Supporting Information for:

Aggregation-Induced Emission Assembled Ultrathin Films for White Light-Emitting Diodes

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Experimental Section

Materials. All reagents in experiments were of analytical grade and used without further purification. NaOH, H_2SO_4, HCl, H_2O_2, etanol, acetone, tetrahydrofuran (THF) were purchased from Beijing Chemical Reagent Company (Beijing, China). Gold(III) chloride trihydrate ($HAuCl_4 \cdot 3H_2O$) was purchased from J&K Chemical Co. Ltd. (Beijing, China). Glutathione reduced form (GSH) was purchased from Solarbio (Beijing, China). 1, 4-Butane sultone, 4-hydroxybenzophenone and polyethylene (PE) powder were purchased from Alfa Aesar (Ward Hill, MA, USA). Titanium tetrachloride and sodium ethoxide were purchased from Aladdin Chemical. Co. Ltd. (Shanghai, China). Poly(allylamine hydrochloride) (PAH) and Zn dust were purchased from HWRK Chem Co. Ltd. (Beijing, China). Ultra-pure water was obtained from the Milli-Q purification system (Barnstead, CA, USA).

Preparation of 1,2-bis[4-(3-sulfonatopropoxyl)-phenyl]-1,2-diphenylethene sodium salt (BSTPE).[1] A mixture of Zn dust (2.9 g, 44 mmol) and 4-hydroxybenzophenone (2.0 g, 10 mmol) was dissolved in 100 mL anhydrous THF in a 250 mL flask and protected by nitrogen. The suspension was then cooled to 0 ^oC, and $TiCl_4$ (2.5 mL, 10 mmol) was subsequently injected into the flask. The resultant mixture was warmed to room temperature for 4 h, followed by filtration with diethyl ether and drying with sodium sulfate. The obtained filtrates were evaporated and purified by a silica gel column, and the eluent was petroleum ether/ethyl acetate (1: 1 by volume, R_f = 0.50). The yield of the obtained 1,2-bis(4-hydroxyphenyl)-1,2-diphenylethene was about 88% (1.61 g). 1H NMR (600 MHz): δ (ppm) 9.29-9.31 (d, 2H), 7.04-7.17(m, 6H), 6.92-6.97(m, 4H), 6.70-6.75(m, 4H), 6.47-6.53(m, 4H). The as-prepared 1,2-bis(4-hydroxyphenyl)-1,2-diphenylethene (0.44 g, 1.2 mmol) was dispersed in 20 mL anhydrous ethanol under the protection of nitrogen. Afterwards, NaOEt (0.23 g, 3.4 mmol) was dissolved in 20 mL anhydrous ethanol and added into 1,2-bis(4-hydroxyphenyl)-1,2-diphenylethene dropwise. The mixed suspension was stirred for 1 h. As the colorless suspension turned to orange-red, the solution of 1,4-butane sultone (0.31 g, 2.3 mmol) in 20 mL absolute ethanol was added into the reaction mixture. The reaction mixture was stirred intensely for 12 h until the white solid was precipitated out. The BSTPE was collected by filtration and washed with acetone and ethanol twice, and 60% yield (0.49 g) for BSTPE was obtained. 1H NMR (600 MHz): δ (ppm) 7.30-7.39(m, 2H), 6.84-7.10(m, 12H), 6.64-6.71(m, 4H), 3.87-3.99(m, 4H), 2.53-2.57(t, 4H), 1.91-1.98(m, 4H).

Preparation of gold nanoclusters (Au NCs). The synthesis of the Au NCs is carried out according to the previous work with a few modifications.[2] Firstly, the mixed aqueous solution of $HAuCl_4\cdot3H_2O$ (20 mM, 0.5 mL) and GSH (100 mM, 0.15mL) was added into 4.35 mL ultrapure water at 25 ^{o}C. Then, the solution was heated to 70 ^{o}C and stirred with a low speed for 24 h. Finally, the yellow-colored Au NCs solution with orange fluorescence emission was obtained and stored at 4 ^{o}C for further use.

