



The stable perovskite solar cell prepared by rapidly annealing perovskite film with water additive in ambient air



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ABSTRACT

In this work, an efficient and stable inverted planar perovskite solar cell (PSC) with water additive has been prepared in ambient air. Adding moderate water additive to $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ precursor solution leads to growth in quality of $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ film, because the boiling point of water is lower than that of N, N-dimethylformamide. The annealing temperature of the $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ film is controlled to 105 °C (slightly higher than the boiling point of water) to obtain the dense perovskite film rapidly in ambient environment with ~ 40% relative humidity. The highest power conversion efficiency (PCE) of PSC with water additive is 14.02%, higher than that (11.17%) of the reference device without water additive. In addition, the aqueous $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ solution is transformed into the stable perovskite film through the method of rapid annealing. The stable $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x \cdot n\text{H}_2\text{O}$ system can resist the decomposition of perovskite film and weaken iodide ion migration in PSCs, which enhance stability of the device under the atmospheric environment. After being stored in the air for 120 h, the PSC device with water additive still has an PCE of 12.01%, higher than that (1.64%) of the reference device.

1. Introduction

Solar energy, new renewable energy, has aroused people's extensive interest and attention. In recent years, the efficiency of lead halide perovskite solar cells (PSCs) has been improved rapidly, thereby becoming one of the hottest research areas throughout the world. $\text{CH}_3\text{NH}_3\text{PbX}_3$ exhibits a strong light absorption of photon whose energy is greater than the forbidden band width (1.5 eV for $\text{CH}_3\text{NH}_3\text{PbI}_3$, and 2.3 eV for $\text{CH}_3\text{NH}_3\text{PbBr}_3$, etc.). What's more, the absorption coefficient of $\text{CH}_3\text{NH}_3\text{PbX}_3$ is closer to that of amorphous silicon [1]. The low exciton binding of the perovskite material can separate photogenerated electron-hole pairs within the material at room temperature [2,3]. With the low recombination rate and high carrier mobility rate in the perovskite material, electron and hole have a considerable diffusion length [4,5], which greatly contribute to the development of the high-efficiency, low-cost and simple-preparation solar cells. The first organic-inorganic lead halide perovskite solar cell with the power conversion efficiency (PCE) of 3.81% was reported in 2009 [6]. Subsequently, the research on PSCs has made remarkable breakthroughs [7–10]. At present, the highest and certified PCE of 22.1% in the PSC device has been achieved [11]. With increasingly improving efficiency of PSCs, top priority has been given to the stability [12,13]. What's more, the

theoretical-experimental results provide a new method that can improve the structural stability performance of PSCs [14].

In general, the crystal quality of perovskite film is mainly influenced by the solvent choice, precursor composition, deposition temperature, and so on. The morphology and coverage of crystalline perovskite film could be improved by controlling nucleation and growth of the perovskite [15–17]. The crystallinity of perovskites could be enhanced by adding some additives to the precursor solution, which results in a better efficiency [18–20]. High-performance PSCs are conventionally fabricated in a glove box to strictly avoid interference of water and oxygen because the perovskite films would be degraded gradually with time in air [21–23]. A dry atmosphere significantly increases costs of preparation of PSCs, while the ambient air can substantially simplify preparation processes, lower costs and promote commercial production of PSCs. In recent years, increasing researches have been conducted to fabricate PSCs under ambient air, which implies that the moisture plays an important role in the growth of performance of cells [24,25]. Furthermore, many studies have noted that the appropriate content of water can contribute to the crystallization of perovskite during the annealing process [26,27]. What's more, some studies explain how the water affects the stability of cells [28–30].

In this study, appropriate water additives are added to the

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perovskite precursor solution by mixing anhydrous N, N-dimethylformamide (DMF) and deionized water together. What's more, an effective and stable PSC device is prepared by optimizing annealing temperature of the perovskite film in the air environment. Compared with the reference device without water additive (11.17%), the device with water additive has a better PCE of 14.02%. More interestingly, after being stored in the air for 120 h, the unencapsulated device without water additive still has a PCE of 12.01%, which maintains 85.7% of the initial efficiency. Whereas, the reference device just maintains 14.7% of the initial efficiency. Adding suitable water additives to perovskite precursor solution can lead to a better stability of the device in ambient air, compared with the reference devices.

