

Perovskite Solar Cells

Interface Molecular Locking Synergized with Self-Assembled Monolayers for Efficient Perovskite Solar Cells

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Abstract: The uniformity of self-assembled monolayers (SAMs) and the interfacial defects in perovskite films significantly affect the performance of inverted perovskite solar cells (IPSCs). Herein, we develop an innovative interface molecular locking (IML) strategy synergized with SAMs to enhance the properties of buried interface. Specifically, two SAMs—(4-(3,6-dimethoxy-9H-carbazol-9-yl)phenyl)phosphonic acid (MeO-PhPACz) and 5-indoleboronic acid (5-IBA)—are employed to combine their advantages and form an enhanced SAM (E-SAM). Due to the strong π - π interactions between MeO-PhPACz and 5-IBA, the E-SAM exhibits a denser and more uniform morphological coverage. Introducing thiabenzazole (TBZ) additive into the perovskite precursor further ameliorates the buried interface properties through its self-assembly behavior, owing to its large molecular dipole moment and strong interactions with the E-SAM. This strategy not only achieves favorable energy level alignment but also improves the crystallinity and reduces the trap density of perovskite films, thereby significantly enhancing hole extraction and suppressing non-radiative recombination. Consequently, both $(\text{FA}_{0.95}\text{MA}_{0.05})_{0.95}\text{Cs}_{0.05}\text{Pb}(\text{I}_{0.95}\text{Br}_{0.05})_3$ and $\text{FA}_{0.95}\text{Cs}_{0.05}\text{PbI}_3$ -based IPSCs achieve high efficiencies exceeding 26.0%, along with significantly enhanced stability. Notably, $(\text{FA}_{0.95}\text{MA}_{0.05})_{0.95}\text{Cs}_{0.05}\text{Pb}(\text{I}_{0.95}\text{Br}_{0.05})_3$ solar cells deliver a high voltage of 1.21 V, one of the highest reported among IPSCs with a 1.56 eV bandgap. Our findings provide unique insights into achieving high-performance IPSCs by synergistically engineering buried interface.

Introduction

Inverted perovskite solar cells (IPSCs) have emerged as promising candidates for scalable photovoltaic applications due to their high efficiency, low-temperature solution processability and compatibility with tandem architectures.^[1–4] However, achieving long-term operational stability remains a critical barrier to large-scale commercial deployment.^[5,6] For IPSCs, the key challenge lies in mitigating interfacial degradation issues, particularly at buried interface where non-radiative recombination and ion migration dominate performance losses.^[7] The hole-selective layers (HSLs), which simultaneously govern charge extraction kinetics, perovskite

crystallinity, and interfacial defect passivation,^[8,9] serves as a critical determinant of device efficiency and stability.

Self-assembled monolayers (SAMs) have revolutionized HSL design by enabling atomic-level control of interface.^[10,11] Structurally, SAMs integrate three essential components: anchoring groups—such as carboxylic acids (-COOH), sulfonic acids (-SO₃H), and phosphonic acids (-PO(OH)₂)—which provide robust bonding to metal oxide surfaces; linker groups that impart molecular flexibility; and terminal groups that govern electronic properties to fulfill their intended functions.^[12] The emergence of various novel SAMs has greatly promoted the efficiency breakthrough of IPSCs. However, conventional phosphonic acid-based SAMs face

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 Supporting Information section

inherent limitations: (i) Defect-prone deposition—surface roughness of ITO substrates and insufficient anchoring strength of phosphate headgroups lead to void formation, creating non-radiative recombination centers;^[13–15] (ii) Solubility constraints—amphiphilic SAM molecules tend to aggregate into micelles in alcoholic or polar solvents, restricting dense molecular packing;^[16–18] (iii) Interfacial instability—acidic phosphonic groups induce ITO corrosion, compromising device stability.^[19,20] Collectively, these factors exacerbate interfacial energy disorder and leakage current, disrupt charge transfer at the interface, intensify the non-radiative recombination, and ultimately reduce the photovoltaic performance.^[21–23]

Recent advances in co-adsorption strategies and post-assembly modifications have partially addressed these challenges. For instance, Yan et al. demonstrated that doping methyl-4-pyridinium phenyl (Me-4PACz) with choline chloride (PC) to form a co-SAM significantly enhances interfacial coverage and effectively reduces leakage current.^[24] Yan et al. incorporated 4-mercaptobenzoic acid (SBA) and 4-(trifluoromethyl) benzoic acid (TFA) as additives into MeO-2PACz to fabricate a co-SAM, which exhibits excellent energy level alignment with perovskite, thereby improving carrier extraction efficiency.^[25] Similarly, Zhou et al. employed 4-fluorobenzoic acid (FBA) as an additive to effectively fill the gaps between Me-4PACz molecules, forming a denser HSL that facilitates the passivation of the buried perovskite interface.^[26] Yu et al. developed a post-assembly chelating molecular bridge strategy using 5-(9H-carbazol-9-yl) isophthalic acid (CB-PA) to modify the buried perovskite interface. Specifically, CB-PA chemically couples with MeO-2PACz through π - π stacking between carbazole groups while forming bidentate Pb-binding chelates ($=O \cdots Pb$ bonds) with the perovskite, thereby constructing a bridging interface that enhances carrier extraction.^[27] Kyaw et al. incorporated 11-mercaptopundecylphosphonic acid (MPA) into the SAM system to construct a co-SAM, while simultaneously doping the perovskite precursor with an ionic liquid (IL) monomer. The accumulation of IL cations at the buried interface triggers an in situ polymerization reaction, forming a POL-AVM polymer at the perovskite/SAM interface, and achieving a high efficiency of 26.25%.^[12] Despite these advances, simultaneously achieving high open-circuit voltage (V_{OC}), fill factor (FF), and stability remains elusive due to unresolved interfacial energy level mismatches and persistent defect states.

