Supporting Information

Reduced open-circuit voltage deficit in wide-bandgap perovskite solar cells enabled by thiazolidine-based interfacial engineering

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Experimental Section

Materials

N,N-dimethylformamide (DMF), dimethyl sulfoxide (DMSO), chlorobenzene (CB), isopropanol (IPA), Cesium iodide (CsI), and $Pb(SCN)_2$ were purchased from Sigma-Aldrich. Diethyl ether (DE) was purchased from Chengdu Chron Chemical Corporation, Ltd.. Lead iodide (PbI_2), formamidinium iodide (FAI), and lead bromide ($PbBr_2$) were purchased from Advanced Election Technology Co., Ltd. Poly(bis(4-phenyl)(2,4,6-trimethylphenyl)amine) (PTAA) was purchased from Xi'an Polymer Light Technology Corporation. C_{60} was purchased from Nano-C, bathocuproine (BCP) was purchased from Jilin OLED and copper (Cu) was purchased from ZhongNuo Advanced Material (Beijing) Technology Co., Ltd. Tetrakis(dimethylamino) tin (IV) (99.9999%) was bought from Nanjing Ai Mou Yuan Scientific Equipment Co., Ltd. All the chemicals were used without further purification.

Perovskite Precursor Preparation

The 1.2 M $FA_{0.8}Cs_{0.2}Pb(I_{0.8}Br_{0.2})_3$ WBG perovskite precursor was prepared by dissolving 165.12 mg of FAI, 387.24 mg of PbI_2, 132.12 mg of $PbBr_2$, and 62.4 mg of CsI in 1 mL mixed solvent of DMF and DMSO with a volume ratio of 3 : 1. An additional 4.8 mg of $Pb(SCN)_2$ was introduced to promote grain growth. The precursor solution was stirred at 60 ℃ for 3 h before use.

The Fabrication of WBG Perovskite Solar Cells

The ITO substrates were ultrasonically cleaned with detergent, deionized water, and ethanol for 15 min, respectively. Before depositing PTAA (2 mg/mL in CB) hole transport layer (HTL), the ITO substrates were treated with ultraviolet-ozone for 15 min. The ITO substrates were then transferred to the nitrogen-filled glovebox. The PTAA solution was spin-coated on the ITO substrates at 4000 rpm for 30 s, and the substrates were annealed at 100 ℃ for 10 min. To improve the surface wettability of PTAA HTL, 50 μL DMF was spin-coated at 4000 rpm for 10 s without annealing. To obtain the perovskite film, 60 μL of the perovskite precursor solution was dropped on the pre-wetted substrate and then spin-coated at 500 rpm for 2 s and at 4000 rpm for 60 s with 600 μL diethyl ether dripping at the 25 s of the second step. Then the as-prepared films were annealed at 60 ℃ for 2 min and 100 ℃ for 10 min. For the devices with the ThHCl treatment, the ThHCl isopropanol solution at different concentrations of 0.5, 1,

2, and 3 mg/mL was spin-coated on the as-prepared perovskite substrates at 3000 rpm for 30 s, and then the samples were annealed at 100 ℃ for 5 min. To complete the device fabrication, all the substrates were transferred to the thermal evaporation chamber. 20 nm of C_{60}, 5 nm of BCP, and 100 nm of Copper were evaporated successively at 3×10^{-4} Pa. For the semitransparent cells, 20 nm of ALD SnO_2 was used instead of BCP. The precursors for ALD SnO_2 were tetrakis(dimethylamino) tin (IV) and deionized water. 120 nm ITO was sputtered under Ar pressure of 2 mTorr. The active area of devices is 0.0975 cm^2, as defined by the overlapped region between the back electrode and the patterned ITO substrate.

