Progress in Major Thin-film Solar Cells: Growth Technologies, Layer Materials and Efficiencies

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Abstract- Thin film solar cells are desirable due to minimal material usage, cost effective synthesis processes and a promising trend in efficiency rise. In this review paper, remarkable progresses of five major types of thin film solar cell (TFSC) - amorphous silicon (a-Si) solar cell, copper indium gallium selenide (CIGS) solar cell, copper zinc tin sulfide (CZTS) solar cell, cadmium telluride (CdTe) solar cell and dye-sensitized solar cell (DSSC) have been presented from their inception to the state-of-the-art development. Cell configurations, different layers of these cells, their growth procedures, function and modification for working solar cells have also been explored. Critical issues that limit the performance of these cells as well as current scenario have also been addressed. Finally, a summary of this work has been presented as a comparative study among the five major types of TFSCs in terms of the state-of-the-art data for structural, optical, and electrical properties, material availability, toxicity, stability and highest efficiency for helping readers decipher the major challenges for popularization and commercialization of this technology.

Keywords a-Si:H; CIGS; CZTS; CdTe; DSSC; thin-film solar cell.

1. Introduction

The sun is the most abundant source of renewable energy to meet up the present and future global energy demands in tera-watt (TW) scale deployment. In this direction, solar cell is one of the best possible means for directly converting sunlight into usable energy without producing any harmful by products. In general, there are two approaches for the conversion of harvested sunlight into electricity namely, single junction and multijunction solar cells [1]. Among all types of solar cells, second generation thin film cells are favorable because of their low cost material usage and promising trend in efficiency rise. Although first generation silicon (Si) solar cells are still dominant players in photovoltaic (PV) literatures and covering around 95% of the PV market share globally [2], due to various limitations of this Si-solar cell such as weight, rigidity, cost of Si wafer, expensive processing technologies, alternative low cost materials and device fabrication technologies have been attracted growing interests over the past five decades. This led to the invention of the second generation light weight low cost thin-film solar cells (TFSCs). In TFSCs family, cuprous sulfide-cadmium sulfide (Cu₂S/CdS) single junction is the first reported solar cell developed for space applications that showed high efficiency (~9.1%), and several start-up companies have been established to commercialize this

technology but diffusion of copper into CdS matrix and doping of the CdS layer led to long-term performance degradation, for which most of the research activities on Cu₂S/CdS cells were declined [3, 4]. Eventually, other genre of TFSCs took over the researchers and manufacturers interests. Among these TFSCs, amorphous silicon (a-Si) solar cell is preferable to the researcher because of its availability. non-toxicity. material low processing temperature and low cost. Meanwhile, chalcopyrite based CIGS solar cells showed comparable efficiencies to the polycrystalline Si solar cell, but the production cost of this cell is reported to be much lower than that of the polycrystalline Si solar cell [5]. One of the major barriers in commercialization of this cell is the resource limitation, since Indium (In) and Gallium (Ga) are considered as scarce materials. On the other hand, quaternary semiconducting compound Copper Zinc Tin Sulphide (CZTS) is anatomically similar to CIGS but uses the earth-abundant elements that alleviate the material scarcity issue precluding CIGS cell potentials. Recently, a promising binary material: Cadmium Telluride (CdTe) has been showing much promises due to its frequently reported higher efficiencies comparable to polycrystalline Si solar cell [6, 7] despite its growth nature and crystal structure is remarkably different from other p-n junction based TFSCs. As a result, CdTe received more attentions than other TFSC materials as an absorber for achieving highly efficient and low-cost solar cell. Besides, non p-n junction based DSSC cells are unique among the TFSCs in terms of structure, light absorption capacity, electron and hole transport mechanism etc. It is also a promising candidate in TFSC technology, because of its cheap constituent materials without requiring higher purity, therefore, lowers the fabrication cost [8, 9].

Due to the above mentioned strategic key features in selecting solar cell materials, different research groups have been engaged in intensive research and development works as well as periodic literature review for knowing the latest advances in TFSCs. In recent time, review works on (a-Si:H) [10], CIGS solar cell [11], CZTS [12, 13], CdTe [14] and DSSC [15] are seen to be reported separately in the literature. Beside these single cell based review work, scientists also published some specific issue based review work of two or three prominent solar cells, such as, T.D. Lee and A.U. Ebong [16] reviewed the technologies and challenges of three major types (a-Si:H, CIGS and CdTe) TFSC. But, to the best of our knowledge, there is no such type of review work that covers five major types of TFSC in a single report and provides required information of these cells from their inception to the state-of-the-art development. So, in this paper, we briefly report and compare the evolution of growth technologies, constituent cell materials, critical issues that limit the cell performance, remarkable progress of their efficiencies, and present scenario of five major types of thin film solar cells.

2. Crystalline Silicon (c-Si) Solar Cell

As mentioned above, crystalline silicon (c-Si) PV cells are the dominant players in global photovoltaic market. Usually these cells are developed from the microelectronics technology. The first practical silicon solar cell with 6% efficiency was developed in Bell laboratory in 1954 [17, 18]. And since of its inception, this technology has been treated as one of the important renewable energy sources that could replace the environmentally detrimental fossil fuels. Silicon solar cells are made up of monocrystalline or polycrystalline silicon wafers. To date, the reported highest laboratory energy conversion efficiency of monocrystalline and polycrystalline solar cells are 26.7% and 22.3% respectively [6]. Besides, recently thin-film silicon solar cells are also drawing much attention among the PV community because this technology reduces the amount of active raw material usage in devising the cell. The chemical vapor deposition method is used for depositing silicon thin-film solar cell on suitable substrates from silane gas and hydrogen gas. Thus amorphous silicon, polycrystalline silicon or nanocrystalline silicon can be yielded depending on the deposition parameters and appropriate conditions [19]. In the following section we only discuss the amorphous silicon solar cell, because this cell is promising due to its strategic potentials, technological advantage, and overall low manufacturing cost.

