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A Multifunctional additive of Scandium Trifluoromethanesulfonate to Achieve Efficient Inverted Perovskite Solar Cells with High Fill Factor of 83.80%

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EXPERIMENTAL SECTION

Materials Preparation:

P3CT-Na was synthesized by the reaction of poly[3-(4-carboxylbutyl) thiophene (P3CT, Rieke Metals) with sodium hydroxide (molar ratio 1:1) in H_2O at room temperature under stirring for 2 day according to literature report. Scandium trifluoromethanesulfonate (Sc(OTF)3) (Adamas, >99.99%), PbI_2 (*p*-OLED, >99.99%), MAI (*p*-OLED, 99.5%), $PbCl_2$ (*p*-OLED, >99.99%), C_{60} (*p*-OLED), BCP (*p*-OLED), DMF (Sigma-Aldrich, 99.8%), DMSO (Sigma-Aldrich, 99.8%) and CB (Sigma-Aldrich, 99.8%) were used without further purification.

Device Fabrication

The indium tin oxide (ITO) glass substrates were sequentially washed by sonication using detergent, deionized water, and ethanol and dried by nitrogen flow, Then the dry ITO substrates were treated by O_2-plasma for 180 s before using. The prepared HTL P3CT-Na (1mg/mL in H_2O) was spin-coated on the ITO substrates at 4000 rpm for 60 s followed by annealing at 140°C for 30 min. After cooling down to room temperature, the substrates were transferred into glove box filled with N_2. $CH_3NH_3PbI_{3-x}Cl_x$ precursor solution (1.26 M PbI_2, 0.14 M $PbCl_2$ and 1.4 M MAI in DMF:DMSO mixed solution with a v/v of 9:1, stirred overnight at room temperature with different addition amounts of $Sc(OTF)_3$ (0, 0.1, 0.5, 1 mg/ml) were spin-coated on P3CT-M substrates at 400 rpm for 3 s and 4000 rpm for 30 s. During this spin coating process, the chlorobenzene (170 μL) was dripped directly on the substrates after spin-coated for 11 s, followed by annealing at 50°C for 2 mins and 85°C for 25 mins. C_{60} (40 nm) and BCP (6 nm) were evaporated under high vacuum on top of active layer served as electron transport layer and buffer layer respectively. At last, a 100 nm thick Ag electrode was deposited through a shadow mask.

Characterization

The current-voltage (*J-V*) curves were measured under 100 Mw/cm^2 (AM 1.5 G) simulated sunlight using Keithley 2400 in conjunction with a Newport solar simulator (94043A). The external quantum efficiency (EQE) was calculated from the photocurrent measurement under monochromatic illuminations at different wavelengths, using a lock-in amplifier (SR-830). UV-vis spectra was measured on a Shimadzu UV-2450 absorption spectrophotometer. XRD was performed on a Japan Shimadzu XRD-7000 diffractometer. The morphology of perovskite films were characterized by FE-SEM images (JSM-7800F). AFM images were collected in air on a Bruker. Steady-state PL spectra were recorded on HORIBA Scientific FluoroMax $^+$. The EIS were measured with CHI in the dark at a bias of 0.96V. Film thickness was measured with Surfcorder ET150. The current-voltage (*J-V*) characteristics of SCLC measurement were measured in dark conditions by a Keithley model 2400 source measure unit. Stability tests were performed on the devices in glove box at room temperature in dark conditions.

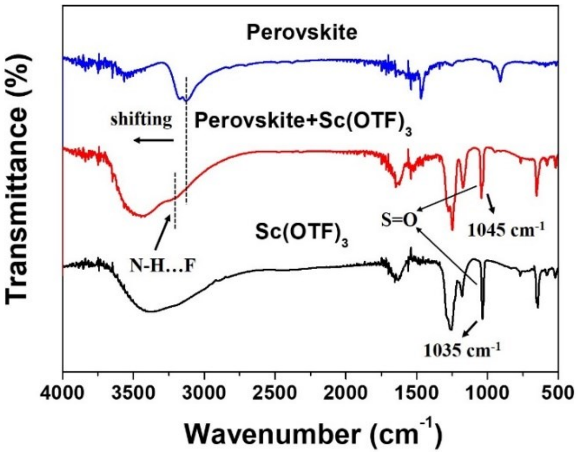


Figure S1. FTIR spectra of Sc(OTF)$_3$, pristine and Sc(OTF)$_3$ incorporated perovskite.

