EFFECTS OF MG INCORPORATION ON X-RAY DIFFRACTION, U-VAND PHOTOLUMINESCENCE PROPERTIES OF ZNO THIN FILMS PREPARED BY SOL-GEL METHOD

Mrs.Seema Singh*

Mrs.Maninder Kaur*

ABSTRACT

As Well-crystallized MgZnO alloy thin films with hexagonal wurtzite structure were fabricated by sol-gel method. With the band gap increases, the surface roughness and the grain size reduces. It is worth noting that the intensity of the band-edge luminescence of Mg doped films enhances with the increase of the Mg content. The microstructure and photoluminescence mechanism have been discussed based on X-ray diffraction patterns, atomic force microscopy images, ultraviolet-visible absorption spectra, photoluminescence spectra and Fourier transform infrared spectra.

KEY WORDS: MgZnO thin film; Optical properties; Sol-gel method.

* Dept.of ECE, Doaba Institute of Engineering and Technology, Kharar, Mohali, Punjab,India.

1. Introduction

Nowadays, zinc oxide has gained much interest because of its potential multifunctional applications ranging from transparent electrodes in solar cells[1], thin film gas sensors[2], spintronic devices[3], photo detectors[4], and surface acoustic wave devices[5], to light emitting diodes[6], based on its wide direct band gap (»3.34 eV) and large exciton binding energy (»60 meV) at room temperature. In order to extend its application in broader wave-length region, ones have attempted to introduce proper ions such as Cd and Mg into ZnO lattice to adjust the energy band structure. Until now, MgZnO thin films have been prepared by many techniques such as spray pyrolysis, sputtering, pulsed laser deposition (PLD), and sol-gel process. The reports have demonstrated that the ternary alloys with a wide range of Mg2+from 0 to 36% still maintained the hexagonal lattice structure due to the similar ionic radius of Mg2+ (0.057 nm) and Zn2+ (0.060 nm), and the corresponding energy band gap could be increased from 3.34 eV to 3.96 eV. Moreover, the MgZnO thin films could produce a bright ultraviolet luminescence at room temperature due to the band edge exciton recombination. This makes the MgZnO alloy thin film acts as an excellent candidate for ultraviolet opto-electronic devices such as UV laser and UV detector.

Among the fabrication methods mentioned above, the sol-gel technique has many advantages, such as simplicity, low cost, and excellent compositional control. In this study the MgZnO thin films were success-fully prepared by sol-gel technique. Besides typical X-ray diffraction (XRD), photoluminescence (PL) and ultraviolet-visible (UV-vis) absorption analysis. Which is beneficial to understanding the chemical composition of the MgZnO thin films? Based on the FT-IR spectra, a new and simple N incorporation method might be found.

2. Experimental

The MgZnO thin films were prepared by sol-gel method. At first, Zn (CH3COO)2.2H2O and Mg(CH3COO)2.4H2O were dissolved together in methanol (CH3OH) at room temperature up to 15-20 min, stir the solution until get transparent solution and then the diethanolamine (HN(CH2CH2OH)2, DEA) was added to the solution as the stabilizer. The total concentration of the metal ions was controlled at 0.5 mol/L in the solution, and the molar ratio of the DEA to the total metal ions was 1:1. The Mg2+ concentration was chosen as 0.00, 0.05and 0.10 mol/L for different samples (1A, 2A, 3A,), respectively. The solution was stirred for 1 h at 60ºC by a ultra -

sonic stirrer until a clear and transparent solution was obtained and then spin-coated onto Si - substrates, with rotating rate of 3000 r/min. The as-coated film was immediately pre-heated at 150 º C to evaporate the solvent and other organic component in the film. Finally, the coated substrates were annealed in a tube furnace at 500ºC for 5 h in air.

X-ray diffraction (XRD) analyses were conducted to determine the crystal structure of the MgZnO alloy thin films by using an X-ray diffract meter (XD-3A, Shimadzu) using Cu$K\alpha$ radiation at 40 kV and 30 mA. The AFM photographs of the MgZnO thin films were given by Molecular Imaging, SPM 3000. Optical absorption spectra were examined by using a spectrophotometer (UV-3600, UV-VIS-NIR Spectrophotometer, Shimadzu). PL measurements were performed by employing spectrometer.

