COMMENTARY

Stable and efficient wide-bandgap perovskite for perovskite-organic tandem solar cells

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The ongoing enhancement of perovskite and interfacial layers characteristics are propelling power conversion efficiencies (PCEs) of metal halide perovskite solar cells (PSCs) close to the Shockley-Queisser limit. To achieve an efficiency breakthrough, one of the most feasible and effective approaches is to construct a tandem solar cell (TSC) by integrating two or more single-junction solar cells with complementary absorption. In contrast to PSCs, organic solar cells (OSCs) display distinctive benefits, such as superior extinction coefficients and PCEs in the near-infrared region, making OSCs the perfect counterpart to PSCs. Nevertheless, the perovskite layer can effectively filter out most of the ultraviolet light to help improve the photostability of OSCs. Hence, perovskite-organic TSCs, comprising wide-bandgap (WBG) (1.7 ~ 1.9 eV) PSCs as the front sub-cell and narrow-bandgap (NBG) (1.1 ~ 1.3 eV) OSCs as the rear sub-cell, hold significant promise for achieving high PCEs and excellent stability.[1]

PEROVSKITE-ORGANIC TSCS

The optimization of perovskite-organic TSCs performance is comprehensively divided into three parts: the performance of WBG PSCs, the performance of OSCs, and the design of interconnection layers (Figure 1). The advent of acceptor-donor-acceptor structured non-fullerene acceptors (NFAs) has significantly narrowed bandgaps compared to fullerene derivatives such as $PC_{61}BM$ and $PC_{71}BM$. Moreover, with considerable efforts

