Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Interfacial defect passivation by diethyl phosphate salts for high-efficiency and stable perovskite solar cells

Xuan Sha,[ab] Jiang Sheng,*[b] Weichuang Yang,[b] Jingsong Sun,[bc] Chunhui Shou,[c] Luyan Zhang,[b] Ningjun Zhang,[b] Zhiqin Ying,[b] Xi Yang,[b] Hongbin Zhao,*[a] Jichun Ye*[b]

[a] Department of Chemistry & Institute for Sustainable Energy/College of Sciences, Shanghai University, Shanghai 200444, China

[b] Zhejiang Provincial Engineering Research Center of Energy Optoelectronic Materials and Devices, Ningbo Institute of Materials Technology & Engineering, Chinese Academy of Sciences, Ningbo 315201, China

[c] Key Laboratory of Solar Energy Utilization and Energy Saving Technology of Zhejiang Province, Zhejiang Energy Group R&D Institute Co., Ltd, Hangzhou, Zhejiang 310003, China

*Corresponding Authors: shengjiang@nimte.ac.cn; hongbinzhao@shu.edu.cn; jichun.ye@nimte.ac.cn

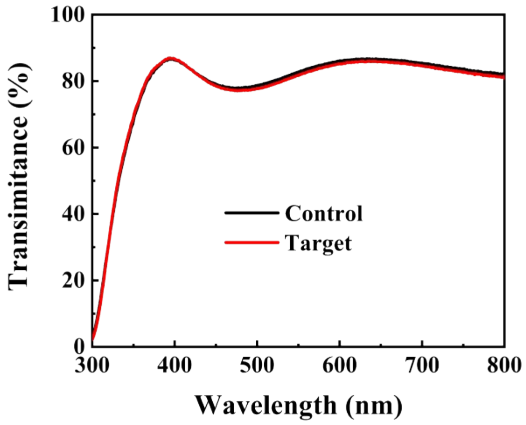

Fig. S1 The transmittances of the SnO_2 ETLs and EMIM DEP-SnO_2 deposited on ITO glasses. Obviously, the transmittance is almost similar, which means the EMIM DEP is free of negative impact on transmittance on the SnO_2 film.

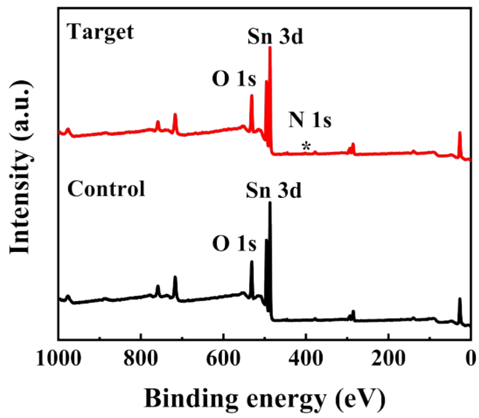

Fig. S2 XPS full spectra of SnO_2 film with and without EMIM DEP. Compared to the spectrums, both have the peak of Sn 3d and O 1s. Compared with the SnO_2 full-spectrum, SnO_2/EMIM DEP has an additional N 1s peak and a weak peak of P 2p.

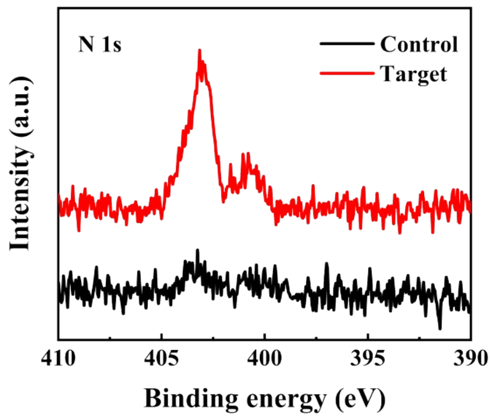

Fig. S3 N fine spectra of XPS of SnO_2 film with and without EMIM DEP. Compared with the N 1s peak of the control SnO_2 film, the EMIM DEP SnO_2 film has an obvious N 1s peak.

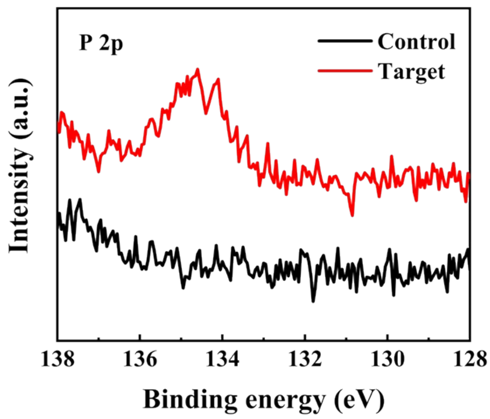

Fig. S4 P fine spectra of XPS of SnO_2 film with and without EMIM DEP. Compared with the P 2p peak of the SnO_2 film, the EMIM DEP-SnO_2 film has an obvious P 2p peak.

