

Role of Mo:Na layer on the formation of MoSe₂ phase in Cu(In,Ga)Se₂ thin film solar cells

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ABSTRACT

This study investigated the influence of Mo:Na on the formation of MoSe₂ in Cu(In, Ga)Se₂ (CIGS) thin film solar cells and its overall effect on the performance of cells with a structure of Ti/Mo:Na/Mo/CIGS/CdS/i-ZnO/ZnOAl/Al. Varying the thickness of the Mo:Na layer enabled the systematic control of the diffusion of Na into the CIGS layer. Experimental results demonstrate that the thickness of MoSe₂ phase decreases with an increase in the thickness of the Mo:Na layer. Reducing the thickness of the MoSe₂ layer enhanced cell efficiency. Additionally, Ga distribution in the CIGS layer may vary with a change in the thickness of the Mo:Na layer. When Mo:Na/Mo=500/500 nm, the Ga content and the thickness of the MoSe₂ layer were close to optimal with regard to cell performance. This paper proposes a model to explain the impact of Na doping on the formation of MoSe₂ phase formation in polycrystalline CIGS. It appears that the addition of Na leads to the formation of Na₂Se_x at the grain boundaries in the absorber layer, which reduces the diffusion of Se to form MoSe₂.

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1. Introduction

CIGS thin film solar cells with the highest efficiency are those produced using a substrate of soda lime glass, which contains considerable quantities of sodium in the form of NaO₂. It has been shown that the presence of sodium during the growth of CIGS absorber layers is beneficial to cell performance, particularly with regard to open-circuit voltage (V_{oc}) and fill factor (FF) [1,2]. In addition, the cell performance of CIGS grown with Na is generally more homogeneous over a large range of Cu concentrations [3,4]. However, exceeding a particular threshold content of Na causes a drop in cell performance due to the small grain size and the porosity of the films [5,6]. Na is usually incorporated in the layers via diffusion from the soda-lime glass substrate; however, the diffusion process is difficult to control with any degree of precision. The application of Na-doped Mo as a source of sodium is another approach to the production of cells of large area, an approach that tends to result in a more uniform distribution of Na [7,8].

Selenization has been shown to result in the formation of a MoSe₂ layer at the CIGS/Mo interface. A specific quantity of MoSe₂ is required to ensure a good electrical contact with the back Mo contact layer [9,10]. Nonetheless, the formation of excessive MoSe₂ can lead to delamination of the film [11] with adverse effects on

the V_{oc} and FF of CIGS solar cells due to the high resistance of the MoSe₂ [12]. The production of high efficiency CIGS solar cells requires that the thickness of the MoSe₂ layer be reduced to below a particular range [13–15]. The thickness of the MoSe₂ layer can be influenced by residual stress in Mo contacts [16], selenization conditions [17,18], the inclusion of a barrier layer [13], sputtering pressure [19], sputtering power [15], and the effects of hydrogen-assisted solid Se vapor selenization [20]. However, the degree to which the Mo:Na layer (Na content) influences the MoSe₂ phase during selenization remains unclear. This study investigated how the Mo:Na layer influences the MoSe₂ phase with regard to the performance of the resulting CIGS thin film solar cells. We also propose a model to explain the role of Na doping on the formation of MoSe₂ in polycrystalline CIGS. Additionally, this study prepared Cu-Ga/In metal precursors with various thickness ratios to determine the device-quality stoichiometry requirements for the CIGS absorber layer.

2. Experimental details

The Mo:Na film was deposited by a Mo-5% Na target as the bottom layer of a bi-layer Mo back contact on Ti metal substrate of $1.5 \times 1 \times 1 \text{ cm}^3$. The used target has a Na atomic concentration of 5%, and the Na was added to the target as Na₂MoO₄ powder. Mo contacts were sputtered onto Mo:Na film to a thickness of 500 nm, at a work pressure of 0.533 Pa, and a power of 250 W, at a process

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Table 1

Composition and composition ratios of CIGS absorber layer following selenization at 798 K for 40 min. The Mo:Na/Mo thickness ratio was fixed at 500/500 nm and the Cu-Ga/In thickness ratios were varied.

