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Efficiency enhancement by changing perovskite crystal phase and adding a charge extraction interlayer in organic amine free-perovskite solar cells based on cesium

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ABSTRACT

Improvements of overall photoconversion efficiencies under simulated 1 sun illumination are reported for inorganic perovskite solar cells based on cesium lead halide, CsPbI₃, as perovskite absorber material. By modifying the perovskite crystal structure and inserting a particular hole extraction interlayer, a remarkable efficiency enhancement is achieved from 0.09% that was reported previously to 4.68%. Additionally, we demonstrated that the nature of the perovskite crystallinity as well as the charge extraction material play a decisive role in the establishment of the degree of hysteresis. Particularly, fundamental analysis verified that upon the existence of MoO₃ layer as a hole extraction interlayer, the *I*–*V* hysteresis effect was suppressed. Therefore, this model proposes that the hysteresis is also originated by the interface phenomena and not only by the perovskite film or the structure of the solar cell, mesoporous and planar, effects as have been reported. Our experimental observation suggests that the morphology of the perovskite film in addition to selective interface cathode electrode play a critical role for recording an enhancement of photoconversion efficiencies in perovskite solar cells.

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1. Introduction

Organic–inorganic hybrid-perovskite solar cells have made an impressive progresses in last few years, mainly by their high power conversion efficiencies over 20%, profit-making, and easily manufactured for marketing purposes [1]. The best efficiencies that have been recorded with these perovskite materials use a general formula such as AMX₃, where A is the organic component cation as methylammonium (MA), M is the metal cation (Pb) and X corresponds to the halide (I) or a mixture of halides (Cl, Br, I) anions [1,2]. Currently, the perovskite solar cell research is focused on finding new routes to synthesize new perovskite absorbers which improve their optical and physical properties as well as their stability issue. To that end, one of the components that can be exchanged instead of MA is organic cations such as ethylammonium and formamidium in order to alter the optical and electronic properties [3,4]. However, reports based on inorganic cations in AMX₃ perovskite structure has been infrequent. In particular, all-inorganic cesium lead halide perovskite (CsPbI₃) solar cell shows improvements in the optical and electrical properties,

keeping the fundamental characteristics of the MAPbI₃ [5]. However, it has been reported that CsPbI₃ achieves poor photovoltaic properties (0.09%) and the combination of MA and Cs was still required to show better photovoltaic performances [5].

Additionally, inorganic cesium lead halide perovskite materials have attracted considerable attention because they do not show dipoles; inversely correlated with the MAPbI₃ because the MA cation may be the origin of the hysteresis in the current–voltage (*I*–*V*) curve. This phenomena is still under debate due to several possible sources may cause the hysteresis in the *I*–*V* characterization which leads over- and underestimated efficiency values [6]. This phenomenon is attributed to the slow dynamic processes because high capacitances at low frequency range are detected by impedance spectroscopy under light in perovskite solar cells [7,8]. This issue has been addressed from different points of view for organic–inorganic hybrid-perovskite solar cells (MAPbI₃), such as experimental processes [9], architecture/interface phenomenon [9,10], ion migration [11], trapping of charge carrier [10], and ferroelectricity [12,13].

Here we report improvements of photoconversion efficiencies for amine free-lead perovskite solar cells (CsPbI₃) fabricated on meso-porous TiO₂ (mp-TiO₂) as scaffold layer under AM1.5G light intensity. Several annealing temperatures for the perovskite film were checked in order to achieve an appropriate perovskite crystal structure. Other important parameter to achieve better overall

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efficiencies is the electrode materials in order to extract efficiently the charge. Mainly, this manuscript focuses on the hole extraction layer. Considering the energy level matching, molybdenum oxide (MoO_3) was selected as a suitable material to extract hole carrier of the cesium perovskite solar cells. In addition, to understand the impact upon the shape of the I - V curves for CsPbI_3 solar cells was undertaken. The hysteresis phenomenon was controlled by effective charge extraction interfaces.

2. Experimental details

2.1. Materials

All chemicals were used as received without any further purification. The CsPbI_3 was synthesized in our laboratory, using the precursors cesium iodide (CsI, 99.9%) and lead (II) iodide (PbI_2 , > 98.0%) from Alfa Aesar, and TCI Tokyo Chemical Industry CO., LTD, respectively. The precursors were solved with dimethyl sulfoxide (DMSO) from Sigma-Aldrich ($\geq 99.5\%$). The hole transport material (HTM) was regioregular poly(3-hexylthiophene-2,5-diyl) (P3HT) from Sigma-Aldrich (99.995%). The HTM solution was solved with chlorobenzene (Anhydrous, 99.8%, Sigma-Aldrich).

2.2. Preparation of cesium perovskite films

A mixture of CsI and PbI_2 in DMSO as a solvent (20% weight) was stirred at 60 °C until complete dissolution. An aliquot of 100 μL of perovskite solution was spin coated at 2000 rpm for 30 s. This perovskite film required an annealing treatment according to the sequence of (i) at 60 °C for 30 min and (ii) at 350 °C for 30 min to acquire the appropriate crystal structure. Additionally, other morphologies of CsPbI_3 perovskite were also analyzed, such as the perovskite formation was spin coated twice or three times depending on the experiment, being the second surface heating carried out at 150 °C, 250 °C or 350 °C for 30 min.

