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Homogenized Optoelectronic Properties in Perovskites: Achieving High-Efficiency Solar Cells with Common Chloride Additives

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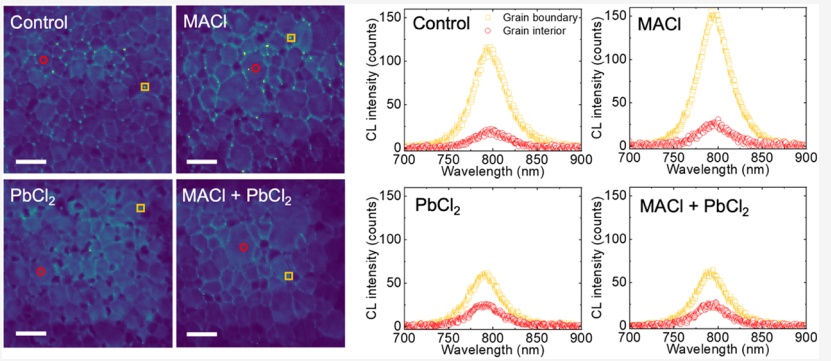

ABSTRACT: Improving the bulk quality of perovskite films is critical for achieving higher-performance photovoltaic devices. Chloride-containing additives, including lead chloride ($PbCl_2$) and methylammonium chloride (MACl)—standard additives widely adopted in perovskite photovoltaics—are effective for controlling crystallization kinetics and grain morphology. However, the distinct impacts of different forms of chloride additives on nanoscale phase uniformity and luminescence homogeneity remain underexplored. Here, we provide new insights into how the choice and combination of chloride additives influence phase transitions and spatially uniform carrier dynamics within perovskite films. We demonstrate that strategically combining MACl and $PbCl_2$ improves crystallinity and optoelectronic uniformity across dimensions spanning micrometers to millimeters. Leveraging these findings, we fabricated inverted (p-i-n) perovskite solar cells achieving certified quasi-steady-state efficiencies of 26.4% and 24.5% at device areas of 0.05 and 1 cm^2, respectively. Furthermore, these devices exhibit robust operational stability, retaining 88% of their initial performance after 1200 h of continuous maximum power point tracking at elevated temperatures (65 °C) under simulated AM1.5G illumination. Our results elucidate the mechanistic differences between chloride additive forms, providing a viable strategy for advancing large-area, high-efficiency, and thermally stable perovskite photovoltaics.

INTRODUCTION

Metal-halide perovskites are semiconductor materials that can be fabricated via cost-effective solution-[1,2] or vacuum-based[3,4] methods. Their remarkable optoelectronic properties[5,6] have positioned them as promising candidates for photovoltaic devices capable of efficiently converting sunlight into electricity.[7] In the past decade, substantial progress has been achieved in single-junction perovskite solar cells (PSCs), with power conversion efficiencies (PCEs) now exceeding 27%[8−11]—comparable to those of state-of-the-art silicon solar cells.[12,13]

Significant advancements in PSC performance have primarily resulted from reduced interfacial losses,[14−19] achieved through mitigating defect sites at perovskite heterojunctions and minimizing nonradiative recombination losses.[20−25] To further enhance device performance, precise control over local composition and optoelectronic uniformity within the perovskite absorber is essential.[26−30] Recent studies have highlighted that inhomogeneous phase distributions from micro- to millimeter scales are detrimental to device efficiency

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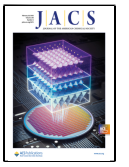

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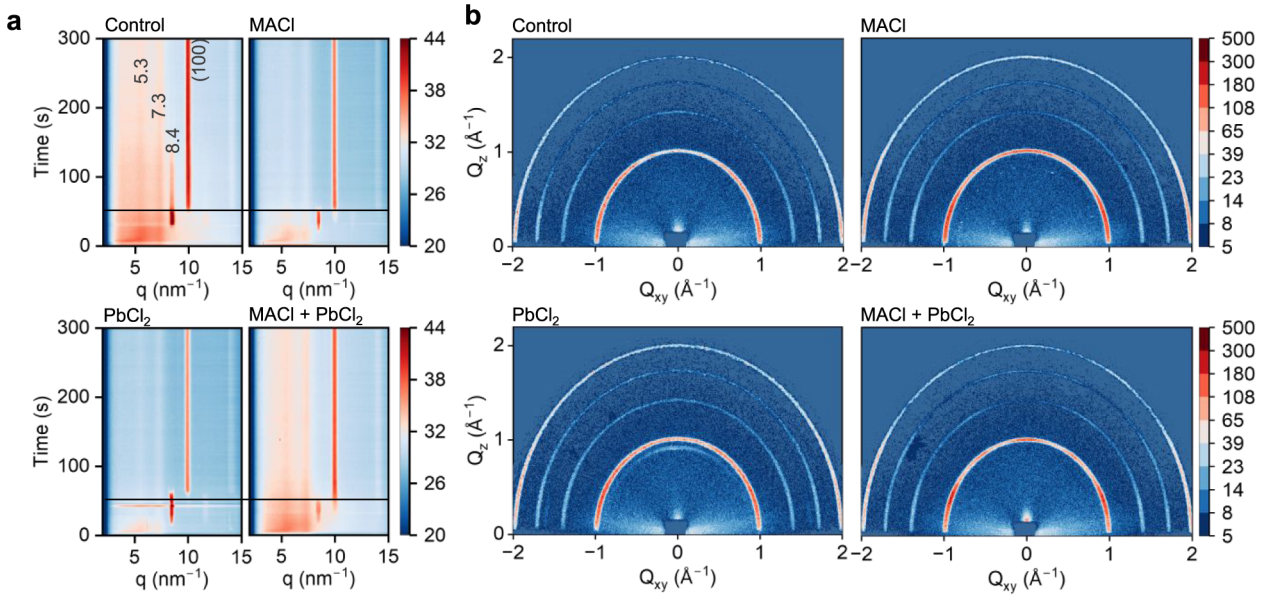

