

RESEARCH ARTICLE OPEN ACCESS

Maximum Stability Point Tracking Stabilizes Wide-Bandgap Mixed-Halide Perovskite Solar Cells

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Received: 21 April 2026 | **Revised:** 18 June 2026 | **Accepted:** 14 July 2026

Keywords: charge trapping | halide segregation | maximum stability point tracking | operational stability | wide-bandgap perovskites

ABSTRACT

Mixed-halide wide-bandgap (WBG) perovskites with bandgaps above 1.65 eV are essential top-cell absorbers for tandem photovoltaics. However, bromide-rich WBG perovskites suffer from photo-induced halide segregation, while its origin under operating conditions remains unclear. Here, we identify trapped-charge accumulation as a key factor triggering phase segregation during operation and develop a practical operational strategy to mitigate this process. Time-evolving photoluminescence and depth-profiling x-ray photoelectron spectroscopy reveal that trapped-charge accumulation accelerates ionic redistribution and halide segregation under illumination. Based on this insight, we introduce a maximum stability point tracking (MSPT) strategy that periodically relieves charge accumulation during device operation. WBG perovskite solar cells (PSCs) operated under MSPT retain approximately 90% of their initial power after 700 h of continuous operation, whereas devices operated under conventional maximum power point tracking (MPPT) rapidly degrade during the early stages of operation. Post-operational analyses further reveal that MSPT preserves the alloyed perovskite phase, whereas MPPT induces pronounced phase segregation. As an operation-stage strategy that mitigates charge accumulation without altering material composition, MSPT is compatible with existing material approaches and offers a route toward more stable perovskite photovoltaics. These findings provide an operational guideline for stabilizing WBG PSCs and advancing tandem photovoltaic technologies.

1 | Introduction

Metal-halide perovskite solar cells (PSCs) have advanced rapidly, and efforts are now focused on surpassing the performance limits of single-junction devices by moving toward tandem cell architectures [1, 2]. In such tandem configurations, mixed-halide wide-bandgap (WBG) perovskites ($E_g > 1.65$ eV) are commonly employed as the top cell absorber, and extensive research has been devoted to their development [3, 4]. These materials are particularly attractive for tandem photovoltaics because their

bandgap can be readily tuned by adjusting the halide composition (I^- and Br^-) and they are compatible with diverse fabrication processes [5, 6]. This application-driven demand has accelerated the development of bromide-rich mixed-halide perovskites as the primary route to realizing WBG compositions.

However, WBG perovskites face a fundamental challenge known as halide segregation, or photoinduced phase segregation [7, 8]. Under continuous illumination, nominally uniform I^-/Br^- perovskite alloys spontaneously separate into iodide-rich and

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bromide-rich regions [9, 10]. This process is manifested by splitting in photoluminescence (PL) spectra, degradation in absorption features, and progressive electronic inhomogeneity [11, 12]. The detrimental impact of halide segregation extends beyond optical spectral changes. In particular, the formation of iodide-rich, lower-bandgap domains creates carrier funneling and local recombination centers, effectively reducing the open-circuit voltage (V_{OC}) [13]. For that reason, WBG PSCs have shown more rapid degradation during actual operation than normal-bandgap (NBG) devices, which is mainly explained by the effect of photo-induced halide segregation [14]. It has been reported that the driving forces behind halide segregation may arise from a complex interplay of photoinduced thermodynamic effects, ionic migration, hole trapping, defect landscapes, and local structural heterogeneity [15–19]. However, the fundamental reason for halide segregation in operating WBG devices still seem debatable.

Meanwhile, maximum power point tracking (MPPT) is considered a prerequisite for efficient operation in most photovoltaic systems. In fact, this assumption originates from long-term stable devices such as crystalline silicon photovoltaics [20, 21]. Additionally, at the system level, where energy storage systems (ESS) and grid-interactive operation are considered, MPPT is no longer an ideal tracking strategy because the ESS inherently regulates the DC-link voltage and power flow [22, 23]. Therefore, enforcing strict MPPT can be counterproductive for photovoltaic systems whose long-term stability is insufficiently secured. If a tracking way improves operational stability by suppressing halide segregation for WBG perovskite devices, it could substantially accelerate the commercial viability of such WBG perovskite technologies.

To identify the origin of halide segregation of WBG perovskites, we first studied phase segregation in WBG perovskites using a half-device architecture, systematically comparing its evolution under continuous light illumination in the presence and absence of the SnO_2 electron transport layer (ETL), as well as under inert conditions or under oxygen-containing ambient conditions. We experimentally found that trapped holes induce rapid halide segregation even in the absence of atmospheric species, although the process is reversible to some extent [24]. Furthermore, a synergetic effect of oxygen caused irreversible degradation due to iodide loss as a consequence of severe halide segregation. These observations imply that trapped charges would be fundamental origins of halide segregation and further lead to device degradation when combined with air exposure. Therefore, suppressing charge trapping within the perovskite layer during operation is crucial for improving device stability. It is interesting to note that the trapped charges were previously revealed as a fundamental cause of both extrinsic and intrinsic degradations of NBG PSCs [25, 26].

Building on these observations, we finally developed a new stability-point tracking approach, termed maximum stability point tracking (MSPT), which mitigates trapped charges and enables more reliable long-term energy harvesting compared with conventional MPPT. Utilizing MSPT, the WBG devices retained approximately 90% of their initial power over 700 h without encapsulation. Finally, comparative analyses after MPPT and MSPT operation revealed that MSPT substantially suppresses

phase segregation and results in a lower trap density. These findings offer a practical guideline for improving operational stability not only in intrinsically instability-prone WBG PSCs but also in other photovoltaic systems requiring sustained, long-term performance.

2 | Results and Discussion

2.1 | Optimization of Photovoltaic Performance in WBG PSCs

Before examining our new operation protocol of MSPT for WBG PSCs, we first prepared a robust platform of WBG PSCs for systematic stability analysis. As illustrated in Figure 1a, the WBG PSCs were fabricated in an *n-i-p* configuration, employing a perovskite absorber with a bandgap of 1.67 eV and a composition of $\text{Cs}_{0.05}(\text{FA}_{0.77}\text{MA}_{0.23})_{0.95}\text{Pb}(\text{I}_{0.77}\text{Br}_{0.23})_3$. The ETL was optimized by improving its coverage on the FTO substrate through the use of chemical bath deposition (CBD) of SnO_2 . As shown in Figure 1b, devices based on CBD SnO_2 exhibited similar V_{OC} and short-circuit current density (J_{SC}) compared to those employing colloidal SnO_2 , but achieved a significantly higher fill factor (FF), resulting in a champion PCE of 21% which is comparable to the state-of-the-art performance reported for *n-i-p* WBG PSCs (typically 21%–22% [27–29], with only a limited report exceeding 23%) [30]. The cross-sectional SEM image in the inset of Figure 1b confirms that the CBD SnO_2 devices form a uniform WBG perovskite layer with a thickness of approximately 350 nm. It was verified that the external quantum efficiency (EQE) spectrum in Figure 1c shows good agreement between the integrated photocurrent and the *J-V* derived current density, while also confirming the optical bandgap. The device performance statistics presented in Figure 1d,e indicate that the CBD devices exhibit improved reproducibility, attributable to the enhanced coverage and quality of the SnO_2 layer. Following ETL optimization, we implemented a surface passivation strategy using a 2D perovskite layer to stabilize the WBG perovskite surface. Guided by previous reports that fluorinated 2D perovskites impart superior moisture stability, we applied this 2D passivation approach and confirmed its effectiveness for both colloid- and CBD-based ETLs, as shown in Figure 1d,e and Figure S1 [31, 32]. Notably, improvements in V_{OC} and FF were observed for both device types. As a result, the CBD-based devices consistently showed the highest overall performance due to the combined effects of ETL enhancement and 2D passivation, and exhibited negligible hysteresis between forward and reverse scans (Figure S2).