Film and Device Characterizations

WBG perovskites with and without ThHCl treatment were characterized by XRD, (Bruker D2 Phaser) with Cu-Kα (λ = 0.154 nm) radiation at 30 kV and 10 mA excitation. SEM images were taken with FE-SEM, Regulus-8230, Hitachi. Absorbance spectra of perovskite films were measured by ultraviolet-visible (UV-vis) spectrophotometer (PerkinElmer Lambda 950). PL and TRPL were performed by FLS980 (Edinburgh Inc.) PL and TRPL were measured using a supercontinuum pulsed laser (Wuhan Yangtze Soton Laser Co. Ltd.) with a wavelength of 532 nm. PL mapping was taken by alpha300 Raman Imaging Microscope (WITec). XPS was measured using a photoelectron spectrometer (ESCALAB 250Xi, Thermo Fisher Scientific). *J-V* curves were measured using Keysight B2901A sourcemeter under AM1.5G (100 mW cm^{-2}) illumination (Enlitech, SS-F5) in an N_2-filled glove box. All *J-V* measurements were performed via a mask with an aperture area of 0.0576 cm^2. The spectral response was measured by a quantum efficiency measurement system (QE-R, Enlitech). The electrochemical impedance spectra (EIS) were recorded on a IviumStat.h electrochemical workstation (Tianjin Deshang Technology Co. Ltd) with a frequency range of 1 MHz - 1 Hz at a bias of 1.0 V at dark. *C-V* measurements were carried out with a voltage scan range of 0 - 1.2 V and a frequency of 1 kHz at dark by using the IviumStat.h electrochemical workstation (Tianjin Deshang Technology Co. Ltd).

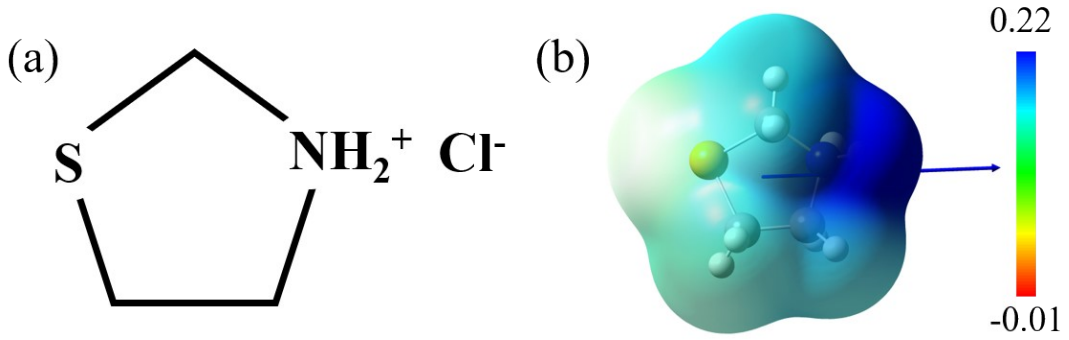

Fig. S1 (a) Structural formula of 1,3-thiazolidine hydrochloride (ThHCl). (b) Electrostatic potential distribution of ThH^+.

