

State of the art review on the Cu(In,Ga)Se₂ thin-film solar cells

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ABSTRACT Solar cell efficiency and durability are some of the critical research areas in the field of solar photovoltaic (PV) technology. Thin-film PV cells have been increasingly used in many industries and applications such as wearable electronics, self-energizing systems, on the roof of electrical vehicles, trains, solar boats, etc. Various types of thin-film solar cells have been investigated, produced, and exploited around the world. Different types of chalcopyrite semiconductors are used in high-efficiency thin-film solar cells, specifically in flexible PV cells. Cu(In,Ga)Se₂ (CIGS) solar cells are promising technology among the thin-film solar cells due to their unique features. However, there are many challenges towards the development of the photovoltaic performance of thin-films and ultra-thin-film solar cells. The thickness and surface roughness of the thin-film layer which has a significant effect on the performance of cells, electron recombination, interface contact between thin-film layers, grain size, and grain boundaries can be mentioned as major problems. Many solutions for the cited problems have been investigated and reported. Alkali post-deposition treatment, for example, was noted to reduce the thickness of the absorber. Reduction in absorber thickness leads to a significant decrease in electron recombination which improves the PV performance of CIGS solar cells. Cs post-deposition treatment (Cs-PDT), on the other hand, improves cell performance by creating conduction band upward and valence band downshifts. Low thickness of absorber increase crystallinity and surface roughness of the CIGS absorber. A high amount of Ga concentration is reported to reduce the grain size and major loss of charge carrier. In addition to the high Ga amount, sulfurization is also responsible for decreasing the grain size and the fill factor (FF) of the PV cell.

Keywords: Chalcopyrite semiconductors, Cu(In,Ga)Se₂ (CIGS), Thin-film solar cells, Flexible solar cells, Alkali treatment

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1. INTRODUCTION

Since the industrial revolution, the energy demand has been mostly supplied by fossil energy resources, worldwide. Using fossil energy resources for a couple of centuries resulted in environmental and human health problems such as global warming, and so climate change. The burning of fossil fuels is responsible for three-quarters of greenhouse emissions and causes massive air pollution and health problems such as at least 5 million premature deaths annually. Based on these concerns and major implications, reducing CO₂ emission and local air pollution are strongly recommended [1, 2]. Therefore,

there is increasing adoption of renewable energy technologies as well as nuclear energy to address these problems.

As nuclear energy has its concerns like potential disaster effects such Chernobyl nuclear power plant tragedy (nuclear accident on April 26, 1986), and water pollution by radioactive pollutant as in Fukushima reactor, Japan due to the earthquake followed by tsunami (March 11, 2011). To reduce carbon emissions and prevent such tragedies; solar energy is one of the solutions as it is a renewable, sustainable, clean, and environmentally friendly energy resource. Nowadays,

renewable energy is a long-term strategy to ensure a stable and adequate supply of electrical energy, especially after the Paris Agreement in 2015 [3] which is focusing on the reduction of carbon dioxide emissions [4]. On the other hand, the fast decline of fossil energy resources is another important reason for the increasing implementation of alternative energy sources. As in 2016, the share of renewable energies is around 24 % of worldwide energy consumption [5] Solar energy conversion is part of this strategy. Photovoltaic technology is the only direct conversion technology of sunlight (photon energy) into electrical energy. Various types of PV cells have been investigated and produced during past decades across the world. Among the

versatile solar cells, thin-film solar cells have received more attention from scientists and researchers.

This manuscript, therefore, aims to provide a brief review on thin-film photovoltaic technologies, and the recent progress in CIGS solar cell variants in detail along with the current challenges and expectations.

2. CLASSIFICATION OF THIN-FILM PV TECHNOLOGIES

In general, thin-film PV technology is classified based on materials used in the structure: Si-based, compound semiconductor-based, and novel materials based ones. The following subsections are intended to give brief information about the subcategories of these thin-film PV technologies as shown in Figure 1.