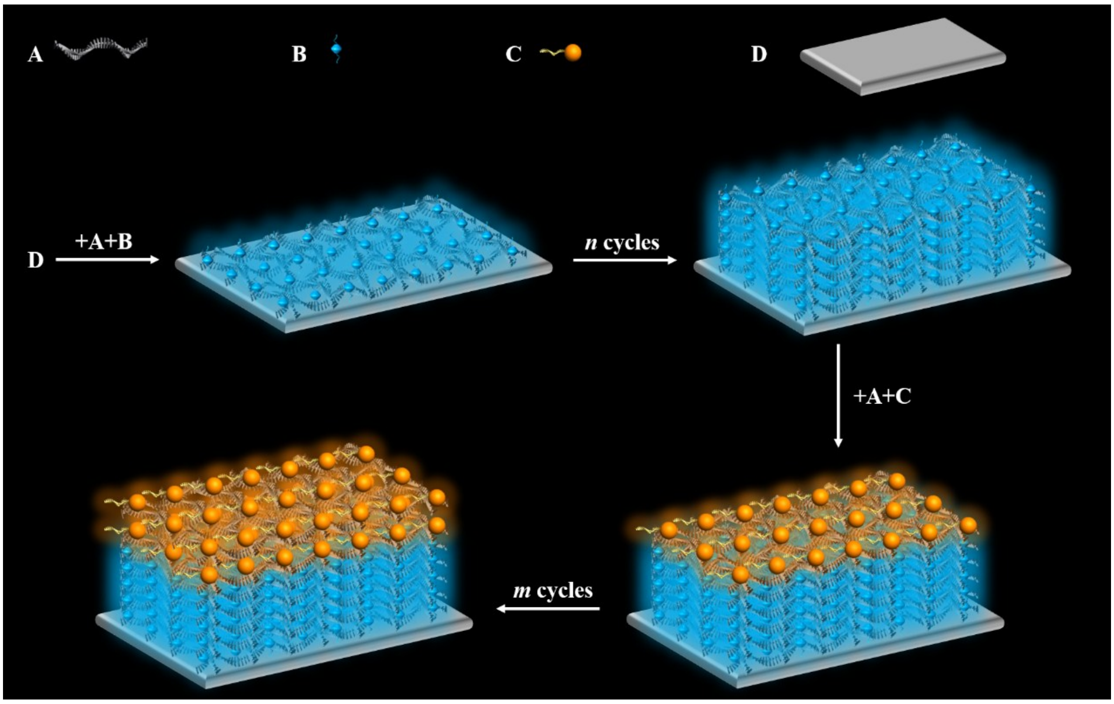
Fabrication of the mono-color luminescence ultrathin films (UTFs). The quartz glass substrate was cleaned by immersion in concentrated H_2SO_4-H_2O_2 (7:3 in volume) solution for 30 min and then ultrasonic treated with the ethanol and deionized water, respectively. The multilayer UTFs of $(BSTPE/PAH)_n$ were prepared by layer-by-layer

(LbL) assembly method. The cleaned substrate was firstly dipped into 0.1 % PAH solution for 10 min followed by washing thoroughly, and then the substrate was dipped into the positively charged BSTPE aqueous solution (100 mM) for another 10 min. Multilayer films of $(\mathrm{BSTPE/PAH})_n$ were prepared by the repeated alternation in PAH solution and BSTPE solution for *n* cycles. The resulted UTFs were dried with nitrogen gas at 25 °C. Similarly, the $(\mathrm{Au\ NCs/PAH})_m$ UTFs were prepared by the repeated assembly of Au NCs (0.4 g/L) and PAH alternatively.

Fabrication of the multi-color and white-light emission luminescence UTFs. The strategy of preparation for multi-color luminescence UTFs is similar to that of mono-color UTFs. Firstly, the blue-emission $(\mathrm{BSTPE/PAH})_5$ UTFs were prepared, and the as-prepared films were dipped into the Au NCs and PAH for deposition of orange fluorescent unit. The multi-color UTFs of $(\mathrm{BSTPE/PAH})_5/(\mathrm{Au\ NCs/PAH})_m$ were obtained by repeated procedures for *m* cycles.

