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PREPARATION, GROWTH, and OPTICAL PROPERTIES OF of (Al/Mg) Co-DOPED ZnO THIN FILMS

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ABSTRACT
$Zn_{0.95-x}Mg_{0.05}Al_xO$ ($0.0\leq x<0.1$) solutions were prepared by sol-gel synthesis using 2, 4-pentanedionate and acetate precursors which were dissolved into solvent and chelating agent. $Zn_{0.95-x}Mg_{0.05}Al_xO$ thin films with different thickness were grown on glass substrates using sol-gel deep coating. The nanoparticles and thin films were then annealed at 600 ℃, were tried to observe the doping ratio, temperature, and thin films effect on structural and optical properties. The particle size, crystal structure, surface morphology and film thickness were characterized by X-ray diffraction, and Scanning Electron Microscope. Optical transmittance spectra of the Al/Mg Co-doped ZnO films were analyzed at room temperature by using a UV-NIR (Shimadzu-2101 PC). The structure, particle size, surface morphology, and optical properties of nanoparticles and the thin films with different doping ratio, thickness (number of dipping), temperature and time of annealing process are presented.
Keywords: Sol-gel Chemistry, X-ray diffraction, Thin Films

1. INTRODUCTION

Zinc oxide (ZnO) attracting an increasing intense attraction due to its technological applications such as transparent conductive electrodes, displaying deviceds, LED’s, gas sensors, solar cells, laser systems etc. Therefore ZnO based films hve been prepared by many groups using various methods such as magnetron sputtering, sol-gel process, pulsed laser deposition, molecular beam epitaxy, spray pyrolyses etc. Among these methods, the sol-gel methods have a adventages such as inexpensive method, low cost, simplicity and better homogeneity [1-5].

The goals of the present work are: to investigate processing, characterization and sol-gel parameters with varying (Al/Mg) Co-doped ratio of the $Zn_{0.95-x}Mg_{0.05}Al_xO$ films on glass substrate using sol-gel coatings technique; to grow crack free, pinhole-free, uniform $Zn_{0.95-x}Mg_{0.05}Al_xO$ thin films; to investigate doping ratios and film thickness on optical properties of thin films.

Fig. 1. The sol-gel coating system.

2. EXPERIMENTAL PART

The mixed oxides $Zn_{0.95-x}Mg_{0.05}Al_xO$ were prepared as solutions with various compositions ($0.0 \leq x \leq 0.1$) using sol-gel technique. Zinc acetate dehydrate (Zn) and Mg 2, 4 pentanedionate (Mg) and Al acetate tetrahydrate (Al) were used as precursor materials and methanol, glacial acetic acit (GAA) and acetyl acetone (AA) were as solvents and chelating agent. The appropriate weighing amount of the Zn, Mg and Al were put all together in a Pyrex container and dissolved in methanol. Following this, GAA was added to containing acid solution. Consisting of metal ions to form complex compound was added AA in environment. They mixed with a magnetic stirrer at room temperature until a transparent solution was obtained. The details of the preparation of ZnMgAlO nanoparticles were discussed in the previous studies [4, 5]. The as prepared powders were ground and annealed individually in air at 600 °C using box furnace.

Microscope glass slides used as substrate cleaned under different conditions and optimum cleaning conditions are decided using scanning electron microscope (SEM) and microscope. These glass slides were dipped into the mixed ZnMgAlO solution at a withdrawal rate of 0.65 m/min and then pulled the vertical furnace at 300 C for 1 min as shown in Fig.1. ZnMgAlO films with different Al concentrations coated on glass slides. ZnMgAlO nanoparticles and the obtained coated glass substrates were annealed in air at temperature 600°C using box furnace. XRD scans were recorded using a Rigaku diffractmeter with Cu K_{α} radiation. Microstructure properties of prepared samples were observed using scanning electron microscope (SEM) (JEOL, JSM-5910LV). Optical properties of $Zn_{0.95-x}Mg_{0.05}Al_xO$ thin films obtained by using PG Instruments UV-Vis-NIR spectrophotometer in 250 – 600 nm range.

3. RESULTS AND DISCUSSION

$Zn_{0.95-x}Mg_{0.05}Al_xO$ nanoparticle powders were obtained with various compositions ($0.0 \leq x \leq 0.1$) using sol-gel technique. $Zn_{0.95-x}Mg_{0.05}Al_xO$ ($0.0 \leq x \leq 0.1$) nanoparticles were annealed at 600 °C from 30 minute under air using box furnace. The X-ray diffraction of $Zn_{0.95-x}Mg_{0.05}Al_xO$ nanoparticles at various compositions ($0.0 \leq x \leq 0.1$) at 600 °C for 30 min in the air is shown in Fig.2. The reflections correspond to ZnO hexagonal wurtzite structure.

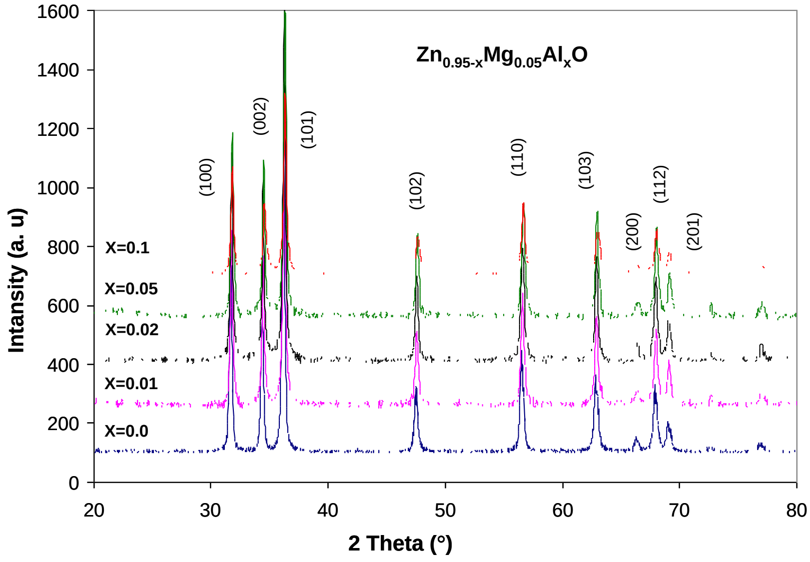

Fig.2. XRD patterns of the $Zn_{0.95-x}Mg_{0.05}Al_xO$ nanoparticles were annealed at 600 ℃ for 30 min.

