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Supplementary Information

Alkylammonium passivation for 2D tin halide perovskite field-effect transistors

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Figure S1. Tauc plots of (PEA)₂SnI₄ thin films with and without additives (PAI and PDAI₂).



Figure S2. SEM images of (a) pristine (PEA)₂SnI₄ films, (b) (PEA)₂SnI₄ film with PAI, and (c) (PEA)₂SnI₄ film with PDAI₂ (scale bar: $10 \mu m$).



Figure S3. Non-contact mode atomic force microscope (AFM) images of (a) pristine (PEA)₂SnI₄ films, (b) (PEA)₂SnI₄ film with PAI, and (c) (PEA)₂SnI₄ film with PDAI₂, along with their corresponding height profiles in (d), (e), (f), respectively.



Figure S4. SCLC measurements of hole-only devices based on (PEA)₂SnI₄ thin films with and without additives (PAI and PDAI₂).



Figure S5. UV photoelectron spectroscopy (UPS) analysis. (a) Secondary electron cut-off region of (PEA)₂SnI₄ film with and without additives (PAI and PDAI₂). (b) Onset region of (PEA)₂SnI₄ film with and without additives (PAI and PDAI₂). (c) Energy band structures of (PEA)₂SnI₄ film with and without additives (PAI and PDAI₂).



Figure S6. Transfer characteristics $(V_G - \sqrt{|I_D|})$ of (PEA)₂SnI₄ FETs with and without additives (a) Pristine (b) PAI added (c) PDAI₂ added. The green dashed lines represent the slope used to calculate the claimed mobility ($\mu_{claimed}$) under the forward scan.



Figure S7. Transfer curves of 30 different (PEA)₂SnI₄ FET devices with PDAI₂ ($V_D = -40$ V) (forward scan).



Figure S8. Maximum trap density within (PEA)₂SnI₄ FET devices with and without additives (PAI, PDAI₂, MAI, and BAI).



Figure S9. Transfer characteristics of (PEA)₂SnI₄ FET devices as a function of additive concentration. (a) PAI. (b) PDAI₂.



Figure S10. Bias-stress stability of (PEA)₂SnI₄ FETs with and without additives (PAI and PDAI₂) for over 2000 s under constant bias condition ($V_{\rm G} = -40$ V and $V_{\rm D} = -40$ V).



Figure S11. (a) Bond lengths of Sn–axial I in 2D perovskites with the addition of MA, EA, PA, BA, MDA, EDA, PDA, and BDA cations. (b) Schematic figure of axial I in the inorganic framework. The axial I is represented by red square.

Note 1. Calculations of mobility values of (PEA)₂SnI₄ FETs

In the saturation region of a FET device, threshold voltage (V_{th}) and field-effect mobility (μ_{FE}) can be calculated using the following equations.[1]

$$I_{\text{D,sat}} = \frac{\mu_{\text{FE,sat}} W C_{\text{i}}}{2L} (V_G - V_{th})^2, |V_G - V_{th}| < |V_{DS}|$$
$$\mu_{FE,\text{sat}} = \frac{2L}{W C_{\text{i}}} \left(\frac{\partial \sqrt{|I_D|}}{\partial V_G}\right)^2$$

where $V_{\rm G}$ and $I_{\rm D}$ are the source-to-gate voltage and the source-to-drain currents, respectively. And *L*, *W*, *C*_i represent the channel length, width, and unit capacitance of the oxide dielectric layer, respectively.

Due to nonidealities such as contact effects, charge trapping, and ion migration, careful evaluation is required for accurate mobility extraction. To minimize potential errors in mobility assessment, we calculated the reliability factor (r_{sat}), along with the effective mobility (μ_{eff}).[2]

$$r_{\text{sat}} = \left(\frac{\sqrt{|I_{\text{D}}|^{\text{max}}} - \sqrt{|I_{\text{D}}^{\text{o}}|}}{|V_{\text{G}}|^{\text{max}}}\right)^2 / \left(\frac{\partial\sqrt{|I_{\text{D}}|}}{\partial V_{\text{G}}}\right)^2_{\text{claimed}}$$

 $\mu_{\rm eff} = r_{\rm sat} \times \mu_{\rm claimed}$

where $|I_D|^{\text{max}}$ and $|I_D^0|$ are the maximum source-to-drain current at the maximum gate voltage $(|V_G|^{\text{max}} = -40 \text{ V})$ and at $V_G = 0 V$. The effective mobility values and the reliability factor for (PEA)₂SnI₄ FETs, with and without additives (PAI and PDAI₂), are summarized in Table S1.

		Pristine	PAI	PDAI ₂
$\frac{\mu_{\text{claimed}}}{(\text{cm}^2\text{V}^{-1}\text{s}^{-1})}$	Forward	0.62	1.37	2.60
	Reverse	0.86	2.46	3.31
r _{sat} (%)	Forward	12.1	54.6	76.1
	Reverse	13.8	38.5	45.8
$\mu_{\text{effective}}$ (cm ² V ⁻¹ s ⁻¹)	Forward	0.08	0.75	1.98
	Reverse	0.12	0.95	1.52

Table S1. Evaluation of field-effect mobility values for (PEA)₂SnI₄ FETs with and without additives (PAI and PDAI₂). Data are obtained from the highest-performing device under each condition.

References in Supplementary Information

1. S. M. Sze and K. K. Ng, *Physics of Semiconductor Devices*, 3rd edn, John Wiley & Sons, 2007.

2. H. H. Choi, K. Cho, C. D. Frisbie, H. Sirringhaus and V. Podzorov, *Nat. Mater.*, 2018, **17**, 2–7, DOI: 10.1038/nmat5035.