

Supplementary Information for

Minimizing Interfacial Energy Losses via Multifunctional Cage-like

Diammonium Molecules for Efficient Perovskite/Silicon Tandem Solar

Cells

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Yihan Sun¹, Meili Zhang¹, Xuchao Guo¹, Yuheng Zeng¹, Xi Yang^{1,2,*}, Jichun Ye^{1,2,*}

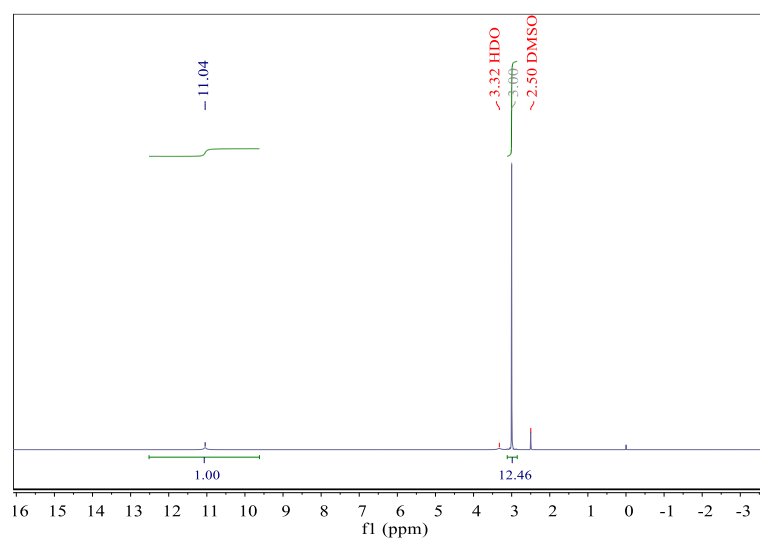
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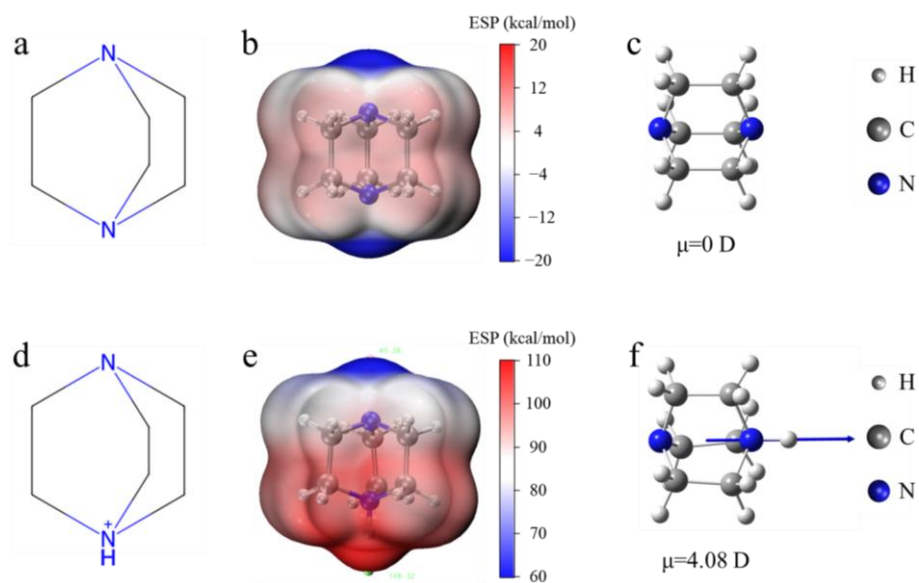
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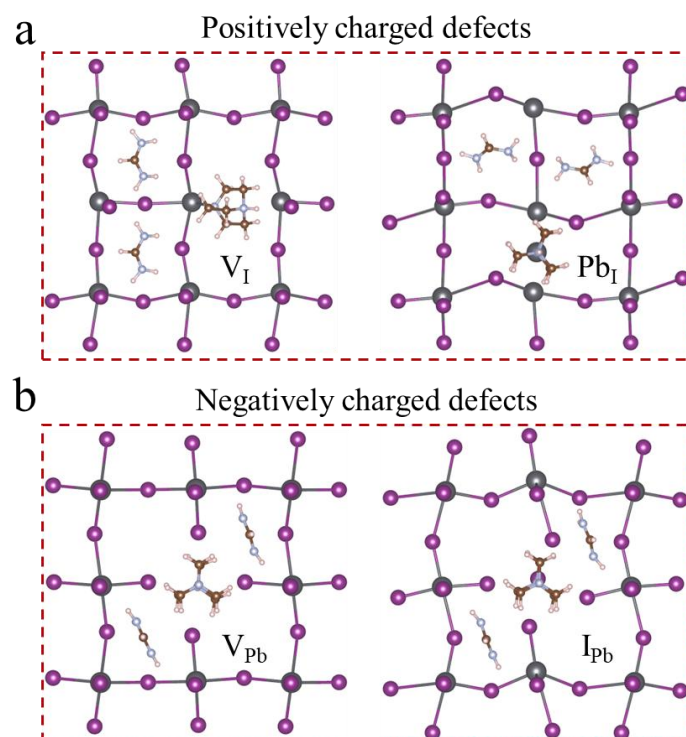
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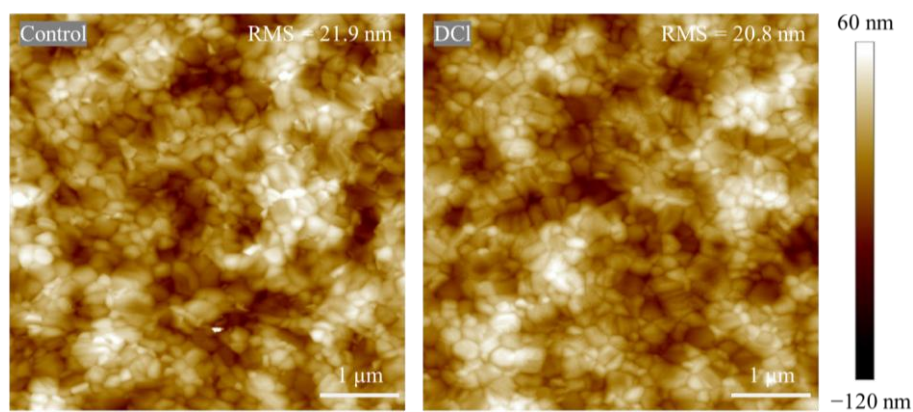
Supplementary Fig. 1 ¹H NMR characterisation of DCI after synthesis.



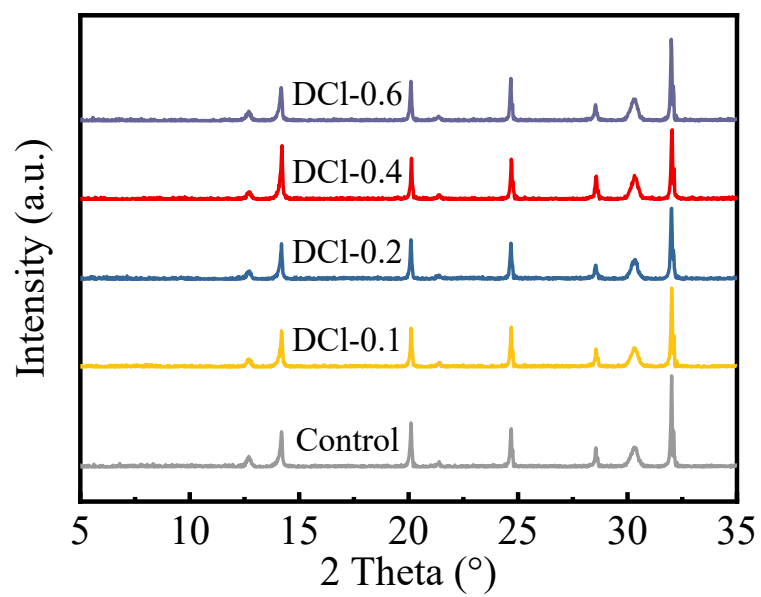
Supplementary Fig. 2 (a, d) Chemical structures, (b, e) electrostatic potentials (ESPs) and (c, f) corresponding electric dipole moments of both (a-c) 1,4-diazabicyclo[2.2.2]octane and (d-f) cage-like diammonium cation. Light gray, dark gray and blue balls represent the H, C, and N atoms, respectively.



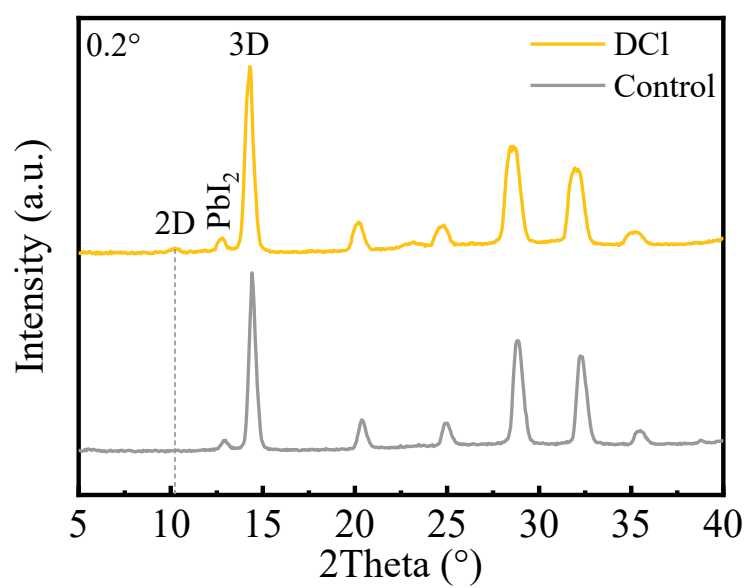
Supplementary Fig. 3 Top-view of various types of defects passivated by DCl. (a) Positively charged defects. (b) Negatively charged defects. Pink, light brown, light gray, violet, and gray balls represent the H, C, N, I and Pb atoms, respectively.



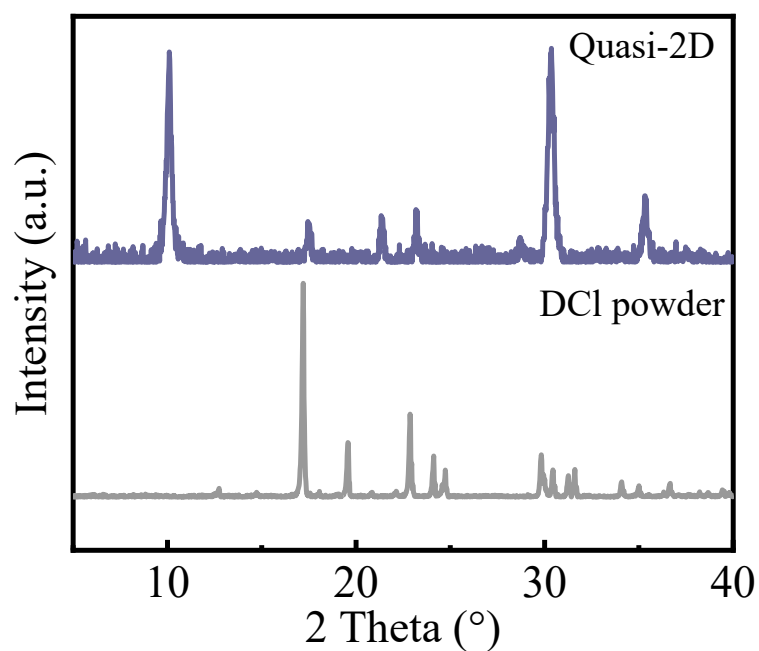
Supplementary Fig. 4 AFM images of control and DCl-treated perovskite films with a scan area of $5\ \mu\text{m} \times 5\ \mu\text{m}$.



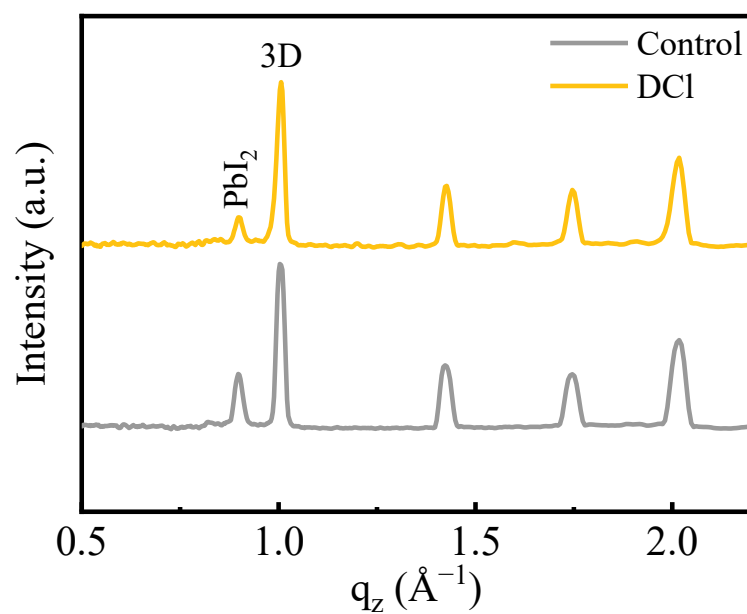
Supplementary Fig. 5 XRD patterns of the control and DCl-treated perovskite films.



