



# Cathode modification with solution-processed hybrid electron extraction layer for improved charge collection of planar heterojunction perovskite solar cells

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## ABSTRACT

Optimal interface modification of perovskite solar cells is critical to achieve efficient and balanced charge transport and collection. We herein demonstrated that a solution-processed hybrid cathode interfacial layer composed of polyethyleneimine ethoxylate and a lithium quinolate complex improves photovoltaic performance of the planar heterojunction perovskite solar cells. The hybrid cathode modifier effectively lowered work function of ITO cathode, which afforded efficient electron transport and collection at ITO. Furthermore, surface roughness of ITO was significantly decreased, leading to enlarged grain size in densely-packed perovskite thin film. Consequently, the perovskite solar cells with hybrid electron extraction layer generated maximum power conversion efficiency up 15.21%, which is 25% improved value than that without hybrid electron extraction layer. Furthermore, highly-flexible flexible devices with a hybrid electron extraction layer exhibited a promising efficiency of 14.41%, demonstrating its potential for high performance perovskite solar cells.

## 1. Introduction

Organic–inorganic lead halide perovskite semiconductors have gained intense interest as one of the most promising light absorbing materials for photovoltaic application [1–5]. The distinct advantages that include an appropriate band gap with a broad and intense light absorption, a long carrier diffusion length and low exciton binding energy afforded efficient photon harvesting and charge generation/transport. These fascinating features allow impressive advance of device performance of the perovskite solar cells (PSCs) in recent years [6]. There have been thorough investigations on crystal growth, thin film morphology and composition optimization of perovskite semiconductors for the purpose of further improvement of the photovoltaic performance of the PSCs. Meanwhile, it is also recognized that the device engineering of PSCs is another effective approach to improve device performance [7–12]. In particular, interface engineering of the PSCs, which not only reduces the energetic barrier between the perovskite absorber and the electrodes but also increases the selectivity of favorable charge carriers to the corresponding electrode, thus plays a critical role to determine the efficiency and stability of PSCs [13]. In

general, an ideal interfacial engineering for PSCs is expected to allow a suitable energy levels, high electrical conductivity and high transparency to minimize parasitic absorption loss and surface recombination at the interface of the devices. In addition, simple solution process at low temperature is another requirement for low-cost production of PSCs.

So far, the most widely used device architecture consists of *n-i-p* junction in which electron extraction layer was used as a scaffold layer for perovskite thin film. The electron extraction layers for these devices, so called “normal structure devices”, usually make use of nano-crystalline metal oxides in the form of mesoporous and compact films [14]. In spite of their outstanding optoelectronic properties for electron-selective contacts of perovskite solar cells, multiple sintering processes at high temperature impedes the practical use of metal oxide in PSCs on flexible substrate [15]. On the other hand, *n*-type organic semiconductors, such as [6,6]-phenyl-C61-butyric acid methyl ester (PCBM), have attracted much interest as an alternative to conventional metal oxides for efficient PSCs because of its simple solution processability at low temperature which is desirable for low-cost production of the devices. Moreover, it allows quick process of the device fabrication of PSCs in “normal architecture”. However, PCEs of the PCBM-

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based normal PSCs are usually inferior as compared to the devices using metal oxide electron extraction layers due to its low electron mobility [16]. In addition, the energy level mismatch between the valence band minimum of perovskite absorber and the lowest unoccupied molecular orbital of PCBM limits the maximum open circuit voltage ( $V_{OC}$ ) generation of PCBM-based devices as compared to metal oxide-based counterparts [17]. The low affinity of PCBM molecule for ITO substrate would be another important problem, because it would limit the optimal film morphology formation of the PCBM layer for efficient electron transport and extraction. Furthermore, the instability of interfacial adhesion between ITO electrode and fullerene layer would not provide robust substrate for perovskite solar cells, which would result in physical deterioration of the devices. In light of this, it is essential to optimize the interface cathode to improve the PCE of the PCBM-based normal PSCs.

In light of this, we herein report that the device performance of the normal PSCs can be significantly improved by using organic-inorganic hybrid electron extraction layer (PEIE-LiQ). PEIE and LiQ have been reported to improve the interface properties and resulting device performance of the planar-heterojunction of the perovskite solar cells [18–20]. However, the insulating nature of PEIE may limit efficient charge transport in the devices, while LiQ exhibits tendency to self-aggregate. Very recently, PEIE-LiQ, a hybrid interfacial materials made of PEIE and LiQ, has been demonstrated as a promising interfacial layer for electron transport and extraction in not only organic solar cells but also organic light emitting diodes, because of the synergetic effect of highly conductive nature of alkaline metal complex (LiQ) and excellent film formation of polyethyleneimine ethoxylate (PEIE). PEIE-LiQ possesses unique advantages including excellent film forming property, high optical transparency, good electrical conductivity and high dipole moment that can tune the energy level of the electrode [21,22]. For the normal structure PSCs, PEIE-LiQ would also provide improved photocurrent generation and fill factor (FF), which can be attributed to the optimal electron transport/hole blocking capability. As a result, the device employing PEIE-LiQ as a cathode interfacial layer exhibited improved open circuit voltage ( $V_{OC}$ ), resulting in improved power conversion efficiency up to 15.21% on ITO/glass substrate. More importantly, the low-temperature solution processability of PEIE-LiQ is desirable to be used in flexible substrate, leading to promising PCE value of 14.41% of flexible PSCs on ITO/PEN, demonstrating the efficacy of hybrid electron extraction layer for practical cathode modifier to fabricate compact and continuous cathode interface of high-performance PSCs.

