

# In Situ Dual-Interface Passivation Strategy Enables The Efficiency of Formamidinium Perovskite Solar Cells Over 25%

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Perovskite solar cells (PSCs) are promising candidates for next-generation photovoltaics owing to their unparalleled power conversion efficiencies (PCEs). Currently, approaches to further improve device efficiencies tend to focus on the passivation of interfacial defects. Although various strategies have been developed to mitigate these defects, many involve complex and time-consuming post-treatment processes, thereby hindering their widespread adoption in commercial applications. In this work, a concise but efficient in situ dual-interface passivation strategy is developed wherein 1-butyl-3-methylimidazolium methanesulfonate (MS) is employed as a precursor additive. During perovskite crystallization, MS can either be enriched downward through precipitation with  $\text{SnO}_2$ , or can be aggregated upward through lattice extrusion. These self-assembled MS species play a significant role in passivating the defect interfaces, thereby reducing nonradiative recombination losses, and promoting more efficient charge extraction. As a result, a PCE >25% (certified PCE of 24.84%) is achieved with substantially improved long-term storage and photothermal stabilities. This strategy provides valuable insights into interfacial passivation and holds promise for the industrialization of PSCs.

## 1. Introduction

Perovskite materials are promising candidates as new-generation photovoltaic materials due to their favorable characteristics, including a tunable bandgap, a high light absorption coefficient, and excellent charge carrier transport properties.<sup>[1–5]</sup> To date, power conversion efficiencies (PCEs) of >26% have been achieved for perovskite solar cells (PSCs), and as a result, they have received growing attention from the photovoltaic community.<sup>[6]</sup> State-of-the-art PSCs are usually prepared by low-temperature solution methods, which are high-throughput, low-cost industrialization processes.<sup>[7,8]</sup> However, perovskite films prepared via such rapid low-temperature approaches tend to contain large numbers of interfacial defects. Compared to bulk defects, interfacial defects can be more detrimental to the performance of a device due to their significantly higher densities.<sup>[9,10]</sup> These interfacial defects, which can lead to significant non-radiative recombination

losses, constitute a crucial impediment to approaching the Shockley–Queisser limit (SQ limit) of the PCE.<sup>[11,12]</sup> In addition, these interfacial defects promote the degradation of PSCs, which must be addressed to achieve the industrialization of PSCs.<sup>[13–15]</sup>

To reduce the interfacial defect density, various defect passivation strategies have been developed, and these can be classified into top and buried interfacial passivation approaches depending on the functioned spatial position.<sup>[16–21]</sup> In the top interface approach, volatile organic components can be released from the perovskite film during thermal annealing, which can cause deficiencies in the coordination of lead or halide vacancies.<sup>[22]</sup> To address this issue, halide salts have been used to induce recrystallization, and low-dimensional perovskite passivation layers have been employed to modify the perovskite surfaces.<sup>[23,24]</sup> In terms of the buried interface, the abundance of trap sites on the surfaces of charge-transport layers (e.g.,  $\text{SnO}_2$  or polymer poly(bis(4-phenyl)(2,4,6-trimethylphenyl)amine) (PTAA), can have detrimental effects on perovskite crystal growth, interfacial charge transport, and the device stability.<sup>[25]</sup> To optimize the buried

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