RESEARCH HIGHLIGHTS

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Stronger together: perovskite/silicon tandem solar cells

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Solar energy, as a renewable resource, is an effective solution to the current global energy shortage problem. To actively respond to the call for "carbon peak" and "carbon neutrality", solar cell industry has experienced unprecedented development. The full utilization of solar energy resources remains an urgent issue to be addressed.

Crystalline silicon solar cells, at present, benefit from significant cost reduction over the past decades due to advancements in fabrication technologies. In contrast, perovskite solar cells (PSCs), owing to their low-temperature solution processing and compatibility with scalable manufacturing methods, demonstrate great potential for fabrication cost reduction. Recent studies have highlighted that PSCs, especially in scenarios of large-scale production, could achieve lower costs[1]. The certified power conversion efficiency (PCE) of single-junction PSCs has exceeded 26%, approaching the Shockley–Queisser (S–Q) limit. However, stability and efficiency still lag behind those of traditional silicon solar cells. To fully utilize solar energy and overcome the restrictions of the S–Q limit, perovskite/silicon tandem solar cells (PSTSCs) emerged, leading the future direction of photovoltaics technology.

The basic principle of tandem solar cells involves superimposing subcells with different band gaps to achieve more efficient spectral utilization. Generally, PSTSCs have three device structures, as shown in Fig. 1(a): two-terminal (2T), three-terminal (3T), and four-terminal (4T)[2]. In 2T PSTSCs, the wide-bandgap top perovskite solar cell and the bottom silicon solar cell are connected through an interconnecting layer, effectively reducing parasitic absorption and manufacturing costs, making it the most popular perovskite/silicon tandem design. Both 3T and 4T PSTSCs offer promising pathways for improving device efficiency. However, they share certain drawbacks, including additional costs associated with their more complex structure and greater parasitic absorption due to additional layers. While the 4T PSTSCs benefit from independent subcells, addressing current matching issues, the 3T design potentially offers better integration with existing silicon technologies, albeit with more complex interconnection requirements. Compared to other tandem solar cells, PSTSCs exhibit higher efficiency and a more mature industrial chain, making them a crucial pathway to achieve photovoltaic industrialization.

In Fig. 1(b), compared to traditional single-junction perovskite solar cells, the efficiency of PSTSCs has increased from an initial 23.6% to 34.6% in just ten years[3]. In 2015, McGehee *et al*. prepared the first 2T PSTSC by connecting perovskite top solar cells and silicon bottom cells using tunnel composite junctions, achieving a PCE of 13.7%[4]. This work demonstrated the feasibility of stacking perovskite solar cells and traditional crystalline silicon solar cells, thereby accelerating the development of PSTSCs. In the early stages, limited by interconnecting layers and significant light loss, the efficiency of 2T PSTSCs consistently lagged traditional silicon solar cells. In 2016, McGehee's team used indium tin oxide (ITO) as the interconnecting layer and prepared the double oxide layer using atomic layer deposition (ALD), achieving a certified efficiency of 23.6% for the tandem cell, with an impressive open-circuit voltage (V_{OC}) of 1.65 V, a short-circuit current density (J_{SC}) of 18.1 mA·cm^{-2}, and a fill factor (FF) of 79.0% (Fig. 1(c)). This represents a significant step towards high-performance PSTSCs[5].

Since then, PSTSCs have benefited from the rapid development of perovskite top cells as well as silicon bottom cells and optimization of the interconnecting layer, achieving several milestone breakthroughs. In 2022, Zhou *et al*. mitigated the effects of damage-etched rough surfaces via poly(N,N′-bis-4-butylphenyl-N,N′-bisphenyl)benzidine (polyTPD) passivation, optimizing reflection and resistive losses using ray tracing, and redesigning c-Si structures for tandem integration. Incorporating a TOPCon-based c-Si design, advanced coatings, and interface engineering, the tandem device achieves 27.6% efficiency[6]. In 2024, Ye *et al*. developed a highly passivated p-type TOPCon structure by optimizing the oxidation conditions, boron in-diffusion, and aluminium oxide hydrogenation. Consequently, integrating with perovskite top cells, 1 cm^2 n–i–p perovskite/silicon TSCs exhibit V_{OC} exceeding 1.9 V and a high efficiency of 28.20% (certified 27.3%)[7].

