



## Boosting efficiency of hole conductor-free perovskite solar cells by incorporating p-type NiO nanoparticles into carbon electrodes

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### ABSTRACT

Carbon-based hole conductor-free perovskite solar cells (PSCs) have attracted great attention due to the simple process, low cost and relatively high stability. However, the power conversion efficiency (PCE) is considerably lower than that of the standard PSCs using spiroOMeTAD as hole-conductor material and Au (or Ag) as counter electrodes. Herein, by means of incorporating p-type NiO nanoparticles into carbon electrodes, the PCE of the hole conductor-free PSCs was significantly boosted to 13.26% from 10.29% of that based on pure carbon electrode, because of the enhancement of hole transfer. In addition, the carbon-based hole conductor-free PSCs showed 85% of the initial efficiency after 800 h in ambient atmosphere. The results indicate that the p-type NiO nanoparticles can enhance hole transfer from perovskite into carbon electrodes, and the carbon electrodes can prevent the water in the air to improve stability. While the PSCs using Spiro-OMeTAD as hole conductor and Ag as electrodes have slightly lower PCE of 13.24% and lower stability. Hence, it is an effective strategy of incorporating p-type nanomaterials into carbon electrodes to enhance the carbon-based hole conductor-free PSCs.

### 1. Introduction

Organic-inorganic hybrid perovskite materials possess excellent optical and electronic properties, such as direct optical bandgap, high absorption coefficient, ambipolar transport, long carrier diffusion length and low excitation binding energy. In addition, due to the organic functional groups the organic-inorganic hybrid perovskite materials can dissolve in various organic solvents, and further the simple low-temperature solution strategy can guarantee the high-quality films. For examples, during annealing the perovskite at about 100 °C, DMF was successfully used to enhance the grain size of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> [1], due the strong coordination effect between DMSO molecules and precursor ions, DMSO was widely taken to retard the crystallization and improve the quality of perovskite films [2], and other solutions such as ethanol [3], acetonitrile [4], and isopropanol [5] were also investigated. Therefore, organic-inorganic hybrid perovskite solar cells (PSCs) have captured widespread attention, and the power conversion efficiency (PCE) has increased to 22% from 3.8% in the recent years [6–9]. Importantly, simultaneously requirements of low cost, high efficiency and

stability for PSCs is guideline to promote the industrial production [10–14].

To date, Spiro-OMeTAD has been usually employed in PSCs as hole transporting materials [15]. However, the expensive Spiro-OMeTAD has low carrier mobility, and additives are needed to enhance charge transfer. Alternatives have been developed, such as CuI, CuSCN, Cu<sub>2</sub>O, NiO and PEDOT:PSS [16–21], on which the expensive Ag or Au have been deposited as electrodes [22,23]. However, the high-cost vacuum technologies and noble metals limit the large-scale applications [24–27]. In addition, the stability is not good under air environment. Thus, a variety of strategies have been proposed to reduce the cost and improve the stability of PSCs. In our previous paper, we summarized the positive roles of carbon materials in the applications at every part of PSCs due to the low cost, extensive sources, high electrical conductivity and chemical stability [28]. The low-cost carbon materials not only enhance the charge transport, but also improve the stability of PSCs. Thus, carbon-based hole conductor-free PSCs have been successfully developed to meet low cost and high stability. And the work function about –5.0 eV makes carbon the ideal electrode for PSCs, and a lot of

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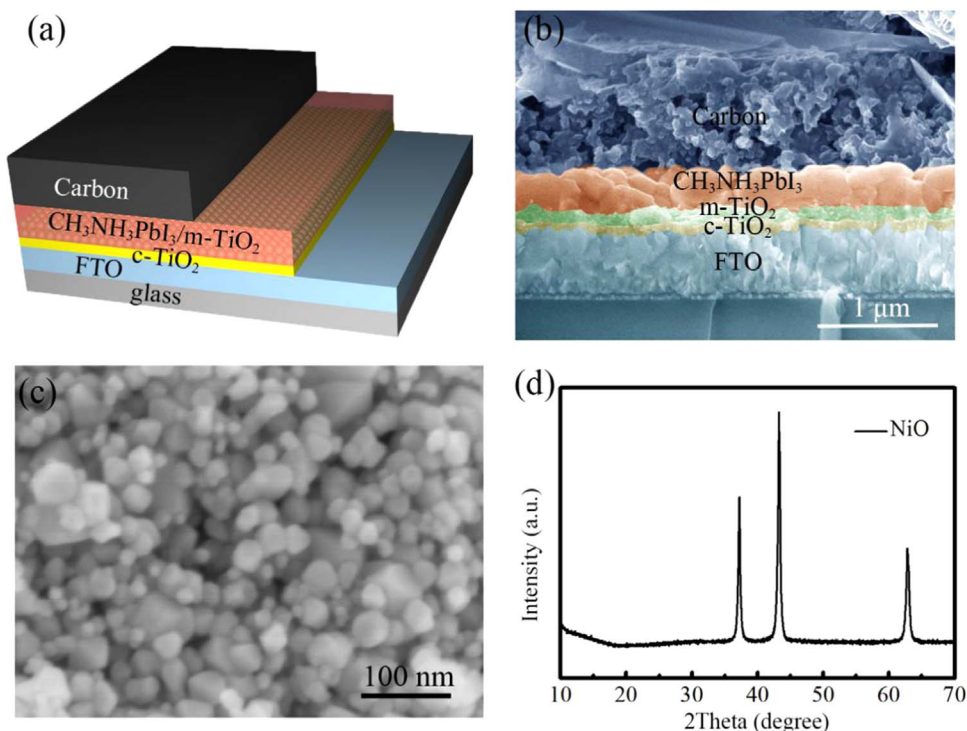


