

The structural and material properties of CuInSe_2 and $\text{Cu}(\text{In,Ga})\text{Se}_2$ prepared by selenization of stacks of metal and compound precursors by Se vapor for solar cell applications

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ABSTRACT

CuInSe_2 films and related alloys were prepared by thermal evaporation of Cu, InSe and GaSe compounds instead of elemental sources. Band-gap tailoring in $\text{Cu}(\text{In,Ga})\text{Se}_2$ -based solar cells is an interesting path to improve their performance. In order to get comparable results, solar cells with Ga/(In+Ga) ratios $x = 0$ and 0.3 were prepared, all with a simple two-step sequential evaporation process. The morphology of the resulting films grown at 550 °C was characterized by the presence of large faceted chalcopyrite grains, which are typical for device quality material. It is important to note that absorber films with elemental gallium resulted in a significant decrease in the average grain size of the film. The X-ray diffraction (XRD) diffraction pattern of single-phase $\text{Cu}(\text{In,Ga})\text{Se}_2$ films depicts diffraction peaks shifting to higher 2θ values compared to that of pure CuInSe_2 . The photoluminescence (PL) spectrum of $\text{Cu}(\text{In,Ga})\text{Se}_2$ thin films also depicts the presence of the peak at higher energy that is attributed to the incorporation of gallium into the chalcopyrite lattice. As the band gap of CIGS increases with gallium content, desirable effects of producing higher open-circuit voltage and low current density devices were achieved. A corresponding increase in device efficiency with gallium content caused by a higher fill factor was observed. The best results show passive area efficiencies of up to 10.2% and open-circuit voltage (V_{oc}) up to 519 mV at a minimum band gap of 1.18 eV.

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1. Introduction

Chalcopyrite compounds of CuInSe_2 and related alloys are promising materials with low cost and high efficiency for solar cells. Small-area solar cells, based on alloys between CuInSe_2 and $\text{Cu}(\text{In,Ga})\text{Se}_2$, have achieved record efficiencies close to 19.2 [1–4]. The band gap of CuInSe_2 can be tailored by the systematic replacement of In with smaller group III atoms such as Ga or Al. This results in a decrease in lattice parameters of the chalcopyrite lattice and ultimately an increase in the band gap. These high-efficiency $\text{Cu}(\text{In,Ga})\text{Se}_2$ devices have a typical Ga/(In+Ga) ratio x of 0.1–0.3 and band gap energy E_g between 1.0 and 1.2 eV. In classical two-step growth processes, Cu–In–Ga metallic precursors are selenized in elemental Se vapor or a $\text{H}_2\text{Se}/\text{Ar}$ gas mixture to form $\text{Cu}(\text{In,Ga})\text{Se}_2$. A seemingly insurmountable disadvantage of these processes is that the resultant semiconductor absorber films are heterogeneous. Due to the difference in the reaction rates between the binary selenides, the bulk of the material contains discrete CuInSe_2 and CuGaSe_2 phases [5]. Alternatively, regardless of its location in the precursor stack, graded film structures are

obtained with most of the gallium located at the back of the film and indium at the film surface [6]. This implies that gallium does not increase the band gap of the absorber in the active region of the solar cell, and complete devices are therefore limited to relatively low open-circuit voltages.

In order to investigate and ultimately solve these reported growth-related problems, the concentration of gallium in the quaternary alloys was altered by variation in the relative thickness of GaSe with respect to InSe and Cu during the formation of the precursors. From these systematic studies, it was possible to predict optimum growth conditions for the deposition of single-phase $\text{Cu}(\text{In,Ga})\text{Se}_2$ with a modified band gap.

2. Experimental procedure

2.1. Absorber formation

In this study various stacked layers were deposited by the thermal evaporation method. CuInSe_2 precursors composed of stacks of InSe/Cu/InSe were prepared on glass/Mo substrate at 200 °C and material properties of the resulting compounds were analyzed. These precursors were deposited on Mo films, prepared

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under identical and optimized conditions, so that the underlying Mo layer had very little influence on the morphology of the final alloys. To prepare $\text{Cu}(\text{In,Ga})\text{Se}_2$, gallium-containing precursors were deposited by sequential evaporation of respective elements or compounds. In order to obtain monophasic $\text{Cu}(\text{In,Ga})\text{Se}_2$ thin films, the GaSe layers needed to be deposited at or above 300°C , while the InSe and Cu layers were deposited at 200°C [7]. All CIGS thin films had $\text{Cu}/(\text{In}+\text{Ga})$ atomic ratios of approximately 0.9. The relative thickness of InSe with respect to GaSe was varied to achieve various $\text{Ga}/(\text{Ga}+\text{In})$ ratios. These precursor films were

subsequently reacted with elemental Se vapor at 550°C for 60 min. In this study two series of absorbers namely, pure CuInSe_2 and $\text{CuIn}_{0.7}\text{Ga}_{0.3}\text{Se}_2$, were prepared to compare structural and material properties using various characterization techniques.