Utilizing these optimized WBG PSCs with enhanced photovoltaic properties, we next evaluated their operational stability under conventional MPPT conditions following standardized ISOS-L-1 protocols [33], as shown in Figure 1f. Despite the performance improvements achieved through ETL and surface engineering, the CBD-based WBG devices were unable to maintain their initial efficiency for more than 2 h under MPPT operation, exhibiting rapid degradation. Notably, the enlarged view of the first 3 h of operation (inset, Figure 1f) reveals that the device fails to maintain T_{90} and undergoes a rapid decline in performance shortly after the start of MPPT operation. These results highlighted the inherent instability of WBG PSCs under conventional operating

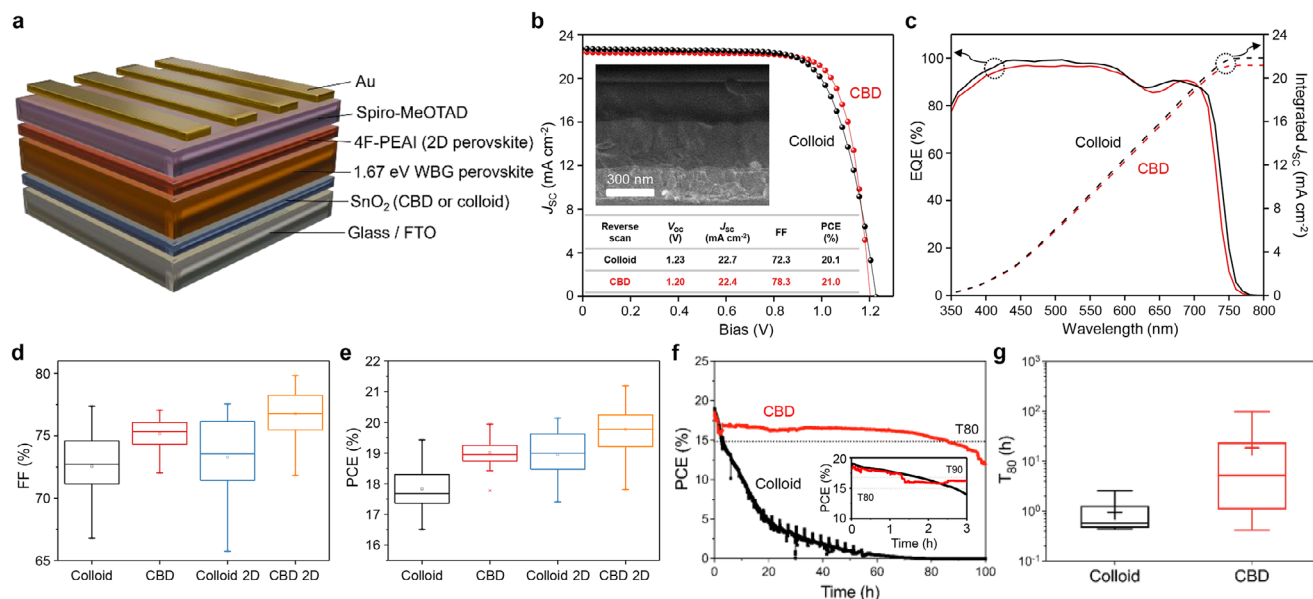


FIGURE 1 | (a) Schematic illustration of the *n-i-p* structured WBG PSCs used in this study. (b) *J-V* curves of WBG PSCs fabricated with colloidal and CBD SnO_2 ETLs, along with cross-sectional SEM images (inset). The table summarizes the photovoltaic parameters of the champion devices measured under reverse scan. (c) EQE spectra of colloidal and CBD SnO_2 -based WBG PSCs (left axis) and the corresponding integrated J_{sc} values (right axis). (d,e) Histograms of FF and PCE for WBG PSCs employing colloidal or CBD SnO_2 with 2D perovskite passivation. A total of 16 devices were fabricated and analyzed for each condition. (f) MPPT stability curves of WBG PSCs employing colloidal and CBD SnO_2 ETLs, under continuous illumination, showing the evolution of PCE over time. The inset presents an enlarged view of the initial 3 h of operation, clearly demonstrating that the device fails to maintain 90% of its initial performance within 2 h. (g) Box plots of the time required to retain 80% of the initial efficiency (T_{80}) for devices based on colloidal and CBD SnO_2 ETLs (logarithmic scale). The statistical distribution was obtained from 10 independent devices for each condition.

conditions and motivated further investigation into the underlying degradation mechanisms.

2.2 | Light-Induced Halide Segregation Driven by Charge Carriers in WBG Perovskites

In Figure 2, we systematically analyzed the factors responsible for the poor operational stability of the WBG PSCs described earlier, despite their improved photovoltaic performance. Our goal was to clarify the origin of light-induced halide phase segregation. Figure 2a–c presents the behavior of halide segregation under nitrogen atmosphere. To examine the influence of the ETL, we employed a half-device structure and compared WBG perovskite films on colloidal and CBD-processed SnO_2 , along with a control sample consisting of a bare glass substrate coated only with the WBG perovskite layer.

As shown in Figure 2b,c and Figure S3a, PL measurements taken after 96 h of continuous illumination reveal a clear red shift in all three samples, confirming the occurrence of halide segregation [7, 8]. However, the degree of red shift was significantly more pronounced in the samples incorporating ETL, indicating that charge carriers play a critical role in driving the segregation process. In the presence of ETL, holes are blocked at the interface, leading to hole carrier localization. These trapped holes more effectively attract iodide anions, which in turn accelerates halide segregation, which agrees with previous reports [19, 28, 30]. Between the two ETL types, the colloidal SnO_2 sample exhibits more severe segregation than the CBD SnO_2 sample, which can be attributed to its poorer uniformity. This nonuniformity

further enhances charge carrier localization even in direction parallel to the interface, thereby promoting halide segregation [34]. These findings are further supported by the time-resolved photoluminescence (TRPL) measurements shown in Figure S4. As the illumination time increased and the PL peak red-shifted due to the formation of iodide-rich domains, the carrier lifetime was also observed to increase, consistent with the accumulation of low-bandgap regions during halide segregation. To further investigate the reversibility of segregation, we examined recovery behavior after storing the samples under nitrogen in the dark for 24 h. PL measurements showed that all samples exhibited a shift back toward their original peak positions, demonstrating partial recovery. However, consistent with the trends observed during illumination, the sample without ETL recovered most closely to its initial state, whereas the colloid-based ETL displayed noticeable peak broadening compared to the CBD sample. These observations reinforce that both ETL presence and its interfacial quality strongly influence the degree and reversibility of halide segregation. This trend is visually supported in Figure S5, and the absorbance spectra in Figure S6 further indicate that films on colloidal SnO_2 experience such severe degradation that the perovskite phase is largely lost, in agreement with the PL results.

