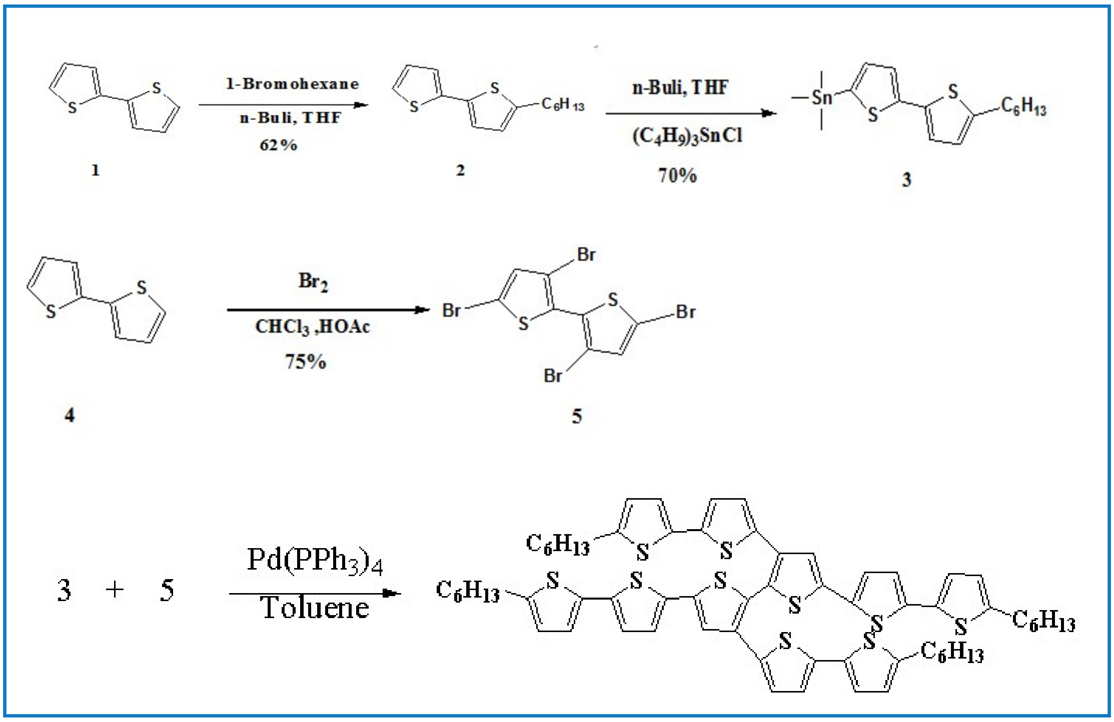
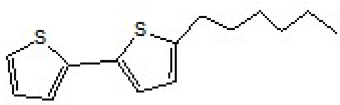
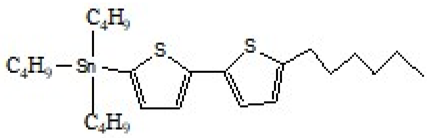
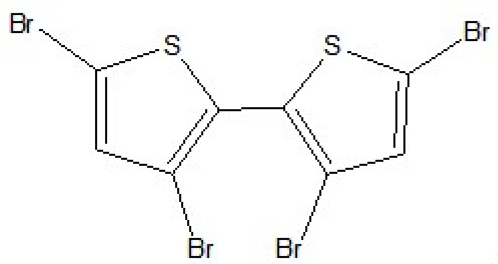




1HNMR (500MHZ, CDCl3, δ): 7.31-7.34 (m, 1H), 7.28-7.29 (m, 1H), 7.10-7.19 (m, 2H), 6.82-6.84 (d, 1H), 2.95 (t, 2H), 1.87 (m, 2H), 1.49-1.59 (m, 6H), 1.12 (t, 3H). 13CNMR: 145.40, 138.21, 135.01, 127.84, 124.90, 123.80, 123.53, 123.11, 31.87, 31.83, 30.39, 29.08, 22.90, 14.40.



1HNMR (500MHZ, CDCl3, δ): 7.31 (d, 1H), 7.13 (d, 1H), 7.07 (d, 1H), 6.75 (d, 1H), 2.87 (t, 2H), 1.65-1.80 (m, 10H), 1.40-1.49 (m, 14H), 1.21 (t, 4H), 1.01 (m, 10H). 13CNMR: 144.95. 144.44, 136.07, 135.67, 135.13, 124.73, 124.25, 123.10, 31.71, 31.70, 30.27, 29.15, 29.07, 28.98, 28.89, 27.62, 27.39, 27.16, 22.72, 14.22, 13.80, 12.34, 12.28, 10.94, 10.92, 9.57.



