Fabrication of CdTe Thin Films by Close Space Sublimation

C. Potamialis, F. Lisco, J. W. Bowers and J. M. Walls
Centre for Renewable Energy Systems Technology (CREST), School of Electronic, Electrical and Systems Engineering, Loughborough University, Loughborough, Leicestershire, LE11 3TU, UK.
Corresponding author: C.Potamialis@lboro.ac.uk , Tel: +44 (0)1509 635361

Abstract

The effects of substrate and source temperatures, deposition disruption and material preparation of CdTe thin films, fabricated in a home-made CSS system are investigated. It is demonstrated that packing CdTe powder prior to deposition has a positive effect on growth rate. Furthermore, grain size, thin film thickness and uniformity strongly depend on substrate and source temperatures. Growth disruption during the sublimation process showed island formation, island size increase and island coalition in consecutive steps.

Introduction

CdTe is an II-IV group semiconductor with high optical absorption of $>10^4 cm^{-1}$, and a direct band gap of 1.45 eV, ideal for solar cell applications. These properties have made CdTe a promising candidate for production of low- cost PV with high energy conversion efficiencies. Currently, the laboratory efficiency record is 21.4% achieved by First Solar Inc [1]. CdTe absorbers can be deposited by various methods demonstrating the flexibility of this material; however, close-space sublimation (CSS) has demonstrated a process which allows uncomplicated processing apparatus, moderate operating pressures, high efficiency devices, high deposition rates and efficient material utilisation [2], Such characteristics makes CSS a very attractive deposition technique to large scale manufacturing.

Here we report the investigation on the effects of substrate and source temperatures, deposition time and material preparation of CdTe thin films, fabricated in a home-made CSS system built at CREST, Loughborough University.

The System

Deposition of CdTe thin films was carried out in a home-made CSS system illustrated at Figure 1. The system consists of a horizontal quartz tube supported by a stainless steel reactor. Source material (CdTe powder 99.999% purity) was placed on a Corning Eagle glass support for sublimation onto a substrate. A silicon carbide (SiC) coated graphite block was used as a heat susceptor to heat up the Eagle glass. The substrate was placed on a second SiC coated graphite block and kept in close proximity to the source material. Both susceptors are supported by a quartz holder and quartz spacers were used to provide separation between the source and substrate. Thermocouples were inserted inside the graphite blocks to control the temperature. Heating of the susceptors was provided by seven 1-kW IR lamps (USHIO) in a two-set configuration in an encapsulated heating element. Three lamps were used for heating the substrate graphite block and four lamps provided the heating for the source graphite block. All lamps are placed in reflectors to concentrate the light on the susceptors. Independent temperature control for the two-set configuration was provided by two PID controllers (Eurotherm) each communicating with a power controller. A gas inlet was used to introduce an appropriate amount of nitrogen and an evacuation pump (Edwards RV12) to evacuate the deposition chamber. Pressure was manually adjusted with the use of a butterfly valve, and a mass flow controller. A quartz insert is positioned immediately inside the horizontal quartz tube to protect it from material condensation. Using this configuration the system can be easily cleaned from excess material condensing on the walls of the system.

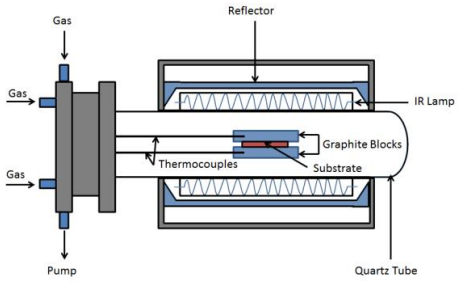

Figure 1: CSS schematic diagram

Experimental Procedures

Prior to thin film deposition the substrates (5.5 mm x 5.5 mm Eagle glass) were ultrasonically cleaned in a DI solution containing 10% IPA and 10% Acetone, for 1.5 hours at 60°C.

Source plate fabrication consists of placing CdTe powder on Eagle glass and then sublimating on a substrate prior to thin film deposition. This process is carried out to prevent spitting of CdTe micro-particles on the substrate during thin film deposition [3] Source plate optimisation was carried out by varying substrate and source temperatures, nitrogen partial pressure, substrate-source separation, deposition time and CdTe powder preparation. Deposition parameters of different source plates are summarised in Table 1 below.

