


Review

Passivation Strategies through Surface Reconstruction toward Highly Efficient and Stable Perovskite Solar Cells on n-i-p Architecture

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Abstract: Perovskite solar cells have achieved remarkable enhancement in their performance in recent years. However, to get an entrance to the photovoltaic market, great effort is still necessary to further improve their efficiency as well as their long-term stability under various conditions. Among various types of approaches (including compositional engineering, dopant engineering, self-assembled monolayers (SAMs), et al.), interfacial engineering through passivation treatment has been considered as one of the most effective strategies to reduce the non-radiative recombination within the PSCs. Thus, this short review summarizes recent efforts on chemical interfacial passivation strategies from a different perspective owing to their common phenomena of reconstructing the perovskite surface via the formation of three-dimensional perovskite, low-dimensional perovskite and synergistic effect provided by a mixed-salt passivation system, respectively.

Keywords: passivation; surface reconstruction; perovskite solar cells



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1. Introduction

Solar cells based on organic–inorganic hybrid perovskite structured materials have attracted tremendous attention in recent years, in which both efficiency and stability have been dramatically enhanced. In particular, the state-of-the-art record efficiency has reached 25.5% with n-i-p architecture [1–4], which is comparable with the well-established crystalline silicon based solar cells (with efficiencies around 26%) and better performance than the thin-film based solar cells (with power conversion efficiency (PCE) around 23% as indicated in National Renewable Energy Laboratory (NREL) efficiency chart), making perovskite solar cell (PSC) a promising candidate in the PV market. However, there are still many defect trap states and charge transport barriers presented at the grain boundaries, bulk and interfaces within the PSCs, which not only hinders the further improvement in PCE towards its theoretical limit but also affects the stability of the devices, especially under exposure to moisture, oxygen, heating and light illumination [5,6]. Therefore, to address such issues, extensive work has been conducted on investigating various materials as well as device engineering.

Passivation is considered to be one of the most powerful approaches to enhance the device performance and long-term stability, mainly by improving charge carrier extraction and suppressing defect trap states [7–9]. It refers to a material becoming “passivate”, according to the International Union of Pure and Applied Chemistry (IUPAC). Thus, passivation layers normally act as shielding layers to protect the active semiconductor surface from being affected or corroded by the surrounding environment. For PSCs, the passivation layer aims at either reducing the defect trap states at the grains and grain boundaries through chemical passivation or isolating adjacent functional interlayers through physical passivation to protect in order to avoid device degradation from exposing

to the external environment, thus leading to an enhanced long-term stability. So far, a large amount of literature has been dedicated to summarizing various passivation materials and their effect in PSCs [5–11]. However, this perspective, on the other hand, provides a different way of clarifying the chemical passivation effect through surface reconstruction, which is one of the most effective approaches to reduce the surface defects of the perovskite as well as the interface between perovskite and the hole-transporting layer (HTL) via the formation of a three-dimensional, low-dimensional interlayer, as well as the synergistic effect provided by a mixed-salt passivation system.

2. Surface Reconstruction with Formation of Three-Dimensional Perovskite Interlayer

Excess lead-iodide (PbI_2) has been widely used as a passivation additive in perovskite films for high performance PSCs. In 2014, Yang and co-workers reported a self-induced passivation technique with the proper amount of PbI_2 in the perovskite film of $\text{CH}_3\text{NH}_3\text{PbI}_3$, which effectively improves its carrier behavior owing to the suppressed recombination at the grain boundaries (GBs) and adjacent interface with TiO_2 (as illustrated in Figure 1), leading to a pronounced enhancement in device performance [12]. Self-induced passivation with excess/unreacted PbI_2 has been investigated as a common strategy in improving device performance of PSCs by the other works, since PbI_2 will help to reduce halide vacancies and passivate the defects in perovskite GBs [13,14]. However, the presence of unreacted PbI_2 results in an intrinsic instability of the perovskite film, especially under continuous light illumination, which accelerates the formation of decomposition products of lead and iodine in the perovskite films [15]. The degradation mechanism and detrimental effect under long-term stability has been recently further investigated by Tumen-Ulzii and co-workers [16].

