One-Step Sputtering CuInGaSe 2 (CIGSe) Process for Thin Film Solar Cells: Progress and Challenges

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Abstract. The one-step sputtering for fabricating CuInGaSe2 (CIGSe) solar cells has been gaining attention due to its potential for simplifying the manufacturing process, large area uniformity, and environmentally friendly as less reliance on toxic Se precursors such as H 2 Se. Despite these advantages, several drawbacks remain. To date, devices fabricated by quaternary sputtering without additional selenization have been limited in efficiency to about 16%, and realizing bandgap grading in order to match the performance of the best evaporated devices presents a challenge. We discuss the prospects for quaternary sputtering as a fabrication technique for CIGSe and highlight areas of research that may result in improved performance. It also delves into the challenges faced in optimizing the material properties and device performance, and the ongoing research efforts to overcome these hurdles. The paper concludes with a perspective on the future directions for the field, emphasizing the importance of this research in the context of sustainable energy solutions.

Keywords: one-step sputtering, performance enhancement, challenges

1. Introduction

The Copper Indium Gallium Selenide (CIGSe) solar cells industry is currently experiencing significant growth and development. The global CIGSE solar cell market size is projected to grow from USD 1.02 Billion in 2017 to a significant value by 2028, at a Compound Annual Growth Rate (CAGR) of 13.5% during the forecast period². The market growth can be attributed to the increasing demand for renewable energy sources and rising concerns about greenhouse gas emissions and climate change.

This thin film solar cell (SC) has high efficiency similar to crystalline silicon SC, but presenting lower cost due to thinner absorber layer (2-2.5 μ m thickness) (Ramanujam et al., 2020). This material, part of the direct band gap chalcopyrite semiconductor compounds family, boasts exceptional optical properties such as a high absorption coefficient ($\alpha \sim 105~{\rm cm^{-1}}$), long-term stability, and high radiation tolerance. Furthermore, the ability to engineer its band gap through the partial substitution of indium with gallium adds a layer of flexibility, allowing for the manipulation of optical absorption. There are several techniques currently in use for the fabricating of CIGSe absorber layers. These are co-evaporation (Chantana et al., 2016), sputtering (Brozak et al., 2017; Chen et al., 2012), pulsed laser deposition (Nicolaou et al., 2020), as well as non-vacuum methods like ink-printing, spin coating, and electrochemical deposition 6. Despite the variety of methods available, the production of high-quality CIGSE thin films for use in laboratory-scale or module-level solar cells is typically achieved through the use of either co-evaporation or sputtering processes.

One-step sputtering deposition of CIGSe absorber, a method that prepares both the metals and Se in a single target, has been suggested as a potentially scalable deposition

technique for manufacturing. This method provides straightforward process control, uniformity over large areas, and eliminates the need for an expensive and potentially dangerous H2Se gas step. However, devices fabricated by quaternary target sputtering without additional salinization have been limited in efficiency to about 11% (CIGSe Solar Cell Market Report). Thus, make it remain on laboratory research. However, ongoing research and development efforts are aimed at improving this efficiency and realizing bandgap grading to match the performance of the best evaporated devices.

In this review, we discuss the ongoing research, future potential, and challenge of one-step sputtering of CIGSe. Section II offers an overview of single-step deposition techniques for CIGSe, outlining the benefits and hurdles of this method. Section III provides an in-depth explanation of the various approaches that have been done to enhance performance of one-step CIGSe process SCs. Lastly, Section IV contemplates the challenge of one-step sputtering CIGSe, addressing current performance limitations and potential solutions.

2. Overview of One-Step Sputtering CIGSe Process

The chalcopyrite materials, CIGSe, is a direct transition bandgap p-type semiconductor. The term "chalcopyrite" comes from the mineral CuFeSe2, which forms in the tetragonal crystal system space group I42d, as depicted in Fig. 1(a). While these I—III—VI2 ternary compounds and their quaternary alloys encompass a wide variety of material systems, our main focus is on CuInxGa1xSe2 systems. In this quaternary mixed alloy systems, the lattice constants, a and c, change linearly with respect to x. A composition of x = 0.8 is observed to achieve the ideal c = 2a ratio. As a promising absorber material, it has attracted considerable attention due to its direct band $(1.0 \sim 1.12 \text{ eV})$, high absorption co-efficient (10^5 cm^{-1}) and low wasted material.

