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A large-area hole-conductor-free perovskite solar cell based on a low-temperature carbon counter electrode

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ABSTRACT

We demonstrate the application of low-temperature carbon counter electrode in fabricating large-area hole-conductor-free perovskite solar cell. A mesoscopic TiO₂ film was deposited on a transparent conductive fluorine-doped tin oxide substrate by screen printing method. Then commercial conductive carbon slurry was printed to prepare the counter electrode at a temperature of 130 °C. The light absorber, CH₃NH₃PbI₃, was formed instantly inside the pores of the entire TiO₂/carbon layer upon single-step deposition of CH₃NH₃PbI₃ precursor. The devices were characterized with XRD, SEM, and UV–vis absorption spectroscopy. As a result, an energy conversion efficiency of 3.3% with an active area of 2.25 cm² was got under AM1.5G condition and there was no obvious decay during a testing period of 624 h. The carbon counter electrode has the features of low-cost and low-temperature preparation, giving it potential for application in the large-scale flexible fabrication of perovskite solar cells in the future

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1. Introduction

Global energy demand has been continually increasing with industrial development and population growth in recent decades, especially for conventional energy resources such as fossil oil, coal and natural gas. Nowadays, over 80% of energy consumption stems from fossil, which will lead to environmental problems and global-warming issue. What's more, because of the excessive exploitation by human being, these fossil energies are on the edge of exhaustion. Perovskite solar cell has caught people's attention since it was proposed for the first time in 2009, because it exhibited excellent property in spectral absorption and carrier transportation [1]. In the last four years, perovskite solar cell has achieved tremendous improvement in photo-current conversion efficiency (PCE) from 3.8% to 19% [2] by using Spiro-MeOTAD or polytriarylamine polymer [3] as the hole conductor. This kind of performance is comparable to that of the commercial silicon solar cells. More importantly, when compared to traditional organic solar cells, perovskite nanocrystalline as the core photoelectric conversion material is cheap and can be made by solution

methods, making it possible for the large-scale fabrication of solar cell. However, compared to other components, the use of hole transfer materials (HTMs) bears a large proportion of cost for such solar cells, limiting the approach to low-cost photovoltaic devices. Recently, the investigations about using CH₃NH₃PbI₃ perovskite nanocrystals as both a light harvester and a hole conductor at the same time were reported, [4–8] implying a great potential to bring down the cost of energy production. In the fabrication processes of perovskite solar cell, noble metal such as Au with the work function –5.1 eV was used for the counter electrodes (CEs), usually prepared by thermal evaporation under high vacuum condition. The high-cost metallic CE and the high energy-consuming evaporation process hinder the industrialization of solar cell to a large extent. Carbon, which is abundant on the earth, is found to have the similar work function (–5.0 eV)[9] and can be used as the CE of solar cells. Ku et al. and Han et al. prepared HTM-free solar cells and achieved a certified power conversion efficiency of 12.8%, in which the CE were screen printed on the photo anode by a kind of special carbon slurry which was composed by carbon black, graphite and ZrO₂ at a mass ratio of 1:3:1 [9–17]. The carbon CE need a high temperature of 400 °C to volatilize the organic solvent. What's more, the flexibility has been a key characteristic gradually for the electronic devices. Many researchers are working on the flexible or wearable energy-harvesting devices by using Au, Ag or Al as the CEs of the solar cells. [18–23] However, the high

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temperature for fabricating the CEs is challenging, as it often exceeds the affordable temperature limit of flexible substrates, such as PET/ITO or PEN/ITO.

Herein, we introduce a low-temperature carbon CEs for fabricating large-area perovskite solar cells, where the CE has a very small square resistance and keeps better contact with the FTO substrate. The morphology and structure for the carbon CE/Mesoscopic TiO₂ heterostructure was examined, and the current-voltage characteristics and monochromatic incident photon-to-electron conversion efficiency (IPCE) were also tested to analyze the performance of the solar cells. As a result, an efficiency of 3.3% under an active area of 2.25 cm⁻² was obtained under AM 1.5 illumination of 100 mW cm⁻². The carbon counter electrode has the features of low-cost and low-temperature preparation, giving it potential for application in the large-scale flexible fabrication of perovskite solar cells in the future.

2. Experimental details

In the experiments, the transparent conductive fluorine doped tin oxide (FTO) glasses with high transparency in the visible range were cut into small pieces of 20 mm × 25 mm and etched by diluted hydrochloric acid (2 mol/L) and zinc powder. The FTO substrates were ultrasonically cleaned in acetone and ethanol for 10 min, respectively, and then rinsed with deionized (DI) water. A hole-blocking layer of compact TiO₂ was deposited by spin-coating a solution of tetrabutyl titanate in ethanol, and annealed at 500 °C for 30 min. Spin-coating was carried out at 2500 rpm for 60 s. After cooling down to room temperature, a 10 μm TiO₂ nanocrystalline layer (DSL 18NR-T, 20 nm, Dyesol, Australia) was deposited on top of the compact layer by doctor-blade method. After that, a commercial conductive carbon slurry (XSF-02) was printed on top of the TiO₂ nanocrystalline layer successively, and a 16 μm mesoscopic carbon layer was formed. The film was drying on the hot plate at 130 °C for 1 h. Finally, 50 μL of the CH₃NH₃PbI₃ precursor (0.395 g CH₃NH₃I and 1.146 g PbI₂ mixed in 2 mL γ-butyrolactone) was dipped on the top of the carbon layer. All

the steps above were carried out in ambient air. The infiltrated devices were then put into the vacuum drying oven for 2 h where the temperature was fixed at 70 °C so as to promote the crystallization of perovskite. During the drying procedure, the color of the devices changed from light yellow to black, implying the accomplishment of the solar cell.