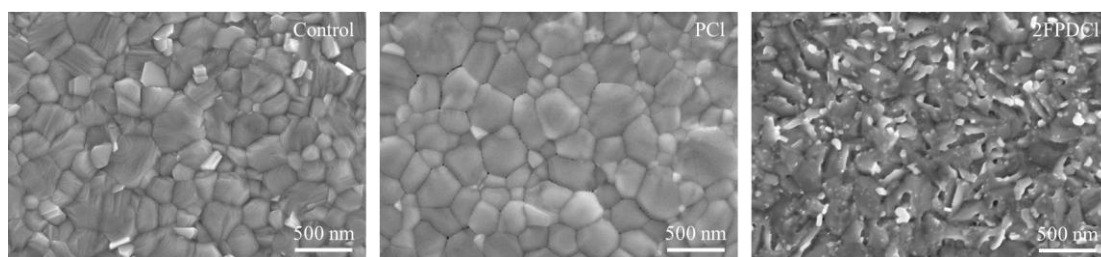
Supplementary Fig. 6 GIXRD patterns of the control and DCI-treated perovskite films with a grazing incidence angle of 0.2°.



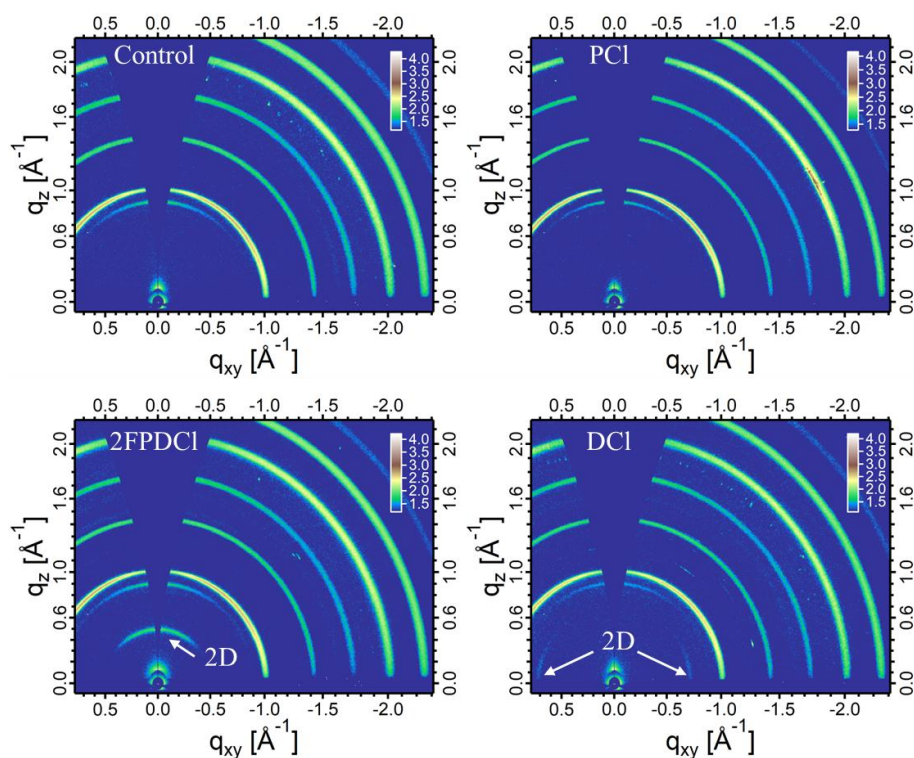
Supplementary Fig. 7 XRD patterns of DCl powder and synthesized single-crystal 2D perovskite. Notably, small grains on the DCl-treated perovskite surface (Fig. 1a and b) should be ascribed to the formation of a 2D phase rather than the post-treated DCl material itself, as evidenced in Supplementary Fig. 7. The peak position at 10.2° of the synthesized single-crystal 2D perovskite precisely matches that observed in the GIXRD measurement (Supplementary Fig. 6).



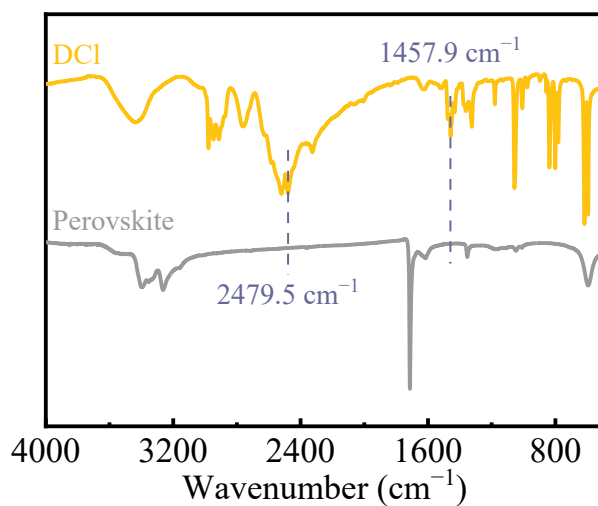
Supplementary Fig. 8 The (001) out-of-plane line cuts of GIWAXS images of the control and DCl-treated perovskite films.



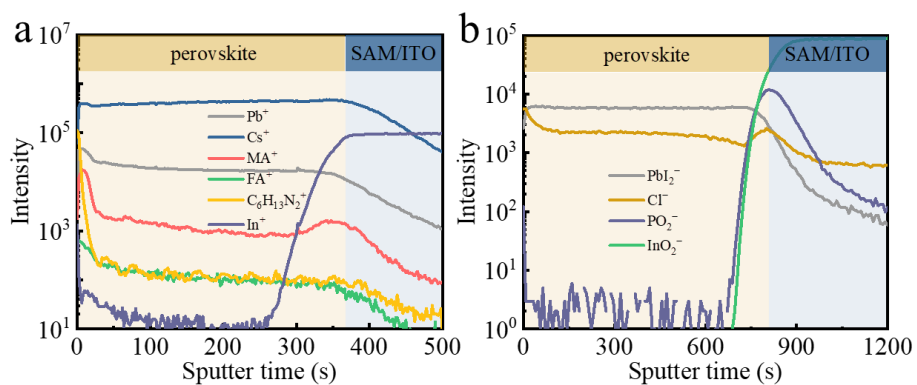
Supplementary Fig. 9 SEM images of the control, PCl-treated and 2FPDCl-treated perovskite films. To highlight the uniqueness of our designed DCI, we additionally selected other two similar molecules-4,4-difluoropiperidine hydrochloride (2FPDCI)^{1,2} and piperazinium hydrochloride (PCl)³ containing the same Cl anion as DCI for comparison. Similar to DCI, both 2FPDCI and PCl incorporate Lewis acid and Lewis base functional groups within their cationic moieties. For the control sample, the perovskite surface consists of clear steps (or terraced edges) and discontinuous PbI₂ filling the grain boundaries. In contrast, the PCl treatment drastically smooths out these steps and reacts with residual PbI₂, which is in line with our previous report³. For the 2FPDCl-treated perovskite film, platelet-like substances with significantly different sizes are observed on the surface compared to the control and PCl-treated samples, which is due to the formation of (2FPD)₂PbI₄ 2D phase on the surface of the 2FPDCl-treated sample^{1,2}. Obviously, both PCl-treated and 2FPDCl-treated samples show different surface morphologies in comparison with the DCI-treated sample (Fig. 1a).



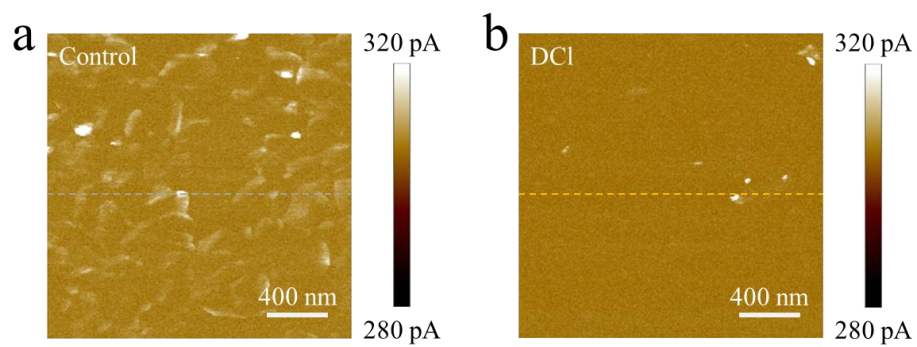
Supplementary Fig. 10 2D GIWAXS patterns of the control, PCl-treated, 2FPDCI-treated and DCI-treated perovskite films with a grazing incidence angle of 0.2° . Careful observation, reveals a new signal at $q_z = 0.49 \text{ \AA}^{-1}$ in only the 2FPDCI-treated sample and a new signal at $q_{xy} = 0.72 \text{ \AA}^{-1}$ in only the DCI-treated sample. Evidently, the crystallographic orientation of the 2D phase at the DCI-treated perovskite surface is entirely different from that at the 2FPDCI-treated perovskite surface, indicating that the orientation of 2D perovskite is also determined by the spatial dimensionality of the molecules, which has rarely been highlighted before.



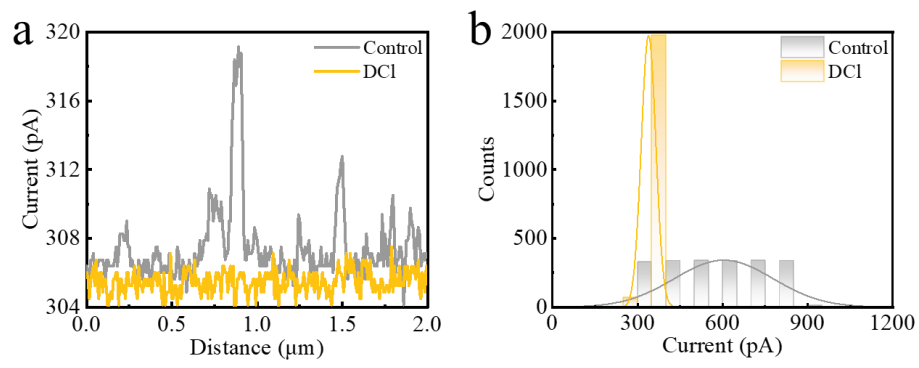
Supplementary Fig. 11 FTIR spectra of the pristine perovskite film and DCI powders. Fourier transform infrared (FTIR) spectroscopy reveals a distinct peak at 1457.9 cm^{-1} for DCI molecules, which serves as a characteristic optical response for detecting their nanoscale distribution in IR mapping profiles.



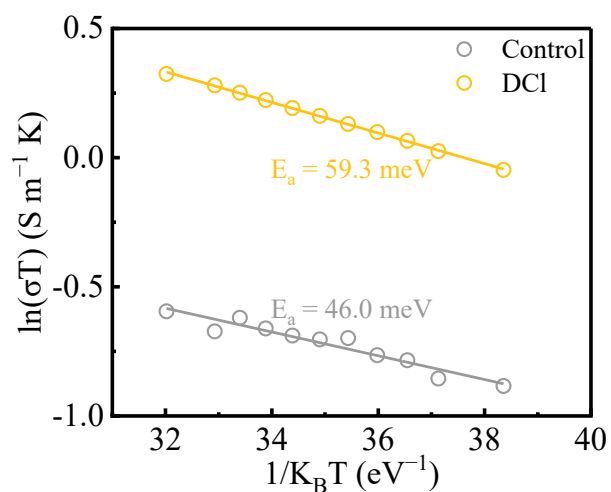
Supplementary Fig. 12 ToF-SIMS 2D depth profiles of DCI-treated perovskite film. (a) Cationic mode. (b) Anionic mode. Pb^+ and PbI_2^- represent the perovskite layer, Cs^+ represents Cesium cations, MA^+ represents Methylammonium cations, FA^+ represents Formamidinium cations, $\text{C}_6\text{H}_{13}\text{N}_2^+$ represents cage-like diammonium cations, Cl^- represents chloride anions, PO_2^- represents the MeO-2PACz self-assembled monolayer, In^+ and InO_2^- represent the indium tin oxide (ITO) substrate.



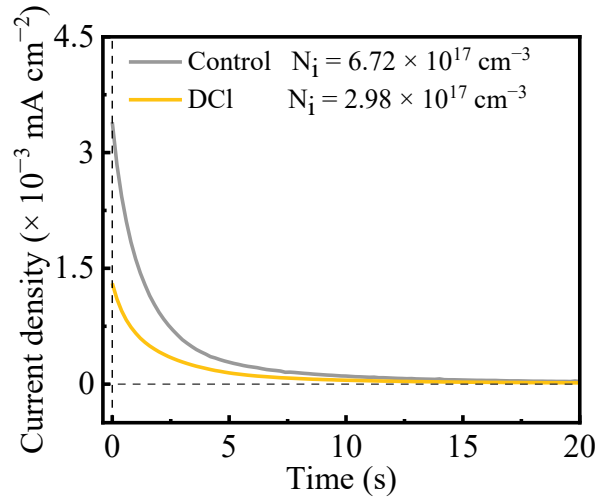
Supplementary Fig. 13 C-AFM images of (a) control and (b) DCl-treated perovskite films.



