Supporting Information

Synergistic effect of anions and cations in additive for highly efficient and stable perovskite solar cells

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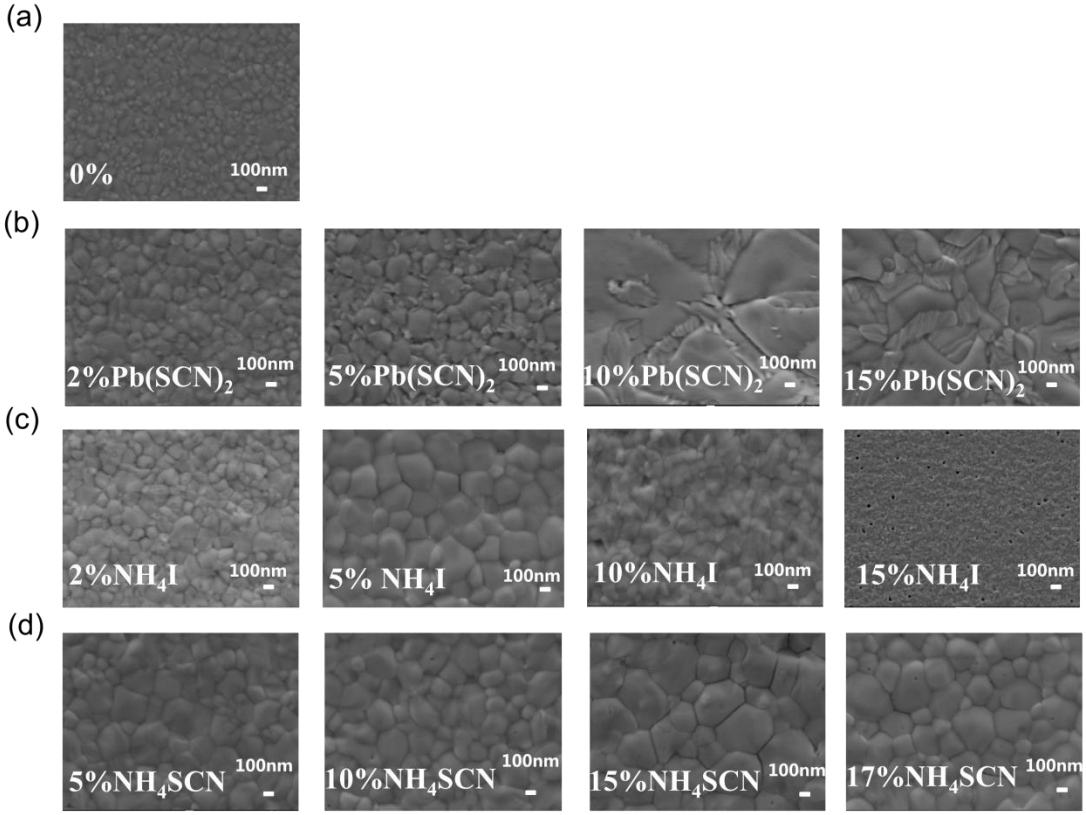

Figure S1. The top-view SEM images of perovskite films. a) Pure perovskite film, b) Perovskite films with 2%, 5%, 10%, and 15% $Pb(SCN)_2$, c) Perovskite films with 2%, 5%, 10%, and 15% NH_4I, d) Perovskite films with 5%, 10%, 15%, and 17% NH_4SCN.

