

Influence of thickness of active layer on the performance of CBD grown Cu-CdS/PbS hetero-junction

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Abstract

Solar cells with the structure ITO-Cu-CdS/PbS-Ag were fabricated by heat induced Chemical Bath Deposition (CBD) technique. Cu-doped CdS/PVA nanocomposite thin film of thickness 260 nm was used as the window layer. The PbS absorber layer of different thickness (528.3 -1250.8 nm) for different molar concentration of lead nitrate (0.05, 0.1, 0.15, 0.2 and 0.3 M) was then grown on ITO/CdS to fabricate the junction. The effect of molar concentrations on the optical and structural properties of the corresponding PbS films and solar cells were investigated. The optical bandgap of the PbS films was found to decrease with the increase of the molar concentration. The photovoltaic parameters such as short circuit current, open circuit voltage, fill factor and efficiency of the CdS/PbS solar cells were evaluated from the J-V characteristics under one sun illumination intensity (100mW/cm²). The changing molar concentration enhanced the performances of the cells and a highest efficiency (1.38%) obtained at 0.3M molar concentration. Copyright © 2016 VBRI Press.

Keywords: CBD, molar concentration, solar cell, power conversion efficiency.

Introduction

Thin film semiconductors from IV-VI group have been drawing attention over the past few years in the fabrication of various hetero-junction solar cells [1-2] as it can provide high absorption coefficient in visible and near infrared region and secondly, a smaller quantity of material is needed for complete absorption of solar radiation and thus leading to lower cost [3]. Lead sulfide (PbS) is an attractive binary semiconductor material from IV-VI groups with a narrow band gap of ~ 0.41eV at 300K. It has relatively large excitation Bohr radius (EBR) of 18nm [4]. Band gap in PbS is very sensitive to grain size and thus making it a good candidate for nanostructured devices [5]. Recently, multiple exciton generation has been detected in PbS quantum dot (QD) and thus rendering it a potential candidate for solar cell applications [6, 7]. PbS thin films can be deposited by a variety of methods such as electrodeposition [8], chemical bath deposition (CBD) [9,10], sonochemical [11], spray pyrolysis [12] and successive ionic layer adsorption and reaction method [13]. However, among these, CBD technique is one of the simple and energy-efficient techniques to synthesize PbS thin film as it is easy to handle, convenient for large area deposition, cost effective and does not require any special set-up or sophisticated instrument [14, 15]. So far, different morphologies of PbS nano structures were obtained such as quantum dots (QD), nanorods, nanotubes and nano-

multi-pods [11, 16-19]. However, the most successful nanocrystals used in solar cells and photoconductors are QDs. Because, QDs has large absorption cross-sections and so incoming light can be efficiently absorbed and converted into electron-hole pair. Moreover, the thickness of the active layer plays an important role in the efficiency of thin film solar cell as it is closely related to the absorption co-efficient of the material. The minimum thickness of the absorbing layer formed by the photovoltaic material in a solar cell is of the order of the inverse of the optical absorption coefficient. The optical, structural and electrical properties are also greatly influenced by thickness. Therefore, determination of the optimum thickness of active layer with suitable optical, structural and electrical properties is an important issue in the fabrication of thin film solar cell.

There are only a few reports on the fabrication of CdS/PbS hetero-junction in which PbS QDs or PbS nanorods has been employed as an active layer for harvesting light energy. Z. Wang *et al.* [20] reported the synthesis of MDMO-PPV capped PbS nano-rods and applied as acceptor in the active layer of bulk heterojunction solar cell. P. Khagendra *et al* [21] reported the first heterojunction solar cells based on PbS-QDs and RF magnetron supported CdS thin film, demonstrating an achieved efficiency of 3.3%. Fabrication and synthesis of n-CdS/p-PbS heterojunction solar cells using microwave-assisted chemical bath deposition technique was reported by Obid *et al* and they achieved a maximum efficiency of

1.68 [22]. M. J. Choi *et al* showed that simple oxidation of PbS QDs at the PbS/metal interface significantly improves the device performance and air stability in schottky junction QDSCs [23]. However, fabrication of high efficiency and low cost solar cells is still a challenge for the scientists.

