

Two-Terminal Perovskite Tandem Solar Cells: from Design to Commercial Prospect

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Abstract: Tandem Solar Cells (TSCs) with multi-junction are capable to break the SQ limit and achieve high PCE through absorbing larger range of light wavelength by multiple absorber layers with different band gaps. Perovskite solar cells are ideal light absorbing materials for TSC because of its high PCE, high suitability with other absorbers, low cost and easy fabrication. Perovskite-based TSCs have so far outperformed single-junction devices in PCE, garnering considerable interest from both academia and material industry. In this review, the basic science of perovskite Tandem Solar Cells (PTSCs) is presented, as well as the construction and properties of PSC as a top cell. Then three main types of PTSCs are introduced: Perovskite/Si, Perovskite/CIGS, and Perovskite/Perovskite including their design, challenges and fabrication methods. Finally, the current status and future prospects for commercialization of PTSCs are also discussed. According to recent developments, PTSCs are considered to be one of the most promising solar cells. Research on PTSCs could contribute to the development of desirable clean energy sources in order to solve the energy crisis and environmental problems of human beings.

Keywords: Perovskite, Solar Cells, Photovoltaics, Multi-Junction, Power Conversion Efficiency.

1. Introduction

Environmental problems caused by the use of fossil fuels are increasingly serious. The carbon, sulfur and nitrogen produced by burning fossil fuels can lead to harmful climate change and environmental pollution. Under such circumstances, how to obtain clean energy has become an urgent problem for mankind now. Among the typical renewable energy sources such as solar, wind, and hydro, the solar energy is believed to be the most likely to replace the fossil fuels in the future, primarily because the sun is constantly providing more energy than needed for humans. Over the past decade or so, the number of solar cell installations has increased significantly due to technological advances and cost reductions. With a 26.6% energy conversion efficiency, Si-based solar cells have taken the top spot in the PV industry [1]. Since the 29.4% theoretical limit is currently close, though, the room for improvement is very little.

Solar cells, as semiconductor devices, are capable of absorbing the photons carrying energy exceeds their band gaps. However, when the electrons in the excited state lose energy and fall to the bottom of the conduction band, the lost energy is released as thermal energy, which not only loses energy but also damages the device as the system heats up. Moreover, photons with sub-band gap energy can also be absorbed, generating undesired heat energy. The major issue single-junction solar cells are now experiencing is striking a balance point between increasing photon absorption and minimizing heat loss. Due to the tradeoff between heat loss and increased photon absorption, the maximum possible power of a solar cell with only one p-n junction will be lower than the Shockley-Queisser (S-Q) limit. Recently, scientists have proposed a number of physical concepts in order to break through this limit, one of which is the Tandem Solar Cell (TSC). Two sub-cells in a tandem cell complement the absorption range of the spectrum, absorb more photons as a whole, and minimize heat loss. Among the different types of TSCs, the most promising is the perovskite-based tandem solar cells (PTSC).

Metal-halide perovskite is a kind of materials with structure ABX_3 usually used for solar cells (Figure 1). In tandem cells, both the top and bottom subcells can be made of perovskite. In the structure, the monovalent cations within A sites is frequently selected from: $CH_3NH_3^+$, FA^+ :

$\text{HC}(\text{NH}_2)_2^+$, Cs^+ , Rb^+ ; the ions within B sites is commonly the three divalent metals: Pb^{2+} , Ge^{2+} , Sn^{2+} or the mixture of them; the monovalent anions within X sites is typically selected from the three halide elements: Cl^- , I^- , Br^- , or the mixtures of them [2].

Perovskite is a material with high structure tolerance, which is defined by The Goldschmidt tolerance factor (t) as below [3]:

$$t = \frac{r_A + r_B}{\sqrt{2}(r_B + r_X)} \quad (1)$$

According to research and experience, the tolerance factor of metal halide perovskite is between range of $0.81 \leq t \leq 1.00$. which allows for a large tunability on lattice constant of the perovskite for fitting with other layers under the circumstance of keeping stability of perovskite [4]. This characteristic allows the perovskite to easily achieve the strict lattice matching with the other subcells. In addition to the tunable lattice constant, the perovskite materials also have tunable band gaps from compositional engineering [5]. At the same time, the PSCs can be fabricated by low-temperature methods to maintain stability of other layers. Because of such characteristics, the Perovskite can be used as outstanding materials for tandem devices. In this decade, many researchers threw themselves into development of PSC and PSC related tandem device due to its potential industrial and commercial worth.

