



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:PC₇₀BM polymer without any tandem structure. The new integrated perovskite/polymer hybrid structure of ITO/PEDOT:PSS/perovskite/PCDTBT:PC₇₀BM/PC₇₀BM/TiO_x/Al provides higher power conversion efficiency (PCE) of devices compared with conventional perovskite cell structure. With the optimized PCDTBT:PC₇₀BM 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 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_x/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:PC₇₀BM blending polymer and found that the hybrid tandem efficiency reached 8.62%, which greater than both perovskite and PBDTT-DPP:[6,6]-phenyl-C₇₁-butyric acid methyl ester (PC₇₀BM) 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/CH₃NH₃PbI₃/(PDPP3T-PC₆₁BM)/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_{oc}). Furthermore, the position of lowest unoccupied molecular orbital (LUMO) energy level of PCDTBT is above PC₇₀BM (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 CH₃NH₃PbI_{3-x}Cl_x perovskite light absorber material can be processed by a solution approach. Conventional perovskite cells (reference cells) with PC₇₀BM as a electron transporting layer were fabricated (device 1, Fig. 1b). Integration of PCDTBT:PC₇₀BM bulk heterojunction into conventional perovskite solar cell was carried out by using 2 different approaches: (1) replacing PC₇₀BM layer by PCDTBT:PC₇₀BM layer (so called “perovskite/polymer hybrid solar cell”) (devices 2, Fig. 1c) and (2) adding PCDTBT:PC₇₀BM layer between perovskite and PC₇₀BM 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 (CH₃NH₃PbI_{3-x}Cl_x, for Air Processing), PCDTBT, PC₇₀BM 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_x 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:PC₇₀BM and P3HT:PC₇₀BM solutions (40 mg/ml) were mixed in 1:0.8 weight ratio and dissolved in DCB. A mixed PTB7:PC₇₀BM (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_x 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 (N₂) 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, PC₇₀BM was spin coated 1,500 rpm for 30 s. TiO_x was coated on the top of PC₇₀BM 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 × 10⁻⁶ Torr) to complete the device by a thermal evaporator.

For perovskite/polymer hybrid solar cells, after coated perovskite layer, PCDTBT:PC₇₀BM, solution (35 μl) was additionally spin coated at 2500 rpm for 30 s (depending on the desired thickness of PCDTBT:PC₇₀BM, 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:PC₇₀BM, P3HT:PC₇₀BM and PTB7:PC₇₀BM 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/cm², 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 cm². 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