1HNMR (500MHZ, CDCl3, δ): 6.97 (s, 2H). 13CNMR: 132.99, 129.54, 114.84, 112.21.

BT-BTH

1HNMR (500MHZ, CDCl3, δ): 7.36 (s, 2H), 7.10 (d, 2H), 7.01 (m, 4H), 6.94 (d, 2H), 6.87 (m, 4H), 6.70 (d, 2H), 6.61 (d, 2H), 2.72-2.82 (m, 8H), 1.61-1.71 (m, 12H), 1.28-1.33 (m, 20H), 0.85-0.92 (m, 12H). 13CNMR:145.95, 145.49, 138.99, 138.04, 137.70, 136.55, 136.07, 135.30, 134.31, 130.96, 126.17, 125.10, 124.93, 124.83, 124.73, 123.65, 123.60, 123.47, 123.24, 123.19, 31.62, 30.19, 29.76, 28.83, 22.7, 14.14.

Fig. S1. Synthesis routes of BT-BTH and 1H NMR, 13C NMR spectra.

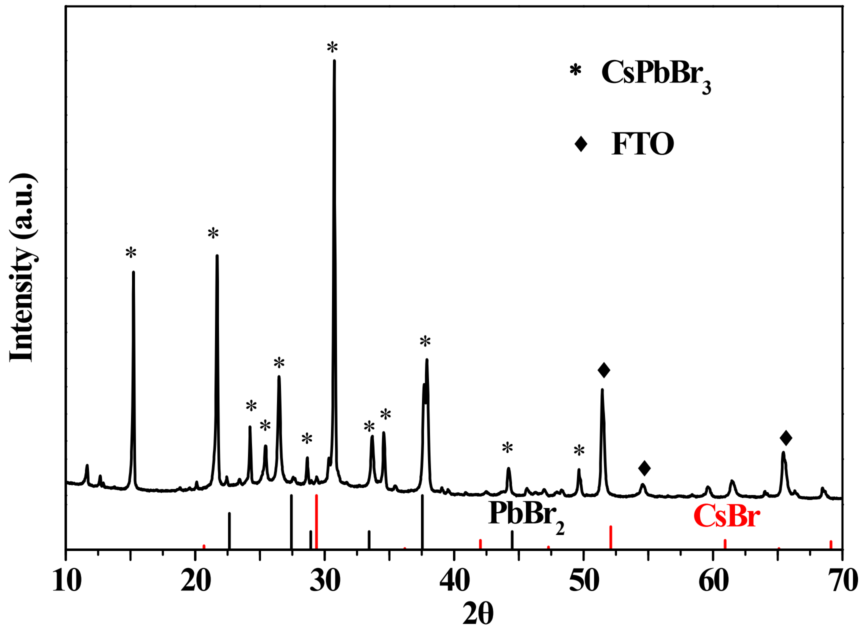


Fig. S2. The X-ray diffraction (XRD) pattern of $CsPbBr_3$ film in FTO glass.

