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# Studied on the graded band-gap copper indium diselenide thin film solar cells prepared by electrochemical route



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#### ABSTRACT

A graded band-gap CuInSe<sub>2</sub> (CIS) thin film solar cell (TFSC) having glass/FTO/CdS/CIS multilayer/Au structure has been fabricated. A simple and low-cost electrodeposition technique is used to deposit the multilayers of CIS onto fluorine doped tin oxide (FTO) coated glass substrate. A conventional three-electrode geometry consisting, FTO, graphite and Ag/AgCl as a working, counter and reference electrodes, respectively was used for electrodeposition. Structural characterization was carried out using X-ray diffraction (XRD) and Raman spectroscopy, which revealed the chalcopyrite tetragonal CIS structure with a quite Cu-rich surface which reduces upon selenization. The morphology of the as grown and selenized CIS multilayer thin films was studied by using atomic force microscopy (AFM) which shows the compact and uniform layer formation. The depth profile distribution of individual elements in both as-grown and selenized CIS multilayer thin films has been determined using secondary ion mass spectroscopy (SIMS). SIMS results revealed that the proposed graded band gap structure is retained even after selenization. The presence of Cu<sup>+</sup>, In<sup>3+</sup> and Se<sup>2-</sup> oxidation states were confirmed using Xray photoelectron spectroscopy (XPS). A single layer and multilayer CIS solar cell devices yielded ~5.10% and  $\sim$  7.20% power conversion efficiency, respectively. In the present work, pH 3 buffer solution helps to improve the morphology of CIS layer which gives the better power conversion efficiency as compared to the previously reported value.

## 1. Introduction

The inadequacy of fossil fuel resources, population growth and global warming facilitate to the requirements of new renewable energy sources, such as solar energy. The copper indium diselenide (CIS) based chalcopyrite solar cells are more advantageous among the all photovoltaic technologies due to the direct and tuneable band gap, large mean free path of charge carriers, high absorption coefficient and extraordinary stability [1,2]. The maximum efficiency of CIS based device has been achieved up to  $\sim 22.6\%$  by vacuum deposition technique [3]. The vacuum based system are expensive and their day to day maintenance is high, which is increases the cost of the TFSC devices. The low-cost solution processable techniques have attracted attention owing to the low-temperature growth, less manufacturing cost and possibility to have the high throughput [4-7]. Bhattacharya et. al, reported ~15.4% efficiency of CIGS solar cell using electrodeposition method with excess Ga addition by physical vapour deposition [8].

Despite of significant economic and technical advantages, CIS based solar cells suffers relatively low-efficiency compared to the theoretical

estimated efficiency  $\sim 32.21\%$  [9]. Thus, in order to improve the performance of CIS based TFSC, it is essential to develop different strategies. Researcher now extensively focussing on the improvements of solar cells by enhancing their electrical and optical properties by harvesting shorter wavelength photons to minimize the absorption losses [10]. Several reports are available on dealing with various aspects of CIS based solar cells [11-13]. Recently the development of nanoparticles based solar cell achieves the efficiency  $\sim 12\%$  [14]. The graded band gap structure of CIS is proposed by several groups with an enhancement in the efficiency of TFSC [15-16]. The development of a graded band gap CIS layer is reported by Chaure et al using electrodeposition technique with efficiency  $\sim 6.5\%$  [17]. It is well known that the energy band-gap of CIS depends on the content of individual elements [18]. This property of CIS can be utilized to tailor the band gap which not only help to prevent the recombination in solar cell and help to harvest the maximum solar spectrum. The contents of individual element in the deposit can be varied by changing the growth potential. Recently, we have also reported the effect of rapid thermal annealing and normal selenization onto the CIS TFSC devices prepared by

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**Fig. 1.** Typical schematic used to grow the multilayer CIS thin films on FTO substrate using electrodeposition at various growth potentials with respect to Ag/AgCl reference electrode.

electrochemical route [19]. We have reported the effect of electrodeposition potentials on the properties on CIS layers [20].

Herein, the work on development of CIS solar cells using graded band-gap structure is described. The concept of graded band gap based CIS solar cell is demonstrated by the formation of p and n-type conducting CIS layers having different energy band gap values. This type of structure could be employed to any chalcopyrite material to get high performance for TFSC devices.

#### 2. Experimental details

## 2.1. Chemicals

Copper chloride (CuCl<sub>2</sub>), indium chloride (InCl<sub>3</sub>), selenous acid (H<sub>2</sub>Se<sub>2</sub>O<sub>3</sub>) and Lithium chloride (LiCl) were purchased from Sigma-Aldrich of purity at least 99.9%. Fluorine doped tin oxide (FTO) coated glass substrates of sheet resistance  $10-15 \Omega/\Box$  were purchased from Pilkington glass company, UK. The double distilled deionized water (DDDW) was used as a solvent.

#### 2.2. Electrodeposition of CIS layers

Potentiostatic electrodeposition was used to deposit CIS thin films. A convectional three-electrode assembly consisting Ag/AgCl, graphite plate and FTO coated glass substrates were used as reference, counter and working electrode, respectively. The substrates were cleaned thoroughly with DDDW in ultrasonic bath. Finally the substrates were ultrasonicated in acetone and *iso*-propanol. The electrolytes contained 2 mM CuCl<sub>2</sub>, 4 mM InCl<sub>3</sub> and 3 mM H<sub>2</sub>Se<sub>2</sub>O<sub>3</sub> were dissolved in pH hydrion buffer (pH 3) solution. Lithium chloride (LiCl) was used as a supporting electrolyte. Cyclic voltammetry (CV) measurement was recorded to study the electrochemical behaviour of CIS system at scan rate 5 mV/sec. The electrodeposition of CIS thin films were carried out at different deposition potentials with respect to Ag/AgCl reference electrode without external agitation at room temperature. A schematic of multilayer CIS structure is depicted in Fig. 1. Immediately after deposition, the samples were rinsed in warm DDDW to remove the loosely



Fig. 2. (a) Cyclic voltammogram for CIS system recorded at 5 mV/sec, (b) chronoamperometric curves for multilayer CIS deposition, (c) plot for atomic concentration for elements in individual CIS layers and (d) PEC signals obtained for CIS layers grown at various deposition potentials.

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