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COMMUNICATION

Effects of formamidinium and bromide ion substitution in methylammonium lead triiodide toward high-performance perovskite solar cells

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Abstract
Compositional engineering of organic-inorganic hybrid perovskite has attracted great research interests recently for seeking a better perovskite system to address existed challenges, such as the thermal and moisture instability, anomalous hysteresis, and toxic lead contamination, etc. In this study, we systematically investigated the structural, optophysical, and photovoltaic properties of the compositional $MA_xFA_{1-x}Pb(I_yBr_{1-y})_3$ perovskite by sequentially introducing FA^+ and Br^- ions into the parental $MAPbI_3$ to elucidate their respective roles when they were inserted into the perovskite lattice. We unraveled that such dual compositional tuning in perovskite can improve the crystallinity of the resultant film and thus reduce its density of defect states as evidenced by admittance spectroscopy, resulting in a prolonged carrier lifetime over 500 ns. As a result, a promising average PCE (PCE_{AVG}) of 17.34% was realized in the optimized $MA_{0.7}FA_{0.3}Pb(I_{0.9}Br_{0.1})_3$-based PVSC with little hysteresis and stable photocurrent output. More significantly, another compositional $MA_{0.7}FA_{0.3}Pb(I_{0.8}Br_{0.2})_3$ perovskite with a large bandgap of 1.69 eV can yield an impressively high PCE_{AVG} over 15%. To the best of our knowledge, this performance is among the state-of-the-art large bandgap (~1.7 eV) PVSCs reported so far, which paves the way for the development of high-performance tandem cells using efficient large bandgap PVSCs as the top subcells. This study not only manifests the pivotal roles of dual compositional tuning in $MA_xFA_{1-x}Pb(I_yBr_{1-y})_3$ perovskites but

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also highlights the importance of compositional engineering for developing an even more efficient perovskite.
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Introduction

Organic–inorganic hybrid perovskites have emerged as one of the most intriguing light-harvesting materials for solar energy due to their exceptional semiconducting properties and cost-effective solution processability [1-5]. Methylammonium lead triiodide perovskites ($MAPbI_3$) have received much attention, and high power conversion efficiencies (PCEs) of over 17% were reported recently [6,7]. However, $MAPbI_3$ still faces several complex challenges that need to be addressed, such as the thermal and moisture instability [8-11], hysteretic current-voltage characteristics [12,13], and toxic lead contamination [14-16], etc. In this regard, many researchers in this field have shifted their focus toward composition engineering of hybrid perovskites to seek a better system for photovoltaic applications [9,17-20], Tuning the organic cations or the halide anions of hybrid perovskites are the most common methods reported thus far.

Notably, formamidinium lead triiodide ($FAPbI_3$) perovskite shows promise as a photovoltaic material because its narrower bandgap (E_g) of 1.48 eV lies closer to the Shockley-Queisser ideal bandgap of ~1.40 eV for single-junction solar cells, meaning even higher PCEs can be realized [21,22], However, controlling the phase purity of $FAPbI_3$ thin films is a significant hurdle to fabricating efficient $FAPbI_3$-based perovskite solar cells (PVSCs). The appearance of the yellow non-perovskite δ-phase accompanies the formation of black α-phase $FAPbI_3$ during annealing due to its relatively lower formation energy hinders the performance of the derived PVSCs due to its insulating properties [1]. Therefore, the most critical issue is to eliminate the formation of the δ-phase to obtain a pure black α-phase $FAPbI_3$. Recently, Grätzel et al. partially substituted MA^+ with FA^+ ions in $MAPbI_3$ and reported higher PCEs for the perovskite $MA_{0.6}FA_{0.4}PbI_3$ than the parental perovskites [19].

