Supporting Information

Ultrastable Germanium Oxide Tuned Induced Self-Crystallization of $CsPbBr_3$@Glass for White Light-Emitting Devices and Wide Color Gamut Backlight Display

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1. Experimental Section

1.1 Materials

Silicon dioxide (SiO_2, Sinopharm, 99%), germanium dioxide (GeO_2, Meryer, 99.999%), aluminum oxide (Al_2O_3, Aladdin, 99.9%), lithium carbonate (Li_2CO_3, Aladdin, 99.99%), potassium carbonate (K_2CO_3, Meryer, 99%), cesium carbonate (Cs_2CO_3, Meryer, 99.9%), lead bromide ($PbBr_2$, Meryer, 99%), sodium bromide (NaBr, Aladdin, 99%), ethylene-vinyl acetate copolymer (EVA, Ruixiang Plastic Co., LTD), K_2SiF_6: Mn^{4+} phosphor (K_2SiF_6: Mn^{4+}, Hangzhou Yingke new material Co., LTD)

1.2 Properties of $CsPbBr_3$@Glass

Firstly, the glass raw materials and $CsPbBr_3$ perovskite precursor were meticulously mixed in a grinding bowl, then transferred into a crucible, and melted at 1300 °C for 20 minutes to attain uniform molten glass, the molten glass was rapidly quenched in water. Secondly, the water-quenched glass was dried at 80 °C and subsequently ground. Finally, the ground glass samples underwent a heat treatment process at temperatures ranging from 490 °C to 550 °C for 5 h (10 h) or 450 °C to 490 °C for 10 h.

1.3 Synthesis of $CsPbBr_3$@Glass@EVA film and K_2SiF_6: Mn^{4+}@EVA film

Taking green film as an example. Firstly, mixed the $CsPbBr_3$@Glass powder and EVA powder in different proportions. Secondly, placed mixture on a plate vulcanizer to heat up and press the film. Finally, taken out after cooling. The proportion of green film were 0%, 20%, 40%, 60%, 80% (the proportion of red film was 10%). The heating temperature was 140 °C, the heating time was 20 minutes, and the cooling time was 15 minutes.

1.4 Fabrication of WLED

The prepared green emitting $CsPbBr_3$@Glass and commercial phosphor of K_2SiF_6: Mn^{4+} were mixed with evenly according to the proportion. And then homogeneously dispersed in the mixture of silica gel (gel A: gel B = 3:1), and then dropped on the InGaN blue chip.

1.5 Fabrication of $CsPbBr_3$@Glass@EVA film and K_2SiF_6: Mn^{4+}@EVA film display device

The light-emitting layer composed of the mini-blue-chip and the light guide plate was put on the top of the reflective layer, and then the $CsPbBr_3$@Glass@EVA composite film and K_2SiF_6: Mn^{4+}@EVA film was put on the light guide plate as the light conversion layer. Fabrication a wide color gamut display device by using a commercial TFT LCD (Thin Film Transistor Liquid Crystal Display) backlight.

1.6 Characterization

X-Ray diffraction (XRD) was measured using Bruker D8 Advance measurements, wherein Cu Kα radiation was working environment at voltage 40 kV and current 40 mA in the boundary of 10–60° (2θ) at the velocity of 0.03° s^{-1}. Transmission electron microscopy (TEM), High Resolution Transmission Electron Microscopy (HRTEM) images and energy-dispersive X-ray spectroscopy (EDX) were measured using a FEI Tecnai G2F20S-TWIN transmission electron microscope. The ultraviolet-visible (UV-Vis) spectrum was measured by PerkinElmer 750 (UV-Vis) spectrometer in the boundary of 200–800 nm (λex = 365 nm). The PL spectra was measured by a Horiba Jobin Yvon Fluromax-4P Spectrofluorometer (λex = 365 nm). The Fourier-transform infrared spectroscopy (FTIR) spectrum was measured by Nicolet iS50 FT-IR between the wave number range of 400 cm^{-1} and 4000 cm^{-1}. The temperature-dependent fluorescence spectrum was measured using a THMS 600 fluorescence spectrophotometer with temperature control. Using time-correlated single-photon counting (TCSPC) lifetime spectroscopy system and nanosecond-pulsed diode laser (nano LED-370) as a single wavelength excitation light source, the lifetime of PL was measured. X-ray photoelectron spectroscopy (XPS) was collected using an Axi Ultra DLD spectrometer with single-color Al Ka radiation as the excitation source. Differential Scanning Calorimeter (DSC, Germany NETZSCH, DSC214) was used for thermal characterization of $CsPbBr_3$@Glass at a heating rate of 10°C /min from room temperature to 700 °C in Ar atmosphere.

