Pyridine-functionalized fullerene derivative as an independent electron transport layer enabling efficient and hysteresis-free regular perovskite solar cells

Ye Wang1 | Bairu Li2 | Lingbo Jia2 | Bo Zhang1 | He Zhang2 | Panfei Liang1 | Muqing Chen2 | Hua Yang1 | Xinqing Wang1 | Shangfeng Yang2

1 College of Materials and Chemistry, China Jiliang University, Hangzhou 310018, China
2 Hefei National Laboratory for Physical Sciences at Microscale, CAS Key Laboratory of Materials for Energy Conversion, Department of Materials Science and Engineering, University of Science and Technology of China, Hefei 230026, China

Correspondence
Muqing Chen and Shangfeng Yang, Hefei National Laboratory for Physical Sciences at Microscale, CAS Key Laboratory of Materials for Energy Conversion, Department of Materials Science and Engineering, University of Science and Technology of China, Hefei 230026, China.
Email: mqchen@ustc.edu.cn; sfyang@ustc.edu.cn
Xinqing Wang, College of Materials and Chemistry, China Jiliang University, Hangzhou 310018, China.
Email: wxqnano@cjlu.edu.cn
Ye Wang, Bairu Li, and Lingbo Jia contributed equally to this work.

Funding information
National Key Research and Development Program of China, Grant/Award Numbers: 2017YFA0402800, 2019YFF0217202; National Natural Science Foundation of China, Grant/Award Numbers: 51925206, U1932214, 51602097

Abstract
Suitable electron transport materials bearing good interfacial contact, improved electron transport ability, and matched energy levels are indispensable for developing efficient perovskite solar cells (PSCs). Herein, regular (n-i-p) planar Cs0.05FA0.83MA0.12PbI2.55Br0.45 (CsFAMA) PSC devices were fabricated using a pyridine-functionalized fullerene derivative (C_{60}-3-BPy) as an independent electron transport layer (ETL), delivering a decent power conversion efficiency (PCE) of 18.22%, which is dramatically higher than that of the control device based on [6,6]-phenyl-C_{61}-butyric acid methyl ester (PCBM) ETL (15.70%). The energy level offset between C_{60}-3-BPy and the perovskite is smaller than that based on PCBM ETL, which is beneficial for efficient ohmic contact in ETL/perovskite interface and improved open-circuit voltage (V_{oc}). Moreover, C_{60}-3-BPy affords strong coordination interactions with perovskite, leading to an improved film quality of the perovskite layer with enlarged grain size and decreased trap state density, which contribute to facilitated electron extraction as reflected by the increases of both the fill factor (FF) and the short-circuit current (I_{sc}). C_{60}-3-BPy-facilitated electron extraction further results in hysteresis-free devices.

KEYWORDS
electron transport layer, fullerene, interfacial engineering, perovskite solar cells, pyridine

1 INTRODUCTION

Organic-inorganic hybrid perovskite solar cells (PSCs) have gained increased interest in recent years, improving their power conversion efficiency (PCE) and device stability owing to the outstanding properties of perovskite materials, which are beneficial for the sufficient utilization of the solar spectrum. Through collaborative
innovation of improving device structure and modifying the perovskite/interface layer interface, tuning the perovskite composition, crystalline phase and morphology of the perovskite film, the certified PCE of PSCs have exceeded 25.5%. [7] Although great progress has been made on device efficiency and stability, continuous efforts are still being devoted to improving the development of high-performance PSCs. Regular (n-i-p) structure PSCs have exhibited a faster evolution compared to that of inverted (p-i-n) structure PSCs, and the classic device structure of n-i-p PSCs consists of a perovskite layer sandwiched between an underneath electron transport layer (ETL) and an upper hole transport layer (HTL). [8,9] Specifically, ETLs featuring an excellent electron-transporting ability, good optoelectronic properties and a compatible conduction band are beneficial for the charge transfer, morphology and crystallization regulation of perovskite films. [10,11] The most commonly used ETLs within high-efficiency n-i-p PSCs consist of titanium dioxide (TiO2). [2,10,11] However, the compact TiO2 ETL obtained via a high-temperature sintering treatment (>450°C) significantly limits their application in flexible devices. [12,13] Moreover, the TiO2 ETL obtained from a low-temperature procedure has the disadvantage of presenting low electrical conductivity and a high trap state density, severely hampering the PCE elevation as well as the long-term stability of the devices. [14,15]

A series of strategies, including morphology optimization, doping, and interface modifications applied in the low-temperature solution-processed TiO2 ETL have been proposed to address the above-mentioned issues [11,16,17]. Therefore, developing novel metal-oxide-free ETLs to avoid the drawback of TiO2 ETLs for improved device performance has become a desirable alternative.