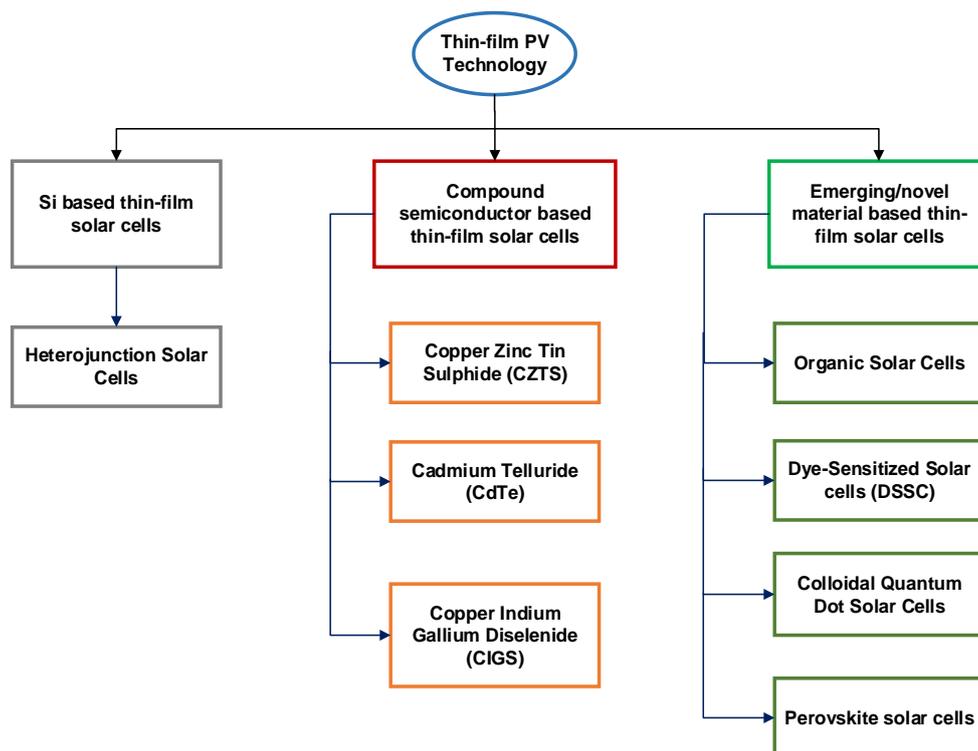


Fig. 1. Classification of thin-film PV technologies

2.1. Silicon-Based Thin-film Solar Cells

These types of solar cells are named also as heterojunction solar cells. In this type of solar cells, hydrogenated amorphous silicon (a-Si:H) as shown in Figure 2 is used as a coating through different post-deposition methods. Plasma enhanced chemical vapor deposition (PECVD) technique is well known for a-Si:H thin-film growth on a crystalline silicon (c-Si) substrate [6]. The a-Si:H is widely studied in literature due to, its high optical characteristics [7], photovoltaic performance, and high stability (less light-induced degradation) [4,7]. The highest cell efficiency has recently been reported by National Renewable Energy Laboratory (NREL) – USA. The report shows that heterojunction silicon solar cells have an efficiency of 26.7 %. However, the highest efficiency recorded by multi-junction (four junctions) solar cells is 47.1 % in 2020 [8].

2.2. Novel Material Based Thin-film Solar Cells

As illustrated in Fig.1, emerging solar cells or novel material based solar cells are part of thin-film solar cells which are also called as the third generation of PV technology. This generation of PV technology includes organic solar cells, dye-sensitized solar cells (DSSC), colloidal quantum-dot solar cells, and perovskite solar cells [8, 9]. This category of thin-film solar cells is, very promising as they are colorful, transparent, and flexible [10, 11].

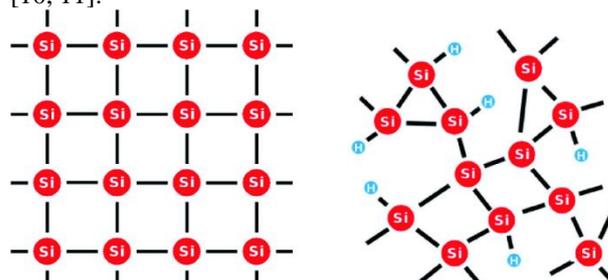


Fig. 2. Structures of crystalline silicon: c-Si (left), and hydrogenated amorphous silicon: a-Si:H (right) [7]

Due to these unique features, it has found several applications including in the roof solar panels for vehicles, locomotives, solar boats, building glasses, and so on. Organic solar cells [12] (Fig. 3), also known as plastic or polymer-based solar cells, consist of conductive polymer substrate as electrodes, and semiconductor material embedded into natural/organic dye [9]. The highest efficiency for organic solar cells is reported as 17.4 % [8].

