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Thermal Energy Storage

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Madrid Institute of Advanced Studies, Madrid, Spain

CSP, Central Receiver, Heat Transfer Fluids, High-temperature Solar Reactor



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Sandia National Laboratories, Albuquerque, New Mexico, United States

CSP, Receivers



Wojciech Lipiński

Australian National University, Canberra, Australia

Solar Chemistry



Peter G. Loutzenhiser, PhD

Georgia Institute of Technology, Atlanta, Georgia, United States



Jayanta Nayak, Ph D

Indian Institute of Technology Bombay, Mumbai, India

CSP, Parabolic Trough



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Photovoltaics, PV and Cell Physics, Renewable/Solar



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Photovoltaics, Photocatalysis, Perovskite Solar Cells, Organic Solar Cells, DSSC



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Dye Sensitized, Organic Solar Cells, Hybrid Solar Cells



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Green energy, Solar cells, Electronic materials, Characterization, Materials Science



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solar radiation, solar energy, numerical weather prediction, satellite observation, clouds, aerosol



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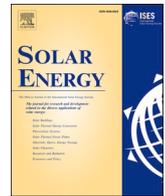
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## Review of CIGS-based solar cells manufacturing by structural engineering

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### ABSTRACT

The design and development of the next-generation power-efficient CIGS solar cells are at the research forefront due to their potential applications in renewable energy. Due to rich fundamental properties such as chemical and physical structures of the CIGS layer, cell scaffolding, and its promising applications like low cost, easy integration, and high efficiency, the CIGS-based solar cell systems are of considerable interest and received tremendous attention. In this article, we review the CIGS solar cells from the point of view of structural engineering. We explain the intrinsic parts of crystalline, optical, and electronic structures of the CIGS absorber layer up to the extrinsic part of the cell multilayer structure. For intrinsic structure, we primarily review the modification of the crystallinity or chemical composition of the CIGS and the effects that these modifications have on the physical properties such as the adjustment of the bandgap grading, effect of impurity or doping, selenization, oxidation processes, and the surface morphology and structure orientation. For extrinsic structure, the effect of substrates, electrical back contact, windows, *n*-buffer, grid, and antireflection layers will be discussed further, as well as the possibility of their tandem use with other solar cell thin films.

### 1. Introductions

Chalcopyrite Cu(In, Ga)Se<sub>2</sub> (CIGS)-based solar cells are promising and widely used solar cells because of their remarkable efficiency, low cost, and easy integration (Noufi and Zweibel, 2006; Ramanujam and Singh, 2017). This is related to their tunable bandgap of approximately 1.0–1.12 eV and high absorption coefficient up to 10<sup>5</sup> cm<sup>-1</sup> (Guillemoles, 2002; Noufi and Zweibel, 2006; Ramanujam and Singh, 2017). Solar cells based on CIGS have high efficiency that is similar to that of crystalline silicon (c-Si) solar cells but are less expensive because CIGS can absorb light using only ~2.0–2.5 mm layer thickness, which decreases the use of raw materials (Guillemoles, 2002; Noufi and Zweibel, 2006; Ramanujam and Singh, 2017). The CIGS-based solar cells are easy to fabricate compared to c-Si based solar cells by growing it on various rigid and flexible substrates by vacuum and non-vacuum techniques; thus, CIGS-based solar cells are promising candidates for the next-generation power-efficient solar cells (Adel et al., 2016; Badgular et al., 2015; Chen et al., 2017a, 2014; Choi and Lee, 2007; Delahoy et al.,

2004; He et al., 2019; Kaelin et al., 2004; Kuo et al., 2016; Kushiya et al., 2001; Lee et al., 2011; Liu et al., 2012; Nakada et al., 1999; Park et al., 2003; Repins et al., 2008; Tsai et al., 2013; Venkatachalam et al., 2008). In addition to CdTe thin films, CIGS is included in the second-generation thin-film solar cells but CIGS is non-toxic compared to CdTe (Noufi and Zweibel, 2006; Ramanujam and Singh, 2017). Besides its tunable bandgap, CIGS is an excellent semiconductor material for creating tandem solar cells. Some theoretical and experimental studies have been conducted to obtain high efficient CIGS-based solar cells. Among them, structural modifications of crystalline and electronic structures to tune its functionalities are considered to be a general route to enhance the CIGS-based solar cell efficiency (Asaduzzaman et al., 2016; Chen et al., 2017b; Chirilă et al., 2013; Han et al., 2012; Ishizuka et al., 2011; Liao et al., 2013; Liu et al., 2011; Malitckaya et al., 2017; Puyvelde et al., 2014; Salomé et al., 2013; Su et al., 2011; Sun et al., 2017).

Nevertheless, compared to c-Si, CIGS-based solar cells are not as widely commercialized due to the difficulty of finding high quality and large-scale of CIGS-based solar cell. It was found that the efficiency of

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the commercial CIGS module much lower than the obtained lab-scale CIGS (Ramanujam and Singh, 2017). Electronic inhomogeneity is considered to be an important key for efficiency limitation (Ramanujam and Singh, 2017; Werner et al., 2005). A correct Cu-In-Ga and Se composition are required to obtain appropriate band-gap, while those of Cu-In-Ga-Se composition is very sensitive to the growth condition during the fabrication process. Thus, considerable research and development efforts have been conducted to overcome the limitations of CIGS, particularly on their chemical and electronic structures. In this review article, the exploitation of CIGS solar cells is described with an emphasis on the chemical and physical issues of the CIGS layer and other components of the cell. We review recent fabrication approaches and provide suggestions to improve the efficiency and stability of CIGS solar cells from the point of view of structural engineering to develop the functionalities of CIGS-based solar cells for renewable energy applications. We begin by reviewing the intrinsic structure such as grading composition to obtain a bandgap grading. The effects of impurity or doping are also briefly discussed including direct doping by the addition of impurities to the CIGS layer directly during or after the coating process and indirect doping, which is facilitated by the diffusion of related materials from the substrates and/or buffer layers (Asaduzzaman et al., 2016; Chirilă et al., 2013; Malitckaya et al., 2017; Puyvelde et al., 2014; Salomé et al., 2013; Shirakata, 2017; Sun et al., 2017). Moreover, selenization and annealing processes, which are considered to be important for tuning the electronic structure, are explained (Chen et al., 2017b; Ramanujam and Singh, 2017). Thus, the design of the CIGS layer morphology, which is used to control the reflection behavior, is discussed (Cai and Qi, 2015; Han et al., 2012; Ishizuka et al., 2011; Liao et al., 2013; Liu et al., 2011; Shirakata, 2017; Su et al., 2011). Furthermore, the extrinsic or multilayer structure of CIGS-based solar cells is also reviewed including the effect of substrates and challenges of developing flexible solar cells, the influence and necessity of electrical back contact, the search for a proper *n*-type buffer layer, and the effects of an additional layer (e.g., grid, window layers and antireflection) on the performance of solar cells (Bhattacharya and Ramanathan, 2004; Dhere et al., 2004; Heriche et al., 2016; Kessler and Rudmann, 2004; Kushiya, 2004; Nakada et al., 2004; Qiao et al., 2018; Reinhard et al., 2013). Finally, further development and challenges are briefly reviewed such as the potential combination of the CIGS layer to form tandem solar cells (Blanker et al., 2016; Chae et al., 2016; Elbar et al., 2015; Elbar and Tobbeche, 2015; Guchhait et al., 2017; Lee et al., 2018; Moon et al., 2015; Shen et al., 2018; Todorov et al., 2016, 2015; Werner et al., 2018; Yamaguchi et al., 2018).

## 2. Fabrication of CIGS-based solar cells

The CIGS-based solar cells can be fabricated on both rigid and flexible substrates by various vacuum and non-vacuum techniques. For example, co-evaporation (Repins et al., 2008), physical vapor deposition (PVD) (He et al., 2019), pulsed laser deposition (PLD) (Tsai et al., 2013), chemical vapor deposition (CVD) (Park et al., 2003), metalorganic chemical vapor deposition (MOCVD) (Choi and Lee, 2007), electron beam deposition (EBD) (Venkatachalam et al., 2008), molecular beam epitaxy (MBE) (Nakada et al., 1999), and sputtering (Delahoy et al., 2004; Kushiya et al., 2001) are vacuum techniques that can be utilized to fabricate CIGS-based solar cells. CIGS solar cell fabrication starts with the deposition of the electrical back contact layer on the substrates and later finished by the coating of the window layer which is primarily deposited using vacuum deposition methods, while, the *n*-buffer layer can be coated using both vacuum and non-vacuum techniques (Choi and Lee, 2007; Delahoy et al., 2004; He et al., 2019; Kushiya et al., 2001; Nakada et al., 1999). The vacuum techniques can be distinguished by how the material is deposited. Specifically, the material can be deposited element by element with several stages of deposition (Choi and Lee, 2007; Nakada et al., 1999; Repins et al., 2008) or by direct deposition, where, we can deposit the CIGS and other component layers in a single

