Perovskite/silicon tandem solar cells based on different layers and structures

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Abstract. Perovskite/silicon tandem solar cells offer great potential for achieving high-efficiency solar energy conversion. Through the absorption of a wide range of wavelength sunlight, they are not limited by the Shockley-Queisser limit in single-junction silicon solar cells. They have shown impressive progress in recent years, reached a power conversion efficiency (PCE) of 32.5%, and have a bright prospect in the future. In light of the condition of the solar industry today, this article mainly introduces the development status and the main progress direction of the perovskite/silicon tandem solar cells in recent years. In terms of efficiency improvement, it primarily uses perovskite unit band gap tuning and recombination layer optimization. Then the typical cell structures (2-terminal and 4-terminal) and some innovations (3-terminal) are introduced. The summary of these frontier researches is valuable and can guide the direction for later improvement. Lastly, the perovskite/silicon tandem solar cells still have many opportunities and challenges and are expected to become more mainstream solar cells.

Keywords: perovskite/silicon tandem solar cells, power conversion efficiency, band gap tuning, recombination layer, innovative structures.

1. Introduction

Solar cells have a long history of more than one hundred years. Due to the limited reserves of petrochemical energy worldwide and the severe environmental pollution of petrochemical energy, renewable energy development is significant. Renewable energy has many advantages, including a wide range of energy sources, easy access, apparent safety, and technical support, and solar energy meets most of the standards. Compared with other renewable energy sources, the photovoltaic (PV) industry has higher generation efficiency and matches the electric system in enterprises nowadays. They are why solar energy is regarded as the most promising form of renewable energy. Crystalline silicon solar cell is the most popular variety, but its PCE is difficult to exceed the Shockley-Queisser limit of 33.77%. Thus, emerging PV developed rapidly, and perovskite/silicon tandem solar cell (TSC) is the most promising one, with great potential to break through this limitation. The limit of the PCE of double-junction TSC can reach 46.1% [1].

The tandem technique in perovskite/silicon TSC combines the silicon bottom cell with a tight band gap and the perovskite top cell with a large band gap. Thus, TSC combines the advantages of both two sub-cells. Solar spectrum of both long and short wavelengths can be effectively covered by the bottom silicon and top perovskite sub-cells. It can be seen in Figure 1. Furthermore, it has an adjustable band

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gap, low photon thermalization, and high open circuit voltage. These good qualities help perovskite/silicon TSC receive the attention of many scientists and have a bright prospect for development. The conversion efficiency increased rapidly, from 23.6% in 2016 to 32.5% in 2022 [2].

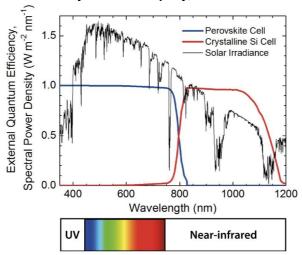


Figure 1. Spectrum of perovskite/silicon TSC. Perovskite absorbs in the visible region, and silicon absorbs in near-infrared light [3].

Perovskite/silicon tandem solar cell has two most common structures, 4-terminal (4T) and 2-terminal (2T), presented in Figure 2. The former consists of two independent cells with four divided electrodes. In this structure, perovskite's band gap range is broader because two sub-cells do not interfere with each other. And the operational processes of the two units are separated completely. By using the best manufacturing conditions of their own, the quality of the two cells can be better. In the 2T structure, the top perovskite unit deposits directly on the bottom silicon unit, connecting with the recombination junction (RJ). This configuration has a lower cost, requiring only one broad spectral transparent electrode, and its parasitic absorption losses are lower. But it requests current matching because the unit with a larger current is limited. Both two mainstream structures have been widely studied, and their efficiency has improved significantly recently.

The PCE of perovskite/silicon TSC still has excellent potential for development. In addition to some general improvement methods in solar cells, such as transparent electrodes, surface reflection reduction, parasitic absorption reduction, etc., the most advanced improvements focus on the following aspects. Perovskite band gap tuning help TSC absorb sunlight of broad spectral. The recombination layer is a critical part of the 2T structure. And some structural innovations provide more potential for efficiency progress.

