



Influence of growth temperature of transparent conducting oxide layer on Cu(In,Ga)Se₂ thin-film solar cells

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ARTICLE INFO

Article history:

Received 16 February 2011

Received in revised form 22 August 2011

Accepted 24 August 2011

Available online 31 August 2011

Keywords:

CIGS solar cell

ITO thin film

Growth temperature

Series resistance

Cd diffusion

ABSTRACT

We have studied the influence of growth temperature (T_G) in the deposition of an indium tin oxide (ITO) transparent conducting oxide layer on Cu(In,Ga)Se₂ (CIGS) thin-film solar cells. The ITO films were deposited on i-ZnO/glass and i-ZnO/CdS/CIGS/Mo/glass substrates using radio-frequency magnetron sputtering at various T_G up to 350 °C. Both the resistivity of ITO and the interface quality of CdS/CIGS strongly depend on T_G . For a $T_G \leq 200$ °C, a reduction in the series resistance enhanced the solar cell performance, while the p–n interface of the device was found to become deteriorated severely at $T_G > 200$ °C. CIGS solar cells with ITO deposited at $T_G = 200$ °C showed the best performance in terms of efficiency.

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1. Introduction

The Cu(In,Ga)Se₂ (CIGS) thin-film solar cell is one of the promising candidates for second-generation photovoltaic devices due to its high performance and the possibility of low-cost commercial production. Recently, an efficiency of 20.1% has been reported from a small-area CIGS cell [1]. Thus far, CIGS solar cells have been extensively studied [2–4] not only to achieve a higher efficiency of over 20%, which is approaching the theoretical limit, but also to develop various applications [5–7]. To achieve these goals, it is prerequisite to understand the physical mechanism of the core CIGS absorber [8,9] as well as the role of other layers such as the transparent conducting oxide (TCO) window layer [10,11]. Until now, however, few studies have reported on the influence of growth conditions of TCO on the performance of CIGS solar cells.

Although ZnO:Al is widely used as a window layer of commercial devices, indium tin oxide (ITO) is known to be the best window layer for a CIGS solar cell [12,13] due to its excellent physical properties, including a wide band-gap (3.5–4.3 eV), high electrical conductivity (2500–5000 $\Omega^{-1} \text{cm}^{-1}$) [14], and good stability in high temperature and humidity conditions [15]. ITO films are typically prepared using radio-frequency (RF) magnetron sputtering [16,17]. Since the substrate temperature during sputtering influences the film growth of the TCO layer, one should be careful to manage the temperature

when obtaining both high conductance and optical transparency, which are related with film quality [18,19]. On the other hand, there are reports that the post-annealing of fully-fabricated CIGS solar cells at high temperature also causes permanent device failure [20,21].

In this paper, we report the relationship of ITO films grown on i-ZnO/CdS/CIGS at various growth temperatures (T_G) with the device performance of CIGS solar cells. A complete analysis is presented for the behavior of solar cells as a function of temperature.

2. Experimental

We prepared CIGS thin-film solar cells using a cluster-type deposition system that consists of load-lock, DC/RF sputtering, co-evaporation, and central transfer modules. A substrate (100 × 100 mm² max.) is first placed into the load-lock chamber. The central transfer module brings the specimen to the desired location for deposition: DC sputtering for a molybdenum back contact layer; RF dielectric sputtering for a front contact or TCO layer of i-ZnO, ZnO:Al, and ITO materials; and co-evaporation of Cu, In, Ga, and Se for an absorption layer. The base pressure of all the process chambers was maintained at $< 6.7 \times 10^{-5}$ Pa. The substrate temperature was adjusted up to 300 °C and ~600 °C for the DC/RF sputtering and evaporation chambers, respectively.