step (Chen et al., 2014; Delahoy et al., 2004; Kushiya et al., 2001; Tsai et al., 2013). Using co-evaporation (e.g., PVD, MOCVD, and MBE techniques), the CIGS layer growth can be achieved in several steps of depositions at various rates with a constant Se flux. The growth of CIGS in several steps is essential to achieve a grading composition and a Cu-poor layer on the CIGS surface. This can be achieved by the separated deposition of In and Ga with Cu and also by controlling the amount of Cu in the first and last stages of deposition (Ramanujam and Singh, 2017). Of note, such graded composition to obtain a graded bandgap and Cu-poor interface to reduce recombination are important to enhance cell efficiency. This aspect will be explained in the next section. Nevertheless, a single steps process is essential to time and cost-efficiency. Specifically, PLD and sputtering can be completed by a single process due to the unavailability of multiple element sources (Chen et al., 2014; Delahoy et al., 2004; Kushiya et al., 2001; Tsai et al., 2013). The CIGS can be directly grown by evaporating a quaternary CIGS target that is made from the fine composition of a Cu-In-Ga powder in the constant Se environment. Using PLD, CIGS can be produced only in a small area; thus, this method is only suitable for lab-scale (Chen et al., 2014; Tsai et al., 2013). The sputtering method can produce large CIGS areas, which are suitable for industrial production (Delahoy et al., 2004; Kushiya et al., 2001). Though high-quality samples can be obtained by vacuum deposition, these approaches have several disadvantages such as high cost and time consumption and poor uniformity over a large area owing to a cosine flux distribution, which results in a sharp change in film composition and lower Se incorporation (Kaelin et al., 2004; Ramanujam and Singh, 2017). Vacuum methods are more efficient if the cells are fabricated without a vacuum break in a full-stack deposition tool (He et al., 2019). However, almost all deposition methods require a post-selenization process (Chen et al., 2017b). Thus, the vacuum has to be broken to perform additional selenization in rapid thermal processing or reactive annealing to achieve the optimal formation of CIGS composition.

The cost of production and complex vacuum processes compels researchers to develop low-cost and simple non-vacuum methods (Adel et al., 2016; Badgujar et al., 2015; Chen et al., 2017a, 2017b; Kaelin et al., 2004; Kuo et al., 2016; Lee et al., 2011; Liu et al., 2012). The non-vacuum techniques are better in and stoichiometric control and material utilization, require low energy input and exhibit high compatible processing compared to the vacuum techniques (Badgujar et al., 2015; Chen et al., 2017a; Kuo et al., 2016; Lee et al., 2011; Liu et al., 2012). Non-vacuum techniques can be used in a two-step process, for example, deposition or printing of the CIGS precursor layer at low temperature and then followed by the selenization process at high temperature (Adel et al., 2016; Badgujar et al., 2015; Chen et al., 2017a; Kaelin et al., 2004; Kuo et al., 2016; Lee et al., 2011; Liu et al., 2012). Moreover, according to the deposition process, the first step to obtaining the precursor layer can be classified into electroless deposition (chemical bath deposition and electrodeposition) (Adel et al., 2016; Kaelin et al., 2004) and particulate/solution deposition (spin/spray coating and paste coating; screen/inkjet printing, doctor-blade coating, and curtain coating) (Badgujar et al., 2015; Chen et al., 2017a; Kuo et al., 2016; Lee et al., 2011; Liu et al., 2012). In the electroless deposition, the precursor layer is deposited using complexing agents to decrease all individual precursor element potentials or make them similar to each other (Adel et al., 2016; Kaelin et al., 2004; Ramanujam and Singh, 2017). Whereas, in the particulate/solution deposition, the precursor material is initially reduced to the particulate form with the desired stoichiometry. Thus, the material can be deposited directly by a liquid binder as a transfer media onto a substrate at specific substrate temperatures (Badgujar et al., 2015; Chen et al., 2017a; Kaelin et al., 2004; Kuo et al., 2016; Lee et al., 2011; Liu et al., 2012). Particulate materials can be prepared by several methods such as solution precipitation, laser pyrolysis, and laser ablation (Adel et al., 2016; Kaelin et al., 2004). After the deposition using both electrodes and particulate/solution depositions, the CIGS layer is performed by sintering the precursor layer in a controlled Se atmosphere

(Kaelin et al., 2004). Recent work using a non-vacuum-based route has been developed in our laboratory, namely the hot injection method. We found that by relatively simple preparation and low-cost, we have successfully grown a high-quality CIGS layer which comparable with previous work of CIGS fabricated by other techniques (Dewi et al., 2020).

The particulate/solution techniques are commonly used deposition of non-vacuum techniques. Wherein, large-scale production with high speed, less material waste, and low cost can be realized. However, the efficiency is not sufficiently high because of high series resistance at the electrical back contact and CIGS interface (Kaelin et al., 2004; Ramanujam and Singh, 2017). In general, the quality of the samples synthesized by the non-vacuum techniques is not as good as the vacuum techniques which reflect the low cell efficiency. Nevertheless, a non-vacuum technique offers wider avenue and relatively easy to develop the physics and chemical structures of the CIGS based-solar cell to enhance efficiency. For instance, simply tuning compositions of the Cu, In, Ga and the amount of doping during synthesizing of CIGS precursor, controlling the Se composition during the selenization process, as well as modifying the surface structure of the CIGS layer by heat treatments (Han et al., 2012; Kuo et al., 2016; Ramanujam and Singh, 2017; Sun et al., 2017).

We have compiled the efficiency of CIGS based solar cells fabricated using various techniques as shown in Fig. 1. The fabrication techniques used to grow the CIGS layer have enormous effects on cell efficiency. Despite the complicated process of the vacuum techniques, the CIGS based solar cell fabricated by using this method possesses cell efficiency up to 20% (Contreras et al., 1999; Reinhard et al., 2013; Repins et al., 2008). In general, the cell efficiency of CIGS based solar fabricated by vacuum techniques has higher cell efficiency compare to non-vacuum techniques due to better crystallinity of the CIGS layer. Nevertheless, the efficiency of the cell fabricated by non-vacuum techniques can be increased in numerous ways. For instance, the efficiency of the cell fabricated by electrodeposition can be raised to 15.4% by adding the In and Ga compositions thus annealed at high temperature (Bhattacharya et al., 2000). The efficiency of the cell fabricated by ink printing also can be optimized by femtosecond-laser annealing treatment which can be up to 11.05% cell efficiency (Chen et al., 2017a). Moreover, the efficiency of the cell fabricated by the doctor's blade coating also can be increased by up to 13.6% by controlling the Ga composition (Kapur et al., 2003). We will discuss the physics and chemical structures modification related to composition grading, doping, selenization, and surface morphology of the CIGS based solar cell in more detail in the next sections.

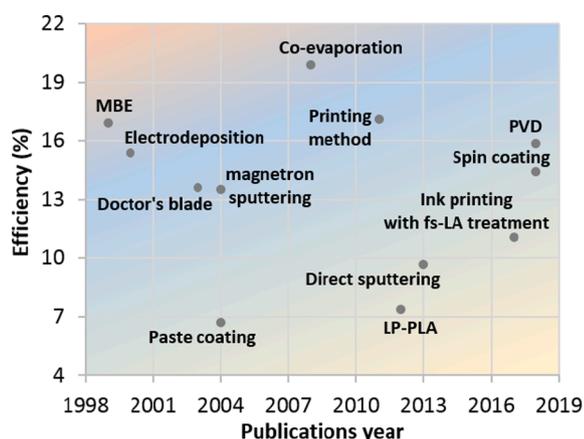


Fig. 1. The efficiency of CIGS based solar cell fabricated using various techniques. The fs-LA stands for femtosecond annealing process and LP-PLA is liquid-phase pulsed laser ablation. (Bhattacharya et al., 2000; Brown et al., 2012; Chen et al., 2017a; Delahoy et al., 2004; Guo and Liu, 2012; He et al., 2018; Kaelin et al., 2004; Kapur et al., 2003; Liu et al., 2013; Nakada et al., 1999; Park et al., 2018; Repins et al., 2008).

### 3. Tuning optical and electronic properties by structural engineering

The optical and electrical bandgaps of the CIGS absorber and multilayer components are essential to obtain desirable efficient solar cells. This relates to optimum bandgap for a CIGS is approximate  $\sim 1.14$  eV as the activation energy for an electron to jump from the valence band to the conduction band. Absorber bandgap of 1.4 eV is required to absorb the solar spectrum and the CIGS efficiency decreases when the bandgap is higher (Ramanujam and Singh, 2017). The CIGS bandgap can be tuned by changing the intrinsic structure and of the multilayer structure of the solar cells (Asaduzzaman et al., 2016; Bhattacharya and Ramanathan, 2004; Cai and Qi, 2015; Chirilă et al., 2013; Han et al., 2012; Heriche et al., 2016; Ishizuka et al., 2011; Kushiya, 2004; Liao et al., 2013; Liu et al., 2011; Malitckaya et al., 2017; Nakada et al., 2004; Puyvelde et al., 2014; Salomé et al., 2013; Shirakata, 2017; Sun et al., 2011; Sun et al., 2017). As shown in Fig. 2, we separated the structural manipulation into two parts, intrinsic (orange) and extrinsic (purple) regions. The substrate and electrical back contact (green) are essential for the indirect tuning of the intrinsic structure of the cell by the incorporation of Na into the CIGS layer from the soda-lime glass (SLG) substrate, which can improve the optical and electric properties of CIGS (Asaduzzaman et al., 2016; Ramanujam and Singh, 2017; Repins et al., 2008; Salomé et al., 2013; Sun et al., 2017).