To further elucidate this behavior, we investigated halide segregation under illumination in oxygen-containing ambient air rather than in a nitrogen atmosphere (Figure 2d–f). As schematically illustrated in Figure 2d, the presence of oxygen is expected to accelerate iodide oxidation, leading to more severe iodine loss from the perovskite film. To isolate the effect of oxygen, both environments were maintained at identical relative humidity (1%) and temperature (25°C), ensuring that the only variable

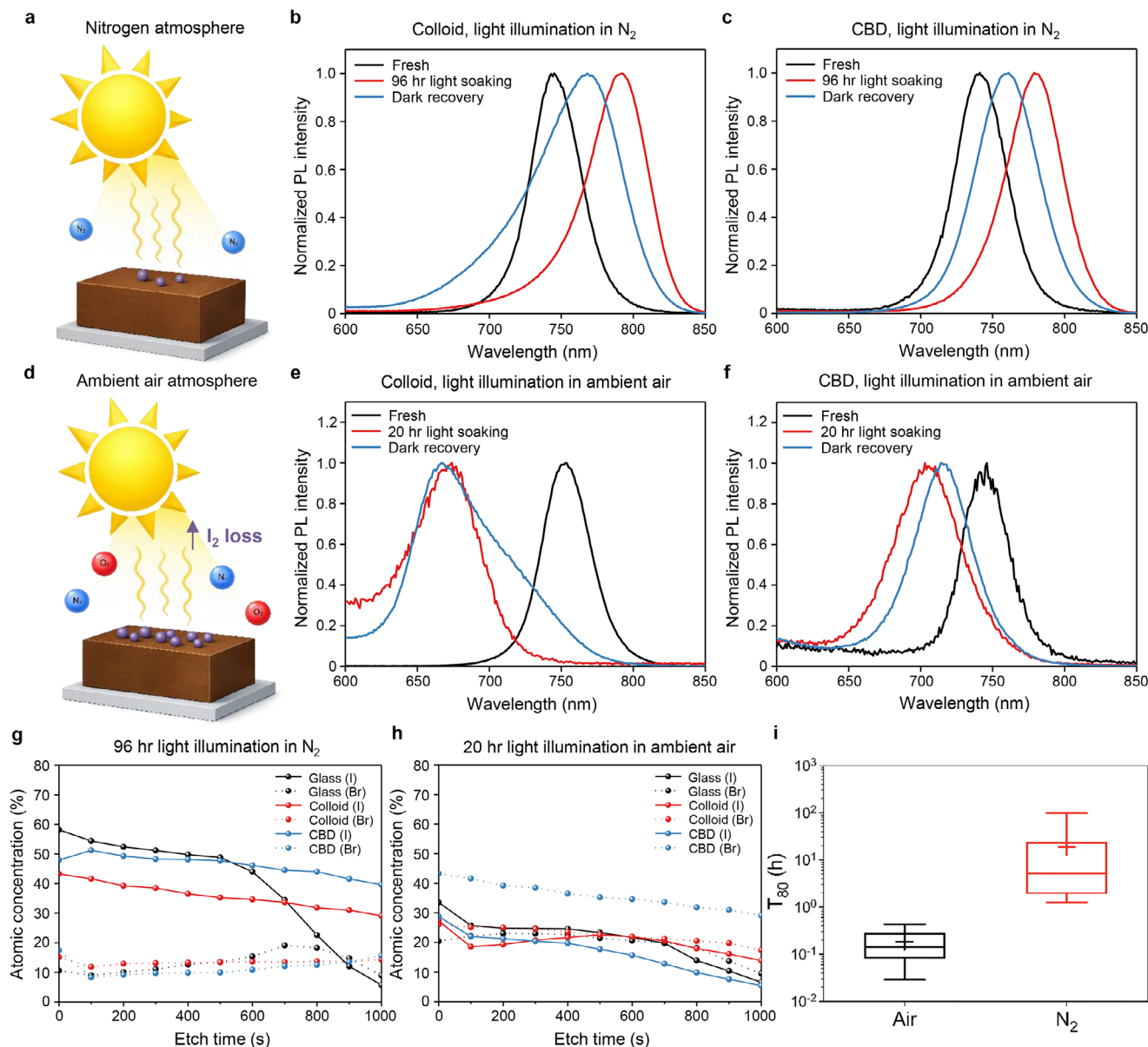


FIGURE 2 | (a) Schematic illustration of light illumination under 1 sun in a nitrogen atmosphere using a half-device configuration, indicating that iodine loss induced by halide segregation is not significant under nitrogen conditions. (b,c) PL spectra of half-device structures employing (b) colloidal and (c) CBD SnO₂ ETLs, measured in the pristine state, after 96 h of light-induced degradation under a nitrogen atmosphere, and after 24 h of recovery in the dark, to investigate light-induced halide segregation. (d) Schematic illustration of light illumination under ambient air conditions containing oxygen, highlighting the more pronounced iodine loss compared to that under a nitrogen atmosphere. (e,f) PL spectra of half-device structures with (e) colloidal and (f) CBD SnO₂, measured in the pristine state, after 20 h of light-induced degradation under ambient air, and after 24 h of dark recovery. (g,h) XPS depth profiling results obtained after light-induced degradation for (g) 96 h under a nitrogen atmosphere and for (h) 20 h under ambient air conditions, used to verify iodine loss associated with halide segregation. (i) Comparison of the operational lifetime (T₈₀) of WBG PSCs under ambient air and nitrogen, highlighting the accelerated degradation under oxygen-containing conditions.

was the presence or absence of oxygen. When the illumination atmosphere was switched to ambient air, the PL results revealed that halide segregation progressed much more rapidly, and in fact, the PL peak exhibited an unexpected blue shift after only 20 h of illumination (Figure 2e,f; Figure S3b). Consistent with the nitrogen-environment results, samples integrated with ETL showed a more severe shift, with the colloid-based ETL exhibiting the most pronounced degradation. This blue shift arises because iodide oxidation is significantly accelerated in the presence of oxygen, leading to the escape of iodine species from the film

and the development of bromide-rich domains [35]. Due to the substantial loss of iodide, the extent of dark recovery after storage was considerably reduced under oxygen-containing conditions compared to the nitrogen environment, indicating that the degradation becomes largely irreversible once iodide depletion occurs.

