

Thickness dependent studies of hetero-junction solar cell synthesized on quartz substrate by spray pyrolysis technique

R Shashidhar* & Nityanand Choudhary

Department of Physics, J S S Academy of Technical Education, Bengaluru 560 060, India

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Variable thickness hetero-junction solar cells synthesized on quartz substrate by spray pyrolysis technique synthesized quartz based solar cells have been found to possess cell parameters such as open circuit voltage, saturation current density, fill factor and efficiency in the range of 191 – 449 mV, 2×10^{-9} – 0.11×10^{-6} A cm⁻², 15 – 20 % and 0.2×10^{-9} – 3.35×10^{-6} % at 206 mW/cm² (at air mass 5.6), respectively. Series and shunt resistance of the solar cells have been found to vary with the thickness of semiconductor films. Thick film solar cells are found to possess reasonably good cell parameters compared to thin film solar cells due to enhancement of charge concentration, mobility, grain size, variation of optical band gap, excesses atomic percentage of Ti, Cu, Te, Sn elements in TiO₂, CuO, CdTe, SnS films and less structural defects, increment of activation energy as well as existence of mixed phase at the Schottky barrier. The optical response of hetero-junction solar cells is found to be thickness dependent. Good rectifying characteristic has been reported in the present work for both thick and thin hetero-junction solar cells. All the measurements have been performed in air without protection against oxygen gas or moisture, which shows the stability of spray pyrolyzed thin films.

Keywords: Spray pyrolysis, Semiconducting films, Film thickness, Quartz, Hetero-junction solar cells

1 Introduction

Photovoltaic energy is future for harnessing renewal energy when conventional energy source is fast exhausting. With rising pollution worldwide due to vast industrialization, affordable and eco-friendly energy sources are the need of the day to save the existence of humanity. Energy generated by photovoltaic conversion is eco-friendly and affordable. Photovoltaic cells are synthesized using suitable semiconductors which are the most promising class of materials for converting solar energy directly into electrical energy¹. Wide ranges of semiconductors have been explored for their prospective utilization in photovoltaic applications²⁻⁵. The common yardstick for selection of a semiconductor material is their availability, toxicity, efficiency, cost effectiveness and the environmentally friendly. The selection of materials is carefully done depending upon the availability of materials, processing techniques and photovoltaic properties that can be attained at ambient conditions⁶. One of the prominent requisite in photovoltaic research is reducing the cost of solar cell. To minimize the cost, thin film technology has been evolved to ensure

suitable photovoltaic properties employing appropriate semiconducting materials possessing proper band gap and hence, to achieve maximum possible efficiency of a cell as per the Shockley-Queisser limit. A quartz substrate poses high purity, uniformity, smoothness, flatness, good heat and chemical resistance, high transmission and no fluorescence over a wide range of wavelengths from UV to IR. It has a lower coefficient of thermal expansion and negligible dielectric loss around GHz frequencies range^{7,8}. Therefore, quartz has been taken as substrate for fabrication of solar cell. As evident from recent developments in solar cell research, solar cells fabricated using promising absorber and window layers such as CdTe, CuO, SnS and TiO₂ are observed possessing reasonably better efficiency. CdTe, CuO, SnS semiconducting films are especially important due to their band gap of 1.5 eV, 1.2 eV, 1.3 eV which are near to the most appropriate value for photovoltaic conversion⁹⁻¹¹. Constituents of these compounds are not toxic except CdTe and the band gap of these films fall in the optimum range⁹ of terrestrial photovoltaic applications. CdTe films possess higher absorption coefficient, therefore, only a few microns of material thickness are required for photovoltaic applications. Recently, efficiency of 0.0017 %¹², 0.1 %¹³ and

*Corresponding author (E-mail: shashidharjss@gmail.com)

1.62%¹⁴ has been reported with TiO₂ / CdTe, TiO₂ / SnS and TiO₂ / CuO hetero junction cell structure.

