Supporting Information

Grain and stoichiometry engineering for ultra-sensitive perovskite X-ray detectors

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Detailed procedure for the preparation of the title compound

First, 1 g MAI powder was dissolved in 10 ml IPA at 80 °C，then 2 g of PbI_2 powder was slowly added into 10 ml of MAI solution (100 g/L) in IPA under stirring at 80 °C. After stirring 2 h at this temperature, the solution was centrifuged and washed by IPA three times, and then the precipitate was placed into a drying oven of 60 °C. Finally, black $MAPbI_3$ perovskite powder was obtained. 1 g $MAPbI_3$ powder and 6.4 mg MAI powder (the molar ratio of MAI to $MAPbI_3$ powder is 2.5%) were mixed and fully ground in a mortar. Finally, ground powder was hot-pressed at 250 °C for 1 h as mentioned in pellets preparation.

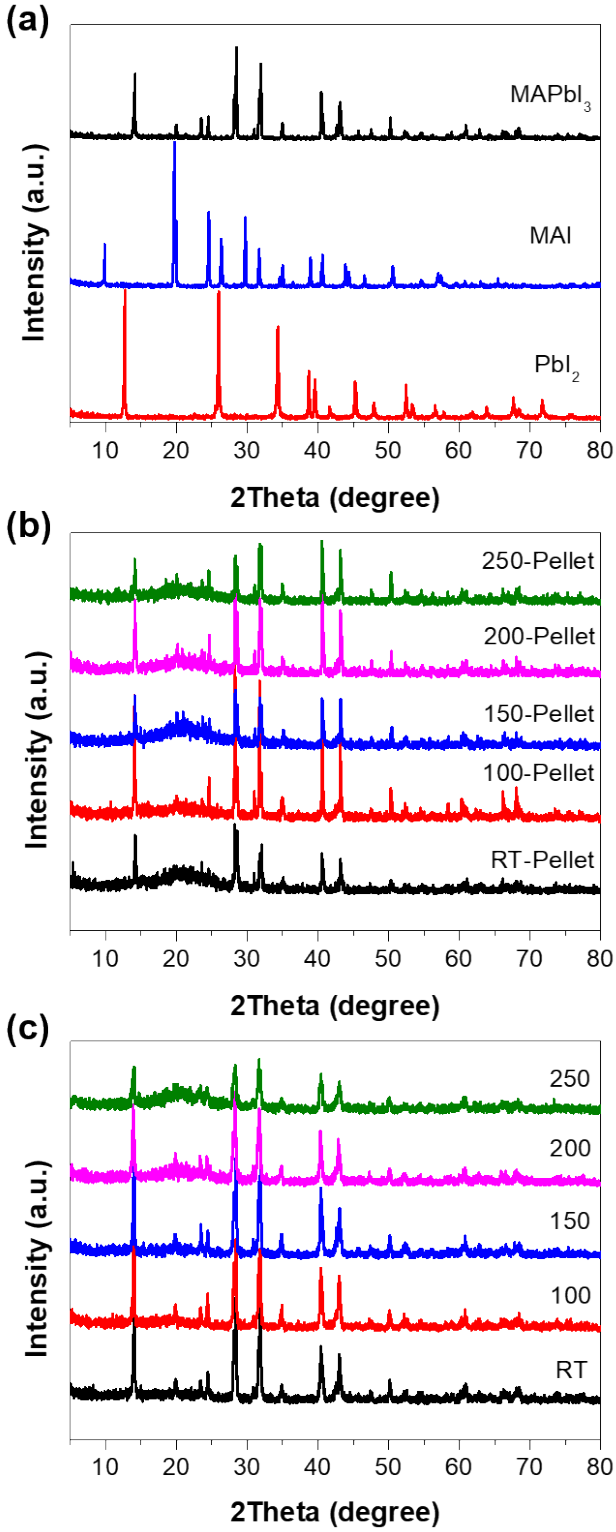

Figure S1. The X-ray diffraction (XRD) patterns of $MAPbI_3$ powder, MAI powder and PbI_2 powder (a), XRD patterns of $MAPbI_3$ pellets obtained at different hot-press temperatures (b) and ground powder from pellets (c).