To further confirm the halide segregation described above, we examined the extent of iodide loss under each atmospheric condition using x-ray photoelectron spectroscopy (XPS) depth

profiling. In the nitrogen environment, as shown in Figure 2g and Figure S7a, all three samples exhibited nearly identical iodide and bromide concentrations in their fresh states. Even after 96 h of illumination, the overall halide composition remained largely unchanged, except for a slight reduction in iodide content for the colloid-based sample, which is consistent with its more pronounced degradation.

In contrast, when illumination was carried out in oxygen-containing ambient air, the XPS profiles revealed severe iodide loss, with the iodide concentration becoming nearly comparable to that of bromide (Figure 2h). Consistent with these results, XPS depth profiling of metallic Pb, a well-established indicator of perovskite degradation, showed the largest increase in the samples illuminated under ambient air (Figure S7b).

We further examined whether such atmosphere-dependent halide segregation directly impacts device stability. As shown in Figure 2i, PSCs operated under ambient air exhibited a markedly reduced T_{80} compared to those under nitrogen. This rapid lifetime decay is consistent with enhanced charge trapping and oxygen-assisted iodide oxidation, which synergistically accelerate irreversible halide segregation. Collectively, these results demonstrate that oxygen dramatically promotes phase segregation and the accompanying iodide loss, leading to substantially more severe degradation than under nitrogen. Accordingly, the extent and irreversibility of halide segregation in WBG perovskites are strongly governed by charge trapping and the illumination atmosphere, with oxygen playing a decisive role in driving irreversible decomposition.

2.3 | Strategy for Stabilizing WBG PSCs, Maximum Stabilized Point Tracking (MSPT)

As revealed from film degradation test in Figure 2, it is inevitable to completely prevent accumulation of photo-generated charges at heterojunctions. The conventional MPPT approach for PSCs allows sufficient charge accumulation to induce splitting of quasi-Fermi levels and develop internal electric potential, which may lead to strong effects of halide segregation. Our approach starts from the fact that a tracking algorithm does not need to be always conventional MPPT especially where degradation speed limits efficiencies over time. As mentioned in Introduction, for example, ESS coupled solar panels should be regulated for best battery-charging conditions, which means that strict MPPT is not required for real-system.

In this respect, when a device has ten-year scale lifetime as for c-Si photovoltaics, PCEs are assumed to be constant over time. Then, MPPT in principle offers maximized energy harvesting with consideration of time as shown in Figure 3a. On the other hand, if a solar cell undergoes obvious degradation within a year, MPPT will not be the best way to harvest much energy as PCEs decay. Accordingly, stability-limited operation necessitates the introduction of a new tracking metric that explicitly accounts for device degradation and maximizes the harvested energy (not instantaneous power). In this work, we assume that degradation follows a single-exponential decay governed by the degradation rate, α , and construct a theoretical model based on this assumption. The resulting metric derived from this framework is termed

the stability metric, $S(V)$, and is expressed as follows:

$$S(V) = \frac{V \cdot J(V)}{\alpha} \quad (1)$$

where V and J denote applied voltage and the corresponding current density, respectively (Figure 3b and detailed explanation in Note S1). Here, α is experimentally found to be approximately proportional to the applied voltage, V , as shown in Figure S8 [36]. Within an equivalent circuit framework, the trapped charge was modeled by the effective interfacial capacitance C , in which α was estimated to scale as $\alpha \propto Q_{\text{trap}} \approx CV_{12}$ [37]. Since V_{12} can be expressed as $V + JR_s$, the stability metric is given by:

$$S(V) = \frac{V \cdot J(V)}{V + J(V) \cdot R_s} \quad (2)$$

where R_s is series resistance. Based on Equation 2, our model can theoretically suggest a specific, real-time point just using basic photovoltaic characteristics extracted by J - V sweeps.

To confirm the effectiveness of the suggested stability metric, we measured energy harvested from WBG devices depending on applied bias voltage as depicted in Figure 3c. It is interesting to observe that harvested energy per hour of WBG devices tested for the first 10 h follows the trend of stability metric ($S(V)$, green curve in Figure 3c) at higher biases, while it is rather consistent with the power metric (red curve in Figure 3c) at lower biases. Extending the testing period is expected to yield results that more closely match the model predictions. Hereafter, we define the peak of the stability metric curve as the maximum stability point (MSP).

Based on the results, we devise the MSPT method that explicitly incorporates device degradation dynamics into the bias selection process as shown in Figure S9. In this algorithm, J - V sweeps and harvested power as a function of time were recorded at fixed time intervals of 3 h, thereby continuously updating a new applied voltage for maximizing stability. (Figure S10) The optimal operating voltage (V_{MSP} , hereafter) is mathematically defined as the point satisfying $\frac{dS(V)}{dV} = 0$, corresponding to the maximum of the stability metric curve. As expected, V_{MSP} is always located slightly lower than V_{MPP} . Simply, this algorithm identifies the point that achieves the longer lifetime than MPPT but without a significant power loss. Furthermore, abrupt variations of the operational voltage are known to induce accelerated degradation and interfacial damage in PSCs [38, 39]. To prevent such bias-induced stress, the MSPT algorithm constrains voltage updates within a safe ramping window, ensuring $|\Delta V| < 0.15$ V during tracking.

Figure 3d shows the strong efficacy of MSPT when applied to WBG PSCs by exhibiting dramatically improved survival rates of T_{80} . The survival rate is defined as the fraction of devices retaining more than 90% of their initial efficiency as a function of elapsed time. These stability results, obtained under standardized ISOS-L-1 testing conditions, confirm that applying a voltage approximately 0.1 V lower could maximize T_{80} . These results demonstrate that bias optimization based on trap-controlled degradation kinetics effectively suppresses trap-driven halide segregation and extends operational lifetime. Unlike conventional MPPT, which maximizes instantaneous power output, MSPT