3. Amorphous Silicon (a-Si) Solar Cell

3.1. Inception and remarkable progress

The first amorphous silicon (a-Si) layer was prepared as thin films of Si from silane (SiH₄) gas, deposited in a radio frequency glow discharge chamber in 1965 [20]. Ten years later, University of Dundee reported that the conductivity of a-Si can be increased by several times by doping performed in a gas mixture of phosphine or diborane in the glow discharge ambient [21]. This pure a-Si contains high concentration of defects termed as dangling bonds. Such defect densities could be reduced from 10^{21} cm⁻³ to 10^{15} -10¹⁶ cm⁻³by flowing hydrogen gas into the deposition chamber. The passivation of these deleterious dangling bonds [22] is called hydrogenation of a-Si (denoted by (a-Si:H)). The first a-Si:H solar cell was fabricated with a reported efficiency of 2.4% by Carlson and Wronski in 1976 [23]. However, due to Staebler-Wronski effect [24], the light-induced change in a-Si:H solar cell has been reported to degrade the efficiency (single-junction cell losses about 30% and triple-junction module losses about 15% of its initial efficiency after about 1000 hours [25, 26]). To overcome this cell degradation over the time, an intrinsic layer was incorporated within the a-Si:H cell for devising a final *p-i-n* structure. The remarkable chronological development of a-Si:H solar cells and materials used in the cell structures are summarized in the table 1.

Table 1. Remarkable chronological development of a-Si:H solar cells

Year	Efficiency improving mechanism employed	Reported efficiency	Researcher/ Organization
1981	First notable progress of efficiency for amorphous silicon solar cell was achieved. For that modifying the film of a-Si:H, the amorphous silicon carbide (a-SiC:H) was fabricated by the plasma decomposition of $[SiH_4(1-X)+CH_4(X)]$ in the presence of B_2H_6 or PH_3 dopant gas system and by utilizing this a-SiC:H, the first a-SiC:H/a-Si:H heterojunction solar cell was developed.	7.14% (S)	Osaka University [27]
1986	Intentional insertion of a thin graded interface layer at the p/i interface of p-i-n device, the overall performance of p-i-n cells was improved.	>8% (S)	Solarex Corp. [28]
1992	Besides single junction p-i-n cells, efficiency of a-Si:H cell was further enhanced by micromorph (Tandem or triple junction) approach. This was done by implementing an a-SiGe:H alloy in tandem and triple junction solar cell structures.	11.2% (M)	USSC [22]
1994	The first 'mixed stacked' a-Si:H/µc-Si:H tandem cell of the world was developed.	9.1% (M)	University of Neuchatel [29]
2000	By combining the active poly-Si cell with a-Si:H cell, a-Si:H/poly-Si/poly-Si cell was fabricated by plasma chemical vapor deposition (CVD) to improve the overall efficiency.	12% (M)	Kaneka Corporation [30]
2004	a-Si:H p-i-n single-junction (S) and tandem (T) cell or multijunction (termed as Micromorph) solar cells have been fabricated by very high frequency low pressure CVD.	9.47% (S) 12.3% (M)	NREL [31]
2015	Highest efficient single junction a-Si:H with improved p-i-n device structure has fabricated using diode and triode PECVD with various deposition rate.	10.2% (S)	AIST [32]
2015	State-of-the-art world highest stabilized efficiency achieved from a-Si:H/ μ c-Si:H/ μ c-Si:H triple-junction cells that is fabricated on periodically textured substrate.	13.4% (M)	AIST[33]

*S stands for Single junction and M stands for Micromorph

3.2. Cell Materials, Configuration and Fabrication procedure

To date, a-Si:H solar cell is configured in either the p*i-n* superstrate or the *n-i-p* substrate sequence. Layer materials of this type of structure have been modified for the performance improvement over the years. The a-Si:H thin film has a direct optical bandgap (Eg) and could be tailored in the range 1.6 - 1.8 eV by changing the substrate temperature or the dilution of silane by hydrogen during processing [34, 35]. In early a-Si:H solar cell, a p-type hydrogenated amorphous silicon carbide (a-SiC:H) absorber layer was incorporated with a comparatively wide bandgap material known as window layer [27]. In general, a conventional active device consists of a *n*-type window layer of a-SiC:H, an intrinsic a-Si:H absorber layer, and an *p*-type a-Si:H layer. In the latest a-Si:H cell configuration [36], absorber p-i-n layer is deposited on ZnO ($E_g \approx 3.20 - 3.40$ eV [37] transparent conducting oxide (TCO) layer [32, 38]. Initially, indium tin oxide (ITO) layer was used as TCO layer but to

date, cheaper alternative: fluorine doped tin oxide (FTO) has been widely used. In addition, this FTO (SnO₂: F) layer has been applied for reducing reflection loss and it serves as the top electrode. The front electrodes should have low electrical resistivity and high optical transmittance in the visible wavelength range for minimizing the ohmic contact loss and optical loss respectively. Usually, Al doped ZnO (AZO) is applied between n-layer and transparent front contact, but in another words, to increase the short-circuit current, the low resistive and high optical transparent ZnO:B [39] layer is applied between n-layer and silver as transparent contact. However, ZnO/Ag has the capability to overcome the adhesion problem between a-Si and metal contact. A schematic diagram of a-Si:H solar cell configuration is shown in Fig. 1

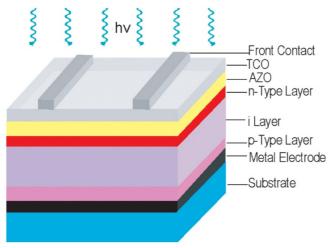


Fig. 1. Schematic diagram of a-Si:H solar cell adopted from [40]

Apart from this single junction configuration, a-Si solar cell could be configured as tandem or micromorph form. Here, it should be noted that hydrogenated amorphous silicon germanium (a-SiGe:H) is a desirable absorber material for stacked or micromorph solar cells due to its reported low bandgap ($E_g = 1.39$ –1.53 eV) [27, 41].

