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for *Adv. Sci.*, DOI 10.1002/advs.202204486

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Simultaneous lattice engineering and defect control via cadmium incorporation for high-performance inorganic perovskite solar cells

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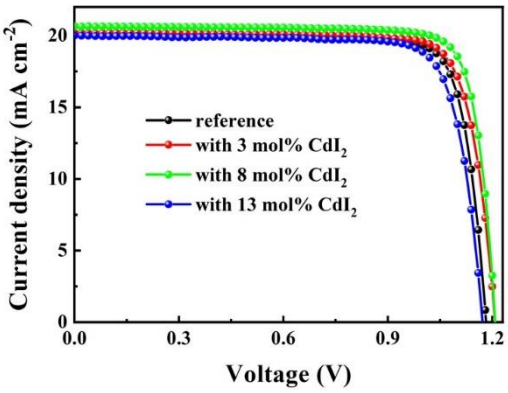

Figure S1. *J-V* curves of $CsPbI_{3-x}Br_x$ inorganic perovskite solar cells with different concentration of Cd-doping. The measurement was performed under one sun irradiation with an aperture area of 0.09 mm^2.

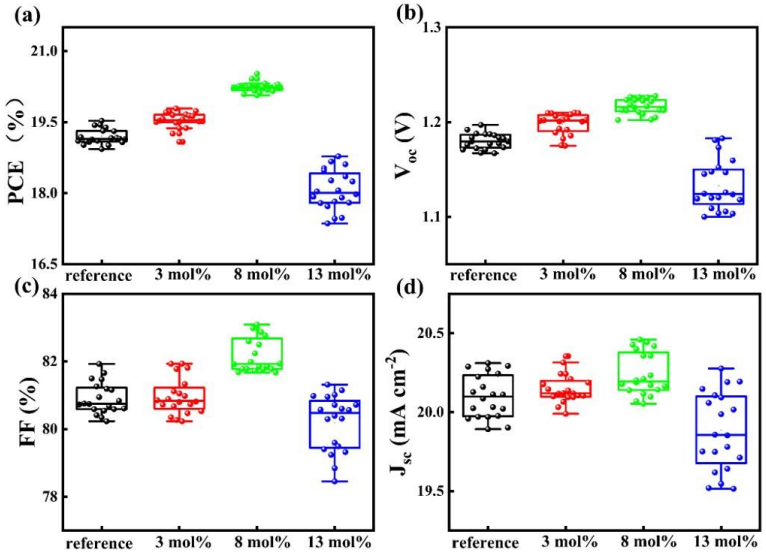

Figure S2. Statistics of photovoltaic performance for $CsPbI_{3-x}Br_x$ inorganic PSCs with different concentration of Cd-doping. (a) PCE; (b) V_{OC}; (c) FF; (d) $J_{SC.}$

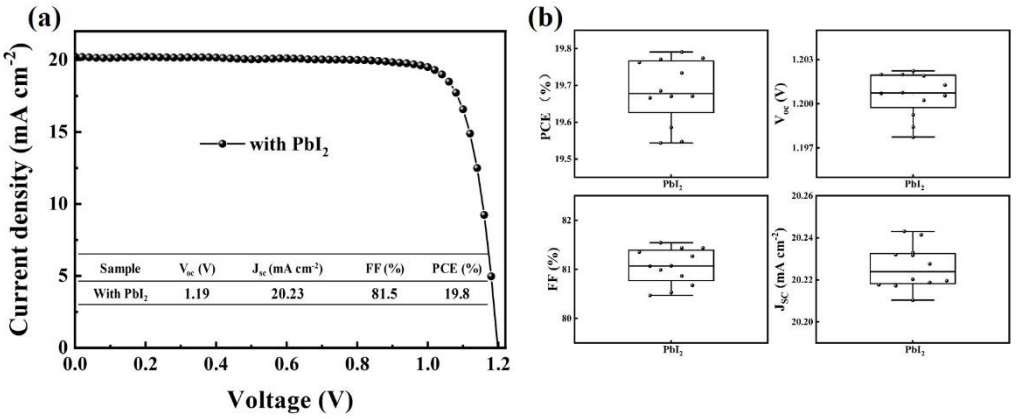

Figure S3. (a) Photovoltaic performance for $CsPbI_{3-x}Br_x$ inorganic perovskite solar cells with 8 mol% PbI_2 as additive. The measurement was performed under one sun irradiation with an aperture area of 0.09 cm^2. (b) The statistics of photovoltaic performance of PbI_2-containing PSCs.