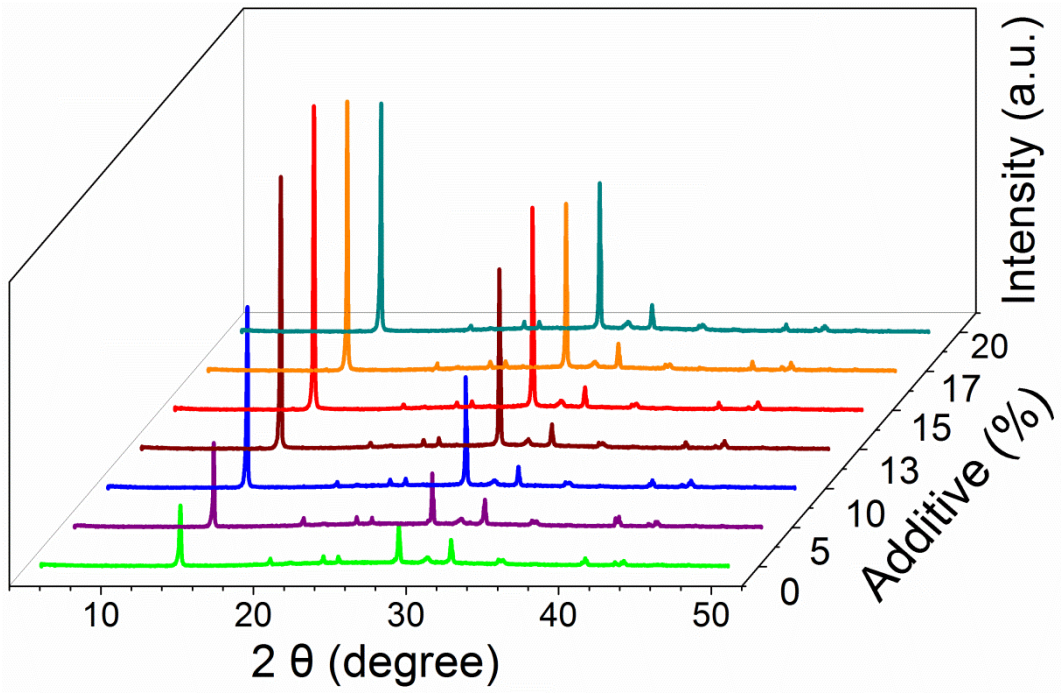

Figure S2. XRD patterns of the perovskite films with 0%, 5%, 10%, 13%, 15%, and 17% NH_4SCN.

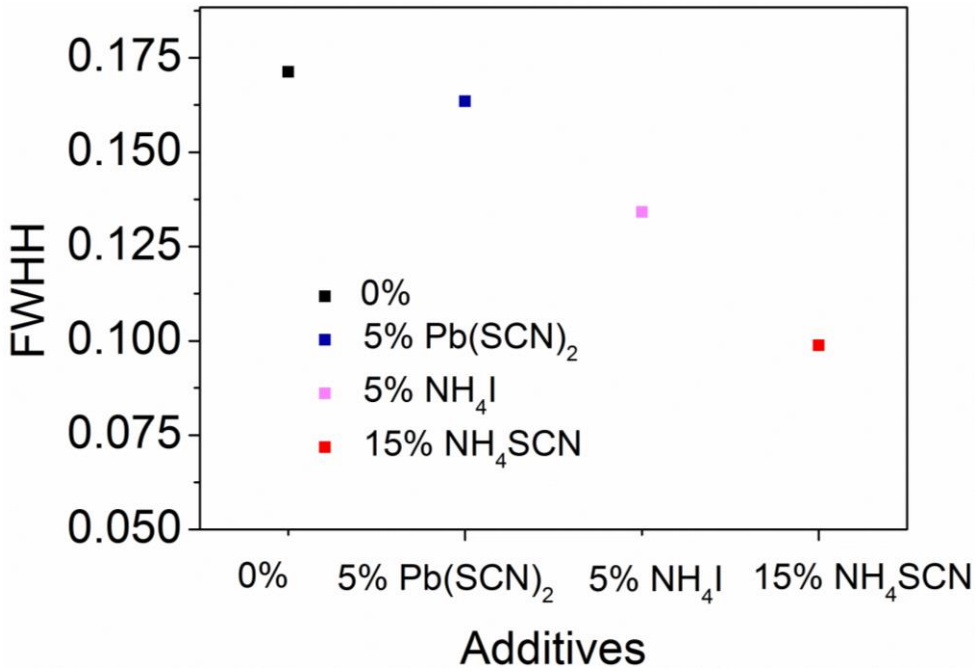

Figure S3. The Full Width of Half Height (FWHH) of (110) diffraction peaks of films with different additives.

