Electronic Supplementary Information

High-performance Perovskite Solar Cells from Large Perovskite Grain Size by the Urea Additive

Qingbin Cai[1], Chao Liang[2], Zhichao Lin[1], Wenqi Zhang[1], Guibin Shen[1], Hongye Dong[1], Xiangning Xu[1], Haoyi Wang[1*], Cheng Mu[1*] and Guichuan Xing[3]

1 Key Laboratory of Advanced Light Conversion Materials and Biophotonics, Department of Chemistry Renmin University of China, Beijing, 100872, P. R. China.

2 MOE Key Laboratory for Nonequilibrium Synthesis and Modulation of Condensed Matter, School of Physics, Xi'an Jiaotong University, Xi'an 710049, P. R. China.

3 Joint Key Laboratory of the Ministry of Education, Institute of Applied Physics and Materials Engineering, University of Macau, Avenida da Universidade, Taipa, Macau 999078, P. R. China.

Table S-1. The fitted parameters of TRPL spectra (excitation at 475 nm) of perovskite films with additive different concentrations of urea deposited on quartz glass.

Sample	τ_{ave}(ns)	A_1 (%)	τ_1(ns)	A_2 (%)	τ_2 (ns)
0M	326.6	43.6	173.3	56.4	386.7
0.1M	944.0	22.5	262.4	77.5	996.1
0.2M	1687.0	4.6	335.6	95.4	1699.9
0.3M	583.2	25.6	213.8	74.4	626.6

Table S2. Calculated perameters and trap density (Nt) of the perovskite films without urea and with 0.2 M urea.

	L (nm)	ε	V_{TFL} (V)	Nt (x10^{15}cm^{-3})
0M	710	42.3	0.41	3.8
0.2M	710	42.3	0.25	2.3

Table S3. Photovoltaic parameters of PSCs devices with additive different concentrations of urea

	J_{sc} (mA/cm^2)	V_{oc} (V)	FF (%)	PCE (%)
0M	23.99±0.04	1.098±0.005	72.6±0.27	19.12±0.4
0.1M	24.42±0.03	1.136±0.007	75.2±0.25	20.86±0.4
0.2M	24.58±0.02	1.142±0.005	78.8±0.23	22.12±0.3
0.3M	24.06±0.04	1.118±0.006	72.7±0.22	19.56±0.3

Table S4. The photovoltaic parameters and hysteresis indexes for PSC without and with 0.2 M urea doping, the hysteresis index is defined as $(PCE_{reverse} - PCE_{forward})/PCE_{reverse}$.

	Direction	J_{SC} (mA/cm^2)	V_{OC} (V)	FF (%)	PCE (%)	Hysteresis index
0 M	reverse	23.99	1.098	72.6	19.12	7%
	forward	23.94	1.082	68.7	17.79	
0.2M	reverse	24.58	1.142	78.8	22.12	3.8%
	forward	24.50	1.136	76.5	21.29	

Table S-5. The fitted parameters for IS measurements PSCs with additive different concentrations of urea

