## **Supplementary Materials for**

## Enhanced efficiency and environmental stability of planar perovskite solar cells by suppressing photocatalytic decomposition

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Figure S1. The transmittance of Al-ZnO and RF-ZnO. We can see that the transmittance of ZnO film with Al interlayer has no obvious difference with bare ZnO, and this will minimize the incident light power loss.



Figure S2. (a)The J-V curve and photovoltaic parameters of the S-ZnO control device. Compared with RF-ZnO and Al-ZnO devices, the performance of S-ZnO control device shows a stark drop. This decrease is mainly from the severely surface recombination. (b). The EQE spectra of PSC based on S-ZnO. The Jsc calculated from EQE data is 14.4 mA/cm<sup>2</sup>, which is consisted with the Jsc from the J-V test. The EQE spectra show a response in range of 350 nm - 700 nm. However, compared with the samples based on Al-ZnO and RF-ZnO, the whole spectrum response is significantly decreased.



Figure S3. The J-V curves of best performance PSC based on Al-ZnO measured by forward and reverse scans. As can be seen, the Jsc, FF and PCE obtained from forward scan slightly decreased and small hysteresis between two scan directions was observed.



Figure S4. The steady state photocurrent output of the best performing PSC based on Al-ZnO, measured at the maximum power point (0.824V). The stable photocurrent obtained is  $20.8 \text{ mA/cm}^2$ , and the stable PCE is 17.05%.



Figure S5. The regression of S-ZnO device photovoltaic parameters over time. All the photovoltaic parameters of S-ZnO control device decrease immediately after the start of the aging process. Complete device failure is observed in just 7 hours of aging test.



Figure S6. The UV-VIS absorption of MAPbI<sub>3</sub> on S-ZnO, and inset photos of the film aging over time. The missing absorption corresponding to PbI<sub>2</sub> before aging demonstrates the full conversion of MAPbI<sub>3</sub> precursor. As the increase of aging time, the background absorption increase over time during aging process because the decomposition of MAPbI<sub>3</sub>. At the same time, the appearance of MAPbI<sub>3</sub> film changed obviously, the color of MAPbI<sub>3</sub> film change from dark brown to yellow during aging process, like the photos shown in inset.



Figure S7. The evolution of ideal factor and reverse saturated current density over time.



Figure S8. The SEM of surface and crosection morphology of MAPbI<sub>3</sub> on S-ZnO. As we can see, unlike dense and uniform film on RF-ZnO and Al-ZnO, there are many holes in MAPbI<sub>3</sub> film on S-ZnO. The holes increase the chance of direct contact between ETM and HTM, which will lead the short circuit of the device. And at the same time, the size of MAPbI<sub>3</sub> grain decrease. Small MAPbI<sub>3</sub> grain will increase the series resistance, which is unfavorable to PSCs.



Figure S9. The  $V_{TFL}$  of the electron-only device based on S-ZnO. Higher  $V_{TFL}$  means the high trap density in MAPbI<sub>3</sub> film.



Figure S10. (a) The photoluminescence (PL) spectra and (b) the time-resolved photoluminescence (TRPL) spectra of perovskite films on Al-ZnO, RF-ZnO and S-ZnO. As can be seen, the perovskite film on Al-ZnO shows the weakest spectra intensity and shortest decay time, which mean the quality of perovskite film and electron extraction efficiency were improved by the  $AlO_x$  modification.

	Before Aging				After Aging			
	PCE	Voc(V)	Jsc(mA/cm <sup>2</sup> )	FF	PCE	Voc(V)	Jsc(mA/cm <sup>2</sup> )	FF
S-ZnO	9.2%	1.06	14.4	60%	0.0064%	0.23	1.25	2.24%
RF-ZnO	14.62%	1.05	20.4	68.25%	7.07%	1.00	14.9	47.43%
Al-ZnO	17.17%	1.053	21.55	75.91%	15.03%	1.034	21.15	68.72%

Table S1. Detail photovoltaic data of Al-ZnO, RF-ZnO and S-ZnO devices before and after aging.