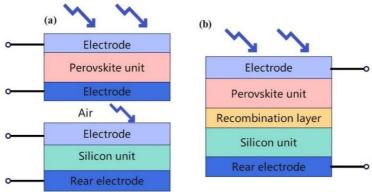


Figure 2. Mainstream perovskite/silicon PSC structures. (a) 4-terminal structure. (b) 2-terminal structure.

This review will focus on these progress strategies and present some recent research benefits to efficiency. In addition, the challenges and opportunities for future development are also foreseen.

2. Perovskite band gap tuning

The band gap of perovskite in ABX₃ structure needs to match silicon for more efficient solar absorption, and its tuning can be achieved by element replacement with a wide range. In most cases, cation A is organic, cation B is metal, and anion X is halogen. The band gap of the most prevalent substance, MAPbI₃, is around 1.6 eV. The number deviates from the optimal band gap width in theoretical simulation. Replacing different elements is the most critical factor for PCE and other performance parameters.

The mainstream methods to tune band gaps are the replacement of metal A and halide X. For example, by bromine substitution of different ratios of iodine, the band gap of MAPb(I_xBr_{1-x})₃ may be tuned under control from about 1.6 eV to 2.3 eV continuously. And a small part of the chlorine can also be used to replace it, but it cannot effectively broaden the band gap because of the significant ionic radius difference. Jixian Xu et al. used triple-halide alloys (I, Br, Cl) to increase Cl solubility, and carriers' lifetime and mobility became twice. Notably, the phase separation was significantly suppressed at 100 times sunlight illumination intensity and 4% reduced degradation after 1000 hours of operation at 60 °C's maximum power point. After integrating with the silicon bottom cell, this TSC reached 27% PCE for a 1 square centimeter device [4]. However, perovskite solar cells with wide band gap have stability problems, leading to higher open circuit voltage (Voc) loss caused by illumination, water, heat, etc. Perovskites composed of Br and I are especially sensitive. Lattice defects can lead to charge accumulation, nonradiative recombination, and other adverse effects, which are the main reasons for reduced stability. Improving stability is the leading research direction, which can effectively enhance the device use time while achieving higher efficiency. Daehan Kim et al. used (FA_{0.65}MA_{0.2}Cs_{0.15})Pb(I_{0.8}Br_{0.2})₃ as the material and its wide band gap at 1.68 eV. By adding a mixture of I and SCN as PEA-based additives, a 2D phase was created, and various PV parameters can be adjusted with the different ratios of SCN and (SCN + I). After being illuminated for over 1000 hours, the efficiency maintained at 80% of the initial [5]. Furkan H. Isikgor et al. used a global passivation strategy. Multi-functional materials such as phenformin hydrochloride (PhenHCl) can passivate surface defects effectively in both rich and poor electron domains. High Voc enhancement occurred in single-junction perovskite devices. However, in perovskite/silicon TSC, this effect of V_{OC} did not present obviously, but the PCE still reached 27.4% [6]. Yuxin Yao et al. made an improvement at 4T TSC. They used ammonium diethyldithiocarbamate (ADDC) to reduce I₂ to ions I and inhibited defect proliferation and migration. At the same time, this method decreased the environmental hazards by I2. In this research, Voc reached 1.24 V in semitransparent perovskite TSC with outstanding stability. And in 4T perovskite/silicon TSC field, solar cells integrated this technology achieved the highest efficiency (30.24%) [7].

Another method is to replace cation using smaller ions to twist the lattice structure in control. The band gap can broaden or narrow by tilting a crystal cell or simply contracting the lattice [8]. Additionally, heavy metals' environmental pollution and toxicity, such as lead, are significant problems. Thus, B-site cations also replace to reduce this effect. Tin is usually chosen as a substitute because it has a similarly high efficiency but without toxicity. Rahul Pandey et al. used MASnI₂Br as perovskite material, and this 2T TCS had V_{OC} of 2.14 V and PCE of 30.7%. However, the instability of Sn ion oxidation can lead to the degradation of device performance, so further optimization is required [9]. Perovskite oxide ferroelectric material is another promising new photovoltaic material, represented by BiFeO₃. Because of its unique bulk PV effect, it receives more research attention. However, the PCE is lower because the band gap of perovskite oxide is much broader than the optimum band gap of TSC. S. Zhang et al. used an artificial superlattice structure consisting of BiFeO₃ and BiCrO₃ and replaced a part of oxygen with sulfur. This method successfully reduced the band gap and maintained the ferroelectricity, making it a better prospect for use in the photovoltaic field [10].

