



Enhancing the perovskite solar cell performance by the treatment with mixed anti-solvent



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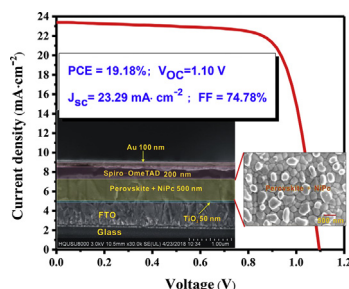
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HIGHLIGHTS

- A simplified anti-solvent treatment is developed for high-quality perovskite films.
- The high-quality film improves the charge separation and transport efficiency.
- Power conversion efficiency of the solar cell is enhanced from 17.4% to 19.18%.

GRAPHICAL ABSTRACT



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ABSTRACT

At present, the surface modification of perovskites by solvent treatment has become the most effective way to improve the performance of planar perovskite solar cells. Here, phthalocyanine nickel as an additive was introduced into the anti-solvent chlorobenzene to form a mixed anti-solvent. Prior to completion of the perovskite crystals, a mixed anti-solvent was infiltrated into the perovskite surface to improve the interfacial contact of the perovskite with the hole-transporting layer. This method improved the crystal quality of the perovskite film, thus enhanced the charge transfer efficiency, and suppressed the recombination of carriers effectively. The perovskite solar cell constructed with this perovskite films treated by the optimal concentration of nickel phthalocyanine in the anti-solvent solution yielded a power conversion efficiency of 19.18% and the filling factor of 74.38% under $100\text{ mW}\cdot\text{cm}^{-2}$ illuminations.

1. Introduction

In recent years, organic-inorganic hybrid perovskite materials have triggered a wave of research in the field of solar cells. Since Miyasaka's team first started using $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite in solar cells in 2009,

the energy conversion efficiency of the perovskite solar cells (PSCs) have increased from 3.8% to 22.1% [1–7]. Halogenated perovskite materials have become the “dream material” for photovoltaic applications due to their high optical absorption coefficient, high carrier mobility and long carrier diffusion length.

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At present, the most successful organic-inorganic perovskite material has an ABX₃ structure comprised of an organic cation, A = (methylammonium CH₃NH₃⁺; formamidinium CH₃(NH₂)₂⁺); Cs⁺, a divalent metal, B = (Pb²⁺; Sn²⁺; Ge²⁺), and an anion X = (Cl⁻; Br⁻; I⁻; SCN⁻) [8–10]. The perovskite solar cell structure has a classical mesoporous n-i-p junction and a mesoporous “planar” structure [11]. Planar heterojunction PSCs have been extensively used as one of the most efficient configurations of PSCs. The improvement of the properties of PSCs mainly depends on the crystal quality, morphology and interfacial contact of perovskite films. Currently, perovskite films are prepared by one-step spin-coating [12], dip coating [13], two-step interdiffusion [14], and vacuum assisted evaporation [15]. Among them, the one-step spin-coating method is an effective process to regulate the morphology, the grain size and the crystal quality of the perovskite film by dropping the anti-solvent (chlorobenzene, toluene, ethyl acetate) during the spin coating.

Generally, interfacial engineering is an efficient approach to solve the problems of defects and the energy-barrier mismatch, thus improving the photovoltaic performance of PSCs [16–21]. For example, further annealing in dimethylformamide steam increased the size of the perovskite grains [22]. Removal of the remaining chlorobenzene in the perovskite film with isopropyl alcohol solution reduced the grain boundary density of the perovskite film, resulting in a smooth surface topography for the large-grained perovskite [23]. The improvement of interface engineering improved the crystal quality of perovskite films, enhanced the charge transport, and effectively suppressed the carrier recombination [24–26]. Esmail Nouri et al. used soluble tetrabutyl copper phthalocyanine as a hole transporting material, and graphene oxide as a blocking split channel to promote the transfer of holes between the perovskite and the hole transporting layer [27]. Phthalocyanine is a p-type semiconductor, which is widely used in thin film field effect transistors and photovoltaic devices because of its high hole mobility, thermal stability and chemical stability [28–30]. As a hole transport layer (HTL) with long-term durability, its derivatives have been widely used in PSCs [31,32].

In this work, we prepared the mixed anti-solvent by doping phthalocyanine nickel (NiPc) with anti-solvent chlorobenzene (CB) and fabricated the perovskite thin films in a glove box by one-step spin-coating to study the influence of different concentration of NiPc on the photoelectric properties of perovskite solar cells. It made the perovskite phase film closely contact with the HTL and reduced the defects of the perovskite interface, which was favorable for extracting charge carriers and improving the photovoltaic performance. The PSC devices based on the NiPc yielded very competitive power conversion efficiency (PCE) up to 19% under 100 mW cm⁻² illuminations.