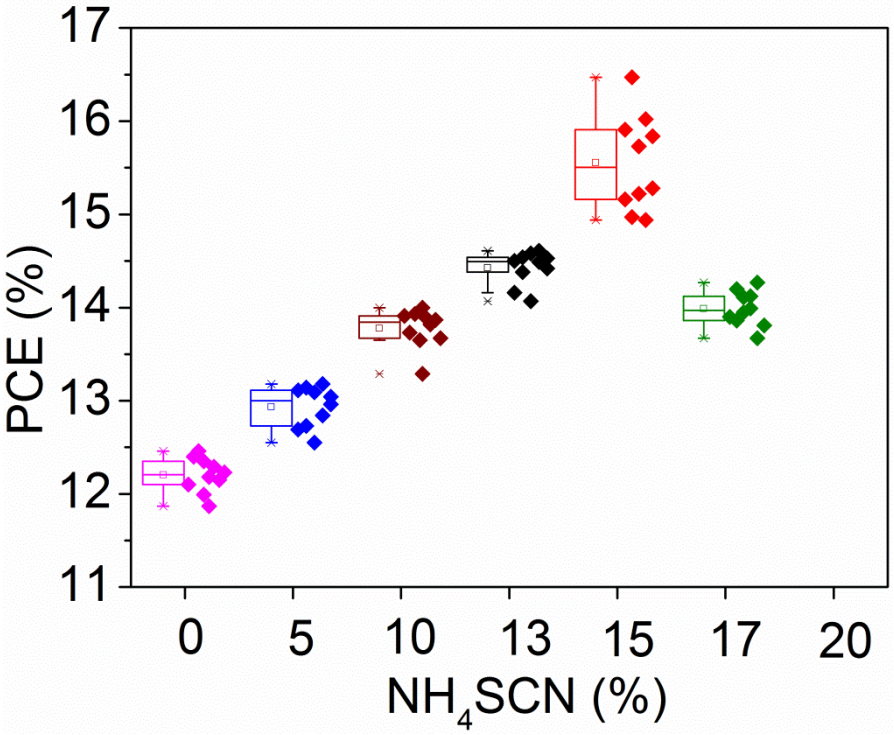

Figure S4. The statistical diagram of PCEs of the devices prepared by adding different amounts of NH_4SCN additive in the precursor solution.

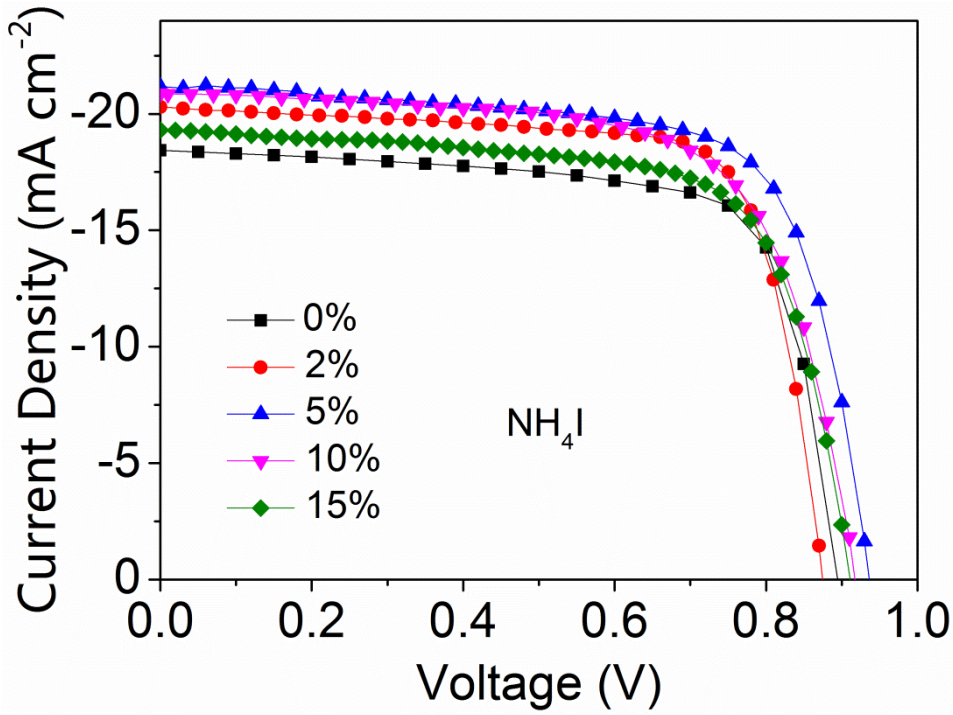

Figure S5. The current density–voltage (J–V) curves of the devices with 0%, 2%, 5%, 10% and 15% NH_4I additive.

