Supporting Information

Using Ligand Engineering to Produce Efficient and Stable Pb-Sn Perovskite Solar Cells with Antioxidative 2D Capping Layers

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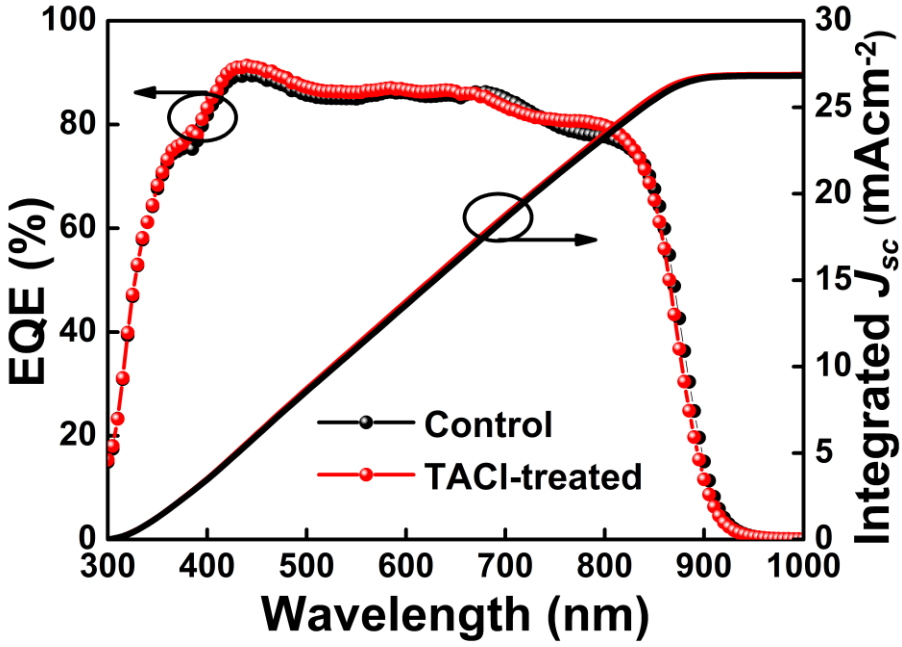

Figure S1. The bandgap is determined to be 1.38 eV using the Tauc plot in Figure 2a and 1.34 eV using the external quantum efficiency (EQE) spectra. The integrated current densities shown in Table S3 agree well with the J_{sc}s obtained by J-V measurement (derivation below 1%).

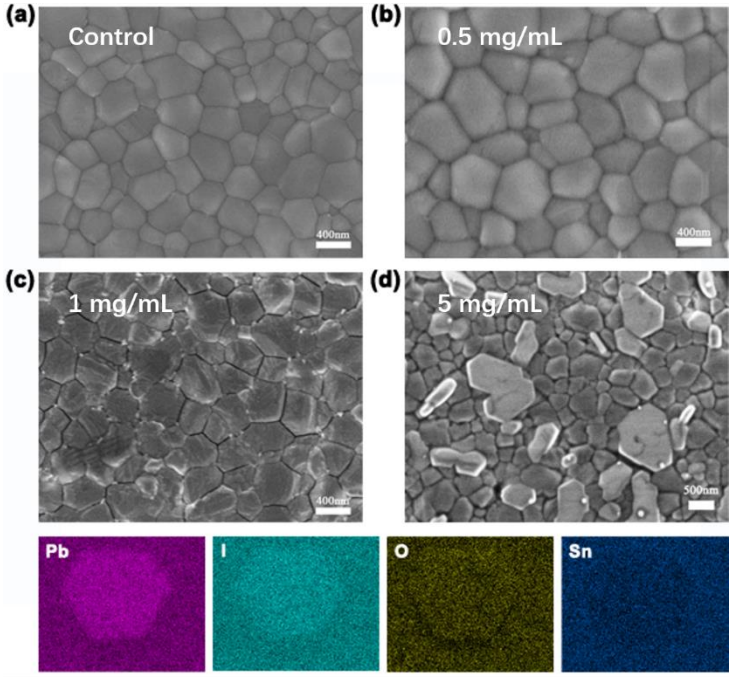

Figure S2. a-d) the surface SEM images of $FA_{0.85}MA_{0.15}Pb_{0.7}Sn_{0.3}I_{2.55}Br_{0.45}$ film treated with different TACl solution. From a-d we can see that the TACl treatment at low concentration (0.5 and 1 mg mL^{-1}) didn't change the perovskite surface morphology obviously while high

concentration (5mg mL^{-1}) TACl induced a large amount of hexagonal flake structures on the perovskite surface. The bottom 4 images are the EDS mapping of the hexagon region emerged after TACl treatment, supporting the formation of TA based 2D perovskite.

