The Road to Breaking Through Limits: Improving Photoelectric Conversion Efficiency of Perovskite Solar Cells

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Abstract. With the rising energy demand, solar cells, renowned for their sustainability, have developed rapidly due to their pollution-free characteristics. The pursuit of lower electricity costs and higher conversion efficiency remains the core goal of the advancement of solar cells. At present, the photoelectric conversion efficiency (PCE) of crystalline silicon cells, which currently the mainstream of commercial technology, is approaching its theoretical limit, leading to intense market competition. In contrast, there is significant room for improvement in the PCE of perovskite cells. This article compares the performance of three types of solar cells, including single junction perovskite cells, crystalline silicon/perovskite stacked cells, and perovskite/perovskite stacked cells, prepared through experiments. The experimental result confirms that perovskite has much greater potential as an absorbing material with enhanced conversion efficiency than crystalline silicon. This research highlights an effective method to break the Shockley Quether limit of single crystal silicon cells for stacked cells such as crystalline silicon/perovskite and perovskite/perovskite. This work positions the perovskite stacked cells as a transformative technology with high efficiency and cost advantages, poised for significant industrial growth.

Keywords: Perovskite cell; solar stacked cell; photoelectric conversion efficiency; crystalline silicon cell.

1. Introduction

Solar power generation is known as one of the most ideal new energy sources, with characteristics such as cleanliness, pollution-free, and renewability. However, its disadvantages such as low energy distribution density and low conversion efficiency limit the widespread application of solar cells [1,2].

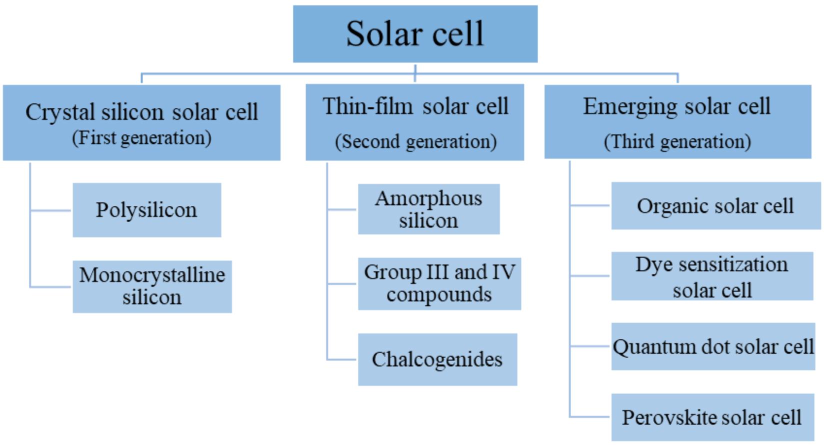

Fig. 1 Development History of Solar Cells (Picture credit : Original)

With the core goal of reducing costs and increasing efficiency, solar cell technology continues to update, gradually evolving from the first generation to the third generation of solar cells. As shown in Fig. 1 and Table 1, the most mature mainstream solar cell currently is the first generation of crystalline silicon solar cells based on silicon materials. At present, crystalline silicon batteries maintain the highest conversion efficiency of this generation of batteries, but due to the limitation of the absorption coefficient of crystalline silicon itself and the impact of energy and material consumption, their room for improvement is becoming smaller and smaller, gradually entering a bottleneck period of development.

The second generation is thin-film solar cells, represented by cadmium telluride (CdTe), copper indium gallium selenide (CIGS), and gallium arsenide (GaAs) cells. With advantages such as light weight, transparency, and flexibility, they have gained certain development space in application scenarios such as photovoltaic building integration. However, due to the scarcity of material reserves, toxicity, complex preparation processes, and low mass production conversion efficiency, they are difficult to produce on a large scale and their market share is far inferior to that of crystalline silicon cells.

The third generation is emerging solar cells, among which perovskite solar cells are increasingly favored due to their excellent properties such as adjustable bandgap (as shown in Fig. 2), high light absorption coefficient, long carrier transport distance, high defect tolerance [3], and easy film formation. In addition, they have the advantages of simple preparation process, low cost [4], and minimal negative impact on the environment [5].