2 Experimental Details

Various thicknesses of CdTe, CuO, SnS and TiO₂ films were synthesized using a simple, economical spray pyrolysis deposition (SPD) technique and subsequent films were characterized. For our present work, lower temperature deposition¹⁵ is one of the requirements, therefore, SPD technique has been used which will ensure wide area uniform deposition at lower temperature and ease of changing atomic ratio percentage of elements¹⁶. Semiconductor thin films of various thicknesses were prepared by changing the time of deposition. The composition of the films was optimized¹⁷ by controlling thickness to achieve better resistivity and photosensitivity of solar cells. Film composition, morphology, and electrical characteristics were studied by employing EDX, SEM and hot probe method.

2.1 Fabrication of solar cells

In present work TiO₂/CdTe, TiO₂/CuO, and TiO₂/SnS hetero-junction solar cells were synthesized on ITO coated quartz plates by spray pyrolysis technique at optimum conditions¹⁸. Anelva, SPF-332H make RF sputtering system (with specifications, such as plate voltage of 1-2 kV, plate current of 60-110 mA, incident power of 40-150 W, reflecting power of 5-25 W, pre-sputtering time of 5-20 min, target-substrate distance of 5.2 cm and a pressure of 6 m Torr) was employed for deposition of ITO films on cleaned quartz plates to be used as the substrates for the formation of hetero-junction. ITO films have high transmission and low sheet resistance (150 nm at substrate temperature of 150 °C and 300 °C, 35 Ω/□). Partly covered ITO quartz substrates were kept over a hot plate, maintained at a temperature of 350 °C. Required chemicals for synthesizing window and absorber layers are titanium tetra iso Propoxide AR (TTIP), (C₁₂H₂₈O₄Ti), acetyl acetone AR (AcAc), (CH₃-CO-CH₂-CO-CH₃), tellurium oxide (TeO₂), ammonia solution (25 % extra pure, sp.gr.0.91 abt, S D fine), hydrazine (mono) hydrate (N₂H₄H₂O), HCl, de-ionized water, cupric acetate (monohydrate) AR ((CH₃COO)₂ CuH₂O), stannous chloride AR (SnCl₂ 2H₂O), thiourea AR (CS (NH₂)₂) which were procured from Leo Chem, India, and absolute ethanol (C₂H₅OH), cadmium chloride (CdCl₂H₂O) from Changshu Yangyuan Chemical and Loba Chemie Pvt Ltd, India, respectively. Window layer of titanium

oxide (TiO₂) thin films of thickness 100 nm and 1 μm was deposited onto an exposed portion of the ITO by spraying a solution of 0.1 M TTIP and 0.2 M AcAc dissolved in absolute ethanol. Annealing time and temperature were estimated by performing isochronal and isothermal studies on each film to minimize resistivity. Finally, the TiO₂ coated ITO substrates were annealed in air at 400 °C for 30 min for reducing the resistivity of the films. Subsequently, air annealed TiO₂ coated ITO substrates were used for preparing multiple hetero-junctions by coating 1 to 10 μm of CdTe, CuO and SnS absorber layers of films at temperature of 350 °C, 300 °C and 270 °C, respectively. Cadmium Telluride (CdTe) thin films were prepared at 350 °C by spraying 0.01 M solution of Cadmium chloride and Tellurium oxide dissolved in a mixture of 1 part of ammonia solution and 4 parts of de-ionized water. In above prepared solution 1.5 ml of Hydrazine (mono) hydrate and 1 ml of HCl were added to get comprehensive solubility of precursors. Copper oxide (CuO) thin films were prepared by spraying a 0.01 M of cupric acetate (monohydrate) dissolved in double distilled water at 300 °C. Tin sulfide (SnS) thin films were prepared at 270 °C by spraying 0.01 M solution of stannous chloride and thiourea dissolved in de-ionized water. To achieve complete solubility of precursors, few drops of HCl were mixed in the above prepared solution. Thin films of TiO₂, CuO, CdTe and SnS were synthesized by spraying the respective precursors at optimum conditions. Solar cells annealed in air give better crystallinity and removes the chemisorbed water leads to achieve better efficiency than in vacuum. Performance of photovoltaic cell is further improved because of better positioning of work function and conduction band minimum of ITO which eventually enhance charge transportation¹⁹. For reducing resistivity CdTe, CuO and SnS films coated onto air annealed TiO₂ coated ITO substrates were subsequently annealed in air at 400 °C, 300 °C and 200 °C for durations of 120, 60 and 15 mins, respectively. Before metallization processes, various metals were deposited on prepared semiconductors to check for Ohmic or Schottky, metal-semiconductor (M-S) contact at the interface. After removing above annealed junctions from the hot plate, it is fixed inside a vacuum coating chamber of RF sputtering unit to deposit the gold, copper and aluminum metal contacts on the absorbing layers of junctions for better ohmic / Schottky contact. Gold and copper are coated over absorbing layers of junctions which are grown on