Figure 1. Film crystallization process. a, In situ (ψ = 0.4°) and b, ex situ (ψ = 0.4°) 2D GIWAXS patterns for the control, MACl, $PbCl_2$, and MACl + $PbCl_2$ samples. For the in situ measurements, the films are deposited on glass substrates in ambient air. The control sample is annealed at 150 °C for a full phase conversion and other samples are annealed at 120 °C. All films for ex situ GIWAXS measurements are deposited under inert conditions with an annealing temperature of 120 °C.

and stability, particularly when upscaling to larger areas.[31−34] Within solution-processing methods, achieving balanced crystallization among mixed-cation and mixed-anion components is critical to improve phase homogeneity.[30,35] Chloride-based additives have been proven effective for controlling crystallization kinetics and improving the semiconductor quality of perovskite films;[6,36−40] however, their influence on local optoelectronic uniformity remains insufficiently understood.

Here, we investigate the influence of different forms of chloride additives on the phase distribution and optoelectronic uniformity in perovskite films. By comparatively evaluating two widely adopted chloride additives[18]—methylammonium chloride (MACl) and lead chloride ($PbCl_2$)—we confirm that chloride incorporation notably accelerates the transition from the inactive yellow phase to the active black perovskite phase. We observe that MACl promotes the out-of-plane compositional heterogeneity, resulting in variable carrier dynamics at micrometer scales. In contrast, $PbCl_2$ more effectively incorporates chloride uniformly within grain interiors, homogenizing local optoelectronic properties but introducing detrimental lead iodide (PbI_2) grains that hinder charge transport. We demonstrate that a synergistic combination of MACl and $PbCl_2$ significantly improves overall film crystallinity and achieves spatially homogeneous carrier dynamics from microscale to millimeter-scale dimensions. Consequently, we fabricate inverted (p-i-n) PSCs that achieve efficiencies of up to 27.0% (certified 26.4%) at an active area of approximately 0.05 cm^2, and 25.5% (certified 24.5%) at 1 cm^2. These devices retain 88% of their initial PCE after 1200 h of continuous maximum power point tracking (MPPT) under simulated AM1.5G illumination at about 65 °C. Although chloride-assisted crystallization and the transition from yellow to black perovskite phases using MACl or $PbCl_2$ are well-documented, our study provides a systematic investigation into their distinct impacts on compositional and morphological uniformity at both micrometer and millimeter scales. We demonstrate how these pathways govern local phase and luminescence homogeneity, as well as carrier dynamics, offering a mechanistic understanding of how high-efficiency and stable perovskite photovoltaics can be achieved using widely accessible and commonly utilized chloride additives.

RESULTS

Phase Transition of Chloride-Engineered Perovskites

We prepared perovskite films with an initial composition of $Cs_{0.05}FA_{0.90}MA_{0.05}PbI_3$ (FA: formamidinium) using solution processing as control samples. To study the mechanism of chloride incorporation, we fabricated films with chloride additives: MACl (13 mol %), $PbCl_2$ (3 mol %), and their combination (13 mol % MACl + 3 mol % $PbCl_2$, relative to PbI_2) after initial device optimization (Supplementary Figures 1−2). Synchrotron radiation-based in situ grazing incidence wide-angle X-ray scattering (GIWAXS) was employed to track film crystallization during spin coating and subsequent annealing (Figure 1a). In the initial stage of spin coating (0 to 40 s), we observe two distinct diffraction peaks at scattering vectors (q) of 5.3 and 7.3 nm^{-1} regardless of using chloride additives, indicating low-dimensional intermediate phases composed of lead iodide-centered adducts with a specific number of solvent molecules or FAI-rich perovskite compounds.[41−43] Upon initiating antisolvent dripping at around 40 s, an additional diffraction peak emerges at q = 8.4 nm^{-1}. Subsequently, as annealing commenced at around 60 s, a new diffraction develops at q = 10 nm^{-1}. The diffraction peaks at 8.4 and 10 nm^{-1} represent the hexagonal 2H polytype (yellow δ-phase) and the black 3D perovskite phase, respectively.[41] Notably, the control sample exhibits incomplete conversion from the yellow to the black phase, maintaining strong diffraction intensity of the 2H phase even after 150 s at 150 °C. In contrast, samples incorporating chloride additives display significantly reduced intensity and a shorter duration of the 2H phase, completely eliminating the peak at 8.4 nm^{-1} within 60 s

