

Microstructural and optical properties of CuIn_3Te_5 thin films for solar cells

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ABSTRACT

CuIn_3Te_5 thin films were deposited onto soda-lime glass (SLG) substrates by using a molecular beam epitaxy (MBE) system. Microstructural properties were examined by means of scanning electron microscopy (SEM), X-ray diffraction (XRD) and Raman scattering. The substrate temperature (T_{sub}) dependence of film properties was investigated in the range of 100–400 °C. Large-grain CuIn_3Te_5 thin films with a defect chalcopyrite structure were dominantly grown at 200–400 °C. Preferred (1 1 2) orientation was observed even at the low substrate temperature of 200 °C. The optical bandgaps of these films were determined to be 1.01–1.05 eV from transmission and reflection measurements. The best solar cell fabricated using CuIn_3Te_5 thin films grown at 200 °C yielded an efficiency of 4.64% with $V_{\text{oc}}=318$ mV, $J_{\text{sc}}=29.0$ mA/cm² and FF=0.504.

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1. Introduction

Cu-III-VI₂ chalcopyrite semiconductors are feasible candidates for application as photovoltaic cells [1], photodetectors (PDs) [2] and light-emitting diodes (LEDs) [3]. Among Cu-based chalcopyrites, a high efficiency of 19.9% has been achieved for Cu(In,Ga)Se₂ (CIGS) based thin film solar cells with small areas (< 1 cm²) [4]. However, no reports on CuInTe_2 (CIT) thin film solar cells have been reported so far, nevertheless this material has a direct energy bandgap in the range of 0.91–1.0 eV at approximately 300 K [5,6]. The energy bandgap of CuInTe_2 thin film is slightly narrower in comparison to that of CuInSe_2 (CIS) thin film (1.04 eV) [7]. The narrow bandgap absorber ($E_g < 1$ eV) is essential in order to make use of bottom cells for multi-junction (tandem) solar cells.

Several deposition methods for CuInTe_2 thin films have been reported so far, including a single-source evaporation [5,8,9], post annealing of Cu/In/Te stacked layers [6] and electrodeposition of Cu–In–Te based precursor layers [10].

However, there have thus far been no reports on the deposition and characterization of Cu-deficient CIT thin films such as CuIn_3Te_5 and CuIn_5Te_8 with defect chalcopyrite structures, known as ordered defect compounds (ODCs) or ordered vacancy compounds (OVCs), regarding their application as solar cells. In fact, telluride films have received comparatively little attention, with the exception of CdTe thin films, which have a suitable bandgap ($E_g=1.5$ eV) for solar cells [11,12].

In the present work, co-evaporation with three elemental sources was performed in order to study the growth of CuIn_3Te_5 thin films by means of a molecular beam epitaxy (MBE) system. The substrate temperature (T_{sub}) dependence of the films was then investigated from the viewpoint of the both microstructural and optical properties. The fabrication and performance of CuIn_3Te_5 solar cells are also described in detail.

2. Experimental

CuIn_3Te_5 thin films were deposited onto soda-lime glass (SLG) substrates by one-stage co-evaporation using a conventional MBE system (EV100, Eiko). The deposition chamber was pumped down using a diffusion pump with a liquid-N₂ trap to base pressure below 4×10^{-6} Pa. High purity 6 N (99.9999%)-metals (Cu, In) and 4 N (99.99%)-Te ingots were used as elemental sources. The Te ingots included a very small quantity of Se (approximately 3 ppm) as an impurity. All sources were simultaneously evaporated from p-BN crucibles in the separate effusion cells. All fluxes were kept constant during the deposition. The typical flux ratio of Te/(Cu+In) was approximately 8–10, as determined by a beam flux monitor. The substrate temperature (T_{sub}) was varied from 100 to 400 °C and was measured by a thermocouple attached to the backside of the SLG substrates. The deposition time was the same for all CuIn_3Te_5 thin films (20 min). The typical deposition rates of the CuIn_3Te_5 films were 1.1–1.4 nm/s at substrate temperatures above 200 °C. The surface microstructure of the films was investigated by scanning electron microscopy (SEM, S4800, Hitachi). The crystalline quality and orientation were characterized by X-ray diffraction (XRD, RINT2000, Rigaku) using

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Cu-K α radiation in theta-2theta scan-mode. Raman scattering measurements were performed at room temperature in a back-scattering configuration (InVia Reflex, Renishaw) using the unpolarized green line of an Nd:YVO₄ laser ($\lambda=532$ nm) as a light excitation source. Using a 50 \times objective lens, the laser spot size was found to be approximately 2 μ m in diameter. The average chemical composition of the films was determined by inductively coupled plasma emission spectroscopy (ICP, SPS7700, SII). The optical bandgaps of the films were determined from the transmission and reflection measurements using a double-beam spectrophotometer (UV-3101PC, Shimadzu).

The CuIn₃Te₅ thin film solar cells with ZnO:Al(0.5 μ m)/ZnO(0.1 μ m)/CdS(80 nm)/CuIn₃Te₅(1.5 μ m)/Mo(1.0 μ m)/SLG structure were fabricated. Al front electrodes were deposited onto ZnO:Al layers and then annealed in air at 200 $^{\circ}$ C for 5 min. Current–voltage (J–V) characteristics of the cells were measured at 25 $^{\circ}$ C under AM1.5, 100 mW/cm² illumination after light soaking with the same illumination conditions for 5 min. The light source of solar simulator (YSS-80 A, Yamashita-denso) was calibrated by a standard Si solar cell.