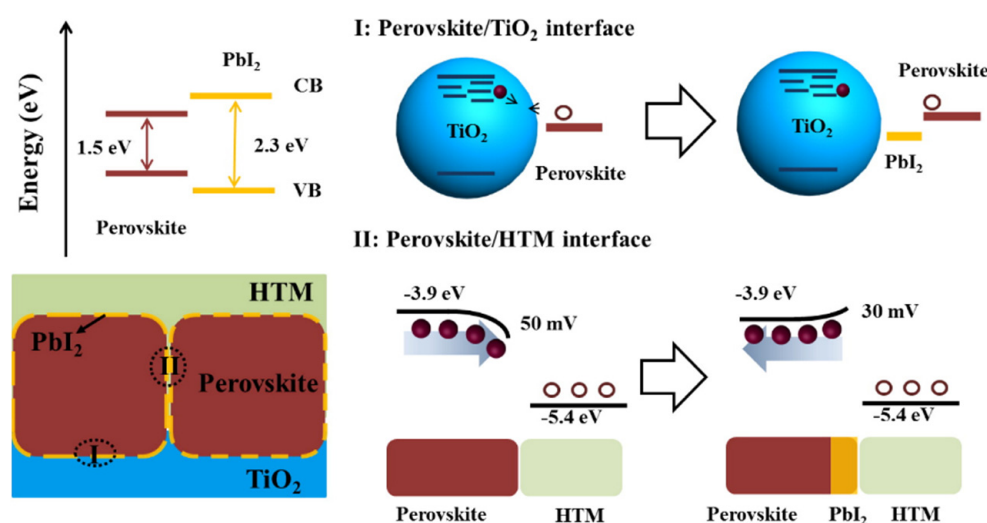


Figure 1. Energy alignment and bandgap of PbI_2 and perovskite is shown on the top left. The schematic n-i-p structure is shown on the bottom left. (I) The interface of perovskite/ TiO_2 before and after the introduction of PbI_2 is described on the top right. (II) The interface of perovskite/HTM before and after the introduction of PbI_2 is shown on the bottom right. Reprinted with permission from reference [12]. Copyright (2014) American Chemical Society.

As discussed above, a series of studies have proven that unreacted PbI_2 in the perovskite films can bring a positive effect on device performance, while PbI_2 crystals is one of the reasons for accelerating the degradation of PSCs. Therefore, many efforts have focused on surface reconstruction with formation of an additional perovskite layer through reactions with the excess of PbI_2 from the pristine perovskite layer for further improvement in device performance as well as long-term stability. For example, Cho and co-workers introduced an interfacial passivating modifier of formamidinium bromide (FABr), which constructs an additional $\text{FAPbBr}_{3-x}\text{I}_x$ layer via reaction with excess PbI_2 on top of the

primary $(\text{FAPbI}_3)_{0.85}(\text{MAPbBr}_3)_{0.15}$ film [17]. Such post-treatment resulted in a perovskite surface reconstruction towards a denser and more homogenous film, as compared to the as-prepared perovskite film, which is indicated in the high-resolution scanning electron microscope (SEM) (as shown in Figure 2). Consequently, the reconstructed $\text{FAPbBr}_{3-x}\text{I}_x$ layer also acted as an electron blocking layer, leading to an improved PCE. However, the shelf-stability of the passivated device is similar to that of the control device. Other self-induced passivation agents, such as methylammonium bromide (MABr), have also been investigated to improve the perovskite film morphology after post-treatment of the perovskite film, which will passivate the surface defects and further improve the PCEs of the target devices [18]. MABr is also developed to treat as-prepared FAPbI_3 film, which helps to increase the perovskite crystallinity as well as to optimize its absorption edge [19].