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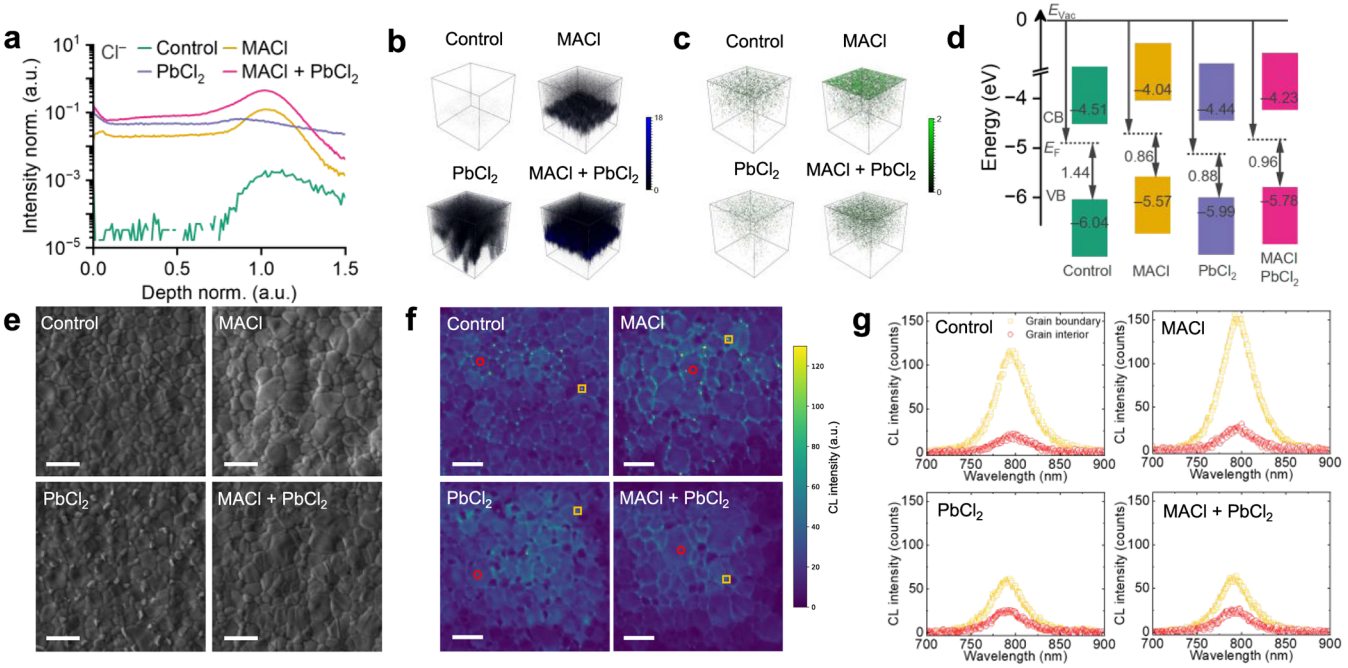

Figure 2. Phase distribution and nanoscopic emission of perovskite films. a, ToF-SIMS depth profiles of chloride anions. 3D tomography of b, chloride anion and c, MA cation. d, Energy level diagram generated from the UPS spectra of perovskite films fabricated without (control) and with additives. e, SEM, f, CL emission intensity images, and g, CL spectra of grain boundary (yellow square) and grain interior (red circle) regions of perovskite films fabricated without (control) and with additives. Scale bars in e and f are 1 μm. More CL spectra from 10 different pixels are shown in Supplementary Figure 14.

at 120 °C. We propose that the suppressed retention of the yellow 2H polytype is due to a kinetic effect introduced by chloride-containing additives. In the precursor stage, chloride ions can bind to the surfaces of the emerging face-sharing 2H crystallites.[44,45] Following antisolvent dripping, the chloride ions that coordinate with Pb^{2+} ions at the octahedral terminal corners destabilize the face-sharing polytype and promote the formation of corner-sharing 3D-like structures. The resulting seeding layer facilitates the complete conversion of black 3D phases in the bulk material. The trends in the 3D structure conversion rate are consistent with in situ photoluminescence (PL) measurements, in which the combination of MACl and $PbCl_2$ (hereafter denoted as MACl + $PbCl_2$) shows the fastest redshift and the highest 3D peak emission intensity, even without thermal annealing[46] (Supplementary Figures 3 and 4). Interestingly, the MACl + $PbCl_2$ film maintains intermediate phases ($q \leq 7.3$ nm^{-1}) throughout annealing up to 300 s, similar to the control film. Conversely, films with only MACl or $PbCl_2$ show reduced presence of these solvent-involved intermediate phases. Preserving intermediate phases while facilitating rapid δ-phase conversion can positively impact the crystallization pathway, leading to higher-quality perovskite crystals.[42] Ex situ GIWAXS measurements further assess the crystallinity of annealed perovskite films (Figure 1b). We observe no low-dimensional phases in any films after annealing. Films containing MACl, particularly those with MACl + $PbCl_2$, exhibit a significantly enhanced (100) diffraction intensities in both in-plane (Q_{xy}) and out-of-plane (Q_z) directions compared to the control film. In contrast, $PbCl_2$ alone provides limited improvement in crystallinity and promotes the undesirable formation of PbI_2 residue (q = 0.9 Å^{-1}).[3,47,48] These crystallographic analyses confirm that chloride additives universally facilitate the formation of the desired 3D perovskite phase. Importantly, combining MACl and $PbCl_2$ effectively preserves intermediate phases for an optimal duration, resulting in superior crystallinity of the final perovskite films (Supplementary Figures 5–6 and Supplementary Table 1).