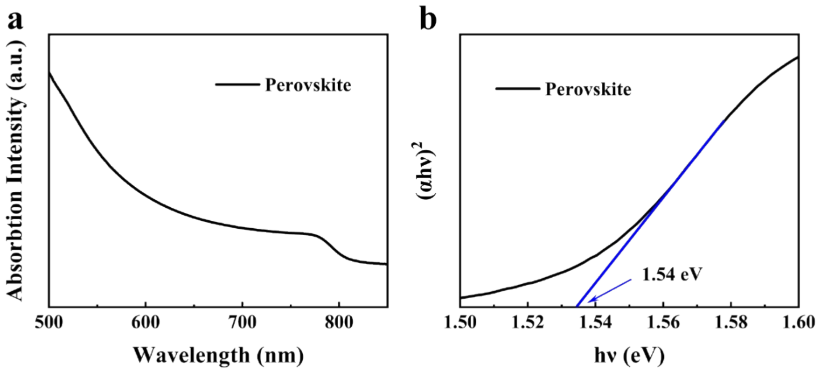

Fig. S5 (a) Absorbance spectra for the perovskite layer. (b) Tauc plot for the perovskite layer. We obtained the band gap of the active layer from the absorption curve by the tangent method. The Tauc curve was obtained using 1240/λ as the horizontal coordinate and (1240*A/λ) ^2 as the vertical coordinate. The tangent line was plotted at the steepest point, and the intersection with the X-axis was the Eg of the active layer.

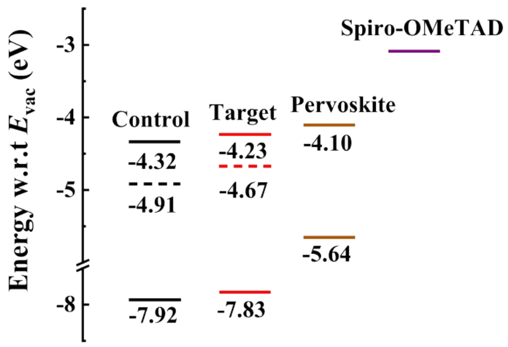

Fig. S6 Energy level diagram of SnO_2 film with and without EMIM DEP treatment. After the modification of EMIM DEP, the conduction band becomes shallower, closer to the conduction band of the perovskite layer, and the interfacial transport becomes stronger. The band gap of SnO_2 used in the energy level calculation is 3.6 eV, and the band gap of perovskite in the energy level calculation is 1.54 eV. The calculation process is as followed:

The EF is calculated as:

$$E_F = E_{cutoff} - h\nu \tag{1}$$

The VB is calculated as:

$$VB = E_F - E_{FT} \tag{2}$$

The CB is calculated as:

$$CB = VB + E_g \tag{3}$$

E_F means Fermi level, hv is 21.22 eV, E_{cutoff} is cut-off edge tangent value, E_{FT} is Fermi edge tangent value, VB means valence band, and CB means conduction band.

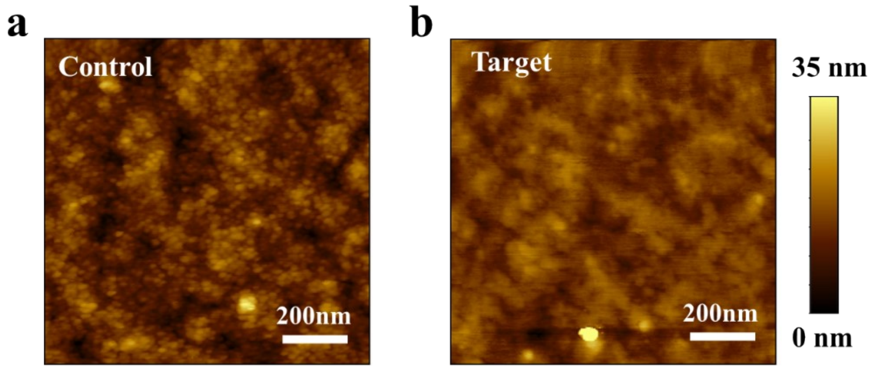

Fig. S7 AFM images of SnO_2 film with and without EMIM DEP. the treated tin oxide film R_q is 1.45 nm, compared with the control sample (Rq = 1.61 nm), the surface roughness of EMIM DEP-SnO_2 becomes smaller, which can provide more nucleation sites to be beneficial to the film formation of perovskite.

