Synopsis of the Thesis Entitled

Development and Fabrication of Laser Scribed Copper Indium Gallium Diselenide (CIGS) Thin Film Solar Cell Module

To be submitted to

The Maharaja Sayajirao University of Baroda

For the Degree of

Doctor of Philosophy

In

Applied Physics

By

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January 2020

Development and Fabrication of Laser Scribed Copper Indium Gallium Diselenide (CIGS) Thin Film Solar Cell Module

An objective of this thesis research work is to develop and fabricate Copper Indium Gallium Diselenide (CIGS) thin film solar cell module on a glass substrate using laser scribed monolithic interconnection. Stack of multilayer consists of Glass substrate/Molybdenum/Copper Indium Gallium Diselenide (CIGS)/Cadmium Sulphide (CdS) /intrinsic-Zinc Oxide (i-ZnO)/Aluminum doped Zinc Oxide (Al-ZnO) have been deposited using various methods like Direct Current (DC) sputtering, Radio Frequency (RF) sputtering, Thermal evaporation and Chemical Bath deposition techniques. To fabricate CIGS solar module, laser scribing process has been adopted to create monolithic interconnection between top window layer and ohmic back contact. Finally, physical properties, optical properties, electrical properties and performance of each layer as well as device were estimated.

An exponential increase in demand of energy worldwide asked researchers to explore multiple innovative technologies to fulfill the deficit between demand and supply of energy. This demand and supply are fulfilled by renewable and non-renewable energy resources. Compared to renewable resources, non-renewable energy resources have demerits such as they will be extinct soon, cost and efficiency of technology, huge menace to environment which can cause ecological hazard, increment in carbon dioxide (CO_2) emission and highly unsustainable. Natural oil, coal, natural gas, petroleum, fossil fuels are non-renewable energy resources that will not replenish from their sources [1, 2]. According to present consumption of energy resources due to innovation and advancement in technology its demand is escalated at very high rate and it will keep constantly rising in future also. To reduce the gap between supply and demand of energy resources, an alternative option is renewable energy resources. The renewable energy resources generate energy from solar power, wind power, wave and tidal power, geo-thermal energy and hydro-power which have high sustainability, less CO_2 emission and less hazards to ecological environment. Among all renewable resources, solar power generated from sun is most preferable choice due to its availability all over the world with no cost. To use solar energy in day to day life, sun's light energy is converted into electric energy with the help of materials responding to photoelectric effect [3]. In year 1839, a French scientist Antonie Cesar Becquerel developed solar technology. The first solar cell was made up of amorphous silicon developed by Chaplin, Fullere and Pearson in year 1954. Continuous increment in demand of low cost, multivariant applicable hazard free and highly efficient solar cell provoked researchers to explore new emerging technologies for developing solar cell. Initially only silicon technology has monopoly over solar energy research and development as well as in electrification development [4]. Therefore, silicon technology is said to be first generation of solar photovoltaic cell. Czochralski process float zone, Bridgman technique, Siemens process and Pyrolysis process were used to develop Mono-crystalline and Poly-crystalline Silicon solar cell [5, 6]. These processes give high conversion efficiency, but having demerits such as wastage of Si wafers, high probability defect formation in solar cell and high processing cost. Mono-crystalline and Poly-crystalline silicon solar cell has achieved 26.7±0.5 % and 22.3±0.4 % solar conversion efficiency developed by Kaneka corporation and FhG-ISE respectively [7]. To overcome the demerits of first-generation solar cell, new innovative idea of reducing the thickness of active or absorber layer has boomed out. Cadmium Telluride (CdTe), Copper Indium Gallium Diselenide (CIGS), Amorphous silicon (a-Si) and Gallium Arsenide (GaAs) have proven themselves as prominent choices of absorber layer to develop solar photovoltaic cells. These solar cells are known as second generation solar photovoltaic cells [8]. Their thickness ranges from 0.1 μm to less than or equal to 1 μm. Thermal evaporation, RF sputtering, DC sputtering, Co-evaporation technique, Metal Organic Chemical Vapor Deposition (MOCVD), Molecular Beam Epitaxy (MBE), etc. are the deposition techniques used in developing second generation solar photovoltaic cell [9-12]. A remarkable world record has been noted in ALTA device on account of GaAs thin film solar cell 29.1±0.6 % efficiency. Solar Frontier has marked 22.9±0.5 % efficiency on CIGS solar cells of 1 cm^2 area. Single Junction terrestrial CdTe solar cell on glass substrate developed by First Solar has achieved efficiency of 21.0 %. Researchers at National Institute of Advanced Industrial Science and Technology, Tokyo, Japan has developed a-Si and micro-crystalline cells having solar conversion efficiency about 10.2±0.3 % and 11.9±0.3 % respectively [13-16]. Miasole has developed CIGS solar panel of 1 m^2 with 15.7±0.5 % solar conversion efficiency. Disadvantage of second-generation solar cell for industrial production is unavailability of less abundant elements like Te, Cd, Ga, In etc. Also, elements like Te and Cd are highly toxic and hazardous to human health [17]. Till date, mass production of second-generation solar cell is not affordable and adaptable in terms of high cost of material, manufacturing and installation solar panels. To reduce toxicity of chemical elements, Third Generation solar cell has come up with Dye-sensitized solar cells (DSSC), Organic/Polymer Photovoltaic cells and Copper Zinc Tin Sulphide (CZTS) solar cells [18, 19]. One of the major advantages of third generation solar cell is having low manufacturing cost. Third generation solar cells are manufactured by roll to roll printing, screen printing and other vacuum and non-vacuum techniques [20, 21]. Sharp Corporation has fabricated DSSC with single solar cell, mini module of 7 serial cells and sub module of 26 serial cell with efficiency of 11.9±0.4 %, 10.7±0.4 % and 8.8±0.3 %, respectively. This organic dyes which can generate electricity through illumination in electrochemical cells [22]. Researchers at University of South Wales, Australia, have reported that heterojunction heat treatment process of CZTS solar cell improves the performance due to inter-diffusion of elements and lead to solar efficiency of 11 % for small area cells (0.23 cm^2) and 10 % for a standard sized cell (1.11 cm^2) [23]. Toshiba has developed organic solar cell and minimodule of series of 8 cells with solar efficiency of 11.2±0.3 % and 9.7±0.3 %, respectively [24]. To enhance the performance, stability and efficiency of solar cell multiple junction solar cells with broad spectral absorber wavelength have been developed. Multijunction solar cells such as GaInP/GaAs/Si monolithic (22.3±0.8 %), Perovskite/Si monolithic (27.3±0.8 %), InGaP/GaAs/InGaAs (31.2±1.2 %) are about to reach Shockley-Quiesser limit (33 %) [22, 25].

