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Effect of Thickness on the Properties ZnO Thin Films

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ABSTRACT

Zinc oxide (ZnO) is a promising material in nanotechnology applications, for example in nano-electronics and nano-robotic technology. The objectives of our work are to study the effect of thickness on the structural, optical, electrical and mechanical properties of ZnO thin films. ZnO thin films were coated using chemical bath technique for various thickness. All the coated films were characerized by XRD, FTIR, UV, PL and electrical measurements. The results are discussed herein.

Keywords: Zinc oxide (ZnO), X- ray diffraction (PXRD), nanostructure

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INTRODUCTION

[4-JP]ZnO is a versatile (opto)-electronic material with direct band gap of 3.37 eV and a large exciton energy of 60 meV, which attracted a lot of attention recently. The high exciton binding energy in Zno crystal can ensure efficient excitonic emission at room temperature [1]. Most prominent crystalline structure of ZnO is wurtzite type, although, it also exists in the cubic zinc blende and rocksalt structures.In wurtzite type, each Zn ion is surrounded by a tetragonal coordination. This give rise to polar symmetry along the hexagonal axis, which is responsible for a number of properties of ZnO.Thin films of Zno have been widely used in transparent electrodes, surface acoustic wave devices, field effect transistors and display devices**[2].**Nano scale porous structures of ZnO with high surface area find applications in chemical sensors and solar cells**[3].** However obatining ZnO films with superior optical and electrical properties suitable for device applications is still a technological challenge. Several new, approaches, use of variety of substrate and experimental conditions are enabling a wide range of optical and electrical properties of ZnO thin films. Flavio Leandro et al **[4]** found that the film thickness has a strong influence on the optical absorption of nanostructured α-Fe_2O_3 nano thin films. They reported that, stress induced due to increased thickness, generates defects in the crystal lattice of the hermatite film, which in turn increases the e-h recombination process. The use of photochemical cells in solar energy conversion has gained importance in the last few decades **[5]**. ZnO is also an attractive material for PEC cell for water splitting mainly due to its high electro chemical stability. Moreover defect induced ZnO can act as a best solar absorbing material with higher effeciency.In view of this in our present work, we made an attempt to prepare ZnO nano structured thin films with various thickness.

MATERIALS AND METHODS

2.1 Prepartion of ZnO thin Films

A traditional method of preparing ZnO nano particles is achieved using chemical bath techniques. Zinc chloride and NaOH were used as the starting materials. Ammonium Solution was used to maintain the PH at 9.ZnO was prepared by double distilled water. Heating the liquid phase at 60^{o}C with continuous stirring yield the ZnO particle in average nano meter size. Now the cleaned glass substrate is immersed in above processed sol to coat over the substrate. This method is based on the sequence of reaction on the substrate surface. ZnO is formed by the following reaction

$(CH_3COO)_{2Zn} \longrightarrow 2CH_3COO^- + Zn^{2+}$

$NH_2\text{-}CO\text{=}NH_2 + H2O \longrightarrow O^{2-} + H_2O$

$Zn^{2+} + O^{2-} \longrightarrow ZnO$

The coating time is varied from 30 mins to 3 hrs in order to vary the thickness of the ZnO films. Following drying and heat treatment is to generate porous structured films on the substrate. Residual organics are removed by washing the films with acetone. This method does not require sophisticated and expensive instruments. Also by this method we have prepared pin-hole free thin films.

