

Review Article

Review on Substrate and Molybdenum Back Contact in CIGS Thin Film Solar Cell

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Copper Indium Gallium Selenide- (CIGS-) based solar cells have become one of the most promising candidates among the thin film technologies for solar power generation. The current record efficiency of CIGS has reached 22.6% which is comparable to the crystalline silicon- (c-Si-) based solar cells. However, material properties and efficiency on small area devices are crucial aspects to be considered before manufacturing into large scale. The process for each layer of the CIGS solar cells, including the type of substrate used and deposition condition for the molybdenum back contact, will give a direct impact to the efficiency of the fabricated device. In this paper, brief introduction on the production, efficiency, etc. of a-Si, CdTe, and CIGS thin film solar cells and c-Si solar cells are first reviewed, followed by the recent progress of substrates. Different deposition techniques' influence on the properties of molybdenum back contact for CIGS are discussed. Then, the formation and thickness influence factors of the interfacial MoSe₂ layer are reviewed; its role in forming ohmic contact, possible detrimental effects, and characterization of the barrier layers are specified. Scale-up challenges/issues of CIGS module production are also presented to give an insight into commercializing CIGS solar cells.

1. Introduction

Copper Indium Selenide (CuInSe₂ or CIS) is a ternary compound p-type absorber material belonging to the I-III-VI₂ family [1]. The very first CIS material being synthesized was in 1953, and then, an efficiency of 12% has been reported for single crystal CuInSe₂-based solar cells [2]. In 1976, the first CIS thin film solar cell with buffer layer CdS was fabricated with an efficiency of 4–5% by evaporating CuInSe₂ powder in the presence of excess Se vapor (coevaporation) [3]. CIG-based thin film solar cell started to receive even more attention in 1981 when Mickelsen and Chen achieved an efficiency of 9.4% by using coevaporation technique from elemental sources [4]. From that onwards, numbers of emerged technologies such as alloying CIS with gallium

(Ga) to become Copper Indium Gallium Selenide (CIGS), incorporating sodium (Na) into the CIGS absorber layer, and replacing thick cadmium sulfide (CdS) buffer layer with thin CdS layer have boosted the efficiency significantly. Copper Indium Gallium Selenide (CIGS) thin film solar cell currently holds a record efficiency of 22.6% since 2016 [5]. To accomplish the record efficiency, research institute ZSW has developed a new method to deposit a layer of potassium fluoride (KF) between CIGS and buffer layer (CdS) to improve the electrical properties of the solar cells, in particular the V_{oc} .

The cell structure of CIGS is known as substrate configuration where the light enters the cell through Transparent Conducting Oxide (TCO), passes through the buffer layer, is absorbed by the CIGS, and then reaches the

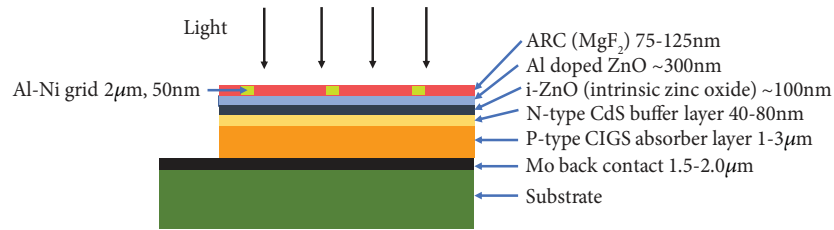


FIGURE 1: Structure of CIGS thin film solar cell.

back contact, usually molybdenum, which is deposited on the substrate. The typical structure of CIGS solar cell consisting substrate/Mo/p-type CIGS/n-type CdS/i-ZnO/ZnO:Al/ARC/metal-grid is shown in Figure 1 [1]. The reason CIGS has been one of the most promising absorber layer for thin film photovoltaic devices is due to its high absorption coefficient for solar radiation and compatibility of its bandgap (1.6 eV–1.0 eV) [6]. The advantages of CIGS-based solar cells over CIS-based solar cells are as follows: (i) the bandgap can be tuned by adjusting the Ga/In ratio to match the solar spectrum. If all indium (In) is replaced by gallium (Ga), the CIGS bandgap increases from about 1.04 eV to 1.68 eV [7]. It has been stated that CIGS absorber layer can absorb most parts of the solar spectrum with a thickness of $1\ \mu\text{m}$ [1]. Hence, a layer thickness of $\sim 2.0\text{--}2.5\ \mu\text{m}$ will be sufficient for the completed device, and a thinner layer device means reduction in raw material usage and lower production cost incurred. (ii) Ga incorporation can also improve the open-circuit voltage V_{oc} of CIGS since $V_{oc} \sim E_g/2$ (E_g is referring to bandgap) [1].

Moreover, CIGS thin film solar cell has very high potential to overcome the cost level of conventional PV crystalline silicon (c-Si) technology [8]. The c-Si modules with efficiencies of 19–23% will have a production cost of $\$0.6\text{--}\$0.7/\text{Wp}$ [9]. Whereas for CIGS modules, manufacturing cost of $\$0.75/\text{Wp}$ can be achieved at 50 MW/yr production capacity with an average efficiency of 12% [10].

The substrate in CIGS has a crucial role in the development of the whole device. Deposition of the molybdenum back contact on rigid or flexible substrate will define the selenization condition. It has to be considered that flexible substrate cannot withstand high process temperature over 500°C , but high process temperature is required in crystallizing the CIGS absorber layer. Lower temperature processes have to be developed when dealing with flexible materials. Recently, an efficiency as high as 20.4% was achieved on flexible polymer substrate [11]. This has revealed that the flexible solar cells with performance close to rigid solar cells can be developed.