Supplementary Fig. 14 (a) Line profiles of local current flow for the gray and yellow dotted lines in the c-AFM images (Supplementary Fig. 13). (b) Statistical plots of the current extracted from the C-AFM images for the control and DCI-treated perovskite films.



Supplementary Fig. 15 Temperature-dependent conductivity of control and DCl-treated perovskite films. The activation energy value of the control perovskite is determined to be 46 meV, whereas for the DCl-treated film, it increases to 59.3 meV. This result indicates that DCl have a substantially boosted energy barrier for ion migration.

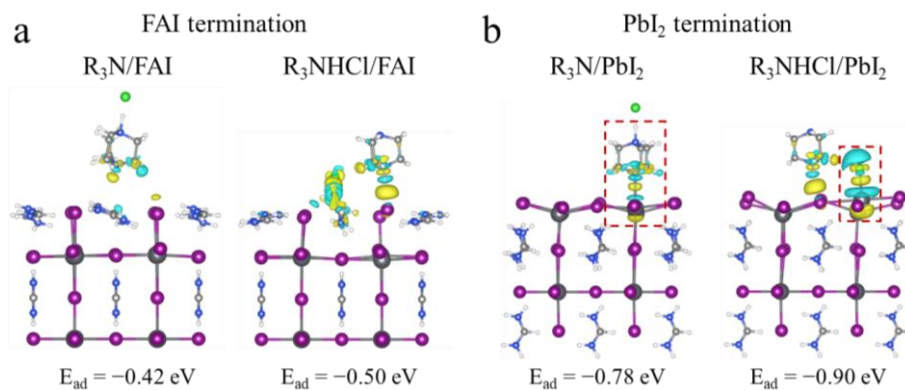


Supplementary Fig. 16 Transient ionic currents (TIC) for extracting mobile ion concentration within the control and DCI-treated perovskite films. We estimated the concentration of mobile ions using a mobile ion charging–discharging method, revealing a decrease from $6.72 \times 10^{17} \text{ cm}^{-3}$ in the control device to $2.98 \times 10^{17} \text{ cm}^{-3}$ in DCI-treated device. The mobile ion concentration in the DCI-treated device is approximately two times lower than that in the control device as the result of enhanced defect passivation after DCI incorporation. The decreased mobile ion concentration, combined with the increased ion migration activation energy (Supplementary Fig. 15) are conducive to suppressing phase segregation and enhancing device stability.

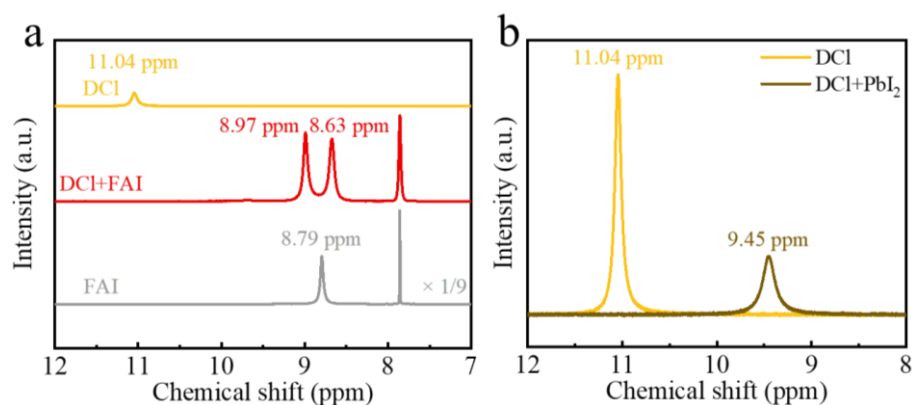
The concentration of mobile ions in the perovskite film was estimated from the TIC measurement based on the following formula⁴:

$$n = \frac{\int_{t_1}^{t_2} J dt}{eL}$$

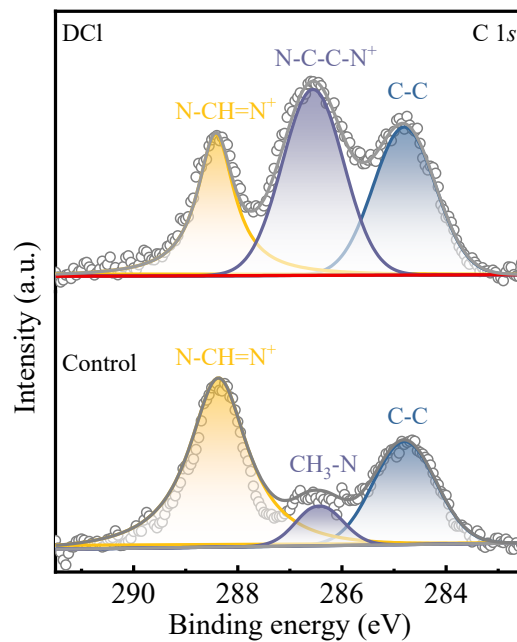
where t is the time, J is the current density, e is the elementary charge and L is the thickness of the perovskite film. The transient ionic relaxation current was measured in the dark under an external forward bias at V_{OC} for 60 seconds. The observed current primarily originates from the redistribution of mobile ions within the perovskite layer after the removal of the applied voltage, as dictated by the dark test conditions.



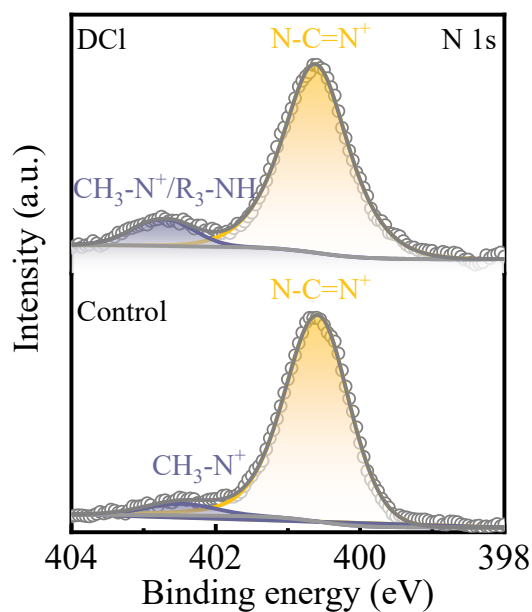
Supplementary Fig. 17 DFT modeling of the interaction of the DCI with (a) FAI-terminated and (b) PbI_2 terminated (100) perovskite surfaces. White, light gray, blue, violet, green and gray balls represent the H, C, N, I, Cl and Pb atoms, respectively. Charge density difference: Yellow iso-surface signifies electron accumulation, while blue represents electron depletion.



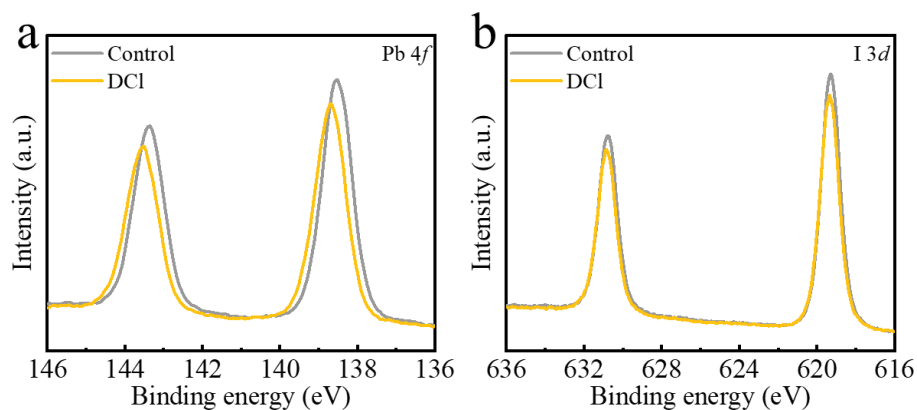
Supplementary Fig. 18 (a) ^1H nuclear magnetic resonance (NMR) spectra of DCl, FAI and DCl+FAI. (b) ^1H nuclear magnetic resonance (NMR) spectra of DCl and DCl+PbI₂. Liquid-state ^1H nuclear magnetic resonance (^1H NMR) was carried out to further explore the interaction mechanism of DCl with perovskite. The chemical shift at 11.04 ppm corresponds to the R_3NH^+ in DCl. Upfield shift trends of ^1H signal for the R_3NH^+ group in DCl was found when mixing DCl with FAI, as depicted in Supplementary Fig. 18a. ^1H signal of the R_3NH^+ group in DCl moves from 11.04 ppm to 9.68 ppm. In addition, the resonance signal (8.79 ppm) of protonated ammonium in FAI splits into two peaks at 8.97 ppm and 8.63 ppm, respectively. These chemical shifts indicate the formation of hydrogen bonding between DCl and FAI⁵. Upon addition of PbI₂ to DCl, ^1H signal of the R_3NH^+ group shows an obviously upfield shift from 11.04 ppm to 9.45 ppm (Supplementary Fig. 18b), which is attributed to the shielding effect induced by anion exchange from Pb-I bond to Pb-Cl bond and the direct ionic binding of Cl^- with the undercoordinated Pb^{2+6} .



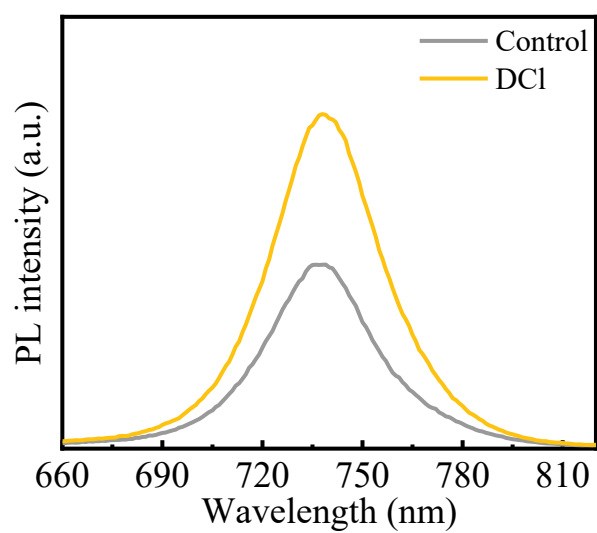
Supplementary Fig. 19 C 1s spectra of the control and DCI-treated perovskite films. In the C 1s XPS core (Supplementary Fig. 19), the binding energies centered at ~284.8, 286.6, and 288.4 eV are assigned to C-C, N-C-C-N, and N-C=N, respectively.



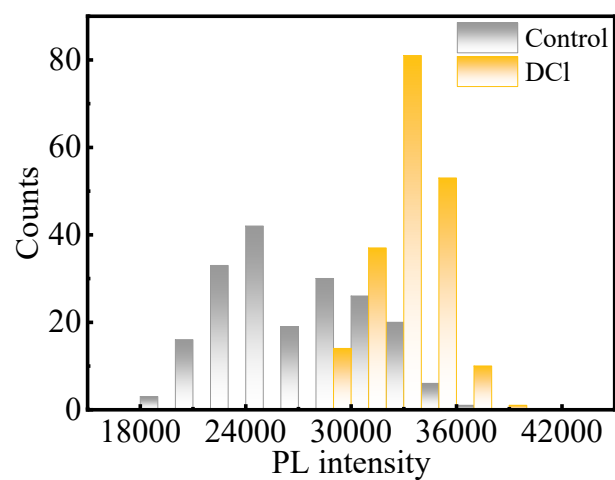
Supplementary Fig. 20 N 1s spectra of the control and DCl-treated perovskite films. An obvious increase in the higher binding energy peak at 402.7 eV is detected, likely corresponding to $\text{R}_3\text{-N}$ bond. The obvious N-C-C-N (Supplementary Fig. 19) and $\text{R}_3\text{-N}$ (Supplementary Fig. 20) characteristic peaks for DCl-treated film collectively indicates the existence of cage-like diammonium cation on the perovskite surface and its dominant interaction with the perovskite.



