

Performance Analysis of CuSbSe₂ Thin-Film Solar Cells with Cd-Free Window Layers

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ABSTARCT

This study investigates novel thin-film solar cells featuring CuSbSe₂ (CAsE) with ZnSnO and ZnMgO window in the layer superstrate structure. For glass/ITO/ZnMgO/CAsE/Cu+Au, the J-V measurements reveal a short-circuit current density (J_{sc}) of 19.4 mA/cm², an open-circuit voltage (V_{oc}) of 0.28 Volts, a fill factor (FF) of 39.14%, and a power conversion efficiency (η) of 2.13%. Similarly, glass/ITO/ZnSnO/CAsE/Cu+Au exhibits J_{sc} around 19.6 mA/cm², V_{oc} around 0.31 Volts, FF around 40%, and a power conversion efficiency (η) of 2.43%. This paper stands out as a pioneering contribution, introducing novel thin-film solar cells with a distinctive superstrate structure utilizing CuSbSe₂ (CAsE) in conjunction with ZnSnO and ZnMgO windows. The comprehensive study presents the first-ever characterization and performance evaluation of these innovative configurations, shedding light on their unique potential in advancing sustainable solar energy technology.

Keywords; *Thin Films, Physical vapour deposition, Energy storage and conversion*

INTRODUCTION

In recent years, the quest for sustainable energy sources has become a crucial aspect of scientific and technological pursuits. This drive stems from the necessity to combat air pollution caused by the extensive use of fossil fuels and to maintain ecological equilibrium. Among various renewable energy resources, solar energy stands out as a highly promising solution. Solar systems are increasingly proving to be cost-effective and environmentally sustainable, positioning themselves as a valuable long-term energy solution [1,2]. The conversion of solar irradiation into electricity through photovoltaic effects is a key aspect of solar power technology. Traditional multi-crystalline silicon (Si) photovoltaics, with an efficiency of around 26.7% [3], face challenges due to their indirect bandgap, resulting in reduced light absorption and the need for thicker silicon absorber layers, thus increasing overall costs [4]. To address this issue, thin-film technologies have been developed, relying on the direct bandgap of semiconductors like CuSbSe₂. CuSbSe₂ It is recognized for its exceptional absorptive properties, boasting a high absorption coefficient ($\alpha > 10^5 \text{ cm}^{-1}$) and a direct band gap of approximately 1.1 eV at short wavelengths [5]. These attributes make CuSbSe₂ a remarkable absorber material for photovoltaic devices, potentially achieving photoelectric conversion efficiencies exceeding 25% due to its advantageous band gap. By reducing the absorber layer thickness and benefiting from a high absorption coefficient, CuSbSe₂ solar cells can enhance carrier migration/diffusion and increase carrier collection efficiency. Moreover, the compositional elements of CuSbSe₂ (CAsE) are abundant, inexpensive, and non-toxic, meeting the criteria for the future development of photovoltaic devices. This underscores the ongoing exploration for materials that are Earth-abundant, cost-effective, and free from toxicity in the pursuit of sustainable energy solutions [6]. This study enhances the development of CAsE thin film solar cells by incorporating novel n-type window layers made of Sn-doped ZnO (ZnSnO) and Mg-doped (ZnMgO). Notably, these ecologically sustainable resources exhibit comparable performance to traditional CdS and pose no harm to humans and the environment. This signifies the initial incorporation of these materials into the substrate device architecture, a significant breakthrough in sustainable energy technology. The resulting solar cells consist of an n-type layer that does not contain Cadmium (Cd) and a p-type absorber layer that is abundant in Earth elements. The objective of this pioneering strategy is to develop ecologically sustainable and efficient solar energy solutions.

EXPERIMENTAL DETAILS

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Figure 1 shows the schematic for the two CAsE thin film solar cells that are fabricated using physical vapor deposition techniques; these cells have an n-type layer of ZnMgO and ZnSnO, respectively. Solar cells that have a ZnMgO layer are called *SC-1*, while those with a ZnSnO layer are called *SC-2*. The two cells are deposited on glass substrates that were coated with indium tin oxide (ITO). Using the magnetron sputtering technique, doped ZnO layers were deposited. Based on the optimization parameters from the previous study, the co-evaporation method is used to fabricate the CAsE layer, as the cell absorber layer [7]. The fabrication process ended with Cu/Au metal contacting process. (See the Supplementary file for the further information of about the deposition of each layer).