The following layer in CIGS after substrate is the molybdenum (Mo) back contact which acts as an optical reflector to reflect the light back to the absorber layer in CIGS solar cell [12, 13]. Molybdenum (Mo) is a preferred back contact material for CIGS solar cells because it does not react strongly with CIGS; it forms low-resistivity ohmic contact to CIGS, and the conductivity of Mo does not degrade during deposition of CIGS at high substrate temperature [14–17]. Mo has high conductivity and is more chemically stable

and mechanically stable during CIGS growth (selenization) than other materials such as W, Ta, Nb, Cr, V, Ti, and Mn [18–23].

The layer after Mo back contact is Copper Indium Gallium (CIG) before going through the process of selenization. During the selenization process, selenium (Se) vapor will react with CIG to become CIGS and react with Mo to form the MoSe_2 layer. This interfacial layer between Mo and CIGS is beneficial in terms of having a wider bandgap (1.35–1.41 eV) than CIGS, hence it can absorb more near-infrared light to improve the cell performance [24]. The formation of MoSe_2 layer does not depend only on the selenization condition but also on the properties of the Mo film [25, 26]. Therefore, improving the properties of the Mo film can promote the growth of MoSe_2 layer. Recently, substrates used in CIGS, either rigid or flexible, together with the properties of the Mo back contact and the MoSe_2 interface layer were discussed in various papers [8, 25, 27, 28]. This paper aims to focus on the mentioned area by first providing an overview of comparison between conventional PV and thin film solar cells followed by reporting the recent progress of the substrates in CIGS, specifically regarding the available substrates. This paper will then converge towards the Mo layer in CIGS and further discuss about the deposition techniques and effect of deposition condition on the properties of Mo back contact. Then, the formation and thickness influence factors of the interfacial MoSe_2 layer will be reviewed in this paper. Scale-up issues of CIGS module production will also be presented to give an insight into commercializing CIGS solar cells.

2. Brief Introduction on the Production, Efficiency, etc. of a-Si, CdTe, and CIGS Thin Film Solar Cells and c-Si Solar Cells

Global production of photovoltaics (PV) has been expanding drastically in the past decades, moving from 202 MW in 1999, 17 GW in 2010, until the recent production over 78 GW in 2016 [29–31]. About 92% of the commercial modules are made from Si while thin film modules contributed 8% in the market share of which <1% for amorphous silicon (a-Si) modules, 5% for Cadmium Telluride (CdTe) modules, and 2% for CIGS modules [32, 33]. Considering the 8% market share in 78 GW global PV production, the total annual production for thin film solar cell will be ~ 10.26 GW. The estimated total energy world consumption in years 2050 and 2100 are 28 and 46 TW, respectively [34].

Looking at the current PV technologies with an average growth rate of 30–40% per year, significant fraction of the future world energy demand can be satisfied [35]. The time where thin film PV technologies started to grow rapidly was during the silicon feedstock shortage that happened back in mid 2000s [30]. This scenario has caused the price of the conventional PV module to be increased and thus opened up an opportunity for the researchers and investors to explore further in the thin film technologies. According to the latest research cell record efficiency chart reported by the National Renewable Energy Laboratory (NREL), crystalline silicon (c-Si) has a record efficiency (lab scale $2 \times 2 \text{ cm}^2$) of 25.7% and 24.4% efficiency for commercially available module [36, 37]. For thin film solar cell, amorphous silicon (a-Si), CdTe, and CIGS, each cell respectively has a record efficiency (lab scale) of 14%, 22.1%, and 22.6%; at the same time, the best module efficiency for each cell falls at 12.3%, 18.6%, and 15.7% [36, 38, 39].

Crystalline silicon (c-Si) currently plays a major part in thin film energy production with the highest module efficiency. Thick and rigid c-Si wafer ($180 \mu\text{m}$) is required for a good absorber (high absorption) in module production [1]. This is because silicon (Si) is a poor absorber due to its indirect bandgap nature and its low absorption coefficient (10^4 cm^{-1}) [40]. Whereas for CIGS, its absorption coefficient is beyond 10^5 cm^{-1} making the thickness of CIGS to be 100 times less than the thickness of c-Si wafer. In addition, the module production of CIGS solar cell requires a lower thermal budget ($\sim 550^\circ\text{C}$) than the c-Si solar cell ($\sim 1100^\circ\text{C}$) [1, 41]. CIGS solar cells provide an alternative to Si solar cells, and it is highly competitive as less raw material, time, and cost are involved in module production. On the basis of material, less material usage will lead to lower manufacturing cost for CIGS solar cell and hence inducing a shorter energy payback time (~ 1 year) as compared to the c-Si solar cell (~ 2 years) [10, 42]. Aside from the conventional c-Si cells, there are also other commercially available thin film modules in the market, including amorphous silicon (a-Si), Cadmium Telluride (CdTe), and, as mentioned, Copper Indium Gallium Selenide (CIGS).

Amorphous silicon (a-Si) has a direct bandgap and only uses 1% of the material (Si) needed for crystalline silicon cells production [43]. a-Si module can be made flexible and lightweight which later enable various possibilities when it comes to application such as mounting on uneven surface, incorporating into small devices, and being portable. One of the a-Si solar cell advantages is the high actual power output in hot climate by having a low temperature coefficient [44, 45]. However, a-Si solar cells have difficulty in the solar market because the price of conventional PV (c-Si) has been decreasing dramatically [46]. The main issue in a-Si solar cell technology is having a low conversion efficiency. The 14% stabilized research efficiency (by National Institute of Advanced Industrial Science and Technology, AIST) and 12.3% module efficiency have clearly shown its limitation to compete in the PV sector. Until now, a-Si technology is matured and commonly being used in the application of consumer products (e.g., calculators, watches, and other noncritical outdoor applications) [43].

