

# Supporting Information

## A Thiocyanate Containing Two-Dimensional Cesium-Lead Iodide Perovskite, $\text{Cs}_2\text{PbI}_2(\text{SCN})_2$ ; Characterization, Photovoltaic Application, and Degradation Mechanism

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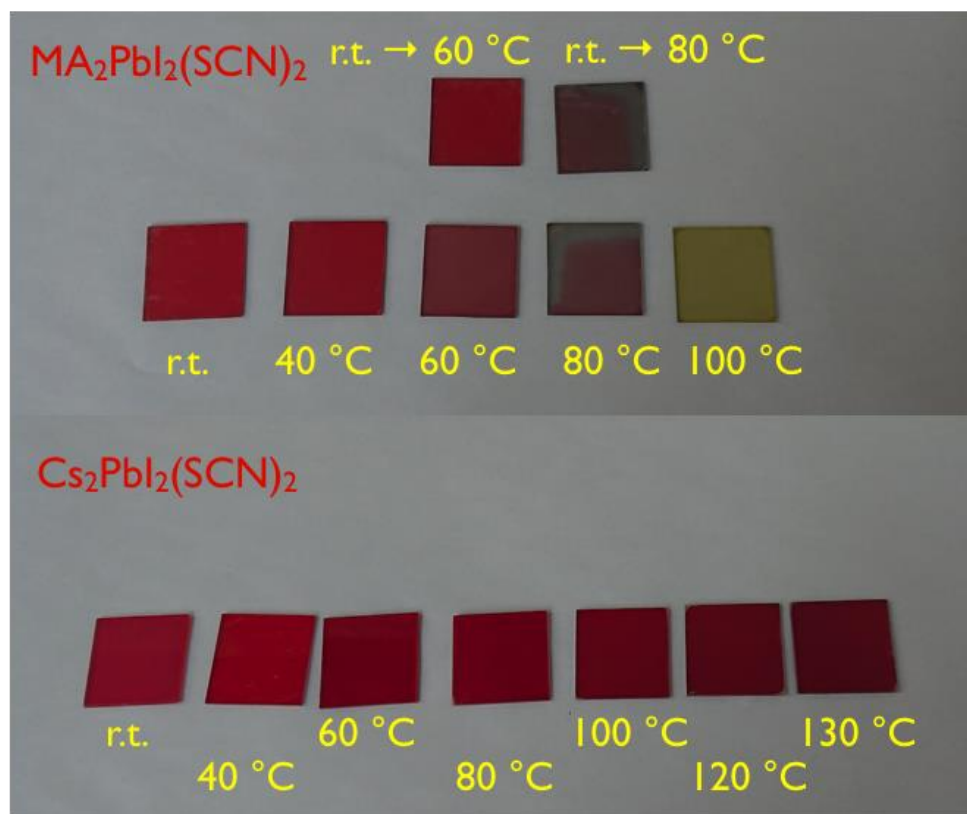
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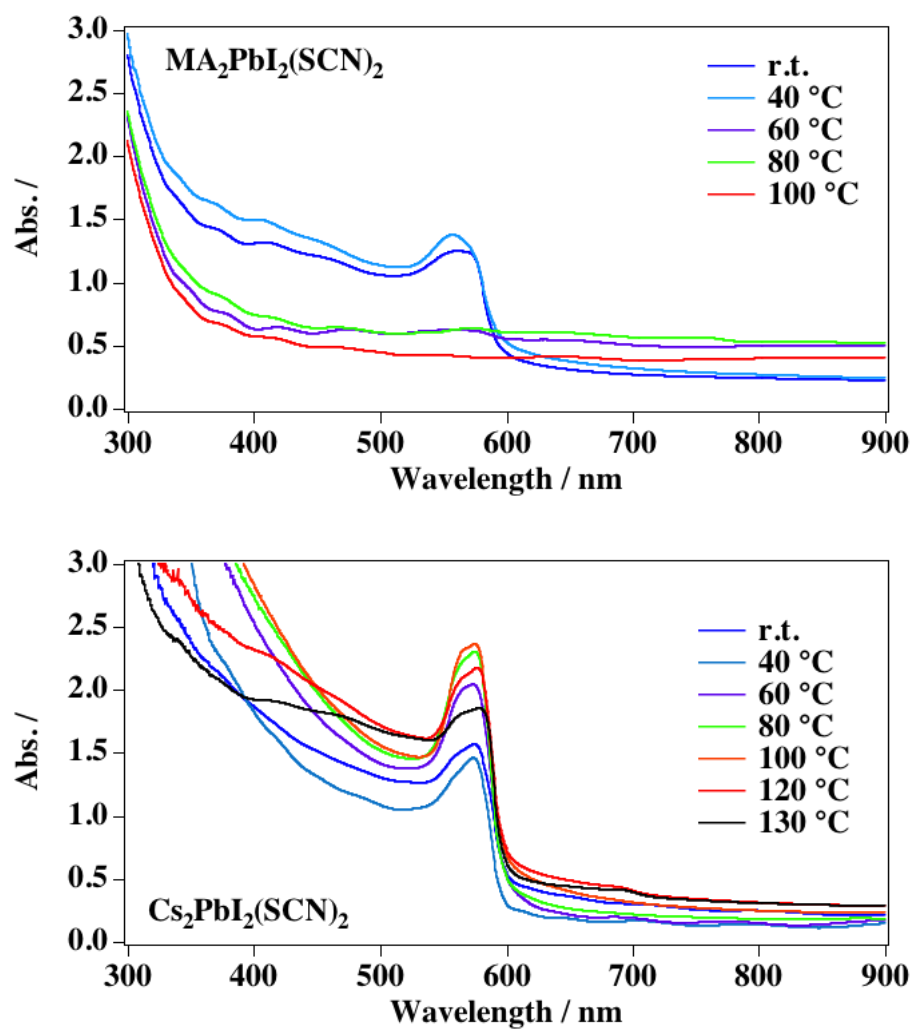
Y. Numata, [y\\_numata@dsc.rcast.u-tokyo.ac.jp](mailto:y_numata@dsc.rcast.u-tokyo.ac.jp) and T. Miyasaka, [miyasaka@toin.ac.jp](mailto:miyasaka@toin.ac.jp).



