Supplementary Information

Low-Cost Coenzyme Q10 as Efficient Electron Transport Layer for Inverted Perovskite Solar Cells

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Experimental details

1) PL measurement

The steady PL spectra and TRPL decay measurements were performed using an Flou Time 300(PicoQuant). The TRPL and steady-state PL measurements were conducted using prepared samples: glass/perovskite or glass/perovskite/Q10 or FTO/perovskite/$PC_{61}BM$. The excitation wavelength of 560nm for PL measurement and 809nm for TRPL measurement. The PL measurements were conducted in the wavelength range from 500 to 900 nm. The PL decay constants were calculated using an equation:

$$y = A_1 \exp(-\frac{t}{\tau_1}) + A_2 \exp(-\frac{t}{\tau_2}) + y_0 \quad (1)$$

where, A is the decay amplitude, and τ is the decay time.

2) Device characterization

The *J-V* characteristics were measured by Keithley 2400 source and the solar simulator with standard AM 1.5G (100 $mW.cm^{-2}$) under ambient conditions. The *J-V* curves of all devices were measured by reverse (1.1 V to -0.1 V) or forward (-0.1 V to 1.1 V) with 0.028 $V\ s^{-1}$ of scan rate. The active area of device is 0.096 cm^2. The conductivity of ETLs was calculated by following equation:

$$\sigma = \frac{Id}{VA} \quad (2)$$

where, A is the active area (0.096 cm^2), d is the thickness of the samples, V is the apply bias, and I is real-time current.

3) SCLC measurement

The device structure of FTO/perovskite/ETL/Ag was measured using CHI 660E electrochemical workstation (Ch Instrument Company) (0 V to 3V) to evaluate the trap densities of devices. The trap densities were calculated using the following equation:

$$V_{TFL} = \frac{qn_t d^2}{2\varepsilon\varepsilon_0} \quad (3)$$

where V_{TFL} is the trap-filled limit voltage, q is the elementary charge, n_t is the trap density, d is the film thickness, ε is the dielectric constant, and ε_0 is the vacuum

permittivity.

4) Mott–Schottky measurement

The device structure is FTO/perovskite/ETL/Ag, using CHI 660E electrochemical workstation (Ch Instrument Company) (0 V to 1V). Doping density was calculated using the Mott–Schottky equation, as follows:

$$\frac{1}{C^2} = \frac{2}{\varepsilon\varepsilon_0 q A^2 N}(V_{bi} - V) \tag{4}$$

$$W_p = \left[\frac{2\varepsilon\varepsilon_0}{qN}(V - V_{bi})\right]^{0.5} \tag{5}$$

where C is the capacitance, ε is dielectric constant 46.9, ε_0 is the permittivity of free space, q is the elementary charge, A is the active area (0.096 cm^2), N is the doping density of the semiconductor, V is the applied bias, and W_p is depletion zone width.

5) DFT calculation details

The cutoff energy of plane-wave in these calculations is set to 500 eV. The calculated supercell combines coenzyme Q10 or $PC_{61}BM$ and a $FAPbI_3$ supercell of 1 × 8 × 8 in the unit of its primitive cell. The Brillouin zone was sampled using Γ point only, which is decided by considering the large size of this supercell and our limited computational resources. The charge transfer happens at the interface between coenzyme Q10 or $PC_{61}BM$ and $FAPbI_3$. The charge density difference plotting in Figure.3(a) and (c) shows that there are significant charge transfers at their interfaces. The charge transfers mainly take place across the interfaces.

To get quantity insights of the charge transfers at the interface, Bader charge analysis code[1-4] have been used to quantify the charge variations of all atoms which are labeled on the atomic structure plots of Figure. 3(b) and (d).