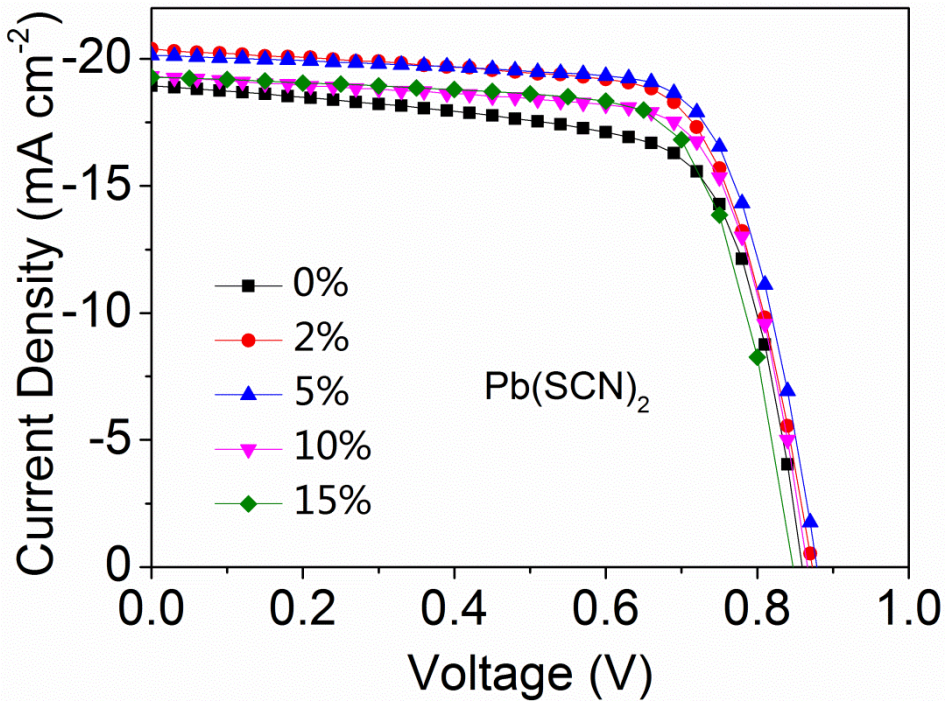

Figure S6. The current density–voltage (J–V) curves of the devices with 0%, 2%, 5%, 10%, 15% $Pb(SCN)_2$ additive.

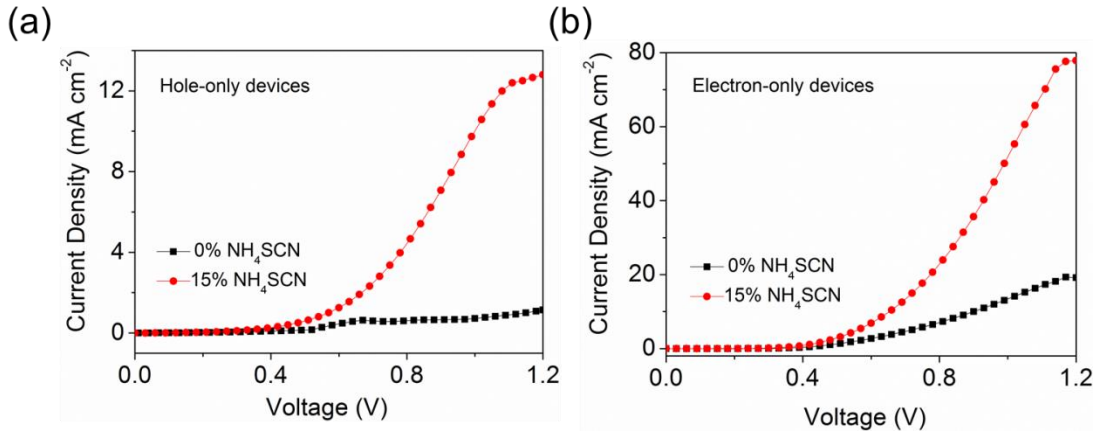

Figure S7. The current density-voltage curves of the hole-only devices with the structure of ITO/PEDOT:PSS/perovskite/Poly-TPD/Al and electron-only devices with the structure of ITO/SnO_2/perovskite/PCBM/Al.