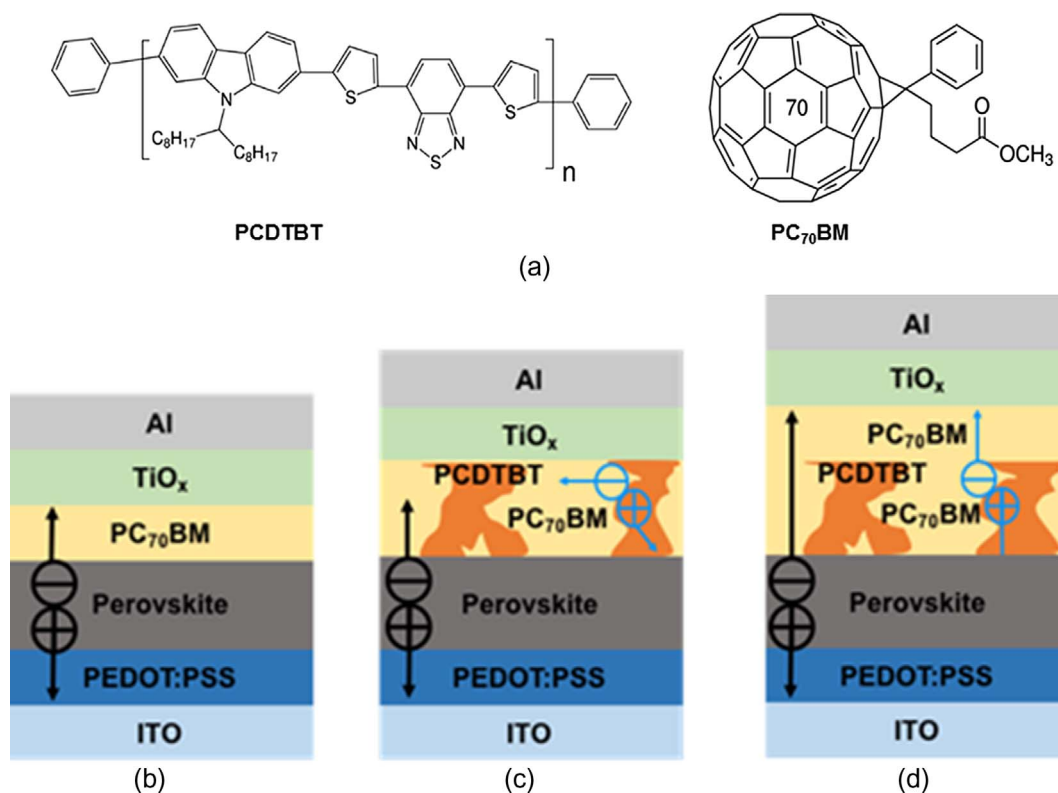


Fig. 1. (a) Structures of PCDTBT and PC₇₀BM; (b) structures for perovskite solar cell; (c) perovskite/polymer hybrid solar cell and (d) integrated perovskite/polymer hybrid solar cell.

Basic software. The thickness of thin film was measured using a spectroscopic reflectometer (AvaLight-Dhc Full-range Compact Light Source, Avenes Enlightening Spectroscopy). The coating thicknesses were determined by a white light interference and the reflectance spectra were captured and analyzed by software algorithms. The process of sampling measurement initially conducted the measurement of an uncoated substrate (reference) and then followed by the coated substrate using the same measurement conditions. Therefore, the measured data was translated and analyzed into the optical thickness by the mathematical calculation software.

3. Results and discussion

Fig. 3a and Table 1 show photovoltaic characteristics of perovskite solar cell, perovskite/polymer hybrid solar cell, and integrated perovskite/polymer hybrid solar cell. PCE of the perovskite solar cell

(device 1, thickness ~ 360 nm) was 8.18%. This PCE is relatively lower compared with other reports [13,14], due to quite high relative humidity in Thailand with one-step deposition technique in this work. Increase in thickness of perovskite layer could not lead to a higher PCE. The results show that when PCDTBT:PC₇₀BM layer replaced PC₇₀BM layer (device 2), PCE was dropped from 8.18 to 7.15%, comparing with the conventional perovskite solar cell (device 1). Device 2 consisted of PCDTBT:PC₇₀BM bulk-heterojunction layers, when excitons (electron-hole pair) were created, they might be incompletely dissociated at a donor-acceptor interface, which led to charge recombination. In previous reports, the surface roughness at interface of perovskite/polymer was directly linked to both charge extraction and recombination [18,19]. Moreover, the surface contact between perovskite and electron transporting layers (PC₇₀BM) was reduced and PC₇₀BM layer was changed to channels in bulk-heterojunction structure. As the results, electron and hole cannot directly transfer to charge collectors. On the

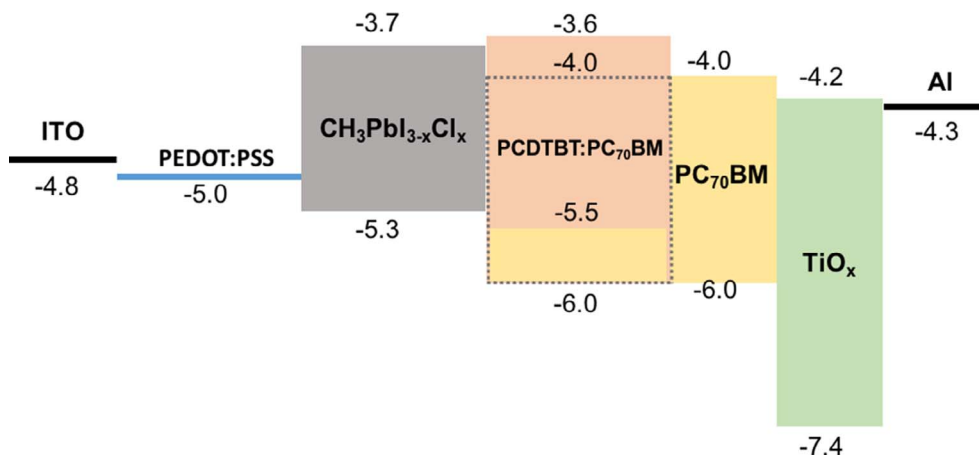


Fig. 2. Energy level diagram of integrated perovskite/polymer hybrid solar cell.

