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Hybrid vapor–solution processing and evaporated dual-interface engineering enable FAPbI₃-based solar cells with thermal stability beyond 10 000 hours

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The hybrid evaporation/solution-coating method holds great promise for perovskite manufacturing and textured tandem architectures. Nevertheless, it has predominantly been applied to wide-bandgap (>1.65 eV) perovskites, while FAPbI₃-based compositions remain underexplored. Here, we establish a hybrid vapor–solution FAPbI₃-based perovskite platform and identify key roles of precursor composition and annealing conditions in perovskite formation. Building on this platform, an evaporated dual-interface CsBr engineering strategy is demonstrated, combining precise interfacial modification with enhanced device performance and thermal stability. Ultrathin CsBr layers deposited by thermal evaporation at both the bottom and top interfaces effectively hinder defect formation during thermal aging. This approach yields p–i–n perovskite solar cells based on sputtered NiO_x with power conversion efficiencies exceeding 23%. Most notably, devices retain over 88% of their initial efficiency after more than 10 400 h of thermal aging at 85 °C under ISOS-D-2I (dark, inert atmosphere) conditions.

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1. Introduction

Perovskite solar cells (PSCs) have recently achieved power conversion efficiencies (PCEs) of 28.0% for single-junction cells and 35.2% in tandem configuration with crystalline silicon.¹ The most commonly used perovskite deposition technique to achieve such high efficiencies in the laboratory is spin coating. An alternative, less explored method is the hybrid vapor–solution deposition, where inorganic halides are first deposited by thermal (co-)evaporation, followed by solution coating of organic halides in alcohol solvents such as ethanol or isopropanol. This hybrid deposition method presents advantages

over fully solution-based approaches, as vacuum-based deposition enables precise and uniform control over the inorganic layer thickness and morphology across large areas, which may facilitate improved control over the perovskite conversion and crystallization. In addition, the hybrid method avoids the use of toxic solvents and the need for a quenching step, features that may be advantageous for large-scale manufacturing. Even though spin coating has been commonly used as the second step in the vapor–solution method, the fully scalable route evaporation/blade-coating has been demonstrated through adequate control of the coating dynamics.^{2–6} Moreover, this technique has been demonstrated to conformally deposit perovskite films on top of substrates with different roughness, such as random pyramid silicon texturing, making it highly attractive for multijunction applications.^{7–10} For this reason, the hybrid vapor–solution method has been predominantly applied for compositions with bandgaps greater than 1.65 eV, mainly intended for their application in perovskite-silicon tandem devices. However, extending this method to perovskites with lower bandgaps (~1.49–1.58 eV) would be greatly relevant for single-junction cells,¹¹ bifacial perovskite-silicon tandem cells,¹² and for middle cells in all-perovskite and perovskite-perovskite-silicon triple-junction architectures.^{13,14}

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At present, two-step hybrid processes have reported single-junction PCE records of 24.3% (certified 23.9%) for an n-i-p device¹¹ and 22.64% for a p-i-n device.¹⁵ It has been demonstrated that the interfaces between the perovskite and the charge transport layers are major contributors to performance losses and instability in PSCs.¹⁶ In this context, the two-step hybrid method differs from conventional spin-coating by involving a heterogeneous solid-liquid reaction pathway that gives rise to distinct surface compositions and buried interfaces. Consequently, passivation strategies developed for solution-processed perovskites may not be directly transferable and may require adaptation to the specific characteristics of the hybrid-processed system.¹⁰

On the one hand, at the hole transporting layer (HTL)/perovskite interface, several passivation and interface-engineering strategies have been developed in PSCs to improve energy level alignment, regulate the perovskite crystallization, and reduce interfacial defects.^{17–19} One clear example is the nickel oxide (NiO_x)/perovskite interface, for which strategies, such as Lewis acid–base organic salts,²⁰ dye molecules,²¹ alkali salts,^{22,23} and self-assembled monolayers,^{24–28} have been employed to passivate defects and impede redox reactions with perovskite cations.^{29–31} For perovskite films prepared *via* vapor-solution processing, inorganic interlayers such as CsBr^{5,32} or CsI² have been incorporated prior to the deposition of either PbI₂ or co-evaporated PbI₂:CsBr. Zhang *et al.* reported that CsBr interlayers can simultaneously modify the NiO_x surface and influence the PbI₂ crystal growth orientation.⁵ In addition, CsBr may react with residual PbI₂ at the bottom of the perovskite layer, forming a vertical composition gradient that enhances energy-level alignment with the HTL.³² On the other hand, treating the perovskite/electron transport layer (ETL) interface is also critically important. It is well known that the direct contact of perovskite with C₆₀ results in the formation of deep trap states causing significant non-radiative recombination.³³ In addition, other defects at the perovskite surface and grain boundaries, such as halide vacancies, Pb²⁺ dangling bonds, and unreacted PbI₂, compromise both photovoltaic performance and device stability.^{34,35} Importantly, most surface modification research so far has relied on spin coating organic-salt based passivation agents, significantly improving device performance, but having limited potential for large-area devices.^{35–40} Thermal evaporation offers a potentially scalable alternative for the uniform deposition of interfacial modification layers with precise thickness control at the sub-nanometer scale. Inorganic salts, such as caesium halides, are particularly suitable for this approach due to their sufficient volatility, thermal stability, and clean evaporation behavior without decomposition, enabling reproducible and uniform interfacial treatment.^{41,42} However, the implementation of fully evaporated interfacial treatments in hybrid-processed perovskite systems remains comparatively unexplored, particularly for FAPbI₃-based devices.

In this work, we establish a hybrid vapor-solution FAPbI₃-based platform and identify key factors governing the solid-liquid perovskite formation process. Building on this platform, the integration of evaporated CsBr interfacial layers addresses degradation-relevant features associated with the hybrid film

formation process, enabling improved device performance, with PCE values exceeding 23%, and enhanced thermal stability under accelerated aging conditions, with devices maintaining over 88% of their initial efficiency after more than 10 400 h at 85 °C in an inert atmosphere and in the dark.

2. Results and discussion

2.1. Establishing a hybrid FAPbI₃-based perovskite platform

Fig. 1a illustrates the three steps that compose the hybrid process used in this work to form perovskite films. First, a lead iodide (PbI₂) seed layer is thermally evaporated on indium tin oxide (ITO)-coated glass substrates pre-coated with an HTL. Unless stated otherwise, the HTL consists of a bilayer of sputtered NiO_x modified with [4-(3,6-dimethyl-9H-carbazol-9-yl) butyl]phosphonic acid (Me-4PACz). In the second step, formamidinium iodide (FAI) and other organic precursors are dissolved in ethanol and spin-coated onto the seed layer. Lastly, the films are annealed in ambient air (45 ± 5% relative humidity) for precursor reaction, solvent evaporation, and perovskite crystallization and grain growth.