Table 1. Three Scheme Comparing

Characteristic	First generation crystalline silicon	Second generation			Third generation perovskite
		GaAs	CIGS	CdTe	
Theoretical efficiency	29.40%	27%	33%	28%	33%(single knot)
Lifetime	25 years	20 years (CIGS、CdTe)			To be verified
Stability	Very good	Relatively good			Sensitive to external environment
Material cost	High purity silicon raw materials	Some raw materials are difficult to obtain			Low raw material prices and low purification requirements
Technological process	There are multiple links and steps in the industrial chain	——			Short industrial chain
Energy consumption	High temperature requirements and high energy consumption	Relatively low			The maximum temperature only requires 150℃

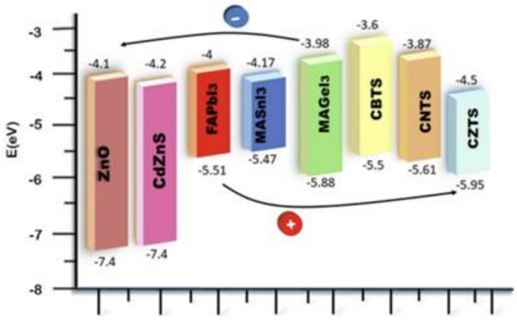

Fig. 2 Range of Bandgap Regulation for Perovskite [6]

2. Theoretical Analysis

2.1. Photoelectric conversion efficiency（PCE）

Conversion efficiency is an important indicator of the progressiveness of solar cell technology. Due to additional losses caused by various factors such as reflection and interface defects, the conversion efficiency of solar cells has always been low. According to the Shockley Queisser limit [7], the band gap of a single crystal silicon cell is approximately 1.12 eV, and the theoretical maximum conversion efficiency is 29.4%. The ideal band gap of a single junction perovskite cell is about 1.4 eV, and the theoretical maximum conversion efficiency is 33.7%.

As shown in Fig. 3, taking the current mainstream crystalline silicon cells as an example, the highest efficiency of crystalline silicon cells produced under the best laboratory conditions is 27.09%. This record was achieved by Longi Green Energy in December 2023, and the conversion efficiency of crystalline silicon cells is gradually approaching the efficiency ceiling, with the marginal benefits of technological improvements becoming smaller and smaller; The conversion efficiency of mass-produced crystalline silicon cells may decrease, with the optimal range reaching 19%, typically between 16% and 18%.

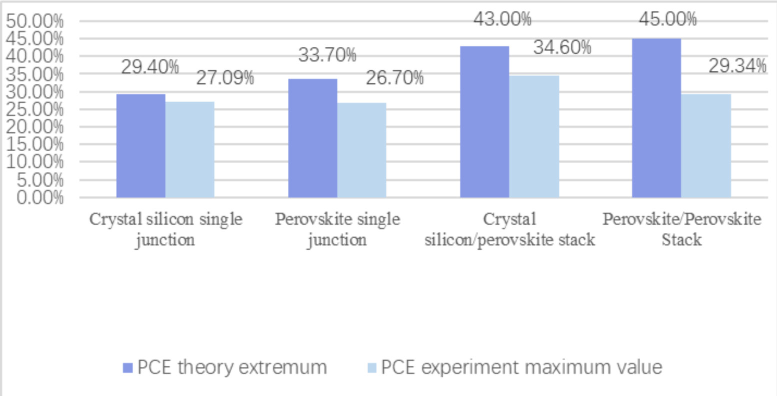

Fig. 3 Range of Bandgap Regulation for Perovskite [6]

By comparison, as of July 22, 2024, updated data on the official website of the Natural Resource Ecology Laboratory (NREL) shows that single junction perovskite solar cells can achieve a certified steady-state efficiency of 26.7% (Fig. 4). In May 2024, Guangyin Technology collaborated with a research team from Shanghai Jiao Tong University to increase the highest conversion efficiency record of perovskite/perovskite stacked cells to 29.34%; In July 2024, Longi Green Energy achieved a breakthrough in the conversion efficiency of crystalline silicon/perovskite stacked cells, with a conversion efficiency of 34.6%.