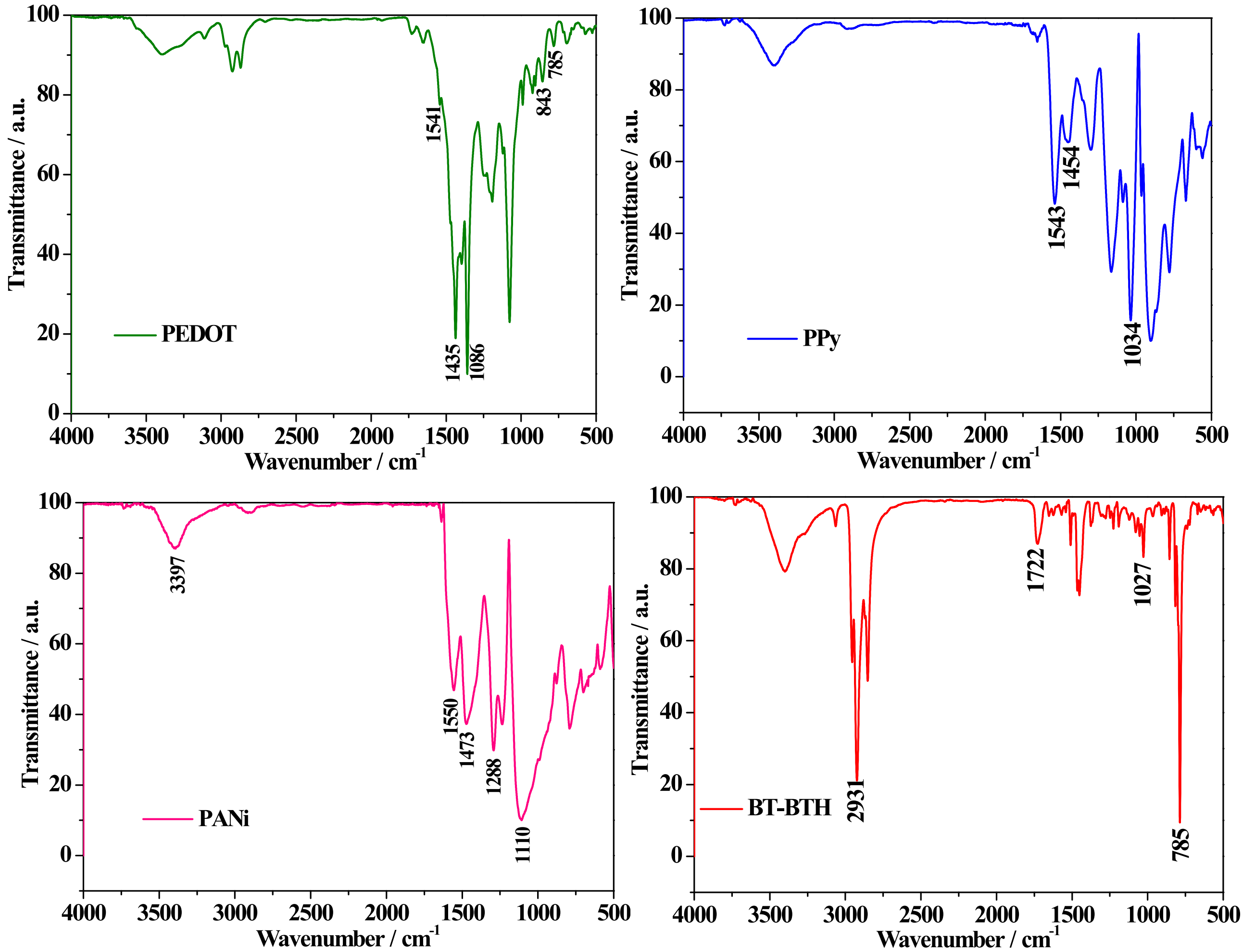


Fig. S3. FTIR spectra of BT-BTH, PEDOT, PPy and PANi.

FTIR absorption spectrum for the polymers and small-molecule BT-BTH are shown in Fig. S3. The bands around 1435 cm^{-1} are typical of the C=C symmetric stretching vibrations of thiophene ring and the appearance of peaks at around 785 cm^{-1} is C–H out of plane vibration. In addition to, the presence of characteristic absorption bands at 1555 cm^{-1} (C=C stretching vibration of the quinoid ring), 1476 cm^{-1} (stretching vibration of C=C of the benzenoid ring) and 1288 cm^{-1} (C-N stretching vibration). The bands in the range of 3000-3500 cm^{-1} correspond to secondary amine stretching (N-H) vibrations.

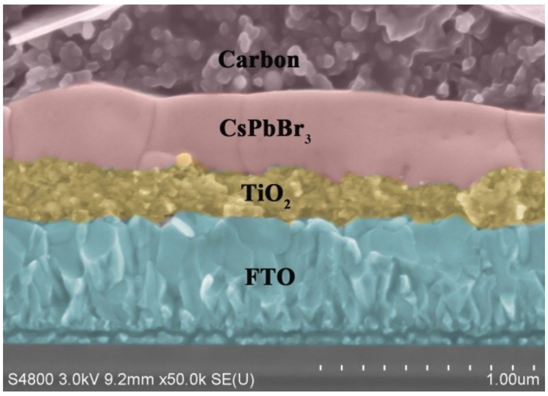


Fig. S4. Cross-sectional SEM image of a PSC device.

Fig. S4 shows the cross-sectional SEM image of a PSC device with $FTO/TiO_2/CsPbBr_3$/HTM/carbon structure.

