**REVIEW ARTICLE** 

# Possible top cells for next-generation Si-based tandem solar cells

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**Abstract** Si-based solar cells, which have the advantages of high efficiency, low manufacturing costs, and outstanding stability, are dominant in the photovoltaic market. Currently, state-of-the-art Si-based solar cells are approaching the practical limit of efficiency. Constructing Si-based tandem solar cells is one available pathway to break the theoretical efficiency limit of single-junction silicon solar cells. Various top cells have been explored recently in the construction of Si-based tandem devices. Nevertheless, many challenges still stand in the way of extensive commercial application of Si-based tandem solar cells. Herein, we summarize the recent progress of representative Si-based tandem solar cells with different top cells, such as III-V solar cells, wide-bandgap perovskite solar cells, cadmium telluride (CdTe)-related solar cells, Cu(In,Ga)(Se,S)<sub>2</sub> (CIGS)-related solar cells, and amorphous silicon (a-Si) solar cells, and we analyze the main bottlenecks for their next steps of development. Subsequently, we suggest several potential candidate top cells for Si-based tandem devices, such as Sb<sub>2</sub>S<sub>3</sub>, Se, CdSe, and Cu<sub>2</sub>O. These materials have great potential for the development of high-performance and low-cost Si-based tandem solar cells in the future.

**Keywords** photovoltaic market, Si-based solar cell, efficiency limit, tandem, top cell

# 1 Introduction

As global environmental pollution and the energy crisis are increasing, renewable energy sources are playing a more important role in people's daily lives. Solar cells, which convert solar energy into electricity directly without extra

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pollution, have attracted extensive attention over the years. The photovoltaic market has developed rapidly with exponentially grown installed capacity. As shown in Fig. 1(a), the global cumulative installed capacity was only 173 MWp in 1996 but reached 392 GWp in 2017 with a > 40% compound annual growth rate [1]. Notably, in the past few years, crystalline Si (c-Si) technology, including monocrystalline Si (mono-Si) and multicrystalline Si (multi-Si), has dominated the photovoltaic market with > 90% of global annual production, and this has resulted in a gradual increasing market share [2]. In contrast, thin-film technologies, such as cadmium telluride (CdTe) and Cu(In,Ga)(Se,S)<sub>2</sub> (CIGS), possess only 5% of global annual production with 4.5 GWp of installed capacity in 2017 [2]. Until the fourth quarter of 2017, the estimated cumulative production of c-Si photovoltaic modules was ~405 GWp, which is much larger than  $\sim$ 33 GWp for thin-film photovoltaic modules (Fig. 1(b)) [2]. Additionally, owing to the optimization of the fabrication process, the module price of c-Si declined from ~4.5 EUR/Wp in 2006 to ~0.36 EUR/Wp in 2017, which was even lower than that of the thin-film photovoltaic module (~0.44 EUR/Wp) [2]. Remarkably, the photovoltaic module prices of both c-Si and thin-film technologies will drop off as their cumulative production continuously increase [2,6]. Apparently, in the near future, c-Si solar cells will still be the dominant portion of the photovoltaic market.

As shown in Fig. 1(c), as of now, the best mono-Si solar cell and module prepared in the laboratory have achieved 26.7% and 24.4% power conversion efficiencies (PCEs), respectively, which are very close to the practical efficiency limit of 29.4% for single-junction crystalline silicon solar cells [3,7,8]. Meanwhile, multi-Si solar cells reached a record efficiency of 23.2%, and its module efficiency exceeded 20% in 2019 [7]. However, in the past 20 years, the record efficiency of c-Si solar cells only increased by 3% [4]. Long progress plateaus have shown

the difficulties related to further improvement of the c-Si solar cells efficiency.

One feasible way to surpass the Shockley-Queisser efficiency limit is to develop tandem solar cells. By stacking several solar cells with different bandgaps, tandem solar cells can alleviate the energy losses that result from carrier thermalization [9]. Typically, in a twojunction tandem solar cell, there is a "top cell" with a large bandgap that mainly absorbs the short-wavelength photons and a "bottom cell" with a small bandgap to absorb longwavelength photons. This type of structure can facilitate the utilization of the solar spectrum and produce a higher efficiency compared with single-junction solar cells. As shown in Fig. 1(d), the theoretical efficiency limit of the four-terminal two-junction tandem solar cell can reach more than 46%, when the bandgaps of the bottom and top cells are 0.94 and 1.73 eV, respectively [10]. Si has a suitable bandgap of 1.12 eV; thus, Si-based tandem solar cells can achieve a theoretical 45% efficiency in conjunction with a top cell that has a bandgap of  $\sim 1.7$  eV [10]. Moreover, c-Si solar cells have the advantages of low manufacturing costs, high device performances, and outstanding stability, making them ideal bottom-cell candidates for two-junction tandem solar cells [11]. The large potential for developing high-efficiency and low-cost Si-based tandem solar cells has attracted many researchers. In fact, various materials, such as III-V semiconductors, wide-bandgap perovskites, and II-VI compounds, have been explored to construct Si-based tandem solar cells in the past few years [8,11].

This review presents the recent developments related to Si-based tandem solar cells and introduces some potential candidates for the top cell. First, we briefly review the past and current situation of the photovoltaic market and discuss the future developing direction of Si-based solar cells. Then, the developments related to typical Si-based tandem solar cells are summarized, and the main hindrances for their commercialization are emphasized. Finally, we highlight some potential candidates that are suitable for constructing a tandem device with c-Si solar cells.