3. Recombination junction

The recombination layer is a critical component of 2T perovskite/silicon TSC. It is important in connecting the perovskite top and silicon bottom units. Here, majority carriers of two subcells recombine, but minority carriers cannot. The voltage in a series structure is the product of two components, while the current is the smaller of two values. Thus, the current matching is significant in TSC. As the bridge between two subcells, the critical influencing factors of RJs are high transmittance and conductance to realize the effective composite of many carriers and low reflectivity. To achieve this effect, the longitudinal resistance of the material should be as small as possible, and the transverse resistance should be as considerable as possible to decrease the transverse current.

The common structures of the Si bottom cell are passivated emitter rear-cell (PERC), tunnel oxide passivated contact (TOPCon), and heterojunction (HJT) cells. The former can be large-scale produced, while the latter two have a better potential for improvement due to their passivation contact, and the materials of all these structures need to adjust. The transparent conductive oxide (TCO) layers are chosen in the PERC structure as significant recombination layers. TCO is frequently made from compounds like indium tin oxide (ITO) and indium zinc oxide (IZO). To get better properties, thickness matching is a significant part. For example, the optimal thickness of the IZO recombination layer is about 40-50 nm, where light absorption is minimized, and excellent current matching consequences are achieved. Outstanding optical and electrical properties are presented in them. However, sputtering deposition in the TCO manufacturing process can cause damage to the silicon bottom unit, and the high temperature can raise costs and affect the quality of the cells. Scientists usually use front design, like Si-based materials, a-Si: H, nc-Si: H. Figure 3 shows one representative of these structures. It can prevent the need for and harm from sputtering processes. Passivation methods are not only used in bottom manufacturing but can also be used to reduce passivation damage in composite layers. Silvia Mariotti et al. combined passivating polycrystalline silicon on oxide (POLO) front junction and PERC bottom cell. This technology could reduce sputtering damage and fit into current industrial technologies. This TSC had a potential PCE of 29.5% based on optical simulation [11]. Woojun Yoon et al. implemented an ITO recombination layer on poly-Si passivated by SiO_x. The damage could be mitigated effectively at a high temperature of 250 cent degrees. In this technology, a PCE of 25% was obtained [12].

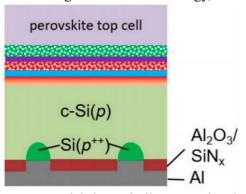


Figure 3. Schematic of a perovskite/PERC TSC with n^+/p^+ -Si [13].

For TOPCon, ITO materials have similar sputtering damage. The recombination layer is easy to form an opposing p/n junction. It can be solved by heavy doping in silicon. Chunhui Shou et al. deposited an extra a-Si:H doped with boron as the front design on the surface of n^+ -poly-Si and made ohmic contact with low resistance by rapid thermal anneal (RTA). This method could effectively reduce the interdiffusion of B-doped and P-type silicon. At 700 cent degrees, the RTA method optimized the recombination junction, achieving the best results. The minimal contact resistance at this point was only $16 \text{ m}\Omega/\text{cm}^2$. In addition, a forming gas annealing (FGA) process could recover the declining passivation quality caused by RTA [14].

In the HJT recombination layer, the ITO layer also fits well. However, the refractive index is mismatched with that of other layers. The nc-Si:H is a better substitute. It has a more suitable index to match, and this layer is manufactured in plasma enhanced chemical vapor deposition (PEVCD) technique, avoiding deposition damage in the ITO process. MgF₂ antireflection coating (ARC) is another

material to reduce the reflection of the recombination layer. Incorporating it in the appropriate place can reduce the problem of refractive-index mismatch. Lujia Xu et al. observed decreased short-circuit current due to the glass and thermoplastic polyurethane, and an optical model was designed using refractive index engineering. In this improved structure, MgF₂ ARC was moved from the recombination layer's surface to the glass's outer surface. As a result, the front reflection of sunlight of almost all the wavelengths and the PCE of all the TCS reached 26.2% [15].

