

RESEARCH ARTICLE

SOLAR CELLS

Initializing film homogeneity to retard phase segregation for stable perovskite solar cells

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The mixtures of cations and anions used in hybrid halide perovskites for high-performance solar cells often undergo element and phase segregation, which limits device lifetime. We adapted Schelling's model of segregation to study individual cation migration and found that the initial film inhomogeneity accelerates materials degradation. We fabricated perovskite films ($\text{FA}_{1-x}\text{Cs}_x\text{PbI}_3$; where FA is formamidinium) through the addition of selenophene, which led to homogeneous cation distribution that retarded cation aggregation during materials processing and device operation. The resultant devices achieved enhanced efficiency and retained >91% of their initial efficiency after 3190 hours at the maximum power point under 1 sun illumination. We also observe prolonged operational lifetime in devices with initially homogeneous $\text{FACsPb}(\text{Br}_{0.13}\text{I}_{0.87})_3$ absorbers.

With an endless compositional space that is almost continuously tailorable in the full region, halide perovskite materials present tunable electronic and optical properties (1–5). Although perovskite solar cells can have high power conversion efficiency (PCE), their limited device lifetime remains a substantial challenge to commercialization (6–8). The use of mixed cations and anions has led to a more appropriate tolerance factor of the crystal for improved

structural stability and extra functionality to strengthen chemical stability in the resultant absorber thin films (9–11). However, mixed perovskite absorbers often undergo element and phase segregation that can decrease device efficiency and lifetime (12, 13).

Most studies of phase segregation of mixed perovskites focus on film aging to understand cation and anion migration, the formation and development of nanoscale clusters (14–16), thermodynamic driving forces (17, 18), and their impacts on film properties and device performance (12, 19–23). Effective strategies such as relaxing residual strain and incorporating low-dimension perovskites can retard phase segregation by suppressing ion migration (5, 24, 25). However, relevant metrics are needed to investigate the atomistic aggregation of individual ions to correlate their collective behavior that led to film degradation.

We adapted a physical analog of Schelling's model, which was used to illustrate agent (such as isolated particles) segregation caused by even low levels of individual preference in the context of social economics (26) and physics (27). It bridges the microscale analysis of cation aggregation in perovskites and macroscale observations of their phase separation and film degradation. In the simulation, the initial film homogeneity affected cation aggregation duration and thus device lifetime. On the basis of these findings, we grew homogeneous α -phase $\text{FA}_{1-x}\text{Cs}_x\text{PbI}_3$ (FA, formamidinium) that resulted in a solar cell with enhanced efficiency and prolonged operational lifetime. Furthermore, for the wide-bandgap $\text{FACsPb}(\text{Br}_{0.13}\text{I}_{0.87})_3$, the corresponding device

retained >80% of its initial PCE after 340 hours at maximum power point (MPP) tracking. It indicates that this model is applicable to various perovskite formulations such as mixed halide perovskites, which demonstrates the generality of the Schelling model for halide perovskites.

Cation aggregation and phase transition

The α -phase $\text{FA}_{1-x}\text{Cs}_x\text{PbI}_3$ (FACs) perovskites have an optimal bandgap and high thermal stability from the resultant crystal structure, with an ideal Goldschmidt tolerance factor (*11*, 21, 28–31). However, reported devices based on FACs perovskite absorbers have had limited lifetimes, resulting from material degradation that is largely the result of phase separation (32). In Fig. 1, A, C, and D, we show the two-dimensional (2D) photoluminescence (PL) mapping by wavelength, intensity, and full width at half-maximum (FWHM), respectively, of the $\text{FA}_{0.9}\text{Cs}_{0.1}\text{PbI}_3$ perovskite film. The emission spectra display peak variability between the selected regions. Moreover, although the emission of region 1 was concentrated at 795 nm (attributed to $\text{FA}_{0.9}\text{Cs}_{0.1}\text{PbI}_3$) (12), both regions 2 and 3 exhibited splitting of the emission peak (Fig. 1B). This feature was indicative of phase segregation within the perovskite film.

We investigated the segregation evolution of films aged under continuous light illumination for 20 min at 80°C at the same location in ambient atmosphere. As shown in the mapping spectra (Fig. 1E), the phase segregation of the film intensified with increasing areas of both the yellow domains (which represent the longer-wavelength, FA-rich composition) and red domains (which represent the shorter-wavelength, Cs-rich composition), which indicated that the local Cs/FA ratio deviated from that observed in the pristine films. Region 1 showed peak splitting at 799 and 789 nm, with increased peak intensity at longer wavelengths. By contrast, for regions 2 and 3, the peak intensity increase was observed at shorter wavelengths (783 and 781 nm, respectively). Moreover, the FWHM increased notably (Fig. 1H), which indicates the broadening of the PL emission peak after aging. After comparison with the pure FAPbI_3 film, for which no obvious change on the PL mapping was observed after aging (fig. S1), we hypothesized that Cs^+ cations in region 1 migrated to regions 2 and 3 in the mixed-cations composition of the perovskite with a single halide. This hypothesis was confirmed through time-of-flight secondary-ion mass spectrometry (TOF-SIMS) mapping (fig. S2).