Phase Distribution and Nanoscale Optical Properties

Following the crystallographic analysis, we investigate the influence of additives on phase distribution using time-of-flight secondary ion mass spectroscopy (ToF-SIMS). The perovskite film incorporating 3 mol % $PbCl_2$ exhibits a notably higher chloride content than the film containing 13 mol % MACl (Figure 2a and Supplementary Figures 7–8). This corresponds to a slight increase (~20 meV) in the photovoltaic bandgap (Supplementary Figure 9), likely reflecting the efficient integration of $PbCl_2$ into the perovskite grain interiors.[49] Moreover, 3D tomographic analysis of the $PbCl_2$-modified film reveals a uniform chloride distribution throughout its bulk (Figure 2b). Conversely, the MACl-containing film displays pronounced chloride accumulation near the buried interface and enrichment of MA cations toward the top surface (Figure 2c). This coincides with a modest increase in Cs^+ at the bottom interface and I^- at the top surface, respectively, which may help retain local electroneutrality in the bulk film (Supplementary Figure 10). We believe this phenomenon is due to the volatility of MACl during annealing. Chloride-containing species near the surface can escape more easily, while those deeper in the film are kinetically trapped,[50] leading to an apparent enrichment at the bottom. At the same time, MA-related species from both the MAI precursor and MACl additive can redistribute and are only partially vaporized, leaving some residual MA near the surface after annealing. These observations indicate that MACl is relatively inefficient at incorporating chloride within perovskite grains, resulting in significant compositional inhomogeneity. Additionally, we recognize that the chemical depth profile is sensitive to

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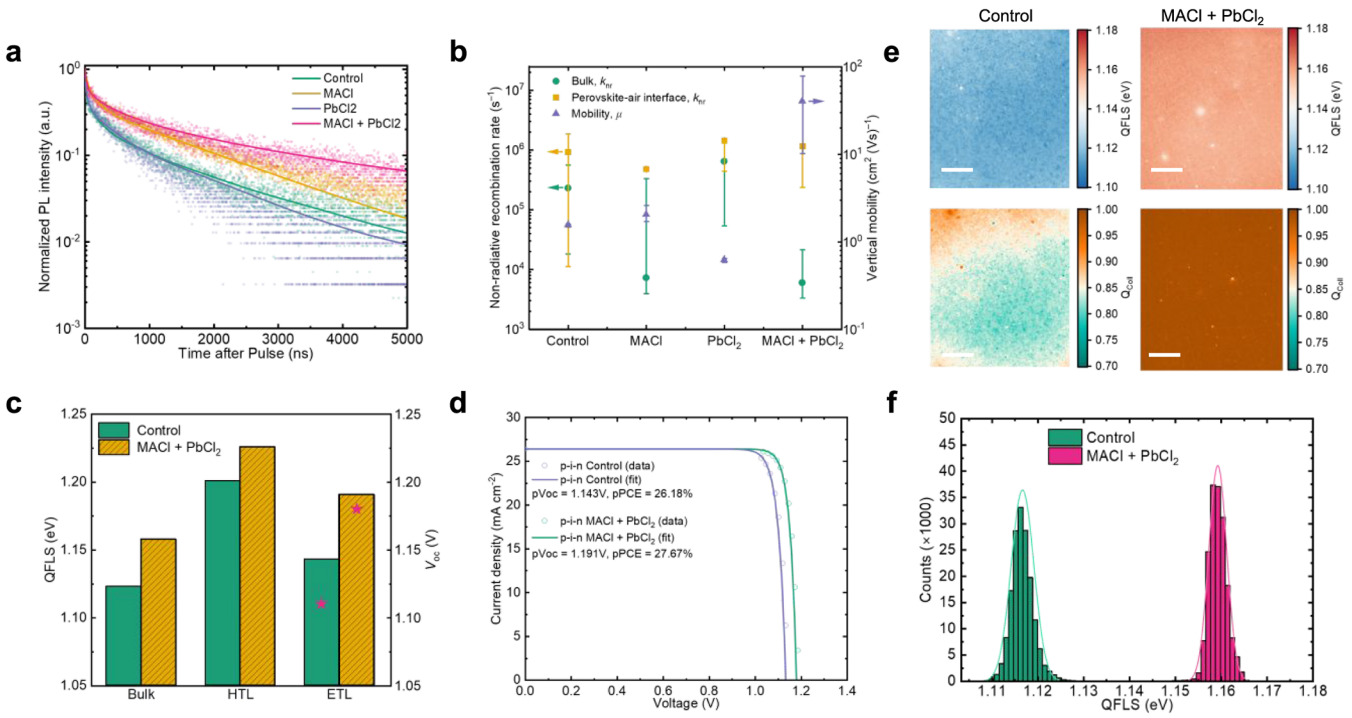

