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Al-doped ZnO: Synthesis, Characterization, Photoconducting nature for efficient DSSCs

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Abstract: In recent years the synthesis of metal doped nanoparticles has attracted particular attention owing to their structural, morphological, optical and photoconducting properties. In the present work, a simple sol gel method is followed to synthesize Aluminium (Al) doped Zinc Oxide (ZnO) nanoparticles. The as- synthesized nanoparticles were characterized by X-Ray Diffraction (XRD), High Resolution Scanning Electron Microscopy (HRSEM) equipped with Energy Dispersive X-Ray Spectroscopy analysis (EDAX), Ultra Violet-Diffuse Reflectance Spectroscopy (UV-DRS) and Brunauer–Emmett–Teller (BET) Analysis which evidenced the formation of highly crystalline, homogeneous nanoparticles with an average crystallite size of 23.15nm, band gap of 2.57eV and surface area of 7.68m^2 /g respectively. Also investigation of the electrical properties of the sample using field dependent dark and photoconductivity measurements presented a significant increase in photoconductivity. Thus the results show that Al doped ZnO would be a promising candidate for photovoltaic applications especially as photoelectrodes in Dye-Sensitized Solar Cells (DSSCs).
Keywords: Band Gap, Doping, Optical Properties, Photoconductivity, Sol-gel

1. Introduction

Semiconductor metal oxides have been under extensive investigation due to their wide applications in energy storage and environmental remediation [1]. Recently, ZnO is emerging as an important metal oxide owing to its cheap abundant raw material, direct band gap, large exciton binding energy, high transmittance in the visible region and non-toxic nature [2]. The structures shared by the ZnO crystal are wurtzite, zinc blende and rock salt type. The hexagonal wurtzite structure was selected for this study since it exists at normal conditions (pressure, temperature) and thus is more meaningful on being investigated [3]. For the purpose of improving the electrical conductivity and optical transmittance of ZnO, group III elements such as boron, aluminum, gallium and indium are introduced to ZnO [4]. The objective of this work was to study the effects produced by Al doping upon the morphological, structural, surface area and photoconducting nature of ZnO wurtzite lattice.

2. Experimental details

For the synthesis of Al doped ZnO, small amount of Zinc Acetate Dehydrate ($C_4H_{10}O_6Zn$) was added with 100 ml of Ethanol and allowed to stir for few minutes. Few grams of Aluminium Chloride ($AlCl_3$) was added with 5 ml of distilled water and stirred for some time. The Zinc acetate dehydrate solution and the Aluminium Chloride ($AlCl_3$) solution is mixed together and allowed to stir for few minutes. Sodium hydroxide is added with distilled water and stirred separately for 15 minutes and then NaOH solution is added in drops to the Zinc Acetate Dehydrate and $AlCl_3$ mixed solution till the pH value reaches 12. The mixed solution is allowed to stir continuously for 12 hours. Then the mixed solution is kept undisturbed for sedimentation. Later it was washed with ethanol for several times, filtered and kept in furnace at 120□ C for 12 hours. The powder is calcined at 500□ C for 1 hour resulting in the formation of Al doped ZnO nanopowder [5]. Powder X-ray diffraction (XRD) was performed on a Rich Siefest 3000 X-Ray Diffraction equipped with a Cu Kα monochromatic radiation source (λ = 1.54187 Å). HRSEM images of as-synthesized sample were taken on a HITACHI S-4800 – IIT Madras, equipped with an Energy Dispersive X-ray Spectrophotometer. The optical studies were carried out by Lamda- 35 UV-Vis / DRS Spectrophotometer. Surface area, pore size distribution was measured by Quantachrome Nova-1000 Instrument. The field-dependent dark and photoconductivity studies were carried out using Keithley Picoammeter 6485 [6].

3. Results and Discussion

Fig.1 shows the XRD pattern of ZnO nanoparticles, prepared by sol gel method. All the obtained peaks in the XRD pattern are well matched with the wurtzite phase of ZnO crystals and well consistent with the JCPDS card (card no.36-1451) [7, 8]. No other phase peaks were detected, indicating the purity of the as-synthesized sample. The average crystalline size of the sample was found to be 23.15nm calculated using the Scherer's equation,

$$D = k\lambda/\beta \cos\theta$$

where, D is the crystallite size, k is a constant (shape factor, about 0.9), λ the X-Ray wavelength used (1.540Å) and β the full width half maximum of the diffraction angle [9].

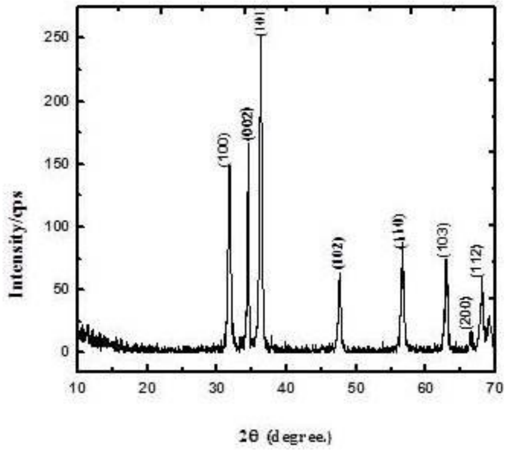

Fig. 1 XRD Pattern of Al doped ZnO

From the HR-SEM images in Fig. 2, it can be seen that the nanoparticles are highly dispersed without any aggregation between them and are uniformly distributed and the average particle size was found to be 38.3nm. Fig. 3 shows the EDAX of the as synthesized sample confirming the presence of elements such as Zinc, Aluminium and Oxygen.