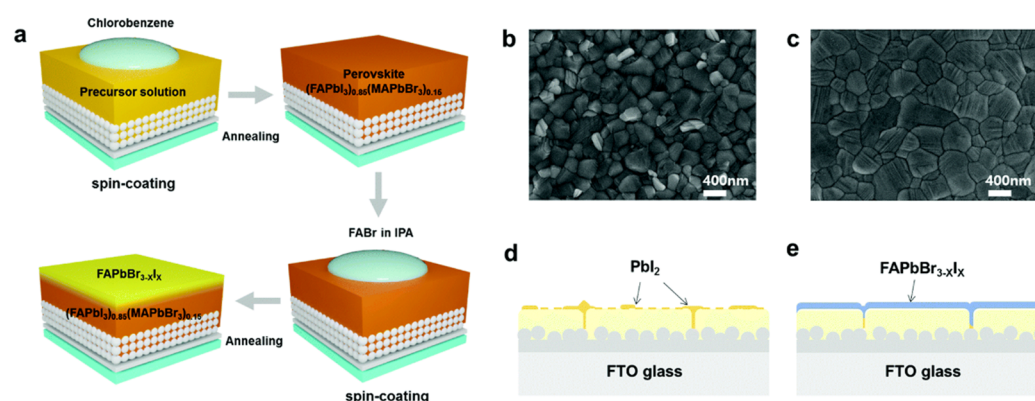


Figure 2. (a) Illustration of the fabrication method of FAPbBr passivated perovskite film. Top-view SEM images of (b) reference film and (c) FAPbBr passivated film. Proposed change of film cross sectional structures of (d) reference film and (e) FAPbBr passivated film. Republished with permission of Royal Society of Chemistry, from reference [17].

3. Surface Reconstruction with Formation of Low-Dimensional Perovskite Interlayer

Low-dimensional perovskites exhibit better environmental stability and photostability compared with their three-dimensional (3D) counterparts, but owing to its poor charge transport and wide bandgaps, perovskite PSCs based on low-D structure show relatively low efficiency [20]. Therefore, efforts have been focused on the structure of low-D/3D perovskite through interfacial engineering on the 3D-based perovskite with large size organic cation spacers. The newly formed low-D perovskite interlayer can help to further improve the performance of PSCs. Meanwhile, they also exhibit excellent long-term stability under various conditions. For example, Soek's group demonstrated a systematic interfacial study on the effect of different alkyl chain length of ammonium iodide (butylammonium iodide (BAI), octylammonium iodide (OAI), and dodecylammonium iodide (DAI)) with the formation of a 2D perovskite on the top of 3D layer [21]. They realized that the chain's electron-blocking ability and humidity resistance increase dramatically as the alkyl chain length increases. Consequently, the PSC treated with OAI has achieved a certified stabilized PCE of 22.9% and enhanced long-term stability under high humidity, with a relative humidity of 85%. Yoo et al. deposited a novel precursor/solvent combination (alkyl ammonium bromides/chloroform) on top of the 3D perovskite layer for the synthesis of the 2D layer. Such treatment resulted in a less faceted and distinctive grain surface and boundaries compare to the pristine 3D perovskite, leading to a certified stabilized PCE of 22.6% with the longest chain length for ammonium bromides [22]. Liu et al. employed pentafluorophenylethylammonium iodide (FEAI) on the surface 3D perovskite. The introduction of an F-rich 2D layer not only exhibited outstanding humid stability, but also achieved the perovskite solar cells with over 22% efficiency [23]. Except the 2D/3D PSCs, Chen's group developed an in situ cross-linking polymerizable propargylammonium on the top of 3D absorption perovskite surfaces [24]. The new formed 1D/3D perovskite heterostructure

could efficiently enhance the device performance and stability of PSCs. They evaluated the ISOS-D-1 stability of the unencapsulated solar cells under ambient condition with a humidity of 40–70% for 1500 h. The target devices retained 97% of their initial performance, whereas the control cell dropped to 43%. Moreover, after maximum power point tracking for more than 3000 h under continuous illumination, the passivated devices still retained 93% of their initial efficiency. More ammonium salts, such as butylammonium iodide (BAI), phenethylammonium iodide (PEAI), along with other types of molecules have also been reported to form low-D perovskite layers at the interface between perovskite and HTL, which present better PCEs by reducing the defects, suppressing the non-radiative recombination, and filling iodine vacancies of both the surfaces and GBs of perovskite films. Alongside this, the strategies show outstanding improvement in long-term stability, especially under a humid atmosphere [25,26].