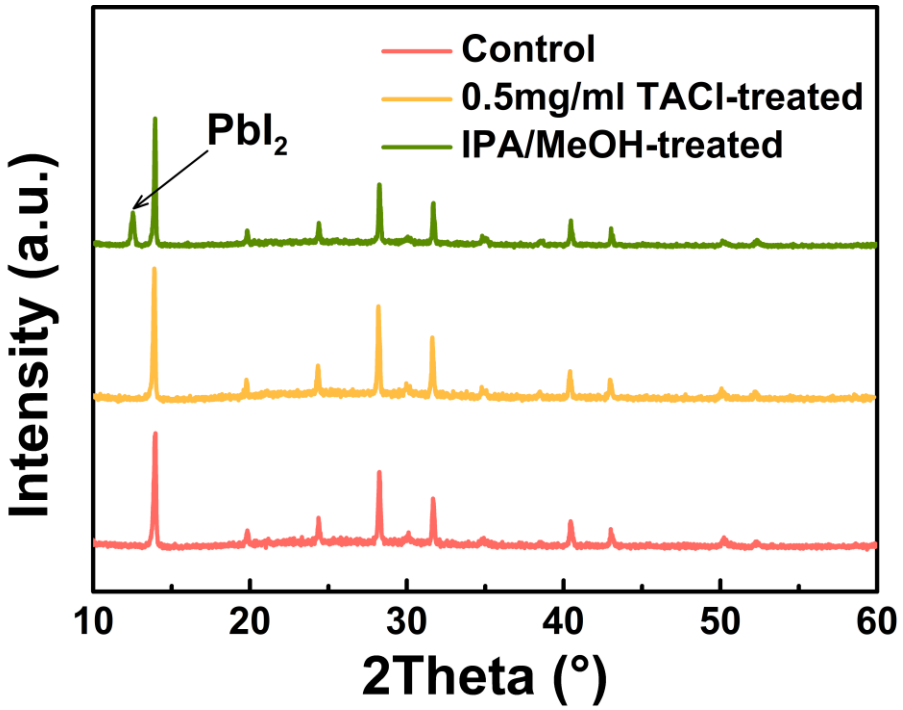

Figure S3. The XRD of Pb-Sn based perovskite film without and with TACl treatment. Due to the limited solubility of TACl in IPA solvent, the 5mg mL^{-1} TACl solution needs to be prepared using IPA/Methanol (10% vv^{-1}) mixed solvent. The green curve is the XRD result of perovskite film treated by IPA/MeOH.

