International Journal of Trend in Scientific Research and Development (IJTSRD)
Volume 9 Issue 1, Jan-Feb 2025 Available Online: www.ijtsrd.com e-ISSN: 2456 – 6470

Magnesium-Doped ZnO Films by Sol-Gel Spin Coating Method

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ABSTRACT

Undoped and Mg doped ZnO films were deposited on glass substrates by sol-gel spin coating method. This method is a simple, economical and effective method of making high quality films. The structural, morphological and optical properties of the films were investigated using X-ray diffraction (XRD), field emission scanning electron microscopy (FESEM) and UV-visible spectroscopy, respectively. The structure, surface morphology, optical transmittance and band gap were found to be dependent on the Mg content of the film. The XRD results showed that the films were of polycrystalline nature and FESEM images proved that the films were homogenous and compact. It was observed that the band gap increases with Mg doping.

KEYWORDS: *Sol-gel method, Mg, ZnO, Films, Doping etc*

How to cite this paper: Awadhesh Kumar | Umakant Prasad "Magnesium-Doped ZnO Films by Sol-Gel Spin Coating Method" Published in International Journal of Trend in Scientific Research and Development (ijtsrd), ISSN: 2456-6470, Volume-9 | Issue-1, February 2025, pp.1086-1089, URL: www.ijtsrd.com/papers/ijtsrd76187.pdf

IJTSRD76187

1. INTRODUCTION

Zinc oxide (ZnO) is one of the most important materials of II-VI semiconductors with an energy band gap of 3.3 eV (Bhattacharya et al. 2004) and large exciton binding energy of 60 meV (Caglar et al. 2010). ZnO is widely used in a wide range of technological applications. Examples are solar cells (Minemoto et al. 2000),light emitting diodes (Saito et al. 2002), photodetectors (Liang et al. 2001), etc. Mg doped ZnO films made by alloying ZnO with MgO showed that the band gap energy varies with Mg concentration (Cho et al. 2011). The band gap of MgZnO alloys can be tuned from 3.3 to 4 eV by doping ZnO with Mg(Ohtomo et al. 1998). Due to the similar ionic radius of Mg2+ and Zn2+, substitution of Zn by Mg does not significantly change the lattice structure (Caglar et al. 2010). MgZnO films with wide optical band gap feature have many advantages for electro-optical application such as coatings of light emitting diodes (LEDs), buffers for thin film solar cell (TFSC) devices and TCO layers (Kaushal et al. 2009 and Shimakawa et al. 2008). MgZnO alloy is an excellent material system for potential application to exciton-related photonic devices in the UV region (Wang et al. 2008). MgZnO films can be prepared by many methods such as RF magnetron sputtering (Minemoto et al. 2000), pulsed laser deposition (Shrama et al. 1999), ultrasonic spray pyrolysis (Zhang et al. 2005), sol–gel method (Moon et al. 2013), etc. In this study, undoped and Mg doped ZnO films were deposited by sol–gel spin coating method. The structural and optical properties and surface morphology of the films were investigated.

2. Experimental Details

Undoped and Mg doped ZnO films were deposited on glass substrates by sol-gel spin coating method. Zinc acetate dihydrate for ZnO and magnesium acetate tetrahydrate for Mg doping were used as starting precursors. 2-Methoxyethanol and monoethanolamine (MEA) were used as solvent and stabilizer, respectively. The concentration of the solution was 0.5 M and the concentration of Mg doping was 25%. The resulting solution was stirred at 60° C for one hour on a magnetic hot plate. After preparing the solution, the undoped ZnO solution was dropped on the glass substrate which was rotated at 3000 rpm for 30 seconds and the Mg doped solution was dropped on the glass substrate which was rotated at 3500 rpm for 30 seconds using a spin coater. Both films were dried at 300°C for 10 minutes. This process was repeated 9 times and then the films were kept in 500°C air for 2 hours and then some of their physical properties were compared.

3. Results and Discussion

3.1. Structural and morphological properties

The crystal structure and preferred crystal orientation of undoped and Mg doped ZnO films were investigated by X-ray diffraction (XRD) measurements. XRD measurements were carried out by Panalytical Amperion X-ray diffractometer using CuKα (λ=1.5405 Å) radiation in the 2θ range 30o - 80o with a scanning speed of 2o/min. Diffractometer images of the films were taken at room temperature. An X-ray tube operated at 45 kV and 40 mA. Figure 1 shows the XRD patterns of undoped and Figure 2 shows the XRD patterns of Mg doped ZnO films on glass substrate annealed at 500 C in air for two hours. Figure 3 compares the XRD patterns of undoped and Mg doped ZnO films. The sequencing of the patterns is according to hexagonal (wurtzite) ZnO and cubic MgO. The diffraction peak of hexagonal ZnO and the peak associated with cubic MgO are indexed based on ICDD (International Centre for Diffraction Data): 98-003-1052 and ICDD: 98-064-2712, respectively. The XRD spectra for undoped and Mg doped ZnO films indicated that the films are of polycrystalline nature. As can be seen in Figure 2, the Mg doped ZnO film shows a distinct (002) MgO peak, indicating that these films generate segregation phase structure. The surface morphology of the films was studied by FESEM (Zeiss Supra 40VP). Figure 4 shows FESEM images of undoped and Mg doped ZnO films. As can be seen in Figure 4(a), the shape of the ZnO film structure is hexagonal and the surface morphology is homogenous and dense. The doped Mg structures can be seen in Fig. 4(b). Similarly, the surface morphology is also uniform.