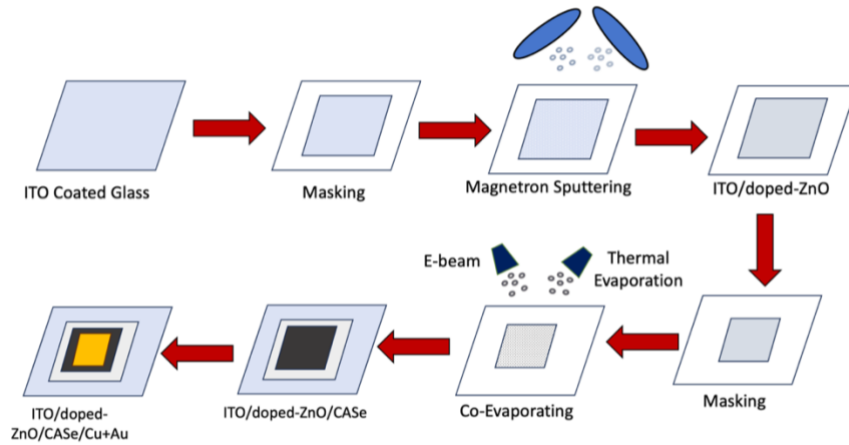


Figure-1: Fabrication Steps of ITO/doped-ZnO/CAsE/Cu+Au Thin Film Solar Cell

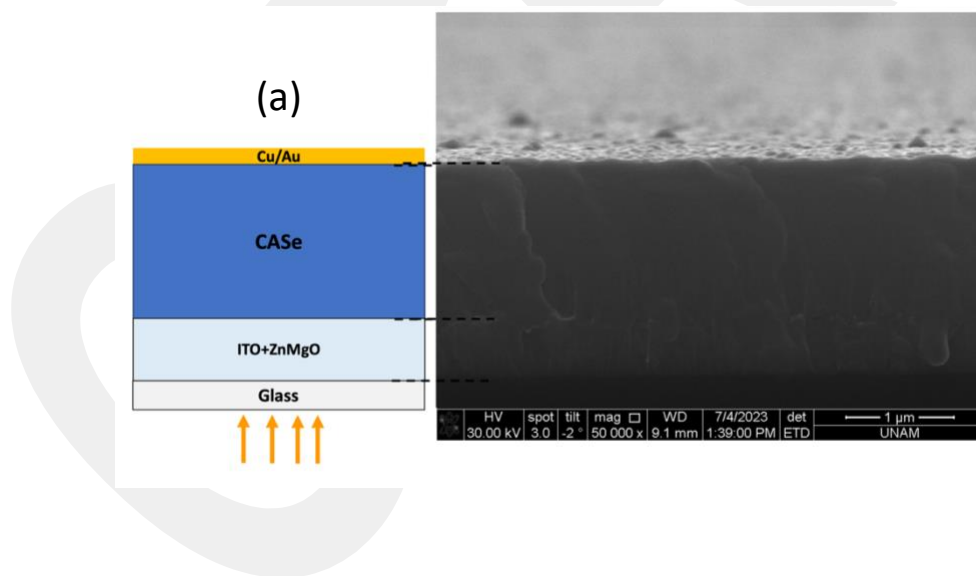
RESULTS AND DISCUSSIONS

The characterization of the layers in both solar cells were outlined in detail in the supplementary file. **CAsE thin film shows crystalline nature with the (013) orientation peak around 28.8° (See FigureS-1) [7].** ZnMgO thin films possess a crystalline structure, with the main alignment occurring along the (002) plane at an approximate angle of $2\theta=35^\circ$ (see Figure **S-2**). The Raman analysis reveals additional peaks at 550 cm^{-1} and 1100 cm^{-1} in the ZnMgO thin film, which corresponding to A_{LO} and $2A_{LO}$ phonon modes, respectively (refer to Figure **S-3**). The optical characterization reveals a film band gap of 3.25 electron volts (eV), as shown in Figure **S-4**. In contrast, ZnSnO thin films predominantly display a primary orientation at an angle of 35° (2θ) along the (311) plane, conforming to the space

group $Fd\bar{3}m$, JCPDS 24-1470 (see Figure S-5). Raman spectroscopy reveals the presence of similar vibrational modes in both ZnMgO and ZnSnO thin films (see Figure S-6). The band gap of ZnSnO thin films is determined as 3.16 eV, as shown in Figure S-7. The Cu+Au back contact for CAsE layer characterized by Current-Voltage (I-V) measurements. The analysis reveals that the Cu+Au/CAsE configuration demonstrates ohmic behavior (see Figure S-8), underscoring its potential for effective electrical contact in the solar cell structure.