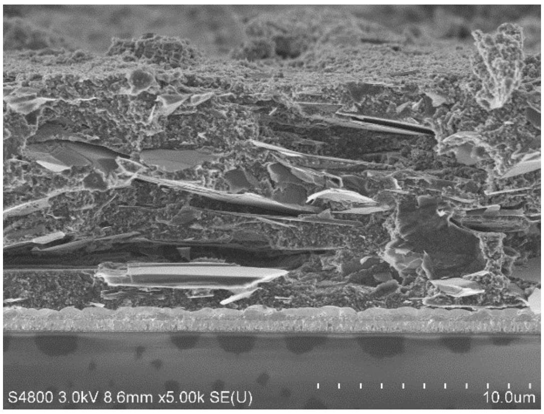


Fig. S5. The magnified SEM image of a total PSC device.

The thicknesses of carbon electrode is about 15 um.

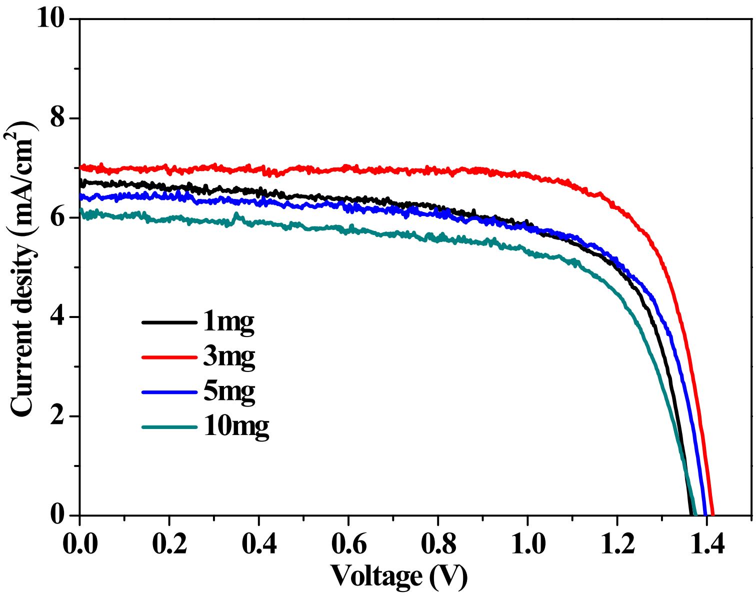


Fig. S6. Photo *J-V* curves of inorganic $CsPbBr_3$ PSCs by coating BT-BTH solutions with different concentrations.

Fig. S6 shows the *J-V* curves of inorganic $CsPbBr_3$ PSCs. It can be concluded the device has excellent performance by coating the BT-BTH solution with a concentration of 3 mg/mL.

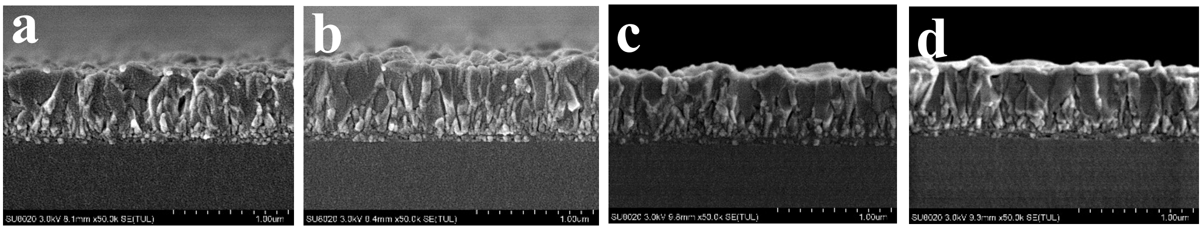


Fig. S7. The cross-sectional images of FTO/BT-BTH, and the concentrations of BT-BTH solutions are (a) 1 mg/mL, (b) 3 mg/mL, (c) 5 mg/mL, (d) 10 mg/mL,.

The cross-sectional views of BT-BTH films by coating the solutions with concentrations of 1, 3, 5 and 10 mg/mL, as shown in Fig. S7, suggesting average thicknesses of 50, 70, 90 and 120 nm, respectively.

