Supporting Information

Multifunctional Organic Ammonium Dopants to Electron Transport Layer for Efficient Inverted Perovskite Solar Cells

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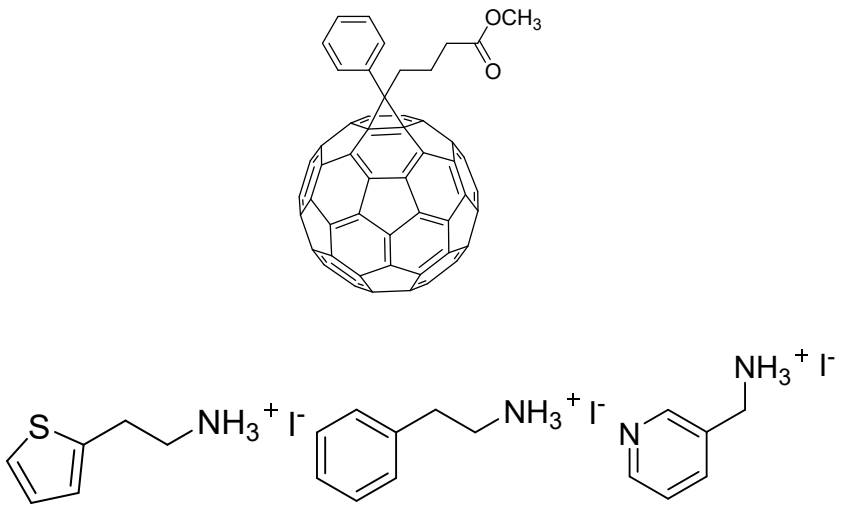

Fig. S1. Molecular structure of $PC_{61}BM$, TEAI, PEAI and PyAI.