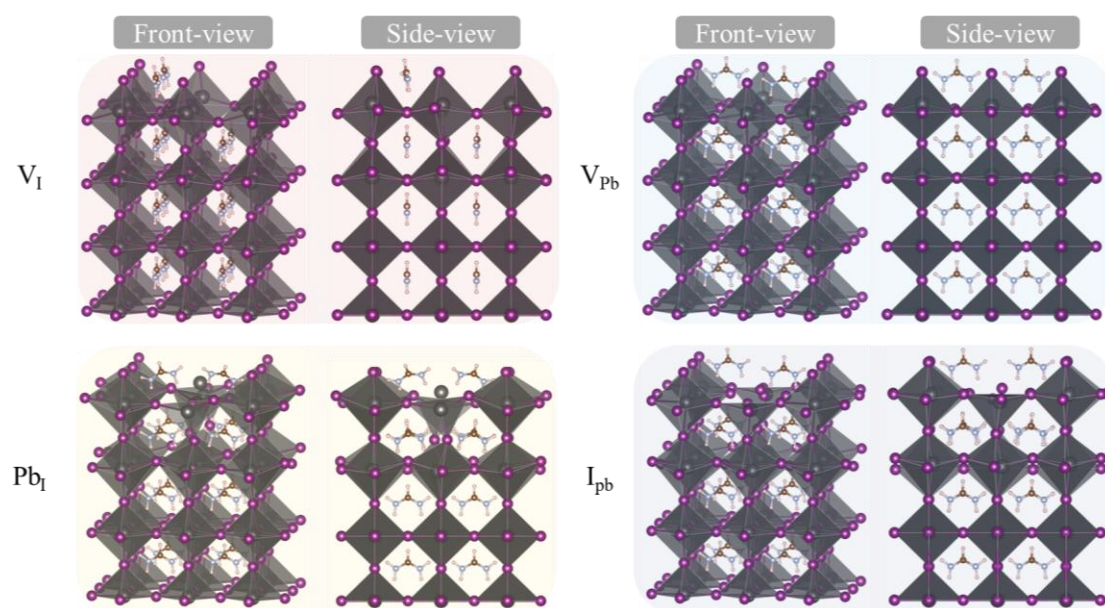
Supplementary Fig. 21 (a) Pb 4f XPS spectra of the control and DCl-treated perovskite films. (b) I 3d XPS spectra of the control and DCl-treated perovskite films. We notice that with the incorporation of DCl, the characteristic peaks in the Pb 4f XPS spectra shift toward the higher binding energy (from 143.37 and 138.51 eV for $4f_{5/2}$ and $4f_{7/2}$ to 143.55 and 138.66 eV, respectively), which may originate from the bond formation between the uncoordinated Pb^{2+} in perovskite or PbI_2 and DCl (Supplementary Fig. 21 and Supplementary Table. 1). Similar upward shift of the corresponding characteristic peaks in the I 4d XPS spectra is also observed (Supplementary Fig. 21b and Supplementary Table. 2), in which I $3d_{3/2}$ (630.71 eV) and I $3d_{5/2}$ (619.26 eV) for the control film slightly shift toward higher binding energy position of 630.77 and 619.31 eV, respectively, after DCl post-treatment, which could be ascribed to the incorporation of Cl^- .



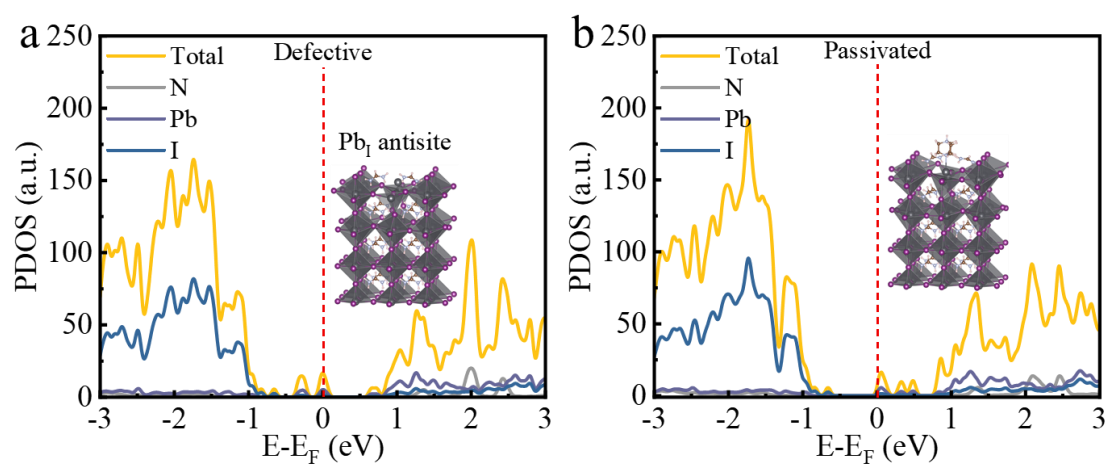
Supplementary Fig. 22 Steady-state PL spectra of the control and DCI-treated perovskite films.



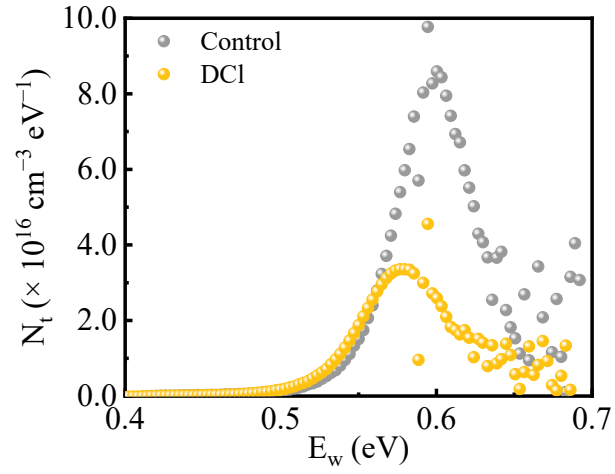
Supplementary Fig. 23 Histogram of PL intensity of the control and DCI-treated perovskite films.



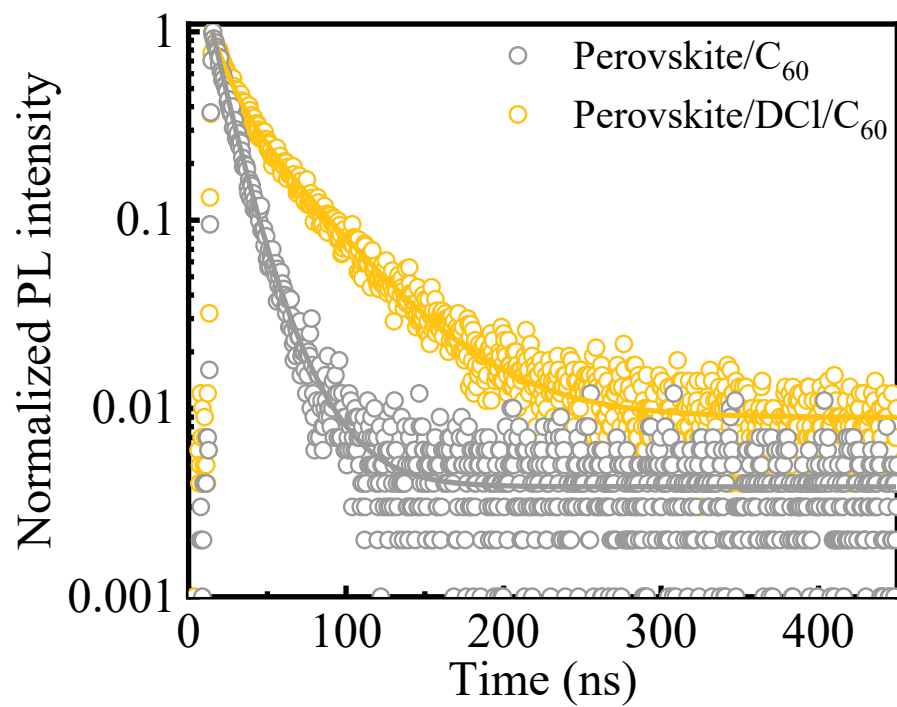
Supplementary Fig. 24 Front-view and side-view of four prevalent surface defects: V_I , Pb_I , V_{Pb} and I_{Pb} . Pink, light brown, light gray, violet and gray balls represent the H, C, N, I and Pb atoms, respectively.



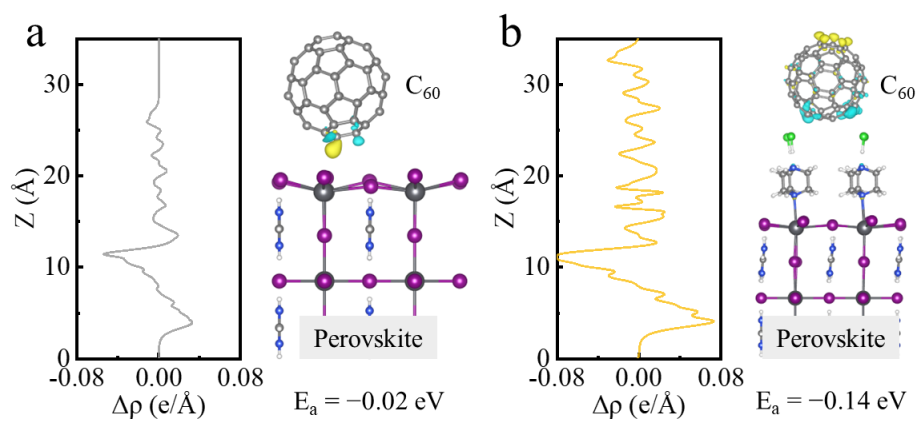
Supplementary Fig. 25 The partial density of states (PDOSs) of (a) defective perovskite (PbI antisite) and (b) passivated perovskite by DCl.



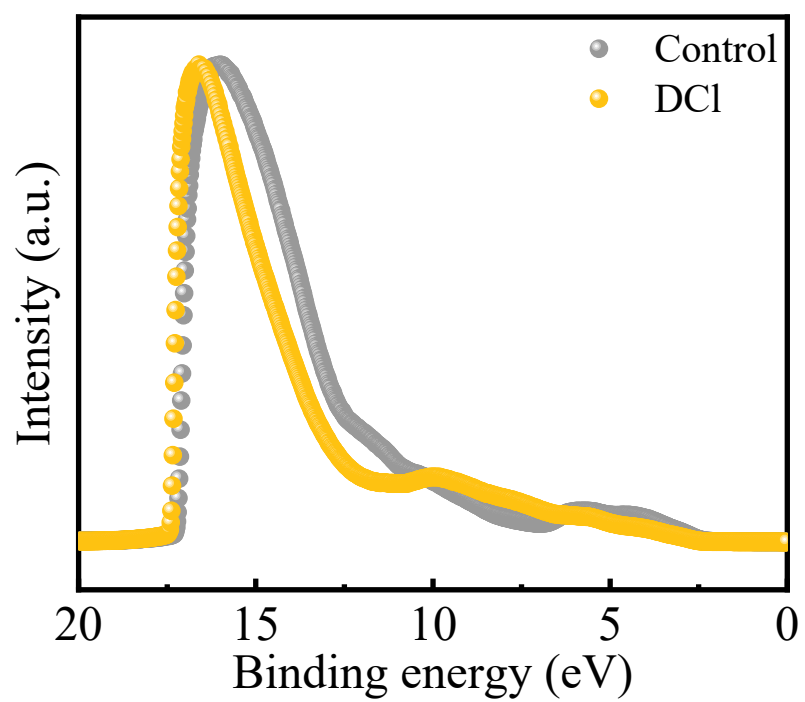
Supplementary Fig. 26 Trap density of states (tDOS) spectra of the control and DCl-treated devices.



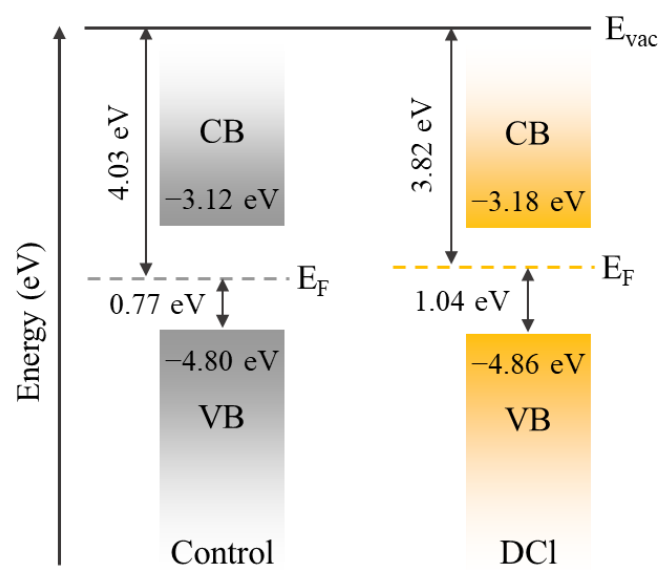
Supplementary Fig. 27 TRPL decay curves of the control and DCI-treated perovskite films capped with a C₆₀ layer.