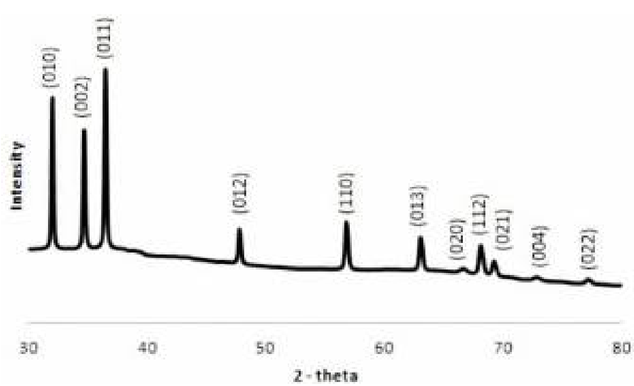

Figure 1. XRD pattern of undoped ZnO film

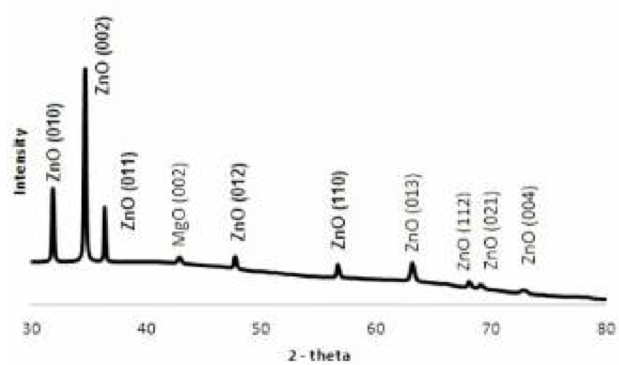

Figure 2. XRD pattern of Mg doped ZnO film

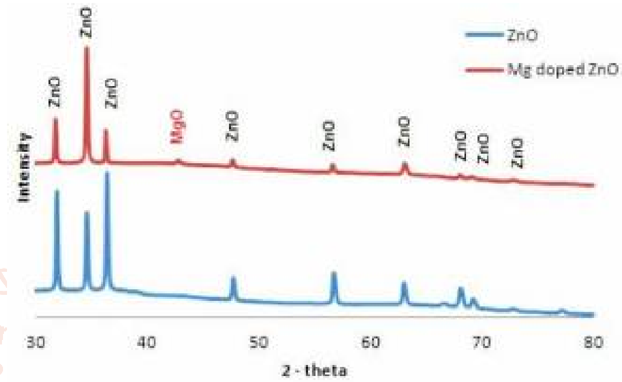

Figure 3. XRD pattern of undoped and Mg doped ZnO films

3.2. Optical properties

Absorption studies were carried out in the UV-Vis region at room temperature to determine the optical band gap of the films. The UV-Vis spectroscopy measurements were analyzed by a Perkin Elmer Lambda 25 UV-Vis spectrometer between 300–1100 nm wavelength. The absorption spectra of undoped and Mg doped ZnO films can be seen in Figure 5. The fundamental absorption edge of the films corresponds to the electron transition from the valence band to the conduction band and this edge can be used to calculate the optical band gap of the films. Undoped and Mg doped ZnO films have a direct transition band gap. In the direct transition, the absorption coefficient can be expressed as (Pankov 1971):

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

where A is a constant, hυ is the photon energy and Eg is the optical band gap. Plots of $(\alpha h\upsilon)^2$

vs. hυ are shown in Figure 6. The E_g values of the films were calculated from these plots and are given in Table 1. As can be seen from these plots, the E_g values of the films increased with Mg doping.