Upon extending the aging duration to 1000 hours (supplementary materials), we observed crystal phase transformation in the film. In addition to the cubic perovskite peak (14.0°), two extra diffraction peaks at 9.7° and 11.6° were dominant in the x-ray diffraction (XRD) patterns (figs. S3 and S4), which indicates

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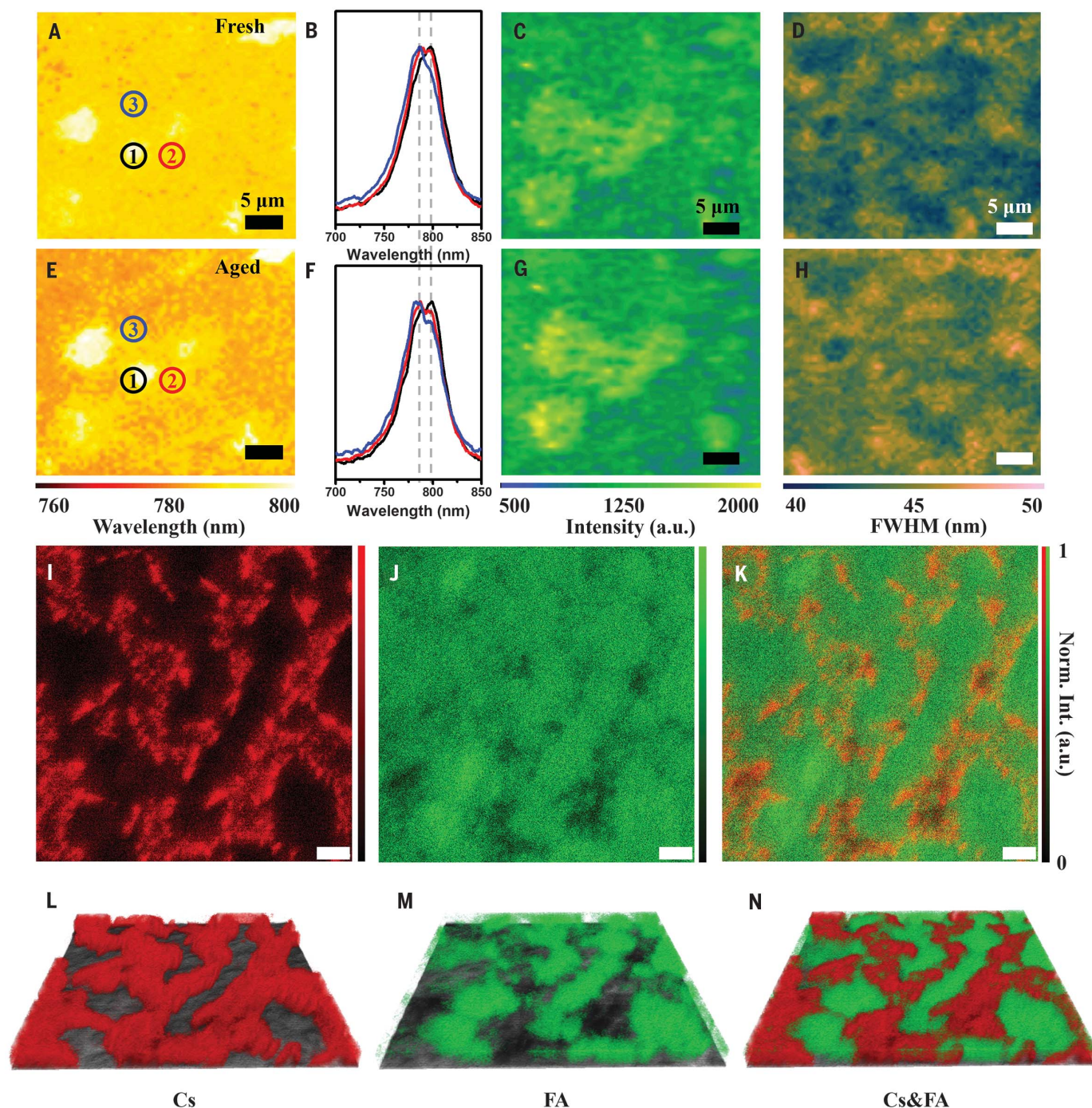


Fig. 1. Degradation mechanisms for binary FACs perovskites. (A to H) 2D PL mapping spectra (wavelength) of perovskite film before (A) and after (E) degradation. [(B) and (F)] PL spectra of selected regions from (A) and (E), respectively. [(C), (D), (G), and (H)] The corresponding 2D PL intensity and

FWHM mapping spectra of (A) and (E). a.u., arbitrary units. [(I to K)] The (I) Cs and (J) FA intensity mapping for the degraded film measured with TOF-SIMS and (K) the overlay mapping spectra. Scale bar, 10 μm. [(L to N)] 3D topographic of Cs and FA for the degraded films measured by TOF-SIMS.

the formation of nonphotoactive δ -CsPbI₃ and δ -FAPbI₃ phases (18, 33, 34). To correlate the composition of each region to its crystal structure, we conducted 2D TOF-SIMS mapping on the fully degraded film. The Cs distribution (Fig. 1I) showed domains at the scale of several to several tens of micrometers, which occurred in parallel to FA seg-

regation (Fig. 1J). The separated Cs/FA domains were also spatially complementary as expected (Fig. 1K), given the binary cation nature of the perovskite system. Moreover, depth-dependent TOF-SIMS measurement (Fig. 1, L to N) showed that vertical direction distribution was relatively uniform, so cations at different depths of the bulk ag-

gregated in a manner similar as that at the surface (fig. S5).