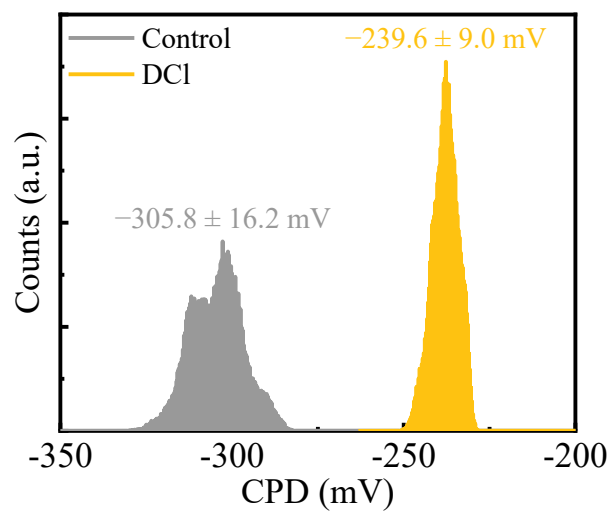
Supplementary Fig. 28 Charge density difference (yellow, accumulation; blue, depletion) along with the corresponding plane-averaged profiles for C_{60} ETL anchoring onto both the (a) control and (b) DCI passivated (100) perovskite surfaces. The calculated adsorption energy (E_a) is also shown. White, light gray, blue, violet, green and gray balls represent the H, C, N, I, Cl and Pb atoms, respectively.



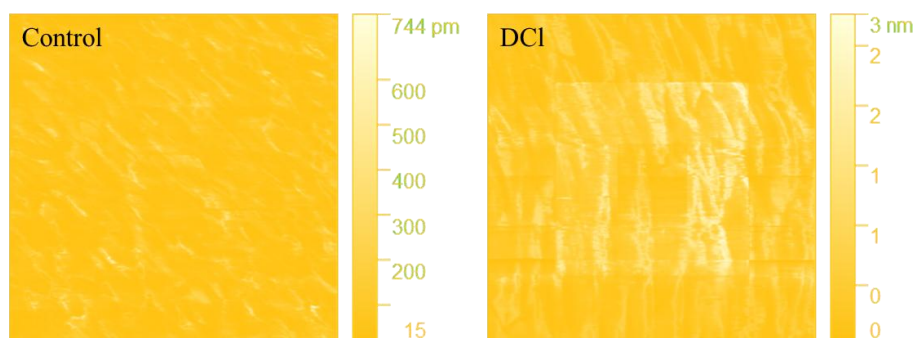
Supplementary Fig. 29 UPS spectra of the control and DCl-treated perovskite films.



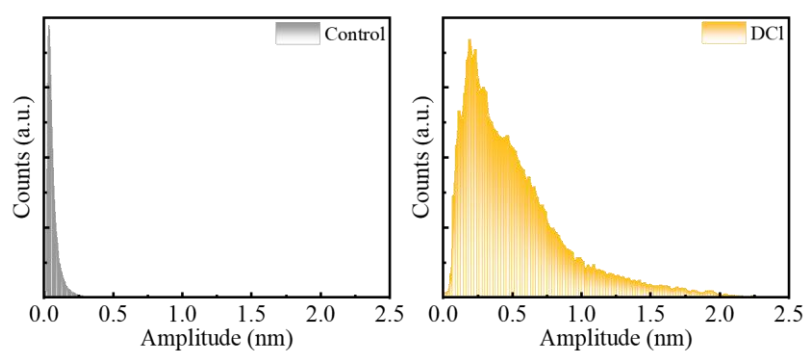
Supplementary Fig. 30 Energy-level scheme based on the UPS spectra.



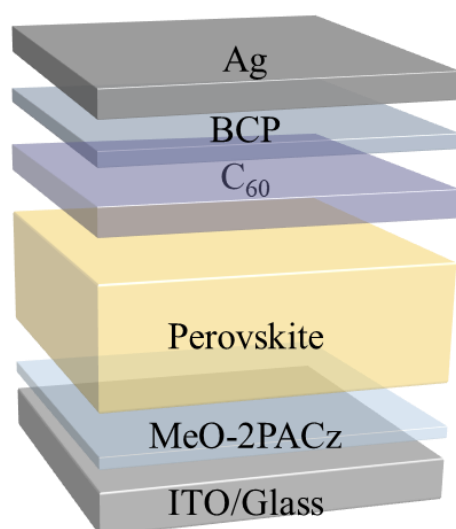
Supplementary Fig. 31 CPD statistical distributions derived from the KPFM images of the control and DCI-treated perovskite films.



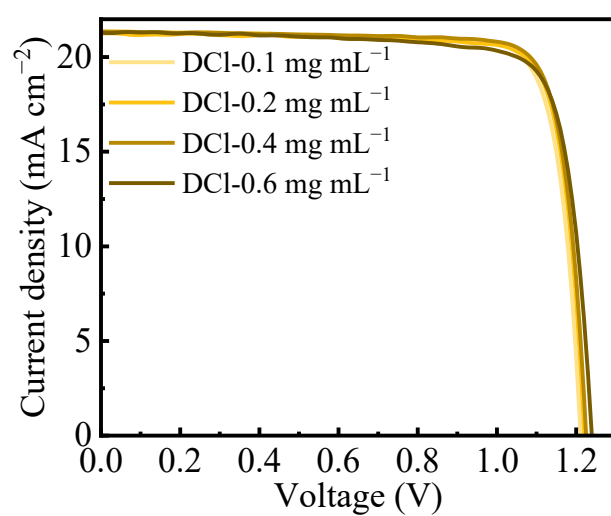
Supplementary Fig. 32 PFM amplitude images of the control and DCL-treated perovskite films.



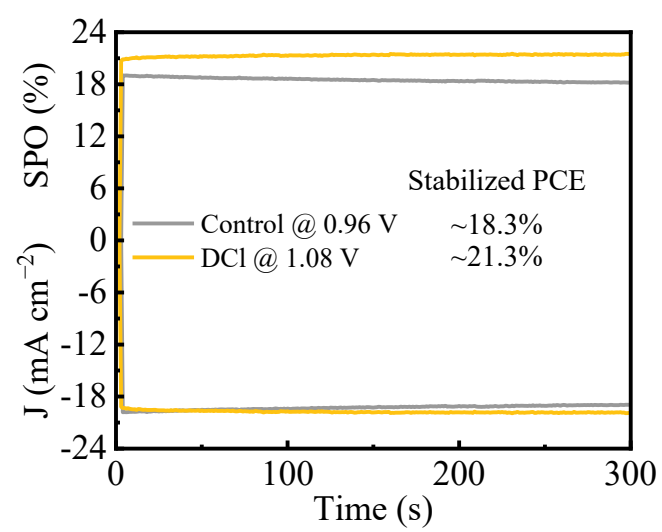
Supplementary Fig. 33 The corresponding amplitude distribution histogram of the control and DCI-treated perovskite films.



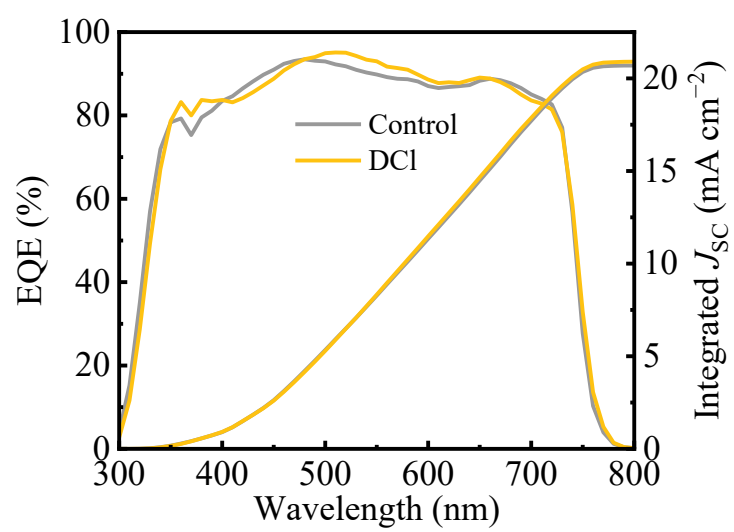
Supplementary Fig. 34 Device architecture of the single-junction perovskite solar cells.



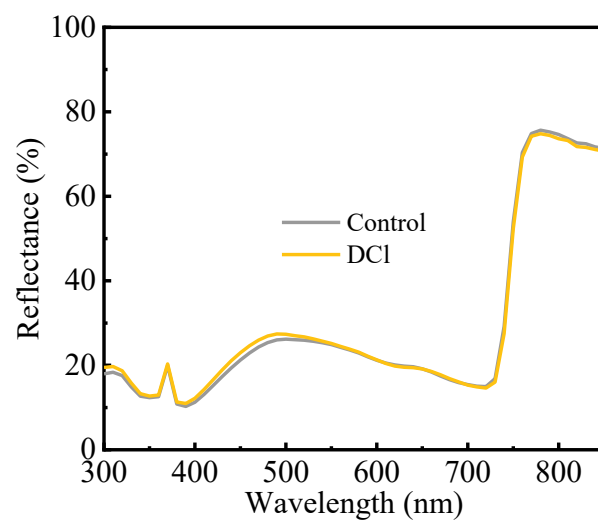
Supplementary Fig. 35 J - V curves of champion devices treated with 0.1, 0.2, 0.4 and 0.6 mg mL⁻¹ of DCl.



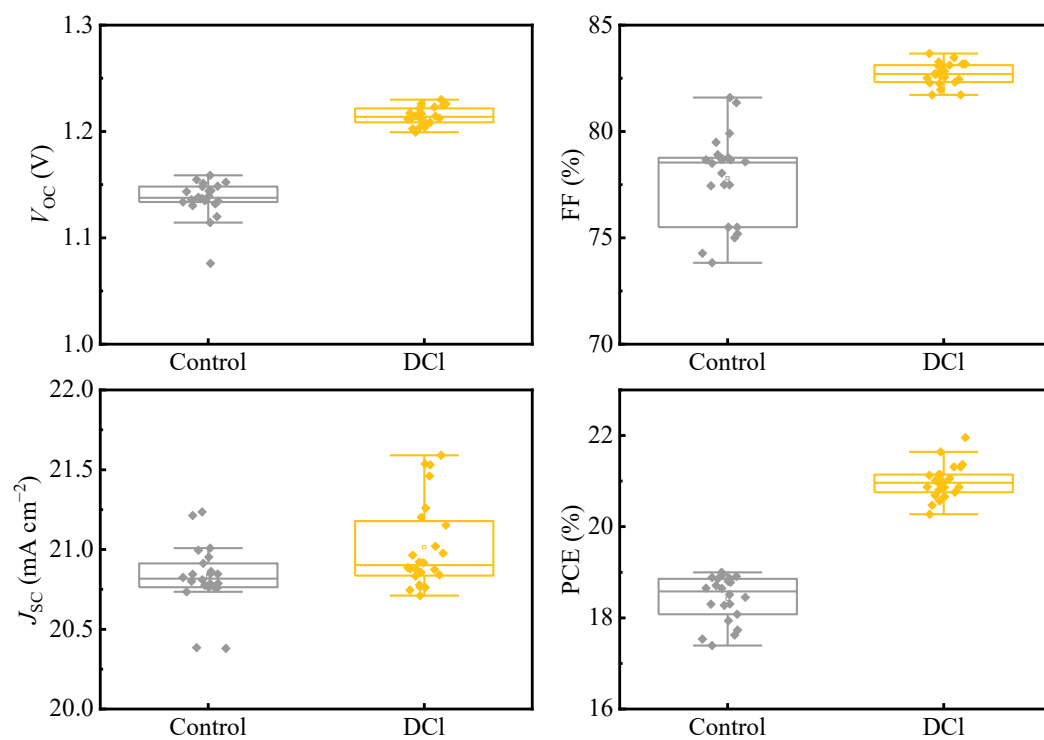
Supplementary Fig. 36 Steady-state output of PCEs for the champion control and DCI-treated perovskite solar cells.



