



Optimizing film morphology and crystal orientation of perovskite for efficient planar-heterojunction solar cells by slowing crystallization process



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ABSTRACT

The morphology of perovskite film plays a key role in determining photoabsorption, carrier transport and ambient stability in perovskite solar cells. However, due to the fast crystallization kinetics, the perovskite film fabricated by solution-processing technique usually suffers from low crystallinity and poor orientation. Here, both the morphology and the ambient stability of perovskite films were improved through regulating crystallization rate and enhancing intermolecular interaction by adding α , ω -diamino poly (ethylene glycol) (PEG-NH₂) into precursor solution (mixture of CH₃NH₃I and PbCl₂). The PEG-NH₂ and Pb²⁺ tended to form a new chelated compound Pb²⁺-PEG-NH₂. The stable multidentate chelation between Pb²⁺ and PEG-NH₂ retarded the crystal growth rate to half of the one of the film without PEG-NH₂, which is determined by the *in situ* UV–vis absorption spectra and *in situ* two-dimensional detection capability. The slow crystallization process is beneficial for realizing a uniform perovskite film with enhanced crystallinity almost 3 times of the pristine film, and increased crystal size from smaller than 100 nm to larger than 300 nm. In addition, the content of perovskite crystals adopted (h 0 0) orientation increased from 9.59% to 20.70% due to the flexible PEG-NH₂ chains. Moreover, the ambient stability of perovskite films was also enhanced because the Pb-halogen bond in perovskite crystals became stronger as well as the film became defect-free. As a result, the device fabricated with PEG-NH₂ attained a high efficiency of 15.6% with remarkably prolonged ambient stability without encapsulation. This work demonstrates that introducing the polymer that can coordinate to Pb²⁺ is a feasible way to simultaneously enhance both efficiency and stability of perovskite solar cells.

1. Introduction

Hybrid solar cells based on organometal halide perovskites solar cells have attracted much attention because of the low-cost, ease of fabrication and excellent photovoltaic properties [1,2]. The power conversion efficiency (PCE) of perovskite solar cells has been improved from the initial value of 3.8%–22.7% during the past decade [3–5]. Although the record for the highest efficiency perovskite solar cell was achieved based on the mesoporous architecture, some fundamental issues such as how to manufacture high efficiency devices with long ambient stability have to be addressed urgently to meet the commercial requirement.

The poor perovskite morphology has been cited as very detrimental to device performance, because it not only causes electrical shorting but also has adverse effect on charge dissociation/transport/collection [6].

Controlling nucleating and crystallizing process during film deposition and annealing process is an attractive route to improve perovskite film quality. The heterogeneous nucleation induced by chloridion, polymers or poor solvent in the perovskite precursor film can greatly improve the crystal size, crystallinity and coverage of the film [7,8]. On the other hand, promoting homogenous nucleation by using a mixed solvent, such as N,N-dimethylformamide (DMF), γ -butyrolactone (GBL) and 1,8-diodooctane (DIO), or increasing the solution concentration can also produce smooth perovskite film with uniform crystal domains [9–12]. In addition, slowing down the crystallizing process of perovskite is another way to improve the crystal size and film coverage [13,14]. Not only the film quality but also the crystal orientation has profound influence on device performance, because it partially determines the carrier transport of the device. It has been demonstrated that the (0 0 1) crystal orientation is good for the carrier transportation, which

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permits the carrier transporting along the Pb-I covalent bond [15,16]. Liang et al. produced perovskite crystal highly oriented at (100) facet (which is the same to (001) crystal orientation due to the perovskite crystal belongs to orthorhombic crystal) and improved the PCE to 16.6% [17]. Tsai et al. attained PCE of 15% via tuning the crystal orientation by using slow crystallization method [14].

Despite the success in boosting the efficiency of perovskite solar cells, the ambient stability is still a challenge and it was demonstrated by the degradation of perovskite and the falling down PCE values [18]. The perovskite degradation can be caused by ingress of moisture, oxygen, exposure to UV illumination, and high temperature [19]. The extrinsic efforts researchers can do is fabricate the perovskite in a controlled atmosphere and adding a good encapsulation to avoid the factors that cause the degradation of perovskite. Guangda and his coworkers used aluminum oxide to modify the perovskite, inhibiting the perovskite degradation from moisture [20]. Using $\text{CH}_3\text{NH}_3\text{PbBr}_3$ or FAPbI_3 (FA represents formamidinium) instead of $\text{CH}_3\text{NH}_3\text{PbI}_3$ can also improve the moisture and thermal stability of perovskites [21,22]. Adopting stable Gold (Au) electrode can also improve the device stability compared that of silver (Ag) electrode [23]. Furthermore, the intrinsic stability of the perovskite is equally important to the encapsulation technology promotion. The intrinsic stability represents the perovskite crystal structure stability. Jong Hong and his coworkers improved the non-encapsulated perovskite stability by blade coating fabrication method [24]. Liang et al. prolonged the perovskite crystal degradation time by using solvent vapor annealing processing method [17]. We can infer that the improved stability was attributed to the optimization of the film morphology, which is enhanced crystallinity, enlarged crystal size and compact crystal packing. Hence, we conclude that the film morphology is of crucial importance for device performance and stability.

