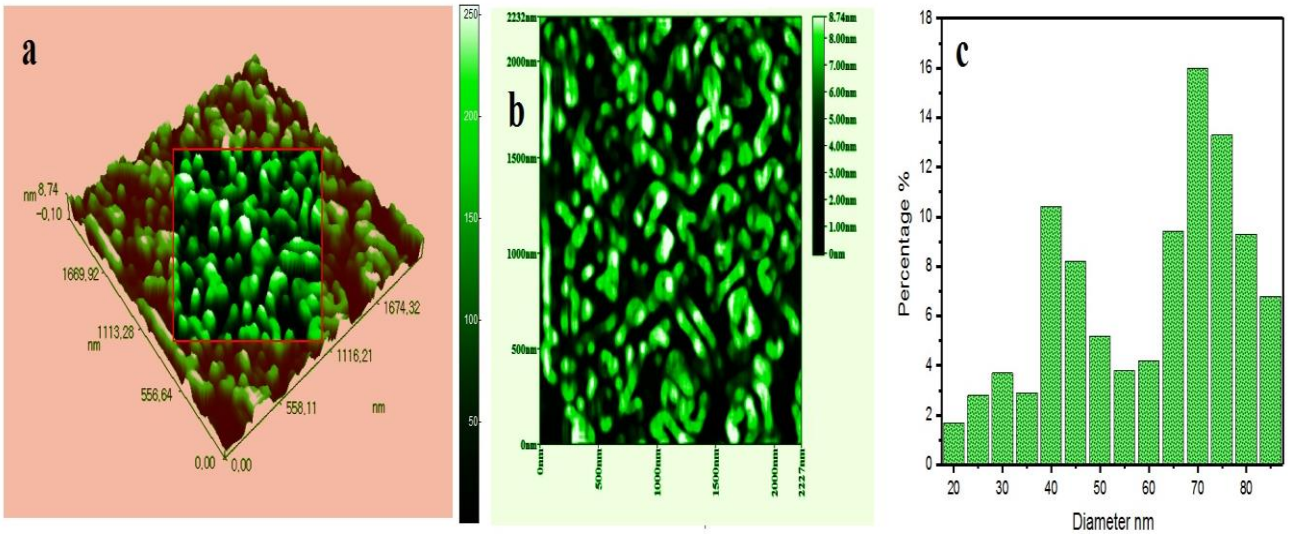

Figure 4. AFM Images of growing films at 400 ºC.