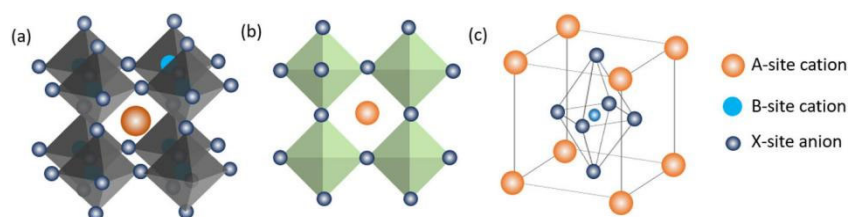


Figure 1. (a) 3D. (b) 2D. (c) Chemical.[6]

There are several materials that can be applied for bottom cell integrated with perovskite top cell, such as Si, CIGS and perovskite. In this review, Perovskite/Si, Perovskite/CIGS, Perovskite/Perovskite TSC with 2T configuration will be introduced. Basic information, the challenges and the solutions of these 2T PTSCs made by such materials will be summarizes.

2. Working mechanism

The TSCs are formed by the combination of two single-junction solar cells (Figure 2), the type integrated solar cells mechanically is 4-terminal (4T), while the type integrated solar cells monolithically is 2-terminal (2T). Compared to 4T, 2T TSC required less function layers, inverters and cablings. Correspondingly, the processing steps of 2T TSC is also less than 4T, as well as costs of fabrication and materials. Additionally, due to low temperature fabrication of PSCs, the stability of other layers is relatively higher as well as overall/system stability. Consequently, 2T TSC is more appealing due to these outstanding features in both academics and industry.

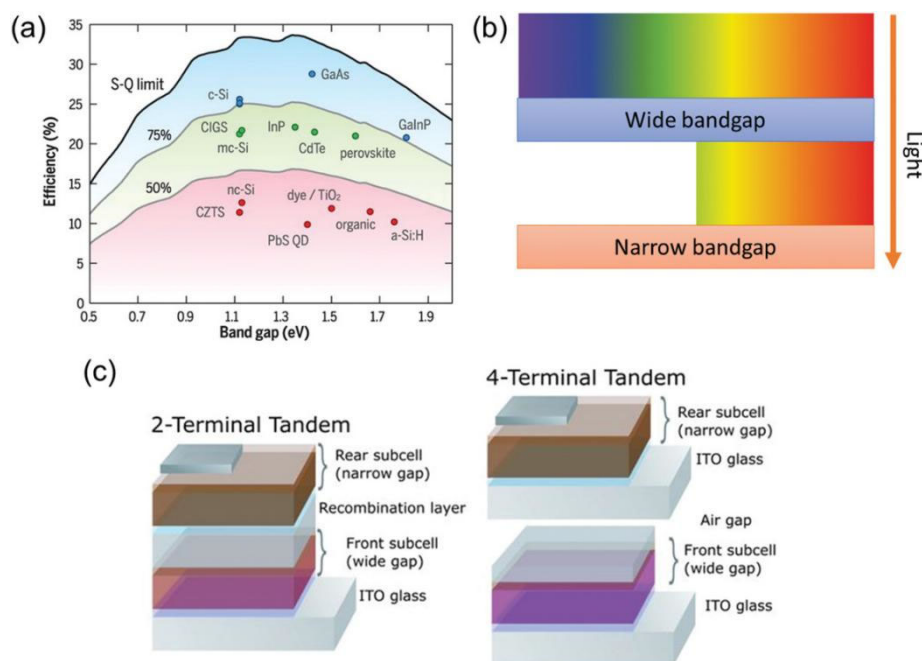


Figure 2. a) the Shockly-Queisser limit and record of PCE for materials b) schematic diagram of absorption process in a tandem device. c) a 2T and 4T tandem structure schematic.[7]

A 2-terminal TSC is a monolithically integrated multi-junction solar cell designed by stacking one single-junction cell on another single-junction cell or series connected the two cells. The common structure of 2T TSC is: TCO/ Top Cell/ RL or TJ/ Bottom Cell [8]. In the next sections, this review will present several layers in TSC in order of importance which is top cell, bottom cell and RL/ TJ.