Cation segregation dynamics by the Schelling model

On the basis of the XRD, PL mapping, and TOF-SIMS results, we argue that the degradation of the binary system follows the route

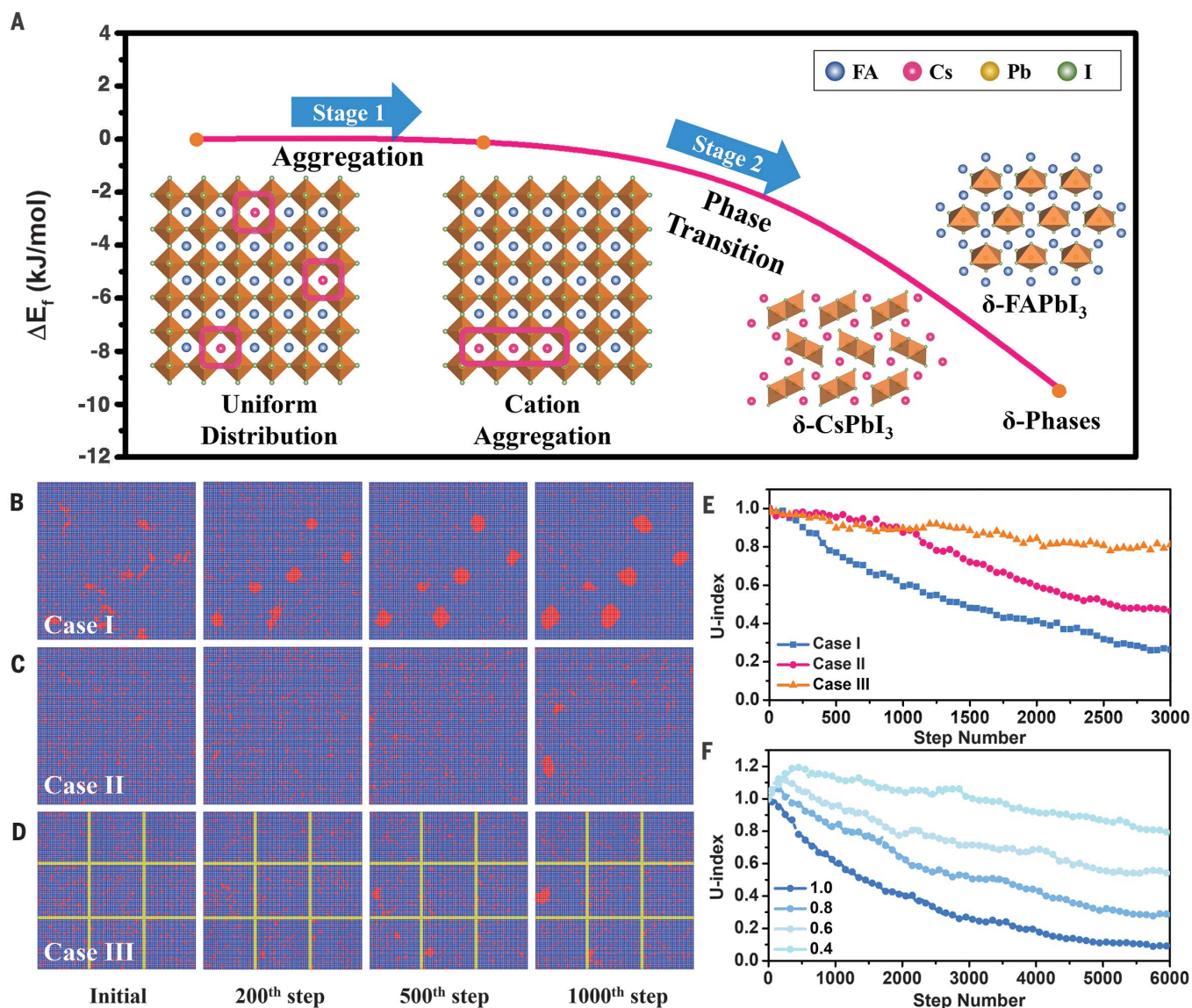


Fig. 2. Schelling model simulations of cation segregation dynamics. (A) Energy change of the degradation processes from a uniform distribution to phase segregation to δ -phases, by means of DFT. (B to D) Simulations of segregation process on the basis of the Schelling model. (B) An initially segregated

distribution (case I, ISP = 27%). (C) An initially less segregated distribution (case II, ISP = 2%). (D) A 1D and 3D (or 2D and 3D) fused system (case III, ISP = 2%). (E and F) U index evolution of red particles (indicates Cs) derived from (E) cases I to III and (F) cases with different moving tendencies.