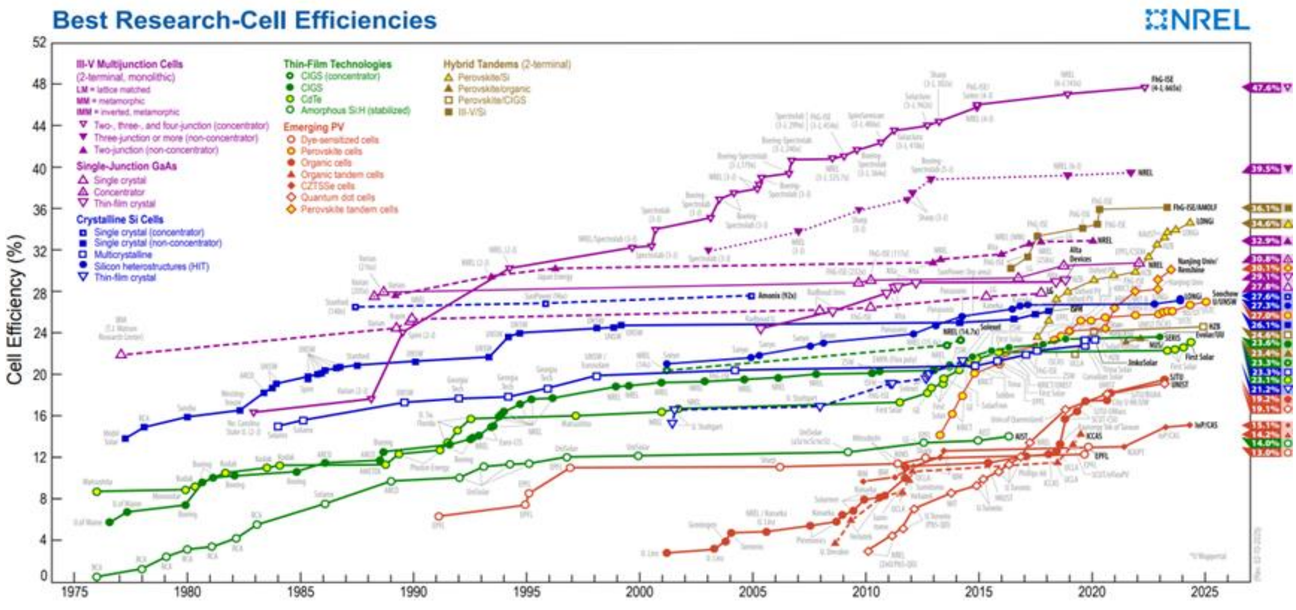

Fig. 4 Efficiency Chart of Emerging Solar Cell Champions [8]

2.2. Stacked batteries are a breakthrough in improving conversion efficiency

The utilization ability of a single semiconductor material for solar energy is limited, while materials with different band gaps have different utilization abilities for different wavelength parts of solar light. As shown in Fig. 5, placing materials with wider band gaps above to mainly absorb and utilize short wavelength light, and materials with narrower band gaps below to mainly utilize long wavelength light, can improve the absorption rate of the entire spectrum of solar light, maximize the utilization of solar energy, and achieve higher conversion efficiency.

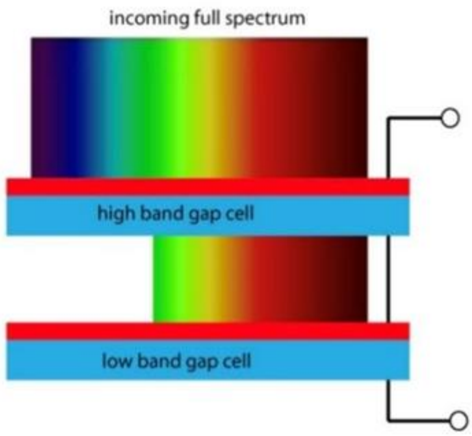

Fig. 5 Principle of Light Utilization in Stacked Cells [9]

Stacked cells are composed of multiple sub cells with different band gaps, which can effectively broaden the range of solar energy spectrum utilization and improve conversion efficiency. The adjustable band gap of perovskite enables it to serve not only as a wide band gap absorber layer, but also as a narrow band gap absorber layer, which is beneficial for the preparation of crystalline silicon/perovskite stacked cells and perovskite/perovskite stacked cells. In order to allocate the absorption spectra of each single junction cell reasonably and achieve higher efficiency in stacked batteries, it is necessary to solve the problem of matching between the upper and lower cells [10].