2.2 Characterizations Made

The average thickness of the coated films were measured by Semi-Consoft in thickness measuring unit.As synthesized ZnO powder was subjected to powder X- ray diffraction (PXRD) analysis, using an X- ray powder diffractometer, PAN alytical with scintillation counter and monochromated Cu K_α (λ = 1.54056 Å) radiation.. In order to understand the chemical compositions of the prepared sample in the present study, we have carried out EDS measurements using JEOL SEM Model, JSM – 5610LV scanning electron microscope .The optical absorption spectra of coated films were recorded in the range of 200-800 nm using Consoft Inc **Spectrophotometer.**

RESULTS AND DISCUSSION

3.1 Structural Analysis

In order to confirm the material of the grown film and to determine the particle size, powder X-ray diffraction (PXRD) data were collected for the powder scratched from the film. The observed PXRD data were indexed by matching with the data available for ZnO Grain or cluster sizes were determined using the Scherrer formula.

$$D = \frac{K\lambda}{\beta \cos \theta},$$

where D is the mean diameter (size) of the grains, K (=0.9) is the size factor, β is the full width at half maximum (in radians), λ is the wavelength of the X-radiation used and 2θ is the angle at which the maximum intensity was observed. The grain size obtained in the present study was 8.621nm which confirm the nanostructure of the synthesized material. The energy peak at XXX eV confirms the prepared material as ZnO.

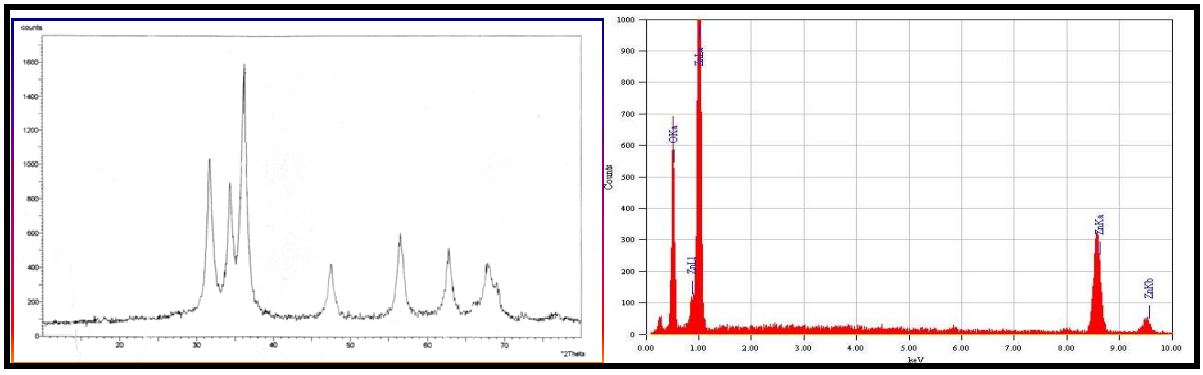

Figure 1: PXRD and EDAX spectrum of ZnO nano powder

3.2 Thickness Measurements

Thickness and coating time for all the prepared samples are tabulated in Table 1.

As the time increases, the thickness also increases.

Sample code	Coating time (min)	Thickness(nm)
Film1	30	**25.321**
Film2	60	**55.525**
Film3	90	**78.230**
Film4	120	**141.615**
Film5	150	**161.267**

3.3 AFM Measuremnts

In order to understand the surface features and size distribution of prepared films in the present study, we have carried out AFM analysis using a Shimadzu atomic force microscope.AFM images of the 5 films are shown in figure .The surface roughness decreases with increasing thickness. Also it confirms the grain size obtained from XRD patterns.

