Halogen bond mediated inhibition of ion migration for stable Sn-Pb perovskite solar cells

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Experiment Section

Materials and Solvents. All chemicals are purchased directly from the distributor and are ready for use upon receipt without further purification and processing. Methylammonium iodide (MAI, ⩾99.5%), bathocuproine (BCP, > 99%), PEDOT:PSS (CLEVIOS P VP AI 4083 from Heraeus) were purchased from Xi'an Polymer Light Technology Corp. Formamidine iodide (FAI), lead (II) iodide (PbI_2) was purchased from Advanced Election Technology Co. Ltd. Tin (II) iodide (SnI_2, 99.999%, Sigma), cesium iodide (CsI, > 99.0%, TCI), Perfluoro Octane Iodide (PFOI, > 98.0%, Sigma). C_{60} and PCBM were purchased from Luminescence Technology Corp. N, N-Dimethylformamide (DMF, 99.8%, Acros), dimethyl sulfoxide (DMSO, 99.9%, Acros), isopropanol (IPA, 99.5%, Acros), chlorobenzene (CB, 99.8%, Acros) and toluene (Acros) were purchased from Beijing Innochem Science & Technology co., LTD. Ag was bought from Zhong Nuo Advanced Material (Beijing) Technology Co., Ltd.

Device Fabrication for Sn-Pb PSCs. In this experiment, we were prepared with $Cs_{0.1}MA_{0.2}FA_{0.7}Pb_{0.5}Sn_{0.5}I_3$ (1.8 M) of Sn-Pb perovskite solar cells. First, 1 ml of PEDOT: PSS was filtered using 0.22μm PTFE, and 45 μl of PEDOT: PSS was spun on ITO conductive glass at 4500 rpm at a rotational speed of 30s. After spinning, it was annealed on a hot bench at 150℃ for 15 min. After annealing, transfer it to the N_2 glove box for reserve. Subsequently, the filtered precursor solution was spin-coated on the prepared HTL at 1000 rpm, 10 s with an acceleration of 500 rpm/s, and then CB drops were added to the spin-coated precursor solution at the appropriate rate 20 s before the

end of the second step (4500 rpm, 50s). The film was then transferred to a hot bench at 100°C and annealed for 10 minutes.Then, after the annealing was completed and the films were cooled to room temperature, the PCBM was spin-coated onto the films at 1200 rpm for 30 s, followed by annealing at 100°C for 3 min. After the film was annealed and cooled to room temperature, the hole barrier layer BCP was spun on the film at a rotational speed of 4500rpm and 30s. Finally, the films were transferred to a vapor deposition machine to deposit 80 nm Ag.

Devices Characterizations. The *J-V* curves of the devices (with a scan rate of 20 mV/30 ms) and the steady-state photocurrent at the maximum power output bias were tested using a solar simulator (PV Measurements Inc.) under AM 1.5G illumination at 100 mW/cm^2. The *J-V* curves of all devices were obtained after masking using a metal with an aperture area of 0.09 cm^2. X-ray photoelectron spectroscopy (XPS) was performed on the films using a Thermo Fisher ESCALAB 250 in an air environment and the data were fitted using Avantage software. Ultraviolet photoelectron spectroscopy (UPS) tests are performed using a He-I source ($h\upsilon$ = 21.22 eV) (AXIS ULTRA DLD, Kratos, UK) to obtain valence band energy and Fermi levels of perovskite films. The measurement of variable temperature conductivity is obtained by measuring the change of conductivity with temperature at different temperatures (-80~50℃) and at a given voltage (1 V), and plotting the test results, calculating the ion migration activation energy (E_a) by fitting the trend of the conductivity with temperature change.

Supplementary Figures

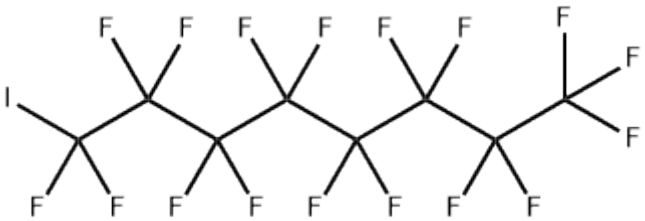

Figure S1. Molecular structure formula of PFOI.