Substrate Temperature Range (°C)	560 - 600
Source Temperature Range (°C)	680 - 700
Separation (mm)	2
Partial Pressure (Torr)	0.4 - 0.7
Powder Preparation	Spread or Compact

Table 1: Summary of source plate's fabrication deposition parameters

Thin film deposition was carried out using the same CSS system. The substrate and source plate were loaded in the chamber and the sublimation procedure was initiated. The chamber was pumped down to a base pressure of approximately 9 mTorr. Nitrogen was introduced in the chamber at a deposition pressure of 700 mTorr. Source and substrate were ramped to 300 °C for a 5 minute annealing to dry off any residual water present in the chamber. Then the substrate was heated to 515°C. Source heating was initiated when the substrate was at a minimum temperature of 450°C to prevent sublimation of source material during the warm up time. When the source temperature reached 630°C sublimation was initiated. For the entire length of this experimental procedure the substrate temperature was kept constant at 515°C while the source temperature and deposition time were varied in the range of 610°C to 630°C and 1 to 5 minutes respectively in order to investigate the effects on CdTe grain growth. Spacing between the source plate and substrate was kept constant at 2 mm.

Characterisation of CdTe films and CdTe Source plates were carried out using Scanning Electron Microscopy (SEM), X-Ray Diffraction (XRD) and Energy-Dispersive X-Ray Spectroscopy (EDX).

Results

Two source plates at identical deposition parameters were created to investigate the effect of CdTe powder preparation on the CSS process. In both cases, substrate and source temperatures, nitrogen pressure and separation were kept constant at 560°C, 680°C, 400 mTorr and 2 mm respectively for 30 minute deposition. Equal amounts of CdTe powder (0.8 g) were spread on Eagle glass or compacted using a soda lime glass as a presser. Results showed that void free CdTe films can be produced with an average grain size of 12 μm. However, compacting the source material prior to deposition has a positive effect on deposition rate (deposition rate increased from 1.2 μm/min to 3.2 μm/min) resulting in thicker films. EDX characterisation showed a stoichiometric composition on both source plates. Further increase of substrate and source temperatures to 600°C and 700°C respectively resulted in higher deposition rates (5.25 μm/min) and much larger grains as expected [4]. However, source plates exhibited poor uniformity with the presence of voids visible as illustrated in Figure 2.

Voids on CSS films at high temperatures (>500°C) can be formed either by re-sublimation of CdTe vapour from the substrate during the cooling cycle [5], or by interruption of process before completion of island growth into a continuous film [6]. The sublimation process was ended by flooding the chamber with N_2 and increasing the pressure to >300 Torr, while the temperature was rapidly decreased from 700°C to 550°C in 35 seconds, In this occasion it can be safely assumed that the presence of voids was related to insufficient CdTe powder, which resulted in interruption of island growth and grain coalescence. To further investigate this effect thin films were deposited from a 100 μm void free source plate.

The first set of experiments were designed to investigate the growth mechanism of CdTe films. Figure 3 shows SEM results

for CdTe films grown at three different source plate temperatures while pressure, substrate temperature and deposition time

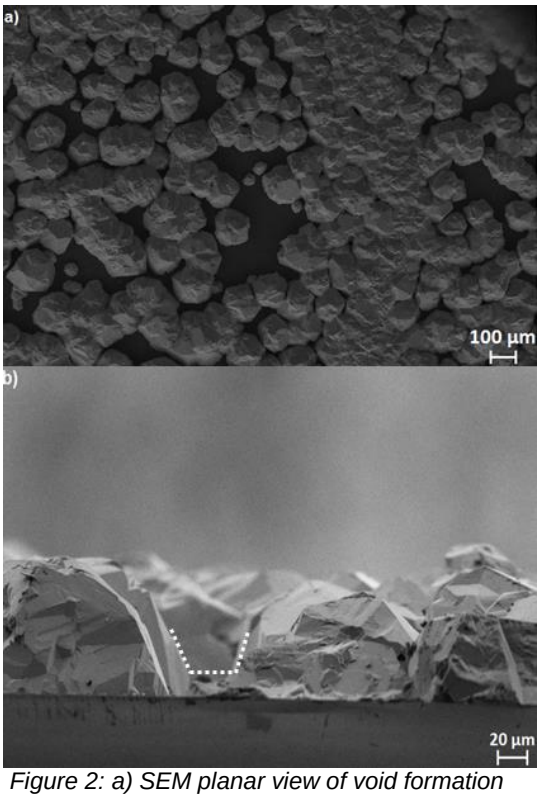

Figure 2: a) SEM planar view of void formation on the fabricated source plate and b) SEM cross section of voids (white dotted line)