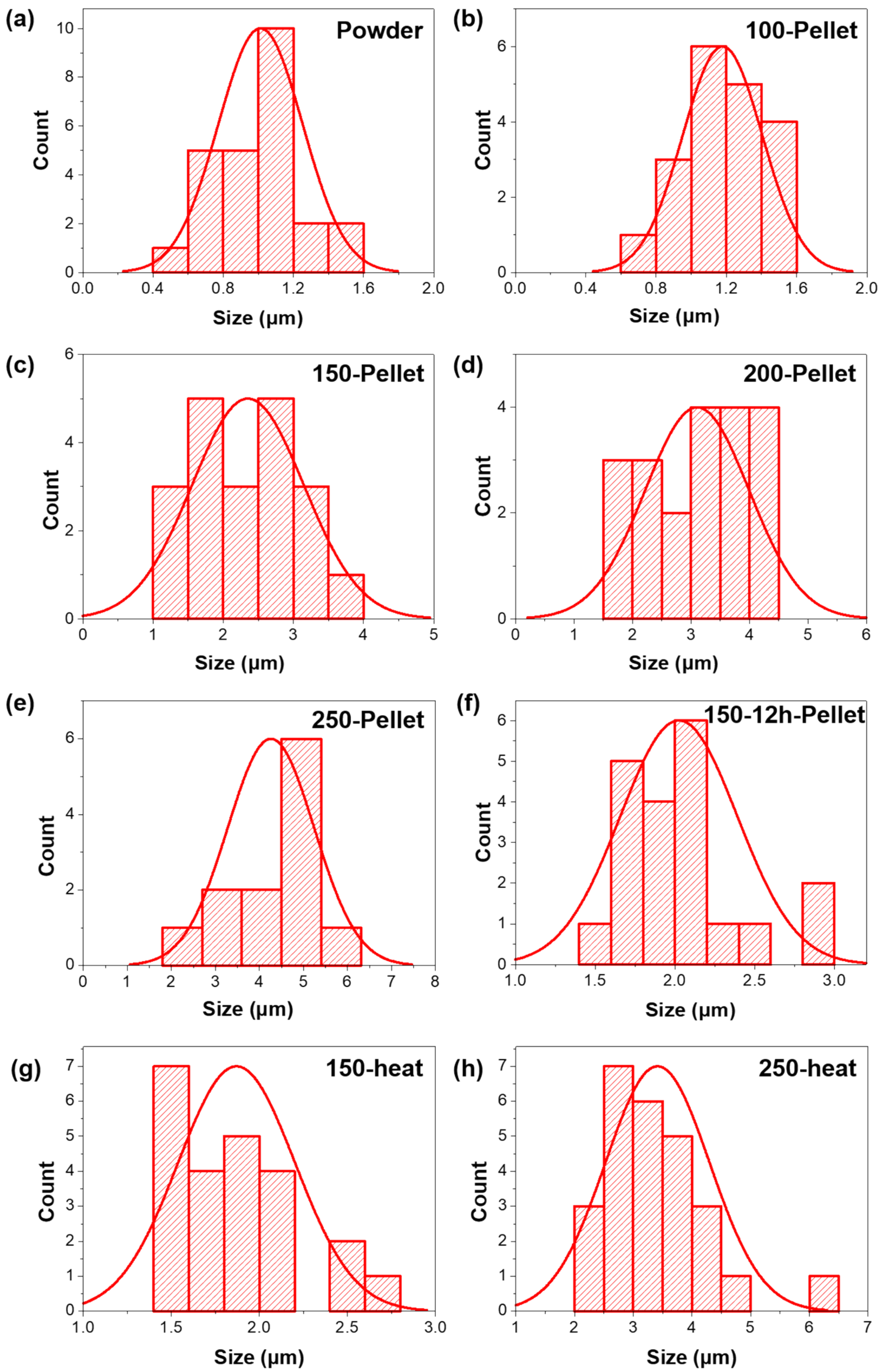

Figure S2. Grain size statistics of (a) powder; (b) 100-Pellet; (c) 150-Pellet; (d) 200-Pellet; (e) 250-Pellet; (f) hot-pressed at 150 ˚C for 12 h, (g) heat treatment at 150 ˚C and (h) heat treatment at 250 ˚C.