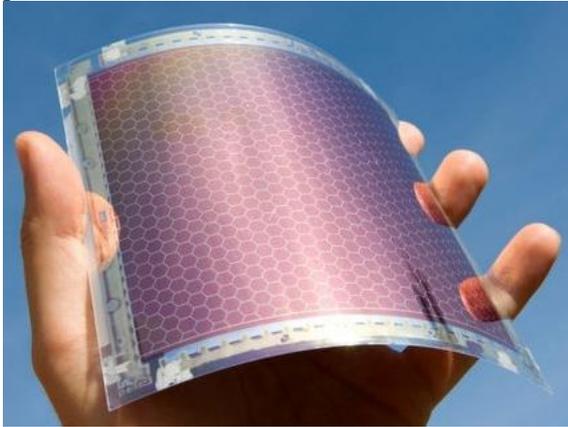


Fig. 3. Organic photovoltaic cells convert solar energy into electric power through organic molecules [12]

Dye-sensitized solar cells (DSSC) are among the emerging thin-film solar cells with the multi-layered structure which consists of 1) two transparent conductive oxide glass or polymer, 2) ZnO, TiO₂ or other types of semiconductor along with organic/synthetic dye, 3) solid-state/liquid electrolyte as electron donor to the dye, and 4) a counter electrode as the back surface of the cell [8, 13, 14, 15]. The working principle of the DSSC is illustrated in Figure 4 [16]. Basic DSSC efficiency is currently reached to 12.6% while the Perovskite solar cells which are developed form of DSSC, and among the high-efficiency solar cells (25.2 %) as reported by NREL.

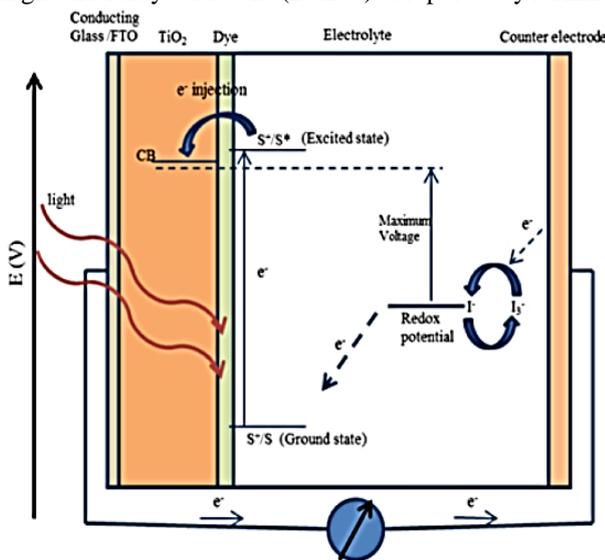


Fig. 4. Working principle of DSSC [16]

The working principle of DSSC can be described in four basic steps. In the first step, the sensitizer (dye) is used to improve the light absorption of the semiconductor. By the absorption of photons, the energy level of the dye is changed from the ground state to the

excited state. In the second step, the excited electrons with a lifetime of nanosecond range are injected into the conduction band of the semiconductor (which is covered by dye molecules). In the third step, the injected electrons are transported between nanoparticles of the semiconductor and moving by diffusion toward the transparent conductive oxide which is acting as an anode electrode. In the fourth step, the electrons reach from the outer circuit to the counter electrode. Then, those electrons reduce tri-iodide (I⁻³) to iodide (I⁻) by a redox reaction. Thus, regeneration takes place due to the acceptance of electrons from the I⁻ ion, then I⁻ is oxidized to the I⁻³ state [8,13,14 and 15].

2.3. Compound Semiconductor Based Thin-film Solar Cells

Compound semiconductor (in the form of chalcopyrite structure) based thin-film solar cells are promising technologies due to their unique features. These thin-film technologies are lightweight, flexible, easy to manufacture, and cheaper than conventional PV technologies or Si-based thin-film technologies. Copper Indium Gallium Diselenide “Cu(In,Ga)Se₂” (CIGS), Cadmium Telluride (CdTe), Copper Zinc Tin Sulphide (CZTS) are in the group of compound material based thin-film solar cells. In this group of thin-film PV technology, CIGS thin-film solar cells can be mentioned as one of the favorites due to their high optical absorption coefficient, modifiable bandgap from 1.02 eV for CuInSe₂ (CIS) up to 1.68 eV for CuGaSe₂ (CGS) [17] and benign grain boundaries [18].

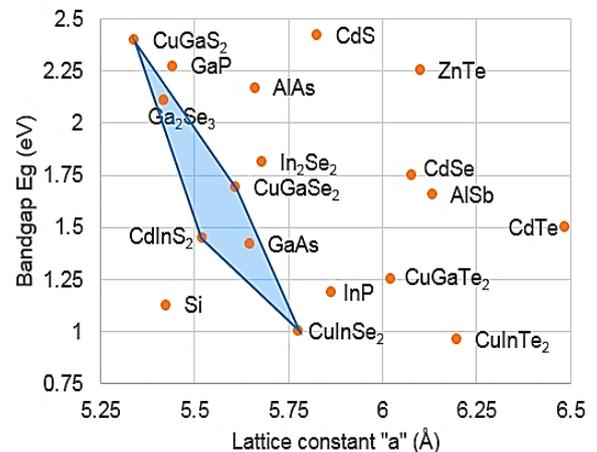


Fig. 5. CIGS technology: modifiable bandgap of CIGS solar cells [After 20]

