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## Hybrid solar cells composed of perovskite and polymer photovoltaic structures

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### ABSTRACT

Organic/inorganic lead halide perovskite solar cells have recently attracted much attention in photovoltaic research, due to the devices show promising ways to achieve high efficiencies. The perovskite devices with high efficiencies, however, are typically fabricated in tandem solar cell which is complicated. In this research work, we introduce a solar cell device with the combination of  $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$  perovskite and bulk heterojunction PCDTBT:PC<sub>70</sub>BM polymer without any tandem structure. The new integrated perovskite/polymer hybrid structure of ITO/PEDOT:PSS/perovskite/PCDTBT:PC<sub>70</sub>BM/PC<sub>70</sub>BM/TiO<sub>x</sub>/Al provides higher power conversion efficiency (PCE) of devices compared with conventional perovskite cell structure. With the optimized PCDTBT:PC<sub>70</sub>BM thickness of ~70 nm, the highest PCE of 11.67% is achieved. Variation of conducting donor polymers in this new structure is also preliminary demonstrated. This study provides an attractively innovative structure and a promising design for further development of the new-generation solar cells.

### 1. Introduction

Solar energy is one choice of interesting renewable energies, because it has lower environmental impact compared with fossil fuels [1]. Solar cells or photovoltaic devices can directly convert solar radiation into electricity. Currently, the commercially accessible crystalline silicon (Si) solar cells have shown PCE up to 25%, but these cells are still produced by complex methods at quite high temperatures [2]. Therefore, many researchers have tried to find new photovoltaic materials to produce alternative solar cells, which expose high performance with large scale production and low cost [3]. Organic-inorganic hybrid material, named perovskite, as a new light absorber has been attracted considerable interest, because of their great properties for the development of low-cost thin-film solar cells [4]. The perovskite material performs high absorption coefficients, suitable direct band gaps, small

exciton binding energies, high carrier mobilities, long diffusion lengths, and superior defect tolerances [1–5]. The structure of perovskite material is  $\text{ABX}_3$ , which A is cation (metal or hydrocarbon), B is a metal cation, and X is halide atoms. Examples of perovskite are organometallic halide perovskites, e.g.  $\text{CH}_3\text{NH}_3\text{PbI}_3$  and  $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$  [5].

Nowadays, devices with combination of two or more completed photovoltaic layers in one cell, called tandem cells, have been expected to get high efficiency with the cells connected in series, which absorb light in different wavelength ranges. Giles et al. [6] exhibited an infrared absorbing wider bandgap  $\text{FA}_{0.83}\text{Cs}_{0.17}\text{Pb}(\text{I}_{0.5}\text{Br}_{0.5})_3$  material in two and four-terminal perovskite/perovskite tandem solar cells, which achieved in 17.0% and 20.3%, respectively. This infrared absorbing perovskite material also showed excellent thermal and atmospheric stability. Reza et al. [7] demonstrated a high PCE approaching ~25% from bifacial Si heterojunction/perovskite tandem solar cell at an

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optimum thickness of perovskite of 135 nm. Philipp et al. [8] introduced a four-terminal tandem cell based on a perovskite top cell and a c-Si heterojunction bottom cell with a transparent MoO<sub>x</sub>/ITO hole contact. The top cell transmitted ~55% near-infrared spectral region. This architecture reached PCE of 13.4% (top cell 6.2%, bottom cell 7.2%). Moreover, perovskite/polymer tandem solar cells have been developed to achieve high performance. Yao et al. [9] showed the perovskite/polymer tandem solar cell with PCE of 16.0% from ~90 nm perovskite thickness, which assisted in use of less lead content in the device. Chun-Chao et al. [10] introduced a new IR-sensitive block copolymer, PBSeDTEG8, which this polymer:PCBM blend (as the bottom subcell) enhanced thermal stability in the tandem device. This perovskite/polymer monolithic hybrid tandem solar cell achieved PCE of 9.13%, which is more than the perovskite and polymer single-junction devices. Jian et al. [11] presented perovskite-organic hybrid tandem solar cell in two-terminal tandem cell with PBDTT-DPP:PC<sub>70</sub>BM blending polymer and found that the hybrid tandem efficiency reached 8.62%, which greater than both perovskite and PBDTT-DPP:[6,6]-phenyl-C<sub>71</sub>-butyric acid methyl ester (PC<sub>70</sub>BM) single-junction cells. Unfortunately, perovskite/polymer tandem solar cell has some disadvantages, i.e. its interlayers make losses in photon transmittance for the bottom sub-cell and tandem device fabrication is complicated [12]. Additionally, the difficulty in fabricating perovskite-polymer hybrid tandem cell is to optimize the interconnecting layer [11].

Recently, integration of solar cell combining both perovskite and bulk-heterojunction (BHJ) photovoltaic layers into a simple photovoltaic device has been interested to achieve high efficiency with simple fabrication techniques. Chuantian and Liming [13] introduced PDPP3T:PCBM blending polymer into perovskite solar cells with a structure of ITO/PEDOT:PSS/CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>/(PDPP3T-PC<sub>61</sub>BM)/Ca/Al. They found that the photoresponse can absorb closely to the near-infrared (NIR), but the efficiency of integrated perovskite/blending polymer device cannot be enhanced. Long et al. [14] employed PDPP3T:PCBM layer into perovskite solar cells and found that the perovskite/polymer hybrid solar cell efficiency can be improved with a 1:4 weight ratio of PDPP3T:PCBM.