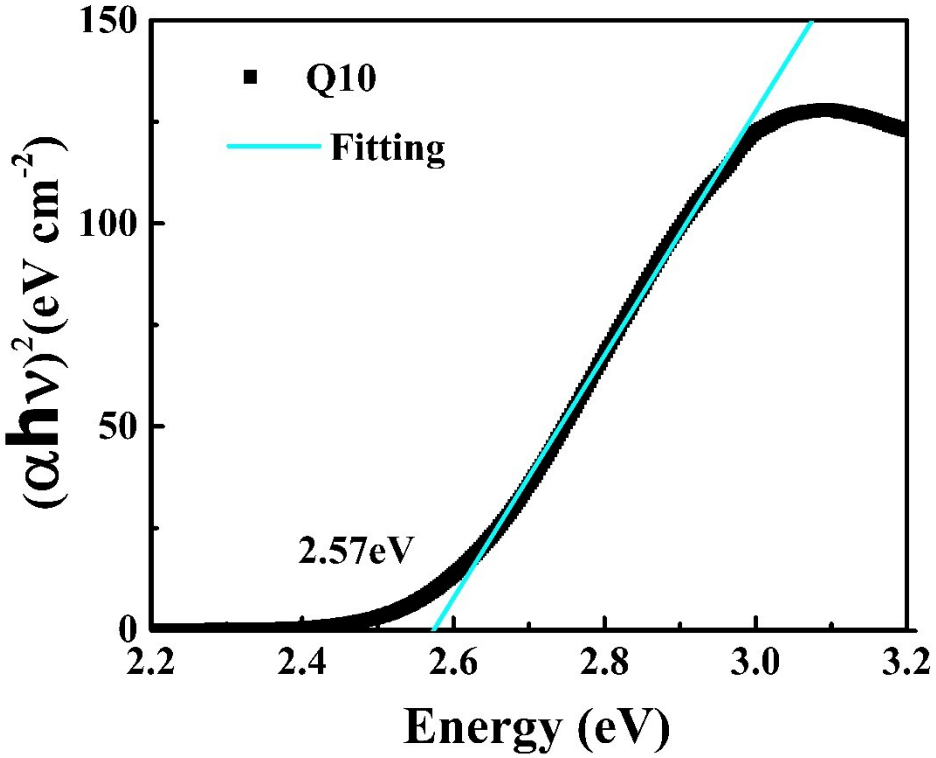

Fig. S1 Absorbance spectra of Q10.

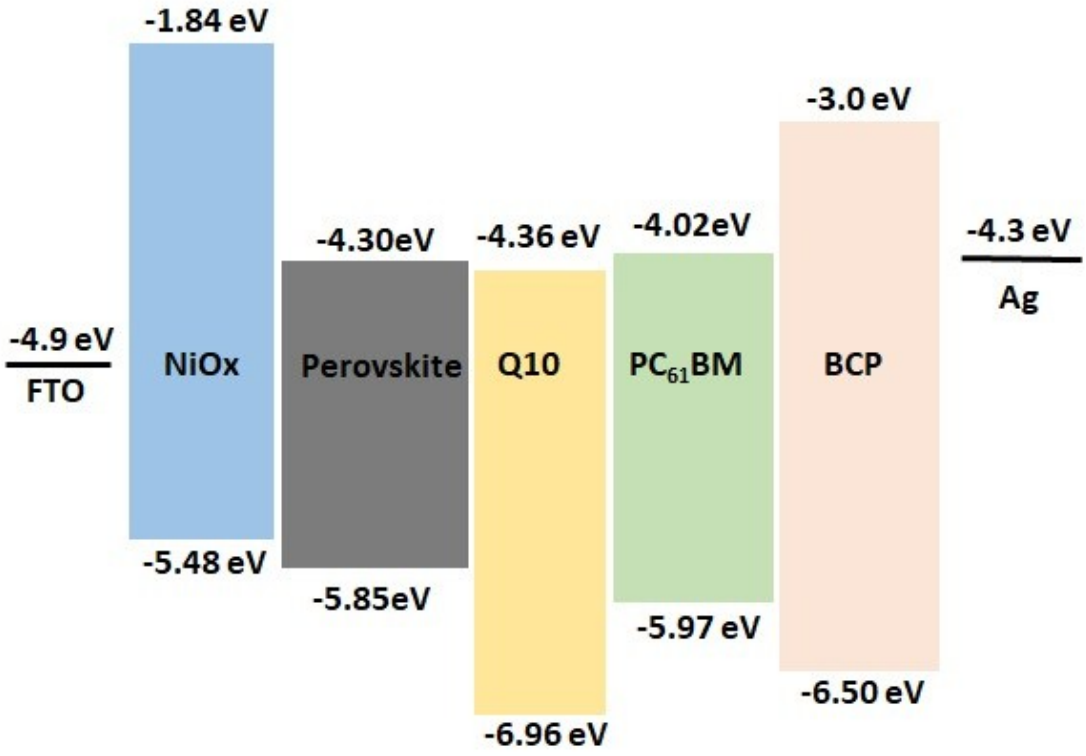

Fig. S2 The energy diagram of the PSC with Q10 and $PC_{61}BM$ ETLs.