The three-exponential function can be expressed as:

$$A(t) = A_1exp(-t/\tau_1) + A_2exp(-t/\tau_2) + A_3exp(-t/\tau_3)$$

2. Supporting Figures and Tables

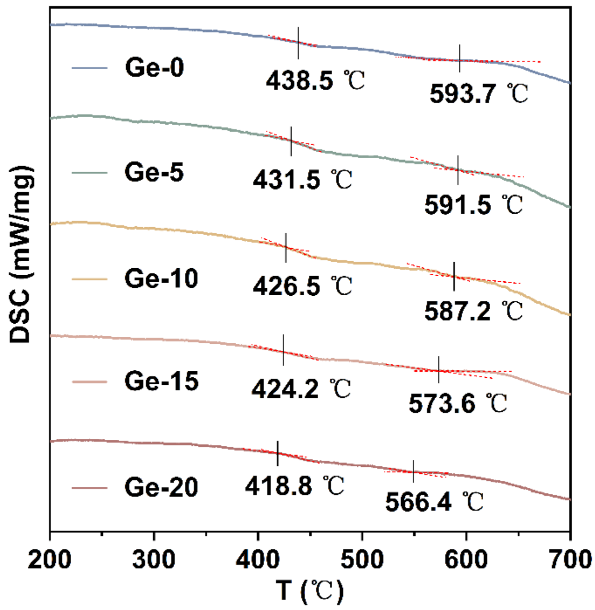

Figure S1 DSC curves of five kinds of $CsPbBr_3$@Glass with different GeO_2 contents without heat treatment.

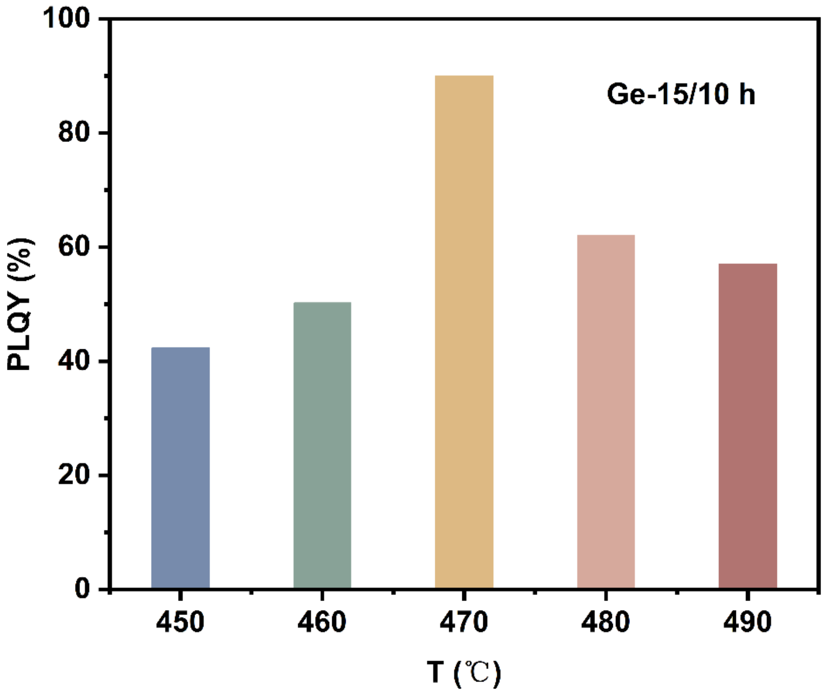

Figure S2 PLQY Intensity of $CsPbBr_3$@Glass samples with different heat treatment temperature.