In this study, we improved the device performance and ambient stability by incorporating PEG-NH₂ into the perovskite precursor solution. We demonstrated the PEG-NH₂ tended to chelate to Pb^{2+} , forming a new Pb^{2+} -PEG-NH₂ chelated compound. The Pb^{2+} -PEG-NH₂ compound retarded the kinetics in both nucleation and crystal growth process of perovskite, thus enhanced the film crystallinity and crystal size. In addition, the multidentate chelation of PEG-NH₂ acted as scaffold for perovskite crystals and confined the crystal orientation, leading to an increased amount of perovskite crystals adopted (100) orientation and compact packing of perovskite crystals. The improved film quality and crystal orientation are beneficial for photoabsorption, carrier extraction and carrier transport, ultimately resulting in an increased performance of planar-heterojunction perovskite solar cells. Furthermore, the optimized film morphology can also inhibit the degradation of perovskite in the air, thus improves the ambient stability of perovskite crystals and the device.

2. Experimental section

2.1. Materials

N,N-dimethylformamide (DMF) was purchased from Sigma-Aldrich. Isopropanol and chloroform were obtained from Beijing Chemical Factory, China. Methylammonium iodide (MAI) and lead chloride (PbCl_2) were purchased from Yingkou Optimal Choice Trade, China. [6,6]-phenyl-C61-butyric acid methyl ester (PC_{61}BM) was purchased from American Dye Source. The bis-functionalization of C70 (Bis-C70) was purchased from 1-Material, Canada. α , ω -diamino poly (ethylene glycol) (PEG-NH₂) (molecular weight, $M_w = 10\,000$; polydispersity index, $\text{PDI} = 1.3$) was synthesized in our laboratory. All materials were used as received.

2.2. Device fabrication

The solar cells were fabricated on ITO-coated glass substrates. The

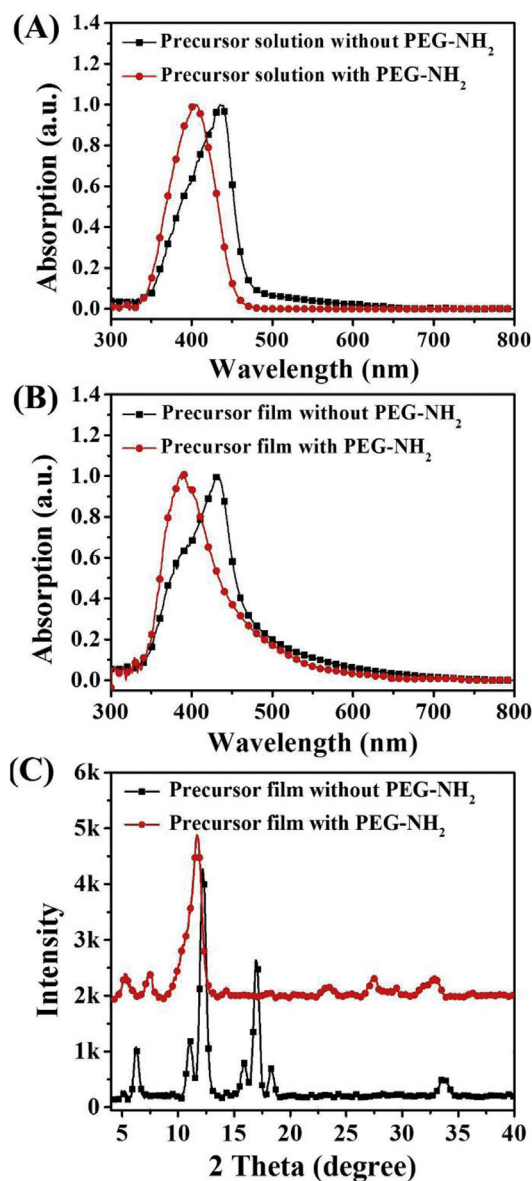


Fig. 1. UV-vis spectra of precursor solutions (A) and precursor films (B) with and without PEG-NH₂. (C) XRD spectra of perovskite precursor films with and without PEG-NH₂.

ITO-coated glass substrates were first cleaned with detergent, then ultrasonicated in deionized water, acetone, and isopropanol, respectively, then dried by nitrogen flow. After treating the ITO substrate with UV ozone for 25 min, a 30 nm thick poly-(ethyleneedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, Baytron P4083) layer was spin-coated on this well-cleaned ITO glass and dried at 140 °C for 30 min. The substrates were transferred into a N₂-filled glovebox. Then the solution containing a mixture of MAI:PbCl₂ (MAI: PbCl₂ with a molar ratio of 3:1 in DMF and the total concentration is 30 wt%) with different content of PEG-NH₂ was spin-cast on top of the PEDOT:PSS layer to produce a 300 nm thick active layer and annealed at 80 °C for 5 h. Afterward, the PC₆₁BM (15 mg/mL in chloroform) and C70-bis surfactant (1 mg/mL in isopropanol) were sequentially deposited on the active layer by spin coating at 3000 rpm for 60 s, respectively. Finally, a layer structure of Al (100 nm) was deposited at top of the active layer by thermal evaporation in a vacuum of 2×10^{-4} Pa to complete the device fabrication.

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