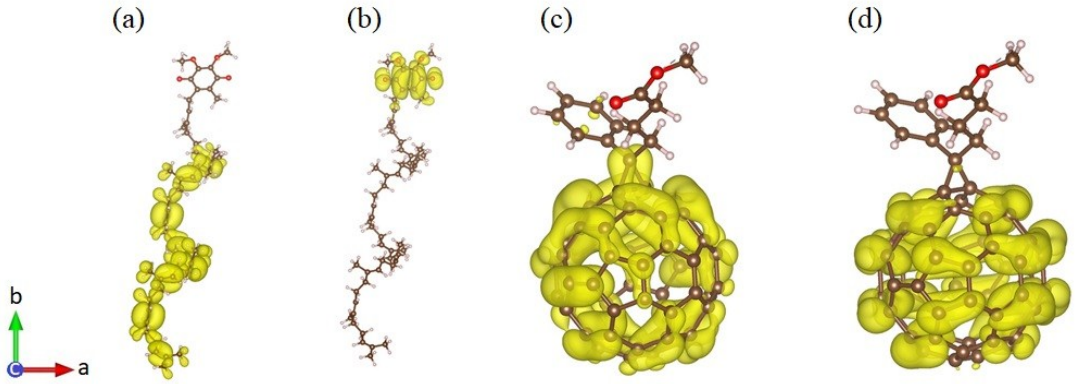

Fig. S3 The charge distribution of HOMO (a) and LUMO (b) of Q10, HOMO (c) and LUMO (d) of $PC_{61}BM$.

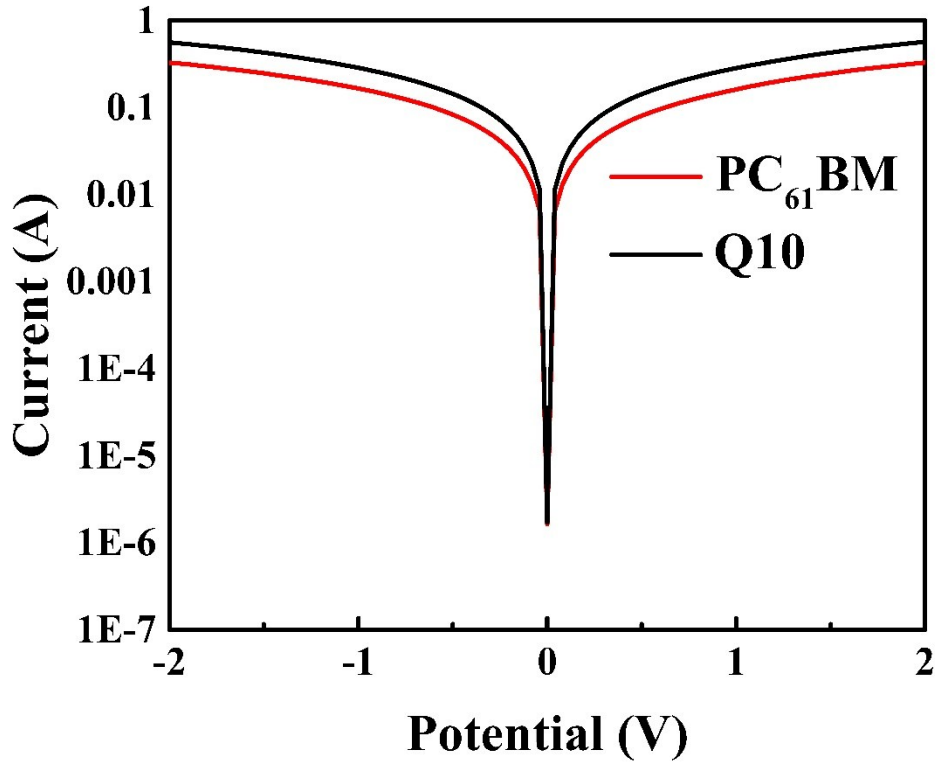

Fig. S4 *I-V* curves of conductivity measurements of Q10 and $PC_{61}BM$ under dark conditions.