Based on the quantum theory of semiconductors, the band gap of solar cells is directly related to the range of sunlight energy that can be absorbed. The high energy range of the sunlight will be absorbed by the top subcell with wider band gap, while the range of lower energy absorbed by the bottom subcell with narrower band gap [9]. Therefore, the whole device can absorb a larger range of photon energy to convert as electricity.

To integrate the top and bottom subcells into a monolithic tandem, the Recombination Layer (RL) is located between them. Meanwhile, the two subcell is connected in series via RL. As a result, a well-functioning RL is crucial to achieve current matching of the two subcells and high open-circuit voltage (V_{OC}). Besides the function of current matching, a RL is the position of recombination of the carriers, which determines carriers recombination efficiency [10].

A part of photons are absorbed by penetrating the glass and HTL of the front subcell, and then the rest part of the light is absorbed by the rear cell. Both two cells generate electrons and holes which are separated as free carriers. Electrons will be collected by ETL and transported to rear electrode or recombination layer (RL) through the conduction band of ETL, while the holes will be transported through valence band of HTL to front electrode or RL, depending on the structures of the whole device. RL is series connected between the two subcells. After the carrier pairs separate as free carriers, RL collect one kind of carriers (electrons or holes) from the rear of the top cell and collect the other kind of carriers (holes or electrons) from the front of bottom subcell. For example, in device with two p-i-n junction cells, the electrons move towards RL from the top cell and holes move towards RL from bottom cell [10]. Such movement of the carriers generate the electric current inside the device. Then the different kind of carriers recombine in RL to generate overall charge neutrality. As a result, RL should be with good selectivity of carriers and high transparency of near infrared (NIR) for allowing the NIR to pass through and be absorbed by bottom cells.

3. Top cell: perovskite

Metal-halide perovskite top cells are single-junction film devices with wide band gaps, high absorption efficiency, which can be easily integrated with other Photovoltaic (PV) materials. Generally, the perovskite absorber thickness is ~ 500 nm [2]. Moreover, for perovskite, the tunability of the band gap is a span from 1.17 to 3.00 eV, which can perfectly adapt the requirement of the ideal top cell band gap (~ 1.67 -1.75 eV) [5]. The perovskite top cell can be p-i-n or n-i-p structures, and mesoscopic or planar structures, which makes the perovskite top cells to be fit in various conditions and requirements. Several typical materials of perovskite top cell are MAPbI_3 , FAPbI_3 , CsPbBr_3 and so on. In addition to the typical materials mentioned above, there are other materials doped with Sn, Cl or other elements through compositional engineering to make the PSVK to be suitable with other layers. Besides tunable band gap, the lattice constant of perovskite can be tuned through substitution of halide elements. Comparatively, a significant difference exists between the lattice parameters (a) of MAPbCl_3 (5.78 Å), MAPbBr_3 (6.04 Å) and MAPbI_3 (6.39 Å) [11]. In general, the tunable lattice constant means tunability on material thickness. Because of high structure tolerance of perovskite, substitution of halide elements would not break the crystalline structure of perovskite. Such diverse materials offer a wide range of options for subcells pairing. The PSC structure is shown in Figure 3.

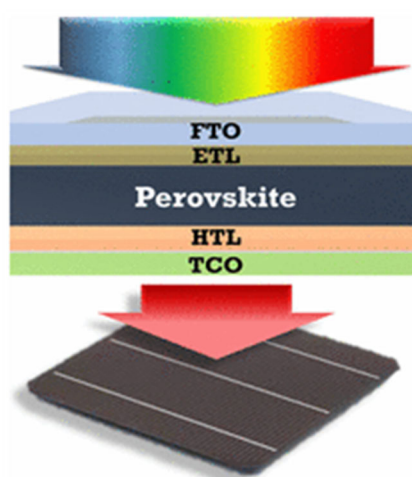


Figure 3. Perovskite solar cell structure. [12]

