



Flexible kesterite $\text{Cu}_2\text{ZnSnS}_4$ solar cells with sodium-doped molybdenum back contacts on stainless steel substrates

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ABSTRACT

In this work we report the Na incorporation from Na-doped Mo (Mo-Na) back contact for kesterite $\text{Cu}_2\text{ZnSnS}_4$ solar cells on flexible stainless steel substrates. It is demonstrated that Na can be effectively incorporated into CZTS by inserting Mo-Na layer at back contact. Direct contact of CZTS and Mo-Na layer leads to poor homogeneity and adhesion. The thickness of MoS_2 formed at the back contact depends on the presence of Na and whether Mo contacts with CZTS directly. Back contact configuration with a Mo capping layer on Mo-Na layer is found to be helpful to maintain the advantages of Mo back contact and control the thickness of MoS_2 interface. As a result, CZTS device fabricated on this configuration yields higher conversion efficiency of 6.2%. However, this efficiency is still far lower than that on traditional soda lime glass substrate which shows efficiency over 8%. The loss mechanism of device fabricated on stainless steel is investigated and analyzed according to the device performance and electrical parameters.

1. Introduction

Kesterite $\text{Cu}_2\text{ZnSnS}_4$ (CZTS) is a promising thin film photovoltaic absorber material that contains earth-abundant and non-toxic constituents and has a near-optimum direct band gap energy of about 1.5 eV and a high absorption coefficient ($> 10^4 \text{ cm}^{-1}$), which has been proposed as a potential alternative to traditional chalcopyrite $\text{Cu}(\text{In,Ga})\text{Se}_2$ (CIGSe). Very encouraging achievements have been obtained on CZTS solar cells based on classical rigid soda lime glass (SLG) substrates. To date, the confirmed highest energy conversion efficiencies of 11% for pure sulfide CZTS and 12.6% for Se containing CZTS_{Se} solar cells have been demonstrated by UNSW and IBM, respectively [1,2], which both show substantial commercial promise. Compared with solar cells on rigid substrates, flexible thin film solar cells own the charm of low cost, light weight, high specific power density and flexible nature of the modules. These unique advantages enable them to be applied to some novel fields like building integrated photovoltaics (BIPV), space power and automotive applications. Another merit of flexible PV technologies is their potential to be implemented in cost-effective roll-to-roll processes, making them extremely attractive from the industrial

prospect. Recently, many efforts have been focused on the development of flexible kesterite CZTS solar cells on a variety of suitable substrates such as polymers [3], stainless steel (SS) [4,5], aluminum foil [6], flexible glass [7], and molybdenum foil [8,9]. In order to achieve high efficiency CZTS solar cells, the alkali metal (s) is well recognized as a must-to-have dopant for CZTS absorber, which typically is sodium (Na). For traditional soda-lime glass substrates, Na is sourced from the soda lime glass itself where Na diffuses through the Mo back contact into the growing CZTS absorber during the high temperature process. For Na-free substrates, extrinsic Na supply is needed and different routes can be employed to dope Na into the CZTS absorbers, such as NaF evaporation [4] and Na-doped Mo back contact sputtering [5]. The later option has been well employed in $\text{Cu}(\text{In,Ga})\text{Se}_2$ solar cells [10,11] and reported to yield better device performance for $\text{Cu}_2\text{ZnSnS}_4$ solar cells [12]. This route, incorporating Na into the Mo back contact sourced from Na-containing Mo sputtering target (Mo-Na), could provide homogenous and controllable Na incorporation into absorber according to the previous experience in chalcopyrite CIGSe [11,13], which would be very convenient for industrial implementation since it can be easily integrated into back contact sputtering step.

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In this study, Na-containing Mo layer at back contact by sputtering method is employed as Na doping source for kesterite CZTS absorber on stainless steel substrates. Various Na-containing Mo back contact configurations are explored and the properties of overlying CZTS and performance of associated CZTS devices are compared. Finally, conversion efficiency of 6.2% has been obtained for flexible kesterite CZTS solar cells with a Mo-Na “sandwiched” back contact configuration.

2. Experimental section

Ferrite stainless steel (SS, 430) foils provided by Baosteel (China) were used as substrates. The substrates were cleaned with cleaning agent and rinsed with deionized water, followed by RCA-1 (RCA clean is a standard set of wafer cleaning steps developed by Werner Kern at RCA laboratories.) standard cleaning process [14] which is widely used in cleaning silicon wafer to remove organic and particles. After that, the substrates were rinsed with deionized water and dried with N_2 flux. 50 nm thick Ti layer deposited by DC magnetron sputtering (AJA International, Inc., model ATC-2200) with the sputtering power of 150 W under Ar pressure of 1.5 mTorr was used as the iron diffusion barrier between stainless steel and Mo back contact and to improve the adhesion of the Mo. Five different back contact configurations are investigated in this work (see Fig. 1). Both Mo-Na and Mo layers were deposited by DC-sputtering with power density of 5.44 W/cm^2 (deposition rate of 5.5 nm/min) and 4.35 W/cm^2 (deposition rate of 9.5 nm/min), respectively. Commercially available Mo-Na sputtering targets with Na concentrations of 10 at% was used and the Na was incorporated into the Mo target material in the form of sodium molybdate (Na_2MoO_4). Besides, Mo-deposited soda line glass (SLG) substrate also has been employed as reference. CZTS precursors were prepared by co-sputtering Cu (with direct current (DC) power supply at power density of 0.37 W/cm^2 and deposition rate of 1.80 nm/min), ZnS (with radio-frequency (RF) power supply at power density of 1.54 W/cm^2 and deposition rate of 3.53 nm/min) and SnS (with RF power supply at power density of 0.83 W/cm^2 and deposition rate of 3.74 nm/min) using the same sputtering system and working pressure as above, targeting thickness of 700–800 nm and a Cu-poor and Zn-rich composition of ($\text{Cu}/(\text{Zn} + \text{Sn}) = 0.8$, $\text{Zn}/\text{Sn} = 1.2$). The targets we used are 3 in. in diameter and the purities of the targets are all 4 N. The precursors were then subjected to sulfurization using Rapid Thermal Processor (AS-One 100) in sulfur & SnS atmosphere at 560°C . The sulfurization temperature profile is to ramp up to 560°C at 3°C/s and retain at 560°C for 5 min and cool down to room temperature naturally. Device fabrication was completed by sequentially depositing 60 nm CdS via chemical bath deposition (CBD), 60 nm i-ZnO and 200 nm ITO by RF and DC magnetron sputtering, respectively. Finally, aluminum grids were used as top contact by evaporation and final cells with areas of 0.24 cm^2 were defined by mechanical scribing.

