

# All-solid solar cells with hybrid perovskite absorbers and graphene electron transport layers



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## ABSTRACT

This work reports the perovskite/titanium dioxide (TiO<sub>2</sub>) heterojunction solid state solar cells (SSSCs) with a hole transport material (HTM) and graphene electron transport layer. The effects of a nanostructure CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite thin film, the HTM, and graphene electron transport layer in SSSC structure were examined. The SSSCs prepared with the optimal parameter exhibited a short-circuit current density (J<sub>sc</sub>), open-circuit voltage (V<sub>oc</sub>), and power conversion efficiency (η) of 17.89 mA/cm<sup>2</sup>, 0.89 V, and 6.91%, respectively. Obvious improvements in power conversion efficiency of the SSSCs were observed by using the HTM and graphene electron transport layer. The HTM and graphene thin films provide a great hole and electron transfer channel for the photogenerated carriers to external circuit, respectively.

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## 1. Introduction

Dye sensitized solar cells (DSSCs) based on titanium dioxide (TiO<sub>2</sub>) have been under extensive research in recent years. [1–5] DSSCs consist of a dye at the interface between a mesoporous TiO<sub>2</sub> wide-bandgap semiconductor and a liquid electrolyte. DSSC with ruthenium complex sensitizer has been demonstrated highest power conversion efficiency. However, the cost of the ruthenium complex sensitizer is very expensive. On the other hand, the liquid electrolyte, which the most commonly used redox electrolyte in DSSC structure is iodide/tri-iodide (I<sup>-</sup>/I<sup>3-</sup>), is highly corrosive, volatile, and photoreactive, interacting with metallic electrodes and sealing materials, such that it affects the reliability of the DSSC. [6–9] Therefore, a promising alternative is to develop a new solid-state absorber material.

Graphene is a potential material for many applications because of their high electron mobility, outstanding optical properties, and thermo conductivity, and mechanical stability [10–13]. Several articles have been reported DSSCs used graphene as electron transport layer prepared by Hummers method, chemical vapor deposition, powder blend, and sputtering [5,14–17].

Therefore, in this article, characteristics of the perovskite CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> (MAPbI<sub>3</sub>)/titanium dioxide (TiO<sub>2</sub>) bulk heterojunction solid state solar cells (SSSCs) with graphene electron transport layer were studied. Besides, we report the fabrication and performance of the DSSCs with the graphene electron transport layer.

Finally, we will discuss the electron transport mechanism in SSSCs with the graphene electron transport layer.

## 2. Experimental

Fluorine-doped tin oxide (FTO) glass slides were cleaned with acetone, ethanol and deionized water. Subsequently, 60 nm-thick graphene as the electron transport layer was sputtered onto the FTO glass substrates sonicated with ethanol for 5 min, by the radio frequency magnetron sputtering system. The deposition rate of the sputtered graphene is 60 nm/min. The details of this process and the characteristics of the graphene have been reported elsewhere. [2,17] TiO<sub>2</sub> slurry was prepared by mixing 6 g nanocrystalline powder (Degussa, P25 titanium oxide, Japan), 0.1 mL Triton X-100, and 0.2 mL acetylacetone. TiO<sub>2</sub> photoelectrode was spin-coated onto the FTO/graphene substrates at a speed of 6000 rpm for 60 s. The resulting films were annealed at 550 °C for 30 min. After that, perovskite (CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> or MAPbI<sub>3</sub>) precursor solution was coated onto the TiO<sub>2</sub>/graphene/FTO substrates by spinner at a speed of 2500 rpm for 30 s. The precursor solution was prepared by 1.2 M PbI<sub>2</sub> and 1.2 M methylammonium iodide (MAI) in a cosolvent of dimethyl sulfoxide (DMSO) and  $\gamma$ -butyrolactone (GBL) (vol. ratio = 1:1). Then, the substrate was annealed at 70 °C for 30 min. After the following, substrates were cooled down to room temperature, and coating 2,2'-7,7'-tetrakis-(N,N-di-p-methoxyphenylamine)-9,9'-spirobifluorene (spiroMeOTAD) as hole transport material (HTM) onto the perovskite/TiO<sub>2</sub>/graphene/FTO substrates. For a deposition of HTM, a solution consisting of 40 mg spiroMeOTAD in 500  $\mu$ l of chlorobenzene was prepared. This solution

