Supporting Information

Facile synthesis strategy for cesium tin halide perovskite crystals toward light emitting devices and anti-counterfeiting flexible fiber

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1. Experimental sections

1.1 Chemical and materials: Cesium carbonate (Cs_2CO_3, 99.99%), tin chloride ($SnCl_2$, 99%), ethanol, and hydrochloric acid (HCl, AR) were purchased from Aladdin Chemical Co., Ltd. (Shanghai, China). Tellurium tetrachloride ($TeCl_4$, 99%) was purchased from Shanghai Macklin Biochemical Co., Ltd. (China). Polyphenylene sulfide resin (PPS, melting temperature = 285 ℃; Mw = 48,000) was supplied by Deyang Chemical Co., Ltd. (China). Aramid chopped fibers (ACFs) were supplied by China BlueStar Chengrand Research Institute of Chemical Industry. Polyethylene oxide (PEO, Mw = 2×10^6) was purchased from Sumitomo Keiretsu Co., Ltd., Japan. Anionic polyacrylamide (APAM, Mw = 3×10^6), acetone, and sodium dodecylbenzene sulfonate (SDS) were purchased from China Pharmaceutical Group Chemical Reagent Co., Ltd. Silicone defoamer was provided by Lusen Chemical Co., Ltd. (China). 385 nm near-ultraviolet (n-UV) chips (3 W, 600 mA) were provided by Suzhou Uking Photoelectric Technology Co., Ltd. (China). Silicone glue was purchased from Shenzhen Tongxin Lida Technology Co., Ltd. (China). DC-regulated power supply (0-15 V) was purchased from Dongguan Maisheng Power Technology Co., Ltd. (China). All chemicals were used directly without further purification.

1.2 Synthesis of $Cs_2Sn_{1-x}Te_xCl_6$ perovskite crystals (PCs): $Cs_2Sn_{1-x}Te_xCl_6$ PCs were synthesized by a more convenient hydrothermal method. *x* mmol of $TeCl_4$ and 0.50 - *x* mmol of $SnCl_2$ were placed within a 100 mL three-neck flask. 3 mL of HCl and 16 mL of ethanol were injected into the three-neck flask. The solution was heated to 80 ℃ with stirring for 30 min until the solution became clear. Subsequently, 0.5 mmol of Cs_2CO_3 (dissolved in 1 mL of HCl) was swiftly injected into the solution, and the $Cs_2Sn_{1-x}Te_xCl_6$ PCs were immediately precipitated. The solution was kept at 80 ℃ with stirring for 60 min for the complete reaction. The solution was purified by centrifugation at 6000 rpm for 5 min. The precipitate was collected, and the supernatant was discarded. Finally, the $Cs_2Sn_{1-x}Te_xCl_6$ PCs were washed with ethanol and dried at 60 ℃ for 6 h. Solid crystals were collected for the characterization of

optical properties and other properties. Meanwhile, we kept some of the original $Cs_2Sn_{1-x}Te_xCl_6$ PCs solution to post-process the ACFs/PPS compound fiber paper.

1.3 Preparation of ACFs/PPS compound fiber paper: First, the ACFs and PPS microfiber non-woven fabrics were cleaned with acetone to remove impurities. PPS ultrafine fiber pulp was made of PPS non-woven fabric with a valley-type beating machine. Then, APAM, SDS and PEO (with a mass ratio of 1:3:6) are stirred at room temperature in deionized water for 4h to obtain a uniform dispersion solution. Next, PPS ultrafine fiber pulp and ACFs (with a mass ratio of 3:7) were added to the dispersion and dispersed by a fiber dissociator. Finally, the mixed fiber suspension was directly used for wet papermaking on the paper machine to obtain ACFs/PPS composite fiber paper.

1.4 Preparation of ACFs/PPS compound fiber paper modified with $Cs_2Sn_{1-x}Te_xCl_6$ PCs: The prepared ACFs/PPS compound fiber paper was cut into regular shapes. Subsequently, the paper was post-processed with $Cs_2Sn_{1-x}Te_xCl_6$ PCs solution for 4 h. Then, the modified paper was dried in a drying oven at 60 ℃ for 6 h. The ACFs/PPS compound fiber paper with fluorescent anti-counterfeiting was prepared.