Functional additives have been widely used to promote the formation and stabilization of α -FAPbI₃ by modifying precursor coordination and crystallization pathways.^{43–46} In particular, methylammonium chloride (MACl) has been extensively employed in solution-processed systems, including one- and two-step deposition approaches.^{44,47,48} In the context of the hybrid method, its effects have been contrasted with other additives, such as urea and formamidinium chloride (FACl),⁴⁹ and methylammonium thiocyanate (MASCN),⁵⁰ in double-cation (CsFA), double-anion (IBr) perovskites.

In this work, we investigated the role of methylammonium iodide (MAI) and MACl as additives in the organohalide solution. Perovskite films were prepared from three different organic precursor solutions: (i) FAI, (ii) FAI + MAI, and (iii) FAI + MAI + MACl. Because additives can alter crystallization pathways and thereby shift perovskite formation temperatures,^{9,48,49} the films prepared with the different precursor solutions were annealed at either 130 °C or 150 °C for 20 minutes. As shown in Fig. S1, photographs taken after spin coating in an inert atmosphere, and before annealing in air show that the MACl-containing samples underwent a visible color change from the yellow hue of PbI₂ to brown, contrasting with the FAI-only and FAI + MAI cases. This color change might indicate that MACl could promote the formation of α -FAPbI₃ even at room temperature. After annealing either at 130 °C or 150 °C, all films exhibited the expected black appearance.

X-ray diffraction (XRD) analysis revealed important differences in crystallinity and phase composition. At 130 °C, the FAI-only sample exhibits a strong (001) PbI₂ peak at 12.68°, indicating incomplete conversion of the evaporated PbI₂ film into α -FAPbI₃ (Fig. 1b). Addition of MAI (7 mol%) moderately reduced the residual (001) PbI₂ signal, whereas the combination of MAI (7 mol%) and MACl (25 mol%) resulted in a substantially reduced residual PbI₂ signal, together with enhanced perovskite crystallinity. Photoluminescence (PL) measurements (Fig. 1d and S2a) are consistent with these observations, showing higher

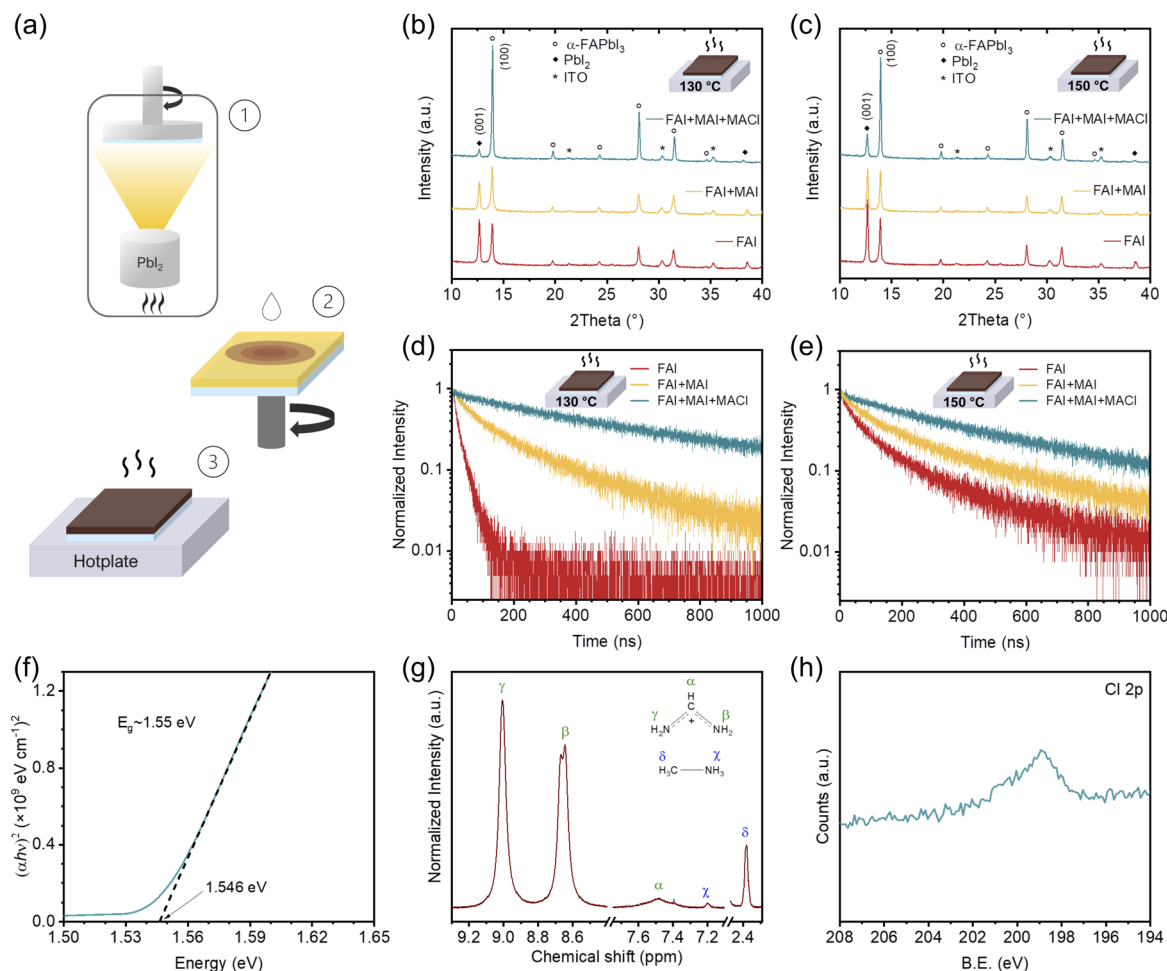


Fig. 1 Impact of methylammonium halides on perovskite conversion. (a) Schematic of the hybrid-deposition process. (b and c) XRD diffractograms, and (d and e) time-resolved PL decay of perovskite films, on glass/ITO/HTL, annealed for 20 minutes at 130 °C (b and d) and 150 °C (c and e), using FAI, FAI + MAI, or FAI + MAI + MACl in the second step ②. (f) Tauc plot, (g) ¹H-NMR spectrum, and (h) XPS spectra of the Cl 2p core level of perovskite film made with FAI + MAI + MACl organohalide solution and annealed at 130 °C for 20 min.

