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Effect of processing parameter on structural, optical and electrical properties of photovoltaic chalcogenide nanostructured RF magnetron sputtered thin absorbing films

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ABSTRACT

The aim of this work was to develop high quality of $CuIn_{1-x}Ga_xSe_2$ thin absorbing films with x (Ga/In+Ga) < 0.3 by sputtering without selenization process. $CuIn_{0.8}Ga_{0.2}Se_2$ (CIGS) thin absorbing films were deposited on soda lime glass substrate by RF magnetron sputtering using single quaternary chalcogenide (CIGS) target. The effect of substrate temperature, sputtering power & working pressure on structural, morphological, optical and electrical properties of deposited films were studied. CIGS thin films were characterised by X-ray diffraction (XRD), Field emission scanning electron microscope (FE-SEM), Energy dispersive X-ray spectroscopy (EDAX), Atomic force microscopy (AFM), UV–vis–NIR spectroscopy and four probe methods. It was observed that microstructure, surface morphology, elemental composition, transmittance as well as conductivity of thin films were strongly dependent on deposition parameters. The optimum parameters for CIGS thin films were obtained at a power 100 W, pressure 5 mT and substrate temperature 500 °C. XRD revealed that thin film deposited at above said parameters was polycrystalline in nature with larger crystallite size (32 nm) and low dislocation density (0.97×10^{15} lines m^{-2}). The deposited film also showed preferred orientation along (112) plane. The morphology of the film depicted by FE-SEM was compact and uniform without any micro cracks and pits. The deposited film exhibited good stoichiometry (Ga/In+Ga=0.19 and In/In+Ga=0.8) with desired Cu/In+Ga ratio (0.92), which is essential for high efficiency solar cells. Transmittance of deposited film was found to be very low (1.09%). The absorption coefficient of film was $\sim10^5\ cm^{-1}$ for high energy photon. The band gap of CIGS thin film evaluated from transmission data was found to be 1.13 eV which is optimum for solar cell application. The electrical conductivity ($7.87\ \Omega^{-1}\ cm^{-1}$) of deposited CIGS thin film at optimum parameters was also high enough for practical purpose.

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1. Introduction

$CuIn_{1-x}Ga_xSe_2$ (CIGS) has emerged as one of the most important polycrystalline semiconducting material in solar cell application because of their stability against photo degradation. It belongs to the group I–III–VI_2 semiconducting material and possesses chalcopyrite structure. It exhibits high optical absorption coefficient ($\sim10^5\ cm^{-1}$) for high energy photons (1.5 eV). Also, its high conversion efficiency and excellent anti irradiation performance make it possible to reduce the thickness of absorbing layer in few micrometre [1]. Its tuneable band gap energy (1.0–1.7 eV) allows us tailoring the optical band gap for

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high efficiency solar cell [2]. The efficiency of photovoltaic (PV) cell strongly depends on many factors including stoichiometry, microstructure, presence of impurity phases and surface roughness for a given area of deposited film. The basic requirement for high efficiency solar cell based on CIGS is high mobility, chalcopyrite structure with good stoichiometry and microstructure with columnar grains [3]. Apart from this, Cu deficient absorbing layer is the favourable condition for high performance of solar cell because majority carrier (hole) concentration increases by increasing the density of Cu vacancies [4]. Solar cells based on CIGS have shown long term stability. The PV cell using CIGS as absorbing layer have reached the record efficiency and recently it has been grown up to 19.9% and 20.3% reported by NREL [5] and Zentrum fur wasserstoff [6] respectively. Various researchers have used different techniques to deposit CIGS thin films for high efficiency solar cell. Among these techniques, co-evaporation has emerged as main deposition technique [7]. Three stage co-evaporation processes for CIGS thin films deposition involve the evaporation of Cu, In, Ga and Se using Cu-rich as well as Cu- poor growth condition. However, this method requires precise control of individual source elements during deposition. Also, this technique involves some disadvantages like costly equipment and slow deposition rate hence; it is difficult to apply this technique for large scale production. The module efficiency based on co-evaporation process has reached only 12–13%. Another method for CIGS thin films fabrication is post selenization of precursor layer [8]. The module efficiency using post selenization process has reached up to 16%. The benefit of this technique is the maintenance of uniformity of film over large area with high deposition rate. However, this process involves toxic gases (Se or H_2Se). Hence, this technique is harmful concerning the environment as well as health. Various scientists have also used different deposition techniques for CIGS thin films deposition like chemical bath deposition[9], electrodeposition [10], electron beam evaporation [11], inkjet printing [12] and laser assisted deposition [13]. Several groups have also used magnetron sputtering of ternary and quaternary alloy followed by post selenization [8]. Post selenization was performed to improve the crystallinity as well as to get the high quality chalcopyrite product. This is a simplified fabrication method because of reducing the number of targets.