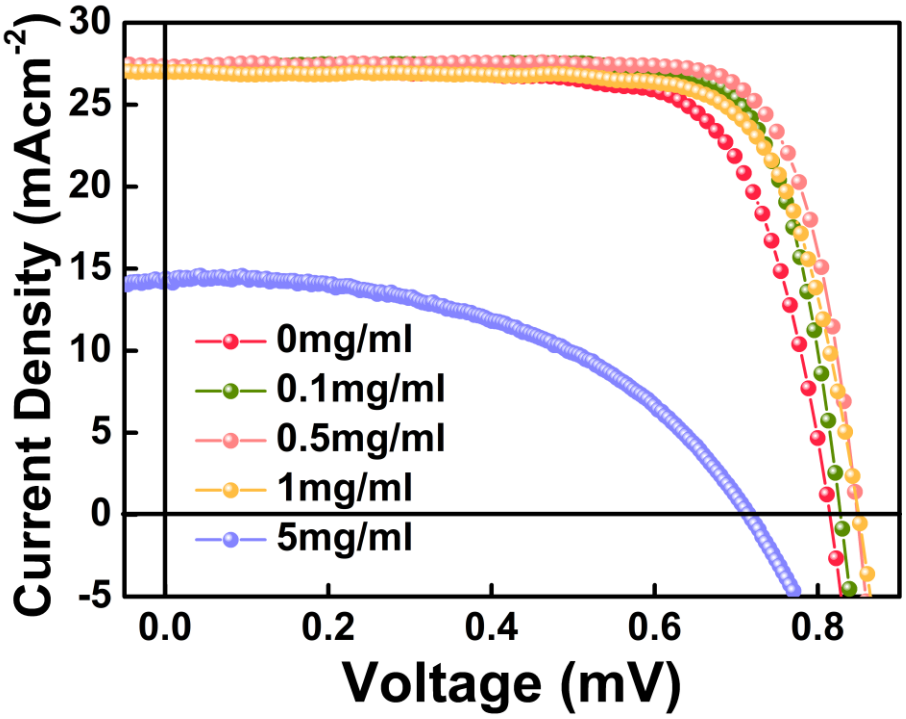

Figure S4. JV curves of Pb-Sn based perovskite solar cells with different concentration TACl treatment.

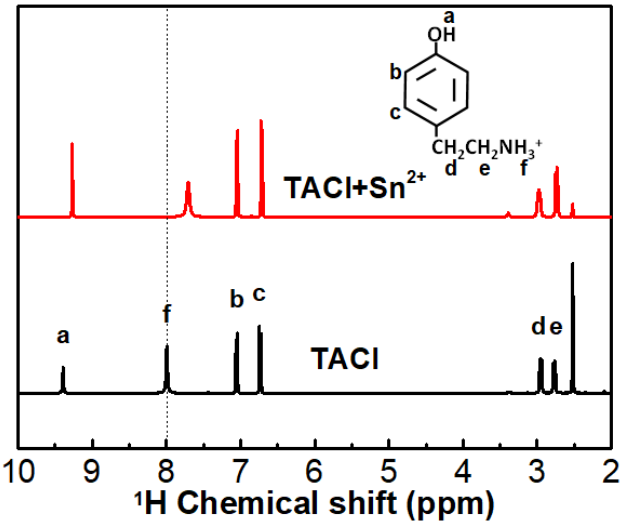

Figure S5. ^{1}HNMR of TACl and TACl-Sn^{2+} mixture in D_6-DMSO solvent.

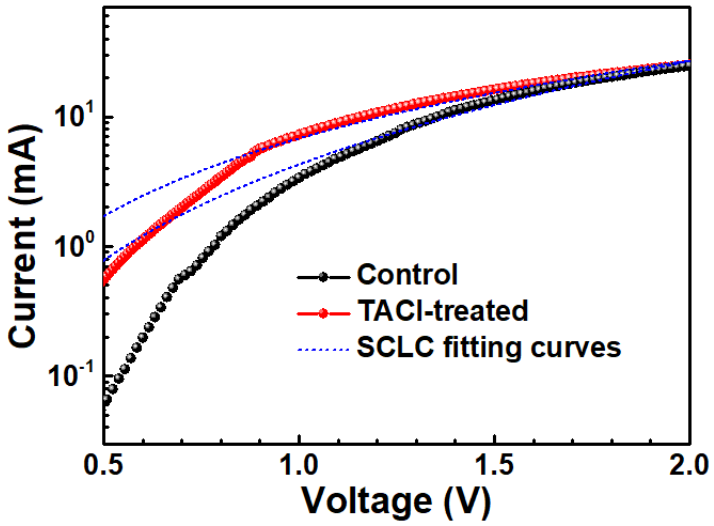

Figure S6. Dark I-V curves and the SCLC fitting curves of hole only device based on control and TACl-treated Pb-Sn perovskite film.

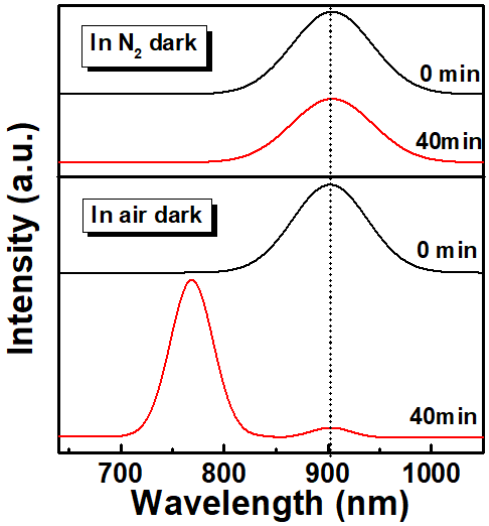

Figure S7. PL spectra of control perovskite films stored under air and N_2 conditions (in dark) for 40 min, respectively.