	$R_s(\Omega\ cm^2)$	$R_{rec}(\Omega\ cm^2)$
0M	26.1	2481
0.2M	6.7	5785

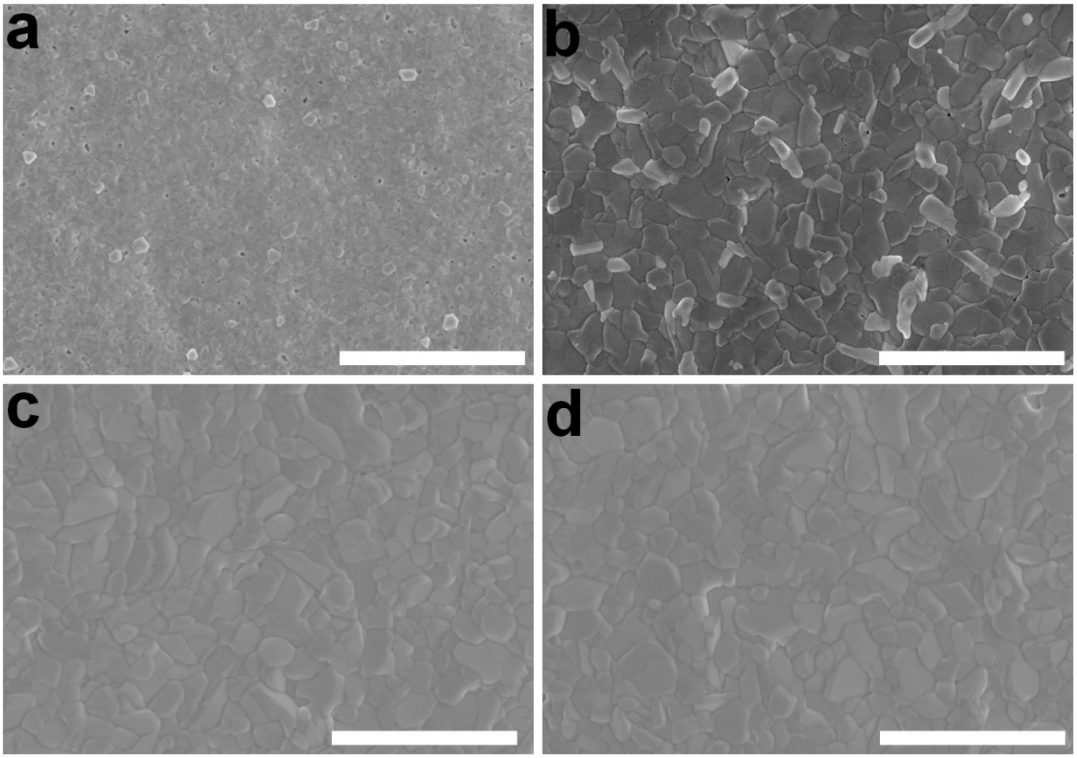


Fig. S1. SEM images of PbI_2 films prepared under different urea concentrations, (a) urea-0 M, (b) urea-0.1M, (c) urea-0.2M, (d) urea-0.3M. The scalebar is 2 μm.

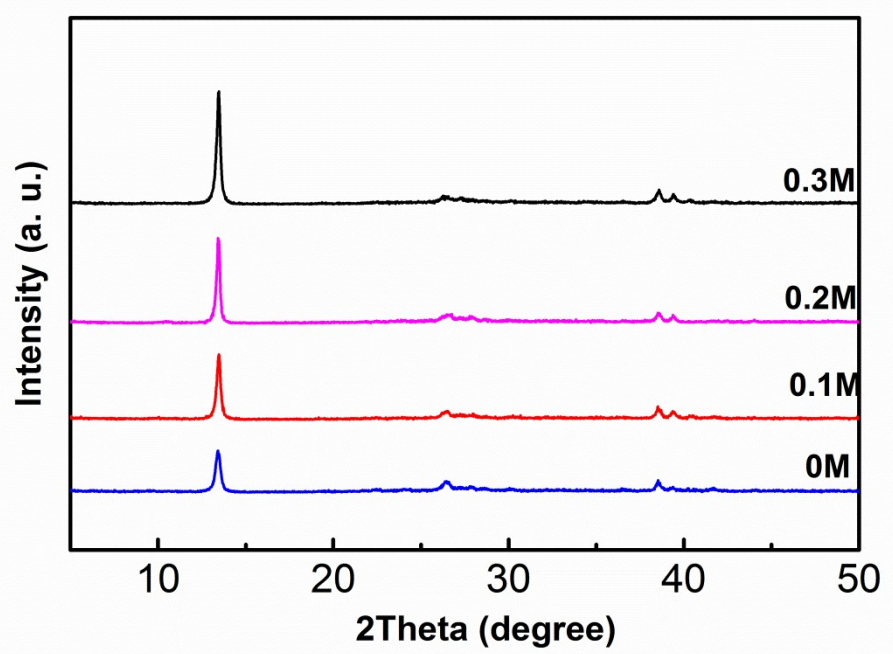


Fig. S2. XRD patterns of PbI_2 films with additive with different concentrations of urea. The FWHM of (001) plane is 0.336 °, 0.272 °, 0.253 ° and 0.247 ° as the urea doping concentration increased, respectively.