4. Surface Reconstruction with Mixed-Salt Passivation through Synergistic Effect

As mentioned above, tremendous efforts have been made on designing functional molecules for interfacial treatment between perovskite and HTL. With the exception of the signal salt that was used to passivate the perovskite films, mixed-salt passivation has also been developed. The mixed-salt passivation provides a new strategy of designing new passivated materials, which could get benefits from signal material as well as the combined synergistic effect of two salts. Cho and co-workers reported a mixture of *iso*-butylammonium iodide (iBAI) and formamidinium iodide (FAI) to post-treat the as-prepared 3D perovskite layer, which enhanced both the efficiency and the long-term stability of PSCs after the treatment [27]. It is observed via scanning electron microscopy (SEM) that, as shown in Figure 3, the mixed-salt treatment significantly changed the perovskite surface morphology as the iBAI content increased with the formation of an interfacial layer. The authors also speculated that the passivation mechanism that the FAI reacts with the excessive PbI_2 from the 3D perovskite reacts under the assistance of iBAI during the crystallization process, whereas FAI itself without iBAI cannot achieve the same results due to the insufficient annealing temperature. In addition, the formation of such an interfacial energetic barrier can effectively reduce the ionic transport, leading to a significantly suppressed photocurrent hysteresis. More recently, we proposed and investigated a systematic study of the mixed-salt passivation strategy of different F-substituted alkyl lengths of ammonium iodide, $\text{CF}_3\text{CH}_2\text{NH}_3\text{I}$ (TEAI), $\text{CF}_3(\text{CF}_2)_2\text{CH}_2\text{NH}_3\text{I}$ (HBAI) and $\text{CF}_3(\text{CF}_2)_5\text{CH}_2\text{NH}_3\text{I}$ (THAI) coupled with formamidinium bromide (FABr) [28,29]. We realized that the introduction of FABr resulted in a surface reconstruction with excessive PbI_2 from the 3D perovskite, leading to the formation of an ultra-thin layer of $\text{FAPbI}_x\text{Br}_{3-x}$, which can be observed from SEM images and XRD patterns, as shown in Figure 4. Meanwhile, F-substituted the ammonium iodide passivated not only the surface but also the perovskite bulk through penetration into the grain boundaries, as evidenced by the energy dispersive X-ray spectroscopy (EDS) and X-ray photoelectron spectroscopy (XPS) depth profiling shown in Figure 4. In addition, it is worth noticing that the shorter the length the alkyl ammonium iodide employs, the deeper it diffuses into the underlying 3D perovskite, resulting in a champion PCE of 23.5%. Furthermore, all devices passivated with TEAI·FABr, HBAI·FABr or THAI·FABr shown more moisture tolerance with a water droplet contact angle of around 60° on the perovskite surface, owing to the introduction of F-substituted alkyl ammonium salts. As a result, the best performing device with TEAI·FABr treatment retained a remarkable 98% PCE of its initial value after exposing to a humidity of 30–60% in the dark for 800 h. Remarkably, the target devices also present excellent operational stability under continuous one sun illumination for 600 h with only 8% PCE drop, whereas the control device suffered from a 15% PCE drop of its initial value. This result illustrates the important of the synergistic effect provided by the mixed-salt passivation system in obtaining high efficiency and long-term stability simultaneously.