3. Results and Discussion

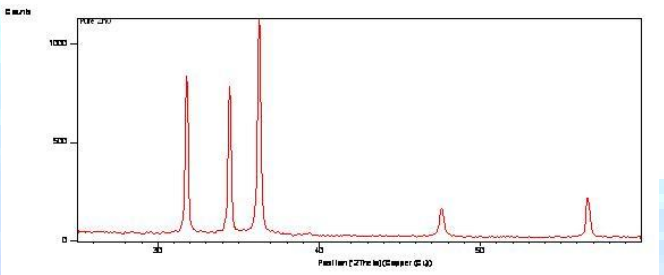

Fig. 1 XRD Pure ZnO (1A)

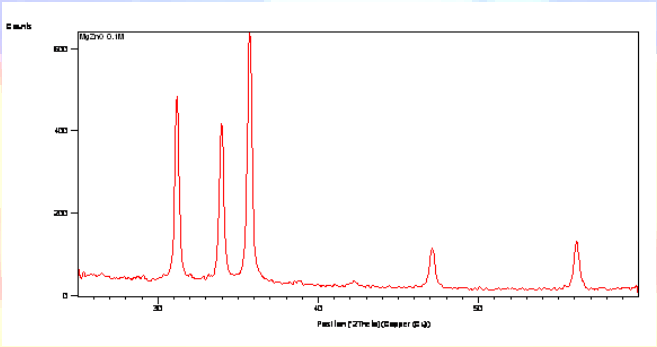

Fig.2 XRD ZnO DOPED WITH Mg (0.05M) (2A)

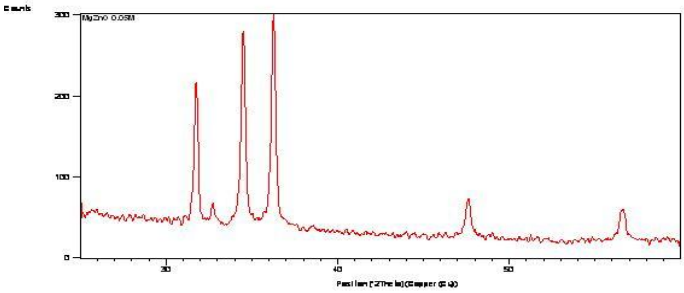

Fig.3 XRD ZnO DOPED WITH Mg (.1M) (3A)

Measurement Conditions: ***(Bookmark 1)***

Dataset Name Pure ZnO

File name C:\X'Pert Data\OCT2011\Pure ZnO.xrdml

Comment Configuration=Flat Sample Stage, Owner=jagtar, Creation date=6/11/2007 3:57:00 PM Goniometer=PW3050/60 (Theta/Theta); Minimum step size 2Theta:0.001; Minimum step size Omega: 0.001 Sample stage=PW3071/xx Bracket

Diffractometer system=XPERT-PRO

Measurement program=PU, Owner=jag tar, Creation date=4/15/2008 1:52:59 PM

Measurement Date / Time 13/05/201212:51:13 PM

Operator Punjab University

Raw Data Origin XRD measurement (*.XRDML

Scan Axis Gonio

Start Position [°2Th.] 25.0084

End Position [°2Th.] 59.9774

Step Size [°2Th.] 0.0170

Scan Step Time [s] 40.7048

Scan Type Continuous

PSD Mode Scanning

PSD Length [°2Th.] 2.12

Offset [°2Th.] 0.0000

Divergence Slit Type Fixed

Divergence Slit Size [°] 0.957

Peak List: (Bookmark 3)

Pos. [°2Th.]	FWHM [°2Th.]	d-spacing [Å]	Rel. Int. [%]	Area [cts*°2Th.]
31.7700	0.1752	2.81431	73.63	141.97
34.4204	0.1805	2.60343	68.52	137.84
36.2570	0.1997	2.47566	100.00	220.07
47.5558	0.2957	1.91050	12.50	39.86
56.6188	0.1962	1.62431	19.21	46.56