PL intensity and longer carrier lifetimes for the FAI + MAI + MACl films. At 150 °C, the same organohalide solution-dependent trend was observed, but residual PbI_2 signals are more pronounced across the different compositions (Fig. 1c). Interestingly, the higher annealing temperature of 150 °C has a favourable effect on the PL response of the additive-free FAI film but a negative one on the FAI + MAI + MACl sample (Fig. 1e and S2b). This reduced PL response and the more intense PbI_2 XRD signals suggest that excess thermal energy may drive the volatilization of MA- and Cl-containing species, hindering further conversion or leading to PbI_2 reformation.^{3,47,48,51} Solar cell measurements (fabrication details in SI) were consistent with the PL and XRD trends (Fig. S3).

PL analysis shows that, at 130 °C, the PL peak maxima of the FAI + MAI and FAI + MAI + MACl samples are blue-shifted by 3 nm and 10 nm, respectively, relative to the FAI-only sample (Fig. S2a). At 150 °C, the corresponding blue shifts are 5 nm and 8 nm, respectively (Fig. S2b). XRD measurements also revealed a small shift of the (100) α -FAPbI₃ peak to higher 2θ for the FAI + MAI + MACl samples (Fig. S4); however, this observation is not

used as standalone evidence for MA⁺ or Cl[−] incorporation into the perovskite lattice. Tauc plot analysis, shown in Fig. 1f, gives a bandgap of $\sim 1.55 \text{ eV}$ for the FAI + MAI + MACl sample annealed at 130 °C, consistent with the expected effect of partial MA⁺/Cl[−] incorporation.^{48,52} Proton nuclear magnetic resonance (¹H-NMR) spectroscopy provides direct evidence of partial incorporation of MA⁺, with an estimated 7% MA⁺ fraction in the final perovskite film (Fig. 1g and Table S1). In this work, the absorber is referred to as FAPbI₃-based to distinguish the resulting composition from a purely FA-based composition. Finally, X-ray Photoelectron Spectroscopy (XPS) revealed a faint but distinguishable Cl 2p signal peaking at 198.8 eV (Fig. 1h), within the reported Cl 2p_{3/2} binding energy range for metal chlorides (~ 198.5 – 199 eV),⁵³ indicating that not all chloride volatilizes during annealing.

To gain a deeper understanding of the conversion process and the origin of the residual PbI_2 phase observed in XRD measurements, we investigated the effects of organohalide solution concentration, PbI_2 morphology, and annealing time. In this hybrid process, sufficient interaction between the

thermally evaporated PbI_2 layer and the subsequently deposited organohalide solution is essential to achieve a uniform perovskite film. While functional additives facilitate this interaction, successful film formation ultimately requires fine-tuning of the processing conditions across all three stages: evaporation, spin coating, and annealing.

One key factor influencing precursor interdiffusion is the morphology of the evaporated PbI_2 layer, particularly its compactness and crystal orientation.^{5,54–57} The way the evaporated PbI_2 film grows is strongly affected by the underlying substrate. On glass/ITO/ NiO_x /Me-4PACz substrates, scanning electron microscopy (SEM) imaging showed that PbI_2 forms sheet-like structures with both in-plane and out-of-plane orientations (Fig. 2c). Despite being the immediate growth layer, the ultrathin NiO_x /Me-4PACz bilayer ($\sim 15/1$ nm) is not the main driver for the PbI_2 morphology. On glass/ NiO_x /Me-4PACz substrates, the resulting PbI_2 film becomes more compact and exhibits fewer out-of-plane structures than when ITO is present (Fig. S5). These differences in precursor morphology are reflected in the subsequent conversion behavior.

Given a fixed PbI_2 film thickness and morphology, the extent of conversion can be tuned by varying the concentration of the organohalide solution. Films prepared for this series were annealed at 130 °C for 20 min. As shown in Fig. 2a and S6, increasing precursor concentration, while keeping the molar fractions of MAI and MACl constant, reduced the residual PbI_2 signal and intensified the perovskite signals, indicating a more complete conversion of the evaporated PbI_2 layer. However, higher concentrations also promoted the formation of

disordered, mesh-like structures (Fig. S7), which were accompanied by reduced open-circuit voltage (V_{OC}) and overall device performance (Fig. 2d and S8). Interestingly, the lowest precursor concentration yielded the highest V_{OC} values despite the presence of more PbI_2 , which may contribute to the passivation of grain-boundary and surface defects.^{58–62} Together, these observations identified an intermediate precursor-concentration window that balances PbI_2 conversion and perovskite film morphology. Additional experiments using denser PbI_2 films resulted in lower conversion levels (Fig. S9), consistent with reduced solution infiltration and precursor interdiffusion.

We next investigated the effect of annealing time on PbI_2 conversion and film morphology using a FAI concentration of 0.522 M and an annealing temperature of 130 °C. As shown in Fig. 2b and S10, shorter annealing times resulted in lower residual PbI_2 signals and smaller PbI_2 domains, suggesting that most of the conversion occurs within the first 10 minutes of annealing. Extended annealing resulted in increased residual PbI_2 , consistent with volatilization of MA- and Cl-containing species and subsequent PbI_2 reformation, as previously reported.^{3,47,48,51} The corresponding device data (Fig. 2e) shows that films annealed for 10 minutes yielded the highest average efficiency and V_{OC} , indicating that prolonged annealing provides no further benefit in conversion and may instead promote degradation or defect formation.

Based on these observations, the organohalide solution concentration was reduced to 0.505 M FAI to improve the balance between conversion and film morphology. The