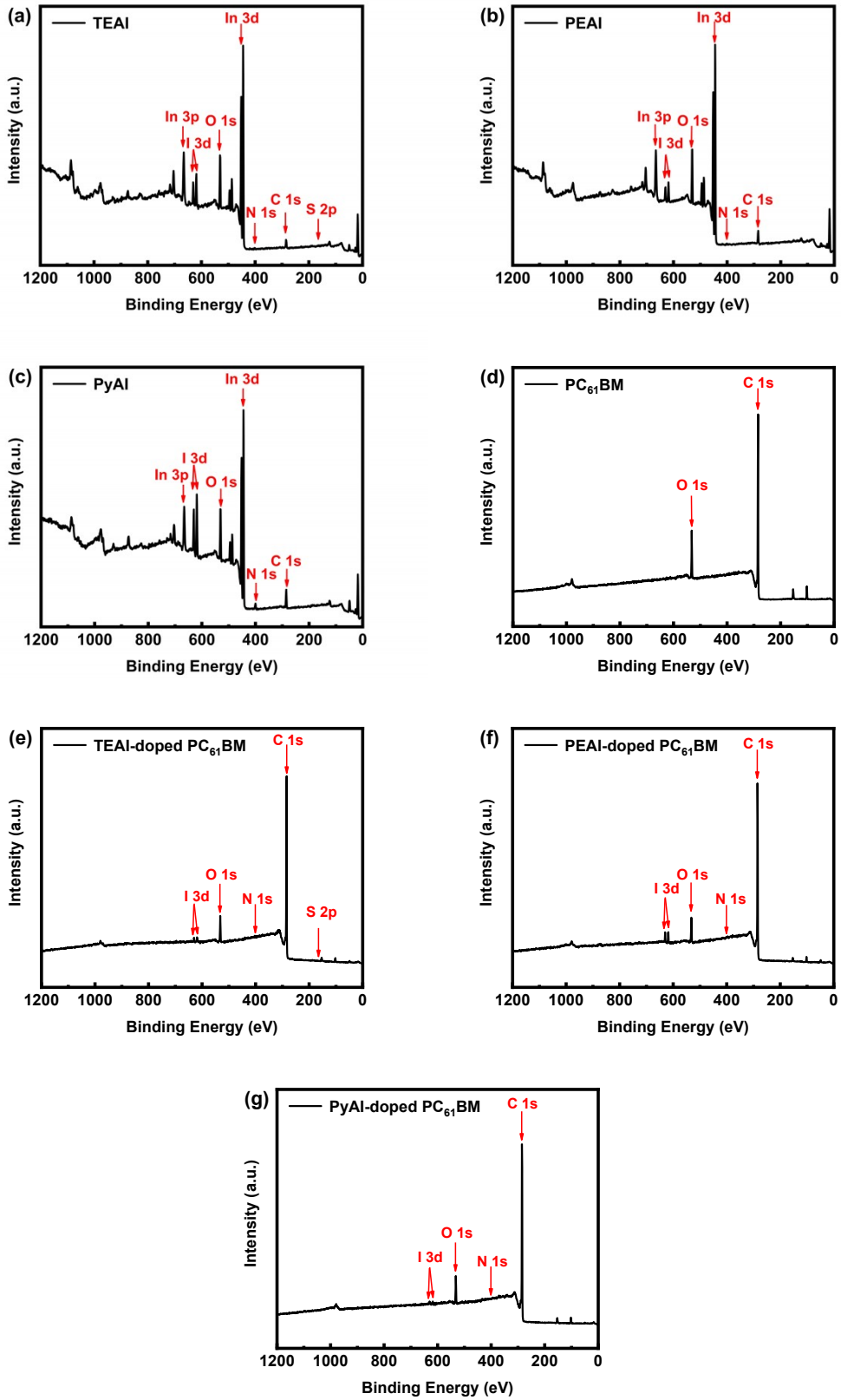

Fig. S2. XPS spectra of pristine TEAI, PEAI, PyAI, pristine $PC_{61}BM$ and doped $PC_{61}BM$.

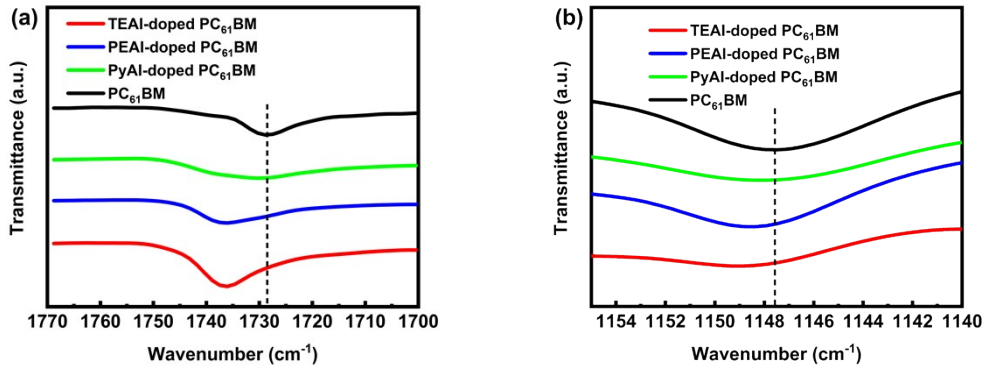

Fig. S3. FTIR spectra of C=O bonds (a) and C-O-C bonds (b) of different ETMs.