Fullerene derivatives have become efficient ETLs applied in n-i-p PSCs due to the advantages such as excellent electron mobility, low-temperature solution process, and tailored molecular structure and energy level. [18–20] Seok et al. deposited [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) onto a flexible FTO/PEI substrate to construct n-i-p MAPbI3-based PSCs, achieving a PCE of 11.1%. [21] Snaith et al. employed cross-linked fullerene derivative (C60-3-BPy) to fabricate CH3NH3PbI3–xClx-based n-i-p PSCs, achieving a PCE of 18.29%, which outperformed that of the control devices based on PCBM ETL (15.70%). The superior performance of PSCs with C60-3-BPy ETL was attributed to the fact that C60-3-BPy enables effective regulation of the work function of the ITO cathode as well as the crystalline quality of the perovskite film.

## 2 | RESULTS AND DISCUSSION

The preparation procedure for the n-i-p planar CsFAMA PSCs and the device architecture is shown in Figure 1A. Briefly, the chlorobenzene solution containing PCBM or C60-3-BPy with a concentration of 5 mg ml-1 was spin-coated onto the underneath ITO substrates. Thereafter, a CsFAMA perovskite precursor solution was spin-coated onto the fullerene derivative ETLs. After depositing the Spiro-OMeTAD HTL and Au electrode, the resultant PSCs were measured to evaluate the performance of the ETLs. The current-voltage curves of the PSCs with PCBM or C60-3-BPy as ETLs are shown in Figure 1B and the detailed photovoltaic parameters were recorded in Table 1. The PSCs with C60-3-BPy ETL achieved an average PCE of 17.43%, which is much higher than that of devices (14.87%) with PCBM as the ETL, obtained from a Voc of 1.07 V, a Jsc of 21.82 mA cm-2, and an FF of 74.86%. The best PCE of PSCs with C60-3-BPy ETL achieved so far reached 18.22%. To ensure accuracy in device measurements, more than 30 independent devices bearing PCBM ETL or C60-3-BPy ETL were prepared, and the statistical photovoltaic parameters are portrayed in Figure S1. The narrower PCE distribution for the device with C60-3-BPy as the ETL shown in the histograms (Figure. S2) suggests the reliability of the device fabrication procedure. Furthermore, according
FIGURE 1  A, Schematic structure of the n-i-p PSC device and the chemical structures C$_{60}$-3-BPy. B, J-V curves of the n-i-p PSC devices with PCBM, C$_{60}$-3-BPy as ETLs. The measurements were carried out under illumination of an AM 1.5 solar simulator (100 mW cm$^{-2}$) in air, and reverse scanning (from open-circuit voltage to short-circuit) with a speed of 100 mV s$^{-1}$ was applied. C, EQE spectra of the devices with PCBM, C$_{60}$-3-BPy as ETLs measured in air.

TABLE 1  Photovoltaic parameters of regular CAFAMA PHJ PSCs with PCBM and C$_{60}$-3-BPy as ETLs

| Device | ETL       | $V_{oc}$ (V) | $J_{sc}$ (mA cm$^{-2}$) | FF (%)  | PCE (%) | $Rs$ ($\Omega \cdot$ cm$^2$) | $Rsh$ ($\Omega \cdot$ cm$^2$) |
|--------|-----------|--------------|-------------------------|---------|---------|-----------------------------|-----------------------------|
| A      | PCBM      | 1.03 ± 0.03  | 21.27 ± 0.47            | 68.18 ± 3.81 | 14.87 ± 0.77 | 7.1                        | 2451.3                      |
| B      | C$_{60}$-3-BPy | 1.07 ± 0.02  | 21.82 ± 0.30            | 74.86 ± 1.27 | 17.43 ± 0.48 | 18.22                      | 4.5                         | 4446.6                     |

