Supplementary Data

Strategic Defect Control of Perovskite Nanocrystallites with Octylammonium Iodide Toward Efficient Red Perovskite Light-Emitting Diodes with High Operative Stability

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Table S1. XRD peak parameters from perovskite films with and without OAI.

Sample	$Peak_{(100)}$ (2θ, degree)	$d_{(100)}$ (Å)	$FWHM_{(100)}$ (2θ, degree)	$D_{(100)}$ (nm)	$Peak_{(200)}$ (2θ, degree)	$d_{(200)}$ (Å)	$FWHM_{(200)}$ (2θ, degree)	$D_{(200)}$ (nm)
Pristine	14.32	3.117	0.83357	10.04	28.92	1.593	1.9315	4.44
2mol% OAI	14.34	3.113	0.63848	13.10	28.98	1.591	1.06925	8.02

Table S2. Fluorescence analysis parameters of perovskite films based on multi-ligand systems with varied OAI concentration.

Con. OAI (mol%)	Steady-state PL		TRPL				
	λ_{PL} (nm)	$FWHM_{PL}$ (nm)	τ_1 (ns)	τ_2 (ns)	f_1 (%)	f_2 (%)	τ_{avg} (ns)
N/A	643	53.23	2.17	10.41	63.90	36.10	5.15
1	640	45.84	2.49	12.75	55.49	44.51	7.06
2	639	45.37	3.23	25.30	40.14	59.86	16.44
3	637	47.57	2.63	12.63	58.98	41.02	6.73
4	634	48.26	2.25	11.33	58.15	41.85	6.05

Table S3. Summary of PL and EL properties of perovskite films and corresponding PeLEDs with varied concentration of OAI.

Con. OAI (mol%)	V_{on} (V)	EL_{Max} (cd m^{-2})	EQE (%)	CIE (x, y)	LT50 (min)
N/A	1.59	685.08	0.92	(0.715, 0.284)	348
1	1.59	840.60	2.44	(0.712, 0.287)	-
2	1.59	962.15	8.62	(0.711, 0.288)	1080
3	1.59	783.00	5.58	(0.712, 0.287)	-
4	1.59	778.33	3.72	(0.712, 0.288)	-

Table S4. Summary of EL properties of red-emitting PeLEDs reported elsewhere.

Ref.	Perovskite composition	V_{on} (V)	EL_{Max} (cd/m^2)	λ_{EL} (nm)	FWHM (nm)	EQE (%)	LT50
This work	OAI(PEAI, IBAB, and PDAB) $CsPbI_xBr_{3-x}$	1.59	962	657	35.5	8.62	18 h
S1	$(PEA/m\text{-}F\text{-}PEA)_xNMA_{1-x}CsPb_2I_7$	2.5	1300	680	39	25.8	34 min
S2	$CsPbI_{3-x}Br_x$	2.6	3100	666	-	21.2	4807 h
S3	$FA_{0.33}Cs_{0.67}Pb(I_{0.7}Br_{0.3})_3$	3	400	694	37	20.9	14 h
S4	$CsPbI_3$	2.9	60	692	36	14.8	20 h
S5	$CsPbI_{3-x}Br_x$	2.9	3100	637	-	4.5	45 min
S6	$CsPbI_{3-x}Br_x$	1.6	2859	659	-	8.94	2.2 min
S7	$FA_{0.47}Cs_{0.53}Pb(I_{0.87}Br_{0.13})_3$	3.3	8547	662	-	4.9	3 h
S8	$FA_xCs_{1-x}Pb(I_yBr_{1-y})_3$	1.6	1000	692	32	17.1	563 min
S9	$CsPbI_3$	2.0	440	688	-	3.7	5 h
S10	$POEA_2Cs_{n-1}Pb_nI_{3n+1}$	2.5	2545	653	-	18.5	131 min
S11	$PEA_2(Cs_{0.3}MA_{0.7})_2(Zn_xPb_{1-x})_3I_{10}$	1.7	453	658	-	9.5	3.2 min
S12	$CsPbBr_{0.6}I_{2.4}$	1.6	1359	670	34	9.1	20 h
S13	$(PEA{:}NMA)_2Cs_{n-1}Pb_nI_{3n+1}$	2.8	1453	635	42	12.41	103 min
S14	$(PBA_xMBZA_{1-x})_2Cs_{n-1}Pb_nI_{3n+1}$	3.0	1724	640	42	10.8	25 min
S15	$CsPbI_{3-x}Br_x$	1.9	10745	671	28	17.03	15.4 h
S16	$CsPbI_3$	2.0	340	700	-	10.4	20 min
S17	$CsPbI_3$	2.6	1272	699	-	15.03	1.7 h
S18	$Cs_{1-x}EA_xPbI_3$	1.7	403	694	-	17.5	4 min
S19	$CsPbI_3$	3.2	800	691	36	14.8	6 h
S20	$FA_{0.47}Cs_{0.53}Pb(I_{0.87}Br_{0.13})_3$	2.0	1408	690	34	8.7	8 h
S21	$CsPbBr_{0.6}I_{2.4}$	1.6	7798	683	-	7.8	95 min
S22	$CsPbI_3$	2.8	210	682	-	8.65	6 h
S23	$POEA_2CsPb_2I_7$	2.8	2377	650	42	18.7	476 min