Cadmium Telluride (CdTe) currently holds the highest module efficiency at 18.6% due to one of its advantages of nearly ideal bandgap (1.45 eV) for solar terrestrial photo-conversion [36, 47]. The module production status of CdTe is currently ahead of a-Si and CIGS thin film solar cell as it can be produced at a cost of $\$0.75/\text{Wp}$ (watt peak), and the production cost at year 2020 is expected to be around $\text{€}0.5/\text{Wp}$ [48, 49]. Amorphous silicon (a-Si) module can be produced at $\text{€}0.75/\text{Wp}$ but in a lower efficiency ($\sim 9\%$) while CIGS is still trying to lower its production cost to below $\text{€}1/\text{Wp}$ [49]. Cadmium Telluride (CdTe) is a favourable technology in terms of prospects in PV sector except the public has raised concern about the core material used in CdTe, cadmium, which is extremely toxic. It has been stated that cadmium forms a very stable compound with tellurium and hence is not banned as a hazardous substance, but before forming a compound, cadmium itself can lead to a variety of adverse health effects including cancer [50]. More advance technology is needed to handle the cadmium and hence creating another challenge for CdTe in lowering the production cost.

Copper Indium Gallium Selenide (CIGS) solar cell is one of the best thin film candidate to look into because its lab scale efficiency (22.6%) has just surpassed CdTe's lab scale efficiency (22.1%) based on the latest research cell record efficiency chart reported by NREL [37]. For CIGS solar cells, a large efficiency gap occurred between the lab scale cells (22.6%) and commercially available modules (15.7%). Although the CIGS module production presently lags behind of CdTe module due to process complexity, nonetheless, CIGS technologies still have a higher efficiency potential versus CdTe, and this can potentially make them more cost-effective than CdTe solar cells [1, 48]. The theoretical efficiency limit for CIGS bandgap of 1.14 eV is 33.5% [51]. According to the CIGS current record efficiency (22.6%), cell efficiency as high as 25% can be reached in the near future. In terms of module efficiency, CIGS (15.7%) is definitely not far behind from CdTe (18.6%) (Table 1).

3. Recent Progress of Substrates

Soda-lime glass (SLG) is a type of rigid substrate being used widely in the CIGS thin film industry due to its material properties which can supply sufficient amount of Na to the absorber during coevaporation or selenization process [53–56]. The supply of sodium (Na) at 0.1 at% (atomic percentage) is reported to be beneficial for CIGS solar cells in terms of increasing the open-circuit voltage and fill factor that lead to an enhancement in solar cell efficiency [53, 57]. The improvement in device efficiency is mainly due to sodium (Na) that passivates the defects at the cadmium sulfide (CdS) and CIGS junction [58–60]. In addition, SLG meets most of the requirements needed such as good adhesion, low weight, and able to work on suitable temperature. SLG substrate also has an optimal coefficient of thermal expansion (CTE) for CIGS which is desired between 5×10^{-6} and $12 \times 10^{-6} \text{ K}^{-1}$ to avoid adhesion problem or crack formation during deposition of CIGS at high temperature [27]. Solar cell company like Solibro has achieved a

TABLE 1: Brief introduction on the production, efficiency, etc. of conventional PV (c-Si) and thin film solar cells (a-Si, CdTe, and CIGS) [10, 32, 33, 36, 37, 40, 42, 51, 52].

	Conventional PV		Thin film solar cells	
	c-Si	a-Si	CdTe	CIGS
Best research cell efficiency	25.7% [37]	14% [37]	22.1% [37]	22.6% [37]
Best module efficiency	24.4% [36]	12.3% [36]	18.6% [36]	15.7% [36]
Theoretical efficiency limit	29.43% [52]	20% [51]	32.8% [51]	33.5% [51]
Absorption coefficient	10^4 cm^{-1} [40]	$(5 \times 10^4) \text{ cm}^{-1}$ [40]	10^5 cm^{-1} [40]	$>10^5 \text{ cm}^{-1}$ [40]
Current PV market share	92% [32]	<1% [32]	5% [32]	2% [32]
Annual production	~71.76 GW	~0.78 GW	~3.9 GW	~1.56 GW
Energy payback time	~2 years [42]	~1.5 years [42]	~7 months [42]	~1 year [42]
Major manufacturer	Jinko Solar [33]	Sharp [10]	First Solar [10]	Solar Frontier [10]

current record efficiency of 21% (single-junction terrestrial cell) and 18.7% on minimodule using SLG as a substrate [61, 62]. The first time ever, in 2010, the substrate involved in pushing the efficiency of CIGS beyond 20% was SLG [63]. Other than the standard glass SLG, specialty glasses have been explored by several research groups which mainly focus on high-temperature glasses to avoid softening of the substrate (SLG) during CIGS absorber deposition [64, 65]. High temperature condition will not only enhance the growth of absorber layer but also favour alkali-diffusion from substrate to absorbers. Thus, specialty glass serves as a medium to provide fine control of alkali-diffusion without softening at high temperature [66, 67]. The CIGS world record efficiency back in 2014, 21.7%, and current world record efficiency since 2016, 22.6%, were both achieved by the same research group ZSW on a specialty alkali-aluminosilicate glass that incorporate Na during the CIGS growth process [5, 61, 68].