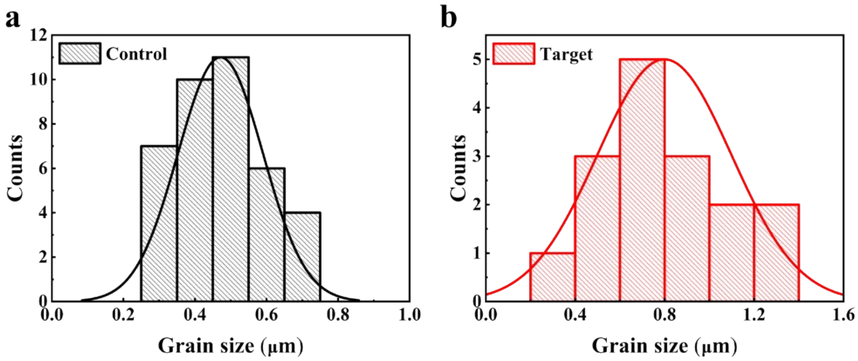

Fig. S8 Number distribution of perovskite grain size in SEM with and without EMIM DEP. The average size of perovskite grains in the control group is 470 nm, while the average size of EMIM DEP-based perovskite grains increases to 810 nm.

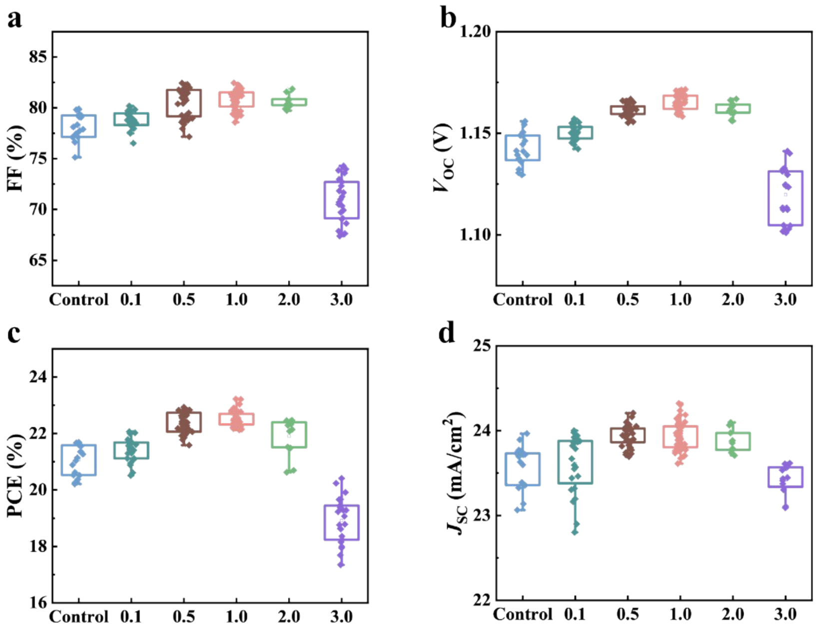

Fig. S9 Distribution of FF, V_{OC}, PCE, and J_{SC} of devices with different concentrations of EMIM DEP. When the content of EMIM DEP is 1mg/mL, the photovoltaic performance is best.

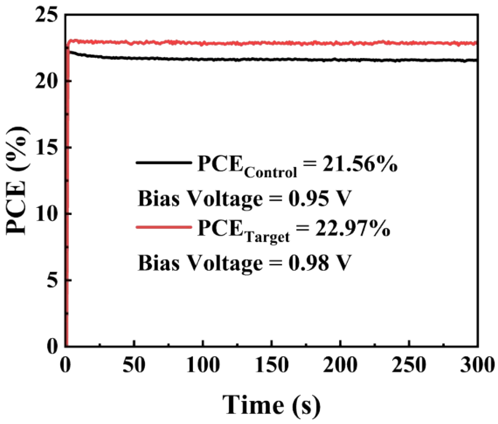

Fig. S10 Steady-state output (SPO) of the photocurrent of the devices for 300 s at the maximum power point (MPP). The output power of the EMIM DEP-based device is stable at 22.97% while gradually decreasing output power is noticed for the control device, resulting in an enhancement of the operating stability.

