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Influence of Dopant Concentration on the Properties of Aluminium Doped Zinc Oxide Nano Structured Thin films

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Abstract: Aluminum doped Zinc Oxide (ZnO:Al, AZO) thin films were prepared by the Sol–gel spin coating method. The Sol solution was prepared with different Aluminum concentration corresponds to 0.002 to 0.01 mol %. The XRD pattern of the films exhibits a preferential c-axis orientation along the (002) plane. Optical properties of the AZO films were studied from the Transmittance Spectra in the UV-Visible region, where the maximum transmittance evaluated was around 80%.The electrical resistiviy of these films have been found to reduce due to Al incorporation in the ZnO lattice.The SEM images of the films illustrate the morphology and showed the particle size getting affected by dopant concentration. These results open a new avenue of research to appraise the suitability of AZO thin films for Solar Cell Applications.
Keywords: AZO thin films; Dopant concentration; Spin coating; band gap; Resistivity.

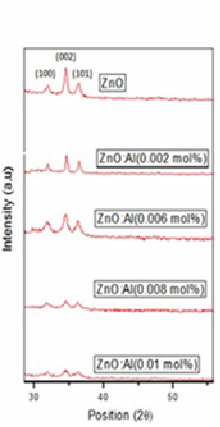
Introduction and Experimental

Zinc oxide (ZnO) belongs to the class of transparent conducting oxides which can be used as transparent electrodes in solar cells, electronic devices and heated windows. ZnO nanostructures are emerging as the key building blocks for nanoscale optoelectronic devices [1]. Doped zinc oxide films have a number of attractive applications, such as gas sensor devices, piezoelectric devices, transparent conducting electrode materials, photo voltaic devices(2) etc. The sol-gel method is of interest, as it enables film deposition in conventional lab environments without the need for expensive equipment.

ZnO thin films doped with Al were prepared by Sol gel spin coating method. The Sol solution was prepared by dissolving Zinc acetate dehydrate [$Zn(C_2H_3O_2)_2$] in the solvent mixture ethanol and monoethanol amine [$H_2NC_2H_4OH$] . The molar ratio of monoethanolamine to Zinc acetate was 1:1 and the concentration of Zinc acetate dehydrate was 0.36 mol/L. Aluminium chloride was used as the source material for doping. Varying concentrations of Aluminiumchloride,0.002, 0.006, 0.008, 0.01 mol % were considered for preparing the Sol solution. The resultant solution is refluxed at 60 °C for 1 hour and then allowed for aging in the ambient. The coating was made on the third day of aging which was the optimized critical time for coating. The Sol solution was spin coated on glass substrate up to 10 layers at spin rate 3500 rpm with coating time of 10 seconds. The thin films were subjected to heat treatment after each coating at 350°C for 5 min duration to evaporate the solvent and to remove the organic residuals. All the prepared films were then annealed at 450 °C in vacuum furnace for one hour.

Results and Discussion

The X-Ray diffraction pattern and the corresponding structural parameters of undoped and Al doped ZnO thin films are shown in fig.1. XRD pattern of all the films found to have three diffractive peaks (100),(002) and (101) which are the characteristic orientations of hexagonal ZnO wurtzite structure. The highly intensed peak at the diffraction angle 34° corresponds to (002) orientation as found in the XRD of pure and doped films, represents the formation of C-axis normal to the substrate [2]. The diffractive peak intensity of undoped film is higher than that of Al doped films, which may be due to the replacement of Zn ion by Al ion or Al ion inserts itself into interstitial site of ZnO lattice[3], resulted in change of lattice and therefore reduces the crystalline quality of the films with increasing Al concentrationas observed by earlier results[4].The intensity of the referred orientations are higher for the film coated with Al concentration at 0.006 mol%, when compared with other concentrations, indicates the formation of best crystallite orientations at this dopant concentration. The lattice constants a and c calculated for this film are 3.238 and 5.211A˚ respectively which matches with JPDS data file no. 05-0664, conforms the hexagonal phase of the grown ZnO crystallites on the thin film. The Crystallite size calculated for the AZO thin films using Scherer formula varies between 10 and45 nm. The calculated values of Texture coefficient found to be more than 1 for (002) planes, which also conforms the preferred C-axis orientation.



Al (dopant) concentration (mol%)	Lattice Orientation h k l	Texture Coefficient TC	Lattice constants a(A˚)	c(A˚)	Particle size D (nm)
0	1 0 0	0.523136	3.2329	5.19678	17.5359
	0 0 2	2.039212			25.21841
	1 0 1	0.437652			22.18885
0.002	1 0 0	0.507688	3.2351	5.17822	45.40348
	0 0 2	1.859507			25.55158
	1 0 1	0.632805			34.92772
0.006	1 0 0	0.664908	3.2376	5.18206	14.61151
	0 0 2	1.544835			35.31547
	1 0 1	0.790256			35.47899
0.008	1 0 0	0.79652	3.2448	5.18284	8.624083
	0 0 2	1.400793			12.41019
	1 0 1	0.802687			8.727428
0.01	1 0 0	0.843367	3.2385	5.21184	8.626231
	0 0 2	1.603903			8.673111
	1 0 1	0.55273			8.720972