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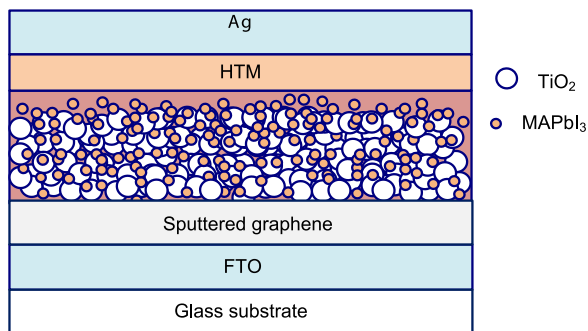


Fig. 1. Schematic diagram of the cross section of the solid state solar cells.

was mixed with 14.4  $\mu\text{l}$  TBP (4-tert-butylpyridine) and 8.75  $\mu\text{l}$  Li-bis(trifluoromethanesulfonyl)imide(LiTFSI)/acetonitrile (520 mg/mL). The spin coating process was performed at 1500 rpm for 60 s. The spin-coated samples were dried in vacuum for 3 h. Finally, an Ag anode ( $\sim 220$  nm) was deposited onto the top of the HTM of spiroMeOTAD through a shadow mask to give a device area of  $0.4 \times 0.4$   $\text{cm}^2$  by evaporator. Fig. 1 schematically depicts the complete structure. The photocurrent voltage ( $I$ - $V$ ) characteristics were measured using a Keithley 2420 programmable source meter under  $100$   $\text{mW}/\text{cm}^2$  irradiation. The forward scan parameters are scan speed ( $0.2$  V/s) and delay time ( $40$  ms). Simulated sunlight was provided by a  $500$  W Xenon lamp (Hong Ming Technology Co, Ltd, Taiwan) that had been fitted with an AM-1.5 filter. Also, in this study, we prepared a DSSC for comparison. The assembly procedure of the DSSC as following: a  $\text{TiO}_2$  film was coated on the ITO glass substrate spin coating. Thereafter, annealing was performed by rapidly thermal annealing at  $450$   $^\circ\text{C}$  for  $30$  min. When the sample had been cooled to room temperature, it was soaked in the N719 dye absorption. Then, a platinum layer was sputtered onto the ITO substrate as a counter electrode. Finally, the electrolyte was then injected into the space between the two electrodes. The details of the DSSC have been reported elsewhere [17].

### 3. Results and discussion

Fig. 2 presents the cross-sectional FESEM image of the HTM/ $\text{TiO}_2$  + perovskite/graphene films on FTO glass substrate. It clearly indicates the presence of each layer of the graphene ( $60$  nm), blend layer of  $\text{TiO}_2$  and perovskite  $\text{CH}_3\text{NH}_3\text{PbI}_3$  ( $600$  nm), perovskite  $\text{CH}_3\text{NH}_3\text{PbI}_3$  layer ( $200$  nm), and HTM layer ( $40$  nm). As

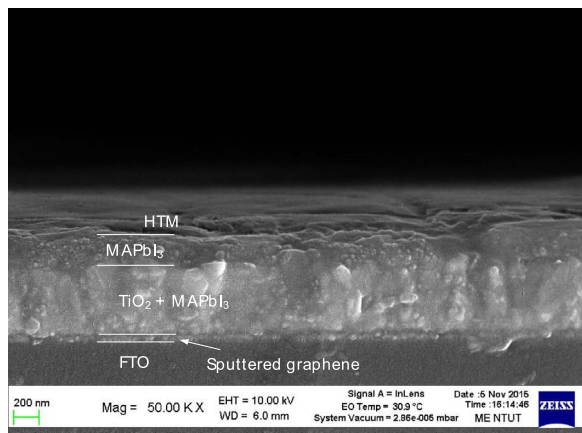


Fig. 2. Cross-sectional FESEM image of the HTM/ $\text{TiO}_2$  + perovskite/graphene films on FTO glass substrate.