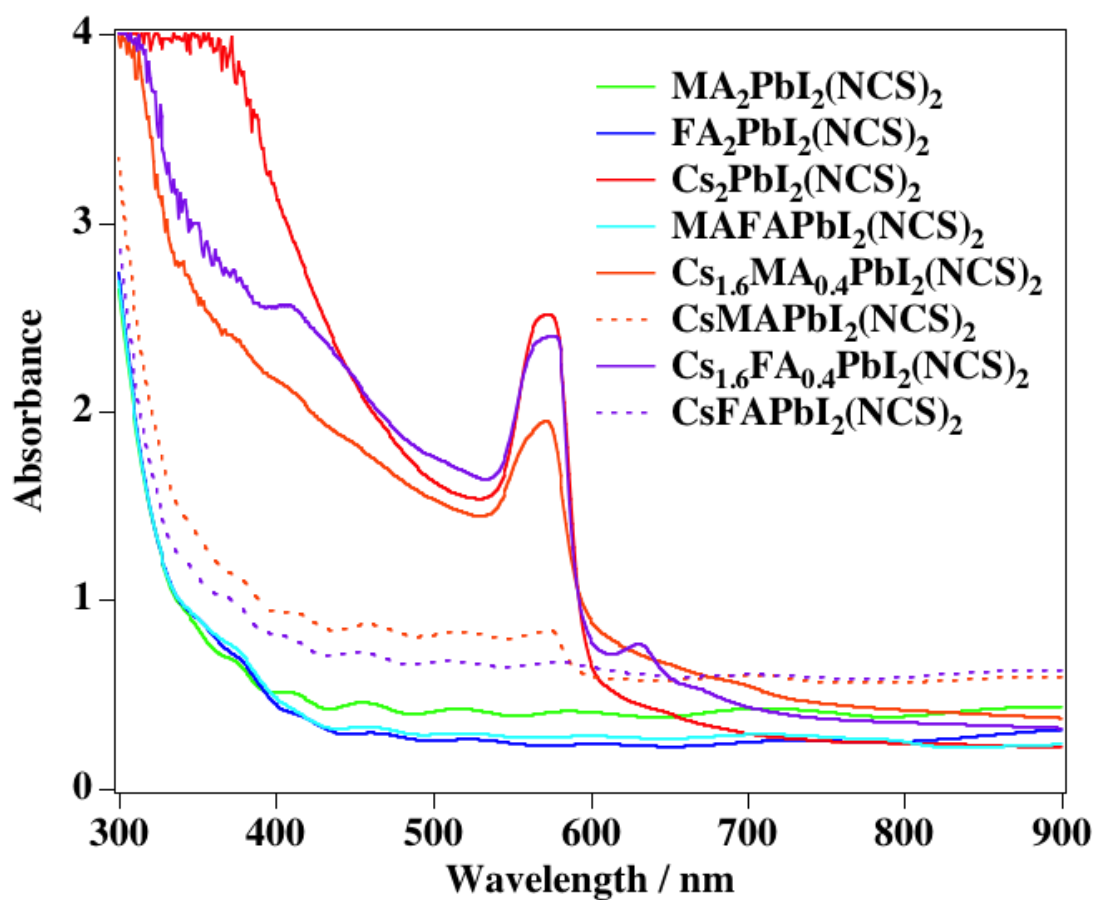
**Figure S1.** Photograph of  $AA'PbI_2(SCN)_2$  films on mesoporous  $TiO_2$  films (annealed at 100 °C for 1 min).