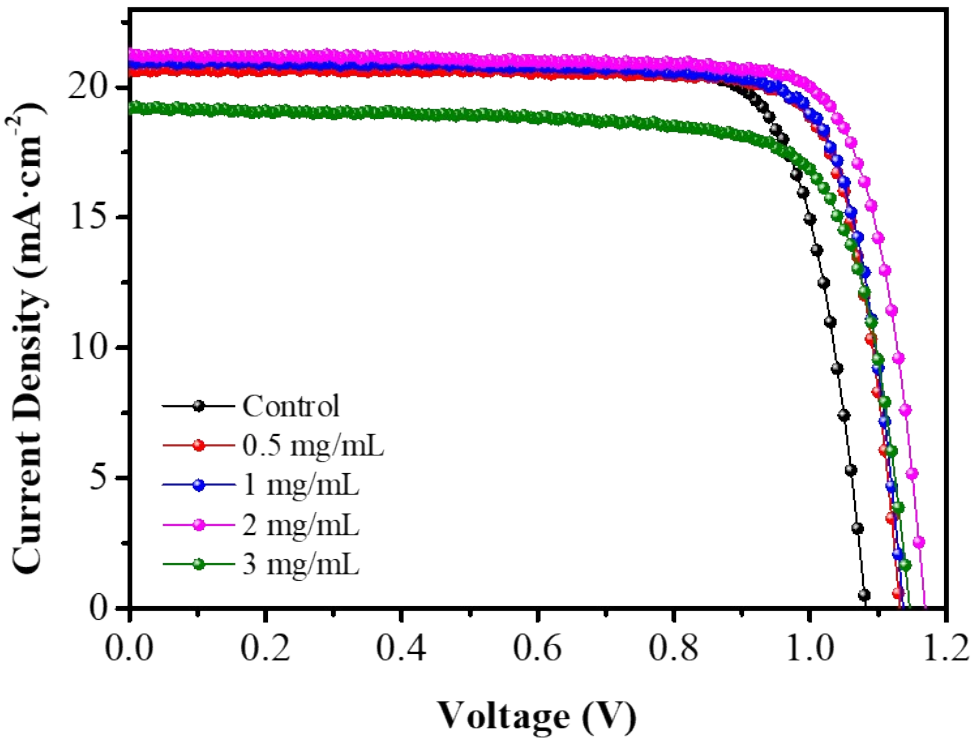

Fig. S2 *J-V* curves of WBG PSCs with different concentrations of ThHCl solution.

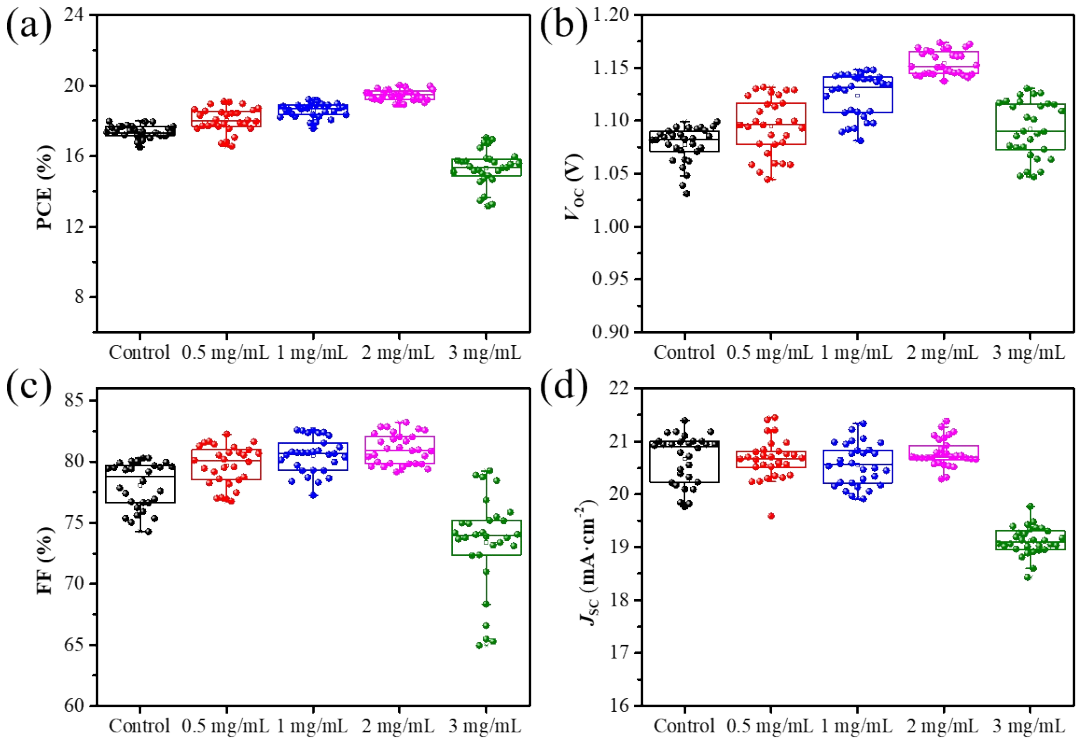

Fig. S3 Performance optimization of WBG $FA_{0.8}Cs_{0.2}Pb(I_{0.8}Br_{0.2})_3$ PSCs treated with different concentrations of ThHCl solution. (a) PCE, (b) V_{OC}, (c) FF, (d) J_{SC}.