In the intrinsic region, manipulation of the band gap can deal with tuning of the Cu/Ga/In composition to obtain a graded bandgap to improve the optical and electronic properties (Asaduzzaman et al., 2016; Ramanujam and Singh, 2017; Repins et al., 2008). Specifically, at the CIGS/buffer interface, band bending causes the conduction band grading offset. Thus, if the Ga concentration is high, minor carrier collection from the CIGS absorber reduces. The grading conduction band profile can be tuned by decreasing the Ga atoms diffusion on the surface. Therefore, it is important to vary Ga concentration carefully to achieve the right bandgap with high efficiency. Moreover, doping by an alkali metal such as Li, Na, K, Rb, and Cs or doping by Zn, and Sn can be used to manipulate the bandgap of the CIGS absorber layer (Asaduzzaman et al., 2016; Chirilă et al., 2013; Malitckaya et al., 2017; Puyvelde et al., 2014; Salomé et al., 2013; Shirakata, 2017; Sun et al., 2017). In addition to the incorporation from the soda-lime glass (SLG) substrate, Na doping can be achieved using an additional buffer layer before the CIGS layer deposition (Salomé et al., 2013; Sun et al., 2017). It is well known that doping Na and Zn can increase the free carrier density of the CIGS absorber layer (Asaduzzaman et al., 2016; Chirilă et al., 2013; Malitckaya et al., 2017; Salomé et al., 2013; Shirakata, 2017; Sun et al.,

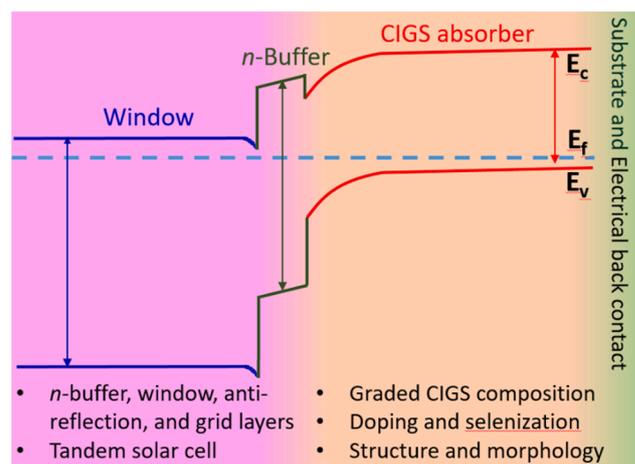


Fig. 2. Bandgap diagram of CIGS-based solar cells. The optimization of solar cell efficiency can be obtained by manipulating the bandgap structure using structural engineering.

2017). Besides, doping Na improves the texturing of the CIGS morphology, which increases cell efficiency (Malitckaya et al., 2017; Salomé et al., 2013; Sun et al., 2017). Similarly, K doping modifies the CIGS surface morphology and facilitates diffusion of Cd in the Cu-depleted surface, which results in the improved quality of the CIGS/CdS interface (Chirilă et al., 2013; Malitckaya et al., 2017). Modification of the CIGS surface morphology is essential for enhancing cell efficiency (Han et al., 2012; Ishizuka et al., 2011; Liao et al., 2013; Liu et al., 2011; Su et al., 2011). These goals require research, which includes the modification of the morphology into nanotip arrays to tune the optical properties of CIGS (Liao et al., 2013; Liu et al., 2011).

Furthermore, the bandgap can be manipulated by modifying the structure of the multilayer components of the cells. For example, the influence of Cd in CdS, which is known as a donor that induces *n*-type doping of the absorber surface region, modifies the bandgap at the surface of CIGS. The choice of a proper *n*-buffer layer is interesting for research, wherein, CdS is the best choice for the *n*-type buffer layer, although Cd is toxic (Ramanujam and Singh, 2017; Repins et al., 2008). Nevertheless, Cd-free layers, such as Zn(O, S, OH)<sub>x</sub> and Zn(O, S), have been used as an *n*-buffer layer, wherein, its high bandgap can improve the blue response of the solar cells (Bhattacharya and Ramanathan, 2004; Kushiya, 2004; Kushiya et al., 2001; Ramanujam and Singh, 2017). The decrease in the CdS layer thickness and the combination with window layers, such as ZnO, can be used to minimize CdS usage (Bhattacharya and Ramanathan, 2004; Kushiya, 2004; Kushiya et al., 2001; Ramanujam and Singh, 2017). CIGS is a promising material for tandem solar cells because of its tunable bandgap. The combination of CIGS with organic and inorganic solar cells yields different bandgap energies induce wider energy regions of the solar spectrum that can be converted into electricity, therefore the cell efficiency increases (Bailie et al., 2015; Blanker et al., 2016; Chae et al., 2016; Elbar et al., 2015; Elbar and Tobbeche, 2015; Guchhait et al., 2017; Kaigawa et al., 2010; Lee et al., 2018; Moon et al., 2015; Nakada et al., 2006; Nanayakkara et al., 2017; Schmid et al., 2010; Shen et al., 2018; Todorov et al., 2016, 2015; Werner et al., 2018; Xiao et al., 2010; Yamaguchi et al., 2018).

### 3.1. Intrinsic structure

Many attempts to enhance cell efficiency of CIGS-based solar cells have been made such as double bandgap grading engineering (Asaduzzaman et al., 2016; Ramanujam and Singh, 2017; Repins et al., 2008), doping (Asaduzzaman et al., 2016; Chirilă et al., 2013; Malitckaya et al., 2017; Puyvelde et al., 2014; Salomé et al., 2013; Shirakata, 2017; Sun et al., 2017), and control of the surface structure/morphology (Han et al., 2012; Ishizuka et al., 2011; Liao et al., 2013; Liu

et al., 2011; Su et al., 2011). The CIGS absorber layer development is important due to its main contribution to light absorption. CIGS is a ternary compound *p*-type semiconductor that belongs to the I–III–VI<sub>2</sub> family and crystallizes with the structure of tetragonal chalcopyrite CuXY<sub>2</sub> (X = In, Ga, Al, and Y = Se, S), as shown in Fig. 3a. The lattice parameter is related to the In/Ga composition of  $c = 56\text{--}58 \text{ \AA}$  ( $x = 0\text{--}1$ ) and  $a = 1.1\text{--}1.15 \text{ \AA}$  ( $x = 0\text{--}1$ ), which is known as tetragonal distortion that originates from Cu–Se, Ga–Se or In–Se bonds. The variation of In/Ga composition induces an alteration in the bandgap (Ramanujam and Singh, 2017). Thus, the optical bandgap can be tuned to match the solar spectrum to improve open-circuit voltage (Voc). Tuning the bandgap to change the conduction band offset is referred to as bandgap grading, wherein, the bandgap increases from 1.04 eV to 1.7 eV when Ga replaces all In. this graded bandgap composition can reduce recombination losses and improve the electronic properties. In the CIGS layer, the high-efficiency solar cells can be achieved with the (Cu)/(In + Ga) ratio in the range of 0.88–0.92 and low Ga atomic ratio [Ga/(Ga + In)] around 0.26. The number of defects in the CIGS layer increases with Ga content. In order to obtain high Voc and efficiency, Ga concentration has to be carefully varied to obtain a wider bandgap > 1.14 eV (Ramanujam and Singh, 2017). The backside grading can increase carrier collection and short-circuit current density (Jsc) and decreases the recombination of bulk/surface at the back contact interface created by Ga grading. A graded bandgap of CIGS films can be fabricated by the deposition of CIS/CIGS bilayer or CGS/CIS/CGS trilayer systems (Noikaew et al., 2018).

Bandgap grading can reduce recombination losses and ensure a highly efficient collection of charge carriers. Besides, doping is another way to enhance cell efficiency by manipulating the chemical composition of CIGS (Asaduzzaman et al., 2016; Chirilă et al., 2013; Malitckaya et al., 2017; Puyvelde et al., 2014; Salomé et al., 2013; Shirakata, 2017; Sun et al., 2017). In this article, doping is categorized into direct and indirect doping. Indirect doping includes external structural manipulation, which will be discussed in the next section. Direct doping by adding an impurity to CIGS helps develop the CIGS performance; however, this method is less popular than indirect doping facilitated by substrates (i.e., Na doping) (Asaduzzaman et al., 2016; Salomé et al., 2013; Sun et al., 2017), *n*-buffer (i.e., Cd doping) (Nanayakkara et al., 2017), and additional layers such as a window layer (i.e., Zn doping) (He et al., 2019). Direct doping can be realized by adding an impurity during or after the CIGS growth. The addition of impurity after the CIGS layer process is known as post-deposition treatment (PDT), can be executed by evaporating the impurities under a selenium atmosphere on top of the CIGS layer, as depicted in Fig. 3b (Chirilă et al., 2013; Malitckaya et al., 2017).

