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# Review: Advances in the CIGS Thin Films for Photovoltaic Applications

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## Abstract

The copper indium gallium selenium (CIGS) thin film is widely acknowledged as the most promising material for photovoltaic applications. Mainly due to appealing chemical and physical structures properties, low fabrication cost, high efficiency, and uncomplicated integration especially with the advancement in the use of the flexible substrate. Promising results have been achieved in CIGS-based solar cells in the last few years and these devices could be key in unlocking the potential of green energy. Therefore, it is necessary to understand the parameters that are critical to improving the efficiency of these devices. Parameters such as doping concentration, thickness, substrates, and energy bandgap. In this review, we comprehensively report on these parameters with an aim of showing the recent progress on the various methods used to optimize them, all geared towards efficient and low cost solar cells for PV applications.

## Keywords

CIGS, Thin Films, Photovoltaics, Efficiency

## 1. Introduction

With growing worldwide demand for energy and environmental impact associated with conventional sources, projections show a very probable energy crisis shortly. This has led to intense research on efficient sources of green energy like wind energy [1], thermal energy [2] and tidal energy [3], hydropower [4], and solar energy [5]. Trends in the last 35 years show 8-fold growth in the generation and consumption of green energy technologies (see **Figure 1(a)**). Though each one of them has received considerable attention, their applications are still not as
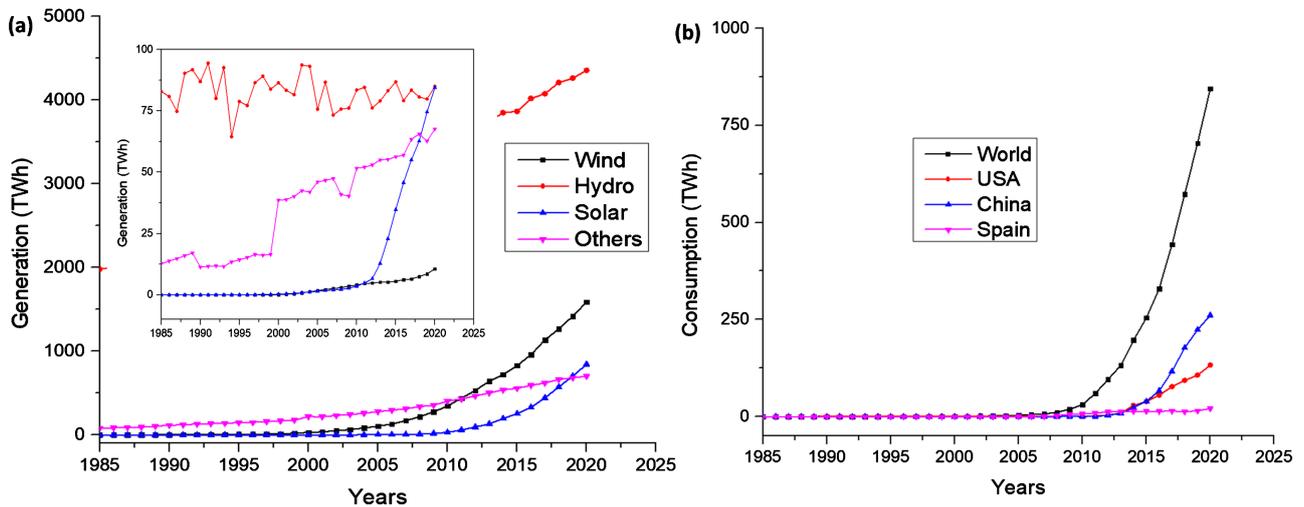
**Figure 1.** (a) Shows the world's share of renewable energy technologies in the last 35 years. Insert shows, the breakdown of these renewable technologies in Japan. (b) Shows the global solar energy consumption and the leading consumer countries [8].

competitive to the fossil sources of energy. For example, although hydropower plants are fundamental to the sustainable growth of renewable energy [4], the increased demand for water and other sources like irrigation may be in direct conflict with the objectives of river conservation. Wind power has been gaining worldwide attention as a large-scale energy source, however, its reliability is a serious challenge due to the intermittent nature of wind power [6]. To this end, the solar energy option offers great potential especially considering that the solar power received on earth is $\sim 10^4$ times larger than humanity's mean consumption [7]. In the last 12 years, the demand for solar energy has soared [8], which calls for intense research in this field to meet this demand.