Wei et al. reported that the best solar cell efficiency can be achieved with x (Ga/In+Ga) ≤ 0.3 [14]. It is also reported that when $x > 0.3$, the efficiency is decreased because open circuit voltage is not proportionally increased with band gap. Hence $CuIn_{1-x}Ga_xSe_2$ thin film works effectively only for a limited range of x. There is a limited study on CIGS thin films with $x = 0.2$. Apart from this, there is no systematic research that deals with optimisation of processing parameter of RF magnetron sputtering to achieve high quality of $CuIn_{0.8}Ga_{0.2}Se_2$ thin films. In present work, $CuIn_{0.8}Ga_{0.2}Se_2$ (CIGS) thin films were fabricated on soda lime glass by RF magnetron sputtering using single quaternary target without any selenization. The desired phase as well as microstructure, good stoichiometry, low band gap and high conductivity has been optimised by varying the processing parameter including substrate temperature, sputtering power and working pressure.

2. Experimental

$CuIn_{0.8}Ga_{0.2}Se_2$ (CIGS) thin films were deposited on ultrasonically cleaned soda lime glass (SLG) substrate by RF magnetron sputtering. A 2 in. diameter, single quaternary chalcogenide CIGS target with purity 99.999% was used as sputtering target. The elemental composition (atomic %) of CIGS target was Cu = 25%, In = 20%, Ga = 5% and Se = 50%. CIGS target and SLG substrate was placed inside the vacuum chamber. The distance between target and substrate was kept 6 cm. Prior to the film deposition; chamber was first evacuated to high vacuum of 3×10^{-6} Torr with the help of turbo molecular pump backed by rotary pump. Ar gas was used as sputtering gas with a flow of 20 sccm. During deposition substrate temperature, power and pressure was varied keeping deposition time 40 min. Before each thin film deposition, pre sputtering was performed for 15 minutes. The deposition parameters for CIGS thin films are shown in Table 1.

3. Characterisation

Structural analysis of deposited CIGS thin films was carried out by Bruker D8 Advance X-ray diffractometer (XRD) using CuK_α radiation ($\lambda = 0.154016$ nm) at a tube angle of 2°. Surface morphology as well as cross-sectional view of CIGS thin films were analysed by field emission scanning electron microscopy (FE-SEM: FEI Quanta 200 F). Elemental composition of CIGS thin films was investigated by energy dispersive X-ray analysis (EDAX) attached with FE-SEM. Surface roughness of deposited films was evaluated using Atomic force microscopy (AFM: NTEGRA) operated in semi contact mode. Optical transmittance and absorbance of CIGS thin films were measured by UV–vis–NIR spectrophotometer (Varian Cary 5000) in 400–1500 nm wavelength range. Electrical conductivity of deposited CIGS thin films was measured by four probe method equipped with Kithley electrical measurement unit (2182 A nanovoltmeter, 6221 current source).

Table 1
Deposition parameters for CIGS thin films.

Target	Substrate	Base pressure (Torr)	Working pressure range (mTorr)	Power range (Watt)	Substrate temperature range (°C)	Ar gas flow (sccm)	Deposition time (min)
$CuIn_{0.8}Ga_{0.2}Se_2$	SLG	3.0×10^{-6}	3.5–9	40–100	RT-500	20	40

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