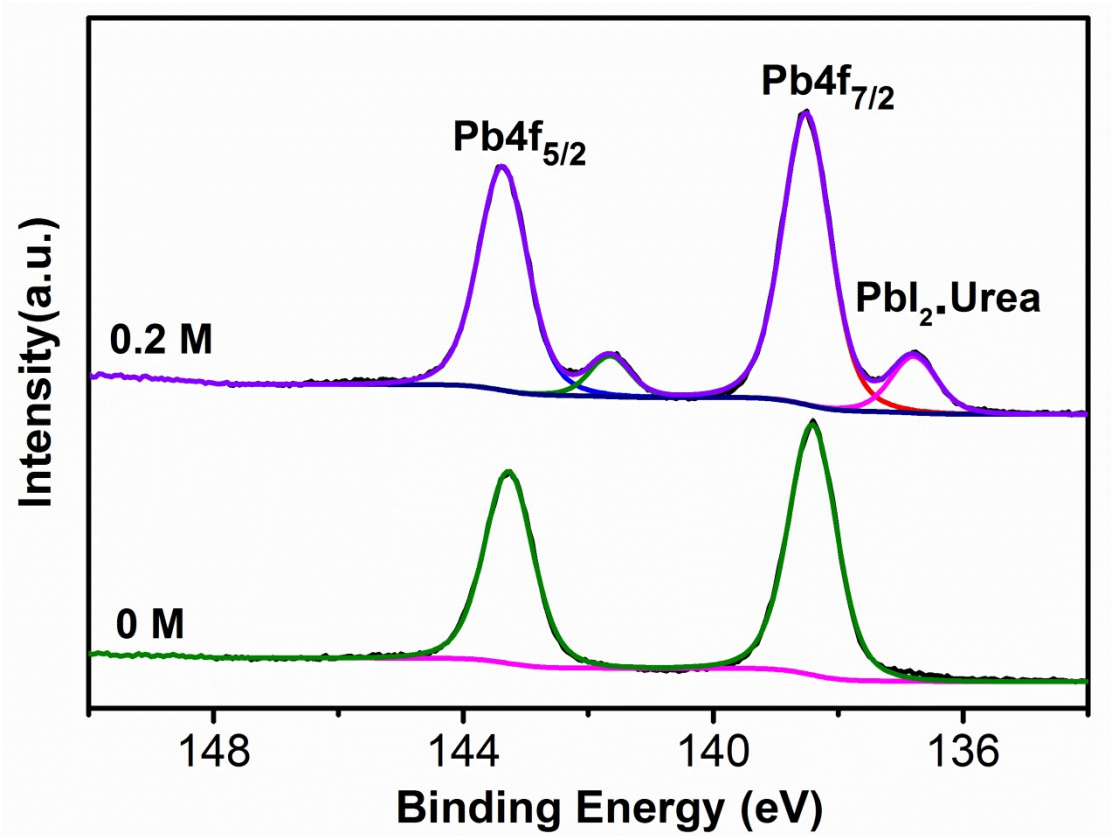


Fig. S3. XPS of PbI_2 films without urea and with 0.2 M urea.