Alkali metals such as Li, Na, K, Rb, and Cs can tune the physical

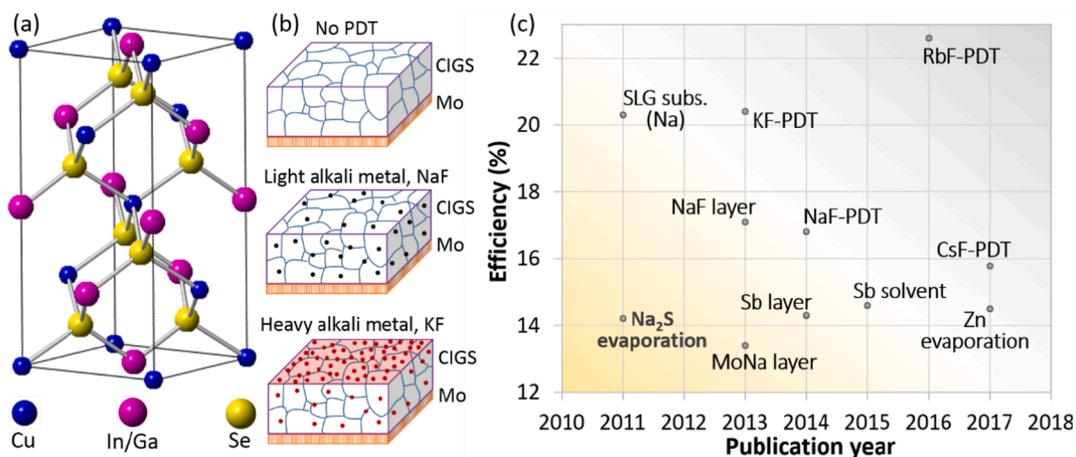


Fig. 3. (a) Crystal structure of CIGS. (b) Different roles of light and heavy alkali metal doping on the CIGS layer. (c) Compilation of CIGS solar cells with doping and its sources (Asaduzzaman et al., 2016; Chirilă et al., 2013; Jackson et al., 2016, 2011; Malitckaya et al., 2017; Mansfield et al., 2015; Pianezzi et al., 2014; Puyvelde et al., 2014; Salomé et al., 2013; Shin et al., 2011; Shirakata, 2017; Sun et al., 2017).

properties of CIGS, which enhance cell efficiency; this has been confirmed theoretically and experimentally (Chirilă et al., 2013; Malitckaya et al., 2017). Na element is a common impurity that enhances the performance of CIGS (Asaduzzaman et al., 2016; Repins et al., 2008; Salomé et al., 2013; Sun et al., 2017). The doping with the light element (Li, and Na) and heavy elements (Rb and Cs) alkali metals differently impact the structure of CIGS, while, K has effects that are similar to both alkali metal types (Malitckaya et al., 2017). Li and Na prevail as impurities in the grain interior and prefer to occupy Cu sites as substitutional neutral impurities or as positively charged impurity pairs via the interstitial migration mechanism that increases Cu depletion due to the out-diffusion of Cu and the in-diffusion of the dopant (Malitckaya et al., 2017). Whereas, the heavy metal like Rb and Cs can make secondary phases formation with Se and In close to the surface. The morphology of the buffer surface can be improved by these secondary phases can improve by enabling alignment of the band. Moreover, they have large energy band gaps which can improve the electrical properties of the device. Besides, it is reported that doping of K relieves Cd diffusion in the Cu-depleted of the CIGS layer, which improves the CIGS/CdS interface quality and lowers optical losses, which yield cell efficiency of up to 20.4% (Chirilă et al., 2013; Malitckaya et al., 2017). In addition to alkali metals, several other impurities (e.g., Sb and Zn) have been added to enhance cell efficiency. The Sb element is used to promote the growth of the preferred grand structure of CIGS film under relatively low-temperature conditions and to improve carrier concentration and the lifetime of the solar cells (Mansfield et al., 2015; Puyvelde et al., 2014). Moreover, the Zn can be used to control carrier concentration where Zn can be formed as an acceptor to yield the ZnCu donor, which makes the CIGS film into *n*-type. The three-stage deposition process was used to obtain the correct composition of Zn to achieve a *p*-type CIGS absorber layer with high carrier concentration. As previously reported, Zn-doped CIGS exhibits an efficiency of 14.5% and Voc of 0.658 V (Shirakata, 2017). We summarize the recent work on CIGS doped by alkali metals, Zn, and Sb using various techniques such as using an SLG substrate, introduction of an additional buried layer under CIGS, PDT, evaporation during the growth process, and the addition of solvent in the Cu–In–Ga precursor, as depicted in Fig. 3c.

To enhance the cell performance, manipulation of surface morphology of the CIGS absorber layer can be adopted (Han et al., 2012; Ishizuka et al., 2011; Liao et al., 2013; Liu et al., 2011; Su et al., 2011). This approach relates to the deposition of *n*-type buffer layers on CIGS layer, elemental inter-diffusion at CIGS/buffer layer interfaces, presence of ordered vacancy defects, compositional deviation, and formation of concomitant *p*–*n* junction (Han et al., 2012; Ishizuka et al., 2011; Su et al., 2011). Controlling Se flux during CIGS growth affect the CIGS film morphology, and thus concomitant solar cell parameters such as Voc and fill factor (FF) due to the high reactivity of the Se (Ishizuka et al., 2011). Moreover, the morphology of CIGS also depends on the preparation of Cu–In–Ga precursors; wherein, the rough surface of the precursor results in the CIGS thin films with poor crystallinity (Han et al., 2012; Su et al., 2011). A smooth surface of the CIGS film increases reflection, which is lowering cell efficiency (Liao et al., 2013; Liu et al., 2011). Therefore, an antireflection layer is deposited to reduce Fresnel reflections on the cell surface. However, the thermal mismatch between the antireflection layer and the device can affect reliability degradation. Thus, the nanostructured surface morphology of CIGS was developed to overcome this problem. Specifically, a surface with conical arrays was created that has a perfect antireflection effect on the gradual refractive index, as depicted in Fig. 4 (Cai and Qi, 2015; Liao et al., 2013; Liu et al., 2011; Shirakata, 2017). The nanostructured CIGS film surfaces such as with nanopillar, nanowire, and nanotip arrays induce the broadband and characteristics of omnidirectional light-harvesting that enable shorter carrier diffusion length and higher electron-hole pairs to increase the cell efficiency. Compared with conventional CIGS (Fig. 4a), nanostructured CIGS (Fig. 4b) is predicted to possess lower reflectance and higher light absorption, which enhances cell efficiency even with a much thinner

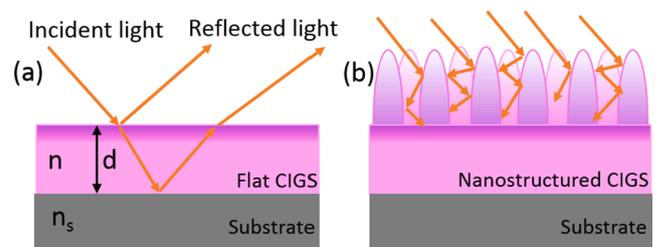


Fig. 4. Modification of the surface structures and morphology of CIGS thin-film affects its optical properties. (a) Propagation of incident light through a flat surface of the CIGS layer ( $n_s > n$ ). (b) Multiple internal reflections of incident light in the nanostructures CIGS layer. Adopted from (Cai and Qi, 2015).

thickness. Besides, an elemental concentration modification at the nanostructured surface decreases shunt leakage and series resistance and facilitates the formation of CdCu at the CdS/CIGS interface to enhance carrier extraction, interface passivation, and better adhesion of the window layer on the surface (Liao et al., 2013; Liu et al., 2011).

### 3.2. Extrinsic structure

The optimization of the cell efficiency can be reached by manipulating the optical and electronic structure through the external components of the cell, which consists of the substrate/electrical back contact/CIGS/*n*-buffer layer/electrode window layer, as depicted in Fig. 5a. In the basic configuration of CIGS-based solar cells, the *n*-buffer layer transmits light to the CIGS where the electron-hole pairs are mainly generated. Electrons within the diffusion length region are flowing from *p*-type CIGS absorber layer to the *n*-type semiconductor buffer layer and collected by the electrode window layer due to a built-in electric field across the *p*–*n* junction interface. In the same way, holes are flowing from the *n*-type semiconductor layer to the *p*-type absorber CIGS layer and collected by the electrical back contact. An additional mechanism in the CIGS layer is the back surface field (BSF) created by a Ga gradient that reflects electrons toward the *p*–*n* junction and finally collected by the *n*-type electrode BSF reduces minor carrier recombination at the interface of CIGS and the electrical back contact side of the device (Ramanujam and Singh, 2017; Singh et al., 2014; Yadav et al., 2015).