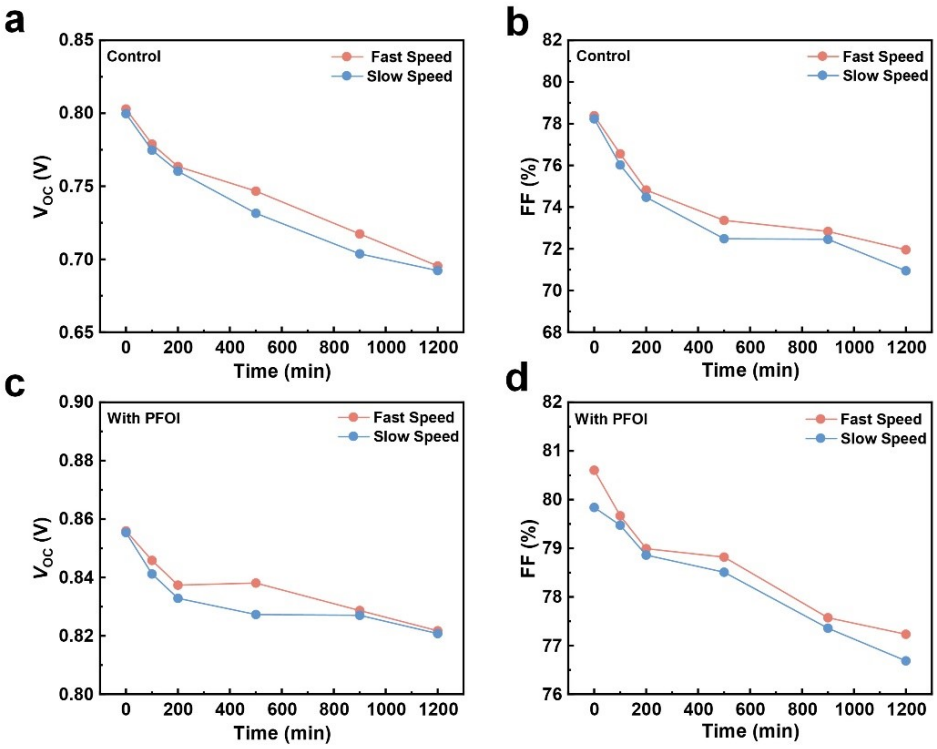

Figure S2. The changes of (a) V_{OC} and (b) FF with aging time of the device without PFOI treatment at different sweep speeds. The changes of (c) V_{OC} and (d) FF with aging time of PFOI-treated devices at different sweep speeds.

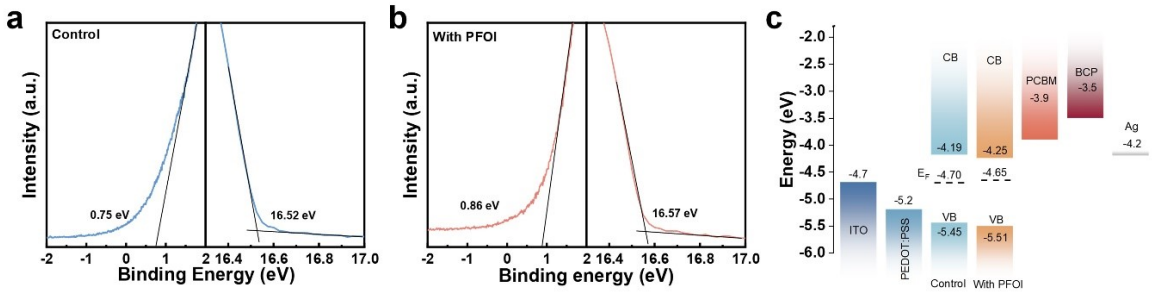

Figure S3. UPS data of the perovskite films (a) without and with (b) PFOI. (c) Energy level of each layer in devices.

