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Preparation and characterization of transparent conducting ZnTe:Cu back contact interface layer for CdS/CdTe solar cell for photoelectrochemical application

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FSEC Photovoltaic Materials Laboratory has developed a photoelectrochemical (PEC) cell using multiple band gap tandem of thin-film photovoltaic (PV) cells and a photocatalyst for hydrogen production by water splitting. CdS/CdTe solar cell, a promising candidate for low-cost, thin-film PV cell, is used as one of the thin-film solar cells in a PEC cell. The back contact has been developed for a CdS/CdTe solar cell which involves the deposition of a primary *p*-type ZnTe:Cu back contact interface layer followed by the deposition of transparent and conducting ZnO:Al and Ni–Al outer metallization layer. This article presents preparation and characterization of ZnTe:Cu back contact interface layer deposited by hot wall vacuum evaporation (HWVE) technique. HWVE technique has produced highly stoichiometric ZnTe:Cu thin films with cubic phase having {111} texture orientation and produced better transparency in the near infrared region on a glass substrate. © 2006 American Vacuum Society. [DOI: 10.1116/1.2201053]

I. INTRODUCTION

As a part of the NASA Hydrogen Project for Florida universities, the FSEC Photovoltaic Materials Laboratory has developed a photoelectrochemical (PEC) cell using a multiple band gap combination of a thin-film photovoltaic cell and photocatalyst. A PEC cell is used to produce hydrogen by splitting water into hydrogen and oxygen using the electric current generated by solar cells. A PEC setup is shown in Fig. 1. It consists of two illuminated solar cells (CdTe and CIGS2), a photoanode deposited on titanium sheet for oxygen evolution, and a platinum foil cathode for hydrogen evolution. The effort presented in this research is different and distinct from the development of two-junction thin-film photovoltaic (PV) tandem cells typically with individual absorber band gaps of ~ 1.0 and 1.7 eV that have to be grown one on top of the other. Instead, the PEC cell uses a multiple band gap tandem of thin-film solar cells and a photoanode. The band gap of thin-film solar cells (CdTe and CIGS2) is in the range of ~ 1.5 – 1.7 eV. Therefore, photons of energy > 1.5 – 1.7 eV are absorbed by the PV cell but photons below this range remain unabsorbed. They can be utilized by using a photoanode of band gap of 1.1 eV, thus opening the possibility of an efficient multiple band gap combination of thin-film photovoltaic cell and thin-film photocatalyst for production of hydrogen and oxygen by water splitting.¹

Polycrystalline thin-film cadmium telluride (CdTe) solar cells, one of the PV cells used in this PEC setup, are amenable to economic mass production. CdTe solar cell has also shown laboratory efficiencies of 16.5% and module performance of over 10%.² In addition, CdTe solar cells with effi-

ciencies over 11% have been made by a variety of deposition methods, and several of these methods have been adapted to module manufacturing. In a PEC cell, in order to transmit photons below the band gap (< 1.5 eV) of CdS/CdTe solar cell, a transparent and conducting back contact has to be employed to PV cell for multiple band gap tandem of PV cell and photocatalyst.

One of the major issues in fabricating a high efficiency CdS/CdTe device is to form a stable and low-resistance back contact for CdTe. In materials such as *p*-CdTe, Ohmic metal-semiconductor contacts do not form readily.³ Because of the high ionization energy of CdTe and the difficulty of achieving high doping levels in polycrystalline *p*-type CdTe, a low-resistance Ohmic contact cannot be formed by simple metallization. ZnTe, which is expected to have a small valence band discontinuity with CdTe and can be doped degenerately with Cu, has been used as a back contact interface layer to develop transparent and conducting back contact for superstrate-type CdS/CdTe thin-film solar cell.