Preparation of the white light-emitting diodes (WLEDs) by $(\mathrm{BSTPE/PAH})_5/(\mathrm{Au\ NCs/PAH})_2$. A flexible substrate of PE films is prepared by heating the PE powder in the double-roller mixer at 120 ^{o}C, followed by molding at the same temperature for 1.5 min. The white-light emission $(\mathrm{BSTPE/PAH})_5/(\mathrm{Au\ NCs/PAH})_2$ UTF was prepared on the flexible PE substrate. Afterwards, the as-prepared white-light emission film was coated on a commercialized UV-LED to obtain a WLED.

Sample Characterizations. The fluorescence spectra were measured by a F-7000 fluorescence spectrophotometer (Hitachi, Japan) with the slit of 5.0 nm and the scanning rate of 1200 nm/min. The UV-vis absorption spectra were obtained on the Shimadzu UV-3600 spectrophotometer (Tokyo, Japan). The quantum yield values were measured with the reconvolution fit analysis (Edinburgh F980 analysis software) equipped with the integrating sphere. The color coordinates of the multi-color luminescence UTFs were measured by the Edinburgh FLS 980 fluorescence spectrophotometer. The measurements of color coordinates of WLEDs were carried out by Konica Minolta CS-2000 spectroradiometer. The morphology data of atomic force microscopy (AFM) were implemented on the NanoScope 9.1 (Bruker, Germany) instrument. Transmission electron microscope (TEM) images were recorded by the JEOL JEM-3010 with the accelerating voltage of 100 kV. 1H-nuclear magnetic resonance (1H-NMR) spectra were determined by a 600 MHz Bruker (Germany) spectrometer at room temperature.



Scheme S1 Schematic representation for the LbL assembly process of white-light emission UTFs based on BSTPE, Au NCs and PAH.

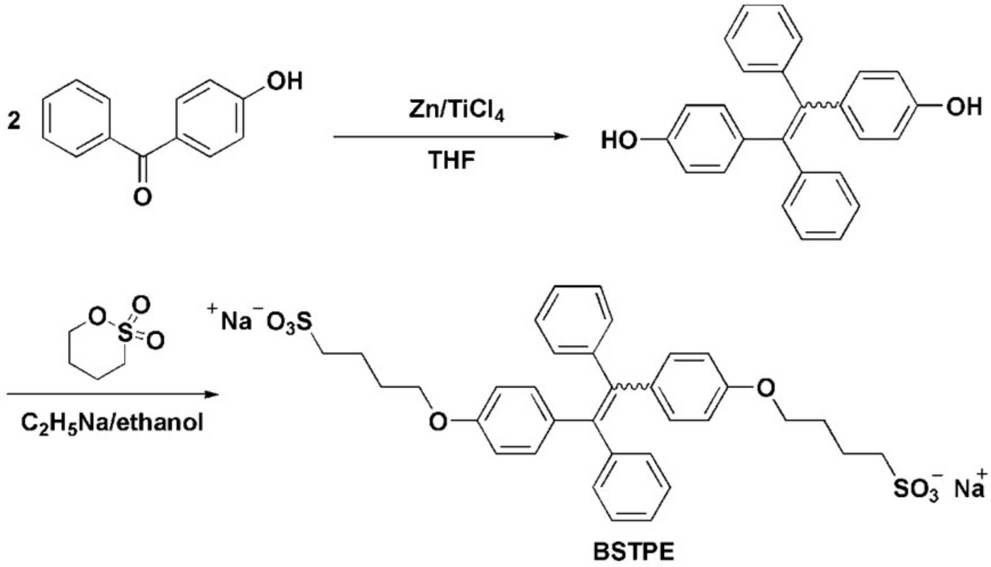


Fig. S1 Synthetic route of BSTPE molecules.

