

Improvement of $\text{CH}_3\text{NH}_3\text{PbI}_3$ thin film using the additive 1,8-diiodooctane for planar heterojunction perovskite cells



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ABSTRACT

The thin-film quality is critical for obtaining high-performance perovskite solar cells (PSCs). The additive 1,8-diiodooctane (DIO) was used to control the morphology and structure of $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite thin films, and planar heterojunction (PHJ) PSCs with an architecture of ITO/PEDOT: PSS/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ /PCBM/Al was fabricated. It was found that the DIO played an important role on $\text{CH}_3\text{NH}_3\text{PbI}_3$ thin-film quality and the performance of PHJ-PSCs. With the optimal volume ratio of 2%, the compact and uniform high-quality $\text{CH}_3\text{NH}_3\text{PbI}_3$ thin films with enhanced crystallinity and less roughness were achieved, resulting in the great improvement of power conversion efficiency (*PCE*) from about 4.5% to over 9.0%. The research results indicate that the additive DIO is a simple and effective method to produce high-quality perovskite thin film and accordingly develop high-performance PHJ-PSCs.

1. Introduction

The researchers have been focused on the development of photovoltaic materials and devices during the past several decades, including silicon solar cells, multiple compound solar cells, dye-sensitized solar cells (DSSC), organic solar cells (OSCs), and organic-inorganic hybrid perovskite solar cells (PSCs), and so on. The silicon solar cells have been commercialized, but the preparation process requires high energy consumption and expensive equipment, resulting in the cost of roughly 10 times higher than that of traditional power generation [1–3]. The low-cost, solution-processed PSC have attracted much attention and developed as a rising star since being initially explored by Miyasaka et al. in 2009 with a power conversion efficiency (*PCE*) of around 3.8% [4]. Then the *PCEs* were dramatically improved from 3.8% to about 10.0% in 2012, about 15.0% in 2013, and over 20.0% in 2015, resulting from the great development in thin film fabrication, device engineering, interface progress and physics mechanism [5–7]. To date, the *PCE* of PSCs has reached 22.1% [8].

There are normally two kinds of device configurations, i.e., mesoporous architectures and planar heterojunction architecture. However, the mesoporous layer requires a high-temperature treatment over 450 °C, which is not compatible with flexible and low-cost devices fabricated by large-scale roll-to-roll (R2R) printing process [9–17]. Then low-temperature, planar heterojunction (PHJ) PSCs have been

developed by several groups [18–22]. In particular, the low-temperature processed PSCs using poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT: PSS) as a hole transport layer and the fullerene-derivative phenyl-C61-butyric acid methyl ester (PCBM) as the electron transport layer have gained much attention [21,22]. These interface layers are compatible with low-cost printable or R2R manufacturing in the future, and they can avoid hysteresis under different scanning conditions. In addition, some organic components could be employed as the additives to control the morphology of perovskite films [23,24]. The main obstacles countered in perovskite thin film fabrication is the control of the crystallization process and its effect on the film quality. Non-uniform perovskite morphology has been proved to be very inadvisable to the device performance because it does not only cause electrical shortening but also harmfully impact charge dissociation, transport and recombination [25–31]. Solvent additives have efficient enhancements in the device performance of bulk-heterojunction (BHJ) OSCs by modulating BHJ morphology [32–34]. Therefore, it is possible to improve the morphology of perovskite thin film by the additives for fabricating high-performance PSCs. Herein, the additive 1,8-diiodooctane (DIO) was added into $\text{CH}_3\text{NH}_3\text{PbI}_3$ solution for controlling thin film morphology, crystallinity, and final device performance [16,35]. Based on the optimal volume ratio of 2%, compact and uniform $\text{CH}_3\text{NH}_3\text{PbI}_3$ thin film with enhanced crystallinity and smaller roughness was obtained, resulting in the improvement of *PCEs* based on low-

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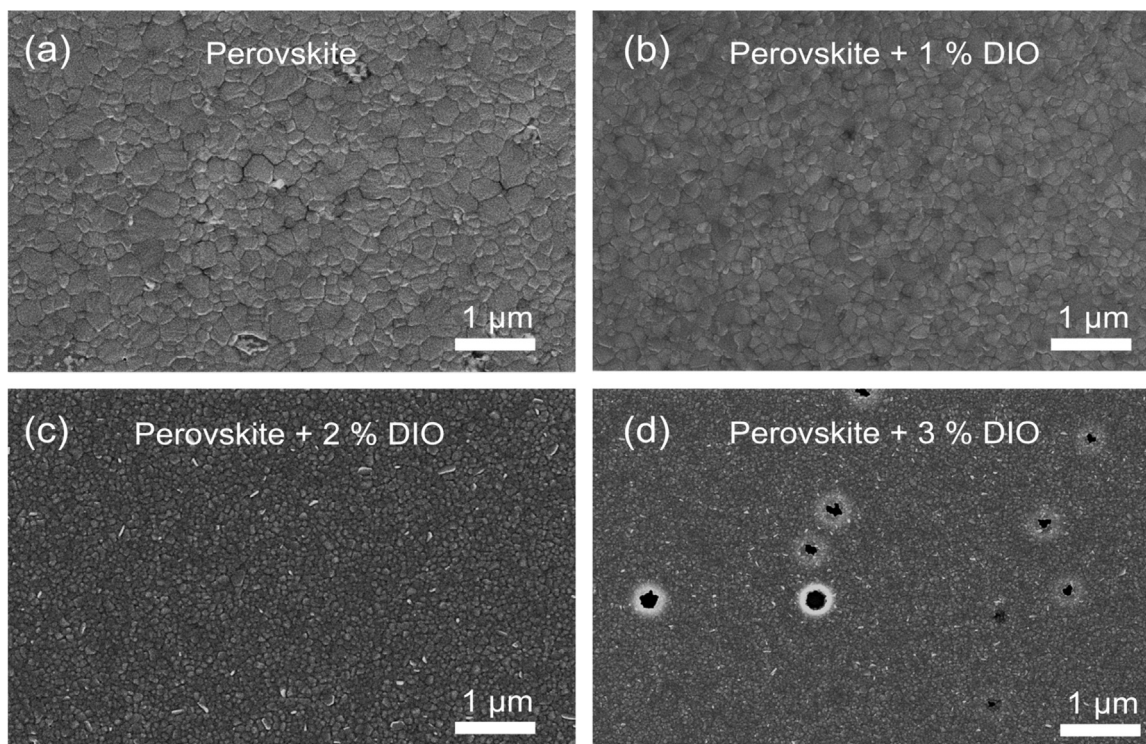