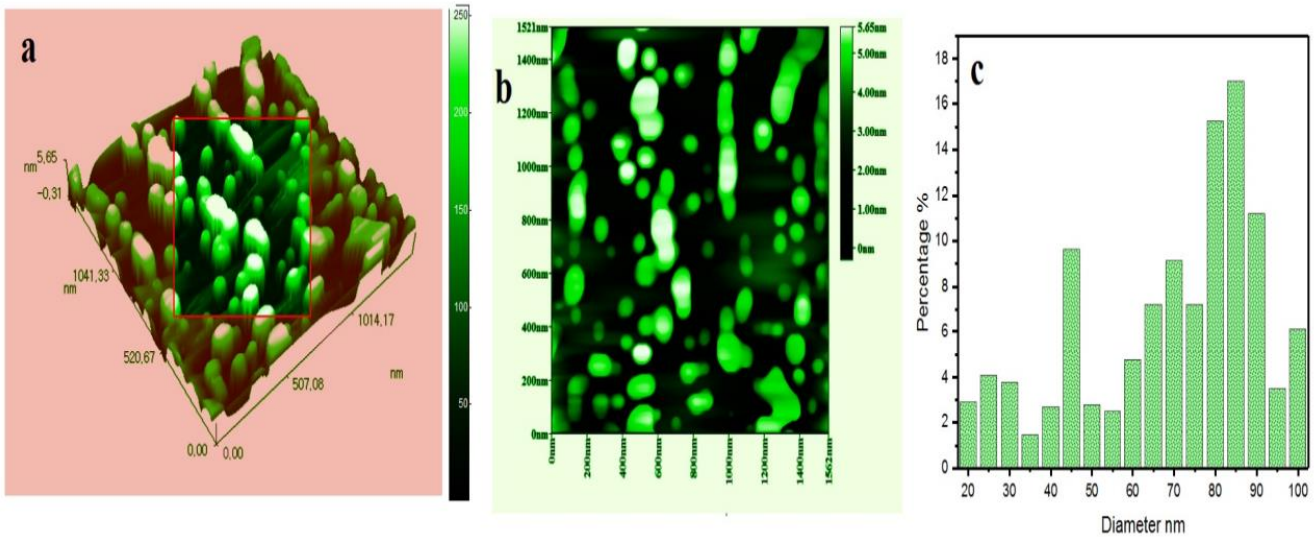

Figure 5. AFM Images of growing films at 500 ºC.

Table2. Surface topography of ZnS films at various T_a.

Samples (ºC)	Average particle size (nm)	Average roughness (nm)	RMS (nm)
300	50.16	6.19	7.22
400	60.12	2.02	2.37
500	80.97	1.49	1.72

Transmittance (T) spectra of ZnS films are displayed in Figure 6. Transmittance increases in the range of (55-80) % with increasing T_a up to 300˚C (55-80) % from 500-900nm.

The absorption coefficient (α) was calculated using the equation [30-33]:

$$\alpha = \frac{1}{t}\ln\frac{1}{T} \qquad -------(4)$$

Where t is the film thickness. Fig. 7 displays α of samples at various T_a. The absorbance increased slightly with increasing the T_a of the deposition and the values of ($\alpha > 10^4$ cm^{-1}). The absorption coefficient is shifted toward lower wavelength of the grown films at low T_a. These results are fit well with references [34,35].

Tauc's relation was used to set band gap energy E_g [36-39], via a plot of $(\alpha h\nu)^2$ as a function of $h\nu$ (as shown in Figure 8).

$$(\alpha h\nu) = A\left(h\nu - E_g\right)^n \qquad ----(5)$$

For direct transition n = ½,. Where A is a constant, α is the absorption coefficient, $h\nu$ is the energy of incident photon. It was noticed that E_g value decreases with increasing T_a; 3.19 eV for 300˚C compared to 3.06 eV for 500˚C. This could be assigned to quantum size effect.

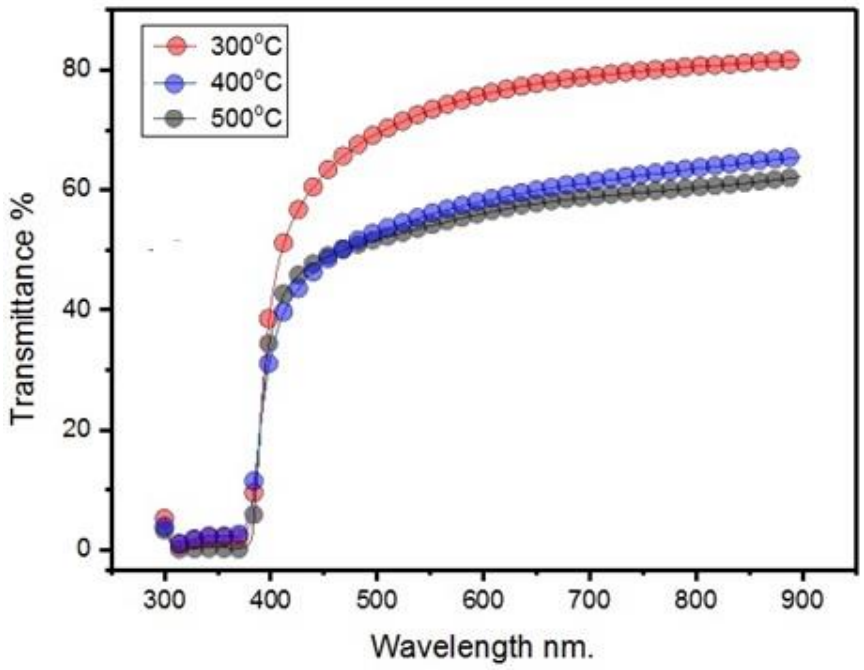

Figure 6. Transmittance of growing films at various T_a.