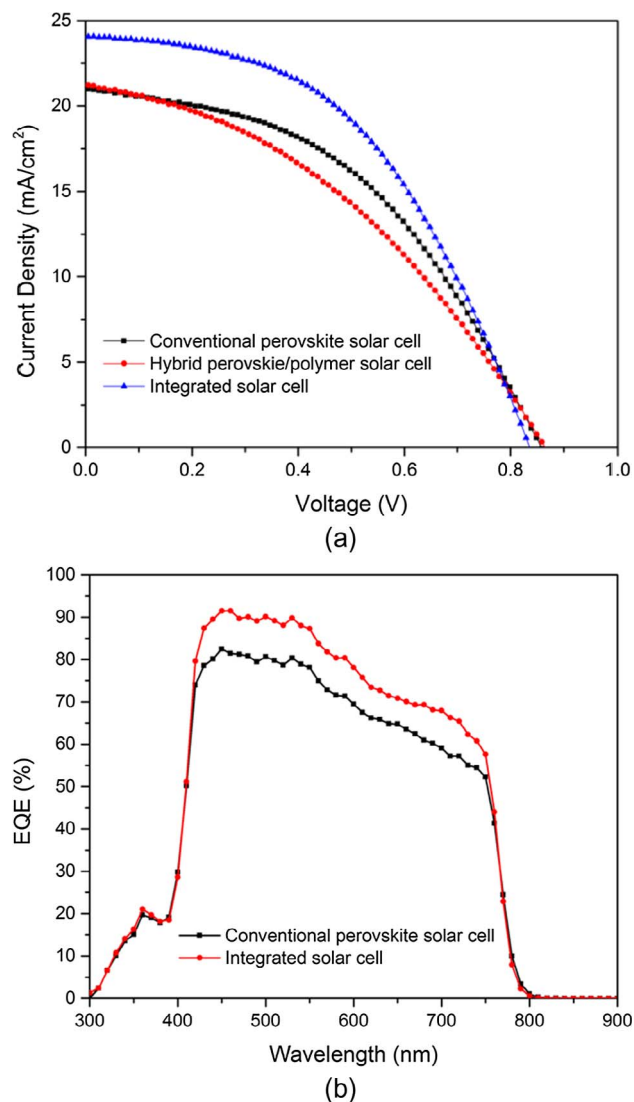


Fig. 3. (a) Photocurrent density-voltage (J - V) curves of different structures of planar perovskite solar cells, perovskite/polymer hybrid solar cell, and integrated perovskite/polymer hybrid solar cell, under AM 1.5, 100 mW/cm²; and (b) EQE spectra of conventional perovskite solar cell and integrated perovskite/polymer hybrid solar cell.

Table 1

Various perovskite solar cell device structures with their performances and photovoltaic characteristics (J_{sc} , V_{oc} , FF , and PCE) of the cells.

| Device | Structure | J_{sc} (mA/ cm ²) | V_{oc} (V) | FF | PCE (%) |
|--------|---|---------------------------------------|--------------|------|-----------|
| 1 | ITO/PEDOT:PSS/Perovskite/ PC ₇₀ BM/TiO _x /Al | 20.99 | 0.85 | 0.48 | 8.18 |
| 2 | ITO/PEDOT:PSS/Perovskite/ PCDTBT:PC ₇₀ BM/TiO _x /Al | 21.24 | 0.86 | 0.38 | 7.15 |
| 3 | ITO/PEDOT:PSS/Perovskite/ PCDTBT:PC ₇₀ BM (spin-coating speed 1500 rpm)/PC ₇₀ BM/TiO _x /Al | 24.08 | 0.83 | 0.48 | 9.63 |

contrary, PCDTBT:PC₇₀BM heterojunction layer was added to the conventional perovskite structure between perovskite and PC₇₀BM layers (device 3). Device 3 showed a short-circuit current density (J_{sc}) of 24.08 mA cm⁻², an open-circuit voltage (V_{oc}) of 0.83 V, a fill factor (FF) of 0.48, and PCE of 9.63% (higher than that of conventional perovskite solar cell (device 1)). This was due to the fact that the device composed of 2 active layers (perovskite and PCDTBT:PC₇₀BM), which the excitons

could be created in both two active layers (adding interfacial surfaces for light absorption) and PC₇₀BM could assist in accepting and conducting of the electrons as an electron transporting layer. Hence, PCDTBT:PC₇₀BM heterojunction acted as a selective gateway in perovskite cell (electron and hole have their own channels in transports). Additionally, this configuration device could generate great charge mobility, which can be seen from the energy level diagram (Fig. 2).

