

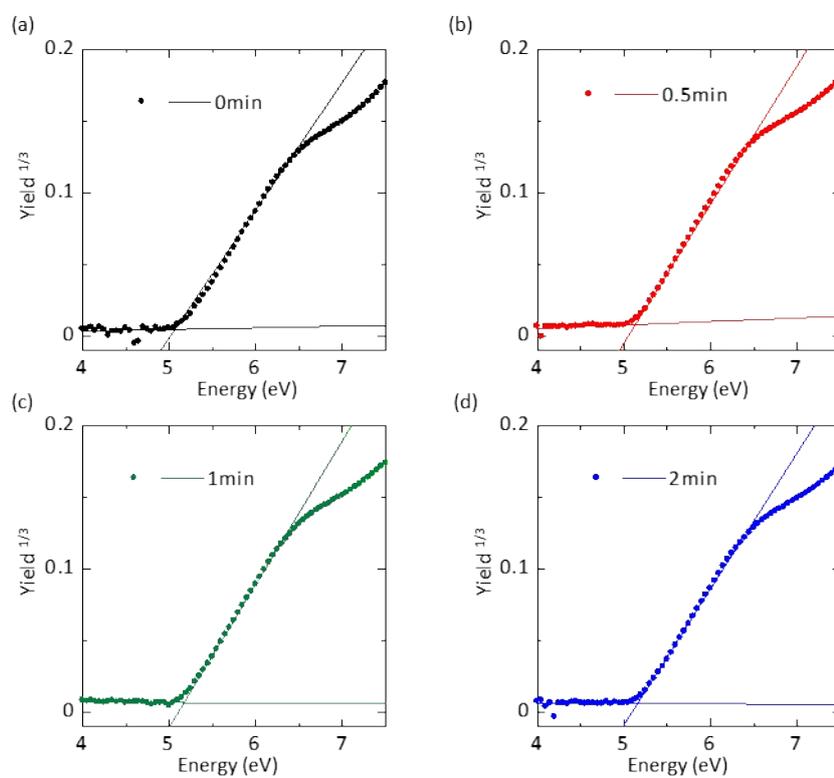
Supplementary Materials

UV ozone treatment for oxidization of Spiro-OMeTAD hole transport layer

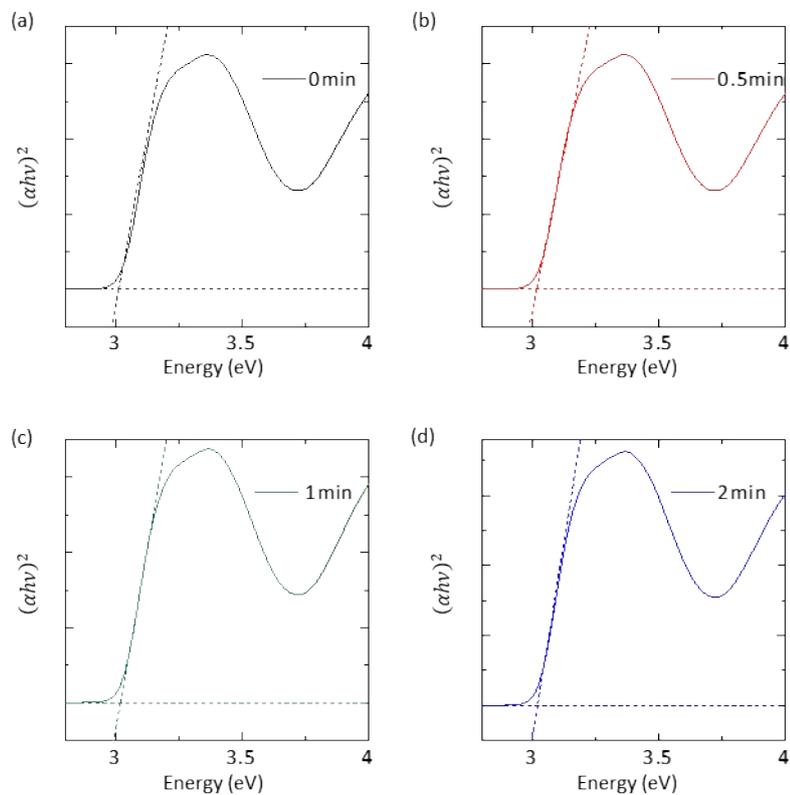
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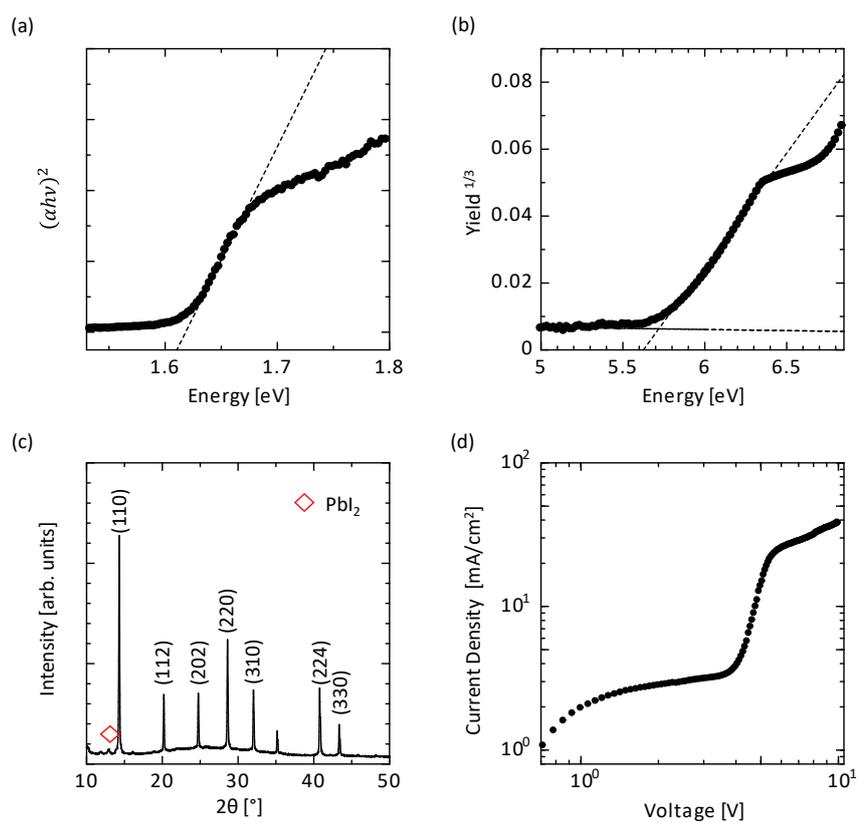
- Figure S1** Photoelectron yield spectra for spiro-OMeTAD films.
- Figure S2** Tauc plot analyses for the absorption spectra of spiro-OMeTAD films.
- Figure S3** Optical, structural, and electrical characteristics of perovskite films.
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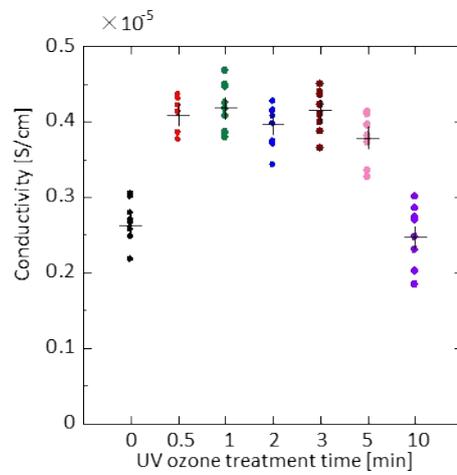
**Figure S1** Photoelectron yield spectroscopy results for spiro-OMeTAD with UV ozone exposure for (a) 0 seconds, (b) 30 seconds, (c) 1 minute, and (d) 2 minutes. The HOMO energy is estimated from the crossing point of two extrapolated lines of experimental data. As shown in Fig. 3 in the main manuscript, the HOMO energy shows a decreased trend with UV ozone treatment time.



