Annealing-Free Efficient Vacuum-Deposited Planar Perovskite Solar Cells with Evaporated Fullerenes as Electron-Selective Layers

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Abstract

We present efficient metal oxide-free and annealing-free planar perovskite solar cells with the regular cell structure using vacuum-deposited fullerenes C_{60} and C_{70} as the electron-selective layers and vacuum-processed perovskites as the light absorbers. The devices with an ultrathin C_{60} layer (5.5 nm) yielded an average power conversion efficiency of 14.3% and a maximum efficiency of 15.7%. The best-performing cell produced a steady-state efficiency of 14.6%. The high performance is attributed to the efficient blocking of holes and extraction of electrons by C_{60} due to a favorable energy level alignment between the C_{60} and the fluorine-doped tin oxide electrodes. With the realization of efficient cells, the annealing-free vacuum deposition of perovskite absorbers and C_{60} or C_{70} electron-selective layers and intermediate layers demonstrates its power for fabricating all-perovskite tandem solar cells.

Introduction

Organic-inorganic lead halide perovskite based solar cells have been extensively investigated over the past few years due to the excellent photovoltaic properties of perovskites such as high optical absorption coefficient, long charge carrier lifetime, high carrier mobility, and band gap tunability [1-5]. Moreover, perovskite solar cells can be made by low cost solution process at low temperature, enabling roll-to-roll manufacturing using flexible substrates. Perovskite-based solar cells have been considered recently as the most promising photovoltaic technology [6, 7], with their power conversion efficiencies (PCEs) rapidly rising from 3.8% to a certified 20.1% [8-15].

The high efficiencies of perovskite solar cells are largely due to the high open circuit voltages (V_{oc} s), which are attributed to the long carrier lifetime [1, 2]. It has been shown that the dominant defects do not create deep energy levels in the band gaps of perovskite absorbers [3, 4], distinctively different from that seen in the common inorganic solar cell materials. The V_{oc} deficit, defined by $E_g/q - V_{oc}$, of the record perovskite solar cells, is as small as 0.42 V, approaching that of the best single crystal GaAs thin film solar cell [12]. Therefore, the V_{oc} of the high-efficiency perovskite solar cells has nearly reached its theoretical value. Unlike V_{oc} , the short-circuit current density (J_{sc}) of the high-performing perovskite solar cells still has considerable room for improvement. Typically, high-efficiency perovskite solar cells use approximately 350-450

nm thick absorbers, which are not sufficiently thick to fully absorb the incident light. Unfortunately, increasing the thickness of the perovskite absorbers could only lead to lower efficiencies, indicating that intrinsic electronic property issues exist for the low temperature processed polycrystalline perovskite thin films. Fabricating tandem cells is a legitimate option for further improving the overall device efficiency and has recently attracted great attention. So far, most perovskite tandem cells reported consist of perovskite solar cells as the top subcells and inorganic solar cells such as Si and Cu(In,Ga)Se₂ cells as the bottom subcells [16-18]. Efficient all-perovskite tandem cells, a preferred approach, have not been reported yet. Part of the challenge stems from depositing intermediate layers without damaging the existing underlying subcells.

The planar cell architecture is preferred for fabricating tandem cells. A typical planar cell consists of a solution-processed or vacuum-processed perovskite absorber layer sandwiched between an electron-selective layer (ESL) and a hole-selective layer (HSL). To avoid any damage to the existing subcell, the ESL or HSL and the perovskite absorber layer should be deposited preferably by vacuum processes and without additional annealing. Furthermore, compared to solution processes, vacuum processes are expected to result in thin films with better uniformity, smoother surfaces, complete coverage, and more accurately controlled thicknesses. However, to date efficient annealing-free vacuum-processed perovskite solar cells have not been reported. The current high-

performing perovskite cells generally use either high-temperature processed metal oxide layers such as TiO_2 [19, 20], solution-processed low temperature materials such as ZnO [21], SnO_2 [22], and Cs_2CO_3 [23], or organic materials such as [6,6]-phenyl-C₆₁-butyric acid methyl ester (PC_{60}BM) and C_{60} as the ESL [24]. The high temperature and/or solution processes could potentially either thermally decompose the existing perovskites or dissolve the underlying subcells and therefore would be incapable of producing high-efficiency tandem cells.