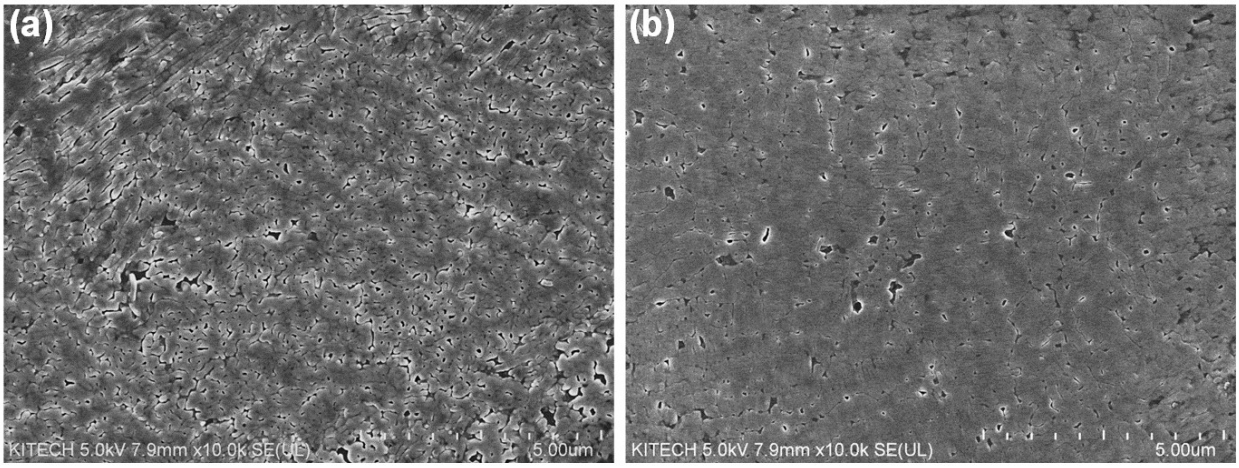

Figure S1. (a) SEM image with lower magnification for pristine and (b) OAI-modified perovskite films.