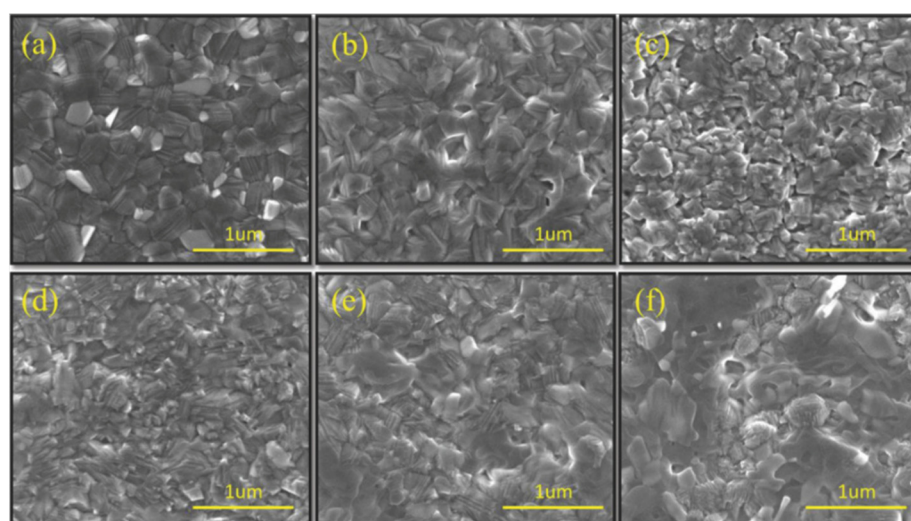


Figure 3. Top-view SEM images of (a) pristine perovskite film and (b–f) post-treated perovskite film with the increase ratio of iBAI. Republished with permission of John Wiley and Sons, from reference [27].

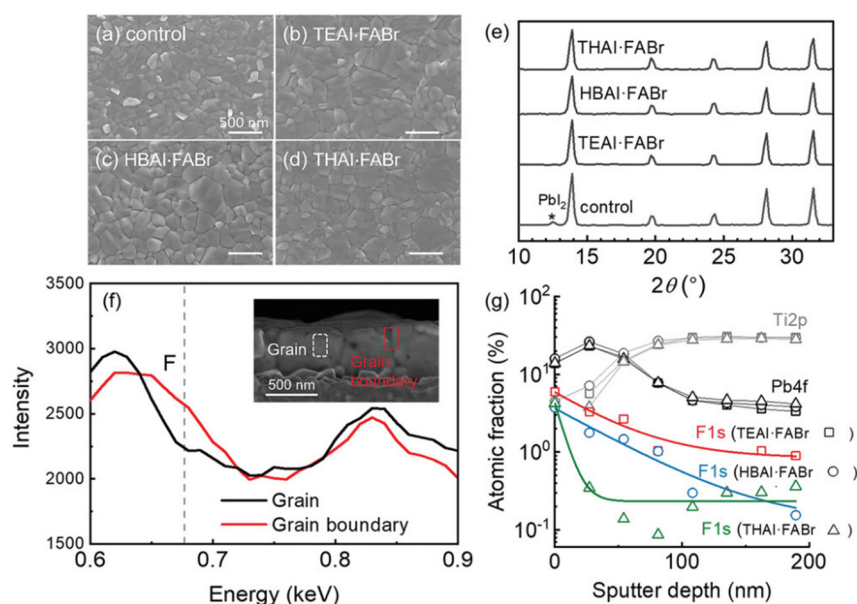


Figure 4. Top-view HR-SEM images of perovskite. (a) Control films and perovskite film with treatment of (b) TEAI-FABr, (c) HBAI-FABr and (d) THAI-FABr. (e) GIXRD patterns of pristine film and passivated perovskite film with TEAI-FABr, HBAI-FABr and THAI-FABr. (f) EDS spectra of the grain boundary and grain of the TEAI-FABr treated perovskite film. (g) XPS depth profiling spectra of the post-treated films with TEAI-FABr, HBAI-FABr and THAI-FABr. Copyright from reference [29].

5. Summary and Outlook

Efficiency and stability have been identified as the most important challenges for the commercialization of perovskite solar cells. In this review, we expounded various passivation approaches of perovskite film through surface reconstruction to make efficient and stable perovskite solar cells on n-i-p configuration. In general, such passivation treatment improves the device performance of a PSC in the following ways:

- (1) Reconstructing the perovskite surface through the formation of an optimized layer of either 3D or low-dimensional perovskite aiming to reduce trap defects, hinder ion migration and suppress the non-radiative recombination centers.

- (2) Adjusting the energy levels through the newly formed interlayer to improve the charge carrier extraction and/or block electron transfer.
- (3) Enhancing the surface hydrophobicity through the formation of a more robust low-dimensional perovskite layer, which consequently improves the device stability, especially under humid conditions. In comparison, the formation of an additional 3D perovskite interlayer shows limited improvement in long-term stability due to the inherent instability of the 3D perovskite against moisture.
- (4) Combined improvements provided by mixed-salt system through synergistic effect.

Although tremendous efforts have been spent on optimizing PSCs with passivation treatment, there is still a lack of systematic studies on developing general rules and criteria for material selection and their passivation mechanisms. In addition, more characterization and theoretical calculation methods need to be further developed.

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