substrate maintained at a temperature of 150 °C and aluminum at 300 °C, respectively. Thus, the hetero-junctions of the type ITO/TiO₂/CdTe/Cu (30 nm) - Au (120 nm), ITO/TiO₂/CuO/Au (150 nm) and ITO/TiO₂/SnS/Al (150 nm) were obtained.

2.2 Sample characterization

TiO₂, CdTe, CuO, SnS semiconducting films prepared by spray pyrolysis technique and were characterized through SEM (scanning electron microscope) and EDX (energy dispersive X-ray). SEM imaging and chemical composition study was performed by using the ray line. The nature of semiconductor property was studied by hot probe technique. Structural and electrical characterizations of the films were performed using XRD, AFM, Raman, PL, four probe techniques and reported elsewhere²⁰⁻²⁴.

3 Results and Discussion

3.1 Chemical composition

Chemical composition of films was investigated using EDX analysis. Figure 1(a-d) illustrates EDX analysis of thin films and comparative study is tabulated in Table 1. It is found that impurity

elements are not present, and presence of silicon, titanium, oxygen, cadmium, tellurium, copper, tin, and sulfur are distinct. Atomic % of Ti, Cu, Te, Sn elements in TiO₂, CuO, CdTe, SnS films is observed increasing with film thickness. Stoichiometric films showed crystallinity^{25,26}.

3.2 Morphology

The morphology of films can be observed in SEM Figs 2(a-d). The average grain size of both thin and thick films is found to be 11.17 nm, 12.28 nm, 31.26 nm, 15.63 nm and 200 nm, 300 nm, 300 nm, 500 nm. It is found that grain size is increasing with increasing thickness of film²⁷.

3.3 Hot probe measurement

The hot-probe method by employing a standard multimeter and a heated probe is a simple and efficient way to differentiate between n and p-type semiconductors. The testing is done by placing cold and hot probe on the surface of the given semiconductor and both the probes are connected to a sensitive electrometer^{28,29}. Positive and negative voltage confirms, n-type and p-type, respectively on application of hot and cold probes on the surface of the semiconductor. In our present study, hot probe