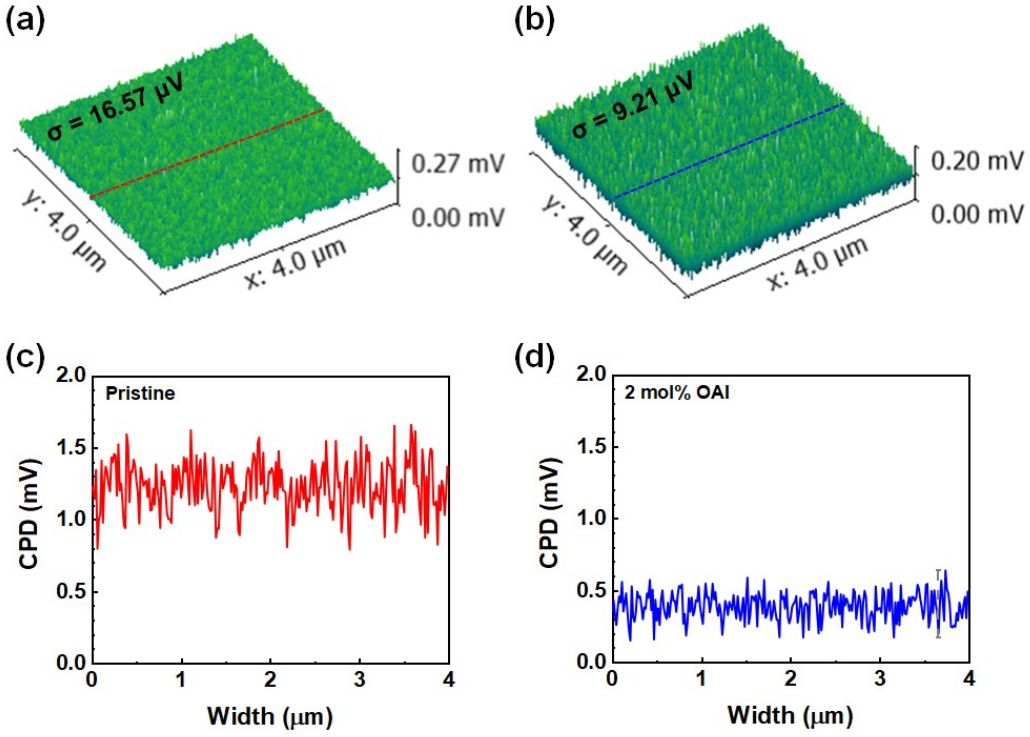

Figure S2. (a) 3D view of KPFM images for the pristine perovskite film and (b) OAI-modified perovskite films. (c) Line profile of CPD for pristine and (d) OAI-modified film.

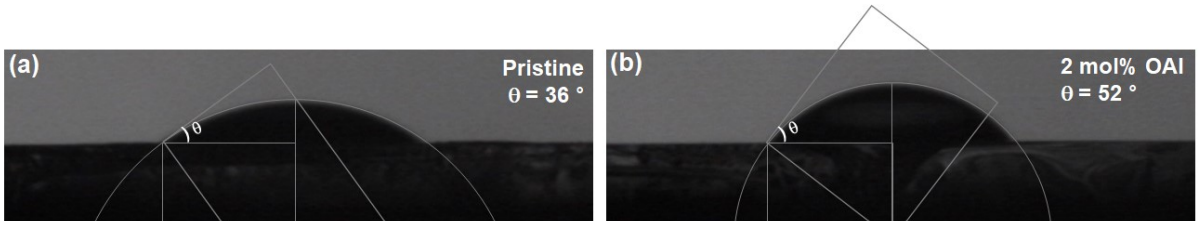

Figure S3. Contact angle measurement of water droplet onto the perovskite films without and with OAI incorporation (2 mol%).