2.3.1. CIGS Thin-film Solar Cells

Cu(InGa)Se₂ (CIGS) is a direct bandgap compound semiconductor used as a photon absorber in PV technology. CIGS is an alloy of CuInSe₂ and CuGaSe₂ that crystallize in the form of a chalcopyrite structure. The advantages of CIGS PV technology are low cost due to low-cost material and less energy consumption, flexibility in design, modifiable bandgap, and lightweight. Its efficiency is 23.4 % [7, 18]. The chalcopyrite crystal structure retains the tetrahedral bonding as a solid solution with the variety of the bond lengths according to the alternative element, which results in shifted lattice constants (Fig. 5 and Fig 6) [20]

Therefore, the bandgap of the semiconductor is modifiable, as illustrated in Figure 5 as well [21].

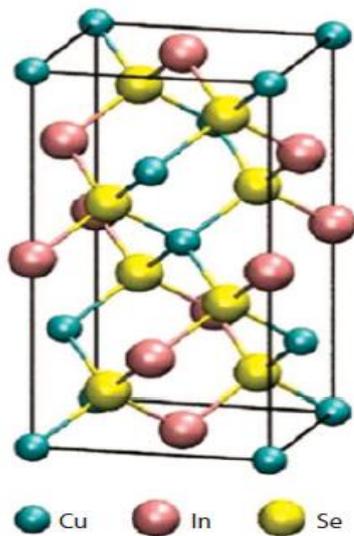


Fig. 6. Tetrahedral bonding as a solid solution with the variety of the bond lengths [After 35]

Modifiability of the bandgap of the chalcopyrite semiconductor makes it possible to process and deal with various materials within the composition, crystallinity, and surface characteristics of the CIGS absorber [20].

Recent reports show that recently Ag is used instead of Cu as basic material for CIGS absorber [21]. CIGS thin-film photovoltaic cell is promising thin-film solar cell due to its high conversion efficiency and long-term stability as it has been recently announced a new record of efficiency of 23.35% for $\text{Cu}(\text{InGa})(\text{SSe})_2$ (CIGSS) [22], while in 2017, the $\text{Cu}(\text{InGa})\text{Se}_2$ (CIGS) thin-film solar cell had a world-record of thin-film solar cells efficiency of 22.9% [23]. The historical development of CIGS solar cells has been shown in Figure 7 and CIGS with/without Cd buffer layer in Figure 8 [24]. Based on recent researches and development, it is observed that thin-film photovoltaic technology is continuously being developed. CIGS thin-film PV cells can be produced using fewer materials and simpler processes compared to conventional Si-based modules. Over 5 GW of CIGS modules have been produced by Solar Frontier K.K. (Tokyo, Japan), a subsidiary of Idemitsu Kosan Co., Ltd. (Tokyo, Japan), and distributed across the world to date [24].

In terms of economical processing, high material utilization, and low material wastes, several techniques have been investigated and suggested for the development of thin-film technology based on CIGS semiconductor. Vacuum co-evaporation, non-vacuum absorber formation, chemical spray pyrolysis, electrodeposition, paste coating can be mentioned as applicable techniques in the processing of CIGS thin-film solar cells [18]. Non-vacuum deposition processes have been preferred for high-quality CIGS according to a report by M. Kaelin et al. Despite the significant achievements in CIGS, there are still various problems in CIGS research including grain boundaries, sodium, inverted CIGS phase at surface, defects, etc. (Table 1) [25]. To refine surface defects, and improve the performance of CIGS, alkali metal post-deposition

treatment (PDT) including Na, Rb, K, and Cs is suggested to improve efficiency [22]. Surface sulfurization of the top layer of the CIGS cell, which has two distinct sulfur phases on the absorber layer, was reported to reduce the cell performance. Based on the results, the performance of the cell highly depends on the thermodynamic stability of the chalcogenide structure of the upper layer [26]. Bandgap grading of compound semiconductor in CIGS cell was also studied and reported in many papers as well in recent researches [23]. Heavy alkali metals such as rubidium and cesium have influences on the performance of CIGS thin-film solar cells and are noted to increase photovoltaic efficiency up to 23% [27]. In addition to those alkali metal treatments of CIGS absorber, crystallinity and thickness of CIGS thin-film solar cell are other effective parameters on the performance of the CIGS thin-film solar cell.

