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Influence of Aluminum Doping on Structural, Morphological and Optical Properties of AZO Thin Films Prepared by Sol-Gel Dip Coating

KEYWORDS: AZO; Sol-gel; Transmittance

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ABSTRACT *Aluminum doped Zinc Oxide (AZO) thin films with 0-5at.% aluminum content were prepared by Sol-gel dip coating technique. The thickness of the films measured using alpha step method. The structural and morphological properties are studied respectively using X-Ray Diffraction (XRD) technique and Scanning Electron Microscopy (SEM). Higher intensity Zinc Oxide (ZnO) peak (002) is observed in 1at.% Aluminum doped film with 450°C of annealing temperature. The grains are more densely packed in the films doped in 1at.% Aluminum content. Optical properties of AZO thin films are tested by UV-Visible Spectroscopy, while comparing with all the films 0.5at.% Aluminum doped film produces more than 90% of transmittance.*

1. INTRODUCTION

Transparent Conducting Oxides (TCOs) based on Zinc Oxide (ZnO) is promising material for Photovoltaic (PV) cells and various optoelectronic devices [1]. ZnO thin films have interested as transparent conductor, because the n-type ZnO thin films has a wide band gap semiconductor (Eg=3.2eV), and high transmission in the visible range, and ZnO thin films can take place of Tin dioxide (SnO_2) and Indium Tin Oxide (ITO) because of their structural and optical properties and its excellent stability which has been mentioned widely [2-3]. ZnO thin films have been prepared with different process such as pulsed-laser deposition, chemical vapor deposition, spray pyrolysis and sol-gel process etc. among them, the sol-gel dip coating technique offers possibility of preparing a small to large area coating of ZnO thin films at low cost for technological applications[4-8]. The technology related to ZnO based TCOs with low resistivity and high transmittance by simple and low temperature deposition technique critically determines on whether the ZnO based TCOs were utilized in touch panels, liquid Crystal Displays (LCD) and other electronic devices [9-12].

In several studies, the structural and optical properties of AZO thin films could be obviously improved by optimized deposition conditions and controlled doping. The primary objective of this work is to investigate the influence of the preparation conditions on structural and optical properties of AZO films prepared by sol gel dip coating method. Alpha step method is used to measure the film thickness. The structural characteristics have studied using X-Ray Diffraction (XRD), the morphological features have studied using Scanning Electron Microscopy (SEM) and the optical properties are investigated by using UV-Visible Spectrometry (UV-Vis) respectively.

2. EXPERIMENTAL DETAILS

Zinc acetate dihydrate ($C_4H_6O_4Zn.2H_2O$) and diethanolamine ($C_4H_{11}NO_2$) are used as a precursor material and the solvent, respectively. Diethanolamine (DEA) was first dissolved in isopropanol (iProH), Zinc acetate dihydrate (ZnAc) was added under stirring, and heated under reflux for 1 hour at 70°C. Zinc acetate was dissolved in DEA by the molar ratio of 1 (DEA/Zn=1).

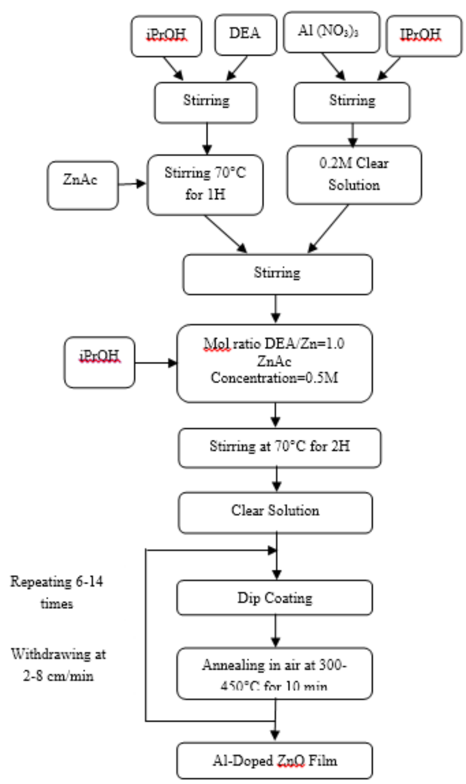

Figure.1 Flowchart showing the procedure for preparing the aluminum doped ZnO films