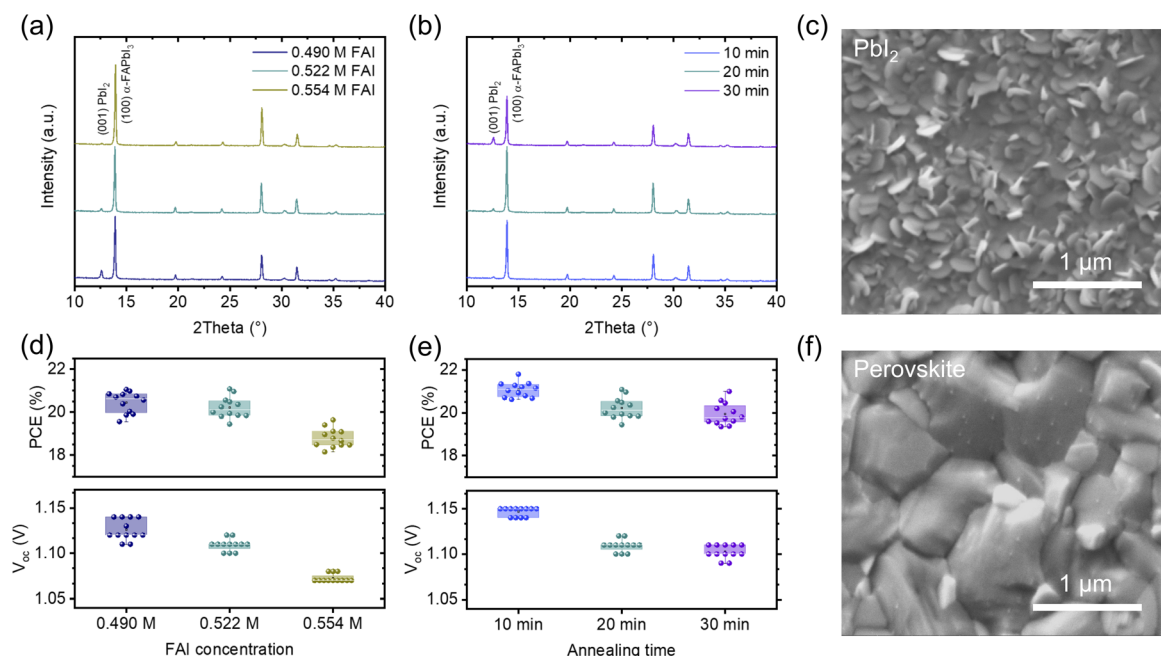


Fig. 2 Impact of FAI + MAI + MACl organohalide solution concentration and annealing time on perovskite conversion. (a and b) XRD diffractograms of absorbers with a variation in organohalide solution concentration (a), and with a variation in annealing time (b). (c) SEM image of PbI_2 layer grown on glass/ITO/ NiO_x /Me-4PACz. (d and e) Photovoltaic parameters of perovskite solar cells with a variation in solution concentration (d), and with a variation in annealing time (e). (f) SEM image of a perovskite film on glass/ITO/ NiO_x /Me-4PACz grown using 0.505 M FAI and 10 min annealing at 130 °C. SEM images were acquired with a secondary electron (SE) Everhart–Thornley detector.

resulting films no longer exhibited the disordered structures observed at higher concentrations. Fig. 2f shows the morphology of such perovskite films, annealed for 10 minutes at 130 °C, with domain sizes around 1 μm and smaller grains ($\sim 0.2\text{--}0.3\ \mu\text{m}$) suspected to be PbI_2 , as supported by back-scattered electron imaging (Fig. S11).

Using the processing conditions established above, p-i-n devices were fabricated with the architecture depicted in Fig. 3a, consisting of glass/ITO/ NiO_x /Me-4PACz/perovskite/LiF/ C_{60} /BCP/Ag. A MgF_2 antireflection coating was applied to the light-incident glass surface for the devices discussed below unless otherwise specified. The resulting champion device achieved a PCE of 22.30%, with J_{SC} of $24.1\ \text{mA cm}^{-2}$, V_{OC} of 1.15 V, and FF of 80.60%. Fig. 3b shows the characteristics of the current density–voltage (JV) curves of the champion device ($0.125\ \text{cm}^2$). The external quantum efficiency (EQE) spectrum of the device is shown in Fig. 3c. The integrated photocurrent with $24.1\ \text{mA cm}^{-2}$ matches well with the value obtained from the JV measurement. Furthermore, perovskite minimodules with an aperture area of $4\ \text{cm}^2$ were fabricated on $9\ \text{cm}^2$ glass substrates using the same layer stack. The schematic diagram of the monolithic series connection between cells in these modules and the measurement of the loss area are illustrated in Fig. S12. As shown in Fig. 3d, the champion minimodule device reached an aperture-area PCE of 19.86%.

2.2. Evaporated dual-interface CsBr engineering

Building on the established hybrid-processed FAPbI_3 -based platform, we next investigated whether targeted interfacial engineering could improve device performance and stability. In this work, ultrathin thermally evaporated CsBr layers were introduced at both the buried perovskite/HTL interface and the top perovskite surface. These samples are labelled as “Dual-CsBr”, while samples with CsBr only at the buried or top interface are called “Bottom-CsBr” and “Top-CsBr”, respectively. A systematic thickness-dependent study of CsBr layers (Fig. S13 and S14) showed that optimal device performance is obtained for ultrathin layers ($\sim 5\ \text{nm}$ at the buried interface and $\sim 2.5\ \text{nm}$ at the top interface), whereas further increasing the CsBr thickness leads to reduced V_{OC} , FF, or both. These results indicate that the beneficial effect of CsBr is retained only within an ultrathin thickness range, while thicker layers adversely affect device performance. The 5 nm CsBr seed layer did not significantly affect the morphology of the evaporated PbI_2 film, as seen in SEM (Fig. S15) and atomic force microscopy images (Fig. S16). However, steady-state PL measurements revealed a slight increase in PL intensity for CsBr-treated films, consistent with improved optoelectronic quality (Fig. 4a). The emission peak of Bottom- and Dual-CsBr samples also showed a blue shift compared to the Control. From the EQE derivative, the bandgap has been found to stay around $\sim 1.55\ \text{eV}$, slightly blue-shifted compared to the Control sample (Fig. S17). The observed