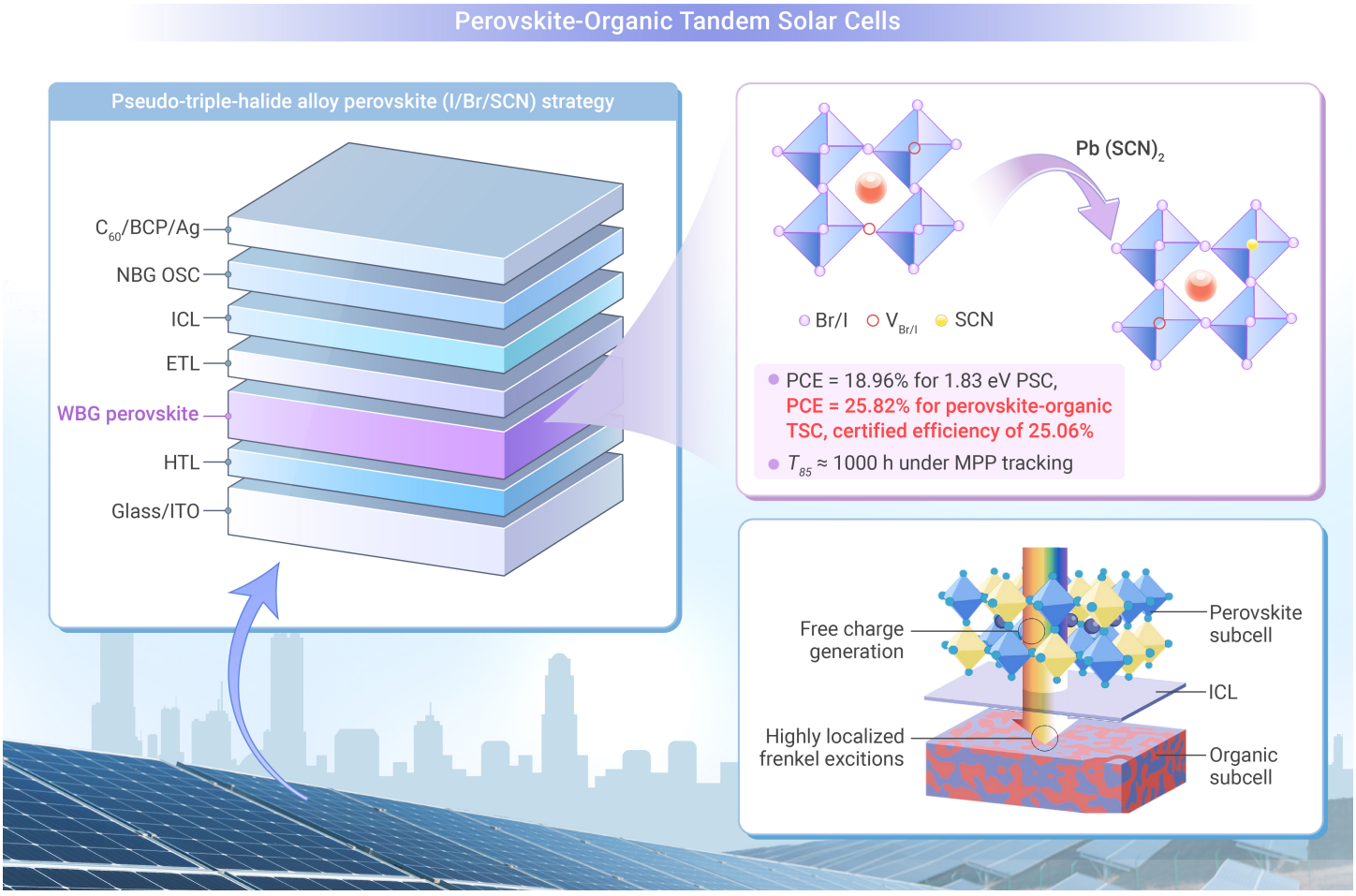

Figure 1. Pseudo-trihalide WBG perovskite for perovskite-organic TSCs.

focused on optimizing NFAs' photovoltaic properties and active layer micromorphology, OSCs have now exceeded 19% PCEs achieved by Y6 derivatives, ensuring a better alignment of both bandgaps and efficiencies with WBG PSCs for fabricating efficient perovskite-organic TSCs. For all-perovskite solar cells, a dense metal interconnect layer is necessary to prevent solvents used in the back-cell from dissolving the front-cell, a limitation that has hindered the development of such solar cells. However, this challenge does not apply to perovskite-organic TSCs, as the small polar solvents, such as

chloroform and chlorobenzene, used in processing the active layers of OSCs do not cause re-dissolution of the perovskite front-cell. Consequently, the requirements for the buffer (or recombination) layer are minimal, and only a thin metal layer is enough, thereby imposing fewer restrictions on the development of perovskite-organic TSCs.[2] Overall, the preparation of efficient and stable WBG PSCs has become a pivotal issue for advancing perovskite-organic TSCs.

WBG PSCS

Due to instable crystal structure and uncontrollable crystallization process, WBG perovskite crystals tend to generate lattice defects that will cause ions migration inside the crystals and interface recombination, leading to greater energy losses and poor device performances in WBG PSCs. Since 2020, perovskite-organic TSCs have experienced rapid growth, with PCEs increasing from 17% to 25.82% (certified 25.06%), mainly due to the steady enhancements in the photovoltaic performances of WBG PSCs. In 2020, Chen et al. achieved a high-efficiency perovskite-organic TSC with a PCE of 20.6% (certified 19.54%) by combining a 1.77 eV bandgap PSC with a 1.41 eV bandgap OSC.[3] In 2022, Chen et al. further improved the PCE of perovskite-organic TSC to 23.6% (certified 22.95%) by passivating the surface of a NiO_x hole transport layer with benzylphosphonic acid to reduce surface recombination losses in a 1.79 eV bandgap perovskite that achieved a maximum PCE of 17.08%.[4] To obtain high-performance perovskite-organic TSCs, further investigation into wider bandgap (> 1.80 eV) PSCs is necessary. However, wider bandgap perovskites are prone to severe halogen segregation, leading to significant non-radiative recombination.