Thickness ratios of Cu-Ga/ In layer (nm/nm)	Compositions (at%)				Composition ratios	
	Cu	Ga	Se	In	Cu/III	Ga/III
200/300	22.11	3.62	50.17	24.10	0.80	0.13
245/300	22.43	1.22	51.70	24.65	0.87	0.05
290/300	24.58	1.78	48.91	24.73	0.93	0.07
335/300	26.19	0.91	47.89	25.01	1.01	0.04
380/300	28.25	0.51	47.33	23.90	1.16	0.02

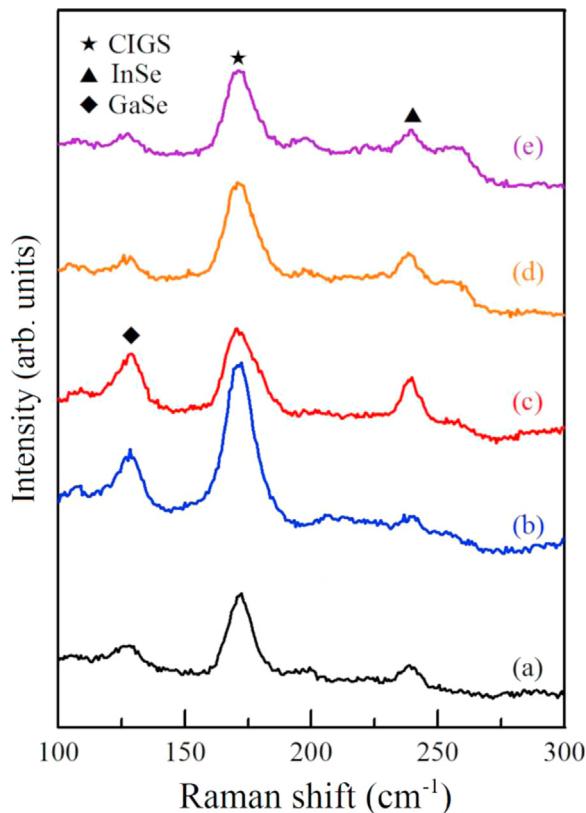


Fig. 1. Raman shifts obtained with Mo:Na/Mo thickness ratio of 500/500 nm after selenization at 798 K for 40 min using a Cu-Ga/In precursor with various thickness ratios: (a) 200/300 nm; (b) 245/300 nm; (c) 290/300 nm; (d) 335/300 nm; (e) 380/300 nm.

temperature of 373 K. Various Mo-5% Na films were prepared with thicknesses ranging from 300 nm to 700 nm with various thickness ratios under a working pressure of 0.533 Pa, and a power of 100 W, at a process temperature of 373 K. Bi-layer In/Cu_{0.7}Ga_{0.3} metal precursor films were deposited on Mo contacts beginning with elemental In, followed by Cu_{0.7}Ga_{0.3}, yielding a Cu-Ga-In film with a total thickness of approximately 600 nm [21]. The In metal precursor film was deposited using evaporation at a temperature of 1273 K and work pressure of 2.2×10^{-3} Pa. The Cu-Ga metal precursor film was prepared by sputtering at a power of 80 W and work pressure of 0.399 Pa. We then initiated a two-stage annealing process at 533 K for 40 min and a second-stage annealing at a higher temperature of 798 K or 848 K for 40 min. Annealing of the CIGS sample was conducted in a tube furnace in Ar (purity 99.995%) atmosphere using elemental Se. The CIGS absorber layer of thickness approximately 1.8 μm was obtained after selenization.