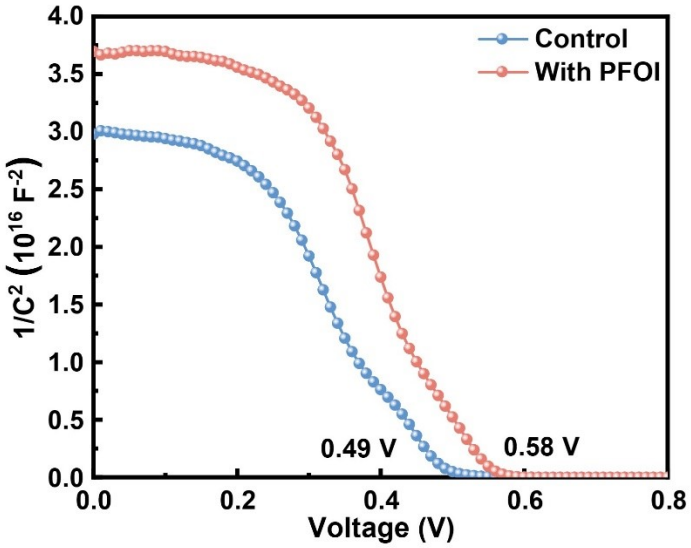

Figure S4. Mott-Schottky plots of the device with and without PFOI treatment.

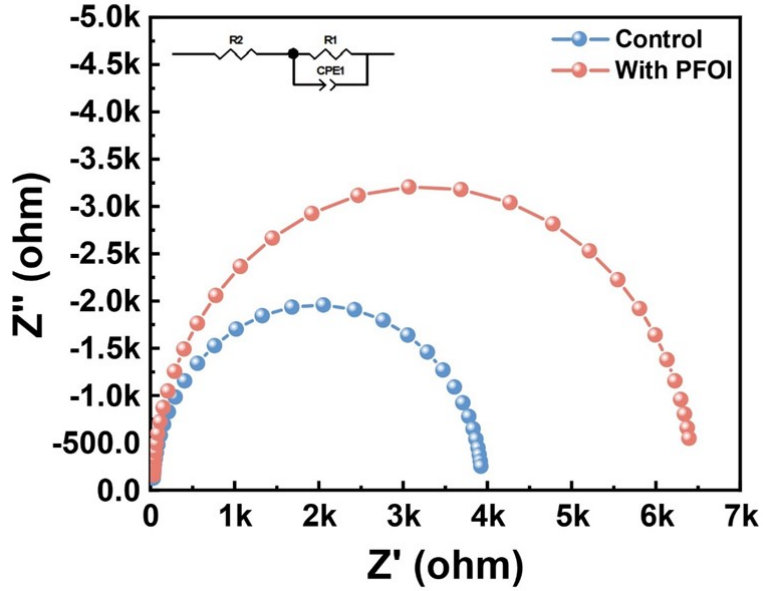

Figure S5. Nyquist plots of the device with and without PFOI treatment.

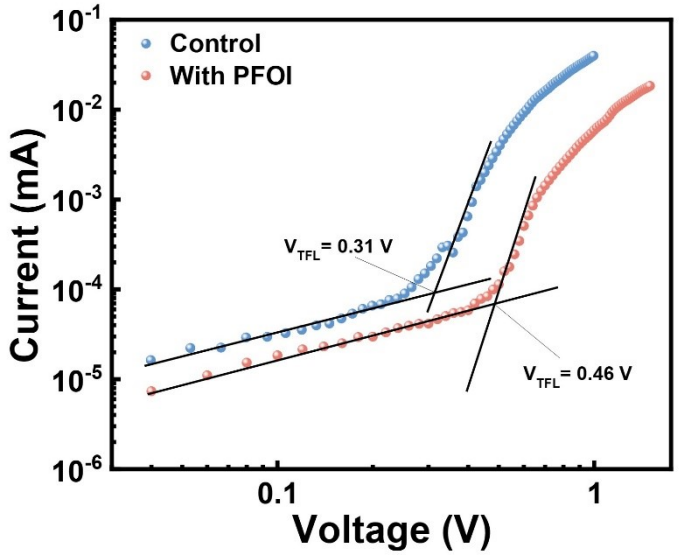

Figure S6. SCLC curves of hole-only devices (ITO/PEDOT: PSS/perovskite/Spiro-OMeTAD/Ag) in the device treated with and without PFOI.