Solar energy harvesting forms what is referred to as photovoltaic energy harvesting and deals with the direct conversion of photons to electrons being key to meeting the world's demands for clean, sustainable, and abundant energy. In the recent past exponential development of photovoltaic (PV) technologies (polycrystalline thin-film like copper-indium-gallium-diselenide, perovskite solar c ls, silicon solar cells, dye-sensitized solar cells or organic solar cells) has led to significant reduction in the price of solar electricity, making it a potential competitor to the commonly used power sources [9].

The application of PVs however can further be enhanced by having more efficient solar cells. According to the Solar cell efficiency tables (Version 55) [10] the efficiency of solar cells (measured by a recognized test centers) is 26% and 38.8% for single and multiple junctions cells respectively. However, various research groups have reported efficiencies as high as 43% [11]. The key challenge is to not only improve the efficiency of solar cells but also make them affordable. Currently, the bulk semiconductor dominates the market due to their easier processing and manipulation [12]. Among them is the important copper-indium-gallium-diselenide (CIGS) thin film solar cells which are characterized by high absorption coefficient and adjustability of graded band gap for solar spec-

trum [13]-[19].

Expanding applications of CIGS in PV devices demands for the optimization of parameters that are keys to the enhancement of the efficiency of these devices. To fully realize the potential of CIGS for PVs applications parameters such as doping concentration, thickness, substrates and energy band gap must be well understood. In addition to effect of forming CIGS based heterojuctions for enhanced performance. Therefore, this review mainly focuses on recent (mostly within past 10 years) progress on the improving the performance of CIGS based devices by optimizing the parameters.

## 2. CIGS Parameters

1) Thickness

The standard thickness of the CIGS layer is ≈2 μm. Reducing this thickness can be a key in reducing the overall production cost and the materials usage. However reducing the thickness can reduce the amount of light absorbed by the layer and back contact recombination because of electrons being generated near the back coat. Soumaila *et al.* [20] investigated the influence of absorber back surface region grading [20] in CIGS solar cells. To achieve the optimal performance, thickness of the back surface grading layer and the absorber bulk thickness were varied. The results showed that back surface grading greatly improved the performances of CIGS [20] [21] [22]. **Figure 2(a)** shows that the efficiency of the device increased with increasing the absorber bulk thickness *d*. Primarily due to an increased short-circuit current density ($J_{sc}$), which varies by 3.2 mA/cm$^2$ from 29 mA/cm$^2$ when d changed from 0.5 μm - 2 μm (see **Figure 2(b)**). Other parameters such as fill factor (FF) and open-circuit voltage ($V_{oc}$) are also enhanced as d was increased because of increased light absorption and the reduction of the back contact recombination. Increasing the grading height (ΔGGI) is a key in suppressing the back interface recombination. **Figure 2(c)** shows that absorbers with high ΔGGI have relatively longer lifetimes, suggesting a reduction in back interface recombination.