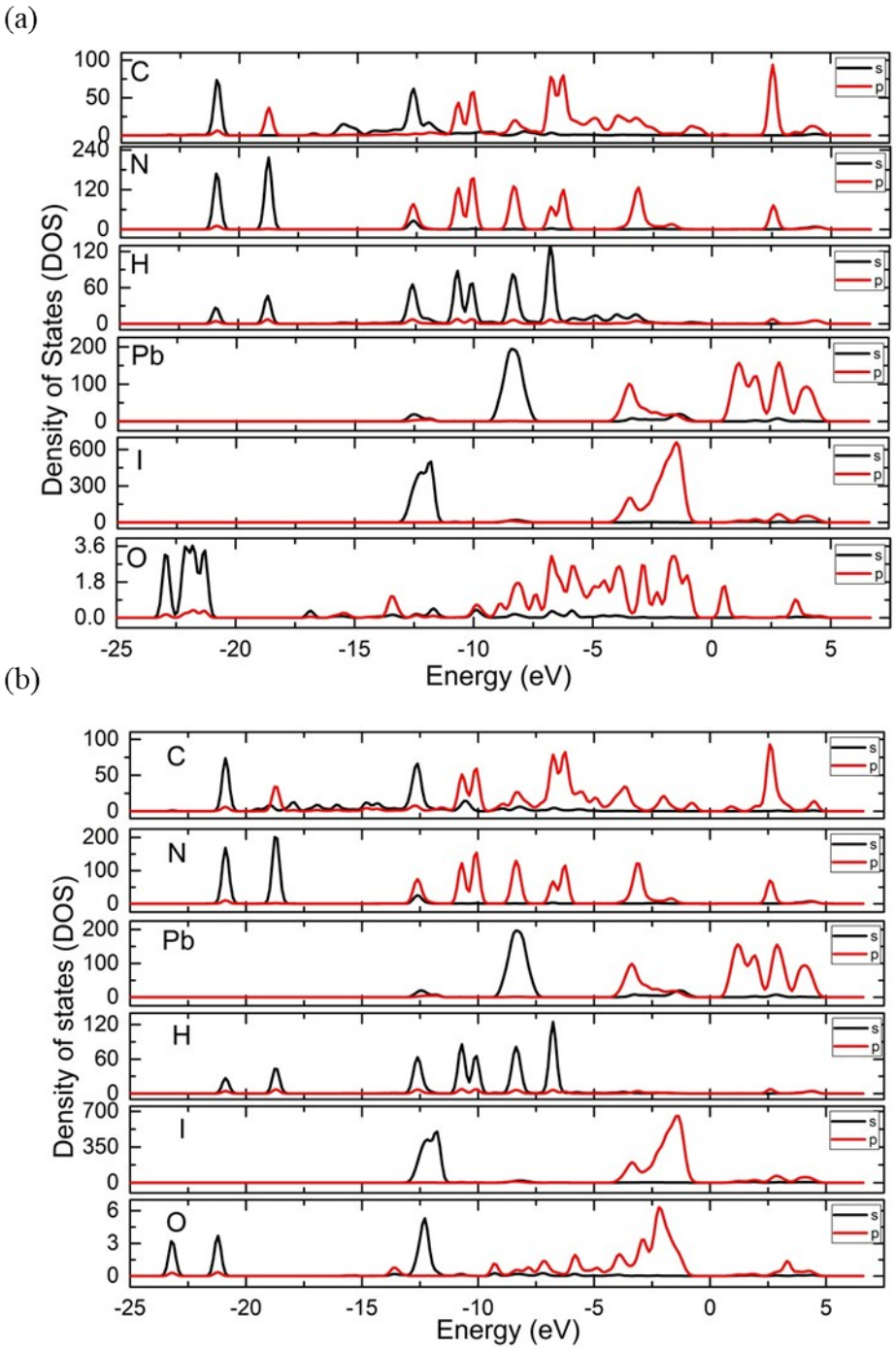

Fig. S5 Partial density of states for (a) Q10/$FAPbI_3$ and (b) $PC_{61}BM/FAPbI_3$.