First, a 900 nm-Mo back contact layer was deposited on a soda lime glass substrate with an area of 25 mm × 25 mm at ambient temperature using a DC sputtering system. Subsequently, CIGS was grown through a three-stage process [22] in which a sequence of In-Ga-Se/Cu-Se/In-Ga-Se co-evaporation was performed at substrate

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temperatures of 330 °C (1st stage) and 570 °C (2nd and 3rd stages). The thickness of the CIGS layer was typically $\sim 2.3 \mu\text{m}$. As the buffer layer, CdS with a thickness of $\sim 50 \text{ nm}$ was deposited using chemical bath deposition in an alkaline aqueous solution consisting of cadmium salt (0.015 M), ammonia (15 M), and thiourea (0.75 M). Intrinsic (i-)ZnO (70 nm) and ITO layers were then deposited successively using RF sputtering in Ar/O₂ and pure Ar gas, respectively. In i-ZnO deposition, the substrate temperature, RF power, Ar/O₂ flow, and operating pressure were ambient temperature, 400 W, 30/10 sccm, and 4.0 Pa, respectively. Finally, we completed the CIGS devices with Au/Ni grid contact using thermal evaporation. Six solar cells with an active area of 0.47 cm^2 were fabricated in each substrate with an area of $25 \text{ mm} \times 25 \text{ mm}$.

To prepare the ITO films, we used a sintered ceramic target of 99.99% In₂O₃:SnO₂ = 90:10 wt.%, and a diameter of 10.1 cm. The target-to-substrate distance was 20 cm. The sputter deposition was done under the conditions of RF power, Ar flow, and an operating pressure of 100 W, 50 sccm, and 0.67 Pa, respectively. 150 nm-thick ITO films were deposited at various values of T_G ranging from ambient temperature to 350 °C.

The current–voltage (I – V) characteristics of the solar cell devices were measured via an I – V source meter (Keithley 2400) under a global AM 1.5 spectrum for 1000 W/m^2 irradiance at room temperature. The electrical and optical properties of ITO/i-ZnO were measured by an Accent HL5500 hall system and HITACHI U-4001 ultraviolet/visible/near-infrared spectrophotometer, respectively. The structural property of ITO/i-ZnO was measured using an X-ray diffraction (XRD, RIGAKU RU-200BH) method with Cu-K α X-ray source ($\lambda = 1.541 \text{ \AA}$) at 40 kV accelerating voltage and 60 mA current. A secondary ion-mass spectrometer (SIMS, CAMECA IMS 6f) analysis of the CIGS cells was carried out using O²⁺ primary ion bombardment included an 100 nA primary current rastered over a $250 \mu\text{m} \times 250 \mu\text{m}$ area with 7.5 keV impact energy. A cross-sectional image of the CIGS solar cell was measured using scanning electron microscopy (SEM, FEI SIRION 400) at an accelerating voltage of 10 kV.

3. Results

3.1. CIGS device characterization

CIGS solar cells of an ITO/i-ZnO/CdS/CIGS/Mo/glass structure were fabricated at different values of T_G for ITO thin films. Fig. 1(a)–(d) shows the relationship between the solar cell parameters and T_G . The solid and dashed lines represent the highest and average values of each data set, respectively. As shown in Fig. 1(a), the efficiency (η) of the CIGS solar cells reveals a strong dependence on T_G , and the highest efficiency of 15.24% was achieved at $T_G = 200 \text{ }^\circ\text{C}$. While η increases with T_G , it drops rapidly above $T_G = 200 \text{ }^\circ\text{C}$. On the other hand, the open circuit voltage (V_{oc}) and short circuit current (J_{sc}) are nearly constant below $T_G = 200 \text{ }^\circ\text{C}$. For $T_G > 200 \text{ }^\circ\text{C}$, both parameters are deteriorated as in η . The fill factor (FF) shows a similar behavior with T_G as in η .