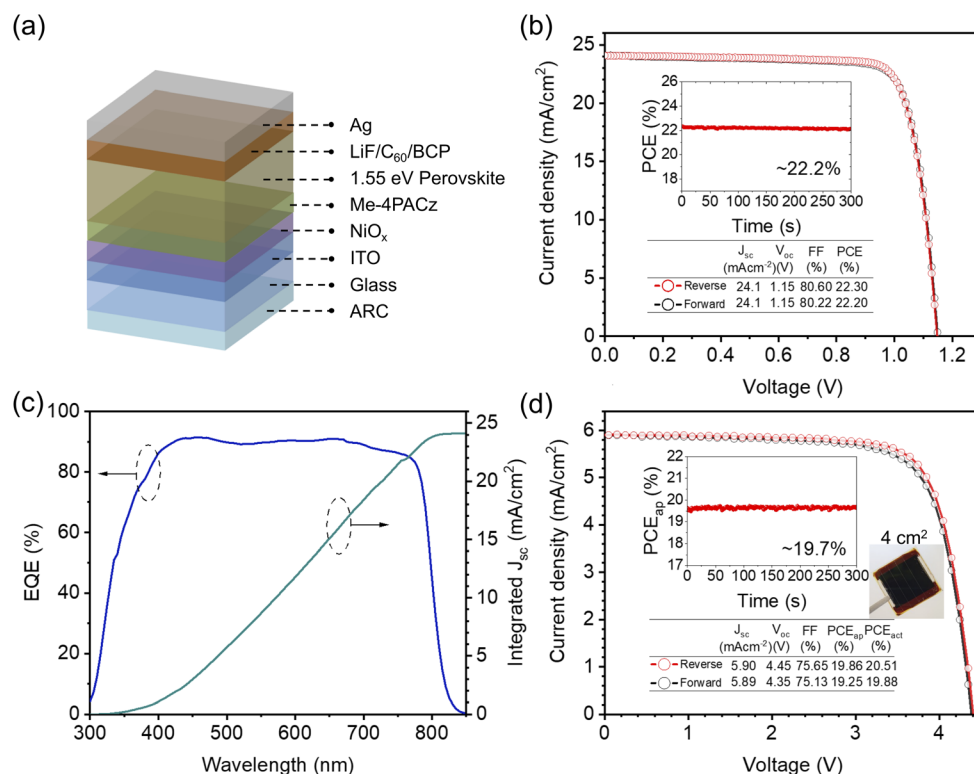


Fig. 3 Opto-electrical properties and performance of perovskite devices. (a) Schematic of solar cell architecture. (b) JV curves, and (c) EQE spectra of a champion perovskite solar cell using the hybrid method. (d) JV curves of perovskite solar minimodule. Insets in (b and d) show maximum power point (MPP) tracking of the devices in an inert atmosphere. A photograph of the fabricated minimodule is also shown as an inset in (d).

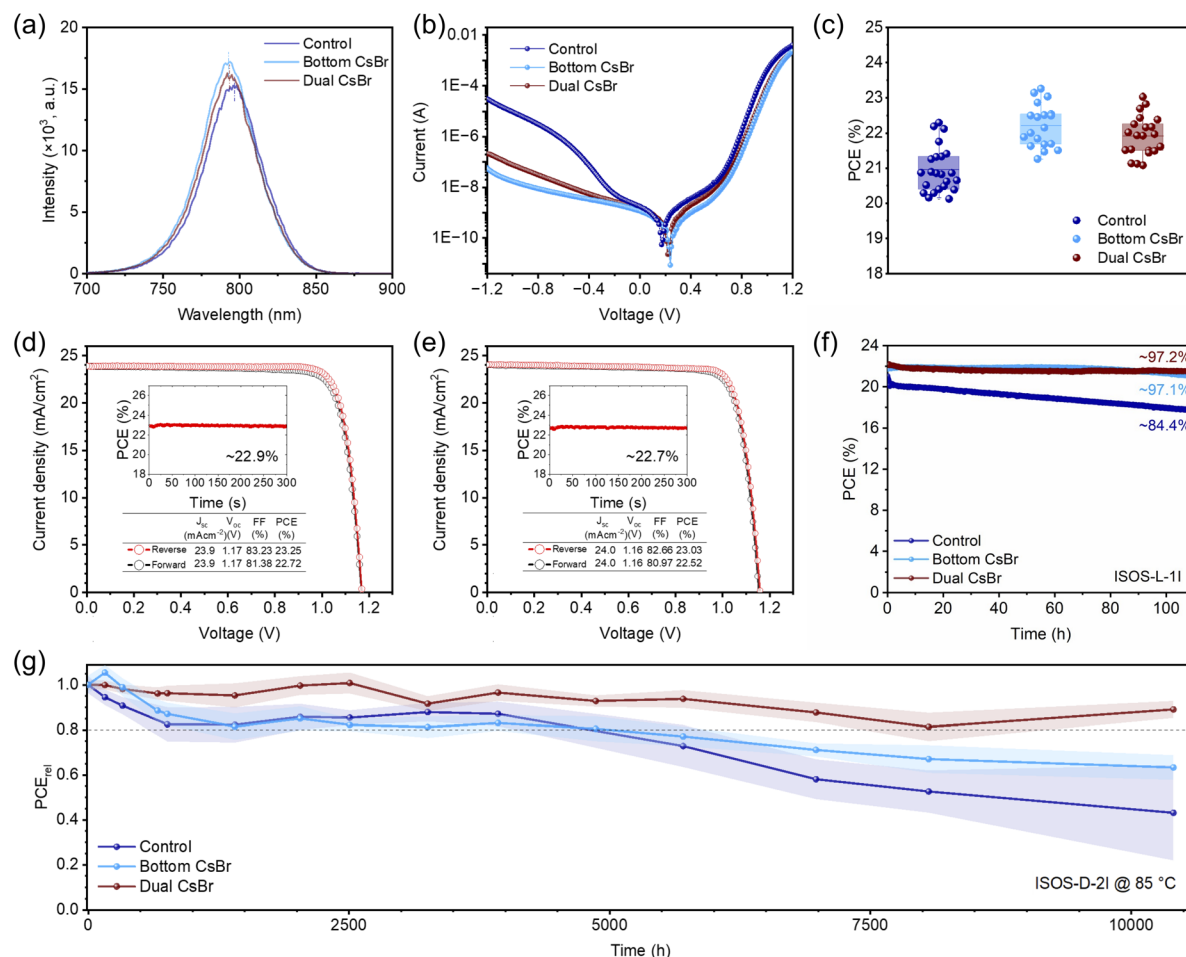


Fig. 4 Opto-electrical properties and performance of CsBr-treated perovskite devices. (a) PL spectra, (b) dark I/V curves, and (c) light I/V statistics on PCE of Control, Bottom-CsBr and Dual-CsBr perovskite devices. (d and e) JV curves of champion Bottom-CsBr (d) and Dual-CsBr (e) solar cells. (f) MPP tracking under continuous 1 sun AM1.5G illumination (ISOS-L-1) for Control, Bottom-CsBr and Dual-CsBr devices. (g) Stability tracking of devices kept at 85 °C in N_2 and in the dark (ISOS-D-2I). The shaded area indicates the standard deviation of 8 devices. Insets in (d and e) show MPP tracking of the devices in an inert atmosphere.

PL and EQE shifts are consistent with the compositional modification associated with CsBr incorporation. As shown in Fig. 4b, devices treated with CsBr also showed reduced dark current compared to the Control, suggesting lower recombination rates within the solar cell.