3. Results and discussion

3.1. Microstructural properties

The surface morphologies of CuIn₃Te₅ thin films deposited onto SLG substrates at various temperatures are shown in Fig. 1. A smooth surface with small grains was obtained at a substrate temperature of 100 $^{\circ}$ C. The lateral grain size of the films increased drastically at 200 $^{\circ}$ C, and many of these grains with sharp edges grew on the film surface. Therefore, it is suggested that the crystallization of CuIn₃Te₅ thin films was promoted even at a low temperature of 200 $^{\circ}$ C. At the moderate temperature of 300–400 $^{\circ}$ C, the surface roughness of the films was found to increase significantly because of the growth of large grains. The films exhibited a densely packed surface, which included faceted triangular grains. It was confirmed that the lateral grain size of the films tend to increase monotonically at higher substrate

temperatures. However, it has been revealed that the surface morphologies of CuIn₃Te₅ thin films are quite different from those of co-evaporated CuInSe₂ thin films [13,14]. In fact, it has been reported that the crystallites of CuInTe₂ thin films are larger and more faceted than CuInSe₂ thin films under the same growth conditions [14]. The well-developed morphological feature of CuIn₃Te₅ thin films may be caused by a migration of Te atoms (or molecules). It contrasts well with the vapor flux of Se, which contains mainly Se₅, Se₆ and Se₇ clusters with periodic structures such as chains and/or cycles at a crucible temperature of 200 $^{\circ}$ C, since Te flux contains mainly Te₂ and a small amount of Te₅ clusters without the periodic structures at a crucible temperature of 400 $^{\circ}$ C [15].

Fig. 2 shows XRD patterns of CuIn₃Te₅ thin films deposited onto SLG substrates at various substrate temperatures. Elemental In and numerous Te lines (JCPDS#85-0554) were observed even at 100 $^{\circ}$ C. It is notable that the (1 1 2) and (3 3 2) lines of the defect (ordered vacancy) chalcopyrite CuIn₃Te₅ structure [16] were confirmed at 100 $^{\circ}$ C. As excess Te atoms began to re-evaporate above 150 $^{\circ}$ C in this study, there was no Te phase in the films deposited at 200 $^{\circ}$ C. At this temperature, typical lines of (1 1 2), (2 2 0), (3 1 2), (3 3 2) and (4 2 4) of the defect chalcopyrite CuIn₃Te₅ were observed clearly. Therefore, the promotion of CuIn₃Te₅-film crystallization is straightforward at lower temperatures compared to CuInSe₂ films. This notable feature of the films can relate to the physical properties of a relatively low epitaxial temperature (620 K) [9] and melting point (789 $^{\circ}$ C) [17] for CuInTe₂. The peculiar lines of (1 1 0), (2 0 2) and (2 1 2) of the defect chalcopyrite CuIn₃Te₅ were observed above 300 $^{\circ}$ C. The defect chalcopyrite phase of CuIn₅Te₈ (JCPDS#24-0362) was not detected at all substrate temperatures. Concerning the film orientation, preferred (1 1 2)-oriented CuIn₃Te₅ thin films with high (1 1 2)/(2 2 0) ratios above 3–4 were obtained at 200–400 $^{\circ}$ C.

Raman spectra of CuIn₃Te₅ thin films shown in Fig. 2 are illustrated in Fig. 3. As a tetragonal chalcopyrite structure (space group I4̄2d) has eight atoms of VI-group elements such as Se and Te in one unit cell, 24 vibration modes, which consist of 21 optical modes and 3 acoustic modes, were present in total [18]. It is well known that 13 vibration modes ($A_1+2A_2+3B_1+3B_2+6E$) can be observed at the Γ point by Raman analysis, in which the A₂ mode

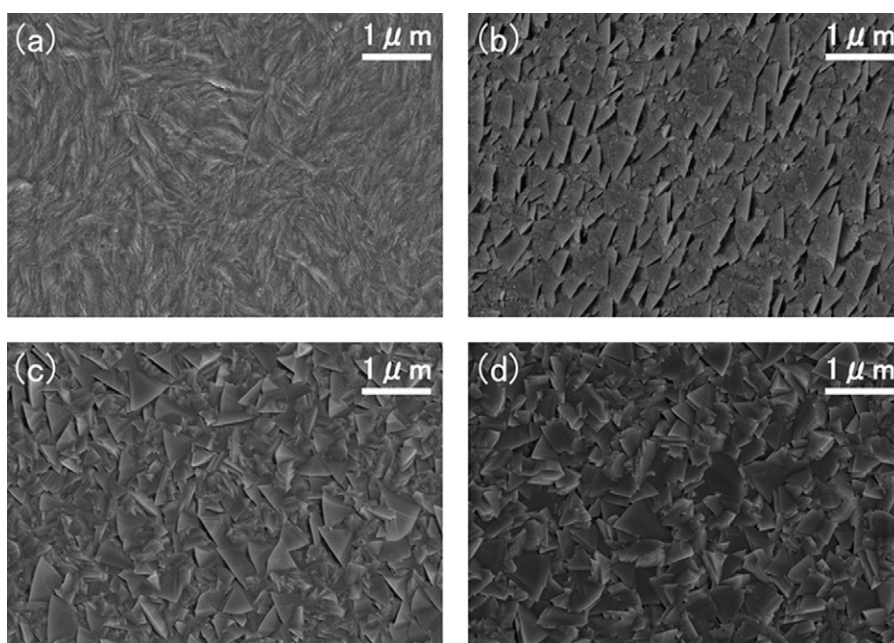


Fig. 1. SEM micrographs of CuIn₃Te₅ thin films (Cu/In=0.23–0.26) deposited onto SLG substrates at substrate temperatures (T_{sub}) of (a) 100 $^{\circ}$ C, (b) 200 $^{\circ}$ C, (c) 300 $^{\circ}$ C and (d) 400 $^{\circ}$ C.

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