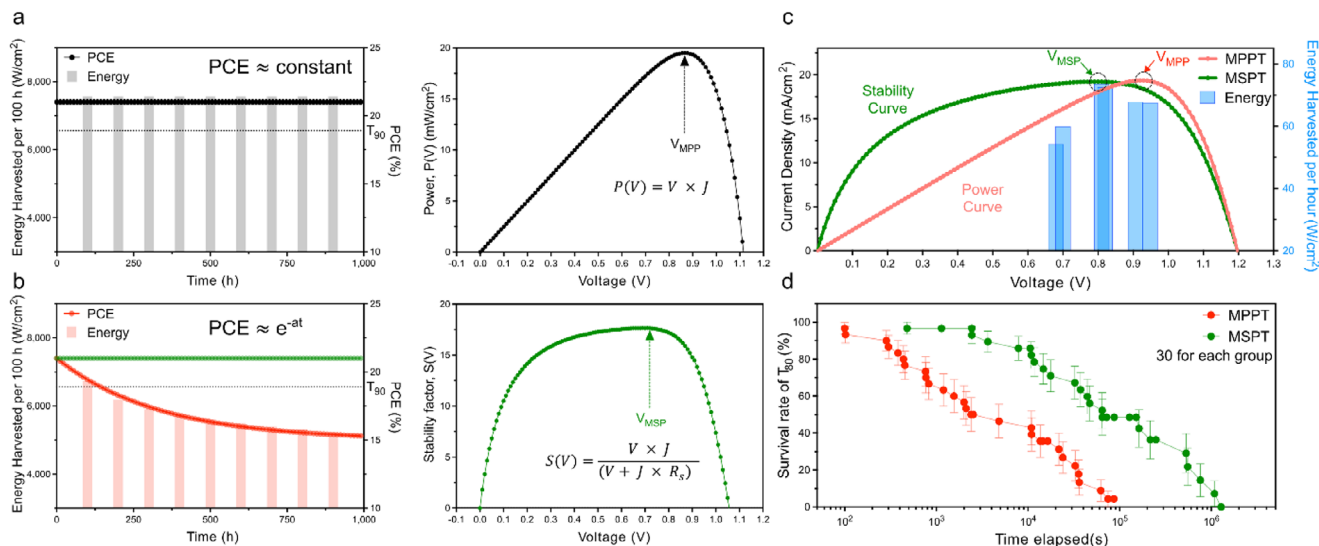


FIGURE 3 | (a) Schematic illustration of photovoltaic performance of an ideal device with zero degradation, exhibiting a constant PCE during prolonged operation. (left) an ideal bias point for operation to harvest maximized energy, which is corresponding to a standard MPPT (right), (b) Expected photovoltaic performance of an actual device with a constant degradation rate (α) under prolonged operation (left) and a stability metric curve ($S(V)$) as a function of voltage bias voltage (middle). The maximum point of the curve (denoted as V_{MSP} , MSP indicates maximum stability point) suggests the highest total harvested energy in consideration of degradation effects. (c) Stability curve vs power curve of a measured $J-V$ curve of a WBG PSC, suggesting the V_{MSP} point 0.1 V lower than its V_{MPP} . When degradation effects considered, averaged energy harvesting (skyblue bars) shows the highest value for the case of device operation at V_{MSP} . (d) Survival rates of T_{80} for a total of 30 sets of devices, showing an approximately one-order-of-magnitude enhancement in operational lifetime under MSPT compared with MPPT.

explicitly considers the trade-off between power generation and voltage-dependent degradation kinetics to maximize long-term harvested energy under degradation-limited operation.

2.4 | Confirming Suppression of Halide Segregation via MSPT

To assess the efficacy of the MSPT method, we performed spatially resolved micro-photoluminescence (μ -PL) mapping on three device samples: fresh, MPPT-operated, and MSPT-operated devices, respectively. (Figure 4a–c) The fresh device exhibited a peak position at 762 nm, confirming its WBG characteristic. (Figure 4a) After 150 h of MPPT operation, the μ -PL map revealed a pronounced red shift accompanied by a broadened wavelength distribution, indicating that iodide-rich domains were formed as a result of halide segregation as shown in Figure 4b. It also led to phase inhomogeneity across the WBG perovskite. Consistently, the corresponding histogram (Figure 4b) exhibits a significant broadening of the peak distribution (increased full width at half maximum (FWHM)), reflecting enhanced compositional fluctuation and spatially non-uniform phase segregation. In stark contrast, the μ -PL mapping after 150 h of MSPT operation showed a narrow, single-peaked wavelength distribution with a slight blue-shift (Figure 4c). The narrower histogram in Figure 4c further confirms suppressed wavelength dispersion and improved compositional homogeneity under MSPT operation. These observations confirm that MSPT effectively suppresses compositional segregation even for continuous long-term operation of 150 h. In addition to the wavelength distribution, spatially resolved PL mapping (Figure S11) reveals that devices operated under MSPT retain higher average emission intensity compared to MPPT-operated devices. Considering that PL intensity correlates with

radiative recombination efficiency, this result is consistent with suppressed defect formation and reduced non-radiative recombination under MSPT operation. Figure 4d–f consistently identify that halide segregation can be suppressed by our MSPT operation. The MSPT-operated device largely preserved the characteristic diffraction peaks of the perovskite phase, closely resembling the fresh reference. In contrast, the MPPT-driven device exhibited clear peak separation around both 28.5° and 32° of scattering angles. Those are evidence of halide segregation as a result of the MPPT operation. Strikingly, the MSPT operation led to suppression of halide segregation, thereby improving operational stability. It should be noted that the observed structural changes may originate from multiple ionic redistribution processes within the perovskite lattice, including not only halide migration but also A-site cation migration. Since ionic migration in perovskites is generally facilitated by trapped-charge accumulation and the associated local electric fields, reducing charge accumulation through MSPT may also potentially influence A-site cation migration [40]. However, A-site cations generally exhibit higher migration barriers than halide ions and are therefore expected to be less mobile under operating conditions [41]. Furthermore, mixed FA/MA compositions have been reported to exhibit even higher cation migration barriers due to cooperative interactions between the two organic cations [42]. Therefore, halide migration is expected to remain the dominant ionic process governing phase segregation in the present mixed-cation mixed-halide perovskite system.

We further advanced the MSPT algorithm to achieve fully automatic tracking independent of time constraints. To ensure a fair and accurate comparison, two identical pixels were integrated into a single device, on which MPPT and MSPT were operated under standardized ISOS-L-1 protocols for more than 800 h as