This mechanism can be maintained by choosing proper multilayer parts to obtain a desirable structure with optimal performance. The substrates have a significant impact on the performance of CIGS (Badgujar et al., 2015; Kessler and Rudmann, 2004; Ye et al., 2010). For example, the incorporation of indirect doping of Na from the SLG substrate affects the inter-diffusion kinetics of Cu–In–Ga–S elements, which result in a changed gradual bandgap profile and electronic properties, as shown in Fig. 5b. The incorporation of Na elements improves solar cell parameters (Voc, FF, and efficiency) due to reducing interfacial recombination, wherein, Na bonded to Se as NaSe<sub>x</sub> creates acceptor-type NaIn defects and forming NaCu defects, which reduces Cu-deficient at the surface and charge compensation. The Na reduces the number of compensating V<sub>Se</sub> donors, which enhances hole concentration and improves conductivity. Moreover, Na tends to react with oxygen and induces the indium oxide and gallium oxide formations. Na also can decrease the Ga/(Ga + In) ratio toward the center of the CIGS layer and created a gradient of bandgap in the CIGS layer. Moreover, the defect formation by Na improves the structural morphology of CIGS grains (Ramanujam and Singh, 2017).

Similar to the SLG substrate, CdS, which is usually used as an *n*-buffer layer, can also improve cell performance by the incorporation of Cd through CdS/CIGS interfaces, as shown in Fig. 5c. Cd penetrates far into the CIGS, which results in an *n*-type material and leads to the formation of a buried *p*–*n* homojunction in CIGS. However, the incorporation of Cu possibly occurs and results in Cu<sub>2</sub>Se, which is a known semiconductor with a bandgap of approximately 1.2 eV (He et al., 2019). The Cu<sub>2</sub>Se

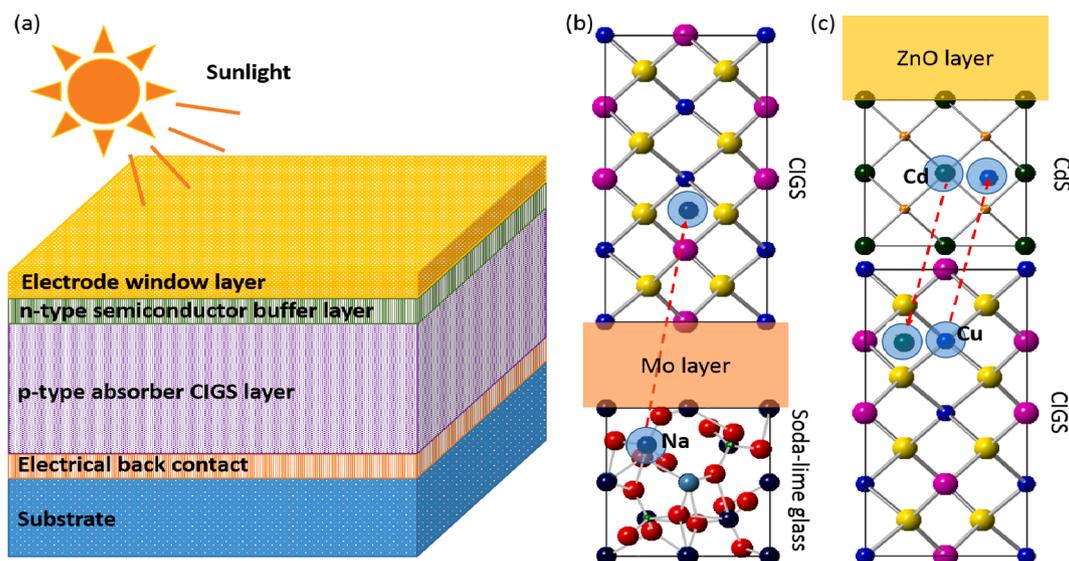


Fig. 5. (a) Basic multilayer structures of the CIGS thin film-based solar cell. (b) SLG substrate can enhance the efficiency of CIGS by incorporating Na. (c) Incorporation of Cd and Cu into the CIGS and CdS buffer layers, respectively.

which is present on the surface is highly conductive lowers the cell performance. The  $\text{Cu}_2\text{Se}$  segregation into the grain boundaries lowers shunt resistance, which results in low solar cell parameters. Therefore, the performance of Cu-rich CIGS solar cells is limited because of high interface recombination (Depredurand et al., 2011; Depredurand et al., 2014). In contrast, Cu-poor CIGS solar cells reduce recombination between the CIGS/CdS interface, which results in better solar cell efficiency. In addition, controlling the thickness of the CdS layer is also important because the CdS buffer layer absorbs high energy photons ( $\sim 2.4$  eV) and transmits them into CIGS where the electron-hole pairs are primarily generated. Thus, it is essential to growing a thick CdS layer. Nevertheless, the growth of a CdS layer followed by the intrinsic and doped ZnO layer, which acts as a window layer, can be adopted to reduce the production cost by eliminating toxic Cd waste (Heriche et al., 2016). Using this approach, the use of CdS can be reduced by decreasing the CdS layer thickness, while ZnO works as an emitter layer. The influence of Cd as a donor can be achieved by incorporation Zn impurities, which are also donors. Several materials have been tested as substitutions for the CdS layer such as  $(\text{Zn}, \text{Mg})\text{O}/\text{Zn}(\text{O}, \text{S}, \text{OH})$ ,  $\text{ZnS}(\text{O}, \text{OH})$ ,  $\text{Zn}(\text{O}, \text{S})$ ,  $\text{ZnSe}$ , and  $\text{In}_2\text{S}_3$  (Bhattacharya and Ramanathan, 2004; Kushiya, 2004; Kushiya et al., 2001). Specifically, a Zn-based buffer layer improves  $J_{sc}$  owing to its wider bandgap of  $E_g = 3.3$  eV compared to that of CdS bandgap with  $E_g = 2.42$  eV (Bhattacharya and Ramanathan, 2004; Kushiya, 2004). However, when there is an excess of oxygen during the deposition of the ZnO layer, this results in the formation of  $\text{CdZnSO}$  in the CdS layer, which leads to the uncontrollable diffusion of Zn further into the CIGS and in increased junction recombination and reduced Voc (He et al., 2019).

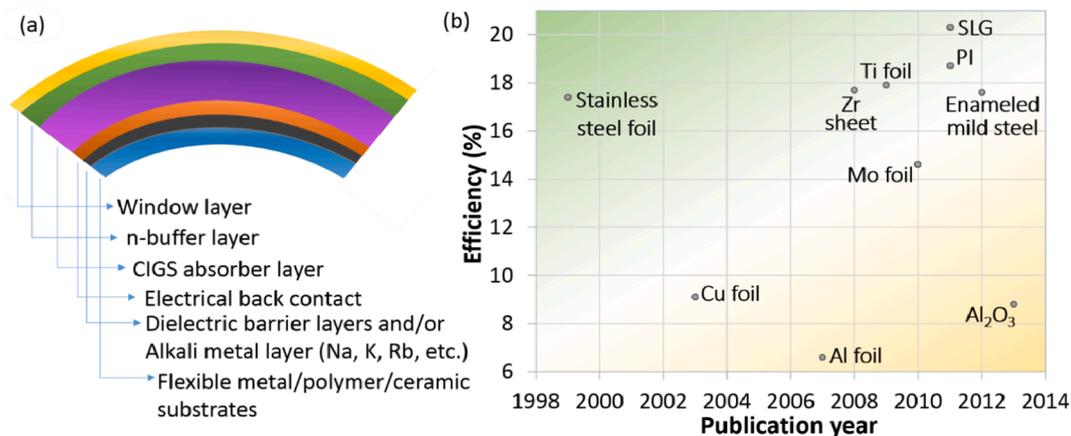
As previously mentioned, indirect doping used to enhance the efficiency of the cell can be realized with a buried layer. The control of Na incorporation into the CIGS film can be achieved by an additional layer before growing CIGS. Hence, it is possible to use flexible and rigid metal, polymer, or ceramic substrates. Moreover, additional layers, such as antireflection and grid layers, need to be account to obtain high efficiency.  $\text{MgF}_2$  is frequently used as an antireflection layer to reduce and to control Fresnel reflection between air and the solar cells device interface (Bhattacharya and Ramanathan, 2004; Ramanujam and Singh, 2017; Repins et al., 2008).

Mo is a common material that is used as an electronic back contact. Besides, Mo can act as a reflector to reflect unused light back into the absorber. Wherein, an improvement in reflection from the backside is exhibited when a Mo–Cu alloy is employed as a rear contact

(Ramanujam and Singh, 2017). Some groups use ITO as back contacts, which performs almost the same function as metallic Mo back contacts. ITO substrate is also common material that is used as a front contact, and its function is similar to that of a grid (Nakada et al., 2004). The deposition of a nontransparent but highly conductive metal grid is employed to collect current and to reduce resistive losses from the low conductivity top contact and to reduce long-term effects of degradation (Bhattacharya and Ramanathan, 2004; Li et al., 2011; Repins et al., 2008). An increase in efficiency has been shown in CIGS-based solar cells with a grid compared to that of the ungrided design (Li et al., 2011).