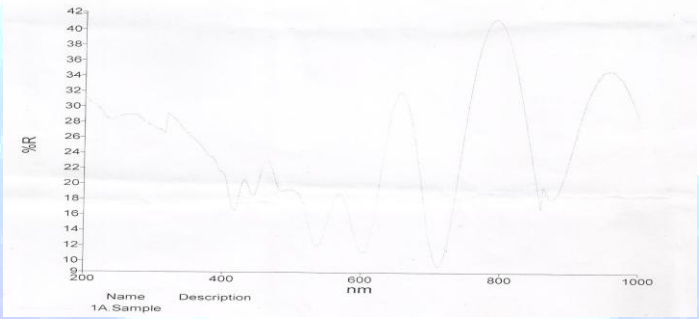

Fig.4 UV-vis REFLECTANCE OF SAMPLE -1A

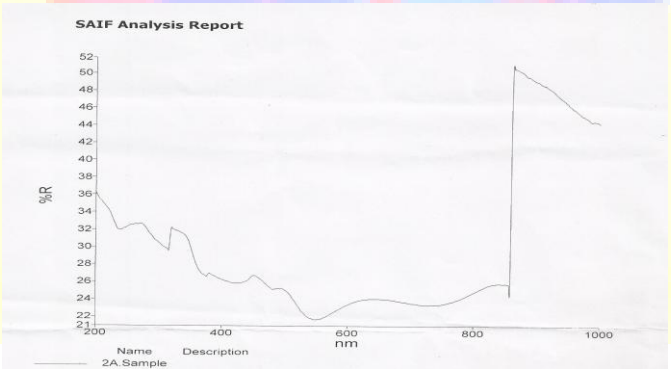

Fig.5 UV-vis REFLECTANCE OF SAMPLE -2A

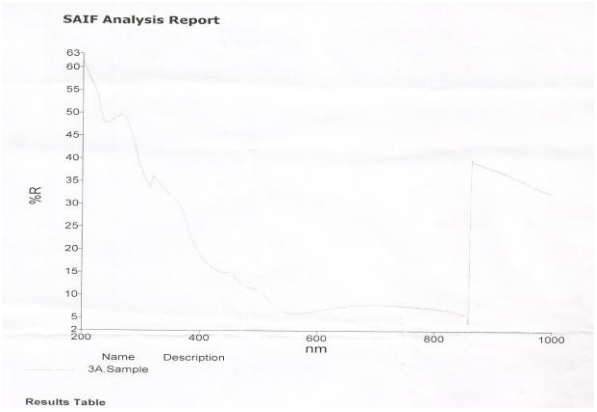

Fig.6 UV-vis REFLECTANCE SAMPLE -3A

PL (Photoluminescence)

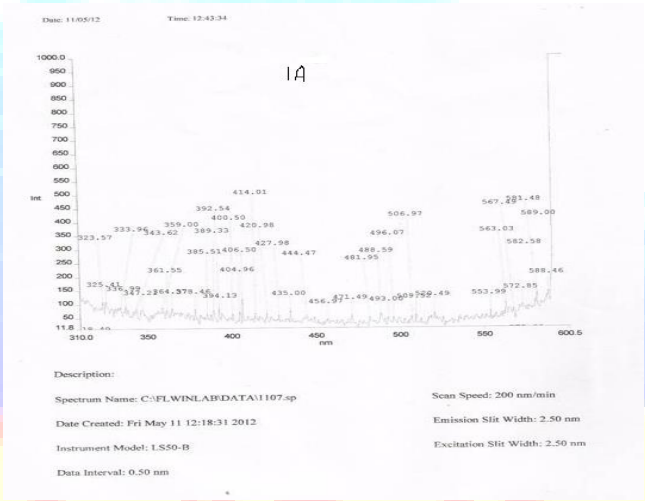

Fig.7 PL OF SAMPLE- 1A

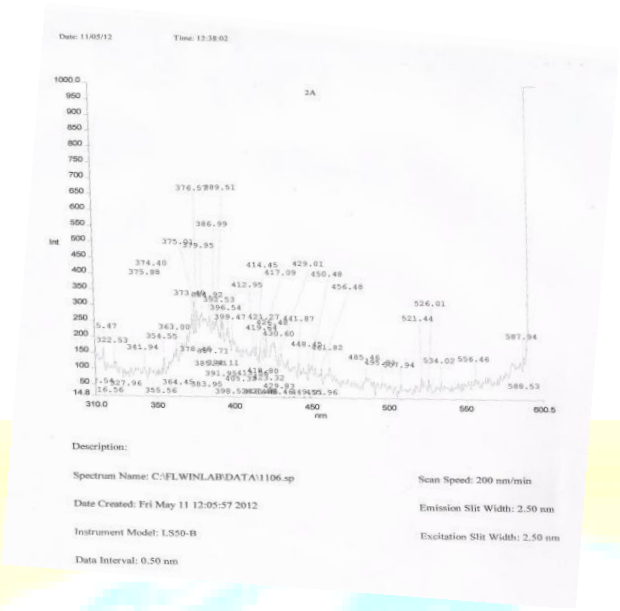

Fig.8 PL OF SAMPLE- 2A

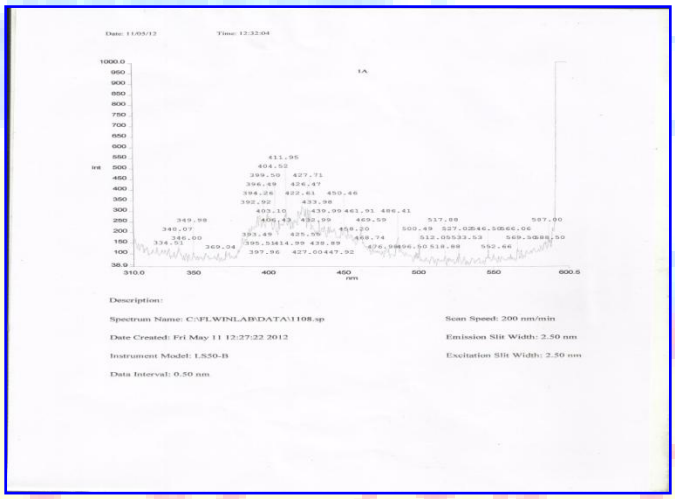

Fig.9 PL OFSAMPLE- 3A

X-ray diffraction patterns of the samples A, B and C are shown in Fig. 1(a). Although the Mg content is increased, the diffraction patterns present three similar main peaks at 31.7±, 34.5± and 36.2±, which are consistent with diffraction peaks from (1010), (0002) and (1011) planes of wurtzite ZnO. No second phase was found within the limit of the XRD measurement although the