Fig. 3b shows EQE spectra of two structures of devices, i.e. conventional perovskite and integrated solar cells. They responded to light in the same wavelength regions (300–750 nm). EQE of integrated solar cell was higher than that of conventional perovskite device. It is ensured that the PCDTBT:PC₇₀BM layer could contribute to the greater absorption of light and acts as an additional active layer, which it can work effectively without any interlayer. The results obtained from J - V measurements, confirm a higher J_{sc} of the integrated device compared with that of the conventional perovskite device (J_{sc} of conventional perovskite device and integrated device are 20.99 and 24.08 mA cm⁻², respectively), integrations of EQE curves were carried out. Obtained J_{sc} from the curve integrations of the integrated perovskite/polymer hybrid device was found to be higher than that of the conventional cell (i.e. 18.02 and 16.22 mA cm⁻², respectively).

Optimized thickness of bulk heterojunction layer between perovskite and electron transporting layers was further examined (Fig. 4 and Table 2). PCDTBT:PC₇₀BM with different thicknesses could be fabricated by control of spin coating speeds of the PCDTBT:PC₇₀BM solutions (Fig. 5). A perovskite solar cell without PCDTBT:PC₇₀BM (device 1) achieved a minimum PCE of 8.18%. At high PCDTBT:PC₇₀BM film thickness (95.8 nm, device 2), higher PCE (9.63%) could be obtained as described above. PCE was found to increase with decreasing the film thickness to a certain value, because of reduction of charge-recombination in this charge selective gateway. Meanwhile, the deposition speed of 2500 rpm with film thickness around 71.6 nm (device 4) showed the best PCE of 11.67% with outstanding J_{sc} (25.23 mA/cm²), V_{oc} (0.89 V), and FF (0.51). To check the reliability of these results, eight referent cells (device 1) and eight cells with the same conditions with the best PCE cells (device 4) were repeatedly fabricated and characterized. Average PCE s of the referent cells compared with the best PCE cells were $7.97 \pm 0.19\%$ and $11.28 \pm 0.43\%$, respectively. The thickness of 71.6 nm (device 4) was appropriate for light absorption and not too thick, so electrons could transfer to the acceptor layer (PC₇₀BM) at an interface and further transferred to the cathode for charge collection. In other sides, holes could move throughout the polymer passed through the main active layer (perovskite) and were

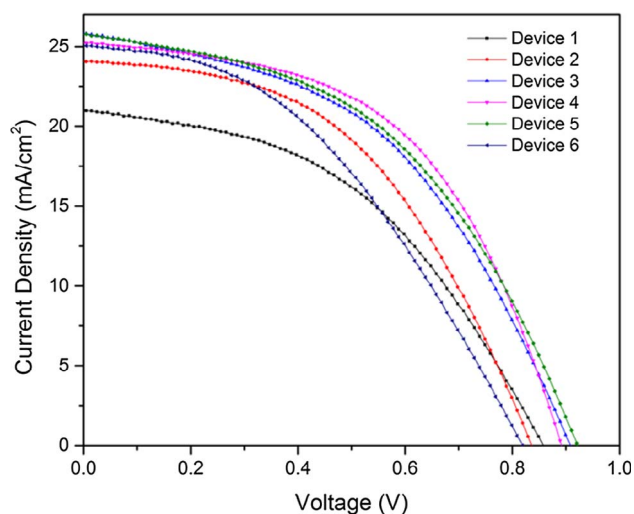
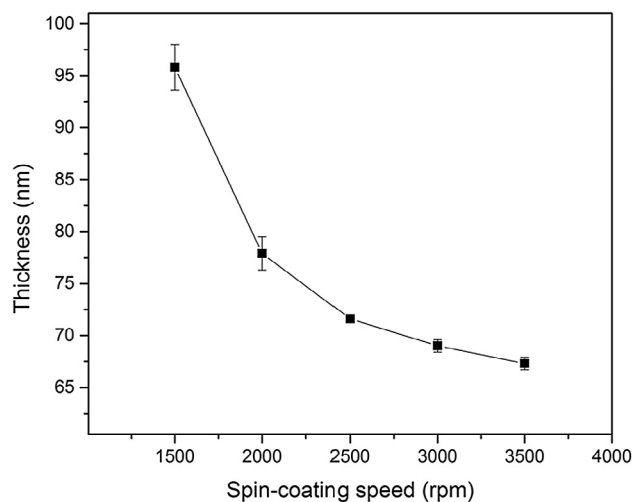


Fig. 4. Photocurrent density-voltage (J - V) curves of devices with different thicknesses of PCDTBT:PC₇₀BM (device configuration: ITO/PEDOT:PSS/perovskites/PCDTBT:PC₇₀BM/PC₇₀BM/TiO_x/Al).