Among second generation solar cell, thin film technology played prominent role in solar industries. Different vacuum and non-vacuum technologies have been incorporated for improvement of solar cell efficiency. Thin film photovoltaic (TFPV) solar devices based on Copper Indium Gallium Diselenide (CIGS) provide an opportunity to decrease the material utilization. Industry have adopted techniques such as thermal evaporation, flash evaporation, RF and DC sputtering, co-evaporation, sequential evaporation and selenization, closed space vapor transport, spray pyrolysis [26-28]. CIGS based solar cells have good chemical stability and doping versality [29].

In this research work, the CIGS solar cell module was developed in the form of multilayer stack deposited on glass substrate consisting of Mo/CIGS/CdS/i-ZnO/Al-ZnO/grid. The multilayer was formed by using various deposition techniques like DC sputtering, RF sputtering, Chemical Bath Deposition and Thermal Evaporation. Each layer has its own key role in performance of CIGS solar cell module. Significance of each layer and its deposition technique is described below:

- Soda Lime Glass Substrate
  - A substrate at the bottom of solar cell module
  - The dimension of substrate is 2 in^2 area and 1 mm thickness
- Molybdenum (Mo)
  - An ohmic back-contact layer
  - To collect photo-generated carriers
  - Low resistive, high adhesion, low recombination and inertness to corrosive atmosphere
  - Direct current sputtering technique is used for deposition
- Copper Indium Gallium Diselenide (CIGS)
  - An absorber layer
  - To absorb photons and generation of charge carriers
  - It has suitable band gap to absorb solar spectrum, proper doping and enough diffusion length
  - CIGS has absorption coefficient 10^5 cm^{-1} and good chemical stability
  - RF sputtered CIGS thin film was deposited using in - house fabricated CIGS sputter target
- Cadmium Sulphide (CdS)
  - A buffer layer
  - To provide good junction formation, protection against damage of absorber layer due to sputtering deposition of window layer, reduce interface recombination
  - It has optimum thickness, low electrical resistance, minimum optical absorption loss
  - Thermal evaporation and Chemical Bath Deposition (CBD) techniques were used to develop CdS thin film
- intrinsic- Zinc Oxide (i-ZnO)/Aluminum doped Zinc Oxide (AZO)
  - A Bi-layer used as top window layer
  - To collect photons and to drive out photogenerated carriers
  - i-ZnO is an insulator layer while AZO is transparent conductive oxide layer having wide direct band gap high optical transmittance and conductivity