Halide substitution in $MAPbI_3$ has been widely investigated to date. Generally, the inclusion of chlorine in $MAPbI_3$ (denoted as $MAPbI_{3-x}Cl_x$) mainly results in the improved film coverage and crystallinity compared to the pristine $MAPbI_3$ as considering the fact that no detectable Cl is residual in the resultant film due to the release of MACl during thermal annealing [18,23,24], while the inclusion of bromine ($MAPbI_{3-x}Br_x$) can effectively increase the E_gs of the resultant perovskite thin films [9]. Recently, Seok et al. demonstrated a PVSC with a certified PCE of 20.1% using a perovskite with the composition of $(FAPbI_3)_{0.95}(MAPbBr_3)_{0.05}$, indicating the importance of composition engineering of perovskites [17]. Given the opposing impacts of the FA^+ and Br^- substitutions on the E_g of $MAPbI_3$, the respective roles of these incorporated ions are still not readily understood at this stage.

In this communication, we systematically investigate the structural, photophysical, and photovoltaic properties of the compositional $MA_xFA_{1-x}Pb(I_yBr_{1-y})_3$ perovskites and elucidate the respective roles of FA^+ and Br^- ions by sequentially introducing them into the compositional perovskites. The incorporation of FA^+ ions into $MAPbI_3$ to form $MA_xFA_{1-x}PbI_3$ improved the crystallinity, carrier lifetime, and absorption of the resultant perovskite thin films. More importantly, the perovskite phase conversion temperature could be maintained at 100 °C despite the presence of FA^+ ions, which is much lower than that for forming pure black α-phase $FAPbI_3$ (150 °C). Subsequently, Br^- ions that were introduced to partially replace I^- ions improved the crystallinity of the resultant film to reduce the density of trap states (N_T) as evidenced by the admittance spectroscopy. This enabled the optimized compositional $MA_{0.7}FA_{0.3}Pb(I_{0.9}Br_{0.1})_3$ perovskite to possess a very long charge carrier lifetime of over 500 ns. As a result, a high average PCE (PCE_{AVG}) of 17.34% was realized in the $MA_{0.7}FA_{0.3}Pb(I_{0.9}Br_{0.1})_3$-based PVSC with little hysteresis and stable photocurrent output, outperforming the cells derived from the parental perovskites and those with other compositions. More encouragingly, a compositional $MA_{0.7}FA_{0.3}Pb(I_{0.8}Br_{0.2})_3$ perovskite with a large E_g of 1.69 eV exhibited a high PCE_{AVG} over 15%. To the best of our knowledge, this performance is among the state-of-the-art large E_g (~1.7 eV) PVSCs reported so far [25]. The synergistic effects in tuning perovskite composition revealed in this work signify that perovskites with different compositions in addition to $MAPbI_3$ deserve more in-depth understanding from physical and crystallographic aspects.

Experimental section

Material preparation

Lead iodide, lead bromide, nickel acetate tetrahydrate, copper acetate monohydrate, $PC_{61}BM$ and C_{60} were purchased from Sigma-Aldrich, and $PC_{61}BM$ was purified before use. Methyl ammonium iodide and formamidinium iodide was synthesized according to previously reported methods [18,20]. Bis-C_{60} was synthesized as reported in our previous work [26].

A 0.1 M NiO_x precursor solution was prepared by dissolving nickel acetate tetrahydrate into ethanol, followed by adding 0.6 vol% ethanolamine into the solution. Separately, A 0.1 M Cu precursor solution was prepared by dissolving copper acetate monohydrate into ethanol. The final Cu-doped NiO_x precursor was prepared by mixing the NiO_x precursor and Cu precursor solutions at volume ratio of 95:5 [6]. Compositional perovskite precursors (1 M) were prepared by dissolving MAI, FAI, PbI_2 and $PbBr_2$ with different ratio into a mixed solvent of dimethylsulfoxide and γ-butyrolactone (volume ratio of 3:7) and stirred at 60 °C for 12 h. Mixed $PC_{61}BM$ and C_{60} solutions (15 mg mL^{-1}) were prepared by dissolving $PC_{61}BM$ and C_{60} into dichlorobenzene with a weight ratio of 1:1. Bis-C_{60} surfactant solution (2 mg mL^{-1}) was prepared by dissolving Bis-C_{60} into isopropyl alcohol followed by sonication for one hour.

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