



Efficient perovskite solar cells based on novel three-dimensional TiO_2 network architectures

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Abstract Mesoscopic lead halide perovskite solar cells typically use TiO_2 nanoparticle films as the scaffolds for electron-transport pathway and perovskite deposition. Here, we demonstrate that swelling-induced mesoporous block copolymers can be templates for producing three-dimensional TiO_2 networks by combining the atomic layer deposition technique. Thickness adjustable TiO_2 network is an excellent alternative scaffold material for efficient perovskite solar cells. Our best performing cells using such a 270 nm thick template have achieved a high efficiency of 12.5 % with pristine poly-3-hexylthiophene as a hole transport material. The high performance is attributed to the direct transport pathway and high absorption of scaffolds, small leakage current and largely reduced recombination rate at interfaces. The results show that TiO_2 network architecture is a promising scaffold for mesoscopic perovskite solar cells.

Keywords Perovskite · Solar cell · TiO_2 · Template · Atomic layer deposition

1 Introduction

Solar cells directly converting solar energy into electricity have been recognized as an alternative energy source for our planet in the future considering the global shortage of fossil fuels. In recent decades, silicon solar cells have a rather high market share worldwide, but their further development is facing a great challenge to balance the power conversion efficiency (PCE) and the cost. It is known that crystalline silicon solar cells have a relatively high PCE and a high cost. However, thin film silicon solar cells are much cheaper due to the use of fewer raw materials. In spite of this, relatively low PCEs limit their large scale production. Developing solar cells with low cost and high PCEs is in great demand nowadays. Among the various types of solar cells, organic-inorganic hybrid perovskite solar cells are most likely to stand out [1]. On the one hand, perovskite solar cells can be fabricated by solution processing or chemical vapor deposition, both of which are beneficial for cost reduction and mass production [2, 3]. On the other hand, solar cells of this type are undergoing a rapid development since they have taken only about five years to raise the PCE from 4 % to 20 % [4], a value comparable to that of crystalline silicon solar cells. The most fascinating gemini of organic-inorganic hybrid perovskites for solar cells are $\text{CH}_3\text{NH}_3\text{PbI}_3$ and $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$, which have a suitable bandgap about 1.55 eV for visible light absorption. To realize easy fabrication and excellent performance of organic-inorganic hybrid perovskite solar cells, much efforts have been devoted to designing a more suitable architecture and modifying the interface property of different contact layers [5, 6].

Hao Lu and Kaimo Deng contributed equally to this work.

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Typical device architectures of perovskite solar cells include six parts: conductive glass (fluorine-doped tin oxides (FTO) or indium tin oxides (ITO)) substrate, hole-blocking layer, scaffold materials, hole-transport materials (HTM), organometal-halide perovskite layer and metal cathode (Au or Ag). TiO_2 is the most frequently used charge-injecting and hole-blocking oxide due to its large energy band gap, matched energy level and high carrier mobility. In a typical process, two fabrication steps are required before perovskite deposition [7, 8]. First, a compact TiO_2 layer is initially formed on the conductive glass substrate to function as hole-blocking layer. Then, a TiO_2 nanoparticle layer serves as both the electron-transport pathway and mesoporous frame structure for perovskite deposition. Snaith and co-workers [9] reported that Al_2O_3 nanoparticle layer could also function well as the mesoporous frame structure although it is an insulator. The advantage of mesoscopic type solar cells is that the mesoporous nanoparticle layer can favor the formation of a thick perovskite layer, resulting in increased light capture and a high current density. In recent years, the planar type solar cells which do not use the nanoparticles layer have also been studied, for its simple fabrication method [3, 10]. Thus it is quite desirable to achieve a high efficiency of perovskite solar cells with a simple architecture by combining the merits of the planar and mesoscopic type solar cells.

The thickness of TiO_2 mesoscopic scaffolds should be lower than the optimum thickness of the perovskite layer, which is typically about 300–500 nm in consideration of the diffusion length of electrons and holes in perovskite solar cells. This requirement limits the choice of template scaffolds. Owing to the facile synthesis and controlled height, one-dimensional nanorod/nanowire arrays have been widely investigated as potential alternatives to nanoparticles. Bi et al. [11] reported ZnO nanorod arrays based perovskite solar cell with efficiency about 5 %, which was much lower than traditional planar or mesoporous cells. Mahmood et al. [12] presented ZnO nanostructures based perovskite solar cell with the highest efficiency about 10 %, in which a number of growth processes should be controlled. Kim et al. [13] showed perovskite solar cell based on TiO_2 nanorod arrays with an efficiency of 9.4 %. Recently, Fakhruddin et al. [14] reported TiO_2 nanorods based perovskite solar cells with efficiency about 10.5 %, which used an interface engineering during growth processes and an optimized laser pulse for patterning. Therefore, it is challenging to explore more efficient nanoporous materials as mesoscopic scaffolds with excellent hole-blocking and electron-transporting property. Selective swelling-induced pore generation of amphiphilic block copolymers (BCPs) has emerged as a facile and efficient strategy for the preparation mesoporous networks with three-dimensionally interconnected porosities. The thicknesses of the porous networks can be continuously tuned from nanoscale to bulk simply by

controlling the coating techniques and coating parameters [15, 16]. Because of the interconnected mesoscopic porosities and adjustable thicknesses, thus-obtained BCPs porous networks are expected to be used as interesting sacrificial templates for the fabrication of perovskite solar cells with direct charge transport pathways. In order to copy the specific morphology of the BCP templates, it is important to choose an appropriate method for the deposition of TiO_2 on the BCPs templates. Atomic layer deposition (ALD) is a powerful tool for depositing thin film with great accuracy in thickness and uniformity at a controllable rate below 1 nm per cycle. The self-limiting and surface saturated process of ALD endows it the perfect ability to produce conformal ultrathin films, even on materials with high aspect ratios [17–19]. ALD technique is generally more expensive than commonly used sol-gel deposition method and ALD instrumentation is not available in some settings.

Herein, we employ ALD technique to coat TiO_2 films with different thicknesses on swelling-induced porous BCPs templates. After a calcining process, BCPs are removed and mesoporous TiO_2 networks are formed with precisely modulated thicknesses. For the first time, this novel TiO_2 network is used as the mesoporous scaffold for loading perovskite films. The thicknesses of frame can be adjusted precisely for effective infiltration of perovskite precursors. Usually a compact hole-blocking layer is necessary before depositing a mesoporous frame, however, present solar cells do not need this extra step. The $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ and pristine poly-3-hexylthiophene (P3HT) are served as the light absorbing layer and HTM, respectively. P3HT is cheap and easy to use and has been considered as a suitable HTM for perovskite solar cells than spiro-MeOTAD, which needs oxidation or doping treatment in ambient atmosphere. The cell (glass/FTO/ALD- TiO_2 nanostructures/ $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ /P3HT/Ag) shows a PCE as high as 12.5 % compared with 9.83 % of planar TiO_2 cells. It is worthy to note that the achieved efficiency is comparable or even higher than values reported previously in perovskite solar cells with pristine P3HT as HTM (Table S1 online). The underlying mechanism for excellent performance has been studied by investigating the cell parameters including the thickness of ALD TiO_2 layer, morphology of perovskite, transmittance, leakage current and charge transfer and recombination processes.

2 Materials and methods

2.1 Preparation of mesoporous templates

The FTO glasses were etched by Zn powder and dilute hydrochloric acid (HCl, 36.5 %–38.0 %, Alfa) and the

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