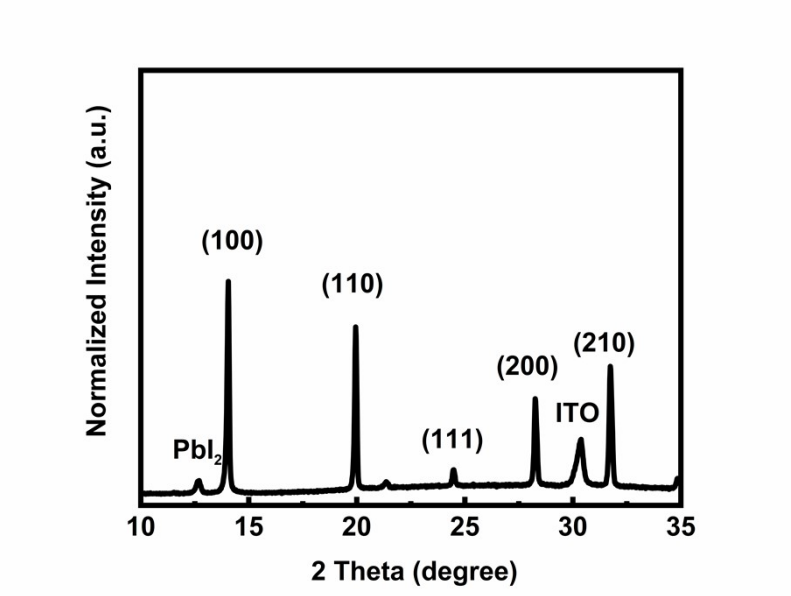

Fig S4. XRD spectra of the perovskite film.

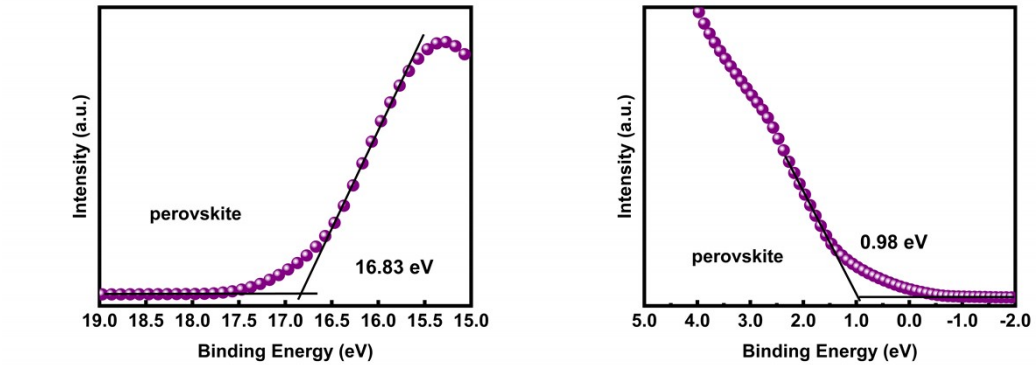

Fig. S5. UPS plots of pure perovskite.

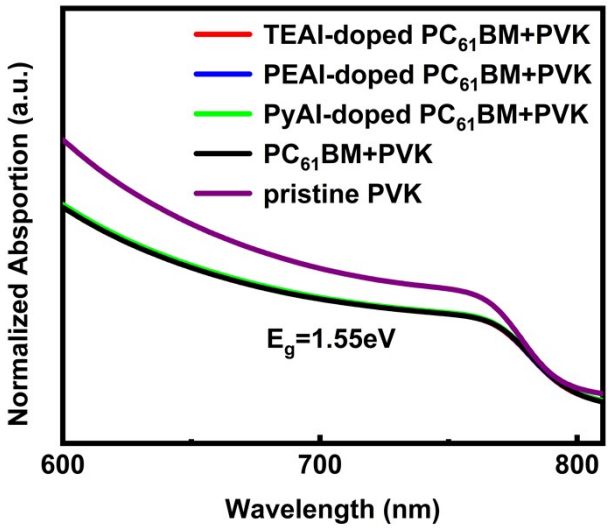

Fig. S6. UV-visible absorption spectra of pristine perovskite and perovskite film with doped $PC_{61}BM$ and pristine $PC_{61}BM$.

