Supporting Information

Large-scale Synthesize and Screen Printing of Upconversion Hexagonal-phase $NaYF_4$: Yb^{3+}, Tm^{3+}/Er^{3+}/Eu^{3+} Plates for Security Application

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Table S1 The reaction parameters of the $NaYF_4$: 18% Yb^{3+}, 2% Tm^{3+} (UCMPs) and corresponding morphology and average dimension.

Sample	Process time /h	TSC /mmol	NH_4F /mmol	NaCl /mmol	OA:EG /ml	Morphology	average dimension (L/T)/ µm	Aspect ratios[a]
S1	6	2	6	2.88	10:5	hexagonal microplates	1.127/0.883	1.27
S2	0.5	2	6	2.88	10:5	nanoparticles	/	/
S3	1	2	6	2.88	10:5	nanoparticles + hexagonal microplates	/	/
S4	2	2	6	2.88	10:5	hexagonal microplates	0.660/0.245	2.69
S5	12	2	6	2.88	10:5	hexagonal microplates	1.125/0.888	1.26
S6	6	1	6	2.88	10:5	irregular microprisms	11.599/4.067	2.85
S7	6	4	6	2.88	10:5	hexagonal microplates	0.774/0.200	3.87
S8	6	2	4	2.88	10:5	nanoparticles	/	/
S9	6	2	12	2.88	10:5	hexagonal microprisms	1.437/1.670	0.86
S10	6	2	6	0	10:5	hexagonal microplates	1.331/1.079	1.23
S11	6	2	6	1	10:5	hexagonal microplates	1.198/0.917	1.31
S12	6	2	6	2.88	7.5:7.5	hexagonal microplates	1.200/0.656	1.83
S13	6	2	6	2.88	5:10	hexagonal microplates	0.921/0.482	1.91

[a] determined that the aspect ratio is equal to the average length divided by the average thickness.

Ink viscosity and tension

Firstly, the kinematic viscosity of the ink was measured using a 0.4-0.5 mm diameter Capillary Tube Viscometer. We can get the kinematic viscosity through the following formula (eg. (1)). Then, viscosity of UCMPs inks is calculated *via* the conversion formula (eg. (2)). Here, the test temperature of the experiments is 20 °C.

$$\upsilon = c * t \qquad \text{eg.(1)}$$

$$\eta = \upsilon * \rho \qquad \text{eg.(2)}$$

where v is the kinematic viscosity of UCMPs inks (mm^2/s), c is the viscometer constant (0.004471 mm^2/s^2), t is the time of certain volume liquid through capillary (s), η and ρ are the viscosity (mPa•s) and density of UCMPs inks (1.13 g/cm^3), respectively. Then, the experimental parameters of viscosity test are presented in **Table R1.** The calculated viscosity of UCMPs inks is 113.11 mPa•s. The surface tension of PET is 47 mN/m, and the measured surface tension of UCMPs inks is 32 mN/m. The surface tension of substrate is higher than the ink, which is beneficial to the printing.

Table S2 The experimental parameters of viscosity test.

t (s)	c (mm^2/s^2)	v (mm^2/s)	ρ (g/cm^3)	η (mPa•s)
22388.00	0.004471	100.10	1.13	113.11