**Fig. 1** (a) Evolution of global cumulative installed capacity and percentage of global annual production of three major photovoltaic technologies: thin-film, multi-Si, and mono-Si. Adapted from Refs. [1,2]. (b) Photovoltaic module price evolution versus cumulative production. Adapted from Ref. [2]. (c) Record efficiencies of c-Si solar cells and their theoretical efficiency limits [3–5]. (d) Theoretical efficiency limit of four-terminal tandem solar cells as a function of the bandgaps of top and bottom cells based on the Shockley–Queisser limit

# 2 Si-based tandem solar cells

Thus far, different types of Si-based tandem solar cells have been explored, but they are still far away from widespread commercial application because of the limit of the top cells. An efficient top cell is crucial for improving the PCE of the total system. Assuming a c-Si bottom cell with 25% efficiency and a top cell with an optimum bandgap of 1.7 eV, 13% efficiency is required for the top cell to compensate for the power lost from the c-Si bottom cell, while an 18% efficient top cell would yield a tandem efficiency of 30% [12]. Moreover, the manufacturing costs and stability of the top cell should also be taken into account. In this section, we discuss the most common ideas, which can be divided into groups based on the top cell material, and we discuss the main challenges for their future development.

#### 2.1 III-V semiconductors as top cell

III-V semiconductors represent an important class of highefficiency solar cells. Most have a direct bandgap with a large absorption coefficient, high mobility, and excellent anti-irradiation properties [11]. As a representative, GaAsbased single-junction solar cells have achieved 29.1% PCE, which is the highest value among all single-junction solar cells created thus far [7,13]. Despite their outstanding solar-to-electricity performance, III-V solar cells are not welcome in the terrestrial photovoltaic market because of the high manufacturing cost; thus, they are usually adopted for space applications or concentrated photovoltaic systems [11]. For tandem photovoltaic devices, sixjunction III-V tandem solar cells have also obtained a high efficiency of 39.2% under AM 1.5G illumination [14]. In the case of Si-based tandem solar cells, III-V semiconductors still hold the record efficiency of 32.8% for two-junction devices and 35.9% for three-junction devices [7,15]. Though researchers have selected a lowcost mechanical stacking solution to prepare tandem solar cells, the estimated fabrication costs were still much higher than commercial c-Si solar cells with the same production volumes [15]. Thus, high material and fabrication costs still hinder the commercial application of III-V/Si tandem solar cells.

#### 2.2 Wide-bandgap perovskites as top cell

Perovskites generally refer to a family of materials with a crystal structure of ABX<sub>3</sub>, which originates from CaTiO<sub>3</sub> [16]. Organic–inorganic hybrid perovskite solar cells developed rapidly in recent years and have attracted extensive attention. The highest efficiency of single-junction perovskite solar cells has reached 25.2% based on the (FAPbI<sub>3</sub>)<sub>0.95</sub>(MAPbBr<sub>3</sub>)<sub>0.05</sub> absorber, and it is close to the record efficiency of single-crystalline Si solar cells (26.7%) [4,7,17]. It has been reported that the bandgap of

perovskite could be tuned from 1.2 to 2.3 eV by controlling the composition, such as methylammonium (MA), cesium (Cs), and formamidinium (FA) at the A site; lead (Pb) and tin (Sn) at the B site; and iodine (I) and bromine (Br) at the X site [16,18]. With their advantages of a tunable bandgap, low-temperature fabrication process, strong light absorption, excellent device performance, and low manufacturing costs, wide-bandgap perovskites are widely expected to be suitable top cells for pairing with c-Si in tandem. We list several reported perovskite/Si tandems with a > 25%certified PCE in Table 1. Moreover, in 2020, the Helmholtz Center for Materials and Energy (HZB) declared that they obtained a certified PCE of 29.1% for perovskite/Si tandem solar cells; however, they have not disclosed their device structure or detailed parameters yet [4]. Similarly, the concrete structure of the 28%-efficient device is unknown as well [7]. Remnant high-efficiency perovskite/Si solar cells are mainly a two-terminal structure that have a monolithic design, which could benefit from the mild preparation process of perovskites. Within only 5 years of development, the device efficiency of perovskite/Si tandem solar cells has broken a number of world records, and seemingly, its upward trend is still far from saturation [4,32]. Notably, the stability of the perovskite top cell is one severe bottleneck for its commercial application, and the toxicity of its inclusive heavy metal could result in problems [33]. Although some investigations have suggested that the stability could be improved by introducing formamidinium (FA) or cesium (Cs) and optimizing the device interfaces, perovskites are still not eligible for practical application [18,33].

#### 2.3 CdTe and II-VI alloys as top cell

CdTe, which has an appropriate direct bandgap of 1.49 eV for single-junction solar cells, has achieved a world-record efficiency of 22.1%, which is close to multi-Si's record of 23.2% [4,34]. CdTe is the most successful commercial thin-film technology in the photovoltaic market, and impressively, the CdTe photovoltaic module has reached 19.0% efficiency [7,35]. Long-term commercial application indicates its operational stability and competitive cost advantage [7]. Although the bandgap of CdTe is not as compatible with Si-based tandem solar cells, it can be adjusted to an expanded range by alloying with Zn, Mg, and Se [36]. However, currently, only CdZnTe with its 1.78-eV bandgap has been integrated on c-Si and constructed as a two-terminal tandem solar cell with a 16.8% PCE (Table 1) [27]. A low device performance mainly resulted from the unoptimized tunnel junction and the low-efficiency c-Si bottom cell [27,28]. To date, singlecrystal CdZnTe and CdMgTe solar cells with a bandgap of ~1.7 eV have demonstrated 16.4% and 11.2% device efficiencies, respectively [27,28,37]. Nevertheless, polycrystalline CdZnTe and CdMgTe thin-film solar cells have shown much lower performance owing to alloy