Vacuum-processed perovskite-based solar cells have been demonstrated by several groups, and the devices have exhibited promising efficiencies in both regular and inverted architectures [25-31]. The first successful vapor-deposited perovskite solar cells with 15% efficiency were presented in a regular structure and used high temperature processed compact TiO_2 as the ESL [25]. Most high-efficiency vacuum-processed perovskite based solar cells used the inverted structure [26, 27, 31], among which the best reported cells with the highest PCE of 16.5% employed solution-processed poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) and various polymers as the HSLs [28]. Other groups have also reported efficient cells using vacuum-processed perovskite absorbers [26, 27, 30, 31]. Likewise, these cells used either high temperature processed compact TiO_2 or solution-processed HSL/ESLs.

Here, we demonstrate efficient annealing-free vacuum-processed perovskite solar cells using thermal evaporation of both the fullerene

C₆₀ as the ESL and the perovskite as the light absorber. We used fullerene C₆₀, because it can function as an ESL [32] and can also be thermally evaporated [33, 34]. Thermally evaporated C₆₀ films exhibit good electronic properties without post-deposition annealing [35]. The devices with an ultrathin C₆₀ layer (5.5 nm) showed an average PCE of 14.3% and a maximum PCE of 15.7%. The best-performing cell produced a steady-state efficiency of 14.6%. Our detailed device characterization revealed that the high performance of our annealing-free vacuum-processed perovskite cells resulted from the use of an ultrathin C₆₀ layer which effectively transfers the electrons and blocks the holes due to the favorable energy level alignment between C₆₀ and fluorine-doped tin oxide (FTO) electrodes. With the realization of efficient cells, the annealing-free vacuum deposition of perovskite absorbers and C₆₀ or C₇₀ electron-selective layers and intermediate layers demonstrates its power for fabricating all-perovskite tandem solar cells.

Experimental section

Perovskite synthesis

The deposition of the perovskite films was carried out using a thermal evaporator integrated with a glove box. CH₃NH₃I and PbI₂ were loaded into their individual crucibles. Two quartz crystal microbalances are placed close to each crucible, monitoring the rate of each evaporation source. In our approach, it is difficult to calibrate and monitor the CH₃NH₃I rate; therefore, it was preferable to monitor the CH₃NH₃I vapor phase pressure

instead. After the crucibles are filled with $\text{CH}_3\text{NH}_3\text{I}$ and PbI_2 , the chamber is evacuated to a base pressure of 3×10^{-7} Torr. The PbI_2 crucible is heated until its deposition rate reaches our target value. Meanwhile, the $\text{CH}_3\text{NH}_3\text{I}$ crucible is heated until the vacuum pressure is 5×10^{-5} Torr. For an optimal deposition condition of the perovskite film, the $\text{CH}_3\text{NH}_3\text{I}$ vapor pressure is kept at 5×10^{-5} Torr by controlling the temperature of $\text{CH}_3\text{NH}_3\text{I}$ crucible and the deposition rate of PbI_2 was kept at $0.75 \text{ \AA/s}$, a procedure which achieves the best crystalline perovskite films. The samples for TRPL measurement were coated by poly(methyl methacrylate) (PMMA).

Device fabrication

Devices are made on FTO substrates cleaned by sonication in deionized water, acetone, and isopropanol, each for 15 minutes. C_{60} or C_{70} layers with variable thicknesses were thermally deposited on the FTO substrates, followed by $\text{CH}_3\text{NH}_3\text{I}$ vapor-assisted perovskite deposition with the growth conditions of PbI_2 rate at $0.75 \text{ \AA/s}$ and $\text{CH}_3\text{NH}_3\text{I}$ pressure at 5×10^{-5} Torr. After the perovskite deposition, the substrates are removed from the chamber. A solution of electron blocking material was spin-coated on the perovskite film at 6000 rpm for 60 s, which consists of 68 mM 2,2',7,7'-tetrakis-(N,N-di-p-methoxyphenylamine)-9,9'-spirobifluorene (Spiro-OMeTAD) (Shenzhen Feiming Science and Technology Co., Ltd., 99.0%), 55 mM tert-butylpyridine (TBP) (Sigma-Aldrich, 96%), and 26 mM lithium bis(trifluoromethanesulfonyl)imide (Li-TFSI) (Sigma-Aldrich, 99.95%) dissolved in a mixed solvent of acetonitrile and chlorobenzene with a

volume ratio of 1:10. Finally, 40 nm Au was thermally deposited as top electrode, generating the active area of 0.08 cm^2 for all devices.