The fabrication of perovskite is easier than c-Si and lower cost. Commonly, the perovskites are fabricated on transparent conductive oxide (TCO) coated glass substrates with low vertical surface roughness less than 5nm [10]. Coating techniques such as spin-coating, inkjet printing, blade, and slot-dye coating are used to fabricate perovskites on TCO [10]. Among these techniques, spin-coating is generally used to small-scale fabrication in laboratory, while blade and slot-dye coating are used to large-scale industrial fabrication [13]. Besides solution methods, vacuum fabrication is also an efficient method to grow perovskite onto the bottom cells. These diversified fabrication methods of perovskites supply various choices when integrating perovskites with different bottom cells, further enhancing feasibility perovskites top cells.

4. Rear cells

4.1 Crystalline-silicon (c-Si)

Crystalline-Silicon (c-Si) is a technologically mature material in PV market, accounting for more than 90% of market share [14]. In addition, c-Si appeals to numerous researchers by the 1.12 eV band gap that it can absorb range of light with energy lower than the light range absorbed by top sub-cell to enlarge the range of absorption of the whole device [15]. Most common research is devoted to the

development of silicon heterojunction (SHJ) solar cells for bottom cells due to its high PCE and high V_{oc} . According to the newest report, the PCE of Perovskite/Si tandems achieves 28.7% in lab [10].

The top layer of SHJ is ITO which is able to contact with p-i-n structured PSC. In SHJ, double textured side structure is commonly used to reduce reflective loss, because the textured surface is beneficial for Near Infrared (NIR) light trapping. Generally, random pyramid textures are employed onto the surfaces [16]. However, the textured surface is incompatible with spin-coating which is used to grow PSC onto a flat surface. Therefore, to make the surface planar enough to facilitate PSC fabrication, an extra chemical-mechanical polishing (CMP) is necessary [17].

If a double-side-polished (DSP) SHJ is applied as a bottom cell of TSC, the perovskite will be easily grown onto the Si surface by low-temperature fabrication. However, due to the large amount of reflective loss and poor current matching, DSP is not competent for excellent bottom cell materials. As the fabrication of PSC does not impose restrictions on the rear side of bottom cell, a rear textured and frontally polished SHJ is qualified to be the bottom cell. A PSC with C60 as ETL and Spiro-OMeTAD as HTL can be grown on the polished front surface. The comparison between several SHJs with different surfaces is shown in Figure 4. Such a method has achieved the following remarkable results. Huang et al. spin-coated

$\text{Cs}_{0.15}(\text{FA}_{0.83}\text{MA}_{0.17})_{0.85}\text{Pb}(\text{I}_{0.7}\text{Br}_{0.3})_3$ onto a ITO layer of polished-front side SHJ to produce a 2T PSC/SHJ TSC with a PCE of 25.4% and a V_{oc} of 1.80V [18]. Xu et al. spin coated $\text{FA}_{0.78}\text{Cs}_{0.22}\text{Pb}(\text{I}_{0.85}\text{Br}_{0.15})_3$ on an ITO layer of SHJ to achieve 1 cm^2 TSC PCE of 27.1% [19]. Regarding commercial and industrial availability, the highest efficiency of large area Perovskite/SHJ of 57.4 cm^2 is 22.6% with a V_{oc} of 1.72v reported by Kamino et al [20].

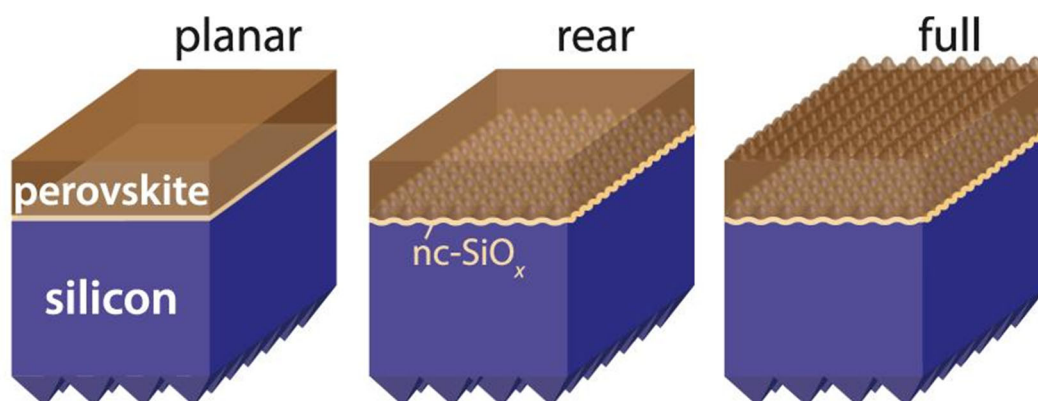


Figure 4. Process of planar top cell; process of nc-SiO_x layer; process of textured top cell.[21]

