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Commentary

Enhancing perovskite/silicon tandem solar cells via nuclei engineering

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Perovskite solar cells (PSCs) have emerged as a promising candidate for next-generation photovoltaic devices and offer potential applications as an alternative to conventional fossil fuels. Over the past decade, solution-processable organic–inorganic halide perovskites have achieved remarkable power-conversion efficiencies (PCEs), comparable to those of their silicon counterparts, owing to their high absorption coefficients and charge densities [1,2]. Notably, integrating wide-bandgap (WBG) perovskites with silicon solar cells to construct perovskite/silicon tandem solar cells (PSTs) renders a significant commercial potential, with recent advances resulting in the PCEs of PSTs exceeding 33% [3]. However, despite rapid improvements in PCEs, WBG PSCs, which function as the top cell in PSTs, exhibit unsatisfactory crystallinity and a weak texture with disordered orientations, resulting in phase segregation and mixed halide-cation ion migration [4].

Hence, Chen et al. devised an innovative nuclei engineering approach to enhance the crystallinity and stability of WBG absorbers [5]. As shown in Fig. 1(a), this approach effectively suppressed the formation of bromine-rich and hexagonal (2H, δ) phase clusters by favoring the formation of the cubic (3C, α) phase. By merely adding long-chain alkylamine ligands (e.g., oleylammonium iodide, OAmI) to the precursor solution, researchers achieved 3C perovskite with uniform nucleation, which enabled the development of films with enhanced crystallinity and texture. These long-chain alkylamine ligands uniformly nucleate the desired 3C perovskite phase by reducing the surface energy of the perovskite crystal facets. The ligands serve as a passivation layer that inhibits the formation of bromine-rich aggregates and promotes a uniform halide distribution. The optimized PSTs achieved PCEs of 32.5% and 29.4% for active areas measuring 1 and 25 cm^2, respectively, while demonstrating remarkable operational stability (Fig. 1(b)). The encapsulated devices retained more than 90% of their initial efficiency after 800 h of operation at 50 °C, thus underscoring the practicality of this approach.

To characterize the optimal nucleation of the WBG absorbers, in-situ grazing incidence wide-angle X-ray scattering (GIWAXS) and photoluminescence (PL) measurements were performed. By adding OAmI nuclei, distinct (001) perovskite crystal facets with enhanced diffraction intensity, superior crystallinity, and strong texture could be observed. This approach minimized energy losses, which were quantified by quasi-Fermi-level splitting, and demonstrated applicability across various WBG perovskite compositions. Compared with the control films, which exhibited polycrystalline structures and a weak texture owing to DMSO-based 2H phase formation, the engineered films showed a strong texture with a preferred (001) orientation. In contrast to the Br-rich nuclei-dominated 2H phase formation in the control films, the target films indicated predominantly 3C perovskite nucleation, as evidenced by in-situ PL measurements. Moreover, time-resolved photoluminescence measurements revealed an extended charge-carrier lifetime, which indicated a reduction in nonradiative recombination centers in the target films. Consequently, the target absorbers exhibited enhanced resistance to ion migration and phase segregation under continuous illumination and thermal stress, which can be attributed to their superior crystallinity and strong texture.

Pursuing stability remains the cornerstone of PST research. This study provides a robust understanding of the mechanisms correlating crystallinity and phase stability in WBG absorbers. The application of nuclei engineering to precisely customized crystal growth underscores the critical role of crystallinity in WBG perovskites.

In conclusion, the nuclei engineering approach represents a transformative advancement in the field of perovskite photovoltaics, and it effectively suppresses ion migration and improves texture, thereby mitigating the primary causes of phase segregation in WBGs. By addressing critical challenges related to crystallinity and stability, this study not only extends the efficiency boundaries of PSTs but also provides new avenues for their commercialization and broader application in energy generation.

Outlook

The nuclei engineering approach represents a significant advancement in the development of stable and efficient PSTs. By adopting

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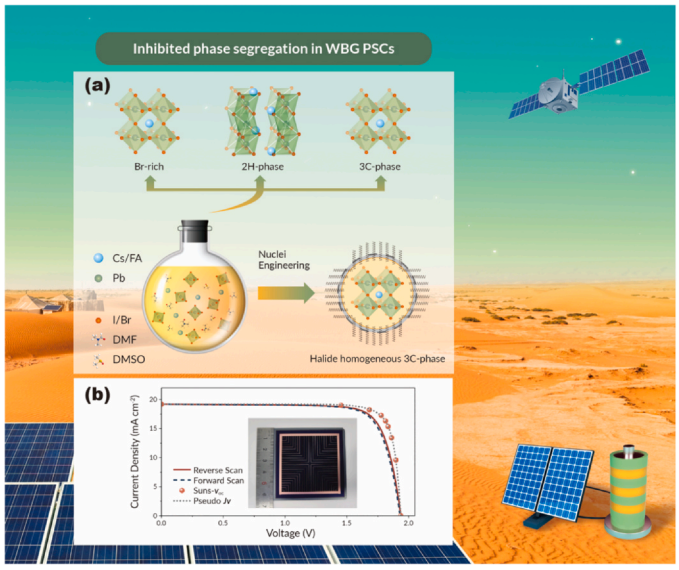

Fig. 1. **(a)** Scheme of nuclei engineering for control and target WBG absorbers. **(b)** *J–V* curves of champion 25 cm^2 PST cell measured under standard AM 1.5G sunlight under ambient conditions. (Inset) Photograph of 25 cm^2 PST cell. Reproduced with permission from Ref. [5]. Copyright 2024, AAAS.

nuclei engineering to control the crystallization process, this investigation facilitates future innovations related to WBG perovskites.

Moreover, operational stability remains a significant challenge in the field of PSCs. Although nuclei engineering has demonstrated superior crystallinity and phase stability compared with control devices, the operational and damp-heat stabilities of PSTs are inadequate for market-scale production. Future research should focus on inhibiting light-induced phase segregation and achieving decades-long operational stability using robust encapsulation techniques. This study highlights the critical role of crystallinity in WBG PSCs and provides new avenues for the development of perovskite-based optoelectronic devices. Although significant progress has been achieved in nuclei engineering for PSTs, several challenges remain to be addressed, as follows:

1. **Energy Loss in Charge-Selective Layers**. Energy loss caused by charge-selective layers must be minimized. Improving the energy-level alignment at the interfaces between the perovskite and both the hole transport layer and electron transport layer (ETL) is crucial.
2. **Parasitic Absorption in Organic ETLs**. High parasitic absorption losses, particularly from widely used organic fullerene-based ETLs, such as C_{60} and PCBM, continue to pose challenges. Addressing this issue may involve investigating alternative materials for ETLs, such as inorganic SnO_x and TiO_x.
3. **Stability**. Although nuclei engineering has enhanced device efficiency and crystallinity, stability during damp-heat operation and real-world operation remains a key issue. Pathways for testing and recording these stability issues should be addressed.
4. **Market development**. Achieving the scalable fabrication of high-performance PSTs will require further advancements in robust encapsulation techniques and a reduction in the levelized cost of energy.

CRediT authorship contribution statement

Gang Li: Resources. **Ruijie Ma:** Writing – review & editing, Supervision, Project administration. **Peter Müller-Buschbaum:** Writing – review & editing. **Xinyu Jiang:** Writing – review & editing. **Dongyang Li:** Writing – original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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