The highest theoretical conversion efficiencies of crystalline silicon/perovskite and perovskite/perovskite stacked cells can reach 43.00% and 45.00%, respectively. The theoretical efficiency advantage is obvious, and it is currently a feasible solution that is expected to break through the theoretical limit of single junction solar cell conversion efficiency and achieve a conversion efficiency of over 30%.

In the specific experimental process, the perovskite/crystalline silicon stacked cell is a layer of wide band gap perovskite material stacked on top of the crystalline silicon cell, with minimal investment in perovskite cost. It can break the efficiency barrier of existing solar cell products without changing the existing crystalline silicon industry chain. Considering the actual production economy, perovskite/crystalline silicon stacked batteries are currently the main technological breakthrough direction in the industry.

The theoretical conversion efficiency of perovskite/perovskite stacked cells is high, and they can break free from the cost and performance constraints of crystalline silicon, fully utilizing the advantages of perovskite materials such as low cost, strong light absorption, and low-temperature processing. These characteristics make perovskite/perovskite stacked cells an important development direction for future solar cells, and even one of the ultimate paths for the future industrial development of solar cells.

Based on the characteristics of perovskite materials and the theory of stacked cells, this article designs experiments to confirm that perovskite has the property of adjustable band gap width. By adjusting the band gap, perovskite materials are applied to the preparation of stacked cells as both a wide band gap and narrow bandgap absorbing layers, thus exploring the role of perovskite in improving the conversion efficiency of stacked cells

3. Experimental Section

3.1. Main indicators of solar cell performance

The main performance indicators of solar cells include open circuit voltage (Voc), short-circuit current density (Jsc), fill factor (FF), photoelectric conversion efficiency (PCE) and so on. The open circuit voltage indicates the voltage of the solar cell when there is no input power. Short-circuit current is the current of the solar cell when the output power is rated. The fill factor represents the number of electrons per unit area that a solar cell fills when its output power is rated. These metrics collectively define the efficiency, charge transport capability, and overall performance of a solar cell.

3.2. Data and analysis of single junction perovskite cells

In this experiment, 5 sets of single junction perovskite cells were prepared, and the perovskite absorbing materials were $FA_{0.9}Cs_{0.1}PbI_3$, $FA_{0.852}Cs_{0.148}PbI_{2.7}Br_{0.3}$, $FA_{0.84}Cs_{0.16}PbI_{2.625}Br_{0.375}$, $FA_{0.828}Cs_{0.}172PbI2_{.55}Br_{0.45}$ and $FA_{0.702}Cs_{0.298}PbI_{2.44}Br_{0.56}$.The band gaps are 1.52 eV, 1.58 eV, 1.60 eV, 1.61 eV and 1.68 eV, respectively, referred to as P-1, P-2, P-3, P-4 and P-5. Six batteries were prepared simultaneously for each group, and the parameters obtained are shown in Table 2.

As shown in Figure 6, among the 5 groups of perovskite cells, the maximum PCE value is 20.28%, which is lower than the world record for conversion efficiency (26.7%) but is already at the upper limit of PCE for mass-produced crystalline silicon cells, reflecting the potential of perovskite to improve PCE compared to crystalline silicon.

