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Optimization of DMSO-based precursor solution by H_2O additive for performance enhancement of kesterite photovoltaic devices

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ABSTRACT

Dimethyl sulfoxide (DMSO) based solution has been proven promising in preparing high-quality $Cu_2ZnSn(S,Se)_4$ (CZTSSe) absorbers. In the present work, H_2O is used as a simple and effective additive to optimize the film formation of DMSO based CZTS precursor. It is demonstrated that a small amount of H_2O introduction into the DMSO solution reduced the oxygen content in the CZTS precursor film, and thus limited the formation of SeO_2/$Sn(O,Se)_x$ secondary phase in the final CZTSSe layer. As a result of improved morphology and carrier collection ability of absorber layer, the power conversion efficiency (PCE) is enhanced significantly from 7.45% for CZTSSe solar cell device prepared with pure DMSO precursor solution to 9.30% for that with an optimized DMSO/H_2O ratio. Applying the same preparing strategy to the Ag containing CZTSSe device, we finally achieved a PCE of 10.3% under 1-sun illumination.

1. Introduction

$Cu_2ZnSn(S,Se)_4$ (CZTSSe) related materials have drawn intense research attention due to their excellent light-absorbing abilities and earth-abundant constituent elements [1–4]. Up to now, both vacuum- and non-vacuum deposition methods have been successfully utilized to obtain CZTSSe solar cell devices with power conversion efficiency (PCE) higher than 10% [5–10]. The current record efficiency of 12.6% was obtained via the hydrazine-based solution processing [5], which is not suitable for large-scale production due to safety concerns. To avoid using toxic solvent, dimethyl-sulfoxide (DMSO)-based processing stands out as a promising deposition route from the molecular-based precursor solution. Using DMSO precursors, efficient solar cell with up to 11.8% efficiency was achieved recently on an active device area of 0.10 cm^2 [11]. However, numerous grain boundaries, secondary phases, and the presence of microscopic voids are commonly observed in sintered absorber layers and limit further improvement of device performance [12]. Various secondary phases, including $Cu_x(S,Se)$, Zn(S,Se), $Sn(S,Se)_x$ or $Cu_2Sn(S,Se)_3$, have been reported detrimental to the performance of solar cells, especially the open circuit voltage (V_{OC}) and fill factor (FF) [13–19]. Oxides such as SeO_2, which is likely formed during the air annealing of CZTSSe, have also been observed on CIGSSe and CZTSSe surfaces [20–22], although their effects on the device performance are not clear yet. To address the above-mentioned issues, a three-stage annealing process was utilized during recrystallization stage to dramatically improve the film crystallinity, which results in the 11.2% efficiency and a low V_{OC} deficit of 0.57 V [23]. On the other hand, since the reaction medium plays a crucial role in the solution preparation of CZTSSe layer [24], attempts to control the generation of second phase and defects during grain growth were performed through optimizing precursor properties [25]. F. Aslan et al. found that the pH value of precursor solution can affect the physical properties of Cu_2SnZnS_4 (CZTS) thin film [26]. Acetone was introduced into the DMSO precursor solution to lower the drying temperature and improve the wetting behavior on substrates [27]. It is also reported that adding precursors into the DMSO solvent in a step-by-step order to get the so-called redox-equilibrated precursor solution effectively leads to better quality CZTS, and yields the higher efficiency of 8.32% [24].

In this work, H_2O was introduced as one of the most simple and common additives into the DMSO-based precursor solution to optimize the film formation of CZTSSe. It is demonstrated that a small amount of H_2O effectively improves the morphology, crystallinity, composition distribution and electrical properties of the CZTSSe absorber. SeO_2 secondary phase on the surface of CZTSSe film is also dispelled upon the addition of H_2O in the precursor solution, thereby promoting the efficiency of CZTSSe thin film solar cells from 7.45% to 9.30% with the

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optimum amount of H_2O introduction. A best conversion efficiency of 10.3% is finally achieved as the approach is applied in the processing of $(Ag_xCu_{1-x})_2ZnSn(S,Se)_4$ (ACZTSSe) solar cell.

2. Experimental

2.1. Thin-film preparation

Mo-coated soda lime glass substrates were slightly oxidized before depositing absorber layer to avoid over selenization of the molybdenum electrode during the selenization process [28]. Polycrystalline CZTSSe thin films were prepared by spin coating DMSO-based precursor solutions onto substrates in nitrogen-filled glove box ($[O_2] < 1$ ppm). Briefly, CZTS precursor solution was prepared by dissolving Cu $(CH_3COO)_2{\cdot}H_2O$ (52.5 mmol), $Zn(CH_3COO)_2{\cdot}2H_2O$ (33 mmol), $SnCl_2$ (30 mmol) and CH_4N_2S (231 mmol) into 10 ml dimethyl sulfoxide and stirring at room temperature for 3 h to get a clear yellow solution. H_2O with varying amount from 0 to 3 ml was added into 10 ml precursor solution. The targeting Cu-poor and Zn-rich compositions with Cu/(Zn + Sn) = 0.83 and Zn/Sn = 1.1 in the precursor solution were maintained for all sample. For the preparation of ACZTS thin film, an optimized amount of $AgNO_3$ was added into the CZTS precursor solution to realize the Ag/(Cu + Ag) ratio of 0.16 in the absorber as following our previously reported synthesis route [29]. The obtained precursor films were loaded into a graphite susceptor together with 0.32 g selenium powder and nominally sealed with a susceptor lid. The selenization was accomplished in a rapid thermal processing (RTP) furnace at 530 °C for 10 min duration under a constant N_2 flow. More detailed precursor solution preparation and coating procedure have been described elsewhere [29].