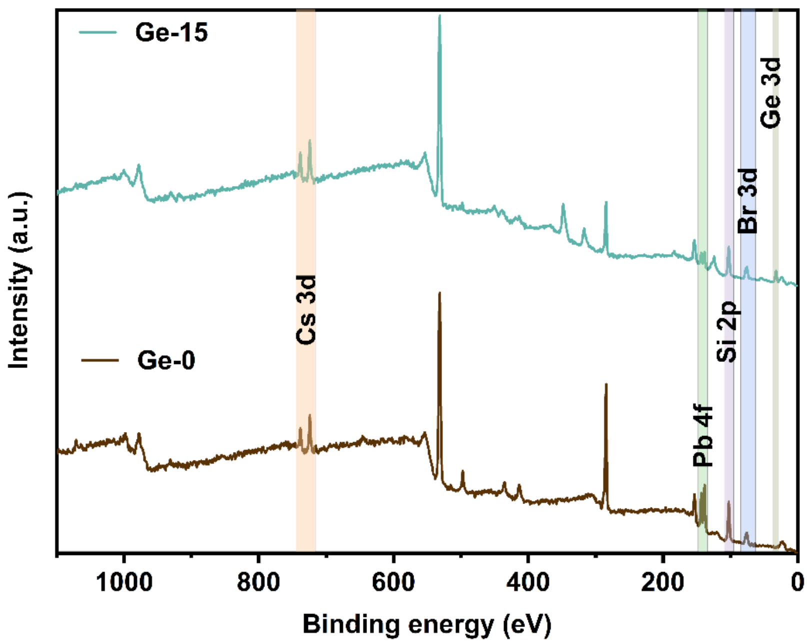

Figure S3 XPS survey spectra of Si 2p, Ge 3d, Cs 3d, Pb 4f and Br 3d in Ge-0, Ge-15 samples.

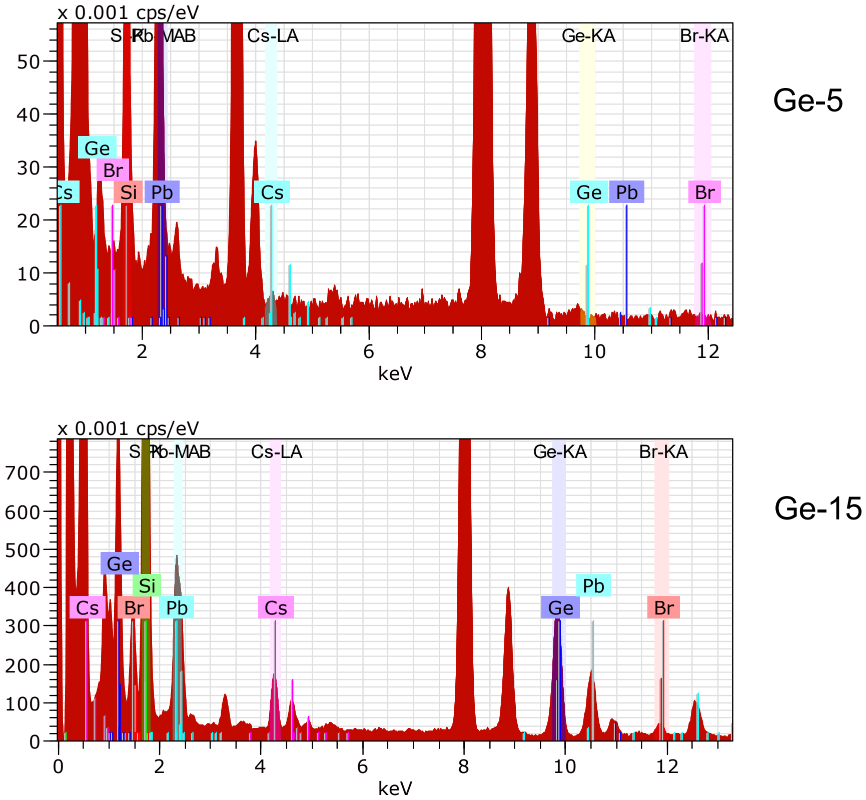

Figure S4 EDX spectrum of the as-prepared Ge-5 and Ge-15 glass sample, showing the existence of Cs, Pb, Br, Si and Ge elements.