CIGS (In = 0.7, Ga = 0.3) thin films with two thickness 500 nm and 1000 nm were deposited by thermal evaporation technique on glass, silicon and ITO substrates at room temperature [24] and later annealed at three different temperatures. The change of current vs. voltage (I-V) explained the basic properties of the solar cell device. The I-V characteristic of the solar cells showed that the best composition was when the thin film was deposited on glass substrate with thickness of 1000 nm. Besides, doubling the thin film thickness from 500 increased the energy gap from 1.82 eV to 1.87 eV. In general, the carrier concentration and Hall mobility was enhanced with the increased thickness. Serap *et al.* [23] deposited CIGS ultrathin films of different thicknesses (52 nm, 89 nm, 183 nm and 244 nm) which showed a single crystal structure and increased crystal sizes with the increasing thickness. Interestingly, the increased particle size leads to the enhanced absorption but band gaps were decreased. The simulated results
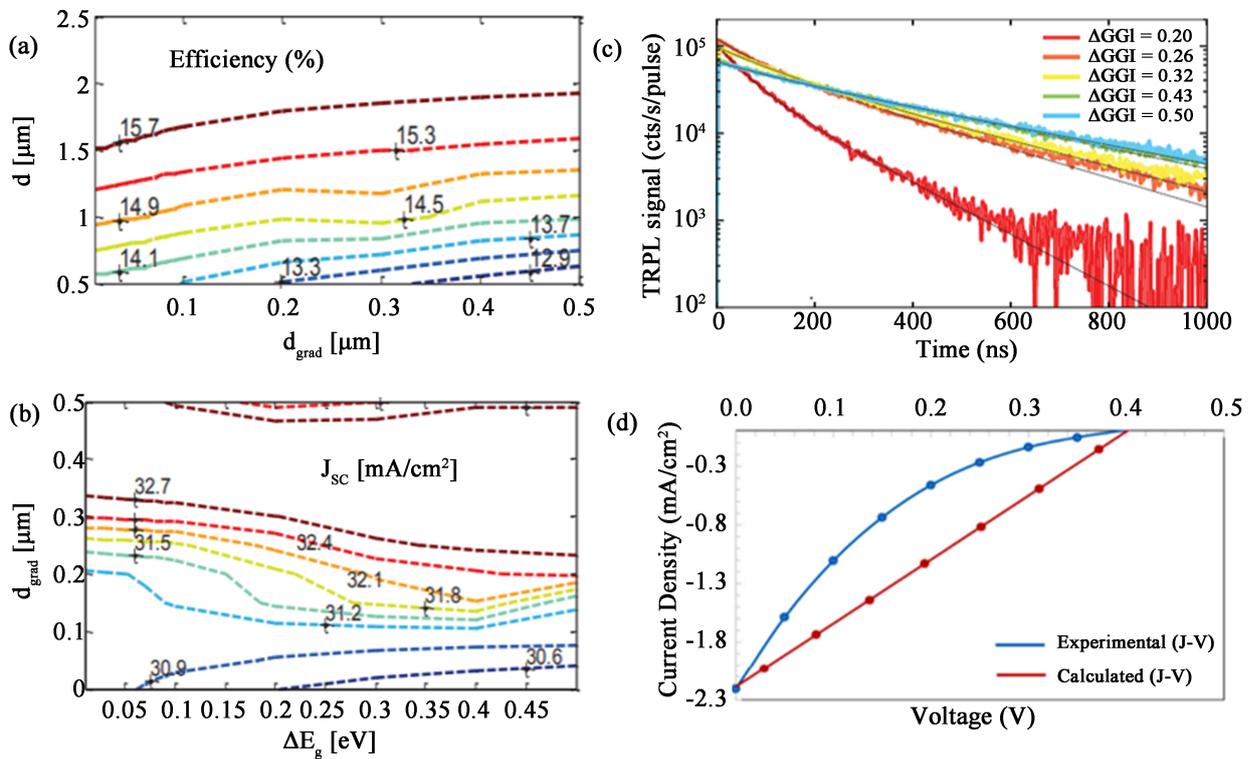
**Figure 2.** The (a) conversion efficiency and (b) short-circuit current density ($J_{sc}$) of the solar cells as function of the thickness of the back surface grading layer d grad and back surface region band-gap variation ($E_g$) [20]. (c) Time-resolved photoluminescence (TRPL) transients of absorbers with different ΔGGI (the black lines are best exponential fits) [21]. (d) J-V characteristic of experiment and simulated CIGS-3/n-Si hetero junction solar cells [23].

for the current density versus voltage show in **Figure 2(d)** a good agreement with the experimental values. Nevertheless, the fill factor varied considerably for the calculated and experimental values, which could be attributed to the loss mechanisms due to factors like ohmic contact resistance and the grain boundary defects.

2) Substrate effect

The substrate in CIGS has a critical role in the development of the whole structure. A rigid Soda-lime glass (SLG) substrate is widely applied in the CIGS thin film sector because of its material properties, which can supply favorable amounts of Na to the absorber during evaporation or selenization process. Solar cells prepared through a multistage process on glass substrates showed high efficiency to the tune of 16.0%. Whereas those on steel sheet substrates without a diffusion barrier showed efficiency levels of 0.2%. Interestingly, changing the process to a single stage enhance the efficiency levels in the steel substrate but reduced it in the glass substrate [25].