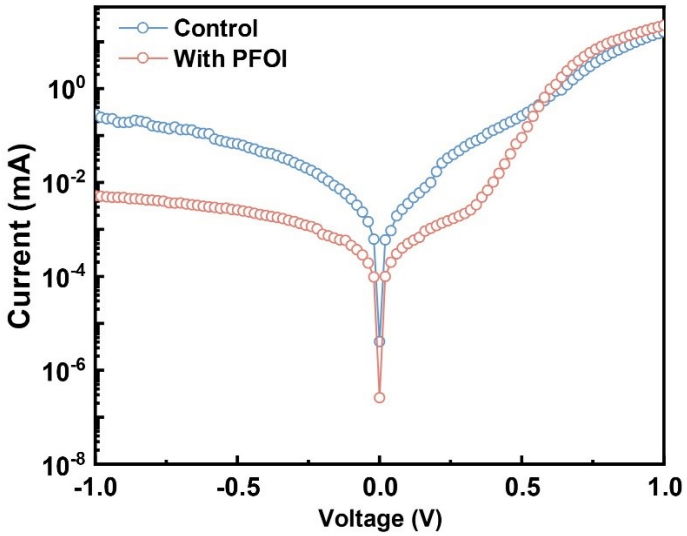

Figure S7. Dark J-V curves of with and without PFOI-treatment devices.

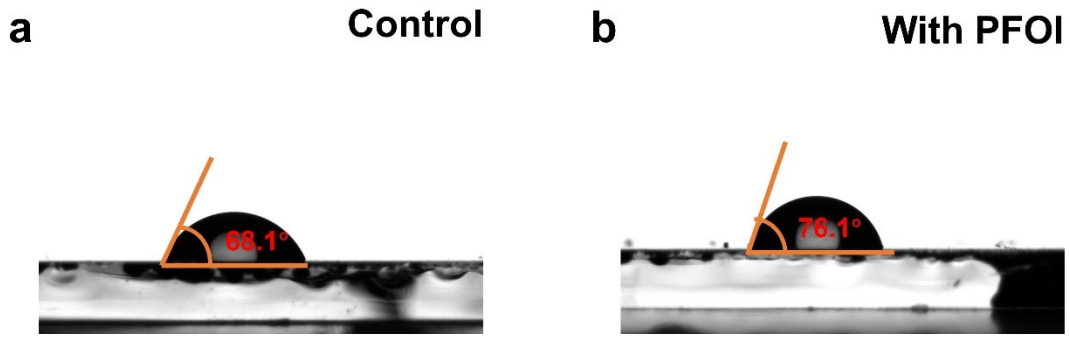

Figure S8. The contact angle of the perovskite film treated (a) without and (b) with PFOI.

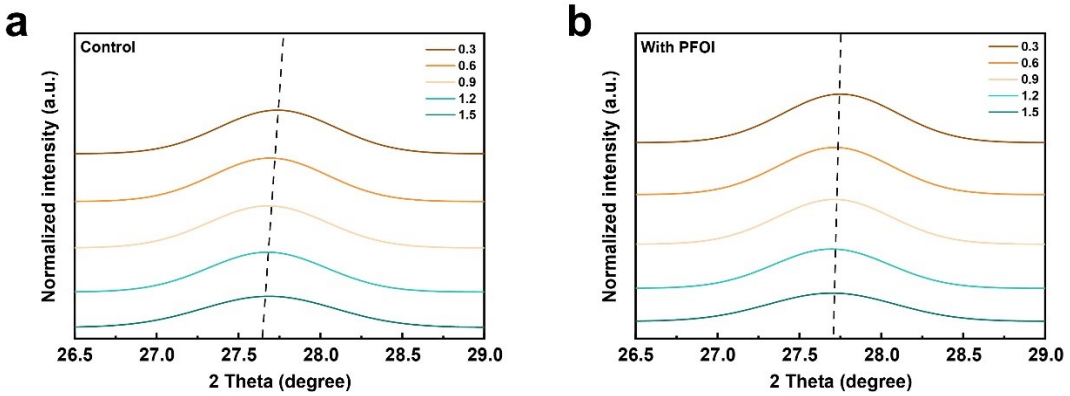

Figure S9. GIXRD images (0.3°-1.5°) of (a) without and (b) with PFOI-treated perovskite films.