double slash (//) means that subcells are	nal and two-terminal tandem structures,	
$\circ$ characterized under a AM 1.5G spectrum (1000 $W/m^2).$ The	nolithic growth. 4T and 2T are abbreviations for four-termi	
table results on Si-based tandem solar cells. All these solar cells wer	iereas a single slash (/) denotes that subcells are integrated by mo	at the data are not available
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Table 1         Summary of notable res	sults on Si-based	l tandem solar cells. Al	I these solar cells were	characterized under	a AM 1.5G s <sub>l</sub>	sectrum (1000 W/m <sup>2</sup> ). T	The double slash (//) means that subcells are
mechanically stacked, whereas a respectively. NA means that the d	single slash (/) ata are not avail	denotes that subcells lable	are integrated by mon-	olithic growth. 4T a	and 2T are ab	breviations for four-ter	minal and two-terminal tandem structures,
device structure	PCE/%	$V_{\rm OC}/V$	$J_{ m SC}/( m mA\cdot cm^{-2})$	FF/%	area/cm <sup>2</sup>	test center, date	note
GaAs//Si (1.42//1.12 eV)	32.82	1.092//0.683	28.90//11.07	85.0//79.2	1.003	NREL, 12/2016	NREL/CSEM/EPFL, 4T [15]
GaInP//Si (1.81//1.12 eV)	32.45	1.454//0.694	15.78//23.11	87.0//77.9	1.005	NREL, 12/2016	NREL/CSEM/EPFL, 4T [15]
GaAsP/Si (1.72/1.12 eV)	20.1	1.673	14.94	80.3	3.940	NREL, 05/2018	OSU/SolAero/UNSW, 2T [19]
AlGaAs/Si (1.6/1.12 eV)	25.2	1.55	27.9	58	1	not certified, 04/ 2012	UTokyo, 2T [20]
GaInP/GaAs//Si (1.81/1.42//1.12 eV)	35.91	2.520//0.681	13.61//11.03	87.5//78.5	1.002	NREL, 02/2017	NREL/CSEM/EPFL, 4T [15]
GaInP/AIGaAs/Si (1.90/1.43/1.12 eV)	34.1	3.177	12.4	86.4	3.987	FhG-ISE, 08/2019	FhG-ISE, 2T [7,21]
GaInP/GaAs/Si (1.90/1.43/1.12 eV)	24.3	2.662	12.2	74.5	3.987	FhG-ISE, 06/2019	FhG-ISE, 2T [7,22]
perovskite/Si	28.0	1.802	19.75	78.7	1.03	NREL, 12/18	Oxford PV, 2T [7]
perovskite/Si (1.67/1.12 eV, CsFAMAPbIBrCI)	27.13 (26.08)	1.886 (1.87)	19.12 (18.4)	75.3 (74.9)	1 (0.999)	not certified, 2019 (NREL, 08/2019)	CU/NREL/USTC, 2T [16]
perovskite/Si (1.68/1.12 eV, CsMAFAPbIBr)	25.71	1.781	19.07	75.36	0.832	FhG-ISE, 03/2020	UofT/KAUST/NREL/SDSU, 2T [23]
perovskite/Si (1.63/1.12 eV, CsFAMAPbIBr)	25.43	1.792	19.02	74.6	1.088	FhG-ISE, 02/2019	HZB/Oxf/Oxford PV, 2T [24]
perovskite/Si (1.6/1.12 eV, CsFAPbIBr)	25.24	1.788	19.53	73.1	1.419	FhG-ISE, 06/2018	EPFL/CSEM, 2T [25]
perovskite//Si (1.72//1.12 eV, CsFAPbIBr)	27.1	1.22//0.678	15.4//24.1	70.1//81.2	0.13	not certified, 12/ 2018	IMEC/KU Leuven/UG/TU/e, 4T [26]
CdZnTe/Si (1.78/1.12 eV)	16.8	1.75	16	60	NA	not certified, 01/ 2010	EPIR, 2T [27,28]
CuGaSe <sub>2</sub> /Si(1.7/1.12 eV)	5.1	1.32	9.0	43	NA	NREL, 03/2003	NREL, 2T [29]
CuGaSe <sub>2</sub> /Si(1.7/1.12 eV)	9.7	1.328	12.3	59.4	0.5	not certified, 11/ 2017	KIST/UST [30]
a-Si//Si(1.7//1.12 eV)	16.8	0.867//0.545	13.4//23.2	61.0//76.6	0.033	not certified, 05/ 1990	Osaka University, 4T [31]
a-Si/Si(1.7/1.12 eV)	15.04	1.478	16.17	63.0	0.065	not certified, 05/ 1990	Osaka University, 2T [31]

degradation during CdCl<sub>2</sub> passivation [38]. Developing a more practicable passivation method could result in highefficiency CdZnTe and CdMgTe thin-film solar cells, thus decreasing the manufacturing costs of Si-based tandem devices.

2.4 CIGS-related compounds as top cell

As another successfully commercialized thin-film technology, CIGS photovoltaic devices have been widely investigated in recent decades. As of now, the champion CIGS thin-film solar cell and module have achieved 23.35% and 19.2% efficiencies on Cd-free buffer layers, respectively [7,39]. The CIGS photovoltaic modules have exhibited decent stability, and it has been reported that the degradation rate of CIGS modules is slightly higher than CdTe and c-Si solar cells [40]. Similar to CdTe alloys, the bandgap of CIGS can be tuned from 1 to 2.5 eV by controlling the contents of In, Ga, and S [36]. However, the highest efficiency of CIGS/c-Si tandem solar cells achieved to date was only 9.7% using CuGaSe<sub>2</sub> as the top cell with its  $\sim 1.7$  eV bandgap [30]. The low device performance mainly resulted from the low efficiency of the CuGaSe<sub>2</sub> top cell [30]. Some researchers have explored perovskite/CIGS tandem devices and increased the device efficiency to 23.3%; other researchers have explore all-CIGS tandems, but these resulted in low device performances (<10%) [7.36,41]. Overall, a high-efficiency wide-bandgap CIGS top cell is necessary for developing CIGS/c-Si tandem solar cells. Whereas the record efficiency of CuGaSe<sub>2</sub> is 11.11%, a pure sulfide Cu(In, Ga)S<sub>2</sub> solar cell has reached 15.5%, but its bandgap is merely ~1.57 eV [42,43]. Another key problem to be solved is the high-temperature treatment during the CIGS preparation process, which could deteriorate the device performance of the c-Si bottom cell [44]. Otherwise, a four-terminal structure is more feasible to construct CIGS/ c-Si tandem solar cells.