Fig. 6a shows the possible construction of flexible CIGS-based solar cells. Flexible CIGS-based solar cells are a special topic that will be briefly explained in this section. Roll-to-roll manufacturing of CIGS-based solar cells on flexible substrates enables the use of compact, high throughput, low thermal budget, lightweight, and more flexible deposition equipment than that used to produce rigid cells. Currently, flexible substrates can be categorized into metal, polymer, and ceramic. Metals can withstand very high deposition temperatures. However, they usually have high density, are rough, and contain metallic impurities; yet, their reactivity with Se is essential for the solar cell performance. Therefore, the deposition of intermediate dielectric barrier layers is usually used to provide electrical insulation between electrical back contact and the substrate, serves as a diffusion barrier against impurities from the substrate, and also lowers surface roughness. Compared to metals, polymer substrates have lower roughness and density; polymers are lightweight and electrically insulating. Polymer substrates cannot sustain high temperatures above  $500$  °C, which is commonly used for high-efficiency cell manufacturing. Thus, low-temperature deposition is used, which results in a low-quality absorber layer. To solve this problem, ceramics, such as zirconia, have been recently used as a flexible substrate with solar cell efficiency that is comparable to those on enameled mild steel, stainless steel, or Ti (Dhere et al., 2004; Kessler and Rudmann, 2004; Qiao et al., 2018; Reinhard et al., 2013). However, their brittle behavior may be an issue for large-scale industrial production.

We compile a recent work on CIGS-based solar cells grown on flexible and rigid substrates in Fig. 6b. Wherein, the cell efficiency of using flexible substrates can be similar to that of the cells fabricated on rigid substrates. Development and optimization can be conducted to obtain higher efficiency flexible CIGS-based solar cells, such as by direct and/or indirect doping to the CIGS layer, selective intermediate dielectric barrier layers, manipulation of CIGS surface morphology, control of CIGS



**Fig. 6.** (a) Flexible CIGS-based solar cells. (b) The efficiency of the CIGS-based solar cells grown on several substrates (Brémaud et al., 2007; Chirilă et al., 2011; Contreras et al., 1999; Ishizuka et al., 2008; Jackson et al., 2011; Niki et al., 2010; Sun et al., 2017; Tober et al., 2003; Wuerz et al., 2012; Yagioka and Nakada, 2009).

structure orientation, reduction in optical and electronic losses. Reduction in optical and electronic losses can be done by optimizing the *n*-buffer, grid, window, and antireflection layers by adjusting the bandgap grading (Dhere et al., 2004; Kessler and Rudmann, 2004; Qiao et al., 2018; Reinhard et al., 2013).

Both intrinsic and extrinsic structure engineering have advantages and disadvantages. Herein, we compile them in Table 1 for consideration to fabricate a highly efficient CIGS-based solar cell. Particularly, a CIGS-based tandem solar cell will be explained more specifically in the next section. Both intrinsic and extrinsic structure engineering is important to obtain CIGS-based solar cells with magnificent performances, even though, they have a disadvantage. By structural engineering using appropriate fabrication techniques, the combination of various materials, substrates and tandem cells can be carefully selected, which undoubtedly would lead to high efficiency CIGS-based solar cells for renewable energy applications.

#### 4. CIGS-based tandem solar cells

The development of tandem CIGS solar cell structure has attracted attention due to the possibility of overcoming the Shockley–Queisser limit of single-junction devices (Chae et al., 2016; Moon et al., 2015). As shown in Fig. 7a, a tandem solar cell consists of the top wide bandgap and bottom narrow-bandgap cells that absorb the short- and long-wavelength parts of the light, respectively (Elbar et al., 2015; Elbar and Tobbeche, 2015). To construct a tandem solar cell, it is essential to obtain an optimal bandgap combination of a two-junction device (Todorov et al., 2016). CIGS is an excellent material for tandem cells since it has tunable bandgap which depends on composition ratios. However, it is difficult to attain high-efficiency tandem cells such as monolithic CIGS/CIGS tandem cells due to the destruction of the subcell during the manufacture of the top cell (Chae et al., 2016; Moon et al., 2015). Nevertheless, numerous types of solar cells can be combined with CIGS cells, such as other thin film-based solar cells such as CIGS/CuGaSe<sub>2</sub> (CGS) and CdZnTe/CIGS, which have theoretical efficiencies of 25% and 26%, respectively (Elbar et al., 2015; Elbar and Tobbeche, 2015). Moreover, organic-based solar cells include dye-sensitizer solar cells (DSSCs) (Chae et al., 2016; Moon et al., 2015), perovskite-based solar cells (Guchhait et al., 2017; Shen et al., 2018; Todorov et al., 2016, 2015), and Si-based solar cells (Blanker et al., 2016; Lee et al., 2018) which have been recently produced in tandem with CIGS-based solar cells.

For example, CIGS cells satisfy the requirements for the top cell to be formed with the CIGS cell, where the fabrication of this tandem solar cell is simple and cheap due to the inexpensive raw materials. The tandem solar cell of CGS/CIGS was reported to achieve a high Voc of 1.18 V with

an efficiency of 7.4%. However, the current density mismatch between the subcells induces a significant current loss. Therefore, an optimization by adjusting the thickness of the top CGS absorber is conducted to obtain an optimal value of Jsc (Elbar et al., 2015; Elbar and Tobbeche, 2015). Moreover, the CIGS cell can be combined with a DSSC subcell, wherein, the optical bandgaps of DSSC of around 1.7 eV and CIGS of around 1.1 eV are suitable for use as top and bottom cells, respectively. From the manufacturing costs viewpoint, tandem solar cells of DSSC/CIGS would be competitive solar cells due to their simple preparation using solution processes. In DSSC/CIGS, the voltage and power conversion efficiency was enhanced compared to that of single-junction solar cells, and the cells showed an efficiency of 15%. However, the use of iodide electrolyte leads to a serious instability issue because of the *p-n* junction corrosion (Diantoro et al., 2019; Moon et al., 2015). Thus, deposition of the ZnO/TiO<sub>2</sub> protection layer, a soft deposition of Pt on the CIGS subcell, and substitution of the cobalt complex-based redox electrolyte have been conducted to overcome this problem (Chae et al., 2016; Moon et al., 2015).

Perovskite solar cells are ones of high efficient solar cells with high absorptivity, small exciton binding energy, and long carrier diffusion lengths (Guchhait et al., 2017; Maryam et al., 2019a). However, corrosion and decomposition are the main issues that need to be overcome in perovskite solar cells similar to DSSC solar cells. Nevertheless, perovskite solar cells can be used as a tandem to enhance cell efficiency. Perovskite/CIGS solar cells have excellent potential because they can be manufactured on lightweight and flexible substrates, which offers wider applications and low-cost production (Maryam et al., 2019a, 2019b; Todorov et al., 2016). The recently reported efficiencies for the mechanically stacked two-terminal (2-T) and 4-T perovskite/CIGS tandem solar cells are 19% and 23.9%, respectively (Guchhait et al., 2017; Shen et al., 2018; Todorov et al., 2015). Moreover, another approach for tandem solar cells is to produce a-Si:H/CIGS solar cells. The construction of a top a-Si:H subcell does not affect the bottom CIGS subcell because it requires a low-temperature deposition, which can be as low as 200 °C. Using the top a-Si:H solar cell, the CIGS cell thickness can be reduced more, and the use of expensive material, In, can be minimized. Yet, the use of CdS/AZO window layers in CIGS cells can be replaced because a-Si:H can absorb a large part of the photons with its wide bandgap. Recently, a-Si:H/CIGS showed a Voc = 1.23 V, FF = 64%, Jsc = 9.95 mA/cm<sup>2</sup>, and an efficiency of 7.9% (Blanker et al., 2016; Lee et al., 2018). We compiled the performance of several CIGS-based tandem solar cells as shown in Fig. 7b, and we observed a gap between simulation and experimental results. It is also important to note that the cell efficiency of the CIGS-based tandem solar cells was influenced by the architectures. The efficiency of CIGS-based tandem solar cells can be explored more by modifying the crystalline and electronic structures of