Commonly used crystalline silicon cells have pyramidal texture structures, and perovskite films prepared by solution-processed spin-coating technique have difficulty in uniformly covering the surface of silicon cells. To overcome this challenge, researchers have made significant efforts. In 2023, Chin *et al.* addressed the issue of uniformly covering perovskite on textured silicon surfaces by involving hot evaporation of inorganic sources and subsequently spin-coating organic salts. They achieved an efficiency of 31.25% for the 2T PSTSC (Fig. 1(d))[8]. In the same year, Yao *et al*. employed a two-step mixed deposition method to prepare perovskite films, followed by surface passivation using dynamic spray coating (DSC) of phenyl-ammonium sulfide fluoride. This approach achieved a certified efficiency of 30.89% for the PSTSCs (Fig. 1(e))[9]. While the combination of evaporation and spin

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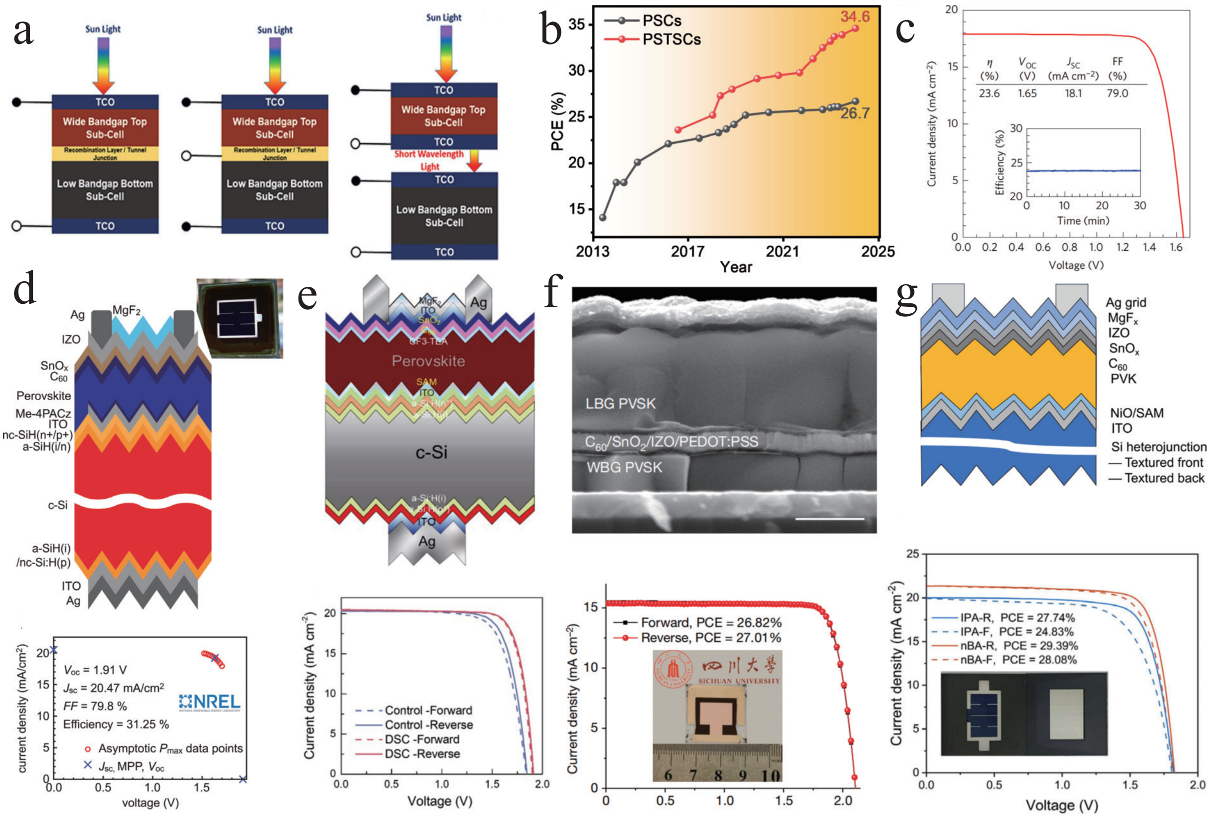