later optical spectra proved that the Mg element was doped significantly into the ZnO lattices. This indicates that the ternary compounds of MgZnO were formed and they still keep the wurtzite structure. Even so, it is still noted the slight shift of the (0002) peaks toward the high angle occurred with increase of Mg content in the MgZnO thin films, as shown in the enlarged (0002) peaks in Fig.2, which indicates the lattice constant decreases accordingly. The pure phase of the XRD and the slight shift of the peak position imply that the Mg atoms were effectively doped into ZnO lattices because the radius of Mg2+ is smaller than Zn2+. Maintaining the same ratio of Zn:Mg, the samples were prepared on quartz substrates under the same condition to measure the optical absorption spectra. Figure 4,5and6 shows the reflectance spectrum of each sample reveals a sharp edge in the UV region and nearly transparent in the visible region. Because the energy band gap of MgO is 7.7 eV, ZnO is 3.34 eV, the corresponding energy band gap of the MgZnO ternary compounds was increased. As a result of the incorporation of Mg, a gradual shift of the reflectance edge towards shorter wavelength is noticed. The band gaps of the different samples were estimated from the absorption spectra. The optical absorption coefficient α of the direct band gap semiconductor ZnO can be derived as the following equation:

$$\alpha = A(h\nu - Eg)$$

The band gaps of sample A, B, C and D were deduced as about 3.31 eV, 3.41 eV, 3.49 eV, and 3.51 eV for the four samples, respectively. It clearly demonstrates the band gap broadening with increase of Mg incorporation in ZnO thin films. The EDX measurement demonstrates that the atomic percentage of Mg in the sample 1A, 2A and3A is 0, 1.47%, 2.08%, The unnormalized and normalized (the inset) PL spectra of the 1A, 2A, 3A, samples on Si substrates at room temperature are shown in Fig. 7,8 and 9. It can be observed that the UV peak position obviously shift towards the higher energy region when Mg content increased from the normalized PL spectra. The emission peaks locate approximately at 377 nm, 365 nm,356 nm and 354 nm, for sample 1A, 2A, 3A respectively. It is worth noting that the band-edge emission of the Mg doped films enhances drastically as the Mg content increases from the un normalized PL spectra.

It is assumed that the N element may originate from the diethanolamine (HN(CH2CH2OH)2, DEA) as the stabilizer in the sol- gel process. This might provide an easy approach for N doped into ZnO and might be beneficial to achieving p-type ZnO thin film. Further investigations are required for the N doping mechanism, spatial distribution and related conductive type.

4. Conclusions

In summary, the effects of Mg incorporation on the microstructure and optical properties of the ZnO thin films prepared by sol-gel method were investigated in detail. The grain size and the surface roughness were reduced due to the incorporation of Mg, while the band gap and the absolute intensity of the band edge emission were increased. The chemical composition of the MgZnO thin films were studied by FT-IR spectroscopy, which expanded understanding of the MgZnO thin films.

Acknowledgements

This work is supported by the Director Principal Dr. Daljit Singh (DIET Kharar Mohali Punjab India),my guide Er. Maninder Kaur , co-guide Er. Ashok kumar (H.O.D ECE Dept. Mitthapur Ambala) , Mr. S.C Sood and my family.

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AUTHOR -1

I Seema Singh awarded diploma in Electronics and communication from B.P.S Mahila Polytechnic , Khanpurkalan Sonipat , Haryana ,India. AMIE in ECE from IEI Kolkatta. Pursuing M.Tech in ECE from Doaba Institute Of Engineering and Technology ,Kharar , Mohali Punjab India .My research interests include digital signal processing , wireless communication and VLSI. Presently working as a Lecturer in ECE Dept. of ICL Institute of Engineering and Technology, KUK Kurukshetra Haryana India

AUTHOR-2

Guide:

Er, Maninder Kaur B.Tech (EIE) from PTU Jalander. M.Tech communication systems from Guru N anak Dev University Amritsar Punjab India. Presently working as Assistant Professor in ECE Dept. D.I.E.T Kharar Mohali. Area of interest: OFC, Image Processing and Micro wave.