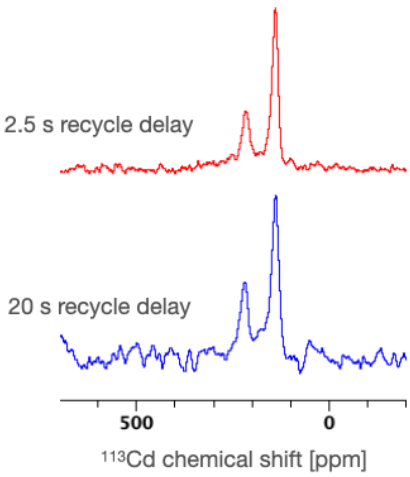

Figure S4. ^{113}Cd MAS NMR spectra of $CsPb_{0.92}Cd_{0.08}I_{2.8}Br_{0.2}$ made by solid-state mechanosynthesis, recorded with a shorter (2.5 s; 32768 scans, 22.8 h total acquisition time) and longer (20 s; 2340 scans, 13 h total acquisition time) recycle delay.

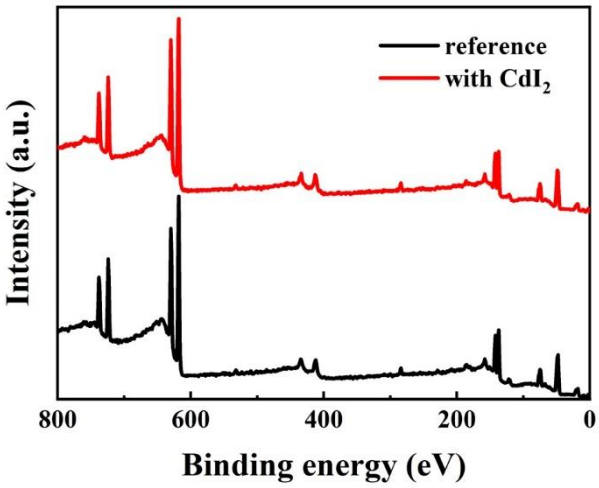

Figure S5. XPS full scan of the $CsPbI_{3-x}Br_x$ inorganic perovskite films without and with CdI_2.

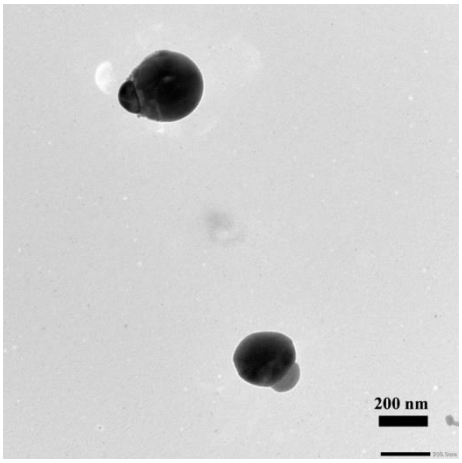

Figure S6. TEM image of $CsPbI_{3-x}Br_x$ perovskite sample with Cd-doping. The scale bar is 200 nm.

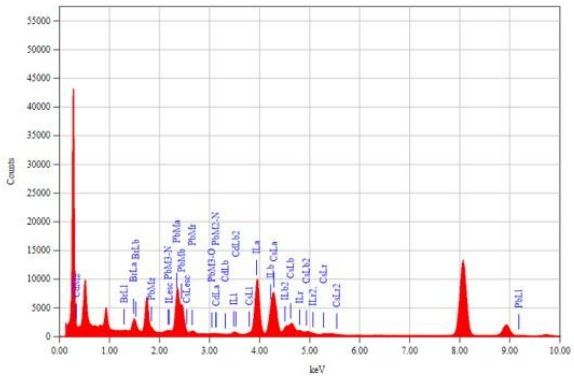

Figure S7. Energy-dispersive X-ray (EDS in a TEM) elemental mapping of $CsPbI_{3-x}Br_x$ perovskite sample with 8 mol% Cd-doping.

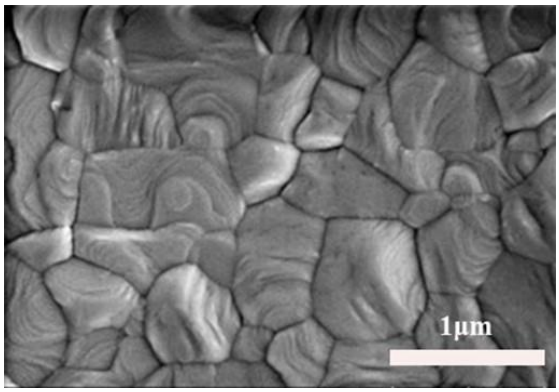

Figure S8. Top-view SEM image of $CsPbI_{3-x}Br_x$ perovskite film with 8 mol% PbI_2 addition. The scale bar is 1 μm.