In a-Si:H thin film solar cell, absorbed photon in intrinsic layer creates an electron-hole pair and electric field induced across the intrinsic layer causes the electrons to drift towards the n-layer and the holes towards the p-layer. For improving the cell performance, a thin graded interface layer has been used to reduce the p/i interface defects which is responsible for low short circuit current and open circuit voltage [28, 38]. However, conventional a-Si:H cells are deposited on either SnO₂ or ZnO coated glass as front contact TCO. Plasma enhanced chemical vapor deposition (PECVD) is the widely used deposition method for devising this solar cell. However, other fabrication methods such as hot-wire CVD, photo CVD, sputtering, and electron cyclotron resonance chemical vapor deposition (ECR CVD) have also been applied in fabricating high efficient solar cells [42-45].

3.3 Critical issues

In our present study, we have found that the three most critical issues regarding material properties and device structure that impeding the performance and reliability of a-Si:H cell are: (1) light management schemes, (2) Staebler-Wronski Effect (SWE) and (3) Faster deposition processes. To increase the performance of this thin film cell significantly from the current level, these critical issues must be addressed. For the improvement of light scattering properties, optoelectronic quality of the front TCO material must be improved [10]. Staebler-Wronski effect is still under discussion, so it must be better understood for finding a suitable way to impede light-induced evolution of bondedhydrogen [46] from the cell material. Development of faster deposition processes are required for high- throughput manufacturing of this solar cell with same level efficiency achieved so far[47].

4. CIGS Solar Cell

4.1. Inception and remarkable progress

History of CIGS thin-film solar cell accounts that, the first CuInSe₂ (CIS) thin film was synthesized by Hahn in 1953 [48] and Bell Laboratories proposed this material as a photovoltaic material in 1974 [49]. The first CuInSe₂/CdS heterojunction photovoltaic devices were developed by coating of an evaporated n-type CdS atop the single crystal of p-CuInSe₂ in 1974 [50] and CIS solar cells started to receive much attention when the first high-efficiency (9.4%)thin film cells were reported by Boeing in 1981 [51]. To date, NREL is a leading research organization in CIGS solar cell development. Time to time they report remarkable improvements of efficiency of CIGS solar cell on rigid glass substrate. The record efficiencies of this solar cell reported by NREL were 17.1% in 1995 [52], 18.8% in 1999 [53] and 19.9% in 2008 [54]. For depositing absorber layer, glass substrate is the most commonly used substrate; however, current trend is to develop flexible solar cells on metal foils [55-57] and polyimide flexible substrate [58-60]. In 1992, Mo, Ti, and Al foils have been used for the first time by International Solar Electric Technology (ISET) for flexible solar cells [61] and state-of-the-art highest efficiency CIGS solar cells were prepared on polymer foil 2013 [62]. Notable chronological development of CIGS solar cells deposited on both rigid glass substrate (R) and flexible glass substrate (F) are summarized in table 2.

Year	Efficiency improving mechanism employed	Reported efficiency (Substrate type)	Researcher/ Organization
1976	First notable progresses of efficiency for CIGS thin film solar cell were achieved. The CIGS (p-CuInSe ₂ /n-CdS) photovoltaic cell was developed through two modes of operation (illumination through CuInSe ₂ or through CdS.) under 100 mW/cm ² tungsten-	4.5% (R)	University of Maine [63]

	halogen illumination for 1.2 cm ² devices.		
1981	First high efficiency $CuInSe_2$ solar cell was made by vacuum deposition coating of an evaporated n-type CdS atop the crystalline p-CuInSe ₂ onto inexpensive substrates.	9.4% (R)	Boeing [51]
1993	The absorber layer has been deposited on flexible Mo foil by using an e-beam evaporation process with subsequent selenization.	8.3% (F)	AIST [61]
1995	Incorporation of Ga into the CIS matrix to raise the absorber bandgap, open-circuit voltage and a fill factor that led to achieve high efficiency CIGS cell fabricated by PVD of the constituent elements on the Mo-coated soda lime glass substrate.	17.1% (R)	NREL [52]
1996	Using alternative of metal foils substrate, the first report in the use of polyimide as a flexible substrate and a CIGS cell was developed by a two-step process of sputtering metal precursors.	8.7% (F)	ISET [58]
1999	A ZnO/CdS/Cu(In,Ga)Se ₂ /Mo polycrystalline thin-film solar cell was deposited on glass substrate that led to additional improvement of efficiency.	18·8% (R)	NREL [53]
2008	The performance of this cell has been further improved by the reduction of recombination during CIGS deposition. This reduction of recombination was achieved by terminating the three stage deposition processing with a Ga-poor (In-rich) layer.	19.9% (R)	NREL [54, 62]
2010	The highest efficiency achieved for Mo substrate where CIGS absorber layer deposited on Mo flexible substrate by using a three-stage co-evaporation process.	14.6% (F)	AIST [64]
2011	The highest efficiency achieved for Al foil substrate where CIGS cells were fabricated on flexible Al foil by using non-vacuum low cost printing setup.	17.1% (F)	Nano Solar & EMPA [5]
2012	The highest achieved efficiency for Mo/Ti-coated stainless steel (SS) substrates where CIGS cells were fabricated without an additional metal-oxide or metal-nitride impurity diffusion barrier using roll-to-roll processing.	17.7% (F)	EMPA [65]
2013	Based on flexible substrate, highest achieved efficiency for CIGS solar cells were prepared on polymer foil.	20.4% (F)	EMPA [62]
2017	Champion CIGS solar cell was fabricated by CIS absorber engineering and enhanced surface treatment of the absorber layer using a co-evaporation process on rigid glass substrate.	22.5% (R)	AIST [66]

Note: AIST: Institute for Advanced Industrial Science and Technology, ZSW: Zentrum für Sonnenenergie- und Wasserstoff-Forschung, ISET: International Solar Electric Technology, Inc., EMPA: Swiss Federal Laboratories for Materials Science and Technology.