Zinc telluride films have been fabricated by a variety of techniques such as thermal evaporation of ZnTe and Cu from two sources,⁴ rf sputtering,⁵ thermal evaporation,⁶ and electrodeposition.⁷ The economics of PV manufacturing requires deposition techniques designed for much large areas such as sputtering or vapor transport. In the present study, hot wall vacuum evaporation (HWVE) technique has been developed for deposition of ZnTe:Cu layer. This technique has obtained high-quality luminescent and electro-optic films of II–VI materials with better carrier concentration and mobilities. It can facilitate growth of thin films under conditions close to thermodynamic equilibrium with a minimum loss of material and has also obtained highly stoichiometric compound thin films.⁸

In this article we present the preparation and characterization of *p*-type ZnTe:Cu transparent conducting layer by

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FIG. 1. PEC setup consisting of two thin film PV cells and a photocatalyst.

HWVE technique for CdS/CdTe superstrate-type of thin-film PV cells for photoelectrochemical applications.

II. EXPERIMENT

ZnTe:Cu layer was deposited using HWVE technique. A schematic diagram of the hot wall setup developed at the FSEC Photovoltaic Materials Laboratory is shown in Fig. 2. The principal advantages of hot wall technique are economy of material, better stoichiometry through the maintenance of adequate evaporant partial pressure, and better uniformity of film thickness.⁹ The hot wall was constructed using fused quartz glass tube. Kanthal heating element ribbon was used to heat quartz glass tube. The glass tube was surrounded with a pair of stainless steel metal shields to prevent heat from

FIG. 2. Schematic diagram of hot wall vacuum evaporation setup.

reaching towards the vacuum chamber wall. Type K, chromel-alumel thermocouples were used to measure temperatures of hot wall and substrate. The distance between source and substrate was maintained at 15 cm. ZnTe:Cu layer was prepared by doping ZnTe with Cu and with Cu₂Te to compare the optical properties of ZnTe:Cu layer. A molybdenum boat was used for evaporation of ZnTe grains (purity 99.999%) and a tungsten boat was used for evaporation of Cu powder (purity 99.999%) and Cu₂Te powder (purity 99.9%). Depositions were carried out on 10 × 10 cm² size glass substrates. The thickness of ZnTe:Cu layer was optimized at 5000 Å with the following deposition parameters: hot wall temperature of 350 °C, substrate temperature of 300 °C, deposition time of 7 min, copper concentration of 2 at. %, and annealing after deposition at 300 °C for 30 min. Experiments were also carried out to deposit ZnTe films without a hot wall with the same deposition parameters to compare the material yield, thickness uniformity, and stoichiometry of ZnTe films obtained with a hot wall.

CdS/CdTe solar cells were completed with the following configuration: glass/SnO₂:F/CdS/CdTe/ZnTe:Cu/ZnO:Al/Ni–Al. CdS/CdTe samples were procured from a solar cell manufacturer with the configuration glass/SnO₂:F/CdS/CdTe. Here soda lime glass substrate coated with transparent and conducting SnO₂:F layer is used to form a front contact. CdS heterojunction partner layer (*n* type) and CdTe absorber layers (*p* type) are coated by vapor transport deposition process. This is followed by CdCl₂ treatment. ZnTe:Cu back contact interface layer was deposited using HWVE technique using the same deposition parameters optimized on a glass substrate. CdS/CdTe solar cells were completed by deposition of ZnO:Al layer using rf magnetron sputtering followed by deposition of Ni–Al contact fingers by e-beam evaporation technique.

ZnTe:Cu thin films on glass were characterized with electron probe microanalysis (EPMA) for composition, x-ray diffraction (XRD) for structural properties, and optical transmission spectroscopy for optical transmittance. Dektak profilometer was used to measure thicknesses of thin films. CdS/CdTe solar cells were characterized for composition-depth profiling by secondary ion mass spectroscopy (SIMS).