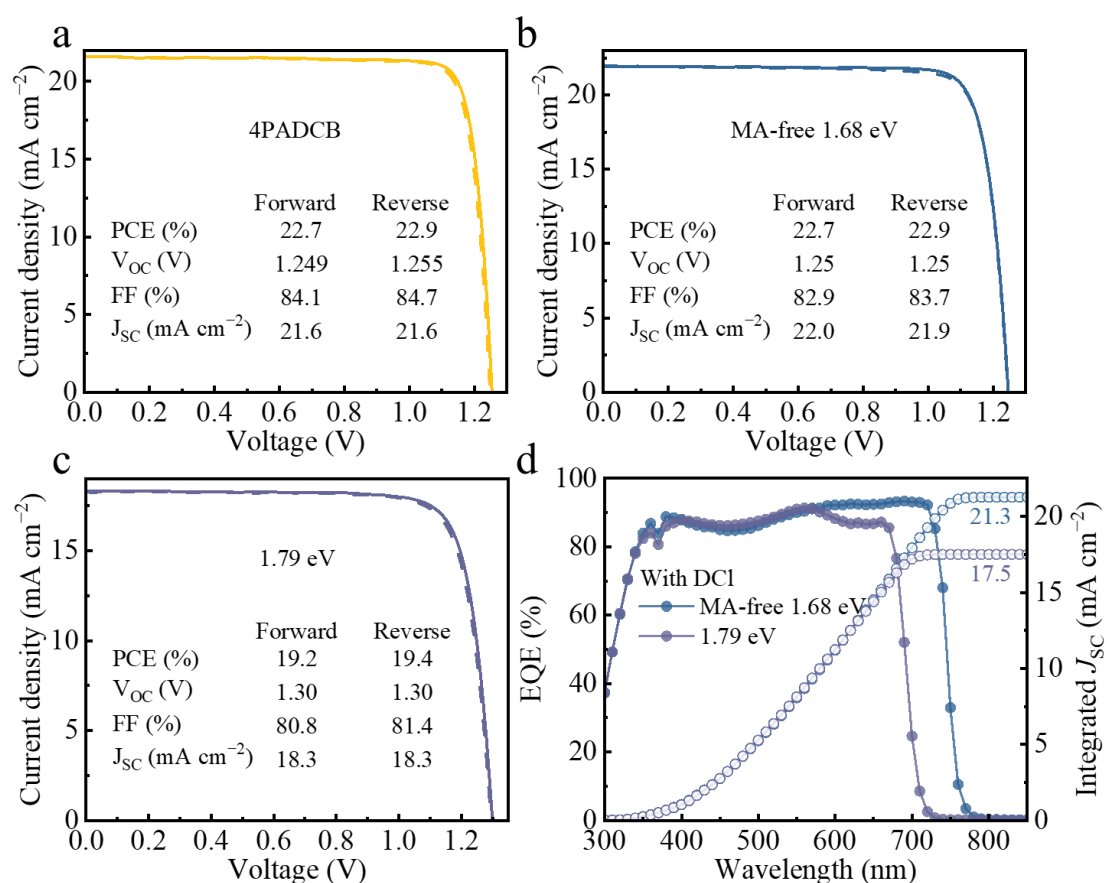
Supplementary Fig. 37 EQE spectra of the control and DCl-treated perovskite solar cells.



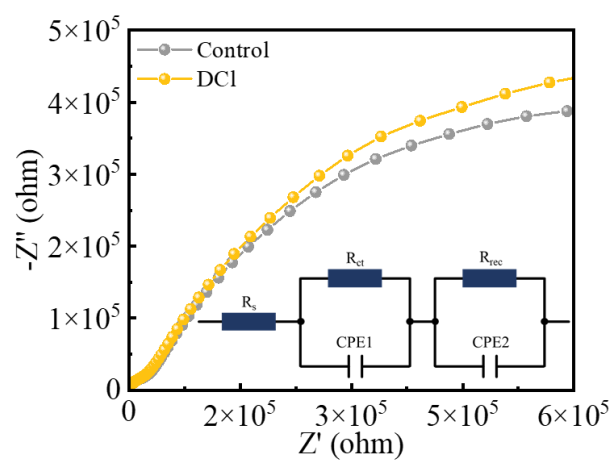
Supplementary Fig. 38 Reflectance spectra of the control and DCI-treated devices.



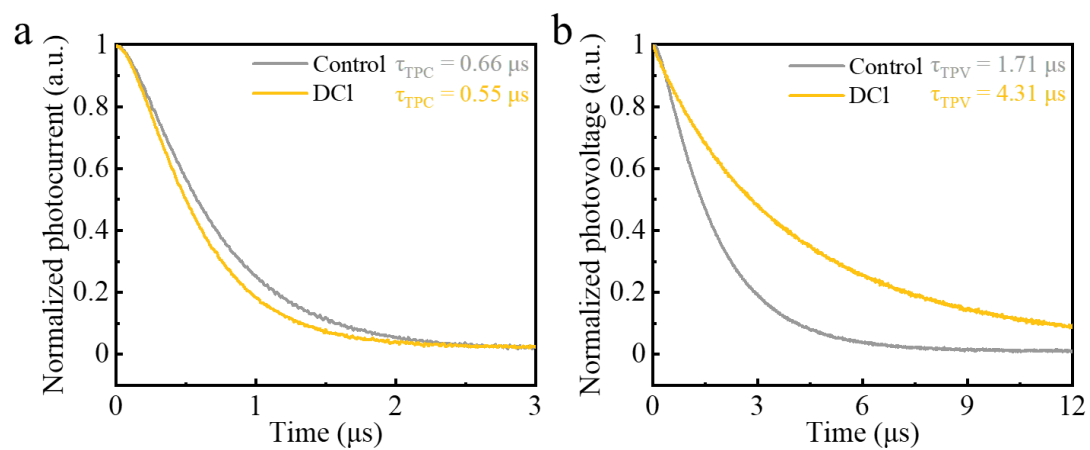
Supplementary Fig. 39 Statistical distributions of PV parameters for control and DCI-treated perovskite solar cells.



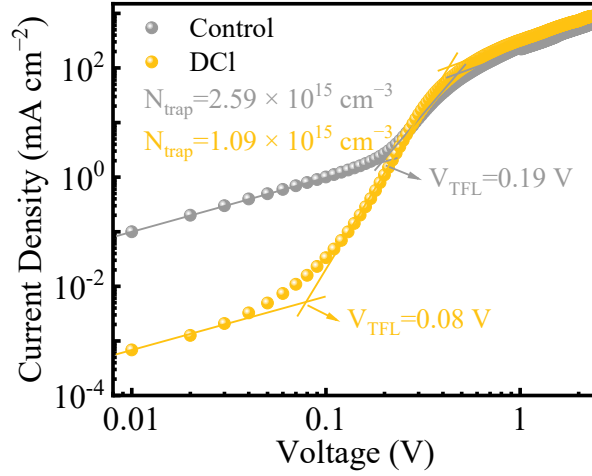
Supplementary Fig. 40 (a) J - V curves of the DCI-treated perovskite solar cells based on 4PADCB hole transport layer. (b, c) J - V curves and (d) EQE spectra and the corresponding integrated J_{SC} s for 1.68 eV MA-free and 1.79 eV wide-bandgap perovskite solar cells.



Supplementary Fig. 41 EIS spectra of the control and DCl-treated devices (inset: equivalent circuit).



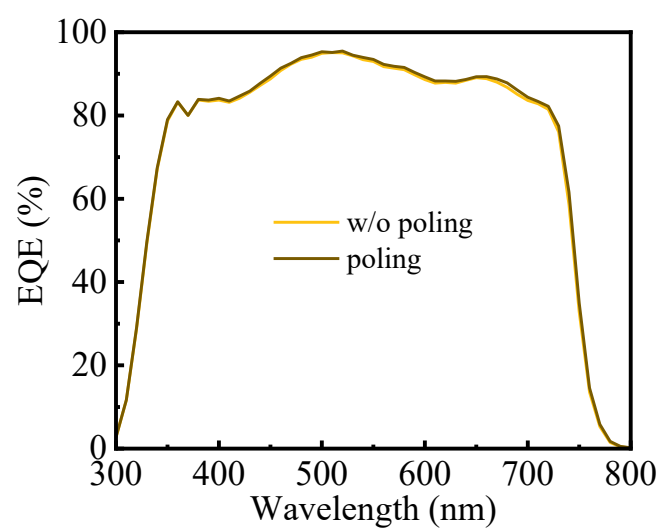
Supplementary Fig. 42 (a) TPC and (b) TPV decay curves of the control and DCI-treated devices.



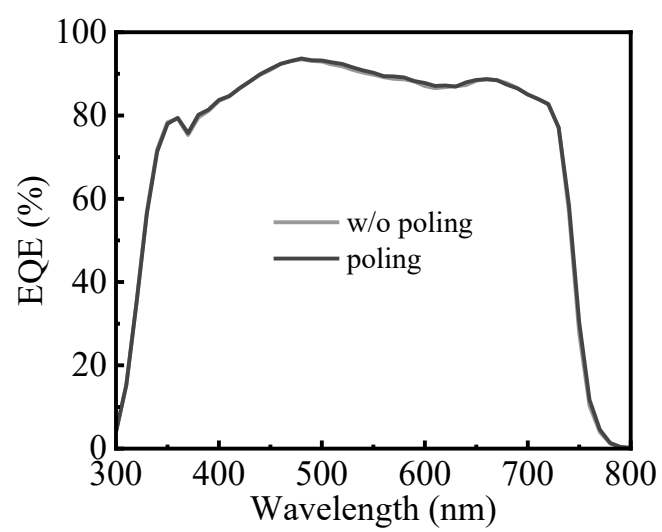
Supplementary Fig. 43 Dark J–V curves of electron-only devices (Glass/ITO/ALD-SnO_x/perovskite/C₆₀/BCP/Ag) with and without DCl treatment. The defect state density (N_{trap}) can be estimated using the following equation:

$$N_{\text{trap}} = \frac{2\epsilon_r\epsilon_0 V_{\text{TFL}}}{2qL^2}$$

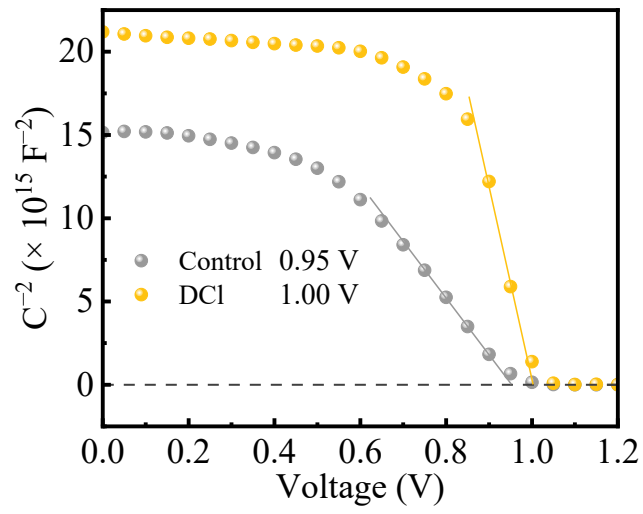
Where q is the elementary charge, V_{TFL} is the trap filling limit voltage, ϵ_r signifies the relative dielectric constant of the perovskite, and ϵ_0 is the vacuum dielectric constant. L represents the perovskite film thickness.



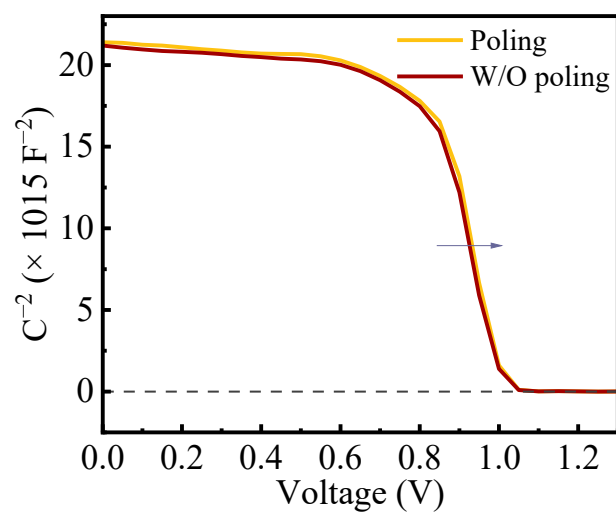
Supplementary Fig. 44 EQE spectra of the DCI-treated perovskite solar cells with and without poling (20.9 mA cm^{-2} without poling and 21.1 mA cm^{-2} with poling).



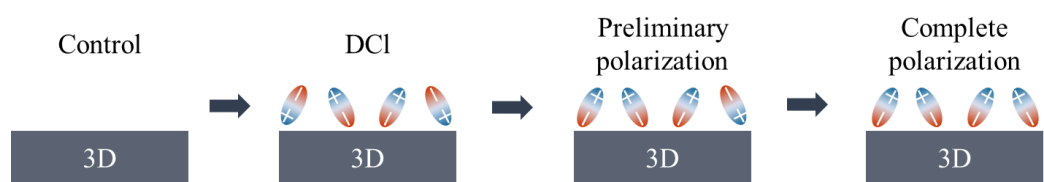
Supplementary Fig. 45 EQE spectra of the control perovskite solar cells with and without poling (20.7 mA cm^{-2} without poling and 20.8 mA cm^{-2} with poling).



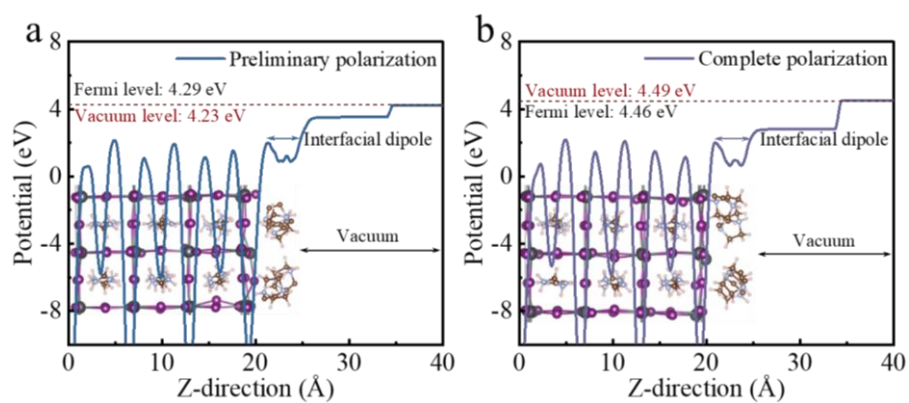
Supplementary Fig. 46 Mott–Schottky plots of the control and DCl-treated perovskite solar cells. The curves on top of the data were obtained by linear fitting the drop region of Mott–Schottky plots, and the built-in electric field was extracted via the intercept of the straight line with the x axis.