4.2. Cell materials, configuration and fabrication procedure

The active materials of CIGS solar cells are composed of direct band gap tetrahedrally bonded compound-element semiconductors having chalcopyrite crystal structure. CIGS solar cell has been formed by alloying any proportion of CuInSe₂ with CuGaSe₂. However, most of the high performance devices maintain a ratio of Ga to (In + Ga) in the range of 0.25-0.35 [67] and this Ga content is varied to tune the band gap to values 1.04 eV for CuInSe₂ and 1.68 eV for CuGaSe₂ [35, 68]. At the very beginning, CuInSe₂ solar cell was a simple p-CuInSe₂/n-CdS heterojunction [63] but present day CIGS solar cell has been configured in structure: substrate/Mo/CIGS/CdS/i-ZnO/AZO/metal contact (Al) [69-72]. A typical configuration of a CIGS solar cell is shown in fig. 2.

For a rigid substrate, soda lime glass (SLG) is commonly used. However, flexible substrates such as polyimide or metal foil also have also been intensively used in this technology [73]. In CIGS solar cell, a sputtered molybdenum (Mo) layer is used as a back contact as well as a reflector to bounce the most unabsorbed light back into the absorber layer. A p-type CIGS absorber layer is grown on sputtered Mo layer. In state-of-the-art high efficiency CIGS solar cell, the absorber layers are grown by physical vapor deposition such as thermal co-evaporation at high temperature (~ 600 $^{\circ}$ C) and a two stage process such as sputtering of precursor layers followed by selenization in Se vapor or H₂Se gas [5].

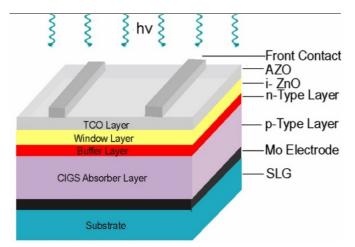


Fig. 2. Schematic diagram of CIGS solar cell adopted from [74]

A thin n-type buffer layer is put atop the absorber. Usually chemical bath deposited CdS is used as buffer layer but in consideration of environmental and hazardous effect, extensive research has been conducted for developing cadmium (Cd) free buffer layers. Moreover, ZnS, ZnSe, ZnO, (Zn,Mg)O, In(OH)₃, In₂S₃, In₂Se₃, InZnSe_x, SnO₂ and SnS₂ have been used as buffer layer and these layers are deposited by various growth methods such as metal organic chemical vapor deposition (MOCVD), chemical bath deposition (CBD), electrodeposition, sputtering, thermal evaporation, atomic layer deposition (ALD), ion layer gas reaction (ILGAR) [75], and pulsed laser deposition(PLD).

For avoiding surface damage of CdS layer an in intrinsic ZnO layer (i-ZnO) which is capped by a relatively thicker Al:ZnO(AZO) layer is added atop the buffer layer. Chemical vapor deposition method is merely used to deposit the ZnO layer [76]. The AZO layer is used as a TCO to collect and transport electrons out of the cell while transmitting as much light as possible.

4.3. Critical issues

In this review, we have identified that the critical issues of CIGS solar cells are: (1) developing a viable deposition process onto a flexible substrate at lower temperatures, (2) CdS buffer layer conventionally deposited by CBD that presents the greatest health concerns due to the use of carcinogenic Cd, thiourea, and the generation of waste solutions. Selenium (Se) does not show highly toxicity but compounds of Se such as H_2Se are extremely toxic [77]. (3) Material availability is also a major concern of the CIGS solar cell. Most of the constituent elements of this solar cell are abundant in the earth crust except the scarce metal Indium (In) and Gallium (Ga). Therefore, resource limitations as well as ecological issues shed a negative light on the practical potential of the CIGS solar cells in the case of terawatt scale deployment (and refs. there in). The search of alternative less toxic materials compared to CIGS solar cell gave birth to the CZTS solar cell made out of entirely earth abundant elements.

5. CZTS Solar Cell

5.1. Inception and remarkable progress

The first CZTS material was developed in 1967 by Nitsche et al. [78] using the chemical vapor transport method and its photovoltaic effect was observed by Ito et al. [79] in 1988. The first CZTS thin-film solar cell with efficiency of 0.66% reported by Katagiri et al. [80, 81] in 1997 using a vacuum-deposition method. The remarkable progress in the development of CZTS solar cells are presented in table 3.

 Table 3 Remarkable choronological development of CZTS solar cells

\sim		Progress of Efficiency	Researcher/ Organization
1997	First notable improvement of efficiency of CZTS solar cell was achieved by developing a thin film of Cu ₂ ZnSnS ₄ with a standard CdS/ZnO window layer by thermal evaporation of the elements or chalcogenide binaries in high vacuum.	2.3%	ZSW [82]
2007	An inline-type vacuum apparatus was introduced for the first time to achieve better quality of CZTS films than previously developed which truly lead to increase the efficiency. This CZTS solar cell was fabricated by three RF sources co-	5.7%	Nagaoka National College of Technology [83]

	sputtering continued with vapor phase sulfurization method.		
2009	Efficiency of CZTS was further notably increased by the same research group. Here absorber layer of CZTS was developed using three radio frequency sources co-sputtering followed by annealing in sulfurized atmosphere.	6.7%	Nagaoka National Colleg of Technology [84]
2010	Applying another novel techniques, kesterites CZTS solar cell was prepared from earth abundant materials by using composite liquid deposition approach which merging the concepts of solution and particle-based coating for chalcogenide materials.	9.6%	IBM [85]
2012	High efficiency kesterite CZTS solar cell was developed by hydrazine process, which improved the open circuit voltage, short circuit current, fill factor etc. that ultimately increased the device performance.	11.1%	IBM [86]
2013	State-of-the-art champion CZTS solar cell has been developed by pure hydrazine solution along with targeted Cu-poor and Zn-rich conditions that improved CZTSSe absorber quality which yield the heights efficiency.	12.6%	IBM [87]