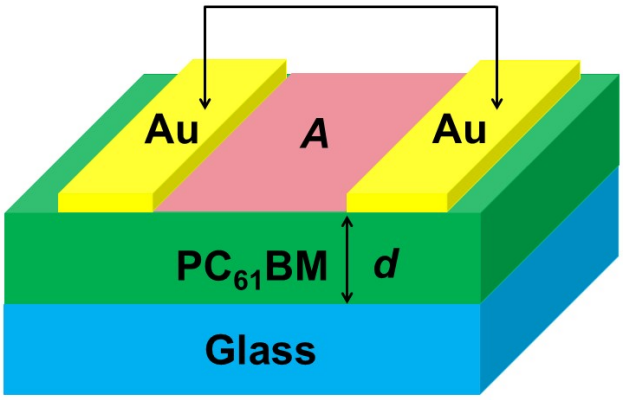

Fig. S7. Device structure and test method of electrical conductivity measurements. Electrical conductivity (σ_0) is defined as the formula, $\sigma_0=I\cdot d/(A\cdot V)$. A is the active area between two gold electrodes, d is the thickness of $PC_{61}BM$ layer, I and V are the test parameters during the electrical conductivity measurements.

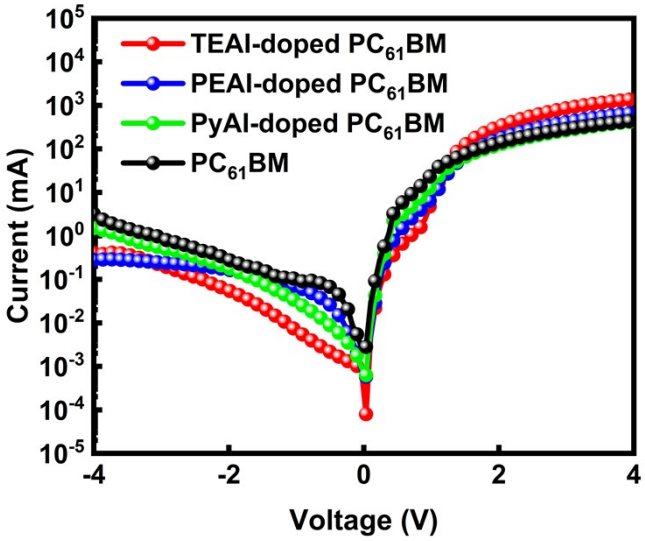

Fig. S8. *J-V* curves of the devices with pristine $PC_{61}BM$ and ammonium-doped $PC_{61}BM$.

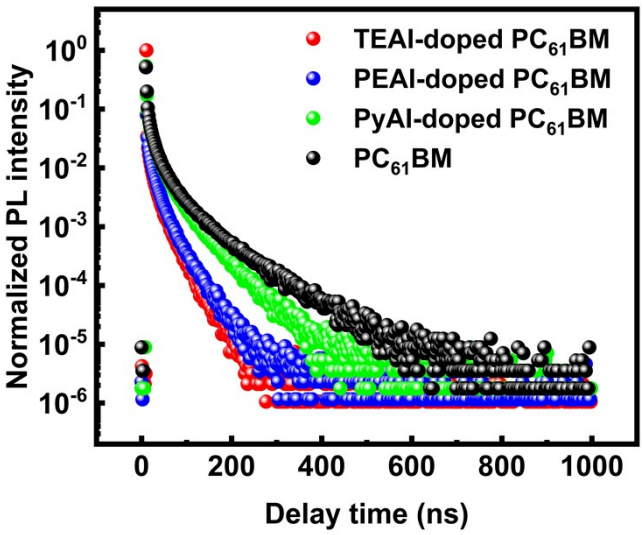

Fig. S9. TRPL spectra of different ETMs spin-coated on the top of perovskite.

We utilized the function, $y=y_0 + A_1e^{-x/t_1} + A_2e^{-x/t_2} + A_3e^{-x/t_3}$ to fit the TRPL spectra. The average decay time (t_{ave}) was calculated by the formula, $t_{ave}= (A_1t_1^2 + A_2t_2^2 + A_3t_3^2)/(A_1t_1 + A_2t_2 + A_3t_3)$. Detailed parameters were shown in Table S3.