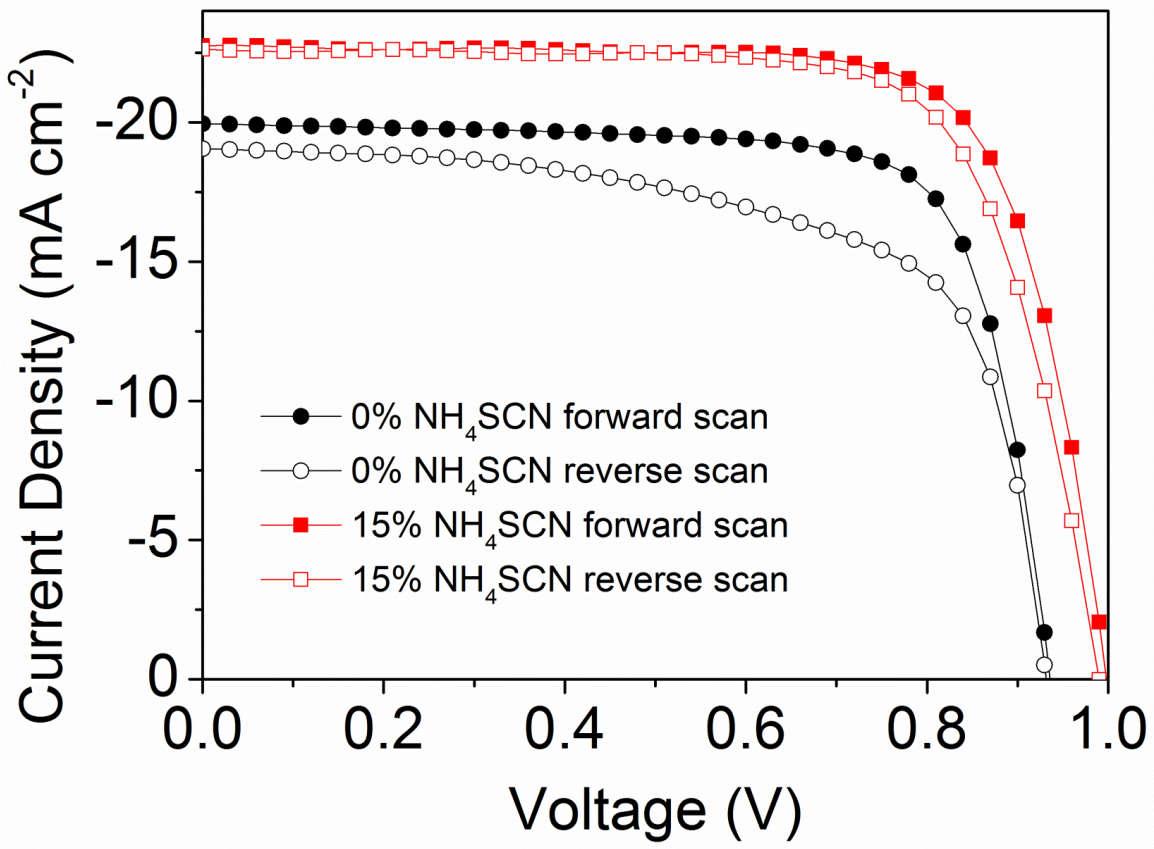

Figure S8. Hysteresis effect of devices with the device structure of ITO/SnO_2 (20 nm)/perovskites with or without NH_4SCN additive/ Spiro-OMeTAD (100 nm)/MoO_3 (5 nm)/Ag (120 nm).

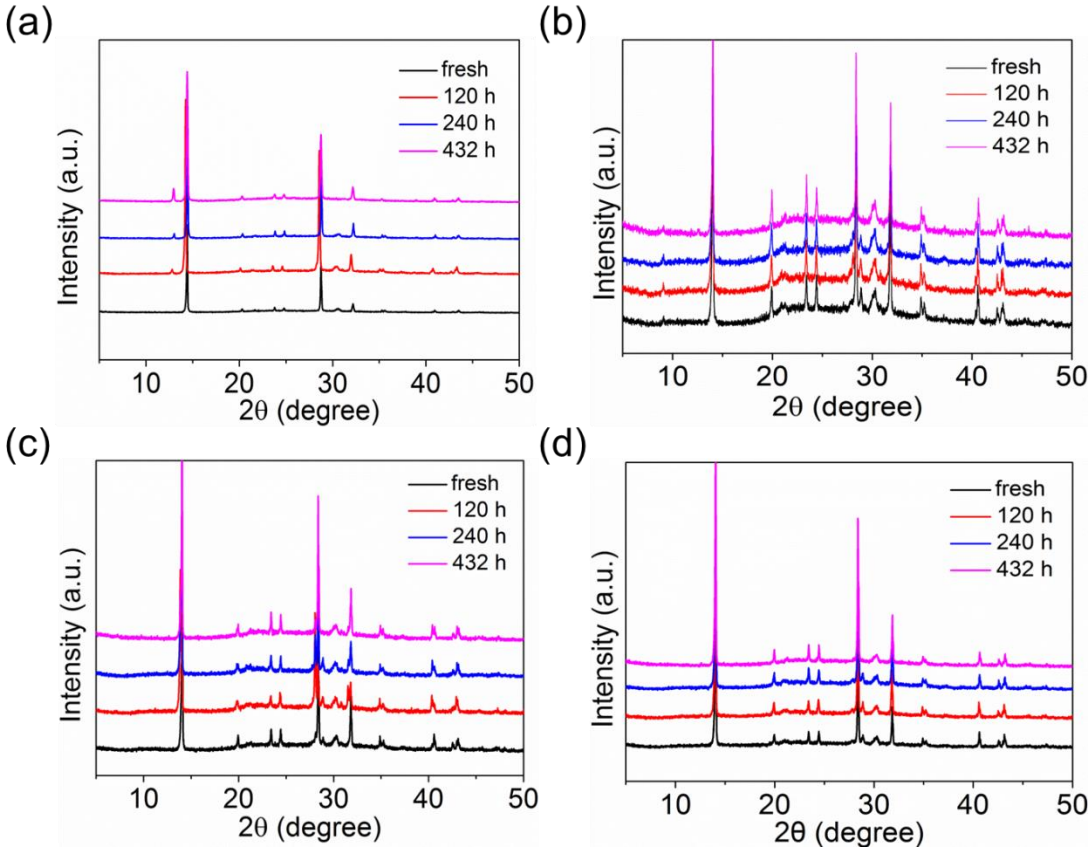

Figure S9. XRD patterns evaluation of the perovskite films with a) 0% additive, b) 5% $Pb(SCN)_2$, c) 5% NH_4I, d) 15% NH_4SCN.