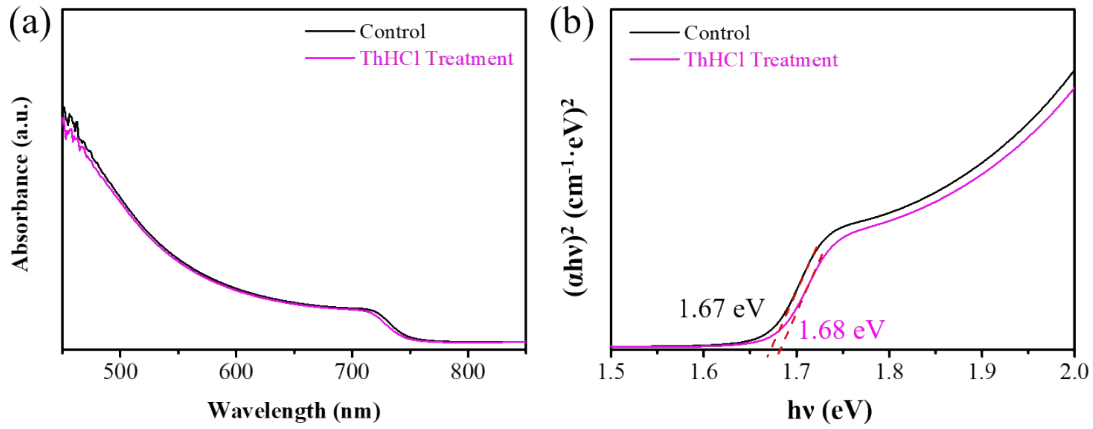

Fig. S4 (a) Absorption spectra and (b) Tauc plots of WBG perovskite films with and without ThHCl treatment.

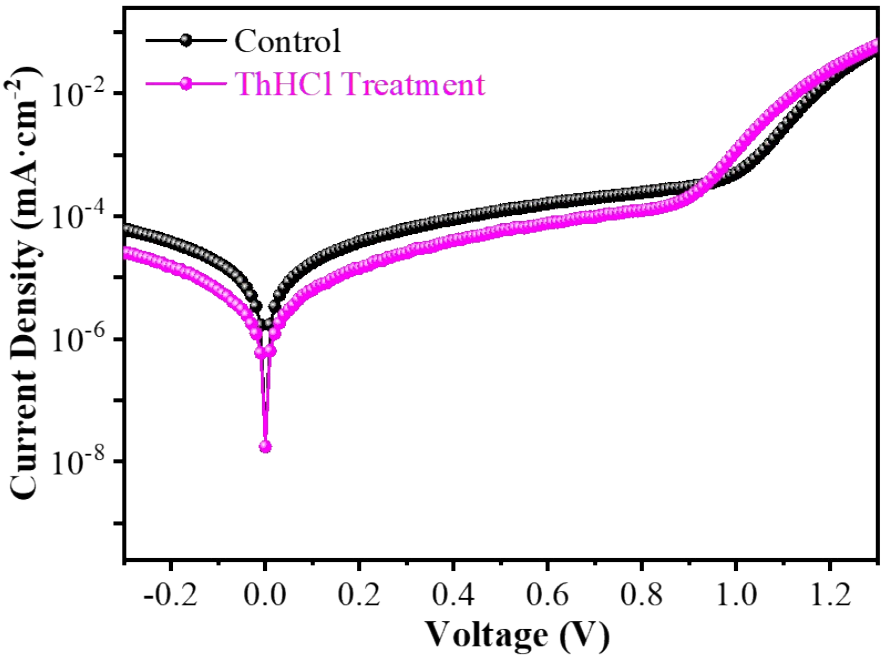

Fig. S5 Dark *J-V* curves of WBG PSCs with and without ThHCl treatment.