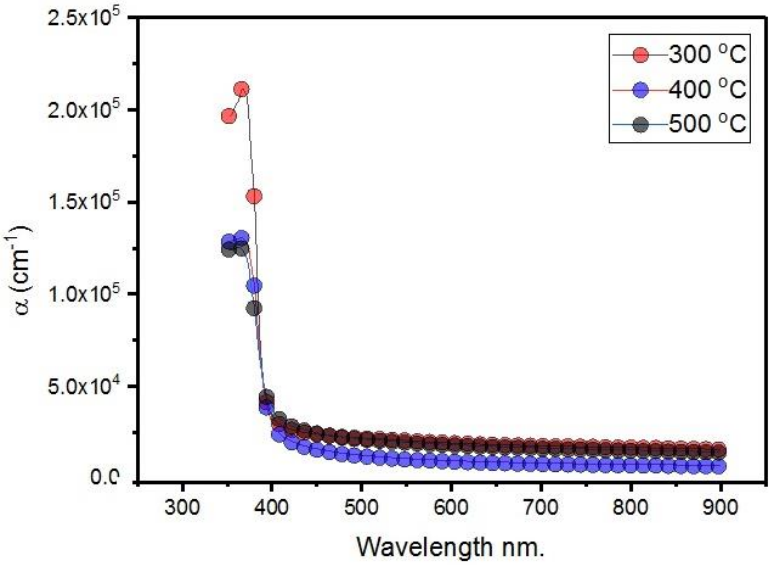

Figure 7. Absorption coefficient of growing films at various T_a.

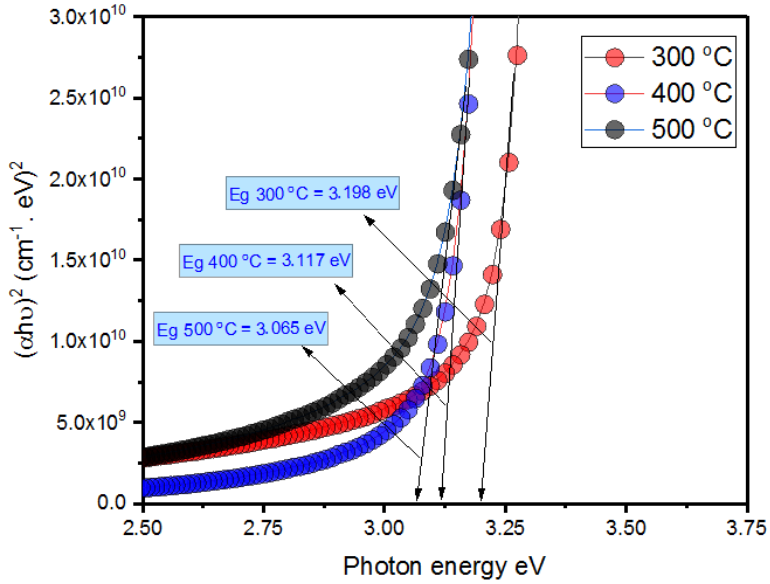

Figure 8. Energy gap of growing films at various T_s.

Conclusions

ZnS thin films are grown by a CSP method onto glass substrate at various T_a of (300, 400 and 500) ºC. The XRD analysis emphasizes the formation of polycrystalline ZnS with high peak intensity along (200) plane, which increases with the increase in T_a. Microstrain and dislocation density was decreased with an increase in T_a .Transmittance increases in the range of (55-80) % with increasing T_a the value of band gap energy E_g decrease with increasing T_a. The absorption coefficient is shifted toward lower wavelength for as-deposited at low substrate temperature. AFM result illustrates that average particle size increase (50-80) nm and average roughness decrease (6.19-1.49) nm with increase of substrate temperature indicating that improvement in film crystallinity.

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