



Toward Stable and Efficient Mixed-Cation Mixed-Halide Perovskite Solar Cells

Gulab Singh Verma, Divya Tripathy, Puspanjali Hota, Sashikant, Toleshwar Prasad Rajwade, Alope Verma

Abstract: Mixed-cation, mixed-halide lead halide perovskites have rapidly progressed from laboratory curiosities to contenders for next-generation photovoltaics. By judiciously alloying A-site cations (Cs^+ , FA^+ , MA^+) and halide anions (I^-/Br^-), these materials marry outstanding optoelectronic quality and bandgap tunability, enabling single-junction devices exceeding 26% and hybrid perovskite-silicon tandems to approach ~35% certified efficiency. Despite these gains, light-induced halide segregation, ion migration, interfacial recombination, and environmental/thermal instability remain central challenges for scale-up and lifetime. This review synthesises the historical context and recent progress on triple-cation, mixed-halide absorbers; surveys at least twenty key studies spanning passivation such as FAPb treatments, defect/strain management, and tandem integration; and outlines practical, research-grade synthesis/processing steps for $\text{Cs-FA-MA Pb (I, Br)}_3$. We discuss characterization workflows via XRD, GIWAXS, PL/TPV/TRPL, UPS/Kelvin probe, JV under MPP tracking, EQE-EL reciprocity, and ISOS durability protocols, outline consensus findings, and map future directions, including wide-bandgap perovskites for stable tandems, ion-migration-aware design of devices, and AI-guided compositional/process discovery. Certified record data from NREL anchors the current efficiency landscape.

Keywords: Perovskite Solar Cells; Triple-Cation Cs-FA-MA; Halide Segregation; Perovskite-Silicon Tandem; Passivation; Stability; Efficiency Records.

Nomenclature:

XRD: X-Ray Diffraction

QFLS: Quasi-Fermi Level Splitting

UPS: Ultraviolet Photoelectron Spectroscopy

I. INTRODUCTION

The modern perovskite photovoltaics (PV) story began

With dye-sensitized architectures employing hybrid lead halides as sensitizers, soon pivoting to solid-state, thin-film devices, as unexpectedly long carrier diffusion lengths and benign defect physics emerged [1]. Rapid efficiency escalations from single digits to >20% within a few years were catalyzed by compositional engineering, interface control, and processing innovations [2]. A milestone for phase stability, crystallization robustness, and device reproducibility included the introduction of triple-cation compositions (typically written as $\text{Cs}_x\text{FA}_y\text{MA}_{1-x-y}\text{Pb}(\text{I}_{1-z}\text{Br}_z)_3$), which outperformed single-cation analogues [3]. Saliba and coworkers' demonstration that adding small fractions of inorganic cesium to FA/MA matrices yields more stable black-phase perovskites helped establish the field's compositional "default" for high performance [4]. Concurrently, mixed-halide alloying (I/Br) provided a practical lever to tune band gaps into the 1.65-1.8 eV window desirable for the top cells of monolithic perovskite silicon tandems [5]. Notably, this also highlighted the problem of light-induced halide phase segregation, in which illumination drives I-rich/Br-rich domain formation, red-shifts emission, and erodes open-circuit voltage [6]. Therefore, substantial effort has focused on mitigating segregation via strain management, entropy-stabilised formulations, passivation, and carrier/ion management strategies [7]. Today's efficiency landscape reflects this maturation. NREL's research-cell chart logs ~27.0% for single-junction perovskites and ~34.9% for certified 2-terminal perovskite-Si tandems (July 2025), positioning hybrid tandems as the near-term path beyond the Shockley-Queisser limit of single-junction Si. Meanwhile, III-V (GaAs) remains the single-junction benchmark (~29.1%), thin-film CIGS and CdTe set ~23-24% records, and OPV/DSSC benchmarks sit lower, useful yardsticks for contextualizing perovskite progress [8]. Alongside device records, the research frontier increasingly targets stability and scalability: controlling ion migration, alleviating interfacial non-radiative losses, engineering robust wide-bandgap chemistries, and translating lab-scale spin-coating to slot-die, blade, and vapour routes [9]. Very recent reports explore FAPb post-treatments, bifacial passivation, and poly-Si tunnelling recombination layers for tandems; complementary perspectives dissect ion migration's dual role [10]. Percolating AI/ML workflows accelerate recipe discovery across vast compositional and process-design spaces [11].

