Study of the Structural Properties of Sol-Gel Deposited Zinc Oxide Thin Film

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Abstract: The present paper is study of structural properties of Zinc oxide. ZnO is an attractive material for applications in electronics, photonics, acoustics, and sensing. Thicknesses of the thin films were varied by increasing the number of deposited layers. As part of our characterization process, XRD and FE-SEM were used to characterize the structural properties

Keywords: *Zinc Oxide, Thin film, Structural properties, Sol-gel*

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I. INTRODUCTION

Recently, wide band gap semiconductors have actively been studied for their application in optoelectronic devices. GaN and ZnO are examples of wide band gap semiconductors which have been used for light emitting diodes and laser diodes. ZnO is mostly chosen for its better excitonic properties compared to GaN, with an excitonic binding energy of 60 meV at room temperature [1]. Besides that, because of its high transparency in the visible region and its high conductive properties, ZnO is widely used as a transparent conducting oxide (TCO), in photovoltaic devices and sensors. ZnO is naturally n-type due to defects such as oxygen deficiency and zinc excess [2]. It is chemically and mechanically stable, non-toxic, and high abundant.

Various deposition methods have been applied to grow high quality ZnO thin films such as molecular beam epitaxy (MBE) [3], thermal chemical vapor deposition (CVD) [4], metal-organic cvd (MOCVD) [5], r.f. sputtering [6], and sol-gel methods [7]. Among the methods, sol-gel spin coating is favorable because of its simple, non-expensive preparation of homogeneous thin film, along with excellent compositional control, lower crystallization temperature and uniform film thickness [8]. In order to produce high quality ZnO thin film, it is necessary to study the parameters that can affect the properties of the thin films, such as pre and post-heating temperature, concentration of the sol, and thickness of films. Among these factors, films thickness is crucial as it is directly related to the resistivity of the ZnO thin film. The crystallites' size and orientation which depend on the film thickness play a big role in controlling the electrical conductivity of the polycrystalline ZnO film [9].

II. MATERIALS AND METHODS

In present study MILMAN Spin coater Model 2000 S Spinner with speed range 100-10,000 rpm and spinning duration up to 120 seconds was used in this research. The coating solution will be dropped onto glass substrates, which were rotated speed of 3000 rpm for 60 seconds by using spin coater. Spin coating is a typical process that has been used for several decades as a method for deposition of thin films [11-12]. A typical spin process used for deposition of thin films consists of four steps namely. a) Substrate loading; b) Dispense liquid; c) Spinning; d) Drying. Initially pre-cleaned substrate on a substrate holder of spin coater was loaded. Afterwards a small amount of solution onto the substrate surface was dispensed. Deposition by Static dispense was simply a small puddle of fluid on or near the center of the substrate. That could range from 1 to 10 cc depending on the viscosity of the fluid and the size of the substrate to be coated. After the dispense step, the substrate were then accelerate to form a thin films with desired thickness at relatively high speed. The combination of spin speed and time selected for this step was generally defined the final film thickness. In general, higher spin speeds and longer spin times create thinner films. Finally prepared film was dried. The process from coating to annealing was repeated 2 to 6 times to produce different thickness of films [13-15].

III. RESULTS AND DISCUSSION

The thicknesses of the films resulting from the 2 to 6 repetitions of spin coating and annealing were found to be 14.3 + 0.2 nm, 20.9 + 0.3 nm, 42.8 + 0.5 nm, 51.5 + 0.4 nm, and 61.7 + 0.3 nm. The XRD pattern of the ZnO films for various film thicknesses are shown in Figure 4.1. All the peaks of the ZnO films correspond to the peaks of standard ZnO (JCPDS #36-1451). Many researchers have produced

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highly c-axis oriented ZnO thin films using sol-gel methods using MEA as a stabilizer [16, 17].

However, here it was observed that there were three main peaks in the XRD pattern, which are (1 0 1), (0 0 2) and (1 0 0), which indicates that the films were polycrystalline and randomly oriented. According to Xu et. al [18], the strain of the film cannot be eliminated during a 30 minute annealing duration, so the crystal quality of the thin films were not as good as reported by those who used MEA stabilizers with longer annealing time. But because of the layer-by-layer application method, the intensity of the peak increased with the film thickness, which suggests that the quality of the thicker film improved.

FE-SEM micrographs of the ZnO thin films deposited are shown in Figure 4.2. From the micrographs, it is observed that nanoparticle structures were formed in the films, which is a normal characteristic of films derived from sol-gel [19, 20]. The average grain sizes measured from the FE-SEM micrographs were found to be increase with the film thickness. It was also observed that the grains become more uniform and dense as the thickness increased.

Table 1: values of hkl, 2θ and d for prominent peak of Zno thin film

hkl	2θ	d(A^0) (XRD)	d(A^0) (PDF)
100	31.9	2.799	2.814
002	34.4	2.597	2.603
101	36.3	2.471	2.476
110	56.6	1.623	1.624

Table 2: Film code, number of repeats and average thickness

Sr. No	Film Code	No. of repeats	Average thickness
I.	a	2	14.3 nm
II.	b	3	20.9 nm
III.	c	4	42.8 nm
IV.	d	5	51.5 nm
V.	e	6	61.7 nm

The arrangements of atoms in further layers get gradually modified by preceding layers. As a result, the growth takes place in a more definite way, resulting in a particular type of preferred growth, and differences in the intensities of peaks when the substrate temperature or other production conditions are changed are observed [21, 22].

The lattice constants of ZnO film can be calculated from the most prominent peaks. The lattice constants for the hexagonal structure are taken as a=b≠c. The a, b, and c lattice constants of ZnO with the hexagonal Wurtzite structure were 0.32316 nm (a = b), 0.51943 nm, respectively. It is clear that the calculated lattice constants are found to be in good agreement with the PDF standards for ZnO powder [23]

The grain size (D) and dislocation density (δ) are calculated for the preferential orientation to obtain information about its crystalline level. The broadening of the full width at half maximum is inversely proportional to the grain size according to Scherrer's formula. The grain size D can be calculated with the following equation [24]

$$D = \frac{0.9\,\lambda}{\beta\ \mathrm{Cos}\ \theta}$$

With the angle position of the peak (δ) and the FWHM (β). In Equation (1), λ is the wavelength of the incident X-ray. The value of the crystallite size is estimated from Scherrer's formula (Eq. 1) as 15.103 nm. Our calculated value is nearly the same value as in the literature [25–28].

The dislocation density (δ) is described as the length of dislocation lines per unit volume of the crystal [29]. The dislocation density of the crystal gives information about the crystal structure. The dislocation density for preferential orientation can be calculated using the formula given below [30],

$$\delta = \frac{1}{D^2}$$

The dislocation density is obtained from Equation (2) as 4.3916 ×10−3 (nm)−2. The smaller the dislocation density is the better the crystallization of the film. It is found to be in well agreement with previous study of S. Aydogu et al. [31].

IV. CONCLUSION

Thicknesses of the thin films were varied by increasing the number of deposited layers on glass substrate. Because higher spin speeds and longer spin times create thinner films. FE-SEM micrographs showed that nanoparticle structures were formed in the films, which is a normal characteristic of films derived from sol-gel.

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