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# Effect of substrate temperature on the morphological and optical properties of nanocrystalline ZnO films formed by DC magnetron sputtering

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**Abstract:** *Zinc oxide (ZnO) thin films were formed by DC magnetron sputtering onto glass substrates held at different temperature. The temperature of the substrate was in the range 473 – 673 K. The temperature dependence, morphological and optical properties of ZnO films were systematically investigated. Atomic force microscopic analysis revealed that the growth of nanostructure in all the films. The root mean square roughness of the films increased from 0.61 to 3.83 nm in the substrate temperature range of investigation. The average optical transmittance of the films was about 78 % in the visible region. The optical band gap of the ZnO films decreased from 3.349 to 3.357 eV with increase of substrate temperature 473-673 K respectively and have a direct-transition type.*

**Key words:** *Zinc oxide, transparent conducting oxide, DC magnetron sputtering, optical transmission and structure properties.*

## I. Introduction

Zinc Oxide (ZnO) is an attractive semiconducting material due to its promising properties of high electrical conductivity and good optical transparency. It has advantages of nontoxicity, low cast and abundance of raw material with high stability. ZnO was found various technological applications such as solar cells [1, 2], active material for highly sensitive humidity and oxygen gas sensors [3], transparent electrodes for flat –panel displays [4], p-n junction diodes, UV photodetectors [5] and ultraviolet light emitting diodes [6]. In the recent years, the novel ZnO nanostructures are reported to have potential applications for optical emission, catalysis, sensing, actuation, and drug delivery. The increase in surface area and the quantum confinement effects have made nanostructure materials are quite distinct from their bulk form in both electrical and optical properties. Various one-dimensional structures of ZnO, such as nanowires, nanorods, and nanobelts, have attracted much attention [7-9].

ZnO thin films have been grown by various deposition techniques such as thermal oxidation of zinc [10], chemical bath deposition [11], spray pyrolysis [12], sol-gel process [13], electron beam evaporation [14], pulsed laser deposition [15], metal organic chemical vapor deposition [16], and DC [8, 17, 18] and RF [1, 2] magnetron sputtering a. The physical properties of the formed films depend on the deposition method employed and process parameters maintained during the growth of the films. Among these deposition techniques, magnetron sputtering is industrially adopted thin films preparation method because of the advantages in the generation of uniform and large area films. The physical properties of the sputtering films depends mainly on the process parameters such as oxygen partial pressure, sputter power, substrate temperature, substrate bias, thickness and post deposition annealing [3].

The properties of sputtered ZnO thin films are known to depend on deposition parameters such as r.f. power, substrate temperature, type of substrate, pressure, gas atmosphere and thickness. Therefore, when one attempts to grow ZnO films of high quality by using r.f. sputtering, it is necessary simultaneously consider both defect formation and film growth behavior in optimizing ambient O2 pressure in the growth chamber[3, 4].

ZnO has a strong potential for various short-wavelength optoelectronic device applications. However, to realize these applications, a reliable technique for fabricating high quality ptype ZnO and p-n junction needs to be established. Compared with other II-VI semiconductor and GaN, it is a major challenge to dope ZnO to produce p-type semiconductor due to self-compensation from native donor defects and/or hydrogen incorporation. Great efforts have been made to achieve p-type ZnO by mono-doping group-I elements(Li, Na and K), group-IB elements(Ag and Cu) or group-V elements (N, P, As, and Sb) and co-doping III–V elements with various technologies, such as evaporation/sputtering process, ion implantation, pulsed laser deposition, thermal diffusion of As after depositing a ZnO film on GaAs substrate, and hybrid beam deposition [7, 18].

In the present investigation, ZnO films with various substrate temperatures in the range 473 – 673 K by using DC magnetron sputtering techniques. The effect of substrate temperature on the morphological and optical properties of ZnO films was systematically studied and reported the results.