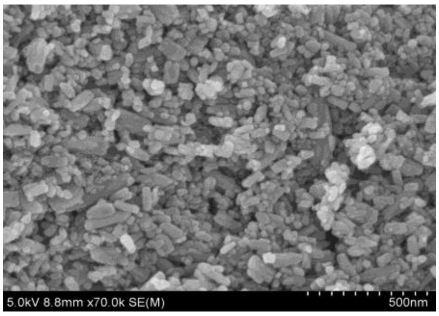

Fig. 2 HRSEM Micrograph of Al doped ZnO

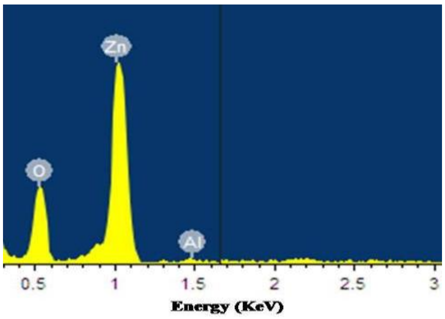

Fig. 3 EDAX spectrum of Al doped ZnO

The absorption spectrum of Al doped ZnO was measured between 200 and 800 nm range as shown in Fig. 3. The exciton absorption is at about 287nm. The optical band gap of the sample was calculated by Kubelka-Munk function $[F(R)\ h\nu]^2$ versus photon energy (hν) [10]. From the optical absorption edge, the band gap of Al doped ZnO was found to be 2.57eV.

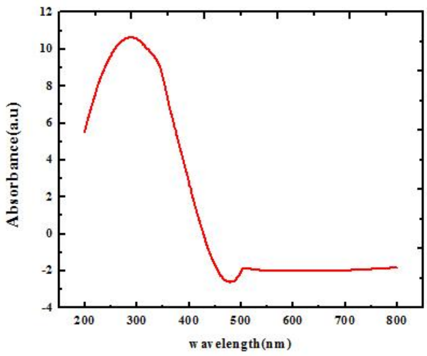

Fig. 4 Absorption spectrum of Al doped ZnO

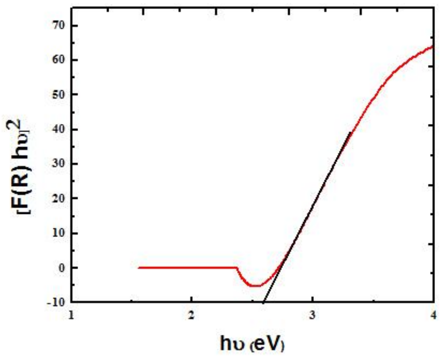

Fig.5 Kubelka Munk Plot of Al doped ZnO

Brunauer–Emmett–Teller (BET) analysis is carried out to study the specific surface area of the as-synthesized samples. The nitrogen adsorption- desorption isotherms and BJH pore size distributions derived from the desorption branch of the adsorption isotherms of the prepared sample are shown in Fig. 6. The sample can be categorized isotherms of type IV [11], indicating the presence of mesoporous materials according to the International Union of Pure and Applied Chemistry classification. The BET surface area was found to be 7.68 m^2/g with a pore diameter of 15.05 nm. The Field dependent dark and photoconductivity studies as in Fig. 12 indicate a linear increase of current in the dark and visible light illuminated samples with increasing applied field depicting the ohmic nature of the contacts as in Fig. 7. The low values of dark current and insignificant rise in photocurrent upon visible light illumination are as expected.

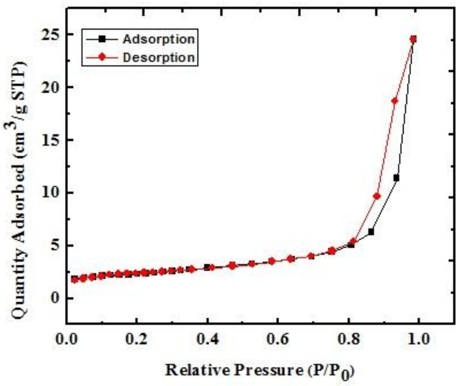

Fig. 6. Nitrogen adsorption isotherm of Al doped ZnO

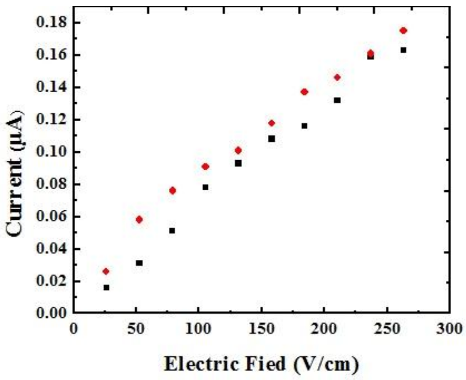

Fig. 8. Field depended dark and photoconductivity of Al doped ZnO

4. Conclusions

Al doped nanoparticles were synthesized by a low cost sol-gel method. Effect of Al doping on the structure, morphology, optical absorption properties, surface area and the photoconducting nature of ZnO were studied. Structural studies have shown a highly crystalline and pure sample with a very small crystallite size of about 23.15 nm. Morphological studies showed the homogenous nature of the sample. The band gap of ZnO was tuned because of the addition of Al which in turn showed a high surface area and thus enhanced the photoconducting nature of ZnO. Thus it could be concluded that Al doped ZnO would be a promising candidate for photovoltaic application especially as photoelectrodes in Dye-Sensitized Solar Cells (DSSCs).

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