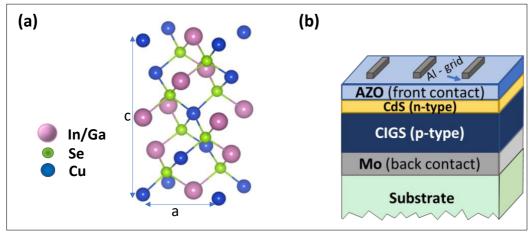


Figure 1. (a) CIGSe chalcopyrite crystal structure, and (b) CIGSe solar cell structure.

Figure 1 (b) illustrates a CIGSe-based solar cell with a structure that includes substrate/Mo/p-CIGSe/n-CdS/intrinsic ZnO/ZnO:Al/ARC/metal-grid. In this architecture, the light enters the cell through the Transparent Conductive Oxide (TCO), and a back contact is applied to the substrate. It is important to note that in the superstrate configuration, light enters the cell through a glass substrate, which serves as a window for illumination. The operation of the CIGSe solar cell is as follows: The n-type CdS buffer layer (with a bandgap energy, Eg, of approximately 2.4 eV) allows light up to 2.4 eV to reach the absorber, where the majority of electron-hole pairs are generated. Photons

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with energy greater than 2.4 eV are absorbed by the CdS layer. Due to the built-in electric field at the p-n junction interface (between CIGSe/CdS), electrons within the diffusion length region are driven from the p-type absorber to the n-type buffer layer and collected by the n-type electrode. Similarly, holes are driven from the n-type layer to the p-type layer and collected by the p-type electrode.

An additional mechanism is the back surface field (BSF) in the CIGSe layer, created by a Ga gradient near the Mo back contact. This BSF reflects electrons towards the p-n junction, where they are ultimately collected by the n-type electrode. The BSF also reduces minority carrier recombination at the rear side of the device, near the CIGSe and Mo interface. More details about the BSF can be found in other sources.

A single quaternary target, CIGSe, was prepared by hot sintering process from the uniformed mixture of high purity Cu₂Se, In₂Se₃, and Ga₂Se₃ powders at 600°-800° sintering temperature (Chen et al., 2012). Elements composition has percentage close to its stoichiometry with Cu₂S% (known as rich copper target) or <23% (known as poor Copper target), In 17.5%-19%, Ga₃7-8% and Se₃51-52% (Chen et al., 2012; Lin et al., 2013; Zhang et al., 2017). The CIGSe film are growth on Mo/substrate via magnetron sputtering (Frantz et al., 2016) (Hsu et al., 2017; A. J. Zhou et al., 2012), pulse DC sputtering (Wang et al., 2021), or Radio Frequency sputtering (Brozak et al., 2017; Kim et al., 2020) at high temperature substrate 500°C-550°C (Chen et al., 2012; D. Zhou et al., 2016). Although the deposition is simple, less toxic and a good film uniformity, poor Se content becomes challenge to obtain high quality CISGS absorber. Thus, many approaches have been done to improve device efficiency in order to make this process reliable for mass production.

3. Efficiency Enhancement Approach

Since one-step process has a lack of Se which cause Se vacancy (V_{Se}) that result to poor absorber, adding enough Se is necessary to have preferable performance. Shi et al. fabricated CIGSe SCs by implemented post deposition selenization step using solid Se source after growing CIGSe layer (Hsu et al., 2017; Shi et al., 2011). Recently, efficiency of 16.7% has been achieved by utilizing H₂Se after absorber (Ouyang et al., 2015). Although these methods are significantly improving the efficiency, environmentally friendly process is preferable. Some ways to enhance single-step CIGSe deposition is elaborated in the section below.