## II. Experimental work

The ZnO thin films were prepared by using DC- magnetron sputtering source Edwards 306 pumping system. The sputtering condition are : Target- anode distance (30)cm , substrate type (glass), substrate temperature in the range 473-673 K, magnetic field 760G, DC power gas pressure $7\times10^{-2}$ Torr and at atmosphere of argon 100%.
ZnO thin films were prepared by DC reactive magnetron sputtering process. The metal ZnO target (99.9% purity, 60mm diameter and 2mm thickness) is fixed on a magnetron-effect cathode, Glass was used as substrate. The distance between the target and the substrate was 30mm. The vacuum chamber was evacuated using a turbomolecule pump to $6\times10^{-2}$Torr before deposition. The target was cleaned by sputtering with power of 50W for 10min with pure Ar atmosphere at the same time the substrate was covered with the shield. The following sputter conditions were used in this work: DC power of 50W, the Substrate temperature (Ts) was 473- 673 K. The parameters of the films were prepared by sputtering on glass substrates were show in table 1.

**Table 1:** Deposition parameters for the growth of ZnO films

| | |
|---|---|
| Sputter target | ZnO |
| Target to substrate distance | 60 mm |
| Base pressure | $2\text{x}10^{-4}$ Torr |
| Sputter pressure | 5 Pa |
| Sputter power | 50 W |
| Substrate temperature | 473, 513, 573 and 673 K |

## III. Results and discussion

The optical transmittance spectra in the wavelength range of 200 to 900 nm of ZnO films deposited at different substrate temperatures are shown in Figure 1. The films deposited at 473K have relatively higher transparency, and small shoulders were observed in the absorption line. However, the images still exhibit an average transmittance of above 77%. The film formed at 573K was relatively lower than the spectral transmittance for the other films prepared at other growth temperatures; moreover, the average transmittance in the visible region was above 70%. The reason for this is that the film was fabricated with a high degree of crystallinity, as indicated in Figure 1. The optical transmittance was also decreased along with the increase in substrate temperature to 513 K. The shift in the absorption line towards a higher energy side can also be attributed to the increase in substrate temperature [6].
Sharp absorption edge was observed at the wavelength of about 390 nm and shifted towards higher wavelengths side with increase of substrate temperature range of investigation. The optical absorption coefficient (α) was calculated from the film thickness (t) and optical transmittance data using the relation [8, 12]:

$\alpha = (1/t) \ln T$ …………..1

Figure 2 shows the plot of $(ah\nu)^2$ versus hν, where a is the optical absorption coefficient, and hν is the energy of the incident photon. The optical band gap (Eg) is calculated from the following expression by assuming a direct transition between valance and conduction bands [4, 17]:

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

where A is a constant, and $E_g$ is estimated by extrapolating the straight-line portion of the spectrum to a zero absorption coefficient value. The optical band gap of the film deposited at 473Kwas 3.356 eV. As the growth temperature increased from 473K to 513K, the optical band gap shifted from 3.356to 3.357 eV. The band gaps at the film formed at 573K (Eg = 3.350 eV). The band gap of the film also decreased with the temperature up to 673K (Eg = 3.349 eV). These results indicate that an increase in the substrate temperature improves the band gap energy of the films at 513K substrate temperature as shown in table 2.