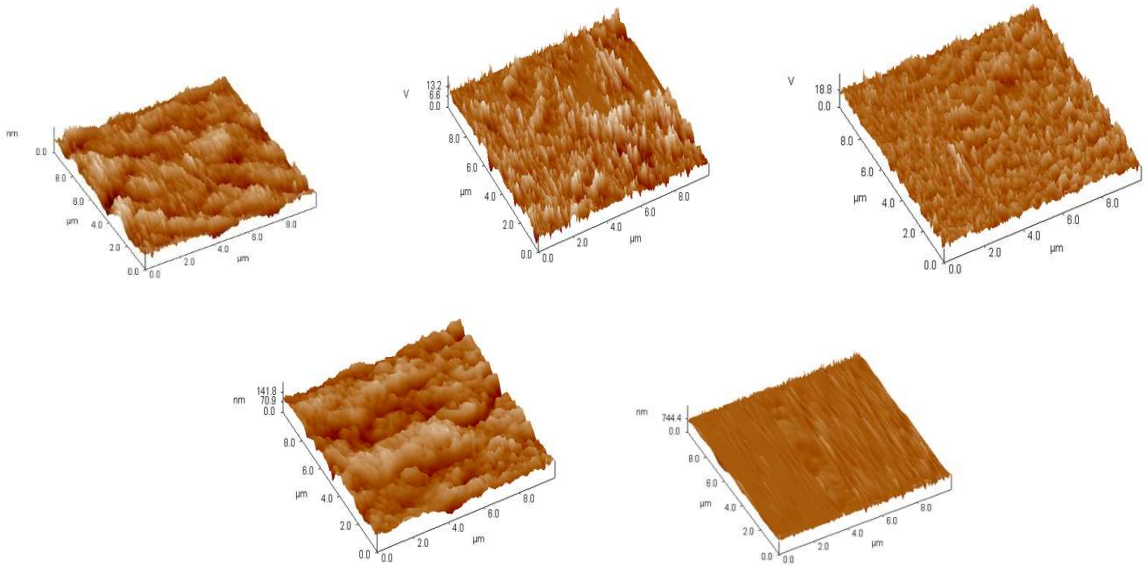

Figure 2: AFM images of ZnO thin films

3.4 UV-spectral Measurements

Optical characterization of thin films gives information about other physical properties, e.g. band gap energy and band structure, optically active defects etc., and therefore may be of permanent interest for several different applications. The spectra were recorded with medium scan speed and sampling interval 0.5 in the wavelength range190-800 nm. The UV-Vis spectrum is shown in Figure

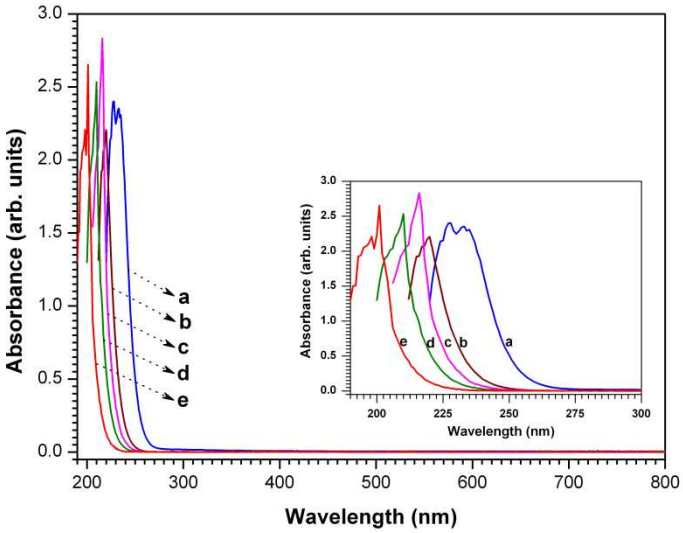

Figure 3: UV-Vis spectra of ZnO thin Films

Plot for determining optical bandgap from UV-Vis absorption data of ZnO films with variable thickness are shown in Figure3. The fundamental absorption, which corresponds to electron excitation from the valence band to conduction band, can be used to determine the value of the optical bandgap. The absorption coefficient α was determined using the relation,

$$\alpha = \frac{2.303 \log (\text{absorption})}{\text{Thickness of the sample}}. \qquad \dots\dots\rightarrow \qquad (3.1)$$

The relation between the absorption coefficient (α) and the incident photon energy (hυ) can be written as **[7]**:

$$(\alpha h\upsilon) = A\left(h\upsilon - E_g\right)^n, \quad \text{.....} \rightarrow \quad (3.2)$$

Where, A is a constant, E_g is the bandgap energy of the material and exponent n depends on the type of transition **[8]**. The value of optical bandgap was calculated by extrapolating the straight line portion of $(\alpha h\upsilon)^{½}$ *vs* hυ graph to hυ axis. The values obtained for the optical band gap is given in table 2.