Fig. 1. SEM morphology images of $\text{CH}_3\text{NH}_3\text{PbI}_3$ thin films without (a) and with DIO additive at 1% (b), 2% (b) and 3% (c).

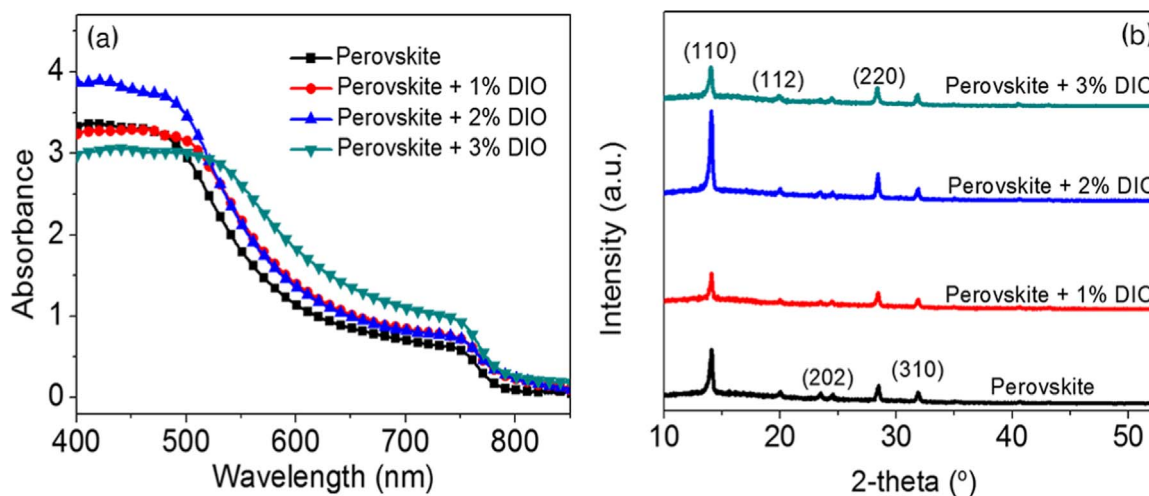


Fig. 2. (a) UV-Vis absorption spectra of $\text{CH}_3\text{NH}_3\text{PbI}_3$ films with different volume ratio of additive DIO. (b) XRD patterns of $\text{CH}_3\text{NH}_3\text{PbI}_3$ films with different volume ratio of additives DIO.

temperature, solution-processed PHJ-PSCs from about 4.5% to over 9.0%.

2. Experiments

The methylammonium iodide ($\text{CH}_3\text{NH}_3\text{I}$, jingge, Wuhan) and lead iodide (PbI_2 , Zhengpin, shanghai) were dissolved in dimethylformamide (DMF, J & K Seal) with a concentration of 450 mg/ml ($\text{CH}_3\text{NH}_3\text{I}:\text{PbI}_2 = 1:1$). The additive 1,8-diiodooctane (DIO), purchased from (Sigma & Aldrich), was added into $\text{CH}_3\text{NH}_3\text{PbI}_3$ precursor solution with the different molar ratio. The fullerene derivative [6, 6]-phenyl-C61-butyric acid methyl ester (PCBM, Dye Source, American) was dissolved in chlorobenzene (CB, J & K Seal) with a concentration of 15 mg/ml. All solutions were vigorously stirred for 12 h at 70 °C in gloves box.

The ITO substrate was cleaned by ultrasonically in acetone,

detergent, distilled water and isopropyl alcohol for 15 min, respectively. The substrate was dried by N_2 flow and was treated the UV-ozone for 20 min. The PEDOT: PSS was coated on the clean substrate at 3000 rpm for 30 s, and treated at 150 °C in air for 15 min. The PEDOT: PSS coated substrate was moved into gloves box for depositing $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite layer. The perovskite film was fabricated by one-step deposit methods. i.e., the perovskite precursor solution was spin-coated onto the top of PEDOT: PSS film at a speed of 4000 rpm for the 30 s, and then perovskite film was annealed at 100 °C for 10 min. Then, the PCBM layer was deposited onto the perovskite film at 3000 rpm for the 30 s. Finally, an Al electrode with a thickness of about 100 nm was deposited onto the PCBM layer, resulting in an active area of 0.09 cm^2 .

Scanning electron microscope (SEM, FEI Helios Nanolab 600i, USA) was used to study the morphology of $\text{CH}_3\text{NH}_3\text{PbI}_3$ films. Ultra violet-visible

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