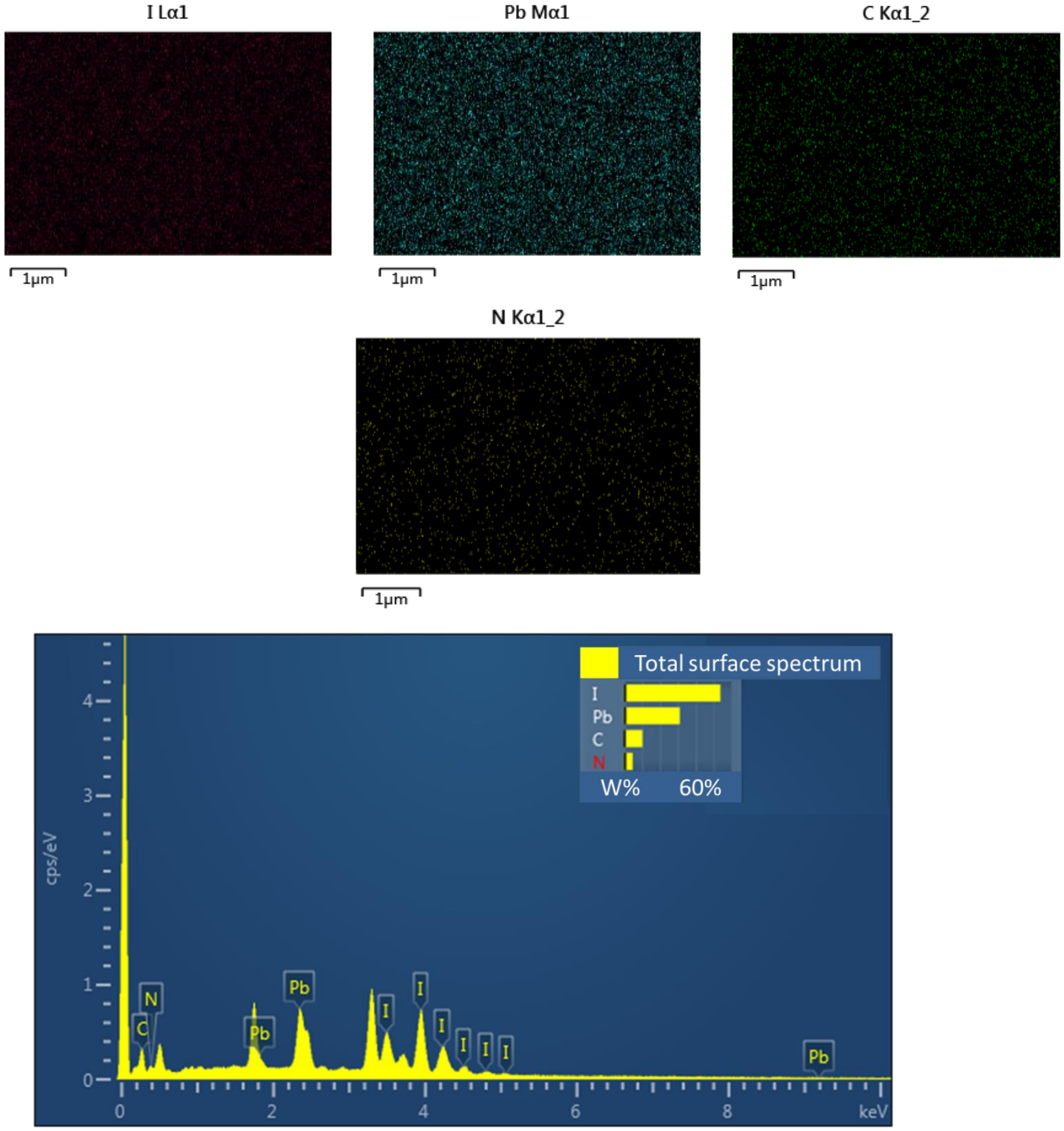

Figure S10. The elemental distribution in the perovskite films without additive analyzed by Energy Dispersive Spectrometer (EDS).