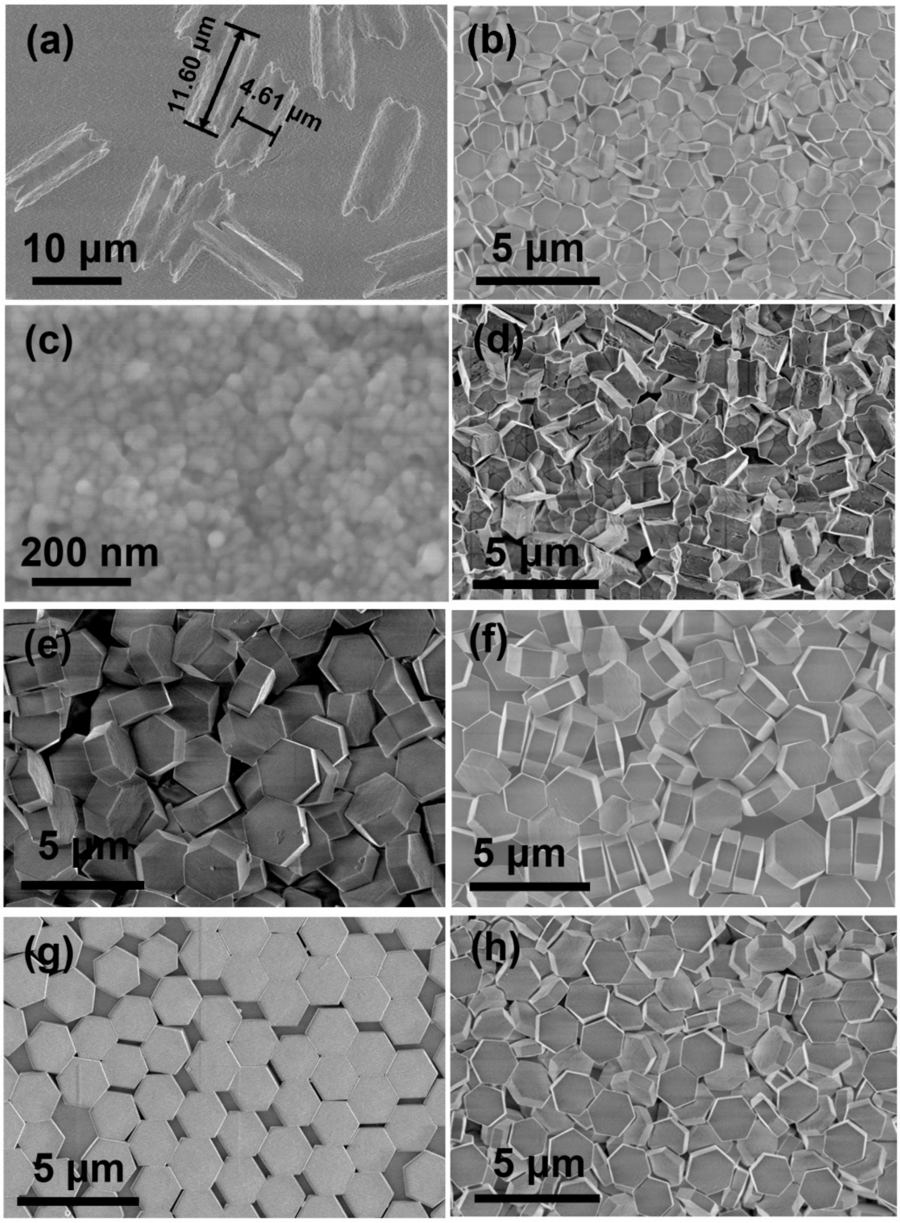

Figure S1 SEM images of $NaYF_4$: Yb^{3+}, Tm^{3+} UCMPs under different reaction conditions. (a) 1 mmol and (b) 4 mmol TSC, (c) 4 mmol and (d) 12 mmol NH_4F, (e) 0 mmol and (f) 1 mmol NaCl, (g) 7.5: 7.5 mL and (h) 5: 10 mL OA: EG, respectively.