2.2. Characterization

The surface morphologies and crystalline phases were examined by scanning electron spectroscopy (SEM) and X-ray

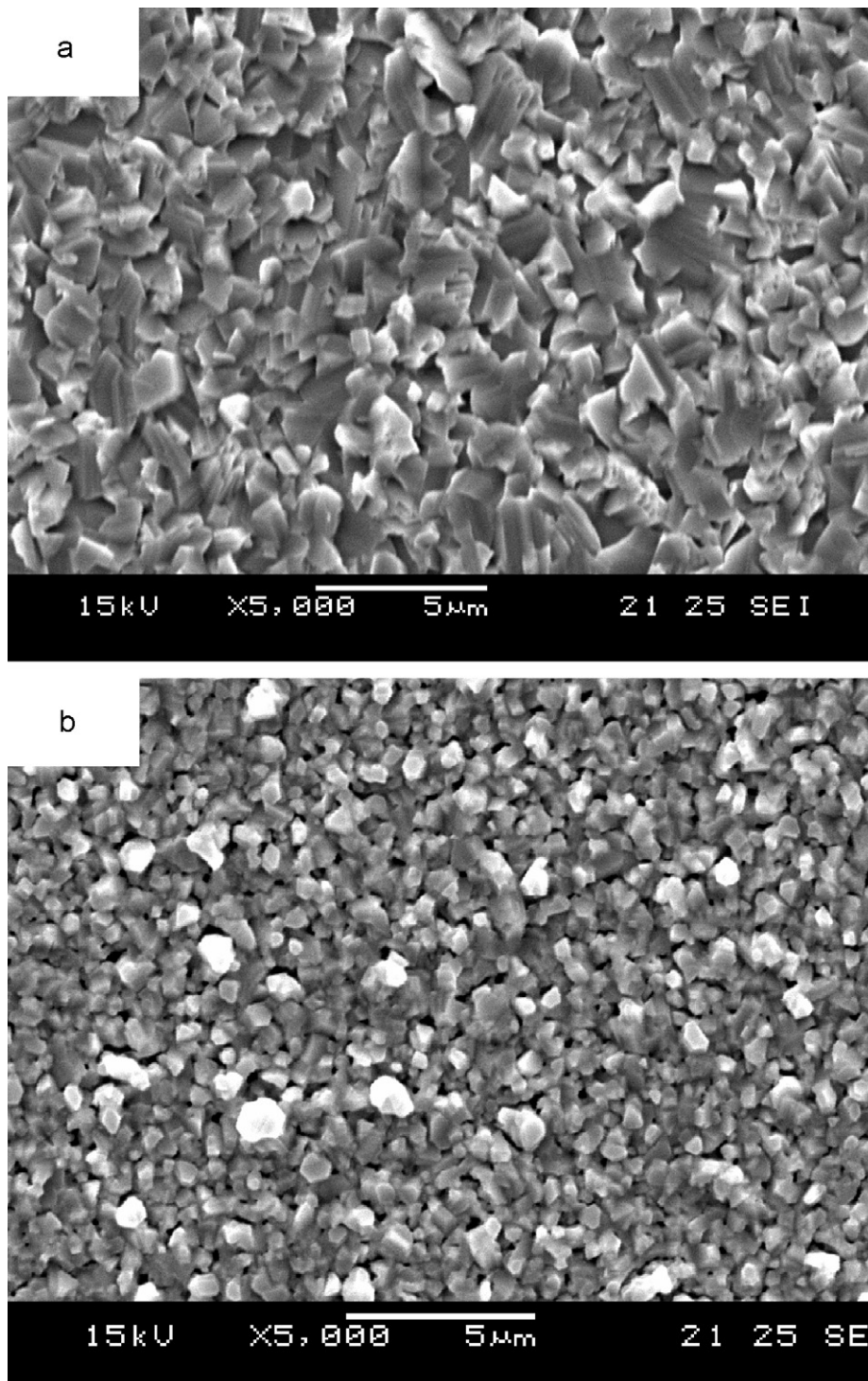


Fig. 1. SEM micrographs depicting the typical morphological features of single-phase (a) CuInSe_2 and (b) $\text{CuIn}_{0.7}\text{Ga}_{0.3}\text{Se}_2$ thin films.

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