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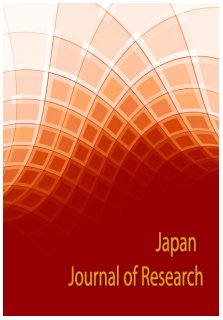

Pressure-Engineered CdS Thin Films via CBD for Enhanced Solar Cell Performance

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Abstract

Cadmium sulfide (CdS) thin films are used as window material in photovoltaic solar cells technology, where their optical transmittance, crystallinity, and surface morphology significantly influence overall device performance. In this study, we report a novel approach to modulate the structural and optoelectronic properties of CdS thin films by varying the reactor pressure during Chemical Bath Deposition (CBD) process. CdS films were deposited on SnO_2:F (FTO) substrates in areas of 100 cm^2. Comprehensive characterization using X-ray diffraction (XRD), scanning electron microscopy (SEM), UV-Vis spectroscopy and Photoluminescence measurements reveal a clear correlation between reactor pressure with optoelectronic properties. Notably, CdS films deposited at elevated pressure exhibited a good crystalline property, atomic force microscopy (AFM) revealed non-uniform morphology on samples. The pressure inside the reactor container led to good CdS growth with thickness around 40 nm to 90 nm an electrical resistivity value around of 10^3 Ω.cm. Photovoltaic CdTe solar cells were developed by using the CdS thin films obtained as a window layer. The electrical properties of the devices were significantly enhanced and photovoltaics efficiency increased from 8% to 15% because of increased pressure within the reactor. This study demonstrates that the reactor pressure is a powerful and previously underexplored parameter in CBD process, providing a scalable and cost-effective strategy for improving the efficiency of CdS based solar cells.

Introduction

Cadmium sulfide (CdS) thin films have interest in photovoltaic and optoelectronic applications, often serving as buffer or window layers due to their favorable direct bandgap (2.4 eV) and high optical transparency. The chemical bath deposition (CBD) method remains one of the most prevalent and cost effective techniques for producing uniform, large-area CdS films, benefiting from its simplicity, low temperature operation, and scalability. Historically, CBD of CdS has relied on aqueous alkaline solutions containing a cadmium salt, a sulfur precursor (commonly thiourea), and a complexing agent to control release of Cd^{2+} ions generating controlled supersaturation and high-quality film growth [1].

While most research has focused on the influence of bath composition, temperature, pH, and deposition time, variations in pressure conditions particularly in sealed or pressurized systems remain an underexplored parameter with significant potential. Empirical evidence shows that conventional CBD parameters, such as temperature and deposition time, have a pronounced impact on film structure and properties. For instance, extended deposition times have been reported to induce phase transitions from cubic to hexagonal structures and to modify optical bandgap values [2]. Moreover, post deposition treatments like annealing and substrate arrangement have been shown to improve crystalline quality and morphological uniformity [3,4]

By contrast, pressure, especially above ambient atmospheric pressure, has been largely neglected in CBD discussions. However, insights from analogous hydrothermal or autoclave assisted syntheses suggest that elevated pressure can increase precursor solubility, raise reaction kinetics, and promote denser, more adherent films. Although specific studies on pressure- controlled CBD of CdS are sparse, exploring this parameter could provide novel pathways to tailor film morphology, crystalline, and electronic properties.

Accordingly, this paper aims to systematically investigate the effects of pressure during the CBD process of CdS thin films considering two chemical processes, the first one traditional CBD technique with an open container and the second one a sealed container inside thermostatic bath. By comparing films to grow under atmospheric condition, ambient environment ,

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and elevated pressure in sealed bath, we systematically analyze variations in morphology, crystalline phase, optical properties and electrical characteristics. These insights are expected to reveal new tunable pathways for optimizing CdS thin films for next generation optoelectronic applications.