Film and device characterization

Perovskite films were imaged with a high resolution field emission scanning electron microscope (SEM) (Hitachi S-4800). Atomic force microscopy (AFM) images were acquired on a Veeco Nanoscope IIIA instrument. The crystal structure of the $\text{CH}_3\text{NH}_3\text{PbI}_3$ film on the C_{60} -coated FTO was examined by X-ray diffraction (XRD) (Rigaku Ultima III) with $\text{Cu K}\alpha$ radiation under operating conditions of 40 kV and 44 mA. The absorption spectra of the perovskite $\text{CH}_3\text{NH}_3\text{PbI}_3$ film on C_{60} -coated FTO and the C_{60} layers with varying thicknesses on FTO substrates were measured by a UV-Vis spectrophotometer (CARY5000, Varian). All layer thicknesses were determined using a Dektak surface profiler and calibrated by spectroscopic ellipsometry. Current density-voltage (J - V) curves were measured in air under AM1.5G solar irradiation (PV Measurements Inc.) with a Keithley 2400 Source Meter. EQE spectra were performed on a commercial QE system (PV Measurements Inc.). Transient photoluminescence decays were measured with a time correlated single photon counting system, where the light source is a Fianium Supercontinuum high power broadband fiber filter (SC400-2-PP). The excitation wavelength was 500 nm with a power of $\sim 25\text{ }\mu\text{W}$ on a spot size of 0.02 mm^2 . The decay curve was fitted using a double exponential model.

Results and discussion

Figure 1a depicts the geometry of our perovskite solar cells in the regular cell structure using thermally evaporated compact C_{60} as the ESL on an FTO coated glass substrate and dual evaporated $CH_3NH_3PbI_3$ perovskite as the absorber. A 2,2',7,7'-tetrakis-(N,N-di-p-methoxyphenylamine)-9,9'-spirobifluorene (spiro-OMeTAD) as the HSL was spin-coated on the perovskite layer. Finally, the device was finished by the thermal evaporation of Au as the top electrode. It has been reported that the roles of C_{60} in inverted planar perovskite solar cells are to facilitate the electron transport as a buffer layer and smoothen the $PC_{60}BM$ surface [24, 36]. In organic solar cells, direct contact between C_{60} and electrodes is not preferred due to the hole-injection induced quenching of excitons at C_{60} /electrode interfaces, leading to large charge carrier recombination and leakage currents [33, 37, 38]. Previous research has used C_{60} and $PC_{60}BM$ in combination with metal oxides or polymers as the ESLs, such as TiO_2 /self-assembled monolayer (SAM)- C_{60} [39, 40], $ZnO/PC_{60}BM$ [41], polyethylenimine (PEI)/ $PC_{60}BM$ [42] in the regular cell architecture to enhance the electron transfer. $PC_{60}BM$ and C_{60} alone can also function as effective ESLs in both regular and inverted cell architectures [28, 32, 43]. However, $PC_{60}BM$ cannot be deposited by vacuum techniques, whereas it is well known that C_{60} thin films can be deposited by thermal evaporation. So far, only solution-processed C_{60} as the ESL has been applied in perovskite solar cells with the regular cell architecture [32]. To achieve efficient cells, it would be advantageous if the deposition of the C_{60} and perovskite films

could be conducted sequentially without breaking vacuum, ensuring a high-quality interface between these two layers.