Statistical device analysis (Fig. 4c and S18) shows that the average device V_{OC} increased from 1138 ± 8 mV for the Control to 1166 ± 9 mV and 1160 ± 6 mV for the Bottom-CsBr and Dual-CsBr devices, respectively. Similarly, the average fill factor (FF) went from $77.6 \pm 1.9\%$ for the Control to $80.6 \pm 1.8\%$ and $79.8 \pm 1.6\%$ for the Bottom-CsBr and Dual-CsBr devices. The highest V_{OC} of 1.18 V was achieved for Bottom-CsBr devices. The champion Bottom-CsBr device reached a PCE of 23.25%, with a J_{SC} of 23.9 mA cm^{-2} , V_{OC} of 1.17 V, and FF of 83.23%. The champion Dual-CsBr device yielded a similar PCE of 23.03%, with a J_{SC} of 24.0 mA cm^{-2} , V_{OC} of 1.16 V, and FF of 82.66%. The corresponding JV curves are shown in Fig. 4d and e, respectively. Notably, devices employing Me-4PACz directly on ITO reached a maximum PCE of 23.73% (Fig. S19), demonstrating the performance potential of the hybrid FAPbI₃-based platform.

However, these devices exhibited reduced operational stability compared to devices incorporating sputtered NiO_x , motivating the focus of the CsBr interfacial engineering and long-term stability study on the NiO_x -based platform. The achieved PCEs are among the highest reported for p-i-n devices based on the two-step hybrid deposition, as well as figuring among the highest overall efficiencies for sputtered NiO_x -based PSCs (SI Table S2).

Interestingly, as shown in Fig. 4c, the introduction of CsBr at the perovskite surface did not lead to a noticeable further improvement in photovoltaic performance compared to Bottom-CsBr devices. While the impact on photovoltaic performance is limited, the influence of the Top CsBr treatment on device stability is considerably more pronounced. Maximum power point (MPP) tracking measurements under continuous 1 sun AM1.5G illumination (ISOS-L-1) were performed for Control, Bottom-CsBr and Dual-CsBr devices (Fig. 4f). Under these operationally relevant conditions, Bottom-CsBr and Dual-CsBr devices retained $\sim 97.1\%$ and $\sim 97.2\%$ of their initial efficiency after 108 h of continuous operation, respectively,

whereas the Control devices retained $\sim 84.4\%$, confirming a clear suppression of early-stage degradation under illumination.

For the long-term aging experiments at 85°C in N_2 and in the dark (ISOS-D-2I),⁶³ the BCP and Ag top contacts were replaced with an ALD SnO_2 layer and ITO, respectively, to minimize degradation associated with the metallic top electrode and to allow for a clearer assessment of the intrinsic thermal stability of the perovskite layer and its interfaces.^{26,64,65} Under these conditions, the Dual-CsBr devices showed an outstanding stability, retaining about 88.9% of their initial PCE after $\sim 10\,400$ hours of aging, whereas the Control and Bottom-CsBr devices dropped below 80% around the ~ 5000 hours mark (Fig. 4g).

Additional measurements of Top-CsBr devices under thermal-stress conditions indicate an intermediate thermal stability improvement compared to Control and Bottom-CsBr configurations, further supporting the role of the top interface in mitigating long-term degradation (Fig. S20). The dual-interface treatment helped mitigate J_{SC} and FF losses during aging (Fig. S21). Mini-module devices (active area 4 cm^2) show the same qualitative stability trend, with Dual-CsBr retaining 85.8% of the initial PCE after ~ 1400 h, compared to 68.9% for Control (Fig. S22). The reduced absolute lifetime relative to pixel

devices may arise from multiple factors, including spatial non-uniformity associated with the larger-area spin-coating process, which can lead to variations in perovskite conversion and film quality, as well as potential additional degradation pathways associated with laser-scribed regions and exposed device edges introduced during cell interconnection.⁶⁶

To gain insight into the mechanisms underlying the remarkable thermal stability of the Dual-CsBr-treated devices, we investigated the morphological, electronic, and compositional properties of perovskite surfaces before and after aging at 85°C . Top-view SEM images of pristine Control and Dual-CsBr perovskite films (Fig. 5a and c) reveal comparable grain sizes, ranging from several hundred nanometers to approximately $1\text{ }\mu\text{m}$. However, only the Control sample exhibits small, bright surface features, which are attributed to residual PbI_2 grains left after annealing. In contrast, the Dual-CsBr film appears more uniform, suggesting that the CsBr top layer promotes (re-) conversion of surface PbI_2 into perovskite during post-deposition annealing. This effect is not observed for the Bottom-CsBr configuration, where PbI_2 grains persist at the surface (Fig. S23).

Kelvin-Probe Force Microscopy (KPFM) measurements were performed on pristine and aged perovskite films. The corresponding topography images can be found in Fig. S24. For the