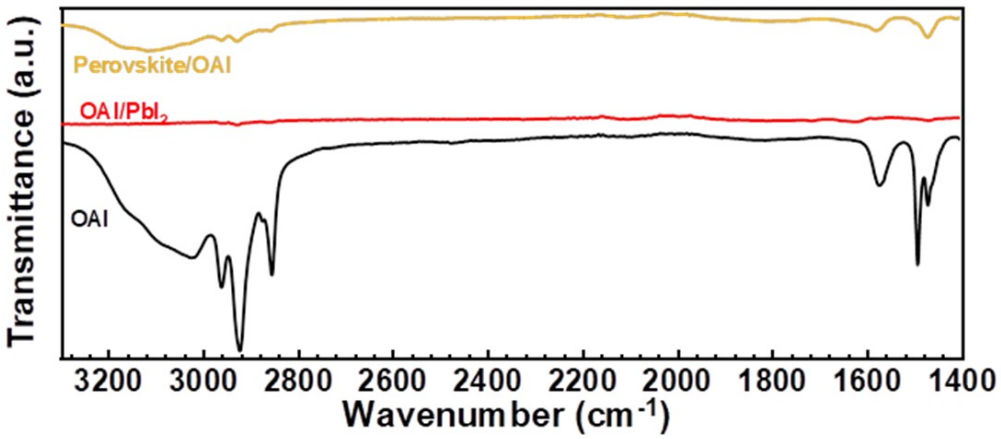

Figure S4. FT-IR full-scan spectra of OAI, OAI mixed with PbI_2, and OAI mixed with perovskite films.

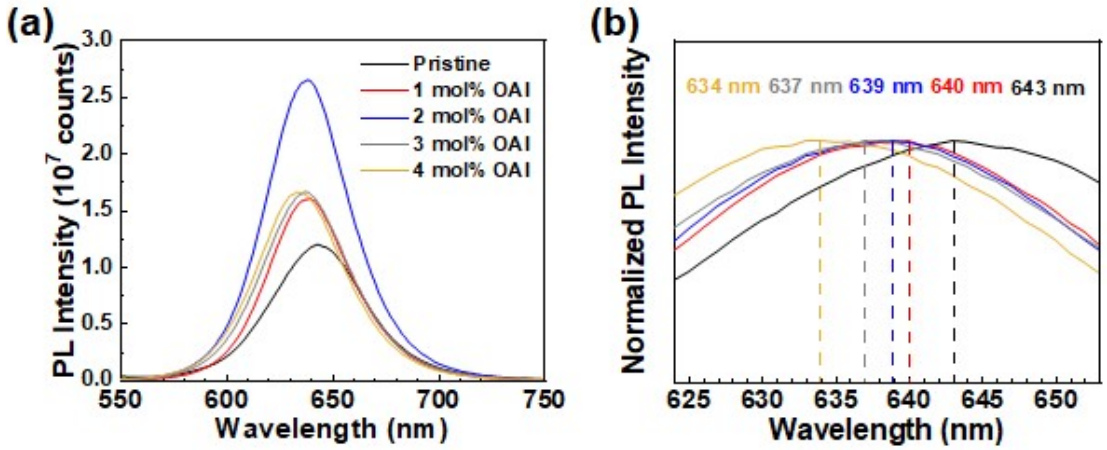

Figure S5. (a) PL spectra, and (b) normalized spectra of perovskite films with varied OAI concentration.

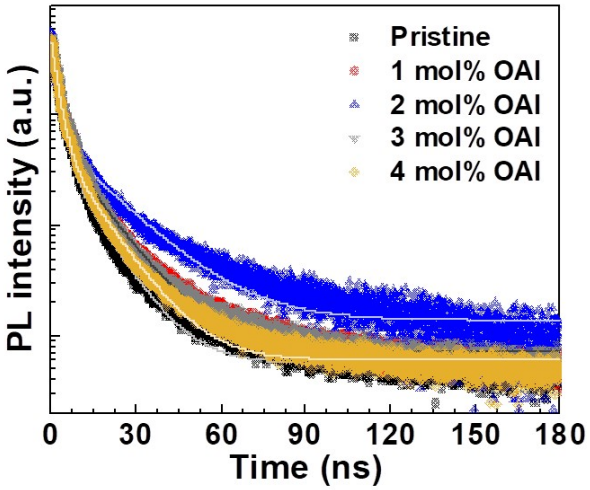

Figure S6. TRPL decay curves of perovskite films with varied OAI concentration.