In organic (polymer) solar cells, the cells made of poly(N-9'-hepta-decanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)) (PCDTBT) (Fig. 1a) as an electron donor material have been widely employed. This was because of deep highest occupied molecular orbital (HOMO) energy level (~ -5.5 eV) of PCDTBT, which was below the air oxidation threshold (-5.27 eV). Consequently, the system could be air-stable and low value HOMO assures a high open-circuit voltage (V<sub>oc</sub>). Furthermore, the position of lowest unoccupied molecular orbital (LUMO) energy level of PCDTBT is above PC<sub>70</sub>BM (electron acceptor material) (Fig. 1b) at least 0.2–0.3 eV, which electron from the donor could effectively transfer to the acceptor [15,16]. The cells composed of PCDTBT:PCBM have shown higher PCE (> 7%) than PDPP3T:PCBM with internal quantum efficiencies approaching 100% [17]. Therefore, PCDTBT is considered to be a promising polymer for improvement of efficiency of perovskite/polymer hybrid devices.

In this study, we demonstrated that CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3-x</sub>Cl<sub>x</sub> perovskite light absorber material can be processed by a solution approach. Conventional perovskite cells (reference cells) with PC<sub>70</sub>BM as a electron transporting layer were fabricated (device 1, Fig. 1b). Integration of PCDTBT:PC<sub>70</sub>BM bulk heterojunction into conventional perovskite solar cell was carried out by using 2 different approaches: (1) replacing PC<sub>70</sub>BM layer by PCDTBT:PC<sub>70</sub>BM layer (so called “perovskite/polymer hybrid solar cell”) (devices 2, Fig. 1c) and (2) adding PCDTBT:PC<sub>70</sub>BM layer between perovskite and PC<sub>70</sub>BM layers (so called “integrated perovskite/polymer hybrid solar cell”) (device 3, Fig. 1d). Energy level diagram of integrated perovskite/polymer hybrid solar cell is shown in Fig. 2.

## 2. Experimental

### 2.1. Materials

PEDOT:PSS aqueous dispersion (Clevios PH1000) was purchased from Heraeus (Germany). Perovskite precursor ink I101 (CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3-x</sub>Cl<sub>x</sub>, for Air Processing), PCDTBT, PC<sub>70</sub>BM and PTB7 were purchased from Ossila (UK). P3HT was purchased from Rieke Metals (USA). Solvent (1,2-dichlorobenzene, DCB and 1,8-Diiodooctane, DIO) was obtained from Sigma-Aldrich (USA). TiO<sub>x</sub> was synthesized by sol-gel method. Isopropyl alcohol (IPA, Fisher scientific) was used to clean substrates. Indium doped tin oxide (ITO) patterned glasses (5 Ω, Luminescence Technology Corporation) and high purity Al are commercially available products.

### 2.2. Substrate preparation

ITO-coat glass substrates were cleaned by sequential ultrasonic treatment in a detergent, deionized water, and isopropyl alcohol (IPA) for 20 min and dried by nitrogen flow. Then the pre-cleaned glass substrates were treated by UV treatment for 30 min.

### 2.3. Sample preparation

Perovskite precursor was stirred at 70 °C for at least 2 h to allow for complete dissolution of solute. PCDTBT:PC<sub>70</sub>BM and P3HT:PC<sub>70</sub>BM solutions (40 mg/ml) were mixed in 1:0.8 weight ratio and dissolved in DCB. A mixed PTB7:PC<sub>70</sub>BM (40 mg/ml) was prepared in 1:1.5 weight ratio and dissolved in DCB and DIO solvent mixture. All conjugated polymers were stirred at 70 °C for 1 h. TiO<sub>x</sub> sol-gel solution was diluted with IPA in a ratio of 1:8.

### 2.4. Device fabrication

PEDOT:PSS solution (20 μl) was coated on an ITO-substrate by convective deposition method at a deposition speed of 3000 μm/s with 2-time coating in air and annealed at 120 °C for 30 min. Then, the PEDOT:PSS film was transferred to a glove box (N<sub>2</sub>) to heat the substrate on a hot plate at 100 °C for 15 min. Perovskite precursor ink (50 μl) was spin coated at 2,200 rpm for 30 s and annealed at 100 °C for 90 min. Then, PC<sub>70</sub>BM was spin coated 1,500 rpm for 30 s. TiO<sub>x</sub> was coated on the top of PC<sub>70</sub>BM by spin-coating method at 2500 rpm for 30 s and then the film was annealed at 80 °C for 20 min. Finally, aluminum (Al) cathode (~120 nm) was evaporated under high vacuum (5 × 10<sup>-6</sup> Torr) to complete the device by a thermal evaporator.

For perovskite/polymer hybrid solar cells, after coated perovskite layer, PCDTBT:PC<sub>70</sub>BM, solution (35 μl) was additionally spin coated at 2500 rpm for 30 s (depending on the desired thickness of PCDTBT:PC<sub>70</sub>BM, spin rates of 1500, 2000, 2500, 3000 and 3500 rpm for 30 s were applied) on top of perovskite film. For variation of conducting donor polymer:PC<sub>70</sub>BM, P3HT:PC<sub>70</sub>BM and PTB7:PC<sub>70</sub>BM were additionally spin coated at 2500 rpm for 30 s on top of perovskite layer.

### 2.5. Sample characterizations

Power conversion efficiency and photocurrent density-voltage (*J-V*) curves were measured under ambient atmosphere and simulated solar light, AM 1.5, 100 mW/cm<sup>2</sup>, using a solar simulator with a xenon lamp. The light intensity of illumination source was calibrated using a standard 2 cm × 2 cm silicon reference cell. The active area of the device was defined with a metal aperture mask of approximately 0.2 × 0.5 cm<sup>2</sup>. Plots of the incident photon-to-current conversion efficiency (IPCE) as a function of excitation wavelength (λ = 300–1100 nm) were determined by using the QEPVSI-b measurement system under the irradiation of a 300 W Xenon lamp equipped with a monochromator controlled via USB connection through TracQ

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