#### 2.5 A-Si as top cell

Amorphous silicon (a-Si) is a theoretically promising top cell material owing to its suitable bandgap of ~1.7 eV when it is stacked with c-Si to made a tandem solar cell [45]. However, only a few studies have focused on a-Si/c-Si tandem devices [31,46,47]. To date, the highest efficiencies of a-Si/c-Si tandem solar cells have been 16.8% for a fourterminal device and 15.04% for a two-terminal device in 1990 [31]. No progress has been made over the next several decades. The main reason can be attributed to the low PCE of a-Si solar cells. Although a a-Si photovoltaic device has been developed since the 1970s, its record efficiency of 10.2% is still much lower than other mainstream solar cells, such as multi-Si, mono-Si, and CdTe solar cells [7,45]. The current device performance of the a-Si solar cell is lower than the requirement for the top cell of Si-based tandems to compensate for the efficiency loss [12]. Unfortunately, it has been reported that the practical limit of the a-Si solar cell is only 16% compared with its Shockley–Queisser limit of 25% because of the existence of band tail states [48–50]. Moreover, the a-Si solar cell still suffers from light-induced degradation, which further limits its application under long-term illumination in outdoor environments.

# **3** Potential candidates as top cell for Si-based tandems

Many challenges still stand in the way to commercializing Si-based tandem solar cells, although the search for more compatible materials has not stopped. Herein, we list several major preconditions for potential candidates, and a detailed screening strategy is illustrated in Fig. 2. First, the bandgap of the top cell must ensure a high theoretical efficiency. As shown in Fig. 1(d), considering mechanically stacked four-terminal two-junction Si-based tandems, a top cell with a bandgap ranging from 1.6 to 2.1 eV could yield a  $\geq 44\%$  efficiency. Second, the top cell must be stable enough to meet the application requirements of c-Si solar cells. As of now, the most recent photovoltaics, such as organic, dye-sensitized, and perovskite solar cells, still suffer from low stability in outdoor environments, which is the biggest obstacle for their commercial application [51,52]. In contrast, inorganic thin-film solar cells, such as CdTe and CIGS solar cells, have demonstrated a statistically approximate degradation rate compared with c-Si solar cells [40]. Indeed, most inorganic planar thinfilm solar cells have the advantage of a good stability, which provides the potential for developing Si-based tandems. Third, the manufacturing costs should be low enough to enable commercializing application. Earthabundant raw materials and low requirements for facilities are beneficial for reducing costs. Additionally, the state-ofthe-art PCE of the candidates also have a big influence on their development prospects. As summarized in Table 2,

screening of potential candidates



Fig. 2 Schematic of the screening process for potential candidates for the development of Si-based tandem solar cells

we propose several potential candidate solar cells that meet the aforementioned requirements. All are typical widebandgap inorganic thin-film solar cells with over 5% PCE.

Antimony sulfide  $(Sb_2S_3)$  is an emerging inorganic photovoltaic material with a one-dimensional crystal structure, and it has an indirect bandgap of ~1.7 eV [44,59]. Both Sb and S are earth-abundant elements, making  $Sb_2S_3$  free from the problem of supply shortages of raw materials [60]. Previously, Sb<sub>2</sub>S<sub>3</sub> was mainly developed for sensitized solar cells and has achieved a record PCE of 7.5% [53]. Recently, more studies have focused on planar thin-film Sb<sub>2</sub>S<sub>3</sub> solar cells. Several research groups have increased their PCEs to >6% within the last few years [61,62]. The large open-circuit voltage  $(V_{OC})$  deficit  $(\sim 1 \text{ V})$  is the main obstacle for obtaining high-efficiency  $Sb_2S_3$  solar cells [44]. Some simulations have implied that the high interfacial defect density and poor carrier transportation drastically limit the device performance of Sb<sub>2</sub>S<sub>3</sub>-based photovoltaics; thus, the present efficiency can be improved by controlling the orientation of Sb<sub>2</sub>S<sub>3</sub> films [44,63–66]. Moreover, in 2020, Zhang et al. demonstrated a proof-of-concept four-terminal tandem device with a 7.93% efficiency using Sb<sub>2</sub>Se<sub>3</sub> and Sb<sub>2</sub>S<sub>3</sub> solar cells as the bottom and top subcells [67]. This attempt implied that Sb<sub>2</sub>S<sub>3</sub> has the potential for application in tandem solar cells. Overall, the matched bandgap, earth-abundant raw material, and benign synthesis conditions give Sb<sub>2</sub>S<sub>3</sub> considerable potential to be used as the top cell of Si-based two-junction tandem solar cells.

Selenium (Se), the oldest photovoltaic absorber, has the advantages of a ~1.95 eV direct bandgap, low-cost, nontoxicity, and low synthesis temperature, and it has potential in the development of tandem solar cells [54,68]. In 2017, research demonstrated a record efficiency of 6.5% on ultrathin Se solar cells and pathways toward a higher device performance [54]. It was proposed that reducing the cliff at the interface by optimizing the buffer layer, increasing the carrier lifetime of the Se film, and alleviating optical loss could further increase the device performance [54]. Additionally, it was reported that the bandgap of Se could be tuned to a small value by alloying with tellurium (Te), thus provided a better match with the c-Si bottom cell [69]. Therefore, Se solar cells have exhibited compatibility with most bottom cells in tandem devices but still require further improvements in terms of device performance.

Cadmium selenide (CdSe) has an appropriate bandgap of  $\sim$ 1.7 eV and a high absorption coefficient in the visible

light region [70]. In 1982, Rickus achieved over 6% efficiency for CdSe thin-film solar cells using a vacuum evaporation method [55]. However, over the next several decades, only a few studies concentrated on CdSe thin-film solar cells, and no efficiency improvement has been reported [71–73]. Otherwise, owing to its excellent semiconductor properties, CdSe has been widely developed in other fields, such as in photoelectrochemical solar cell sand colloidal quantum dots [74,75]. The device performance of CdSe thin-film solar cells could be enhanced to a higher degree upon further optimization. Notably, the toxicity of the heavy metal, Cd, could restrict the application of CdSe solar cells in the future because of the potential environmental impact.