In the present work we have fabricated a glass/ITO/Cu-CdS/PbS/Ag heterojunction fully grown by CBD technique without using any sophisticated equipments and hence it may reduce the production cost. Cu-CdS/PVA thin film was used as a window layer and CBD grown PbS thin film as the absorber layer. We have deposited CdS/PVA window layer employing previously reported heat induced thermolysis technique [24]. But, the presence of PVA in CdS/PVA thin film may cause low order of conductivity due to dielectric behavior of PVA [25, 26]. In order to improve the performance of the devices, doping is a widely used method as doping with a suitable impurity concentration could effectively reduce the materials resistivity and as well as affect the band gap energy and crystalline structure. CdS can be doped with different transition metals ions such as Mn^{2+} , Fe^{2+} and Cu^{2+} to improve the electrical conductivity of the CdS/PVA nanocomposite thin films as PVA has dopant dependent electrical and optical properties [27-29]. Further, the Influence of lead Nitrate concentration on the particle size, thickness and properties of PbS thin film is investigated for improving the efficiency of Cu-CdS/PbS solar cell.

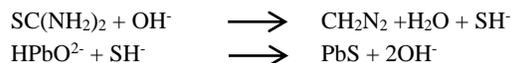
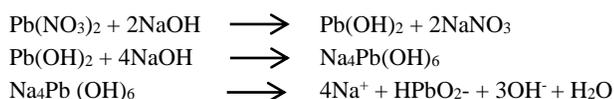
Experimental

Materials details

Cadmium acetate [$Cd(CH_3COO)_2 \cdot 2H_2O$], lead Nitrate, thiourea [$CS(NH_2)_2$], polyvinyl alcohol(PVA), sodium hydroxide [NaOH] and Silver (Pure) were purchased from Merck (India) Ltd. The reagents were used directly as received. Deionized water was used throughout the experiments.

Material synthesis

The PbS thin films were grown using a simple Chemical bath deposition technique. For the deposition of PbS thin films, lead nitrate and thiourea were used as the source of Pb^{2+} ion and S^{2-} ion respectively; sodium hydroxide was used as the complexing agent. In a typical reaction, 1M NaOH solution was added drop wise into 10 ml aqueous solution of lead nitrate with constant stirring. On addition of NaOH, initially, a white precipitate was observed due to the formation of $Pb(OH)_2$. The precipitate then dissolved gradually as more NaOH was added. Eventually, the precipitate was totally disappeared and the solution turned into clear and transparent when all $Na_4Pb(OH)_6$ complex ion was dissolved in the solution. A set of five reaction baths were prepared for five different molar concentrations of lead nitrate such as 0.05M, 0.1M, 0.15M, 2M and 3M.



For the deposition of PbS thin films, chemically clean glass substrates were dipped into the reaction bath containing the precursors and then subjected to heat treatment at 60°C in a microprocessor controlled digital oven. At the beginning, the deposition bath was colourless and in due course of time its colour has changed from brown to black within 15-20 minutes. After 30 minutes, the substrates were covered with a shiny brown film. The coated substrates were subsequently taken out, rinsed thoroughly with distilled water, dried in an oven at 60°C for 10 minutes. The resulting films were homogeneous, well adherent to the substrate. The prepared samples were abbreviated as S1, S2, S3, S4 and S5 for lead nitrate concentrations of 0.05M, 0.1M, 0.15M, 0.2M and 0.3M respectively. Thickness of the film was found to increase from 528.23nm to 1250.8 nm as the molar concentrations of lead nitrate was gradually increased from 0.05 M to 0.3 M. However, beyond 0.3 M concentration of lead nitrate, the film quality deteriorates.