The use two steel substrates *i.e.* Stainless chromium (Cr) steel and Cr-free steel sheets [25] was investigated in terms of the diffusion of iron (Fe) and other substrate elements into the CIGS layer by Secondary Ion Mass Spectrometry (SIMS). The influence of the impurities on the solar cell parameters was determined by current voltage (JV) and external quantum efficiency (EQE) mea-

surements. The diffusion of substrate elements into the CIGS layer was found to be more pronounced in the case of unalloyed steel substrates as Compared to Cr steel. This was because Cr oxide layer at the surface of the Cr steel foil acted as a Fe diffusion barrier. Other substrates have also been used, for example polycrystalline $CuInSe_2$ thin films were deposited on a flexible metal foil substrates using the selenization technique. Here, E-beam evaporated Cu-ln precursor layers were reacted with an atmosphere containing $H_2Se$ gas at around 400˚C [26]. Mo, Ti and Al foils were considered as possible substrate materials for $CuInSe_2$ solar cells. From the results, Al was found to be the most reactive of the three foil substrates studied.

An interesting development in the applications of the CIGS solar cells is the advancement of the flexible substrates, which can open new markets for indoor and outdoor applications. For example, flexible modules facilitate easier installation features on integrated photovoltaics with minimal transport, portable consumer devices and automobiles [27]. Good progress has been made on flexible CIGS solar cells especially using the polyimide-based substrates. Generally, flexible substrates can be of three types: metallic foils, polyimide sheets, and Zirconia [14]. Liu *et al.* [28] developed a flexible CIGS solar cell on stainless steel substrates by using Ti/TiN composite structures as the diffusion barrier layer. Despite the several drawbacks of the flexible substrates such as being fragile and heavy. Conversion efficiency of 8.9% of CIGS/SS solar cells with Ti/TiN composite structure were achieved which was close to the 9.1% value of CIGS cells fabricated on soda-lime glass substrates. Higher efficiency of 20.4% on a polyimide film has been reported giving optimism on the ability of these flexible substrates based solar cells matching the efficiency of solar cells on rigid substrates [29] [30].

3) Effects of temperature

Varying temperatures during CIGS thin film deposition can be used to determine the effect of thin film growth in relation to solar cell efficiency. Stuckelberger *et al.* [31] investigated the complex temperature dependence of defects and voltage in $CuIn_xGa_{1-x}Se2$ thin solar cells. The growth temperature ranged from room temperature to 100˚C and the thin films were deposited by Mia-Sole on flexible stainless steel substrates. They concluded that a crucial understanding of light-induced and heat-induced metastabilities at the microscale is vital in relation to the overall module performance especially the efficiency values. In addition, the deposition of polycrystalline CIGS thin films onto Mo-coated soda-lime glass substrates using the three-stage co-evaporated process was done [32]. And, carried out at the substrate temperature ($T_{sub}$) varying from 350˚C to 550˚C coupled with independent control Cu, In, Ga and Se sources show that that the cell efficiency increased with the increase in growth temperature. Interestingly, the simulated results revealed that the solar cell performances depended on the increase in operating temperatures for all CIGS semiconductor with varying temperature gradients in comparison to the experimental values.

To further enhance the efficiency of the solar cell device, varying substrate temperature can be coupled with the use of a buffer layer. For example, zinc sulfide (ZnS) thin films were sputtered under various substrate temperatures ranging from 100˚C to 400˚C. The results revealed that the sputtered ZnS thin films with an optimized substrate temperature of 100˚C could serve as a buffer layer for heterojunction thin film solar cells with quaternary compound semiconductor-based absorber layers [33]. Another study used cadmium sulphide (CdS) buffer layer, and showed that the solar cell performance is affected by the operating temperature [34].

