Electronic Supplementary Information

Low power density 980 nm-driven ultrabright red-emitting upconversion nanoparticles *via* synergetic Yb³⁺/Tm³⁺ cascade-sensitization

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Fig. S1 TEM images: (A) NaErF₄:Yb(98%); (B) NaErF₄:Yb(85%); (C) NaErF₄:Yb(60%). (D-F): Corresponding NaErF₄:Yb@NaLuF₄ core/shell structured UCNPs.



Fig. S2 Powder X-ray diffraction patterns: (A) NaErF₄:Yb(98, 85, 70, 60%), NaErF₄:Yb(98%)@NaLuF₄ (~2.5 nm), NaErF₄:Yb/Tm(19/1%) and standard β-NaYF₄ (JCPDS 16-0334); (B) NaLuF₄ coated NaErF₄:Tm(0.5%) and NaErF₄:Yb/Tm(69/1%) with different shell thickness. Fast Fourier transform (C) and high angle annular dark field scanning transmission electron microscopy (D) images of the as-prepared β-NaErF₄:Tm(0.5%)@NaLuF₄, supporting the formation of core/shell structured β-UCNPs.



Fig. S3 Decay curves of ${}^{4}S_{3/2}(Er^{3+})$ at 540 nm for core/shell structured NaErF₄:Yb(70%)@NaLuF₄ with different shell thickness (~2.5 nm, ~4 nm) under excitation at 980 nm.



Fig. S4 TEM images: (A) $NaErF_4:Yb/Tm(89/1\%)$; (B) $NaErF_4:Yb/Tm(79/1\%)$; (C) $NaErF_4:Yb/Tm(69/1\%)$; (D) $NaErF_4:Yb/Tm(59/1\%)$; (E) $NaErF_4:Yb/Tm(19/1\%)$; (F) $NaErF_4:Tm(0.5\%)$. Corresponding $NaErF_4:Yb/Tm@NaLuF_4$ (G-K) and $NaErF_4:Tm(0.5\%)@$ $NaLuF_4$ (L) core/shell structured UCNPs.



Fig. S5 TEM images: (A) NaYbF₄:Tm(1%) and (B) NaYbF₄:Tm(1%)@NaLuF₄ UCNPs. (C) Upconversion emission spectrum of NaYbF₄:Tm(1%)@NaLuF₄ UCNPs under excitation at 980 nm (0.2 W cm⁻²).



Fig. S6 Power dependence characteristics of Er^{3+} emissions at 540 and 654 nm in NaYF₄:Yb/Er (20/2%)@NaLuF₄ under excitation at 980 nm.



Fig. S7 TEM images: (A-B) NaErF₄:Yb/Tm(69/1%) with ~4 nm and ~6 nm NaLuF₄ shell. (C-D) NaErF₄:Tm(0.5%) with ~4 nm and ~6 nm NaLuF₄ shell.



Fig. S8 Upconversion emission spectra of $NaErF_4$:Tm(0.5%) with 6 nm $NaLuF_4$ shell under 980 nm excitation.



Fig. S9 Experimental setup for the *in vitro* penetration simulation model.



Fig. S10 Upconversion emission spectra of $NaLuF_4$ coated $NaErF_4$:Yb (98%) and $NaYF_4$:Yb/Er(20/2%) UCNPs.