In CIGS solar cells, the most commonly used flexible substrates are metals and polymers [27, 60]. The essential aspects to be considered for a suitable flexible substrate for CIGS films are dependent on different physical and chemical properties, such as thermal stability, vacuum compatibility, suitable coefficient of thermal expansion (CTE), humidity barrier function, chemical inertness, and surface smoothness [57]. Metals are able to withstand very high deposition temperature but they possess a rather high roughness, density, and CTE especially Al and Cu [27]. Furthermore, most of the metals like steel contains metallic impurities (Fe and Cr) that affect the device performance [27, 69, 70]. Therefore, a metal oxide barrier is used in order to provide electrical insulation between the substrate and the Mo back contact at the same time serves as a diffusion barrier against impurities from the metal substrate [27, 69, 71, 72]. Typical examples of barrier layer materials are Al_2O_3 and SiO_x [73–75]. It has been announced that the highest cell efficiencies reached so far using stainless steel (SS) and titanium (Ti) foil as substrate are 17.7% and 17.9% [71, 76, 77]. Whereas the maximum cell efficiency (area of 0.5cm^2 with antireflective coating) achieved so far on an enamelled steel is 18.6% by ZSW research group [78]. These results prove the potential of metal as an alternative substrate to rigid glass.

Polymers as substrates have a much lower density, roughness than metals [27]. It allows direct monolithic integration

of solar cells and roll-to-roll deposition process that can reduce manufacturing cost [27, 60]. Other than that, polymer has a high power-to-weight ratio and excellent radiation-hardness which makes polymer an ideal candidate for space application, but polymers cannot sustain high temperature of 550–660°C due to their limited thermal stability [79, 80]. Thus, low process temperature is required, and this generally leads to deterioration of absorber quality [81, 82]. Polyimide films are one of the few polymer films that can sustain temperature close to or above 450°C but not more than 500°C for a short period of time, but this type of polymer has high CTE and it varies depending on suppliers [27, 60]. Despite the challenges faced in using polymer foils as a substrate, the Swiss Federal Laboratories for Material Science and Technology (Empa) has successfully developed thin film solar cells, CIGS, on flexible polyimide foil with an efficiency of 20.4% [83]. The Empa research group was able to modify the properties of the CIGS layer so that it can be grown at low temperature without compensating the light absorption of the CIGS layer which contributes to the photo-current in solar cells [11]. Since 2010 until 2016, ceramics have also been used as flexible substrate [75, 84]. A submodule efficiency of 15.9% has been achieved by AIST in Japan using flexible zirconia-based ceramic sheet as a substrate [75]. Ceramic substrate is able to withstand higher temperature than the soda-lime glass and polymers due to its higher chemical, mechanical, and thermal stability and low porosity [84, 85]. However, its brittle behaviour might be an issue for industrial production on large scale (Table 2).

4. Different Deposition Techniques' Influence on the Properties of Molybdenum Back Contact for CIGS

Current deposition techniques available in the thin film industry for Mo are Ion-beam sputtering, Direct Current (DC) sputtering, Radio Frequency (RF) sputtering, and also High-target-utilization sputtering (HiTUS) [25, 86–88]. The common techniques being used are DC and RF sputtering as they have been discussed profoundly in several papers [12, 86]. Since the characteristics of Mo thin film depend strongly on deposition method and deposition parameters, therefore, a comparison has been made to investigate the

TABLE 2: Summary of current record efficiency on different substrates (CIGS solar cell) [5, 11, 61, 62, 71, 75, 76, 78].

Substrate	Current record efficiency	Description
<i>Rigid</i>		
Soda-lime glass	21% [61]	Solibro (single-junction terrestrial cell) [61]
	18.7% [62]	Solibro (minimodule) [62]
Alkali-aluminosilicate glass	22.6% [5]	ZSW [5]
<i>Flexible</i>		
Titanium	17.9% [76]	Aoyama Gakuin University, Japan [76]
Stainless steel	17.7% [71]	Empa [71]
Enamelled steel	18.7% [78]	ZSW [78]
Polymer	20.4% [11]	Empa [11]
Zirconia-based ceramic sheet	15.9% [75]	AIST (17-cell-integrated submodules) [75]

properties of Mo layers based on DC and RF sputtering techniques. DC sputtering is a well-established industrial process with high throughput capability and requires a cheaper set up cost, whereas RF sputtering deposits a more reflective Mo thin films but with added expense on RF power supplies and impedance matching networks [12, 14, 18, 88, 89]. DC sputtering technique has higher deposition rate than the RF sputtering technique, and DC sputtered films possess good uniformity and adhesion properties over large surface areas [24]. Low deposition rate that arises in RF sputtering technique is due to the number of cycles involved during deposition where RF sputtering only deposits in the second cycle of the AC supply. However, RF sputtered films are found to be more conductive with improvements in open-circuit voltage (V_{oc}) and short circuit current (I_{sc}) [90].

A desirable Mo back contact for CIGS solar cells is addressed to conductive, stress-free, well-adherent, uniform, and crystalline molybdenum (Mo) thin films with preferred orientation (110) on large area glass substrates [91]. By altering the deposition condition on discharge power, working pressure, substrate temperature, and target-to-substrate distance, the desired properties of Mo back contact in terms of physical, optical, and electrical can be obtained [20, 91, 92]. In this paper, the properties of the Mo back contact on both DC and RF sputtering will be further reviewed. The focus deposition parameters will be on discharge power and working pressure, as these parameters have major effect on the properties of the DC and RF sputtered films.