5.2. Cell materials, configuration and fabrication procedure

layer to form p-n junction (beside this CdS buffer layer, another high bandgap materials such as CdZnS also found in

CZTS is a direct bandgap quaternary semiconducting compound. Kesterite crystal structure is most common structure for CZTS cells; beside this, chalcopyrite structure [88], wurtzite structure [89] and stannite structure [90] are reported in literature. However stannite and kieserite both forms can be coexist [91, 92]. The optical bandgap of stoichiometric kesterite-CZTS estimated theoretically is 1.50 eV [91] and experimentally, it is varied in the range 1.4 - 1.5 eV [93]. Its universally recognized raman shift peak is 338 cm⁻¹ [94]. The electrical resistivity of CZTS thin films is varied from $10^{-3}\Omega \cdot \text{cm}$ to $10^{-1}\Omega \cdot \text{cm}$ [91].

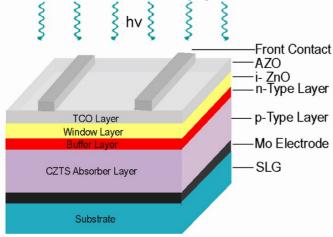


Fig. 3. Schematic diagram of CZTS solar cell adopted from [95]

Most of the CZTS cell consists of a sputteringdeposited Mo coated soda lime glass, a p-type CZTS absorber layer in contact with CBD grown n-type CdS buffer literature [96]), a thin layer of AZO is used as a window layer atop the CdS layer for preventing leakage. Metallic contacts made up of Ni/Al grid is separately deposited on both TCO and Mo layer. A schematic diagram of a typical CZTS solar cell is shown in fig.3. Several fabrication methods including vacuum-base, vacuum-free and solution process have been commonly used for CZTS absorber layer deposition [97-100]. The frequently used physical vapor methods are e-beam evaporation [93, 101], direct current (DC)/radio-frequency (RF) magnetron sputtering deposition [102], co-evaporation or electron beam evaporation [93], hybrid sputtering [94], thermal evaporation [103], pulsed laser deposition [104].

5.3. Critical issues

The major advantage of CZTS is that its raw materials are low cost, earth abundant and nontoxic [98] and about five times cheaper than CIGS [7]. However, in this review work, we have found a number of critical issues such as (1) toxic element of CdS buffer layer, (2) defect control of absorber, (3) pure CZTS phase generation and secondary phase detection and (4) bandgap engineering. Two toxic elements such as selenium (Se) and cadmium (Cd) are frequently used in CZTS precursors to grow high quality CZTS thin films. Although Se and Cd are toxic element but CZTS solar cell does not show toxicity. Besides, other non-toxic solventbased process has been used for the deposition of a CZTS absorber layer [105].

6. CdTe Solar Cell

6.1. Inception and remarkable progress

CdTe crystal was synthesized chemically for the first time by Margottet in 1879 [106] and its *p*-type and *n*-type conductivity was reported for the first time by doping foreign

impurity by Jenny and Bube in 1954 [107] and Loferski proposed it as a photovoltaic material in 1956 [108]. First single-crystal homojunction CdTe solar cell was demonstrated by Rappaport of about 2% efficiency fabricated by diffusion of Indium into *p*-type CdTe crystals in 1959 [109]. The historical development of CdTe solar cells are summarized in the table 4 below.

Year	Efficiency improving mechanism employed	Reported Efficiency	Researcher/ Organization
1972	The first notable progress of efficiency for a CdTe thin film heterojunction solar cell was reported. This graded gap thin film cell was prepared in a 3-step process, where high temperature vapor phase deposition (VPD) was used for CdTe film and high vacuum evaporation was used for CdS film.	6%	Bonnet and Rabenhorst [110]
1977	To prepare CdS/CdTe heterojunction solar cell, a CdS film was deposited on a chemically polished CdTe absorber by vacuum evaporation and introducing window layer (In_2O_3 : Sn) by RF sputtering.	7.9%	Stanford University [111]
1979	Fabricated CdTe thin film solar cell by close-space vapor transport deposition of p-type arsenic-doped CdTe films onto n-type crystals.	>7%	CNRS [112]
1982	Fabricated a CdTe shallow homojunction cell by close space vapor transport deposition technique to form p-type CdTe epitaxial layer upon a single crystal n-type CdTe layer.	11%	CNRS [113]
1993	Another remarkable progress of CdTe solar cell is to combine the $CdCl_2$ with thin- film CdS/CdTe heterojunction cell. Where n-CdS buffer layer was prepared by chemical bath deposition and <i>p</i> - CdTe absorber layer was deposited by close- spaced sublimation.	15.8%	University of South Florida [114]
2001	Employing vapor CdCl ₂ treatments and refinements of window-layer processing had led to additional improvements of this cell deposited on mesa glass substrate.	16.5%	NREL [115]
2009	Beside the CdTe solar cell deposition on rigid glass substrate, a CdS/CdTe cell was deposited on flexible lightweight polyimide film by using low temperatures (<450°C) vacuum evaporation technique and a subsequent annealing step in air where the materials are heated up and cooled down to harden them.	12.4%	EMPA [62]
2011	Significant improvement on the efficiency of heterojunction CdTe solar cell was reported compared to the previous cells developed by EMPA . Here the absorber layer of CdS/CdTe cell was developed by close space sublimation method.	17.3%	First solar Inc. [116]
2016	State-of-the-art highest efficient heterojunction CdTe solar cell has been developed by close space sublimation method.	22.1%	First Solar Inc. [6]