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II. EVOLUTION AND ADVANCES IN MIXED-CATION MIXED-HALIDE PEROVSKITE SOLAR CELLS

Mixed-cation, mixed-halide hybrid perovskite solar cells have emerged over the past decade amid intense development in halide chemistry and photovoltaic engineering. The early attempts focused on addressing the instability issues of MAPbI₃ by incorporating multiple A-site cations, thereby enhancing crystallinity and suppressing volatility. The real breakthrough came with the triple-cation system: Cs-FA-MA. Superior reproducibility, tolerance to environmental conditions, and phase stability set the stage for further achievements. This effectively stabilised the desirable α -phase of FAPbI₃, improved carrier lifetime, and optimised grain quality while minimising trap-state formation through partial Cs incorporation [12].

In parallel with cation alloying, mixed-halide engineering was developed to tune the bandgap, enabling the development of wide-bandgap absorbers suitable for tandem solar architectures. Regrettably, halide segregation upon light illumination remained a persistent issue, driven by ionic mobility, lattice strain, and light intensity, resulting in iodide- and bromide-rich domains that are detrimental to device uniformity. Later, it was demonstrated that such segregations can be suppressed by defect and strain engineering; compositional control improved the photophysical uniformity in α -FAPbI₃ films. Further ion migration studies elucidated its role in hysteresis, phase instability, and long-term degradation, now recognised as significant challenges to device durability [13].

Other significant developments include progress in surface and interface passivation. Organic salts, secondary crystallisation methods, and halide re-distribution increased grain size, improved film density, and prolonged carrier lifetimes. Besides, Cs-modified hole-transport-layer engineering enabled complementary pathways toward intrinsic device stability by further improving charge extraction and reducing recombination losses [14].

Efforts have also focused on incorporating these perovskite absorbers into tandem architectures that go beyond the limits of single-junction efficiency. The development of robust interconnection layers has allowed for efficient optical coupling and mechanical stability in perovskite/silicon tandems. Devices reported to date have achieved efficiencies above 33%, close to the theoretical limit of more than 42%. Flexible tandem variants have also achieved high performance in lightweight, portable applications favoured nowadays. Wide-bandgap mixed-halide absorbers with bromine content in the 20–30% range have so far proved very effective for highly efficient tandem top cells [15].

Recent updates from international efficiency databases confirm that single-junction perovskite devices now reach around 27% certified efficiency, while two-terminal perovskite/silicon tandems approach 35%. These values have been reproduced by several independent laboratories, which indicates readiness for pre-commercial deployment of this technology. The emphasis of contemporary research is on overcoming the remaining challenges in long-term operational stability, scalable fabrication, and defect-tolerant material processing. Extensive reviews now point out that the strategies of entropy stabilisation, interface design, and

crystallisation assisted by additives have turned mixed-cation mixed-halide perovskites into a versatile materials platform. Early compositional tuning has given way to advanced strategies revolving around defect passivation and the formation of homogeneous microstructures. In general, integrating chemical design, process optimisation, and interfacial engineering has made these materials a promising route toward next-generation, high-efficiency, and cost-effective solar energy technologies [16].

III. EXPERIMENTAL METHOD

In general, the fabrication of mixed-cation, mixed-halide perovskite solar cells follows a well-optimised solution-processed route that yields smooth, crystalline, and defect-minimised thin films. Target absorber composition in this context is generally represented as $\text{Cs}_x\text{FA}_{1-x}\text{Pb}(\text{I}_{1-z}\text{Br}_z)_3$, where Cs⁺, FA⁺, and MA⁺ occupy the A-site of the perovskite lattice. The halide sublattice consists of iodide and bromide in controlled ratios [17]. Typical compositions include Cs fractions of 5–15%, FA fractions of 70–90%, and the remaining portions in MA. Tuning of the halide ratio, particularly the bromine fraction within the range of 20–50%, can be used to adjust the optical bandgap from about 1.55 to 1.75 eV, meeting the requirements for high-efficiency single-junction devices and top cells in perovskite-silicon tandem configurations [18]. The synthesis generally starts with the preparation of a high-purity precursor solution containing lead halides, PbI₂ and PbBr₂, along with organic halides such as FAI, MAI, and CsI. Solvents such as DMF and DMSO are mixed in a 4:1 volume ratio to dissolve the salts [19]. Additives such as methylammonium chloride or thiocyanates are commonly added to control crystallisation kinetics while favouring grain enlargement. Post-treatment with formamidinium bromide has been used in several optimised recipes to heal halide vacancies, smooth surface defects, enhance carrier lifetimes, and suppress non-radiative recombination [20].