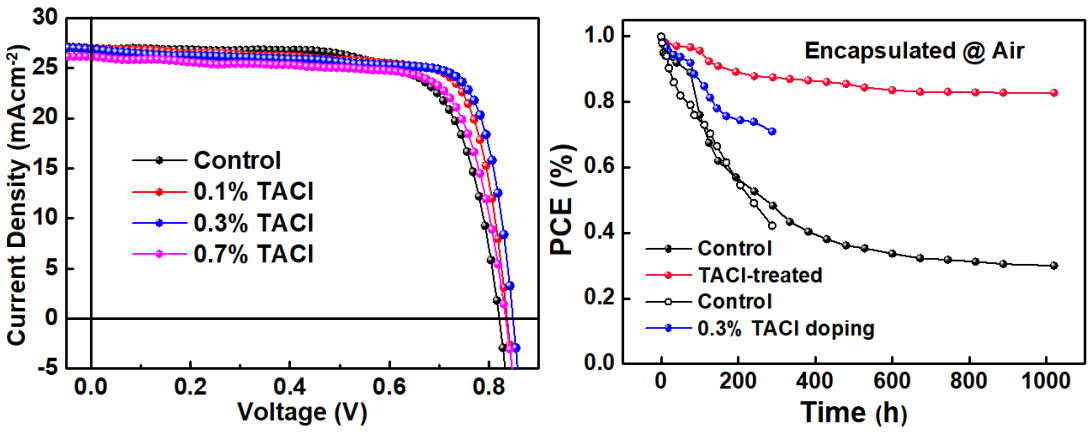

Figure S8. (a) *J-V* curves of Pb-Sn based perovskite solar cells with different concentration TACl precursor doping. (b) Shelf stability of encapsulated solar cells based on control, TACl post-treated and precursor doping .

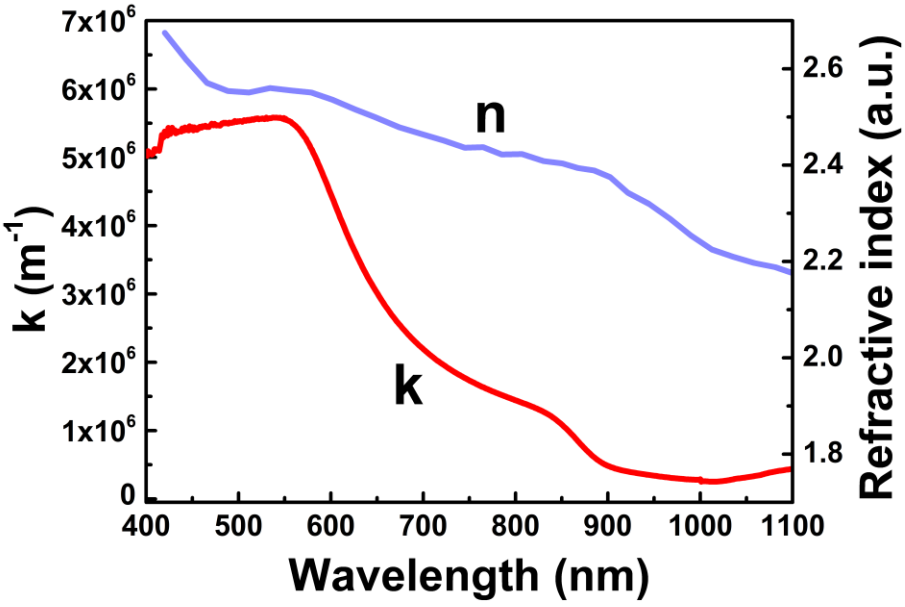

Figure S9. Optical parameters of Pb-Sn mixed perovskite layer.