Device fabrications

To complete the solar cell, indium tin oxide (ITO) coated glass slides ($<15 \Omega/sq$. Zhuhai Kaivo Electronic components C. Ltd.) were thoroughly washed with a mixed solution of deionized water, acetone, and dilute HCl solution (volume ratios of 1:1:1) under sonication for 60 min. CdS/PVA nanocomposite thin film (thickness 260 nm) were then deposited on the top of ITO by heat induced thermolysis technique as reported in our earlier publication [24]. The CdS coated ITO substrates were then dipped into 0.1M $Cu(NO_3)_2$ solution for 2 minutes, taken out of the bath and heated for 1hour at 60°C. The PbS layer was grown onto the Glass/ITO/Cu-CdS substrate by the CBD techniques as described above. A set of five cells (C1 to C5) were fabricated by using PbS thin film of different thickness (ranging from 528.3 nm to 1250.8 nm) to investigate the effect of thickness of PbS layer on the efficiency of the cell. Finally, silver (Ag) metal was deposited over the PbS thin films via thermal evaporation method under 10^{-5} torr pressure using HIND High Vacuum Coating Unit (M:12A4D). The active area of the device was $1 \times 1 cm^2$.

Results and discussion

Optical properties

The UV- Vis absorption spectra of the PbS thin films (S1 to S5) are shown in Fig. 1A. The overall absorbance in the PbS thin films (in the wavelength range 450-750nm) were found to increase when the film thickness was increased from 528.3nm to 1250.8nm. However with the increase of thickness, the absorption peak is shifted to the higher wavelength side (red shift). The observed red shift could be attributed due to the increase in crystallite sizes of the films with the increase of film thickness.

The transparency of the PbS (S1 to S5) film were in the range from 10% to 23% in the near IR region. However in the visible range the film is almost opaque. This indicates

that, as prepared PbS films are good absorber layer in the wavelength range 450-750nm and as such can be considered as potential materials for solar cell applications. The optical band gaps (E_g) of the PbS thin films were obtained by using the following Eq. (1) for a semiconductor [30],

$$\alpha = K (h\nu - E_g)^{m/2} / h\nu \quad (1)$$

where, α is the absorbance, K is a constant and m is equal to 1 for direct transition and 2 for indirect transition. Linearity of the plots of $(\alpha h\nu)^2$ versus photon energy for the PbS thin films indicate that the material is of direct band gap type.

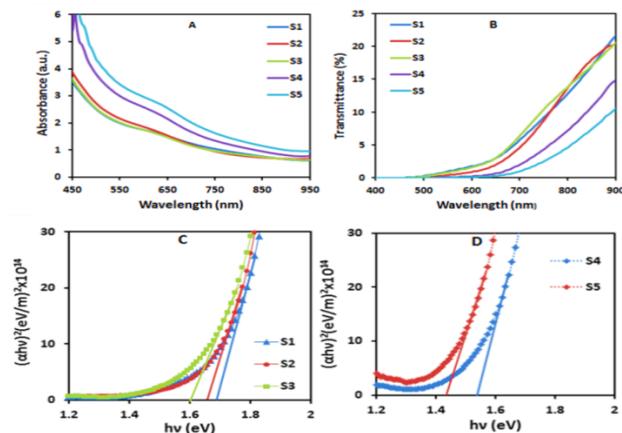


Fig. 1. (A) UV-vis absorption spectra of PbS thin films (B) Transmittance spectra of PbS thin films (C-D) energy band gap values of PbS thin films.

The extrapolation of the straight line to $(\alpha h\nu)^2 = 0$ axis gives the energy band gap (E_g) of the film material. The E_g values obtained are summarized in **Table 1**. From **Table 1**, it is observed that, band gap of PbS thin films (S1 to S5) are in the range 1.67 - 1.42 eV and are red shifted compared with the bulk PbS band gap of ~0.41 eV.