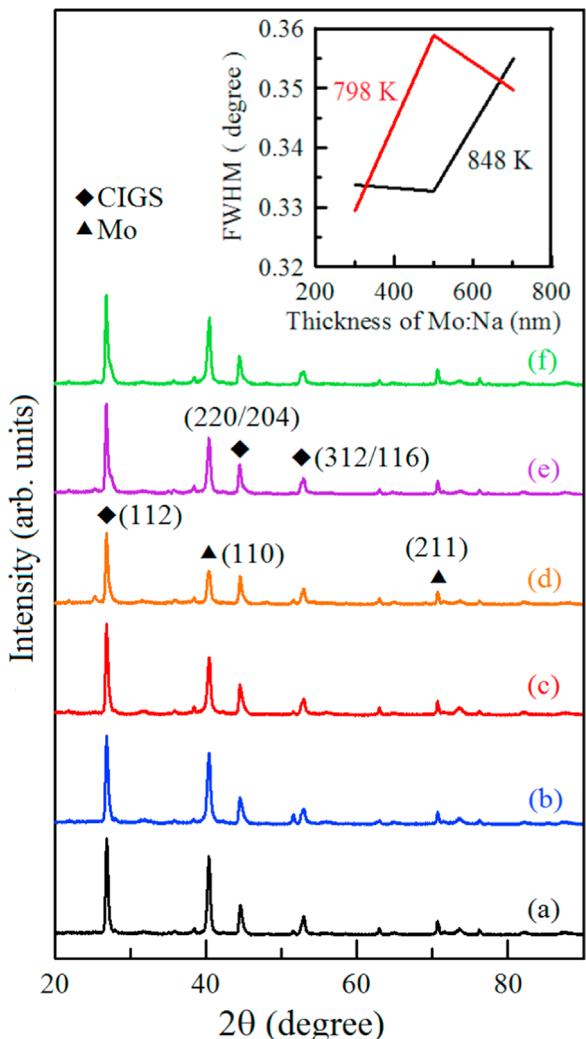


Fig. 2. XRD results obtained from samples with various Mo:Na/Mo thickness ratios that underwent selenization at various temperatures: (a) Mo:Na/Mo = 300/500 nm, at 798 K; (b) Mo:Na/Mo = 500/500 nm, at 798 K; (c) Mo:Na/Mo = 700/500 nm, at 798 K; (d) Mo:Na/Mo = 300/500 nm, at 848 K; (e) Mo:Na/Mo = 500/500 nm, at 848 K; (f) Mo:Na/Mo = 700/500 nm, at 848 K. The inset presents the FWHM of the (112) peaks obtained from CIGS associated with various Mo:Na/Mo thickness ratios and selenization temperatures.

A field-emission scanning electron microscope (FE-SEM, JEOL JSM-6700F) was used to observe the microstructure of films and measure the thickness of MoSe₂ films. X-ray diffraction (XRD, JEOL TF-SEM JSM7000F, CuKα, $\lambda = 1.54052$ Å) and a Raman analyzer (Ramboss 500i Micro-Raman System) were used to investigate the crystalline structure and phase transformation of the CIGS absorber layer using 532 nm excitation wavelength. Secondary ion mass spectroscopy (SIMS, IMS-6f) was used to investigate the depth profiles of the thin film elements. Extraction voltages were set at 10 and 12.5 keV, respectively. Additionally, the current of the O²⁺ ion was set to 80 and 120 nA, which impacted the surface of the samples with 5.5 and 8 kV of energy, respectively. Transmission electron microscopy (TEM, JEOL, JEM-ARM200FTH) was used in conjunction with energy dispersive X-ray analysis (EDAX) to compare the chemical composition of grains and grain boundaries in the absorber layer, with the aim of verifying the presence of Na₂Se_x. TEM sample preparation was performed using a dual-beam focused ion beam system (FEI Helios Nanolab 600i System).

Following CIGS absorber layer, we performed chemical bath deposition of CdS buffer layers. Highly resistive layers of intrinsic

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