**Table 1**  
Advantages and disadvantages of intrinsic and extrinsic structures engineering.

Structures engineering	Advantages	Disadvantages
<b>Intrinsic structure</b>		
Composition grading	<ul style="list-style-type: none"> <li>Improves optical and electronic properties (Ramanujam and Singh, 2017)</li> <li>Usable to obtain a precision and desire bandgap of the CIGS layer (Ramanujam and Singh, 2017)</li> </ul>	Complicated fabrication and difficulty in control Cu and Ga + In compositions (Noikaew et al., 2018; Ramanujam and Singh, 2017).
Doping	Improvement of CIGS layer microstructures lead to better cell performances (Asaduzzaman et al., 2016; Chirilă et al., 2013; Malitckaya et al., 2017; Puyvelde et al., 2014; Ramanujam and Singh, 2017; Salomé et al., 2013; Shirakata, 2017; Sun et al., 2017)	Uncontrollable doping leads to the formation of detrimental impurity (He et al., 2019)
Surface morphology	<ul style="list-style-type: none"> <li>Improve the antireflection effect (Cai and Qi, 2015; Liao et al., 2013; Liu et al., 2011; Shirakata, 2017)</li> <li>Modify elemental concentration of CIGS layer surfaces result in better interface junction with <i>n</i>-buffer as well as a window layer (Liao et al., 2013; Liu et al., 2011)</li> </ul>	<ul style="list-style-type: none"> <li>Difficult to obtain homogenous nanostructured CIGS film over a large area</li> <li>Damage elemental composition and difficult to control composition grading</li> </ul>
<b>Extrinsic structure</b>		
<b>Substrate</b>		
Rigid	SLG substrate is known as the most efficient substrate to achieve high-efficiency cell of 20.3% through the incorporation of Na into a CIGS layer (Asaduzzaman et al., 2016; Ramanujam and Singh, 2017; Repins et al., 2008; Salomé et al., 2013; Sun et al., 2017)	The cell cannot be modified for wider applications, particularly for flexible solar cell
Flexible	High throughput, low-thermal budget, lightweight and more flexible in use than rigid cells (Reinhard et al., 2013)	<ul style="list-style-type: none"> <li>Metal substrate are high density, rough, contain metallic impurities, and very reactive with Se (Reinhard et al., 2013)</li> <li>Narrow thermal deposition for polymer substrate</li> <li>The ceramic substrate is brittle (Ishizuka et al., 2008; Reinhard et al., 2013)</li> </ul>
<b><i>n</i>-buffer layer</b>		
CdS	Cd in CdS known as the best donor inducing <i>n</i> -type doping on the CIGS layer surface (Ramanujam and Singh, 2017; Repins et al., 2008)	Toxic (Ramanujam and Singh, 2017; Repins et al., 2008)
Zn-based	Improves the Jsc and the blue response of the solar cells due to its wider band gap compare than CdS (Bhattacharya and Ramanathan, 2004; Kushiya, 2004)	Uncontrollable Zn incorporating on CIGS layer leads to the formation of detrimental impurity (He et al., 2019)
<b>Additional layers</b>		
Back contact	<ul style="list-style-type: none"> <li>Mo metal has good electrical properties and low resistivity. Mo also can reflect unused light back into the CIGS layer (Ramanujam and Singh, 2017)</li> </ul>	<ul style="list-style-type: none"> <li>Mo metal layer is expensive and required an additional fabrication process. Mostly metal substrate is high density and rough (Reinhard et al., 2013)</li> <li>ITO has poor electric properties compare to Mo</li> </ul>

**Table 1 (continued)**

Structures engineering	Advantages	Disadvantages
	<ul style="list-style-type: none"> <li>ITO glass is transparent, cheaper than Mo metal and acts as an electrode substrate thus no need additional bottom electrode layer reducing fabrication cost (Nakada et al., 2004)</li> </ul>	metal. ITO has no elemental doping such as Na as SLG does, hence required direct or indirect doping to CIGS layer
Buried layers	<ul style="list-style-type: none"> <li>A layer containing alkali metals elements, Zn and Sb act as doping seeds (indirect doping) to improve CIGS layer microstructures leads to better cell performances (Bhattacharya and Ramanathan, 2004; Kushiya, 2004; Mansfield et al., 2015; Puyvelde et al., 2014; Shirakata, 2017)</li> <li>Dielectric barrier layer acts as electrical insulation between the metal substrate and electrical back contact and serves as a diffusion barrier against impurities from the substrate (Herz et al., 2002)</li> </ul>	Uncontrollable Zn incorporating on CIGS layer leads to the formation of detrimental impurity (He et al., 2019)
Window	Zn-based window layer reduce CdS usage and avoid front surface recombination (Bhattacharya and Ramanathan, 2004; Kushiya, 2004; Kushiya et al., 2001; Ramanujam and Singh, 2017)	<ul style="list-style-type: none"> <li>Leads to the formation of detrimental impurity (He et al., 2019)</li> <li>Reliability degradation due to thermal mismatch with the device</li> </ul>
Antireflection	Control and reduce Fresnel reflection at the interface between air and the device to enhance the efficiency (Bhattacharya and Ramanathan, 2004; Ramanujam and Singh, 2017; Repins et al., 2008)	<ul style="list-style-type: none"> <li>Reliability degradation due to thermal mismatch with the device (Liao et al., 2013)</li> </ul>
Grid	To collect current, reduce resistive losses and reduce long-term effects of degradation (Bhattacharya and Ramanathan, 2004; Li et al., 2011; Repins et al., 2008)	<ul style="list-style-type: none"> <li>Reliability degradation due to thermal mismatch with the device</li> </ul>
<b>Tandem solar cells</b>		
Thin film	Typical thin-film solar cell such as CGS cell is particularly satisfied due to ease fabrication and less usage of raw material components (Elbar et al., 2015; Elbar and Tobbeche, 2015)	Damage of the bottom sub-cell during the construction of the top sub-cell, especially when we use monolithic tandem construction (Elbar et al., 2015; Elbar and Tobbeche, 2015)
Organic	Low manufacturing costs due to its simple preparation of solution processes (Chae et al., 2016; Moon et al., 2015)	Corrosion and decomposition (Chae et al., 2016; Moon et al., 2015)
Perovskite	Its excellent cell efficiency compare than other class solar cells results in magnificent cell efficiency combination (Guchhait et al., 2017; Shen et al., 2018; Todorov et al., 2016, 2015)	Corrosion and decomposition
Si-based	Construction of top Si-based sub-cell will not affect the bottom CIGS sub-cell because it requires a low-temperature deposition, especially when we use monolithic tandem construction (Blanker et al., 2016; Lee et al., 2018)	<ul style="list-style-type: none"> <li>Tunnel recombination junction lowering electrical performance</li> <li>Cracks and a poor roughness induced by the grains of CIGS bottom sub-cell, especially when we use monolithic tandem construction</li> </ul>

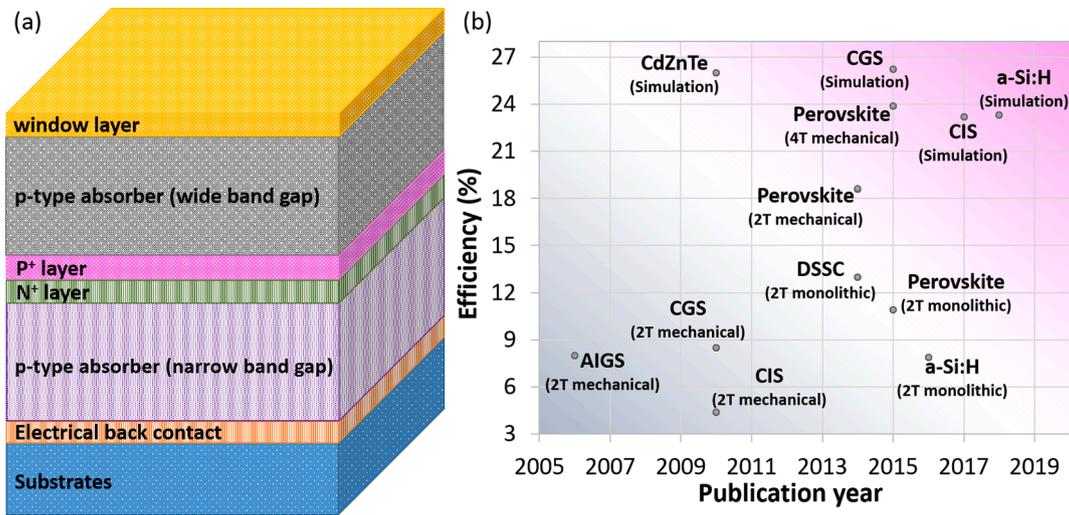


Fig. 7. (a) Basic structure of the tandem solar cell. (b) Compilation of cell efficiencies (theoretical and experimental) related to different tandem materials with CIGS-based solar cells using a variety of tandem architectures (Bailie et al., 2015; Blanker et al., 2016; Brémaud et al., 2007; Chae et al., 2016; Chirilă et al., 2011; Contreras et al., 1999; Elbar et al., 2015; Elbar and Tobbeche, 2015; Guchhait et al., 2017; Ishizuka et al., 2008; Jackson et al., 2011; Kaigawa et al., 2010; Lee et al., 2018; Moon et al., 2015; Nakada et al., 2006; Nanayakkara et al., 2017; Niki et al., 2010; Schmid et al., 2010; Shen et al., 2018; Shen et al., 2003; Todorov et al., 2016; Werner et al., 2018; Wuerz et al., 2012; Xiao et al., 2010; Yagioka and Nakada, 2009; Yamaguchi et al., 2018).

both subcells, as previously explained. Hence, the efficiency of CIGS-based tandem solar cells can approach the theoretical results.