Sample name	Optical band gap energy eV	Cutoff wavelength nm
Film 1	4.94	251
Film 2	5.31	234
Film 3	5.46	227
Film 4	5.60	222
Film 5	5.87	211

As the thickness increases, the band gap also increases which in turn increases the optical absorption.

3.5 Photoluminscence Measurements

All the five films prepared in the present study exhibit five prominent peaks (two peaks coupled) in the PL spectra. The peaks are observed at around 408, 446, 486, 490 and 530 nm. The PL spectra indicate clearly that the films have very high PL yield.

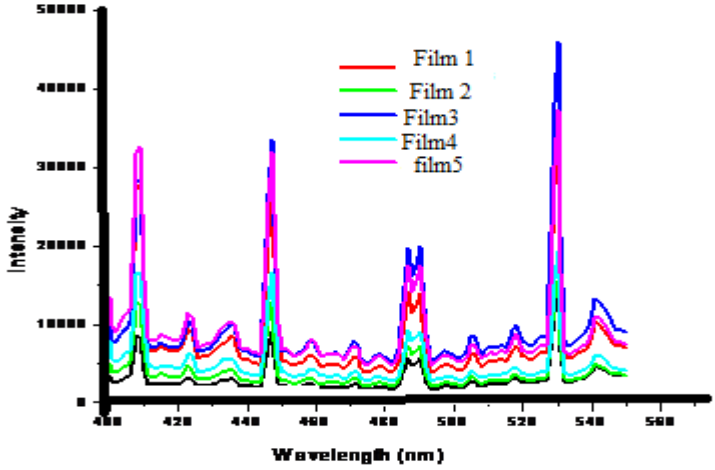

Figure 4 : PL spectra of ZnO thin films for various thickness

3.6 Photo conductivity Measurements

With increasing thickness of the ZnO films both the conductivities (I_d and I_p) increase linearly with the applied field, but I_p is found to be greater than that of I_d **[9]**. This indicates the positive photoconductive nature of all the fourteen systems considered in the present study **[10]**. The positive photoconductivity, in general, is attributed to the generation of mobile charge carriers caused by the absorption of photons **[11].** If the effect of light is primarily either to increase the density of free carriers or to decrease the resistance of barriers in the given material, then photoconductivity results **[12]**. The action of light in this case is to decrease the height of the barrier, thus permitting greater charge carrier flow between different regions of the photoconductor **[13]**.