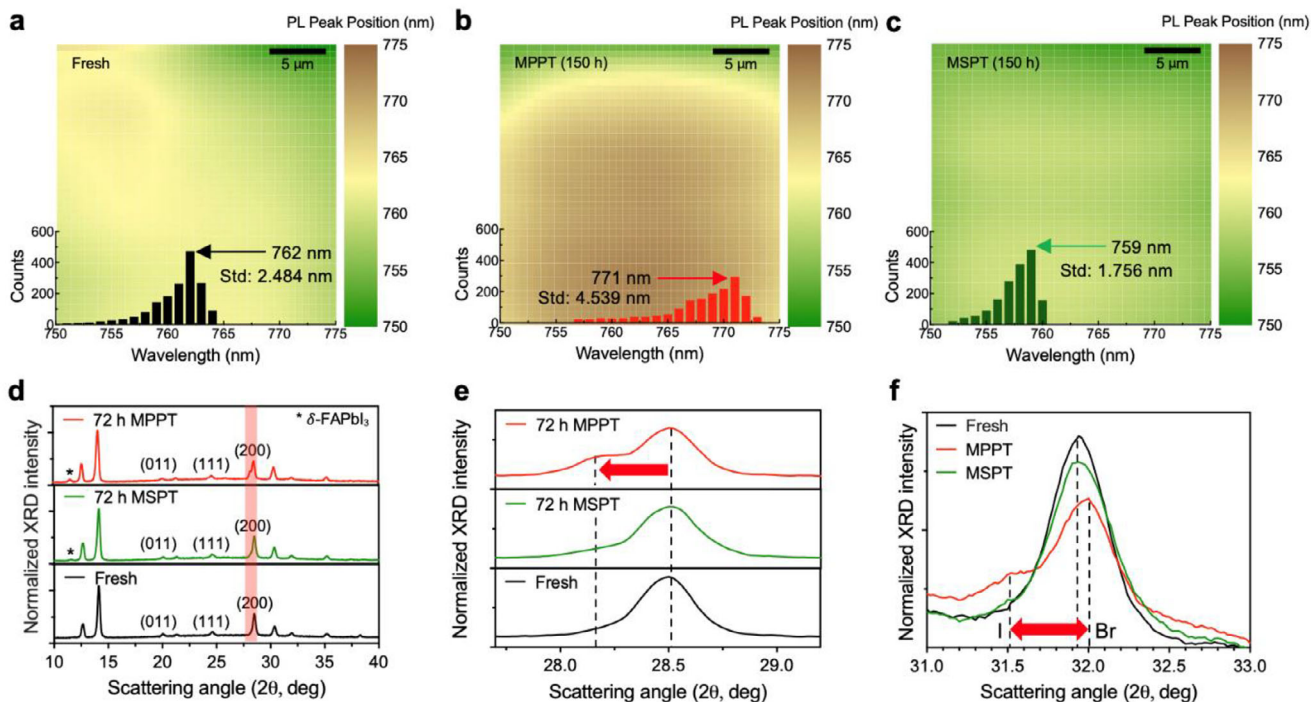


FIGURE 4 | Spatially resolved micro-photoluminescence mapping (top) and histograms of peak wavelength (bottom) of (a) fresh device, (b) 150-h MPPT-operated device, and (c) 150-h MSPT-operated device. These observations indicate that the MPPT operation led to red-shifted PL emission due to halide segregation whereas, the MSPT suppressed halide segregation by mitigating trapped charges. (d) X-ray diffraction patterns of the three devices, the fresh (blue), the 72-h MPPT-operated device (red), and the 72-h MSPT-operated device (green). magnified XRD patterns at 28.4° (e) and 32.0° (f) both corresponding to the (200) plane, showing halide segregation only in the case of MPPT operation.

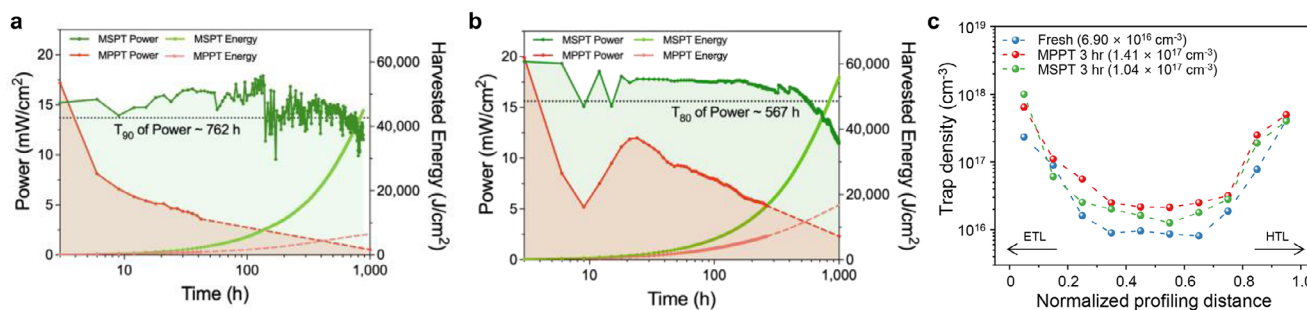


FIGURE 5 | (a, b) Long-term operational stability and cumulative harvested energy of devices operated under MPPT and MSPT conditions under continuous one-sun illumination. In (a), the MSPT-operated device retained 90% of its initial power for more than 750 h, whereas the MPPT-operated device underwent rapid burn-in and complete degradation within 100 h. In (b), the MSPT-operated device retained 80% of its initial power for more than 550 h under identical testing conditions. The corresponding cumulative harvested energy curves demonstrate that MSPT enables greater total energy generation through enhanced operational stability. In both panels, the dotted lines indicate projected degradation trends obtained by extrapolation of the measured data. (c) Trap density profiles extracted from deep-level capacitance profiling (DLCP) for the control device and devices operated for 3 h under MPPT and MSPT conditions.

shown in Figure 5a,b. The most stable pixel retained 90% of the initial power until over 750 h of continuous operation under one sun illumination, while the simultaneously MPPT-operated device showed complete degradation even before 100 h of operation. In a separate but identically tested sample (Figure 5b), the MSPT-operated device retained 80% of its initial power for approximately 567 h (T_{80}), again demonstrating substantially enhanced operational stability compared with MPPT. The cumulative harvested energy curves further highlight the practical

advantage of MSPT (Figure 5a,b). The rapid degradation under conventional MPPT resulted in a substantially lower cumulative energy yield, whereas MSPT enabled greater total harvested energy through enhanced operational stability. The corresponding PCE evolution data are provided in Figure S12. Other sets of stability test can be found in Figure S13, which consistently shows dramatic enhancement of device lifetime. To further examine whether the enhanced performance of MSPT originates solely from the update frequency, MPPT operation was additionally

evaluated using update intervals of 3 and 6 h together with conventional perturb-and-observe (P&O) operation (Figure S14). Introducing periodic updates improved both operational stability and cumulative harvested energy compared with conventional P&O tracking, while shorter update intervals further enhanced device performance owing to the pronounced burn-in degradation occurring during the initial stage of operation. Importantly, even when the same 3 h update interval was employed, MPPT-operated devices still exhibited inferior operational stability compared with MSPT-operated devices, demonstrating that the enhanced performance of MSPT arises from operation at V_{MSP} .

Furthermore, MSPT improved the operational stability of both colloidal- and CBD-based WBG PSCs (Figure S15). The observation that MSPT remains effective even in the inherently more stable CBD-based devices suggests that the benefits of MSPT are additive to existing material- level stabilization approaches. MSPT was also successfully applied to *p-i-n* WBG PSCs, retaining 97.6% of the initial efficiency after 1000 h of continuous operation (Figure S16). Moreover, MSPT was extended to NBG PSCs, yielding a T_{90} lifetime exceeding 1500 h (Figure S17), whereas MPPT-operated devices stabilized at approximately 70% of their initial PCE after pronounced burn-in degradation. Collectively, these findings suggest that MSPT can function synergistically with existing stabilization strategies and effectively enhance operational stability across different device architectures and bandgaps, further supporting the notion that reducing charge accumulation mitigates degradation during device operation.

