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Synthesis and Characterization of aluminum dope zinc sulfide (Al:ZnS) thin films by chemical bath deposition techniques

Abstract

A well adherent thin films of Aluminum doped zinc sulphide (Al:ZnS) has been deposited on silica glass substrates using acidic chemical bath deposition (CBD) containing zinc acetate, Aluminum Chloride, and thioacetamide. EDTA was used as complexing agent to control the free ion concentration of the thin films. Aluminum concentrations were doped by 2%, 4%, and 6% while keeping other deposition parameters constant as deposited Al:ZnS thin films. The samples were characterized by Bruker*D*8 diffractometer with cu$K\alpha$ (λ=1.5406A°) radiation working at 40 mA and 40 kV, JOEL-2300 Analysis Station Scanning electron microscope (SEM), and Perkin Elmer Lambda 950 UV-vis/NIR spectrophotometer. The structural characterization of the samples show that no intense peaks were observed indicating the amorphous nature of the films. The surface morphology studies of as deposited Al:ZnS thin films shown the films were uniform, dense, and composed of spherical shaped grains. EDAX shows the elemental composition of Zn, S, and Al. The ratios of Zn/S in Stoichiometric even though the concentration of Aluminum is increased. Optical absorbance of the films decreased with increase Aluminum concentration. The large band gap makes them good materials for application as a window layer for solar cells.

Keywords: thin film, sulphide, semiconductor and aluminum chloride

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Abbreviations: CBD, chemical bath deposition; SEM, scanning electron microscope; Al:ZnS, aluminum doped zinc sulphide

Introduction

The energy band gap of semiconductor is the most distinct property of semiconductors separate them from insulators and metals. It determines wavelengths of light that can be absorbed or emitted by the semiconductors. The ionicity becomes even larger and more important in the II-VI compounds such as ZnS. As a result, most of the II-VI compound semiconductors have band gaps larger than 1 eV. The exceptions are compounds containing the heavy element mercury (Hg). Mercury Telluride (HgTe) is actually a zero band gap semiconductor (or a semimetal) similar to gray tin. While the large band gap II-VI compound semiconductors have potential applications for displays and lasers, the smaller band gap II-VI semiconductors are important materials for the fabrication of infrared detectors.[1,2]

Recently, there were many advances enabled by thin film deposition techniques in a wide range of technological applications: laser diodes, solar cells, electroluminescent displays etc.[3] The main advantages of using ZnS are :i) thin film coating in the optical and microelectronic industries ii) ZnS reduces engenders toxic to our environment when compared with CdS because of its high toxic, and iii) the use of the wide band gap material ZnS (3.7 eV) as a buffer layer could decrease the window absorption losses and improve the short-circuit current of the cells and can be replace CdS (2.4 eV) increasing the current generated.[4,5] To obtain its full behavior of conductivity, the electrical resistivity needs to be reduced since un-doped ZnS films have electrical very high electrical resistivities (typically 107 Ωcm). This is possible by doping these layers with suitable impurity atoms without affecting the optical properties of the layers too much. Efforts have been made by several research groups to dope ZnS films with elements such as In, Al, Cu and Cl. In this study, Al-doped ZnS films have been deposited by a simple chemical bath deposition (CBD) method. This method has been successfully used to deposit a variety of metal chalcogenides and oxides as this method is expected to dope the films uniformly. This paper reports on the synthesis of Al-doped ZnS layers using different dopant concentrations and studies of the physical and chemical properties of the grown films.[6,11]

Experimental details

Zinc acetate, thioacetamide and aluminium chloride were used as the precursors for Zn, S and Al respectively. The zinc acetate was dissolved in methanol to make a 0.1M solution, whereas the aluminium Chloride and thioacetamide were also dissolved in distilled water to make 0.1 M of solution.[7] Appropriate amounts of the solutions were mixed to prepare the reaction bath. Since ZnS precipitation is reported to occur even for very low zinc and sulphur ion concentrations in solution, a suitable complexing agent EDTA was used to control the release of the ions in the solution. In the present study, ammonia and hydrazine hydrate were used as complexing agents to control the free ion concentration. The pH of the solution was maintained as 5.8 during the deposition (measured using an INSAT Instruments pH meter). Ultrasonically cleaned glass were used as substrates. These were placed vertically inside the beaker containing the reaction bath. Stirring of the solution continued during the deposition using magnetic stirrer. The thin films were deposited at a bath temperature of 75°C for a duration of 60 min. Finally the films were taken out of the reaction bath, washed with distilled water and dried in a hot air oven for 15 min. The surface topology had determined by using JOEL-2300 Analysis Station Scanning electron microscope (SEM). The composition of the films had determined using JOEL-2300 Analysis Station (EDAX) energy analyzer. Structural Characterization of the film was carried out by BrukerD_8diffractometer with cuKα(λ=1.5406A°) in 2θ range, 20°– 70°, radiation working at 40 mA and 40 kV. The electrical resistivity of the layers were identified using Perkin. Elmer Lambda