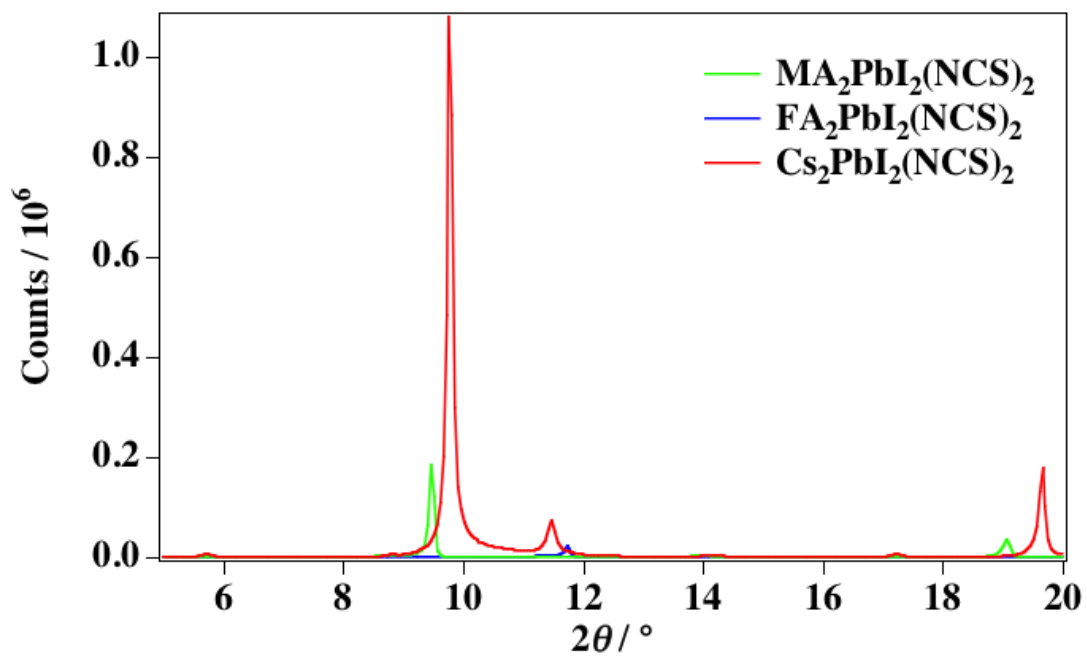
**Figure S2.** Photograph of (top)  $\text{MA}_2\text{PbI}_2(\text{SCN})_2$  and (bottom)  $\text{Cs}_2\text{PbI}_2(\text{SCN})_2$  films on mesoporous  $\text{TiO}_2$  films annealed at different temperatures. (For  $\text{MA}_2\text{PbI}_2(\text{SCN})_2$ , black part of the 80 °C sample is residual high temperature phase and yellow color of the 100 °C sample is due to the long time annealing than the pale-red sample in Figure S1).