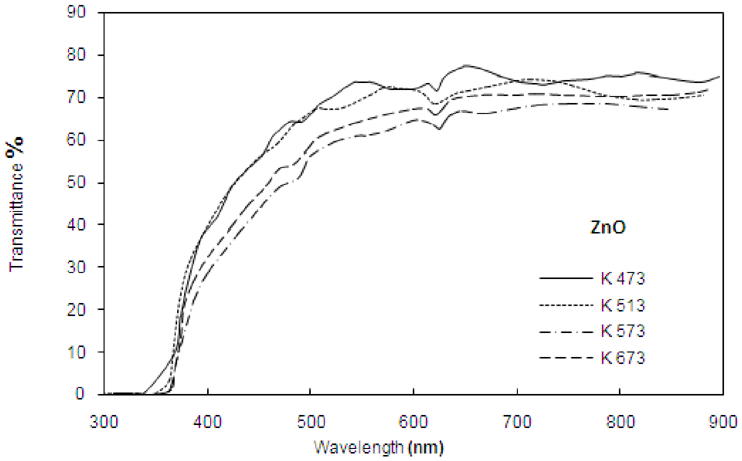



**Fig. 1:** Optical transmittance spectra of ZnO thin film on fused quartz glass.

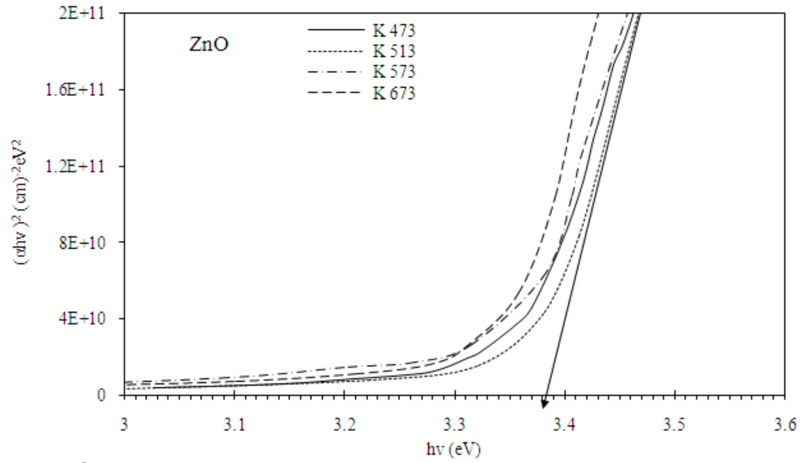



**Fig. 2:** Plot of $(ah\nu)^2$ vs. photon energy (hν) for ZnO films grown at various substrate temperatures

The refractive index n and extinction coefficient k of ZnO film on glass substrate as a function of photon energy were shown in Figure 3 and 4. In Figure 3, the refractive index of ZnO thin film increases as the photon energy increase in the range of 0.5eV to 3.1eV (normal dispersion). The extinction coefficient k increase as photon energy increases and it is very small at low energy part, where the film is transparent. The extinction coefficient (k) of the films was also calculated by using the relation [10, 14]:

$$n = \frac{1+R}{1-R} + \left[\left(\frac{1+R}{1-R}\right)^2 - (k^2 - 1)\right]^{1/2} \qquad \ldots\ldots\ldots\ldots\ldots 3$$

$$k = \frac{\alpha\lambda}{4\pi} \qquad \ldots\ldots\ldots\ldots\ 4$$

The extinction coefficient of the ZnO films first increased from 0.0042 to 0.006 with increase of substrate temperature from 473K to 513K thereafter decreased to 0.005 at high temperature about 673K (Fig. 3), as shown in the Table 2.