Recently, Li's group proposed a pseudo-trihalide alloy perovskite model incorporating iodide (I), bromide (Br), and thiocyanate (SCN) (Figure 1). By introducing SCN^-, adjustments can be made to both the macroscopic crystal morphology and microscopic lattice components of WBG perovskite, which comprehensively inhibits the migration and phase separation of halogen ions in the lattice and film to some extent, thus reducing the energy loss of single-junction WBG PSCs. The introduction of a small amount of SCN^- into the perovskite delayed crystallization and thus reduced grain boundaries and increased carrier diffusion length. Additionally, a trace amount of SCN^- remaining in the bulk phase can preferentially enter the perovskite lattice to occupy halogen vacancies and form an I/Br/SCN alloy, limiting halogen ions migration within the lattice through steric hindrance.[5] Compared with traditional passivation methods, SCN^- provides a more comprehensive defect passivation effect and increases the halogen migration activation energy. Consequently, PSCs with a 1.83 eV bandgap achieved an 18.96% PCE along with a very high open-circuit voltage of 1.32 V. Perovskite-organic TSCs based on this WBG perovskite as the front sub-cell achieved the PCEs of 25.82% (certified efficiency of 25.06%) and 24.45% respectively with the active areas of 0.0580 and 1.004 cm^2, representing the highest efficiencies reported to date for perovskite-organic TSCs. The outstanding PCE of the device with the active area of 1.004 cm^2 demonstrates the feasibility of the strategy proposed by Li's group for developing large-area devices. Furthermore, the tandem device maintained over 85% of its initial efficiency after 1,000 hours of maximum power point tracking.

The passivation strategy based on this quasi-ternary halide perovskite effectively mitigates halogen segregation and phase separation, which typically occur due to high Br content (50%) in perovskite with a bandgap exceeding 1.80 eV, far surpassing the photostability threshold (20% Br content) of halide perovskite. The introduction of the Lewis base $Pb(SCN)_2$ regulates WBG perovskite crystallization and diminishes grain boundary paths for halide ion migration. Simultaneously, a trace amount of SCN^- infiltrates the perovskite crystal lattice, occupying halide ion vacancies. This Lewis base, with its comprehensive passivation function, offers a more effective approach to controlling crystallization and defect passivation in WBG perovskite.

CONCLUDING REMARKS

WBG perovskite containing mixed halogens is conducive to the fabrication of perovskite-organic TSCs. Introducing the pseudo-halogen SCN^- ion inhibits halogen ion migration, promotes crystallization, and diminishes grain boundaries. SCN^- ions can also enter the crystal lattice to occupy iodide (I^-) vacancies, inhibiting ion migration and reducing energy loss in WBG PSCs through the steric hindrance of SCN^-. The quasi-trihalide alloying strategy in WBG PSCs will facilitate the development of stable and efficient perovskite-organic TSCs. Due to the straightforward preparation process of perovskite-organic TSCs, they are also expected to facilitate the construction of flexible and large-area devices.

REFERENCES

1. Brinkmann, K. O., Wang, P., Lang, F., et al. (2024). Perovskite-organic tandem solar cells. Nat. Rev. Mater. **9**: 202–217. DOI: 10.1038/s41578-023-00642-1.
2. Wu, S., Yan, Y., Yin, J., et al. (2024). Redox mediator-stabilized wide-bandgap perovskites for monolithic perovskite-organic tandem solar cells. Nat. Energy **9**: 411–421. DOI: 10.1038/s41560-024-01451-8.
3. Chen, X., Jia, Z., Chen, Z., et al. (2020). Efficient and reproducible monolithic perovskite/organic tandem solar cells with low-loss interconnecting layers. Joule **4**: 1594–1606. DOI: 10.1016/j.joule.2020.06.006.
4. Chen, W., Zhu, Y., Xiu, J., et al. (2022). Monolithic perovskite/organic tandem solar cells with 23.6% efficiency enabled by reduced voltage losses and optimized interconnecting layer. Nat. Energy **7**: 229-237. DOI:10.1038/s41560-021-00966-8.
5. Zhang, Z., Chen, W., Jiang, X., et al. (2024). Suppression of phase segregation in wide-bandgap perovskites with thiocyanate ions for perovskite/organic tandems with 25.06% efficiency. Nat. Energy **9**: 592-601. DOI:10.1038/s41560-024-01491-0.

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DECLARATION OF INTERESTS

The authors declare no competing interests.