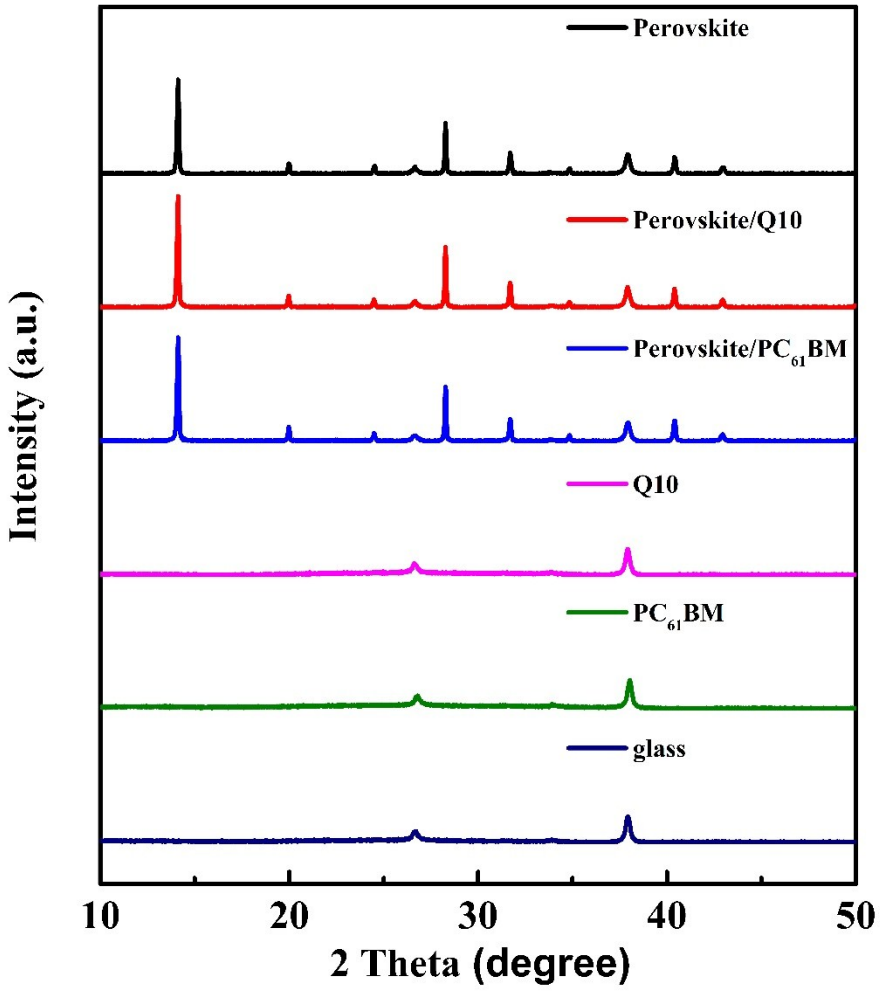

Fig. S6 XRD pattern of FTO, $PC_{61}BM$, Q10, $FAMAPbI_3$.