III. RESULTS AND DISCUSSION

The transparent and conducting back contact developed on CdS/CdTe solar cells for PEC multiple band gap tandem cell must be transparent in the infrared region (~750–1150 nm). ZnTe:Cu thin film developed for CdS/CdTe solar cells was developed on glass initially, observed for its visual appearance, and characterized for optical transparency. The films deposited at 300 °C were visibly reddish in color and showed mirrorlike shining appearance while films deposited at room temperature were dark red in color and did not show mirrorlike shining appearance. The films deposited at 300 °C and annealed for 30 min showed the same visual appearance as films deposited at 300 °C without any annealing treatment. Observation under a high resolution optical microscope showed that the films were ex-

FIG. 3. Optical transmittance data for ZnTe:Cu and ZnTe:Cu₂Te films deposited on glass.

tremely smooth. This was observed for all the films and was found to be independent of film thickness. The results presented below are for a typical representative sample.

Gessert *et al.*¹⁰ have produced CdS/CdTe devices with an efficiency of 11%–12% employing ZnTe:Cu back contact layer with an optimum thickness of 5000 Å. Experiments were carried out by setting the hot wall temperature at 350 °C, substrate temperature at 300 °C, and Cu concentration of 2 at. % to optimize the thickness of ZnTe:Cu layer at 5000 Å. Films were annealed at 300 °C for a duration of 30 min as postdeposition annealing reduces resistivity of the films. ZnTe:Cu films deposited at 300 °C have shown better carrier concentration, mobility, and conductivity.³ The transparency spectra of ZnTe doped with Cu and Cu₂Te are shown in Fig. 3. It showed sharp increase in transmittance at the wavelength of 548 nm for ZnTe doped with Cu, equivalent to a band gap of 2.26 eV. Transparency of the film increased as wavelength increased from 548 to 900 nm. Transparency in the range of 548–700 nm was ~35%–50%. However, for a multiple band gap tandem PEC cell, transparency above 820 nm has to be taken into consideration as band gap of CdTe is ~1.5 eV. Therefore most of the photons having a wavelength of up to 820 nm would be absorbed by the CdTe absorber layer. Transparency obtained for the ZnTe:Cu film was ~55%–65% in the range of 700–900 nm. Transparency obtained for ZnTe doped with Cu₂Te was ~30%–55% in the range of 548–700 nm and ~55%–75% in the range of 700–900 nm. ZnTe doped with Cu₂Te showed better transparency as compared to ZnTe doped with Cu and hence was used for final deposition for obtaining a CdS/CdTe cell.

The economics of PV manufacturing requires deposition techniques designed for large areas and low cost. HWVE technique used in the present research is a relatively simple technique with low cost and has produced films of quality acceptable for optoelectronic device application. Experiments were carried out to compare material yield, thickness uniformity, and composition of ZnTe thin films obtained with hot wall to those without hot wall. Results obtained are shown in Table I. To obtain ZnTe thin films with 5000 Å thickness, samples with hot wall required 0.26 g of ZnTe and

TABLE I. Comparison of ZnTe grown with and without hot wall.

Properties (substrate size of 4 × 4 in. ² , thickness of 5000 Å)	With hot wall	Without hot wall
Material required (g)	0.26	0.32
Thickness uniformity (%)	±4.5	±9.8
Average stoichiometry (at. %)	Zn=50.2, Te=49.8	Zn=53.0, Te=47.0

samples without hot wall required 0.32 g of ZnTe. Thickness uniformity was ±4.5% for thin films obtained with a hot wall while it was ±9.8% without a hot wall. Material yield and thickness uniformity increase may be due to conditions during the growth, provided by the hot wall. As the distance between source and substrate is enclosed by a hot wall, the Zn and Te atoms which tend to go away and also reevaporate and bounce back from the substrate are sent back by the hot walls which are kept at elevated temperatures, increasing material yield and thickness uniformity in the film. The stoichiometry of ZnTe films prepared with and without a hot wall was also compared as it plays an important role in forming highly *p*-type ZnTe:Cu thin films. Average atomic concentrations measured at 20 kV for samples with a hot wall showed a Zn:Te proportion of 50.2:49.8 while it was 53.0:47.0 without a hot wall. EPMA results were also obtained for ZnTe:Cu film deposited with hot wall technique. Average atomic concentrations measured at 20 kV showed Zn:Te:Cu proportion of 46.2:51.2:2.6. This proportion indicated that zinc atoms were replaced by copper atoms in the ZnTe layer, thus increasing the possibility of obtaining a highly *p*-type ZnTe:Cu layer.

