Advanced partial nucleation for single-phase $\text{FA}_{0.92}\text{MA}_{0.08}\text{PbI}_3$ -based high-efficiency perovskite solar cells

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ABSTRACT To date, extensive research has been carried out, with considerable success, on the development of high-performance perovskite solar cells (PSCs). Owing to its wide absorption range and remarkable thermal stability, the mixed-cation perovskite $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ (formamidinium/methylammonium lead iodide) promises high performance. However, the ratio of the mixed cations in the perovskite film has proved difficult to control with precursor solution. In addition, the $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ films contain a high percentage of MA^+ and suffer from serious phase separation and high trap states, resulting in inferior photovoltaic performance. In this study, to suppress phase separation, a post-processing method was developed to partially nucleate before annealing, by treating the as-prepared intermediate phase $\text{FAI-PbI}_2\text{-DMSO}$ (DMSO: dimethylsulfoxide) with mixed FAI/MAI solution. It was found that in the final perovskite, $\text{FA}_{0.92}\text{MA}_{0.08}\text{PbI}_3$, defects were substantially reduced because the analogous molecular structure initiated ion exchange in the post-processed thin perovskite films, which advanced partial nucleation. As a result, the increased light harvesting and reduced trap states contributed to the enhancement of open-circuit voltage and short-circuit current. The PSCs produced by the post-processing method presented reliable reproducibility, with a maximum power conversion efficiency of 20.80% and a degradation of ~30% for 80 days in standard atmospheric conditions.

Keywords: perovskite solar cell (PSC), mixed cations, partial-nucleation, single phase, defects, stability

INTRODUCTION

In recent years, organic-inorganic perovskite solar cells (PSCs) have made their mark in the field of solar energy conversion materials with a rapid boost in power conversion efficiency (PCE) from 3.8% [1] to 24.2% [2]. The ratio of organic cations, such as FA^+ (formamidinium) and MA^+ (methylammonium), in mixed-cation perovskite has a huge influence on its band gap and stability. The benefit of the FA^+ cation is its wide-angle light absorption, whereas MA^+ is necessary to achieve the useful black phase of mixed-cation perovskite [3–5]. However, several factors were found to be harmful to the preparation of $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$. First, the high annealing temperature (~150°C) led to the decomposition of methylammonium iodine (MAI) molecules into CH_3I and NH_3 gas, where poor surface morphology of the film was usually observed [6]. An increase in the FA^+ content might have benefited the thermal stability, but the undesired non-perovskite (δ -phase) by-product, $\delta\text{-FAPbI}_3$, was formed, which compromised the phase purity of the obtained $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ [5,7]. The observed defects in mixed-cation (MAFA) perovskite usually derive from the impurity phase and poor film morphology, which induce non-radiative recombination and low carrier mobility [8–10]. Moreover, with the increase in FA^+ content, the FA^+ was accommodated easily in the lattice structure, making the preparation of high-quality film extremely difficult [6,11,12].

Recently, $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ perovskite with high FA con-

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tent has been prepared using different methods in the mixed-cation ($\text{FA}_x\text{MA}_{1-x}$) system. However, increasing the FA content in the mixed system has resulted in a change in the quality of the perovskite film. Han *et al.* [13–16] proposed a novel vertical recrystallization technique to prepare high FA content of $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ perovskite. This method achieved an FA content of as high as 0.95, but the highest efficiency reported was an FA ratio of 0.86. Li *et al.* [17] reported a dynamic compositional tuning process to prepare high FA content by dissolving formamidinium iodine (FAI), MAI, PbI_2 , and MAX in specific ratios and producing an $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ perovskite as a result. The best FA ratio from this method was stated to be 0.8, although the highest was 0.9. The mechanism of this method involved spontaneous ion exchange in the precursor solution. However, the mixed precursor-based processes remained difficult to control and proved to be insufficient in the production of high-quality perovskite films with high FA content. In addition, these films contain vast trap states and result in serious carrier recombination, which attribute to poorer open voltage and circuit current. Therefore, producing a better preparation technique for mixed-cation perovskite film with high FA content without phase separation, which might result in high-quality film and enhanced optoelectronic properties, is challenging. It is important to improve the cation exchange process of mixed-cation perovskites to obtain high-quality and high FA^+ content perovskite film.