Cuprous oxide (Cu<sub>2</sub>O) is one of the oldest discovered semiconductor materials, and it has a wide bandgap of  $\sim 2.1$ eV [36]. Preventing the formation of a Cu or CuO phase when preparing Cu<sub>2</sub>O solar cells is essential to obtaining pure-phase Cu<sub>2</sub>O [76]. High-quality Cu<sub>2</sub>O can be prepared by thermally oxidizing Cu sheets, and based on this method, Minami et al. obtained a 8.1% PCE in Cu<sub>2</sub>Obased heterojunction solar cells in 2016, which is impressive in contrast to its ~20% Shockley-Queisser limit efficiency [49,50,56]. Further efficiency improvement relies on applying a surface treatment to decrease leakage current and constructing an n-i-p device [56]. Additionally, a low-temperature process of atomic layer deposition was also developed to fabricate Cu<sub>2</sub>O solar cells, and this achieved a certified 3.97% PCE with a remarkable 1.2 V V<sub>OC</sub> [57,77–79]. The low PCE mainly resulted from the low short-circuit current density  $(J_{SC})$ and fill factor (FF) caused by inefficient photo-generated carrier collection [77]. Overall, Cu<sub>2</sub>O still has the potential for use in the development of monolithic integrated Sibased tandem solar cells.

## 4 Conclusions

As the dominant part of the current photovoltaic market, c-Si solar cells are approaching their practical limit. Constructing tandem solar cells with a wide-bandgap material as a top cell is a feasible way to exceed this practical limit. Although various types of top cells have been investigated, there is still a long way toward commercial application. III-V/Si tandem solar cells exhibit notable high efficiency and stability but have drawbacks of

Table 2 List of potential candidates for use in Si-based tandem solar cells

material	$E_{\rm g}/{\rm eV}$	PCE/%	$V_{\rm OC}/{ m V}$	$J_{\rm SC}/({\rm mA}\cdot{\rm cm}^{-2})$	FF/%	description
Sb <sub>2</sub> S <sub>3</sub>	1.7	7.5	0.711	16.1	65.0	chemical bath deposition [53]
Se	1.95	6.51	0.969	10.6	63.4	thermal evaporation [54]
CdSe	1.72	~6	0.65	18	50	vacuum evaporation [55]
Cu <sub>2</sub> O	2.1	8.1	1.2	10.4	65	Cu sheet oxidization [56-58]

a high manufacturing cost. Perovskites/Si tandems developed very rapidly and achieved approximately 30% efficiency, but perovskites still suffer from their low stability in outdoor environments. CdZnTe/Si, CuGaSe<sub>2</sub>/Si, and a-Si/Si tandem solar cells have not exceeded the record efficiency of single-junction mono-Si solar cells thus far, and the low performance of the top cell compared with III-V and wide-bandgap perovskites is the key problem that needs to be resolved. Therefore, we propose four potential candidates for developing Si-based tandem solar cells including Sb<sub>2</sub>S<sub>3</sub>, Se, CdSe, and Cu<sub>2</sub>O. They have an appropriate bandgap to enable a high theoretical efficiency, decent stability to meet the requirements of the Si-based bottom cell, earth-abundant raw materials to reduce costs, and >5% state-of-the-art device performance to ensure development prospects. Notably, the most important goal is to increase the device performance to a new level, which can make it more applicable for configuring tandem devices with Si-based solar cells.

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### References

- 1. BP p.l.c. BP Statistical Review of World Energy. UK. 2019, 51
- Fraunhofer Institute for Solar Energy Systems. Photovolatics Report. Germany. 2019, 21–45
- Richter A, Hermle M, Glunz S W. Reassessment of the limiting efficiency for crystalline silicon solar cells. IEEE Journal of Photovoltaics, 2013, 3(4): 1184–1191
- 4. NREL. Best Research-Cell Efficiencies. 2020
- Polman A, Knight M, Garnett E C, Ehrler B, Sinke W C. Photovoltaic materials: present efficiencies and future challenges. Science, 2016, 352(6283): aad4424
- Reese M O, Glynn S, Kempe M D, McGott D L, Dabney M S, Barnes T M, Booth S, Feldman D, Haegel N M. Increasing markets and decreasing package weight for high-specific-power photovoltaics. Nature Energy, 2018, 3(11): 1002–1012
- Green M A, Dunlop E D, Hohl-Ebinger J, Yoshita M, Kopidakis N, Ho-Baillie A W. Solar cell efficiency tables (Version 55). Progress in Photovoltaics: Research and Applications, 2020, 28(1): 3–15
- Yu Z S, Leilaeioun M, Holman Z. Selecting tandem partners for silicon solar cells. Nature Energy, 2016, 1(11): 16137
- Leijtens T, Bush K A, Prasanna R, McGehee M D. Opportunities and challenges for tandem solar cells using metal halide perovskite semiconductors. Nature Energy, 2018, 3(10): 828–838
- 10. Bremner S, Levy M, Honsberg C B. Analysis of tandem solar cell efficiencies under AM1.5G spectrum using a rapid flux calculation

method. Progress in Photovoltaics: Research and Applications, 2008, 16(3): 225-233

