

Formamidinium-Caesium Perovskite Solar Cells and Modules from Lead Acetate-Based Precursors

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Abstract

Perovskite solar cells (PSCs) have attracted widespread attention as a promising photovoltaic technology due to the outstanding photovoltaic properties of perovskite active layers. The power conversion efficiency (PCE) of PSCs is rapidly approaching their theoretical limit. However, scalability and stability remain the main obstacles restricting their commercialization. Lead acetate-based precursors have produced ultrasoft perovskite thin films with only short annealing times and without the need for an antisolvent. The accelerated perovskite crystallization process and facile processability make lead acetate a promising lead source for large-scale fabrication of PSCs. However, to date, it has only been used as a precursor for the synthesis of methylammonium (MA) or caesium (Cs) based perovskites due to a limited understanding of side-reactions occurring in the precursor solution, which limits its application in the field (Figure 1A). For the lead acetate route to yield high purity perovskite thin films, a compatible cation is required for effective thermalization of the volatile components. Here we outline why the conventionally used methylammonium cation cannot be used in conjunction with formamidinium (FA) and show that replacing it with ammonium (NH_4^+) addresses this issue. This expands the lead acetate route to all formamidinium-caesium (FACs) perovskites, and hence makes it viable for all high-performing and industrially relevant perovskite compositions.

Moreover, we also compared FACs perovskite thin films prepared from our newly developed PbAc_2 route (target) to films with the same stoichiometry produced following the conventional PbI_2 route (reference). We performed scanning electron microscopy (SEM) to study the morphologies of the perovskite films. Top view images are shown in Figures 2A and 2B. The reference and target films both show compact, uniform and pinhole-free surfaces, but with significantly larger apparent grain sizes in the target films. We further investigated the charge carrier lifetime of the films on the microscale ($10 \mu\text{m} \times 10 \mu\text{m}$) by performing time-resolved confocal PL lifetime mapping measurements (Fig. 2C and 2D). Compared with the reference film (blue region), the target film exhibited a much longer lifetime (green region). The increased PL lifetime and uniformity further accentuate the enhanced optoelectronic performance and effective suppression of non-radiative recombination when using the PbAc_2 route.

Furthermore, high-quality large-area FACs mixed-cation perovskite films were produced by blade-coating a lead acetate-based precursor formulation in an ambient laboratory environment, with the use of NH_4^+ as a volatile cation to drive off acetate during annealing (Figure 1B), leading to formation of PSCs with a power conversion efficiency (PCE) of up to 21.0%. Blade coated mini-modules with an aperture area of 10 cm^2 displayed PCEs of up to 18.8% (Figure 3). The encapsulated PSCs showed excellent thermal stability, with no evidence of efficiency loss after 3300 hours at 65° C (Figure 4). These results and insights will stimulate further work on the use of lead acetate to facilitate stable and efficient large area PSCs and thereby contribute to the commercialization of this technology.

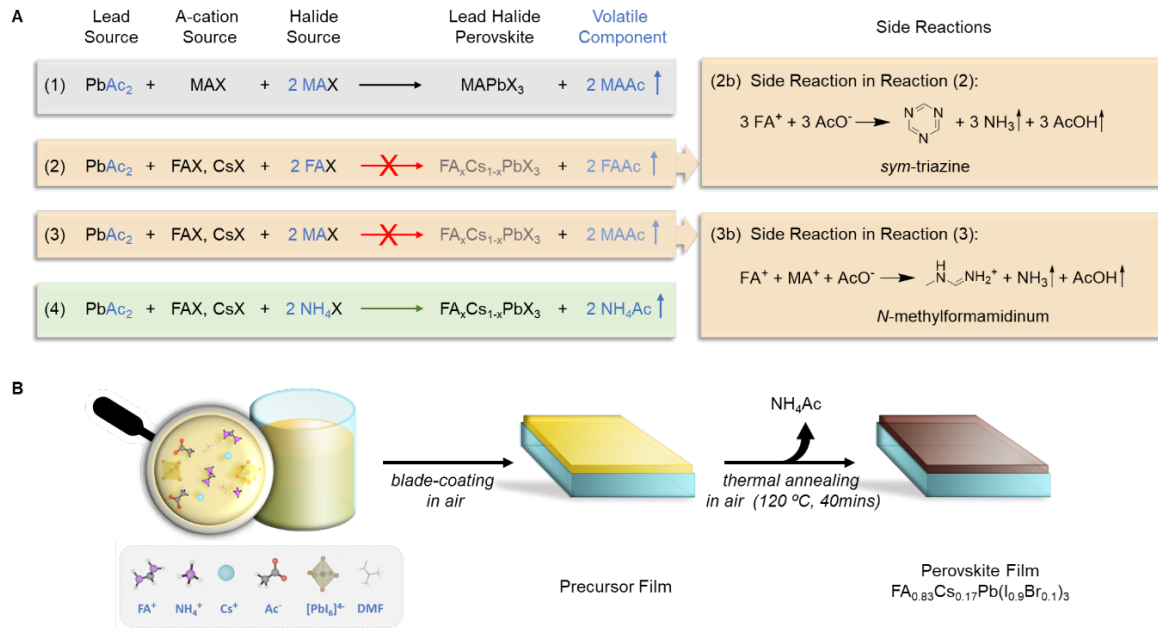


Fig. 1 Effect of different volatile cations in PbAc_2 -based precursors on the formation of FACs perovskite. (A) Different synthetic avenues for the synthesis of MA- and FACs-perovskites using the PbAc_2 route and proposed side reactions. (B) Schematic diagram of the fabrication process of the FACs perovskite from a PbAc_2 -based precursor solution with NH_4^+ as the volatile cation (abbreviated to A-Ac-FACs).