remain constant (515 °C, 700 mTorr, and 5 minutes respectively).It was found that growth rate strongly depends on the source temperature. At a source temperature of 610°C, films (Figure 3.a) exhibit poor area coverage with voids between grains. Island formation and the island coalition are clearly visible. The average island size is around 35 µm. Increasing the temperature to 625°C (Figure 3.b) island size increased to an average of 41 µm while island coalition density in the film also increased, however, full substrate coverage was not realised. Further increase of source temperature to 630°C, (Figure 3.c) resulted in a 30 µm thick void free film, with an average grain size of 26 µm. XRD characterisation showed a zinc blende CdTe structure highly oriented along the (111) direction in agreement with literature[7].

The same deposition parameters which provided the 30 µm film were used to investigate the effect of deposition time on the growth mechanism of CdTe films. Figure 4 shows SEM results for CdTe film

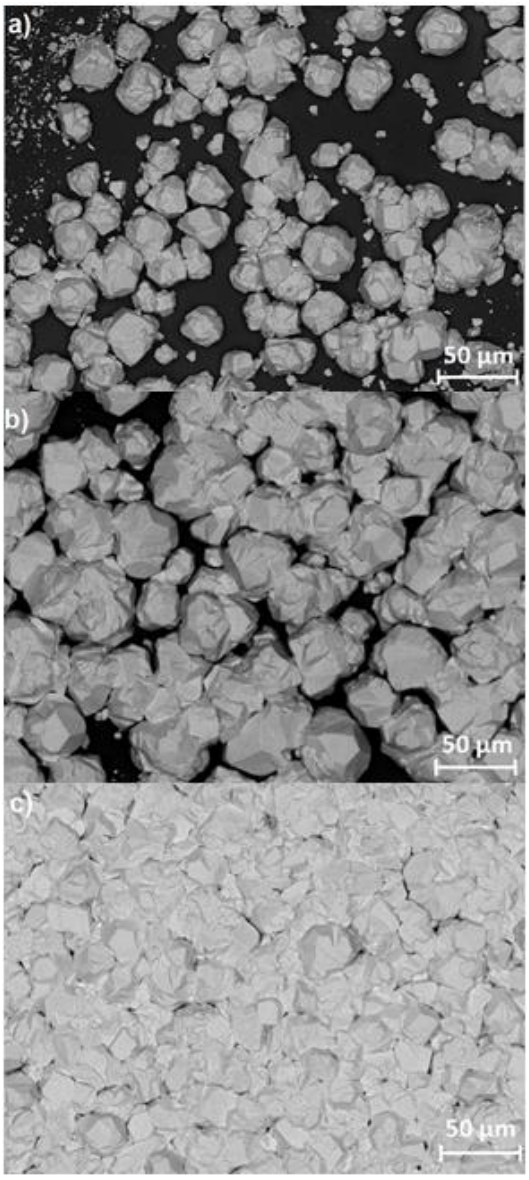

Figure 3: SEM of CdTe Films deposited at a) 610°C, b) 625°C and c) 630°C

growth at 1, 2.5 and 5 minutes of deposition. Substrate and source temperature were kept constant at 515°C and 630°C respectively. As expected after 1 minute of CdTe deposition, resulting films showed that just a few grains were deposited on the substrate. Figure 4.a illustrates this effect, average grain size was found to be around 25 µm; this can be assumed to be the stage where formation of individual islands occurs [6]. By increasing the time to 2.5 minutes thin film coverage was increased but still void governed. In figure 4.b the coalition of islands forming into a single island of larger size is visible. At this stage individual island growth has reached maximum size of around 40 µm, in this process, there is an initiation of individual

islands coalescing and forming larger grains. At the same time secondary island formation is continued in accordance to Volmer-Weber growth model [6]. Further continuation of the deposition process to 5 minutes resulted in the formation a 30 µm thick void free CdTe film (Figure 4.c).