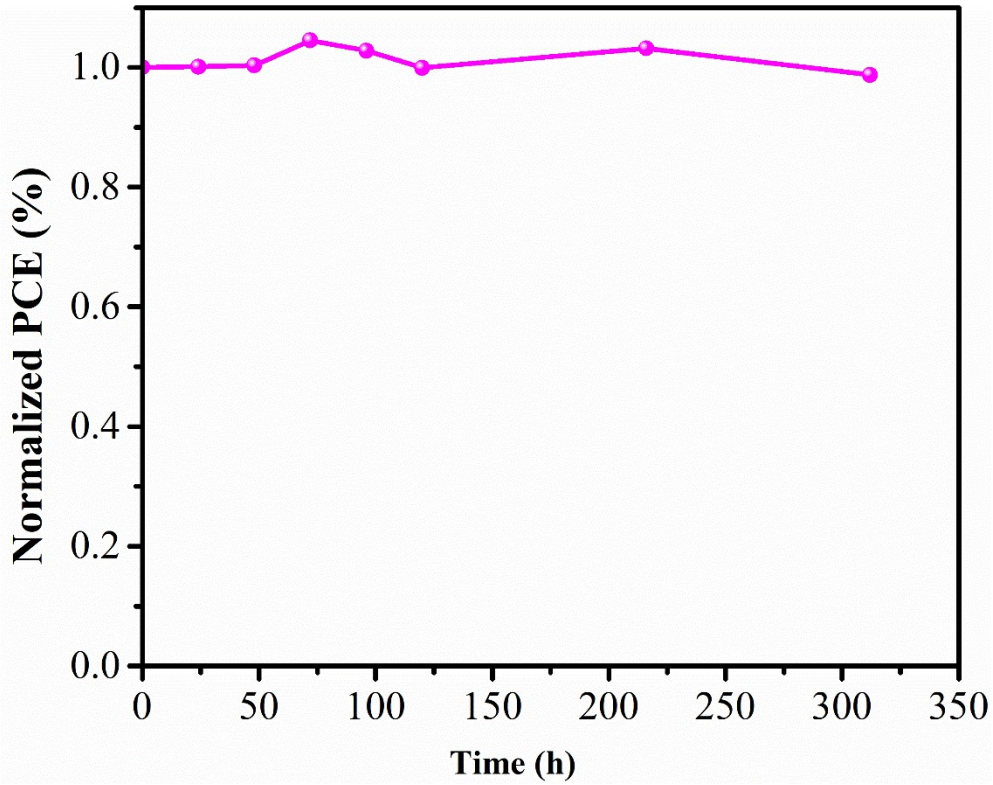

Fig. S6 Long-term stability of WBG PSC with ThHCl treatment stored in N_2-filled glovebox.