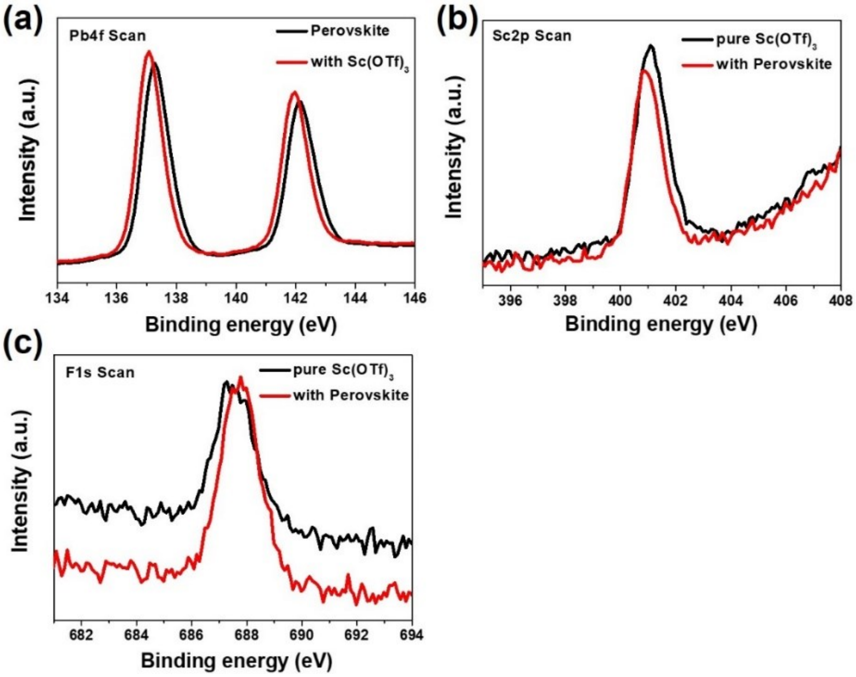


Figure S2. XPS spectra of elements (a) Pb, (b) Sc and (c) F of pure Sc(OTF)$_3$ and perovskite films doped with Sc(OTF)$_3$, respectively.

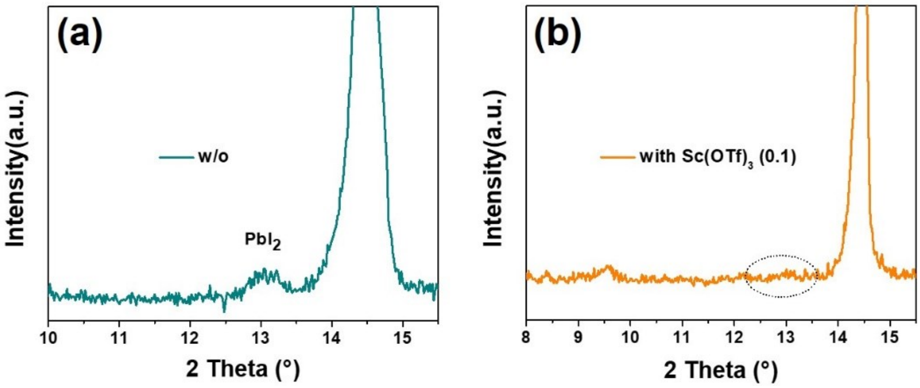


Figure S3. Magnified XRD images of (a) 0, and (b) 0.1mg/ml doped perovskite films.

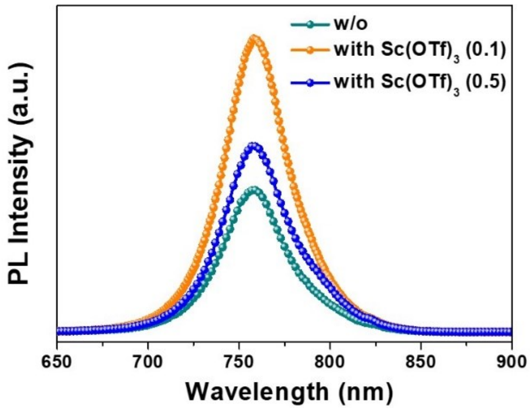


Figure S4. PL spectra of perovskite films doped with different addition amounts of Sc(OTF)$_3$.