In this work, we developed an innovative interface molecular locking (IML) strategy synergized with SAMs for IPSCs. This strategy synergistically combines a uniformly HSLs and a perovskite additive. First, an enhanced SAM (E-SAM) with uniform morphological coverage was achieved by combining the advantages of two SAMs: (4-(3,6-dimethoxy-9H-carbazol-9-yl)phenyl)phosphonic acid (MeO-PhPACz) and 5-indoleboronic acid (5-IBA). Meanwhile, thiabendazole (TBZ) was incorporated into the perovskite precursor. Driven by its functional groups ($-NH-$, $=N-$, and $-S-$), TBZ molecules coordinate with perovskite components and preferentially accumulate and lock at the buried interface due to their large dipole moment and strong interaction with E-

SAM, further improving the crystallinity and reducing trap density of the perovskite films. This strategy significantly strengthened interfacial adhesion, improved charge transport properties, and reduced interfacial non-radiative losses. Consequently, the champion devices achieved remarkable PCE of 26.00% and 26.14% for $(FA_{0.95}MA_{0.05})_{0.95}Cs_{0.05}Pb(I_{0.95}Br_{0.05})_3$ and $FA_{0.95}Cs_{0.05}PbI_3$ IPSCs, respectively. Furthermore, the unencapsulated devices exhibited exceptional long-term operational stability, retaining 87% of their initial efficiency after 1000 h of maximum power point tracking (MPPT) under the ISOS-L-1 protocol. The strategy also endows the devices with significantly enhanced ultraviolet (UV) and thermal stability.

Results and Discussion

The inherent amphiphilic nature and steric hindrance effects of SAMs predispose them to molecular aggregation, leading to interfacial inhomogeneity and the formation of adsorption vacancies on conductive substrates.^[28,29] Such imperfections induce residual stress in perovskites, significantly degrading photovoltaic performance.^[30] To this end, we developed a strategy that combines the advantages of two SAM components to achieve atomic-level interface engineering. First, the primary SAM (MeO-PhPACz), containing a phosphonic acid anchoring group, was deposited onto the ITO surface to ensure robust substrate attachment.^[31] Subsequently, 5-indoleboronic acid (5-IBA), featuring boronic acid anchoring groups, infiltrates interstitial voids in the primary SAM layer through π - π interactions, constructing a densely packed interface with high coverage. This can effectively improve the contact at the perovskite/HSL interface, and optimize the interfacial energy level alignment, thereby significantly enhancing hole extraction and reducing interfacial charge recombination.^[32] This enhanced SAM is referred to as E-SAM in the following discussion. As illustrated in Figure 1a, the molecular structures and electrostatic surface potentials of the two SAMs reveal distinct electronic characteristics, with the highest occupied molecular orbital/lowest unoccupied molecular orbital (HOMO/LUMO) levels and dipole moments further analyzed in Figure S1. Atomic force microscopy-based infrared spectroscopy (AFM-IR) measurements (Figure 1b) reveal that the E-SAM film exhibits a significantly enhanced and sharpened IR resonance at 1560 cm^{-1} , compared to 1400 cm^{-1} for the control. This resonance corresponds to the $C\equiv C$ stretching in aromatic rings. The intensification and shift of this peak indicate enhanced electron coupling between the aromatic rings of MeO-PhPACz and 5-IBA, a phenomenon commonly observed in strongly π - π stacked systems.^[33,34] Time-of-flight secondary ion mass spectrometry (TOF-SIMS) further indicates that 5-IBA and MeO-PhPACz form a blended state (Figure S2). We performed X-ray photoelectron spectroscopy (XPS) measurements on bare ITO and ITO coated with different SAMs, specifically monitoring the In 3d core-level signals originating from the ITO substrate. As shown in the Figure S3a, the intensity of the In 3d peaks is significantly attenuated in the E-SAM sample compared to both the bare ITO and

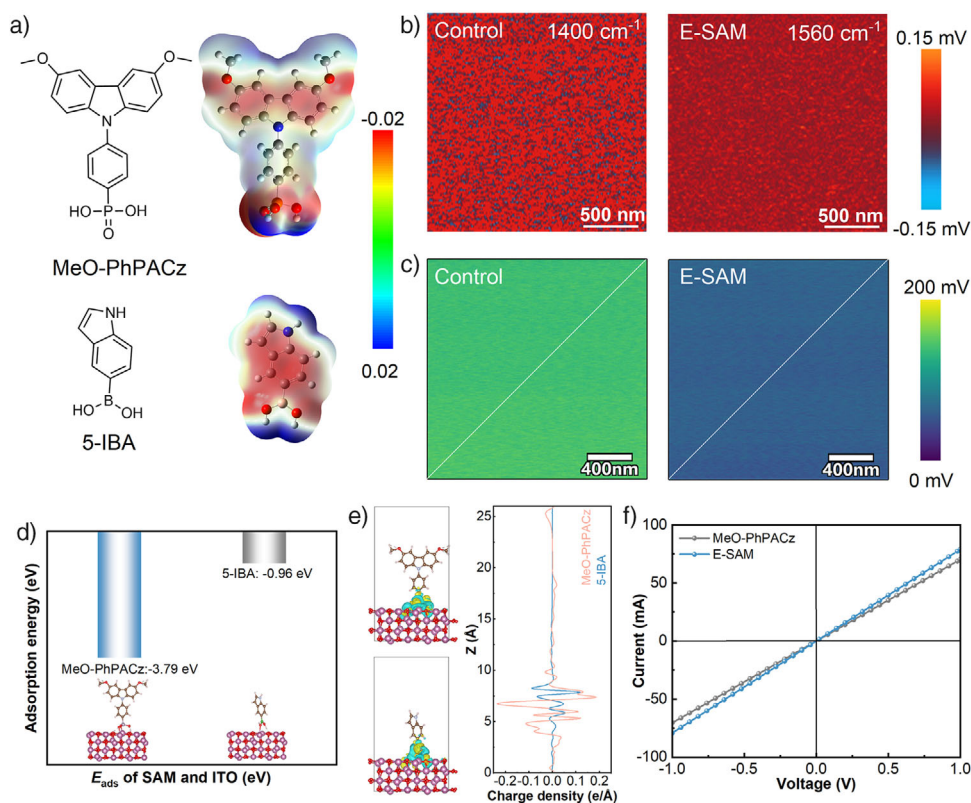


Figure 1. a) Molecular structures and electrostatic surface potential maps. b) AFM-IR images of the SAM films: the control film exhibits characteristic P–C stretching at 1400 cm⁻¹ (corresponding to the MeO-PhPACz); the E-SAM shows enhanced C=C aromatic resonance at 1560 cm⁻¹, indicating dense molecular packing. c) KPFM images of MeO-PhPACz and E-SAM films. d) Statistics on the E_{ads} of SAMs on ITO. e) Optimized configurations of MeO-PhPACz (top left) and 5-IBA (bottom left) on the ITO surface with differential charge density; corresponding charge integration curves along the z -axis (right). f) I - V curves of the ITO/HSLs/MoO_x/Au layer.