1.5 Fabrication and characterizations of LED: Normally, 20 mg of $Cs_2Sn_{0.94}Te_{0.06}Cl_6$ PCs and 200 mg of AB silicone glue were uniformly mixed together. Then the mixture was coated on the 385 nm n-UV chip to obtain the LED. The device was cured at 60 ℃ for 4 h. Electroluminescence spectrum was measured by debugging the fluorescence spectrophotometer (HITACHI, F-4700). Specific operations can be summarized as follows:

The emission port of the fluorescence spectrophotometer was closed. The voltage of the LED to 5 V and the current to 600 mA were set with the external DC-regulated power supply, and then the LED was faced with the receiving port of the fluorescence spectrophotometer to measure the EL spectrum of LED.

1.6 Characterizations: X-ray diffraction (XRD) patterns were measured by an X-ray Powder diffractometer (PANalytical B.V., Empyrean) to determine the crystal structure of samples. The elemental composition and chemical state of samples were identified by X-ray photoelectron spectroscopy (XPS, Thermo Scientific, K-

Alpha). The morphology and lattice spacing of samples were observed by scanning electron microscope (SEM). The element distribution was analyzed by energy-dispersive spectrometry (EDS, attached on the SEM, TESCAN MIRA LMS/TESCAN MIRA4 (Czech Republic)). The photoluminescence (PL), photoluminescence excitation (PLE) spectra, electroluminescence (EL), and temperature-dependent PL spectra were measured with the fluorescence spectrophotometer (HITACHI, F-4700). The photoluminescence quantum yield (PLQY) and PL decay were measured with fluorescence spectrometers (Edinburgh, FLS1000). Thermo gravimetric analysis (TGA) was measured with a thermal gravimetric analyzer (NETZSCH, TG209F1).

Tab. S1 Chemical composition (at %) of $Cs_2Sn_{1-x}Te_xCl_6$ perovskite crystals measured by EDS.

x	Cs	Sn	Te	Cl
0	24.72	11.72	0	63.56
0.01	24.77	11.74	0.3	63.19
0.02	23.55	11.63	0.32	64.5
0.03	24.1	11.7	0.49	63.71
0.04	24.58	11.7	0.68	63.04
0.05	24.32	11.13	0.59	63.96