Table 2. Three Scheme Comparing

TYPE	Perovskite absorbing material	Band Gap (eV)	Voc (V)	Jsc(mA/cm^2)	FF	PCE (%)
P-1	$FA_{0.9}Cs_{0.1}PbI_3$	1.52	1.10782	22.29574	82.1038	20.2794
			1.033592	22.39329	78.9912	18.2829
			1.08702	22.3119	80.6748	19.5664
			1.074484	22.46671	80.3187	19.389
			1.050473	22.39518	77.7832	18.2989
			1.068277	22.46137	81.4982	19.5555
P-2	$FA_{0.852}Cs_{0.148}PbI_{2.7}Br_{0.3}$	1.58	1.119625	20.98187	80.3319	18.8714
			1.126687	21.03221	80.4656	19.0677
			1.116542	20.81879	79.5208	18.4847
			1.110322	20.77622	78.2128	18.0424
			1.115731	20.8337	79.0416	18.3731
			1.110048	20.97456	80.2371	18.6814
P-3	$FA_{0.84}Cs_{0.16}PbI_{2.625}Br_{0.375}$	1.60	1.139917	20.01417	77.414	17.6616
			1.137361	19.60335	77.5453	17.2896
			1.127813	19.65948	77.4497	17.1723
			1.12076	19.9535	78.2976	17.5098
			1.121892	20.08155	78.2571	17.6308
			1.131761	20.50237	80.0775	18.581
P-4	$FA_{0.828}Cs_{0.172}PbI_{2.55}Br_{0.45}$	1.61	1.160004	19.23661	78.6707	17.555
			1.148372	19.15895	77.727	17.1012
			1.139503	19.42317	77.9554	17.2537
			1.1329	19.66235	78.4242	17.4694
			1.134049	19.62405	78.5742	17.4864
			1.134632	20.14314	80.0472	18.2948
P-5	$FA_{0.702}Cs_{0.298}PbI_{2.44}Br_{0.56}$	1.68	1.167309	19.73208	81.1265	18.6862
			1.154651	19.86013	81.0598	18.5882
			1.153501	19.97059	81.5103	18.7768
			1.154529	20.22049	82.2517	19.2018
			1.158004	20.19396	81.8139	19.1319
			1.158499	20.19619	81.7155	19.1192

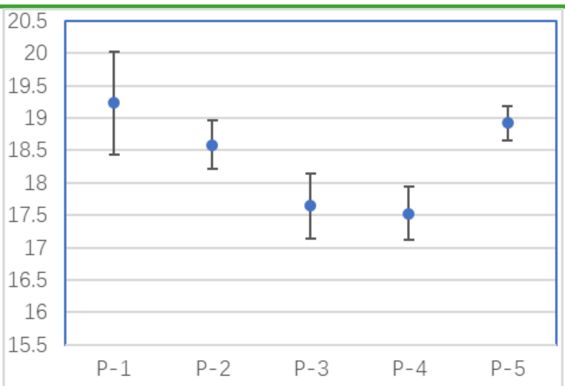

Fig. 6 PCE (%) of 5 Sets of Single Junction Perovskite Cells (Picture credit : Original)

1.1 Data and analysis of perovskite/perovskite stacked cells

According to the preparation process of perovskite/perovskite stacked cells, 16 sets of perovskite/perovskite stacked cells were prepared in this experiment, with 6 cells prepared simultaneously for each set. The composition of perovskite/perovskite stacked cells is: FTO/NiO_x/Ph_4PACz/PVSK/PEACl/PCBM/SnO_2/IZO/PEDOT/NBG/PEACl/PCBM/SnO_2/Ag.

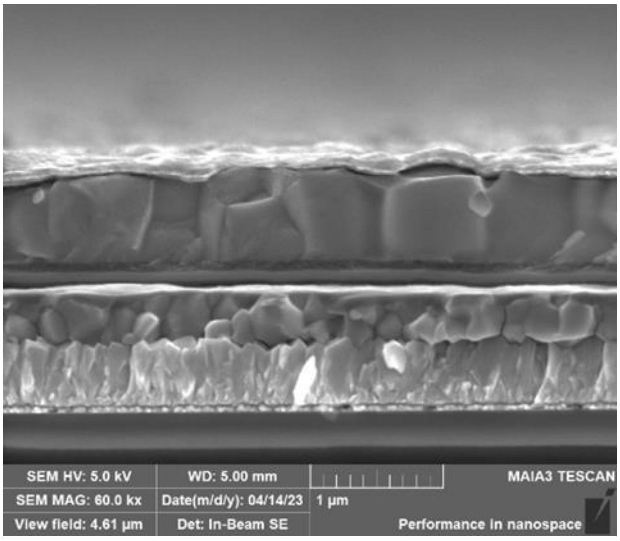

Fig. 7 Electron Microscopy Photo of Perovskite/Perovskite Stacked Cells (Picture credit : Original)

Fig. 7 is an electron microscope image of a perovskite/perovskite stacked cell with a narrow band gap perovskite absorption layer on the upper area of the picture and a wide band gap perovskite absorption layer on the lower area. Table 3 shows the data of the highest PCE cell, and Fig. 8 shows the J-V parameter graph of the highest PCE cell. As shown in Table 3, the maximum PCE of perovskite/perovskite stacked cells is 25.04%, which is close to the highest conversion efficiency of crystalline silicon cells prepared under optimal laboratory conditions (27.09%). From this, it can also be seen that perovskite, as an absorbing material for solar cells, has greater potential for improving conversion efficiency than crystalline silicon.