Fig. 1. (Color online) (a) Schematic diagram of PSTSCs: 2T (left), 3T (middle), and 4T (right). Adapted from Ref. [2]. Copyright 2022, Elsevier. (b) Evolution of the efficiency in PSCs and PSTSCs. (c) *J*–*V* curve and efficiency at the maximum power point (inset) of the champion tandem device. Adapted from Ref. [5]. Copyright 2015, Springer Nature. (d) Schematic illustration of the PSTSC and corresponding picture of the perovskite top cell on textured Si (up). Asymptotic maximum power scan (down). Adapted from Ref. [8]. Copyright 2023, American Association for the Advancement of Science. (e) Schematic view of a fully textured monolithic perovskite-silicon tandem (up). *J*–*V* scans of the control and DSC-treated tandem cells (down). Adapted from Ref. [9]. Copyright 2024, Wiley. (f) Cross-sectional SEM image of an all-perovskite TSC (up). *J*–*V* curve of the best-performing TSC (down). Adapted from Ref. [10]. Copyright 2023, Springer Nature. (g) Schematic diagram of perovskite/silicon heterojunction tandem solar cell (up). *J*–*V* curves of the tandem device under reverse and forward bias (down). Adapted from Ref. [12]. Copyright 2024, Springer Nature.

coating effectively addresses the challenges of depositing film on textured silicon surfaces, the wastage of evaporated materials and contamination of the evaporation chamber continues to hinder its widespread application.

In recent years, the emergence of self-assembled monolayer (SAM) has significantly improved the efficiency of both inverted single-junction and tandem perovskite solar cells. In 2023, we reported a promising SAM molecule, 4-(7H-dibenzo[c,g]carbazol-7-yl butyl)phosphonic acid (4PADCB). 4PADCB was used as a hole transport layer for a wide-bandgap perovskite solar cell, achieving the highest recorded efficiency (26.4%) for 1-cm^2 all-perovskite tandem solar cell at that time (Fig. 1(f))[10]. SAM also significantly enhances PSTSCs. In 2023, De Wolf's team used the common SAM, [2-(9H-carbazol-9-yl)ethyl]phosphonic acid (2PACz), as the hole transport layer for the PSC and optimized the interconnecting layer to enhance optical properties. This resulted in a certified efficiency of 32.5%, the highest efficiency achieved in the laboratory at that time[11]. In 2024, Tan *et al.* used nickel oxide/SAM as the hole transport layer and solvent engineering to develop PSTSCs that can be prepared in large areas under ambient conditions (Fig. 1(g))[12]. SAMs can enhance the efficiency of tandem solar cells as hole transport layers of perovskite top cells, but their structural instability also restricts their widespread application.

The PSTSCs have been rapidly developing in the past decade, which efficiency surpasses that of silicon solar cells and gradually approaches the S–Q limit. However, several problems still need to be solved. Firstly, the efficiency problem persists. Although the emergence of SAM and the design and regulation of the interconnecting layers can improve device efficiency and reduce optoelectrical losses to some extent, suitable solutions to uniformly preparing perovskite films on textured silicon surfaces are lacking. One might deeply investigate the crystallization processes of perovskite films on the surface of textured silicon and compositional engineering, as well as. Secondly, poor stability remains a significant challenge for commercialization. Further development is needed to address stability issues in PSTSCs to meet the 25-year device lifespan required for commercial applications by suppressing the phase segregation of wide-band-gap perovskites via additive engineering and interfacial modification, as well as blocking the ion migration and stabilizing the ions at the interface. Additionally, more efforts should also be directed towards scalability issues to accelerate commercialization, especially in vacuum deposition preparation technology, which can scale up the production of top PSCs on textured silicon cells with good reproducibility and stability. Meanwhile, the toxicity of lead should be carefully considered. Pb-based PSCs are frequently utilized as wide-band-gap top cells in PSTSCs. However, lead is a toxic heavy metal with detrimental effects on human health and the environment. Therefore, addressing the issue of lead leakage is crucial for accelerating the commercialization of PSTSCs, which can be achieved through composition engineering to reduce or replace lead, as well as additive engineering to absorb excess lead. We still have a lot of works to do in the future to promote PSTSC technology for photovoltaic building integration, automotive applications, medical electronics, space equipment, etc. We believe that these challenges could be overcome, and PSTSC technology will lead the future development of photovoltaic technology.

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