Table 1. Band gap energy (E_g) of PbS thin films estimated from the absorption spectra.

PbS film	Molar conc. (Lead Nitrate)	Molar conc. (Thiourea)	Thickness (nm)	Energy band gap (eV)
S1	0.05M	0.4M	528.3	1.67
S2	0.1M	0.4M	646.7	1.65
S3	0.15M	0.4M	769.6	1.6
S4	0.2M	0.4M	963.6	1.52
S5	0.3M	0.4M	1250.8	1.42

XRD analysis

The X-Ray diffractogram of CBD PbS thin films of different thicknesses are shown in **Fig. 2**. The presence of different peaks at 2θ values of 25° , 30° , 43° , 50° , 62° and 71° corresponds to reflections from (111), (200), (220), (311), (400) and (420) planes of cubic structure of PbS, which coincide well with the JCPDS data [05-0592]. The presence of sharp structural peaks in the XRD spectra reveals that the films are polycrystalline in nature. Impurity phases (such as elemental Pb, S, PbO and SO₂) were not detected in the XRD pattern, confirming the purity of PbS thin film.

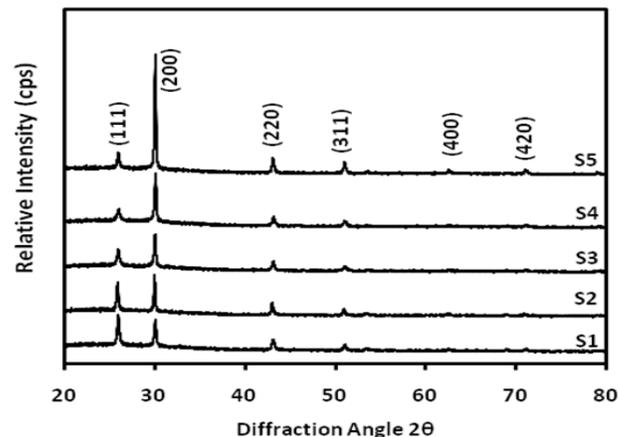


Fig. 2. XRD spectra of PbS thin films deposited at various molar concentrations of lead nitrate.

The intense peak at 2θ value of 30° confirms the presence of PbS compound and clearly indicates the significant quantities of Pb²⁺ and S²⁻ ions. As the thickness increases the diffraction peaks are not shifted either in the direction of lower or higher 2θ values, thereby it has been concluded that the PbS thin films have the same composition irrespective of their thickness. In case of sample S1, the intensity of (111) plane is a little higher than the intensity of (200) plane. A gradual and significant increase of intensity of the (200) plane in the PbS thin film was observed with the increase of film thickness. Two new peaks (400) and (420) at 2θ values of 62° and 71° respectively have also appeared in the PbS thin film of higher thickness (S5). The increase of peak intensity and emergence of new peaks may be due to thickness induced grain growth.

Surface morphology analysis

The SEM micrographs of the PbS thin films deposited at different lead nitrate concentrations are shown in **Fig. 3(A-E)**.