In this study, to prepare a single-phase high-content FA-based perovskite, a beneficial technique known as mixed-cation post-substitution (MCP) was introduced to realize “partial nucleation” before annealing. Different from the conventional method of adding mixed cations to the precursor solution, this molecular exchange process is controlled by post-treating intermediate $\text{FAI-PbI}_2\text{-DMSO}$ (DMSO: dimethylsulfoxide) with mixed molecular (FAI/MAI) solution. After FAI/MAI dripping, the film skipped the initial phase transformation, which could be proved by the measurements (optical microscopy and transmission electron microscopy (TEM) images) from the nucleation process. It was important to omit the initial stages because a shorter nucleation process would reduce the probability of phase separation. $(\text{MAI})_x(\text{FAI})_{1-x}\text{-PbI}_2\text{-DMSO}$ is produced by the MAI substituting the FAI in the intermediate phase. Upon annealing, the formed $\text{MAI}_x\text{FAI}_{1-x}\text{-PbI}_2\text{-DMSO}$ undergoes further phase transformation, leading to the black $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ perovskite (α -phase) being a smooth and compact film. Compared with the existing methods, our technique achieved an FA content of 0.92, which is also the optimized ratio for the

highest efficiency (20.80%) of fabricated mixed-cation perovskite. In addition, the prepared devices exhibited excellent stability even after 80 days of exposure to a humid (~25%) environment with ~30% performance degradation.

RESULTS AND DISCUSSION

High-quality $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ perovskite films were prepared using the MCP technique, which involved post-treating on $\text{FAI-PbI}_2\text{-DMSO}$ intermediates with the mixed-cation solution (Fig. 1a) and the as-obtained sample labeled as MCP- $\text{FA}_x\text{MA}_{1-x}$ (x was the calculated ratio of FA in the films). Contrarily, $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ films were also prepared using the traditional mixed precursor method (Fig. 1b); the calculated highest FA ratio of the film using the traditional method was defined as Tra- $\text{FA}_x\text{MA}_{1-x}$.

As presented in Fig. 1a, the perovskite precursor (FAI/PbI_2 (1:1) in DMSO/DMF (DMF: dimethyl formamide)) solution was spin-coated on the pre-sprayed $\text{TiO}_2\text{/FTO}$ substrate [18]. The chlorobenzene (CB) was then dripped as an anti-solvent to form the $\text{FAI-PbI}_2\text{-DMSO}$ [19,20]. The FAI/MAI solution was then rapidly dripped onto the spinning $\text{FAI-PbI}_2\text{-DMSO}$ colloid-like film. After annealing at 105°C for 20 min, the color of the film changed from yellow to black with a smooth surface [21–23]. The X-ray diffraction (XRD) pattern presented in Fig. S1 clearly indicated the presence of 110 and 220 facets, which indexed the mixed-cation (MCP- $\text{FA}_x\text{MA}_{1-x}$) perovskite without phase separation of $\alpha\text{-FAPbI}_3$ or MAPbI_3 . The prepared $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ perovskite images *via* post-treating with different mixed-cation (FAI/MAI) ratios are presented in Fig. S2. It was noted that when treated with different concentrations of FAI/MAI (5, 10, and 15 mg mL^{-1}), the films turned deep black. However, as the concentration of the FAI/MAI increased to 20 mg mL^{-1} , the film color changed from black to reddish brown. This indicated that there existed a non-perovskite by-product that was produced when higher FA content in the mixed-cation solution was dripped over intermediate products and that the by-product restricted the growth of the perovskite films.

Fig. 2 presents the nucleation processes in the MCP and the traditional methods during the thermal annealing stage at different time intervals. In the case of the film prepared using the traditional method, heated at 105°C , the color turned slowly from yellow to dark brown in six stages of different time intervals (Fig. 2a–f). However, it is worth noting that there were only three stages in the MCP- $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ perovskite film, which started from

Figure 1 (a) Schematic illustration of step-wise fabrication process for FA_xMA_{1-x}PbI₃ perovskite film *via* MCP technique (MCP-FA_xMA_{1-x}PbI₃). (b) Traditional method to fabricate mixed cation perovskite (Tra-FA_xMA_{1-x}PbI₃).

