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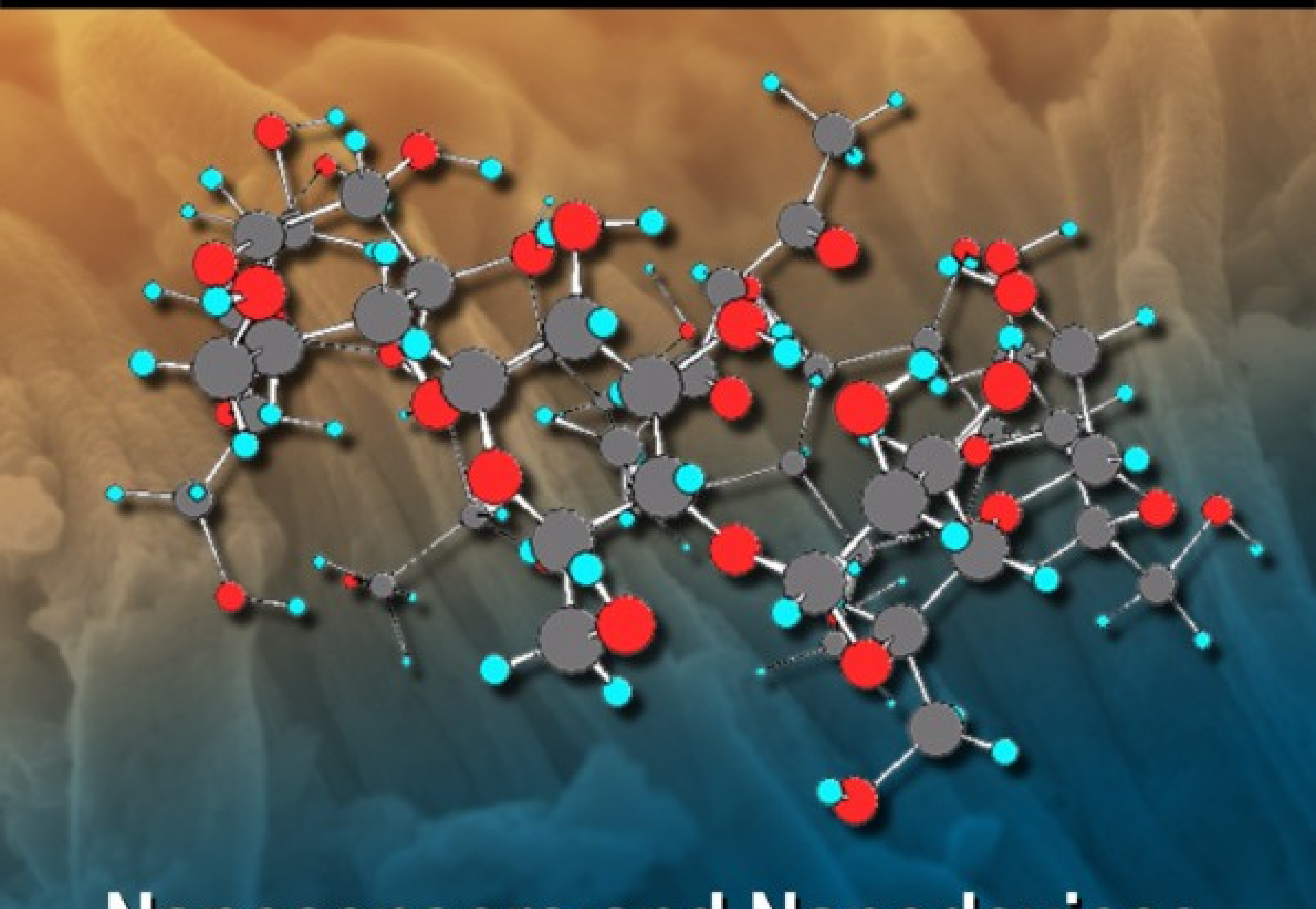

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Structural, Morphological and Optical Properties of Spray Deposited Nanocrystalline ZnO Thin Films: Effect of Nozzle to Substrate Distance

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Abstract: The present work deals with the effect of nozzle to substrate distance on structural, morphological and optical properties of nanocrystalline zinc oxide thin films deposited by chemical spray pyrolysis technique. The XRD analysis revealed that all the films are polycrystalline and are preferentially oriented along (002) plane. This study further the grain structure has considerably improved at nozzle to substrate distance of 28 cm. The morphological observations supported well about importance in grain size with changing nozzle to substrate distance and revealed an average particle size 250 nm. Further, analysis of optical data indicated that the band gap of ZnO films is lying in the range of 3.32 eV to 3.19 eV and found to be dependent of nozzle to substrate distance.