The fabricated solar cells are schematized in Figure 2. Figure 2(a) displays the SEM image of the CAsE/ZnMgO+ITO interface at a magnification of 50,000X. The image clearly shows the separate layers of the CAsE thin film and ZnMgO window layer. The films are clearly distinguishable, with the CAsE thin film having a thickness of around 1.2 μm , which aligns with the scale shown on the figure (3 μm). The image prominently shows the harmonious interaction between the layers.

Figure 2(b) indicates the scanning electron microscope (SEM) image of the interface between CAsE and ZnSnO+ITO, magnified at 50,000X. The active surface area for both solar cells, SC-1 (incorporating a ZnMgO layer) and SC-2 (incorporating a ZnSnO layer), is uniformly set at 1x1 cm^2 after being brought into contact.



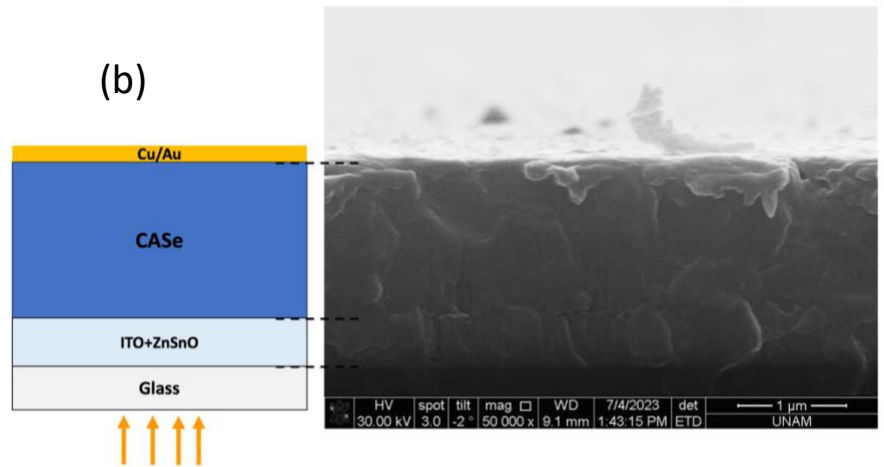


Figure-2: (a) The schematic diagram of SC-1 and SEM image for ITO/ZnMgO/CaSe interface, (b) The schematic diagram of SC-2 and SEM image for ITO/ZnSnO/CaSe interface.

Valuable device parameters are obtained by conducting current density-voltage (J-V) measurements on the fabricated solar cells. Figure 3(a) depicts the J-V graph and Incident Photon to Current Efficiency (IPCE) spectrum with integrated photocurrent (J_{int}) calculated under AM 1.5 G solar irradiation for SC-1. It shows a short-circuit current density (J_{sc}) of 19.4 mA/cm², an open-circuit voltage (V_{oc}) of 0.28 Volts, a fill factor (FF) of 39.14%, and a power conversion efficiency (η) of 2.12%. Figure 3(b) displays the J-V measurements and Incident Photon to Current Efficiency (IPCE) spectrum with integrated photocurrent calculated under AM 1.5 G solar irradiation for SC-2 in a similar manner. SC-2 demonstrates the value of J_{sc} around 19.6 mA/cm², the value of V_{oc} around 0.31 Volts, The value of FF around 40%, and a power conversion efficiency (η) of 2.43%. As can be seen from Figure 3, the J_{sc} values obtained from the J_{int} calculation are close to the values obtained from the J-V measurements.

Prior research in the field has shown the diverse synthesis methodologies, encompassing physical vacuum techniques and chemical approaches, resulting in noteworthy yet constrained power conversion efficiencies (PCEs). Welch et al. used self-organized sputtering with molybdenum (Mo) as the back contact to achieve 3% PCE, and two years later, they set a record with 4.7%. [8], [9].

Wang et al. achieved an efficiency of 3.04% [10]. In another study, Rampino et al. published a study on a thin film solar cell made of CuSbSe₂ with 3.8% [11]. The predominant chemical method employed for fabricating prototypes
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of thin film solar cells based on CuSbSe_2 relies on the utilization of hydrazine. Xue et al. fabricated a thin film solar cell using fluorine-doped SnO_2 (FTO) and achieved an efficiency of 1.32% [12]. The present study uses superstrate solar cell architecture with a Cd-free layer to achieve power conversion efficiencies of 2.13% and 2.43%, demonstrating advances in thin-film solar cell technology.