As a comparison, we also compile the efficiency of the CIGS based solar cell with and without tandem as shown in Fig. 8. Superior cell efficiency could be obtained in tandem solar cell, however, degradation of the cell efficiency also could be emerged in this manner. The efficiency of single CIGS-based solar cells even higher than CGS/CIGS and a-Si:H/CIGS tandem solar cells presumably due to electronic inhomogeneity (Ramanujam and Singh, 2017; Werner et al., 2005). The fabrication process of subsequent subcell tandem on top of CIGS could be affected by the uncontrollable of the chemical structure of the CIGS layer. While a corrosion issue still the main problem when we use organic and perovskite as subcell tandem.

The performance of CIGS-based tandem solar cells also depends on their architecture, which is categorized on the interconnection scheme, and the fabrication sequence, for example, monolithic, mechanical stacks, and spectrum-split architecture as shown in Fig. 9a, the monolithic stack integration consists of a bottom and top cells, which are connected with 2-T in a series configuration. The subcells are electrically

connected by a conductive layer to transport the carrier through a recombination layer or tunnel junction from one subcell to another. The operation of these tandem devices may be similar to that of a single solar cell, which simplifies their integration in a photovoltaic system and requires fewer materials, fewer deposition steps, and lowers the production costs. Besides, only a single substrate is required, which reduces the series resistance losses associated with large-area modules. Based on Kirchhoff's law, the subcells that are connected in series equal to the sum of two subcells resulting in high voltages, which reduces resistive losses in the photovoltaic system. However, the series connection implies the need for current matching, which limits the choices of materials in terms of bandgaps and makes the system more sensitive to spectral variations. Thus, current matching must be maintained to have a similar photocurrent because the device performance will be lowered by the cell with the lowest current. Moreover, because the top cell is deposited onto the bottom cell, the fabrication process should not affect the bottom cell and the bottom cell must prevail as a suitable substrate with a suitably textured surface (Todorov et al., 2016; Werner et al., 2018; Yamaguchi et al., 2018).

Furthermore, a mechanically stacked tandem device consists of vertically stacked two separately developed cells. This architecture allows independent fabrication and offers process simplicity. Thus, optimal fabrication conditions that are specific to each subcell can be used (e.g., cell polarity, substrate roughness, process temperature, and solvents). Mechanically stacked devices can be divided into two designs depending on how many terminals are used, i.e., 2-T (Fig. 9b) and 4-T (Fig. 9c). Wherein, the tandem is constructed as a stack of two separate top and bottom cells but they are connected in series with two-terminal or four-terminal outputs, respectively. This tandem is not required for interfacial tunneling, and the current matching condition can be controlled, which widens the choice of the subcell bandgap and makes the system less sensitive to spectral variations. However, for this architecture to be viable, it is essential to minimize parasitic absorption and fabrication costs because of the doubling of all power electronics such as cables, inverters, and bifacial transparent electrodes (Todorov et al., 2016; Werner et al., 2018; Yamaguchi et al., 2018).

Moreover, spectrum-split architecture, which is still a 4-T device that consists of a dichroic filter or mirror that splits the light toward the high and low bandgap subcells, is shown in Fig. 9d. The subcells are operated individually without any integration. Thus, the choice of subcells and manufacturing system integration is very flexible. Each device can be

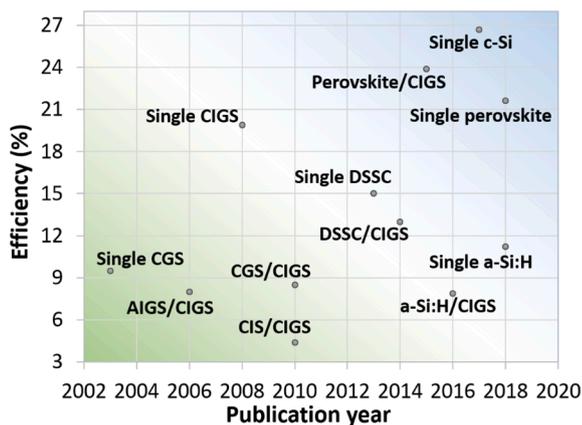


Fig. 8. A comparison of single CIGS-based solar cell and CIGS-based tandem solar cells. (Blanker et al., 2016; Chae et al., 2016; Fortes et al., 2018; Green et al., 2018; Kaigawa et al., 2010; Moon et al., 2015; Nakada et al., 2006; Niki et al., 2010; Repins et al., 2008; Schmid et al., 2010; Shen et al., 2018; Upadhyaya et al., 2013; Xiao et al., 2010; Yang et al., 2018; Young et al., 2003).

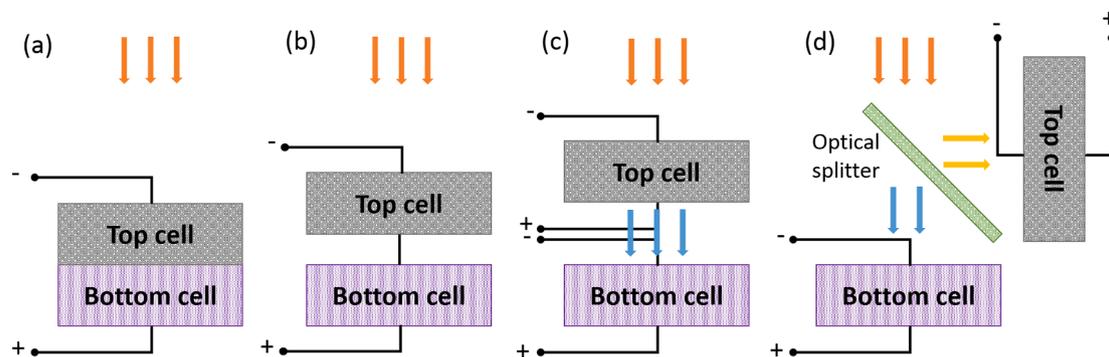


Fig. 9. Architectures of tandem solar cells. (a) 2-T monolithic, (b) 2-T mechanically stacked, (c) 4-T mechanically stacked, and (d) 4-T spectrum-split.

optimized as if it operates as a stand-alone solar cell and only requires two conductive layers, which reduces the optical loss. This architecture is advantageous because standard cells can be used without any specific adaptation. However, the added cost of a filter limiting the economic viability of this tandem architecture for nonconcentrated photovoltaic systems, and 4-T limits the commercial potential of this architecture. Such approaches, which require solar tracking, cannot efficiently collect the diffuse light present in the solar spectrum, and their performance may be strongly affected by module soiling. An example of this type of tandem has been demonstrated with an impressive total potential efficiency of 28% (Todorov et al., 2016; Werner et al., 2018; Yamaguchi et al., 2018).

## 5. Summary

The optimization and development of CIGS-based solar cells allow achieving over 20% light-to-electricity efficiency using various vacuum and non-vacuum deposition techniques. Modifications of intrinsic and extrinsic structures to tune their functionalities are considered to be a general route to enhance cell efficiency. The fabrication of intrinsic structures, such as composition grading, can be realized by controlling the amount of Cu–In–Ga on the front and back parts of the CIGS layer during the deposition process. Thus, similar to composition grading, the addition of an impurity or doping, such as alkali metal elements, during or after the deposition is a promising way to enhance cell efficiency. Doping can be achieved by incorporating the elements from substrates or an additional buried layer below the CIGS layer. Composition grading and doping are essential to manipulate the physical properties and structures of the CIGS layer. The structure and morphology of CIGS are important not only for tuning the optical and electrical bandgaps but also for the formation and elemental inter-diffusion at the *n*-buffer layer/CIGS interfaces, the concomitant *p*-*n* junction formation, the presence of compositional deviations or ordered vacancy defects and reflection behavior. Therefore, nanostructured CIGS presents a solution in addition to optimizing the proper *n*-buffer, window, grid, and anti-reflection layers. The choice of combination layers with the CIGS layer and modification of the extrinsic structure of the CIGS solar cell remains interesting research areas. Other promising research directions include the search for a CdS free layer, a transparent conducting layer, substitution of the famous SLG substrate to avoid the presence of an additional Na buried layer, and the selection of proper flexible substrates. The flexible substrate is essential for the development of functionalities of CIGS solar cells. Thus, several types of flexible substrates (e.g., metal, polymer, and ceramic) have been tested to grow flexible CIGS-based solar cells. The development of extrinsic structures leads to the creation of CIGS-based tandem solar cells. The combined cell efficiency increases because the wider energy regions of the solar spectrum can be converted into electricity. Several architectures of tandem solar cells (e.g., monolithic and mechanical stacks, and spectrum-split architectures) can be used to achieve high conversion efficiencies. Besides, CIGS solar

cells can be combined with other solar cells (e.g., thin-film, DSSC, perovskite- and Si-based solar cells) to realize an excellent tandem solar cell.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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