2.3.1.1. The working principle of the CIGS solar cell

A CIGS solar cell with the n-type CdS buffer layer which has a bandgap energy of $E_g \approx 2.4$ eV, transmits the incident light up (≥ 2.4 eV) into the absorber where electron-hole pairs are mainly generated. High energy incident light (photons) is absorbed by the CdS layer. Due to an electric field built across the p-n junction interface between absorber and buffer layer, the electrons within the diffusion length region are swept away from the p-type absorber to the n-type buffer layer then collected by the n-type electrode. On the other side, similarly, holes are swept away from the n-type layer to the p-type layer and collected by the p-type electrode (Fig. 10).

A back surface field (BSF) in the CIGS layer created by a Ga gradient (close to Mo back contact) is an additional mechanism that reflects electrons towards the p-n junction to be finally collected through the n-type electrode. The BSF reduces minority carrier recombination at the rear (close to the CIGS and Mo interface) side of the device [28]. Performance of the solar cells is commonly characterized by the short circuit current (I_{sc}) and open-circuit voltage (V_{oc}) produced by a PV cell, which is called I-V curve (Fig. 9).

Power conversion efficiency η (in %) is one of the important characterization factors of the PV cells, and can be computed as in Eq. 1, where P_{max} is the maximum power produced by the cell, FF is fill-factor, and P_{in} is the incident power into the unit cell or into the module. The fill-factor (in %) is another important characterization parameter for PV cells, and it can be computed as given in Eq.2.

$$\eta = \frac{P_{max}}{P_{in}} FF = \frac{I_{sc} \cdot V_{oc} \cdot FF}{P_{in}} \quad (1)$$

$$FF = \frac{P_{in}}{P_{max}} \eta \quad (2)$$

2.3.1.2. Effective Parameters on the Performance CIGS PV Cell

Chalcopyrite thin-film solar cells based on CIGS compound semiconductor yielded a very promising performance as noted in the literature.