Figure 2 Optical microscopy images of (a–f) the Tra-FA_xMA_{1-x} film and (g–i) the MCP-FA_xMA_{1-x} film recorded at different time intervals during annealing stage.

the fourth stage compared with Tra-FA_xMA_{1-x}PbI₃ (Fig. 2g–i). This demonstrates that the MCP method could increase the speed of nucleation and help to promote the nucleation of the perovskite seed even before the annealing stage. When the mixed-cation (FAI/MAI) solution was dripped onto the FAI-PbI₂-DMSO intermediates, the FAI/MAI might substitute the previous FA⁺ and create a new intermediate product, which might be more analogous to the final perovskite structure [5,24]. In the case of the MCP method, the FAI/MAI treatment solution played a role not only by participating in the

substitution reaction but also by reducing the Gibbs free energy of the original FAI-PbI₂-DMSO system [25]. The reaction started spontaneously, and the nucleation began before thermal annealing, which accorded well with Fig. 2.

To further verify the formation of partial nucleation, the TEM images were measured. Fig. 3a shows that the intermediate phase of Tra-FA_xMA_{1-x} was in disarray; it could transform gradually to the partial-nucleation phase (Fig. 3b) and then the perovskite structure while annealing. However, in the case of the MCP-FA_xMA_{1-x},

Figure 3 TEM images of (a) intermediate phase of Tra-FA_xMA_{1-x}PbI₃, (b) MCP induced partial-nucleation ((FAI)_x(MAI)_{1-x}PbI₂-DMSO), (c) FA_xMA_{1-x}PbI₃, (d) XPS of the MCP-FA_xMA_{1-x}PbI₃ powders post-treated with different mixed cation (FAI/MAI) solutions. (e) Raman measurement for the MAPbI₃, MCP-FA_xMA_{1-x}PbI₃ and FAPbI₃ films. (f) The formation mechanism of MCP-FA_xMA_{1-x}PbI₃.

when the FAI/MAI solution was dripped onto the colloidal-like films, it changed at once to the partial-nucleation phase. After annealing, complete perovskite films were achieved with clear, elaborate lattice fringes (Fig. 3d).

The formed intermediates of MCP-FA_xMA_{1-x} after the substitution process were proposed to be (FAI)_x(MAI)_{1-x}PbI₂-DMSO [26], which was proven by X-ray photoelectron spectroscopy (XPS), as presented in Fig. 3d. The shift in the Pb 4f_{7/2} peak from 138.4 to 137.9 eV and the Pb 4f_{5/2} peak from 143.3 to 142.7 eV indicated the change in the binding energy [26]. Previous reports demonstrated that the Pb 4f_{7/2} and Pb 4f_{5/2} peaks of pure MAPbI₃ perovskite were at 137.9 and 143.3 eV, respectively [26]. When additional MAI was doped into the two-dimensional (2D) structure (FAI-PbI₂-DMSO), the binding

energy for Pb was likely to decrease because the high content of substituted MAI could break the dipole moment, given that DMSO and FAI have a similar molecular structure [27]. Compared with the FAI-PbI₂-MAI intermediate, the binding energy for Pb in the FA_xMA_{1-x}PbI₂-DMSO intermediate was likely to reduce as the MAI entered the structure, resulting in a left shift in the XPS spectra, which was also consistent with the Raman measurements in Fig. 3e. The shift in the Raman peak also proved that the dipole moment was weakened [28]. Finally, the pre-deposited FAI-PbI₂-DMSO intermediate layer was able to directly convert into FA_xMA_{1-x}PbI₂-DMSO intermediate by post-substitution, which transformed to the FA_xMA_{1-x}PbI₃ thin film on annealing at 105°C.

In the case of excess or insufficient FAI/MAI con-

centration, the perovskite films showed different FA ratios and different bond vibrations. As presented in Fig. 3f, we have proposed the following formation mechanism of MCP-FA_xMA_{1-x} based on the cation exchange reaction [3]:

The perovskite precursor was in colloidal form (FAI + PbI₂ + DMSO), and when CB was dripped, the free ions (FAI) would combine with PbI₂ to form the intermediate of FAI-PbI₂-DMSO [27]. When the FAI/MAI (IPA) solution was dripped rapidly onto the film, it allowed MA⁺ to enter the 2D structure and to substitute FA⁺ [26]. It was easy for MA⁺ to substitute FA⁺ because the ionic radius of FA⁺ (2.53 Å) is larger than that of MA⁺ (2.17 Å) [5,13]. However, in the directly mixed precursor system, a small number of MA⁺ ions might be inserted into the lattice structure, and this might lead to the formation of separate phases [29,30]. The FA content in the final perovskite film depends on the different ratios of FAI/MAI in the post-treatment solution.