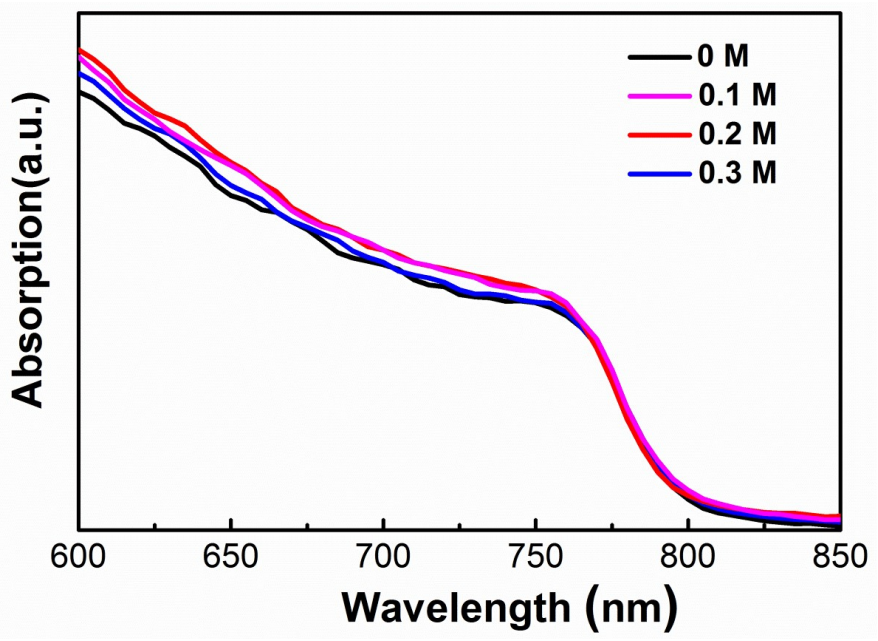
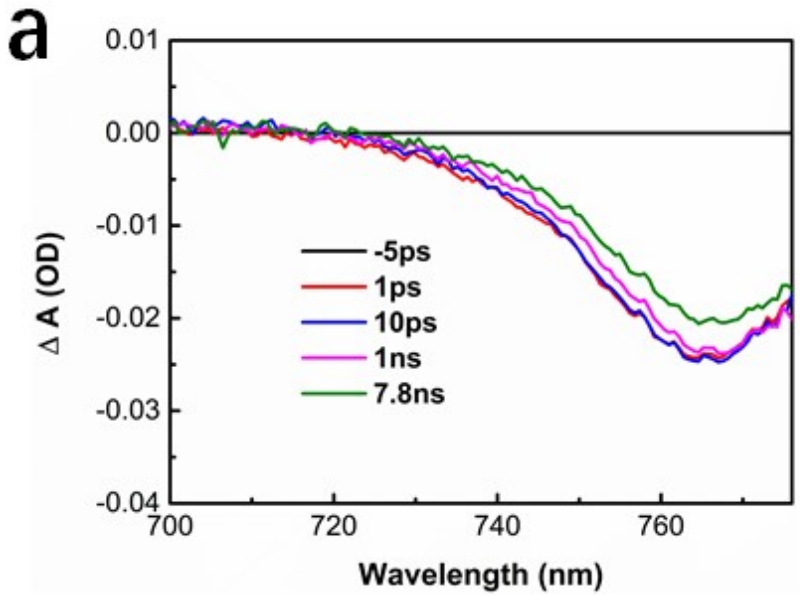


Fig. S4. UV of perovskite films with different concentrations of urea doping.