To complement these observations with device-level evidence, we assessed defect formation in MPPT- and MSPT-operated devices using deep-level capacitance profiling (DLCP), as shown in Figure 5c [43]. The depth-resolved trap density profiles reveal a pronounced increase in trap density across the device following MPPT operation. Compared with fresh devices, the trap density increases substantially both near the transport layers and within the perovskite bulk. Such increase in trap density is consistent with accelerated degradation driven by sustained high-bias operation. In contrast, MSPT operation markedly suppresses trap generation throughout the absorber layer. Compared to the MPPT-driven device, the MSPT-driven device exhibits a substantially lower trap density across the entire profiling depth, including both the bulk perovskite region and the ETL/HTL interfaces. The extracted average trap densities are $6.90 \times 10^{16} \text{ cm}^{-3}$ for the fresh device, $1.41 \times 10^{17} \text{ cm}^{-3}$ after 3 h of MPPT operation, and $1.04 \times 10^{17} \text{ cm}^{-3}$ after 3 h of MSPT operation, clearly demonstrating that stability-oriented driving significantly mitigates defect formation even at early operation times [44]. Importantly, the reduced trap density under MSPT provides a quantitative basis for the preserved photoluminescence intensity, prolonged carrier lifetime, and suppressed compositional segregation observed in Figure 4a–c. By limiting the accumulation of recombination-active defects in both the bulk and interfacial regions, MSPT fundamentally alters the defect evolution pathway of WBG PSCs, thereby sustaining optoelectronic integrity and enabling the enhanced operational stability and lifetime demonstrated in Figure 3. To further assess the reversibility of degradation following prolonged MSPT operation, devices operated under MSPT for 1000 h were stored in a dark nitrogen environment,

and their steady-state PL spectra were subsequently measured (Figure S18). The PL peak position exhibited substantial recovery toward that of the fresh device after dark storage, indicating that a reversible component of halide segregation remains even after extended MSPT operation.

For broader context, we compared the operational stability achieved in this work with previously reported single-junction mixed-halide WBG PSCs (1.65–1.71 eV) employing an *n-i-p* architecture under continuous MPPT operation (Figure S19). Although testing conditions and evaluation protocols differ among studies, the comparison provides a representative benchmark for assessing device stability. The stability achieved in this work is comparable to those reported for similar *n-i-p* WBG PSCs under continuous operation. Overall, these results highlight MSPT as a broadly applicable operation-stage strategy that suppresses charge accumulation, delays degradation, and enhances long-term energy harvesting without requiring modifications to device composition or architecture.

3 | Conclusion

In this work, we investigated the operational instability of mixed-halide WBG perovskites, a major barrier to their use as top-cell absorbers in tandem photovoltaics. PL and depth-profiling XPS analyses revealed that trapped charges play a decisive role in initiating halide segregation, driving the formation of iodide-rich and bromide-rich domains even without external atmospheric species.

Guided by this mechanistic understanding, we introduced MSPT as a degradation-informed alternative to conventional MPPT that balances power generation and degradation to maximize long-term harvested energy. MSPT minimizes carrier accumulation and suppresses segregation-driven degradation, enabling WBG devices to retain 90% of their initial power over 700 h, while MPPT leads to rapid efficiency loss and strong phase separation. Post characterizations further confirmed that MSPT effectively maintains alloy uniformity and reduces trap density.

Overall, MSPT offers an effective operational approach that improves the durability of inherently unstable WBG perovskite absorbers. These results emphasize the importance of operation-mode engineering and offer a clear pathway toward durable, high-performance tandem photovoltaic technologies.

4 | Experimental Section

4.1 | Materials

ITO ($9.5 \Omega \text{ sq}^{-1}$) and FTO ($8 \Omega \text{ sq}^{-1}$) substrates were purchased from AMG and Asahi, respectively. Formamidinium iodide (>99.99%), methylammonium bromide (>99.99%), methylammonium chloride (>99.99%), and 4-fluoro-phenethylammonium iodide (4F-PEAI, >99.5%) were acquired from Greatcell Solar Materials. Lead iodide (99.999%) was obtained from Alfa Aesar. Colloidal SnO_2 solution (20 wt.% in H_2O) were purchased from Sukgyung AT. Lead bromide (>98.0%) and Me-4PACz (>99.0%) were obtained from Tokyo Chemical Industry Co. Spiro-OMeTAD

and Co(III) TFSI were purchased from Luminescence Tech. Tin(II) chloride dihydrate ($\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$, 98%), hydrochloric acid (HCl, 37 wt.% in water), thioglycolic acid (TGA, > 99%), urea (99.0–100.5%), N,N-dimethylformamide (99.99%), dimethyl sulfoxide (99.50%), chlorobenzene (99.8%), isopropanol (IPA, 99.5%), diethyl-ether (99.9%), acetonitrile (99.8%), 4-tert-butylpyridine (99.90%), Li TFSI (99.95%), and cesium iodide (99.999%) were purchased from Merck. Deionized (DI) water was acquired from Samchun. NiOx ink (2.5 wt.% in ethanol) was purchased from Nano Clean Tech. All the chemicals were used without further purification.

4.2 | Device Fabrication

Pre-patterned ITO and FTO substrates were cleaned by ultrasonication using detergent, acetone, isopropyl alcohol, and DI water for 15 min, consecutively. The cleaned substrates were then dried in an oven at 70°C. For the chemical bath deposition (CBD) of the SnO_2 layer, precleaned substrates were first masked at one edge with Kapton tape to protect the electrode contact area, followed by oxygen plasma treatment for 10 min. Subsequently, the substrates were immersed in a glass container filled with a solution composed of 600 mL of DI water, 1.575 g of $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$, 7.5 g of urea, 7.5 mL of HCl, and 150 μL of TGA, and reacted in an oven at 90°C for approximately 10 h. After the deposition, the substrates were thoroughly rinsed with DI water for 30 min and thermally annealed at 150°C for 1 h. For the colloidal SnO_2 deposition, substrates were UV ozone-treated for 20 min before the coating procedure. SnO_2 solution was prepared by diluting SnO_2 colloidal solution with DI water (1:4 of volume ratio). The SnO_2 solution were then spin coated at 4000 rpm for 30 s, followed by an annealing at 150°C for 30 min. The resulting SnO_2 -deposited substrates were treated with UV-ozone for 15 min to improve wettability prior to perovskite film coating. The WBG perovskite precursor solution was prepared with a composition of $\text{Cs}_{0.05}(\text{FA}_{0.77}\text{MA}_{0.23})_{0.95}\text{Pb}(\text{I}_{0.77}\text{Br}_{0.23})_3$ with an addition of 5 mol% MACl. The precursor solution concentration was 1.1 M and the solvent mixture consisted of DMF and DMSO at a 4:1 volume ratio. The WBG perovskite solution was spin-coated onto the SnO_2 layer at 4000 rpm for 20 s. 0.5 mL of diethyl-ether was dripped on the substrate at the beginning of 8 s as an antisolvent. The perovskite coated substrates were annealed at 100°C for 20 min. For 2D perovskite passivation, 1.0 mg of 4F-PEAI was dissolved in 1 mL of IPA. The 2D solution was processed using a dynamic spin-coating method; during spin coating at 5000 rpm for 20 s, the solution was dripped onto the substrate 8 s after the start of spinning. Immediately afterward, the hole transport layer (HTL) was deposited. The spiro-OMeTAD solution was prepared by dissolving 72 mg of spiro-OMeTAD in 0.8 mL of chlorobenzene, followed by the addition of 31.6 μL of 4-tert-butylpyridine, 18.4 μL of Li TFSI solution (520 mg of Li TFSI in 1 mL of acetonitrile), and 10 μL of Co(III) TFSI solution (375 mg of Co(III) TFSI in 1 mL of acetonitrile) as dopants. The HTL was deposited by spin-coating at 2500 rpm for 30 s. Finally, a 100-nm-thick Au electrode was deposited by thermal evaporation.