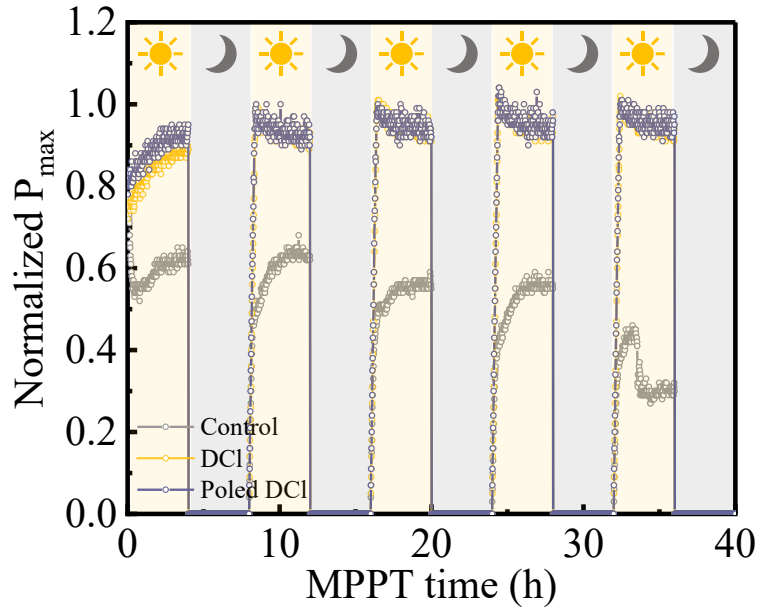
Supplementary Fig. 47 Mott-Schottky plots of the DCI-treated perovskite solar cell before and after poling.



Supplementary Fig. 48 Schematic illustration of electric dipole switching under external polarization in the DCI-treated perovskite surface.

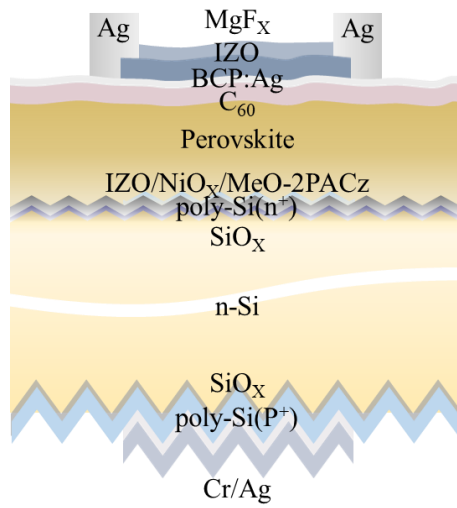


Supplementary Fig. 49 DFT-calculated electrostatic potential and surface work functions of perovskites the adsorption of cage-like cations. (a) Preliminary polarization, (b) Complete polarization.

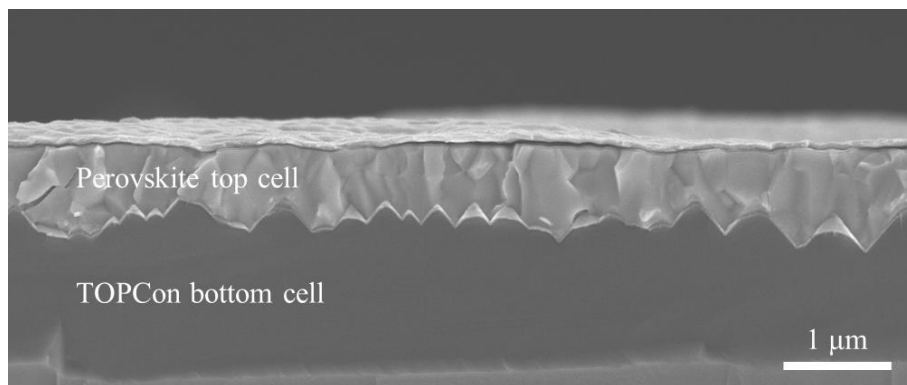


Supplementary Fig. 50 Operational stability of control, DCI-treated and poled DCI-treated devices by MPP tracking with an artificially induced day-night cycle. As presented in Supplementary Fig. 50, the control device shows a continuous drop in maximum power point ($P_{\max}$) without any recovery after storage in the dark, indicating poor stability. In addition, the control device consistently exhibits a gradual increase in $P_{\max}$ during each light cycle, which is detrimental to achieving optimal energy output under realistic light-dark cycling conditions. In contrast, the DCI-treated and poled DCI-treated devices maintain stable performance without noticeable degradation during 40 hours of day-night cycle testing. In case of DCI-treated and poled DCI-treated devices, although the $P_{\max}$ drops to 90-95% of its initial value for each light cycle of 4 hours, it fully recovers to its original level after a subsequent 4-hour dark period. This reversible behavior demonstrates a self-healing effect, which could be practically beneficial for commercial perovskite solar cell operation, as it enables nighttime self-recovery from the ~5-10% photodegradation accumulated during daytime use⁸. Moreover, in each light cycle, the $P_{\max}$ values of the DCI-treated and poled DCI-treated devices immediately reach their optimal levels at the onset of illumination, which is favorable for maximizing energy output under realistic light-dark cycling conditions. Notably, the poled DCI-treated device delivers a higher output power in the initial light cycle but converges to the same performance level as the DCI-treated device in the following cycles, indicating that the ferroelectric effect contributes to a slight, yet short-lived, enhancement in energy yield. This behavior may be ascribed to the transient ferroelectric characteristic of the DCI-mediated quasi-2D perovskite. In fact, a permanent ferroelectric state would be energetically unfavorable for 2D perovskites, as it would inevitably compromise the structural stability of the 2D perovskite. Before poling, the ferroelectric organic cations predominantly occupy the A-sites to maintain the structural integrity of the quasi-2D lattice. Upon applying an external electric field, these cations undergo ferroelectric switching and deviate from their equilibrium positions, generating an internal depolarization field within the ferroelectric spacer layers that facilitates charge transport. However, during the dark period, the organic

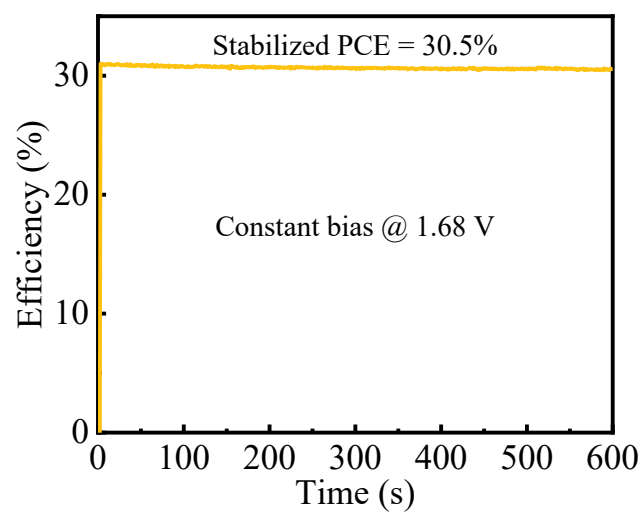
cations gradually return to their original A-site locations to restore the structural stability of the 2D perovskite. This dynamic process may explain why the poled DCI-treated device delivers enhanced output power during the initial light cycle but converges to the same performance level as the unpoled DCI-treated device in subsequent cycles.



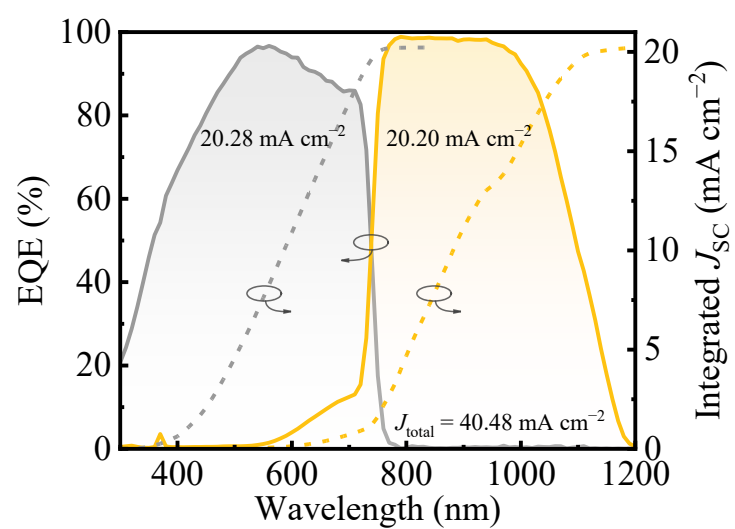
Supplementary Fig. 51 Schematic illustration of the perovskite/silicon tandem solar cell architecture.



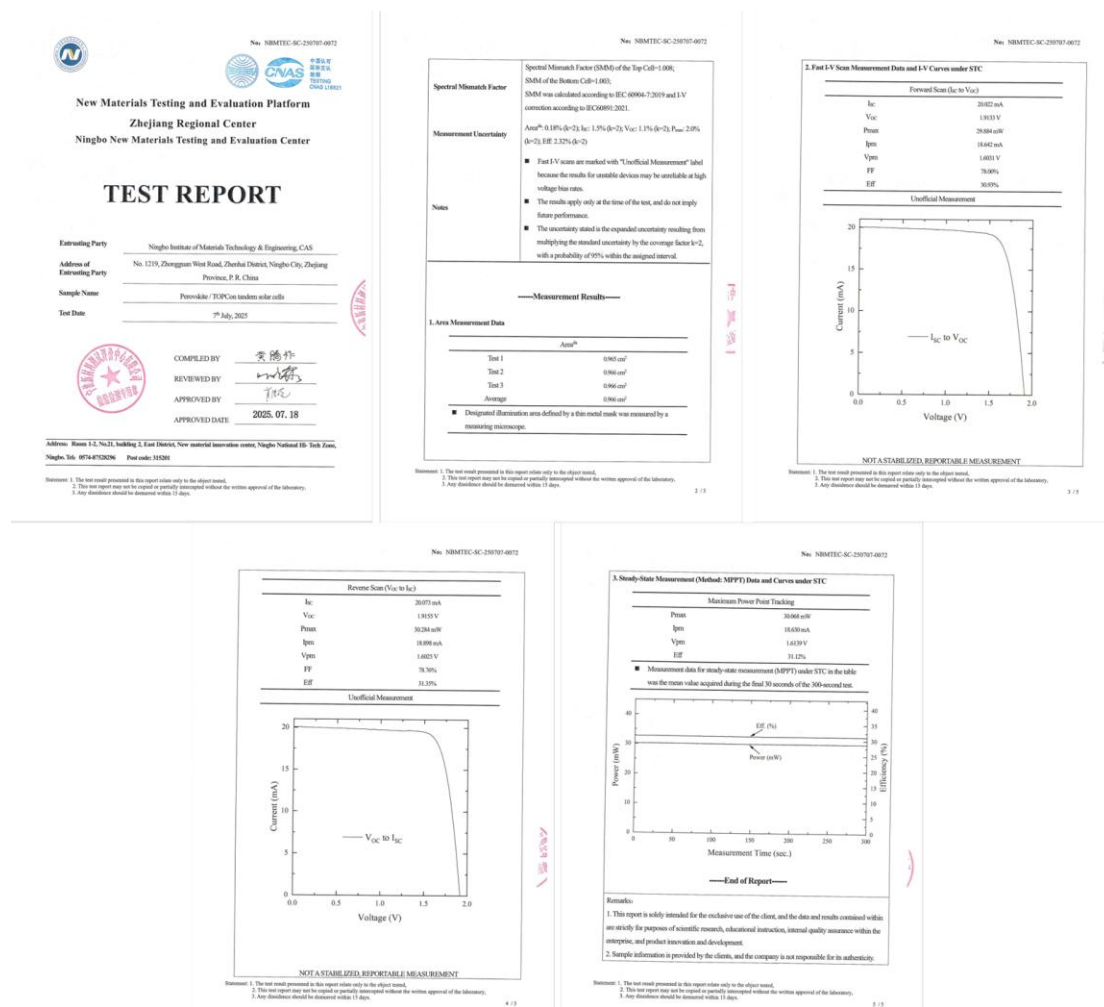
Supplementary Fig. 52 Cross-sectional SEM image of the DCl-treated perovskite/silicon tandem solar cell.