Figure 1b shows the energy band diagram of our perovskite solar cells. On one hand, the lowest unoccupied molecular orbital (LUMO) level of C_{60} is slightly below the LUMO level of $CH_3NH_3PbI_3$, allowing the smooth electron transfer from $CH_3NH_3PbI_3$ to FTO. On the other hand, the highest occupied molecular orbital (HOMO) level of C_{60} is much lower than the HOMO level of $CH_3NH_3PbI_3$, effectively blocking the holes from $CH_3NH_3PbI_3$. C_{60} is therefore an ESL, reducing charge carrier recombination and shunt currents.

Figure 2a shows the top-view scanning electron microscopy (SEM) image of an evaporated perovskite film on the C_{60} -coated FTO substrate. The SEM image presents a uniform and compact surface with complete coverage and an average grain size of 150-200 nm. On the cross-sectional SEM image (**Figure SI, SI**), a smooth perovskite film is shown with a thickness of approximately 320 nm. Relatively thin perovskite layers are necessary in all-perovskite tandem solar cells in order to achieve current matching [44], which is hence our focus in this work. Such thin films can also generate respectable photocurrents. The surface topography of the perovskite film was measured by using tapping-mode atomic force microscope (AFM). **Figure 2b** shows the 3D image of the surface morphology, demonstrating a root mean square (RMS) roughness of 15 nm. Our vacuum-processed perovskite layer is pinhole-free and sufficiently dense to

prevent the penetration of spiro-OMeTAD through the perovskite layer to directly contact C₆₀/FTO electrode, thus avoiding any leakage. The absorption spectrum of the perovskite film is shown in **Figure 2c**, which exhibits a broad absorption ranging from 400 to 800 nm where an absorption tail occurs. The X-ray diffraction patterns in **Figure 2d** confirm that the perovskite film is crystalline with the tetragonal structure and random orientation [27]. The peaks at 14.05° and 28.3° correspond to (110) and (220) planes. It is also found that a small amount of PbI₂ (peak at 12.7°) present in our perovskite films can effectively benefit the device performance and reproducibility as reported previously [36].

In order to investigate the effect of C₆₀ layer thickness on the device performance, the current density-voltage (*J-V*) characteristics of the devices without C₆₀ and with varying thicknesses of the C₆₀ layer under AM1.5 G 100 mW/cm² illumination are shown in **Figure 3a**. The performance summary of corresponding devices is tabulated in Table **I** and the performance statistics of these devices are shown in **Figure S2 (SI)**. As a control, the device without C₆₀ has a PCE of 4.4% with a *V*_{oc} of 1.04 V, a *J*_{sc} of 9.4 mA/cm², and an FF of 44.7%, exhibiting very poor performance and reproducibility due to ineffective electron extraction and hole blocking at the FTO/perovskite interface. This is evidenced by the low shunt resistance (*R*_{sh}) of 226 Ωcm² and also the large leakage current under reverse bias as shown in the dark *J-V* characteristics (**Figure S3b, SI**). Although a series resistance (*R*_s) of 2.0 Ωcm² is reached, the FF is quite low.

The devices with C₆₀ inserted exhibit simultaneous enhancement of all primary performance parameters.

It is worth highlighting that the thermal evaporation technique is advantageous compared with the spin-coating technique since the materials can be evenly and uniformly coated on substrates, especially for an ultrathin layer, i.e. the deposition follows the surface topology of the substrates. Therefore, there is no distinct difference in the surface roughness (root mean square (RMS) roughness) between bare FTO and C₆₀-coated FTO with various thicknesses, and similar morphology occurs in all samples, as shown in **Figure S4 (SI)**. A thickness variation of the C₆₀ layer does not strongly influence the V_{oc} , however, remarkable changes of J_{sc} and FF have been observed.