$^a$Averaged over 30 devices fabricated independently.
$^b$Rs and Rsh are obtained by the PCE measurement system.

to the in-depth comparison of these photovoltaic parameters, it is clear that the PCE enhancement of the device with a C$_{60}$-3-BPy ETL is mainly due to the significantly increased FF (from 68.18% to 74.86%, ~9.79% enhancement) and slightly increased $J_{sc}$ (from 21.27 mA cm$^{-2}$ to 21.82 mA cm$^{-2}$, ~2.58% enhancement) and $V_{oc}$ (from 1.03 V to 1.07 V, ~3.88% enhancement), relative to those of the PSCs with PCBM ETL. The change in $J_{sc}$ was verified by the comparison of the external quantum efficiency (EQE) for the devices with PCBM or C$_{60}$-3-BPy ETL (Figure 1C). The device with the C$_{60}$-3-BPy ETL shows a more predominant EQE response in the region of 580-700 nm and a more integrated $J_{sc}$ relative to that of the control device with PCBM ETL, indicating improved effective photon utilization after using the C$_{60}$-3-BPy ETL.

The current-voltage hysteresis of PSCs with PCBM ETL or C$_{60}$-3-BPy ETL was thereafter studied by the J-V curve measurements in different scan directions (Figure S3). The hysteresis index can be calculated from Equation 1 as:

$$\text{Hysteresis index} = \frac{\text{PCE (reverse)} - \text{PCE (forward)}}{\text{PCE (reverse)}}$$  (1)
The PSCs with C$_{60}$-3-BPy ETL presented a hysteresis index of 1.65%, which is lower than that (6.99%) of the control device with PCBM ETL (see Table S1), indicating negligible hysteresis for the PSCs with C$_{60}$-3-BPy ETL as well as a reduced trap state and charge accumulation in the perovskite layer.

Ultraviolet photoelectron spectroscopy (UPS) measurements were performed on the C$_{60}$-3-BPy/ITO and PCBM/ITO films to reveal the influence of these deposited fullerene derivatives ETLs on the performance of PSCs (Figure 2). The obtained work function (W$_{F}$) from the UPS was -4.14 eV and -4.09 eV for ITO/PCBM and ITO/C$_{60}$-3-BPy, respectively. The W$_{F}$ of the pristine ITO was -4.39 eV. Notably, the fullerene derivatives deposited on the ITO substrate significantly decreased the W$_{F}$ of pristine ITO. Moreover, C$_{60}$-3-BPy renders a stronger interaction with the ITO substrate relative to that of the PCBM, leading to a decreased energy offset between the W$_{F}$ of the ITO and the conduction band energy level (CB) of perovskite, suggesting that a lower energy loss is favorable for electron transport. Furthermore, the smaller energy offset between the ITO/C$_{60}$-3-BPy and the perovskite film is beneficial for the formation of an ohmic contact, facilitating electron transport from the perovskite film to the ITO electrode and resulting in increased $V_{OC}$.

The morphology of the ETLs has been unveiled to possess a considerable effect on the upper perovskite film quality. Therefore, the film morphology of ITO/PCBM and ITO/C$_{60}$-3-BPy was investigated by atomic force microscopy (AFM) (Figure S4), showing a smoother surface for the ITO/C$_{60}$-3-BPy with a root-mean-square of 0.92 nm while that of ITO/PCBM was 1.01 nm. The smoother surface of the ITO/C$_{60}$-3-BPy is beneficial for reduced interface traps and the growth of large-size grains of perovskite. Thereafter, the influence of the ITO/PCBM and ITO/C$_{60}$-3-BPy substrates on the morphology regulation of the CsFAMA perovskite layer was studied by scanning electron microscopy (SEM). Figure 3A,B show that the CsFAMA perovskite precursor deposited on the ITO/PCBM and ITO/C$_{60}$-3-BPy substrates delivers a compact surface perovskite film without obvious pinholes. According to the statistic grain-size distribution shown in Figure S5, the ITO/C$_{60}$-3-BPy/perovskite film possesses a grain size of 238 nm, which is larger than that of ITO/PCBM/perovskite with a value of 226 nm, suggesting better film quality of perovskite after the incorporation of the C$_{60}$-3-BPy ETL.