950 UV-vis/NIR spectrophotometer that the band gap is decreased as the percentage of aluminium is increased and show that electrical resistivity of Al:ZnS is decreased as energy band gap decrease with increment of conductivity of Al:ZnS.

Results and discussion

The surface morphology of the deposited Al:ZnS films were whitish or pale in appearance, uniformly distributed on the surface of substrate with enlarged irregular shape with smaller pin holes, while it shows uniform, compact, homogenous, and composed of nearly spherical shaped grains distribute on the surface of substrate as it shown in Figures 1&2.[8,9] The elemental composition of 2% Al:ZnS thin film were Zn=44.31%, S=43.29%, and Al=12.39%; where as the composition of 6% Al:ZnS thin film were Zn=40.29%, S=39.32%, and Al=20.39%. The presence of other elements such as carbon, sodium, silicon, oxygen, Calcium, magnesium, and aluminum are suspected to be as a result of the microscope glass slide used as the substrate. The percentage of aluminium in the films are greater than added in the initial mother solution.[10] This may be due to the impurity in the substrate and the sample holder of the EDAX machine which is made from aluminium as shown in Figure 3. For this reason, the percentage of aluminium impurity become large as its percentage increases within the deposition solution of Al:ZnS thin films.

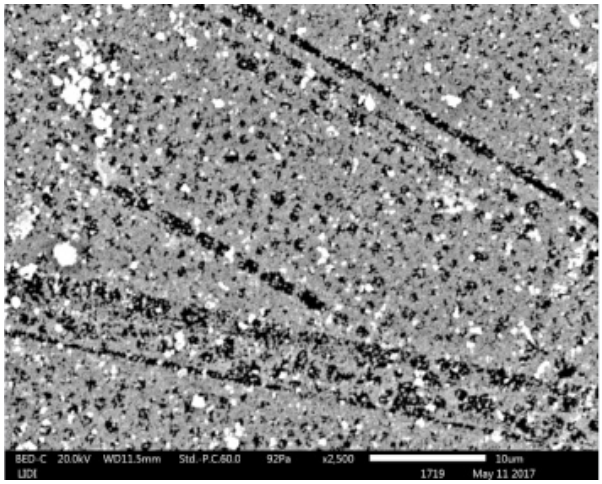

Figure 1 SEM micrograph of as deposited Al:ZnS thin films with 2% Al.

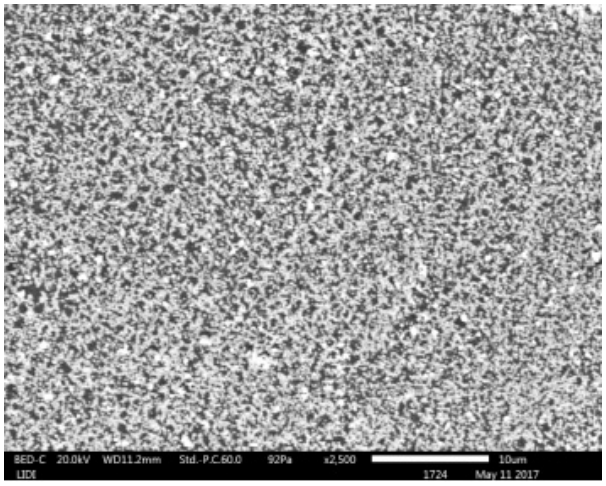

Figure 2 SEM micrograph of as deposited Al:ZnS thin films with 6% Al.