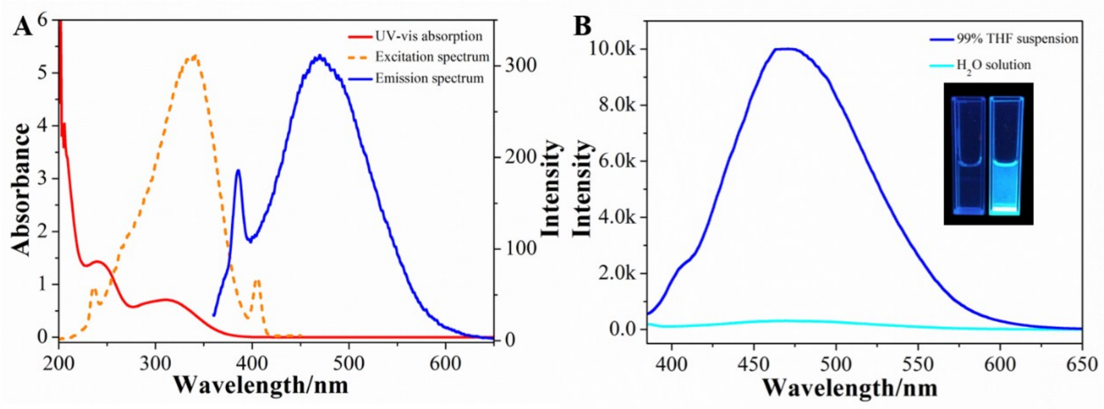


Fig. S2 (A) UV-vis absorption (red line), excitation (orange line, λ_{em}=470 nm) and emission (blue line, λ_{ex}=340 nm) spectra of BSTPE (100 μM) aqueous solution; (B) fluorescence spectra of BSTPE (100 μM) in H_2O (cyan line) and 99% THF (blue line), the inset shows the photographs of BSTPE in H_2O (left) and 99% THF (right) under UV irradiation (365 nm).

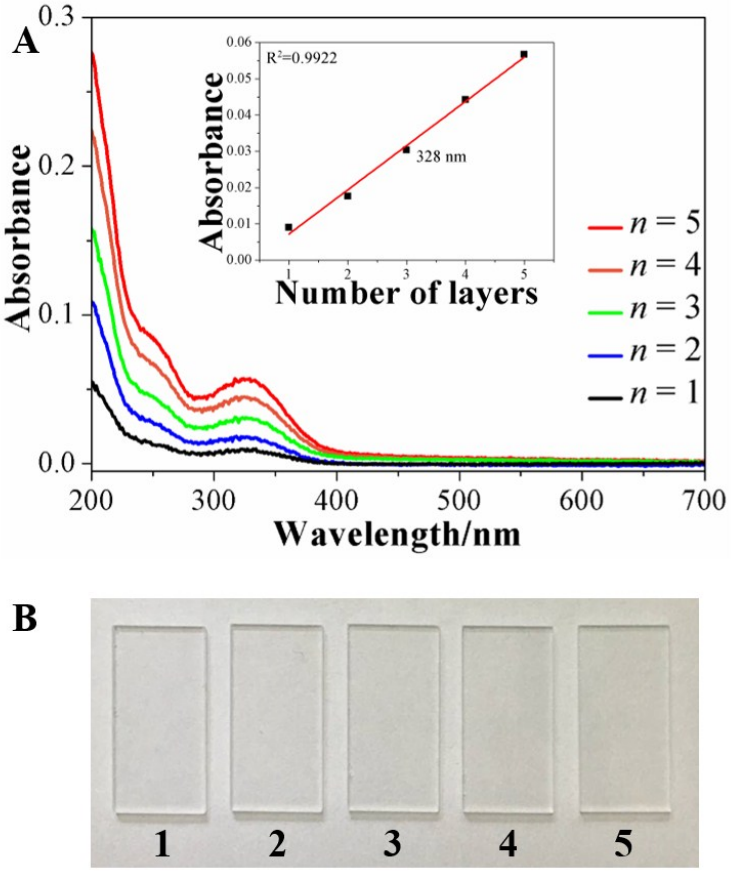


Fig. S3 (A) UV-vis absorption spectra of (BSTPE/PAH)$_n$ UTFs (n = 1−5), and the inset shows linear correlation between absorbance and bilayer number; (B) photographs of (BSTPE/PAH)$_n$ UTFs (n = 1−5) taken under daylight.