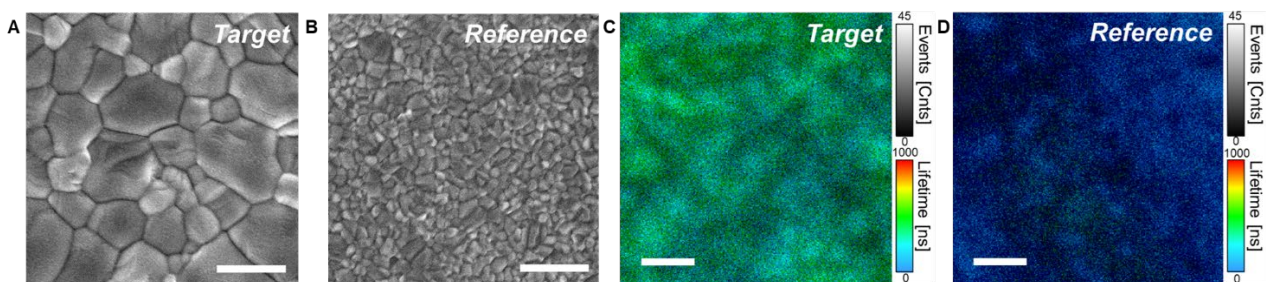


Fig. 2 (A and B) Top-view scanning electron microscope (SEM) images of the (A) target and (B) reference. (C and D) Time-resolved confocal PL lifetime maps of the (C) target and (D) reference $\text{FA}_{0.83}\text{Cs}_{0.17}\text{Pb}(\text{I}_{0.9}\text{Br}_{0.1})_3$ films. Scale bar, $2 \mu\text{m}$.

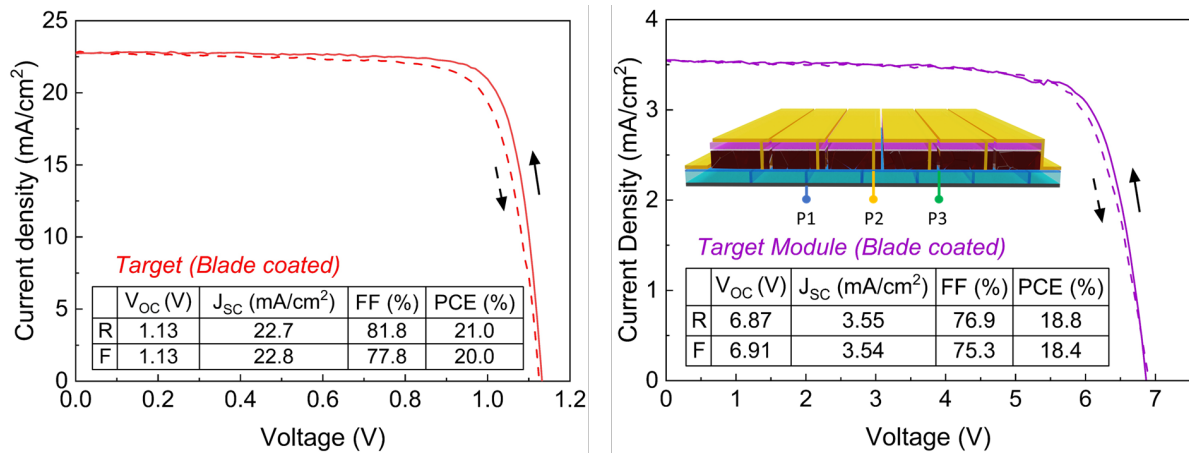


Fig. 3 J-V characteristics of champion blade coated $FA_{0.83}Cs_{0.17}Pb(I_{0.9}Br_{0.1})_3$ device (Mask area: 0.16 cm^2) and mini-module (Mask area: 10 cm^2)

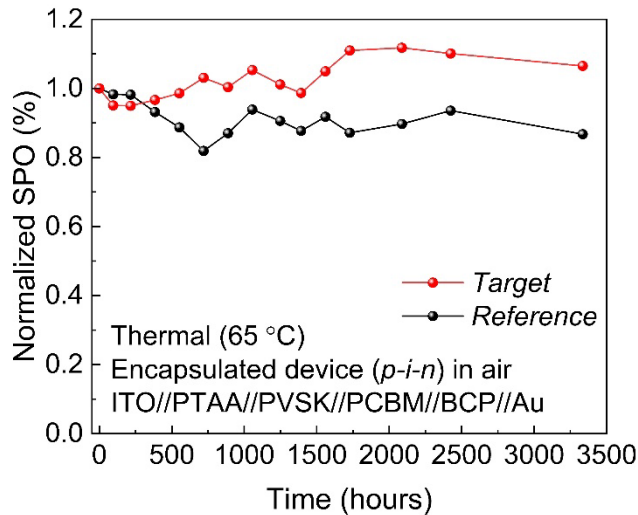


Fig. 4 Thermal stability of inverted PSCs (p-i-n) (encapsulated device aged at 65°C in an ambient atmosphere under dark conditions).