Figure 3. Optoelectronic properties of perovskite films. a,b, Representative TRPL response (a), the inferred nonradiative bulk and surface recombination rate, and the vertical mobility (b) for the control, MACl, $PbCl_2$, and MACl + $PbCl_2$ perovskite deposited on glass/ITO/HTL substrates. For clarity, only one TRPL measured from the glass side is shown (excitation fluence 1.3×10^9 cm^{-2}). The fitted curves from our global inference are shown in Supplementary Figures 21−24. In (b), the solid dots represent median inferred values and the error bars indicate the first and third quartiles. c, QFLS of control and MACl + $PbCl_2$ perovskite films deposited on bare glass, HTL half stack and p-i-n layout. The stars represent the measured device V_{OC} prepared in the same batch. d, Pseudo $J-V$ characteristics of control and MACl + $PbCl_2$ films processed in a p-i-n layout. e, QFLS imaging recorded at V_{OC} conditions and the charge collection quality mapping by comparing the PLQY at V_{OC} and J_{SC} conditions for control and MACl + $PbCl_2$-added PSCs. The scale bars are 1 mm. f, QFLS histograms of samples displayed in Figure 3e.

precursor stoichiometry and processing conditions. Changes in the intermediate adduct phases during antisolvent quenching and the redistribution of volatile species, such as MACl, during thermal annealing may lead to different apparent ion distributions.[51]

We further analyze the surface electronic structure using ultraviolet photoelectron spectroscopy (UPS, Figure 2d and Supplementary Figure 11). Compared to the control films, we identify the largest upward shift (0.47 eV) in the valence band maximum (VBM) for the MACl-containing films, followed by a smaller shift (0.26 eV) in the MACl + $PbCl_2$ films. The $PbCl_2$ film shows negligible shifts near the band edges. Correlating with the ToF-SIMS data, we attribute the pronounced VBM shift primarily to surface accumulation of MA cations, which lowers the ionization energy. Additionally, we observe a decreased offset between the Fermi level (E_F) and VBM in all chloride-engineered films, suggesting the formation of a more intrinsic, electronically neutral top surface. Based on the measured photovoltaic bandgap values, we estimate the conduction band minimum (CBM) to be −4.51, −4.04, −4.44, and −4.23 eV for the control, MACl-, $PbCl_2$-, and MACl + $PbCl_2$-added films, respectively. Considering the lowest unoccupied molecular orbital (LUMO) energy level (−4.20 eV) of the electron transport layer (ETL) C_{60},[52] the film modified with MACl + $PbCl_2$ presents the smallest interfacial energy mismatch on the electron extraction side.

To correlate film morphology with optical emission properties, we perform scanning electron microscopy (SEM) coupled with cathodoluminescence (CL) imaging at submicrometer resolution (Figure 2e−g). Top-view SEM images show that chloride additives effectively increase the apparent average grain size from 298 ± 96 nm (control) to 318 ± 151 nm (MACl), 272 ± 108 nm ($PbCl_2$), and 412 ± 171 nm (MACl + $PbCl_2$) (Supplementary Figure 12). Cross-sectional SEM images show that the film with only MACl has greater surface roughness due to the increased nonuniformity in grain morphology (Supplementary Figure 13). Correspondingly, CL imaging under continuous electron beam excitation reveals distinct photon emission behaviors. For the control films, emission near 800 nm (associated with the 3D perovskite phase) is stronger at grain boundaries compared to grain interiors, observed across multiple grain regions (Figure 2f,g and Supplementary Figure 14−16). Interestingly, MACl-added films exhibit an even more pronounced contrast, with grain boundaries emitting up to six times higher intensity than the interiors. This observation is unexpected since grain boundaries generally harbor higher defect densities and are known as nonradiative recombination sites. However, we find no clear correlation between grain size and bulk nonradiative recombination rate, suggesting grain boundaries are not dominate in nonradiative recombination processes (Supplementary Figure 25). Additionally, the overall averaged CL intensity and maximum emission wavelengths show minimal variations among the different samples (Supplementary Figure 17), highlighting that the observed emission contrast is highly localized to grain boundaries. We rule out beam-induced halide segregation as a possible cause of brighter grain boundaries[53] since no energy shifts are detected between grain boundary and interior regions (Figure 2g and Supplementary Figure 15−16). While beam-induced degradation within the grain