- Yamaguchi M, Lee K H, Araki K, Kojima N. A review of recent progress in heterogeneous silicon tandem solar cells. Journal of Physics D, Applied Physics, 2018, 51(13): 133002
- White T P, Lal N N, Catchpole K R. Tandem solar cells based on high-efficiency c-Si bottom cells: top cell requirements for > 30% efficiency. IEEE Journal of Photovoltaics, 2014, 4(1): 208–214
- Kayes B M, Nie H, Twist R, Spruytte S G, Reinhardt F, Kizilyalli I C, Higashi G S. 27.6% conversion efficiency, a new record for single-junction solar cells under 1 sun illumination. In: Proceedings of the 37th IEEE Photovoltaic Specialists Conference. Seattle: IEEE, 2011, 000004–000008
- Geisz J F, Steiner M A, Jain N, Schulte K L, France R M, McMahon W E, Perl E E, Friedman D J. Building a six-junction inverted metamorphic concentrator solar cell. IEEE Journal of Photovoltaics, 2018, 8(2): 626–632
- 15. Essig S, Allebé C, Remo T, Geisz J F, Steiner M A, Horowitz K, Barraud L, Ward J S, Schnabel M, Descoeudres A, Young D L, Woodhouse M, Despeisse M, Ballif C, Tamboli A. Raising the onesun conversion efficiency of III–V/Si solar cells to 32.8% for two junctions and 35.9% for three junctions. Nature Energy, 2017, 2(9): 17144
- 16. Xu J, Boyd C C, Yu Z J, Palmstrom A F, Witter D J, Larson B W, France R M, Werner J, Harvey S P, Wolf E J, Weigand W, Manzoor S, van Hest M F A M, Berry J J, Luther J M, Holman Z C, McGehee M D. Triple-halide wide-band gap perovskites with suppressed phase segregation for efficient tandems. Science, 2020, 367(6482): 1097–1104
- Jung E H, Jeon N J, Park E Y, Moon C S, Shin T J, Yang T Y, Noh J H, Seo J. Efficient, stable and scalable perovskite solar cells using poly(3-hexylthiophene). Nature, 2019, 567(7749): 511–515
- Lal N N, Dkhissi Y, Li W, Hou Q, Cheng Y B, Bach U. Perovskite tandem solar cells. Advanced Energy Materials, 2017, 7(18): 1602761
- Grassman T J, Chmielewski D J, Carnevale S D, Carlin J A, Ringel S A. GaAs<sub>0.75</sub>P<sub>0.25</sub>/Si dual-junction solar cells grown by MBE and MOCVD. IEEE Journal of Photovoltaics, 2016, 6(1): 326–331
- Tanabe K, Watanabe K, Arakawa Y. III-V/Si hybrid photonic devices by direct fusion bonding. Scientific Reports, 2012, 2(1): 349
- Cariou R, Benick J, Feldmann F, Höhn O, Hauser H, Beutel P, Razek N, Wimplinger M, Bläsi B, Lackner D, Hermle M, Siefer G, Glunz S W, Bett A W, Dimroth F. III–V-on-silicon solar cells reaching 33% photoconversion efficiency in two-terminal configuration. Nature Energy, 2018, 3(4): 326–333
- Feifel M, Lackner D, Ohlmann J, Benick J, Hermle M, Dimroth F. Direct growth of a GaInP/GaAs/Si triple-junction solar cell with 22.3% AM1.5G efficiency. Solar RRL, 2019, 3(12): 1900313
- 23. Hou Y, Aydin E, De Bastiani M, Xiao C, Isikgor F H, Xue D J, Chen B, Chen H, Bahrami B, Chowdhury A H, Johnston A, Baek S W, Huang Z, Wei M, Dong Y, Troughton J, Jalmood R, Mirabelli A J, Allen T G, Van Kerschaver E, Saidaminov M I, Baran D, Qiao Q, Zhu K, De Wolf S, Sargent E H. Efficient tandem solar cells with solution-processed perovskite on textured crystalline silicon. Science, 2020, 367(6482): 1135–1140

- 24. Mazzarella L, Lin Y H, Kirner S, Morales-Vilches A B, Korte L, Albrecht S, Crossland E, Stannowski B, Case C, Snaith H J, Schlatmann R. Infrared light management using a nanocrystalline silicon oxide interlayer in monolithic perovskite/silicon heterojunction tandem solar cells with efficiency above 25%. Advanced Energy Materials, 2019, 9(14): 1803241
- 25. Sahli F, Werner J, Kamino B A, Bräuninger M, Monnard R, Paviet-Salomon B, Barraud L, Ding L, Diaz Leon J J, Sacchetto D, Cattaneo G, Despeisse M, Boccard M, Nicolay S, Jeangros Q, Niesen B, Ballif C. Fully textured monolithic perovskite/silicon tandem solar cells with 25.2% power conversion efficiency. Nature Materials, 2018, 17(9): 820–826
- Jaysankar M, Raul B A, Bastos J, Burgess C, Weijtens C, Creatore M, Aernouts T, Kuang Y, Gehlhaar R, Hadipour A, Poortmans J. Minimizing voltage loss in wide-bandgap perovskites for tandem solar cells. ACS Energy Letters, 2019, 4(1): 259–264
- Carmody M, Mallick S, Margetis J, Kodama R, Biegala T, Xu D, Bechmann P, Garland J, Sivananthan S. Single-crystal II–VI on Si single-junction and tandem solar cells. Applied Physics Letters, 2010, 96(15): 153502
- Sivananthan S, Garland J W, Carmody M W. Multijunction singlecrystal CdTe-based solar cells: opportunities and challenges. In: Proceedings of Energy Harvesting and Storage: Materials, Devices, and Applications. Orlando: SPIE, 2010, 76830N
- 29. Noufi R, Young D L, Coutts T J, Gessert T, Ward J S, Duda A, Wu X, Romero M, Dhere R, Shama J A. Toward a 25%-efficient polycrystalline thin-film tandem solar cell: practical issues. In: Proceedings of the 3rd World Conference on Photovoltaic Energy Conversion. Osaka: IEEE, 2003, 12–14
- Jeong A R, Choi S B, Kim W M, Park J K, Choi J, Kim I, Jeong J H. Electrical analysis of c-Si/CGSe monolithic tandem solar cells by using a cell-selective light absorption scheme. Scientific Reports, 2017, 7(1): 15723
- Matsumoto Y, Miyagi K, Takakura H, Okamoto H, Hamakawa Y. a-Si/poly-Si two-and four-terminal tandem type solar cells. In: Proceedings of IEEE Conference on Photovoltaic Specialists. Kissimmee: IEEE, 1990, 1420–1425
- 32. Shen H, Walter D, Wu Y, Fong K C, Jacobs D A, Duong T, Peng J, Weber K, White T P, Catchpole K R. Monolithic perovskite/Si tandem solar cells: pathways to over 30% efficiency. Advanced Energy Materials, 2020, 10(13): 1902840
- Todorov T, Gunawan O, Guha S. A road towards 25% efficiency and beyond: perovskite tandem solar cells. Molecular Systems Design & Engineering, 2016, 1(4): 370–376
- Scheer R, Schock H W. Chalcogenide Photovoltaics: Physics, Technologies, and Thin Film Devices. Weinheim: John Wiley & Sons. 2011, 178
- Bosio A, Rosa G, Romeo N. Past, present and future of the thin film CdTe/CdS solar cells. Solar Energy, 2018, 175: 31–43
- Todorov T K, Bishop D M, Lee Y S. Materials perspectives for nextgeneration low-cost tandem solar cells. Solar Energy Materials and Solar Cells, 2018, 180: 350–357
- Becker J J, Campbell C M, Tsai C, Zhao Y, Lassise M, Zhao X, Boccard M, Holman Z C, Zhang Y. Monocrystalline 1.7-eVbandgap MgCdTe solar cell with 11.2% efficiency. IEEE Journal of