from cation aggregation to phase transition (Fig. 2A). Local aggregation of cations initiates phase transition that leads to the complete process of phase segregation. The thermodynamic driving force for the phase segregation was investigated by means of density functional theory (DFT) calculations (fig. S6). DFT results identified that the $\text{FA}_{0.89}\text{Cs}_{0.11}\text{PbI}_3$ perovskite exhibited an energy change of only $-0.133 \text{ kJ mol}^{-1}$ from the homogeneous distribution to cation aggregation (Fig. 2A, stage 1), which is possibly due to the reduction in interfaces between the FA and Cs domains. However, in the subsequent process, the segregated domains experienced phase transition to form δ -FAPbI₃ and δ -CsPbI₃ phases (Fig. 2A, stage 2), in which the

energy change of $-9.427 \text{ kJ mol}^{-1}$ acted as the energy sink corresponding for the irreversible segregation. Our observations showed that cation aggregation occurred before phase transition, which provides an opportunity to intervene before δ -phase formation occurs (20, 21).

The Schelling model of segregation can be applied to isolated particles (such as molecules or ions) on the basis of a few simple physical assumptions (see supplementary text S1) (27). We adapted the Schelling model to study the cation dynamics in perovskite films by means of a Monte Carlo algorithm (fig. S7). Given that cations at different depths of the film aggregated in the same manner as at the surface (Fig. 1, I to N), we simplified the simulation

to a 2D case (35, 36) with a 100 by 100 lattice to accommodate binary cations of Cs (Fig. 1, red particles) and FA (Fig. 1, blue particles) that could randomly swap positions with neighbors.

In the simulation for any step, each single cation has a probability to exchange its position with neighbors. We defined this probability as moving tendency that is the product of the thermodynamic parameter of the phase transition (η_T) and kinetic parameter of cation aggregation (η_K). DFT results revealed the phase formation energy to identify η_T in the Schelling model. Particularly, η_T values were assigned to be 1 for all spontaneous processes with a negative value for energy change. The kinetic parameter η_K was determined by the

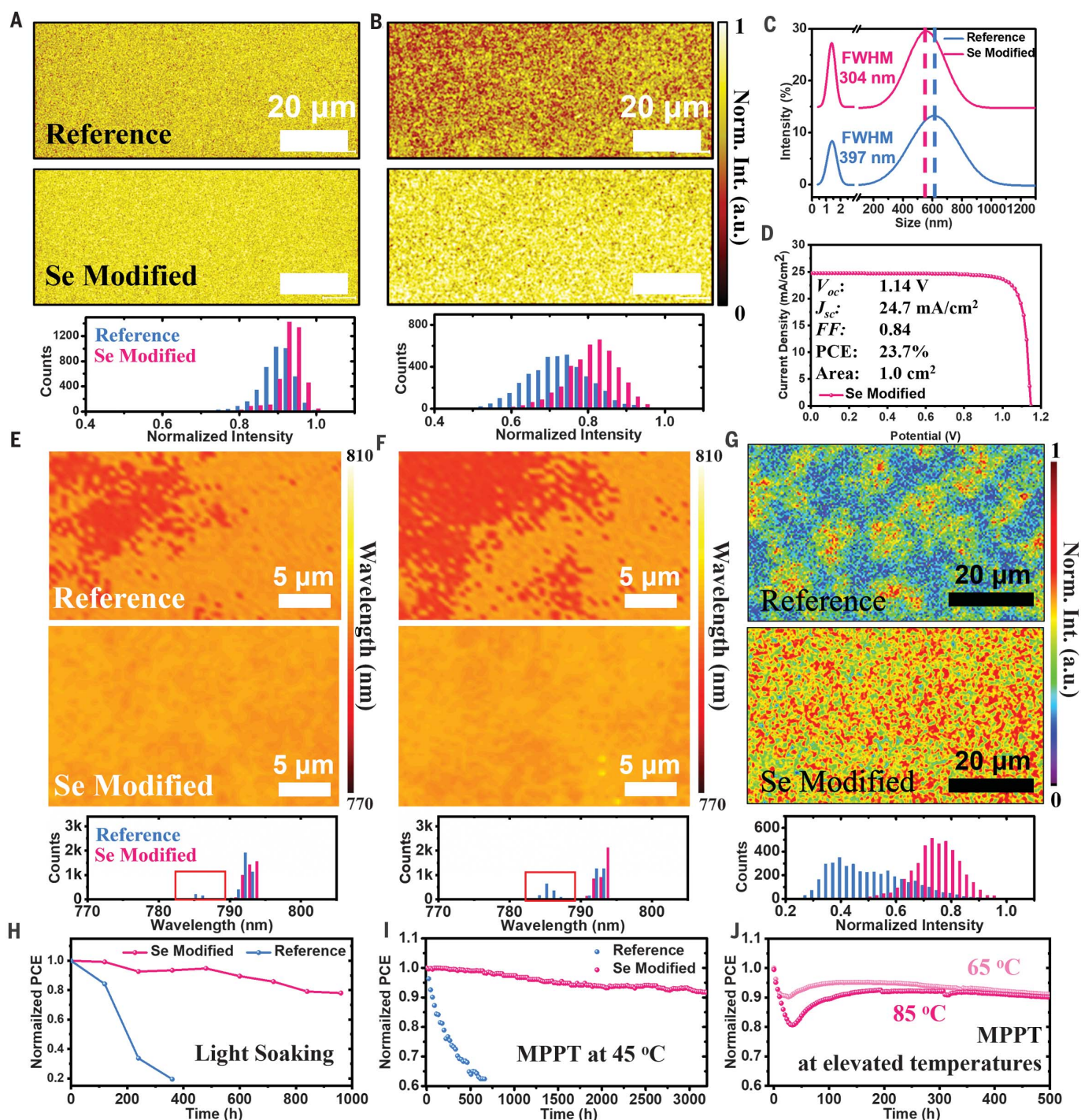


Fig. 3. Film homogeneity of mixed cation perovskite and the impacts.