Fig. 1. (a) Schematic representation of the structure of perovskite solar cell as “glass/FTO/c-TiO<sub>2</sub>/m-TiO<sub>2</sub>/CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>/carbon”. (b) Cross-sectional SEM image of the perovskite solar cell. (c) SEM image and (d) XRD spectra of the NiO nanoparticles.

carbon materials have been applied in PSCs [29,30]. Especially, carbon electrodes are successfully applied in the fully printable mesoscopic PSCs, which need high-temperature procedure for carbon electrode [31]. In addition, low-temperature carbon electrode is another trend for PSCs. However, for most low-temperature carbon-based PSCs, the PCE was less than 10%. Therefore, it is significantly important to study the mechanism and develop low-temperature carbon-based PSCs.

In this work, p-type NiO nanoparticles are incorporated into low-temperature carbon electrodes to improve interfacial hole transfer from perovskite to carbon electrode for enhanced hole conductor-free PSCs. For pure carbon electrodes, the PCE of PSCs was 10.29% under AM 1.5 G conditions. With incorporation of NiO nanoparticles, the PCE of PSCs was increased to 13.26%, which is slightly higher than 13.24% of the devices using Spiro-OMeTAD as hole conductor and Ag as electrode. Meanwhile, the stability of the carbon-based hole conductor-free PSCs can keep 85% of the initial PCE after 800 h in ambient atmosphere.

## 2. Experimental details

### 2.1. Fabrication of NiO nanoparticles

NiO nanoparticles with 10–30 nm size were synthesized by a sol-gel method. Typically, 0.02 M Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and 0.04 M oxalic acid were dissolved into 100 ml and 50 ml deionized water under stirring at 50 °C for 1 h and room temperature, respectively. Then, the later solution was dropped-wise into the former. The above mixed solution was heated to form a gel at 100 °C for 30 min under stirring, which was followed to dry overnight in air to form green powder. Lastly, the green precipitate was annealed at 450 °C for 2 h in air to obtain the NiO nanoparticles.

### 2.2. Fabrication of perovskite solar cells

Fluorine-doped tin oxide (FTO, ~7 Ω/□<sup>2</sup>, Nippin) glasses were patterned with 2 M HCl solution and Zn powder, cleaned with deionized water, acetone, and ethanol using an ultrasonic bath for 20 min, respectively, dried under N<sub>2</sub> gas flowing, and treated with ultraviolet-ozone for 20 min. Next, for a compact TiO<sub>2</sub> layer (60 nm), the cleaned FTO glasses were spin-coated on a precursor solution at 2500 rpm for

30 s and dried at 125 °C for 15 min, which were processed by a solution method. Titanium(IV) isopropoxide (369 μl) was dissolved in isopropanol (2.53 ml) under stirring at room temperature, and then a solution of 2 M hydrochloric acid (35 μl) in isopropanol (2.53 ml) was added dropwise. The reaction mixture was stirred for 1 h at room temperature. Before being used, the precursor solution was filtered through a 0.45 μm syringe filter. Then, for a mesoporous TiO<sub>2</sub> layer (120 nm), a diluted TiO<sub>2</sub> paste (Dyesol 18 NR-T, 1: 6 attenuation by weight in ethanol) was spin-coated at 4500 rpm for 25 s, which was dried at 125 °C for 15 min, and annealed at 500 °C for 30 min in air condition. CH<sub>3</sub>NH<sub>3</sub>I powder was synthesized according to previous report [32]. In brief, 30 ml of methylamine (40% in methanol) and 32.3 ml of hydroiodic acid (57 wt% in water) were stirred for 2 h in an ice-water bath. The precipitate was washed with diethyl ether three times, and dried at 60 °C in vacuum overnight. For perovskite precursor, 462 mg PbI<sub>2</sub> and 159 mg CH<sub>3</sub>NH<sub>3</sub>I were dissolved in a mixed solution of 0.3 ml DMSO and 0.7 ml DMF with stirring at 70 °C overnight. The precursor solution was spin-coated on TiO<sub>2</sub> substrates at 500 rpm for 6 s and 4000 rpm for 20 s in a N<sub>2</sub>-filled glovebox. During the spinning process, 0.2 ml toluene as anti-solvent was dripped to rush the film. After being dried at 75 °C for 15 min and annealed at 105 °C for 20 min, a brilliant black film of perovskite film (400 nm) was obtained. Then, a carbon paste directly deposited on perovskite film by a doctor-blading method, and dried at 105 °C for 20 min in air to form carbon electrodes (10 μm). The schematic diagram of doctor-blading technique is shown in Fig. S1 (Supporting information). The dispersion of NiO nanoparticles into carbon paste was achieved by a grind method in an agate mortar. Here, the weight ratio of added NiO nanoparticle powder relative to carbon paste is expressed as A: B, where A and B are the mass of NiO powder and carbon paste, respectively, and which will be represented the electrodes.

### 2.3. Characterization

Morphology of the samples was characterized by scanning electron microscopy (SEM, FEI NOVA NanoSEM 450). The NiO phase was examined for the samples by X-ray diffractometer (XRD, Bruker D8 Advance X-ray diffractometer) using Cu-Kα radiation (λ = 1.5418 Å).

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