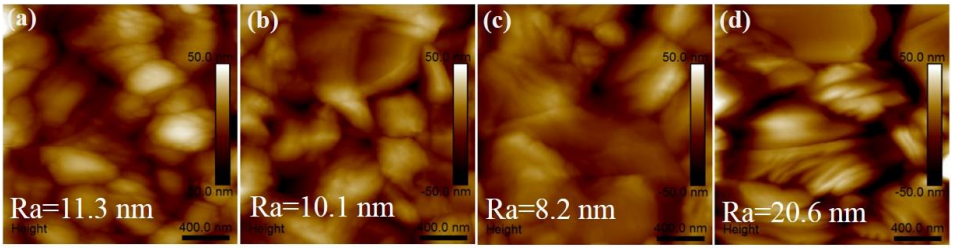

Figure S9. AFM characterization of $CsPbI_{3-x}Br_x$ perovskite film with different concentration of CdI_2. The scale bar is 400 nm. Ra represents root mean square (RMS). (a-d): reference; 3 mol%; 8 mol%; 13 mol%.

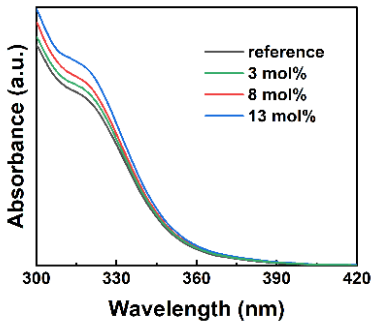

Figure S10 UV-vis spectra of perovskite precursor solution with different concentrations of CdI_2.

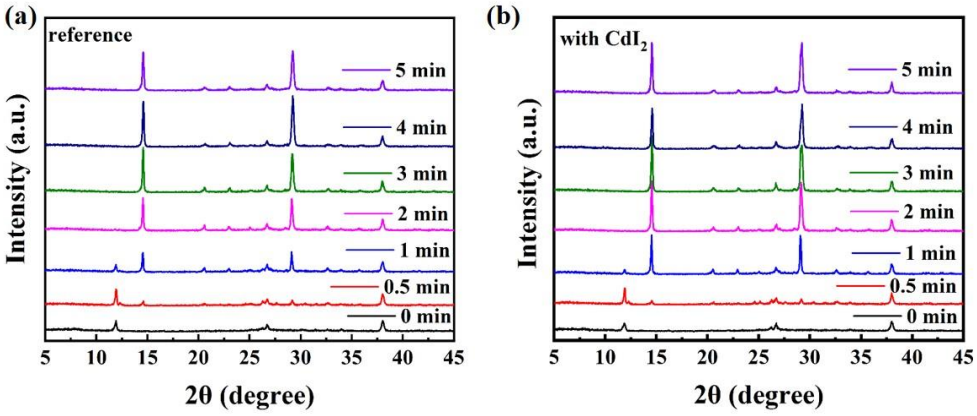

Figure S11. In-situ XRD characterization of $CsPbI_{3-x}Br_x$ perovskite film with different annealing time. (a) reference, (b) with 8 mol% CdI_2.

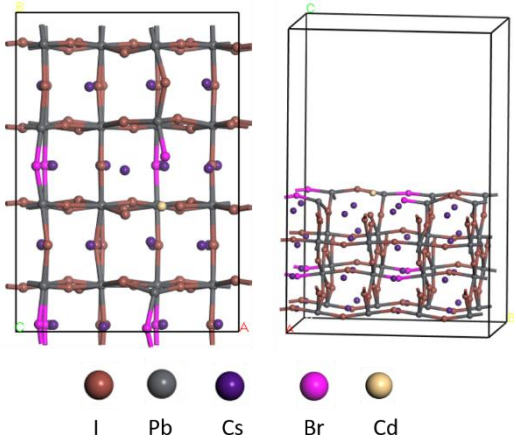

Figure S12. Top view of the iodide vacancy defects upon Cd-doping in (100) CsPbCdIBr-slab using DFT calculation.