the single MeO-PhPACz. This attenuation provides direct evidence that the E-SAM forms a denser molecular layer, which more effectively shields the underlying ITO substrate from the X-ray probe, indicating superior surface coverage. In addition, we indirectly verified the coverage of SAM on ITO by measuring the peak area ratio of adsorbed OH groups and lattice O atoms (In–O and Sn–O) in the O 1s core energy level peak in XPS spectra. As shown in Figure S3b,c, the proportion of OH groups in E-SAM (39.63%) are significantly higher than that of MeO-PhPACz (30.15%), indicating a higher coverage of SAM.^[16] Kelvin probe force microscopy (KPFM) analysis (Figures 1c and S4) further confirmed that the E-SAM possessed a more uniform surface potential than the ITO coated solely with MeO-PhPACz, a characteristic favorable for hole transport.^[26]

The energy levels of the underlying SAMs were systematically investigated using ultraviolet photoelectron spectroscopy (UPS) (Figure S5). The work function (WF) of the E-SAM was measured at 4.88 eV, surpassing that of the MeO-PhPACz control sample (4.58 eV) (Figure S6). This elevated WF indicates the enhanced p -type character of the E-SAM, which is beneficial for improving hole extraction efficiency from the perovskite layer.^[35] Density functional theory (DFT) calculations (Figure 1d) quantified the binding energies between different SAMs and ITO. The adsorption energies (E_{ads}) of MeO-PhPACz and 5-IBA molecules on ITO

were determined to be -3.79 and -0.96 eV, respectively. The moderate E_{ads} of -0.96 eV for 5-IBA ensures stable anchoring without competitively displacing MeO-PhPACz, thereby facilitating its role as an interstitial filler. Meanwhile, 5-IBA contains an indole ring, an electron rich heterocyclic aromatic system with a favorable π -conjugated structure. This structure facilitates the delocalization of holes, thereby providing efficient charge transfer channels. Collectively, the strategically tailored E_{ads} of 5-IBA balances interfacial adhesion and charge transport efficiency. Differential charge density analysis (Figure 1e) revealed that the sharp charge accumulation at the 5-IBA/ITO interface was significantly reduced compared to MeO-PhPACz, indicating mitigated interfacial charge localization.^[27,36] To evaluate the electrical impact of 5-IBA post-assembly, we calculated the conductivity (σ) using the equation^[26]:

$$\sigma = \frac{d}{AR}$$

where d is the SAM layer thickness, A is the active device area, and R is the resistance derived from I - V curves. As shown in Figure 1f, the E-SAM-based device (ITO/MeO-PhPACz/5-IBA/MoO_x/Au) exhibited a conductivity of 9.67×10^{-4} mS cm⁻¹, which is 2.06 times higher than that of the single MeO-PhPACz layer (4.69×10^{-4}

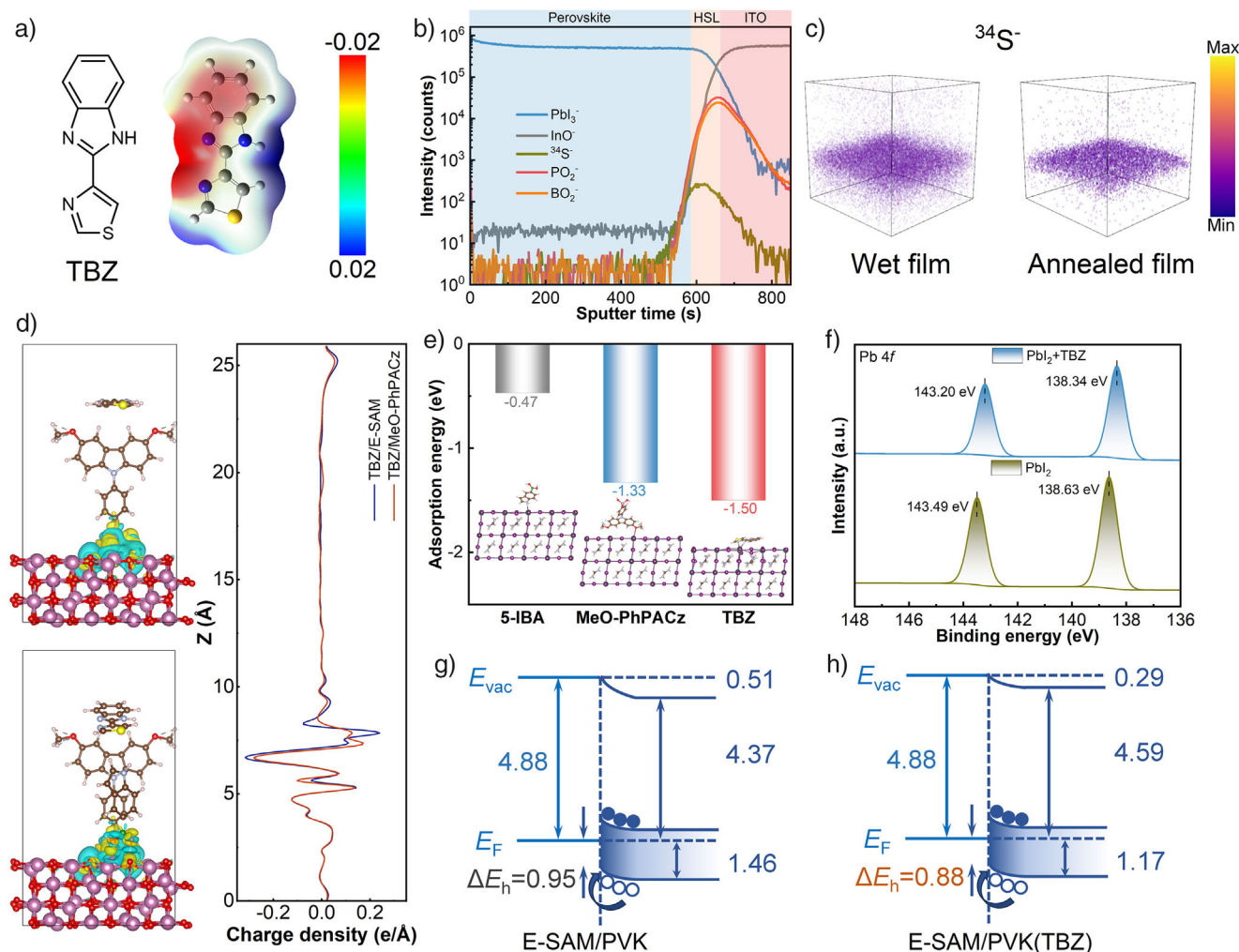


Figure 2. a) Molecular structure and electrostatic surface potential map of TBZ. b) TOF-SIMS analysis of the target film. c) Three-dimensional (3D) distribution of $^{34}\text{S}^-$ ions in perovskite films before and after thermal annealing. d) Differential charge density of TBZ on MeO-PhPACz (top) and E-SAM (bottom) in ITO surface. Blue isosurface indicates electron depletion, while yellow denotes electron accumulation. e) Schematic of adsorption configurations between the perovskite (001) plane and SAMs, with corresponding E_{ads} . f) XPS spectra of Pb 4f for PbI_2 and $\text{PbI}_2 + \text{TBZ}$ films. g) and h) Illustrations of the energy levels.

mS cm^{-1}). This significant enhancement underscores the role of the E-SAM in improving the overall charge transport property of the interface. The well-aligned energy level and enhanced conductivity facilitate efficient hole transfer and reduce charge accumulation at the HSL/perovskite interface, promoting higher open-circuit voltage (V_{OC}) and fill factor (FF) in PSCs. Consistently, E-SAM-based PSC devices achieved a PCE of 24.85%, surpassing the control devices with bare MeO-PhPACz HSL (23.91%) (Figures S7 and S8).