2.2. Solar cell device fabrication

To complete the device, ~ 30 nm CdS buffer layer was deposited onto CZTSSe film by a standard chemical bath deposition, followed by RF magnetron sputtering of ~ 30 nm i-ZnO window layer and DC magnetron sputtering of ~ 200 nm ITO top transparent contact layer. Aluminum grids were deposited by thermal evaporation for front contacting. The device area of ~ 0.11 cm^2 was defined by mechanical scribing. No antireflection coating was used.

2.3. Characterization

X-ray diffraction (XRD) measurement of CZTSSe films was performed using a PANalytical EMPYREAN X-ray diffractometer with Cu Kα radiation. HORIBA Jobin-YvonLabRAM H800 was utilized to record Raman spectra at 532 nm excitation wavelength. Sample mophology was observed through a JSM-6701F Field-emission scanning electron microscopy (FE-SEM), and energy dispersive spectroscopy (EDS) data was collected through an IE 300 X energy-dispersive X-ray spectroscopy detector. The optical absorption spectra were obtained using a Shimadzu UV-2600 ultraviolet-visible (UV–vis) spectrometer equipped with an integrating sphere detector. A Physical Electronics PHI-5702 spectrometer was applied to record the X-ray photoelectron spectrum (XPS) with an Mg K_α X-ray source (250 W, pass energy of 29.35 eV). All of the binding energies were calibrated by the C 1s peak at 284.6 eV. Under a calibrated AM 1.5G illumination generated by an AAA-class solar simulator (Oriel Sol3A 94023A, from Newport), current-voltage (I-V) curves were recorded on a Keithley 4200-SCS semiconductor measurement system. The external quantum efficiency (EQE) was measured using a 150 W xenon lamp (Oriel) with a Cornerstone 74004 monochromator. The capacitance-voltage (C-V) and drive level capacitance profiling (DLCP) characterizations were also conducted using a

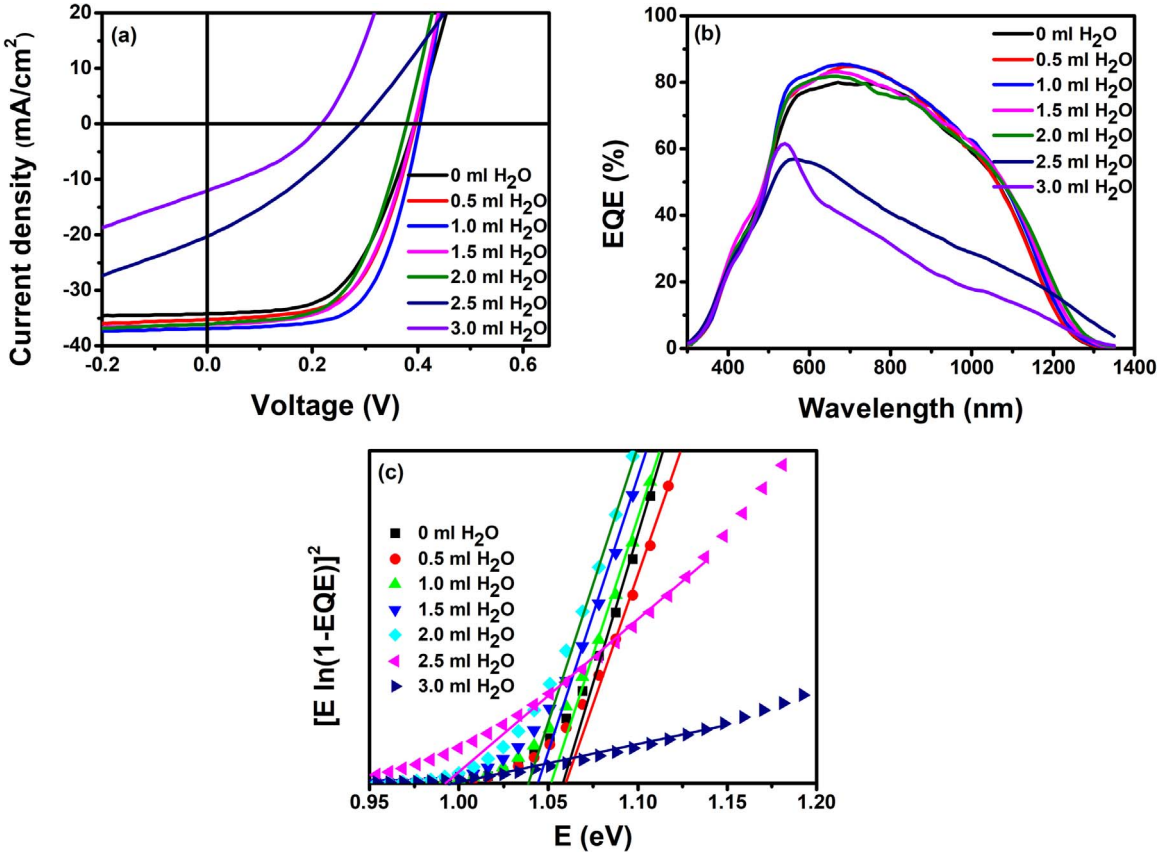

Fig. 1. (a) Current–voltage characteristics of CZTSSe solar cells with different H_2O contents in precursor solution. (b) External quantum efficiency spectra of corresponding devices. (c) Determination of Eg of these CZTSSe absorbers based on external quantum efficiency response.

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