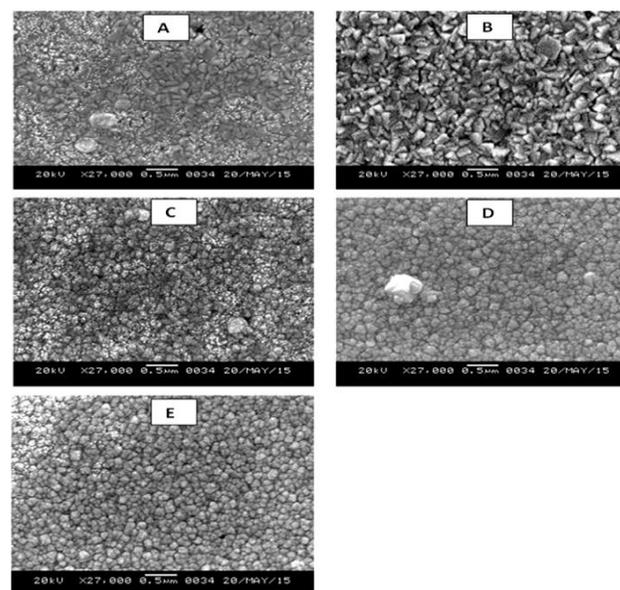


Fig. 3. SEM micrographs of PbS thin films deposited at different lead nitrate concentrations: (A) 0.05M, (B) 0.1M (C) 0.15M (D) 0.2M (E) 0.3M.

The film morphology shows a clear dependence on lead nitrate concentrations. Samples prepared at lower molar concentrations of lead nitrate (0.05M, 0.1M) are composed of non-uniform clusters with well-defined boundaries [31, 32]. On the other hand, uniform and compact PbS films were produced when the lead nitrate concentration was increased from 0.15M to 0.3M.

Device performance

The current–voltage (I–V) behavior of the as fabricated glass/ITO/Cu-CdS/PbS/Ag cells (C1 to C5) was recorded by Keithley 2400 source meter under one sun illumination (Fig. 5). The five different cells (C1 to C5) have the same device architecture as shown in Fig. 4; however, the thickness of the active layer (PbS) was varied from 528.3 nm to 1250.8 nm. Table 2 summarizes the photovoltaic parameters [fill factor (FF), short-circuit current density (J_{sc}), open-circuit voltage (V_{oc}), and efficiency (η)] of glass/ITO/Cu-CdS/PbS/Ag cells fully developed by CBD technique.

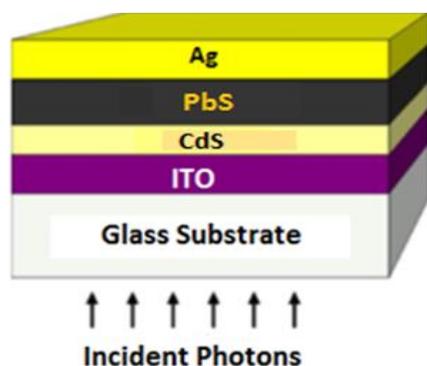


Fig. 4. Structure of CdS/PbS solar cell.

The results show the positive impact of increasing the thickness of the PbS active layer on the cell parameters and on overall efficiency of the cell. The efficiency (η) of the glass/ITO/Cu-CdS/PbS/Ag solar cells was found to increase from 0.52% to 1.38% with the increase of PbS film thickness from 528.3 nm to 1250.8 nm, where the highest value was obtained for the sample with 0.3 M molar concentration having a lower energy gap, while the lower efficiency value was achieved for the cell with 0.05 M. It is observed that when the molar concentration increases, the energy gap decreases which led to increase of electron hole pair [33]. The power conversion efficiency of the cells is also greatly affected by the series and shunt resistances. An ideal cell should have a series resistance close to zero and shunt resistance close to infinity. The calculated values of series resistances and shunt resistances of the cells (C1-C5) are included in Table 2. In the present study, high series resistance ($\sim 24.0 \times 10^3 \Omega$) and low shunt resistance ($\sim 60 \times 10^3 \Omega$) was found for the cell with 0.05M molar concentration and exhibited the lower η of 0.52%, while low series resistance ($\sim 7.29 \times 10^3 \Omega$) and high shunt resistance ($\sim 198.1 \times 10^3 \Omega$) was observed for the cell with 0.3 M molar concentration which demonstrated the maximum η of 1.38% with corresponding values of short circuit current density (J_{sc}), open-circuit voltage (V_{oc}) and fill

factor (FF) are 9.2 mA/cm², 285 mV and 53% respectively.