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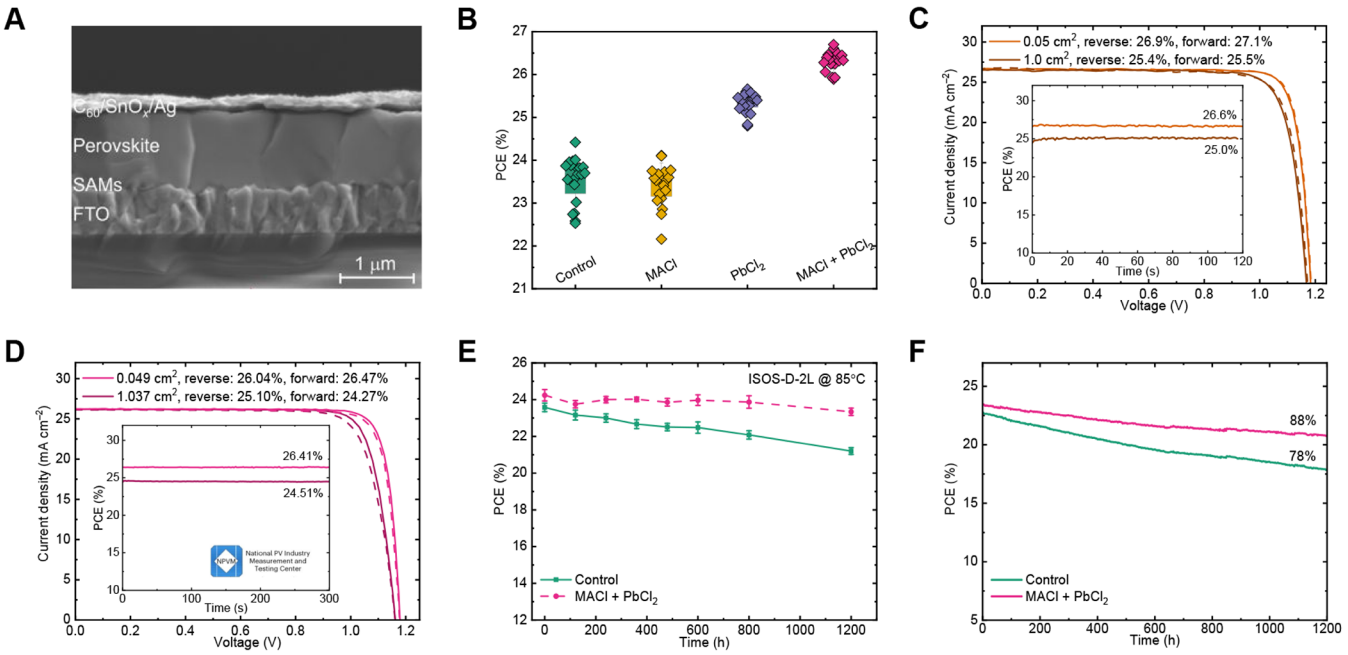

Figure 4. Photovoltaic and stability performance of PSCs. a, Cross-sectional SEM of a p-i-n cell stack. The scale bar is 1 μm. b, PCE statistics (reverse scan) for 20 control, MACl, $PbCl_2$, and MACl + $PbCl_2$ added devices. c, d, $J-V$ curve of the champion devices (~0.05 or 1.0 cm^2) measured in-lab and at the National PV Industry Measurement and Testing Center. The insets display steady-state PCEs under MPPT. e, ISOS-D-2L device stability during storage at 85 °C. Statistics are taken from 8 devices for each condition. f, MPP stability tracking of the encapsulated device under simulated 1-sun illumination at 65 °C.

interiors may be a potential factor,[54] we propose enhanced surface roughness from MACl incorporation increases scattering and optical out-coupling efficiency, contributing to greater local emission brightness.[55,56]

In contrast, $PbCl_2$- and MACl + $PbCl_2$-films exhibit significantly narrower variations in CL emission intensity across grain boundaries and interiors. This enhanced uniformity in emission characteristics reflects improved morphological and optoelectronic homogeneity. However, $PbCl_2$-added films featured distinct needle-shaped features attributed to excess PbI_2, visible in CL maps filtered within the 510−550 nm spectral range (Supplementary Figures 18−20). Combined with our ToF-SIMS analysis, these results indicate MACl alone promotes morphological disorder and nonuniform optoelectronic properties, whereas $PbCl_2$ contributes to uniform emission properties but introduces unwanted PbI_2 phases. Ultimately, the combination of MACl and $PbCl_2$ additives provides a balanced approach, yielding highly uniform carrier dynamics across micrometer-scale regions.

Carrier Dynamics of Perovskite Films and Devices

We further investigate the carrier dynamics within perovskite films using time-resolved photoluminescence (TRPL) measurements. Consistent with our previous approach,[57] we acquire four TRPL decay curves using two different excitation fluences (1.3×10^9 and 1.3×10^{10} cm^{-2} per pulse) on both the top and bottom surfaces of the perovskite samples (Figure 3a and Supplementary Figure 21−24). Employing a comprehensive physical model coupled with Bayesian inference, we extract eight device-relevant parameters, including the bulk and surface recombination velocities. We calculate the median bulk recombination rates to be 2.3×10^4, 7.1×10^3, 6.3×10^4, and 5.9×10^3 s^{-1} for the control, MACl, $PbCl_2$, and MACl + $PbCl_2$ samples, respectively. We interpret that improved film uniformity does not necessarily lead to a reduced average bulk recombination rate. While $PbCl_2$ facilitates more uniform chloride incorporation, it also promotes the formation of PbI_2 phases (Supplementary Figures 6 and 20), which can introduce nonradiative recombination centers. Interestingly, the MACl + $PbCl_2$ condition balances these competing effects, resulting in highly crystalline, phase-pure films with homogeneous carrier dynamics and reduced nonradiative bulk recombination. In comparison, median surface recombination rates are found to be 9.2×10^5, 4.7×10^5, 1.4×10^6, and 1.2×10^6 s^{-1} for the control, MACl, $PbCl_2$, and MACl + $PbCl_2$ samples, respectively, suggesting a comparatively minor influence of chloride treatments on surface recombination processes (Figure 3b). Additionally, we estimate the vertical charge-carrier mobility of the films, considering spectral changes caused by self-absorption during pulsed photoexcitation (Methods and Supplementary Figure 26).[57,58] Remarkably, the MACl + $PbCl_2$-modified film shows enhanced vertical mobility of 40.2 cm^2 V^{-1} s^{-1} compared to substantially lower values (0.6−2.0 cm^2 V^{-1} s^{-1}) in other films. Hence, the combined use of MACl and $PbCl_2$ considerably improves charge transport properties beneficial for photovoltaic applications.