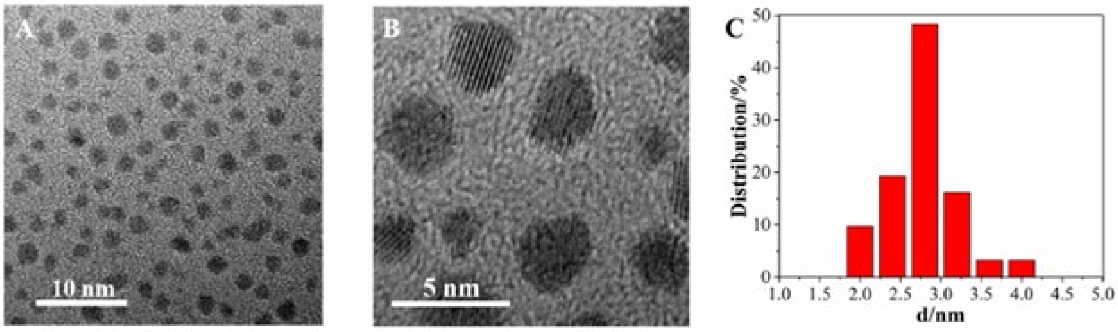


Fig. S4 High resolution TEM images of Au NCs: (A) low magnification and (B) high magnification (with crystal lattice); (C) size distribution of Au NCs.

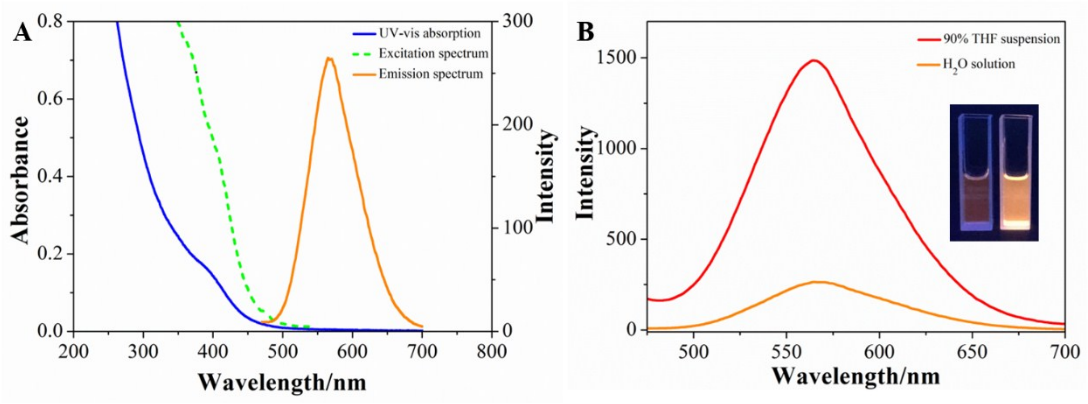


Fig. S5 (A) UV-vis absorption (blue line), excitation (green line, λ_{em}=565 nm) and emission (orange line, λ_{ex}=370 nm) spectra of Au NCs (40 mg/L) in water; (B) fluorescence spectra of Au NCs (40 mg/L) in H_2O (orange line) and 90% THF (red line), the inset shows the photographs of Au NCs in H_2O (left) and 90% THF (right) under UV irradiation (365 nm).