Keywords: Semiconductors, Thin films, Spray pyrolysis, X-ray techniques, Optical properties.

1. Introduction

The increasing use of transparent conducting oxide (TCO) films for liquid crystal display, silicon solar cells, energy windows has promoted many studies on the preparation and characterization of the less expensive and stable TCOs [1]. Among these TCOs, ZnO is an outstanding semiconductor attracting much attention due to its interesting properties such as wide band gap of 3.37 eV at room temperature with large exaction binding energy (60 meV), high chemical stability, low dielectric constant, large electrochemical coupling coefficient and high luminous transmittance [2]. It offers noticeably high chemical, mechanical and thermal stabilities [3]. Due to which it has become a promising material for several technological applications such as ultraviolet (UV) / blue emission devices, solar cells, piezoelectric devices, acousto-optical devices, acoustic resonators, chemical sensors, electro-luminescence displays, heat mirrors and Schottky diodes [4-6]. It also has other major advantages, such as the amenability to wet chemical etching, a high radiation resistance, and relatively low costs [2]. Due to high electron mobility and good transparency it is utilized in paints, rubber, catalysts, sensors, varistors, and so forth [2].

Several techniques have been used for growing ZnO films, like metal organic chemical vapor deposition, spin coating, thermal oxidation, magnetron sputtering [7], molecular beam Epitaxy, sol-gel process and spray pyrolysis [8]. Among them, the chemical spray pyrolysis method is one of the most promising routes due to its low cost, environment friendly. Main objective of present work is to investigate the effect of nozzle to substrate distance on structural, morphological and optical properties of ZnO thin films.

2. Experimental Details

In this investigation, the ZnO thin films were deposited on preheated amorphous glass substrates using P C controlled spray pyrolysis set up supplied by Holmark (Cochin, India). A 0.5 M solution of zinc acetate (AR grade) in a mixed solvent (75 % methanol and 25 % double distilled water) was used as a precursor. Compressed air was used as the carrier gas. The ZnO films were deposited at optimized temperature of 450 ^{0}C by varying the nozzle to substrate distance to 30 cm, 28cm and 26 cm and the deposited films were termed as ZD1, ZD2 and ZD3 respectively. The precursor solution was atomized into the fine droplets and carried to the preheated glass substrates. The various optimized preparative parameters in the film deposition are listed in Table 1.

Table 1. The optimized preparative parameters for the spray deposited ZnO thin films.

Spray parameters	Optimum value / item
Substrate temperature	450 ^{0}C
Molar concentration	0.5 M
Spray rate	3 ml/min.
Spray time	10 min
Solvent	Distilled water
Carrier gas	Compressed air
Gas pressure	1.2 kg/cm^2
Nozzle	Glass

X-ray diffractometer (Rigaku Mini- II) was used to analyze the crystalline orientation and the crystalline plane spacing of the films. The XRD measurements were performed in a standard θ-2θ scan using a Cu kα radiation (λ = 1.5406 Å) over the range of 20^{0} to 80^{0}. The microscopic features were observed through a scanning electron microscope (JEOL-JSM-5600). The optical studies were carried out by using UV-VIS spectrophotometer (Schimatzu Japan) in the 300 to 950 nm wavelength range.

3. Results and Discussions

3.1. Structural Analysis

The XRD patterns of as deposited ZnO thin films with varying nozzle to substrate distance are shown in Fig. 1.