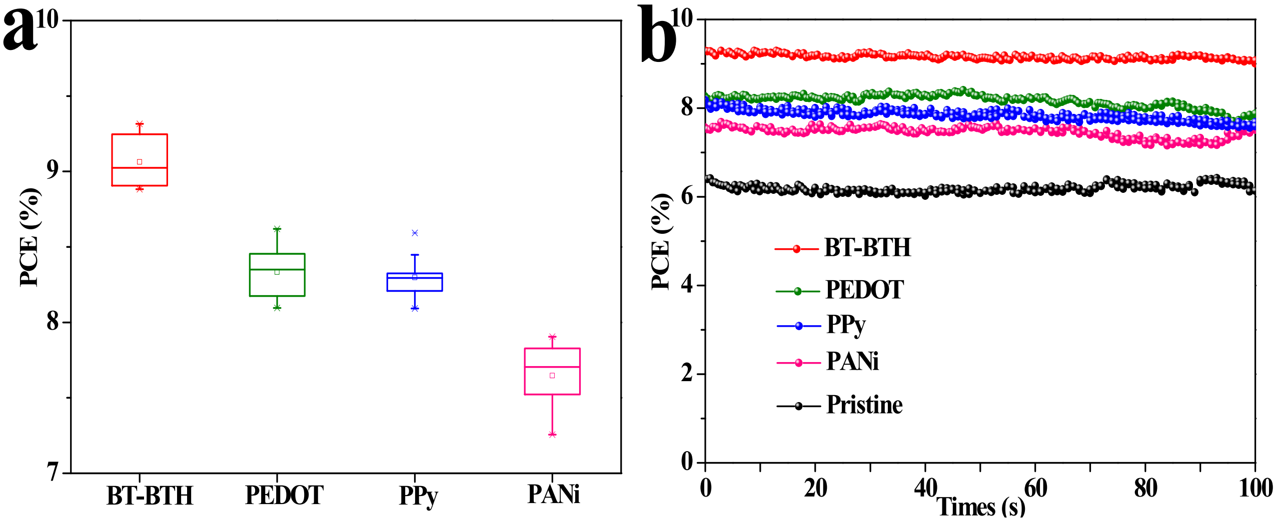


Fig. S8. (a) The PCE distribution of 10 random devices for each device with different HTMs. (b) Steady-state power outputs at the maximum power point for HTM-free and HTM tailored PSCs.

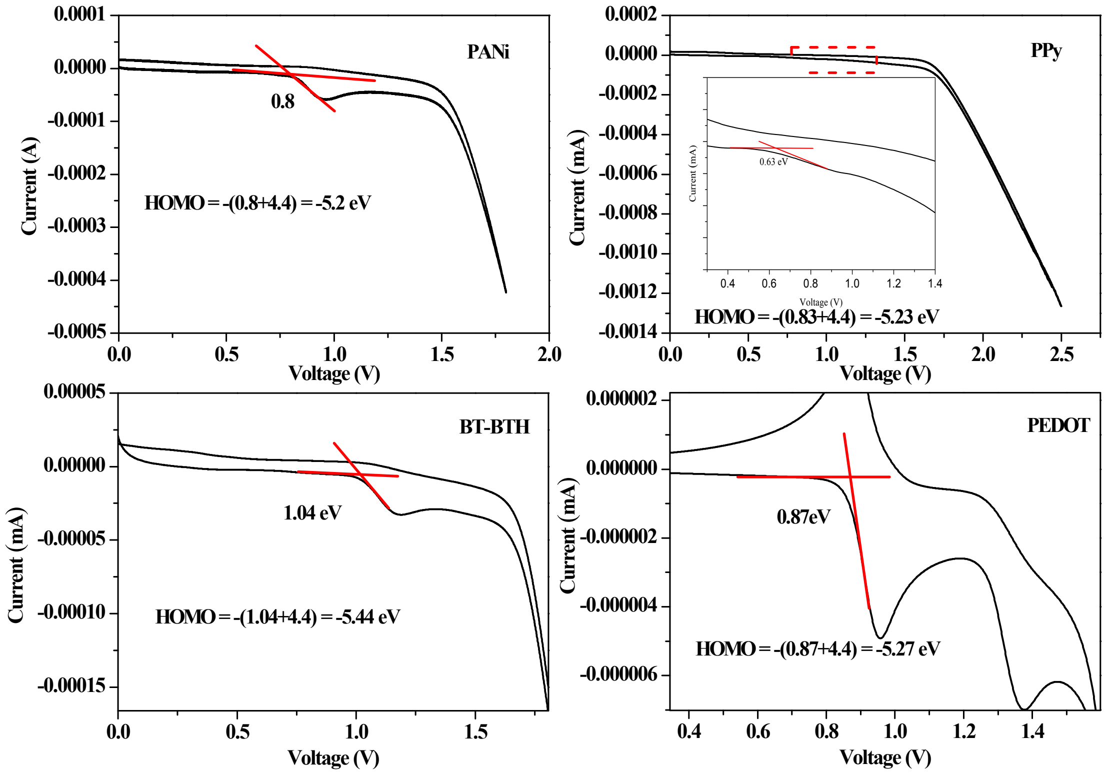


Fig. S9. CV curves of different HTMs.