(A and B) 2D TOF-SIMS mapping of Cs⁺ for (A) the first step of PbI₂/CsI film fabricated by reference and selenophene-modified precursor solutions and (B) perovskite films fabricated by reference and modified precursor solutions, as well as the corresponding intensity distribution. (C) Colloid size distribution in the perovskite precursor solutions obtained from DLS. (D) *J*-*V* curves of the champion devices from homogeneous film with an active area of 1 cm². (E and F) In situ 2D PL mapping spectra evolution of perovskite films (E) before and (F) after aging under 80°C thermal treatment in an ambient environment

with a relative humidity of 40% and the corresponding statistics diagrams. (G) 2D TOF-SIMS mappings of perovskite films (Cs) after continuous thermal treatment in an N₂-filled glovebox at 85°C for 144 hours with the corresponding statistics diagrams. (H) Time-dependent PCE measurements under continuous light illumination (LED source with 100 mW/cm²) held at the open circuit in an N₂ atmosphere. (I and J) MPP tracking of devices under 100 mW/cm² LED source in an N₂-filled glovebox without encapsulation at (I) 45° ± 5°C and (J) elevated temperatures of 65° ± 5°C and 85° ± 5°C. MPPT, MPP tracking.

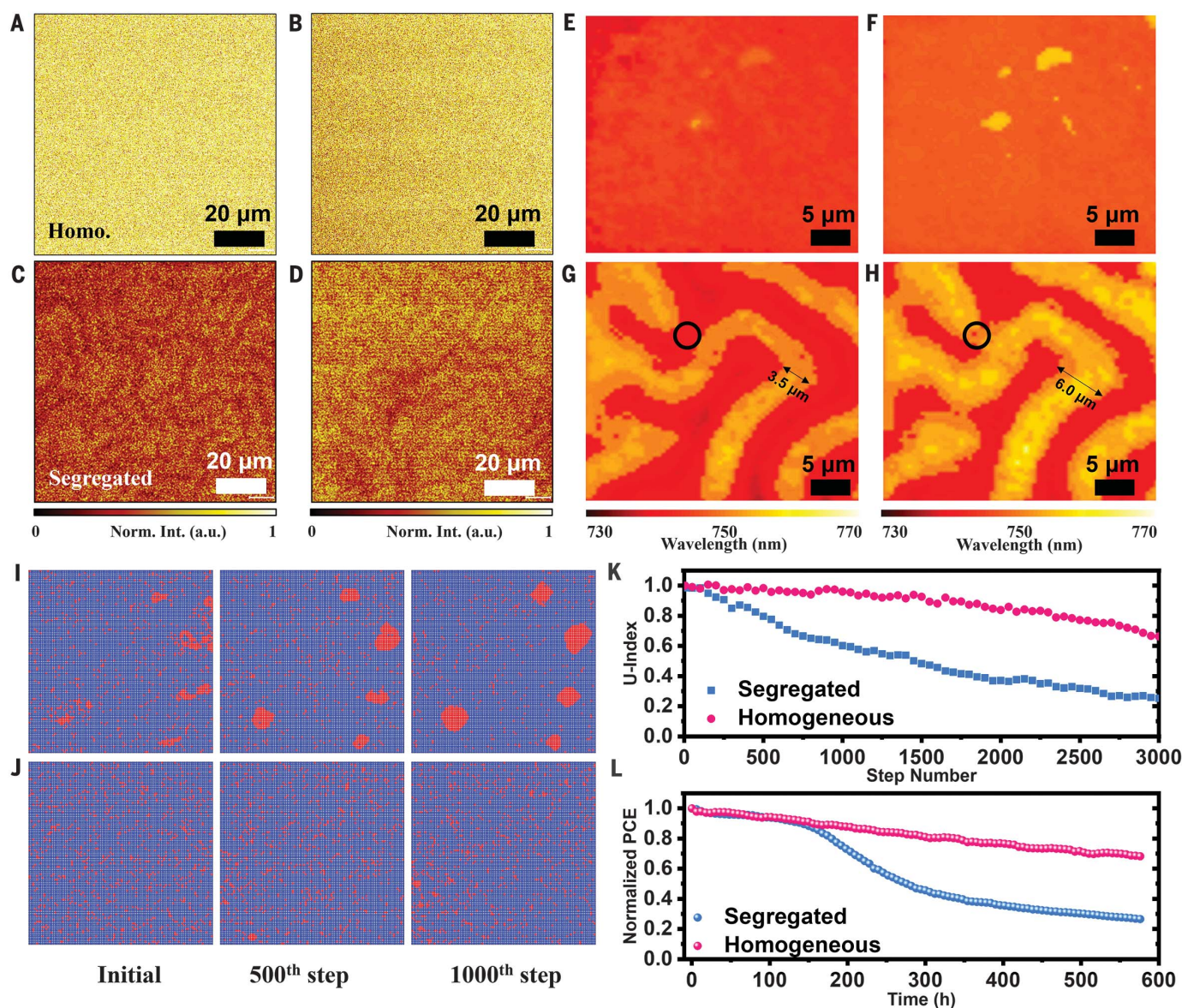


Fig. 4. Film homogeneity of mixed halide perovskite and the impacts. (A to D) 2D TOF-SIMS mapping of I^- in the mixed halide perovskite films (A) without and (C) with initial segregation and [(B) and (D)] perovskite films aged under continuous light illumination at $45^\circ \pm 5^\circ\text{C}$ in an N_2 -filled glovebox for 24 hours (B) without and (D) with severe initial iodide segregation. (E to H) In situ 2D PL mapping spectra (wavelength) evolution of perovskite films [(E) and (F)] without and [(G) and (H)]

with severe iodide segregation, under light illumination for 20 min in an ambient environment with relative humidity of 60%. (I and J) Simulation of segregation process of perovskite films in (I) an initially segregated distribution (ISP = 28%) and (J) an initially less segregated distribution (ISP = 3%). (K) U index evolution of red particles (Br) derived from simulation cases. (L) MPP tracking under 100 mW/cm^2 LED illumination in an N_2 -filled glovebox without encapsulation.

ion migration activation energy, depending on the cation properties and their local chemical environment. In a particular scenario for simulation, we simply normalized the migration activation energy for different cations from DFT or experimental results to obtain η_K ranging from 0 to 1. To provide a quantitative measure of the film homogeneity, we defined unsegregated index (U index) as the ratio of presently unsegregated Cs ions to the initially unsegregated ones. Regarding any case

of interest, the heavier segregation can be reflected on the smaller U index (Fig. 2, B and E).

The initial state of cation aggregation affected the phase segregation trajectories. We investigated phase segregation with different initial segregation percentages (ISPs), which stands for the ratio of initially segregated Cs to the total amount of Cs. The values of other core parameters are summarized in tables S1 and S2. The cation aggregation with an arbitrary ISP of 27% was simulated for clear demon-