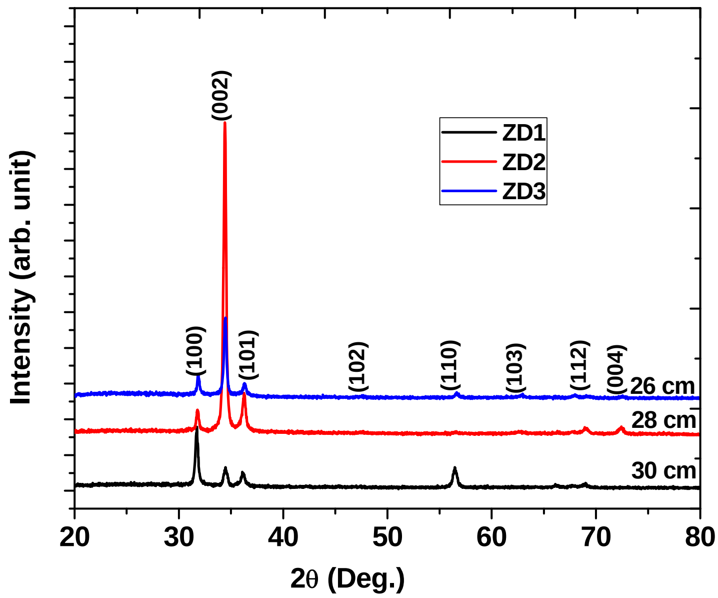


Fig. 1. XRD patterns of as depostied ZnO thin films.

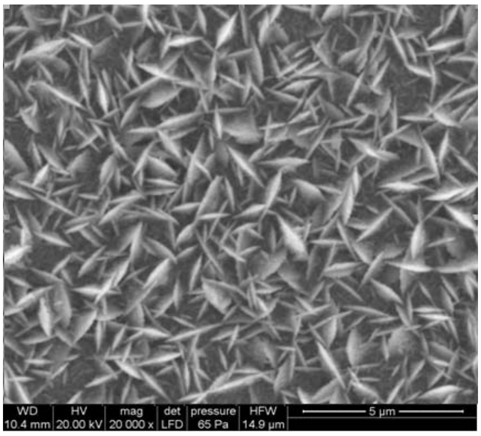
The sharp and intense peaks indicate the samples are polycrystalline in nature. The XRD peaks for (100), (002) and (101) planes showed hexagonal wurtzite structure for as deposited ZnO films. It is evident form XRD patterns that as nozzle to substrate distance decreases from 30 cm to 28 cm, the intensity of (002) peak increases sharply and with further decrease in distance to 26 cm, intensity of (002) peak decreases.

The crystallite sizes (D) calculated using Scheers' formula was found to be in nm range which proves the nanocrystalline nature of ZnO films. Further it is observed that, the crystallite size went on increasing with decrease in nozzle to substrate distance from 30 to 28 cm becomes 27.7 nm and further decreasing the distance to 26 cm, the crystallite size decreased. The films thickness listed in Table 2 shows that thickness increases with decreasing nozzle to substrate distance becomes maximum at 28 cm and further decreased with decrease in nozzle to substrate distance. This trend in thickness supports well the observations marked by XRD analyzed crystallite sizes. The lattice constants 'a' and 'c' were calculated. The observed 'a' and 'c' values are in good agreement with the standard values taken from the Joint Committee of Powder Diffraction Standards (JCPDS) card 75-0576. Typically, 'a' changed from 3.2332 Å to 3.2762 Å and 'c' from 5.1802 Å to 5.2434 Å as nozzle to substrate distance was changed from 30 cm to 26 cm.

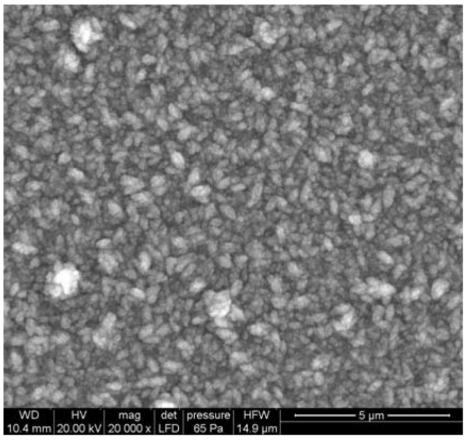
3.2. SEM Studies

Fig. 2a-c represents micrographs of ZD1, ZD2 and ZD3 samples. Micrograph shows a randomly oriented flake like grains with three different grain sizes. Intergranular space is visible along the surface and it contributes to surface roughness. Micrograph of ZD2 for nozzle to substrate distance