- Both i-ZnO and AZO thin films were deposited using RF sputtering techniques
- For deposition of i-ZnO thin films in-house 2-inch (diameter) sputter target was prepared and for AZO thin films AZO sputter target (American Elements) (2 % Al_2O_3) was used

A state of art for developing highly efficient CIGS solar cell module with low electrical and optical loss is monolithic integration process. This process helps to reduce intrinsic losses due to dead areas and series resistances. It includes uniform separation of cells and provide interconnection between front contact and back contact. There are different types of patterning to create separation of cell and due to which permutation and combinations of series and parallel of cells in module will produce desired output power. At three levels, patterning is created; first P1 patterning on Mo layer to create electrical isolation, P2 on CdS layer to create series interconnection between adjacent cell and finally P3 above top window contact to create cell isolation with adjoining cell [31, 32]. Patterning is a scribing process which is done either by mechanical scribing or laser scribing technologies. In mechanical scribing process a sharp pointed tool is used to scribe, but there are certain disadvantages in using this process such as cost of tip, replacement of tip, slow process and interposing of slinters at the edges as well as in scribe width area. This reduce active area and therefore performance of module is deteriorated. To overcome this, Laser scribing process has been introduced. Laser scribing is a selective laser ablation process in which material is removed due to heat energy supplied by laser pulse. Depending upon wavelength and pulse duration, different lasers can be used to create monolithic interconnection [33, 34]. With the help of laser fine, narrow, clean, burr free and reduced dead area patterning can be carried out. Laser scribing is cost effective technology because it is low maintenance process unlike mechanical scribing. In this research work, laser system with a source of 1064 nm Nd: YAG Pulsed Fibre Laser (IPG Photonics) from M/s. Sahajanand Laser Technology Limited, Gujarat, India, have been used [35]. Average output power is 50 W with beam quality (M^2) 2.0 and pulse duration 120 ns at 50 kHz. Different layers have different physical, optical and mechanical properties, depending on these, laser parameters such as laser power, pulse repetition rate, speed, pass delay and Z - focus was optimized to achieve clean, narrow and burr free scribe.

Chapter 1:

Remarkable achievement in development of photovoltaic cell technology is presented in the chapter. Current scenario of different photovoltaic devices and road map towards high efficiency is well described. An outline of role of material properties of Molybdenum, Copper Indium Gallium Diselenide, Cadmium Sulphide, intrinsic- Zinc Oxide and Aluminum doped Zinc Oxide are well described. Importance and key feature of glass substrate is also explained in this chapter. Merits and demerits of different types of interconnection of solar cell is also summarized.

Chapter 2:

The chapter deals with variety of deposition methods and characterization techniques used for development and fabrication of CIGS solar cell module. Thickness, morphology, elemental composition, structural, optical and electrical properties of different layers as well as device was characterized. The techniques used for characterization are X-ray diffraction, Atomic Force Microscopy, Scanning Electron Microscopy, Electron Dispersive X-ray Analysis, Transmission Electron Microscopy, Photoluminescence and Raman Spectroscopy. The van der Pauw method and Four Point Probe method are used for electrical, Current-Voltage (I-V) characteristics, Capacitance-Voltage (C-V) characteristics were analyzed for detail assessment of optoelectronic properties of each layer and CIGS solar module device.

Chapter 3:

The chapter reports the deposition method and optimization of Mo layer used in the CIGS solar module. An ohmic back contact for CIGS solar cell module was developed using refractory material i.e. Molybdenum (Mo) deposited by DC sputtering. Single as well as bilayer Mo thin film was deposited at constant argon flow. For single layer deposition pressure was varied as 10 mTorr and 1 mTorr while for bilayer, bottom layer at 10 mTorr and top layer 1 mTorr. Each layer has 0.5 μm thickness. DC sputter power varied as (50, 100, 150 and 200 W) and two different substrate temperature (Room temperature and ST-215 ˚C) was kept constant. Morphological, structural, optical and electrical studies were carried out. Adhesion was tested using mechanical and laser technology. For monolithic interconnection P_1 process was optimized using Nd: YAG 1064 nm laser by varying parameter viz. pulse repetition frequency (PRF), power, speed, and pass delay. All above mentioned deposition parameters were tuned to obtain high reflectivity, low resistive and highly adhesive back ohmic contact with burr free clean electrical isolation.