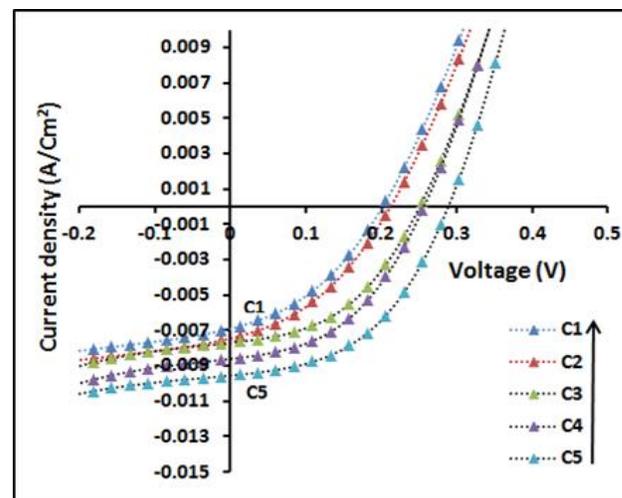


Fig. 5. Current-Voltage characteristics of CdS/PbS solar cells.

Table 2. Summary of device performance for different PbS active layer thicknesses under 100 mWcm⁻² simulated solar illumination.

Cells	Thickness of PbS layer (nm)	Voc (mV)	Isc (mA cm ⁻²)	Fill Factor (%)	Efficiency (%)	Series Resistance (Ω)	Shunt Resistance (Ω)
C1	528.3	205	7.0	37	0.52	24.0×10^3	60×10^3
C2	646.7	215	7.5	39	0.63	20.58×10^3	71×10^3
C3	769.6	241	7.9	46	0.91	18.4×10^3	91.743×10^3
C4	963.6	252	8.6	48	1.05	11.23×10^3	129.36×10^3
C5	1250.8	285	9.2	53	1.38	7.29×10^3	198.1×10^3

Conclusion

We report the fabrication of Cu-CdS/PbS heterojunction solar cells based on CBD technique, demonstrating an achieved efficiency of 1.38%. The effect of molar concentration of lead sources on the optical and structural properties of PbS thin films were investigated with the help of UV-Vis spectroscopy, XRD and SEM analysis. The thickness of PbS thin film was found to increase from 528.3nm to 1250.8nm as the molar concentration increases from 0.05 to 0.3 M and this may be due to increase of ions reaction with increasing molar concentrations. XRD analysis reveals that all the prepared thin films are polycrystalline. However, the PbS thin film prepared at 0.3 M molar concentrations has a good crystalline quality. The overall absorbance in the PbS thin films (in the wavelength range 450-750nm) were found to increase with the increase of film thickness. But with the increase of film thickness, the absorption peak is shifted to the higher wavelength side and it could be attributed due to the increase in crystallite sizes with increasing film thickness. To understand the influence of thickness of PbS layer on the energy conversion process, various cells with varying active layer thicknesses were compared. It is observed that, the device efficiency increases with increasing PbS thickness (by achieving a little high J_{sc}) and at the same time, increasing V_{oc} (due to increasing absorption in the long wavelength region). The maximum efficiency (1.38%) was achieved for the PbS thickness of 1250.8 nm.

Acknowledgements

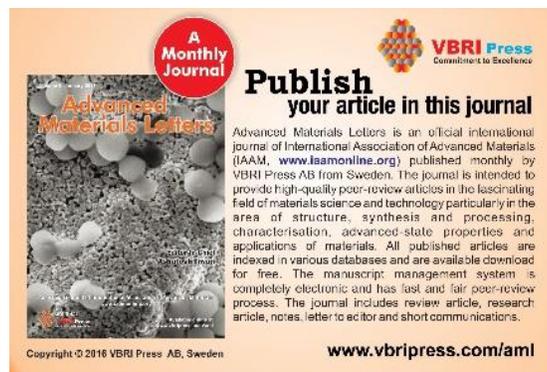
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Author's contributions

DS, PP - Performed the experiments, PP, MRD - Data analysis and DS-Wrote the paper. It is declared that Authors have no competing financial interests.

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