4) Effects of post-selenization and use of a precursor

The quality of CIGS thin films can be improved by deposition of the precursors followed by post- selenization which in the process improves the cell efficiency. Post-selenization of copper, gallium and indium precursors to fabricate $CuIn_xGa_{1-x}Se_2$ (CIGS) thin films can be achieved by the use of Se vapour, diethylselenide or $H_2Se$ gas. Using Cu–In–Ga precursors and $H_2Se$ gas Cu(In1_x-Gax)Se2 (CIGS) thin films fabricated [35]. To improve the optoelectronic properties, a high temperature selenization and in situ annealing process was conducted. Morphological and crystal characterization showed that the films had large grain size and with improved crystallinity. Conversely, sputtering of CuIn-Ga precursors followed by chalcogenization was done to fabricate CIGS thin films. Two stage selenization processes were employed and then the microstructural characteristics of CIGS films studied [36]. The selenization temperature for the two processes was varied between 450˚C and 580˚C to establish the relationship between the microstructural characteristics and compositions of the CIGS films. From the results, the CIGS thin films formed using isothermal selenization were found to have dense grain structure whose grains increased in size after an increase in the selenized temperature. However, the Se/(Cu + In + Ga) ratios of the films indicated that Se was distributed non-uniformly in the films.

Further investigation employed a rapid thermal process of stacked elemental layers. Here, the properties of the Cu,Ga and In layers deposited by DC-sputtering were studied [37]. By varying the thickness ratio of the In/CuGa layer, the chemical compositions of the metallic precursor were optimized. The optimized precursor was then selenized under various temperatures after which the performance of the fabricated CIGS solar cells could be investigated and analyzed. The experimental results showed that the performance of the CIGS solar cells enhanced at higher selenization temperatures. The use off non-vacuum coating techniques for CIGS thin films is an interesting thing and many efforts have been made to develop for solar cell applications. The approach may either use solution type precursors or particle-based precursors [38]. For instant, Gas flow sputtering of CIGS with slightly Cu-poor stoichiometry was performed with two opposing CIGS targets i.e selenium only provided by target and additional selenium from an elemental source inside the sputtering system [39]. From the results, the thin films deposited without extra selenium produced cells of efficiency

2% while films with additional selenium produced cells of up to 12% efficiency. These results proved that extra supply of selenium during sputtering improves efficiency of solar cell.

Even at nano-scale, post-selenization and use of a precursor remains key parameters ffor improving the efficiency of the CIGS based solar cells. For example, CIGS nanoparticles with uniform size of 15 nm in diameter and a chemical composition of $Cu_{0.9}In_{0.64}Ga_{0.23}Se_{2.00}$ were synthesized using spray pyrolysis technique. Selenization of the spray deposited CIGS nanoparticles was conducted in a homemade two-zone rapid thermal annealing_RTA_ furnace using Se pellets as a source of Se Vapor. The study revealed that achievement of higher Se supply to CIGS nanoparticles was as a result of either increasing Se evaporation temperature or by increasing the flow rate of carrier gas resulting to larger CIGS grains with higher degree of crystallinity [38].

Varying of annealing temperatures is known to enhance the crystal growth in CIGS thin films. Zhang *et al.* [40] prepared CIGS thin films by directly sputtering a CIGS quaternary target consisting of Cu:In:Ga:Se = 25:17.5:7.5:50 at %. Structural and composition properties of the CIGS thin films were explored after annealing at a temperature of 550˚C under vacuum and Se-containing atmosphere [41]. Recrystallization of CIGS thin films was found to occur with a preferred orientation in the (112) direction. The results proved that the CIGS thin films annealed under vacuum exhibited a portion loss of Se while those annealed under Se containing atmosphere revealed compensation of Se. Another study to check on the structural, morphological, optical and electrical properties of electrodeposited CIGS thin films were also investigated by considering two different annealing atmospheres such as vacuum and $N^2$ + Se vapor. The films were electrodeposited from an aqueous-based solution at room temperature in a three electrode cell configuration whereby platinum plate was used as the counter electrode and a glass/ITO substrate as the working electrode [42]. The results revealed that the crystal structure remained the same for all the samples but the grain size varied with the variation of the annealing atmosphere leading to the conclusion that the annealing process had a great influence on grain nucleation and growth.