This hypothesis was verified by powder X-ray diffraction (XRD) and synchrotron-based grazing incidence X-ray diffraction (GIXRD) measurements. The XRD patterns of ITO/PCBM/perovskite and ITO/C$_{60}$-3-BPy/perovskite films show featured diffraction peaks at 14.2°, 28.5°, and 31.9° assigned to the (001), (002), and (012) diffraction planes of perovskite, respectively (Figure 3C,D). The perovskite film deposited on the ITO/C$_{60}$-3-BPy substrate exhibits enhanced diffraction intensity along with a drastically decreased PbI$_2$ diffraction peak at 12.8°, indicating the improved crystalline quality of the perovskite film. The 2D-GIXRD patterns of the ITO/PCBM/perovskite and ITO/C$_{60}$-3-BPy/perovskite films at an X-ray incident angle of 1.0° show scattered rings at q=10, 20, and 22.2 nm$^{-1}$ assigned to the (001), (002), and (012) diffraction planes of the perovskite crystals, respectively. The ITO/C$_{60}$-3-BPy/perovskite film exhibits brighter and
sharper scattered rings that the ITO/PCBM/perovskite film, suggesting improved film quality. Furthermore, the weaker scattered rings assigned to PbI₂ within the ITO/C₆₀-3-BPy/perovskite film suggests the effective suppression of PbI₂ harmful to device performance.\cite{29}

Steady-state photoluminescence (PL) measurements and time-resolved photoluminescence (TRPL) spectroscopy measurements were performed to unveil the charge transport property of the PSCs with PCBM or C₆₀-3-BPy ETLs, as shown in Figure 4A,B. The steady-state PL spectra of the ITO/PCBM/perovskite and ITO/C₆₀-3-BPy/perovskite films show the featured PL peak at 766.4 nm of CsFAMA perovskite under an excitation wavelength of 532 nm (Figure 4A). It is noteworthy that the C₆₀-3-BPy ETL sandwiched between the perovskite layer and the underneath ITO electrode results in
remarkable PL quenching relative to that of the counterpart device with PCBM ETL, indicating the more powerful electron extraction and transport capability of C$_{60}$-3-BPy ETL due to the special coordination interaction and the suitable energy level between C$_{60}$-3-BPy and the perovskite layer.$^{[30–32]}$ In addition, the PL peak of the ITO/C$_{60}$-3-BPy/perovskite film shows a blue-shift from 766 nm to approximately 757 nm relative to that of the ITO/PCBM/perovskite film, suggesting decreased trap states within perovskite with C$_{60}$-3-BPy ETL resulted from the improved perovskite crystallinity, the reduced grain boundaries, and Lewis acid-base reaction between the pyridine group and Pb$^{2+}$ of the perovskite.$^{[20,27,28,33–35]}$

The TRPL measurements were further used to study the charge transfer kinetics from the perovskite to the fullerene derivatives (Figure 4B and Table S2). The life of 21.12 ns for C$_{60}$-3-BPy/perovskite film is shorter than that of the PCBM/perovskite film (23.59 ns), suggesting the effective charge transfer from perovskite to C$_{60}$-3-BPy ETL.$^{[36]}$

Space charge limited current measurements were performed using electron-only devices to further quantify the trap state density of perovskite after the incorporation of PCBM or C$_{60}$-3-BPy ETLs, and the structure of ITO/PCBM or C$_{60}$-3-BPy ETL + perovskite/PCBM/Ag was analyzed.$^{[37,38]}$ The obtained J-V curves in the dark are depicted in Figure 4C. According to the bias voltage at the kink point, which is defined as the trap-filled limit voltage (V$_{TFL}$), the corresponding trap-state density ($n_t$) can be determined as 1.59 × 10$^{15}$ cm$^{-3}$ and 1.24 × 10$^{15}$ cm$^{-3}$ for the PSC device with PCBM and C$_{60}$-3-BPy ETL, respectively (Supporting Information S7). The decreased trap-state density of the PSC device with C$_{60}$-3-BPy ETL demonstrates that the C$_{60}$-3-BPy ETL enables a high-quality perovskite film and constructs an optimal ETL/perovskite interface. Such an improvement on the quality of perovskite film is expected to benefit the improvement of the ambient stability of the PSC devices.