CIGS Absorber Modification

Absorber crystallinity and its crystal prefer orientation determines the device performance. Peng et al. controlled CIGSe layer growth by vary Cu composition, post annealing, and having bilayer absorber. This group found that double layer, rich and poor Cu layer followed by annealing thievingly improve performance by almost 3% due to larger grain size (see Fig. 2) of absorber and higher current density compared to annealed one-layer CIGS (Peng et al., 2017). Sputtering rich copper content CIGSe followed by In₂Se₃ layer are able to change absorber into Cu poor surface which result in higher short circuit current and bigger grain size (Peng et al., 2018). Inserting of Sb in the bottom and middle of CIGS layers could significantly increase the grain size (Zhang et al., 2017). In addition, controlling CIGSe prefer orientation is also important to improve device performance. This can be done by inserting a thin In₂Se₃ precursor before growing absorber (Yan et al., 2016), controlling Mo orientation (Jingxue et al., 2016; Kamikawa-Shimizu et al., 2009; Lin et al., 2012).

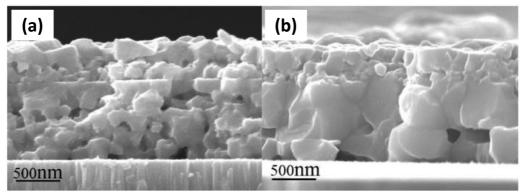


Figure 2. (a) the single layer annealed CIGS thin film consisting of 1100 nm-thickness bottom layer of CIGS with Cu content of 0.97 and 500 nm-thickness top layer with Cu content of 0.82; (b) the bilayer annealed CIGS thin film consisting of 1100 nm-thickness bottom layer of CIGS with Cu content of 0.97 and 500 nm-thickness top layer with Cu content of 0.82 (Peng et al., 2017)

Alkali Metals Incorporation

The effects of diffusion of alkali metal such as Na, K, Cs, Rb on CIGS SCs been studied (Cho et al., 2018; Li et al., 2017; Sun et al., 2017). Hsu and teams have revealed that efficiency can be boosted up to 11% by introducing NaF after CIGSe layer to suppress effect of Se-deficient (Hsu et al., 2015). The same group also has done double post deposition of alkali by depositing NaF and KF in sequent (Hsu et al., 2017). The effect of Na has been widely studied. It increases the net acceptor concentration (Sun et al., 2017, 2017), it affects grain growth (Hsu et al., 2015), surface passivation, the diffusion of Ga towards back contact, and resulting better p-type properties (Salomé et al., 2015). When Na or K is incorporated, Na can reduce both InCu and VSe, while K can significantly promote the oxidation of VSe which can be observed on short circuit Voltage (Voc) improvement (Fig. 3(a)). However, when NaF deposited followed by KF after CIGSe, it further enhances absorber quality because of effective passivation of both defects (Hsu et al., 2017) which shown on Photo-luminescence (PL) spectra Fig 3(b).

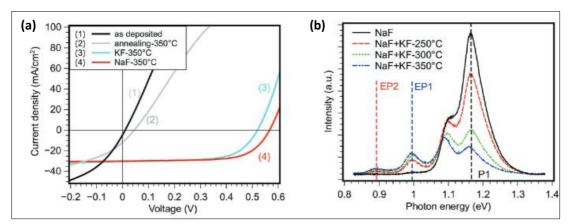


Figure 3. (a) J-V curves of CIGSe solar cell with different post-treatment, (b) PL spectra of Na-PDT without and with KF-PDT (Hsu et al., 2017)

Band-Gap Engineering

The bandgap engineering of CIGSe absorber layers proves to be highly beneficial

in boosting cell performance. This is because it minimizes recombination losses and maximize carrier collection within the cell (Park et al., 2016). In co-sputtering method, bandgap of CIGSe can be easily tuned by adjusting Ga/(Ga+In) ratio during absorber deposition. In the process of quaternary target sputtering, the capacity to alter the elemental ratio of the sputtered atoms from a single target is somewhat restricted. To achieve compositional variation, one must either employ a combination of ternary targets or use stoichiometric quaternary and elemental targets. Ga grading or controlling Ga concentration to replace In in single step CIGSe deposition can be done by using a combinatorial magnetron sputtering system from a CIS/CGS stoichiometric single target (Park et al., 2016; Lyu et al., 2019).