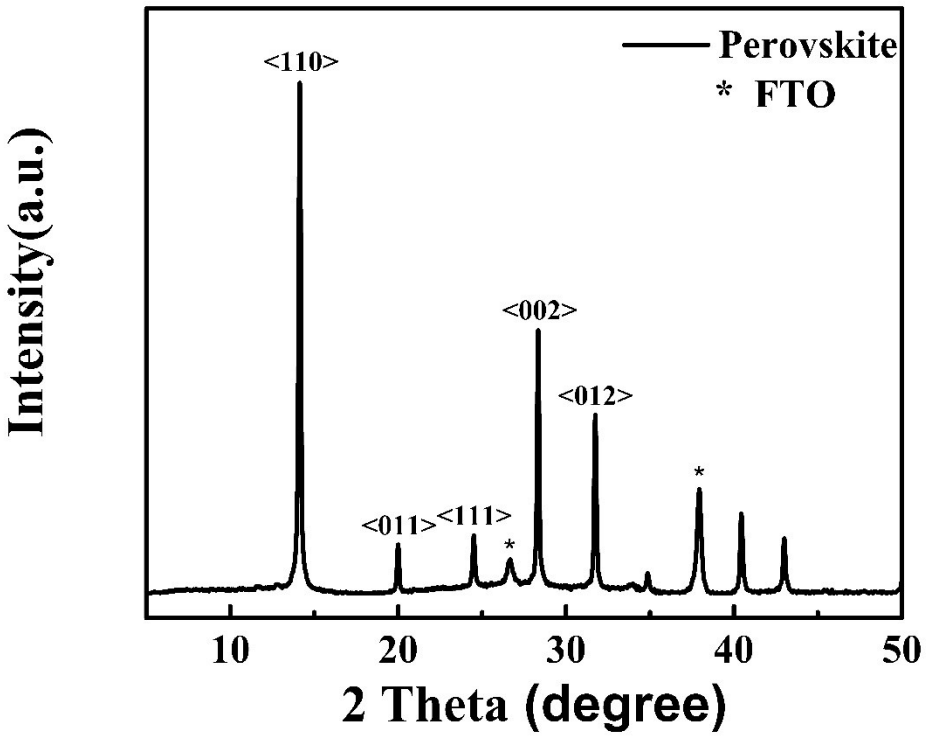

Fig. S7 XRD pattern of $FAMAPbI_3$.

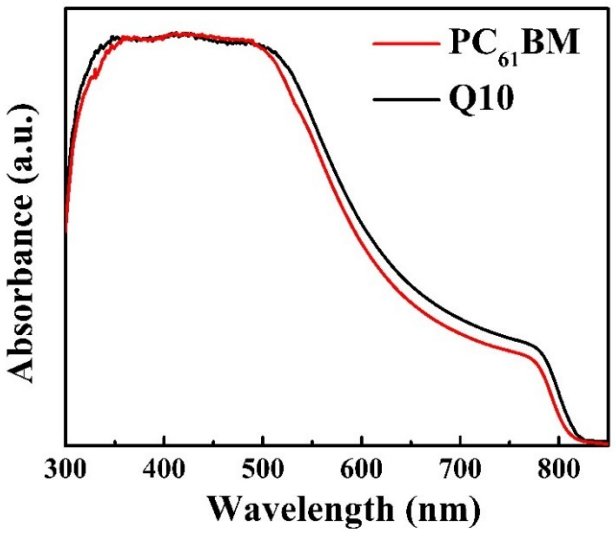

Fig. S8 UV-vis absorption spectra of FTO/NiO_x/$FAMAPbI_3$/ETL/BCP.

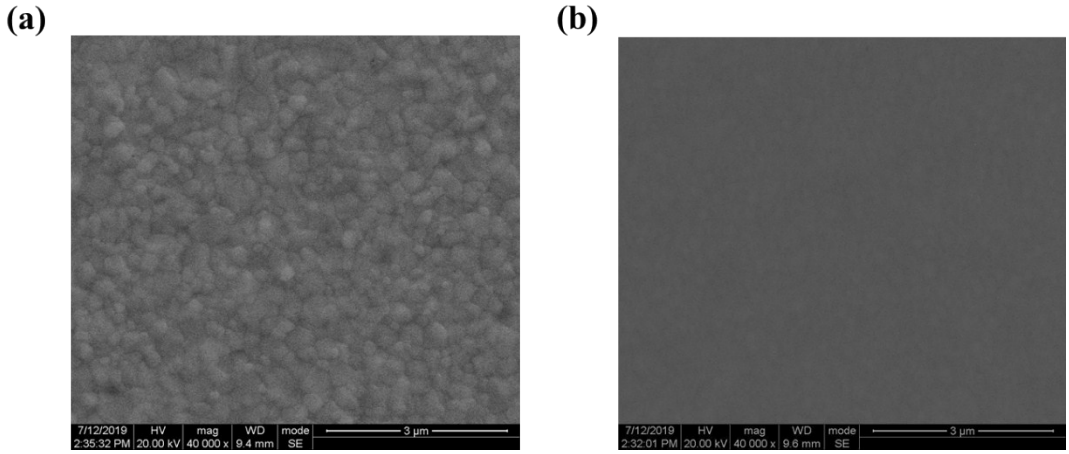

Fig. S9 Top-view SEM images of perovskite (a) and Q10 covered perovskite (b), the concentration of Q10 solution is 7mg/mL in CB.