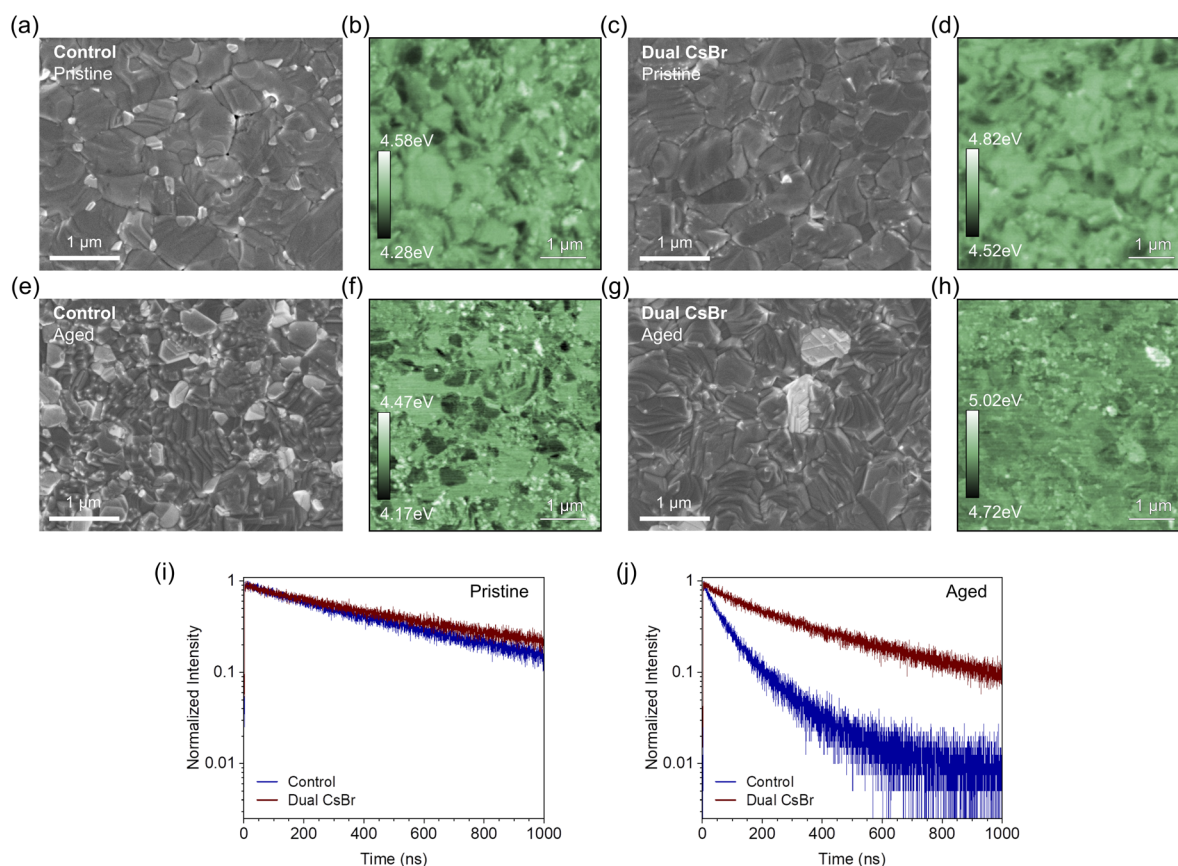


Fig. 5 Impact of Dual-CsBr treatment on the perovskite's morphological and optoelectronic properties before and after thermal aging. (a, c, e and g) SEM images, (b, d, f and h) KPFM work function images, and (i and j) time-resolved PL decay of pristine (a–d and i) and thermally aged (e–h and j) Control and Dual-CsBr perovskite films, on glass/ITO/HTL. SEM images were acquired with an in-lens SE detector.

pristine films (Fig. 5b and d), the Dual-CsBr film showed a more homogeneous surface potential than the Control film, along with slightly higher average work function (4.6 eV) for the Dual-CsBr compared to Control (4.4 eV). Consistent trends are observed by UPS measurements (Fig. S25), yielding work functions of 4.76 eV and 4.51 eV for Dual-CsBr and Control, respectively. While the impact of work function shifts depends on the complete device energetics, the work function map observations are consistent with reduced surface electronic heterogeneity and suppressed non-radiative recombination pathways.

After thermal aging, the differences between samples become markedly more pronounced. SEM images of aged Control films (Fig. 5e) reveal severe morphological degradation, including the formation of distinct secondary phases and nanoscale precipitates that roughen the perovskite grain surfaces. Energy-Dispersive X-ray Spectroscopy (EDX) confirms that these bright domains are Pb-rich and depleted in nitrogen, consistent with PbI_2 formation due to thermally induced perovskite decomposition (Fig. S26).^{3,67} In contrast, aged Dual-CsBr films (Fig. 5g) retain a comparatively uniform morphology, with only isolated secondary-phase domains observed. Bottom-CsBr films show intermediate degradation, with partial suppression of nanoscale precipitates but continued presence of bright domains (Fig. S27).

Post-aging KPFM measurements corroborate these trends: the Control film (Fig. 5f) develops strong spatial inhomogeneities in

surface potential, including regions of reduced work function consistent with PbI_2 accumulation, whereas the Dual-CsBr film (Fig. 5h) maintains a relatively uniform potential distribution. The absolute average work function values of the aged samples need to be treated with care since tip-sample interactions were present after the aging (Fig. S28).

Time-resolved photoluminescence (TRPL) measurements further support the improved stability conferred by the Dual-CsBr treatment. In pristine conditions, both samples show comparable carrier lifetimes (Fig. 5i). Upon aging, however, the Control film exhibits a substantial reduction in lifetime, consistent with increased non-radiative recombination due to defect formation (Fig. 5j). A similar behavior was observed with the Bottom-CsBr film (Fig. S29). In contrast, the Dual-CsBr film displays a substantially smaller decline in carrier lifetime, suggesting better preservation of optoelectronic quality.

To probe the chemical origins of these effects, Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) depth profiling was performed on pristine and aged films. Interfacial transition regions at the surface and at the perovskite/HTL interface are indicated in the profiles; signals within these regions are affected by matrix-dependent ion yields and sputter-induced mixing, and compositional trends are therefore discussed within the perovskite bulk (*i.e.*, unpatterned gray area in Fig. 6 and S30–S32). Determination of transition regions is exemplified in Fig. S30.

For pristine films, Br^- depth profiles (Fig. S31a) reveal incorporation throughout the perovskite layer, with gradients

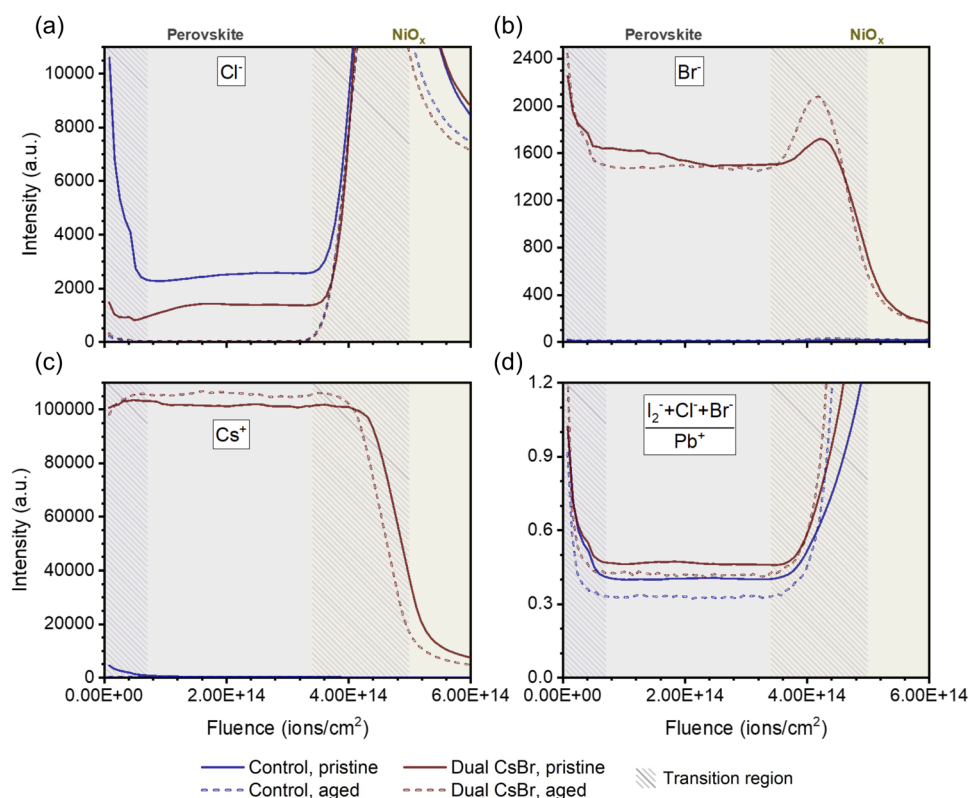


Fig. 6 Depth-resolved compositional evolution of perovskite films before and after thermal aging. ToF-SIMS depth profiles for (a) Cs^+ , (b) Br^- , (c) Cl^- , and (d) $(\text{I}_2^- + \text{Cl}^- + \text{Br}^-)/\text{Pb}^+$ of pristine and aged Control and Dual-CsBr perovskite films.