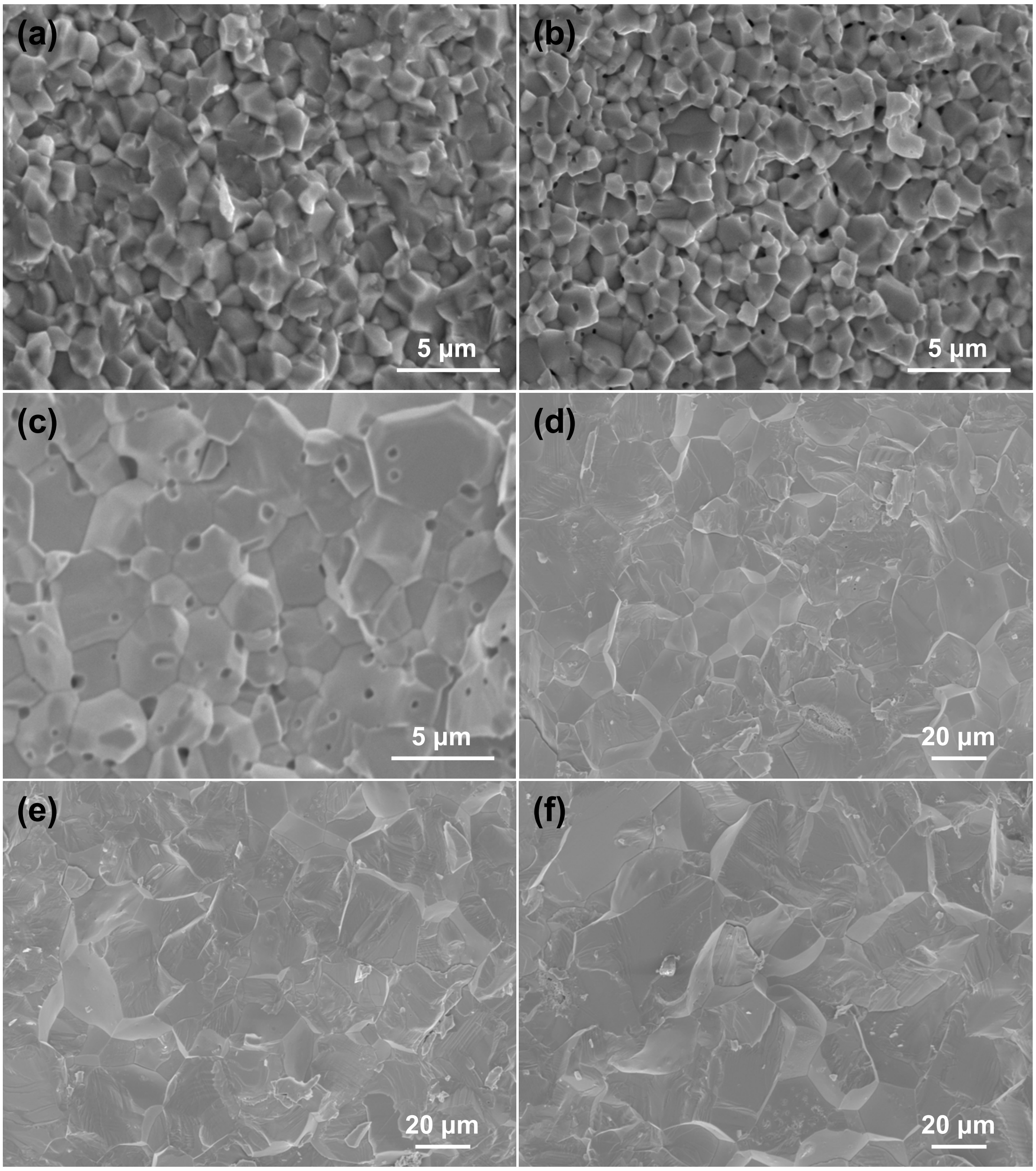

Figure S3. Scanning electron microscopy images of $MAPbI_3$ pellets: (a) 150-heat pellet; (b) 250-heat pellet; (c) hot pressed for 12 h; (d) 1.25-250 pellet; (e) 2.50-250 pellet; (f) 5.00-250 pellet.