Electrochemical impedance spectroscopy (EIS) measurements were performed to investigate the charge transport dynamics in the dark and a reverse potential of 1.0 V. The obtained Nyquist plots of the devices and the corresponding fitted curves through an equivalent circuit are shown in Figure 4D.$^{[39–41]}$ The fitted parameters from the Nyquist plots including the series resistance ($R_S$) and charge transfer resistance ($R_{ct}$) are summarized in Table S3. Moreover, it was determined that the constant phase element is related to the non-ideal chemical capacitance. The distinct discrepancy on $R_{ct}$ for the devices with PCBM ETL (1053.6 Ω·cm$^2$) and those with C$_{60}$-3-BPy
ETL (472.9 Ω cm\(^2\)) suggest that C\(_{60}\)-3-BPy ETL enables suppressed hole-electron recombination and facilitated charge transport.

3 | CONCLUSIONS

Regular n-i-p CsFAMA PSCs using C\(_{60}\)-3-BPy as an independent ETL afforded a decent PCE of 18.22%, which outperformed the control device with PCBM ETL (15.70%). C\(_{60}\)-3-BPy plays an important role in modifying the work function of the ITO electrode and providing a smooth surface, which is beneficial for obtaining high-quality perovskite films and the matched energy alignment of the device. Furthermore, the high electron affinity of C\(_{60}\)-3-BPy contributes to improved charge extraction. Moreover, the PSC with C\(_{60}\)-3-BPy ETL exhibits negligible current-voltage hysteresis. This study demonstrates the potential of fullerene derivatives in realizing large-area, low-temperature, and flexible PSC devices.

4 | EXPERIMENTAL SECTION

4.1 | Materials

The ITO-coated glass substrates with a sheet resistance of 10 Ω sq\(^{-1}\) were purchased from Shenzhen Nan Bo Group, China. The FTO-coated glass substrates with a sheet resistance of 13±1.5 Ω sq\(^{-1}\) were purchased from NSG Group, Japan. All the chemicals were used as received without further purification, including methylammonium bromide (MABr, Dyesol), formamidinium iodide (FAI, Dyesol), lead iodide (PbI\(_2\), Alfa Aesar), cesium iodide (CsI, Xi’an Polymer Light Technology Corp.), Spiro-OMeTAD (1 M company), Li-bis(trifluoromethanesulfonyl)imide lithium salt (Li-TFSI, 520 mg mL\(^{-1}\) in acetonitrile). The contact angles were measured using a CAM instrument (Data Physics, Germany). AFM measurements were performed on a XE-7 scanning probe microscope in noncontact mode (Park Systems, Korea). UPS measurements were carried out at the Catalysis and Surface Science station at the BL11U beamline in the National Synchrotron Radiation Laboratory. The GIXRD measurements were performed at the BL14B1 beamline of Shanghai Synchrotron Radiation Facility using X-ray with a wavelength of 1.38 Å. The XRD patterns were