6.2. Cell materials, configuration and fabrication procedure

In CdTe crystal, the tetrahedral atomic coordination leads to diamond structure in monatomic solids and zincblende and wurtzite structures in binary solids [117]. The reported value of the bandgap of CdTe found in the literature are 1.44 eV; 1.464 eV; 1.50 eV; 1.51 eV and 1.56 eV at 300 K [22]. This CdTe solar cells could be configured both in substrate and superstrate arrangement. However, the state-ofthe-art high efficiency cells are found to be configured in superstrate arrangement [118, 119]. In early configuration, vacuum-evaporated CdS window layer and chemical vapor deposited CdTe absorber layer deposited on Mo substrate [110]. But, to date, the TCO works as window layer and CdS deployed as buffer layer. SnO₂, In₂O₃: Sn, ZnO, Cd₂SnO₄ are frequently used as TCO seen in literature. For TCO, typically fluorine-doped tin oxide (FTO) and undoped tin oxide (TO) bilayers has been deposited on the glass by MOCVD. The TCO serves as the front contact and lateral current carrying conductor. The CdS window layer could be deposited by CBD [119], sputter deposition [120] and close-spaced sublimation (CSS) [121]. Most of the CdTe absorber layer has been deposited by CSS on glass substrate and CdTe layer is then CdCl₂-vapor treated by CSS. It is reported that CdCl₂ treatment increases grain size, which significantly increase the conversion efficiency [121]. A schematic diagram of a typical CdTe solar cell is given in figure-4.

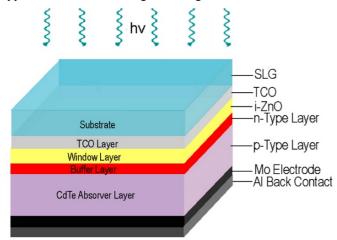


Fig. 4. Schematic diagram of CdTe solar cell adopted from [122]

The ZnTe:Cu/Ti back contact is deposited using a multisource vacuum processing chamber. Several methods have been employed to deposit CdTe absorber layer for solar cell

Table 5 Remarkable chronological development of DSSC	Table 5 Remarkable cl	hronological d	development of	DSSCs
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[35], such as physical vapor deposition (PVD), CSS, sputter deposition, electrodeposition, MOCVD, spray deposition, screen-print deposition and sintering but the best performing CdTe-based devices are fabricated using CSS [118, 123].

6.3. Critical Issues

Constituent elements of CdTe cell Cd and Te, respectively, are toxic and scarce materials. Cd is obtained as a by-product of zinc refining process from ore and at present it is an abundant material. In contrast, Te is obtained as a by-product of copper refining process from ore and it is a rare earth material [124]. As mentioned earlier, Cd is recognized as a carcinogenic elements, and classified as an environmentally hazardous material, but the compound CdTe itself is reported to be less toxic and very stable material [125]. Again the main perceived risks during the active lifetime of a CdTe solar panel are leaching out of broken modules and fire hazards. So toxicity of Cd and scarcity of Te are main critical issues limiting the commercialization potentials of this kind of solar cell.

7. DSSC Solar Cell

7.1. Inception and remarkable progress

Research on the photoelectric effect on organic dye was discovered at the end of nineteenth century. James Moser first observed that the photoelectric effect on silver plates was enhanced in the presence of erythrosine dye in 1887 [126] and in 1968, it was discovered that illuminated organic dyes can generate electricity at oxide electrodes in electrochemical cells [127]. Based on the experiment of understanding and simulating of the primary processes in photosynthesis, the dye sensitization solar cell (DSSC) principle was demonstrated and discussed in 1972 at University of California (UC), Berkeley [128]. However, in 1988, today's dye sensitized solar cell was co-invented by Brian O'Regan (who first invented the TiO₂ film) and Michael Grätzel (who invented the organic dye) at UC Berkeley and later, based on dye-sensitized colloidal TiO₂ films, a low cost, high efficiency cell has been developed by the aforementioned researchers at the École Polytechnique Fédérale de Lausanne (EPFL), Switzerland in 1991[129] and coined the term Grätzel cell. The summary of remarkable progress in the development of DSSC solar cell is given in table 5.

Year	Efficiency improving mechanism employed	Reported efficiency	Researcher/ Organization
1991	First notable improvement of efficiency for DSSC was achieved. This device was created based on an optically transparent film of TiO2 particles of $10-\mu$ m-thick and few nanometers in size. It was coated with a monolayer of charge-transfer dye to sensitize the film, a low cost, high efficiency cell has been developed.	7.1-7.9%	EPFL [129]

2003	For achieving the high temperature stability an amphiphilic ruthenium dye Z-907 { <i>cis</i> -RuLL'(SCN) ₂ (L= 4,4'-dicarboxylic acid-2,2'-bipyridine, L' = 4,4'-dinonyl-2,2'-bipyridine)} was used in conjunction with a quasi-solid-state polymer gel electrolyte. By this way a stable quasi-solid-state dye-sensitized solar cell was fabricated.	6.1%	EPFL [130]
2006	Instead of liquid electrolytes, first successful solid-hybrid dye- sensitized solar cells were reported . Backside illuminated dye sensitized solar cells was fabricated using N719 sensitized TiO_2 nanotube which was synthesized using anodization of Ti foils in electrolyte containing KF, NaHSO ₄ and trisodium citrate.	4.24%	Pennsylvania State University [131]
2007	Intensive research with a wide variety of low-cost organic dyes based on porphyrin was conducted. The porphyrins were used for preparing dye-sensitized TiO ₂ low-cost excitonic cells using an iodide/tri-iodide (I^-/I_3^-) redox electrolyte.	5.6%	Massey University [132, 133]
2008	A new solvent-free liquid redox electrolyte consisting of three salts was used in replacement of organic solvents. This ternary melt was used in conjunction with a TiO_2 film and the amphiphilic heteroleptic ruthenium complex Z907Na. This lead to achieve an excellent stability.	8.2%	EPFL, Changchun Institute of Applied Chemistry [134][137]
2011	Yella et al, reported a mesoscopic porphyrin-sensitized (designated YD2- o -C8) solar cells with cobalt (II/III)–based redox electrolyte instead of using I ⁻ /I ₃ ⁻ redox electrolyte that limited the efficiency of dye-sensitized solar cells.	12.3%	EPFL, National Chung Hsing University [135]
2013	Michael Graetzel et al. reported the state-of-the-art highest efficient solid state mesoscopic dye sensitized solar cell. They fabricated this cell using a hybrid perovskite CH ₃ NH ₃ PbI ₃ dye that subsequently deposited in two-step method from the separated solutions of CH ₃ NH ₃ I and PbI ₂ .	15.0%	EPFL, Max-Planck- Institute for Solid-State Research [136]