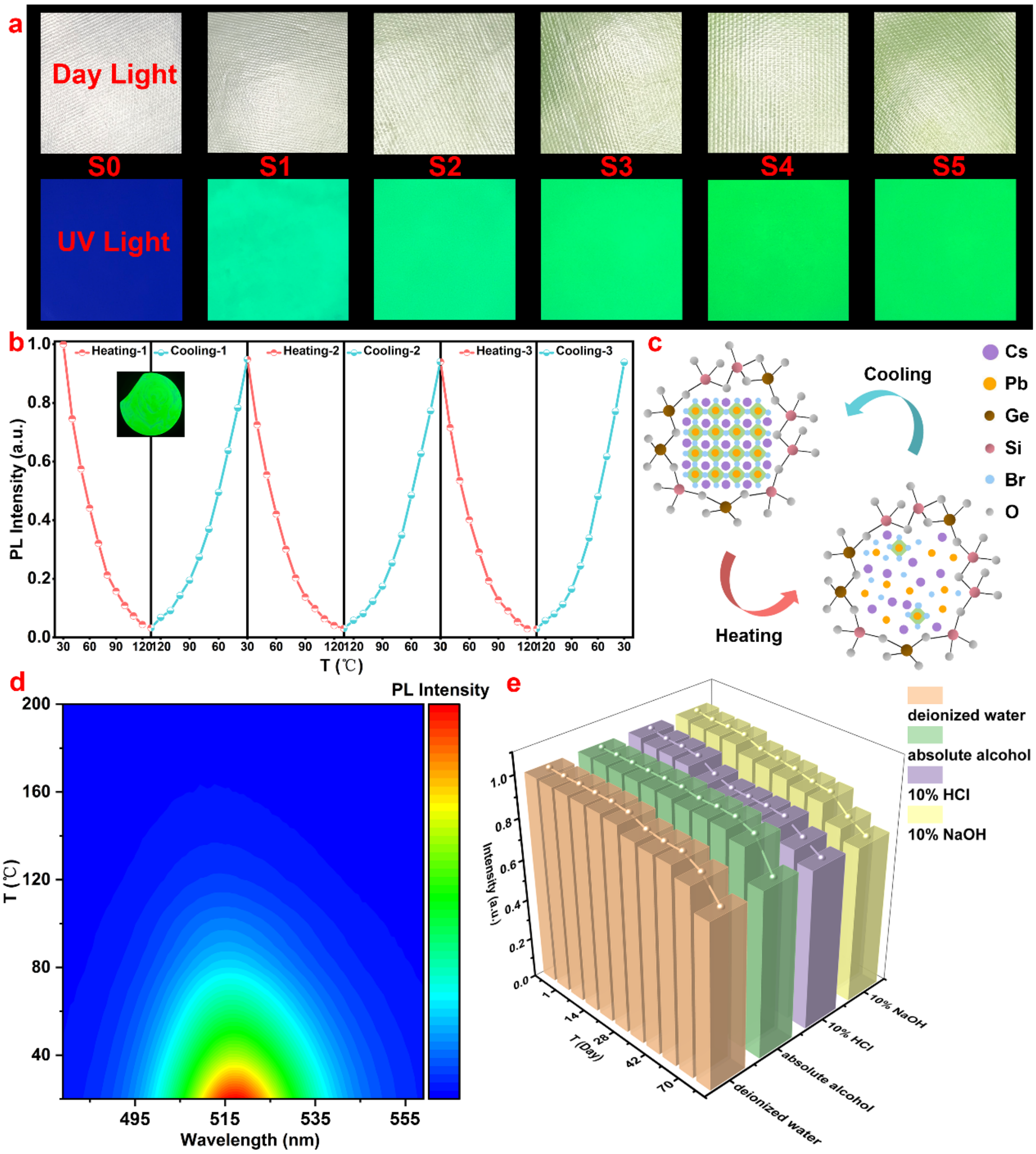

Figure S5 The photographs with different proportions of the $CsPbBr_3$@Glass@EVA film samples under ultraviolet (365 nm) irradiation. (b) Relative PL intensity for the $CsPbBr_3$@Glass sample experiencing three-time heating/cooling cycles between 30 °C and 120 °C, inset was the photograph of $CsPbBr_3$@Glass sample under UV light. (c) The mechanism diagram of $CsPbBr_3$@Glass heating/cooling cycle treatment. (d) The corresponding temperature-dependent PL emission spectra of the $CsPbBr_3$@Glass between 20 °C and 200 °C. (e) The relative luminescence intensity of $CsPbBr_3$@Glass@EVA film in four different solutions for 80 days.