3.2. ITO characterization vs. growth temperature

Fig. 2 shows the resistivity (ρ), mobility (μ), and carrier concentration (N) of the ITO/i-ZnO/glass samples as a function of T_G . The resistivity, ρ , exhibited a monotonic decrease with T_G , and was leveled off at $2.1 \times 10^{-4} \Omega \text{ cm}$. This shows roughly a relationship of $\rho \propto \exp(1/T_G)$, where T_G is in Kelvin. This behavior results from the simultaneous enhancement of N and μ as related in $\rho = 1/N\mu q$. It is believed that as T_G increases, the Sn ions substituted into the lattice of In₂O₃ become dense and release more electrons to increase N [19]. On the other hand, the mobility increases to $35 \text{ cm}^2/\text{V}\cdot\text{s}$ at $T_G = 200 \text{ }^\circ\text{C}$ and decreases by 23% beyond this temperature. As reported in the literature [19], the grain boundary electron scattering and ionized impurity

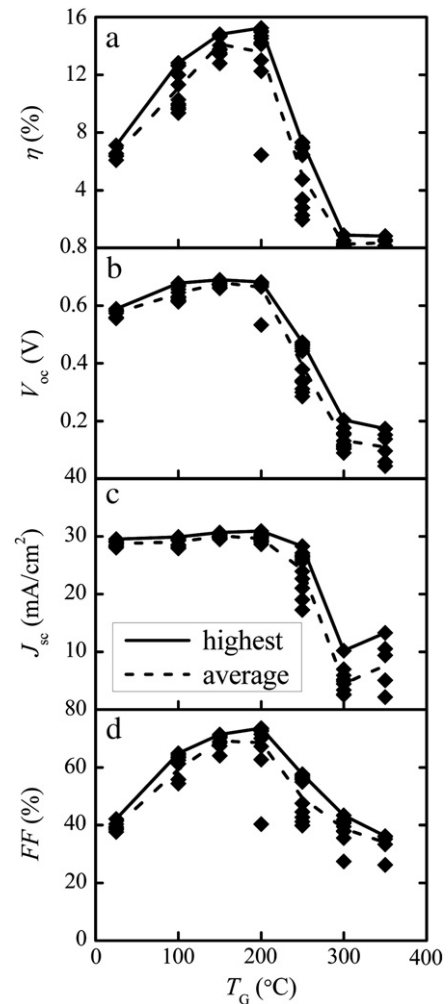


Fig. 1. Solar cell performance parameters vs. the growth temperature, T_G , of ITO films: (a) efficiency (η), (b) open circuit voltage (V_{oc}), (c) short circuit current (J_{sc}), and (d) fill factor (FF). The solid and dashed lines indicate the highest and average values of each cell, respectively.

scattering were believed to be dominant mechanisms to decrease the mobility at the entire range of growth temperature.

Fig. 3 shows the XRD spectra of i-ZnO/ITO films grown on the soda lime glass substrates as a function of T_G . The XRD patterns of ITO films showed strong (222) diffraction peaks which indicates a polycrystalline structure with a (222) preferential orientation. Unlike in a previous work [19], however, we observed no distinguishable improvement of

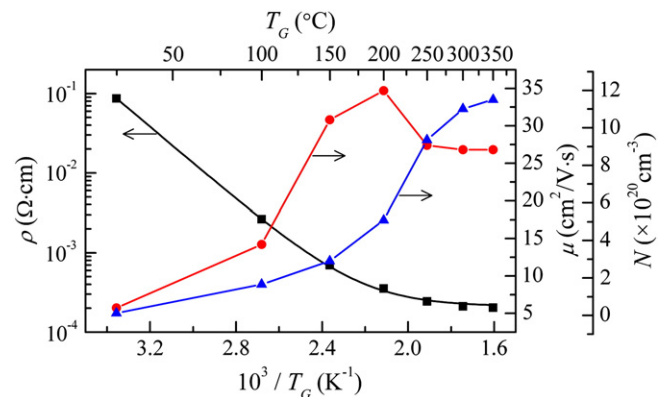


Fig. 2. The resistivity (\blacksquare), mobility (\bullet), and carrier concentration (\blacktriangle) of ITO on i-ZnO/glass substrates vs. $10^3/T_G$ (bottom axis) and T_G (top axis).

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