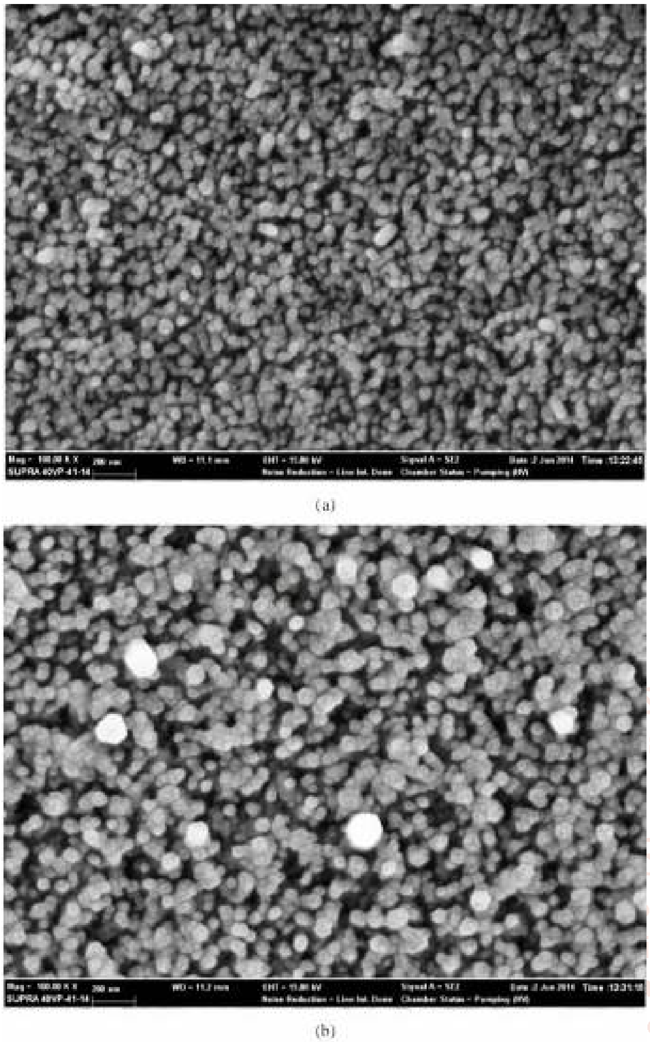

Figure 4. FESEM images of (a) undoped ZnO (b) Mg doped ZnO films

Table 1. Energy band gap values

Films	E_g (eV)
ZnO	3,32
Mg doped ZnO	3,48

Optical transmittance measurements were analyzed by Perkin Elmer Lambda 25 UV-Vis spectrometer between 300-1100 nm wavelength. The optical transmittance spectra of the films are shown in Figure 7. The transmittance value of the films decreased with Mg doping.

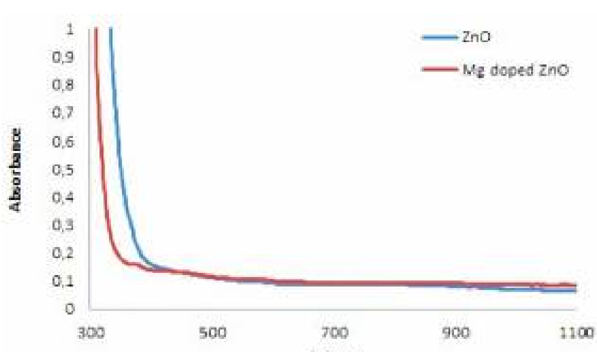

Figure 5. Absorbance spectra of undoped and Mg doped ZnO film

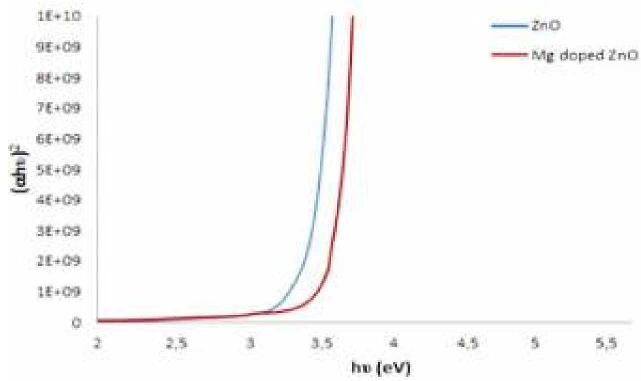

Figure 6. The plots of $(\alpha h\upsilon)^2$ vs. hυ of undoped and Mg doped ZnO film

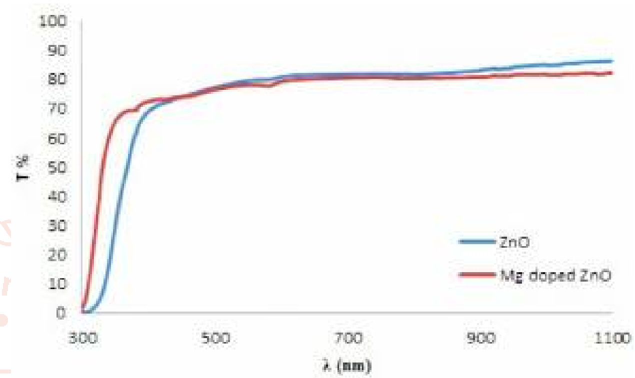

Figure 7. Optical transmittance spectra of undoped and Mg doped ZnO film

4. Conclusion

Undoped and Mg doped ZnO films were deposited on glass substrates by sol-gel spin coating method. The effect of Mg doping on the structural, morphological and optical properties of these films was investigated. XRD spectra indicate that the films are of polycrystalline nature. In the XRD spectra of Mg doped ZnO films, Mg is segregated as MgO(002). FESEM images clearly show the homogenous and compact nature of the samples. Moreover, doped Mg structures can be observed. The band gap has been successfully tuned from 3.32 to 3.48 eV with Mg doping. This band gap tuning is well acknowledged by the shift of the (002) peak in the XRD spectra. Films prepared by this method are easy to prepare, low cost and effective, hence these can be a good candidate for many applications.

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