2. Experimental

2.1. Materials and preparation

Chlorobenzene (C_6H_5Cl), hydrogen iodide (HI) and methylamine (CH_3NH_2) were purchased from Shanghai Chemical Industry Co.. Poly (3, 4-ethylene dioxy-thiophene)-poly (styrene sulfonate) (PEDOT:PSS) was bought from Heraeus (Germany). $PbCl_2$ (99.999%) and DMF (anhydrous, amine free; 99.9%) were purchased from Alfa-Aesar. Phenyl- C_{61} -butyric acid methyl ester (PCBM) and 4,7-diphenyl-1,10-phenanthroline (Bphen) powders were produced by Nichem Fine Technology Co., Ltd. (Taiwan). CH_3NH_3I was synthesized by the chemical reaction of CH_3NH_2 and HI [31]. Aqueous DMF solvent was prepared by adding different volume ratios of deionized water to DMF. $CH_3NH_3PbI_{3-x}Cl_x$ solution was fabricated by solubilizing CH_3NH_3I and $PbCl_2$ in aqueous DMF solvent with a molar ratio of 3:1, and agitating the compound at 60 °C for 12 h in ambient air. The $CH_3NH_3PbI_{3-x}Cl_x$ solutions were filtered by a 0.45 μm Polytetrafluoroethylene (PTFE) filter before fabrication. PCBM solutions were manufactured by dissolving PCBM powder into chlorobenzene and churning the mixture for 12 h in ambient air, with an optimum concentration of 20 mg mL⁻¹. Bphen solutions were prepared by lysing Bphen powder into ethanol and rabbling the admixture for 12 h in ambient air, with an optimized concentration of 0.5 mg mL⁻¹.

2.2. Device and characterizations

13 × 15 mm ITO-coated glass (Fine Chemicals Industry Co. 10 Ω sq⁻¹) was etched through method reported [32]. ITO-coated glass was cleaned by ultrasound in deionized water, ethanol and acetone for 15 min or more, respectively. After being washed, the substrate was treated in UV-Ozone cleaner for 10 min. A PEDOT:PSS film was spin-coated onto ITO substrate at 3500 rpm for 40 s and then annealed at 140 °C for 20 min. In ambient air, the $CH_3NH_3PbI_{3-x}Cl_x$ precursor solutions with and without water were spin-coated on PEDOT:PSS films at 2500 rpm for 40 s respectively. Perovskite layer films were dried at 105 °C for 40 min, not through typical gradient increased temperature method [33,34]. Then, the PCBM and Bphen solutions were deposited sequentially by spin-coating at 2000 rpm for 40 s and dried at 60 °C for 10 min. Finally, the substrates were transferred to a vacuum chamber for evaporating silver electrode (defining single device area of

6.25 mm²). Fig. 1 shows the preparation process of PSCs with water additives in ambient air.

The surface morphology of $CH_3NH_3PbI_{3-x}Cl_x$ films and section morphology of PSC were characterized by a field emission scanning electron microscope (FESEM, Quanta 200 FEG, FEI Co.). The Energy dispersive spectroscopy (EDS) measurements with elemental mappings and EDS spectra were carried out by an EDS device connected to FESEM. The current density–voltage (J – V) curves of PSCs were measured (2400 Series Source Meter, Keithley Instruments) under simulated Air-Mass (AM) 1.5 sunlight at 100 mW/cm² (Newport, Class AAA solar simulator, 94023A-U). The incident-photon-to-current efficiency (IPCE) measurement was carried out through the system that combines the monochromator, chopper xenon lamp, and lock-in amplifier with calibrated silicon photodetector. Steady-state photoluminescence (PL) measurements were conducted by using an Edinburgh FLS980 fluorescence spectrophotometer. The X-ray diffraction (XRD) patterns of $CH_3NH_3PbI_{3-x}Cl_x$ films were recorded on a Rigaku D/MAX-2400 diffractometer. X-ray photoelectron spectroscopy (XPS) measurements of $CH_3NH_3PbI_{3-x}Cl_x$ films were performed by using an AXIS Ultra instrument (Kratos UK) at a base pressure of $\sim 10^{-8}$ Torr and 295 K.