stration (Fig. 2B, case I). After 200 steps, the U index decreased from 1.00 to 0.94. After 500 and 1000 steps, the U index dropped to 0.77 and 0.59, respectively, and heavier aggregation and enlarged domains were observed (movie S1).

We compared the initially more homogeneous film (case II, ISP of 2%) with the inhomogeneous one (case I) through simulation. After 200 steps, the U index for case II only dropped to 0.98 (Fig. 2C, middle left), in which

the aggregation is negligible as compared with the initial state. After 500 and 1000 steps, the U index remained at 0.95 and 0.88, respectively (movie S2). With linear regression, we obtained the U index decay rate of 0.012 per 100 steps for case II, whereas that for case I is 0.041 per 100 steps. The initial homogeneous film clearly exhibited slower segregation (Fig. 2, B and C, and fig. S8), which suggests a longer lifetime in the corresponding devices.

Increasing the activation energy of ion migration is proven to prolong device lifetime (10), which can be reflected as a lower η_K in our model. Simulations for the group of films with the same initial homogeneity (ISP of 27%) but decreased η_K (1, 0.8, 0.6, and 0.4) showed reduced decay rates of 0.041, 0.022, 0.017, and 0.007 per 100 steps in their linear decay regions, respectively (Fig. 2F, fig. S9, and table S2). The initial increase of U index could be observed in some cases, in which a large number of small segregation domains were dissolved to form larger ones (supplementary materials). It reveals the determinant impact of the initial state on the segregation dynamics, which were in good agreement with the experimental data reported elsewhere (23).

We also simulated the aggregation dynamics of ternary cation perovskites (fig. S10) and 1D and 3D fused perovskites (Fig. 2, D and E, and fig. S11). For the fused perovskites (case III), the immobile large cations created barriers between each sublattice and retarded cation migration. According to previous observations (25, 37), we reasonably assumed the η_K of large cations (yellow) to be zero. These boundaries retarded phase segregation (movie S3), and the decay rate further declined to 0.006 per 100 steps, which was just half of that of case II and in agreement with reported experimental data (37, 38). The stability enhancement in 1D and 3D perovskites is generally attributed to the prevention of moisture by large hydrophobic cations, but our findings suggest that retarded phase segregation is caused in part by partially immobilized cations.

Initializing film homogeneity and device performance

We modulated the Cs aggregation in FACs perovskite thin films with a two-step deposition process to introduce CsI when depositing the PbI_2 film in the first step (fig. S12) (39). The Coulomb interaction between the Cs^+ cation and the PbI_4^{2-} -based complex led to the formation of CsI– PbI_2 colloids in solution (fig. S13) and subsequent Cs-rich aggregates in the precursor film after thermal annealing at 70°C (40). The top-view mapping of Cs^+ on the PbI_2/CsI films that were characterized by TOF-SIMS (Fig. 3A) revealed the inhomogeneous distribution of Cs-rich domains on the micrometer scale. After the organic components were

added, the CsI/PbI_2 was converted to perovskite films when annealed at 150°C. The inhomogeneous Cs^+ distribution was still observed with TOF-SIMS mapping (Fig. 3B) and the PL measurements (fig. S14).