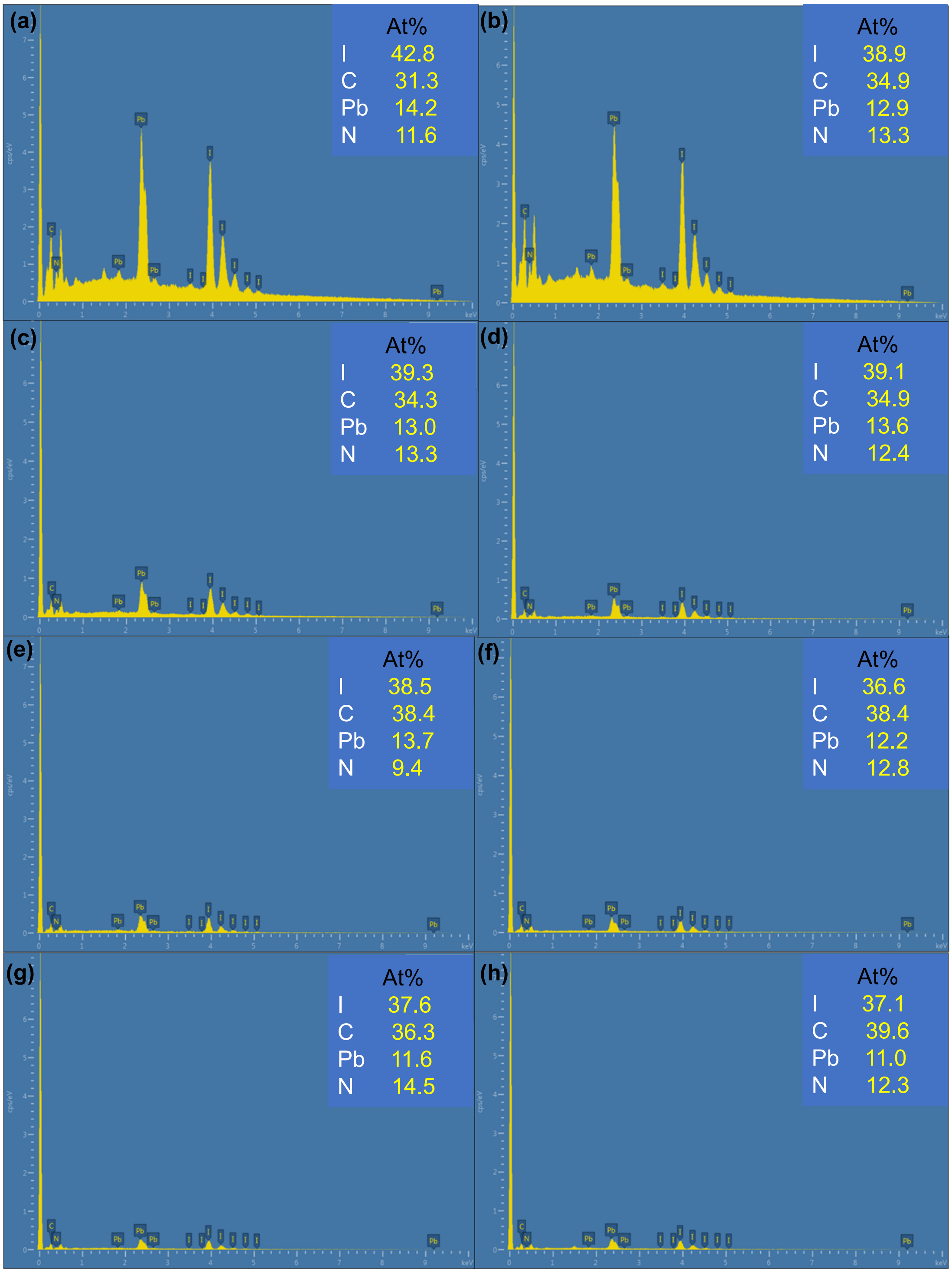

Figure S4. Atomic% of different elements in (a) RT-pellet; (b) 100-pellet; (c) 150-pellet; (d) 200-pellet; (e) 250-pellet; (f) 1.25-250 pellet; (g) 2.50-250 pellet; (h) 5.00-250 pellet.