When a 2.5 nm thick C₆₀ layer is inserted, the cell efficiency greatly increases to 13.4% with a V_{oc} of 1.05 V, a J_{sc} of 18.0 mA/cm², and an FF of 71.1%. Merely 2.5 nm thick C₆₀ layer can effectively transfer electrons, block holes, and suppress charge recombination at the FTO/perovskite interface, leading to a significant enhancement of J_{sc} and FF. Its R_s decreases to 1.2 Ω cm² and R_{sh} increases to 1303 Ω cm². With further increasing the thickness of the C₆₀ layer to 5.5 nm, the FTO surface is completely covered, evidenced by the continuous improvement in efficiency (Table 1) and device reproducibility (**Figure S5, SI**). The device with 5.5 nm C₆₀ achieves a PCE of 14.9% with a V_{oc} of 1.06 V, a J_{sc} of 18.3 mA/cm², and an FF of 77.0%. To the best of our knowledge, such FF value is the highest among those reported for vacuum-

deposited perovskite solar cells. This is primarily attributed to the minimized charge carrier recombination at the interface between the perovskite and C₆₀ layer as well as the effective electron extraction, evidenced by largely increased R_{sh} (2444 Ωcm^2) and small R_s (2.4 Ωcm^2). Compared with the devices without C₆₀, the devices with 5.5 nm C₆₀ have negligibly low current under reverse bias, as shown in the dark J - V characteristics (**Figure S3b**, SI). This is attributed to the efficient hole blocking by the C₆₀ layer.

Thicker C₆₀ layers (10 and 15 nm) lead to a reduction in device efficiency, primarily due to the decreased J_{sc} and FF (for the cell with 10 nm C₆₀: $R_s = 2.6 \Omega\text{cm}^2$ and $R_{sh} = 2599 \Omega\text{cm}^2$; for the cell with 15 nm C₆₀: $R_s = 3.0 \Omega\text{cm}^2$ and $R_{sh} = 2811 \Omega\text{cm}^2$). It should be noted that C₆₀ has a band gap of 1.7 eV, causing excessive light loss if the layer is too thick, as shown in **Figure 4**. The C₆₀ layers with 2.5 and 5.5 nm have higher transmittance throughout the spectral range; however, the thick C₆₀ layers with 10 and 15 nm have significant light absorption from 350 to 550 nm, which decreases the incident light reaching the perovskite layer and leads to the reduction in J_{sc} . Moreover, with the increase of C₆₀ layer thickness, R_s is increased correspondingly due to the relatively low electron mobility in C₆₀ compared with inorganic metal oxides [35], resulting in excess R_s and slightly reduced FF.

Figure 3b shows the external quantum efficiency (EQE) spectra of perovskite cells incorporating various thickness of C₆₀ layers with as the

ESLs. The devices with 2.5 and 5.5 nm thick C₆₀ layers exhibit higher EQE values than those with 10 and 15 nm C₆₀ due to the light loss caused by absorption of thick C₆₀ layers in the range of 350-550 nm, demonstrated by the transmittance spectra shown in **Figure 4**. The device with a 5.5 nm thick C₆₀ possesses the highest EQE, producing an EQE-integrated J_{sc} of 18.3 mA/cm², in good agreement with J_{sc} from the J - V curve. It is interesting that all EQE curves of our evaporated perovskite solar cells present an obvious dip ranging from 650 to 750 nm, which is likely ascribed to optical cavity effect [28]. Therefore, the photocurrent of our vapor-assisted perovskite solar cells can be further enhanced by careful photon management.

To evaluate the reproducibility of our devices using an ultrathin C₆₀ as the electron-selective layer, 30 devices with 5.5 nm C₆₀ layers in several batches were fabricated. As shown in the histogram of the corresponding device data in **Figure S5**, the average PCE for these devices is 14.3% and the highest PCE is 15.3%. Therefore, our cells using the ultrathin C₆₀ interface layers and evaporated perovskite absorbers appear to have good reproducibility.

One of the crucial issues of perovskite solar cells is the hysteresis in J - V curves, which leads to a strong dependence of device performance on the voltage scanning direction, the scanning rate, and the device architecture [45]. Such hysteresis is likely to overestimate the device performance. **Figure 5a** shows the J - V characteristics of the device with 5.5 nm C₆₀ as the ESL under 100 mW/cm² illumination applying different voltage