For less inhomogeneity, we modulated the Pb-solvent interactions by introducing a donor molecule to reduce the CsI – PbI_2 colloid size (41). Common Lewis bases (such as dimethyl sulfoxide) lead to strong coordination with Pb and form crystalline intermediates (42). To achieve a moderate coordination with Pb and disturb those crystalline intermediates, two weak donors, thiophene and selenophene, were separately added to the PbI_2/CsI precursor solutions to grow films. The 2D TOF-SIMS mapping of both films revealed a more homogeneous distribution of Cs than that of the reference sample (Fig. 3A and fig. S15 for selenophene and thiophene, respectively), although the selenophene was more homogeneous.

The intensity distribution analysis of CsI – PbI_2 film (Fig. 3A) showed a reduced FWHM for the selenophene-modified film (0.04 versus 0.08 for the reference), which indicates the smaller intensity difference of Cs at each scanning point (~ 0.1 by $0.1 \mu\text{m}^2$). In addition, the x-ray photoelectron spectroscopy (XPS) mapping (fig. S16) showed a narrow intensity variation in the modified film, which also demonstrated an enhanced film homogeneity. No phase structure difference was detected from the XRD patterns (fig. S17A). As expected, the resultant perovskite film (selenophene modified) was more uniform than the reference (Fig. 3B), with the FWHM of the perovskite peak from XRD patterns at 14.0° decreasing from 0.19 to 0.14 (fig. S17B). No obvious phase segregation was detected in the PL mapping measurement for the modified film (fig. S14).

We studied the interaction between selenophene and Pb in the precursor solution with several methods. Carbon nuclear magnetic resonance (^{13}C -NMR) revealed that the chemical shift of the α carbon in selenophene at 131.95 parts per million (ppm) moved downfield to 132.05 ppm, which is indicative of chemical bonding between selenium (Se) and PbI_2 (fig. S19). Extended x-ray absorption fine structure (EXAFS) spectra of the Pb L_3 -edge (fig. S20 and table S3) showed that the coordination numbers of Pb in Pb-I before and after the introduction of selenophene were 4.2 ± 0.6 and 3.6 ± 0.5 , respectively. The EXAFS results are not conclusive because of its limited sensitivity but are in line with the ^{13}C -NMR results, which is indicative of an interaction between Pb and Se in the precursor (43). Dynamic light scattering (DLS) showed that the mean colloidal size decreased from 622 nm in the reference to 552 nm (Fig. 3C). We argue that selenophene interacted with Pb to disturb the formation of large colloid clusters in the precursor. As compared with large clusters,

smaller clusters effectively diluted the aggregation of Cs, which mitigated the heterogeneity in the resultant film.

A series of devices, with the configuration of indium-doped tin oxide (ITO)/ $\text{SnO}_2/\text{Cs}_{0.1}\text{FA}_{0.9}\text{PbI}_{3/2,2',7,7'}$ -tetrakis[N,N-di(4-methoxyphenyl)amino]-9,9'-spirobifluorene (Spiro-OMeTAD)/Au, were fabricated by tuning the initial film homogeneity (fig. S22), and the current-voltage (J - V) curves of champion devices are shown in fig. S23 with the key parameters summarized in table S4. On the basis of the enhanced initial film homogeneity with selenophene, the device performance improved from 20.9 to 23.4% (0.08 cm^2 , average PCE from 20.1 to 22.6%). We observed improved photoelectronic properties in the resultant device with suppressed carrier recombination (fig. S24), lower trap density (fig. S25), enhanced mobility (fig. S25), and better charge extraction (fig. S26). With further optimization, the device with an active area of 1.00 cm^2 showed a certified PCE of 23.7% (reverse scan), with an open-circuit voltage (V_{oc}) of 1.14 V, a short-circuit current density (J_{sc}) of 24.7 mA per cm^2 , and a fill factor (FF) of 0.84 (Fig. 3D). The device was also subjected to continuous illumination by holding at 1.03 V and achieved a stabilized efficiency of 22.4% after 300 s in ambient atmosphere without encapsulation (fig. S27).

The absorber films with improved initial homogeneity (with selenophene) exhibited enhanced stability as compared with the reference film. We conducted in situ PL mapping measurements to the films during aging under thermal treatment. After 20 min of thermal aging in an ambient atmosphere at 80°C, the reference sample exhibited two emissive regions, of which the ones at $\sim 785 \text{ nm}$ grew larger (Fig. 3, E and F). This change was most likely caused by the formation of Cs-rich and Cs-poor domains, in which the local Cs-FA stoichiometry deviated from that in the pristine films. By contrast, the perovskite film with enhanced initial homogeneity exhibited a constant and uniform emission at 794 nm with retarded Cs aggregation. These results were confirmed by means of TOF-SIMS mapping (Fig. 3G) of films after aging for 144 hours. Scanning electron microscopy (SEM) images also revealed that the initially segregated film underwent substantial morphology changes after the thermal treatment, whereas no obvious changes were observed for the selenophene-treated film (fig. S28).