4. Innovative structures

The two most common structures of perovskite/silicon TSC are 4-terminal and 2-terminal. In the 4T structure, perovskite and silicon subcells are divided into two separate units. In addition to mechanically stacking the two cells directly, spectral separation methods can absorb different wavelengths of sunlight by perovskite and silicon, respectively. In this structure, the currents of the two cells do not need to be precisely matched, and fewer transparent electrodes are required, but the cost of this method is relatively high. In the 2T structure, the recombination junction connects two subcells. However, this structure is subject to more restrictions. In recent years, some innovative configurations can avoid the disadvantages and have better potential.

The bifacial structure can significantly boost short-circuit current, especially for monolithic perovskite/silicon TSC. In bifacial cells, sunlight from the ground reflection can be absorbed through the rear transparent electrode. Figure 4 presents a typical bifacial structure. This configuration improves light energy utilization in the actual industrial power generation process. This effect is particularly pronounced in areas with highly reflecting ground, such as sand, snow, etc. The bifacial structure requires a smaller band gap for better current enhancement. A smaller band gap in the perovskite/silicon TSC means less bromine replacement is used in the tuning process, resulting in better stability. At the same time, the current matching problem can be effectively solved. The research of this configuration has been studied for a long time and has recently been used in TSC improvement. If TSC uses the bifacial structure, it is calculated that the output power could be 66% more than the normal mono-facial solar cell. Moreover, the optimized band gap of perovskite top cells with bifacial structure is 1.55 eV and 1.12 eV for silicon units [16].

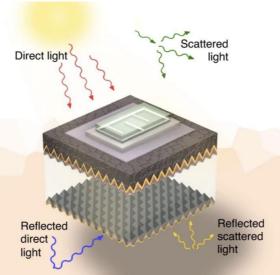


Figure 4. Sunlight absorption of bifacial perovskite/silicon TSC [17].

Besides the classic 4T and 2T structures, some innovative designs like 3T are presented. Rudi Santbergen et al. introduced the three-terminal structure. A novel design was created, with one terminal in the front part and two more in the rear position in this arrangement, and the bottom and top cells have the same or opposite polarity. This 3T tandem structure combined the advantages of conventional two-and four-terminal forms. It could reduce parasitic absorption losses and accommodate a wide variety of

perovskite thicknesses because just one transparent conductive layer is required. It was found that the parasitic loss can be effectively reduced when the hole transport layer is under the perovskite material, but the reflection was increased. By combining and improving the structure, the best result was achieved. In the simulation, the combination of a perovskite cell with 22.7% efficiency and a silicon cell with 24.9% efficiency could give a PCE of 32% when connected in series [17]. Philipp Wagner et al. designed a 3T perovskite/silicon TSC in 2022. It was similar to the 2T structure, but an additional electrode was added to perform the current regulation. As shown in Figure 5, staggered rear contacts and p/n recombination connections were put between two units. The maximum PCE, at 24.9% in the experiment, was achieved when the photocurrent reached a maximum, regardless of the effect of current matching. Additionally, the voltage was unrelated to the spectral, improving the robustness [18].

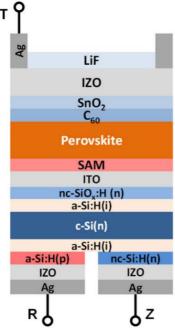


Figure 5. Schematic of the 3T perovskite/silicon TSC with interdigitated rear contacts and a p/n RJ [15].

5. Conclusion

Perovskite/silicon TSCs have the highest efficiency among emerging PV devices. In recent research, the improvement mainly focuses on these aspects. For perovskite band gap tuning, halide and cation replacement can regulate effectively to absorb sunlight in broad spectrum in control. In addition, it can reduce toxicity and other negative impacts. For recombination layer improvement, three different structures of silicon bottom subcell have specific ways to improve RJs. The front design usually enhances passivation and reduces sputtering deposition damage. Besides traditional 4T and 2T structures, innovative 3T designs avoid current matching problems and have better potential, and bifacial electrodes can improve the output power.

In the future, besides above directions, the challenges of preparing perovskite/silicon TSC with higher PCE are presented in these main aspects. The problem of crystal defects still needs to be solved to enhance the carrier diffusion length and photothermal stability. And ion migration problem of the broadband gap PSCs still needs to be solved effectively. Although perovskite/silicon TSC have high efficiency in laboratories, reducing their production cost and making them more valuable is significant.

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