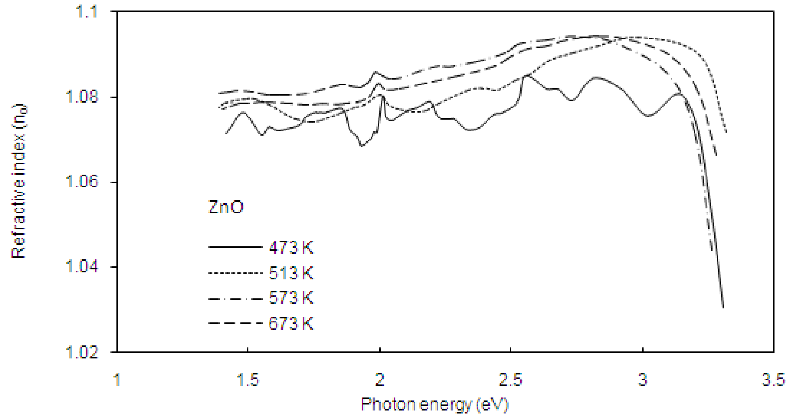



**Fig. 3:** Variation of refractive index with photon energy.

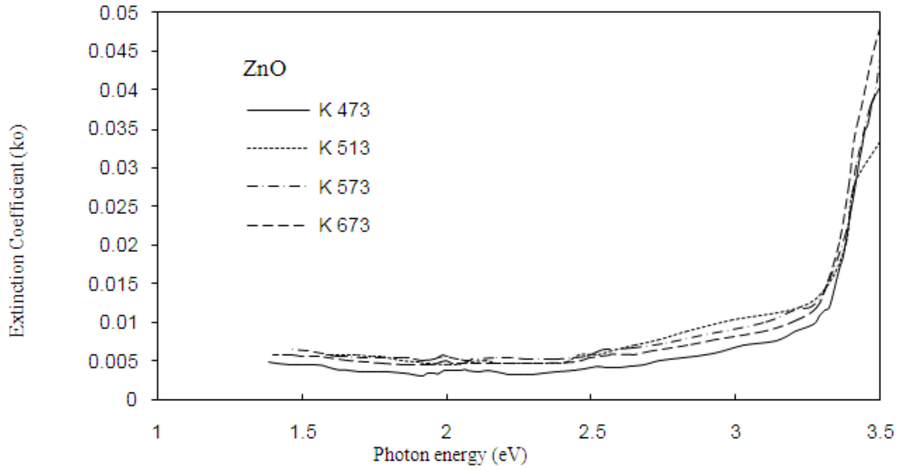


**Fig. 4:** Variation of extinction coefficient with photon energy.

**Table 2:** Optical and structure parameters of ZnO thin films.

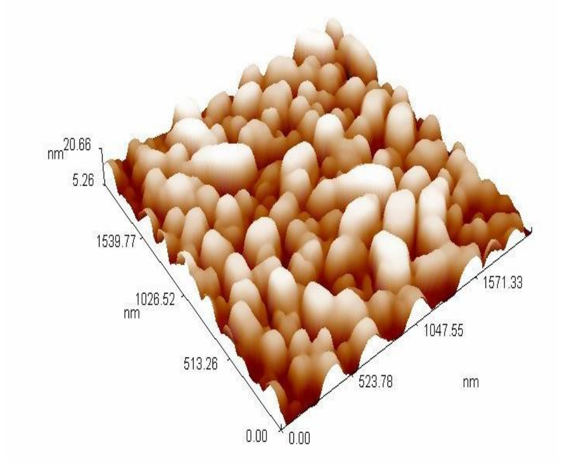
| Temperature (K) | Optical band gap (eV) | Transmittance % | Extinction coefficient |
|---|---|---|---|
| 473 | 3.356 | 78 | $42\times10^{-4}$ |
| 513 | 3.357 | 74 | $60\times10^{-4}$ |
| 573 | 3.350 | 68 | $56\times10^{-4}$ |
| 673 | 3.349 | 70 | $50\times10^{-4}$ |

The three dimensional AFM images taken at a Fig. 3, represent the surface morphology of the ZnO thin films. After acquiring the AFM images, they were subjected to a flattening procedure using the NOVA image processing software. According to a quantitative analysis of the roughness deduced from AFM measuring (Table 2), the values of average roughness (Ra), root mean square (RMS) and coefficients of kurtosis (RKU) changed in relation to the substrate temperature. The films are uniform, dense and well packed between particles. The films deposited at various substrate temperatures show the columnar structure and the particles are arranged uniformly.
The AFM topography of the as-deposited at 573 K revealed that the film surface is rather smooth and compact (Fig. 5 c). A significant difference occurs at temperature of 473K where the film surface exhibits a higher roughness and clear grains can be seen (Fig. 5 a,b).
The atomic force microscopy was used to determine average size of the grains nucleated in the ZnO films as show in table 3, it show that the film have lower value of grain size was 82nm at 473 K (substrate temperature) . For the substrate temperature 513K the root mean square roughness (RMS) has lower value was 0.61 nm. With increase of substrate temperature to 573 K the RMS roughness is gradually increased to 1.91 nm. The increase of RMS with the increase of film substrate temperature was due to the larger size grains formation as well as increase in the porosity of the films [39]. [28]. The increase of RMS of ZnO films leads significant effect on the industrial applications such as gas sensors.