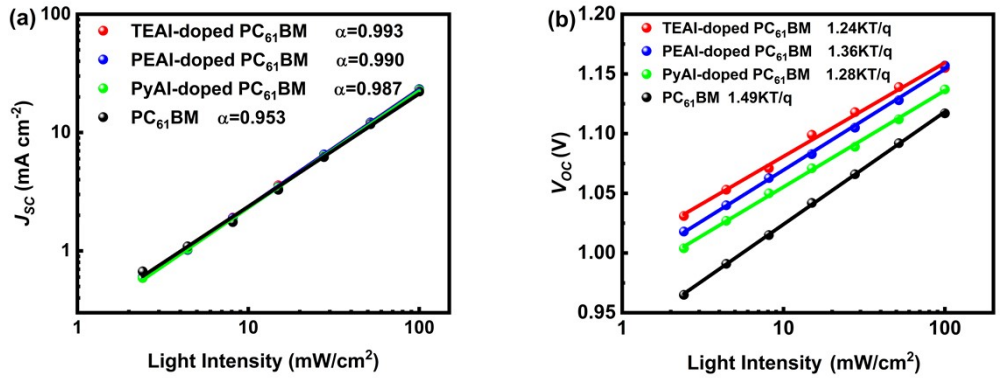

Fig. S10. The dependence of J_{SC} (a) and V_{OC} (b) on the light intensity for devices with different $PC_{61}BM$.

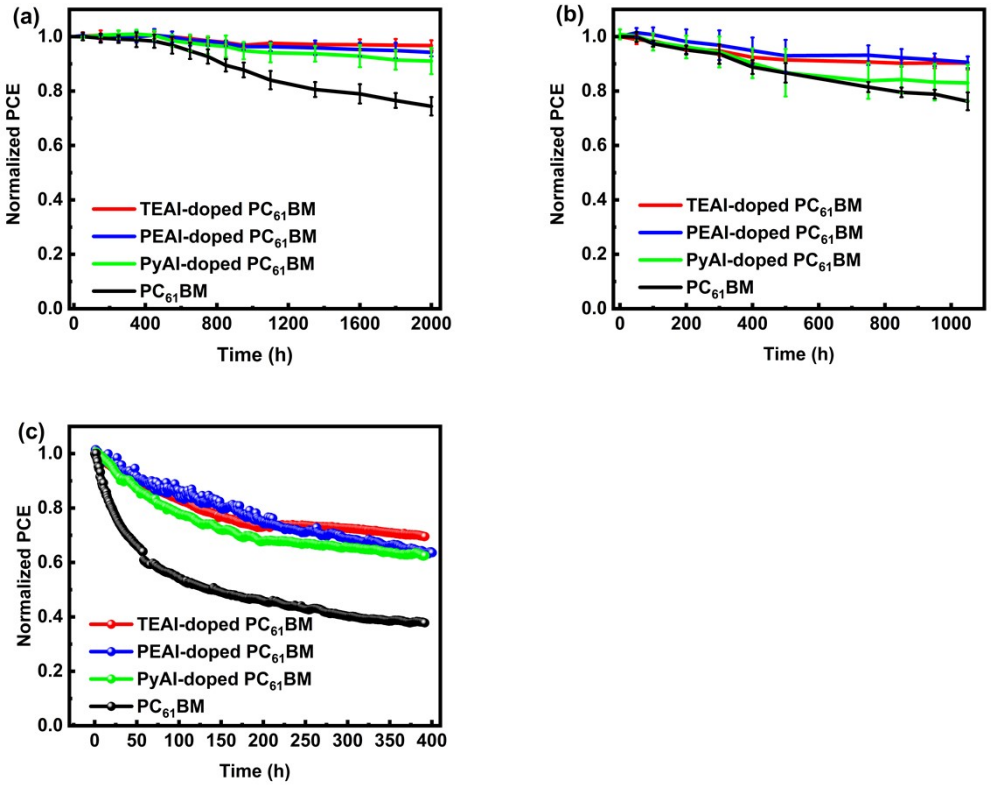

Fig. S11. (a) Storage stability of devices in N_2 glovebox under room temperature. (ISOS-D-1I) (b) Thermal stability of devices in N_2 glovebox under 85 °C. (ISOS-D-2I) (c) MPP tracking test of devices in N_2 glovebox under room temperature with white LED, whose light intensity (100 mW/cm^2) is automatically calibrated with a Si reference diode. (ISOS-L-1I)

Table S1. UPS data of perovskite, doped $PC_{61}BM$ and pristine $PC_{61}BM$.