We subsequently perform quasi-Fermi level splitting (QFLS) analysis via absolute PL to evaluate the intrinsic efficiency potential of perovskite films, without the influence of series resistance losses (Figure 3c). Comparing neat films processed on bare glass with those integrated into half-cells using NiO_X/[4-(3,6-dimethyl-9H-carbazol-9-yl)butyl]-phosphonic acid (Me-4PACz) as the hole transport layer (HTL), we observe QFLS enhancements of 78 and 68 meV for the control and MACl + $PbCl_2$-modified films, respectively. Correspondingly, the implied open-circuit voltage (iV_{OC}) reaches 1.201 V in the control sample and 1.226 V in the MACl + $PbCl_2$ film, consistent with the passivation effect from

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the self-assembled monolayer[25] and suggesting better optoelectronic quality for the latter samples. Interestingly, the subsequent deposition of a C_{60} layer (forming a p-i-n structure) tremendously reduces the iV_{OC} of the control sample to 1.143 V, whereas the MACl + $PbCl_2$ sample retains a much higher iV_{OC} of 1.191 V. Simultaneously, the control sample allows for a pseudofill factor (*p*FF) of 86.8%, compared with a higher 88.1% obtained for the MACl + $PbCl_2$ device. This indicates that the MACl + $PbCl_2$ additive reduces nonradiative recombination and restores fast charge extraction when in contact with a C_{60} layer atop.[25] Accordingly, pseudocurrent density−voltage (*pJ*−*V*) characteristics imply efficiency potentials of 26.18% for control and 27.67% for MACl + $PbCl_2$-based device.

To bridge local optical properties with macroscopic device performance, we perform absolute PL imaging on complete solar cells under 1-sun equivalent illumination, both at open-circuit (OC) and short-circuit (SC) conditions (Methods). At OC, the MACl + $PbCl_2$ device exhibits a higher mean QFLS (1.16 eV) compared to 1.12 eV for the control device (Figure 3e, f). This aligns with the PLQY data and suggests that the MACl + $PbCl_2$ device exhibits improved luminescence quality over millimeter length scales. We note that a reduced surface energy-level mismatch from the UPS data may also contribute to the increased iV_{OC}.[59,60] At SC, devices with higher charge extraction efficiency should exhibit lower PL signals.[60,61] Therefore, comparing the QFLS maps at OC and SC allows us to spatially resolve the charge collection quality (Q_{coll}) of the solar cells.[61] We observe that the control device displays more inhomogeneities, resulting in an approximate Q_{coll} of 85%. In comparison, the MACl + $PbCl_2$ device shows more uniform morphology, achieving an increased Q_{coll} of about 95%. These findings align closely with the CL analysis, underscoring that enhanced luminescence uniformity in MACl + $PbCl_2$ films promotes efficient charge extraction and higher achievable device V_{OC}.

Photovoltaic Performance

To evaluate the impact of chloride additives on the photovoltaic performance, we fabricate p-i-n PSCs with the architecture of FTO/NiO_x/2PACz:Me-4PACz/perovskite/C_{60}/SnO_x/Ag (Figure 4a). The thickness of the perovskite layer is approximately 1 μm. Statistical analysis of 20 devices prepared in the same batch indicates a notable increase in PCE, rising from 23.5 ± 0.5% (control) to 25.3 ± 0.2% ($PbCl_2$-treated) and further to 26.3 ± 0.2% (MACl + $PbCl_2$-treated) (Figure 4b). Correspondingly, both the averaged V_{OC} and FF are improved from 1.11 V (control) to 1.16 V ($PbCl_2$) and 1.18 V (MACl + $PbCl_2$), and from 0.79 (control) to 0.82 ($PbCl_2$) and 0.84 (MACl + $PbCl_2$), respectively (Supplementary Figure 27). The enhancement in FF can be attributed to reduced transport losses, compared to the resistance-free pseudo-FF values (Figure 3d and Supplementary Table 5). We attribute the superior photovoltaic performance of the MACl + $PbCl_2$ devices to enhanced film uniformity and efficient charge extraction across micro- to millimeter scale dimensions. We note that the change in bandgap due to the chloride addition (20 meV, Supplementary Figure 9) is modest compared to the substantial V_{OC} increase in the $PbCl_2$ and MACl + $PbCl_2$ treated devices. Interestingly, devices treated solely with MACl show limited performance gains despite improved film crystallinity. As shown in the UPS data (Figure 2d), a cliff-like band alignment between the MACl perovskite and C_{60} may facilitate electron transfer but can lower the V_{OC} due to increased interfacial nonradiative recombination.[59] In contrast, for $PbCl_2$, a moderate spike-like offset with C_{60} can help suppress recombination by blocking the backward flow of injected electrons while still allowing for efficient carrier extraction.[62,63] In our case, films made with only MACl also show increased roughness and local heterogeneity, which exacerbates interface-limited losses in the p-i-n architecture. To disentangle the specific role of additional MA and Pb cation from chloride effects, we fabricate reference devices incorporating 13 mol % MAI, 3 mol % PbI_2, or their combination (13 mol % MAI + 3 mol % PbI_2), relative to Pb (Supplementary Figure 28). We find that individual addition of MAI or PbI_2 adversely affects device performance; only their combination slightly increases the PCE from 23.1 ± 0.2% (control) to 24.1 ± 0.4%. These results confirm that performance gains primarily result from chloride incorporation rather than additional MA or Pb cations.