2. Experimental

2.1. Materials

Anhydrous alcohol (99.7%) was from Sinopharm Chemical Reagent Co, Ltd. Acetonitrile (99.8%), 1-Butanol (99.8%), *N,N*-Dimethylformamide (DMF, 99.8%), dimethyl sulfoxide (DMSO, 99.8%), chlorobenzene (99.8%), *tert*-butyl pyridine (TBP, 96%), Nickel phthalocyanine (85%), bis(trifluoromethane) sulfonamide lithium salt (Li-TFSI, 99.95%) and titanium diisopropoxide bis(acetylacetonate) (75 wt % in isopropanol) were purchased from Sigma-Aldrich. Cesium Iodide (CsI), formamidinium iodide (FAI) and methylammonium bromide (MABr) were purchased from Xi'an Polymer Light Technology Corp, China. Lead iodide (PbI₂, 99.9985%) were purchased from Tokyo Chemical Industry. The Spiro-OMeTAD as HTL was purchased from Luminescence Technology Corp, Taiwan, China. All materials were used without further purification unless otherwise stated. All steps were performed under ambient conditions, but the perovskite and hole-transport material solutions were mixed inside an Argon glove box.

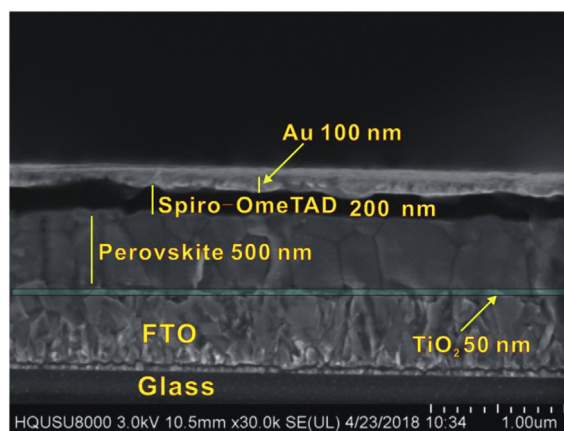


Fig. 1. The cross-sectional FE-SEM image of a complete perovskite solar cell.

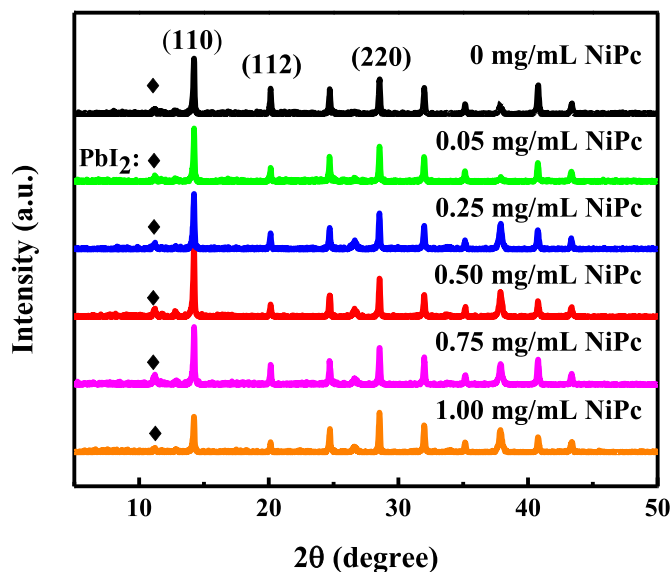


Fig. 2. XRD patterns of the CB and NiPc/CB-treated perovskite films.

2.2. Preparation of TiO₂ photoanode

Fluorine-doped tin oxide (FTO)-glass substrates etch into the desired device with a laser etcher prior to cleaning and then cleaned by ultrasonication in Hellmanex (2%, deionized water) for 30 min, rinsed thoroughly with acetone, isopropanol, de-ionized water and ethanol (each for 10 min), consecutively. Afterwards, the FTO glasses were treated with UV ozone for 30 min and plasma cleaning for 5 min respectively. To prepare a dense TiO₂ electron transport layer, the cleaned FTO glass was coated with a TiO₂ solution by a two-step spin-coating method with commercial titanium diisopropoxide bis(acetylacetonate) (75% in 2-propanol, Sigma-Aldrich) diluted in 1-butanol solution (3:40, volume ratio), the first step was 700 rpm for 5 s with an acceleration of 1000 rpm s⁻¹, the second step was 2500 for 15 s with a ramp-up of 2000 rpm s⁻¹. The compact TiO₂ coated FTO substrates were dried at 120 °C for 15 min and annealed at 550 °C for 30 min and then allowed to cool down to room temperature slowly. The thickness of the obtained TiO₂ electron transport layer (ETL) is about 50 nm.

2.3. Preparation of perovskite

A perovskite precursor solution was obtained through using one-step spin-coating procedure. The Cs_{0.05}(MA_{0.17}FA_{0.83})_{0.95}Pb(I_{0.83}Br_{0.17})₃ precursor solution was prepared containing PbI₂ (1.1M),

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