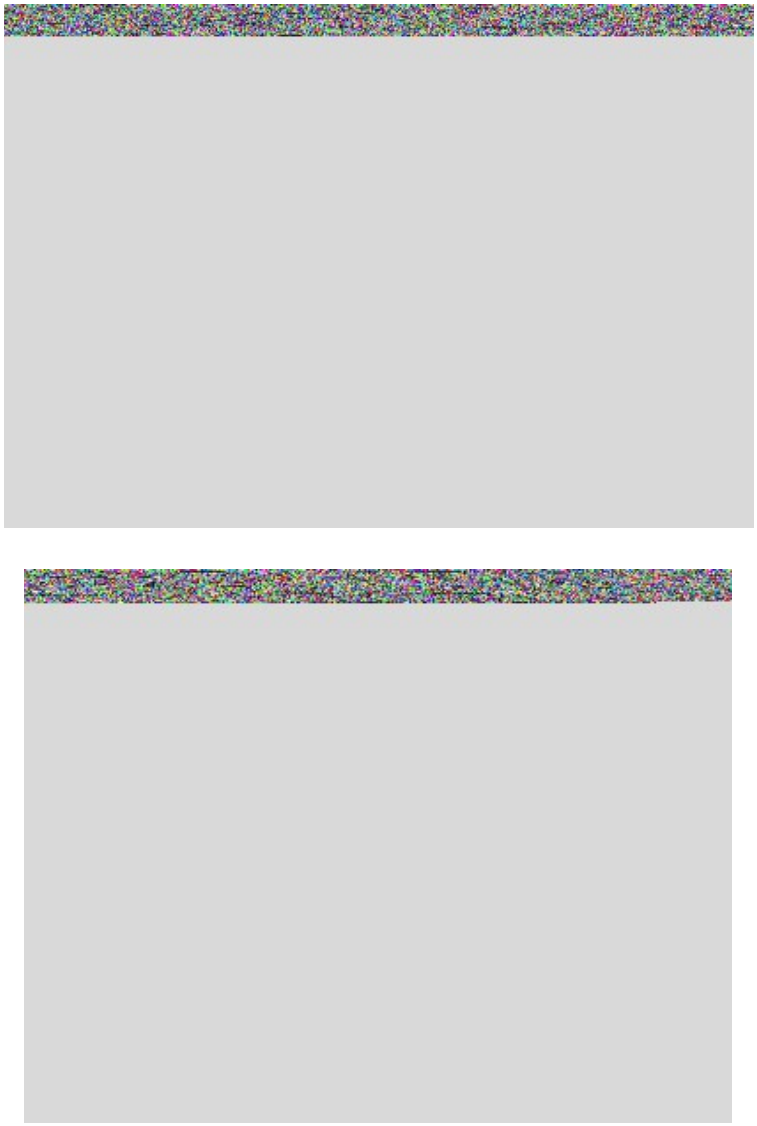


Fig. S5. Chirp corrected femtosecond transient absorption (TA) spectroscopy of pristine perovskite (a) and 0.2M urea incorporated (b) perovskite films at selected probe delay times. (c) The corresponding normalized bleaching kinetics at 770 nm for the pristine perovskite and 0.2M urea incorporated perovskite films following excitation at 650 nm (~2 μJ cm^{-2}).

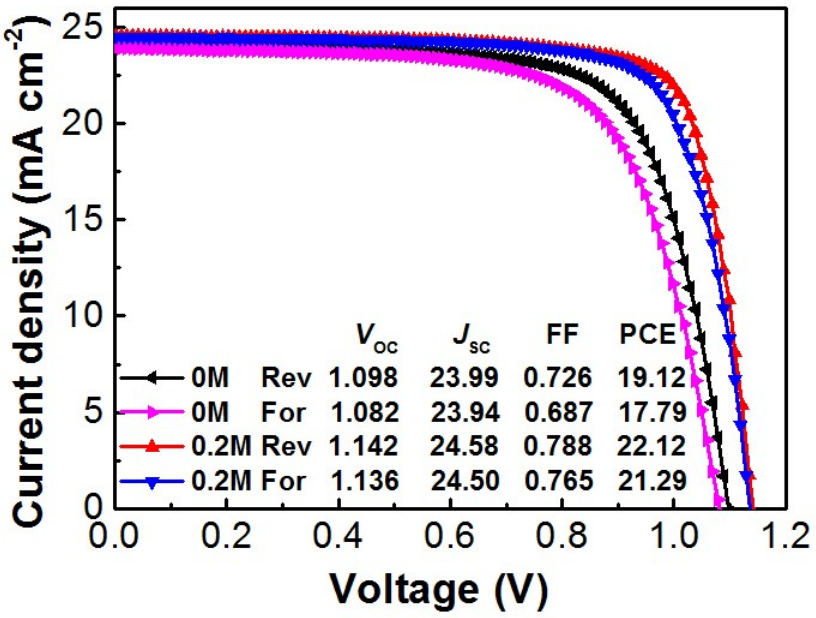


Fig. S6. *J-V* curves of PSC without and with 0.2 M urea doping.