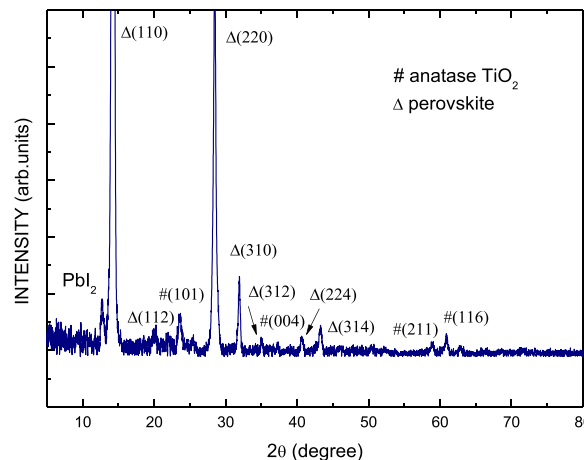


Fig. 3. XRD patterns of the blend of perovskite  $\text{CH}_3\text{NH}_3\text{PbI}_3$  and  $\text{TiO}_2$  nanocrystal films deposited on FTO substrates.

shown in Fig. 2, the perovskite  $\text{CH}_3\text{NH}_3\text{PbI}_3$  penetrated into the  $\text{TiO}_2$  layer to form the blend layer.

Fig. 3 shows the XRD pattern of the blend of perovskite  $\text{CH}_3\text{NH}_3\text{PbI}_3$  and  $\text{TiO}_2$  nanocrystal films deposited on FTO substrates. The spectra in this study reveal three peaks at the position of  $14.23^\circ$ ,  $28.34^\circ$ , and  $31.67^\circ$ , which correlate well with (110), (220), and (310) planes of the  $\text{CH}_3\text{NH}_3\text{PbI}_3$  perovskite phase. Besides, several weak peaks about  $\text{TiO}_2$  phase were observed in the XRD pattern. They are located at  $23.67^\circ$ ,  $37.34^\circ$ ,  $58.86^\circ$ , and  $60.88^\circ$  corresponding to anatase  $\text{TiO}_2$  (101), (004), (211), and (116) phase, respectively. This result suggests that the blend of  $\text{TiO}_2$  nanocrystal power and perovskite  $\text{CH}_3\text{NH}_3\text{PbI}_3$  absorber is homogenous, and suitable for as an active layer of SSSCs. Also,  $\text{PbI}_2$  phases can be observed in the  $\text{CH}_3\text{NH}_3\text{PbI}_3$  perovskite layers. This is due to the thermal decomposition of  $\text{CH}_3\text{NH}_3\text{PbI}_3$ .

Fig. 4 displays the UV–visible absorption spectra of DSSC with dye N719 and SSSC with  $\text{TiO}_2$ /perovskite absorber bulk heterojunction. To compare these two spectra, there are two absorption edges, labeled A and B, located  $400$  nm and  $550$  nm corresponding to the absorption of  $\text{TiO}_2$  and dye N719, respectively. Similarly, there are two absorption edges, labeled A and C, located  $400$  nm and  $760$  nm corresponding to the absorption of  $\text{TiO}_2$  and  $\text{CH}_3\text{NH}_3\text{PbI}_3$  perovskite absorber, respectively. The absorption of wavelengths in the range  $400$ – $760$  nm is attributed to the perovskite  $(\text{CH}_3\text{NH}_3)\text{PbI}_3$  particles, which have a band gap  $1.6$  eV. [18] Clearly, the solar cell with the  $\text{CH}_3\text{NH}_3\text{PbI}_3$  perovskite absorber exhibited the greatest increase in absorption ranging from  $400$  nm

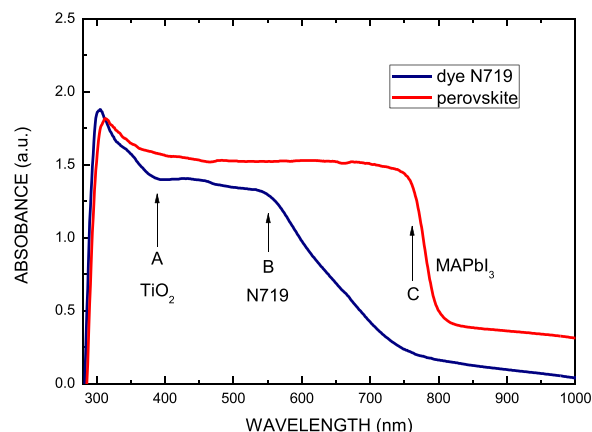


Fig. 4. UV–visible absorption spectra of (a) DSSCs with dye N719, and (b) solid-state solar cell with perovskite active layer.

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