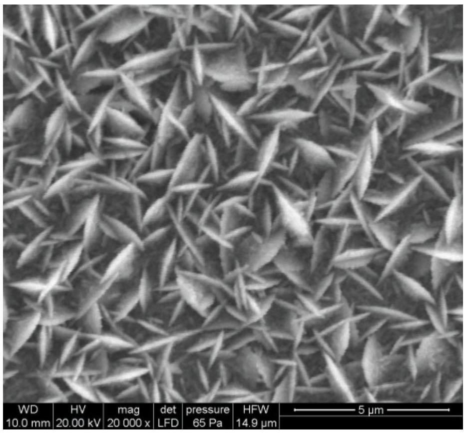
28 cm is found to be consisting of aggregations of small rounded grains with 250 nm average size, separated by small voids with decrease in distance to 26 cm, surface morphology changed and which is more or less similar to that of ZD1 (Fig. 2a). A clear tendency in change of morphology is seen with changing the nozzle to substrate distance. These surface properties have strong impact on the optical properties such as transmittance, absorbance and reflection.



(a) ZD1



(b) ZD2



(c) ZD3

Fig. 2 (a-c). SEM images of ZnO thin films at various nozzle to substrate distances.

3.3. Optical Properties

Fig. 3 shows the optical transmittance spectra of ZnO films, deposited at different nozzle to substrate distances. All the as deposited layers showed good transmittance in the visible region. With decreasing nozzle to substrate distance from 30 to 28 cm, the transmittance increased to 90 % and with further decrease in distance to 26 cm, the transmittance reduced to 80 %. The decrease in transmittance may be due to the enhanced surface scattering, surface roughness and thickness induced absorption [9-10].

The plot of $(\alpha h\nu)^2$ vs. $h\nu$ for all the ZnO samples is shown in Fig. 4. The plot was linear and the extrapolation of the straight portion in graph to the energy axis at $\alpha = 0$, gave the energy band gap. The energy band gap values are tabulated in Table 2. It is observed that band gap is dependent on nozzle to substrate distance.

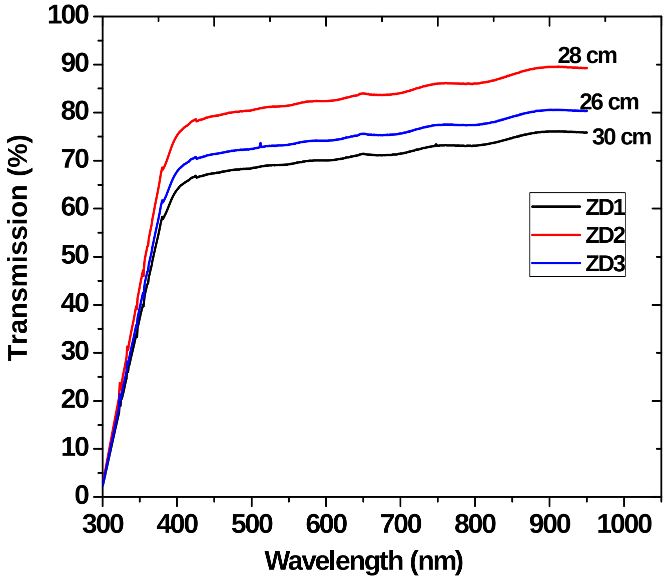


Fig. 3. Optical transmittance spectra of spray deposited ZnO thin films.

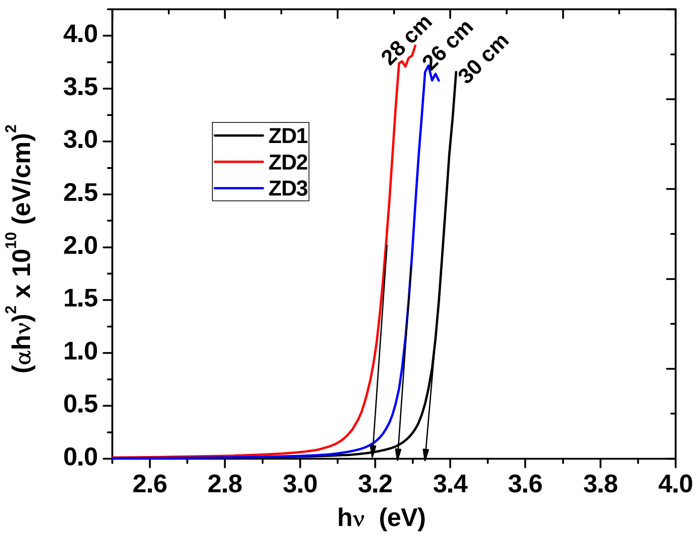


Fig. 4. Variation of $(\alpha h\nu)^2$ versus hν for ZnO thin films.