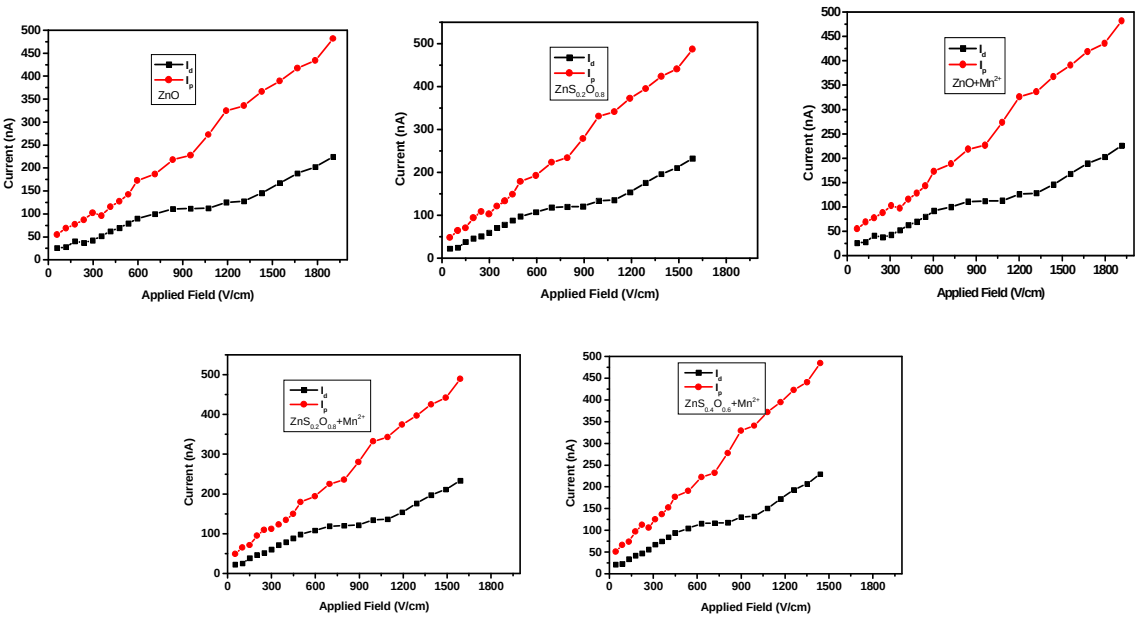

Figure 5: Photo conductivity of ZnO thin films

3.7 Dielectric Measurements

The measurement of dielectric constant and dielectric loss as a function of frequency and temperature is of interest both from theoretical point of view and from the applied physics **[14]**.

The ε_r values obtained for the thinner film is very small when compared to that observed for thicker one **[15]**. All the three electrical parameters increase with the increase in temperature for all the five films **[16].**