XRD patterns of the perovskite films post-treated by different solutions of FAI/MAI are presented in Fig. S3. It is evident that a noticeable peak at 12.6° indicates the presence of excess PbI₂ in the MCP-FA_{0.28}MA_{0.71} film, suggesting that there is insufficient FAI to react with PbI₂ [31–33]. However, as the concentration of FAI/MAI increased to 10 mg mL⁻¹ in the post-treatment solution, the peak of PbI₂ disappeared, which was due to sufficient FAI/MAI cations reacting with excess lead halide to form MCP-FA_xMA_{1-x} [34].

Fig. 4a presents the sectional XRD patterns of MCP-FA_xMA_{1-x}PbI₃ film post-treated with different con-

centrations of FAI+MAI (IPA). A noteworthy blueshift in the 110 peaks was observed, from 14.15° for the MCP-MA (MAPbI₃) film to 13.98° for the MCP-FA_{0.92}MA_{0.08} film, indicating an enlarged lattice structure to accommodate the FA⁺ [35,36]. However, it was noted that the perovskite (MCP-FA_{0.9}MA_{0.1} and MCP-FA_{0.92}MA_{0.08}) peak (110) showed no further blueshift when post-treated with high concentrations of FAI/MAI (15 and 20 mg mL⁻¹). This proved the difficulty in accommodating more FA⁺ into the 2D lattice structure (FAI_xMAI_{1-x}-PbI₂-DMSO) [37,38]. According to the Bragg equation, the lattice parameter functions as the FA ratio in the MCP-FA_xMAI_{1-x} (Fig. 4b). In addition, the absorption spectra of MCP-FA_xMAI_{1-x}PbI₃ exhibit a redshift as the concentration of the FAI/MAI solution increases (Fig. 4c). Interestingly, in the traditional method, it is difficult to achieve the same broadened absorption edge as in the MCP method (Fig. S4). The MCP method therefore is proved to be more efficient for preparing high FA content MCP-FA_xMAI_{1-x}PbI₃ perovskite [39].

To further analyze the intramolecular exchange properties between FA⁺ and MA⁺, two different MCP-FA_{0.92}MA_{0.08} and Tra-FA_{0.9}MA_{0.1} perovskite films were prepared using the MCP and traditional methods, respectively. The scanning electron microscopy (SEM) images presented in Fig. 5a show that smooth, pinhole-free MCP-FA_{0.92}MA_{0.08} films with large grain size (500 nm) were obtained. Large grain size and compact, even, smooth perovskite film are prerequisites for high-performance devices. Compared with the MCP-FA_{0.92}MA_{0.08} film, the Tra-FA_{0.9}MA_{0.1} film exhibited a rough, uneven surface (Fig. 5b), which could then result in trap states, especially deep-level defects with increased non-radiative charge recombination [27]. Cross-sectional SEM images show that each layer was uniform and could be clearly distinguished (Fig. 5c, d). The estimated thicknesses of

Figure 4 (a) Extended XRD fingerprint region of (110) plane of MCP-FA_xMA_{1-x}PbI₃ films post-treated with different FAI/MAI concentrations. (b) Lattice parameters of MCP-FA_xMA_{1-x}PbI₃ with different FA contents. Red line is the linear fitting of the lattice parameters. The lattice parameter is calculated from the Bragg equation: $2d\sin\theta=n\lambda$ ($\lambda=1.54056$ Å). (c) UV-vis absorbance spectra of MCP-FA_xMA_{1-x}PbI₃ film.

Figure 5 Top-view SEM images of (a) MCP-FA_{0.92}MA_{0.08} film and (b) Tra-FA_{0.9}MA_{0.1} film. Cross-sectional SEM images of (c) MCP-FA_{0.92}MA_{0.08} film and (d) Tra-FA_{0.9}MA_{0.1} film.

both perovskite layers were ~ 400 nm.

To thoroughly investigate the film quality and phase separation, atomic force microscopy and photoluminescence (PL) mapping measurements were taken [40]. As presented in Fig. 6, the surface roughness of the MCP-FA_{0.92}MA_{0.08} film was ~ 16.0 nm (Fig. 6a), whereas the value increased to 25.0 nm for Tra-FA_{0.9}MA_{0.1} (Fig. 6b). This was because the MCP method is beneficial for an even morphology and uniform dense layer [41,42].