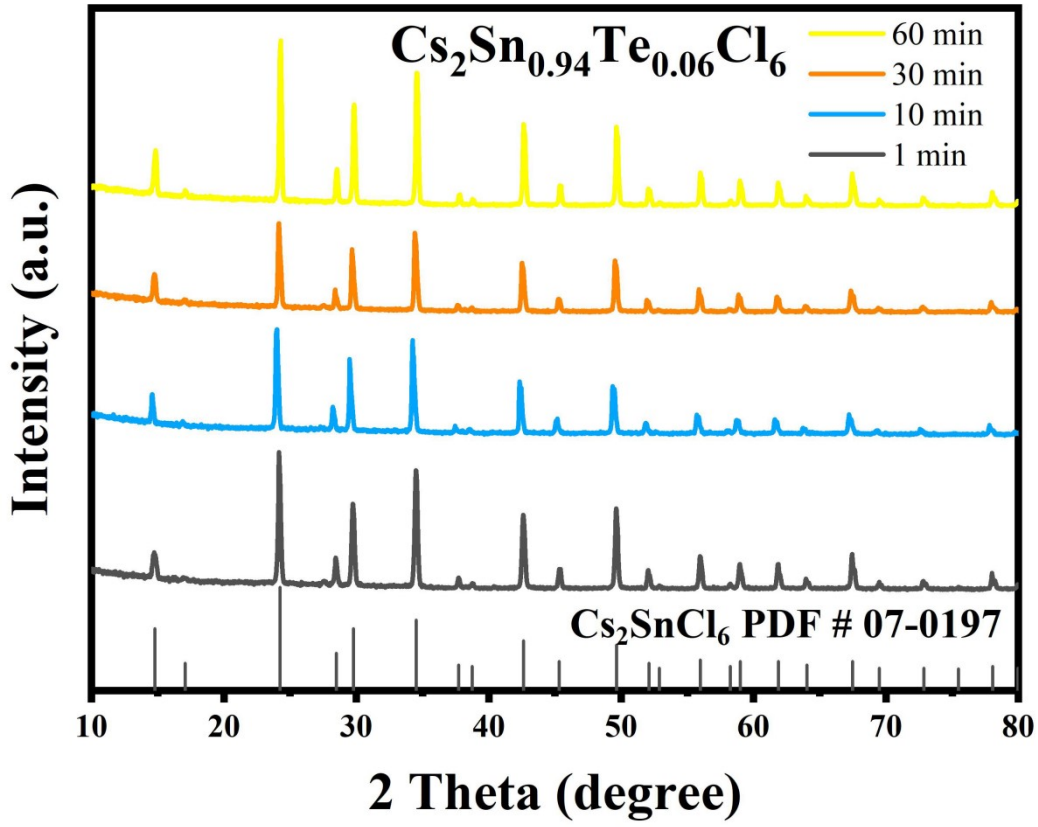

Fig. S1 The XRD patterns of $Cs_2Sn_{0.94}Te_{0.06}Cl_6$ with different reaction time.

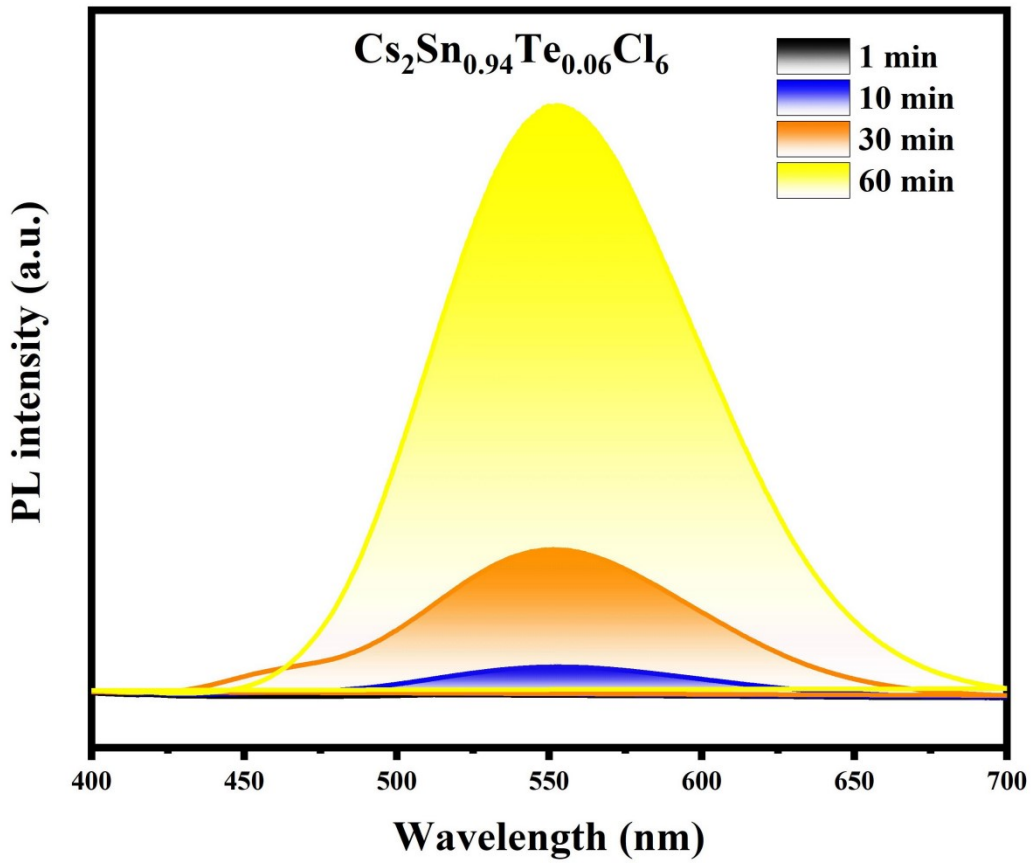

Fig. S2 The PL spectra of $Cs_2Sn_{0.94}Te_{0.06}Cl_6$ with different reaction time.