In DC sputtering at low working pressure, surface with dense microstructures is observed while surface with loosened microstructures is observed at high working pressure [18, 55]. At low working pressure, atoms that obtained high kinetic energy are able to travel and bombard onto the substrate surface to form a compact Mo layer, whereas at high working pressure, insufficient kinetic energy has caused the number of atom bombardment towards the substrate surface to reduce, thus resulting in porous microstructure. The high-pressure Mo with loosened microstructure will eventually lead to greater inclusion of impurities such as oxygen that can increase the resistivity of the Mo film. The direct effect of working pressure of RF sputtering on Mo microstructure has not been reported yet, but low RF power was stated necessary to minimize stress and obtain a compact

Mo microstructure despite high RF power deposits good quality of Mo [93]. The Mo deposited with high RF power caused delamination of the absorber layer after the selenization process, and this can be due to the presence of microstresses that existed on the Mo layer.

Another physical property to evaluate is the grain size of Mo. The trend of grain size is found to increase by increasing sputtering power and decreasing sputtering pressure in both DC and RF sputtering Mo films [88, 94–96]. In high power and low working pressure, the increase of Mo grain size causes space between grains to reduce and thus correlates well to the formation of densely packed Mo microstructure. Additionally, along all applied pressures, the grain size of DC sputtered Mo film is always larger than the RF sputtered Mo film [97]. One possible way to explain the formation of large grain size is probably higher power will induce higher flux, and high deposition rate of DC sputtering tends to increase the probability of the Mo particles to nucleate with each other.

Crystal structure of sputtered Mo back contact also plays a fundamental role in Mo quality determination. The lower the working gas pressure, the better the crystallinity of the DC sputtered film [88]. At lower working gas pressure (higher gas power), the atoms gain higher energy due to lesser scattering, and the atoms will then impact the substrate surface with sufficient energy which enhance the atoms mobility in order to facilitate atom diffusion and microvoid fill up, thus creating a conducive requirement for large grain growth and better crystallinity. The same crystallite behaviour was observed in RF sputtered films at low working pressure, but the degree of crystallization appeared to be lower as compared to the DC sputtered films [95, 97]. This can be related to the deposition rate of both RF and DC sputtering technique. Lower deposition rate of RF sputtering generally requires longer deposition time to achieve the same thickness as DC sputtered films, and within the deposition period, impurities such as oxygen can be introduced which can potentially restrain the process of atom diffusion and deteriorates the crystallite property of Mo film.

Surface morphology is one of the properties to be investigated, as it will affect the adhesion, optical, and electrical properties of the solar cell [20]. The surface roughness of DC and RF sputtered Mo films were found to be increased

TABLE 3: Effect of DC and RF sputtering techniques on molybdenum properties for CIGS [12, 86, 95, 97].

DC sputtering	Mo properties	RF sputtering
Larger grain size [97]	Grain size	
	Optical reflectance	Higher optical reflectance [86]
Better crystallization [95]	Crystallization	
Better surface morphology [97]	Surface morphology	
	Resistivity	Lower resistivity [12]

as the working pressure increased [24, 95]. At lower working pressure, the average roughness was due to large grain size formed under high kinetic energy gained by the atoms whereas the roughness at higher working pressure was attributed to loosened microstructure with voids formed under low energy excitation. Although both DC and RF sputtering portray similar surface roughness trend, RF sputtered films possess smoother surface morphology than the DC sputtered films at same working pressure [86]. Smoother morphology also indicates less void formation and porosity effect which contribute to a higher optical reflection in the Mo film; hence, RF sputtered films are more reflective than DC sputtered films as proven in several literatures [93, 98]. In other words, Mo films deposited under low working pressure will have higher optical reflection and conductivity due to densely packed microstructure and large grain size formed (lesser void formation yield to reduced incorporation of foreign atoms).

Furthermore, sputtering pressure has a major influence on the sheet resistance of DC and RF sputtered Mo films over sputtering power [12, 88]. As the working pressure increases, the sheet resistance increases and vice versa, whereas, the sheet resistivity of the Mo film is inversely proportional to the sputtering power [24]. The DC sputtered films also have higher resistivity as compared to the RF sputtered one in all applied working pressures [86]. This phenomenon is interrelated to the microstructure and surface morphology properties. As DC sputtered Mo films are rougher, it verifies the formation of larger voids (more porous microstructure) along the grain boundary, and this will allow impurities such as oxygen to occupy the voids resulting in an increase in resistivity. However, low-resistivity Mo films deposited under low working pressure often gets delaminated, and Mo deposited at high working pressure has better adhesion but less conductive [24]. Mo back contact that exhibits low resistivity and adhesive properties are challenging to simultaneously achieve in single Mo layer [20]. Thus, Mo multilayers like bilayer structure are usually applied to deposit a well-adhered and conductive Mo layer [99–101]. This was done by depositing the first bottom layer with high working pressure (to achieve better adhesion) and the second top layer at lower working pressure (to achieve lower resistivity).

Besides that, single Mo layer can also cause excessive tensile or compressive stress which lead to surface cracking and delamination between the Mo film and the substrate [18]. Bilayer structure is used as a solution to reduce excessive residual stress that exists on the Mo layer [24, 102]. The bottom Mo layer of tensile stress (formed under high pressure) and top Mo layer of compressive stress (formed under low

pressure) contribute an overall residual stress compensation to avoid delamination of the Mo film from the substrate. It is an undeniable fact that depositing two layers of Mo can resolve adhesion and resistivity problem until certain extent, but taking into consideration of other properties such as microstructure, grain size, crystal structure, and surface morphology will further improve the quality (electrical and optical) of the Mo films. Referring to Table 3, RF sputtered Mo films are more reflective and conductive while DC sputtered Mo films have larger grain size, better crystallization, and better surface morphology. A combination mode of both sputtering techniques to form multilayer molybdenum (DC/RF) can be realised to optimise the Mo back contact accordingly. The potential of simultaneous DC and RF sputtering on Mo deposition can also be examined as this technique has been used by employing an induction coil in the DC path (to avoid short circuit to the RF voltage) for depositing ZnO/ZnO:Al window and contact layer with an improvement in V_{oc} and I_{sc} [90] (Table 3).