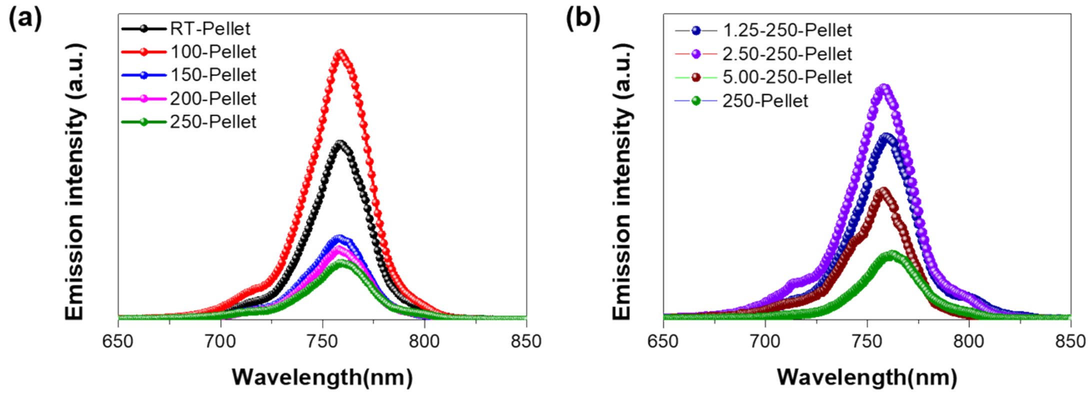

Figure S5. (a) Photoluminescence (PL) of pellets pressed at temperatures varying from RT to 250 °C, and (b) pellets pressed at 250 °C with different amounts of excess MAI from 0 to 5%.

