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Synthesis of sphere-like-crystal CdS powder and thin films using chemical residue in chemical bath deposition (CBD) for thin film solar cell application

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ABSTRACT

CdS microcrystal, with sphere like morphology, was synthesised from the residue obtained after the chemical bath deposition (CBD) of CdS thin layers usually deposited for CdTe and/or CIGS thin film solar cells, specifically by recycling the chemical waste. The residue was collected and reused to deposit cadmium sulphide (CdS) thin films on glass substrate by thermal evaporation. The films were subsequently annealed in vacuum at 250 °C, 350 °C and 420 °C for 40 min. The structural and optical properties were characterized by XRD, SEM, and UV–Vis spectrometry. All the films were found to exhibit high transmittance (over 70 to 90%). The optical band gap energy was found to be in the range of 2.28–2.37 eV, which is very suitable to be used as buffer or window for CdTe or CIGS thin film solar cells.

1. Introduction

Cadmium sulphide (CdS) is a group II-VI compound semiconductor material with a direct optical band gap of 2.482 eV at room temperature (Martienssen and Warlimont, 2006). It has high optical transmittance and ease of deposition making it very attractive for optoelectronic device applications. CdS grows intrinsically as n-type semiconductor (Richter et al., 2013; Oliva, 2001). CdS thin films are widely used in solar cells as the window layer in cadmium telluride (CdTe) recording efficiencies of 21.4% for cells and 17.5% for modules (Green, 2016). It is also used as buffer layer in copper indium gallium selenide, CuInGaSe$_2$)CIGS(based hetero-junction solar cells with efficiencies of 21.6% for cells and 18.7% for modules (Green, 2016; Chopra et al., 2004).

CdS buffer layer shows high window functionality for emerging new thin film PV technologies such as CZTS thin film solar cells with recent efficiencies 12.6% (Jean, 2015; Tao, 2016).

Several techniques have been used to deposit CdS thin films including chemical bath deposition (CBD) (Shah, 2012), sputtering (Lee and Lee, 2007), close spaced sublimation (CSS) (Ferekides, 2000), and thermal evaporation (Sahay et al., 2007). Even though the highest efficiency solar cells were fabricated using CdS by CBD, this technique is not suitable for large-scale production and involve a fairly slow process (Romeo, 2004). CBD also produces hazardous waste that needs to be recycled or safely disposed to safeguard the environment. Recovering or recycling the CdS from the CBD waste has many advantages from economic and environmental point of view.

In this work CdS crystal, with sphere-like morphology, was first synthesized using the chemical residue from CBD process, as the chemical bath deposition (CBD) waste was recycled. The prepared CdS crystal was used to deposit CdS thin films by low vacuum thermal evaporation method. Thermal evaporation is considered as a simple and low cost technique compared to other thin film deposition techniques. In window layer applications, it is desirable that the CdS layer should be as thin as possible (Morales-Acevedo, 2006), where preferable thickness is between (50–100) nm (Durose et al., 1999). In this work, CdS films with 50 nm and 60 nm thicknesses were obtained and characterized.

2. Methodology

Commercially available cadmium sulfate ($CdSO_4$), Thiourea ($SC(NH_2)_2$), ammonium hydroxide (NH_4OH), and ammonium sulfate

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$(NH_4)_2SO_4$ were used without further purification (Sigma-Aldrich Co.). The deposition of CdS thin films was then carried out by chemical bath deposition (CBD) with the following the process. 0.002 M of $CdSO_4$, 0.006 M of$(NH_4)_2SO_4$was weighed and then mixed in the aqueous alkaline solution of NH_4OH and then 0.05 M of thiourea, SC $(NH_2)_2$ was added to the aqueous solutions and stirred constantly. The reaction temperature was controlled within the range of (338–353) K and the reaction time was set to 20–30 min. More details of the prepared CdS thin film using CBD method was discussed elsewhere (Yusoff, 2015). At the end of the process, the orange precipitate of CBD waste was collected. This precipitate was separated by centrifugation, washed several times with ethanol and distilled water, and then dried at 60 °C under vacuum overnight. The resultant CdS powder was used for preparing CdS thin films using thermal evaporation technique on cleaned soda lime glass substrate under low vacuum. The glass substrates used in the deposition were ultrasonically cleaned in acetone, ethanol, and de-ionized water successively and finally dried with N_2 before used.

Thermal evaporation is a simple technique for depositing material onto a substrate as thin films. It uses an electric resistance heater and has tungsten boat to heat the source material. The temperature of the boat increases with the increase in current eventually heating the source material which starts to evaporate. Hence, CdS is deposited using the thermal evaporation method and the entire process is done in low vacuum chamber allowing the molecules to evaporate freely in the chamber and they subsequently condense on the substrate as CdS thin films. The deposition parameters of CdS are mentioned in the following Table 1.