Equally important is the preparation of the substrate. The TCO substrates, generally ITO-coated glass, are successively cleaned with detergent, deionised water, and isopropanol, treated with ultraviolet ozone to remove organic residues and improve wettability [6]. The HTL is then deposited, typically a sol-gel-derived NiOx film that offers superior energy alignment and chemical compatibility with perovskites [3]. Researchers found that doping a small amount of CsI into NiOx improved the film's crystallinity and reduced the number of interface trap states [11]. The perovskite active layer is put down using a two-step spin-coating process [17]. To spread the precursor solution, it is spun slowly (about 1000 rpm) to achieve a uniform thickness, then spun quickly (about 4000 rpm) [2]. During the last few seconds of spinning, a non-polar antisolvent such as chlorobenzene or anisole is dripped onto the surface to accelerate crystallisation by pulling the solvent out [3]. After this process, the wet film is heated to 100–120°C for 20–30 minutes to form a uniform, photoactive perovskite phase [5]. In this step, you can optionally do FABr post-treatment by spin-coating with a thin FABr solution (1–3 mg/mL in isopropanol) and then heating

it for a short time [6]. This promotes uniform halides, lattice passivation, and improved protection against moisture [7]. To get the charge out, the whole device stack is built up by putting down ETLs one at a time. These are usually C_{60} (about 20 nm), a thin BCP interlayer (about 6–8 nm), and a reflective Ag or Au top contact (about 100 nm), all deposited by thermal evaporation. To protect the device from oxygen and moisture, it is crucial to encapsulate it in an inert nitrogen atmosphere with UV-curable epoxy and a glass coverslip. Instead of spin coating, blade coating or slot-die printing can be used for large-area fabrications. This allows depositing films on heated substrates while controlling solvent evaporation [8]. This method yields dense, pinhole-free films with high crystallinity and improved environmental stability. XRD, UV-Vis spectroscopy, PL, and SEM indicate that black-phase perovskites have formed and that the movies are uniform in shape. Optimised $\text{Cs-FA-MA Pb(I,Br)}_3$ films exhibit high absorption, long carrier diffusion lengths, and low trap densities. This means that single-junction configurations have PCEs above 25%, while tandem architectures have PCEs above 34%. This synthesis method is the best approach for making high-performance, stable, scalable, and efficient mixed-cation, mixed-halide hybrid perovskite solar cells [9].

IV. RESULT AND DISCUSSION

A lot of research has been done on mixed-cation, mixed-halide hybrid perovskite solar cells, and it shows that adding both inorganic and organic A-site cations makes the cells much more stable, with better optoelectronic properties, and more resistant to environmental conditions [10]. The most studied system, $\text{Cs}_x\text{FA}_{1-x}\text{MA}_{1-x}\text{Pb(I-zBrz)}_3$, is an excellent example of how changing the composition can change the crystallographic phase, the defect landscape, and the dynamics of charge carriers [11]. X-ray diffraction (XRD) and grazing-incidence wide-angle X-ray scattering analyses have consistently validated that the yellow, non-photoactive δ -phase of formamidinium lead iodide is inhibited upon the incorporation of a minor fraction of caesium and bromine. Adding an inorganic Cs^+ ion reduces the lattice size and the degree of octahedral distortion, thereby making the black phase more stable, even at high humidity. Also, adding mixed halides increases the optical bandgap to 1.65–1.75 eV, raising the open-circuit voltage and enabling the use of silicon bottom cells in tandem architectures [13].

Microscopic and spectroscopic analyses demonstrate significant improvements in film morphology and electronic uniformity. SEM characterisations show that triple-cation perovskites can make larger grains, about 500–800 nm in size, with fewer pinholes than single-cation perovskites. This increase in grain size is associated with lower trap density and longer carrier diffusion lengths, as shown by TRPL and SCLC measurements. Treatment with or addition of formamidinium bromide during or after film formation further promotes surface passivation, as evidenced by longer carrier lifetimes exceeding one μs and narrower Urbach energies below 15 meV. These signatures suggest a modest decrease in non-radiative recombination, which should directly improve device efficiency and stability [14].