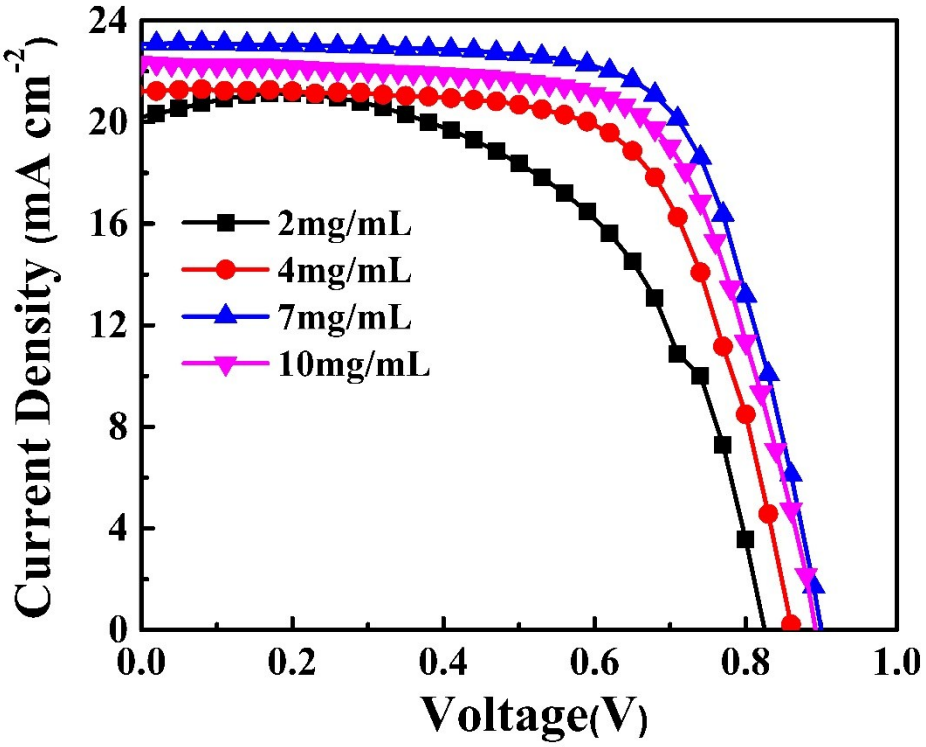

Fig. S10 *J-V* curves for PSCs with different Q10 concentration.

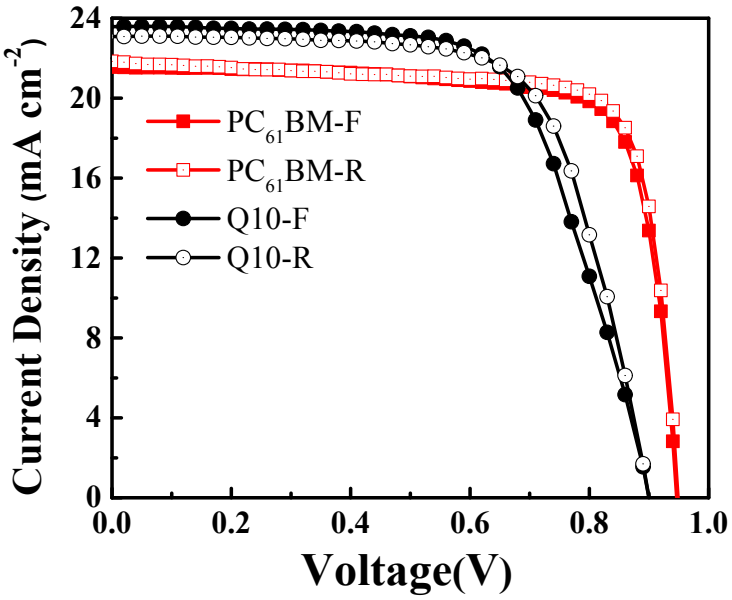

Fig. S11 *J-V* curves of the champion Q10-based (black) and $PC_{61}BM$-based (red) device, F means forward scan and R means reverse scan.

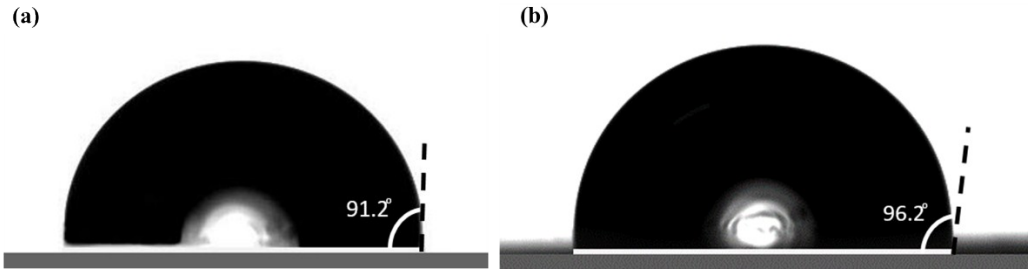

Fig. S12 The water contact angle of $PC_{61}BM$ (a) and Q10 (b) deposited on a perovskite film surface.