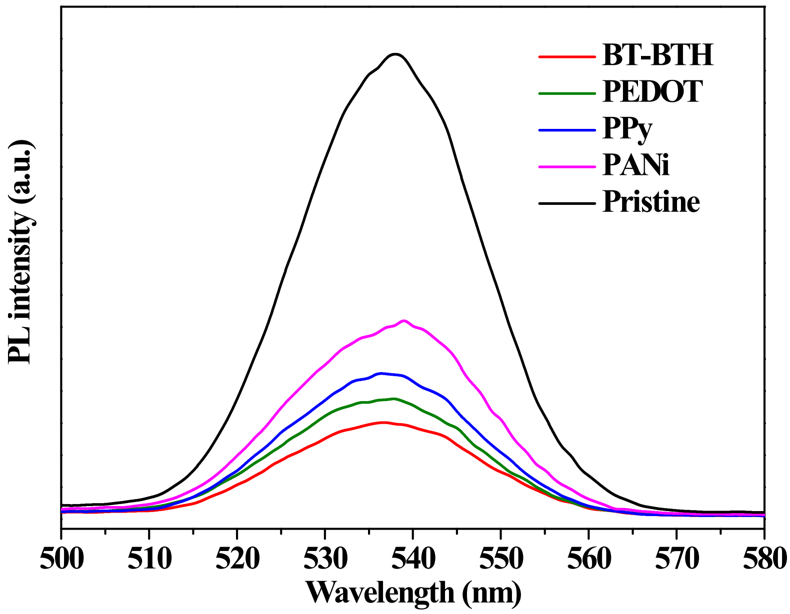


Fig. S10. The steady-state PL spectra of pristine device and BT-BTH, PEDOT, PPy, PANi based devices.

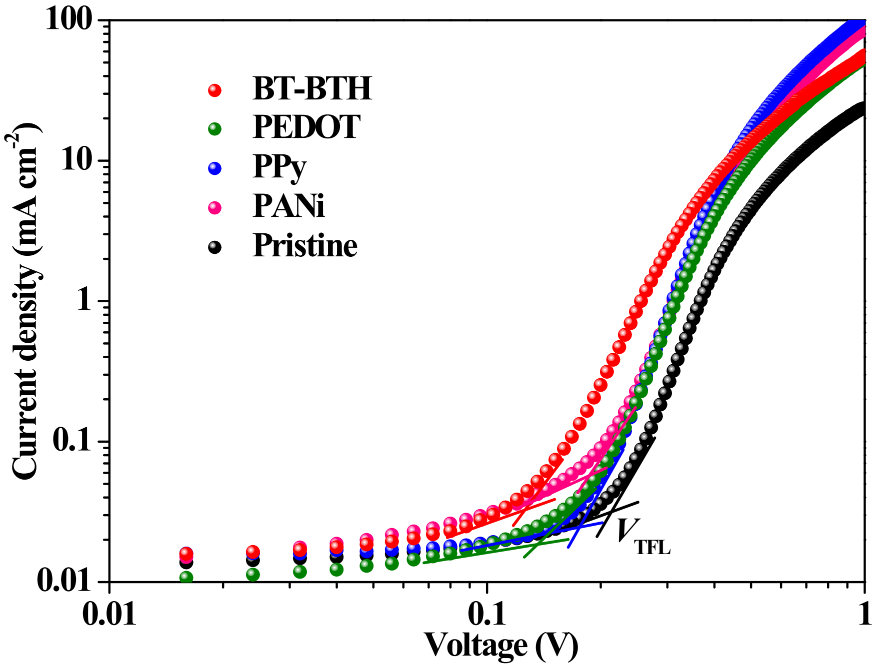


Fig. S11. The space-charge limited current (SCLC) characterizations of pristine and coated with different HTM layers.

The holes mobility of the cesium lead bromide films is further studied using a space-charge limited current (SCLC) model. Figure S8 shows SCLC curves of five devices. The trap density can be obtained according to $V_{TFL} = en_tL^2/2\varepsilon\varepsilon_0$, V_{TFL} is trap-filled limit voltage. The corresponding V_{TFL} values of BT-BTH, PEDOT, PPy, PANi, Pristine device are 0.126 V, 0.136 V, 0.169 V, 0.187 V, 0.215 V, respectively. Therefore, BT-BTH device has the highest device according to the proportional relation between n_t and V_{TFL}.