7.2. Cell materials, configuration and fabrication procedure

With respect to the materials and layer structure, DSSC is unique from the other types of thin-film solar cell. The DSSC is comprised of multiple layers stacked between two electrodes. Each electrode has a TCO film. Initially Indium-Tin-Oxide (ITO) is used as TCO layer, but, ITO has a low thermal stability of resistance, for this limitation, at present FTO is widely used. Moreover, other material also found in literature such as ZnO, Nb₂O₅ [137]. TiO₂ nanoparticles are sintered and bound to the FTO layer of an electrode.

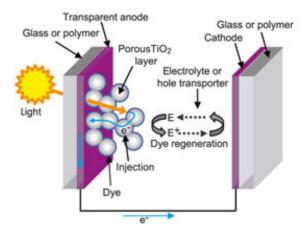


Fig. 5. A typical structure of a dye-sensitized solar cell

Dye is then absorbed into the TiO_2 layer. Likewise, platinum nanoparticles are sintered onto the FTO layer of the opposite electrode and both electrodes are then connected with either a two or three part liquid epoxy or a polymer sealant melts. Cell construction is completed with the incorporation of the

electrolyte into the center of the cell [138]. A general structure of a typical DSSC is shown in Figure-5.

A ruthenium dye was initially employed in the DSSC, but, to date, several organic dyes [139, 140] and inorganic dyes [141] have been reported for this genre. Grätzel first used organic liquid electrolyte containing LiI/I2. Later, many kinds of liquid electrolytes containing I^{-}/I_{3}^{-} redox couple and high dielectric constant organic solvents such as acetonitrile (AcN), ethylene carbonate (EC), 3-methoxypropionitrile (MePN), propylenecarbonate (PC), γ-butyrolactone (GBL), and N-methylpyrrolidone (NMP) have been investigated and found the photovoltaic effectiveness to some present day DSSCs[142, 143]. In replace of liquid electrolyte as the holetransporting medium, solid-state hole-transporting medium has also been used in this solar cell [144, 145]. To the best of authors' knowledge, widely used preparation techniques of TCO process by spin-coating or dip-coating, hydrothermal layer found in literature listed as spray pyrolysis technique, chemical vapor deposition (CVD), sol-gel method [146]. For TiO₂ deposition, screen printing [147], spray coating [148], electrophoretic deposition [149], spin coating [150] are used method in this field.

7.3. Critical Issues

Most of the raw materials of DSSCs are cheap and environmentally benign and fabrication procedure of this thin film cell is very simple. But a number of expensive materials such as platinum and ruthenium are still used in this cell. Long-term durability of the dye, leakage of liquid electrolyte for DSSC poses a major challenge in commercialization of this kind of cell usable in all weather. DSSCs are still in research and development stage, but for these cells, recently used polymer or metal foil substrates instead of glass substrate promote them towards the commercialization[151, 152].

8. Discussion

The main focus of this review work is to elucidate the remarkable progress of efficiencies of major five TFSCs along with the evolution of their fabrication procedures. Notable chronological improvements of these cells have been depicted in fig. 6.

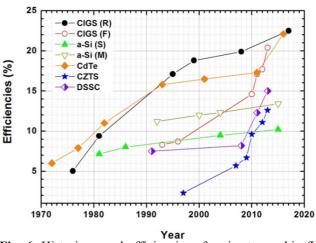


Fig. 6. Historic record efficiencies of major types thin film solar cells (Lab scale)

As can be seen, two separate efficiency lines (both single junction (S) and micromorph (M) cell) have been drawn for a-Si:H solar cells. The a-Si:H solar cell was initially fabricated as single junction solar cell but two- and three- junction (multijunction) cell configurations have also been developed for increasing the efficiency as well as the reliability [36]. In fact, multijunction solar cells are designed to capture the entire solar spectrum and overcome the single junction solar cell efficiency limitation [1, 153, 154]. However, due to higher bandgap and absorption coefficient. 90% of the visible radiation would be absorbed only in 1 micrometer amorphous thin film [22], but, due to the short crystalline order in a-Si film and the dangling bonds, the performance of this solar cell is not viable compared to the crystalline silicon solar cell. So, for improving the efficiency of this cell, reduction of reflection loss and enhancement of optical absorption is required. At the same time, surface textured substrate layer in combination with an antireflection coating must be introduced [155].