Thermally evaporated CdS thin films were annealed under vacuum at 250 °C, 350 °C, and 420 °C for 40 min, respectively. Table 2 summarizes the sample description for the CdS thin films that were annealed at different temperatures.

The structural investigation of CdS powder was carried out using X-ray powder diffractometer (Bruker AXS Germany, D8 Advance) with Cu - $K_\alpha$radiation wavelength λ = 1.5406 Å and angle (2θ = 20° to 60°) at room temperature. Grain size, surface morphology and cross-sectional view were observed by using Carl Zeiss Merlin field emission scanning electron microscope (FESEM) which was operated at 3 kV. The electrical parameters such as carrier concentration, mobility, and resistivity were measured by Hall Effect measurement system, HMS ECOPIA 3000 with a magnetic field of 0.57 T and UV–Vis spectrometry (Perkin-Elmer Lambad 35) in the wavelength range of 300 nm to 900 nm.

3. Results and discussion

Fig. 1 shows the attained cubic zinc-blende structure of CdS powder with space group *F-43m (2 1 6),* which was expected as it is commonly known that low temperature CBD CdS thin films were often found in cubic phase (Lisco, 2015; Mariappan, 2012). The peaks match to the JCPDS card numbered 01–0890440, where the peaks are associated with the (1 1 1), (2 2 0), and (3 1 1) planes.

The mean crystallite size (D) of CdS powder was also calculated using Scherrer equation as given in Eq. (1). The micro-strain (ε) was calculated from the XRD peaks using Eq. (2) (Guinebretière, 2013).

Table 1
Thermal evaporation deposition parameters.

Thermal Evaporation Parameters	Value/Range
Source	45 mg CdS recycled powder
Substrate	Soda lime glass
Boat to substrate distance	5 cm
Deposition current	25 A
Deposition Time	20 min
Deposition pressure	5 Pa

Table 2
Summary of annealing process parameters.

Sample ID	Annealing Temperature °C	Time of annealing min
As deposited	–	–
A	250	40
B	350	40
C	420	40

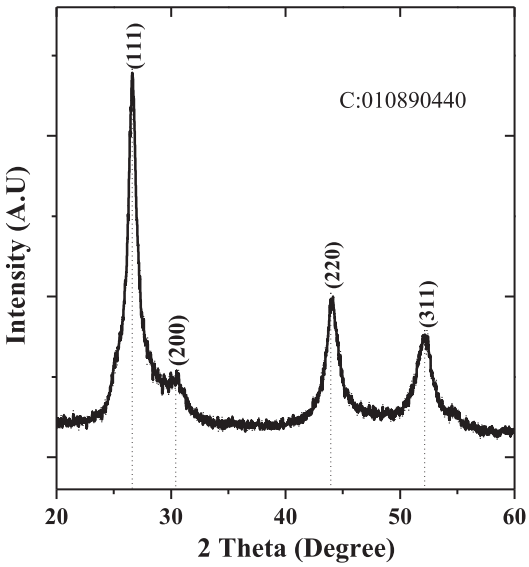

Fig. 1. XRD pattern of recycled CdS powder.

$$D = \frac{0.9\lambda}{\beta cos\theta} \quad (1)$$

$$\varepsilon = \frac{\beta}{4tan\theta} \quad (2)$$

where

D: the crystallite size,
λ: X-ray wavelength (1.54 Å),
β: Full width at half maximum (FWHM) (rad).

The crystallite size and micro-strain final calculations were done using the TOPAS (BrukerAXS, 2008) software. Results are summarised in Table 3 with goodness of fit (GOF = 1.31).

The corresponding lattice parameter of the CdS film as stated in the JCPDS card no. 01-0890440 is found to be a_0 = 5.80537 Å. The corresponding formulae for the lattice parameter of a cubic crystal system with inter-planar distance, *d*, is shown in Eq. (3). Using the formulae, the lattice parameter here was found to be a = 5.8295 Å (Pecharsky and Zavalij, 2009).

$$a^2 = d^2(h^2 + k^2 + l^2) \quad (3)$$

FESEM images in Fig. 2(a)–(d), show that CdS microstructure was

Table 3
Parameters from XRD calculation.

2° θ	Miller indices	Crystallite size (nm)	Microstrain ($\times 10^{-3}$)
26.6	)111(	26.4	17.5
44.1	(220(	54.4	14
52.1	(311)	87	15.5