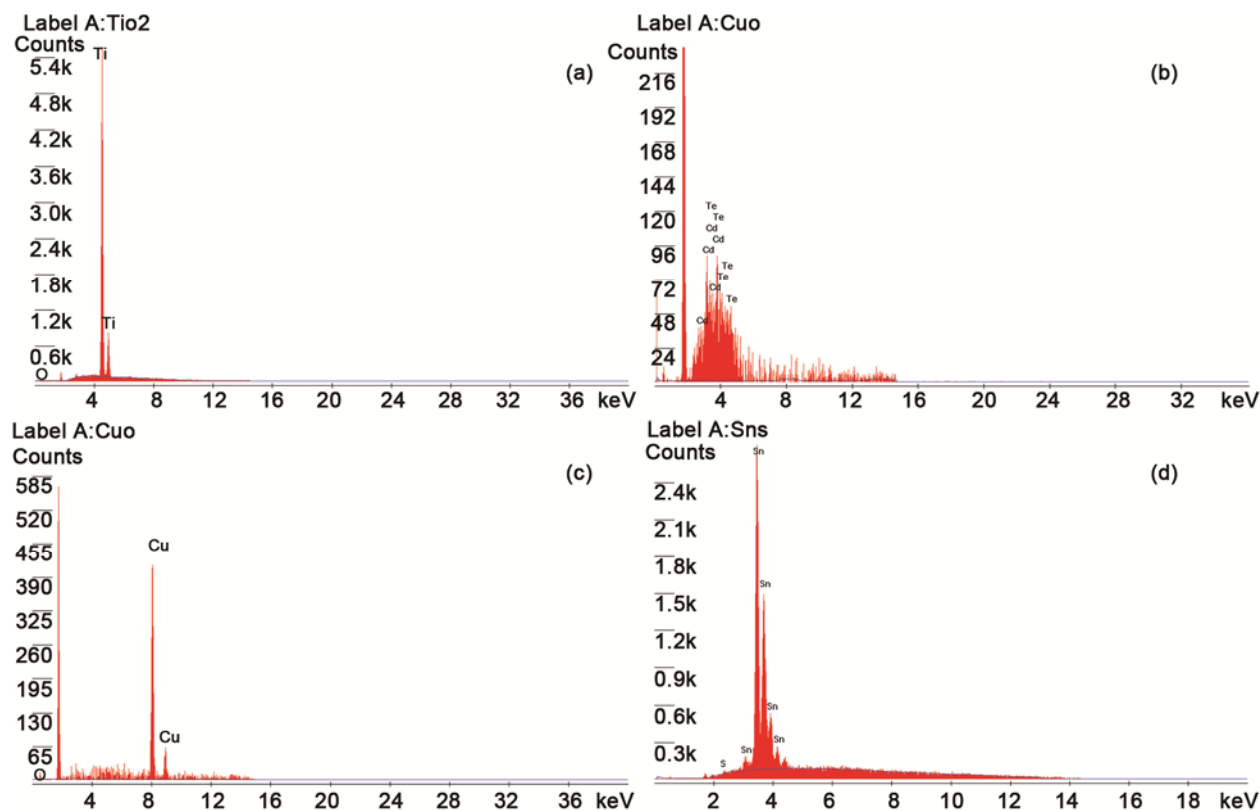


Fig. 1 — EDAX patterns of (a) TiO₂, (b) CdTe, (c) CuO and (d) SnS thick films.

Table 1 — EDAX comparative analyses of thick and thin films.

| Thick film | Element | Wt % | At % | Thick film | Element | Wt % | At % |
|------------------|---------|--------|--------|------------|---------|-------|-------|
| TiO ₂ | O K | 00.00 | 0.00 | CuO | O K | 0.35 | 1.36 |
| | Ti K | 100.00 | 100.00 | | Cu K | 99.65 | 98.64 |
| TiO ₂ | O K | 1.42 | 4.15 | CuO | O K | 0.44 | 1.73 |
| | Ti K | 98.58 | 95.85 | | Cu K | 99.56 | 98.27 |
| CdTe | Cd L | 40.93 | 44.03 | SnS | S K | 0.18 | 0.65 |
| | Te L | 59.07 | 55.97 | | Sn L | 99.82 | 99.35 |
| CdTe | Cd L | 37.19 | 40.20 | SnS | S K | 0.31 | 1.13 |
| | Te L | 62.81 | 59.80 | | Sn L | 99.69 | 98.87 |
| Thin film | Element | Wt % | At % | Thin film | Element | Wt % | At % |
| TiO ₂ | O K | 32.60 | 59.15 | CuO | O K | 10.22 | 31.13 |
| | Ti K | 67.40 | 40.85 | | Cu K | 89.78 | 68.87 |
| TiO ₂ | O K | 33.08 | 59.68 | CuO | O K | 12.56 | 36.32 |
| | Ti K | 66.92 | 40.32 | | Cu K | 87.44 | 63.68 |
| CdTe | Cd L | 34.39 | 37.31 | SnS | S K | 3.42 | 11.59 |
| | Te L | 65.61 | 62.69 | | Sn L | 96.58 | 88.41 |
| CdTe | Cd L | 37.96 | 40.99 | SnS | S K | 2.27 | 7.92 |
| | Te L | 62.04 | 59.01 | | Sn L | 97.73 | 92.08 |