Table 3. Three Scheme Comparing

TYPE	Voc(V)	Jsc(mA/cm^2)	FF	PCE(%)	Band Gap(eV)
WBG	1.29	18	82.42	19.17	1.78
NBG	0.83	29.45	80.93	19.69	1.25
All	1.99	15.16	82.69	25.04	

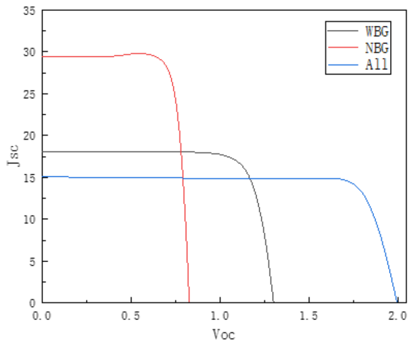

Fig. 8 Electron Microscopy Photo of Perovskite/Perovskite Stacked Cells (Picture credit : Original)

The preparation of perovskite/perovskite stacked solar cells faces many challenges, one of which is the heterogeneous crystallization of wide band gap and narrow band gap perovskite layers. High PCE perovskite cells require a dense, uniform, and suitable high-quality perovskite absorber layer with consistent grain size. However, due to the short coating time window, perovskite films often cannot form uniformly, and the crystallization process is not synchronized up and down, which can easily lead to a large number of defects at the bottom of the film. Especially for narrow band gap perovskite films, the crystallization process is too fast and difficult to control, and the problem of uneven films is more likely to occur, all of which can affect efficiency. In addition, light reflection, parasitic absorption, and large dead zones in modules can also cause varying degrees of optical loss, leading to a decrease in efficiency.

3.3. Data and Analysis of Crystal Silicon/Perovskite Stacked Cells

In this study, wide band gap perovskite sub cells were stacked on top of crystalline silicon sub cells to form a crystalline silicon/perovskite stacked cell. The perovskite materials were $FA_{0.9}Cs_{0.1}PbI_3$, $FA_{0.852}Cs_{0.148}PbI_{2.7}Br_{0.3}$, $FA_{0.84}Cs_{0.16}PbI_{2.625}Br_{0.375}$, $FA_{0.828}Cs_{0.172}PbI_{2.55}Br_{0.45}$, $FA_{0.702}Cs_{0.}298PbI2_{.44}Br_{0.56}$. The band gaps are 1.52 eV, 1.58 eV, 1.60 eV, 1.61 eV, 1.68 eV, respectively, referred to as Si-1, Si-2, Si-3, Si-4, Si-5. Five sets of crystalline silicon/perovskite stacked cells were simultaneously prepared with six cells.

Table 4. Three Scheme Comparing

TYPE	Voc(V)	Jsc(mA/cm^2)	FF	PCE(%)	Perovskite based solar cell absorbing materials	Band Gap(eV)
Si-1	1.705082	13.0957	76.103	25.6895	$FA_{0.9}Cs_{0.1}PbI_3$	1.52
	1.635921	13.3833	75.002	23.6911		
	1.657022	13.3119	76.675	24.7102		
	1.684484	13.3856	76.319	24.8911		
	1.640473	13.3641	73.783	23.8001		
	1.662771	13.2602	77.498	24.9488		
Si-2	1.728625	13.9798	76.323	24.5695	$FA_{0.852}Cs_{0.148}PbI_{2.7}Br_{0.3}$	1.58
	1.734687	14.032	76.477	24.7702		
	1.725542	13.82	75.532	24.1785		
	1.719322	13.776	74.224	23.7419		
	1.724731	13.834	75.053	24.0702		
	1.719048	13.974	76.247	24.3796		
Si-3	1.785917	14.515	73.413	23.7409	$FA_{0.84}Cs_{0.16}PbI_{2.625}Br_{0.375}$	1.60
	1.745351	14.6997	73.564	23.3702		
	1.735613	14.6601	73.458	23.2532		
	1.730176	14.95	74.29	23.5901		
	1.730791	14.0801	74.264	23.7111		
	1.740661	14.5999	76.088	24.6411		
Si-4	1.770005	14.237	74.669	23.6115	$FA_{0.828}Cs_{0.172}PbI_{2.55}Br_{0.45}$	1.61
	1.757373	14.16	73.733	23.1622		
	1.749601	14.4227	73.963	23.3142		
	1.740129	14.66	74.432	23.5122		
	1.743048	14.63	74.57	23.5506		
	1.745633	14.1422	76.052	24.3542		
Si-5	1.776308	15.7298	77.134	25.1332	$FA_{0.702}Cs_{0.298}PbI_{2.44}Br_{0.56}$	1.68
	1.764553	15.8599	77.06	25.0094		
	1.766501	15.97	77.52	25.2059		
	1.763429	15.2215	78.26	25.4311		
	1.768105	15.19	77.814	25.5002		
	1.769398	15.1972	77.727	25.5414		