## 2. Experimental

### 2.1. Preparation of materials

Methylamine (Junsei Chemical), hydroiodide (Duksan Chemical) and  $PbI_2$  (Acros) were used as received.  $CH_3NH_3I$  was synthesized and purified according to the method reported elsewhere. The perovskite precursor solution was prepared by dissolving  $PbI_2$  (461 mg, 1.0 M) and  $CH_3NH_3I$  (159 mg, 1.0 M) in the mixture of DMF and DMSO (1.0 ml, 3:7 in volume), and stirred at 70 °C for 30 min. The precursor solution of the hybrid electron extraction layer was prepared by mixing a PEIE solution (0.5 wt% in 2-methoxyethanol) and a LiQ solution (0.1 wt% in 2-methoxyethanol) in 9:1 ratio in volume at room temperature [23]. Spiro-OMeTAD solution doped with lithium bis(trifluoromethanesulfonyl)imide (Li-TFSI) and 4-ter-butyl pyridine was prepared as reported elsewhere [24].

### 2.2. Fabrication of devices

The solar cell devices were fabricated with a normal architecture: ITO/PEIE-LiQ/PCBM/ $CH_3NH_3PbI_3$ /Spiro-OMeTAD/Ag. The ITO-coated glass (10  $\Omega/\square$ ) or ITO-coated PET substrates (ca. 50  $\Omega/\square$ ) were

cleaned by ultrasonication in detergent water, followed by acetone and isopropanol. After drying with nitrogen gas, the glasses were treated with ultraviolet-ozone for 15 min. Then, PEIE-LiQ was coated by spin coating of the precursor solution to form a thin film with thickness of  $\sim 3$  nm. And then PCBM (2 wt% in chlorobenzene) was coated by spin coating at 2000 rpm followed by thermal annealing at 80 °C for 10 min. The perovskite film was coated by anti-solvent washing process, as reported elsewhere [25]. It should be noted here that spin coating of the perovskite precursor solution did not destroy underlying PCBM layer, as examined in Fig. S1 [26]. After the Spiro-OMETAD film was coated as reported elsewhere, Ag (100 nm) was thermally deposited to complete the device preparation. The active area (6.00 mm<sup>2</sup>) was defined using shadow masks.

### 2.3. Characterization

The film morphology of the PEIE-LiQ was observed using an atomic force microscope (Park Systems) in tapping mode. The perovskite thin film was observed by scanning electron microscopy (SEM) (JSM-6010, JEOL). The photoluminescence (PL) of the perovskite thin film was investigated by Fluorolog FL-3 (Horiba). J–V curves were obtained from 4200-SCS (Keithley) under AM 1.5 G (100 mW/cm<sup>2</sup>) illumination, which was calibrated using a NREL-certified photodiode. The incident photon-to-current density efficiency was measured in the lock-in amplifier (Stanford Research Systems SR830) system which records the short-circuit current density under chopped monochromatic light. The bending test of the flexible device was performed using in-house measurement setting with a curvature radius of approximately 5 nm. To measure the ambient stability of the device performance, the device was kept under ambient conditions under complete darkness.

## 3. Results and discussion

Fig. 1a and b shows schematic illustrations of the solar cell devices and materials studied in this work. As depicted in Experimental section, small amount of LiQ was added in PEIE matrix to form a solution processable composite interfacial material. The composite interfacial materials exhibit good optoelectronic and mechanical properties, affording improved device performance in polymer or perovskite solar

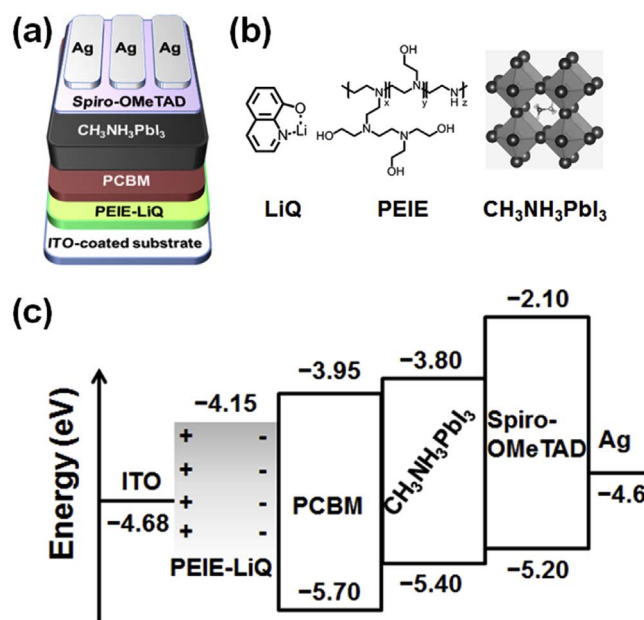


Fig. 1. Schematic device architecture (a), materials (b) and corresponding energy level diagram (c) and SEM cross-sectional image of the perovskite solar cell device (d) studied in this work.

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