



The influence of perovskite layer and hole transport material on the temperature stability about perovskite solar cells



Haiying Zheng^{a,b}, Guozhen Liu^{a,b}, Changneng Zhang^a, Liangzheng Zhu^{a,b}, Ahmed Alsaedi^c, Tasawar Hayat^{c,e}, Xu Pan^{a,*}, Songyuan Dai^{a,d,*}

^a Key Laboratory of Photovoltaic and Energy Conservation Materials, Institute of Applied Technology, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei 230031, China

^b University of Science and Technology of China, Hefei 230026, China

^c NAAM Research Group, Department of Mathematics, Faculty of Science, King Abdulaziz University, Jeddah 21589, Saudi Arabia

^d State Key Laboratory of Alternate Electrical Power System with Renewable Energy Sources, North China Electric Power University, Beijing 102206, China

^e Department of Mathematics, Quaid-I-Azam University, Islamabad 44000, Pakistan

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ABSTRACT

Organometal halide perovskite has recently emerged as a promising solar cell material. However, the instability of perovskite solar cells when exposed to moisture, light, oxygen, UV and temperature remains a key challenge to the commercialization. Among these factors, temperature instability has been a major obstacle to fabricating the long-term operational device. In this study, we tried to explore the effect of perovskite layer and hole transport material on the temperature stability of solar cell at different three temperatures (20, 60 and 85 °C). The thermogravimetric analysis and absorption intensity variation characterization results on the perovskite layer indicated that the perovskite layer is not the main factor that leading to the temperature instability. Furthermore, temperature-dependent X-ray diffraction measurements indicated that the temperature has no remarkable effect on the crystal structure of perovskite materials. We also performed an aging test of the device by removing additives from spiro-OMeTAD HTM and the solar cells exhibited an improvement of temperature resistant, but it did not solve the problem of temperature instability. Finally, by using P3HT without additives to replace spiro-OMeTAD as HTM, we achieved the devices which can remain initial PCE after 800 h at 20, 60 and 85 °C and display outstanding temperature resistant. These findings demonstrated that the temperature instability of perovskite solar cells is mainly affected by HTM and additive, not the perovskite layer.

1. Introduction

Recently, organic–inorganic halide perovskite solar cells have achieved rapid development because of their low-cost input materials, superior material properties such as tunable bandgap, high open-circuit voltage (V_{oc}), long electron–hole diffusion lengths, and excellent absorption. These make them very attractive for solar energy. Since 2016, the power conversion efficiency (PCE) of organic–inorganic halide perovskite solar cells has surpassed 22%, bringing substantial interest in the technology (Grätzel and Park, 2014; Kim et al., 2012; Barrows et al., 2016; Yang et al., 2015; Bi et al., 2016; Burschka et al., 2013; Kojima et al., 2009).

Despite PSC has high PCE, the problem of instability under moisture, light, oxygen, UV and temperature has hampered its commercial development. There are many reports about improving the

stability of perovskite solar cells (Saliba et al., 2016; Aitola et al., 2017; Zhao et al., 2017; Oku et al., 2014; Kim et al., 2016; Chang et al., 2016; Nam et al., 2017; Zhang et al., 2017; Sun et al., 2017). Saliba et al. (2016) fabricated the temperature stable perovskite solar cells which can maintain 95% of their initial performance at 85 °C for 500 h under full illumination by the incorporation of rubidium cations and polymer-coated. Based on carbon nanotube hole contact, Aitola et al. (2017) achieved the high temperature-stable perovskite solar cell that shows slight PCE loss with an extrapolated lifetime of 580 h. Via introducing an ALD oxide electron extraction layer, Kim et al. (2016) reported the heat-resistant hybrid perovskite photovoltaics that exhibit a stable operation over at least 10 h when subjected to 100 °C in ambient environments. Whereas these studies do not clearly show the influence factors of stability and fundamentally solve the problem of instability.

Among these unstable factors, the humidity instability and UV light

* Corresponding authors at: State Key Laboratory of Alternate Electrical Power System with Renewable Energy Sources, North China Electric Power University, Beijing 102206, China (S. Dai).

E-mail addresses: xpan@rntek.cas.cn (X. Pan), sydai@ncepu.edu.cn (S. Dai).

instability can be improved by encapsulating and adding UV filter membrane, respectively (Han et al., 2015; Niu et al., 2014). However, the temperature instability is determined by the material itself and there are still no good measures to avoid at present. Therefore, the temperature instability became the major challenge on the development of perovskite solar cells (Kim et al., 2015; Misra et al., 2015; Ming et al., 2016). The research results demonstrate that perovskite materials may start to transform from the tetragonal into cubic structure at a slight high temperature (Niu et al., 2015; Bush et al., 2016; Juarez-Perez et al., 2016; Song et al., 2015; Berhe et al., 2016; Dualeh et al., 2014). Especially, Dualeh et al. (2014) reported that the high temperature can accelerate the decomposition of MAPbI₃ and then impede photovoltaic processes. It was found that the degradation may happen at high temperature, when spiro-OMeTAD with additives was used as hole transporter material (HTM) (Hawash et al., 2015; Ono et al., 2014; Jung et al., 2015; Wang et al., 2015). These all may result in the declining of photovoltaic performance. Hence, we need to understand which layer mainly leads to the temperature instability of perovskite solar cells and further exclude the causes of instability for enhancing the temperature resistant.