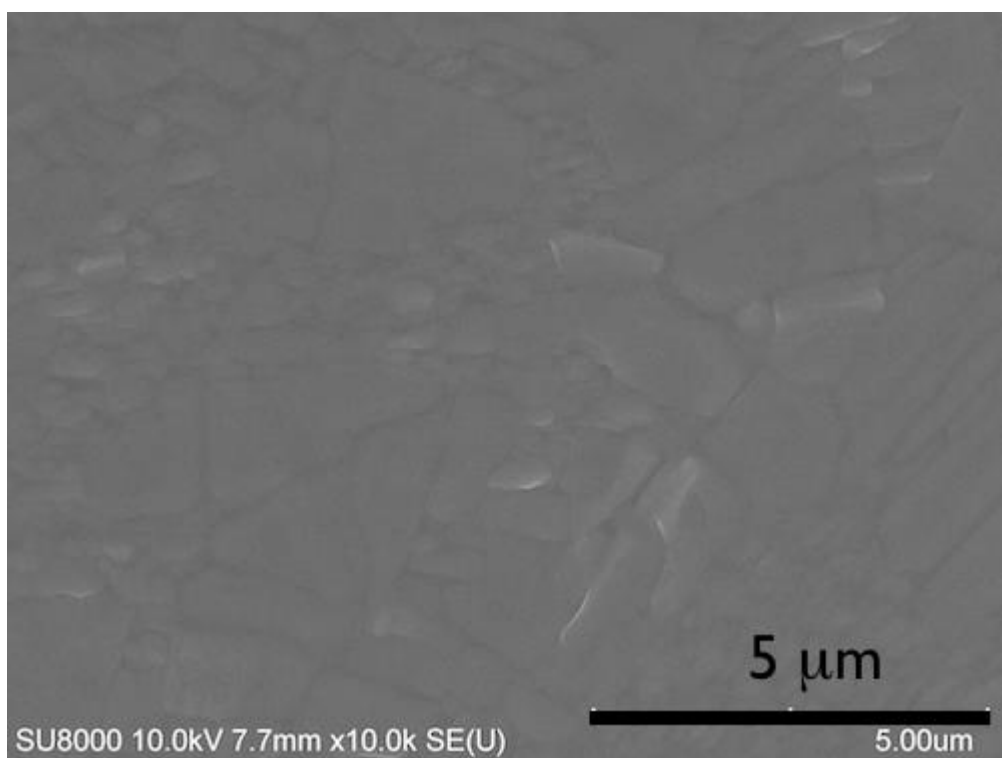
**Figure S3.** Absorption spectra of (top)  $\text{MA}_2\text{PbI}_2(\text{SCN})_2$  and (bottom)  $\text{Cs}_2\text{PbI}_2(\text{SCN})_2$  films on mesoporous  $\text{TiO}_2$  films annealed at different temperatures shown in Figure S2.



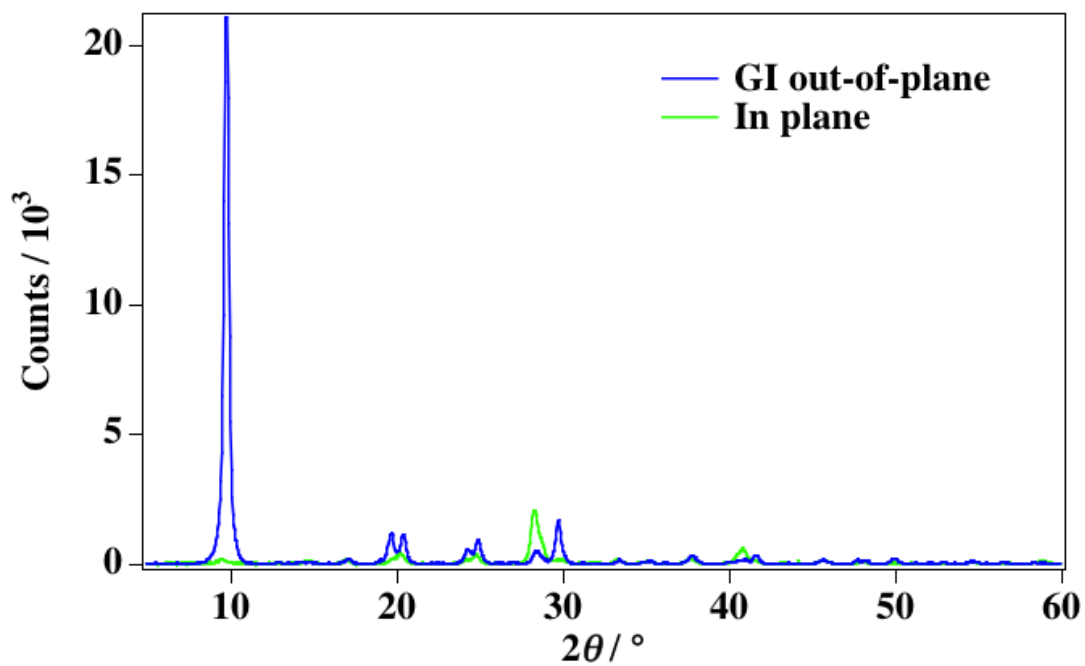
**Figure S4.** Absorbance of  $A_2PbI_2(NCS)_2$  films on mesoporous  $TiO_2$  films dried at 100 °C.