2.3. Device fabrication

The substrates were FTO (Fluorine-doped Tin Oxide) coated glass (Nippon Sheet Glass Co. Ltd, 10 Ω/sq) with dimension of $2.5 \times 2.5 \text{ cm}^2$ and were etched with Zn powder (Wako) and 6 M HCl aqueous solution. The surface was cleaned by ultrasonification and plasma system. The TiO_2 back layer (bl- TiO_2) was deposited on the FTO glass by spray pyrolysis at 300 °C of the precursor solution titanium diisopropoxide bis (acetylacetonate) (75 %wt isopropanol, Sigma-Aldrich) diluted in ethanol (Super-dehydrate, 99.5%, Wako) (1:39 by volume ratio), using nitrogen as a carrier gas. The mesoporous TiO_2 , mp- TiO_2 , paste (PST-30NRD pursued by JGC Catalysts and Chemicals Ltd.) was diluted by ethanol (1:7 by weight ratio) and deposited on the substrates by spin coating at 5000 rpm for 30 s. After drying on the hot plate at 120–150 °C for 10 min, the film was annealed into the oven at 500 °C for 30 min. Immediately afterwards, the mp- TiO_2 films were transferred into a glove box and all following processes were carried out under nitrogen atmosphere. On the top of the mp- TiO_2 film, the perovskite film was synthesized as stated above. Then, an aliquot (100 μL) of P3HT solution with 15 mg/mL of concentration was deposited on the top of the perovskite film by spin coating at 1000 rpm or 2000 rpm for 30 s. Once the film of the HTM is completely dry, an annealing heating was required at 130–150 °C for 5 min. Finally, thermal evaporation was used to deposit $x \text{ nm}$ MoO_3 (being x either 0 or 8 nm) and 60 nm Au for the back contact. All solar cells were encapsulated using poly(methylmetacrylate) (PMMA) by spin coater. In Fig. 1 is shown the architecture of the CsPbI_3 -based solar cells with 8 nm MoO_3 as an electron blocking layer.

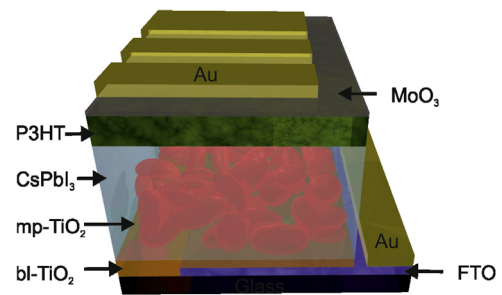


Fig. 1. Schematic architecture of the cesium perovskite solar cell as glass/FTO/bl- TiO_2 /mp- TiO_2 / CsPbI_3 /P3HT/ MoO_3 /Au. A thin MoO_3 interlayer was added between the perovskite and the polymer films as a function of hole transporting and electron blocking material.

2.4. Device characterization

Current-voltage (I - V) measurement. A solar simulator was used to measure the photocurrent-voltage characteristics of the solar cells which was equipped with two light sources at AM1.5G 100 mW cm^{-2} irradiance (CEP-2000SRR, Bunkoukeiki Inc). The irradiance was calibrated using a reference cell of silicon. The photovoltaic performance were recorded using a mask to define an active area of the devices, which was in this case 0.12 cm^2 and 0.08 cm^2 . In order to analyze the hysteric phenomena, two scan directions are desired such as forward sweep (FS) and reverse sweep (RS) at scan rate 0.1 V/s. The FS measurement was carried out from short-circuit to open-circuit and reversely, and the RS measurement was carried out from open-circuit to short-circuit.

Incident photon to current efficiency (IPCE). The IPCE was measured by using CEP-2000SRR (Bunko Keiki) equipped with 300 W Xe lamp. Monochromatic light was exposed by DC mode and the current was taken every 100 ms after the light exposure (10 nm interval). The monochromatic light was adjusted to $1 \cdot 10^{16} \text{ mW cm}^{-2}$ which was monitored by Si photodiode.

X-Ray Diffraction (XRD). The substrates for the XRD measurements were glass/ CsPbI_3 /PMMA which two films of CsPbI_3 perovskite were synthesized at 150 °C, 250 °C, and 350 °C for 30 min. The X-Ray diffractometer was RINT-Ultima III, Rigaku.

Optical characterization. UV-visible spectroscopy was measured using V-670 JASCO spectrophotometer and the structure of the substrates were glass/FTO/bl- TiO_2 /mp- TiO_2 / CsPbI_3 /PMMA. The perovskite CsPbI_3 (2 films) morphology was changed by different annealing treatments at 150 °C, 250 °C, and 350 °C for 30 min.

3. Results and discussion

3.1. Perovskite films characterization

The crystal phase of the CsPbI_3 perovskite film was altered experimentally with the purpose to know how it affects the cell performance, and consequently the photoconversion efficiencies. Under this scenario, a variety of baking temperatures such as 150, 250 and 350 °C were checked in complete perovskite solar cells.

As shown in the X-Ray Diffraction (XRD) patterns in Fig. 2a, CsPbI_3 perovskite film shows a direct dependent between crystallization phase and surface annealing treatment. In particular, the XRD peaks at 150 °C and 250 °C illustrated an orthorhombic pattern (Fig. 2b); in contrast, a cubic pattern (Fig. 2c) was observed for the cell prepared at 350 °C. Additionally, similar results have been reported such as a phase transition at 330 °C from orthorhombic $Pnma$ crystal (δ - CsPbI_3) to cubic $Pm3m$ crystal (α - CsPbI_3) [14]. Therefore, CsPbI_3 obtained at 150 °C and 250 °C was assigned to δ - CsPbI_3 , and the perovskite obtained at 350 °C was referred to α - CsPbI_3 .

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