Experimental Details

CdS thin films were deposited on Fluorine doped tin oxide (SnO_2:F) coated sodalime glass substrates (FTO). Prior to deposition, the substrates were treated with a 0.1 M HCl solution. Cadmium sulfide (CdS) thin films were deposited using the chemical bath deposition (CBD) technique, employing $CdCl_2$ (0.1 M) and thiourea, $SC(NH_2)_2$ (0.3 M), as precursor solutions. To facilitate the formation of complex compounds, NH_4Cl (0.2 M) and NH_4OH (2 M) were added to the solution. The CdS films were deposited as monolayers (M) and bilayers (B) onto FTO in areas of 100 cm^2 at 70 ± 2 °C. Two processes were used for CdS layers; the first involved CdS deposit as bilayers using the traditional CBD (CdS_T) with an open solution container, where substrates were positioned vertically inside the container. The deposition time was 8 min and 2.8 L of precursor solution was used. The second process consisted of a hermetically sealed container (H) containing 500 mL of precursor solution. The FTO substrates were arranged horizontally at the bottom of the reactor container. This setup was placed inside a thermostatic bath with a deposition time of 4 min, for this process, the CdS monolayers and bilayers will be referred to as CdS_{HM} and CdS_{HB}, respectively. After deposition, all CdS samples were thermally annealed in air at 450°C for 60 min.

The film thicknesses were determined using a contact profilometer (Sloan Dektak II). Optical transmittance measurements were carried out with a Shimadzu UV-Vis spectrophotometer (model UV 2401-PC). Electrical properties were assessed using a 6517B electrometer/high- resistance meter. Structural characterization was performed via X-ray diffraction (XRD) using a Bruker D8 diffractometer equipped with a Cu-Kα radiation source (λ = 1.5418 Å). Photoluminescence (PL) spectra were recorded using a SPEX 500 spectrometer coupled to a Hamamatsu photomultiplier. A He-Cd laser (Kimmon Koha IK3102R-G) with an excitation wavelength of 325 nm and an output power of approximately 50 mW was used as the excitation source. Surface morphology was examined by scanning electron microscopy (SEM) using a JEOL JSM7X series microscope operated at an acceleration voltage of 5 kV, providing a resolution of up to 100 nm. Elemental composition was analyzed through energy-dispersive X- ray spectroscopy (EDS), employing a detector with a spectral resolution of 129 eV and a sensitivity of 0.001 cps/eV, using a 10 kV acceleration voltage. Both K- and L-lines were utilized for elemental identification. The working distance ranged from 9 to 10.1 mm, with a magnification of up to 60,000×. PB-ZAF correction was applied to improve quantification accuracy. Current–voltage (I–V) characteristics were measured using a Sol3A Class AAA solar simulator (IEC/JIS/ASTM standards) equipped with a calibrated Xenon lamp delivering 100 mW/cm² under AM1.5 illumination conditions. Spectral response and external quantum efficiency (EQE) were obtained using a QEPVSI-b system from Oriel Instruments.

Results

Morphological properties

Figure 1 shows the SEM and AFM images of the obtained CdS films. Figure 1a presents the FTO substrate. Figure 1b shows a uniform CdS film deposited using a traditional CBD technique. Figures 1c and 1d show a CdS monolayer and bilayer deposited under higher pressure inside the reactor container, respectively. A non-uniform surface was obtained with white grains associated with CdO, and CdS grains size less than 100 nm. The presence of CdO was further confirmed through X-ray diffraction (XRD) analysis. On the other hand, AFM images show three-dimensional morphology. Figure 1e, presents the FTO morphology, Figure 1f shows the CdS uniform growth using conventional CBD, while Figures 1g and 1h illustrates how increased reactor pressure substantially modifies the morphology, resulting in less uniform films with large cavities on the order of micrometers.