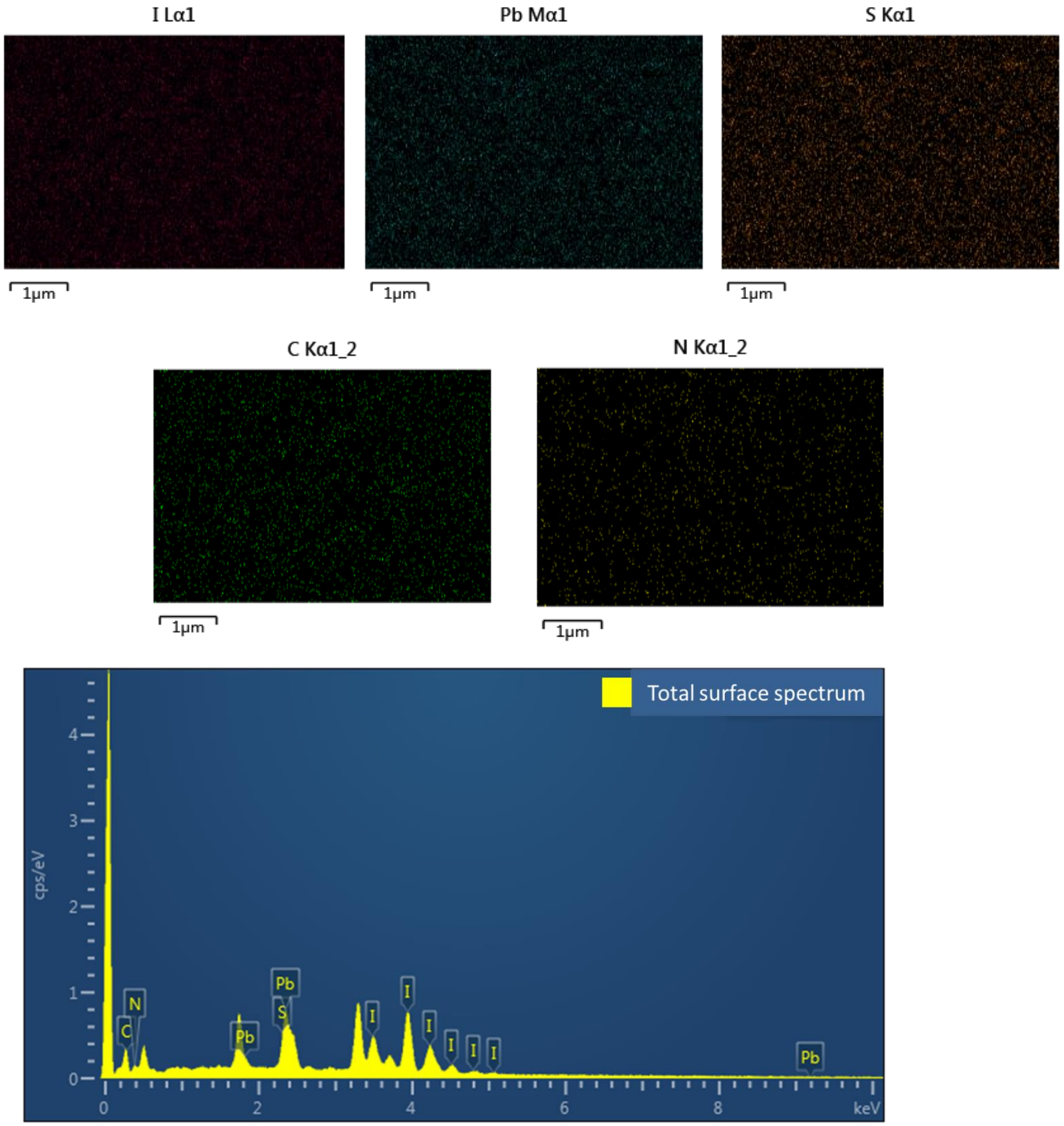

Figure S11. The elemental distribution in the perovskite films with 15% NH_4SCN additive analyzed by EDS.