Table 2. Some physical parameters of spray deposited ZnO thin films.

Sample Code	Thickness (nm)	Cryst. size (nm)	Lattice parameters ZnO (Hex.)			Energy band gap (eV)
			a	c	c/a	
ZD1	410	23.8	3.2332	5.1802	1.602	3.32
ZD2	450	27.7	3.2762	5.2434	1.600	3.19
ZD3	425	25.5	3.2433	5.2078	1.605	3.25

4. Conclusion

The effect of nozzle to substrate distance on structural and optical properties of ZnO is discussed. The crystallite sizes, intensity of 002 plane ad lattice parameters are influenced by nozzle to substrate distance. Morphological studies also showed that intergranular gap and size of grain is altered with nozzle to substrate distance. The optical studies revealed the energy band gap and transmittance are nozzle to substrate dependent.

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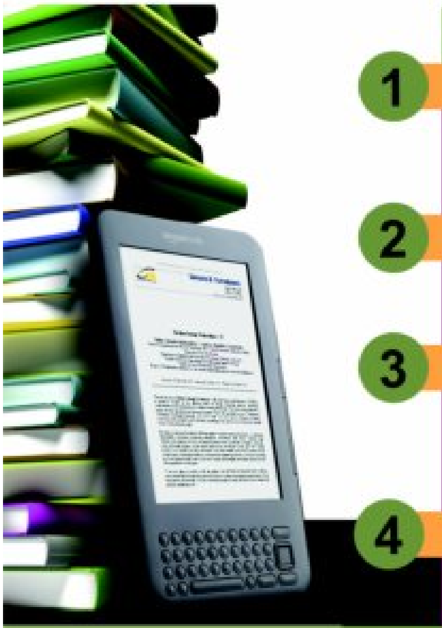

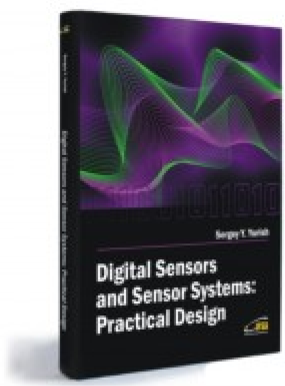
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Digital Sensors and Sensor Systems: Practical Design

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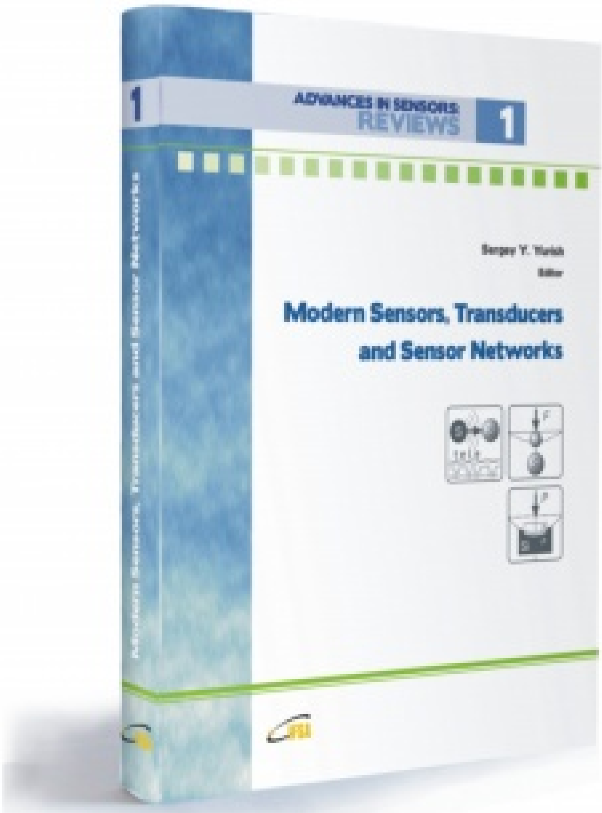

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