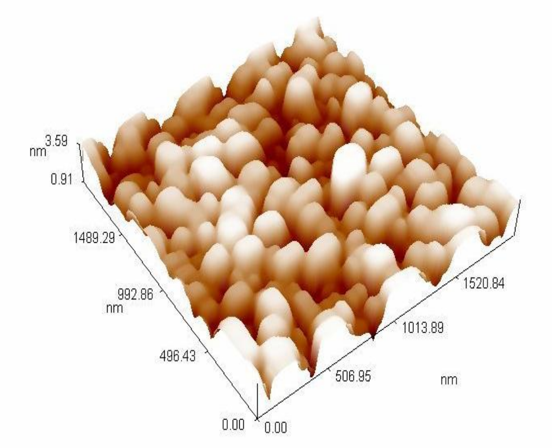
**Table 3:** Structure parameters of ZnO thin films.

| Temperature (K) | Grain size (nm) | RMS (nm) | Roughness (nm) | Peak-peak (nm) |
|---|---|---|---|---|
| 473 | 82 | 3.88 | 3.28 | 15.4 |
| 513 | 88 | 0.52 | 0.52 | 2.68 |
| 573 | 98 | 1.46 | 1.46 | 10.9 |
| 673 | 111 | 2.51 | 2.51 | 24.5 |

a




b



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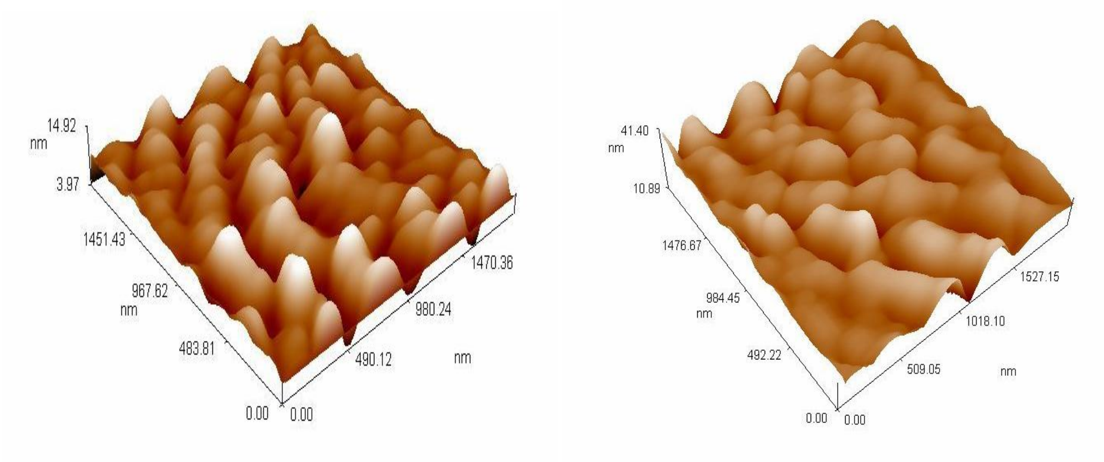


**Fig. 5:** AFM studies on deposited ZnO thin films at different substrate temperature a)473K, b)513K, c)573K and d)673K showing different structure.

## IV. Conclusion

The substrate temperature dependence of morphological and optical properties of nanocrystalline ZnO films deposited by DC magnetron sputtering on glass was systematically studied. The grain size was 22nm and the roughness was increase by increasing the substrate temperature reach to 573 K. The optical studies revealed the maximum transmittance of 78 % found in substrate temperature 513K. The optical band gap of ZnO films decreased from 3.355 to 3.350 eV with increase of substrate temperature from 537 to 673 K.

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