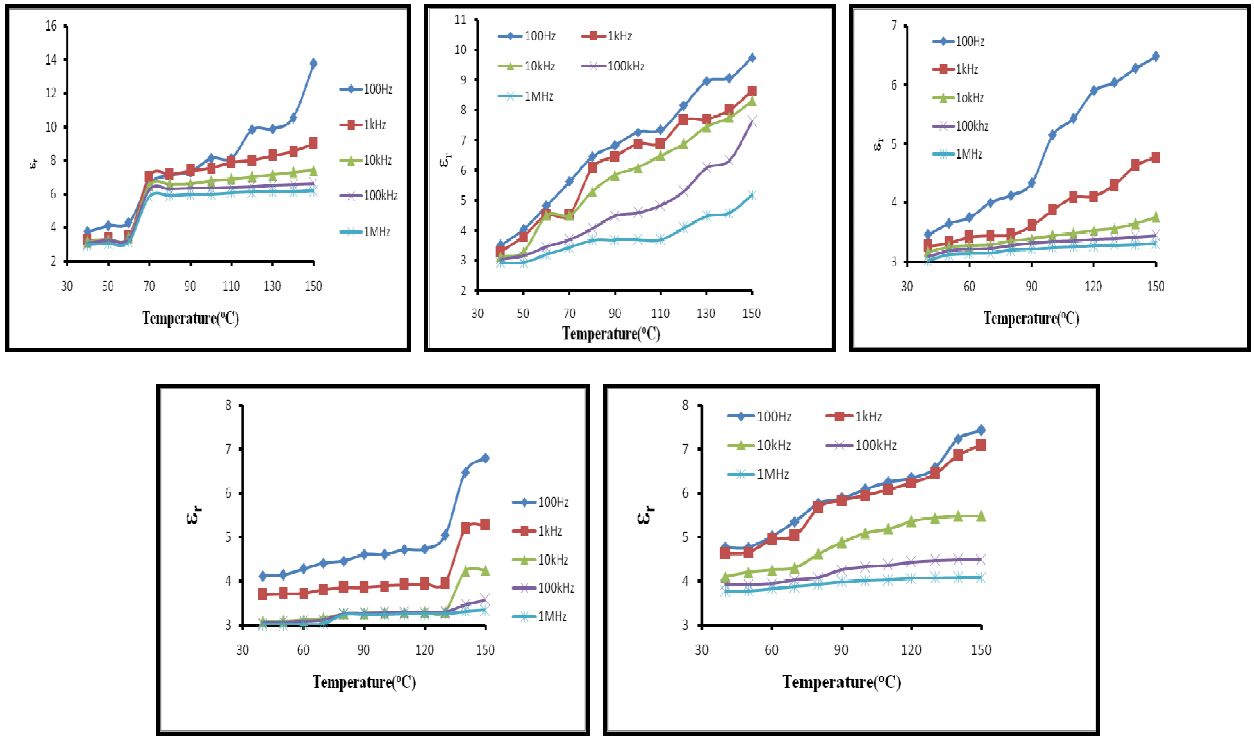

Figure 6: Variation of dielectric constant with temperature and thickness for various frequencies

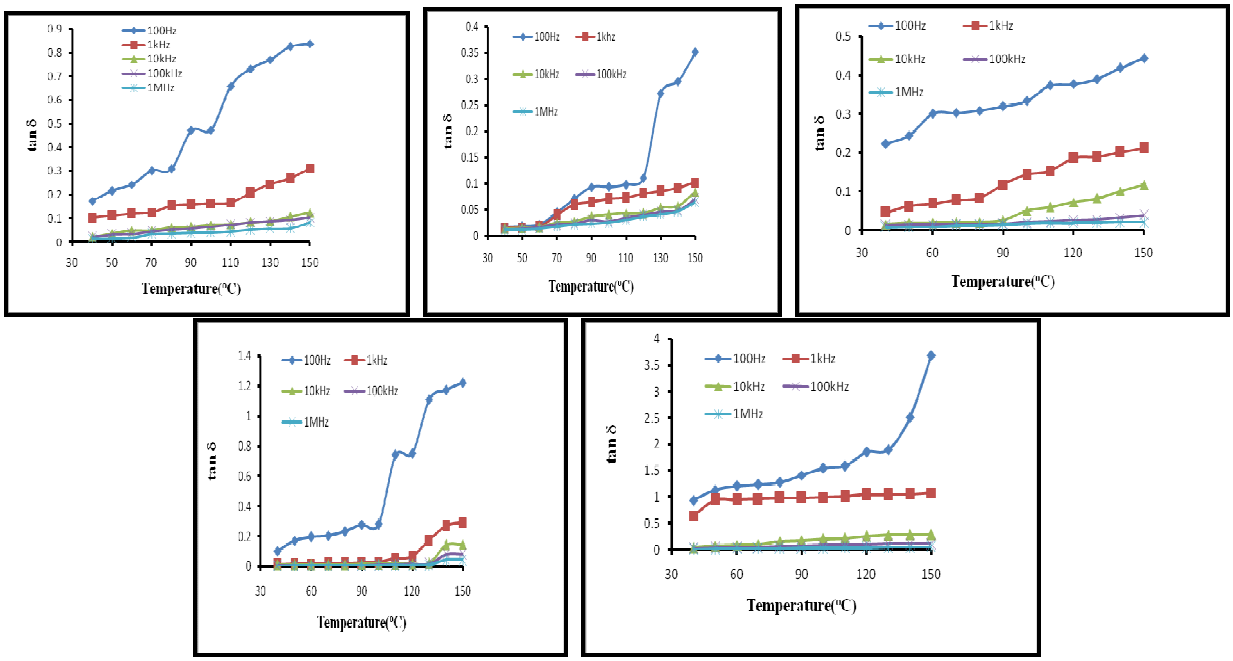

Figure 7: Variation of dielectric loss factor with temperature and thickness for various frequency

CONCLUSION

The feasibility of ZnO thin film nanostructure preparation in an aqueous medium by a simple chemical bath technique was demonstrated. As the coating time increases linearly, thickness increases with nonlinear variations. XRD spectra show that, all the five films belong to wursite structure also thickness variation produces change in intensity of the diffracted beam. The UV spectral measurement confirms that higher thickness film has higher band gap due to more electron – hole production. But the thinnest films show low ε_r value.

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