In the research of enhancing PCE of TSC, the low temperature tolerance of SHJ, which is less than 200°C, is a challenge of attempting on different materials combination. This characteristic limits fabrication feasibility of PSC, for example, an annealing temperature higher than 400°C is required to process efficient PSCs containing c-TiO₂ or m-TiO₂ [22]. To address this issue, a passivation by poly-Si/SiO_x (TOPCON) is available.

4.2 CIGS

In addition to silicon, another outstanding candidate is CIGS ($\text{Cu}(\text{In,Ga})\text{Se}_2$), which has a bandgap in between 1.08 and 1.15 eV. Similar to perovskite, the bandgap of CIGS is tunable from 1.00 to 1.70 eV through compositional engineering [23]. At the same time, due to that both of PSC and CIGS are thin-film, PSC/CIGS TSC can be high power with low mass (higher power to weight ratio than other configurations of PTSCs [10].) As a result, PSC/CIGS TSC can be potentially used in mobile devices and building integrated photovoltaics. Furthermore, the PSC/CIGS TSC is propitious to space

condition because both PSC and CIGS are proton radiation-hard [24]. Commonly, a CIGS solar cell is structured as substrate/ Mo/ CIGS/ CdS/ ZnO, where p-n junction is formed by CIGS and CdS. Since this architecture is designed in such a polarized order, as shown in Figure 5, only the inverted configured PSCs (p-i-n) can be the top cell of TSCs. (While Si bottom cell can match with both of the two junction types) However, this condition is not a complex problem to Perovskite. Through research and experiments, the inverted PSCs are more capable to reduce optical loss [13].

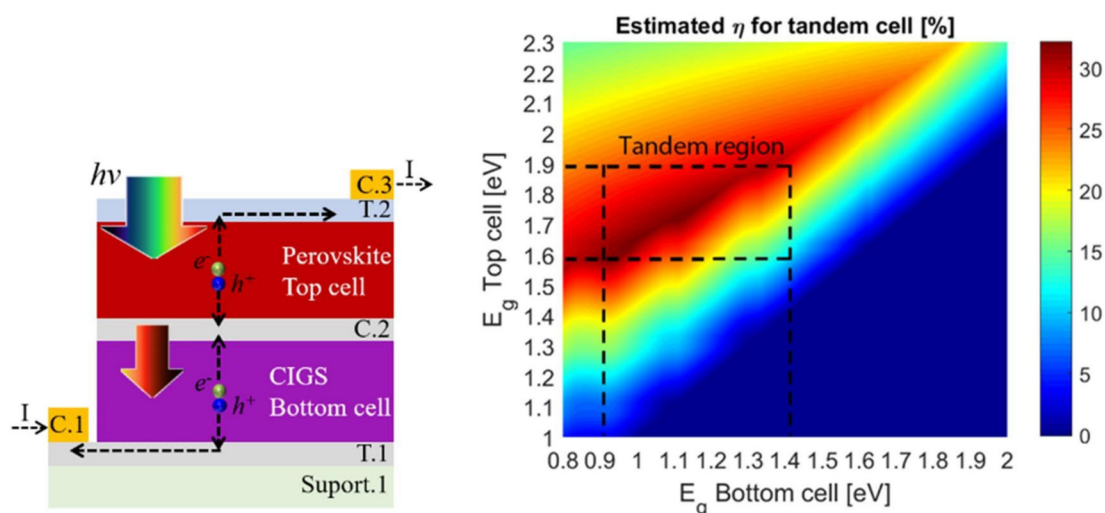


Figure 5. The 2-terminal Perovskite/CIGS architecture, and the estimate of optimum PV performance of the architecture.[25]

Compared to Si, advantages of CIGS bottom cell are:

- (1) Tunable bandgap of both top and bottom cell to potentially achieve higher PCE
- (2) Less sensitivity to impurities, which lower the strict fabrication requirement
- (3) Direct bandgap and thin-film, which reduce material and energy cost
- (4) Easily fabricated on flexible substrate.