In addition, CIGS thin films were prepared onto different substrates by thermal evaporation technique in a high vacuum system of $(10^{-5})$ torr [43]. The deposited thin films were then annealed at (100, 200 and 300)˚C temperature. The structural and optical properties of the deposited thin films revealed that the surface morphology and optical band gap increased with the annealing temperature. Besides, annealing in selenium-free atmosphere show that the grain growth of CIGS films was enhanced with the increase of the annealing temperature ranging from 450˚C to 525˚C [40]. Another approach is to incorporate the copper into indium selenide thin-films. Here, an ion-exchange reaction was used to incorporate Cu ions from aqueous solution into indium selenide thin-films. A precursor structure was prepared for conversion into $CuInSe_2$ (CIS) layers suita-

ble for solar cell processing [44]. This process resulted to thin films with a graded compositional depth-profile containing the crystalline phases $\beta$-Cu$_{2-x}$ Se and $\gamma$-In$_2$Se$_3$. Annealing of these layers in the presence of Se vapour showed to homogenise the compositional depth-profile of the layers and to form chalcopyrite CIS. CIGS absorber layers were deposited by multi-stage evaporation process at a substrate temperature below 500°C on Mo coated soda-lime glasses [45]. Sodium fluoride post deposition treatment was applied on CIGS layer during cool-down of the substrate. The results showed that the microstructure of CIGS composition depth GGI composition profiles changed depending on the [Se/metal] conditions.

5) Doping effect

Doping concentration of different layers plays an important role in relation to maximizing the efficiency and minimizing the fabrication cost of any solar cell. A theoretical study of a CIGS thin film solar cell which produced a maximum efficiency of 24.27% [46] shows that the optimized efficiency can be obtained by determining the optimum band gap of the absorber and varying the doping concentration of constituent layers. The Ga content denoted by x = Ga/(In + Ga) was selected as 0.35 which provided the optimum band gap of absorber layer as 1.21 eV. A one-dimensional simulator ADEPT/F 2.1 was used to analyze the fabricated device parameters. The simulation results revealed that the efficiency increased with increase in band gap. It was noted that after certain band gap level (1.21 eV), the efficiency decreases despite an increase in the band gap. A proof that CIGS cell suffers from lattice mismatch effect for the Ga/(In + Ga) ratio is above 0.35. A study on the doping concentration in CIGS thin-film solar cells, done at each layer of the cell with an aim to obtain the optimum doping concentration was done using ADEPT 2.0, a 1D simulation software. The results revealed that energy conversion efficiency decreases with increasing doping concentration in the window layer (ZnO) and the buffer layer (CdS). Conversely, it was observed that cell efficiency increases with increasing doping level in the absorption layer (CIGS). Therefore it can be concluded that doping concentration play significant role on the performances of the CIGS solar cell (ZnO/CdS/CIGS) structures [47].

Sodium is another interesting candidate for doping. Ideally, CIGS absorbers have the following shortcomings; including poor crystallinity, large porosity, and rough surfaces, which result in lower power conversion efficiency as compared to vacuum-based CIGS solar cells. Therefore, promoting absorber grain growth is fundamental to enhancing the performance of these devices especially the solution-based solar cell. The use of Sodium which is alkali based has been shown to improve the grain growth and enhance the absorbing ability of the CIGS layer [48]. Specifically it leads to morphological changes leading to improved carrier collection and minority carrier lifetimes. Another way of doping is the use of Cs-PDT although the mechanism of doping remains controversial. The entry of Cs makes more Na enter the absorber besides, the Cs atoms entering the film not

only locate at the grain boundary but also enter the grains. Implying that they could passivate the defects both at the grain boundary and grain interior, improving the hole carrier concentration and minority carrier lifetime [49].

## 3. Conclusion

From the above discussion, it shows that CIGS quaternary compound is good candidate for use as an absorber layer in high efficiency thin films solar cells. However, the efficiency of the CIGS thin films solar cells is dependent on the following factors: substrate and growth temperature, deposition techniques and the stoichiometry composition. Unlike the other compounds, in thin films of the alloy CIGS copper, indium, and gallium typically redistribute during growth to create composition profiles in the final layers completely different from their initial distribution. Therefore, it is necessary to discuss the essential materials such as gallium and its impact on the structural properties of CIGS solar cells. This provides a better understanding of the relationship between the emitter and absorber bulk in relation to electronic fields, carrier transport, and recombination processes that determine device performance. In this review, we have discussed these parameters with an aim of showing the recent progress on the various methods used to optimize them, all geared towards efficient and low cost solar cells for PV applications.

## Acknowledgments

## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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