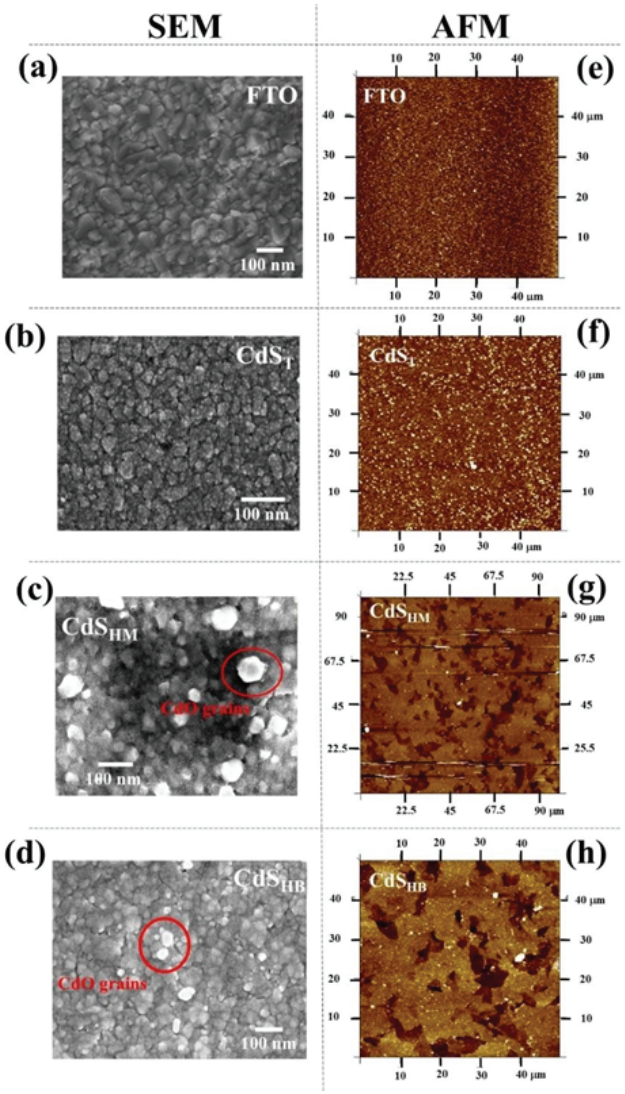

Figure 1. *SEM and AFM images for FTO substrate (1a and 1e) respectively and all CdS samples obtained using an open container under traditional and conventional CBD technique (CdST) (see b and f) and a hermetically sealed container placed in a thermostatic bath (see c, d, g and h) respectively.*

To detect the presence of Cd, S and O elements in each sample, energy-dispersive X-ray spectroscopy (EDS) was performed over the entire surface of the CdS thin films and on the white grains enclosed in a red circle. For the CdST sample, a Cd content of 61% and a sulfur content of 39% were obtained.

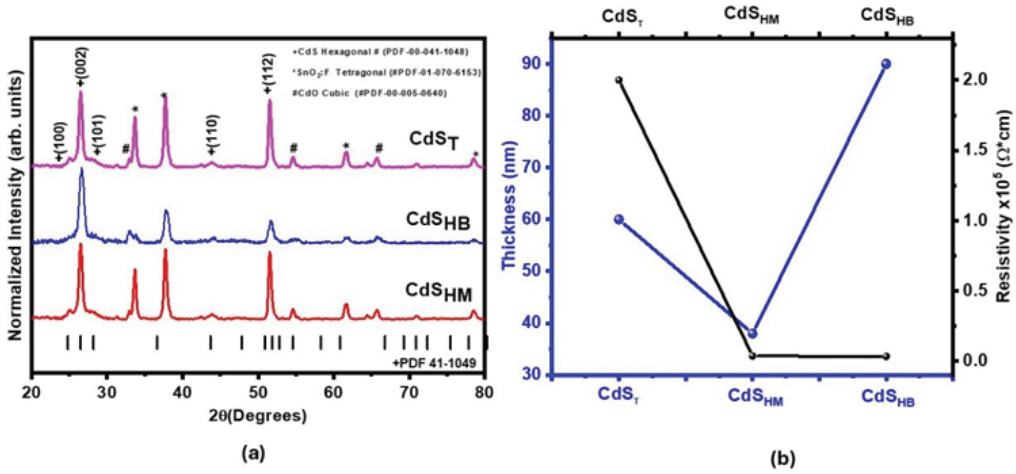

Figure 2. *(a) Diffraction patterns and (b) Thickness and electrical properties of CdST, CdS_{HM} and CdS_{HB} respectively.*

Analyzing the mono layers and bilayers of CdS a Cd content of 65 % and a sulfur content of 35 % were obtained respectively. The white grains correspond with CdO compound, with a Cd content of around 44% and 56% of oxygen.

Crystalline and electrical properties

Figure 2 shows the XRD diffractograms of all CdS samples. The diffraction peaks observed in each sample were indexed using the following standard crystallographic reference patterns: PDF# 41-1049 for hexagonal Wurtzite-type CdS, PDF# 05-0640 for cubic CdO, and PDF# 41- 1445 for tetragonal SnO_2, all of which are star-quality standards.

A Comparison of the experimental X-ray diffraction (XRD) patterns with the simulated diffraction pattern of CdS reveals that the experimental films exhibit strong diffraction peaks at 2θ = 26.6°, corresponding to the (002) plane and at 2θ = 52° corresponding to the (112) plane of hexagonal wurtzite CdS. The sharp and narrow nature of these peaks indicates a high degree of crystallinity in the films deposited. For the CdS_T and CdS_{HM} samples, additional intense peaks are observed at 2θ = 33.8° and 37.8°, characteristic of SnO_2, suggesting the presence of this secondary phase. Moreover, weaker peaks corresponding to the (100) and (101), planes of hexagonal CdS are also present, confirming the polycrystalline nature of the films.