To further ameliorate the buried interface, where accommodates various defects including lead vacancies (V_{Pb}), iodine vacancies (V_{I}), and uncoordinated Pb^{2+} ions that can induce non-radiative recombination and instability issues,^[37–40] we doped the perovskite precursor with an additive, thiabenzazole (TBZ), which can self-assemble at the buried interface. The molecular structure and electrostatic surface potential of TBZ are shown in Figure 2a, and the HOMO/LUMO levels and dipole moments are presented in Figure S9. TBZ contains functional groups, such as $-\text{NH}-$, $=\text{N}-$, and $-\text{S}-$.

These groups can influence the crystallization process, thereby enhancing both the crystallinity and morphology of the target perovskite film.^[41] Additionally, they are expected to strongly coordinate with the defects, such as uncoordinated Pb^{2+} at the grain boundaries, significantly reducing defect density and suppressing non-radiative carrier recombination. These advantages can collectively contribute to high-quality perovskite films with exceptional optoelectronic properties.

TOF-SIMS revealed that only a minor fraction of TBZ resides at grain boundaries within the bulk film (Figures 2b,c, S10, and S11), while it is predominantly enriched at the perovskite buried interface. To elucidate the dynamic deposition mechanism, we performed in situ XPS analysis by tracking the S 2p peak on the surface of the wet (Figure S12a) and annealed (Figure S12b) perovskite films. These analyses corroborated that TBZ molecules initially distribute uniformly throughout the perovskite bulk and interfacial regions, then progressively migrate to the perovskite buried interface during solvent evaporation—a behavior likely driven by its

large molecular dipole moment (4.34 D).^[42] TBZ contains aromatic benzene and thiazole rings, which can undergo π – π interaction with the MeO-PhPACz layer, allowing TBZ to firmly adsorb onto it. The methoxy group at the MeO-PhPACz end exhibits electron donating ability and possesses a dipole moment. The heteroatom rings and functional groups within TBZ molecules also have their own dipoles, enabling dipole–dipole interactions that further stabilizing their relative positions. The nuclear magnetic resonance (NMR) measurement (Figure S13) for the mixture of MeO-PhPACz and TBZ revealed that the chemical shift of the -NH group in TBZ moved from 12.88 to 13.00 ppm, accompanied by a corresponding shift in the methoxy group signal of MeO-PhPACz. This suggests the occurrence of molecular interactions between TBZ and MeO-PhPACz, likely involving the formation of hydrogen bonds. The π – π , dipole–dipole, and hydrogen bond interactions collectively contribute to the stabilization of the molecular assembly at the interface. Importantly, differential charge density analysis (Figure 2d) indicates that TBZ enrichment at the SAM surface enhances π – π interactions while having a negligible impact on charge transport across the SAM/ITO interface. DFT calculations further quantified interfacial interactions (Figure 2e), the E_{ads} of TBZ on the FAPbI₃ (001) surface was calculated as –1.50 eV, surpassing those of MeO-PhPACz (–1.33 eV) and 5-IBA (–0.47 eV). Intermolecular E_{ads} values among the three components, MeO-PhPACz, 5-IBA, and TBZ, were systematically mapped (Figure S14). The robust dual interaction capability of TBZ with both perovskite and MeO-PhPACz indicates that TBZ can serve as an intermediary agent to strengthen interactions at the buried perovskite/SAM interface. Meanwhile, the functional groups of TBZ likely improve the crystallinity of the perovskite film and passivate interfacial defects. Collectively, these optimizations will enhance the interfacial stability of the PSC devices.

We performed XPS measurement to elucidate the interaction between perovskite and TBZ molecules.^[30] As shown in Figures 2f and S15, the Pb²⁺ 4f_{5/2} (143.49 eV) and 4f_{7/2} (138.63 eV) peaks in pure PbI₂ films shifted to 143.20 and 138.34 eV, respectively, upon TBZ incorporation. Similarly, the I 3d_{3/2} (630.79 eV) and 3d_{5/2} (619.33 eV) signals shifted to 630.85 eV and 619.37 eV. This interaction was also confirmed through nuclear magnetic resonance (NMR) (Figure S16). UV–vis absorption spectroscopy revealed that the incorporation of TBZ did not alter the perovskite bandgap (Figure S17). The UPS measurements (Figures S18 and S19) revealed a deeper work function in TBZ-modified perovskite films (–4.59 eV) compared to pristine films (–4.37 eV). This enhanced *p*-type character improves energy level alignment with E-SAM, thereby reducing the hole transport barrier. Band-bending analysis (Figure 2 g,h) demonstrated that, in the case of E-SAM/perovskite (Figure 2g), the perovskite band bends upward, which can promote hole extraction and block electrons; the offset ΔE_{h} is relatively high at 0.95 eV. For E-SAM/perovskite (TBZ) (Figure 2h), owing to its more pronounced *p*-type character and a deeper Fermi level, the perovskite band also bends upward, but ΔE_{h} is reduced to 0.88 eV, lowering the energy barrier and facilitating more efficient hole transport.^[33,43]

To investigate the charge carrier dynamics at the SAM/perovskite interface, transient absorption spectroscopy (TAS) measurements were performed.^[27,35] As shown in Figure 3a–c, two-dimensional TAS spectral maps reveal distinct exciton dynamics in perovskite films deposited on ITO/SAM substrates. From the decay kinetics fitting (Figure S20 and Table S1), the average decay lifetimes (τ_{ave}) were determined to be 2099 ps (control), 1838 ps (E-SAM), and 1183 ps (target). Here, τ_1 corresponds to the ultrafast exciton dissociation at the interface, while τ_2 represents the exciton diffusion toward the interface prior to dissociation. The amplitude ratio (A_1/A_2) reflects the relative contribution of these competing processes, with the shortest τ_{ave} in the target film confirming enhanced interfacial hole extraction efficiency.