5. Formation and Thickness Influence Factors of MoSe₂ Interface in CIGS Solar Cell

MoSe₂ is an interface formed between molybdenum back contact and CIGS absorber layer during the process of selenization at high temperature (above 723 K) [103, 104]. The selenium diffuses into the Mo back contact and reacts to form MoSe₂ along the process of selenization [105]. The MoSe₂ layers consist of polycrystalline grains with columnar structure and lattice spacing which improve the adhesion between Mo and CIGS [106–108]. Instead of that, MoSe₂ has a wider bandgap of 1.41 eV than the CIGS absorber that forms a back surface field which can hinder the recombination of electrons and holes [109–111]. The CIGS/Mo heterocontact including the MoSe₂ layer leads to a favourable ohmic-type contact by the evaluation of dark I-V measurement at lower temperature [25, 109]. Without the interface layer, a Schottky contact will be formed at the Mo/CIGS contact, causing significant problem in resistive losses [112]. However, excessive formation of MoSe₂ can lead to the delamination of the film and adverse effect on V_{oc} and FF of the completed CIGS solar cells due to high resistance of the MoSe₂ [113, 114]. Therefore, a range of specific MoSe₂ thickness between 100 nm and 200 nm is required to ensure good adhesion and electrical contact between Mo/CIGS [115–117].

The thickness of the MoSe₂ layer can be influenced by several factors such as sputtering conditions [117], residual stress in Mo layer [118], characterization of the barrier layer (TiN, MoN_x, and MoO_x) [115, 116, 119] and selenization

conditions [120–122]. The quality of the Mo crystals improves with sputtering power, thereby reducing the resistance of the Mo back contact. As the sputtering power increases, the thickness of MoSe₂ also increases [117]. This is due to the fact that increase in sputtering power enhances the diffraction intensity and crystallinity of Mo (110), (211) which facilitate the transformation of the cubic crystal structure Mo back contact into a hexagonal crystal structure MoSe₂ layer [123, 124]. However, varying the sputtering power to achieve the desired MoSe₂ thickness is not practical since high and low sputtering power are required to deposit a well-adhered and conductive Mo layer (bilayer structure) [99]. The MoSe₂ layer thickness is also dependent on the in-grain density of Mo which is interrelated to the residual stress. In a typical sputtered Mo layer, the tensile residual stress of Mo increases with the sputtering pressure up to 10 mTorr and decreases with further increase of pressure [118]. The increase in pressure reduces the mean free path and decreases the ion energy of the Mo particles, causing an increase in tensile stress (Region I). As the pressure further increases beyond 10 mTorr (Region II), the atomic attraction across grain boundary increases, creating porosity which eliminates the grain boundary attraction, thus reducing tensile stress. The reduction of the tensile stress in Region II will increase the in-grain density of the Mo back contact and decrease the MoSe₂ reactivity (thickness) during the selenization process.

In recent years, barrier layers are used to control the excessive formation of MoSe₂. A thin Titanium Nitride (TiN) barrier layer with a thickness of ~20 nm was grown on a Mo-coated soda-lime glass under vacuum condition prior to CZTSe deposition (CZTS has the same solar cell structure and works similarly to CIGS) [115]. Transition metal nitrides are used as a barrier layer because several researches showed that transition metal nitrides worked better in preventing diffusion at higher temperature than transition metals (Ta, Ti, W, and Mo) [125–127]. The TiN barrier layer has successfully suppressed the growth of MoSe₂ from ~1300 nm to ~200 nm significantly improving the V_{oc} , J_{sc} , and FF of the device. Other than TiN, the same concept has been applied by forming a molybdenum nitride (MoN_x) barrier layer to passivate the Mo back contact against selenization [116]. With MoN_x thickness of ~120 nm, the thickness of MoSe₂ formed was ~150 nm without consuming the entire upper thin Mo layer (20–30 nm left). One previous study has also demonstrated that the MoN_x barrier layer is able to control the formation of MoSe₂ and improve FF and current-voltage characteristic of the device, but thicker barrier layer will increase the series resistance of the device [128]. Other than the transition metal nitride barrier layer, molybdenum oxide (MoO_x) was proposed to control the growth of MoSe₂ [119]. It has been reported that a thin layer of MoO₂ as low as 10 nm is able to prevent the Mo back contact from overselenization and improve the V_{oc} , FF, and shunt resistance (R_{sh}) of the device [129]. In addition, oxygen allows diffusion of alkali metals during selenization which will enhance the performance of the device [130–134]. The behaviour of the MoO₂ barrier layer to control the formation of MoSe₂ can be explained with their corresponding Gibbs

free energy. At 900 K, the Gibbs free energy of the reaction between Mo and oxygen (−423 kJ/mol) is lower than the one of Mo with selenium (−129 kJ/mol) causing the reaction of MoO₂ with selenium (+294 kJ/mol) to be thermodynamically unfavoured, validating the passivation effect of MoO₂ on Mo back contact against selenization [119].