Photovoltaics, 2018, 8(2): 581-586

- 38. Swanson D E, Reich C, Abbas A, Shimpi T, Liu H, Ponce F A, Walls J M, Zhang Y H, Metzger W K, Sampath W S, Holman Z C. CdCl<sub>2</sub> passivation of polycrystalline CdMgTe and CdZnTe absorbers for tandem photovoltaic cells. Journal of Applied Physics, 2018, 123(20): 203101
- Nakamura M, Yamaguchi K, Kimoto Y, Yasaki Y, Kato T, Sugimoto H. Cd-Free Cu(In,Ga)(Se,S)<sub>2</sub> thin-film solar cell with record efficiency of 23.35%. IEEE Journal of Photovoltaics, 2019, 9 (6): 1863–1867
- Jordan D C, Kurtz S R. Photovoltaic degradation rates-an analytical review. Progress in Photovoltaics: Research and Applications, 2013, 21(1): 12–29
- Yi J. New generation multijunction solar cells for achieving high efficiencies. Current Photovoltaic Research, 2018, 6(2): 31–38
- 42. Ishizuka S, Yamada A, Fons P J, Shibata H, Niki S. Impact of a binary Ga<sub>2</sub>Se<sub>3</sub> precursor on ternary CuGaSe<sub>2</sub> thin-film and solar cell device properties. Applied Physics Letters, 2013, 103(14): 143903
- 43. Hiroi H, Iwata Y, Adachi S, Sugimoto H, Yamada A. New worldrecord efficiency for pure-sulfide Cu(In,Ga)S<sub>2</sub> thin-film solar cell with Cd-free buffer layer via KCN-free process. IEEE Journal of Photovoltaics, 2016, 6(3): 760–763
- Kondrotas R, Chen C, Tang J. Sb<sub>2</sub>S<sub>3</sub> solar cells. Joule, 2018, 2(5): 857–878
- Sai H, Matsui T, Kumagai H, Matsubara K. Thin-film microcrystalline silicon solar cells: 11.9% efficiency and beyond. Applied Physics Express, 2018, 11(2): 022301
- Navaraj W T, Yadav B K, Kumar A. Optoelectronic simulation and optimization of unconstrained four terminal amorphous silicon/ crystalline silicon tandem solar cell. Journal of Computational Electronics, 2016, 15(1): 287–294
- 47. Matsui T, Maejima K, Bidiville A, Sai H, Koida T, Suezaki T, Matsumoto M, Saito K, Yoshida I, Kondo M. High-efficiency thinfilm silicon solar cells realized by integrating stable a-Si:H absorbers into improved device design. Japanese Journal of Applied Physics, 2015, 54(8S1): 08KB10
- Tiedje T. Band tail recombination limit to the output voltage of amorphous silicon solar cells. Applied Physics Letters, 1982, 40(7): 627–629
- Rühle S. Tabulated values of the Shockley–Queisser limit for single junction solar cells. Solar Energy, 2016, 130: 139–147
- Shockley W, Queisser H. Detailed balance limit of efficiency of p-n junction solar cells. Journal of Applied Physics, 1961, 32(3): 510– 519
- Castro-Hermosa S, Yadav S K, Vesce L, Guidobaldi A, Reale A, Di Carlo A, Brown T M. Stability issues pertaining large area perovskite and dye-sensitized solar cells and modules. Journal of Physics D, Applied Physics, 2017, 50(3): 033001
- 52. Owens C, Ferguson G M, Hermenau M, Voroshazi E, Galagan Y, Zimmermann B, Rösch R, Angmo D, Teran-Escobar G, Uhrich C, Andriessen R, Hoppe H, Würfel U, Lira-Cantu M, Krebs F C, Tanenbaum D M. Comparative indoor and outdoor degradation of organic photovoltaic cells via inter-laboratory collaboration. Polymers, 2015, 8(1): 1
- 53. Choi Y C, Lee D U, Noh J H, Kim E K, Seok S I. Highly improved

 $Sb_2S_3 \ sensitized-inorganic-organic \ heterojunction \ solar \ cells \ and \ quantification \ of \ traps \ by \ deep-level \ transient \ spectroscopy.$  Advanced Functional Materials, 2014, 24(23): 3587–3592