For the CdS_{HB} sample, the intensity of the (112) diffraction peak decreases, indicating a possible preferential orientation along the (002) plane, reflecting a degree of texture development in the film.

Additionally, four extra peaks corresponding to face-centered cubic CdO are detected at (111), (200), (220), and (311), suggesting the formation of a minor CdO phase, likely due to oxidation during deposition or drying. A comparative analysis of the two deposition systems indicates that the newly implemented deposition method produces CdS thin films with crystallinity comparable to or even superior that obtained using the conventional chemical bath deposition (CBD) technique. Importantly, this approach significantly reduces the volume of chemical waste generated, providing a more sustainable and environmentally friendly alternative.

The relative differences in peak intensities may also indicate partial recrystallization or further texture development along the (002) plane, potentially influencing the crystalline quality of the films. Such orientation dependent structural variations are relevant for optimizing thin film properties in photovoltaic applications, where crystal alignment can significantly affect carrier transport and optical absorption.

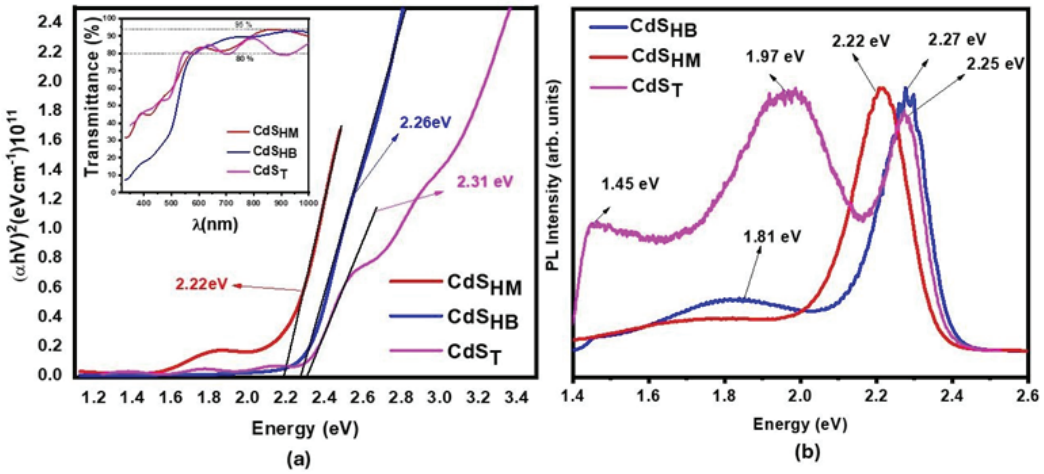

Figure 3. *(a) UV-Vis and (b) Photoluminescence (PL) spectra for all CdS samples.*

Optical Properties

Figure 3 presents the UV-Vis and photoluminescence (PL) spectra for all CdS samples. The inset in Figure 3a shows the transmittance values range from approximately 80% to 90%. The CdS_{HB} sample displays a well-defined absorption edge indicating enhanced polycrystalline characteristics (see Figure 3a). The optical band gap values were calculated, obtaining values from 2.22 eV to 2.31 eV.

The PL spectra (see Figure 3b) were recorded at 10 K using an argon laser with an excitation wavelength of 488 nm (254 eV). The laser beam was focused onto the sample through a cylindrical lens to minimize heating effects. Both the CdSHM and CdSHB samples exhibit well- defined emission bands centered between 2.22 and 2.27 eV. These bands, which closely match the CdS band gap energy (E_g), are commonly referred to as the "green band" and are attributed to band-to-band electronic transitions

Additionally, weaker emission bands are observed near 1.81 eV, attributed to sulfur vacancy defects (VS) and commonly referred to as the "red band." The CdS_T reference film exhibits a low-energy emission at 1.45 eV, associated with intrinsic defects within the material. It also shows a more intense emission band at 1.97 eV, identified as the "yellow band," which arises from radiative recombination between donor levels and the valence band, likely induced by interstitial cadmium atoms. These results indicate that CdS films deposited via hermetically close DBQ method display optical quality comparable to or exceeding that of conventionally deposited CdS films, while featuring distinct defect related emissions that could influence their photovoltaic performance.