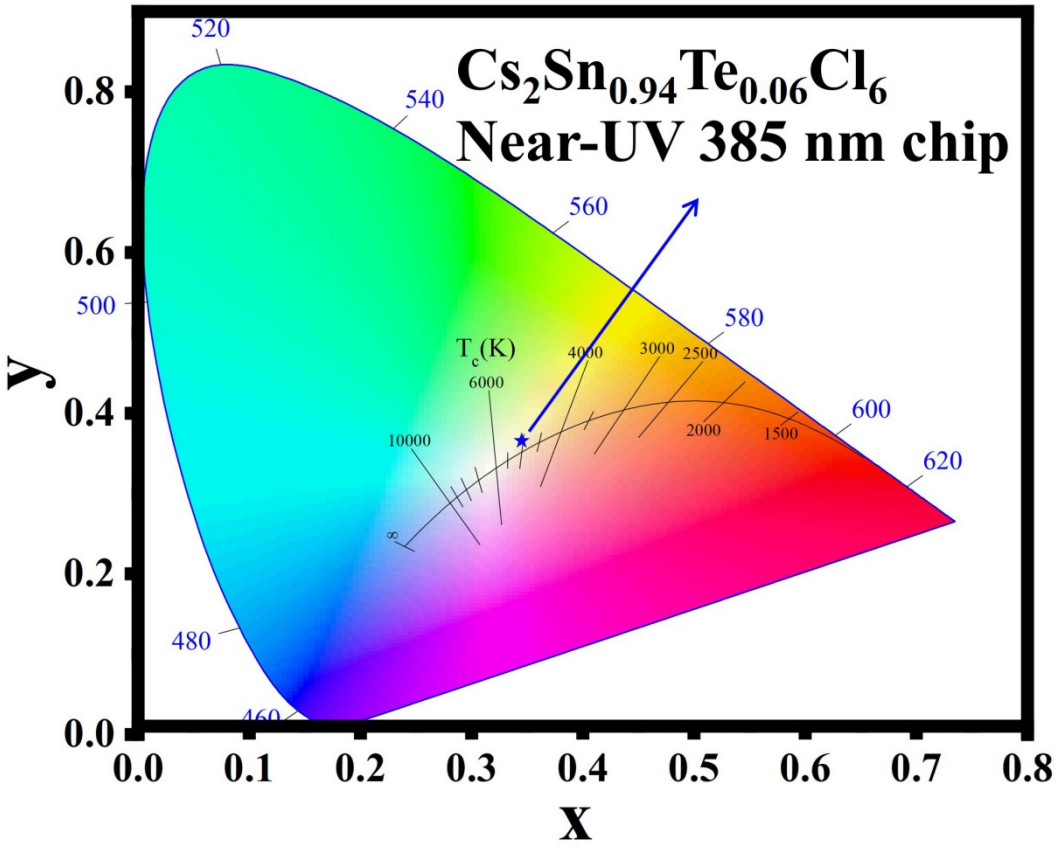

Fig. S3 Chromaticity coordinates (CIE 1931) of light-emitting diode (LED) lamp fabricated via the combination of a near-ultraviolet 385 nm chip and $Cs_2Sn_{0.94}Te_{0.06}Cl_6$ perovskite crystals.

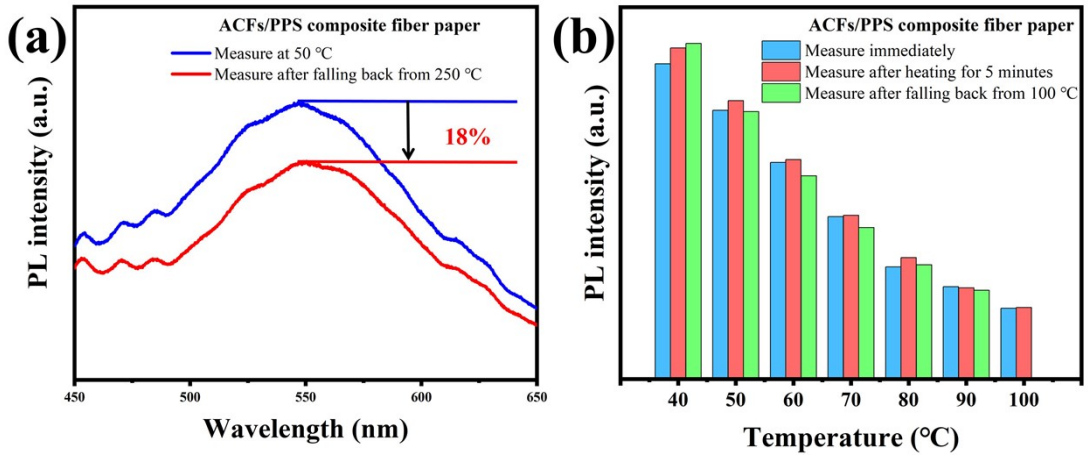

Fig. S4 (a) PL spectra of ACFs/PPS composite fiber paper were measured at 50 ℃ and after falling back from 250 ℃. (b) PL intensities of ACFs/PPS composite fiber paper were measured at different temperatures.