scanning directions. It can be seen that our devices exhibit very small hysteresis, producing a reverse-forward PCE of 14.9% with a V_{oc} of 1.06 V, a J_{sc} of 18.3 mA/cm², and an FF of 77.0% and a forward-reverse PCE of 14.1% with a V_{oc} of 1.06 V, a J_{sc} of 18.1 mA/cm², and an FF of 73.7%. To evaluate the accuracy of device performance, the steady-state efficiency measurement is a reliable method that reflects the actual performance under operation. **Figure 5b** shows the steady-state J_{sc} and PCE of the device with 5.5 nm C₆₀ as the ESL at a constant bias of 0.861 V under AM1.5 G 100 mW/cm² illumination. A steady-state J_{sc} of approximately 17 mA/cm² and stabilized output power of 14.6% is achieved. The hysteresis and steady-state data for devices with 10 nm C₆₀ as the ESL are also shown in **Figure S6 (SI)**. However, the devices without a C₆₀ layer have serious hysteresis in the J - V curves (**Figure S3a, SI**). Our results on significantly reduced hysteresis and steady-state efficiency close to J - V efficiency indicate that our evaporated perovskite films not only have a compact structure with low densities of defects that could trap electrons and holes [4], but also have good interfacial contact to the C₆₀ layer with surface passivation [43].

To understand the charge carrier recombination in the devices with C₆₀ as the ESL, the J - V characteristics were measured under different light intensities ranging from 1 to 100 mW/cm², as shown in **Figure 6a**. **Figure 6b** shows the power law dependence of J_{sc} on light intensity ($J \propto I^\alpha$), which exhibits a linear relation on a double logarithmic scale with $\alpha = 0.99$. A solar cell with no space charge effect will have an α value close to 1 [46-48]. Therefore, our devices with C₆₀ as the ESL do not appear to be space

charge limited. In contrast, the devices without C_{60} have a linear relation with $\alpha = 1.20$, indicating significant space charge built-up in the devices and charge carrier losses occur as shown in **Figure S7 (SI)** [49-52]. **Figure 6c** shows V_{oc} as a function of logarithmic light intensity. V_{oc} appears to increase monotonically with logarithmic light intensity, indicating that trap-assisted Shockley-Read-Hall recombination is prevalent in C_{60} -based perovskite solar cells [36, 49]. The dependence of FF on light intensity is shown in **Figure 6d**. A maximum FF of 77.9% is reached at 20 mW/cm^2 , implying that the charge carrier recombination is minimal at a relatively lower light intensity due to the low carrier concentration generated. The results suggest that an ultrathin layer of C_{60} can efficiently extract the charge carriers and suppress the charge recombination.

Figure 7 compares the charge carrier lifetime as measured by transient photoluminescence (TRPL) in perovskite deposited on FTO to that in perovskite deposited on FTO/ C_{60} . The TRPL decay curves were fitted assuming a bi-exponential decay model. For the FTO/perovskite sample, τ_1 is 14.9 ns (44.1%) and τ_2 is 75.9 ns (55.9%), leading to a weighted average of approximately 49.0 ns. For the FTO/ C_{60} /perovskite sample, τ_1 is 2.0 ns (74.5%) and τ_2 is 8.7 ns (25.5%) with a weighted average of approximately 3.7 ns. Thus, it is clear that the insertion of a thin C_{60} layer greatly enhances the electron extraction at the perovskite/FTO interface. This is consistent with the device characteristics discussed above.

By optimizing the thickness of spiro-OMeTAD layer, the maximum PCE of our cells can reach 15.7% with a V_{oc} of 1.10 V, a J_{sc} of 18.9 mA/cm², and an FF of 75.4%. Its J - V characteristics and EQE spectrum are shown in **Figure 8**. The EQE-integrated J_{sc} can reach 18.5 mA/cm², close to that from J - V curve. Its steady-state photocurrent and efficiency at a bias of 0.906 V are shown in **Figure S8**. Moreover, the dip of EQE spectrum shifts to blue range slightly due to the cavity effect by the spiro-OMeTAD thickness variation.