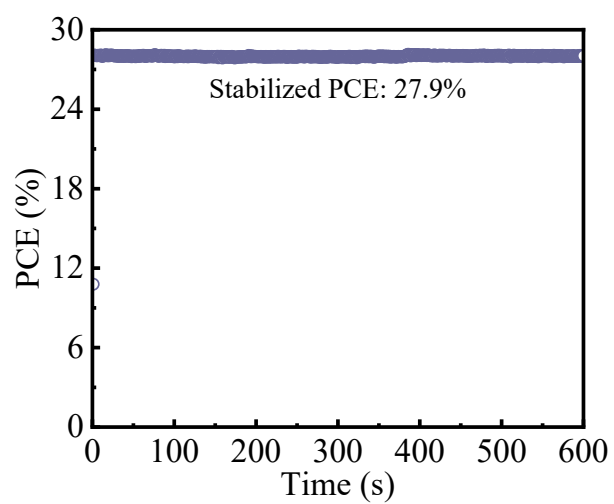
Supplementary Fig. 53 Steady state PCE of the champion DCl-treated perovskite/silicon TSC under 100 mW cm^{-2} AM1.5G illumination.



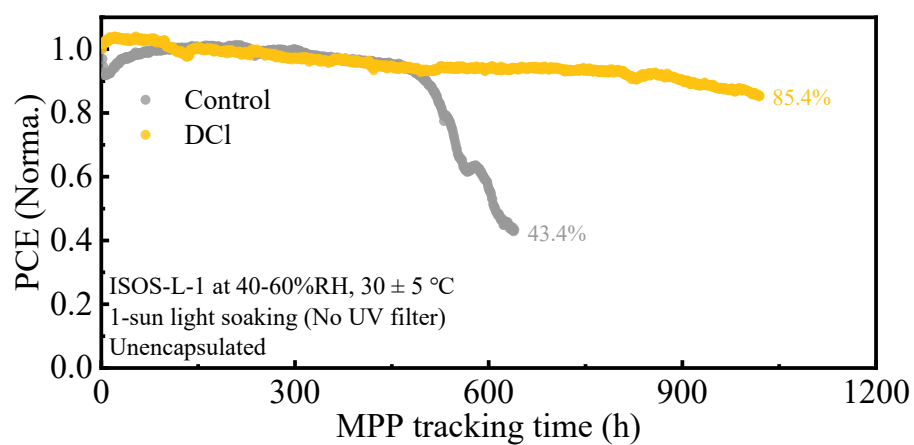
Supplementary Fig. 54 EQE spectrum of the DCI-treated perovskite/silicon tandem solar cell.



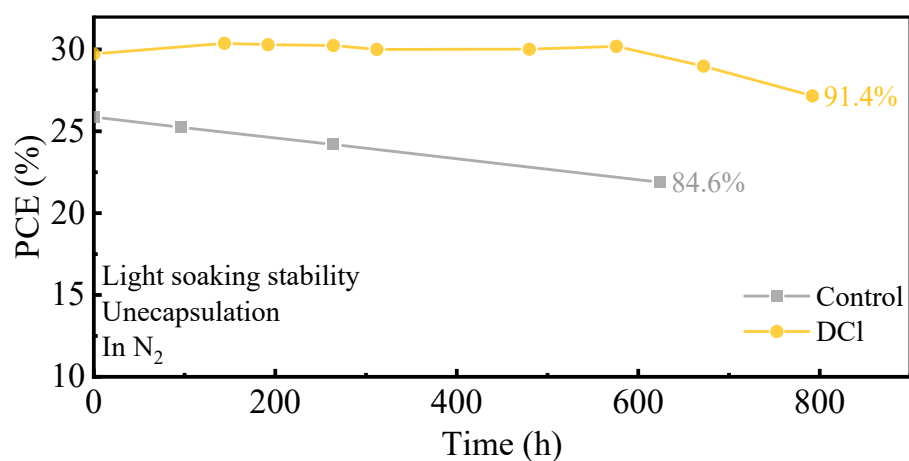
Supplementary Fig. 55 Certified results of DCI-treated perovskite/silicon tandem from an accredited photovoltaic certification laboratory.



Supplementary Fig. 56 Steady-state PCE with a constant bias of 1.58 V for the large-area monolithic perovskite/silicon TSCs with an active area of 4 cm².



Supplementary Fig. 57 Continuous MPP tracking of the unencapsulated control and DCI-treated perovskite/silicon TSCs under 1-sun illumination (100 mW cm^{-2}) with a xenon lamp without UV filter in ambient air ($30 \pm 5 \text{ }^{\circ}\text{C}$, 40–60% RH).



Supplementary Fig. 58 Light soaking stability of unencapsulated control and DCI-treated perovskite/silicon TSCs under 1-sun illumination (LED, 100 mW cm⁻²) in N₂ atmosphere at open-circuit conditions.

Supplementary Table. 1 Binding energy of Pb 4f XPS spectra for the control and DCl-treated perovskite film.

Pb 4f	Control	DCl
4f 5/2 (eV)	143.37	143.55
4f 7/2 (eV)	138.51	138.66

Supplementary Table. 2 Binding energy of I 3d XPS spectra of the control and DCI-treated perovskite film.

I 3d	Control	DCI
3d 3/2 (eV)	630.71	630.77
3d 5/2 (eV)	619.26	619.31

Supplementary Table. 3 TRPL parameters of the control and DCl-treated perovskite films with a C₆₀ layer by the bi-exponential fitting.

Devices	A ₁	τ_1 [ns]	A ₂	τ_2 [ns]	τ_{avg} [ns]
Perovskite/C ₆₀	1.02	10.77	0.17	22.91	12.50
Perovskite/DCl/C ₆₀	0.49	10.48	0.52	43.01	27.23

Supplementary Table. 4 Summary of reported perovskite solar cells with ferroelectric materials.

Ferroelectric materials	Structure	Strategy	Ref.
P(VDF-TrFE)	Chain-like	Additive	9
P(VDF-TrFE)	Chain-like	Additive	10
P(VDF-TrFE):DH	Chain-like	Additive	11
R-1-(4-chlorophenyl)ethylamine	Plane-type	Additive	12
Trimethyliodomethylammonium tris(μ 2-iodo)-lead	1D	Additive	13
R-1-(4-chlorophenyl)ethylamine	Plane-type	Post-treatment	14
3-pyridinylmethylammonium	Ring-like	Additive	15
4,4-difluoropiperidine hydrochloride	Ring-like	Post-treatment	16
4,4-difluoropiperidine hydrochloride	Ring-like	Post-treatment	17
1,4-diazabicyclo[2.2.2]octane chloride	Cage-like	Post-treatment	This work

Supplementary Table. 5 J - V parameters of perovskite single-junction solar cells treated with 0 (control), 0.1, 0.2, 0.4 and 0.6 mg mL⁻¹ of DCl.

Concentration [mg mL ⁻¹]	V_{oc} [V]	FF [%]	J_{sc} [mA cm ⁻²]	PCE [%]
Control	1.137	78.7	21.2	19.0
0.1	1.211	82.3	21.4	21.3
0.2	1.221	82.7	21.3	21.5
0.4	1.226	83.2	21.5	21.9
0.6	1.240	80.5	21.3	21.3

Supplementary Table. 6 J - V parameters of the best control and DCl-treated perovskite solar cells before poling.

W/O poling	V_{oc} [V]	FF [%]	J_{sc} [mA cm ⁻²]	PCE [%]
Control	1.134	78.5	21.2	18.8
	1.137	78.7	21.2	19.0
DCl	1.225	82.9	21.5	21.8
	1.226	83.2	21.5	21.9

Supplementary Table. 7 The average and standard deviation of device parameters for single-junction perovskite solar cells without poling.

Sample	V_{oc} [V]	J_{sc} [mA cm ⁻²]	FF [%]	PCE [%]
Control	1.137±0.017	20.83±0.20	77.80±2.12	18.42±0.50
DCI	1.214±0.004	21.01±0.27	82.70±0.53	20.99±0.38

Supplementary Table. 8 EIS fitting parameters of the control and DCI-treated devices.

Sample	R_s (Ω)	R_{ct} ($\times 10^4 \Omega$)	R_{rec} ($\times 10^4 \Omega$)
Control	8.2	97	3.4
DCI	8.1	3.5	86

Supplementary Table. 9 The average and standard deviation of device parameters for single-junction perovskite solar cells after poling.

Sample	V_{oc} [V]	J_{sc} [mA cm ⁻²]	FF [%]	PCE [%]
Control	1.141±0.003	21.04±0.37	78.26±0.57	18.76±0.35
DCI	1.224±0.009	21.53±0.35	83.32±0.63	21.94±0.44

Supplementary Table. 10 J - V parameters of the best control and DCl-treated perovskite solar cells after poling.

Poling	V_{oc} [V]	FF [%]	J_{sc} [mA cm ⁻²]	PCE [%]
Control	1.138	78.9	21.3	19.1
	1.139	78.8	21.4	19.2
DCl	1.235	83.1	21.9	22.5
	1.238	83.6	21.8	22.6

Supplementary Table. 11 Summary of large-area monolithic perovskite/silicon tandems based on the spin-coating method reported in literatures. HJ: homojunction; SHJ: silicon heterojunction; TOPCon: tunnel oxide passivated contact.

PCE [%]	J_{sc} [mA cm ⁻²]	V_{oc} [V]	FF [%]	silicon	type	Area [cm ²]	Ref.
20.5	16.1	1.68	78.0	HJ	n-i-p	4	18
21.8	16.2	1.74	78.0	HJ	n-i-p	16	19
22.6	17.5	1.72	75.0	SHJ	p-i-n	57.4	20
23.1	16.5	1.78	81.0	HJ	n-i-p	4	21
27.1	19.1	1.88	75.5	SHJ	p-i-n	3.8	22
17.3	14.1	1.78	67.0	TOPerc	n-i-p	25	23
28.3	19.1	1.84	80.2	TOPCon	p-i-n	4	24
27.1	/	/	/	SHJ	n-i-p	24	25
26.4	19.0	1.91	72.6	SHJ	p-i-n	8.9	26
28.4	19.4	1.91	76.8	TOPCon	p-i-n	4	This work

Supplementary Table. 12 Summary of MPP stability for the state-of-the-art monolithic perovskite/silicon TSCs reported in literatures.

Structure	Environment	Encapsulation	Light condition	PCE retention	Ref.
p-i-n	In ambient air; 25–35%RH; 30 °C	no	1-sun illumination	T ₉₂ =100 h	27
p-i-n	In ambient air; 30–40%RH; 25 °C	no	LED with wavelengths of 470 and 940 nm	T _{95.5} =300 h	28
p-i-n	In N ₂ ; 25±1 °C	no	1-sun illumination	T ₉₀ =100 h	29
p-i-n	In ambient air; 20%RH; 25 °C	no	1-sun illumination	T ₉₈ =300 h	30
p-i-n	In N ₂ ; 25±1 °C	no	1-sun illumination	T ₁₁₈ =100 h	31
p-i-n	In N ₂ ; 25 °C	no	White LED light source (940 nm LED light for extra compensation)	T ₉₈ =200 h	32
p-i-n	In ambient air; 40–70%RH; 20–35 °C	no	Xenon lamp (100 mW cm ⁻²)	T _{93.6} =450 h	33
p-i-n	In ambient air; 25–75%RH, most often ≈60%RH; 20–35 °C	no	Xenon lamp (100 mW cm ⁻²)	T _{86.6} =306 h	34
p-i-n	In N ₂ ; 25 °C	no	1-sun illuminations	T ₈₀ =1200 h	35
p-i-n	In ambient air; 30–40%RH; 25°C	no	1-sun illuminations	T ₁₀₀ =700 h	36
p-i-n	In ambient air; 25%RH; 25°C	no	1-sun illuminations	T ₉₆ =527 h	37
p-i-n	In N ₂ ; 25 °C	no	1-sun LED	T ₉₀ =1200 h	38

			illumination		
p-i-n	In ambient air; 40%RH; 25°C	no	1-sun illumination	T _{90.6} =100 h	39
p-i-n	In N ₂ ; 25 °C	no	1-sun illumination	T ₉₅ =280 h	40
n-i-p	In N ₂ ; 25 °C	no	1-sun LED illumination	T ₈₀ =700 h	41
p-i-n	In ambient air; 40–60%RH; 25–35 °C	no	1-sun xenon-lamp illumination (without a UV filter)	T ₈₀ =755 h	3
p-i-n	In ambient air; 25 °C	yes	AM 1.5G illumination	T _{96.2} =1068 h	42
p-i-n	In ambient air; 40%RH	yes	0.8-sun infrared-enhanced white LED	T ₉₅ =1000 h	26
p-i-n	In ambient air; 25%RH; 25 °C	no	1-sun illumination	T ₉₆ =527 h	43
p-i-n	In ambient air; 40–60%RH; 25–35 °C	no	1-sun xenon-lamp illumination (without a UV filter)	T _{85.4} =1020 h	This work

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