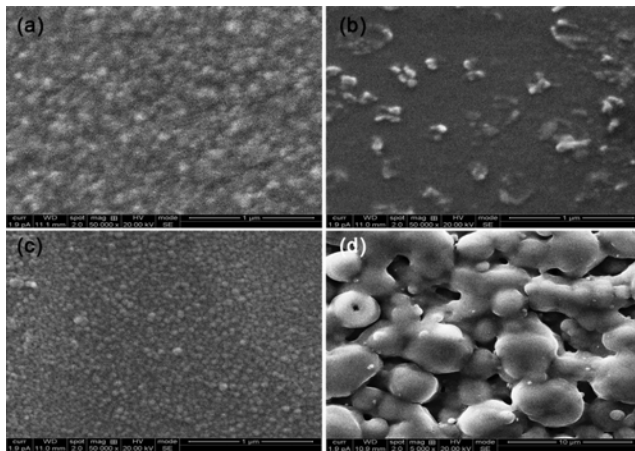


Fig. 2 — SEM patterns of (a) TiO₂, (b) CdTe, (c) CuO and (d) SnS thick films.

measurements reveal that the TiO₂ and CdTe, CuO, SnS are semiconductor films and exhibit n-type and p-type conductivity, respectively.

3.4 Optical response studies of solar cell

Semiconductor parametric analyzer (Agilent 4155 C) was used for electrical studies. The setup has been arranged as shown in Fig. 3 which shows the cross-section and band diagram of synthesized quartz based hetero junction solar cells. Solar cells were illuminated with 206 mW/cm² (air mass 5.6) through the quartz, ITO window and absorber layers. The plot of dark light I-V characteristics of quartz based both thin and thick film TiO₂/CdTe, TiO₂/CuO and TiO₂/SnS hetero junction solar cells are shown in Figs 4(a-c), 5(a and b), 6(a-c) and Fig. 4(d), 5(c) and

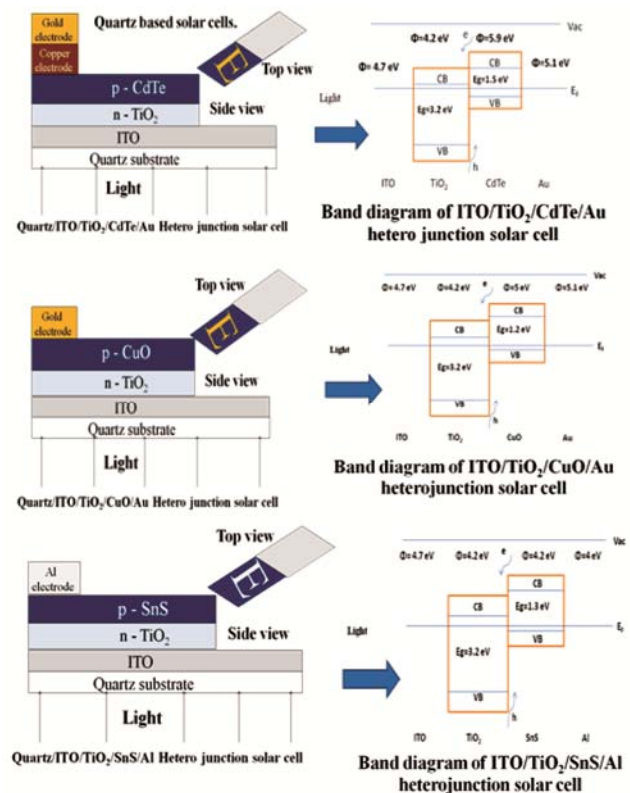


Fig. 3 — Cross section and band diagram of synthesized quartz based hetero junction solar cells.