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Doping of the solution will obtain by adding a 0.2M solution of aluminum nitrate in ethanol. The molar ratio of dopant in the solution (Al/Zn), varies between 0.5% to 5%. Isopropanol is added to adjust the solution concentration to 0.5Mol/L of ZnAc. The molar ratio of DEA to ZnAc was maintained 1at.%. The solution was stirred at 70°C for 2 hours to yield a clear and homogeneous solution, which served as the coating solution after cooling to room temperature. The coating was usually made one or two days after the solution was prepared. Figure.1 shows the procedure for preparing the AZO films.

3. STRUCTURAL AND MORPHOLOGICAL CHARACTERISTICS

X-Ray Diffraction Analysis

The x-ray diffraction analysis (XRD) was used to investigate the physicochemical process involved in the sol-gel deposition of ZnO. The XRD patterns of these films with different aluminum contents are shown in Figure.2. The ZnO (002) XRD peak is the only peak observed for all the aluminum doped films, irrespective of the aluminum content, which indicates that preferred orientation of the crystals is present with the c-axis perpendicular to the substrate surface. Higher intensities of the (002) peak are observed in a limited aluminum concentration range of 0.5 to 1.0 at.%. Higher intensities of the (002) peak are observed in a limited aluminum concentration range of 0.5 to 1.0 at.%.

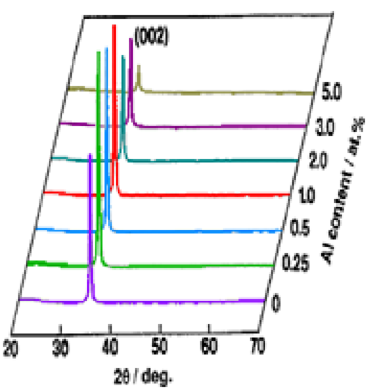

Figure.2 XRD patterns of the aluminum doped ZnO films with different aluminum contents

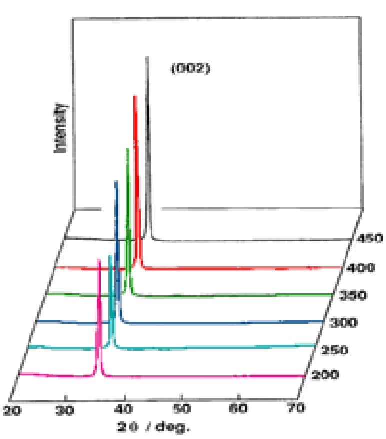

Figure.3 XRD patterns of the 1.0at.% aluminum doped ZnO films prepared for various annealing temperatures

Figure.3 shows the XRD patterns of these 1.0 at.%. aluminum doped films prepared with different annealing temperatures. All the films show a preferential crystal orientation, and the intensity of the ZnO (002) peak increases as the annealing temperature increases, up to 450°C, and show a maximum at that temperature, which indicates the highest degree of crystalline orientation attained by preheating the gel films at 450°C.

3.2. Morphological Analysis

The morphological feature was studied by scanning electron microscope. Figure 4(a)–4(d) shows SEM micrographs of the different aluminum contents. The grains have a tendency to decrease in size as the aluminum content increase, as shown by the surface morphology.

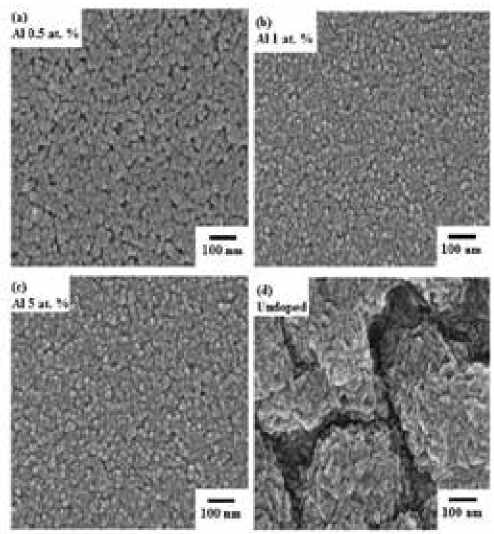

Figure.4 SEM images of doped and un-doped ZnO thin films with different aluminum contents,