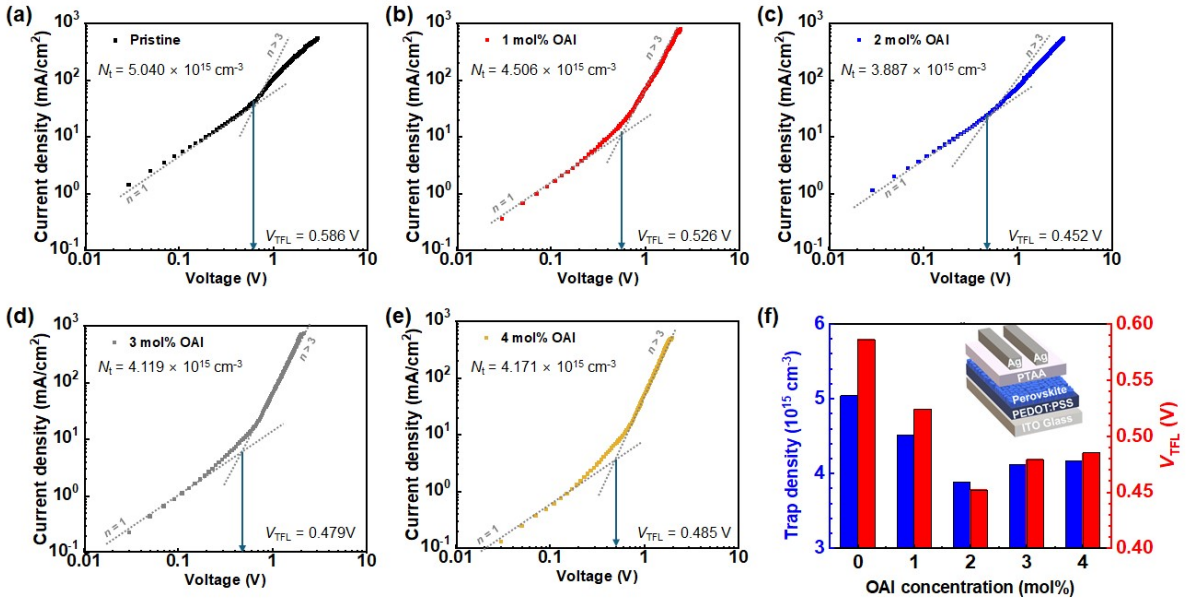

Figure S7. (a) Schematic illustration of hole-only device, and (b) *J-V* curves of hole-only devices based on the perovskite films with and without OAI incorporation.

References

[S1] J. Jiang, Z. Chu, Z. Yin, J. Li, Y. Yang, J. Chen, J. Wu, J. You, X. Zhang, Adv. Mater. 2022, 34, 2204460.

[S2] Y. Ye, Y. Li, X. Cai, W. Zhou, Y. Shen, K. Shen, J. Wang, X. Gao, I. Zhidkov, J. Tang, Adv. Funct. Mater. 2021, 31, 2105813.

[S3] Z. Fang, W. Chen, Y. Shi, J. Zhao, S. Chu, J. Zhang, Z. Xiao, Adv. Funct. Mater. 2020, 30, 1909754.

[S4] G. Cheng, Y. Liu, T. Chen, W. Chen, Z. Fang, J. Zhang, L. Ding, X. Li, T. Shi, Z. Xiao, ACS Appl. Mater. Interfaces 2020, 12, 18084.

[S5] K. Wang, L. Wang, Y. Liu, Y. Song, Y. Yin, J. Yao, J. Yang, J. Wang, L. Feng, Q. Zhang, Q. Zhang, H. Yao, Adv. Opt. Mater. 2020, 2001684.

[S6] M. Jiang, Z. Hu, L. K. Ono, Y. Qi, Nano Res. 2021, 14, 191. 17

[S7] Y. Ke, N. Wang, D. Kong, Y. Cao, Y. He, L. Zhu, Y. Wang, C. Xue, Q. Peng, F. Gao, W. Huang, J. Wang, J. Phys. Chem. Lett. 2019, 10, 380.