Materials	E_{sec} (eV)	E_f (eV)	ϕ_{hole} (eV)	HOMO (eV)	LUMO (eV)
perovskite	16.83	-4.39	0.98	-5.37	-3.82
TEAI-doped $PC_{61}BM$	16.69	-4.53	1.35	-5.88	-3.88
PEAI-doped $PC_{61}BM$	16.63	-4.59	1.32	-5.91	-3.91
PyAI-doped $PC_{61}BM$	16.64	-4.58	1.32	-5.90	-3.90
$PC_{61}BM$	16.60	-4.62	1.38	-6.00	-4.00

Table S2. Devices parameters of TEAI-doped, PEAI-doped, PyAI-doped and control devices.

ETLs		V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
$PC_{61}BM$		1.13 (1.12±0.01)	23.43 (23.37±0.55)	78.6 (77.5±1.4)	20.78 (20.39±0.39)
TEAI-doped $PC_{61}BM$	0.05 mg/mL	1.14 (1.13±0.02)	24.81 (24.18±0.66)	77.8 (77.2±2.5)	21.99 (21.15±0.84)
	0.1 mg/mL	1.15 (1.15±0.02)	24.94 (24.51±0.34)	80.6 (79.7±2.3)	23.17 (22.80±0.47)
	0.2 mg/mL	1.16 (1.16±0.01)	24.23 (24.27±0.72)	80.1 (79.7±2.1)	22.57 (22.39±0.66)
	0.5 mg/mL	1.16 (1.16±0.02)	23.05 (23.07±0.74)	80.4 (78.9±2.0)	21.58 (21.04±1.24)
PEAI-doped $PC_{61}BM$	0.05 mg/mL	1.14 (1.14±0.01)	23.14 (22.90±0.24)	81.0 (78.7±2.3)	21.48 (20.61±0.87)
	0.1 mg/mL	1.15 (1.15±0.01)	24.26 (24.12±0.35)	80.8 (80.2±1.8)	22.61 (22.34±0.44)
	0.2 mg/mL	1.15 (1.15±0.02)	23.70 (23.23±0.76)	79.1 (79.2±1.7)	21.62 (21.25±0.37)
	0.5 mg/mL	1.15 (1.16±0.01)	22.44 (22.83±1.18)	80.3 (78.0±2.3)	20.98 (20.55±0.57)
PyAI-doped $PC_{61}BM$	0.05 mg/mL	1.13 (1.13±0.01)	23.93 (23.63±0.66)	78.7 (77.5±3.0)	21.32 (20.73±0.59)
	0.1 mg/mL	1.15 (1.15±0.01)	24.19 (23.78±0.41)	80.1 (80.5±1.1)	22.31 (22.06±0.25)
	0.2 mg/mL	1.15 (1.15±0.01)	22.96 (23.13±1.17)	80.9 (79.2±2.5)	21.42 (20.99±0.45)
	0.5 mg/mL	1.15 (1.16±0.01)	23.31 (23.04±0.34)	79.6 (79.1±1.3)	21.36 (20.95±0.44)

Table S3. Parameters of TRPL fitting line.

ETM	t_1 (ns)	t_2 (ns)	t_3 (ns)	A_1	A_2	A_3	t_{ave} (ns)
TEAI-doped $PC_{61}BM$	0.73	8.12	28.50	156734	22862	5455	13.24
PEAI-doped $PC_{61}BM$	0.81	9.29	32.34	136085	25150	5129	14.97
PyAI-doped $PC_{61}BM$	1.59	13.00	46.69	95255	39038	7523	23.01
$PC_{61}BM$	2.18	15.20	69.46	104576	37647	4813	28.58