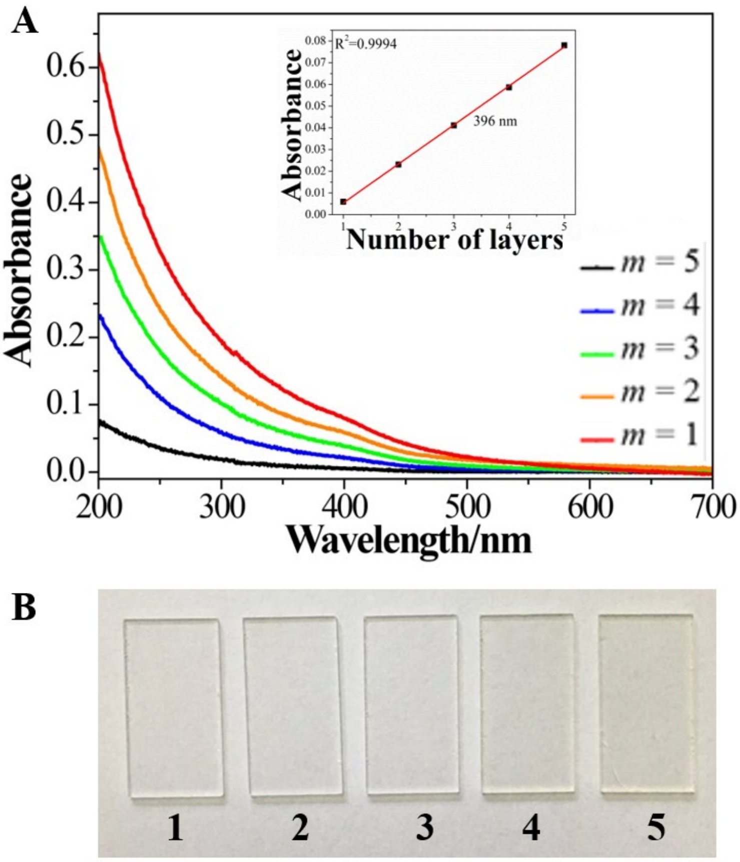


Fig. S6 (A) UV-vis absorption spectra of (Au NCs/PAH)$_m$ UTFs (m = 1−5), and the inset shows linear correlation between absorbance and bilayer number; (B) photographs of (Au NCs /PAH)$_m$ UTFs (m = 1−5) taken under daylight.

Table S1 The quantum yields of mono-color (BSTPE/PAH)$_5$ (n = 1−5) UTFs

Samples	$n = 1$	$n = 2$	$n = 3$	$n = 4$	$n = 5$
QYs	37.37%	37.49%	38.06%	39.19%	40.99%

Table S2 The quantum yields of mono-color (Au NCs/PAH)$_5$ (m = 1−5) UTFs

Samples	$m = 1$	$m = 2$	$m = 3$	$m = 4$	$m = 5$
QYs	20.53%	21.36%	21.47%	21.69%	22.89%

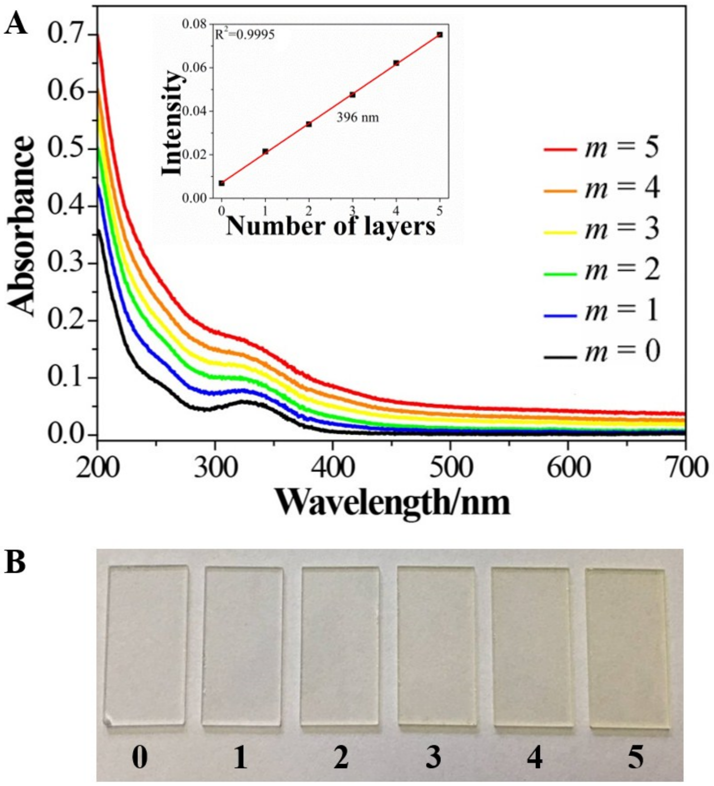


Fig. S7 (A) UV-vis absorption spectra of (BSTPE/PAH)$_5$/(Au NCs/PAH)$_m$ UTFs (m = 0−5), and the inset shows the linear correlation between absorbance and bilayer number; (B) photographs of (BSTPE/PAH)$_5$/(Au NCs/PAH)$_m$ UTFs (m = 0−5) taken under daylight.