- 54. Todorov T K, Singh S, Bishop D M, Gunawan O, Lee Y S, Gershon T S, Brew K W, Antunez P D, Haight R. Ultrathin high band gap solar cells with improved efficiencies from the world's oldest photovoltaic material. Nature Communications, 2017, 8(1): 682
- Rickus E. Photovoltaic behaviour of CdSe thin film solar cells. In: Proceedings of the 4th EC Photovoltaic Solar Energy Conference. Dordrecht: Springer, 1982, 831–835
- 56. Minami T, Nishi Y, Miyata T. Efficiency enhancement using a  $Zn_{1-x}Ge_x$ -O thin film as an n-type window layer in Cu<sub>2</sub>O-based heterojunction solar cells. Applied Physics Express, 2016, 9(5): 052301
- Wong L H, Zakutayev A, Major J D, Hao X, Walsh A, Todorov T K, Saucedo E. Emerging inorganic solar cell efficiency tables (Version 1). Journal of Physics: Energy, 2019, 1(3): 032001
- Minami T, Nishi Y, Miyata T. High-efficiency Cu<sub>2</sub>O-based heterojunction solar cells fabricated using a Ga<sub>2</sub>O<sub>3</sub> thin film as ntype layer. Applied Physics Express, 2013, 6(4): 044101
- Lei H, Chen J, Tan Z, Fang G. Review of recent progress in antimony chalcogenide-based solar cells: materials and devices. Solar RRL, 2019, 3(6): 1900026
- Mavlonov A, Razykov T, Raziq F, Gan J, Chantana J, Kawano Y, Nishimura T, Wei H, Zakutayev A, Minemoto T, Zu X, Li S, Qiao L. A review of Sb<sub>2</sub>Se<sub>3</sub> photovoltaic absorber materials and thin-film solar cells. Solar Energy, 2020, 201: 227–246
- 61. Han J, Wang S, Yang J, Guo S, Cao Q, Tang H, Pu X, Gao B, Li X. Solution-processed Sb<sub>2</sub>S<sub>3</sub> planar thin film solar cells with a conversion efficiency of 6.9% at an open circuit voltage of 0.7 V achieved via surface passivation by a SbCl<sub>3</sub> interface layer. ACS Applied Materials & Interfaces, 2020, 12(4): 4970–4979
- Jiang C H, Tang R F, Wang X M, Ju H X, Chen G L, Chen T. Alkali metals doping for high-performance planar heterojunction Sb<sub>2</sub>S<sub>3</sub> solar cells. Solar RRL, 2019, 3(1): 1800272
- Wang L, Li D B, Li K, Chen C, Deng H X, Gao L, Zhao Y, Jiang F, Li L, Huang F, He Y, Song H, Niu G, Tang J. Stable 6%-efficient Sb<sub>2</sub>Se<sub>3</sub> solar cells with a ZnO buffer layer. Nature Energy, 2017, 2 (4): 17046
- 64. Zhou Y, Wang L, Chen S, Qin S, Liu X, Chen J, Xue D J, Luo M, Cao Y, Cheng Y, Sargent E H, Tang J. Thin-film Sb<sub>2</sub>Se<sub>3</sub> photovoltaics with oriented one-dimensional ribbons and benign grain boundaries. Nature Photonics, 2015, 9(6): 409–415
- 65. Deng H, Zeng Y, Ishaq M, Yuan S, Zhang H, Yang X, Hou M, Farooq U, Huang J, Sun K, Webster R, Wu H, Chen Z, Yi F, Song H, Hao X, Tang J. Quasiepitaxy strategy for efficient full-inorganic Sb<sub>2</sub>S<sub>3</sub> solar cells. Advanced Functional Materials, 2019, 29(31): 1901720
- 66. Cao Y, Zhu X, Jiang J, Liu C, Zhou J, Ni J, Zhang J, Pang J. Rotational design of charge carrier transport layers for optimal antimony trisulfide solar cells and its integration in tandem devices. Solar Energy Materials and Solar Cells, 2020, 206: 110279
- Zhang J, Lian W, Yin Y, Wang X, Tang R, Qian C, Hao X, Zhu C, Chen T. All antimony chalcogenide tandem solar cell. Solar RRL, 2020, 4(4): 2000048

- Zhu M, Niu G, Tang J. Elemental Se: fundamentals and its optoelectronic applications. Journal of Materials Chemistry C, Materials for Optical and Electronic Devices, 2019, 7(8): 2199– 2206
- Hadar I, Hu X, Luo Z Z, Dravid V P, Kanatzidis M G. Nonlinear band gap tunability in selenium tellurium alloys and its utilization in solar cells. ACS Energy Letters, 2019, 4(9): 2137–2143
- Bagheri B, Kottokkaran R, Poly L P, Sharikadze S, Reichert B, Noack M, Dalal V. Efficient heterojunction thin film CdSe solar cells deposited using thermal evaporation. In: Proceedings of the 46th IEEE Photovoltaic Specialists Conference (PVSC). Chicago: IEEE, 2019, 1822–1825
- Mahawela P, Jeedigunta S, Vakkalanka S, Ferekides C S, Morel D L. Transparent high-performance CDSE thin-film solar cells. Thin Solid Films, 2005, 480–481: 466–470
- 72. Patel N G, Panchal C J, Makhija K K, Patel P G, Patel S S. Fabrication and characterization of ZnTe/CdSe thin film solar cells. Crystal Research and Technology, 1994, 29(2): 247–252
- Shenouda A Y, El Sayed E S M. Electrodeposition, characterization and photo electrochemical properties of CdSe and CdTe. Ain Shams Engineering Journal, 2015, 6(1): 341–346
- 74. Murray C B, Norris D J, Bawendi M G. Synthesis and characterization of nearly monodisperse CdE (E = sulfur, selenium, tellurium) semiconductor nanocrystallites. Journal of the American Chemical Society, 1993, 115(19): 8706–8715
- 75. Frese K W Jr. A high-efficiency single-crystal cdse photoelectrochemical solar-cell and an associated loss mechanism. Applied Physics Letters, 1982, 40(3): 275–277
- 76. Giraldo S, Jehl Z, Placidi M, Izquierdo-Roca V, Pérez-Rodríguez A, Saucedo E. Progress and perspectives of thin film kesterite photovoltaic technology: a critical review. Advanced Materials, 2019, 31(16): 1806692
- 77. Lee Y S, Chua D, Brandt R E, Siah S C, Li J V, Mailoa J P, Lee S W, Gordon R G, Buonassisi T. Atomic layer deposited gallium oxide buffer layer enables 1.2 V open-circuit voltage in cuprous oxide solar cells. Advanced Materials, 2014, 26(27): 4704–4710
- Eisermann S, Kronenberger A, Laufer A, Bieber J, Haas G, Lautenschläger S, Homm G, Klar P J, Meyer B K. Copper oxide thin films by chemical vapor deposition: Synthesis, characterization and electrical properties. Physica Status Solidi (a), 2012, 209(3): 531– 536
- Lee Y S, Heo J, Siah S C, Mailoa J P, Brandt R E, Kim S B, Gordon R G, Buonassisi T. Ultrathin amorphous zinc-tin-oxide buffer layer for enhancing heterojunction interface quality in metal-oxide solar cells. Energy & Environmental Science, 2013, 6(7): 2112–2118



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