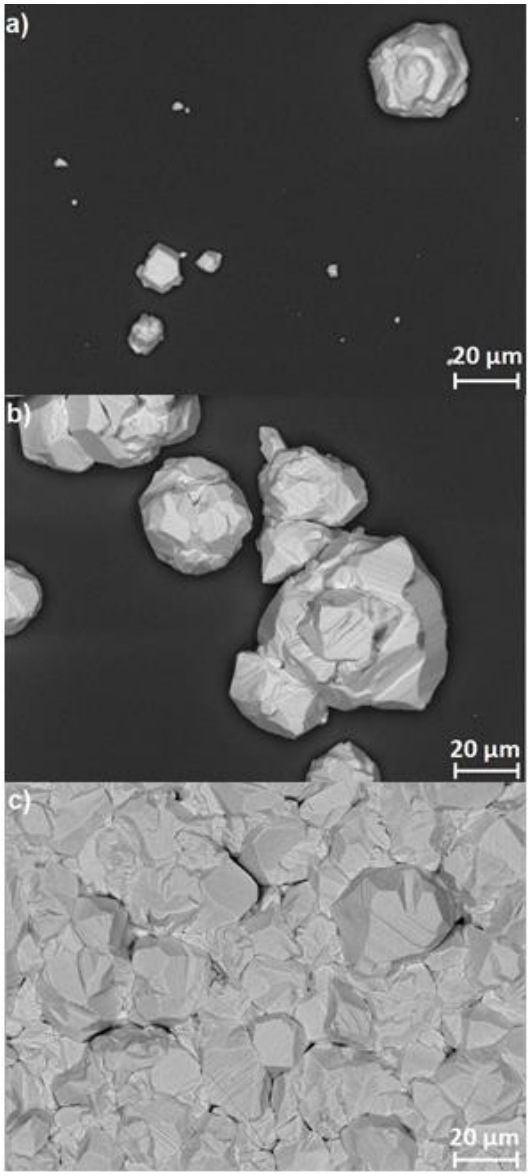

Figure 4: SEM of CdTe Films deposited at a) 1 minute, b) 2.5 minutes and c) 5 minutes

Conclusions

CdTe thin films were successfully deposited using a home-made CSS system. Source plate optimisation showed that compacting CdTe powder prior to initiation of the sublimation process resulted in higher deposition rates while grain size remained unaffected.

Furthermore, growth interruption of CdTe films during the sublimation process showed the successive steps preceding the formation of a continuous film, which include island formation, growth and grain coalition in agreement with the Volmer-Weber growth model. Further source temperature increase resulted in higher growth rates and grain size. CdTe thin films deposited on Eagle glass show a zinc blende cubic structure highly oriented along (111) direction.

References

[1] F. S. Inc, “FIRST SOLAR SETS WORLD RECORD FOR CDTE SOLAR CELL EFFICIENCY,” *August 5*, 2014. [Online]. Available: http://investor.firstsolar.com/releasedetail.cfm?ReleaseID=864426. [Accessed: 01-Jan-2001].

[2] J. Britt and C. Ferekides, “Thin-film CdS/CdTe solar cell with 15.8% efficiency,” *Appl. Phys. Lett.*, vol. 62, no. 22, pp. 2851–2852, 1993.

[3] N. Romeo, A. Bosio, R. Tedeschi, and V. Canevari, “Growth of polycrystalline CdS and CdTe thin layers for high efficiency thin film solar cells,” *Mater. Chem. Phys.*, vol. 66, no. 2, pp. 201–206, 2000.

[4] J. L. Cruz-Campa and D. Zubia, “CdTe thin film growth model under CSS conditions,” *Sol. Energy Mater. Sol. Cells*, vol. 93, no. 1, pp. 15–18, 2009.

[5] N. R. Paudel, C. R. Grice, C. Xiao, and Y. Yan, “High temperature CSS processed CdTe solar cells on commercial SnO2:F/SnO2 coated soda-lime glass substrates,” *J. Mater. Sci. Mater. Electron.*, pp. 4708–4715, 2015.

[6] J. D. Major and K. Durose, “Early stage growth mechanisms of CdTe thin films deposited by close space sublimation for solar cells,” *Sol. Energy Mater. Sol. Cells*, vol. 95, no. 12, pp. 3165–3170, 2011.

[7] J. D. Major, Y. Y. Proskuryakov, K. Durose, G. Zoppi, and I. Forbes, “Control of grain size in sublimation-grown CdTe, and the improvement in performance of devices with systematically increased grain size,” *Sol. Energy Mater. Sol. Cells*, vol. 94, no. 6, pp. 1107–1112, 2010.