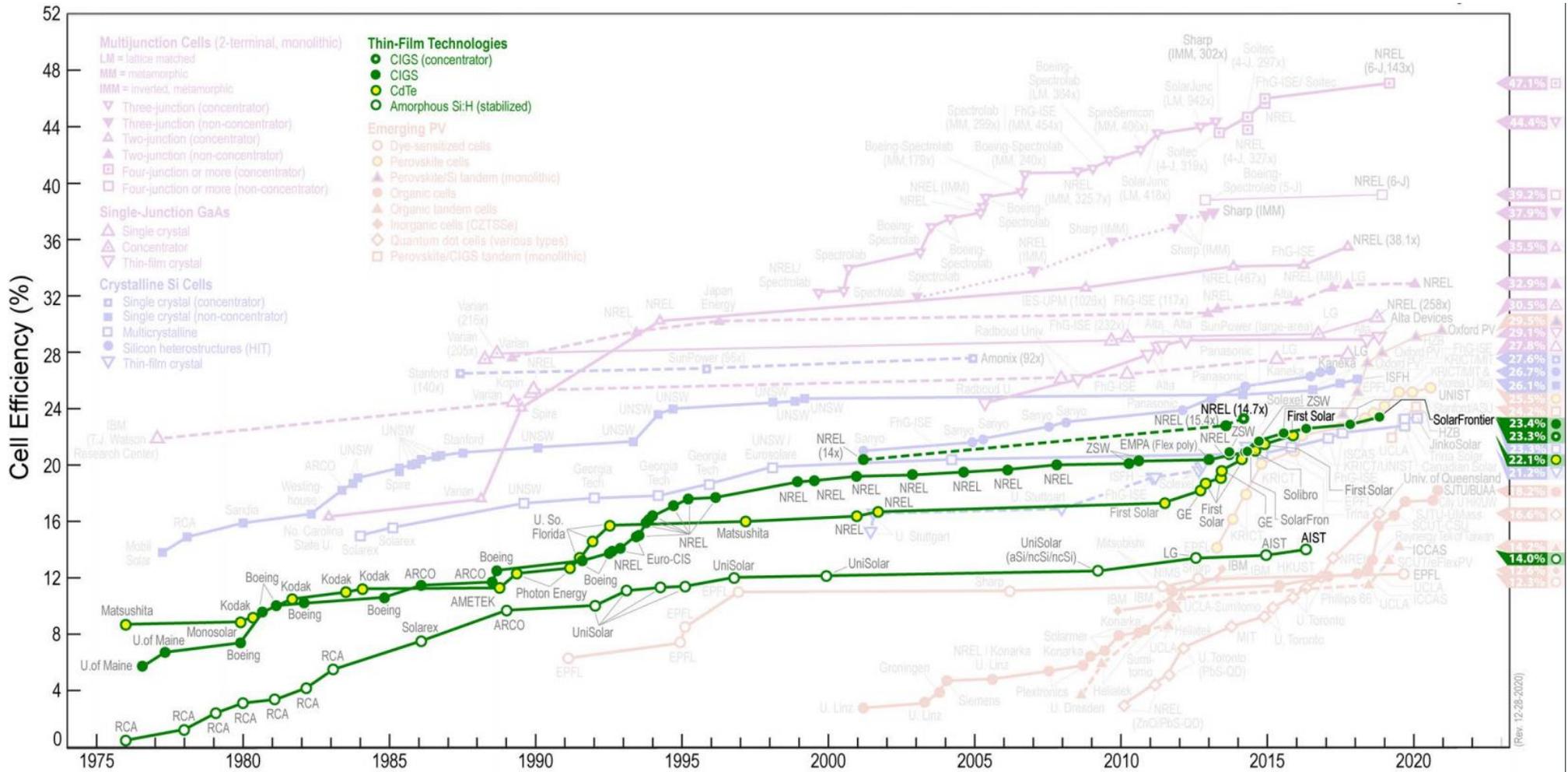


Fig. 7. Historical development of the CIGS solar cells efficiencies based on different technologies [8]

AIST: National Institute of Advanced Industrial Science and Technology, ARCO: Atlantic Richfield Company, Boeing: The Boeing Co., EMPA: Swiss Federal Laboratories for Materials Science and Technology, First Solar: First Solar Inc., LG: LG Electronics, NREL: National Renewable Energy Laboratory, SolarFron: Solar Frontier, Solibro: Solibro GmbH, U. So. Florida: University of South Florida, ZSW: Zentrum für Sonnenenergie- und Wasserstoff-Forschung Baden-Württemberg (Centre for Solar Energy and Hydrogen Research Baden-Württemberg)

CIGS can be made of fewer materials and different compositions. The performance of CIGS solar cells strongly depends on the composition of the materials, surface defects and surface morphologies, manufacturing methods, microstructure, the thickness of the absorber layer, bandgap, and many other parameters of the consisting layers of the cell [16, 20, 21 and 27]. Reduction of charge recombination is a big challenge in the development of CIGS as well as other thin-film solar cells. Effective factors on the performance of CIGS solar

cells are listed in Table 1 and detailed in the following paragraphs.

a) Amount of Ga concentration

The amount of Ga content in the absorber material plays an important role and affects the crystal quality and fabrication of CIGS. A high amount of Ga may cause a reduction in grain size. Therefore, a major loss of carrier collections occurs due to increasing grain-grain or random grain concentration at interfaces [29].

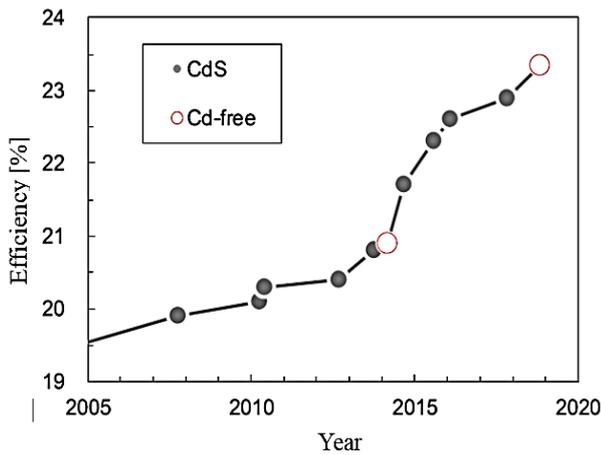


Fig. 8. Historical development of CIGS cells with/without Cd buffer layer [24].

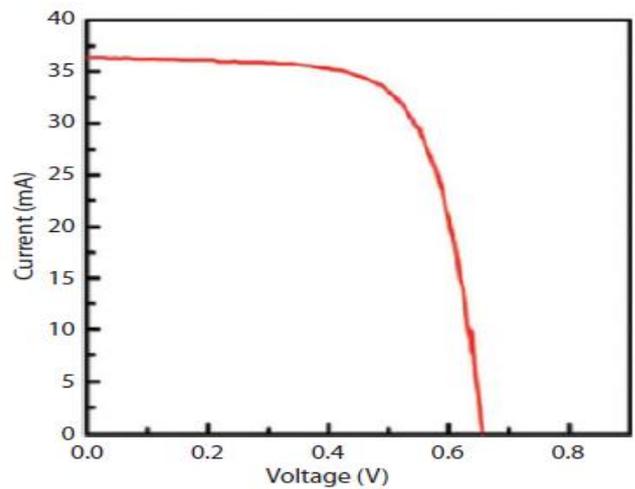


Fig. 9. I–V curve of CIGS/Zn(O,S) with 14% s content under 1.5 AM illumination condition[35].