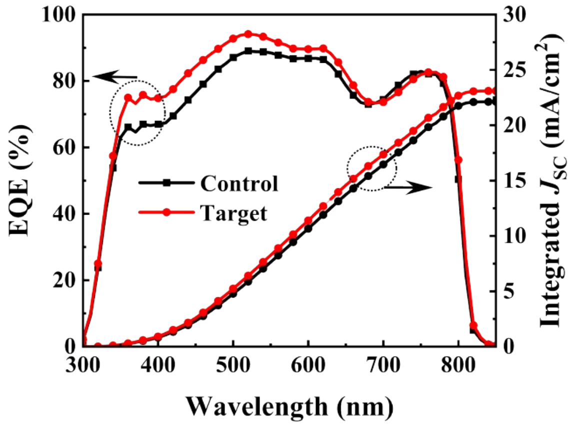

Fig. S11 The external quantum efficiency (EQE) spectrum of the corresponding devices. The integrated current density of the SnO_2 and SnO_2/EMIM DEP-based devices is 22.13 and 23.10 mA/cm^2 (the EQE test has a 5% error with the *I*-*V* test instrument).

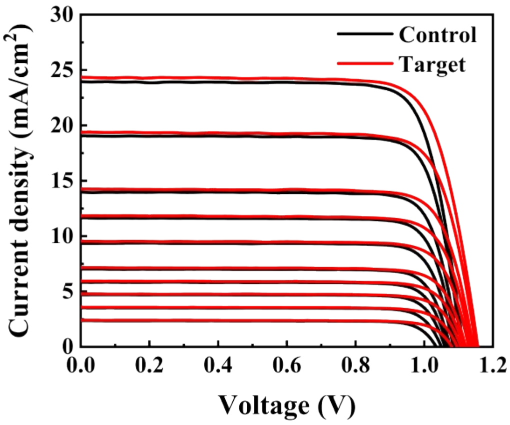

Fig. S12 *J*-*V* curves of the devices with and without EMIM DEP treatment at different light intensities (0.999 sun, 0.798 sun, 0.587 sun, 0.489 sun, 0.395 sun, 0.296 sun, 0.246 sun, 0.198 sun, 0.149 sun, 0.101 sun.) respectively.

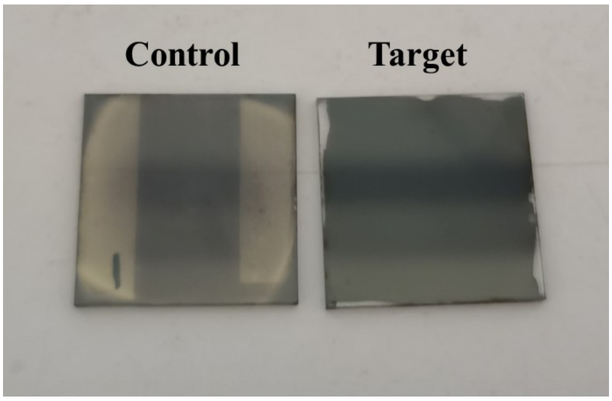

Fig. S13 Photos of the perovskite films after 7 days in ambient air with 70% ~ 80% relative humidity.

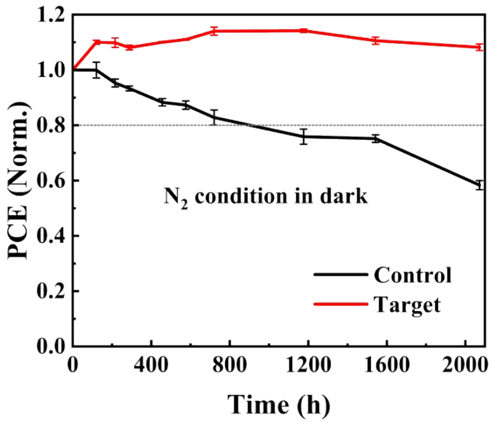

Fig. S14 Normalized PCE evolution of the unencapsulated devices based on SnO_2 and SnO_2/EMIM DEP stored in N_2 ambient. The PCE of the EMIM DEP-based device remains 2072 h without degradation.

Table S1. The energy level parameters of SnO_2 with and without EMIM DEP treatment.

	E_{cutoff} (eV)	E_{CB} (eV)	E_{VB} (eV)	E_{fermi} (eV)
Control	-16.53	-4.32	-7.92	-4.91
Target	-16.77	-4.23	-7.83	-4.67

Table S2. The fitted data of TrPL curves.

	A_1	A_2	τ_1 (ns)	τ_1 (ns)
Control	0.38	0.20	101	643
Target	0.14	0.58	48	1013

Table S3. The fitted data of EIS tests.

	R_s (Ω)	R_{ct} (Ω)	R_{rec} (Ω)
Control	28.66	481.60	47.90
Target	21.65	341.60	100.90