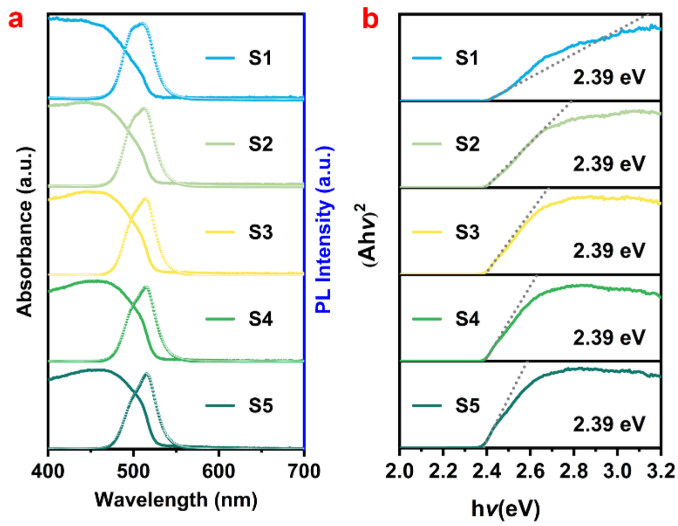

Figure S6 (a) The fluorescence spectra and UV-Vis absorption spectra and (b) bandgap spectra of the $CsPbBr_3$@Glass@EVA film samples.

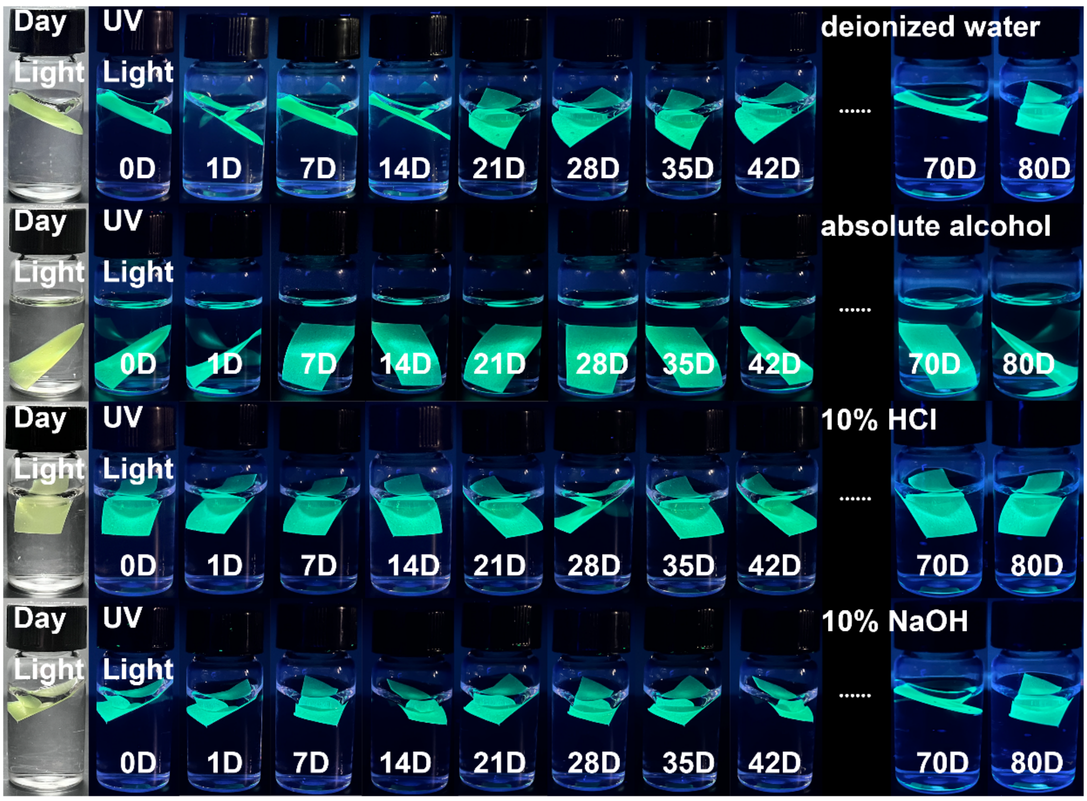

Figure S7 The day light and UV light photographs of $CsPbBr_3$@Glass@EVA film in four different solutions for 80 days.