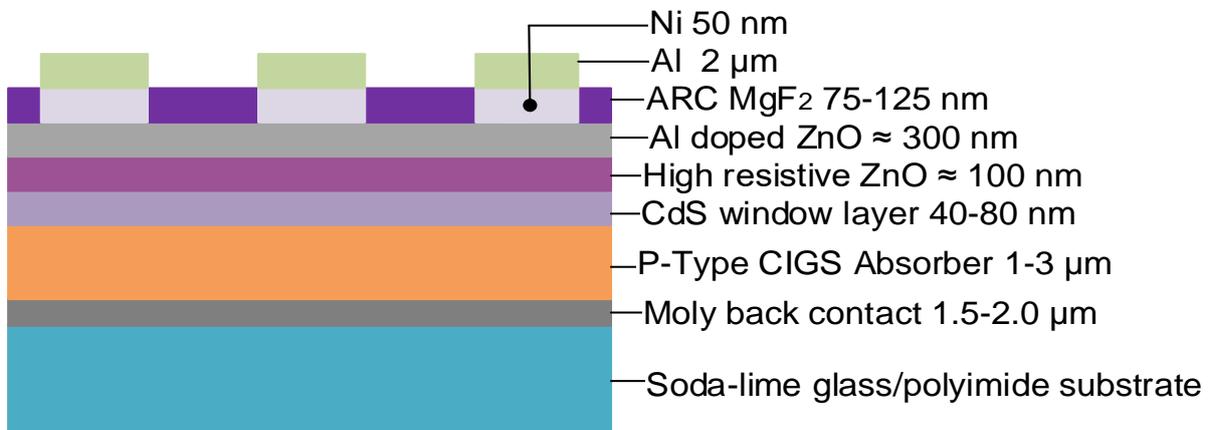


Fig. 10. Schematic of the basic structure of a typical CIGS solar cell with a CdS buffer layer [after 28].

Table 1. Effective parameters on the performance of CIGS solar cells

No.	Parameter	Effects	Reference
1	Surface roughness of absorber	Rough surface has better performance.	[27]
2	High Ga content	Reduces grain size, major loss of charge carrier	[25]
3	Low thickness of absorber	Increases crystallinity, reduce recombination, improve performance	[25]
4	Thicker CIGS absorber	Reduces carrier collection length	[25] [35]
5	Crystallinity of CIGS	Increases performance	[25], [28]
6	Sulfurization	Decreases grain size, reduces fill-factor (FF), does not affect alkali metal treated surface, improves bandgap.	[29]
7	Cs-PDT	Improves performance, creates conduction band upward, valence band downshift	[28]
8	Material composition	Modifiable bandgap	[17]
9	Temperature/Selenization	Does not affect crystallinity and grain size, affects energy gap of CIGS absorber layer	[30]
10	Tandem CIGS absorber layer	Significantly influences cell performance	[17]

Li Weimin *et al.*, reported that crystallinity of wide bandgap CIGS is increased through reducing the thickness, and the carrier collection increases gradually with thin CIGS absorber [29]. The most recent research based on numerical solutions and bandgap optimization of CIGS tandem solar cell notes a considerable achievement. It shows that the CIGS/CIGS tandem cells with various CIGS absorbing layer thickness gained theoretical efficiency of 27.30% [17]. This recent report considered the thickness of CIGS absorbing layer and its bandgap as key parameters to improve the performances of CIGS/CIGS tandem solar cell. It was noted that the optimum thickness for top absorbing layer is 0.18 μm while it is 3.2 μm for bottom layer, considering optimal bandgap combination of 1.68 eV for top cell, and 1.13 eV for bottom cell. There is a numerical investigation on CIGS cell focused on the impact of buffer layers (CdS and ZnS) as well as back materials mirror (Mo, Cu, Au and Ag) on the efficiency. It was revealed that copper nano-pillar geometry reduces parasitic losses [33].

b) Surface morphology

Surface morphology of CIGS film in CIGS cell is another important issue. Surface morphology of CIGS thin-film with RbF-PDT and CsF-PDT were studied and

reported by Paper *et al.* [27]. In both cases, CIGS film showed rough surface morphology. The effects of annealing conditions, as well as selenization process on the CIGS phase, morphology of the absorber layer and optical characteristics of the layer, were studied by E. Ghanbari *et al.* [32]. According to this study, the heating process does not have any effect on CIGS's phase of the nanoparticles while the phase of CIGS is affected by the selenized layer. Based on that study, the heated layer with the energy gap of 1.5 eV and grain size of 30.2 nm had a uniform surface, whereas the bandgap and grain size of the selenized surface increased up to 1.74 eV and 37.6 nm, respectively. It is attributed, that selenized surface of the CIGS layer was rougher than the heated layer.

c) Alkali metal post-deposition treatment

Alkali-metal post-deposition treatment was suggested in many studies to refine surface defects of CIGS absorber which leads to a reduction in electron accumulation at the back contact of the cell [22].

