Modified Surface States of NaGdF₄:Yb³⁺/Tm³⁺ Upconversion Nanoparticles via a Post-chemical Annealing Process

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Fig. S1 (a) XRD patterns of NaGdF₄: $x\%Yb^{3+}$, $1\%Tm^{3+}$ NPs; (b) the up-conversion spectra of NaGdF₄: $x\%Yb^{3+}$, $1\%Tm^{3+}$ NPs under the excitation of 980 nm laser (x= 0.39, 0.44, 0.49, 0.54 and 0.59).



Fig. S2 (a) XRD patterns of NaGdF₄: 49%Yb³⁺, y%Tm³⁺ NPs; (b) the up-conversion spectra of NaGdF₄: 49%Yb³⁺, y%Tm³⁺ NPs under the excitation of 980 nm laser (y= 0.005, 0. 01, 0.015, 0.02 and 0.025).



Fig. S3 The HRTEM images of NaGdF₄ bare core NPs.



Fig. S4 XRD patterns of the post-chemical annealed NaGdF₄: 49%Yb³⁺, 1%Tm³⁺ NPs at different temperature (T=220, 240, 260, 280 °C).



Fig. S5 The HRTEM images of 17-nm (a) and 55-nm (b) NPs after the post-chemical annealing at 200°C.



Fig. S6 The TEM (left) and HRTEM (right) images of $NaGdF_{4}$ -260 NPs



Fig. S7 The TEM (left) and HRTEM (right) images of NaGdF₄-280 NPs.



Fig. S8 The logarithmic relationship of Tm^{3+} emission intensities versus power density of $NaGdF_4$ bare core NPs (a) and $NaGdF_4$ -250 NPs (b).



Fig. S9 The fluorescence decay curves of (a) ${}^{3}F_{2,3}$ and (b) ${}^{3}H_{4}$ level in NaGdF₄ bare core NPs and the post-annealed NPs. (the annealing temperature is 200, 230, 250 and 280 °C, respectively.)



Fig. S10 (a) XRD patterns of NaGdF₄: Er^{3+} bare core NPs and the post-annealed NP; (b) The up-conversion spectra of NaGdF₄: Er^{3+} bare core NPs and post-annealed NPs under the excitation of 980 nm (the post-chemical annealing temperature is 200, 230, 250 and 280°C, respectively.).



Fig. S11 (a) XRD patterns of NaGdF₄: Eu^{3+} bare core NPs and the post-chemical annealed NPs; (b) The downconversion spectra of NaGdF₄: Eu^{3+} bare core and post-annealed NPs under the excitation of 393 nm (the postchemical annealing temperature is 200, 230, 250 and 280°C, respectively.).



Fig. S12 The intensity ratio of MD/ED of Eu^{3+} as a function of the post-chemical annealing temperature (T=200, 220, 230, 240, 250, 260, 280°C)

Pulse-width(µs)	Bare Core	NaGdF ₄ -250
50	(0.4366, 0.3931)	(0.4234, 0.4009)
100	(0.4263, 0.3889)	(0.4071, 0.3914)
200	(0.4211, 0.3747)	(0.3617, 0.3499)
300	(0.4155, 0.3672)	(0.3233, 0.3186)
400	(0.4045, 0.3580)	(0.2951, 0.2938)
500	(0.3926, 0.3488)	(0.2805, 0.2773)
600	(0.3859, 0.3406)	(0.2660, 0.2595)
700	(0.3758, 0.3324)	(0.2540, 0.2495)
800	(0.3659, 0.3264)	(0.2437, 0.2384)
900	(0.3574, 0.3189)	(0.2401, 0.2335)
1000	(0.3533, 0.3163)	(0.2340, 0.2270)

Table 1. The color coordinates of $NaGdF_4$ bare core NPs and $NaGdF_4$ -250 NPs