Optoelectronic characterisation has demonstrated a notable enhancement in the quasi-Fermi level splitting (QFLS), approaching the radiative limit for these materials. Steady-state photoluminescence (PL) spectra of mixed-halide triple-cation perovskites exhibit blue-shifted, stronger emission than that of binary or single-cation analogues. This means that charge transport is more balanced and there are fewer deep traps. Ultraviolet photoelectron spectroscopy (UPS) and Kelvin-probe studies show that the valence and conduction bands are better aligned at the interfaces with NiOx and C_{60} , respectively. This makes it easier to extract carriers and reduces hysteresis. Devices using Cs-modified NiOx exhibit higher fill factors ($\text{FF} \approx 82\text{--}84\%$) and open-circuit voltages up to 1.24 V, underscoring the importance of aligning energy levels at the interface [15].

Nevertheless, photo-induced halide segregation remains one of the most frequently discussed concerns. Under constant light irradiance, iodide and bromide ions may drift, resulting in local composition gradients that cause a redshift in photoluminescence and/or a decrease in V_{OC} . However, with entropy-rich compositions and effective strain management, this process is considerably delayed. In situ PL mapping and light-bias EQE tracking reveal that minimising surface strain and halide disorder can suppress phase separation for several hours of operation. Compositional engineering, incorporation of large organic cations at the grain boundaries, and robust encapsulation have also recently been shown to restrain ionic migration [16].

Electrical characterisation under one-sun illumination has revealed PCEs of over 25% for single-junction inverted architectures and has provided evidence of stabilised output via MPP tracking. The V_{OC} deficit, quantified by $(E_{\text{g}}/q - V_{\text{OC}})$, was pushed below 0.35 V, which means near-ideal radiative efficiency. In this context, wide-bandgap perovskites (≈ 1.7 eV) in tandem with crystalline silicon have reported certified efficiencies as high as 34.9%, which is a massive leap beyond the Shockley-Queisser limit for single-junction photo-voltaic conversion. Performance milestones are summarised in the NREL 2025 efficiency chart, where perovskite-based tandems stand alongside the world's highest-efficiency photovoltaic devices [17].

ISOS protocol stability tests confirm that, when properly encapsulated, the TCMH cells can retain more than 90% of their initial efficiency under continuous illumination at 65°C for 1000 hours. Degradation analyses suggest that most residual performance losses are due to interfacial reactions between the perovskite and charge-transport layers, rather than intrinsic perovskite decomposition [18]. These have now been mitigated with strategies such as inserting ultrathin ALD-SnO_x or poly-Si tunneling recombination layers. The results show that triple-cation, mixed-halide perovskites strike a delicate balance between their electronic performance and structural strength [19]. They deliver strong absorption in the visible range, long carrier diffusion lengths, and minimal trap-assisted recombination, while maintaining tolerance for thermal and environmental stress. Correspondingly, the synergistic roles of Cs^+ incorporation, Br^- substitution, and post-treatment passivation converge in high-efficiency devices that are steadily increasing toward commercial

reliability. Further tuning of the composition, interfacial chemistry, and encapsulation process, permitting AI-guided process optimisation, continues to enhance efficiency and operational lifetime, moving these materials toward large-scale, next-generation photovoltaic deployment [20].

V. CONCLUSION

In modern photovoltaics, hybrid perovskites with mixed cation-halide compositions have emerged as a breakthrough technology owing to their exceptional balance of efficiency, stability, and tunability. Incorporation of more than one A-site cation like Cs^+ , FA^+ , and MA^+ with halides I^- and Br^- tends to stabilise the perovskite lattice, improve film quality, and allow bandgap control suitable for single-junction and tandem applications. Surface passivation and interfacial engineering further enhance device durability by reducing defects. Recently, power conversion efficiencies of about 27% for single-junction cells and 34.9% for perovskite/silicon tandem cells have been realised, providing a new benchmark for thin-film solar technologies. Compositional tuning, encapsulation, and advanced process control have progressively mitigated the challenges of ion migration and halide segregation. This perspective considers triple-cation, mixed-halide perovskites as one of the best routes toward highly efficient, low-cost, stable solar cells. Continued research on scalable fabrication, improved stability, and AI-assisted optimisation is up-and-coming to accelerate further the transition of these devices from laboratory prototypes to commercial solar technologies.

DECLARATION STATEMENT

After aggregating input from all authors, I must verify the accuracy of the following information as the article's author.

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