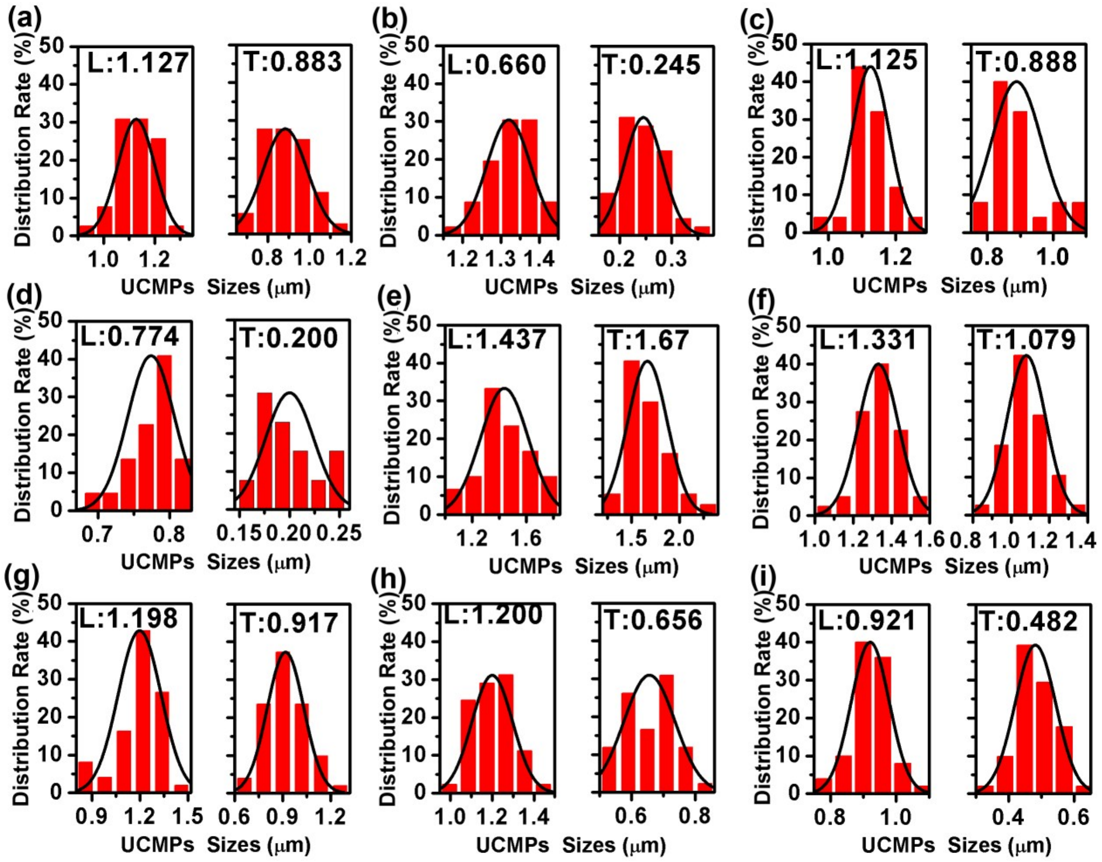

Figure S2 Sizes distribution statistics of $NaYF_4$: Yb^{3+}, Tm^{3+} UCMPs under different reaction conditions.(a) 6 h, (b) 2 h, (c) 12 h; (d) 4 mmol TSC; (e) 12 mmol NH_4F; (f) and (g) 0 mmol, 1 mmol NaCl; (h) and (i) 7.5: 7.5 mL, 5: 10 mL OA: EG, respectively.

The energy transition formulas of Tm^{3+}, Er^{3+}, $Eu^{3+}$and Yb^{3+} dopant ions.

$$^2F_{7/2}(Yb^{3+}) + h\nu(980nm) \xrightarrow{GSA} {^2F_{5/2}}(Yb^{3+}) \quad \text{(eg.1)}$$

$$^2F_{5/2}(Yb^{3+}) + {^3H_6}(Tm^{3+}) \xrightarrow{ETa} {^2F_{7/2}}(Yb^{3+}) + {^3H_5}(Tm^{3+}) \quad \text{(eg.2)}$$

$$^3H_5(Tm^{3+}) \rightarrow {^3F_4}(Tm^{3+}) + {^2F_{5/2}}(Yb^{3+}) \xrightarrow{ETb} {^3F_2}(Tm^{3+}) \quad \text{(eg.3)}$$