4.2 | Device fabrication

The patterned ITO-coated glass was cleaned by sequential ultrasonic treatment in detergent, deionized water, acetone, and isopropanol for 15 minutes, and then treated with ultraviolet-ozone for 20 minutes. C\(_{60}\)-3-BPy was synthesized via a facile one-step Prato reaction at 80°C as reported previously.\(^{[26,27]}\) C\(_{60}\)-3-BPy was dissolved in chlorobenzene with concentrations of 5 mg ml\(^{-1}\) and spin-coated onto the ITO substrates and annealed at 100°C for 10 minutes. Next, CsFAMA perovskite precursor solution (1.3 M dissolved in DMSO and DMF with a volume ratio of 2:8, with a molar ratios of PbI\(_2\)/PbBr\(_2\) = 1:1.2, FAI: MABr = 1:0.2, CsI/(FAI+MABr) = 0.05:0.95, PbI\(_2\)/FAI = 1:1, PbBr\(_2\):MABr = 1:0.2) was spin-coated onto the substrates with a two-step procedure.\(^{[17]}\) The first step was 2000 rpm for 10 seconds with an acceleration of 200 rpm s\(^{-1}\). The second step was 6000 rpm for 30 seconds with a ramp-up of 2000 rpm s\(^{-1}\). At 15 seconds before the end of the second procedure, 100 µL chlorobenzene was dropped on the spinning substrate. The substrate was then immediately transferred on a hot plate and heated at 100°C for 60 minutes. After the perovskite films cooling down to room temperature, the HTL was deposited on top of the perovskite film by spin coating at 3000 rpm for 30 seconds using a chlorobenzene solution which contain 73.2 mg mL\(^{-1}\) of Spiro-OMeTAD and 28.8 µL mL\(^{-1}\) of tert-butylpyridine, as well as 18.8 µL mL\(^{-1}\) of bis(trifluoromethane)sulfonylimide lithium salt (Li-TFSI, 520 mg mL\(^{-1}\) in acetonitrile). Finally, the device was transferred into a vacuum chamber (10\(^{-6}\) torr), and an Au electrode (ca. 55 nm thick) was thermally deposited through a shadow mask to define the effective active area of the device (0.10 cm\(^2\)).

4.3 | Measurements and characterization

Current density-voltage (J-V) characterizations were conducted using a Keithley 2400 source measurement unit under simulated AM 1.5 irradiation (100 mW cm\(^{-2}\)) with a standard xenon lamp-based solar simulator (Oriel Sol 3A). The solar simulator illumination intensity was calibrated with a monocrystalline silicon reference cell (Oriel P/N 91150 V, with KG-5 visible color filter) calibrated by the National Renewable Energy Laboratory. EQE measurements were carried out on an Oriel Intelligent Quantum Efficiency 200TM measurement system which was equipped with a tunable light source. SEM images were obtained using a field emission scanning electron microscope (Zeiss Gemini SEM 500, Germany). The contact angles were measured using a CAM instrument (Data Physics, Germany). AFM measurements were performed on a XE-7 scanning probe microscope in noncontact mode (Park Systems, Korea). UPS measurements were carried out at the Catalysis and Surface Science station at the BL11U beamline in the National Synchrotron Radiation Laboratory. The GIXRD measurements were performed at the BL14B1 beamline of Shanghai Synchrotron Radiation Facility using X-ray with a wavelength of 1.38 Å. The 2D-GIXRD patterns were obtained by a MarCCD detector mounted vertically at a distance of around 274 mm from the sample with an exposure time of 50 seconds at a grazing incidence angle of 1°. The XRD patterns were
recorded on a Rigaku SmartLab X-ray diffractometer with Cu-Kα radiation (0.154 nm). The steady-state photoluminescence (PL) spectra were recorded using an Edinburgh Instruments FLS920 fluorescence spectrometer with an excitation wavelength of 460 nm. For TRPL spectra, the samples were excited by a 543 nm picosecond pulsed diode laser with a pulse width of 104 ps (Picoquant Gmbh Solea Supercontinuum laser using time-correlated single-photon counting method, and TimeHarp 260 software was used to record the decays.) Impedance spectroscopy (EIS) measurements were performed using an electrochemical workstation (Autolab 320, Metrohm, Switzerland) in a frequency range of 1 Hz to 1 MHz under 1.0 V in the dark. Alternating current 20 mV perturbation was applied with a frequency from 1 MHz to 1 Hz. The obtained impedance spectra were fitted with Z-View software.

ACKNOWLEDGMENTS
This work was supported by the National Key Research and Development Program of China (2017YFA0402800, 2019YFF0217202), and the National Natural Science Foundation of China (51925206, U1932214 and 51602097).

CONFLICT OF INTEREST
The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT
Research data are not shared.