The crystallinity and morphology of perovskite films were characterized by X-ray diffraction (XRD), grazing-incidence wide-angle X-ray scattering (GIWAXS), and scanning electron microscopy (SEM) measurements. The target perovskite film exhibits increased diffraction intensity, indicating enhanced crystallinity (Figure S21). GIWAXS measurements at 0.9° incidence (Figure 3d–f) revealed enhanced crystallinity in target perovskite films, evidenced by intensified (001) diffraction rings compared to control and E-SAM films. Quantitative orientation analysis using the normalized orientation parameter^[44,45]:

$$f_{\text{ori}} = \frac{A_{\text{ori}}}{A_{\text{ori}} + A_{\text{iso}}}$$

where A_{ori} and A_{iso} represent the integrated areas under the fitted Gaussian peaks corresponding to the oriented and isotropic crystallites, respectively, normalized by the total integrated area (Figure 3g). It is worth noting that the orientation degree of the target perovskite film was 50.24%, which was higher than the control (44.83%) and E-SAM (46.52%), indicating the preferential orientation of the (001) plane. As the incident angle increases from 0.3° to 1.2°, the diffraction peaks of the control perovskite film gradually shift towards smaller angles (Figure S22), which may be attributed to the presence of interfacial gaps or lattice relaxation. In contrast, the peaks of the target film demonstrate negligible shift, indicating effective release of residual strain. These results confirm that the IML strategy successfully mitigates the residual stress–strain in perovskite films caused by excessive lattice shrinkage. The grain size of the target perovskite film is larger than those of the control and E-SAM-based samples. The increased grain size signifies fewer grain boundaries. Most importantly, the SEM images of the buried interface demonstrate that there are remarkable pinholes in the control sample, possibly due to the inferior coverage and uniformity of the bare MeO-PhPACz HSL (Figure S23). The pinholes in the E-SAM-based sample are significantly reduced. In contrast, the target sample shows no observable vacancies at the buried interface, as a result of crystallization modulation achieved by TBZ incorporation. These results corroborate that the effectiveness of the synergistic interface engineering, which greatly improves the quality of the buried interface.

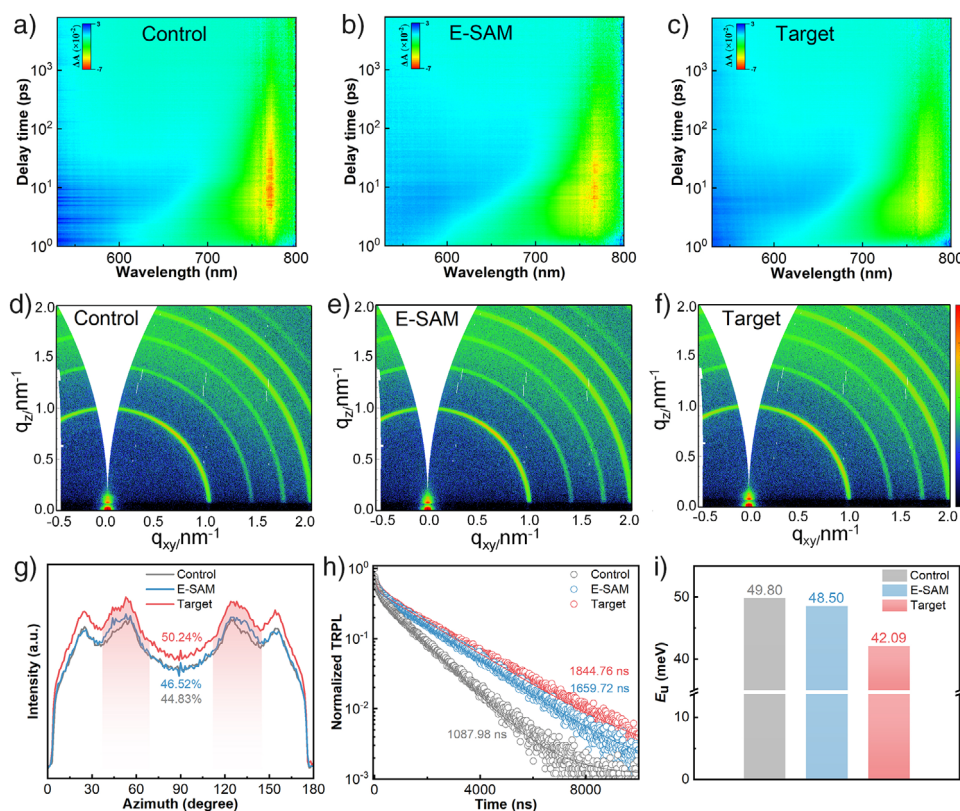


Figure 3. TAS pseudo-color maps of perovskite films on: a) ITO/MeO-PhPACz/PVK, b) ITO/E-SAM/PVK, and c) ITO/E-SAM/PVK(TBZ). d)–f) GIWAXS patterns of three perovskite films: d) Control, e) E-SAM, and f) Target. g) Corresponding azimuth integral curves of the (001) scattering ring extracted from GIWAXS results. h) TRPL decay curves of perovskite films deposited on SAMs. i) E_u values calculated for perovskites consisting of ITO/HSL/perovskite.

To further evaluate the non-radiative recombination behavior in the perovskite films, photoluminescence (PL) and time-resolved photoluminescence (TRPL) measurements were conducted on glass/SAM/perovskite samples. As shown in Figure S24, the annealed target film exhibited significantly stronger PL intensity, indicating a reduction in trap-assisted non-radiative recombination. Consistent with this observation, the TRPL spectra revealed a carrier lifetime of 1844.76 ns for the target film, which was substantially longer than that of the control film (1087.98 ns) and the E-SAM sample (1659.72 ns) (Figure 3h and Table S2). This prolonged carrier lifetime, attributed to the TBZ-induced (001) preferential orientation, effectively suppresses non-radiative recombination and enhances the overall optoelectronic quality of the perovskite.

Additionally, the Urbach energy (E_u), which reflects the width of the sub-bandgap absorption tail and serves as a key indicator of electronic disorder and defect density within the semiconductor, was measured to evaluate the quality of the perovskite films. A lower E_u value is generally associated with reduced energetic disorder, improved electronic quality, and diminished non-radiative recombination losses.^[46] As shown in Figures 3i and S25, the target film exhibited the lowest E_u (42.09 meV), indicating an improvement in perovskite film quality and a reduction in V_{OC} loss.