$Zn_{0.95-x}Mg_{0.05}Al_xO$ thin films were deposited on glass substrate using the sol-gel dip coating system. The quality of thin film depends on withdrawal rate, drying, heat treatment condition and sol structure such as chemical composition, purity of precursor solvent catalyst materials and pH value of starting and stabilized solution.

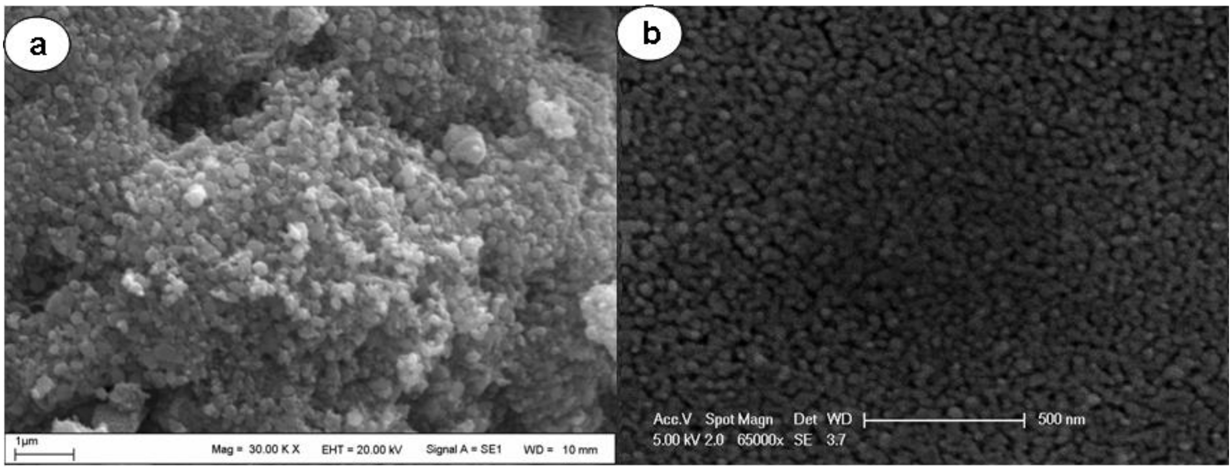

Fig. 3. depicts the SEM picture of a) $Zn_{0.93}Mg_{0.05}Al_{0.02}O$ nanoparticles and b) $Zn_{0.94}Mg_{0.05}Al_{0.01}O$ thin films.

Fig. 3a depicts morphologies of $Zn_{0.93}Mg_{0.05}Al_{0.02}O$ nanoparticles. The grain size of the nanoparticles was observed to be around 90-100 nm. As seen Fig. 3b surface is smooth, dense and without crack and porosity. Thickness of the coating was controlled by viscosity of solution, number of dipping, changing withdrawal rate and temperature of the furnace.

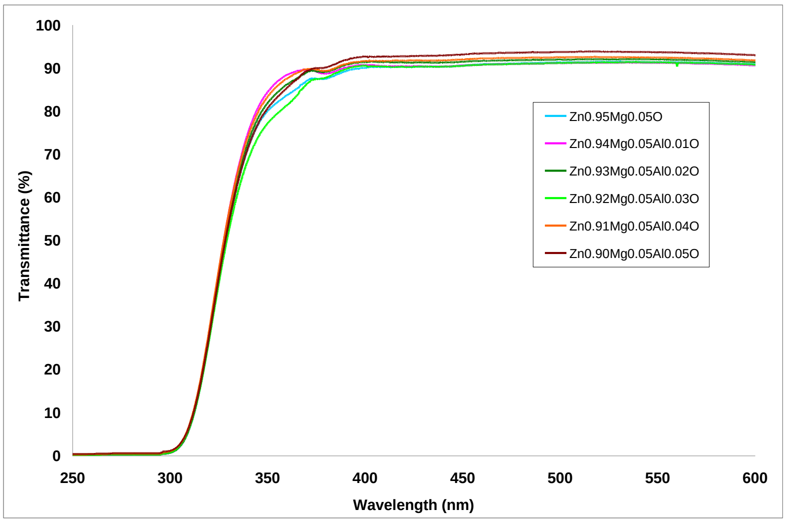

Fig 4. Room temperature optical transmittance spectra of $Zn_{0.95-x}Mg_{0.05}Al_xO$ thin films deposited on glass substrate.

4. CONCLUSIONS

$Zn_{0.95-x}Mg_{0.05}Al_xO$ thin films were coated on the glass substrate using sol-gel dip coating system. The thickness of the Al/Mg Co-doped ZnO thin film increases by increasing the number of dipping, withdrawal speed, and solution density. Smooth, dense and crack free thin film was produced. The $Zn_{0.95-x}Mg_{0.05}Al_xO$ layer was observed uniform on the glass substrate by using SEM. The grain size of $Zn_{0.95-x}Mg_{0.05}Al_xO$ nanoparticles were 90-100 nm. From XRD analysis, it was shown that the reflections correspond to ZnO hexagonal wurtzite structure without secondary phases.

Acknowledgements

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