$$^3F_2(Tm^{3+}) \rightarrow {^3F_3}(Tm^{3+}) \rightarrow {^3H_6}(Tm^{3+}) + h\nu(696nm) \quad \text{(eg.4)}$$

$$^3F_3(Tm^{3+}) \rightarrow {^3H_4}(Tm^{3+}) + {^2F_{5/2}}(Yb^{3+}) \xrightarrow{ETc} {^1G_4}(Tm^{3+}) \quad \text{(eg.5)}$$

$$^1G_4(Tm^{3+}) \rightarrow {^3H_6}(Tm^{3+}) + h\nu(475nm) \quad \text{(eg.6)}$$

$$^{1}G_{4}(Tm^{3+}) \rightarrow {}^{3}F_{4}(Tm^{3+}) + h\nu(648nm) \qquad \text{(eg.7)}$$

$$^{2}F_{5/2}(Yb^{3+}) + {}^{2}I_{15/2}(Er^{3+}) \xrightarrow{ETa} {}^{2}F_{7/2}(Yb^{3+}) + {}^{2}I_{11/2}(Er^{3+}) \qquad \text{(eg.8)}$$

$$^{2}F_{5/2}(Yb^{3+}) + {}^{2}I_{11/2}(Er^{3+}) \xrightarrow{ETc} {}^{2}F_{7/2}(Yb^{3+}) + {}^{4}F_{7/2}(Er^{3+}) \qquad \text{(eg.9)}$$

$$^{2}F_{7/2}(Er^{3+}) \rightarrow {}^{2}H_{11/2}(Er^{3+}) \rightarrow {}^{4}I_{15/2}(Er^{3+}) + h\nu(522nm) \qquad \text{(eg.10)}$$

$$^{2}F_{7/2}(Er^{3+}) \rightarrow {}^{2}S_{3/2}(Er^{3+}) \rightarrow {}^{4}I_{15/2}(Er^{3+}) + h\nu(542nm) \qquad \text{(eg.11)}$$

$$^{2}F_{7/2}(Er^{3+}) \rightarrow {}^{2}F_{9/2}(Er^{3+}) \rightarrow {}^{4}I_{15/2}(Er^{3+}) + h\nu(656nm) \qquad \text{(eg.12)}$$

$$^{2}F_{5/2}(Yb^{3+}) + {}^{7}F_{0}(Eu^{3+}) \xrightarrow{CU} {}^{2}F_{7/2}(Yb^{3+}) + {}^{5}D_{1}(Eu^{3+}) \qquad \text{(eg.13)}$$

$$^{5}D_{1}(Eu^{3+}) \rightarrow {}^{7}F_{0}(Eu^{3+}) + h\nu(523nm) \qquad \text{(eg.14)}$$

$$^{5}D_{1}(Eu^{3+}) \rightarrow {}^{5}D_{0}(Eu^{3+}) \rightarrow {}^{7}F_{1}(Eu^{3+}) + h\nu(592nm) \qquad \text{(eg.15)}$$

$$^{5}D_{0}(Eu^{3+}) \rightarrow {}^{7}F_{2}(Eu^{3+}) + h\nu(615nm) \qquad \text{(eg.16)}$$

$$^{5}D_{1}(Eu^{3+}) \rightarrow {}^{5}D_{4}(Eu^{3+}) \rightarrow {}^{7}F_{2}(Eu^{3+}) + h\nu(656nm) \qquad \text{(eg.17)}$$

$$^{5}D_{4}(Eu^{3+}) \rightarrow {}^{7}F_{5}(Eu^{3+}) + h\nu(546nm) \qquad \text{(eg.18)}$$

$$^{5}D_{4}(Eu^{3+}) \rightarrow {}^{7}F_{6}(Eu^{3+}) + h\nu(475nm) \qquad \text{(eg.19)}$$

Figure S3 The image of screen printing plate.