A well-crystallised CIGS absorber layer requires a process temperature of at least 500°C and above for the selenization process [118]. Nonetheless, at high process temperature, the thickness of the MoSe₂ layer will increase significantly [117]. If the process temperature is lowered to reduce the thickness of MoSe₂, deterioration of the absorber layer (electrical properties) will occur [135]. Besides the mentioned thickness-influencing factors, sodium (Na) might be the dominant factor to control formation of MoSe₂ layer [25, 123]. An experiment has been conducted by varying the amount of Na (using SiO_x barrier layer) diffuses from the substrate across the Mo back contact to the CIGS absorber layer [123]. The experiment concluded that Na aids in the formation of MoSe₂, but if the Na content is too high, it passivates the grain boundaries at CIGS layer to form Na₂Se_x; thus, lesser Se atom is available to react with Mo, and this will retard the formation of MoSe₂. Comparing these thickness-influencing factors, barrier layer appeared to be the most effective method to control the thickness of MoSe₂, allowing sodium (Na) diffusion while minimizing the adverse effect on cell performance.

6. Scale-up Challenges/Issues of CIGS Thin Film Modules

Since 2006, thin film solar cell production in the U.S. has outperformed the production of c-Si solar cell, becoming the least expensive technology to be manufactured [35, 48]. The existing thin film PV technologies especially CIGS have reached over 1.6 GW of cumulative module production in 2015 and is reported to have a high cost reduction potential at high production volumes [136, 137]. For a production capacity of 1000 MW/yr with 15% module efficiency, the CIGS module production cost as low as \$0.34/Wp can be achieved [10]. Due to photovoltaics cost declining with maturity of the conventional PV technologies and new entry of China into the market in 2010, the future challenges for CIGS production will be combining high production volumes with high throughput, sufficient yield, and superior quality. Meaning that the production cost has to be brought down to make it competitive with conventional sources.

Currently, the leading CIGS module manufacturer Solar Frontier claims that their CIS module is able to output more electricity than the conventional crystalline silicon (c-Si) in real-world conditions such as better performance at high temperatures, low-light condition, light-soaking effect, and also shadow tolerance [138]. With mass production of modules having efficiencies ranging from 11.8% to 13.8%, Solar Frontier was able to achieve an annual production of ~1 GW and over 3 GW of shipments worldwide in 2015 [139, 140]. The statement made on surpassing the performance of c-Si solar cells is according to the cadmium- and lead-free CIS module (SF 150-170S Series)

which provides an efficiency of 13.8% for a total area of 12,280 cm² [140, 141]. The current technology offered by Solar Frontier with high energy yield leads to a shorter energy payback time which fulfil the requirement to be competitive towards c-Si solar cells by producing high-quality module at lower cost. On the other hand, Solar Frontier has also attained a higher efficiency of 19.2% on cadmium-free CIGS minimodule [36, 142]. This research direction affirms the potential of cadmium-free CIGS module with consideration on environmental issues (reduce usage of cadmium) as the energy production increases.

The production of CdTe PV modules is ahead of CIGS PV modules at present stage due to simplicity of the process. However, different approaches have been used to boost up the CIGS cell efficiency such as applying Post Deposition Treatment (PDT) on the CIGS surface with alkali elements, incorporating more gallium into the absorber layer (CIGS) and also combining with other materials such as perovskites for multijunctions [1]. PDT is the process developed by ZSW to reach 22.6% efficiency in CIGS solar cells, but the compatibility and practicality between new technologies and scaling it up into mass production should be taken into account earnestly. Therefore, the challenges/issues in CIGS solar cell production are discussed as below:

- (i) Uniformity of CIGS absorber film over large areas: Uniformity is essential for electrical and optical properties of high-efficiency solar cells, and it directly influences the yield in production. The yield determines actual production volume in MW/yr and production cost in ¢/Wp [35]. Coevaporation can be an appropriate technique for large area substrate because the coevaporated CuGaIn precursor has a higher selenization rate than the cosputtered one, and it has been actively developed by many manufacturing companies [122]
- (ii) Standardizing the cell fabrication process: In the industry, Mo and ZnO films were sputtered (vacuum system); buffer layer CdS was deposited using Chemical Bath Deposition (nonvacuum system), and absorber layer CIGS was deposited with the coevaporation method (vacuum system) [143]. Fabricating the cell in and out from vacuum and nonvacuum process will cause difficulty in troubleshooting once a problem occurs. Substrate handling in open air environment and between different tools can contaminate the substrate. This can be avoided by using a vacuum process along the fabrication of the whole cell
- (iii) Presence of moisture in CIGS modules: Water vapor will oxidise the back contact molybdenum (Mo) causing Mo to degrade [8]. To solve this issue, a robust encapsulation technology with the properties of durability, adhesion, thermal stability, etc. is required
- (iv) Long processing time for CIGS and TCO layers: The processing time for CIGS (including deposition and selenization) and Transparent Conducting Oxide (TCO) layers should be reduced to meet the required industrial production time (10 min [1]) when it involves large volume production. The processing time can be improved by using a thinner CIGS absorber layer and high-speed deposition technique for TCO
- (v) Cadmium used in the CIGS buffer layer (CdS) is toxic, and disposal of the cadmium-containing product causes detrimental effect to human health [144]. Moreover, the use of CdS buffer layer leads to optical absorption loss [145]. Hence, the CdS buffer layer can be replaced by other appropriate wider bandgap buffer materials to improve the short-circuit current (J_{sc}) of the device
- (vi) Indium scarcity on CIGS module: Production of indium currently relies in by-products of mining and refining of other material (in particular, zinc) [146]. There is a concern raised upon scarcity of indium might escalate the price and can be a threat to CIGS ambitions for production cost reduction and cost competitiveness in the wider PV market. Addressing this issue, CIGS layer of $\leq 1 \mu\text{m}$ should be used without compromising J_{sc} of the device [1].