Table 4 shows that the highest PCE value of the five sets of crystalline silicon/perovskite stacked cells is 25.69%, with an average value of 24.30%, which is close to the theoretical limit PCE of single crystal silicon cells (29.40%). The potential for PCE improvement in crystalline silicon/perovskite stacked cells is highlighted. The highest PCE value of crystalline silicon/perovskite stacked cells is lower than the current world record of 34.6% PCE for crystalline silicon/perovskite stacked cells, due to the use of crystalline silicon sub cells with a PCE of 16.39% (see Table 5), which is lower than laboratory prepared and high-performance crystalline silicon cells; matching degree between bandgap of perovskite sub cells and crystalline silicon cells; and factors such as preparation process and experimental skills.

The maximum PCE value of the crystalline silicon/perovskite stacked cell prepared in this experiment is 25.69%, and the maximum PCE value of the perovskite/perovskite stacked cell is 25.04%, which is only about two percentage points lower than the highest efficiency value of the monocrystalline silicon solar cell produced under the best laboratory conditions. Experiments have shown that both crystalline silicon/perovskite stacked, and perovskite/perovskite stacked cells can be options for improving the PCE of solar cells (Fig. 9).

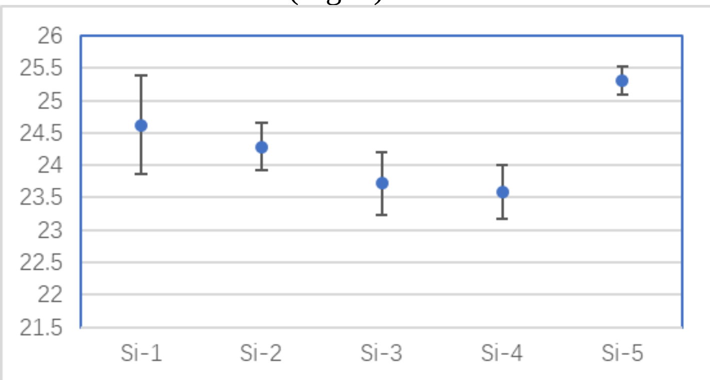

Fig. 9 PCE of 5 Sets of Crystalline Silicon/Perovskite Stacked Cells (Picture credit: Original)

This study further selected one cell from each of the five sets of crystalline silicon/perovskite stacked cells mentioned above and tested the relevant parameters of the crystalline silicon sub cells after passing through different band gap perovskites. The data is shown in Table 5.

Table 5. Important Parameters of Crystalline Silicon Sub Cells after Perovskites with Different Band Gaps

Type	Voc(V)	Jsc (mA/cm^2)	FF	PCE(%)	PCE Remain (%)	Perovskite based solar cell absorbing materials	Band Gap (eV)
Si (100%)	0.62845	37.6834	69.2045	16.3891	/	/	/
Si-1	0.60592	13.0889	68.4719	5.4303	33	$FA_{0.9}Cs_{0.1}PbI_3$	1.52
Si-2	0.60961	13.6399	68.5792	5.7024	35	$FA_{0.852}Cs_{0.148}PbI_{2.7}Br_{0.3}$	1.58
Si-3	0.60808	14.5731	68.6291	6.0816	37	$FA_{0.84}Cs_{0.16}PbI_{2.625}Br_{0.375}$	1.60
Si-4	0.60977	14.2915	68.9223	6.0063	37	$FA_{0.828}Cs_{0.172}PbI_{2.55}Br_{0.45}$	1.61
Si-5	0.61058	15.2651	69.0514	6.436	39	$FA_{0.702}Cs_{0.298}PbI_{2.44}Br_{0.56}$	1.68