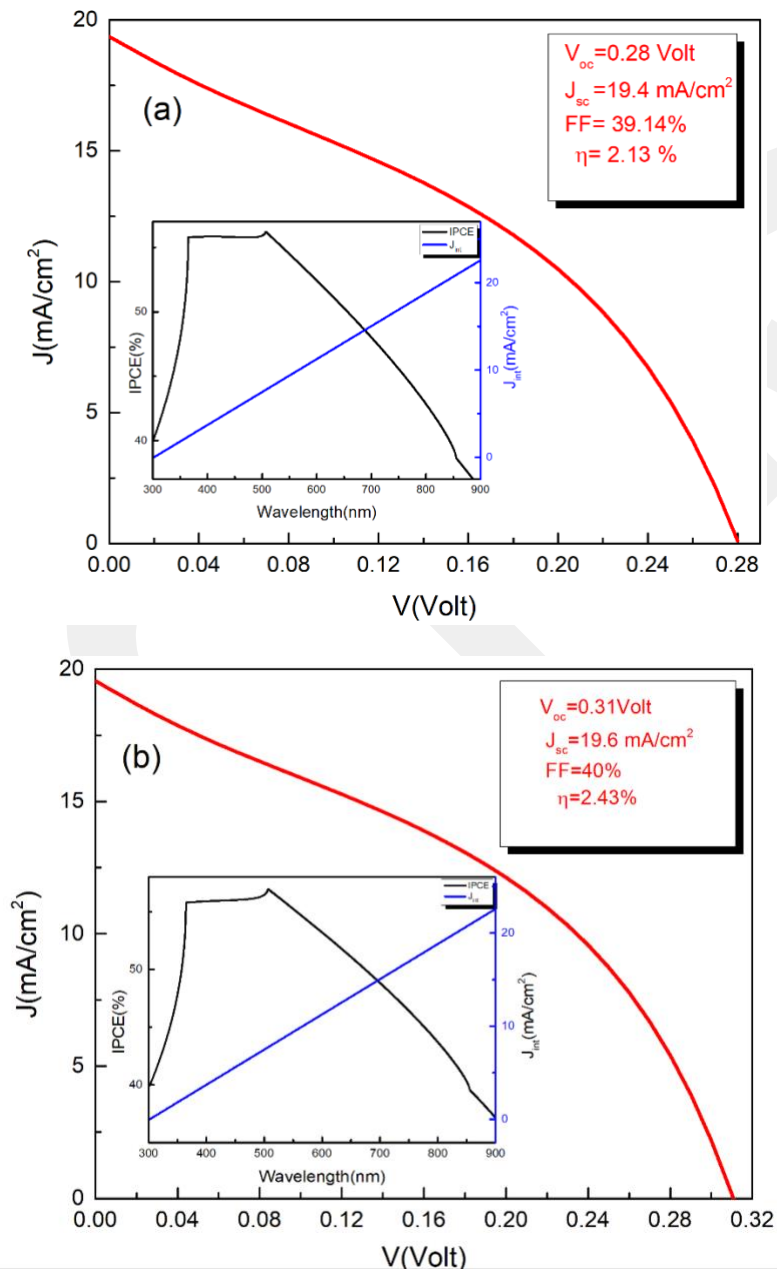


Figure-3: The J-V plots for (a) SC-1 and (b) SC-2 (Insets on each plot show IPCE spectra and integrated photocurrent calculated under AM 1.5 G solar irradiation).

CONCLUSION

A comparative analysis of the fabricated *SC-1* and *SC-2* revealed that *SC-2* have been exhibited superior performance with higher J_{sc} , open-circuit voltage V_{oc} , and overall power conversion efficiency value when compared to *SC-1*. This observation underscores the enhanced efficiency of employing ZnSnO as the window layer in CAsE thin-film solar cells, contributing to a notable improvement in cell efficiency. The present study's innovative approach, featuring a glass/ITO/doped-ZnO/CAsE/Cu+Au top layer structure and the exclusion of toxic materials like CdS, resulted in an impressive efficiency of 2.13% and 2.43%. Although the figure may appear slightly lower, it is crucial to emphasize the innovative design, the absence of toxic materials, and the potential for additional improvements. These factors show a contribution to the advancement of environmentally friendly and safer solar energy technologies.

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Declaration of generative AI and AI-assisted technologies in the writing process

During the preparation of this work the authors used QuillBot to improve readability. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

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