The cross-sectional structure of the solar cells were monitored by the field emission scanning electron microscopy (FESEM, JSM-7600F, JEOL). The formation of the perovskite absorber layers has been further confirmed by XRD analysis (PANalytical PW3040/60) with Cu Kα radiation (λ = 1.5406 Å) from 10° to 60°. The CH₃NH₃PbI₃ was dip-coated on mesoscopic TiO₂ layer with FTO. UV-vis spectrophotometer (UV 2600, Shimadzu) was utilized to obtain the absorption spectra of the perovskite solar cells.

Current density-voltage (*J*-*V*) curves were measured by an electrochemical station (Autolab PGSTAT302N, Metrohm Autolab, Utrecht, The Netherlands) under simulated AM 1.5 sunlight at 100 mW cm⁻² irradiance generated by solar simulator (Oriel 94043A, Newport Corporation, Irvine, CA, USA). The active areas of the cells were 2.25 cm². The incident photon-to-electron conversion efficiency (IPCE) was tested under illumination of monochromatic light from a xenon lamp coupled with a monochromator (TLS1509, Zolix). In order to investigate the stability of the solar cells stored in dry air at room temperature without any encapsulation. Long-term stability test was carried out on one low-temperature carbon CE perovskite solar cell with the initial efficiency of 2.35% for over 600 h.

3. Results and discussion

CH₃NH₃PbI₃, which has a tetragonal perovskite structure, is a relatively underexplored alternative for light-absorption that provides a framework for binding organic and inorganic components into a molecular composite (Fig. 1a).[24] In CH₃NH₃PbI₃ nanocrystals, Pb²⁺ cations adopt the octahedral coordination of I⁻ anions together with the cuboctahedral coordination of CH₃NH₃⁺ anions. [25] As shown in Fig. 1b, a

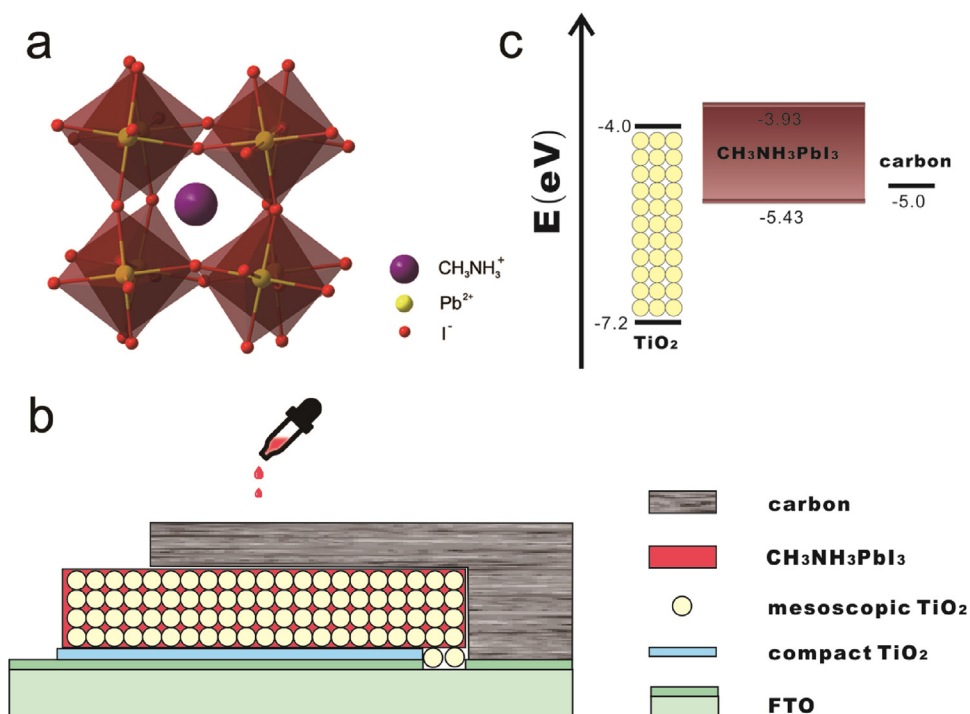


Fig. 1. a) The crystal structure of CH₃NH₃PbI₃ perovskite. b) A schematic structure of a carbon based monolithic device. c) Schematic of the relative energy levels of each layer.

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