At the same time, the rough surface is a greater challenge for CIGS than for Si. There are two strategies to circumvent the challenge, i.e., polish TCO RL or directly planarize the surface of CIGS by solution fabrication. However, the high PCE of CIGS comes from vacuum deposition, while solution fabrication would cause a crucial damage to PCE. Therefore, polishing on RL is a more feasible method to give consideration to maintain the high PCE of CIGS subcells and fabrication of PSC. Han et al [26]. used CIGS with structure of B-doped ZnO (BZO)/i-ZnO/CIGS/CdS/Mo/glass as the bottom cell, where the vertical roughness of the layer of BZO is 250nm. However, the maximal thickness of perovskite absorber is 300–600nm. Therefore, it is difficult to grow homogeneous PVSK on such a rough surface. An ITO layer with 300 nm of thickness is deposited on the BZO surface and a CMP process with SiO₂ slurry is employed on the surface of ITO. As BZO layer is not affected by the polishing process, the structure retained. Through the process, the roughness of PSC growing surface is reduced to 40 nm from 250 nm, which is enough for PSC fabrication. At the same time, PCE of bottom cell decreases to 16.76% from 18.73% with a V_{oc} of 674-676 mV, which is an acceptable loss. They attained 22.4% PCE from the entire tandem device of Cs_{0.09}FA_{0.77}MA_{0.14}Pb(I_{0.86}Br_{0.14})₃/CIGS (with E_g of 1.59 eV /1.00 eV).

4.3 Perovskite

In the above two sections, two different materials for bottom cells are introduced. However, two PSCs can also be combined together as a tandem device through tuning bandgaps. Compared to Si and CIGS, the easily tunable bandgap, flexible substrate selection and low cost make PSC another powerful candidate of bottom cell. To achieve a high PCE, 0.8-1.2 eV is the suitable range of bandgap for bottom cells. Because of the related studies on PSC/Si and PSC/CIGS devices, it is acknowledged

that the single-junction PSC can achieve high PCE with 1.53-1.62 eV of bandgap. Therefore, the main challenge of all PSC tandem device is developing a suitable bottom cell with appropriate bandgap. Replacing the Pb ions with Sn is an effective method to reduce the bandgap from more than 1.55 eV to 1.20-1.35 eV. However, due to the oxidization of Sn^{2+} ions, the PCE of the bottom cell will be reduced [27]. According to research, alloying with Pb can help to restrain oxidization of Sn ions [28]. For bottom cell, the B ion of ABX_3 is composited by Sn and Pb. Another issue is current matching of the two subcells. Because of low absorption cross-section of Sn-Pb based PSC, in order to attain 80% EQE, the thickness of the bottom cell should be more than $1\text{ }\mu\text{m}$.

To circumvent the issues above, Snaith et al. used $\text{FA}_{0.75}\text{Cs}_{0.25}\text{Sn}_{0.5}\text{Pb}_{0.5}\text{I}_3$ (with E_g of 1.2 eV) as bottom cell to combine with top cell $\text{FA}_{0.83}\text{Cs}_{0.17}\text{Pb}(\text{I}_{0.5}\text{Br}_{0.5})_3$ (with E_g of 1.8 eV) to achieve a PCE of 17.0%, with a J_{sc} for top cell equal to $14.1\text{ mA}\cdot\text{cm}^{-2}$ and a J_{sc} for bottom cell equal to $15.8\text{ mA}\cdot\text{cm}^{-2}$. A scanning electron micrograph of this design is shown in Figure 6. According to the results, the PCE is limited by current mismatching, and the current is limited by the top cell. To improve this situation, Snaith et al [29], introduced Cl into Sn-Pb perovskite to enlarge the bottom cell thickness for completely absorbing light passing through the top cell and match the current. The introduced Cl increased the bandgap to 1.25 eV and grain size and reduced electron disorder. The PCE of bottom cell achieved 18.4% with thickness of bottom cell absorber of 750 nm as Cl occupies 2.5% of content. Then, the bottom cell is combined with $\text{FA}_{0.8}\text{Cs}_{0.2}\text{Pb}(\text{I}_{0.7}\text{Br}_{0.3})_3$ (with E_g of 1.75 eV) and RL: Ag /MoOx /ITO. Finally, the device achieved 21.0% for top cell, and 20.7% for bottom cell.