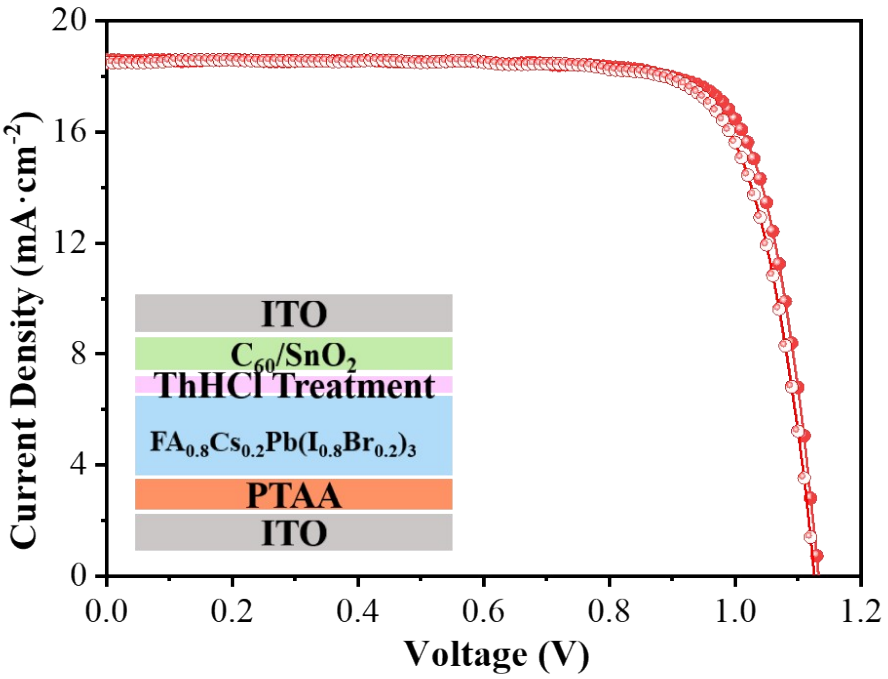

Fig. S7 *J-V* curves of semitransparent WBG PSC with ThHCl treatment. The inset is the corresponding device configuration.

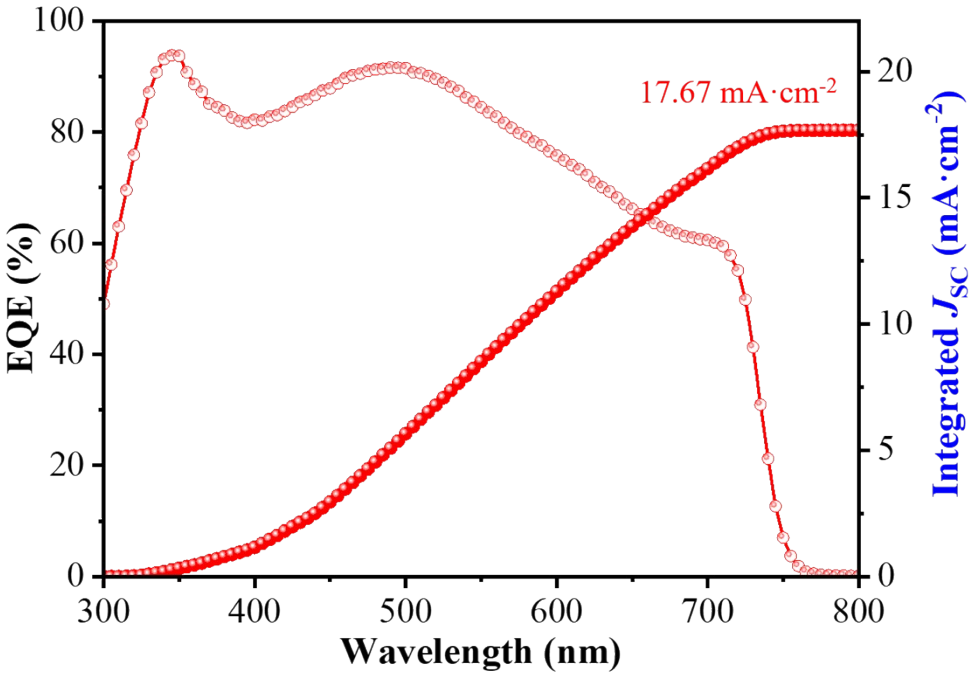

Fig. S8 EQE spectrum and EQE-integrated J_{SC} of semitransparent WBG PSC with ThHCl treatment.