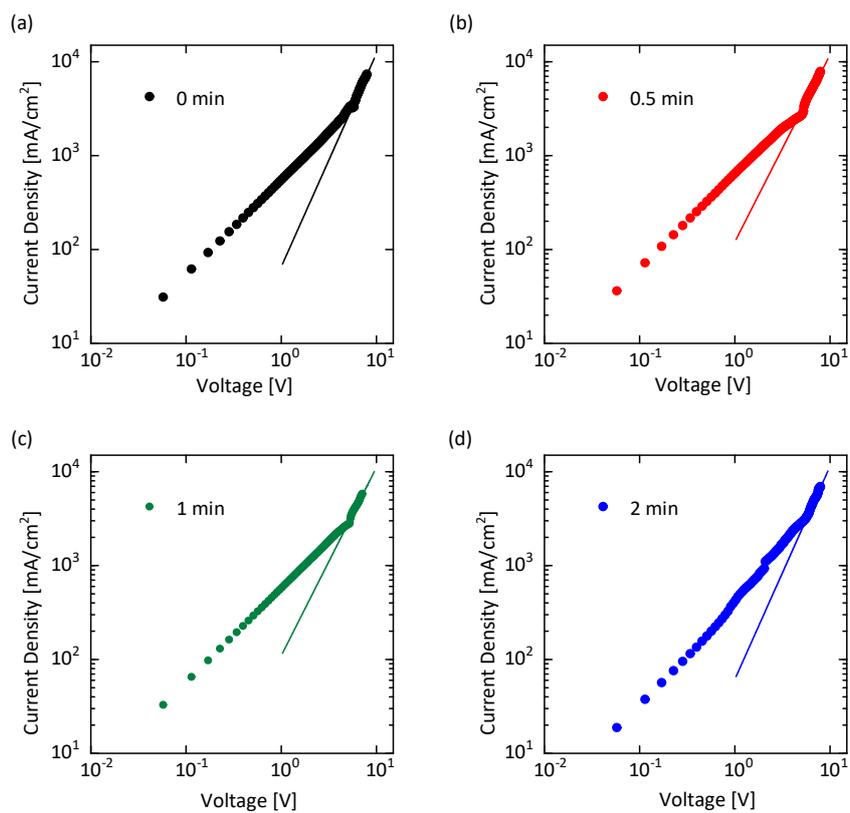
**Figure S2** Tauc plot analyses for the absorption spectra of spiro-OMeTAD with UV ozone exposure for (a) 0 seconds, (b) 30 seconds, (c) 1 minute, and (d) 2 minutes. We assume a direct arrowed transition. The estimated band gap energy does not significant variation against UV ozone treatment time.



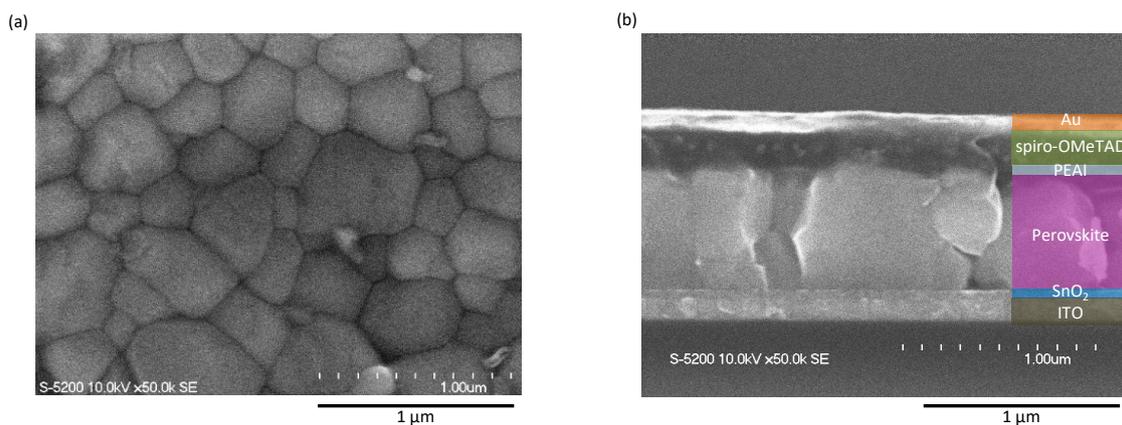
**Figure S3** Fundamental properties of perovskite film fabricated in this study. (a) Tauc plot analyses of absorption spectra. The band gap energy is estimated as  $\sim 1.62$  eV. (b) Photoelectron yield spectra. The valence band maximum energy is evaluated to be  $\sim -5.73$  eV. (c) X-ray diffraction pattern. A polycrystalline film with a cubic crystal structure is confirmed. (d) Current density-voltage profile of a hole-only device with a ITO/PEDOT:PSS/perovskite/spiro-OMeTAD/Au structure, exhibiting ohmic, trap-filling, and SCLC regimes at low-, middle, and high-voltage regions, respectively.