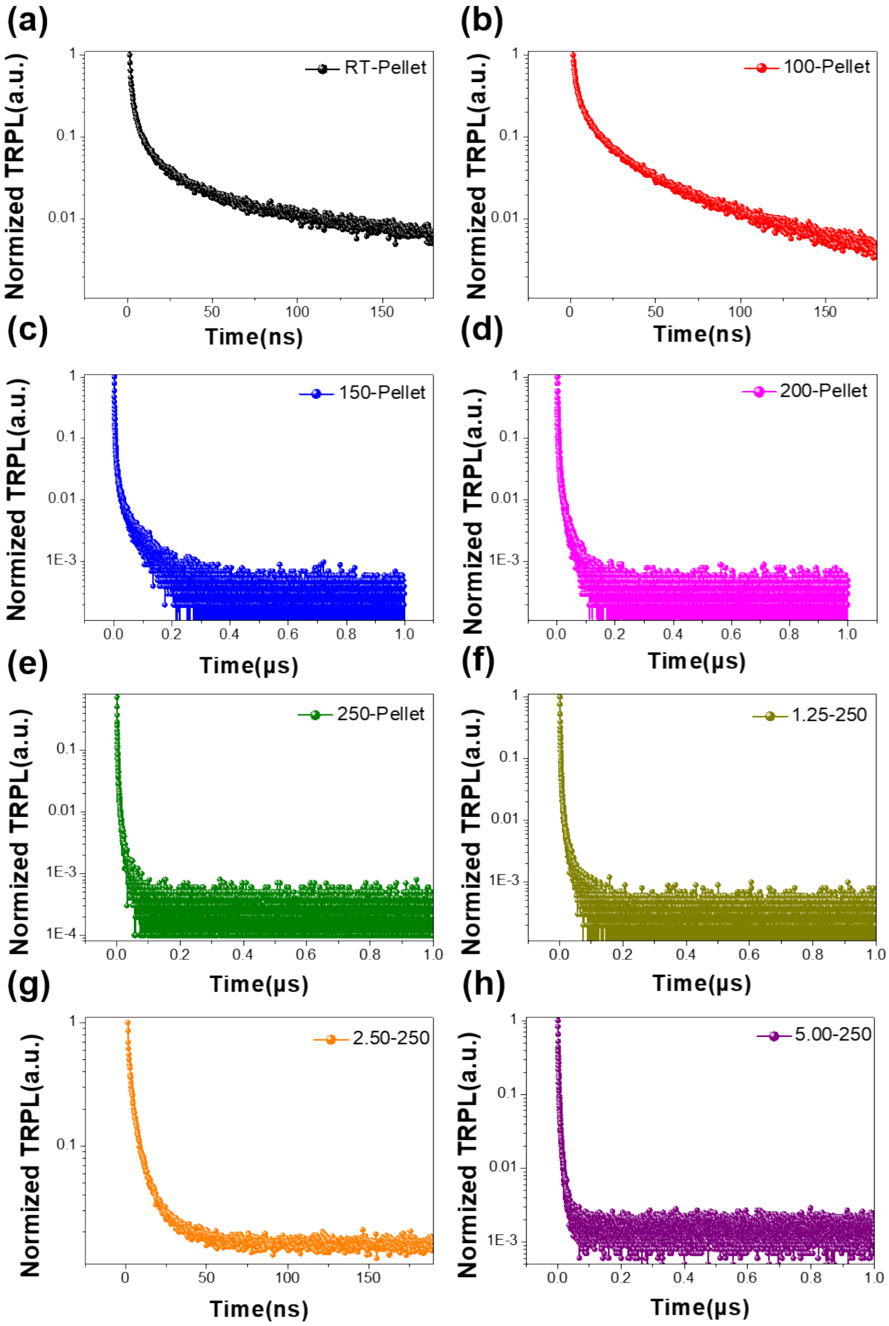

Figure S6. Time-resolved photoluminescence of (a) RT-Pellet; (b) 100-Pellet; (c) 150-Pellet; (d) 200-Pellet; (e) 250-Pellet; (f) 1.25-250; (g) 2.50-250; (h) 5.00-250.

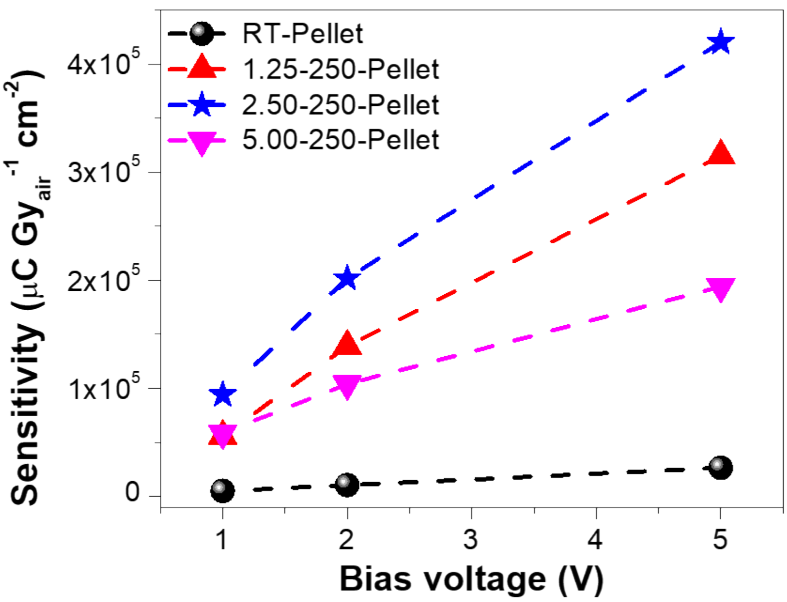

Figure S7. Sensitivities of devices based on RT, 1.25-250, 2.50-250, 5.00-250 pellets at biases of 1 V, 2 V and 5 V