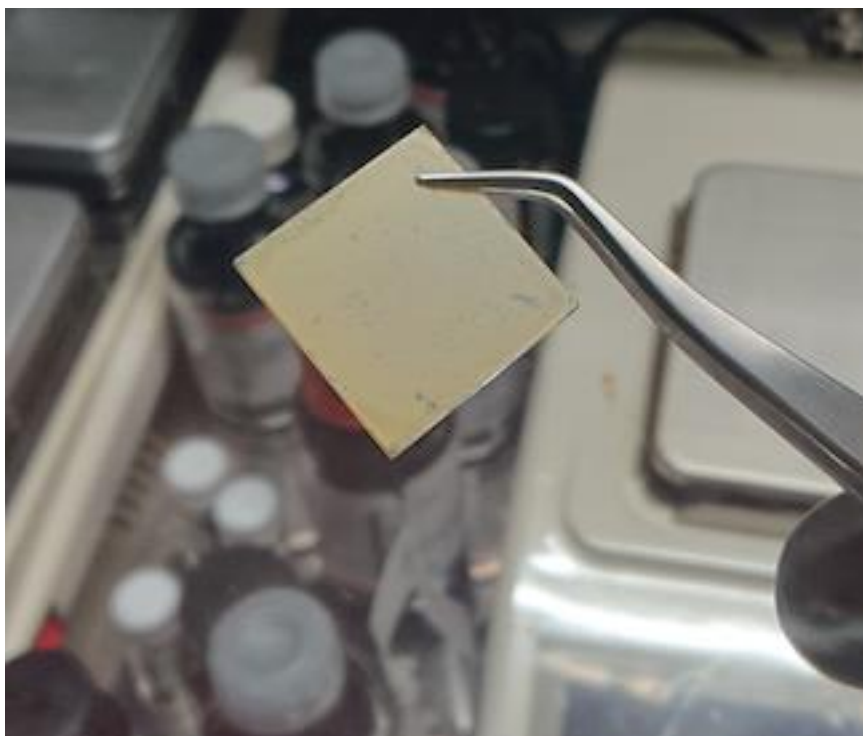
**Figure S5.** XRD chart of A<sub>2</sub>PbI<sub>2</sub>(SCN)<sub>2</sub> perovskite films on mesoporous TiO<sub>2</sub> substrate annealed at 100 °C for 2 min.



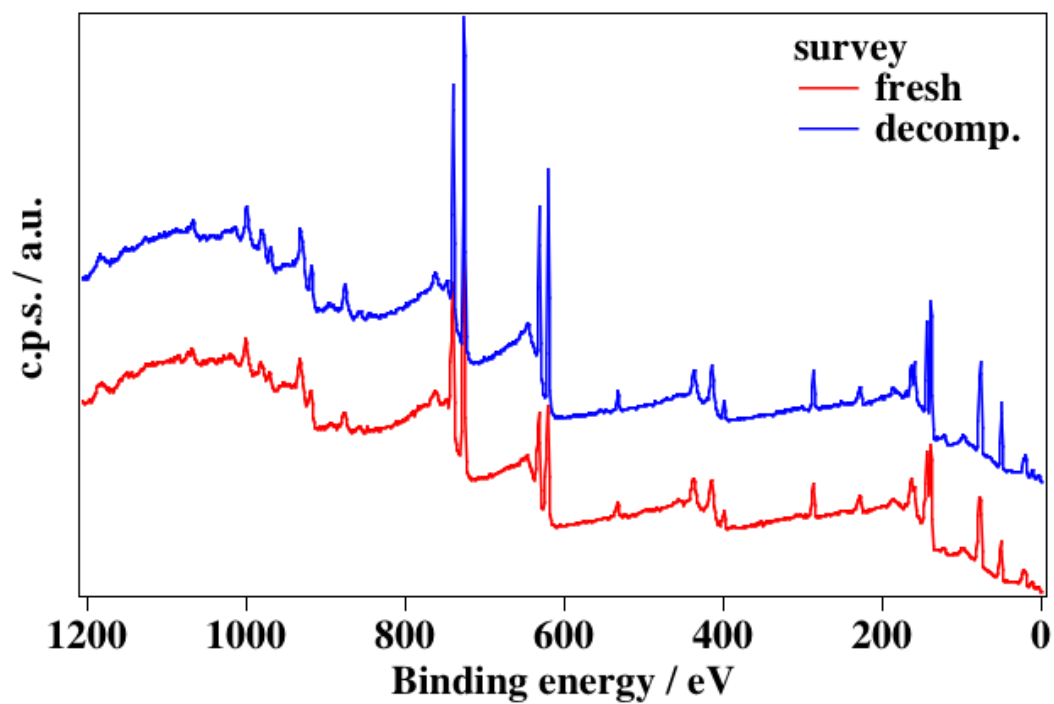
**Figure S6.** Surface SEM image of  $\text{Cs}_2\text{PbI}_2(\text{SCN})_2$  film casted on a mesoporous  $\text{TiO}_2$  film.