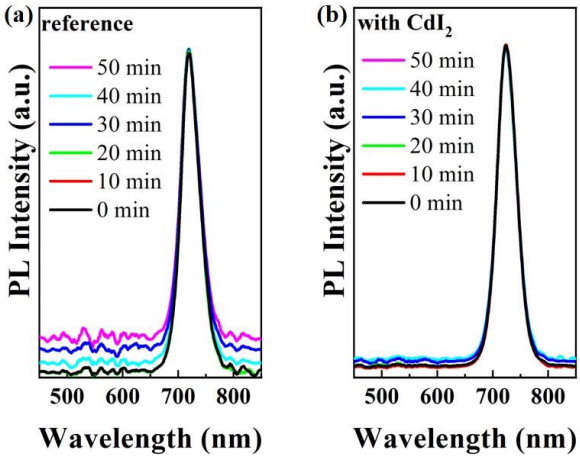

Figure S13. Time dependence of PL curves of perovskite films (a) reference; (b) with 8 mol% CdI_2.

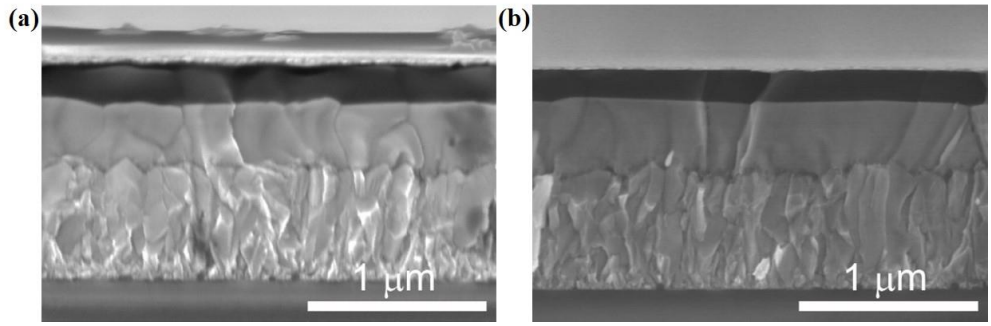

Figure S14. Cross-sectional SEM images of inorganic PSCs (a) reference; (b) with Cd-doping. The scale bar is 1 μm.

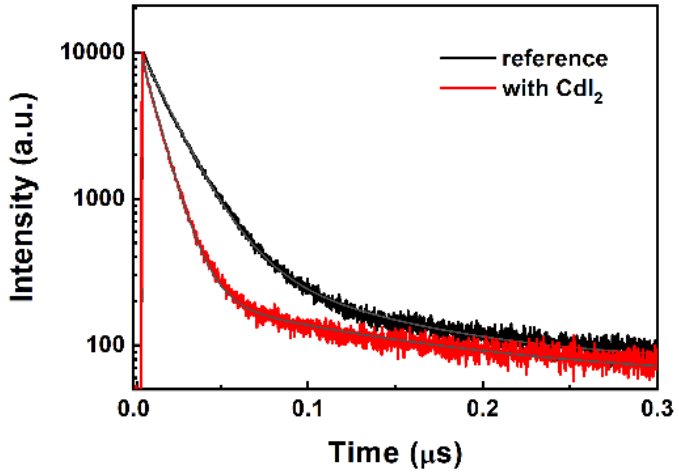

Figure S15 TRPL curves of perovskite with and without CdI_2 addition. The films were prepared on TiO_2 substrate.

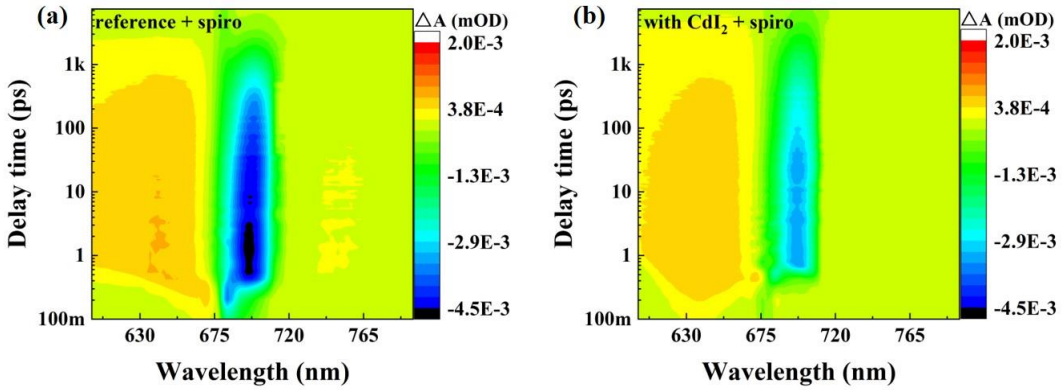

Figure S16. 2D contour plot of TAS of the photoinduced absorption (ΔA) as a function of wavelength and delay time for films a) reference; b) with Cd-doping. Both perovskite films were capped with spiro-OMeTAD hole transport layer.