The structural features of the fabricated CZTS absorbers were studied by X-ray diffraction (XRD, Rigaku D/max 2500) and Raman scatter spectroscopy (LabRAM HR800) with excitation wavelength of 514 nm at room temperature. The morphologies of the CZTS absorbers were

examined with a scanning electron microscope (SEM, FEI Nova NanoSEM 450). Compositional depth profile for both bulk and trace elements were acquired by time of flight secondary ion mass spectroscopy (TOF-SIMS, TOF-SIMS5 from ION-TOF GmbH, Germany). An Cs^+ ion gun operating at 1 keV, current of 116.8 nA and raster size of $300 \times 300 \mu\text{m}^2$ was used for sputtering. Elemental composition information was analyzed using a 15 keV pulsed Bi_3^+ primary analysis ion gun at an area of $100 \times 100 \mu\text{m}^2$. Cross-sectional transmission electron microscopy (TEM) specimens were prepared by a focused ion beam (FIB) system (the xT Nova NanoLab 200). Note that a $\sim 20 \text{ nm}$ Au layer and a $\sim 1 \mu\text{m}$ Pt layer were deposited consecutively on top of specimens before cutting process for the sake of preventing the ion damage. JEOL JEM-ARM200F (200 kV) aberration-corrected scanning transmission electron microscope (STEM) system was used for acquiring HAADF (high-angle annular dark-field) image.

The current density-voltage (J-V) curves of CZTS solar cells were measured using a solar simulator to provide simulated 1 sun AM 1.5G illumination, which was calibrated with a standard Si reference cell. External quantum efficiency (EQE) data were collected by a QEX10 spectral response system (PV measurements, Inc) calibrated by the National Institute of Standards and Technology (NIST)-certified reference Si and Ge photodiodes. This system uses monochromatic light chopped at 120 Hz.

3. Results and discussions

The appearance of the CZTS absorbers on five back contact configurations and on reference SLG was examined by optical microscope (see Supplementary Material, Fig. S1) equipped in Raman measurement system. For reference sample on SLG, the CZTS absorber has a homogeneous appearance. The CZTS absorbers on type I, III and V configurations also show homogeneous appearance, similar to the reference sample. For type II configuration, there are many black domains on the surface of CZTS absorber, which corresponds to a mixture of CZTS and Cu-S secondary phases confirmed by Raman measurement (see Fig. S2). In type IV configuration, the adhesion of the CZTS absorber is worse than the other samples, suffering from cracks and peeling-off issue, which cannot be used for further characterizations and device fabrication. Therefore, no data come from type IV samples will be presented in further discussion. Similar peeling-off issue has been reported for CIGSe deposited directly on Mo-Na [13], indicating direct contact of CZTS with Mo-Na would lead to poor adhesion and homogeneity. Fig. 2 displays the surface morphologies of the CZTS absorbers on type I, II, III and V configurations with reference CZTS on SLG for comparison. For type I configuration without Na addition and type II with CZTS contacting Mo-Na directly, the CZTS absorbers exhibit very rough and loose morphology, and the grains cannot be clearly distinguished, implying a poor crystallinity. CZTS absorbers on type III and V configurations show very compact and uniform morphology, demonstrating isolated grains with grain size over $1 \mu\text{m}$ and well-defined boundaries, which exhibits similar features with that of reference on SLG but relatively smaller grain size.

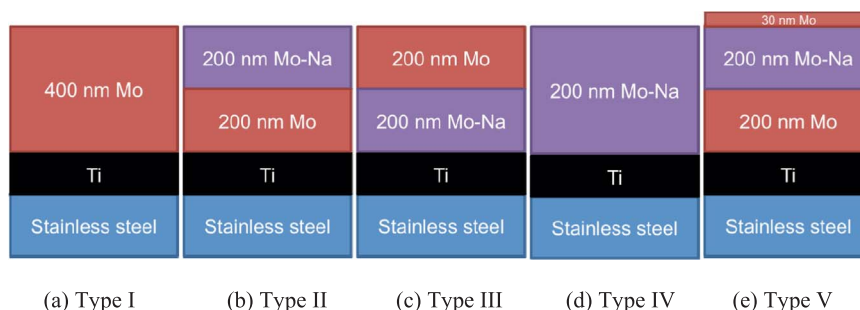


Fig. 1. Schema of five different back contact configurations studied in this work. The layers are not drawn to scale.

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