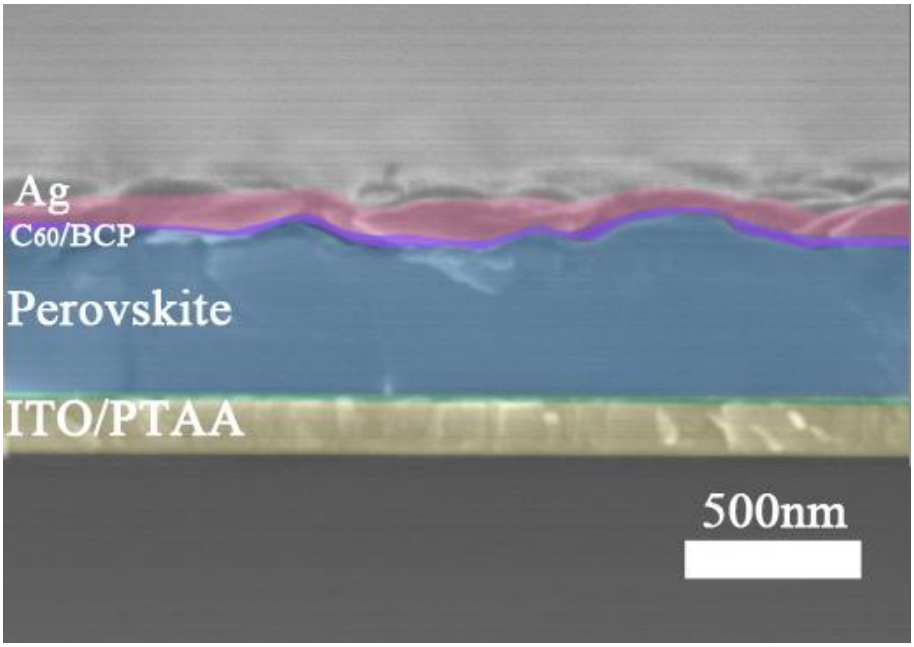

Figure S10. Cross-sectional SEM image of the entire perovskite solar cells.

Table S1. The TRPL fitting results.

	Y_0 (Counts)	A_1 (%)	τ_1 (ns)	A_2 (%)	τ_2 (ns)
Control	23.5	8.2	13.6	91.8	454.6
TACl-treated	34.8	6.4	25.0	93.6	581.2

Table S2. Photovoltaic parameters of devices based on perovskite treated with different concentration TACl.

TACl concentration (mg mL^{-1})	0	0.1	0.5	1	5
J_{sc} (mA cm^{-2})	27.14	27.22	27.36	27.05	14.57
V_{oc} (V)	0.82	0.83	0.85	0.85	0.72
FF	0.71	0.78	0.78	0.74	0.48
PCE (%)	15.77	17.56	18.14	17.01	5.05

Table S3. Photovoltaic parameters of the best-performing control and TACl-treated devices extracted from Figure 6a.

	V_{oc}(V)	J_{sc}(mA cm^{-2})	*FF* (%)	*PCE* (%)	Integrated J_{sc} (mA cm^{-2})	hysteresis
Control (forward)	0.82	27.10	73.1	16.25	26.38	0.159
Control (reverse)	0.79	27.29	62.6	13.66		
TACl-treated (forward)	0.85	27.36	78.6	18.28	26.70	0.010
TACl-treated (reverse)	0.85	27.40	77.7	18.10		

Table S4. Photovoltaic parameters of devices based on perovskites with different concentration TACl precursor doping.

Perovskite	V_{oc} (V)	J_{sc} (mA/cm^2)	FF	PCE (%)
Control	0.82	26.98	0.72	15.93
0.1% TACl	0.84	26.94	0.77	17.42
0.3% TACl	0.85	27.02	0.78	17.87
0.7% TACl	0.84	26.18	0.74	16.33

Table S5. The device parameters extracted from the gpvdm simulation.

Parameter	Control device	TACl-treated device	Units
Charge carrier mobility	2.0×10^{-06}	3.7×10^{-06}	$m^2V^{-1}s^{-1}$
Trap density	4.6×10^{23}	1.5×10^{23}	$m^{-3}eV^{-1}$
Free electron to Trapped electron	5.3×10^{-19}	8.9×10^{-20}	m^{-2}
Trapped electron to Free hole	1.7×10^{-19}	3.5×10^{-20}	m^{-2}
Shunt resistance	1.1×10^{03}	4.1×10^{03}	$Ohmsm^2$
Series resistance	53.252	32.763	*Ohms*
Contact charge density	4.3×10^{24}	4.7×10^{24}	m^{-3}
Photon efficiency (η)	0.753	0.773	*0-1*