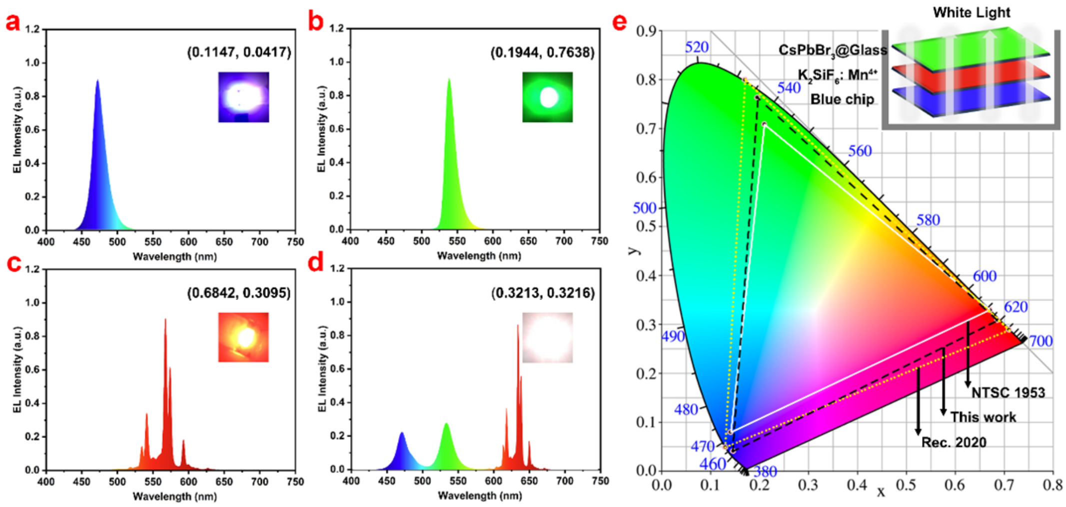

Figure S8 The PL spectra of blue chips, green $CsPbBr_3$@Glass composites, red K_2SiF_6: Mn^{4+} phosphor and WLED, insets included blue, green, red and white light, respectively. (e) Color gamut of NSTC 1953 standard (white line), the Rec. 2020 standard (yellow line) and this work (black line) in the CIE diagram, inset was the structural schematic of WLED.

Table S1. PLQY, stability, and color gamut data of PNCs@Glass.

Sample	PLQY/%	Stability	Color Gamut	Ref.
$CsPbBr_3$@Glass	70	in water 120 h, PL remained 90%	/	17
$CsPbBr_3$@Glass	81.1	in water 7 days, PL remained 97%	/	21
$CsPbBr_3$@Glass	72.9	/	/	25
$CsPbBr_3$@Glass	63	in water 30 days, PL remained 90%	NTSC 123%	26
$CsPbBr_3$@Glass	14.8	in water 60 days, PL remained ~100%	/	27
$CsPbBr_3$@Glass	48	in water 42 days, PL remained ~55%	NTSC 103.1%	28
$CsPbBr_3$@Glass	40	in water 40 days, PL	/	29

		remained 60%		
$CsPbBr_3$@Glass	90	in water 80 days, PL remained 75%	123%	this work

Table S2. PL lifetimes data of $CsPbBr_3$@Glass.

Samples $CsPbBr_3$@Glass	τ_1 (ns)	τ_2 (ns)	τ_3 (ns)	Rel%-1	Rel%-2	Rel%-3	$\tau_{average}$ (ns)
450°C	4.27	21.39	108.17	19.48	47.64	32.87	46.58
460°C	4.92	28.79	158.04	18.40	43.23	38.36	73.97
470°C	5.84	36.01	203.09	16.49	40.06	41.30	99.27
480°C	5.85	37.95	228.37	16.26	39.29	44.45	117.37
490°C	6.06	38.33	239.56	14.73	33.39	51.88	137.97

Table S3. CIE color coordinates and the calculated color gamut for Rec.2020 standard, NTSC 1953 standard and this work.

	Red	**Green**	**Blue**	**Area**
NTSC 1953	(0.62, 0.33)	(0.21,0.71)	(0.14, 0.08)	0.158 (100%)
Rec. 2020	(0.71,0.29)	(0.17,0.80)	(0.13,0.05)	0.212 (134%)
This work	(0.68,0.31)	(0.19,0.76)	(0.11,0.04)	0.195 (123%)

The area ratio was defined as the color gamut, which compares the RGB triangular area of the LCD screen with the triangular area of the NTSC 1953 standard. The definition was described as

$$\text{Color Gamut Area} = \frac{A_{screen}}{A_{standard}}$$

A_{screen} and $A_{standard}$ are the RGB triangular area of the LCD screen and that of the NTSC1953 standard, respectively, which are calculated by the equation as follow:

$$A_{screen} \text{ or } A_{standard} = \frac{1}{2}\begin{vmatrix} x_R & y_R & 1 \\ x_G & y_G & 1 \\ x_B & y_B & 1 \end{vmatrix}$$