[S8] Y.-C. Ye, Y. Li, Y. Tian, X.-Y. Cai, Y. Shen, K.-C. Shen, X. Gao, F. Song, W. Wang, J.- X. Tang, Nanoscale 2021, 13, 340.

[S9] S. Zhang, C. Yi, N. Wang, Y. Sun, W. Zou, Y. Wei, Y. Cao, Y. Miao, R. Li, Y. Yin, N. Zhao, J. Wang, W. Huang, Adv. Mater. 2017, 29, 1606600.

[S10] X. Liang, Z. Liu, J. Zhang, H. Chen, Q. Gu, W. Zhang, C. Shen, Z. Xiao, Y. Wang, J. Liao, X. Wen, J. Xie, L. Yao, W. Cai, Y. Mo, J Qing, S. Su, L. Hou

[S11] D. Liu, X. Liu, G. Sun, F. Meng, Z. Liu, C. Shen, M. Li, S.J. Su, ACS Appl. Mater. Interfaces 2021, 13, 55412.

[S12] P. Liu, W. Cai, C. Zhao, S. Zhang, P. Nie, W. Xu, H. Meng, H. Fu, G. Wei, Adv. opt. mater. 2021, 9, 2101419.

[S13] L. Yang, Y. Zhang, J. Ma, P. Chen, Y. Yu, M. Shao, ACS Energy Lett. 2021, 6, 2386.

[S14] J. Qing, S. Ramesh, Q. Xu, X.K. Liu, H. Wang, Z. Yuan, Z. Chen, L. Hou, T.C. Sum, F. Gao, Adv. Mater. 2021, 33, 2104381.

[S15] N. Li, S. Apergi, C. Chan, Y. Jia, F. Xie, Q. Liang, G. Li, K. Wong, G. Brocks, S. Tao, N. Zhao, Adv. Mater. 2022, 34, 2202042.

[S16] C. Yi, C. Liu, K. Wen, X.-K. Liu, H. Zhang, Y. Yu, N. Fan, F. Ji, C. Kuang, B. Ma, C. Tu, Y. Zhang, C. Xue, R. Li, F. Gao, W. Huang, J. Wang, Nat. Commun. 2020, 11, 4736.

[S17] Y. Miao, X. Liu, Y. Chen, T. Zhang, T. Wang, Y. Zhao, Adv. Mater. 2021, 33, 2105699.

[S18] G. Sun, X. Liu, Z. Liu, D. Liu, F. Meng, Z. Li, L. Chu, W. Qiu, X. Peng, W. Xie, C. Shen, J. Chen, H.-L. Yip, S.-J. Su, Adv. Funct. Mater. 2021, 31, 2106691.

[S19] G. Cheng, Y. Liu, T. Chen, W. Chen, Z. Fang, J. Zhang, L. Ding, X. Li, T. Shi, Z. Xiao, ACS Appl. Mater. Interfaces 2020, 12, 18084.

[S20] Y. Ke, N. Wang, D. Kong, Y. Cao, Y. He, L. Zhu, Y. Wang, C. Xue, Q. Peng, F. Gao, W. Huang, J. Wang, J. Phys. Chem. Lett. 2019, 10, 380.

[S21] H. Shi, Z. Wang, H. Ma, H. Jia, F. Wang, C. Zou, S. Hu, H. Li, Z. Tan, J. Mater. Chem. C 2021, 9, 12367.

[S22] B. Han, B. Cai, Q. Shan, J. Song, J. Li, F. Zhang, J. Chen, T. Fang, Q. Ji, X. Xu, H. Zeng, Adv. Funct. Mater. 2018, 28, 1804285.

[S23] Y. Kuang, L. Yang, J. Ma, T. Bie, D. Zhang, Y. Xue, N. Zhou, M. Shao, ACS Mater. Lett. 2023, 5, 2922-2928.