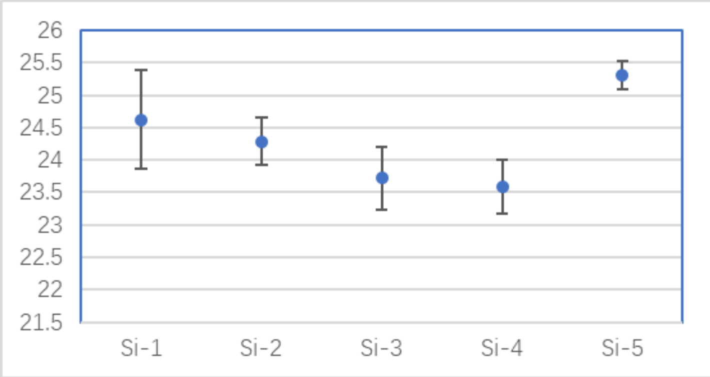

Fig. 10 Remaining Efficiency of Crystalline Silicon Sub cells after Perovskites with Different Band Gaps (Picture credit : Original)

Table 5 and Fig. 10 show that after passing through perovskites with different band gaps, the remaining PCE of the crystalline silicon sub cell is between 33% and 39% and increases with the increase of the perovskite band gap. This study shows that the larger the band gap, the better the matching between perovskite and crystalline silicon.

4. Conclusion

The pursuit of high PCE is the driving force behind the development of solar cells. The PCE of crystalline silicon batteries is approaching its theoretical limit, and the closer it is to the theoretical limit, the fewer ways to improve battery efficiency and the greater the difficulty of improving efficiency. Compared to other solar cell materials, the excellent performance of perovskite has attracted much attention. The maximum PCE value of the single junction perovskite cell prepared in this article is 20.28%, which has exceeded the current upper limit of PCE for mass-produced crystalline silicon cells, reflecting the potential of perovskite to improve PCE compared to crystalline silicon.

Compared with Shockley Quether limit efficiency, the PCE of stacked cells has great potential for improvement. The preparation experiment of perovskite stacked cells in this article shows that based on the adjustable gap characteristics, the advantages of perovskite are better reflected in stacked cells. The highest PCE values of the perovskite/perovskite stacked cell and crystalline silicon/perovskite stacked cell prepared in this experiment were 25.04% and 25.69%, respectively, which were only about two percentage points lower than the highest PCE value of the single crystal silicon cell prepared under the optimal experimental conditions, and four percentage points lower than the limit PCE of the single crystal silicon cell. The potential of perovskite as a solar cell absorbing material in terms of conversion efficiency far exceeds that of crystalline silicon.

This experiment confirms that crystal silicon/perovskite stacked cells can stack wide band gap perovskite materials on top of crystal silicon cells, with minimal investment in perovskite costs, without changing the existing crystal silicon industry chain, and can break down the efficiency barriers of existing solar cell products. Considering the actual production economy, crystalline silicon/perovskite stacked batteries are currently the main technological breakthrough direction in the industry. Compared with crystalline silicon/perovskite stacked cells, perovskite/perovskite stacked cells have higher theoretical conversion efficiency, can break free from the cost and performance constraints of crystalline silicon, and can fully utilize the advantages of perovskite materials such as low cost, strong light absorption, and low-temperature processing.

Perovskite/crystalline silicon and perovskite/perovskite stacked cells have achieved high PCE, but in order for perovskite stacked batteries to become mainstream, they still need to overcome some challenges, including improving the long-term stability of the battery, solving the problem of toxic elements such as Pb in the material, maintaining high efficiency after large-area preparation of stacked components, and further reducing production costs. However, considering the enormous potential of perovskite in terms of efficiency and cost, once the long-term stability of the device is solved, coupled with its flexibility, perovskite stacked cells are likely to become one of the important technological directions to break the current pattern of the solar cell industry and usher in explosive growth.

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