



# Device architecture for efficient, low-hysteresis flexible perovskite solar cells: Replacing TiO<sub>2</sub> with C<sub>60</sub> assisted by polyethylenimine ethoxylated interfacial layers

Jaewon Ha<sup>a</sup>, Hoyeon Kim<sup>a</sup>, Hyunwoo Lee<sup>a</sup>, Kyung-Geun Lim<sup>b</sup>, Tae-Woo Lee<sup>c</sup>, Seunghyup Yoo<sup>a,\*</sup>

<sup>a</sup> School of Electrical Engineering, Korea Advanced Institute of Science and Technology (KAIST), 291 Daehak-ro, Yuseong-gu, Daejeon 34141, Republic of Korea

<sup>b</sup> Department of Materials Science and Engineering, Pohang University of Science and Technology (POSTECH), 77 Cheongam-ro, Nam-gu, Pohang, Gyeongbuk 37673, Republic of Korea

<sup>c</sup> Department of Materials Science and Engineering, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea

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## ABSTRACT

We here report methylammonium lead-iodide-based perovskite solar cells (psSCs) in which a TiO<sub>2</sub> layer (a *de-facto* electron transport layer of psSCs) is replaced with a layer of C<sub>60</sub> evaporated onto indium tin oxide layers covered with polyethylenimine ethoxylated (PEIE) layers. Unlike TiO<sub>2</sub> layers requiring a high-temperature sintering, C<sub>60</sub> is deposited while the substrate is held at room temperature, making it compatible with various plastic substrates for flexible psSCs. The PEIE layers are shown to play key roles, not only as an electron-collecting interfacial layer but also as a surface modifier that helps maintain the integrity of the C<sub>60</sub> layers during spin-coating of the perovskite active layers. Using the proposed device architecture, we demonstrate flexible psSCs that exhibit power conversion efficiency as high as 13.3% with low hysteresis.

## 1. Introduction

With the increasing awareness of environment pollution and energy crisis, solar energy has been recognized as an important renewable source of energy, which can be converted and stored through various technologies such as photoelectrochemical cell, photocatalysis [1,2], and photovoltaic (PV) cells. Organic-inorganic lead halide perovskite solar cell (psSC) technology, in particular, has recently gained immense attention as one of the most promising low-cost PV energy-generation technologies [3–7]. With intensive efforts devoted to this emerging PV technology, the power conversion efficiency (PCE) of psSCs has dramatically improved from 3.8% [8] to 22.1% [9] in the past few years. Growing highly crystalline perovskite photoactive layers was shown to be a critical factor in realizing high-PCE psSCs. In addition, most psSCs in a normal configuration, where the cathode is on the substrate side, generally contain a hole blocking and electron transporting layer (ETL) of compact and/or mesoporous titanium oxide (TiO<sub>2</sub>) layers. These layers, however, require a high-temperature around 450–500 °C for sintering process [8,10–12], making it challenging to realize flexible psSCs. Initial efforts were thus made based on a mesoporous-TiO<sub>2</sub>-free inverted planar geometry, in which “inverted” refers to a case where an anode is placed on the substrate. Such trials

include a device geometry where a conducting polymer of poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) or its variant is used as a bottom hole-transporting buffer layer and fullerene derivatives serve as a top electron transport layer [13–16]. Nevertheless, those flexible psSCs exhibited PCE < 10%; this may be regarded consistent with the fact that the PCEs of the psSCs in an inverted planar geometry have been found lower than those of the psSCs in a normal geometry in most of the reports found in the literature [17,18]. Note that having both flexibility and high efficiency could be a critical feature that could differentiate psSCs from other PV technologies. It would thus be highly beneficial to find an alternative ETL processable at a temperature compatible with most plastic substrates. It would be even better, if it could work with normal geometry, in which a solution-based growth of high-quality perovskite active layers could be possible without damaging the underlying ETL. In this regard, various low-temperature processes were proposed for ETL with normal geometry [7,19–21].

Another issue that is important in psSCs is the frequently observed hysteresis behavior in their current density-voltage (*J-V*) hysteresis, which leads to an undesirable PCE difference depending on voltage scan direction and scan rate [22–25]. In general, low PCE due to decrease in the fill factor is observed with forward scan (short-circuit to

\* Corresponding author.

E-mail address: [syoo@ee.kaist.ac.kr](mailto:syoo@ee.kaist.ac.kr) (S. Yoo).