To date, CIGS cells have the comparable efficiencies to the polycrystalline Si solar cell. These cells could be deposited on both rigid glass substrate or flexible substrate and it was observed that the efficiency is higher when the cell is deposited on rigid substrate, i.e., soda lime glass substrate due to the positive effect of Na. In figure 6, two separate efficiency lines also can be seen for CIGS solar cell, one is based on rigid substrate (R) and another is for flexible substrate (F). Notice that the efficiency of CIGS cells deposited on flexible substrate gradually progressing towards the cell deposited on rigid substrate. It is already mentioned that manufacturing cost of this cell is lower than that of crystalline solar cell but scarcity of In and Ga is still the vital challenge. Most of the buffer layer of CIGS contain toxic cadmium content, which is another bottleneck limiting the potential of this solar cell. Similar to (CIGS), the CdTe is one of the most investigated materials among the second generation thin film solar cells. However, there are some issues regarding to its cost of raw materials and toxicity. CZTS is another promising thin film solar cell which is anatomically similar structured like CIGS. That is, fabrication procedure of different layers such as buffer,

window and TCO layer of CZTS is also similar to CIGS cell. Beside these, DSSC cells are unique among the TFSCs in terms of cell structure and constituent materials. This solar cell is the promising likewise the CZTS because of earth abundance of its constituent elements and their environmentally benign nature. Therefore, CZTS is one of the most desirable and adorable PV materials despites its' reported efficiency is lower compared to the CIGS and CdTe. A summary of the five TFSCs research works has been presented in table 6 as a comparative data for structural, optical, and electrical properties, material availability, toxicity, stability and highest efficiency.

Thin-film solar cells cover around 4.5% of worldwide photovoltaic market share where a-Si:H covers 0.3%, CIGS covers 1.9% and CdTe covers 2.3% of the market share in 2018 [1]. Both CIGS and CdTe solar cell enjoy the advantage of their matured fabrication technology, presently their

efficiency is comparable with multicrystalline silicon solar cell but their cost is comparatively lower than this type of Si solar cell. Both of these cells can be deposited on either rigid glass substrate or flexible glass substrate, both can be configured in either superstrate or substrate stacks. However, both of them use scarce and toxic materials which are the major limitations of these cells. Again, efficiency of a-Si:H is relatively lower in compare with CIGS and CdTe. and reduced after a certain time. In contrast, CZTS and DSSC are most promising solar cells in thin film family, because of raw materials' abundance, environmentally benign nature, lowcost as well as easy to synthesis and processing techniques. These solar cells are still in research and development phase, so its market share is not mentionable but their progress slowly moving forwards for commercializing in global PV market.

Table 6 Comparison of different properties of five major types thin film solar cell

Properties	a-Si:H Cell	CIGS Cell	CZTS Cell	CdTe Cell	DSSC Cell (approx.)
			Kesterite,		
Crystal			Chalcopyrite,	Diamond,	
Structure	NA	Chalcopyrite	Wurtzite [89],	Zincblende,	NA
		1.2	Stannite [90]	Wurtzite	
Lattice	NA	a = 0.56 - 0.58 nm	a = 0.5435 nm	0.648 nm	NA
constant		c = 1.10 - 1.15 nm	c = 1.0843 nm		
Bandgap	1.6–1.8 eV	1.04eV (CuInSe ₂)	1.4~1.5eV	1.50 eV [22]	3.2-3.35eV
	[35, 38]	1.68eV	[92, 93, 95]		TiO ₂ [156]
		$(CuGaSe_2)[68]$			1.7 eV Dye [157]
Absorption	$>10^5 \mathrm{cm}^{-1}$ [22]	$>10^{5} \mathrm{cm}^{-1}$ [158]	$10^4 \mathrm{cm}^{-1}$ [86]	$>5\times10^{5}$ cm ⁻¹ [159]	Adjusted [160]
Co-efficient					
Acceptor	$4.3 \times 10^{16} \mathrm{cm}^{-3}$	$10^{15} - 10^{16} / \text{cm}^3$	10^{16}cm^{-3} to		10^{18}cm^{-3} for
Concentr	Ionized [161]	[162]	10^{18}cm^{-3}	$>10^{14} \mathrm{cm}^{-3}$ [123]	Hole transport
Ation			[163-165]		medium [160]
Hole	$0.3 \text{ cm}^2/\text{V.s}$	250cm ² /V.s	$0.1-30 \text{ cm}^2/\text{V.s}$	$50-80 \text{ cm}^2/\text{V.s}$	1350×10^{-3}
Mobility	[166]	CuGaSe ₂ [167]	[79]	[168]	cm ² /V.s [160]
Electron	$2 \text{ cm}^2/\text{Vs}$ [166]	90-900 cm ² /Vs	$>5 \text{ cm}^2/\text{Vs}$	500–1000 cm ² /Vs	450 × 10 ⁻⁴
mobility		[169]	[157]	[168]	cm ² /Vs [160]
Toxicity	Non-toxic	Less toxic	Non-toxic	Toxic	Non-toxic
Material	Available	In and Ga	Available	Cd is available Te	Available
availability		are limited		is limited	
Stability	Efficiency	Very stable	Very stable	stable	Less stable
	Less stable	-	-		(Dye)
Highest Efficiency	13.4%	22.5%	12.6%	22.1%	15%
Market Share	0.3%	1.9%	R&D phase	2.3%	R&D phase

*Exact data is not available

9. Conclusion

In this article, remarkable development, constituent cell materials, fabrication procedures of major types thin film solar cells have been reviewed. Modification of the layers in cell configuration from the primitive cell to the state-of-theart cell has been discussed with possible inclusion of new raw materials and fabrication process. Limitations for which performance of these cells are still below market-dominant silicon solar cell have also been discussed. The record efficiency, to date, for a-Si:H, CIGS, CZTS, CdTe and Dye sensitized solar cell (DSSC) is 13.4%, 22.5%, 12,6%, 22.1% and 15% respectively. Among them, CZTS and DSSC are

still in research and development phase while the efficiencies of CIGS and CdTe solar cell are comparable with the c-Si solar cells efficiency. However the manufacturing process of c-Si cell poses more challenges than thin film solar cells. So, thin film PV technologies are believe to be the most promising for terawatt scale PV deployment among the existing renewable energy technologies that could mitigate present as well as future energy crisis. Therefore, extensive research efforts must be given to overcome the critical issues related to the TFSCs for popularization as well as for viable commercialization.

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