To sustain a profitable business in the PV market, PV module manufacturers will have to adopt or innovate technologies that offer a production cost $< \$0.5/\text{watt}$ with the capability to match c-Si module performance. The manufacturing cost of CIGS solar cell can be further reduced by increasing process yield, production capacity, and efficiency while the raw material scarcity can be eased by improving material utilisation (thinner absorber layer) during the deposition process. Nonetheless, due to the maturation of solar PV sector, the cost effectiveness of solar cell can no longer be determined solely on manufacturing cost ($\text{\$/Wp}$) and efficiency. Aspects such as annual electricity yield, cost of PV modules, Balance of System (BOS) ($\text{\$/W}$), and cost incurred for the PV system are critical to compute the minimum price at which energy must be sold to break-even the solar PV project [147–149]. This concept is also known as Levelized Cost of Energy (LCOE). Between 2010 and 2017, the LCOE for utility scale crystalline silicon (c-Si) PV plants have reduced from 0.36 $\text{\$/kWh}$ to 0.10 $\text{\$/kWh}$, mainly driven by the reduction of module prices [150]. From the year 2010 to 2012, LCOE at the price of 0.12–0.20 $\text{\$/kWh}$ to 0.11 $\text{\$/kWh}$ was achieved by the thin film PV plants, and the LCOE price is estimated to be decreased to about 0.06–0.10 $\text{\$/kWh}$ by 2020 [151–153].

The major manufacturer of CdTe thin films, First Solar, was able to obtain a LCOE at 0.15 $\text{\$/kWh}$, and the LCOE was targeted to be brought down to 0.08 $\text{\$/kWh}$ with the system cost of 2 $\text{\$/W}$ [154]. Whereas CIGS (14% efficiency) has attained a decent LCOE at 0.084 $\text{\$/kWh}$ for a 100 MW scale PV system [155]. Besides that, the CIGS LCOE reported at 0.084 $\text{\$/kWh}$ was being compared with c-Si solar cell (16% efficiency) in an identical location and system, and the LCOE

of the c-Si solar cell was at 0.80 \$/kWh, lower than CIGS but comparable to each other [155]. This can be explained by the fact that c-Si solar cell has better module efficiency causing the Balance of System (BOS) to be reduced meanwhile CIGS offers better performance under high temperature and low-light condition thus diminishing the LCOE differences between c-Si and CIGS solar cell. The LCOE of CIGS is also believed to be higher than the CdTe thin films at this moment; it is because CIGS thin film possesses lower module efficiency and requires more complex system that tends to increase both cost on BOS and PV system. Nevertheless, CIGS technology is still feasible and promising as its record efficiency (22.6%) has surpassed CdTe (21.6%), and the theoretical efficiency limit of CIGS (33.5%) is higher than the c-Si (29.43%) solar cells [37, 51, 52]. This implies that CIGS solar cell with greater efficiency potential will result in lower material usage, lower system cost, and higher energy yield which eventually contribute to an exceptional Levelized Cost of Energy (LCOE).

7. Conclusion

CIGS solar cells are believed to have a very high potential against c-Si and CdTe solar cells in achieving low production cost with high module efficiency as the CIGS possesses better absorption coefficient (lower material usage), requires lower thermal budget than c-Si solar cells, and its record efficiency (22.6%) has just surpassed CdTe (22.1%). The rigid substrates such as soda-lime glass and alkali-aluminosilicate glass give rise to higher efficiency with direct Na incorporation. However, flexible substrates have proven its capability to be an alternative to rigid substrates by achieving a decent efficiency comparable to the rigid substrate solar cells by applying Post Deposition Treatment. DC sputtered Mo films are favouring physical properties while RF sputtered Mo films are favouring electrical properties. Thus, multilayer molybdenum (DC/RF) or simultaneous DC and RF sputtering mode can be explored to optimise the molybdenum back contact. On the other hand, the MoSe₂ layer is important in forming ohmic-type contact and improving the adhesion between CIGS and Mo layers. Excessive formation of MoSe₂ can cause delamination problem and increase in resistivity of the solar cell. Addressing this issue, the most effective method to control the thickness of MoSe₂ is forming a barrier layer in between the bilayer structure Mo back contact. The scale-up challenges/issues of CIGS thin film modules discussed are possible to overcome as current CIGS technology has started to offer better performance at high temperature and low-light condition than the c-Si solar cells in real-world conditions. Lastly, with greater efficiency potential and higher total lifetime power produced, CIGS technology will eventually attain an outstanding Levelized Cost of Energy (LCOE) against c-Si and CdTe solar cells.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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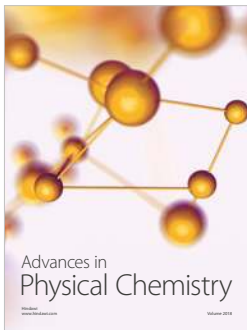
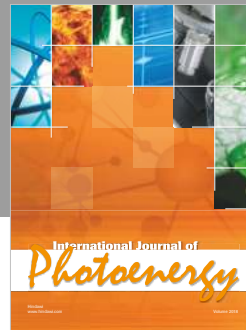
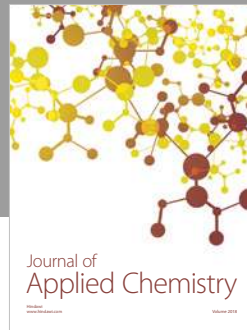
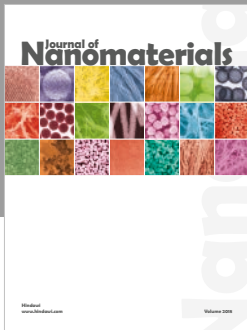
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