3. Results and discussion

In order to study the role of water additive in the crystal growth of perovskite films and the stability of PSC devices in ambient air with $\sim 40\%$ relative humidity, we has fabricated a conventional inverted planar heterojunction architecture of indium tin oxide (ITO)/PEDOT:PSS (40 nm)/ $CH_3NH_3PbI_{3-x}Cl_x$ (270 nm)/PCBM (50 nm)/Bphen (10 nm)/Ag (120 nm) by the method mentioned above. The cross-section FESEM image of PSC prepared is shown in Fig. S1. Bphen film between PCBM and Ag cathode is used as an efficient material of interfacial modification [35]. To analyze the effect of water additives on performance of cells prepared in ambient air, we use the PSCs without water additives as reference device.

The photoelectric performance parameters of PSC device have significantly been improved by adding water additives. Fig. 2a shows the J – V characteristics of the PSCs with and without water additives. Reference device has a PCE of 11.17%, short circuit current density (J_{SC}) of 18.65 mA cm⁻², open circuit voltage (V_{OC}) of 0.90 V and a fill factor (FF) of 0.69, while device with water additives has a better PCE of 14.02%, J_{SC} of 20.19 mA cm⁻², V_{OC} of 0.95 V and FF of 0.73. By the method of adding water additives, significant improvements in J_{SC} and V_{OC} were achieved, which resulted in the increase of PCE from 11.17% to 14.02%. In order to analyze the hysteresis behavior of PSCs, the devices are measured by applying a forward and reverse bias at a scan rate of 0.01 V/s. J – V curves of devices with and without H₂O are shown in Fig. S2. Compared with reference device, the devices with H₂O show a weaker hysteresis. To confirm the enhancement of the J_{SC} in the PSCs with water additives, the IPCE spectra of the PSCs with and without water additives is measured (Fig. 2b). The IPCE intensity of PSC is higher than that of the reference device, within a wide wavelength range between 360 and 750 nm. The J_{SC} 20.05 mA cm⁻² of the PSCs with water additives is calculated via IPCE intensity, which is within the allowed error range, compared with J_{SC} 20.19 mA cm⁻² from the J – V curve. In order to understand nonradiative recombination in the perovskite films, the steady-state PL spectra and time-resolved PL spectra of perovskite films with and without water additives are shown in Fig. 2c and d, respectively. An enhancement in steady-state PL indicates that the nonradiative decay is significantly suppressed through water additives. Furthermore, time-resolved PL clearly shows that water additives have exerted an important influence on PL lifetime. The perovskite film with water additives gives an increased PL lifetime of about 63.4 ns, compared with the perovskite film without water additives (41.1 ns). Here, the steady-state PL and time-resolved PL measurements show that the nonradiative recombination channels are greatly inhibited, therefore the number of defects is reduced by adding

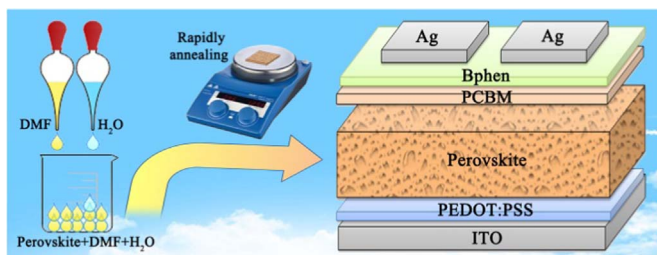


Fig. 1. The operation process chart of preparing PSC with water additive in ambient air.

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