**Figure S7.** GI-out-of-plane and in-plane XRD charts of  $\text{Cs}_2\text{PbI}_2(\text{SCN})_2$  perovskite film.

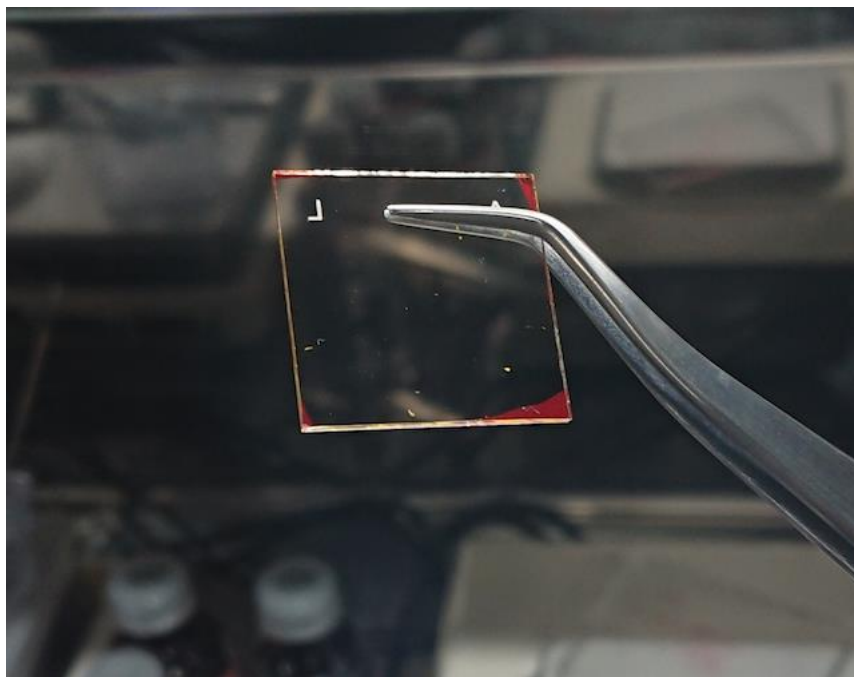


**Figure S8.** Photograph of  $\text{MA}_2\text{PbI}_2(\text{SCN})_2$  perovskite film stored in the GB for 5 days.

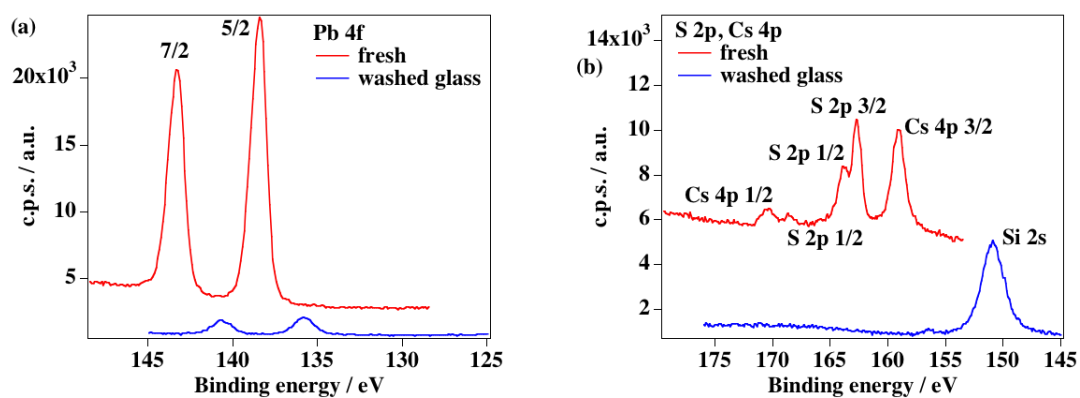


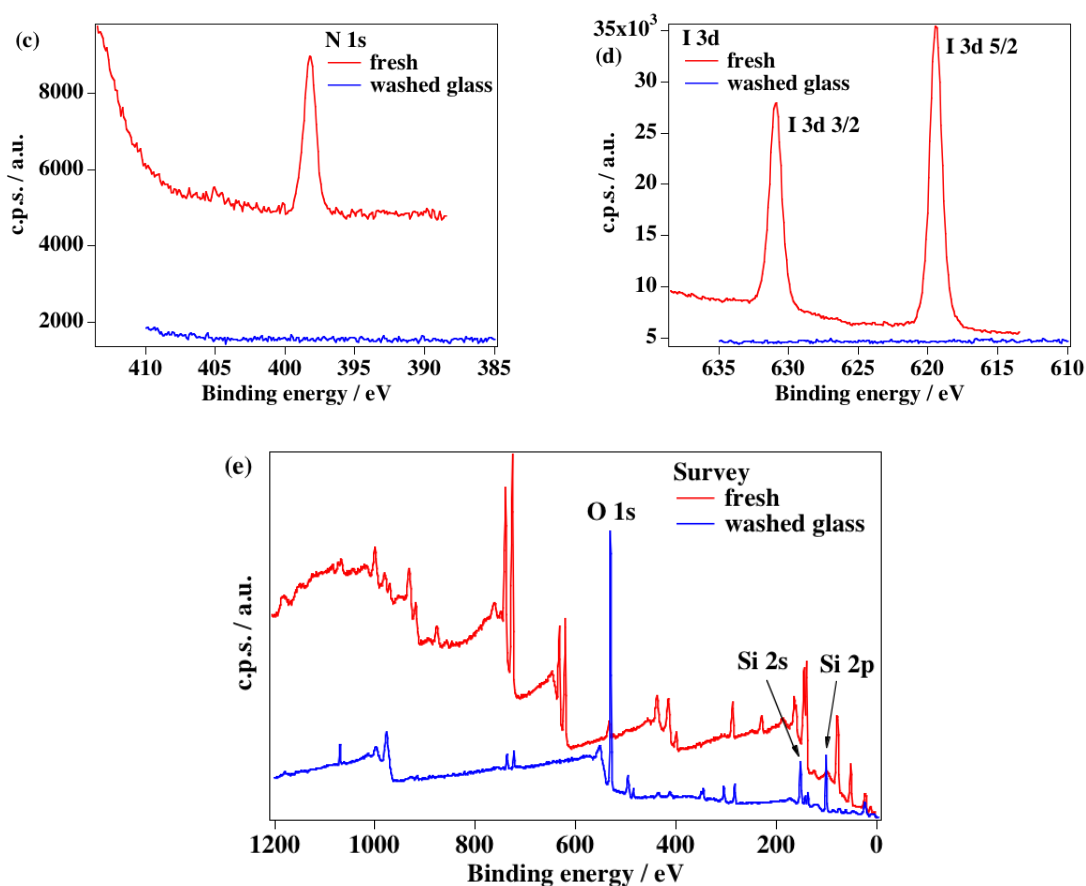


**Figure S9.** Survey XPS spectra of fresh and decomposed  $\text{Cs}_2\text{PbI}_2(\text{SCN})_2$  films.



**Figure S10.** Photograph of a glass substrate after wash off  $\text{Cs}_2\text{PbI}_2(\text{SCN})_2$  perovskite film by water.





**Figure S11.** XPS spectra of (red) fresh  $\text{Cs}_2\text{PbI}_2(\text{SCN})_2$  film and (blue) washed off glass substrate shown in Figure S8. Spectral ranges of (a) Pb 4f, (b) S and Cs, (c) N 1S, (d) I, and (e) survey, respectively.