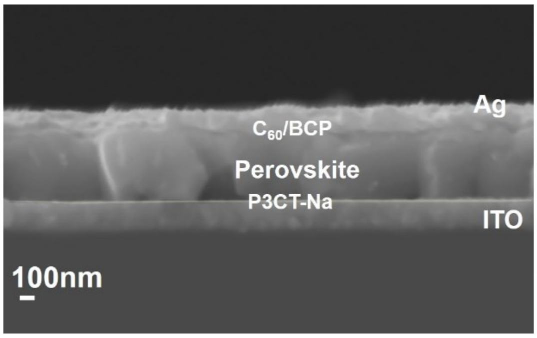


Figure S5. (a) Cross-sectional SEM image of the device.

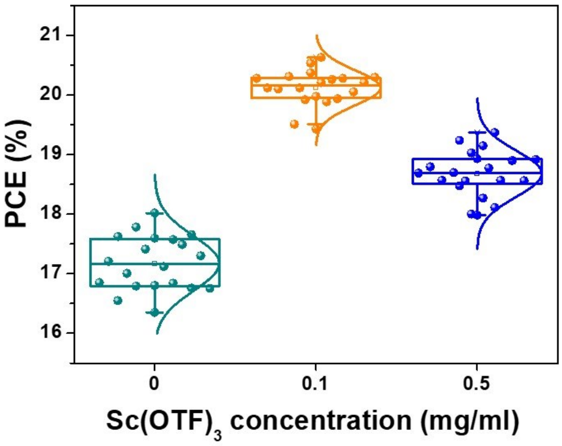
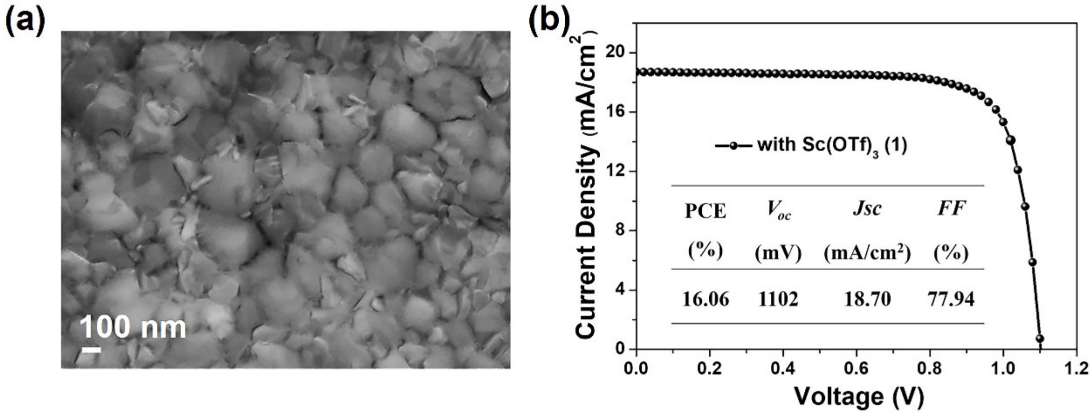


Figure S6. The PCE value dispersion of devices based on different addition amounts of Sc(OTF)$_3$ for 20 individual devices.



PCE (%)	V_{oc} (mV)	*Jsc* (mA/cm²)	*FF* (%)
16.06	1102	18.70	77.94

Figure S7. (a) SEM images of perovskite films doped with 1 mg/ml Sc(OTF). (b) Current density-voltage (*J*-*V*) curves of PSC based on 1 mg/ml $Sc(OTF)_3$ doping.

Table S1. Summary of device reverse scan performance with different amounts of $Sc(OTF)_3$ additive.

$S_C(OTF)_3$	**V_{oc} / (mV)**	**J_{sc} / (mA/cm^2)**	***FF* / (%)**	**PCE / (%)**
0	1080	20.43	79.41	17.52
0.1	1132	21.68	83.49	20.49
0.5	1115	20.73	82.42	19.05