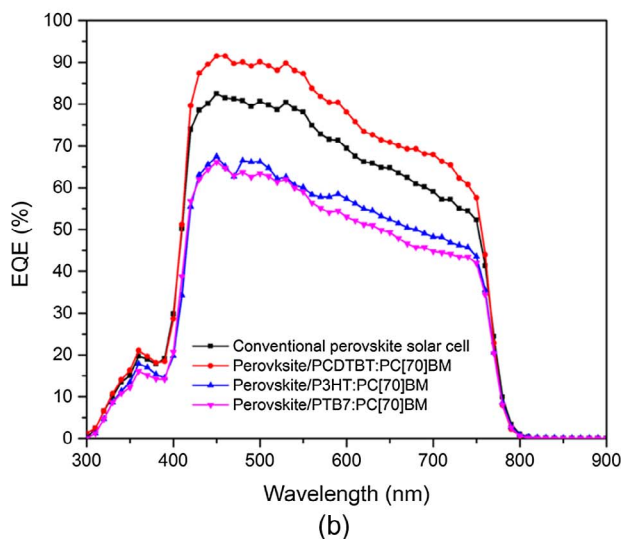
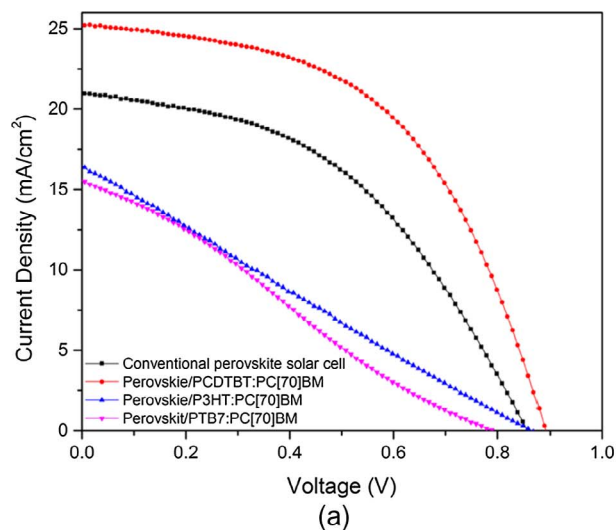
Table 2Performances and photovoltaic characteristics of various perovskite devices with different thicknesses of integrated PCDTBT:PC₇₀BM.

| Device | Active layer | Spin-coating speed of PCDTBT:PC ₇₀ BM (rpm) | Thickness of PCDTBT:PC ₇₀ BM layer (nm) | J_{sc} (mA/cm ²) | V_{oc} (V) | FF | PCE (%) |
|--------|---------------------------------------|--|--|--------------------------------|--------------|------|---------|
| 1 | Perovskite/PC ₇₀ BM | – | – | 20.99 | 0.85 | 0.48 | 8.18 |
| 2 | Perovskite/PCDTBT:PC ₇₀ BM | 1500 | 95.8 ± 2.2 | 24.08 | 0.83 | 0.48 | 9.63 |
| 3 | Perovskite/PCDTBT:PC ₇₀ BM | 2000 | 77.9 ± 1.6 | 25.84 | 0.90 | 0.46 | 10.87 |
| 4 | Perovskite/PCDTBT:PC ₇₀ BM | 2500 | 71.6 ± 0.4 | 25.23 | 0.89 | 0.51 | 11.67 |
| 5 | Perovskite/PCDTBT:PC ₇₀ BM | 3000 | 69.0 ± 0.6 | 25.75 | 0.92 | 0.46 | 11.13 |
| 6 | Perovskite/PCDTBT:PC ₇₀ BM | 3500 | 67.3 ± 0.6 | 25.07 | 0.81 | 0.41 | 8.57 |

**Fig. 5.** Thicknesses of PCDTBT:PC₇₀BM active layers at various speeds of spin coating (1500–3500 rpm).

collected at the anode. However, when the film thickness of PCDTBT:PC₇₀BM further decreased to 69.0 and 67.3 nm (devices 5 and 6), PCE was found to decrease, because of less additional active layer (charge selective gateway). This result complied with the previous study of thickness of PCDTBT:PC₇₀BM in organic solar cells [16,20].