Photovoltaic Solar cells of CdTe

Considering the obtained results, the CdS with the best physical properties was CdS_{HB}. Therefore, serial photovoltaic devices were fabricated using CdS_T and CdS_{HB} as window material to compare their electrical properties and the photovoltaic performance. The device structure was as follows: FTO/different CdS as window material/CdTe (3µm)/Cu (5nm)/ Au (100 nm).

The CdTe absorber material was deposited by thermal evaporation technique using CdTe powder, with a thermal gradient of 100°C for 3 min on argon and oxygen inert atmosphere at 100 mtorr. Subsequently CdTe samples were thermally treated on $CdCl_2$ process with a ΔT= 150 °C for 30 min. Figure 4 shows the I-V characteristics of the photovoltaic devices, and Table 1 summarizes the electrical parameters for each device.

The photovoltaic efficiency was significantly enhanced by employing CdS bilayers deposited in a hermetically sealed CBD system (see Figure 4a) compared with the device using CdS_T as window layer increasing from 8% to 15.3 %. Figure 4b presents the quantum efficiency (QE) as function of wavelength for all devices in the 300 nm to 900 nm range, representing the ratio of generated electron-hole pairs to incident photons. A clear modification of the CdS/ CdTe junction response is observed in the wavelength region between 550 nm to 850 nm.

Moreover, devices incorporating CdS_{HB} exhibit enhanced response in the UV region and improved junction quality between CdS and CdTe, leading to increases int the open circuit voltage (Voc) from 520 mV to 792 mV and the fill factor

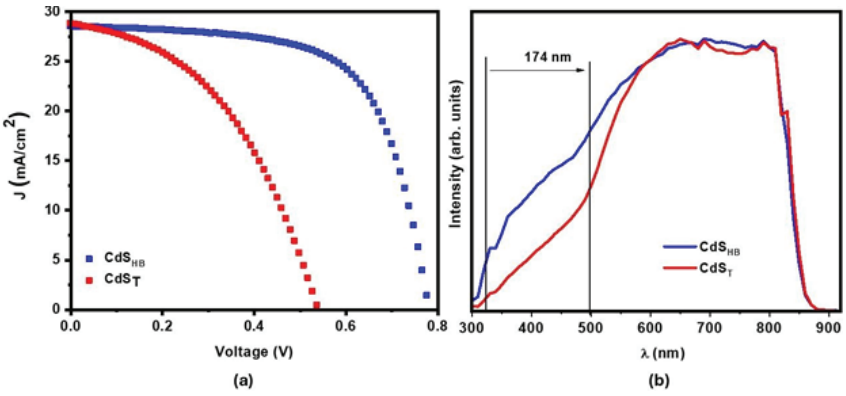

Figure 4. *CdTe solar cells (a) I-V behavior using CdS_T and CdS_{HB} as window material respectively, the photovoltaic efficiency was improved with CdS_{HB} and (b) QE vs wavelength plot for all devices,*

Table 1*. Electrical parameters for CdS/CdTe solar cells (contact area of 0.03 cm²).*

Devices	CdS Thickness (nm)	Voc (mV) ±1mV	Jsc (mA/cm²) ±3mA/cm²	R_{series} (Ω) ±0.1Ω	R_{shunt} (Ω) ±0.1Ω	FF± (%) 0.6%	η(%) ±0.8%
FTO/CdST/ CdTe/Cu/Au	~ 40	520	29.94	186	3417	47.07	8.1
FTO/CdS_{HB}/ CdTe/Cu/Au	~90	792	29.91	131	23944	64.84	15.3

(FF) from 47% to 64.84%. Additionally, both series and shunt resistance values show noticeable improvement (see table 1).

Conclusions

CdS thin films were successfully synthesized using a hermetically sealed chemical bath deposition (CBD) system, enabling internal pressure during film growth. The increasers pressure improves the crystalline quality, as evidence by XRD diffraction results. Photoluminescence analysis revealed a reduction in defect related emissions, indicating enhanced optoelectronic properties.

When employed as the window layer in CdTe solar cells, CdS films deposited using the conventional CBD method typically yielded photovoltaic efficiencies of approximately 8%. In contrast, devices fabricate with CdS layers grown under hermetically closes pressure assisted CBD achieved efficiencies of up to 15% representing a substantial improvement in device performance. Overall, this pressure assisted CBD technique represents a scalable, low cost and environmentally friendly strategy for producing high quality CdS thin films, with strong potential for next generation thin films for photovoltaic applications.

Acknowledgments

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