Here, we took the perovskite (FAPbI₃)_{0.85}(MAPbBr₃)_{0.15} (Simplified as FA) or Cs⁺ doping which is often used as perovskite precursor with high PCE and more temperature resistant than MAPbI₃ (Aharon et al., 2015) as perovskite layer. Whereas, the unsealed and sealed devices based on FA and FA with 10% Cs⁺ exhibited poor temperature resistant. Then, we performed thermogravimetric analysis (TGA), temperature-dependent X-ray diffraction (XRD) measurements and absorption intensity variation at different temperatures of normal perovskite. The results indicated that perovskite layer has neglectful influence on the temperature stability at less than 100 °C. To further test the effect of HTM, it was found that the device without additives in HTM exhibited the improvement of temperature stability, but the PCE decreased to about 20% after 800-h storage at 85 °C. Furthermore, we achieved outstanding temperature stable device which can remain initial PCE after 800 h at 20, 60 and 85 °C by changing spiro-OMeTAD into P3HT without additives. The results indicated that HTM and additives in HTM have enormous effects on the temperature stability, while the perovskite layer has slight influence at less than 100 °C.

2. Experimental

2.1. Preparation of formamidine iodide

The preparation of formamidine iodide (FAI) was by reacting formamidine acetate with hydroiodic acid in the round-bottom flask, and then the reactants were stirred at 0 °C for 2 h. The light-yellow powder was gained from rotary evaporation of the solvent at about 60 °C under reduced pressure. Then the product was purified by dissolving in methanol and collected by air pump filtration. The purification process was conducted twice to get pure FAI powder. The product was completely dried at about 60 °C in vacuum oven.

Other materials were purchased from Alfa without further purification.

2.2. Device fabrication

FTO-coated glass was cleaned by sonication in detergent and rinsed three times with ultrapure water and ethanol. A TiO₂ compact layer was coated on the FTO substrate by spray pyrolysis at 450 °C and the precursor solution is included with 0.4 mL bis(acetylacetonate) and 0.6 mL titanium diisopropoxide in 7 mL isopropanol. TiO₂ mesoporous layer was spin-coated on TiO₂ compact layer with the speed of 4000 r.p.m. for 20 s, from a 30 nm TiO₂ paste that is dispersed by ethanol (TiO₂:ethanol = 1:5.5). Then, mesoporous TiO₂ were slowly sintered from room temperature to 510 °C for 3 h. The 1.4 M Pb²⁺ precursor solutions of (FAPbI₃)_{0.85}(MAPbBr₃)_{0.15} were gained from dissolving the corresponding component powders in DMSO and DMF mixed solvent (DMSO:DMF = 1:4 by volume) with stirring at 70 °C for 30 min. Next, the precursor solutions were spin-coated onto the mesoporous layer at firstly 1100 r.p.m. for 15 s, secondly 4600 r.p.m. for 33 s in an air flowing glovebox. About 0.2 mL of chlorobenzene was quickly dripped on the substrate after the second step for 20 s. The as-prepared film was annealed at 100 °C for 50 min on a hotplate. The HTM solution with spiro-OMeTAD (73 mg), 4-tert-butylpyridine (29 μL), Li⁺ salt (17 μL) and cobalt(III) salt (8 μL) in chlorobenzene was deposited onto the substrate at 3000 rpm for 20 s. At last, about 60 nm of Au were coated by thermal evaporating on the HTM layer.

2.3. Characterization

Thermogravimetric analysis (TGA) was recorded on TG/DTA 7300 (SEICO INST.). The measurement was carried at rate of 10 °C/min under a under ambient N₂. Common XRD and temperature-dependent XRD patterns of the perovskite films were carried out by an X'Pert PRO (PANalytical) in the 2θ range 5–70° at room temperature. Absorption spectra were measured on an ultraviolet–vis (UV–vis) spectrophotometer (U-3900H, HITACHI, Japan). *J*–*V* curves and PCE were recorded by using a solar simulator (Newport, Oriel Class A, 91195A) with a source meter (Keithley 2420) at 100 mW/cm² illumination AM 1.5G. The active area of the device was 0.09 cm² by masking a black mask. The temperature aging tests were performed in three containers with 20, 60 and 85 °C, respectively. The containers remained less than 10% RH and keep in dark.

3. Results and discussion

Firstly, we studied the PCE variation of unsealed and sealed FA perovskite solar cells with common mesoporous structure (shown in Fig. 1a) at different temperatures. Fig. 2 shows the normalized efficiency variation curves of unsealed and sealed perovskite solar cells at 20, 60 and 85 °C, respectively. The starting photovoltaic performance and *J*–*V* curves are exhibited in Table S1 and Fig. S2. As demonstrated

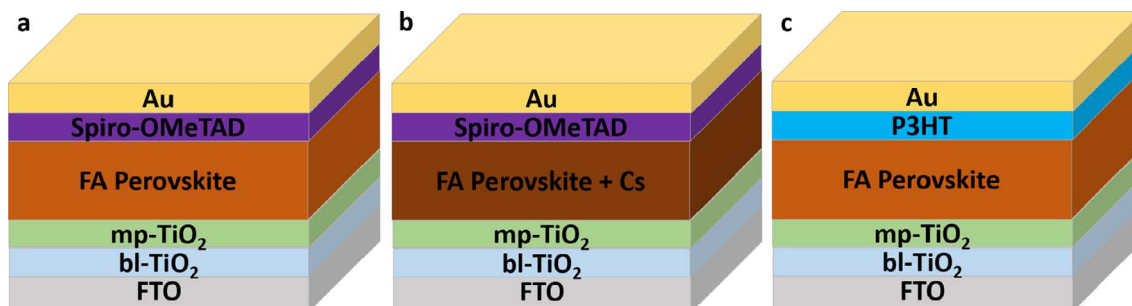


Fig. 1. Schematic structure of perovskite solar cells studied in this work, (a) FA perovskite layer (spiro-OMeTAD as HTM with additives), (b) FA perovskite layer containing Cs⁺ (spiro-OMeTAD as HTM with additives), (c) FA perovskite layer (P3HT as HTM without additives).

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