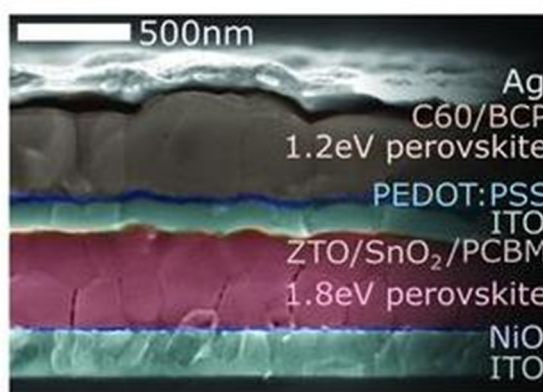


Figure 6. all-perovskite tandem micrograph. [29]

5. Discussion

PTSCs are expected to achieve 35% of PCE by improvement of PSC, RL and electrodes. However, developing PTSCs to a commercial level relies not only on increasing PCE, but large-scale production and stability are even bigger challenges. The following are several possible approaches to overcome these challenges and advance PTSCs to a commercial level.

In industry, excellent PV performance of large area PTSCs relies on high homogeneity. Furthermore, a high homogeneity of PTSCs comes from large-scale fabrication. However, the currently commonly used technique, spin-coating, is not able to achieve a high uniformity of PCs in large-scale fabrication, and it is not the best for flexible devices either. As a result, large-scale deposition is studied to overcome the problem. Among the various depositions, automated-slot dye and spray coating are the most promising methods. These two fabrication methods can maintain homogeneity while processing a large area PTSC [13].

In addition to the problem of homogeneity, the process of TCO deposition possibly damages to bottom subcell in large-scale fabrication. There are two feasible strategies to address this issue, which

are (1) develop a new type of orthogonal solvent system and (2) develop a sufficiently functional barrier layer to protect the bottom cells [30].

Similar to single-junction solar, the stability of PTSCs affected by intrinsic factors. Since replacing metal electrode by TCO, the stability of PTSCs is higher than single-junction solar cells. However, stability of PTSCs can be further improved, with the possibility of reaching the expected 25 years. For the most of PTSCs, the stability can be improved mainly through improvement of PSC subcells, because the stability of Si, CIGS subcells are already high enough to reach 25 years. To enhance the stability of PSCs, MA can be replaced by Cs, Rb and FA at A position, while I can be replaced with Br at X position [21].

6. Conclusion

To summarize, in this review, the author reviews the development of three 2T PTSC device designs, as well as the challenges and solutions in the process. Their current status and the challenges of elevating the PTSCs to the commercial level are also summarized. For the latter, author introduces and discusses several solutions that have been envisioned or are being studied. The main challenge is the improvement on stability of the Perovskite layer itself through compositional engineering, and discover certain fabrication technologies to be developed for large-scale fabrication to drive the PTSCs to commercial level. Regarding the question "Which type of PTSCs is the best?", the answer is ambiguous. As mentioned in the previous sections, each type of PTSCs has its own unique advantages, convenience, and applicable environment. Moreover, since the technology of PTSCs is not yet mature, each type of PTSCs has considerable potential. Among the different types, the most likely to attain the theoretically expected 35% PCE are the three cells mentioned in this review, which are Perovskite/Si, Perovskite/CIGS and Perovskite/Perovskite tandems devices. If PTSCs can be produced on a large scale while maintaining a high PCE, PTSCs will hopefully become a new product to replace Si-based cells in the PV market, and enable the replacement of traditional energy sources with solar clean energy on a boarder scale. Thanks to recent rapid advances in PTSCs, related technologies and research are expected to alleviate environmental problems and the energy crisis.

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