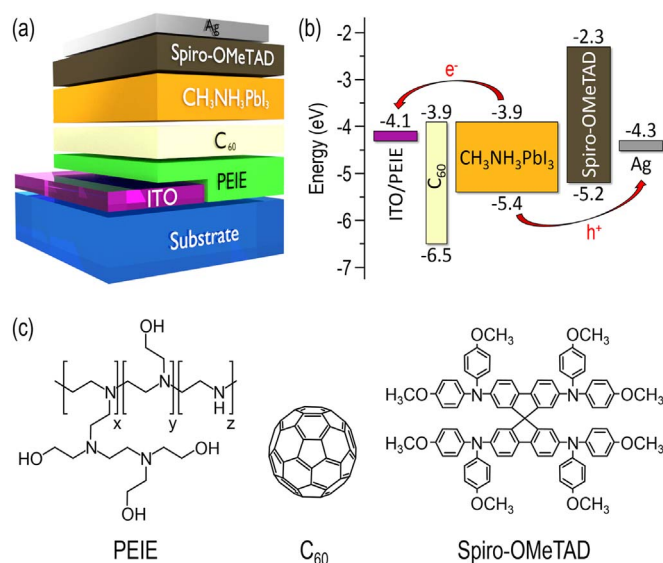
open-circuit), and a fast scan rate tends to increase the PCE difference between forward and reverse scan (open-circuit to short-circuit) [24,25]. Because of its significant practical implications, it is critically important to make sure any new structure or layer configuration, at the least, would not aggravate; but much better, would reduce or suppress this hysteresis effect.

Here, we explore flexible psSCs in a normal geometry for which the bottom ETL of TiO<sub>2</sub> is replaced with evaporated C<sub>60</sub>, well-known in organic electronics for its excellent electron transport properties [26,27]. A previous report by Kim et al. [28] suggested that C<sub>60</sub> can work as an ETL also in the psSCs although it was demonstrated as an ETL prepared on top of perovskite materials in fully vacuum-evaporated psSCs. This left a question about whether it would also work as a bottom ETL in psSCs with solution-processed perovskite layers. In a recent report by Snaith and his coworkers [29], a solution-processed C<sub>60</sub> film was employed to replace a bottom TiO<sub>2</sub> layer, suggesting that it might be plausible. Recently, Yoon et al. [30] reported that thermally evaporated C<sub>60</sub> can indeed work as a bottom ETL, further corroborating the potential of C<sub>60</sub> films as an alternative ETL. Nevertheless, the typical two-step spin-coating method could not be used in both of these studies; instead, the deposition of the lead iodide (PbI<sub>2</sub>) layer was done via thermal evaporation in the former, and one-step spin-coating with diethyl ether dripping method was used in the latter, both of which were motivated mainly by a concern about partial dissolution of C<sub>60</sub> films during spin-coating of PbI<sub>2</sub>. It would thus be helpful to develop a device architecture that is based on C<sub>60</sub> but can work with common two-step spin-coating method as well.

In this study, it turns out that C<sub>60</sub> layers deposited on a bare indium tin oxide (ITO) layer are subject to damage during the spin-coating process for methylammonium lead iodide (CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>; MAPbI<sub>3</sub>) based perovskite photoactive layers. However, those grown on ITOs coated with polyethylenimine ethoxylated (PEIE) [31] are shown to keep their film integrity. It is found that they work not only as ETLs and but also as effective templates for growth of high-quality MAPbI<sub>3</sub>-based perovskite layers. With this method, we demonstrate flexible psSCs with efficiency as high as 13.3% and low hysteresis.

## 2. Experimental

A schematic device structure and energy band diagram [13,30,32,33] of the proposed psSC device and the molecular structure of key materials used therein are shown in Fig. 1(a)–(c), respectively. It



**Fig. 1.** (a) Schematic device structure and (b) energy band diagram of the proposed perovskite solar cell (psSC). (c) Molecular structure of the materials used in this work.

is based on a conventional psSC structure [34] except for the fact that a thermally evaporated C<sub>60</sub> layer is herein used as an ETL instead of mesoporous TiO<sub>2</sub> layers. As a photoactive layer, MAPbI<sub>3</sub>-based perovskite films were prepared using the so-called two-step spin-coating method [13], which consists of sequential spin-coating of PbI<sub>2</sub> and methylammonium iodide (CH<sub>3</sub>NH<sub>3</sub>I; MAI) solutions. For a cathodic interfacial layer, PEIE film was inserted between ITO and the C<sub>60</sub> layer [31].