Figure S17. Stability of $CsPbI_{3-x}Br_x$ perovskite films with different CdI_2 concentrations. These films were stored in an environment with a RH between 40% and 50%. (1: reference; 2: 3 mol%; 3: 8 mol%; 4: 13 mol%)

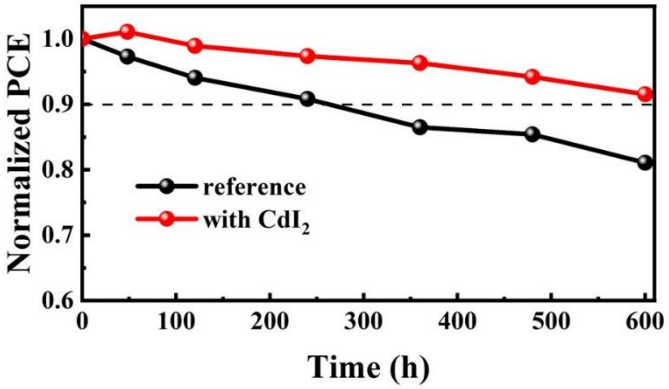

Figure S18. Device stability without and with 8 mol% CdI_2 addition. The devices were stored in an ambient environment with a RH of 20%.

Table S1. ^{113}Cd NMR acquisition and processing parameters for the materials shown in Figures 2d and S4. Magnetic field strength: 11.7 T, MAS spin rate: 12.5 kHz.

Material	Pulse sequence	Number of scans	Recycle delay (s)	Total experiment time (h)	Apodization (Hz)
$CsPb_{0.92}Cd_{0.08}I_{2.8}Br_{0.2}$	Hahn echo	32,768	2.5	22.8	1000
$CsPb_{0.92}Cd_{0.08}I_{2.8}Br_{0.2}$	Echo with tanh/tan refocusing	2,654	20	14.7	1000
$Cs_2CdI_{3.73}Br_{0.27}$	Echo with tanh/tan refocusing	2,695	2.5	1.9	500

Table S2. ^{133}Cs NMR acquisition and processing parameters for $CsPb_{0.92}Cd_{0.08}I_{2.8}Br_{0.2}$ in Figures 2d and 2e. Magnetic field strength: 11.7 T, MAS spin rate: 12.5 kHz.

Material	Pulse sequence	Number of scans	Recycle delay (s)	Total experiment time (h)	Apodization (Hz)
$CsPb_{0.92}Cd_{0.08}I_{2.8}Br_{0.2}$	Hahn echo	4	600	0.7	100
$CsPbI_{2.8}Br_{0.2}$	Hahn echo	8	600	1.3	100
$Cs_2CdI_{3.73}Br_{0.27}$	Hahn echo	4	300	0.3	50

Table S3 Hysteresis behavior of PSCs with and without CdI_2. The measurement was carried out under 100 mW cm^{-2} light intensity with a scan rate of 30 mV s $^{-1}$. The aperture area is 0.09 cm^2.

Sample	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
reference-reverse	1.18	20.14	81.9	19.4
reference -forward	1.16	20.14	80.1	18.7
with CdI_2-reverse	1.21	20.64	83.2	20.8
with CdI_2-forward	1.19	20.68	82.9	20.3

Table S4 The optical properties of perovskite thin films with and without CdI_2 addition by UPS characterization.

	$E_{cut-off}$ (eV)	E_V-E_F (eV)	E_F (eV)	E_V (eV)	E_C (eV)	E_g (eV)
reference	17.28	1.34	3.94	5.28	3.53	1.75
with CdI_2	17.48	1.43	3.74	5.17	3.43	1.74

Table S5 Charge lifetimes within perovskite films with and without CdI_2 addition from TRPL measurement.

	A_1 (%)	T_1 (ns)	A_1 (%)	T_1 (ns)	T_{ave} (ns)
reference	17.0	46.8	82.9	14.3	27.4

with CdI_2	19.1	124.2	80.8	23.8	79.2

Table S6 Charge lifetimes of perovskite films with and without CdI_2 addition from TRPL measurement. The films were prepared on TiO_2 substrate.

	A_1 (%)	**T_1 (ns)**	**A_1 (%)**	**T_1 (ns)**	**T_{ave} (ns)**
reference	13.56	32.5	86.44	20.3	21.8
with CdI_2	18.01	28.4	81.99	19.7	22.7