Table S3 The coordinates of multi-color $(BSTPE/PAH)_5/(Au\ NCs/PAH)_m$ (m = 0−5) UTFs

Sample	$m = 0$	$m = 1$	$m = 2$	$m = 3$	$m = 4$	$m = 5$
x	0.183	0.258	0.333	0.422	0.478	0.496
y	0.268	0.306	0.339	0.381	0.406	0.414

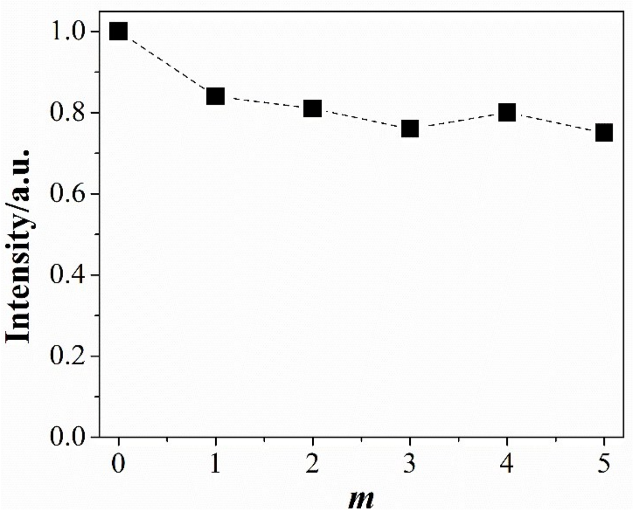


Fig. S8 Fluorescence intensity changes of BSTPE (480 nm) for (BSTPE/PAH)$_5$/(Au NCs/PAH)$_m$ (m = 1−5) UTFs.

Table S4 Thickness data for $(BSTPE/PAH)_5$, $(Au\ NCs/PAH)_5$ and $(BSTPE/PAH)_5/(Au\ NCs/PAH)_2$ UTFs

Samples	$(BSTPE/PAH)_5$	$(Au\ NCs/PAH)_5$	$(BSTPE/PAH)_5/(Au\ NCs/PAH)_2$
Thickness/nm	9.639	25.663	23.597

Table S5 Comparisons of the QYs and CIE coordinates with white-light ultrathin films from literatures.[3-10]

Samples	QYs	CIE (*x*, *y*)	References
Nanoclusters	18%	(0.34, 0.29)	*ACS Appl. Mater. Interfaces*, 2016, **8**, 33993
Quantum dots	60%	(0.363, 0.324)	*ACS Appl. Mater. Interfaces*, 2014, **6**, 5744
Quantum dots	--	(0.3973, 0.3948)	*Phys. Chem. Chem. Phys.*, 2016,**18**, 31634
Quantum dots	--	(0.32, 0.33)	*J. Mater. Chem. C*, 2016, **4**, 1665
Organic dyes	90%	(0.3216, 0.3447)	*RSC Adv.*, 2016, **6**, 22111
Polymers	72.8%	(0.30, 0.31)	*J. Mater. Chem. C*, 2014, **2**, 9035
Polymers	--	(0.33, 0.31)	*J. Phys. Chem. C*, 2015, **119**, 28701
Polymers	1.85%	(0.29, 0.36)	*Macromolecules*, 2010, **43**, 5925

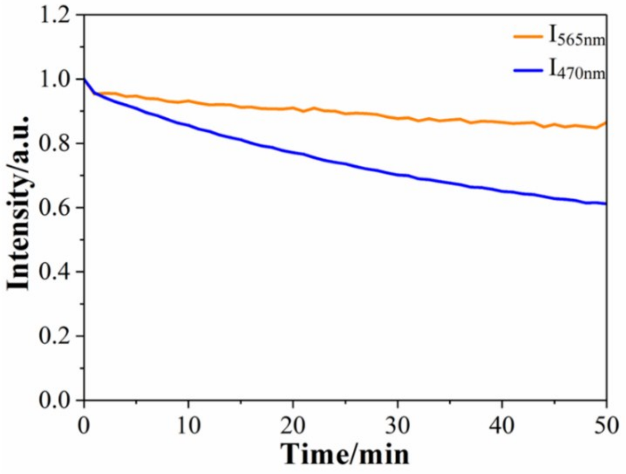


Fig. S9 Photo-stability of the $(BSTPE/PAH)_5/(Au\ NCs/PAH)_2$ UTF traced by the fluorescence intensity at 565 nm (orange line) and 470 nm (blue line).

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