The homogeneous films also exhibited enhanced light stability. After aging under continuous illumination for 144 hours in an N_2 -filled glovebox, notable morphology changes occurred in the reference at the grain boundaries as shown in SEM images (fig. S29, A and B), and the δ -FAPbI₃ phase was identified by means of XRD (fig. S29, C and D). By contrast, no

obvious morphology change occurred on the homogeneous films, and we observed a retarded formation of δ -FAPbI₃. The 2D PL mapping measurements show that the PL peak of reference film exhibited blue shift from 790 to 785 nm with the emergence of nonphotoactive regions, possibly owing to the formation of δ -FAPbI₃. However, the film with improved initial homogeneity showed only slight changes of the peak position and distribution, which illustrates the enhanced light stability (fig. S30).

The DFT simulations suggested that the remaining selenophene can benefit the suppressing of Cs migration with increased activation energy (fig. S31). XPS data showed that after being stored in the ambient environment for 12 hours, the perovskite films still exhibited the symmetric peaks of Pb (II) at 138.5 and 143.4 eV that are attributed to Pb 4f_{7/2} and Pb 4f_{5/2}, respectively. It indicates that the film surface was protected from oxidation, which may have originated from the coordination of residual selenophene (fig. S32). However, the remaining amount is small according to the TOF-SIMS measurement (fig. S33), so the enhanced film stability could be mainly attributed to the increased initial film homogeneity.

Selenophene-treated perovskite devices were aged under various conditions according to the modified International Summit on Organic Photovoltaic Stability (ISOS) stability protocols (44) and presented enhanced stability than that of reference devices. The selenophene-treated perovskite device showed good stability without PCE degradation more than 2000 hours after being stored in the dark in an N₂ atmosphere (fig. S34). The devices that were aged at 85°C in N₂ in the dark (fig. S35) retained ~80% of their initial PCE after 1750 hours. When aged over 1-sun continuous illumination [light-emitting diode (LED) source, 100 mW/cm²] at open circuit, the devices could retain ~80% of their initial PCE after 960 hours (Fig. 3H and fig. S36). For the MPP test at 45° ± 5°C in an N₂ atmosphere, the device retained 91% of its highest PCE after 3190 hours. In addition, the MPP test was performed at elevated temperatures in the N₂ atmosphere, and the devices retained 91 and 89% of their highest PCE after 500 hours at 65° ± 5°C and 85° ± 5°C, respectively (Fig. 3J and fig. S37).

For mixed-halide perovskites, the main device issue has been the poor light stability. We fabricated FACsPb(Br_{0.13}I_{0.87})₃ of a bandgap of 1.66 eV with different initial halide segregation. We compared 2D TOF-SIMS mapping on the absorber films, which were subjected to continuous light illumination for 24 hours at 45° ± 5°C in an N₂-filled glovebox. The film without severe initial segregation that was fabricated under the protocol that is reported elsewhere (46) (supplementary materials, materials and methods) retained a homogeneous I⁻

distribution (Fig. 4, A and B). However, for the film with heavier initial segregation, the larger I⁻-deficient regions (~5 μm) were identified on the resultant film (Fig. 4, C and D).

In addition, we conducted in situ PL mapping as films were being aged under light illumination. Similar to cation segregation, the film with initially homogeneous halide distribution showed better stability under continuous illumination (Fig. 4, E and F). By contrast, the film with heavier initial halide segregation showed a marked change on the PL mapping spectra (Fig. 4, G and H, and fig. S38). We observed that the width of longer-wavelength regions (Fig. 4, G and H, yellow) was increased from ~3 to 6 μm. Moreover, the emission peak was clearly redshifted from 739 to 751 nm (Fig. 4, G and H, circled region), which indicates the fast halide segregation. The Schelling model simulation results are in good agreement with the experimental observation. As shown in Fig. 4, I to K, the segregated case (ISP = 28%), presented a decay rate of 0.040 per 100 steps, which is more than four times higher than that of the initially homogeneous film of 0.008 per 100 steps (ISP = 3%).

We further fabricated devices based on these two absorbers, which showed PCEs of 18.78% (homogeneous films) and 17.93% (reference), respectively, as shown in fig. S39. The mixed halide device with initially homogeneous halide distribution achieved an impressive operational stability during the MPP test at 45° ± 5°C in N₂, which retained 80% of its initial PCE after 340 hours (168 hours for the reference device). To date, this stability result is among the best device operational lifetimes (T₈₀) based on mixed halide perovskites (46). In addition, at an elevated temperature of 85° ± 5°C, the mixed halide perovskite device with improved homogeneity achieved a T₈₀ lifetime of 80 hours (fig. S40).

We have demonstrated that the Schelling model is a powerful tool to bridge theoretical analysis of perovskites at the atomic scale and macroscale observations of their phase separation and film degradation. From simulation and experiment results, we found that the initial film homogeneity, in terms of elemental distribution, has shown substantial influence on film and device stability. Benefiting from homogeneous films by tailoring precursor chemistry with selenophene, we developed high-performance devices based on mixed perovskites that achieved a long-term stability during the MPP test even at elevated temperatures.

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SUPPLEMENTARY MATERIALS

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Materials and Methods

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