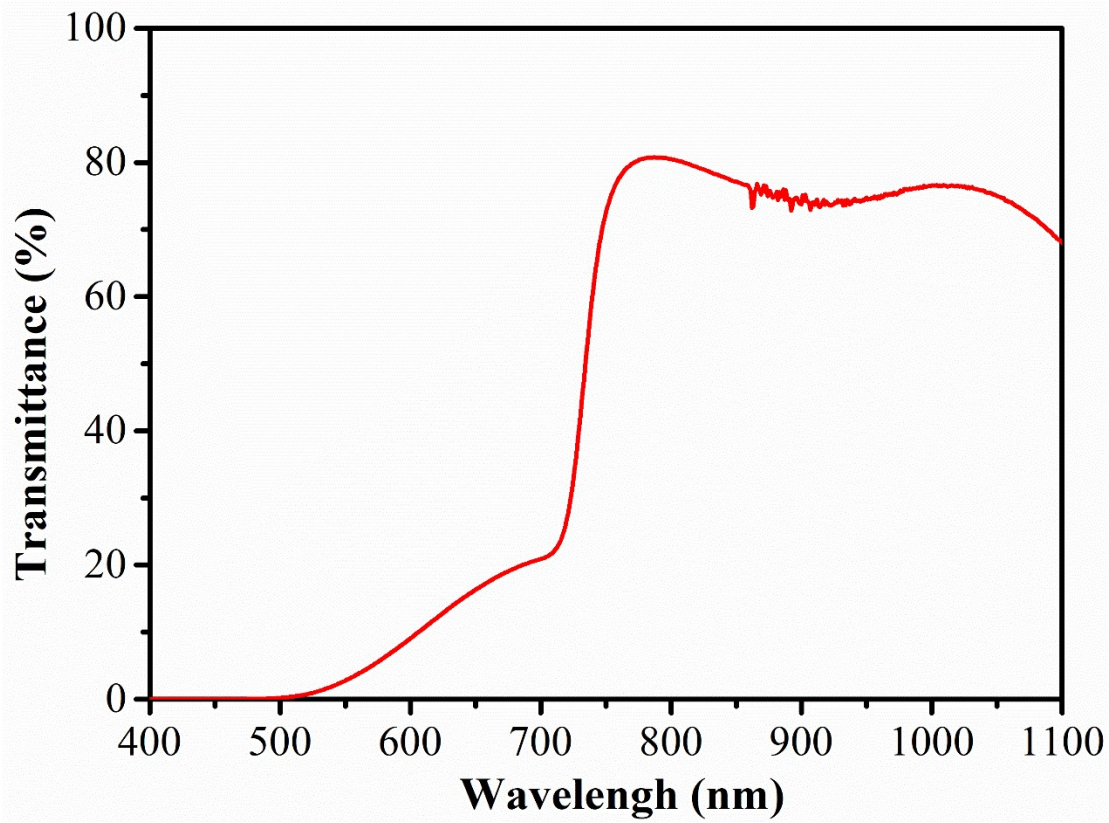

Fig. S9 Transmittance spectrum of semitransparent WBG PSC with ThHCl treatment.

Table S1 Photovoltaic parameters of $FA_{0.8}Cs_{0.2}Pb(I_{0.8}Br_{0.2})_3$ WBG PSCs treated with different concentrations of ThHCl solution.

Samples		PCE (%)	V_{OC} (V)	FF (%)	J_{SC} (mA·cm^{-2})
Control	Average	17.37 ± 0.35	1.07 ± 0.02	78.07 ± 1.89	20.66 ± 0.46
	Champion	17.98	1.08	79.95	20.79
0.5 mg/mL	Average	18.03 ± 0.68	1.09 ± 0.03	79.69 ± 1.61	20.67 ± 0.37
	Champion	19.09	1.13	81.63	20.67
1 mg/mL	Average	18.60 ± 0.41	1.12 ± 0.02	80.52 ± 1.44	20.56 ± 0.39
	Champion	19.21	1.14	80.55	20.97
2 mg/mL	Average	19.43 ± 0.32	1.15 ± 0.01	81.04 ± 1.23	20.77 ± 0.26
	Champion	20.02	1.17	80.48	21.27
3 mg/mL	Average	15.33 ± 1.01	1.09 ± 0.03	73.38 ± 3.87	19.12 ± 0.26
	Champion	16.97	1.14	77.23	19.17