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Fabrication of $Cu(In,Ga)Se_2$ thin films solar cell by selenization process with Se vapor

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Abstract

CIGS films were prepared on Mo-coated glass by sputtering and selenization processes. The metallic precursors were selenized under higher pressure in selenium vapor instead of H_2Se. In order to improve the performance of CIGS thin film solar cells, the morphologies of CIGS thin films were studied carefully by various temperature profiles. The relationship between temperature decrease rate and fill factor (FF) of solar cells was investigated thoroughly. On the other hand the value of open circuit voltage (V_{oc}) was improved by increasing the gallium content near the surface of CIGS thin film. A glass/Mo/CIGS/CdS/ZnO cell was fabricated and the conversion efficiency of 9.4% was obtained without antireflective film.
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Keywords: Selenization; CIGS solar cell; Solid Se source

1. Introduction

$CuIn_{1-x}Ga_xSe_2$(CIGS) chalcopyrite compound semiconductors of various composition have received much attention as the absorbing layers in high-efficiency polycrystalline thin-film solar cell structures (Tarrant and Gay, 2002; Delahoy et al., 2002). In this study, device-quality $Cu(In,Ga)Se_2$ thin films were prepared by a relatively simple two-stage growth technique. The metallic precursors were deposited sequentially on Mo/glass substrates using Cu–Ga alloy and In targets by DC magnetic sputtering system. These metallic precursors were then selenized with solid Se pellets in a closed vacuum furnace. This technique has the advantage of lowering danger by eliminating the use of toxic H_2Se gas. We had investigated physical and electric properties of the CIGS thin films by different temperature profiles under controlled Se vapor pressure.

Various selenization temperature has been applied in order to produce $Cu(In,Ga)Se_2$ chalcopyrite thin films with device-quality. In the interest of improving material properties of the final compound films, a large number of absorbing layers

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were grown and processed into solar cells during this study. We experimented with various heating rates during selenization. It was found that the thickness of CIGS films prepared by fast heating rate was smaller than other samples prepared by slow heating rate, the difference is about 0.15 μm. It may mean that the fast increasing rate of temperature has a good influence upon the density of CIGS thin films. On the other hand, we studied the effects of temperature decreasing rate on the fill factor (FF). Faster decreasing rate results in bad adhesion between Mo substrate and CIGS film, and the FF of solar cell well degrade severely. Fabricating by the best selenization temperature profile, the CIGS films show a smooth surface morphology and contains a single chalcopyrite phase.

Recent years, many research institutes proposed that the V_{oc} of solar cell might be improved by controlling the grade of gallium content in CIGS thin film. The fill factor loss was eliminated by introducing a Ga/(Ga + In) grading distribution, which also resulted in an increased open-circuit voltage of 20–30 mV for all CIGS solar cells (Lundberg et al., 2003). A back-surface field (BSF) is achieved since a higher concentration of gallium, which mainly increases the conduction band level in the CIGS film (Dullweber et al., 2000). A reduced back-contact recombination has earlier been demonstrated by the use of such a BSF (Dullweber et al., 2001). In addition, the higher gallium concentration nearing films surface may improve the V_{oc} of solar cells to a great extent (Britt et al., 2000). In our study, the gallium content gradient was controlled by various sputtering sequences and time, and we found that the gallium content nearing surface of CIGS thin films has a great influence on the performance of solar cells.

2. Experiment

A Mo back contact with a thickness of 800 nm was deposited on a soda-lime glass substrate by DC magnetic sputtering in Ar gas. The soda-lime glass substrates were not heated during the sputtering deposition. A CIGS absorbing layer was deposited by two-step method. Firstly, the Cu/Ga/In alloy precursors of approximately 800 nm thickness were deposited on Mo coated glass substrates using DC magnetic sputtering. Secondly, the metallic precursors were selenized with Se vapor in a closed space. The advantage of this method is that the excessive element Se allows for fabrication of superior crystalline quality of CIGS films. Selenization was accomplished by a one-step fast heating stage and a constant temperature process, the thickness of CIGS films is about 1.85 μm. In order to investigate the effects of selenization temperature and time, the selenization processes were carried out by various temperature profiles. The time of constant temperature was applied from 20 min to 40 min, the selenization temperature from 500 °C to 550 °C was used, the decreasing rate of temperature after selenization is about 6 °C/min.

The processes completing the solar cells after CIGS selenization consist of sequential deposition of a 50 nm thickness CdS film by chemical bath deposition (CBD) from an aqueous solution containing 0.015 M $CdSO_4$, 1.5 M NH_4OH, and 0.75 M thiourea. Two transparent-conducting layers of 50 nm undoped ZnO and 700 nm doped ZnO with Al_2O_3 were deposited by RF magnetic sputtering. Followed by finally, a Ni–Al metal grid was deposited through an aperture mask. A total cell area of 1.2 cm^2 was defined by mechanical scribing. I/V curves of the cells were measured under standard AM1.5 and 100 mW/cm^2 illumination at 25 °C, and there are not MgF_2 films as an antireflective coating on our solar cells surface.

The surface morphologies and crystalline phases were examined by scanning electron microscopy (SEM) and X-ray diffraction (XRD), respectively. The gallium content nearing surface of CIGS thin films was determined by secondary ion mass spectrometry (SIMS). The composition of CIGS films was confirmed by X-ray fluorescence (XRF) measurements.

3. Results and discussion

The ultimate aim of this study was the development of a relatively simple and reproducible growth technique for the production of device-quality $Cu(In,Ga)Se_2$ thin films. So the attention was focused mainly on the optimization of technical parameters during the precursor's formation as well as the selenization parameters during the reaction stage.

Fig. 1 shows XRD patterns of CIGS thin films obtained from various sellenization temperature and time. It can be seen that all the films have the basic chalcopyrite crystal structure and very similar XRD pattern, diffraction peak 112 was the strongest, only single-phase chalcopyrite $CuIn_{0.75}Ga_{0.25}Se_2$ was detected. However, there is a noticeable change

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