The structural properties of ZnTe:Cu thin films were characterized by x-ray diffraction (XRD) technique and are discussed elsewhere.¹¹ The XRD pattern showed a highly textured thin film with the reflections of (111) at $2\theta=25.90$, (222) at $2\theta=52.42$, and (333) at $2\theta=81.96$, all from the (111) set of planes. The XRD pattern confirmed the cubic structure of ZnTe:Cu with strong {111} orientation. Further, the nature of XRD plots remained the same for film samples which were subjected to heat treatment at 300 °C for 30 min and those which were not subjected to any heat treatment. This behavior could be attributed to the conditions during growth, provided by the hot wall, favoring the growth of crystallites and their orientation. The impact of Zn and Te particles on the walls of the cylinder, enclosing the sources, provided necessary additional thermal energy till they reached the substrate held at a higher temperature. This helped to achieve higher migrational energy of the adatoms resulting in better nucleation. Also the accommodation coefficient, which is governed by energies corresponding to the source, the substrate, and the particle temperatures, could be expected to be higher. This could have resulted in an increased smoothness of the films.

CdS/CdTe solar cells were completed with the following configuration: glass/SnO₂:F/CdS/CdTe/ZnTe:Cu/ZnO:Al/Ni–Al. Currently one of the major issues facing

FIG. 4. SIMS depth profile of CdS/CdTe device.

CdS/CdTe solar cell devices is long term instability of the device due to Cu diffusion from the back contact layer. Composition-depth profiling of Cu in a CdS/CdTe device was carried out by SIMS technique to observe any diffusion of Cu into the device from the ZnTe:Cu back contact interface layer during HWVE processing. Figure 4 shows the SIMS profile of a CdS/CdTe device without Ni–Al contact fingers. The SIMS profile suggests that Cu has diffused into the device through ZnTe:Cu back contact interface layer. Cu concentration is nearer the CdTe/ZnTe interface and it reduces further in the CdTe region but again increases in the CdS layer which could be detrimental for the long term stability of the device. Diffusion of Cu may be attributed to incorporation of Cu during CdCl₂ treatment prior to HWVE processing.¹² Also, diffusion of Cu may be due to deposition of ZnTe:Cu at elevated substrate temperature and postdeposition annealing of the film. Presently efforts are underway to carry out analysis by depositing ZnTe:Cu layer at different substrate temperatures and to reduce the step of postdeposition annealing. Analyses through these experiments may help in reducing Cu diffusion into the device and thus aid in increasing long term stability of the device.

IV. CONCLUSION

HWVE proved to be a successful technique for obtaining highly stoichiometric and textured ZnTe:Cu thin films with better thickness uniformity and material yield. ZnTe:Cu layer obtained by doping ZnTe with Cu₂Te showed more (~65%–75%) transparency as compared to ZnTe doped with Cu (55%–65%) on glass in the near-IR region required for PEC applications. ZnTe:Cu layer was deposited successfully on superstrate-type CdS/CdTe thin-film PV cells and was analyzed for Cu diffusion into CdS/CdTe device by SIMS technique. Initial study indicates that there is Cu diffusion from ZnTe:Cu back contact interface layer into *p*-type CdTe and *n*-type CdS layer and further work will be required to carry out for reducing the Cu diffusion into the device during HWVE processing to enhance the long term stability of the CdS/CdTe device.

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