In previous reports, various bulk-heterojunction materials as active layers in conventional organic solar cells have been investigated and achieved effective performances. These cells were made from various conducting polymers, such as poly(3-hexylthiophene-2,5-diyl) (P3HT), poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophenediyl]] (PTB7), etc. [21–24]. Therefore, we further used other conducting donor polymers (P3HT and PTB7) to fabricate polymer:PC₇₀BM layer and insert between perovskite and PC₇₀BM layers to fabricate integrated perovskite solar cells in the configuration of ITO/PEDOT:PSS/perovskite/conducting donor polymer:PC₇₀BM/TiO_x/Al. Photocurrent density-voltage (J - V) curves and photovoltaic results are shown in Fig. 6a and Table 3. It was found that the performances of cells made of alternative conducting donor polymers (P3HT and PTB7) were significantly lower (PCE of 3.48 and 3.15%, respectively), compared with conventional perovskite device and integrated perovskite/PCDTBT:PC₇₀BM hybrid device. FF of both cells decreased (0.22 and 0.25 for cells made of P3HT and PTB7, respectively), because of incompatibility between layers contacting in these devices. Moreover, their J_{sc} dropped obviously, which showed only 16.45 and 15.52% mA cm⁻² for cells made of P3HT and PTB7, respectively). The main reason for the decreased efficiencies might be non-optimization of bulk-heterojunction layer for these P3HT and PTB7. EQE spectra (Fig. 6b) also confirms that both perovskite/P3HT:PC₇₀BM and perovskite/PTB7:PC₇₀BM devices responded to light in the similar range of wavelengths, but at lower values (max EQE ~65%) compared with perovskite/PCDTBT:PC₇₀BM (max EQE ~90%). Moreover, EQE spectra show the same trend with performance data from J - V curves. In future

**Fig. 6.** Photocurrent density-voltage (J - V) curves of (a) conventional perovskite and integrated perovskite hybrid devices with different conducting donor polymers in polymer:PC₇₀BM bulk-heterojunction layers, under AM 1.5, 100 mW/cm²; and (b) EQE spectra of conventional perovskite and various integrated perovskite hybrid devices with different conducting donors in polymer:PC₇₀BM bulk-heterojunction layers.

works, the new conducting donor polymers and optimal conditions for more-efficient integrated perovskite/polymer hybrid solar cells should be studied to solve the interfacial problems.

4. Conclusions

We developed efficient photovoltaic devices based on perovskite cell by combining perovskite material with PCDTBT:PC₇₀BM heterojunction from organic solar cells to make integrated perovskite/polymer hybrid solar cells (ITO/PEDOT:PSS/perovskite/

Table 3

Performances and photovoltaic characteristics of conventional perovskite and various integrated perovskite hybrid devices with different conducting donors in polymer:PC₇₀BM bulk-heterojunction layers.

| Device | Bulk-heterojunction layer | J_{sc} (mA/cm ²) | V_{oc} (V) | FF | PCE (%) |
|--------|--|--------------------------------|--------------|------|---------|
| 1 | Reference (no bulk-heterojunction layer) | 20.99 | 0.85 | 0.48 | 8.18 |
| 2 | PCDTBT:PC ₇₀ BM | 25.23 | 0.89 | 0.51 | 11.67 |
| 3 | P3HT:PC ₇₀ BM | 16.45 | 0.86 | 0.22 | 3.48 |
| 4 | PTB7:PC ₇₀ BM | 15.52 | 0.81 | 0.25 | 3.15 |

PCDTBT:PC₇₀BM/PC₇₀BM/TiO_x/Al). This proposed device achieved higher PCE compared with conventional perovskite cells, because PCDTBT:PC₇₀BM contributes to the greater absorption of light. The PC₇₀BM could also assist in accepting and conducting electrons as an electron transporting layer. This additional heterojunction structure acted as a selective gateway, which affected positively electron and hole collections. The highest performance of the integrated hybrid device was 3.49% higher than that of the conventional perovskite device, which used only a thin PCDTBT:PC₇₀BM layer (~70 nm). Variation of conducting donor polymers in this new structure was also preliminary demonstrated. Further systematical development could be achieved by fine-tuning the conditions for these conducting donor polymers in the integrated perovskite/polymer hybrid devices.

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