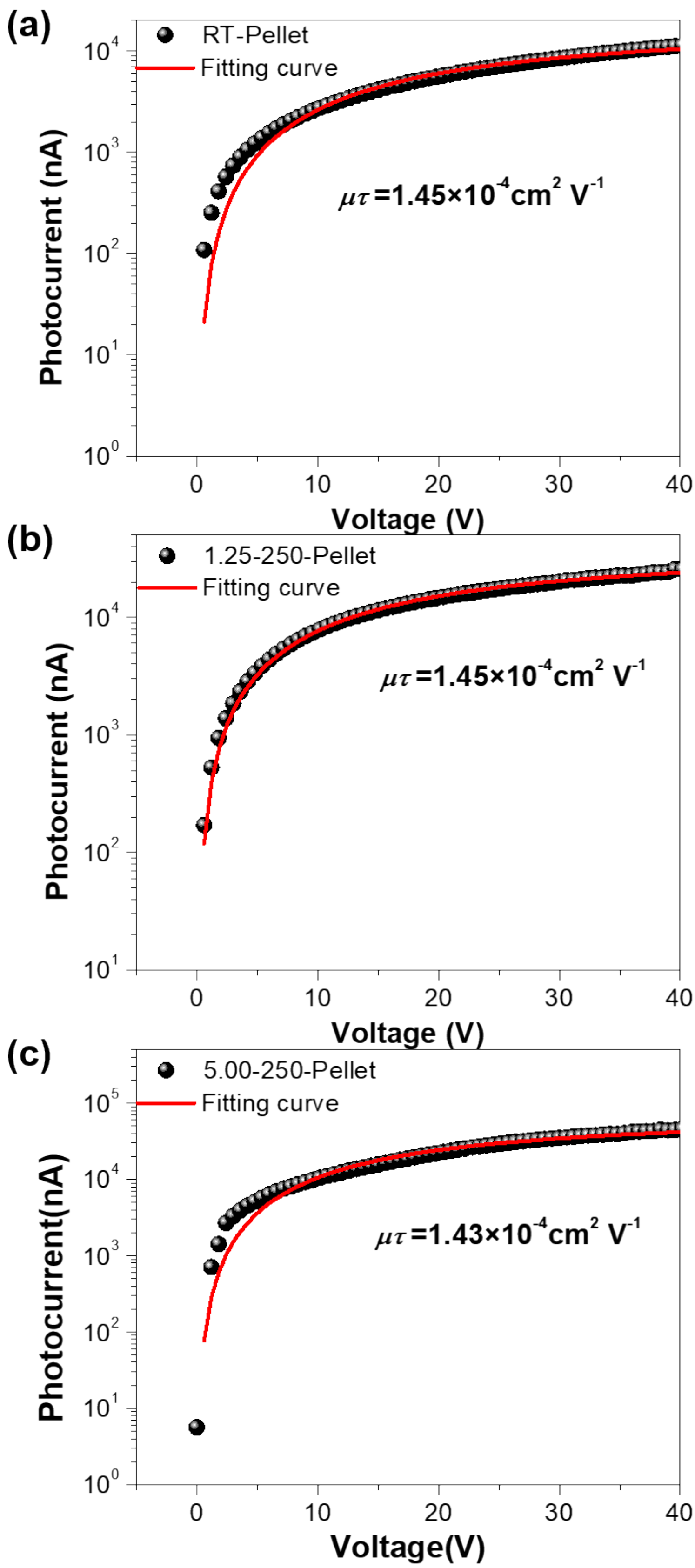

Figure S8. Photoconductivity curves of (a) RT, (b) 1.25-250 and (c) 5.00-250 devices.

Table S1. Summaries of fitting parameters for time-resolved photoluminescence (TRPL) for a cross-section of different pellets.

Sample	Average τ(ns)	τ1 (ns)	Amplitude τ1(%)	τ2 (ns)	Amplitude τ2(%)
RT-Pellet	19.19	28.36	12.97	2.292	87.03
100-Pellet	22.14	29.997	19.46	2.973	80.54
150-Pellet	15.62	34.50	3.77	1.856	96.23
200-Pellet	7.18	15.97	6.00	1.6063	94
250-Pellet	3.07	6.594	7.07	0.766	92.93
1.25-250	4.72	12.184	4.63	1.282	95.37
2.50-250	6.10	8.802	23.38	1.6164	76.62
5.00-250	4.07	8.115	10.64	1.5456	89.36

Table S2. Gain factors of different devices at biases of 1 V, 2 V and 5 V.

Device	Gain at 1V	Gain at 2V	Gain at 5V
RT-Pellet	65.28	140.59	348.64
1.25-250	850.40	2135.44	4930.70
2.50-250	1347.48	2913.25	6126.35
5.00-250	955.17	1720.62	3316.88

Table S3. Key parameters of polycrystalline perovskites-based X-ray detectors

Device structure	E_{ph} (keV)	E (V mm^{-1})	μτ (cm^2 V^{-1})	S (μC $Gy_{air}^{-1}$$cm^{-2}$)	LoD (nGy_{air} s^{-1})	Ref
Ag/ZnO/PCBM/ $MAPbI_3$/PEDOT: PSS/ITO	38	200	2×10^{-4}	2527	-	1
ITO/TFT/PI-$MAPbI_3$/$MAPbI_3$/PI-$MAPbBr_3$/ITO	(100 kV)[a]	60	1×10^{-4}	1.1×10^{4}	-	2
Au/$MAPbI_3$/PCBM/Au	(40 kV) [a]	12.5	3.8×10^{-4}	1.2×10^{5}	-	3
FTO/$CsPbBr_3$/Au	30	5	1.3×10^{-2}	5.6×10^{4}	215	4
Au/$Cs_2AgBiBr_6$/Au	30	500	5.5×10^{-3}	250	95.3	5
Au/$MAPbI_3$/Au	-(40 kV) [a]	1.32	5.46×10^{-3}	9.4×10^{4}	350	this work

[a]Only the acceleration voltage of the X-ray sources rather than its peak photon energy is provided.

References

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