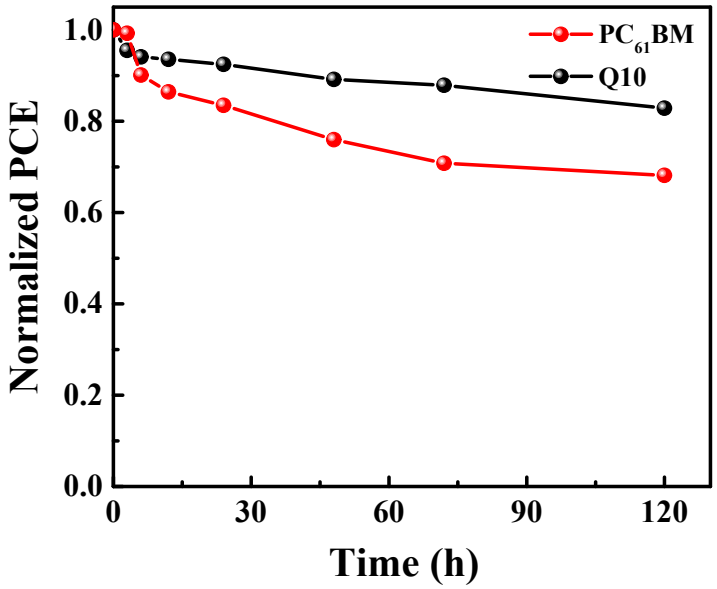

Fig. S13 Stability test of the unencapsulatd devices in ambient air with Q10 or $PC_{61}BM$ as ETL (stored under air condition of 30 C, 25% relative humidity).

Table S1. Calculated conductivities of each ETL.

ETL	Slope	Thickness (nm)	Conductivity (mS cm^{-1})
Q10	2975.97	122.64	3.650E-02
$PC_{61}BM$	1720.70	96.64	1.663E-02

Table S2. Summery electron lifetimes of each structure.

structure	A_1(%)	τ_1 (ns)	A_2(%)	τ_2 (ns)	τ_{av}(ns)
Perovskite	23.62	6.44	76.38	51.17	40.60
Perovskite/Q10	57.42	4.06	42.58	29.61	14.94
Perovskite/$PC_{61}BM$	43.10	3.26	56.90	21.18	13.46

Table S3. V_{TFL} and n_t trap densities of perovskite/ETL interface.

ETL	d (nm)	V_{TFL} (V)	n_t （cm^{-3}）
Q10	482.5	0.953	2.12 E+16
$PC_{61}BM$	469.5	0.963	2.26 E+16

Table S4. Slope of the Mott–Schottky plots and doping densities of ETLs.

	slope	N_d（cm^{-3}）	V_{bi}（V）	W_p(nm)
Q10	-2.658E+08	1.227E+20	0.082	1.861
$PC_{61}BM$	-6.597E+08	4.944E+19	0.119	3.532

Table S5. Detailed photovoltaic performance of different density of Q10-based PSCs.

	J_{sc} (mA cm^{-2})	V_{oc} (V)	FF (%)	PCE (%)
2mg/mL	20.22	0.82	58.36	9.72
4mg/ mL	21.26	0.86	67.15	12.26
7mg/ mL	23.11	0.90	68.94	14.34
10mg/ mL	22.30	0.89	67.39	13.42

References:

1. G. Henkelman, A. Arnaldsson and H. Jónsson, *Comput. Mater. Sci.*, 2006, **36**, 354-360.
2. E. Sanville, S. D. Kenny, R. Smith and G. Henkelman, *J. Comput. Chem.*, 2007, **28**, 899-908.
3. W. Tang, E. Sanville and G. Henkelman, *J. Phys.: Condens. Matter*, 2009, **21**, 084204.
4. M. Yu and D. R. Trinkle, *J. Chem. Phys.*, 2011, **134**, 064111.