For normal-bandgap PSCs, the only modification was the perovskite precursor solution, which was prepared by dissolving 705.4 mg of PbI_2 , 240.8 mg of FAI, 33.8 mg of MACl, 1.4 mg of MABr, 4.4 mg of PbBr_2 in 900 μL of DMF and 112.5 μL of DMSO.

For the fabrication of the *p-i-n* devices, a NiOx layer was first deposited onto ITO substrates by spin-coating a 1:1 (v/v) diluted solution of NiOx ink and ethanol at 3000 rpm for 40 s, followed by annealing at 250°C for 30 min. Subsequently, Me-4PACz was dissolved in IPA at a concentration of 0.3 mg mL^{-1} and deposited onto the NiOx-coated substrates. After dispensing the solution and allowing it to rest for 10 s, the substrates were spin-coated at 3000 rpm for 30 s and then annealed at 100°C for 10 min to form the HTL. The WBG perovskite layer was deposited under the same conditions as those used for the *n-i-p* devices. Finally, C_{60} (20 nm) and BCP (6 nm) layers were sequentially deposited by thermal evaporation, followed by the deposition of a 100 nm Cu electrode to complete the device fabrication.

4.3 | Device Characterization

J-V curves were acquired using a solar simulator (Sol 3A 64023A, Oriel) equipped with an AM1.5G filter and a source meter (Keithley 2400). Illumination intensity of 100 $\text{mW} \cdot \text{cm}^{-2}$ was calibrated using a reference Si cell equipped with a KG5 window (91150-KG5, Newport). Scan voltage ranged from −0.1 to 1.3 V in forward scan and from 1.3 to −0.1 V in reverse scan. The scan rate was set at 100 $\text{mV} \cdot \text{s}^{-1}$. The active area of the device was defined using a metallic aperture mask (0.0625 cm^2). All *J-V* characterizations were conducted in a nitrogen glovebox at room temperature without encapsulation, unless otherwise specified. The EQE spectra were measured by using a quantum efficiency measurement system (IQE-200B, Oriel). An impedance analyzer (MFIA, Zurich Instruments) was utilized to perform drive-level capacitance profiling (DLCP) on the WBG PSCs. The capacitance of the cells was measured at a given 10 kHz frequency by changing DC and AC voltages from 0 to 1.3 V and 20 to 200 mV, respectively.

4.4 | Operational Stability Measurements

All operational stability measurements were conducted in accordance with the ISOS-L-1 protocol for PSCs, ensuring standardized evaluation under continuous one-sun illumination (AM1.5G, 100 $\text{mW} \cdot \text{cm}^{-2}$). LED-based solar simulator (Ossila, G2009B1) was installed inside an N_2 -filled glovebox for the measurement. Devices were measured in an unencapsulated state. The light intensity was calibrated at the device plane using a certified Si reference cell and meter (Newport 91150-KG5). For a direct and simultaneous comparison, isolated pixels within a single device were independently operated under MPPT and MSPT using two Keithley 2401 source meters under identical illumination conditions. The operating points for both MPPT and MSPT were updated every 3 h. All measurements were conducted at room temperature (25°C).

4.5 | Maximum Stability-Point tracking (MSPT) Algorithm

The MSPT algorithm was developed by considering the cumulative energy output under degradation-limited operation. The temporal evolution of the output power was approximated by

an exponential decay, $P(t) = P_0 e^{-\alpha t}$, where the degradation rate α reflects voltage-assisted trapping. Based on an equivalent circuit model, the trapped charge density was approximated as $Q = CV$, leading to the assumption that α scales proportionally with the trapped charge and thus with the operating voltage. Under this framework, the long-term extracted energy becomes inversely related to the degradation rate, motivating the definition of a stability metric $S(V) = \frac{VJ}{V+J R_s}$. The operating voltage was selected by satisfying the extremum condition $dS/dV = 0$. To avoid bias-induced stress and abrupt degradation, an additional voltage-limiting rule was incorporated into the tracking algorithm, constraining the voltage update step to $|\Delta V| < 0.15 V$. This restriction prevents sudden operational voltage shifts that are known to accelerate degradation in PSCs under dynamic bias conditions. A detailed derivation and algorithmic implementation are provided in Note S1.

4.6 | 2D Micro-Photoluminescence (μ -PL)

The μ -PL measurements were carried out using a 532 nm laser as the excitation source. The PL intensity and peak position were spatially mapped over an area of $20 \times 20 \mu\text{m}^2$ using a raster-scanning method. Spatially resolved μ -PL mapping was performed on three device states: fresh devices, devices operated under MPPT and devices operated under MSPT. The μ -PL mapping was conducted after 150 h of continuous operation for the MPPT- and MSPT-operated devices. The PL peak position and its spatial distribution were extracted from the μ -PL maps to assess changes in emission wavelength and spectral homogeneity across the device area.

4.7 | Film Characterization

XRD patterns were obtained using a SmartLab (Rigaku) by monochromatic 9 kW Cu-K radiation. A Cary 5000 UV-vis-NIR (Agilent) spectrometer was used to measure the absorption spectra. A FluoroMax-4 spectrofluorometer (Horiba) was used to measure steady-state PL and TRPL. A laser with a 376 nm wavelength was used as an excitation source for both steady-state PL and TRPL measurements. Surface and cross-sectional images of perovskite films and devices were obtained utilizing a FESEM Sigma 300 (Zeiss). XPS depth profiling was performed utilizing an Axis Supra (Kratos) system with a monochromatic Al K α source (1486.6 eV), and Ar⁺ ions (5 keV) were used for etching.

Acknowledgements

This work was supported by the Global Frontier R&D Program of the Center for Multiscale Energy Systems and the National Research Foundation (NRF) under the Ministry of Education, Science and Technology, Korea (RS-2023-00279529 and RS-2023-00282896). Additional support was provided by the Institute of Advanced Machines and Design and the Institute of Engineering Research at Seoul National University.

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

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

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

Supporting File: aenm71358-sup-0001-SuppMat.pdf.