that depend on the CsBr deposition configuration (top and/or bottom). A pronounced Br enrichment near the surface is observed when CsBr is deposited on the already-formed perovskite (Top CsBr and Dual CsBr), whereas Br introduced at the buried interface shows a more uniform distribution, consistent with enhanced diffusion during perovskite annealing at 130 °C. Cs⁺ depth profiles show a clear presence of Cs throughout the full perovskite thickness, even when CsBr is introduced at only one interface (Fig. S31b). While matrix effects and partial signal saturation may limit a quantitative assessment of Cs⁺ concentration gradients, the detection of Cs deep within the film, including in Top-CsBr samples subjected only to brief low-temperature annealing, indicates significant Cs redistribution within the perovskite layer. Thus, although CsBr is introduced at the defined interfaces, the resulting compositional modification is not confined to these regions, and the effects of the treatment should therefore be considered in terms of both interfacial modification and Cs/Br redistribution within the perovskite layer.

Comparison of Control and Dual CsBr samples before and after aging reveals marked differences in halide chemistry (Fig. 6). In pristine CsBr-treated films, Cl[−] signals are strongly suppressed relative to Control, consistent with reduced residual MACl incorporation (Fig. 6a). A vertical Cl gradient observed in pristine films, characterized by lower Cl content near the surface, suggests partial retention of MACl during crystallization and incomplete volatilization during annealing. Critically, prolonged thermal stress at 85 °C leads to near-complete loss of detectable Cl in both samples.

Importantly, Br[−] remains in the perovskite bulk and is distributed uniformly after aging (Fig. 6b), while Cs⁺ is again detected across the full film thickness (Fig. 6c). This behavior suggests a stabilization pathway in which Br redistribution is associated with improved halide retention during aging, thereby mitigating the formation of halide-deficient domains that would otherwise favor PbI₂ reformation. To quantify this phenomenon, Fig. 6d presents the total halide-to-Pb ratio, using I₂[−] signal as a proxy for iodine due to saturation of I[−] signals. Dual-CsBr samples exhibit a consistently higher halide-to-Pb ratio in both pristine and aged states. Notably, the halide content of aged Dual-CsBr remains higher than that of the pristine Control film, consistent with suppressed perovskite decomposition. In accordance with these trends, organic-related fragments (CH₃N⁺) are relatively better preserved in the near-surface region of Dual-CsBr films compared to Control, further supporting suppressed perovskite decomposition (Fig. S32).

To further probe chemical degradation pathways, XPS analysis was performed on pristine and aged films. A low-binding-energy Pb 4f component consistent with metallic Pb⁰ formation was detected only in the aged Control sample, while no Pb⁰ signal was observed for the aged Dual-CsBr film (Fig. S33). This observation supports the suppressed perovskite decomposition in CsBr-treated devices.

While these results consistently indicate improved compositional stability in CsBr-treated films, direct quantification of

ion migration kinetics would be required for a more complete mechanistic description.

3. Conclusion

In summary, we demonstrate high-efficiency and exceptionally thermally stable p-i-n perovskite solar cells by combining a hybrid vapor-solution deposition route with a dual-interface engineering strategy based on evaporated CsBr layers. Through optimization of additive chemistry and processing conditions, uniform perovskite films with large grains and minimal residual PbI₂ were obtained, enabling power conversion efficiencies exceeding 23%, with a maximum value of 23.25% in devices employing sputtered NiO_x hole transport layers. Introducing CsBr at both the buried and top perovskite interfaces effectively suppresses defect formation, improves interfacial electronic homogeneity, and markedly enhances operational stability. As a result, Dual-CsBr devices retain, on average, over 88% of their initial efficiency after more than 10 400 h of thermal aging at 85 °C under ISOS-D-2I conditions, representing one of the longest reported stability demonstrations for p-i-n perovskite solar cells with sputtered oxide contacts. Correlative compositional, morphological, and optoelectronic analyses indicate that Cs/Br redistribution and reduced residual Cl content are associated with improved halide retention and suppressed PbI₂ reformation during aging, consistent with the observed preservation of surface morphology and carrier lifetimes. Overall, this work highlights the integration of vacuum-deposited interface engineering within hybrid-processed FAPbI₃-based systems as an effective and industry-relevant approach to achieving both high efficiency and long-term stability, offering a practical pathway toward durable and manufacturable perovskite photovoltaic technologies.

4. Experimental section

A detailed description of the materials and methods is available in the SI.

Author contributions

Cristian A. Villalobos Meza contributed to the conceptualization and development of the research idea and was responsible for methodology, investigation, formal analysis, data curation, visualization, and writing – original draft. Aslihan H. Babayigit and Sunil Suresh contributed equally to investigation, data curation, supervision, and manuscript review and editing. Gabriel Belchior Miranda, Jan D'Haen, Dharendra P. Singh, and Evandro Martin Lanzoni performed advanced characterization and contributed to data analysis and interpretation. Tamara Merckx, Aranzazu Aguirre, Johannes Lambooij, and Valerio Zardetto contributed to investigation and technical support. Thierry Conard and Alex Redinger contributed to characterization methodology, analysis, and interpretation. Anurag Krishna contributed to data analysis and scientific discussion. Yinghuan Kuang contributed to the conception of the project idea,

secured funding, and supervised the research together with Tom Aernouts, Bart Vermang, and Jef Poortmans. All authors reviewed and approved the final manuscript.

Conflicts of interest

The authors declare no conflicts of interest.

Data availability

The data supporting this article have been included as part of the supplementary information (SI). Supplementary information: experimental details and supplementary characterization data, including XRD, PL, NMR, SEM, AFM, EQE, EDX, XPS, UPS, ToF-SIMS, and photovoltaic device measurements and statistics. See DOI: <https://doi.org/10.1039/d6ta05732b>.

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