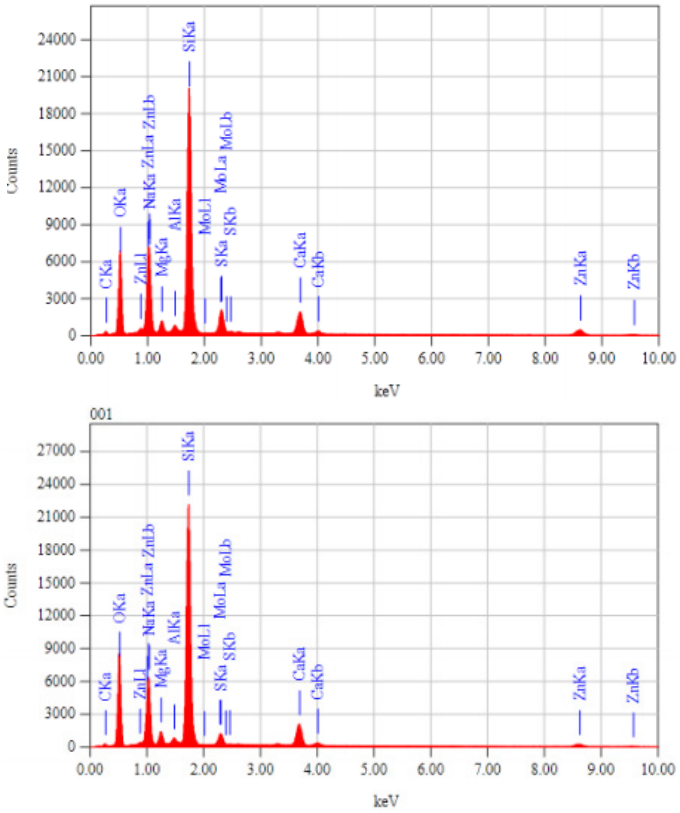

Figure 3 EDAX elemental analysis of as deposited Al:ZnS thin films.

The x-ray diffraction pattern of ZnS films doped with different 'Al' concentrations are shown in Figure 4. The X-ray diffraction pattern of ZnS films doped with different Aluminium concentrations are shown in Figure 4 show that no intense peaks were observed as the synthesized layers are non crystalline (amorphous) indicating that the sample has no long range order and is macroscopically identical in nature.[12]

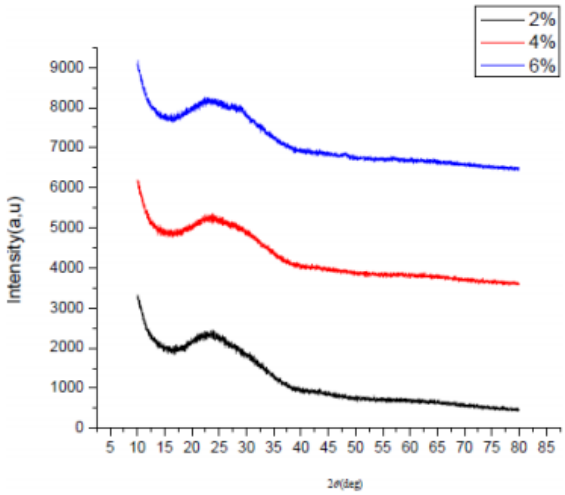

Figure 4 X-ray diffraction pattern of Al:ZnS thin films.

Figure 5 shows the optical transmittance spectra of as-deposited ZnS: Al films with different Al dopant concentrations. It shows the absorption spectra of thin films of ZnS and Al:ZnS. The films have low absorption for wavelength greater than 550 nm such property makes

the films suitable materials for thin film solar cell window layers. The optical band gap of the thin films were estimated from absorption data as a function of wavelength by using the stern Relation. The energy band gap and transition type was derived from mathematical treatment of data obtained from optical absorbance versus wavelength with the Stern (1963) give as

$$Ahv = (hv - E_g)^{\frac{n}{2}} \quad \text{.....(1)}$$

Where *hv*, is the photon energy, E_g, the optical band gap and A is a constant.

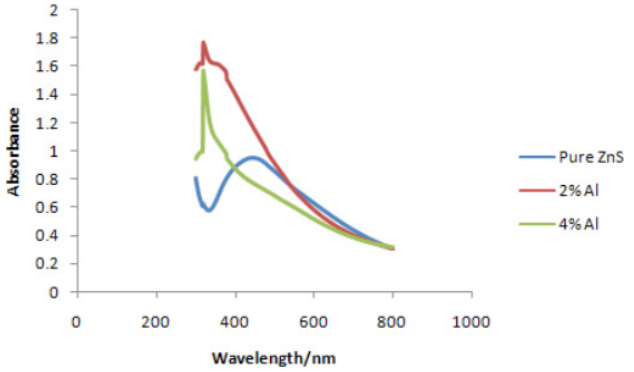

Figure 5 A plot of absorbance versus wavelength of ZnS and Al:ZnS thin films.