6(d), respectively. It demonstrates rectifying response of both thin and thick solar cells. The performance results of quartz based both thin and thick hetero junction solar cells are tabulated in Table 2. Overall, result shows that quartz based thin and thick hetero junction solar cells have both lower short circuit

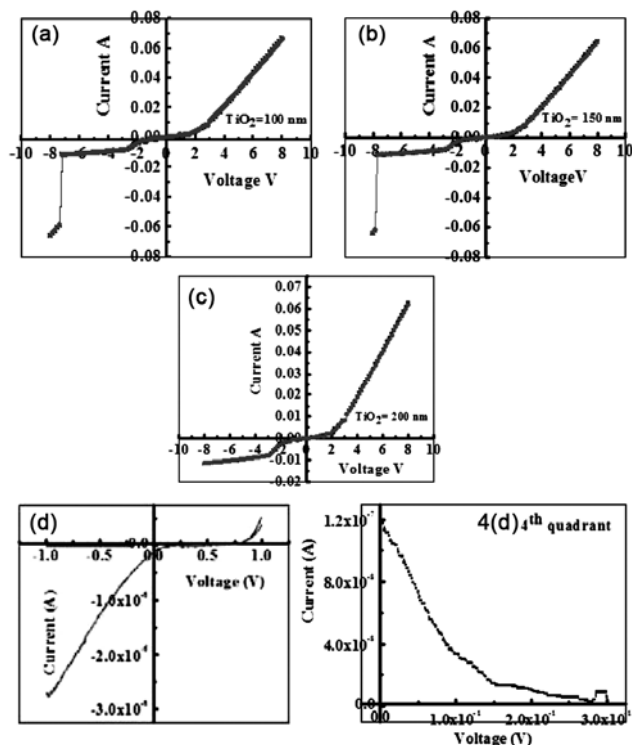


Fig. 4 — Dark-light I-V characteristics showing rectifying behavior of thin and thick film TiO_2/CdTe quartz based hetero-junction solar cells.

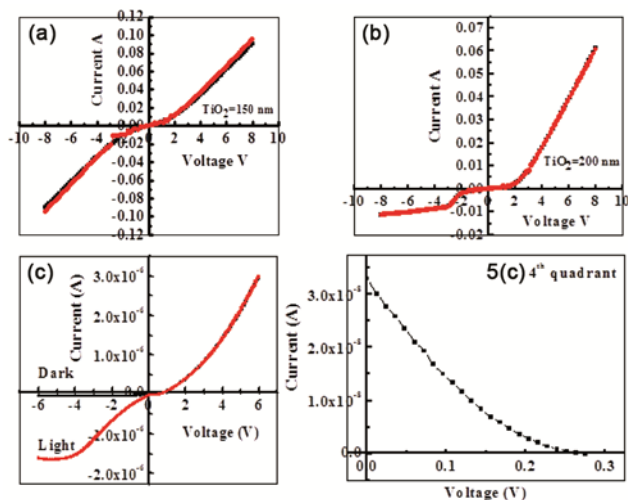


Fig. 5 — Dark-light I-V characteristics showing rectifying behavior of thin and thick film TiO_2/CuO quartz based hetero-junction solar cells.

current (I_{sc}) and open circuit voltage (V_{oc}), low fill factor (FF) and therefore, small efficiency (η). Low shunt resistance at the junction is another key aspect which leads to decrement in both open circuit voltage and the fill factor. Lower shunt resistance may be due to the dominant mechanism of interface