Chapter 4:

The chapter describes the synthesis, deposition method and characterization of CIGS thin film as an absorber layer. Single quaternary chalcopyrite Copper Indium Gallium Diselenide (CIGS) sputter target was fabricated in-house using high-energy ball milling technique followed by vacuum sintering process. Using the prepared CIGS sputter target, thin films were deposited by RF sputtering technique on 2 sq-inch glass substrate. Influence of RF power, deposition pressure and substrate temperature on optical and electrical properties of CIGS thin films were studied. Also, annealing of CIGS thin films were carried out to study heat treatment effect on grain size and etching out of binary phase. Composition, Structure, optical property and electrical property were evaluated to optimize CIGS thin film as an absorber layer.

Chapter 5:

The chapter reports the deposition methods and characterization of Cadmium Sulphide (CdS) buffer layer. Here, two different techniques i.e. (1) Thermal evaporation and (2) Chemical Bath Deposition (CBD) was used to deposit CdS buffer layer. Both deposition techniques were used to deposit CdS thin film on 2 sq-inch glass substrate. For thermal evaporation deposition CdS powder was used and substrate temperature was varied (RT, 100, 125, 150 and 175 ˚C). In CBD deposition method the study was carried out to know the effect of Sulphur to Cadmium ratio which was varied as 1, 2.5, 5 and 7. Characterizations such as structural, optical, Urbach energy, light and dark resistivity and photoluminescence analysis were carried out in order to optimize CdS buffer layer used for development of CIGS solar module.

Chapter 6:

The chapter discusses the influence of deposition parameters viz. deposition pressure, RF power, substrate temperature and thickness on structural, optical, electrical, topographical and microstructural analysis of AZO thin films and i-ZnO thin films. The structure symmetry and presence of defects, impurities in presence of variable deposition parameters were also analyzed using Raman and photoluminescence spectroscopic analysis. Both i-ZnO and AZO thin films were sputtered using RF sputtering technique on 2 sq-inch glass substrate. For i-ZnO thin film in-house 2-inch (diameter) sputter target was fabricated using ZnO powder (99.9 %) while for AZO thin films AZO sputter target (American Elements) (2 % Al_2O_3) was used.

Chapter 7:

The chapter describes fabrication of CIGS solar cell module on 2 sq-inch glass substrate. For developing CIGS solar stack, optimized ohmic back contact (Mo), absorber layer (CIGS), buffer layer (CdS), top window layer (i-ZnO and AZO) was deposited followed by intermediate laser scribing process for electrical isolation of each layer of solar cell module. I-V and C-V (open circuit voltage V_{oc}, short circuit current I_{sc}, fill factor FF and quantum efficiency η %) characteristics of CIGS thin film solar cell module was analyzed to estimate the performance.

Conclusion and Future Scopes:

This chapter is an accomplishment of the work done in the thesis. Along with that, we also discuss the future prospects of research in the area of monolithic integration of CIGS thin film solar cell for multijunction structure.

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Endorsement of the Supervisor:

Synopsis is approved by me.

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List of Publication

Papers in Peer Reviewed Journals

1. Priya S. Suryavanshi, C. J. Panchal, "**Investigation of Urbach Energy of CdS Thin Films as Buffer Layer for CIGS Thin Film Solar Cell", *Journal of Nano - And Electronic Physics,* 10** (2), 02012(5pp) (2018).

2. Priya S. Suryavanshi, C. J. Panchal, **"Urbach Energy, Burstein Moss Shift and Physical Properties of RF Sputtered i-ZnO and AZO Thin Film for CIGS Solar Cells", *Solar Energy* (Communicated) (2019).**

3. Priya S. Suryavanshi, C. J. Panchal, **"Nanosecond Laser Patterning and Impact of Deposition Parameter on DC sputtered Bilayer Molybdenum Thin films", *(Manuscript under preparation*)**

Papers in Conference proceedings

1. Priya S. Suryavanshi, Hardik Khunt, Bharati Rehani, C. J. Panchal, **"Texture Analysis of Flash Evaporated CIGS Thin Films for Photovoltaic Devices", *Materials Today: Proceedings,* 4 12500–12504 (2017).**

2. Priya S. Suryavanshi, C. J. Panchal, **"Influence of Thickness on Physical and Optoelectronic Properties of DC Sputtered Bilayer Molybdenum Thin Films for CIGS Solar Cell", *Materials Today: Proceedings,* (Communicated) (2019).**