As a result, the optimized MACl + $PbCl_2$ device exhibits a maximum PCE of 27.1% (0.05 cm^2) and features a V_{OC} of 1.185 V, a FF of 0.858, and a J_{SC} of 26.6 mA cm^{-2}, with minimal hysteresis (Figure 4c). The corresponding 1 cm^2 device exhibits a maximum PCE of 25.5%, with a V_{OC} of 1.172 V, a FF of 0.816, and a J_{SC} of 26.7 mA cm^{-2}, with minimal hysteresis. Certified quasi-steady state (QSS) efficiencies of 26.4% and 24.5% are obtained for device areas of 0.05 and 1 cm^2, respectively, validated by the National Photovoltaic Industry Measurement and Testing Center (NPVM) (Figure 4d and Supplementary Figures 29−30). Furthermore, accelerated durability tests (ISOS-D-2L) at elevated temperatures show enhanced stability for encapsulated MACl + $PbCl_2$ devices, retaining over 96% of initial performance after 1200 h at 85 °C, compared to 90% for control devices. Additionally, under continuous maximum power point (MPP) operation at 65 °C and ∼50% relative humidity, MACl + $PbCl_2$-treated devices maintain 88% of initial PCE after 1200 h, significantly outperforming control devices (78% retention). The enhanced stability of MACl + $PbCl_2$ devices can be attributed to the improved phase purity and more uniform optoelectronic properties of the perovskite absorber.[40] Chloride additives help accelerate the transition from the yellow 2H to the more stable 3D phase. In our case, they also improve film crystallinity, luminescence uniformity, and vertical charge transport. These improvements are expected to reduce local recombination centers and suppress interfacial charge accumulation during operation, which can otherwise lead to ion migration and degradation under thermal and light stress. This interpretation is consistent with recent research indicating that optimizing the chloride distribution can enhance both device performance and operational stability.[51]

We would like to point out that chloride-containing additives cover a diverse range of chemical options, and in many cases, the accompanying cation can influence crystallization and interfacial chemistry beyond merely supplying Cl^- ions. In this study, we select MACl and $PbCl_2$ as model systems because they are among the most commonly used chloride additives across p-i-n and n-i-p device architectures. This allows us to compare different pathways for chloride incorporation without introducing extra functionalities and perovskite phases. Our findings indicate that beyond MA^+, substituting larger organic ammonium cations, such as propylammonium (PA^+)[44] and phenethylammonium (PEA^+),

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leads to decreased performance in our p-i-n devices (Supplementary Figure 31). This correlates with a greater tendency to form mixed reduced-dimensional perovskite phases, which can hinder charge collection.[64] Despite this, our findings indicate that Cl^- anions generally lead to better device performance in compared to I^- anions. These results underscore that while Cl^- typically has a beneficial effect on the quality of the perovskite absorber, the molecular structure of the cation becomes crucial when it leads to the formation of lower-dimensional perovskites.

CONCLUSIONS

In this study, we elucidated the critical role of chloride-based additives in controlling phase uniformity, compositional homogeneity, and luminescence properties of perovskite films, revealing their significant influence on photovoltaic device performance. We demonstrated that MACl additives increase compositional heterogeneity, causing spatially nonuniform carrier dynamics within perovskite grains. Conversely, $PbCl_2$ proved more effective at homogenizing local optoelectronic properties across the films but introduced undesirable PbI_2 phases, detrimental to efficient charge extraction. Importantly, the strategic combination of MACl and $PbCl_2$ enabled optimal crystallinity and uniform carrier dynamics extending from the microscale to millimeter-scale regions. Consequently, this approach significantly reduced interfacial recombination losses, enabling efficient charge-carrier extraction and achieving exceptionally high photovoltaic performance.

Although the chloride additives investigated here are standard in perovskite photovoltaics, our study offers new mechanistic insights by distinguishing the impacts of these commonly used additives. We reveal that different forms of chloride additives introduce unique pathways for enhancing perovskite film quality, emphasizing the importance of additive form and combination strategies. Our findings underscore the possibility of achieving high efficiencies using widely accessible additives, thus opening new avenues for further optimization and scalable deployment of high-performance perovskite solar cells.

ASSOCIATED CONTENT

Data Availability Statement

The raw data for this article is available on Oxford University Research Archive at https://ora.ox.ac.uk.

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/jacs.5c18303.

Materials, experimental methods, and additional device statistics, in situ PL, XRD, XPS, ToF-SIMS, UPS, CL, TRPL analysis, and solar cell certification reports (PDF)

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Notes

The authors declare the following competing financial interest(s): H.J.S. is the co-founder and CSO of Oxford PV Ltd. A.W. is the co-founder and CSO of Enecoat Technologies Co., Ltd. All other authors declare no competing interests.

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