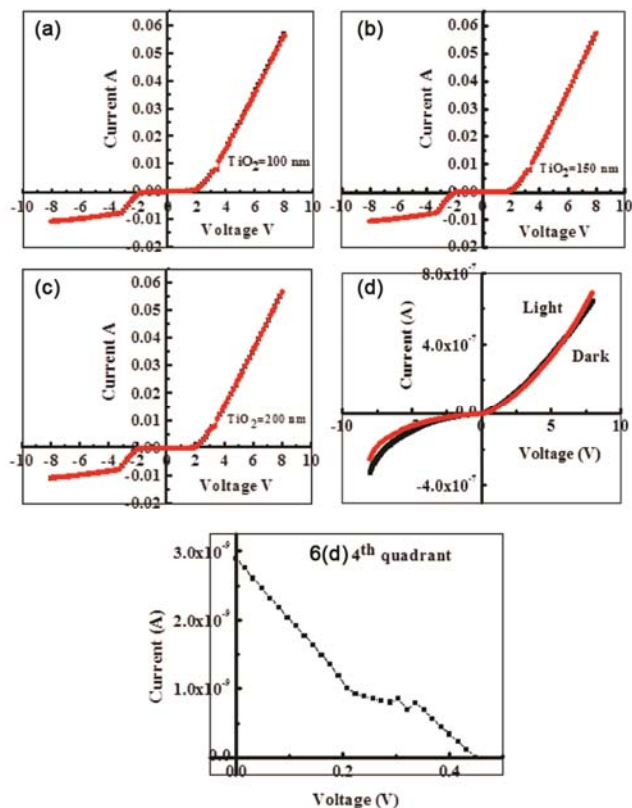


Fig. 6 — Dark-light I-V characteristics showing rectifying behavior of thin and thick film TiO_2/SnS quartz based hetero-junction solar cells.

recombination at the junctions and a reunion of charge carriers at the grains and defects that exist in large numbers in spray pyrolyzed films³⁰. Lower cell efficiencies are due to the study carried out at higher air mass of 5.6, which leads to solar irradiance peak shift towards higher wavelength and attenuation of shorter wavelengths due to Rayleigh's scattering³¹, smaller values of V_{oc} and FF. Smaller fill factor is due to enhancement of series resistance and more over open circuit voltage depends upon the material rather than the cell structure³². Thick film solar cells are found to possess reasonably good cell parameters due to improved film properties such as resistivity, conductivity, charge concentration and mobility. Therefore, stable cell performance has been observed even experiment has been carried out in the open air, without protection against moisture. The lower resistivity of TiO_2 , CuO , CdTe , SnS films may be due to excesses element At % of Ti, Cu, Te, Sn^{33,24} and enhancement of carrier concentration due to increasing thickness. However, lower film thickness leads to poor crystallinity with few atomic layers in

Table 2 — The performance results of thin and thick film quartz based hetero-junction solar cells (Incident power $P_{in} = 206 \text{ mW/cm}^2$, Area of solar cell $A = 0.5 \text{ cm}^2$).

| Film | Hetero-junction solar cell | R_{Series} (dark) Ω | R_{Series} (light) Ω | R_{Shunt} (dark) Ω | R_{Shunt} (light) Ω | V_m mV | i_m μA | V_{oc} mV | i_{sc} μA | FF % | Eff 10^{-6} % |
|-------|--|---------------------------------|----------------------------------|--------------------------------|---------------------------------|-------------|------------------------|----------------|---------------------------|---------|--------------------|
| Thin | TiO ₂ (100 nm) / CdTe (1 μm) | 197.53 | 164.93 | 186.05 | 158.41 | - | - | - | - | - | - |
| | TiO ₂ (150 nm) / CdTe (1 μm) | 250 | 168.42 | 192.77 | 172.04 | - | - | - | - | - | - |
| | TiO ₂ (200 nm) / CdTe (1 μm) | 320 | 133.33 | 202.5 | 183.91 | - | - | - | - | - | - |
| Thick | TiO ₂ (1 μm) / CdTe (10 μm) | 9.47 M | 0.79 M | 8.58 M | 0.26 M | 74 | 0.0467 | 277 | 0.1197 | 10.42 | 3.35 |
| Thin | TiO ₂ (100 nm) / CuO (1 μm) | - | - | - | - | - | - | - | - | - | - |
| | TiO ₂ (150 nm) / CuO (1 μm) | 140.35 | 105.26 | 727.27 | 571.42 | - | - | - | - | - | - |
| | TiO ₂ (200 nm) / CuO (1 μm) | 800 | 262.29 | 197.53 | 195.12 | - | - | - | - | - | - |
| Thick | TiO ₂ (1 μm) / CuO (10 μm) | 600 M | 0.79 M | 34.68 M | 7.89 M | 108 | 0.0134 | 274 | 0.0326 | 16.19 | 1.405 |
| Thin | TiO ₂ (100 nm) / SnS (1 μm) | 301.88 | 213.33 | 258.06 | 246.15 | - | - | - | - | - | - |
| | TiO ₂ (150 nm) / SnS (1 μm) | 243.27 | 241.15 | 238.66 | 215.63 | - | - | - | - | - | - |
| | TiO ₂ (200 nm) / SnS (1 μm) | 243.98 | 225.76 | 256.16 | 245.70 | - | - | - | - | - | - |
| Thick | TiO ₂ (1 μm) / SnS (10 μm) | 16.6 M | 12.0 M | 115 M | 77.5 M | 336 | 0.0007 | 449 | 0.0028 | 20.3 | 0.0002 |