We find that thermally evaporated fullerene C₇₀ can also act as the ESLs in our vacuum-processed regular perovskite solar cells. As listed in **Table 1**, the device with 5.5 nm C₇₀ shows a PCE of 14.9% with a V_{oc} of 1.03 V, a J_{sc} of 18.6 mA/cm², and an FF of 77.7%, as shown in **Figure S9a**. Its EQE spectrum is also shown in Figure S9b. The J_{sc} of the cells with C₇₀ ESLs is slightly higher than that of the cells with C₆₀ ESLs, which is due to the fact that C₇₀ has a slightly larger band gap (2.0 eV) than C₆₀ (1.7 eV), leading to less light loss. Therefore, there is still room to enhance our annealing-free vacuum-processed regular perovskite solar cells.

Conclusion

We have demonstrated high-efficiency vacuum-processed regular perovskite solar cells with an ultrathin metal-oxide-free and annealing-free C₆₀ or C₇₀ as the electron-selective layer. The devices with C₆₀ interfacial layers exhibit high fill factors and high performance, which can be attributed to minimal recombination and fast electron extraction at the interface between the perovskite and C₆₀ layers. The C₆₀ layer not

only serves as a hole blocking layer and an electron-selective contact but also it forms an intimate contact with the perovskite layer and promotes the growth of a well crystallized film. Light intensity dependent characteristics and TRPL results indicate increased charge carrier transfer at the interface, leading to a maximum efficiency of 15.7% and a steady-state efficiency of 14.6%. With the realization of efficient cells, the annealing-free vacuum deposition of perovskite absorbers and fullerenes ESLs shows its promise for fabricating all-perovskite tandem solar cells on flexible substrates.

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Figure 1. (a) Device structure and (b) Energy band diagram of our perovskite solar cells with C_{60} as the ESLs.

Figure 2. (a) SEM image and (b) tapping-mode AFM image of an evaporated perovskite film on a C₆₀-deposited FTO substrate, (c) UV-Vis absorption spectrum of the perovskite film, and (d) X-ray diffraction patterns for perovskite films on C₆₀-coated FTO along with FTO and C₆₀-coated FTO patterns for reference.

Figure 3. (a) J - V characteristics under AM1.5G 100 mW/cm² illumination and (b) EQE spectra of perovskite solar cells with varying C₆₀ layer thickness.

Figure 4. Transmittance spectra of C_{60} layers with different thickness.

Figure 5. (a) J - V characteristics of the device with 5.5 nm C_{60} as electron-selective layer under $100 \text{ mW}/\text{cm}^2$ illumination under different voltage scanning direction: Reverse-Forward and Forward-Reverse and (b) Steady-state J_{sc} and PCE of the device with 5.5 nm C_{60} as the electron-selective layer at a constant bias of 0.861 V under $100 \text{ mW}/\text{cm}^2$ illumination.

Figure 6. Light-intensity dependence of perovskite solar cell performance with 5.5 nm C_{60} as the electron-selective layer: (a) J - V characteristics, (b) J_{sc} versus light intensity, (c) V_{oc} versus light intensity and (d) FF versus light intensity. The light intensity ranges from $1 \text{ mW}/\text{cm}^2$ to $100 \text{ mW}/\text{cm}^2$

Figure 7. Transient photoluminescence (TRPL) measurements comparing charge carrier lifetime in the perovskite films deposited on FTO and on FTO/C₆₀ (5.5 nm).

Figure 8. (a) J - V characteristic under AM1.5G 100 mW/cm² illumination and (b) EQE spectrum of our best-performing perovskite solar cells with 5.5 nm C₆₀ as the ESL.

Table 1. Summary of performance metrics of the devices with varying thicknesses of C₆₀ layer as the ESL and with 5.5 nm C₇₀ as the ESL.

	J_{sc}	V_{oc}	FF	PCE
	[mA cm ⁻²]	[V]	[%]	[%]
C ₆₀ = 0.0 nm	9.4	1.04	44.7	4.4
C ₆₀ = 2.5 nm	18.0	1.05	71.1	13.4
C ₆₀ = 5.5 nm	18.3	1.06	77.0	14.9
C ₆₀ = 10.0 nm	17.3	1.07	75.2	13.9
C ₆₀ = 15.0 nm	16.1	1.06	70.4	12.0
C ₇₀ = 5.5 nm	18.6	1.03	77.7	14.9

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Graphical abstract