The PEIE solution was diluted by mixing PEIE (Sigma-Aldrich) with 2-methoxyethanol to a weight concentration of 0.4 wt%, as described by Zhou et al. [31]. Then, PbI<sub>2</sub> (Sigma-Aldrich, 99%) and MAI (1-Material, 99.5%) were dissolved in *N,N*-dimethylformamide (DMF, Sigma-Aldrich, anhydrous, 99.8%) at a concentration of 462 mg/mL and 2-propanol (IPA, Sigma-Aldrich, anhydrous, 99.5%) at a concentration of 20 mg/mL, respectively. The PbI<sub>2</sub> solution was placed on a hot plate held at 100 °C during the entire spin-coating process. Next, 2,2',7,7'-tetrakis(*N,N*-di-*p*-methoxyphenylamine)-9,9'-spirobifluorene (spiro-OMeTAD, Derthon, 99.5%) was dissolved in 72.3 mg/mL chlorobenzene (Sigma-Aldrich, anhydrous, 99.8%). Then 28.8 μL/mL of 4-tert-butylpyridine and 17.5 μL/mL of lithium bis(trifluoromethylsulfonyl)imide (Li-TFSI, Sigma-Aldrich) solution (520 mg/mL Li-TFSI in acetonitrile, 99.8%) were added 30 min before spin-coating.

ITO-coated substrates (AMG glass, 10 Ω/sq. for glass and PECF-IP, Pecell Technologies Inc., 15 Ω/sq. for plastic substrates) were sequentially cleaned by ultrasonication using detergent, deionized water, acetone, and isopropanol for 20 min each. The ITO-coated plastic substrates were based on polyethylene naphthalate (PEN; 200 μm in thickness). Before the cleaning process, the half of ITO pre-coated on a 25×25 mm<sup>2</sup> substrate was patterned out by wet etching process to later define the device area by the overlapped area between ITO and metal cathodes, which were typically about 0.1 cm<sup>2</sup>. The cleaned-substrates were treated with air plasma (PDC-32G, Harrick Plasma) for 5 min. In order to avoid bending during the spin-coating process, the flexible PEN substrates were attached to a temporary carrier glass. The PEIE solution was spin-coated onto a plasma-cleaned ITO substrate at 5000 r.p.m. for 55 s and annealed on a hotplate at 100 °C for 10 min, in ambient air. The PEIE-coated ITO substrates were then loaded into a vacuum thermal evaporator (HS-1100, Digital Optics & Vacuum, < 10<sup>-6</sup> Torr) for deposition of the C<sub>60</sub> (Niche, 99.9%) ETL (25 nm thick), which was shown to decrease to 15 nm after spin-coating of the PbI<sub>2</sub> solution. The C<sub>60</sub>-evaporated substrates were transferred to the outside of the vacuum thermal evaporator for spin-coating of the perovskite photoactive layers and hole transporting layers (HTLs) [13,34]. Subsequently, PbI<sub>2</sub> solution, heated to 100 °C, was spin-coated on a C<sub>60</sub> layer at 8000 r.p.m. for 30 s, and then annealed on a hotplate at 100 °C for 5 min. After the substrates were cooled to room temperature (RT), the MAI solution was spin-coated at 3000 r.p.m. for 30 s and annealed at 100 °C for 5 min. After annealing, a dark brown MAPbI<sub>3</sub> film was formed. Spiro-OMeTAD solution was then spin-coated onto the MAPbI<sub>3</sub> perovskite layer at 4000 r.p.m. for 20 s and dried without any additional thermal annealing. Spin coating and annealing of the PbI<sub>2</sub>, MAI, and spiro-OMeTAD layers were all done inside an environment-controlled chamber, which was maintained at relative humidity (RH) of 30%. These samples were then loaded into the vacuum thermal evaporator for deposition of the silver (Ag, Ulet, 99.99%) layers as top anodes.

The *J-V* characteristics were measured with a sourcemeter unit (Keithley 2611A) under 100 mW/cm<sup>2</sup> illumination (AM1.5G) from a spectral response system (CEP-25ML, Bunkoukeiki) with the uniform irradiation area larger than 27×27 mm<sup>2</sup>. The irradiance of the incident light was verified periodically using a calibrated Si photodiode (BS-520BK, Bunkoukeiki). Measurements of *J-V* proceeded under the following conditions: voltage scan step of 10 mV, and scan delay time of 50 ms between measurement points (scan rate of 0.2 V/s) for reverse scan (from open-circuit to short-circuit) and forward scan (from short-

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