disordered form and therefore, a large number of defects evolves as a result of incomplete bonding as the atoms at the poor crystallized regions are disordered. Due to large defects, mobile charge carriers are electrically trapped inside the defect, leading to the formation of energy barrier which eventually avoids the motion of charge carriers from one crystallized region to another.

Hence, it has been found that the decrement in the carrier concentration at the poor crystallized region with thickness, which increase film resistivity³⁴⁻³⁶. Improved conductivity may be due to enhancement of grain size as a function of thickness and subsequent variation of the optical band gap. Thicker films contain less structural defects compared thinner films leading to enhancement of activation energy at the interface³⁷.

TiO₂/ SnS hetero-junction solar cell, is found to possess almost zero efficiency which may be because of the strong dependence of sulfur content in the film, which changes depending upon the temperature of the previous runs, leading to irreproducibility, as one of the main limitations for the development of SnS based devices¹¹.

In present work, all hetero-junctions including TiO₂/ CdTe solar cell study is being done at 206 mW/cm² (AM 5.6), therefore, estimated value of series and shunt resistance of cell is high. However, the same can be further reduced by minimizing the light intensity near (AM 1.5), as the I-V characteristics of a solar cell is light dependent. The reported result in the present work is in good agreement³⁸.

TiO₂/ CuO heterojunction solar cell is found to possess 3.35×10^{-6} % efficiency, which is in good agreement of previous work³⁹. TiO₂ being a n-type semiconductor have a wide bandgap, the affinity of 3.2 eV, 1.6 eV as an active material, capture photon generating electron hole pair, while CuO, a p-type semiconductor with a narrow band gap of 1.2 eV widen the spectrum of light of TiO₂ to capture a large number of photons. Excess copper having a work function of 4.7 eV, provides many tracks for the electrons that have formed on the conduction band of TiO₂. Across the TiO₂-CuO Copper interface, electrons forward quickly with ease towards the electrode by thermionic process due to the Schottky barrier⁴⁰⁻⁴³ formed at the interface. This is because the work function of copper is higher than the

semiconductor affinity of TiO₂ semiconductor, in the absence of copper electron reach electrode by diffusion process only⁴⁴. Enhancement of cell efficiency might be due to improvement of copper content leading to better transportation of electrons by the thermionic process at the interface¹⁴. Higher cell efficiency may also be due to lower resistivity in CuO thin films, which may be due to excess copper atomic percentage or carrier degeneracy as a result of continuous distribution of defect states or formation of mixed phase of Cu₂O-CuO in the grain or enhancement of hole concentration and mobility^{33,24}.

4 Conclusions

Quartz based hetero junction thin films and thick films solar cells synthesized by low cost non-vacuum spray pyrolyzed technique showed excellent rectifying behavior and reasonably good response. The composition of the films is found to change with the film thickness leading to better value of resistivity and photo sensitivity. Thick film solar cells are found possessing reasonably good cell parameters, compared to thin film solar cells. Even though, the measurements of cell parameters have been done in open air, against the protection of air and moisture, obtained stable cell parameters shows better stability of spray pyrolyzed films. TiO₂/CuO hetero-junction solar cells are found to possess better optical and electrical cell response as compared to other cells due to the Schottky barrier formation at the interface. The efficiency of the solar cells can be further improved by improving the quality, stoichiometry, crystallinity, reducing the resistivity of the sprayed semiconductor thin film layers by further annealing in air or vacuum or doping, and performing cell study as a function of Air Mass which may lead to mass production of non-silicon based solar cell.

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