The IPSC devices with the structure ITO/SAM/perovskite/PCBM/BCP/Ag were fabricated to evaluate the effect of the IML strategy (Figure 4a). Figure 4b,c illustrate the schematic diagrams of the formation of E-SAM and the IML strategy, respectively, based on the aforementioned characterizations. The bare MeO-PhPACz SAM exhibits incomplete coverage on the ITO substrates, which can lead to direct contact between the perovskite films and the exposed ITO surface. In contrast, the 5-IBA SAM successfully compensates for these uncovered areas, forming an E-SAM with a denser and more uniform morphology due to the strong interactions between MeO-PhPACz and 5-IBA molecules. This eliminates pinhole-induced shunts and creates a low-resistance pathway for hole extraction, as confirmed by the enhanced stack conductivity (Figure 1f). This step is crucial for achieving high fill factor and current density. Furthermore, the large molecular dipole moment of TBZ, the strong interaction between the TBZ molecule and the E-SAM, and their coordination with the perovskite collectively lock them at the buried interface of the perovskite. This interaction further enhances the crystallinity of the perovskite films and improves the defect passivation effect at the buried interface. Current density-voltage ($J-V$) curves of the champion devices (Figure 4d) revealed a maximum PCE of 23.91% for the control device, with a V_{OC} of 1.165 V, a short-circuit current density (J_{SC}) of 25.10 mA cm⁻², and an FF of 82.71%. To verify the repeatability of device

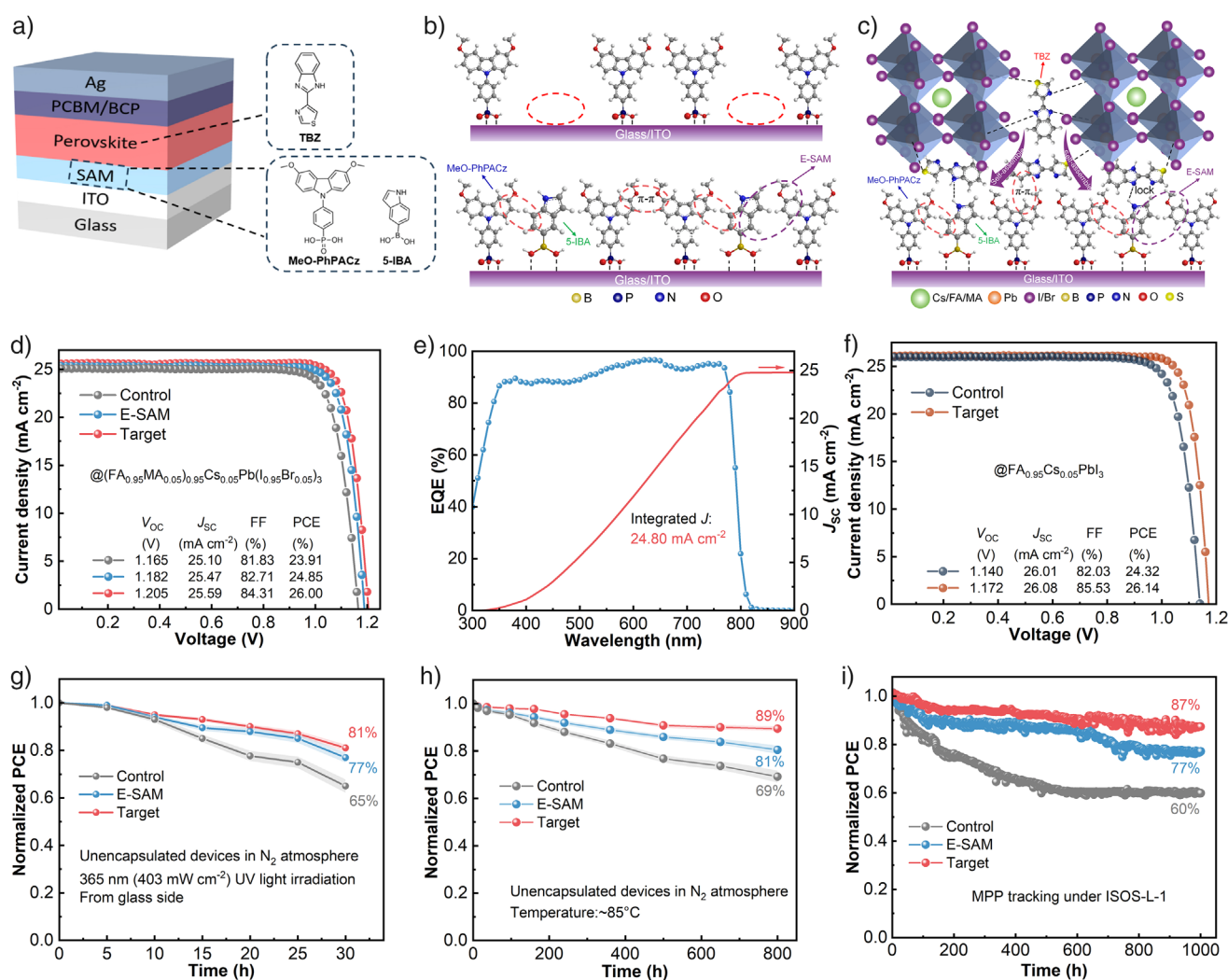


Figure 4. a) Device architecture of the IPSCs. Schematic diagram of E-SAM formation b) and IML strategy (c). d) $J-V$ curves. e) EQE spectra and integrated current density of target device. f) $J-V$ curves of FACs-based PSCs. g) UV-light stability tests of PSCs under 365 nm UV light (403 mW cm^{-2}) from the glass side in a N_2 atmosphere. h) Normalized PCE of PSCs aged in an 85°C N_2 glove box. i) Maximum power point tracking was performed for 1,000 h under continuous 1 sun illumination in N_2 environment.