However, Ga₂Se₃ as co-sputtering target may lead to Ga₂O₃ formation which degrades device's efficiency. Wang et al. and coworkers have used different co-sputtering targets, CuGaSe₂ (Wang et al., 2020) and In₂Se₃ (Wang et al., 2021) with CIGSe to modified the bandgap. The prior co-sputtering technique inhibit the creation of secondary phases and attain a customizable normal grading which results in a peak efficiency of 15.63%, achieved without the need for post-selenization on flexible substrates (Wang et al., 2020). While the later effectively passivated surface Se vacancies (VSe) due to optimize surface sulfurization (Wang et al., 2021). With increasing Ga/(In + Ga) ratio, diffraction peak positions shifted to high angle for both (112) and (220) planes. Similar trend also observed on Raman spectra.

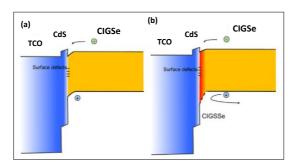


Figure 4. Diagram illustrating the band structure of samples: (a) without any treatment, and (b) treated with In2S3 (Wang et al., 2021)

Increasing Ga content leads to an increase in VOC as result of reduction of recombination and a decrease in JSC due to absorber quality improvement, witnessed on both studies. From Fig. 4 (a)(b), enlargement of surface bandgap formed due to formation of a thin layer of CIGSSe at the interface between buffer layer and absorber layer. Moreover, the presence of this layer on the surface results in a downward shift of the valence band. This, in turn, reduces the interface recombination.

4. Challenges

Improving the efficiency of CIGS solar cells using a one-step sputtering technique can present several challenges:

- a. Impurity issue: in the sputtering process produces a film composition that mirrors the target composition, meaning any impurities present in the target will be transferred to the final film. Thus well care need to taken to make sure target fabrication and sputtering chamber free from contaminant (Frantz et al., 2016)
- b. Optimization of deposition parameters: The deposition pressure and RF-Power used during the sputtering process can significantly affect the properties of the

- CIGS thin film². Finding the optimal parameters to achieve a smooth surface and good crystallinity can be challenging (Desarada et al., 2023).
- c. Post-deposition annealing: After the CIGS thin film is deposited, a post-annealing process is often required to improve the crystallinity, grain size, and compactness of the CIGS samples (Desarada et al., 2023; Suryavanshi & Panchal, 2023). The choice of annealing technique and parameters can greatly influence the properties of the CIGS thin film.
- d. Stoichiometry control: The energy bandgap of CIGS semiconductors can be tuned in the range of 1.05eV to 1.65 eV by changing the stoichiometry of indium (In) to gallium (Ga) of the sample. However, achieving the desired stoichiometry can be difficult in a one-step sputtering process (Suryavanshi & Panchal, 2023).
- e. Formation of defects: High Ga contents can lead to the formation of deep-level defects, which can degrade the performance of the CIGS solar cells (Huang et al., 2018).
- f. Cost and scalability: While the one-step sputtering technique can potentially reduce the cost and increase the scalability of CIGS solar cell production, achieving high efficiency at a low cost and large scale remains a challenge (Desarada et al., 2023).

These challenges highlight the complexity of the one-step sputtering process for CIGS solar cells and the need for ongoing research and development to optimize this process.

5. Conclusion

The prominent advantages of single step CIGSe sputtering is suitable for large area deposition due to its good uniformity and simplicity. However, the solar cells efficiency is still below from co-sputtering or multistep methods. In order to achieve feasibility for scale up, many approaches have need implemented. CIGSe layer quality can be improved by controlling Mo layer phase, inserting wetting layer or barrier layer. Secondly, alkali metal atoms deployment, Na or K, improves carrier concentration and give some passivation effects on CIGSe surface and later reduce recombination at interface, thus the Voc of devices. The last, bandgap grading can be introduced into single-step process by co-sputtering with other targets to adjust Ga/(Ga+In) ratio. This results in an elevation of VOC due to the reduction in recombination, and a decline in JSC as a consequence of the improvement in absorber quality. Despite the performance slowly elevated, there are some challenges remained. Impurity levels in the target, stoichiometry issue, defects formation, and cost for scaling up are need to be solved in order to be able to compete with other CIGSe thin film SCs fabrication methods.

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