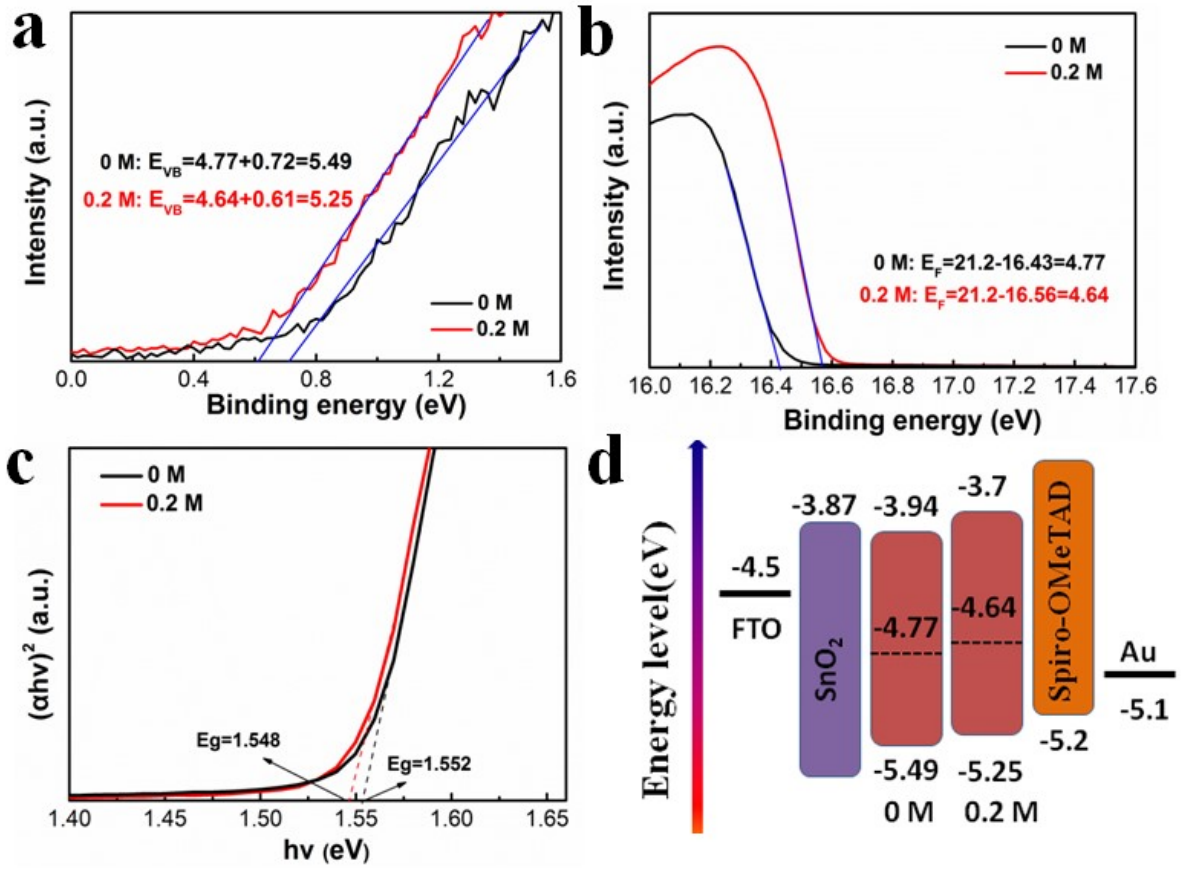


Fig. S7. (a) and (b) are UPS spectra of perovskite films without and with 0.2 M urea doping.(c) Tauc plot results of the without and with 0.2 M urea doping perovskites films.(d) Energy level diagram of a typical PSCs.