Several alkali metals including heavy alkalis such Cs and Rb as well as other alkalis like Na or K can be applied to improve the cell performance by modifying the bandgap, crystallinity, and refining surface defects [16, 21].

d) Crystallinity

The crystallinity of the absorber is an important factor in CIGS cells, too. Crystallinity and crystal quality can be affected by the amount of Ga in material composition. The high amount of Ga in the material composition reduces the grain size and it causes a major loss of charge carrier because of random grain concentration interface of grain-grain concentration interface. On other hand, the crystallinity of wide bandgap CIGS is reducing the thickness of the absorber [29]. The carrier collection length is gradually reduced by increasing the thickness of the CIGS absorber. In case of high gallium content, CIGS absorber along with the Mo back contact, deposition of CIGS, fine grains near Mo back contact act as dead grain to collect current. It is indicated that good crystallinity of high Ga amount in CIGS absorber is very important to obtain high efficiency. It was noted that reducing the thickness led to improved crystallinity in high bandgap CIGS solar cells [29].

e) Surface sulfurization

Sulfurization is not regarded as a successful treatment for refining surface defects and grain boundary defects as it affects the performance of CIGS solar cells negatively and degrades fill factor (FF) up to 6% [26]. This negative effect can be caused due to the correlation between the sulfur incorporation and the grain size of the CIGS absorber layer. It shows that high sulfur content refines the grain size in the microstructure of the CIGS absorber [26]. H. Aboulfadl *et al.* reported that they expected the formation of sulfur as a thin layer on the surface as a result of the sulfur application while it was shown that sulfur had some outward diffusion into the absorber in their experiments. It caused increasing sulfur concentration in small grains due to effective diffusivity [34].

Treated surfaces with alkali metal Na, K, Rb, and Cs, were not affected by sulfurization on their grain boundaries. It can be considered as an advantage of sulfurization in the case of tuning the bandgap at the neighborhood of P-N junction since it is understood that differentiation of alkali metals at the grain boundaries has positive effects on device performance [26].

Heavy alkali metal Cs has a high influence on the distribution of other alkali-metals such as Na and K along the grain boundaries. Cesium contained grain boundary may create conduction band upward and valence band down-shift. Thus, the carrier recombination can be suppressed by the energy downshift of the valence band at the grain boundaries. However, the electron accumulation can be induced by the conduction band upward, then intensifies the recombination. The controlling thickness of the Cs content layer and tuning the absorber to maintain a strong valence band offset field can reduce or avoid the charge accumulation [22].

3. CONCLUSION

Cu(InGa)Se₂ thin-film solar cells are promising photovoltaic technology among the other thin-film solar cells due to the unique characteristics of the CIGS absorber which has a chalcopyrite structure. CIGS is a direct bandgap semiconductor with a tunable bandgap, and it has a very high absorptivity of sunlight in the interval of visible light. The development of CIGS highly depends on the microstructure and composition of materials used in manufacturing. It was noted that the crystallinity of the CIGS absorber and crystal quality highly influence the cell performance. The crystallinity can be controlled by the amount of gallium in material composition in an optimum range to avoid reducing grain size and major loss in charge carrier. The thickness of the absorber layer is another important physical parameter of the CIGS that affects cell performance [35]. A decrease in thickness increases crystallinity and reduces carrier recombination due to the increase in carrier collection length. The role of alkali metal treatment on the performance cannot be underestimated and it is a key function toward the development of the CIGS thin-film solar cells. As CIGS is a modifiable bandgap cell, sulfurization may play an important role to improve absorber bandgap. Sulfurization can increase the bandgap of the absorber. Nevertheless, it decreases the grain size which is a negative aspect of sulfurization. Therefore, further research on the sulfurization method is required to find a better solution to eliminate its negative effects on the grain size of the CIGS absorber.

Current studies reveal that unique characteristics of CIGS solar cells such as high photon absorptivity, flexibility, modifiable bandgap, variety of materials composition, low cost, easy manufacturing, and high conversion efficiency make it more interesting for future research and development in the field of photovoltaic technology.

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Biographies



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