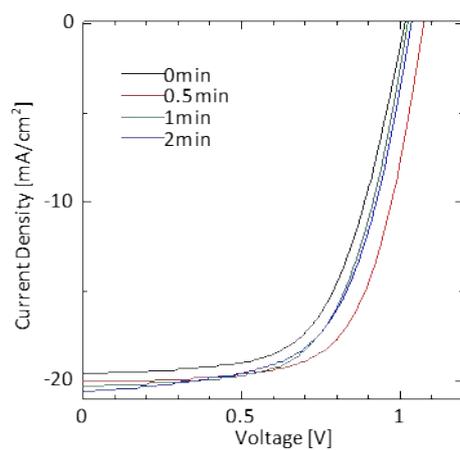
**Figure S4** Conductivity of spiro-OMeTAD films as a function of with UV ozone treatment time (30 s~10 minutes). Closed circles and crosses show experimental results and their mean values, respectively. It is found that the long UV ozone exposure time results in degradation of conductivity.



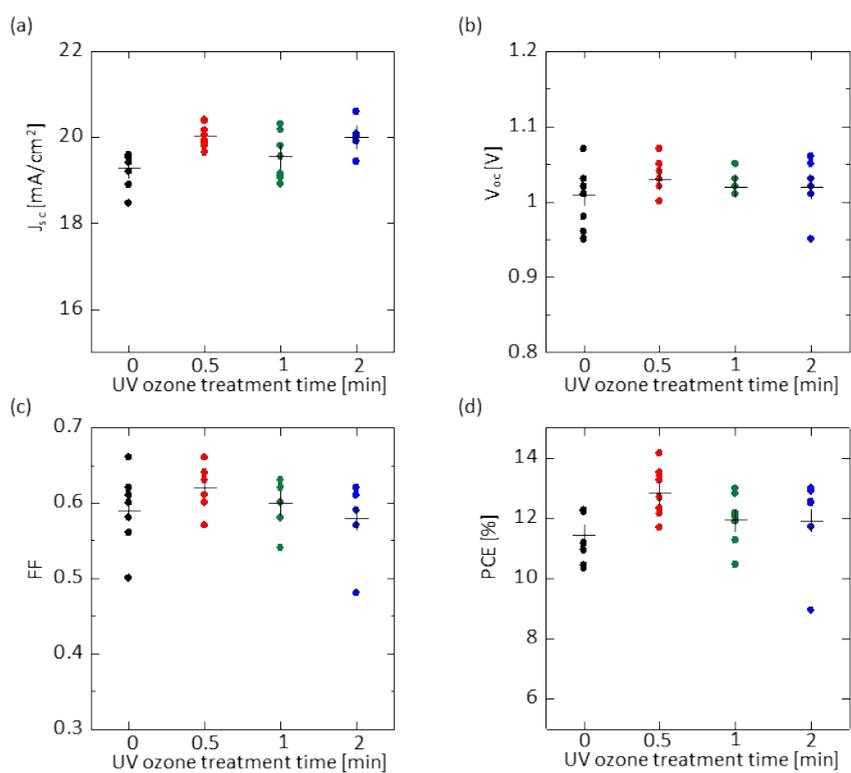
**Figure S5**  $J$ - $V$  characteristics showing space-charge-limited current (SCLC) regime of spiro-OMeTAD films with UV ozone exposure for (a) 0 seconds, (b) 30 seconds, (c) 1 minute, and (d) 2 minutes. The SCLC regime is observed above  $\sim 5$  V. The mobility is evaluated from a quadratic function exhibited by a solid line.



**Figure S6** Scanning electron microscopic (SEM) observations of perovskite solar cell device. (a) Surface SEM image of perovskite layer deposited on a SnO<sub>2</sub>/ITO structure. The film consists of perovskite polycrystal with grain sizes of 200 – 500 nm. (b) Cross-sectional SEM image of the solar cell device. PEAI, oxidized spiro-OMeTAD, and Au layers are deposited onto this surface. The thicknesses of perovskite and spiro-OMeTAD are ~600 and ~250 nm, respectively.



**Fig. S7.** Current density-voltage curves under the AM 1.5G illumination of the best solar cell samples. The best device performance is obtained at 0.5 min of UV ozone treatment time for spiro-OMeTAD hole transport layer.



**Figure S8** Photovoltaic parameters of solar cell devices with UV ozone treated spiro-OMeTAD hole transport layer. (a) Short-circuit current density, (b) open-circuit voltage, (c) filling factor, and (d) power conversion efficiency. Closed circles and crosses show experimental results and their mean values, respectively.