ORCID
Shangfeng Yang https://orcid.org/0000-0002-6931-9613

REFERENCES
1. W. Yang, B.-W. Park, E. H. Jung, N. J. Jeon, Y. C. Kim, D. U. Lee, S. S. Shin, J. Seo, E. K. Kim, J. H. Noh, S. I. Seok, Science 2017, 356, 1376.
2. W. Yang, J. H. Noh, N. J. Jeon, Y. C. Kim, S. Ryu, J. Seo, S. I. Seok, Science 2015, 348, 1234.
3. Z. Xiao, C. Bi, Y. Shao, Q. Dong, Q. Wang, Y. Yuan, C. Wang, Y. Gao, J. Huang, Energy Environ. Sci. 2014, 7, 2619.
4. W.-Q. Wu, D. Chen, R. A. Caruso, Y.-B. Cheng, J. Mater. Chem. A 2017, 5, 10092.
5. T. Liu, K. Chen, Q. Hu, R. Zhu, Q. Gong, Adv. Energy Mater. 2016, 6, 1600457.
6. L. Meng, J. You, T. Guo, Y. Yang, Acc. Chem. Res. 2016, 49, 155.
7. https://www.nrel.gov/pv/assets/pdfs/best-research-cell-efficiencies.20200925.pdf
8. Y. Rong, Y. Hu, A. Mei, H. Tan, M. I. Saimadinov, S. I. Seok, M. D. McGehee, E. H. Sargent, H. Han, Science 2018, 361, 6408.
9. J.-P. Correa-Baena, A. Abate, M. Saliba, W. Tress, T. Jesper Jacobsen, M. Grätzel, A. Hagfeldt, Energy Environ. Sci. 2017, 10, 710.
10. Y. Li, J. K. Cooper, W. Liu, C. M. Sutter-Fella, M. Amani, J. W. Beeman, A. Javey, J. W. Ager, Y. Liu, F. M. Toma, I. D. Sharp, Nat. Commun. 2016, 7, 12446.
11. H. Zhou, Q. Chen, G. Li, S. Luo, T.-h. Song, H.-S. Duan, Z. Hong, J. You, Y. Liu, Y. Yang, Science 2014, 345, 542.
12. J. Wang, J. M. Ball, E. M. Barea, A. Abate, J. A. Alexander-Webber, J. Huang, M. Saliba, I. Mora-Sero, J. Bisquert, H. J. Snaith, R. J. Nicholas, Nano Lett. 2014, 14, 724.
13. D. Liu, T. L. Kelly, Nat. Photonics 2013, 8, 133.
14. C. Yang, M. Yu, D. Chen, Y. Zhou, W. Wang, Y. Li, T. C. Lee, D. Yun, Chem. Commun. 2017, 53, 10882.
15. F. Cai, L. Yang, Y. Yan, J. Zhang, F. Qin, D. Liu, Y.-B. Cheng, Y. Zhou, T. Wang, J. Mater. Chem. A 2017, 5, 9402.
16. M. Zhang, T. Li, G. Zheng, L. Li, M. Qin, S. Zhang, H. Zhou, X. Zhan, Mater. Chem. Front. 2017, 1, 2078.
17. W. Hu, W. Zhou, X. Lei, P. Zhou, M. Zhang, T. Chen, H. Zeng, J. Zhu, S. Dai, S. Yang, S. Yang, Adv. Mater. 2019, 31, 1806095.
18. C. Cui, Y. Li, Y. Li, Adv. Energy Mater. 2017, 7, 1601251.
19. L.-L. Deng, S.-Y. Xie, F. Gao, Adv. Electron. Mater. 2018, 4, 1700435.
20. L. Jia, M. Chen, S. Yang, Mater. Chem. Front. 2020, 4, 2256-2282.
21. S. Ryu, J. Seo, S. S. Shin, Y. C. Kim, N. J. Jeon, J. H. Noh, S. I. Seok, J. Mater. Chem. A 2015, 3, 3271.
22. K. Wojciechowski, I. Ramirez, T. Gorisse, O. Dautel, R. Dasari, N. Sakai, J. M. Hardigree, S. Song, S. Marder, M. Riede, G. Wantz, H. J. Snaith, ACS Energy Lett. 2016, 1, 648.