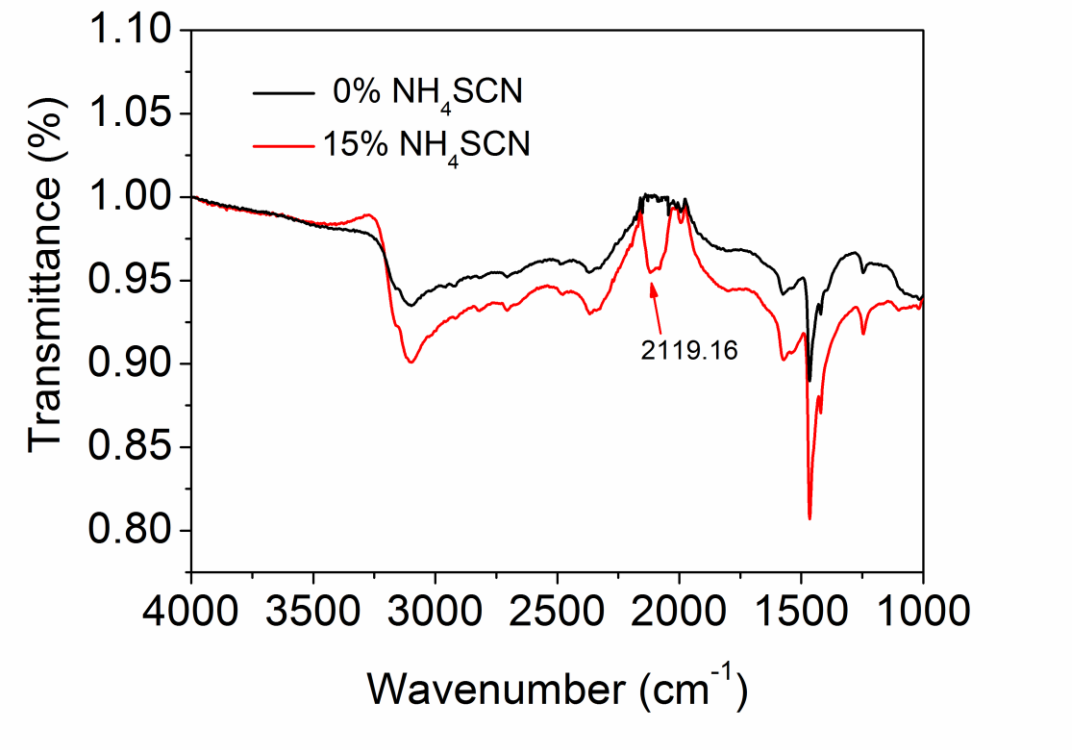

Figure S12. FTIR spectra of perovskite powders with and without NH_4SCN additive.

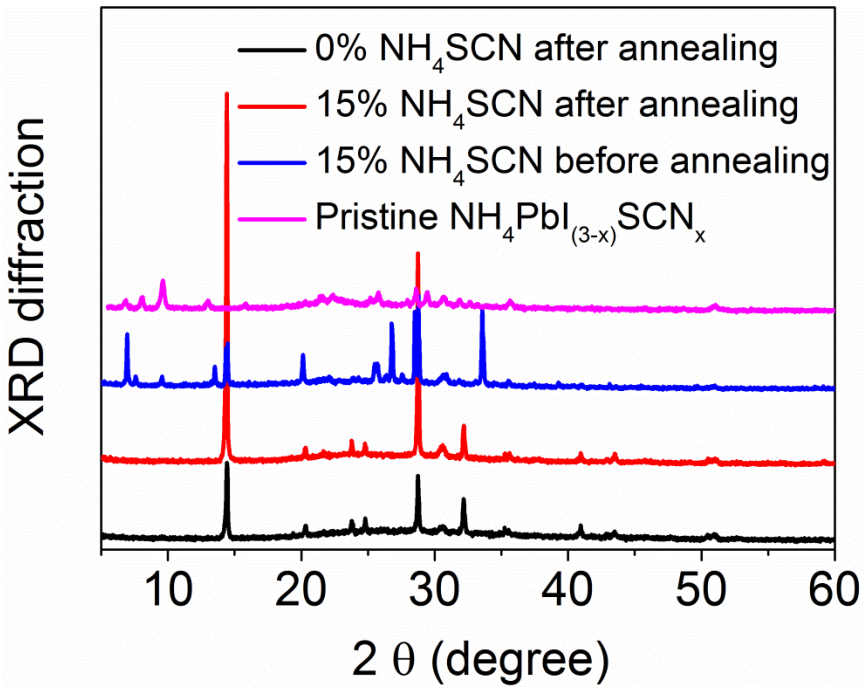

Figure S13. The XRD patterns of NH_4SCN based perovskite films before and after annealing, the pristine $NH_4PbI_{3-x}SCN_x$ perovskite, and $MAPbI_3$ without additives.

Table S1. The calculation of the formation enthalpy (∆Hf) of the following reactions: $NH_4I + PbI_2 \rightarrow NH4PbI_3$ and $MAI + PbI_2 \rightarrow MAPbI_3$.

Compounds	Energy (eV)
NH_4I	-1140.6337047659
CH_3NH_3I	-1335.5293553123
PbI_2	-3603.1894938924
$MAPbI_3$	-4939.9162464459
NH_4PbI_3	-4745.1273744954

$\Delta H_f(MAPbI_3)$ = -1.197397241 (eV)
$\Delta H_f(NH_4PbI_3)$= -1.304175837 (eV)

Ab-initio calculations are performed using density functional theory in the generalized gradient approximation (GGA) with Perdew-Burke-Ernzerhof parametrization for the exchange-correlation functional [1], as implemented in the Quantum-ESPRESSO package [2]. We employ ultrasoft pseudopotentials and a 408 eV plane wave energy cutoff. The atomic coordinates are fully relaxed until all atomic forces have declined below 0.01 eV/Å. The Brillouin zone is sampled on a 4×4×4 k-point grid.

[1] B. Hammer, L. B. Hansen, and J. K. Nørskov, Phys. Rev. B 1999, 59, 7413.

[2] Giannozzi, P., et al., QuantumESPRESSO: A Modular and Open-Source Software Project for Quantum Simulations of Materials. J. Phys. Condens. Matter, 2009, 21, 395502.