Fig.1. XRD pattern and Structural properties of undoped And Al doped ZnO films

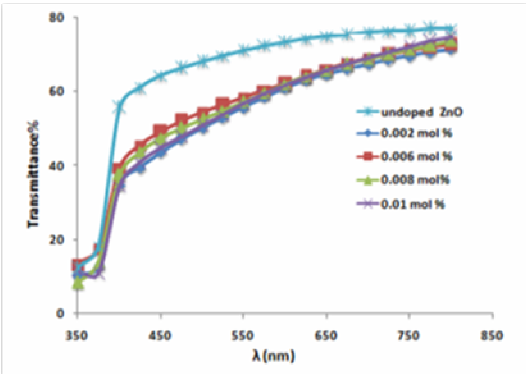


Fig. 2. Transmittance spectra of AZO films at different Al concentration

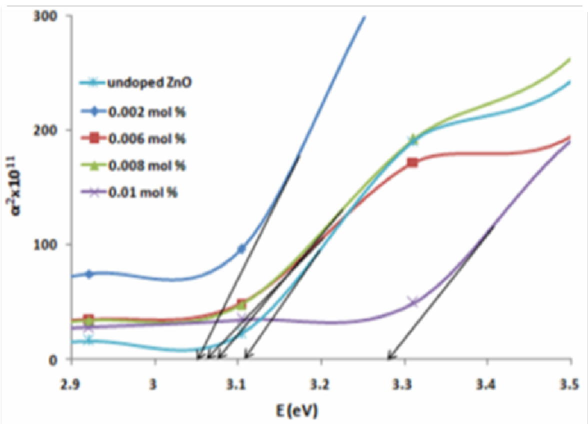


Fig.3. Band gap energy of AZO thin films at different Al concentration

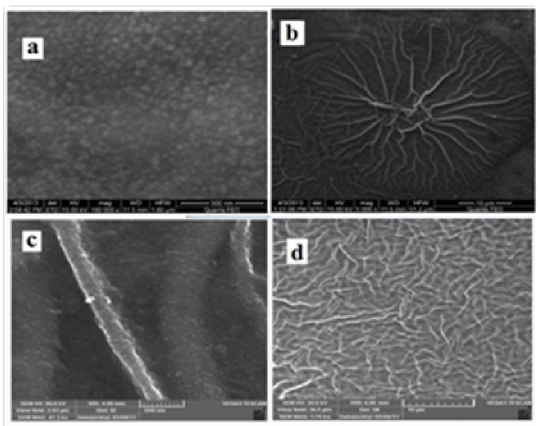


Fig.4. SEM micrographs of(a)&(b) undoped and (c)&(d)Al doped (0.006 mol %) ZnO thin films.

The optical transmittance spectra recorded in the wavelength region 300-850 nm for AZO films are shown in fig.2, displaying that all films were highly transparent in the visible region. The maximum transmittance percentage observed was around 75 % for all the AZO films with different Al concentrations. There is a slight decrease in transparency of the doped films as compared to the undoped film. This may be due to the variation in grain size as well as the clustering of particles as the dopant material introduced [5]. This can be understood from the determined particle size values from the XRD data. The value of absorption coefficient (α) was determined and the band gap energy of AZO thin films were determined using the plot between α^2 and photon energy hν as shown in fig.3. The measured band gap energy of the films varies between 3.0 eV to 3.25eV and is well in agreement with reported results [4]. It was also observed that there is an increment [6]in the band gap energy value of doped thin film and maximum [7]at Al concentration of 0.01mol % as reported by Wen-Jauhchen et al[4].

The resistivity of AZO thin film coated at 0.006 mol % were measured as 35 M Ω and for other dopant concentration its value ranges up to 120 MΩ. This lowering of resistivity [8]with increase in Al dopant concentration leads to the enhancement in the electrical conductivity.The film morphology analysis was carried out with scanning electron microscope. Micrographs of undoped and Al doped film at concentration 0.01 mol% with different magnifications are shown in fig.4(a),(b), (c)&(d).The surface nature of these films was compared. The grain size appears to be reduced for doped film than that of pure film[5] as seen in the microstructures of higher magnification. The images obtained at lower magnification for undoped film shows fiber-like streaks or wrinkles and for the doped film it shows homogeneous worm like structure [9] and observed the continuous coverage of the substrate with the material. The grain size measured by SEM ranged at 50 nm is larger than those measured by XRD, which shows the grains appeared in SEM [9]are the domains formed by aggregation of nanosize crystallites, that is due to different grain size criteria underlying different method[10].

Conclusion

Transparent oxide thin films of Al doped thin films were successfully fabricated on glass substrates using Sol-gel method. From the characteristic analysis, it is evident that the Al doping at different concentration in ZnO thin films remarkably affects the structural and optical properties. The thin films coated with 0.006 mol % Al concentration exhibited the strong(002) reflection and observed Al doping has not changed the hexagonal wurtzite structure of ZnO. At this dopant concentration the energy band gap of the film was measured as 3.2 eV. Among the thin films investigated in the present study the 0.006 mol % Al doped ZnO thin films exhibited decrement in resistivity, the best average transparency with uniform grains in surface. This study is helpful in finding the suitability of ZnO thin films in Solar cell applications in terms of optimized usage of Al dopant.

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