efficiency, a batch of 20 control, E-SAM, and target devices were fabricated, and corresponding statistical data were plotted in Figure S26. The simultaneous improvement of V_{OC} and FF is the main reason for the PCE enhancement of the target device among the key photovoltaic parameters. We introduced TBZ molecules as an additive into the perovskite precursor and combined them with MeO-PhPACz layer, achieving a PCE of 25.32% (Figure S27). Furthermore, when only 5-IBA was employed as the HSL, the PCEs of devices fabricated with and without the TBZ additive were 23.20% and 18.75% (Figures S28 and S29), respectively. Finally, by combining E-SAM and optimizing the concentration of TBZ (Figure S30 and Table S3), the target device based on $(\text{FA}_{0.95}\text{MA}_{0.05})_{0.95}\text{Cs}_{0.05}\text{Pb}(\text{I}_{0.95}\text{Br}_{0.05})_3$ perovskite achieved a PCE of 26.00% ($V_{OC} = 1.205 \text{ V}$, $J_{SC} = 25.59 \text{ mA cm}^{-2}$, $\text{FF} = 84.31\%$), surpassing the E-SAM-based device (24.85%). The composition of the E-SAM was rigorously optimized by systematically varying the concentration of the secondary SAM, 5-IBA. As summarized in Figure S31, the device

performance exhibited a pronounced dependence on the 5-IBA concentration, with an optimum at 0.3 mg mL^{-1} . The typical Gaussian distribution trend underscores the synergistic nature of the E-SAM: an insufficient amount of 5-IBA leaves voids in the monolayer, while an excess may lead to unfavorable molecular packing. The optimal concentration yields the highest PCE, V_{OC} , and FF, confirming that achieving a precise molecular ratio is essential to maximizing the interfacial benefits. In addition, we also examined the mixture of MeO-PhPACz and 5-IBA (Figure S32), finding that the V_{OC} and PCE of the device fabricated with this blend were lower than those fabricated using the IML strategy. This may be due to the crystallization interference between the two materials during solution spin coating, which prevented the formation of a perfectly dense HSL. In addition, we also performed spin coating of the TBZ layer on E-SAM (Figure S33), where the champion efficiency of the device (25.69%) was lower than that achieved with the IML strategy (26.00%). Notably, the target device

exhibited significantly reduced hysteresis compared to the controls (Figure S34; Table S4). The integrated J_{SC} extracted from the external quantum efficiency (EQE) spectrum aligns well with the J - V measurement (Figure 4e). Moreover, this strategy was successfully extended to $\text{FA}_{0.95}\text{Cs}_{0.05}\text{PbI}_3$ -based PSCs (Figure 4f), boosting the PCE from 24.32% to 26.14% (certified steady-state efficiency of 25.65%, Figure S35). And in the 1.78 eV wide-bandgap PSCs, the device efficiency increased from 17.78% to 20.18% (Figure S36).

The stability of perovskite devices was systematically evaluated through three accelerated degradation protocols. Due to the excellent ultraviolet (UV) stability of MeO-PhPACz,^[31] the devices were exposed to intense UV radiation continuously in a nitrogen (N_2) environment at room temperature. As shown in Figure 4g, after 30 h of prolonged UV irradiation, the PCE of the target device remained at approximately 81% of its initial value. In contrast, the E-SAM and control devices retained only about 77% and 65%, respectively. Additionally, the thermal stability of unencapsulated devices was evaluated at 85 °C in a N_2 glove box. After 800 h of continuous heating, the target device retained 89% of its initial PCE, significantly outperforming the E-SAM (81%) and control device (69%) (Figure 4h). The operational stability of unencapsulated devices was monitored for 1000 h under LED illumination (100 mW cm^{-2}) in an N_2 glove box. As shown in Figure 4i, the target device maintained approximately 87% of its initial PCE, outperforming the E-SAM (77%) and control device (60%). These results quantitatively validate the multi-faceted stability enhancement provided by IML strategy, achieving synergistic improvements in UV, thermal, and operational stability.

To probe interfacial charge transfer dynamics at the perovskite/HSL interface, the relationship between V_{OC} and light intensity was investigated (Figure 5a).^[47] A smaller slope in the V_{OC} versus light intensity plot indicates to a lower probability of trap-assisted recombination. The target device exhibited a slope of $1.21 K_B T q^{-1}$, which is lower than those of the control ($1.45 K_B T q^{-1}$) and E-SAM ($1.26 K_B T q^{-1}$) devices, indicating effective suppression of trap-assisted bimolecular recombination in the target device.^[48] In addition, carrier transport dynamics were further investigated through transient photovoltage (TPV) and transient photocurrent (TPC) decay measurement. As shown in Figure 5b, the target device exhibited an extended TPV carrier lifetime, consistent with the inhibition of non-radiative recombination at high V_{OC} . The TPC results revealed that the target device achieved the shortest charge extraction time of 0.75 μs (Figure 5c), indicating significantly accelerated interfacial charge extraction. The electrical characteristics of the devices were examined through electrochemical impedance spectroscopy (EIS) (Figure 5d). The recombination resistance (R_{rec}) values for the control, E-SAM, and target devices were calculated as 1297, 2139, and 3662 Ω , respectively. Concurrently, the series resistance (R_s) exhibited a decreasing trend. The marked increase in R_{rec} , coupled with the reduced R_s , contributed to suppressed charge recombination and enhanced carrier collection efficiency. Consequently, these improvements directly led to significant improvements in both V_{OC} and FF observed in the target device.

Furthermore, the investigation of FF loss relative to the Shockley–Queisser (S–Q) limit indicates a combination of non-radiative recombination and charge transport losses, as shown in Figure 5e. The maximum achievable FF (FF_{max}) under conditions that neglect charge transport losses is derived from the following equation^[49,50]:

$$FF_{max} = \frac{v_{OC} - \ln(v_{OC} + 0.72)}{v_{OC} + 1}$$

$$v_{OC} = \frac{V_{OC}}{nK_B T/q}$$

The maximum achievable FF (FF_{max}) values in the absence of charge transport loss were calculated to be 85.18% for MeO-PhPACz, 86.85% for E-SAM, and 87.49% for the target devices. By comparing the measured FF and FF_{max} to the S–Q theoretical limit, it was demonstrated that the target devices exhibit minimized non-radiative recombination and reduced charge transport losses. This improvement can be attributed to excellent interfacial contact with the perovskite layer and superior film quality. The photoluminescence quantum yield (PLQY) measurement was employed to evaluate the extent of non-radiative recombination in perovskite films deposited on SAM substrates. The target perovskite films showed a significantly higher PLQY of 2.96%, compared to 1.99% for E-SAM and 1.06% for the control films (Figure 5f). This increase in PLQY reflects a reduction in non-radiative recombination channels and an enhancement in radiative recombination. Consequently, the non-radiative V_{OC} loss was substantially reduced from 118 to 91 mV (Figure S37). Additionally, the quasi-Fermi level splitting (QFLS) was derived from the PLQY measurements, yielding values of 1.183 V for the control, 1.199 V for E-SAM, and 1.210 V for the target perovskite. The increased QFLS in the target device underscores optimized energy level alignment and reduced internal defects, thereby contributing to an improved V_{OC} .^[51] Statistical analysis of the FAMACs-based devices (Figure 5g and Table S5) further confirmed that the target architecture achieves one of the highest V_{OC} values reported for this bandgap range (1.55–1.58 eV). The dark current J - V characteristics of the devices (Figure 5h) demonstrated that the target device exhibits markedly lower leakage current, more efficient hole transfer, and superior diode behavior in the voltage range of ± 2 to 0 V, consistent with enhanced interfacial quality and reduced defect-mediated recombination.