Micro PL characterization was carried out for both the MCP-FA_{0.92}MA_{0.08} and Tra-FA_{0.9}MA_{0.1} thin films, which allowed the coupling of the photo-electronic properties with the information on morphology and composition

Figure 6 Atomic force microscope (AFM) images of (a) MCP-FA_{0.92}MA_{0.08} and (b) Tra-FA_{0.9}MA_{0.1}. PL mapping images for peak position of (c) MCP-FA_{0.92}MA_{0.08} and (d) Tra-FA_{0.9}MA_{0.1}. (e) PL mapping intensity image of MCP-FA_{0.92}MA_{0.08} integrated from 800 to 810 nm. (f) PL mapping intensity image of Tra-FA_{0.9}MA_{0.1} film integrated from 715 to 815 nm. AFM area or the active mapping area is $10 \times 10 \mu\text{m}^2$ and each mapping step is $0.2 \mu\text{m}$.

[43]. PL maps with emission wavelengths of 800–810 nm for MCP-FA_{0.92}MA_{0.08} and 715–815 nm for Tra-FA_{0.9}MA_{0.1} are presented in Fig. 6c, d, respectively [8,44]. For the MCP-FA_{0.92}MA_{0.08} film, the range in the main PL peak was wide, which proved the absence of the second phase. However, the main peak in the Tra-FA_{0.9}MA_{0.1} film was in a wide wavelength range, which indicated its non-homogeneous material composition. This demonstrated the presence of the second phase in the Tra-FA_{0.9}MA_{0.1}, because both MA⁺ and FA⁺ could react with PbI₂, which would produce the δ -phase in the films. In addition, the PL intensity in the MCP-FA_{0.92}MA_{0.08} film was quite uniform (Fig. 6e), and in the range of 4.2E5 to 5.0E5, which confirmed the single phase of mixed cations with fewer defects in the MCP-FA_{0.92}MA_{0.08} film. By contrast, the PL intensity decreased in the micro-domain of Tra-FA_{0.9}MA_{0.1} film, and the intensity distribution was uniform, indicating the presence of the second phase.

To further explore the defects in the MCP-FA_{0.92}MA_{0.08} and Tra-FA_{0.9}MA_{0.1} films, time-resolved PL (TRPL) spectra were measured with perovskite films on a glass substrate to avoid charge injection. As presented in Fig. 7a, the charge lifetime was calculated by fitting a double-index model; it was found that the average life-

time of the MCP-FA_{0.92}MA_{0.08} film was longer (15.287 ns) than that of the Tra-FA_{0.9}MA_{0.1} perovskite film (9.202 ns) [2,31,45,46]. This improved carrier lifetime suggested that the enlarged perovskite grains reduced the grain boundaries and that the defects significantly decreased as a result of the MCP-FA_{0.92}MA_{0.08} film's smooth, even surface. The steady-state PL measurement in Fig. 7b further highlighted the broadened absorption edge and reduced non-radiative recombination [47]. The characteristics of the PL were employed to illustrate the defects in the perovskite films, and the space-charge limited current in Fig. 7c was also tested to study the dynamics of bulk defects in perovskite [48]. The onset voltage of trap filled limit (V_{TFL}) was obtained by fitting the respective curves, as follows:

$$V_{\text{TFL}} = \frac{n_t e d^2}{2 \epsilon \epsilon_0}, \quad (2)$$

where the trap density (n_t) is proportional to V_{TFL} because the $ed^2/\epsilon\epsilon_0$ is constant for the same materials [49]. The MCP-FA_{0.92}MA_{0.08} film showed a smaller V_{TFL} value (0.36 V) than the Tra-FA_{0.9}MA_{0.1} film (0.78 V), demonstrating that the trap density of the former was less; this was also supported by the electrochemical impedance

Figure 7 (a) TRPL decay (excitation at $\lambda=485$ nm) and (b) the steady-state PL (excitation at $\lambda=532$ nm) curves of the Tra-FA_{0.9}MA_{0.1} and MCP-FA_{0.92}MA_{0.08} films deposited on the glass substrates. (c) The space-charge-limited current curves are measured with a structure of FTO/TiO₂/perovskite/Au. (d) The EIS spectra of the devices based on MCP-FA_{0.92}MA_{0.08} and Tra-FA_{0.9}MA_{0.1} films, using the structure of FTO/TiO₂/perovskite/spiro/Au.

spectroscopy (EIS) in Fig. 7d [42,50]. According to the equivalent circuit model, the R_s is the transfer resistance of the device, which is the point of intersection of the EIS and the abscissa. Both R_s are similar in Fig. 7d, but the R_{rec} , which represents the recombination resistance, is larger for the MCP-FA_{0.92}MA_{0.08} film than for the Tra-FA_{0.9}MA_{0.1} film [51,52]. Higher recombination resistance would help the MCP-FA_{0.92}MA_{0.08} film to boost photovoltaic performance by effective charge separation at respective collection layers.