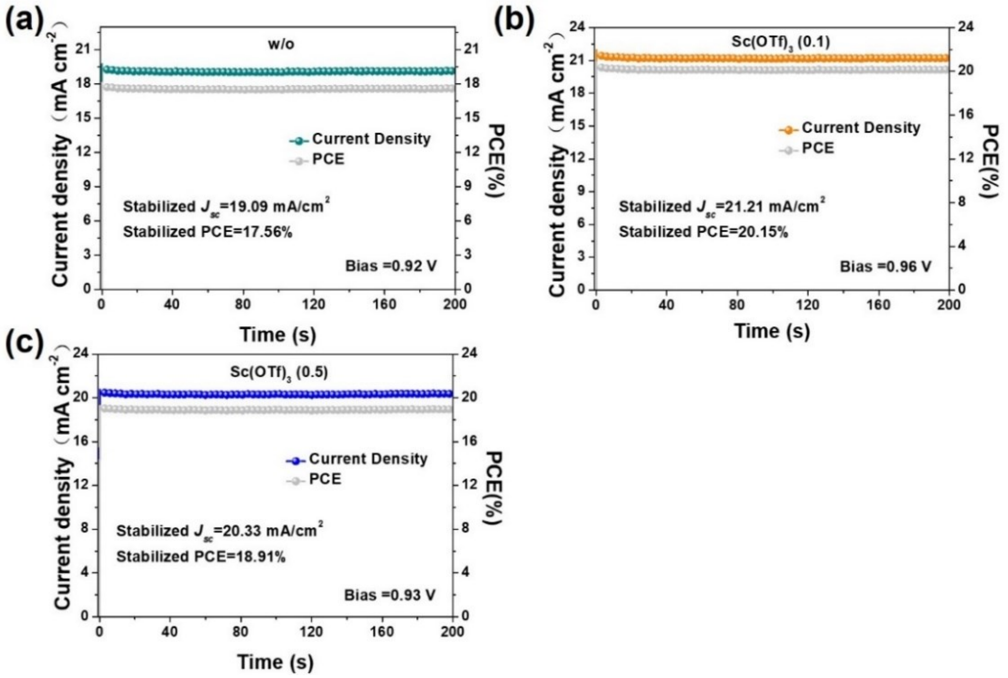


Figure S8. The stabilized J_{sc} and PCE of the devices based on different addition amounts of Sc(OTF)$_3$.

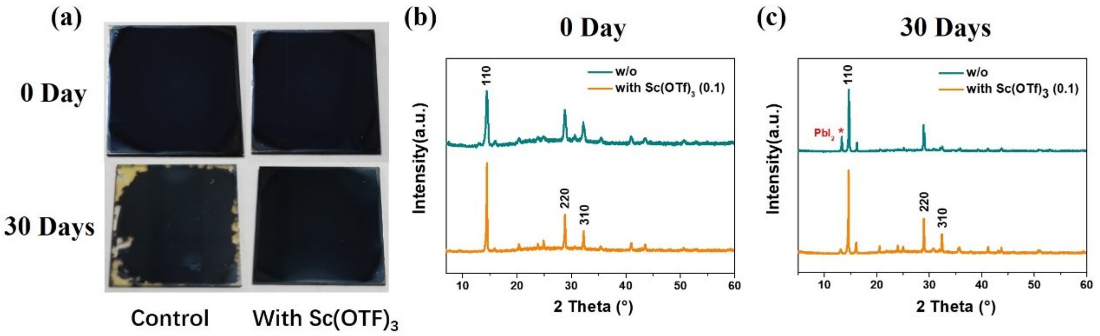


Figure S9. (a) Images and (b-c) XRD patterns of films doped with different addition amounts of Sc(OTF)$_3$ before and after aging in the air with 50% relative humidity.

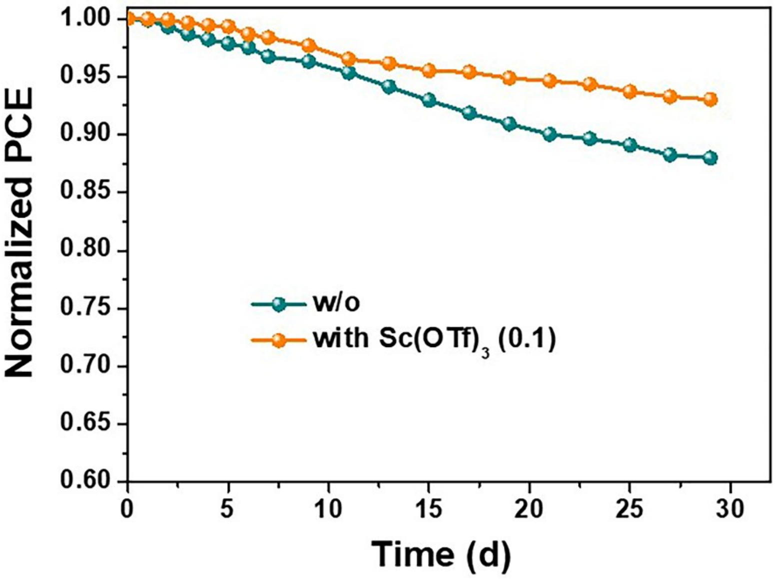


Figure S10. the stability measurement of devices based on different addition amounts of Sc(OTF)$_3$.