To analyze the trap-state density (N_{trap}) in different perovskite films, the space-charge limited current (SCLC) measurements were performed on the hole-only devices with the structure ITO/SAM/PVK/MoO₃/Ag. The N_{trap} was calculated using the following formula^[27,29]:

$$N_{trap} = \frac{2\epsilon_r\epsilon_0 V_{TFL}}{qL^2}$$

As shown in Figure 5i, the trap-filled limit voltage (V_{TFL}) for the control, E-SAM and target devices was measured to be 0.21, 0.18, and 0.14 V, respectively. The corresponding N_{trap} values were calculated as 1.77×10^{15} , 1.39×10^{15}

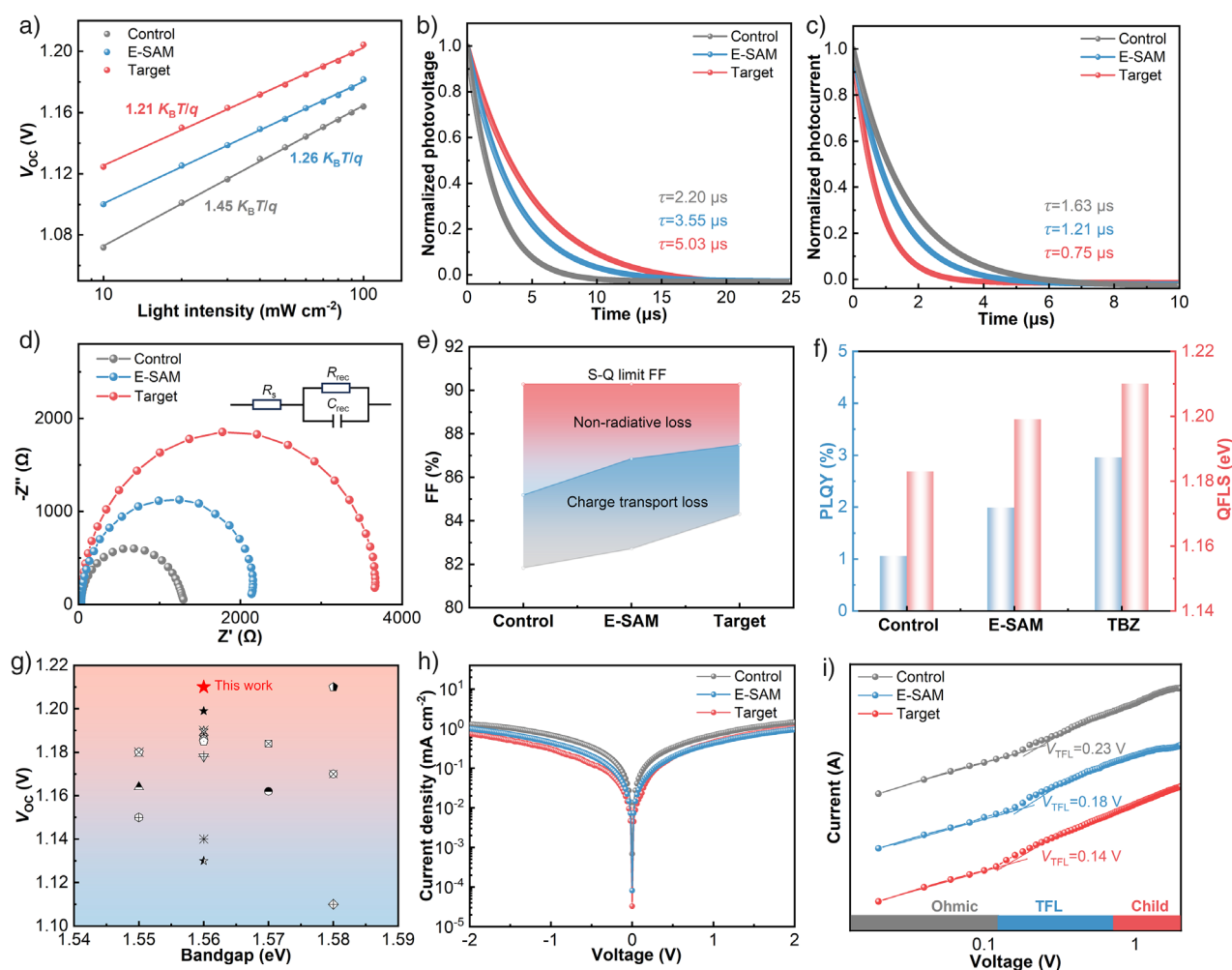


Figure 5. a) Light intensity dependent V_{OC} test. b) TPV and c) TPC measurements. d) Nyquist plots of the devices. e) FF loss analysis, analyzing the impact of non-radiative and charge transfer losses. f) PLQY and corresponding QFLS. g) Statistical V_{OC} distributions of the target FAMACs-based devices. h) Dark $J-V$ and i) $I-V$ curves of the hole-only devices.

and $1.08 \times 10^{15} \text{ cm}^{-3}$, respectively. These results clearly indicate a reduction in defect density within the target device, confirming the effective suppression of trap-assisted recombination.

Conclusions

In summary, we developed an innovative IML strategy that significantly enhances hole extraction and suppresses non-radiative recombination in IPSCs. This strategy utilizes MeO-PhPACz and 5-IBA to form a uniform E-SAM with high coverage on the ITO substrates. Simultaneously, the incorporation of TBZ additive into the perovskite precursor promotes its accumulation and lock at the buried interface, owing to its large inherent dipole moment and strong interactions with the E-SAM. This further modifies the perovskite buried interface and reduces defect density. Consequently, champion devices employing the IML strategy achieved PCE of 26.00% and 26.14% for FAMACs- and FACs-based IPSCs,

respectively. Moreover, the devices demonstrated exceptional operational stability, retaining 87% of their initial efficiency after 1000 h of continuous illumination under the ISOS-L-1 protocol. Our findings offer valuable insights into achieving high-performance IPSCs by synergistically engineering the buried interface.

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Conflict of Interests

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords: Buried interface • Efficiency • Inverted perovskite solar cells • Molecular locking • Self-assembled monolayers

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