The incident photon-to-current conversion efficiency (IPCE) measurements are presented in Fig. 8a; as expected, the absorption edge of the MCP-FA_{0.92}MA_{0.08} is broader than that of Tra-FA_{0.9}MA_{0.1}. The MCP-FA_{0.92}MA_{0.08}-based device exhibited a notable lift in integrated J_{SC} compared with that of the TRA-FA_{0.9}MA_{0.1}-based device, reaching 22.5 mA cm⁻², which is consistent with the actual $J-V$ characteristics. The representative current density-voltage ($J-V$) characteristics of the devices are presented in Fig. 8b and summarized in Table S1. The Tra-FA_{0.9}MA_{0.1}-based device showed a maximum performance of J_{SC} (23.53 mA cm⁻²), V_{OC} (1.04 V), and FF (75.01%), contributing to the highest PCE of 18.43%. In contrast, the MCP-FA_{0.92}MA_{0.08}-based device exhibited an

improvement in photovoltaic parameters, especially V_{OC} and J_{SC} , which were significantly enhanced to 24.19 mA cm⁻² and 1.10 V, respectively. All these improvements are likely to have originated from a broadened absorption edge and reduced deep-level traps to achieve a higher PCE. The statistical trend of PCEs of 26 cells from both methods, together with the Gaussian fitting, is presented in Fig. 8c. The MCP-processed devices showed a higher average efficiency; their efficiency evolution is presented in Fig. 8d. The Tra-FA_{0.9}MA_{0.1}-based device showed a considerable drop, reflecting more facile decomposition of the absorber layer, due possibly to the presence of defects [53–55]. In the MCP-FA_{0.92}MA_{0.08}-based device, the stability curve remained linear with little slope throughout the observed period, which supports the notion of reduced defects, as indicated by the TRPL, PL, and EIS results discussed above.

CONCLUSIONS

Partial nucleation was introduced to synthesize and prepare high-quality single-phase FA_xMA_{1-x}PbI₃ films with high FA content in the lattice structure. It was realized through the MCP method, which effectively promoted the analogous ion-exchange process on a pre-deposited

Figure 8 (a) IPCE spectra of both cases with the corresponding integrated currents. (b) Current density-voltage curves of both kind of mixed cations PSCs with different scan directions. (c) Statistical plot of PCEs (26 samples for each case); (d) Stability plot of devices for 80 days. All the devices were stored in the dark at room temperature (25°C) and humidity of 40%.

intermediate film *via* intramolecular cation substitution. A uniform film with significantly reduced defects and greatly enhanced stability was obtained. It achieved a reproducible efficiency of 20.80% by the MCP method, which was 10% higher than that produced by the traditional method. Our work exposed a useful experimental approach to partially nucleation in advance, which significantly reduced carrier recombination and improved carrier migration and injection properties. This study provides a simplified way of preparing $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ with high FA content with atmospheric stability for 80 days.

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部分预成核以制备单相基于 $\text{FA}_{0.92}\text{MA}_{0.08}\text{PbI}_3$ 的高效太阳能电池

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摘要 目前为止, 对于高性能钙钛矿太阳能电池(PSCs)的开发已经进行了大量的研究, 并取得了显著的成果. 基于 $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ 的混合阳离子钙钛矿具有广泛的吸光范围和显著的热稳定性, 是获得高性能器件的理想材料. 然而, 很难通过前驱体来控制钙钛矿膜中混合阳离子的比例. 此外, 高 MA^+ 含量的 $\text{FA}_x\text{MA}_{1-x}\text{PbI}_3$ 薄膜存在严重的相分离和大量的陷阱态, 导致光伏性能较差. 为了抑制相分离, 本文提出了一种退火前部分成核的后处理方法, 用混合的甲酰胺碘/甲胺碘(FAI/MAI)溶液处理制备的FAI-PbI₂-DMSO中间相, 最终得到的钙钛矿为 $\text{FA}_{0.92}\text{MA}_{0.08}\text{PbI}_3$, 发现由于类似的分子结构引发了钙钛矿后处理薄膜中的离子交换, 从而促进了部分成核, 缺陷大大减少. 因此, 增加的光捕获和减少的缺陷态有助于提高开路电压和短路电流. 用后处理方法制备的PSCs在大气条件下, 最大PCE为20.80%, 80天后降解率为30%, 具有可靠的重现性.