By plotting a graph of $(Ahv)^2$ as ordinate and *hv*as abscissa, the optical band gap E_g can be determined by extrapolating the linear portion of $(Ahv)^2$ versus *hv* to the energy axis at $(Ahv)^2=0$.[13] As it obtained from stern relation the band gap energy of pure ZnS is 3.74 eV as shown in Figure 6, where asAl:ZnS deposited at varied concentrations of Aluminium (2%, 4%) are between 3.7 eV and 3.5 eV shown in Figures 7&8 respectively. The results show that the band gap is decreased as the percentage of aluminum is increased.

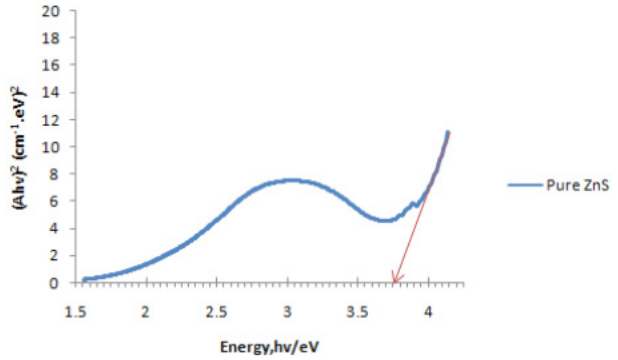

Figure 6 Plot of $(Ahv)^2$ versus photon energy, *hv* showing pure ZnS.

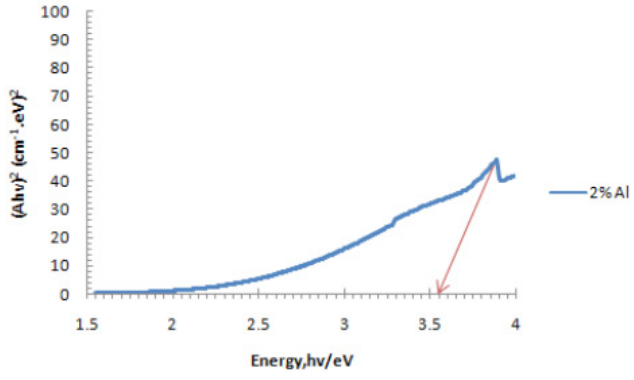

Figure 7 Plot of $(Ahv)^2$ versus photon energy, *hv* with 2% of Aluminium.

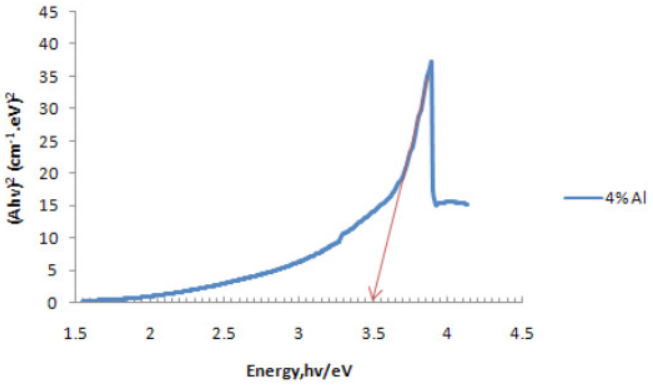

Figure 8 Plot of $(Ahv)^2$ versus photon energy, *hv* with 4% of Aluminium.

Conclusion

An adherent thin film of Al:ZnS has been deposited on silica glass substrates from acidic bath containing Zinc acetate, Aluminium Chloride, and Thioacetamide. EDTA was used as complexing agent to control the free ion concentration of the thin films. The XRD studies show that no intense peaks were observed. SEM micrograph of the as deposited Al:ZnS thin films show the films were uniform, dense, and composed of spherical shaped grains. EDAX results shows the elemental compositional of Al, Zn, and S. The optical absorbance decreased with increasing aluminium concentration. The optical absorbance of the films were also determined from the absorption spectra for wavelength greater than 550 nm. The band gap values were also decreased as the concentrations of aluminium is increased. The optical band gaps value of pure ZnS was 3.74 eV, where as the band gap values of Al:ZnS films were 3.54 eV and 3.5 eV.

Acknowledgments

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Conflicts of interest

Authors declare that there is no conflict of interest.

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