23. Z. Wang, D. P. McMeekin, N. Sakai, S. Reenen, K. Wojciechowski, J. B. Patel, M. B. Johnston, H. J. Snaith, Adv. Mater. 2017, 29, 1604186.
24. Y.-C. Wang, X. Li, L. Zhu, X. Liu, W. Zhang, J. Fang, Adv. Energy Mater. 2017, 7, 1701144.
25. B. L. Watson, N. Rolston, K. A. Bush, T. Leijtens, M. D. McGehee, R. H. Dauskardt, ACS Appl. Mater. Interfaces. 2016, 8, 25896.
26. H. Liu, S. Li, L. Deng, Z. Wang, Z. Xing, X. Rong, H. Tian, X. Li, S. Xie, R. Huang, L. Zheng, ACS Appl. Mater. Interfaces. 2019, 11, 23982.
27. B. Li, J. Zhen, Y. Wan, X. Lei, Q. Liu, Y. Liu, L. Jia, X. Wu, H. Zeng, W. Zhang, G.-W. Wang, M. Chen, S. Yang, ACS Appl. Mater. Interfaces. 2018, 10, 32471.
28. B. Li, J. Zhen, Y. Wan, X. Lei, L. Jia, X. Wu, H. Zeng, W. Zhang, G.-W. Wang, M. Chen, S. Yang, ACS Appl. Mater. Interfaces. 2016, 8, 14503.
29. S. Feng, Y. Yang, M. Li, J. Wang, Z. Cheng, J. Li, G. Ji, G. Yin, F. Song, Z. Wang, J. Li, X. Gao, ACS Appl. Mater. Interfaces. 2016, 8, 14503.
30. N. K. Noel, A. Abate, S. D. Stranks, E. S. Parrott, V. M. Burlakov, A. Goriely, H. J. Snaith, ACS Nano 2014, 8, 9815.
31. L. Zuo, Q. Chen, N. D. Marco, Y.-T. Hsieh, H. Chen, O. Sun, S. Chang, H. Zhao, S. Dong, Y. Yang, Nano Lett. 2017, 17, 269.
32. N. Ahn, D.-Y. Son, I.-H. Jang, J. Am. Chem. Soc. 2015, 137, 8696.
33. S. Shao, Z. Xiao, C. Bi, Y. Yuan, J. Huang, Nat. Commun. 2014, 5, 5784.
34. D. Yang, X. Zhou, R. Yang, Z. Yang, W. Yu, X. Wang, C. Li, F. Liu, R. P. H. Chang, Energy Environ. Sci. 2016, 9, 3071.
35. D. Yang, R. Yang, X. Ren, X. Zhu, Z. Yang, C. Li, S. Liu, Adv. Mater. 2016, 28, 5206.
36. W. Chen, J. Zhang, G. Xu, R. Xue, Y. Li, Y. Zhou, J. Hou, Y. Li, Adv. Mater. 2018, 30, 1800855.
37. J. Jiang, Q. Wang, Z. Jin, Adv. Energy Mater. 2018, 8, 1701757.
38. T. Niu, J. Lu, R. Munir, J. Li, D. Barrat, X. Zhang, H. Hu, Z. Yang, A. Amassian, K. Zhao, S. Liu, Adv. Mater. 2018, 30, 1706576.
39. J. Zhen, W. Zhou, M. Chen, B. Li, L. Jia, M. Wang, S. Yang, J. Mater. Chem. A 2019, 7, 2754.
40. Y. Lin, L. Shen, J. Dai, Y. Deng, Y. Wu, Y. Bai, X. Zheng, J. Wang, Y. Fang, H. Wei, W. Ma, X. Zeng, X. Zhan, J. Huang, Adv. Mater. 2017, 29, 1604545.
41. J. Jung, C.-C. Chueh, A. K. Y. Jen, Adv. Energy Mater. 2015, 5, 1500486.

**SUPPORTING INFORMATION**
Additional supporting information may be found online in the Supporting Information section at the end of the article.

**How to cite this article:** Y. Wang, B. Li, L. Jia, B. Zhang, H. Zhang, P. Liang, M. Chen, H. Yang, X. Wang, S. Yang. Nano Select 2021, 1. https://doi.org/10.1002/nano.202100050