(a) 0.5 at. % AZO , (b) 1 at. % AZO, (c) 5 at. % AZO and (d) Undoped ZnO

The grains are more densely packed in the films with higher aluminum contents. The surface micrograph reveals a lower porosity for films with higher aluminum content, which is the consistent with this observation.

4. OPTICAL PROPERTIES

The optical properties are investigated by UV-Visible Spectrophotometry. Figure.5 indicates that the transmittance of the doped and un-doped ZnO films is always higher than 80% and the transmittance of the doped ZnO films is higher than that of the un-doped films.

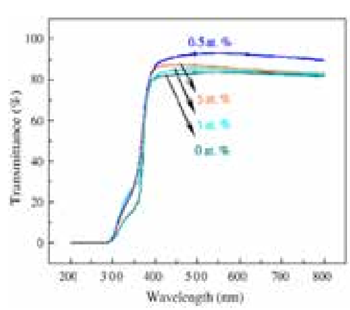

Figure.5 Optical transmittance of the Un-doped and Doped ZnO thin films

Moreover, the transmittance of doped film with 0.5 at.% Al is nearly 90% for wavelengths over 400nm, and is higher than that of the doped film with 1 at.% and 5 at.%. This may be due to the fact that the film with 0.5 at.% doping presents more voids than the films with 1 and 5 at.% doping. This may lead to a decrease in optical scattering. According to Figure.6, when the thickness of the AZO films is increased from 400nm to 1600nm, the transmittance of the AZO films decreased gradually.

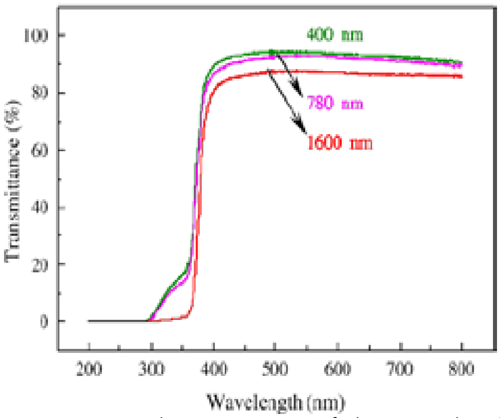

Figure.6 Optical transmittance of the AZO thin films for various film thicknesses

5. RESULTS AND DISCUSSION

Aluminum doped Zinc Oxide (AZO) thin films were prepared by a Sol-gel dip coating technique and the films are under went to annealing in air followed by cooled to the ambient atmosphere. Films were annealed to tune and enhance the desired properties. The X-ray diffraction technique reveals that, the higher intensity of the (002) peaks are observed in a limited aluminum concentration range of 0.5at.% to1at.%. When the annealing temperature was increased from 200-450°C, the films were oriented more preferentially along the (002) direction, the grain size of the films increased. According to the SEM results 0.5at.% aluminum doped film exhibits a porous microstructure and the spherical crystalline particle size is approximately 38nm. It is observed from the results of UV visible spectroscopy, all the aluminum doped zinc oxide thin films have the optical transparency of greater than 80%. The transparency of 0.5at.% aluminum doped films has more than 90% transmittance over visible region and also it reveals that, when the thickness of AZO films is increased from 400nm to 1600nm, the optical transmittance of the films decreased gradually.

6. CONCLUSION

The present work is focused on the synthesis of undoped zinc oxide and Aluminum doped zinc oxide thin films with the Aluminum ratios of 0.5%, 1% and 5.0% and its structural, morphological and optical properties are investigated. It reveals that the structure, morphology and maximum optical transparency of AZO thin film got potential applications as TCOs in photovoltaic cells and solid-state optoelectronic devices etc.

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